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MLM-2609

Commercial Cyclone Incinerator Demonstration Program: October 1978-March 1979

Barbara M. Alexander

April 30, 1979

MASTER

Monsanto

MOUND FACILITY
Miamisburg, Ohio 45342

operated by
MONSANTO RESEARCH CORPORATION
a subsidiary of Monsanto Company

for the
U. S. DEPARTMENT OF ENERGY

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Foreword

Under the sponsorship of the DOE Office of Nuclear Waste Management, Mound is responsible for the development of design criteria for a volume reduction system to treat combustible solid and liquid waste generated in the nuclear industry.

This report is submitted by W. T. Cave, Director, Nuclear Operations, and B. R. Kokenge, Manager, Nuclear Technology, from contributions prepared by members of the Nuclear Waste Technology Section: K. V. Gilbert, Manager, and the Volume Reduction Technology Group: J. W. Doty, Leader.

Previous reports related to this project are listed below:

Volume Reduction System for Solid and Liquid TRU Waste from the Nuclear Fuel Cycle:

October-December 1977	MLM-2509
January-March 1978	MLM-2517
April-June 1978	MLM-2539
July-September 1978	MLM-2569

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Abstract

The commercial cyclone incinerator program was designed to study the effects of burning low-level waste contaminated with beta and gamma emitters in a cyclone system. The ultimate program goal is the demonstration of a cyclone incinerator at a nuclear power plant.

During the past six months, the first three tasks in the project milestone schedule were completed; the project schedule itself covers the period from January 1979 through June 1981. The plan to demonstrate the feasibility of incinerating beta- and gamma-contaminated waste was developed utilizing an extensive literature search, making design modifications to the laboratory cyclone incinerator, and designing an experimental plan. A summary of the feasibility plan was then written and reviewed. It is currently being revised for submission to the Nuclear Regulatory Commission. Following informal NRC review of the feasibility plan, laboratory-scale experiments will begin.

Introduction

The commercial cyclone incinerator program was designed to study the effects of burning low-level waste contaminated with beta and gamma emitters in a cyclone system. The ultimate goal of this program is the demonstration of the Mound cyclone incinerator at a nuclear power plant by the end of FY-1983. The first major event toward achieving this goal is to secure NRC approval of a Topical Report for the cyclone incinerator.

Approval of the Topical Report would signify that the NRC recognizes the benefit to be derived from incineration of power plant waste and initially concurs that the system is safe for operation in a nuclear power plant. This approval would accelerate the licensing process for installation of the incinerator at a specific nuclear reactor facility. By the end of January, 1979, a detailed project milestone schedule identifying five major project objectives and associated work tasks was completed. These major objectives must be achieved before NRC approval of the Topical Report can be successfully obtained. The five major objectives along with anticipated completion dates are listed in Table 1.

Information and performance data gained from experiments performed during the feasibility program utilizing the laboratory-scale incinerator and the demonstration program performed on the full-scale incinerator will provide vital input to publication of the Topical Report for the cyclone incinerator. To ensure that the information thus obtained will be adequate for NRC evaluation, the feasibility plan summary will be submitted for NRC preliminary review before the experimental program is initiated.

Table 1 - MAJOR OBJECTIVES LEADING TO TOPICAL REPORT APPROVAL FOR MOUND CYCLONE INCINERATOR

	Objective	Completion Date
A)	NRC Preliminary Review-Feasibility Plan	August 1979
B)	Complete Incinerator Feasibility Plan	March 1980
C)	Demonstrate Full-Scale Plan	September 1980
D)	Complete Design Criteria	December 1980
E)	Topical Report Completed	June 1981

Task A1 - project milestone schedule completed

The project milestone schedule for securing NRC approval of the cyclone incinerator Topical Report was written during the month of January. It lists five major objectives which must be accomplished before this project milestone can be successfully reached. Topical Report approval is anticipated by the end of June 1981. The timetable for achieving the objectives is given in Figure 1.

