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Volume I

Vijendra P. Kothari

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Morgantown, West Virginia

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Proceedings of the Environmental Technology Development Through Industry Partnership Conference
Volume I

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Sponsored by

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October 3-5, 1995

MASTER

Foreword

The Environmental Technology Development Through Industry Partnership was held October 3-5, 1995, at the Morgantown Energy Technology Center in Morgantown, West Virginia. This meeting was sponsored and hosted by the Morgantown Energy Technology Center (METC) of the U. S. Department of Energy.

The overall objective of this conference was to review the latest Environmental and Waste Management technologies being developed under the sponsorship of Environmental Management — Office of Science and Technology (OST) Program at METC. The focus of this conference was also to address the accomplishments and barriers affecting private sector, and lay the groundwork for future technology development initiatives and opportunities. Three hundred attendees from industry, academia, National Laboratories, DOE, and other Government agencies participated in a 3-day conference. Twenty-six papers were presented in the EM-OST focus areas:

- Mixed Waste Characterization, Treatment, and Disposal (5 papers)
- Contaminant Plume Containment and Remediation (8 papers)
- Decontamination and Decommissioning (13 papers)

In addition to the presentations, ten Focus Area presentations, thirty one posters of all Focus Areas were displayed. Two panel discussions were also held on the (1) Mixed Waste Characterization, Treatment, and Disposal Issues; and (2) The Application, Evaluation, and Acceptance, of In-Situ and Ex-Situ Plume Remediation technologies.

The papers presented in these proceedings have been reproduced from camera-ready manuscripts provided by the authors. They have been neither refereed nor extensively edited.

V. Y. Kothari

Vijay Kothari
Conference Chairperson

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Appendices

* No paper was submitted.

Session 1

Opening Session

The GETE Approach to Facilitating the Commercialization and Use of DOE-Developed Environmental Technologies

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Introduction/Needs

The Global Environmental Technology Enterprise (GETE)* was conceived to develop and implement strategies to facilitate the commercialization of innovative, cost-effective Department of Energy (DOE)-developed environmental technologies. These strategies are needed to aid DOE's clean-up mission; to break down barriers to commercialization; and to build partnerships between the federal government and private industry in order to facilitate the development and use of innovative environmental technologies.

Objectives/Problem

DOE's environmental clean-up mission is huge and complex. For forty years, DOE weapons facilities designed, manufactured, and tested nuclear weapons to support the national defense. As was the case with most manufacturers that operated before an understanding of environmental stewardship was developed, this weapons production resulted in soil and groundwater contamination. In 1989, at the close of the Cold War, DOE set a goal of cleaning up its weapons complex to bring all sites into compliance with applicable environmental regulations by the year 2019.

The DOE Office of Environmental Restoration and Waste Management (EM) is responsible for remediating its contaminated sites and managing DOE's waste inventory in a safe and efficient manner.[i] DOE EM faces the challenge of cleaning up more than 100 contaminated installations in 36 states and territories. This includes 3,700 sites, representing over 26,000 acres containing hazardous or radioactive contaminated surface or groundwater, soil, or structures.[ii] The cost to remediate these sites, and others which have not yet been formally classified, is not well-defined. DOE estimates have placed the cost at over $200 billion over 30 years.[iii] One private study has estimated the cost of remediation at $250 billion.[iv]

Many factors will influence the cost of this clean-up. Among these factors is the cost of developing and utilizing remediation technologies. The overall cost could be reduced with the use of more cost-effective technologies, many of which are believed to reside in Department of Energy facilities. However, real and perceived barriers exist which prevent these technologies from being deployed at DOE sites or by other agencies and the private sector. These barriers include: 1.) lack of awareness and

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*The GETE Project is supported by the U.S. Dept. of Energy's Morgantown Energy Technology Center, under cooperative agreement DE-FC21-94MC31179, with the Global Environment & Technology Foundation, 7010 Little River Turnpike, Suite 300, Annandale, VA 22003; phone: 703-750-6401.

access by the private sector to these technologies; 2.) unwillingness to use new technologies due to environmental regulations and enforcement or the *perception of those factors;* 3.) lack of financing by potential partners which prohibits investments in new environmental technologies; 4.) unfamiliarity with and a lack of consideration in many government facilities of commercialization objectives; and 5.) issues relating to property rights of publicly-developed technologies. Potential economic value is often defined by the intellectual property rights associated with a particular technology. When these rights are left unprotected or undefined, commercial partners fear that intellectual property rights may be compromised.

