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FOREWORD

The Editor of the Proceedings of the 21st DOE/NRC Nuclear Air Cleaning Conference extends thanks to all authors, session chairmen, panelists, and the many speakers from the floor who, after the Conference, carefully reviewed their contributions and promptly returned their edited texts thereby making it possible to complete the preparation of the Proceedings in a four month period. The purpose of completing the Proceedings in the shortest possible time is to make important information widely available while still highly relevant for current operations.

The principal tasks associated with preparing the Proceedings in a timely manner were performed ably by Mrs. Joan K. Sullivan who also provided essential administrative services before and during the Conference. I am certain that I speak for everyone associated with the Conference in extending a thank you to Mrs. Sullivan for many jobs well done.

Financial support for the 21st DOE/NRC Nuclear Air Cleaning Conference was provided by the U.S. Department of Energy (Office of Nuclear Safety) and the U.S. Nuclear Regulatory Commission (Office of Nuclear Regulatory Research), both successor agencies of the U.S. Atomic Energy Commission that sponsored the first thirteen Conferences in this series. The Nuclear Regulatory Commission undertook responsibility for printing and distributing the Conference Proceedings. For the first time, the 21st Conference was also supported financially by ISNATT (International Society of Nuclear Air Treatment Technologies) a newly formed private industry association of individuals and companies interested in all aspects of nuclear air cleaning technology. Thanks are extended to all three for their contributions of funds and, especially, for their valuable advice and guidance.

Julia First's kind attention to the hospitality functions designed for the family members accompanying those attending the technical sessions is noted and greatly appreciated.

Melvin W. First
17 December 1990

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AIR CLEANING REQUIREMENTS FOR EVOLUTIONARY AND ADVANCED REACTORS
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CLOSING COMMENTS OF SESSION CO-CHAIRMAN MOELLER

WELCOME AND OBJECTIVES OF THE CONFERENCE

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I am Chairman of the Program Committee for the 21st DOE/NRC Nuclear Air Cleaning Conference and it is my pleasant duty to open the Conference by extending a personal, as well as an official, welcome to all here in the room.

As the Conference title indicates, this is the 21st meeting of nuclear air cleaning professionals sponsored by Harvard University and one or more official agencies of the U.S. Government. The first meeting took place at the Harvard Air Cleaning Laboratory in June of 1940. Therefore, not only is the Conference celebrating its 40th year, it has simultaneously attained its 21st year majority. Long may it continue as an important international forum for nuclear air and gas cleaning technology. I introduced the word "international" purposefully to acknowledge the large representation of conference delegates from outside U.S. and their exceedingly important contribution to the technical program that will run throughout the four days of the Conference. At the 17th Conference, I stated that we in the U.S. owe a considerable debt to our colleagues from other countries for sharing with us their technical developments in spent fuel rod reprocessing for the recovery of valuable elements inasmuch as this activity seems to be becoming a lost art in the U.S. At the present time, we are also being enriched technically from abroad by reports of containment venting studies and installations.

The first Conference, sponsored by the U.S. Atomic Energy Commission 40 years ago, was during a period of great secrecy about all matters concerned with nuclear energy and attendance was 100% from the U.S. In fact, some sessions excluded U.S. citizens who did not have security clearance. No Proceedings were published. I mention this to draw attention to the fact that about 40% of the papers that will be presented at the 21st Conference are by authors from other countries and there will be no restrictions on attendance at every session.

The 21st Conference introduces us to an additional sponsoring agency, the International Society of Nuclear Air Treatment Technologies (ISNATT). I think this is an important innovation for two reasons: (1) The financial sponsorship of the conferences has been broadened by formalizing, in a very vital way, the long history of commercial interest and involvement in the Air Cleaning Conferences formerly expressed as a Government-Industry Meeting on Filters, Media, and Filter Testing and (2) a mechanism has been created for interested parties outside the U.S. to help support future Air Cleaning Conferences and to make them even more of an international event than heretofore. Therefore, we welcome the active participation of ISNATT. I urge you to join and become active participants in the Society's activities, thereby helping to support future Air Cleaning Conferences by making them international in reality.

Air and gas cleaning systems specifically designed for in-containment nuclear power plant applications have developed into a mature technology over the past 40 years. To inattentive observers of this scene, it may appear that little more remains to be accomplished in this area. However, studies of the physical and chemical nature of iodine releases following core disruptive accidents (initiated after the TMI-2 accident) have radically altered our previously held ideas about iodine, and these new ideas are stimulating a great deal of rethinking about the relative importance and functionality of the traditional elements of emergency air and gas cleaning trains. Nuclear Regulatory Guides 1.3 and 1.4 assumed that 90% of the iodine released would be in elemental form. After the accident at TMI-2, a reassessment indicated that iodine would likely be released into the containment as CsI, possibly with some HI. This is a very important transformation of ideas and I anticipate that we will learn much more about it from this week's activities.

The generally satisfactory nuclear containment experience at TMI-2 has not seemed to dampened

enthusiasm for adding filter systems to water cooled reactors to relieve hypothetical containment vessel overpressures following a core disruptive accident. Construction of these systems has taken place in Europe and a great deal of study of the technology has taken place here and elsewhere. We have an entire session on the subject at this Conference. As the only U.S. contribution to this session will be a co-chairman, it bears out my impression that containment venting has not been as well regarded in the U.S. as in Europe. It is not known to me whether U.S. reluctance to plunge into containment venting reflects greater confidence in the strength of our containment vessels and operating protocols (that were improved after TMI-2 through the efforts of INPO and others) or looking ahead to the new fail-safe modular designs now on the horizon, or for still other reasons.

I think you will find it interesting to learn that an inquiry made by the ASME sponsored Committee on Nuclear Air and Gas Technology (CONAGT) to the designers of the eagerly anticipated fail-safe power reactors to obtain information on their projected needs for air and gas cleaning equipment with the thought that CONAGT might as well get a headstart on developing suitable equipment codes, brought back the rather startling response that the power reactor designers foresaw no need for air and gas cleaning equipment in their new reactors. You can understand that I was feeling pretty sad about this response, anticipating that the nuclear air and gas cleaning industry would soon disappear, like the buggy whip industry, and eventually would become the butt of bad jokes about obsolescence. However, I was greatly heartened two weeks ago when I read in the Boston newspapers that our friends, the Union of Concerned Scientists, seriously questioned the infallibility of the fail-safe reactor designs. The Union didn't say, "Put in air and gas cleaning equipment," but perhaps the Nuclear Regulatory Commission will do so, and nuclear air and gas cleaning will continue as a modern technology for a while yet.

I believe that another recent inquiry by CONAGT will be of interest to this group, also. It asked whether there will be a need for new and different air and gas cleaning guidance documents to prepare for reactor life extension needs. The answer to this inquiry was that current codes and standards are believed to be entirely adequate for all foreseeable life extension purposes. This implies, I think, that there is a widespread perception that current activities on the preparation of codes, standards, and guides are adequate for current and future needs in the nuclear power industry. This is certainly a comforting thought. We are now looking forward to the imminent issuance of a new U.S. NRC Regulatory Guide 1.52 that should carry us through the next decade or two, as did the last revision.

When we turn our attention to the parts of the nuclear industry that are not civilian power reactors, we become aware that far less attention has been paid to their needs for standardized procedures. An outstanding example is nuclear waste management. On the program this morning, we have scheduled two papers on important aspects of nuclear waste management in the U.S. and Canada that will be of great interest throughout the world. An aspect of nuclear waste management about which we have less information to offer you concerns applications of nuclear air and gas cleaning technology to the U.S. Department of Energy's own site cleaning activities. The task has been adopted as a special commitment of Secretary Watkins. All remedial activities at these sites are of intense interest and importance, not only because of critical health and safety considerations, but because of the enormous costs involved. Costs are estimated to be 28.6 billion over the next five years just to get started. The current DOE goal is to clean all waste sites and bring all facilities into environmental compliance in 30 years, at a cost estimated to be between $ 150 and $ 200 billion dollars. We will shortly hear about DOE's plans for the environment from a very authoritative source and I don't want to encroach on his talk, but I would like to say that although only a tiny fraction of these enormous funds will be spent for air and gas cleaning equipment and services, the importance of the technology for protecting personnel and the environment cannot be overestimated or measured by the relatively small fraction of the money that will be spent in this area. I think DOE appreciates this fact, as we here do, because of their willingness to be a principal sponsor of the Nuclear Air Cleaning Conferences since the U.S. Atomic Energy Commission ceased to exist. Needless to day, we hope this interest continues.

The sorts of problems facing the Department of Energy that call for advanced air and gas cleaning technology as a principal containment barrier range from high level liquid waste concentration, solidification, and vitrification (a part of DOE's commitment to environmental restoration) to more

efficient operation of 40 year old HEPA filter installations at production sites as part of the effort to minimize future waste production, a major thrust of DOE's current waste management program. I had occasion recently to review plans for sampling tank 101-SL at Hanford; it is one that generates hydrogen by radiolysis. It is also one that recently came to the attention of a congressional committee, and from them to the attention of the public in a most unfavorable light through the news media. You will be pleased to know that the tank is vented through HEPA filters and that maintenance of this barrier throughout all manipulations in the tank occupies a very prominent part of their program planning for a sampling operation. Many more examples of the importance of nuclear air and gas cleaning technology and equipment in areas outside the civilian nuclear power industry could be cited but this one illustrates very well the importance of air cleaning technology in nuclear waste management and the modest costs involved in the application of nuclear air cleaning equipment and technology.

I cannot express too strongly my appreciation for the enormous assistance and support provided by the Program Committee over the past two years for this, the 21st Conference. Needless to say, a conference of this size and complexity takes a great deal of planning, many thoughtful suggestions, and a good deal of downright hard work selecting speakers, reviewing abstracts, and responding to the Chairman's frequent requests for assistance. The names of the Program Committee members are listed at the front of the Proceedings to remind all of us of their good work.

Naturally, I worry about whether the Conference will go smoothly and pleasantly and whether it will be a hugh technical success. Generally, the closer I get to the Conference, the more I worry; after all, that's what a Chairman is for. The other evening, I ate at a nearby Chinese restaurant and with the check I got my fortune cookie. The message was very brief and I would like to share it with you, "Complete success in all undertakings." So, I happily conclude that the Conference has been launched on a favorable tide and cannot fail to please. I hope you all enjoy your stay at this beautiful place and in addition, partake liberally of the intellectual nourishment that is available for the taking.

DEPARTMENT OF ENERGY DIRECTIONS IN ENVIRONMENT, SAFETY, AND HEALTH

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Abstract

The Secretary of Energy has issued several initiatives for the Department in the areas of environment, safety, and health. Together, these initiatives establish the framework for Department of Energy (DOE) programs designed to protect the environment and the health and safety of workers and the public surrounding DOE facilities. Much of the responsibility for carrying out these initiatives lies with the Office of Environment, Safety and Health (EH). This paper addresses recent and planned changes in DOE's organization and procedures responsive to the Secretary's initiatives and focuses on new programs within EH designed to further the Secretary's overall goals in this area. Specifically, focus is placed upon EH's response to the initiatives set forth in the Secretary's Ten-Point Initiative, including discussion of Tiger Team assessments, DOE compliance with Occupational Safety and Health Act (OSHA) standards, improvements in DOE compliance with the National Environmental Policy Act (NEPA) and other environmental requirements, and improvements to DOE's line management. The paper concludes by addressing changes in DOE's epidemiology program.

I. Introduction

I am happy to be here today at the 21st DOE/NRC Nuclear Air Cleaning Conference to talk about the Department of Energy's Directions in Environment, Safety, and Health. In 1985, Secretary Herrington established DOE's Office of Environment, Safety and Health in order to focus more of the Department's attention on environmental protection and safety at all major DOE sites and facilities. Our current Secretary of Energy, James D. Watkins, has taken the Department even further in this endeavor by initiating a new culture at DOE. The Secretary's concern for the protection of the environment, and the health and safety of the public and the Department's contractors and employees has manifested itself in a new operational philosophy. This philosophy encompasses the welcoming of and response to constructive criticism directed towards the Department, a dedication to professional excellence in all of the Department's activities, and an emphasis on the compatibility between nuclear weapons production and applicable environmental, safety, and health standards.

The Secretary realizes that the United States is now paying the price for its past focus on nuclear materials and weapons production at the expense of efficiency and safety. Through the initiatives that I will be discussing, the Secretary has worked to give attention to those programs that have been neglected or implemented improperly in the past and require immediate attention. Further, it is through these initiatives that the framework for DOE's new directions in environment, safety, and health have been established.

II. The Secretary's 10-Point Plan

On June 27, 1989, Secretary Watkins announced the implementation of a 10-Point Plan designed to increase Departmental compliance and accountability in the areas of environment, safety, and health.

Points directly affecting the EH function include the following:

- Establishing the "Tiger Team" program currently implemented through the Office of the Assistant Secretary for Environment, Safety and Health (EH).
- Mandating compliance with the standards of the Occupational Safety and Health Act at all Department facilities.
- Reviewing and making recommendations for the revision of Departmental policies on the National Environmental Policy Act (NEPA), including the coordination of DOE's major NEPA-related activities with the governors of States that host Department facilities.
- Strengthening the technical knowledge and capabilities of line management and oversight personnel in the areas of environment, safety, and health to enable such personnel to ensure compliance with related standards.
- Establishing an independent advisory panel to determine the changes needed in the Department's epidemiology program.
- Developing a comprehensive epidemiology data repository to include information on past and present DOE employees for use by qualified researchers.

III. Changes in ES&H Procedures Responsive to the Ten-Point Initiative

A. Tiger Teams

Through the Tiger Team assessments, EH is providing oversight of DOE facilities and assurance of facility compliance

with environmental, safety, and health requirements throughout the DOE complex. These Tiger Team compliance assessments were put in place by the Secretary to serve as the basis for the Department's new culture of self-assessment and accountability at all its facilities.

The assessments performed by these Tiger Teams include reviews of the operation, management, planning, and overall performance of DOE facilities in meeting environment, safety, and health requirements and complying with schedules. Each Tiger Team assessment requires at least thirty well-trained, highly qualified professionals. The teams spend approximately four to six weeks on site, and usually three to four weeks in preparation and follow-up procedures. As of August 1, 1990, sixteen Tiger Team compliance assessments had taken place at DOE sites. These include the following: Rocky Flats Plant, Feed Materials Production Center, West Valley Demonstration Project, Y-12 Plant, Pantex Facility, Mound Plant, Portsmouth Gaseous Diffusion Plant, Nevada Test Site, Kansas City Plant, Pinellas Plant, Savannah River Site, Lawrence Livermore National Laboratory, Brookhaven National Laboratory, Sandia National Laboratory (Livermore), Hanford Site, and Paducah Gaseous Diffusion Plant.

It is the experience of the Department that the facilities that have undergone Tiger Team assessments are responding well to such evaluations. They have moved quickly to address the problems identified, and in cases where direct and immediate action has not been possible, interim steps have been taken. Some of the high priority issues identified in the performance of the first Tiger Team assessments include:

- the need for clarification of the authority and responsibilities for meeting environment, safety, and health requirements;

- the identification of the necessary discipline and formality for management to function effectively in both the field and at Headquarters;

- the need to obtain adequate staff, both in quality and size, to perform program activities;

- instances of noncompliance in several areas, including construction, machine guarding, electrical safety, toxic substances use, and walking/working surfaces; and

- deficiencies in radiological protection, emergency preparedness, waste management, inactive waste site planning/oversight, and environmental monitoring.

The Secretary has instructed appropriate line managers to take the actions necessary to respond to these issues. He has also directed the Department to pay close attention to these issues

when conducting internal reviews of the DOE facilities and in developing programs to deal with the Department's environment, safety, and health concerns.

Unlike previous DOE oversight efforts, the Tiger Team assessments work to determine the root cause of management and other problems at Department facilities as well as their symptoms. The identification of these underlying causes has allowed the Department to initiate appropriate changes in the management of environment, safety, and health programs at these facilities. It has also allowed these facilities to adopt an early action approach for interacting with regulators on issues of concern to the Department.

B. OSHA Compliance

In accordance with the commitments made vis a vis the Occupational Safety and Health Act in the Ten-Point Initiative, the Department is in the process of establishing intensive occupational safety and health programs within EH. These programs integrate radiological protection and industrial hygiene, along with other Department programs in the areas of worker safety and health, into the Department's Office of Environment, Safety and Health. It will be a goal of the Office of Environment, Safety and Health's program to increase emphasis on the relationship among radiation protection, industrial hygiene, medical surveillance, and epidemiology studies.

In furtherance of this initiative, the Department has established an agreement with the Occupational Safety and Health Administration (OSHA), signed on September 22, 1989, that has significantly increased interaction between OSHA and DOE, especially as it relates to worker and facility safety. In order to help identify noncompliance with OSHA standards relating to industrial safety at Department facilities, OSHA compliance officers have been accompanying the Tiger Teams on their assessments of Department defense nuclear facilities since the Fall of 1989, including those at Mound, Pantex, and the Y-12 Plant. OSHA has also been helping the Department to establish a training program for effective monitoring of workplace safety, including such practices as hazard evaluation and workplace safety compliance reviews. Department and contractor employees have been undergoing training at the OSHA Training Institute in Illinois as well as at worksites under OSHA jurisdiction. OSHA has agreed to perform an overall review of the Department's worker safety oversight program, and the Secretary of Labor is expected to provide a written report containing recommendations in this regard later in the year.

C. NEPA and Environmental Compliance

Pursuant to the Secretary's initiative, a comprehensive review of the Department's National Environmental Policy Act (NEPA) practices has been performed and a new policy has been

approved. The policy provides for early participation by host States in DOE's NEPA process and helps to ensure that both the public and the Department as a whole will be better able to evaluate the environmental impacts of proposed Departmental projects. Proposed revisions to the Department's NEPA Guidelines may be issued this fall as draft rules for public comment. Upon their finalization, comprehensive Departmental procedures for NEPA compliance will be issued for the first time as regulations.

In addition to increased emphasis on NEPA compliance, the Department is also focusing on achieving compliance with other environmental laws and regulations. Many environmental requirements facing DOE are externally driven by Federal and State laws and regulations with which DOE and DOE contractors must comply. The Department's policy is that compliance with all applicable environmental laws is an integral part of operating DOE facilities.

Consistent with the Department's policy and Secretary Watkins' initiatives to assure that DOE facilities are operated in compliance with environmental laws and regulations, the Department has been actively negotiating and executing compliance agreements and agreements to clean up and restore the environment at many facilities with EPA and host states. Since 1979, DOE has executed 53 agreements and is currently negotiating 24 agreements. In addition, the Department is negotiating Agreements in Principle with host states to provide greater access to the facilities and promote additional environmental oversight and monitoring by the states.

To resolve complex environmental compliance issues and further improve compliance at its facilities, the Department, through the Office of Environment, Safety and Health, frequently coordinates and works with the EPA and host states. Work groups or negotiating teams composed of DOE program, field and oversight staff are established as appropriate to work with counterparts from EPA and the states to resolve environmental issues, such as the application of the Resource Conservation and Recovery Act (RCRA) land disposal restriction requirements to DOE mixed waste; the application of the Toxic Substances Control Act (TSCA) to DOE's three gaseous diffusion plants which are contaminated with polychlorinated biphenyls (PCBs); and the use of model language in compliance and clean up agreements with EPA and states.

D. Strengthening Line Management and Oversight

In furtherance of the Ten-Point Initiative's line management reforms, a Secretarial Notice was issued on September 5, 1989, identifying the need of the Department's line management for more skilled technical support. Despite the fact that much of the Department's work is performed by contractors, DOE's line management is still responsible for adherence to standards and requirements. Secretary Watkins announced his intention to create permanent positions within DOE to be filled by persons

with the necessary skills and experience to support line management in the fulfillment of its oversight responsibilities. These new positions will help to solidify one of the Secretary's main goals: primary accountability and responsibility for one's actions at all levels of the Department, especially with regard to environment, safety, and health.

One revision related to the line management reforms specifically affecting the Office of Environment, Safety and Health, is the recently implemented change in EH's responsibilities. Nuclear safety policy development for nuclear reactor and nonreactor nuclear issues, once the jurisdiction of EH, has been reassigned to the Assistant Secretary for Nuclear Energy. This decision was influenced by the newly increased awareness and support for DOE nuclear facility safety as evidenced by the establishment of the Defense Nuclear Facilities Safety Board. The Department also intends to continue its oversight in other designated safety and environmental areas. Having been relieved of direct nuclear safety policy responsibility places EH in a position whereby it will provide greater concentration on its oversight in such areas as environmental protection, radiological protection, epidemiology and health surveillance, facility non-nuclear safety, and worker occupational safety at DOE defense and non-defense facilities.

To further the goals of the Secretary's Ten-Point Initiative, independent external and internal oversight systems have also been established by the Secretary to ensure Departmental compliance with applicable environment, safety, and health requirements. Such independent oversight participation includes that of the Advisory Committee on Nuclear Facility Safety and the Defense Nuclear Facilities Safety Board as well as the improved State monitoring of Departmental operations called for in the Ten-Point Initiative. However, it should be emphasized that through the Secretary's establishment of a new line management system, it is the direct and complete responsibility of the line managers to ensure Departmental compliance with applicable laws and regulations.

Another form of independent, internal oversight designed to ensure structural and operational safety at Department nuclear facilities is the establishment of the Nuclear Self-Assessment Offices within the program offices. The Secretary has mandated that such offices be established within the offices of Nuclear Energy, Defense Programs, and New Production Reactors. The purpose of these Nuclear Self-Assessment Offices is to conduct independent design, construction, and operational evaluations of Department nuclear facilities. Reports will be made directly to each offices' Program Secretarial Officer.

Other new offices established by the Secretary include the Office of Environmental Restoration and Waste Management and the Office of Nuclear Safety. The Office of Environmental Restoration and Waste Management - - called for in the

Secretary's Environmental Restoration and Waste Management Five-Year Plan - - is taking the lead in DOE's environmental cleanup efforts and in discovering permanent solutions for our waste management concerns. The Office of Nuclear Safety is acting as an advisory body for line managers, overseeing their efforts to ensure nuclear safety in operations. This office has been given an extensive capacity for auditing and monitoring line management's ability to ensure nuclear safety in order to better advise managers of adequate safety levels.

As an example of how my office, the Office of Environment, Safety and Health, will provide independent internal assessments I will describe briefly our involvement in the pending readiness review for resumption of plutonium operations at the Rocky Flats plant.

As an independent oversight organization we will first evaluate and comment on the proposed Operational Readiness Review (ORR) which will be conducted by the Departmental element responsible for facility Defense Programs (DP). Independent of DP's review we will conduct selected independent verifications to assess the adequacy of DP's conclusions. Of special concern to us is the assessment of the adequacy of both the contractor's execution of their phased resumption and contractor and DOE line management (DP) oversight. Our review will be documented in a report completed at a time consistent with the resumption schedule and provided to the Secretary of Energy for his action.

Our review will include Occupational Safety and Health Administration (OSHA) standards compliance, fire protection, configuration control, adequacy of safety-related systems and components, criticality safety, radiation protection, and discipline of operations. These are all operational safety considerations. To assess the adequacy of these safety issues, we will overview the DP activities and perform a limited verification-type inspection of the workplace. A substantial part of our review will be verification of a small sampling of safety-related procedures and worker training performance.

E. Epidemiology Program

Now I would like to turn my attention to those EH health initiatives responding to the report issued by the Secretarial Panel for the Evaluation of Epidemiology Research Activities (SPEERA). In his Ten-Point Initiative, Secretary Watkins called for the establishment of an independent panel to recommend necessary changes to the Department's epidemiology program. In August 1989, the Secretarial Panel for the Evaluation of Epidemiology Research Activities (SPEERA) was appointed. Through site visits, public meetings, invited testimony, and document review, SPEERA completed an evaluation of the Department's epidemiology activities. On March 27 of this year, the Panel issued its report containing over fifty recommendations.

In its report, the Panel expressed concern for the fragmented state of the Department's epidemiology and other occupational health programs. The Panel emphasized the need for a coordinated program of occupational and environmental health and safety and provided the following recommendations:

- consolidation of all aspects of occupational and public health under my office, the Office of the Assistant Secretary for Environment, Safety and Health
- development of a strong descriptive-type epidemiology program to assure pro-active worker/health surveillance
- creation of an outside relationship with the Department of Health and Human Services (HHS) to manage the longer-term, analytic-type epidemiologic research
- extensive use of committees for advice and guidance on program priorities
- improved communication of epidemiologic information to the workers and the public

Upon receipt of the Panel's final report, the Secretary issued several directives in response to the report's most significant recommendations. The foremost of these directives establishes a new Deputy Assistant Secretary for Health under the Assistant Secretary for Environment, Safety and Health. This new office will be responsible for all occupational health and epidemiology programs within the Department. It will also oversee the occupational medical programs of Department contractors.

A review of all on-going Departmental epidemiology projects and a list of all possible new study projects have been completed to help determine the appropriate direction, size, and scope of the DOE's expanded epidemiology research needs. The review was completed with the aid of DOE's internal Epidemiologic Research Coordinating Committee (ERCC). As Assistant Secretary for Environment, Safety and Health, I chair this committee. The purpose of DOE's internal committee is to coordinate epidemiologic research activities. A near-term objective is to transfer all on-going and future analytic-type epidemiologic research to HHS for their management. To enable this we will transfer funding to HHS under a Memorandum of Understanding (MOU). The new Office of Health has already started negotiations with HHS to establish a long-term MOU and it is projected to be signed by the end of this month. We expect HHS to commence actual management responsibilities for analytic studies in FY 1991.

Some notable examples of DOE-funded, analytic epidemiology studies include the on-going Hanford Environmental Dose Reconstruction (HEDR) Project and the recent University of Utah

study of leukemia incidence in populations surrounding the Nevada Test Site. In both of these studies the DOE is fostering new research into dose reconstruction using the very latest skills in atmospheric pathway modeling in order to assess the hypothetical doses received by the general population surrounding the facilities of interest. This task is somewhat complicated by the fact that it relies heavily on historical records not only of effluent releases and environmental monitoring but also of the social-economics, demographics, and life style practices during a period extending back almost forty years. In the case of the Hanford dose reconstruction, the actual health studies will be carried out upon the successful completion of the dosimetry work to be carried out over the next two years. HHS has already initiated a thyroid disease study which will be completed in parallel to the HEDR project and many more similar studies of this nature are expected. In the case of the Utah study, the preliminary dose assessments had previously been completed and the researchers carried out an evaluation of health and mortality records on a defined cohort that led them to conclude that the excess leukemia incidence observed in southern Utah may be attributable to exposure due to fallout. These are but a few of the more than twenty epidemiologic studies now under way at the DOE.

Other directives issued by the Secretary in response to the SPEERA report include the creation of a strengthened Departmental program for health surveillance and the development of policies and protocols for the release of the Department's epidemiological data to researchers. These directives provide for the surveillance of occupational disease and disability as well as the health concerns of nearby communities. They also work to improve the communication of resultant data both within DOE and with the public.

Once fully implemented, these directives will greatly improve the Department's control of worker and public health. They will also enable the Department to communicate epidemiological and other health-related information more readily. Through this reorganization and improved communication, the Department will be in a better position to assume its responsibility for worker and public health and safety.

With regard to the development of a comprehensive epidemiologic data resource (CEDR), a draft program plan has been developed. An interim system with data on approximately 70,000 workers has already been installed for testing. Technical working groups for the repository in the areas of site surveys, demographic data, dosimetry information, and vital statistics have provided recommendations. An integral part of this CEDR objective is the creation of a Department-wide approach to consistent application in health surveillance. Joint efforts by the Office of Epidemiology and Health Surveillance and Occupational Medicine are already underway to achieve this objective. Also under way are efforts to obtain permission to

release cause of death data provided by the States to the Department as well as to other researchers. The goal of the CEDR program is to establish a way to organize and store data from all epidemiology studies conducted by the Department of Health and Human Services (HHS) in association with DOE or by DOE itself for use by bonafide researchers. The interim CEDR system is expected to be available for use by June 30, 1991, with phased implementation of the full system as data becomes available.

V. Conclusion

The initiatives being issued by the Secretary and the resultant changes within the Office of Environment, Safety and Health depict the Department's evolution towards a new culture of environmental preservation, protection of public and worker health and safety, and assurance of National security. DOE is working to make these goals compatible by revising line management, expanding data collection and communication, increasing internal and external oversight, and emphasizing worker responsibility and accountability. It is conferences such as this one that will help DOE as it progresses through these changes. Exchanging information about clean air problems and solutions is important to all the Department's nuclear facilities. With the proper standards put in place by groups such as this one, DOE and others may focus their energy and resources on individual compliance issues.

DISCUSSION

FIRST: Because of our teaching activities at Harvard over the many years, it is obvious we have a lot of friends at most of the DOE nuclear facilities, not necessarily all in the very highest echelons. What I hear, and I hear it over and over again, is that as a result of TIGER Team activities and other similar activities, morale is at a very, very low level at the sites. Instead of being "buoyed up" by this attention people are being discouraged. I wonder if there is someone at DOE who recognizes that there has to be some encouragement for these people to do their best rather than a long, bitter series of condemnations, not only by the reports themselves but also by the press. Now let me just say that I recognize that there are things that are wrong at these sites, but I would like to think that the effort is intended to help the sites rather than to tear them down. Do you have a comment on that?

ZIEMER: The point is well taken and I must tell you that I have heard the same things. I am new in the DOE, so I have heard them from the outside. I have a number of former students who work at DOE facilities or in the DOE, as well as many colleagues of many years who are at these sites. It is quite true that when these types of problems arise there is the possibility that morale will be affected. After all, it is a type of criticism of what has been done or what is being done. That type of criticism is always hard to take, especially by those who have, in fact, worked hard for many years and

have tried to do a good job. In fact, there are many who have done a good job, and we know that. There is a lot of talent in those facilities. It is also true that many of them, particularly in our areas of environmental safety and health, have not always been able to do the things that they have felt are important because of certain decisions that were made in the past. So, there are those in the labs who now welcome the fact that these areas, which so many of us have felt needed attention, are, in fact, finally being given that attention. Nonetheless, the criticism does hurt a bit. However, it should be mentioned that the critiques are done by peers. Largely, the teams that go in are from the other national laboratories, so they are critiquing each other. There are also some headquarters people and some outside consultants; it is a mix. The critiques are done by peers who are experts in the various areas, such as OSHA, NEPA, or health. In that sense, the critiques have to be seen as valid.

You asked, "Is there something to buoy us up amongst this type of criticism?" I think there are two things. Although I came onboard officially a day ago, I have, of course, had the opportunity to spend a fair amount of time at the Department over the past several months to become acquainted with the operations. One of the first questions I asked, Dr. First, was that very question. Are we only criticizing? What if they are doing something right? One thing the TIGER teams are doing is they are identifying what you might call good practices. And, in fact, the TIGER team reports contain not only critiques of where things need to be improved, or changed, or corrected, but they also contain a section called Noteworthy Practices. Our office of special projects, which basically coordinates the TIGER teams, has put together a document, the draft of which I have seen just a few days ago, that brings together all the good practices from the many labs into one document which will be distributed widely within the system and outside to point out what the labs are doing right and what they are doing well. So there is an effort to do just that. The second thing is that as corrections are made, and as action plans (there are always action plans) go into place, as things get underway, and the practices turned around, I think that will, in itself, help morale. Criticism, if it is constructive and brings about a change overall, can be good and that is what we are looking for.

MOELLER: I think it is very clear to everyone that Paul faces a tremendous challenge. Paul, we are proud that you are there and we wish you every success.

AIR CLEANING REQUIREMENTS FOR EVOLUTIONARY AND ADVANCED REACTORS

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Abstract

The NRC is reevaluating its approach to licensing for evolutionary and advanced nuclear power plants. The objectives of this reevaluation are (1) safety improvements due to advances in the technology and operating experience with the current generation of reactors, (2) improvements in communication between the NRC and industry groups on technical issues, and (3) consolidating the issues open to litigation to make the process more efficient and effective. This process raises important questions about the role of, and requirements for, air cleaning systems in future nuclear power plants. This paper describes the context in which these questions will be addressed.

I. The Present: A Time of Transition

No new nuclear power plants have been ordered in the last decade, hence the design and the licensing pipelines are virtually empty. The new requirements that resulted from the evaluation of the TMI accident essentially all have been implemented, therefore this major task is out of the way. The result is an opportunity to reconsider what we have done and to try to make improvements.

Much has been learned in the 47 years since the "Atoms for Peace" program was initiated. The various research programs have made major contributions to our knowledge. Plant designs and the regulatory systems have evolved. Extensive experience has been acquired with both plant and regulatory systems. This knowledge should enable us to do a better job with the next generation of nuclear power plants.

The nuclear industry is taking advantage of this opportunity to present new plant designs that reflect the advanced state of the designers art. The NRC is actively reviewing General Electric's "Advanced Boiling Water Reactor." Other "evolutionary" plant designs are in various stages of development. Furthermore, EPRI is developing a "requirements document" for "passive" plants and the designs are expected in the next few years. Thus there is extensive activity on the part of the industry to improve plant designs and to develop new approaches to safety issues.

The NRC has been active in reassessing regulatory positions. The establishment of official safety goals was a major development

[1]. The safety goals provide an essential basis for the evaluation of regulatory requirements. Also, the Commission has approved a fundamental revision of its basic "Standards for Protection Against Radiation" (10 CFR Part 20), a change that will have a significant impact on the nuclear industry. The NRC has gone a long way in confronting the severe accident issue. Consideration of severe accidents began well before the publication of WASH-740 in 1957 [2]. The Reactor Safety Study [3] marked the transition to the use of PRA; we have progressed to the Severe Accident Risks study [4] and the Individual Plant Evaluations (IPE) [5, 6]. The NRC is now undertaking a "decoupling" effort, designed to divorce the plant engineered safety feature requirements from the design basis dose calculations and the dose criteria of 10 CFR Part 100 [7].

II. The Current Licensing System

Evolutionary Development

Early in the reactor development program, it was recognized that exclusion distance was one of the most reliable measures for protecting the public from the ill effects of a severe accident. By 1950, the exclusion radius (R, miles) was considered adequate [8] if it was related to the thermal power (P, kW) by:

$$R \geq 0.01 \sqrt{P}$$

This limitation was indeed adequate to prevent catastrophic consequences, but caused problems for many possible applications. Even for a reactor as small as 50 MWt, the exclusion radius had to be greater than 2 miles. For a large modern plant, the exclusion radius would approach 20 miles.

One of the first steps away from reliance on separation distance was taken in September 1948 when the Atomic Energy Commission (AEC) approved the construction of a Naval reactor prototype at the West Milton site near Schenectady, New York. The reactor was built inside a containment sphere that was designed to contain the radioactivity that might be released by a severe accident. By 1955, when the first applications were made for construction permits, containment was a well established feature of nuclear power plants. Indian Point-1, Dresden-1, Yankee Rowe, Humbolt Bay, and Big Rock Point all had containments. Hallam, with a sodium-cooled reactor, was the only US nuclear power plant licensed to operate without a containment.

The first construction permits were issued on the basis of judgement, including consideration of the radiation doses that might result from a severe accident. In May 1959, the Commission issued a "Preliminary Notice of Proposed Rulemaking," indicating that reactor siting might be addressed in a new regulation, 10 CFR Part 100 [9]. The notice indicated that the factors involved in the acceptability of a site were (1) exclusion distance, (2) population density, including distance from towns and cities,(3)

meteorological considerations, (4) seismological considerations, and (5) hydrology and geology. The draft rule, which was published February 15, 1961, stated the basic objectives as being (1) to prevent serious injury from credible accidents and (2) to keep the number of people killed from being catastrophic in the event of an accident "not normally considered credible" [10]. The draft rule stated that the site and the reactor design would be considered together but the emphasis of the rule was on exclusion radius, low population distance, and doses. The final rule [7], issued April 12, 1962, was similar to the draft except that the appendix on dose calculation was deleted and replaced by a reference to TID-14844 [11]. The "Statement of Consideration" indicated that a desire to (1) limit the cumulative dose to "a level that might be considered low in comparison to what might be considered reasonable for a total population dose" and (2) to protect "people in large centers, where effective protective measures may not be feasible."

TID-14844 ("the TID") provided a source term and methods for calculating doses to determine the exclusion radius and the low population distance [11]. The TID was explicit about the conservatism in the method, such as giving no credit for containment sprays or other air cleaning systems. According to this method, 80% of the plants already approved were acceptable. Few, if any, of the plants licensed since 1962 would be acceptable using the TID methodology. For a large modern plant, the TID methodology yields exclusion radii of over 2.5 miles and low population distances of about 40 miles.

To meet the need for plants larger than 1500 MWt and exclusion radii less than 0.9 miles, credit had to be given for the air cleaning effects of containment sprays. The construction permit issued March 2, 1964 for San Onofre-1 reflected credit for sprays.

Standardized Accident Consequence Analysis

Part 100 provides dose criteria but the criteria have little meaning unless the calculational methods are defined. The extent to which dose calculation standards are needed is indicated by the safety analysis reports submitted by General Electric in the 1960s; the General Electric calculations often yielded dose results thousands of times lower than the Commission results.

The Commission quickly departed from the TID dose calculational methodology by giving credit for air cleaning by containment sprays and other air cleaning systems. In 1970 the revised dose calculation methods for loss-of-coolant accidents were documented in Safety (now called Regulatory) Guides 1.3 and 1.4 [12, 13]. Guides for other accidents were developed later and consolidated in "Standard Review Plan" (SRP) Chapter 15 [14].

There are three key considerations in the Commission's dose calculation methods. The first factor is that these are design basis calculations; this means the system being evaluated must be

assumed to function. For example, there is no point in assuming containment failure when evaluating the acceptability of the containment design leak rate. The second factor is the desire to be conservative; this generally means allowing less credit for a system than its designer would be likely to claim. Credit is limited because rarely has effectiveness been demonstrated under accident conditions and because experience shows that system effectiveness frequently degrades with time after testing. The third factor is the desire to avoid excessive reliance on any one system or component. If maximum credit were given for one system, it might eliminate the need for other systems. This would conflict with the defense-in-depth philosophy and result in an undesirable vulnerability.

Interconnected Requirements

As experience was gained with nuclear power and the public became more concerned about safety, the Commission also focused more attention on safety. In the early 1960s, Commissioner Thompson expressed the position succinctly:

Public safety accidents:	There **must be none.**
Economic Accidents:	Should be prevented.
Industrial Personnel Accidents:	Should be rare as possible.
Operational Problems:	Should be minimized.

The defense-in-depth philosophy indicated that minimizing the frequency of lesser events, the more serious accidents could be made highly improbable. Furthermore, experience had shown that problems were principally in conventional, rather than nuclear, areas. The extensive experience with nuclear accidents prior to the publication of Part 100 was documented in the SIFTOR project after the SL-1 accident [15]. This provided the background for the "General Design Criteria" (Appendix A to Part 50) and the "Standard Content and Format For Safety Analysis Reports" (Reg. Guide 1.70) [16] that were being developed in the early 1960s. The scope of regulatory review grew to include a wide range of accidents and the dose criteria of Part 100 provided guidance for the acceptability of various systems.

The Development of Siting Requirements

Part 100 provided a basis for trade-off between site characteristics and design features. Taken to the extreme, this could imply that metropolitan siting could be accepted. Applications were submitted for six metropolitan sites (Bolsa Island, Burlington, Edger, Newbold Island, New Rochelle, and Ravenswood), but none were approved [17]. It was soon recognized that, no matter what the design, some exclusion area and low population distance would be required to permit expedient emergency action in case of a severe accident. At that juncture, it became evident that, if the unstated siting requirements were met, the design could be adjusted so the dose criteria of Part 100 also would be met.

The National Environmental Policy Act of 1969 (NEPA) and the Commission's implementing regulation (10 CFR Part 51) expanded the public protection considerations in siting. These criteria, which are delineated in Regulatory Guide 4.7, [18] are much greater constraints on site selection than are the Part 100 dose criteria.

After the accident at TMI, greater emphasis was placed on emergency preparedness. The criteria are presented in 10 CFR §50.47, Appendix E, and NUREG-0654 [19]. The emergency preparedness requirements, such as prompt notification and an "emergency planning zone" of approximately 10 miles radius, effectively preclude metropolitan siting.

The Role of the Part 100 Dose Criteria

The Part 100 dose criteria were no longer a serious factor in site selection but the criteria became very important in the design and operation of numerous systems, including the following:

Containment leak rate,
Containment spray cleanup rate,
Reactor coolant cleanup system,
Main steam isolation valve closure time,
Closure times for other valves,
Steam generator leak rate,
Decontamination factors of various air cleaning systems,
Setpoints of numerous monitoring systems,
Standby gas treatment systems, and
Stack heights.

In addition to the design implications, the Part 100 dose criteria play important roles in both operation and regulation. The dose criteria provide the bases for many technical specifications such as the limits on radioactivity in reactor coolant, radioactivity release limits from BWR air ejectors, and surveillance requirements for the previously mentioned design features. The regulatory program places heavy emphasis on compliance with the technical specifications. Furthermore, when escalated enforcement is considered, one important factor is whether the violation could have led to the dose criteria being exceeded.

III. The PRA Experience

Early Progress

From the beginning of the nuclear power program, it has been recognized that the probability of occurrence is a major factor in any hazard assessment. There are calls for analysis of the "worst possible accidents," [20] but such analyses provide little information about the risks. It has long been evident that the need is for analyses that relate probabilities to the calculated consequences of nuclear accidents. In 1957, for example, WASH-

740 [2] offered an assessment of the probability of a nuclear accident with severe off-site consequences that is remarkably consistent with current estimates.

The application of probabilistic risk analysis (PRA) to nuclear systems dates back at least to the work of T. G. Bassett in 1957; he applied PRA to parts of the Ford reactor at the University of Michigan [21]. In 1964, Bob Hart and his colleagues at Atomics International performed a PRA of the SNAP-10A reactor and used that PRA to obtain approval for its launch and operation in space [22].

The Reactor Safety Study

Despite earlier attempts [23], PRA did not make its formal appearance in the regulatory program until 1975, when the "Reactor Safety Study" was published [3]. That study offered important insights into the possible accident sequences that contributed significantly to risk. It was, of course, subject to considerable criticism for the final probability numbers it contained. There was, and still is, no standard or generally accepted method for performing such analyses and, great uncertainties are present. Considerable judgement is involved in performing the analyses. Whatever its shortcomings, however, there is a consensus that the Reactor Safety Study contributed greatly to our understanding of reactor accident analysis.

Recent Progress

In the past decade, the NRC and the nuclear industry made substantial advances in the use of PRA as well as in the recognition of its limitations. In the absence of standardized methods for performing such analyses it has not been deemed practical to base regulatory requirements directly on PRA results. However, the studies have been found to provide important insights into vulnerabilities, deficiencies and potential problem areas. The new "Severe Accident Risks" study (NUREG-1150) is a significant advance from the Reactor Safety Study.

Many people are convinced that the most important PRA contributions to reactor safety will come from the Individual Plant Evaluation program (IPE). In this program, each licensee is being called upon to perform a systematic evaluation of his own plants to identify vulnerabilities and to find opportunities to make significant safety improvements. This is a two part program. The IPE proper is directed to internal events such as component or system failures [5]. The second part is directed toward external events such as earthquakes [6]. The entire program will require the devotion of considerable resources, but it is expected to make an important contribution to safety, both by improvements in the plants and by enhanced understanding by the licensees and the regulators.

Some of the more significant insights from past PRAs are that (1) accidents in which containment integrity is maintained present essentially no off-site hazard, (2) if containment integrity is maintained for a few hours the hazard is small, (3) there remains a small but significant probability of early containment failure and a substantial off-site hazard, and (4) there is a very high probability of awareness of impending core damage to permit early evacuation and the resulting elimination of off-site injury. Thus the most important contributions to safety can be made by, first, eliminating any plant specific weaknesses and, second, by reducing the likelihood of early containment failures.

IV. Opportunities for Improvement in the Regulatory System

Improved Communications

With the present licensing system, the severe accident potential is not addressed directly. Instead, extensive efforts are made to fine-tune ESF parameters and site characteristics so the Part 100 dose criteria are met using the calculational methods of the Standard Review Plan. The NRC staff uses the flexibility provided in the SRP to accommodate plant specific problems and to address perceived deficiencies, but the artificiality of the process is manifest. The dominant fact is that:

> **If the engineered safety features do not experience gross malfunction, the real off-site doses will be trivial even if the doses calculated by the SRP methods exceed the Part 100 criteria.**

Working in this environment has complicated communications between the NRC staff, applicants, and other interested parties. Thus there is an important opportunity to improve communications by directly addressing the safety objectives and concerns.

More Effective Utilization of Resources

Demonstration of compliance with the specific NRC requirements entails use of significant resources from both the NRC staff and the applicants. To the extent that these resources are directed to coping with artifacts of the models, the resources are not really contributing to the safety of the plants. This inefficiency can be reduced by directing requirements to actual safety concerns.

More Effective Requirements

The dose calculation models of the Standard Review Plan include the instantaneous release of the radioactive material from the core to the containment. This unrealistic assumption has the advantages of simplifying the calculations, being conservative, and avoiding the difficult question of just how quickly a release might occur. The disadvantages include the imposition of unnecessarily severe requirements on various systems. The requirements include valve closure times and startup times for air cleaning systems such

as containment sprays and standby gas treatment systems. These requirements obviously affect system design but they also are reflected in the technical specifications, which means the requirements continue to affect maintenance, surveillance, and test requirements throughout the life of the plant.

As mentioned earlier, the dose calculations affect the requirements for a number of systems. The dose calculations play the important role of providing a basis for interrelating requirements for several systems. The principal result has been a high level of safety. A secondary result, however, has been the imposition of requirements that are not fully justified by safety considerations. Thus there is an opportunity to establish more effective requirements by more explicitly addressing severe accident issues.

V. The Decoupling Effort

The NRC has initiated an effort to decouple design requirements from the dose criteria of Part 100. Actually, the goal is to separate design requirements and siting requirements as completely as practicable. Ideally, a proposed site could then be approved independent of the reactor design, a standard reactor design could be certified without regard to the site, and the specific licensing process would be greatly simplified. In practice, complete decoupling of site and design appears impracticable. Site characteristics affect design through soil parameters, seismic considerations, ultimate heat sink availability, source of cooling water, etc. A single design that would fit all acceptable sites, if feasible, would be unduly expensive to construct. Even if decoupling cannot be complete, avoiding the use of dose calculations to establish design requirements seems attainable and a step in the right direction.

The Source Term Issue

Accident consequence calculations generally begin with the calculation or assumption of a release of radioactivity from the core to the containment atmosphere. This release is called the "source term" and it is a major factor in the calculated doses. The characteristics of a source term are (1) the fraction of each nuclide released, (2) the rate of release, and (3) the chemical form of the released material.

It is difficult, if not impossible, to accurately predict the source term for a severe accident. This is true because the release is affected by factors such as the pre-accident condition of the fuel, the rates of heat-up and cool-down, the temperatures reached, and the fuel's environment. Furthermore, the source term will be different for each accident. Fission product release experiments, beginning with the work of Spedding et al. in 1942 [24], established some basic principles, e.g.:

1. Essentially all the noble gases will escape if the cladding is damaged, even at normal operating temperatures;
2. Halogens also escape readily but the rate of release is affected by conditions such as temperature, rate of change in temperature, and the presence of water;
3. Nuclides of "intermediate volatility" (cesium and tellurium) also escape readily if the temperature is slightly elevated;
4. Other nuclides (Ru, Tc, & Mo) are volatile under highly oxidizing conditions; and
5. Other elements of low volatility, especially the "refractory" group show little propensity for escape.

After the Windscale accident in 1957 [25], radioiodine received special attention. Radioiodine, especially iodine-131, is an important contributor to radiation dose because it tends to escape readily from the fuel and because it concentrates in a very small organ, the thyroid. In 1958, Brewer advanced the idea that iodine would not readily escape from the fuel in a large power reactor because it would react with cesium and form the non-volatile compound cesium iodide [26]. The theory did not seem consistent with experience in normal operations [27] or in accidents such as those at Chalk River, Hanford and SL-1. Consequently the theory did not garner great support.

By the time TID-14844 was written, it was clear that a wide range of accidents were possible and that it would be impractical to deal with different source terms for each accident. Accordingly, a single source term was postulated for use in design basis accident dose calculations. The TID source term (100% of the noble gases, 25% of the iodine, and 1% of others) was much smaller than the total release postulated for WASH-740, but still large enough to be considered sufficiently conservative for licensing purposes. The TID did not address the chemical form of the radioiodine. When credit was given for air cleaning by containment sprays, it was appropriate to assume some fraction of the iodine to be in organic form and so resistant to the effects of the sprays. The Safety Guides established the organic fraction at 4%, a value which seems conservative for fresh fission products but not for older releases [27].

After the TMI accident, a concerted effort was made to get the NRC to reevaluate the source term giving credence to the cesium iodide theory [28]. The NRC staff has done considerable research in the source term area, looking at both fission product releases and accident conditions. The results are less encouraging than some people had expected. Some results are consistent with the cesium iodide theory but there are no definitive results [29]. Furthermore, it seems that some accident sequences are capable of releasing as much iodine as the TID source term plus a significant quantity of cesium and other particulates. A contractor is now working on the development of a new source term. The outcome is uncertain but iodine seems likely to be an important part of the inventory.

Explicit Site Requirements

Part 100 is entitled "Reactor Site Criteria" but in its present form, it impacts design more than site selection. The NRC staff believes the present site requirements are adequate, but some criteria are in Regulatory Guide 4.7 [25] rather than in a regulation. Furthermore, the emergency preparedness requirements influence site selection. Therefore, it seems desirable to consolidate the site requirements in a revised Part 100. It is expected that the revision will include elimination of dose criteria for design basis dose calculations.

New Engineered Safety Feature Design Requirements

If the dose criteria are removed from the regulations, other provisions must be made for the numerous design requirements now based on the dose calculations. This is expected to be a formidable task because so many systems are affected, there is considerable uncertainty about the new designs, and an attempt is being made to maintain some of the flexibility inherent in the dose criteria. The development of these requirements is further complicated by the fact that the dose criteria now provide the bases for numerous technical specifications and play a central role in the enforcement process.

The new approach and new requirements are intended to be for the new plants. It is not likely that backfit rule requirements could be met for making the new requirements applicable to existing plants. There is, however, the possibility that existing plants will want to take advantage of some of the new provisions.

Efforts to develop these new requirements are just beginning and it is not yet clear that the effort will be successful. The "fallback position" is that dose criteria could be introduced into Part 50. This could constitute some improvement over the present situation in that the design basis role of the dose criteria would be made clear.

Implications for Air Cleaning

Air cleaning systems are expected to continue to play an important role in the safety of nuclear power plants. The precise nature of that role, however, is likely to be significantly different for the next generation of plants. It is not yet clear just what changes may occur but it is important to recognize that changes are being made.

VII. Summary

New designs for nuclear power plants are being advanced and the licensing system is undergoing a major change. There will be changing demands on air cleaning, but there are no clear indications as to what those changes will be.

References

1. *"Safety Goals for the Operation of Nuclear Power Plants," Federal Register, 30028, August 21, 1986.*

2. *Beck, C. K., et al., "Theoretical Possibilities and Consequences of Major Accidents in Large Nuclear Power Plants," WASH-740, March 1957.*

3. *Rasmussen, N., et al., "Reactor Safety Study: An Assessment of Accident Risks In U. S. Commercial Nuclear Power Plants," WASH-1400, October 1975.*

4. *"Severe Accident Risks: An Assessment for Five U.S. Nuclear Power Plants," NUREG-1150, Draft for Peer Review, June 1989.*

5. *"Individual Plant Examination: Submittal Guidance, Final Report," NUREG-1335, August 1989.*

6. *"Procedural and Submittal Guidance for the Individual Plant Examination of External Events (IPEEE) for Severe Accident Vulnerabilities," NUREG-1407, Draft Report for Comment, July 1990.*

7. *"Part 100 -- Reactor Site Criteria," Federal Register, pg 3609-11, April 12, 1962.*

8. *"Summary Report of the Reactor Safeguards Committee," WASH-3, Atomic Energy Commission Report, 1950.*

9. *"10 CFR Chapter 1, Notice of Proposed Rule Making," Federal Register, May 23, 1959.*

10. *"10 CFR Part 100, Notice of Proposed Guides," Federal Register, pg 1224-6, February 11, 1961.*

11. *DiNunno, J. J., F. D. Anderson, R. E. Baker & R. L. Waterfield, "Calculation of Distance Factors for Power and Test Reactor Sites," Atomic Energy Commission Report TID-14844, March 23, 1962.*

12. *"Assumptions used for Evaluating the Potential Radiological Consequences of a Loss of Coolant Accident for Boiling Water Reactors," Regulatory Guide 1.3, Rev.2, June 1974.*

13. *"Assumptions Used for Evaluating the Potential Radiological Consequences of a Loss of Coolant Accident for a Pressurized Water Reactor," Regulatory Guide 1.4, Rev. 2, June 1974.*

14. *"Standard Review Plan for the Review of Safety Analysis Reports for Nuclear Power Plants, LWR Edition," Nuclear Regulatory Commission Report NUREG-800, July 1981 (Originally issued as NUREG-75/087).*

15. *Thompson, T. J. and J. G. Beckerley, Eds., The Technology of Nuclear Reactor Safety, Vol. 1, 1964 & Vol. 2, 1973, Cambridge, MA: MIT Press.*

16. *"Standard Format and Content of Safety Analysis Reports For Nuclear Power Plants (LWR Edition)," Nuclear Regulatory Commission Regulatory Guide 1.70, Rev. 0, February 1972; Rev. 3, November 1978.*

17. *Bunch, D. F., "Metropolitan Siting -- A Historic Perspective," Nuclear Regulatory Commission Report NUREG-0478, October 1978.*

18. *"General Site Suitability Criteria for Nuclear Power Stations," Nuclear Regulatory Commission Regulatory Guide 4.7, Rev.0, Sept. 1974; Rev. 1, November, 1975.*

19. *"Criteria for Preparation and Evaluation of Radiological Emergency Response Plans and Preparedness in Support of Nuclear Power Plants," NUREG-0654, Rev. 1, October 1980, Supplement 1, November 1987.*

20. *Webb, R. E., The Accident Hazards of Nuclear Power Plants. Amherst, MA: U. Mass. Press, 1976.*

21. *Bassett, T. G., "A Hazard Evaluation of the Ford Nuclear Reactor," Unpublished Masters Thesis, University of Michigan, May 1957.*

22. *Hart, R. S., et al., "Final SNAPSHOT Safeguards Report," NAA-SR-10022, (CRD) July 1964.*

23. *Willis, C. A., "Statistical Safety Evaluation of Power Reactors," AEC Report AI-65-TDR-212, October 1965.*

24. *Spedding, F. H., et al., "Removal of Fission Products from Molten Uranium by Diffusion and Chemical Reactions," U. of Chicago Metallurgical Lab. Report MUC-NS-3068, 1942.*

25. *Horan, J. R. & W. P. Gammill, "Health Physics Aspects of the SL-1 Accident," Health Physics, 9, pp 177-186, 1963.*

26. *Brewer, L., "Vaporization Processes in a Runaway Reactor," General Atomic Report GAMD-919, August 1959.*

27. *Willis, C. A., et al., "Monitoring Gaseous Radioiodine at Power Plants," 19th DOE/NRC Nuclear Air Cleaning Conference, NUREG/CP-0086, May 1987.*

28. *Stratton, W. R., A. P. Malinauskas and D. O. Campbell, Letter to NRC Chairman J. Ahearne, August 14, 1980.*

29. *Silberberg, M., et al., "Reassessment of the Technical Bases for Estimating Source Terms," NUREG-0956, July 1986.*

AIR CLEANING TECHNOLOGIES FOR THE MANAGEMENT AND DISPOSAL OF RADIOACTIVE WASTES

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Abstract

Air cleaning systems are a mandatory and essential part of most all portions of the processes and activities involved in the management and disposal of radioactive wastes. Such systems are integral to the compaction, solidification, adsorption and/or evaporation of low level wastes, and to the consolidation, solidification, transportation and ultimate disposal of high level wastes. With the need to reduce the volumes of low level wastes, due to the dramatic increases in disposal costs, and with the delays that have occurred in the confirmation of plans for the development and operation of a geologic repository for the disposal of high level wastes, the demands on air cleaning systems are likely to increase. Compounding this situation are regulatory changes, such as those being proposed by the Environmental Protection Agency relative to their National Emission Standards for Hazardous Air Pollutants: Radionuclides. This continues to be a challenging field with much work remaining to be done. Since it will be beneficial to all concerned, efforts to establish sound mechanisms for the feedback of operating experience, and to encourage the maximum exchange of information on this subject at the international level, should continue.

I. Introduction

Air cleaning systems are an essential part of most all portions of the processes for the management and disposal of radioactive wastes. In the case of low level wastes, solids are generally shredded and compacted and/or incinerated, liquids are concentrated by ion exchange or evaporated with the resins and/or residues being solidified, and gases and airborne particulates are removed by adsorption or filtration. Each of these steps has an associated need for an air cleaning system. In the case of high level wastes, spent fuel and liquid wastes are currently stored on-site. Plans call for these wastes to be suitably treated, as necessary, and later transported to a monitored retrievable storage facility for interim storage, followed by final disposal in a geologic repository. As in the case for low level wastes, each stage of this process includes some requirement for an air cleaning system (Figure 1).

II. Low Level Radioactive Wastes

One of the basic air cleaning systems employed in the initial handling of low level radioactive wastes is that associated with equipment designed to collect dry radioactive dust and debris, or to remove sludge and other types of radioactive deposits from the bottoms of water pools and storage basins. The latter units have an initial drum for collecting the liquids and associated solids, followed by three stage filters for removing radioactive material in the accompanying air (vacuum) flow. One such system includes a disposable 16-layered cellulose fiber dust bag as a first filtration stage to entrap large particles of debris or dirt that may be carried over from the primary collection tank or silo. The second filtration stage is a 10 micrometer polyester prefilter that provides protection for a final pleated HEPA filter (Figure 2).[1]

LOW-LEVEL RADIOACTIVE WASTES

Solids:	Shredding, Compaction and/or Incineration
Liquids:	Ion Exchange or Evaporation Followed by Solidification of Resins or Residues
Gases and Airborne Particles:	Adsorption or Filtration

HIGH-LEVEL RADIOACTIVE WASTES

Spent Fuel:	On-Site Pool or Dry Cask Storage With or Without Consolidation
Liquids:	Tank Storage, Conversion to Salt Cake or Vitrification
Transported Waste:	To Monitored Retrievable Storage; To Geologic Repository

Figure 1 Steps in the management and disposal of radioactive wastes that require air cleaning support.

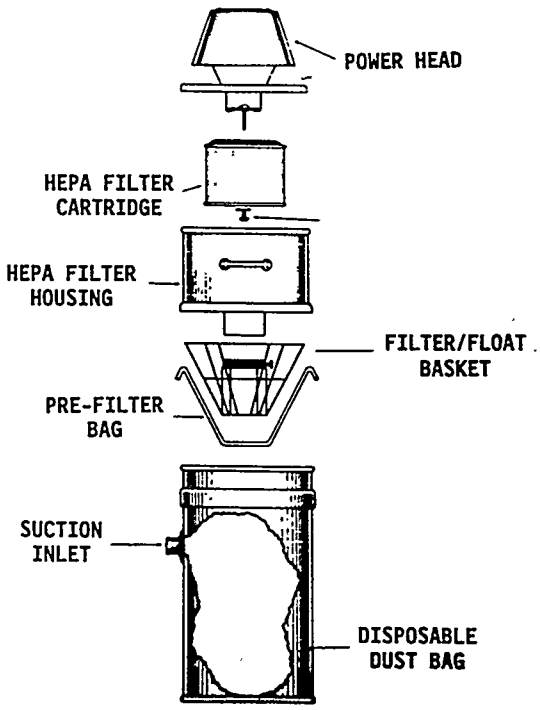

Figure 2 Air cleaning units for nuclear grade vacuum system.[1]

Once collected, low level wastes must generally be treated prior to disposal. To assure that the volumes are minimized, many waste producers today shred, compact and/or incinerate their solid low level radioactive wastes. Each of these processes has associated air cleaning requirements. Shredding and compaction are generally conducted in an enclosure with a local exhaust ventilation system equipped with filters designed for the retention of particulates and radioiodine. Where there is a potential for release of airborne materials, workers must be provided with protective clothing and air supply respirators.

The primary goal in the incineration of low level radioactive wastes is to achieve essentially complete combustion of the wastes with as total retention as possible of the accompanying radionuclides. This represents a special problem for mixed wastes that contain both radioactive materials and toxic chemicals. Newer technologies range from the common types of excess and controlled air incinerators to the advanced concepts of high-temperature slagging, pyrolysis, and acid digestion (Figure 3).[2] All such units include off-gas scrubbing and particulate filtration systems to treat the associated airborne releases. Such systems have four principal functions: (a) particle control, which generally includes scrubbers and/or filters; (b) gas control, which may include both wet and dry gas adsorption processes; (c) temperature control, which may include water quenching, cooling through a heat exchanger, or the addition of ambient air to the off-gas stream; and (d) moisture control, which includes demisters and follow-up heaters for humidity control.

Systems for the treatment of the off-gases are a necessary adjunct to a low-level radioactive waste incineration facility and their design is a challenging, but feasible, engineering task. The incineration process, itself, makes air cleaning a complex operation since, in addition to the radionuclides and other hazardous materials, there are large quantities of volatile gases, water vapor, fly ash and other combustion products that must be removed from the off-gas stream. Furthermore, because the off-gas stream leaves the final combustion chamber at high temperature, it usually must be at least partially cooled before it is permitted to enter the air cleaning system. This is especially true if the air cleaning system is a non-aqueous type that includes heat sensitive HEPA filters.[2] Detailed information on current designs for off-gas cleaning technologies for use in incinerator facilities for low level radioactive wastes has been provided in a recent publication by the International Atomic Energy Agency.[3]

Low level liquid radioactive wastes are generally treated either by evaporation or ion exchange. Where evaporation is practiced, there is a need for entrained moisture separators. The facilities in which the residues and/or ion exchange resins are solidified must also be equipped with proper air cleaning systems.

III. High Level Radioactive Wastes

The major sources of high level wastes in the U. S. today are spent fuel from operating nuclear power plants and liquid wastes generated in the chemical processing of spent fuels from military operations (such as at Savannah River, Idaho and Hanford) and certain past civilian operations (such as at West Valley). In the main, the spent fuel generated as a result of civilian nuclear operations is being stored in water pools at the power plants sites. Such pools are within the containment or fuel handling building and are therefore served by the safety grade air cleaning systems associated with those facilities. With spent fuel pools at many nuclear plants now reaching full capacity, several nuclear utilities are designing and constructing dry cask containers for storing spent fuel onsite. Each such container is provided with an inert inside gas cover that is monitored for leaks.

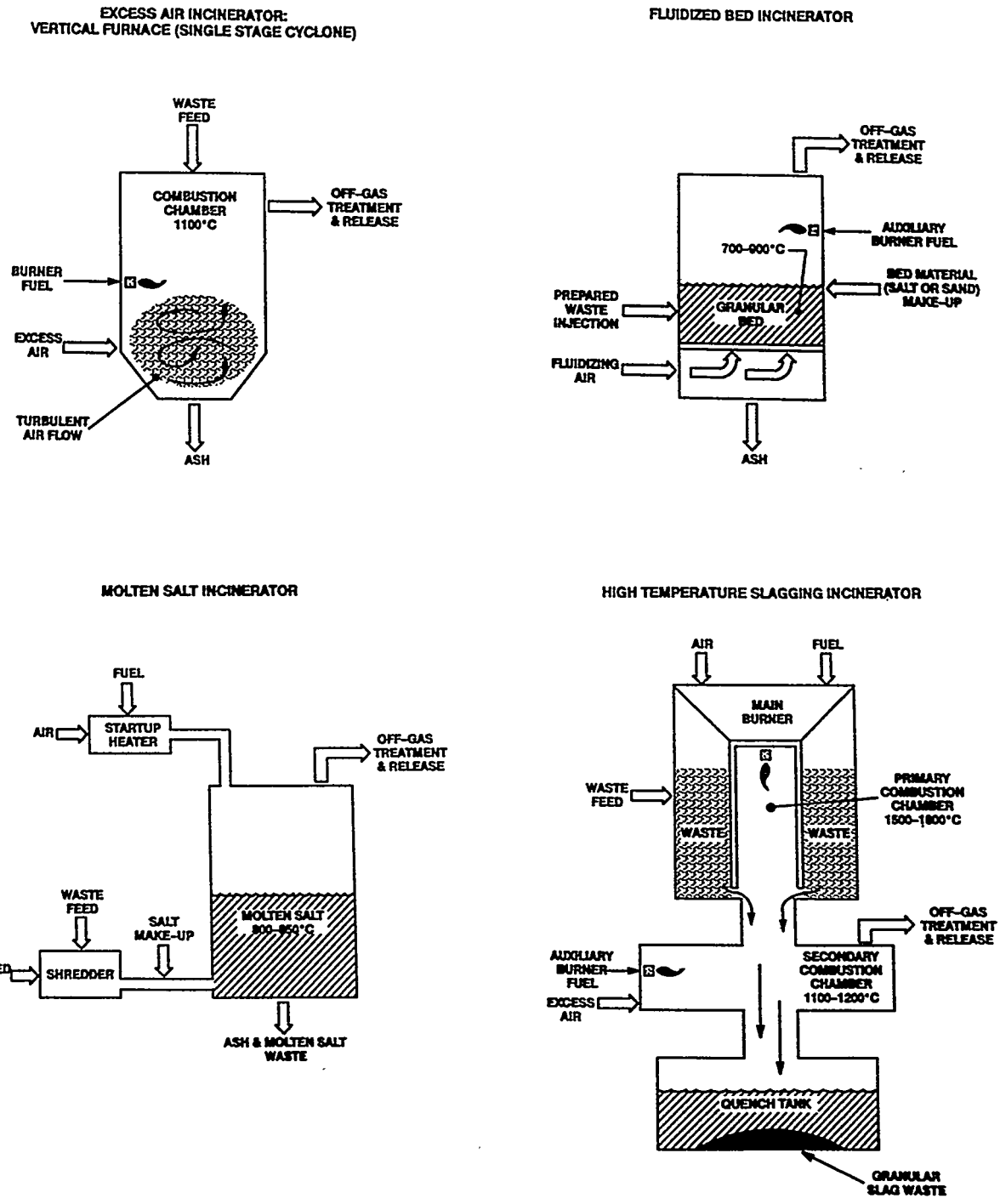

Figure 3 Selected types of incinerator technologies.

High level radioactive liquid wastes, such as those at West Valley and those produced by the chemical processing of spent fuel from military operations, are being stored in large underground tanks. Because such wastes initially generate a considerable amount of heat, the tanks in which they are stored must be cooled and equipped with an air cleaning system. This includes a forced air purge to maintain hydrogen and flammable vapor concentrations well below the lower explosive limit. Such systems generally include a refrigerated condenser to remove condensible vapors, and a high-efficiency mist eliminator to capture aerosol particles and entrained water before the off-gas enters a final HEPA filter.[4] Shown in Figure 4 is the system used on the high level waste tanks at Savannah River.[5] An emergency backup system is provided in case of failure of the primary system. Heated air is also supplied to the annuli of all double wall tanks to prevent condensation and minimize corrosion. The warm air also helps retard seepage whenever small cracks develop in the primary tank wall by causing salt encrustation to form around the point of leakage. The exhaust from this system is filtered to avoid dispersal of radioactive material to the atmosphere.[5]

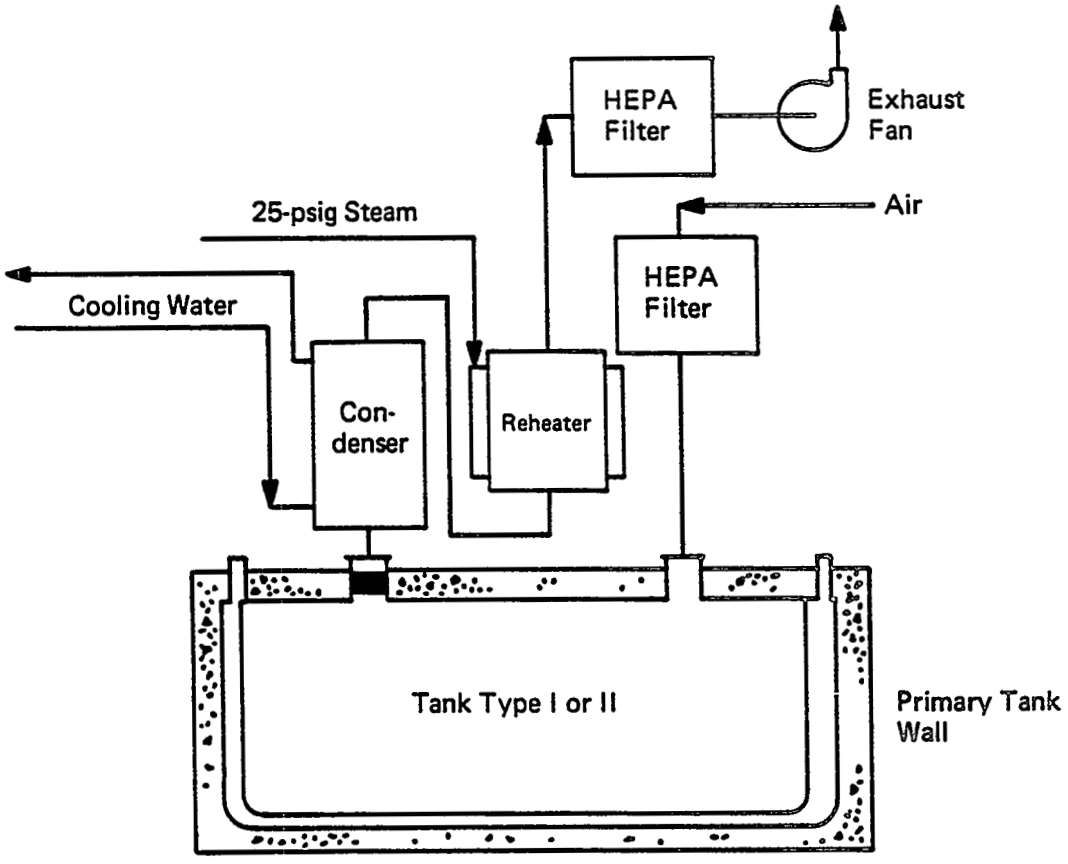

Figure 4 Ventilation system for high-level liquid waste tanks at Savannah River.[5]

As previously mentioned, plans call for high level liquid wastes subsequently to be solidified for ultimate disposal in a geologic repository. In most cases, such solidification will involve vitrification of the wastes into borosilicate glass. This process will generate airborne releases which will require an elaborate air cleaning system. Such a system will include a film cooler, submerged bed scrubber, and high efficiency mist eliminators, followed by double HEPA filters and a catalytic system for removal of the oxides of nitrogen (Figure 5).[6] The film cooler maintains a clear and unobstructed line between the melter and the scrubber by injecting air and steam into the off-gas line. The submerged bed scrubber consists of a packed bed submerged in a water tank. The scrubber condenses the process stream and captures the gross particulates. The mist eliminator removes fine particulates and droplets of liquid. The off-gas is then sent through a heater/HEPA filter assembly. Prior to entering the HEPA filters, the off-gas is heated above the dew point to prevent any condensation.[6]

The melter off-gas system at Savannah River normally flows at a rate of about 400 cfm and consists of micrometer sized particles of radioactive materials, plus mercury, steam and noncondensible gases. The main radioactive components of the melter feed are ^{137}Cs and ^{90}Sr. Two to three percent of the ^{137}Cs fed to the melter vaporizes and condenses in the off-gas as a 0.2 to 1 micrometer aerosol. Strontium-90 is emitted from the melter as entrainment. The treatment equipment is designed to have an overall decontamination factor of about 8×10^8 and it reduces the releases of ^{137}Cs to less than 3 mCi per year. The equipment is designed to operate continuously because, even during the idle mode when there is no feed, the melter generates a considerable volume of contaminated off-gas.[5]

The air cleaning system for the melter at Savannah River consists of two parallel trains, one providing primary cleanup and one serving as a backup. Treatment of the off-gas is switched to the backup system whenever any portion of the primary system is down for replacement or repair. Automatic switchover occurs if the primary off-gas line becomes plugged or if power is lost to either the primary exhauster or primary quencher pump. To provide protection against accidental releases and enhance the overall removal efficiency, the system at Savannah River includes a final sand filter.[5]

As an initial step, high level liquid wastes at the Idaho Chemical Processing Plant are being reduced in volume and solidified using a waste calcining process. The wastes will later be converted into a more durable solid form for ultimate disposal. The off-gas system for the waste calcining facility is designed to remove particulates and ruthenium and begins with the off-gas passing from the calciner through baffle plates to a primary cyclone. A quench tower cools the off-gas using sprays of recycled scrub solution, and a high-efficiency venturi scrubber is used to remove the particulates. The off-gas then passes through a centrifugal separator followed by a condenser, a mist eliminator, and three parallel adsorber beds. Selective catalytic reduction provides for removal of the oxides of nitrogen. A second mist eliminator collects any condensed moisture that is carried over from the adsorbers, and a heater is used to increase the temperature of the gas before it enters the four remotely removable parallel prefilter/HEPA filter units that provide final cleanup.[7][8]

Of considerable interest is the fact that scientists at the Pacific Northwest Laboratories are proposing that the vitrification process be used not only to solidify high level liquid radioactive wastes that are in storage, but also for the in situ vitrification of low level concentrations of radionuclides in the ground (earth). This is accomplished by inserting four electrodes into the contaminated sections of the ground, and then passing sufficient current through them to vitrify the earth and the accompanying radionuclides. A portable diesel generator provides power to a trailer-mounted mobile system that includes an off-gas containment hood that is placed over the area being treated. Housed in the trailer unit is an off-gas removal system. It includes a

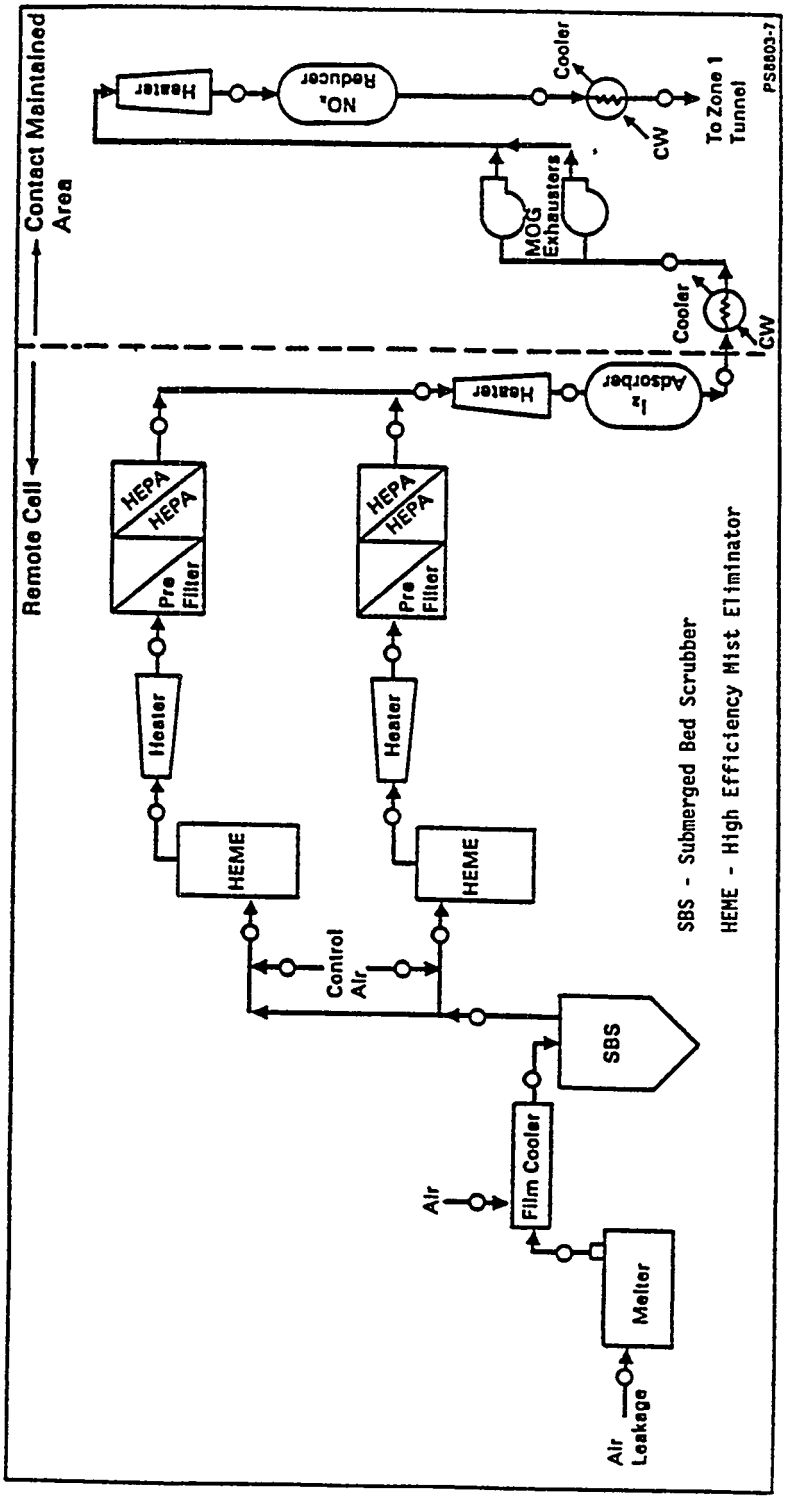

Figure 5 Air cleaning system for a high level waste vitrification facility.(6)

venturi-ejector scrubber and separator, a Hydro-Sonic scrubber, separator, condenser, another separator, heater, two stages of HEPA filters, and a blower (Figure 6).[9] Liquid to the two wet scrubbers is supplied from two independent recirculation tanks, each equipped with a pump and heat exchanger. Data show higher than 99% retention of all radionuclides within the vitrified zone; soil-to-off-gas decontamination factors averaged greater than 4000 for the transuranic elements, and ranged from 130 for ^{137}Cs to 3100 for ^{90}Sr.[9]

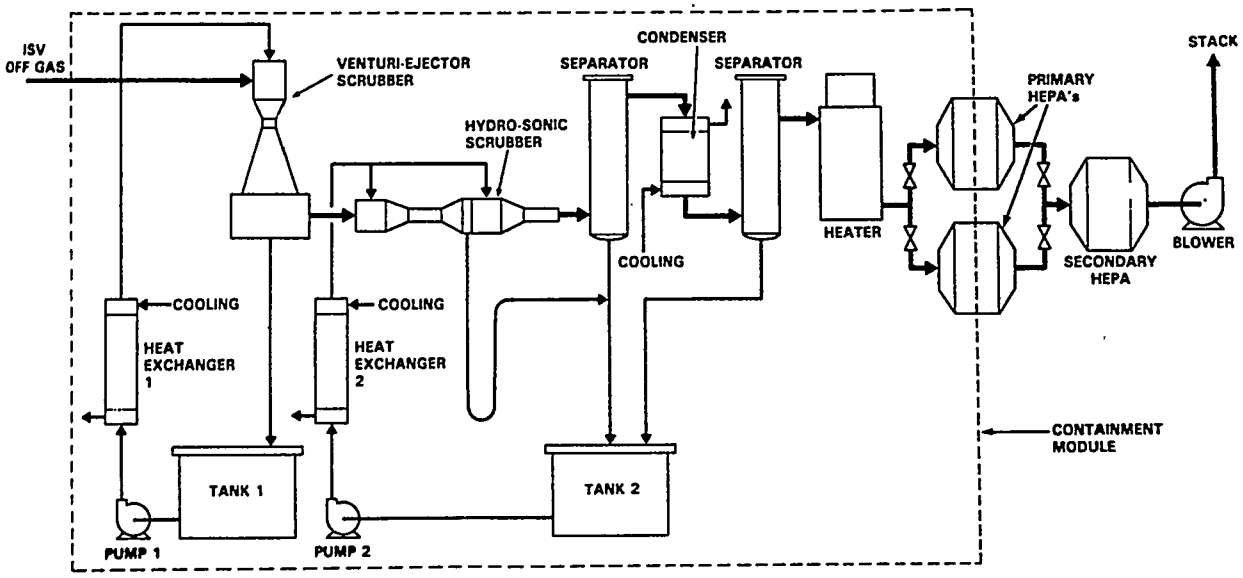

Figure 6 Air Cleaning System for <u>In Situ</u> Waste Vitrification Process.[9]

As mentioned above, plans call for solidified high level wastes and the spent fuel from commercial nuclear plants to be placed in a geologic repository for final disposal. As an interim step, these wastes may be transported to a monitored retrievable storage (MRS) facility for storage and processing. One of the steps that may be applied to the spent fuel is consolidation. This will include taking the fuel assemblies apart and consolidating the fuel rods into smaller packages. Since this could result in some release of the volatile fission gases, it will have to be performed in a facility equipped with an off-gas treatment system, capable of retaining airborne particulates, radioiodine and fission gases.

In addition, there will be air cleaning considerations associated with the transport of certain types of wastes to a geologic repository. One example is the container currently under development for the transport of transuranic (TRU) wastes to the Waste Isolation Pilot Plant in New Mexico. As part of this container, a filtered pressure equalization system has been developed that is designed to equalize the pressure drop across the structure of the container package while, at the same time, it effectively contains any airborne particles present in the released gas (Figure 7).[10] Use of a filtered vent allows for a more lightweight containment vessel and limits the buildup of hydrogen.

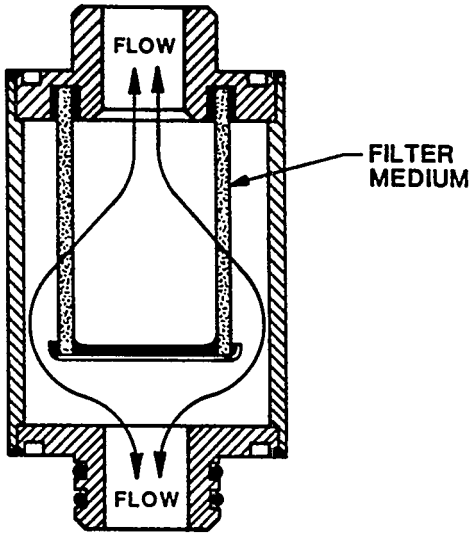

Figure 7 Glass Fiber Two-Way Filter Cartridge for Use in TRUPACT-I Shipping Container.[10]

Use of such a vent, however, leads to certain complications. For example, atmospheric pressure variations due to changes in the weather or elevation, temperature changes of the internal gas, or pressure buildup due to gas generation by the waste contents, may create pressure differences in the container. Since the cited factors may cause either a positive or negative pressure across the waste package, the accompanying filter is required to be capable of accommodating flow in either direction. Particle retention is especially important for flow from inside the package to the outside. Further complicating this situation is that the filter must perform its required functions under a full range of accident conditions. These include shock, vibration, pressure, heat, cold, moisture, and exposure to chemical substances. Initial tests indicate that a filter with a glass fiber filtration medium may meet these requirements.[10]

Access to the Waste Isolation Pilot Plant for placement of the TRU wastes is through several vertical shafts. These shafts also provide for ventilation of the facility. Some of the challenges in the design and construction of this facility have included the development of a monitoring system that collects a representative sample from the exhaust stack,[11] and setting up an air cleaning and monitoring system that can handle mass concentrations that range from 300 ug to 50 mg per cubic meter, at flow rates varying from 33 to 200 cubic meters per second.[12]

The candidate site for the nation's first geologic repository for the disposal of spent fuel and solidified high level wastes has been proposed for Yucca Mountain, Nevada. Associated air cleaning requirements will include those for the waste handling building and for the repository, itself. As currently proposed, ventilation of the repository at Yucca Mountain will be provided through two exploratory shafts, vertical openings extending from the earth's surface down into the repository. Current plans are to transport the wastes into the repository through a "Waste Ramp" that leads in through an opening in the side of the Mountain (Figure 8).

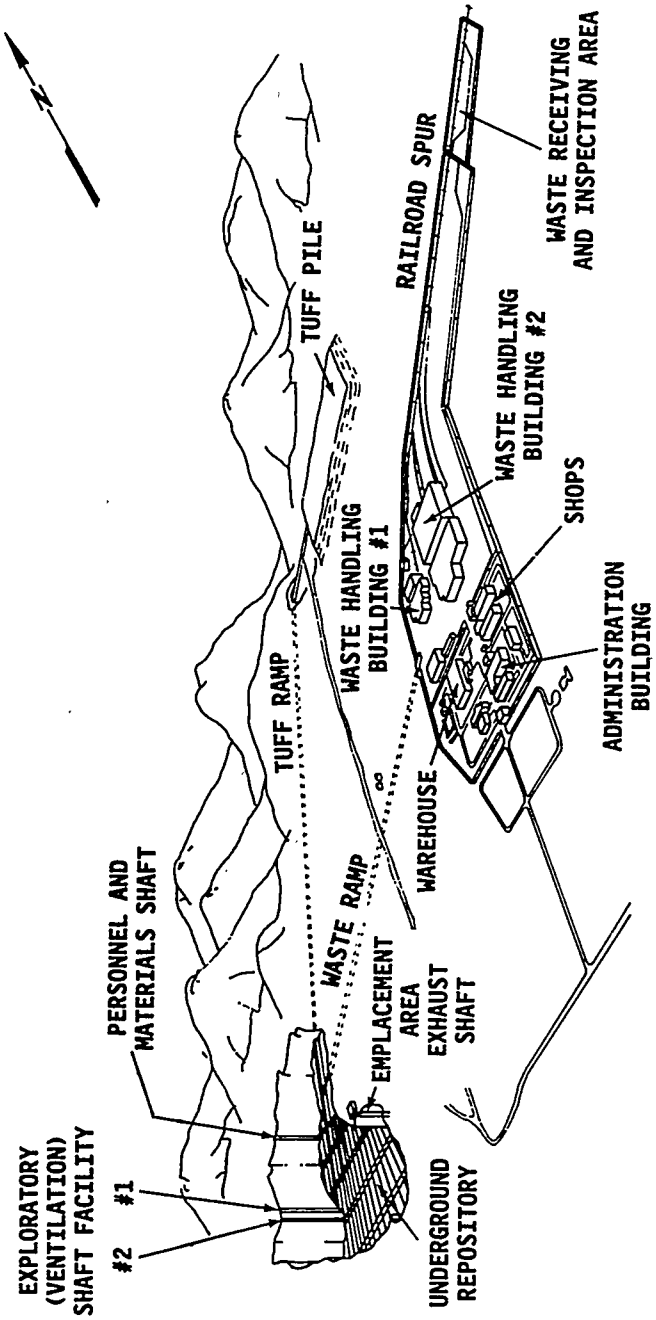

Figure 8 Overview of support facilities for proposed geologic repository at Yucca Mountain.

IV. Uranium Mill Tailings

Many nuclear air cleaning specialists are familiar with the problems cited above. There are, however, other challenges in the waste management field. One of these involves the development of programs for the stabilization of, and control of airborne releases from, tailing piles that have been generated as a result of various mining and milling operations in the U. S. These include the processing not only of uranium, but also of phosphate ores and lignites.

Under the regulations established by the U. S. Environmental Protection Agency,[13] the following conditions must be met with respect to airborne releases of radon from uranium mill tailings:

1. The disposal site must be designed to control releases from the tailing piles for 1000 years to the extent reasonably achievable and, in any case, for at least 200 years; and

2. The disposal site design must prevent the release of ^{222}Rn at rates exceeding an average of 20 pCi per square meter per second, or from increasing the annual average concentration of ^{222}Rn in the air outside the disposal site by more than 0.5 pCi per liter.

In order to meet these requirements, the U. S. Department of Energy (which has the responsibility for the remediation of uranium tailings piles) generally takes the following approaches:

1. The disposal area (pile) should be covered by a seven foot thick cap which includes an infiltration/radon barrier to reduce radon emissions and to limit groundwater infiltration; and

2. Above this should be placed a sand filter layer, a thick layer of compacted select fill for frost protection; a filter and bedding layer to promote lateral run-off; and a rip-rap layer to protect against erosion.

After completion of this remedial action, the disposal site must be fenced and posted with appropriate warning signs to discourage human intrusion. In addition, the site must be surveyed and monitored periodically by a custodial agency licensed by the U. S. Nuclear Regulatory Commission.

V. Commentary

As may be noted by the above review, there is hardly a facet of the operations involving the management and disposal of low level or high level radioactive wastes that does not require extensive air cleaning support. These requirements range from the more common systems for the removal of airborne particles and the adsorption of gases generated in facilities where low and high level wastes are being handled and/or processed, to the requirements associated with the application of controls to the releases of ^{222}Rn from uranium mill tailings piles. Changes pending in certain Federal regulations, such as the "National Emission Standards for Hazardous Air Pollutants: Radionuclides" of the U. S. Environmental Protection Agency,[14] hold promise for requiring even greater reductions in airborne emissions from waste handling activities.

Although nuclear air cleaning specialists have met many of these challenges in an excellent manner, problems remain. If these problems are to be solved, there is a need for better reporting and feedback of operating experience, and for an increased exchange of information with experts in other countries who are facing similar problems. Examples of the latter include the recently released summary report of waste management activities in the U.S.S.R.,[15] several reports issued under the auspices of the International Atomic Energy Agency,[16][17] and a report from the United Kingdom on the use of foams for the removal of radioactive dust from gas streams.[18] Although the latter approach had been extensively investigated in the U.S. several decades ago, the U.K. paper proposes new approaches such as the use of the process gas to generate the foam. As demands for a cleaner environment increase, compounded by other factors such as the potential impacts of the BEIR-V report,[19] there will be demands for air cleaning systems that are more effective and efficient. At the same time, air cleaning specialists must not lose sight of the fact that certain components and processes incorporated into cleanup systems often add to the volume of nuclear wastes being generated. These include spent filters, the solid residues collected in scrubbers, residual charcoals from off-gas systems, and associated ductwork.

Acknowledgments

The author wishes to thank Andrew J. Bieniawski, Intern Fellow, and Stewart W. Long, Fellow, Advisory Committee on Nuclear Waste/Advisory Committee on Reactor Safeguards, U. S. Nuclear Regulatory Commission, for their help in the development of the background material used in this paper.

References

1. "Nuclear waste vacuum systems," Power Products and Services, Inc., P. O. Box 207, Forest, VA (1990).

2. Long, S. W., "The incineration of low level radioactive waste," Advisory Committee on Nuclear Waste, Report NUREG-1393, U. S. Nuclear Regulatory Commission, Washington, DC (June, 1990).

3. "Treatment of off-gas from radioactive waste incinerators," Report Series No. 302, International Atomic Energy Agency, Vienna, Austria (1989).

4. Brouns, T. M., and Peterson, M. E., "Performance evaluation of the pilot -scale, double-shell tank ventilation system using simulated aerosol streams," Report PNL-7188, Pacific Northwest Laboratories, Richland, WA (December, 1989).

5. Private Communication, Donald A. Orth, Westinghouse Savannah River Company, Aiken, SC (May, 1990).

6. Perez, J. M., Jr., Goles, R. W., Nakaoka, R. K., Sevingy, G. J., Larson, D. E., and Kruger, O. L., "Liquid-fed ceramic melter testing for the Hanford waste vitrification plant," paper presented at the Annual Meeting of the American Ceramic Society, Indianapolis, Indiana (April 23 - 27, 1989).

7. Bingham, G. E., and Wheeler, B. R., "A production facility for the solidification of high-level radioactive wastes," Special Report, Allied Chemical Corporation, Idaho Chemical Programs, Idaho Falls, Idaho (1974).

8. May, K. W., and Loo, H. H., "A remotely replaceable and testable off-gas filter system for the new waste calcining facility," Proceedings of the 26th Conference on Remote Systems Technology, pages 250 - 254 (1978).

9. Oma, K. H., and Timmerman, C. L., "Off-gas treatment and characterization for a radioactive in situ vitrification test," Proceedings of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, Report CONF -840806, Vol. 1, pages 683 - 701 (March, 1985).

10. Kincy, M. A., Joseph, B. J., and Humphreys, D. L., "TRUPACT I filter development status report," Report SAND88-3271, Sandia National Laboratories, Albuquerque, NM (June, 1989).

11. Rodgers, J. C., "Exhaust stack monitoring issues at the Waste Isolation Pilot Plant," Report EEG-37, Environmental Evaluation Group, Health and Environment Department, State of New Mexico (November, 1987).

12. Shenk, K. J., "The development of the airborne radioactivity effluent monitoring system for the Waste Isolation Pilot Plant," Proceedings of the 20th DOE/NRC Nuclear Air Cleaning Conference, Report CONF-880822, Vol. 2, pages 805 - 813 (May, 1989).

13. U. S. Environmental Protection Agency, "Health and environmental protection standards for uranium mill tailings," Title 40, Part 192, Code of Federal Regulations, Federal Register, Vol. 48, No. 3 (January 5, 1983).

14. U. S. Environmental Protection Agency, "National emission standards for hazardous air pollutants: radionuclides; Final Rule and Notice of Reconsideration, Title 40, Part 61, Code of Federal Regulations, Federal Register, Vol. 54, No. 240 (December 15, 1989).

15. Nikiforov, A. S., Nikipelov, B. V., and Kulichenko, V. V., "Policy and principles of radioactive waste management in U.S.S.R.," State Committee for Utilization of Atomic Energy, Paper presented at the 1989 Joint International Waste Management Conference, Tokyo, Japan (1989).

16. "Design and operation of off-gas cleaning and ventilation systems in facilities handling low and intermediate level radioactive material," Internal Atomic Energy Agency, Vienna, Austria (1988).

17. "Design and operation of off-gas cleaning systems at high level liquid waste conditioning facilities," International Atomic Energy Agency, Vienna, Austria (1988).

18. Mitchell, J. P., "Decontamination by foams: A promising treatment for the removal of radioactive dust from gas streams," Report AEEW-R2520, Chemistry Division, Winfrith Technology Centre, United Kingdom (June, 1989).

19. "Health effects of exposure to low levels of ionizing radiation," Committee on the Biological Effects of Ionizing Radiation, BEIR-V report, National Academy of Sciences, National Academy Press, Washington, DC (1990).

DISCUSSION

FIRST: I was very interested in observing the details of your slides detailing nuclear waste preparation steps that include incineration, vitrification, and so on. We note that there is always an air cleaning train that includes multiple stages of scrubbers, heaters, mist eliminators, and HEPA filters. For many years I, and I am sure others, have tried to point out to the technical people designing these nuclear waste processing systems that splitting the waste into four or five different process streams and then having to cope with the disposal of each of them gases, liquids, and solids at the end of the line makes no sense at all. Along with handling all of these waste streams, the operators must deal with very large complicated equipment that gets thoroughly contaminated and gives almost insurmountable maintenance problems. Surely, the way to go is to stay dry all the way and, thereby simplify the equipment and the disposal problems. Do you have anything to say on that subject?

MOELLER: I think you have said it very well.

THE PERMANENT DISPOSAL OF NUCLEAR FUEL WASTES IN CANADA

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Abstract

This paper summarizes the Canadian concept for the permanent disposal of nuclear fuel wastes in the plutonic rock of the Precambrian Canadian Shield. The concept uses a combination of engineered and natural barriers to ensure that the used fuel remains isolated from the environment. Waste vault designs are being evaluated using a probabilistic systems assessment code, which uses simplified models of the performance of the engineered systems, the geosphere, and the biosphere. The simplified models are based on more detailed models and experimental information. Research and development is being carried out to elucidate the important processes relating to the performance of the engineered and natural barriers, the stability of the waste form, and contaminant transport.

Introduction

Canada generates about 15% of its electricity from nuclear power using CANDU pressurized heavy water reactors. Most of this capacity is in the province of Ontario, which derives about 50% of its electricity from nuclear reactors. In addition, there are reactors in Quebec and New Brunswick. At the end of 1989, about 14 000 tonnes of used fuel had accumulated in Canada, and about 1800 tonnes are being produced each year. The used fuel is currently being stored at the reactor sites using both wet and dry storage methods.

For the past several years, Canada has been developing the technology to permanently dispose of its nuclear fuel wastes in deep geologic formations in the Precambrian Canadian Shield. The decision to focus on plutonic rock of the Canadian Shield as the medium for the disposal vault was made in the late 1970's.[1] Several considerations justify this decision: 1) The Canadian Shield has been relatively stable for at least 600 million years, and most of the Shield has not experienced major orogenic activity for 2.5 billion years. Therefore, it is reasonable to conclude that the region would remain stable for the required lifetime of a disposal vault. 2) Regional topographic gradients in the Shield are low and, therefore, the natural driving force for groundwater flow deep in the rock should be weak. 3) It is believed that there are large volumes of plutonic rock with extremely low porosity and permeability. This would limit groundwater flow to the waste form, and would inhibit movement of contaminants through the rock. In addition, minerals in plutonic rock are known to react with many of the radionuclides in nuclear fuel waste, which would further reduce their mobility. 4) The large area of the Canadian Shield, and the large number of exposed plutons, ensure that there would likely be considerable flexibility when selecting a site.

In 1981, the Canadian and Ontario governments approved a generic (non-site specific) research program to assess the permanent disposal of nuclear fuel wastes in plutonic rock of the Canadian Shield. The results of this generic research will be reviewed under the Federal Environmental Assessment and Review Process, which is now underway. The environmental impact statement for the disposal concept will be given a thorough review by technical experts, and by the public through public hearings. No effort relating to the selection of potential disposal sites can be initiated until a decision is made by the government following the review.

In this paper, we summarize the Canadian concept for the permanent disposal of nuclear fuel wastes. We also discuss some of the research and development work that is being used to characterize and evaluate the concept.

Conceptual Disposal Vault

A schematic of the conceptual disposal vault and details of a typical disposal room are shown in Figure 1.[2] The reference vault would occupy an area of about 4 km^2 and would consist of 480 disposal rooms, each of which would be 220 m long, 8 m wide, and 5.5 m high. The vault would be constructed at a depth between 500 and 1000 m, depending on the characteristics of the host rock mass. Such a design would accommodate about 190 000 tonnes of used CANDU fuel, which would be placed in thin-walled containers, as shown in Figure 2. The reference container has 6.35-mm thick titanium walls and holds 72 CANDU fuel bundles. Support against external pressure is provided by packing glass beads in the spaces between the fuel assemblies and the outer wall. Prototype containers of this design have withstood hydrostatic tests at external pressures greater than 10 MPa at 150°C and, therefore, they meet the structural requirements for disposal in a vault at a depth of 1000 m.[3] The design target for the containers is to isolate the used fuel from contact with groundwater for at least 500 years, the period of time required for most of the radionuclides to decay to negligible levels.

Each waste container would be placed in a 1.25-m diameter borehole in the floor of the disposal rooms, and would be surrounded by a compacted mixture of bentonite and sand. An alternative approach would be to place the containers directly in the rooms as opposed to using boreholes. Once the rooms were filled, they would be backfilled by compacting a mixture of clay and crushed granite in the lower portion of the room, and by pneumatically filling the top portion with bentonite and sand. Concrete bulkheads would seal the room entrances, and grouting material would be used to ensure integrity. Closure of the vault would be achieved by backfilling the access tunnels in the same manner as the rooms. The shafts leading to the access tunnels would be backfilled with compacted clay and crushed granite, separated by a series of supporting concrete bulkheads.

The disposal concept is being assessed using a probabilistic systems assessment code (SYVAC3), to characterize the long-term radiological safety of a hypothetical vault.[4] The component models that run under the SYVAC executive code are shown in Figure 3. The vault model has been developed to assess the performance of engineered

barriers, and includes container failure, release of radionuclides from used fuel, and mass transport of released radionuclides through the clay-based sealing materials surrounding the waste containers.[5]

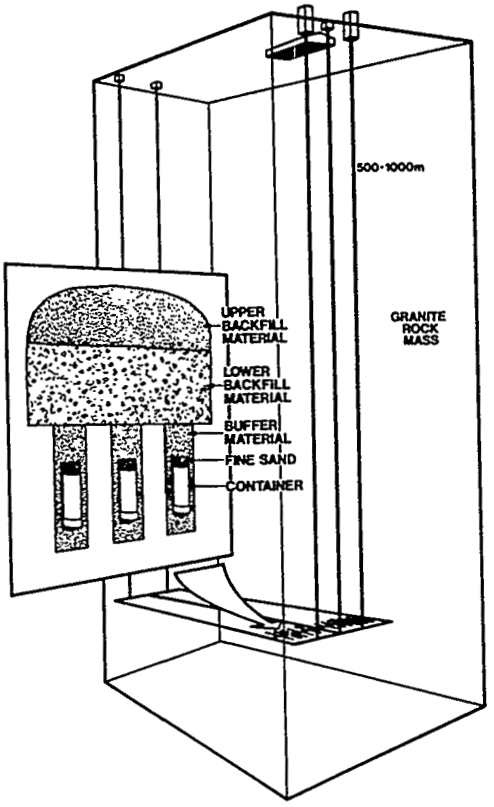

Figure 1 Conceptual disposal vault.

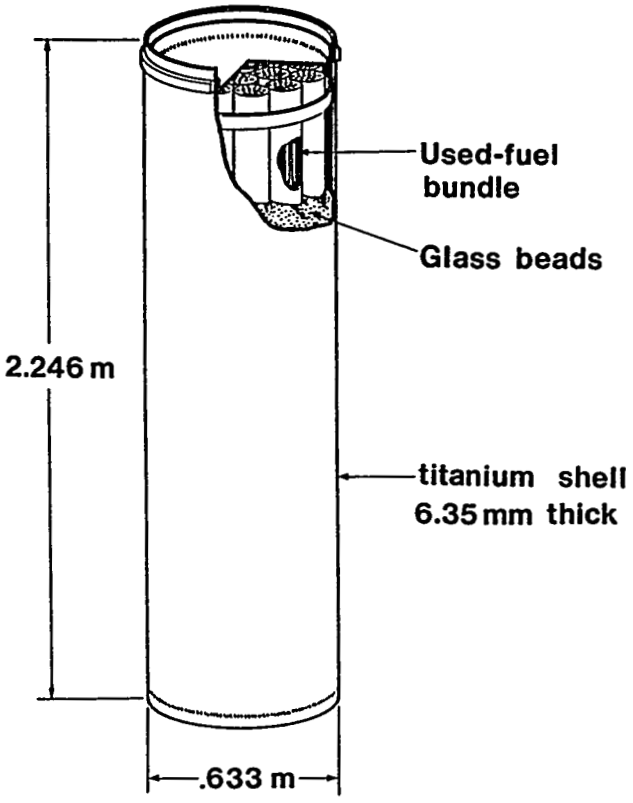

Figure 2 Fuel container.

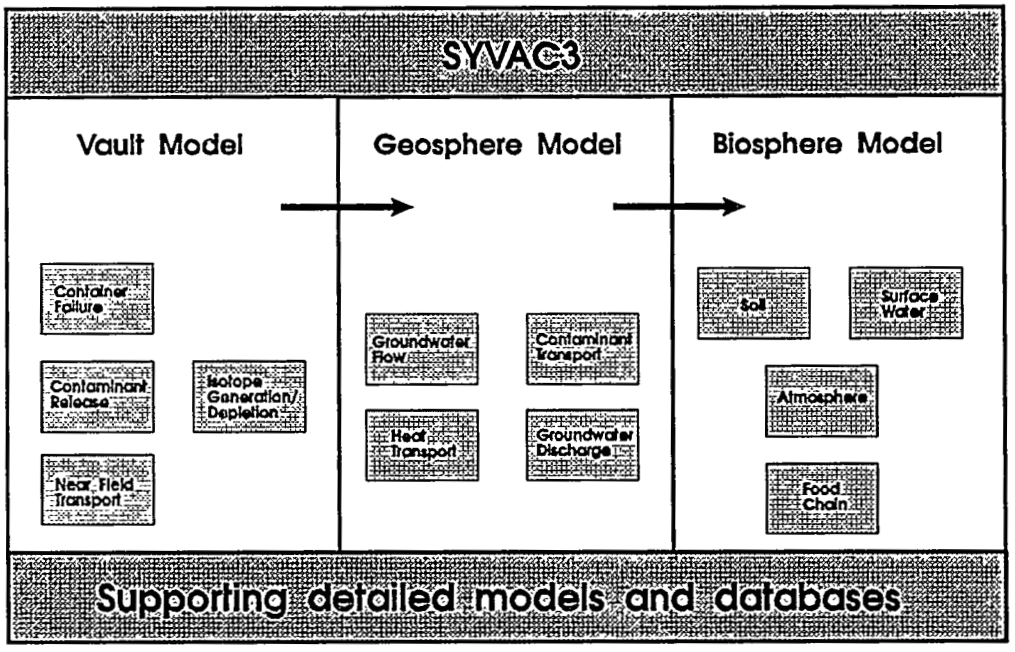

Figure 3 Models for disposal vault assessment.

The geosphere model is site specific, and includes sub-models for contaminant transport, groundwater flow, and heat transport. A SYVAC geosphere model of a hypothetical site (such as a research area) begins with the characterization of all relevant geological, geophysical, geochemical, and hydrogeological data for the region. Based on this information, a detailed conceptual model of the hydrogeological regime is formulated. Contaminant transport models are then used to determine a suitable form for the SYVAC geosphere model.[6]

The biosphere model describes the transport of contaminants throughout a watershed supplying a surface body of water. The model calculates the resulting concentrations of contaminants in lake, soil, and atmosphere environmental compartments, and the radiological consequences for humans dependent on resources from the affected area.[7-9]

SYVAC3 directs repeated simulations of the total waste disposal system, selecting values for the parameters in the three models from probability distribution functions, and predicts a dose-frequency histogram. This involves considerable amounts of computation time, and it is necessary to use relatively simple models of the various processes. Detailed models have been developed to describe the relevant phenomena, and the results of these analyses are then used to develop simplified assessment models, which produce results consistent with the detailed models over the ranges of conditions required for the assessment. These simplified models are then used in SYVAC.

In addition to the modeling work a comprehensive experimental program is being performed in which both the fundamental science and engineering scale phenomena are being addressed. In the following sections, we describe some of the work being done in these programs.

Engineered Barriers

Engineered barriers consist of the used fuel, the fuel container, and the buffer/backfill materials that surround the fuel container. The only credible mechanism for release and transport of contaminants from the vault is via groundwater flow. Therefore, we have been characterizing the performance of the engineered barriers by considering in detail the phenomena summarized in Figure 4.

Fuel Performance

Most of the radionuclides in spent fuel are trapped within the uranium oxide grains, and would only be released if the fuel were to dissolve in groundwater. In addition, fuel in contact with groundwater would release about 2% (on average) of the cesium and iodine when the fuel sheath fails, and about 6% (on average) of these elements would be slowly released due to preferential dissolution at the grain boundaries.[10] It is clear, therefore, that the stability of uranium dioxide in groundwater is extremely important.

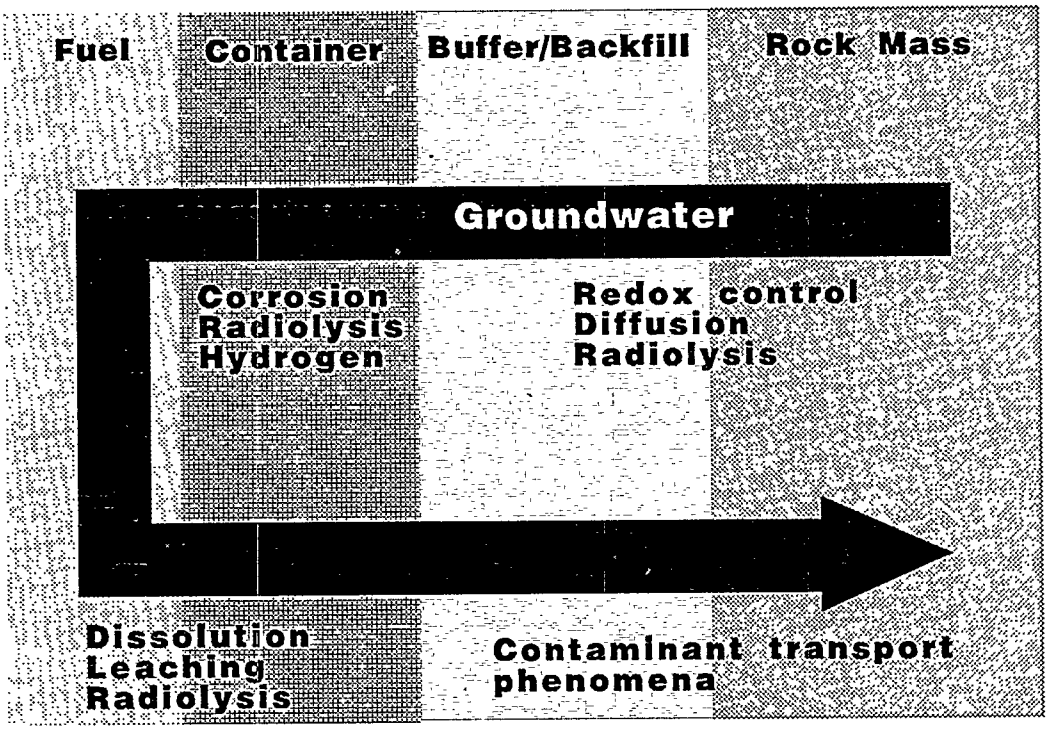

Figure 4 Groundwater interactions in the vault.

Our current understanding of the chemical behaviour of used fuel in groundwater is summarized in Figure 5.[11] The shaded area in the upper part of the figure indicates the oxidation potential range expected just after the vault is sealed. As the oxygen is consumed, the oxidation potential rapidly drops to a value somewhere in the range indicated by the shaded area in the bottom part of the figure. Investigations have shown that uranium oxide compositions below $UO_{2.33}$ have extremely low solubilities.[12] Therefore, the fuel is likely to be highly stable in the groundwater conditions expected in the vault. Figure 5 also indicates the effects of α-radiation on the composition, as determined from electrochemistry experiments.[13] Since the radiation fields when the fuel would come into contact with groundwater would be equivalent to those produced by the 5-10 μCi source in these experiments, it is unlikely that α-radiation would have a major effect on fuel dissolution. Finally, experiments with used fuel have shown that uranium concentrations in very dilute to moderately saline groundwaters are only about 5-30 μg/L, and the dissolution rates are found to be extremely low.[10]

Natural analogs can also provide useful information on the stability of uranium oxides in groundwater. We have been examining the Cigar Lake uranium deposit in northern Saskatchewan, which was formed about 1.3 billion years ago, and is situated at a depth of 430 m at the interface between the host sandstone formation and the underlying basement rock.[14] The ore body contains about 150 000 Mg of high-grade ore in the form of individual grains of uranium oxide mixed with clay minerals. The average concentration of the ore is 12%, with local concentrations reaching 60%. Investigations have

shown that the surface oxidation state of the uranium oxide grains is below $UO_{2.33}$, which is consistent with the redox conditions of the groundwater in contact with the ore body.[15] As a result, water Samples only 5 m away from the ore body contain uranium concentrations that are below the levels specified for drinking water.

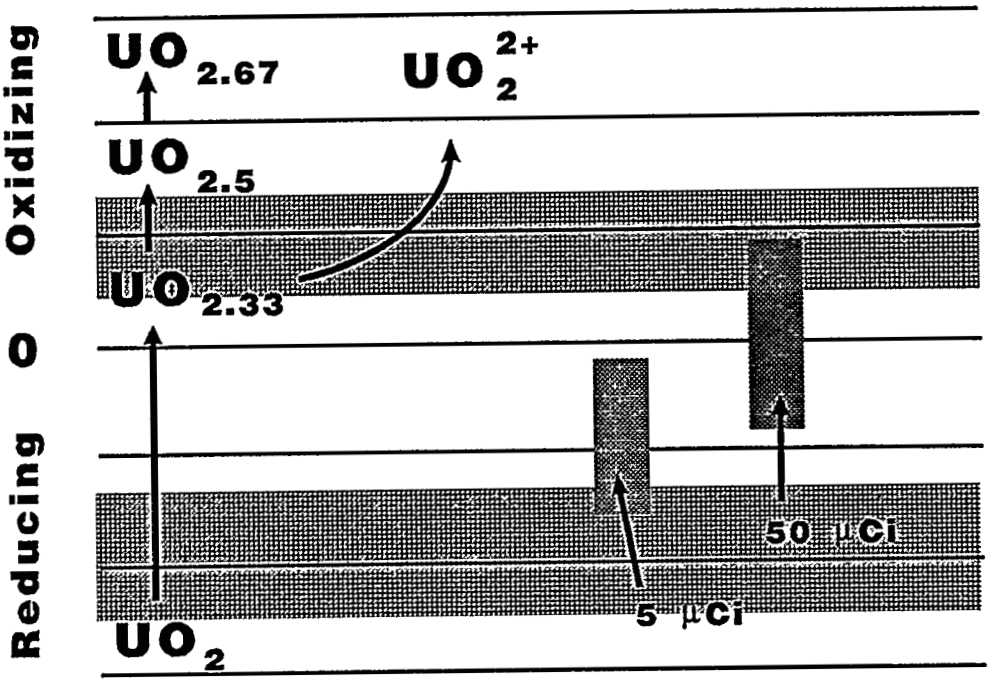

Figure 5 Effect of oxidation potential on fuel behaviour.

Containers

Titanium alloys and copper are being examined as container materials since they are found to be particularly resistant to the chloride-rich groundwaters found deep in the Canadian Shield.

There are three principal mechanisms by which the reference ASTM Grade 2 titanium containers could fail, and these mechanisms are being characterized in detail. The first mechanism, uniform, or general, corrosion, is found to proceed extremely slowly (< 1µm/a) under conditions expected in the disposal vault (groundwater temperature < 150°C and chloride content < 1 mol/L).[16] This extremely low corrosion rate is due to the formation of a passivating oxide layer on the surface of the titanium. Therefore, bulk corrosion is not likely to be a major failure process.

If the passivating layer breaks down, then localized effects, such as crevice corrosion, would occur. Electrochemical studies are showing that the propagation of crevices may be prevented by the re-establishment of the protective oxide film. In particular, it has been found that crevice corrosion does not occur at all for ASTM Grade 12 titanium at temperatures below 70°C.

The third failure mechanism is hydrogen-induced cracking which also requires breakdown of the oxide layer. There is no evidence that hydrogen-induced cracking occurs in titanium until temperatures are below 30°C. Therefore, this mechanism is only likely to occur at short times before temperatures have risen, or at relatively long times when the temperature has decreased.

Figure 6 illustrates a conservative model for the failure of thin-walled titanium-2 containers over long periods of time. For the failure mechanisms involving corrosion, it is assumed that mechanical failure would occur whenever the container wall thickness is reduced to 2 mm at any location. It is also assumed that 1 in 5000 containers would fail immediately due to manufacturing defects. During the first 1000 years, less than 1% of the containers would fail due to hydrogen cracking. The model assumes that this would occur for the Ti-2 containers located near the edges of the vault where temperatures would remain low. Over the next several thousand years, about 95% of the containers are assumed to fail by crevice corrosion. However, it is not clear that crevice corrosion would occur since the oxygen concentrations in the vault would be very low. In addition, very aggressive conditions have been necessary in the laboratory to initiate crevice corrosion in Ti. Finally, it is assumed that the remaining containers, whose crevice corrosion rates would be slow, would ultimately fail due to hydrogen-induced cracking. Therefore, even assuming a very conservative failure model, there is little doubt that the containers would isolate the used fuel from groundwater for at least 500 years.

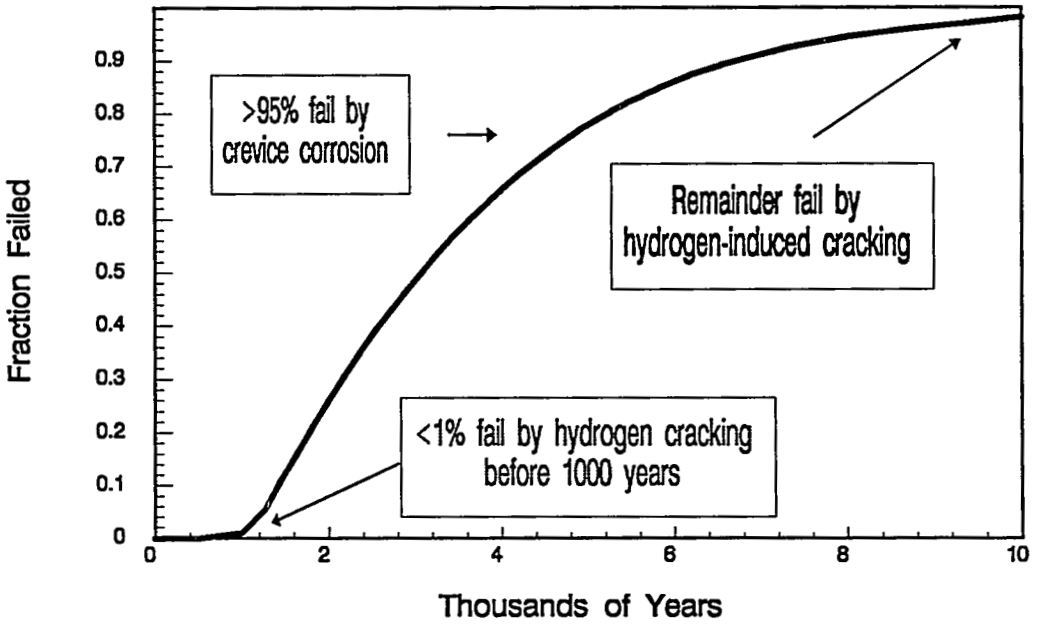

Figure 6 Conservative model for the failure of Ti-2 containers.

Backfilling Materials

The reference buffer material surrounding the fuel containers is a mixture of 50 wt% sodium bentonite clay and 50 wt% silica sand, which is found to be an optimum mixture for many physical properties such as strength, hydraulic and thermal conductivities, and drying shrinkage.[17] Bentonite has been selected for mixing with the sand owing to its high swelling capacity, its low permeability to groundwater, and its high sorption capacity for many radionuclides.[18] In addition, the effective diffusion coefficients for anions that do not interact strongly with clays, such as I^- and TcO_4^-, are very small.[19] Finally, field and laboratory studies of natural bentonite deposits in southern Saskatchewan have confirmed that bentonite has maintained an acceptably high swelling potential and low permeability millions of years after its deposition.[20]

The reference material for the main backfill is a mixture of 25 wt% Lake Aggassiz clay and 75 wt% crushed granite. This mixture has been found to have the maximum density and the minimum hydraulic conductivity after dynamic compaction.[21] It is also a highly effective barrier to radionuclide transport.

Geosphere

The migration of contaminants into the surrounding rock would be controlled by the flow rate of groundwater around the vault, the fracture density and porosity of the rock, and the diffusion of contaminants through the buffer. Since the movement of groundwater around the vault would be exceedingly slow, and the porosity and fracture density of the rock very small, the rate of transfer into the geosphere would be low. Once contaminants were in the surrounding rock, their rate of movement would be affected by sorption onto minerals and by dispersion and dilution in the groundwater system. These processes would tend to reduce the concentration of contaminants in the groundwater.

Application of Geoscience Methodology

The performance of any particular rock mass can be assessed by applying a structured methodology that integrates various geoscience disciplines.[22]

1. Field investigations are carried out to determine geological features of the rock mass that affect groundwater flow.

2. The data from these investigations are used to develop a conceptual model of the groundwater flow systems.

3. Based on the conceptual model, a detailed three-dimensional mathematical model of the flow system is used to predict changes caused by natural and man-made perturbations of the rock mass and flow system. The observed changes are then compared to the model predictions, and the models are refined so that together they provide a realistic representation of the actual groundwater flow system.

4. The validated mathematical model is then used to predict contaminant transport for various vault designs in the rock mass.

The above methodology can be illustrated by examining its application to the volume of rock containing the Underground Research Laboratory, which is described in the next section. Over 130 boreholes were drilled into the rock to depths up to 1100 m. Fractures were characterized in the boreholes using several techniques, including detailed core-logging methods, in-hole television camera equipment, and by a variety of standard and innovative borehole geophysical logging techniques. Hydraulic conductivity measurements were made in selected zones in individual boreholes. Also, the interaction between boreholes was measured by either injecting or withdrawing water from one borehole, while groundwater pressures were measured in isolated zones in neighboring boreholes. These data provided an understanding of the hydraulic conductivity of the rock mass located between the boreholes, and have allowed us to characterize the major fracture zones.[23]

The conceptual groundwater flow model resulting from these measurements was then described using the finite-element, three-dimensional MOTIF code.[24,25] MOTIF represents the relatively intact rock by an equivalent porous medium composed of three-dimensional continuum elements. The high conductivity fracture zones are described by special planar elements that are embedded in the intact rock background. The three-dimensional flow field for this system is described by porous medium flow equations.

As expected, it was found that the shortest groundwater transit times to the surface occurred in the fracture zones. If any contaminants would reach a fracture zone where the direction of the groundwater flow were towards the surface, the transit time could be of the order of a thousand years. However, the hydraulic conductivity of the relatively intact rock was found to be so small that radionuclide migration would occur only by diffusion. This means that contaminant transport times of the order of 1000 years/m would be expected. Therefore, in an actual vault, the location of the disposal rooms relative to any fracture zones would be a major parameter with respect to contaminant transport to the surface.

Underground Research Laboratory

The Underground Research Laboratory (URL) is located in the Lac du Bonnet Batholith, a previously undisturbed granitic pluton located near the Whiteshell Laboratories. The URL site was thoroughly characterized before and during construction to ensure that any changes in the hydrogeology of the region are well-documented. The laboratory, which is close to completion, consists of a 443 m deep shaft with shaft stations and working levels at various depths. The final configuration for the laboratory is shown in Figure 7.

Research programs in the URL have been designed to provide realistic environment and boundary conditions for understanding integrated processes, to examine scaling effects, to conduct long term experiments on a scale that is impracticable in the laboratory, and to

provide a database for model development and validation. The research will be carried out over the next 10 years, and consists of the following major experiments:

1) Solute transport in fractured rock.

2) Buffer/container experiments.

3) Grouting and shaft sealing experiments.

4) Multicomponent tests using a backfilled room.

5) Mine-by experiment to assess the effect of excavations.

6) URL site characterization.

These experiments will allow us to develop and test the methodology that will be needed to construct a waste vault.

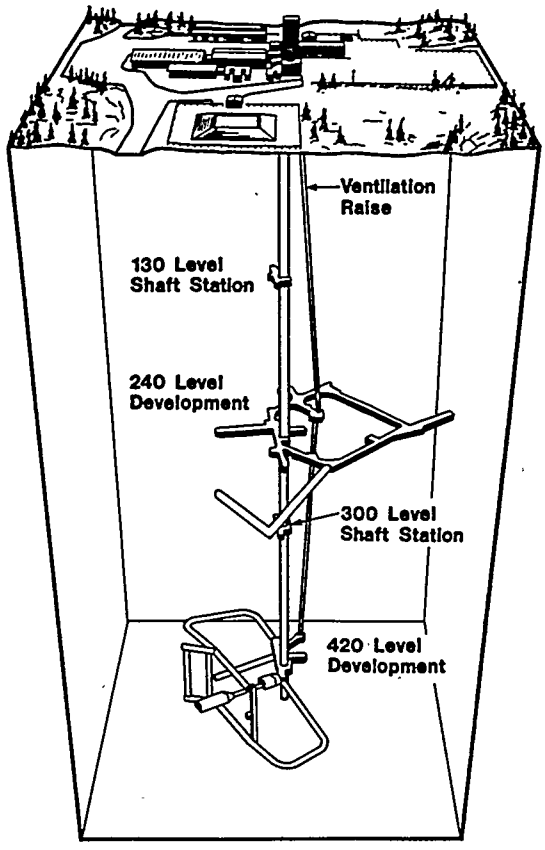

Figure 7 Underground Research Laboratory.

Biosphere

A comprehensive research program is being carried out in environmental research to improve our understanding of important processes, and to use this information in the biosphere model.[26] The research includes contaminant behavior in soils, wetlands, surface waters, plants, the atmosphere, and food chains. A brief description of some of this research is given below.

The transport of contaminants in soil is an important pathway to humans. Over the past several years, soil cores have been spiked with various radioactive tracers and placed in small lysimeters to observe the manner in which the tracers have been redistributed under the action of leaching and capillary rise.[27] The tracers have been added to the surface of some core samples to simulate an irrigation or deposition source, and to the base of other cores to simulate contact with contaminated groundwater. The groundwater cores showed that Tc, I, Mo, and Cr are the most mobile contaminants. A detailed soil solute transport model has been developed that agrees well with experimental data.[28]

The transfer of contaminants from soil to plants is being studied in the field using natural trace elements, and in the laboratory using radioactive tracers.[29,30] Contaminant concentrations in the soil and plants are measured and used to calculate concentration ratios for use in the food-chain submodel of the biosphere model.

Contaminant transport in food chains is being investigated using natural radioactive potassium and fallout cesium as tracers, for two food chains that culminate in beaver and in moose. The data are used to determine transfer factors for the food-chain submodel, and to identify uncertainties due to species variability.[31] In addition, the contribution of inhalation of contaminants by food animals to human doses has been reviewed for 57 radionuclides.[32]

Other work on the biosphere includes the volatilization of contaminants by plants, dispersion of gases released from underground sources, and iodine tracer studies in wetlands. All of this work is leading to an improved generic understanding of the transport of radionuclides through the biosphere.

Conclusions

The Canadian program for the permanent disposal of nuclear fuel wastes is focussing on a conceptual waste vault located in the Canadian Shield. Before site selection can begin, it must be demonstrated that the conceptual waste vault can meet the regulatory requirements, and the concept must undergo extensive public and scientific review. A comprehensive R&D program has been carried out over the past 12 years to develop the required understanding and technology. This research has involved detailed examinations of engineered barriers, the geosphere, and the biosphere. In each case, fundamental research is being done to elucidate underlying processes, large scale integrated tests are being done to understand scaling effects and the interactions between the various phenomena, and models

are being developed that can be applied to site characterization. The results of this research are giving us confidence that it will be possible to identify many locations in the Canadian Shield that would be technically suitable for the permanent disposal of nuclear fuel wastes.

Acknowledgements

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References

1. Aiken, A.M., Harrison, J.M. and Hare, F.K., "The Management of Canada's Nuclear Wastes", Energy, Mines and Resources Canada Report EP-77-6, 1977.

2. Baumgartner, P. and Simmons, G.R., "Disposal Centre Engineering for the Canadian Nuclear Fuel Waste Management Program," Radioactive Waste Management and the Nuclear Fuel Cycle, Vol 8, Numbers 2-3, p 219, (1987).

3. Teper, B., "Feasibility Assessment of the Particulate-Packed, Thin-Walled Container", AECL Research Technical Record TR-350, Vol 1, p 21, 1985.

4. Dormuth, K.W. and Sherman, G.R., "SYVAC - A Computer Program for Assessment of Nuclear Fuel Waste Management Systems, Incorporating Parameter Variability", AECL Research Report, AECL-6814, 1981.

5. Garisto, N.C. and LeNeveu, D.M., "The Vault Model for the Disposal of Used CANDU Fuel: Documentation and Analysis of Scoping Calculations", AECL Research Report, AECL-9578, 1989.

6. Chan, T., "An Overview of Groundwater Flow and Radionuclide Transport Modeling in the Canadian Nuclear Fuel Waste Management Program", Proc of Conf on Geostatistical, Sensitivity, and Uncertainty Methods for Ground-Water Flow and Radionuclide Transport Modeling, San Francisco, p 39, 1987.

7. Zach, R., "LIMCAL: A Comprehensive Food Chain Model for Predicting Radiation Exposure to Man in Long-term Nuclear Waste Management, AECL Research Report AECL-6827, 1982.

8. Sheppard, M.I., Dzik, E.J., and Mehta, K.K., "SCEMR1 Soil Model for SYVAC", AECL Research Technical Record TR-363, 1985.

9. Fleer, V.N. and Barnard, J.W., "The Biosphere Submodel for SYVAC3", AECL Research Technical Record TR-375, Vol 2, p 353, 1986.

10. Johnson, L.H., Shoesmith, D.W., and Stroes-gascoyne, S., "Spent Fuel: Characterization Studies and Dissolution Behaviour under

Disposal Conditions", Scientific Basis for Nuclear Waste Management, Vol. XI, Pittsburgh, p 99, (1988).

11. Shoesmith, D.W., private communication, 1990.

12. Shoesmith, D.W., Sunder, S., Bailey, M.G., Wallace, G.J. and Stanchell, F.W., "Anodic oxidation of UO_2. IV. X-ray photoelectron spectroscopic and electrochemical studies of film-growth in carbonate-containing solutions", Appl. Surf. Sci., Vol 20, 39 (1984).

13. Bailey, M.G., Johnson, L.H., and Shoesmith, D.W., "The effects of the alpha-radiolysis of water on the corrosion of UO_2", Corros. Sci., Vol 25, p 233, (1985). Also, D.W. Shoesmith, private communication.

14. Cramer, J.J., "A Natural Analog for a Fuel Waste Disposal Vault", Proc. 2nd International Conference on Radioactive Waste Management, Canadian Nuclear Society, Winnipeg, p 697, 1986.

15. Sunder, S., Taylor, P., and Cramer, J.J., "XPS and XRD studies of uranium rich minerals from Cigar Lake, Saskatchewan", Scientific Basis for Nuclear Waste Management, Vol XI, Pittsburgh, p 465, (1988).

16. Ikeda, B.M. and Clarke, C.F., "Titanium Corrosion under Canadian Nuclear Waste Disposal Conditions", Proc 2nd International Conference on Radioactive Waste Management, Canadian Nuclear Society, Winnipeg, p 593, 1986.

17. Dixon, D.A. and Gray, M.N. "The Engineering Properties of Buffer Material - Research at Whiteshell Nuclear Research Establishment", AECL Research Technical Record TR-350, p 513, 1985.

18. Oscarson, D.W. and Cheung, S.C.H., "Evaluation of Phyllosilicates as a Buffer Component in the Disposal of Nuclear Fuel Waste", AECL Research Report AECL-7812 (1983).

19. Cheung, S.C.H., "Evaluation of Methodologies for Determining Ionic Diffusion Coefficients in Clays", Proc. of the Canadian Engineering Centennial Convention, Montreal, p 195, 1987.

20. Oscarson, D.W., Dixon, D.A. and Gray, M.N., "Swelling capacity and permeability of an unprocessed and processed bentonite clay", J of Eng Geology, Vol 28, p 281 (1990).

21. Dixon, D.A., Gray, M.N., and Thomas, A.W., "A study of the compaction properties of potential clay-sand buffer mixtures for use in nuclear fuel waste disposal", Eng. Geol. 21, p 247 (1985).

22. Whitaker, S.H., "Geoscience research for the Canadian Nuclear Fuel Waste Management Program", Radioactive Waste Management and the Nuclear Fuel Cycle, Vol 8, Numbers 2-3, p 145 (1987).

23. Davison, C.C., "Hydrogeological Characterization of the URL Site", AECL Research Technical Record, TR-299, p 231, 1985.

24. Davison, C.C., Chan, T., and Scheier, N.W. "Experimental Activities of the Canadian Nuclear Fuel Waste Management Program to Validate Geosphere Models", Proc of Geoval-87, Stockholm, 1987.

25. Guvanasen, V., "Development of a Finite-Element Code and Its Application to Geoscience Research", AECL Research Technical Record TR-299, p 554, 1985.

26. Zach, R., Amiro, B.D., Champ, D.R., Cornett, R.J., Davis, P.A., Killey, R.W.D., Lee, D.R., Moltyaner, G.L., Osborne, R.V., Sheppard, M.I., and Sheppard, S.C., "Environmental research for the Canadian Nuclear Fuel Waste Management Program", Radioactive Waste Management and the Nuclear Fuel Cycle, Vol 8, p 197 (1987).

27. Sheppard, M.I., Thibault, D.H., and Mitchell, J.H., "Element leaching and capillary rise in sandy soil cores: experimental results", J. of Env. Quality, Vol 16, No 3, p 273 (1987).

28. Sheppard, M.I. and Sheppard, S.C., "A soil solute transport model evaluated on two experimental systems", Eco. Modelling, Vol 37, p 191 (1987).

29. Sheppard, S.C., Evenden, W.G., and Pollock, R.J., "Uptake of natural radionuclides by field and garden crops", Can J. Soil Sci., Vol 69, p 751 (1989).

30. Sheppard, S.C. and Evenden, W.G., " Critical compilation and review of plant/soil concentration rations for uranium, thorium and lead", J. Environ. Radioactivity, Vol 8, p 255 (1988).

31. Zach, R., Hawkins, J.L., and Mayoh, K.R., "Transfer of fallout cesium-137 and natural potassium-40 in a boreal environment", J. Environ. Radioactivity, Vol 10, p 19 (1989).

32. Zach, R., "Contribution of inhalation by food animals to man's ingestion dose", Health Physics, Vol 49, No 5, p 737 (1985).

DISCUSSION

CEDERQVIST: As you probably know, the plans in Sweden are to encapsulate the fuel in copper. Could you please elaborate on why you have chosen titanium? I guess that might have to do with water supply, groundwater, or something?

TORGERSON: We have two reference materials, titanium and copper. Our groundwater is such that we find some advantages in using titanium. I believe in the case of Sweden you have more saline groundwaters. The chemistry is such that going to very thick cooper has advantages for you. There are several engineering reasons why titanium turns out to be better for our program. Basically, it comes down to compatibility with the types of groundwater that one gets in the Canadian shield.

FIRST: I noticed, Dr. Torgerson, that you mentioned toward the end of your talk that there had been some cooperation in your activities from DOE. Would you kindly indicate what the cooperative arrangements are between the Canadian program and the U.S. program. Are they going their separate ways, or is there close coordination being exercised?

TORGERSON: I joined the Waste Management Program back in December. What I have found since then is that there is a lot of cooperation internationally. With respect to the US DOE, we have had a cooperative effort on the underground research lab. You probably recall that at one point there were two media being considered in the United States. One was granitic-type crystalline rock, the approach that we are taking. The other was TVF, the rock formation that the DOE is currently proposing. At an earlier time, therefore, there was a lot more cooperation than there is currently. However, there are still some areas where generic cooperation may be beneficial. Therefore, we have had ongoing discussions with the US DOE to work out our future interactions.

PLUMB: I am always reminded that perfection is the enemy of various things including the practicum. We already have a little dissension occurring in the canister material, question of whether it should be copper or titanium. Of course, the next obvious question is, "Why not zircaloy, the original choice for LWR containment?" Just a little bit more about this question of ultimate proof which gets very complicated. What are practical options and limitations?

TORGERSON: Again, I will have to point out that there are probably many container materials that are appropriate. I don't think there is any one "best" container material; however, container materials must be compatible with the conditions that you have in your particular geologic medium. So, when people tell me that one container is better than another container, I don't buy that unless they can tell me all the conditions under which they are going to be disposing of the fuel. Clearly, there are some materials you don't want to use such as those that generate hydrogen. You have to pay careful attention to materials problems in a waste vault, and I think that the international waste management RHO community is doing so.

Radiological Health Effects Models for Nuclear Power Plant Accident Consequence Analysis -- An Update (1990)

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Harvard School of Public Health

Jeremy L. Sprung
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Abstract

In 1975 the U.S. Nuclear Regulatory Commission (NRC) issued the Reactor Safety Study (RSS). The health effects models developed for the RSS provided the basis for most of the NRC's early assessments of the health risks associated with nuclear power plant accidents.

Ten years later the NRC published Health Effects Models for Nuclear Power Plant Accident Consequence Analysis. The new radiological health effects models differed from the original RSS models in several important ways: (i) Weibull functions were used to assess the risks of early health effects, (ii) relative risk projection was recommended for estimating the risks of several cancers -- including breast, gastrointestinal and lung, (iii) data were included that enabled the analyst to project both incidence and mortality from radiation-induced cancers, (iii) genetic risks were estimated using linear-quadratic models of dose-response, and (iv) uncertainty was addressed by providing central, lower and upper estimates of the risks of most effects.

Recently, the NRC completed an effort to further revise their health effects models. The revised models build upon those developed in 1985, but include several significant modifications: (i) threshold doses have been estimated for the early health effects models, (ii) dose rate dependent LD_{50}'s have been developed for the bone marrow and pulmonary syndromes, (iii) upper estimates of the risks of several cancers -- including breast, gastrointestinal, and lung -- are based on the assumption that relative risk depends on age at exposure, and (iv) models for estimating the peri-implantation pregnancy losses ("spontaneous" abortions early in pregnancy) due to radiation-induced genetic damage have been developed. Here we briefly summarize NRC's revised health effects models.

Introduction

In 1975 the U.S. Nuclear Regulatory Commission (NRC) issued the Reactor Safety Study (RSS). The health effects models developed for the RSS provided the basis for most of the NRC's early assessments of the health risks associated with nuclear power plant accidents.

Ten years later the NRC published Health Effects Models for Nuclear Power Plant Accident Consequence Analysis. This document described new radiological health effects models that differed from the original RSS health effects models in several ways:

- Weibull functions were used to assess the risks of early health effects,
- Models were added to permit analysis of the risks of skin burns, sterility and mental retardation,
- Relative risk projection was recommended as the basis for estimating the risks of several cancers -- including breast, gastrointestinal and lung,
- More recent data from the followup of the survivors of the bombings of Hiroshima and Nagasaki were considered in developing the parameters of the cancer risk models,
- Data were included that enabled the analyst to project both incidence and mortality from radiation-induced cancers,
- Uncertainty was addressed by providing central, lower and upper estimates of the risks of most effects, and
- Information about the severity, duration and impact on longevity of each radiological health effect was given for each risk assessment model.

Recently, the NRC completed an effort to further revise their health effects models. A revised edition of the document Health Effects Models for Nuclear Power Plant Accident Consequence Analysis has just been published (NRC, 1990). It builds on the 1985 models, but includes several significant modifications:

- Threshold doses have been estimated for the early health effects models,
- Dose rate dependent LD_{50}'s have been developed for the bone marrow and pulmonary syndromes,
- Upper estimates of the risks of several cancers -- including breast, gastrointestinal, and lung -- are based on the assumption that relative risk depends on age at exposure, and
- Models for estimating the peri-implantation pregnancy losses ("spontaneous" abortions early in pregnancy) due to radiation-induced genetic damage have been developed.

Here we attempt to briefly summarize NRC's revised health effects models. Readers interested in the scientific bases for the models should consult the complete report -- Health Effects Models for Nuclear Power Plant Accident Consequence Analysis, Revision 1 (NRC,1990).

Model Description

The NRC's radiological health effects model is actually a collection of models. The collection includes three broad classes of effects -- early effects, late somatic effects and genetic effects.

The models were developed largely by four working groups and were reviewed both by the members of our scientific advisory committee and by several scientists who had not participated in the NRC model development project.

The models are intended to permit estimation of the risks associated with exposure to low-LET radiation. The NRC is currently developing additional models for exposure to high-LET radiation.

Uncertainty

The health risks associated with exposure to ionizing radiation cannot be predicted precisely. The uncertainties are of two types -- parameter uncertainties and model uncertainties. Parameter uncertainty arises due to the need to draw inferences from small samples about processes which are to some extent random (or are observed with error). Standard statistical approaches permit one to estimate the magnitude of parameter uncertainties. Model uncertainty arises from the need to rely on analogy. Estimates of the magnitude of model uncertainty are necessarily subjective -- they are related to the validity of the analogy.

The working groups have taken a first step toward addressing uncertainty by providing three risk estimates for each health effect -- a central estimate, a lower estimate and an upper estimate. The central estimates are intended to be realistic -- reflecting the judgment of the scientists involved in model development. The upper and lower estimates are intended to reflect alternative views that are reasonably consistent with available evidence and that may be preferred by some scientists.

In view of the rather large uncertainties in estimating radiological health risks it is important that accident consequence analysts consider the implications of the full spectrum of possible consequences rather than focusing attention on the central estimates.

Early Effects

In the event of a severe nuclear power plant accident, those living nearby may receive doses large enough to suffer from the "early and continuing" effects of radiation. The early effects -- which include the potentially lethal hematopoietic, pulmonary and gastrointestinal syndromes and several less severe effects such as vomiting, diarrhea and skin burns -- typically occur within the first few days or weeks after exposure. Continuing effects -- such as hypothyroidism, pneumonitis, diminution of sperm count/supression of ovulation, and cataracts -- may require somewhat longer to develop or may involve symptoms that persist for several years after exposure. Irradiation of women pregnant at the time of the accident may also lead to increased risk of embryo loss, fetal death or mental retardation among those babies that survive the initial exposure.

Knowledge of the risks of these effects derives largely from four sources -- studies of radiation related side effects among humans exposed therapeutically, analyses of the experience of the survivors of the atomic bombings of Hiroshima and Nagasaki, examination of the health effects observed among the relatively small numbers of individuals who have received large doses in various accidents, and investigations of the effects observed in animals experimentally exposed to radiation.

The NRC models for early and continuing effects were developed by Drs. Bobby Scott and Fletcher Hahn of the Lovelace Inhalation Toxicology Research Institute (ITRI). The models are based in part on data concerning human radiation injury which was reviewed and interpreted by Drs. Neil Wald, Joseph Watson and Albert Spritzer of the University of Pittsburgh. Information on thyroid effects was provided by Dr. Harry Maxon of the University of Cincinnati and several of his colleagues.

Scientific understanding of the biological nature of early effects indicates that they are threshold effects -- i.e., in any individual the effect will not be experienced unless a threshold dose is exceeded. Population dose-response functions for these non-stochastic effects are simply reflections of the distributions of individual thresholds (tolerances) among the population.

The risks of early and continuing effects are modeled using hazard functions. The relationship between risk and hazard is given by:

$$r = 1 - e^{-H}$$

where r is the probability that a person will exhibit the effect of interest and H is the cumulative hazard function. The relationship between dose and risk is implicit in the relationship between dose and hazard. The cumulative hazard functions used to predict early effects are two-parameter Weibull functions of the form:

$$H = 0.693 \left[d / D_{50} \right]^{v} \qquad \text{for } d > T$$

where d is the (average absorbed) dose to the relevant organ, D_{50} is the dose at which half of the exposed population experiences the effect, v is the shape parameter and T is the population threshold dose. All doses are given in Gray (Gy).

Table I gives the values of LD_{50}, T and v recommended by the early effects working group for analyzing the risk of death from the hematopoietic, pulmonary and gastrointestinal syndromes following brief exposure at high dose rate. Two sets of values are given for the hematopoietic syndrome -- one appropriate for those receiving supportive treatment and another for those receiving only minimal medical treatment. Supportive treatment includes reverse isolation, antiobiotic therapy, blood transfusions, electrolyte replacement and parenteral feeding.

For most early effects, dose received at low dose rate is much less effective than dose received at high dose rate. This phenomenon can be accounted for by adjusting the value of the median lethal (or effective) dose. The simplest adjustment is one in which two values of D_{50} are used -- one appropriate for dose received at high dose rate, and another for dose received at low dose rate. Using this approach, the cumulative hazard is evaluated as:

$$H = 0.693 \left[\left(d_b / D_{50,b} \right) + \left(d_p / D_{50,p} \right) \right]^{v}$$

where d_b is the brief dose (received at high dose rate) and d_p is the protracted dose (received at low dose rate).

This is the approach recommended by the early effects working group for estimating the risks of early morbidity. The parameters suggested for analyzing the risks of prodromal symptoms (vomiting, diahrrea), thyroiditis, hypothyroidism, skin burns (erythema, transepidermal injury), reproductive effects (permanent supression of ovulation, temporary diminution of sperm count), and cataracts are given in Tables II and III. As noted in the tables, a dose rate cutoff of 0.05 Gy per hour has been recommended to distinguish "high" and "low" dose rates.

Table I. Early Mortality from Brief Exposure[a,b]

Effect	Risk Estimate								
	Central			Lower[c]			Upper[c]		
	LD_{50}	T^d	v	LD_{50}	T^d	v	LD_{50}	T^d	v
Hematopoietic Syndrome									
Minimal Treatment	3	1.5	6	3.5	2	8	2.5	1	4
Supportive Treatment	4.5	2	6	5	3	8	4	1.5	4
Pulmonary[e] Syndrome	10	5	12	12	6	14	8	4	9
Gastrointestinal Syndrome	15	8	10	20	8	10	10	8	10

a - Doses are organ-specific absorbed dose (Gy). Some values have been rounded to avoid conveying a false sense of precision.

b - Brief exposure parameters are appropriate for dose received at high dose rate. Modifications needed for application to low dose rate are described in the text.

c - The larger values of LD_{50} and T lead to lower estimates of risk.

d - Available human data are too weak to support clear choices of population threshold doses. One reviewer recommended setting the thresholds at 10 to 20% of the corresponding LD_{50}'s.

e - These parameters are thought to be appropriate for healthy young adults. Older persons and individuals with respiratory disease may be twice as sensitive.

Table II. Early Morbidity from Brief Exposure[a,b]

Effect	Risk Estimate								
	Central			Lower			Upper		
	ED_{50}	T	v	ED_{50}	T	v	ED_{50}	T	v
Vomiting	2	0.5	3	2.5	0.5	3	1.5	0.5	3
Diarrhea	3	1	2.5	4	1	2.5	2.5	1	2.5
Hypothyroidism[c]	60	2	-	60	2	-	60	2	-
Erythema	6	3	5	7	4	6	5	2	4
Transepidermal Injury	20	10	5	25	12	6	15	8	4
Supression of Ovulation	3.5	0.6	3	4.5	1	4	2.5	0.2	2
Supression of Sperm Count	0.7	0.3	10	0.8	0.4	11	0.6	0.2	9
Cataracts	3	1	2	7	1.5	3	2	0.5	1

a - Doses are organ-specific absorbed dose (Gy). Some values have been rounded to avoid conveying a false sense of precision.

b - Brief exposure parameters are appropriate for dose received at high dose rate -- i.e., > 0.05 Gy/hour.

c - The thyroid effects working group recommends estimating hypothyroidism risks following x-ray exposure using a linear model with a slope of 85×10^{-4} cases per Gy and a threshold of 2 Gy. One reviewer indicated that there was little empirical evidence supporting the threshold.

Table III. Early Morbidity from Protracted Exposure[a,b]

Effect	Risk Estimate								
	Central			Lower			Upper		
	ED_{50}	T	v	ED_{50}	T	v	ED_{50}	T	v
Vomiting	5	1.5	3	6	1.5	3	4	1.5	3
Diarrhea	6	2.5	2.5	7.5	2.5	2.5	5	2.5	2.5
Thyroiditis	1200	200	2	1200	200	2	1200	200	2
Hypothyroidism[c]	300	10	-	300	10	-	300	10	-
Erythema	20	6	5	30	8	6	10	4	4
Transepidermal Injury	80	40	5	100	50	6	60	30	4

a - Doses are organ-specific absorbed dose (Gy). Some values have been rounded to avoid conveying a false sense of precision.

b - Protracted exposure parameters are appropriate for dose received at low dose rate -- i.e., < 0.05 Gy/hour.

c - The thyroid effects working group recommends estimating hypothyroidism risks following ^{131}I exposure using a linear model with a slope of 17×10^{-4} cases per Gy and a threshold of 10 Gy. One reviewer indicated that there was little empirical support for the threshold.

Although simple, this approach may yield relatively imprecise estimates of risk -- especially when the median lethal dose is a strong function of the dose rate. Better estimates can be obtained by increasing the number of terms in the model. In the limit, a continuous form of the model is reached:

$$H = 0.693 \left[\int \dot{D} / D_{50}(\dot{D})\ dt \right]^{V}$$

where $\dot{D}$ is the instantaneous dose rate at time t, $D_{50}(\dot{D})$ is the median lethal dose applicable to dose received at dose rate $\dot{D}$ and H is the cumulative hazard function. This is the approach recommended by the early effects working group for computing the risks of death from the hematopoietic and pulmonary syndromes.

The relationship between median lethal dose and dose rate is given by:

$$D_{50}(\dot{D}) = \theta_{\infty} + \theta_1/\dot{D}$$

where θ_{∞} is the limiting value of the median lethal dose, $\dot{D}$ is the dose rate (Gy/hr) and θ_1 is a parameter reflecting the sensitivity of the median lethal dose to the dose rate (Gy^2/hr).

The recommended values of θ_{∞} are simply the LD_{50} values that were given in Table I. For the hematopoietic syndrome, the recommended values of θ_1 are approximately 2% of the corresponding θ_{∞}. For the pulmonary syndrome, the working group recommends that θ_1 be set at 2 to 4 times θ_{∞}. Using these values, the LD_{50} for hematopoietic syndrome mortality is essentially independent of dose rate for values of $\dot{D}$ above 1 Gy/hour. Similarly, the LD_{50} for pulmonary syndrome is essentially constant for dose rates above 100 Gy/hour. However as dose rates drop below these levels estimates of risk will rapidly diminish. For example, at a dose rate of 0.1 Gy/hour the LD_{50} for bone marrow syndrome mortality is approximately three times its limiting value and the LD_{50} for pulmonary syndrome mortality is between 20 and 40 times its limiting value.

Two effects of special concern are mental retardation and loss of an embryo or fetus. For both of these effects risk is a strong function of developmental age at the time of exposure. Table IV gives the parameters recommended for estimating the risks of these effects.

Late Somatic Effects

Those who survive the early effects of radiation or who receive doses to small to induce early effects may be at an increased risk of developing one of several forms of cancer.

Estimates of cancer risks are based primarily on the findings of studies of human populations exposed to ionizing radiation. Examples of such populations include -- the survivors of the atomic bombings of Hisoshima and Nagasaki, women treated with x-rays for postpartum mastitis, children treated by x-ray for

Table IV. Early and Continuing Effects from Brief Exposure in Utero[a]

Effect	Risk Estimate								
	Central			Lower			Upper		
	D_{50}	T	v	D_{50}	T	v	D_{50}	T	v
Severe Mental[b] Retardation									
8-15 Weeks	1.5	0	1	1	0.1	2	1	0	1
16-25 Weeks	7	0	1	2	0.5	2	3	0	1
Death of Embryo or Fetus									
0-18 Days	1	0.1	2	1.5	0.5	2.5	0.5	0	1.5
18-150 Days	1.5	0.4	3	2	0.5	4	1	0.2	2
> 150 Days[c]	-	-	-	-	-	-	-	-	-

a - Dose is the absorbed dose (Gy) received by the embryo or fetus. Some values have been rounded to avoid conveying a false sense of precision.

b - The D_{50} values for central and upper risk estimates correspond approximately to the doses at which the linear models recommended by the working group reach risks of 50%. Although the working group recommended that linear models also be used to derive lower estimates of risk, for these models we have included thresholds and have selected parameters consistent with quadratic fits of the data.

c - In this period the fetus and its mother are assumed to have about the same radiosensitivity. Parameter values should be selected from Table I or determined using the dose rate dependent models.

ringworm of the scalp, patients treated for ankylosing spondylitis, women given fluoroscopic examinations of the chest, persons treated with ^{131}I for Graves' disease and other thyroid conditions, and children born to women who received x-ray pelvimetry during pregnancy.

Many of these populations were exposed to relatively high doses at high dose rates. Few of the studies are complete -- i.e., many of those exposed are still alive. Thus, two key issues in the interpretation of these studies are how to extrapolate the results for use in situations involving much lower doses (and dose rates) and how to estimate the impact of incomplete followup.

The NRC models for late somatic effects were developed largely by Dr. Ethel Gilbert of Battelle Pacific Northwest Laboratories with input from Dr. Roy Shore - New York University, Dr. George Hutchison - Harvard School of Public Health, Drs. Gilbert Beebe and Charles Land - National Cancer Institute, Dr. Edward Webster - Massachusetts General Hospital, Dr. Jacob Fabrikant - University of California and Lawrence Berkeley Laboratory, Dr. William Bair - Battelle Pacific Northwest Laboratories, Dr. Marvin Goldman - University of California (Davis), Dr. Warren Sinclair - National Council on Radiation Protection and Measurements, Dr. Edward Radford - Radiation Effects Research Foundation, and Dr. Nan Laird - Harvard School of Public Health. The thyroid effects working group directed by Dr. Harry Maxon of the University of Cincinnati developed the models for thyroid cancers and benign thyroid nodules.

To derive central risk estimates for most cancers, the late somatic effects working group recommends use of a linear-quadratic model of dose-response in which the parameters are chosen so that doses received at low dose rates are 30 percent as effective as equivalent doses received at high rates. This central estimate of the low dose rate effectiveness factor was chosen from a range of values -- 10 to 50 percent -- given in NCRP Report 64 (NCRP, 1980). Many European accident consequence calculation codes rely on a low dose rate effectiveness factor of 45 percent. To reflect the uncertainty in this choice, the working group's lower risk estimates for most cancers are based on an effectiveness factor of 10 percent and their upper estimates assume that doses received at low dose rates are as effective as doses received at high rates.

Computationally, the effects of dose and dose rate are accounted for by using dose-response models of the form:

$$r = (a + bd)\ cd$$

where d is the dose (Gy), c is the unit risk coefficient (cases of cancer or cancer deaths per 1000 persons per Gy) derived from epidemiological studies at high dose and dose rate, a is the low dose rate effectiveness factor described above (0.3, 0.1 or 1 for central, lower and upper estimates), and b is a parameter selected to ensure that the modifying term, a + bd, equals 1 at a dose of 1.5 Gy.

When doses are received at low dose rates, the bd component of the modifying term is dropped and the model becomes:

$$r = acd$$

The two models most commonly used for projecting the impact of incomplete followup are the absolute risk projection model and the relative risk projection model. Both models allow for a latency period -- during which there is no radiation-induced cancer risk -- and a plateau (or expression) period -- during which the effects of exposure to radiation are expressed. The plateau period may be of fixed length -- e.g., 25 years -- or the risk may be assumed to persist for the remainder of the exposed individual's life. The key difference between the absolute and relative risk projection models is the assumption made with respect to the pattern of radiation-induced risk during the expression period. Under the absolute risk projection model the risk is assumed to be constant. Under the relative risk projection model the risk is assumed to be a constant fraction of the baseline age-specific risk. Because background rates for most cancers strongly increase with age, the relative risk model tends to yield projections of risk that are higher than those derived using the absolute risk model.

A secondary issue related to the application of the relative risk model is whether the fractional increase in cancer risk associated with a specific dose depends on the age at which the dose is received. Although existing data do not clearly resolve this point, several recent analyses suggest that relative risks decrease with increased age at exposure. Therefore, while central estimates of the risks of most solid tumors are based on the assumption that relative risks do not depend on age-at-exposure, the upper estimates for these same cancers are calculated assuming that relative risks for those younger than 20 at the time of exposure are three times as large as for those over 20.

Table V gives the cancer mortality models recommended by the working group. Separate models are provided for estimating the risks of leukemia, bone cancer, breast cancer, lung cancer, gastrointestinal cancers (including cancers of the esophagus, stomach, colon, rectum, pancreas and liver), thyroid cancer, a residual category of "other" cancers (which is intended to reflect cancers of the bladder, kidney, brain, female reproductive organs and lymphomas), and for both leukemias and other cancers associated with <u>in utero</u> exposure. This site-specific approach was taken in recognition of the non-uniformity of the organ doses that may occur in nuclear power plant accidents. All doses are in Gray (Gy).

Table V. Cancer Mortality[a]

Effect	Lifetime Risk (deaths/1000)		
	Central	Lower	Upper
Leukemia	$1.4\ d + 2.3\ d^2$	$0.5\ d + 2.9\ d^2$	$4.8\ d$
- In Utero[b]	$0.12\ d$	$0.12\ d$	$0.3\ d$
Bone Cancer	$0.06\ d + 0.09\ d^2$	$0.02\ d + 0.12\ d^2$	$0.2\ d$
Breast Cancer[c]	$6.0\ d$	$0.4\ d + 2.4\ d^2$	$8.7\ d$
Lung Cancer	$2.0\ d + 3.1\ d^2$	$0.5\ d + 3.0\ d^2$	$25\ d$
GI Cancer	$5.7\ d + 8.9\ d^2$	$0.9\ d + 5.4\ d^2$	$33\ d$
Thyroid Cancer[d]	$0.7\ d$	$0.7\ d$	$0.7\ d$
Other Cancers	$2.9\ d + 4.5\ d^2$	$0.5\ d + 3.0\ d^2$	$17\ d$
- In Utero[b]	$0.12\ d$	$0.12\ d$	$0.3\ d$

a - Doses are organ-specific absorbed dose (Gy).

b - Apply to population. Risks to children in utero are 100 times larger.

c - Apply to population. Risks to women are twice this large.

d - Dose from I^{131} is less effective. Such dose is reduced by a factor of 3 in derivation of central estimates, and by a factor of 10 in calculation of lower estimates.

e - Includes lymphoma, multiple myeloma, and cancers of the brain, kidney, bladder and uterus. Excludes skin and prostate cancer and all cancers for which separate risk models have been developed.

The cancer models described in the 1985 report were based primarily on information discussed in BEIR III (NAS, 1980) and data published subsequently by Kato and Schull (1982) and Wakabayashi et al. (1983). The revised models presented in Table V reflect information contained in the Report of the National Institutes of Health ad hoc Working Group to Develop Radioepidemiological Tables (NIH, 1985) and more recent data from the ongoing followup of the survivors of the bombings at Hiroshima and Nagasaki (Preston, 1987). However they have not been adjusted to account for any impact of the reassessment of the doses received by the Japanese survivors.

Our complete report (NRC, 1990) includes models for cancer morbidity and data useful for estimating the impact of each type of cancer -- i.e., life years lost per case, typical interval between diagnosis and death. For most cancers, the estimates of morbidity and mortality risks are roughly equivalent -- because they are almost always lethal. Notable exceptions are thyroid cancers -- only 1 in 10 results in death -- and breast, gastrointestinal and "other" cancers -- which yield 2 to 3 times as many cases as deaths.

Genetic Effects

A slight increase in the incidence of genetic disease would also be expected following a nuclear power plant accident. The genetic risk would be expressed both directly -- i.e., as an increase in the incidence of birth defects among the children of the exposed population -- and indirectly -- i.e., through latent mutations expressed in their grandchildren, great grandchildren and in subsequent generations. In addition, there would be small increases in the rates of spontaneous abortion -- primarily occurring within the first few days of pregnancy before the fertilized ovum is implanted in the uterine wall.

The NRC models for genetic effects were developed by a working group led by Dr. Seymour Abrahamson of the University of Wisconsin that included Dr. Michael Bender - Brookhaven National Laboratory, Dr. William J. Schull - University of Texas, and Dr. Carter Denniston - University of Wisconsin.

The working group relied heavily on analyses provided in the BEIR I and BEIR III reports of the National Academy of Sciences (NAS, 1972, 1980) as well as those described in recent reports of the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 1977, 1982). These basic approaches have been modified in several important respects to reflect new scientific information and improvements in analytic methodologies for risk assessment.

The possible genetic effects are too numerous to be considered individually. Models of major classes of genetic disease have been developed that reflect the key differences in induction, transmission and significance of these conditions. The three major classes of genetic disease considered in this report are single gene disorders, chromosome anomalies and multifactorial diseases.

Muscular dystrophy and Huntington's chorea are examples of single gene disorders. About 1 percent of all children suffer from some form of single gene defect. Down syndrome and both Kleinfelter and Turner anomalies reflect chromosome aberrations. About 0.5 percent of children are born with chromosome anomalies. Multifactorial genetic diseases include congenital malformations (e.g., spina bifida and cleft palate), constitutional diseases and degenerative diseases. This is, by far, the largest class of genetic disease -- affecting about 9 percent of the population.

Estimates of radiation-induced genetic risks are based on extrapolations from animal models. The limited human data relevant for genetic risk assessment comes from studies of the children of survivors of the atomic bombings of Hiroshima and Nagasaki. Although these studies have not revealed an excess incidence of genetic defects, they are not powerful enough to reject current models of radiation-induced risk.

The responses observed in the spermatogonial cells of the mouse serve as an indication of the effects that would be expected to occur in the spermatogonial cells of men. Unfortunately there is no adequate mammalian model of genetic damage in the human female. The working group's central and upper estimates of risk are based on the assumption that damage to oocytes and spermatogonia is equivalent. Their lower risk estimates are derived from an analysis that assumes oocytes are not susceptible to radiation-induced damage.

When dose is received at low dose rate, the risk of genetic damage is thought to be proportional to the dose received. However evidence from many different experimental studies -- i.e., *Drosophilia oogonia* mutations, *Trandescantia* mutations -- indicates that when dose is received at high dose rate the yield of mutations is better described by a linear-quadratic relationship. Such a result is consistent with radiobiological understanding of the mechanism of damage -- i.e., that the majority of radiation-induced mutations in higher organisms are tiny submicroscopic deletions, inversions or insertions encompassing parts of one or more genes; single nucleotide changes appear to be extremely rare.

When a linear-quadratic relationship -- e.g., (a + bd) cd -- is fit to the data on specific locus recessive mutations in the spermatogonia of mice (with dose in Gy) the coefficients *a* and *b* are found to be virtually identical. This result has been used as the rationale for estimating the risks of most genetic effects using models of the form:

$$r = (1 + d)\ acd$$

where d is the gonadal dose (Gy), the product *ac* is the risk coefficient observed at low dose rates, and the term *1 + d* modifies the risk estimate to account for the effects of high dose rate. When an accident scenario involves only chronic exposure at low dose rate, the modifying term is dropped and the risk is computed as:

$$r = acd$$

This same simplification may be used when some of the dose is received at high dose rate -- as long as the total dose is reasonably small. For example, at a dose of 0.5 Gy the risk would be underestimated by only 50% using this approach. For lower doses, the bias is even smaller and is negligible in comparison with the fundamental uncertainties in current estimates of the risk coefficients.

Table VI gives the models recommended for estimating genetic risks. The values shown in the table are estimates of the integrated risk -- i.e., the risk summed over all future generations. All of the numerical chromosome aberrations and two-thirds of the structural anomalies will be expressed within one generation (about 30 years) of the accident. Half of the single gene defects are expected to occur within the first three generations. The dynamics of multifactorial diseases are poorly understood and, therefore, no estimate of their timing was developed.

It should be noted that the simple forms of the genetic risk models given in the table assume that the doses received by the mother and father are equal. The modifications necessary to allow for differences in maternal and paternal doses are described elsewhere (NRC, 1990).

These results assume population stability, an intergenerational interval of 30 years, and a crude birthrate of about 16 births per 1000 persons per year (1/2 birth per person per generation). Were the population to increase, the absolute impact -- i.e., number of effects -- would increase accordingly.

Discussion

Revised health effects models have been developed for nuclear power plant accident consequence analysis.

Models for early health effects have been modified to include thresholds. For the two dominant causes of early death -- the hematopoietic and pulmonary syndromes -- a new approach has been devised to account for the dependence of the median lethal dose on the dose rate. All parameters for estimating the risks of early effects were reviewed, and where appropriate revised, to ensure consistency with the available data on human radiation injury. In addition, upper and lower estimates have been developed for all early and continuing health effects.

Models for cancer have been revised to reflect the possibility that relative risk is highest for those who are young at the time of exposure. The upper estimates of the risks of lung cancer, gastrointestinal cancer and "other" cancers have been modified accordingly.

Table VI. Genetic Risk[a]

Effect	Integrated Risk (cases/1000)		
	Central	Lower	Upper
Single Gene Defects			
Dominant	$7.5\ d + 7.5\ d^2$	$1.2\ d + 1.2\ d^2$	$22\ d + 22\ d^2$
X-linked	$2.2\ d + 2.2\ d^2$	$0.5\ d + 0.5\ d^2$	$9\ d + 9\ d^2$
Chromosome[b] Anomalies			
Numerical	$0.5\ d$	0	$1.5\ d$
Structural	$0.9\ d + 0.9\ d^2$	$0.1\ d + 0.1\ d^2$	$6.5\ d + 6.5\ d^2$
Multifactorial	$7.2\ d + 7.2\ d^2$	$0.45\ d + 0.45\ d^2$	$45\ d + 45\ d^2$
Pregnancy Losses[b]			
Numerical	$4.5\ d$	0	$13.5\ d$
Structural	$8.1\ d + 8.1\ d^2$	$0.9\ d + 0.9\ d^2$	$58\ d + 58\ d^2$

a - Doses are organ-specific absorbed dose (Gy). Integrated risk is summed over all future generations and is expressed as the number of cases per 1000 persons receiving a gonadal dose d.

b - Chromosomal anomalies may result in severe physical and mental defects or they may lead to loss of pregnancy.

Models of genetic effects have been expanded to facilitate calculation of the increased risk of peri-implantation pregnancy losses (spontaneous abortions occuring primarily during the first few days of pregnancy) caused by severe chromosome anomalies. In addition, the estimates of uncertainty in calculating some genetic risks have been revised.

Although these revised models represent an improvement over the RSS and original NUREG/CR-4214 models, they must be periodically revised to reflect new developments in epidemiology and radiobiology. In particular, the models for assessing cancer risk must be reconsidered in light of the reassessment of the doses received by the survivors of the atomic bombings of Hiroshima and Nagasaki.

Acknowledgements

These models were developed with funding from the U.S. Nuclear Regulatory Commission through a cooperative agreement with Sandia National Laboratories. The report reflects the collaborative efforts of many individuals. Primary among these are the working group leaders -- Dr. Bobby Scott, Dr. Ethel Gilbert, Dr. Seymour Abrahamson and Dr. Harry Maxon. The original project leaders -- Dr. Douglas Cooper and Dr. Dade Moeller -- also deserve special mention. Many other scientists have made significant contributions to the development of the models. Drs. Neil Wald, Albert Spritzer, and Joseph Watson reviewed and interpreted the data on human radiation injury that provides the underpinnings of the analysis of early health effects. Drs. Roy Shore and Nan Laird provided technical information and analyses that supported the development of skin and thyroid cancer models. Dr. Syed Mansur performed many of the calculations needed to develop population-based cancer and genetic risk models.

The members of our scientific advisory committee -- whose names are listed below -- critically reviewed both the 1985 report and the more recent revision of the report. Every effort has been made to respond to their comments. Nonetheless, membership on the advisory committee should not be interpreted as implying endorsement of the models.

Dr. Seymour Abrahamson
Dr. William Bair
Dr. Michael Bender
Dr. Bruce Boecker
Dr. Victor Bond
Dr. Richard Cuddihy
Dr. Keith Eckerman
Dr. Jacob Fabrikant
Dr. Marvin Goldman
Dr. George Hutchison
Dr. Dade Moeller
Dr. Edward Radford
Dr. Eugene Saenger
Dr. Warren Sinclair
Dr. Neil Wald
Dr. Edward Webster
Dr. Shlomo Yaniv

Additional review was provided by the following experts -- Dr. Gilbert Beebe, Dr. Thomas Cochran, Dr. James Crow, Dr. Troyce Jones, Dr. Charles Land, Dr. Clarence Lushbaugh, and Dr. Robert Young.