Task A2 - feasibility plan developed

Preparation of technical data base

A survey of the available literature on nuclear reactor waste revealed that approximately 45% of all solid waste produced by boiling water reactors (BWR's) and pressurized water reactors (PWR's) is dry. This dry solid waste, however, contains only 5% of the total activity found in solid waste from the power plants [1]. About 80% of the dry solid waste is combustible [2], so that a great reduction in volume could be achieved by cyclone incineration without producing highly active residues.

Dry solid waste from BWR's and PWR's contains a mixture of beta and gamma emitters. The most common isotopes are given in Tables 2 and 3 [1]. The most troublesome contaminant in the waste is radioactive iodine. As is shown in Tables 2 and 3, iodine is either nondetectable or present only in very small quantities in dry solid waste from nuclear reactors, but its removal from incinerator offgas is of great concern. Radioiodine poses a threat to human health because it concentrates in the thyroid gland, and at the same time it is difficult to trap from incinerator offgas because its common forms (such as I_2 and CH_3I) are highly volatile [3].

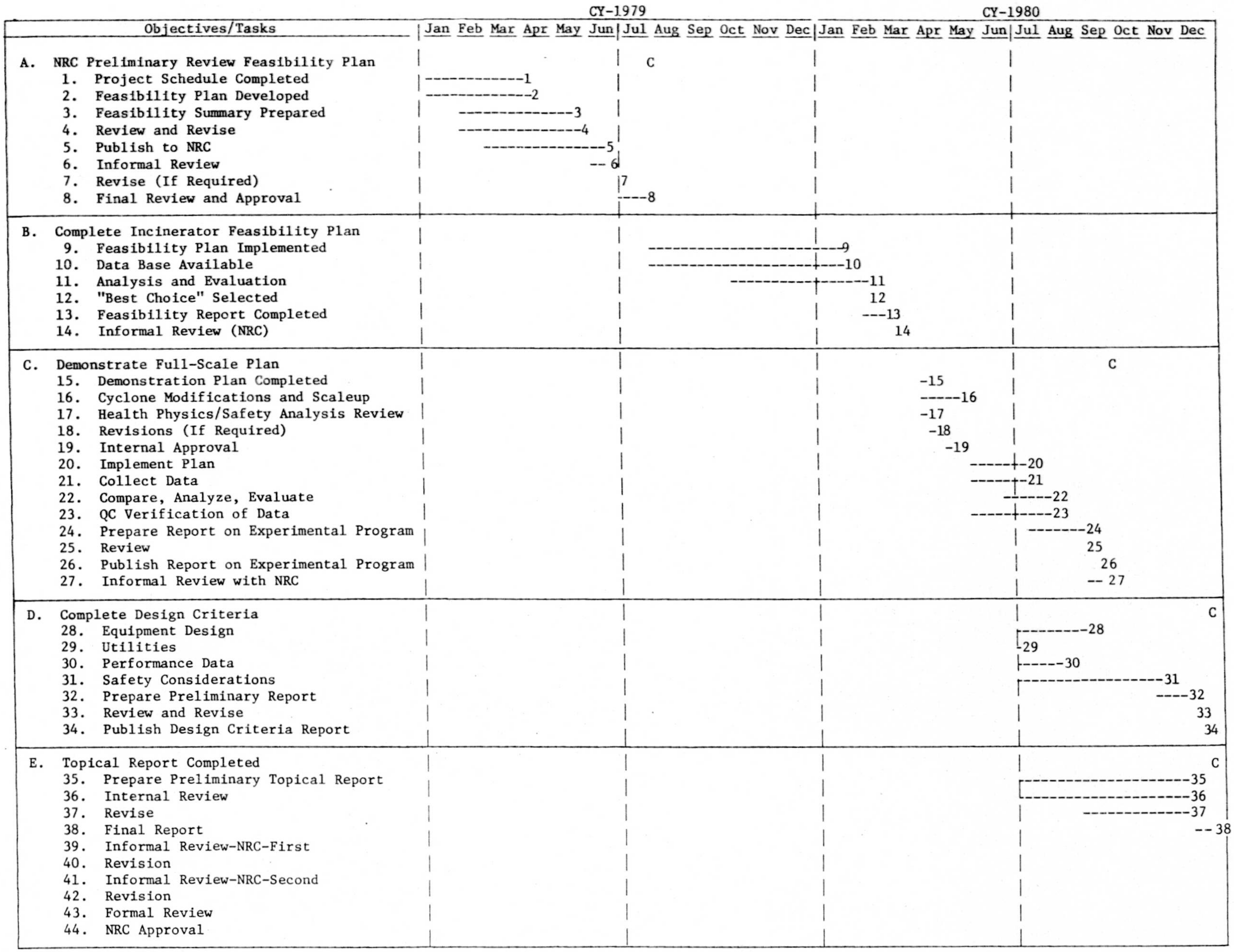

FIGURE 1 - Project milestone schedule for FY-1979, FY-1980, and FY-1981 to obtain commercial incinerator Topical Report approval by NRC.

Table 2 - RADIOISOTOPES PRESENT IN DRY SOLID WASTE FROM PRESSURIZED WATER REACTORS[a]

Isotope	Content Found in 1975 Study[b] (Ci)	(%)	Content Found in 1976 Study[c] (Ci)	(%)
Co-57	0.6	0.9	-	-
Co-58	24.3	35.2	17.1	16.5
Co-60	21.5	31.2	18.2	17.6
Cs-134	6.1	8.8	15.1	14.6
Cs-137	11.7	17.0	28.9	27.9
Mn-54	3.6	5.2	1.1	1.1
Nb-95	0.1	0.1	0.6	0.6
Nb-97	-	-	0.5	0.5
Sb-125	0.3	0.4	-	-
Other	0.8	1.2	22.0	21.2