Approach/Solution

The deployment of innovative DOE-EM technologies could provide more cost-effective treatments than remediation technologies and methods currently in use. By building partnerships between the public and private sectors, the technological expertise of federal researchers and engineers and the commercial incentive of industrialists and other non-government interests can combine to help move these technologies "off the shelf" and toward use at contaminated sites.

The GETE approach to achieving this end is: 1.) to develop a process which identifies and assesses DOE-developed technologies from the perspective of commercial marketability; assists in bringing these technologies to the attention of the private sector; and aids, if necessary, in business planning and start-up activities; 2.) to establish a state-of-the-art electronic communications system, the Global Network of Environment & Technology (GNET), to disseminate information on DOE-developed technologies, as well as information on capital and financing availability, regulatory matters, and commercialization objectives to potential business partners and others; and 3.) to undertake public participation and outreach activities designed to address barrier reduction and public acceptance issues.

Program Description/Technology

1. EM's Office of Technology Development (OTD) supports applied research and demonstration efforts to develop and transfer innovative, cost-effective technologies to its site clean-up and waste management programs.[v] The GETE process seeks, at a minimum, to identify and facilitate the use of those technologies that meet the needs of the OTD Focus Areas and which show the best commercial promise. These Focus Areas are: Contaminant Plume Containment and Remediation; Mixed Waste Characterization; Treatment and Disposal; High-Level Waste Tank Remediation; Landfill Stabilization; and Facility Transitioning, Decommissioning, and Final Disposition.

The GETE technology commercialization process consists of three phases: **Technology Identification**, in which technologies are identified, screened, and selected; **Technology Qualification**, in which in-depth assessments of technologies are conducted with regard to technological and commercial potential; and **Technology Presentation**, in which technologies are "brokered," or matched with public and private partners. This phase also includes business planning, start-up, and financial development assistance.

Technology identification entails meeting and working closely with OTD Focus Area Project Managers, and establishing technology identification criteria specific to the internal success factors of DOE; thus, technology identification is a malleable process which can vary from one field office to another. Once identification criteria have been established, GETE personnel work with Focus Area personnel to develop an initial list of technologies that appear to be fairly advanced in the DOE development process. This process is made up of stages defined by DOE "Technology Development Gates," which begin with basic research (Gate 0) and extend to production and implementation (Gate 5). In this case, GETE is most interested in technologies which are at Gates 3 through 5: engineering development, demonstration, and production and implementation.

The initial list of technologies is discussed by GETE with Focus Area personnel, and analyzed in accordance with a standardized "Process Acceptance Screening," a list of questions to help quickly assess a technology's potential. This initial screening is general and is aimed at removing from consideration any technology that could be hampered by issues that make successful commercialization unlikely, such as problems with intellectual property rights. Those technologies which show promise are subjected to a more comprehensive screening focused on gathering more information relevant to a technology's commercial potential. Technologies which are not eliminated are passed on to the next phase, technology qualification.

Technology qualification entails a more formal assessment which includes not only a rigorous technical evaluation by an independent third-party evaluator, but also an examination of potential commercial viability, competition, and other such factors in determining if the technology is ready for the marketplace. This step takes between six and ten weeks. When the evaluator believes that they have been able to accomplish the study to a 50 percent completion level, a preliminary report is submitted by the evaluator to GETE. This not only provides a status report on the assessment, but also includes a recommendation on whether to proceed further. If a favorable evaluation is made at the conclusion of the technology assessment, GETE will move forward to prepare a comprehensive market study of the technology to determine the size and scope of its potential market. This market study analyzes potential demand for the technology by DOE and other federal agencies, state and local governments, and commercial and international customers. This step takes two to three months, and facilitates the teaming of the technology with a commercial partner and financing.

The next step is the commercial development phase. In this stage, GETE works to find an appropriate commercial partner for a technology, if one has not already been identified. In many cases, technologies at the later DOE Technology Development Gates have found partners. If a technology has no business partner, GETE can actively seek one through such venues as the Commerce Business Daily or the Global Network of Environment & Technology (GNET, described later in this paper). In all cases, GETE maintains fairness of opportunity through dissemination of information on technologies via the Internet, printed material, and participation in public forums.