References

1. Nuclear Regulatory Commission (1975): Reactor Safety Study: An Assessment of Accident Risks in U.S. Commercial Nuclear Power Plants, Appendix VI, WASH-1400, NUREG-75/014, United States Nuclear Regulatory Commission, Washington, DC.

2. Nuclear Regulatory Commission (1985): Health Effects Model for Nuclear Power Plant Accident Consequence Analysis, NUREG/CR-4214, United States Nuclear Regulatory Commission, Washington, DC.

3. Nuclear Regulatory Commission (1990): Health Effects Model for Nuclear Power Plant Accident Consequence Analysis, Revision 1, NUREG/CR-4214, Rev 1, United States Nuclear Regulatory Commission, Washington, DC.

4. NCRP 64 (1980): Influence of Dose and Its Distribution in Time on Dose-Response Relationships for Low-LET Radiations, NCRP Report 64, National Council on Radiation Protection and Measurements, Washington, DC.

5. BEIR III (1980): The Effects on Populations of Exposure to Low Levels of Ionizing Radiation, National Research Council Committee on the Biological Effects of Ionizing Radiations, National Academy of Sciences, National Academy Press, Washington, DC.

6. Kato, H. and Schull, W.J. (1982): "Studies of the Mortality of A-Bomb Survivors, 7. Mortality, 1950-1978: Part I. Cancer Mortality," Radiation Research, 90: 395-432.

7. Wakabayashi, T., Kato, H., Ikeda, T. and Schull. W.J. (1983): "Studies of the Mortality of A-Bomb Survivors, Report 7, Part III. Incidence of Cancer in 1959-1978, Based on the Tumor Registry, Nagasaki," Radiation Research, 93:112-146.

8. NIH (1985): Report of the NIH Ad Hoc Working Group to Develop Radioepidemiological Tables, NIH Publication 85-2748, U.S. Department of Health and Human Services, Washington, DC.

9. Preston, D.L., Kato, H., Kopecky, K.J. and Fujita, S. (1987): "Studies of the Mortality of A-Bomb Survivors, 8. Cancer Mortality, 1950-1982," Radiation Research, 111: 151-178.

10. BEIR I (1972): The Effects on Populations of Exposure to Low Levels of Ionizing Radiation, National Research Council Committee on the Biological Effects of Ionizing Radiations, National Academy of Sciences, National Academy Press, Washington, DC.

11. UNSCEAR (1977): Sources and Effects of Ionizing Radiation, United Nations Scientific Committee on the Effects of Atomic Radiation, United Nations, New York.

12. UNSCEAR (1982): Ionizing Radiation: Sources and Biological Effects, United Nations Scientific Committee on the Effects of Atomic Radiation, United Nations, New York.

DISCUSSION

FIRST: Can you tell us how the BEIR V report will impact on these model results?

EVANS, J.: Although we didn't hold up the publication of the report for BEIR V, the two things that influenced BEIR V primarily were the dosimetry revision and the extended followup. The data from the extended followup was available in a Preston and Pierce publication in 1987 and that was the basis for revising our upper estimates of these solid tumors. I think really what would happen if we were to reconsider, would be that our central estimates would be moved towards the upper estimates. In view of the consensus about BEIR V, we would reflect linearity for the solid tumors (which we are already doing in our upper estimates) and greater emphasis on relative risk with age-at- exposure specific risk coefficients.

CLOSING COMMENTS OF SESSION CO-CHAIRMAN MOELLER

We have heard three papers in this opening portion of the initial session. Dr. Melvin W. First provided us with an outline of the history of the Conference and set forth some of the goals we hope to achieve during this meeting. This was followed by a presentation by Dr. Paul L. Ziemer, Assistant Secretary for Environment, Health and Safety, U.S. DOE, who described the new attitude or "culture" that has been adopted within his agency, with the objective being to focus more of the attention of the Department on providing protection for the health and safety of workers and facilities, as well as for members of the general public living in surrounding areas. Specific initiatives include improved health related data collection and analyses and the promotion of the independent conduct of related epidemiology studies. The latter is to be accomplished through transferring the responsibility for such studies to the U.S. Department of Health and Human Services. Another part of the program is to have DOE installations comply with portions of the Occupational Safety and Health Act and the National Environmental Policy Act. The Agency is also moving to be more responsive to the interests and concerns of State and local agencies, and the establishment of better communications and openness with the public.

In a follow-up presentation, Charles A. Willis traced the history of siting and safety regulations for nuclear power plants. He pointed out that the development of such regulations has been an evolutionary process and that the present pause in applications for the construction and operation of new plants has provided time to reflect on and re-evaluate what the NRC has been doing. This effort is being directed primarily to the potential licensing of advanced nuclear power plants and includes (1) seeking to achieve safety improvements through the application of advances in technology and operating experience, (2) improving communications between the NRC and industrial groups on technical issues, and (3) consolidating the issues open to litigation to make the process more efficient and effective. A part of this process is to address the important questions it raises relative to the role of, and requirements for, air cleaning systems in the next generation of nuclear power plants.

SESSION 2

FILTER TESTING

Monday: August 13, 1990
Co-Chairmen: W. L. Anderson
W. Bergman

OPENING COMMENTS OF SESSION CO-CHAIRMAN BERGMAN

NEW INJECTION SYSTEM FOR A SHORT MIXING OF TEST AEROSOLS AND GAS TRACERS INSIDE VENTILATION DUCTS
Ph. Cassette, N. DuPoux, J. C. Laborde

REAL-TIME DETECTION OF A FLUORESCENT AEROSOL. APPLICATION TO THE EFFICIENCY MEASUREMENTS OF HEPA FILTERS
Ph. Mulcey, P. Pybot, J. Vendel

AEROSOL CONCENTRATIONS PRODUCED BY A LASKIN NOZZLE GENERATOR A COMPARISON OF SUBSTITUTE MATERIALS AND DOP
D. W. Crosby

CHARACTERISTICS OF LASKIN NOZZLE GENERATED AEROSOLS
Xiaowei Yan, M. W. First, S. N. Rudnick

SAFE REPLACEMENT MATERIALS FOR DOP IN "HOT SMOKE" AEROSOL PENETROMETER MACHINES
H. R. Carlon, M. A. Guelta

DEVELOPMENT OF FILTER SYSTEMS - PART 2, INJECTION SYSTEMS AND MULTI-POINT SAMPLER EVALUATION
D. Loughborough, D. Morris, S. Capon

THE DUST HOLDING CAPACITY OF HEPA FILTERS
D. Loughborough

SIMULATED PERFORMANCES OF A SINGLE PARTICLE LASER LIGHT SCATTERING ANALYZER
E. Neri, V. Massacurati, M. Platini

CLOSING COMMENTS OF SESSION CO-CHAIRMAN ANDERSON

OPENING COMMENTS OF SESSION CO-CHAIRMAN BERGMAN

Welcome to the session on Filter Testing. It is an honor to co-chair this session with Wendell Anderson, who is one of the pioneers in this field. Today we have eight papers dealing with various aspects of filter testing. Two of the papers deal with proper mixing of aerosols with the air stream to assure representative measurements of aerosol concentration. This topic has been a recurrent theme at several of the recent air cleaning conferences, but it is an important one because of its criticality in terms of accurate results. We have four papers on aerosol generation, three of which pertain to DOP test methods, using hot DOP and Laskin nozzle generated DOP. One deals with a uranine aerosol generation method. Although these topics have been covered at other air cleaning conferences, the vitality of the subject and the new information that has been obtained in recent years are such that tests methods are continually improving. We are looking forward to some new improvements here, also. We have one paper on dust holding capacity of HEPA filters. This is a very critical topic that has not found its way into test standards. We have a test standard for filter efficiency, but we do not have one for dust holding capacity. I am reminded of the research that Professor First did years ago trying to establish dust loading capacity for a variety of filter designs. There were dramatically different results depending on the filter design. This is a critical topic. Finally, we have a paper on a new light scattering technique. It will be a variation of former studies done in this area.

NEW INJECTION SYSTEM FOR A SHORT MIXING OF TEST AEROSOLS AND GAS TRACERS INSIDE VENTILATION DUCTS

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Abstract

The use of tracers, in the form of gases or aerosols, to characterize a physical system (ventilated rooms, air cleaning devices, ...) necessitates, when injected inside ducts, being able to detect them at an adequate distance from the point of emission to ensure that the "carrier-tracer" fluid mixture is homogeneous. In many practical instances, it is difficult to have an adequate distance available for proper homogenizing and it proves necessary to reduce the mixing length to the maximum possible extent.

The paper presents a system of injection developed at the Saclay Nuclear Studies Centre. The principle is based on high-speed injection which is perpendicular to the flow and on a multiplicity of jets.

The results obtained from Helium injection (representative of tracer gases), from soda fluorescein and from DOP (representative of test aerosols used for efficiency measurements of HEPA filters) are given for cylindrical and parallelepipedic duct configurations.

This technique allows an homogeneous mixture to be obtained at a distance of the order of 3 to 5 diameters, whatever the flow velocity.

I. Introduction

Tracers in the form of gases or aerosols are frequently used in the nuclear industry to characterize the condition of a ventilation system (measurements of air flow, air change rates,...) transfers of contamination linked to this ventilation (transfer time, transfer coefficients, ...) and air cleaning systems (efficiency measurements of High Efficiency Particulate Air filters).

When using a tracer, the characterization of a physical system is generally the result of the measurements of the tracer concentration at a given point. These measurements require an homogeneous mixture of the tracer in the carrier fluid. For this, it is necessary to take a sample of tracer at a sufficiently long distance from the injection point, which is then called as the mixing length. It is defined as the minimum distance downstream from the injection cross-section beyond which the injected tracer is sufficiently well distributed over a cross-section to enable the concentration to be measured with the accuracy required[1].

Taking account of the impossibility, in practice, of having adequate homogenizing lengths available, they may be reduced by using various methods of injection and types of injectors: annular injection, injectors with multiple orifices, high-speed jets, vortex generators. The method of injection presented in this paper and developed at the Saclay Nuclear Studies Centre, combines high-speed injection perpendicularly to the flow with a multiplicity of jets. In order to characterize the performance of the injector, tests were carried out using first of all a gaseous tracer (Helium) then particulate tracers (soda fluorescein and DOP).

II. Description of the mixing length and the theory of jets

Mixing length

The mixing length is expressed in the case of central injection and on the assumption of a constant diffusion coefficient and a constant velocity by the following correlation[1]:

$$\frac{X}{D} = 1.18 \sqrt{\frac{8}{\lambda}} \left(2.94 - \frac{\ln \Delta C}{2.3}\right) \qquad (1)$$

X : straight length under consideration

D : diameter of duct

λ : friction factor

ΔC : maximum variation of the concentration in the cross-section of the duct, expressed in %

Figure 1 represents the variations of X/D as a function of ΔC for a Reynolds number $R_e = 10^5$ and a smooth pipe-system.

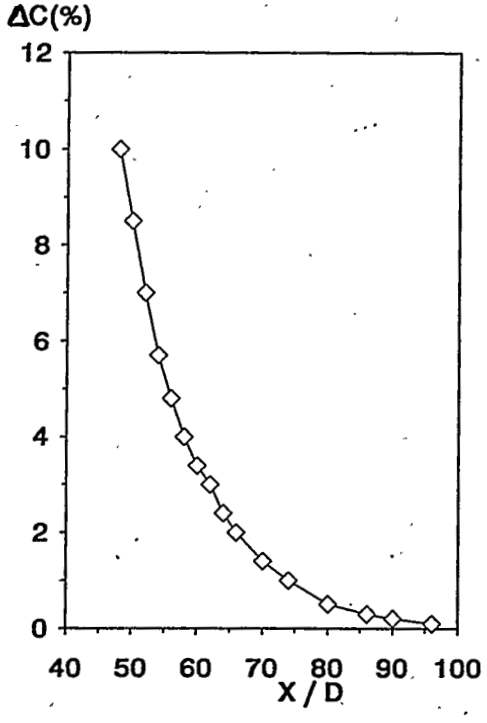

Figure 1 Mixing length

It will be noted that a maximum variation of concentration of 10% requires 48 diameters. For 5%, it needs 56 diameters.

Any impediment to the flow producing turbulence (bends, valves, turbofans, ...) produces an improved blending of the mixture and therefore reduces the mixing lengths by a factor which is difficult to evaluate experimentally.

Theory of jets emitted perpendicular to the flow[2]

Let us consider a circular jet of diameter d and velocity V_0 emitted perpendicular to a flow of velocity V_1. Because of the turbulent mixture developing at the periphery of the jet, the outer layers lose a part of their momentum; the jet is deflected and in cross-section takes on the characteristic bean-like shape (figure 2).

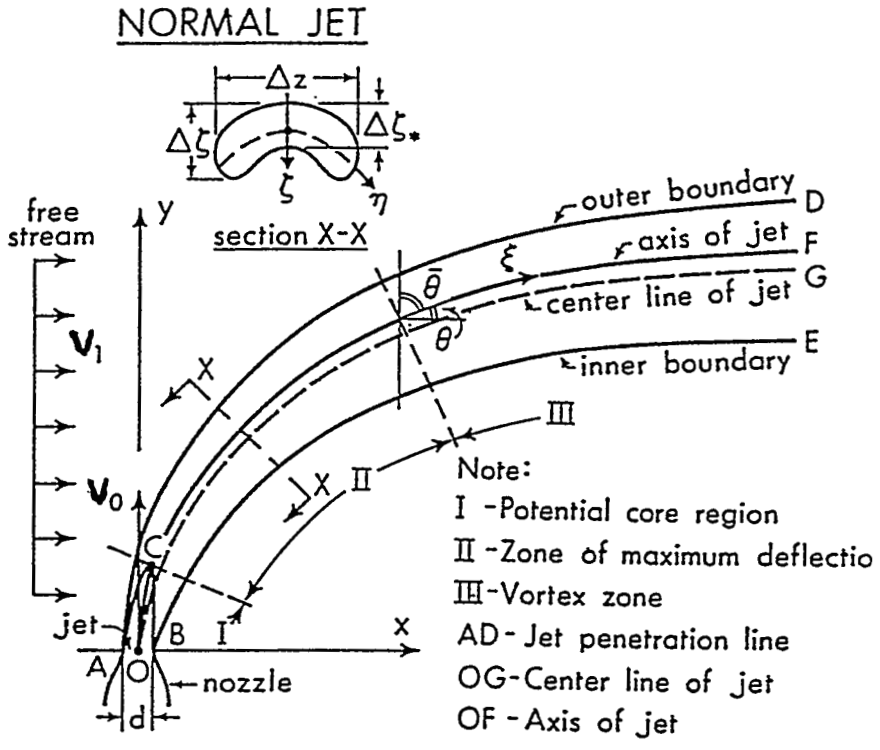

Figure 2 Jet deflection

Many authors have studied the deflection of the jet and have expressed it using empirical relations of the form:

$$\frac{y}{d} = f\left(\frac{x}{d}, \alpha\right) \text{ where } \alpha = \frac{V_0}{V_1}$$

Pratte and Baines established three relationships which express respectively the deflection of the outer layer AD (known as the jet penetration), the deflection of the inner layer BE and the deflection from the central line OG.

Outer layer: $y_E/\alpha d = 2.63\ (\alpha/Yd)^{0.28}$

Inner layer: $y_I/\alpha d = 1.35\ (\alpha/Yd)^{0.28}$

Central line: $y_c/\alpha d = 2.05\ (\alpha/Yd)^{0.28}$

From this we can deduce that the obtaining of an homogeneous mixture of a tracer injected perpendicularly to the flow will be a function of the diameter of the jet, the ratio of the jet velocity to the flow velocity and the distance between the sampling and injection points.

III. Design of a multiple-jet injector

Taking into account the behaviour of a jet emitted perpendicularly at high speed into a flow, several of these jets were brought together to cover the whole cross-section of the duct and, through the interaction of the jets with one another, improving the blending of the mixture. After testing several designs of injectors incorporating various numbers of orifices with various diameters and shapes, the system shown in figure 3 was retained.

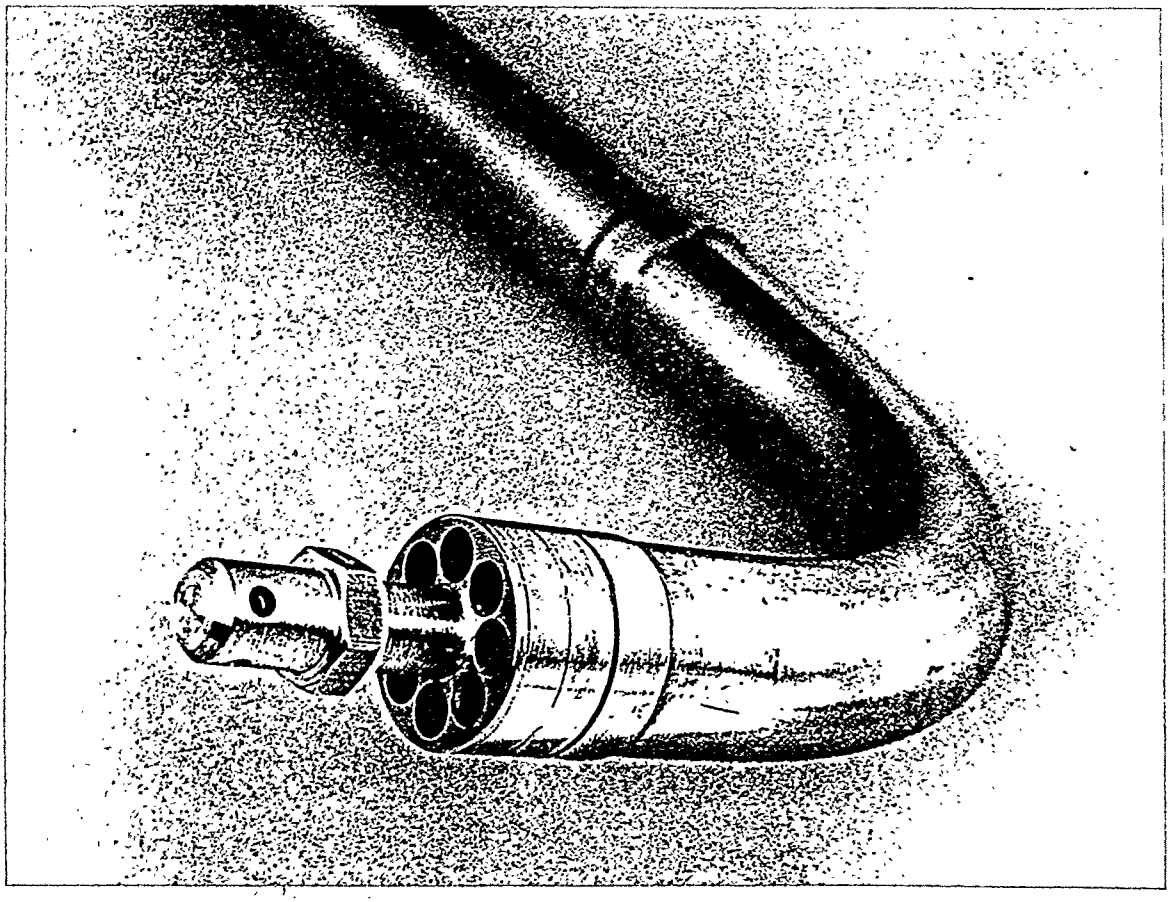

Figure 3 $I_{1.5}$ Injection system

The injection apparatus $I_{1.5}$ consists of a brass cylinder, in the thickness of which 4 orifices, arranged at 90° have been drilled. Before being ejected from the nozzle, the jet passes through a straight section in order to homogenize the velocity field in the flow. This part is screwed into a hollow pipe through which the nozzle is supplied with tracer-air mixture. In the case of aerosol injection, the injection pipe is fitted with a central tube through which the nozzle is supplied with compressed air, and a concentric tube of an higher diameter through which the aerosol is introduced. The additional air flow rate, measured using a mass flow meter, may be varied in order to increase or reduce the ejection velocity of the jet without changing the reference concentration of the tracer in the flow. The 4 jets are thus emitted at velocity V_0 into the duct perpendicularly to the flow, the velocity of which is V_1; injection is carried out at the centre of the duct.

IV. Characterization of the performance of the multiple-jet injector

IV.1 Operating conditions

Geometry of ducts

In practice, when a tracer is used to characterize the operational condition of a ventilation system or air cleaning systems, ducts are encountered with varying geometry and dimensions. The injector was therefore tested in various duct configurations:

- circular cross-sections[3]: diameter: between 0.30 m and 0.60 m
- rectangular cross-sections[4]: dimensions: 0.40 m x 0.25 m

Tracers used

In order to be representative of gaseous and particulate tracing techniques, three tracers were used; their characteristics are summarised in the following Table 1.

Test rig

To evaluate the performance of the injector, the test rig, shown in figure 4, was used. It includes:

- lengths of ventilation ducts of varying geometry,
- a HEPA filter for working with clean air,
- a fan for air circulation
- a production and detection system suited to the tracer used,
- the injection system being studied.

Several sampling points are inserted in order to characterize the homogeneity of the mixture

Table 1

Tracer	Helium	Soda fluorescein	DOP
Nature	Gas	Solid aerosol	Liquid aerosol
Size		Mass median diameter: 0.15 μm Standard deviation: 1.4	Mass median diameter: 0.6 μm Polydispersed
Production of aerosol	He pressurized + mass flow meter	Pneumatic spraying	Pneumatic spraying
Detection	Mass spectrometer	Condensation Nucleus Counter	Condensation Nucleus Counter
Main Applications	- flow metering - leak detection - operating time distribution - air change rate	- HEPA filter testing - Simulation of contamination transfers	HEPA filter testing

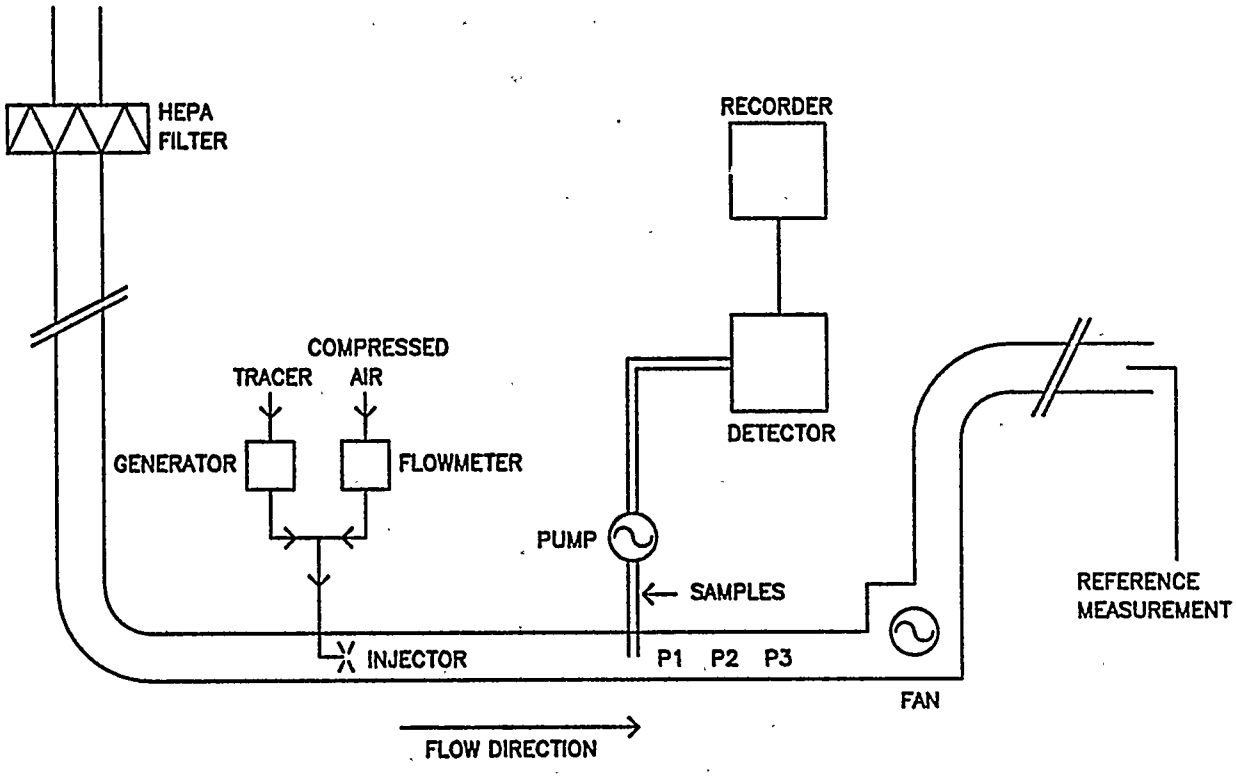

Figure 4 Test rig

Operational method

A metal tube is introduced into the duct at various sampling points where it is desired to measure the tracer concentration. By moving it vertically inside the duct, an exploration may be made following one diameter and the concentration of tracer is so determined along this diameter. The average concentration of tracer thus measured is then compared to the reference concentration measured at a point where the mixture is truly homogeneous.

IV.2 Characterization test carried out

Many tests were carried out in order to characterize the development of the jet, to study the influence of the distance crossed by the tracer inside the injection nozzle and to analyse the influence of the jet and flow velocities.

Moreover, the performance of the new injector was compared to that of other apparatus.

Table 2 below summarizes the characteristics of the significant tests made during the study.

Table 2

Test n°	Duct Cross-Section - Dimension (m)	Tracer	Injector Type	V_0 (m/s)	V_1 (m/s)	L/ad
1	Circular - 0.32	He	New injector $I_{1.5}$ 4 holes	200	7.6	8.1
2	Circular - 0.54	He	New injector $I_{1.5}$ 4 holes	250	5.6	8.1
3	Circular - 0.32	He	New injector I_2 4 holes	148	7.6	8.2
4	Circular - 0.36	Soda fluorescein	New injector $I_{1.5}$ 4 holes	113	3.8	8.1
5	Circular - 0.36	DOP	New injector $I_{1.5}$ 4 holes	97	3.8	9.3
6	Circular - 0.32	He	Injection parallel to the flow - 1 hole	204	7.6	8
7	Circular - 0.32	He	Injection perpendicular to the flow - 1 hole	204	7.6	8
8	Circular - 0.36	Soda fluorescein	Injection perpendicular to the flow - 1 hole	7.9	7.7	44
9	Rectangular 0.40 x 0.25	He	New injector $I_{1.5-2.4}$ 4 holes -towards wall	288	8.1	7.5
10	Rectangular 0.40 x 0.25	He	New injector $I_{1.5}$ 4 holes at 115° and 65° - towards angles	412	7.5	9

Observations

1) L: characteristic dimension of the duct

- L = D in the case of a duct of circular cross-section
- L = distance between 2 opposite walls when the jet is emitted in the direction of the walls (rectangular cross-section)
- L = Diagonal of the rectangular cross-section when the jet is emitted in the direction of the angles of the duct.

2) Test 8 corresponds to injection conditions normally used for HEPA filter tests.

V. Characteristic results

The performance of the injection is characterized by the relationship: C/C_{ref}

C : concentration of tracer at the point being analysed

C_{ref}: reference concentration at a point where the mixture of the tracer is homogeneous

At a given distance X from the point of injection, the variations of the C/C_{ref} relationship are represented in a section perpendicular to the flow.

Ducts with a circular cross-section

Figure 5 show the variations of C/C_{ref} for various values of the relationship X/D for ducts with a circular cross-section.

For a duct with a diameter D = 0.32 m, it can be observed that at 3D from the injection, an homogeneous mixture is obtained in the central part of the conduit. In the area immediately surrounding the wall, the difference between the measured and reference concentrations does not exceed 30%. At 5D, this maximum difference is 10% at 2 cm from the wall, which is quite negligible; it may therefore be considered that the mixture of the tracer in the flow is, at this distance, homogeneous. At 7D, the mixture is completly homogeneous, even in the area around the wall.

These observations are also valid for the homogeneity of the mixture in the case of a duct with a diameter D = 0.54 m.

The criterion for homogeneity is independent of the type of tracer as is shown by the results obtained by injecting DOP or soda fluorescein (figure 5c and 5d): the mixture is homogeneous at 6D.

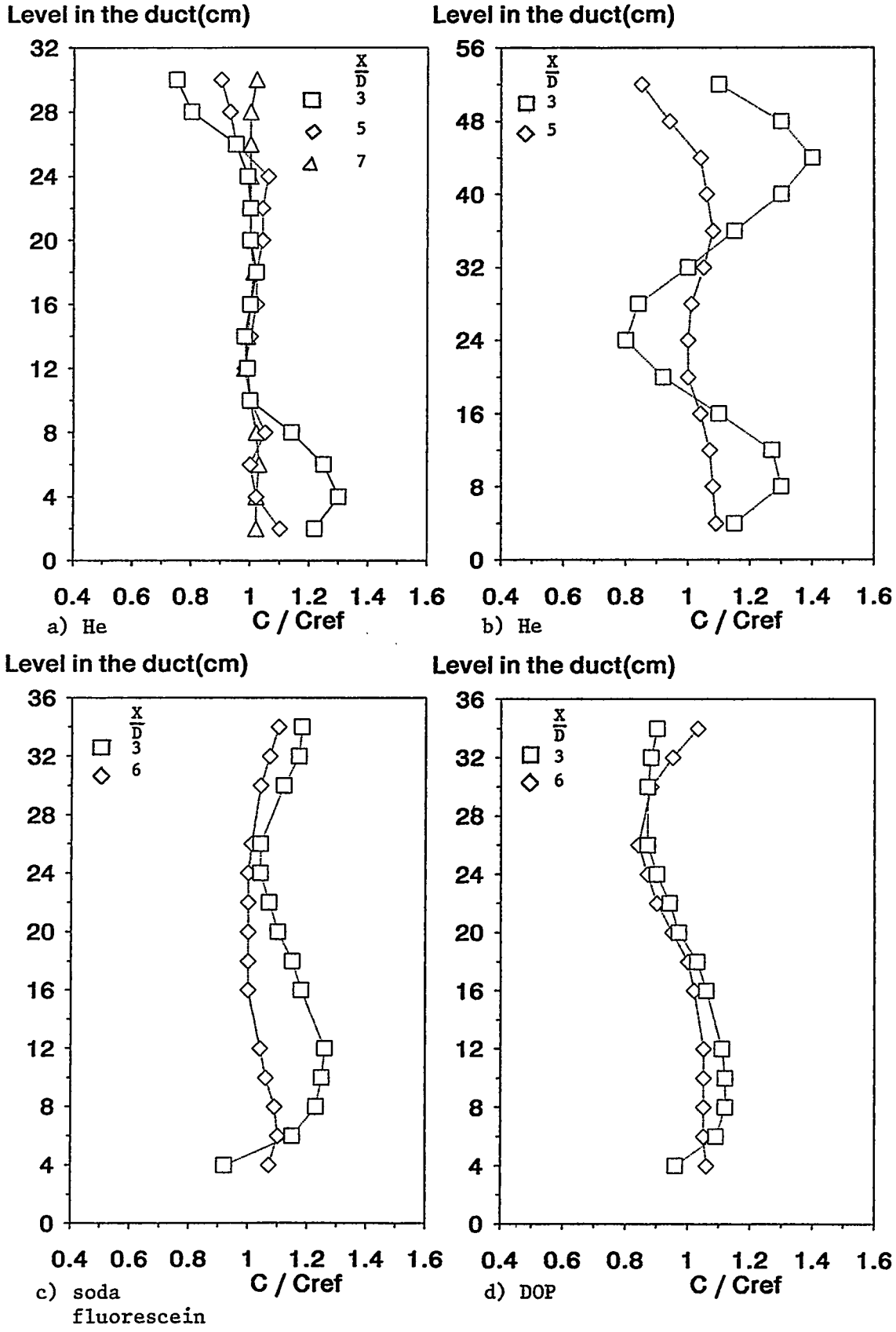

Figure 5 Producing an homogeneous mixture with the $I_{1.5}$ injection

This conclusion is valid with submicronic aerosols (case of DOP and soda fluorescein used for the HEPA filters testing). For aerosols with diameters above 3 μm, tests are necessary to confirm previous results taking into account the influence of inertial and gravitational phenomena.

Duct with a rectangular cross-section

Two series of tests were carried out: in the first, the efficiency of an injector incorporating 2 orifices with a diameter of 1.5 mm and 2 orifices with a diameter of 2.4 mm arranged alternately and forming angles of 90° between them was tested. The jets are emitted towards the walls. In the second series of tests, the injector incorporated 4 orifices, forming angles of 115° and 65° between them respectively. The jets were emitted towards the angles of the duct.

Figure 6 show the results obtained at a distance of 4D, for samplings taken within the width and height of the duct. In the case of a duct with a rectangular cross-section D corresponds to the hydraulic diameter defined by:

$$D_H = \frac{4 \times \text{cross-sectional area}}{\text{wetted perimeter}}$$

The results obtained show that the best mixtures correspond to conditions in which the jets are directed towards the angles of the duct.

Influence of the diameter of the orifices of the injector

Tests carried out with various orifice diameters show that this parameter has a negligible influence.

Comparison of the performance of $I_{1.5}$ injector with traditional injection methods

Figure 7 allows the performance of injector $I_{1.5}$ to be compared with the simple methods of injection frequently used:

A) Injector comprising an orifice 1.5 mm in diameter. Injection of Helium in the direction of the flow at the centre of the duct (test 6).

B) Injector comprising an orifice 1.5 mm in diameter. Injection of Helium at the centre of the duct perpendicularly to the flow (test 7).

C) Injector comprising an orifice 8 mm in diameter. Injection of aerosols at the centre perpendicularly to the flow (test 8).

A better homogeneity of the mixture is observed using our injector $I_{1.5}$.

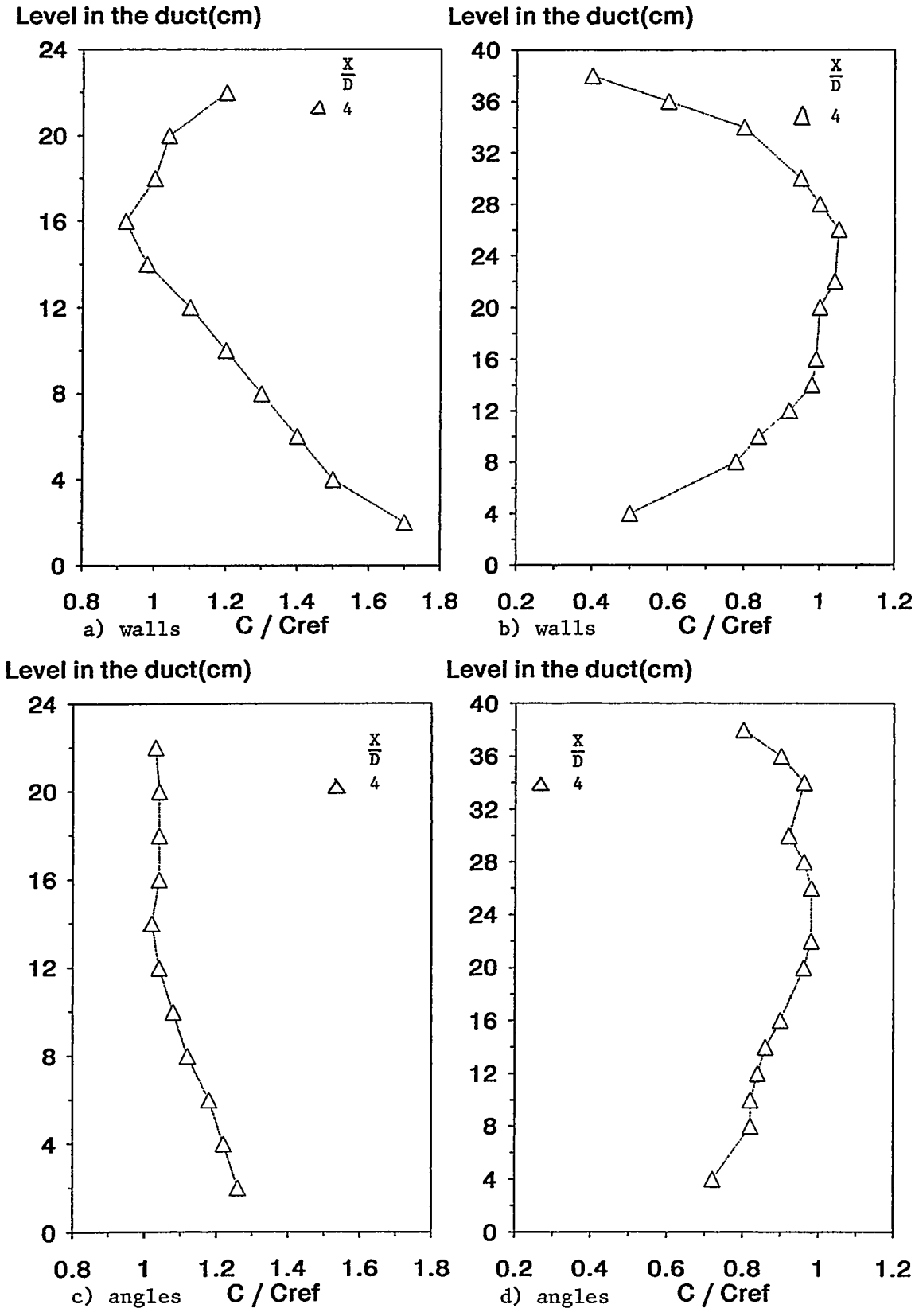

Figure 6 Jets in direction of walls and angles

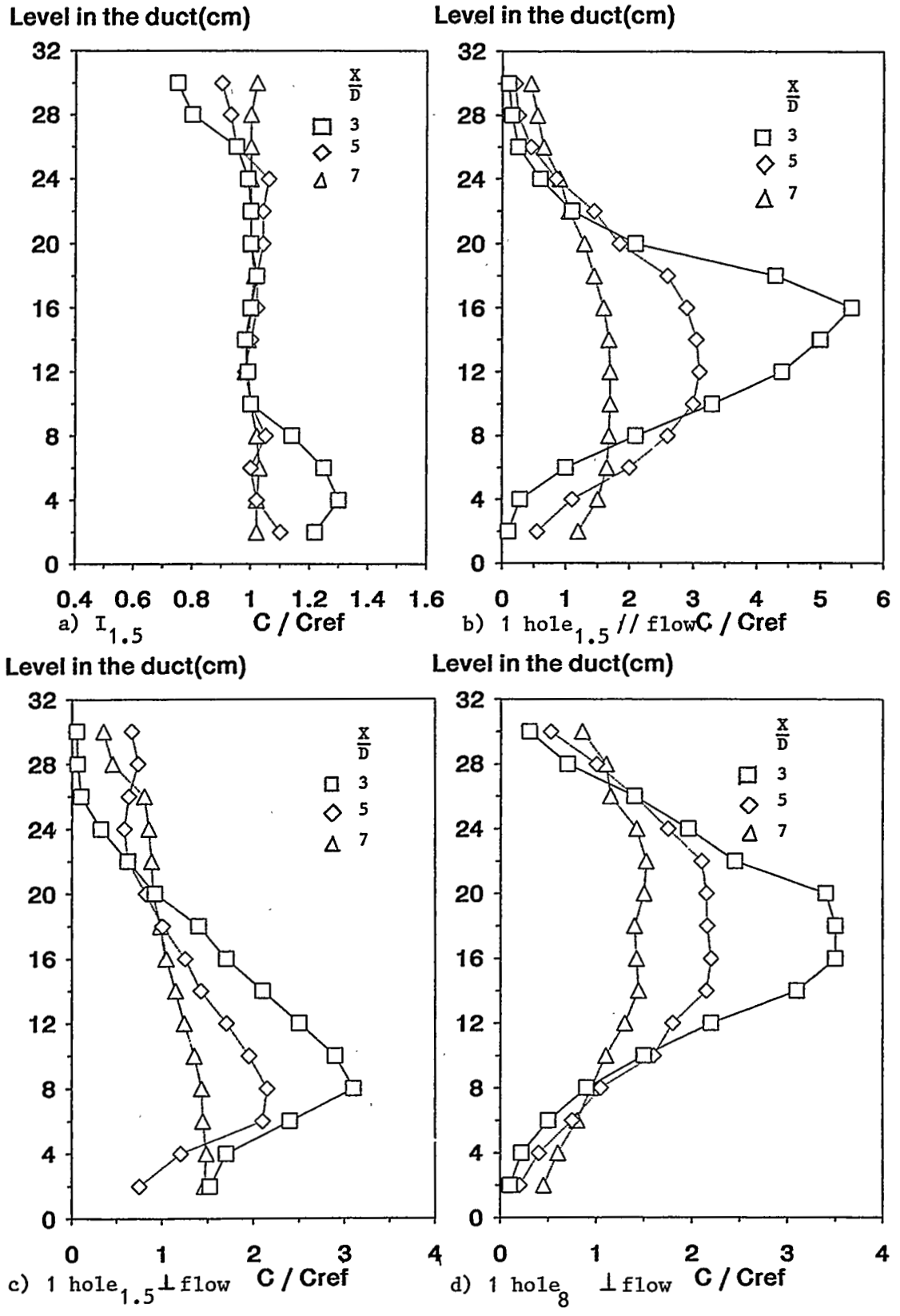

Figure 7 Comparison to the performance of $I_{1.5}$ injector with traditional injectors

Characteristic parameter of $I_{1.5}$ injector operation

The theory of jets imposes the value $\alpha d = V_0/V_1\ d$ (paragraph II).

In our study we were interested in the adimensional number $I = L/\alpha d$

L: characteristic dimension of the duct into which injection is carried out.

For the three types of duct used during the tests, we determined the velocities V_0 of ejection required to obtain an homogeneous mixture at 5D as a function of the velocity V_1 and then calculated the values of I.

Table 2 presented in paragraph IV.2 summarizes the values of I obtained for some significant tests.

It will be seen that the values of I are very close (average value: 8.3 - standard deviation: 0.6) for very different test configurations. It therefore seems to be an invariant characteristic parameter for the entire phenomenon. For a given application, it will thus enable the additional air flow rate from the injector to be calculated, given that we know the geometry of the duct, the flow velocity into the duct and the diameter of the orifices of the injector.

VI. Conclusion

The method of injecting a tracer into a gaseous flow which we have developed enables the mixing length to be reduced by a factor of approximately 10. It is based on the principle of injection with multiple jets perpendicularly to the main flow.

The technique may be adapted to any duct and enables to an homogeneous mixture of the tracer to be obtained at a short distance from the injection (of the order of 5 diameters) with a high degree of accuracy, whatever the flow velocity. This simple system, which is inexpensive and easy to use, only disturbs the flow slightly and offers the advantage in relation to many techniques of not causing any extra pressure drop. It may be used for the injection of gaseous or particulate tracers. The parameters of injection of the system developed may be determined on the basis of knowing the characteristic dimension of the duct L (diameter or diagonal) and/or the flow velocity V_1 and/or the diameter of the injection orifices d and/or the jet velocity V_0 knowing that $\frac{L\ V_1}{d\ V_0} \approx 8.3$ for optimal use of the injector.

Acknowledgements

The writers of this document would like to express their gratitude to Mr Paul MOREL and Camille VAVASSEUR, without whom this study could not have taken place.

References

(1) "Mesure de débit des fluides dans les conduites fermées" Norme AFNOR X10-141

(2) N. RAJARATMAN "Turbulent jet" - Elsever Scientific Publising Company, New York 1976

(3) N. DUPOUX, P. MOREL, C. VAVASSEUR "Mise au point d'une méthode d'injection d'un traceur dans un écoulement permettant d'obtenir un mélange homogène à faible distance" CEA Report, SEIP/6.3.1.2, October 1987

(4) N. DUPOUX, P. MOREL "Mélange homogène à faible distance - Cas des canalisations rectangulaires" CEA Report, SEIP/6.3.1.2, September 1988

DISCUSSION

ORTIZ: The question I have, Dr. Vendel, is in reference to your aerosol injection tubes. We have found that some of our injection tubes, which are not very similar to yours, show particle losses. Did you find particle losses associated with your injection tubes and if so, what sizes were affected?

VENDEL: There are certainly losses in the system but it is not a problem because it is used for measuring the decontamination factors. We sample upstream and downstream of the filter. It is not a problem if we have some losses in the particle injector. We want an homogenous concentration. Particle size was not considered.

GUEST: I must congratulate you on a simple solution to a long-standing problem. Is it possible to borrow or buy engineering drawings of the injector?

VENDEL: It is not commercial at this time.

MORRIS: I always find it gratifying when somebody else is doing the same sort of work that I am doing. In fact we have come up with almost exactly the same design. It is commercial and we will be marketing it shortly. The question I have is, do you find differences when you use different numbers of nozzles and what sort of differences in performance do you get when you use a large number of nozzles with a high pressure? In particular, can you tell me what pressure you use behind your nozzles?

VENDEL: We tested several nozzles with different numbers of orifices. For the presentation, the system was four orifices for the injection. The results of this study showed large differences between the number of orifices used.

MORRIS: We found that the principal factor is the critical pressure ratio through the nozzle.

VENDEL: The compressed air pressure used for four orifice holes was about 6 bar in this case.

MORRIS: Is that the pressure immediately behind the nozzle or is that the pressure in the reservoir, the main tank holding the compressed air?

VENDEL: I termed this device a laboratory device.

MORRIS: It is not.

VENDEL: It is not an in-situ device.

REAL TIME DETECTION OF A FLUORESCENT AEROSOL APPLICATION TO THE EFFICIENCY MEASUREMENTS OF HEPA FILTERS

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Abstract

This paper introduces a new technique for measuring mass concentration of a fluorescent aerosol. The study focuses, in particular, on the detection of a uranine (fluorescein sodium salt) aerosol, implemented during standardized efficiency tests of HEPA filters.

This aerosol exhibits the full range of characteristics researched for a test aerosol. Its fluorescent property gives it a specific nature with regard to its detection and measurement, by means of a fluorimetric method, offers the advantage of attaining a mass response with great sensitivity (some 10^{-10} g). Its physical properties: a solid, spherical aerosol, barely hygroscopic, with a mass median diameter of 0.15 μm and geometric standard deviation of 1.6 provide the possibility of carrying out the efficiency test for the maximum penetration range of HEPA filters.

The current implementation of the method is suffering from a delayed response in the measurement of the aerosol's mass concentrations upstream and downstream from the filter being tested. Whilst preserving the qualities of the test aerosol, the suggested technique allows measurements of these concentrations to be carried out in real time. It is based upon the principle of achieving different measurement sequences (sampling on a filter, dissolution of aerosol collected and measurement of the solution's fluorescence by means of a fluorometer) in one single phase.

The appliance operating at a flow rate of 3 l/min involves three sets of apparatus:

- an air-humidifier - condenser in which the solubilization of the particles of uranine into droplets, is achieved. These are around 2 μm in size and are obtained by condensation of glycerol vapour on each fluorescent particle,
- an impactor, composed of an accelerating nozzle set facing a collecting surface, assures the collection of these droplets with optimum output,
- an optoelectronic system intended on the one hand to select the excitation wavelenght of fluorescein, in order to illuminate the droplets deposited on the collecting surface and, on the other, to gather and process the signal of fluorescence obtained.

The detailed description and qualification of each stage will be presented. Test results will be carried out on the operation of the appliance integrating the three stages.

Introduction

The measurement of the collection efficiency of High Efficiency Particulate Air (HEPA) filters or the highlighting of transfers of airborne contamination brought about by ventilation inside premises requires the use of special tracer aerosols.

In the case of standardized efficiency tests[1] on HEPA filters, the French tracer aerosol is soda fluorescein (uranine) which is solid, spherical and not very hygroscopic, with a mass median diameter ($\bar{d}p$) equal to 0.15 μm and a geometric standard deviation (σg) of 1.6. Its property of fluorescence permit measurement by a fluorometric method giving a response proportional to the mass concentration.

The application of this method requires a sequence of operations such as: sampling on a filter, putting the aerosol collected into a known volume of solution at PH 9 and measuring the response in terms of fluorescence by means of a fluorometer. This sequence of operations involves a delayed response in the determination of mass concentrations upstream and downstream from the element being tested. Moreover, this technique, although very sensitive, does not permit the detection of masses smaller that 10^{-10} g of a fluorescent matter deposited on the filter. In order to obtain a real time response with better sensitivity, whilst retaining the advantages of the test aerosol, a new detection system has been developed.

The composition of this apparatus and its qualifications are described below:

Measurement concept:

The fluorescence response being more sensitive in liquid form, it is necessary to convert the solid test aerosol into droplets in order to ensure the solubilization of the fluorescent matter. This first stage is carried out by condensation of a suitable vapour on the solid particles inside a particle growth system (PGS).

The second stage is the collection of the droplets formed in a geometrically well defined zone. At the exit from the PGS the average size of the drops being of the order of 2 μm, sampling is carried out by impaction on a glass collection plate.

The third stage is the analysis by fluorescence of the quantity of matter deposited. The deposited left on a transparent sheet is illuminated by a laser beam. The fluorescent light emitted is gathered and sent to a photoelectron multiplier, the signal from which is recorded, and is proportional to the mass of matter deposited.

This apparatus is the subject of a patent registered in France and Europe[2], all of the various stages are shown in diagram-form in figure 1.

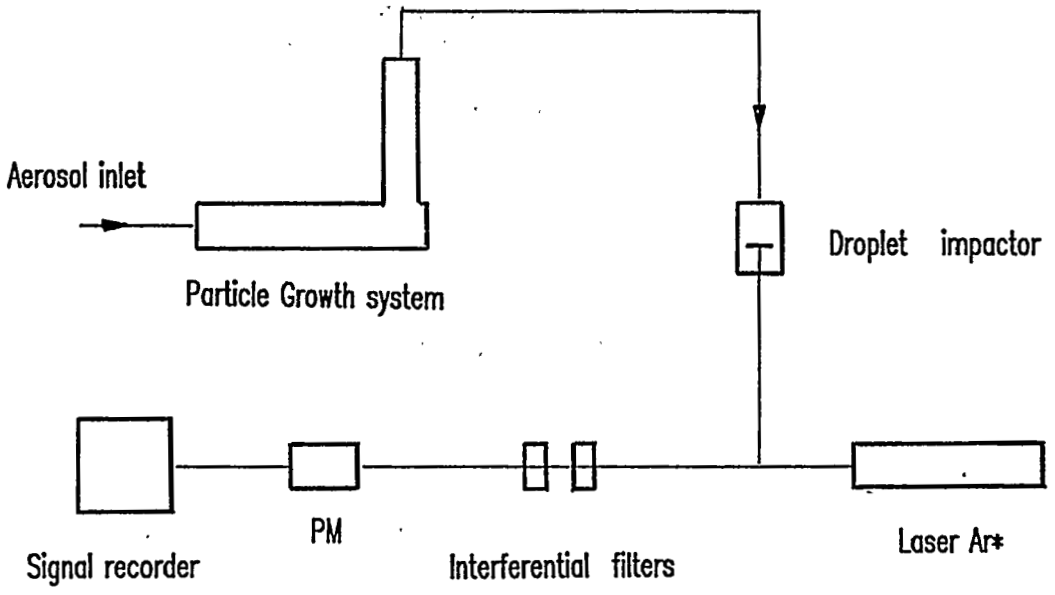

Figure 1 Schematic diagram of the measurement concept

The particle growth system

The role of the PGS is, on one hand, to increase the size of the test aerosol so that it may be collected by impaction and, on the other hand, to convert it from the solid state to the liquid state in order to ensure its detection by fluorescence.

The problem posed by detection of ultrafine particles has been resolved by a process which makes the particles enlarge in an atmosphere which is supersaturated with a vapour. This method, which is used in condensation nucleus counters (CNC) consists of circulating the primary particles inside a zone which is saturated with vapour (the saturator) then directing the flow towards a cool zone (the condenser) in order to ensure condensation of the vapour on the particles.

For the application described in this study, the liquid selected to produce the vapour is glycerol. Tests on putting uranine into solution in the glycerol showed that the solubilization limit is of the order of 2 g/l.

A theoretical study[3] led to the creation of a prototype (figure 2), of which the characteristics are as follows:

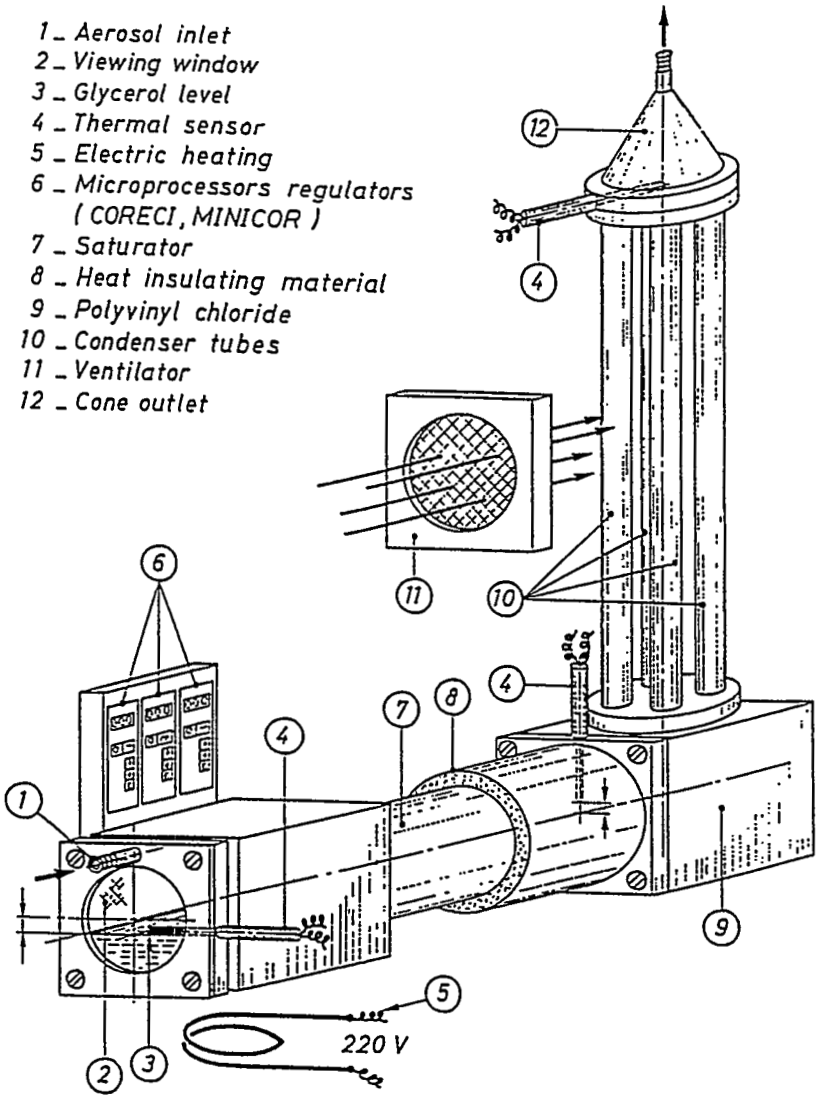

Figure 2 Schematic of the Particle Growth System (PGS)

Saturator

This is a cylindrical pipe of length 300 mm and diameter 80 mm made of duraluminium of thickness 10 mm. It has a draining orifice and a filling orifice. The inlet is a flat plate fitted with a sampling tube and a viewing window for checking the level of glycerol. At the begining the volume of glycerol is approximately 450 cm^3. Holes allow the insertion of thermal sensors for measurement of the temperature of the glycerol bath and of the fluid at the outlet of the saturator.

Thermal insulation between the saturator and condenser is made by means of polyvinyl chloride.

Heating of the liquid is ensured by an electric heating with a resistance placed at the bottom of the saturator next to the fluid entrance.

Condenser

The condenser is made of copper with four cylindrical pipes 400 mm in length and 20 mm in diameter and a cone is installed at the outlet. Inside the latter, hole allows the insertion of thermal sensor for measurement of the temperature of the fluid at the condenser outlet.

The condenser wall temperature is not adjusted; a ventilator simply accelerates the evacuation of heat released by the gas.

Airtightness betwen the different parts of the system is ensured by O-rings. Microprocessors regulators (Model Coreci, Minicor) of 1% precision are used to adjust the temperature of the liquid inside the saturator and to measure the temperature of the fluid at the outlets of the saturator and the condenser.

The sensors are platinum probes (100 Ω, 0°C). Note that regulation performed on the saturator liquid, is of the "constant temperature" type and not of the "constant energy flux" type. This means that the sampling rate could be varied while maintaining a constant saturator temperature.

The final size of the droplets produced from a uranine aerosol (dp = 0.15 μm), by the enlarging system operating at 11 l/min was determined by an aerodynamic particle analyser (APS 33 TSI) as a function of the temperature of the saturator. An example from the granulomectric spectra in number and mass of the droplets formed is represented in figure 3.

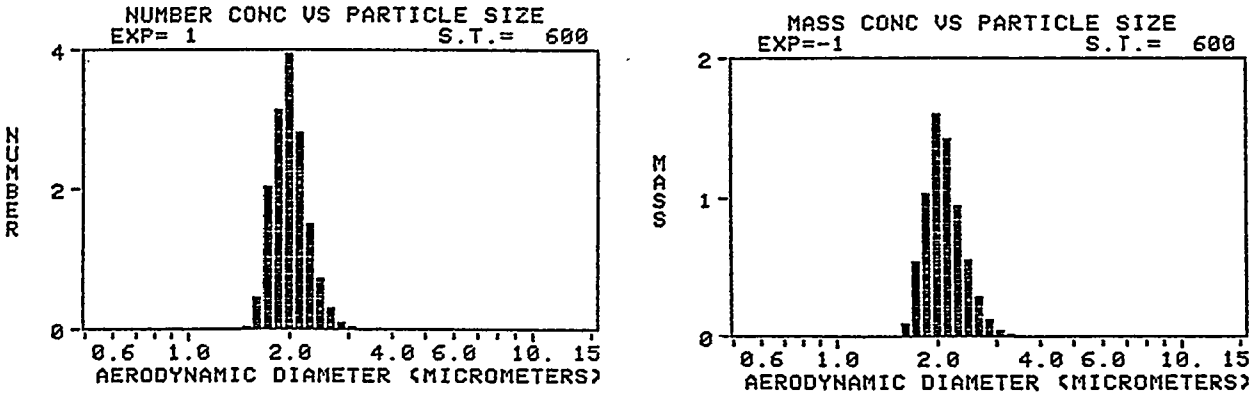

Figure 3 Droplets size distribution. Saturator temperature 77°C

The average diameter of the droplets (Dg) and the geometric standard deviation (Sg) of the distribution are calculated from the cumulative distributions established for various temperatures of the saturator (figure 4). Evaluation of the average diameter Dg as a function of temperature is represented in figure 5. The geometric standard deviation is independent of the temperature and remains below 1.15.

Under the operating conditions of the PGS, the tests as a whole enable the temperature of the saturator supplied with glycerol to be fixed at 77°C in order to obtain droplet diameters of 2 µm.

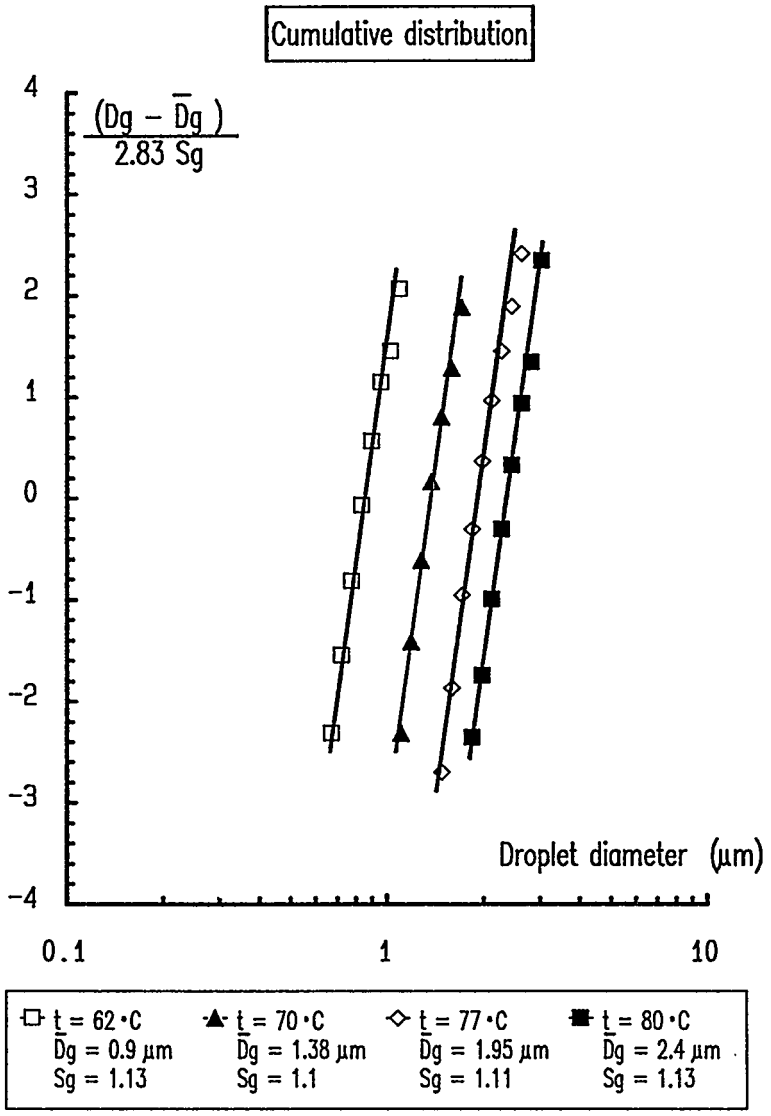

Figure 4 Droplets cumulative distribution

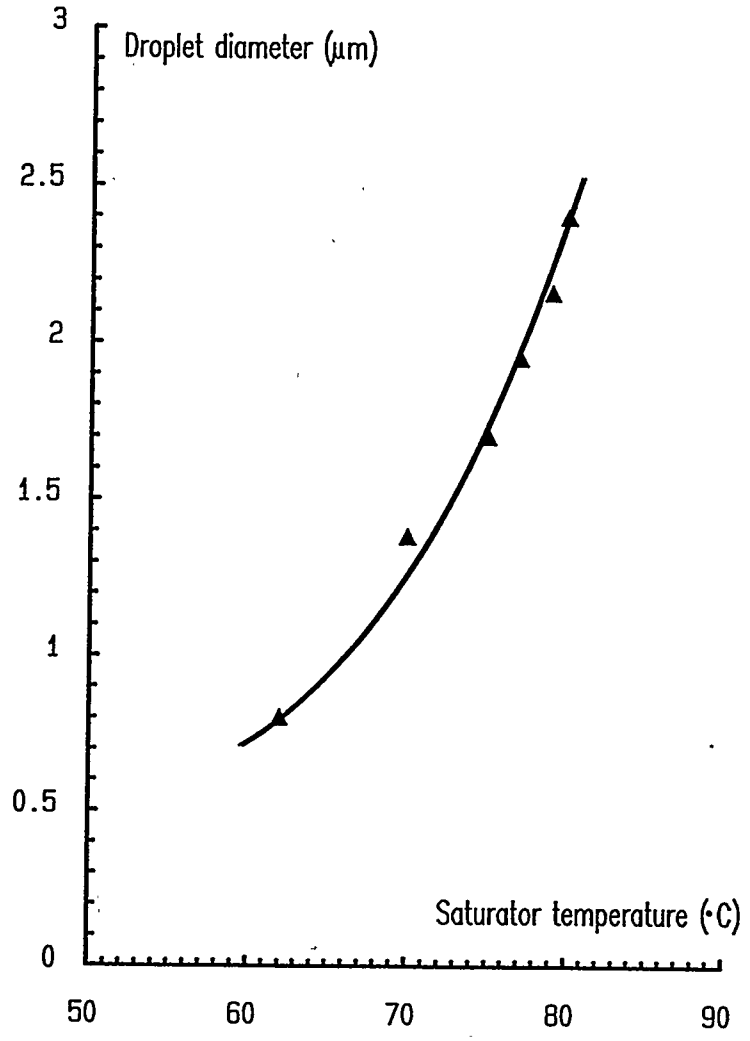

Figure 5 Droplet diameter VS saturator temperature

The impactor

The size of the droplets produced at the outlet from PGS (Dg = 2 µm, Sg ≈ 1.15) enables us to contemplate a system of collection by impaction. This technique means that the deposit of droplets can be localised in a zone of limited area. A single-nozzle impactor of circular geometry was made in accordance with the dimensioning laws put forward by Marple[4]. Calculation of the characteristics of the accelerator nozzle was carried out so that the particles of a diameter greater that 1 µm would be collected with a 100% efficiency. This condition is achieved when the Stokes number (Stk) of the particle corresponds to relation (1).

$$\sqrt{Stk} = 0.7 \qquad (1)$$

The Stokes number is defined by relation (2):

$$Stk = \frac{4\ \rho_p\ C\ Q\ dp^2}{9\ \mu\ D_b^{\ 3}\ \pi} \qquad (2)$$

where ρ_p = particle density (kg/m^3)

C = Cunningham's correction factor

dp = particle diameter (m)

Q = impactor flowrate (m^3/s)

μ = dynamic air viscosity (Pa.s)

D_b = diameter of accelerator nozzle (m)

The diameter of the accelerator nozzle is worked out from equations (1) and (2)

$$D_b = \left(\frac{4\ \rho_p\ C\ Q\ dp^2}{9\ \pi\ \mu\ (0.7)^2} \right)^{1/3} \qquad (3)$$

Hence, for operating conditions fixed at 3 l/min, an accelerator nozzle diameter equal to 0.105 cm is determined. This dimension satisfies Reynold's number criterion ($Re = 4.10^3$) which according to[4] should be around 3,000 in order to avoid impairing the collection characteristics of the impactor.

Collection of the droplets is carried out on glass plates (microscope cover-glasses). The distance e between the accelerator nozzle and the impaction base is given by relation (4):

$$\frac{e}{D_b} < 5 \qquad (4)$$

This distance was set at 5 mm.

Measurements of impaction efficiency were taken by means of uranine aerosol converted into droplets by the PGS. The efficiency is determined by measuring, with a fluorometer, the quantity of matter collected on the impaction plate and on a HEPA filter on the lower side of the impactor. The efficiency is defined by relation (5):

$$Re\ (\%) = \frac{Mp}{(Mp + Mf)} \cdot 100 \qquad (5)$$

where Mp and Mf are respectively the mass of fluorescent matter collected on the impaction plate and on the HEPA filter.

The results show, for glycerol droplets 2 μm diameter, a collection efficiency of the order of 90% when the impactor is operating at an output of 3 l/min.

Detection system

The detection system plays a dual role: firstly it illuminates the build-up of droplets by means of a light beam, the wavelength of which is fixed at the excitation wavelength of the soda fluorescein (λ = 488 nm); secondly it permits collection of the fluorescence light emitted, of which the intensity is proportional to the mass of fluorescent matter collected. For uranine the maximum emission wavelength is about 512 nm.

Excitation

Taking account of the excitation wavelength of uranine, illumination of the deposit is performed by means of an ionised argon laser, the wavelength of which is set at 488 nm. To achieve better illumination of the zone to be analysed, an axial optic 0° was rétained by completely separating the laser beam from the collection optic. Illumination of the deposit is achieved by a miniprism which reflects the laser beam at 90°.

Detection

In order to precisely determine the surface analysed by the detector, an image of the droplets is created by two objectives and defined by an iris diaphragm.

Two interferential filters placed in series carry out filtration of the laser exciting line. A transmission of 2.10^{-9} for the wavelength 488 nm is arrived at for this assembly.

The real image of the sample at the level of the iris is an achromatic doublet; the iris lying in the focal plane which is the object of this lens, the interferential filters are placed in a parallel beam which is directly focussed on the photoelectron multiplier by a final lens.

An overal diagram of the optical system is given in figure 6.

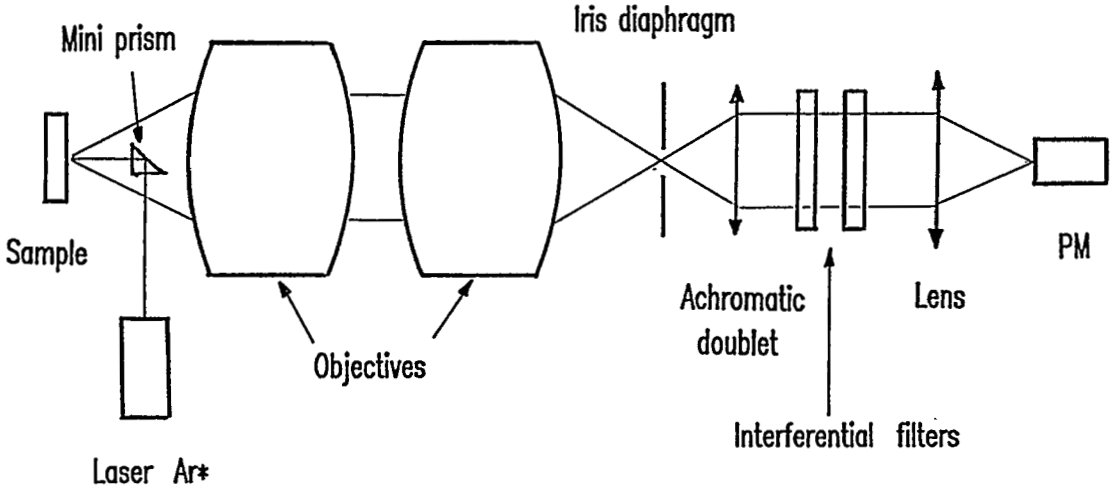

Figure 6 Schematic diagram of the optical system

Calibration of the measurement apparatus

An experimental study was carried out in order to calibrate the response of the photoelectron multiplier as a function of the mass of fluorescent matter collected on the impaction plate. Calibration was effected, at a laser power of 2.1 mW and a voltage of 700 V applied to the photoelectron multiplier, by means of the experimental set-up shown diagramatically in figure 7.

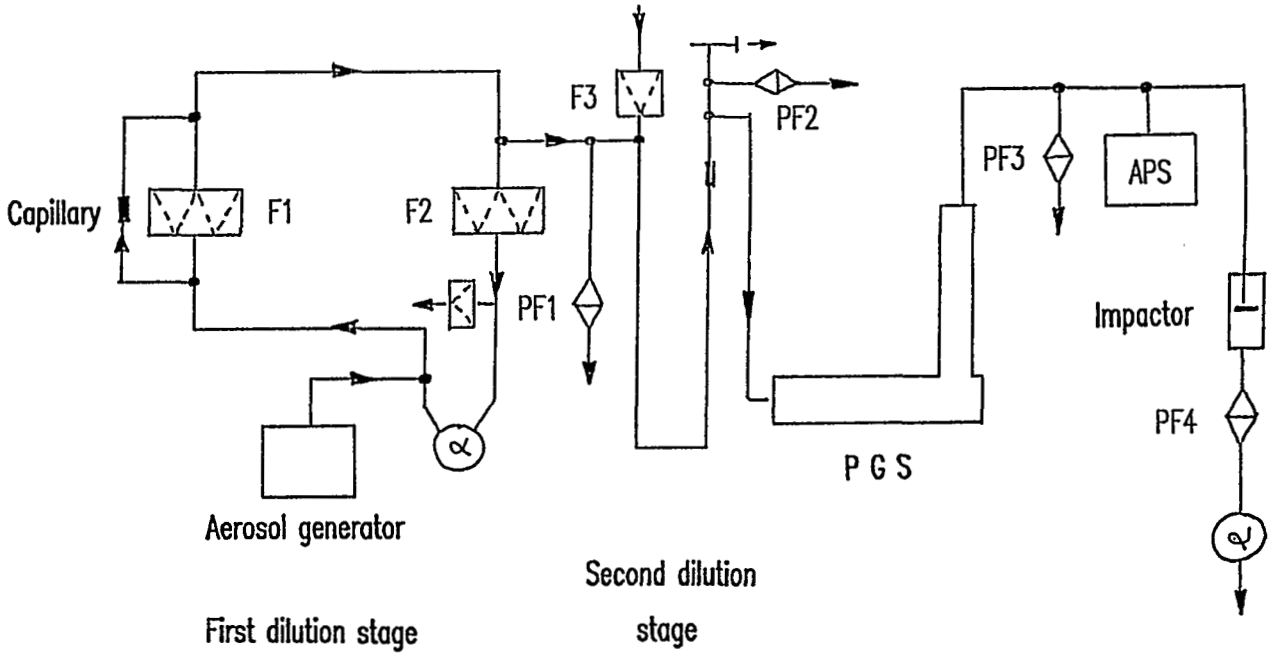

Figure 7 Experimental set up

It comprises two successive stages of dilution supplied by an aerosol generator delivering an output of 30 mg/h of soda fluorescein. The mass concentration in the second stage of dilution is determined by a traditional fluorimetric method by means of PF_2 sampling. A sampling effected at 11 l/min is sent to the PGS, the droplets formed at the outlet are taken off at the rate of 3 l/min on the impactor. The mass deposited on the impaction plate is determined by the operating efficiency of the impactor and by the time when sampling takes place. The masses deposited varied from $1.5\ 10^{-13}$ g to 5.10^{-10} g and the response from the photoelectron multiplier was recorded for each mass deposited.

The overall result are given in figure 8, the y-axis represents the response reading of the photoelectron multiplier and the x-axis the mass of matter collected on the plate.

The detection limit of the traditional fluorimetric method (Eppendorf fluorometer) and the background noise induced by the collection plates used, are also given in this figure.

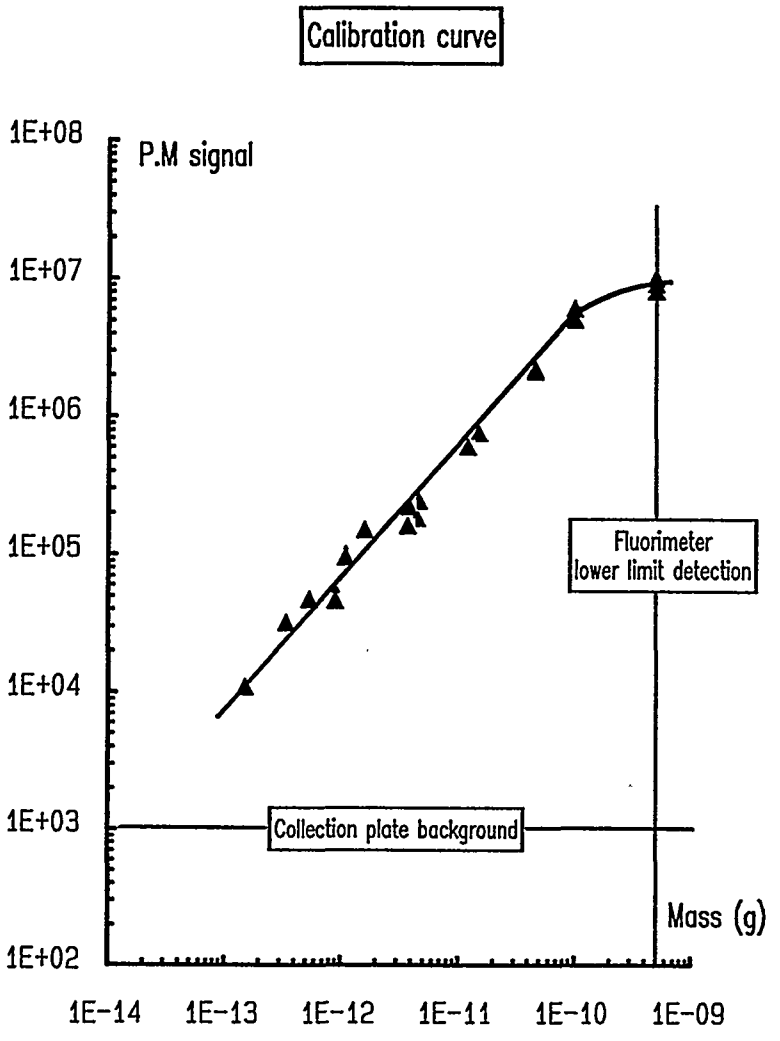

Figure 8 Calibration curve

Application for measuring the decontamination factor of a HEPA filter

Determining the decontamination factor (DF) of a HEPA filter requires measurements of the weight concentration of the test aerosol upstream and downstream from the filter.

The test was conducted on a circuit (see figure 9) handling 3,400 m^3 per hour, supplied via a soda fluorescein generator providing 30 mg/hr at a flow rate of 10 m^3/hr.

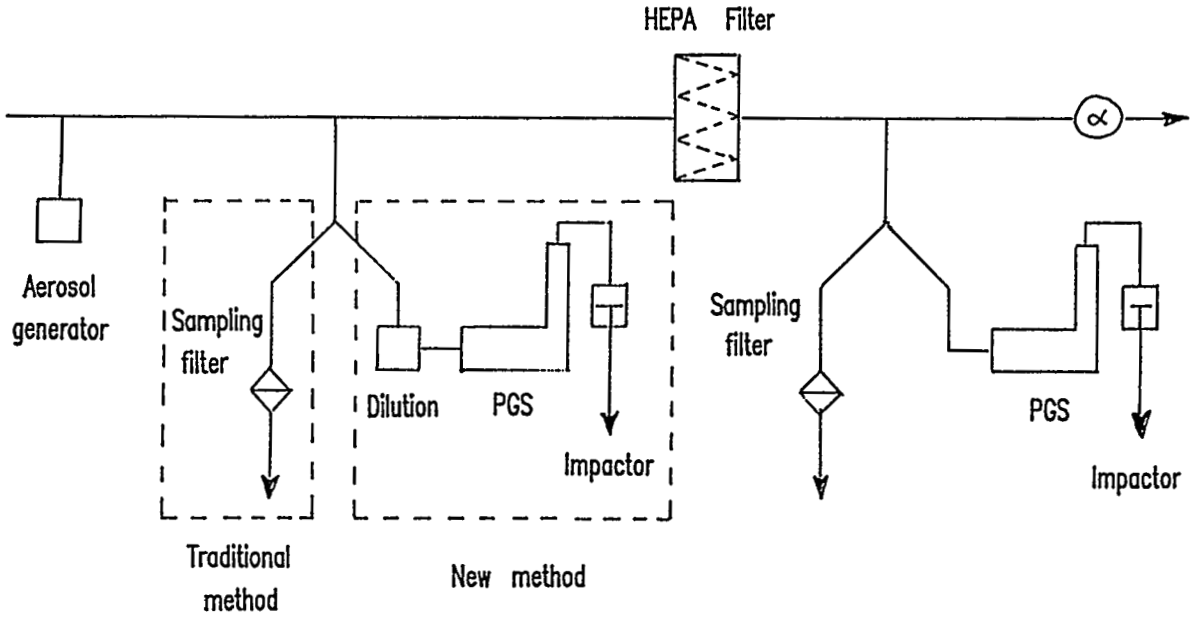

Figure 9 Experimental set up for testing HEPA filter

Table 1 indicates the weight concentration values measured using both the traditional method and the new proposed method.

Table 1 Comparison between traditional and new method

	Traditional method (TM)	New method (NM)	V(TM)/V(NM)	C(TM)/C(NM)
Upstream concentration (g/m^3)	$7.7\ 10^{-6}$	10^{-6}		7.7
Volume sampled (1)	500	3	166	
Downstream concentration (g/m^3)	3.10^{-10}	$1.1\ 10^{-10}$		2.7
Volume sampled (1)	5,000	2	2,500	

The comparison between the concentration measured using the two method shows a large difference between the results, of approximately a factor of 8 and 3 respectively for upstream and downstream concentrations.

The difference observed is explained by the fact that the illumination of the deposit is not obtained for the entire sample, the answer therefore is a function of the number of droplets illuminated.

However, considering the high sensitivity of the new method, the results in the table 1 show that sampling times can be considerably reduced. For downstream concentrations, the sampling times required for the traditional method are approximately 30 to 60 minutes, whereas for the new method typical time is approximately one minute.

The results of this test indicate that the application of the new method provides a much quicker response than the traditional method, but that the response suffers yet too much difference from the standard one to reliably determine the HEPA filter decontamination factor.

Conclusion

The traditional method of evaluating the efficiency of THE filters suffers currently from a delayed response, taking account of the process of evaluating the matter taken off. This method offers the advantage of being carried out with an aerosol which possessed the qualities required for a strict test. The new technique of measuring the mass of fluorescent material, used during these tests, offers the advantage of retaining the traditional test aerosol and combining the various stages (sampling, putting into solution and analysis) into one single mechanism.

Using the Particle Groth System has shown that it is possible to both enlarge the uranine particles from 0.15 μm to 2 μm and solubilize the fluorescent matter in these glycerol droplets. The impaction system permits collection of the droplets so formed, in a zone of limited dimensions, with a satisfactory efficiency. As for the detection mechanism, the combined use of an ionised argon laser beam and a separated collection optical unit permits detection of masses of the order of 10^{-13} g (around 3 orders of magnitude below the present limit) and delivering a linear response as a function of the mass deposited up to 10^{-10} g.

Research is underway to enlarge the accuracy of this method and to integrate all parts of this apparatus (PGS, impactor and detector) to make it operational on site on a routine basis.

Acknowledgements

The part of this study concerning fluorescence detection was carried out in co-operation with the instrumentation and spectrometry Group of the University of Lille, under Contract SA 9008/BD, our thanks go to Professor DELHAYE, Mr. DA SILVA and the DILOR Company.

References

(1) Ventilation separator. Method of measurement of the efficiency of filters by means of a uranine aerosol (soda fluorescein).
Approved French Strandard NF.X 44011, May 1972

(2) Real time measurement apparatus for contents of gas in aerosol form.
Patent n° 87 029 34, March 1987

(3) M. ASSA, D. BOULAUD and Y. METAYER
"Ultra fine particle detection by vapour condensation" in Aerosols Formation and Reactivity P. 837
Pergamon Press Oxford 1986

(4) MARPLE
Fine particle
ed. B.Y.H. LIU, 1976, Academic Press

DISCUSSION

GUEST: Has the system been used on a power station system or is it still under laboratory testing? Do you think it will be rugged enough for power station use?

VENDEL: No, it is only under laboratory testing. At this time, problems remain on the computation system. We have to integrate all the ports to make a rugged enough system for power installation.

CONCENTRATIONS PRODUCED BY A LASKIN NOZZLE GENERATOR A COMPARISON OF SUBSTITUTE MATERIALS AND DOP

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ABSTRACT

DOP, dioctylphthalate [or di (2-ethylhexyl) phthalate (DEHP)], commonly used as a plasticizer, has been used to produce test aerosols for in-place filter testing of HEPA filters and for quantitative respirator fit testing for over 30 years. Some questions arose about 10 years ago regarding the safety and potential health hazards of this material. Therefore, over the years many substitute materials for test aerosols have come into use in the field.

A study was undertaken recently at the Harvard School of Public Health[1] evaluating particle size distribution of the substitute materials using a Laskin[2] nozzle aerosol generator.

In concurrent preliminary studies performed at Air Techniques it was determined that the substitute liquids do not give the same concentration levels as DOP. This study will report on the various DOP alternatives and the concentrations of aerosols produced in the Laskin nozzle type aerosol generator in comparison with the concentration normally produced using DOP. Substitutes include di (2-ethylhexyl) sebacate (DOS), mineral oil, Ondina EL, Kaydol, corn oil, Emery 3004 and polyethylene glycol.

INTRODUCTION

In the past few years, there have been many substitutes for DOP used for testing in various air filtration fields. These substitute liquids are used in a Laskin nozzle type aerosol generator for producing an aerosol to challenge the integrity of high efficiency particulate air (HEPA) systems, and for quantitative respirator fit testing. Very recently, the European community has begun to use Ondina EL oil as a substitute for DOP. Ondina EL is available in Europe, through the Shell Oil Company, but is not available in the United States. Shell Oil of America recommends Kaydol, a brand name of the Whitco Chemical Corporation[3] of New York as a substitute. Also in Europe, paraffin oils, such as ENERPAR M, available from BP Oiltech GmbH[4] in Hamburg, Germany, are being used. It is not available in the U.S. In recent studies done by the Army Chemical Center, another new substitute has been found. This substitute is Emery 3004, which is available from the Henkle Chemical Corporation[5] located in Cincinnati, Ohio. General Properties of DOP and these substitute materials are given on a subsequent page. (See Table 1)

Air Techniques is a manufacturer of Laskin nozzle type aerosol generators and has many customers worldwide using these generators. Over the years we have received many inquiries from customers regarding two main subjects. The first question is, "If the liquid level varies, does it have any effect upon the output of the Laskin nozzle generator?" The second question is, "What is the Laskin nozzle generator output when using substitutes for DOP?" Thus the practical reasons for this study.

This study is prepared in two parts. The first part looks at the concentration output of a TDA-4A Laskin nozzle aerosol generator while varying the liquid level above the nozzle collar. The level was measured from the top of the nozzle collar to the top of the liquid. The generator output was measured both gravimetrically and photometrically using an Air Techniques model TDA-2E Aerosol Photometer. The TDA-2E Photometer was calibrated using a one hundred microgram per liter concentration of polydispersed DOP aerosol from a Laskin nozzle generator.

In the second part of the study, the same TDA-4A Aerosol Generator with a Laskin nozzle was employed. The aerosol generator was tested using DOP and various other substitute liquids. A one-inch liquid level was maintained above the collar on the Laskin nozzle. The outputs were measured both gravimetrically and photometrically. The photometric result provided instantaneous concentration indications to compare with the gravimetric result for each of the substitute liquids.

PROCEDURES

In accordance with the Echols and Young report, the output of a type IIIA Laskin nozzle generator with 20 pounds per square inch gauge pressure applied, will deliver 5.1 milligrams per liter of aerosol concentration. If this output is diluted with 135 cubic feet per minute of air, a concentration of one hundred micrograms per liter will be obtained. This is the basic standard that is used universally in the air filtration industry. Therefore, all test data is based on a TDA-4A Aerosol Generator utilizing a type IIIA Laskin nozzle generator operating at 20 psig.

For this study, a test rig was fabricated (See Figure 1) similar to the type used by the Harvard School of Public Health and was adjusted to supply 135 cubic feet of air per minute. The test rig was fitted with a Stairmand disk to ensure thorough mixing of the air and aerosol prior to the sampling port. A calibrated orifice plate manufactured by Daniel Flow Products Incorporated[6] in conjunction with a Dwyer[7] inclined manometer, was used to measure the air flow. A blast gate valve, located before the inlet of the blower, was used to control the air flow. Pressure to the aerosol generator was verified using a U-Tube mercury manometer (40.72 inches/Hg). The gravimetric concentration flow was measured with a Dwyer 0-100 standard cubic foot per hour flowmeter. The flowmeter operating head was measured with a Dwyer U-Tube manometer and the flow was corrected to 14.7 psia. The aerosol samples were collected in Gelman Sciences[8] extra-thick, glass-fiber, 47 millimeter filters (#66078) contained in a Gelman Sciences 47 millimeter gravimetric sample holder. A Gast[9] 1531 pump provided the sample flow. Each gravimetric sample was run for a ten minute duration at an average flow rate of approximately 1 cfm to assure sufficient pick-up for accurate results.

For the second part of the study, the same aerosol test apparatus and aerosol generator were used. The TDA-4A aerosol generator was tested first with DOP and then subsequently using the various substitute liquids. The liquid level was maintained at one inch above the top of the Laskin nozzle collar. Each time the aerosol generator was refilled with an alternate substitute liquid, it was first drained and flushed with denatured alcohol, and then cleaned. The aerosol generator's pressure was adjusted to 20 psig, and then a reading was taken with the TDA-2E Photometer. This aerosol photometer was calibrated against a 100 microgram per liter concentration of DOP aerosol. We felt it would be interesting to see if the photometer's response was the same for DOP as it was for substitutes for DOP. The gravimetric samples were taken in the same manner as the first part of the study.

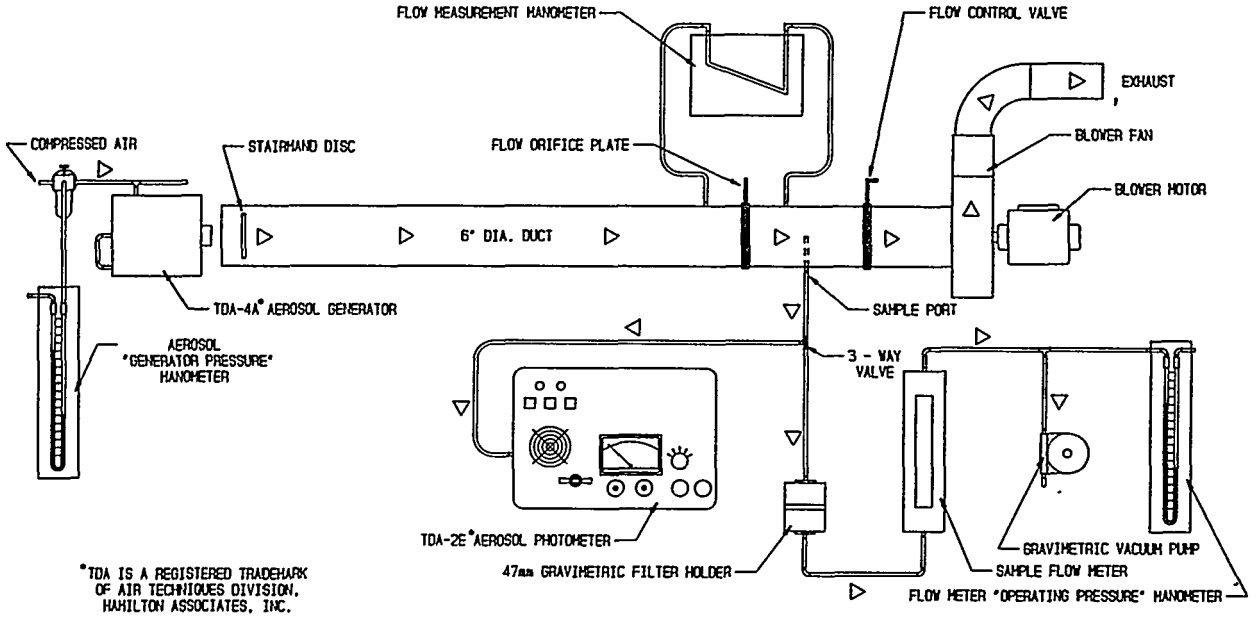
TABLE 1

Some Properties of DOP and Substitute Materials

Material	Specific Gravity	Viscosity (CP)	Boiling Point (°C)	Refractive Index
Di (2-ethylhexyl) phthlate (DOP)	0.983	82 @ 20°C	350	1.485
Di (2-ethylhexyl) sebacate (DOS)	0.915	17.4 @ 25°C	240	1.448
Mineral Oil, Heavy	0.845 - 0.905	>125 @ 20°C	360	1.471
Ondina (EL), Extra Light	.853	37 @ 20°C	—	1.467
Kaydol	<1.0 @ 15°C	67 @ 40°C	230	1.4787
Corn Oil	.918	(A)	(B)	1.464
Emery 3004	.819	16.9 @ 40°C	401	1.4556
Polyethylene Glycol PEG 400	1.128	105 @ 25°C	(B)	1.465
Paraffin Oil (BP ENERPAR M-002)	.848 @ 15°C	14.8 @ 40°C	301	1.466

(A) A natural product with varying properties. (B) Boils over a broad temperature range.

FIGURE 1 - SCHEMATIC DIAGRAM OF AEROSOL TEST APPARATUS



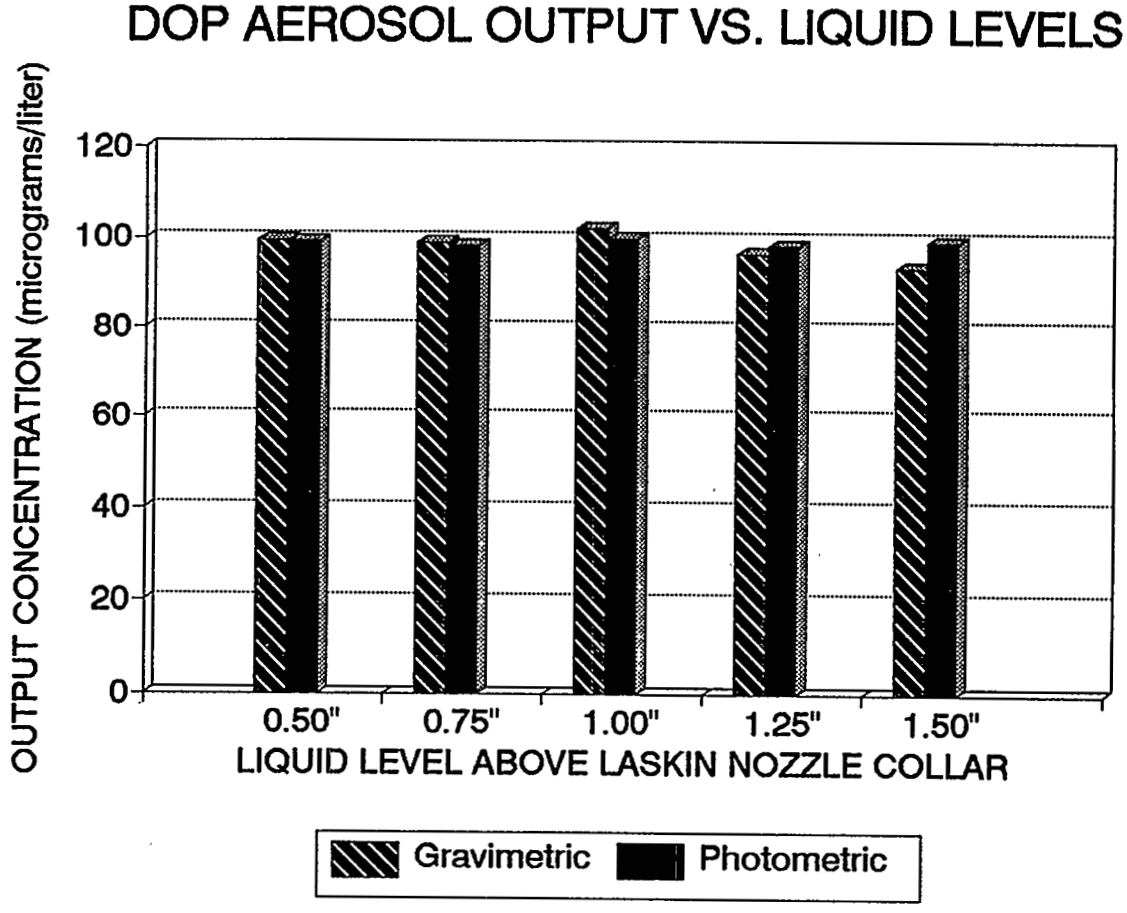
DATA

The results of the tests performed in the first part of the study are displayed both in table form and graphically. Table 2 gives the actual data and Figure 2 graphically displays the results. The bar graph shows both the differences in the liquid levels and the difference in the instantaneous photometric indications versus the actual gravimetric concentrations measured.

TABLE 2

Generator Liquid Level	Gravimetric Concentration (micrograms/liter)	Photometric Concentration Reading (%)
0.50 inch	99.3	99
0.75 inch	98.6	98
1.00 inch	102.0	100
1.25 inch	96.3	98
1.50 inch	93.2	99

FIGURE 2



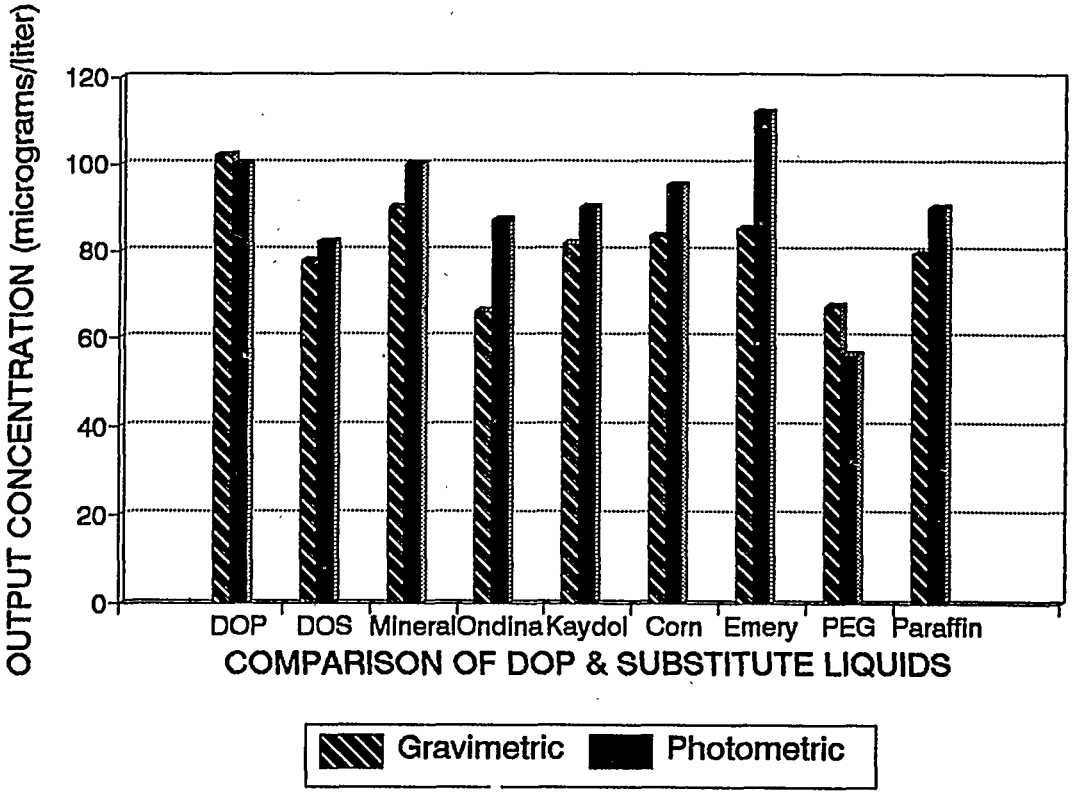
For the second part of the study, the data is again displayed both in table form and graphically. Table 3 displays the actual data and Figure 3 graphically displays the results. The bar graph shows the concentrations of the various substitute liquids and the instantaneous photometric indications versus the actual gravimetric measurements.

TABLE 3

Generator Liquid Level	Gravimetric Concentration (micrograms/liter)	Photometric Concentration Reading (%)
DOP	102.0	100
DOS	77.7	82
Mineral Oil	90.0	100
Ondina EL	66.0	87
Kaydol	81.6	90
Corn Oil	83.3	95
Emery	85.0	112
Peg 400	67.1	56
Paraffin Oil	79.4	90

FIGURE 3

AEROSOL OUTPUT-DOP & SUBSTITUTE LIQUIDS



CONCLUSIONS

It is apparent from the data developed in the first part of this paper that the liquid level does not significantly affect the concentration output of the generator. Both the photometer instantaneous indications and the gravimetric concentration readings show that there is very little change in concentration output from the generator, even though the liquid level is varied above the Laskin nozzle. This data also indicates that there were only some minor differences between the actual gravimetric reading and the instantaneous photometric indication. The photometer readings were taken for approximately 10 to 15 seconds. The readings were "eyeball averaged" due to minor fluctuations. The gravimetric results were run for ten minutes. The gravimetric result is simply a time weighted average of the actual photometer readings. We expected the photometric and gravimetric results to be close, though not identical, since we used the same liquid for each of the tests in the first part.

From the data developed in the second part of this paper it is apparent that the different substitute liquids do give concentrations that vary over a wide range. More importantly, photometric reaction to the various substitute liquids does not seem to correspond to their gravimetric concentrations. The photometer used for photometric measurement was calibrated against DOP aerosol. Aerosol photometers utilize the forward light scattering principle and the light scattered forward is a function of the aerosol diameter. Therefore, the aerosol of these substitute liquids may possibly have some differences in aerosol mass median aerodynamic diameter. Then, the aerosol photometer readings would not track with the actual gravimetric reading unless the photometer was calibrated initially against that particular aerosol. Regardless, when using a substitute liquid for DOP, the variation in concentration should be considered. Also, perhaps the photometer and generator should be calibrated together on a "mock test system" using the actual substitute liquid to insure accurate test results.

Many users of substitute liquids have encountered both operational and maintenance problems. Corn oil seems to be the most difficult substitute liquid to work with from a maintenance standpoint. Feedback from personnel using Laskin-type aerosol generators in the field indicate that the material tends to gel, and eventually turns to the consistency of RTV after it has cured. If the corn oil is allowed to get to this stage, the Laskin nozzle will be ruined and will have to be replaced. The best way to prevent problems and excessive maintenance on the aerosol generator, when using substitute liquids, is to drain them immediately after use. Then flush the nozzles and aerosol reservoir with a solvent.

REFERENCES

1 W.C. Hinds, J.M. Macher & M.W. First, "Size Distribution of Aerosols Produced by the Laskin Aerosol Generator Using Substitute Materials for DOP," American Industrial Hygiene Association Journal, J.44(7);495-500 (1983), pp. 495-500.

2 W.H. Echols & J.A. Young, "Studies of Portable Air-Operated Aerosol Generators," Report NRL-5929, July 26, 1963.

3 Whitco Chemical Corporation, Sonneborn Div., P.O. Box 336, Petrolia, PA 16050, TEL: 412-756-2210

4 BP Oiltech GmbH, Neuhoefer Brueckenstrasse 127-152, Hamburg 93, Germany, TEL: 49-40-751970

5 Henkel Chemical Corporation, Emery Div., 4900 Este Avenue, Cincinnati, OH 45232, TEL: 800-543-7370

6 Daniel Flow Products, Inc., 9720 Katy Road, Houston, TX 77224, TEL: 713-467-6000

7 Dwyer Instruments, Inc., P.O. Box 373, Michigan City, IN 46360, TEL: 219-879-8000

8 Gelman Sciences, Inc., 600 South Wagner Road, Ann Arbor, MI 48106, TEL: 313-665-0651

9 Gast Manufacturing Corporation, P.O. Box 97, Benton Harbor, MI 49022, TEL: 616-926-6171

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CHARACTERISTICS OF LASKIN NOZZLE GENERATED AEROSOLS

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Abstract

This study examines the aerosol concentration and light scattering intensity produced by Laskin nozzles at 20 psig with DOP and alternative liquids. Knowledge of both characteristics is critical when using an internal reference light set with a DOP aerosol at a concentration of 100 μg/L. This study supplements information on alternative liquids used in a prior study[1] and adds data on three additional liquids: (1) Shell Ondina Oil, extra light (EL), (2) a synthetic aliphatic hydrocarbon (Emery 3004), and (3) Kaydol White Mineral Oil. After each alternative liquid was aerosolized at 20 psig with Laskin nozzles into a controlled flow wind tunnel, (1) concentration was measured gravimetrically by sampling onto filter papers, (2) light scattering power was measured with a total aerosol light scattering photometer standardized with a fixed internal reference light and (3) particle size distribution was measured with a laser single particle counter and sizer. Data for each alternative liquid are compared with equivalent DOP characteristics and factors are derived to make possible accurate measurements with alternative liquids when using a photometer with an internal reference light calibrated with DOP.

I. Introduction

Routine in-place testing of high efficiency filter systems was initiated at Oak Ridge National Laboratory in mid-1962.[2] The testing instruments and methods developed at ORNL have continued in use to the present time in the nuclear industry with very little change and have been adopted for filter integrity testing in clean rooms, medical facilities, biological safety cabinets, and respirator quantitative fit tests. The method calls for a liquid test aerosol generated by an air-operated nebulizer, usually referred to as a Laskin nozzle,[3] and a total aerosol scattering photometer of the forward scattering type to measure filter penetration.[4] Until 1981, the aerosolized liquid aerosol in use was di-(2-ethylhexyl)phthalate usually referred to as DOP, an acronym for dioctylphthalate. In 1981 the National Toxicology Program published a technical report citing an association between exposure to 3,000 to 12,000 ppm DOP in the diet and the development of hepatocellular carcinomas or neoplastic nodules in mice and rats.[5] Although the toxicological findings were deemed somewhat speculative and the ingestion route of administration not especially relevant for inhalation exposures, the National Institute for Occupational Safety and Health designated DOP a "potential human carcinogen".[6] This stimulated an intensive search for innocuous substitutes that would give the same size distribution as DOP when aerosolized with Laskin nozzles. A number of acceptable alternative liquids were found, as shown in Table 1 from Hinds et al.,[1] and it was thought that the matter had been resolved satisfactorily. And so it had for all situations where it was possible to make an upstream aerosol reading to program the 100% photometer setting against which to measure the downstream penetration value.

A complication arises, however, when a photometer internal reference light that has been calibrated with a DOP aerosol at a concentration of 100 μg/L is used with an alternative liquid aerosol. In principle, an internal reference light makes it possible to adjust the photometer gain setting to the same position it would have had if the upstream aerosol concentration had been sampled directly by using calculations based on the number of Laskin nozzles in operation at 20 psig air pressure and the diluting air volume. The ability to substitute calculation for measurement of the upstream aerosol concentration avoids a need to physically enter a contaminated filter plenum or to withdraw a sample from contaminated duct or plenum, a safety measure in conformity with the ALARA principle (as low as reasonably achievable).

Table 1 Properties of DOP and Alternative Liquids

Material	Specific Gravity	Viscosity	Refractive Index
MATERIALS USED IN 1980 (Ref. 1)			
Di(2-ethylhexyl)phthalate Hatcol DOP	0.983	82 cps @ 20°C	1.485
Di(2-ethylhexyl)sebacate Uniflex DOS	0.915	17.4 cps @ 25°C	1.448
Mineral Oil Arcoprime 200	0.8608	52.4 cps @ 100°F	1.471
Corn Oil Fisher USP	0.918	-	1.464
Polyethylene Glycol Union Carbide PEG-400	1.127	105 cps	1.455
MATERIALS USED IN THIS STUDY			
Di (2-ethylhexyl)phthalate Eastman Kodak (DOP)	0.983	82 cps	1.485
Ondina Oil, EL Food Quality Mineral Oil, Shell	0.869	14.3 cps	1.467
Kaydol White Mineral Oil Whitco Mfg. Co.	0.877	67.1 cps	1.479
Emery 3004 Synthetic Aliphatic Hydrocarbon	0.819	-	1.456
Corn Oil Purity Supreme Supermarket	0.918	-	1.464
Polyethylene Glycol PEG-400 Union Carbide Corp.	1.127	105 cps	1.455

The uncertainty that arises when using DOP substitutes becomes apparent from a review of Table 1 where it can be seen that there are differences in viscosity and specific gravity that can affect the amount of aerosol generated by Laskin nozzles at a fixed air pressure plus differences in refractive index that can affect the light scattering function of equal aerosol mass concentrations and size distributions.

Recent revisions of established standards covering in-place filter testing have backed off from the original requirement for an upstream aerosol concentration of 100 μg/L. The 1989 revision of ASME N510, Testing of Nuclear Air Treatment Systems,[7] merely requires the photometer to be capable of detecting "penetration smaller than the maximum penetration allowed by the test program" and is silent on a specific upstream concentration requirement. NSF Standard Number 49, Class II (Laminar Flow)

Biohazard Cabinetry,[8] calls for an upstream aerosol concentration not less than 18 μg/L. For both standards, it is important to be certain the upstream concentration is at least 10^5 times greater than the least downstream concentration that can be measured with confidence. To resolve the matter of the upstream sample concentration in a field situation, an internal reference light calibrated for 100 μg/L DOP aerosol can be used as a standard to measure the concentration in μg/L of a sampled DOP aerosol. If, however, the upstream aerosol has been generated from an alternative liquid, the same uncertainties enumerated for using the internal reference light for adjusting the photometer gain setting by calculation apply to measuring the generated aerosol concentration.

To resolve all uncertainties regarding the equivalence of aerosols generated from alternative liquids to the standard DOP aerosol, a study program was developed to measure and compare aerosol generation rate, size distribution, and light scattering intensity under uniform aerosol generator operating conditions. All comparisons were made using standard Laskin nozzles operated at 20 psig air pressure and diluting the aerosol to a DOP equivalent of approximately 30 μg/L. Table 1 lists the characteristics of the liquids used in this study program.

II. Experimental Method

A. Equipment

The experimental apparatus consisted of the following:

1) Two commercial Laskin nozzle aerosol generators of identical size and construction, TDA 4A Aerosol Generator.*
2) Two commercial total light scattering photometers equipped with an internal reference light calibrated to produce a reading of 100 on the 100% range scale when sampling a DOP aerosol concentration of 100 μg/L, TDA 2-EL Particulate Detection Unit.*
3) Duct sampling train consisting of a duct probe, a 50-mm filter-holding cassette equipped with ultra-high efficiency all-glass fiber filter papers, rotameter, pressure measuring manometer, flow regulating valve, and a sampling pump. The experimental arrangement of apparatus is shown in Figure 1.
4) Cahn electrobalance
5) ASAS-X laser spectrometer instrument for particle size measurements in the 0.09-0.3 μm range. Particle Measuring Systems, Inc., Boulder, CO.
6) A wind tunnel capable of handling 1,200 cfm and equipped with a flow control damper and a calibrated airflow measuring meter.

B. Procedures

Step 1 was to verify that the two aerosol generators delivered precisely the same aerosol light scattering intensity and mass concentration of DOP. The purpose was to keep one generator filled with DOP as a reference aerosol source while the other was filled in turn with each of the alternative liquids. Verification was based on (1) duct sampling for 10 min at 19 Lpm and weighing filters after temperature and humidity stabilization, before and after sampling (2) measuring light scattering intensity when a single Laskin nozzle generator at 20 psig was discharged into the same wind tunnel airflow at 450 cfm, and (3) measuring particle size distribution of the aerosol. The results of these comparisons are shown in Table 2. They indicate that the two generators gave aerosol characteristics that were virtually identical when operated alternately under experimental conditions as closely the same as possible.

Step 2 was to compare the aerosol characteristics of each of the alternative liquids to DOP on the basis of concentration, light scattering intensity, and particle size distribution when generated at 20 psig and diluted to the DOP equivalent of approximately 30 μg/L. Simultaneous measurements were made with the reserved DOP generator for comparison purposes.

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* ATI, 1801 Whitehead Rd., Baltimore, MD 21207.

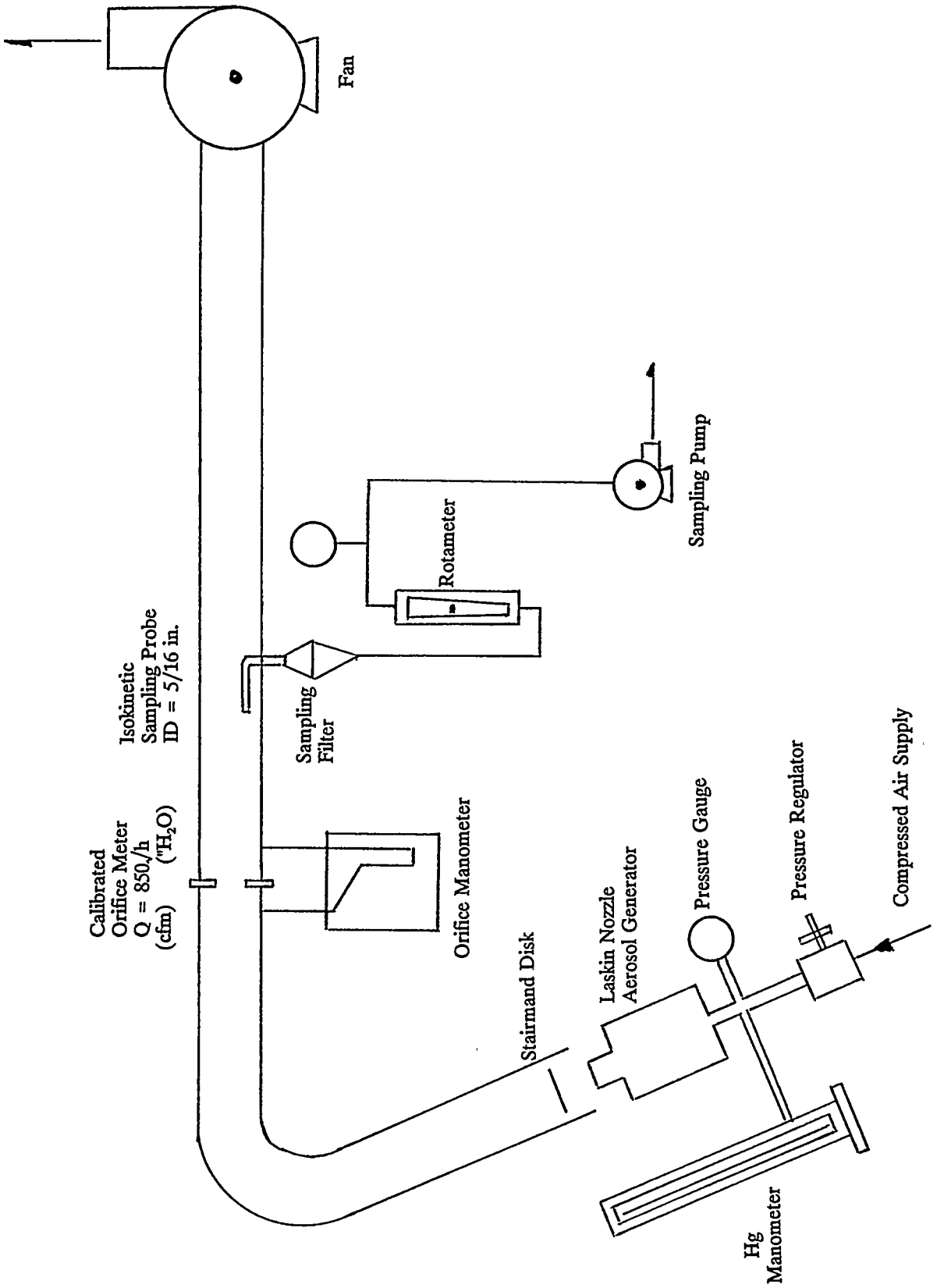

Figure 1 Apparatus for Aerosol Concentration Sampling

Table 2 Characteristics of Two Laskin Nozzle Aerosol Generators with DOP (Dilution Airflow 450 cfm: Nozzle Pressure 20 psig)

Generator Number	Mass Concentration* (ug/L)	Light Scattering Reading**	Count Median Diameter	Geometric Standard Deviation***
#1	29.6 ± 1%	30	0.28	1.54
#2	29.6 ± 1%	30	0.29	1.52

III. Experimental Results

A. Aerosol Concentration

With one of the two identical aerosol generators filled with DOP and the other one successively filled with each of the selected alternative liquids, five replicate measurements were made of each during a single day. In all, 25 replicate DOP measurements were made during 5 different days. They are shown in Table 3. The average daily DOP concentration produced by one Laskin nozzle discharged into a 450 cfm airstream ranged from 27.9 to 29.6 μg/L. The concentration measurements shown in Table 2 are within this range. The average of the 25 samples in Table 3 is 29 μg/L. Data published by Echols and Young in 1963[3] and verified many times since shows that one Laskin nozzle operated with air pressure at 20 psig delivers 75 Lpm of DOP aerosol containing 5.1 μg/L. When diluted to 450 cfm in air, one Laskin nozzle should, therefore, produce a calculated DOP concentration of 30 μg/L. The measured values, derived from air sampling and gravimetric analysis, were considered to be acceptably close to the calculated value.

It may be seen in Table 3 that tests for each alternative liquid were conducted with a simultaneous set of DOP tests and that there were small differences in the daily average DOP concentrations measured simultaneously. The differences are believed to reflect small experimental variations in the aerosol generator compressed air pressure setting, the wind tunnel dilution air flow setting, the aerosol sampling rate setting, plus weighing variations caused by small differences in temperature and humidity equilibration. Because the magnitude and direction of these irreducible experimental variations were likely to be the same for all measurements conducted on the same day, it was considered more accurate to compare the results for each alternative fluid with the corresponding simultaneous DOP results rather than to use the average for all DOP measurement relationships and these values are shown in Table 3.

The average concentration of corn oil and Emery 3004 most closely matched that of DOP. They gave concentration ratios to DOP of 0.98 and 0.97, respectively. Ondina oil and Kaydol oil were alike, delivering 0.85 and 0.87 as much aerosol as DOP. PEG 400 delivered only 0.76 as much aerosol as DOP on a mass basis, a serious shortfall.

B. Light Scattering Intensity

The light scattering intensity of the aerosols generated for concentration measurements (Table 3) was read simultaneously by adjusting the photometer gain setting with the aid of the internal reference light to a reading on the 100% range scale exactly equal to a DOP aerosol concentration in μg/L. As was expected, all DOP aerosol readings were at or close to 30 on the photometer

* with 95% confidence limits based on 10 samples for each generator.

** Photometer gain set to read 100 for a DOP concentration of 100 μg/L.

*** Calculated from Equation in text.

Table 3 Mass Concentration and Light Scattering Intensity of DOP and Alternative Liquids
(Dilution Airflow: 450 cfm, Nozzle Pressure: 20 psig)

Date	Liquids	Mass Concentration* (μg/L)	Mass Ratio (Alternative/DOP)	Light Scattering Reading** (100% Position)	Photom. Ratio (Alternative/DOP)	Photometer Reading/μg/L Aerosol	Correction Factor
7/3	PEG-400	22.6 ± 3%		22.2		0.98	0.97
	DOP	29.6 ± 0%	0.76	30.0	0.74	1.01	
7/5	Ondina	25.0 ± 1%		32.7		1.31	1.28
	DOP	29.3 ± 1%	0.85	30.0	1.09	1.02	
7/13	Kaydol	24.9 ± 4%		32.0		1.29	1.23
	DOP	28.7 ± 1%	0.87	29.9	1.07	1.04	
7/11	Emery	27.5 ± 1%		42.9		1.56	1.51
	DOP	28.5 ± 1%	0.96	29.4	1.46	1.03	
7/9	Corn Oil	27.4 ± 1%		36.7		1.34	1.26
	DOP	27.9 ± 1%	0.98	29.7	1.24	1.07	

* with 95% confidence limist based on 5 measurements for each liquid.

** Photometer gain set to read 100 for a DOP concentration of 100 μg/L.

scale. Using exactly the same photometer gain setting, the 5 alternative liquids gave the light scattering intensities shown in Table 3. It was expected that light scattering intensity would be influenced by the aerosol concentration generated by the method described earlier and, indeed, the 22.2 scattered light intensity reading for PEG-400 (equivalent to 22.2 μg/L had it been DOP aerosol) was almost identical to the 22.6 μg/L gravimetric measurement. The other four alternative liquids, by contrast, produced much greater scattered light intensity readings than did DOP. The readings could not be accounted for by the aerosol concentrations that were measured gravimetrically (Table 3). Therefore, it is clear that the internal reference light, calibrated with known concentrations of DOP aerosol, does not give accurate concentration readings with these four alternative liquids. Ondina and Kaydol oils gave light scattering readings 9 and 7% greater than DOP in spite of having a 15 and 13% lower aerosol concentration, respectively. Corn oil gave 24% higher and Emery gave 46% higher scattered light intensity reading for only slightly lower aerosol weight concentrations than DOP.

C. Aerosol Size Particle Distribution

The scattered light intensity of aerosol clouds is influenced by particle size, aerosol concentration, and refractive index of the liquid. With equal mass concentration, smaller particle sizes should give increased light scattering intensity of the cloud. The particle size distributions of DOP and each of the alternative liquids tested are shown in Table 4.

The count median diameter, CMD, is the 50% size. The geometric mean diameter, GMD, and the geometric standard deviation, GSD, were calculated from the following formulas:

$$\text{For GMD: } d_g = \exp\left[\frac{\Sigma n_i \ln d_i}{\Sigma N_i}\right]$$

$$\text{For GSD: } \sigma_g = \exp\left[\frac{\Sigma n_i(\ln d_i - \ln d_g)^2}{\Sigma N_i - 1}\right]^{1/2}$$

where: d = particle diameter
n = number
σ = standard deviation
g = geometric
i = i^{th} diameter size category
$N = \Sigma n_i$

The average count mean diameter, CMD, held steady at 0.25 μm for each of the five DOP data sets in Table 4, as did the geometric standard deviation, GSD, at 1.62. When the DOP GSDs were recalculated as the ratio of the 84% size divided by the 50% size (following the method used in Ref. 1), the values ranged from 1.52 to 1.60. Each of the alternative liquids gave slightly different CMDs (0.24-0.30 μm vs 0.25) and GSDs (1.50-1.63 vs 1.62). Even though PEG-400 gave higher scattered light intensities for lower mass concentration than DOP, it showed a somewhat higher CMD. The other liquids matched the DOP CMD very well but had somewhat smaller GSDs.

Table 4 Particle Size Parameters for DOE and Alternative Liquids
(Nozzle Pressure: 20 psig)

Date	Liquids	Particle Size Parameters				
		CMD (μm)	GMD (μm)	GSD	D(84%)/D(50%)	MMD (μm)
7/24/90	PEG-400	0.30	0.31	1.63	1.53	0.80
	DOP	0.25	0.26	1.62	1.52	0.69
7/24/90	Ondina Oil	0.24	0.25	1.51	1.46	0.55
	DOP	0.25	0.26	1.62	1.56	0.69
7/25/90	Kaydol	0.26	0.27	1.57	1.46	0.78
	DOP	0.25	0.26	1.62	1.52	0.69
7/25/90	Emery	0.25	0.26	1.51	1.44	0.58
	DOP	0.25	0.26	1.62	1.60	0.69
7/26/90	Corn Oil	0.24	0.25	1.50	1.46	0.58
	DOP	0.25	0.26	1.62	1.58	0.69
1980	DOP	0.25			1.53	
1980	Mineral Oil	0.27			1.52	
1980	Corn Oil	0.25			1.53	
1980	PEG-400	0.30			1.48	

NOTES:
1. Particle size measurements were made with aerosols containing 3,000-6,000 particles/cc.
2. GSDs were calculated from the distribution equation in the text. The other measure of dispersion given is the ratio D(84%)/D(50%) that is applicable for a distribution that gives a straight line on log-probability plotting paper. In our experiments, the distribution plots were not straight.
3. MMD is the mass median diameter calculated as the cumulative percent to the 50% point.

IV. Discussion

When using the internal reference light, it becomes possible to measure the correct aerosol concentration of each of the alternative liquids accurately by calculating a factor that relates light scattering intensity to concentration. The result is shown in Table 3 as light scattering intensity per μg/L of concentration after the photometer gain has been set to read 100 for 100 μg/L of DOP. The unit light scattering intensity ranges from 0.98 light scattering intensity units per μg/L of concentration for PEG-400 to 1.56 light scattering units per μg/L for the Emery product. To use this information correctly, first, the photometer gain must be set with the internal reference light to gave a reading of 100 for an aerosol concentration of 100 μg/L of DOP; second, the upstream aerosol photometer reading

must be taken; third, the alternative liquid aerosol concentration may be calculated by dividing the scattered light intensity reading by the factor given in Table 3. For example, if, after setting the photometer gain as noted above, the aerosol light scattering reading was 85 while using the Emery product, the aerosol concentration would be 85 ÷ 1.51, or 56 μg/L.

To set the correct gain into the photometer by calculation when using a liquid other than DOP: first, the aerosol concentration must be calculated as though DOP was being used, second, the aerosol concentration must be corrected from the mass ratio data in Table 3, third, the light scattering intensity must be modified by the correction factor in Table 3. If one were using Ondina oil EL, the aerosol concentration in μg/L would be only 0.85 times as high as it would have been had DOP been used. However, the light scattering intensity would be 1.28 times that of DOP per unit of aerosol concentration. This means that the calculated concentration for DOP must be multiplied by 0.85 for Ondina oil EL and the product multiplied by 1.28 to give the correct gain setting. For example, using two Laskin nozzles at 20 psig diluted in 450 cfm of air would give a calculated aerosol concentration of 60 μg/L were the liquid DOP, but for Ondina oil EL, the aerosol concentration would be 60 X 0.85, or 51 μg/L. Fifty one μg/L of Ondina oil EL gives 1.28 times the light scattering intensity of an equal mass concentration of DOP, or the same scattered light intensity as 51 x 1.28 = 65.3 μg/L of DOP. Therefore, the photometer gain should be set as though the aerosol contained 65.3 μg/L of DOP to give an upstream reading of 100% with an aerosol containing 51 μg/L of Ondina oil EL.

The aerosol particle size data obtained during the 1980 study[1] are shown in Table 4 with the size data collected during the present study. It may be seen that the CMDs and GSDs for DOP, corn oil, and PEG-400 were found to be closely the same in both studies. The mineral oil used in the 1980 study gave a CMD and GSD similar to those for Ondina Oil EL and Kaydol oil.

V. Conclusion

Many alternative liquids can be used as substitutes for DOP provided appropriate factors are used to correct light scattering intensity and concentration readings.

References

1. Hinds, W., Macher, J and First, M.W., Size Distribution of Aerosols Produced from Substitute Materials by the Laskin Cold DOP Aerosol Generators, in Proceedings of the 16th DOE Nuclear Air Cleaning Conference, M.W. First, Ed., NTIS, Springfield, VA, Feb. 1981, pp. 125-38.

2. Parrish, E.G. and Schneider, R.W., Review of Inspection and Testing of Installed High-Efficiency Particulate Air Filters at ORNL, in Treatment of Airborne Radioactive Wastes, Intern. Atomic Energy Agency, Vienna, 1986, pp.143-64.

3. Echols, W.H. and Young, J.A., Studies of Portable Air-Operated Aerosol Generators, NRL Report 5929, Naval Research Lab., Washington, DC, July 26, 1963.

4. Knudson, H.W. and White, L., Development of Smoke Penetration Meters, NRL Report P-2642, ibid, Sept. 14, 1945.

5. National Toxicology Program, NTP, Technical Report on the Carcinogenesis Bioassay of Di(2-ethylhexyl)Phthalate (CAS No. 117-81-7), Research Triangle Park, NC, 15 October 1980.

6. NIOSH, Occupational Safety and Health Guideline for Di-2-Ethyhexyl Phthalate (DEHP) Potential Human Carcinogen, U.S. Dept. Health and Human Services, 1988.

7. ASME, Testing of Nuclear Air Treatment Systems, ASME N510-1989, The Am. Soc. of Mech. Eng., New York City, 1989.

8. NSF, Standard 49 for Class II (Laminar FLow) Biohazard Cabinetry, National Sanitation Foundation, Ann Arbor, MI, 1987.

DISCUSSION

HANSON: What effect does the air pressure on the DOP generator have on the particle size generation? Does the various pressure affect the size of DOP?

FIRST: We have not made measurements at pressures other than at 20 psi.

HANSON: I was wondering if the number of nozzles used in the ATI generator has much effect on the size generated.

FIRST: The generator we used has a range of one to seven nozzles. There is a possibility that some agglomeration may occur at higher concentrations before the aerosol is diluted. It is very important to dilute the aerosol instantaneously as it comes out of the jet or the size spectrum will change significantly.

SAFE REPLACEMENT MATERIALS FOR DOP IN "HOT SMOKE" AEROSOL PENETROMETER MACHINES

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Abstract

For many decades dioctyl phthalate (DOP), a common industrial material, has been used by the U.S. Army and other agencies to simulate aerosol behavior in non-destructive gas mask and filter serviceability testing, and for related test purposes. Techniques are completely standardized. But DOP has been labeled a hazardous material. The research reported here, which was performed using ATI Q-127 and TDA-100 "hot smoke" aerosol penetrometer test machines, identified relatively innocuous, inexpensive replacement materials for DOP. One of these, a synthetic hydrocarbon (poly alpha olefin (PAO)), can be used to replace DOP directly with minimum impact upon existing hardware and procedures. Of more than two dozen candidate replacement materials that were tested, isostearic acid and oleic acid also performed well as DOP replacements. All three materials also show promise for cold smoke testing applications.

I. Introduction

Di (2-ethylhexyl) phthalate, also called dioctyl phthalate, di-sec octyl phthalate, DOP, or DEHP, is a widely used industrial material. Over ninety percent of the material produced is used as a plasticizer, primarily for PVC plastics. The properties of DOP that make it useful as a plasticizer, including low vapor pressure, chemical stability, and insolubility in water, also make it useful as a test aerosol. DOP aerosols are used in respirator fit testing, HEPA filter testing, aerosol research, aerosol instrument calibration, and other applications. These uses involve human occupational exposure to submicrometer-sized DOP aerosols, often briefly but in moderately high concentrations.[1]

Concern about the potential health effects to people working with DOP test aerosols has led to a search for substitute materials.[1] This search has taken a number of different directions, depending in part upon the specific test applications for which a DOP replacement has been sought. For example, Hinds, et al,[2] looked at size distributions of test aerosols of corn oil, di (2-ethylhexyl) sebacate (DOS), mineral oil, and polyethylene glycol (PEG), and compared these to DOP. Gerber[3] has published a detailed study of glycols as safe DOP replacements. Other comparative studies of size distribution and filter penetration of corn oil,[4] mineral oil, PEG, and DOP have been reported.[5] Interest has been revived in the use of solid aerosols including salts as test media,[6] and their performance compared to DOP aerosols.

The U.S. Army routinely performs 100% quality control testing of filter canisters manufactured for use with field-issue gas masks, and periodic sampling and testing of canisters stored in its supply depots. In April, 1986, the U.S. Army Surgeon General placed severe restrictions upon testing with DOP; agencies were also informed that dioctyl sebacate (DOS) would no longer be acceptable as a DOP replacement material, and that similar restrictions would apply for both. These restrictions included occupational

exposure monitoring of workers exposed to DOP aerosols and liquid, medical surveillance, issue of personal protective equipment, formal notification to workers of associated risks, and labeling of work areas as "cancer suspect agent areas."

Clearly, these actions placed severe restrictions upon routine, 100% quality assurance testing of filters and other equipment. For this reason, in 1988 the U.S. Army initiated a detailed study of the problem of finding an acceptable substitute material for DOP that could meet all standard military test specifications while itself being a non-carcinogen and, ideally, having other attributes including acceptable acute inhalation toxicity, low cost, ready availability, and the ability to replace DOP directly in machines at test installations without retrofit or other modification of these machines.

This paper describes the experimental procedures and results of that study which are applicable primarily to "hot smoke" aerosol penetrometer machines including the Army-standard "Q-127" machine that is currently produced as the Model TDA-100 by Air Techniques, Inc. (ATI).[7] Several dozen samples of promising materials were obtained and tested in two filter-penetrometer machines: (1) a standard Q-127 "hot smoke" machine, and (2) the Los Alamos Monodispersed Aerosol Prototype Penetrometer (LAMAPP), a state-of-the-art developmental machine that produces smokes from liquids at room temperature. The two most promising materials were later tested in two brand new TDA-100 machines and compared with DOP smokes from a third brand new TDA-100 machine at the factory, by invitation of the manufacturer, ATI.

While this paper will discuss briefly the testing carried out and results obtained with both kinds of penetrometer machines, it will concentrate most upon "hot smoke" technology. Replacement materials for "cold smoke" applications will be discussed fully in a later paper.

II. Test Smoke Specifications

Present U.S. Army test specifications for acceptable "hot smokes" prescribe a geometric mean diameter (GMD) of between 0.18 um and 0.33 um, with a geometric standard deviation (GSD) equal to or less than 1.30, and a mass concentration at the test chuck, where filters are held for penetration measurements, of 100 $\pm$ 20 mg/m^3. The mass concentration of an aerosol in milligrams per cubic meter is given by the equation:

$$mg/m^3 = \frac{\pi}{6} \times 10^{-3} \; d \; N \; D_{um}^3 \qquad (1)$$

where d is the density of the material, N is the aerosol particle population per cubic centimeter, and D_{um} is the particle diameter in micrometers. In Figure 1, Equation 1 is plotted for unit density spheres for three values of the GMD. It is seen that as the aerosol mass concencentration approaches 100 mg/m^3, the number of 0.3 um particles per cubic centimeter approaches 7 x 10^6, and that this number for 0.2 um particles is well in excess of 10^7.

Populations as large as these exceed the operating ranges of laser aerosol spectrometers like the one discussed below that was used in our studies. Typically, these instruments can count 1.7 x 10^4 particles with 90% accuracy in a one cubic centimeter sample per second. Thus an accurate

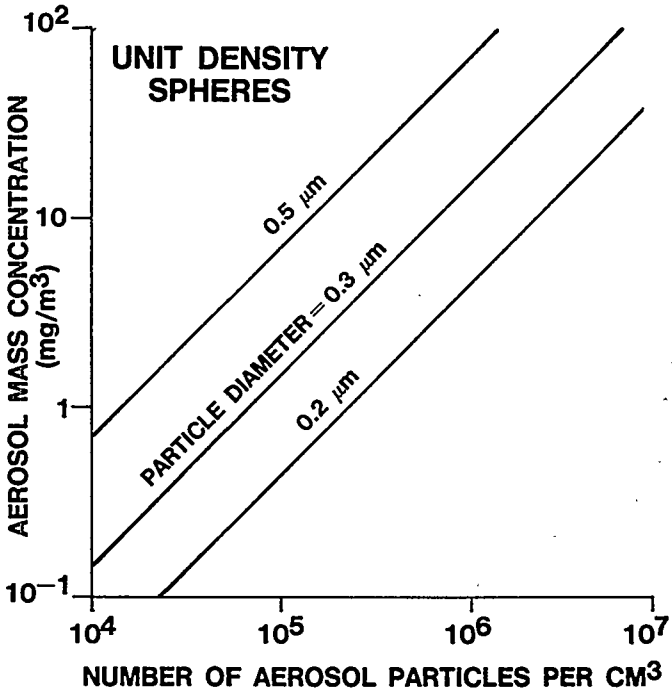

Figure 1. Plot of Equation 1 for Unit Density Spheres.

aerosol dilution of several hundred times is required to count the particles delivered to a Q-127 machine canister test chuck at 100 mg/m^3. In the research reported here, such aerosol dilutions were made routinely using precision equipment.

Our Q-127 machine was a refurbished older model of the similar but redesigned and simplified "monodispersed aerosol penetrometer" presently being marketed by ATI under the model number TDA-100.[7] These machines generate "hot smokes" by the vaporization and recondensation of DOP or other liquids, having suitable vapor pressure and other physical properties, which are placed in a heated reservoir. Hinds, et al,[8] described "hot DOP" aerosol size distributions produced by the ATI Q-127 machine more than a decade ago, and gave a good description of changes in aerosol output corresponding to various settings of the mechanical analyzer (modified nephelometer) or "Owl" that is standard equipment on all Q-127s and TDA-100s. They found that the output aerosol could be varied over the GMD range 0.23-0.30 um corresponding to angular settings of the Owl from 29^o to 45^o, respectively. These results can be compared to ours, which are reported below.

A PMS Model LAS-X laser aerosol spectrometer[9] was used in our research to measure GMD of test aerosols in four diameter ranges between 0.09 um and 3.00 um. Fifteen channels of size resolution were provided in each range. (Hinds, et al,[8] used a PMS Model ASASP-300 aerosol spectrometer; Skaats[10] performed similar DOP studies using an LAS-200 laser aerosol spectrometer in tests to Army specifications with Q-127, Q-107, and Q-76 machines). We used precision gas diluters to prepare samples for size measurement from the Q-127, which were drawn from the analyzer (Owl) chamber, for introduction directly into the LAS-X system.

The LAMAPP system is shown in Figure 2. It is described in greater detail in Ref. 12. A fine polydisperse aerosol is generated by a Laskin nozzle in a "cold pot" containing the test liquid. In a similar pot containing an NaCl solution, droplets are produced that evaporate to form salt condensation nuclei as they flow through a vaporization tube, where the DOP or other liquid aerosol is simultaneously vaporized. Upon cooling, the liquid recondenses on the salt nuclei to form droplets of controlled size and small GSD. These enter an aging chamber from which they are drawn for filter test purposes. Provision is made for calibration using polystyrene latex (PSL) or other standard aerosols. A laser aerosol spectrometer (LAS-X) and microcomputer permit GMD and GSD to be determined and printed on a strip chart.

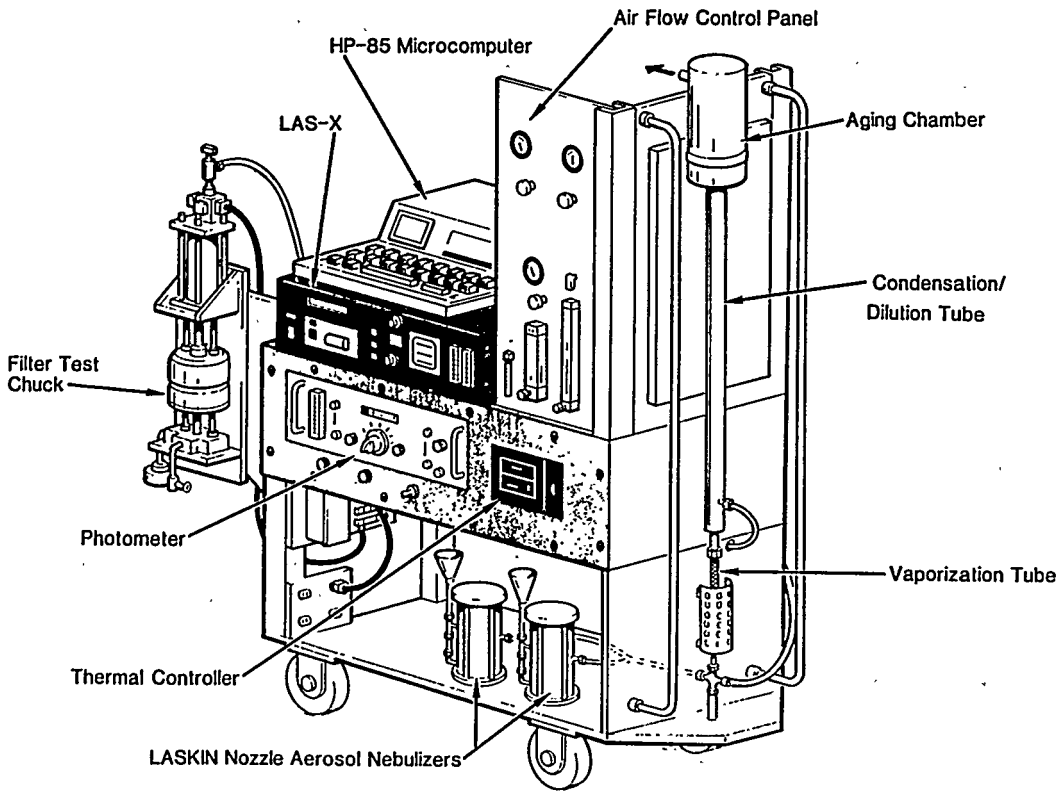

Figure 2. LAMAPP System.

Toxicological properties of candidate materials were of paramount importance in our study. Classes or families of materials known to be non-carcinogenic and relatively non-toxic were required to be screened for possible candidates which, at the same time, had physical properties such that they might produce acceptable aerosols in the penetrometer machines. And, ideally, they should offer other inducements to their use such as low cost.

Material Safety Data Sheets (MSDS) were compiled for promising materials, which included straight-chain saturated hydrocarbons (cosanes), moderately branched-chain saturated hydrocarbons, fatty acids, fatty acid esters, and glycols. The MSDS indicated that virtually all of these materials were classed as having very low toxicity to humans. Many, in fact, were approved for use in foods and cosmetics. But virtually no data were found to exist concerning the inhalation toxicity to humans of these materials when breathed as fine aerosols or smokes.

The U.S. Army Chemical Research, Development and Engineering Center (CRDEC) has developed computerized data bases of many chemical compounds that can be searched for toxicological and physical properties. Search routines were executed to determine which compounds from the classes of materials mentioned above were in these data bases. These materials were in turn searched to optimize matches of physical properties that were thought to be important in aerosol generation by vaporization/recondensation processes.

III. Selection and Testing of Candidate Materials

Several candidate replacement materials for DOP were identified on the basis of presumed inherent low toxicity. The approach was two-fold: (1) to find those materials that might work best in the machines, considering reproducible particle size, size distribution, smoke concentration, market availability, and cost; (2) to subject those materials to replacement testing in production and/or product assurance penetrometer machines, and seek final approval for the use of any successful material(s) to replace DOP.

Other factors which must be considered in choosing a material to replace DOP are: (1) the impact of a new material upon existing testing hardware must be minimal; (2) machine maintenance- it is undesirable to use a material that will clog the testers and/or will support fungus growth; (3) destructive vs. non-destructive testing- DOP penetration testing is considered non-destructive, and a new material must also be non-destructive in the sense that it will not damage filters in standard test aerosol concentrations (even DOP might damage filters in massive concentrations); (4) a new material must exhibit adequate stability and aging characteristics, especially at elevated reservoir temperatures found in Q-127 and TDA-100 machines.

Many physical properties appear to play some role in the behavior of candidate materials when they are used in filter penetrometer machines such as the Q-127. Among these physical properties are vapor pressure, surface tension, viscosity, and density.

Figure 3 presents a Clausius-Clapeyron plot for several potential candidate materials, relating the logarithm of vapor pressure to the reciprocal of absolute temperature in degrees Kelvin. The upper scale on the abscissa also gives temperature in degrees Celsius. The negative slope of the curve for each material corresponds to its heat of vaporization divided by the gas constant. Such curves are readily constructed if vapor pressures for a given material are known at two or more temperatures.

In Figure 3, it can be seen that the curve for DOP lies below the others, indicating that DOP has a lower vapor pressure at a given temperature than the other materials shown. As the molecular weight increases in a family of candidate chemical compounds, the vapor pressure decreases. Thus as the carbon chain length in capric, lauric and myristic acids increases from C = 10 to 12 to 14, respectively, the curves in Figure 3 for these acids approach the lower curve for DOP.

This simple analysis suggests that as the fatty acid carbon chain length increases a better match is made with the vapor pressure characteristics of DOP. Thus palmitic acid (C =16) or stearic acid (C = 18), which are not shown in Figure 3, should most closely match DOP in this respect. But the fatty acids just discussed are all solids at room temperature; their melting points range from 31°C for capric acid to 68°C for stearic acid. As their vapor pressure characteristics become more like those of DOP, they become increasingly difficult to work with. They will melt in the Q-127 machine pot and will vaporize, but the recondensation aerosols that they produce will become increasingly unpredictible with increasing chain length and melting point.

It is much easier to work with candidate materials that are liquids at room temperature. But such materials usually have two characteristics that are undesirable: (1) they are unsaturated; i.e., they contain C:C double bonds that

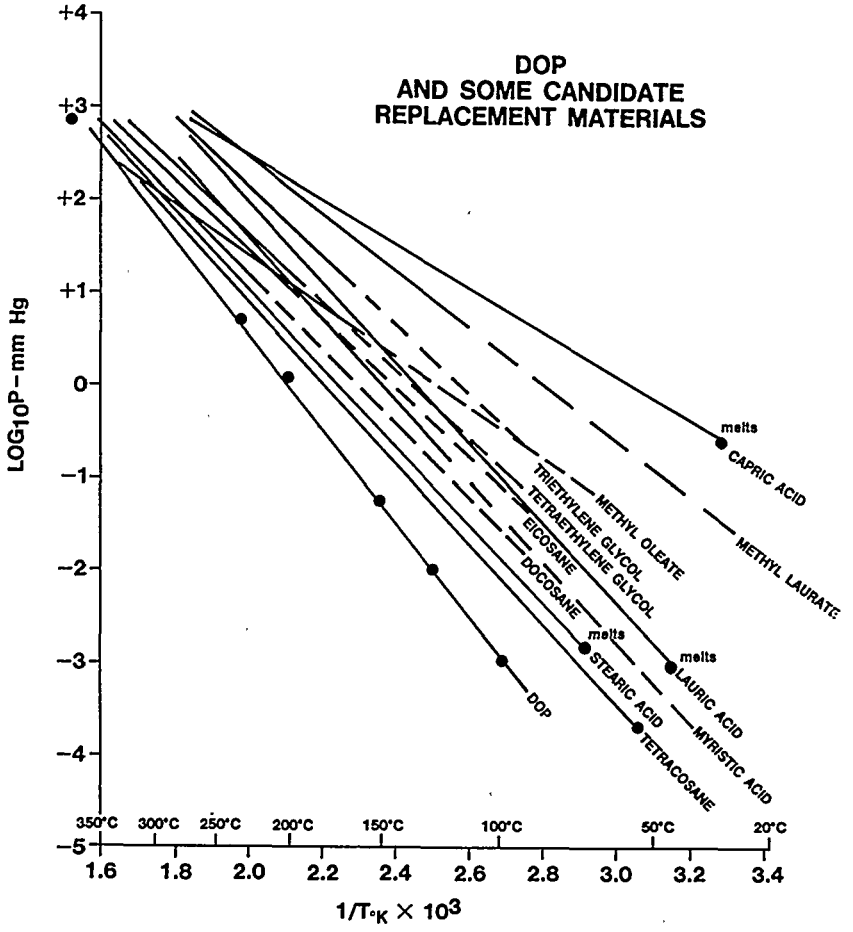

Figure 3. Clausius-Clapeyron Plot: DOP and Some Candidate Replacement Materials.

are sites for chemical attack leading to instability with aging, especially at elevated temperatures, and thus rancidity and fungus growth could result; (2) their carbon chains are branched, rather than straight as for the fatty acids just discussed, and this increasingly complex structure increases the probability that human toxicological problems will be encountered in their use.

Figure 3 suggests how compromises might be made to find a simulant for DOP to use in the Q-127 machine. Suppose, for example, that the Q-127 machine pot is normally maintained at 150°C (shown on the upper abscissa scale in Figure 3) when it contains DOP. This corresponds to an ordinate value of -1.3, or a DOP vapor pressure of 0.050 mm Hg. Moving horizontally to the right at the -1.3 ordinate value, we encounter the myristic acid curve at a temperature of about 100°C. This indicates that we should obtain the same vapor pressure with myristic acid at a pot temperature of about 100°C that we would obtain with DOP at 150°C.

There are other complications. For example, the Q-127 machine pot temperature might not be conveniently adjusted to a temperature as low as 100°C. Even if this were possible, the acid might not recondense into an aerosol under conditions achievable using other Q-127 machine settings, or it might not yield an aerosol with the proper specifications. Even if the acid were to perform well, it would still freeze during shut-down in the Q-127 machine pot and fittings, because its melting point is 52°C. Its inhalation toxicological properties would still remain to be investigated.

Our Q-127 and LAMAPP penetrometer machines were supplied by air from a compressor. The inlet air line was fitted with a refrigerative dehumidifier to remove moisture and other contamination, such as oil, from the air supply.

The machines were provided with two precision capillary air diluters in series to sample aerosols for analysis by the LAS-X laser aerosol spectrometer. Each air diluter was adjustable for dilution ratios up to 100:1 (10,000:1 maximum in series) at flow rates up to 5 standard lpm. Each diluter employed a closed system using filtered air from the original sample to mix with and reduce the concentration of the aerosol at the output. The dilution air passed through two in-line high-efficiency particulate aerosol (HEPA) filters.

LAS-X data for the Q-127 machine were compared directly with readings from the optical "Owl" device on the Q-127 machine which was set to a given angular reading (usually 29°) corresponding to a "standard" DOP test aerosol. In this way, Q-127 machine operators could be instructed as to which control settings should be used to obtain proper test aerosols from DOP replacement materials, since they would not normally have access to a LAS-X system in filter quality testing.

A detailed Standing Operating Procedure (SOP) was prepared to permit safe machine operation with DOP, as well as with candidate replacement materials. Candidate samples were aged in an oven at 140°C concurrently with their use in the penetrometer machines. In this way the stability and aging characteristics of promising candidate materials could be studied over periods ranging from weeks to months, even though their time of use in the machines would be limited to days because of the pressing schedule of candidate material testing.

A discussion of Q-127/TDA-100 and LAMAPP operating adjustments and procedures is beyond the scope of this brief presentation. Some information is contained in a paper submitted to the AAAR journal.[11] Complete details are contained in a CRDEC report that has been cleared for public release; a copy is available upon request from the authors.[12]

In summary, the required performance criteria for candidate materials to successfully replace DOP are clear: they must be toxicologically and environmentally innocuous when dispersed as aerosols, and they must have physical and chemical properties sufficiently similar to those of DOP to allow them to produce aerosols like those of DOP when subjected to evaporation and recondensation in filter penetrometer testers such as TDA-100 and Q-127 machines. Ideally, they should also work in machines that operate at room temperature.

About three dozen candidate materials were obtained from several suppliers including the primary manufacturers.[13] These materials were either liquids or solids at room temperature. A test priority was established such that the materials were evaluated in the sequence: (1) liquids in the Q-127 machine; (2) solids in the Q-127 machine, in order of increasing melting point; (3) liquids in the LAMAPP machine.

Our experiments identified several materials that are viable candidates to replace DOP in "hot smoke" penetrometer machines such as the Q-127/TDA-100, and in "cold pot" machines like LAMAPP. These are summarized in Table 1, with sources of supply.[13] The materials identified here as DOP alternatives or replacements are generally inexpensive, and readily available. Aging tests at elevated temperatures, which are presently underway, should identify candidate materials that are thermally unstable.

Most candidates should be quite stable in long-term testing, if not contaminated in use. Indeed, some candidate materials that contain significant percentages of "impurities" (compounds similar to the primary compound, but more volatile) actually seem to improve in performance with aging at elevated temperatures.

Table 1. Recommended Replacement Materials for DOP in Q-127 and TDA-100 Machines, and in the LAMAPP Machine, Ranked in Order of Probable Success.

	Q-127 and TDA-100 Machines		LAMAPP Machine	
Ranking*	Chemical Name	Manufacturer[13] or Source	Chemical Name	Manufacturer[13] or Source
1	synthetic hydrocarbon Emery 3004	Emery Group Henkel Corp.	synthetic hydrocarbon Emery 3002	Emery Group Henkel Corp.
2	isostearic acid (76%) Emersol 875	Emery Group Henkel Corp.	isostearic acid (76%) Emersol 875	Emery Group Henkel Corp.
3	isostearic acid (66%) Emersol 871	Emery Group Henkel Corp.	methyl oleate stearate Emery 2219	Emery Group Henkel Corp.
4	synthetic hydrocarbon Emery 3006	Emery Group Henkel Corp.	synthetic hydrocarbon Emery 3004	Emery Group Henkel Corp.
5	oleic acid (71%) Industrene 206LP	Humko Chem. Div., Witco Chem. Corp.		
6	oleic acid (74%) Emersol 233LL	Emery Group Henkel Corp.		

* Highest rankings have highest probability of success.

IV. Discussion

Isostearic acid has the structural formula:

$$CH_3-\underset{\displaystyle CH_3}{CH}-(CH_2)_{14}-COOH \qquad (2)$$

where the single branched methyl group usually occurs in the position shown but also can occur at any other position along the carbon chain with a much lower probability. Thus it is an isomer of stearic acid, but the two acids have distinctly different physical properties. Isostearic acid is a light yellow liquid at room temperature with a melting point of 12-15°C, depending

upon its purity, and it has a vapor pressure of 50 mm Hg at 265°C. Its flash point is approximately 182°C, open cup.

Two samples of differing purity were used in this work. The purest sample was 70-76% isostearic acid, with the remainder consisting of myristic, isopalmitic, and palmitic acids, in that order. The less pure sample was 60-66% isostearic acid, with the remainder consisting of isooleic, oleic, stearic, and isopalmitic acids, in that order.

Both samples worked very well in our Q-127 machine, with the purer material yielding slightly higher aerosol concentrations. The purer material also worked very well in the LAMAPP machine. This can be observed in Table 1. The oxidation stability of isostearic acid appears to be excellent compared to other candidate materials. A weak acid, isostearic acid is not recommended for use in contact with chemically-active metals such as brass and iron, especially at elevated temperatures. Stainless steel, or other inert materials should be specified for the fabrication of the "hot pot", e.g., on TDA-100 machines, if isostearic acid were to be used owing to its other desirable properties. For this reason a synthetic hydrocarbon, described below, was given first priority for use in hot smoke machines, as is shown in Table 1.

Synthetic hydrocarbons include poly-alpha olefins (PAOs), which are used as synthetic lubricants, and in other applications. These versatile, saturated, synthetic hydrocarbons are produced by direct oligomerization of decene-1. Linear alpha olefins are polymerized and hydrogenated to manufacture PAOs. Three PAOs were investigated in our studies; these are designated Emery 3002, 3004 and 3006. Data are summarized in Table 2.

Table 2. Properties of Poly-Alpha Olefins (PAOs).

Trade Name	Pour Point, °C	Flash Point, °C	Fire Point, °C	Auto-Ignition Point, °C	Specific Gravity
Emery 3002	-65	164	178	324	0.80
Emery 3004	-69	225	250	343	0.82
Emery 3006	-64	243	266	371	0.83

See Ref. (13)

In the Q-127 machine, Emery 3004 performed best, giving a GMD adjustable from 0.2 to 0.3 um, with a GSD of 1.23 and an adequate aerosol yield. The "pot" temperature was 180°C. Emery 3002 was too volatile, and produced large aerosol yields that could not be adjusted to GMDs below 0.3 um, and which had GSD values of 1.40-1.67 or more. Emery 3006 had to be heated to 195°C to yield 110 mg/m^3 of aerosol with a GSD value of 1.20, but the GMD could not be adjusted above 0.21 um.

Emery 3002 performed well in the LAMAPP machine, giving smokes with specifications as good as those for Emery 3004 in our Q-127 machine, and thus it was given first priority for use in the LAMAPP, compared to fourth priority for Emery 3004 in that machine.

Oleic acid has the structural formula:

$$CH_3-(CH_2)_7-CH=CH-(CH_2)_7-COOH \qquad (3)$$

where one double bond exists between the ninth and tenth carbons of an 18-carbon chain. The molecule is most easily chemically attacked at this double bond, making this acid less stable during aging at elevated temperatures than saturated fatty acids. Nevertheless, it performs well in the Q-127 machine with the exception that the aerosol concentration is so great that is is sometimes difficult to control. Oleic acid is not recommended for use in the LAMAPP machine.

The oleic acid used here was 71-74% pure, with the remainder consisting mainly of palmitoleic and linoleic acids, in that order. It is a light yellow liquid at room temperature, with a slight odor. It melts at 11-14^{o}C, and has a vapor pressure of 10 mm Hg at 224^{o}C. Its flash point is approximately 184-189^{o}C closed cup, and 193^{o}C open cup.

Methyl oleate stearate is a mixture of 58% methyl oleate, whose structural formula is:

$$CH_3-(CH_2)_7-CH=CH-(CH_2)_7-COO-CH_3\ , \qquad (4)$$

24% methyl stearate, whose structural formula (where n = 18) is:

$$CH_3-(CH_2)_{n-2}-COO-CH_3 \qquad (5)$$

14% methyl linoleate, and 4% methyl palmitate (n = 16 in the formula above). It melts at 18^{o}C, and is a light yellow liquid. Its vapor pressure is 10 mm Hg at 205^{o}C, and its flash point is approximately 173^{o}C open cup.

This material produced a thin smoke in the Q-127 machine that could not be read by the Owl. The GMD range was 0.24-1.1 um, and the GSD range was 1.40-1.50. However, the material performed well in the LAMAPP machine, for which it was given third priority for use as indicated in Table 1.

Our primary mission in this program was to identify a safe replacement material for hot smoke machines including Q-127/TDA-100s that could be used to replace DOP directly in existing machines with a minimum of downtime and/or machine modification. That best material (Emery 3004) would then have to pass toxicological screening so as to, ultimately, obtain approval by the U.S. Army Surgeon General for use Army-wide. The remainder of this paper thus concerns itself with hot smoke machines; the LAMAPP will receive little further attention here but is considered in detail elsewhere.[12]

We were invited by the management of Air Techniques, Inc.[7] to carry out comparative testing of DOP and our two best candidate replacement materials in three brand-new TDA-100 (formerly Q-127) penetrometer machines on their production line, during "break-in" in the early months of 1990. Data obtained from these tests were extremely valuable.

The materials selected for comparison with DOP were Emersol 875 isostearic acid, and Emery 3004 synthetic hydrocabron (poly alpha olefin, or "PAO"). The reservoir ("hot pot") of each machine was was filled with one of the three materials. The machines were adjusted to give smoke yields at the test chucks of 100 mg/m^3, in accordance with Army specifications. The DOP machine was

adjusted according to ATI specifications for new machines, and the machines containing isostearic acid and PAO were initially set up using the control setting from our research, which were then modified slightly as needed to obtain the required smoke yields. The machines, which were identical, were then operated for 100 hours on eight-hour daily, 40-hour weekly, schedules. Samples of the materials were collected into glass vials from the hot pots after approximately every 20 hours of operation, for future chemical analyses to determine whether decomposition, polymerization, or other changes occured during this thermal "aging."

DOP and PAO were observed to have similar aging properties. Both remained clear for 10-20 hours of operation at temperature, and then exhibited a deepening honey color which turned to yellowish-brown after 100 hours. The isostearic acid, however, began to chemically attack brass parts used in the ATI machine hot pots. Within a few hours the liquid had turned greenish, and within 10-20 hours a deep greenish-black color developed. At the end of 100 hours, the material was black and quite viscous, indicating that polymerization might have been catalyzed by iron and copper complexes that were taken into solution. ATI engineers expressed an interest in experimenting with modified hot pots of stainless steel, glass, ceramic, or other inert construction, that could eliminate these corrosion problems. Such machine modifications might also reduce discoloration and sludging of DOP and other liquids used in the pots, thus increasing elapsed times to the accumulation of sludge, with required periodic maintenance, and requiring less frequent replacement of the material itself.

After the break-in period, our LAS-X and diluter systems were taken to the ATI factory for detailed measurements of the test smokes from the three machines. Trials were first run with the aged materials in the machines. The hot pots were then drained and refilled with fresh materials, and a second set of trials was run.

The new TDA-100 machines are designed around standard DOP specifications, and their operating temperatures are controlled by fixed-wattage heaters. Nevertheless, it was demonstrated that Army test specifications could be met with the new materials, without modification of the machines. ATI engineers indicated that if it were desired to deliver a new machine with, e.g., PAO instead of DOP, only a minor modification would be required to match the heater wattage to the proper operating temperature range of the replacement material, thus further improving the "monodispersity" of the of the test smoke.

After the machines were refilled with fresh materials, it was found that the specifications of the DOP and PAO smokes were very similar. Within the range of operating controls, the PAO smoke produced had a GMD of 0.22 um with a GSD of 1.29, while the DOP smoke, with optimum machine settings, had a GMD of 0.21 um with a GSD of 1.29. The fresh isostearic acid also was found to be able to produce a smoke that met Army specifications, within the available range of machine operating controls, but the latter material was not subjected to further detailed testing. Primarily because of its non-corrosiveness and better aging properties, PAO was selected as the tentative candidate of choice to replace DOP in existing Q-127 and new TDA-100 penetrometer machines with a minimum of downtime and machine modification.

Further testing at ATI with Emery 3004 allowed operators to become familiar with the range of TDA-100 adjustments and their interactions, so that smoke specifications were gradually improved using this new material. In most recent tests, Emery 3004 produced smokes with GSDs smaller than 1.25.

Very recently manufacturers of PAOs[13] including Emery 3004 have reissued MSDSs, as required by law when previously unknown but pertinent information becomes available, to reflect the 1982 findings of Guiney[14] on the acute toxicity assessment of PAOs.

The new Emery MSDS for PAOs claim that although the toxicities estimated in Guiney's study were significant, the test conditions were such that the test animals (rats) were subjected to great physical insults, sufficient that observed histopathical changes may have been a response to these insults rather than being related to a specific compound toxicity effect. The MSDS indicate that PAOs may be considered non-hazardous, for all practical purposes, by inhalation. Indeed all oils, including the purest of mineral oils, are capable of causing "oil pneumonia" and/or death by suffocation when inhaled at very high aerosol concentrations for periods of more than several minutes. Since oils cannot be cleansed easily from the lungs due to their extreme hydrophobicity, and a build-up and coating of the lung surfaces is inevitable. Humans and animals subjected to such physiological insults respond by coughing, and by exiting the contaminated area. A confined test animal, on the other hand, would be subjected to great physical and psychological trauma in the presence of very high, suffocating oil aerosol concentrations, and could succumb rapidly for a variety of reasons.

To investigate the exact nature of conditions to which the test rats were subjected, visual and other data observations from Guiney's paper were analyzed using optical constants from a paper[15] containing optical extinction (i.e., attenuation) coefficients at wavelengths including the visible, for oil smokes and aerosols. From the extinction coefficients, the actual oil aerosol concentrations to which the rats were subjected were calculated. The test visibility conditions were such that the heads of the rats confined in test chambers could be seen under normal lighting conditions, but their hind quarters could not. This would be roughly equivalent to subjecting oneself to an oil aerosol or "fog" so thick that one could not see a hand six inches in front of his or her face. The aerosol mass concentration was estimated at 5,000 mg/m^3 (5 g/m^3), or roughly 1,000 times the PEL or TLV values normally given as safe for occupational exposure limits for oil mists.

V. Conclusions and Recommendations

In summary, it would appear that the test conditions under which PAOs were considered to be potentially toxic by inhalation were completely unlike those reasonably expected to be found in actual experience. The acute toxicities of the PAOs are comparable to that of DOP. Since Emery 3004 presents no evidence of carcinogenicity,[16] is not toxic under normal operating conditions, and can be used to replace the suspected carcinogen DOP directly in operating machines (with no mission interruption and while meeting or exceeding accepted Army filter test standards), it must be considered the prime candidate for approval as a DOP replacement by the U.S. Army Sugeon General.

References

1. Hinds, W.C., Macher, J.M., and First, M.W., "Size distribution of aerosols produced by the Laskin aerosol generator using substitute materials for DOP," Am. Ind. Hyg. Assoc. J. 44(7):495-500 (1983).

2. Hinds, W., Macher, J., and First, M., "Size distributions of test aerosols produced from materials other than DOP," J. Environ. Sci. 25:20-21 (1982).

3. Gerber, B.V., "Selected polyethylene glycols as 'DOP' substitutes," Proc. 16th D.O.E. Nuclear Air Cleaning Conference, San Diego, CA, 20-23 October 1980, U.S. Dept. of Energy and the Harvard Air Cleaning Laboratory, M.W. First, ed., February 1981:109-124 (1980).

4. Sharaf, M.A., and Troutman, S.J., "Comparative size distribution and filter penetration measurements of DOP and corn oil aerosols," Particulate Science and Technology 6:207-217 (1988).

5. Gerbig, F.T., and Keady, P.B., "Size distributions of test aerosols from a Laskin nozzle," Microcontamination. July 1985:56-61 (1985).

6. Murrow, J.L., and Nelson, G.O., "HEPA-filter testing: Comparison of DOP and NaCl aerosols," Proc. 12th A.E.C. Air Cleaning Conference; Vol. 12:808-816 (1973).

7. Air Techniques, Inc. (ATI), Division of Hamilton Associates, Inc., 1716 Whitehead Road, Baltimore, Maryland 21207.

8. Hinds, W., First, M., Gibson, D., and Leith, D., "Size distribution of 'hot DOP' aerosol produced by ATI Q-127 aerosol generator," Proc. 15th D.O.E. Nuclear Air Cleaning Conference Boston, MA, 7-10 August 1978:1130-1144 (1978).

9. Particle Measuring Systems (PMS), Inc., 1855 South 57th Court, Boulder, Colorado 80301.

10. Skaats, C.D., "A study of dioctyl phthalate particles (DOP) generated in penetrometers and the devices used currently to measure their size," Proc. 15th D.O.E. Nuclear Air Cleaning Conference, Boston, MA, 7-10 August 1978:1127-1129 (1978).

11. Carlon, H.R., Guelta, M.A., and Gerber, B.V., "Some candidate replacement materials for DOP in 'hot smoke' aerosol penetrometer machines," submitted to Aerosol Science and Technology (1990).

12. Carlon, H.R., Guelta, M.A., and Gerber, B.V., "A study of candidate replacement materials for DOP in filter-testing penetrometer machines," Technical Report CRDEC-TR-053, U.S. Army Chemical Research, Development and Engineering Center, Aberdeen Proving Ground, MD 21010-5423, March 1989.

13. "Emery" and "Emersol" products are available from Emery Group, Henkel Corp., 11501 Northlake Drive, Cincinnati, OH 45249, (513) 530-7300; "Industrene" products are available from Humko Chemical Div., Witco Chemical Corp., P.O. Box 125, Memphis, TN 38101, (901) 320-5941.

14. Guiney, P.D., "Acute toxicity assessment of polyalphaolefin (PAO) synthetic fluids," Proc. Symposium on Synthetic and Petroleum Based Lubricants, Div. of Petroleum Chemistry, Amer. Chem. Soc., Las Vegas, NV, 28 Mar - 2 Apr 1982: 381-389 (1982).

15. Carlon, H.R., Anderson, D.H., Milham, M.E., Tarnove, T.L., Frickel, R.H., and Sindoni, I., "Infrared extinction spectra of some common liquid aerosols," Applied Optics 16, 1598-1605 (1977).

16. Tancrede, M., Wilson, R., Zeise, L., and Crouch, E.A.C., "The carcinogenic risk of some organic vapors indoors; A theoretical survey," Atmospheric Environment 21(10):2187-2205 (1987).

DEVELOPMENT OF FILTER SYSTEMS - PART 2

Injection Systems and Multi-Point Sampler Evaluation

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ABSTRACT

Two essential requirements for accurate in-situ efficiency testing of HEPA filters are a uniform challenge to the whole of the upstream filter face and representative sampling of the upstream challenge and the downstream leakage aerosol. Uniform challenge is defined as a concentration variation of aerosol across the plane of the duct of **less than +/-10%.**

Where systems have primary and secondary filters which have only very short distances between them, say less than 2 duct diameters, efficiency testing of individual filters to a satisfactory level would be impossible if normal practices were followed.

Work carried out at the Harwell Laboratory and reported here, investigates a method to individually efficiency test filters in a system which employs 4 circular filters in a 2 x 2 bank (2 primary and 2 secondary), with the primary and secondary circular filter housings connected directly to each other.

Initial work established the relationship between the number of injection points and mixing length and a simple multi-point injector achieved a mixing length of 8 duct diameters with only a minimal pressure drop penalty. Some design criteria previously used were shown to grossly over estimate the effect of increasing the number of injection points.

Further work was carried out to investigate a multi-point sampler which could be positioned at the outlet to the primary housing to achieve an efficiency result as accurate as if a single point sampler were positioned some distance downstream in a fully mixed air flow.

I. INTRODUCTION

The ventilation plant of an active facility at Harwell Laboratory is being refurbished. The old system, which employed rectangular HEPA filters rated at 1700m^3/h is to be replaced by circular filters rated at 3400m^3/h. The fume cupboard extract which forms part of this new system was erected in a development laboratory for pre-commissioning trials and development work.