Total	69.0	100.0	103.5	100.0

[a]From Reference 1.

[b]Based on study of 812.7 m^3 of waste.

[c]Based on study of 877.4 m^3 of waste.

In the designing of the experimental laboratory incinerator system, special efforts were taken to ensure that all iodine in the incinerator feed will be removed from the offgas. All other radioisotopes commonly found in dry solid waste from PWR's and BWR's are nonvolatile and should be totally contained in the incinerator offgas treatment system by HEPA filtration.

Volatile radioiodine compounds are commonly controlled by adsorption on a solid adsorbent, such as activated charcoal or silver zeolite. Zeolite was chosen for installation in the laboratory incinerator offgas system, because of its superior performance and safety. It provides safety because, unlike charcoal, it is not combustible. Zeolite is also insensitive to process upsets, retaining a high removal efficiency for iodine even under conditions of high temperature and high humidity that render charcoal ineffective. At 125°C and >98% relative humidity, the decontamination factor for methyl iodide (CH_3I) is 1000 using silver zeolite as the adsorbent, as opposed to 10.1 using charcoal impregnated with KI and I_2 [4]. The performance of activated charcoal is also adversely affected by

Table 3 - RADIOISOTOPES PRESENT IN DRY SOLID WASTE FROM BOILING WATER REACTORS[a]

Isotope	Content Found in 1975 Study[b] (Ci)	(%)	Content Found in 1976 Study[c] (Ci)	(%)
Co-58	0.5	1.1	0.36	1.1
Co-60	13.3	28.9	19.6	60.1
Cr-51	1.5	3.3	1.6	4.9
Cs-134	4.0	8.7	0.3	0.9
Cs-137	6.6	14.3	3.0	9.2
Fe-59	-	-	0.01	0.03
I-131	-	-	0.1	0.3
Mn-154	12.1	26.3	4.2	12.9
Nb-95	0.02	0.04	-	-
Zn-65	1.0	2.2	1.09	3.3
Zr-95	0.27	0.6	-	-
Other	6.73	14.6	2.33	7.2
Total	46.0	100.0	32.6	99.93

[a]From Reference 1.

[b]Based on study of 507 m^3 of waste.