The commercial development phase also includes business planning and start-up activities. These can include assistance in the preparation or review of a commercial partner's business plan. Business planning activities must be agreed upon by all concerned, and for that reason, GETE has developed commercial non-disclosure and teaming agreements. Financial development assistance, in which GETE assists in locating and negotiating financial backing, is also provided, if needed.

2. The GETE commercialization process is complemented by the information resources available to the public through GNET. GNET is an Internet-accessible system for sharing and disseminating environmental technology information, and includes features which allow for interaction between users.

GNET has sites on both the World Wide Web (WWW) and The Microsoft Network, and provides timely, easily-accessible information on environmental technologies (DOE and non-DOE-developed), market opportunities, current events, business and financing assistance, and regulatory and legal matters.

One of the goals of GNET is to generate significant private sector interest in the site, in order to provide greater visibility, and to generate interest in innovative environmental technologies, resulting in more technologies being deployed for use. GNET will accomplish this by providing value-added, timely, pertinent information in a convenient, easily-accessible format. This information will range from federal government reports to abstracts of articles published in other sources to databases of technology and program information.

3. GETE outreach activities are designed to assist stakeholders in their efforts to utilize and/or bring to market DOE-developed environmental technologies, and to identify and address existing commercial barriers. These activities include participation in conferences, the production of printed material, and the development of GNET on-line information resources and forums. This activity also includes development of a national affiliates program, and in-depth investigation of strategies to address intellectual property, fairness of opportunity, and conflict of interest issues that arise during the commercialization process.

Accomplishments

GETE accomplishments after one year include:

1. The GETE commercialization process has identified, within DOE OTD Focus Areas, over 185 DOE technologies, and has completed in-depth technical assessments of four technologies. GETE has provided commercial development services to 30 companies. This includes establishing seven teaming agreements, 12 non-disclosure agreements, and three strategic partnerships. General business assistance has been provided to eight companies.

2. GNET is fully operational on the World Wide Web and The Microsoft Network and is continuing to develop a full suite of information resources. GNET was cited by Vice President Al Gore as the network for federal interagency environmental technology collaboration and information-sharing activities. GNET provided an on-line interactive communications service for the White House's Technology for a Sustainable Future Initiative, facilitating development of the national environmental technology strategy, "Bridge to a Sustainable Future." GNET services are also used by the Interagency Environmental Technologies Office, U.S. Environmental Protection Agency Common Sense Initiative, as well as other governmental agencies, non-governmental agencies, and businesses in the environmental technology arena.

3. GETE public outreach activities have included playing a key role in the White House Conference on Environmental Technology, and assisting with 30 local and regional workshops for the White House Technology for a Sustainable Future Initiative. GETE has participated in the White House Sustainable Power Conference, Rocky Flats Environmental

Institute Microbacterial Purification Conference, Weapons Complex Monitor Applied Research and Technology Colloquium, Sandia National Laboratory Environmental Decision Coalition, and other conferences. GETE utilizes GNET to disseminate information on environmental technologies and government matters of interest. In addition, a GETE Advisory Council, a group of experts who will develop methods to remove barriers to commercialization, has been formed.

Applications/Benefits

The overall benefit of GETE is derived from moving environmental technologies from National Laboratories and into the marketplace by facilitating and accelerating the commercialization process. The increased use of innovative, cost-effective environmental technologies can help reduce the overall cost of remediating contaminated sites.

Future Activities

Planned activities for FY 1996 include: continuing technology identification, qualification, and presentation activities; adapting GNET so that it can begin to generate revenues; and conducting a major technology trade fair to highlight DOE-developed technologies.

Acknowledgments

The GETE project is supported by the Department of Energy-Morgantown Energy Technology Center, under the three-year cooperative agreement DE-F21-94MC31179, commencing October 1, 1994, with the Global Environment & Technology Foundation, 7010 Little River Turnpike, Suite 300, Annandale, VA 22003; phone: 703-750-6401. Subcontracting partners include the Environmental Export Council, National Association of State Development Agencies, National Technology Transfer Center, Professional Services International, Inc., and Science Applications International Corporation. Kelly Pearce of the Environmental & Waste Management Division, Morgantown Energy Technology Center is the Contracting Officer's Representative. GETE would also like to acknowledge the DOE Office of Science and Technology Development at METC and Denise Riggi, Contract Specialist, for continued leadership and support of this effort.

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[i] U.S. Department of Energy Office of Environmental Restoration and Waste Management - Office of Technology Development, Technology Catalog, First Edition, February 1994, page 1.