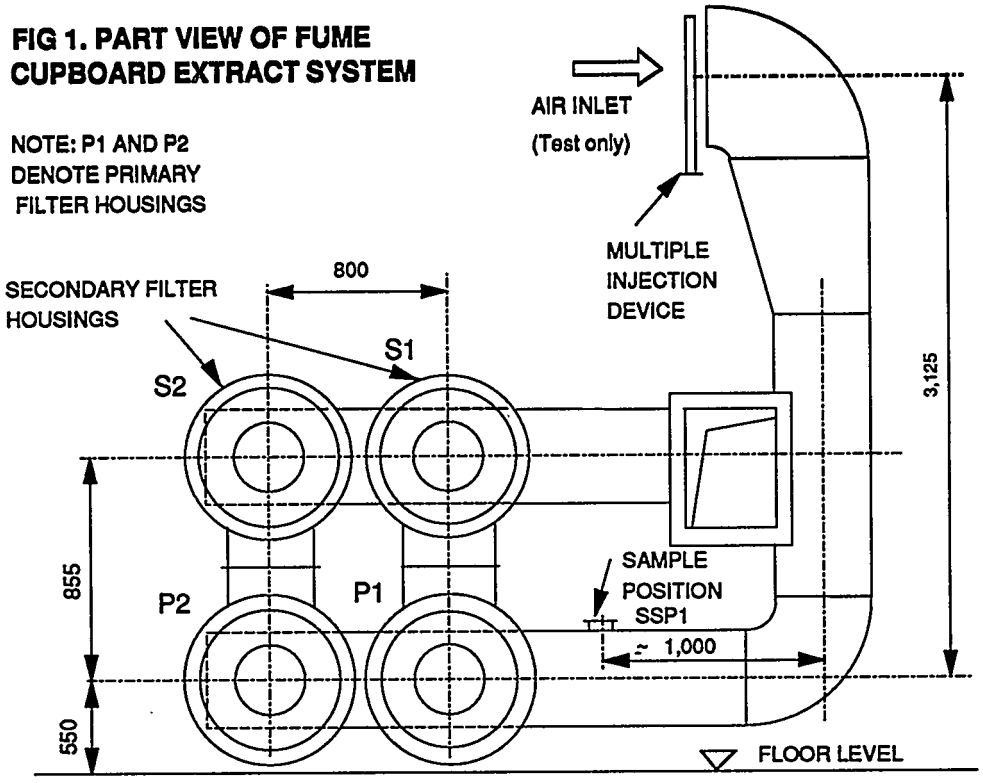
The fume cupboard extract system, which is shown in Figures 1 and 2, consists of 4 circular filters in a 2 x 2 bank (2 primary and 2 secondary), and has an operating flowrate of 6,800m^3/h (4,000cfm). Because of space constraints from the existing building design it was not possible to design the installation for efficient in-situ testing using the normal design concept of long mixing lengths or high pressure drop devices, eg Stairmand discs.

The mixing length available upstream of the primary filters is only 10 duct diameters and the primary and secondary filter housings are close-coupled, with a separation of only 1 duct diameter. It was necessary, therefore to develop low pressure drop injection devices which would meet the mixing requirements within the 10 diameter constraint, and also to develop a multi-point sampling system which would function between the 2 stages of filtration and enable the in-situ efficiency of the primary filters to be determined.

The work programme can conveniently be split into 3 sections:

- Characterisation of the system in terms of velocity profiles, volumetric flowrates and pressure drops through the system.
- Development of a low pressure drop multi-point injection system capable of providing a uniform challenge to the filters under test.
- Development of a multi-point sampling system for measuring the aerosol concentration between the primary and secondary filters.

FIG 1. PART VIEW OF FUME CUPBOARD EXTRACT SYSTEM



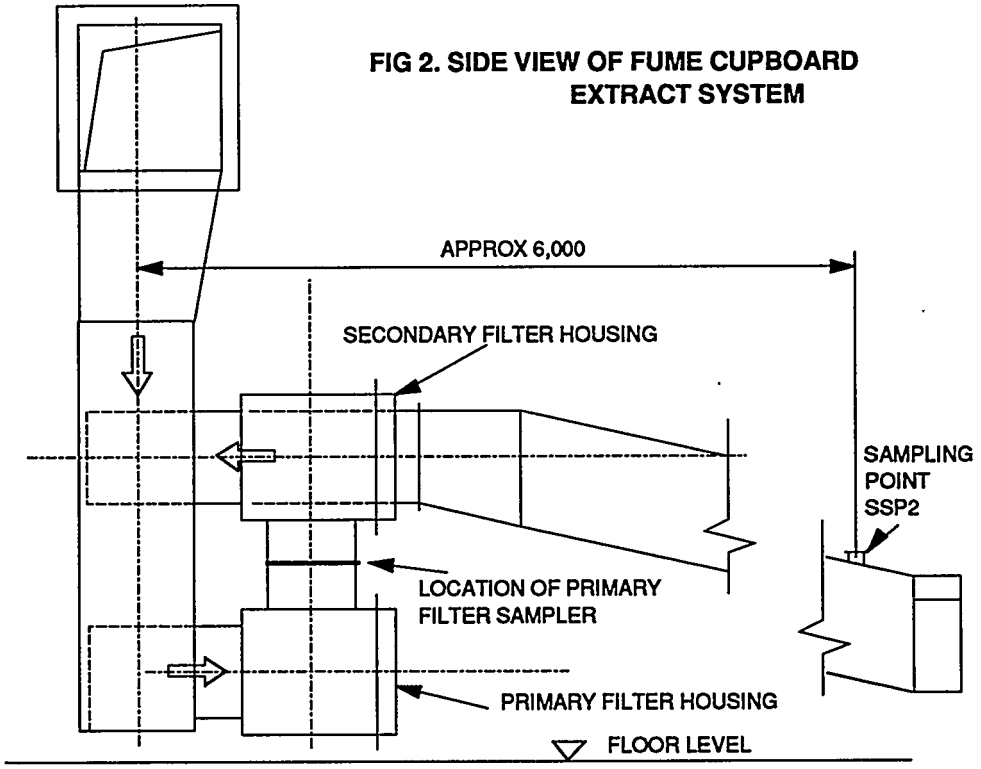


FIG 2. SIDE VIEW OF FUME CUPBOARD EXTRACT SYSTEM

II. SYSTEM DESCRIPTION

System Description

The system under test is shown in Figures 1 and 2. It consists of a centrifugal fan which draws air through the ductwork. The fan is controlled by an adjacent damper. The system employs low temperature circular filters rated at 3400 m^3/h (2000 cfm) which are in accordance with AESS 30/95100[2]. The filters are mounted in standard "rear entry" housings[3] available from M. C. Air Filtration Ltd. The nominal flow rate is 6,800m^3/h (4000cfm), each of the four filters running at rated flow.

The two primary filters are installed such that their airflow is in the normal direction i.e. in through the centre of the filter and out radially. The secondary filters are installed such that they operate in the reverse direction to facilitate the close coupling of the units and keep the plant room size to a minimum.

The distance between the inlet to the system erected for these trials - which is also the furthest upstream an injector could be positioned - and the inlet to the primary housings is approximately 10 duct diameters. The separation distance between the primary and secondary filters is approximately 1 duct diameter.

III. BASIC OPERATING CHARACTERISTICS

Velocity Profiles

An air velocity meter which utilises hot wire anemometry was used to indicate the air velocity along the horizontal and vertical centre line of the duct at the sampling positions SSP1 and SSP2. Results are shown in Table 1 below and in Figures 3 and 4. The profiles appear relatively flat and are consistent with a well developed flow. Positions 1 and 2 at the upstream sample position (values of 5.9 and 7.4m/s) are lower than the other results which may be attributed to additional turbulence associated with the bend located immediately upstream of this point.

TABLE 1. Velocity Profile Results

	Velocity in m/s			
	Upstream Position (SSP1)		Downstream Position (SSP2)	
Position	Vertical	Horizontal	Vertical	Horizontal
1	5.9	8.5	8.0	8.0
2	7.4	8.9	9.4	9.2
3	8.4	9.0	9.5	9.3
4	9.0	8.7	9.0	9.4
5	9.0	8.7	8.7	9.6
6	9.2	8.8	8.4	9.6
7	9.6	8.8	7.7	8.5

Pressure Differential

The pressure differential for the whole system, i.e. between SSP1 and SSP2, was measured using standard micromanometers at a range of volume flows. The pressure differential across each of the 4 filter housings was also recorded at rated flow.

Results are shown in Table 2 and 3 below. The overall pressure differential of the system at its rated flow, with four clean filters in position, was recorded as 825 Pascals (82.5 mm W.g).

TABLE 2. Pressure Drop Across System

Flowrate	m^3/h	3,400	4,250	5,100	5,950	6,800	7,522
	(cfm)	2,000	2,500	3,000	3,500	4,000	4,425
Pressure Drop	Pa	305	435	530	660	825	950

FIGURE 3: VELOCITY PROFILES (Vertical Scan)

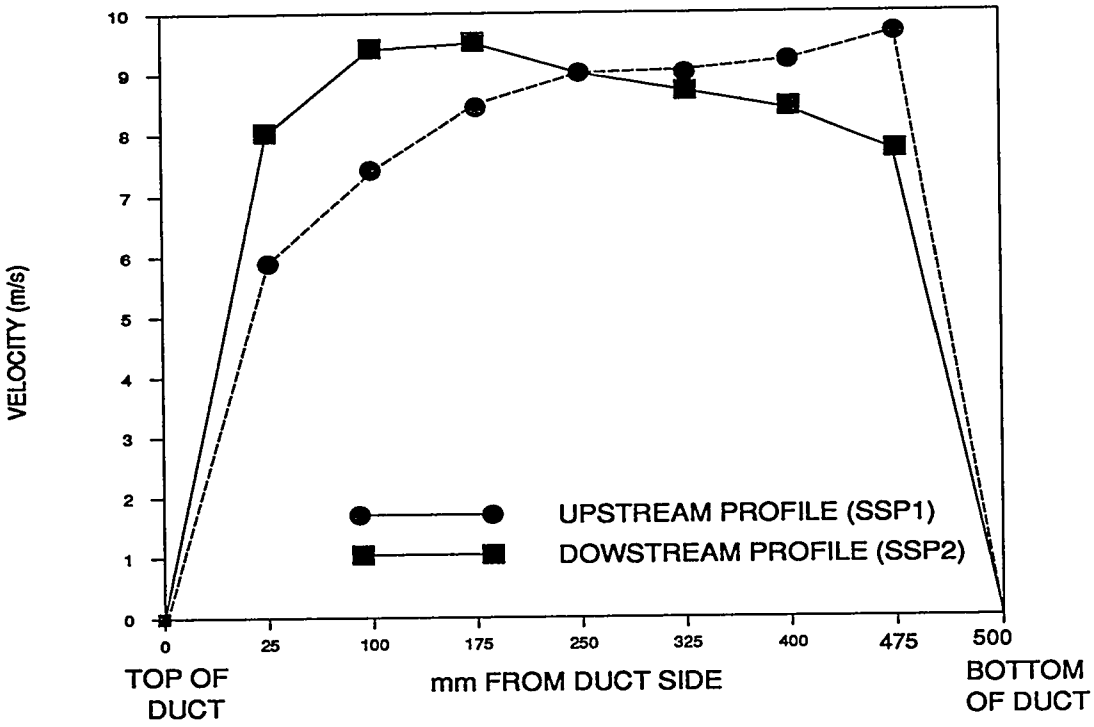


FIGURE 4: VELOCITY PROFILES (Horizontal Scan)

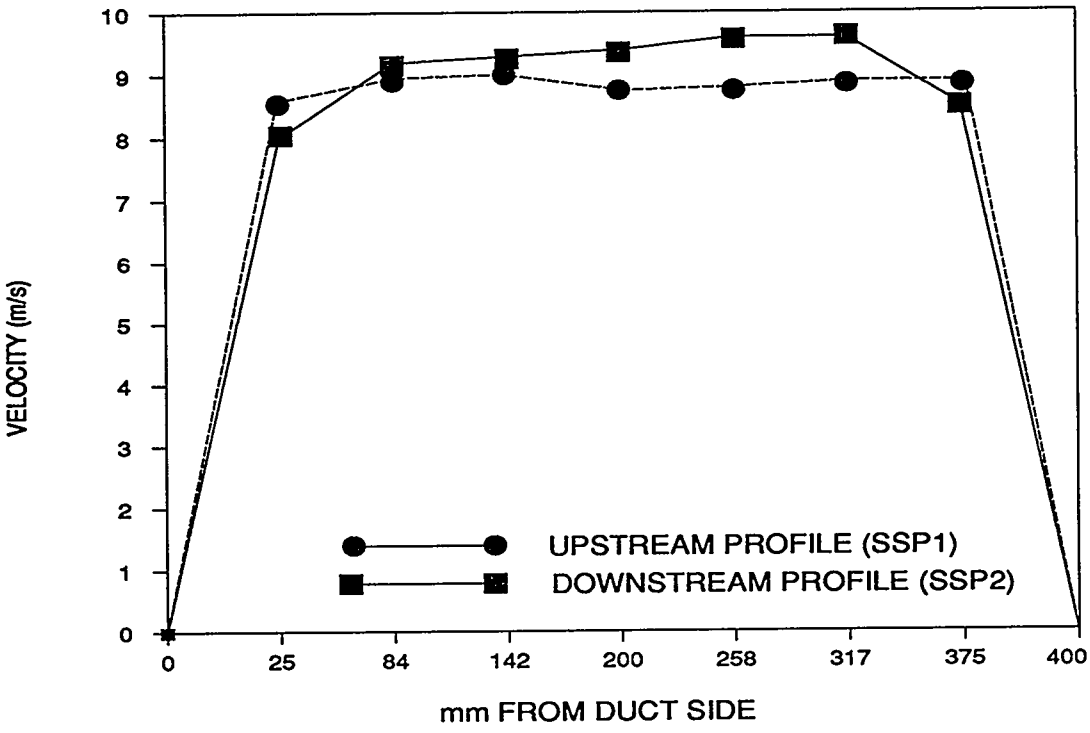


TABLE 3. Pressure Drop Across Housings
(at 6,800 m^3/h)

		Primary Filter		Secondary filter	
Housing		P1	P2	S1	S2
Pressure Drop	Pa	405	360	330	350

Note: Typical results for this design of housing with clean filters operating in the normal and reverse flow directions are, 410 Pa and 360 Pa respectively[4]. P1, P2, S1 and S2 refer to Primary and Secondary housings, see Figure 1.

System Efficiency

Using standard in-situ testing techniques and equipment (Aerosol photometer, Ondina-EL aerosol[5] the efficiency of the system between SSP1 and SSP2 was as detailed below:

	Efficiency	Penetration	Decontamination Factor (DF)
At 6,800m^3/h:	> 99.9995%	< 0.0005%	> 2.0×10^5

As the DF of a single filter might be as high as 3×10^4 (99.997%), it is possible that one of the two filters in series is outside the specified 2×10^3, but the overall resolution of the test technique is insufficiently sensitive to verify this. It is therefore necessary to develop an injection and sampling system so that the site installed efficiency can be verified

On system start up the technique would be to install the secondary filters and measure their efficiency between SSP1 and SSP2. If acceptable, the primary filter would then be installed and its efficiency measured between SSP1 and the multi-point sampler installed between the two filter banks.

During service, the primary filter can be retested when changed, the frequency of change is anticipated to be 4 times that of the secondary. Without the development of a suitable sampler testing of the primary could only be undertaken when the secondary filters were changed.

IV. DEVELOPING A MULTI-POINT INJECTOR

Introduction

In the system only 10 duct diameters of ventilation ductwork were available upstream of the primary filter housing for the injection and mixing of the test aerosol.

The traditional method of achieving a uniform upstream challenge - using a Stairmand disc mixer - whilst achieving a mixed flow in approximately 10 duct diameters[6] has the major disadvantage of a high pressure drop: over 200 Pa at 9 m/s duct air velocity. An alternative - low pressure drop - method of injection/mixing was required as the additional resistance and added complexity of "feathering" a Stairmand disc mixer when not in use was unacceptable.

In order to be accepted any alternative method must give a uniform challenge within the 10 duct diameters limit and have a significantly lower pressure differential than the Stairmand disc. The use of a multi-point injection array is one such alternative, effectively reducing the duct cross section served by each injection hole. To enable a suitable injector to be designed it was important to establish the relationship between the number of injection points and the mixing length achieved.

Previous Design Guide Recommendations

Earlier experimental work had shown that the decrease in mixing length is not as great as might be expected from the simple theory used in a number of Design Guides and Codes of Practice[7,8]. These suggest that a single injection point would achieve a mixed flow in only 12 duct diameters and that using 4 symmetrically arranged injection points, (no mixing device), would reduce the required distance to less than 6 duct diameters (De).

However, test results [6] have shown that using a single point injector requires upwards of 25 De to achieve mixed flow and a mixing length of 10 De can only be achieved with a single injection point in conjunction with a Stairmand disc mixer. Therefore, if the referenced design guides[7,8] are followed this would lead to a reduction in the accuracy of the measured in-situ efficiency value and in some cases a defective filter could be left in place.

Test Results

Tests were carried out in 600 mm square ductwork at duct air velocities between 4 m/s and 10 m/s. The results indicate that using 25 points of injection, each 3 mm in diameter, with a leg diameter of 15 mm will mix the aerosol, to within +/-10% of the mean concentration, within 10 De, see Table 4. Using a 22 mm leg diameter will give a 15% improvement in performance with mixing achieved in only 8.5 De.

The aerosol generator used for these tests was a Taylor TDA-4A (compressed air) unit which has the capacity to operate with up to 8 jets at a supply pressure up to 40 psi.

Table 4. Multiple Point Injector Results.

No. of Legs	No. of Holes	Dia. of Holes (mm)	Dia of Legs (mm)	Mixing Length (De)
3	9	3	15	>15
4	16	3	15	14
5	25	3	15	10
5	25	3	22	8.5

Effect of Hole Size

Tests have been carried out on a 22 mm diameter 5 legged, 25 point injector with varying hole sizes as indicated in Table 5. Results indicate that a 3 mm hole diameter is the most appropriate for the injectors tested. The most appropriate ratio (hole to leg) of cross sectional area (CSA) is 1:10.

Table 5. Effect of Hole Diameter.

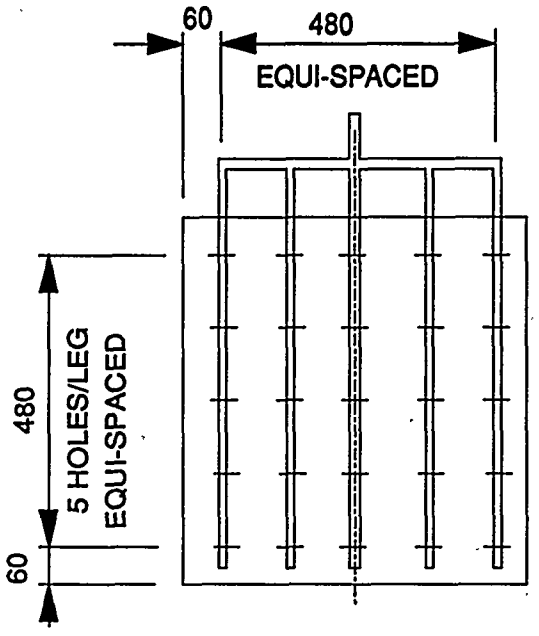
Diameter of leg (mm)	Diameter of hole (mm)	C.S.A. of leg (mm^2)	Hole C.S.A. per leg (mm^2)	Approx Ratio of C.S.A. per leg.	Mixing length De
22	3	380	35	1/10	8.5
22	4	380	63	1/6	10
22	5	380	98	1/4	12
22	7	380	192	1/2	>15

Design Considerations

When considering the use of this type of multiple point injector the following additional points are worth noting:

- Such multi-point devices can either be placed permanently in the ductwork (with only one break in the ductwork) or where old plant is involved they can be designed so that they can be retro-fitted, see Figure 5.
- This type of device is particularly useful where the duct cross section is non standard. They have been successfully retro-fitted into active plant where the mixing lengths were very short, in one case only 6 De, and where the duct was very narrow but wide (0.2 m x 1.2 m).
- The mixing lengths achieved are specific to tests carried out using the compressed air type of aerosol generator. A feature of this system is that compressed air used to generate the aerosol is driven out of the generator and into the injector/duct. Other systems such as the thermal or "hot" aerosol generators do not subject the aerosol to the same level of "forced" injection. Preliminary findings suggest an increase in mixing length in the region of 50% is required where there is no forced injection.

FIGURE 5: ARRANGEMENT OF 5 LEGGED 25 POINT INJECTOR



NOTE: For retro-fitting the top leg may be positioned inside the duct.
Only one break in the duct would therefore be required.

THIS DESIGN FOR 600 mm SQUARE DUCT

HOLE DIAMETER = 3mm
LEG DIAMETER = 15/22mm
(See text for details as sizes vary)

Design solution for this application

The unit used in this development work, see Figure 5, is of the 25 point, 5 legged, 22 mm diameter type each hole being 3 mm in diameter. For the system shown in Figures 1 and 2 tests indicated that mixing to within +/- 10% of the mean concentration was achieved in the distance available.

Testing of this device in a 600 mm square test rig, at a duct air velocity of 9 m/s, recorded a pressure drop of 60 Pa.

V. Developing a Multi-Point Sampler

Introduction

This section deals with the work carried out in developing a method for testing the two primary filters without the need to remove them from the system. This involved developing a multi-point sampler capable of obtaining a representative sample in the short section of duct between the two housings.

The sampler will be required to measure accurately the efficiency of a clean, unused filter with no leaks, typically 99.997% efficient, as well as lower efficiency values, in particular those of the order of 99.95% which is the current failure criteria. At this lower level the possibility of streaming of leakage aerosol - from a filter media or seal leak - is an added complication which must be considered.

The "standard" method of evaluating the performance of a new test method involves comparing the known penetration value of filters (as new and artificially damaged) against those obtained from testing the same filters using the new test method/sampler. The known penetrations are obtained in a standard test rig where both the aerosol challenge and leakage aerosol are fully mixed.

For this work the required effectiveness of the sampler is a deviation from the actual (or standard) penetration of less than +/-0.02% i.e. if the actual penetration is 0.05% the measured value must not be outside the range 0.03 - 0.07%.

Test Filters

Defects were added to two standard circular filters to provide the known and consistent penetrations required for this evaluation.

FIGURE 6: SECTIONAL VIEW OF PRIMARY FILTER & HOUSING
Showing relationship between location of defects and outlet

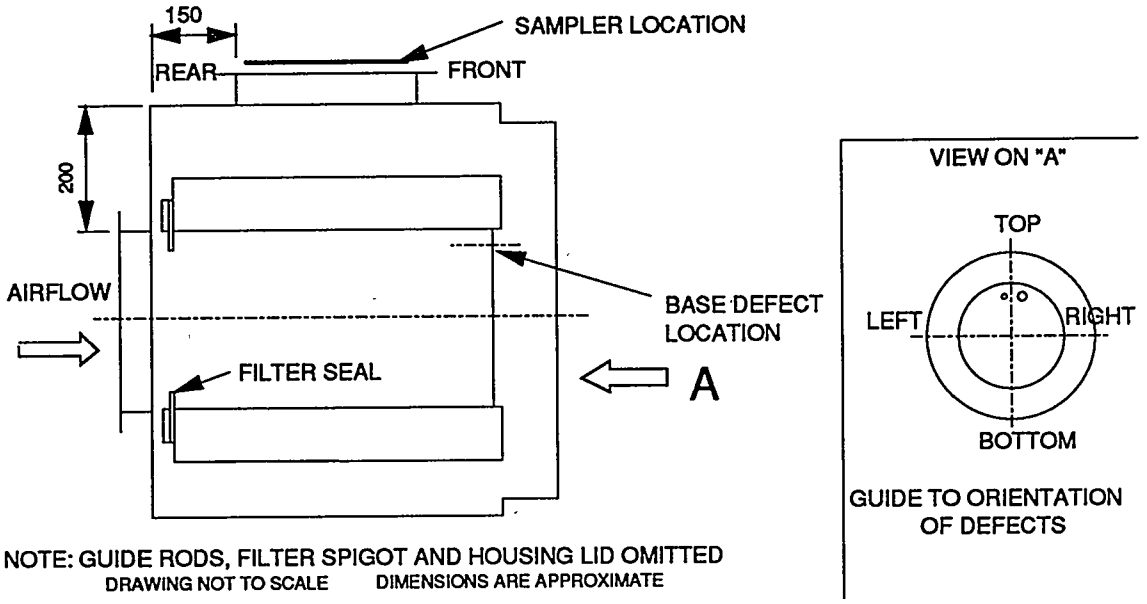
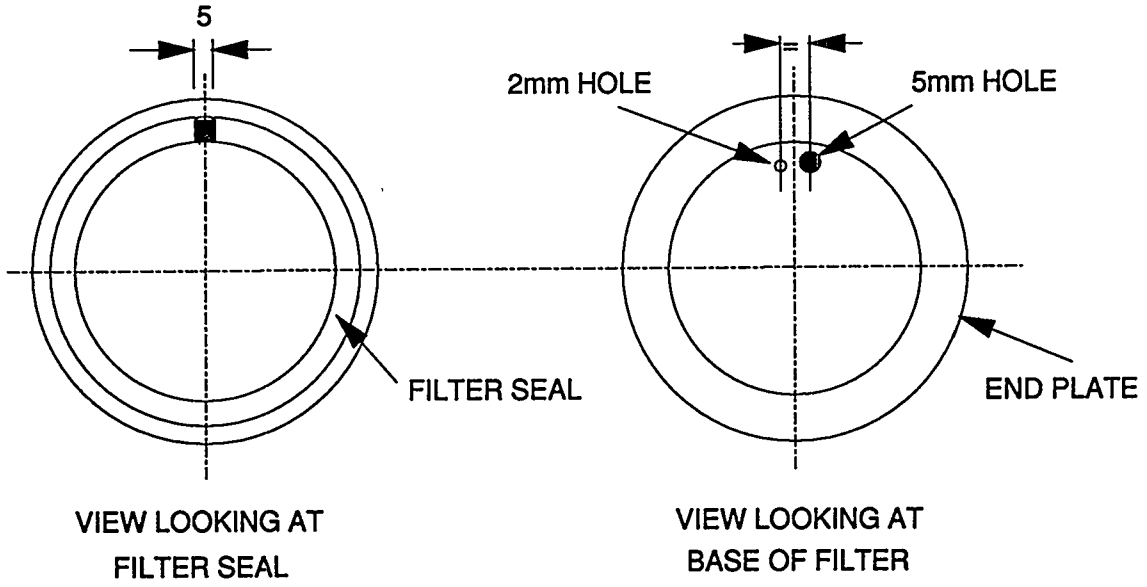


FIGURE 7: DETAILS OF ADDED DEFECTS



Figures 6 and 7 detail the added defects and their location, which have been chosen in order that the sampler is subjected to some of the most difficult leaks to detect re: a seal defect and a simulated defect in the media at the base of the filter. Piercing the media was not considered to be a suitable method for ensuring a consistent leak as its size and penetration will not be constant, hence the defect - one 2 mm and one 5 mm hole - are actually located in the base of the filter.

A second filter had a 5mm split cut into its seal to give a penetration of approximately 0.05%. The "standard" penetration values for each of the two defects (base and split seal) in 4 orientations designated, "top", "bottom", "left" and "right" as shown in Figure 6 are detailed in Table 6.

TABLE 6. Standard Efficiency Test Results

Defect	Orientation	Penetration (%)
As New		0.0025
Base Leak		
2.0	Top	0.0065
5.0	Top	0.030
2+5	Top	0.036
2+5	Right	0.044
2+5	Bottom	0.040
2+5	Left	0.048
Split Seal		
	Top	0.026
	Right	0.072
	Bottom	0.048
	Left	0.048

The highest result (0.072% - split seal) was initially thought to be due to the size of the split varying as the filter was rotated (off the spigot). Re-testing however confirmed this value and because, in relative terms, it is considerably higher than the 3 others recorded it is likely to be a good indicator of the sensitivity of any chosen sampler (see Table 8).

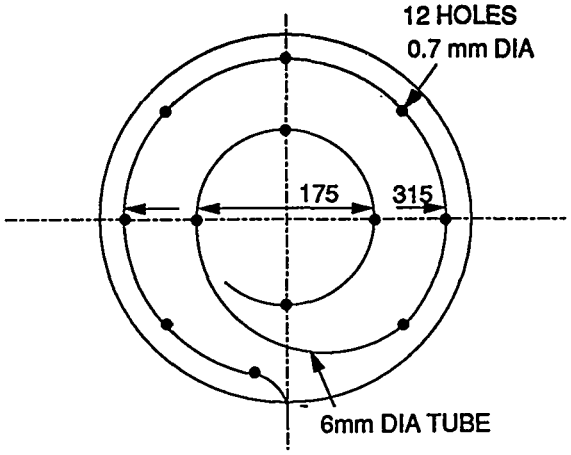
Currently available Multi-Point Samplers

Testing was initially carried out on two currently available multi-point samplers developed for other specific applications. One sampler[9] (Type A), consists of 6mm diameter tubing with 12 holes 0.7mm diameter spirally wound as shown in Figure 8. The sampler has been shown to be effective at distances of about 4 duct diameters downstream of the filter face.

The second (Type B), shown in Figure 9, consists of 15mm diameter tubing with 5 holes 3mm diameter on a 250mm PCD.

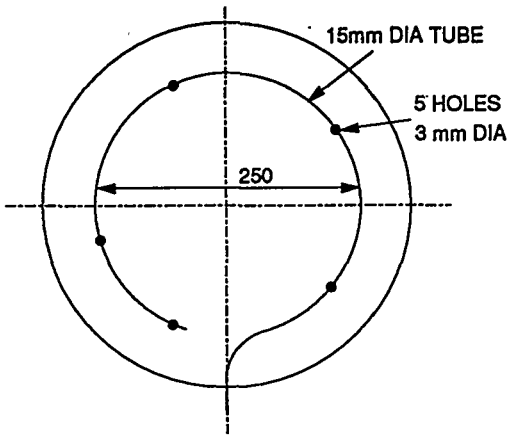
Penetration results for these are shown in Table 7 below. The Type A sampler has only 3 of the 8 values recorded showing a deviation greater than +/-0.02%. However, all 3 are below the actual value and give an optimistic penetration reading which, for an in-situ efficiency test, is not recommended.

FIGURE 8: TYPE "A" SAMPLER



Duct diameter = 350 mm

FIGURE 9: TYPE "B" SAMPLER



Duct diameter = 350 mm

TABLE 7. Penetration Results for Currently Available Samplers

Defect	Position	Standard Result	Type A Sampler		Type B Sampler	
			Pen (%)	Error (+/- %)	Pen (%)	Error (+/- %)
Base Leak						
2+5	Top	0.036	0.032	-0.004	**0.19**	**+0.154**
2+5	Right	0.044	0.047	+0.003	**0.18**	**+0.136**
2+5	Bottom	0.040	0.021	-0.019	0.034	-0.006
2+5	Left	0.048	**0.012**	**-0.036**	0.033	-0.015
Split Seal						
	Top	0.026	0.014	-0.012	0.008	-0.018
	Right	0.072	**0.036**	**-0.036**	**0.020**	**-0.052**
	Bottom	0.048	0.033	-0.015	**0.016**	**-0.032**
	Left	0.048	**0.027**	**-0.021**	**0.019**	**-0.029**

The alternative sampler with 5 out of 8 results deviating by more than +/-0.02%, the largest being +0.154% (a 0.036% penetration was recorded as 0.190%) is obviously unacceptable for this particular application.

Concentration Testing at the Sample Position

Concentration testing at the sample position was carried out to determine the area(s) of high aerosol concentration relating to a specific defect. This was intended to give an idea of the overall coverage of the duct cross section required by a multi-point sampler and to show the degree of streaming, if any, of the leakage aerosol. It also gave an insight into the reasons for the high/low readings of the two samplers tested to date.

Figures 10 and 11 show the results of this testing using the 2+5 and split seal defects when located in 4 orientations by scanning across the duct section using a single point sampler. Distinct evidence of streaming can be seen. Adding these areas together as shown in Figure 12 gives an idea of the minimum coverage required by any sampler.

An examination of the results in more detail gave some indication as to the reasons for the discrepancies noted when testing two samplers. For the Type B sampler with the 2+5 defect in the top and right positions - the highest recorded penetration values - it is likely that 2 of the 5 sampling holes (40%) were situated in areas of high concentration. This is illustrated in Figure 13. Similar arguments can be made for the low recorded values.

FIGURE 10: AREAS OF HIGH CONCENTRATION - BASE LEAK

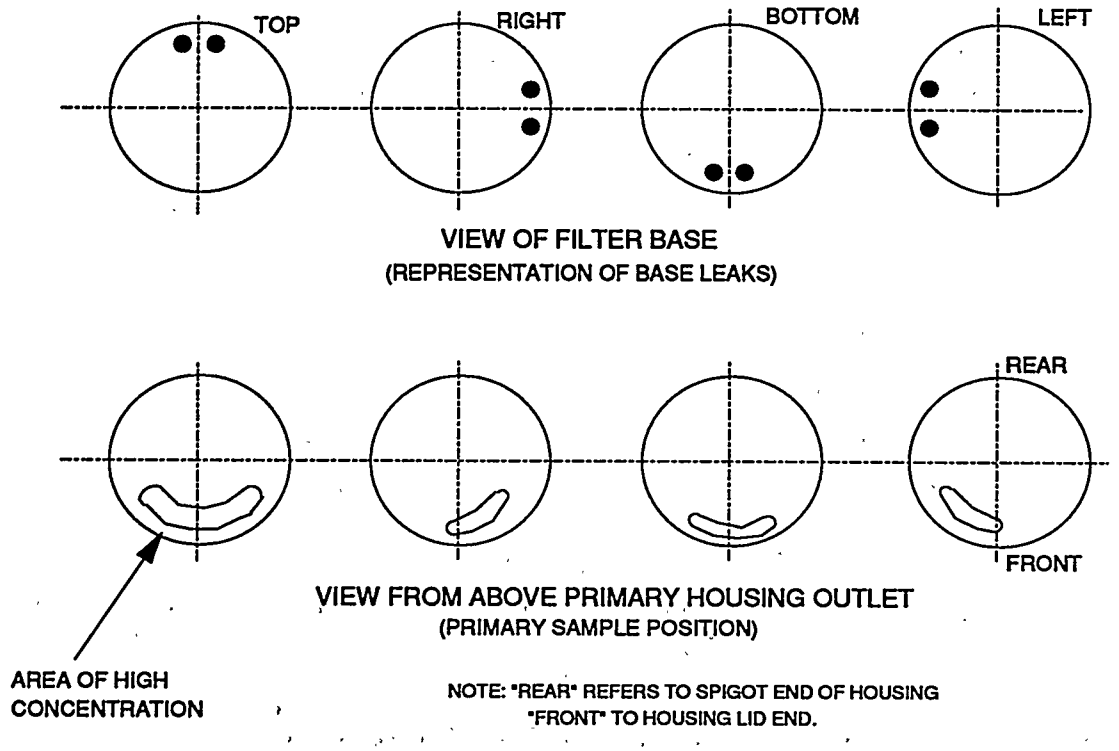


FIGURE 11: AREAS OF HIGH CONCENTRATION - SEAL LEAK

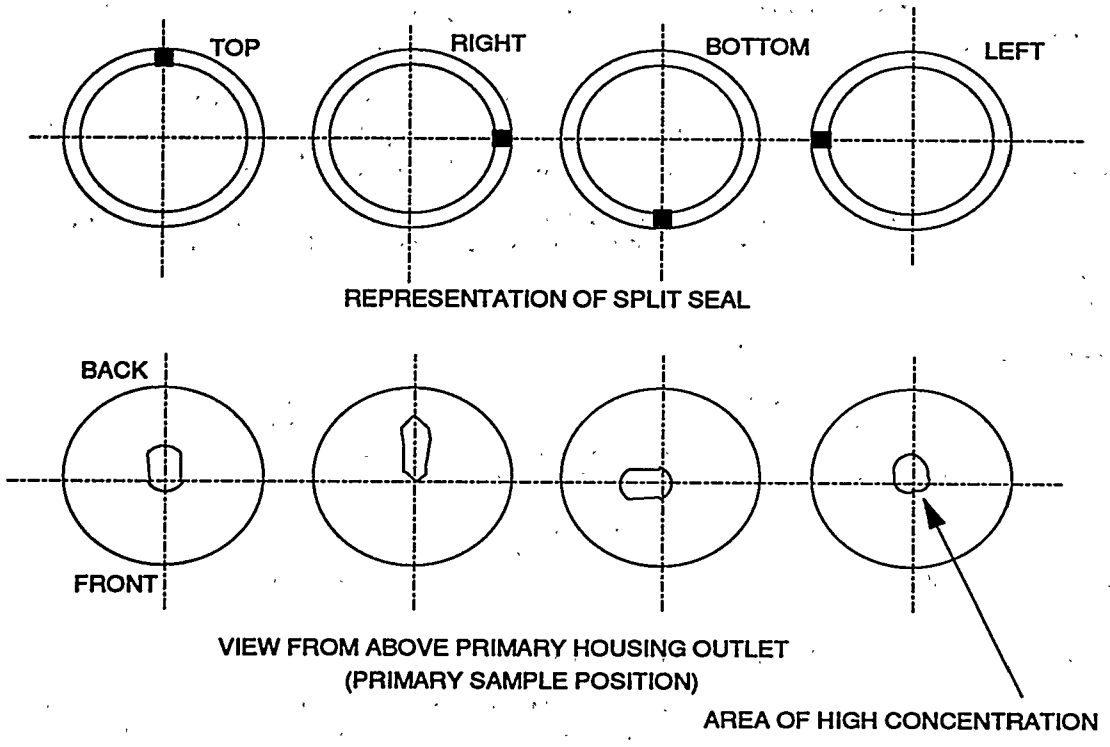


FIGURE 12: COMBINED AREAS OF HIGH CONCENTRATION

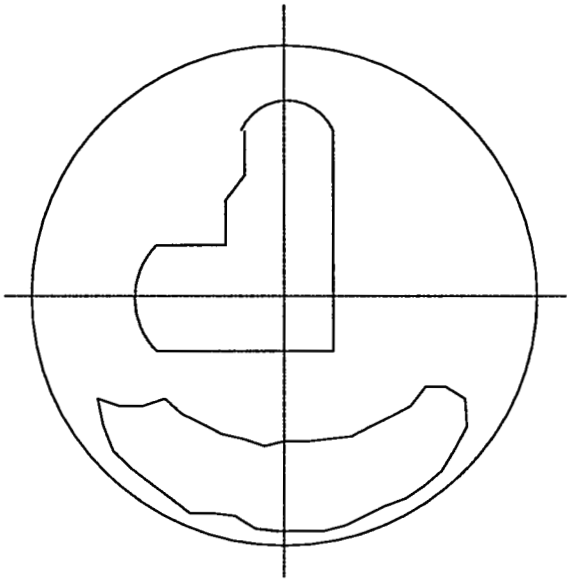


FIGURE 13: EXISTING SAMPLERS vs AREAS OF HIGH CONCENTRATION

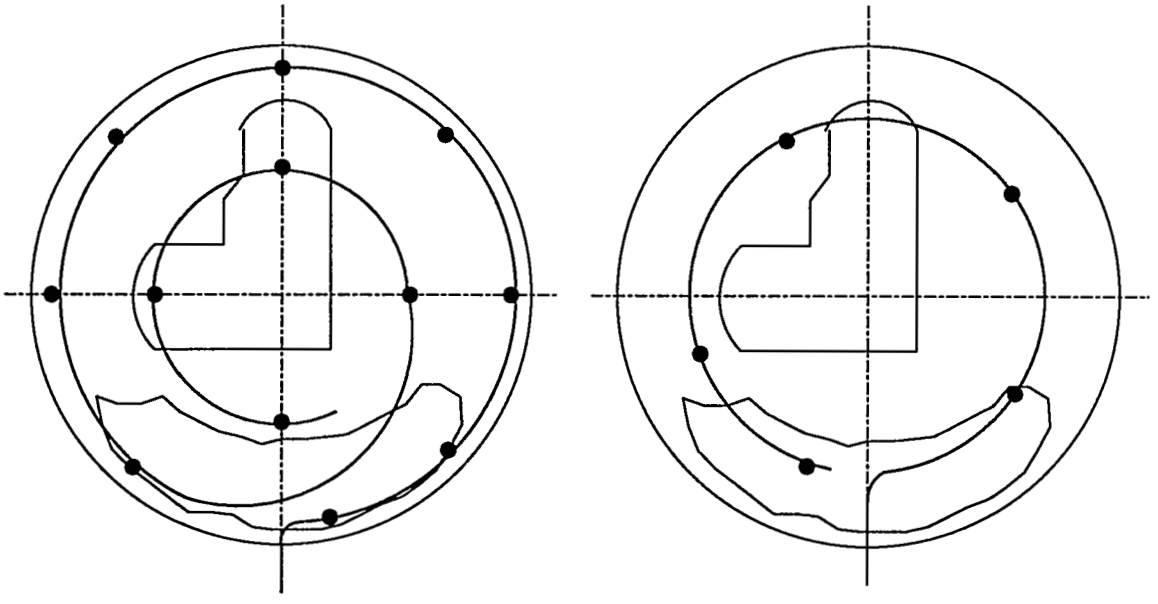


Figure 13 also illustrates the layout of the Type A sampler compared to all the identified areas of high concentration and shows the outer of the two rings (PCD of 315mm) lying on the very edge of the high concentration area.

It is apparent that a sampler with a single ring of sampling holes will not provide adequate coverage of the sampling area. It also evident that any new sampler should have two sampling rings and that the outer ring should lie on a diameter of less than 315 mm.

Testing of New Samplers

Following further development work and assessment the sampler shown in Figure 14, designated AEA II, was fully tested. It is manufactured from 15 mm diameter tubing, with 12 holes of 3 mm diameter in two concentric rings. Results for the penetration tests carried out using the AEA II sampler are shown in Table 8, below.

TABLE 8. Penetration Results for the AEA II Sampler

Defect	Position	Standard Result	AEA II Sampler	
			Pen (%)	Error (+/- %)
As New		0.0025	0.0027	+0.0002
Base Leak				
2	Top	0.0065	0.0043	-0.0022
5	Top	0.030	0.011	-0.019
2+5	Top	0.036	0.019	-0.017
2+5	Right	0.044	0.045	+0.001
2+5	Bottom	0.040	0.026	-0.014
2+5	Left	0.048	0.052	-0.004
Split Seal				
	Top	0.026	0.034	+0.012
	Right	0.072	0.073	+0.001
	Bottom	0.048	0.052	+0.004
	Left	0.048	0.049	+0.001

The measured penetration of the clean filter is extremely close to that of its actual value. With the base or split seal defect no recorded penetration varied by more than +/-0.02% from the actual value, in fact only 3 results deviated from the actual value by more than +/-0.005%. The layout of the sampler compared to the areas of high concentration is also shown in Figure 14

In terms of the overall requirements of this system the AEA II sampler is considered acceptable. In addition these results indicate that it may be suitable for use in other systems where circular filter housings and 350 mm ductwork are employed.

VI. FUTURE DEVELOPMENTS

There is an outstanding requirement to develop a multi-point injection system such that a satisfactory in-situ test can be made on the secondary filter. Initial work has concentrated on fitting 2 multi-point injector tubes into the air collection annulus downstream of the primary filter but within the primary filter housing (see Figure 15).

In such a system the concentration of aerosol challenging the secondary filter is measured by the installed multi-point sampler used for the primary filter downstream measurement.

To date such a system has been developed which is capable of achieving an aerosol concentation variation (at the sample point) within +/- 30%. Further work is required to enable further development and verification of this design approach to be carried out.

FIGURE 14: AEA II SAMPLER vs AREAS OF HIGH CONCENTRATION

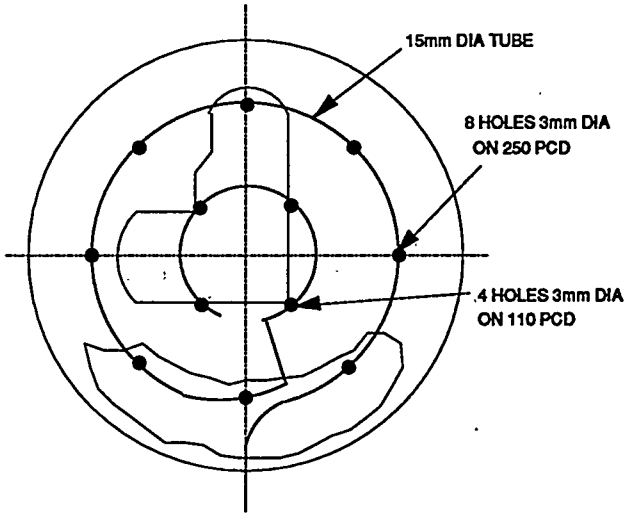
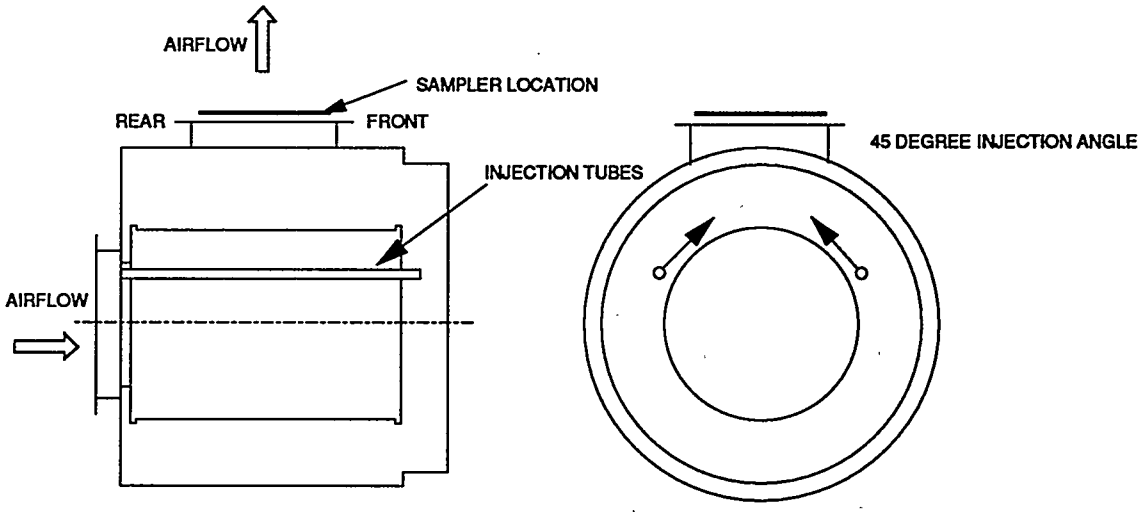


FIGURE 15: LOCATION OF SECONDARY FILTER UPSTREAM INJECTOR



NOTE: GUIDE RODS AND HOUSING LID OMITTED
DRAWING NOT TO SCALE DIMENSIONS ARE APPROXIMATE

VII. CONCLUSIONS

The following conclusions can be drawn from this work:

Basic Sytem Characteristics

1. The fume cupboard extract system offers a compact design with acceptable pressure drop and velocity profiles with a developed injection and sampling system suitable for testing the Primary filters in position.

Multi-Point Injection

1. To achieve adequate mixing in about 8 duct diameters a 22mm diameter 5 legged, 25 point injector is acceptable in 600mm square ductwork at duct air velocities between 4 and 10 m/s.

2. The most suitable injection hole diameter for such an injector is 3mm.

Developing a Primary Filter Test Method

1. The AEA II sampler, Figure 14, had over 50% of measured penetration values within +/-0.005% of the actual penetration. None were outside the +/-0.02% requirement.

2. The AEA II sampler is a suitable device for measuring, to within an acceptable level of accuracy, the penetration of filters when positioned at or very close to the outlet of a circular filter housing.

Future Development

1. More work is required to develop a suitable injector for in-situ efficiency measurements for the secondary filters.

IIX. ACKNOWLEDGEMENTS

Acknowledgements are given to Mr R.P Pratt who was the Project Manager in charge of the design and installation of this active facility and to Miss R. French who assisted in the practical work reported here.

IX. REFERENCES

(1) Atomic Energy Code of Practice (AECP) 1054 April 1989 "Ventilation of Radioactive Areas" - Provisional.

(2) Atomic Energy Standard Specification (AESS) 30/95100 June 1987 "Filter inserts, high efficiency particulate air (HEPA) circular plug-in 470 and 950 l/s capacities" (Note: re-issued Feb 1990)

(3) AESS 72001 Feb 1990 "Filter housings for circular plug-in HEPA filter inserts (internal seal) 950 l/s capacity"

(4) D. Loughborough - unpublished work, 1987/8.

(5) AEA EE 0030 "Investigations and experience related to in-situ filter testing using DOP or Ondina-EL aerosols and photometric detection" D.C Stevens, R.C Parker, D. Loughborough and RB Bosley, AEA Technology.

(6) "Development of Filter Systems - Injection Systems and Mixing Device Evaluation" 20th DOE/NRC Nuclear Air Cleaning Conference, Boston, USA. Paper 13-7. Aug 1988 D. Loughborough

(7) MoD - Not for Publication

(8) BNFL - Not for Publication

(9) "The mixing of Aerosols during in-situ filter testing" European conference: Gaseous effluent treatment in Nuclear installations Oct 1985 J.L Cunningham & Ms B.L Green UKAEA Harwell.

DISCUSSION

HOMICH: If velocity distribution was measured between filters, was it correlated with concentration found with multiple point sampler?

LOUGHBOROUGH: Yes, velocity profiles were taken across the sample position. I do not have the information immediately available. Since I do not recall the exact results, I must assume that such readings are not significant.

THE DUST HOLDING CAPACITY OF HEPA FILTERS

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ABSTRACT

The choice of HEPA filter for main extract ventilation systems, with the advent of large circular filters rated at 3400 m^3/h and the development of a 1700 m^3/h 0.125m (5") deep panel filter, is no longer the relatively simple choice it was only five years ago. Rapidly increasing waste storage and disposal costs in the United Kingdom coupled with this greater variety has significantly increased the interest in filter dust holding capacity.

Work carried out at the Harwell Laboratory and reported here, investigates the dust holding capacity versus pressure drop characteristics of four filter designs. Two test dusts - carbon black, and sodium chloride - have been used to load filters at volume flowrates ranging from 850 m^3/h up to 3400 m^3/h (500 - 2000 cfm).

The work identified that many deep pleat filters had a significantly reduced dust holding capacity to those available in 1984/85. It is further shown that both the circular and deep pleat filters perform significantly better than the mini-pleat and panel filters in terms of dust holding capacity when used as primary filters. The disparity between the dust holding capacity of the various filter types indicates that plant designers should give careful consideration to the loading conditions anticipated before recommending the use of high capacity mini-pleat or panel filters in primary plant locations.

When used as secondary filters all four filter types appear similar in terms of dust holding capacity. Other factors such as unit costs, volume flowrate and ease of installation must therefore be taken into account when considering the optimum choice for such a plant location.

I. INTRODUCTION

Currently all High Efficiency Particulate Air (HEPA) filters used by the UKAEA and British Nuclear Fuels plc are purchased against specifications[1,2,3], which details requirements for materials of construction, basic design and general performance criteria. Whilst the specifications stipulate a "minimum effective area of filtration medium" for a given volume flowrate of air (equivalent to stating a maximum velocity through the medium) they do not require evaluation or performance testing with regard to the life of the filter.

The advent of large circular filters rated at 3400 m^3/h with all their well published advantages[4,5], and the recent development of a 1700 m3/h, 0.125 m (5") deep panel filter, has made the choice for main extract ventilation filters no longer the simple alternative between the traditional deep pleat filter rated at 1700 m^3/h and the high capacity mini-pleat filter rated at 3060 m^3/h. This greater variety coupled with the increasing financial and technical implications of radioactive waste treatment, storage and disposal has significantly increased the interest in the dust holding capacity of such filters.

Whilst the relevant advantages and disadvantages, in terms of dust holding capacity, between deep pleat and mini-pleat filters and the effect of the arrangement of the media have previously been investigated[6,7], a comparison between all four main types of ventilation extract filter had not been made.

The work reported here establishes and compares the dust holding capacity of these 4 designs of HEPA filter at rated flow and at additional flowrates in order that plant designers and engineers are better able to decide on the most appropriate design for a particular plant application.

II. SCOPE OF WORK

The only requirement laid down in current Atomic Energy Standard Specifications regarding aerosol collection is a strength requirement to withstand a minimum differential pressure (3 kPa) after dust loading. The quantity of aerosol and particle size is not specified.

The reason for this omission is that many parameters affect this characteristic, and a suitable test i.e. both meaningful and practical, is difficult to devise. Having chosen a test dust or dusts, a dispersion method, a loading rate and a maximum (or nominal filter change) pressure drop, then the relative dust holding capacity of different filters and filter designs can be determined and from this suitable comparisons made.

It is important to note that a single test/dust is unlikely to provide sufficient data to enable a full evaluation of any one filter design to be made. This is because in ventilation extract systems primary and secondary filters (ie filters in primary and secondary locations) are, in general, subjected to entirely different conditions, both in terms of overall dust loading, loading rate, particle size distribution and even flow rate.

Whilst it would be impractical, if not impossible, to devise a test that covers every and all situations, it is possible to envisage two tests that, in the general sense, simulate the two situations.

Since a filter in a secondary location will normally be subjected to a predominantly sub-micron challenge, it can be argued that a wholly sub-micron aerosol with a mean particle size in the range normally associated with the most penetrating particle size for HEPA media - 0.15 - 0.4 microns[8] would form the basis for a loading test in this application. A thermally generated sodium chloride aerosol is used in this test work.

In the case of a filter in a primary location, which would normally be subjected to a much wider range of particle sizes, then it can be argued that a carbon black test dust, - although nominally sub-micron in practice significant agglomeration occurs resulting in a much wider particle size distribution - may form the basis of a loading test, for comparison purposes as long as a standard dispenser is specified.

III. TEST EQUIPMENT AND FILTERS

Test Rig

The test rig, Figure 1, consists of circular ducting with an open inlet to enable dust to be dispensed, a standard filter housing and downstream ducting connected to an extract fan. Volume flowrate is measured by a standard orifice plate and controlled using a damper. Pressure differential across the filter is measured by a micromanometer.

A second housing specifically for large circular filters and conforming to the dimensions in the circular filter specification[2], was also used for this work. This special in-line housing - a Harwell development - adds less than 70 Pa (7 mm Wg) to that of the filter at 3400 m^3/h (2000 cfm).

Test Filters

For this work 4 designs of commercially available HEPA filter have been tested, namely:

a) Conventional deep pleat filters rated at 1700 m3/h (1000 cfm)

b) High capacity mini-pleat filters rated at 3060 m3/h (1760 cfm)

c) Large plug-in circular filters rated at 3400 m3/h (2000 cfm)

d) 0.125 m (5") deep panel filters rated at 1700 m3/h (1000 cfm).

Conventional deep pleat filters from 4 manufacturers, mini-pleat filters from 2 manufactures and circular filters from 1 manufacturer, conforming in most respects to the relevant Atomic Energy Standard Specification[1,2] have been tested for this work. In addition 2 manufacturers have supplied 125 mm (5") deep panel filters which conform in most respects to the deep pleat specification[1] except for the filter dimensions, which for a 1700 m^3/h panel filter are 610 x 610 x 125 mm (24 x 24 v 5").

All filters had a minimum efficiency of 99.95% to BS 3928[9] and have been manufactured with either Hollingsworth and Vose grade F39ZAE/R (formerly F39Z/R) or Lydall grade L3255 media. The basic features of the deep pleat, mini-pleat and circular are illustrated in Figures 2, 3, and 4. Figure 5 illustrates the differences in the size and spacing of the media pleats.

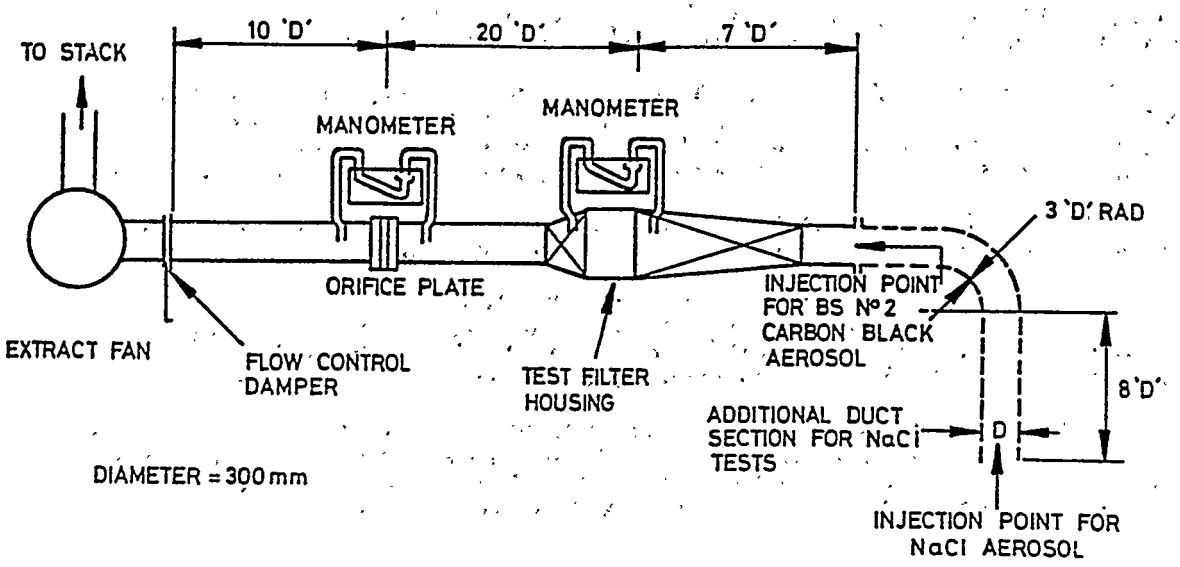


FIG 1. GENERAL ARRANGEMENT OF TEST RIG

- Standard unit
- Used since the 1940's
- Size: 610 x 610 x 292 mm (24"x24"x12")
- Flowrate: 1700 m^3/h (1000 cfm)

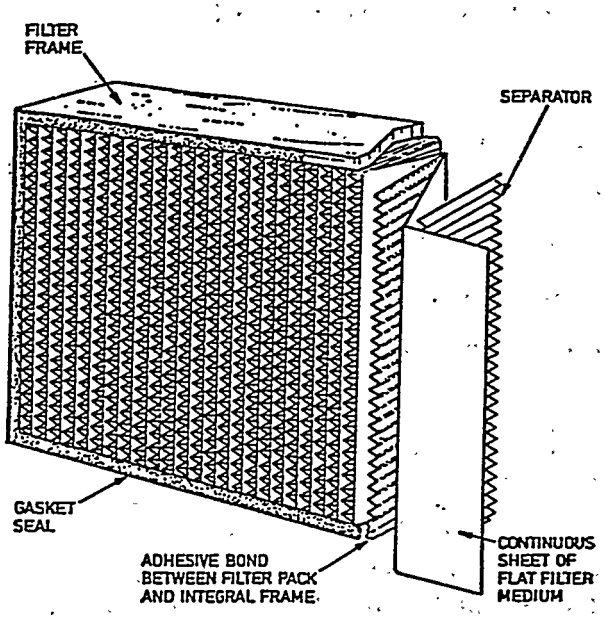


FIG 2. VIEW OF STANDARD 1700 m^3/h DEEP PLEAT FILTER

- Standard unit
- Size: 610 x 610 x 292 mm (24"x24"x12")

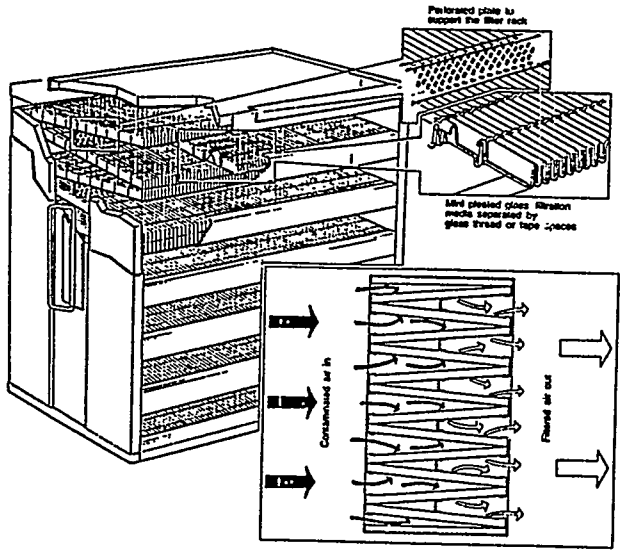


FIG 3. VIEW OF STANDARD 3060 m³/h MINI-PLEAT FILTER

- Standard unit
- Size: 534 O/D x 634 mm long
- Normal flow: In to out

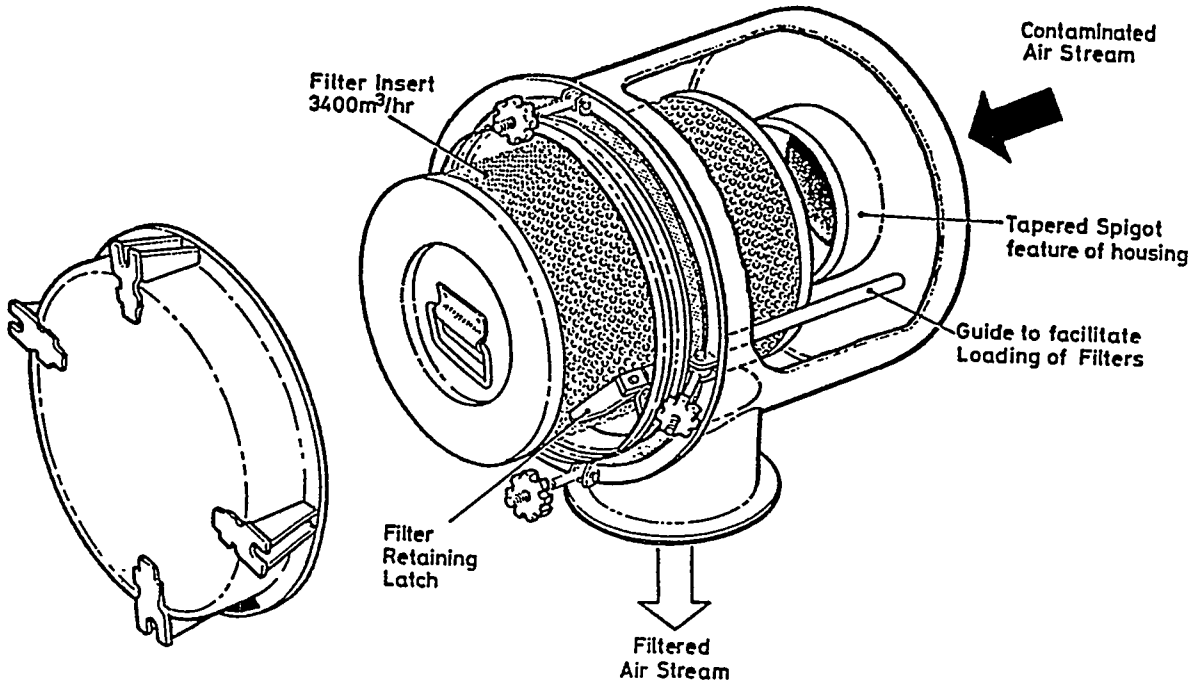


FIG 4. VIEW OF 3400 m³/h CIRCULAR FILTER & HOUSING

Test Aerosols

The two test aerosols used in this work are:-

a) Sodium Chloride with a MMD of 0.14 microns[10]. The CDE portable thermal generator used in this work produces the aerosol by vaporising sticks of sodium chloride in an oxy-propane flame and is capable of dispensing particulate material in the range 0.32 to 6.7g/min.

b) Carbon Black with a nominal MMD of 0.6 microns. Measurements carried out using an Anderson MK III impactor[11] gave a MMD of 2 microns (35% below 1 micron), which confirms the view from earlier work[6] that significant agglomeration of the particles occurs. The standard ASHRAE dispenser used is capable of dispensing carbon black in the range 0.3 to 2.4 g/min.

IV. CARBON BLACK LOADING RESULTS

All filters were tested at their rated flow. Additional tests were carried out to evaluate the effects of operating at flowrates above and below rated flow.

The conventional deep pleat filter - average media area of 20 m^2 - has been used as a standard in order to make useful comparisons between the various filter designs. In addition a nominal filter change pressure drop of 1000 Pa (4" Wg) is assumed.

Deep pleat filters

The results shown in Table 1 below and illustrated in Figure 6 show the extent to which nominally similar Deep pleat filters can vary in dust holding capacity.

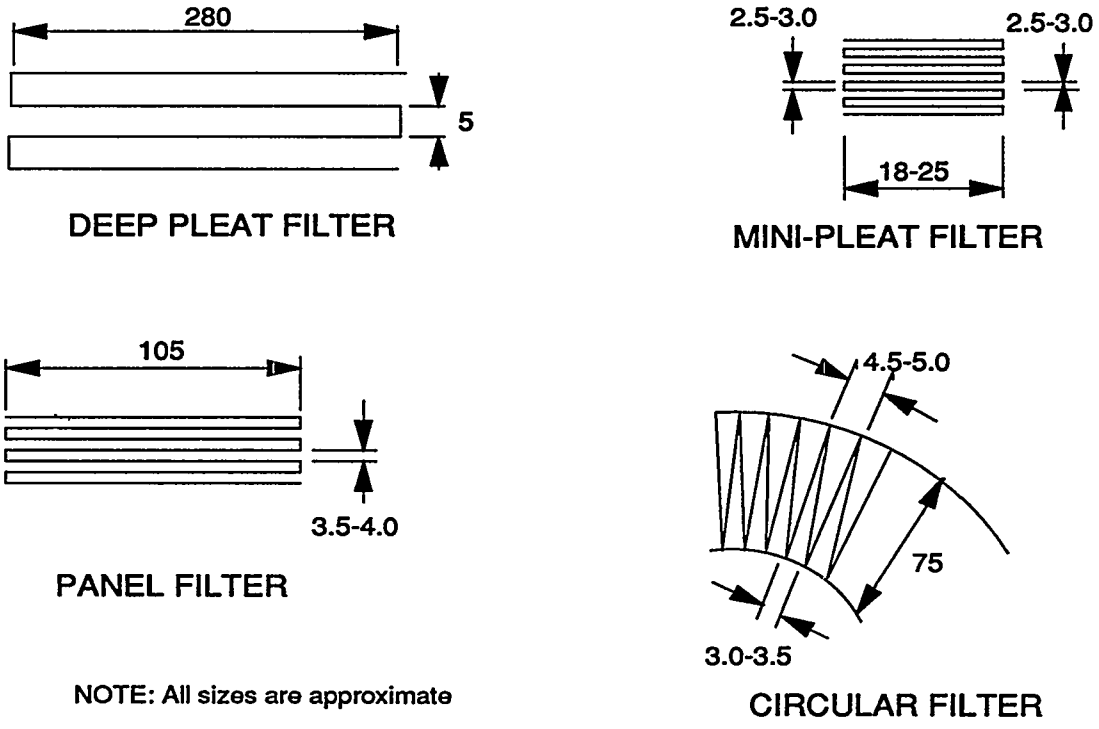
TABLE 1: Dust Load vs Pressure Drop: Carbon Black - Deep Pleat Filters

		Deep Pleat Filter Reference							
		A1	A2	B1	B2	C1	D1	D2	B3
Feed Rate	g/min	0.3	0.6	0.3	2.4	2.4	2.4	2.4	2.4
Medium	m^2	18.1	18.1	19.3	18.7	20.1	21.1	21.1	20.21
Test Flow	m^3/h	1700	1700	1700	1700	1700	1700	1700	850
grams		Pressure Drop (Pascals)							
0		245	235	225	230	230	200	205	110
60		320	310	295	280	280	240	245	135
120		400	400	360	360	360	286	280	170
180		465	500	440	440	440	340	330	210
240		620	615	550	540	540	400	385	250
300		750	745	670	650	650	455	455	290
360			900	750	750	750	540	535	350
420			1060			870	640	615	430
480			1180			1020	740	720	500
750						1170	1550	1440	970
900							2050	1840	1500
1050							2950	2560	
1200								3000	

Note: Filter media quantity values are approximate and refer to the effective media area.

Prefix to filter reference eg "A" denotes filter supplier

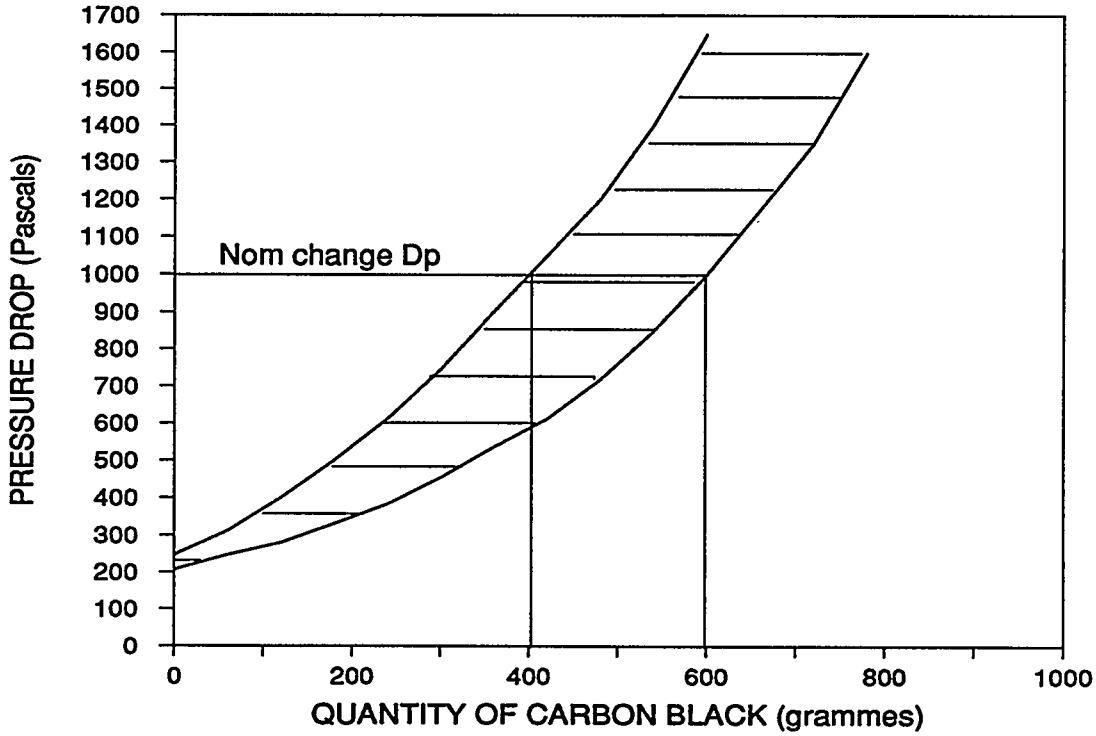
FIGURE 5: COMPARISON OF MEDIA PLEATS



NOTE: All sizes are approximate

FIGURE 6: DUST LOAD vs PRESSURE DROP

DEEP PLEAT FILTERS - CARBON BLACK LOADING



At a nominal change pressure drop of 1000 Pa (4" Wg) the dust holding capacity was found to vary from 380g to 600g (filters A2 and D1 respectively). Further examination of the data indicates the difference is related to relatively small variations in filter media area.

The current AESS minimum media area requirement is 18.8 m^2 for this type of filter. Filter D1 has 112% of the minimum required, whilst filter A2 has only 96%, the difference in practical terms is equivalent to 9 media pleats. As a result under these loading conditions filter A2 (18.1 m^2) has only 64% of the dust holding capacity of filter D1 (21.1 m^2).

If a comparison between these two filters were made at a pressure drop of 750 Pa (3" Wg) - 300 g and 480 g respectively - the reduction in dust holding capacity is 50%.

Two further important points are worth emphasising:

1. The variation recorded is large, but not as large as that recorded by Pratt/Green[6] who used filters which, at the time, were not subject to the same media area requirements. AESS specifications prior to 1984 required a minimum media area of 20 m^2 for a 1700 m^3/h filter.

2. More importantly there has been a significant overall reduction in the dust holding capacity of filters currently available (to the UK's nuclear industry) due to the reduced media area requirement of 18.8 m^2 from the 20 m^2 requirement specified prior to 1984.

The introduction of the current minimum media area requirement whilst ensuring that a "minimum standard" is achieved, has resulted in a general reduction in dust holding capacity and hence in-service filter life.

On the basis of these tests and earlier work[12] it has been recommended that the standard minimum (effective) area of filtration media be increased from 18.8 m^2 to 20.6 m^2. In practical terms this means an increase of approximately 6 pleats, from an average of 66 to 72 pleats per filter (1 pleat = 2 x single sheets).

High capacity mini-pleat filters

Results for the mini-pleat filter are shown in Table 2, see Figure 8, and indicate that in comparison to the average (20 m^2) deep pleat filter they have just over half the dust holding capacity. This confirms the view from earlier work[6] concerning the need for caution when specifying this type of filter for use in primary plant locations where this type of loading may be predominant.

TABLE 2: Dust Load vs Pressure Drop: Carbon Black - Mini-Pleat Filters

		Mini-Pleat Filter Reference						
		A3	A4	A5	D4	A6	A7	A8
Feed Rate	g/min	0.6	0.6	2.2	0.3	1.2	1.2	2.4
Medium	m^2	31	31	31	32	31	31	31
Test Flow	m^3/h	3060	3060	3060	3060	1700	1700	3400
grams		Pressure Drop (Pascals)						
0		255	260	260	310	135	150	380
60		315	320	320	380	165	187	490
120		404	410	400	475	210	235	690
180		550	550	520	615	285	310	960
240		750	760	765	790	410	420	1300
300		1220	1100	1150	1050	670	600	1800
360		1500	1500	1700	1450			
420		(330g)	(345g)		1900			
450					2500			
520					3000			

FIGURE 7: COMPARISON OF DUST HOLDING CAPACITY

DEEP PLEAT FILTERS - CARBON BLACK LOADING

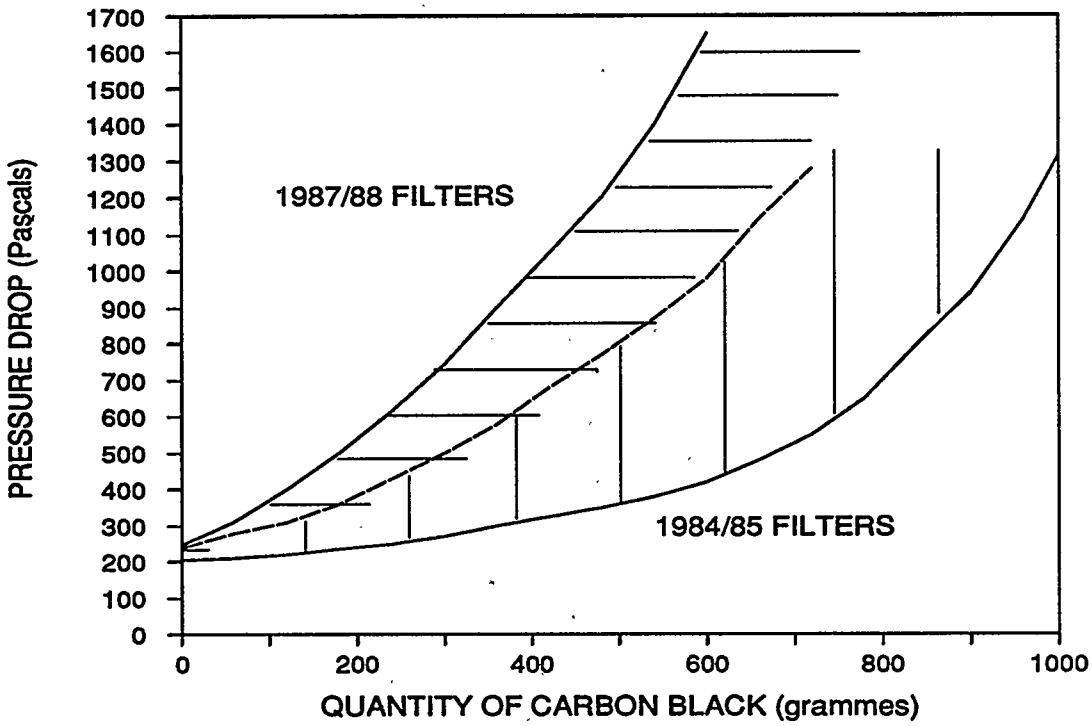
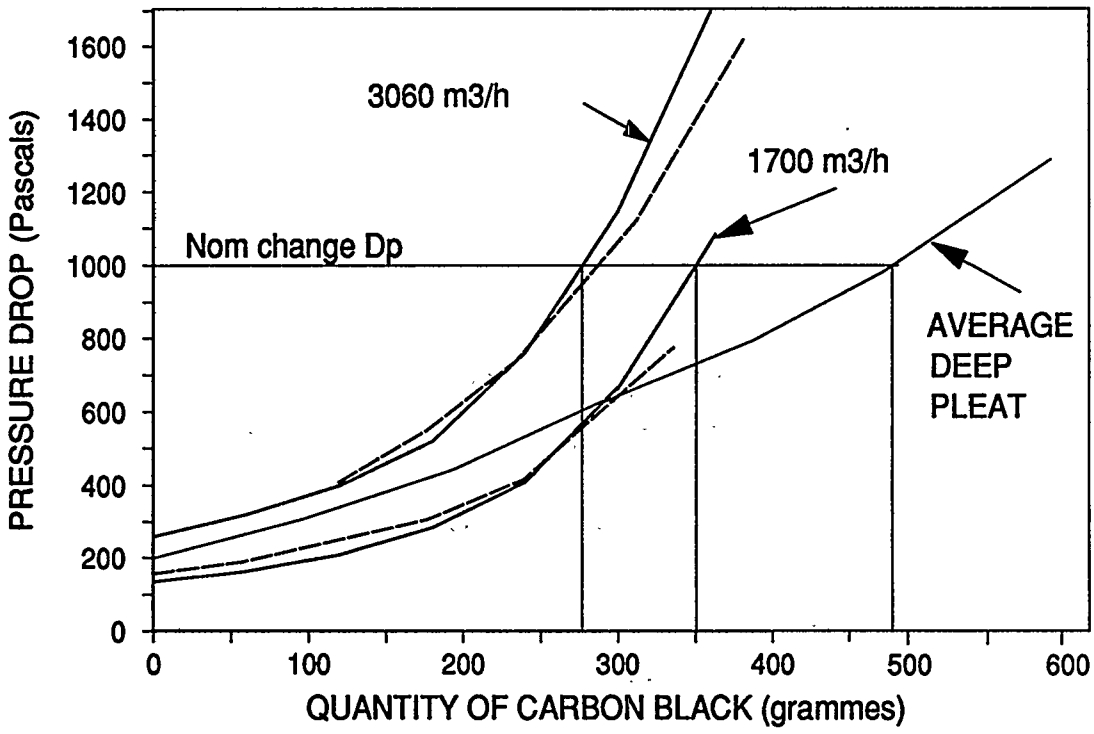


FIGURE 8: DUST LOAD vs PRESSURE DROP

MINIPLEAT FILTERS - CARBON BLACK LOADING



Operating the filter at 1700 m^3/h results in a dust holding capacity which is still only 75% of the (average) deep pleat filter, indicating that under these loading conditions - nominal primary loading - that this type of filter appears to offer no extension in filter life. It also has a penalty in terms of the additional cost of purchase as it is currently approximately 3 times more expensive than the deep pleat filter.

Panel filters

This particular design of filter has only recently been introduced in the 610 x 610 mm format using a 105 mm pleat depth (125 mm over the casing) and has a media area of approximately 19 m^2 and weighs approximately 9.5kg (21 Ibs).

The panel filter results shown in Table 3, see Figure 9, indicate that although they show a small (10%) increase in dust holding capacity over the mini-pleat filters tested at 3060 m^3/h, they are still substantially less effective than the deep pleat filter in terms of dust holding capacity.

TABLE 3: Dust Load vs Pressure Drop: Carbon Black - Panel Filters

		Panel Filter Reference			
		E1	A9	A10	A11
Feed Rate	g/min	2.4	1.2	1.2	2.4
Medium	m^2	20.7	21.2	17.5	20.7
Test Flow	m^3/h	1700	1700	1700	850
grams		Pressure Drop (Pascals)			
0		210	295	330	150
60		270	395	425	190
120		380	470	515	230
180		550	570	635	300
240		750	720	770	380
300		950	925	980	580
360		1270		1350	880
420		1930		1500	1350
480		2900			2000

This size of unit offers a volume flowrate of 1700 m^3/h (1000 cfm) with only one third of the volume of a conventional deep pleat/mini-pleat filter thus considerably reducing the volume of waste to dispose of. The unit costs are similar to the deep pleat and the consequent savings in storage and disposal costs, although not yet fully evaluated, may be substantial. Its lighter weight makes it easier to handle and therefore change, thus reducing the time of change and ultimately man exposure.

Improvements in dust holding capacity are essential if the benefits of this unit are to be fully exploited and before it is more readily accepted.

Circular Filters

The results of the circular filter loading are shown in Table 4, and Figure 10, which shows, at 3400 m^3/h, an almost identical dust holding capacity to that of the deep pleat filter.

Slight variations in the dust holding capacity between apparently identical circular filters are attributable to minor differences in design eg, modifications to the grille (from perforated plate to expanded metal) and the formation of the pleat.

FIGURE 9: DUST LOAD vs PRESSURE DROP

PANEL FILTERS - CARBON BLACK LOADING

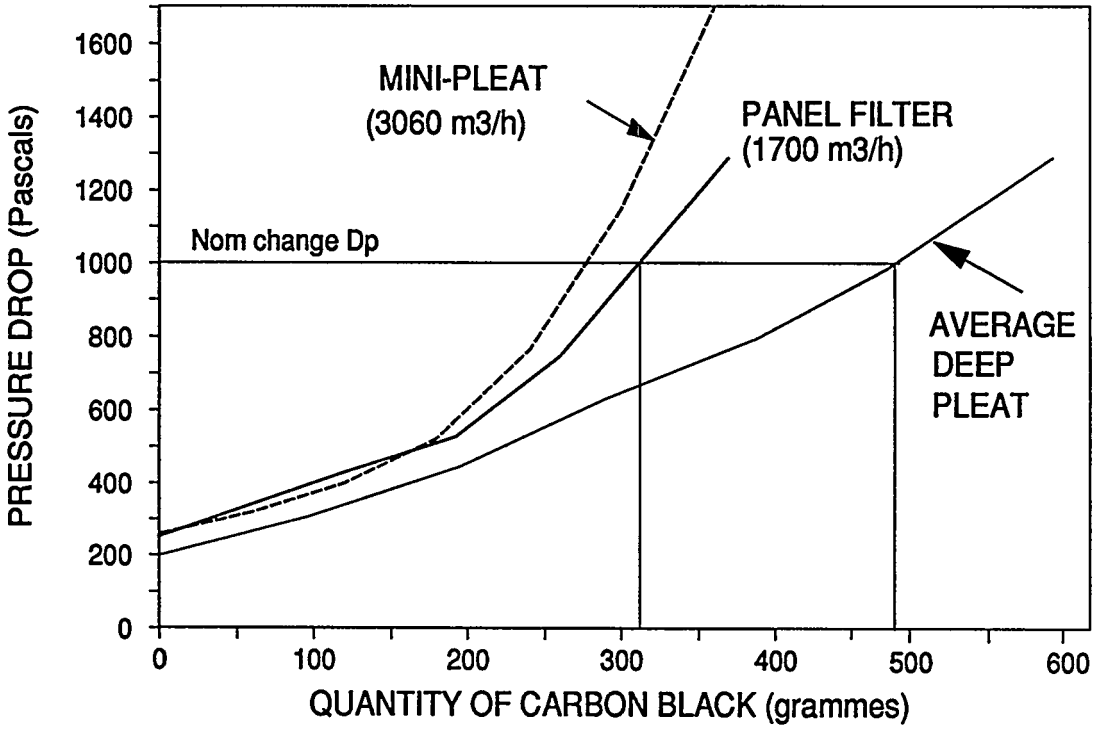


FIGURE 10: DUST LOAD vs PRESSURE DROP

CIRCULAR FILTERS - CARBON BLACK LOADING

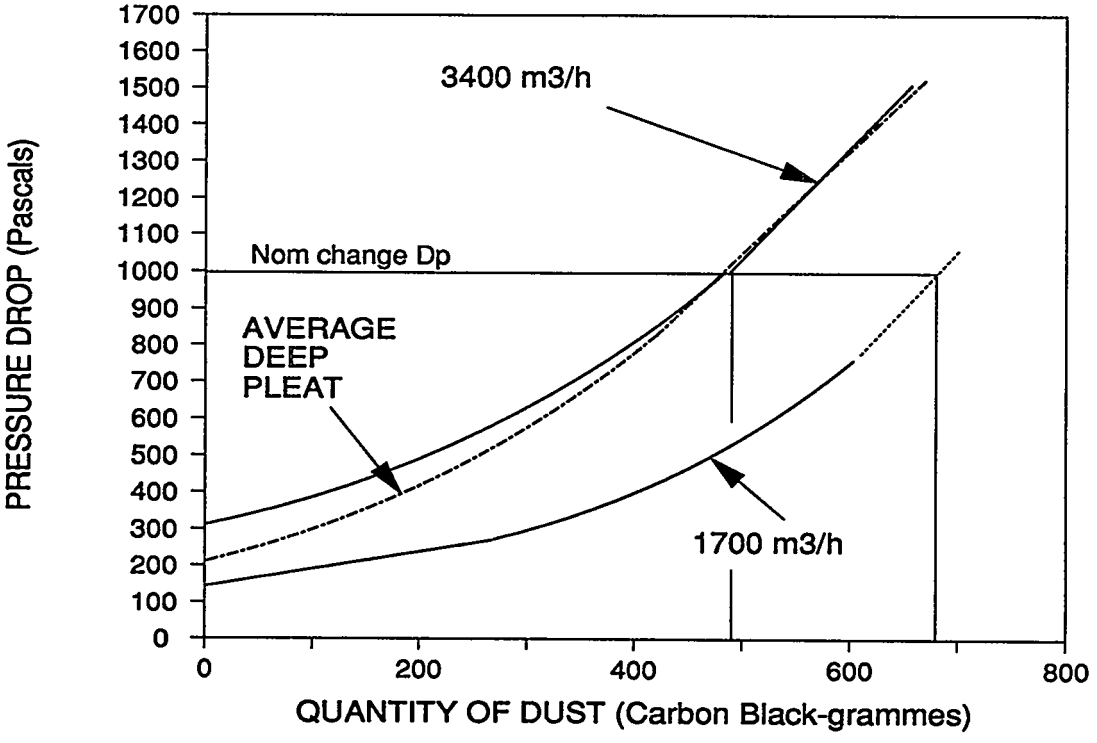


TABLE 4: Dust Load vs Pressure Drop: Carbon Black - Circular Filters

		Circular Filter Reference				
		A12	A13	A14	A15	Mk 3
Feed Rate	g/min	2.4	2.4	1.2	0.3	1.2
Medium	m^2	28.5	28.5	28.5	28.5	28.5
Test Flow	m^3/h	3400	3400	1700	1700	3000
grams		Pressure Drop (Pascals)				
0		310	300	152	143	200
60		370	360	175	169	240
120		430	420	195	200	280
180		490	480	216	224	340
240		570	540	252	270	450
300		665	620	290	305	650
360		760	720	330	365	1000
420		920	840	370	420	1450
480		1100	970	430	525	2000
540		1300	1150	500	620	2650
600		1500	1300	550	750	3200
660		1720	1500	665		
720		1900		750		
780		2200				
840		2550				
900		2900				
960		3200				

Figure 11 compares the current standard circular filter, pleat depth of 75 mm, with an early development circular filter, the MK 3 (1986), rated at 3000 m^3/h with a pleat depth of 55 mm. Although the difference in dust holding capacity is relatively small up to a pressure drop of 1000 Pa, when the filters are compared at 3000 Pa the difference is extremely large.

When compared to the deep pleat the MK 3 is equally unsatisfactory and would have made a poor, if not unacceptable, alternative.

Current circular filters combine the advantages of the mini-pleat design - high volume flowrate - with the high dust holding capacity of deep pleat filters. These two advantages plus a simple and easy to effect seal and their suitability for disposal (sized for direct placement in a disposal drum) give these units a distinct advantage when designing a ventilation system for new plant.

A comparison at various flowrates

Whilst all the filters have been directly compared to the average deep pleat filter at their rated flows a further comparison between filters operating at 3400 m^3/h, 1700 m^3/h and 850 m^3/h is discussed below.

Operating at 3400 m3/h. For a ventilation extract system which requires primary filters to operate at 3400 m^3/h (2000 cfm) a direct choice between circular and mini-pleat filters must be made. Whilst the units cost are approximately the same the circular filter exhibits, under these loading conditions, a filter life of over 100% greater, see Figure 12.

Whilst a full comparison requires other factors such as plant size, cost of housings/ductwork, ease of sealing etc to be considered the case for using circular filters - based on dust holding capacity alone - is a strong one. This is particularly so when the current trend, to reduce man exposure and minimise waste arisings (of which the HEPA filter may be a relatively large proportion) is taken into account.

FIGURE 11: COMPARISON OF TWO CIRCULAR FILTERS

CARBON BLACK LOADING

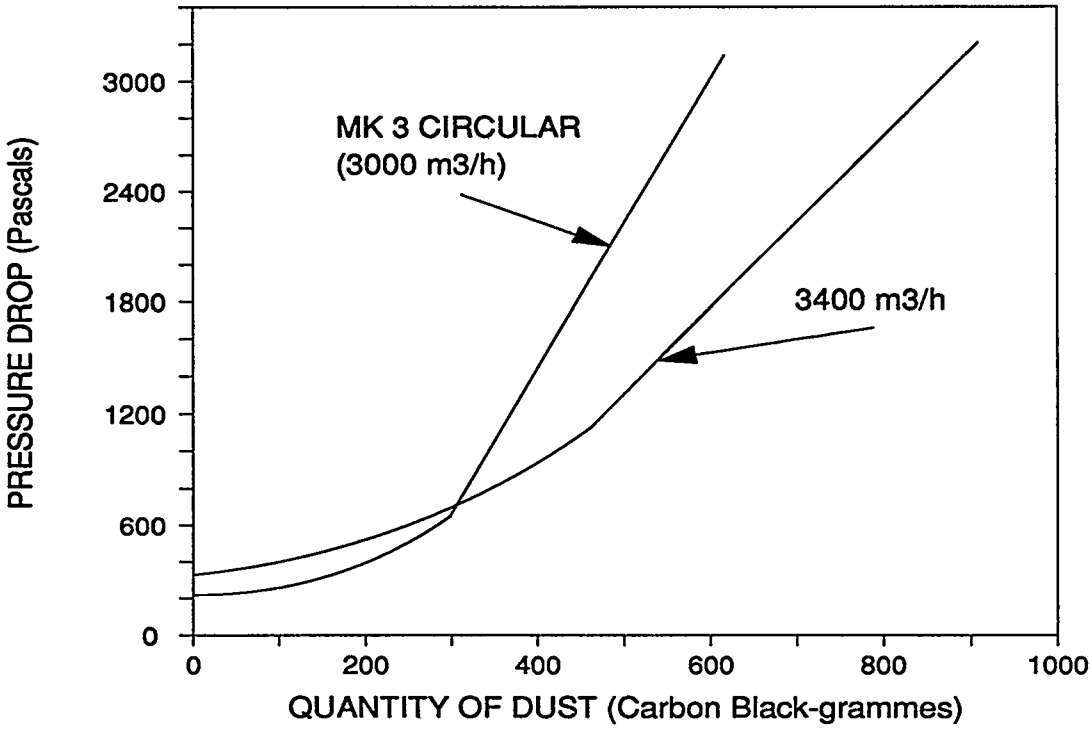
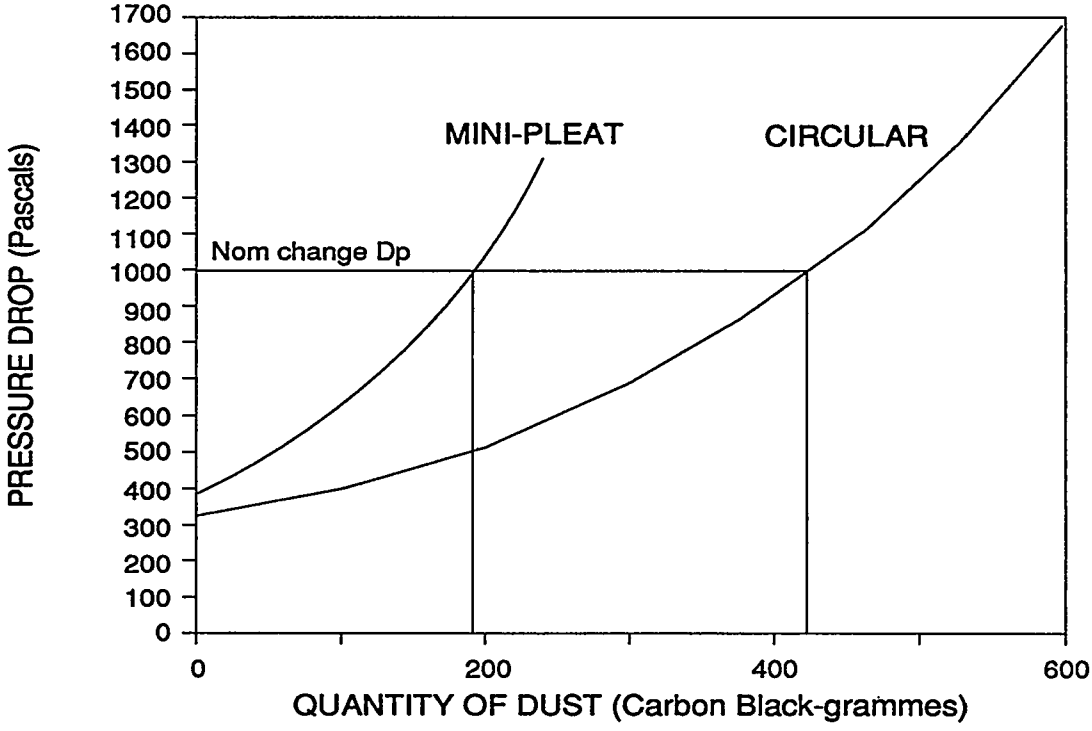


FIGURE 12: COMPARISOM AT 3400 m^3/h (2000 cfm)

CARBON BLACK LOADING



Operating at 1700 m3/h. At a system flow rate of 1700 m^3/h (1000 cfm) the choice is more varied as both mini-pleat and circular filters are included. If the average deep pleat is again taken as standard both panel and mini-pleat filters would have - under these loading conditions - only half the anticipated in-service life, see Figure 13. At this flowrate the circular filter shows a 40% increase over the deep pleat filter and should be able to stay in-service over twice as long as both the panel and mini-pleat filters.

Where a high dust holding capacity is required, perhaps where it is essential to prolong the time between filter changing, then the circular filter appears to be the best option.

In existing primary plant locations designers and/or plant managers may be tempted to specify mini-pleat filters, to replace deep pleat filters being run at 1700 m^3/h, with a view to extending the time between filter changing. The results of this test work suggests that unless the filter change pressure drop is substantially below 500 Pa, which is the point at which the dust load vs pressure drop curves of the two filters cross, see Figure 13, then it would not be recommended. Above 500 Pa and the frequency of filter changing would increase.

Operating at 850 m3/h. Figure 14 compares the dust holding capacity of the deep pleat and panel filters when operated at 850 m^3/h. At the nominal change pressure drop of 1000 Pa the deep pleat has a dust holding capacity of twice the panel filter indicating that for ventilation extract systems operating at this flowrate the deep pleat is the most viable option.

V. SODIUM CHLORIDE LOADING RESULTS

Table 5 details the results of loading the four filter types using the sub-micron sodium chloride aerosol, Figure 15. The absolute dust holding capacity of all the filters is considerably lower than those obtained using carbon black with, on average, only 120g needed to reach the nominal change pressure drop of 1000 Pa (filters at rated flow). This average, which is also the mini-pleat result at 3060 m^3/h, is comparable with those reported elsewhere[6].

The results indicate that operating the deep pleat filter at half rated flow 850 m^3/h (500 cfm) - and obviously half the face velocity - doubles the filters life. Increasing the rated flow by 50% to 2550 m^3/h (1500 cfm) - a 50% increase in face velocity - decreases the filter life by half. These results confirm the earlier work by Pratt[6].

However, when comparing the dust load per unit area of the deep pleat at rated flow, (8.4 g/m^2), it is double that recorded by the three other filter designs when operated at approximately the same face velocity. The 170g of dust needed to increase the pressure drop to 1000 Pa is consequently double that recorded by Pratt[6].

The increase in effectiveness may be related to electrostatic effects and the quantity of aluminium in a normal deep pleat (almost equivalent to the paper area) and to the surface coating/finish of the aluminium. Further work in this area is required and may involve testing filters with non-metallic spacers.

The mini-pleat and circular filters are very similar under these loading conditions but only have a dust holding capacity of 75% of the deep pleat filter (A12), although the circular offers the benefit of twice the volume flowrate.

The panel filters require only 80g of sodium chloride dust to reach 1000 Pa and have only 50% of the dust holding capacity of the deep pleat filter.

It is possible to make further comparisons with the earlier carried out by Pratt[6]. In respect of this work the following points can be made:

- The mini-pleat and panel filters operating at a face velocity of approximately 1.62 both achieve a dust load per unit area in the 4.0 - 4.5 range which is identical to the results reported by Pratt[6].
- The differences recorded in the mini-pleat filters tested, a reduction in face velocity from 1.65 to 0.91, increased the dust load per unit area by a factor of 2.5 is also in agreement.

FIGURE 13: COMPARISON AT 1700 m^3/h (1000 cfm)

CARBON BLACK LOADING

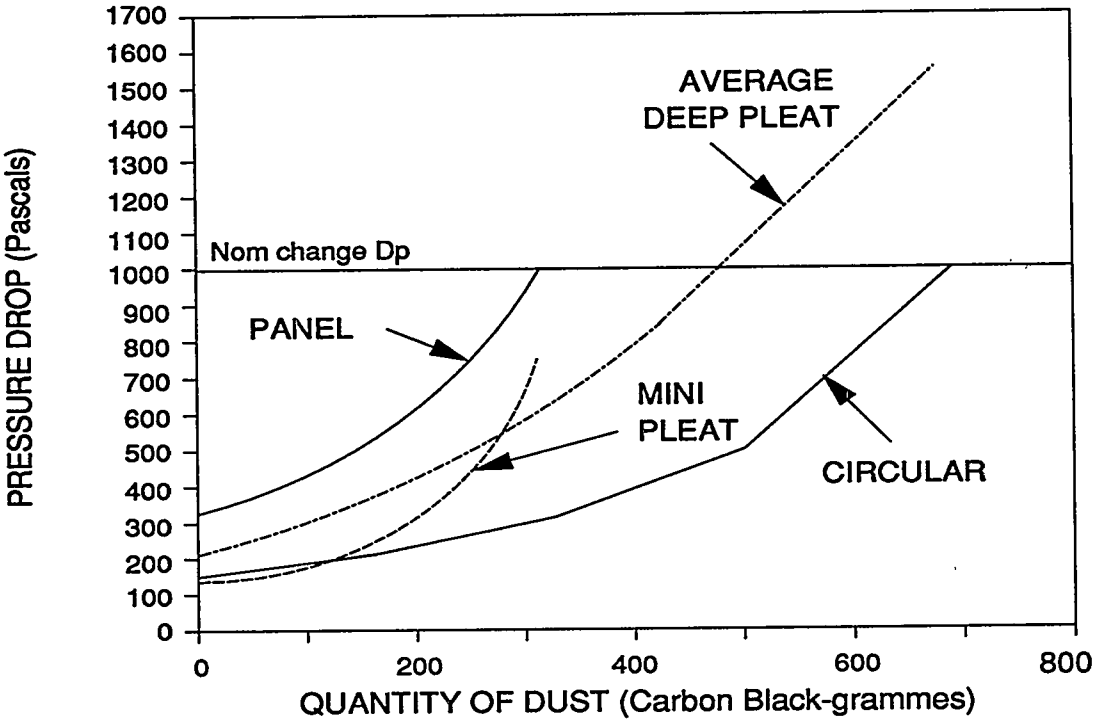
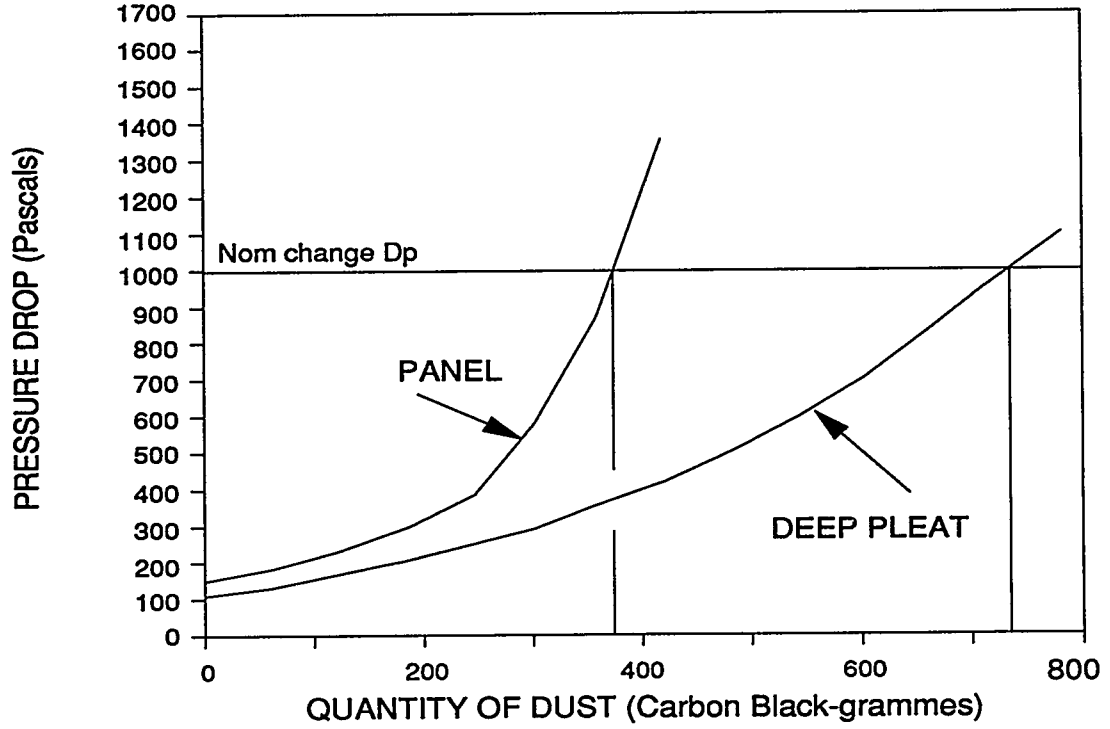


FIGURE 14: COMPARISON AT 850 m^3/h (500 cfm)

CARBON BLACK LOADING



- The two circular filters, one tested at twice the face velocity of the other had a dust load per unit area differing by just over a factor of two, which is also in agreement with the general findings of this earlier work.

TABLE 5: Dust Load vs Pressure: Sodium Chloride - Various Filters

	Filter Reference								
	DEEP PLEAT			MINI PLEAT		CIRCULAR		PANEL	
	B4	A16*	B5	A17	A18	A19	A20	E2	A21
Feed Rate g/min	3.6	3.6	3.6	3.6	3.6	3.6	0.3	0.3	3.6
Medium m^2	20.5	19.1	20.5	31	31	28.5	28.5	22	17.5
Test Flow m^3/h	2550	1700	850	3060	1700	3400	1700	1700	1700
Face Velocity m/min	2.1	1.5	0.72	1.65	0.91	1.99	0.99	1.29	1.62
Dust Load/unit area (1000 Pa)	4.4	8.4	15.6	3.9	9.7	3.9	8.4	4.1	4.6
grams	Pressure Drop (Pascals)								
0	340	200	150	290	155	320	150	210	335
20	430	310	175	350	210	440	265	330	485
40	600	385	225	440	265	590	315	530	620
60	805	470	285	560	340	720	370	750	800
80	940	555	335	710	400	840	450	960	1000
100	1150	600	410	860	450	940	500		
120	1295	740	450	980	510	1120	575		
140	1445	840	510		560	1280	610		
160	1550	960	560		620	1500	640		
180		-	620		680		725		
200			680		730		810		
220			730		790		890		
240	90g=	170g=	795		830		975		
260	1000Pa	1000Pa	840		890				
280			890		950				
300			950		1000				
320			990						

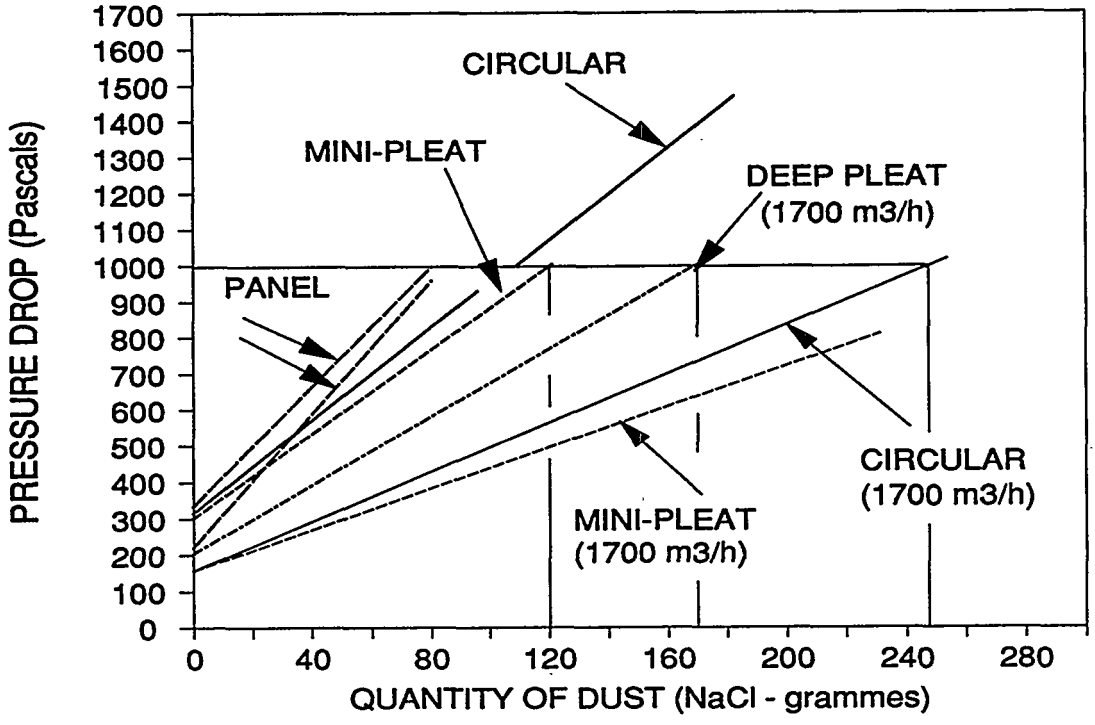
* Test repeated on 3 additional filters. At 1000 Pa the quantity of dust required differed by no more than +/- 10g.

In high flow installations the choice of secondary filter is between mini-pleat and circular filters. For new installations the circular filter is the most suitable for reasons not connected with dust holding capacity as mentioned earlier in this report and reported elsewhere[4].

Downrating the circular and mini-pleat filters to 1700 m^3/h is seen to increase the filter life by at least a factor of 2 giving the filters a 50% longer life - under these loading conditions - than the deep pleat filter. However, as in all these cases a more detailed study of all the parameters involved in selecting a particular design of filter needs to be made before it can be determined which is the optimum filter to use and at what flowrate.

FIGURE 15: DUST LOAD vs PRESSURE DROP

VARIOUS FILTERS - SODIUM CHLORIDE LOADING



Any evaluation of this type must consider at least some of the items detailed in Table 6 below.

TABLE 6: Factors to be considered in the choice of a Secondary Filter

Factor	Points to consider			
	D. pleat	Panel	M. pleat	Circ
Unit costs:	1	1	2-3	2-3
Volume flowrate m^3/h	1700	1700	3060	3400
Ease of sealing	POOR	POOR	POOR	BEST
Ease of handling	AVE	BEST	POOR[1]	BEST
Ease of volume reduction/disposal	POOR	BEST	POOR	BEST

*1 Mini-pleat filter scores badly due to its excessive weight. Currently available mini-pleat filters weigh of the order of 30kg (66lb) for the high temperature version.

VI. CONCLUSIONS

From this work the following conclusions can be drawn:

Primary Filter Applications (C. Black)

1. The circular and deep pleat filters tested performed significantly better than mini-pleat and panel filters in terms of dust holding capacity.

2. The circular filter, rated at 3400m^3/h, offers the twin benefits of a high volume flowrate and a high dust holding capacity. Downrating to 1700 m^3/h offers a significant improvement in dust holding capacity.

3. Downrating the mini-pleat filter to 1700m^3/h did not significantly improve its performance compared with the deep pleat filter.

4. Careful consideration should be given to the loading conditions anticipated in ventilation extract plant before recommending the use of mini-pleat or panel filters.

Secondary Filter Applications (Sodium Chloride)

1. The absolute dust holding capacity of all 4 filter types is significantly lower than achieved with the carbon black test dust.

2. At rated flow the deep pleat has the highest overall dust holding capacity.

3. Downrating the mini-pleat and circular filters to 1700m^3/h could lead to significant improvements in dust holding capacity.

4. In secondary plant locations however it is normally the case that the overall dust holding capacity is not overly significant because of the extremely low loading (and loading rate) encountered by filters in these areas, and other factors are important in selection of filter type.

5. A special case is decommissioning operations[13] where a primary filter may be subjected to principally sub-micron aerosols, say from the oxy-propane cutting of a reactor shell, and where the absolute level of dust holding capacity is important because of the large quantities of aerosol produced.

VII. ACKNOWLEDGEMENTS

The authors would like to express their thanks to British Nuclear Fuels plc and the Authority Fuel Processing Directorate (AFPD) via the Filter Development and Standards Working Party (FDSWP) for providing funding for this work.

Thanks are also expressed to Mr R Pratt and to Mr F Johnson, Miss R French and Mr A. Taylor for their assistance in this programme of work.

In addition acknowledgements are given to AAF Ltd, M C Air Filtration Ltd, Vokes Ltd, Ozonair and Trox Brothers who supplied the filters for this work.

IIX. REFERENCES

1. AESS 30/93402 "Filter inserts, High Efficiency Particulate Air (HEPA), Rectangular" (Nov 1984).
2. AESS 30/95100 "Filter inserts, High Efficiency Particulate Air (HEPA) circular plug-in 470 and 950 l/s capacities" (June 1987).
3. AESS 30/93400 "Filter Medium for use in HEPA Filters" Aug 1989
4. R.P.Pratt/B.L.Stewart UKAEA "Application of Circular Filter Inserts" 20th DOE/NRC Nuclear Air Cleaning Conference
5. R.P.Pratt, UKAEA & B.Tinklin, M.C.Air Filtration "Development of Circular Filters for Active Facilities" Gaseous Effluent Treatment in Nuclear Installations, Luxembourg, Oct 1985.
6. R.P.Pratt/B.L.Green UKAEA. "Collection of aerosols in HEPA filters". 19th DOE/NRC Nuclear Air Cleaning Conference. Paper 12-4
7. R.P.Pratt UKAEA. "A preliminary assessment of the dust holding versus pressure drop characteristics of high capacity HEPA filters". 16th DOE/NRC Nuclear Air Cleaning Conference.
8. M.Pierce, R.Guimond, N.Lifshutz Hollingsworth and Vose Co "A General Correlation of DOP Penetration with Face Velocity as a Function of Particle Size using the FTS-200" 20th DOE/NRC Nuclear Air Cleaning Conference
9. BS 3928 : 1969 "Method for Sodium Flame test for Air Filters"
10. R.Poynting CDE. "Comparisons of the Sodium Chloride Aerosols used by the CDE for air filter tests (U) June 1979. CDE tech note No. 393.
11. Mr C.Hamblin, UKAEA - Unpublished work
12. Mr D.Loughborough, UKAEA - Unpublished work
13. Dr K.S.Robinson, C.Hamblin UKAEA "Measurement and removal of steel flame cutting aerosols (0.01-1 micron) in WAGR Decommissioning.

DISCUSSION

BERGMAN: Are the two filter loading tests British Standards and how long do the tests take?

LOUGHBOROUGH: The loading tests are not British Standards, but a carbon black loading test is now included in all HEPA filter specifications issued by the UKAEA. The tests can last as little as 22 min for sodium chloride loading to several hours for some of the carbon black tests. Sufficient data is available in the report to determine the time for each test. You should note that since loading in the range 0.3 to 2.4 g/min gives identical results for both test dusts, there is no advantage gained from choosing a slow loading rate.

SIMULATED PERFORMANCES OF A SINGLE PARTICLE LASER LIGHT SCATTERING ANALYZER

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Abstract

The subject of this paper is the realistic simulation of an "*in situ*" laser light scattering apparatus which will be realized to measure the physical characteristics (number density, size and refractive index) of a filter testing aerosol in its actual flow regime.

The apparatus is conceived for analyzing simultaneously the intensity of the light scattered at different angles by each particle passing through an optically defined measuring volume Vs (hereafter called scattering volume).

In this way, the univocal determination of both size and refractive index may be obtained provided the incoming light flux is kept constant, within 15%, inside the scattering volume.

The complete simulation of the optical apparatus has been performed by supposing that only one particle at a time is analyzed and that the particles are spherical, thus allowing the Mie theory to model their scattering behavior. The realistic simulation of the experimental data has been performed taking into account both the statistical nature of the photon emission process and the statistical fluctuations related to the gain of the photomultiplier tubes. Further, an electronic circuitry noise of the order of 1% has been supposed.

The fitting procedure as been performed using the MINUIT χ-square minimization program[2].

Particle size (0.1÷3.0 μm), complex refractive index (real and imaginary part) and light flux incident onto the particle are considered as the four variable parameters.

Preliminary trial runs performed with the aim of determining the minimum number of collecting angles necessary to obtain the convergence of fitting procedure to the expected values of the unknown parameters, showed that this convergence could be only occasionally obtained even if up to 10 collecting angles had been adopted. This occurrence demonstrated the existence of a strong correlation between the four parameters.

Subsequent runs performed for non conductive materials gave, on the contrary, better results. In this case the remaining three unknown parameters can be determined with a 10% precision by reference to the size and 15% precision by reference to real part of the refractive index, provided that the simulated data are affected by an error less than 10% in correspondence of a set of six collecting angles.

Introductory remarks

Since the features of the instrument we intended to develop were addressed to the investigation of HEPA filters penetration as a function of filter parameters, our efforts were focused in obtaining a high resolution within the submicron size range (0.1÷1.0 μm) together with a good number counting accuracy, without perturbing the aerosol flow conditions.

To this end, we arranged a simple optical assembly which allowed to associate the size of a particle of known refractive index with the intensity of the light scattered at 90°, independently of the particle trajectory within the scattering volume Vs[1].

The best results were obtained by focusing into Vs the image of a finely adjustable slit that allowed the transmission of a narrow central portion of the gaussian laser beam (TEMoo). In this way an approximately rectangular intensity distribution was obtained.

The scattering volume size was kept so small as to fulfill the "*single particle*" mode operation within the particle concentration working range, namely $10^5 \div 10^6$ p/cm^3.

This simple collecting geometry didn't allow to univocally relate the instrument response to both the particle size and refractive index which needs to be already known. Moreover, a non monotonic relation between detected amplitudes and particle diameters was found while performing the measurements for particle sizes within 0.1÷1.0 μm range. This kind of drawback made it difficult to obtain a proper calibration of the measuring apparatus. Consequently, we decided to improve the technique by analyzing the intensity of the light scattered as a function of the collecting angle. In such a way, the univocal determination of both size and refractive index may be obtained.

On account of the complexity of such a realization, a complete realistic simulation of the behavior of an optical apparatus which performs the analysis of the light pulses scattered by a single particle at different angles has been carried out.

By means of a minimization program any set of simulated experimental data is then fitted with the Mie scattering functions to determine the particle size and refractive index.

The scattering volume is supposed to exhibit the optical characteristics determined in the previous laboratory set-up[1].

The simulation results refer to an hypothetical set-up, whose general features are described in the next section.

New optical system conceptual design

In fig.1 a particular configuration of the optical set-up is shown.

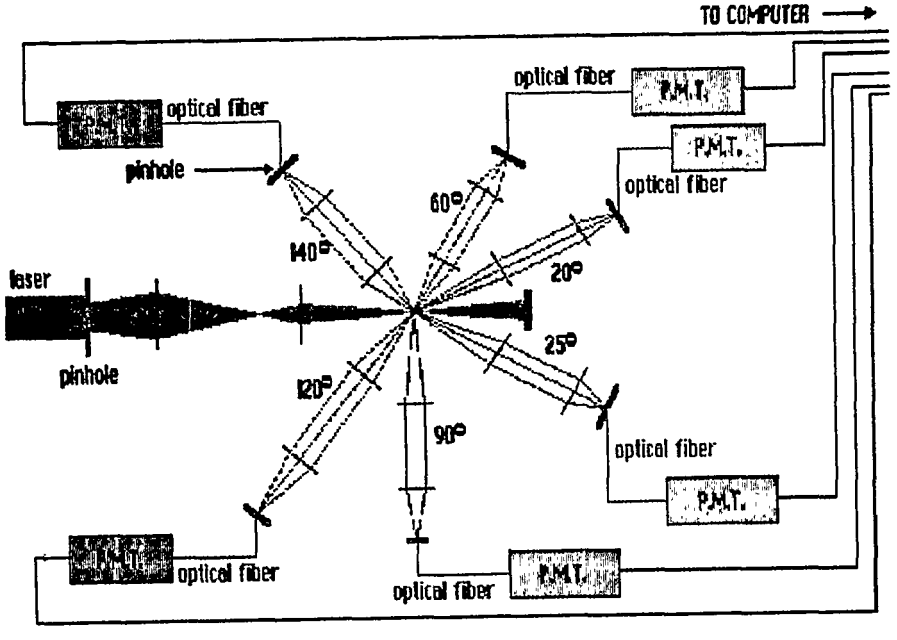


Figure 1 System conceptual design

An argon-ion laser beam of radius **b** illuminates a pinhole of diameter **p**, whose reduced image is focused in the center of the measuring cell by a confocal lens system. The pinhole is used to select the central region of the gaussian beam distribution (TEMoo) in order to obtain a "*top-hat*" laser beam profile in the transverse plane by reference to the beam propagation direction (fig.2).

The collection arms are composed by confocal lens systems of given *f-numbers*; pinholes of suitable diameter are set in the back focus of each collection system and all the light passing through them is transmitted to the photodetectors via optical fibers. This arrangement allows the use of photomultiplier tubes as detectors even if the light is collected at close angles.

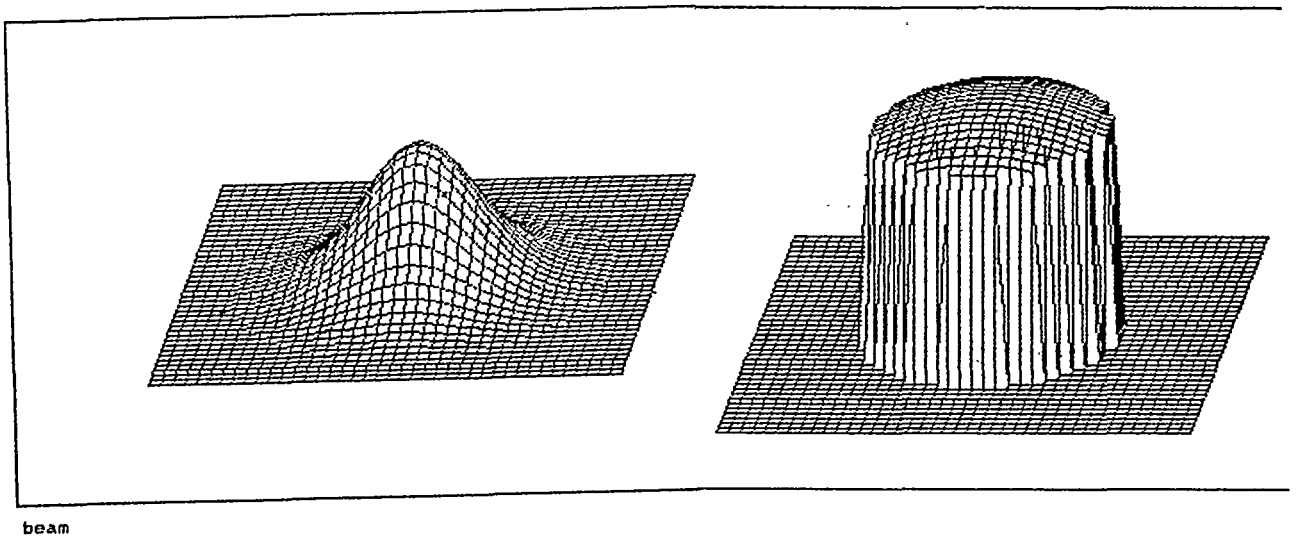


Figure 2 a) gaussian beam distribution; b) "*Top hat*" distribution.

The technique involves the simultaneous measurement of the peak amplitude of the signals detected at the selected collecting angles which are proportional to the scattered light intensity, integrated over the accepted solid angles $\Delta\Omega_i$:

$$V_i = K \int_{\Delta\Omega_i} J(x,y,z) \frac{\partial\sigma(\alpha,\theta,\tilde{m})}{\partial\Omega} d\Omega$$

where **K** is a constant that depends on the optics transmission and PMT cathode quantum efficiency, **J(x,y,z)** is the incident photon flux and $\partial\sigma(\alpha,\theta,\tilde{m})/\partial\Omega$ is the Mie differential scattering cross-section.

The measured values on the left hand side of the equation are usually affected by systematic errors caused by background noise (scattering due to density of matter present within **Vs**), and by random errors associated with the statistical nature of photon emission process, along with statistical noise associated with the detectors and built-in electronic circuitry.

In order to reduce the resulting uncertainty in the determination of the parameters under measure, without using sophisticated inversion algorithms[3], the relative intensity

of the light, simultaneously scattered by a particle at different angles, will be analyzed in a such way as to obtain the particle "signature" from the angular scattering pattern.

The apparatus is conceived to perform the analysis of only one particle at a time, thus providing a set of data only affected by the experimental errors.

This set of data, together with their errors, constitute the inputs for a χ-square minimization program to the Mie scattering function.

The computation is carried out by minimizing the function

$$\chi^2(\bar{p}) = \sum_{i=1}^{n} \left[\frac{V_i - T_i(\bar{p})}{\sigma_i} \right]^2$$

which is the sum of squares of deviations between simulated experimental values V_i affected by their errors σ_i, and theoretical values $T_i(\bar{p})$, containing as variable parameters:

-particle size
-refractive index (real part, imaginary part)
-light intensity flux incident onto the particle.

Although the constraint of achieving an approximately uniform illumination throughout the scattering volume is less critical, since the angular scattering pattern is univocally related to particle size and complex refractive index, the "*top-hat*" light intensity distribution makes it possible to obtain a well defined scattering volume at 90° collecting geometry, thus allowing an accurate determination of the number density and comparable experimental errors in the measurement of the characteristics of similar particles. Further, it allows to optimize the photodetectors dynamic range because comparable signal amplitudes are detected in correspondence of similar particles.

Simulation

The measure of both size and refractive index of spherical submicron particles has been simulated making use of a personal computer fitted out with math. coprocessor 80287.

FORTRAN language has been used to code the subroutine which compute the Mie theory differential scattering cross-section for whatever particle size and refractive index.

Since BASIC is the actual working language, the above-mentioned subroutine was compiled and linked as a BASIC library.

The calculation is performed by supposing that:

-only one particle at a time is present within the scattering volume Vs;
-the particle are spherical, thus they allow the use of Mie theory for the differential scattering cross-section computation.

The numerical algorithm has been developed taking into account the existing literature concerning this subject[5,6,7,8].

It makes use of the following input data:

-light intensity flux (ph./cm^2 sec) that hits the particle when it travels through Vs (random variable)
-laser beam characteristics (power, wavelength)
-particle size and both real and imaginary part of the refractive index.

The realistic simulation of the experimental data, that is the detector's output signals obtained in correspondence of the selected collecting angles, has been performed taking into account that the Mie scattering cross-section is a probabilistic quantity. Thus, if one takes **N** as the number of photoelectrons produced at a given angle within the integration time τ, and **G** as the gain of the PMT, the measured voltage signals can be expressed as:

$$V_i = \frac{\alpha}{\tau} \sum_{k=1}^{N} G_k$$

where $\alpha = e\ R$ (e = electron charge = $1.6\ 10^{-19}$ C; R=resistence of the detector output).

When **N** increases, we know from the central limit theorem that the resulting V_i distribution should approach a gaussian distribution characterized by a mean value

$$\langle V_i \rangle = \frac{\alpha}{\tau} \langle N \rangle \langle G \rangle \qquad (1)$$

and by a variance

$$\sigma_i^2 = \frac{\alpha^2}{\tau^2} \langle N \rangle \left[\langle G \rangle^2 + \sigma_G^2 \right] \qquad (2)$$

For these reasons, in order to simulate a vector of realistic experimental data, gaussian distributed random variables characterized by the indicated values of $\langle V_i \rangle$ and σ_i^2 have to be generated.

Further, to take into account the uncertainty affecting the light intensity flux that hits the particle, which is caused by the variable light intensity level within the Vs, a "*clean top-hat*" gaussian beam profile has been supposed. The chosen random variable takes into account this uncertainty since it simulates a random off-axis displacement of the particle trajectory.

Ultimately, an electronic circuitry noise of the order of 1% has been taken into account.

The validity of the chosen model to the end of representing the real experiment is closely related to the limit that can be found when applying the Mie theory to spherical particles. The model is thus adequate by reference to the size range under measurement.

Scattering volume analysis

The scattering volume dimensions are closely related to the maximum achievable particle number density. This limit is approximately equal to the inverse of the scattering volume Vs, that is:

$$\rho_{max} \leq \frac{4P_i}{V_s}$$

where P_i represents the probability of having two or more simultaneous scattering events[4].

In the adopted optical configuration each detector will collect light from its own scattering volume *Vsi*, and consequently, the effective regions from which the light is collected could not coincide. Moreover, a different collection efficiency could result at different collecting arms.

These problems can be solved provided that the multiangular PMT array is triggered by a coincidence circuit driven by the 90° collection system. In this way, one can be sure that the scattered light, collected at different angles, is actually produced by the particle under measurement.

Further, if the laser beam dimension is much smaller than the detector's pinhole diameters (fig.3), a suitable combination of the optical parameters of the whole collecting system will assure well overlapped scattering volumes.

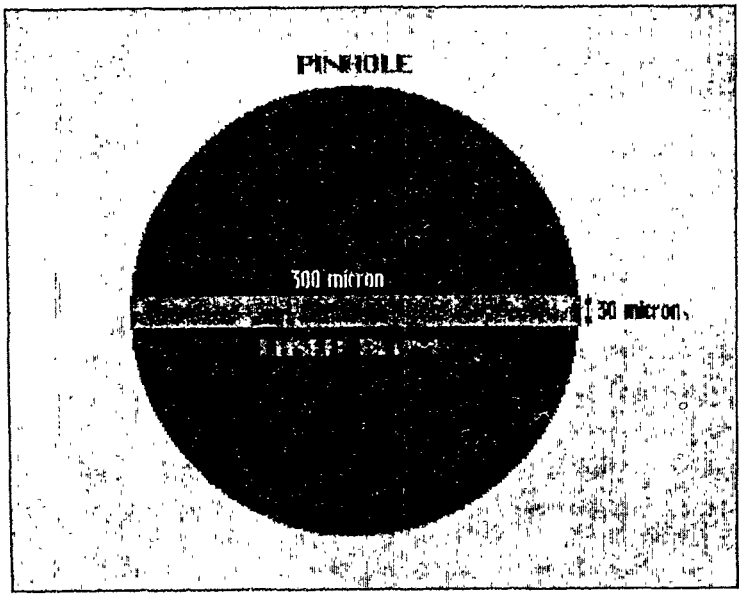


Figure 3 Beam diameter vs. pinhole diameter

Detector's pinholes are all imaged by confocal lens systems of definite f-numbers in the central region of the measuring cell.

The computation results concerning the collection efficiency distribution are shown in fig.4 for the most critical receiving geometry, set at an angle of 20°.

As can be observed in the figure, a slightly varying collection efficiency in the collection direction is obtained by an appropriate choice of the receiving optical system f-number and pinhole diameter.

The alignment of the whole optical system is supposed to be performed in such a way as to get the images of detector's pinholes set in the center of the measuring cell. The scattering volume dimensions will be determined by intersecting the "*top-hat*" gaussian beam profile of the laser beam with the efficiency distribution of the 90° collector.

The percent variation in the Vs dimensions, in correspondence of the most unfavorable geometry set at 20°, is expected to be about 25%.

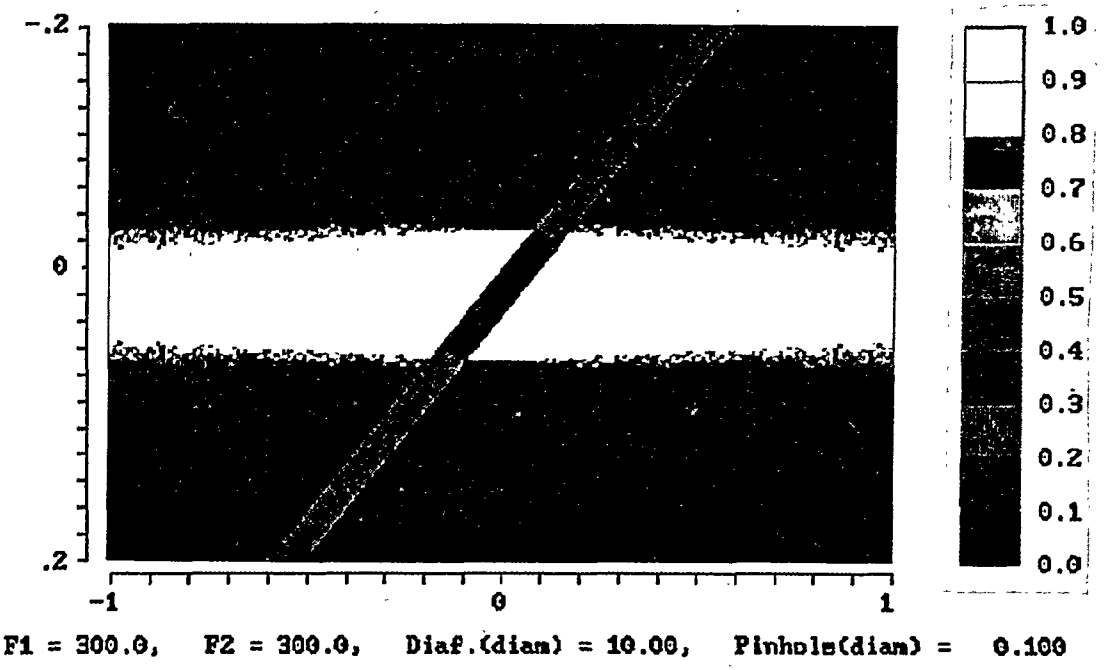


Figure 4 Collection efficiency distribution computed for the 20° collecting geometry.

Description of the first results obtained.

The computation has been carried out under the following assumptions:

-an argon-ion laser (2W all line) is used as light source;
-the scattering plane is perpendicular to the polarization of the incident light.

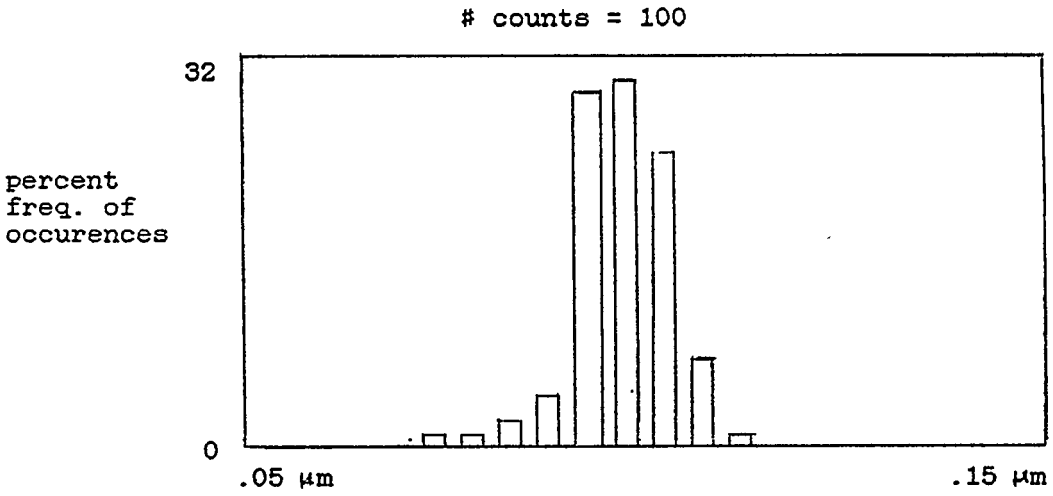
Many subsequent trial runs were carried out with the aim of determining the minimum number of collecting angles required to obtain the convergence of the fitting procedure to the expected values of the four unknown parameters (size, real and imaginary part of the refractive index, incident light flux).

Unfortunately, even if up to ten collecting angles had been adopted, this convergence was only occasionally obtained, thus showing a strong correlation between the four parameters.

On the contrary, when the imaginary part of the complex refractive index is chosen to be zero, a least number of six different collecting angles is needed in order to determine the remaining three unknown parameters.

The histograms shown as an example in fig.5a,b and fig.6a,b illustrate the results obtained in correspondence of particles characterized by different diameters and real part of the refractive index. As can be observed in fig.5a,b, an acceptable resolution is attainable also for particles in the lower end of the instruments size range (i.e. for 0.1 μm particles characterized by a refractive index of 1.3 + i0.0). In this case, however, the value of integration time t that makes the corresponding set the simulated experimental data be affected by errors within 10% (eq.1, 2) could be not fully consistent with the physical constraints imposed by the aerosol flow conditions.

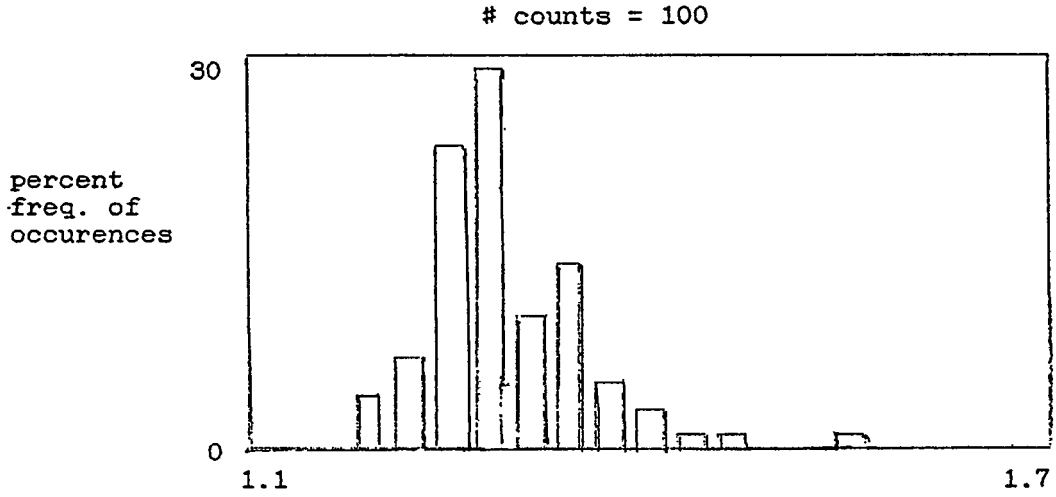
In general, the following occurrences have been found out:



a) particle diameter (μm)

mean(Diam) = 0.0995 μm stdev(Diam) = 0.0059 μm

$$\frac{\text{stdev(Diam)}}{\text{mean(Diam)}} = 5.9004\ \%$$

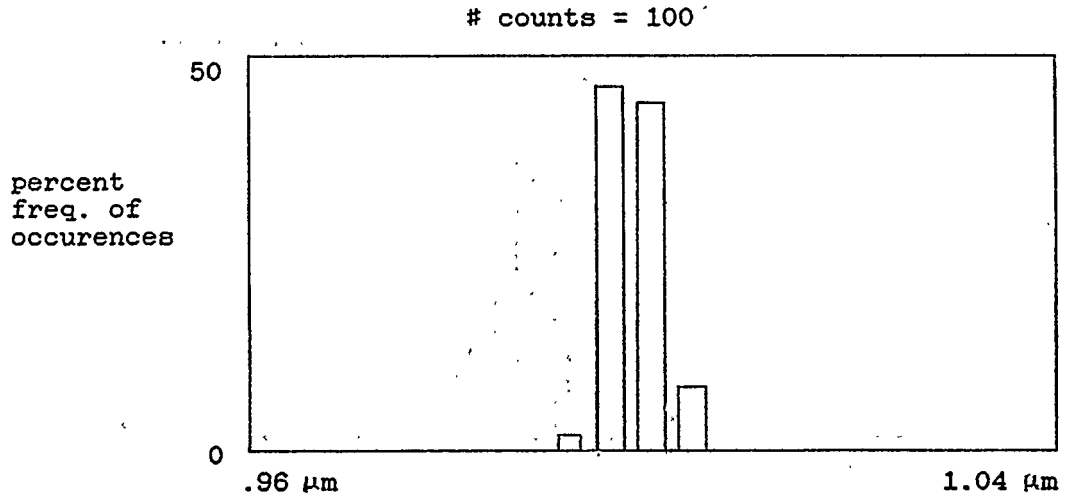


b) real part of refractive index

mean(Realind) = 1.3084 stdev(Realind) = 0.0704

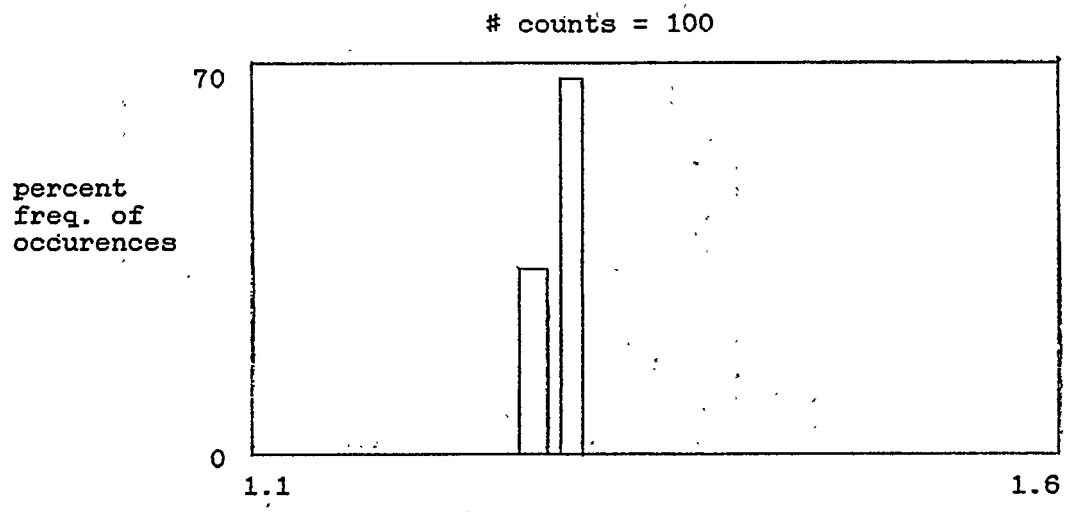
$$\frac{\text{stdev(Realind)}}{\text{mean(Realind)}} = 5.3773\ \%$$

Figure 5 Distribution resulting from the measure of 100 particles characterized by: ϕ = 0.1 μm; m = 1.3 + i 0.000



mean(Diam) = 1.0003 μm stdev(Diam) = 0.0023 μm

$$\frac{\text{stdev(Diam)}}{\text{mean(Diam)}} = 0.2326\ \%$$



mean(Realind) = 1.3005 $\text{stdev(Realind)} = 9.9889\ 10^{-4}$

$$\frac{\text{stdev(Realind)}}{\text{mean(Realind)}} = 0.0768\ \%$$

Figure 6 Distribution resulting from the measure of 100 particles characterized by: ϕ = 1.0 μm; m = 1.3 + i 0.000

-the determination of the unknown parameters can be obtained in the whole tested size range (0.1÷3.0 μm) provided that the simulated experimental data are affected by an error less then 10%;

-wider error fluctuations may be handled if single-line laser operation is adopted, but the resulting power density within the scattering volume is so low as to cause the fitting procedure to fail when particles at the lower end of useful size range are analyzed;

-subsequent runs performed for particles of definite size and refractive index indicate a reciprocal dependence in the determination of these two parameters, however, the resulting precision for both the parameters is maintained within 15%, if the collected light has the statistical fluctuations lower then 10%;

-the determination of the light flux is less critical for the fitting procedure, as could be expected, because it doesn't depend on the angular scattering pattern. Nevertheless, it can be different from the expected value, meaning that the minimization program prefer to adjust the overall intensity, once it has obtained the best values of the other two parameters from the shape analysis.

Conclusions

The convergence of the fitting procedure is assured, provided that only the real part of the complex refractive index is taken as variable input parameter. In this case, the determination of the unknown parameters for non conductive materials is attainable with a precision ranging from 0.5% to 15% depending on the errors which affect the data.

The analysis of particles with diameter less then 0.1 μm may be performed by either increasing the light density power within Vs or decreasing the aerosol flow velocity.

The simulated apparatus has been found to be sensitive to the out of range particle sizes beyond the upper limit of the useful range. Many runs, performed to evaluate how this fact could affect the measurements, showed that every time the resulting value of the χ-square function was very high. Therefore a simple criterium has been found that allows to reject these spurious data during the phase of data processing.

Although the simulation code as not been completely explored the results obtained up to now give sufficient indications concerning the general design features of a pilot single particle laser light analyzer that will allow to validate the assumptions on which is based the computation.

References

(1) Mazzacurati, V. et al.; "A laser spectrometry technique for the determination of aerosol characteristics". Proceedings of the 20th DOE/NRC pp.1179÷1188.

(2). James, F.; "MINUIT function minimization". Reprinted from the proceedings of the CERN Computing and Data Processing School, Pertisau, Austria, 10-24 September 1972 (CERN 72-21).

(3) Holve, D. J.; Annen, K. D.; "Optical particle counting, sizing, and velocimetry using intensity deconvolution". Opt. Eng. Vol.23 No.5 pp.591÷603 (1984).

(4) Holve, D. J.; "A Single Particle Counting Diagnostic System for Measuring Fine Particulates at High Number Densities in Research and Industrial Applications". SAND83-8246.

(5) Van de Hulst, H. C.; "Light Scattering by Small Particles". Wiley, New York (1957).

(6) Bohren, C. F.; Huffman, D. R.; "Absorption and Scattering of Light by Small Particles". Wiley-Interscience, New York (1983).

(7). Dave, J. V. "Subroutines for Computing the Parameters of the Electromagnetic Radiation Scattered by a Sphere". IBM Order Number 360D-17.4.002 (1968).

(8) Wiscombe, W. J.; "Improved Mie scattering algorithms". Appl. Opt.,19, 1505-1509 (1980).

CLOSING COMMENTS OF SESSION CO-CHAIRMAN ANDERSON

This session has concentrated on several aspects of filter testing; characteristics of different aerosols were explored and devices/methodology for their uniform insertion into operating or test systems were developed.

Two papers addressed the need for devices that would present uniform aerosol challenges to operating systems. Mr. Cassette, from France proposed a unique high-speed injection system using a nozzle which is perpendicular to the flow direction and has a multiplicity of jets; Mr. Loughborough used a distributed multi-point array for aerosol insertion. Both authors presented positive confirmation experimental data for their techniques but stated that their efforts thus far were laboratory type results and extrapolation to operations needs to be documented.

Two papers explored the suitability of DOP substitutes in air operated generators currently used for in-place filter testing. Mr. Crosby of Hamilton Associates found that different substrate liquids give different mass and photometric responses when used under identical operating conditions, while Dr. First of Harvard confirmed the different photometric responses while measuring the particle size parameters of the alternate liquids. Both papers concluded that alternates can be substituted for DOP provided appropriate factors are used to correct light scattering and generation rate/size differences.

Mr. Mulcey, from France, introduced a new technique for measuring mass concentration of a uranine aerosol during HEPA filter testing. This method reduces the response time to one minute and increases the sensitivity of the uranine testing; still it is deficient in the reliable determination of HEPA filter decontamination factors.

Mr. Carlon from Aberdeen Proving Grounds presented similar results for substitutes for DOP in "hot" generators used for QA/QC testing. This paper has rank ordered their substitute materials for both "hot" and "cold" aerosol testing.

Mr. Loughborough, from England, reported on his results of carbon black and sodium chloride loading of HEPA filters. Data was presented on lading factors for deep and mini-pleated units as well as panel and circular filters. This disparity of dust holding capacity of the various filter types indicates that plant designers should give careful consideration to operational conditions before selecting the optimum filter type for each application.

Mr. Neri, from Italy, described an "in-situ" laser light scattering technique for the measurement of the size and refractive index of aerosols. Much of the work performed thus far has been theoretical and exploratory to determine instrument parameters. Only operational exposure to real world particle populations and sizes will determine the limits, accuracy, and usefulness of the system.

SESSION 3

PANEL: CODE ON NUCLEAR AIR AND GAS TREATMENT ASME/ANSI AG-1

Monday:	August 13, 1990
Moderator:	W. H. Miller, Jr
Panel Members:	Thomas J. Vogan Steven G. Banton Sue A. Hobart

OPENING COMMENTS OF PANEL MODERATOR MILLER

INTRODUCTORY STATEMENT
W. H. Miller, Jr.

OVERVIEW OF DIVISIONS I AND II
T. J. Vogan

FIELD TESTING OF NUCLEAR AIR TREATMENT AND GAS PROCESSING SYSTEMS
S. G. Banton

AG-1, DIVISION IV, SECTION TB: FIELD TESTING OF GAS PROCESSING SYSTEMS
S. A. Hobart, F. J. Cannito, L. B. Nesbitt, D. P. Werkheiser, J. M. Pleva

PANEL DISCUSSION

OPENING COMMENTS OF PANEL MODERATOR MILLER

My name is Bill Miller and I am the Chairman of CONAGT. I have been with CONAGT now since 1975, some 15 years, and it has been a very rich and rewarding experience and at times downright challenging.

I would like to make a commercial announcement for CONAGT before we get started. CONAGT is the Committee on Nuclear Air and Gas Treatment. We have 80-100 active members depending on how our industry sponsors are supporting us in a given period. We meet twice a year as a Main Committee but a number of our Subcommittees are meeting up to four times a year. We have responsibility for three major documents: ANSI N509, ANSI N510, and the Code AG-1. This session today concerns AG-1. However I do want to talk about N509 and N510 since they were recently revised and issued for use. This is the second major maintenance revision of those documents which get a lot of use out in the utility industry. Try as we might to supplant N509 and N510 with the AG-1 code, these documents refuse to die. We have now acquiesced to the fact that N509 and N510 may live forever.

In planning this session, we thought it would be worthwhile today to familiarize you with AG-1 so that you go back to your organization and spread the word on AG-1. We are very concerned that since AG-1 is not referenced in some of the important DOE directives that people working in the nuclear air cleaning business in DOE facilities without knowledge of AG-1's existence. It is a better document than the N509 and N510 documents in many ways. It is not as system oriented but on a component basis it is much more complete.

In the way of a general disclaimer I must mention that none of the panel members or I are authorized to speak officially on behalf of ASME. Our statements today are our personal opinions.

PANEL SESSION: CODE ON NUCLEAR AND GAS TREATMENT ASME/ANSI AG-1

W. H. Miller, Jr. Moderator
Sargent & Lundy Engineers
55 E. Monroe Street
Chicago, Illinois 60603

INTRODUCTORY STATEMENT

The AG-1 Code covers the design, fabrication, inspection and testing of nuclear safety related air treatment systems and gas processing components used in nuclear facilities. This panel session will consist of brief presentations by members of the ASME Committee on Nuclear Air and Gas Treatment (CONAGT) on major portions of AG-1 as follows:

Overview of AG-1	W. H. Miller
Divisions I and II	T. J. Vogan
Division IV - Section TA	S. G. Banton
Division IV - Section TB	S. A. Hobart

These presentations are meant to familiarize you with AG-1 and stimulate discussion among the panel members and the audience. Therefore, presentations will be followed by solicitation of questions and comments from the audience. These panels are one means that CONAGT uses to elicit input from users or potential users for Code improvements.

OVERVIEW of AG-1

Air cleaning standards produced by organizations such as the American Society of Heating, Refrigerating and Air Conditioning Engineers (ASHRAE), Underwriters Laboratories (UL) and the Air Conditioning and Refrigeration Institute (ARI) have been recognized for many years. When nuclear power plants became viable in the mid 1960's, there were few specific standards available to the design and testing of nuclear air treatment systems and gas processing systems. In the early 1970's, the American National Standards Institute (ANSI), charged ASME's N45.8 Committee, which dealt with reactor plants and their maintenance, with developing documents for critical air cleaning systems in nuclear power plants. The initial N45.8 Committee scope was to develop standards to cover the design, fabrication, installation, testing and maintenance of high efficiency airborne radioactivity systems. ANSI N510, Testing of Nuclear Air Cleaning Systems was published in 1975 and ANSI N509, Nuclear Power Plant Air Cleaning Units was published in 1976. In 1975, ASME transferred the responsibility for these standards from the N45.8 Committee to the newly formed Committee on Nuclear Air and Gas Treatment (CONAGT). The scope of CONAGT was expanded from the N45 Committee to develop both Codes and Standards for all air cleaning equipment, which included

both HVAC systems and gas processing systems. All components and their supports were included.

CONAGT intended to utilize existing codes, standards, standard specifications and guides, as applicable, for these new and revised codes and standards. However, most of the requirements in these documents were for commercial or industrial service and were not directly useful for the nuclear applications. Early development efforts by CONAGT focused on bringing extensive personal knowledge to the committee and expanding the commercial technology to suit the evolving nuclear plant applications. This led to the development of the ASME AG-1 Code. Development priority was placed on engineered safety features equipment and systems.

This Code is a relatively new document (first published in 1985) that has had limited application since the decline of new nuclear plant construction in the USA. The Code is organized into 13 HVAC equipment sections, 6 gas processing sections, 2 test sections and one general requirements section. Each section was prepared, approved and published separately to get as many requirements into the users hands as possible, and the Code is now available for use in the design and procurement of most replacement or upgraded components. Approximately 50% of the code has been published to date.

DESCRIPTION OF THE CODE

The AG-1 Code is separated into four major divisions, namely General Requirements (Division I), Air Cleaning and Conditioning Equipment (Division II), Process Gas Treatment Equipment (Division III), and Testing (Division IV).

The detailed Table of Contents is repeated here.

Division I		General Requirements
	Section AA	Common Articles
Division II		Ventilation Air Cleaning and Ventilation Air Conditioning
	Section BA	Fans and Blowers
	Section DA	Dampers and Louvers
	Section SA*	Ductwork
	Section RA	Refrigeration Equipment
	Section CA	Conditioning Equipment
	Section FA*	Moisture Separators
	Section FB*	Medium Efficiency Filters
	Section FC	HEPA Filters
	Section FD	Type II Adsorber Cells
	Section FE	Type III Adsorbers
	Section FF	Adsorbent Media
	Section FG*	Mounting Frames
	Section IA*	Instrumentation and Control
Division III		Process Gas Treatment
	Section GA*	Pressure Vessels, Piping Heat Exchangers, and Valves
	Section GB*	Noble Gas Holdup Equipment
	Section GC*	Compressors
	Section GD*	Other Radionuclide Equipment
	Section GE*	Hydrogen Recombiners
	Section GF*	Gas Sampling
Division IV		Testing Procedures
	Section TA*	Field Testing of Air Treatment Systems
	Section TB*	Field Testing of Gas Processing Systems

* Not yet published.

This paper deals with Divisions I, II, and IV. Division III covers Gas Processing Equipment and is in the early stages of preparation. The first part addresses the general requirements and the design of air cleaning and conditioning equipment.

OVERVIEW of DIVISIONS I AND II

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Abstract

The major portion of design requirements for nuclear air treatment system components is contained in Divisions I and II. These divisions of the Code were initiated in 1976 and 1977. Emphasis and priority was placed on the engineered safety features equipment. All of the sections are either published or in advanced stages of preparation. These divisions are very broad in their scope and this paper provides descriptions of their content and an overview of their use.

General Requirements

Section AA of the Code contains articles which are common to all other sections of the Code. The topics covered are contained in the Introduction (AA-1000); Referenced Documents (AA-2000); Materials (AA-3000); Structural Design (AA-4000); Inspection and Testing (AA-5000); Fabrication (AA-6000); Packaging, Shipping, Receiving, Storage and Handling (AA-7000); Quality Assurance (AA-8000); and Nameplates & Stamping (AA-9000). Article AA-4000 is a major part of this section and contains the detailed structural design requirements for all equipment and supports. Design by analysis and design by test rules are defined.

Design by Analysis: Load combinations for various service levels are provided in a format similar to that used in the Boiler and Pressure Vessel Code, namely service levels A,B,C, and D. For each service level, rules are defined for both maximum stress theory and maximum deflection theory. Formulae and acceptance criteria are detailed for plate- and shell-type components for maximum stress, maximum deflection and allowable buckling stress. Linear type components (supports) are designed by rules similar to that provided in AISC and Section III, Subsection NF; application formulas and acceptance criteria are provided.

Design by Test: Detailed rules for seismic design verification testing explain single and multiple frequency testing, single and multiple axis testing, and test durations. These rules compliment ANSI/IEEE 344 requirements.

Another major article in this section is Article AA-6000 which addresses fabrication, joining, welding, brazing, protective coating and installation. The welding and brazing requirements, inspection and acceptance criteria are listed in detail. Special welding instructions pertaining to sheet metal ductwork and housings is

highlighted. This section represents Included are butt welds, fillet welds, spot welds and stud welding.

Nuclear Air Treatment Systems (NATS)

Division II of the AG-1 Code contains the component specific requirements for virtually all of the HVAC equipment and constitutes the bulk of the Code. The equipment covered are fans, blowers, dampers, louvers, compressors, refrigeration heat exchangers, valves, cooling coils, electric heaters, ducts, instruments, control devices, moisture separators, filters, adsorbers and housings. Each section provides references (Article 2000), materials of construction (Article 3000), detailed design (Article 4000), shop inspection and testing (Article 5000), fabrication (Article 6000), packing storage and shipping (Article 7000), quality assurance (Article 8000), and nameplates (Article 9000). The specific parameters affecting each component design and construction are caused to be provided within each component section and Section AA is referenced for design rules.

Experience with the AG-1 Code

The first issue of the AG-1 Code was in 1985. The contents were limited to the general requirements and detailed requirements for fans and refrigeration equipment. The 1989 revision and addenda represent a relatively complete set of Codes. The ductwork, moisture separators, medium efficiency filters and instrumentation will be issued by addenda in the near future. There is some recent activity among US utilities in the area of nuclear air treatment system equipment procurement where sections of AG-1 are referenced for design. In addition, the design of the HVAC systems for new generating units at Yonggwang in The Republic of Korea used AG-1 for specifying NATS equipment. These early uses have not resulted in significant questions or interpretations.

Example of Code Usage

For plants designed to the ANSI N509, 1976 or 1980 requirements, procurement of any replacement component to AG-1 can readily be accomplished by specifying the required process parameters, loading conditions and the specific code section can be referenced for design and fabrication.

For new or replacement NATS fan, the requirements of AG-1, Section BA would be referenced and the following data would be specified by the engineer:

- Fan type and blade shape
- Air flow, actual cfm
- Total pressure to be developed, in-wg
- Maximum discharge velocity, ft/min
- Rated and maximum air density, lb/cu.ft.
- Operating and peak temperature, F
- Intake details
- Fan operating range and duty
- Radiation

- Environmental temperature & pressure
- Seismic response spectra
- Other loading conditions

For a Type III charcoal adsorber, a typical replacement specification to AG-1, Section FE would include:

- Type of gas to be treated
- Volumetric flow rate, actual cfm
- Internal and external design pressure, in-wg
- Pressure time transients
- Temperature range, F
- Relative humidity range, %
- Contaminants to be removed
- Adsorbent type
- Required efficiency
- Face velocity, ft/min
- Bed thickness, inch or residence time, sec
- Differential pressure range across bed
- Sampling requirements
- Adsorbent handling requirements
- Seismic response spectra
- Other loading conditions

In contrast to these specialty items, ordering data for a standard Type II cell for an ANSI N509 system only require the following:

- Type of gas to be treated
- Contaminants to be removed
- Adsorbent type
- Seismic requirements
- Frame mounting detail

All other inputs and conditions are included within the standard design in the Code.

The AG-1 code is not excluded from being used on non-ANSI N509 systems. The code does contain requirements that can be readily adapted for those special conditions encountered. For example, in the filters areas, most differences would be dimension changes from the current standard dimensions. The critical parameters would be translated into the Code requirements and compromise designs that meet the intent of the current Code will be made. Pressure retaining components might show differences in materials and material documentation. Material selections and documentation basis was put into the Code sections to assist the user in matching the existing component designs. As experience grows among the manufacturers, we would expect that the NATS components would be standardized to the AG-1 design rules.

Summary

This presentation has provided a brief overview of the AG-1 Code, Divisions I and II. We have attempted to have as many participants of the Code writing groups as possible available for discussion. The dialogue and comments related to some of the issues will follow hard and fast positions; others involve guidelines and may have varying positions among the technical experts.

We have attempted to address all the issues in the Code in a clear, concise and accurate manner. As the experience base grows and the need for new or modified guidance is identified, the Code will be appended annually to reflect these needs.

FIELD TESTING OF NUCLEAR AIR TREATMENT AND GAS PROCESSING SYSTEMS

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ABSTRACT

The field testing sections of ASME AG-1 have been under development, by the Committee on Nuclear Air and Gas Treatment (CONAGT), since the early 1980's and are scheduled to be issued in late 1990 or early 1991. These sections will provide detailed construction acceptance test, and operating inservice test requirements for safety related nuclear air treatment systems and gas processing equipment built to the design specifications of ASME AG-1. These sections are formatted similar to the ASME pump and valve testing sections (ASME section XI), and can easily be applied to systems designed and built prior to the issue of ASME AG-1. The tests specified in these sections are designed to ensure safe and reliable operation of installed nuclear air treatment and gas processing systems, as well as periodic verification of continued proper operation. This paper provides an overview of the testing requirements specified in these sections and a background of their development.

INTRODUCTION

Section TA of AG-1 was designed to provide field test verification of the components and overall system operation for nuclear air treatment systems. Article TA-4000 specifies acceptance test requirements for the system and it's components following initial installation. Section TA-5000 specified the inservice testing requirements, and the associated interval for these tests to be conducted following facility startup. Approximately one half of the content of TA was derived from the existing guidance in ASME N510 "Testing of Nuclear Air Treatment Systems" which was originally published in 1975 and last revised in 1989. Some of you may remember my presentation on this new revision from the conference in 1988. The remainder of TA was developed for specific components typically contained in nuclear air treatment systems, but not covered in N510. Examples include fans, dampers, refrigeration, and conditioning equipment. Also addressed are the integrated functions and

associated interlocks typically found on nuclear air treatment systems. These tests are designed to verify component and system operability which ensures they will provide the intended function when called upon to do so. System components

TA components and subsystems include the following:

- o Fans and Blowers
- o Dampers
- o Ducts and Housings
- o Filter and Adsorber Mounting Frames
- o Refrigeration subsystems
- o Conditioning subsystems
- o Moisture Separators
- o Prefilters
- o HEPA Filters
- o Adsorber Banks
- o Adsorber Media

Field Tests:

Testing specifications for each component and integrated system are outlined under two major articles in Section TA. Article TA-4000 provides Field Acceptance testing and inspection requirements for nuclear air treatment systems following initial installation, or following component replacement, repair, modification or maintenance. Article TA-5000 provides inservice testing requirements, and is designed to verify nuclear air treatment system continuing operability. The testing is outlined in five major groups, these are:

- o Visual Inspections (visual degradation)
- o Pressure Boundary Tests (structural integrity, leak tightness)
- o Component Functional Tests (individual component function)
- o Equipment Train Functional Tests (system operation)
- o Integrated System Test (automatic modes and interlocks)

Supporting these test requirements are eight appendices which provide detailed guidance on how to perform some of the specific tests. These are:

A: Visual Inspection Checklist
B. Structural Capability Test Procedure
C. Duct and Housing Leakage Test Procedure
D. Mounting Frame Pressure Leak Test Procedure
E. Airflow Distribution Test Procedure
F. Air Aerosol Mixing Uniformity Test Procedure
G. In-Place Bypass Leakage Test Procedure
H. Refrigerant Coil and Piping System Leak Test Procedure

Supporting Details

Section TA also provides detailed guidance on Test instrument calibrations, testing following an abnormal event in which the function of the nuclear air treatment system has received a severe challenge, and corrective action requirements associated with failure to satisfy the requirements of the specific test sections. In addition, acceptance criteria is provided when the results of a test impact the results of another test. However much of the specific component criteria is dependent on the owners project specifications.

Example of TA Usage

Here is a typical example of a system In-service test program using Article TA, section 5000 of the AG-1 Code:

Lets say a plant decides to use TA to formalize the periodic inspection and testing of the plants auxiliary building exhaust system. This system is typical of most plants and has a network of ducts, manual and automatic dampers, a filter housing, an inlet heater for carbon moisture control, a cooler for humidity and temperature control, moisture separators, prefilters, a HEPA filter bank, carbon filter bank, exhaust fan, and associated exhaust system ductwork. The plant develops detailed test procedures that contains the applicable test requirements of TA-5000. For example:

1. System Fans would be visually inspected and vibration would be monitored every quarter, the instruments on the system calibrated every two years, a fan performance test to verify the fan is still operating at or near it's fan curve every two years, and the pressure boundary components would be leak tested every ten years.

2. System control and isolation dampers would be visually inspected, exercised, calibrated, and all mechanical and electrical interlocks verified every two years.

3. System ducting would be visually inspected every two years and pressure boundary ductwork would be leak tested every ten years.

4. System heaters would be visually inspected every quarter, and the electrical circuits tested every two years.

5. System cooler would be visually inspected every quarter for leakage, and cooler water side differential pressure and flowrates every two years. Any power operated isolation valves or control valves would be tested on a two year frequency.

6. The moisture separator, prefilters, and HEPA filter would be checked for differential pressure loading every month and visually inspected at least every two years. The HEPA filter would be tested for bypass leakage every two years.

7. The carbon filter bank would be monitored for differential pressure loading each month, visually inspected and tested for bypass leakage every two years, and a carbon sample would be analyzed in a test lab every two years or 720 hours of operation.

8. Every two years, the entire integrated system function will be thoroughly tested to verify all auto starts, interlocks and automatic re-alignment functions occur as designed.

If the system or portions of the system are replaced, repaired, modified or has maintenance performed on it, Section 4000 provides additional testing requirements to verify proper system operation prior to return to service. These are basically the same requirements for acceptance testing following new system installation.

It is hoped that both Section TA, and Section TB which you will hear about next, will become as widely used and accepted by the nuclear industry as ASME Section XI has. It is intended, that once implemented, these testing standards will provide adequate diagnostics and verification of component operability that will ensure a high degree of reliability for both new and existing nuclear air and gas treatment systems.

DISCUSSION

MILLER: How many Main Committee ballots has TA been through?

BANTON: TA has been through one Main Committee ballot.

MILLER: The reason that this question is relevant is that rarely does a document get through our Main Committee on its first pass. I think that Susan Hobart will give you evidence that is usually a three pass process. That is why it takes us so long to publish these codes.

AG-1, DIVISION IV, SECTION TB: FIELD TESTING OF GAS PROCESSING SYSTEMS

S.A. Hobart, F.J. Cannito, L.B. Nesbitt, D.P. Werkheiser, J.M. Pleva[1]

Abstract

The purpose of this paper is to present and update the status of Section TB of the AG-1 code. This code section covers the field testing of nuclear safety-related gas processing equipment, such as the post-LOCA hydrogen recombiners and hydrogen ignitors used in commercial nuclear power plants. CONAGT was requested by the American Society of Mechanical Engineers (ASME) Board of Nuclear Codes and Standards (BNCS) to provide testing requirements, along with equipment design requirements, for gas processing equipment in two consensus codes--one for nuclear safety-related and one for non-nuclear safety-related nuclear air treatment and gas processing equipment. A subgroup has been working for several years to prepare the gas processing equipment test sections. The need for coverage of nuclear safety-related equipment being the most urgent, the testing section for AG-1 was given priority. Subsequently, the non-nuclear safety-related section that would cover process offgas and waste gas systems has been put on indefinite hold, along with the rest of the code for non-nuclear safety-related nuclear air treatment and gas processing equipment. Extensive loss in plant output due to problems in the offgas and waste gas systems may influence this decision in time. The TB section of AG-1 is scheduled for its third CONAGT main committee ballot this year.

I. Introduction

The purpose of this panel is to present an update of the AG-1 code--its content, intent, and status. This individual summary discusses the subsection of the code related to testing of gas processing equipment. Details of the code section are not presented here, although any details-related questions that might be asked by the audience will certainly be addressed. A previous paper on this code section can be found in proceedings from the 19th Air Cleaning Conference.[2] This current presentation will summarize the scope of Section TB, the specific equipment covered, the organization and requirements of the code, and its current status.

Testing of gas processing equipment (both nuclear safety-related and non-nuclear safety-related systems) in commercial nuclear power plants has been left to the discretion of plant owners and operators, to date. However, the large number of fires and detonations occurring each year in non-nuclear safety-related offgas and waste gas systems has illustrated the need for careful and complete testing programs to verify functionality and safety of any gas processing-type equipment. The potential hazard to both plant personnel and the general public if such a failure should occur in one of the nuclear safety-related systems has prompted the development of a test section for AG-1 that applies specifically to gas processing equipment. This presentation briefly describes the content of that section and its status with respect to approval and publication.

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[2] S.A. Hobart, F.J. Cannito, L.B. Nesbitt, D.P. Werkheiser, J.M. Pleva, and R. Reda, "An ASME Code for Field Testing of Nuclear Safety-Related Gas Processing Systems," Proceedings of the 19th DOE/NRC Nuclear Air Cleaning Conference, paper number 5-4, Seattle, Washington, August 1986.