[c]Based on study of 1015 m^3 of waste.

the presence of three chemical species that are found in incinerator offgas: hydrocarbons and CO_2 will decrease the iodine adsorption efficiency of carbon, and NO_2 may even react explosively with it. Zeolite, on the other hand, is affected only slightly by hydrocarbons and is unaffected by the presence of NO_2 or CO_2. Both charcoal and zeolite, however, must be protected from poisoning by halogenated compounds and sulfur compounds [4, 5]. Silver zeolite is more costly than activated charcoal, but it results in a smaller volume of waste, because of its greater efficiency and higher capacity for iodine retention. The capacity of silver zeolite for methyl iodide retention is 100 mg per gram of zeolite, as opposed to 5 mg per gram of charcoal [6]. Iodine is only physically adsorbed by charcoal and can be desorbed into air or leached into water, but silver zeolite adsorbs iodine into a highly insoluble compound, resistant to leaching or desorption.

To prolong the service life of the silver zeolite bed, means of improving iodine removal in the laboratory incinerator spray scrubber were also investigated. The caustic scrub liquor currently used in the scrubber does have a limited capacity for absorption of iodine. Iodine decontamination factors as high as 160 have been reported for caustic solutions [6].

From a survey of the literature, several chemical additives were found that, when added to the scrub liquor, may improve iodine removal in the spray scrubber. Two additives were chosen for further study: sodium hypophosphite and potassium iodide. Sodium hypophosphite is a strong reducing agent. Another strong reducing agent, sodium thiosulfate, has been used to reduce the volatility of iodine in aqueous solution. Addition of 1 wt % thiosulfate to an aqueous solution containing KOH and iodine has been shown to reduce volatilization of the iodine from 2.5% to less than 0.55% [7]. Hypophosphite will be tested as a scrub liquor additive, rather than thiosulfate, because thiosulfate reacts to form SO_2, which can harm the silver zeolite bed.

Potassium iodide has been widely used as a charcoal impregnant for removal of radioactive methyl iodide and also in aqueous solution to enhance removal of I_2 from air. As a charcoal impregnant, potassium iodide is thought to aid adsorption of radioactive methyl iodide by an exchange reaction between its stable iodine and the radioactive iodine of the methyl iodide molecule [8]. In solution, however, potassium iodide aids adsorption of iodine by reacting with I_2 to form KI_3. The equilibrium constant for this reaction in water near 25°C is 725 [9], indicating that the reaction is very favorable. Potassium iodide will be tested in the scrub liquor of the laboratory incinerator as a possible aid to absorption of iodine.

Design of experimental system

From mid November through the end of February, the laboratory-scale incinerator system was modified to prepare it for experiments on feed spiked with radioisotopes. The system as it existed before modification was described in a previous report [10]. Changes were made to improve the offgas treatment system and to improve the safety of the operation. A photograph of the system following modification is shown in Figure 2.

To prevent the release of fine, light ash from the incinerator combustion chamber during handling, the chamber has been placed inside a fume hood (see Figure 3). Sheets of transite, an insulating material, will shield the glass of the fume hood during burning. Ash remaining in the combustion chamber after each burn will be handled through gloves, with the front of fume hood closed. This will provide contamination control.

A HEPA prefilter has been added to the incinerator offgas system following the spray scrubber. This prefilter, rated as 90-95% efficient for removal of a 0.3-μm particle, will protect downstream adsorbent beds from being plugged by small particles. The first adsorbent bed after the prefilter is silica gel. The former Balston filter housing was modified to accommodate it (see Figure 4).