[ii] U.S. Department of Energy Office of Environmental Restoration and Waste Management - Office of Technology Development, A National Program, October 1993.

[iii] U.S. Department of Energy, Estimating the Cold War Mortgage: The 1995 Baseline Environmental Management Report, March, 1995.

[iv] Nuclear Waste News, July 15, 1993, page 271.

[v] U.S. Department of Energy Office of Environmental Restoration and Waste Management - Office of Technology Development, Technology Catalog, First Edition, February 1994, page 1.

Poster Session I

PI.1 Electromagnetic Mixed Waste Processing System for Asbestos Decontamination

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Introduction

DOE sites contain a broad spectrum of asbestos materials (cloth, pipe lagging, sprayed insulation and other substances) which are contaminated with a combination of hazardous and radioactive wastes due to its use during the development of the U.S. nuclear weapons complex. These wastes consist of cutting oils, lubricants, solvents, PCB's, heavy metals and radioactive contaminants. The radioactive contaminants are the activation, decay and fission products of DOE operations. The asbestos must be converted by removing and separating the hazardous and radioactive materials to prevent the formation of mixed wastes and to allow for both sanitary disposal and effective decontamination. Currently, no technology exists that can meet these sanitary and other objectives.

Objectives

The overall objective of this project is to develop and demonstrate a cost-effective technology for converting contaminated asbestos to a form suitable for disposal in a sanitary landfill or to a form which stabilizes inorganic, radioactive and heavy metals in a solidified silica suspension for disposal as low-level waste.

The electromagnetic mixed waste processing system employs three patented technologies to convert DOE asbestos to a non-hazardous, radionuclide-free, sanitary waste: high-shear acid decomposition (ABCOV Method), metals separation (Westinghouse STC), and radio frequency (RF) heating (KAI Electromagnetic Process). The asbestos is decomposed to an amorphous silica suspension using the high-shear acid process. Radionuclides and heavy metals are separated from this suspension by ion exchange and physical separation processes. The suspension is then heated with radio frequency energy to remove

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Research is sponsored by the U.S. Department of Energy's Morgantown Energy Technology Center under contract DE-AC21-94MC29249 with KAI Technologies, Inc., 170 West Road, Suite 4, Portsmouth, NH 03801, 603-431-2266; telefax: 604-431-4920.

the organic volatiles. Finally the amorphous silica is solidified with sodium silicate for disposal.

The objectives of Phase 1 were to establish the technical feasibility of asbestos decomposition, inorganic radionuclide and heavy metal removal, and organic devolatilization. The technical feasibility involved separate bench-scale testing of:

1. Decomposition of asbestos to an amorphous silica suspension using the ABCOV technology described in U.S. Patent Number 5,041,277; performed by Ohio DSI Corporation in Columbus, OH.

2. Removal of inorganic radionuclides and heavy metals designated as hazardous by the Resource Conservation and Recovery Act (RCRA) using ion exchange and physical separation processes; performed by WSTC in Pittsburgh, PA.

3. Devolatilization of organics using RF-based technology described in U.S. Patent Number 5,065,819; performed by KAI Technologies, Inc. in Portsmouth, NH.

Technical Approach

Apply techniques that have already proved successful in the mining, oil and metal processing industries to the development of a multi-stage process to remove and separate hazardous and radioactive materials from asbestos. This process uses three methods: acid attack which converts the asbestos to a sanitary waste; electrochemical processing for the removal of heavy metals, RCRA wastes and radionuclides; and dielectric heating to volatilize the organic materials. This process will result in the destruction of over 99% of the asbestos; limit radioactive metal contamination to 0.2 Bq alpha per gram and 1 Bq beta per gram; reduce hazardous organics to levels compatible with current EPA policy for RCRA delisting and achieve TCLP limits for all solidified waste.

Technologies

The electromagnetic mixed waste processing system employs three patented technologies to convert DOE asbestos to a non-hazardous, radionuclide-free, sanitary waste; high shear acid decomposition (ABCOV Method), radio frequency heating (KAI Electromagnetic Process) and electrochemical separation(Westinghouse STC). The asbestos is decomposed to an amorphous silica suspension using a high-shear acid process. Radionuclides and heavy metals are separated from this suspension by an electrochemical process and radio frequency heating is employed to remove organics. Finally, the amorphous silica is solidified with sodium silicate for disposal.