II. Scope and Purpose

As is true for the entire AG-1 code, this section applies only to nuclear safety-related equipment. For the purposes of this code, gas processing equipment is defined as equipment that either produces a change of state or composition or alters the concentration of gases. Specifically, nuclear safety-related equipment designed to (a) stabilize the containment atmosphere or (b) process gases containing radioactive material during normal or emergency conditions is addressed by this code. It can be distinguished from nuclear air treatment system (NATS) equipment by its operating conditions of relatively high pressure, low flow, and high radioactivity content. In the case of systems containing both air treatment and gas processing equipment, this code section applies to the gas processing equipment (e.g., compressors, blowers, piping, and valves) and references section TA for testing requirements related to air treatment equipment (e.g., ductwork and dampers). Gas supply and fire suppression systems are excluded from the code.

The code applies to equipment that is assembled and ready for use, and that is installed in its intended use location. Requirements do not apply to manufacturers' pre-shipment acceptance tests.

Functional tests of equipment important for the protection of plant personnel and the general public are commonly addressed by consensus codes and standards. For example, Sections VI, VII, and XI of the ASME Boiler and Pressure Vessel Code (B&PVC) all address functional testing, in addition to pressure boundary tests. The need for a test code for nuclear gas treatment systems was identified by the ASME BNCS for the following reasons: (a) gas processing equipment functionality is important in order to protect plant personnel and the general public; (b) general requirements of the B&PVC currently must be tailored to the specific equipment designed to process gases; (c) minimum test requirements for this type equipment currently do not exist in a consensus code; and (d) test recommendations for gas process equipment are scattered among many different documents.

Because gas processing systems provided by various vendors have significantly different design features, actual detailed test procedures cannot be specified in an industry consensus code. In addition, a consensus code is intended to specify only those minimum requirements necessary to ensure safety. This code section has thus been designed to provide requirements for the types of tests that must be performed and considerations that must be included in preparing test procedures. Test measurements (e.g., temperature, pressure, and gas composition) that must be performed and checked against design specifications in system and component design criteria documents are also identified. Where test methodology is important, the code section either references applicable existing codes and standards or describes the methodology to be used. The system owner is held responsible for developing test procedures that comply with the code.

III. Specific Equipment Covered by the Code Section

At present, the code's definition of nuclear safety-related gas processing equipment applies only to a few systems designed for containment combustible gas control in commercial nuclear power plants: in-containment igniters, recombiners (both in-containment and external systems), and any gas-processing-type equipment found in mixing, diluting, inerting and purge systems. Future revisions to the code may incorporate requirements for filtered containment vent systems. In addition to the principal functional components of these systems (igniters and recombiners), the code covers compressors, blowers, piping components, on-line sampling and analysis equipment, and electrical, instrumentation and control equipment associated with each system.

IV. Code Section Organization and Requirements

The organization of code test section TB was developed to accommodate the unique requirements of testing, as opposed to design. An outline of section TB is shown in Table 1.

TABLE 1
Outline of Code Section TB

1000	Scope
2000	Definitions
3000	Test Requirements
3100	Test Personnel
3200	Test Instruments
3300	Test Boundaries
3400	Integrated Equipment Test Description
3500	Examination Procedures and Techniques
4000	Field Acceptance Tests
5000	Periodic Field Tests
6000	Testing Following an Abnormal Incident
7000	Repair and Retest
8000	Documentation

Subsection 3000 contains generic test requirements applicable to the majority of equipment and systems within the scope of this code section (e.g., visual examinations and pressure and leakage tests). Requirements for test instruments and test personnel are also provided in this subsection.

Details regarding specific systems' tests are given in Subsections 4000 and 5000. These subsections are organized in parallel for each system covered, as shown in Tables 2 and 3. This organization permits easy location of requirements by personnel who will use the code. That is, a paragraph numbered 4xxx will always refer to a field acceptance test, while 5xxx refers to a periodic field test. Within sections 4000 and 5000, a paragraph numbered x2xx will always refer to a recombiner system and one numbered xx3x will always refer to a functional test. Components that are common to more than one system (compressors and blowers, piping components, sampling and analysis equipment, and electrical and instrumentation and control equipment) are discussed separately in order to minimize redundancy.

A field acceptance test is required both for new systems and for existing systems that are subject to the code. The code section requirements for the initial field acceptance test include the visual examination and pressure boundary tests previously mentioned. In addition, component functional tests are required and the entire equipment train is required to be given a functional test to verify each component's operability under design conditions. In cases where design conditions cannot easily be simulated, some exceptions or substitute conditions are allowed for equipment

train functional tests. However, where replication of design conditions is crucial to verifying equipment operability, exceptions have not been permitted.

TABLE 2
Parallel Organization of Code Section TB
Field Acceptance Tests

4100 Igniter Equipment	**4200 Hydrogen Recombiner Equipment**	**4300 Mixing,Diluting, Inerting and Purging Equipment**	
4110 Visual Examination	4210 Visual Examination	4310 Visual Examination	
4120 Pressure Boundary Tests	4220 Pressure Boundary Tests	4320 Pressure Boundary Tests	
4130 Functional Tests	4230 Functional Tests	4330 Functional Tests	
4140 Acceptance Criteria	4240 Acceptance Criteria	4340 Acceptance Criteria	
4400 Sampling and Analysis Equipment	**4500 Blowers and Compressors**	**4600 Piping Components**	**4700 Electrical, Instrumentation and Control**
4410 Visual Examination	4510 Visual Examination	4610 Visual Examination	4710 Visual Examination
4420 Pressure Boundary Tests	4520 Pressure Boundary Tests	4620 Pressure Boundary Tests	4720 Pressure Boundary Tests
4430 Functional Tests	4530 Functional Tests	4630 Functional Tests	4730 Functional Tests
4440 Acceptance Criteria	4540 Acceptance Criteria	4640 Acceptance Criteria	4740 Acceptance Criteria

TABLE 3
Parallel Organization of Code Section TB
Periodic Field Tests

5100 Igniter Equipment	**5200 Hydrogen Recombiner Equipment**	**5300 Mixing,Diluting, Inerting and Purging Equipment**	
5110 Visual Examination	5210 Visual Examination	5310 Visual Examination	
5120 Pressure Boundary Tests	5220 Pressure Boundary Tests	5320 Pressure Boundary Tests	
5130 Functional Tests	5230 Functional Tests	5330 Functional Tests	
5140 Acceptance Criteria	5240 Acceptance Criteria	5340 Acceptance Criteria	
5400 Sampling and Analysis Equipment	**5500 Blowers and Compressors**	**5600 Piping Components**	**5700 Electrical, Instrumentation and Control**
5410 Visual Examination	5510 Visual Examination	5610 Visual Examination	5710 Visual Examination
5420 Pressure Boundary Tests	5520 Pressure Boundary Tests	5620 Pressure Boundary Tests	5720 Pressure Boundary Tests
5430 Functional Tests	5530 Functional Tests	5630 Functional Tests	5730 Functional Tests
5440 Acceptance Criteria	5540 Acceptance Criteria	5640 Acceptance Criteria	5740 Acceptance Criteria

For convenience of reference, requirements for tests are presented in the order normally followed by good engineering practice: visual examination, pressure boundary tests, component functional tests, and equipment train functional tests. Where the sequence of tests is important (e.g., hydrostatic tests of catalytic recombiner systems must be performed prior to installation of catalyst, and mechanical joint leakage tests must be performed after installation of catalyst), the test order is specified. The code requires that equipment train functional tests be performed following the completion of visual examinations, pressure boundary tests and individual component functional tests, but allows for some component functional tests to be performed concurrently with integrated flow testing. This requirement minimizes the potential for undetected damage to nuclear

safety-related equipment from failure of components that have not been inspected and tested prior to system operation, while maintaining flexibility in test scheduling.

Some plant designs allow shared use of a containment recombiner at more than one operating unit and the common equipment is normally kept in a central storage location. To ensure that the recombiner and permanent connections are properly aligned and the soundness of the equipment is not affected by transportation procedures and equipment, the code requires that each shared recombiner be installed at each of its potential use sites and all connections be leak-tested and/or functionally verified. However, integrated equipment train functional testing is required at only one of the potential use sites.

Periodic field tests are essentially repeat performances of the initial field acceptance test. The code requires that data from these tests be compared to both design criteria and values obtained from previous tests, in order to identify either a failure to perform properly or a trend in performance degradation. Required field testing frequencies have been designed to permit performance during plant refueling outages.

- Visual examinations and functional tests of igniters are required every ten years, with the examinations and tests for groups of igniters distributed throughout that period.

- Visual examination is required quarterly for stored recombiner equipment, and every two years for installed equipment. Leakage tests of joints and equipment functional tests are required every two years or every one or two refueling outages.

- Visual examination, leakage tests of joints, and equipment functional tests of mixing, diluting, inerting and purge equipment are required every two years or every one or two refueling outages.

IV. Status of Code Section Approval

The first draft of this code section was complete in 1986. The consensus code review, comment, revision, and approval process has been relatively slow for Section TB of AG-1, but has resulted in significant technical and editorial improvements over the first draft. This fall the section will be submitted for its third round of ballots by the ASME Main Committee of CONAGT. After consensus and approval is reached within CONAGT, the process will begin again with the ASME BNCS. Only after both groups have agreed on the code's content will it be published and available for use. That could occur as soon as next spring, or it may require another several years of negotiation and revision to ensure that an effective and usable test code is published.

DISCUSSION

MILLER: I should make note that there have been some changes in the Board of Nuclear Codes and Standards balloting process. Let me just explain for a minute the way that ASME is organized. ASME has a number of supervisory boards and the one that we report to is the Board on Nuclear Codes and Standards. On that Board sits some very distinguished gentlemen in the nuclear industry. Mr. Guy Arlotto from the Nuclear Regulatory Commission is Vice Chairman. Mr. Robert Dick of Duke Power Company is the Chairman. The Chairman of all of the committees that Board supervisors are member; the O&M Committee, the Quality Assurance Committee, the Qualification of Mechanical Equipment Committee, Section 3 Committee, Section 11 Committee, Crane Committee, and Accreditation Committee. Every document that passes one of the Main Committees is sent to the Board for review and ballot. I not only have to ballot all my Main Committee items, I have to ballot on all other committee items that come to the Board. The Board recently adopted a change in its administrative procedures. What was happening is one or two members on each ballot were holding up a lot of actions with negative ballots. So we devised a streamlined process which encourages committees to address negative ballots quickly and personally in a positive manner which permits the Board member to withdraw his negative ballot.

DISCUSSION

BELLAMY: I have two comments. My first question is for the Chairman of the panel. He asked me to elaborate on the use of the term "safety-related." I don't know what the term "safety-related" means. I confuse "safety-related" with "safety-grade" with "licensee approved" with "Q list" with "engineer safety feature." I am curious whether, this has caused confusion in usage or whether the terms are such well recognized terms that everybody knows what they mean without clear, concise definitions?

MILLER: I use the term in conjunction with "engineered safety feature systems," those systems that are necessary to place the reactor in a safe shutdown condition and maintain it in that condition.

VOGAN: Within AG-1 and ASME lexicons there are definitions that have been extracted for the term "nuclear safety-related." When we use "nuclear safety-related" we mean engineered safety feature systems and equipment that are required to function in a safety-related fashion. I think we use the terms interchangeably.

BELLAMY: I would have to agree. Anybody who has been to the Harvard In-Place Filter Testing Workshop has heard me state my philosophy that the only term that I personally use is "engineering safety feature." By that designation I have always meant any system designed to operate after an accident to mitigate the consequences of that accident and put the reactor in a safe shutdown condition. I interpret both of your comments to mean that there really is no confusion among industry people as to what "safety-related" means. Generally speaking, you use it in the same context as "engineering safety feature."

MILLER: That is correct.

VOGAN: If confusion is prevalent, we should in the next revision of AG-1 look at the use of our terms "safety-related," and "important to safety." I hate to use "important to safety," but if it is there we need to make sure it is well defined. And then focus on what our intent is as engineered safety features equipment and systems.

BELLAMY: I noticed a dichotomy in the comments of Susan Hobart and Steve Banton. I noticed a difference when they were talking about Code surveillance requirements, in that Steve Banton was very careful every time to say, "frequency of surveillance requirements" and even though his written writeup did not have the phrase in there, each time he included the words, "at least," e.g., "at least every two years," "at least quarterly," "at least every five years" (even though the Code does not say "at least every five years," and his writeup does not say at "least every five years"). Sue Hobart didn't do that. Although I acknowledge that she did not read what was on her slide, she did say once that it was a "stated frequency" and not an "at least frequency." I submit that the addition of the words "at least" by Mr. Banton confuses the issue because when you write a code section with minimum requirements, you don't then want to say "at least." It makes our job as regulators exceedingly difficult. The work that I put into the committee had a lot to do with defining what the frequencies should be. Then I have some of my licensees say, "I know that the frequency stated is the minimum, but we am going to do it better." Then, I wind up at a Congressional hearing, or a public meeting, having to justify why licensee X is doing the tests at least every two years whereas licensee Y is doing the same tests every year (that qualifies as "at least every two years") and licensee Z is doing the same tests every two years, exactly. Why did you rachet licensee Y is a frequently asked question. I have noted the difference in philosophy and would simply ask if they have any further comments on this issue.

HOBART: Dr. Bellamy, I hate to disappoint you but our section of the Code says words equivalent to "at least." We have set a boundary and that boundary we believe, was, in fact, the minimum. But we left to the owners' discretion how often they would do the test provided it was done within the frequency we defined. We believed that there were some mitigating circumstances involved in scheduling. Our equipment, by and large, cannot be tested until there is an outage. Because outage frequencies are different from plant to plant, we tried to set the frequency limit long enough that people

could conduct the test it at least once an outage if they were on an 18 or 24 month schedule. But we left open the option for plants to do the tests more frequently if they had a twelve month outage schedule.

VOGAN: The reason I put "at least" into my writeup is because that is what it really means. I know what happens if you go beyond the required surveillance requirement. The matter you didn't mention that is at issue here is that we did not say 18 months. We said 2 years on a lot of the test frequencies because it was important to identify a current industry trend to go to longer and longer cycles between refuelings. Although the Code is written for nuclear facilities in addition to nuclear power plants, the frequencies that are included in the code are intended for nuclear power plants. It is important because we hope that someday we can use the TA and TB sections in place of current technical specifications. The utilities will then have the option to tear out large sections of the technical specifications and replace them with a statement that they will test in accordance with TA or TB. This is the same that everybody did with Section XI.

MILLER: I would also like to make a comment. This past month we were informed by the ASME that their lawyers have taken a look at the scope statements for the various committees and have decided that the term "minimum requirements" doesn't serve the Society well because in this day and age there are too many people that do not want to see "minimum anything" related to the safety, health, and welfare of the public as it relates to nuclear power plants. Although ASME recognizes that the term "minimum requirement," when specifically used to denote a testing frequency or a thickness of material, is appropriate, they said to us, "Get it out of the scope statements." All of CONAGT's documents have those words deeply imbedded in their scope statements.

PARTHASARATHY: Going through AG-1, and even the Table of Contents, an item that is conspicuous by its absence is filter housings. It there any intent to add this topic at a later date?

VOGAN: The design of filter housings is being treated with ductwork. Basically, the rules for design are the same. There are specific acceptance criteria issues that have been debated such as the integration of some of the filtration equipment into the housings, the doors, etc. At present, the intent is to cover housings under the ductwork section.

PARTHASARATHY: I want to say in followup that I worked in the power plant area before and for the least few years I have been working on DOE plants. Filter housings are items of extreme importance, e.g., cost, space, everything. We are dealing with contamination at every level, not potential contamination. The filter housings are almost a number one priority within HVAC systems. Coupling them up with ducting reduces the perception of their importance.

MILLER: Going back to Susan Hobart's analogy, all this work is done by "tin knockers." I believe that people view ductwork used to exhaust a storeroom a lot differently than they view a filter plenum used to house nuclear filters. Those who have gotten used to the ANSI N509 leakage methodology will find that the same methodology has been carried into the AG-1 Code. You will be able to specify the same high integrity, low leakage plenums for your filter housings using the ductwork, SA Section, in the AG-1 Code. The SA Section is close to Main Committee approval. We want that one approved because that ties everything together.

PARTHASARATHY: I made this comment two years ago, also. Each Code section as you noted in your opening statement, is intended to be used not only by nuclear power plants but in all the nuclear facilities. I think that is appropriate. Most of the discussions and comments come back to nuclear power. I have seen AG-1, it is a very good document, but there may be some areas where not enough attention has been paid to DOE-type facilities.

MILLER: I totally agree with you and I would welcome your addition to our Committee to help us out on that regard. We have one DOE member, we have one DOD member, and we are really hungry for Bechtel/Savannah River people to help us with that part of the Code.

MILLER: I have a question to ask Steve Banton. Does it make sense to add margins to equipment design specification to permit degradation for testing?

BANTON: We didn't want to make the same mistake that B&PV Section 11 did, by putting in degradation margins. It was a good idea when you have no other operability limits that you need to worry about. The trouble most plants ran into with degradation margins is that in most case the point of inoperability was significantly below what was allowed by current technical specifications or regulatory commitments. In addressing TA, we left it up to the owner to specify where the point of inoperability lies.

VERSK: I have two questions on which I need clarification. One is regarding TA 4000 & 5000. Have those Sections gone through all the necessary reviews and balloting?

MILLER: They are currently in process, which means that both the TA and TB Sections in their entirety, including the 4000 and 5000 Sections, are making it through the Main Committee approval process. Both need to have a majority of the members on the Main Committee agree that the Code is ready to go up to the Board for approval, after which it would be issued for publication comment. That is a very important point. Every document has a period for public comments before it is published. If you don't feel that you had enough opportunity to express your ideas to people on the Code Committee, you have an opportunity to get a copy of the document from ANSI or ASME and make your comments. We have to consider comments that come in during the public comment period. To the best of my recollection in 15 years of dealing with the public commenting process, there have been no comments from the public. I don't know if that means people do not care or they do not know that the document is available for public comments. Notice appears in the ANSI REPORTER, and, I think, in Mechanical Engineering Magazine.

VERSK: The other question is regarding welds. Is visual inspection all that is necessary for field welds in accordance with AG-1?

HOBART: For piping systems, we don't specifically address visual inspection of welds. We specify a visual inspection of the whole system and then a hydrostatic or pneumatic test to see whether welds function correctly.

MILLER: I think that is only part of the answer. Steve Banton, is this topic in the general requirements section?

BANTON: After thinking about it, I believe Section SA on ductwork probably covers welding requirements and refers back to the AA Section. Whether it is done in the field or at the shop doesn't matter, visual inspection would be covered by AA. People are using a new document, is a nuclear construction industries group document called NICG-01, for visual inspection.

MILLER: That is a welding acceptance criteria specifically addressed to the type of welds we deal with, not high-pressure welds.

PORCO: In light of the stagnant U.S. nuclear market for commercial power plants, what is the Code Committee doing to incorporate new technology into the developing code?

MILLER: I would like to answer that. We specifically reorganized our Committee about two years ago to create a Technology and Training Subcommittee headed up by Mr. Louis Kovach of NUCON. He had a meeting last Thursday afternoon and he regaled the members of his Subcommittee with some very provocative ideas about new technology matrixes which when prepared will permit us to evaluate whether or not we are really doing the job. My belief is that when he is done, we will see that we have some additional work areas, and not just sideload filters. Louis Kovach, what ideas on new technology should we be looking at?

KOVACH: What we are looking for is to be able to cover the entire nuclear fuel cycle, to look at the technologies that are involved and, in some cases, the application of equipment. As an example, the function of the moisture separator in a conventional air cleaning system is very different from the function of a moisture separator in an evaporator. Or, a HEPA filter that is normally used only under accident conditions is different from a filter in a chemical processing plant where it is exposed to various corrosive chemicals. Therefore, we have to look at both the application of conventional components and the development of new components and devices that are being used in various other applications. The basic questions that come up are, "What are the applications of conventional components that are not now covered by the Code?" and, "What are the components that are being used to handle nuclear safety-related applications of air and gas treatment?" I would state here that a conflict exists with the safety definition. We have legalistic definitions of nuclear safety in power plants which are quite different than for other facilities. In many cases, you have a component used in a safety function in a fuel cycle facility and you may have exactly the same component used in another part of the nuclear fuel cycle which is not safety related. There is no clear definition of safety-related in the rest of the fuel cycle. This makes it difficult to understand what is going to be involved. Generally, I think we are going to be involved with air cleaning trains, and components that are used currently in applications such as reprocessing and fuel manufacturing, or, in some cases, possibly long term depository vent systems. These systems will require continuing revision and addition, to existing codes. The AG-1 doesn't cover half of these potential applications as systems, because it has not recognized them as systems, per se. We have components that are not covered in the component part of the Code. There will be a lot of cases that will have requirements not only for maintenance-type provisions, but for field changes in the performance of the code work that will require additional personnel to become actively involved in this field. Currently, companies are skewed toward conventional power plant air cleaning and air handling systems.

MILLER: We have to have talent on the Committee to do it. That means some of the people sitting in this audience have to help us.

EDWARDS: The ANSI N290 Committee, under the auspices of AICHE, wrote the Standard for Ventilation Systems for Mixed Oxide Plants, (O_2 and PUO_2). We were instructed by AICHE not to just put the document out for comment, but to contact as many people as we knew who were interested in commenting on the document. As a result, we more than 200 unsolicited copies of the draft document to those people we thought would have comments about the document. Many did, and we got some excellent suggestions from those who responded. I feel that CONAGT shouldn't just sit back and wait for the public comment, but that it should actively solicit input from the public.

MILLER: I think you point is well taken and certainly our committees are under instructions to go out and seek expert consultation help whenever they believe their committee needs additional input. You can imagine, that with a Main Committee of 23 members we have difficulty in achieving consensus. Maybe what you are saying is if we got more people involved on a correspondences basis we wouldn't have so much trouble getting approval at the top. I think we should take that into consideration.

Will the AG-1 Code be referenced in the new Advanced Light Water Reactor Plant document that EPRI is putting together?

VOGAN: I think the best way to answer that question is to point to the current revision of Regulatory Guide 1.52. We hope it will reference AG-1 in addition to N509 and in lieu of N510, We have the distinction of having two members of the NRC on the CONAGT writing committees, so I think the information and the sections that we have been developing are well understood by the NRC. I think its reference in Regulatory Guide 1.52 would be how it would be included in the Advanced Light Water Reactor document.

ORNBERG: I have been involved with EPRI in writing the HVAC Section of the EPRI Advanced Light Water Reactor Requirements documents. It contains general design criteria for both the new evolutionary and passive plant designs. AG-1, N509, and N510 are very heavily referenced as

well as current ANS Standards that address HVAC systems both in- and outside containment. They are being picked up. The design criteria document, the requirements document in Chapter 9, is about 50-60 pages long and addresses every HVAC and air cleaning system for every type of space in both PWR and BWR plants.

MILLER: Initially, one of the goals of EPRI was to not use any charcoal or any carbon on these plants. They have revised that position, haven't they?

ORNBERG: I haven't seen it lately. Last March I saw some correspondence that says they did remove the initial design intent. They have a general requirement section of the EPRI requirements document that lays out what their design intentions are. One of their intentions was to have no carbon and therefore it was written that way throughout. I think they have now removed that, or softened it so the issue of whether carbon adsorbers will be required would be evaluated on a plant basis.

BELLAMY: Let me state that the overall philosophy of the NRC is to be able to simply reference industry consensus standards. That way, when we go to hearing or licensing boards we can call you people in as expert witnesses, using our vast subpoena powers. That is a general answer to your question. A specific answer is that the NRC position right now is that we are looking for AG-1 to be completed. It is very difficult for us at this time to reference only specific parts of it in an upcoming revision to the Regulatory Guide 1.52. I am stealing some of my thunder from my talk later on this week. We will be very heavily referencing N509 and N510. I do not believe at the present time there is a reference to AG-1 except noting it. I will go through the process when I give my presentation on exactly how we would love to have your comments. In reply to Mr. Edwards, the mailing list for comments for that proposed revision to the Reg. Guide measures well into the thousands not just hundreds. But I am sure that you will not take this opportunity to load us down with many comments that we don't need.

VERSK: AG-1 does not provide any information on HEPA filter loading e.g., pounds' of dirt accumulated vs. cfm and pressure drop. The above knowledge is essential to determine the number of filters required for exhaust systems.

MILLER: The number of HEPA filter elements in a NACU is typically based on air flow considerations, with each 24" x 24" element rated for 1000 cfm. The large particle dirt is removed by pre-filters suitable sized for specific conditions in the air stream being treated. Depending on the fan pressure/flow characteristic, a HEPA filter bank can be run up to twice its initial clean pressure drop.

OPEN SESSION

EUROPEAN COMMUNITY NUCLEAR CODES AND STANDARDS

Monday: August 13, 1990
Co-Chairmen: W. H. Miller, Jr.
J. L. Kovach

OPENING COMMENTS OF SESSION CO-CHAIRMAN MILLER

EUROPEAN COMMUNITY NUCLEAR CODES AND STANDARDS
M. R. Green

OPENING COMMENTS OF SESSION CO-CHAIRMAN MILLER

On December 31, 1992, twelve European nations comprising the European Community are scheduled to create a single market by eliminating internal trade barriers. Projections set this market's population at about 340 million people, once East Germany is admitted. This group of countries has a gross domestic product of 4.7 trillion dollars. And there are a couple of business men in the audience today that just want a small percentage of that - Ben Franklin is one of them.

The subject of EC 1992 is discussed in newspapers daily. What was labelled "Euroscoliosis" a few decades ago, now appears to be "Europhoria". There is a great optimism that business prosperity in Western Europe will expand substantially once linkage is complete. This situation has not gone unnoticed by industry around the world. The kinds of questions that I would like to explore today, with your help, is, will EC 1992 become a reality as scheduled? Will EC 1992 truly succeed in its objectives in the near term? What will be the impact on the codes and standards and products and services? And what strategies will emerge in non-EC countries to successfully adapt to EC 1992.

There has been a great deal of apprehension around the world that EC 1992 will ultimately create "Fortress Europe," precluding substantial involvement by non-EC members. The signals appear mixed on whether this will or will not occur. But such fears are probably unfounded. EC cannot afford to adopt a protectionist attitude. It needs exports to pay for imports and access to the diversified markets of the world. "Fortress Europe" would impede and possibly destroy Europe's role in global trade.

Now, the European Community currently consists of 12 member states: Belgium, West Germany, France, Italy, Luxembourg, The Netherlands, Denmark, Ireland, United Kingdom, Greece, Spain, and Portugal. Other European countries such as East Germany, Austria, Norway, Sweden, Switzerland, Finland, and Iceland are expected to join in the near future. This powerful economic unit is not a recent formation. Its birth came on March 25, 1957 with six countries signing the Treaty of Rome: Belgium, France, The Netherlands, West Germany, Italy and Luxembourg. The European Economic Community became a legal entity on January 1, 1985. So, I guess that forming the European Community is much like writing codes and standards; it takes 15 to 30 years to actually get the job done. The 1987 Single European Act, established a process for policy setting and decision making. A complex structure was formed to propose, discuss, and legislate the Community's regulations and policies. I thought it would be interesting today to review how they set up an organization to govern this hugh European Community.

The four-part governing system works both independently and as an integrated system. The European Commission is comprised of 17 representatives of all member states and they initiate the policies and they have the power to implement decisions of other entities, as well as to enforce the treaties. The Community's Council of Ministers is the final decision making body with members representing the interest of both their own countries while working toward agreements

benefitting the entire European Community. Five hundred and eighteen elected members of the European Parliament review legislation and offer opinions on various issues. These political representatives of 12 member states watch the conduct of the other governing entities, approve the EC's annual budget, and have the power to dismiss the Commission as a whole. The final leg of the system is the Court of Justice. This is much like United States Supreme Court; the judicial unit interprets EC laws for the national courts, and rules on the legal questions regarding treaties. Member states, community institutions, private firms, and individuals are all permitted to take their concerns to a panel of 13 judges, but they must accept the court's ruling.

There seem to be five principal concerns among members of the engineering and science community in this country. They concern reciprocity, ownership, professional licensing, EC public procurement, and standards. Our guest speaker today is going to speak on standards, and then, I hope, there are members in the audience that will help us discuss some of these other issues.

EUROPEAN COMMUNITY NUCLEAR CODES AND STANDARDS

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Mr. Chairman, other panelists and participants, I appreciate this opportunity to address the broad subject of International Standards. As you are aware, the European Community intends to remove the remaining barriers to commerce among its member states by January 2, 1993. We know that international events have caused slippage in the implementation date; however, the United States must change to interact with the European Community.

In testimony before the House of Representatives, Subcommittee on Science Research and Technology and the National Institute of Standards, I testified on behalf of the Council on Codes and Standards to the effect: ASME is one of the world's largest voluntary standards setting organizations and administers numerous accreditation and certification programs. ASME's codes and standards include criteria for nuclear plants and environment, fasteners, boilers, elevators, etc. ASME publishes about 600 standards. Approximately 4,000 volunteers develop ASME codes and standards through consensus committees and serve on boards and committees regarding accreditation and certification.

After a standard has been approved by an ASME Consensus Committee, it is submitted to ANSI for approval as meeting the criteria as an American National Standard. ASME submits all of its codes and standards to ANSI for consideration.

ASME has developed domestic codes since 1884 and has participated in international standards for testing materials since 1898. From this involvement in domestic and international standards, ASME participated in the formation of both the American Society for Testing Materials (ASTM) and the American National Standards Institute, or its predecessor organization.

During the Kennedy round of tariff discussions, it was alleged that ASME's boiler and pressure vessel accreditation program was a "non tariff barrier" because ASME limited its accreditation of manufacturers to those located within the United States and Canada where there existed provincial, state and municipal government oversight.

The United States sued ASME and The National Board of Boiler and Pressure Vessel Inspectors and resolved the suit with a consent decree that provided for ASME to extend its accreditation and the National Board to extend its registration to the rest of the world. ASME looks upon the United States as the host government and through this relationship receives counsel on international matters that involve ASME consultants, designees, and inspectors that are part of the infrastructure of a world class accreditation program. ASME now accredits manufacturers and suppliers in more than 35 countries and

our marks are recognized in about 80 countries. In Europe, 12 countries, ASME accredits: Belgium 25; France 75; Italy 140; Holland 40; Spain 20; UK 75; Germany 125.

During the early years of the last decade, in its update of its strategic plan, ASME's Council on Codes and Standards recognized the movement toward a global economy and revised its strategic plan to state, in part, "Where ASME domestic codes and standards are preempted by international standards as evidenced by little or no demand for the domestic code(s) or standard(s), the Society through technical advisory groups will continue to support the international activity and may disband the domestic standards activity." The strategic plan continues, "domestic standards committees that are replaced by technical advisory groups will be funded by government, industry and the public." The funding requires government participation which is not now provided in the United States but is common in other countries.

The present participation of the U.S. voluntary standards organization in the international arena lacks strong leadership, the United States government is unique in its lack of financial support to domestic and international standards activities.

During the past two decades, there have been situations where a dominance of power by a few in an essential industry has prevented the U.S. from having American National Standards in areas vital to health, safety and resources. Because American National Standards are normally the basis for participating in international standards, this lack of a forum for consensus prevents the United States from having an effective voice in developing international standards.

There is a compelling need for a level of "due process" that goes beyond the private sector requirements and is supportable in the courts. We strongly believe that only the federal government can provide the forum to resolve those issues when the provisions for "due process" in the private sector have been exhausted. Unless we provide this forum, the United States' interest in international standards and related commerce could be harmed.

Another reason why the federal government needs to be more involved in international standards is that there appears to be little likelihood that U.S. industry will provide the necessary financial support for administration of the U.S. secretariats or the qualified people to represent the United States on technical advisory groups. The United States government should take the appropriate action to assure that Americans are represented by qualified people.

Where negotiations relating to standards and accreditation involve foreign governments, it is essential that the U.S. government provide leadership. Because the various states of the United States vary in their reference to voluntary standards and accreditation programs, federal government involvement is necessary to provide a U.S. position. After participating in a U.S. delegation to Brussels on EC-92 issues relating to standards and accreditation, I feel that U.S. government participation is essential in negotiations with governments on standards and accreditation issues.

Based on the aforementioned observations, we recommend that establishment of a federal standards accreditation and certification institute. This institute should develop through consensus, the criteria for standards and accreditation bodies and audit accredited bodies to assure compliance. Such an institute must have oversight and investigative powers to provide for a level of "due process" that is needed to assure lack of bias and competitive opportunity. Membership in the institute should be required only of standards developers and accreditation administrators whose programs are referenced as a means of satisfying regulatory requirements.

This institute would also provide the necessary forum to develop U.S. positions on standards and accreditation issues and represent the U.S. in negotiations with other governments. Such an institute may mean the difference between a piecemeal and disorganized private sector approach to international standards and a healthy worldwide system for standards, accreditation and certification programs.

Although the ASME Council on Codes and Standards feels an urgency to have its recommendation implemented, it recognized that an overall study may be needed. In testimony before the House Subcommittee on Science, Research and Technology, we would support a study Commission created by Congress and appointed by the President. We feel that such a Commission must have a balanced membership from nominees from state, local, and federal government; standards developers; and the public interest groups. Due to the urgency of the problem, authorizing legislation should be as soon as possible and this Commission should make its recommendation within one year of its creation.

We emphasized that our recommendation is a concrete recommendation to help strengthen the consensus standards process through oversight and support; it is not an invitation for government takeover of the private standards process in the United States.

ASME has benefitted from government participation in its codes, standards and accreditation programs - including oversight through audits.

On June 19, 1990, Dr. Robert W. White, Undersecretary for Technology, Department of Commerce, addressing the House Committee on Science, Research and Technology Committee stated, in part, that the Department of Commerce intends to conduct a series of workshops with members of specific sectorial manufacturing parties. These workshops would be held to explore the possibility of developing a national laboratory accreditation scheme for conformity assessment that is consistent with internationally recognized technical as well as legal criteria.

Since June 19, 1990, the American Association of Engineering Societies sponsored a forum attended by the major technical society standards developers and it was concluded that the U.S. government should study the issues involving domestic and international standards. Also, ASME has agreed to be a co-sponsor of a workshop on boiler and pressure vessel accreditation. The purpose of the workshop is to develop criteria against which the DOC can accredit ASME as a notified body for Boiler and Pressure Vessel Accreditation. A notified body is an organization designed by a European country to implement an EC directive.

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For the past twenty years, nuclear codes, standards and related accreditation activities have been a major thrust of ASME programs. In some areas, such as quality materials manufacturers accreditation, more than 50% of the accredited organizations are located outside North America.

Because Europe intends to use International Standard Organization (ISO) and International Electrotechnical Commission (IEC) standards as the basis for CEN and CENELEC standards, it is important that we have input. The mechanism for issues resolution to provide for this input must be put in place through government/private sector partnership.

DISCUSSION

MILLER: As I understand it, one of the reasons that the United States doesn't do well in discussions with the EC 1992 is that there is no one person, or one body that can speak for all the codes and standards organizations. Now one might think that should be ANSI, but, evidently, there are some problems with that. Would you care to talk about that?

GREEN, M: The American National Standards Institute has the role of coordinating standards in the United States and also being a member body of the International Standards Organization, ISO, and the IEC. But once you move out of the U.S., even to Canada; the Canadians, Europeans, they want a government-related organization to negotiate with. I recently was at a meeting with the Canadians on the Canadian-U.S. Treaty and they made it very clear that the U.S. has no focal point for them to negotiate with. I expect that the Canadians are also going to petition the U.S. Government for remedy to this situation. The Europeans make no bones about it. When you are a delegate for the U.S. they will tell you, "until you fellows come over here, we have no one to talk to." They are not coming to ASME, ASTM, or ANSI and talk. And I know these private sector organizations have their offices in Brussels, and London, and they think they are getting within the continental market, but the Europeans look at it much differently. They are looking for the U.S. to set up an Institute. Now I must give credit to the Department of Commerce, there are people down there that are trying to help the private sector. They are promoting the standards. I receive letters from Europe asking us to put our boiler and pressure vessel code into ISO. Well, the Department of Commerce is going a different route; it is suggesting that the boiler and pressure vessel code and the related accreditation part of it be accepted by the European Community as a means of satisfying their regulatory requirements. The DOC is providing leadership in that area, but as for an Institute we don't have one.

MILLER: For us to get involved in ISO standards, it means that the individuals that participate are going to have to travel to Europe in order to attend meetings and we know from our past experience from representatives on ISO efforts that it is just impossible for private industry to support that kind of an effort unless they can really see a return on their investment and right now it is a little unclear.

GREEN, M: That is correct. Many of you probably know Dr. Cooper. Dr. Cooper spent a lot of time trying to get parts of ASME B&PV Code Section III, the nuclear section of the boiler code, accepted by ISO. He said to our Council on Codes and Standards the big problem is to get the right people there to represent the U.S. Quite often you can get the fellow who has the financial wherewithal to go but you don't have the right person there. That is what the U.S. needs when they bargain with the Europeans on the international scene.

MILLER: Are there any members of the audience who have some contributions to make especially if we have some representatives from Europe here, could you shed any further light on what is going on in Europe these days on codes and standards?

KOVACH: First of all, if we are going to have a government agency handle standards, we might as well start with some organization as efficient as the Post Office. They only work efficiently and they do such a good job. They would be a good agency to start with in trying to standardize. In discussing your contribution, you made a little jibe about Hungary, I want you to know that even East Germany found that the way to the common market was through Hungary.

The problem, I think, is a little more complicated; at least from the standpoint that in some areas in the common market, already, there are regulations coming into force which require that certain equipment is sold only by accreditation in, and, solely by, agencies located in the common market areas. And I can see a two-fold problem, one relating to the multi-national organizations that are involved in

activities both in Europe and in the U.S. These organizations, generally, can handle some of these activities much easier; whether they have to test a component in Europe separately, even if they test it in the U.S. also. They have no problem with this. However, I can see significant problems for some of the smaller organizations that make specific items in one country or another, which may not get a proper hearing or proper accreditation at the normal business expense that is currently part of doing business in various parts of the world. From this standpoint, the activities of the Commerce Department are greatly favored by, and promoted by, some of the smaller companies. I know a number of organizations that are actually pushing for the Commerce Department's activity to accept some of the U.S. standards in the countries of Europe, in addition to China, on the basis that it saves also a significant amount of effort in Europe if there is no organized standard in existence at the present time. The question that comes up that there may be 90% of a standard completely acceptable to the Europeans as it is, but they have difficulty with maybe a 10% part of the standard that we have. To assure that these standards can be accepted in Europe, we have to realize that we have to be flexible, also; whether it is at ASME or ASTM. We are working on an ASME code currently which is basically still in English units, on the CONAGT section at least. Some part of it is being switched to the metric system or international standard units also. The areas where we have to contribute is to make sure that any of our standards that we are trying to complete for the rest of the world to consider are reasonable for them and not exclusionary from their standpoint also. But it has to work two ways. I really don't see any way that we will be convinced that we will take only the European standards and we will make them apply in the U.S. and I think it is equally foolish for us to believe that the rest of the world will take ASME or ASTM as a whole and accept it as their standard also. Somewhere some merger of these two will have to come up. I, personally, feel that in any area the more we involve the government, at least from the U.S. end, the more difficult it will be to achieve such an action. The way I would propose is that the U.S. Government assign some sort of an official veil or mantel over a group representing the standard writing organizations solely for the negotiation relating to the standards. That I can foresee much easier than to set up another bureaucracy and train people within the government now, who don't know beans about the standards, to negotiate for us. That method I can see creating all sorts of problems. I think that just as there are experts at various U.N. groups assigned and in a semi-governmental official status, I think that the various standards organizations of the U.S. can put together a group that could have a semi-official status representing the U.S. in this type of activity.

MILLER: Now you have been involved in working with OECD haven't you, Louie, in the past and generating some of the documents that they are using for air cleanup.

KOVACH: Yes. There is no difference in an international consensus activity than in the U.S. There are also people in international groups that speak a different language, not only in the US ASME group.

MILLER: Are the Europeans using N509, N510, and AG-1? Do you have any idea what they are using? The Germans, of course, are using German standards, I suspect. But what about other European countries?

KOVACH: You have several of the countries that are using either the spirit or the format or, in fact, following N509 and N510 fairly closely. I think that we were remiss in some cases as we were revising those issues not to have sent it to those countries and ask them for input. Anybody who uses a standard, after awhile, knows what is wrong with it. I think, again, if we want to go into international activity, before we issue a new version of N509 or N510 we should go back to The Netherlands, Belgium, Italy, or Spain and ask them for input because they work with it also. Maybe they see something that we don't. This is the sort of input that we need.

MILLER: Earlier this afternoon, James Edwards admonished me for not seeking broad enough comments on our AG-1 code and I think I am hearing the same international flavored message

this evening, and that is; we have got to find ways to improve our global communications if we expect our standards to reflect the global spirit.

KOVACH: I will have you an example. The Europeans are doing a tremendous amount of work in containment venting. Now, I think it would be extremely presumptuous on the part of CONAGT to write a standard or a code on how to design, build, and test containment venting systems when we don't have any in the U.S. This would really leave us in the dark.

KUMAR: Just a thought about how to get into the European Community. Go fullblown in the metric way, even though some of the private manufacturers are taking steps, as a national policy we are not really going fullblown. That might be of some help. I would appreciate any comments.

GREEN, M: Those of you that are familiar with the ASME boiler and pressure vessel code, probably recall that in 1983 we published two ASME boiler and pressure vessel codes. On July 1, 1983 the customarily "foot-pound" version became effective. On October 1, 1983 the "SI base" version became effective. That was premised on Canada making it mandatory that any document that they referenced as part of their laws, would need to be with SI units. That was supposed to take effect on October 1, 1983. A conservative government was elected in Canada, and that regulation never was implemented. As a result, we were able to provide one complimentary copy of the SI version of boiler code in one Province of Canada and we sold very, very few. When I was in Brussels, this subject came up. The Europeans alleged that the Americans want us to recognize their standards, but you don't consider the units we have. I reminded them of the 1983 boiler code and that the Europeans did not buy the 1983. They said, "you used the wrong kind of metric; SI is not what is used in Europe." Then when you go to Japan, also. The Japanese did not buy the SI version of the boiler code. Those 1983 boiler codes are lying in bookstores all over the world. Some day they may be worth some money for archives, but they were not used by engineers. Our experience right now does not justify an SI code, unless we only publish an SI version and did away with the customary altogether. All other ASME standards have soft. The Boiler and Pressure Vessel Code has conversion units in the Appendix. That is our experience with SI and the Boiler code.

KOVACH: I have one special request of our speaker. If the Justice Department is overlooking now the accreditation in a technical field as their first priority, could we get ASME to overlook their accreditation of lawyers?

GREEN: I can't comment on that. The Justice Department has served us well for 18 years.

MILLER: One of the other issues that I talked about earlier today had to do with the licensing of professional engineers. That has been a concern that the European Community might adopt licensing requirements that pre-empt engineers from the U.S. participating in design work overseas. It is expected that the EC 1992 implementation will result in a much freer movement of engineers between countries within the european community. Just recently it has been learned it looks like, that as long as a U.S. engineer has graduated from an ABET accredited college or university, and has taken the EIT exam that is administered by the National Council of Engineering Surveyors, that will be accepted by the European Community. I suspect that professional accreditation then is going to cease to be of concern. What about goods and services? Those people in the audience who sell goods and services, like the American Air Filters Ellis & Watts, have you sold that many services or goods in Europe where you are going to refocus your market strategies?

FRANKLIN: We have a number of plants overseas. We have a company in England, for example, and they are doing the containment coolers for Sizewell. They are using ASME Section III, right now. They are building it themselves and they are using those guides but they are not getting an "N" stamp and this seems to be acceptable there. In Spain, we have a company who make some of the

nuclear housings and they use whatever codes Spain uses. We have a factory in Emmon, Holland, making HEPA filters, etc. and they are abreast, if not ahead of what HEPA filters are in the U.S. We have a company in Singapore who are making HEPA filters there. We have a licensee in Japan that is making some of their HEPA filters in Japan. Just speaking of nuclear-related items, our international companies are using the local codes that are in existence. In some of the plants, like Spain, where we used nuclear codes U.S. ANSI N509 and N510 they are making some of those things to that code. Some of the spares are being bought from the United States. We are doing some work in Korea, Taiwan and elsewhere in the Far East to ANSI N509 or N510 because the architect/engineers are using those codes.

MILLER: I read recently that some companies are considering setting up small operations in an European country just so they get the toe-hold in the European Community for 1992. Is that something that is reasonable?

FRANKLIN: Yes, I would think so because any country would rather have localized manufacturing. We tried to do this in China for commercial work. In Mainland China they wanted us to build plants there but to sell to the U.S. not to sell to the local population - to generate more dollars. In many countries you can't get profits out of the area. It is preferable to try to get local participation - to manufacture locally. You do it through offering advice in their technical licensing agreements and royalty agreements, etc., that can be made.

GREEN: I think you have heard both sides of the government issue. My associate, Mr. Kovach, feels like it would be like the Post Office. I feel like we need the level of due process so that we can have a U.S. position. I am not suggesting a take-over. In fact, that would be a big loss to the Association with whom I am employed for a long time if the government took over voluntary standards. So, keep in mind the level of due process, so that the U.S. can have a position. On those rare situations where we cannot come to a consensus, there needs to be an independent body to provide due process.

MILLER: What I have learned in preparing for this session is that we haven't heard the last of EC 1992. I think that there is going to continue to be some real tough discussions among our Department of Commerce, and major industries. You had mentioned that you are meeting with IBM next week and you have met with Westinghouse, so Mel Green is taking his crusade on the road and trying to make sure that people understand where we have a vulnerability in our representation in the European Community.

SESSION 4

CHEMICAL PROCESSING OFF-GAS CLEANING

Tuesday: August 14, 1990
Co-Chairmen: R. T Jubin
A. C. Vikis

OPENING COMMENTS OF SESSION CO-CHAIRMAN VIKIS

RETENTION AND MEASUREMENT OF IODINE-129 AND OF ORGANOIODINE IN THE OFF-GAS STREAMS OF THE KARLSRUHE REPROCESSING PLANT WAK
F. J. Herrmann, V. Motoi, B. Herrmann, A. van Schoor, D. Fang, H. Fies

IODINE-129 DISTRIBUTION AND RETENTION DURING EVAPORATION OF MWL SOLUTIONS
J. Amend, V. Motoi, F. J. Herrmann, J. Furrer

TECHNICAL-SCALE IODINE EXPULSION FROM THE DISSOLVER SOLUTION AND BALANCE STRIKING FOR LIQUID AND GASEOUS IODINE FRACTIONS
J. Furrer, H. Deuber, A. Linek, R. Kaempffer, K. Jannakos

ELIMINATION OF IODINE FROM RADIOACTIVE GASEOUS EFFLUENTS IN A PACKED COLUMN
B. Vignau, J. P. Goumondy, M. Roustan

IODINE AND NO_x BEHAVIOR IN THE DISSOLVER OFF-GAS AND IODOX SYSTEMS IN THE OAK RIDGE NATIONAL LABORATORY INTEGRATED EQUIPMENT TEST FACILITY
J. F. Birdwell

DEVELOPMENT OF A PROCESS FOR ADSORPTIVE SEPARATION OF KR-85 FROM THE OFF-GAS OF NUCLEAR FACILITIES
H. D. Ringel, B. G. Brodda, T. Burbach, R. J. Printz

CLOSING COMMENTS OF SESSION CO-CHAIRMAN JUBIN

OPENING COMMENTS OF SESSION CO-CHAIRMAN VIKIS

Chemical Processing Off-Gas Cleaning is normally associated with the dissolution of irradiated fuel for advanced fuel cycle fuel reprocessing and for military applications. Typical off-gas cleaning requirements include the removal of volatile radionuclides (I-129, Kr-85, C-14 and tritium) and semivolatiles (such as ruthenium) as well as NO_x associated generally with the dissolution of fuel in nitric acid. Iodine, because of its quantity and radiotoxicity, has received most of the attention and, thus, five of the six papers this morning deal with iodine removal. The sixth deals with krypton-85. I should note that all but one of the six papers come from European colleagues, who have traditionally been strong on this topic. I should also note R&D in this field appears to be slowing down; largely, as a result of the present uranium glut and thus a deferred need for uranium economy, but also as a result of the relevant technology reaching a stage of maturity.

RETENTION AND MEASUREMENT OF IODINE-129 AND OF ORGANOIODINE IN THE OFFGAS STREAMS OF THE KARLSRUHE REPROCESSING PLANT WAK

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Abstract

In the Karlsruhe Reprocessing Plant WAK more than 95 % of the iodine-129 inventory of the spent fuel is expelled during dissolution into the dissolver offgas and is fixed on iodine filters. Less than 5 % enters the PUREX process.

The distribution of the iodine-129 in the offgas streams is the following:

- 0.01 % in the dissolver offgas downstream the iodine filter;
- 0.4 to 0.5 % in the vessel offgas of the PUREX process;
- 0.4 to 0.5 % in the vessel offgas of the MAW storage facility;
- 0.1 % in the vent air of the dismantling cell;
- 0.01 % in the vessel offgas of the HAWC storage facility downstream of the iodine filter.

About 85 % of the iodine in the WAK vessel offgas streams has been determined to be bound to organic compounds.

Characterization of these organoiodine compounds yielded n-dodecyl iodide as the main constituent; in addition, short chain alkyl iodides, nitroalkanes and esters of nitric acid have been detected.

Thermodynamically, the organoiodine compound CH_3I is removed without problems both on silver nitrate and on elemental silver. On the other hand, under WAK operating conditions inorganic iodine is not removed on silver nitrate, but solely on metallic silver.

I. Introduction and Objective

As reported at the 20th Air Cleaning Conference ([N]) almost complete iodine-129 retention had been the target design limit for the German reprocessing plant to be built at Wackersdorf.

At the Karlsruhe Nuclear Research Center and in the Karlsruhe Reprocessing Plant WAK test programs were performed which had shown the conditions of iodine-129 release of less than 50 mCi/a.

For a plant throughput of 500 metric tons per year a 50 mCi/a release can be guaranteed only if less than 0.2 % of the iodine inventory in the spent fuel reaches the biosphere.

This release of less than 50 mCi/a can be only achieved if more than 99.9 % of the iodine is expelled during dissolution and fixed with a decontamination factor (DF) of $\geq 10^3$ on iodine filters provided in the dissolver offgas.

If this 99.9 % iodine removal cannot be guaranteed iodine filters must be provided for all iodine carrying gas streams.

With regard to the objectives above an experimental program was performed at WAK which was geared to:

- almost complete iodine expelling during dissolution;
- investigation of the iodine paths in the PUREX process;
- determination of the iodine-129 distribution in the WAK offgas streams;
- characterization of the organoiodine compounds isolated from the WAK offgas;
- thermodynamics of iodine removal under WAK operating conditions;
- reaction mechanism of iodine removal under WAK operating conditions.

In order to attain these objectives sampling devices were developed, especially a cooling trap, which allows the condensable fractions of the offgas to be trapped over several days without requiring maintenance work. Moreover, various sorption materials were developed and tested for their suitability to remove iodine from the offgas. The problem was that very low iodine-129 concentrations of 10^{-9} to 10^{-6} g/m^3 had to be measured in the offgas.

The investigations reported here were made on the combined offgas systems (dissolver offgas, vessel offgas of the PUREX process, and vessel offgas of the medium level liquid waste storage facility) of the Karlsruhe Reprocessing Plant WAK.

N. F.J. Herrmann et al
Testing an Iodine Filter for the Vessel Offgas of the German Industrial Scale Reprocessing Plant
20th DOE/NRC Nuclear Air Cleaning Conference

II. Iodine-129 Distribution in the WAK Offgas Streams

Figure 1 shows the four WAK offgas systems:

- the dissolver offgas with the iodine filter (which contains AC 6120, a silver impregnated SiO_2 base catalyst carrier, as the sorption material);
- the vessel offgas of the PUREX process with a test filter for iodine retention (AC 6120) in a 10 % bypass line;
- the vessel offgas of the medium level liquid waste storage facility;
- the vessel offgas of the high level waste storage facility with the iodine filter (AC 6120).

In addition, the percentage distribution of iodine-129 in the offgas streams has been entered in Fig. 1. It appears that more than 95 % of iodine contained in the spent fuel is expelled during dissolution into the dissolver offgas and retained on the iodine filter ([N]); up to 3 % reach the liquid wastes and approximately 1 % leaves the plant via the gas path.

The 1 % of iodine-129 just mentioned is distributed among the individual WAK offgas lines (Fig. 1) as follows:

- 0.01 % in the dissolver offgas;
- 0.4 to 0.5 % in the vessel offgas of the PUREX process;
- 0.4 to 0.5 % in the vessel offgas of the medium level liquid waste storage facility;
- 0.1 % in the vent air of the fuel dismantling cell (not represented in Fig. 1);
- 0.01 % in the vessel offgas of the HAWC storage facility (this offgas has been passed through an iodine filter).

The iodine-129 distribution described above is correlated to the process:

- During long periods of intervention the portion of iodine in the vessel offgas of the PUREX process decreases because there will be no transfers and no evaporator operations.
  Thus the I-129 entrainment decreases during about 5 months by a factor of 10 during outage times (from approx. 40 Bq/m^3 to 2 to 5 Bq/m^3).
  This "bleeding" is attributed to the solutions still present in the system, especially to the organic solvent.
- The contribution of the vessel offgas of the MAW storage facility is quasi-constant and does not diminish during intervention periods because the storage facility is operated independently of the PUREX process and always includes iodine-129 containing organic and aqueous wastes.

N. The iodine-129 concentration in the dissolver offgas (upstream the iodine filters) is 10^5 Bq/m^3 during dissolution. When dissolution is completed, it takes another 2 to 3 weeks until the value reached is approximately 10^3 Bq/m^3 which further decreases with time. When the sampling pipe is heated the iodine concentration rises sharply. This is attributed to the deposits of iodine on the pipe walls.

III. Discrimination of Inorganic Iodine in the Offgas

A discriminating sampling for inorganic iodine in the total offgas of WAK was made with the DSM 11 sorption material (an SiO_2 base catalyst carrier with 6 % Al_2O_3 and 7 % KI impregnation) during an outage phase. This iodine sorption material is only suited for elemental iodine sorption but not for sorption of organoiodine ([N]).

The outage phase had been selected for the test because with DSM 11 no suitable results are obtained in offgases containing NO_2. During periods in which no dissolving campaigns are performed the NO_2 content of the WAK offgas is < 5 vpm whereas during dissolving operations it may increase to approximately 10,000 vpm.

A partial stream of the total offgas from WAK was passed over the following sorption materials at a rate of 500 l/h (see Fig. 2): four beds with DSM 11 at 80 °C, four beds with AC 6120/12 % Ag at 80 °C, four beds with charcoal impregnated with 1.5 % KI (SS 207 B) at 80 °C, four beds with AC 6120/12 % Ag at 180 °C.

Each filter bed was 32 mm in diameter an 50 mm in depth.

The γ-spectrometric evaluation of the filter beds after 17 days of sampling yielded the following percentage distribution of iodine-129: 15 % on the DSM 11 beds at 80 °C, 78 % on the AC 6120 beds at 80 °C, 2 % on the charcoal beds at 80 °C, and 5 % on the AC 6120 beds at 180 °C.

As a result of these measurements approximately 15 % of the iodine present in the WAK offgas occurs as inorganic iodine.

This result has been confirmed by iodine sampling with an alkaline wash column over a period lasting several weeks.

IV. Contribution to Characterizing Organoiodine Compounds

A 300 to 400 l/h partial stream of the WAK total offgas was passed through a cooling trap (sampling point K1 in Fig. 1). It can be seen from Fig. 1 that the dissolver offgas has already passed the iodine filter.

The temperature of the gas leaving the cooling trap was about -20 °C.

The iodine-129 content in the vessel offgas from the PUREX process of WAK was approximately 40 Bq/m^3. This corresponds to an iodine partial pressure of 7.5×10^{-10} atm (as alkyl iodide).

Due to the different volatilities of methyl iodide and dodecyl iodide the dodecyl iodide as well as long chain alkyl iodides should be preferably removed at -20 °C. Short chain alkyl iodides remain preferably in the gas phase.

It can be expected that some of the volatile alkyl iodides condense too provided that they are miscible with the dodecyl iodide.

An iodine sampling filter was connected downstream of the cooling trap in order to be able to measure the iodine-129 fraction passing through the cooling trap (Fig. 3).

N. H. Deuber
Aspects of Retention and Differentiation of Gaseous Radioiodine Species
Paper presented at the Boris Kidric Institute of Nuclear Sciences, Belgrade (Yugoslavia), Sept. 1981

Generally, approximately 800 ml of aqueous phase and 5 to 25 ml of organic phase are condensed after 30 to 40 m^3 offgas have passed through the trap. During operation periods, a value of 0.5 to 0.8 ml of organic phase per cubic meter of offgas is found whereas during outage periods the value attains < 0.2 ml/m^3. The iodine-129 concentration in the organic phase is 10,000 to 20,000 Bq/l.

About 70 % of the total amount of iodine-129 in the offgas is fixed on the iodine sampling filter downstream the cooling trap.

The isolated organic phase was analyzed by gas chromatography at the Hot Chemistry Institute (IHCH) of the Karlsruhe Nuclear Research Center. n-dodecyl iodide has been identified as the main constituent of the organoiodine compounds, but also alkyl iodides with 2 to 11 carbon atoms, butyl iodide occurring at a slightly elevated concentration relative to the neighbouring alkyl iodides. Besides the alkyl iodides, nitroalkanes and esters of nitric acid of various carboxylic acids were detected depending on the operating conditions.

Batch distillation in the vacuum of the organic phase collected in the cooling trap has further contributed to characterizing the organoiodine compounds from the WAK offgas.

About 17 % of the total iodine-129 quantity was detected in the sump and in the first fraction, respectively. The rest of iodine-129 was distributed among eight intermediate fractions. During distillation the temperature increased by 9 °C.

The results confirm that the organoiodine compounds condensed from the offgas contain, besides the low volatile dodecyl iodide in the sump, also iodine compounds which are more volatile than dodecyl iodide.

V. Termaodynamics of Iodine Removal under WAK Operating Conditions

Contrary to information from the literature, experience at WAK shows that the effective substance for iodine sorption on AC 6120 is not the silver nitrate but the elemental silver.

The iodine filter materials used in the WAK offgas lost about 7 % of their weight after short service lives. Some of the particles shone like silver whereas some others looked grey-black.

Therefore, the thermodynamics of iodine removal on $AgNO_3$ and Ag-impregnated sorption materials was studied considering the WAK operating conditions. The result will be briefly reported here:

Possible reaction equations describing the removal of gaseous inorganic iodine (I_2) and of organic iodine (e.g. CH_3I), respectively, on solid silver or silver nitrate were established. Then the iodine equilibrium pressures pI_2 and pCH_3I, respectively, were calculated.

Thermodynamically, iodine can only be removed if the iodine concentration in the WAK offgas exceeds the value of the calculated iodine equilibrium pressure.

The straight lines in Figs. 4 and 5 represent the iodine equilibrium pressures as a function of the temperature. The detected iodine partial pressures in the WAK offgas (dissolver offgas, DOG, during dissolution of fuel from the German reactors KKS and MZFR and during intervention period, IP; vessel offgas, VOG) for temperatures between 135 and 145 °C are also entered in these figures.

The Fig. 4 shows that the partial pressures of I_2 and CH_3I, respectively, calculated at 413 K (140 °C) for the reaction of metallic silver with gaseous I_2 and O_2, with gaseous CH_3I and with gaseous I_2, respectively, are all lower than the iodine pressures detected in the WAK offgas. This means that thermodynamically iodine is removed in all three cases on metallic silver. These reactions are not influenced by the NO_2 concentration in the WAK offgas.

Looking at the reaction of silver nitrate with gaseous iodine one sees that the iodine partial pressures in the WAK offgas attain the same order of magnitude as the calculated iodine equilibrium pressures at 413 K (140 °C). That means that thermodynamically no iodine is removed on $AgNO_3$ effectively.

For the same reason no iodine is removed from the vessel offgas containing more than 300 vpm NO_2 (Fig. 5).

The reaction of $AgNO_3$ with I_2 forming $AgIO_3$ and NO_2 is not shown in Fig. 5. For this reaction the calculated iodine equilibrium pressures at 413 K (140 °C) are of the same order of magnitude as the iodine partial pressures measured. For this reason this reaction is neither possible in terms of thermodynamics.

Thermodynamically, the organoiodine compound CH_3I can be removed without problems both on $AgNO_3$ (not shown in a figure) and on metallic silver (Fig.4). The calculated iodine equilibrium pressures in these cases attain orders of magnitude which are less than that of the iodine concentration in the WAK offgas.

Kinetics provides an answer to the question whether thermodynamically possible reactions will proceed at the required rates. Due to a kinetic inhibition or due to a very slow reaction rate, a reaction which would be possible thermodynamically does not take place.

The finely distributed silver with a large surface which develops on the sorption material from the $AgNO_3$ impregnation should be advantageous in kinetics terms.

VI. Reaction Mechanism of Iodine Removal under WAK Operating Conditions

A reaction mechanism for the removal of inorganic iodine as I_2 and of organic iodine as CH_3I on $AgNO_3$ impregnated sorption material under WAK operation conditions will be considered below:

1. Inorganic iodine

In contrast to data published in the literature, the results of the thermodynamic calculations and of the experience accumulated at WAK have shown that under WAK operating conditions no inorganic iodine is removed on $AgNO_3$ but rather on elemental silver while forming AgI and $AgIO_3$.

From the thermodynamics point of view silver nitrate cannot undergo thermal decomposition into Ag, NO_2 and O_2 in the WAK offgas. $AgNO_3$ is probably decomposed by reduction with organic compounds. Dodecane, which is present in the dissolver offgas too, might play an important role in that process.

The reaction mechanism which governs the removal of inorganic iodine on $AgNO_3$ impregnated sorption material can be expressed by the following equations:

$AgNO_3(s)$ <==> $Ag(s) + NO_2(g) + 1/2\ O_2(g)$
$Ag(s) + 1/2\ I_2(g)$ <==> $AgI(s)$
$Ag(s) + 1/2\ I_2(g) + 3/2\ O_2(g)$ <==> $AgIO_3(s)$
$AgIO_3(s)$ <==> $AgI(s) + 3/2\ O_2(g)$ (very slow)

The decomposition of silver iodate in the reaction $AgIO_3(s) \rightarrow AgI(s) + 3/2\ O_2(g)$ occurs at temperatures $\geq 127\ ^{o}C$; under WAK operating conditions it must therefore be expected to take place for reasons of thermodynamics. According to literature data the rate of decomposition of $AgIO_3$ is slow.

2. Organic Iodine

Thermodynamically, organoiodine compounds in the form of alkyl iodides (e.g. CH_3I) can be removed both on silver nitrate and on elemental silver under WAK operating conditions.

$AgNO_{3(s)} + CH_3I_{(g)}$ <==> $AgI_{(s)} + CH_3NO_{3(g)}$
$Ag_{(s)} + CH_3I_{(g)}$ <==> $AgI_{(s)} + 1/2\ C_2H_{6(g)}$

It is understood that the reaction mechanism is much more complex than indicated here.

VII. Conclusions

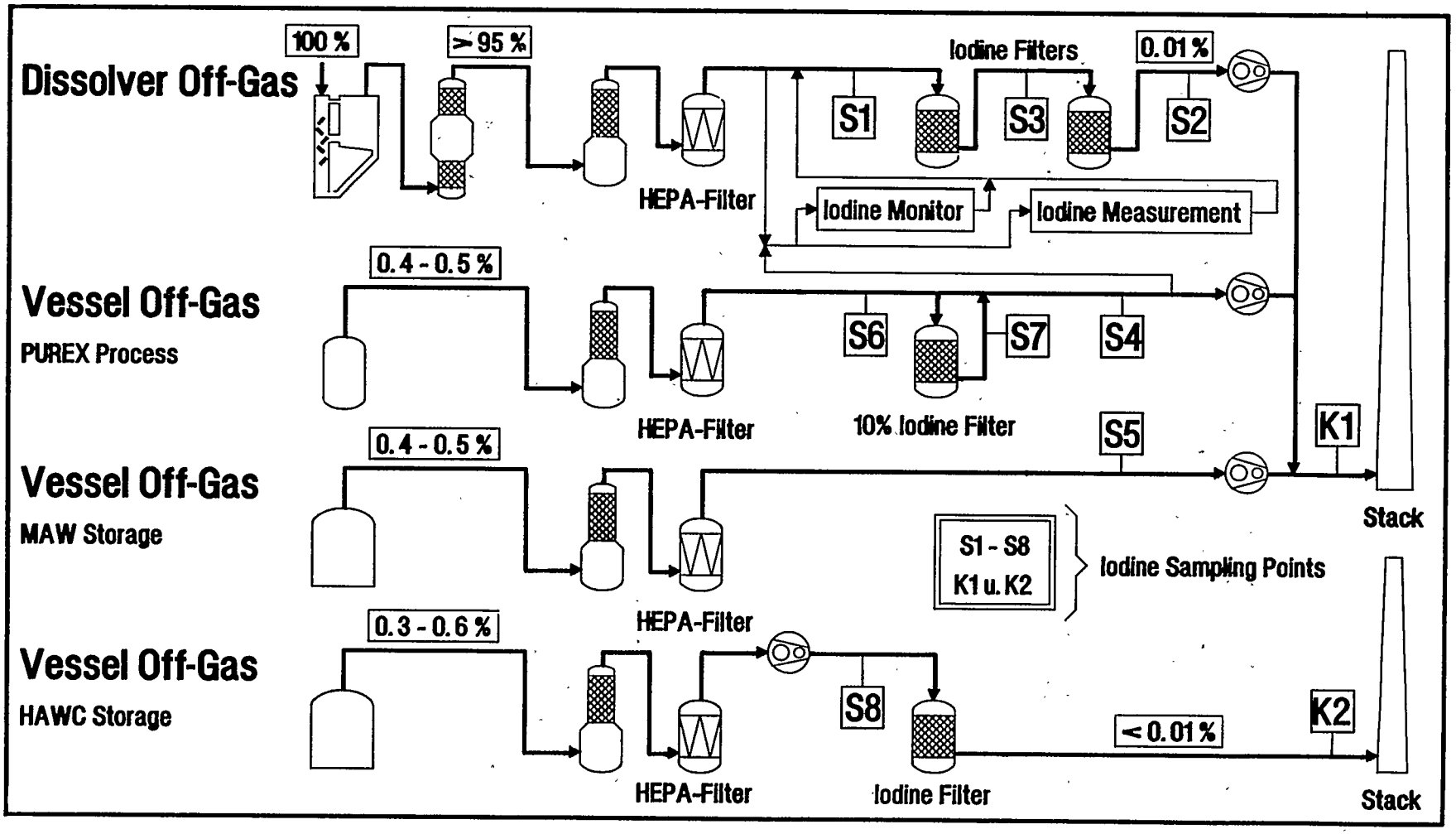
The investigations made at the Karlsruhe Reprocessing Plant WAK have demonstrated that the iodine-129 must be expected to occur in all offgas lines of a reprocessing plant.

In order to retain iodine almost completely all offgas lines must be equipped with iodine filters.

In the WAK vessel offgas streams about 85 % of the iodine has been determined to be bound to organic compounds.

No problems are anticipated in the retention of organoiodine compounds on Ag- and $AgNO_3$-impregnated catalyst carriers, respectively.

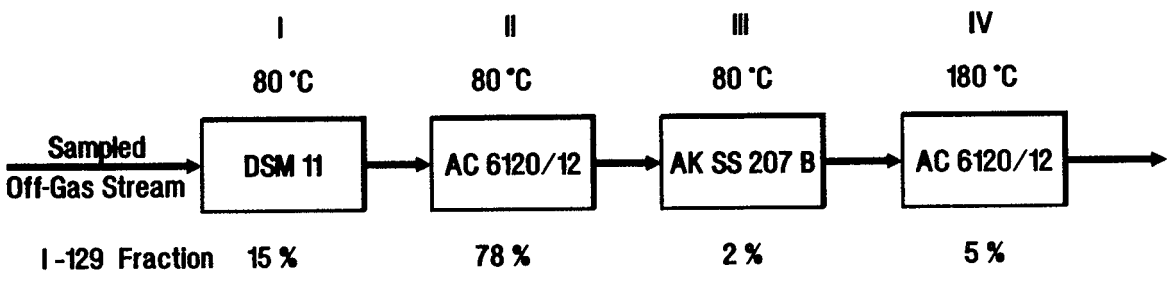
Although the results described are no longer of direct benefit to a German reprocessing plant, they nevertheless offer an orientation for other reprocessing plants if nearly complete iodine retention is required.



WAK

Off-Gas Cleaning in WAK

WAK : ACC/90			
Fig. : 1			
drawn	9.7.90	RKr.	Drawing - Nr.
chckd.	9.7.90	VM.	VM9003



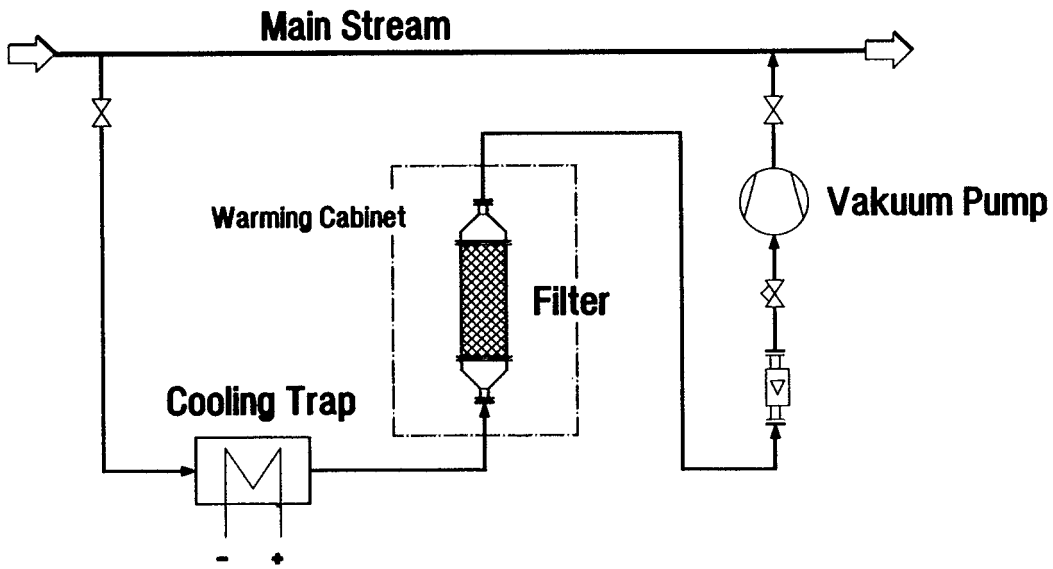
Test conditions :

- Sampling during outage	(NO_2 - content <5 vpm)
- Sampling rate	550 l/h
- Number of Beds	4 for each per location I, II, III, IV
- Diameter of Bed	32 mm
- Depth of Bed	50 mm
- Duration of sampling	17 days

WAK

Discriminating Iodine - 129 Sampling

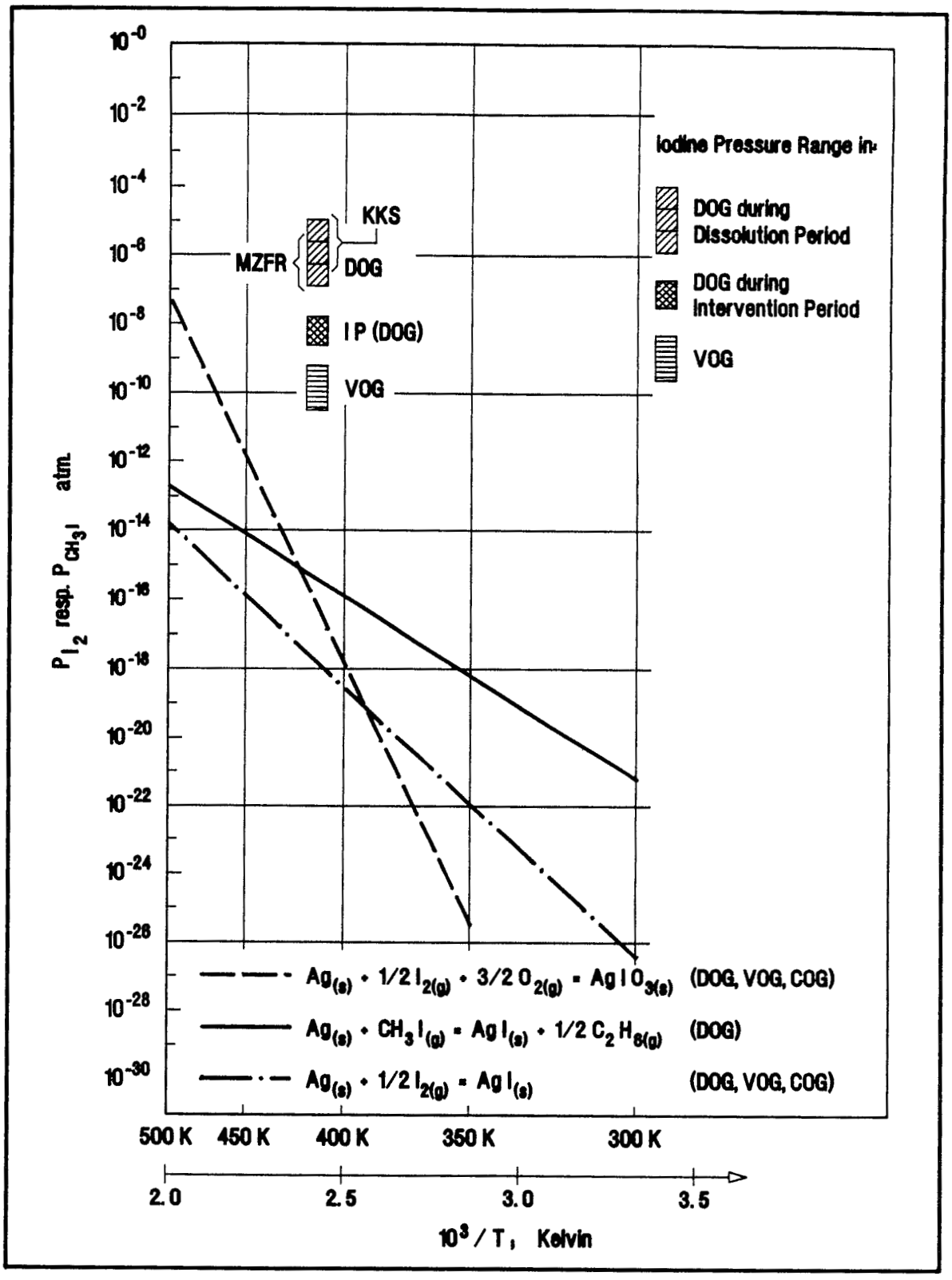
WAK : ACC/90			
Fig. : 2			
drawn	9.7.90	RKr.	Drawing Nr.
chckd.	9.7.90	VM.	VM9005



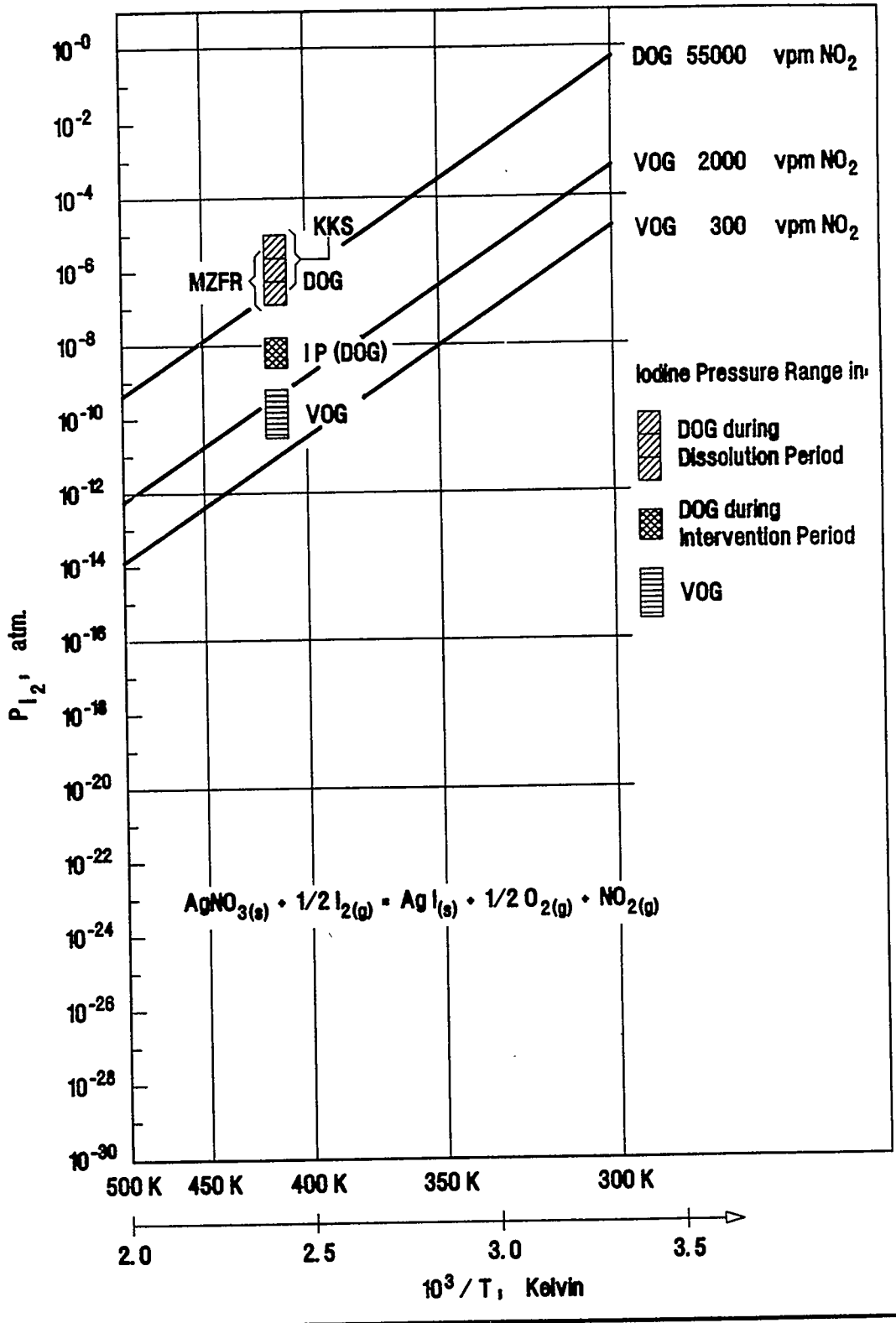
WAK

Iodine Sampling by Cooling Trap

WAK : ACC/90			
Fig. : 3			
drawn	9.7.90	RKr.	Drawing Nr. :
chckd.	9.7.90	VM.	VM9004



WAK	Iodine Equilibrium Pressures Versus Temperature	WAK: ACC/90 Fig.: 4 drawn 8.7.90 RKr. Drawing Nr. checkd. 8.7.90 VM. VM9007



WAK	Iodine Equilibrium Pressures Versus Temperature	WAK: ACC/90		
		Fig.: 5		
		drawn 8.7.90	RKr.	Drawing Nr. VM9006
		chckd. 8.7.90	VM.	

DISCUSSION

JUBIN: What steps are you taking to enhance the evolution of iodine from the dissolver solutions?

MOTOI: We try to expel iodine during dissolution and we have seen that we are not able to do better than 95%, our maximum. We thought we could remove more but it was impossible and, therefore, we have an iodine buildup in the off-gas cleaning system.

JUBIN: I wonder if you used sparging with nitrogen oxides.

MOTOI: We have made tests with nitrogen dioxide additions but we found a need to sparge the solution for a couple of hours at 90° and that we couldn't do.

VIKIS: Where is the dodecyliodide coming from?

MOTOI: The dodecyliodide is coming from our solvent.

IODINE-129 DISTRIBUTION AND RETENTION DURING EVAPORATION OF MLW SOLUTIONS

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Abstract

At the Karlsruhe Reprocessing Plant WAK 15 - 20 metric tons of LWR fuel are processed annually. The amount of aqueous MLW arising is 200 - 400 m^3 which contain 0.5 to 1% of the iodine-129 inventory of the spent fuel reprocessed in the WAK plant.

In 1989 11 MBq iodine-129 were contained in 230 m^3 MLW. During MLW evaporation 36% of iodine-129 reached the distillate, 50% the concentrate, and 14% the offgas.

The silver nitrate impregnated sorption material AC 6120 has proved its worth in retaining iodine from the MLW evaporator offgas. Decontamination factors (DF) between 30 and 40 were measured over 12 months of operation.

I. Introduction and Objectives

The Central Decontamination Plants Department (HDB) of the Karlsruhe Nuclear Research Center (KfK) is responsible for the disposal of wastes arising at the Nuclear Research Center, at the Karlsruhe Reprocessing Plant WAK, and in the state of Baden-Württemberg. Medium-level and low-level, liquid and solid wastes are being processed.

The iodine-129 which gets into the Karlsruhe Reprocessing Plant in the course of dissolution of spent fuel is distributed as follows (1):

- >95% of the iodine-129 inventory introduced together with the fuel is expelled from the dissolver and fixed on iodine filters provided in the dissolver offgas line.
- Approx. 1.5% of the iodine-129 inventory introduced together with the fuel reaches the vessel offgas lines where iodine filtration has been or will be provided.
- 1.7 to 3% of the iodine-129 inventory introduced together with the fuel gets into the medium-level liquid waste solutions which are disposed of by KfK-HDB.

The annual iodine-129 activity releases with the vent air of the facilities from reprocessing operations and from the treatment of aqueous low and medium level wastes are shown in Fig. 1 for the years from 1977 until 1989

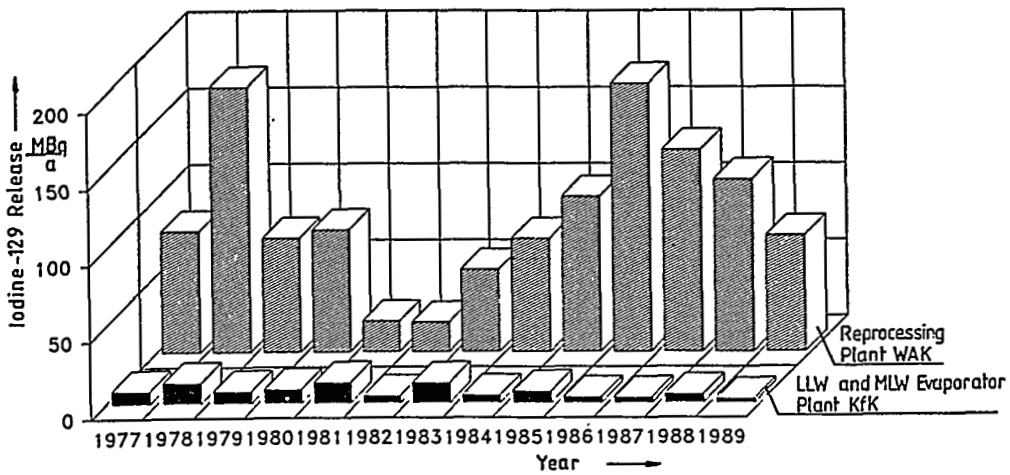

Fig. 1 Annual release of iodine-129

It appears that only a relatively small amount of iodine-129 emission originates in waste treatment.

During the years from 1986 until 1989 WAK and KfK-HDB made investigations into the iodine flowsheet associated with the treatment of liquid wastes. The work was necessary in order to be able to prepare an iodine-129 flowsheet for the German reprocessing plant to be built at Wackersdorf.

Moreover, as the weekly limit of iodine-129 emission was reduced in 1987, the offgas generated during evaporation of aqueous medium-level waste - a step in the treatment of this waste category - had to be filtered for the purpose of iodine removal.

The two requirements above implied the following goals to be pursued:

- determination of iodine-129 distribution in the MLW treatment process;
- followup of iodine-129 emission from the MLW evaporator plant;
- testing a sorption material for iodine filtration in the offgas of the MLW evaporator plant.

Depending on the process operating conditions selected for reprocessing and on the burnup of the fuel, 1.7 to 3% of iodine introduced in the reprocessing plant reaches the waste treatment facilities as follows: 0.2 to 0.4% in LLW, 1.0 to 1.6% in organic MLLW, and 0.5 to 1.0% in aqueous MLW.

The Central Decontamination Plants Department owns two

evaporat ion plants for liquid waste treatment where LLW and MLW solutions are evaporated. The annual throughput is 5000 m^3 in the LLW evaporator system and about 400 m^3 in the MLW evaporator system. During LLW evaporation the volume is reduced by a factor of 40 and in case of MLW evaporation by a factor of 10. The concentrates obtained are cemented in another HDB facility to give a waste form suitable for final repository storage. About 1500 m^3 of solid waste are produced annually which are transferred into the HDB intermediate storage facility until a final repository will be put into service. The steps in the treatment of the aqueous wastes at HDB have been represented schematically in Fig. 2.

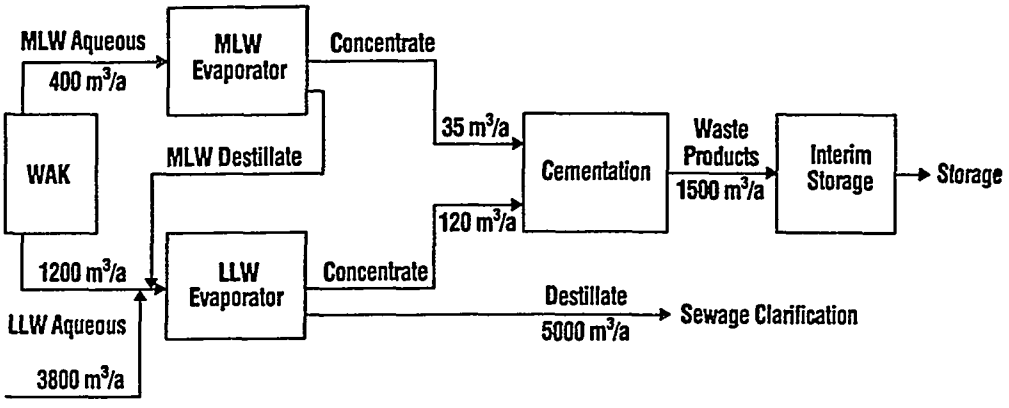

Fig.2 Treatment of aqueous LLW/MLW at KfK

This paper describes in detail the iodine-129 distribution and retention during evaporation of the aqueous medium-level waste.

II. Evaporation of the Aqueous Medium-level Waste

Most of the aqueous MLW originates in WAK, a pilot plant with an annual throughput of approx. 15 - 20 metric tons of burnt up LWR fuel. Every year 200 to 400 m^3 of aqueous MLW are generated at the WAK which are evaporated in the aqueous MLW evaporator plant.

1. Evaporation

The aqueous MLW solutions are transported from WAK to HDB in a tank truck as 2 m^3 batches of nitric acid solutions and supplied via a phase separation vessel (where organic phases present are separated) into the receiver of the MLW system. From that system the solutions are fed batchwise into the evaporator and through evaporation reduced in volume by factors of 6 to 13 (average 10), depending on the acidity,

activity and salt freight. MLW is evaporated in 5 to 10 campaigns per year of four to eight days duration each. During the evaporation campaigns the throughput of the evaporator is 1 m^3/h.

The layout of the MLW evaporator system of HDB (evaporation and offgas cleaning) has been represented schematically in Fig. 3.

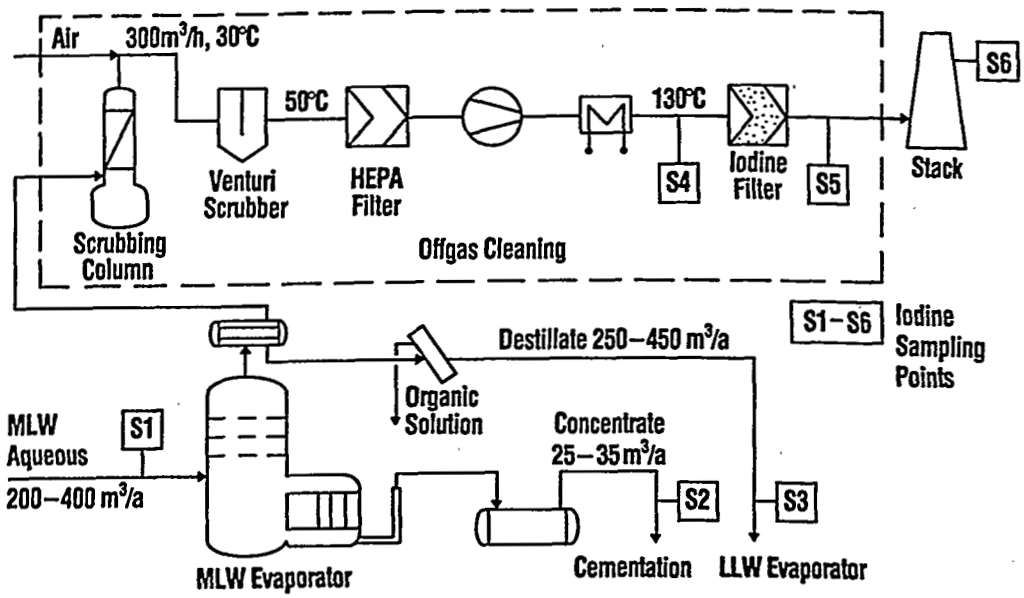

Fig. 3 MLW evaporation plant at KfK

The evaporator is operated by natural circulation and heated with steam (p = 2.8 bar). 1.8 m^3 concentrate at the maximum may accumulate in the evaporator sump.

The rising vapors are washed in a sieve tray column on top of the evaporator and cooled down to approx. 60°C. The condensate is passed through a phase separator where the co-distilled organic constituents are removed. The discharging aqueous distillate is further processed together with the aqueous LLW. The efficiency of that phase separation cannot be quantified because, for the time being, no samples can be collected from the aqueous and organic phases. The separated organic phase is directly carried over into the MLW concentrate.

After the limits of activity and/or the total cation concentration (3.7×10^{10} Bq/l and/or 5 mole/l) have been attained, the concentrate is extracted batchwise or in the continuous mode. It is then cemented in the HDB cementation facility. The evaporator offgas is treated in the series connected offgas cleaning system which is permanently operating.

During evaporation the iodine is distributed, dependent on its concentration, volatility and the operating conditions, among the concentrate, the distillate and the offgas section[2

2. Offgas Cleaning

The offgas section comprises the following major components:

- scrubbing column/Venturi scrubber for nitrogen oxide and droplet removal;
- HEPA filter for solid aerosol removal;
- iodine sorption filter with heater.

The offgas of the MLW evaporator is scrubbed in a scrubbing column and in a Venturi scrubber with a view to removing the nitrogen oxide and droplets, passed over HEPA filters for solid aerosol removal and, finally, over the iodine filter for iodine retention.

After the limit of iodine-129 emission had been reduced from 0.4 to 0.2 MBq/week [3] it was necessary to install an iodine sorption filter in the offgas section in 1987. It was operated exclusively during the evaporator campaign. After termination of a campaign the iodine filter remained operating for another eight hours. Between two evaporator campaigns the iodine filter was bypassed.

Initially, potassium-iodide impregnated activated carbon (SS 207 B) was used as the sorption material at approx. 40°C offgas temperature. The material did not produce satisfactory results which will be explained in Section III.

Therefore, it was decided in 1988 to test a silver nitrate impregnated catalyst carrier (AC 6120). That material has been used with good success in the WAK dissolver offgas since 1975. An electric heater was installed in order to raise the offgas temperature to 130°C and exclusively AC 6120 has been used as iodine absorber since.

The layout of the iodine filter is represented in Fig. 4.

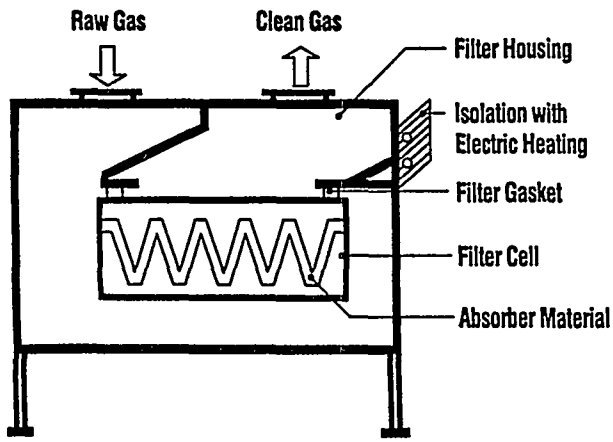

Fig. 4 Iodine filter in the MLW offgas

The KfK stainless steel standard filter version is used as the filter housing and filter cell for accommodation of the sorption material. The face area of the filter cell is 1.6 m^2, the bed depth is 50 mm. The bulk weight of a filter fill is 40 kg in case of activated carbon and 51.5 kg in case of AC 6120 (overall dimensions of the filter housing 800x700x700 mm^3).

The offgas flow in the cleanup section is kept constant at 300 std.m^3/h by dosing in controlling air. The iodine is removed under the following conditions.

- volume flow rate during operation: 440 m^3/h at 130°C
- face velocity (linear): 76 mm/s
- gas residence time: 0.7 s

The iodine-129 emission of the MLW evaporator system is controlled at the vent air measuring location S6 shown in Fig. 2.

Since 1988 iodine sampling locations have been provided upstream and downstream of the iodine filter (S4 and S5 in Fig. 2). These measuring points serve to determine the iodine-129 concentration in the offgas and to determine the decontamination factor of the iodine filter.

The devices for iodine sampling at the locations S4 and S 5 have been represented schematically in Fig. 5.

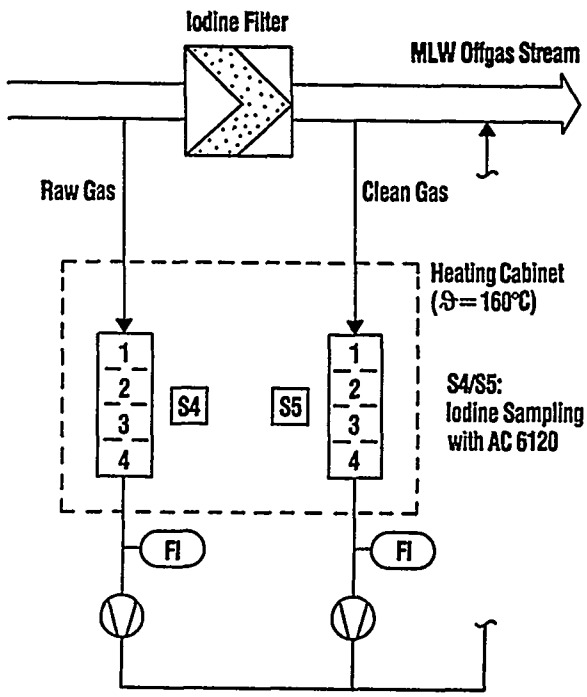

Fig. 5 Iodine sampling in the MLW offgas

Samples are taken simultaneously at a rate of approx. 550 std.l/h upstream and downstream of the iodine filter and carried to an iodine collector heated to 160°C. It is filled with the AC 6120 absorber material and consists of four beds equal in size, 50 mm bed depth, 32 mm in diameter. The gas residence time is 0.2 s per bed. The iodine collector is evaluated every two to four weeks.

III. Results

1. Iodine-129 Emissions during MLW Treatment

The iodine-129 emission during treatment of the aqueous MLW is determined at regular intervals at the vent air measuring point (S6 in Fig. 3).

The annual emissions in the years 1983 until 1989 are represented in Fig. 6. An iodine filter was installed in the offgas cleaning section of the plant in 1987 with a view to reducing the iodine-129 emission.

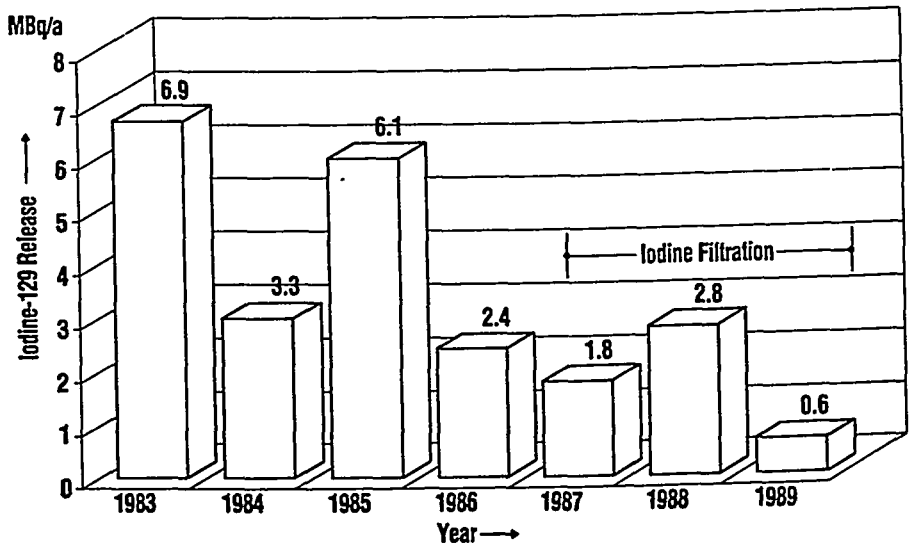

Fig. 6 Annual release of iodine-129 at the MLW evaporator plant

2. Iodine-129 Distribution in the MLW Evaporator Plant

Evaporation

In 1989 a total of 230 m^3 iodine-129 containing aqueous MLW solutions were processed in the course of seven evaporation campaigns.

The aqueous MLW solutions are evaporated in nitric medium. Under these conditions elemental iodine and iodine compounds, respectively, are partly expelled (2). Consequently, both the MLW concentrate and the MLW distillate as well as the plant offgas contain iodine-129.

Iodine-129 balancing of the liquid streams was performed at the sampling points shown in Fig. 3: S1 for the plant feed, S2 for the MLW concentrate, S3 for the MLW distillate. The results of balancing in 1989 are evident from the following Figure 7.

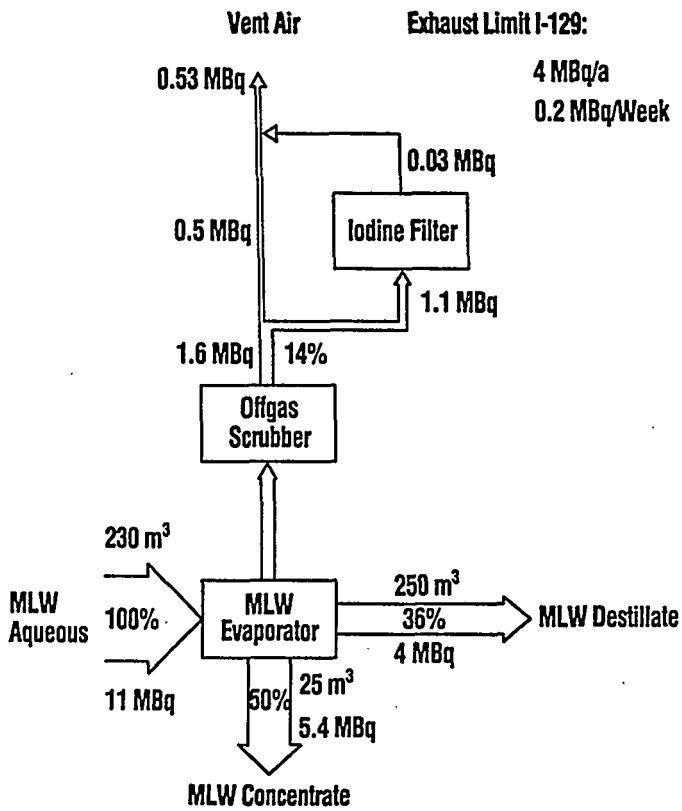

Fig. 7 Iodine-129 flowsheet of the MLW plant in 1989

Offgas Cleaning

The evaporation system is operated at intervals. The outage periods between two evaporator campaigns often last several weeks.

The offgas is passed over the iodine filter solely during evaporator operation. During outage periods the iodine filter is bypassed. In order to be able to study the retention of

iodine also during periods of outage of the evaporator plant the iodine filter was operated in 1989 both in the seven evaporator campaigns and, in addition, during four outage periods. This gives 11 intervals of operation of the iodine filter.

The investigations have shown that the specific iodine-129 concentration of the offgas is clearly higher during evaporator operation than during the outage period: The mean iodine-129 concentration during evaporator operation is 3 to 6 Bq/m^3, during outages 0.3 to 1 Bq/m^3.

3. Retention of Iodine-129 from the MLW Evaporator Offgas

KI impregnated activated carbon (SS 207 B) and $AgNO_3$ impregnated catalyst carrier (AC 6120) were used in the iodine filter of the MLW offgas section.

- KI Impregnated Activated Carbon

In case of iodine filtration using KI impregnated activated carbon iodine breakthrough took place at the end of fourteen days of operation. The short time-to-failure is attributable to humid and nitrogen oxide containing offgas and to the low operating temperature of the iodine filter of 40°C. Moreover, it has been demonstrated that there was no more KI impregnation present after that campaign. As the activated

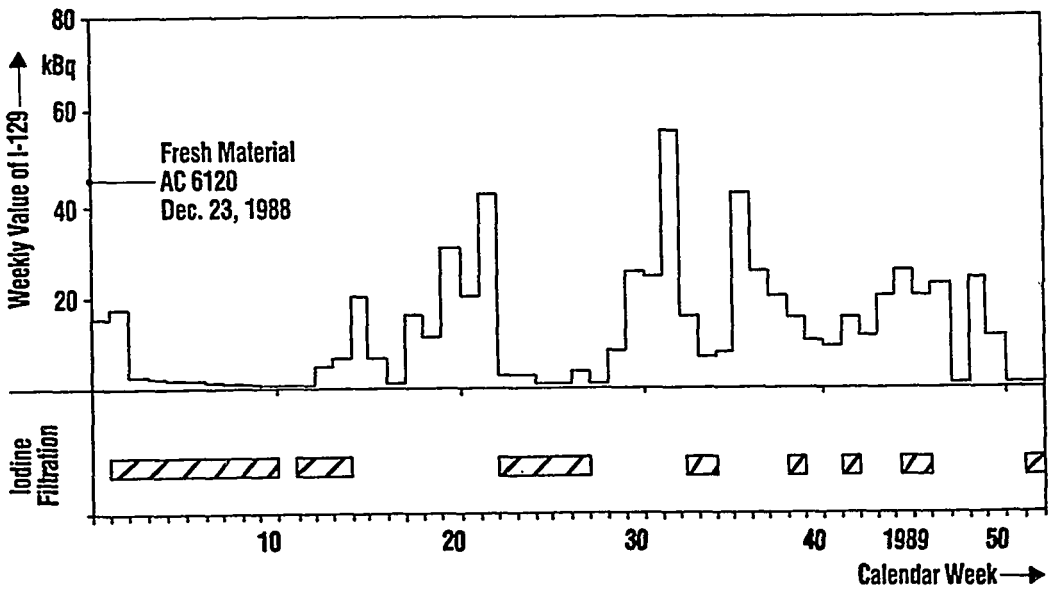

Fig. 8 Weekly iodine-129 release in the vent air

carbon filter could not been operated during a sufficiently long service period, it has not been possible to determine a decontamination factor with the measuring equipment available.

Activated carbon filters can only be considered as adsorption filters with a relatively long time until breakthrough.

. AC 6120

It gets evident from the diagram in Fig. 8 that the iodine filter material was replaced in late 1988. In 1989 the offgas from the MLW evaporator system was passed over the iodine filter during the evaporator campaigns and only in some instances also during the evaporator outage periods.

Immediately after changeout of the iodine filter material a DF of nearly 70 was determined. Even after 10 to 15 weeks decontamination factors $\geq$ 60 were measured (see Fig. 9).

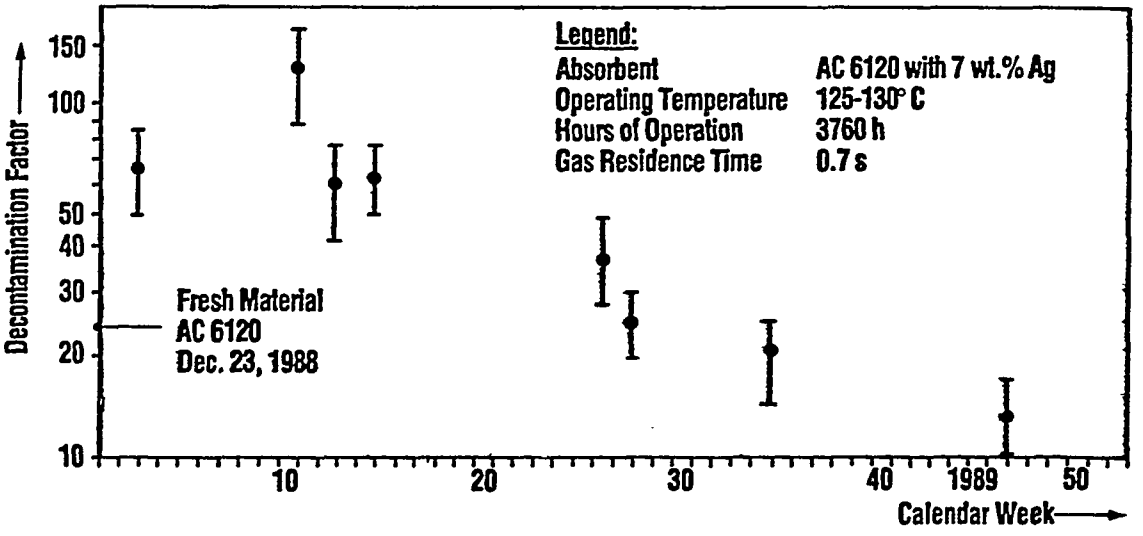

Fig. 9 Iodine-129 decontamination factor in MLW offgas in 1989

It is evident from Fig. 9 that by the middle of the year the iodine absorption capability had not decreased substantially. Only during the second half of the year a decrease in the decontamination factor can be noticed. This decrease cannot be explained by saturation of the material with iodine. It is more plausible that e.g. organic constituents of the offgas have deposited on the filter material.

In late 1989 the iodine filter material was again replaced. Samples were taken from that material in order to be able to characterize it under a subsequent program and to make studies on its regeneration.

The total iodine-129 activity entrained with the offgas was 1.6 MBq in 1989. Due to the intermittent operation of the iodine filter, only 1.1 MBq (approx. two thirds) of it were carried to the iodine filter. The decontamination factor calculated for the iodine filter ranged from 33 to 37 (see Table 1).

Table 1 Operating and Outage Periods of the MLW Evaporator, Iodine Entrainment and DFs of the Iodine Filter

No.	Operation	Duration t [h]	I-129 Activity A [MBq] Raw Gas	I-129 Activity A [MBq] Vent Air	DF
1.	MLW Evaporation with Iodine Filtration	1500	0.8	0.02	33
2.	Outage of Facility with Iodine Filtration	2260	0.3	0.01	37
3.	Outage of Facility without Filtration	5000	0.5	0.5	—
	Total	8760	1.6	0.53	3

The total iodine-129 emission of the MLW evaporator system (i.e.emissions of the offgas fractions with and without iodine filtration) was 0.53 MBq in the year of reference. In case of permanent operation of the iodine filter the expected iodine retention would have been > 97% which is equivalent to a reduction in iodine-129 emission of that facility to 0.05 MBq.

The investigations have shown that the AC 6120 iodine sorption material is suited for iodine filtration in the MLW evaporator offgas, both during evaporator operation and in outage periods.

Thanks to the temporary operation of the iodine filter in 1989 only 13% of the annual limit of emissions were attained.

However, with a view to minimizing the iodine-129 emission it is reasonable to pass the offgas over the iodine filter also during outage of the evaporator.

IV. Outlook

It is presently examined on a sample taken from the installed filter whether the initial retention capability can be regenerated by conditioning with nitrogen dioxide. This would extend the service life of an AC 6120 filter fill and, moreover, the costs of iodine filtration could be much lowered due to the savings in material and disposal costs.

Also, it is being investigated whether the decrease in DF is a result of poisoning of the iodine filter.

Literatur:

/1/ F.J. Herrmann et al
Retention and Measurement of Iodine-129 and Entrainment of Organoiodine with the Off Gas Streams of the Karlsruhe Reprocessing Plant WAK
Proc. 21st Air Cleaning Conference (1990)

/2/ E.Henrich et al
Ioddesorption aus der Brennstofflösung
KfK 4476 (1988)

/3/ W. Koelzer, M. Urban
Jahresbericht 1989 der Hauptabteilung Sicherheit
KfK 4730, S. 63 - 66

DISCUSSION

WILHELM: During the period the iodine filter is not used, the ventilation duct which carries the off-gas is contaminated. Is part of this ventilation duct still used when the iodine filter is needed so that contamination downstream of the filter may add to the contamination of the off-gas? Or is it completely divided so that no cross contamination can occur?

AMEND: There is a by-pass to the iodine filter and the line can shut off with valves. The sampling point to measure the iodine concentration at the iodine filter is in the part which is shut off.

WILHELM: So it shouldn't be contaminated when the filter is not used?

AMEND: Yes, it's correct.

HANDYSIDE: After the adsorbing material comes out of service it then goes into storage, have you looked at the release of iodine from stored AC-6210?

FURRER: We carried out such experiments on the AC 6120 iodine sorption material which had been exposed to I-129 from the dissolver off-gas of the Karlsruhe Reprocessing Plant. Sixty-eight kg AC 6120/12% Ag were filled into a drum together with 880 mCi I-129. After closure of the waste drum it was flushed with air for several weeks at 60°C in order to simulate the temperature in a repository and the exhaust air was passed through a multi-stage iodine filter. Then the sorption material was immobilized in Portland cement and the test was repeated. For the non-immobilized material an I-129 release rate of 5.4×10^{-10}/a was measured and for the material immobilized in Portland cement a value of 3.2×10^{-7}/a.

MULCEY: I would like you to comment on the regeneration of AC6120 with NO_2, especially from the point of view of the chemical in the solvent itself.

AMEND: Three investigations were made at Kernforschungszentrum Karlsruhe and the results showed that it is possible to regenerate AC6120 with nitrogen dioxide.

TECHNICAL-SCALE IODINE EXPULSION FROM THE DISSOLVER SOLUTION AND BALANCE STRIKING FOR LIQUID AND GASEOUS IODINE FRACTIONS

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Abstract

In 34 experiments (five of them performed with recycled acid simulated by addition of tributyl phosphate) carried out in the WAESCHE/PASSAT dissolver offgas simulation and treatment facility both iodine expulsion from the simulated dissolver solution and iodine distribution in the offgas section were studied while varying the addition of carrier iodine (iodide and iodate) with or without subsequent exposure to NO_2 gas. The items to be taken into account included iodine recycling together with the condensate from the condenser installed downstream of the dissolver, optimization of the desorption of iodine from the NO_x-scrubber, iodine absorption in the rinsing liquid during cleaning of the fiber pack demister as well as iodine transport in the offgas lines to the iodine filters.

I. Introduction

The forthcoming reduction of the admissible annual iodine-129 release via the exhaust air path in commercial reprocessing plants (e.g., Karlsruhe up to 6.5 mCi, Cap La Hague, France, 3 Ci, Rokkasho-mura, Japan, 270 mCi) requires the application of suitable measures to expel from the dissolver solution those amounts of iodine that can be justified from the technical point of view and be removed directly from the dissolver offgas for deposition on solid iodine sorption material so as to be amenable to disposal. It is the objective of these investigations to reduce the amount of iodine remaining in the dissolver solution to the extent justifiable in terms of conduct of the process by using substances not interfering with the process and by variation of the chemico-physical conditions. Another goal consists in the determination of iodine distribution in the liquid and gaseous phases of the WAESCHE/PASSAT dissolver offgas simulation and treatment facility. Should it not be possible to expel most of the iodine into the dissolver offgas, a 20-30 times larger offgas volume flow in total must be removed in the individual process stages by filtration after dissolution.

In the Karlsruhe reprocessing plant (WAK) numerous measurements of the residual iodine content of the dissolver solutions were carried out while dissolving spent fuel elements. According to F.J. Herrmann et al. /1/, most of the fission product iodine is expelled from the dissolver during fuel dissolution in the WAK. It is then collected as inorganic iodine on the iodine filters with the AC 6120 iodine

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sorption material /2, 3/ located in the dissolver offgas section. The remaining amount of fission product iodine (< 5%) is subjected to the PUREX process treatment and partially discharged together with the vessel offgas.

II. Investigations of Iodine Expulsion and Distribution in the WAESCHE/PASSAT Dissolver Offgas Simulation and Treatment Facility

The WAESCHE offgas simulation facility was erected in 1983. Trial operation was taken up at the end of that year. This facility consists of a shear dust simulator, a dissolver, a condenser and an NO_x-scrubber and is used to simulate and study the aerosol and iodine formation, behavior and distribution in the head-end under real conditions. After flowsheet studies had been carried out, the PASSAT dissolver offgas purification facility /3-7/ was modified and connected with the WAESCHE facility.

A simplified flowsheet of the WAESCHE/PASSAT dissolver offgas simulation and treatment facility is shown in Fig. 1. The sampling points for the gas and the liquid streams are marked "SG" and "SL", respectively. The previous PASSAT flowsheet /6/ included two heaters. The first heater was located upstream of the HEPA filter and heated the offgas up to 80 °C. The second heater was located upstream of the two iodine filters which were connected in series. This heater was used for further heating of the offgas up to 150 °C. On the basis of the experience gained, better NO_2- and temperature-resistant materials were selected for the adhesive between the filter medium and the frame as well as for the gasket of the HEPA filter. Thus, it was possible to use one heater only for the heating of the offgas up to 150 °C. This heater was to be located upstream of the HEPA filter and the two iodine filters.

The studies using this integrated offgas simulation and treatment facility were aimed at determining the aerosol source term during batch operation of a dissolver designed to accommodate one metric ton of cut spent fuel elements /8, 9/ . Furthermore, it was the objective to investigate iodine distribution and expulsion from the dissolver to the iodine filters in the following years.

Experimental

In 34 experiments (five of them performed with recycled acid simulated by addition of TBP), iodine expulsion from the simulated dissolver solution as well as iodine distribution in the offgas section were studied while varying the addition of carrier iodine (iodide and iodate) with and without subsequent exposure to NO_2 gas. The items to be taken into account included iodine recycling together with the condensate from the condenser installed downstream of the dissolver with the operating temperature adjusted to the optimum value of 60 °C, optimization of the desorption of iodine from the NO_x-scrubber, iodine absorption in the rinsing liquid during cleaning of the packed fiber mist eliminator (HEME) as well as iodine transport in the offgas lines to the iodine filters. Each experiment lasted 7 days, the duration of dissolution and iodine expulsion being fixed at 5 hours (flow rate 200 kg HM/h) and 4 hours, respectively. In the course of several experiments a new process step was introduced, the so-called tempering or treatment by boiling of the iodine containing

solution after addition of about 10% iodine as iodate in order to promote the exchange of isotopes.

In each experiment, 300 g I-127 as NaI (60 g/h) and 0.5 x 10^9 Bq I-131 as NaI, respectively, were fed into the dissolver solution. By means of the I-131 gamma radiation the liquid and gas samples (a total of 100 per experiment) were measured at calibrated measuring stations.

300 l 3-5 molar nitric acid were heated in the dissolver to about 100 °C. Within a period of 5 hours radioactively labeled sodium iodide was added from a storage tank in an α-shielded glovebox by means of an air jet pump. The stirring gas (air or N_2) fed into the dissolver served as carrier gas for the direct feeding of the iodide containing solution into the lower part of the dissolver. For the simulation of fluid mechanic processes, the dissolver is equipped with the cage containing the fuel element hulls in the dissolver pipe and two overflow pipes to the slab tank.

The nitric acid of the dissolver was stirred with 3 m^3/h air or nitrogen with the gas being fed into the lower part of the dissolver pipe. Leakage air was simulated by feeding an amount of 20 m^3/h into the vapor space of the dissolver above the acid. At about 100 °C and 960 mbar abs., evaporation rate of the solution amounted to 10% per hour. 2 m^3/h of NO_2 were supplied for iodine expulsion. The average operating temperature of the tubular heat exchanger installed downstream was adjusted to the optimum value of 60 °C. The condensate arising was measured and led back into the dissolver.

Table 1: Conditions for dissolution of spent elements

Amount of HM	**1 t/dissolution**
Duration	**5 h dissolution** **4 h iodine expulsion**
Dissolver solution	**nitric acid,** **up to 8 molar**
Evaporation rate	**10% of the content/h**
Stirring gas	**3 m^3/h**
Leakage air	**20 m^3/h**
Amount of iodine	**300 g/dissolution**

The NO_x-scrubber installed downstream was a bubble-plate column with a liquid volume of 400 l. Within this column, 300 l/h of diluted nitric acid cooled down to 20 °C were circulated and injected above the sixth and the tenth plate, respectively. For the oxidation of nitrogen monoxide generated during NO_2 absorption as well as for the homogenization of offgas flow through the column, 80 m^3/h air were led into the column via a control valve. The iodine containing offgas was led to the iodine filters connected in series via the separation units of wave plate demister, high efficiency mist eliminator (HEME) and high efficiency particulate air (HEPA) filter. For reasons of safety, cycle operation of the facility was carried out while introducing NO_2 and iodine.

A survey on the dissolution conditions of spent fuel elements is given in Tab. 1. The values indicated were used as a basis for the simulation experiments.

III. Residual Fractions of Iodine in the Dissolver Solution

Impact of the stirring gas fed into the dissolver on the residual fraction of iodine

During dissolution, homogenization of the dissolver solution is promoted by the addition of gas. Initially, those iodine compounds which were difficult to be expelled were assumed to be iodates. In addition to these iodates also elemental iodine is formed during oxidative dissolution of the iodides. Both the use of air and nitrogen as stirring gas was investigated for the displacement of the redox potentials.

As is evident from Fig. 2, the average value taken from several experiments does not indicate an increased iodate formation when using stirring air (1) instead of pure nitrogen (2) as stirring gas. Even subsequent treatment of the dissolver solution by addition of nitrogen dioxide (feeding time 1 h, 2 m^3/h NO_2) only led to minor reductions of the residual iodine contents (3).

Influence of the metered addition of iodide and iodate to the dissolver solution on the residual fraction of iodine

About 10% of the iodine originally present during dissolution of 1 t fuel were added to the dissolver solution at the bottom of the dissolver as dissolved inactive sodium iodide or sodium iodate at the end of dissolution and iodine expulsion. This addition of an excess of iodine ions was carried out with a view to facilitate desorption of the residual iodine fractions in the dissolver solution. After admixture of iodide or iodate, the solution was subjected to a 1 hour boiling process with subsequent exposure to NO_2 gas. As is obvious from Fig. 2, statistical evaluation of several experiments does not indicate that the residual iodine fraction in the dissolver solution is clearly influenced by dosage /4, 5/. While adding iodide, it was directly converted into elemental iodine by means of oxidation. Also the short residence time of the iodate in the dissolver solution and subsequent reduction to elemental iodine by NO_2 addition failed to bring about the expected reduction of the residual iodine constituents in the dissolver solution.

Influence of the iodate residence time on the amount of iodine remaining in the dissolver solution (treatment by boiling)

Obviously, minimization of the amount of iodine remaining in the dissolver solution was not achieved. Moreover, iodate formation in the dissolver solution was not found to be the decisive factor influencing the residual fraction of iodine. As a consequence, additional experiments were carried out in 1989 with the residence time of the iodates in the dissolver being prolonged considerably. After iodine expulsion iodate was added to the dissolver solution by means of boiling. Boiling of the solution and the iodate lasted several hours (iodine treatment by boiling). Then, NO_2 was fed into the dissolver for iodate reduction. The feeding time amounted to 1-2 hours. As a result of this iodine treatment by boiling, the amount of iodine remaining in the dissolver solution was reduced by 80-90%. The prolonged residence time of iodate is bound to give rise to a rapid exchange of isotopes in the boiling solution with an excess of inactive iodate. According to previous findings, most of the non-volatile residual iodine fractions in the dissolver solution are iodine compounds which undergo isotope exchange, but cannot be reduced by addition of nitrogen oxide. In the dissolver solution, isotopic exchange and iodine release take place in accordance with the following formulas (UC-I^{131} unidentified iodine compound):

$$UC\text{-}{}^{131}I + {}^{127}IO_3^- \longrightarrow {}^{131}IO_3^- + UC\text{-}{}^{127}I$$

$$3\ NO_2 + H_2O \longrightarrow NO + 2\ HNO_3$$

$$2\ {}^{131}IO_3^- + 6\ NO \longrightarrow 6\ NO_2^- + {}^{131}I_2$$

IV. Iodine Content in the Scrubbing Solution of the NO_x-Scrubber

The gaseous elemental iodine released from the dissolver solution is led via the condenser to the nitrogen oxide scrubbing column. In this column, the dissolver offgas is subjected to scrubbing during the dissolution time of 5 hours until a residual amount of nitrogen oxides of about 1 vol.% is reached. Parallel to nitrogen oxide retention iodine is dissolved in the scrubbing acid. Iodine distribution in the scrubbing column is found to be in equilibrium. This equilibrium is influenced profoundly by the content of organic traces in the offgas. The use of nitric acid in the dissolver and in the NO_x-scrubber does not result in major differences of the iodine contents of both solutions (Fig. 3, treatment methods 1-5). Metered addition of TBP (92 mg/l) for simulation of a recycled acid, however, is bound to cause iodine concentration of the scrubbing acid to increase dramatically (Fig. 3, treatment method 6). In Fig. 4, the experiments performed with and without TBP are compared statistically. As a result of the use of TBP containing acid the amount of iodine remaining in the NO_x-scrubber solution is increased fourfold. Due to the volatility of TBP at 100 °C, most of it is evaporated in the dissolver solution. In contrast to this, TBP is found to be enriched in the NO_x-scrubber. As a consequence, solubility of iodine in the scrubbing acid is increased. For concentration purposes, the acid of the NOx-scrubber is fed into an evaporator. This evaporator is connected to the dissolver offgas such that the iodine released during

evaporation enters the dissolver offgas. The concentrated acid is used again in the dissolver as recycled acid.

V. Iodine Behavior During Simulation of a Dissolution Process

By way of example, iodine-131 concentration behavior determined in an experiment without TBP is represented in Fig. 5. While adding NaI labeled with NaI-131 over a period of 5 hours, iodine concentration both in the dissolver and in the condensate is nearly constant, whereas iodine-131 concentration in the NO_x-scrubber is increased. After 4 hours of iodine expulsion, iodine concentration in the dissolver and scrubbing column reaches an equilibrium value. Subsequent exposure to gas causes iodine concentration to drop slightly. In the dissolver, however, a slight rise of iodine concentration can be observed. This rise can be explained by depot formation in the dead spaces of the dissolver. As the air flow is not uniform in all dissolver compartments, iodine deposition takes place. In the course of the following 17 hours the deposited iodine is swept back into the solution.

Iodine concentration in recycled acid simulated by TBP addition with iodine treatment by boiling and by admixture of IO_3^- and subsequent exposure to NO_2 gas for a period of 2 hours is represented in Fig. 6. After iodine expulsion, a slight increase in the iodine concentration can be noticed until NO_2 is added. After the addition of NO_2 the amount of iodine remaining in the solution is found to be reduced significantly.

VI. Measurement and Distribution of Gaseous Iodine Fractions in the WAESCHE/PASSAT Facility

Iodine behavior at the high efficiency mist eliminator (HEME) with variable iodine concentrations of the offgas

During normal operation of the PASSAT filter system, the amounts of condensate discharged in the wave plate demister for coarse droplet separation and in the downstream HEME fine droplet separator, respectively, are not worth mentioning. Only cleaning of the HEME unit the filter bed of which consists of a densely packed glass fiber revealed iodine fractions in the drained and collected scrubbing solution which varied as a function of dissolver operation (iodine dosage and iodine expulsion). While cleaning the HEME unit with 60 l of liquid over a period of 15 minutes during main dissolution simulated by metered addition of sodium iodide (60 g iodine/h), 1.5 g of radioactively labeled iodine (0.5% in terms of the total amount of iodine used) were entrained together with the rinsing liquid. The samples were taken at the sampling points SL 5 and SL 6 indicated in Fig. 1. During 4 hours' iodine expulsion following main dissolution, an additional amount of iodine of 0.05% was entrained while rinsing with 60 l of liquid.

Distribution of gaseous iodine

A self-contained iodine balance of the entire facility requires the determination of the iodine fractions both in the solutions and of the gasborne iodine. At the SG sampling points indicated in Fig. 1, partial flow samples were taken and iodine was collected on

the AC 6120 iodine sorption material at 150 °C. However, sampling turned out to be difficult at the sampling points SG 1 between the dissolver and the condenser and SG 2 between the condenser and the scrubbing column, respectively. At SG 1, the gas samples were taken from a moisture saturated gas flow at temperatures of about 95-100 °C. This gas flow consisted of 2 m^3/h of stirring air, 20 m^3/h of leakage air and 54 m^3/h of steam. Condensation at the probe located in the main offgas stream caused the iodine to be washed out of the sampling flow.

Compared to the amount of iodine used, iodine concentration measured at SG 1 was too low by about 40% in all experiments (Fig. 7). As a result of adding 80 m^3/h of control air with room temperature, dew point is underrun and iodine is washed out (dew point of the condenser 60 °C). Consequently, smaller iodine concentration values are measured at SG 2 (Fig. 7).

Satisfactory concentration values were measured at the sampling points SG 3 (dew point of the offgas after having passed the NO_x-scrubber: 25 °C), SG 4, SG 5, and SG 6 (upstream of, between and downstream of the iodine filters heated to 150 °C), respectively. In Fig. 7, the average iodine concentrations measured at 4 different sampling points 26 hours after iodine dosage into the dissolver are shown in relation to the amount of radioactively labeled iodine fed into the dissolver. The mean values indicated were calculated on the basis of 30 experiments performed.

In all experiments, the iodine fractions measured at the sampling points SG 3 and SG 4, respectively, under the same operating conditions differed considerably. 26 hours after the introduction of iodine into the dissolver the amounts of iodine measured upstream of the iodine filters at SG 4 were found to be smaller by 5-8% in all experiments. After 5 days' additional operation of the offgas blower, additional iodine fractions ranging from 2 to 8% were measured at the sampling points SG 2, SG 3, and SG 4, respectively. The following iodine flow results from iodine desorption in the lines and components. During the test period of about 150 hours, nearly the total amount of iodine is desorbed and chemisorbed as silver iodide on the iodine filters. Thus, the iodine balance of the WAESCHE/PASSAT facility is calculated to be 100% in terms of the radioactively labeled iodine used, on the basis of the analysis of the gaseous and liquid iodine fractions.

VII. Summary and Conclusion

- Depending on the treatment method, the amount of iodine remaining in the simulated dissolver solution ranged from 0.1 to 0.35%.
- When using air or nitrogen as stirring gas, the residual iodine content was not affected noticeably.
- During optimum operation of the NO_x-scrubber, the average amount of iodine remaining in the scrubbing liquid was found to be 0.15% in standard experiments. In experiments performed with tributyl phosphate in the dissolver this value rose to 0.55%.

- While rinsing the high efficiency mist eliminator (HEME) during dissolution with 60 l of water in 15 min, about 0.5% of the iodine was entrained together with the rinsing liquid. After 1 hour of iodine expulsion the iodine content measured in the rinsing liquid amounted to 0.05%.

- Over a period of 17 hours after iodine expulsion, an additional amount of iodine of 5-8% was entrained together with the offgas to the iodine filters.

- In several experiments, iodine balance was determined to be 100 ±1 %.

- The addition of iodide as I-127 to the residual iodine and subsequent feeding of NO_2 did not lead to a decrease in the residual iodine fraction. The absence of isotope exchange was caused by the immediate oxidation of iodide to elemental iodine in the presence of nitric acid.

- In case no excess of inactive iodine as iodate was added and NO_2 was fed into the boiling solution, only slight reduction of the iodine fraction could be observed. This means that the residual iodine fraction mainly consisted of unidentified iodine compounds.

- Iodine treatment by boiling with an excess of about 10% of iodine as iodate seemed to be successful in initiating the exchange of isotopes. The decisive factor was the exposure time of potassium iodate with subsequent feeding of NO_2.

It is evident from the above results that the amount of iodine remaining in the simulated dissolver solution can be reduced to 0.1%. To remain well below this level, the parameters of iodine dosage, exposure time and solution temperature have to be optimized for minimization of the residual iodine fraction. Operation of the new IATEMA prototype facility has been taken up with a view to investigate these parameters by variation of their values. Minimization of the residual iodine fraction is to be studied both for batchtype and continuous dissolutions.

VIII. References

/1/ Herrmann, F.J., Motoi, V., Schoor van, A., Kraut, W., Füger, B., Stieglitz, L.,: "Austrag von Organo-Iod-129 mit dem Abgas einer Wiederaufarbeitungsanlage", Proceedings of the Annual Meeting on Nuclear Technology '90, Nürnberg, F.R.G., p. 309, ISSN 0720-9207 (1990).

/2/ Wilhelm, J.G., Schüttelkopf, H.: "An Inorganic Adsorber Material for Offgas Cleaning in Fuel Reprocessing Plants", CONF-720823, Vol. 2, p. 540 (1972).

/3/ Furrer, J., Wilhelm, J.G., Jannakos, K.: "Aerosol and Iodine Removal System for the Dissolver Offgas in a Large Fuel Reprocessing Plant", CONF-780819, Vol. 1, p. 494 (1978).

/4/ Furrer, J., Wilhelm, J.G.: "Iodine Trapping from the Exhaust Air of Reprocessing Plants", Seminar on Iodine Filter Testing, Commission of the European Communities, Directorate Health Protection, Luxembourg (Grand-Duchy), Doc V/559/74 (March 1974), p. 185.

/5/ Wilhelm, J.G., Furrer, J., Schultes, E.: "Head-End Iodine Removal from a Reprocessing Plant with a Solid Sorbent", CONF-760 822 (1977), Vol. 1, p. 447.

/6/ Furrer,J. et al.: "Results of Cleaning Dissolver Off-Gas in the PASSAT Prototype Dissolver Off-Gas Filter System", CONF-801 038 (1981), p. 566.

/7/ Jannakos, K. et al.: "Selected Solutions and Design Features from the Design of Remotely Handled Filters and the Technology of Remote Filter Handling", CONF-801 038 (1981), Vol. 1, p. 317.

/8/ Furrer, J. et al.: "Aerosol Source Term in the Head-End-Dissolver Offgas of a Reprocessing Plant", CONF-860 820 (1987), Vol. 2, p. 688.

/9/ Furrer, J., Weinlaender, W.: "Concepts and Operating Results in Minimizing Radioactive Emissions for the Wackersdorf Reprocessing Plant", CONF-880 822 (1988), Vol. 1, p. 100.

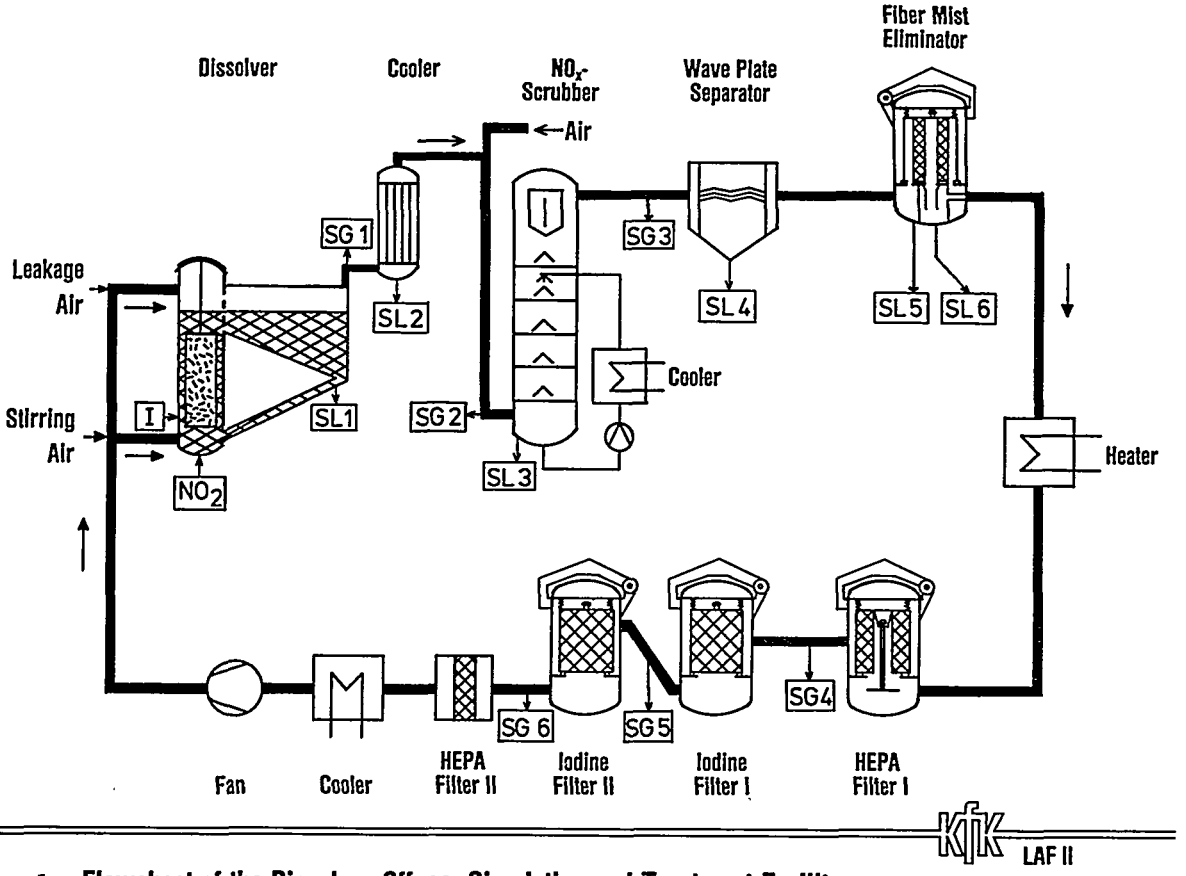

Fig. 1 Flowsheet of the Dissolver Offgas Simulation and Treatment Facility (WAESCHE-PASSAT)

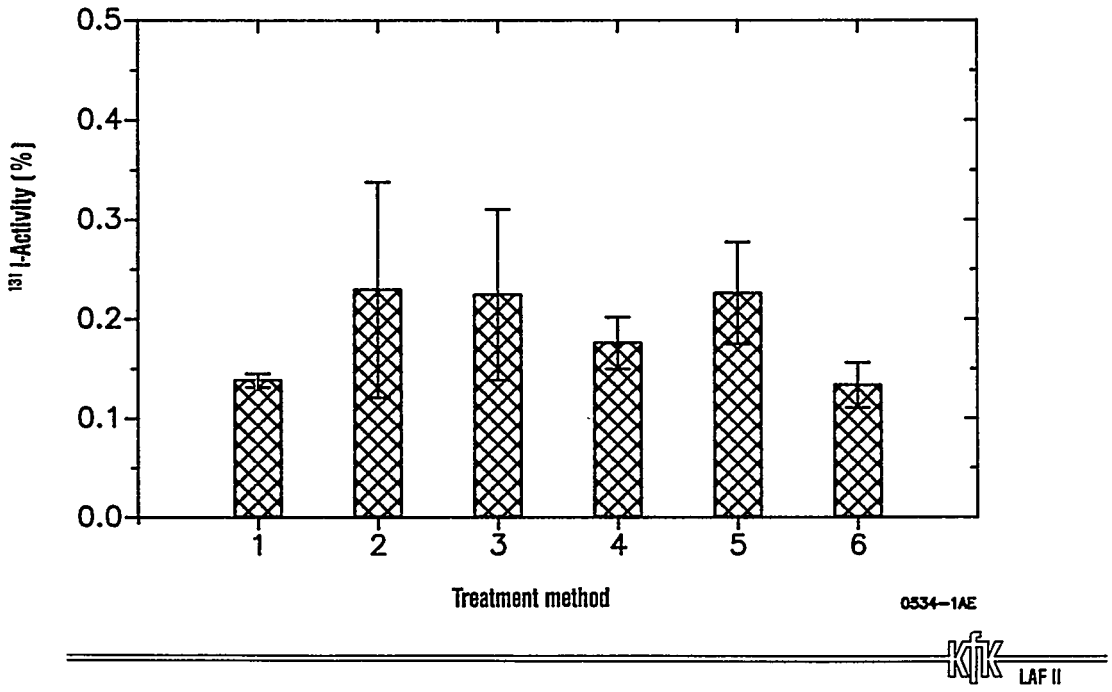

Fig. 2 Average ^{131}I activity and standard error in the dissolver (SL1) after 26 h with different treatment methods:
1: standard, 2: N_2 (as stirring gas), 3: NO_2, 4: NO_2 + NaI, 5: NO_2 + KIO_3, 6: NO_2 + KIO_3 + TBP (test 5-34)

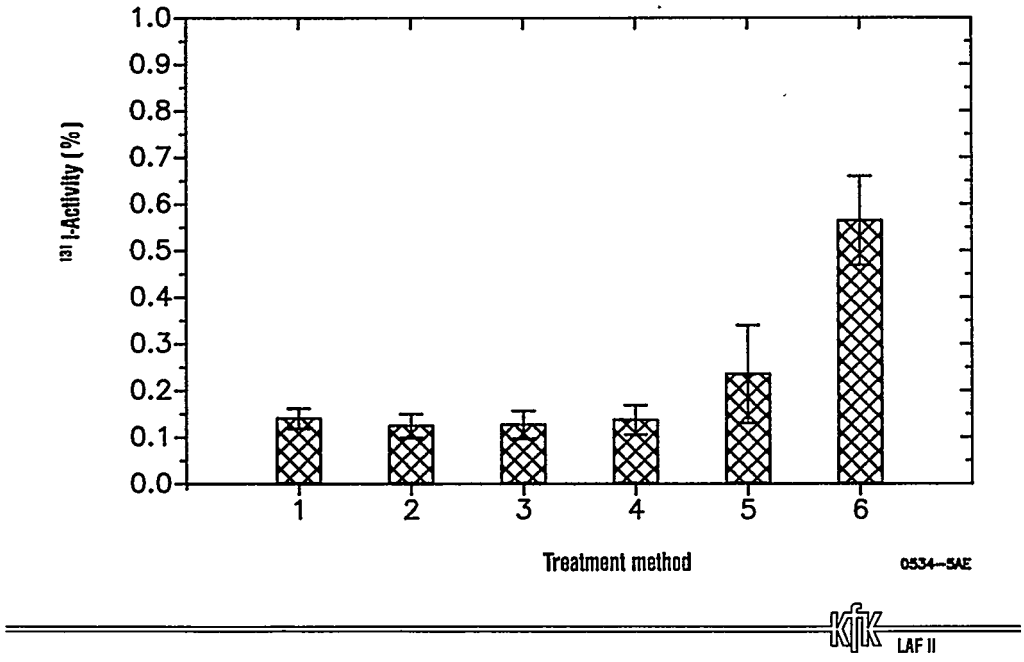

Fig. 3 Average ^{131}I activity and standard error of the ^{131}I activity in the NO_x-scrubber (SL3) after 26 h with different treatment methods:
1: standard, 2: N_2 (as stirring gas), 3: NO_2, 4: NO_2 + NaI, 5: NO_2 + KIO_3, 6: NO_2 + KIO_3 + TBP (test 5-34)

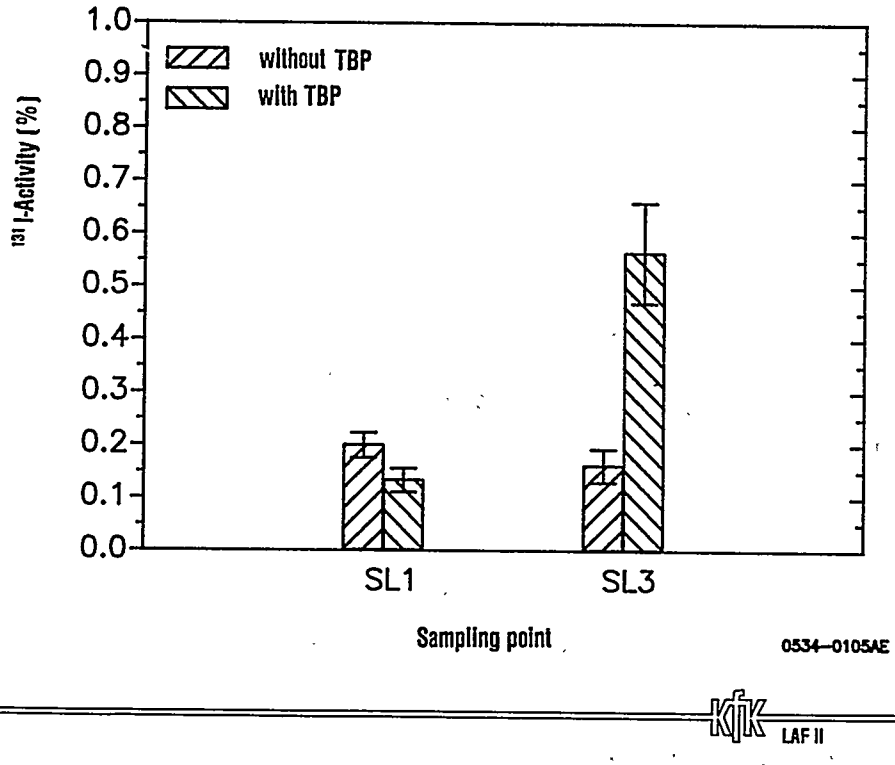

Fig. 4 Average ^{131}I activity and standard error after 26 h without and with TBP at different sampling points: tests 5-34

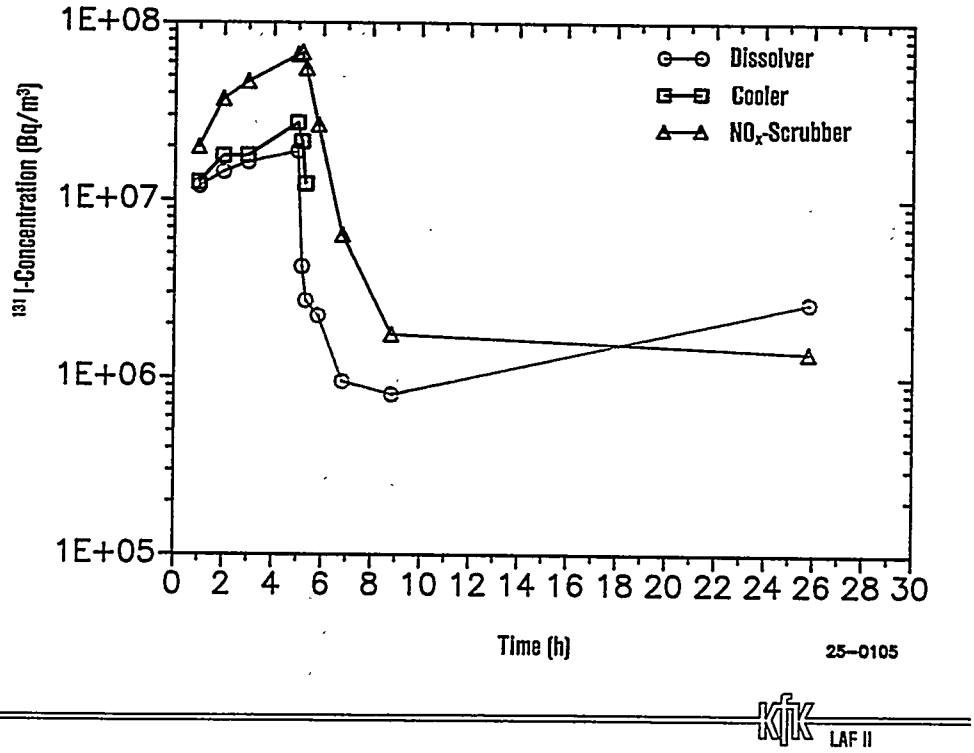

Fig. 5 ^{131}I-Concentration as a function of time in dissolver, cooler, and NO_x-scrubber: test 25

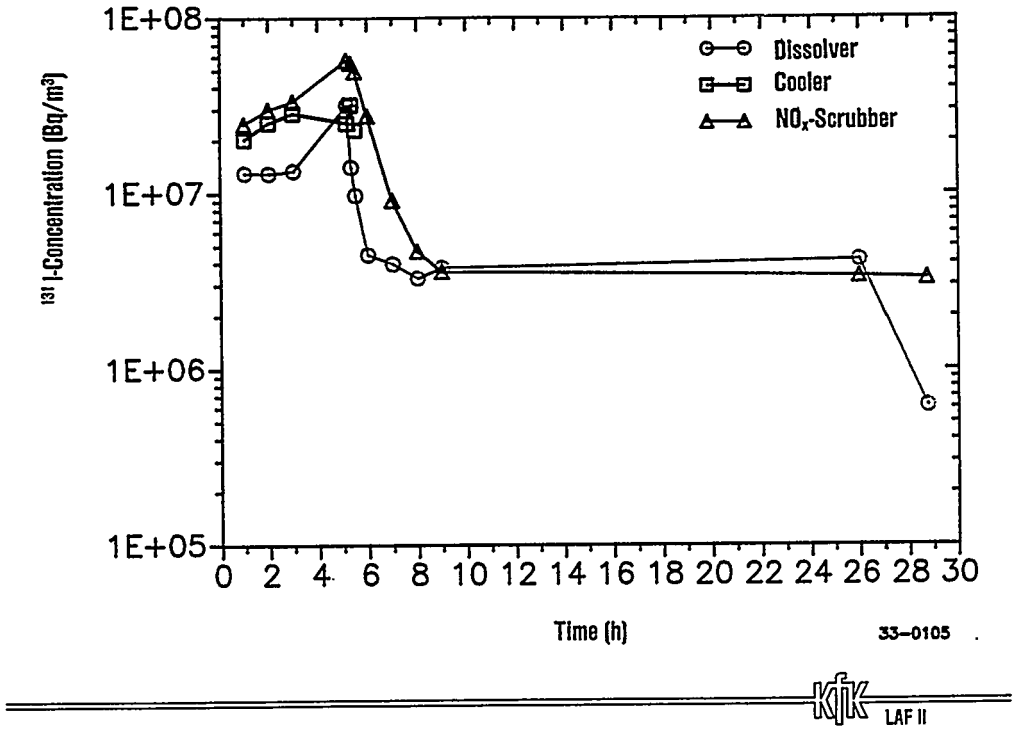

Fig. 6 ^{131}I-Concentration as a function of time in dissolver, cooler, and NO_x-scrubber: test 33

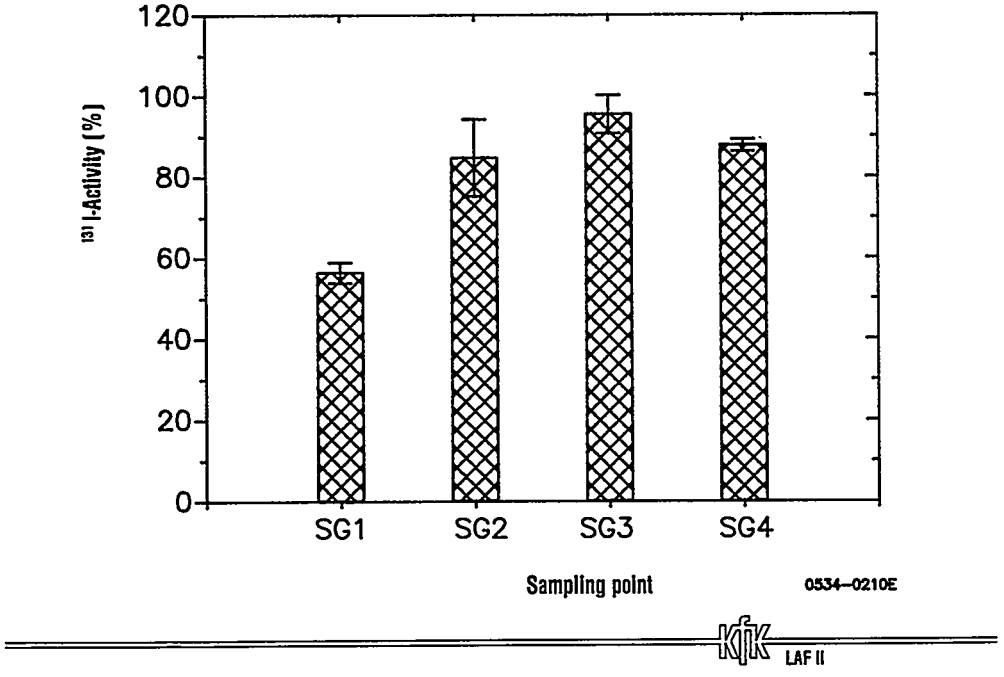

Fig. 7 Average ^{131}I activity and standard error after 26 h at different sampling points: tests 5 - 34

ELIMINATION OF IODINE FROM RADIOACTIVE GASEOUS EFFLUENTS IN A PACKED COLUMN

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Abstract

Iodine is eliminated from the gaseous radioactive effluent of a reprocessing plant in a packed column. Iodine is absorbed in a 0.5 N sodium-hydroxyde solution. The Kinetics of I_2-air-NaOH are known : it is an instantaneous interface reaction where the important parameters are the interface surface area and the gas film mass transfer coefficient k_G.

The hydrodynamic characteristics of different kinds of packing (Raschig rings, Berl saddles, Multiknit) were determined with a column of 0,1 m in diameter and 2 m high. The pressure drop, dynamic holdup, effective interface surface area, axial dispersion coefficient were determined.

The mass transfer coefficient k_G for the I_2-air-NaOH system was investigated in the column filled with glass Raschig rings. About 10 experiments were conducted because it is difficult to make experiments with this system. So we have used another well known system (CO_2-air-NaOH~1.8 N) to determine k_G. The $k_G(I_2)$ and $k_G(CO_2)$ values are shown in figures 8 and 9. They can be correlated by :

$$k_G(I_2) = 0.22\ G^{0.80} \quad \text{and} \quad k_G(CO_2) = 0.34\ G^{0.80}$$

The difference between the constant values can be explained by the ratio of diffusivity coefficients in gas phase ($D(I_2)/D(CO_2) = 0.5$ --> $k_G(I_2)/k_G(CO_2) = 0.7$)

I - Introduction

This study investigated hydrodynamics and mass transfer in a packed column with I_2-air-NaOH system (1).

II - Material and methods

The experimental plant is shown schematically in Figure 1, and the characteristics of different kinds of packings in Table 1. The inside diameter of the column is 100 mm and the packing height is 2 m.

The iodine, carbon dioxyde and carbonate concentration were determined respectively by a potentiometric method with a specific electrode, by chromatography and by pH measurements.

The effective interface surface area was mesuared with a pseudo first-order reaction (2). The flux of the compound A is :

$$N_A = C_{Ai} \, (k_2 \, D_{AL} \, C_{BL})^{0.5}$$

With the CO_2-air-NaOH system the CO_2 flux is independent of the resistance in the liquid film and $k_2.C_{BL}$ = Cte, when the concentrations are :

$$1 \times 10^{-5} < CO_2 < 4 \times 10^{-2} \text{ atm} \; ; \; 0,2 < NaOH < 0.4 \text{ N}$$

The material balance between the top and bottom of the column was used to obtain the effective interface surface area a_E which can be compared with the geometrical interface area a_C.

$$a_E = \frac{H_e \, G_M \, Ln \, (p_E/p_S)}{L P_T \, (D_{AL} \, k_2 \, C_{BL})^{0.5}}$$

The axial dispersion coefficient was measured in each phase. Each phase was marked with radioactive tracer (gas phase : ^{79}kr, liquid phase : ^{82}Br) by a pulse injection. The residence time distribution diagrams were recorded at the top and bottom of the packing. The results were analysed by convolution method :

$$C_2 \, (t) = \int_0^{\infty} C_1 \, (u) \; h \, (t - u) \; du$$

where :

$$h(t) = L \left(\frac{1}{E_i \, \pi \, t^3}\right)^{0.5} \exp \left[- \frac{(L - U_{Ii} \, t)^2}{E^2{}_i \; 4t} \right]$$

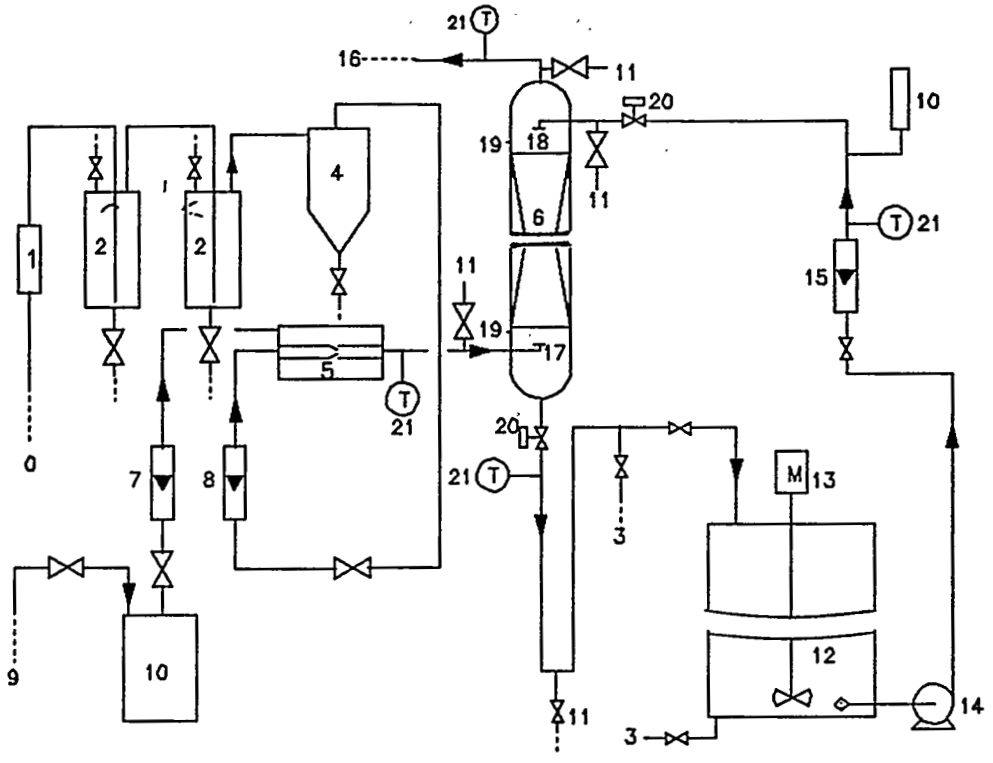
The curve was fit by the least-squares method. From these curves the axial dispersion coefficients E_L and E_G can be determined, as well as the Peclet numbers of each phase (1).

The pressure drop was measured by differential measurement of the dynamic pressures between top to bottom of the packing, and the dynamic holdup by collection of the liquid below the packing after cutting off simultaneous flowrates.

III - Theoretical aspect

Axial dispersion model

The axial dispersion model is applied to the absorption packed column and a material balance on a cross section of the column leads to differential equations for each phase (figure 2).



0. Compressed air
1. Actived charcoal filter
2. Air saturated with water
3. Liquid outlet
4. Cyclone
5. Gas mixing
6. Column
7. CO_2 flow meter
8. Air flow meter
9. CO_2
10. Tank
11. Sampling
12. Liquid tank
13. Mixing system
14. Pump
15. Liquid flowmeter
16. Air extraction
17. Gas inlet
18. Liquid inlet
19. Pressure tap
20. Pneumatic valve
21 Température probe

Fig. 1 : Pilot flow-sheet

Packing	nominal dimension $m*10^3$	external diameter $m*10^3$	height $m*10^3$	thickness $m*10^3$	contact surface a_c $m^{-2}.m^{-3}$	free space $m^{-3}.m^{-3}$	rings number $m^{-1}*10^{-3}$	critical surface tension $N.m^{-1}$
Glass Raschig rings	10	10	10	1	470	0,78	760	0,073
Ceramic Raschig rings	10	10	10	1,75	490	0,69	680	0,061
P.V.C. Raschig rings	10	10	10	1	492	0,78	780	0,040
Stell Raschig rings	13	13	12.8	1.7	342	0,75	330	0,071
Ceramic Berl Saddles (3)	10	--	--	--	670	0,66	690	0,061
Wire mesh packing "Multiknit"	100	--	--	--	1950	0,94	--	0,071

Table 1 : Geometrical characteristics of used packings

After solving these equations (1), we can determine the influence of the axial dispersion over the height of the column versus the column efficiency .

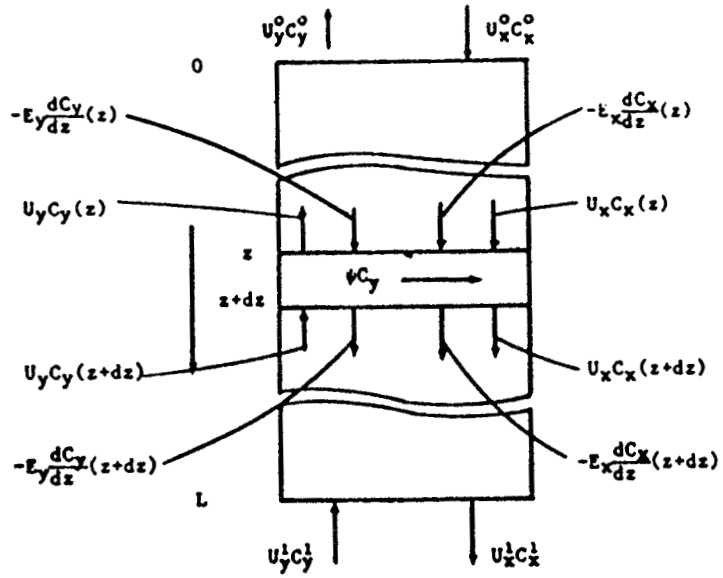


Fig. 2 :
Mass balance on a cross section of the column.

$$E_x \frac{d^2C_x}{dz^2} - U_{Ix} \frac{dC_x}{dz} + K_y a \, (C_y - {}^{e}C_y) = 0$$

$$E_y \frac{d^2C_y}{dz^2} + U_{Iy} \frac{dC_y}{dz} - K_y a \, (C_y - C_y^{e}) = 0$$

<u>I_2-air-NaOH system</u> (1,6)

Iodine adsorption in most materials raises problems for experimentation. The mass transfert of I_2-air-NaOH was investigated in a column packed with glass Raschig rings because iodine is not absorbed on glass.

Iodine is absorbed in a 0.5 N sodium hydroxyde solution. The kinetics of I_2-air-NaOH are known : it is an instantaneous interface reaction. The réaction flux equation is :

$$N_A = k_G \, Y_A$$

A material balance in column element gives :

$$G \, dY = N_A \, a_E \, dh$$

When Y << 1 the equation becomes :

$$k_G = \frac{G}{a_E \, L} \cdot \ln \left(\frac{Y_E}{Y_S} \right)$$

By measuring the concentrations at the top and bottom of the column, it is easy to determine partial mass transfer coefficient k_G in the gas phase.

But it is difficult to perform many experiments with gaseous iodine. We therefore simulated the column operation by another system easier to use. We chose the CO_2-air-NaOH system because it is very well know.

CO_2-air-NaOH system

At hydroxyde concentrations between 1.8 and 2 N it is possible to determine the partial mass transfert coefficient in the gas phase. The physico-chemical constants are know (2,4,5).

The method is the same as for the iodine system. The flux equation is :

$$N_A = \frac{1}{\frac{1}{k_G} + \frac{He}{(k_2\ C_{BL}\ D_{AL})^{0.5}}}\ P_A$$

Integration of the material balance from the top to the bottom of the column gives :

$$\frac{1}{k_G} = \frac{P_T\ a_E\ L}{G\left[\left(\mathrm{Ln}\ \frac{p_A}{P_T - p_A}\right) - \left(\frac{P_T}{P_T - p_A}\right)\right]_{p_{AS}}^{p_{AE}}} - \frac{He}{(k_2\ C_{BL}\ D_{AL})^{0.5}}$$

The measured inlet and outlet concentrations give the partial mass transfert coefficient k_G.

IV - Experimental results

Macroscopic hydrodynamics

Figure 3 shows a typical curve of pressure drop ($\Delta P/Z$) versus gas flowrate G, at constant liquid flowrate. The performance of the Multiknit is more interesting than other kinds of packing. Its capacity is five to ten times greater than classical packing (Berl saddles, Raschig rings). A the same gas-liquid flowrate the pressure drop of the Multiknit packing is ten times lower than Berl saddles and Rashig rings.

A typical curve of dynamic holdup β_d versus gas flowrate is shown in figure 4. After the loading point, β_d increases until the flooding point is reached.

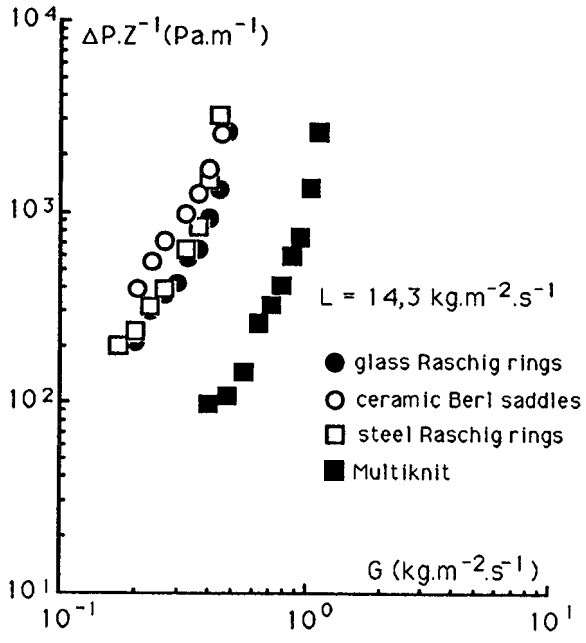


Fig. 3 :
Pressure drop versus gas flowrate.

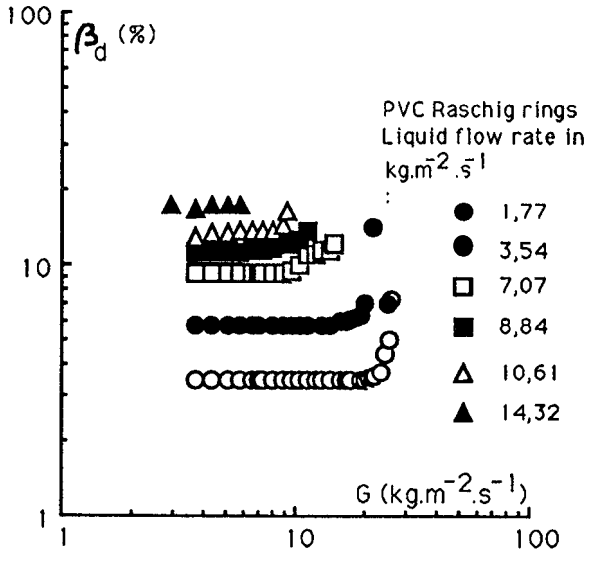


Fig. 4 :
Dynamic holdup versus gaz flowrate at constant liquid flowrate.

For a constant liquid flowrate, the interface area is constant below the loading point when the gas flowrate increases. Between the loading and flooding points the interface area increases as the gas flowrate increases, as dynamic holdup. A constant gas flowrate the interfacial area increases with liquid flowrate, but there is a limit with high liquid flowrate (figure 5a).

The multiknit packing has a different behavior (figure 5b). The interface area decreases when gas flowrate increases at constant liquid flowrate and also decreases when the liquid flowrate increases at constat gas flowrate. At high irrigation density there is a limit for interface area. At low flowrate, the capillary strengths are more important than the strenght of kinetics while the opposite is true at high flow rates.

Multiknit gives an interface area a_E two to five times greater than Berl saddles and Raschig rings.

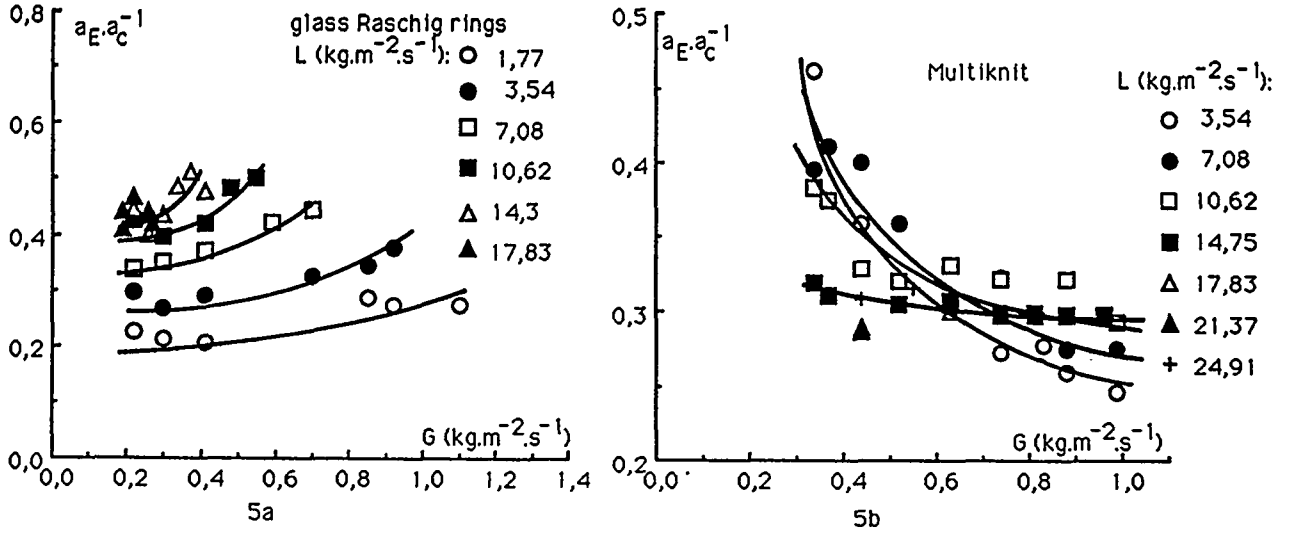


Fig. 5 : Ratio of measured interface area to geometrical interface area.

Microscopic behavior

Some data on axial dispersion E_L and Pe_L in the liquid phase are given in Tables 2a 2d for glass Raschig rings and Multiknit packing. For the same gas and liquid flowrates :

E_L (Raschig) < E_L (Multiknit)

Pe_L (Raschig) > Pe_L (Multiknit)

Tables 2b 2c, gives some axial dispersion values E_G and Pe_G in the gas phase. The same order of magnitude is obtained for both types of packings.

Raschig Rings.

U_G (m.s^{-1}) / U_L (m.s^{-1})*10^3	0		0.188		Δ 0.217		Δ 0.326		0.372		Δ 0.453		Δ 0.449		Δ 0.566		Δ 0.644		Δ 0.69		Δ 0.821		0.846	
	E_L	Pe_L	E_L	Pe_L	E_L	Pe_L	E_L	Pe_L	E_L	Pe_L	E_L	Pe_L	E_L	Pe_L	E_L	Pe_L	E_L	Pe_L	E_L	Pe_L	E_L	Pe_L	E_L	Pe_L
0																								
1.77			29.3	20																				
3.54			23.2	35															17.3	44	20.1	27		
7.08	21.1	54	22.9	48											13.6	72	6.8	103					12.8	52
10.62			23.4	53							11.7	89	14.1	84										
14.30			15.8	100			10.2	127																
17.84			14.7	110	12.5	112																		

Δ between loading and flooding points.