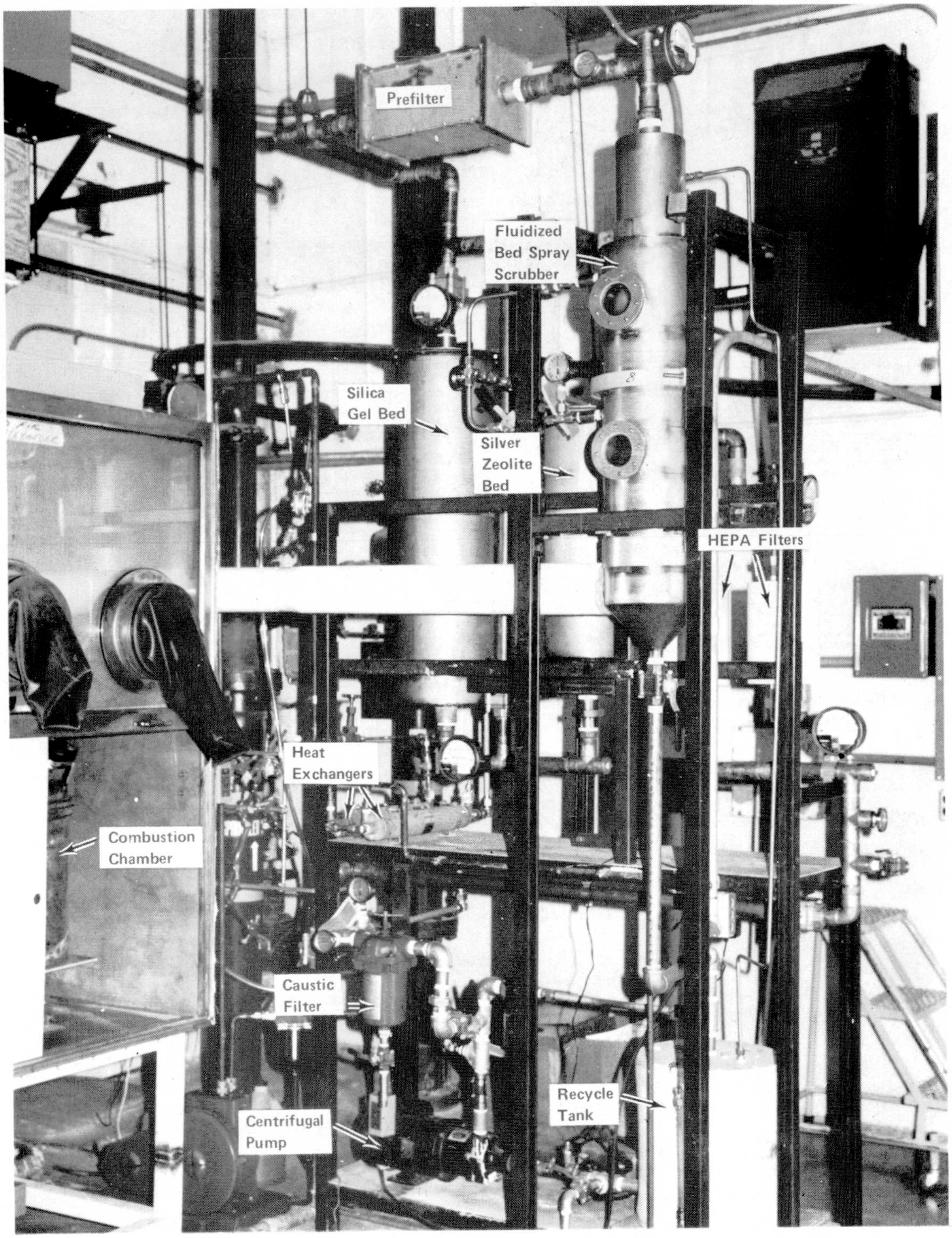

FIGURE 2 - Laboratory-scale cyclone incinerator system for incineration of beta- and gamma-contaminated waste - front view.