The ABCOV Method is a non-burning, simple and economical, chemical and mixing treatment of friable asbestos-containing material (ACM) that renders it harmless in a period of two hours or less. The ABCOV process was developed after years of extensive research at Battelle Laboratories and Georgia Institute of Technology. It uses several chemical formulations to effectively improve the removal of asbestos-containing materials (ACM) and chemically converts asbestos into a non-hazardous substance. It offers the following advantages over conventional removal and disposal methods:

- Removal time is reduced by applying ABCOV-T directly to the ACM, achieving improved wetting and initiating the conversion process. Some studies have documented removal times to be reduced by as much as 40 percent, allowing for labor savings .In addition, after removal of ACM, ABCOV-T can be used to clean substrates of any remaining fibers, eliminating the need for scrubbing the wire brushes.

- Depending on the type of filler material in the ACM, the volume of waste may be reduced by as much as 80%. The removed ACM is immersed in a vat containing formulation ABCOV-C. After 30 or more minutes of agitation using a high-speed dispersion mixer, the mixture may be analyzed for the presence of asbestos fibers. Several previous demonstrations of the process by major utilities and federal institutions have confirmed conversion into a non-ACM substance.
- Since a hazardous substance no longer exists, it becomes unnecessary to transport the remaining material to a required ACM landfill.

Thermal processing is the most effective approach to removing organics from the mixed waste matrix. As thermal processing candidates, both direct incineration and vitrification processing would destroy any organics present in the asbestos, and in particular, vitrification can result directly in a stable waste form for disposal. The KAI radio frequency (RF) desorption technology was selected over these processes for the following reasons:

- Incineration - a) requires more complicated licensing for on-site, mobile operations ;b) requires more expensive off-gas processing/particulate collection system to contain airborne contaminants for an ALARA design for nuclear application (this includes other asbestos fibers and radionuclides); and c) may condition metals present in the asbestos decomposition residue to make radionuclide and heavy metal extraction more difficult.

- Vitrification would destroy organics condensing the asbestos and metals into a single vitrified waste form, but in so doing generates no potential waste minimization through release of a "sanitary" fraction in the form of the ABCOV product silica suspension.

By contrast, RF desorption meets both the needs of the DOE PRDA for a decontaminated, (potentially) sanitary waste form and the EPA requirements for a pollution prevention and (secondary) waste minimization in any waste treatment.

Because RF dielectric heating "couples" directly with the waste at the molecular level, it does not rely on convective or conductive mechanisms for heat transfer. Such coupling is an intrinsic advantage of both radio frequency and microwave heating approaches (for materials with which they couple and which, otherwise, may resist conventional heating due to their insulating characteristics).

This asbestos is, by definition, an insulation material, and so bulk-scale, thermal desorption processing by convective or conductive heating of the asbestos matrix will be extremely inefficient. Through the use of radio frequency coupling, the proposed process system penetrates the asbestos matrix effectively and results in rapid, even heating of organic molecules. Because RF coupling is a "non-invasive" heating process which injects energy directly into the substrate's atoms and molecules, RF-driven desorption reduces the risk of airborne contamination with the results:

- An ALARA design is maintained, critical for all nuclear operations
- Man-ran exposure decreases through reduced maintenance and reduced material handling.
- Risk of asbestos fiber release is reduced.

Westinghouse technology for the various

metals and radionuclide extraction includes a multi-step approach:

- Removal of metals and metallic oxides by flotation or other gravimetric approaches.
- Filtration of the solids from the processing liquor.
- Ion exchange and/or electrochemical processing to process the liquids.

One processing alternative to address hazardous and radioactive metals is direct plasma vitrification of the asbestos matrix (without separations). While feasible (and may even be cost-effective), direct vitrification does not support direct discharge of a sanitary waste product; therefore, we have discarded it for meeting the PRDA constraints.

Another alternative is using chemical lixiviants (based on commercial mining experience) to dissolve the metal contaminants from the asbestos matrix (or its ABCOV decomposition products) as opposed to our proposed physical processing. The primary issue for chemical leaching is the potential mismatch between the contaminant lixiviant chemistry and of the ABCOV decomposition chemistry. In addition, the leaching approach requires additional solid/liquid separations to segregate the decomposition and extraction reagents resulting in

:

- Risks of release/spill
- Increased process complexity
- Greater secondary waste generation

Accordingly, we have discarded leaching for this specific application as well - recognizing that it may well be the process of choice for other applications.