Table 2a

Raschig Rings

U_G (m.s^{-1}) / U_L (m.s^{-1})*10^3	0		0.188		Δ 0.217		Δ 0.326		0.372		Δ 0.453		Δ 0.449		Δ 0.566		Δ 0.644		Δ 0.69		Δ 0.821		0.846	
	E_G	Pe_G	E_G	Pe_G	E_G	Pe_G	E_G	Pe_G	E_G	Pe_G	E_G	Pe_G	E_G	Pe_G	E_G	Pe_G	E_G	Pe_G	E_G	Pe_G	E_G	Pe_G	E_G	Pe_G
0			68.6	58					85.4	94													538	31
1.77			89.7	56																	439	43		
3.54			105.3	48															405	39			585	36
7.08			92.3	58											466	31	598	28						
10.62			88.1	64							645	18	596	20										
14.30			151.4	40			163	63																
17.84			181.4	36	382	19																		

Δ between loading and flooding points.

Table 2b

Multiknit

U_G (m.s^{-1}) / U_L (m.s^{-1})*10^3	0		0.188		0.258		0.372		0.474		0.502		0.690	
	E_G	Pe_G	E_G	Pe_G	E_G	Pe_G	E_G	Pe_G	E_G	Pe_G	E_G	Pe_G	E_G	Pe_G
0							50	135						
7.08			43	93					163	61				
10.62			69	58							99	110	157	95
14.29			65	65			96	84			239	48		
17.84			68	62			220	52						
21.37			68	65			150	57	170	62				
24.91					78	73	182	48			347	38		

Table 2c

Multiknit

U_G (m.s^{-1}) / U_L (m.s^{-1})*10^3	0		0.188		0.258		0.372		0.474		0.502		0.690	
	E_L	Pe_L	E_L	Pe_L	E_L	Pe_L	E_L	Pe_L	E_L	Pe_L	E_L	Pe_L	E_L	Pe_L
0														
7.08			149	11					189	7			127	9
10.62			138	16							113	15		
14.29			162	13			110	17			41	30		
17.84			203	11			110	16						
21.37			189	13			85	22	99	19				
24.91					116	21	214	10			138	12		

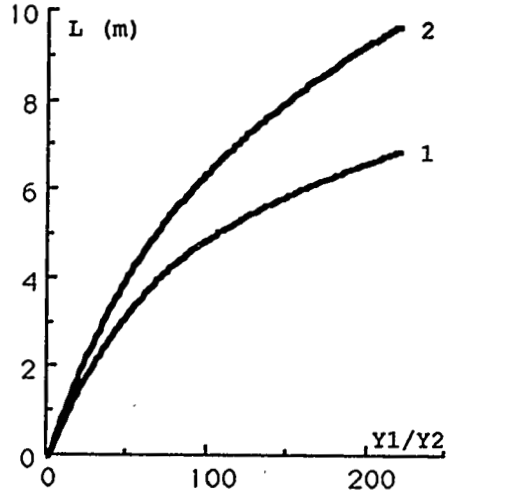
Table 2d

Influence of axial dispersion on column height

Figures 6 and 7 show the influence of the axial dispersion on the column height, compared with the column height obtained with plug flow. Two cases are investigated :

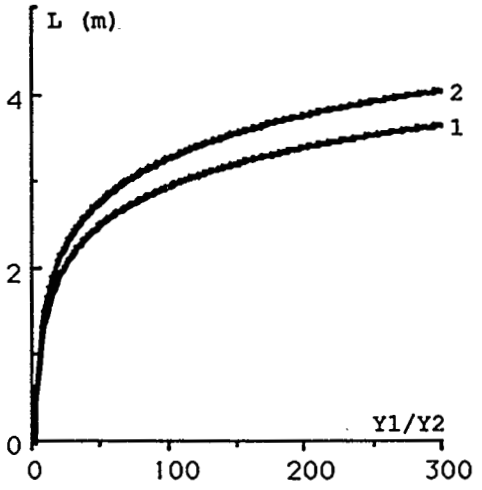
- Case 1 : mass transfert resistances in the two phases (figure 6). When $Y_1/Y_2 > 50$ axial dispersion begins to have some influence on column height..

- Case 2 : mass transfert resistance is located in one phase (fast reaction, instantaneous surface reaction I_2-air-NaOH). The column height is not significally influenced by axial dispersion (figure 7)



	E_G	E_L $(m.s^{-1})$
curve 1	0	0
curve 2	$596*10^{-4}$	$14*10^{-4}$

Fig 6
Column height versus efficiency



	E_G $(m.s^{-1})$
curve 1	0
curve 2	$596*10^{-4}$

Fig. 7
Column height versus efficiency

Operating conditions :

$U_L = 14.15*10^{-3}$ m.s^{-1}
$U_G = 0.34$ m.s^{-1}
$\epsilon = 0.78$
$\beta_G = 0.56$
$a_E = 240$ m^{-1}
$K_G = 5*10^{-3}$ m.s^{-1}

$$\frac{Y1}{Y2} = \frac{\text{inlet conc. phase}}{\text{oulet conc. phase}}$$

Mass transfert coefficient k_G

With I_2-air-NaOH system, the variation of mass transfert k_G ($m.s^{-1}$) versus gas flowrate is shown in figure 8. A regression gives :

$$k_G = 0.22 \quad G^{0.80} \quad (r^2 = 0.92)$$

$m.s^{-1}$ K moles.$m^{-2}.s^{-1}$

The liquid flowrate has no influence on the mass transfer coefficient k_G. This result is correct because the reaction (instantaneous) takes place at the gas-liquid interface and is controlled by diffusion throught the gas film. This correlation can be compared with results obtained with CO_2-air-NaOH which is easier to use. Figure 9 shows the values of k_G ($m.s^{-1}$) versus gas flowrate G for three types of packing. The influence of packing is not very important. So if we correlate k_G versus G we find.

$$k_G = 0.34 \quad G^{0.80} \quad (r^2 = 0,93)$$

$m.s^{-1}$ K moles.$m^{-2}.s^{-1}$

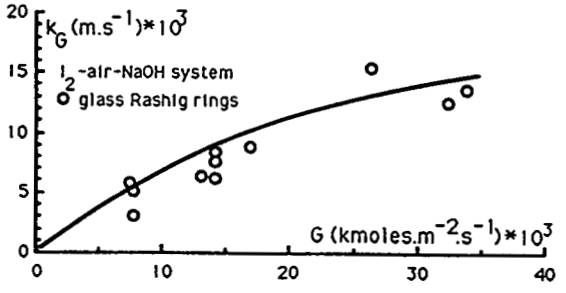


Fig 8
$K_G(I_2)$ versus gas flowrate

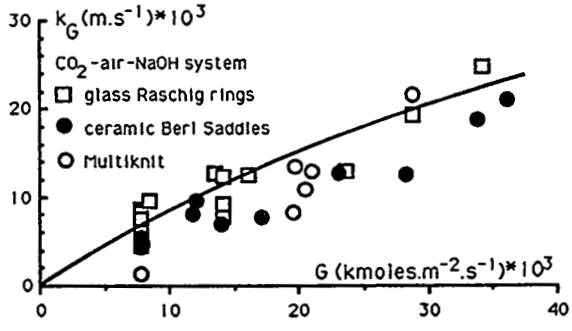


Fig. 9
$K_G(CO_2)$ versus gas flowrate

The $k_G(I_2)$ values are lower than to those obtained for CO_2. That can be explained by the difference between the diffusivity coefficients of CO_2 and I_2 air ($D(I_2)/D(CO_2) = 0.5$). If Higbies'model is valid, $k_G(I_2)/k_G(CO_2)$ is equal to 0.7, which corresponds to the experimental values.

V - Conclusions

The main hydrodynamic characteristics of different types of packing were determined : pressure drop, liquid holdup, interface area, axial dispersion coefficient.

Mass transfer was difficult to investigate with an I_2-air-NaOH system. So another system (CO_2-air-NaOH) was used to calculate values of the gas film mass transfer coefficient k_G (CO_2) and consequently the values of $k_G(I_2)$.

The hydrodynamic and mass transfer data, can then be used to optimize the design of packing columns.

Nomenclature

a_C	geometrical interface area	L^{-1}
a_E	measured interface area	L^{-1}
C_{Ai}	gas concentration at equilibrium	ML^{-3}
C_{BL}	concentration of B in liquid phase	ML^{-3}
D_{AL}	diffusivity of A in liquid phase	L^2T^{-1}
E_G	axial dispersion coefficient in gas phase	L^2T^{-1}
E_L	axial dispersion coefficient in liquid phase	L^2T^{-1}
G_M	molar gas flowrate per unit cross section of column	$ML^{-3}T^{-1}$
He	henry's law constant	L^2T^{-2}
k_2	2nd order Kinetic constant	$M^{-1}L^3T^{-1}$
k_G	gas film mass transfer coefficient	LT^{-1}
L	packing height	L
N_A	matter flux of component A	$ML^{-2}T^{-1}$
P_T	work pressure	$ML^{-1}T^{-2}$
Pe	peclet number	
p_e	partial pressure of CO2 at inlet gas phase	$ML^{-1}T^{-2}$
p_s	partial pressure of CO2 at oulet gas phase	$ML^{-1}T^{-2}$
t	time	T
U_G	superficial velocity of gas phase	LT^{-1}
U_L	superficial velocity of liquid phase	LT^{-1}
U_I	internal velocity of a phase	LT^{-1}
Y	concentration of I_2 in gas phase	ML^{-3}

U.S. Department of Energy.

Reference

1 -B. VIGNAU, "Etude du fonctionnement d'une colonne d'absorption d'iode"
These Doctorat, INSA TOULOUSE, Septembre 1986, n° 27

2 -A. LAURENT, Thèse Doctorat d'Etat, I.N.P.L., Nancy, Décembre 1975, n° AO.12161.

3 -J.M. PERRY, Chemical Engineers Handbook, 4th Edition (1963).

4 -A.D. VIDWANS, MM. SHARMA, Chem, Eng. Sci., 1967, vol. 22, p. 673-684.

5 -W. HAMILTON, G.B. BARTHOLOMAI, British Chemical Engineering and Process Technology (1971), vol. 16, n° 12, p. 1133-1134.

6 -W. EGUCHI, M. ADACHI, Y. MIYAKO, T. SAKAMOTO, Journal of Nuclear Sciences and Technology, 12 (9), p. 567-580, 1975.

IODINE AND NO_x BEHAVIOR IN THE DISSOLVER OFF-GAS AND IODOX SYSTEMS IN THE OAK RIDGE NATIONAL LABORATORY INTEGRATED EQUIPMENT TEST FACILITY

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Abstract

This paper describes the most recent in a series of experiments evaluating the behavior of iodine and NO_x in the Integrated Equipment Test (IET) Dissolver Off-Gas (DOG) System. This work was performed as part of a joint collaborative program between the U.S. Department of Energy and the Power and Nuclear Fuel Development Corporation of Japan.

The DOG system consists of two shell-and-tube heat exchangers in which water and nitric acid are removed from the dissolver off-gas by condensation, followed by a packed tower in which NOx is removed by absorption into a dilute nitric acid solution. The paper also describes the results of the operation of the Iodine Oxidation (IODOX) System. This system serves to remove iodine from the DOG system effluent by absorption into hyperazeotropic nitric acid.

Prior to the most recent experiments, three phases of testing were performed, one in the laboratory and two in the IET facility. Significant modifications in operational and experimental procedures were introduced during the latest test to simplify system operation and constituent analyses. These modifications included installing continuous NO_x analyzers, improving the control of air inleakage into the process, and using 131-I to facilitate iodine analysis by measuring the gamma activity level.

The results of the testing indicate strong and varied interactions between the components in the NO_x-H_2O-HNO_3 system. Due to the range of equilibrium relationships present in this system, the behavior of these components is strongly influenced by the relative concentration levels. The test results also indicate a reasonably high removal efficiency in the NO_x scrubber, even at relatively low feed gas concentrations.

Results from iodine testing indicate that only 2 to 4% of the iodine present in the vapor exiting the dissolver is returned to the dissolver in liquid recycle streams from the off-gas condensers and the NO_x scrubber. These results are consistent with those obtained in previous phases of testing. Some

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*Operated by Martin Marietta Energy Systems, Inc., for the U.S. Department of Energy.

accumulation of iodine in the NO_x scrubber solution was observed. However, the short duration of the iodine test prevented the attainment of steady state in the scrubber with respect to iodine loading and distribution. Iodine removal efficiencies observed in the IODOX system were much lower than anticipated.

1. Background

The Integrated Equipment Test (IET) facility was constructed in the early 1980s as part of the Consolidated Fuel Reprocessing Program. The IET was built to provide a facility for the development and testing of processes and equipment used in the reprocessing of spent nuclear reactor fuel.

A principal chemical process in nuclear fuel reprocessing is the dissolution of the spent fuel, which consists of heavy metal oxides and various fission products. The process (which typically uses nitric acid as the dissolvent) results both in the formation of heavy metal nitrates and oxides of nitrogen (NO_x) and in the release of volatile fission products such as krypton, xenon, and iodine.

From its inception, the IET facility has utilized a continuous dissolution concept. This concept has been realized in the demonstration of a continuous rotary dissolver, which produces an aqueous uranyl nitrate/nitric acid solution when fed continuously with nitric acid and semicontinuously with uranium oxide. Associated with the dissolution process is an off-gas removal system for the treatment of the gaseous effluent. As originally installed, the DOG treatment system consisted of a packed NO_x scrubber and ancillary equipment (Fig. 1). This system serves the dual purposes of removing a large quantity of moisture from the DOG (which exits the dissolver at temperatures in the range 348 to 368 K) and reducing the DOG NO_x concentration to $\leq$ 1%. Both of these steps are prerequisites for effective removal of iodine in the IODOX process (Fig. 2) in which iodine is absorbed into hyperazeotropic nitric acid. Evaporation of the IODOX scrubber liquor results in the crystallization of hydroiodic acid (HIO_3) formed during absorption.

In facilities in which the heavy metal processing rate is relatively low (less than 0.5 tonne/day) the off-gas contribution from the dissolution reaction is small relative to the contributions from inleakage, instrument bubblers, and air spargers. As a result, the NO_x concentration in the DOG may be sufficiently low to have no adverse impact on the iodine removal efficiency of the IODOX process. In cases such as this, the only pretreatment required prior to iodine removal is drying.

Development of the double overhead condenser concept for DOG treatment was directed toward effectively cooling and drying the DOG while minimizing the amount of iodine recycled to the dissolver. Calculations performed using the equation for the

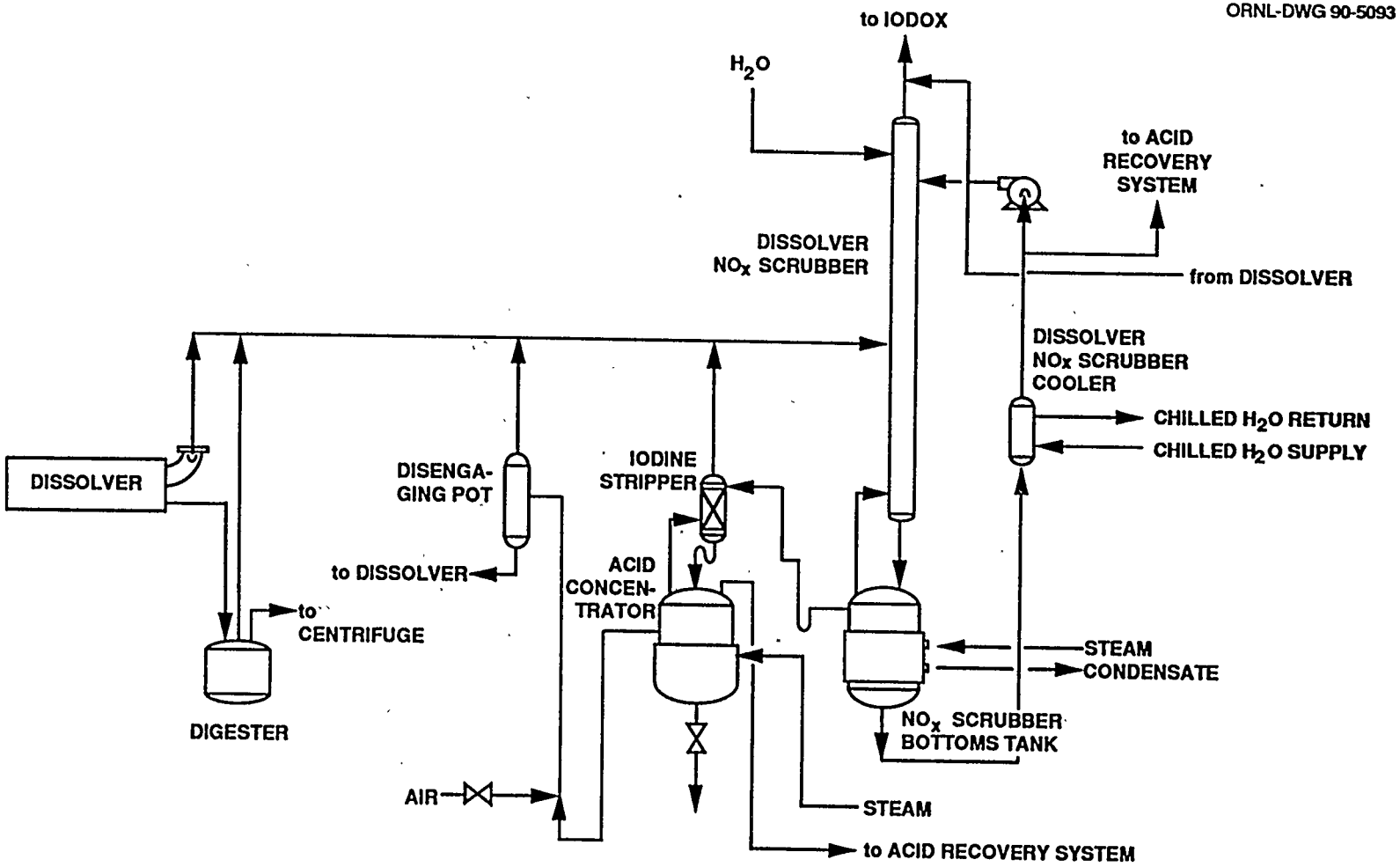

Fig. 1. Original configuration of IET dissolver, NO_x scrubber, and supporting equipment.

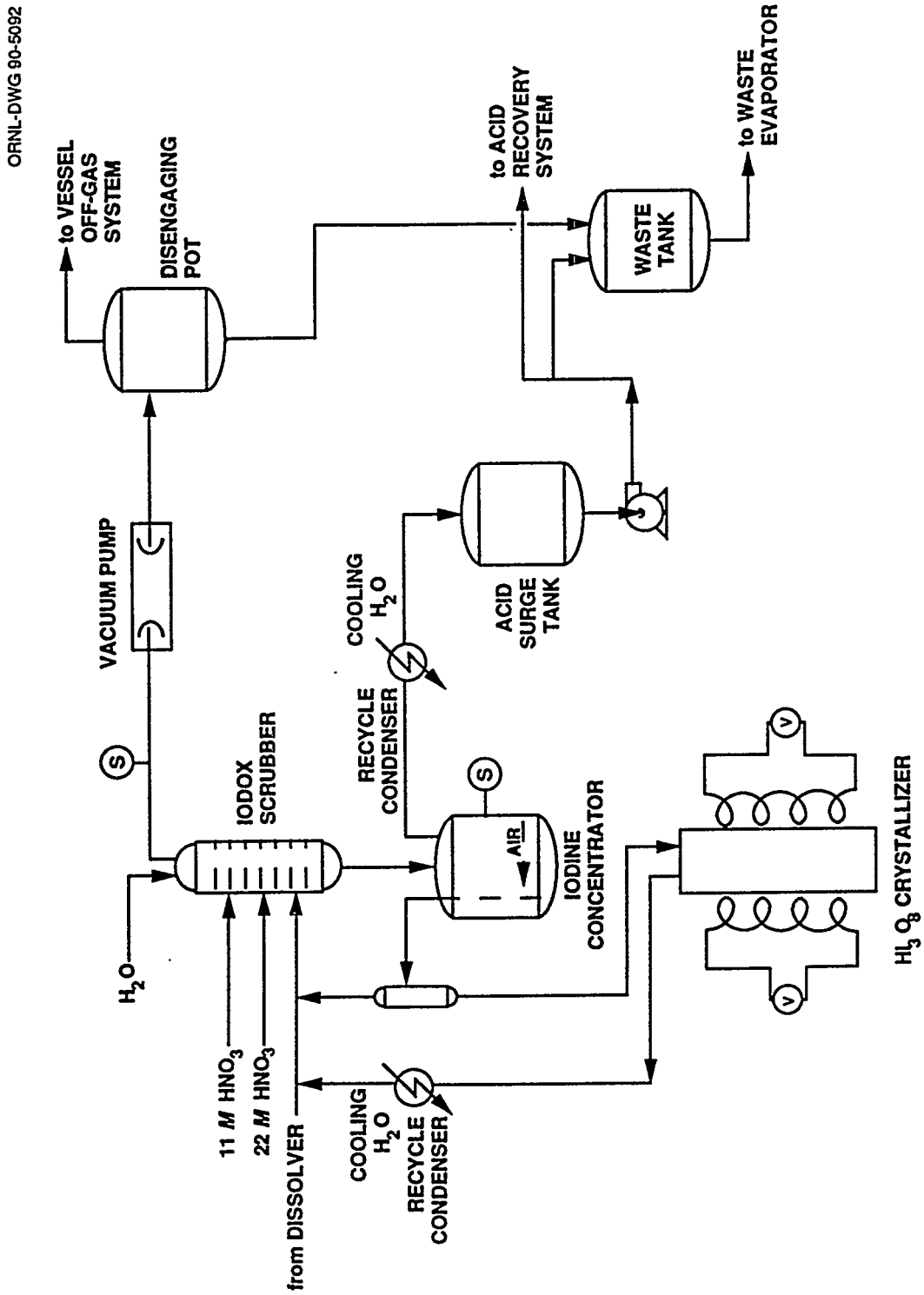

Fig. 2. IODOX system schematic with sample withdrawal points indicated.

iodine distribution coefficient,

$$\ln D = [4793.4 - 150.9(M)](1/T) + 0.547(M) - 11.886 , \quad (1)$$

where

D = I_2 distribution coefficient, (molarity $I_2)_{liq}$/(molarity $I_2)_{gas}$,

T = temperature (K) ,

M = acid concentration in liquid (mol HNO_3/L) ,

indicated that a stepwise cooling of the DOG stream could accomplish these objectives. Based on these calculations, a laboratory evaluation of the viability of the double overhead condenser concept was performed. The results of this study provided qualitative confirmation that cooling and drying of the DOG could be achieved with excellent retention of the iodine in the vapor phase. Subsequently, two shell-and-tube condensers were installed in the IPD dissolver off-gas stream for testing (Fig. 3).[1] This testing was performed in two phases. In the first phase, the dissolver operating conditions were maintained within the normal temperature and pressure ranges without performing uranium oxide dissolution. Iodine was fed to the dissolver in the form of potassium iodide solution. In the second phase, the off-gas system was operated in conjunction with the shear, which delivered uranium oxide and simulated hulls to the dissolution system, generating NO_x.

Results from the first phase of experiments in the IPD indicate that the removal of iodine from the vapor phase is inversely proportional to temperature. This finding is consistent with Eq. (1), which yields increasing liquid/vapor distribution coefficients at decreasing temperatures. While the rate of gas flow did have some effect on the removal of iodine from the vapor phase, this relationship was not as pronounced as the correlation between temperature and liquid/vapor partitioning.

For the two condensers in series, the overall iodine removal efficiency was quite low, ranging from 0.35 to 6.29%. Individual removal efficiencies for the two condensers ranged from 0.03 to 5.78% for the first condenser and from 0.02 to 3.42% for the second condenser. In general, the results of this study indicated that the overall iodine removal efficiency was lowest when the first condenser operating temperature was in the 323 to 343 K temperature range.

The combined NO_x removal efficiency in the two condensers ranged from 5 to 58%, but was generally 20%, $\pm$7%. The overall NO_x efficiency for the DOG system (condensers and scrubber) ranged from 40 to 60%. Scrubber effluent NO_x concentrations ranged from 0.4 to 1.0%. Scrubber removal efficiencies were

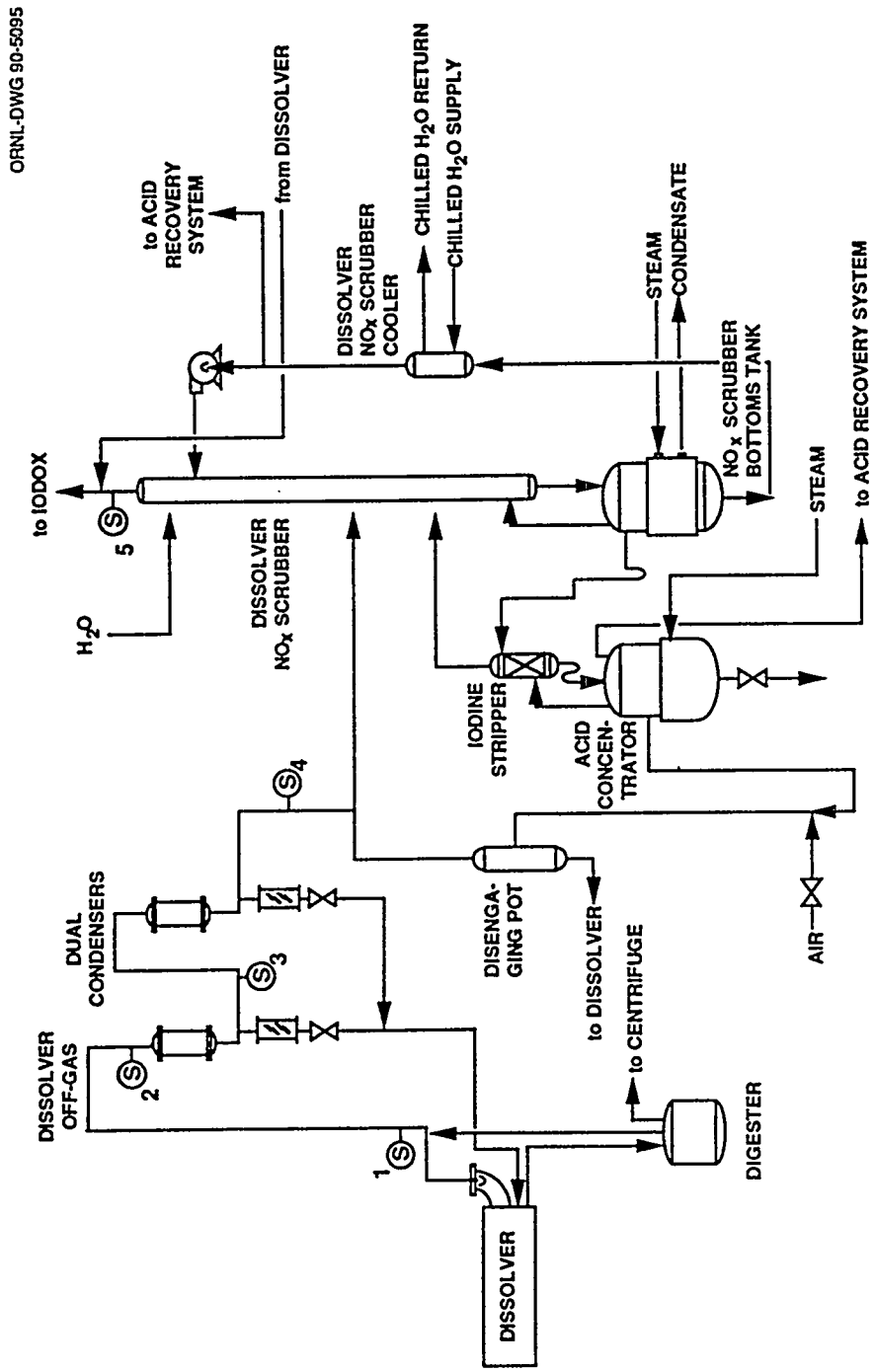

Fig. 3. Current configuration of IET dissolver and NO_x scrubbing system.

generally low because of low concentrations of NO_x present in the effluent from the second condenser.

It was observed that the NO_x removal efficiencies of the condensers and the scrubber complemented each other (i.e., under conditions at which scrubber efficiencies were low the condenser NO_x removal efficiencies were high. The results were the converse when scrubber efficiencies were high). In general, the observed NO_x removal capability of the condensers was greatest when the inlet NO_x concentration was low. This tendency suggested that the removal of NO_x in the condensers could be attributed primarily to the condensation of HNO_3, rather than the absorption of gaseous NO_x species into the liquid.

2. Objectives

The described testing provided useful information on the behaviors of iodine and NO_x in the DOG system. However, operational difficulties and limitations in the sampling and analytical techniques affected the precision and validity of the results. The more recent phases of testing were performed with modifications made in system operation, data collection, and analysis, which were intended to improve the quality of the data collected.

To minimize confusion between the two phases of experiments described in Sect. 1 and later work, the most recent off-gas experiments will be referred to as Phases 3 and 4. The primary objective of Phase 3 was to characterize the off-gas generated by the dissolution of uranium oxide. This test was performed by using the IET Feed Station to deliver feed material to the dissolver incrementally to simulate shear operation. In addition to determining the concentrations of nitrogen oxides in the DOG, gas- and liquid-phase samples were taken throughout the dissolver and DOG systems to obtain data regarding NO_x distribution and behavior in both phases.

The objectives of Phase 4 were (1) to study the behavior of NO_x and iodine in the off-gas system and (2) to evaluate the iodine and NO_x removal capabilities of the IODOX and DOG systems respectively. To both simplify system operation and provide the capability to vary test conditions, generating NO_x by dissolution of uranium oxide was replaced by feeding NO and NO_2 into the dissolver from gas cylinders. The gas feed rates were predetermined to produce NO_x concentrations in ranges approximating those observed in Phase 3 testing. Note that whereas dissolution of uranium oxide under simulated shear conditions generates off-gas in pulses, Phase 4 testing was performed at constant gas feed rates to simplify analysis of the results.

3. Description of Experiments

3.1 Phase 3

As stated in Sect. 2, Phase 3 off-gas was generated by introducing uranium oxide to the dissolver under conditions simulating feeding from a fuel shear device. Under these conditions, a feed increment consisting of uranium oxide and simulated sheared pin segments is fed to the dissolver once every 90 s. Based on an established shear operation scheme, complete shearing of one assembly requires 72 cuts. Therefore, the total duration of a dissolver feed period is 108 min. Again, based on assumed shear operating scenarios, a 72-min shear reload period was simulated during which no additional feed was introduced into the dissolver. Accordingly, a complete simulated shear cycle used in Phase 3 testing lasted 180 min.

NO_x samples were withdrawn at five locations throughout the DOG system: at the dissolver off-gas outlet (Sample Point 1), immediately upstream from the first condenser (Point 2), immediately upstream from the second condenser (Point 3), downstream from the second condenser (Point 4), and downstream from the DOG NO_x scrubbing column (Sample Point 5).

The results of Phases 1 and 2 indicated that moisture was condensing in the gas sample bombs as the samples were collected. Material balance calculations seemed to indicate that this condensation was resulting in the absorption of some NO_x from the sample prior to analysis. To mitigate this problem, a system of infrared NO and NO_2 analyzers was installed to provide online analysis capabilities. As shown in Figs. 4 and 5, the off-gas sample streams were drawn to the analyzers using vacuum pumps located downstream of each analyzer. Two analyzer stations were utilized, each consisting of one NO and one NO_2 analyzer. A valve panel was configured to allow switching between Sample Points 1, 2, and 3 at analyzer station A, and between Points 4 and 5 at analyzer station B. Before entering the analyzers, the gas sample streams from Points 1, 2, and 3 were passed through heat changers to cool and partially dry the gas, preventing condensate formation in the analyzers and the vacuum pumps. The off-gas conditions at Points 4 and 5 were such that additional drying was not required. The sample cells in the analyzers are maintained at a temperature of 328 K as further protection against condensate formation.

In addition to gas samples, liquid samples from the sample line condensers, overhead condensers, scrubber bottoms tank, and the acid concentrator were collected and analyzed for HNO_2 and HNO_3.

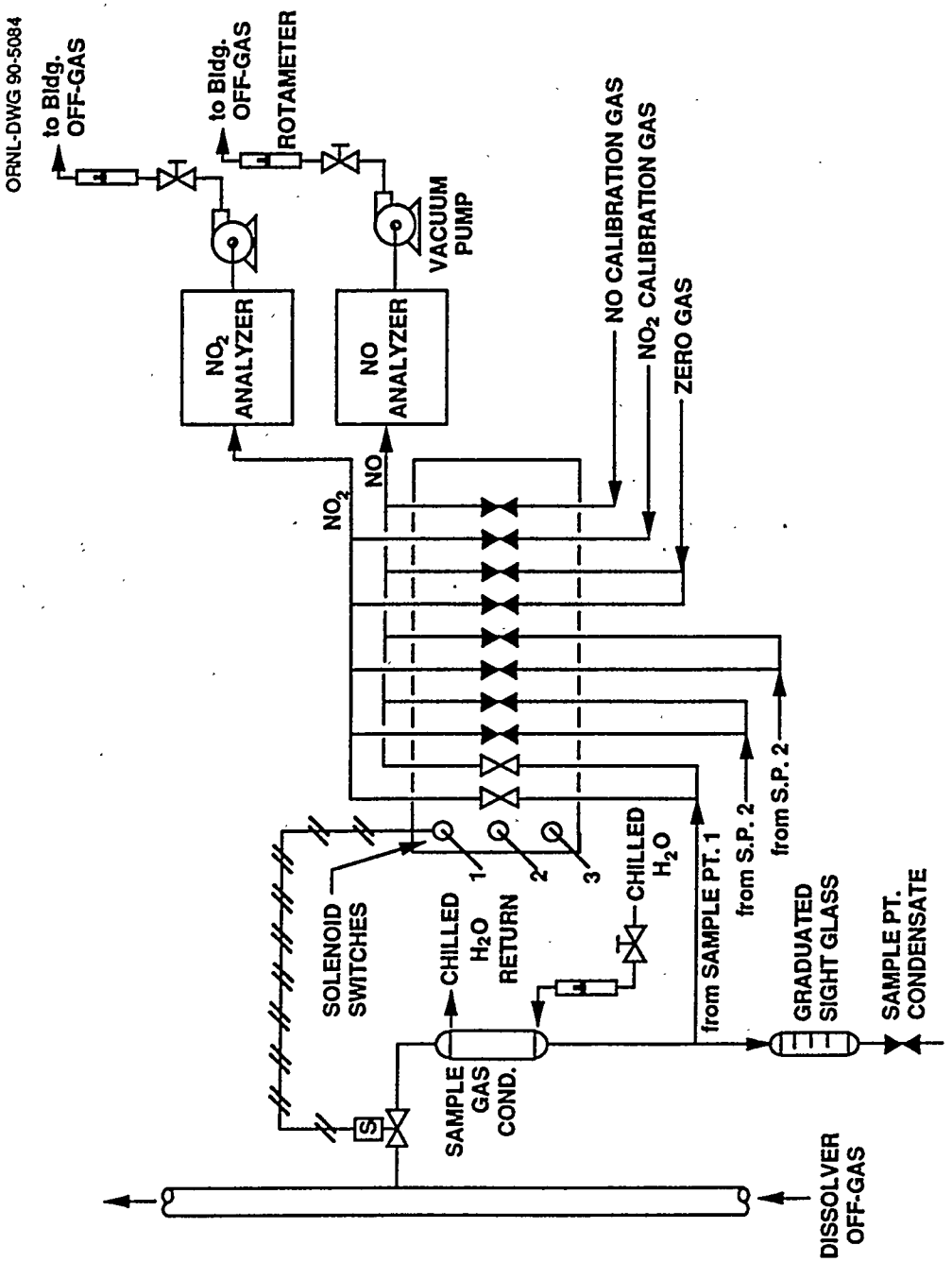

Fig. 4. Analyzer station *A* schematic.

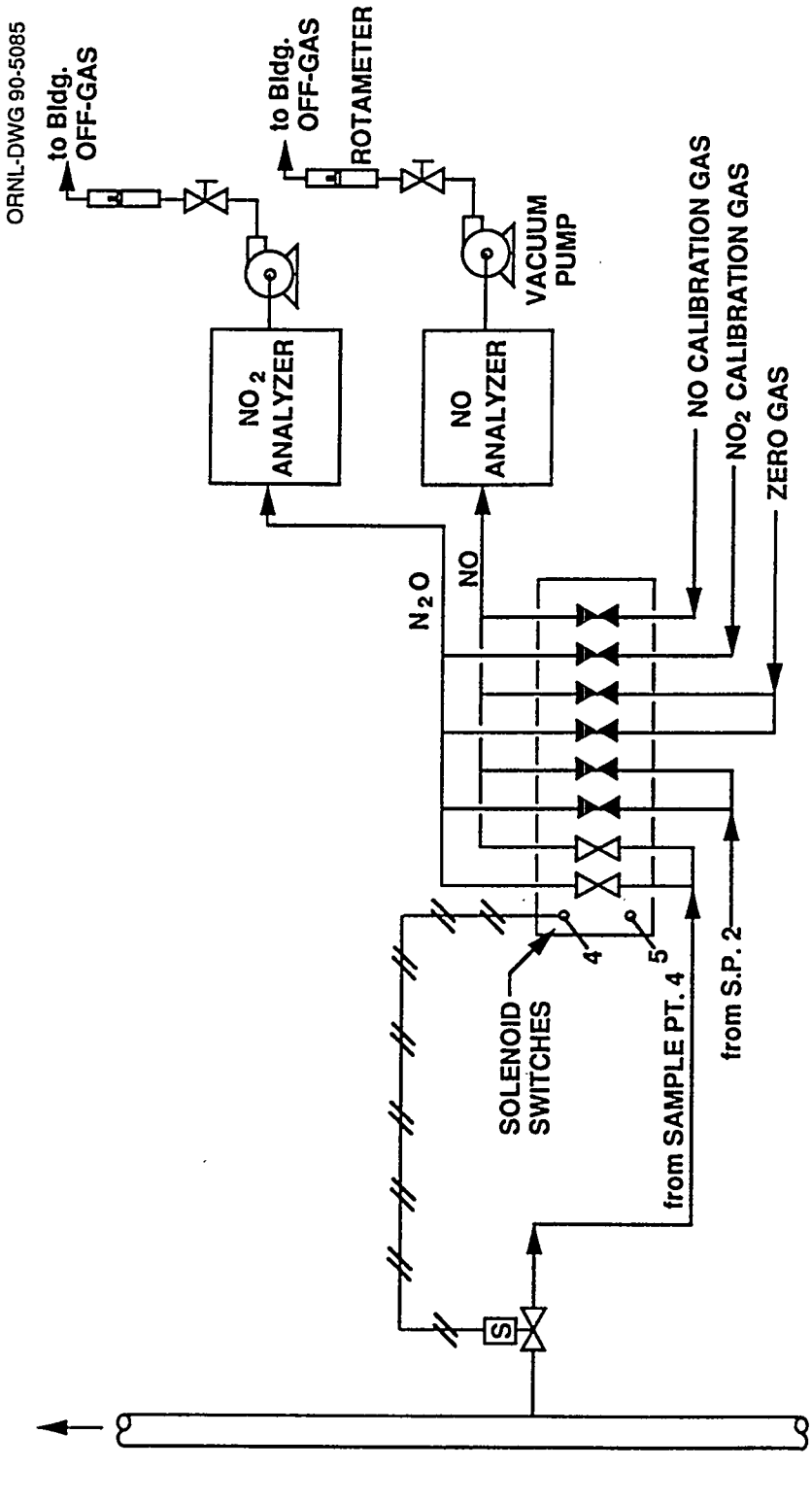

Fig. 5. Analyzer station *B* schematic.

3.2 Phase 4

Phase 4 testing was performed in two parts. In part A, additional data were obtained regarding the behavior of NO_x in the system. Based on the results of the Phase 3 tests, a range of DOG NO and NO_2 concentrations were selected for use in Phase 4. To minimize the likelihood of process upsets and to increase the manpower availability for sampling, uranium dissolution was not performed during this phase. Instead, NO and NO_2 from commercially available gas bottles were used to simulate dissolver vapor-phase effluents.

The NO_x analyzer stations were configured as in Phase 3. In addition to the liquid samples collected in Phase 3, samples of the dissolver product stream were taken from both the digesters and the dissolver seal pot (which is located in the product solution line immediately downstream of the dissolver). To study the effect of gas flow rate on DOG system performance, an air addition line was installed at the acid-feed end of the dissolver. The flow through this line was controlled and metered by means of a rotameter/needle valve combination.

In part B of Phase 4 testing, iodine was added to the dissolver in an aqueous 0.2 M KI, 0.4 M $NaNO_2$ solution. The solution also contained approximately 1.5 mCi of 131-I, which was used as a tracer. Gas-phase iodine samples were collected by adsorption onto activated carbon beds using the sampler configuration shown in Fig. 6. As the figure indicates, each sampler consisted of two adsorption beds in series (to ensure complete adsorption); a rotameter to measure sample gas flow; a vacuum pump to induce flow through the adsorption beds; and an ice bath to dry the sample gas, protecting the vacuum pumps from condensate. Samples were taken at locations corresponding to the NO_x gas sample locations. In addition, iodine gas sampling was performed downstream of the IODOX column. Because of the extremely corrosive nature of iodine in the form HI, the NO_x analyzers were valved out of the off-gas system during this last phase of testing.

During Phase 4, the IODOX system was operated to remove iodine from the NO_x scrubber effluent gas stream. The acid concentration system was also operated in conjunction with IODOX in order to generate the hyperazeotropic acid required for iodine scrubbing.

During Phase 4B, liquid samples were collected from the overhead condensers, the dissolver seal pots (dissolver product stream), the dissolver KI feed solution, the digesters, the NO_x scrubber bottoms tank, the acid concentrator, the iodine concentrator in the IODOX system, and the acid product surge tank in the acid concentration system.

Analyses for iodine content of gas samples and liquid samples were performed by measuring the gamma activity level of

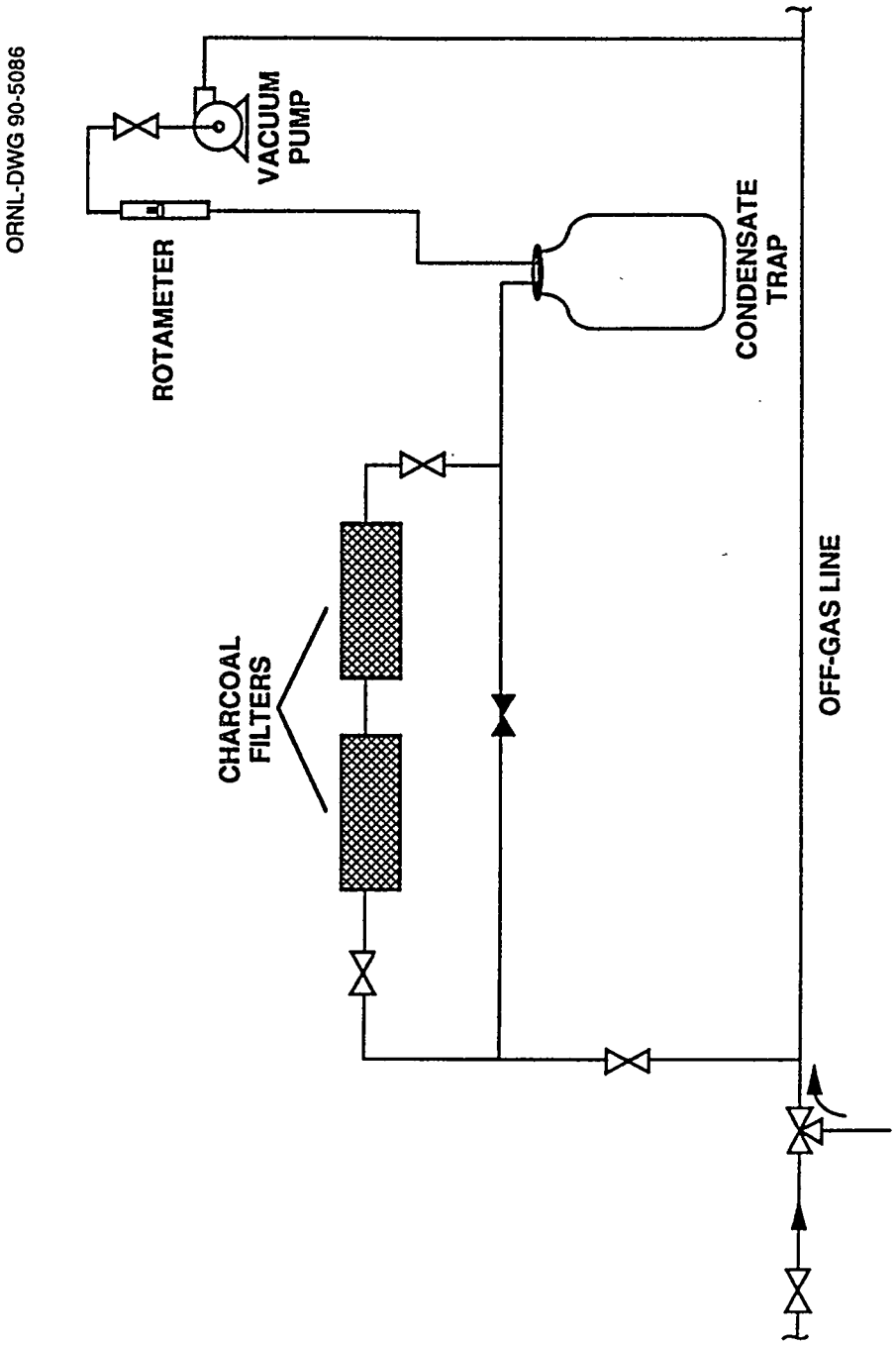

Fig. 6. Vapor-phase iodine sampling system.

the samples. By using the known concentration of the dissolver KI feed solution and the sample volume, the concentration/gamma activity ratio was determined. Similarly, vapor phase iodine concentrations were derived from the observed gamma activity level, the flow rate of sample gas through the adsorption beds, and the duration of flow. In addition to iodine, all liquid samples collected were also analyzed for HNO_2 and HNO_3 content. Liquid-phase iodine samples were analyzed by wet chemical techniques in addition to the gamma activity method.

4. Results

4.1 Phase 3

The results from Phase 3 were somewhat sporadic, because of operational difficulties with both the dissolver and the feed mechanism. These difficulties resulted in test termination after 15 h of data collection; hence, steady state with respect to HNO_2 and HNO_3 liquid-phase concentrations was not achieved in the DOG scrubbing system. However, the vapor-phase results indicate that at a feed rate of 500 kg U/day and over the range of DOG flows from 500 to 1000 SLM (dry gas basis), the NO concentration in the off-gas exiting the dissolver ranged from near zero to 0.1%, and that of NO_2 ranged from 0.5 to 1.25% (Fig. 7).

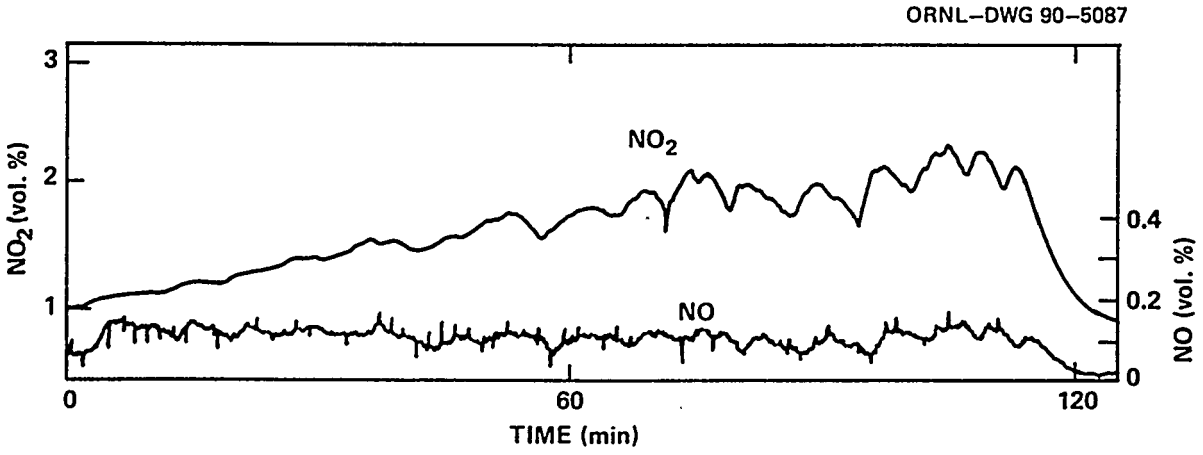

Fig. 7. NO, NO_2 concentrations at the dissolver outlet (Sample Point 1).

NO_2 concentrations recorded at the dissolver exit were observed to increase steadily during a uranium oxide feed period. NO concentrations observed at this point were relatively stable throughout a simulated shear cycle. In contrast, the concentrations of both NO and NO_2 at Sample Points 2, 3, and 4 were observed to rise and fall sharply during a shear cycle, at a frequency corresponding to that of uranium oxide addition. A possible explanation for this observation involves the accumulation of liquid in the sample line between Sample Point 1 and the NO_x analyzers. Because of the relatively high humidity of the off-gas stream exiting the dissolver and the relatively short residence time of the gas in the sample condenser, it is expected that condensate accumulates in this line during testing. This supposition is supported by the surprisingly small amounts of condensate recovered from the Sample Point 1 condenser.

The consistent shift in the concentrations of NO and NO_2 between Sample Points 1 and 2 (Fig. 8) can only be explained by the occurrence of chemical reactions between the dissolver and first condenser. Vapor-phase chemical reactions that may occur in the NO_x-H_2O-HNO_3 system include:[2]

$$2NO_2\ (g) \leftrightarrow N_2O_4\ (g)\ , \tag{2}$$

$$NO\ (g) + NO_2\ (g) \leftrightarrow N_2O_3\ (g)\ , \tag{3}$$

$$NO\ (g) + NO_2\ (g) + H_2O\ (g) \leftrightarrow 2HNO_2\ (g)\ , \tag{4}$$

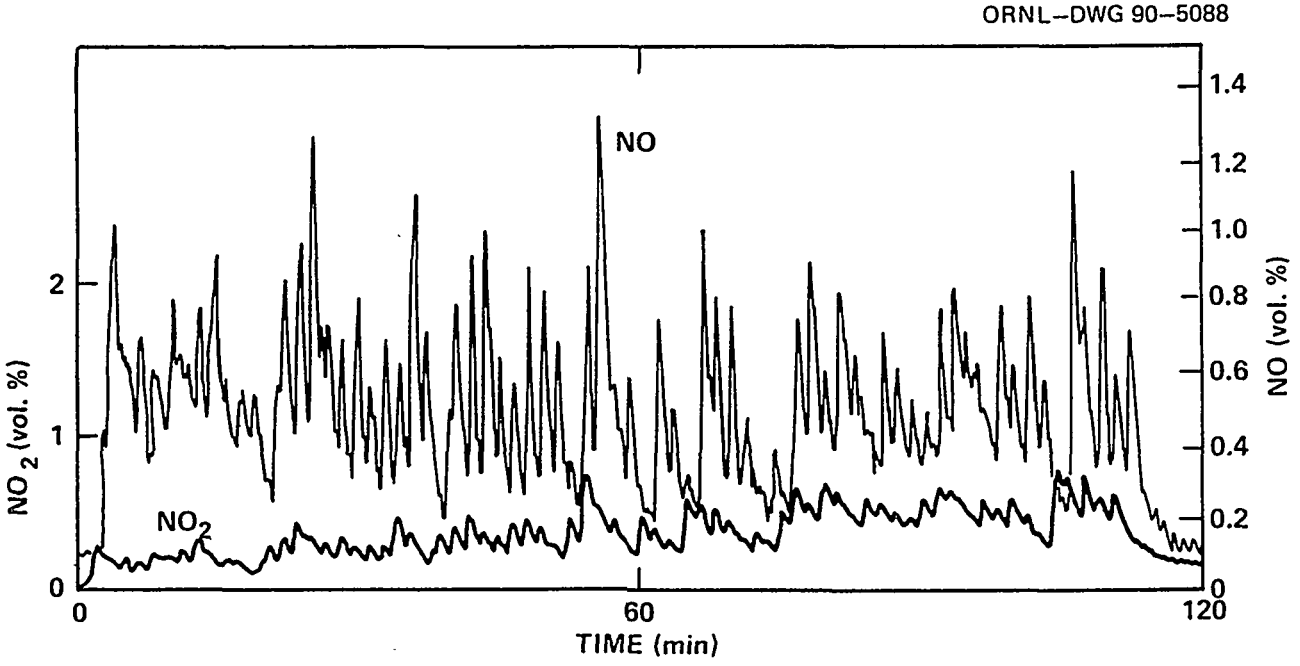

Fig. 8. NO, NO_2 concentrations at first condenser inlet (Sample Point 2).

$$3NO_2\ (g) + H_2O\ (g) \leftrightarrow 2HNO_3\ (g) + NO\ (g)\ , \qquad (5)$$

and

$$2NO\ (g) + O_2\ (g) \rightarrow 2NO_2\ (g)\ . \qquad (6)$$

A proposed stoichiometry for the dissolution reaction of U_3O_8 (the uranium oxide form used during Phase 3) is

$$U_3O_8 + 7.35HNO_3 \rightarrow 3UO_2(NO_3)_2 + NO_2 + 0.35NO + 3.65H_2O\ . \qquad (7)$$

The failure to develop a homogenous off-gas mixture in the dissolver coupled with the probable occurrence of chemical reactions involving NO, NO_2, and H_2O prevents the verification of this equation from the data collected in Phase 3.

Equilibrium constants calculated for reactions (2), (3), and (4) at the nominal dissolver off-gas temperature of 363 K are 0.116 atm^{-1}, 0.031 atm^{-1}, and 0.084 atm^{-2} respectively. Given the typical concentrations of NO and NO_2 in the dissolver off-gas are on the order of 10^{-2} atm, the concentrations of HNO_2, N_2O_4, and N_2O_3 inside the dissolver will be negligible, regardless of rate.

In addition to the gas samples, condensate from the off-gas was collected at each of the first three sample points. The HNO_3 concentration in these samples ranged from 1.2 to 1.54 M at Sample Point 1, from 0.33 to 0.74 M at Sample Point 2, and 1.68 to 1.7 M at Sample Point 3.

To determine the minimum relative humidity of the off-gas at Sample Point 2, an off-gas flow rate at the upper end of the observed range (1000 SLM dry air) is selected. Assuming the air exiting the second condenser is saturated with water at 293 K, the relative humidity of the gas entering the first condenser was determined by measuring the rate of condensate removal at each of the two condensers. During Phase 3 testing, these condensate rates were observed to be 150 ml/min at the first condenser and 70 ml/min at the second condenser, nominally. Therefore, the off-gas entering the first condenser must have contained approximately 240 g of water in approximately 1300 g of dry air, and has a relative humidity at 343 K (the nominal gas temperature at the first condenser inlet) of 75%.

Studies by Smith[3] indicated that the formation of NO_2 from NO and HNO_3 by reverse reaction (5) is apparently catalyzed by NO_2 and H_2O. Studies by Leffers[4] et al. using a wetted wall column contacted NO with aqueous HNO_3. These studies determined that NO is oxidized to NO_2 by vapor-phase nitric acid and then is absorbed into the liquid phase after conversion to N_2O_4.

Given that the presence of a significant amount of HNO_3 in the dissolver off-gas is indicated by the concentration measured in the condensate Sample Point 1, and that a considerable

quantity of water vapor is also present, it appears likely that reaction (5) occurs in the reverse direction. This is especially true at the dissolver outlet where the concentrations of water and nitric acid vapor are at the highest levels in the DOG system. The occurrence of this reaction would account for the deviation between the observed ratio of NO to NO_2 at the dissolver and the stoichiometric ratio indicated by reaction (7). The oxidation of NO to NO_2 by reaction (6) may also be partly responsible the overabundance of NO_2 at the dissolver outlet, although data from a number of researchers indicate that this reaction will occur at a rate one order of magnitude slower than reverse reaction (5).

The depletion of NO_2 and the accompanying increase in NO concentration between Sample Points 1 and 2 are most likely due to the occurrence of reaction (2) in the vapor phase, followed by the equilibrium absorption of NO_2-N_2O_4 according to the expression

$$1.5N_2O_4\ (g) + H_2O\ (l) \leftrightarrow 2HNO_3\ (l) + NO\ (g)\ , \qquad (8)$$

which has been proposed by Carberry.[5] This reaction is facilitated by the condensation of water inside the uninsulated off-gas pipe in the 40 ft section between Sample Points 1 and 2. Additional depletion of NO_2 and formation of NO is probably because of the occurrence of the liquid phase reaction

$$N_2O_4\ (l) + H_2O\ (l) \rightarrow HNO_3\ (l) + HNO_2\ (l)\ , \qquad (9)$$

and the subsequent decomposition of HNO_2 according the expression

$$3HNO_2\ (l) \leftrightarrow HNO_3\ (l) + H_2O\ (l) + NO\ (g)\ . \qquad (10)$$

The occurrence of reaction (10) in Phase 3 testing is indicated by the low concentrations of HNO_2 observed in the liquid phase throughout the off-gas system.

The concentrations of NO in the gas phase immediately upstream of the second condenser (Sample Point 3) were slightly lower than concentrations measured immediately upstream of the first condenser. However, the time-averaged concentrations of NO_2 at Sample Point 3 during two shear cycles were 1.3 and 1.7% (Fig. 9), which are significantly higher than the average of 0.4% measured at Sample Point 2 during two previous cycles. The only apparent mechanism for NO_2 formation without significant changes in the concentration of NO is the decomposition of nitrous acid by reaction (10) followed by oxidation of all NO formed. In fact, the HNO_2 concentration in the condensate samples decreases from Sample Point 2 to Sample Point 3 (from 2.0 to 0.2 mg/ml) while the HNO_3 concentration increases. At the conditions of the off-gas exiting the first condenser, the rate of NO oxidation is .00475 atm/s. At this relatively slow rate, complete conversion of the NO formed by HNO_2 decomposition between Sample Points 2 and 3 is improbable.

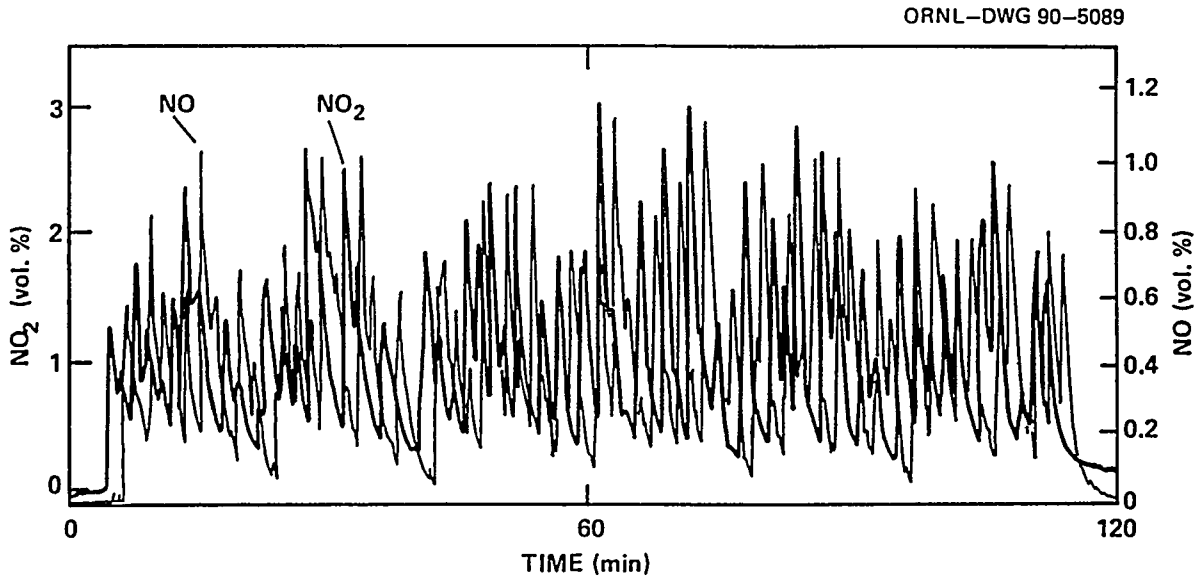

Fig. 9. NO, NO_2 concentrations at second condenser inlet (Sample Point 3).

The off-gas pulses evident at Sample Points 2 and 3 were also observed downstream of the second condenser (Sample Point 4). At this location, the measured concentrations of NO and NO_2 ranged from 1.0 to 1.1% and 0.3 to 0.5% respectively (Fig. 10). The increase in NO and decrease in NO_2 concentration are most likely due to the occurrence of reactions (2), (8), (9), and (10). Reactions (8) and (9) are enhanced between Sample Points 3 and 4 by the condensation of most of the water remaining in the vapor phase downstream of the first condenser. The increase in the HNO_2 concentration in the condensate from the second condenser supports this hypothesis.

The NO and NO_2 concentration spikes seen at Sample Points 2, 3, and 4 disappear at Sample Point 5, located at the exit of the NO_x scrubber. This is most likely due to the gas residence time within this vessel. NO concentrations at the scrubber outlet ranged from 0.2 to 0.4%. NO_2 concentrations ranged from 0.2% to a level not detectible by the infrared analyzer used (Fig. 11). These results are as expected and reflect the absorption of NO_2 into water by reactions (8) and (9). The depletion of NO_2 by absorption then drives reaction (6) toward the oxidation of NO, resulting in the depletion of the oxide.

4.2 Phase 4

NO_x-H_2O-HNO_x System Behavior. The somewhat artificial conditions imposed influenced the results of the NO_x portion of Phase 4 testing which, as a consequence, are not directly representative of off-gas behavior during fuel dissolution. As stated previously, both NO and NO_2 were fed into the dissolver

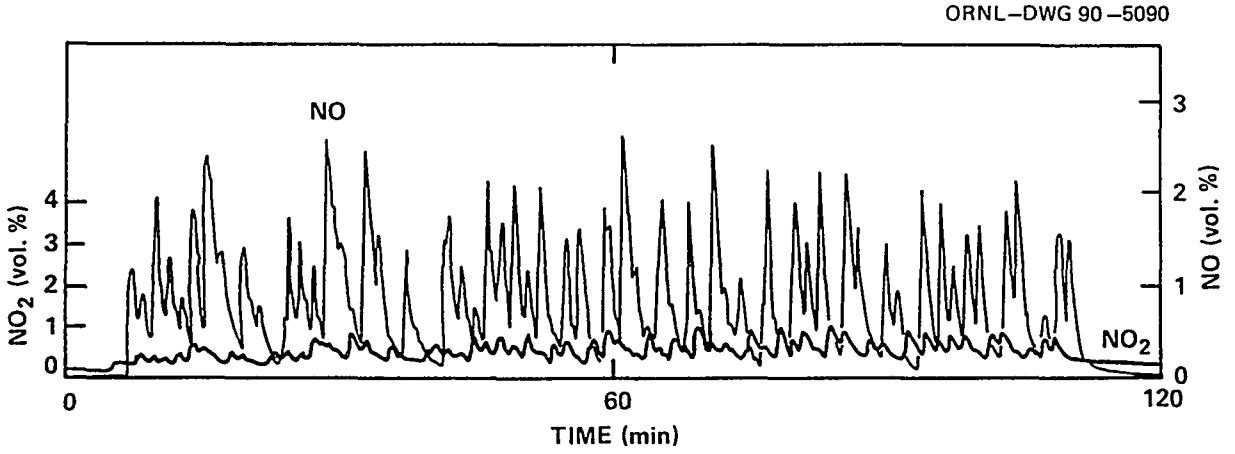

Fig. 10. NO, NO_2 concentrations at second condenser outlet (Sample Point 4).

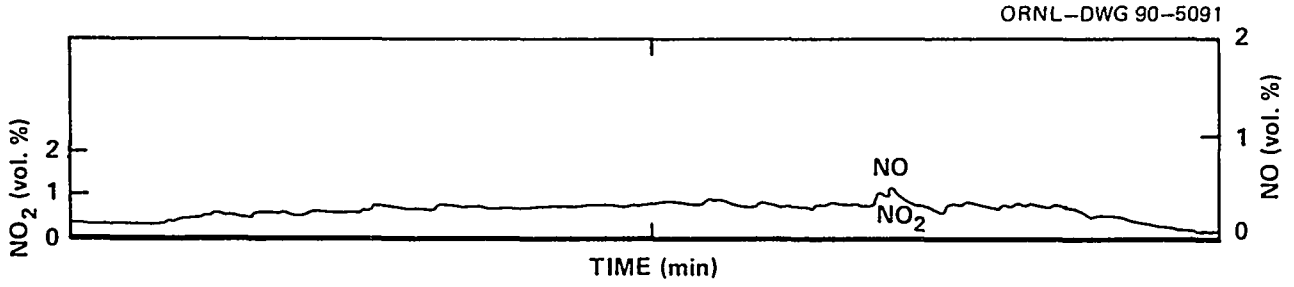

Fig. 11. NO, NO_2 concentrations at NO_x scrubber outlet (Sample Point 5).

from gas cylinders on a continuous flow basis during Phase 4 testing. Both the NO and NO_2 flows into the dissolver were metered to generate NO and NO_2 concentrations in the dissolver off-gas over the range of 1 to 2.5% for each component. The concentrations of the components relative to one another were not adjusted in any way to simulate the U_3O_8 dissolution stoichiometry described in reaction (7). Recognizing that these conditions limit the direct applicability of Phase 4 NO_x results to dissolver off-gas behavior, the elimination of the fluctuations introduced by the intermittent nature of normal dissolver feeding simplifies analysis of interacting NO_x-H_2O-HNO_3 reaction mechanisms.

The amount of NO charged to the dissolver (Table 1) was approximately six times the quantity measured in the off-gas at

Table 1. Dissolver inputs

Sample set	NO_2 feed SLM	NO feed SLM	DOG LPM	DOG K	Material Balance DOG %NO_2	Material Balance DOG %NO
1	0	0	a	a	a	a
2	6.32	8.68	a	a	a	a
3	0	0	a	a	a	a
4	8.60	12.47	1006	348.6	1.02	1.48
5	9.00	12.87	817	351.0	1.32	1.89
6	8.57	8.97	1019	352.3	1.01	1.06
7	11.36	0	744	353.0	1.84	0
8	9.12	15.48	a	a	a	a
9	0	0	686	352.0	0	0
10	10.64	21.82	591	353.1	2.17	4.45
11	8.09	8.89	858	348.0	1.12	1.23
12	8.64	13.34	1008	346.9	1.01	1.57
13	8.58	11.39	1102	346.5	0.92	1.22
14	8.58	15.26	943	346.6	1.08	1.91
15	8.44	11.55	964	344.7	1.03	1.41
16	4.41	17.79	1077	346.8	0.48	1.96
17	9.02	17.93	804	346.1	1.32	2.63
18	11.86	18.16	1185	346.8	1.18	1.81
19	12.06	18.48	998	346.8	1.43	2.19
20	11.47	19.02	803	352.1	1.72	2.85
21	a	0	866	352.0	a	0
22	11.64	0	707	351.2	1.97	0
23	5.28	7.23	859	362.5	0.76	1.04
24	5.29	7.38	945	371.2	0.71	0.99
25	0	6.76	960	364.5	0	0.97
26	0	7.17	842	355.2	0	1.03
27	0	23.27	1013	344.8	0	2.70
28	0	0	1142	363.7	0	0
29	0	19.61	1151	343.2	0	0
30	0	23.83	1041	340.1	0	2.66

[a] No data recorded.

the dissolver off-gas outlet (Table 2). NO_2 results are somewhat inconsistent, with increases in the dissolver NO_2 effluent over the amount charged occurring in approximately half the sample sets collected. As in Phase 3, significant quantities of water and nitric acid were found at the dissolver vapor outlet. These conditions are conducive to the occurrence of reverse reaction (5), in which NO is depleted and NO_2 formed. Under conditions of low NO feed gas flow into the dissolver, the amount of NO_2 exiting the dissolver was found to be equal to or slightly less than the inlet amount. However, at higher inlet NO feed rates, the effluent NO_2 concentration was always higher than would be expected on the basis of the NO_2 feed rate. These findings also support the occurrence of reverse reaction (5) in the dissolver.

Table 2. Sample Point 1 Data

Sample set	Analyzer %NO	Analyzer $\%NO_2$	Temp. (K)	Condensate rate (ml/min)	HNO_2(l) (M)	HNO_3(l) (M)
6	0.15	0.75	352.3	0	[a]	[a]
9	0.15	0.69	352.0	0.0083	2.6E-7	2.32
10	0.15	0.69	353.1	0.0083	7.4E-7	2.94
13	0.00	0.20	346.5	0	[a]	[a]
14	0.30	1.10	346.6	0	[a]	[a]
16	0.12	0.94	346.8	0.558	5.7E-7	2.64
19	0.16	0.87	346.8	0.667	4.3E-7	2.51
22	0.15	0.20	351.2	0	[a]	[a]

[a]No data available.

As in Phase 3, a shift in NO concentration between Sample Points 1 and 2 was observed, with the NO level increasing by from 30 to 100% (Table 3). However, in contrast to the results in Phase 3, a slight increase in NO_2 concentration between Sample Points 1 and 2 was also indicated. HNO_3 concentrations in condensate collected at Sample Point 2 are significantly lower than at Sample Point 1. HNO_2 concentrations in the condensate at Sample Point 2 were generally an order of magnitude greater than at Sample Point 1.

The water content of the gas at Sample Point 2 is still considerable, based on the rate of condensation in the first overhead condenser (Table 4). However, it is suspected that an undetermined amount of water present in the gas at Sample Point 1 condenses before reaching Sample Point 2, because of convective cooling of the off-gas piping between the two points. In addition to water removal, this cooling results in the removal of HNO_3 from the vapor phase, explaining the concentration drop seen between the condensates at Sample Points 1 and 2. The presence

Table 3. Sample Point 2 Data

Sample set	Analyzer %NO	Analyzer $\%NO_2$	Temp. (K)	Condensate rate (ml/min)	HNO_2(l) (M)	HNO_3(l) (M)
4	0.33	0.99	354.8	0.167	1.0E-6	0.69
7	0.45	1.36	354.7	0.080	1.0E-2	0.95
11	0.23	0.70	348.7	0.233	8.3E-7	1.59
14	0.32	1.18	345.4	0.150	1.1E-7	1.15
17	0.57	1.14	346.8	0.250	2.0E-7	0.90
20	0.64	0.57	353.2	0.322	1.1E-6	1.38

Table 4. Condenser Data

	First Condenser			Second Condenser		
Sample set	Condensate rate (ml/min)	HNO_2(l) (M)	HNO_3(l) (M)	Condensate rate (ml/min)	HNO_2(l) (M)	HNO_3(l) (M)
1	a	a	a	a	a	a
2	a	5.3E-3	0.31	219.96	a	a
3	a	3.3E-3	0.21	a	1.6E-2	0.83
4	329.94	6.7E-3	0.26	71.49	2.0E-2	1.51
5	208.96	a	a	60.49	1.0E-2	1.61
6	274.95	5.2E-3	0.15	65.99	1.2E-2	0.99
7	142.97	7.8E-3	0.32	76.99	2.7E-2	2.29
8	219.96	9.6E-3	0.64	71.49	6.5E-7	2.76
9	120.98	5.9E-3	0.27	43.99	3.5E-3	2.68
10	76.99	1.4E-2	0.96	27.50	5.8E-2	3.34
11	115.48	3.7E-3	0.32	49.49	1.3E-2	1.38
12	197.96	3.5E-3	0.25	87.98	1.4E-2	1.40
13	197.96	3.9E-3	0.28	87.98	1.9E-2	1.19
14	131.98	4.6E-3	0.31	93.48	5.4E-3	1.36
15	142.97	3.9E-3	0.26	82.49	1.5E-2	1.13
16	219.96	2.8E-3	0.56	93.48	3.7E-2	2.38
17	197.96	3.5E-3	0.58	71.49	2.4E-2	2.96
18	263.95	8.9E-3	0.68	87.98	2.8E-2	2.70
19	153.97	1.4E-6	0.72	87.98	3.3E-2	2.42
20	219.96	1.4E-2	0.92	60.49	3.4E-2	3.20
21	219.96	1.2E-2	0.74	60.49	2.9E-2	2.90
22	126.48	9.1E-3	0.46	27.50	3.0E-2	1.96
23	197.96	a	a	65.99	a	a
24	241.96	5.2E-3	0.25	65.99	1.8E-2	1.45
25	153.97	2.2E-3	0.15	54.99	1.6E-2	0.85
26	197.96	2.4E-3	0.13	54.99	1.2E-2	0.76
27	170.47	1.5E-6	0.27	54.99	3.9E-3	1.50
28	208.96	2.0E-7	0.05	52.24	4.6E-3	0.26
29	258.45	5.7E-3	0.19	54.99	2.0E-2	1.22
30	203.46	7.0E-3	0.26	62.32	2.4E-2	1.51

[a]No data available.

of condensate in the off-gas line allows the absorption of N_2O_4 into H_2O by the two-phase reaction (8), and to a lesser extent by liquid-phase reaction (9). NO is formed directly in reaction (8), and indirectly by the nitrous acid decomposition reaction (10), which follows reaction (9) in series.

Increases in both NO and NO_2 concentrations were indicated at Sample Point 3 (Table 5). However, these increases in concentration are more than offset by decreases in the volumetric gas flow rate from condensation of water, such that the actual quantities of NO and NO_2 in the vapor have now decreased. Increases in the concentrations of HNO_2 and HNO_3 in the off-gas condensate were also observed. These results indicate the absorption of NO, NO_2, and N_2O_4 to form nitric and nitrous acids by the forward reactions (4) and (5). The slight decrease in NO concentration is the result of reaction (6), which is driven by the depletion of NO_2 by absorption. Formation of HNO_2 and HNO_3 by reactions (8), (9), and (10) is also likely. These reactions should occur primarily at the condenser tube surface, as water present in the off-gas at the dissolver outlet is removed before the gas reaches Sample Point 3. Concentrations of HNO_2 and HNO_3 in the condensate from the first condenser range from 0.0024 to 0.0137 M and from 0.15 to 0.96 M respectively (see Table 4).

Table 5. Sample Point 3 Data

Sample set	Analyzer %NO	Analyzer $\%NO_2$	Temp. (K)	Condensate rate (ml/min)	HNO_2(l) (M)	HNO_3(l) (M)
5	0.15	0.80	332.7	0	[a]	[a]
8	0.37	1.48	333.8	0.067	[a]	[a]
12	0.90	1.35	332.2	0.091	3.0E-7	1.35
15	0.38	0.67	333.2	0.150	1.1E-7	1.44
18	0.48	1.03	333.3	0.450	4.1E-7	2.36

[a]No data available.

There is considerable variation between relative NO concentrations recorded at Sample Point 4 (Table 6) and those measured at Sample Point 3. No distinct trend is discernable.

Table 6. Sample Point 4 Data

Sample set	%NO	$\%NO_2$	Temp. (K)
5	0.48	0.42	294.3
7	0.55	0.70	293.7
11	0.33	0.58	297.6
13	0.30	0.37	298.1
15	0.25	0.25	297.7
17	0.55	0.58	294.0
19	0.51	0.53	297.4
21	0.50	0.65	293.7

However, all recorded NO_2 measurements at Sample Point 4 indicate a significant decrease in concentration from Sample Point 3. Significant increases in the HNO_2 (from 0.0035 to 0.0585 M) and HNO_3 (from 0.76 to 3.34 M) concentrations in the condensate from the second condenser were observed (see Table 4). These results point toward occurrence of NO_2 and N_2O_4 depletion by absorption. It is likely that some of the removal is due to the gas phase reactions (4) and (5). However, given the large quantity of water removed from the off-gas in the first condenser, it is more likely that the decrease in NO_2 concentration is the result of wetted-wall-effect absorption inside the condenser tubes by reactions (8) and (9).

Significant decreases in the quantities of NO and NO_2 present were observed at Sample Point 5, the DOG scrubber vapor outlet (Table 7). The reductions in NO concentration are

Table 7. Sample Point 5 Data

Sample set	%NO	$\%NO_2$	Temp. (K)
4	0.23	0	293.8
6	0.18	0	292.4
8	0.25	0.15	294.2
10	0.02	0.27	294.2
12	0.33	0.30	294.2
14	0.18	0.28	293.6
16	0.23	0	294.1
18	0	0.33	294.1
20	0.18	0	293.8
22	0.15	0	294.0

somewhat inconsistent, ranging from nearly 50% to nearly zero. The data seem to indicate a correlation between higher NO removal efficiencies and higher concentrations of NO_2 in the scrubber effluent gas. This finding indicates the occurrence of NO oxidation according to reaction (6). The large decrease in NO_2 concentration coupled with a consistent upward trend in nitric acid concentration in the recirculated scrubber solution is the result of NO_2 absorption. The favored mechanisms for absorption are reaction (8), which occurs in conjunction with the N_2O_4-NO_2 equilibrium relationship, and reaction (9), because of the relatively low operating temperature of the scrubber (293 K). A lack of accumulation of nitrous acid in the scrubbing solution is evidence of the decomposition of the acid by reaction (10).

Unfortunately, continuous recording of the NO and NO_x readings from the infrared analyzers was not performed during Phase 4. Therefore, the damping observed at Sample Point 1 in Phase 3 cannot be verified.

Iodine Behavior. Iodine samples, both those analyzed by gamma activity levels and by wet chemical techniques, were analyzed for total iodine. Therefore, no observations regarding the presence of specific iodine hydrolysis products can be derived. Note also that the condensates collected at Sample Points 1, 2, and 3 during NO_2 sampling were the result of drying the gas before entry into the infrared NO_x analyzers. Neither the analyzers nor the sample gas condensers were operated during iodine sampling because of potential corrosion problems.

Concentration values for vapor-phase iodine at Sample Point 2 were inconsistent with the values at Sample Points 1 and 3 (Table 8). Specifically, the concentration values at Sample

Table 8. Gas-phase Iodine Concentrations, Sample Points 1 - 3

	Sample Point 1		Sample Point 2		Sample Point 3	
Sample set	Iodine (mol/L)	Gas flow (LPM)	Iodine (mol/L)	Gas flow (LPM)	Iodine (mol/L)	Gas flow (LPM)
23	[a]	971	[a]	937	[a]	576
24	2.31E-6	1060	6379E-8	992	2.81E-6	569
25	3.19E-6	953	1.57E-6	911	3.41E-6	626
26	1.18E-6	934	7.00E-7	911	1.59E-6	561
27	1.52E-6	1104	9.74E-7	1094	2.06E-6	785
28	1.77E-4	1233	1.45E-4	1176	2.34E-4	789
29	2.29E-6	1241	1.03E-7	1251	3.69E-6	789
30	2.09E-6	1142	1.25E-6	1163	2.70E-6	787

[a] No data recorded.

Point 2 yield quantities significantly lower than quantities at either Sample Point 1 (upstream) or Sample Point 3 (downstream). Since there is no mechanism for the reappearance of iodine between Sample Points 2 and 3, the boundaries for the front-end iodine balance were established at Sample Points 1 and 3. Gas-phase concentration values at Sample Points 4, 5, and 6 were generally more consistent than data collected at Sample Points 1, 2, and 3 (Table 9).

Table 9. Gas-phase Iodine Concentrations, Sample Points 4 - 6

Sample set	Sample Point 4 Iodine (mol/L)	Sample Point 4 Gas flow (LPM)	Point 5 Iodine (mol/L)	Point 6 Iodine (mol/L)	Gas flow[a] (LPM)
23	[b]	460	[b]	[b]	407
24	2.30E-6	452	1.96E-6	1.47E-6	400
25	2.04E-6	527	1.32E-6	6.51E-7	465
26	9.46E-7	460	9.36E-7	7.94E-7	407
27	1.21E-6	684	1.63E-6	9.89E-7	600
28	1.78E-4	671	1.05E-4	4.10E-5	607
29	1.91E-6	687	1.41E-6	1.48E-6	603
30	1.54E-6	675	1.43E-6	1.27E-6	592

[a] Flow rates were approximately equal at Points 5 and 6.
[b] No data recorded.

Material balances performed across the first condenser typically close within 20%. Lack of closure is most likely from inconsistencies in the collection of gas samples. More importantly, the analysis of condensate from the first condenser, both by chemical and radiological techniques, detected little or no iodine in 5 of 7 samples (Table 10). In the other two sample

Table 10. Liquid-phase Iodine Concentrations in the DOG system

Sample set	First condenser I (mol/L)	Second condenser I (mol/L)	Scrubber bottoms tank I (mol/L)	Acid concentrator I (mol/L)
23	[a]	[a]	[a]	[a]
24	0	6.70E-4	3.31E-4	0
25	0	2.76E-4	0	0
26	0	1.97E-4	9.46E-5	0
27	0	1.18E-4	0	1.18E-4
28	1.58E-4	1.58E-4	0	1.18E-4
29	0	1.97E-4	0	2.36E-4
30	3.94E-5	1.18E-4	0	1.97E-4

[a] No data recorded.

sets, the amounts of iodine recycled to the dissolver in the liquid from the first condenser were 0.015 and 0.337% of the input iodine.

Material balances around the second condenser generally accounted for only 60% of the input iodine. However, the concentrations of iodine in the condensate from this condenser were consistently on the order of 10^{-4} mol I/min. At the nominal 55 ml/min condensation rate observed during this Phase of testing (Phase 4), the percentage of iodine removed from the vapor stream in the second condenser ranged from 0.31 to 1.80%.

Samples from the NO_x scrubbing system indicate that approximately 2% of the iodine fed to the dissolver was returned in the acid recycle stream (see Table 10). Inexplicably, the concentrations of iodine in the acid concentrator tank were higher than those in the scrubber bottoms tank in about half of the sample sets.

Observed liquid-vapor distribution coefficients for iodine at the first condenser were typically near zero. Distribution coefficients at the second condenser were typically of the order 10^2. The data from the second condenser correspond closely with distribution coefficients calculated using Eq. (1). The data from the second condenser are also consistent with distribution coefficients determined by the simultaneous solution of the equations

$$[HIO] = (K_{19}[I_2]_{aq})/([H^+][I^-]) \quad , \tag{11}$$

$$[H_2OI^+] = (K_{20}[I_2]_{aq})/([I^-]) \quad , \tag{12}$$

$$[I_3^-] = K_{18}[I_2]_{aq}[I^-], \tag{13}$$

$$[I^-] = \{(K_{20}[I_2]_{aq})/(1 + K_{18}[I_2]_{aq}) + (K_{19}[I_2]_{aq})/([H^+](1 + K_{18}[I_2]_{aq}))\}^{1/2} \quad , \tag{14}$$

$$[I_2]_{aq,TOT} = [I_2]_{aq} + 0.5([HIO] + [H_2OI^+] + [I^-] + 3[I_3^-]) \quad , \tag{15}$$

$$D = [I_2]_{aq,TOT}/[I_2]_g \quad , \tag{16}$$

as given by Parsly.[6] These expressions are derived from the equilibrium relationships for the water-iodine system

$$I_2(g) \text{ <--> } I_2(aq) \quad , \tag{17}$$

$$I_2(aq) + I^- \text{ <--> } I_3^- \quad , \tag{18}$$

$$I_2(aq) + H_2O \text{ <--> } H^+ + I^- + HIO \quad , \tag{19}$$

$$I_2(aq) + H_2O \text{ <--> } H_2OI^+ \; I^- \quad . \tag{20}$$

The liquid-phase concentration of iodine from the first condenser condensate was somewhat lower than predicted by Eq. (1). Given the relatively low concentrations of iodine in both phases, this deviation is not unreasonable.

IODOX System Performance. The results of material balances around the IODOX scrubbing column indicate poor performance in iodine removal. Removal efficiencies ranged from 11 to 61%, with the highest efficiency achieved at the highest inlet iodine concentration. The average removal efficiency over 6 sample sets was 34%. The resultant decontamination factor of 1.5 is several orders of magnitude less than values reported by Groenier.[7] Potential errors in the collection of the gas sample are not likely to have produced such a large deviation. The brevity of the IODOX operation period made it unlikely that steady state was approached. For this reason, and the extreme variation between experimental results reported herein and data obtained in earlier work, the validity of the IODOX results obtained in this study is suspect.

5. Conclusions

Results from the evaluation of NO_x behavior in all phases of testing are consistent with reaction mechanisms previously reported. The most significant findings are the strong interactions among the various chemical species in the NO_x-HNO_x-H_2O system at the relatively low component concentrations present in the dissolver off-gas system.

The results of this study indicate that the extent of incidental NO_x treatment, which occurs in the off-gas condensers, is determined by the composition of the entering gas. However, over the range of feed conditions studied, the combined NO and NO_2 concentration in the effluent from the second condenser never exceeded 1.5%.

The NO_x scrubber was surprisingly efficient, given the low concentrations of NO and NO_2 present in the feed gas stream. However, given the low concentrations of NO and NO_2 present downstream of the off-gas condensers, no further NO_x removal is required to effectively remove iodine.

One limitation in the NO_x study was the ability to simultaneously monitor only one sample point at each of the two anaylzer stations. This severely restricts the ability to perform real-time mass balances across individual sections of the off-gas system.

The results from iodine testing in the DOG system confirmed the results obtained previously by Lewis and Jubin.[1] Negligible amounts of iodine were recycled to the dissolver in the condensate from the first condenser. A minimal amount (2% maximum) of the iodine in the dissolver off-gas was recycled in the condensate from the second condenser. In addition, a 2% recycle of iodine from the NO_x scrubbing system was observed. It may be concluded that, at steady state, the double overhead condenser/NO_x scrubber configuration results in a 4 to 5% recycle of iodine and 99% recycle of the nitric acid and water present in the dissolver off-gas. Removal of the NO_x scrubbing system from the loop reduces the amount of iodine recycle to 2 to 3%.

The iodine removal efficiencies observed in the IODOX system were considerably lower than anticipated. Unfortunately, the short duration of the test produced insufficient data to determine the cause of the poor scrubber performance.

REFERENCES

1. B. E. Lewis and R. T. Jubin, "Demonstration of the Iodine and NO_x Removal Systems in the Oak Ridge National Laboratory Integrated Equipment Test Facility," Proceedings of the 19th DOE/NRC Nuclear Air Cleaning Conference, Seattle, Wash., August 1986.

2. R. M. Counce, The Scrubbing of Gaseous Nitrogen Oxides in Packed Towers, ORNL-5675, Martin Marietta Energy Systems, Oak Ridge Natl. Lab., November 1980.

3. J. H. Smith, J. Am. Chem. Soc., 69, 1742 (1947).

4. J. B. Lefers et al., Chem. Engr. Sci. **35**, 145 (1980).

5. J. Carberry, Chem. Engr. Sci. **9**, 189 (1959).

6. L. F. Parsly, Design Considerations of Reactor Containment Spray Systems-Part IV, Calculation of Iodine - Water Partition Coefficients, ORNL - TM - 2412, Part IV, Martin Marietta Energy Systems, Oak Ridge Natl. Lab., January 1970.

7. W. S. Groenier, An Engineering Evaluation of the IODEX Process: Removal of Iodine from Air Using a Nitric Acid Scrub in a Packed Column, ORNL-TM-4125, Martin Marietta Energy Systems, Oak Ridge Natl. Lab., August 1973.

Development of a Process for Adsorptive Separation of Kr-85 from the Off-Gas of Nuclear Facilities

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Abstract

A separation process was developed for the retention of krypton noble gas from gaseous effluents of nuclear installations. In the flow sheet only adsorption beds are applied. The most efficient process scheme is adsorption at temperatures as low as -160°C and thereafter gas chromatography during temperature increase up to +150°C. The industrial scale application of this new gas separation process - called ACHAT process - was demonstrated by the operation of a pilot plant facility with 13 m^3/h STP gas flow. Purity of the separated krypton can be > 99 vol % and the decontamination factor > 1000.

I. Introduction

At the Research Centre Jülich (KFA) a process has been developed for the separation of radioactive krypton from the gaseous effluents of nuclear installations. This new process is called ACHAT process (adsorptive chromatographic separation of krypton on activated charcoal at low temperatures). In recent years further work on developing the process has been carried out in cooperation with the Power Generation Group KWU of Siemens AG and verified by the construction and operation of a pilot plant.

II. Principles of the ACHAT Process and Comparison of Process Variations

The concept of the adsorptive chromatographic krypton separation process is based on a combination of adsorptive pollutant separation from the off-gas flow and subsequent fractionation of the adsorbed gases by gas chromatography (1).

Based on this idea, several process variations have been experimentally studied during recent years. They included variations of the temperature and pressure change techniques and their combination with gas chromatography.

Four process variations have been basically studied which significantly differ by their operating conditions both in the adsorption and the regeneration phases.

Variation 1: loading at normal pressure and room temperature
Variation 2: loading at 10 bar and room temperature
Variation 3: loading at 10 bar with moderate cooling (approx. -70°C)
Variation 4: loading at normal pressure with low-temperature cooling (approx. -160°C).

The first two process variations require a two-stage krypton separation. In the first stage, krypton is adsorptively enriched in the off-gas for being chromatographically separated in the second stage.

On the other hand, if the adsorber columns are cooled to low temperatures during loading, the krypton separation process can be combined in one process step.

The process variations studied are characterized and compared in Table 1 using the example of a facility for a throughput of 100 m^3 gas/h.

Variation	Operating Conditions			Plant Data for a Facility with 100 m^3/h Throughput			
	Loading [pressure, temp.]	Separation [purge gas, pressure, temp.]		Adsorber Volume [m^3]	He Purge Gas [m^3/h]	Decont. Factor	Kr Purity [vol %]
		1st Stage	2nd Stage				
1	1 bar RT	N_2 50 mbar + 180°C	He 1 bar RT	10.8	62	440	87
2	10 bar RT	N_2 50 mbar + 180°C	He 1 bar RT	5	11	400	90
3	10 bar - 150°C	He 1 bar + 120°C		2	30	800	90
4	10 bar - 150°C	He 1 bar + 120°C		0.32	8	≥ 1000	≥ 90

Table 1: Survey of Different Process Variations and Plant Data

The development work (2) eventually led to a process according to which the krypton is first adsorbed from the off-gas by means of activated charcoal in two adsorber columns cooled to low temperatures (minimum -160°C). In the regeneration phase, these adsorber columns are then heated according to a defined temperature scheme (maximum +150°C) and purged with a small amount of helium which causes the adsorbed gases, such as nitrogen, krypton, xenon and others, to slowly flow through the activated charcoal bed. Since the gases have a different tendency to adsorption, they migrate through the column at a different rate so that a multi-stage mass separation process results. The parameters (such as column combination, temperature

characteristic, helium flow etc.) are matched in such a way that only pure krypton leaves the last column with the helium flow for a defined time interval. This helium-krypton mixture can then be passed again through a krypton storage cylinder cooled to low temperatures and filled with activated charcoal or zeolite. The krypton is thus directly separated in the storage cylinder in a single operating process, while the helium is purified and leaves the storage cylinder for being re-used.

III. Laboratory Experiments

Laboratory Facility

Experiments have been carried out with a laboratory facility in order to establish optimal process parameters for the ACHAT process and measure the gas separation achievable with this process. Figure 1 shows the flow principle of the facility. It basically consists of the following components:

- LN_2 inlet heat exchanger
- first activated charcoal column
- LN_2 intermediate heat exchanger
- second activated charcoal column
- krypton collection cylinder.

The two activated charcoal columns are of identical design. A cross section of one column is shown in Figure 2. The column basically consists of a ring-shaped charcoal bed with an annulus thickness of 20 mm. The inner and outer walls of the annulus can be cooled by a cooling medium (LN_2 + N_2 vapour) and heated by an electric resistance heater, which is integrated into both walls. This column design was chosen in order to have a relatively small temperature gradient in the cross section of the column, i. e. perpendicular to the gas flow. This is essential for achieving a relatively undisturbed gas separation process by gas chromatography during the column heat-up sequence.

Results of Laboratory Experiments

Most of the experiments were carried out with N_2 or dry air containing about 100 vpm Kr and 1000 vpm Xe. For this gas composition the optimum process parameters for the facility with two columns of 5 l charcoal were as follows:

For the Adsorption Sequence:

gas flow rate:	6.7 m^3/h STP
adsorption time:	3 h
temperature column 1:	-130°C
temperature column 2:	-160°C

For the Regeneration Sequence:

He flow rate:	6 l/min STP
regeneration time:	3.5 h
max. temperature:	150°C
purity of Kr product:	85 vol %
decontamination factor:	≥ 1000

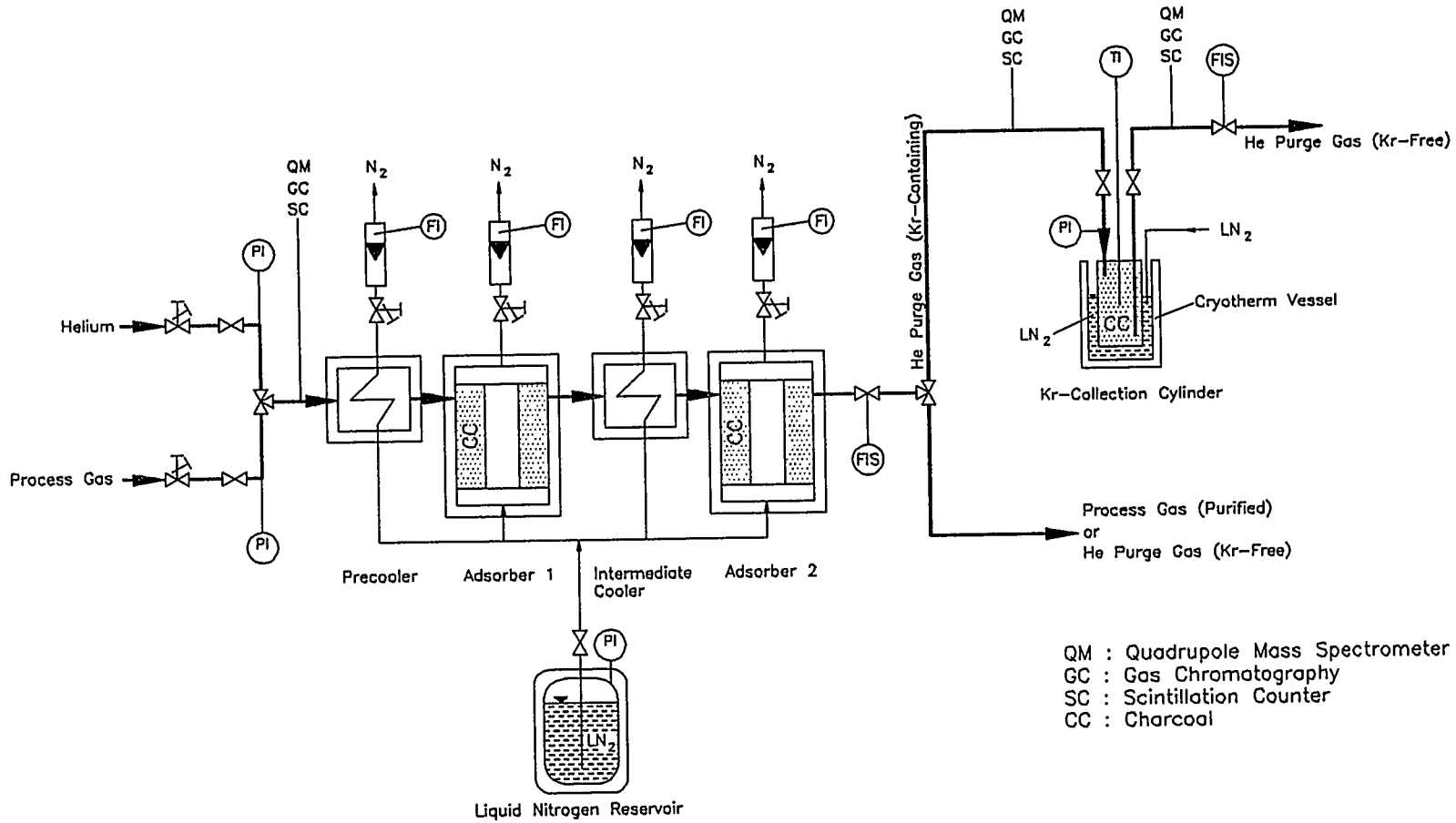

Figure 1: Flow Diagram of the Laboratory Facility for Kr-Separation with Integrated Kr-Collection Cylinder

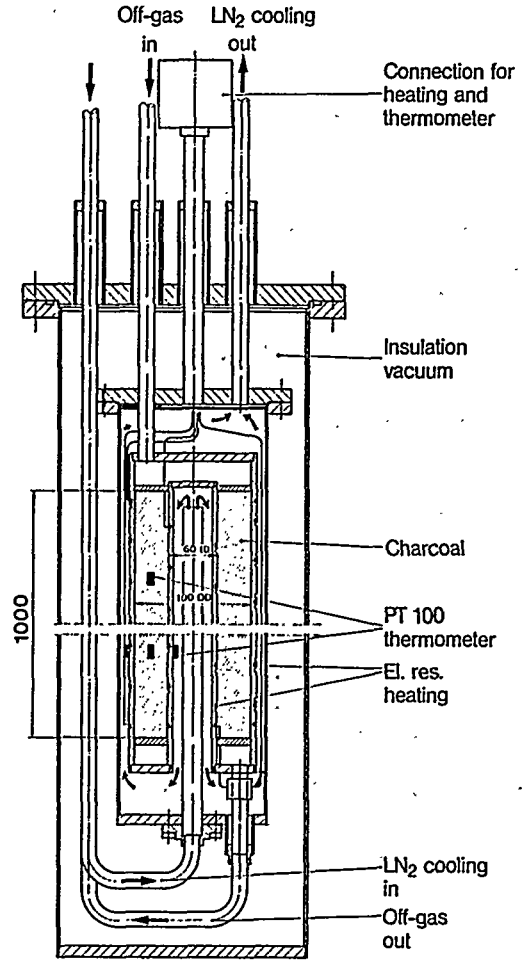

Figure 2: Kr Separation Column for Laboratory Facility

Besides the experiments with typical dissolver off-gas, experiments with dry ambient air (containing 1.14 vpm Kr) and with off-gas (containing 0.28 vpm radioactive Kr) from an electric bulb fabrication line have also been accomplished. These experiments were intended to demonstrate the application of the ACHAT process for very low krypton concentrations and its high and reliable decontamination capacity.

The results with the latter off-gas are illustrated in more detail by the operation diagram in Figure 3. The off-gas from the electric bulb fabrication line had the following composition:

80	vol %	Ar
16	vol %	N_2
4	vol %	O_2
300	vpm	CO_2
40	vpm	H_2
0.28	vpm	Kr with approx. 5 % Kr-85

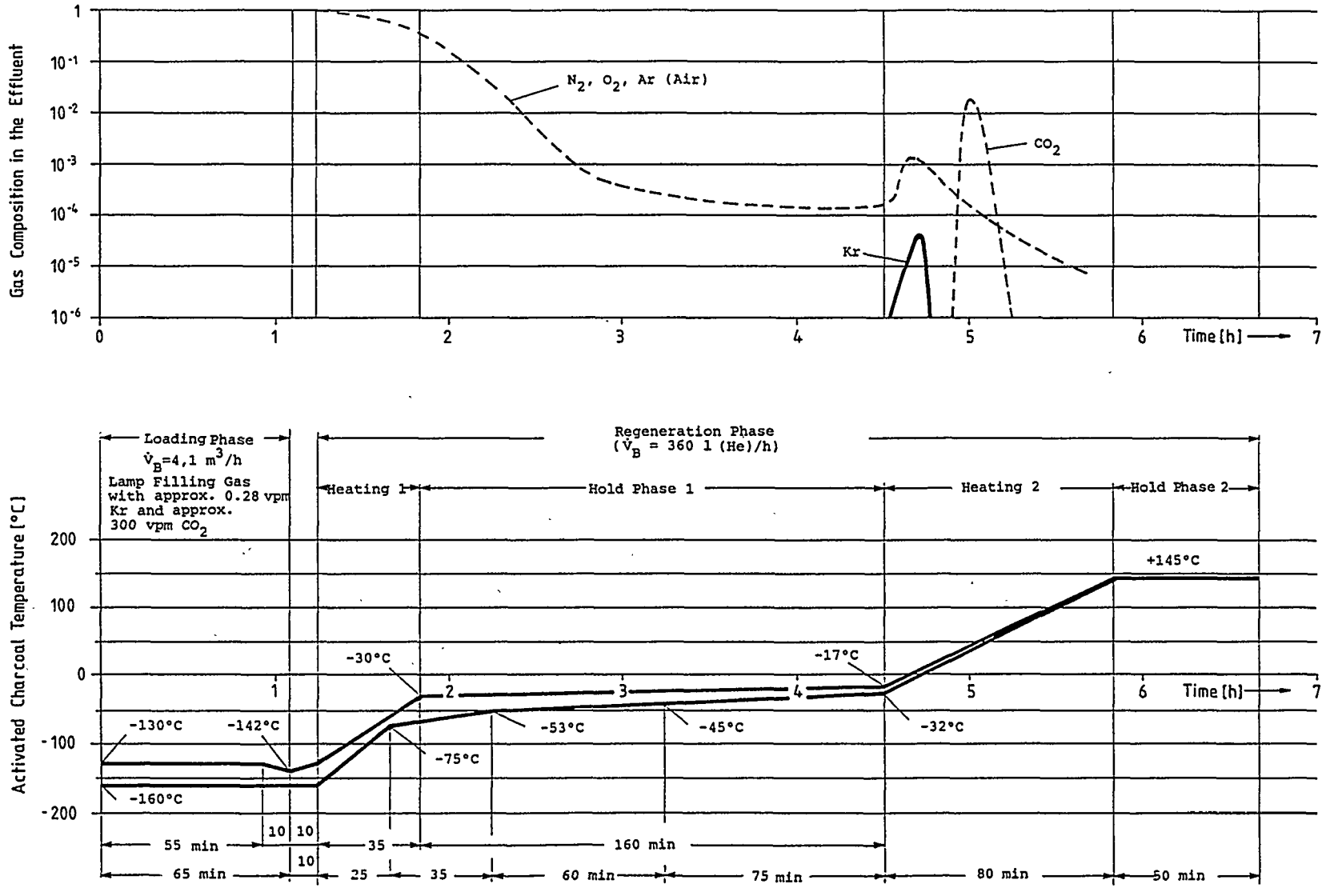

Figure 3: Operation Diagram of Kr-Separation from a Special Lamp Filling Gas using the Laboratory Facility

About 4.4 m^3 STP of this gas with a flow rate of 4.1 m^3/h was routed through the two adsorbers being at a temperature of -130°C and -160°C, respectively.
The separated krypton fraction contained approx. 1 vol % krypton which gives an enrichment factor of about 30,000. For nearly 100 % Kr, this krypton fraction would have to be processed twice in the facility. With an uncertainty of about 10 % practically all krypton from the off-gas was collected in the krypton fraction.

IV. Pilot Plant Experiments

ACHAT Pilot Plant

A dissolver for spent nuclear fuel might have an off-gas flow in the range of 100 m^3/h STP gas. Flows of this order have been used for the basic layout of a full-scale adsorber system (3). On this basis the Siemens AG developed an adsorber system (4), which fulfilled stringent requirements with regard to both process engineering and the quality of equipment, and built a pilot plant which had all the main features of a full-scale industrial facility. Figure 4 shows the pilot plant. It was designed as a package unit with a size of height x width x depth = 3.45 x 2.0 x 1.6 m.

The plant was dimensioned for operation with a 6-hour loading phase at a gas flow rate of 13.4 m^3/h STP. The gas would contain krypton and xenon concentrations of 100 vpm and 1000 vpm, respectively.

The most import part of the plant are the two adsorber units or columns. Figure 5 shows a cross section of an adsorber unit. Each unit comprises an adsorber tube bundle which is filled with the same activated charcoal as used in the laboratory experiments. Besides this a helium/liquid nitrogen heat exchanger, an electric heater and a helium fan are also integrated into the unit. The He-LN_2 heat exchanger and electric heater make up the heating and cooling unit which is arranged vertically beside the adsorber tube bundle; the helium fan is mounted above the tube bundle (5). As a result of this thermodynamic and hydraulic design of the adsorption system it was possible to ensure a very homogeneous temperature field in the area of the adsorber tube bundle during the loading and regeneration phases. This is an important prerequisite for uniform adsorption and desorption in the activated charcoal tubes.

The following technical data are of importance:

- helium circulation rate for heat exchange: 4000 h^{-1}
- pressure loss ratio bundle/total: ≥ 0.95
- bundle temperature difference, vertically: ≤ 1°C
- bundle temperature difference, horizontally: ≤ 4°C

A further important feature of the adsorbers is that they have been designed according to the same principle as used in floating head heat exchangers. All the internals are fixed and supported in the vessel cover; this facilitates thermal expansion in the operation temperature range between -200°C and +150°C. The plant also features two coolers, one upstream of adsorber unit 1 and one between the two adsorber units. Both are operated using liquid nitrogen. Figure 6 shows the schematic of the ACHAT pilot plant.

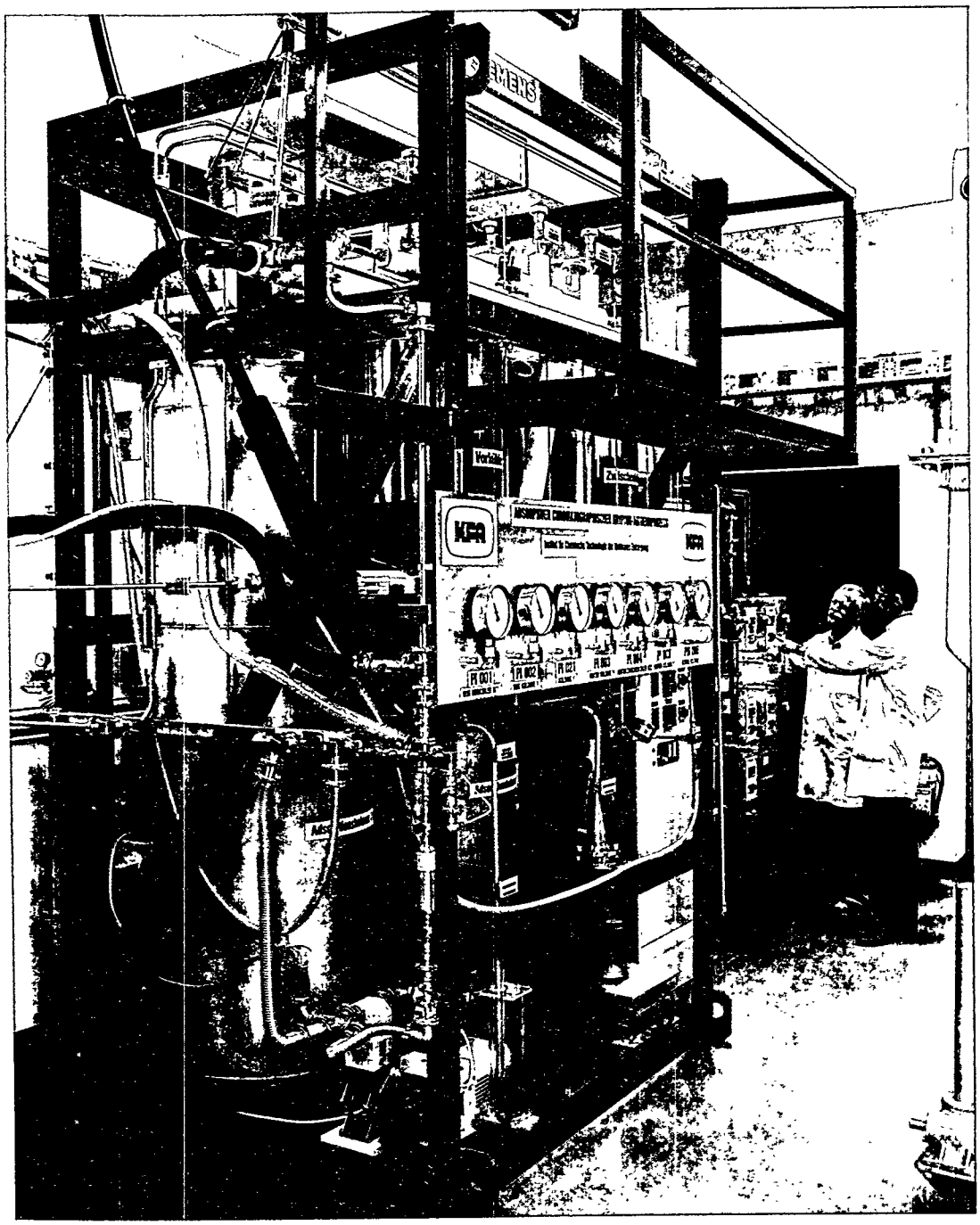

Figure 4: ACHAT Pilot Plant at Jülich Research Centre

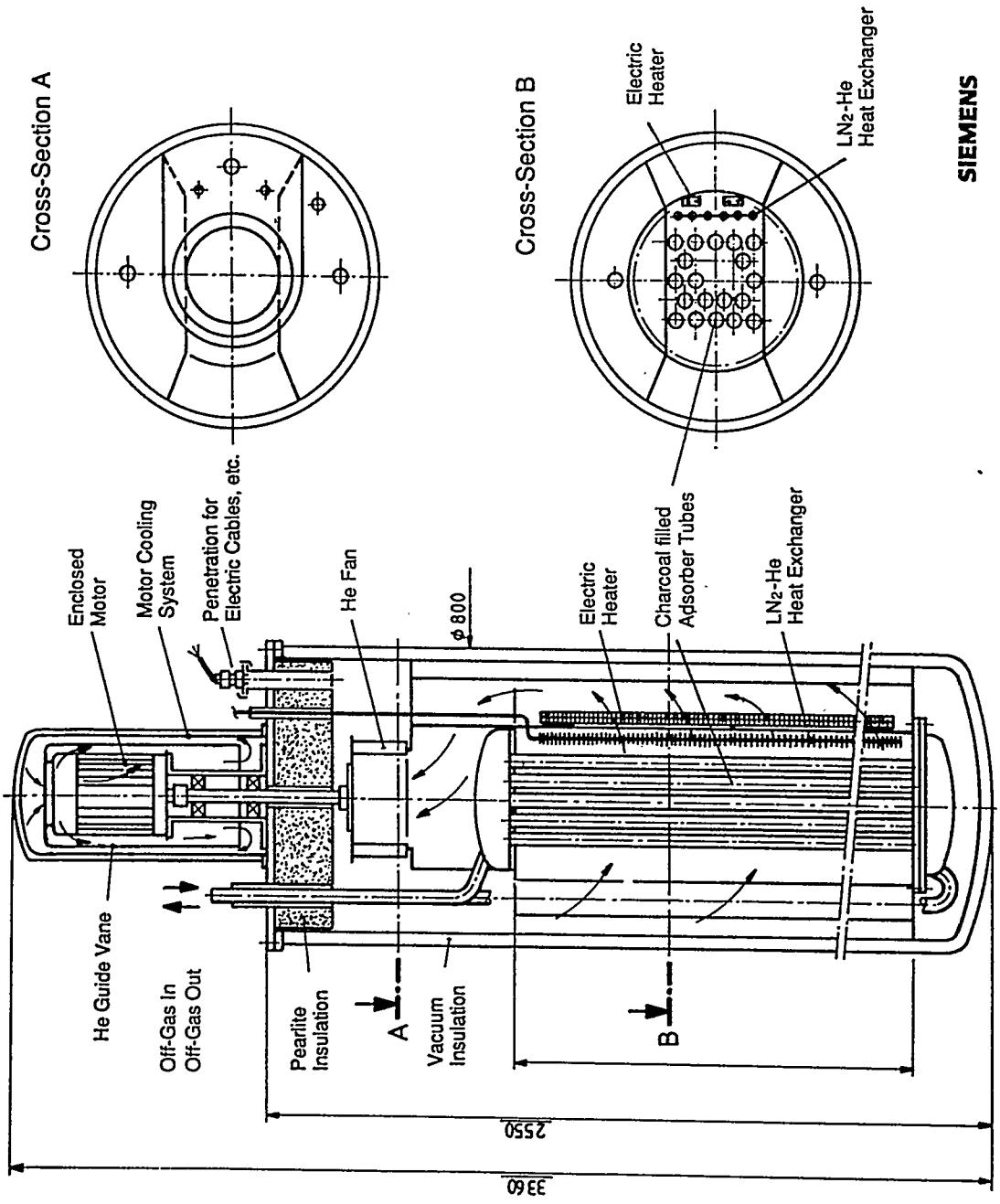

Figure 5: Adsorber Unit of the ACHAT Plant

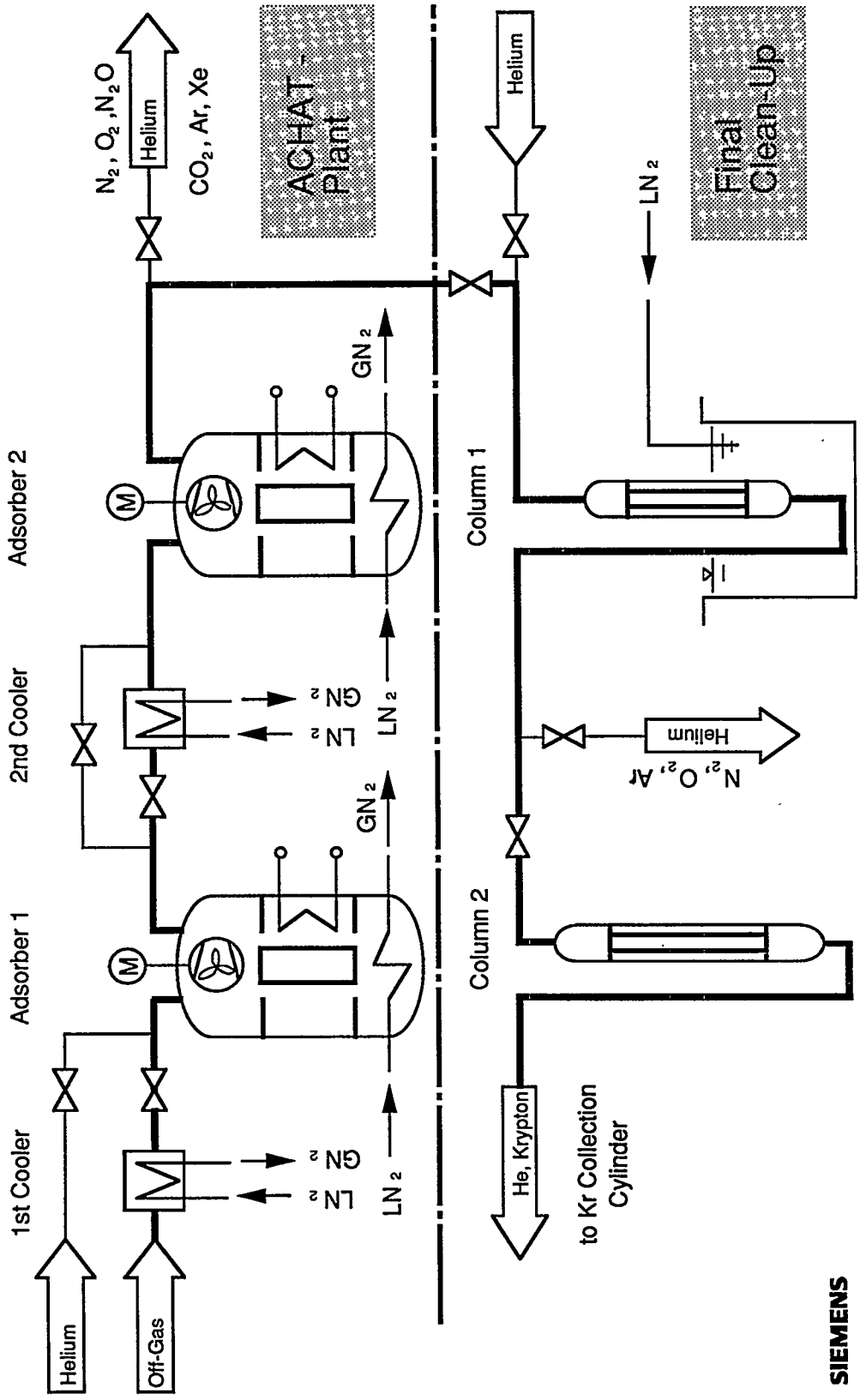

Figure 6: Schematic of the ACHAT Plant Connected to Final Clean-Up System

Operating Results

The tests were performed taking into account the conditions to be expected at a reprocessing plant. To simulate a typical dissolver off-gas, a gas was used containing synthetic air (N_2, O_2), 0.1 vol % of xenon and 0.01 vol % of krypton as well as representative amounts of argon (Ar), carbon dioxide (CO_2) and dinitrogen oxide (N_2O). The adsorbers were loaded over a period of 6 h in accordance with the typical duration of krypton release from a spent fuel dissolver.

The results from the test runs of the pilot plant were consistent with those obtained using an analytical model and confirmed the feasibility of the process on an industrial scale.

As an example the test results for the fractionation of 80 m^3 STP of simulated dissolver off-gas are given. For this experimental run the following parameters are of importance:

For the Adsorption Sequence:

gas flow rate:	13.4 m^3/h STP
gas composition:	synthetic air with 1 vol % Ar, 100 vpm Kr, 1000 vpm Xe, 300 vpm CO_2 and 200 vpm N_2O
loading time:	6 h
overall press. drop during adsorption:	200 mbar
temperature column 1:	-130°C
temperature column 2:	-160°C

For the Regeneration Sequence:

He flow rate:	1.2 m^3/h STP
regeneration time:	6.5 h
max. temperature:	120°C

More detailed information of the run and its result are given by the operation diagram shown in Figure 7. The diagram shows the temperatures of adsorption unit 1 and adsorption unit 2 for the adsorption and regeneration sequences as well as the gas composition of the effluent of adsorption unit 2. As can be seen from the diagram, amounts of N_2, O_2, Ar and CO_2 are contained in the Kr fraction, while N_2O and Xe are completely separated from the Kr. For this case, the purity of the Kr would be 84 vol %.

In order to achieve an increased Kr purity, the effluent from adsorption unit 2 was routed through a final clean-up unit, consisting of two small activated charcoal columns (35 mm ID x 0.5 m length and 35 mm ID x 2.5 m length) as shown in the flow sheet of Figure 6. The impure krypton was first collected in column 1 which was kept at LN_2 temperature. While column 1 was then heated up to room temperature, a secondary gas chromatography process was activated by flushing both columns with a small helium flow. The Kr purity was thus increased to > 99.5 vol %. The decontamination factor for the run was determined as ≥ 1600.

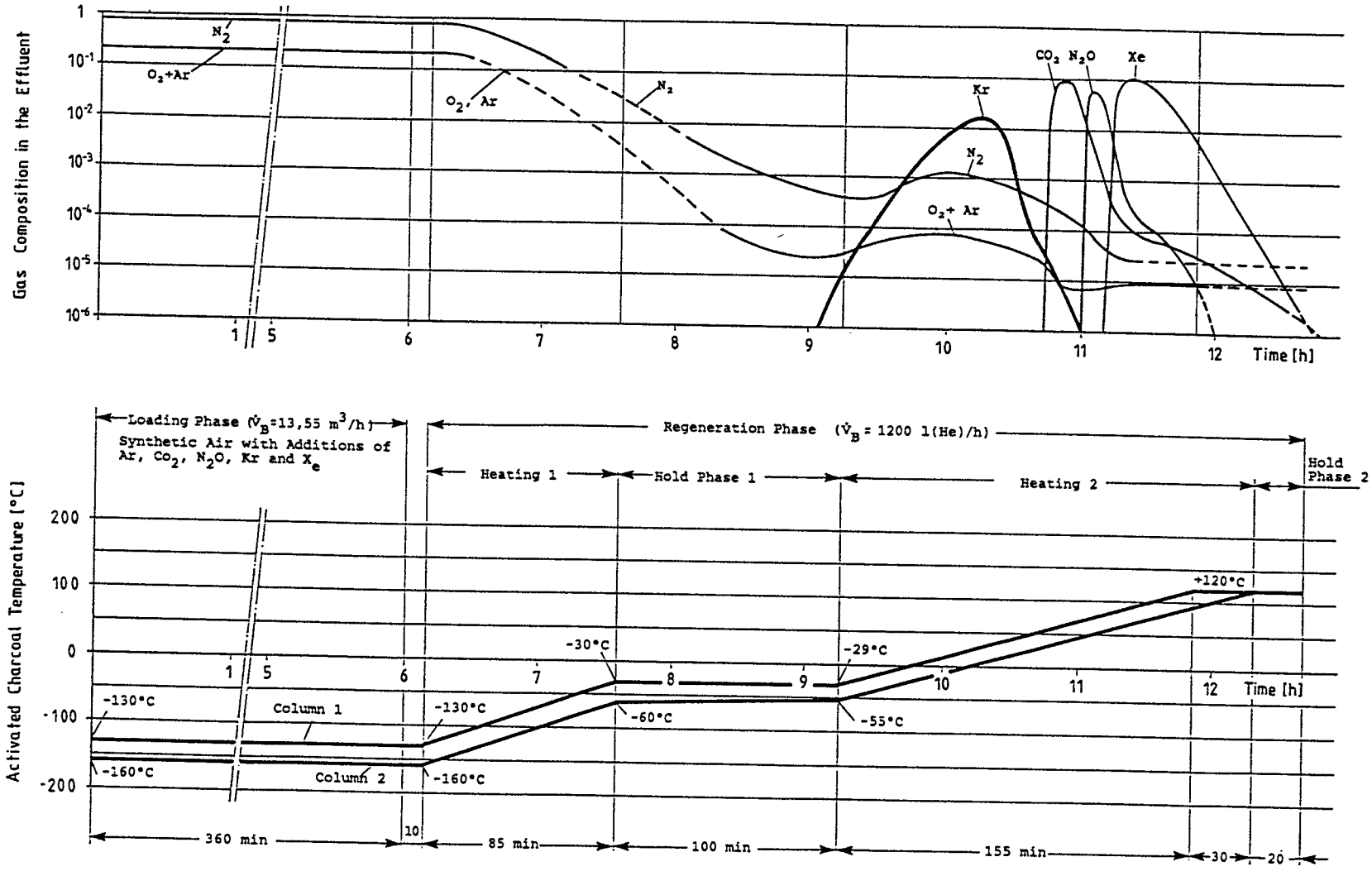

Figure 7: Typical Operation Diagram of Kr-Separation with the ACHAT Pilot Plant

Industrial-Scale Application and Extrapolation of Test Results

During the development of the pilot plant, scale-up considerations were taken into account by the appropriate choice and sizing of the adsorber tube bundle geometry and consideration was given to uniform gas introduction into the tube bundle and flow velocity through the empty tubes. A scale-up to industrial plant size for off-gas flow rates of e.g. 300 m^3/h STP is therefore possible by simply increasing the number of adsorber tubes. No other changes are necessary.

An adsorber of this kind, having a throughput of 300 m^3/h STP, can be easily built with a ring-shaped adsorber tube bundle. By integrating all the functional elements in the adsorber vessel, all the advantages of the pilot plant can be maintained when scaling up to industrial plant size. Such an adsorber unit would have a diameter of approximately 2.2 metres and be about 3.5 metres high (5). Figure 8 shows the design of a full-scale adsorber unit.

The results obtained from operating the ACHAT pilot plant are directly applicable to this industrial-scale plant because all the relevant sorption and thermodynamic design parameters are identical.

V. Conclusions

The ACHAT process is based on the adsorptive chromatographic separation of krypton using activated charcoal at very low temperatures. This new process is therefore entirely based on the physical separation involving the adsorption of contaminants by a solid. This principle is characterized by its reliability and simplicity and has been tried and tested on numerous occasions.

The following advantages of the ACHAT separation process deserve special mention:

- A krypton separation plant designed for continuous operation basically consists of a total of four sorption columns with heat exchangers and valves. These are easy to control and do not require a complex closed-loop control system.
- The process is insensitive to off-gas flow and off-gas composition fluctuations.
- Compared to other processes, the inventory of krypton-85 in the plant is small.
- Nowhere in the plant does concentrated krypton occur. The only concentrated krypton (almost 100 %) is collected in the storage cylinder.
- The capital and operating costs are very attractive compared to other processes. This has been verified by different detailed studies.

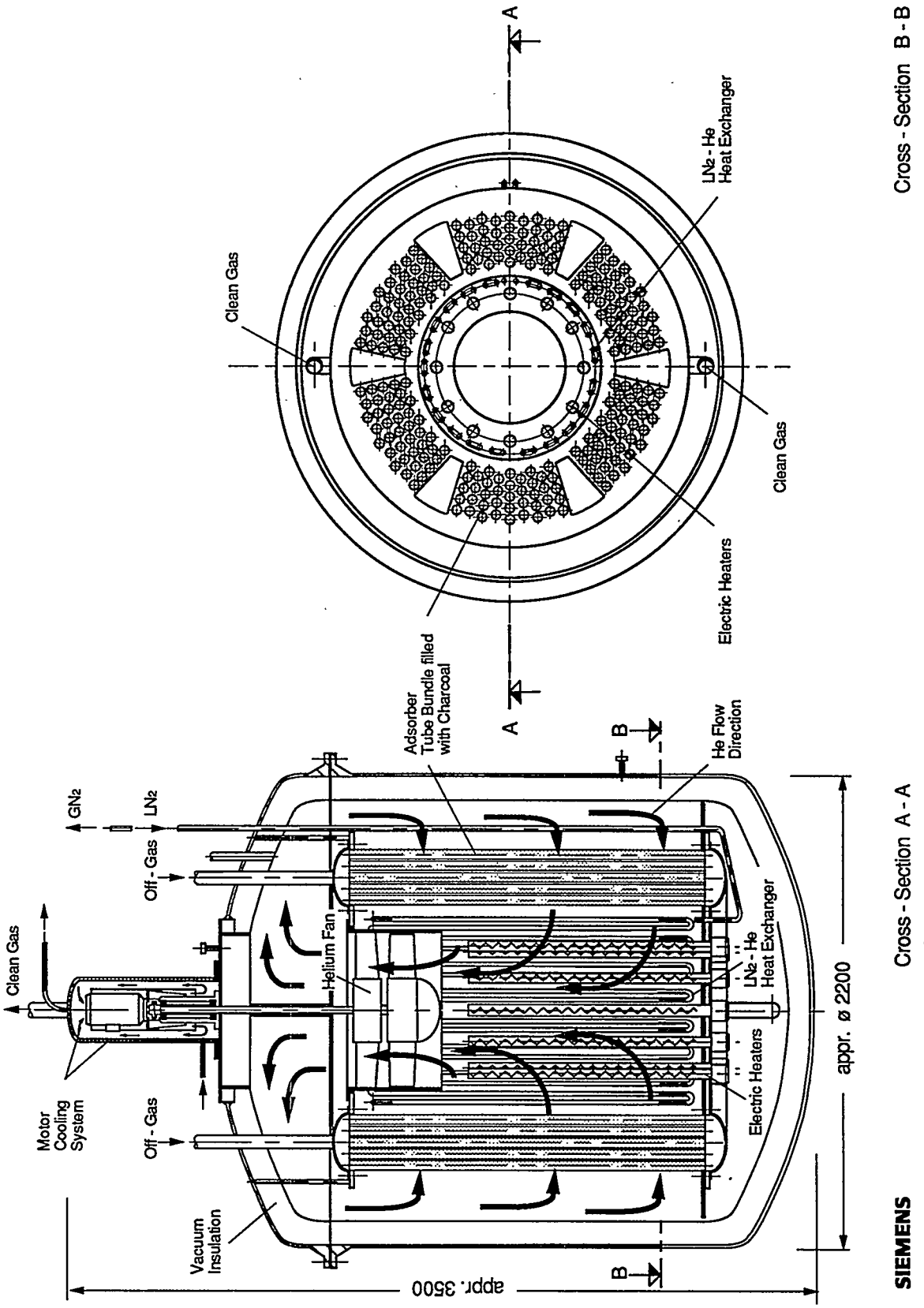

Figure 8: Adsorber Unit Designed for a Flow Rate of 300 m^3/h STP

Acknowledgments

We would like to thank Mr W. Frommelt, Mr K. Hein, Mrs G. Linse and Mr S. Dix for their experimental assistance.

References

(1) Ringel, H. "Process for the Separation of Krypton from a Radioactive Waste Gas Mixture and Arrangement for Implementing the Process"
US Patent Number 4,654,056 (March 31, 1987)

(2) Meßler, M. "Entwicklung eines Verfahrens zur adsorptiven Abtrennung von Krypton-85"
Report: Jül-1988 (1985)

(3) Ringel, H.D.; Printz, R. "Pilot Plant Development for Adsorptive Krypton Separation from Dissolver Off-Gas" 19 th DOE/NRC Nuclear Air Cleaning Conference, CONF-860820, Vol 1, 566-577 (1987)

(4) Eckardt, B.; Burbach, T. "Adsorption Device for Gas Separation"
US Patent Number 4,881,958 (Nov. 21, 1989)

(5) Burbach, T. "Adsorptive Chromatographic Separation of Krypton at Low Temperatures, Process and Plant Description"
Siemens, US 331, (Dec. 6, 1989)

DISCUSSION

HANDYSIDE: How large are the cost advantages of your process? Would you comment on the safety of the process relative to other processes>

RINGEL: Different studies showed that our processes has capital and operational cost advantages. The difference in operational cost is mainly related to power needs. For a 300 m^3/h plant, power needs are 0.6 and 1.2 MW for the ACHAT process and the low temperature rectification process, respectively. The capital cost for one large adsorption unit is of the order of 1 to 2 million DM.

HANDYSIDE: Would you care to comment on the safety of this process compared to the safety of other processes?

RINGEL: Concerning safety, we showed by experiments that a power supply or cooling failure resulted in only a very delayed and only partial Kr release from the columns. In this case, we would have the smallest exposure to plant personal and the environment in respect to other processes. We have no O_3 problem, because O_3 will decompose and will not build up on the charcoal. The only thing is, that charcoal is burnable (and can not be replaced by molecular sieve in the ACHAT process), but we have no ignition conditions throughout the process.

VIKIS: Is it possible to scale-up this technology to levels applicable for removal of Kr-85 during a containment vent following a severe reactor accident?

RINGEL: Scale-up to off-gas flows following a reactor accident will be too expensive. A better way might be to build some kind of a large adsorptive delay bed and then use the ACHAT process for handling the regeneration gas from such a delay bed.

CLOSING COMMENTS OF SESSION CO-CHAIRMAN JUBIN

This morning we head six papers addressing various aspects of chemical off-gas processing. The first three papers addressed recent work in Germany concerning iodine releases from a large nuclear fuel reprocessing facility. This work examined the behavior of iodine in both a large pilot plants and the WAK facility. A key point mentioned in these papers is the need to achieve a high degree of iodine evolution from the dissolver solution to minimize the down stream iodine treatment and retention requirements. Dr. Furrer of Kernforschungszentrum Karlsruhe described tests in which >99.5% of the iodine is routinely evolved from the dissolver into the dissolver off-gas system during pilot plant tests. If these results can be translated in to plant performance, this certainly takes a great deal of the burden off of the vessel off-gas and waste treatment off-gas systems. A detailed description of the treatment of the MAW stream was presented where it was pointed out that 50% of the iodine remained with the MAW evaporator bottoms. The work reported in these papers also pointed out the difficulty in obtaining meaningful samples from the wet off-gas streams near the dissolver.

The paper on iodine removal using caustic scrubbing focused on the microscopic analysis of the process rather than the macroscopic approaches that have been reported at may previous conferences. This work provides an insight into the kinetics and mass transfer behavior through the use of classic chemical engineering analysis and should prove useful in gaining a better understanding of the processes involved and possibly lead to a more optimal scrubber design.

The U.S. paper in this session also addressed the behavior of iodine and NO_x in a large pilot plant facility. This work showed very little iodine recycle back to the dissolver by the dual condenser system. This experimental finding is supported by limited iodine distribution data for the iodine - air - nitric acid system. The very small iodine decontamination factors for the IODOX system are somewhat disturbing and as yet unexplained. I hope further work will be done with this system to obtain an understanding into the observed behavior. It was also somewhat surprising that very little iodine was accumulated in the NO_x scrubber bottoms tank.

The final paper of this session discussed a novel approach to the recovery of krypton from an off-gas stream using new low temperature absorption onto charcoal beds. This provides a high krypton decontamination factor and a high purity krypton product. This type of innovative work appears to offer a significant cost saving and potentially some improvement in safety over previously developed krypton recover processes.

As a final note, I would ask that all of you consider that this area of chemical off-gas treatment be broadened to include studies into the final waste forms. As you all are aware, the recovery step is only the beginning of a complete treatment system. I hope you will consider this area for the next air cleaning conference in two years.

SESSION 5

PANEL: HOW TO USE N510 TESTING METHODS AND ACCEPTANCE CRITERIA FOR AIR TREATMENT SYSTEMS NOT CONSTRUCTED ACCORDING TO N509

Tuesday:	August 14, 1990
Moderator:	R. R. Weidler
Panel Members:	James R. Edwards
	John W. Jacox
	Richard D. Porco
	Peter Olson
	Philip Roberson

OPENING COMMENTS OF PANEL MODERATOR WEIDLER

THE DIFFICULTY OF TESTING NON-509 SYSTEMS TO THE N510 STANDARD
J. R. Edwards

A REVIEW OF THE HISTORY, REVISIONS AND INTERRELATIONSHIPS OF ANSI/ASME N509 AND N510, 1989 EDITIONS
J. W. Jacox

TESTING OF NON-ANSI/ASME N509 SYSTEMS TO ANSI/ASME N510 REQUIREMENTS
R. D. Porco

TESTING OF NON-N509 FILTER SYSTEMS - AUDIT EXPERIENCES AT MCGUIRE NUCLEAR STATION
P. W. Roberson

COMMENTS ON TESTING OF NON-N509 FILTER SYSTEMS - AUDIT EXPERIENCES AT MCGUIRE NUCLEAR STATION
P. Olson

PANEL DISCUSSION

CLOSING COMMENTS OF PANEL MODERATOR WEIDLER

OPENING COMMENTS OF PANEL MODERATOR WEIDLER

I am Ray Weidler from the Duke Power Company in Charlotte, NC and I manage the McGuire Mechanical Systems Group which is involved strictly with McGuire Nuclear Station. I have been involved with nuclear plant HVAC and nuclear air treatment systems for the past 18 years. I am also Vice-Chairman of the ASME Committee on Nuclear Air and Gas Treatment. With me today is an extremely distinguished panel consisting of Jim Edwards, who is President of Charcoal Service Corporation, Bath, NC; Rich Porco, Consultant from Pittsburgh, PA; Jack Jacox, Consultant from Columbus, OH and Pete Olson, who is filling in for Duke Power's Phil Roberson who couldn't be with us. Pete is with Flakt Company and is located at Comanche Peak.

The purpose of this Session is to hear and discuss the problems that we have with the testing of the non-N509 systems to the N510 requirements. Also, I definitely want to involve the audience and hear your problems. We sincerely want to have your input. What we hope to do with this information is go back to the Committee on Nuclear Air and Gas Treatment and determine if we need to incorporate changes in either N509, N510, or the Code, AG-1.

THE DIFFICULTY OF TESTING NON-N509 SYSTEMS TO THE N510 STANDARD

James R. Edwards, President
Charcoal Service Corporation
Bath, North Carolina

Utilities, and others, are under constant pressure to upgrade their test methods to include current technology. It is not unusual to see requirements that an existing non-N509 system be tested to the N510 testing standard.

These requirements are apparently made without knowledge that referring to N510 as the test standard imposes the very rigorous N509 design criteria on a filter system. The N510 standard is specific that only an N509 system can be testable to N510. However, this cooperation between the standards is ignored by many who arbitrarily impose N510 as the testing standard.

In order to adequately address the problem, the test engineer should proceed as follows for each non-N509 system which is to be tested to the N510 criteria:

1. Review all former test procedures and results, including the original acceptance test procedures and results.

2. Perform a visual inspection of each non-N509 system to be tested.

3. Direct the installation of requisite test ports in the non-N509 system.

4. Rewrite the existing acceptance test procedure to encompass the N510 requirements. The acceptance test procedure should be specific to each system to be tested.

5. Perform the acceptance test procedures. Document all failures and successes.

6. Report the results.

7. In addition to Number 6., write an itemized summary of each acceptance criteria which cannot be met according to N510 requirements, with reasons why it cannot be met.

8. Recommend changes that will make the system testable to N510.

Examples of problems which may be encountered during an N510 acceptance test are as follows:

1. Poor Airflow Distribution

2. Poor Air/Aerosol Uniformity

3. Insufficient access to perform the acceptance tests

As a practical matter, utilities which have non-N509 systems authorized by existing tech specs are usually unwilling to spend the money necessary to make major system upgrades. In many cases, the costs of the upgrades to make the systems testable to N510 would be considerable, And, frankly, some of the upgrades would be impractical. These upgrades may include any or all of the following significant changes, or more:

1. Longer approach plenums and duct runs

2. More space between components

3. The addition of baffles and turning vanes

4. Better access to the interior of the system

5. Replacement of internal components, such as HEPA racks

6. The addition of qualified instruments and controls

7. Seismic bracing

It should be noted that any non-N509 system can be in-place tested using N510 as guidance.

In conclusion, mere reference to N510 as the testing standard for a non-N509 system is usually impractical. It is likely that the test engineer will have difficulty achieving the N510 acceptance criteria. In order to meet the N510 criteria, the non-N509 system will need alterations. However, any non-N509 system can be tested per the spirit of N510.

A REVIEW OF THE HISTORY, REVISIONS AND INTERRELATIONSHIPS OF ANSI/ASME N509 AND N510, 1989 EDITIONS

John W. Jacox

President

JACOX ASSOCIATES

1990

ABSTRACT

This paper includes three sections on N509 and N510. The rational for changes made including the public workshops held by the COMMITTEE ON NUCLEAR AIR AND GAS TREATMENT, the actual changes made and their impact on users and vendors and the close interrelationship of these standards and the critical need to reference the correct editions for both fabrication and testing.

The first section is a short history of the standards, the workshops, formal questions received by ASME, discussions with regulatory personnel on the committee and at the workshops and experience of the committee members.

The second subject covers the major changes, the intent of the changes, when a revision is NOT a change but simply stronger wording to improve compliance, and the reasons for the changes. Note that none of these standards are AUTOMATICALLY required to be used but must be invoked by the owners on themselves through the Plant License or by NRC requirements.

The third subject is the critical requirement to be aware that each set of editions was written to be used as a complimentary set of documents. The problems and advantages of using a different testing edition applied to a fabrication edition will be discussed.

The new ASME Code "AG-1" is briefly mentioned as the follow-on replacement and expansion document to N509 and N510.

INTRODUCTION

The standards ANSI/ASME N509 "Nuclear Power Plant Air-Cleaning Units and Components" (1) and N510 "Testing Of Nuclear Air Treatment Systems" (2) are referenced in all operating United States nuclear power plants since United States Nuclear Regulatory Commission (USNRC) Regulatory Guides 1.52 "Design, Testing, and Maintenance Criteria For Atmospheric Cleanup Air Filtration and Adsorption Units of Light-Water-Cooled Nuclear Power Plants" (3) and 1.140 "Design, Testing, and Maintenance For Normal Ventilation Exhaust System Air Filtration and Adsorption Units of Light-Water-Cooled Nuclear Power Plants" (4) both refer to the these standards. Further both standards are well recognized and used throughout the world. This wide spread use makes a full understanding of them very important and requires that they be written as clearly and unambiguously as possible. This need for clarity is the main reason for the three editions rather than major changes in content or intent. Whether or not they are sufficiently clear or if additional formal guidance is needed is the theme of this panel.

HISTORY

The original 1975 and 1976 editions were written by an hoc group of experts who started to meet in the early 1970s. Their efforts came to fruition with the initial publications in 1975 and 1976. Unfortunately since this was an ad hoc effort the group dissolved upon publication and there was no mechanism for questions to be formally answered or revisions to be made. The American Society of Mechanical Engineers (ASME) sponsored a meeting in January of 1976 to form what became the ASME "Committee On Nuclear Air and Gas Treatment" (CONAGT) which has written the later editions. With these standards under the jurisdiction of a formal ASME Code Committee continuity and the ability to formally respond to technical inquires was ensured. This has worked very well in the nearly fifteen years of CONAGT's existence. In addition to the serious need for technical guidance in the area of Nuclear Air Treatment Systems (NATS) the early reference of these documents by the USNRC made them de facto requirements for the US nuclear power industry. They have also been adopted both formally, and as guidance documents by other government agencies, including DOE and other entities around the world.

Given that these were new standards that enjoyed immediate wide spread use, questions of intent, interpretation and a few actual errors quickly became evident. None of these problems were serious but the strict regulation of the nuclear industry requires even minor points be formally addressed. CONAGT has been able to provide such formal assistance with the ASME inquiry procedures. Work on revising standards started in 1976. Additionally the CONAGT committee was given a mandate to produce a totally new Code (AG-1) that covered a greatly expanded and somewhat different scope over that N509 and N510. Considerable in-depth background on CONAGT and the revisions of N509 and N510 is given in a number of past Conference papers. (5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16)

Since it was obvious that a second revision of these standards was necessary to correct some errors, improve the wording, improve the description of instruments to be used for the testing and generally upgrade them, work was started as soon as CONAGT was formed. No change in intent of the documents was planned nor executed. The changes were all, hopefully, simply improved guidance to accomplish the original objectives in essentially the same ways.

In this period there were two major and innovative developments by CONAGT that are worthy of note. The first was the initiation of public workshops to obtain input to allow the standards to be able to meet user needs in the best possible manner. The first was held in February 1985 for one day in Los Angeles. It was so successful that the second held in April 1985 in Charlotte, NC was expanded to a two day affair. (12, 17) These Workshops were extremely valuable in obtaining details of how to upgrade the standards to make them more convenient to use and most importantly how to reduce ambiguous areas of requirements. The second area is the investigation (8, 9) of the then existing ASTM standard (D3803-79) for radioiodine testing. The initial CONAGT investigation brought to light serious questions as to the accuracy of results obtained using this standard. More specific controlled data was needed so CONAGT oversaw a roundrobin test series with an international group of laboratories participating. These results proved there was indeed a problem and the NRC, through a subcontractor, conducted two more roundrobins as well as a number of open meetings. Eventually the standard was revised based on this data and has greatly improved the testing of carbon for radioiodine efficiency.

MAJOR CHANGES IN THE 89 EDITIONS

The revisions in 509/510-89 have been discussed in previous papers (13, 14, 15) but since when these previous papers were written the standards were still only drafts it is worth briefly noting their relevance again. The changes will only be listed to highlight them without going into detail since the comments should be generally sufficient to make the changes understandable.

MAJOR REVISIONS IN N509-89

1. General update and improve wording based on users comments.
2. Improve consistency of required documentation (Design, QA, Installation and Test Reports).
3. Expand reference and definition Sections.
4. Improve and clarify the definition of "pressures".
5. Reference AG-1 "Code On Nuclear Air and Gas Treatment" (19) sections where applicable.
6. Better coordinate with N510-89.
7. Provide testing parameters for N510 Acceptance Testing or state which parameters must be provided by the designer.
8. Upgrade proscriptive requirements for system to be testable per N510.
9. Add test manifold design and testing recommendations/requirements.
10. Generally expand rationales and directions for design.
11. Upgrade adsorbent requirements.
12. Upgrade requirements for adsorbent fire protection and eliminate requirements for automatic actuation of water deluge systems.
13. Clarify adsorbent cooling re water deluge section.
14. Add specific requirement that all valves and dampers to be leak tested must be exercised through their full operating range before a valid leak test may be performed.
15. Improve and upgrade the detailed requirements for housing doors and their hinges and door dogs.
16. Eliminate the requirement for a "down stream" HEPA Bank.

MAJOR REVISIONS IN N510-89

1. General update and improve wording based on user comments.
2. Reduce verbiage.
3. Update to allow current instrument technology.
4. Better coordinate with N509-89.
5. Improve Visual Check List.
6. Improve reporting/documentation requirements.
7. Add 10 year Housing Leak Test requirements.
8. Make Frame Test optional.
9. Incorporate mandatory manifold requirements - eliminate Multiple Sampling as an acceptable test method.
10. Add specific test of bypass duct dampers.
11. Add specific test of entire system.
12. Address ASTM D3803-79 test problems.
13. Incorporate industry standards for duct testing (SMACNA, AABC, NEBB and AMCA).
14. Eliminate individual bank pressure drops and replace with total housing pressure drop for Air Flow Capacity Tests.

INTERDEPENDENCE OF THE TWO STANDARDS, N509 & N510

The importance of proper understanding and use of the interrelationship of these standards can not be over emphasized. They have been written to be used together as complimentary documents. Since the N509 fabrication document is usually used only at the design and fabrication stage and the N510 testing document is used throughout the life of the system there is a high probability that later versions of the test standard will be invoked for systems built to earlier editions. This is certainly acceptable, and usually helpful, IF care is taken to understand where conflicts will occur and correct for them. The edition of N509 is often changed when referring to the carbon specifications since carbon is purchased over the life of the systems. This can be helpful but also must be carefully considered. The most common error is to invoke a later edition of either document IN TOTAL rather than only the sections necessary for the specific update planned. This blanket approach will almost always cause problems if only in conflicting references in other documents such as Plant Technical Specification, the plant Final Safety Analysis Report (FSAR), various other plant License documents that contain system/component parameters, and Regulatory Guides. Obviously there is an open ended list of possible documents that could become problems but these are almost universal when edition references are changed without proper study, understanding and coordination.

For example if the carbon for a safety system (Regulatory Guide 1.52 system) is reordered it will be very helpful and probably more cost effective to order it by the N509-80 or AG-1 specifications. This is not a difficult change to implement but often the change is too broad and the edition of N509 is changed from -76 to -80 (or either edition to 89) across the board in the Plant Technical Specifications. Conflicts in other areas are, therefore, unavoidable since the physical systems will not meet the letter of the later editions of N509. Even to change the test methods for the carbon and keeping the same performance criteria can be complex since the test parameters in the ASTM tests referenced in N509-80 (or 89) are different than those in the old RDT test standards referenced in N509-76.

Probably the most common change is to use the latest edition of N510 for testing. Since N510 has not changed the operational test intent there should be no problems BUT there often are. Even though the intent has not changed the wording has become increasingly proscriptive to require the complete Nuclear Air Treatment System be leak tested. This has made the 89 edition appear to add tests for Bypass Duct Dampers and the System test. These ARE NOT NEW and were always intended to be performed in the earlier editions. The wording clearly was not sufficiently clear given very poor historical compliance experience so the more proscriptive sections were added. Many seem to believe this is a major increase in the testing required by the N510-89 edition. It is not! Only the wording for prerequisites for leak testing the HEPA and adsorber banks has become more proscriptive. Another problem area is the qualification of the systems to allow technically correct DOP and Halide Leak Tests. Again it was felt very proscriptive language was necessary in the 89 editions. The Air Flow Capacity and Air/Aerosol Mixing tests have always been required, and are technically necessary, before a valid bank leak test could be performed but were often not performed. Lack of these prerequisites makes all later DOP and Halide leak tests suspect.

The new CODE AG-1 is being issued in parts with over one half currently published. The scope includes such new areas as Off Gas Systems and much broader testing of the total systems. The next generation of reactor orders should use AG-1 to everyone's benefit. As a point of interest Regulatory Guide 1.52 is being revised to, among other things, refer to current industry standards and hopefully eliminate existing conflicts.

In any case unless standards are read with some reasonable attention they can never be properly used or applied. As a sad example there is currently an RFQ on the street for a 150 cfm HEPA-Carbon-HEPA NATS. It has been issued by one of the major A/E firms with decades of nuclear power plant experience. The specification in the RFQ does not simply reference N509 and N510, both 1989, even though the RFQ states the NATS shall comply with them. Rather, the standards are rewritten along with sections from obsolete references and older editions. This blind "cut and paste" approach provides a specification requiring, among other things, 30X50 inch marine bulkhead doors, permanent access platforms for components over 6 feet from the housing floor and permanently installed ladders. This is for a system that has as inlet and outlet connection 4 inch diameter pipe and an over all cross section of less than 30" X 30". No amount of revisions to a standard or training courses can solve this type of problem. Codes and Standards are not substitutes for good judgement.

CONCLUSIONS

The major conclusions are that, the N509 and N510 standards content has evolved with increasingly detailed proscriptive content. This has been deemed necessary by the CONAGT Committee since the evidence from installed systems design, fabrication and testing proves that the standards have not been fully met in most cases. Whether the 89 editions will be successful in specifying sufficient detail for the system built, operated and tested to meet the writing groups intent remains to be seen.

The continued use of the later editions for testing and carbon purchase for systems built to earlier standards clearly requires much more careful investigation and understanding of the total impact when the referenced edition is changed.

Should ASME prepare new documents to provide guidance in the use of N509, N510 and AG-1? There are certainly different points of view as can been seen from this panel session. My personal feeling is that no formal ASME documents are needed, but more education is. Clearly these conferences are extremely valuable but certainly not alone sufficient. The "ASME Short Course", the Harvard "In Place Filter Testing Workshop" and various commercial courses such as offered by Nuclear Consulting Services, Inc. with a full sized SBGT NATS to use for hands on experience are all valuable resources and should be used by all involved in NATS technology. Only when those who work with this technology have both a basic theoretical AND practical understanding can technically excellent and cost effective results be obtained. It is the education, experience and good judgment of the Responsible Engineer on a project that will ultimately define if a NATS will be Codes and Standards and, more importantly,function safely, efficiently and cost effectively.

REFERENCES

1. ANSI/ASME N509 "Nuclear Power Plant Air-Cleaning Units And Components", 1976, 1980 and 1989 Editions, American Society of Mechanical Engineers, 345 East 47th Street, New York, NY 10017.

2. ANSI/ASME N510 "Testing Of Nuclear Air Treatment Systems", 1975, 1980 and 1989 Editions, American Society of Mechanical Engineers, 345 East 47th Street, New York, NY 10017.

3. Regulatory Guide 1.52 "Design, Testing, and Maintenance For Atmospheric Cleanup System Air Filtration and Adsorption Units of Light-Water-Cooled Nuclear Power Plants, Revision 2, March 1978, USNRC, Washington, DC 20556.

4. Regulatory Guide 1.140 "Design, Testing, and Maintenance Criteria For Normal Ventilation Exhaust System Air Filtration and Adsorption Units of Light-Water-Cooled Nuclear Power Plants, Revision 1, October 1979, USNRC, Washington, DC 20556.

5. "ANSI/N45-8 Nuclear Gas Treatment Systems Treatment Standards", F. D. Leckie & C. A. Thompson, 12th Conference, CONF-720823, US Government Printing Office, P.O. Box 37082, Washington, DC 20013-7082.

6. "CONAGT Progress In Nuclear Standards Development", J.F. Fish, 16th Conference, 1980, CONF-801308, US Government Printing Office, P.O. Box 37082, Washington, DC 20013-7082.

7. "Inplace Testing Of Non ANSI-N509 Designed Systems", W.R. Lightfoot, 18th Conference, CONF-840806, US Government Printing Office, P.O. Box 37082, Washington, DC 20013-7082.

8. "Results of CONAGT-Sponsored Nuclear-Grade Carbon Test Roundrobin", M.W.First, 18th Conference, CONF-840806, US Government Printing Office, P.O.Box 37082, Washington, DC 20013-7082.

9. "CONAGT's Nuclear Carbon Roundrobin Test Program", R.R.Bellamy, 18th Conference, CONF-840806, US Government Printing Office, P.O. Box 37082, Washington, DC 20013-7082.

10. "CONAGT's Place In ASME's Centennial Year", W.H.Miller, Jr., 18th Conference, CONF-840806, US Government Printing Office, P.O.Box 37082, Washington, DC 20013-7082.

11. "Conflicts and Misapplications of ANSI N509 N510 USNRC Regulatory Guides 1.52 and 1.140, J.L.Kovach, 19th Conference, CONF-860820, US Government Printing Office, P.O. Box 37082, Washington, DC 20013-7082.

12. "Report on the CONAGT Field Testing Workshops", D.M.Hubbard, 19th Conference, CONF-860820, US Government Printing Office, P.O. Box 37082, Washington, DC 22013-7082.

13. "Highlights of Proposed Changes To ANSI/ASME N509-80", S.C. Ornberg, 19th Conference, CONF-860820, US Government Printing Office, P.O.Box 37082, Washington, DC 20013-3802.

14. "Highlights of Proposed Changes to ANSI/ASME N510 Testing Of Nuclear Air-Treatment Systems", D.D.Whitney, 19th Conference, US Government Printing office, P.O.Box 37082, Washington, DC 20013-3802.

15. "Changes To ANSI/ASME N510", A.T.Bishara, 20th Conference, CONF-880822, US Government Printing Office, P.O.Box 37082, Washington, DC 20013-3802.

16. "Backfitting ANSI N510 for Non-ANSI N509 Designed Systems, A Utility Responsibility, S.G.Banton, 20th Conference, CONF-880822, US Government Printing Office, P.O. Box 37082, Washington, DC 20013-3802.

17. "Committee On Nuclear Air And Gas Treatment Workshops: Successful User Feedback Method", J.W.Jacox, MECHANICAL ENGINEERING, March 1987, American Society of Mechanical Engineers, 345 East 47th Street, New York, NY 10017.

18. "Standard Method for Testing of Nuclear-Grade Gas-Phase Absorbents", D3803-89 (79), American Society for Testing and Materials, 1916 Race Street, Philadelphia, PA 19103-1187.

19. "Code On Nuclear Air and Gas Treatment" AG-1-88, American Society of Mechanical Engineers, 345 East 47th Street, New York, NY 10017.

TESTING OF NON-ANSI/ASME N509 SYSTEMS TO ANSI/ASME N510 REQUIREMENTS

Richard D. Porco

We are here today to discuss problems in testing systems that do not conform to N509 requirements. After designing hundreds of filter systems for both nuclear and non-nuclear applications, I may have a slightly different perspective than my colleagues.

Let me begin by asking a few questions. Are there any good systems out there that are not N509? I should hope so. Are there any good systems out there that are not N510 testable? Again the answer is yes.

When confronted with a system to be tested, one should ask "Does the system meet the design specification? Have there been qualification and/or acceptance tests previously performed?" How had the designers planned to meet the performance spec. If valid testing was never performed and there is no testing information in the design spec or supplier information, then I suggest modifications to your purchasing and testing practices.

Is the system testable? Forgetting about N510 for a minute, can you perform valid, repeatable tests on the unit to verify performance? Looking through the menu of tests in Table 9.1 of 509 and Table I of 510, over 90% of problems can be attributed to one characteristic - poor air flow distribution. This is also one that is most difficult to correct. In addition to affecting the air-aerosol mixing uniformity during testing, poor air flow distribution will affect performance by increasing media velocity through a portion of the HEPA filter(s) possibly causing increased penetration of sub-micron particles as the media velocity increases above 5 ft/min. The carbon adsorption could also be affected since retention time in a portion of the bed could be reduced.

If the unit was designed with poor flow distribution, the reason was probably insufficient space requirements to accommodate the unit. If this is the case, the design must provide for flow straighteners and baffles.

Assuming you have a testable system, there are various techniques and methods available to prove a system is functional. If you cannot test to the letter of N510, meet the intent. Look for modifications that produce valid and repeatable results.

There are test methods available, both new and existing technology, to prove the reliability and performance of your unit. Scanners, laser particle counters, continuous monitors, etc. can be employed; but, N510 or any other test programs cannot compensate for a poorly designed system.

Should these systems be addressed in the code? Can all the variations be adequately addressed by a code? That is what we are here to determine.

TESTING OF NON-N509 FILTER SYSTEMS- AUDIT EXPERIENCES AT McGUIRE NUCLEAR STATION

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Abstract

Site specific examples of auditor identified discrepancies and station responses in the testing of nuclear air filtration systems per ANSI/ASME N510 requirements are reviewed.

Introduction

This paper is written from the perspective of an "end-user." My background includes 10 years as a testing staff engineer at a commercial nuclear power plant. Nine of those years have been directly involved with ventilation and filtration systems, which has led to my assignment as Nuclear Air Filtration System Expert. I apply the label of end-user to myself in two respects - both in the sense of the operation and testing of the ventilation/filtration systems, and in the interpreting and application of test codes, standards and reg guides to these systems.

The intent of this presentation is to stimulate discussion on the topic of non-N509 system testing. Concrete answers regarding the extent of applicability of N510 to non-N509 systems will NOT be attempted, as those types of interpretations and judgements tend to be plant and system specific. What will be provided is a partial summary of the efforts of one nuclear station to address the concerns of various auditors as regards nuclear air cleaning.

N510 Non-Conformance and Other Auditor Issues

McGuire Background

McGuire Nuclear Station is a two 1200 MW unit, Westinghouse PWR. Specific system design for the station began in 1970, with ground-breaking in 1971. Preoperational testing of ventilation and filtration systems was performed from the late '70s thru 1981. Unit 1 fuel loading occurred in February 1981, with initial criticality in August of that year.

The design timeframe of McGuire precedes the development and adoption of nuclear air filtration design standards (N509-1976), and the preoperational testing timeframes overlap the development, adoption and revision periods of the test standard (N510-1975 and 1980). Nuclear air filtration preoperational testing was primarily performed by vendors, with some support from station personnel.

As part of the broad approach to the topic of audit experiences, the term 'auditors' is being used to address a wide range of personnel and agencies. This includes Quality Assurance, Self-Initiated Technical Audits [SITAs], NRC (resident site inspectors, regional effluents inspectors, independent contractors) and others.

Auditor Issues and Station Responses

One issue that arose from an auditor's review of test data for a specific system's air-aerosol mixing uniformity test regarded acceptance criteria. The test procedure stated that the allowable variation for individual concentrations was ±10% from the average concentration, with N510-1975 cited as the source. The 1975 standard states (after making reference in the preceding steps to the measuring of individual concentrations and calculation of the average for both upstream and downstream sample planes), "9.4.7 If the maximum and minimum readings at the plane passing through the upstream sample point differ by more than 10%, relocate the injection port, or...". The auditor's view was that the context of the previous steps (referring to the average) did not apply, and a correct reading of the standard was that the max and min values could not differ from each other by more than 10%.

By consulting CONAGT committee members and by quoting the N510-1980 revision, which states, "9.4.6 If the maximum and minimum readings differ by more than ±20% of the average reading, relocate the injection port, or...", the station convinced the auditor that the original 1975 criteria had referred to "% difference from average", and the later revision had doubled the allowable range, not quadrupled it.

Auditor scrutiny of preoperational tests also revealed that in some cases, the acceptance criteria and conditional test applicability from the 1980 revision had been used in conjunction with the 1975 standard. For some systems, this mixing of standards resulted in NO preoperational test being performed for air-aerosol uniformity in the HEPA downstream sample plane (NOT required by N510-1980) and also in NO adsorber air distribution test being performed (NOT required by N510-1975). For two single HEPA filter trains, the air distribution test was not performed (since N510-1975 requires comparing the center face velocity of each HEPA), and the air-aerosol uniformity test was not performed (as specifically allowed by N510-1980, Step 9.4.4).

The station and auditors agreed that the mixing of requirements from the two N510 versions could result in a less thorough demonstration of filter system performance. The station committed to comply as fully as possible with the comprehensive requirements of N510-1980.

During efforts to comply with N510-1980, the air distribution profile for the deep-bed adsorber bank of a 54,000 cfm system was measured. A junction box inside the filter housing located near the last inlet slot caused a local disturbance, affecting two readings. Velocity uniformity results based on slot-to-slot averages met the ±20% criteria, but the comparison of the 200 individual velocities did not.

Justification of the acceptability of the adsorber velocity profile was provided by Design Engineering. The small number of points outside the tolerance range, their relative position in the extreme corners of the adsorber bank face and the fact that further equalization of flows would occur within the adsorber slots assuming an approximately uniform carbon density were all used to support the judgement that adsorber flow distribution was acceptable.

Auditor issues have also dealt with the physical limitations of performing tests of filter trains that were built without the N509 specified "access between banks of components." In one specific case, efforts to perform comprehensive testing per N510-1980 were not successful due to the back-to-back construction of the prefilter, HEPA, adsorber bed, and welded discharge damper. Since no access between any individual components was available, a compromise test using N510-1975 methods and conditions, with 1980 acceptance criteria was approved.

Recommendations

The consequences of auditor concerns can usually be decreased by the use of three steps: Consultation - talk to the experts in the industry to ensure your test practices and interpretations are supported. Communication - inform the NRC resident inspector of interpretations and judgement calls. Documentation - ensure that technical judgements regarding test applicability, acceptability, etc. along with sufficient supporting information are recorded and easily retrievable.

Obviously, in an ideal situation, the above steps will have been applied prior to an issue becoming an "audit concern." In those instances where this is not the case, resolving the audit concern will probably involve the same process - which leaves you in better shape for the scrutiny of your next auditor.

COMMENTS ON TESTING OF NON-N509 FILTER SYSTEMS-AUDIT EXPERIENCES AT McGUIRE NUCLEAR STATION

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I do not have a problem discussing Phil Roberson's paper after reading it because I totally agree with his final recommendations. We will get to them in a minute.

The history of the McGuire Nuclear Station is the design began in the early 1970s prior to the existence of N509 or N510. Pre-operational testing of the station was completed in 1981. Looking at the history of nuclear standards, we are aware that N509 and N510 were issued during the 1975-1976 time period and revised in 1980. Regulatory Guide 1.52 was issued in the middle of this period and then we had the TMI situation. First of all, you had sort through all the various standards and their revisions to determine what you had to do. Based on my own experience I can imagine that at the Station they were saying, "Test this system and meet all codes and standards." A recent review of their test program revealed some potential deficiencies relating to what was done during the early 1980s. It concerned testing for air/aerosol distribution and the 10% acceptance criterion for the test. At the time, the station interpreted the 1975 and 1980 versions of N509 and N510 to resolve the immediate concern.

Eventually, startup testing of the system was reviewed and it was found that the test engineer at that time had picked different test methods and acceptance criteria from the 1975 and 1980 versions. It was determined recently to be a dangerous practice, i.e., you must stick with one edition or the other. When you mix the two, you risk not meeting either. Therefore, the station elected to accept to the 1980 edition and agreed to revise their test methods to comply with it. However, because their system was designed during the early 1970s, it did not meet the 1980 N509 criteria. In addition, while reviewing their test methods, they found more areas that had used the "pick and choose" approach. They had not made an aerosol distribution test on the HEPA filter downstream sample points, which were not required by the 1980 edition. They did not do an adsorber distribution test, which was not required by 1975, but was required by the 1980 edition. They did not do an air distribution test on a single HEPA filter bank, which was not required in the 1980 edition. When they went back and did the adsorber distribution test, they found some deficiencies caused by a junction box that obstructed flow into few of the slots of Type 3 adsorber. Engineering had to correct it. It showed that there was a potential deficiency in the test methods. This particular unit had back-to-back HEPA filters and adsorber beds that did not conform to N509 requirements for access between banks of components. Because of this, it could not be tested in conformity to N510 in its entirety. Based on these findings, the recommendations made by Phil, that I totally agree with, are: 1) Document the test program for a N509 or non-N509 system in detail. State what will be done to the system and the technical justification for doing it. 2) Consult with experts in the industry, and those who write the codes. 3) Communicate with your QA/QC inspectors, your region NRC people, your residential NRC people, and the various auditors who will come into your plant. With that technical justification you should be able to justify the test program you have selected.

DISCUSSION

WEIDLER: What I heard from the presentations is that most of the systems were designed prior to the issuance of N509 or that they were not designed totally in accordance with N509. Yet, we are using N510 to test them. When you can't conform to the letter of the law, the key factor appears to be good documentation and good justification. That is the approach we take at Duke and that is the approach recommended by the panel. We want to see if there are code or standard changes that should be made. I think Mr. Porco made a key point when he said, the time to consider testing is when you are designing the systems not when you are getting ready to test it. I am guilty of this and probably every other system designer is, also. I hope we can put the lessons we have learned into effect for the next generation of plants.

BELLAMY: I would like to make the point that we have a perception that when you have a non-N509 system that can't be tested according to N510, you don't know anything about the system. I think we should be more positive than that. I think Mr. Porco stated it admirably when he said that the goal of in-place testing is not to use N510 to prove whether or not we have an N509 system, the goal of in-place testing is to make sure that everybody has the perception that the system is adequate to perform its intended function, whether that be after an accident or whether that be after normal operation. If any selected in-place testing method proves that the system will work, that is really the goal. We shouldn't get obsessed with whether a system is testable according to N510 and whether it does or does not satisfy all the criteria of N509.

WEIDLER: We have to use standard N510 for testing. Whether we call it perception or something else. NRC's resident inspectors are taking a very legalistic approach to both the technical specifications and the standards that we use. Therefore, we have to solve that problem first.

JACOX: Ron Bellamy, I couldn't agree with you more. You stated it very well. Based on discussions with panel members, I think we all agree with you. I also agree with Ray. Most of your NRC colleagues in the field, with a few exceptions, are not as enlightened as you. Given the perception that most utility licencing and QA types have, plus their fear of the Commission, they tend to be blindly legalistic. That was part of the point I was trying to make, there is an absolutely legalistic approach taken by both the utilities and the Commission rather than simply reviewing it as a technical problem that requires a technical solution.

PORCO: I referred to the "black box theory," i.e., if you put DOP in upstream and don't get any downstream, you have a good system. That is a simplistic view but seems to be what you were saying, it is O.K. if it protects the health and safety of the public.

KOVACH: If you put DOP into a black box and DOP does not come out, it does not mean that whatever you normally put in may not come out. The first thing you have to realize is that we are using a simulant. The second is that we have very few personnel present from the NRC at these Conferences. We have very few personnel from the NRC present, Ron Bellamy, Charles Gill, Jack Guttmann, Jack Hayes, Forrest Remick, Robert Shepard, Leonard Soffer, and Charles Willis in code or standard writing groups and who have the depth of knowledge that is required to make a judgement. In the absence of that knowledge, they have a tendency to follow the words rather than the meaning of the standards. Maybe we need to have better training given at the NRC Regions to bring all the inspectors up to some minimum understanding of what it is they are trying to enforce. It is my understanding that some training has been offered but it does not seem to be enough to assure good quality NRC personnel at the sites in regard to in depth understanding of air cleaning issues.