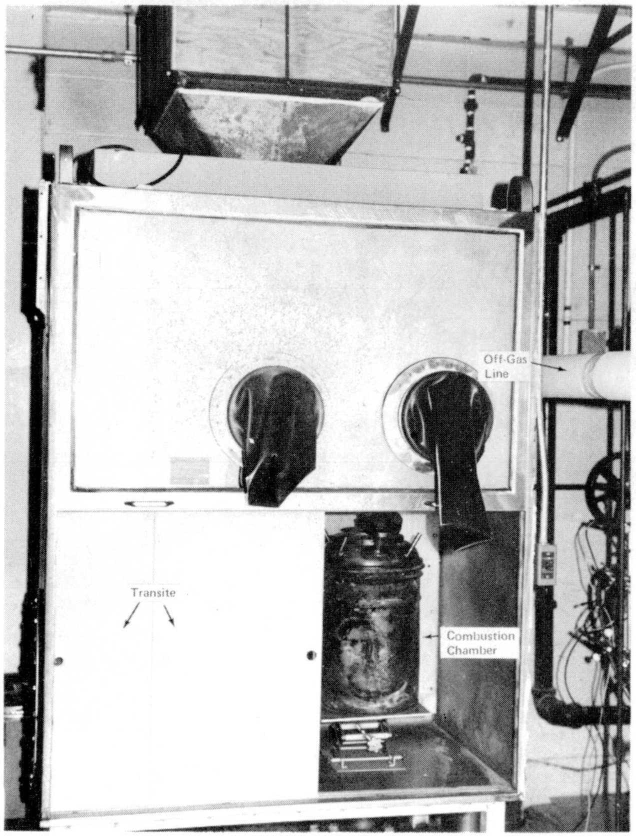

FIGURE 3 - Laboratory-scale cyclone incinerator combustion chamber and containment system.

The silica gel was added to enhance ruthenium removal in the offgas system. Since ruthenium is not found in dry combustible waste from nuclear power plants, it will not be studied immediately, so the offgas will bypass the silica gel bed during the first experiments.

The second adsorbent bed is silver zeolite, added to the offgas system for efficient adsorption of iodine (see Figure 4). This bed will be heated to 125°C by a heating tape wrapped around the outside of the bed. Under these conditions, a methyl iodide decontamination factor greater than 1000 is anticipated.

As a final stage in the offgas treatment system, the offgas will pass through a pair of HEPA filter canisters in parallel (see Figure 4). These filters will remove the last trace of particulate matter and fines entrained from the adsorbent beds before the offgas is exhausted to the building ventilation system.

Design of experimental plan

A plan for implementing the experimental phase of the feasibility plan has been developed in conjunction with the Experimental Design Group at Mound Facility. The experiments will be performed sequentially to permit more efficient use of time and resources. This method will be described in more detail later.

In each experiment, 1500 g of Type I waste will be burned. The composition of Type I waste is given in Table 4. Polyvinylchloride present in the feed will burn to form HCl gas. Most of the HCl will be absorbed in the spray scrubber, but a small percentage of it will be adsorbed in the silver zeolite bed, decreasing its capacity for iodine adsorption. The amount of the decrease will be measured by these experiments to determine whether waste containing polyvinylchloride can be incinerated without foreshortening the service life of the silver zeolite bed.

Before each experiment, the incinerator feed will be spiked with 10 μCi of one of three radioisotopes: cobalt 60, cesium-137, or iodine-131*. Cobalt and cesium were selected for study because

*For reasons of safety, early experiments will be performed using 100 mg of KI as the spike.