Waste Forms

The potential waste forms for the stabilization operation after metals extraction from the asbestos include the following:

- A "fine fraction" consisting of metallic ions adsorbed into the ABCOV generated silica suspension is a possible but unlikely form since we do not anticipate any chemical binding between the asbestos/silica and the contaminants. Further we anticipate that the metallic contaminants will have hydrolyzed to metallic oxides prior to the asbestos removal and D&D operations. The contaminants were originally metal chlorides of U, Ra, Th and others - all of which hydrolyze readily on exposure to moisture or oxidizing conditions. As a result, these oxides should have been separated by the density/gravimetric operations based on density differences since the density of silica is approximately 2.2 g/cc compared to thoria at 9.86 g/cc and urania at 8.3 to 11g/cc; some combination of either floatation classification or magnetic separation should produce a separate, contaminant fraction.

- The metallic oxides fraction from the density/gravimetric separation is likely.

- The "clean" or "sanitary" residue of the asbestos decomposition process - only approximately one-tenth to one-eighth of the original asbestos volume is the major constituent.

- Any ion exchange or filtration media generated during the process must be included as the process secondary waste.

Note that all these streams are compatible with standard LLW radwaste stabilization media - grout, epoxies, polyesters and/or silicates.

Accomplishments

Based on the "success" criteria established in the Phase 1 Laboratory-Scale Test Plan, the objectives of the bench scale testing of the KAI RF organic process were achieved in that a near optimum treatment temperature was achieved for PCE and that the laboratory testing demonstrated that over 90% of an organic contaminant (PCE) could be removed from the silica suspension (converted ACM). During the test a heating rate of 20°C/hour was achieved for the silica suspension using 200 watts of RF power. These tests achieved significant removal of PCE operating near its boiling point of 121°C. The electromagnetic energy required to accomplish the removal of the organic contaminant was quite reasonable at 1.2 kilowatts per kilogram of the silica suspension. The residence time can be significantly reduced by increasing the RF power level. For example, 30 minutes or less is a realistic goal for RF power levels in the range of 2 kilowatts. The total number samples analyzed under EPA method 601 was 30 (2 samples from Test 2 (Control); 14 samples each for Test 3 and Test 4).

A PCB surrogate RF plasma destruction feasibility study was undertaken as an addition to the objectives described in the Phase I KAI Test Plan. The PCB surrogate chosen was 1,2 - dichlorobenzene, and glass wool was chosen as a surrogate for asbestos. RF energy was used to both volatilize and begin decomposition of the PCB surrogate. A Bronson Model PM-310 RF glow plasma discharge chamber operating at the ISM (Industrial, Scientific, Medical) frequency of 13.56 MHz was employed. The reactor vessels were small, 15 cm long x 10 cm in diameter, and the residence time was limited for these preliminary experiment, therefore limiting the extent of destruction possible for the dichlorobenzene. However, the key reactions leading to the fracture of the benzene ring were initiated in the first experiment. This was evidenced by the compounds detected by an independent laboratory evaluation of the products produced using the cold plasma. The initial reactions involve removal of hydrogen and chlorine obstruction from the dichlorobenzene creating free radicals.

These radicals can then combine with gas-phase radicals or lead to fracture of the aromatic ring. Decomposition of the rings (desired result) occurs by additional energy absorption from collisions with photons, ions, electrons and excited atoms and molecules. The additional compounds collected or trapped provide evidence that these reactions are beginning to take place.

The Westinghouse Phase 1 Laboratory-Scale Program established the technical feasibility of inorganic heavy metal and radionuclide removal at the laboratory scale using ion exchange and physical separation technologies. Test results lead WSTC to propose a revised integrated mixed waste processing system design for remediation of contaminated asbestos.

The success criteria for Phase 1 were met:

1.) The feasibility of ion exchange to remove metals from the process stream was demonstrated by obtaining:

- 92% uranium removal in single-contact tests with a commercial ion exchange resin.
- 0.6 equivalents/l resin loading with uranium.
- lead retention in the solid phase for stabilization, allowing separation of the radioactive materials from hazardous contaminants.

2.)The potential to regenerate the ion exchange resin was demonstrated by obtaining 52% regeneration of the resin in one-cycle screening tests.