KUMAR: The technical specification at my plant is absolutely clear about testing according to ANSI N510, such and such a year. The way to handle it is to take exceptions. N510 states that for non-

N509 systems, N510 is to be used as a guide. The technical specifications are what the NRC inspectors look at, they govern surveillance tests.

JACOX: The caution I would offer is, that you should be very specific about what you are going to comply with and how. So that you don't have a problem with your NRC inspector, put it down, write it up, get it signed off, and go with it.

ORNBERG: I would like to add to what Mr. Kumar said. We have had success at a number of our plants where, because of the age of the plant, we were designing to N509-1976, or 1980, or even before 1976, and having to commit to those standards after the fact. What we have done instead is to go through N509 and N510 line by line and identify where we meet the requirements and where we don't, what our justification is for using an alternative test method. We don't put it in the technical specifications but have it available for testing people and regulators to review. It helps focus on where problems might arise. Obviously it is best to do it up front, but we all go through a learning curve. As new people are added, they, too, go through the same learning curve. I think we need panels like this to keep everyone focused on what are the problems. I would also like to say that we have tried to pick up many of the problems people have talked about today through Workshops. One of the things we try to stress is that manufacturers must test units before sending them out into the field. I have gone through many designs where I thought I was specifying an N509 system, I thought I designed the plant layout that way, I thought I specified it that way, and I thought a manufacturer had submitted a proposal in accordance with it, only to find out after it got out into the field that there were portions that were not testable. There have been comments about designing testing capabilities into the system early in the design phase. A recommendation made to me, that I thought was good, was to have an independent review by a testing contractor early in the design. I think that way you can eliminate some of the errors. In addition, the manufacturer should do a lot of the airflow uniformity testing, manifold qualification testing, etc. in the shop to help prevent some problems before they get out into the field.

JACOX: I have a question for the NRC. Mr. Bellamy certainly has technical depth. I see Dr. Gill and Mr. Willis here, they certainly do, also. What is your recommendation when the plant QA/licensing people or an NRC inspector, resident or regional, says "We don't accept the disclaimer in N510. It says you are supposed to do testing according to N510 and that's what we want to see?" That has happened often. As a consultant, I only get calls when there is a problem and I don't know how to answer if a person can't read or refuses to believe what they read, and I don't know of any better way to phrase it. So, I am very seriously soliciting advice as to what you do when you run into such a situation.

BELLAMY: I am a Regional Branch Chief in Region I and I supervise the technical specialists in the Northeast part of the country who go into the field and judge the acceptability of in-place tests for ESF filter systems. I think you gave part of the answer to your question, you said yourself, a consultant only gets called when there is a problem. I repeat my earlier comment. We are into perceptions and you and others only hear about the times that it doesn't work. You don't hear about all the times that a resident inspector will call me up at home at night for guidance. As a matter of fact, I got a call yesterday from Indian Point II about this same issue. We worked it out and it gets worked out a lot of times. I know Dr. Gill and I have spoken on the phone a lot of times about such matters. I think we need to realize that, there are inconsistencies and, there are residents and region-based specialists that will abide by the letter of N510. Before I get to the basic answer, I again want to point out there are many more times when it works out that the NRC inspectors are reasonable and come to a happy solution before the matter needs to be referred to the regional level. Having a disclaimer in N510 to say that you may not be able to use this guidance for all your filter systems isn't sufficient. I believe inspectors are taught the same way plant operators are taught to apply verbatim compliance. The ANSI standard often says, "You shall." When the technical specification state that it must be done in accordance with N510, I turn the burden back on the licensee and say you should do it in accordance

with N510 unless you can convince QA, licensing staff, and NRC staff that what you are doing is equivalent. If that doesn't resolve the issue, it is time to refer it to regional NRC management, who should then get involved.

JACOX: I accept your statement that there are many more cases where there the issue is worked out. I am glad you made it. I find an internal contradiction in our statement about taking N510 verbatim in that part of the test of N510 is the qualification statement. Why is that one paragraph ignored if everything else is to be taken verbatim. It doesn't make semantic sense to me.

BELLAMY: It is too general; how can you say, "Implement N510 and if it doesn't work, you can do what you want." The disclaimer in my opinion does nothing. If you are going to have a disclaimer like that, it is the same as putting in front of the technical specifications (which is a legal, enforceable document), "If you don't like what is in here and if you, as a licensee, don't think you can implement it, you can do whatever you want." I submit there is no legal basis for that disclaimer and, legally, it doesn't do any of us any good. I think the point I made earlier, that, technically there are non-N509 systems that you just can't test to N510, is an absolutely truthful statement. I am sure no one is this room would disagree with that. But disclaimers like the one in N510 just don't help us out when it comes to regulation basis. The NRC strives to utilize its resources most efficiently. We will work with licensees to resolve deficiencies. However, it is not obvious that the disclaimer in N510 (that it only applies to N509 systems) has any legal weight, and it may, in fact, allow any variations from N510 that you want.

WEIDLER: Mr. Bellamy, you made a very good point when you said that you work out problems with your resident inspectors. Region I is blessed with the technical competence to work those problems out. The other regions in the country are not so blessed. Neither is NRR. Most of the other areas of the plant, such as the NSSS systems, are pretty well manned with expertise, either at NRR or at the regions. But HVAC is not. My question is, what is NRC going to do about solving that problem?

BELLAMY: That is an excellent question but I do not have an answer for you. It is an acknowledged problem. By definition, NRC is an auditing agency, we are not available nor able to look at everything every licensee does all of the time. The entire NRC staff numbers approximately 3,000 for the whole. In Region I, we have a budget of 264 individuals that includes overhead people, the management who stay in the region, and resident inspectors. We pick and chose exactly what areas will be emphasized, depending on the potential impact to public health and safety. Over the last 20 years you have seen an enormous increase in the ability of the air cleaning systems to do their job as well as the ability of plant staff to do their job and to test them appropriately. Maybe this is an area where the NRC doesn't have to look so hard in the future.

WILLIS: I am one of the people in NRR that you believe doesn't know how to review these matters. It has been my experience that whenever someone comes in ahead of time to explain a plant specific problem, we have been able to deal with it. When you wait until you have to have the test results and then come in to ask for a deviation the next day, we are likely to have problems. I would like to say that we seem to have done a relatively poor job in standards development in this area. N510 is a beautiful test standard; unfortunately, it seems to have been written for systems that do not exist. It has a disclaimer in the beginning which says, if you can't do it all, do whatever turns you on. If we take that disclaimer seriously, it makes it rather difficult for a regulator to rely on the standard. I would like to ask the panel whether anything has been done about writing a test standard for the systems that do exist, something that we could rely on.

ANNON: For every non-N509 system that is now being manufactured, a test procedure specific to that system is being developed at the same time that submittal drawings are made. The owner can now review the drawings and review the test procedure at the same time. If the owner has a problem

with either, it is the right time to bring the problem to the manufacturer or to the regulatory's attention rather than after it is built and put into service.

JACOX: In defense of N510, I don't think the disclaimer is quite as cavalier as it has been represented. The mandate that the committee received was to write a N509 standard for equipment and, specifically, a test standard for that equipment. That is what the first part of that disclaimer says, that N510 is to be applied rigorously to N509 systems. It is not the fault of the Subcommittee that wrote N510 or the Main Committee that approved N510 that people are not designing and building N509 systems. Neither is it the N509 Committee's fault that, although it has written a good standard, customers are not demanding that designers and fabricators build and install N509 systems. I agree that there are only a few N509 systems. I don't think it is fair to say that it is the fault of the standard preparing group that customers are not demanding that the standard be met. The wording of the N510 standard states that it may be used for technical guidance for testing non-N509 systems and perhaps that can be interpreted as saying, "Do whatever you want" but that was not the intent of the Committee. It is an attempt to stress the fact all the technical guidance in N510 is applicable to non-N509 systems when viewed from an engineering standpoint. I would like to stress the words "engineering standpoint" rather than "legal standpoint." We must look at these problems as technical/engineering problems, not legal problems. I understand the technical specifications are legal documents but if they are approached up front, rather than as a last second crisis, the issues can be worked out. Plant people generally take a legalistic approach out of fear of discussing things with the Commission. There is not a good working relationship when there is fear of asking questions. I think it is incumbent on the utilities and the Commission to do something to improve this situation. Once again, I say, it should be an engineering problem not a legal problem. As long as you think of it as a legal problem, you won't solve anything.

WEIDLER: At Duke Power, we have taken a big step in maintaining communication with NRC. We have an excellent relationship with our resident inspector at McGuire and we do not hesitate to go to him when we discover a problem or to call in a regional inspector. The fear comes after you discover a problem because it becomes a potential enforcement action that no utility wants. We have learned to overcome the communication barrier; I see the problem is the enforcement action that comes later.

ANNON: I think the codes are fine, the people working on them have done a good job. Problems arise when dealing with pseudo N509 systems. Early on, when buying an N509 system, you work with the procurement people, or the architectural engineers, or the mechanical contractors. They want to deliver an N509 system but then they discover a space problem and ask, "Can you cut something out?" Perhaps it is going to effect testing requirements later on. The code is not the problem. We are all concerned with testing our systems according to N510 but keep in mind that just because an N510 test was passed doesn't mean that the system performs well. We should always be looking at the entire system and how it operated rather than just go down a checklist.

PATEL: I want to refer to what Mr. Kumar said earlier about mentioning a non-N509 system in the plant's technical specifications basis. We either have to meet the Regulatory Guide or take exception to it. Both Regulatory Guides 1.52 and 1.140 refer to N509 and N510 but it is already known that the system doesn't meet N509 requirements. So we need a guidance from the local people to solve the problem.

KOVACH: Almost every system that I have seen is testable, but the problem occurs when you run the test and fail it. At that point you start complaining about N510 and say it is not a good test. I have never seen an owner send back a system to the manufacturer because it was not a N509 system. The owner says, "I brought an N509 system, the architect/engineer specified an N509 systems, but what I have is not an N509 system." When they realize they do not have an N509 system, and in some cases it is a non-working system, the endeavor is to come up with a test procedure to show that it is acceptable. I don't think that is what we want to do. I want to break the problem into two separate

portions. One, whether you can perform N510 tests rigorously on that system and two, after the system fails, can you modify the procedure to justify its acceptance. These are two different problems, let's not mix them up.

WEIDLER: That is correct, when you fail the N510 test you must do one of two things, either change the system or justify the results.

JACOX: One situation that is extremely common is bad airflow distribution. There are systems that are called "serpentine design" where flow direction changes 90° or 180° between every bank, sometimes 270°. How are you going to take good airflow distributions readings in this design because there is turbulence? You have performed an N510 test rigorously to prove there is turbulence and the system failed. All N510 provides is a method to secure accurate data for evaluation. That is not often recognized. In the heat of doing testing, the objective seems to be, "How can we get some numbers so we can go to the next test, turn this thing over, and get the plant started up." One million dollars a day to the utility is indeed a problem if you can not start up. But most of the time they end up wasting more time and having more delay by not recognizing the engineering facts. You must either justify the failure or you must modify the equipment. Rewriting the test procedure isn't likely to do much good.

HILLEY: There has been a lot of discussion about N509 and N510 but our technical specifications at McGuire specifically reference another document, Regulatory Guide 1.52. Is there something the NRC can do to provide more guidance in Regulatory Guide 1.52 by clarifying some of the concerns about non-N509 systems being tested according to N510? I would like to hear some comments on that.

BELLAMY: I am not going to answer that question now at all because that is the specific topic of a paper that will be presented tomorrow morning. I will be more that willing to answer that question in detail at that time. I think there will be people in the later session who may want to be a part of the discussion.

HAYES, J.: My question for the panel is, do we have systems which are not capable of performing their intended functions? I think Ron Bellamy made the point I was interested in. Although we have had a lot of discussion about N509 and N510, the important thing is, are the systems capable of performing the function for which they were designed? My first question is, do you find systems that cause you to question their capability to perform their intended function? Second, are there systems that qualify by a N510 test but the question remains to whether they can perform the intended function?

JACOX: Unfortunately, the answer is "yes" to both questions. There are systems that I can't be sure will meet their intended function because I am not able to run a test that I feel comfortable with. For example, if I cannot do an airflow/air aerosol mixing test, I can not be assured that I am challenging all possible leak paths, the gasket area, the frame to wall wells, etc. There are times when you cannot run the test because there is no access. Other times you take data and mixing is inadequate. The expedient solution is to justify running the leak test. The consultant or testing contractor carefully writes it up and puts it in the test report. The customer may say he can justify the exception and ask you to run the next test. If there is real danger, a consultant has a legal obligation to go directly to the Commission with the information but it is hard to justify this action when something is "iffy" at best. Because the plant owner may be able to justify it away, I am unlikely to feel that it is proper for me, as an individual or representing another company, to call the NRC directly until the utility has had a chance to do their engineering justification. Generally, that happens after the consultant is gone. It is a catch-22 as far as the ethical and legal responsibilities lie. You may question whether systems that pass tests have design flaws, but that is outside the N509, N510 criteria. You may have enough experience at working in a plant to say, the system works but it only handles 1,000 cfm when it ought

to be 10,000 cfm. On the basis you may question whether it will perform the intended function, but that is a design question, not a test question.

WEIDLER: I believe the fact that plants continue to operate answers the question affirmatively. At Duke, we take a very serious view of the letter of the law. We either justify test failures or we modify the equipment. We have a problem investigation report process that we use internally and usually the NRC is involved. If we can't resolve the problem, we shutdown. That is why I believe the answer to the question is, "yes." We don't have an ideal situation and we probably have made modifications that we would rather have not, but to make sure the system is completely operational and ready to perform its function, we may defeat certain small interlocks or make necessary repairs. I am not worried that we have unsafe systems.

HAYES, J.: I would like to make two comments. One, I am a project manager for Virgil Summer and I have responsibility for licensing actions for that plant. If any of you experience a problem with your plant, I think one of the appropriate things for you to do, in addition to working with the regional and resident inspectors, is to make sure that you have your project managers on board so that they can get the appropriate people in NRR and the region and the residents to have discussions with you. Through such discussions, I think most problems can be resolved. The second thing I would like to say, is that I think the NRC has taken some hits this morning concerning the capability of its people in HVAC. I want to share with you my perceptions and experience. I don't think most people set out to be HVAC engineers, they sort of fall into it. In other words, it was a job they were assigned. My experience is that it calls for on-the-job training. The people that become involved in this activity seem to get very little respect, like being a Rodney Dangerfield. It may sound facetious but it is very true. I remember going to one plant where we were evaluating one of their systems. We were looking for information but couldn't find anyone at the plant who had any expertise or experience with their HVAC systems. Finally, someone said, "Yes, I had responsibility for one system for about one year and I was real happy I was able to get rid of it." Unfortunately, I think that is the same situation that we find here at the Conference, the expertise is diminishing at the same time qualified people are having difficulty with employment because of cut backs by manufacturers and owners. In addition, NSSS vendors are coming up with advanced light-water designs for which they think they do not need ventilation systems. Therefore, we have a selling job to convince people that this subject is still important. In essence, you are providing one of the major protection barriers for your inplant personnel and your offsite population. We have a job to do to show people that this is, indeed, important. The only way you can do this is through an industry-wide effort. Otherwise, you will have a committee of 20 coming to air cleaning conferences. You will lose expertise because I think most people here would agree that it is hard to make a living anymore practicing nuclear HVAC engineering.

WEIDLER: I appreciate these comments and I agree with them. Jack Jacox pointed out that he, too, has experienced a lack of expertise in the industry. The reason people at this session picked on the NRC is probably because there are more of us than there are of you and that is true for the industry. But the point to be made is that both the industry and the NRC need more expertise to solve the problems that we face.

JACOX: First, I apologize to Jack Hayes who I didn't see when I mentioned your three colleagues as being well qualified in the HVAC area. I certainly would have included your name had I seen you, and included you among the good guys that have ability to speak off the record and with whom one can have an exchange of expert technical information. Second, my personal experience has been there is a lot more trouble with the utility people being rather blindly legalistic with their QA checklist than with the NRC people. That may be because there are a whole lot more utility people, but I think I mentioned that in my introductory talk. The utility people often are less flexible simply because they are so afraid to bringing something up to the NRC, but not at Duke. Third, as an independent consultant, I can certainly agree that there are fewer and fewer employers willing to pay

for expertise in HVAC. Years ago business was good enough that architect/engineers to were willing to hire an HVAC consultant, but today things are so tight they try to be all things to all people. It is hard to convince them to spend money outside. There are people in this room with the necessary expertise who simply are no longer in a position to put it to direct use, they have become senior managers or unemployed consultants.

GLADDEN: I would like to second Mr. Willis' comment that we concentrate too much on N509 systems because our codes and standards are designed around it. However, many people are using N509-type systems for other purposes. The army is particularly involved. Therefore, I think the CONAGT Committee ought to be looking at these other applications by preparing a code or a code section that would apply to non-nuclear toxic or hazardous systems. If there was a good standard for testing those kinds of systems, as a supplement to N510, it would support the public good.

KOVACH: The US Army has lately been the major purchaser of N509-like systems but their specifications require N509 compliance. This means that they thought they ordered N509 systems and what they got were not authentic N509 systems. Now it is necessary to come up with a procedure to test them. Testing will be possible, but it can not turn them into N509 systems.

EDWARDS: A good system begins with good specifications. The architect/engineer is responsible for writing good specifications. After the specifications have been issued, good or bad, there is not much anybody can do except conform to the specifications. When there are poor specifications there will be a poor system. Although Steve Ornberg said his young engineers are at the beginning of their learning curve, there is still a body of knowledge among industry people who have already gone through their learning curve. We should make sure that young engineers don't make the same mistakes that we made 20 years ago.

PORCO: I wish to point out that even though you may not have an N509 system, you may have a functional system and that is what you are after.

CROSBY: I would like some comments from the panel. In the standard that so rigorously outlines procedures for getting a uniform aerosol concentration upstream of the HEPA filter system, there is no requirement for a minimum aerosol concentration challenge. On the downstream side one sample can be taken 8 to 10 duct diameters downstream. This procedure dilutes the leak with clean air which tends to give a lower reading. Why does the nuclear industry use a maximum allowable leakage of 0.05% when all the other industries, pharmaceutical, electronic, etc. use 0.01%?

PORCO: Because an absolute filter is not really an absolute filter, 3 particles out of every 10,000 that filter sees go through the filter. The intent is to remove particles down to a level that is acceptable. Therefore, what you are trying to determine is the aerosol concentration in the mixture.

OLSON: We test to 0.05%. because our technical specifications call for it. The penetration figure is based on allowable leakage for offsite conditions. We have some systems that are acceptable at 0.2% penetration. It depends on the application.

ANNON: I agree there should be a specified upstream concentration.

KOVACH: The standard is very clear that a range of 10^5 is for the detector. If you have difficulty increasing the low end detectability, you must use close to 100 µg/L upstream. If there are no minimum detection and drift problems at the low end, you can probably go as low as 10-20 µg/L upstream, but it will still be necessary to show that you have less than 0.05% penetration. The requirement the current performance requirements relate to is not the technical specifications, but on the required dose

calculations for the population and that is why some may be tighter and some looser. In some cases, 5% leakage can meet the requirement.

CLOSING COMMENTS OF PANEL MODERATOR WEIDLER

I believe it has been a good session. I believe the objectives of the session have been met. We found out that we do not have a lot of problems with the current standard and that we do not need to do a whole lot more work on it. But we discovered that we need more training of personnel. The Committee on Nuclear Air and Gas Treatment is helping by holding a short course this week on N509 and N510. I hope others will follow. We learned that the disclaimer in N510 is not much good and we will look critically at that again. I think we need to review other applications for the standards, such as for the military, and the DOE, to make sure that the standards cover their applications; that will be a continuing project. Finally, we heard that both NRC and industry have an equal need to increase their level of expertise in dealing with these systems.

All in all, I believe we met our objective and I thank you for your presence and your interest.

SESSION 6

WORKING LUNCHEON

Tuesday: August 14, 1990
Chairman: D. W. Moeller

CHANGING WINDS IN NRC'S REGULATORY ENVIRONMENT
Forrest J. Remick

CHANGING WINDS IN NRC'S REGULATORY ENVIRONMENT

Forrest J. Remick, Commissioner
U.S. Nuclear Regulatory Commission
Washington, DC 20555

Good afternoon, ladies and gentlemen.

I'm honored to be addressing you at the 21st Nuclear Air Cleaning Conference. With Congress deliberating over its first good chance to amend the Clean Air Act in the last thirteen years, I figure that I couldn't be in a better place at a more propitious time to share my thoughts on the future direction of nuclear air cleaning technology.

I say this because I believe that in the long run, the action is not in Washington, but out here in the field, where the technology is being developed and applied. The clean air laws and regulations made in Washington unquestionably have a major effect on your professional endeavors, but these laws and regulations are in large part a reflection of the prevailing public attitudes about nuclear technology and the need for better air pollution control. Those attitudes are formed out here in the countryside, not back in the capital, largely in response to people's understandings, or misunderstandings, about how well you are doing your jobs.

If the title of my talk, "Changing Winds in NRC's Regulatory Environment," has given you the sense that I will be focusing my remarks today on this larger political and institutional environment for air cleaning technology, the title has done its job. As I see it, these changing winds bring both significant long-term opportunities and significant short-term risks to the career of this technology.

Let me first speak of the opportunities. I think the nuclear power industry could be at the threshold of a significant renascence. For financial, environmental, and public health reasons, and because of global and domestic factors affecting energy supply, I believe nuclear power could claim an expanding portion of the nation's energy mix in the coming decades.

Certainly, the overall energy demand will be expanding. The Department of Energy, in its Interim Report on the National Energy Strategy cited estimates by investor-owned utilities and other sources that future growth in electricity demand will be about 2 percent annually nationwide, implying a need for at least an additional 100 gigawatts of generating capacity above that currently planned by the year 2000.

A factor nudging these utilities toward choosing nuclear to meet this new electricity demand may be an increasing public awareness of the risks of fossil fuels. Concerns about acid

rain, ozone depletion, and global warming have received increasing attention by media and policy-makers, and even some environmental and conservation organizations have modified their opposition.

In what may be a subtler but potentially more significant development, my intuition tells me that with what many see as a decline in U.S. economic competitiveness, and with the example of successful technological innovations in other countries, Americans may be more willing now to take a more balanced view of technology not only as a source of problems, but a potential solution. The revived interest in electric cars, particularly in the smoggiest U.S. urban areas, is a case in point.

In addition, the growing use of nuclear medicine suggests that people can and do make distinctions about the sources of radiological risk, that they understand that there are tradeoffs between radiological and non-radiological risks, and that they are capable of weighing one kind of risk against another.

In this context, the growing awareness of the costs and risks of fossil energy use, and the example of safe and efficient nuclear power in other industrially advanced countries, may enhance the American public's receptiveness to a greater reliance on nuclear power -- under the right conditions.

This is by no means to suggest that the nuclear industry can afford to relax, however. I would argue just the opposite: that more diligence in making all nuclear applications cleaner and safer is in order. The risks of losing public confidence are as great as the opportunities for gaining it, and the implications for federal air pollution control policies are considerable.

I probably needn't remind you that to begin with, public confidence in things nuclear is not high. Even in some communities that have lived with major nuclear facilities for years, and where there is widespread concern about jobs and the community's economic future, people are unwilling to support new nuclear facilities.

An example particularly close to home for nuclear air cleaning technology in my native state of Pennsylvania is the withdrawal of the effort to site an advanced incinerator in Apollo, a rural host community for a longstanding defense nuclear research and development facility.

A public attitude survey cited in a recent report on low-level waste incineration for the Commission's Advisory Committee on Nuclear Waste found that only 31 percent of local respondents were "very" or "somewhat trusting" of the company that developed the incinerator, 34 percent were "very" or "somewhat trusting" of the state environmental safety office, and 39 percent were "very" or "somewhat trusting" of NRC. I do not take comfort from the

fact that NRC managed to stay out of the cellar of public confidence.

After a thorough examination of the operating experience and developmental status of different incineration technologies, a discussion of off-gas controls, a cost comparison with land disposal, and a look at current state and federal regulatory requirements, the Advisory Committee report went on to conclude that incineration can have a significant role to play in waste management.

The report noted that most of the technical difficulties of incineration have been resolved, and the remaining problems result from poor operations and maintenance practices or a low capacity factor.

The principal remaining obstacle to wider adoption of incineration, however, is public acceptance. It is worth quoting from the report's conclusions on this point:

> "Ironically, the lack of public information about incineration, which results in misconceptions and distrust, is probably the greatest cause of public concern. The technology behind LLW incineration has been carefully engineered and, consequently, should be viewed favorably by a well-informed public -- especially when incineration is compared to current LLW disposal practices."

Clearly, those of you in the business of incineration and its supporting technology have your work cut out for you. Despite the fact that the overall environmental impacts of incineration compare favorably with the impacts of burial; despite the fact that, as the report points out, the Europeans, Canadians, and Japanese are demonstrating that proper treatment of incinerator effluents and ash "can effectively preclude any significant short- or long-term release" of hazardous or radioactive materials to the environment; despite that fact that regulatory requirements can be met, and government policies generally are not unfavorable -- the public isn't buying.

I suspect there are several reasons for the continuing unfavorable winds in the climate of public opinion. One might be a prevailing gut-level view that when you put incineration against land disposal and imagine worst coming to worst, you can get another source of water to drink, but you can't get different air. Never mind the statistical probabilities of either worst case scenario actually coming to pass.

Another source of public skepticism is that we don't yet have enough facts to settle all the questions about the risks of radiation. No doubt many of you have read about the finding of increased cancer risk from radiation in the latest report of the National Academy of Sciences' Committee on the Biological Effects

of Ionizing Radiation, or BEIR V. Using re-evaluated data from studies of the Hiroshima and Nagasaki atomic bomb survivors and other sources, BEIR V concluded that the statistical lifetime risk of contracting fatal cancer from a "low-level" dose of radiation is three to four times higher than previously believed.

Note that this so-called "low-level" dose is actually ten rem. Although ten rem is more than twenty times the average annual dose received by workers in the nuclear power industry, it is at the low end of the range of doses at which researchers have been able to analyse effects on large populations.

The BEIR V report itself pointed out that carcinogenic effects have been observed only at relatively high doses and dose rates, and that studies of populations chronically exposed to what most of us think of as low-level radiation, such as those residing in regions of elevated natural background, "have not shown consistent or conclusive evidence of an associated increase in the risk of cancer."

Nevertheless, the news that radiation, even at the comparatively high dose levels and dose rates experienced by A-bomb survivors, can be three to four times more carcinogenic than previously thought has posed legitimate questions, both to the public and to policy-makers in Washington.

How do we know that future research will not find even greater health risks from a given quantity of radiation? How should we set limits on emissions or effluents that will stand up over time as adequately protective? Do the findings of expert groups like BEIR V mean that we should be more conservative in setting standards? If so, how much more conservative?

These uncertainties will be played out in the current legislation to amend the Clean Air Act. As most of you probably know, last year EPA promulgated standards for radionuclide emissions that are now in effect for all DOE facilities and a number of other civilian operations.

EPA is currently reconsidering application of the standards to NRC and Agreement State licensees, however. Responding to comments on the proposed rule, EPA agreed to entertain petitions for reconsideration of the application of the standards in the final rule. NRC, the National Institutes of Health, and others in the biomedical and industrial community submitted petitions arguing that these standards would impose unwarranted compliance costs and provide no additional public health benefits.

The Senate Clean Air Act bill has an amendment providing that no EPA standard is required for NRC or Agreement State licensees if the EPA Administrator finds that the NRC regulatory program provides an ample margin of safety to protect public health. The House bill has no comparable provision, and the

difference will have to be resolved in the House-Senate conference committee.

No doubt you can imagine the consequences to licensees, and to the nuclear air cleaning profession they support, if the arguments for the existing regulatory regime prove unpersuasive. In the proposed EPA clean air rule last year, for example, one alternative emission standard for licensees was based on a one-in-a-million lifetime risk of fatal cancer -- a risk criterion now in both House and Senate bills for setting standards for other hazardous pollutants. The proposed EPA dose limit based on that one-in-a-million lifetime risk was three-hundredths of one millirem per year.

At the risk of sounding unduly alarmist, I would venture that avoiding an impracticable Clean Air Act standard is by no means assured. I find it significant that, despite arguments to Congress by the National Institutes of Health and EPA itself emphasizing the questionable public health benefits, considerable costs, and unwanted effects of dual NRC-EPA regulation on medical treatment and biomedical research, the House did not attempt to change the status quo, and the Senate rejected an initial amendment that would clearly have removed the authority for continued EPA jurisdiction.

In light of these ominous clouds on the regulatory horizon for the nuclear air cleaning industry, both regulators and regulated need to redouble their efforts to demonstrate a conscientious commitment to protecting the public. Let me turn now to discuss a recent NRC decision bearing on this commitment that will affect all of you.

Just this past spring, the Commission approved a major revision of its general radiation protection regulations, which are known to health physics professionals as "Part 20" because they appear under that part of Title 10 of the Code of Federal Regulations.

The new requirements, which will take effect January 1, 1992, cover all licensees, including those associated with reactors, incinerators, medical and industrial facilities and services, and others. For some of these licensees, these requirements may affect the manufacture and design, construction, operation, and decommissioning of facilities and equipment.

In general, the revisions conform NRC radiation protection requirements to updated recommendations of national and international bodies of expert opinion on health physics.

Here are some of the highlights of the new Part 20 revision:

-- Using revised models for effects on the lung and gastrointestinal tract, and more recent data on metabolic retention, NRC has calculated new Annual

Limits on Intake and Derived Air Concentrations for workers, and new concentration limits for releases to the environment, for more than 750 nuclides, about 500 more than the 260 radionuclides listed in the previous Part 20.

-- To calculate total whole-body doses to their workers, licensees will have to sum the doses from radiation external to the body with internal doses "committed" by the inhalation or ingestion of radioactive materials that continue to contribute radiation doses over time.

-- Under the new standard, the total effective dose equivalent from both external and committed internal radiation may not exceed five rem per year from normal operations. This is a reduction of more than two-thirds from the old limits.

-- There are additional specified limits on accumulated internal doses, and doses to the embryo and fetus of a woman radiation worker who declares herself pregnant. New limits on doses to the public are one-fifth the old limits.

-- Licensees' radiation control programs are now required to have documented measures for reducing doses to workers and the public to levels as low as is reasonably achievable, a concept known as "ALARA." Licensees were told only that they "should" keep doses ALARA under the old Part 20.

I should note here that the ALARA concept, even as a hortatory provision under the old rule, has turned out to be an excellent insurance policy for most licensees. It has kept occupational exposures, and effluents and emissions to the environment, low enough that few licensees should have significant difficulty meeting the tightened new standards.

Reactors, for example, have routinely been able to meet an ALARA limit of five millirem per year to any member of the public -- one twentieth of the new 100 millirem limit. The average measurable annual dose to a worker at a nuclear powerplant in 1989 was .34 rem, less than a tenth of the 5-rem limit on total effective dose equivalent per year under the new rule.

My point here is not that licensees can rest on their laurels as they approach the implementation of these new, more stringent standards. Given the changing winds in the current regulatory environment, I am suggesting that it is always wise to

stay alert to the possibilities for reducing exposures through refining operating procedures or applying new technology, whether the application is to an existing facility or the design of a new one.

Radiation protection is still a changing science, and as the public and political reaction to the latest BEIR report has made clear, there is always the possibility that new information may lead to further tightening of prevailing standards.

In light of this, I believe that the added cost of designing and operating reactors and other facilities to increase the efficient use of inputs and minimize the production of wastes and effluents will continue to be well worth the investment.

I hope the discussion today has given you a feel for the shifting crosscurrents in the political and regulatory atmosphere in which both NRC and the industry must make their way. I hope we can all avoid wind shear. With that, I thank you for your kind attention and will be pleased to answer questions.

SESSION 7

OPEN END

Tuesday: August 14, 1990
Chairman: M. W. First

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CLOSING COMMENTS OF SESSION CHAIRMAN FIRST

OPENING COMMENTS OF SESSION CHAIRMAN FIRST

This is meant to be an opportunity for presenting work that is in progress, or very short reports that are not stout enough for a full 20-minute presentation, or for the purpose of asking the audience for the answers to questions.

QUALITY ASSURANCE REQUIREMENTS FOR NESHAPS COMPLIANCE

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ABSTRACT

The U.S. Congress is reauthorizing the Clean Air Act. Three major issues in the reauthorization are acid rain, air toxics and automobile emission, all of which present air cleaning challenges. Radioactive materials emissions are included in the air toxics category, and in December 1989, a very restrictive new standard was issued by EPA. Compliance with the new standard, and demonstration thereof, presents a technical challenge to the nuclear air cleaning community. This paper discusses the compliance challenge and discusses a reasonable Quality Assurance program which, if implemented, could assure and verify compliance with the new standard.

Background

The Clean Air Act (CAA) is currently being reauthorized, the first attempt to toughen air pollution legislation since the 1970 Clean Air Act was amended in 1977. The pending legislation contains provisions to combat three unresolved issues: acid rain, air toxics, and certain automobile related problems, namely, ozone, carbon monoxide, and particulate matter (1).

This discussion focuses on certain of the air toxics issues. In general, compliance with the CAA reauthorization will require major sources of pollution involving about 200 chemicals to acquire best available control technology by the year 2000. Since 1970, the EPA has regulated only eight airborne toxic substances, specifically by establishing a National Emissions Standard for Hazardous Air Pollutants (NESHAP) for each. For some substances more than one NESHAP has been promulgated, such that there are currently 12 NESHAPs. Obviously, there will soon be many more NESHAPS with which to contend.

In December 1989, EPA issued a revised NESHAP related to radioactive material emissions (2). There are 12 source categories listed in the new NESHAP, several of which are of interest to nuclear air cleaning specialists, because they apply to DOE Facilities, NRC Licensed and non-DOE Federal Facilities, and Uranium Fuel Cycle Facilities. The new NESHAP will have an impact on the Nuclear Industry, especially from a Quality Assurance viewpoint, e.g., demonstration of technical compliance by design, implementation and record.

Technical Issues

Over the years, annual public radiation exposure limits have decreased dramatically. Until about 1980, the annual limits were 500 millirem (mrem) to the general public, with a limit of 170 mrem to a "suitable segment" of the population. When EPA first published its regulations related to the nuclear fuel cycle, the annual exposure limits dropped to a whole body dose of 25 mrem to the general public, with applicable annual organ dose limits of 75 mmrem. DOE's internal Orders system established a policy called "As low as reasonably achievable" (ALARA) which was to limit annual doses to the public to 100 mrem from ALL sources and pathways.

Nuclear regulations (10CFR20), and DOE Orders applicable at the time (AECM 0524) also established air emissions standards in terms of concentration guidelines (uCi/cc) which were related to the annual dose limits of 500 mrem/year. These concentration guidelines have not been changed to reflect the lower dose limits. In addition, during the past ten years certain radiation effects quality factors were increased, and so was the method by which annual doses were calculated and reported. (There are now about nine different expressions for annual dose.) Because the changes were in general, "conservative", (Quality Factors were for the most part raised) they tended to reduce applicable dose limits, and, therefore, the applicable concentration guidelines for radioactive emissions.

The newest NESHAP (12/89), applicable to DOE and NRC licensed facilities, has further lowered the annual dose limit to 10 mrem/year, effective dose equivalent (EDE). Further, all buildings, and operations within one contiguous site are considered to be a single facility; the sum of the annual doses from all stacks or emission points combined must be equal to or less than 10 mrem. To a large degree, the justification for the latest regulatory ratcheting, e.g., the new limits, is that DOE sites, according to their annual reports, appear to meet them. One hopes that the quality of the sampling and measurement techniques and the assumptions and calculations which converted exhaust air emission data to EDEs is adequate to support the reported data. For sites with multiple radioactive materials emissions points, small errors in the same assumptions, measurements, and calculations which provided justification to the new standard could lead to the appearance of noncompliance with the new standard.

For existing facilities, with no major facility additions or modifications planned for a site, or increased activities, the impact of the new standard might be minimal (again assuming proper sampling, measurement and dose assessment). However, planning for new activities, modifying facilities or changing operations will make compliance more difficult. These planned activities require safety analyses, including risk/dose assessments to predict compliance with limits and to define types and numbers of air cleaning devices to be installed in exhaust air emissions systems. Specific and highly restrictive exhaust concentration guidelines for new or

modified facilities must be developed. Depending on how close the overall site is to its 10 mrem annual limit, the additional proposed activities could cause the limit to be exceeded, or existing limits might require changing. For design purposes, there is also the need for some degree of conservatism (ALARA).

Because of the more stringent requirements, there could be a need to review the current standard for HEPA filter efficiency to see if the .003% transmission standard provides adequate environmental pro-tection. Possible impacts of the recent nuclear NESHAP might be to place more severe requirements on air emissions control systems, and measurement systems as well. Demonstration of compliance will rely on QA/QC techniques related to design, installation and opera-tion of these systems, and adequate documentation thereof.

The QA/QC Challenge

The technical challenge presented by the recent 10 mrem NESHAP can be addressed by an overall systems analysis. There are several essential steps in the process; each step of the analysis must contain QA/QC elements. The steps are:

1. Safety Assessment/Operational Analysis
2. Selection and Quality Control testing of critical air cleaning components.
3. In-place (as installed) systems testing.
4. Design, test and calibration of exhaust air sampling and monitoring equipment.

Each item and associated Quality requirements are addressed below.

1. Safety Assessment/Operational Analysis

The analysis provides direct input to design of the exhaust air emissions system. Types, forms and quantities or concentrations of potential hazardous air pollutants released from the system are identified. Based on process knowledge and operational assumptions, the exhaust system flow rates are calculated, and concentrations of haz-ardous air pollutants (radiological and chemical) are compiled. Applicable regulations (40CFR61) are reviewed to identify best developed available technologies (BDAT) or applicable concentration guidelines. For new or modified systems, calculations to predict the public impact in terms of annual effective dose commitments are performed. The impact of the new or modified system is added to the current dose assessment value, and the new total is compared to the NESHAP (10mrem). If it appears probable that the new or modi-fied system will cause the site NESHAP to be

exceeded, further mitigation measures are necessary, either as a new concentration guideline, further design or both. Appropriate decontamination factors for the intended air cleanup equipment will be applied, and based on calculations, the number of needed cleanup devices in series will be identified to predict expected emissions concentrations. New dose assessment calculations, taking credit for the efficiency of the cleanup devices, will be performed to help confirm adequacy of the system design. One tradeoff might be to use more efficient filters or other air cleaning devices to save an additional air cleaning stage.

Quality Assurance is provided by redundant, independent review of assumptions and verification of calculations.

2. Quality Control Testing of Specific Air Cleaning Components

After design and construction of air cleaning systems are complete, Quality Control for critical air cleaning components is most important. At the Department of Energy nuclear facilities, there is a formal QA program in effect consisting of strict procurement requirements, filter media testing, destructive and nondestructive testing by the manufacturer or designated agent, and additional performance testing at the DOE HEPA Filter Test Facilities. Records of all these activities are prepared and maintained. DOE has found that when the entire component system testing program is functional, the quality of the filters remains acceptably high. When a part of the testing system is eliminated, the quality of the filters has appeared to deteriorate to some degree. The FTFs have had a stabilizing effect on the quality of HEPA filters used in DOE nuclear facilities.

Component testing is important because users need to know that the quality of the filters they will install in their air cleaning systems is acceptably high. If defective filters are installed in contaminated systems, and the system fails the in-place test, the newly installed but unused filters must be removed and discarded as nuclear waste, which is both cost and safety ineffective.

3. In-Place Systems Testing

Once the tested components have been installed in air cleaning systems, the entire system must be tested prior to use and periodically thereafter. The systems test is performed to ensure that the systems components have been properly installed such that there is no leakage path around them or through them due to damage incurred during installation. The current test is less stringent than the individual components test, especially in large systems, however, to date, the test has been useful in helping to predict that the air cleaning system is adequate and functional.

4. Air Emissions Sampling and Monitoring

Evidence that air cleaning systems are performing acceptably, e.g., the systems are in compliance with applicable NESHAPS, is provided by stack emissions monitoring. Stack monitoring data at DOE installations were used to help establish the current NESHAPS in that EPA risk assessments were based on DOE's 1986 report of emissions concentrations, actual meteorological data and population distribution. The DOE data indicated that maximally exposed individuals received annual radiation doses of about two millirem in that year. It was assumed that the air emissions sampling or monitoring systems were properly installed, calibrated and operable, and that real time monitoring devices or sample analytical systems were also properly calibrated and operable. In short, it was apparently concluded that the DOE air emissions data was a valid set of data, supported by documented records.

Quality Assurance Program For NESHAPS Compliance

The system described above exists for the most part within the DOE nuclear program and to a lesser degree in NRC licensed programs. The overall system with proper controls constitutes a Quality Assurance program. Safety analyses for design of nuclear air cleaning systems are conducted at both DOE and licensee sites. At DOE, there is an implemented policy which provides direction for procurement of nuclear grade filters, specifies destructive and non-destructive tests for HEPA filters by or under direction of the manufacturer, and requires penetration and resistance testing at a DOE FTF for all HEPA filters used for environmental protection. There is no comparable component testing program requirement for air cleaning systems which handle gases or vapors. The NRC does not currently require independent HEPA filter or other component testing, but appears to have stronger design analysis requirements for its nuclear facilities. However, DOE will soon demonstrate a design analysis system comparable to that of NRC for the new production reactor at Savannah River, and for other new or modified facility projects. In-place testing of installed air cleaning systems with HEPA filters is required at DOE and NRC licensed facilities, but DOE has not always required in-place testing for other types of nuclear air cleaning devices. Stack sampling/monitoring system design, installation, and calibration (initially and periodically thereafter) appears to be a weakness at DOE sites, and may also be a weakness at some NRC licensee sites.

Compliance with the new radiological NESHAP for DOE and NRC can be accomplished and demonstrated by augmenting, integrating and formalizing the system described above. The primary requirement is to define and document a systems program which demonstrates compliance with an annual dose commitment of 10 mrem/year, and the secondary requirement is to create by calculation applicable stack or emissions point concentration guidelines which act as action guides to help ensure compliance. Elevated concentrations must be quickly and accurately detected in order to avoid noncompliance with the NESHAP.

Following is a discussion of how the major elements discussed above can, when implemented, provide documented compliance with the radiological NESHAP or any other.

1. Safety Assessment/Operational Analysis

The basic safety assessment concept is good. Techniques used by some begin with assessment of upper limit accident conditions (bounding case assumptions which are super-credible) followed by more reasonable upper limit accident assumptions and then assessment of expected concentrations for normal operations. Operating emissions limits (alarm points) must be established for all potentially major radioactive materials release points, because the annual dose limit applies to the whole site, not each emission point. These alarm points must relate to relevant concentrations and instrument detectability.

Accident calculations are also necessary to establish the degree of air cleaning necessary to comply with requirements. Contaminant removal efficiencies of all cleaning devices should be substantiated by component test. One possible trade-off is to substitute higher efficiency cleaning devices for numbers of cleaning devices in series. For example, current limits for HEPA filter efficiencies (99.97%) could be evaluated to see if higher efficiency filters (clean room filters) might be required or desirable.

Records of all calculations should be retained, and all calculations and assumptions should be reviewed and verified.

2. Quality Control Testing of Specific Air Cleaning Components

Independent testing of HEPA filters by the DOE Filter Test Facilities has helped maintain the high quality of the filters used for environmental protection at DOE sites. However FTF test capabilities have not quite kept pace with development of new filters. Currently, filter manufacturers have begun to produce filters rated at 2000-2500 CFM, but only one of the FTFs can test at 2000 CFM, and none can test at 2500 CFM. In addition, only one FTF can perform the high humidity/high temperature QA tests. The FTFs should be able to test all filter units manufactured and identified for use in DOE facilities. Further, for NESHAPS compliance purposes, the FTFs could be expanded to provide tests of other air cleaning devices. This capability could become more necessary as the new NESHAPS for non-radioactive hazardous air pollutants are developed, and new types of cleaning devices are designed.

If need for higher efficiency HEPA filters arises, the test method must also be reviewed to determine if measurement of the higher efficiencies is feasible with existing test equipment 0^U(^U ^^(measurement methods. Whatever the outcome, the value of Quality Assurance component testing has been well established. Component testing can pay for itself if defective units are identified before

they are installed in air cleaning systems.

3. In Place Systems Testing

Need for in-place systems testing for HEPA filter installations has been well established in the nuclear industry by the requirements of ANSI N-510. The requirement should be expanded to include in-place testing of other air cleaning systems which are present for NESHAPS compliance.

4. Air Emissions Sampling and Monitoring

This subject might require the most technical attention at first. Existing radiological sampling and monitoring systems should be reviewed to include the following:

1. Collection of representative samples, including periodic calibration of the sampling systems and flow rate verification.

2. Calibration of real time monitoring systems, including:

   a. calibration standards with appropriate emission energies.
   b. corrections for particulate deposition on sampling medium (particulate loading).

3. For systems which require collection of samples with analysis elsewhere, apply EPA "chain-of-custody" Quality Program requirements related to sample collection, packaging, analysis and data validation.

The new NESHAP was derived from DOE stack sampling data. It would be interesting to attempt to validate this data, using qualified, independent entities. The data were collected during a low work activity period in the nuclear field in general, and DOE in particular, possibly resulting in reduced airborne radioactivity releases. If the sampling/monitoring systems were also underestimating airborne radioactivity concentrations, it is possible that the nuclear NESHAP is artificially restrictive, too difficult to comply with, or too difficult to measure.

Conclusions

The recently published nuclear National Emissions Standard for Hazardous Air Pollutants (10 mrem/year), is very restrictive and is apparently based more on DOE stack emissions data and dose calculations than on actual assessment of risks. 10 mrem per year from all site stacks combined is 10% or less of the annual dose commitment due to natural background radiation in all parts of the

United States. Demonstration of compliance with the NESHAP by validated measurement presents a technical challenge. Safety assessments for new or modified facilities to assure adequate air cleaning system design and margin of safety for compliance might indicate need for even higher quality air cleaning system components, and even more sensitive component test methods.

A systems Quality Assurance program which may help assure and verify compliance with the new nuclear NESHAP has been described and appears to be feasible. The QA program is based on the current DOE overall program which includes independent HEPA filter tests, but includes additional suggestions and concerns. The QA program, if adopted, can be readily modified to demonstrate compliance with the approximated 200 or more new non-nuclear NESHAPS to be established by EPA in the next ten years.

REFERENCES

(1) Private communication, Senator Jeff Bingaman (NM) to author 5/1/90.

(2) Summary Article, The Health Physics Society's Newsletter, June 1990.

DISCUSSION

FIRST: You have questioned whether current HEPA filter paper is adequate for monitoring very, very low concentrations of radionuclei. I have two comments. First, the minimum standard for papers is not necessarily the recorded performance of papers; it tends to be considerably higher. My second comment refers to your dissatisfaction with sampling papers that are only 99.97% efficient. I believe 99.97% efficiency will get almost everything that is there with negligible loss.

BRESSON: Although I said that the current standard is 99.97%, filters that are currently available, clean room filters, are better than that. For design of these facilities, we normally have applied some conservatism regarding how many filters in series are required to clean up the air to certain concentration guidelines. My point is that the concentration guidelines are being reduced significantly. That hasn't shown up yet in 10CFR20 or in associated documents, but it will now that the radiological NESHAP, a standard of 10 millirems to the general public, is an applicable dose limit. To comply with this limit, we must calculate air concentration guidelines that are going to be some small fraction of those that currently exist; that is what NESHAPs will do to us. My point is, when we design new facilities, the current HEPA filter efficiency standard may have to be lowered, or we will find ourselves with too many filters in series in order to meet our design requirements.

FIRST: You were talking about sampling filter papers and now you are discussing production facility. I don't think that is the same thing.

BRESSON: If I appeared to be talking about sampling filtration papers, I don't think that was the issue. The issue is the quality of filters, which has been good, may have to get better. In fact, they are better, and all I am saying is that we may want to make the standards even lower with respect to how good the cleanup devices must be in order to properly design systems. We now have different and more restrictive concentration guidelines to deal with.

PROGRESS REPORT ON THE IN-PLACE ACCEPTANCE TESTS OF WALK-IN PLENUM FILTER HOUSINGS WITH CLOSE COUPLE REDUNDANT SERIES FILTERS

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ABSTRACT

Tests were performed on injection and sampling grids in a 12,000 CFM experimental filtration system. These grid assemblies were designed using the guidelines in ASME N509 (1989). The injection assemblies were designed with the capability to selectively challenge portions of the filter bank. The sampling assemblies were capable of being either an upstream or downstream sampler, according to the testing situation. The injection grid exhibited some deficiencies in challenge agent mixing and concentration of aerosol produced during the test. The sampling grid produced representative samples of the challenge agent concentrations, both upstream and downstream, in the test system.

INTRODUCTION

Filter banks in close, serial arrangements present major obstacles to accurate in-place testing. Challenge agents injected at a single point between serial banks of filters do not provide an adequate challenge to all of the downstream filters when the distance between the serial banks of filters is short. This problem is compounded as the filtration system increases in size. A representative single point sample cannot be taken upstream of the second bank of filters in a series unless the challenge agent has been properly mixed prior to the bank. In order to ensure a uniform challenge of the downsteam bank, an injection grid is utilized. In order to take representative samples of the challenge and the penetrant, a sampling grid is utilized.

Description Of Test System

To test the effectiveness of the injection and sampling grid assemblies preliminary tests were conducted in an experimental filtration system constructed of plywood (Fig. 1).

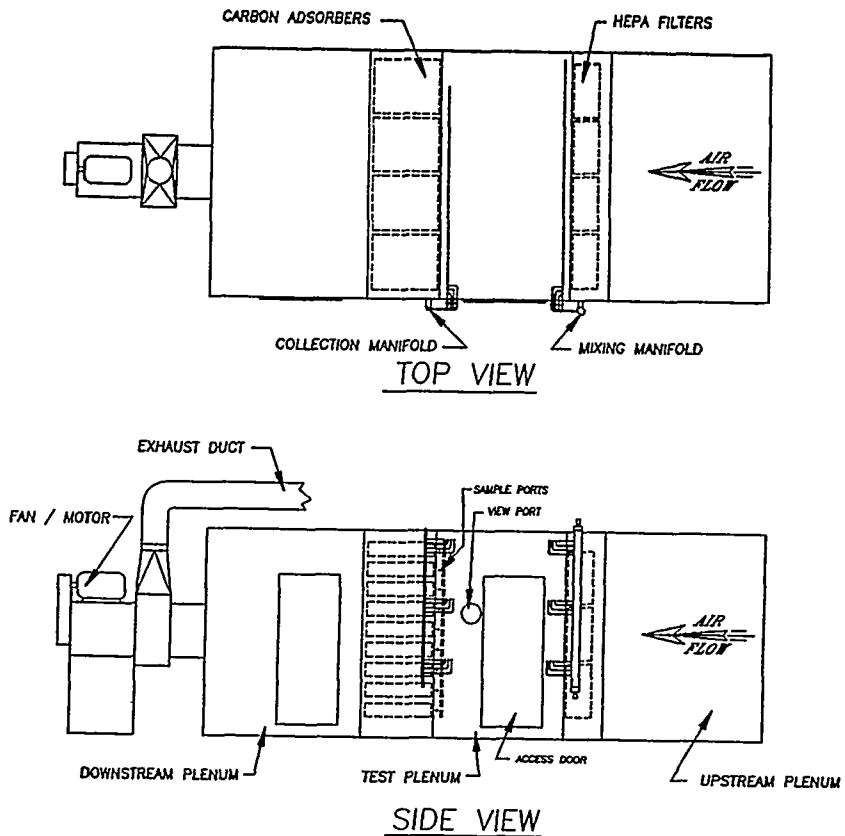

Fig. 1 Test System

The system was designed with an upstream plenum, HEPA filter bank (3H4W), test plenum, downstream carbon bank (36 Type II trays in a 9H4W array), downstream plenum, and fan/motor assembly. The fan was capable of producing a 12,000 CFM airflow through the filtration system. The entire unit was caulked with RTV sealant to prevent leaks into the system. The test plenum was 6 feet long in direction of airflow.

Injection Grid

The injection grid was designed so that challenge agent could be injected behind each filter in the HEPA filter bank. The injection grid was connected to a mixing manifold located outside the filtration system. The mixing manifold served the dual purpose as a point of connection for the aerosol generator and an accumulation chamber to ensure that a uniform concentration of challenge agent was available to all the injection grid supply lines. Each supply line was connected to the mixing manifold. It was possible to control the quantity of challenge agent, by the addition of valves on the supply lines.

Twelve 3/4" PVC supply lines ran from the mixing manifold to a position above the downstream side of each HEPA filter. The injection grid was composed of 3/4" PVC which ran from the supply line vertically on the downstream side of the HEPA filter. Six 1/2" PVC pipes extended horizontally from the vertical pipe to form the injection grid (Fig. 2).

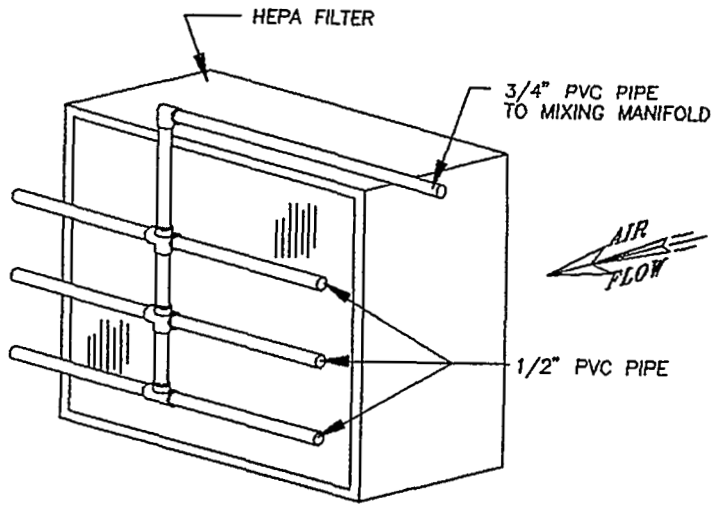

Fig 2 Injection Grid

A total of 42 holes 1/8" in diameter were drilled into the vertical and horizontal pipes of this injection grid. The holes were oriented at a 45° angle into the airflow. Each grid was located approximately six inches downstream of the HEPA filter.

Sample Grid

The sample grid was composed of 1/4" I.D. polyethylene tubing. Each set of 3 Type II adsorbers (24" x 24" area) was supplied with one vertical sample line located in the middle of the upstream face of the adsorbers. A 1/8" hole, positioned at the slot of each Type II tray, was drilled into the plastic tubing. Each of the sample lines (12 total) ran to a sample collection manifold located outside the filtration system. The collection manifold was designed with several sample points to determine if sampling position on the collection manifold had an effect on sampling readings.

The test system had a window and door, and had sample ports to allow conventional sampling with a sample probe from outside the housing.

TEST PROCEDURES

The tests were performed using Dioctyl Sebacate (DOS) aerosol as a challenge agent. The aerosol was produced by a portable, thermal aerosol generator. Aerosol concentration measurements were taken with a forward light scattering photometer.

A first test was performed to determine if the injection grid produced a uniform challenge across the face of the downstream filter bank in the serial filter arrangement. Several sets of aerosol measurements were taken approximately one inch upstream of the air inlet for each Type II adsorber (36 total). The average of these measurements was calculated and a +/- 20% range determined.

The uniformity measurements indicated that the injection grid did not produce aerosol uniformity within the +/- 20% range prescribed by ANSI/ASME N510. Comparing several sets of measurements revealed areas of consistently high or low concentrations of aerosol. Rearrangements of injection grid sections or change in the number or direction of holes in the grid did not change the aerosol concentration pattern. Since the uniformity could not be corrected by changing the injection grid, it was deduced that the high and low areas of aerosol concentrations were a product of varying pressure drops across the HEPA filters. A typical set of uniformity measurements are presented in Fig. 3.

TOP

78	75	70	70
85	75	85	85
96	92	96	95
70	100	95	85
△60	94	90	90
90	74	95	100
70	74	85	85
70	95	100	96
△65	100	100	100

FRONT

BACK

BOTTOM

$\bar{x}$ = 86

Range = 69-103

△ = Reading Outside Range

Fig 3 Typical Uniformity Measurements

The maximum concentration of challenge aerosol that could be produced through the injection grid was approximately 40%, even while using two aerosol generators. It was possible to increase the aerosol concentration readings by introducing a blower between the aerosol generator and the mixing manifold. Aerosol concentrations of 100% were capable using a blower and one aerosol generator.

A second test was performed to determine if the sample grid measurements of the aerosol concentration upstream of the second filter bank in the serial arrangement were representative. While the downstream bank of filters was being challenged with aerosol, measurements were taken with a sample probe upstream of the filter bank. Measurements of the aerosol concentration were also taken from the sample grid and compared to the sample probe measurements.

The sampling grid consistently produced representative samples of the aerosol concentration in the test system. The measurements taken using the sampling grid were normally within +/- 10% of the readings taken with a sample probe. Figure 4 shows comparisons of readings taken with a sample probe and through the test grid.

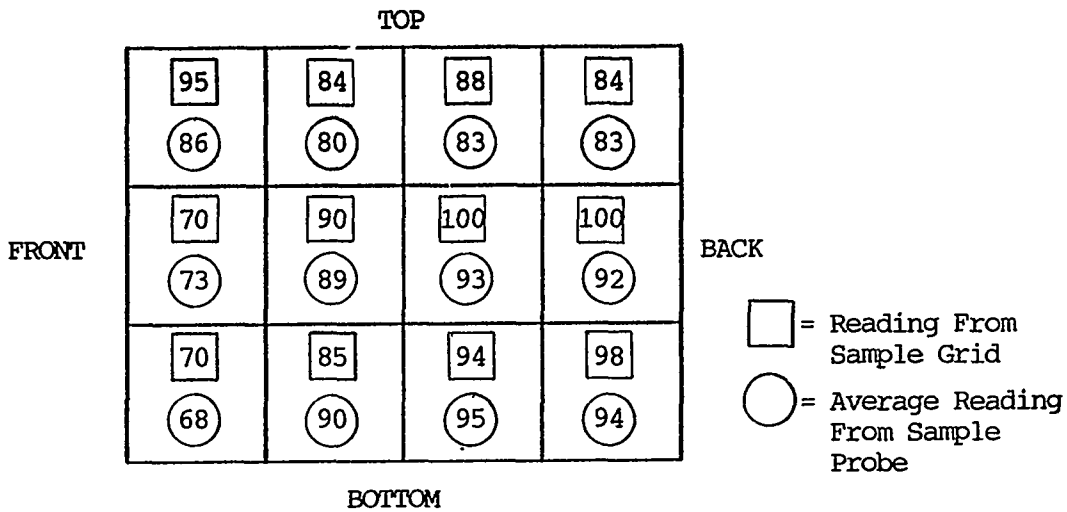

Fig 4 Comparisons Readings

A third test was performed to determine if the sample grid was capable of detecting a leak from the upstream filter bank. A leak was created in the upstream filter bank which allowed a small concentration of aerosol to bypass the bank. Measurements were taken across the face of the downstream filter bank with the sample probe and through the sample grid. Comparisons of the area from which the leak was originating, the actual concentration of aerosol upstream of the second filter bank, and the sample grid measurements were used to determine if it was possible to detect which upstream filter was leaking using only the sample grid.

When a small leak was generated in the upstream HEPA bank, it was possible to narrow down the area from which the leak was coming by using the sample grid. It was not possible to determine which filter was leaking when the leak was near the edge or corner of the HEPA filter. A leak through the middle of the HEPA filter could be distinguished.

CONCLUSION

The injection grid designed by using the N509 guidelines was tested by CSC. The grid would not produce an aerosol concentration which would meet the criteria of N510. The grid would perform as required with the addition of a "booster" blower to help inject the aerosol.

Valves installed outside the filter system, on the aerosol supply lines, could be used to regulate the aerosol.

The sample grid tested was adequate for taking upstream or downstream samples.

The sample grid tested could also be used to identify an area of leakage in a filter bank. The actual leaking filter could not be detected.

A system designed and constructed to the requirements of N509 would produce better airflow characteristics.

DISCUSSION

HANSON: In 1982 I gave I paper similar to the philosophy you have expressed. It had to do with a large filter bank at the Idaho Chemical Processing Plant where we used a different method for introducing smoke into a manifold system. We used a Penberthy compressed air jet pump and sucked off the smoke. We got very good dispersion. I think we had something like 5% variation. One other comment, we made the mistake of making the piping out of PVC and using DOP smoke. We came back a couple of weeks later and found the piping laying on the floor of the housing. It had all deteriorated. Maybe it will not do that with the material you used. What material did you use?

DUNBAR: I used PVC and it did the same sort of thing. I had a lot of trouble. I called it "condensation" at first, melted plastic.

FRANKLIN: You said you did not have an even concentration of the DOP upstream entering the filters due to, you stated, use of old filters. You then changed the pressure drop to get uniformity. Isn't that like knowing the answer and multiplying by a fudge factor to get the answer you wanted? I don't understand how you proved your manifolds were working.

DUNBAR: We were not achieving the uniformity that we thought we were going to achieve with this injection grid not the HEPA filters. We were testing the grid. So, we assumed something was wrong with the grid. We tried changing the hose, changing the position of the hose, changing the direction of the hose, but with everything we changed, we still got bad uniformity. Therefore, we concluded that the bad uniformity was not a result of the grid itself. The bad uniformity was a result of how the air was going through our test system. The test system was really not up to par. We did not really have sufficient sealing surface for the HEPA filters. If I had it to do over again, I would have used all brand new HEPA filters, but at the time we didn't have them. I believe that our bad uniformity was a result of that.

FRANKLIN: You still believe that you would get proper uniformity with that system?

DUNBAR: I believe we would. The sampler that we had was giving us really good results. When we measured in the sampling plane with samplers set side by side upstream and downstream we were getting almost the same readings a lot of the time. When we took measurements there and adjusted them to the uniformity we wanted you could assure yourself of a good challenge of the bank.

INVESTIGATION OF SALT LOADED HEPA FILTERS

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Abstract

The HEPA filters used in the ventilation system of the Waste Isolation Pilot Plant (WIPP) facility near Carlsbad, New Mexico potentially can collect large amounts of salt due to the salt aerosol generated by activities within the mine. Therefore, 0.61×0.61 m HEPA filters of the type used at WIPP were tested to determine the effects of salt loading on filter efficiency. In addition, flow resistance as a function of salt mass loading was determined. Tests were conducted at both high and low relative humidity conditions. For both high and low humidity the filter resistance increased at an increasing rate with filter weight gain. For a given weight increase the filter resistance was greater for the high humidity case. Filter efficiency was essentially unchanged for the low humidity case and always better than 99.99%. Salt loading with high humidity indicated a slight decrease in efficiency with values no lower than 99.98% at the highest loading.

Introduction

The WIPP facility, located southeast of Carlsbad , New Mexico, is a repository to demonstrate the safe disposal of transuranic wastes. The facility is located on and in a large salt formation. Presently 14.5 kilometers of horizontal shafts have been excavated. It is the excavation and the need to trim the shafts due to salt creep that is the principal source of the salt aerosol.

The ventilation system is capable of moving nearly 14,000 m^3/min (500,000 CFM) air, generating velocities more than sufficient to entrain large amounts of salt from the shaft floors. Normally air is discharged directly to the atmosphere without being filtered. In the case of an accident air flow is rerouted to banks of HEPA filters and the flow rate is reduced to 1700 m^3/min (60,000 CFM). Even at this reduced rate substantial amounts of salt areosol can be carried to the filter banks.

No known data existed on the behavior of HEPA filters subjected to salt loading. In particular, the hydroscopic nature of salt could result in significantly different results depending on the humidity levels. Therefore, a series of experiments was carried out that allowed HEPA filters to be loaded with salt in a controlled manner. Both low and high humidity cases were examined.

Experimental Arrangements

Filter Testing Facility

The filter testing was carried out at the Large Scale Ventilation Facility at New Mexico State University. A schematic diagram of the filter facility is shown in Figure 1. A 140 m^3/min

(5000 CFM) blower pulled air through the duct and filters. At the entrance to the duct a pre-filter was installed to avoid entrainment of particulates that would interfere with the depth of penetration (DOP) tests. In addition, a turbulence generator formed from nominal 1.5 cm tubing was placed just downstream of the pre-filter and served to further straighten the flow and enhance mixing. For the high humidity test a cooler pad saturated with water was placed upstream of the pre-filter. For the low humidity test two HEPA filters replaced the cooler pad. In addition a gas fired burner was used to reduce the ambient humidity levels.

Approximately 1 m downstream of the pre-filter the salt was injected. The injector consisted of four separate copper tubes. These tubes joined at a common pipe that was fed an aerosol of salt. The salt aerosol was generated using a BGI, Inc. Aerosol Generator. Between the injector and the filter being tested were 12 m of 0.61 × 0.61 m ducting. The filter itself was mounted to a null balance with an accuracy of ±4 grams. The balance could only be used after the filter was released from the adjoining upstream and downstream ducting. Real time filter weighing was not possible. Downstream of the filter were 7 m of duct that was connected to the blower. The blower discharged into 6 m of 20 cm diameter pipe. Approximately 2 m from the end of this final pipe was mounted a pitot probe for the determination of flow rates.

For the DOP efficiency test sampling probes were located 30 cm upstream and 240 cm downstream of the filter. The aerosol samples were analysed continuously using a Laser Aerosol Spectrometer built by Particle Measuring Systems, Inc.

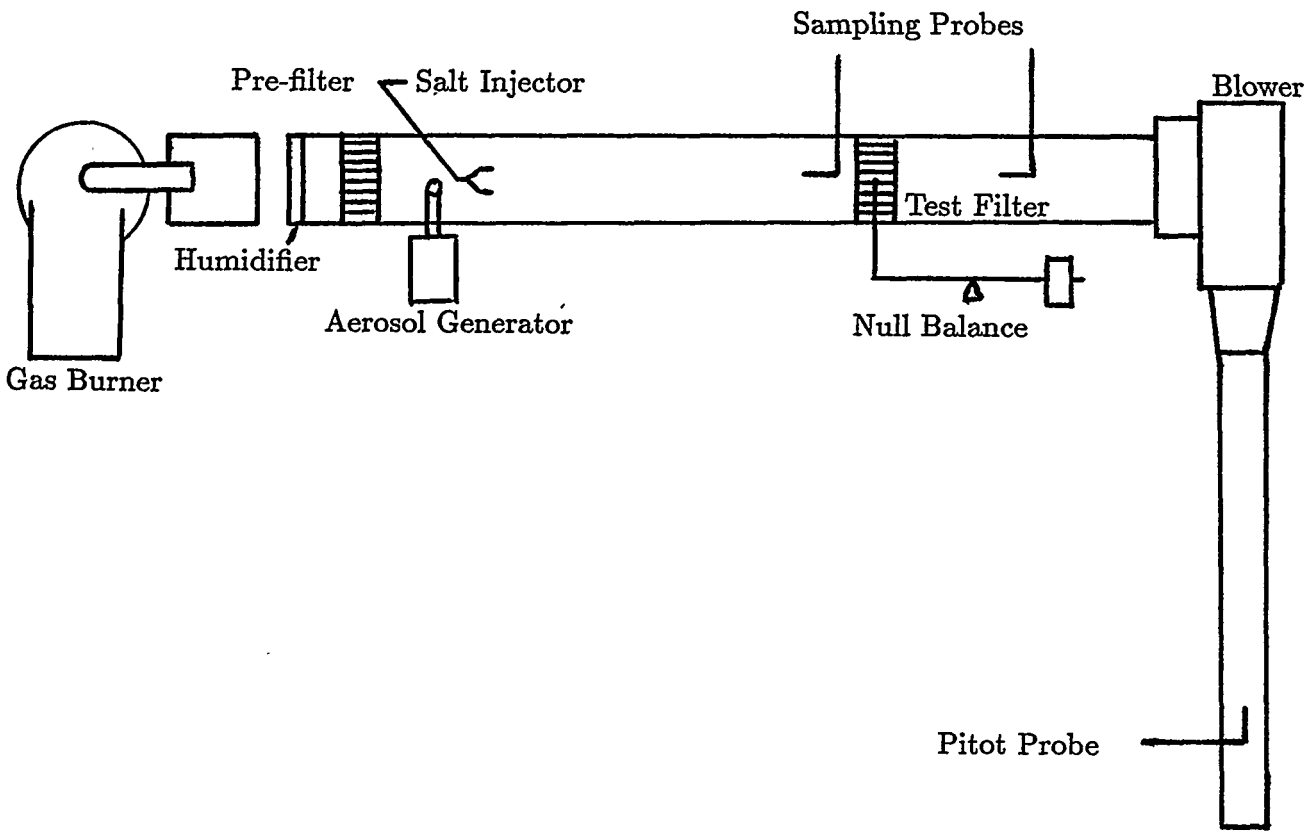

Figure 1. Schematic diagram of filter testing facility at New Mexico State University.

Test Filters

The HEPA filters used in this study were obtained from WIPP. The filters had been previously certified at Rocky Flats to have a DOP penetration of less than 0.01 percent. The filters were mounted in a stainless steel frame and, in addition, were equipped with a steel screen on both sides of the filter. The openings of the screen were 0.64 mm squares and the screen was positioned about 4.5 cm from the filter faces.

Experimental Procedure

High Humidity Case. To raise the humidity to the desired value a cooler pad was mounted at the duct inlet with a small amount of water dripped through it. A slide gate was placed in front of the cooler pad to control the flow rate. A Tycos Wet/Dry Bulb thermometer was used to determine relative humidity. Measurements were taken outside and inside the main building, and about 1 m upstream of the filter being tested.

Pretest investigations of salt aerosol generation revealed that plugging of the particle feeder was a problem. To solve this problem a 140 US standard seive was used to screen the feed salt. This screening process resulted in an upper size limit of about 105 microns. The salt exposed to the atmosphere was found to be absorbing water, and as a result would tend to ball up. This problem was particularly accute on humid days. A process of drying the salt with heat lamps up to the time it was placed in the feeder was adopted to overcome this problem.

The actual experimental run consisted of injecting salt until a significant increase in static pressure drop could be measured across the test filter. At this point the salt generator was turned off, humidity measurements were taken, and the water was shut off to the humidifier. Next the DOP generator was turned on to perform the filter efficiency tests. The DOP system would have to run about 45 minutes before the downstream count was sufficient to give accurate results. After the DOP test the blower was turned off, the filter was carefully unclamped from the ducting, and the mass gain of the filter measured. Lastly, the null balance was re-zeroed and the loading process was repeated.

Low Humidity Case. Ambient humidity levels were higher than desired; thus, it was necessary to heat the air being pulled through the duct. This was accomplished using a Houck gas fired burner. The cooler pad was removed and replaced by two HEPA filters sandwiched together. An additional 4.8 m of ducting was placed upstream of these filters but with a gap of about 20 cm between this ducting and the two HEPA filters. At the inlet of this added ducting the discharge of the Houck burner was placed. The burner was operated at a very low setting that resulted in air temperatures around $65°C$. The burner was not sealed to the ducting therefore air was entrained at the inlet as well as at the 20cm gap. The procedures for the low humidity case were the same as for the high humidity case except for the operation of the humidifier.

DOP Efficieny Determination A complete system for measuring particle size distributions from 0.03 to 3.0 microns was made available by Los Alamos National Laboratory. The unit was equipped with piping and valving so that samples could be obtained from upstream and downstream of the filter, and samples could be diluted in order to reduce the count rate to useable levels. The actual aerosol was generated from di octyl phthalate, a liquid. This liquid was placed in a container. A compressed air stream was passed through six small tubes placed in the bottom of the container. The air entrained liquid droplets and the resulting aerosol entered the ducting through the duct floor. The blower, set at $28\,m^3/min$ (1000CFM), carried the aerosol to the filter. A dilution factor was determined using the upstream sample tube only. Factors around 500 were consistently obtained. An actual penetration determination involved adjusting the aerosol generator to produce a reasonable count rate downstream of the test filter without any dilution. Upstream measurements were made with dilution and corrected with the previously determined dilution factor.

Experimental Results

High Humidity Case

The average relative humidity was around 80% with variations from 65 to 99%. Salt loading of the filter was continued until the filter weight had increased by about 5000 grams. As will be shown later a significant portion of this weight gain was water.

The pressure drop across the filter in inches of water is plotted versus filter weight gain in Figure 2. The trend in the data indicates an increasing slope with weight gain. This trend has been observed in previous work with other aerosol materials [1,2]. The data was fitted with a quadratic function for purpose of correlation. The scatter in the data is due almost exclusively to uncertainties in the filter weight, not in uncertainties in pressure drop. However, it should be stated that the null balance is not considered the source of the uncertainty, but rather the weight gain or loss of water.

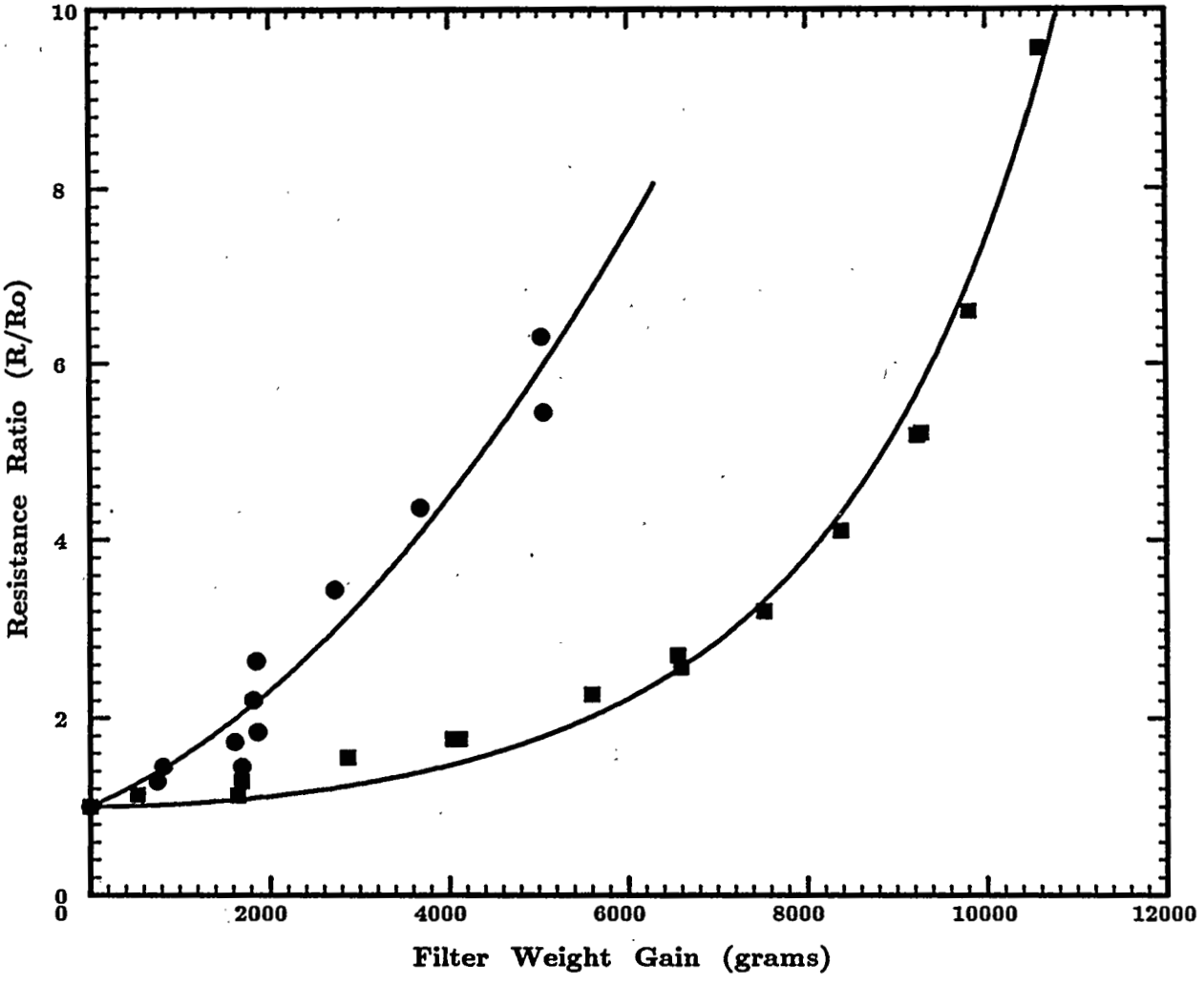

Figure 2. Pressure drop per unit flow rate versus total filter weight gain. Data have been fitted emperically. Squares are data from low humidity case, circles are from high humidity case.

The actual experiment spanned a period of about six days. Salt was injected on four of these days. The length of the experiment was due to the slow rate of salt accumulation on the test filter. As a result of stopping and starting the salt injection, weight loss was observed on four separate occasions. Two of these occasions were observed after the experiment was stopped overnight. Measurements taken the next morning indicated a loss of weight. In one case the loss was about 34 grams out of 1815 grams, in the other case the loss was about 190 grams out of 4870 grams. In both of these cases the humidifier was turned back on and an additional weight measurement was taken after 45 minutes. No salt was injected during these 45 minute periods. For the case in which 34 grams were lost the humidifying resulted in a gain of 46 grams. For the 190 gram loss case the gain was 386 grams. These results clearly indicate that moisture accounts for a non-negligable fraction of the total weight. The other two occasions of weight loss occurred after a DOP test was carried out. For this test the humidifier was shut off but the blower was left on. The drying action of the lower humidity ambient air is believed responsible for the weight loss.

The very last weighing indicated a loss of about 220 grams. Clearly, some loss mechanism was operating since salt was being injected. This mechanism may have been water dissolving salt on the filter screen and filter. When the filter was released from the duct it was observed that water was running down the upstream side of the filter. This water plus salt drained away from the filter and was not weighed. The reason for the presence of liquid water is not completely understood, but is believed to be the result of crystal formation. The injected salt particles absorb water on their surfaces as they are carried toward the filter. These particle fuse together to form the larger crystals that were observed on the filter screen. As the crystals are formed it is surmised that water is displaced from the particle surfaces. Eventually the amount of displaced water is so great that the salt can no longer hold it. At this point the water begins to run off dissolving some of the salt in the process.

Past filter loading work [1,2,3] with soot from polystyrene, PMMA, and wood indicated that filter resistance could be correlated to filter weight gain with a quadratic equation. The equation is of the form:

$$\frac{R}{R_0} = A M_s^2 + B M_s + C \qquad (1)$$

where A, B, and C are fitting constants, M_s is the filter weight gain, R is the filter pressure drop divided by flow rate, and R_0 is the value of R before any salt has been loaded onto the filter. The value of C was chosen to be unity so that the correlation would give the correct value for a clean filter. The values of A and B were determined by least squares to be 1.07×10^{-7} and 4.42×10^{-4} respectively. The quadratic form has no theoretical basis.

DOP Results. Filter penetration data is shown in Figure 3. With the exception of one point, filter efficiency was greater than 99.99% for all salt loadings. The one exception was the last value taken where liquid water was observed. For this point the filter efficiency decreased to 99.98%. A possible explanation for the decrease in efficiency is that water droplets were formed on or near the filter screen, passed through the filter and were counted downstream. Since the upstream probe would not sample any of these water droplets, the apparent filter efficiency would appear to decrease. Further work is required to test this hypothesis.

Low humidity Case

Ambient humidity levels were often in excess of 40 to 50%, considerably greater than the 15% desired. As discussed previously the gas burner was used to heat incoming air and reduce the relative humidity to the desired level. With this arrangement relative humidity in the duct was maintained between 9 and 18%.

Normalized flow resistance versus filter weight gain is shown in Figure 2. The lower humidity allowed more salt to be deposited on the filter than in the high humidity case. Toward the end of the experiment there was some salt loss from the filter screen and face when the filter

was separated from the duct. This loss resulted from unavoidable shaking of the filter which caused some of the built up salt to fall off. However, the amount of salt lost was small, around 130 grams.

The data in Figure 2. shows a smooth increase in pressure drop with filter weight gain. This is in contrast to the high humidity case where water loss and gain caused scatter in the data. As an indication of the importance of humidity changes in the low humidity case are the four double data points at filter weight gains of 1700, 4000, 6600, and 9300 grams. In each of these cases one of the values was taken at the end of a day and the other was taken first thing the next morning. At night the system was shut down completely. The small changes in weight and pressure drop show that stopping and starting the experiment had little effect on the results.

An equation quadratic in filter weight gain did not fit the data satisfactorily. A little experimentation with different functions showed that a two parameter exponential function could correlate the data quite well. The form chosen was:

$$\frac{R}{R_0} = exp(A M_s^B) \tag{2}$$

where the various parameters have the same meaning as before. A and B are fitting parameters with the following values chosen, $A = 1.264 \times 10^{-7}$ and $B = 1.8$. In fitting the data the value of B was chosen and then A was calculated to minimize the square error. The value of B is not the least-square value but is close. Again, it is stressed that the functions chosen are not derived from any filter models.

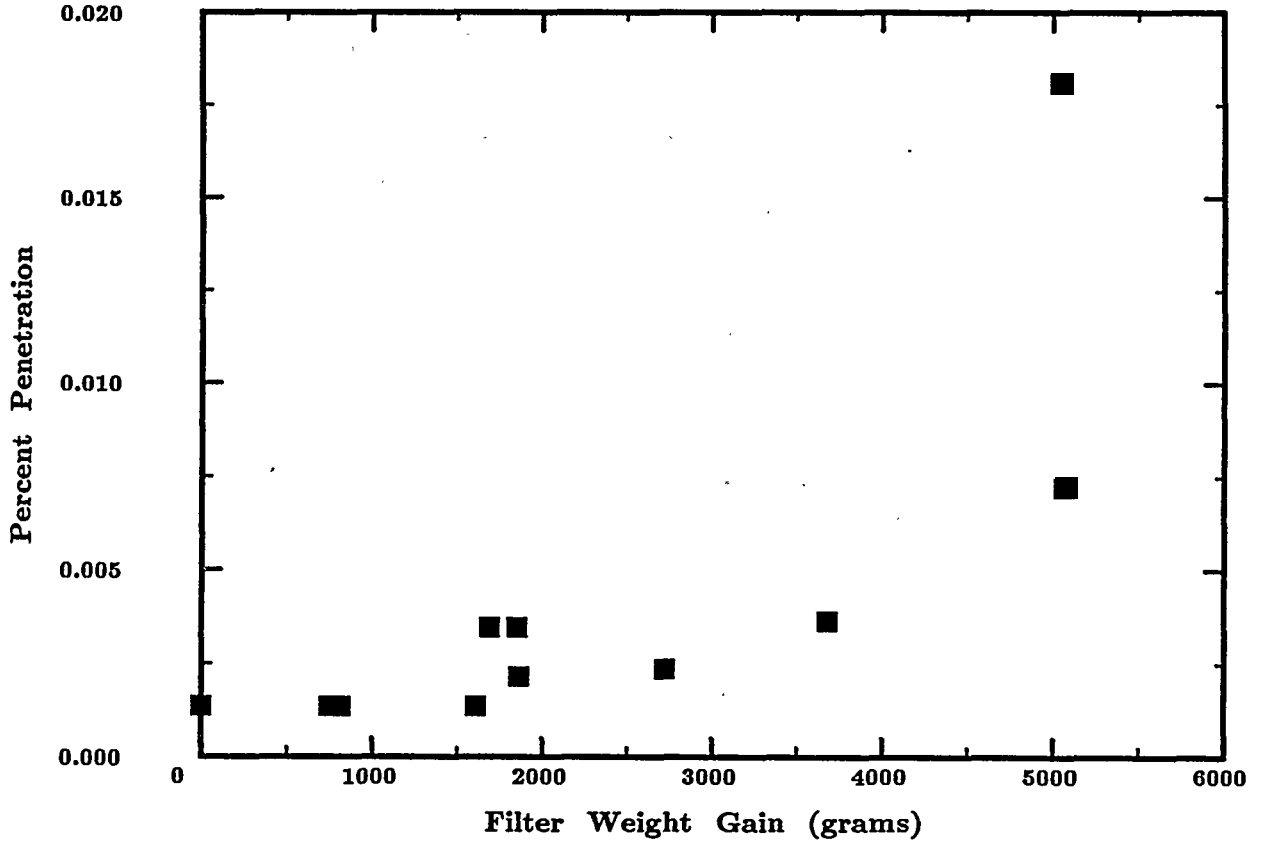

Figure 3. Percent penetration of 0.3 micron particles for high humidity conditions.

DOP Results. Filter penetration data is shown in Figure 4. For all loading levels the filter efficiency was greater than 99.99%. The trend in the data is somewhat unusual in that the penetration remains fairly constant to about 7000 grams, shows a sharp increase and then a decrease to near earlier values. No physical mechanism is obvious to explain these results. A possible explanation is scatter in the data as a result of the measurement technique. Note that the differences in filter efficiency are in the fifth significant figure, *i.e.* 99.99x%. At this time it is questionable whether the combined system of aerosol generator and aerosol particle sizer can provide accurate and repeatable five-significant-figure data. Since the efficiencies never fell below 99.99%, the precise explanation of the trend in the penetration data is considered of secondary importance.

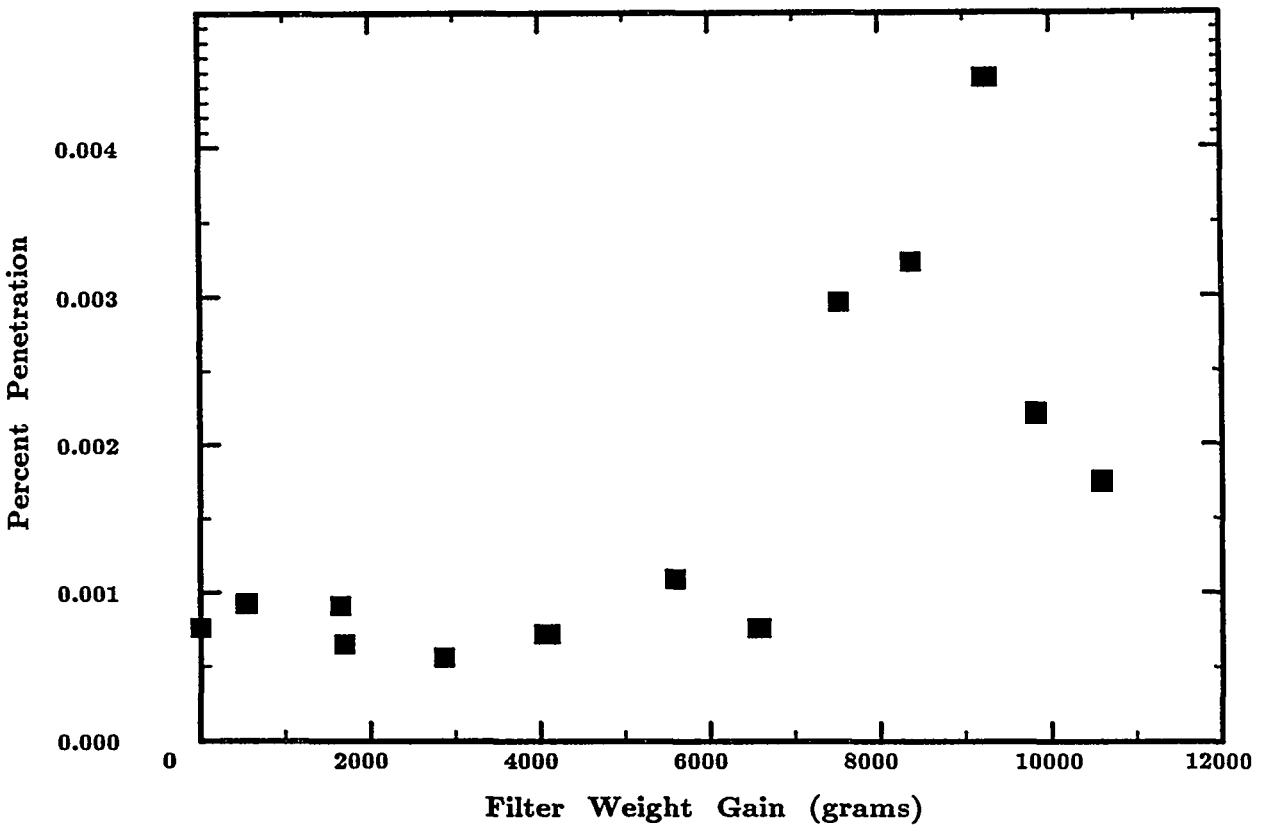

Figure 4. Percent penetration of 0.3 micron particles for low humidity conditions.

Discussion

Filter Loading

The resistance ratio results for salt show a clear difference between the low and high humidity cases. At present, the following explanation is put forward to explain the greater resistance of high humidity loading. Inspection of the screen and filter suggested that a very large fraction of the salt remains on the screen and the upstream face of the filter. In fact, for the high humidity case a portion of the screen was covered with an unbroken layer of salt. This sealing action is no doubt due to the small salt particles growing into larger crystals which in turn eventually fuse together. Salt that does penetrate into the filter is also expected to fuse to form larger crystals. With low humidity the salt particles do not fuse together (at least not to the same degree) and therefore present a more porous structure to the flow.

Further evidence for a majority of the salt being deposited on the filter screen and filter face comes from a comparison of the present results with the soot work of Fenton *et al.* [1] shown in Figure 5. The Figure 5. results are for polystyrene combustion. The two sets of curves are for high and low burning rates of polystyrene (PS). The upper curves are for the higher burning rates which produces smaller soot particles. These particles can penetrate the filter and cause plugging rather effectively. The low burnrate with larger soot particles tended to form a fragile cake on the filter face; hence, the HEPA filter was able to accumulate a larger amount of soot before plugging.

In both cases the mass median aerodynamic diameter of the soot was about 2 microns. For the salt the mass median diameter is larger. An estimate from sizing done at WIPP is 5 microns [4]. Although the differences in plugging behavior between soot and salt no doubt depend on more than just particle size, size does seem to be an important factor. If the low humidity salt case is compared to the the low burn rate PS case one sees that to reach a resistance ratio of 10 requires almost ten times as much salt as PS soot. If the comparison is to the high burn rate PS case then nearly thirty times as much salt is required.

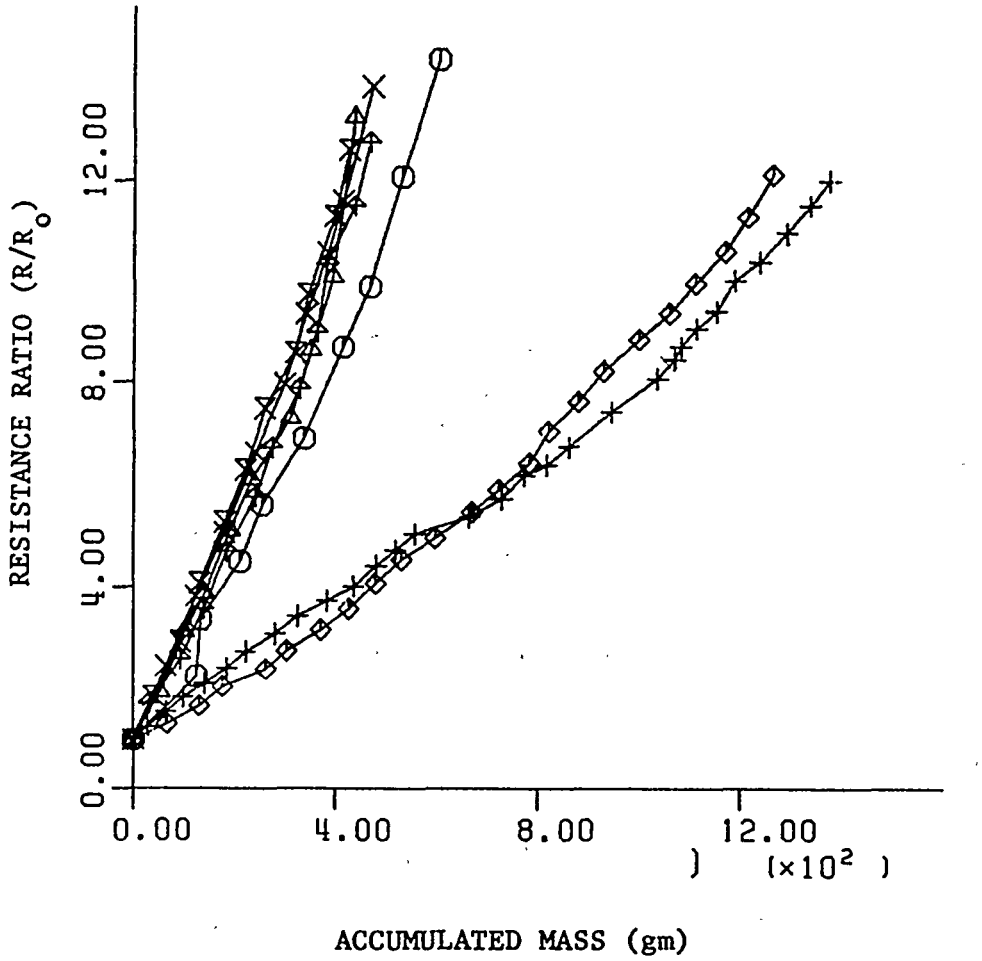

Figure 5. Variation of resistance ratio with accumulated soot mass for polystyrene combustion. Taken from Fenton *et al.* [1].

DOP Efficiency

Very little difference in percent penetration exists between the low and high humidity cases, especially if the comparison is made for weight gain between zero and 4000 grams. Above 4000 grams the high humidity case shows an increase in penetration. However, if the loss of around 200 grams for the last data point is viewed as an experimental error (from too much moisture), and instead of 200 a gram loss, there should have been a weight gain of several hundred grams, then the percent penetration appears to be rising exponentially. But, as stated earlier water drops may have formed at the filter from shear forces and been carried through the filter. Since there was no means to differentiate these particles from those originating at the aerosol generator, the percent penetration would be artificially high.

For the low humidity case the percent penetration rise above 7000 grams may be real, but, on the other hand, may be due to scatter in the data due to changes in aerosol feed rates or changes in the dilution factor for the particle sizer. At this time it is not known if the DOP system is good to five significant figures.

Conclusions

The effect of high humidity is to cause a greater resistance ratio for the same total weight gain as compared to low humidity loading. A tentitive hypothesis is that the presence of significant moisture causes the salt to fuse and more effectively plug the filter. The protective screen in front of the filter is important for the high humidity case since considerable salt buildup was observed there. Compared to soot loading of HEPA filters much more salt is required to achieve the same increase in resistance ratio. Anywhere from 10 to 30 times as much salt by weight is required.

In general, no significant degradation of the filters occur with respect to penetration of 0.3 micron particles. For the low humidity case the filter never had an efficiency less than 99.99%. For the high humidity case one DOP measurement resulted in an efficiency of 99.98%; however, this point as well as one other are potentially suspect because of the presence of liquid water on the filter.

Acknowledgement

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References

1. Fenton, D.L., Gunaji, M.V., Tang, P.K., and Martin, R.A., "Combustion Aerosol Laoding of Hepa Filters," Proceedings of European Conference on Gaseous Effluent Treatment in Nuclear Installations, Luxenbourg, October 14-18, 1985.

2. Gunaji, M.V., "Combustion Aerosol Loading of HEPA Filters," Master of Science Thesis, New Mexico State University, 1984.

3. Smith, P.R. and Leslie, I.H., Los Alamos National Laboratory report in preparation.

4. Newton, G.J., Shenk, K.D., Yin-Fong, S., Hsu-Chi, Y., Hoover, M.D., and Boecker, B.B., "Aerosol Studies for Evaluation of Isokinetic Sampling at the Waste Isolation Pilot Plant: Phase I, Measurements in the 14-ft Diameter Exhaust Shaft," Final Report from Lovelace Inhilation Toxicology Research Institute, July 30, 1987.

DISCUSSION

PARTHASARATHY: Have you had a chance to look at the actual filters at the site to see what kind of particle distribution you might have had on the filters?

SMITH, P.R.: The filters on the site have not been challenged yet by large doses of salt. They have only been tested for very brief periods of time so that the loading has been very minor. We have some of the filters in our laboratory and cannot see anything unusual about them. They do not look like they have been challenged at all. So, they have not had an accident were they actually have picked up much salt yet.

PARTHASARATHY: From what I understand they also have two banks of prefilters ahead of them.

SMITH, P.R.: They have one prefilter ahead of each HEPA filter. One of our next steps is to test them in series.

DORMAN: I would agree with the speaker that resistance rises markedly with high humidity, and as is noted, the curve for low humidity is rather different. Your curve for low humidity would depend very greatly on the particle size of the salt distribution. You have, in fact, noted this elsewhere. I think that Adley and Anderson gave some figures for resistance rise against particle size at the Oak Ridge Conference in 1964. On the other question, tests with high humidity show that pressure drop rises very rapidly at high humidities even when there are no particles there; at 95% R.H. the resistance goes up very rapidly. Sometime ago, I experimented with particles of latex and when we got over 95% humidity, latex particles penetration increased. I think it was because the pressure drop across the filter become so high it forced particles through. You do get an increase in penetration at very high humidities for particles that are not subject to solubility like salt.

SMITH, P.R.: We did see some evidence of that but it was sort of down in the fifth decimal place and we did not want to make a big point of it. At high humidity we did see some decrease in the efficiency, but the filter was still 99.99%, but out on that fifth point it was rising a little bit.

PHOTOCHEMICAL REMOVAL OF RADIOACTIVE IODINE FROM AIR

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Abstract

An advanced method of removing both organic (RI) and inorganic (I_2, HI) forms of radioactive iodine from air, developed by AECL Research is reviewed. The method employs ultraviolet light (200-300 nm) to convert RI and HI to molecular iodine (I_2) and the corresponding oxidized forms of the free radical R (alcohols, aldehydes, and ketones) and the hydrogen atom H (H_2O). Molecular iodine can be further converted by reaction with ozone to iodine oxide, solids which can react with water vapour to form solid and nonvolatile iodine oxyacids or, in the presence of water, dissolve to form nonvolatile iodates. This paper discusses applications of the photochemical method to the construction of scrubbers that can be used to remove volatile radioiodine in air inside a reactor containment or in air released during venting of a reactor containment following an accident.

I. Introduction

In certain severe nuclear accidents, an excessive pressure may build up inside the reactor containment as a result of substantial steam and/or hydrogen production. Excessive pressures may lead to containment failure or uncontrolled leakage of radioactivity. To prevent these undesirable events, in some accident management scenarios, the reactor containment is vented through an engineered system [1,2], designed to filter out radioactivity- laden aerosols and, in some cases, radioactive iodine, which is one of the most hazardous radionuclide that could be released in an accident. Cesium iodide (CsI) aerosols, molecular iodine (I_2), and hydrogen iodide (HI) are removed easily by charcoals as well as a number of other sorbents and scrubbing solutions [3,4]. However, deep beds of charcoal, impregnated with triethylene diamine (TEDA) and/or KI, are required to remove the more penetrating organic iodides. Such charcoals must be frequently monitored and periodically replaced to ensure availability in an accident. In the severe post-accident environment (high radiation fields, high temperature and relative humidity), the performance of impregnated charcoal with respect to organic iodide removal could suddenly be substantially degraded because of poisoning effects [5]. Thus, for emergency filter applications, it is important to find more reliable alternatives.

In this paper, we are exploring the technical and economic feasibility of a photochemical system for removing organic and inorganic radioiodines during venting of the reactor containment following an accident. The system under consideration uses

ultraviolet light to convert organic iodides to molecular iodine. The iodine then reacts with ozone to form solid iodine oxyacids, which can be removed from air with aqueous scrubbers or particulate filtration techniques [6]. Alternatively, fixation of iodine by reaction with ozone can be eliminated and, once the organic iodides are converted to molecular iodine, the latter can be easily removed by even substantially degraded charcoal filters. The proposed system would increase the efficiency and long-term reliability of charcoal filters used in emergency applications and, where venting systems are not equipped with iodine scrubbers, would provide an effective means of radioactive iodine removal. A transportable version of the proposed scrubber is also possible and would have the added advantage of being well maintained in a central location serving more than one reactor. In an emergency, the transportable scrubber would be available for dispatch to the impaired reactor, which, of course, would be equipped with the appropriate linkages for venting the containment.

II. Photochemical Conversion of Organic Iodides

The theory of operation of a photochemical scrubber has been presented elsewhere [6], and thus, we review here only the key elements. Organic iodides (RI) absorb light in the 200-300 nm wavelength region and decompose at the C-I bond to form iodine atoms and organic radicals,

$$RI + h\nu \longrightarrow R + I \ . \qquad (1)$$

Depending on the conditions, a number of reactions may follow the above primary process. However, for low concentrations of organic iodides in air, as in the present context, the only significant reactions would be the oxidation of the organic radicals to form aldehydes and ketones,

$$R + O_2 \longrightarrow \text{oxidation products}, \qquad (2)$$

and the recombination of iodine atoms, which is assisted by air acting as a third body (M), to form molecular iodine,

$$I + I + M \longrightarrow I_2 + M \ . \qquad (3)$$

The rate of photochemical decomposition of organic iodides is given by

$$dC/dt = -\sigma IC \qquad (4)$$

where, C is the concentration of the absorbing species (RI), σ is the absorption cross-section, and I is the incident photon flux. For a steady-state tubular-flow reactor, Equation (4) can be integrated to provide the decontamination factor (DF),

$$\ln DF \equiv \ln C_{in}/C_{out} = \sigma IV/F \quad (5)$$

where

DF = ratio of inlet to outlet concentrations of RI, C_{in}/C_{out},
V = volume of the photochemical reactor,
F = volumetric flow rate,
I = average photon flux in the photochemical reactor, and
σ = absorption cross section.

In most photochemical systems, the light intensity varies significantly throughout the reaction vessel because of the presence of relatively high concentrations of absorbing species. Thus describing the extent of reaction is complex. In the proposed system, however, the absorbing species are present in very low concentrations (parts per million or less). Furthermore, the inside of the photochemical reactor will be an ultraviolet-light reflecting surface, such as aluminum and on average, a photon will be reflected by the reactor walls five to ten times before being absorbed by the wall or by an organic iodide molecule. Hence, the intensity of photon flux can be considered constant throughout the reaction vessel, independent of the distance from the lamps, and the average light intensity can be determined using a photon balance.

At steady state, the lamp output equals the rate of absorption by organic iodides plus the rate of absorption by the walls,

$$n2\pi r I_o l = \pi(R^2 - nr^2)\,\Sigma\, C_i\sigma_i\, LI + 2\pi R(1 - K)LI + 2\pi R^2(1 - K)I \quad . \quad (6)$$

Thus,

$$I = nrI_o l/[\pi(R^2 - nr^2)L\,(\Sigma\, C_i\sigma_i) + R(1 - K)(L + R)] \quad (7)$$

where

I_o = flux of photons in the proper energy range at the lamp surface,
r = radius of the cylindrical lamps,
l = arc length of the lamp,
n = number of lamps,
L = length of the cylindrical reaction vessel,
R = radius of the cylindrical reaction vessel,
C_i = concentrations of organic iodides within the reactor, and
K = fraction of the photons reflected by the reaction vessel walls.

Combining Equations (5) and (7) gives equation (8),

$$\ln DF = \frac{\sigma V n r l I_o}{FI[\pi(R^2 - nr^2)L\ (\Sigma\ C_i \sigma_i) + R\ (1 - K)(L + R)]} . \qquad (8)$$

Because the materials in the reaction vessel compared with the reaction vessel walls, absorb an insignificant fraction of the photons, $\pi(R^2 - nr^2)L\ (\Sigma\ C_i \sigma_i) \ll R\ (1 - K)(L + R)$. Also, for a cylindrical reactor $V = \pi R^2 L$. Thus,

$$\ln DF = \frac{\sigma \pi n r l I_o RL}{F\ (1 - K)\ (L + R)} . \qquad (9)$$

Equation (9) was used to predict the performance of the conceptual designs presented below. Of course, ultimately, an empirical evaluation derived from pilot-scale studies will be necessary. As noted above, a number of assumptions were made in deriving Equation (9); the most important is the assumption that the flow within the reaction vessel is tubular. Conservation of photons and lamp geometry, however, dictated maximizing the reaction vessel radius and hence using a cylindrical rather than a tubular reaction vessel. Fortunately, in the proposed system, the photon flux will not vary significantly within the reaction vessel and there will be no necessity to induce mixing of reactants. It is assumed that near-tubular flow can be achieved through transparent baffles used to minimize mixing while maximizing the effective reaction vessel volume.

III. Fixation of Iodine by Reaction with Ozone

Molecular iodine generated by the photochemical conversion of organic iodides, or already present in the air to be decontaminated, can be reacted with ozone to form solid I_4O_9 at temperatures less than about 370 K, or solid I_2O_5 at higher temperatures [7]. In humid atmospheres, both of these oxides react with water to form solid iodine oxyacids (HIO_3, HI_3O_8),

$$I_2 + O_3 \xrightarrow{<370\ K} I_4O_9 \xrightarrow{H_2O} HI_xO_y \qquad (10)$$

$$I_2 + O_3 \xrightarrow{>370\ K} I_2O_5 \xrightarrow{H_2O} HI_xO_y . \qquad (11)$$

The rate-determining step in the above reactions is the bimolecular reaction of iodine with ozone, which obeys the rate law

$$-d(I_2)/dt = -S^{-1}\, d(O_3)/dt = k(I_2)(O_3) \qquad (12)$$

where k is the rate constant and S = 3.9 ± 0.2 is the reaction stoichiometric factor. The rate constants in the temperature range 293 K to 370 K fit the Arrhenius equation, $k = A \exp(-E/RT)$, where the pre-exponential factor A is equal to $10^{7.76 \pm 0.17}$ $dm^3 \cdot mol^{-1} \cdot s^{-1}$ and the activation energy E is equal to 25.0 ± 1.2 $kJ \cdot mol^{-1}$.

Under conditions of excess ozone, Equation (12) can be integrated to give

$$\ln DF = \ln[(I_2)_{in}/(I_2)_{out}] = k(O_3)t \qquad (13)$$

where $(I_2)_{in}$ and $(I_2)_{out}$ are the inlet and outlet concentrations for a scrubber and t is the reaction time. Typical ozone concentrations required to achieve a DF of 1000 at various temperatures are given in Table 1. Such ozone concentrations can be generated using commercial ozone generators [8].

Table 1 Ozone Requirement for I_2 Removal (DF = 1000) as a Function of Flow Rate and Temperature

Flow rate (m^3/s)	Temperature (°C)	Required O_3 (%)
1	50	0.21
1	100	0.075
1	150	0.035
0.6	50	0.12
0.6	100	0.045
0.6	150	0.021

IV. Photochemical Systems

Two conceptual systems for photochemical abatement of radioiodine are described below. The first, SPARGE (System for Photochemical Abatement of Radioiodine in Gaseous Effluent), is for decontamination of air released from the reactor containment to the environment. The second, SPARC (System for Photochemical Abatement of Radioiodine in Containment), is for decontamination of air within confined spaces in a reactor containment.

SPARGE

SPARGE is shown in Figure 1. It consists of a 15-m^3 cylindrical reaction vessel (3.0 m in diameter and 2.2 m in height) containing 12-kW medium-pressure mercury lamps. These lamps emit about 400 W of ultraviolet light within the 260-nm absorption band of organic iodides. Each lamp is mounted in two concentric quartz

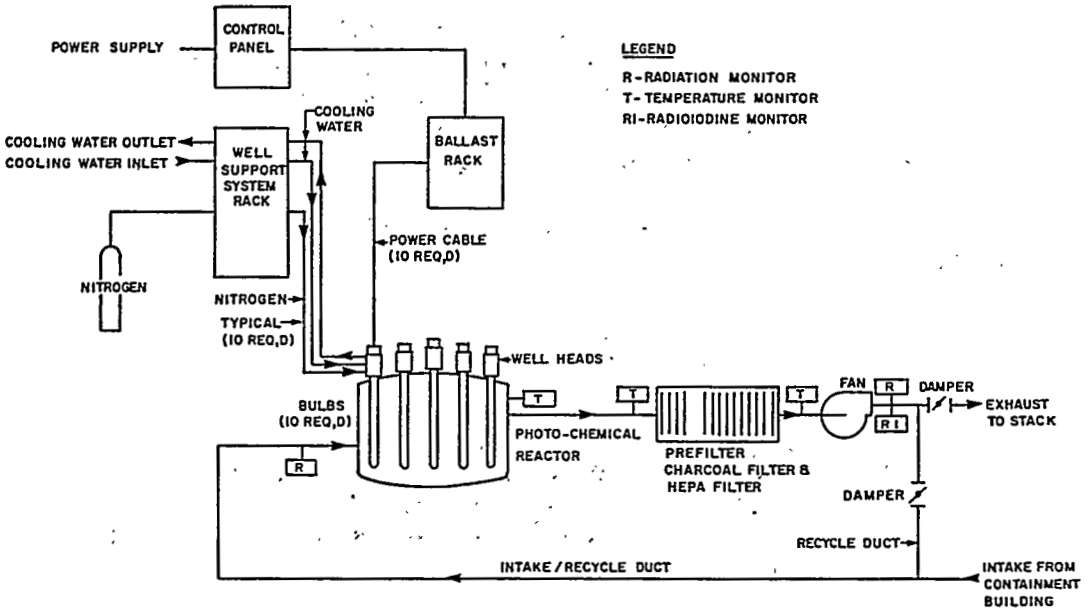

FIGURE 1. Conceptual Flow Sheet of SPARGE

wells used for cooling the lamps through continuous flows of water and nitrogen gas. The inside of the steel reactor vessel is coated with polished aluminum having a high reflectivity. SPARGE also includes a blower, a support system and transformers for the lamps, and a separate control system. It is designed for flow rates of up to 1000 $dm^3 \cdot s^{-1}$ (STP) air. As shown in Table 2, the extent of decontamination depends on the photochemical conversion of the organic iodides, which is a function of the light intensity, the reflectivity of the reaction vessel and the flow rate.

Table 2 Photochemical Conversion of CH_3I to I_2 in SPARGE as a Function of Key Variables

Flow rate (m^3/s)	Number of lamps	Reflectivity (%)	Conversion factor
0.6	15	90	3150
0.6	15	80	60
0.6	10	90	215
0.6	10	80	15
1	15	90	125
1	15	80	10
1	10	90	25
1	10	80	5

A version of SPARGE that includes a charcoal filter is equipped with 60 one-inch beds containing a total of 150 kg of TEDA-impregnated charcoal. This system is capable of accommodating flows of up to 2500 $dm^3 \cdot s^{-1}$ and is capable of removing over 99.9% of the molecular iodine. This charcoal, when new, is capable of removing over 99% of the organic iodides, but, with time, its efficiency deteriorates. Consequently, removal of organic iodides by charcoal is not credited.

SPARGE can be built as a permanent module of a filtered air vent or it can be mounted on a truck, as shown in Figure 2, for transportation and linkage to an impaired reactor. The weight of the transportable version is predominantly due to the transformers and radiation shielding and is estimated to be about 15 t. The total cost of either version of SPARGE is estimated at about $1000 000.

SPARC

SPARC is a mobile system designed for use in confined spaces in containment such as the fuel bay or the reactor vault. It consists of a 1- to 1.5-m^3 reactor vessel containing two 7.6-kW medium-pressure mercury lamps. In this design, two lamps are included for reliability although only one would be used at any one time. We estimate that SPARC can remove 90% of the gas phase organic iodine in a well-mixed 100-m^3 space in about one hour using the design flow rate of 500 $dm^3 \cdot s^{-1}$.

As shown in Figure 3, SPARC together with its own blower and support system would be mounted on a trolley. For operation, SPARC could be towed into the confined space and connected to available sources of cooling water and electrical power, using a robotic system if entry by workers is prohibited by a high level of contamination. SPARC would draw air from the room past the photochemical scrubber and then discharge it back into the room. Operation of the system would be by remote control. During operation, the radioiodine levels in the air entering and leaving the system would be monitored as would the temperatures of the air in the photochemical reactor. The cost of SPARC is estimated to be about $250 000 (Cdn).

V. Conclusions

The underlying principles of a photochemical method of radioiodine abatement were reviewed and used to estimate the performance of conceptual photochemical scrubbers that can be used in reactor accidents. Two systems have been identified: SPARGE, which can be used in series with an engineered system for venting the containment, and SPARC, which is a mobile system suitable for decontaminating confined spaces within containment. Both concepts appear attractive but require further verification at the pilot-scale level.

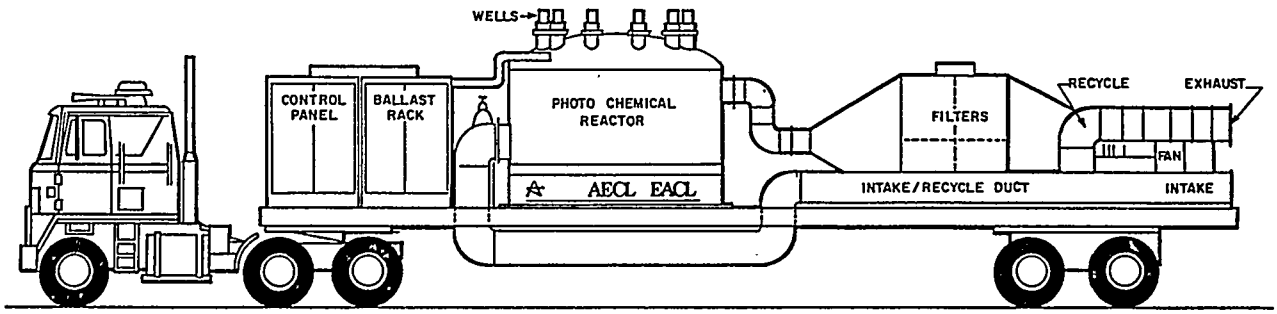

FIGURE 2. Conceptual Design of Transportable Photochemical Scrubber (SPARGE).

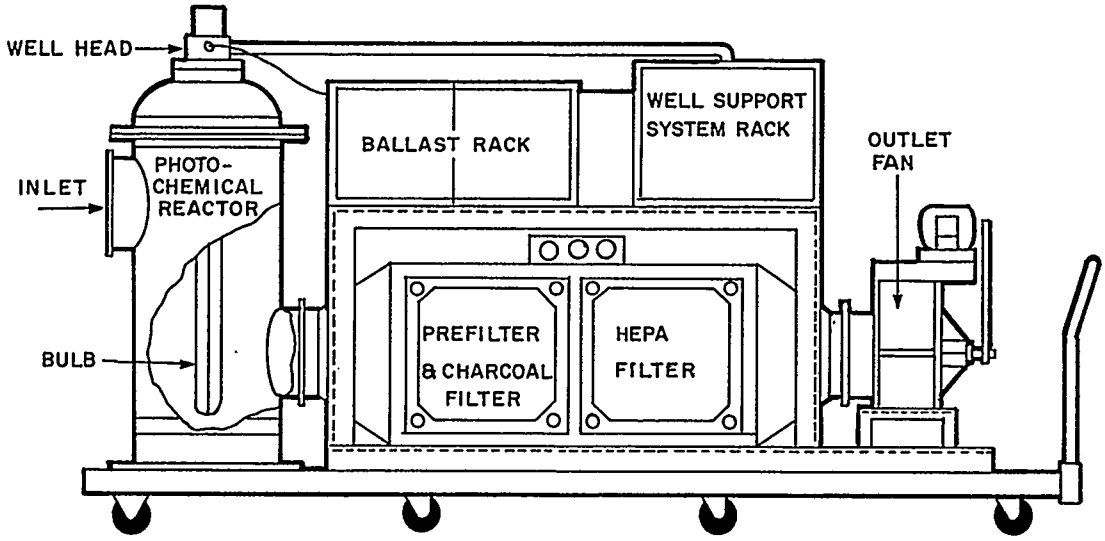

FIGURE 3. Conceptual Design of Mobile Photochemical Scrubber (SPARC).

VI. References

(1) Jefford, J.D. and Fluke, R.J., "Experimental data required for the design and analysis of emergency filtered air discharge systems", Proc. 16th DOE Nuclear Air Cleaning Conference, Vol. 1, 225 (1981).

(2) OECD/NEA, Proceedings of Specialist Meeting on Filtered Containment Venting Systems, CSNI Report 148, Paris, 1988.

(3) "Radioiodine removal in nuclear facilities, methods and techniques for normal and emergency situations", IAEA Tech. Rep. Ser. No. 201, Vienna (1980).

(4) Kovach, J.L., "The evolution and current state of radioiodine control", Proc. 16th DOE Nuclear Air Cleaning Conference, Vol. 1, 417 (1981).

(5) Wren, J.C., Moore, C.J. and Vikis, A.C., "A study of the performance of charcoal filters under post-LOCA conditions", Proc. 20th DOE Nuclear Air Cleaning Conference, Vol. 1, 786 (1989).

(6) Vikis, A.C., "The photochemical method for radioiodine abatement", Atomic Energy of Canada Limited Report, AECL-7819 (1984).

(7) Vikis, A.C. and MacFarlane, R., "Reaction of iodine with ozone in the gas phase", J. Phys. Chem. 89, 812 (1985).

(8) "Ozone chemistry and technology", Advances in Chemistry, Series 21, American Chemical Society, Washington, 1959.

DISCUSSION

EVANS, A.G.: Do you anticipate trying to use charcoal in conjunction with this? One of the hazards of ozone and charcoal is an explosive mixture.

VIKIS: Yes, that is quite correct. In such a situation one may use a sliver coated sorbent. In the case where ozone will be used there is no need at all for using charcoal. If we use ozone that would take us directly to the iodine oxide which can be captured on a particulate filter. If one uses charcoal, there is no need to use ozone or to generate ozone. However, note that if you only wish to convert organic iodides to elemental iodine, which you can capture on charcoal, then you should use ultraviolet light sources that have a vycor jacket. The latter lamps do not emit below 220 mm, thus, they do not generate ozone.

REVIEW OF DEPARTMENT OF ENERGY FILTER TEST FACILITY AND FILTER TEST FACILITY TECHNICAL SUPPORT GROUP ACTIVITIES FY 1986 - FY 1989

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Abstract

All nuclear-grade High Efficiency Particulate Air (HEPA) filters in critical health and environment protection applications at the Department of Energy (DOE) nuclear facilities are quality assurance tested at one of the three DOE Filter Test Facilities (FTFs). Test activities at the FTFs have been governed for six years under a unified set of DOE operating standards. The standards specify an annual review of the FTF test operations and technical consultation performed by the FTF Technical Support Group (TSG).

Los Alamos National Laboratory has been commissioned as the FTF TSG. One of the tasks of the FTF TSG is to compile integrated annual reports based on the semi-annual filter acceptance/rejection reports submitted by the FTFs. The objective of the annual report is to provide information on the performance and quality of High Efficiency Particulate Air (HEPA) filters being acquired and installed at DOE nuclear facilities. Data and conclusions from Fiscal Years (FY) 1986-1989 annual reports are discussed.

From FY 1981 and continuing through FY 1985, the Airborne Waste Management Program Office funded a project at Los Alamos to develop an aerosol test system suitable for use at the three FTFs as a potential replacement for the current di (2-ethylhexyl) phthlate (DEHP) Q107 test system. In response to the DOE's request, Los Alamos developed the High Flow Alternative Test System (HFATS). This system has undergone extensive laboratory prooftesting, operational evaluation, and technology transfer. Results of these evaluations have been published in the Air Cleaning Conference proceedings. The status of implementing and obtaining DOE approval of the HFATS at the three DOE FTFs is discussed.

I. Introduction

The Filter Test Facilities (FTFs) activities to include component qualification testing for FY 1986 - FY 1989, the status of the High Flow Alternative Test System (HFATS)(2,3,4) and the cooperative efforts between the Department of Energy (DOE) and Department of Defense (DOD) in modernization of filter test equipment will be discussed.

II. Filter Test Facility Activities

The FTFs provide 100% quality assurance inspection and testing of HEPA filters procured and used in DOE facilities. The performance of HEPA filters becomes exceptionally critical as more stringent requirements are imposed on DOE facilities(5).

Test activities at the FTFs have been governed for six years under a unified set of nuclear standards(6). The four DOE standards are designated at Nuclear Energy (NE) Standards published by the Performance Assurance Project Office, Oak Ridge National Laboratory. The standards include an Operating Policy of the DOE Filter Test Program(7), Quality Assurance Testing of HEPA Filters(8), DOE Filter Test Facilities Quality Program Plan(9) and Specification for HEPA Filters Used by DOE Contractors(10). The DOE NE F 3-42 through NE F 3-45 standards are currently being reviewed for incorporation as mandatory standards in DOE Order 6430.1, General Design Criteria Manual. The standards have enhanced the operation of the FTFs.

The NE F standards specify component qualification testing. The manufacturer qualification testing is destructive testing performed on the size 5 filter units(10,11) and includes the rough handling test to simulate severe handling conditions, the overpressure resistance test to simulate the pressure and moisture that an installed HEPA filter would experience in the event that the deluge system was activated and the heated air test which simulates the heat that a filter would experience in the event of a fire. Section 7.2 of NE F 3-45(6) provides three specific alternatives for component qualification testing:

1. Manufacturer may qualify the design and components of a HEPA filter through Edgewood Quality Products List (QPL)testing program(8).

2. Manufacturers may perform the qualification testing in house provided that the manufacturers' equipment has been approved for qualification testing.

3. Manufacturers may qualify the HEPA filter design through The DOE at the Rocky Flats FTF.

It is DOE's intent to periodically select stock filters and perform the three specific component qualification tests. To date, the Rocky Flats FTF has supported the component qualification testing in accordance with the NE F Standards and has tested a total of thirty (30) filter model types representing four (4) manufacturers. Further testing is required to substantiate specific manufacturers' filter model types. The intent of DOE is not to publish a Quality Products List but rather continue to have updated component qualification testing data to assist procurement and FTF personnel. Initially, this will require a coordinated effort between the three FTFs with DOE support to procure and test the representative filter model types for all DOE facilities.

Component qualification test results indicate that 16.6% of the total number of manufacturers' filter model types rejected were attributed to Heated Air Test failure, 16.6% for Overpressure Resistance failure, 50% from Rough Handling and 16.6% rejected for more than one of the three tests (5.5 for overpressure resistance and rough handling and 11.1% for Rough Handling and Resistance to Heated Air.)

III. FTF Technical Support Group

The NE F Standards specify an annual review of the FTF operations and technical consultation performed by the FTF Technical Support Group. Los Alamos National Laboratory has been commissioned as the FTF TSG. The FTF TSG provides technical assistance to the FTFs through continuing activities to ensure consistency of measurements at each FTF and to ensure consistency of measurements among the FTFs. The FTF TSG assists in solving technical problems that arise at the FTFs.

The FTF TSG program includes coordination of the Round Robin Testing, summary of the three FTFs acceptance/rejection data in the form of annual report, support in the installation, proof testing and training for the High Flow Alternative Test System (HFATS), Low Flow Alternative Test System (LFATS) and coordinated efforts with DOE and DOD in the modernization of filter test equipment.

Round Robin Testing (RRT)

The purpose of the RRT program is to maintain and improve the consistency of FTF measurements. In 1982, the RRT was initiated by Cliff Burchsted, Oak Ridge National Laboratory. The results of the RRT indicated possible problems in comparability of penetration and pressure drop measurements. The program responsibility was transferred to Los Alamos in March of 1984. The RRT program is in accordance with Section 10 of NE F 3-43(8), is conducted twice each fiscal year and a total of 13 RRT have been conducted to date.

Semi-annual Report Summaries

The task of compiling an integrated annual report based on the semi-annual reports of the three FTFs is assigned to the FTF TSG. The objective of the annual reports are to:

1. Provide a reference frame against which performance statistics of individual FTFs can be evaluated.

2. Provide information on the performance of filters being procured and used in DOE facilities for use in ventilation exhaust and other systems.

There is a wealth of information provided by the FTF semi-annual reports and for purposes of this paper the summary of the annual filter test results, filter test results by FTF, test results grouped by the filter manufacturer and number of filters tested by size for FY 1986 - FY 1989 will be discussed.

Table 1 illustrates the annual filter test results for the FY 1986 - FY 1989. Please note that the highest number of filters were tested in FY 1986, decreasing in FY 1988 and a return increase in FY 1989. The category, number of filters accepted with waiver fluctuates from a high of 6.3% in FY 1988 and to a low of 1.1% in FY 1989. The acceptance with waiver or repair category includes those filter units that have been waived from specification requirements excluding penetration. Acceptance with waiver for resistance includes only the down rating of filter units. The rejection rate remained the same for FY 1987 and FY 1988 and increased in FY 1989 to 2.8%.

TABLE 1

Annual Filter Test Results

FY 1986 - FY 1989

Fiscal Year	No. Tested	No. Accepted	%	No. Accepted w/Waiver	%	No. Rejected	%
1986	14,534	13,297	91	847	5.8	392	2.7
1987	13,656	13,161	96	235	1.7	260	1.9
1988	11,822	10,847	92	745	6.3	230	1.9
1989	12,538	12,042	96	139	1.1	347	2.8
TOTALS	52,550	49,347	93	1,966	3.7	1,229	2.3

Table 2 illustrates a summary of test results by FTF for FY 1986 - FY 1989. It should be noted that for FY 1986 - FY 1988, Rocky Flats, as with past fiscal years tested more filter than the other two FTFs combined. In FY 1989, The Oak Ridge FTF tested the highest number of filters compared with the other two FTFs. Evaluating the number of filters in the accepted with waiver category for each FTF, the Oak Ridge FTF demonstrated the highest percentage with the reported low of 2.0% in FY 1989 to the reported high of 16.0% in FY 1988. In discussing the FY 1988 acceptance with waiver data with Oak Ridge FTF personnel, it was indicated that the high acceptance with waiver rate is indicative of procurement procedures and is not related to the quality of filter being supplied or the operation of the Oak Ridge FTF. Rocky Flats FTF demonstrated a <1% acceptance with waiver rate, the lowest reported. This low acceptance with waiver rate could be attributed to the majority of the filters tested at the Rocky Flats FTF are utilized for use at the Rocky Flats site and are ordered in large quantities.

TABLE 2

Summary

Test Results By Filter Test Facility

FY 1986 - FY 1989

	No. Tested	No. Accepted w/Waiver	%	RANGE %	No. Rejected	%	RANGE %
HANFORD	6,130	98	1.6	1.3-5.2	174	2.7	2.1-3.7
OAK RIDGE	19,977	1,848	9.3	2.0-16	739	3.7	2.2-4.9
ROCKY FLATS	26,443	20	<1	<1	316	1.2	0.6-1.6
TOTALS	52,550	1,966	3.7		1,229	2.3	

Table 3 illustrates a summary of test results as grouped by filter manufacturer. Data is only reported for those filter manufacturers that supplied >100 filters per fiscal year. The table depicts a wide range for the acceptance with waiver criteria, with manufacturer C demonstrating the high of 23% and manufacturer E demonstrating the low of 0%. The number of filters in the rejected category, manufacturer A represented the highest number of filters tested demonstrating the lowest rejection rate with the rejection rate increasing to 2.8% which was observed in 1989.

TABLE 3

Summary

Test Results Grouped by Manufacturer

>100 Filters per year

Manufacturer	No. Tested	No. Accepted w/Waiver	%	RANGE %	No. Rejected	%	RANGE %
A	38,945	1,084	2.8	<1-8.1	696	1.8	<1-2.8
B	7,221	385	5.3	0-18	183	2.5	<1-4.2
C	4,035	361	8.9	0-23	140	3.5	2.3-5.1
D	1,086	70	6.5	1-11	49	4.5	2.6-6.1
E	470	0	0	0	75	16	5-29
F	131	18	13.7	14	1	12.2	<1
TOTALS	51,888	1,918	3.7		1,144	2.2	

Table 4 lists the eight (8) filter sizes with the associated nominal flow in cubic feet per minute (cfm) and ranges of flow to accommodate the separatorless type of filter units.

TABLE 4

Filter Size

Size	Nominal Flow - CFM	Flow Range - CFM
1	25	<37.5
2	50	37.5 - <87.5
3	125	87.5 - <312.5
4	500	312.5 - <750
5	1000	750 - <1125
6	1250	1125 - <1375
7	1500	1375 - <1750
8	2000	≥1750

The summary of the number of filters tested by size is illustrated in Table 5. The size 5 category represent 62.9% of the work load followed by the size two filter unit the standard glove box filter with a nominal flow rate of 50 cfm. The size 8, nominal flow of 2,000 cfm, demonstrates the highest percentage of accepted with waiver. The high acceptance with waiver rate in this category can be attributed to down rating the filter unit to meet the 1.0 in. w.g. resistance requirement. At the present time, acceptance with waiver criteria does not include downrating of filter units. In the future, filter units that exceed the 1.0 in. w.g. pressure drop requirement will be rejected.

TABLE 5

Summary - Number of Filters Tested by Size

FY 1986 - FY 1989

Size	No. Tested	No. Accepted w/Waiver	%	No. Rejected	%	% Total
1	3,357	9	2.7	124	3.7	6.4
2	6,754	123	1.8	153	2.3	12.9
3	3,748	95	2.5	99	2.6	7.1
4	1,487	96	6.5	77	0.05	2.8
5	33,055	1,967	.0	589	0.8	62.9
6	3,515	662	18.8	185	5.3	6.7
7	578	122	21	12	2.1	1.1
8	56	50	89	6	11	0.1
TOTAL	52,550	3,124	5.9	1,245	2.37	100.0

IV. Status of the High Flow Alternative Test System

In FY 1981 and continuing through FY 1985, DOE funded a program at Los Alamos to develop an aerosol test system suitable for use at the three FTFs as a potential replacement for the current DEHP Q107 penetrometer test system (2,3,4). Completion of the FTF TSG project resulted in recommendations for changes in the test systems used at the FTFs. The DOD funded Los Alamos to assist in the adoption and implementation of the HFATS designed to test filters with flow rates between 400 - 2,000 cfm at the three FTFs. A long term HFATS operational evaluation was completed in FY 1986 at the Oak Ridge FTF with the Oak Ridge FTF staff operating the HFATS for a nine month period(4). Review of the data collected during this evaluation indicated that the HFATS had a number of advantages. Extensive prooftesting of the HFATS is performed in conjunction with the installation of the HFATS at each FTF. Currently, the HFATS has been installed at the Oak Ridge FTF in March of 1990 and at the Rocky Flats FTF in May of 1990. Installation and prooftesting of the HFATS at the Hanford FTF is scheduled for September 1990 pending delivery of the diluter/sampler system presently being fabricated at Oak Ridge Gaseous Diffusion Plant.

An initial HFATS RRT is now in progress. The Rocky Flats FTF was designated as the reference laboratory (first participant) for the initial HFATS RRT. The data collected from the proof testing an initial RRT will provide preliminary results and necessary information about the consistency of the HFATS operation at the three FTFs. Analysis and the interpretation of the data collected from the initial HFATS RRT will determine if a more extensive RRT is required. Conclusive results can not be reported from the data collected from the two FTFs at this time.

After completion of the initial HFATS RRT and data analysis, and if the initial HFATS RRT is significant to determine if the HFATS is a comparable test method, notification will be provided to the DOE in accordance with NE F 3-43 Annex 6(8) for petitioning acceptance of the HFATS as a test method. It is anticipated that the initial HFATS RRT will be completed by the first of October with data analysis completed in November.

V. DOE/DOD Cooperative Efforts - Modernization of Filter Test Equipment

In FY 1985, the US Army - Product Assurance Directorate (PAD) DOD, and the DOE jointly agreed to cooperate in the development and evaluation of modern filter test systems. PAD agreed to support the development of the test systems and DOE agreed to support the operational evaluation of the developed system. DOE has completed an operational evaluation of the HFATS and the results have been made available to PAD and findings of the evaluation incorporated in the US Army HFATS to be delivered to PAD in the first quarter of FY 1991.

In FY 1990, funding was provided by the US Army PAD to Los Alamos for development and the adaptation of the LFATS. The LFATS is to be developed and a prototype installed at PAD. The LFATS represents a replacement for the existing Q233 DEHP penetrometer and the Q76 DEHP penetrometer capabilities of testing filters with rated flows from 20 - 250 cfm.

The specific objectives of the LFATS program include:

1. Adaptation of HFATS technology for use in the LFATS.

2. Fabrication of a test stand for the LFATS.

3. Implementation of PAD/LFATS at Aberdeen Proving Grounds, Maryland.

To date, the LFATS test equipment has been procured, the draft test stand specification has been written and the laboratory prooftesting equipment has begun. Laboratory prooftesting includes calibration of the laser spectrometer, calibration of the diluter sampler system, aerosol charge and concentration measurements.

VI. Summary

In summary, the RRT program has been established since 1982 and ensures the consistency of FTF measurements, the annual summary report of the acceptance/ rejection data provides DOE with information regarding HEPA filter performance and quality of HEPA filters that are procured and installed in DOE facilities, the HFATS has been installed and prooftested at the Oak Ridge FTF and Rocky Flats FTF. Installation and prooftesting of the HFATS at the Hanford FTF is scheduled in September of 1990. An initial HFATS RRT has be initiated. The data collected from the HFATS RRT will determine if the HFATS is a comparable test method with notification to DOE in accordance with NE F 3-43 Annex 6(8) for petitioning acceptance of the HFATS as a test method and NE F 3-43 Annex 5(8) to test with the most penetrating particle size. The modernization of filter test equipment has been a cooperative effort between DOE and the DOD with HFATS installed at two of the DOE FTFs and the anticipated installation of the DOD FTFs and the anticipated installation of the DOE HFATS in October 1990.

References

1. Employed by Environmental Health Sciences, Inc., contracted to Los Alamos National Laboratory.

2. R. C. Scripsick, S. C. Soderholm, and M. I. Tillery, "Evaluation of Methods, instrumentation and Materials Pertinent to Quality Assurance Filter Penetration Testing," published in the Proceedings of the 18th DOE Nuclear Airborne Waste Management and Air Cleaning Conference, Baltimore, Maryland (August 12-16, 1984), M./ W. First, ed., March 1985, pp. 1131-1143.

3. R. C. Scripsick and S. C. Soderholm, "Final Report: Evaluation of Methods, Instrumentation and Materials Pertinent to Quality Assurance Filter Penetration Testing," Los Alamos report LA-10748 (1986).

4. R. C. Scripsick, R. L. Smitherman, and S. A. McNabb, "Operational Evaluation of the High Flow Alternative Filter Test System," published in the Proceedings of the 19th DOE/NRC Nuclear Air Cleaning Conference, Seattle, Washington (August 18-21, 1986), M. W. First, ed., May 1987, pp. 863-885.

5. J. F. Bresson, "Quality Assurance Requirements for NESHAP Compliance," to be published in the 21st DOE/NRC Nuclear Air Cleaning Conference.

6. J. F. Bresson, "HEPA Filter Test Activities at the Department of Energy," published in the Proceedings of the 19th DOE/NRC Nuclear Air Cleaning Conference, Seattle, Washington (August 18-21, 1986), M. W. First, ed., May 1987, pp. 863-885.

7. US DOE Nuclear Standard NE F 3-42, Filter Test Facility Standards Writing Group, "Operating Policy of the DOE Filter Test Program," US Department of Energy, October 1988.

8. US DOE Nuelear Standard NE F 3-43, Filter Test Facility Standards Writing Group, "Quality Assurance Testing of HEPA Filters," US Department of Energy, February 1990.

9. US DOE Nuclear Standard NE F 3-44, Filter Test Facility Standards Writing Group, "DOE Filter Test Facilities Quality Program Plan," US Department of Energy, July 1986.

10. US DOE Nuclear Standard NE F 3-45, Filter Test Facility Standards Writing Group, "Specifications for HEPA Filters Used By DOE Contractors," US Department of Energy, October 1988.

11. "Filters, Particulate (High-Efficiency Fire Resistant)," Military Standard MIL-F-51068F, August 1988.

12. Semi-annual Report Summaries for fiscal years 1986 - 1989, R. C. Scripsick, Los Alamos National Laboratory.

DISCUSSION

WEBER: Could you please elaborate briefly on the differences in components of the HFATS and Q127 penetrometer systems?

McINTYRE: The high flow alternative test system is for the Q107 penetrometer and uses a laser spectrometer to determine particle size and concentration coupled with a computer to provide a readout. The aerosol is a cold-generated polydisperse aerosol using a Laskin Nozzle at about 1/5th of the usual concentration combined with a laser spectrometer to measure particle size and concentration and a computer to generate data.

WEBER: Do you use the same liquid aerosols?

McINTYRE: We use dioctylphthate at two of the facilities and dioctylsebacate at one facility.

PORCO: You stated that HFATS will replace the Q107 method. How soon is that going to happen at all three facilities?

McINTYRE: We expect to have the initial roundrobin results soon to provide the needed backup. The procedure will be implemented by January 1, 1991 provided the DOE approves the method.

PORCO: My next question is on NEF 3-43. It was issued in February 1990. It lists the only approved DOE method as the light scattering photometer, Q107. When is it going to come out and be changed?

McINTYRE: In the stated standard the HFATS is referenced in Annex 5 and Annex 6. To approve a new test method or aerosol, you must go through several steps that include notification to manufacturers, and soliciting DOE approval. The Department of Energy has the final say whether the test method or aerosol is accepted or not.

FRANKLIN: Of the facilities where you have conducted roundrobins, does your preliminary data give a good correlation, or don't you know, yet?

McINTYRE: At this time it appears that the data are consistent with those reported in the 19th Nuclear Air Cleaning Conference Proceedings comparing the HFATS and Q107 systems.

A SYSTEMATIC STUDY ON THE EFFECTIVENESS OF MONO ALKYL IODIDES OF CYCLIC DIAMINES AS AN IMPREGNANT OF AN ACTIVATED CARBON OF THE NUCLEAR GRADE

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Abstract

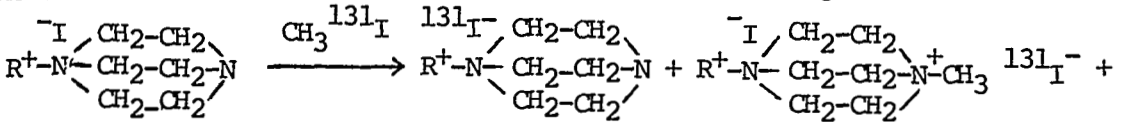
Mono alkyl iodides(R=-CH_3 and -C_2H_5) of two cyclic diamines(DABCO and 1,4-dimethylpiperazine) were prepared and the effectiveness of these chemicals as an impregnant was systematically studied with respects to their reactivities with methyl iodide in both a liquid and a solid phase. Yields of the diiodide formed depending on the reaction time in an alcoholic solvent showed the higher values for DABCO derivatives as compared to those of piperazine analogues. The similar type of the reaction with radio active methyl iodide could show the higher labelled yield of the addition product in cases of DABCO derivatives, whereas those of piperazine analogues were of minor, but their radio isotopic exchange yields were higher than those of DABCO derivatives at a high reaction temperature. Active carbon samples impregnated by these chemicals varying the impregnated weight percentage showed the higher adsorption yield of methyl iodide gas under a dried air stream at 30°C in cases of DABCO derivatives as compared to those of piperazine analogues, but variation of the yield was depended a great deal to the impregnated weight percentage of a sample applied in cases of the former, whereas the latter analogues showed a little variation. At 80°C, the adsorptivity of methyl iodide gas by these carbon samples were decreased, but the variation of the yield depending on the impregnation weight percentage of a carbon sample was of little significance. Penetration ratio(%) of radio active methyl iodide gas through these carbon samples at 30°C and 80°C showed the similar trend of decreasing the ratio as above, and piperazine derivatives were able to show the better penetration performance at 80°C than those of DABCO analogues and the impregnation weight percentage of the carbon sample is not much influenced by the variation of the penetration ratio. Results were discussed for the feasibility of this method to be applied for selecting an effective impregnant of an activated carbon of the nuclear grade.

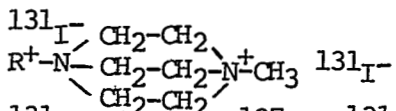
1. Introduction

In the previous study(1), mono methyl iodide of DABCO had shown the non deteriorative quality under a prolonged exposure of a humid air flow at the operational temperature of the ESF filter system of a nuclear power plant(80°C) and it afforded favorable physical and chemical properties as an impregnant of an activated carbon of the nuclear grade as summarized in the table 1. Mono alkyl iodides of DABCO and relevent cyclic diamines were, therefore, considered as one of the potential effective impregnant of an activated carbon to be useful for the carbon filter system of a nuclear power plant, especially for the ESF filter system, which could be used for a prolonged time without being deteriorated its adsorption characteristics. In this study, mono alkyl iodides of DABCO and 1,4-dimethylpiperazine were considered for a systematic survey on their effectiveness as an impregnant. (R= -CH_3 and-C_2H_5)

Performing this systematic approach, chemical reactivities of these compounds with methyl iodide in a liquid phase were surveyed at first to confirm their behaviors as a chemical. This type of compound had shortly been mentioned on its existence in literatures and detailed studies on its chemical reactivity with organic iodides such as methyl iodide were not clearly reported except cases of dialkyl

iodides of DABCO. (2) In alcoholic solvents such as ethanol and methanol,methyl iodide was , therefore, reacted with mono alkyl iodides of the present series at the refluxing temperature.(78.4°C & 64.7°C) Subsequently, studies on their reactivities with radioactive methyl iodide (^{131}I) were conducted in the similar solvent system as above. Radioactive methyl iodide may be expected to form the following labelled product with these mono iodides in which one radioactive exchange and two addition reactions are involved as described in the following.





Since a tracer solution of $CH_3{}^{131}I$ (i.e. $CH_3{}^{127}I + {}^{131}I$) was used in the experiment, the actual reaction sequences were more complicate than the above, which involved two more addition reactions by $CH_3{}^{127}I$ and three more radioactive exchange reactions of these products formed. Efforts were, therefore, concentrated to classify total amounts of labelled addition products and radioactive exchange reaction products thereof. Detailed study along the identification of the individual labelled products may further be attempted in the subsequent research.

Based on these fundamental chemical reactivities, reactivities in a solid phase (i.e. the adsorptivity of methyl iodide gas in the active carbon sample impregnated by these chemicals) were attempted to be confirmed. In the preliminary experiment of this approach, the adsorption of methyl iodide gas by the impregnated carbon was studied under a dry air flow applying the method described for CCl_4 activity of activated carbon in ASTM procedures. (3) The dependence of the adsorption yield on the impregnated weight percentage of a carbon sample was particularly studied so that the data may be referred to optimize the impregnation condition of an activated carbon by these chemicals. Finally, penetration ratio (%) of radioactive methyl iodide gas through the carbon sample was evaluated applying the existent ASTM testing procedure. (4) Again, the dependence of the penetration ratio on the impregnation weight percentage of a sample was closely studied. In this experiment, the input concentration of radioactive methyl iodide gas was adjusted higher than that of ASTM procedure in order to confirm the ratio more accurately.

Results obtained in this systematic study were further evaluated for the feasibility to be adopted for selecting an effective impregnant of an active carbon of the nuclear grade.

2. Experimental Procedures

2.1 Reaction of Methyl Iodide with Mono Alkyl Iodides in an Alcoholic Solvent.

Into a three necked round bottomed flask equipped with a reflux condenser, a magnetic stirrer and an oil bath, a solution of the definite quantities of methyl iodide(0.00112 mole scale) and a mono alkyl iodide(0.01 mole scale) in 110 ml. of anhydrous alcohol was introduced and the resulting reaction solution was refluxed under a vigorous stirring for the definite reaction time. (see table 2) The reaction solution was then filtered hot through a glass funnel and the solid filtered was dried in an oven. The filtrate was further treated to collect solid products formed by the reaction on basis of the data obtained from a separate experiment to identify the solubilities of the diiodide and the mono alkyl iodide applied.

Table 1. Comparison of Properties of TEDA and its Derivatives and Activated Carbons Impregnated by These Chemicals

Type of Compound	$N(CH_2CH_2)_3N^+ CH_3I^-$	$^+N(CH_2CH_2)_3N^+ CH_3I^- \quad CH_3I^-$	$N(CH_2CH_2)_3N + KI$
1. m pt.(°C)	211	260	TEDA 170
2. pH of aq. sol.	6.8[2]	5.3[2]	TEDA:10.2[1] KI : 6.0[1]
3. Isotope product formed by $CH_3{}^{131}I_{(\%)}$[3]	81.86	84.85	TEDA = 78.85 KI = 66.12
4. Impregnated carbon			
a. Impregnated (wt%)	4.43	4.38	3.84 (TEDA = 2.14, KI = 1.70)
b. pH[4]	9.0	7.6	9.6
c. Deterioration at 80°C RH 90%, 15hrs.	None	None	TEDA deteriorated ca. 20%
d. $CH_3{}^{131}I$ 5) penetration (%)	0.08	0.09	0.08

1) Aq. sol. of 0.2g samples in 100ml of water (pH of water : 6.3).
2) Aq. sol. of 0.4g samples in 50ml of water (pH of water : 6.3).
3) In presence of 30ml MeOH and 10ml of water, sample: 0.3g,0.4g,0.025g and 0.3g, respectively. Reaction time 72hrs. Reaction Temp. 25-30°C.
4) According to ASTM D-3838-80.
5) According to ASTM D-3803-86, method A.

The solid product and the unreacted mono alkyl iodide were thus separated, identified for their chemical structures, and weighed to confirm the yield of the diiodide formed. Results were summarized in table 2. A and B.

2.2 Reaction of Radioactive Methyl Iodide(^{131}I) with Mono Alkyl Iodides in an Alcoholic Solvent System.

A. Radio Isotopic Reaction

Into a 50 ml., two necked round bottomed flask equipped with a reflux condenser, magnetic stirrer, and a stirring hot plate, a solution of the definite concentrations of radioactive methyl iodide tracer($CH_3{}^{127}I + CH_3{}^{131}I$) and a mono alkyl iodide in 29.0 ml. of the alcohol was introduced and the resulting reaction solution was refluxed under a vigorous stirring for the definite time.(see tables 3-6) The reaction solution was cooled and transferred to a counting cell. The radioactivity of the solution was measured by means of a Ge-Li detector(EG & G Ortec. 7050-7000 Series) recording the time. The solution was, then, concentrated to remove the unreacted radioactive methyl iodide by heating it under a vigorous stirring and the radioactivity of the concentrated solution was measured upon cooling and the time was recorded again. The radioactive labelling yield of the mono alkyl iodide was calculated by means of comparing these two radioactivities

measured referring their counted times. Results were summarized in tables 3-6.

B. Apparent Yield of the Radioactive Exchange Reaction

The same procedures described in item 1 above were followed to confirm the yield of the addition reaction of methyl iodide under this reaction condition adopting the same reaction parameters of A above and non radioactive methyl iodide was used only. The yield obtained by treating the reaction solution was sustracted from the radioactive labelled yield in A to estimate the apparent yield of the radioactive exchange reaction. Results were summarized in tables 3-6.

2.3 Adsorption of Methyl Iodide with Mono Alkyl Iodide.

A. Adsorption Process

The apparatus and procedures described in ASTM D Standard for carbon tetrachloride activity of activated carbon (3) were applied for the adsorption process of methyl iodide gas with the impregnated active carbon under dried air flow. The adsorption process was terminated when the increased weight of the carbon sample become constant.

B. Desorption Process

The same apparatus and procedures described in A were applied. The desorption process was conducted by means of passing the dried air through the carbon sample treated as above and was terminated when the decrease of weight of the carbon sample approached to a constant level. The carbon sample was further dried in an oven at 100-110°C until the weight of the sample become constant. The total increase of the weight of the carbon sample treated was compared to that of the base carbon treated as shown in A and B processes and the adsorption yield of the sample was calculated. Results were summarized in tables 7,8, and 9. A plot of the molar concentration of the impregnated in the carbon sample v.s. moles of the adsorption product formed in the same sample was shown in fig. 1.

2.4 Penetration of Radioactive Methyl Iodide(^{131}I) through the Impregnated Carbon.

The apparatus developed by this laboratory(5) and procedures described in ASTM D 3803-86(4) were applied. The concentration of the test adsorbate was, however, adjusted to a higher value than that of the ASTM D procedure in order to confirm the penetration ratio(%) more accurately. Results were summarized in tables 10-12.

3. Results and Discussion

3.1 Reactivity of Mono Alkyl Iodide with Methyl Iodide.

As shown in the table 2 A and B, mono alkyl iodide of DABCO could result the higher yield of the diiodide as compared with piperazine analogues in both methanol and ethanol solvent systems. This difference of the reactivity may be attributed to the relative electronic moiety of the tertiary nitrogen atom in the molecule in which the electron density of the tertiary nitrogen of the DABCO derivative is more dense than that of the piperazine analogue. Ethyl derivatives appeared to be more reactive with methyl iodide than those of methyl substituted, which may probably be due to the increased electron donating character of the ethyl group decreasing the positive moiety of the quaternary nitrogen atom. For alkyl derivatives of DABCO, (R=-CH_3,-C_2H_5,-C_3H_7, and -C_4H_9) the yield of the

Table 2. Variation of Yields of Diiodides Depending on the Reaction Time.

A. Methanol** system

Compound*** \ RX Time	1.0	1.5	2.0	3.5	5.0	Remarks
R= - CH_3	36.32	45.96	52.78	59.34	69.00	
- C_2H_5	39.47	49.72	57.52	67.52	75.54	
R'=- CH_3	10.55	14.32	19.09	27.39	34.17	
- C_2H_5	14.81	20.11	24.75	32.99	39.93	

* RX Temp. : Refluxing(64.7°C)
** Solvent : 100 ml.
*** Reactant Scale : 0.01 mole for mono alkyl iodide. ; 0.0112 mole for methyl iodide.

B. Ethanol System

Compound*** \ RX Time	1.0	1.5	2.0	3.5	5.0	Remarks
R= - CH_3	46.72	57.38	64.97	67.91	76.01	
- C_2H_5	57.56	68.05	73.46	76.90	83.95	
R'=- CH_3	24.62	32.40	38.69	46.73	56.03	
- C_2H_5	26.02	33.89	40.14	52.71	59.51	

* Solvent : 100 ml.
** Reaction Temp. : Refluxing(78.4°C)
*** Reactant Scale : 0.01 mole for mono alkyl iodide. ; 0.0112 mole for methyl iodide.

Table 3. Profiles of Reaction between Mono Alkyl Iodide and Radio Active Methyl Iodide(^{131}I) in Methanol Solvent.(A Dilute System)

A. Methyl Iodide Deficient.(2)

R or R'(2)	Yield of Radio Active Products%	Yield of Diiodide Salt(%)(3)	Apparent Radio Active Exchange, Yield(%)(4)
R=-CH_3	90.90	16.36	74.54
-C_2H_5	91.34	16.43	74.91
R'=- CH_3	84.20	1.95	82.25
- C_2H_5	95.37	10.06	85.31

(1) Solvent : 29 ml. RX temp. : Refluxing. RX. Time : 1hr.
(2) Mol. Conc. of Mono alkyl iodide: 0.001 ; Methyl iodide : 0.00048
(3) RX. of the similar type was performed using nonradioactive methyl iodide and yield of the diiodide was measured on basis of methyl iodide used.
(4) Calculated by substracting yield of the diiodide from the yield of radio active product.

B. Methyl Iodide Excess.[2]

R or R'[2]	Yield of Radio Active Products(%)	Yield of Diiodide Salt(%)[3]	Apparent Radio Active Exchange,Yield(%)[4]
R=-CH_3	66.37	12.33	54.04
-C_2H_5	69.48	15.44	54.04
R'=- CH_3	59.96	2.79	57.17
- C_2H_5	78.25	5.93	72.32

(2)-(4) : All reaction parameters were the same as A. above except mol. conc. of methyl iodide.(0.00112, the amount of radioactive methyl iodide was adjusted to the same of A.)

Table 4. Profiles of Reaction[1] between Mono Alkyl Iodide and Radio Active Methyl Iodide(^{131}I) in Ethanol Solvent. (A Dilute System)

A. Methyl Iodide Deficient.[2]

R or R'[2]	Yield of Radio Active Products(%)	Yield of Diiodide Salt(%)[3]	Apparent Radio Active Exchange,Yield(%)[4]
R=-CH_3	90.12	39.25	50.87
-C_2H_5	91.04	41.70	49.34
R'=- CH_3	76.80	7.81	68.99
- C_2H_5	95.53	16.98	78.55

(1)-(4) : All reaction parameters are the same as shown in the table 3-A.

B. Methyl Iodide Excess.[2]

R or R'[2]	Yield of Radio Active Products(%)	Yield of Diiodide Salt(%)[3]	Apparent Radio Active Exchange,Yield(%)[4]
R=-CH_3	66.54	26.97	39.57
-C_2H_5	69.33	30.93	33.40
R'=- CH_3	67.87	7.27	60.60
- C_2H_5	73.91	11.47	62.44

(1)-(4) : All reaction parameters are the same as shown in the table 3-B.

Table 5. Profiles of Reaction[1] between Mono Alkyl Iodide and Radio Active Methyl Iodide (^{131}I) in Methanol Solvent. (A Conc. System)

A. Methyl Iodide Deficient.[1]

R or R'[1]	Yield of Radio Active Products(%)	Yield of Diiodide Salt(%)[2]	Apparent Radio Active Exchange,Yield(%)[3]
R=-CH_3	94.06	35.33	58.73
-C_2H_5	87.97	43.22	44.75
R'=- CH_3	71.39	8.59	62.80
- C_2H_5	83.63	14.86	68.77

(1)-(3) : Mol. conc. of mono alkyl iodide is 0.0025 and that of methyl iodide is 0.0008. All other parameters are the same as shown in the table 3.

B. Methyl Iodide Excess.[1]

R or R'	Yield of Radio Active Products(%)	Yield of Diiodide Salt(%)[2]	Apparent Radio Active Exchange,Yield(%)[3]
R=-CH_3	80.03	36.62	43.41
-C_2H_5	84.17	39.47	44.70
R'=- CH_3	66.66	10.55	56.11
- C_2H_5	83.79	14.81	68.98

(1)-(3) : All reaction parameters are the same as shown in the above A except mol. conc. of methyl iodide(0.00257)

Table 6. Profiles of Reaction between Mono Alkyl Iodide and Radio Active Methyl Iodide(^{131}I) in Ethanol Solvent. (A Conc. System)

A. Methyl Iodide Deficient.[1]

R or R'	Yield of Radio Active Products(%)	Yield of Diiodide Salt(%)[2]	Apparent Radio Active Exchange,Yield(%)[3]
R=-CH_3	83.03	79.29	3.74
-C_2H_5	82.67	83.00	0
R'=- CH_3	71.86	20.31	51.55
- C_2H_5	82.02	26.41	55.61

(1)-(3) : The same as shown in the table 5-A.

B. Methyl Iodide Excess.(1)

R or R'	Yield of Radio Active Products(%)	Yield of Diiodide Salt(%)(2)	Apparent Radio Active Exchange,Yield(%)(3)
R=-CH_3	90.47	46.72	43.75
-C_2H_5	99.10	57.56	41.54
R'=- CH_3	77.92	24.62	53.30
- C_2H_5	81.93	26.02	55.91

(1)-(3) : The same as shown in the table 5-B.

Table 7. Adsorption Pattern* of Impregnated Active Carbon with Methyl Iodide under a Dried Air Flow.

Impregnant**	Impregnated Wt. %	Adsorption*** Product Yield(%)	Impregnant**	Impregnated Wt. %	Adsorption*** Product Yield(%)
A	4.02	53.99	C	3.09	33.40
A	4.74	51.62	C	4.12	33.33
A	5.88	47.13	C	5.98	33.09
B	4.12	56.24	C	7.22	30.88
B	5.46	54.77	D	1.86	31.29
B	6.19	46.98	D	4.33	31.82
C	1.86	34.56	D	6.29	27.94

* Refer to ASTM D 3467-88(CCl_4 Activity). RX. Temp. 30±1 °C. Conc. of CH_3I 750±10 mg/l. Air flow rate : 600± ml/min.

** A : $N(CH_2CH_2)_3N^+CH_3I^-$ B : $N(CH_2CH_2)_3N^+C_2H_5I^-$

C : $H_3CN(CH_2CH_2)_2N^+(CH_3)CH_3I^-$ D : $H_3CN(CH_2CH_2)_2N^+(CH_3)C_2H_5I^-$

*** A complex compound of the N-alkyl iodide applied.

Table 8. Adsorption Pattern* of Impregnated Active Carbon with Methyl Iodide under a Dried Air Flow.

Impregnant**	Impregnated Wt. %	Adsorption*** Product Yield(%)	Impregnant**	Impregnated Wt. %	Adsorption*** Product Yield(%)
A	4.74	39.02	C	4.12	23.31
A	5.88	37.42	C	7.22	19.77
B	5.46	40.09	D	4.33	23.11
B	6.19	37.76	D	6.29	21.53

* Refer to ASTM D 3467-88(CCl_4 Activity). RX. Temp. 80±1 °C Conc. of CH_3I 750±10 mg/l. Air flow rate : 600±5 ml/min.

** A : $N(CH_2CH_2)_3N^+CH_3I^-$ B : $N(CH_2CH_2)_3N^+C_2H_5I^-$

C : $H_3CN(CH_2CH_2)_2N^+(CH_3)CH_3I^-$ D : $H_3CN(CH_2CH_2)_2N^+(CH_3)C_2H_5I^-$

*** A complex compound of the N-alkyl iodide applied.

Table 9. Adsorption Pattern* of Impregnated Active Carbon with Methyl Iodide under a Dried Air Flow. (A Detailed Study)

Impregnant : $N(CH_2CH_2)_3N^+C_2H_5I^-$

Impregnated Wt. (%)	Adsorption Product** Yield (%)	Impregnated Wt. (%)	Adsorption Product** Yield (%)
1.58	54.88	3.50	55.69
1.83	54.74	4.12	56.14
2.33	54.94	5.46	54.46
3.00	55.35	6.00	48.01

* Refer to ASTM D 3467-88(CCl_4 Activity). RX. Temp. 30± °C Conc. of CH_3I 750± 10 mg/l. Air flow rate : 600± ml/min.

** A complex compound of the N-alkyl iodide applied.

Table 10. Examples of $CH_3{}^{131}I$ Penetration* of the Impregnated Active Carbon. (30°C)

Impregnant**	Impregnated Wt. %	Penetration (%)	Impregnant**	Impregnated Wt. %	Penetration (%)
A	2.06	7.95	C	4.02	4.76
A	4.02	8.98	C	5.98	10.22
A	5.88	12.02	D	3.94	9.35
B	2.16	3.31	D	6.19	3.45
B	4.12	5.20	E	1.86	9.62
B	6.19	11.69	E	4.23	7.53
C	1.86	6.27	E	6.29	8.22

* Based on ASTM D 3803-86, Method A except the following parameters; Concentration of test adsorbate 6.1 x 10^{-1} mg/min. RH 90 %.

** A: $N(CH_2CH_2)_3N^+CH_3I^-$ B: $N(CH_2CH_2)_3N^+C_2H_5I^-$

C: $H_3CN(CH_2CH_2)_2N^+(CH_3)CH_3I^-$ D: TEDA + KI

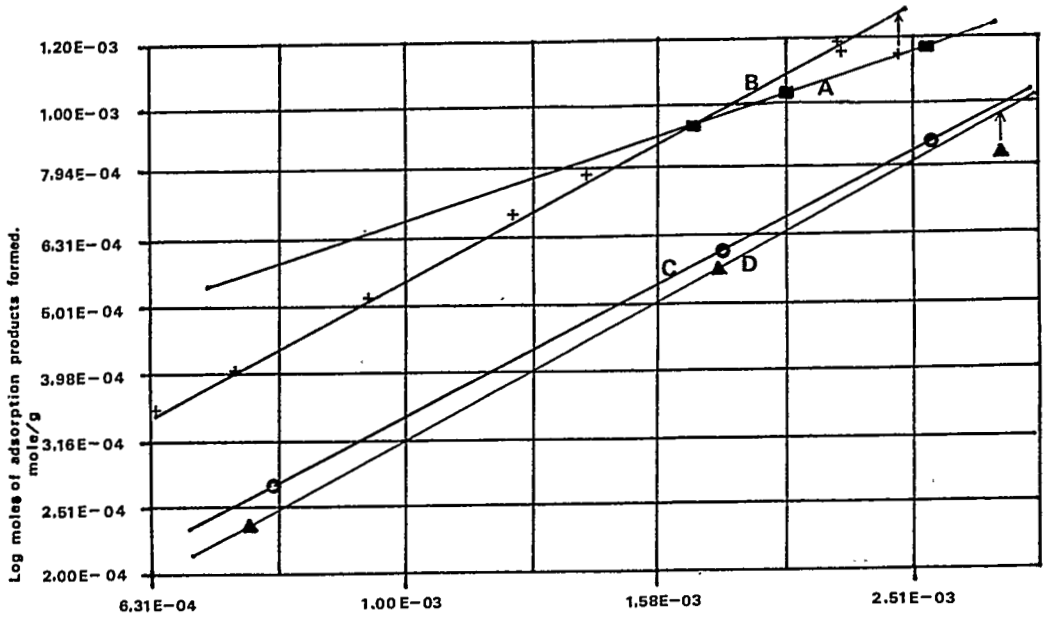
E: $H_3CN(CH_2CH_2)_2N^+(CH_3)C_2H_5I^-$

Table 11. Examples of $CH_3{}^{131}I$ Penetration* of the Impregnated Active Carbon (A Detailed Study, Impregnant: $N(CH_2CH_2)_3N^+C_2H_5I^-$)

Impregnated Wt. (%)	Penetration (%)	Impregnated Wt. (%)	Penetration (%)
Base carbon	50.80	3.09	25.29
1.50	30.26	3.50	21.98
1.83	24.42	4.14	24.65
2.23	25.22	6.19	27.98

* Based on ASTM D 3803-86, Method A except the following parameters; Concentration of test adsorbate 6.1 x 10^{-1} mg/l. Elution temp. 50°C.

Fig. 1. A plot of the Molar Concentration of the Impregnant in the Carbon Sample v.s. Moles of Adsorption Product Formed in the Same Sample. (30±1 °C)



Log moles of impregnant in the carbon sample.
(The logarithm scale is converted to the regular scale by a computer.)

A : $N(CH_2CH_2)_3N^+CH_3I^-$ B : $N(CH_2CH_2)_3N^+C_2H_5I^-$

C : $H_3CN(CH_2CH_2)_2N^+(CH_3)CH_3I^-$ D : $H_3CN(CH_2CH_2)_2N^+(CH_3)C_2H_5I^-$

Table 12. Examples of $CH_3{}^{131}I$ Penetration* of the Impregnated Active Carbon. (80 °C)

Impregnant	Impregnated Wt. %	Penetration (%)	Impregnant	Impregnated Wt. %	Penetration (%)
Base carbon		5.45	C	1.86	1.06
A**	1.86	0.58	C	5.98	0.10
A	6.19	0.12	E	1.86	0.42
B	2.16	0.22	E	6.29	0.09
B	5.36	0.28			

* Based on ASTM D 3803-86, Method B except the following parameters; Equilibrium Time: 2 hrs. Feeding Time: 2 hrs. Elution Time: 2 hrs. and Concentration of test adsorbates 6.1 x 10^{-1} mg/min.

** A,B,C and E denote the compound as shown in the table 10.

diiodide showed an increase in order of R=-C_2H_5 > -C_3H_7 > -CH_3 > -C_4H_9 at the refluxing temperature of methanol and R=-C_2H_5 > -C_3H_7 > -C_4H_9 > -CH_3 at the refluxing temperature of ethanol, respectively, which had been confirmed in the author's separate research.(6) In these cases, ethyl derivative could result the highest yield of the diiodide, which was explained by a δ^+ through space influence of the electron moiety of the quaternary nitrogen atom on that of the tertiary nitrogen atom in the same molecule. Difference of the yield of the diiodide observed in DABCO and piperazine derivatives may further be studied to elucidate it on basis of their molecular structure in subsequent research.

The yield was surveyed in details depending on the reaction time. The kinetic data along this approach will be published elsewhere. Generally, the increase of the yield of the diiodide was more apparent in case of DABCO derivatives than those of piperazine analogues which showed the generality of the enhanced reactivity in the former cases.

The difference of the yield of the diiodide observed in a liquid phase as the present case was considered to be able to be referred for evaluating the relative effectiveness of an impregnated carbon adsorbing gaseous methyl iodide in the solid phase and it was true as described in the followings.

3.2 Reactivity of Mono Alkyl Iodide with Radioactive Methyl Iodide.(^{131}I)

As shown in tables 3-6, the difference of the yield of radioactive labelled products observed between DABCO and piperazine derivatives was studied in details varying the molar ratio of methyl iodide and mono alkyl iodide concerned in order to adjust the reaction rates of addition and radioactive exchange reactions involved. Since the tracer solution applied in this reaction study bears a large quantity of non radioactive methyl iodide as the carrier and addition reaction involved is influenced by the concentrations of both radioactive and non radioactive methyl iodides through a SN_2 type reaction path, the total concentration of methyl iodide in the reaction system would be the essential factor to adjust the rate of addition reactions involved, whereas the radioactive exchange reaction may approximately be concerned with the trace quantity of radioactive methyl iodide itself.(Ref. to Mckay equation. (7))

Results observed in table 3 A and B, where the concentration of methyl iodide is varied keeping the molar concentration of radioactive methyl iodide constant, indicated that the total radioactive labelled yield was increased in order of piperazine (R=-C_2H_5) > DABCO(R=-C_2H_5) > DABCO(R=-CH_3) > piperazine(R=-CH_3) and apparent yield of radioactive exchange reaction was increased in order of piperazine (R=-C_2H_5) > piperazine(R=-CH_3) > DABCO(R=-C_2H_5) > DABCO(R=-CH_3). These observations indicated that piperazine derivatives are in favor of more radioactive exchange reaction than their addition reaction, whereas DABCO derivatives are shown in the reverse way, even though the reaction rate of their addition reactions shown in table 3 A was increased by the methyl iodide deficient reaction condition. Decrease of the total radioactive labelled yield in cases of B as compared with those of A may be due to the decrease of the rates of the overall reactions under the methyl iodide excess reaction condition. At a higher reaction temperature as shown in table 4, the total radioactive labelled yield was increased in order of piperazine (R=-C_2H_5) > DABCO(R=-C_2H_5) > DABCO(R=-CH_3) > piperazine(R=-CH_3) and apparent yield of the radioactive exchange reaction was increased in order of piperazine(R=-C_2H_5) > piperazine(R=-CH_3) > DABCO(R=-CH_3) > DABCO(R=-C_2H_5). The order of the increased yield of the radioactive exchange reaction for DABCO(R=-C_2H_5) and (R=-CH_3)was reversed at this temperature comparing it with those of table 3, which indicated that the

increase of yield of addition reaction of DABCO(R=-C_2H_5) was little higher than that of lower reaction temperature as shown in table 3. The general mode of increasing the yield of the radioactive exchange reaction at this temperature was shown a higher value than that of lower reaction temperature as shown in table 4, which indicated that the radioactive exchange reaction of DABCO derivatives was markedly suppressed due to the enhanced reaction rate of the addition reaction at this temperature. In tables 5 and 6, results obtained in reactions of doubling the concentration of reactants in the system were shown to afford the similar trends of yields observed as above, but the preference of the radioactive exchange reaction for piperazine derivatives were shown vividly in case of A condition of table 6 at a high reaction temperature.