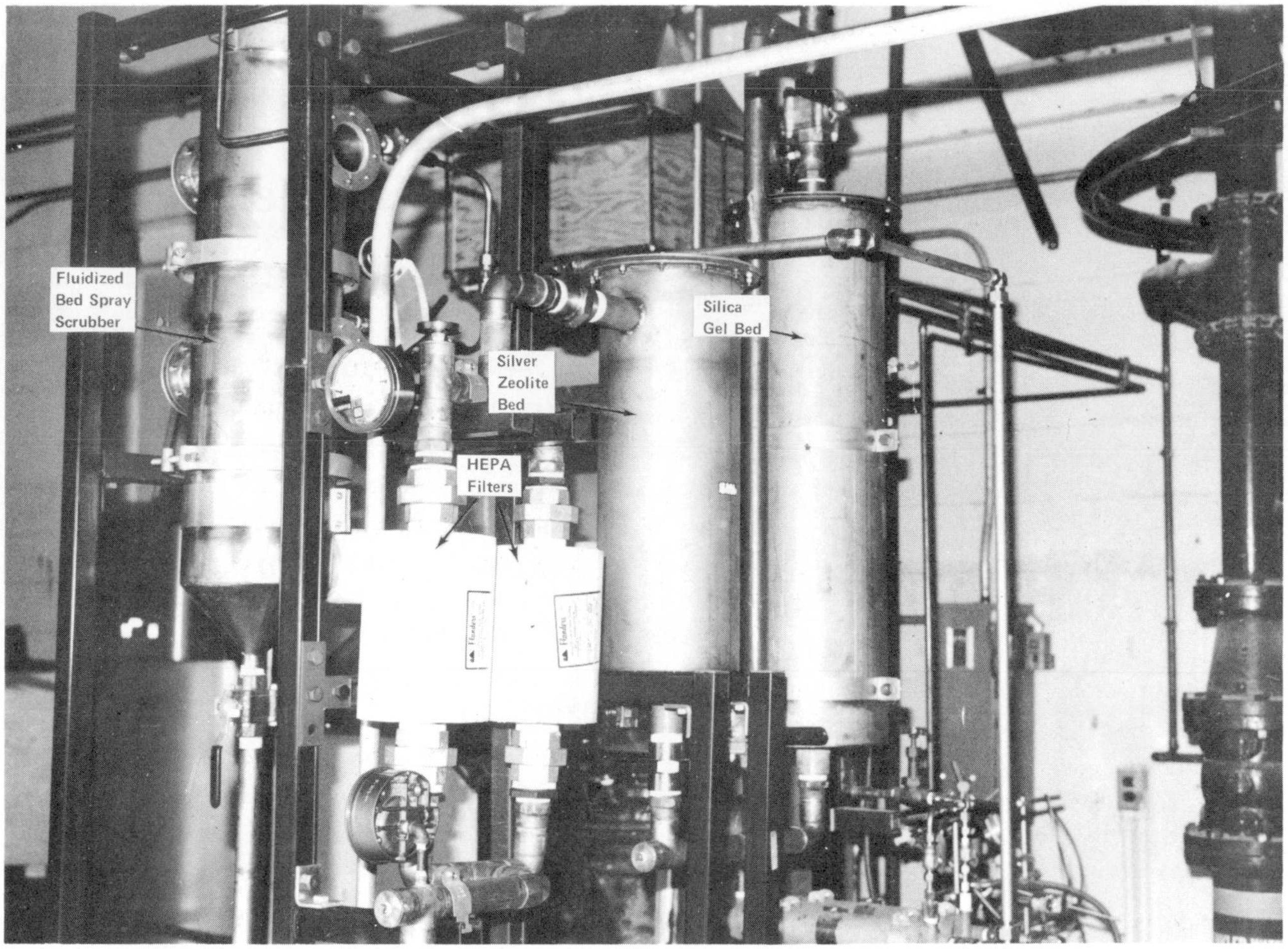

FIGURE 4 - Laboratory-scale cyclone incinerator offgas system - rear view.

Table 4 - COMPOSITION OF EXPERIMENTAL FEED (TYPE I WASTE) LABORATORY CYCLONE INCINERATOR

Component	Amount (wt %)
Paper	34
Plastic	
Polyvinylchloride	9
Polyethylene	31
Polypropylene	9
Rubber	14
Cloth	3
	100

they are two of the most common radioactive species found in solid waste from nuclear reactor facilities. Iodine was chosen because it is a difficult removal problem. Four different scrub liquor compositions will be tested to determine which shows the best decontamination factor for these isotopes. The four scrub liquors to be tested will be caustic only, caustic plus hypophosphite, caustic plus potassium iodide, and caustic plus both hypophosphite and potassium iodide. The experiments are staged sequentially so that the scrub liquors will first be screened to determine which shows the most promise, then a set of experiments will be run to determine the best concentration of the promising scrub liquor(s) to be used. Finally, a full set of experiments will be performed at the chosen concentration to determine how the performance of the scrub liquor changes after repeated use.