Even though more definite kinetic data on reaction rates should further be explored and the apparent yield of the radioactive exchange reaction was calculated on basis of the additivity of radioactive exchange reaction and addition reaction which may closely be evaluated with respect to its feasibility in such competitive reaction path as the present case, piperazine derivatives were proved to be in favor of more radioactive exchange reaction than DABCO derivatives and the ethyl derivative resulted a slight higher yield of radioactive labelling as compared with the corresponding methyl derivatives. These tendencies observed were also experienced in the following solid phase reactions, which were discussed referring data described in this item.

3.3 Adsorption of Gaseous Methyl Iodide in an Impregnated Carbon.

As shown in tables 7 and 8, the adsorption yield of methyl iodide in the carbon sample impregnated with a close equivalent quantity of each impregnant was increased in order of DABCO(R=-C_2H_5)$\rangle$ DABCO(R=-CH_3)$\rangle$ piperazine(R=-CH_3)$\rangle$ piperazine (R=-C_2H_5) and the difference of the yield in cases of piperazine derivatives was of a slight value as compared with DABCO analogues. These tendencies were more clearly observed in fig. 1 where the molar concentration of the impregnant in the carbon sample was plotted against the moles of the adsorption product formed on basis of logarithm scales. The relative reactivity of each impregnant with methyl iodide thus observed was almost in accord with that of 3.1 above. Probably, the reactivity of mono alkyl iodides with methyl iodide was in parallel with a liquid and a solid phase.

However, the adsorption yield of an impregnated carbon could not show a linear relationship with its impregnated weight percentage in which a decrease of the yield was observed as its impregnated weight percentage is increased. This tendency was of more significance in cases of DABCO derivatives as compared with piperazine analogues. At a higher reaction temperature(80°C), this tendency for DABCO derivatives become of less significance as shown in table 8. In fig. 1, the marked decrease of the adsorption yield of the carbon sample impregnated with DABCO(R=-C_2H_5) was clearly observed, which was further studied in details varying its impregnation weight percentage in a small fraction as shown in table 9. Thus, the optimum impregnated weight percentage affording the high adsorption yield would be covered by 2.0-4.0% range in the DABCO(R=-C_2H_5) case. There had been some discussions on the optimum impregnated weight percentage of an active carbon where an excess quantity of an impregnant in a carbon decrease its adsorption of a chemical because of an excessive occupacy of the carbon pore with the chemical molecules.(8) Present observations in regards to the non linear relationship between the adsorption yield and the impregnated weight percentage may be due to the difference in the molecular structure of each impregnant to occupy the carbon pore by its molar volume capacity.

Thus, DABCO(R=-C_2H_5) occupied more space in the carbon pore by its molecular structure than the cases of others.

As described in the experimental procedures, the saturated carbon sample with methyl iodide at the adsorption process was subjected to the desorption process to remove methyl iodide adsorbed physically in the carbon sample. In fig. 2, examples of desorption profiles for DABCO(R=-C_2H_5) impregnated carbons(1.58% and 6.0%) were shown for a reference purpose. Observing these profiles, the desorption rate of methyl iodide was faster in case of 1.58% impregnated than 6.0% impregnated, which may partly be due to the excessive occupacy of the carbon pore by the latter case. Detailed researches along this trend of the desorption process of an impregnant in an activated carbon are now under progressing. A vivid desorption profiles of the exhausted carbons, which had been damaged for their normal pore behaviors, had already been experienced in the previous study.(9)

In conclusion, adsorption profile of an impregnated carbon under dried air flow could be followed by this experiment affording data on the relative reactivity of the impregnant with gaseous methyl iodide, which were in accord with those reactivities observed in 3.1 above. Furthermore, the range of the optimum impregnation weight percentage affording the high adsorption yield was able to be confirmed.

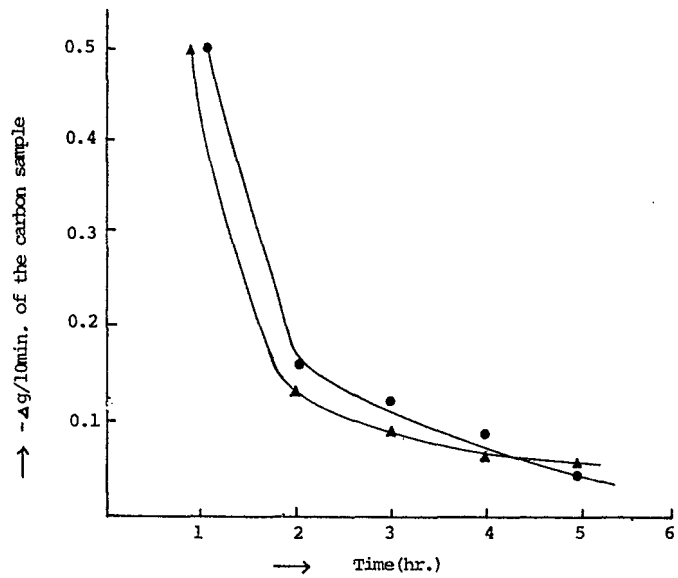


Fig. 2 Desorption of Methyl Iodide from an Impregnated Active Carbon Saturated by the Same Iodide.(Ref. table 9)

● : 1.58% Impregnated Carbon by DABCO(R=-C_2H_5)

▲ : 6.00% Impregnated Carbon by DABCO(R=-C_2H_5)

3.4 Penetration of Radioactive Methyl Iodide(^{131}I) through an Impregnated Carbon.

As summarized in table 10, penetration of radioactive methyl iodide(^{131}I) through the carbon sample impregnated with the close equivalent quantity of each impregnant showed the decrease of the ratio(%) in order of DABCO(R=-C_2H_5)〈piperazine(R=-CH_3)〈 DABCO(R=-CH_3)〈 piperazine(R=-C_2H_5) at the impregnated weight level of

two percent, whereas it showed the decrease in order of piperazine($R=-C_2H_5$)<piperazine ($R=-CH_3$)< DABCO($R=-C_2H_5$)< DABCO($R=-CH_3$) at the level of 6.0%. The former case was corresponded to the relative reactivities of the addition reaction observed in 3.1 and 3.3, and the latter case was equivalent to the total radioactive labelling yield observed in 3.2. These observations indicated that the carbon sample impregnated with the less amount of mono alkyl iodide favored the addition reaction path with radioactive methyl iodide gas, whereas the carbon sample with the high quantity of the iodide favored the radioactive exchange reaction path, especially for piperazine derivatives as observed in table 10. For comparison purpose, the sample impregnated with TEDA + KI was examined for its penetration ratio which afforded a larger ratio than those of mono alkyl iodide impregnated at the impregnated weight level of less than 4%, and a lower value at the level of more than 4.0%. Therefore, it may be said that mono alkyl iodide is the better impregnant for a carbon used at 30°C than the existing TEDA + KI impregnant system within the impregnation level of 4.0% weight.

The penetration ratio could not show a linear relationship with the impregnation weight percentage of the carbon sample, which was studied in details for the case of DABCO($R=-C_2H_5$) as shown in table 11. The optimum impregnation weight percentage giving the low ratio was covered within the range of 2.0-4.0%. This trend was also observed in 3.3 above. These two observations in regards to the optimum impregnation weight percentage may be the important factor to select an effective impregnated carbon of DABCO($R=-C_2H_5$) type to be used at the operation temperature of the normal carbon filter system(30°C).

At a high adsorption temperature(80°C), penetration ratio(%) of the carbon sample was markedly decreased as compared with that of the penetration done at 30°C, and the difference of the ratio observed in each of the impregnant and in various impregnated weight percentages was not of significance. Generally, the ratio was decreased in order of DABCO($R=-C_2H_5$)< piperazine($R=-C_2H_5$)< DABCO($R=-CH_3$)< piperazine ($R=-CH_3$) at the impregnation level of less than 2% weight and in order of piperazine($R=-C_2H_5$)< piperazine($R=-CH_3$)< DABCO($R=-CH_3$)< DABCO($R=-C_2H_5$). Again, these tendencies of decreasing the penetration ratio were in accord with those observed at 30°C, even though the scale of the difference was of a minor value. Difference of the penetration ratio was in parallel with increasing the impregnation weight percentage except the case of DABCO($R=-C_2H_5$), but the deviated difference was of a minor value. Especially, piperazine derivatives showed the lowest penetration ratio at the impregnation level of 6.0% weight, which may be attributed to the enhanced radioactive exchange reaction as observed in cases of table 4 A and table 6 A reaction conditions. At this temperature, the operation temperature of the ESF carbon filter system, the effective impregnant of a carbon may be covered by all of mono alkyl iodides studied and piperazine derivatives at the impregnation level of 6.0% showed the least value of the penetration ratio.

3.5 Overall Review on Data for the Effectiveness of a Mono Alkyl Iodide as an Impregnant.

Reviewing data described in 3.1-3.4 above, the following discussions were considered to be valuable for selecting an effective impregnant of an activated carbon of the nuclear grade. For convenience sake, DABCO($R=-C_2H_5$) and piperazine ($R=-C_2H_5$) were chosen to evaluate their effectivenesses as an impregnant on basis of the ongoing overall review on Data.

A. The relative reactivity data of methyl iodide with mono alkyl iodides in alcoholic solvent systems afforded the basic conception on the relative effectiveness of these chemicals as an impregnant. Thus, DABCO($R=-C_2H_5$) could be considered

as the better candidate for the impregnant than piperazine($R=-C_2H_5$).

B. Radioactive isotope reactions of mono alkyl iodides with radioactive methyl iodide varying the relative concentration of the tracer solution could clarify the relativity between the addition and radioactive exchange reactions in the reaction system. Thus, DABCO($R=-C_2H_5$) and piperazine($R=-C_2H_5$) showed resembled yields of total radioactive products, though the latter compound showed the preference of the radioactive exchange reaction overcoming its poor yield of the addition reaction. Judging from the yield of total radioactive labelled products only, both compounds may be considered as having a resembled quality each other as an impregnant.

C. Adsorption of gaseous methyl iodide in the impregnated carbon under dried air flow showed the similar trend of increasing the adsorption yield in order of each compound as observed in the liquid phase reaction described in A. above, but the yields were dependent on the impregnation weight percentage of a sample. Thus, DABCO($R=-C_2H_5$), when it was impregnated in an activated carbon at the impregnation weight level of 2.0-4.0%, showed the better adsorption yield than that of piperazine($R=-C_2H_5$), but its adsorption yield was decreased rapidly as the impregnation weight percentage was increased up to 6.0%. Therefore, DABCO($R=-C_2H_5$) may be considered to be the better impregnant at the low level of the impregnation weight percentage of the carbon sample.

D. At 30°C, penetration ratio(%) of radioactive methyl iodide through the impregnated carbon at the low level of the impregnation weight percentage(2.0-4.0%) showed the tendency of decreasing the ratio in order of each compound equivalent to the observations in A. and C. above, whereas it became equivalent to the observations in B. above when the impregnation weight percentage was increased up to 6.0%. At 80°C, the difference of the penetration ratio of each carbon sample became of a minor value and less significance to be evaluated. Thus, DABCO($R=-C_2H_5$), when it is impregnated in an activated carbon at the level of 2.0-4.0%, may be the better impregnant to be used at 30°C than piperazine($R=-C_2H_5$), but both compounds could not show a distinct difference between their penetration behaviors at 80°C.

E. Judging from the overall review on data obtained in this systematic study, DABCO($R=-C_2H_5$), when it is impregnated in an activated carbon at 2.0-4.0% weight level, would be considered to be the best impregnant of an activated carbon effective at both 30°C and 80°C. Piperazine($R=-C_2H_5$) would be considered as the next probable impregnant at 30°C, but it could be effective enough to be compared with DABCO($R=-C_2H_5$) at 80°C.

4. Conclusion

Overall review on data obtained in this systematic study could evaluate the advantages of a mono alkyl iodide concerned as an effective impregnant and these advantages evaluated may be able to afford essential reference parameters of selecting an effective impregnant of an activated carbon of the nuclear grade. Compounds having their chemical structures differed from that of mono alkyl iodide may also be able to be evaluated for their effectivenesses as an impregnant on basis of methods developed in this study modifying the study item 1 and 2 depending on their chemical behaviors with methyl iodide.

Acknowledgement

The author express his appreciation for several parts of laboratory works performed by Messrs. Hi Suk Kang and Yong Ho Cho throughout this study.

References

1. Kim, Y.S., " A study on Adsorption Characteristics and Deterioration Pattern of an Impregnated Active Carbon under a Simulated Service Condition of the Filtering System at a Nuclear Power Plant." 20th DOE/NRC Nuclear Air Cleaning Conference, NUREG/CP-008, CONF-880822, vol. 1, p. 602-618. (May, 1989)

2. Oae, S., Hovarth, C.Z., and Harris, R., " Quaternary Ammonium Salts of 1,4-Diazabicyclo(2,2,2,)octane." J. Org. Chem., 24, 1348(1959)

3. ASTM., " Standard Test Method for Carbon Tetrachloride Activity of Activated Carbon." Designation D 3467--8(1988)

4. Indem, " Standard Test Method for Testing of Nuclear Grade Gas Phase Adsorbent.", Designation D 3803-86(1986)

5. Kim, Y.S., " Study on Adsorption Characteristics of Radioactive iodine and its Compounds." The Annual Research Report of KAERI, KAERI/RR-402/83(Dec. 1983, KAERI, Korea)

6. Indem, " Studies on Reactivities of Triethylenediamine to Alkyl Iodides.", Presented at the 31st National Organic Symposium, June 19, 1989(Am. Chem. Soc., Cornell University, Ithaca, N.Y., U.S.A.)

7. Japan Radioisotope Assoc., Lecture and Practice on Radio Isotope., p. 110 (1975, Maruzen, Japan)

8. J.L. Kovach, " Air Cleaning in Accident Condition." 18th DOE Nuclear Air Cleaning Conference, 1984 and subsequent reports concerned.

9. Deitz, Victor R. and Romans, James B., " Testing Iodized Activated Carbon Filter with Non-Radioactive Methyl Iodide." NRL Memorandum Report 4240 (May 30, 1980)

TYPE II HEPA FILTER

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Abstract

A HEPA filter conforming to the dimensional requirements of the ASME AG-I Section FD Type II adsorber cell has been developed. The Type II HEPA meets the efficiency related properties of conventional size HEPA filters, while exceeding the current HEPA filter mechanical strength requirements.

The media is the minipleat configuration in two slabs with a special reinforcing grid holding and protecting the minipleat slab in a frame which has the same external dimensions of the Type II adsorbers.

The airflow resistance of the Type II HEPA is 1.0 inch w.g. at 1000 CFM.

The NUCON^{R} commercial designation NUHEPA^{TM} is intended for use in portable air cleaning units which need to be used for either/or, both particulate and gaseous contamination control, as a method to increase flow capacity of limited cross section air cleaning units and to increase the in-place testability of multiple HEPA banks. The latter is improved by the directional turns of airflow, which greatly improves mixing.

The efficiency, mechanical strength and AG-I Code required data obtained on the NUHEPA will be presented.

Introduction

The early adsorber cells (Type I designation according to IES and other standards (1)) were identical in external dimensions and airflow capacity (1000 CFM) to the standard 24 inch X 24 inch X 12 inch HEPA filters. This dimensional and capacity equivalency has permitted interchangeability of particulate filtering and gas cleaning elements. However, the development of the Type II tray type adsorbers in the US, dictated by the desire to decontaminate organic iodines also - which required an approximate doubling of gas/adsorbent contact time - resulted in an adsorber cell with significantly different shape and airflow capacity from that of conventional HEPA filters.

However, there are numerous cases where HEPA filter - gas adsorber interchangeability is still important, even if the airflow capacities are not identical. There are also often multiple HEPA filter banks in series where the directional change caused remixing of entering and exiting airflow can be a significant advantage both for testing and operational reasons.

The above consideration and the potential reduction in HEPA filter plenum volumes led to the development of the NUCON designation NUHEPA Type II HEPA filter.

Description of the NUHEPA Filter

The NUHEPA frame can be constructed from aluminum, stainless steel or appropriate polymeric materials such as polycarbonate (for incineratable filters.) The framework external dimensions are identical to the dimensions of the Type II adsorber cell as specified by the ASME AG-I Code Section FD (2) which are:

Face Plate: 25 3/4 inch X 7 3/4"

Body: 24 inch X 6 1/4 inch X 30 inch

The media is the minipleat configuration in two "slabs" sealed into the frame channels by conventional potting compounds. A special reinforcing grid holds and protects the minipleat on both sides of each pleat. This construction method increases shock resistance of the minipleat. The face plates and handles of the NUHEPA are similar to that of the Type II adsorber except for the face plate slot which is somewhat larger to decrease the pressure drop and permit flows as high as 1000 CFM through a 30 inch long unit.

The Type II NUHEPA filter meets the 99.97% efficiency for monodisperse 0.3 micron DOP aerosol removal when the minipleats are constructed from appropriate media meeting the requirements of MIL-F-510790 (3). The NUHEPA passes the rough handling test of AG-I Section FC in all three axes. The fire resistance of the new design has not been tested yet, but it is expected that, due to the properties of its components, it will have at least as good a fire rating as a conventional HEPA filter. Figure 1 shows an aluminum frame NUHEPA.

Companion: Particulate Filters and Moisture Separators

Identical Type II shape particulate prefilter family has been developed, designated NUPRE™ filter. For applications requiring low efficiency removable filter media, mats are placed between the grids of the Type II framework. The media is disposable and the framework is reused. For medium efficiency prefiltration, the structure contains an appropriate efficiency pleated media.

The identical shape holding frame with rod reinforcement contains stainless steel or stainless steel/glass fiber knit mesh pads for liquid droplet coalescing use. In moisture separator application, the units are used in a vertical orientation to assure even capture efficiency through the slabs, prevent liquid blowout from the slabs and give even, easy coalesced water drainage. In this design, the bottom of the vertically placed holding framework has drain holes in the face plate.

Applications of the NUHEPA and NUPRE Filters

Figure 2 shows a NUCON portable nuclear air treatment system designated P-NATS™.

The same basic housing and framework can be used for various applications. As an example, the following combinations of filter

and adsorber combinations can be used.

Mode 1: 1st unit 1000 CFM NUHEPA
2nd, 3rd, 4th units in parallel each 333 CFM Type II NUCELL^{R} adsorbers
5th unit 1000 CFM NUPRE medium efficiency filter
(To prevent clogging the HEPA filter, a pleated prefilter is used in the down flow inlet framework.)

Mode 2: 1st unit mat type filter without framework (on empty)
2nd , 3rd, 4th units 1000 CFM NUHEPA each for a total of 3000 CFM high efficiency particulate clean-up
5th unit empty

Mode 3: 1st unit 1000 CFM NUCELL carbon guard bed
2nd, 3rd, 4th units 333 CFM each NUCELL carbon beds
5th unit 1000 CFM medium efficiency NUPRE filter

Therefore, the flexibility of a portable air cleaning unit permits its application to any clean-up function which may occur in high efficiency filtration or adsorption application.

Another example of NUHEPA use is in multiple bank HEPA filtration where the directional turns in the airflow permit easier leak location by sampling in a single plane in front of the inlet slots where a well mixed air/aerosol mixing occurs as shown in Figure 3.

Of course, the potential space saving and dimensional flexibility through the use of NUPRE and NUHEPA filters is obvious, the same face area that holds a conventional 1000 CFM pre or HEPA filter, can hold 3 each NUPRE or NUHEPA filters with a total flow capacity of 3000 CFM.

When the Type II filters are operated in a flow mode through the inlet slot and outward through the slabs any hazardous contaminants would be mainly trapped in the interior permitting the transfer of contaminated filters with less hazard of redispersion.

Conclusion

The development of the pre and HEPA filters in the exact external dimensions of the Type II adsorber cells permits improvements and greater flexibility in the construction of high efficiency air cleaning systems.

References

1) IES CS-8T Standard for High Efficiency Gas-Phase Adsorber Cells.

2) ASME/ANSI AG-1-1988 Code on Nuclear Air and Gas Treatment.

3) MIL-F-51079D Filter Medium, Fire Resistant, High Efficiency.

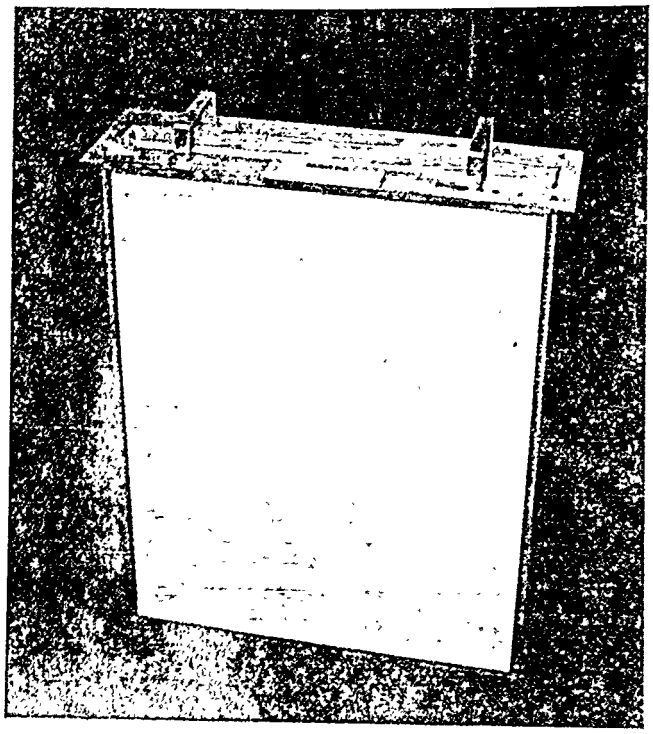


FIGURE 1

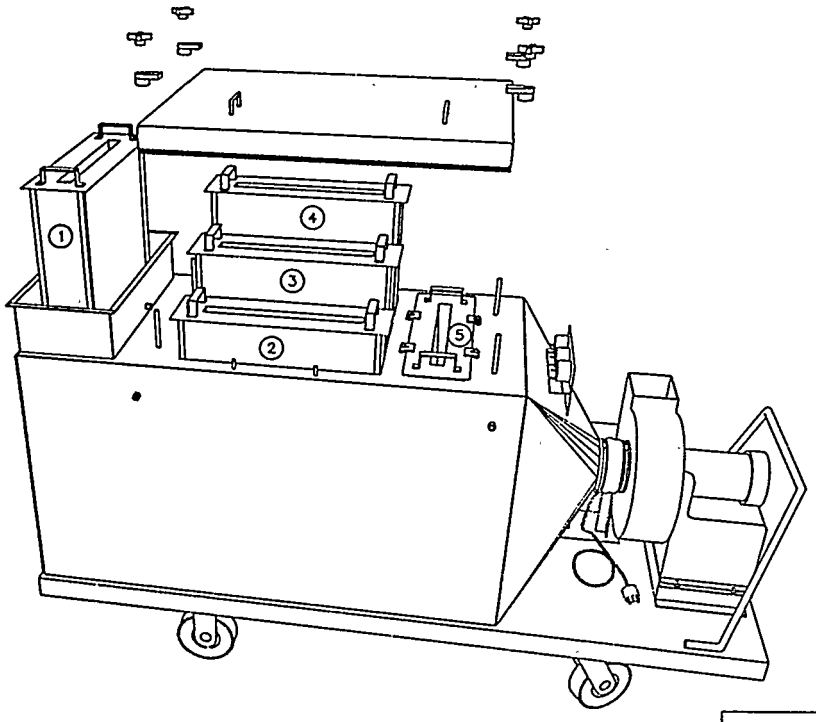


FIGURE 2

NUCON®
7000 Huntley Road Columbus, Ohio 43229-1035

1000 CFM PORTABLE FILTRATION SYSTEM – NATS

DWG TITLE: P–NATS™

DATE: AUG 1990 | DWG No.: 9022–SK50101 | REV: 1

D:\MISC\SK50101.DWG

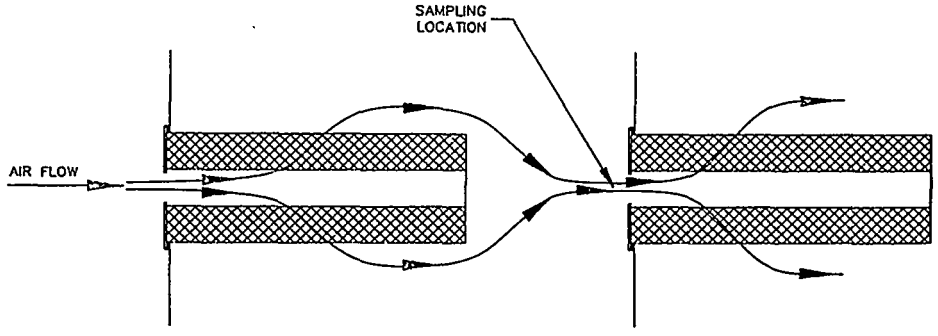


FIGURE 3

NUCON®
7000 Huntley Road Columbus, Ohio 43229-1035
DWG TITLE
DATE DATE
DWG No. 04UA-SK101-01
REV 0
D:\MISC\SK101-01.DWG

DISCUSSION

PORCO: I noticed that on the filter bank there was a separatorless design. Have you done any heated air tests or any tests on strength?

KOVACH: We have not performed the full MIL-282 test yet. The sealing we are using is only around the perimeter and we believe that on a total area basis, we have significantly less glue than in conventional mini-pleat filters. Therefore, we do not expect to run into the high organic content problem on which some of the mini-pleat filters are failing.

PORCO: My comment was not directed to combustion but to strength. When you don't have adhesives in there, you have a weaker slug, especially considering the amount of face area.

KOVACH: There is a much stronger face plate screen than is normally used to hold it together. In fact, the media can rest on the grid safely when vibrations are absent.

CONFIRMATION OF AUTOMATIC FLOW CONTROL FOR A GASEOUS EFFLUENT SAMPLING SYSTEM (U)

by

Stephen A. Epperson
Westinghouse Savannah River Company
Savannah River Site
Aiken, South Carolina 29808

ABSTRACT

The Department of Energy Orders require continuous isokinetic sampling of potential radionuclide release points to the environment. To achieve continuous isokinetic sampling under fluctuating flow conditions, flow control valves are installed to match the sampling flow rate to the exhaust flow rate. If the flow control valves are not operated properly, sampling is not isokinetic. The system's operating performance can be verified by superimposing the plots of the daily release and sampling volumes. When the variance in the daily sampling volume plot properly tracks the daily release volume the system is operating and sampling isokinetically.

Introduction

Effluent Sampling Flow Control

There are two types of control systems for gaseous effluent sampling systems: manual and automatic. Manual control is accomplished by adjusting the sampling flow rate with a flow control valve until it is equivalent to the stack flow rate. Typically, manual control is used on exhaust systems that have a constant flow rate. If the stack experiences fluctuations in flow, as a result of equipment operation (i.e., doors opening and closing, etc.), it may be necessary to install an effluent sampling system with an automatic flow valve.

Basically, automatic flow control is achieved by continuously comparing the flow rates in the stack and sample line. If the two rates are not within a specified range, an automatic control valve is adjusted until the two flow rates are within that range.

System Characteristics

The system of interest is situated on a new facility; therefore, no historical operational data is available. The volumes released and sampled are recorded on a six digit, non-resetable, flow totalizer. Daily, a technician manually records the totalizer reading onto an Environmental Release Sample Data Sheet. These data sheets were used in the analysis.

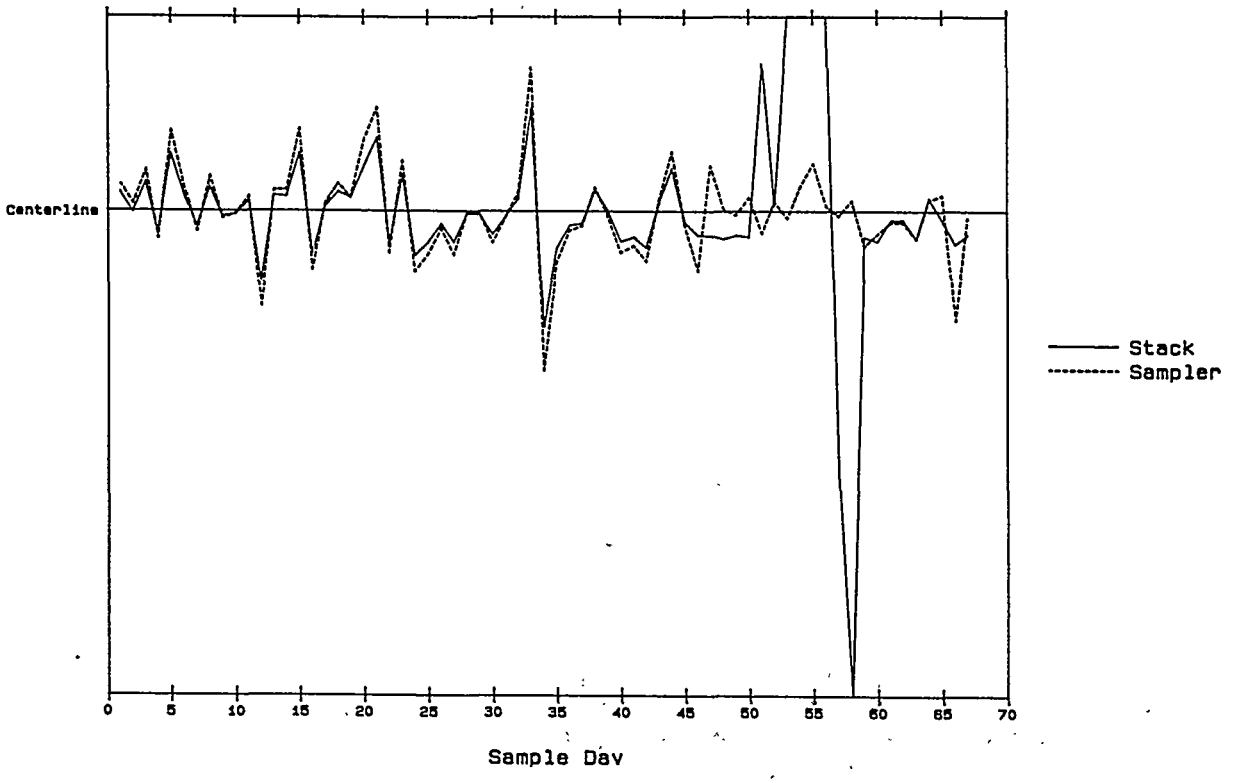
Superpositioning Method

Beginning with the daily totalizer reading for the stack and sample, the daily stack or sample volumes can be easily determined by subtracting the totalizer reading for one day from the following day. It should be noted that if the totalizer is scaled (e.g., "reading" x 10,000), this scaling may be ignored, and the actual totalizer reading may be used for simplicity. The next step is to calculate the average of the stack and sample volumes. Once this is done, find the difference between the Average Sample Volume and the Average Stack Volume. Then by subtracting this difference from the Daily Sample Volume, a new set of data, the Adjusted Daily Sample Volume, is created. Plot this new data set along with the Daily Stack Volume. The resulting graph is a superpositioned graph of the two data sets.

Figure 1 is an illustration of the superpositioned graph. As can be seen, there is excellent correlation between the stack volume that is exhausted and the sampled volume for sample numbers 1-45. As a result, we would expect that the sampling system is correctly identifying the changes in stack flow rate, and adjusting the sampling flow rate properly. It should be noted that in order to show that the stack is continuously being sampled isokinetically, a graph depicting velocity versus time would be more appropriate. In a sense, that is what is shown in Figure 1.

Figure 1

Superposition of Adjusted Daily Stack and Daily Sample Volume



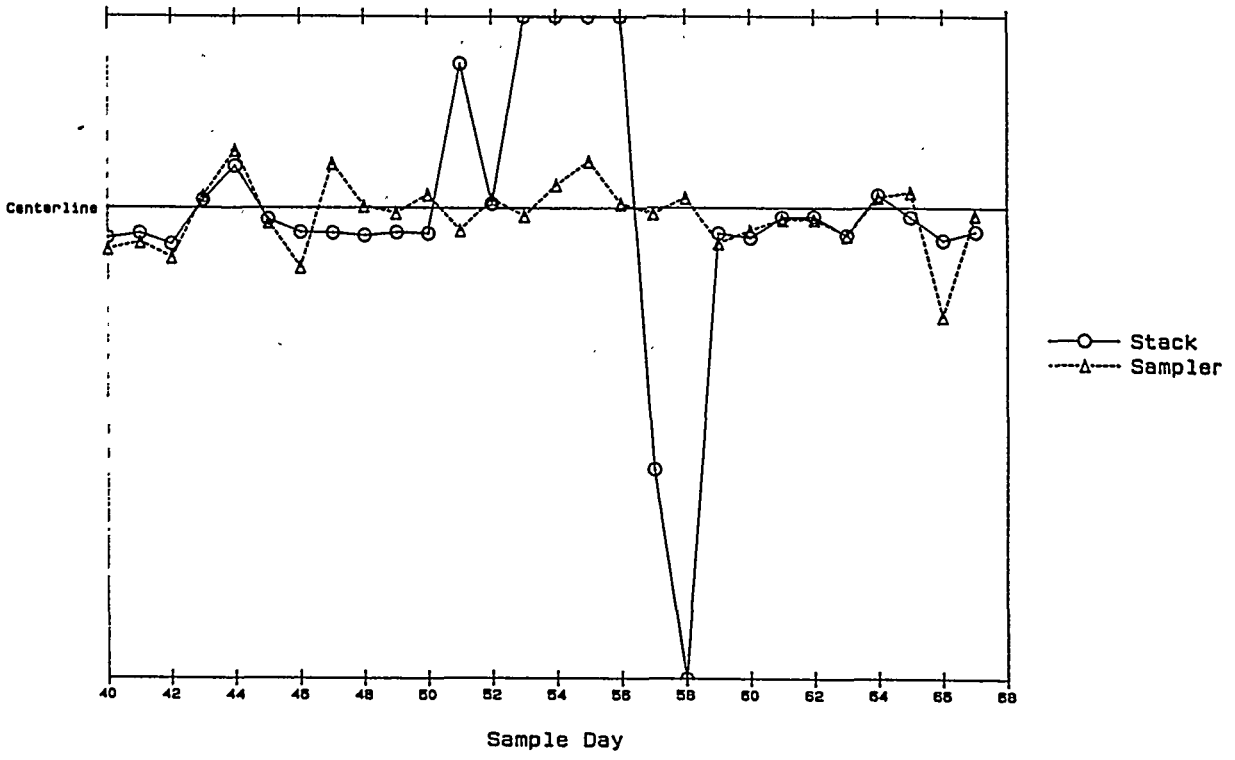
Each sample number corresponds to a day. Since the daily volumes are known, the dimensions for the stack and sampling line could be factored in. However, this would only result in shifting the values of each data point. The resulting graph would be identical to Figure 1 except the daily average stack and sampling velocity versus time would be illustrated.

Analysis of the Graph

As previously stated, Figure 1 shows that the sampling system was operating properly through Sample Numbers 1-45. Figure 2 shows Sample Numbers 40-67 from Figure 1. At sample Number 46, the curves begin to deviate until Sample Number 59 when the system appears to be working properly. In examining Figure 2, one immediately notices the flat section from Sample Number 53-56 on the stack curve. For these days, the stack totalizer reading was incomplete (not all six digits recorded), or not recorded at all. As a result, the volume released was averaged over these days.

Figure 2

Superposition of Adjusted Daily Stack and Daily Sample Volume Sample Days 40-67



The sampling system primarily consists of electronic components, and these components are not designed to detect a failure, and then correct it. Therefore, there are only two possibilities that could explain the temporary erratic behavior of the system: 1) a system failure was detected by the technician and repaired, or 2) The technician was at fault.

For isokinetic sampling, the ratio between the stack and sample volume should remain constant. In examining the original data from the Environmental Release Data Sheets, the ratio of the stack and sample volumes between Sample Numbers 45-59 was determined. This value was compared to the same ratio taken between Sample Numbers 1-15. The two values were within 1.5% of each other. This indicates that over the time period represented by Sample Numbers 46-58, the system was operating correctly and isokinetic sampling did take place. In this case, it appears that human error is to blame for the anomaly.

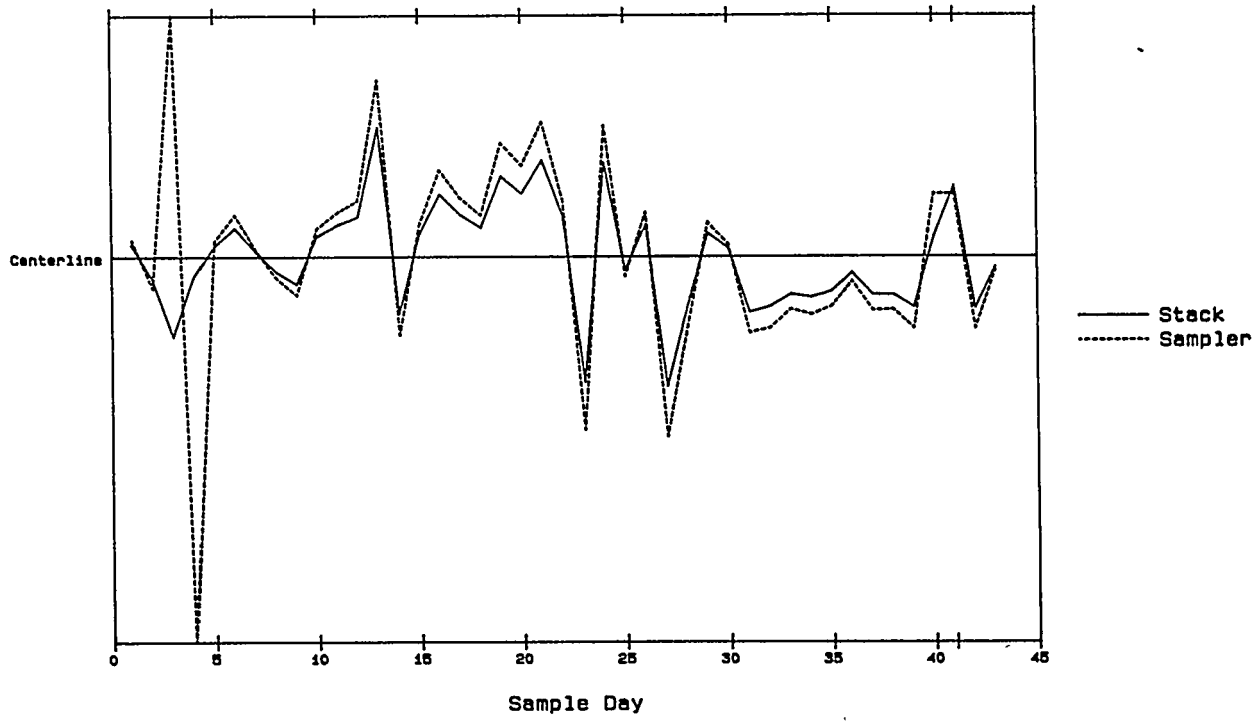
Superpositioning Program

The entire superpositioning method used here is ideal for the computer. Although the calculations are simple, they are repetitious. In conjunction with a graphics package, a program can be written that will take the input data (daily totalizer reading), and produce a graph of the superpositioned curves. Moreover, the program can be designed to search for, average, and mark bad data points. For example, if the totalizer reading is not taken one day, the user could input a code word or character. Upon detection of this error, the program would then average over that range, and mark the graph with an error message.

The graph shown in Figure 3 was created with just such a program. Normally, the graph would be reproduced in color allowing the RED ticks at Sample Numbers 40 and 41, which indicate the averaged data points, to be more apparent.

Figure 3

Superpositioned Graph as Created by OVERLAY



Red ticks indicate averaging of data.

Additional Options, Methods

Since the data has been manipulated to allow for the superpositioning, no values are indicated on the graph. One programmatic option is to graphically show the degree of flow fluctuation. That is, determine ±10%, for example, of the centerline value, and represent these values with gridlines similar to the centerline. With this, the analyst is able to quantify the flow variability.

The method described here is not the only method that can be done that will verify the operating ability of a gaseous effluent sampling system with automatic flow control. As stated earlier, the ratio of the stack and sample volumes should remain constant. Even though a horizontally, straight line should result when the ratio is plotted, some deviation will most likely appear. However, these points should be within the operability range defined by the system's specification.

ADAPTATION OF NUCLEAR FILTRATION TECHNOLOGY TO THE DEMILITARIZATION OF CHEMICAL WEAPONS

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San Francisco, CA 96305

Abstract

JACADS is an acronym for, Johnston Atoll Chemical Agent Disposal System. This presentation is to share, how the Nuclear Air Cleaning Technology is being applied in other areas, specifically the U. S. Army Chemical Agent Munition Disposal Systems. JACADS is located in the central Pacific, near the intersection of the international date line and the equator, and is some 825 miles west-south-west of Honolulu. This tiny atoll, which is a U.S. possession, is ideally suited for a pilot plant of this type, both due to its remoteness and the fact that the trade winds (westerlies) are continuous towards the vast empty Pacific. The destruction of these weapons is a result of bilateral agreements between the U.S. and Soviet Governments for chemical disarmament. The weapons involved, include various rockets, projectiles, mines, bombs, containers and etc. that are filled with any number of "mans inhumanity to man", including nerve agents, both GB and VX, and blistering agents such as mustard gas.

Incineration

JACADS is essentially a set of four high temperature furnaces, and their support utilities. A munition round is brought into the plant, from its downrange storage bunker, where it is prepared for processing and drained of its liquid agent, inside of an Explosion Containment Room (ECR). That agent is then stored in its liquid state, for eventual incineration in the Liquid Incineration Furnace (LIC). The munition is further processed and broken into smaller pieces, then sent to a Deactivation Furnace System (DFS) which again, is behind a blast containment enclosure. Dunnage, accumulated from the transport process, and other potentially contaminated waste is disposed of in the Dunnage Furnace (DUN). Other metal parts, of a non-energetic or explosive nature, go to the Metal Parts Furnace (MPF). Each of these furnace designs incorporates levels of combustion and residence times, with afterburners, to ensure complete destruction of the agent. The furnaces effluent, which is now an industrial waste, is then processed through a Pollution Abatement System (PAS), resulting in salt brines and scrap metals.

Filtration

The JACADS filtration system, is a direct adaption from existing nuclear air cleaning technology. The "bible" for this technology is MIL-HDBK-144, MILITARY HANDBOOK, Engineering Design Handbook For Air Cleaning For Chemical Demilitarization, and is in part excerpted from, ANSI/ASME N509/N510.

Figure 1, depicts the Munition Demilitarization Building (MDB) Cascade Ventilation System. The flow path, from left to right, takes 100% outside air, through demisters, prefilters and bag filters, cooling coils and delivers the conditioned air to the MDB clean areas. The air is then distributed through three pressurization zones, category C, B and A, ranging from approximately a negative quarter inch (C) to approximately a negative 3 inches (A), water column. Each compartment within the building, and from zone to zone, is air balanced to ensure the design mass flow rates (compartment change rates, vary from 6 changes per hour to 20 changes per hour) are maintained. The concept, like any nuclear power plant, is to continuously move the air from clean, to progressively more "dirty" areas, enroute to the filtration system. The air path through the furnace rooms is shown on the bottom, however is not conditioned. Furnace room air is normally exhausted through the respective furnace while in operation, or when shut down to the filters as shown.

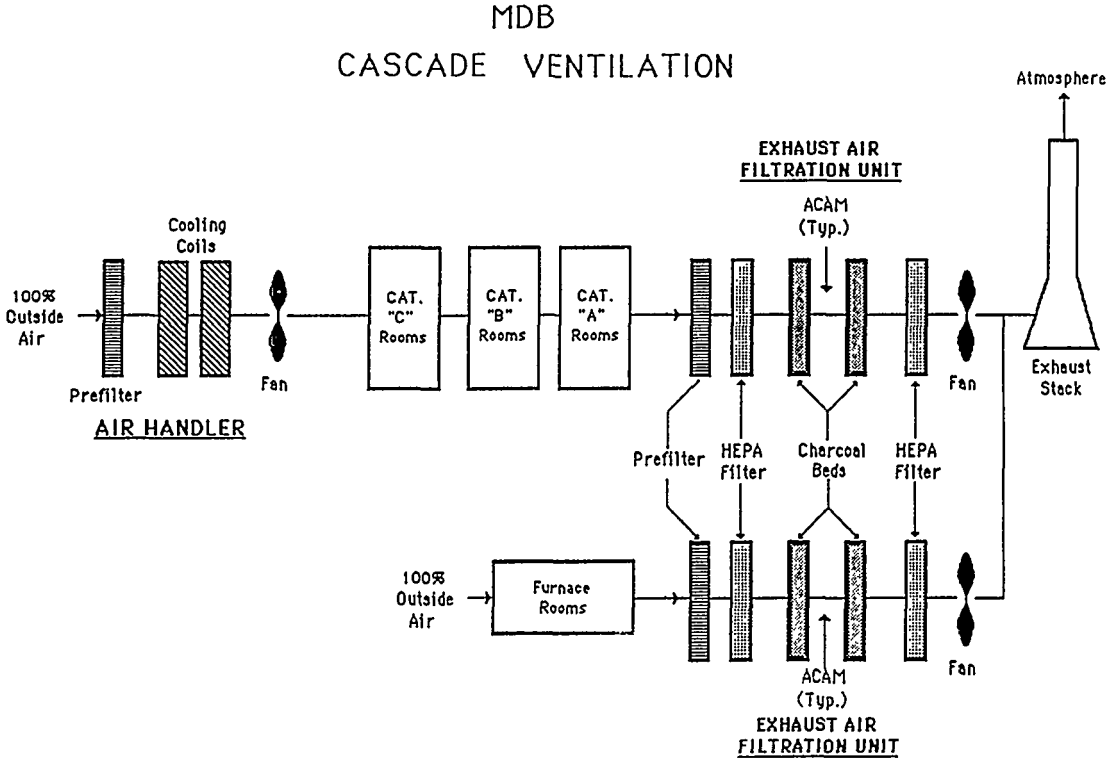


Figure 1

Figure 2, provides more detail of a MDB filter plenum. Monitoring is provided to determine the level of filter loading, by use of differential pressure instruments across the particulate media. Gas phase monitoring is accomplished by ACAMS (Automatic Continuous Air Monitoring System), the heart of which is a gas chromatograph, sampling down stream of the first, second and fourth beds. Each bed consists of 48 Type II trays (2304 total at JACADS) with approximately 113,000 lbs. of unimpregnated coconut based charcoal. These beds are challenged in accord with the precepts of ANSI/ASME N510, except that the level of required efficiency is a minimum of 99.99%. The HEPA filters are challenged to the standard value of 99.97%.

MDB - EXHAUST AIR FILTRATION UNIT

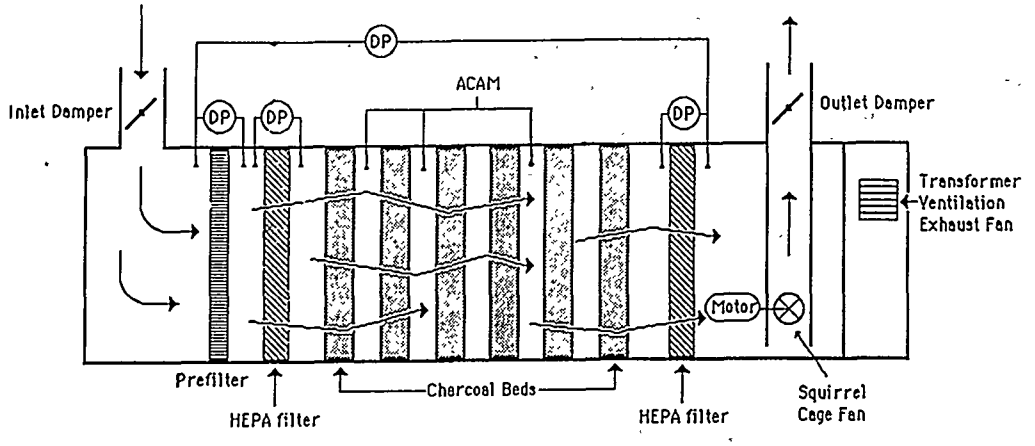


Figure 2

Summary

In summary, figures 3 and 4 depict actual Program Logic Controller (PLC) advisor screens, showing the system status in operation. The supply and exhaust systems reflect the speed, flow, delta P and temperatures as they existed at that point in time, and may be used to determine that the system is meeting the safeguard, Engineering Controls function. These screens are in reality displayed in color, and provide the means to remotely control that equipment. The system(s) status, parameters and alarms are further recorded in the Process Data Acquisition Report (PDAR) for retrieval and review.

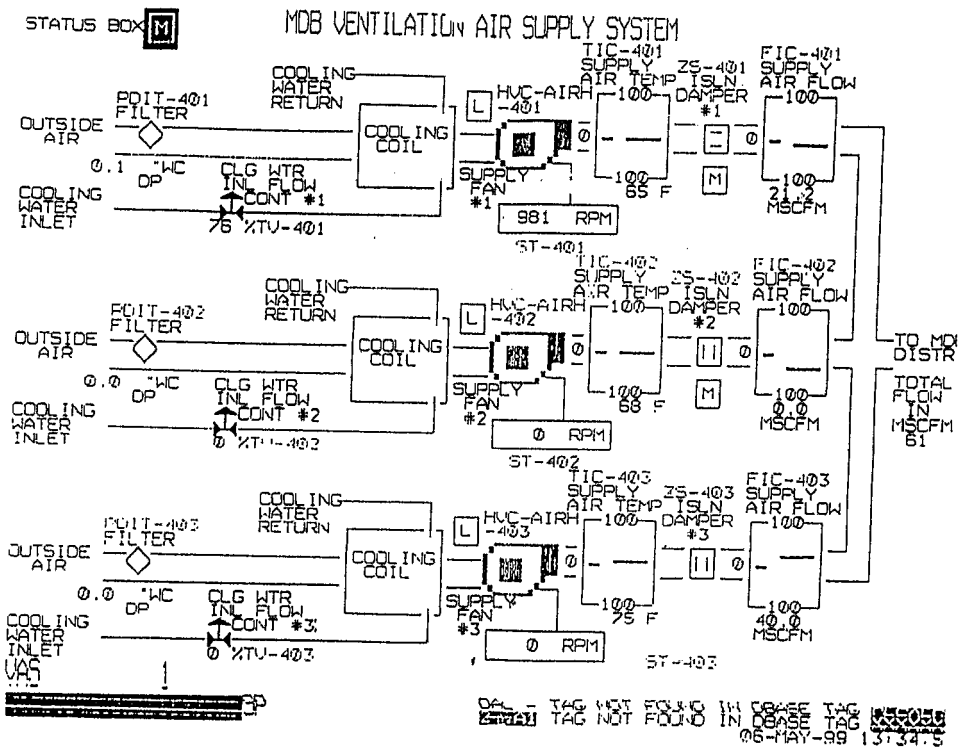


Figure 3

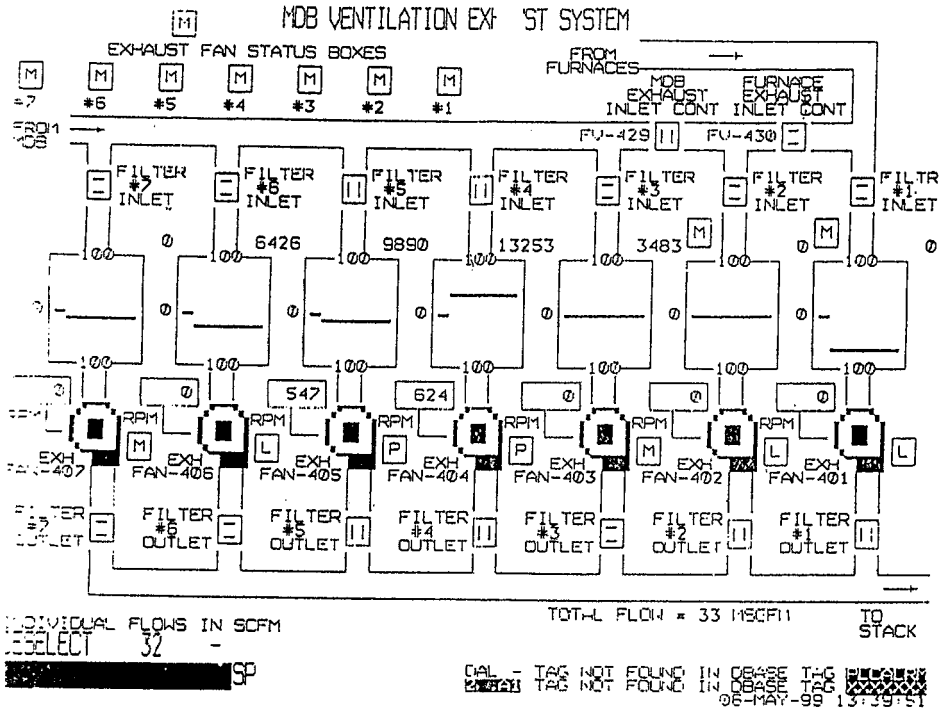


Figure 4

DISCUSSION

FIRST: I think you have the kinds of systems Jim Bresson was asking us to put into effect for the nuclear industries because these are really high-powered. Would you tell us what other locations have similar facilities?

HALL: Tooele, Utah has some prototypes of the pilot plant that exists at JACADS although not quite in the same configuration but with many of the same components. Others are planned through the years at other sites state-side, but I am in no position to discuss where or when they are going in.

TECHNICAL BRIEF: STAINLESS STEEL FILTERS FOR NUCLEAR AIR CLEANING.

Dr. Lawrence D. Weber
Pall Corporation
Scientific and Laboratory Services Department (SLS)
30 Sea Cliff Avenue
Glen Cove, New York 11542

ABSTRACT

Pall Corporation maintains extensive filter test facilities and R&D capability designed to meet technical requirements of nuclear air cleaning. SLS performs numerous standardized DOP, NaCl, and ambient aerosols filter testing in preparing filter media and designs for DOE/DOD certification. Beyond standardized testing, we encourage advancement in the state-of-the-art by responding to highly application-specific performance requirements. Some recent examples are summarized herein.

Recent Tests of Ultra High Efficiency Metal Filters

Results at Lawrence Livermore National Laboratory

In testing by Lawrence Livermore, we successfully met tight space constraints in constructing filter elements capable of 99.999997% particle removal efficiency (3.2×10^{-8} penetration) of SiO_2 aerosol at the diameter of maximum penetration, 0.1μm.[1] Results are shown in Appendix A attached. In these tests at 2CFM, pressure drop was also within specification at 4.2" H_2O.

A Compact System for Reactor Containment Vents

Taking a systems approach, we met stringent requirements for Nuclear Reactor Containment Vents in European Pressurized Water Reactor power plants. A joint effort between Pall's U.S., British, and German subsidiaries, the resulting design incorporates a horizontal pleated element configuration for Accident Mitigation by both strength and particle capture criteria, also comprising passive cooling.[2]

In support of this development, a unique test facility was constructed by Pall to facilitate filter qualification between ambient temperature and 160°C; using wet and dry TiO_2 aerosols, NaCl and Cu aerosols; both in steady or pressure shocked flow; to 5.5 bar.[2] A schematic diagram of the multitemperature and pressure dry particle aerosol test stand is given in Appendix B.[2,3] Test stand performance was calibrated for both NaCl and TiO_2 aerosols as a function of the test parameters. Examples are shown in Appendix C.[2,3]

Test results confirmed the requisite removal efficiency and physical stability of the test filter elements. Removal efficiencies for TiO_2 are reported as ß ratios = (counts in)/(counts out) over a specified size range. Percentage efficiency = ((ß-1)/ß)x100%. Efficiencies for TiO_2 particles $\geq$0.1µm are typically 10,000 or better ($\geq$99.99%), as shown in Appendix D (Figure D-1) over a range of temperatures and pressures. Figure D-2 demonstrates 99.98% efficiency for NaCl aerosol at 0.1µm, 20°C.

Further Applications

Other current applications of Pall porous metal filter technology include vents and process gas circuits of U.K. Magnox and AGR gas-cooled generating stations. Based on tests conducted for U.S. DOE, sintered metal filters were chosen for the off-gas treatment system at the Idaho National Engineering Laboratory Transuranic Waste Treatment Facility (a high temperature slagging pyrolysis incinerator was selected). The test results for over 4900 hours of operation showed Pall sintered metal filters to have an efficiency equal to or better than 99.99999% for fly ash (0-2µm range).

Developments in HEPA Filter Technology

Rationale

Pall is broadening the scope of metallic filter development to include those applications calling for conventional HEPA requirements.

Conventional HEPA filters comprise a well characterized technology but with operational limitations becoming significant in critical applications. Resin bonded glass filters provide limited mechanical strength; and sensitivity to moisture and heat.

In some HEPA applications, sand beds are employed comprising tons of sand and stone. These units cannot be performance tested to DOE specifications prior to their installation; and they are not amenable to seismic design, or useful after the onset of seismic disturbance. Further, as with glass fiber HEPA units, they cannot be regenerated if plugged.

Pall can supply Ultra High Efficiency Metal filters to meet or exceed DOE HEPA specifications for particle removal efficiency, pressure drop, seismic and thermal criteria. In addition to structural stability under extreme service conditions, they are regenerable by chemical and/or mechanical means.

Substantial geometric versatility is also achieved with this technology, at comparable size per CFM as conventional HEPAs, and extending to pleated cylindrical elements typically enclosed within a metal housing. For example, a 1000 CFM customized unit may be 20 inches in diameter by 72 inches long.

Test Results Targeting HEPA Efficiency

To demonstrate HEPA DOP removal capabilities Pall recently submitted three pleated elements to the DOE Filter Test facility at Oak Ridge. All three exceeded HEPA requirements at 0.3μm, 1" H_2O pressure differential, confirming SLS in-house test. A copy of the Test Report is attached as Appendix E.[4]

In summary, Pall draws upon an extensive, growing international data base of independent and in-house test experience in designing metal filters and filtration systems to DOE and application-specific technical requirements.

References

1. Bergman, W., Larsen, G., Lopez, R., Williams K., and Violet C., "High Efficiency Steel Filters for Nuclear Air Cleaning," Proc. 21st DOE/NRC Nuclear Air Cleaning Conference, (1990).

2. Randhahn, H., Taetz, F., Laslop, R.-J. John, "Pall Filter System for Reactor Pressure Release," SLS Study FG6BI, Pall Deutschland GmbH, (1988).

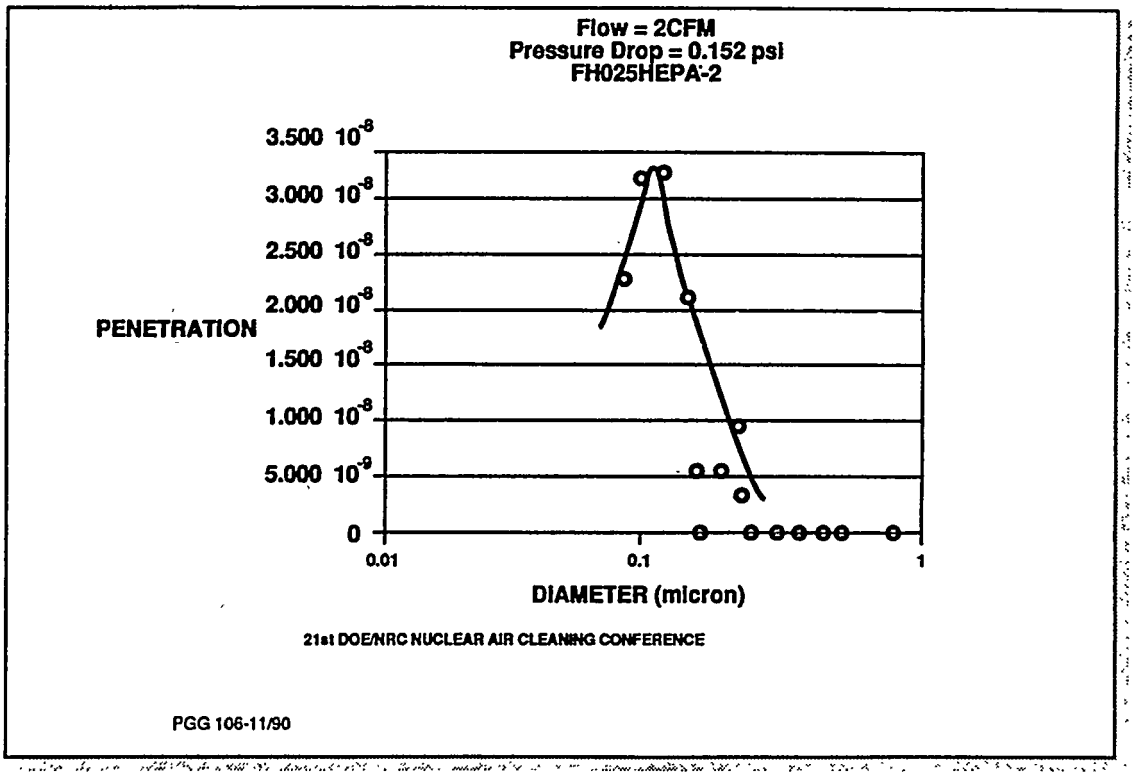
3. Randhahn, H. "The Development of All-Metal Filters for Use in the Nuclear Industry" Proc. 20th DOE/NRC Nuclear Air Cleaning Conference, (1988).

4. Filter Test and Inspection Report, Martin Marietta Energy Systems, Inc., Oak Ridge Gaseous Diffusion Plant, September 28, 1990.

APPENDIX A

FIGURE A-1

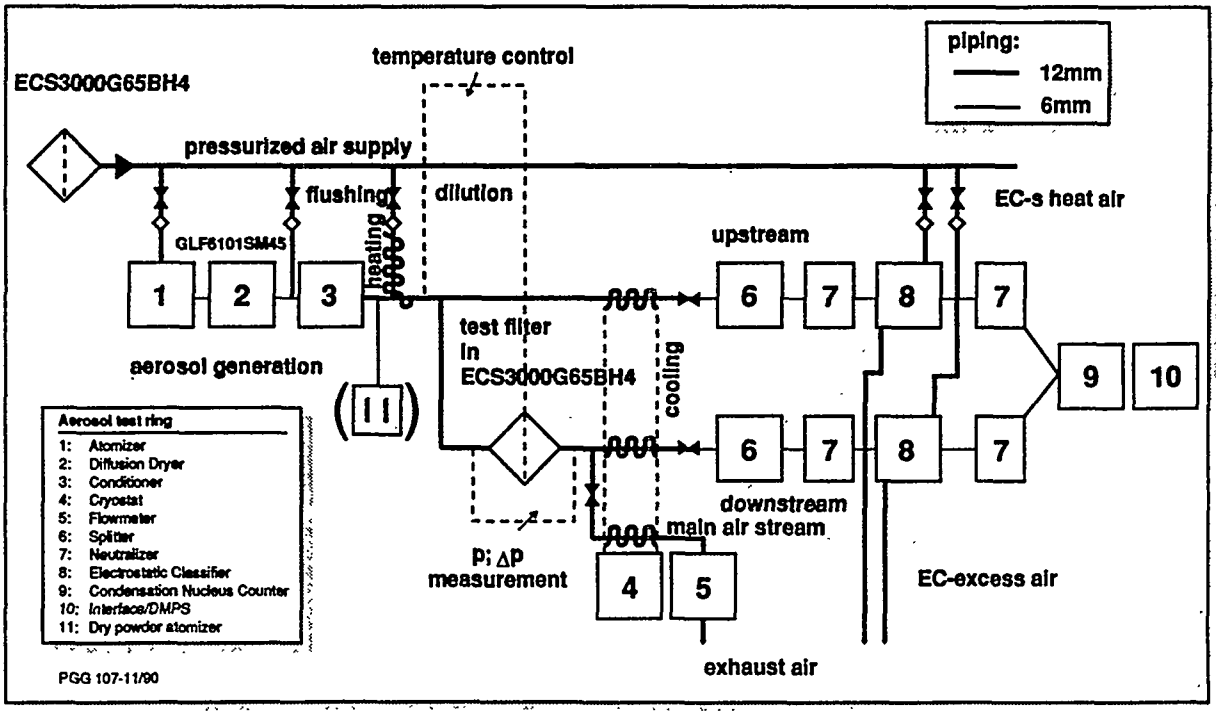
(Bergman & Colleagues, Ref. 1)



APPENDIX B

FIGURE B-1

(Randhahn & Colleagues, Ref. 2,3)



APPENDIX C

FIGURE C-1

GRAPH II

(Randhahn & Colleagues, Ref 2,3)

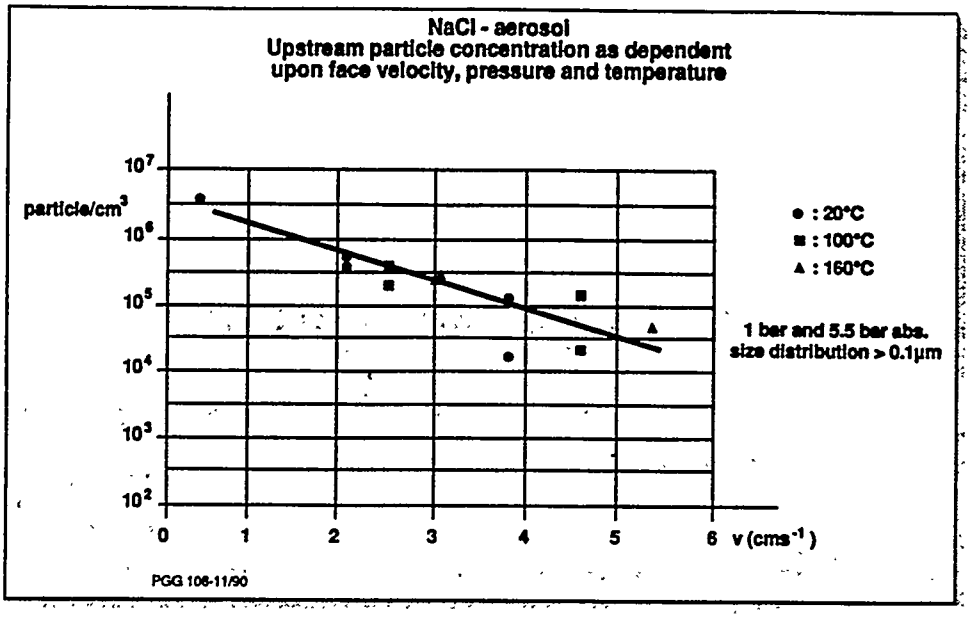
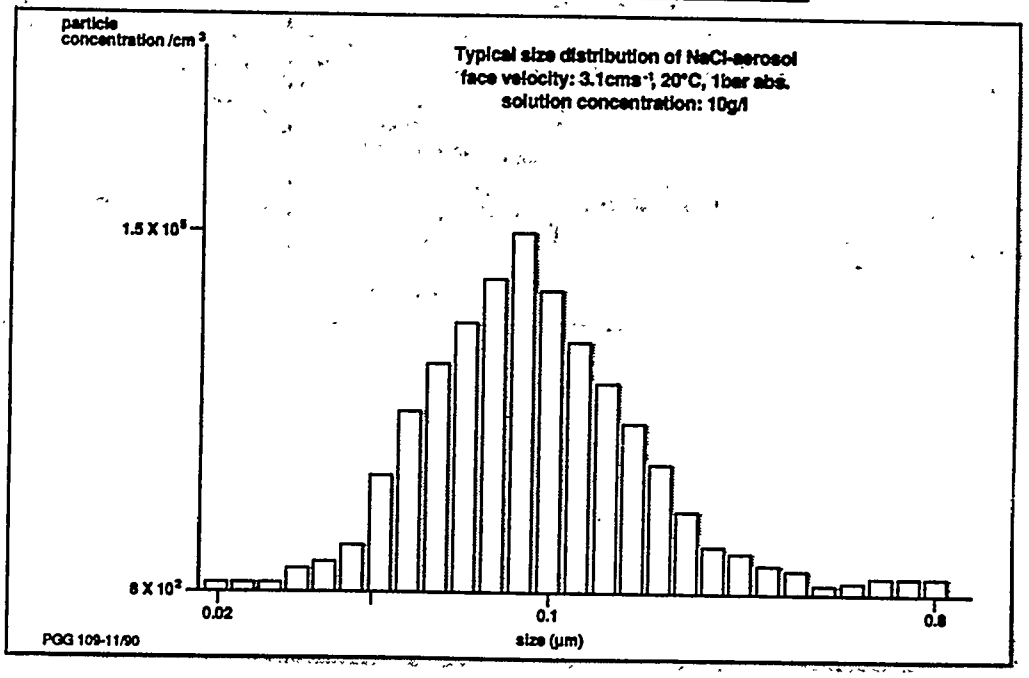


FIGURE C-2

GRAPH III

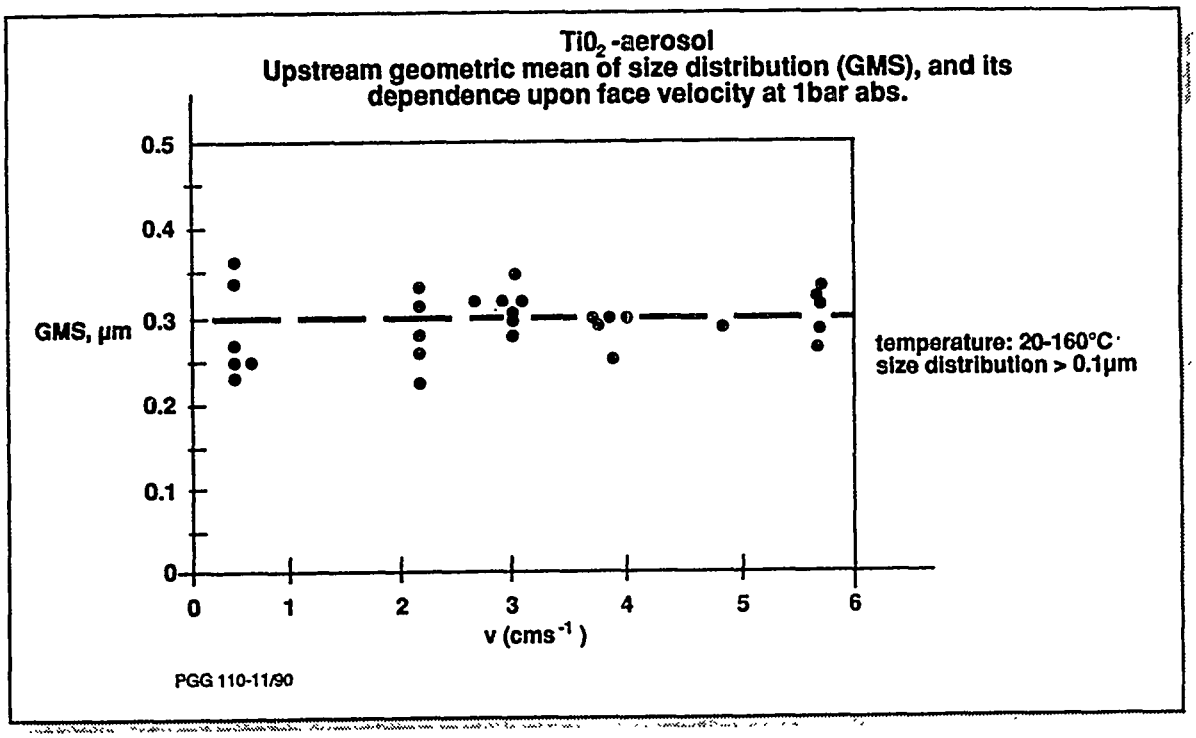
(Randhahn & Colleagues, Ref. 2,3)



APPENDIX C - (continued)

FIGURE C-3

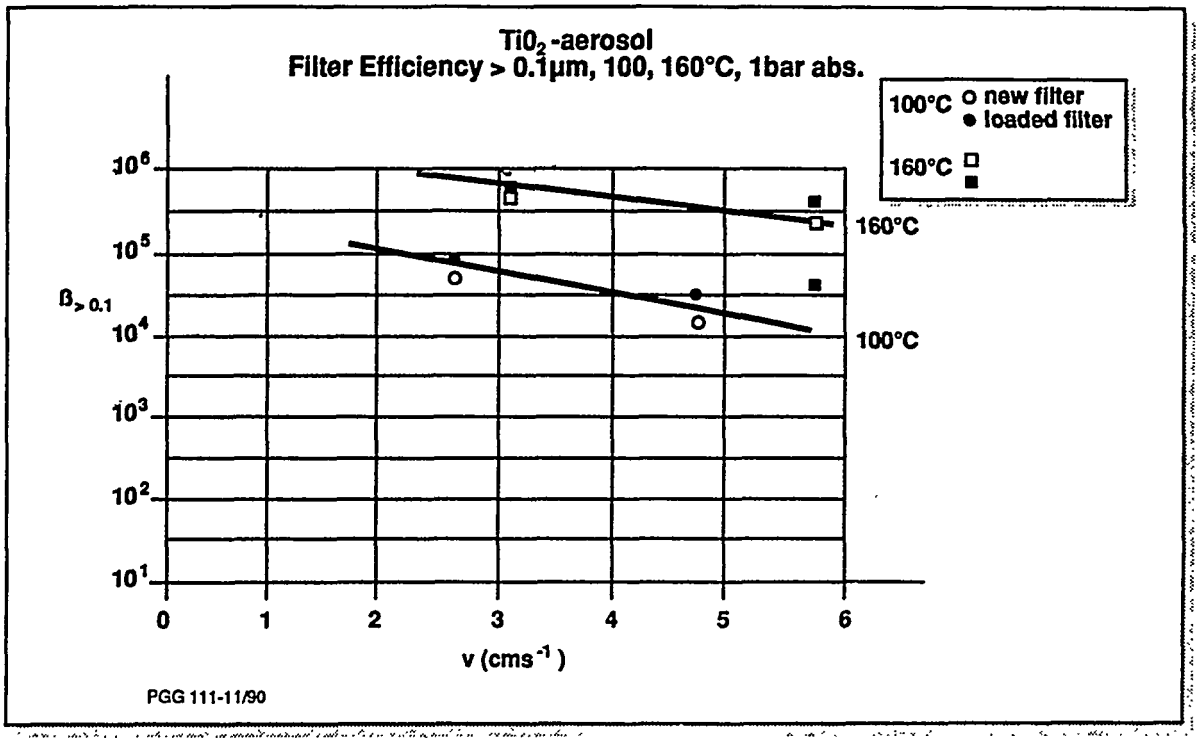
(Randhahn & Colleagues, Ref. 2,3)



APPENDIX D

FIGURE D-1

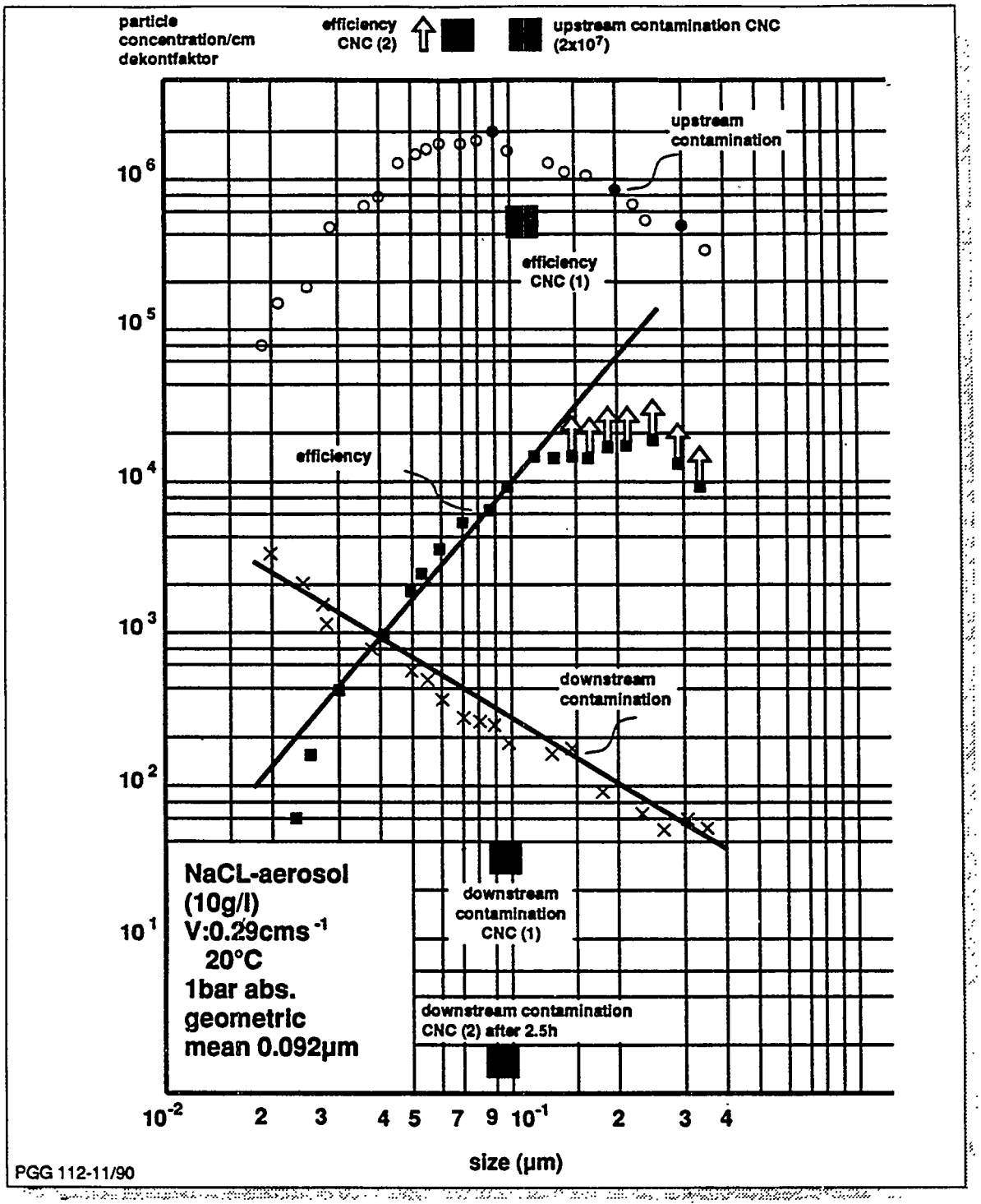
(Randhahn & Colleagues, Ref. 2,3)



APPENDIX D - continued

FIGURE D-2

(Randhahn & Colleagues, Ref. 2,3)



APPENDIX E

MARTIN MARIETTA ENERGY SYSTEMS, INC.

OAK RIDGE GASEOUS DIFFUSION PLANT

FILTER TEST AND INSPECTION REPORT

PAGE 1 OF 1

PURCHASER: Pall	DOP PENETRATION	NUMBER ORDERED	DATE RECEIVED: 9-28-90
FILTER PURCHASE ORDER NO.	RESISTANCE	NUMBER RECEIVED: 3	DATE TESTED: 9-28-90
TEST PURCHASE ORDER NO.: 95530	SPECIFICATION	NUMBER ACCEPT	DATE INSPECTED: 9-28-90
MANUFACTURER: Pall	TEMPERATURE: 71	TEST FLOW: 34 LPM	PENETRATION / RESISTANCE
FILTER DESCRIPTION: Cylindrical 2"x3"	BAROMETRIC PRESS: 75.5	OTHER	DAMAGE / OTHER
MODEL / RATED FLOW			

ITEM No.	FILTER SERIAL NUMBER	INSPECTION RESULTS	TEST RESULTS: RESISTANCE	PENETRATION @ 100% FLOW	PENETRATION @ 20% FLOW
	5		1.0	.005	
	7		1.0	.012	
	9		1.0	.004	

DISTRIBUTION: Mike McNeely, Pall Corp.

FILTER TEST FACILITY

TESTED BY: S.K. McNeely

APPROVED BY:

DISCUSSION

FIRST: Is this similar to the sintered filters that were used at the Rocky Flats incinerator by Zeigler and his people?

WEBER: It is a different type, a lower pressure drop, pleatable porous metal medium more recently formulated.

PORCO: I think there is a need for high-temperature HEPA filtration but I have a question about cost relative to conventional HEPA filters.

WEBER: The cost is higher than conventional HEPA filters in terms of the initial purchase price, but over the long-run the cost can be competitive or even less because the metal filter can be cleaned in place. In addition, it can be used in places where the conventional HEPAs can not.

PORCO: Can you give us an order of magnitude of the purchase cost?

WEBER: I would have to refer you to our commercial people for that, but it will be significantly more expensive than a HEPA of equal airflow capacity.

PORCO: A question about size. You said it was approximately the same size as conventional HEPAs. Did I hear you say 20 in. in diameter by 72 in. long? That would be quite larger than a standard HEPA filter.

WEBER: That is correct for the design format I mentioned, but it can also be modified. The reason is that we typically employ a metallic membrane-like material which we can pleat into very compact geometries giving a high surface area and low pressure drop.

BRESSON: You mentioned regeneration of the filter by chemical cleaning. What kind of chemicals are you talking about for cleaning agents?

WEBER: Any chemicals that will help loosen the contaminants and that are compatible with stainless steel, Inconel, or other alloys of stainless steel. For example, in fuel processing facilities, nitric acid is used in abundance and it would be an excellent candidate that could be recycled into the system. Organic chemicals could be used depending on the nature of the contaminant.

BRESSON: I am wondering about the creation of second generation waste, some of which might be mixed waste.

WEBER: Quite so. I think that has to be addressed. These materials could possibly generate significant concentrations of "mixed" wastes, which are RCRA regulated and present significant costs to handle technically and in a regulatory sense.

Field Measurement of the Performance of Air-Cleaning Systems to Simulated Worst-Case Hazardous Chemical Accidents Using the LLNL "S^3" Cell

James S. Johnson, Ph.D.,*
Donald H. Fearon, and Frank R. Weber
Lawrence Livermore National Laboratory
P. O. Box 808, Livermore, CA 94550

Abstract

We have identified a significant deficiency in the evaluation of critical air-cleaning systems that are planned for use involving hazardous chemical accidents. We have developed a conceptual design to test these components and systems using the Lawrence Livermore National Laboratory's Small Scale Spill (S^3) Cell located at the Department of Energy's (DOE) LGF Spill Test Facility at the Nevada Test Site in Mercury, Nevada.

Introduction

The need to assure a high degree of reliability for air-cleaning systems designed to contain hazardous materials during an accident is a recognized fact. The accepted way to obtain this reliability is by careful selection of construction material and components, as well as the use of redundant systems and components. In many cases, construction materials and components have been evaluated for resistance to the hazardous materials in question.[1–3] In some cases, however, this information is lacking and best guesses are used in the selection process. The performance of aged components in these systems is not addressed, and it has been suggested that in the case of aged HEPA filters, significant degradation in strength and performance is possible.[4] New national legislation to improve the safety of chemical plants as well as the ongoing safety concern about nuclear installations require a change in the way air-cleaning systems are designed and tested to demonstrate worst-case hazardous chemical accident performance. Air-cleaning units that contain all representative components used in the operational systems should be tested in the actual worst-case hazardous chemical atmospheres.

LLNL S^3 Cell

The Hazards Control Department of the Lawrence Livermore National Laboratory (LLNL) has designed and constructed a Small Scale Spill (S^3) Cell at the DOE's LGF Spill Test Facility at the Nevada Test Site in Mercury, Nevada. The S^3 cell (illustrated in Fig. 1 and shown in Fig. 2) is a cooperative effort with construction

*Work performed under the auspices of the U. S. Department of Energy by Lawrence Livermore National Laboratory under Contract W-7405-Eng.-48.

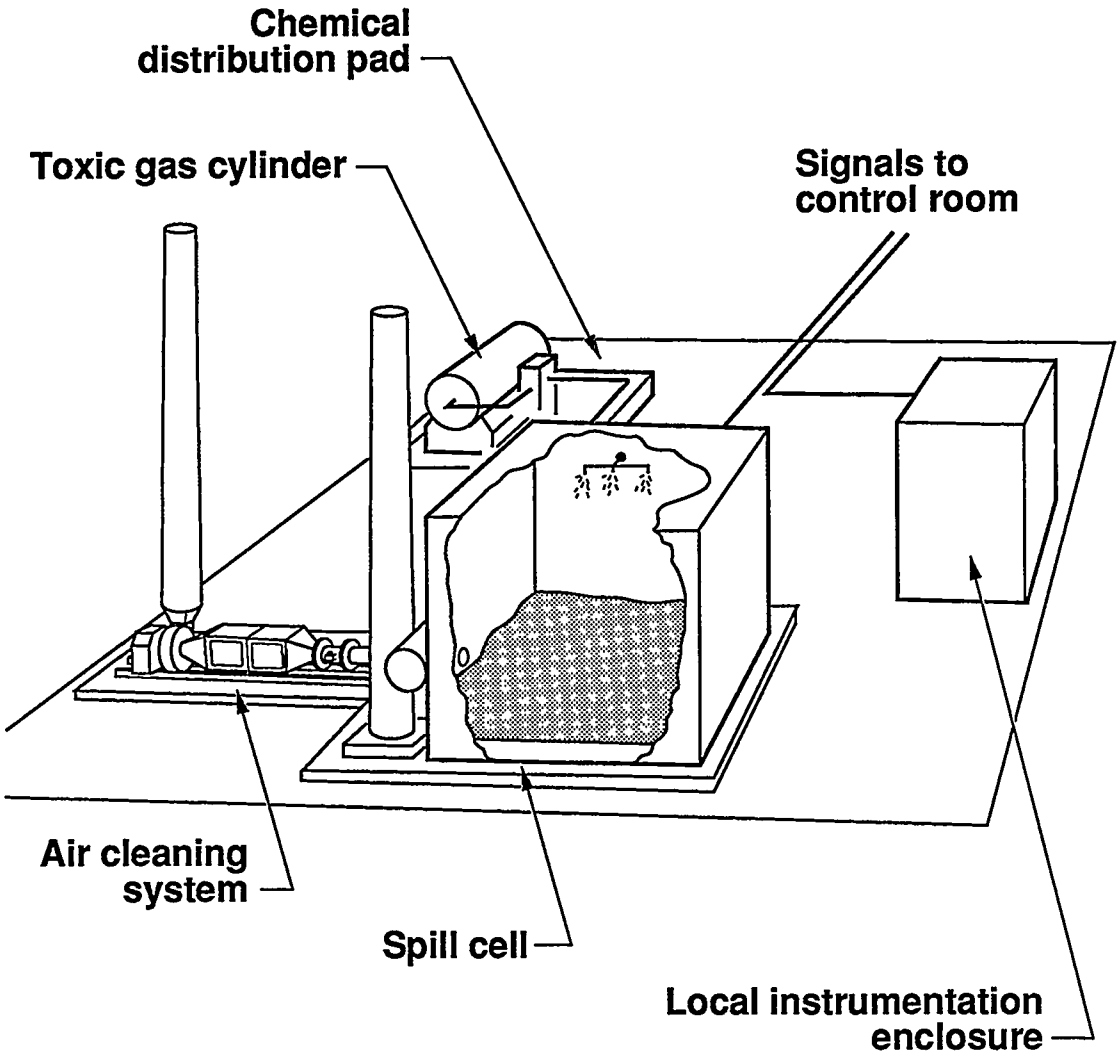


Figure 1. Drawing of the LLNL S^3 cell with proposed air-cleaning module attached.

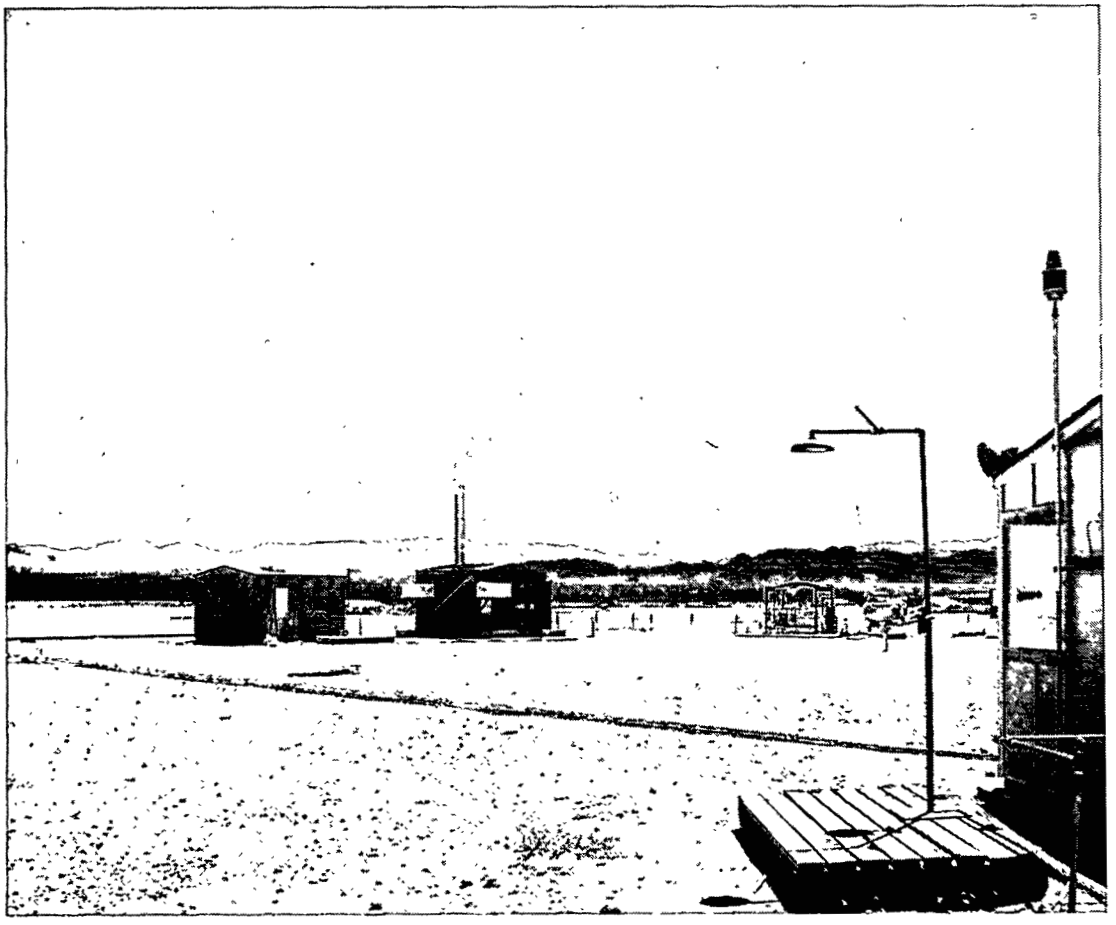


Figure 2. Photograph of the actual LLNL S^3 cell at DOE's Nevada Test Site.

funds supplied by the Occupational Safety and Health Administration, the Department of Transportation, DOE's Fossil Energy Program, and LLNL. Initial experiments using this cell will test the reliability of personal protective clothing used by emergency response staff. Chemicals that will be used in these studies are hydrogen fluoride, chlorine, and ammonia. DOE's LGF Spill Test Facility is a unique national resource that has undergone detailed environmental analysis to obtain permission to release these chemicals directly to the environment in large quantities as well as 27 other hazardous materials. The S^3 cell will be able to accommodate these chemicals as well as others after appropriate environmental review is completed for air-cleaning system testing.

Future Work

The use of the S^3 cell for the evaluation of air-cleaning systems and components will demonstrate unequivocally their ability to function as intended in worst-case hazardous chemical accidents. A conceptual design of a 1000-cfm ventilation system has been formulated. Application of this concept to the evaluation of air cleaning system performance in simulated worst-case hazardous chemical accidents is presently being considered. The effect of component aging on system performance during worst-case accidents must also be evaluated. These studies will demonstrate with actual test data the level of reliability ventilation systems can provide in simulated worst-case hazardous chemical accidents. Without this type of performance data, the reliability of the air-cleaning system will always be in question during emergencies.

References

1. G. W. Brassell and W. G. Thowaldson, "Development and Evaluation of Acid-Resistant HEPA Filter Media," in *Proc. 17th DOE Nuclear Air Cleaning Conf.* (Denver, CO, August 2–5, 1982), CONF-820833, pp. 1125–1133.

2. W. Alken, H. Bella, V. Ruedinger, and J. C. Wilhelm, "Development of a HEPA-Filter with High Structural Strength and High Resistance to the Effects of Humidity and Acid," in *Proc. 18th DOE Nuclear Air Cleaning Conf.* (Baltimore, MD, August 12–16, 1984), CONF-840806, pp. 1085–1095.

3. V. Ruedinger, C. I. Ricketts, and J. G. Wilhelm, "The Realization of Commercial High Strength HEPA Filters," in *Proc. 20th DOE/NRC Nuclear Air Cleaning Conference* (Boston, MA, August 22–25, 1988), CONF-880822, pp. 917–942.

4. J. S. Johnson and D. G. Beason, "The Effect of Age on the Structural Integrity of HEPA Filters," in *Proc. 20th DOE/NRC Nuclear Air Cleaning Conference* (Boston, MA, August 22–25, 1988), CONF-880822, pp. 366–382.

CLOSING COMMENTS OF SESSION CHAIRMAN FIRST

This concludes the Open End Session. By severely restricting presentation time we have been able to learn important aspects about eleven programs and proposals for change within the allotted time. An Open End program of this nature has become a tradition at Nuclear Air Cleaning Conferences and its continuing popularity is attested to by the excellent attendance today. To the best of my knowledge, our Open End is a unique type of session at technical meetings; it is less formal than the usual platform session but much more comfortable than a poster session. I think a short presentation time allotment tends to sharpen a speaker's thoughts so as to get quickly to the essential points of the presentation.

The presentations have offered us many new ideas and the questions and comments following the presentations have sharpened our understanding of the several presentations.

SESSION 8

A - INCINERATION AND VITRIFICATION
B - SYSTEM DESIGN

Wednesday: August 15, 1990
Co-Chairmen: R. L. Shepard
J. R. Yow

OPENING COMMENTS OF SESSION CO-CHAIRMAN SHEPARD

LFCM PROCESSING CHARACTERISTICS OF MERCURY
R. W. Goles, G. J. Sevigny, C. M. Andersen

BEHAVIOR AND CONTROL OF RUTHENIUM DURING OPERATION OF THE NEW WASTE CALCINING FACILITY AT THE IDAHO CHEMICAL PROCESSING PLANT
J. D. Christian

THE INCINERATION OF LOW LEVEL RADIOACTIVE WASTES - CURRENT STATUS
D. W. Moeller, S. W. Long

STEPS TOWARDS THE MINIMIZATION OF PARTICULATE EMISSIONS FROM A LOW-LEVEL WASTE INCINERATION FACILITY
H. Leibold, R. Mai, J. G. Wilhelm

SYSTEM DESIGN

DESIGN BASIS DOCUMENTATION FOR THE CONTROL ROOM VENTILATION/CHILLED WATER SYSTEM AT MCGUIRE NUCLEAR STATION
J. R. Hilley, Jr., R. R. Weidler

DEMONSTRATION OF CARBON-14 REMOVAL AT CANDU NUCLEAR GENERATING STATIONS
S. D. Chang, C. H. Cheh, P. J. Leinonen

CLOSING COMMENTS OF SESSION CO-CHAIRMAN SHEPARD

OPENING COMMENTS OF SESSION CO-CHAIRMAN SHEPARD

We know that vitrification and incineration are important technologies. In fact, Nuclear Engineering International in its March 1990 issue, did an extensive series of articles on vitrification as it relates to the Hanford site, the West Valley site and also the Stellarfield facility of British Nuclear Field Services. We are excited about this session this morning and, we hope we will gain some insight into what is currently taking place in the area of incineration and vitrification.

LFCM PROCESSING CHARACTERISTICS OF MERCURY

R. W. Goles, G. J. Sevigny, and C. M. Andersen
Pacific Northwest Laboratory

Abstract

An experimental-scale liquid-fed ceramic melter was used in a series of tests to evaluate the processing characteristics of mercury in simulated defense waste under various melter operating conditions. This solidification technology had no detectable capacity for incorporating mercury into its borosilicate, vitreous product, and essentially all the mercury fed to the melter was lost to the off-gas system as gaseous effluent. An ejector venturi scrubber condensed and collected 97% of the mercury evolved from the melter. Chemically the condensed mercury effluent was composed entirely of chlorides, and, except in a low-temperature test, Hg_2Cl_2 was the primary chloride formed. As a result, combined mercury accounted for most of the insoluble mass collected by the process quench scrubber. Although macroscopic quantities of elemental mercury were never observed in process secondary waste streams, finely divided and dispersed mercury that blackened all condensed Hg_2Cl_2 residues was capable of saturating the quenched process exhaust with mercury vapor. However, the vapor pressure of mercury in the quenched melter exhaust was easily and predictably controlled with an off-gas stream chiller.

Introduction

Radioactive high-level liquid waste (HLLW), a byproduct of defense nuclear fuel reprocessing activities, is currently being stored in underground tanks at several U.S. sites. Because its mobility poses significant environmental risks, HLLW is not a suitable waste form for long-term storage. High-temperature processes for solidifying and isolating the radioactive components of HLLW have therefore been developed and demonstrated by the U.S. Department of Energy (DOE) and its contractors. Specifically, vitrification using liquid-fed ceramic melters (LFCMs) has become the reference process for converting U.S. HLLW into a borosilicate glass, which is considered a suitably stable matrix for permanent geological storage. Two vitrification plants are currently under construction in the United States: the West Valley Demonstration Project (WVDP) being built at the former West Valley Nuclear Fuels Services site in West Valley, New York; and the Defense Waste Processing Facility (DWPF), which is nearing completion at DOE's Savannah River Plant (SRP) in Aiken, South Carolina. A third facility, the Hanford Waste Vitrification Plant (HWVP), is being designed at DOE's Hanford Site in southeastern Washington.

The Pacific Northwest Laboratory (PNL) has been developing HLLW immobilization technologies under the sponsorship of the DOE and its predecessor agencies for well over 25 years. Since 1973, PNL has

technically supported the design and development of the LFCM vitrification process. Much of this development work has involved nonradioactive experimental studies using a ~1/3-scale LFCM and its associated off-gas processing system. Since 1981, more than 20 LFCM tests involving 3000 h of processing have been conducted at PNL in direct support of the DWPF, the WVDP, and the HWVP.

As a result of the diverse needs and specific processing requirements associated with the HLLW at each of these DOE sites, PNL-LFCM tests have provided a wealth of information regarding the processibility of a variety of waste components under diverse process and operating conditions. The processibility of mercury is of particular importance with respect to melter operability, secondary waste stream management, and environmental compliance modeling due to the (electrical) conductivity, volatility, and toxicity, respectively, of this element and its chemical compounds. Because of the unique challenges the presence of mercury poses to LFCM processing of HLLW, this paper deals exclusively with the empirically determined processing and off-gas behavior of mercury under the various LFCM operating conditions encountered during scaled simulation testing.

Before the operating and processing conditions used during these LFCM tests are discussed, a brief description of the scaled melter and its off-gas processing and sampling systems will be presented along with feed composition data. The nature and magnitudes of melter mercury losses and the impact of these losses on the off-gas processing will then be discussed as a function of test operating conditions. Finally, the results of a destructive post mortem examination of the melter and its off-gas processing system will be used to establish processing system accumulation sites of feed component mercury.

Liquid-Fed Melter Processing and Sampling System Description

This section briefly describes the liquid-fed melter processing and analysis systems developed at PNL in support of DOE's waste isolation programs. Further information concerning the LFCM technology and its associated off-gas processing requirements is documented elsewhere.[1-3]

Melter

Joule-heated, ceramic-lined melters form the basis of the LFCM process, which is illustrated in Figure 1. This type of glass melter, developed for the commercial glass industry, is composed of a heavily insulated, ceramic-lined cavity that contains the molten glass, and corrosion-resistant electrodes. The electrical resistive heating used to maintain the glass in its molten state and to process feed chemicals into more glass is produced by passing an electric current (AC) through the glass pool via the immersed melter electrodes--this process is known as joule heating. In addition to this primary source of power, auxiliary radiant heaters located in the melter plenum above the glass melt surface are also used to increase or boost processing rates. Because joule-heated ceramic melters can attain higher temperatures than conventional melters, high-silica glasses with exceptional chemical stability can be fabricated. This

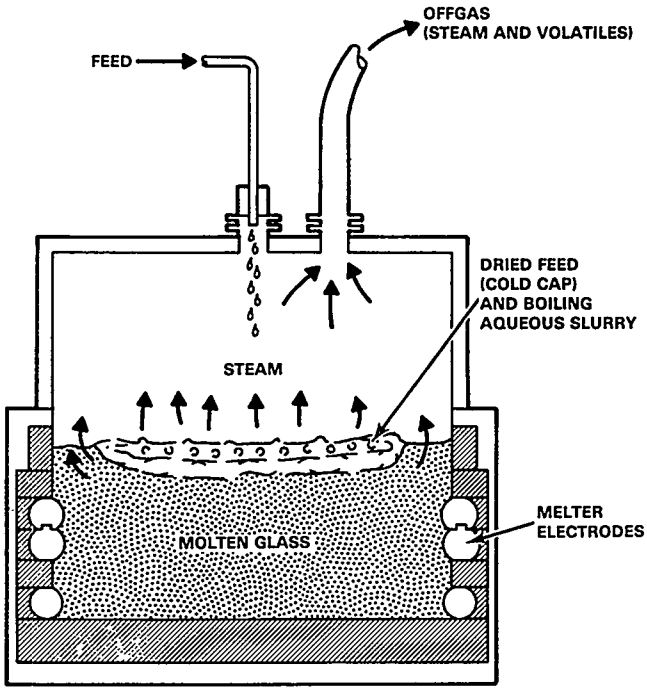


Figure 1. Liquid-Fed Ceramic Melter Process

feature is important to LFCM applications because the vitreous waste form must exhibit low aqueous leaching rates to qualify for long-term geological storage.

The use of joule-heated ceramic melters to vitrify HLLW is a straightforward extension of glass-making technology; the waste components are treated merely as a subset of the chemicals needed to produce a specific glass product. In the LFCM process, major glass-forming chemicals and/or glass frit are mixed with the HLLW stream to produce an aqueous slurry that is continuously fed to the joule-heated melter to produce a molten radioactive glass. The molten melter glass product, which commonly has a waste loading of 25 wt%, is transferred via an overflow port to metal canisters that are subsequently cooled and then sealed to further isolate the immobilized waste form.

Because of the health hazards associated with mercury and its chemical compounds, an experimental-scale ceramic melter (ESCM) was exclusively used for all melter tests involving this toxic element. The ESCM was a 1/50-scale melter with a 0.054 m^2 glass surface area and was small enough to be contained within a walk-in hood. Because it is small, melter inleakage could be successfully controlled, allowing plenum oxidizing conditions to be used as a process parameter. Figure 2 illustrates the experimental arrangement involving the ESCM and its ancillary feed and off-gas processing systems.

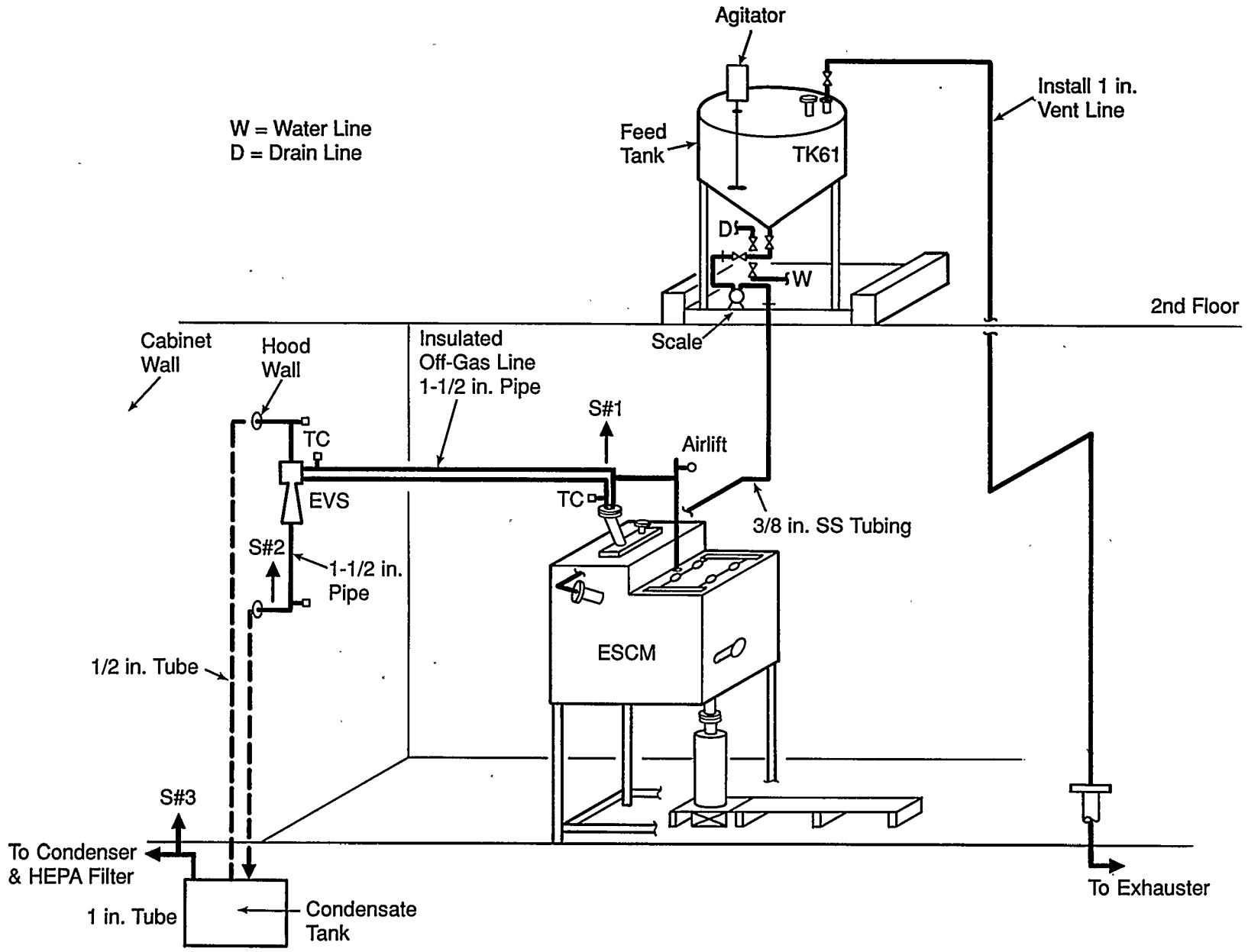


Figure 2. Experimental-Scale Ceramic Melter and Off-Gas Processing System

Feed Composition

The liquid slurry feed used for the ESCM mercury tests was a uniform mixture of simulated defense waste sludge and glass formers (frit). The composition of this slurry and the projected equivalent oxide composition of the resultant process glass are detailed in Table 1. Because mercury does not possess a stable oxide at glass processing temperatures, the process efficiency for vitrifying this element, as shown in the table, was expected to be quite low; demonstrating and quantifying this fact was one of the primary reasons for conducting the ESCM tests in the first place. The effective waste loading of the slurry used was 25 wt% of the total oxides present in the feed.

Although the composition of the liquid feed remained nominally constant throughout the period of testing covered in this report, the quantities of formic acid, NaCl and Na_2SO_4 used in the feed formulations were altered during the testing program to establish their influence upon feed processing rates, glass quality, and effluent emission characteristics. These as well as other process variables used during ESCM testing are summarized in Table 2 along with related concurrent processing conditions.

Off-Gas Processing System

Because of the turbulent conditions produced by introducing an aqueous slurry into a melter whose plenum and glass temperatures are greater than 350°C and 1000°C, respectively, a contaminated gas phase waste (off-gas) stream is generated that must be treated before it can be released to the environment. A melter off-gas (MOG) treatment system provides this function by pulling off these hot, contaminated, steam-laden emissions and decontaminating them. As discussed previously,[1] LFCM process exhaust consists primarily of steam, volatilized effluent, and suspended condensed-phase matter (aerosols). Since most waste components that require long-term environmental isolation are lost to the off-gas system as aerosols, LFCM process off-gas (POG) treatment systems are composed primarily of aerosol emission abatement devices and ancillary support equipment.

Like other high-temperature combustion processes, vitrification produces two distinct and independent classes of aerosol effluent: entrained and semivolatile condensed matter.[1] Because many radioactive semivolatiles such as ^{137}Cs form condensation aerosols that are primarily of submicron size, high-efficiency aerosol removal devices are essential components of LFCM processing systems. However, these devices are not, in general, compatible with high off-gas aerosol and steam loadings or with high melter exhaust temperatures; consequently, aqueous quench scrubbers are universally used to precondition the process exhaust by cooling the exhaust stream, condensing the steam, and removing large-diameter (>1 μm) entrained debris. Because entrained matter can also present an off-gas line blockage problem, off-gas quenchers are usually close coupled to the melter source to minimize or at least localize this problem.

In general, additional stages of off-gas treatment would be required if one or more radioactive waste components formed a gaseous effluent that was thermodynamically stable and was little affected by

Table 1. Simulated LFCM Defense Waste Feed

Feed*			Product		
Component	wt%	g/L	Form	Equivalent, g/L	Glass, wt%
$Fe(OH)_3$	12.79	70.1	FeO	15.7	3.25
			Fe_2O_3	34.9	7.22
$Mn(COOH)_2$	3.10	17.0	MnO	11.8	2.44
MnO_2	0.80	4.36			
$Al(OH)_3$	6.02	33.0	Al_2O_3	25.1	5.19
$Ca(COOH)_2$	1.33	7.28			
$CaCO_3$	0.26	1.40	CaO	6.06	1.25
CaF_2	0.058	0.32			
$CaSO_4$	0.16	0.90	$CaSO_4$	0.90	0.19
$Ca_3(PO_4)_2$	0.047	0.26	$Ca_3(PO_4)_2$	0.26	0.054
$Ni(OH)_2$	0.71	3.91	NiO	3.13	0.65
Na(COOH)	2.99	16.4			
Na_2SO_4	0.082	0.45	Na_2SO_4	0.45	0.093
NaCl	0.53	2.90	NaCl	2.9	0.60
NaI	0.006	0.031	NaI	0.031	0.006
Na_2O	7.96	43.6	Na_2O	54.2	11.21
Na_3PO_4	0.006	0.031	Na_3PO_4	0.031	0.006
NaF	0.031	0.17	NaF	0.52	0.11
$NaNO_3$	1.30	7.13			
RuO_2	0.13	0.71	RuO_2	0.71	0.15
$Cr(OH)_3$	0.12	0.67	Cr_2O_3	0.49	0.10
Cu_2O	0.10	0.54	CuO	0.60	0.12
Hg	0.15	0.83			
Zeolite	3.41	18.7			
KCOOH	0.16	0.90	K_2O	0.51	0.10
$Sr(COOH)_2$	0.037	0.204	SrO	0.119	0.025
Rare Earths	0.066	0.36	RE_2O_3	0.36	0.074
$CsNO_3$	0.10	0.56	Cs_2O	0.40	0.083
B_2O_3	6.48	35.5	B_2O_3	35.5	7.34
Li_2O	4.52	24.8	Li_2O	24.8	5.13
ZrO_2	0.65	3.55	ZrO_2	3.55	0.73
SiO_2	45.25	248.0	SiO_2	257.0	53.15
MgO	0.65	3.55	MgO	3.55	0.73

*Feed Density = 1.363; Feed Solids = 548 g/L;
Glass Product = 484 g/L.

Table 2. ESCM Operation and Production Summary

Process Parameter	Units	Test 1	Test 2	Test 3	Test 4	Test 5
Duration	h	236	107	103	104	112
Feed Processed	kg	1198	344	187	280	450
Average Feed Rate	$l/h\text{-}m^2$	70	46	25	36	54
Glass Produced	kg	426	129	66	103	165
Average Glass Rate	$kg/h\text{-}m^2$	35	23	12	18	27
Hg Processed	kg	0.73	0.21	0.11	0.17	0.27
Plenum Temperature	°C	750	750	550	740	740
Air Inleakage	kg/h	0.73	6.8	0.73	0.76	0.76
Feed NaCl	wt%	0.51	0.51	0.51	0.51	0.76
Feed Na_2SO_4	wt%	0.079	0.079	0.079	0.079	0.36
Feed HCOOH	%ST*	90	86	86	110	110

*The amount of formic acid used is specified in terms of the percent of the stoichiometric amount required to complete acid/base reaction with the waste sludge, plus that required to decompose all carbonates and nitrates.

the aqueous quench scrubbing stage. However, radionuclides such as these (^{3}H, ^{14}C, and ^{129}I) are usually not significant HLLW components (on a curie basis). Process generation of regulated gaseous, chemical effluents such as NO_x, SO_2, and Hg may also require additional treatment stages, depending upon the concentrations of these effluents penetrating the primary quench scrubber.

The process off-gas system used in support of the ESCM testing discussed here utilized three stages of off-gas treatment consisting of an ejector venturi (quench) scrubber (EVS), a water-cooled off-gas condenser, and a final high-efficiency particulate air (HEPA) filter, arranged sequentially in that order. The off-gas condenser was employed to reduce off-gas water loadings before final HEPA filtration and to assess the ability of an off-gas chiller to reduce potential off-gas concentrations of mercury vapor. Since the purpose of the ESCM tests was primarily to characterize the melter effluent behavior of mercury, the performance of the generic off-gas processing system was not exhaustively characterized as was the melter source; however, sufficient data were collected to establish average off-gas operating and performance characteristics.

Off-Gas Effluent Sampling System

The sampling network, which was developed in support of melter emission characterization studies, is conceptually illustrated in Figure 3. The components making up this network were designed to selectively determine the composition of melter exhaust with regard to particulate matter (Fe, Si, Sr, etc.), semivolatile effluent (compounds of B, Cs, Ru, Hg, etc.), and noncondensible gases (O_2, CO, CO_2, SO_2, and Hg).

Sampling for mercury effluent in the unquenched (~350°C) melter exhaust was conducted using a differential effluent sampling system to assess the pathways and magnitudes of condensible and/or scrubbable melter-generated emissions (semivolatile and particulate

effluent). This sampling system was composed of a heated (~200°C) particulate filter, a heat-traced sampling line, a condenser, and a series arrangement of three gas scrubbing units sequentially configured in the order listed (Figure 3). The distribution of semivolatiles across the five discrete sample fractions generated by the system was determined using inductively coupled plasma (ICP) spectrometry, atomic absorption (AA), ion chromatography (IC), and x-ray fluorescence (XRF). Chemical identities of crystalline effluents collected were established by x-ray diffraction techniques.

With the exception of mercury, the gaseous composition of melter off-gas emissions was established using continuous real-time gas analyzers. The gas stream sampled was extracted after the off-gas quencher (site S2 of Figure 2) in the two-phase off-gas line leading to the EVS condensate tank. The moisture-laden gas sample stream was passed through a heated filter assembly to remove airborne particulate matter and then through a permeation dryer to reduce the moisture loading of the gas before it was distributed to the individual gas analyzers by a stainless steel bellows pump.

Noncontinuous batch sampling for gaseous mercury was measured at three locations: after the venturi (site S2), downstream of the quench tank (site S3) and, finally, after the exhaust had passed through a condenser and final HEPA filter. The presence of mercury vapor at these off-gas system sites was measured by two separate types of Hg° monitors. One instrument was based upon ultraviolet (UV) excitation, while the other operated on the basis of gold film amalgamation. Being low-level instruments, neither monitor was suitable for direct sampling of the melter exhaust stream. However, by using instrument calibration techniques that involve controlled dilution of a room-temperature $Hg_{(l)}$ source, off-gas system vapor pressure measurements of mercury were obtained using off-gas grab samples.

In addition to effluent data collected by the sampling network, the glass product and the secondary aqueous waste stream generated by the MOG EVS were routinely sampled and analyzed throughout ESCM testing. Post-test examination of the melter and off-gas system was also conducted in order to establish the presence and magnitudes of mercury accumulation sites, if present.

Melter Performance and Effluent Characterization

Melter performance with regard to effluent emission is commonly expressed in terms of a unitless decontamination factor or DF. By definition, a feed component DF is the ratio of the rate at which that particular component enters a device to the rate at which it exits. Consequently, melter DFs are related to LFCM process efficiencies (1 - 1/DF) for converting feed components into a borosilicate glass. Particulate melter DFs are similarly defined in terms of mass ratios without reference to elemental composition, where only contributions of airborne particulate matter to overall melter effluent losses are considered. Since particulate DFs are strongly influenced by physical entrainment losses, they are very good indicators

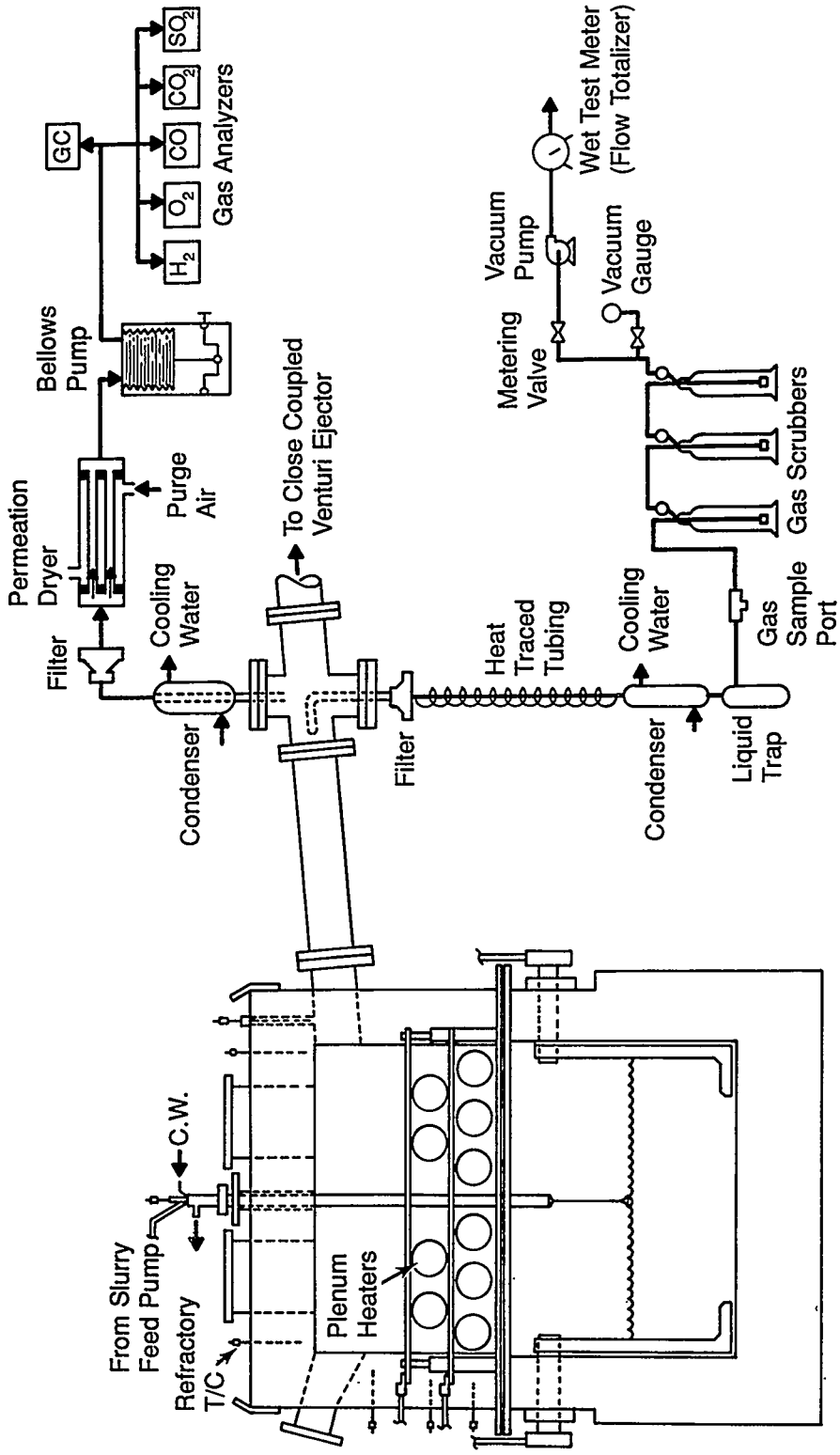


Figure 3. Melter Off-Gas Sampling Network

of processing stability. More importantly, large deviations of individual DFs from corresponding particulate DFs are direct indicators of the semivolatile or volatile properties of the element and/or its chemical compounds.[3]

Table 3 summarizes process and operating conditions used during ESCM testing and the resultant performance parameters obtained. In accordance with pre-testing expectations, processing efficiency for mercury is quite low. No trace of mercury could be found in the glass product under any of the processing conditions used, and analytical sensitivity limits were used with feed makeup data to place an upper bound on melter DFs for this element.

Off-gas sampling results were totally consistent with the low DFs established by the glass composition data; that is, mercury was found to be a major contributor to the overall mass of condensible off-gas effluent exiting the ESCM. This result reflects the fact that a minor feed constituent with very low DF can produce a greater effluent mass loss rate than a major feed component with a significantly higher process DF. Table 4 presents melter effluent loss rates of selected feed components as a percentage of observed mercury losses. These emission rate data clearly show that, on a mass basis, mercury is the major contributor to overall process losses of nonvolatile and condensible effluent. Indeed, the average mass emission rate of mercury observed during all phases of ESCM testing was found to be only 17% lower than the average overall particulate (aerosol) loss rate that has been shown to be the predominant LFCM process loss mechanism.[1]

Although melter DFs for mercury derived from off-gas sampling data were somewhat higher than glass values previously presented (2 vs <1.1), the uncertainties in the documented inhomogeneity of mercury in the feed made the short-term off-gas sampling values indistinguishable from the glass-derived result; i.e., DF ~ 1. Nevertheless, a search for potential accumulation sites for mercury and its compounds within the melter and its off-gas system were conducted upon completion of ESCM testing, as will be discussed later in this paper.

Nature of Melter Mercury Losses

Although it was never expected that the variable process parameters evaluated during ESCM testing would influence melter processing performance for mercury, it was recognized that these variables could be quite influential in establishing the physical and chemical nature of the mercury effluent produced. In order to characterize the pathways and magnitudes of process losses of mercury, a differential sampling system composed of an aerosol filter, condenser, and a series arrangement of three gas-scrubbing units was employed. The manner in which any given element is distributed across the five discrete sampling fractions of this device is indicative of the physical states assumed by the effluent and their relative importance. Sampling to characterize ESCM-generated effluents was conducted in the process exhaust line connecting the melter plenum to the EVS (site S1). The temperature in this jumper was maintained between 300°C and 400°C, using steam injection cooling when necessary.

Table 3. ESCM Processing and Performance Parameters

Test Number	Hours	Prod Rate, kg/hm^2	Plenum Temp, °C	Plenum Composition % O_2	Plenum Composition % CO_2	Plenum Composition % CO	Plenum Composition % H_2O	Feed wt% NaCl	Feed wt% Na_2SO_4	Feed % HCOOH*	Melter DF Hg	Melter DF Fe	Melter DF Part
1	236	35.3	750	1.28	1.05	0.01	92.5	0.51	0.079	90	<1.4	430	820
2	107	23.4	750	9.78	0.47	<0.05	52.3	0.51	0.079	86	<1.4	230	520
3	103	12.3	550	3.26	0.52	0.05	82.8	0.51	0.079	86	<1.4	230	460
4	104	18.3	740	3.33	1.03	0.02	82.9	0.51	0.079	110	<1.1	230	560
5	113	27.0	740	3.11	2.07	0.35	82.7	0.76	0.36	110	<1.1	290	600

*The amount of formic acid used is specified in terms of the percent of the stoichiometric amount required to complete acid/base reaction with the waste sludge, plus that required to decompose all carbonates and nitrates.

Table 4. Comparative LFCM Effluent Loss Rates

	Relative Melter Mass Loss Rates					
Effluent	Test 1	Test 2	Test 3	Test 4	Test 5	Average
Al	0.82	3.13	2.38	2.84	2.33	2.30
B	14.12	10.45	13.06	9.67	11.87	11.84
Cr	0.23	0.60	0.19	0.12	0.23	0.27
Fe	12.99	24.12	24.12	24.34	19.00	20.92
Hg	100.00	100.00	100.00	100.00	100.00	100.00
Mn	3.42	6.65	6.65	6.30	4.79	5.56
Na	9.25	12.02	16.83	6.57	6.94	10.32
Ni	0.06	0.00	0.00	0.01	0.01	0.02
Si	4.21	11.98	10.48	11.98	10.16	9.76
Part.	81.20	128.70	143.37	120.12	112.04	117.09

The sampling results obtained were invariant throughout all phases of ESCM testing. Specifically, all mercury-containing effluent was collected in the condensate fraction of the differential sampling system. Since mercury was never detected in the filter catch, the above sampling results indicate that mercury is primarily a gas phase effluent in the unquenched process exhaust under all the process operating conditions examined. A typical XRF spectrum of insoluble matter collected in the condenser coils of the sampler is displayed in Figure 4. This insoluble matter is almost entirely composed of mercury halides, of which the chloride is clearly the dominant chemical form. The insoluble nature of the halides combined with the relative intensities of mercury and chlorine fluorescent spectral lines establish the major chemical form of the condensible mercury effluent collected as calomel or Hg_2Cl_2. The spectrum also indicates that Hg_2I_2 and/or the mixed halide HgCl·HgI are also present. It should be noted that iodine was a minor halide constituent in the simulated feed stream used during all phases of ESCM testing (see Table 1).

The presence of corrosive sublimate, $HgCl_2$, in the condensate sample fraction was also assessed by examining the concentration of soluble mercury in chloride-rich condensate liquor. The ratio of soluble to insoluble mercury in the condensate sample fractions taken throughout ESCM testing are summarized in Table 5. These data clearly show that insoluble Hg_2Cl_2 is the primary mercury-containing effluent species collected by the off-gas sampling system except during high inleakage and low plenum temperature operating conditions. Since oxidizing conditions are required to produce mercury halides from the Hg° and halide components of the feed, increased oxygen concentrations produced by high melter inleakage conditions should result in conditions favoring production of the higher (mercuric) halide; i.e., $HgCl_2$, in accordance with observations made during ESCM test 2. The effect of plenum temperature upon the relative yields of the two mercury halides is more difficult to interpret, but must necessarily be related to the kinetics of mercury halide formation or, more probably, to effective halide oxidation (X_2 production) rates.

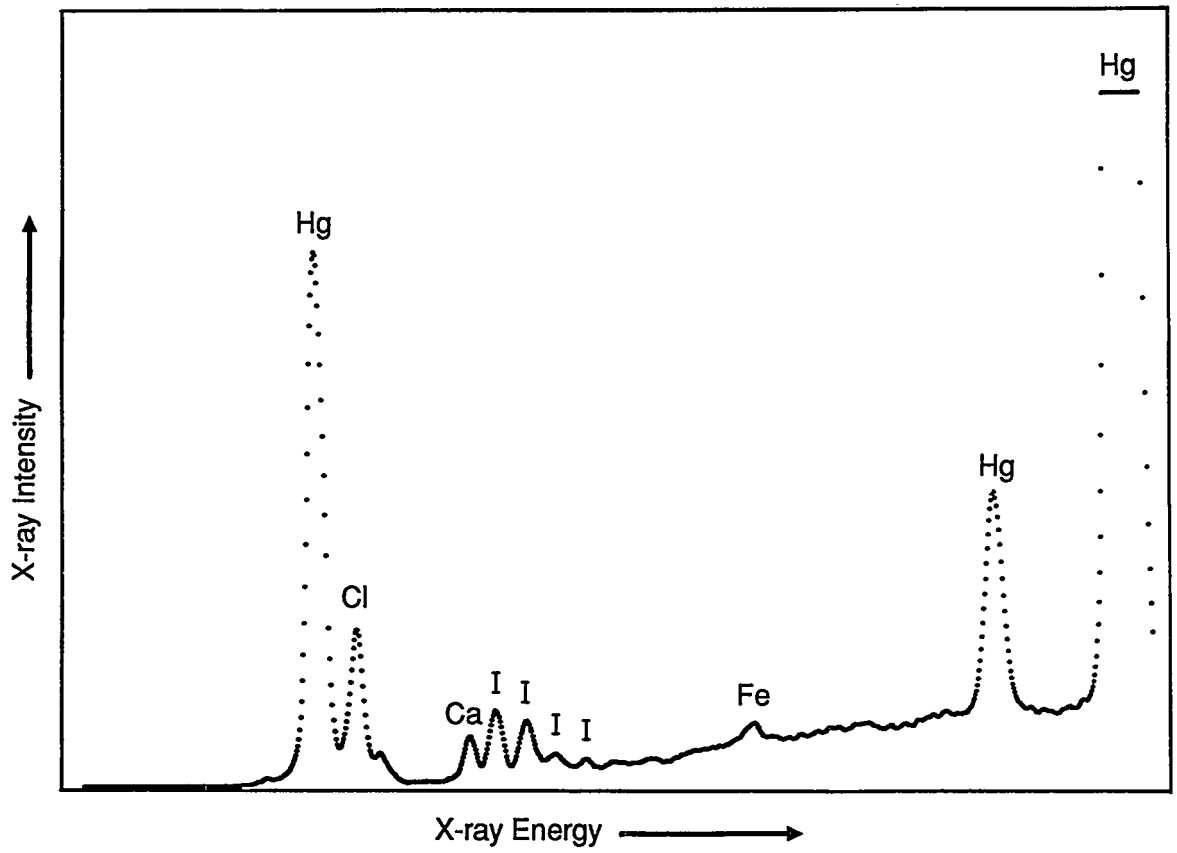


Figure 4. X-Ray Fluorescence Spectrum of Condensed Off-Gas Effluent

Table 5. Sampler Mercury Effluent Composition

Test Number	Air Inleak, kg/h	Plenum, C°	Mercury Halide Composition, % Hg_2Cl_2	$HgCl_2$	Hg_2I_2*
1	0.73	750	86	14	Trace
2	6.8	750	38	62	Trace
3	0.73	550	33	67	Trace
4	0.76	740	82	18	Trace
5	0.76	740	85	15	Trace

*Or mixed halide HgCl-HgI.

Beyond the mercury halides, condensate samples contained little else other than anions formed from aqueous interactions with reactive, acid gas effluents; e.g., X_2, HX, HBO_2, NO_x, and SO_x. No evidence for the presence of HgO was found in any of the samples collected during ESCM testing. Moreover, essentially all mercury effluent collected was chemically combined as a halide. However, the presence of Hg° vapor above all process-generated condensates, detected by environmental mercury analyzers previously described,

unmistakably established the presence of elemental mercury in these samples. However, in no instance was macroscopic, agglomerated mercury (spheres) ever observed in condensate sample fractions or, as will be subsequently discussed, in process quench (EVS) tank residues. It is believed that the presence of the Hg° in these samples was primarily due to the disproportionation reaction, governed by the following equilibrium expression:

$$Hg_2^{++} \rightleftarrows Hg^{++} + Hg;\ K = 2 \cdot 10^{-2} \qquad (1)$$

Because the mercurous ion is only moderately stable to disproportionation in solution, any reagent that preferentially reduces (complexation, precipitation, etc.) the activity of Hg^{++} relative to Hg_2^{++} will drive the above reaction to the right. Indeed the tendency for Hg_2Cl_2 to disproportionate is so great that its name, calomel, is derived from Greek words whose partial meaning is "to blacken." Gradual darkening of condensate sample residues (Hg_2Cl_2) was in fact observed during all ESCM sampling campaigns, further supporting the proposed origin of Hg° in these samples. The very strong vapor signature of this element in all condensate samples collected was a direct result of the extremely large surface area over which the limited (massless) quantities of finely divided mercury were deposited. X-ray diffraction studies of frozen sampler and process condensate residues, which will be discussed in the next section, further confirmed that only microscopic quantities of Hg° were present in these samples.

EVS Scrubbing Liquor Composition

The sampling data just described have shown melter-generated mercury effluent to be in the gaseous state prior to the quenching process. Furthermore, mercury halides were found to be the predominant chemical forms of mercury recovered when the sample stream was quenched. Since these off-gas sampling data cannot conclusively establish that mercury halides were present in the unquenched melter exhaust, it is important to verify that process quenching of the ESCM exhaust will produce the same chemical effluent results that were observed under controlled sampling conditions.

Unlike the differential sampling system that segregated effluents on the basis of their physical states in the unquenched melter exhaust stream, the EVS collects aerosols, condensibles, and chemically scrubbable effluents in a single effluent waste stream--the EVS scrubbing liquid. Because this waste stream is composed of both soluble and insoluble effluents, representative sampling is troublesome, as uniformity of insolubles throughout the bulk of the solution cannot be guaranteed. Nevertheless, samples of the EVS scrub solution effluents were collected and analyzed, assuming uniform stirred tank conditions for ESCM tests 1 through 3. Loss of tank agitation during tests 4 and 5 effectively invalidated the uniform stirred tank approximation and guaranteed nonrepresentative sampling conditions for these last two tests.

Table 6. EVS Mercury Effluent Characteristics

Sample No.	Percent Insoluble		
	Test 1	Test 2	Test 3
1	94	85	67
2	93	89	72
3	89	86	73
4	88	89	66
5	90	94	52
6	87	88	61
7	86	95	31
8	85	96	59
9		88	43
10		93	45
11		87	27
12		90	25
13			32

Table 6 presents the distribution of EVS mercury effluent between soluble and insoluble forms for the first three ESCM tests. With the exception of the second test, these results are quite comparable to the previously described off-gas sampling results if the soluble and insoluble forms of mercury are assumed to be $HgCl_2$ and Hg_2Cl_2, respectively. Since the chloride ion concentration in EVS solutions was always much greater than the mercury content of these solutions, there is little doubt that the weakly dissociated $HgCl_{2(aq)}$ is the major soluble form of mercury present in EVS solutions.

The insoluble forms of mercury present in the EVS scrubbing liquor were assessed using x-ray diffraction techniques. The results of these studies for all ESCM tests conducted are summarized in Table 7. In all cases Hg_2Cl_2 was found to be not only the major mercury compound present but the major (~90%) crystalline compound in the EVS insoluble residues. Moreover, low-temperature x-ray diffraction examinations of the insoluble matter failed to reveal any evidence for the presence of elemental mercury in these samples. The composition of suspended matter in EVS scrubbing liquor, summarized in Table 8, further verifies the conclusions drawn by the x-ray diffraction studies of tank sludges, although, as discussed previously, this suspended matter may not be totally representative of tank insolubles. These compositional results clearly illustrate how a minor process feed stream component with a low melter DF can influence secondary waste stream composition and process recycle strategies.

Given the demonstrated chemical similarities between EVS and off-gas samples, Tables 5 and 6 can be directly compared, keeping in mind that the predominant soluble and insoluble compounds of mercury in the EVS samples are $HgCl_2$ and Hg_2Cl_2, respectively. In general, good agreement between sample results is observed except that the proportion of calomel in the EVS waste stream tends to be higher than in corresponding off-gas sampler effluent. This is especially true

Table 7. X-Ray Diffraction Data for Insoluble Residues Collected During ESCM Tests

ESCM-I Condenser Residue			ESCM-I Venturi Residue			ESCM-II Venturi Residue 8/5/83		
Intensity	Interplanar Distance, °A	ID	Relative Intensity	Interplanar Distance, °A	ID	Relative Intensity	Interplanar Distance, °A	ID
3	5.11	?	82	4.19	HgCl	90	4.17	HgCl
3	4.52	HgI	16	3.36	?	3	3.34	?
80	4.15	HgCl	100	3.195	HgCl	100	3.17	HgCl
7	3.48	HgI	10	2.98	?	2	2.96	?
100	3.17	HgCl	15	2.85	HgCl	20	2.83	HgCl
1	2.96	?	45	2.75	HgCl	50	2.728	HgCl
2	2.90	HgI	18	2.53	?	10	2.523	?
10	2.82	HgCl	50	2.35	Al	2	2.348	?
90	2.72	HgCl	23	2.25	HgCl	25	2.247	HgCl
2	2.46	HgI	65	2.08	HgCl	70	2.070	HgCl
17	2.35	Al	100+	2.03	Al	50	1.963	HgCl
20	2.25	HgI, HgCl	20	1.98	HgCl	2	1.842	?
27	2.07	HgCl	40	1.97	HgCl	7	1.757	HgCl
100	2.03	Al	11	1.76	HgCl	24	1.734	HgCl
25	1.97	HgCl	25	1.735	HgCl	2	1.697	?
	1.96	HgCl	10	1.596	HgCl	10	1.588	HgCl

ESCM-II Venturi Residue 8/12/83			ESCM-III Venturi Residue 10/1/83		
Relative Intensity	Interplanar Distance, °A	ID	Relative Intensity	Interplanar Distance, °A	ID
4	5.11	?	100	4.19	HgCl
75	4.16	HgCl	2	3.70	?
3	3.35	?	100	3.20	HgCl
100	3.17	HgCl	2	2.98	?
20	2.83	HgCl	8	2.85	HgCl
55	2.726	HgCl	20	2.75	HgCl
20	2.526	?	5	2.71	?
28	2.244	HgCl	12	2.53	?
70	2.065	HgCl	20	2.26	HgCl
5	1.99	?	30	2.08	HgCl
55	1.975	HgCl	5	1.99	HgCl
	1.963	HgCl	15	1.97	HgCl
2	1.84	?	2	1.84	?
3	1.757	HgCl	3	1.76	HgCl
20	1.731	HgCl	6	1.74	HgCl
4	1.697	?	2	1.70	?
			4	1.595	HgCl

Table 8. Major and Minor Feed Components in EVS Suspended Solids, wt%

Element	Test 1	Test 2	Test 3	Test 4	Test 5
Al	0.77	0.45	1.06	0.10	0.00
Cr	0.51	1.09	1.34	1.99	0.60
Fe	33.89	29.30	40.10	46.71	32.87
Hg	51.48	59.91	45.47	43.96	60.58
Mn	4.68	5.02	6.42	0.00	0.00
Na	0.51	0.38	0.60	1.85	1.60
Ni	0.00	3.26	3.69	3.87	3.91
Si	8.15	0.60	1.31	1.51	0.43

for ESCM test 2, which involved high inleakage and plenum temperatures. The fact that such a large difference exists between sample results for this test suggests that stable chemical forms of mercury in the unquenched melter exhaust probably did not exist and that the chemical nature of mercury effluent collected may have been a function of quenching conditions. Since the residence time of off-gas effluents was minimized during this test, reaction kinetics may also have been responsible for transitory off-gas compositions and resultant variable effluent results.

Like the mercury effluent collected by the off-gas sampler, the insoluble EVS matter exhibited a very strong vapor pressure of elemental mercury, but no evidence of agglomerated mercury could be found. However, due to the density of elemental mercury, sampling of the scrub solutions may not have been representative for the elemental form of this effluent. Consequently, a search for macroscopic quantities of pooled elemental mercury was conducted within the EVS quench tank. This was accomplished by allowing the scrub solution to settle, pumping out the supernate to within a few inches of the tank bottom, and allowing the remaining water to evaporate. The tank residues were subsequently examined. No sign of the elemental form of this effluent could be found through visual examinations, and low-temperature x-ray diffraction examination of these residues also failed to detect any significant concentrations of Hg° in these tank samples. Nevertheless, like all process samples, these tank residues exhibited an elemental mercury partial pressure that was nominally equivalent to the temperature-dependent vapor pressure of this element. The impact of these process sources of mercury vapor upon off-gas concentrations of this element will be discussed in the following section.

Off-Gas Concentrations of Mercury Vapor

Since mercury is a toxic, controlled effluent, mercury vapor concentrations were measured at various off-gas system locations during ESCM testing in order to assess the impact of gas phase losses upon off-gas process requirements. Mercury vapor concentrations were measured at three different off-gas locations: after the venturi in the two-phase off-gas line leading to the condensate tank (site S2), downstream of the quench tank (site S3), and, finally, after the exhaust had passed through a condenser and final HEPA filter (see Figure 2). These sampling points will be referred to as the venturi, quench tank, and HEPA locations, respectively. Table 9 presents a summary of the mercury data collected at these points during tests 2 and 3, which represented typical off-gas conditions during normal processing operations.

The measurements made after the venturi suggest that a near-saturation condition existed in the quenched exhaust for the duration of both ESCM tests. That is, the partial pressure of mercury measured at sampling site S2 was found to be equivalent to the equilibrium vapor pressure of this element at the temperature of the observation. This result is also found to be independent of the range of off-gas flow rates used during LFCM mercury testing. Nominal saturation conditions were also observed at the other two sampling sites, although the concentrations were significantly lower due to reduced off-gas temperatures. Specifically, the exhaust

Table 9. ESCM Quenched Exhaust Gas Concentrations of Hg

Test	Date	Time	Hg Partial Pressure and Corresponding Vapor Pressure Temperature, Torr x 10^3, °C						Venturi Exhaust, °C
			Venturi		Quench Tank		HEPA		
			Pres	Temp	Pres	Temp	Pres	Temp	
ESCM-2	08/02/83	1000	6.7	41	--	--	--	--	40
	08/03/83	1400	6.5	41	--		0.45	9	40
	08/04/83	1730	4.1	35	1.7	24	0.48	10	40
	08/05/83	1100	5.6	39	1.7	24	0.53	11	41
ESCM-3	08/08/83	2220	3.1	32	0.88	17	0.33	6	32
	08/09/83	1015	2.0	26	1.0	18	0.57	12	30
	08/10/83	0940	2.1	27	1.2	20	0.41	8	30
	08/11/83	1415	1.3	21	0.96	17	0.55	11	30

concentrations of this element were nominally determined by the EVS tank and off-gas condenser exhaust temperatures, ~20°C and 5°C, respectively. However, accumulation of mercury-bearing aerosols on the HEPA filter, which will be described later, definitely influenced measurements collected downstream of this device. The simple temperature dependence of these vapor measurements suggests that an off-gas chiller is apparently all that is required to control off-gas mercury vapor emissions.

As previously mentioned, the EVS waste stream solids were found to exhibit a strong Hg° vapor pressure even though only microscopic quantities of mercury were present. In an attempt to determine the influence of the EVS waste stream upon the overall mercury source term, the Hg° concentration in the venturi exhaust was measured as a function of time after melter feeding was terminated at the conclusion of ESCM test 3. The data collected, which are summarized in Table 10, clearly establish that the Hg-loaded scrub liquor is a major off-gas emission source of Hg° vapor effluent and that EVS operations are capable of maintaining saturation conditions in the dual-phase venturi exhaust line independent of the melter source term.

Table 10. Time Dependence of Hg Vapor Pressure in the Venturi Exhaust Upon Feed Termination (0935 hours)

Time	Hg Partial Pressure, Torr x 10^3	Exhaust, °C	Vapor Pressure Temp, °C
0935	2.2	27	27
0944	2.2	27	27
1004	2.1	27	27
1025	2.0	26	26
1125	1.7	24	24
1315	1.5	22	22
1415	1.8	25	25
1650	1.6	23	23

EVS tank exhaust concentrations of Hg° have also been found to be strongly linked to the turbulence associated with tank agitation. Without agitation, the EVS jet spray provides the only intimate contact between mercury-coated tank residues and the off-gas stream. When the EVS tank agitator failed during tests 4 and 5, tank exhaust concentrations of mercury dramatically dropped below the levels previously achieved with the 5°C off-gas condenser. This result demonstrates that the agitated EVS tank was a significantly greater source of mercury vapor than was the quenched melter exhaust stream. It should be noted that the large effect of tank agitation upon off-gas concentrations of mercury in these tests was somewhat enhanced by the fact that EVS tank inleakage significantly (~2X) diluted the process exhaust stream composition before measurements were made. However, this condition in no way alters the conclusions drawn about the importance of the agitated EVS tank to the overall off-gas concentrations of mercury vapor.

Since the mercury in the melter feed stream was primarily in an uncombined, elemental form, the impact of the agitated feed tank upon mercury vapor concentrations in the vessel ventilation system was also investigated. Unlike the process off-gas system, much greater tank ventilation flow rates were used in characterizing $Hg_{(g)}$ in the vessel vent system. The results obtained from these measurements over a two-day period are summarized in Table 11. As with the EVS tank, the agitated feed tank's exhaust exhibited a partial pressure of mercury vapor that approached the characteristic vapor pressure of this element under low flow conditions. However, with an air purge of 160 l/m (0.4 scfm/ft^2) mercury vaporization rates were insufficient to maintain saturation conditions in the vessel exhaust stream. The average partial pressure of mercury under these high flow rate conditions is equivalent to saturation conditions at 14°C. As shown in Table 11, the observed concentrations are purge-rate-dependent variables, and reductions in tank ventilation rates will be accompanied by corresponding increases in exhaust concentrations of $Hg_{(g)}$.

Process Accumulation Sites of Mercury-Containing Effluent

Physical examinations of the ESCM and its associated off-gas processing system were conducted at the conclusion of the five LFCM processing tests in order to identify unanticipated sites of accumulation of mercury and its associated compounds. Although mercury and

Table 11. Feed Tank Hg Emissions

Sampling Period		Feed Tank Exhaust Conditions		Vapor Pressure	Exhaust Flow,
Date	Time	Hg°, Torr x 10^3	°C	Temperature, °C	l/m(scfh)
10/11/83	1545	0.44	30	10	156 (330)
10/12/83	1650	1.4	31	22	-- low
10/12/83	1730	0.79	31	15	156 (330)
10/12/83	1738	0.86	31	16	156 (330)
10/12/83	2000	0.61	31	13	156 (330)
10/13/83	1600	0.64	28	13	156 (330)

its halogenated compounds have significant vapor pressure at or below LFCM operating temperatures, cool surfaces within the melter cavity and/or melter off-gas system can condense gaseous mercury effluent and act as a sink or accumulation point for mercury and its volatile compounds. Since the condensation of volatiles can and will produce small, highly penetrating aerosols, the final off-gas HEPA filter can also become a significant accumulation site for mercury-bearing compounds. Consequently, the melter, the melter EVS off-gas line, the quench tank, the quench tank exhaust line, and the final HEPA filter were all examined to establish potential buildup sites of mercury and to measure the magnitudes of mercury accumulations, if possible.

Melter

In order to prepare the ESCM melter for visual examination at the conclusion of the fifth and final test, the remaining glass in the melter had to be gravity drained through an inductively heated freeze valve located in the bottom of the melt chamber. To accomplish this, plenum heaters were used to heat the melter cavity to ~1000°C after ancillary equipment (such as the feed nozzle) was removed. A water-cooled condenser was placed in the melter exhaust port to collect any mercury volatilized from the melter during the heatup and/or draining process, and an environmental mercury monitor was used to measure mercury vapor emanating from the molten glass as it was discharged from the melter. The melter itself was disassembled and destructively examined for mercury content subsequent to the glass draining process.

Table 12 summarizes the location and quantity of mercury found in the melter. The largest source of mercury found in the melter examination was observed in the melter exhaust condenser and represents the quantity of mercury that resided within the melter after processing that distilled away during the draining process. As with literally all samples collected during ESCM testing, the volatilized mercury collected was primarily chemically combined, presumably as a halogen. However, the sample exhibited a strong elemental mercury vapor pressure, but no evidence for macroscopic quantities of agglomerated mercury could be found in the condensed effluent. Nozzle and viewport deposits collected after processing but before melter heatup and glass drainage accounted for nominally 5% of the sample mass of mercury collected from the melter.

No elemental mercury vapor could be detected emanating from the molten glass draining from the melter. This was also found to be the case for glass transfers conducted during all phases of ESCM processing of mercury-containing feeds. This result is in accord with previously described compositional data that found no detectable quantities of this element in the vitreous glass product.

Very little evidence of mercury was found within the melter itself after glass drainage. Only nozzle and off-gas ports in the melter lid exhibited any detectable elemental mercury vapor pressure and this emanated from massless deposits. All cracks and penetrations in the melter refractories that extended out to the melter

Table 12. Mercury Content of Post-Test Samples

Source	Sample Number	Sample wt, g	Location	Composition	Hg, g	% Total
Melter	1	0.213	Lower Feed Nozzle	50% HgCl, 50% Fe_3O_4	0.0064	4×10^{-3}
	2	1.609	Upper Feed Nozzle	50% HgCl, 50% Fe_3O_4	0.0193	1.1×10^{-2}
	3	1.232	Feed Nozzle Port	SiO_2, Weak HgCl and Fe_3O_4	0.0296	1.7×10^{-2}
	5	39.04	Viewport	--	0.109	6.3×10^{-2}
	9	--	Thermal Well Top 2"	HgCl	--	--
	10	--	Thermal Well Top 6"	Fe_3O_4 and NaCl	--	--
	11	--	Thermal Well Port	HgCl, Moderate Fe_3O_4	--	--
	19-21	--	Exhaust Quencher	--	3.68	2.1
Quench Tank	4	10.8	Bottom Drain Residue	--	0.824	0.4
	30	--	Tank Bottom Residue	--	141.01	81.2
HEPA	31	--	Filter Sheets	--	28.00	16.2
			TOTALS		173	100

shell were closely examined for deposits of condensed mercury. No evidence of mercury accumulations could be found and no characteristic vapor emanations could be detected from any site other than the nozzle locations previously described.

Since the total quantity of mercury residing in the melter after processing is clearly an insignificant fraction of quantity fed to it during the three processing campaigns (1.5 kg), it is clear that an LFCM is not a significant point of accumulation for mercury or its associated compounds. Moreover, the melter effluent characterization

and post-test examination studies have shown that feed component mercury does not present a credible electrical shortage threat to LFCM operations.

Melter EVS Off-Gas Jumper

The process exhaust line conveying melter off-gases to the EVS quench scrubber was operated at ~350°C throughout all phases of ESCM testing. Consequently, condensation of mercury compounds was not a problem over most of its length. However, the temperature of this line at the inlet of the EVS quench scrubber was sufficiently low to condense volatile mercury halides which, once formed, acted as accumulation sites for other airborne condensed effluent. Off-gas line obstructions instigated by mercury effluent condensation were observed during all phases of ESCM testing and are a reflection of the fact that a temperature transition between 350°C and 40°C has to occur at the off-gas inlet of the process quench scrubber. Because of this and the insoluble nature of calomel, mercury in LFCM feeds will exacerbate off-gas blockage problems associated with the LFCM technology.

Use of high off-gas velocities has been somewhat successful in reducing off-gas effluent deposition rates with non-mercury-containing feeds. Air or steam injection used to boost melter exhaust velocities may, along with resultant volumetric dilution, also be useful in controlling mercury deposition rates, but unless the observed process chemistry changes, mercury-containing feeds will create additional unquenched off-gas system challenges.

Condensate Tank

An ejector venturi was used to scrub and quench the melter exhaust during the LFCM testing with mercury-containing feed. Analysis of melter exhaust and EVS scrub solution compositions has shown calomel, Hg_2Cl_2, to be a major melter effluent and a primary EVS condensate tank insoluble, respectively. If a nominal EVS DF of 10 for ostensibly insoluble iron effluent is assumed,[1-2] the melter DF data for mercury and iron, when combined with the x-ray diffraction results of condensate tank sludges, suggest an EVS scrubbing efficiency of >90% for mercury effluent. Post-test examination of the quench tank contents confirmed this vessel to be a major accumulation point for melter-generated mercury effluent (see Table 12). The quantity of mercury-rich sludge found in this vessel was not at all unusual in that the tank's contents were pumped out without the benefit of tank agitation; as a result, the settled solids quantified during post-test examination represented a large portion of the insoluble solids collected during the final ESCM test. These insoluble tank residues were found to be friable and easily suspended, and no evidence of macroscopic quantities of high-density elemental mercury could be found in these settled solids.

Quench Tank Exhaust Line

The exhaust line leading from the quench tank to upstream processing equipment was examined for mercury-containing residues. This process line was extremely clean; no collectible macroscopic residues

were present. This finding is not at all surprising in that the condensation process, which has been shown to be almost entirely responsible for generating mercury-bearing airborne particulate matter, produces submicron aerosols that have no appreciable tendency to settle or accumulate on surfaces.

Final Off-Gas HEPA Filter

Since vapor condensation can act to produce penetrating submicron aerosols, the final HEPA filter used in the ESCM off-gas processing system was examined to determine the total mass of mercury that successfully penetrated the EVS quench scrubber as a submicron aerosol throughout all phases of ESCM testing. Samples of the filter media were extracted across the HEPA and were subsequently analyzed for mercury. The average value from these samples was used to estimate the total mercury loading on the HEPA filter. These results are also listed in Table 12. Since ~1.5 kg of mercury was fed to the melter during the five tests conducted, submicron aerosol transport of mercury is not a major source of off-gas system losses of mercury and its associated compounds, although it is significantly greater than elemental vapor losses. Based upon the observed filter loading, feed composition data, and off-gas vapor pressure measurements, the average mercury DF achieved by the EVS throughout all phases of ESCM testing is estimated to be ~40. This corresponds to an EVS removal efficiency of 97%, which is in accord with previous estimates based upon condensate sample compositions.

Discussion/Summary

The processing and off-gas emission characteristics of mercury in LFCM feeds have been studied under various operating conditions, including melter inleakage, plenum temperature, and feed composition. The results of these studies have shown typical LFCM-produced borosilicate glasses to have no detectable capacity for mercury. Essentially all the mercury fed to the melter was lost to the process off-gas system in the gaseous state; as a result, mercury, on the average, accounted for greater than 85% of condensible process off-gas system effluent losses. Mercury halides are the primary chemical forms of mercury condensed and collected after process exhaust quenching. Since chlorine was the major halogen in the melter feed, mercury effluent was predominantly composed of its two chloride forms.

Although $HgCl_2$ is the thermodynamically favored chloride over the entire processing temperature range,[4] Hg_2Cl_2 was found to be the predominant halide condensed and collected after off-gas quenching except under low plenum temperature conditions. Since Hg(II) is readily reduced to the Hg(I) state by elemental mercury if stoichiometric quantities of oxidants are not available, the instability of mercury oxides in combination with limited concentrations of halogens is most likely responsible for the thermodynamically unexpected result. In any event, the fact is that Hg_2Cl_2, on the average, was the predominant chemical form of mercury that was condensed and collected by the process quench scrubber during ESCM testing. This result was also observed previously when a similar waste stream was solidified using the related spray calciner in-can melter technology.[5] Unlike the present result, however, small quantities of

agglomerated elemental mercury were identified in quench scrubber insoluble residues. Only finely divided surface deposits of mercury were observed in process samples collected and examined in these LFCM tests. The highly dispersed nature of these deposits strongly suggests that the source of Hg° in these samples is due to disproportionation of Hg(I) (see Equation 1).

In spite of the fact that large sources of elemental mercury are not generated by the processing of mercury-containing feed, the quenched melter exhaust stream was found to be saturated with mercury vapor under normal operating conditions. The agitated quench (condensation) tank was found to be the major source of mercury vapor downstream of the EVS. However, it was also found that the partial pressure of mercury in the off-gas stream could be effectively and predictably controlled through use of an off-gas chiller.

The major point of accumulation of LFCM processed mercury is the process condensate tank. Overall, the EVS collected 97% of the mercury processed (DF ≈ 40) during ESCM testing, and on the average most of this was in the form of insoluble Hg_2Cl_2. Since the LFCM process has no capacity for incorporating mercury (melter DF ≈ 1), Hg_2Cl_2 always represented the largest fraction of insoluble matter in ESCM condensate tank residues. Typically, 90% of the insoluble crystalline matter collected in the EVS condensate tank during ESCM testing was specifically due to Hg_2Cl_2.

The proportion of soluble $HgCl_2$ produced varied with processing conditions. With low plenum temperatures, ~70% of the ESCM mercury effluent that was collected in the EVS condensate tank and in off-gas samples was in the form of $HgCl_2$. Although off-gas sampling data indicated similar effluent compositions under high inleakage conditions, condensate tank compositional data indicated otherwise. These differing results suggest that quenching conditions may influence the chemical composition of the mercury halides collected. However, the process realities are that except for the test conducted without auxiliary lid heat, Hg_2Cl_2 was the primary chemical form of mercury collected by the process quench scrubber, independent of all other operating conditions.

Acknowledgments

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References

1. Goles, R. W., and Sevigny, G. J. 1982. "Off-Gas Characterization of Liquid-Fed, Joule-Heated Ceramic Melters." Proceedings of the 17th DOE Air Cleaning Conference, August 2-5, 1982, CONF-820833, Denver, Colorado.

2. Scott, P. A., Goles, R. W., and Peters, R. D. 1985. Technology of Off-Gas Treatment for Liquid-Fed Ceramic Melters. PNL-5446, Pacific Northwest Laboratory, Richland, Washington.

3. Goles, R. W., and Andersen, C. M. 1986. "LFCM Emission and Off-Gas Performance for Feed Component Cesium." PNL-SA-13954, Pacific Northwest Laboratory, Richland, Washington.

4. Wicks, C. E., and Block, F. E. 1963. Thermodynamic Properties of 65 Elements--Their Oxides, Halides, Carbides and Nitrides. Bulletin 605, Bureau of Mines, Washington, D.C.

5. Holton, L. K. 1981. Behavior of Mercury and Iodine During Vitrification of Simulated Alkaline PUREX Waste. PNL-3964, Pacific Northwest Laboratory, Richland, Washington.

BEHAVIOR AND CONTROL OF RUTHENIUM DURING OPERATION OF THE NEW WASTE CALCINING FACILITY AT THE IDAHO CHEMICAL PROCESSING PLANT

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Abstract

The New Waste Calcining Facility (NWCF) processes acidic fluoride and nitrate high level liquid wastes, converting them to granular solids in a fluidized bed by internal combustion of kerosene. The off gases pass through a venturi scrubber, silica gel bed, and HEPA filters. Normally, less than 4 μCi ^{106}Ru/day are released in the form of particulate material to the stack. Some operations can cause small, but noticeable increases in ^{106}Ru releases. Although resulting lung doses are extremely small, because of the public perceptions and ALARA principles controls have been implemented to prevent any releases significantly above normal.

Evaluations were made of ruthenium behavior during the various operations in the NWCF and parameters that affect its volatility. The results of the evaluations and recommended controls to prevent ruthenium releases, which are described in detail, are as follows. Dissolution of calciner bed in boiling 13 M HNO_3 with the silica gel beds bypassed resulted in 0.17 Ci release (0.004% of inventory). With the silica gel beds on line, this operation does not result in increased Ru release; to prevent challenging the beds, dissolution in 6 M HNO_3 at 90°C is recommended. Recommended conditions for decontaminating lines downstream of the silica gel beds are initial contact with 4 M HNO_3, followed by 6 M HNO_3 at 80°C or up to 10 M HNO_3 at 60°C and, for leaching spent HEPA filters, 6 M HNO_3 at 80°C. Silica gel beds may be bypassed if necessary for washing off collected particles during normal calcination operations. Small ^{106}Ru releases of up to 0.19 μCi have sometimes been observed during this condition, but may be due to the bed washing and drying operation rather than from calcination off gas.

I. Introduction

The New Waste Calcining Facility (NWCF) processes acidic fluoride and nitrate high level liquid wastes, converting them to a granular solid in a fluidized bed heated to 500°C by internal combustion of kerosene. The off gases pass through a venturi scrubber, silica gel beds, and HEPA filters. Various operations have the potential for volatilizing small quantities of ^{106}Ru. Most of any volatilized ruthenium in the calciner system is reduced to particulate RuO_2, due to reducing conditions present in the off-gas lines, and removed in the off-gas cleaning system. However, any potential for ^{106}Ru releases to the stack greater than the normally very small release of a few microcuries ^{106}Ru/day is viewed as undesirable because of negative public perceptions.

A small release of 0.17 Ci ^{106}Ru during nitric acid dissolution of a calcine bed in October 1988 prompted a review of operations with the potential for similar increased releases and establishment of

procedures and controls to prevent such releases. Evaluations were made of ruthenium behavior during the various operations in the NWCF and parameters that affect its volatility. The operations include bypassing and washing the silica gel beds during calcination, bed dissolution, decontaminating off-gas lines downstream and upstream of the silica gel bed adsorbers, decontaminating the cell floor, and leaching of spent HEPA filters in preparation for disposal. Operating, laboratory, and literature data were used to establish practical conditions that will assure control of ruthenium.

II. Calciner System and Operations

The calciner off-gas system is depicted in Figure 1. When fluoride-nitrate wastes are calcined, $Ca(NO_3)_2$ is added to the liquid feed and reacts with the fluorides to form stable, non-volatile CaF_2. Thus, the primary off gas constituents are fluidizing air, combustion products of kerosene, water vapor, oxides of nitrogen, and calcine fines. A high-efficiency cyclone removes a majority of fine particles from the 500°C off gas, which then enters the wet scrubber system consisting of a quench tower, venturi scrubber, venturi scrubber knockout drum, condenser, mist eliminator, and scrub hold tank. The quench tower cools the off gas to about 70°C. The scrub solution is typically 2 to 4 M HNO_3.

The scrubber off gas passes through the adsorber superheater and three parallel silica gel ruthenium adsorber beds. The adsorbers each contain a lower and an upper bed of silica gel. The lower bed acts primarily as a prefilter to remove residual entrained particles and hydrocarbons. The dry upper bed will remove most of any remaining ruthenium vapor species. Two parallel banks of three series HEPA filters each are on line during normal processing; one bank is on line during shutdown periods.

The NWCF process off gas is combined with other plant gases and is finally treated with the Atmospheric Protection System (APS) HEPA filters. Only one of the two stages of HEPA filters shown in Figure 1 has been used.

The stack gases are sampled at the 90 foot level. The sampling system consists of a particulate filter and three gas monitoring beds in series: charcoal, silver zeolite, and silica gel. The particulate filter is pulled daily at 8:00 a.m. for counting. It has a detection limit for ^{106}Ru of 0.63 μCi for the 24 hour sample. The gas sample adsorber beds are pulled biweekly and have a detection limit of 0.41 μCi ^{106}Ru, equivalent to 0.029 μCi per 24 hours average. No ^{106}Ru gas species has ever been detected since operation of the NWCF began in September 1982.

The normal concentration of ruthenium in the high level liquid waste is approximately 10^{-3} M, or less. ^{106}Ru comprises 0.1 to 0.2 percent of the total ruthenium. A steady state calcine bed weighs ca. 2300 kg and contains on the order of a kilogram of ruthenium and 5000 Ci ^{106}Ru.

Laboratory studies[1] have shown that during calcination, 20 to 62 percent of the ruthenium is volatilized, the higher value occurring during smooth burning of the kerosene, as is characteristic of the

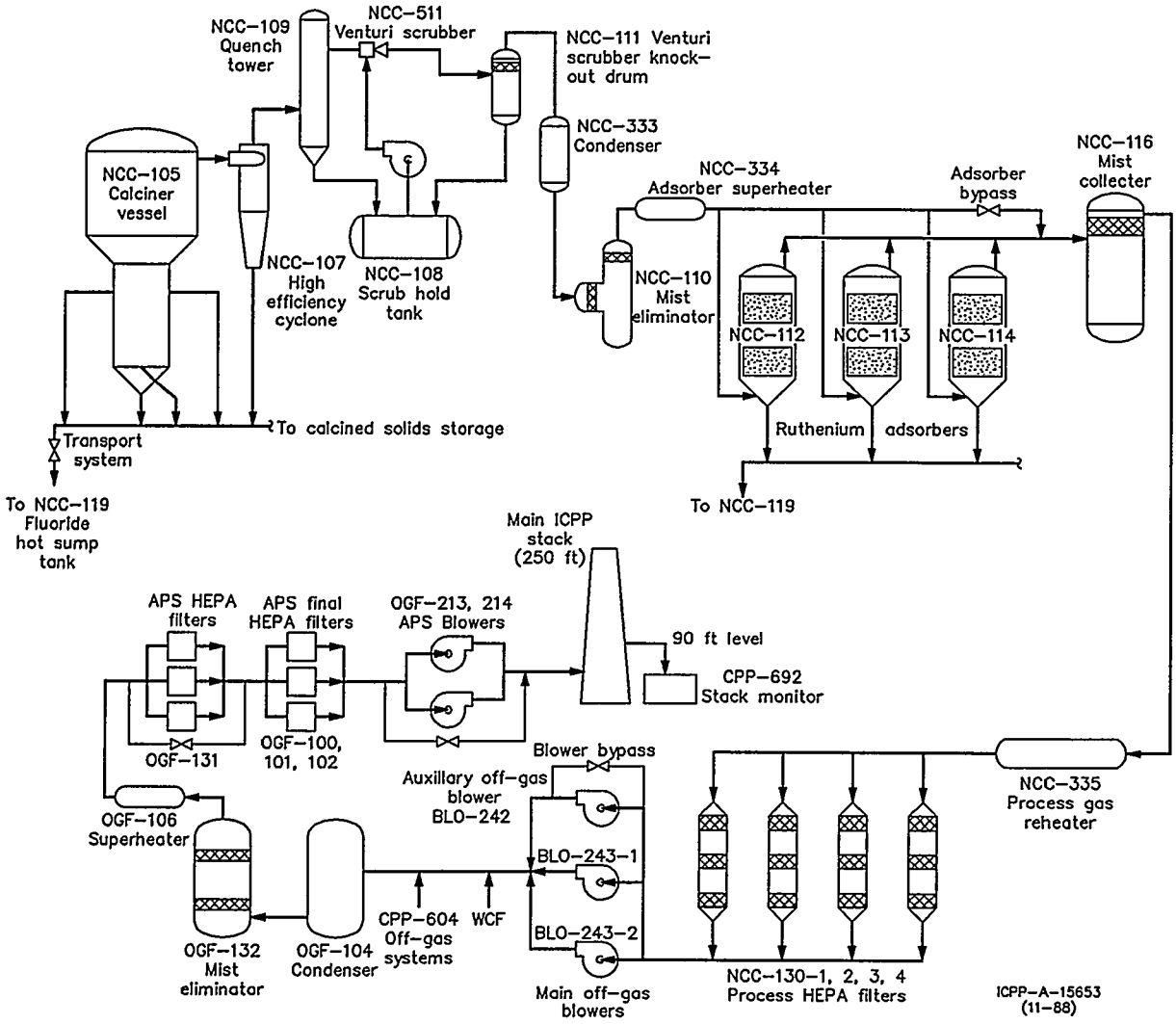

Figure 1 Calciner Process Off-Gas System.

full scale calciner. The volatile form is most likely a nitrato or oxyfluoride species. Generally, gaseous RuO_4 cannot account for the partial pressure of ruthenium observed in calcination processes.[1] The partial pressure and, thus, volatility, decreases with increasing temperature, in contrast to the formation of $RuO_4(g)$ over RuO_2. In the reducing conditions of the off gas from the combustion of kerosene, the vapor species in the calciner is quickly reduced to RuO_2, resulting in efficient removal in the scrubber system and downstream particle removal devices. Thus, the silica gel beds are not normally challenged with gaseous ruthenium species. Certainly, less than 1 percent of the feed ruthenium (maximum feed rate approximately 17 g Ru/hr) reaches the adsorber beds as a vapor. The adsorber capacity of the three silica gel beds exceeds 40 kg ruthenium.

Normal decontamination factors for the off-gas cleaning devices for particulate and gaseous ruthenium species have been estimated from operating data from the original Waste Calcining Facility and are reported elsewhere.[2,3]

III. Release Limits and Dose Consequences

A Technical Standard (TS) safety limit for stack releases of particulate radionuclides has been set to limit off-site exposures to less 7.5 mrem for 50-year committed dose equivalent (CDE) to any organ and 2.5 mrem effective dose equivalent (EDE). The one-hour limit is 0.25 Ci. When releases are uniform over 24 hours, the 24 hour limit is 6.6 Ci.* The limits are based on a standard average radionuclide composition of stack particles, of which ^{106}Ru constitutes 17 percent of the activity. The limiting radionuclides and dose are $^{238,239,240}Pu$ and the associated bone CDE; for 6.6 Ci release of the standard composition, the bone CDE is 7.3 mrem and the EDE is 1.2 mrem. ^{106}Ru provides a lung CDE.

Normal stack total particulate releases are a few millicuries/day, while the typical ^{106}Ru particulate release is a few (less than 5) microcuries/day. Thus, less than 0.5 percent of the characteristic particle activity is due to ^{106}Ru.

For Class F fumigation conditions (worst case), the atmospheric dispersion factor, χ/Q, is 1.9×10^{-6} sec/m^3. Class F fumigation conditions do not last more than one hour and do not occur more than once each 24 hours. For purposes of setting the TS 24-hour release limit, a weighted average χ/Q of 7.4×10^{-8} sec/m^3 is assumed. Based on these values, the maximum off-site dose per curie of particulate ^{106}Ru release during the meteorological conditions would be as summarized in Table 1.

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* The 24-hour limit, on an hourly basis, exceeds the one-hour limit, because the 24-hour worst case average meteorological conditions result in more dilution than the one-hour worst case condition (see text below). For consistency, however, the 24-hour limit is being evaluated for revision to be as restrictive as the one-hour limit.

Table 1 Maximum off-site 50-year committed dose equivalent per curie particulate ^{106}Ru release.

Meteorology	Lung CDE, mrem/Ci ^{106}Ru
Class F fumigation (1-hour maximum occurrence)	2.4
Maximum weighted average for 24 hour period (conservative for Technical Standard Controls)	0.092
Typical 24 hour average (assumed to be 1/3 of maximum weighted average)	0.031

For the TS limit release of 0.25 Ci particles over one hour with 17 percent of "standard" activity due to ^{106}Ru, the calculated 50-year lung CDE to the maximally exposed offsite individual under Class F fumigation meteorological conditions is 0.10 mrem. The 6.6 Ci total particulate, 1.1 Ci particulate ^{106}Ru 24-hour uniform release during the maximum weighted average meteorological conditions would, likewise, result in 0.10 mrem. The effective dose equivalent is 0.30 times the lung CDE.

The Technical Standard limit is just that: a limit that is not expected to be challenged or approached. The operating philosophy is to control releases so that they are a small fraction of the conservatively established safe limit.

IV. Behavior and Control of Ruthenium During Operation

A. Normal Calcination

Normal stack releases of ^{106}Ru during operation of the NWCF have varied from nondetectable (< 0.63 μCi/day) to ca. 1 mCi/mo (30 μCi/day average) as particles (see Figure 2). Typical recent normal releases, since January 1985, have been less than 100 μCi/mo (3.3 μCi/day average). The average 50-year committed dose equivalent to the lung of the maximally exposed offsite individual from a 3.3 μCi/day release is 1×10^{-7} mrem/day. Clearly, these background releases are acceptably small. They provide the basis for comparing potential releases from special operations.

B. Washing of Silica Gel Beds

The silica gel beds must be washed periodically to remove collected particles that increase the pressure drop. They are designed so that a bed can be valved out for washing while the other two remain on line as the calciner operates. However, the isolation valves have begun to leak, resulting in water from the bed being washed entering the off-gas stream via the other beds, with the potential for saturating the gas and wetting the downstream HEPA filters. Therefore, it has been necessary during the later operations to completely bypass the beds for washing as the calciner operates.

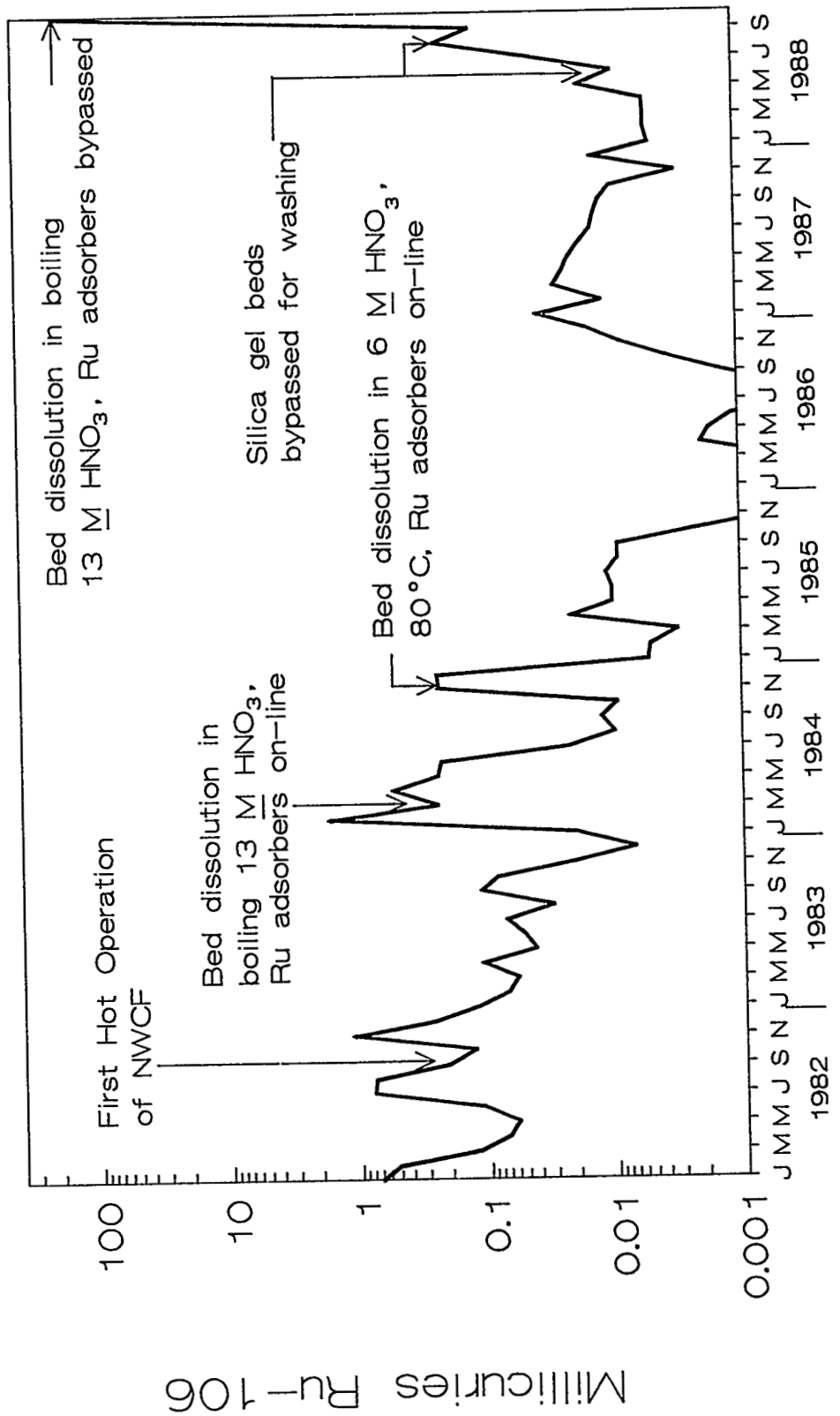


Figure 2 Monthly Ru-106 stack releases.

Until it is possible to replace the valves, an assessment of ^{106}Ru release that can occur while the calciner is operating was required so that a determination could be made of an acceptable operating procedure to permit the silica gel ruthenium adsorber beds to be isolated from the off-gas system to be washed. The results of the assessment are summarized here.

In 1988, the silica gel beds were bypassed while the calciner was processing high level waste on three different occasions. The daily ^{106}Ru releases during these periods, which were only in the form of particles, are shown in Figure 3. The first time that the beds were bypassed and washed, on June 5-7, stack releases did not increase over background at the time of 1 to 3 μCi/day. On the second occasion, August 2-3, a noticeable increase to 0.19 mCi occurred. On August 20-21, a steam outage to the adsorber beds caused moisture to condense in them and, so, the bypass valve was opened (see Figure 3). The beds did remain on-line for almost two days, but very little gas passed through them. A small increase in daily ^{106}Ru particulate release occurred, to 60 and 19 μCi, but when the beds were valved out and washed, the stack release was normal.

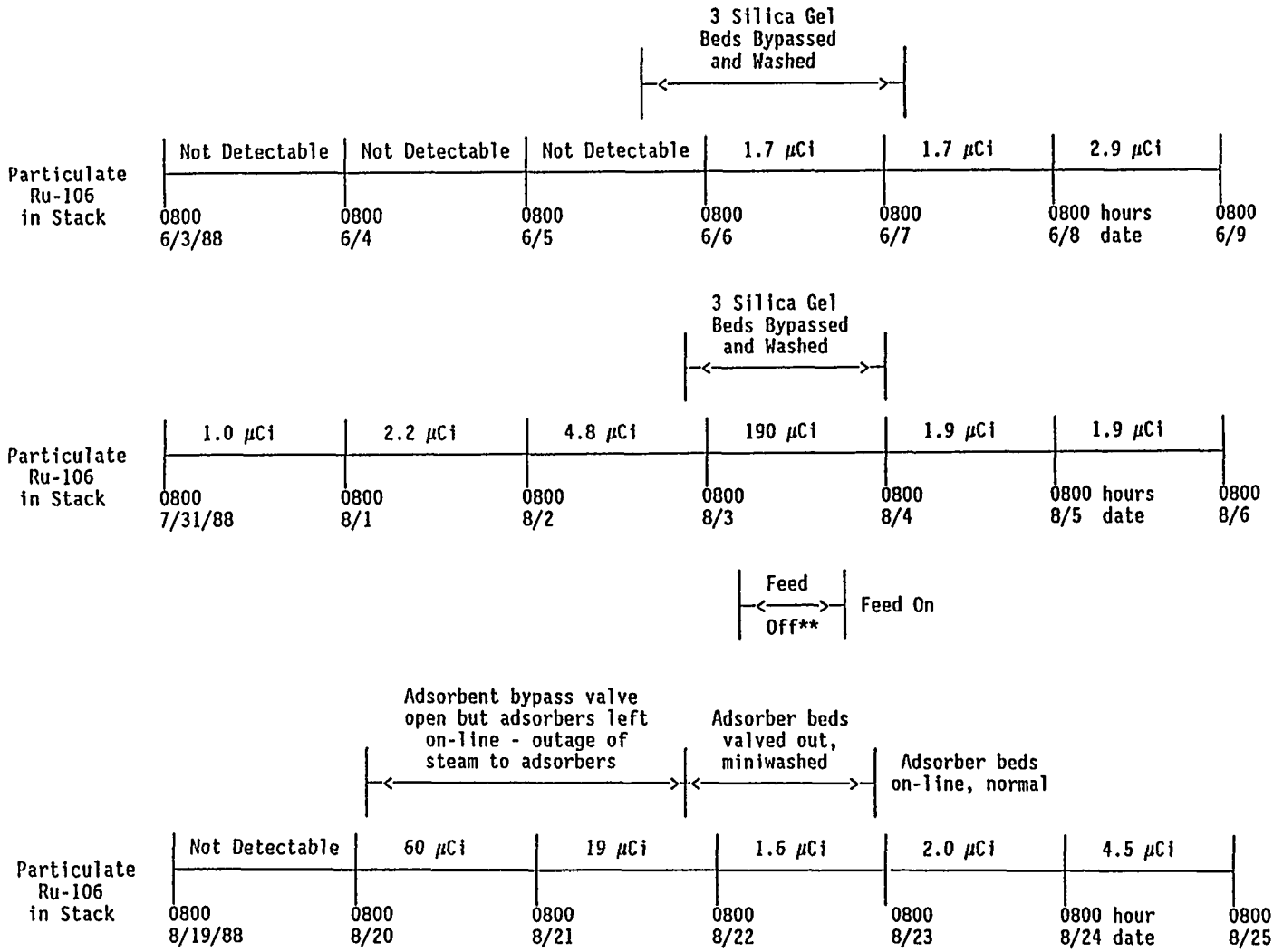
Indications are that the 0.19 Ci 24-hour release during the August 3 bed washing is associated with that activity and probably is indicative of the magnitude of the upper limit expected. The release, while real, is very small and is 0.0029 percent of the Technical Standard safety limit of 6.6 Ci for all particulate radionuclides over 24 hours and only 0.08 percent of the one-hour safety limit particulate release of 0.25 Ci.

A 0.19 mCi ^{106}Ru release over 24 hours that resulted from bypassing the silica gel beds and washing them would result in a maximum off-site lung CDE of approximately 0.000017 mrem. If it should occur within a 1 hour period during which the worst-case Class F fumigation meteorological condition existed, the CDE would be 0.00046 mrem (see Table 1). Clearly, these are negligible compared with the Technical Standard requirements and intent.

These doses may be compared with the approximately 0.00037 mrem/day (bone) limiting CDE resulting from normal 1 mCi/day stack particulate releases of a "standard" composition. On an annual basis, the silica gel beds might be washed six times. If increased ^{106}Ru releases were to occur each time (they haven't in the past), over a 24 hour period, the annual maximum CDE would be unaffected. If they were to occur over one-hour periods during Class F fumigation conditions, the annual average CDE would be increased 0.4 percent; this is extremely unlikely.

An accidental occurrence could be loss of the venturi scrubber pumps during calcination while the silica gel beds were bypassed. This would activate the Rapid Shutdown System (RSS). Conservative estimates of the potential ruthenium-106 release during such an excursion range from 0.0002 to 0.0076 Ci, the former being the more probable (see Appendix). These are well within, by factors of 33 to 1300, of the TS one-hour release limit for total particulates and by factors of 5.6 to 220 of the implied associated release limit for particulate ^{106}Ru (based on ^{106}Ru comprising 17 percent of the normal particle activity).

Figure 3 ^{106}Ru releases during normal fluidized-bed calcination - NWCF.*



* Detection limit: 0.63 μCi on 24-hour sample.
** Feed was shut off by Rapid Shutdown System when off-gas blower stopped.

No Ru-106 has ever been detected on the gas sample adsorber beds (samples pulled biweekly with detection limit of 0.41 μCi, equivalent to 0.029 μCi per 24 hours).

Maximum off-site doses from the release are indicated in Table 2 (using the dose per Ci factors in Table 1).

Table 2 Estimated doses from Ru-106 potentially released during loss of venturi pump with activation of RSS while silica gel beds are bypassed.[a]

Assumed Meteorological Condition	Lung CDE, mrem	Lung CDE as Fraction of TS Limiting Dose	Lung CDE as Fraction of Daily Average CDE
Typical Average	6×10^{-6} to 2×10^{-4}	8×10^{-7} to 3×10^{-5}	0.02 to 0.6
Maximum Weighted Average Used for TS	2×10^{-5} to 7×10^{-4}	2×10^{-6} to 9×10^{-5}	0.05 to 2
Class F Fumigation	5×10^{-4} to 0.018	6×10^{-5} to 0.002	1.3 to 50

[a] 0.0002 to 0.0076 Ci ^{106}Ru particulate release.

Potential doses under any condition are negligible relative to the TS control intent. Only if an accident occurred during Class F fumigation conditions would the CDE significantly exceed the current average daily doses from normal particulate releases. Class F fumigation conditions exist for less than 1 hour per 24 hour period and the release during the RSS sequence would be over a period of less than 2.5 minutes (0.042 hour, see Appendix). The probability of both occurring simultaneously is 1/24 = 0.042. In addition, the probability of RSS occurring while the beds are bypassed is some small fraction, probably smaller than 1/100, and the chance of a dose from an accidental release of ruthenium exceeding the current average daily dose from particulate releases is less than one in one-to-ten thousand.

An additional accident could be loss of the Rapid Shutdown system at the same time that the scrub pump failed while the silica gel beds were bypassed. The potential ^{106}Ru release can be estimated with the following assumptions. While the condition exists, ruthenium will be continuously released at a rate of 0.0002 to 0.0076 Ci per 2.5 minutes (calculated for the 2.5 minute period required for the RSS to shut off feed). The residence time to the stack monitor is approximately 5 to 10 minutes. Consider that, if the silica gel beds are bypassed, a requirement is in place that calciner feed be shut off immediately if the stack monitor alarms. Assume response time of 5 minutes.

The stack monitor is set to give an "early" high alarm when particles collected on the sample filter (which is changed daily) corresponds to a stack release of 0.0063 Ci. After this quantity has been released from the stack, an additional 5 minutes of releases will occur before the feed is shutoff. At that time the off-gas lines between the calciner and the stack contain another 5 to 10 minutes quantity of ruthenium that will be purged to the stack. The

total amount of ^{106}Ru release would be 0.0063 + [(10 to 15)/2.5] x (0.0002 to 0.0076) Ci, or about 0.0071 to 0.052 Ci. This is less than the quantity of 0.063 Ci that would set off the "high" high alarm (which is set after an "early" high alarm and which is the normal level requiring process shutdown) and 2.8 to 21 percent of the TS safety limit for particulate release.

Maximum potential offsite doses are summarized in Table 3.

Table 3 Estimated doses from Ru-106 potentially released during loss of venturi pump and RSS while silica gel beds are bypassed.[a]

Assumed Meteorological Condition	Lung CDE, mrem	Lung CDE as Fraction of TS Limiting Dose	Lung CDE as Fraction of Daily Average CDE
Typical Average	$2x10^{-4}$ to 0.002	$3x10^{-5}$ to $2x10^{-4}$	0.6 to 4.3
Maximum Weighted Average Used for TS	$7x10^{-4}$ to 0.005	$9x10^{-5}$ to $6x10^{-4}$	1.8 to 13
Class F Fumigation	0.017 to 0.12	$2x10^{-3}$ to 0.016	46 to 330

[a] 0.0071 to 0.052 Ci.

As before, the probability of Class F fumigation conditions existing and the scrub pumps failing simultaneously while the silica gel beds are bypassed is less that one in one-to-ten thousand. Further, the probability of the RSS failing, also, is some small fraction, probably smaller than 1/100. Therefore, the probability that a dose from the accidental release exceeding the current average daily dose from particulate releases by more than a factor of 5 or 10 is less than one in 10^5 to 10^6.

One may conclude that the spirit of ALARA would not be violated by allowing the silica gel beds to be bypassed for washing, if it is not operationally feasible to isolate them, while calcining HLW. However, every effort is made to have the silica gel adsorbers on line at all times.

Other actions could potentially be taken while washing the beds. These include feeding inactive waste during the period that the beds are bypassed, shutting off feed, and shutting down calcination operations. The latter may result in additional worker exposures many orders of magnitude greater than the potential off-site doses calculated for continuation of calcination of HLW because of maintenance problems normally associated with shutdowns. Feeding of inactive waste would dilute the radioactive and toxic constituents of the calcine bin contents, increasing overall waste volume. See other considerations in Section IVF.

The feed might be shut off for a period to bypass and wash the silica gel beds. It would be necessary to continue burning some kerosene to avoid creating oxidizing conditions that might volatilize bed ruthenium. While feed is shut off the bed would be partially ground to fines. This could not continue longer than a day. However, even this is very tenuous. The bed temperature soon becomes very unstable and tends to trip the RSS. This option is not recommended.

Whether the 0.19 mCi release during washing of the silica gel beds occurred from calcination operations or from the bed washing procedure, itself, is difficult to evaluate. That no increase in release occurred during the 6/6/88 and 8/22/88 bypasses and washings (Figure 3) is indicative of negligible ruthenium release from the calcination operation. It is possible that the washing operation on August 3 dislodged particles that were blown out when the beds were placed back on line or the drying procedure volatilized ruthenium. To evaluate the mechanism in the future, stack samples will be pulled just prior to and after placing the beds back on line following a washing operation.

In a similar vein, it is possible that during the August 20-21 period when the bypass valve was opened, but the beds were not valved out, water condensed in the beds, dissolving sorbed ruthenium that became entrained in water aerosol droplets.

C. Decontamination of Cell Floor

The cell off gas is treated only by HEPA filtration. If volatile ruthenium were formed during dissolution of calcine material on the cell floor, the absence of scrubbing and silica gel beds would result in a significant fraction of the volatilized material being released. Approximately 0.02 percent of the ruthenium volatilized during the bed dissolution on October 30, 1988 in 13 M HNO_3 was released to the stack (see Section IVG). The decontamination factors of the scrubber system for gaseous and particulate ruthenium are about 13 and 100, respectively.[2,3] If a value of 100 is assumed, then 2 percent might be released in the absence of the scrub system. It is also likely that much of the volatilized ruthenium from the calciner bed was reduced by hydrocarbons present in the off-gas system. Since these are not present in the cell, one may expect an even greater percentage to escape the HEPA filters, perhaps 10 to 25 percent.

If one considers 25 percent release, the quantity of bed material that might be dissolved without controls on acid concentration may be estimated by establishing an acceptable release quantity. For the present purpose, assume it to be that which would yield a maximum off-site dose comparable to that which is typical from normal particulate release (0.00037 mrem/day). Consider, further, that meteorological conditions are average. The allowed ^{106}Ru release would then be (see Table 1) 0.00037 mrem/0.031 mrem/Ci = 0.012 Ci. The allowed source in the calcine would be 0.012Ci/0.25 = 0.048 Ci. Characteristic concentrations of ^{106}Ru in calcine are approximately 2 Ci/kg. Thus, only 0.024 kg of calcine could be dissolved under uncontrolled conditions.

It is obvious that chemical conditions must be controlled to limit ^{106}Ru releases. Available data for evaluating acceptable conditions are provided in Figures 4 and 5. Figure 4 represents conditions for which negligible ruthenium is volatilized in a fuel dissolver over 20 hours.[4] It is somewhat dependent on gas flow rate. The volatilization factor in Figure 5 is defined as the concentration of ruthenium in the vapor phase divided by the concentration in the aqueous phase for boiling nitric acid solutions.[1,5]

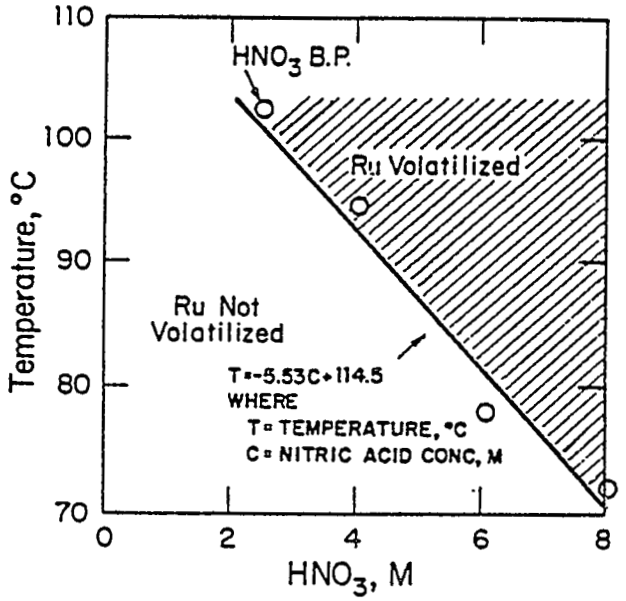


Figure 4 Ruthenium volatility from HNO_3 dissolver.

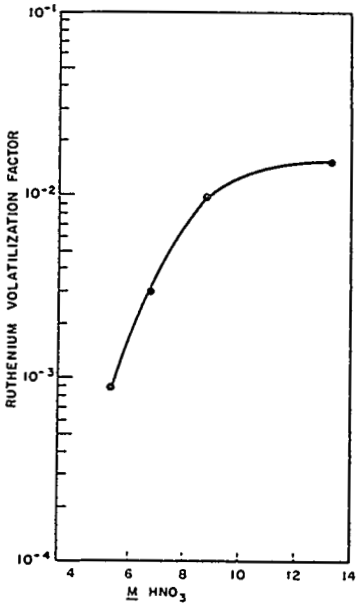


Figure 5 Effect of acid concentration on ruthenium volatility from distilling nitric acid solutions.

If there is sufficient calcine on the floor to result in an exothermic reaction that could result in heating, the acid concentration must be limited. If boiling is assumed, Figure 4 indicates a 1.7 M limit. This is impractical, as reaction would not occur. If a higher concentration is used, although volatility may not be zero according to Figure 4, its departure from zero would be very small, initially. Figure 6 (from the same study as Figure 4)[4] indicates that at 4 M, the 20 hour release would be ca. 0.5 percent, if boiling occurred. Figure 5 indicates that the volatilization from boiling 6 M HNO_3 would be approximately 7 times that at 4 M, consistent with the data of Figure 6. This factor change is probably about the same in going from 2 M to 4 M. Thus, release from boiling 4 M would be approximately an order of magnitude higher than the "zero" release from 1.7 M HNO_3 and, thus, be small.

The provisional recommendation is to use 3 to 4 M HNO_3 for a first "hit" in dissolving calcine from the floor. Every effort should be made to limit the temperature to 92°C for 4 M and 98°C for 3 M. After the calcine has been largely dissolved, additional flushes can be done with 6 M HNO_3. Since nitrate is really the important parameter, rather than acid, if $Al(NO_3)_3$ is used for complexing fluoride, the

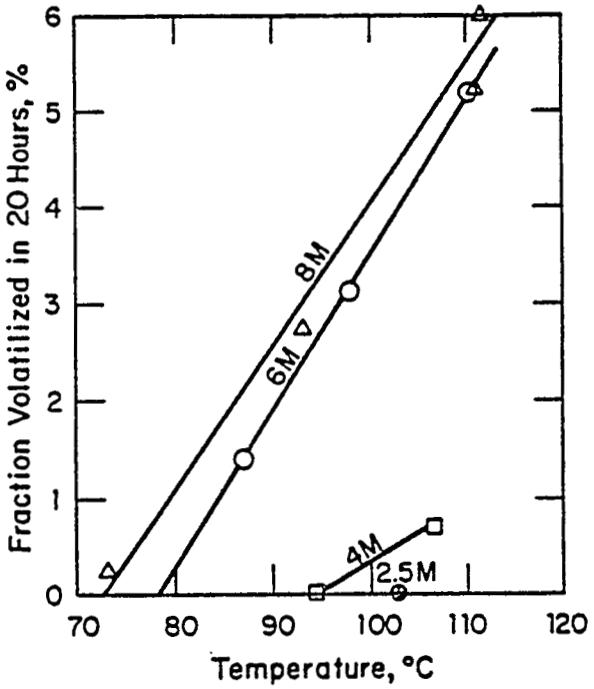


Figure 6 Evolution of Ru from HNO_3 dissolver.

acid concentration should be decreased by 3 times the aluminum concentration. Boric acid (0.2 M) is the preferred complexing reagent.

A final recommendation will be made following dissolution studies of hot calcine when it is available after restart of the NWCF.

D. Decontamination of Off-Gas Lines

Downstream of Silica Gel Adsorbers. Since there are no means to remove volatilized ruthenium, other than reduction by organics on the pipe walls, it is necessary to avoid forming a vapor species. This requires operating in the range where temperature and nitric acid concentration are less than specified by the approximate relation $t = -5.53c + 114.5$ (Figure 4). If 13 M HNO_3 is permitted, the temperature must not be allowed to exceed 40°C. If 6 M HNO_3 is used, the temperature can be 80°C. The total nitrate concentration is actually what should be controlled. Thus, if $Al(NO_3)_3$ is added for complexing, the nitric acid concentration should be reduced by three times the aluminum concentration.

The decontamination solution is normally heated to 100°F (38°C) prior to use, but at times, it has been heated to as high as 150°F (65°C). This may be cooled some during transport to the region being decontaminated. However, accumulation of calcine fines on the pipe can result in an exothermic reaction. The heat generation and temperature cannot be predicted or controlled. The same considerations for dissolving the calcine apply as discussed in Section IVC.

Upstream of Silica Gel Adsorbers. The adsorbers should provide a decontamination factor of 10^3 for volatilized ruthenium. The ratios of ruthenium volatilized from boiling nitric acid solutions at 13 and 6 M versus that from 4 M HNO_3 are about 62 and 6.9, respectively (based on Figure 5). If 13 M or 6 M HNO_3 were used, the volatile ruthenium release relative to that from using 4 M HNO_3 downstream of the adsorbers would be (62 or 6.9)/1000 = 0.062 or 0.0069, respectively. The former could release measurable quantities of Ru-106 if boiling occurred. The latter would be negligible.

A maximum nitrate concentration of 6 M should be used in decontaminating off-gas lines ahead of the silica gel adsorbers. In order not to challenge the adsorbers, if calcine is present that would result in substantial exothermic heating, initial dissolutions should be done with 4 M HNO_3.

E. Filter Leaching

The 100 scfm off gas from filter leaching is treated with a packed column water scrubber, blended with the 20,000 scfm HVAC air, and discharged through HEPA filters to the roof of the NWCF. The scrubber would remove approximately 90 percent of any volatilized ruthenium.

In Section IVC, the assumed allowable release over a 24 hour period to the main stack was 0.012 Ci, which would result in an offsite lung CDE equal to the average daily bone CDE from normal particulate releases. The dispersion from the NWCF to the site boundary would be approximately half that from the stack, which would limit the allowable release accordingly to 0.006 Ci over 24 hours. This would correspond to 0.06 Ci released to the scrubber.

The maximum radiation field observed on a filter is approximately 250 R/hr. This corresponds to a few tenths of a curie of Ru-106, assuming the activity is primarily from that source. One or two filters are leached over a day. The source term, under uncontrolled dissolution conditions, exceeds the proposed guideline for release.

Leaching should be done with 6 M HNO_3 at 80°C to assure that the ruthenium does not volatilize. Other acceptable conditions can be defined by maintaining the nitrate concentration and temperature below values specified by the Figure 4 relationship $t = -5.53c + 114.5$, where t is temperature in degrees centigrade and c is nitrate molarity.

F. Feeding of Cold (Simulated) Waste

A bed that contains ruthenium has the potential for releasing volatilized ruthenium when feeding a nitrate solution. The following data were collected using a 3-inch diameter stainless steel calciner with an off-gas sampling system at the exit of the calciner. The normal measured ruthenium volatility during calcination of ruthenium-containing synthetic (inactive) zirconium waste with in-bed combustion at 500°C was 35 percent but ranged between 20 and 62 percent, depending on efficiency of burning.[1] (This represents initial volatility in the calciner, but in the NWCF the vapor species is quickly reduced to RuO_2 and removed in the scrubber.) When the

calciner was started up using a ruthenium-containing bed and various startup feeds without ruthenium, quantities of ruthenium vapor transported into the off gas, relative to that normally transported while calcining the same volume of ruthenium-containing HLW were as follows.

Startup Feed	Off-Gas Ru During Startup / Off-Gas Ru During Calcination of Ru-Containing HLW
0.7 M H^+, 1.6 M Al^{+3}, 5.5 M NO_3^-	0.080
1.6 M Al^{+3}, 4.8 M NO_3^-	0.012
Zr Waste w/out Ru	1.7

The only measurements of ruthenium volatilized from a bed while feeding non-ruthenium feed at normal calcination conditions were made for a bed of synthetic AGNS (Barnwell) commercial calcine while feeding normal synthetic HLW without ruthenium. At 500°C or higher temperature, the amount of ruthenium transported in the off gas was the same as it would have been had the feed contained ruthenium, whether heating was done indirectly or with in-bed combustion. At 440°C or lower, with indirect heating, it was about 3 percent as much as for the ruthenium feed. Evidently, at the high temperatures, where percent volatility from the feed is low, less than 0.03 percent, the quantity of ruthenium in the off gas is fixed by the partial pressure of ruthenium in a pseudoequilibrium vaporization process. At the lower temperature, where ruthenium volatility exceeds 0.65 percent, this is less important compared to another mechanism.

The lower temperature results may be more applicable to calcination of zirconium waste at 500°C, because of its normally high percent volatility.

The only conclusion that can be definitely drawn is that some bed ruthenium may be volatilized during calcination of or restart with cold waste. The unanswered question is how much. It most likely would be a few percent of that normally volatilized. The quantity can be minimized by minimizing the nitrate concentration in the synthetic feed to the extent possible. Also, minimizing the feed rate and, therefore, volume will have a direct effect.

Considering the negligible impact of feeding HLW while bypassing the adsorber beds for washing (see Section IVB), when the calciner is operating normally, no significant benefit can be gained under such conditions by feeding synthetic waste. It could, however, mitigate maximum potential releases during postulated accident conditions by minimizing the source. Again, the potential benefit is small.

G. Calcine Bed Dissolution

Occasionally, the fluidized bed of calcine will solidify or "clinker" for various reasons. When this happens, it must be dissolved in nitric acid. This has occurred three times in the NWCF. The recorded ^{106}Ru releases (always as particles in the stack monitors) are indicated in Figure 1. On the first occasion, March

1984, the bed was dissolved in boiling 13 M HNO_3 and the silica gel adsorber beds were on-line (along with the scrubber system, which was on-line for all bed dissolutions). The ^{106}Ru release during the month of the bed dissolution was, at 0.25 mCi, slightly below the background for the previous and following months.

The second bed dissolution was done during late November - early December 1984 using 6 M HNO_3 at 80°C, again with the silica gel adsorbers on-line. The ^{106}Ru releases during November and December were 0.23 and 0.24 mCi, compared with background releases during the prior and following months of 0.0095 and 0.0053 mCi. It is difficult to correlate the November-December releases directly with the bed dissolution; the observed releases may be the result of normal variation in the background. At any rate, it is apparent that, when the silica gel adsorber beds are on-line, little difference occurs in ^{106}Ru releases, whether boiling, 13 M HNO_3 or 6 M HNO_3 at 80°C is used to dissolve the bed.

In October 1988, a clinkered bed was 90 percent dissolved in boiling 13 M HNO_3, but with the silica gel beds bypassed. A 140 mCi ^{106}Ru release occurred over one hour. The dissolution was terminated by stopping acid addition after the first 20 minutes as a result of a stack monitor alarm. A total of 170 mCi (0.004 percent of bed inventory) was released over 24 hours, compared with background daily releases at the time of less than 0.005 mCi. It is clear that the boiling 13 M HNO_3 results in volatilization of some ruthenium that finds its way through the off-gas system (exclusive of silica gel adsorbers) to the stack, where, after dilution with plant ventilation air, it is quickly reduced to particulate RuO_2. Furthermore, the silica gel adsorbers effectively remove the volatilized ruthenium, as evidenced by the March 1984 results.

In order to understand the mechanisms leading to the October 1988 release, the fraction of available ruthenium in the dissolved bed that was volatilized was estimated to be one-fourth. This estimate was based on the concentration of ruthenium in the vapor being 0.014 times the concentration in the boiling 13 M HNO_3, as determined from Figure 5, air sparging flowrate and time, and an assumption of saturation of the sparge gas with the ruthenium vapor. If the sparge gas was not saturated, the fraction of ruthenium released was less than one-fourth but likely by no more than an order of magnitude. Of the amount volatilized, approximately 0.02 percent (based on one-fourth volatilized) passed through the off-gas system and reached the stack monitors as particulate ruthenium.

The small release was likely caused by the formation of a volatile nitrato species of ruthenium formed in the boiling 13 M HNO_3. Above ca. 6 M HNO_3, ruthenium is substantially oxidized to a volatile form. The volatility is a maximum in the 10 to 14 M range.[1] Once the nitrato vapor species is formed and swept into the off-gas system, it probably begins to decompose to the particulate RuO_2 form as the partial pressure of HNO_3 is decreased by condensation and dilution. (The mole fraction of HNO_3 vapor above boiling 13 M HNO_3 is 0.21.) For the small fraction that passes through the particulate removal system as a vapor, when it is blended and greatly diluted with plant ventilation air, the subsequent dissociation is rapid and it appears at the samplers as particulate material.

The 0.17 Ci particulate ^{106}Ru release resulted, for the meteorological conditions existing at the time, in a dose to the maximally exposed individual of 0.006 mrem lung collective dose equivalent and 0.002 mrem effective dose equivalent. Had Class F fumigation conditions existed during the hour of 0.14 Ci release, the doses would have been increased by a factor of 56. As small as these are, the operating policy is to control special process operations, when reasonably possible, to maintain radionuclide releases near the normal operating background releases. Thus, the dissolution of the remaining 10 percent of the bed was carried out under conditions that would guarantee no additional release above background and, yet, assure bed dissolution.

A primary change was to require the silica gel beds to be on-line, which the earlier bed dissolutions showed to be effective in collecting any volatilized ruthenium. As a precautionary added measure, chemical conditions were maintained to essentially avoid challenging the silica gel adsorbers.

Based on laboratory studies of dissolution of synthetic clinkers,[7] the maximum dissolution rate occurs in 6 M HNO_3, 0.3 M $Al(NO_3)_3$ at 80°C. [$Al(NO_3)_3$ is present to complex HF released from the dissolution of CaF_2 in HNO_3.] Increasing the temperature or concentration did not substantially increase the rate. (It should be commented that the motivation for using 13 M HNO_3 earlier was to minimize the quantity of resulting high-level liquid waste.) Based on these results, and ruthenium volatility from boiling 6 M HNO_3 relative to that from boiling 13 M HNO_3 estimated from Figure 5, a concentration of 6 M HNO_3, 0.15 M $Al(NO_3)_3$ was selected for dissolving the residual bed material at a temperature between 80 and 90°C. An excess of acid was used such that the concentration during reaction did not decrease below 4 M so that a practical dissolution rate could be maintained. Zirconium, aluminum, and boron constituents of the calcine provided sufficient complexing of HF without the need for the usual 0.3 M $Al(NO_3)_3$, thereby allowing the nitrate concentration to be minimized. Boric acid would be a preferred complexing reagent, but it was not practical to add it.

During the final bed dissolution, the temperature never exceeded 80°C. With the silica gel beds on-line, the ruthenium release, as expected, did not increase above the background. The background at the time, following the 0.17 Ci release, remained higher than normal at 50 to 100 μCi/day for approximately 1½ weeks and then gradually decreased to the normal few microcuries per day over the next three months, during which the calciner was not processing waste.

When the next calcination campaign begins in the fall of 1990, radioactive calcine product will be obtained and used to perform laboratory dissolution studies under varying conditions of HNO_3 concentration, temperature and additives to determine the ruthenium volatilization behavior. The information will be used to establish a permanent bed dissolution procedure that can be used while minimizing the high level waste produced.

V. Conclusions

Ruthenium-106 can be volatilized during certain operations of the NWCF involving nitric acid. The vapor species is stabilized by the presence of nitric acid vapors and is partially reduced to particulate RuO_2 from the presence of reducing agents, such as hydrocarbons, in the off-gas lines and as the nitric acid/NO_x vapors become diluted with process or blended ventilation air. This results in removal of most of any volatilized ruthenium by the particulate removal devices. If silica gel beds are on-line they effectively remove residual ruthenium vapors.

Any residual ruthenium vapor that can enter the stack and resulting doses to the public are very small. However, it is desirable to maintain releases to near background at all times. Based on process, laboratory, and literature data, it has been possible to define practical chemical conditions for all operations in the NWCF that will assure this and, as a defense-in-depth measure, avoid challenging the silica gel adsorber beds.

VI. References

1. Christian, J. D., "Processing behavior and control of ruthenium and cerium," in Controlling Airborne Effluents from Fuel Cycle Plants, Proceedings of ANS-AIChE Meeting, Sun Valley, Idaho, August 1976.

2. Christian, J. D., and Rhodes, D. W., Ruthenium Containment During Fluid-Bed Calcination of High-Level Waste from Commercial Nuclear Fuel Reprocessing Plant, ICP-1091, January 1977.

3. Rhodes, D. W., Freeby, W. A., and Christian, J. D., "Ruthenium containment during fluidized-bed calcination of high-level waste," AIChE Symposium Series, 75, No. 191, 69-75 (1979).

4. Ondrejcin, R. S., "Volatile ruthenium chemistry, an answer to RTA 925-3," DuPont Savannah River Laboratory Internal Report DPST-80-254, January 31, 1980.

5. Wilson, A. S., "Ruthenium volatilization in the distillation of nitric acid," J. Chem. & Eng. Data. 5, 521-4 (1960).

6. Christian, J. D., unpublished results, 1975 and 1976.

7. Newby, B. J., Westinghouse Idaho Nuclear Co., Idaho Falls, Idaho, unpublished results, 1979.

Appendix. Estimate of Potential Ruthenium-106 Release During Rapid Shutdown of Calciner with Scrubber and Silica Gel Beds Out of Service

Should the venturi scrubber pumps fail, the off-gas temperature will increase and the Rapid Shutdown System (RSS) will be activated. This activation occurs when the off-gas temperature increases from the normal 150°F (65°C) to 300°F (149°C). Approximately 1 to 2½ minutes elapse during this excursion. The feed nozzles are then shut off in 1 second, the fuel and O_2 are stopped, and the heaters for the fluidizing air are shut off. Between the time that the scrubber fails and the feed nozzles are shut off, transient conditions might result in volatilized ruthenium reaching the silica gel beds. If they were simultaneously out of service, some small release might occur. Conservative calculations below, based on observed releases with the silica gel beds bypassed during calcination and during bed dissolution, indicate that the potential release is far less than 3 percent of the TS limits.

Assume 2.5 minute response to shut off feed nozzles after scrubber failure and maximum feed rate of 160 gal/hr (600 L/hr) with 0.6 Ci ^{106}Ru/gal (0.16 Ci/L). Source term is 4 Ci during the transient. Two methods are used to estimate the ^{106}Ru potential release.

Method 1

During bed dissolution with the silica gel beds bypassed and the scrubber on, 0.019% of the volatilized Ru was released (see Section IVG). The decontamination factor (DF) of the scrubber for Ru vapor is 10. A conservative calculation assumes that all of the source term is volatilized. This brackets the condition in which the kerosene feed is stopped. Then the potential for release is 10 x 0.019 = 0.19%, or 0.0076 Ci. This is 3 percent of the TS limit. It is extremely unlikely that this much release would occur.

Method 2

During calcination with the scrubber operating and the silica gel beds bypassed, as much as 0.19 mCi Ru-106 was released over 24 hours. The quantity fed over 24 hours, while the feed rate was 150 gal/hr with 0.6 Ci/gal, was 2200 Ci. Thus, 8.6×10^{-6}% was released.

For conservativeness, assume the scrubber efficiency of 600 for particles. Then, without the scrubber, the release would be 600 x 8.5×10^{-6}% = 0.005%. 0.005% x 4 Ci = 0.0002 Ci released, or 0.08 percent of the TS limit.

DISCUSSION

HOLMAN: Whilst I realize that Dr. Christian's paper deals largely with the behavior of ruthenium from this facility, I find it surprising that you can run any form of calcine without NO_x control and iodine control.

CHRISTIAN: Good point. In future years we will not be permitted to operate in that manner. We currently have an NO_x selective catalyst reduction process on a slip stream. It is a pilot plant for our NWCF off-gas system. It will be tested during the next campaign scheduled to start in the middle of September 1990. Until now we have not been required to destroy NO_x. In fact, the site boundary concentrations are well below the clean air standard of 0.05 ppm. However, the State of Idaho and EPA have dictated that we destroy NO_x. I don't know the exact schedule, but sometime in the next 5 or 6 years we will perform selective catalytic reduction of NO_x. Any ruthenium vapor that reaches the SCR bed is reduced by NH_3 and completely removed in the first centimeter of the bed.

HOLMAN: Doesn't the waste itself contain a significant amount of iodine-129?

CHRISTIAN: The quantity of iodine-129 in the processed fuels has been sufficiently small that the site boundary doses resulting from releases during fuel dissolution and calcination of HLLW have been on the order of 0.1 mrem/yr EDE, well within the 10 mrem/yr DOE INEL limit and 1 mrem/yr limit allocated to ICPP. As processing rates increase in the future, it may become necessary to control iodine releases. Assessments are currently being made and technologies are being evaluated for removing iodine from off gases.

BERGMAN: Volatile species often get a free ride on particles and are able to penetrate the gas trapping system. An example of this occurs with tritium getting through tritium traps such as silica molecular sieves by riding on aerosols generated by vacuum pumps. Please comment on the possibility of volatile ruthenium behaving in a similar fashion.

CHRISTIAN: As was indicated in the paper, most of any volatilized ruthenium is, in fact, removed by particulate removal devices. This may be due to a combination of reduction of the ruthenium vapor species to RuO_2 by organics or, when the gases are diluted or cooled, dissociation of the nitrato vapor species as a result of HNO_3/NO_x dilution by ventilation air, and, possibly, some sorption on aerosols. However, once sorbed, ruthenium would likely remain in a reduced state. When the old WCF operated with indirect heating, where reducing conditions were not present, the silica gel beds effectively removed the ruthenium vapor species, as they did during dissolution of the calcine bed in the NWCF in boiling 13M HNO_3. This is quite different from H_2O, which is well known for its ability to sorb on surfaces, especially, as its pressure approaches saturation. If we didn't have particle removal devices , we would probably have a problem.

FURRER: Have you watched for poisoning of the ruthenium filter? I saw that you got a high decontamination factor. What was the lifetime of your ruthenium filter?

CHRISTIAN: Since upstream samplers are not operational, it has not been possible to routinely measure the operating DF of the silica gel beds. As indicated in the paper, when the beds are bypassed for washing during normal calcination, no increase in ^{106}Ru release is usually observed. This, of course, could mean that the beds make no difference, but more likely they simply are not normally challenged. The fact that the ^{106}Ru release during the bed dissolution in boiling 13M HNO_3 in March 1984 when the adsorber beds were on-line did not increase above background and was 1/700 of the release during a similar dissolution October 1988 when the adsorber beds were off-line indicates that the beds, which have been used since startup in September 1982, are still effective (DF 700) at the millicurie/day level, or a fraction of a milligram of ruthenium/day in the few hundred to 2000 scfm

stream. Early development studies by Newby indicated an expected DF of 10^3. Also, experience in the old WCF showed operational DFs of ca. 800. The capacity of the beds is 40 kg ruthenium, so they will never be saturated. The periodic washing of the beds in dilute HNO_3 (to remove particles and organics) appears to maintain their effectiveness.

THE INCINERATION OF LOW LEVEL RADIOACTIVE WASTES - CURRENT STATUS

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Abstract

Incineration is a state-of-the-art, proven volume reduction method that can make a significant contribution to the long-term management of low level radioactive wastes (LLW). The application of incineration at government, industry, institutional, and commercial nuclear power plant facilities has proven to be both technically feasible and efficient, and can be an economical part of a complete LLW management program. A regulatory structure supportive of incineration is in place at the State and Federal levels, although public concerns and limited economic incentives are major issues that must still be addressed before incineration becomes as wide-spread a practice in the U.S. as it is in other countries. This paper summarizes the domestic and international status of LLW incineration and is intended primarily to serve as an overview of the technology and related licensing issues for waste management professionals involved in its use or regulation.

The Technology

Types of Incinerators and General Design Requirements

The technology for the incineration of LLW is a specialized and carefully engineered form of incineration developed to ensure essentially complete combustion of the burnable waste and positive retention of the radionuclides. The current technologies range from the common types of excess and controlled air incinerators, including fluidized bed systems, to the advanced concepts of high-temperature slagging, molten-salt incineration, pyrolysis, and acid digestion. These LLW incinerators are highly efficient at controlling both radionuclide releases and conventional waste emissions such as acid gases, heavy metal compounds, and toxins; in addition, the magnitude of these emissions is far less than the level of similar releases from fossil-fueled or waste-fueled power plants because of the relatively small volumes of waste processed in the incinerators.

Most modern incinerator systems have a number of common basic features. These include:

- Waste sorting & pre-processing system
- Waste feed system
- Combustor: Primary, sealed (or sub-atmospheric) main combustion chamber with auxiliary heaters

- Afterburner: secondary, afterburning combustion chamber with auxiliary heaters (Note: some systems do not have afterburners)
- Ash collection system
- Dry and/or wet off-gas and fly-ash scrubbing/filtering system
- Off-gas release monitoring system

Incinerator systems also have a number of experienced-based limitations on operating parameters and waste input concentrations; although these limitations are not strict design requirements, they do represent good practices that can minimize maintenance and monitoring requirements. Currently accepted practices include:

- Minimum primary and secondary combustion chamber temperatures, typically in the 500-1200 °C range, to ensure complete combustion.

- Maximum allowable input amounts of plastics (such as polyvinyl chloride) and sulfur-bearing wastes to limit the amount of chlorine to about 5-10 wt % and sulfur to about 1-5 wt % (to limit acid-gas formation to manageable levels).

- Maximum allowable amounts of certain radionuclides such as tritium, carbon-14, and sulfur-35, which are weak beta particle emitters difficult to monitor in the off-gas stream.

Typical LLW for Incineration

LLW appropriate for incineration typically consists of radioactively contaminated paper, wood, rubber, rags, plastics, oils, solvents, sludges and resins, as well as industrial, medical and biological research wastes. Table 1 shows the characteristics of radionuclides usually found in nuclear facility waste. Average LLW is about 80-99 percent combustible, although the pre-incineration sorting process may leave small amounts of non-combustibles such as glass and metal parts. LLW is generally classified as dry or liquid waste.

LLW is normally designated by 10 CFR Part 61 as Class A waste in terms of shallow-land burial requirements, but may be sufficiently contaminated to be become Class B or C waste. Wastes contaminated with transuranics (typically alpha-emitting radionuclides) are designated as Class A or Class C waste if the activity levels do not exceed 10 or 100 nanocuries per cubic meter, respectively. Greater-than-Class-C TRU waste is also considered to be LLW because of vagueness in the legal definitions. With very low levels of contamination, TRU waste can be incinerated using standard LLW incineration methodologies. Normally, most industrial, academic, and research LLW has only very low concentrations of radioactive materials. Furthermore, typical medical waste is characterized by even lower concentrations, with the majority of the contaminating radionuclides being short-lived with half-lives ranging from tens of hours to a few months.

TABLE 1

CHARACTERISTICS OF RADIONUCLIDES
ASSOCIATED WITH NUCLEAR FACILITY WASTE

NUCLIDE	HALF LIFE	EMISSION MODE	TYPICAL-PERCENT-ABUNDANCE*		
			PWRs	BWRs	PHWRs
Cr-51	28 d	Gamma		3	14
Mn-54	312 d	Gamma	5	26	
Co-57	272 d	Gamma	1		
Co-58	71 d	Beta,Gamma	35	1	
Fe-59	45 d	Beta,Gamma			
Co-60	5.3 y	Beta,Gamma	31	29	30
Zn-65	245 d	Beta,Gamma		2	7
Sr-90/	29 y	Beta			
Y-90	64 h	Beta,Gamma			
Zr-95/	66 d	Beta,Gamma		<1	5
Nb-95	35 d	Beta,Gamma	<1	<1	5
Ru-103	39 d	Beta,Gamma			
Ru-106	1.0 y	Beta			<1
Cd-109	1.3 y	Gamma			
Sb-125/	2.8 y	Beta,Gamma	<1		
Te-125	58 d	Gamma			
I-125	60 d	Gamma			
I-131	8 d	Beta,Gamma			
Cs-134	2.1 y	Beta,Gamma	9	9	
Cs-137	30 y	Beta,Gamma	17	14	14
Ce-141	33 d	Beta,Gamma			
Ce-144	284 d	Beta,Gamma			11
U-235	7.04E8 y	Alpha,Gamma			
U-238	4.47E9 y	Alpha,Gamma			
Pu-239	2.41E4 y	Alpha,Gamma			

Notes:

PWRs - U.S. Pressurized Water Reactors; 1% of nuclides not specified.
BWRs - U.S. Boiling Water Reactors; 15% of nuclides not specified.
PHWRs - Canadian Pressurized Heavy-Water-Moderated and Cooled Reactors (i.e., CANDUs); 13% of nuclides not specified.

* Radionuclides listed in this table, but not associated with PWRs, BWRs, or PHWRs, may be found in LLW at other types of reactor/nuclear facilities.

Air Cleaning: Off-Gas Treatment

The off-gas stream from a LLW incinerator is a complex mixture of gases, condensible vapors, entrained liquid water, and solids. The high temperature combustion process itself is the first step in chemically stabilizing the input waste, since the large organic molecules tend to be broken down into simpler, less reactive compounds. Tables 2 and 3 describe some of the important combustion products. Many of the radionuclide combustion compounds that form are insoluble, thereby allowing them to be easily filtered (and reprocessed in the case of plutonium). Although the increased chemical stability of the off-gas is a positive feature, most of the resulting combustion products are still noxious and potentially harmful to individuals and to the environment. Therefore, the proper treatment of the off-gas stream is essential in preventing the uncontrolled release of both radioactive and non-radioactive material.

Radioactive materials may be released directly as volatilized radionuclides (e.g., iodine, ruthenium, and cesium) and as radioactive gases (e.g., CO_2, H_2O, and SO_2 formed with C-14, H-3, and S-35, respectively). These materials may also be released indirectly, while combined with organic particulates (i.e., burned/unburned combustion residues) and inorganic salts (i.e., neutralized acid residues and fluidized-bed material).

Off-gas scrubbing and particulate filtration systems for LLW incinerators as described in Table 4 are highly efficient in their primary role of controlling release of radioactive combustion products, as well as in their secondary role of limiting the release of non-radioactive products and protecting the incinerator from corrosion. Off-gas treatment systems have four principal functions related to the control of the off-gas stream:

- **Particulate control**, which consists of scrubbing (using a venturi scrubber) and filtration (using dry mechanical filters and final HEPA filters) for particulate removal

- **Gas control**, which consists of wet and dry treatment for gas absorption (in particular, to remove acid gases by combining them with caustic compounds)

- **Temperature control**, which consists of temperature reduction (though the addition of ambient air to the off-gas stream, water quenching, or cooling through a heat exchanger) and reheating for humidity control (to avoid condensation in the dry filters)

- **Moisture control**, which consists of demisters (for liquid water removal) and reheating for humidity control (as stated above under "temperature control").

TABLE 2

IMPORTANT CHEMICAL PROPERTIES OF LLW RELATED TO INCINERATION

DRY WASTE	PROPERTY
Plastic (PVC)	Contains chlorine which forms acid gas
Paper/Wood/Organic Material	Forms dioxins, furans, and polynuclear hydrocarbons
Metals	Form metal-chlorides
Organic Material Containing Proteins	Forms nitrogen oxides from oxidation of amines

WET WASTE	PROPERTY
No. 4 Fuel Oil	Contains up to 1.5 percent sulfur which forms acid gas
"FIREQUEL" (fire-resistant oil)	Contains up to 8 percent phosphorus which forms acid gas

TABLE 3

ANTICIPATED RADIONUCLIDE COMBUSTION COMPOUNDS

NUCLIDE	COMPOUND*	ALKALI SOLUBILITY
Cr-51	Cr_2O_3	Insoluble
Mn-54	Mn_3O_4	Insoluble
Co-57,58,60	CoO	Insoluble
Zn-65	ZnO	Insoluble
Fe-59	Fe_2O_3	Insoluble
Zr-95	ZrO_2	Insoluble
Nb-95,97	Nb_2O_5	Insoluble
Ru-103,106	RuO_2	Insoluble
I-125,131	I^-, IO_3^-	Very Soluble
Cs-134,137	CsCl	Very Soluble
La-140	La_2O_3	Insoluble
Ce-141,144	CeO_2	Insoluble

* Note: These combustion compounds are being described as they exit the final incinerator chamber, prior to entering the off-gas treatment system. Consequently, since combustion was likely to have occurred in an oxygen-deficient environment and since the off-gas stream is typically saturated with water vapor, less stable products may form. These products include RuO_2, instead of RuO_4, and I^-/IO_3^-, instead of elemental iodine or iodine oxide.

TABLE 4

AQUEOUS VS. DRY OFF-GAS TREATMENT FOR LLW INCINERATORS

TYPICAL AQUEOUS OFF-GAS TREATMENT SYSTEM COMPONENTS:

o Quencher	- Low energy spray towers for cooling of hot off-gas
o Particle Scrubber	- High energy venturi or fibrous bed scrubbers
o Adsorber	- Chemical removal of selected compounds and radionuclides
o Off-gas Conditioner	- Condenser of volatile compounds and off-gas reheater
o Final Filter	- HEPA filter

TYPICAL DRY OFF-GAS TREATMENT SYSTEM COMPONENTS:

o Particulate Remover	- Sintered metal, ceramic, or baghouse filters
o Acid Gas Neutralizer	- In situ fluidized bed caustic compound -or- Off-gas stream spray of caustic compound to cool and neutralize -or- Filter coated with caustic compound
o Gas Cooler	- Spray drying, cool air dilution, or heat recovery
o Final Filter	- HEPA filter

Volume Reduction Ratio

The volume reduction (VR) ratio is one measure of the efficiency of LLW incineration. It is generally defined in one of two ways: First, it is a comparison of the LLW volume before and after incineration (i.e, sorted, combustible LLW volume versus ash and residue volume); second, it is this same comparison except that the pre-incineration volume is based on the unsorted and/or packaged LLW, while the post-incineration volume includes the stabilizing/binding agents, containers, and over-packs. The former of these definitions yields larger VR ratios (since it is closely tied to the physical process of incineration) and it is the value stated by manufacturers. The latter definition, although really more of an LLW stream volume reduction ratio than an incineration volume reduction ratio, yields smaller ratios that can be useful in economic assessments of the overall potential cost savings gained through the use of incineration at a specific site.

Typical incineration VR ratios that are achievable range from 30:1 to 100:1, although some industry reports indicate that actual values of LLW stream VR ratios are closer to 10:1. These LLW stream VR ratios are very plant specific and may be lower than expected because of ash stabilization and packaging requirements, as well as because of difficulties or inefficiencies in separating combustible from non-combustible waste. By comparison, the competing technologies of compaction and super-compaction generally provide a 3:1 and a 8:1 reduction in volume, respectively. Even after the addition of ash stabilizing materials and final packaging (i.e., using the LLW stream definition for the VR ratio), the incineration process is still 2 to 5 times more volumetrically efficient than super-compaction.

Operating Experience

Typical Incinerator Operation

Most incinerators are of the excess or controlled air type, although fluidized bed systems have also been installed at several U.S. nuclear power plant sites. The newer, more advanced incinerators are generally located at national laboratories, nuclear research centers, and fuel reprocessing facilities and are being run on an R&D or demonstration basis.

Typical incinerator operation at power plants or at hospitals and universities involves LLW sorting and shredding (if necessary) at the incinerator. Regional use of an incinerator, such as provided by the Studsvik facility in Sweden, does, however, require pre-sorting and labeling of the waste containers at the site where the waste is generated. Most incinerator facilities use a semi-automatic batch feed system, although automated/continuous feed systems are being developed. In addition, many incinerators have capabilities that exceed the waste quantities to be processed on an annual basis by a factor of as much as ten -- which implies low capacity factors. For this reason, plus the requirement that an

incinerator be pre-heated during initial startup, it is usually most efficient to collect the waste and then conduct a continuous incinerator run of several weeks on a once or twice per year basis. Incinerator maintenance and cleaning are performed in the off-periods between runs. Release limits for emissions on an annual or quarterly basis are still adhered to with this approach of using short periods of continuous operation. Smaller incinerators, such as those at hospitals, may be operated efficiently on a weekly or monthly schedule.

Treatment of the off-gas for removal of fly-ash, particulate matter, and acid-gases is accomplished typically by a two-stage filtering and scrubbing system, with characteristics as noted in Table 4. Given careful waste sorting and proper operation, off-gas treatment systems can function for periods of days without being cleaned or having the filters replaced.

Ash is not normally stored permanently at an incinerator site. It therefore must be packaged, either directly as ash or in solidified form, for transport and burial off-site. After volume reduction by incineration and packaging, most LLW ash/residue is still classified as LLW in terms of its concentrations of radioactive materials.

Technical Problems & Maintenance

A number of technical problems were identified during the development of LLW incinerator technology. In the U.S., much of this development was done at the national laboratories in the 1950s and 1960s. The significant problems identified at that time have been essentially resolved with contemporary designs. Some of the major technical problems associated with incinerators are:

- Incomplete Combustion
- Insufficient Waste Sorting & Preparation
- Radioactive Contamination of the Incinerator
- System Corrosion and Wear
- Non-Optimal Waste Types & High or Low Calorific Values
- Complexity of Off-gas Treatment Systems and Uncontrolled Release of Radionuclides

In addition, maintenance is necessary for all complex systems and LLW incinerators are no exception. In general, however, the maintenance requirements are basic: First, the incinerator and the off-gas treatment system must be kept clean and, second, the incinerator must be operated in a manner that protects the inner surfaces -- especially the refractory lining.

Economics & Utilization

Incinerator costs are highly dependent on the specific application. In addition to capital costs, which are proportional to capacity and the type of incinerator technology, operational and maintenance costs are also incurred. LLW disposal costs are significantly reduced because of the volume reduction factors

achieved, but are not entirely eliminated because of the continuing need to dispose of incinerator ash as well as non-combustible waste. Furthermore, because not all LLW may be appropriate for incineration, a realistic reduction in the volume (and cost) for off-site LLW disposal may be about 50% for a nuclear power plant with a modern radwaste/incineration facility. The real economic figure-of-merit for an incineration facility should therefore be taken as the total cost for incinerator purchase and operation, plus the cost to dispose of both ash and non-burnable waste, as amortized and averaged on an annual basis. Typical costs over the last twelve years for LLW incineration and land burial, adjusted to 1990 dollars, are provided in Table 5. The degree of utilization of a given incinerator is an another important factor in the determination of overall cost effectiveness. Also to be considered, as previously mentioned, is the fact that LLW incinerators are not generally operated at full capacity. Table 6 provides some estimates of capacity/utilization factors.

Domestic & Foreign LLW Incinerators

Tables 7 and 8 list the major nuclear utility and industry incinerator facilities located in this country and overseas.

Licensing & Public Acceptance

Federal and State Licensing Requirements

Licensing and operating requirements for LLW incineration in the U.S. are based on a complex set of Federal laws and Federal/State agreements. (Licensing requirements in foreign countries are similar to the U.S. requirements, but are not addressed in this paper.) The complexity of the licensing process stems from the fact that not only is the incineration process itself regulated, but both the handling of the unprocessed waste and the incinerator by-products are also regulated. In addition, LLW may contain both radioactive and hazardous/infectious materials, with the former coming from a combination of short- and long-lived radionuclides; such a waste combination is sometimes referred to as "mixed waste". Therefore, LLW and mixed waste are regulated at both the Federal and State levels by a broad spectrum of requirements covering possession, processing, and disposal.

The basic requirements for incinerator operation are derived indirectly from the general Congressional legislation that regulates the overall handling of nuclear waste. From this basic legislation, authority has passed to the Nuclear Regulatory Commission (NRC), the Department of Energy (DOE), and the Environmental Protection Agency (EPA) to develop and promulgate specific requirements. The NRC has developed appropriate requirements under Title 10 of the Code of Federal Regulations (CFR) to regulate commercial nuclear activities, while the DOE retains control of defense and government operations. Both the NRC and DOE, however, incorporate into their guides and regulations the requirements published by EPA under Title 40 CFR. The separation

TABLE 5

EXAMPLES OF COSTS FOR INCINERATION AND LAND BURIAL

(Adjusted to Constant 1990 Dollars)

INCINERATION

Type of Facility	Cost*	Year
US nuclear power plant, PWR (Oconee)	$3160 per m^3 of LLW to reduce total annual site LLW volume from 935 to 470 m^3 for off-site burial	1987
Foreign regional facility (Canada)	$1403 per m^3	1978

LAND BURIAL

Type of Site	Cost*	Year
US commercial LLW sites (Barnwell, Beatty, Richland, Sheffield)	$1630 to $3470 per m^3 ($960,000 to $2.0 million for typical, single unit U.S. power plant)	1984
Foreign LLW site (UK)	$945 to $1890 per m^3	1988
Foreign LLW site (Canada)	$1895 per m^3	1978
US nuclear power plant, PWR (Salem)	$4240 per m^3	1990
US nuclear utility (Commonwealth Edison)	$1265 per m^3 (average of two PWR and two BWR plants)	1986

* Note: Costs for incineration and/or burial at specific facilities cannot be compared directly using these data, even though adjustments have been made to 1990 dollars, because actual costs for land and equipment are different for different years and because increasingly stringent waste management regulations make all current methods of LLW treatment and disposal more expensive than similar activities in the past.

TABLE 6

ESTIMATED CAPACITY FACTORS
FOR FOREIGN INCINERATOR FACILITIES

Facility	Waste Processed; Maximum Process Rate; Hours of Operation	Operational Period: Total Available Hours	Estimated Capacity Factor ** (Utilization Factor ***)
Bruce (Canada)	194 tonnes or 1500 m^3; 1 batch (2200 kg) per 36 hrs; 3200 hrs required	1977* = 8760 hrs	36%
	1800 tonnes; 1 batch (2200 kg) per 36 hrs; 29,450 hrs required 58,900 hrs actual	1977-1985 = 78,800 hrs	37% (75%)
Karlsruhe (FRG)	1900 tonnes; 60 kg/hr; 32,700 hrs required 35,000 hrs actual	1971-1983 = 114,000 hrs	28% (31%)
Wurenlingen (FRG)	tonnes unknown; 25 kg per hr; 5000 hrs actual	1975-1984 = 87,600 hrs	(6%)
Hinkley Pt. (UK)	Annually 1300 m^3 (approx 169 te); 70 kg per hr; 2400 hrs required annually	1977-1984 = 8760 hrs annually	28%

Notes:
* First year of operation
** Capacity factor defined as [Required hrs at maximum process rate]/[Total available hrs operation]; a low capacity factor does not necessarily mean poor performance, but more likely implies under-utilization.
*** Utilization factor defined as [Actual hrs of operation]/[Total available hrs operation]

TABLE 7

U.S. FACILITIES WITH LLW INCINERATORS

Federal Government	Industry	Nuclear Power Plants
INEL (ID)	Aerojet (CA)	Byron
Savannah River (SC)	EG&G (ID)	Braidwood
Hanford (WA)	W Columbia (SC)	Oconee
Mound (OH)	GE (NC)	Vogtle
Los Alamos (NM)	B&W (VA)	
Rocky Flats (CO)	C-E (CT)	
	Kerr-McGee (OK)	

TABLE 8

FOREIGN FACILITIES WITH LLW INCINERATORS

Government & Industry Facilities		Nuclear Power Plants
Chalk River (Canada)	CEN Grenoble (FR)	Bruce (Canada)
NV Kema (Holland)	CNRS Strasbourg (FR)	Hinkley (UK)
Mol (Belgium)	Wurenlingen (CH)	Wylfa (UK)
Karlsruhe (FRG)	Studsvik (S)	Bradwell (UK)
Juelich (FRG)	SGEA Seibersdorf(AU)	Tokai-Mura + 12
Fontenay-aux-Roses(FR)	Windscale (UK)	other plants
JAERI Oarai (Japan)	Harwell (UK)	(Japan)
Marcoule (FR)	CNEN Casaccia (I)	
Cadarache (FR)	Ispra (I)	
Bhabha (India)	JGC Corp (Japan)	
Chiba Works (Japan)		

of responsibilities between these Federal agencies is not always well defined as indicated by the recent disagreement between the DOE and the EPA over the need for a license for one of the Rocky Flats incinerators: The EPA requires a license for the incineration of hazardous waste, but the DOE takes the position that their incineration activity is part of a recovery process to reclaim plutonium from plutonium contaminated material (i.e., not a waste process) and therefore does not need an EPA license.

Specific NRC, DOE, and EPA regulations related to LLW incineration are listed in Table 9. Incineration-based legislation of most interest to the commercial nuclear industry may be found in 10 CFR Parts 20, 61, and 50 (Appendix I). Details of these regulations are highlighted in Table 10.

One item of current interest is a proposed rule that would change 10 CFR Part 20.305 to allow incineration of slightly contaminated waste oil at nuclear power plant sites. If approved, this rule would permit the incineration of waste oil without requiring a Part 50 license amendment; however, all radionuclide emission limits per Appendix I of Part 50 would still have to be met. The regulatory analysis for this proposed rule indicates that 1000 to 5000 gallons of waste oil, with typical specific activities in the range of 10^{-7} to 10^{-5} microcuries per milliliter, are generated annually per power plant. For plants at which low level wastes (including contaminated oil waste) are already being incinerated under an amended license, the total dose due just to the oil has been limited to a very small fraction (about 0.1%) of their technical specification release limit. Consequently, the incineration of waste oil is a potential candidate for below-regulatory-concern rulemaking, although the NRC has decided not to include such a decision as part of this rule.

Another unresolved issue concerns the interpretation of Federal regulatory guidance on the disposal requirements for incinerator ash. The Resource Conservation & Recovery Act (RCRA) of 1976 established categories of solid waste as hazardous and non-hazardous, although it is not clear in which category ash belongs. The RCRA defines hazardous materials as having the characteristics of corrosiveness, ignitability, reactiveness, and/or toxicity; of these characteristics, only toxicity is likely to be a feature of ash. The EPA is responsible for administering the requirements of the RCRA, but has not set minimum concentration limits for specific hazardous materials that would determine whether ash or unprocessed waste containing only small amounts of any of these materials would be classified as hazardous waste. Therefore, in the interim, the ash from the incineration of "mixed waste" is likely to continue to be treated as hazardous waste even if the radionuclide concentrations are below the 10 CFR Part 20--Appendix B limits and the hazardous materials are present only in trace quantities.

Overall, Federal legislation favors the minimizing of waste volume as an important aspect of waste management. Advocacy of volume reduction is unambiguous in the Low Level Radioactive

TABLE 9

NRC/DOE/EPA REQUIREMENTS RELATED TO LLW INCINERATION

NRC REGULATIONS FOR COMMERCIAL LICENSEES:

10 CFR Part 20, "Standards for Protection Against Radiation"

10 CFR Part 61, "Licensing Requirements for Land Disposal of Radioactive Waste"

10 CFR Part 50 - Appendix I, "Numerical Guides for PWR Effluents" NRC Low Level Radioactive Waste Policy Statement of 1981

DOE GUIDELINES FOR DEFENSE/GOVERNMENT FACILITY OPERATIONS:

DOE Order 5480.1A, "Environmental Protection, Safety, and Health for DOE Operations"

DOE Order 5480.2, "Hazardous & Radioactive Mixed Waste Management"

DOE Order 5820.2, "Radioactive Waste Management"

DOE/LLW-38T, "Guidance for DOE 5820.2 Chapter III, LLW Management (Feb 85)"

EPA REGULATIONS APPLICABLE TO NRC & DOE:

40 CFR Part 61, "National Emissions Standards for Hazardous Air Pollutants/ Standards for Radionuclides (Feb 85)"

Radiation Protection Guides (issued by the Federal Radiation Council)

TABLE 10

DETAILS OF NRC REGULATIONS RELATED TO LLW INCINERATION

10 CFR 20, STANDARDS FOR PROTECTION AGAINST RADIATION:

20.106(a,b), "Radioactivity in Effluents to Unrestricted Areas": Specific release limits in terms of concentrations of radionuclides as stated in Part 20 - Appendix B, Table 2; provision for higher release limits on case-by-case basis.

20.302, "Method for Obtaining Approval of Proposed Disposal Procedures": Requirements for obtaining approval of disposal processes not specifically allowed by regulations.

20.305, "Treatment or Disposal by Incineration": Requirement that LLW incineration must be approved via 20.106(b) and 20.302, except for C-14 and H-3 in specific wastes as provided for in 20.306.

20.306, "Disposal of Specific Wastes": Authorization to dispose of, without regard to radioactivity, up to 0.05 microcuries of C-14 or H-3 per gram of scintillation material or animal tissue. (Note: A petition for rulemaking was docketed by the Commission in Sept 1988 which would allow disposal of any solid waste containing up to 0.05 microcuries of C-14 or H-3 per gram of waste up to an annual limit of 0.1 curies of C-14 and 1 curie of H-3).

Appendix B, Table II, "Concentrations Limits for Radionuclides in Water and Air".

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10 CFR 61, LICENSING REQUIREMENTS FOR LAND DISPOSAL OF RADIOACTIVE WASTE:

61.55, "Waste Classification": Definitions of Class A,B, and C waste categories.

61.56, "Waste Characteristics": Requirements for waste (e.g., incinerator ash) characteristics to permit shallow land burial; for LLW with hazardous/infectious materials contamination, requirement also to reduce all types of contamination.

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10 CFR 50 - APPENDIX I:

"Numerical Guides for Design Objectives and Limiting Conditions for Operation to Meet 'ALARA' Criterion for Radioactive Material in PWR Effluents": Requires control of radioactive gaseous and liquid effluents to "as low as reasonably achievable" limits as approved on case-by-case basis.

Waste Policy Amendments Act (LLRWPAA) of 1985 which cites the societal benefits of a reduction in the number of waste shipments and the extension of disposal site life-times. Volume reduction can be accomplished, according to the NRC Policy Statement of 1981 on Low Level Waste Volume Reduction, by administrative controls to limit the amount of waste initially produced and by techniques of reducing the volume of the waste which is produced. Incineration, along with evaporation, fluidized bed drying, and compaction, are specifically mentioned in this NRC policy statement.

The States have no mandate to control nuclear waste in general, or waste processing (e.g., LLW incineration) in particular. With the exception of DOE regulated operations, States may elect, however, to assume regulatory authority from the Federal government for certain commercial nuclear activities. The mechanism for transfer of this authority is from the NRC through the Agreement State Program.

State regulations, as developed by the currently existing 28 Agreement States, are unique to each State. Nevertheless, all of their regulations are required to be essentially equivalent to their Federal counterparts and differ primarily in areas where the State has decided to set more conservative regulatory levels (such as for personnel radiation exposures or radioactive/hazardous material emissions) than specified by the Federal regulations.

The State of Illinois is an Agreement State which has established a very active and broad scope radioactive waste management program. The Department of Nuclear Safety is the responsible organization within that State and the rules it has developed are almost identical to those provided in 10 CFR Part 61. The primary basis for the State's regulation of radioactive waste is the Illinois Low Level Radioactive Waste Management Act (LLRWMA) of 1985. Of particular interest is how the management of LLW incineration in Illinois becomes a split Federal/State responsibility; namely, Illinois regulates the LLW disposal sites, while the NRC (for production and utilization facilities such as commercial nuclear power plants) regulates LLW incinerator licensing and operation. A specific example is the incinerator at the Byron Nuclear Power Station: Although this incinerator was authorized by the NRC via a plant license/FSAR amendment, disposal of the waste ash from the incinerator will be regulated by the State through their Department of Nuclear Safety. Illinois currently charges each utility a flat fee of $1,158,000 per year per power plant site for disposal of all LLW. Volume reduction of LLW at the Byron site prior to shipment, even though advocated by the Federal LLRWPAA, is clearly not encouraged by this type of fee structure. Nevertheless, the Illinois LLRWMA does go beyond the Federal legislation in allowing consideration of the concentration of radionuclides in the waste and the degree of treatment that the waste has received (and hence its stability) as additional factors that may influence fees. Illinois will be the host site for the Central Midwest Compact for regional disposal of LLW, so it is highly likely that the fee structure will be adjusted to encourage LLW volume reduction. In terms of non-nuclear/non-hazardous emissions (i.e., normal, fossil

fuel combustion gases and products) from a incinerator such as the one at Byron, the State EPA has authority to issue a site license using Federal EPA based emission levels. In general, the annual or quarterly quantities of these non-nuclear/non-hazardous emissions from a LLW incinerator are so much less than similar emissions from large waste incinerators and fossil power plants that regulation is not needed. A State EPA license was issued for the Byron incinerator, but only because it was specifically requested by the utility. (Some States do regulate opacity of the off-gas in addition to total quantities of pollutants, so even small incinerators would need State EPA licenses in those States.)

Public Acceptance

Public acceptance, or lack thereof, concerning operations in the nuclear industry is difficult to measure. Whether the technical issues have been communicated poorly to the public or whether the issues themselves are inherently uncommunicable in non-technical language, the net result is that public perception is presently based on a combination of their degree of trust in the regulators and their fear of the dangers if the technological safeguards should fail. The basis of this public perception, as applied to LLW incineration, can be seen in the following account of reaction to a plan to build an incinerator in a rural community. While some of these perceptions are narrowly focused on concerns about the possible release of radioactive material, others are more related to aversions about incineration in general.

In 1984, Babcock & Wilcox, Inc. announced its plans to construct a low level radioactive waste reduction facility (including an incinerator) in the town of Apollo in Southwestern Pennsylvania. The local economy needed new jobs and had supported the nuclear industry in the past, so B&W anticipated strong community support. Instead, when the company took a survey, the overall public opinion was decidedly negative, as summarized in Table 11. Generally, 3 out of 4 persons surveyed were very concerned about radiation contamination of the environment, while even higher percentages worried about lowered property values and danger to fish and wildlife. Furthermore, only 1 in 3 believed they could trust the company or the Federal/State regulators. B&W subsequently cancelled its plans for the incinerator as a direct consequence of the public opposition.

This Pennsylvania experience may not necessarily be typical of the U.S. national opinion concerning incinerator operations, but it does clearly highlight the need to foster good public relations and to provide unbiased information to allay genuine public concerns about contamination of the environment.

TABLE 11

LOCAL PUBLIC SURVEY ABOUT PROPOSED LLW INCINERATOR IN RURAL PENNSYLVANIA

CONCERNS ABOUT:	DEGREE OF CONCERN:
Radioactive contamination of water supplies	69% very concerned
Radioactive contamination of soil	71% very concerned
Radioactive contamination of air	77% very concerned
Radioactive release from a truck accident	74% very concerned
Permanence of waste storage at site	81% very concerned

TRUST IN:	DEGREE OF TRUST:
Company	31% very/somewhat trusting
U.S. Nuclear Regulatory Commission	39% very/somewhat trusting
State Environmental Safety Office	34% very/somewhat trusting

LOCAL ECONOMIC IMPORTANCE OF:	DEGREE OF IMPORTANCE:
New jobs	87% very/somewhat needed
New jobs from hazardous industry	38% very/somewhat needed
Proposed incinerator facility	12% very/somewhat needed

PERCEIVED THREAT TO:	DEGREE OF THREAT:
Property values	82% high/moderate threat
Quality of life (e.g., increased truck traffic)	78% high/moderate threat
Fish & wildlife	90% high/moderate threat

Conclusions

The incineration of LLW is a well-developed, if not yet mature, technology that can play a significant role in waste management. There are many benefits to be obtained from the use of incineration, including cost control, ease of waste handling, and a reduction in the dependency on limited-capacity, shallow-land burial sites. The initial capital costs are high compared to direct burial (even though burial costs are rapidly increasing), but the volume reduction levels achievable with incineration more than offset these costs on a long-term basis. In fact, this volume reduction capability has led the Electric Power Research Institute (EPRI) to consider the incorporation of a LLW incinerator into the standard design for the Advanced Light Water Reactor; specifically, an incinerator could help in meeting the EPRI goal of designing a "zero LLW production plant" that would store all of its own LLW on the plant site during an active 60 year operational life.

There have been technical problems associated with LLW incinerator operations, but most have been resolved with experience gained through worldwide incinerator use. As more U.S. utilities begin to operate incinerators at their nuclear power plant sites, their increasing experience level should minimize any remaining operational difficulties.

The incentives for incineration include support from Federal and State laws that explicitly permit and, in some cases, encourage the use of incineration as a volume reduction technique. Some States do restrict incineration only to the LLW generated on the site; however, this type of legislation is directed against the establishment of regional processing facilities (such as those in Europe) and not, for example, against an incinerator used to handle waste at a nuclear power plant. Overall, the licensing process in this country is very comprehensive, and may be lengthy as well, but obtaining a license is still an achievable goal. Federal and State EPA emission regulations can be met by modern incinerators, although it is sometimes prudent to limit the combustion of certain kinds of wastes, including those containing high concentrations of sulfur compounds, tritium, and toxic materials.

Public concerns exist about the risk from both gaseous and particulate releases of radioactive material from incinerators, as well as from the potentially highly toxic nature of incinerator ash. These concerns are valid and must be directly addressed if U.S. industry is to gain the confidence of the public. Not surprisingly, organizations that make a good-faith effort to keep the local communities informed about their safety programs and planned incinerator activities are more likely to have local support for their facilities. The technology behind LLW incineration has been carefully engineered and, consequently, should be viewed favorably by a well-informed public -- especially when incineration is compared to current LLW disposal practices.

Outside of this country, the decision to proceed with incineration has already been made: The Europeans, the Canadians, and the Japanese are demonstrating that proper treatment of incinerator effluents and solidification and/or packaging of incinerator ash can effectively preclude any significant short- or long-term release of radionuclides and hazardous materials to the environment. In the U.S., it is expected that the use of incineration technology will grow, particularly in the commercial nuclear power industry, as the management of LLW disposal sites is taken over by State compacts and unaffiliated States.

Finally, it is worth restating the major advantages of LLW incineration in comparison to direct shallow-land burial. The incineration process results in chemically and biologically stabilized waste and in very reduced waste volumes that can be converted into a highly insoluble form by permanent bonding with inert materials. Without the stabilizing effect of incineration, buried waste -- even compacted or super-compacted waste stored in drums -- will eventually release contaminants into the soil, water, and air. Consequently, incineration can make an important contribution to effective, long-term LLW management.

Reference

US Nuclear Regulatory Commission, The Incineration of Low Level Radioactive Waste: A Report for the Advisory Committee on Nuclear Waste, NUREG-1393, S.W. Long, June 1990.

DISCUSSION

WEADOCK: You cited costs for burial of low-level radioactive wastes in the U.S. that range from about $1,200 to $4,000 per cubic meter. You also cited the figure of about $3,000/m^3 for incineration. Is that before or after the volume reduction that is associated with incineration? Is it cheaper or not?

MOELLER: I am certain those numbers apply to the waste before it is incinerated, so in essence, the costs, at the moment, are comparable.

WEADOCK: Did the Advisory Committee make any specific recommendations in the report to the Commission on what they should do?

MOELLER: The purpose of the report was to summarize the status of the incineration of low-level radioactive wastes. Although the report was prepared at the request of the Advisory Committee on Nuclear Waste, it is a report to the Committee and is designed primarily as a source of information. The report has not been endorsed by the Committee nor does it represent an official Committee position. We simply said that we thought incineration is a viable approach that should be given further consideration.

HANDYSIDE: We do use incineration in the U.K. for some low-level wastes as you stated in your paper. But I would like to point out some cautions before you get too enthusiastic about the process. It is important to recognize that: (a) volume reduction factors may not be as large as expected when account is taken of any need to encapsulate ash for land disposal. (The reduction factor may be similar to super co-impaction. (b) secondary waste arising can be important and it is necessary to take full account of these and in particular their environmental impact. (There may be significant volumes of solid radioactive waste arising from maintenance if the incinerator proves unreliable.), and (c) when the incinerator reaches the end of its service life, it will have to be decommissioned. (This will give rise to radioactive wastes and there will be associated costs.)

MOELLER: Thank you for those comments. Each of the points you make needs to be taken into consideration.

STEPS TOWARDS THE MINIMIZATION OF PARTICULATE EMISSIONS FROM A LOW-LEVEL WASTE INCINERATION FACILITY

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Abstract

Radioactive waste, which is for the most part combustible, results from the operation of nuclear installations, the medical and industrial use of radionuclides as well as research activities. In order to achieve the greatest possible reduction in the volume of burnable waste, a batch-fed shaft reactor type furnace utilizing excess combustion air has been operated at Karlsruhe Nuclear Research Center (KfK) since 1971. An additional facility based on the same proven incineration method has been used for the handling of α-contaminated wastes since 1989. The concept for cleaning the off-gases of this new facility is primarily based on the particle removal processes of the older one. However, changes in the design of the new facility, including the addition of off-gas scrubbing, had to be undertaken with respect to more strict emission standards.

With respect to this so-called α-incineration facility, improvements in the effectiveness of the gas cleaning process and reductions in operational costs were addressed. The size distribution, carbon content and mass concentration of the particulate combustion products at the furnace discharge were characterized. The performance characteristics of two ceramic filter media for the removal of submicron particles from hot gases were established. In particular, fractional efficiency curves, determined under laboratory and actual operating conditions for temperatures up to 800°C, are presented. The experimental results are augmented by mathematical models for variations in the removal efficiencies of the ceramic media due to the influence of a number of filtration parameters. It will additionally be shown that the second stage of the hot-gas filtration and the off-gas scrubbing provided no significant contribution to the particle removal process, thereby indicating that the off-gas cleaning process can be simplified. Furthermore, an optimized and already proven concept for off-gas cleaning will be explained.

I. Introduction

In the operation of nuclear installations, the industrial or medical utilization of radionuclei as well as in the operation of large research facilities, radioactive residual waste is produced, the greater part of which is burnable. To transform it into a chemi-

cally inert form and to get the best possible reduction in volume for the end disposal, waste of this kind has been incinerated in a batchwise operated shaft furnace at Karlsruhe Nuclear Research Center since 1971. In competition with other available methods such as pyrolysis, incineration in a rotary kiln or in a circulating fluidized bed furnace, the incineration in the shaft furnace with excess air has proven itself well, especially because of its simplicity and low maintenance requirements [1].

Based on the good experience gained with the existing facilities for the incineration of β- and γ-contaminated waste, a system for the incineration of α-contaminated waste was designed to address the high waste production at the beginning of the 80's. Extensive demands on the off-gas cleaning have to be taken into consideration on account of environmental laws which have been stiffened repeatedly in recent years. According to the valid regulations [2], for small installations up to 50 MW heat output, a max. average of 30 mg/m^3 of non-radioactive dust are allowed to be emitted, when incinerating waste. As for incineration facilities for special waste like the α-system, limiting values are generally set even lower and depend on the potential threat of the harmful substances involved. Additionally, regulations for the incineration of radioactive waste set limits for peak and yearly maximum emission levels according to isotope and radiation type.

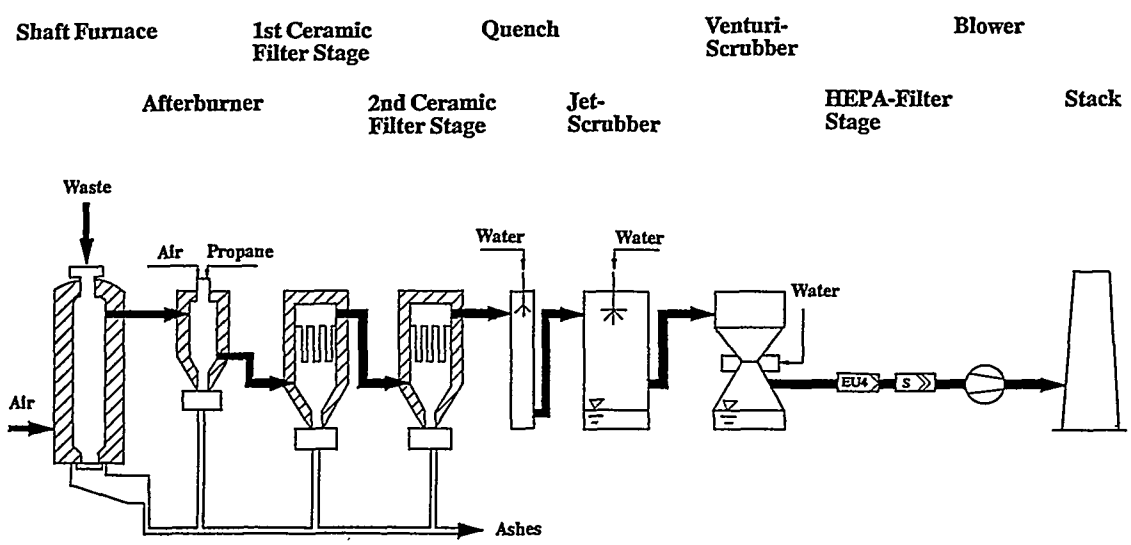

Fig.1: Schematic of the Karlsruhe incineration facility: basic construction of the facility.

Gaseous radionuclei in the burnable waste, mainly C-14, Tritium or I-129 exist in such low concentrations that special minimization measures are not necessary. The permissible emission levels for par-

ticulates, to which α-emitters preferably bind, are in comparison much lower than those for nonradioactive dusts. Since the maximum allowable emission values for harmful gases such as HCl and SO_2 can be met comparatively easily, the main challenge of the off-gas cleaning was seen in the minimization of particulate emissions.

With regard to the design of the new installation for the incineration of α-contaminated residual products, as well as to supplement the existing incineration system, extensive studies were designed to improve the efficiency and cost effectiveness of the off-gas cleaning process.

The main characteristics of the facility are a shaft furnace layed out for a capacity of 50 kg/h and the high-temperature dust filtration. In the discontinuously charged furnace the waste is incinerated at temperatures of 900°C. The off-gas passes the secondary combustion chamber at temperatures up to 1200°C. An essential item of the subsequently added off-gas cleaning is the two-stage hot-gas particle filtration in which the main part of the airborne dust is separated at temperatures of 800°C and 650°C. A quench forms the transition to the subsequent low-temperature part of the off-gas cleaning. In order to make possible the effective separation of harmful gases, especially HCl, HF and SO_2, the original procedure was enlarged by a combination of an acid lined jet scrubber and a basic lined venturi scrubber. The venturi scrubber makes possible an additional particulate filtration. A HEPA filter stage, consisting of prefilters EU4[3] and S-class[4] filters concludes the off-gas cleaning phase. The cleaned off-gas leaves the installation through the blower and the stack.

II. Particulate Emissions of the Furnace

Primary minimization measures, i.e., the reduction of particle formation in the furnace, are preferable to secondary measures based on increasing the filter efficiency. Thorough evaluation of the old furnace operation pointed out that in the first two or three minutes after feeding the furnace with waste having a high oxygen demand, very large particle concentrations appeared (Fig. 2). This formation of soot could be ascribed to the poor intermixture of air and combustion gases. Although there was excess air over the whole charge interval, there were at times notable local under-stoichiometric conditions in the furnace chamber.

The turbulence in the furnace chamber was greatly increased by an improved distribution of combustion air above the height of the furnace and especially by the tangential input of the air with high momentum. It was possible to minimize the concentration of soot particles for the problematic polyethylene waste, for a ten-minute combustion interval, from more than 12 g/m^3 to about 300 mg/m^3. The change in particle formation before and after the furnace modification is shown in Fig. 2. While the volume of the feed material is limited to 30 l for every charge by the bag size, the quantity varies between 2 and 8 kg according to the waste composition. This means, that the favorable emissive values possible will not be reached under all

circumstances. The ceramic filters are thus challenged for short times with higher particle concentrations.

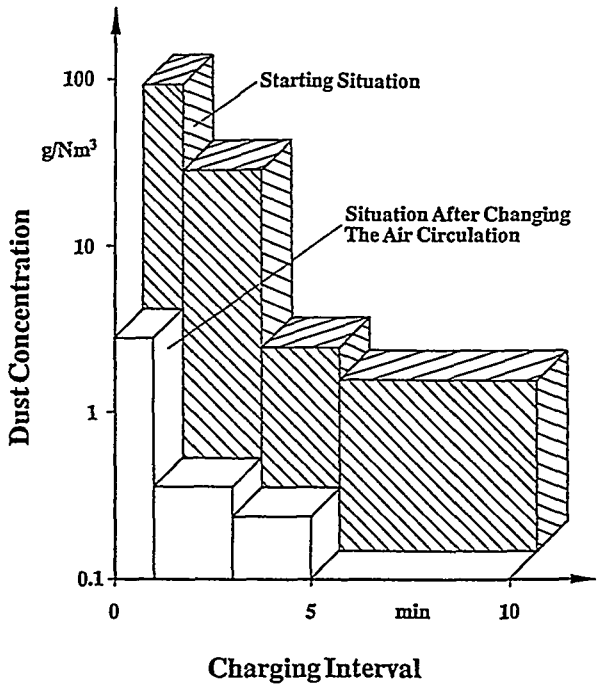

Fig.2: Formation of particles during incineration of waste with a high oxygen demand: initial operating conditions (shaded) and values after changing the air circulation.

Of crucial importance for the filtration characteristics and collection efficiency of all filters in the off-gas cleaning system is the particle size spectrum of the flue gas dust at the afterburner exit. If the particle filters are not coordinated with the spectrum of the dust particle size, an unfavorable performance in the process and particularly a low efficiency can result. This is due to the characteristic ranges of increased penetration for the candle filter, scrubber, and HEPA filter.

The size distributions shown in Fig. 3 (left) and (right) are based on impactor measurements and SEM-analysis of dust samples deposited on Nuclepore sampling filters. In the first three minutes after the charging of the furnace, approximately 75% of the off-gas dust forms (Fig. 3 (left)). Particles < 5 μm form about 90% of the dust and particles < 3 μm in diameter make up 50% of the dust. The particle diameter related to the number of particles is 0.1 μm on the average. This accentuates the extreme fineness of the particles produced in the beginning of incineration. This proportion of small diameter particles decreases very fast with the progress of final combustion (Fig. 3 (right)). By weight, 90% of the dust is composed of particles > 10 μm. Shortly after charging, predominantly soot particles are produced - as indicated by a carbon content of over 95%. The content of C in the remaining 7 minutes of the incineration interval is under 5%.

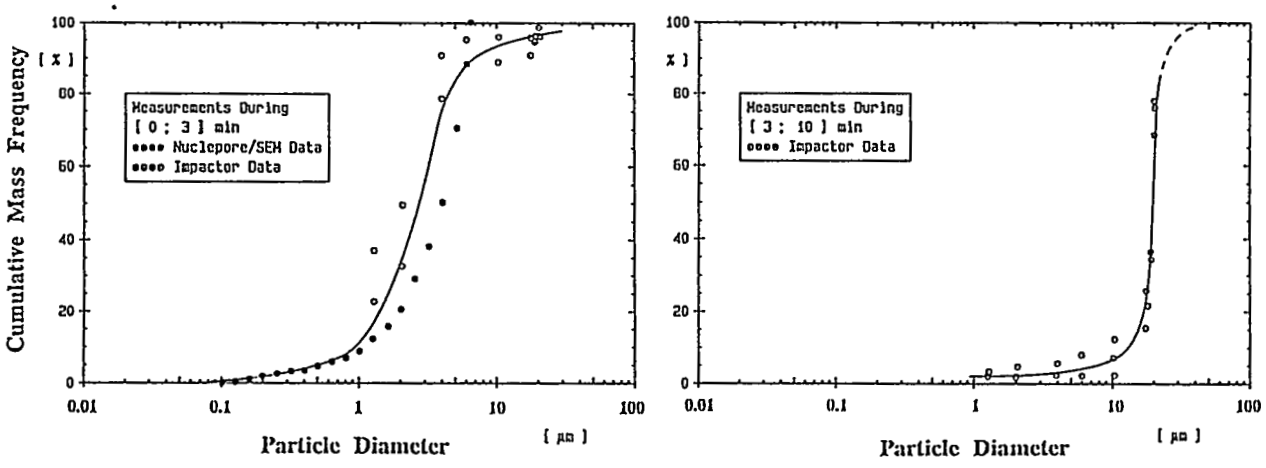

Fig.3: Cumulative mass frequency of particulate in the beginning (left) and during the burnout (right).

III. Hot-Gas Filtration

For applications with large variations in the mass concentration, the particle size distribution, and the composition of the dust in the off-gas, ceramic candle filters are particularly suitable. They offer good performance in collecting fine dust particles. With high off-gas temperatures, deposited soot can burn up on the candles immediately behind the afterburner, without the risk of impairing the filters.

The original concept provided for a two-stage hot-gas filtration in order to attain the highest collection efficiency possible in the candle filters. Both stages showed the same set-up and were equipped with the same candles - 170 elements in each stage. Figure 4 and Table 1 show further details.

Two types of candles were shown to have the necessary temperature resistance and durability, necessary for high-temperature filtration. Both types of candles are SiC ceramics. Type 1 has a homogeneous profile with the characteristics of a deep bed filter (Fig. 5, left). Type 2 consists of two layers and is constructed to be a surface filter. In order to provide for dust collection on the surface, the candle is equipped with a layer of Al_2O_3 fibers approximately 100 µm thick, and with a diameter of 3 µm, on the supporting body out of SiC (Fig. 5, right).

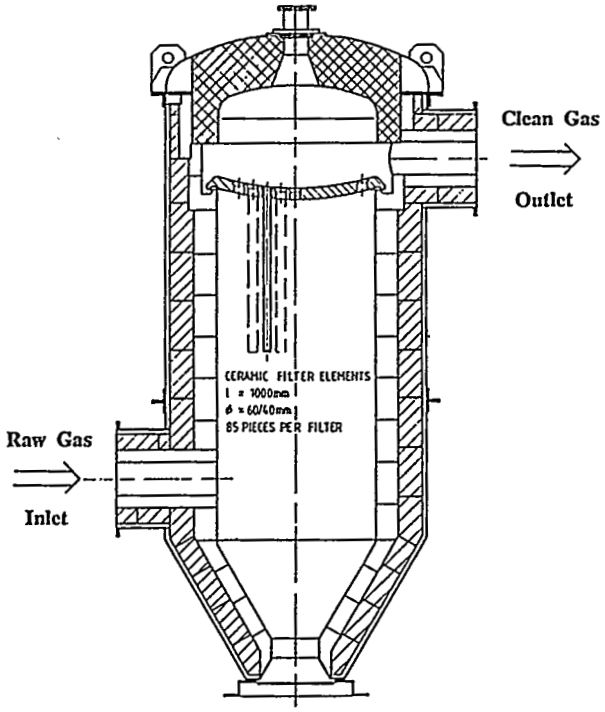

Fig.4: Schematic set-up of the hot-gas filter stage.

The two hot-gas filter stages represent the main components of off-gas cleaning as far as particle removal is concerned. Therefore they play an primary role when optimization is required. Choosing the best type of candle requires a knowledge of the collection efficiency. Since collection efficiency depends above all on the particle size distribution, the shape of the fractional efficiency curve for both types of candles must be known. The influence of temperature and filtration velocity have to be considered as additional parameters.

Table 1: Structure characteristics of grain ceramic and grain/fiber ceramic[5].

		Grain type	Grain/fiber type
Length	(mm)	1000	1000
Outside Diameter	(mm)	60	60
Inside Diameter	(mm)	40	30
Wall Thickness	(mm)	10	15
Grain Diameter	(μm)	120	300 - 500
Fiber Diameter	(μm)	-	3 - 5
Pore Diameter(Matr.)	(μm)	40	100
Pore Diameter(Coat.)	(μm)	-	10 - 20
Filter Area	(m^2)	0.16	0.16
Pressure Drop at 3 cm/s	(Pa)	550	180

Fig.5: SEM photographs of the ceramic candle structures: grain type (left) and grain/fiber type (right).

Therefore, in the existing incineration facility, measurements of the fractional efficiency for both hot-gas filters were carried out in practical operation. These measurements during operation were supplemented by laboratory scale tests. Here temperature and filtration velocity could be better controlled and varied over a wider range than possible under field conditions. To analyze the removal efficiency of the hot-gas filters, an attempt was made to model the particle deposition on both the grain type and grain/fiber type ceramic on the basis of available theory.

For modelling particle removal within the grain layers of both types of candles an approach of [6] was taken as a basis. Here the ceramic is assumed to be a bed of densely packed individual granules in which particle collection follows Eq.(1).

$$T_K(x) = 1 - \exp\left[- 1.5 \frac{1 - \epsilon}{\epsilon} \frac{L}{D_K} E_K \right] \qquad (1)$$

$$E_K = E_{DK} + E_{RK} + E_{IK} \qquad (2)$$

The filtration efficiency E_K, describes the collection on the layer's separate granules by the phenomena of diffusion, interception, and inertia, assuming that the values of the separate effects add up to the total efficiency (Eq. (2)). The grain layer itself is described by the thickness L, the grain diameter D_K, and the average porosity ϵ. Flow conditions are different for the single grain and

corrected when determining E_{DK}, E_{RK}, and E_{IK} by introducing a correction equation depending upon porosity.

For modelling the grain/fiber type ceramic, the assumption was a collection within two layers in series, for which the fractional efficiency is described by Eq (5).

$$T_F(x) = 1 - \exp\left[- \frac{\Delta P \; D_F}{W_F \; \mu \; v} \; E_F \right] \qquad (3)$$

$$E_F = E_{DF} + E_{RF} + E_{DRF} \qquad (4)$$

$$T(x) = T_F(x) + \left[1 - T_F(x) \right] T_K(x) \qquad (5)$$

The fractional efficiency of the layer of fibers, $T_F(x)$, can be determined by Eq (3), according to [7], again starting from conditions of the single collector, i.e., in this case the single fiber. The efficiency is described by the pressure drop of the layer ΔP, the fiber diameter D_F, the drag force W_F, the dyn. viscosity and the velocity of the flow. In contrast to collection on a single grain, an additional combination effect E_{DRF} is considered in Eq. (4).

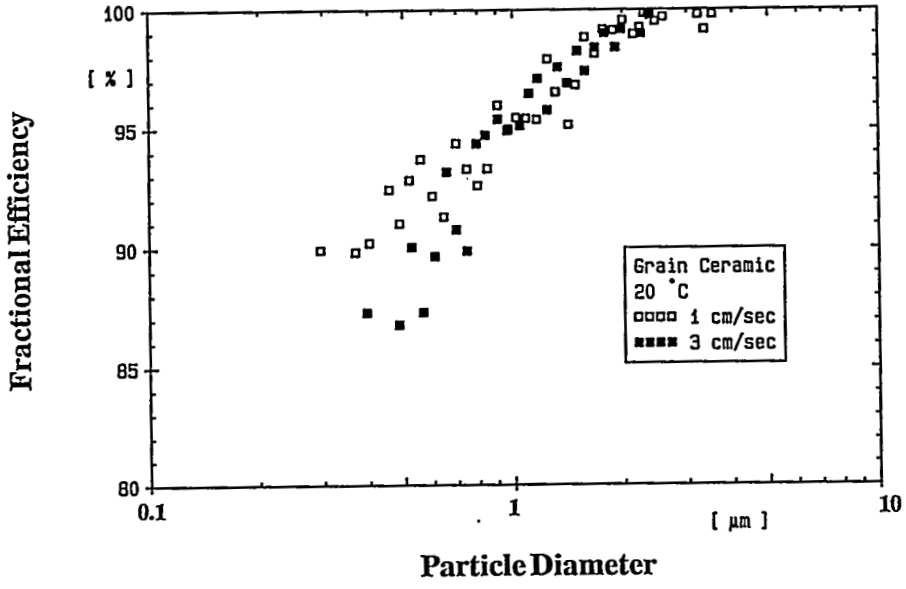

Fig.6: Fractional efficiency of the grain type ceramic at room temperature.

Structure characteristics necessary for applying Eqs. (1) and (3) describing the geometry of the grain layer and the grain/fiber layer, were determined from SEM-photographs.

Figures 6 and 7 show fractional efficiency curves determined experimentally for grain ceramic with a range of particle size from 0.2 to 4 μm. It is seen that the efficiency decreases generally with declining particle size, i.e., collection depends mainly on interception. The inertia phenomenon under the given circumstances, above all the low filtration velocities, does not contribute significantly to collection.

Figure 6 shows this clearly, since the fractional efficiency above 1 μm is independent of velocity. For particles < 1 μm an improved collection is noted with decreasing filtration velocity. This indicates the influence of collection by diffusion effects.

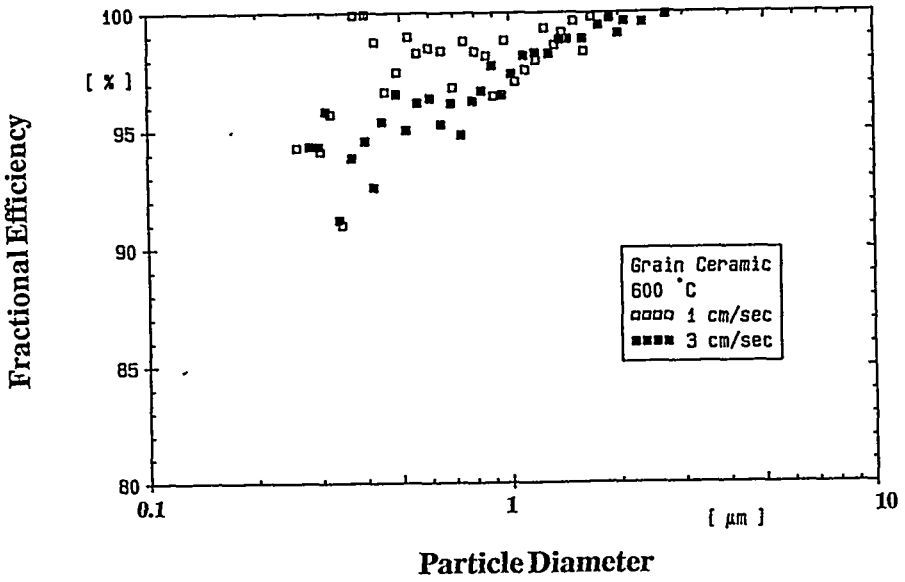

Fig.7: Fractional efficiency of the grain type ceramic at 600°C.

With regard to temperatures of 600°C, the operating temperature of the second hot-gas filter stage, this becomes even clearer. Figure 7 demonstrates that a fractional efficiency of 95% can be obtained even for the particles hardest to collect, while at ambient temperature 87% (3 cm/sec) and 90% (1 cm/sec) were measured. The influence of velocity, which is to be expected at 600°C, is greatly exceeded by the improvement in diffusive collection due to the elevated temperature.

Since the grain/fiber type ceramic seemed to be particularly suited for surface collection of the fine dusts, additional measurements of the fractional efficiency were carried out down to a particle size of 0.015 μm.

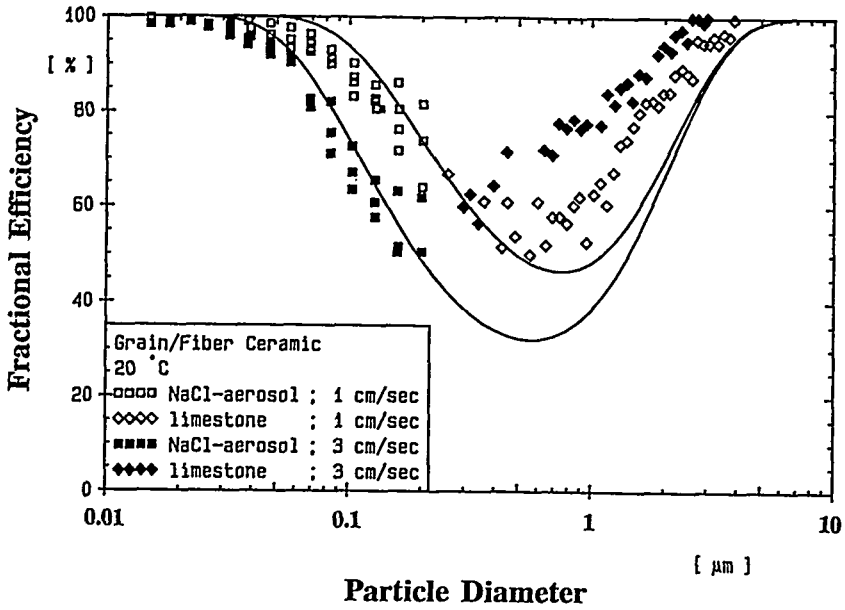

Fig.8: Fractional efficiency of the grain/fiber type ceramic at room temperature: experimental and modelled.

Figure 8 shows fractional efficiency curves for grain/fiber ceramic determined under laboratory conditions with ambient temperature. Remarkable is the fact that this type of candle construction shows a collection minimum in the shape of the fractional efficiency curve far more distinct than is the case with pure grain ceramic. As the diagram shows, increasing filtration velocity from 1 to 3 cm/sec effects a shift parallel to the X-axis. Unlike the conditions with the grain ceramic, increased filtration velocity here leads to an improvement in collection efficiency to the right of the minimum. This suggests that in this case particle collection, by inertia plays an important role in the fiber layer. In the area exclusively controlled by diffusion, which is the part of the fractional efficiency curve to the left of the collection minimum, increased velocity leads to decreased values for the fractional efficiency, as is to be expected. Included in the diagram are curves determined on a theoretical basis. The left branch of the curve can be predicted correctly for both velocities. The right branch, shows a clear underestimate of the measured collection. This shows, that inertia collection was not taken into consideration for the collection in the fiber layer. Accordingly, the deviation of experimental data from theoretically determined values is greater at higher velocities. For the conditions to be expected in operation, 1 cm/sec filtraton velocity and 600 - 800°C, inertia collection in the fiber layer can be neglected. As Fig. 9 shows, the collection performance of grain/fiber type ceramic in operation at 800°C can be modelled with sufficient accuracy.

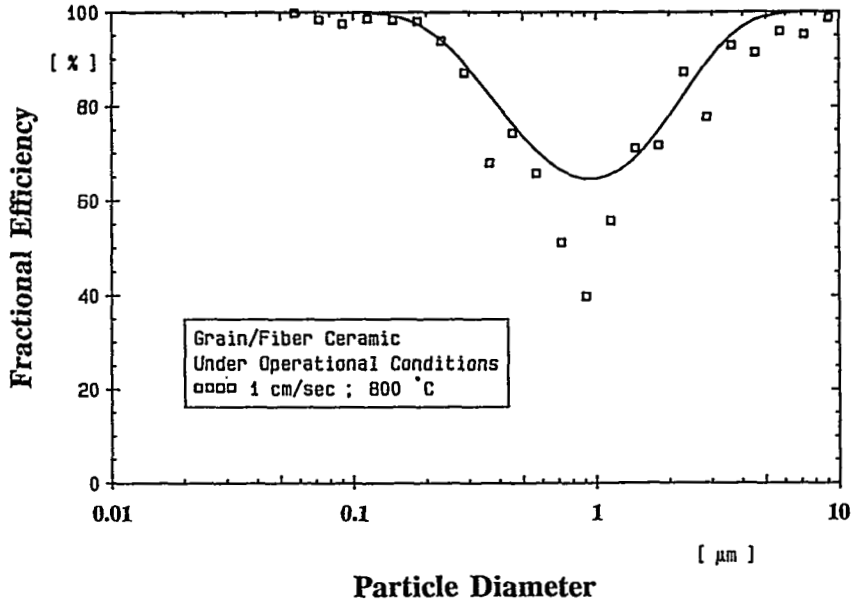

Fig.9: Fractional efficiency of the grain/fiber type ceramic at 800 °C: experimental and modelled.

The theoretical model allows the individual collection efficiencies of the grain and the fiber layer to be modelled. Separate grade efficiency curves for both layers of the grain/fiber ceramic are shown in Fig. 10, together with the resulting overall fractional efficiency curve. On the basis of the particle spectrum in Fig. 2 and by using the separate efficiency curves it is possible to show that coarser particles are predominantly retained in the fiber layer. Submicron particles on the other hand, predominantly penetrate the grain layer. This diagram proves clearly, that the grain/fiber type ceramic offers no advantage in comparison with the grain type ceramic. In both types of candles the difficult to filter fine dusts are predominantly collected in the grain layer.

As proven by extensive measurements and the given separation characteristics, particles transported in the flue-gas are being collected in the first stage with an efficiency of 95 - 98% by weight. A comparably narrow fraction of particles, corresponding to the given range of penetration, passes through the candles of stage 1. Since the fractional efficiency in the second identical filter stage doesn't differ essentially from the first stage, in spite of the temperature difference of 200°C, only a low collection efficiency can result. For the candles of the second stage these dusts are to be considered as particularly critical. They penetrate deeply into the filter layer, are practically not to be cleaned off and therefore irreversibly clog the candles. This explains the relatively short service life of approximately 700 h. Due to the low contribution to overall collection and the short service life, the second hot-gas filter stage has to be considered as uneconomical, no matter which type of candle is employed.

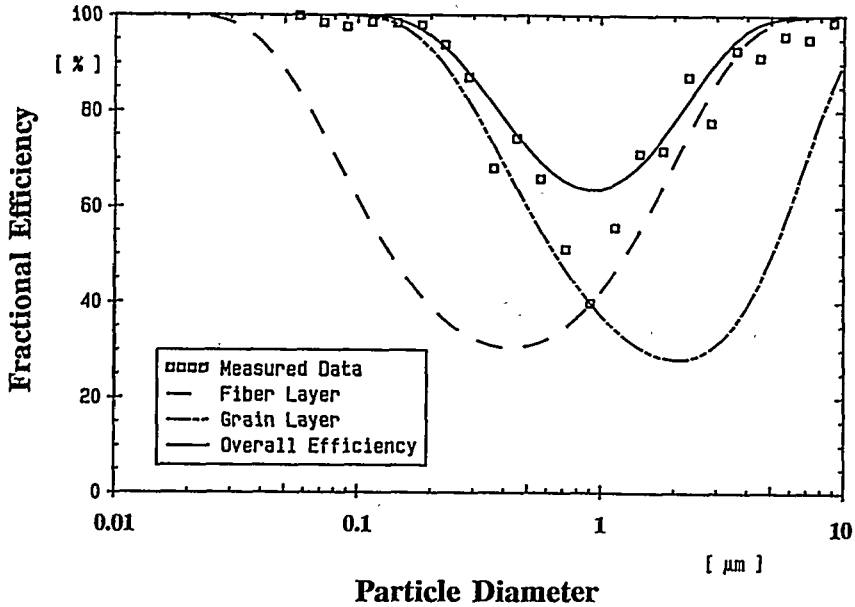

Fig.10: Selective fractional efficiency of the grain and the fiber layer for the grain/fiber type ceramic.

IV. Fine Dust Filtration

Originally, removal of harmful gases and the collection of fine dust had been planned to be combined, in order to relieve the final HEPA filters. Preliminary investigations into the removal characteristics of a scrubber combination for the existing ß-incineration facility, however, led to a change at this stage of the procedure.

For the assessment of the collection performance of jet and venturi scrubbers, which is determined essentially by the question, which dust fractions pass through the preceding hot-gas filter stage, it is suitable to take into consideration the efficiency of the candle filters. If one knows the filtration performance of the HEPA filter, estimates can be made for the expected performance of these filters. Therefore in Fig. 11, the overall fractional efficiency curve of the scrubber combination is shown plotted against the fractional efficiency curves of the hot-gas and HEPA filter stages. The diagram ordinate was chosen to be logarithmic in order to be able to display the widely differing efficiency of the particle filters used. The fractional efficiency curve for the scrubber combination represents (for particles < 1 μm) the range of penetration of the venturi scrubber, as the preceding jet scrubber for sub-micron particles shows practically no effect. It is clearly seen that the fractional efficiency curves of scrubber and hot-gas filter overlap in a wide range of high penetration. The result is, that the greater portion of the fine particles still to be collected after the hot-gas filter,

unhindered. Measurements demonstrated the venturi scrubber's low efficiency.

After off-gas washing a maximum dust content of between 50 and 100 mg/m^3 must be expected. This would have meant a disproportionately high stress on the HEPA filter. Due to its low dust loading capacity for captured particles, with offgas dusts approx. 1 kg per filter element, only an uneconomically short service life can be obtained. As to be seen in Fig. 11, class S HEPA filters show only an extremely narrow range of penetration around 0.1 µm, collection efficiency even here being higher by three orders of magnitude than with the candle filters and the scrubbers. Thereby, an extremely efficient filtration of fine dusts is guaranteed.

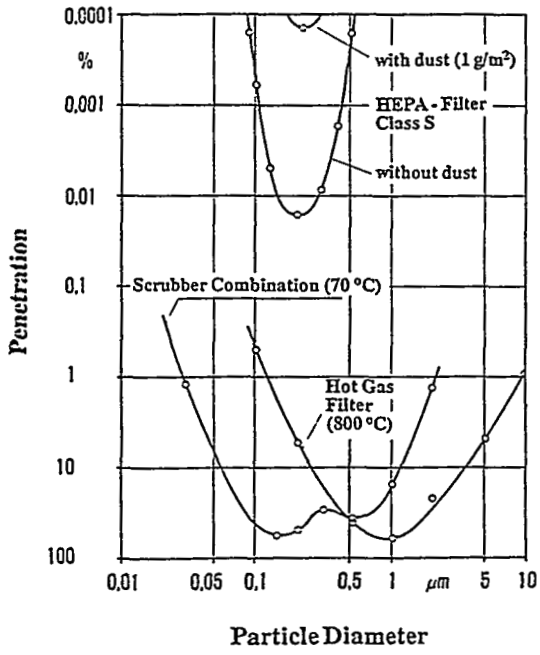

Fig.11: Penetration of the dust collectors in the off-gas cleaning process.

As laboratory measurements on dust-loaded filter media have shown, penetration decreases even more when low dust loadings are already present. For example, a curve for the comparably small loading of 1 g/m^2 is included in the diagramm. It has been possible to show, that EU8 particulate air filter with an initial penetration 300 times higher than that of class S filters can be loaded with 3 - 4 kg of off-gas dusts before reaching the allowable maximum pressure drop. It applies to these filters as well, that the collection efficiency improves with an increasing dust loading. If one replaces EU4 filters for coarse dust particles by filters of type EU8, the HEPA filters can be effectively relieved. The practical evaluation of this filter combination in the incineration facility for liquid waste at KfK with extremely fine dusts in the sub-micron range, produced the expected prolongation of the service life for the HEPA filter stage as a whole. By this measure, fine dust filtration could be

realized much more economically. For increased operational safety of both filter stages, the filter elements of increased structural strength developed at KfK are being used[8].

V. Optimized Concept for Off-gas Cleaning

On the basis of the above results combustion control and off-gas cleaning procedures were improved under the aspect of minimizing dust emissions and operating costs. The optimized concept for the procedure in the α-incineration facility is shown schematically in Fig. 12. By way of a better distribution of air and the creation of strong turbulence in the furnace, formation of soot particles could be minimized. With an unchanged first hot-gas filter stage the second stage could be dispensed with, without a notable loss in collection efficiency. The employment of a second jet scrubber, instead of the venturi scrubber originally planned, saves investment costs and above all, costs of operation. The collection of fine dusts is being carried out with increased efficiency and improved economy using EU8 filters and HEPA filter units. Via large scale investigations into particulate formation and particle separation in operating facilities, the basis for a technically and economically optimized facility could be achieved. The results of this research provide a contribution toward a better ecological compatibility for nuclear installations.

Furthermore, because of the extraordinarily high level of overall collection of the problematic fine dusts, the solutions created here are also of interest for the incineration of conventional hazardous waste.

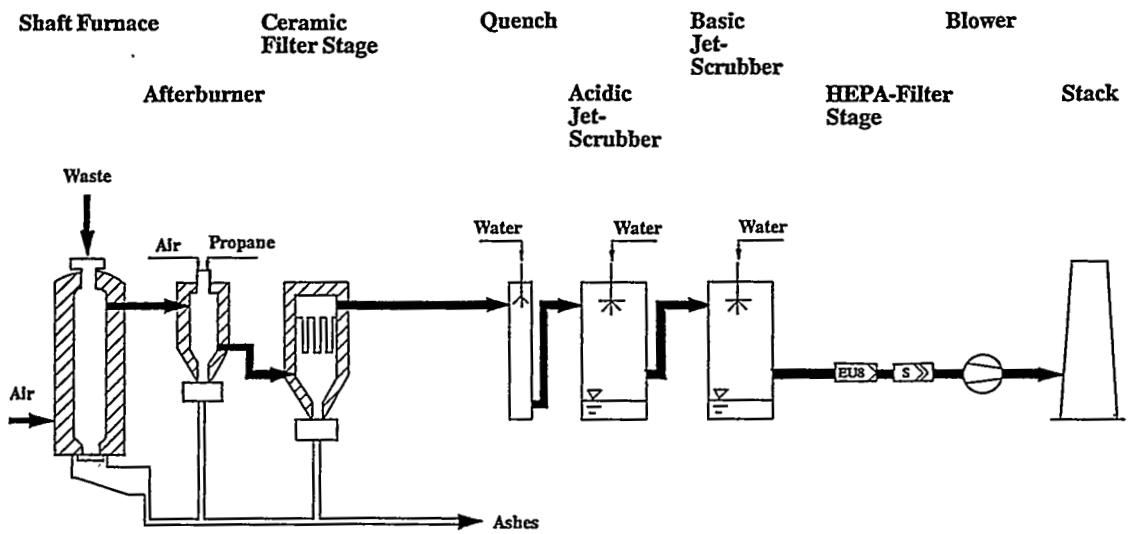

Fig.12: Flow diagram of the α-incineration facility with an optimized off-gas cleaning process.

V. References

(1) Koenig, R.A. and Vavruska, J.S.; "Incinerators used for radwaste"; Proceedings of Incineration of Low Level & Mixed Wastes; Charlotte NC, USA; (1986).

(2) 1. Allg. Verwaltungsvorschrift zum Bundesimmissionsschutzgesetz (Technische Anleitung zur Reinhaltung der Luft); Bundesanzeiger Verlagsgesellschaft; Köln, FRG; 1986.

(3) DIN 24185; Method of testing air filters used in general ventilation; Beuth Verlag; Berlin, FRG; 1980.

(4) DIN 24184; Type test of high efficiency submicron particulate air filter; Beuth Verlag; Berlin, FRG; 1974.

(5) Leibold, H; Dirks, F; Rüdinger, V.; "Particulate emissions from a LLW-incinerator and off-gas cleaning with a new type of ceramic candle filter"; Nucl. and Chem. Waste Mgt.: Vol 9; p. 87 - 94; (1989).

(6) Tardos, G. and Gutfinger, G.; "Theoretical and experimental investigations on granular bed filters"; Atm. Envrm.: Vol. 13; p. 853 ff.; (1979).

(7) Kirsch, A.A. and Stechkina, J.B.; "The theory of aerosol filtration with fibrous filters" in "Fundamentals of aerosol science"; John Wiley & Sons; New York; 1978.

(8) Rüdinger, V., Ricketts, C.I. and Wilhelm, J.G.; "High strength HEPA filters for nuclear applications"; Nuclear Technology: to be published in Oct. 1990.

DISCUSSION

BERGMAN: How was your experimental data generated? Would you comment on some of the procedures you used to develop the efficiency curves?

LEIBOLD: We took samples on the upstream and downstream sides of the hot gas filter, cool them down to 160° C, and analyze them. We used Nuclepore filters for sampling.

WEBER: Have you had a chance to evaluate a finer grade of ceramic candle as a second stage to see if the potential of particles bridging the pores makes this stage practical?

LEIBOLD: I don't think that it would be of great use because the pressure drop would increase very much if we were to use very small grains in the candles. The problem is the penetration. The penetration range of these candles is not the problem. I don't think that the minimum range will change or will shift to a greater particle size.

HANDYSIDE: I had the privilege of visiting your facility some years ago at the early stages of its operation. I recall at that time the radioactive content of the waste that you brought through it was very, very low indeed. There was a problem at that time with the service life of the ceramic filters. Can you tell me what is the maximum alpha content of the waste that you now process through the facility? What do you think would be the service life of the first candle filter in your optimum system?

LEIBOLD: I can only speak about the service life of the candles. Up to now, the filter station has operated about 1,500 hours without significant increase in pressure drop.

HANDYSIDE: That is significantly better than in the early days.

STEVENS: Pall Corporation has provided hundreds of self cleaning-metal off gas filters over the years. Parameters do exist to mitigate irreversible pluggage. The selection of the approximate pore structure and optimized face velocity is a (f) of the particle size distribution of the solids. In general it is better to perform the filtration in a single stage; increasing its size to achieve face velocity and ΔP limits.

DESIGN BASIS DOCUMENTATION FOR THE CONTROL ROOM VENTILATION/CHILLED WATER SYSTEM AT MCGUIRE NUCLEAR STATION

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Abstract

The design bases of the Control Room Ventilation/Chilled Water System at McGuire Nuclear Station have been captured in a more usable and permanent form. The bases for control room pressurization, control room temperature, chlorine detection, and tornado missile protection, were researched and documented.

1.0 BACKGROUND

The code of federal regulations defines design bases as "that information which identifies the specific functions to be performed by a structure, system, or component of a facility, and the specific values or ranges of values chosen for controlling parameters as reference bounds for design." For Duke's nuclear plants, this information currently resides in numerous locations (FSAR, Technical Specifications, licensing correspondence files, SER, design calculations, etc.). The timely retrieval of such information has been difficult. The level of detail of this information also varies considerably. These factors have contributed to or caused delays in responding to operational concerns at Duke's nuclear plants and have contributed to or caused several significant licensing issues in recent years. One of the main issues that prompted the effort to document design bases was a misinterpretation of the relationship between two safety-related systems that controlled hydrogen concentration inside the primary containment after an accident. One of the systems involved was a ventilation system that is designed to mix the containment atmosphere and prevent hydrogen buildup in isolated pockets of containment. In addition, it was discovered that the two trains of the ventilation system were not balanced. A decision was made to capture the design bases of this and other safety-related systems in a more usable and permanent form.

In early 1989, a steering committee was formed to define the content, format, scope, and schedule of the Design Basis Documentation (DBD) effort. Three pilot documents were developed. Subsequent to this, the DBD project was approved and is in the second year of a six year schedule. System design bases, equipment design bases, and system descriptions are included in each DBD. The safety-related system and equipment design bases (the required functions) and the system description are in separate sections.

The first DBD developed outside the pilot effort for McGuire Nuclear Station was for a nuclear air cleaning and conditioning system, i.e., the control room ventilation/chilled water system. This system has received a lot of regulatory attention recently and a self-initiated audit has been completed. Early completion of this DBD helped address some concerns and assisted with the audit. Several interesting issues, addressed during the development of the DBD for this system are described in the following section.

2.0 DESIGN BASES

Issues related to the design bases for control room pressurization, control room temperature, chlorine detection, and tornado missile protection were addressed during the development of this DBD. In addition, an audit of the control room ventilation/chilled water (VC/YC) system was conducted following the release of this DBD. Audit findings are also summarized in this section.

The VC/YC system at McGuire has two trains of equipment and two intakes at each of two locations (four total). The control room must be pressurized to 1/8 inch water gauge with each train taking suction off of two out of the four intakes. The capability to isolate two intakes following the receipt of a radiation signal was confirmed to be a licensing commitment. The outside air intakes are not designed to operate under conditions where two intakes have been isolated on a high radiation signal and a concurrent single failure is assumed.

Also, a new reference location for the pressure test was established. This concern was identified outside of the DBD effort. The reference pressure location was moved from an adjacent room to an area which is at atmospheric pressure. The reference pressure location used originally was at a slightly negative pressure with respect to the outside atmosphere. Therefore, when the location was moved to the outside atmosphere, it became more difficult to pressurize the control room to 1/8 inch water gauge. In order to meet the control room pressure requirement, all four intakes were required to remain open. An operator will open the intakes if they close on a radiation signal. These compensatory measures allow for continued operation until additional leaks in the control room are located and sealed.

McGuire's Technical Specification for control room temperature is currently 120°F. During the development of the VC/YC DBD, it was discovered that there were documented failures of safety-related electronic equipment in the control room at general area temperatures of 90°F. Temperatures in the control area were administratively controlled to 90°F. A revision to the Technical Specifications is currently being pursued.

Chlorine gas protection criteria was established in the Standard Review Plan. The control room operators at McGuire were

previously evaluated for chlorine gas exposure. The height of the outside air intakes at McGuire precludes the intake of chlorine into the control room. Therefore, the chlorine gas detectors are not addressed in the Technical Specifications and are not safety-related. This position was documented in the VC/YC DBD.

One final design basis issue that was addressed during the development of the VC/YC DBD concerned the protection of the outside air intakes from tornado-generated missiles. Under the original design, the outside air intakes are protected from tornado missiles by separation distance and surrounding structures. However, with the compensatory measures taken for the reference pressure concern, i.e., all four intakes are required to be open, there was concern that the intakes may no longer be adequately protected. An investigation determined that the probability of there being a tornado strike on a pair of intakes before or after an event that released radiation, and thus required the operation of the VC/YC system, was extremely low. Therefore, the current design was deemed acceptable.

The VC/YC system was audited shortly after completion of the DBD by a team of Duke engineers made up of representatives from the QA, Design Engineering and Nuclear Production Departments. The DBD was a useful tool for the audit.

3.0 CONCLUSIONS

The design bases documented for the VC/YC system will be helpful in resolving future operability issues, used as design input for plant modifications, training, and as a tool in audits. Initial feedback from station personnel has been positive.

A DBD for the annulus (secondary containment) ventilation system has also been published and another nuclear air cleaning system DBD may be issued in 1991. The DBD effort is scheduled for completion in 1995.

DISCUSSION

RABON: With respect to the analysis for chlorine releases, how acceptable to the regulators was resolving the potential for Cl_2 exposure to the central control room with defending the height of the intake ducts?

HILLEY: The analysis was done many years ago by a fellow named Howard Smith. He did an effluent analysis by looking at the pressure difference between the chlorine canisters and the outside air and provided that to the NRC. At the time, technical specifications were being developed and there was some question whether detectors would appear in the technical specifications. We sent them a letter with the analysis, documented it in FSAR and they approved it. They aren't safety related, but they are regulatory required.

WOOD: When you analyzed the ΔP in the control room with respect to the outside area, did you also look at adjacent areas or just the outside area?

HILLEY: The original reference point on the pressure was an adjacent room. We found it to be slightly negative with respect to the outside atmosphere. Then, we decided it would be more conservative to maintain a positive pressure with respect to the outside area.

DEMONSTRATION OF CARBON-14 REMOVAL AT CANDU NUCLEAR GENERATING STATIONS

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Canada M8Z 5S4

Abstract

In CANDU nuclear generating stations, nitrogen, carbon dioxide and D_2O are used in various applications where they are exposed to thermal neutron irradiation. Under these conditions, nitrogen-14, oxygen-17, and carbon-13 are activated to carbon-14. The carbon-14, so produced, may be in the form of gaseous carbon-14 dioxide, aqueous solution, usually as carbonate or bicarbonate, or as a solid deposit. While the emission of carbon-14 has never exceeded the Derived Emission Limit (DEL) at CANDU NGS, it is, nevertheless, desirable to restrict its uncontrolled release, if possible, because of its long half-life.

Our development of a gaseous carbon-14 removal process, which uses a calcium hydroxide bed at ambient temperature, was reported at the 17th and 18th Nuclear Airborne Waste Management and Air Cleaning Conferences. The process is simple, reliable and safe to operate and the exhausted bed is stable and suitable for disposal. A demonstration unit, using this process, was installed at the NPD NGS in 1987 to remove carbon-14 dioxide from a side stream of the helium moderator cover gas. This unit was subsequently modified and installed at the Pickering NGS in 1988 to remove carbon-14 from the purge stream from the nitrogen-filled annulus gas system. This paper summarizes the results of these full-scale applications at CANDU stations.

At NPD NGS, no carbon dioxide was detected downstream of the removal system with an entering concentration of 90 vppm. At Pickering also, the gasified carbon-14 deposit was quantitatively retained on the calcium hydroxide bed.

Introduction

In CANDU nuclear generating stations, nitrogen, and carbon dioxide, and D_2O are used in various applications where they are exposed to thermal neutron irradiation. Under these conditions, nitrogen-14, oxygen-17, and carbon-13 are activated to carbon-14. The carbon-14, so produced, may be in the form of gaseous carbon-14 dioxide, aqueous solution, usually as carbonate or bicarbonate, or as a solid deposit. While the emission of carbon-14 has never exceeded the Derived Emission Limit (DEL) at CANDU NGS, it is, nevertheless, desirable to restrict its uncontrolled release, if possible, because of its long half life, 5730 years.

The DELs, calculated by considering several factors such as the chemical and physical forms of carbon-14, environmental variables, and behavior in the human body, were reviewed and approved by the Atomic Energy Control Board of Canada. These approved DELs are summarized in Table 1.

Table 1
C-14 Derived Emission Limits for
Ontario Hydro Nuclear Stations (1)

	Derived Emission Limit (DEL)	
Station	Airborne (Ci/7 days)	Waterborne (Ci/mo)
Pickering NGS A	1.4×10^3	3.5×10^2
Pickering NGS B	1.4×10^3	3.5×10^2
Bruce NGS A	1.1×10^3	2.9×10^2
Bruce NGS B	1.1×10^3	3.1×10^2
Darlington NGS*	6.8×10^2*	2.2×10^3*

* Provisional

Carbon-14 is produced in CANDU reactors in moderator systems, nitrogen-filled annulus gas systems (AGS), and, to a minor extent, in the fuel and coolant. The production rates depend on the properties of the target atoms as shown in Table 2 and the neutron flux. The carbon-14 production rates are shown in Table 3.

Table 2
Properties of Target Atoms

ISOTOPE / PROPERTIES	^{14}N	^{17}O	^{13}C
Natural Abundance	0.9963	0.00037	0.0111
Reaction	$^{14}N(n,p)^{14}C$	$^{17}O(n,\alpha)^{14}C$	$^{13}C(n,\gamma)^{14}C$
Thermal Neutron Cross Section (Barnes)	1.81	0.235	0.0009
Sources	Annulus gas-N_2 air, CO_2 impurity Fuel-pellet and filling gas	Annulus gas-CO_2, air Moderator and primary coolant D_2O Fuel-UO_2, pellet gas; filling gas	Annulus gas-CO_2 Fuel-pellet gas CO and CO_2 graphite coating

Table 3
Calculated C-14 Production Rates in Ci/Annum (2)

	Pickering A	Bruce A or B, Darlington
Moderator D_2O	1400	2300
Annulus Gas	920 (460*)	2
Fuel	32	58
Heat Transport D_2O	27	34
TOTAL (Rounded)	2380 (1920*)	2390

* Adjusted production rate assuming conversion of Units 1 and 2 to carbon dioxide from nitrogen.

To comply with the DELs, Ontario Hydro established a stack monitoring program at all operating nuclear generating stations. Table 4 summarizes the carbon-14 gaseous emissions performance of a typical year 1980-1981. We can see from this table that the releases are normally a small fraction of the production rates and, generally, less than one percent of DEL. The NPD NGS, having an air-filled annulus gas system,had a high carbon-14 production rate, but is now shut down permanently. The nitrogen-filled annulus gas systems of Pickering NGSA Units 1 and 2, have been converted to carbon dioxide. This has lowered carbon-14 emissions from the station. Unit 3 is currently shut down for retubing and will be converted to carbon dioxide before restart. Unit 4 is the only unit still operating with a nitrogen-filled annulus gas system.

Table 4
Carbon-14 Gaseous Emissions Performance
1980 - 1981 (2)

STATION	AVERAGE C-14 RELEASE RATE (Ci/wk)	AVERAGE % $^{14}CO_2$	AVERAGE $^{14}CO_2$ RELEASE RATE (Ci/wk)	RELEASE EXPRESSED AS % DEL	RELEASE EXPRESSED AS % PRODUCTION RATE
Pickering NGS A	16.4	80	13.0	0.93	36
Bruce NGS A	1.3	78	1.0	0.09	3
NPD	0.27	98	0.26	0.007	60

One method for trapping carbon-14 dioxide is to use a calcium hydroxide bed. A development program was initially proposed in 1980 and the pilot plant study was approved in 1981. The results of the pilot plant study concluded that carbon dioxide can be absorbed effectively by calcium hydroxide at ambient temperatures if a high relative humidity can be maintained. These results were published in 1982 and 1984(3,4) and US and Canadian patents were granted in 1985(5).

The process is simple, reliable, and safe to operate and the exhausted bed is stable and suitable for disposal. A demonstration unit, using this process, was installed at the NPD NGS in 1987, to remove carbon-14 dioxide from a side stream of the helium moderator cover gas. This unit was subsequently modified and installed at the Pickering NGS in 1988 to remove carbon-14 from the purge stream from the nitrogen-filled annulus gas system. This paper summarizes the results of these full-scale applications at CANDU stations.

Demonstration on the NPD NGS Moderator Cover Gas System

After the activation of nitrogen-14 in the annulus gas system, the activation of oxygen-17 in the moderator heavy water is the major carbon-14 emission source in CANDUs. The carbon-14 produced in the moderator, partitions between the heavy water and the helium cover gas. At steady state, it is present mainly as dissolved carbonate in the liquid phase and as carbon dioxide in the gas phase. Carbon-14 in the liquid is removed in the moderator purification system ion exchange columns. The carbon-14 dioxide in the gas phase may be redissolved in the liquid or be released to the atmosphere via leaks or purges.

As part of Ontario Hydro's Carbon-14 Management Project, 1984, a recommendation was made to install a carbon-14 removal system at Pickering NGSA on the moderator cover gas system. As an alternative, a proposal was made to test a full scale prototype carbon-14 scrubber system at NPD NGS in 1985. The latter proposal was accepted. Design and procurement were undertaken almost simultaneously. The unit was shipped to NPD on April 2, 1986. AECB approval of the design package and for the installation was formally granted in June, 1986. During testing, some deficiencies were identified and corrected. The unit was finally installed in early 1987, and commissioned in March of that year. The unit operated from April 27 to May 22 for a total of 612 hours. NPD was shut down on May 24, 1987 and the decision was made not to restart, thus the experiment was interrupted.

Demonstration Program

The original program objectives at NPD were to demonstrate: (1) the technical viability of the carbon dioxide removal method in an actual industrial environment, (2) the carbon-14 dioxide retaining capability of the calcium hydroxide bed, (3) the effect on the station carbon-14 emissions, and (4) the reliability of the prototype design for use in other locations.

With these objectives, experiments were planned:

1. To route a side stream of the moderator cover gas to the carbon-14 removal unit and to monitor the changes of the carbon dioxide level of the cover gas with time.

2. To relocate periodically the carbon dioxide monitor downstream of the absorber to confirm that most of the carbon dioxide passing through the unit is retained by the absorber.

3. To withdraw cover gas samples periodically to evaluate the effectiveness of the absorber to reduce the concentration of carbon dioxide in the cover gas.

4. To retrieve the used absorber bed after six months of operation to determine total carbon dioxide retention, and the carbon-12/carbon-14 ratio.

5. To condition the used absorber bed for storage or disposal.

Of five items above only the first item was carried out, for a total of 26 days. The results indicated that the system operated as designed. The carbon dioxide content in the moderator cover gas decreased with time from 95 vppm but levelled off quickly to 23 vppm. This trend was expected, since the ingress of carbon dioxide from air under subatmospheric pressure operation was known to be substantial at NPD.

NPD Moderator Cover Gas System

The NPD moderator cover gas system together with the carbon-14 removal unit can be simplified as shown in Figure 1.

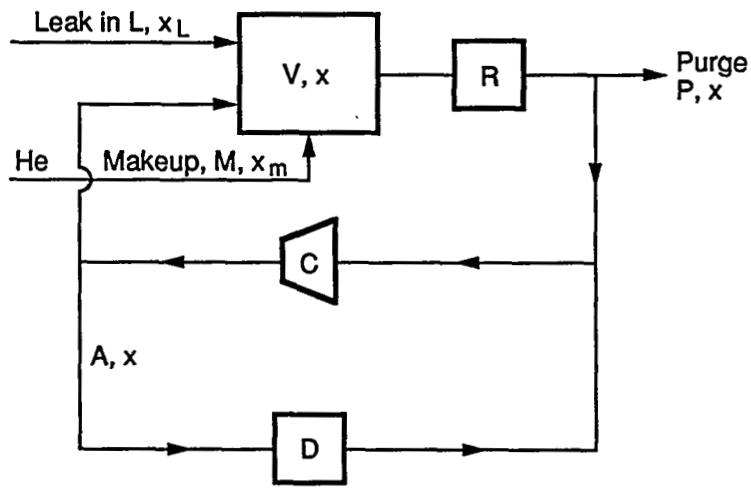

Figure 1: The Simplified Flow Diagram, NPD Moderator Cover Gas System

In Figure 1, V is the volume of the moderator, R is the recombiner, C is the cover gas recirculation compressor, A is the flow rate of the side stream through, D, the carbon-14 removal unit, M is the helium makeup rate with its carbon dioxide concentration x_m and L is the rate of the air ingress with a carbon dioxide concentration of x_L.

From the overall material and the carbon dioxide component balances:

$$L + M - P = 0 \tag{1}$$

and $$L x_L + M x_M - V \, dx/dt - P x - A x = 0 \tag{2}$$

the following equation can be obtained:

$$-V \frac{dx}{dt} - (L + M + A) x + L x_L + M x_M = 0 \tag{3}$$

With the boundary condition of

$$t \dashrightarrow 0, \; x \dashrightarrow x_0$$

The above equation can be solved and expressed as

$$x = \frac{L x_L + M x_M}{L + M + A} + \left(x_o - \frac{(L x_L + M x_M)}{L + M + A} \right) \exp - \frac{(L + M + A)}{V} t \tag{4}$$

From this equation we can see that as $t \dashrightarrow \infty$

$$x = (L x_L + M x_M)/(L + M + A) \tag{5}$$

After 612 hours of operation, the CO_2 levelled off at 23 vppm. From equation 5 with known values: $x_L = 200$ vppm, $M = 0.175$ L/s, $x_M = 5.1$ vppm, $A = 2.35$ L/s, the air ingress rate L is calculated to be 0.323 L/s.

The amount of CO_2 absorbed during the period t can be expressed,

$$CO_2 = \int_o^t A \, x dt \tag{6}$$

where x is function of time and represented by equation 4.

$$\therefore \int_o^t A \, x dt = ANt + \frac{A (x_0 - N) V}{L + M + A} \left(\exp \left(- \frac{L + M + A}{V} t \right) - 1 \right) \tag{7}$$

where $N = (L x_L + M x_M)/(L + M + A)$

For $V = 75 \times 10^3$ L
$t = 612$ h $= 2.2032 \times 10^6$ s
$x_o = 95$ vppm

$$CO_2 = \int_o^t A\, x dt = 234 \times 10^6 \mu g = 234\ g. \tag{8}$$

Demonstration on Annulus Gas Systems

The primary purpose of the annulus gas system in a CANDU NGS is to reduce heat losses from the hot fuel channels to the relatively cool calandria tubes and surrounding moderator. The annulus gas system is also very important for detecting leaks in the pressure and calandria tubes. This is done by monitoring the dew point of the annulus gas.

A simplified flow diagram of the Pickering Unit 3 and 4 annulus gas systems is shown in Figure 2. Dry nitrogen, normally at a dew point of <-40°C, flows through the annuli between the fuel channels and the calandria tubes, via inlet and outlet headers. The hot nitrogen that emerges from the reactor is cooled with an air-to-nitrogen heat exchanger, HX1, for passage through the concrete reactor vault wall. The dew point of the nitrogen is monitored before it is recirculated by metal-bellows compressors to the reactor. Should the dew point exceed -18°C, an alarm is raised in the Control Room.

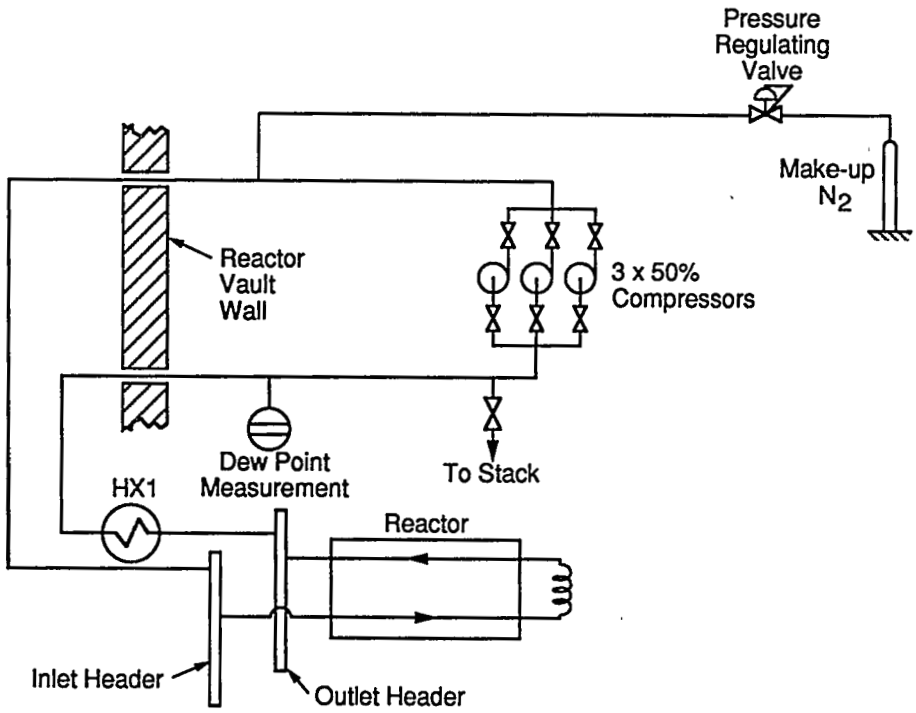

Figure 2: Simplified Annulus Gas System

During replacement of the pressure tubes of Pickering NGS-A Units 1 and 2, carbon-14 contaminated dust was found in the annulus gas system. Elaborate work procedures had to be followed by retubing workers to avoid personal contamination. This affected the retubing schedule. A study was therefore carried out to find a way to decontaminate the annulus gas sytems of Units 3 and 4 prior to retubing.

It was learned that, in a nitrogen filled annulus gas system, carbon-14 forms hydrogen cyanide (HCN), which then polymerizes and crystallizes out on the annulus gas system surfaces. Even at a high temperature the carbon-14 species decomposes very slowly. The decomposition products are H_2O, NH_3, and CO_2 at temperatures up to 300°C. The decomposition of the polymer material is normally incomplete. Pyrolysis, however, is enhanced when a source of oxygen is provided (6). Consequently, it was proposed that oxygen be added to the annulus gas to oxidize the carbon-14 which could then be purged from the system.

At the time of this study, the combined emissions from units 1, 3 and 4 were in the order of 9 curies of carbon-14 dioxide/7-day period. The 1% DEL, the station operating target, is 14 curies/7-day period. This left little room to increase emissions through intentional oxidation and purging. This difficulty can be overcome if the purge lines were equipped with a carbon-14 removal system.

The removal unit built for the NPD cover gas system was adapted for use on Unit 3 at Pickering with the following modifications:

a) Installation of a humidifier upstream of the scrubber bed.

b) Installation of a cryogenic NO_x removal system. When oxygen is introduced into a nitrogen annulus gas circuit nitrogen oxides are generated. The NO_x would compete with carbon dioxide for the calcium hydroxide in the absorber bed.

c) A larger calcium hydroxide bed was required for continuous decontamination of an annulus gas system.

Operation and Results of the C-14 Removal Unit at Pickering NGS

The modified NPD carbon-14 removal unit was installed in the purge stream of the Unit 3 annulus gas system. A simplified schematic flow diagram is shown in Figure 3.

Oxygen was added to the nitrogen feed and the purge was routed through the carbon-14 removal unit. The C-14 activities in the purge stream upstream and downstream of the C-14 removal unit were monitored. The results provided by Pickering NGS(7) are presented in Figures 4 to 8.

Figure 4 shows the history of oxygen addition to annulus gas system. Figure 5 shows the concentration of N_2O presence on the annulus gas system. This correlates well with the oxygen concentration.

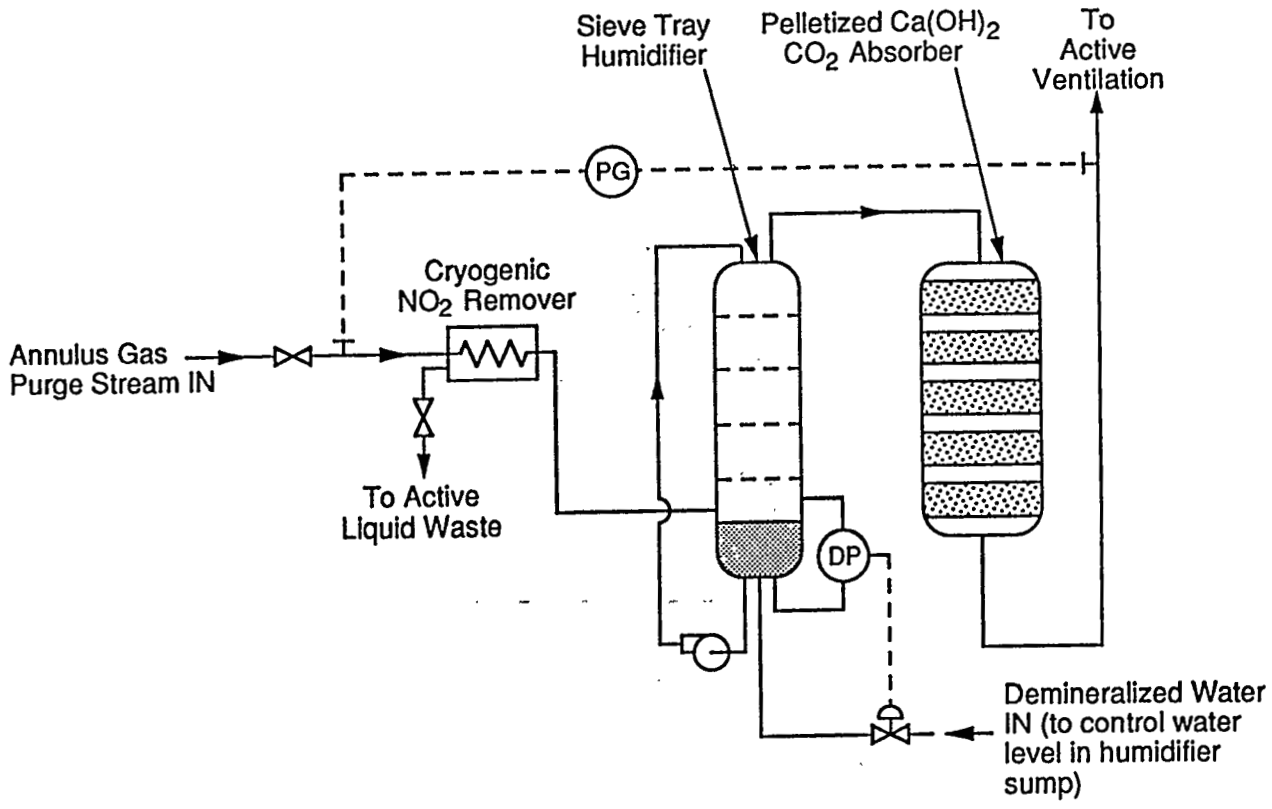

Figure 3: Annulus Gas Purge Stream Carbon-14 Removal System

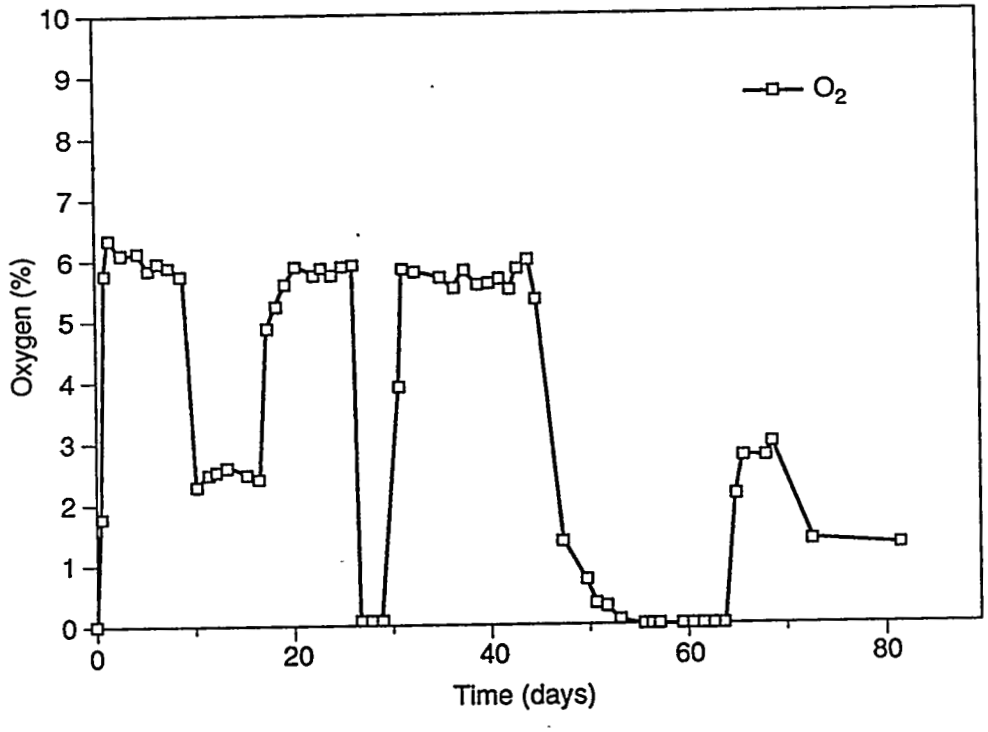

Figure 4: Oxygen Concentration in the PNGS U-3 AGS During C-14 Decontamination by Oxygen Addition

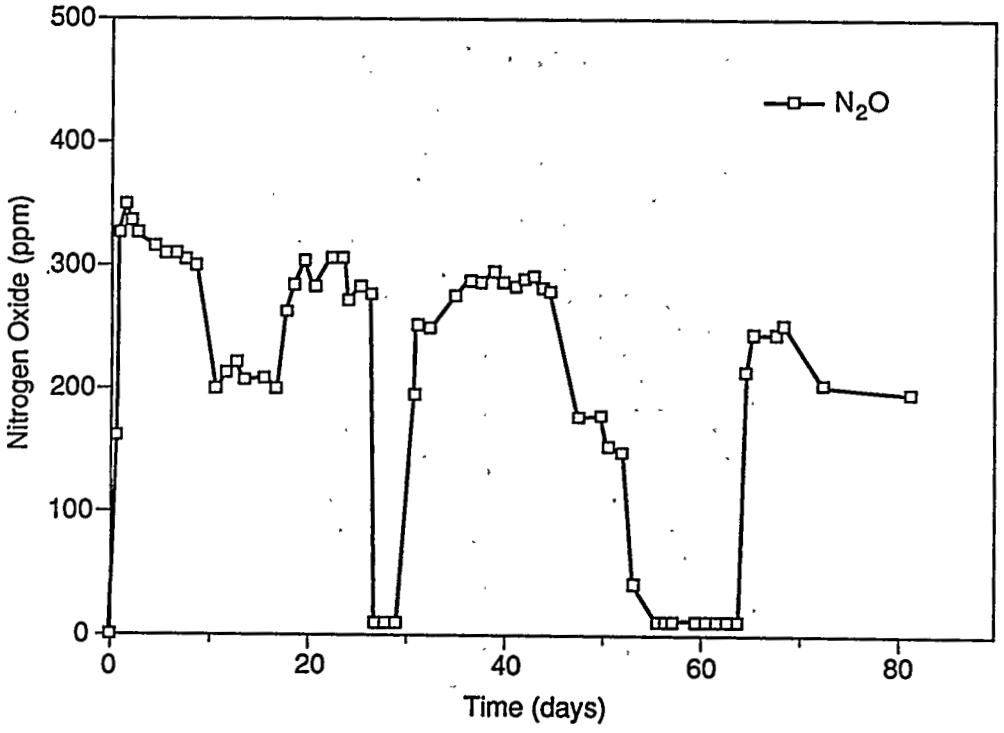

Figure 5: N_2O Concentration in the PNGS U-3 AGS During C-14 Decontamination by Oxygen Addition

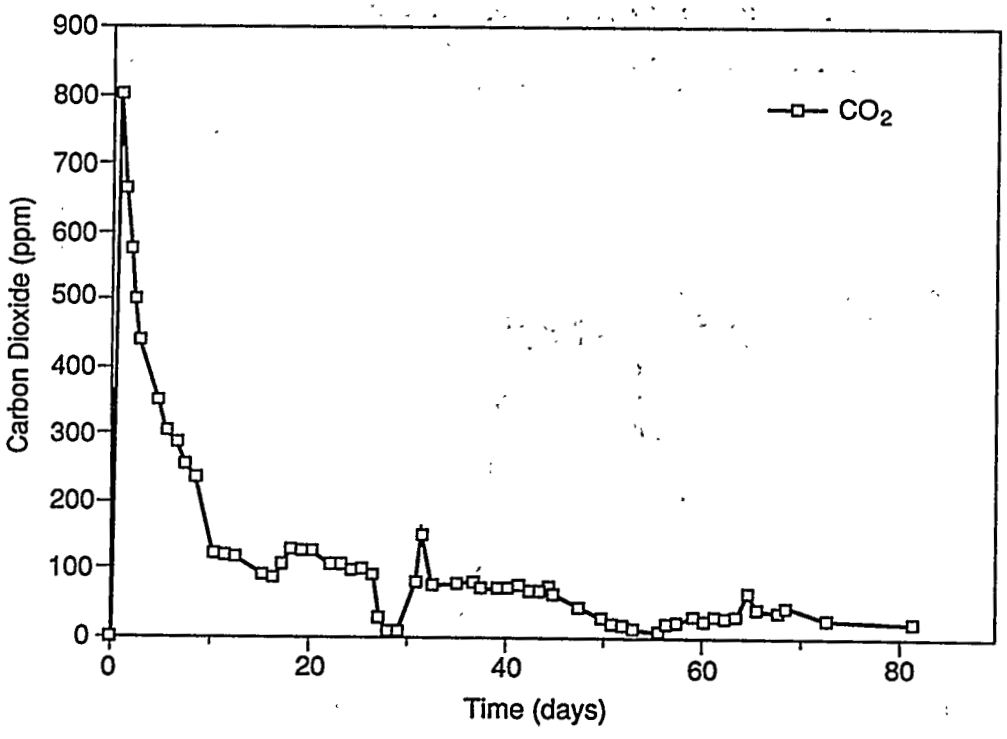

Figure 6: CO_2 Concentration in the PNGS U-3 AGS During C-14 Decontamination by Oxygen Addition

Figures 6 and 7 show that the total carbon dioxide and the activities of the carbon-14 dioxide gas decreased with time. The cumulative removal of carbon-14 is shown in Figure 8. In the first few days the removal rate was high. After 15 days the rate became steady. The carbon-14 removed in the initial period estimated to be about 15 Ci in terms of carbon-14 dioxide was from the inventory. The subsequent steady state period reflected the removal of carbon-14 dioxide from the current production and the rate is estimated to be 4.5 Ci/7-days.

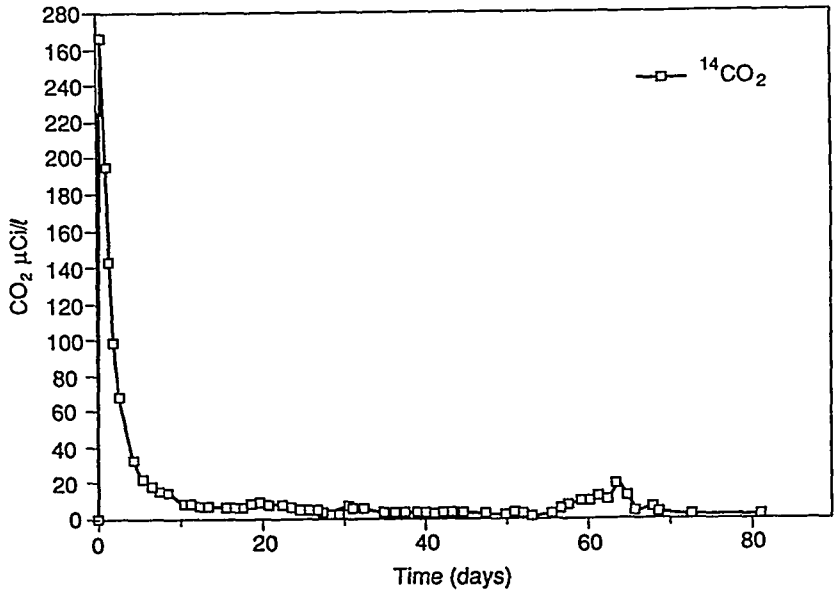

Figure 7: $^{14}CO_2$ Concentration in the PNGS U-3 AGS During C-14 Decontamination by Oxygen Addition

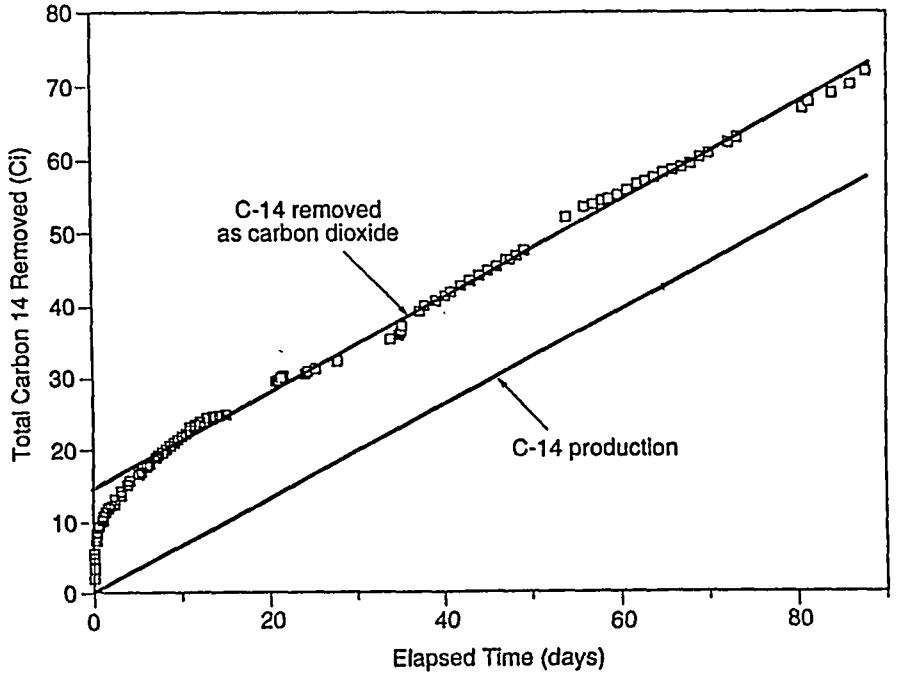

Figure 8: PNGS U-3 AGS C-14 Decontamination Integrated C-14 Removal - Total

Conclusions

At NPD NGS, no carbon dioxide was detected downstream of the carbon-14 dioxide removal system with an entering concentration of 90 vppm. The carbon dioxide concentration in the moderator cover gas was reduced from 90 to 23 vppm. A total removal of 234 g of mixed CO_2 was achieved during the 612 hours demonstration. With the removal unit on line the carbon-14 emission from the moderator cover gas could be reduced to 23/90 or 25% of the level without the removal unit.

At PNGS A Unit 3 under a full load operation, 15 curies of carbon-14 dioxide was removed from the inventory in addition to the current production 4.5 Ci/7 days. No carbon-14 dioxide downstream of the carbon-14 removal unit was detected.

It is demonstrated that the calcium hydroxide bed can be used effectively to clean up carbon-14 dioxide from the gas stream of a CANDU station.

References

1. R.R. Stasko and G.A. Vivian, "An Overview of Ontario Hydro Carbon-14 Control Program", paper presented at International Conference on Radioactive Waste Management, Winnipeg, Canada, September 1982.

2. R.W. Maruska, "A Correlation of Measured Carbon-14 Emissions and Station Operating Parameters", Ontario Hydro Report RMEP-IR-03483-1, November 1981.

3. C.H. Cheh, R.W. Glass and V.S. Chew, "Removal of Carbon-14 from Gaseous Streams", Paper Presented to IAEA Seminar on Testing and Operating of Off-Gas Cleaning System at Nuclear Facilities, Karlsrube, Federal Republic of Germany, May 3-7, 1982.

4. C.H. Cheh, "Removal of Carbon-14 from Nitrogen Annulus Gas" Proceedings 18th DOE Air Cleaning Conference, Baltimore, Maryland (1984).

5. C.H. Cheh et al., "Carbon-14 Removal Method Employing Packed Solid Calcium Hydroxide" U.S. Patent No. 4,492,649 Jan 8, 1985.

6. F.R. Greening, "The Characterization of Carbon-14 Rich Deposits Formed in the Nitrogen Gas Annulus Systems of 500 MWe CANDU Reactors", Radiochimica Acta. 47, 209-217 (1989).

7. Ken Russell, Personal Communication, May 1990.

DISCUSSION

HANDYSIDE: Is it your intention to apply this system or technique to all of your reactors now?

CHEH: Because the annulus gas system is now converted to CO_2, we really don't have a carbon-14 problem in the annulus gas system. If we have to install a system, it would be for the moderator cover gas system. There is no provision for putting in a moderator cover gas carbon-14 removal system in stations such as Bruce and Pickering. At Darlington, as I understand it, the design is such that there is provision for a moderator cover gas system that would allow us to put in a carbon-14 removal system if we so desired.

HANDYSIDE: Did you consider how you will dispose of the carbon-14 labelled waste?

CHEH: The way we are doing it at Pickering is to put calcium hydroxide into stainless steel canisters. These vessels will be shipped to our waste disposal site at Bruce. The intention is to put the stainless steel vessels into drums, pour asphalt around them, and put in the disposal site for storage.

CLOSING COMMENTS OF SESSION CO-CHAIRMAN SHEPARD

I think when we consider all that we have been discussing this morning, both incineration and vitrification and the design of systems, it can be summarized in terms of avoiding polluting the environment. In the area of incineration and vitrification, we could conclude that there is a proven technology that is being used extensively in Europe. Based on Dr. Moeller's presentation, we see that its use in the U.S. is not great but that there are intentions for using it. Notably absent, from the incineration and vitrification portion, as far as I am concerned, was the regulatory framework, especially the high-level waste part. We heard about the low-level part and we saw 10CFR61 and 10CFR20 changes that address it. But I don't recall anything about 10CFR60, and specifically 60.2, which addresses the definition of high-level waste. I don't think that 10CFR60 it or the Nuclear Waste Policy Act addresses mixed high-level waste streams. In other words, it seem to me that more attention is going to have to be given to the separation aspects of high-level vs. non-high-level activity wastes. If that is the case, there has to be a lot of research involved, a lot of creativity. In that regard, we should be able to bring graduate and undergraduate educational research component into the field of incineration and vitrification and create ideas for the continuous training of our graduate and undergraduate students. We can look at this as very important in terms of developing creative research technical ideas.

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11. ABSTRACT *(200 words or less)*

This document contains the papers and the associated discussions of the 21st DOE/NRC Nuclear Air Cleaning Conference. Major topics are: (1) chemical processing systems, (2) reactor operations, (3) incineration and vitrification, (4) particulate filter developments, including filter testing and response to physical and temperature stress, (5) adsorption and testing of activated carbon and adsorber systems, (6) severe accident mitigation including modeling of emergency response systems, (7) nuclear waste management systems, (8) carbon removal, (9) monitoring and measurement systems, (10) the development of standards and regulations and concerns with existing standards and regulations, and (11) nuclear air cleaning activities around the world.

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