Provisions have been made for taking samples of incinerator offgas at four locations: before the prefilter, before the adsorbent beds, and before and after the final HEPA filters. These samples, along with samples of incinerator ash, sludge, and scrub liquor, will be analyzed by gamma-detection, using a Ge(Li) detector with a multichannel analyzer. In this manner, a mass balance of radioisotopes in the laboratory incinerator system can be calculated, and compliance with NRC radioisotope emission limits (Title 10, Code of Federal Regulations) can be verified [11].

From the mass balance data, decontamination factors for each component in the offgas system will be calculated, rate of chemical consumption in the system can be estimated, potential operator exposure from various incinerator components can be identified, and shielding can be placed accordingly. At the end of the set of experiments, the residual iodine adsorption capacity of the silver zeolite will be determined to obtain an estimate of the service life of zeolite in the incinerator offgas.

The scrub liquor that demonstrates the best decontamination factor, based on data from the laboratory-scale experiments, will then be used in a demonstration program on the full-scale cyclone incinerator. A bed of silver zeolite will also be installed in the offgas system of the full-scale unit for this purpose. The demonstration program will help to verify results obtained in the laboratory-scale experiments.

Task A3 - feasibility summary prepared

The Nuclear Regulatory Commission has agreed to informally review the feasibility plan for the Mound Cyclone Incinerator before it is implemented. This will be done to ensure that data from these experiments, which will be used in writing the Topical Report for NRC approval, will be found acceptable.

During the month of February, the first draft of the report describing the experimental plan was completed. It has been reviewed internally and returned with suggested changes. When these changes have been incorporated into the report, it will be sent to the NRC for review, revision, and concurrence.

Quality control

Quality control will be maintained during this program by the use of calibrated instruments and a standard operating procedure. The standard operating procedure will be finalized after the best method for gamma counting incinerator ash and sludge has been determined. This procedure will be followed for each experiment to ensure that all data are obtained under similar conditions.

All deviations from standard procedure will be noted when results are reported, and error limits of all measurements will also be noted to ensure that all conclusions drawn from the data are firmly supported.

References

1. An Analysis of Low-Level Solid Radioactive Waste from LWR's through 1975, U. S. Environmental Protection Agency, Office of Radiation Programs, ORP-TAD-77-2 (November 1977).

2. Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle, U. S. Energy Research and Development Administration, ERDA-76-43 (May 1976).

3. C. A. Burchsted, J. E. Kahn, and A. B. Fuller, Nuclear Air Cleaning Handbook, Oak Ridge National Laboratory, 1976.

4. W. J. Maeck, D. T. Pence, "Application of Metal Zeolites to Radioiodine Air Cleaning Problems," Idaho Nuclear Corporation, presented at the 11th AEC Air Cleaning Conference, CONF-700816.

5. W. J. Maeck, D. T. Pence, and J. H. Keller, "A Highly Efficient Inorganic Adsorber for Airborne Iodine Species," Idaho Nuclear Corporation, presented at the 10th AEC Air Cleaning Conference, CONF-680821.

6. O. O. Yarbro, D. J. Crouse, and G. I. Cathers, "Iodine Behavior and Control in Processing Plants for Fast Reactor Fuels," Oak Ridge National Laboratory, presented at the 11th AEC Air Cleaning Conference, CONF-700816.

7. A. G. Evans and A. H. Dexter, Confinement of Airborne Radioactivity: Progress Report, January-December 1975, Savannah River Laboratory, DP-1430 (September 1976).

8. A. A. Khan, et al., Studies on Removal of Methyl Iodide by Activated Charcoal, Bhabha Atomic Research Center, Bombay, India, 1975.

9. P. Durrant and B. Durrant, Introduction to Advanced Inorganic Chemistry, John Wiley and Sons, Inc., New York, 1962.

10. B. M. Alexander, et al., Volume Reduction System for Solid and Liquid TRU Waste from the Nuclear Fuel Cycle, MLM-2569 (November 1978), 17 pp.

11. Code of Federal Regulations, Title 10 - Chapter 1, U. S. Nuclear Regulatory Commission Rules and Regulations.

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