Ohio DSI Corp. (OHIO) performed a series of asbestos-destruction runs. The purpose of the runs was to demonstrate destruction and to generate treated material for experimental operations by KAI Technologies, Inc. (KAI) and Westinghouse Science & Technology Center . OHIO was tasked to initially analyze the ACM, wet it using ABCOV-T and treat it using ABCOV-C. The resulting solution and solids were then provided to KAI and Westinghouse. OHIO was tasked to provide each contractor with 30 liters of solution resulting from the treatment of 4.1 kilograms of asbestos. The following results were achieved:

- Thirty-three runs were made to treat ACM with ABCOV-C solution. In all 33 cases, the fibers were converted in 45 minutes.

- The final fluoride content, final pH and final volume varied with each run.

- The average amount of make-up ABCOV-R for each run was 310 grams, or 2.77 grams per gram of dry-weight asbestos (1.84 grams per gram of ABCOV-T-wetted material).

- Runs 1 through 32 were packaged into two containers. Runs 1 through 16 were delivered to KAI in Woburn, MA on October 11, 1994. Runs 17 through 32 were delivered to Westinghouse in Pittsburgh, PA on October 11, 1994.

Applications

Laboratory-scale tests on each processing operation have provided the basis to conceptually design a cost-effective, integrated process for remediation of asbestos-containing material (ACM) that is contaminated with organics, heavy metals and radioactive metals. Process characteristics include:

Flexibility: Although the target remediation material for the system is asbestos, the process for inorganic removal has been applied to other types of contaminants. Concrete, demolition debris and soil are material types that could also be remediated using the basic system elements being developed in this program.

Demonstrated Unit Operations: The overall process uses unit operations and technologies that have a proven track record for related remediation problems. For example, the metals separation process is based on Westinghouse soil washing experience. Successful soil washing of uranium-contaminated solution mining site at Bruni, Texas demonstrated the application of similar metals removal processes for soil remediation.

Economic Potential: The process offers significant economic advantage over high-temperature vitrification and mixed waste disposal options for the mixed waste asbestos containing materials.

Environmental Benefits: The process will be designed to have zero fugitive emissions. It will eliminate the need to consider incineration or other high-temperature processes that require stacks and gas cleaning systems. The converted asbestos is a nonhazardous material.

Future Activities

The overall objective of this project is to develop and demonstrate a cost-effective technology that converts contaminated asbestos to a form suitable for disposal in a sanitary landfill, or to a form which stabilizes inorganic radioactive and heavy metals in a solidified silica suspension for disposal as low-level waste.

The objectives of Phase 2 are to optimize the Electromagnetic Mixed Waste Process for removal of inorganic radionuclides and heavy metals from contaminated asbestos and to develop commercial-scale designs and system economics of the Electromagnetic Mixed Waste Process.

The objectives of Phase 3 are to demonstrate the Electromagnetic Mixed Waste Process for removal of inorganic radionuclides and heavy metals in a full-scale system, to gain regulatory acceptance to update system economics, to conduct a market analysis and to develop a commercialization plan.

Contacts

KAI Technologies, Inc. is actively engaged in the innovative use of electromagnetics for this and other environmental problems. For information regarding this project, please contact:

Contractor Project Manager; Raymond S. Kasevich, KAI Technologies, Inc., 170 West Road, Suite 4, Portsmouth, NH 03801, 603-431-2266

DOE's Morgantown Energy Technology Center (METC) supports the Environmental Management (EM) Office of Technology Development by contracting research and development of new technologies for wast site characterization and clean-up. For information regarding this project, the DOE contact is: DOE Project Manager, James Longanbach, 304-285-4659

PI.2 Electrokinetic Decontamination of Concrete

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Introduction:

The U.S. Department of Energy has assigned a priority to the advancement of technology for decontaminating concrete surfaces which have become contaminated with radionuclides, heavy metals, and toxic organics. This agency is responsible for decontamination and decommissioning of thousands of buildings.

Electrokinetic extraction is one of the several innovative technologies which emerged in response to this initiative.

This technique utilizes an electropotential gradient and the subsequent electrical transport mechanism to cause the controlled movement of ionics species, whereby the contaminants exit the recesses deep within the concrete.

Objectives:

The primary mission of this PRDA was to demonstrate the feasibility of this approach as a means to achieve "release levels" which could be consistent with unrestricted use of a decontaminated building.

The secondary objectives were:

- To establish process parameters;
- To quantify the economics;
- To ascertain the ALARA considerations;
- To evaluate wasteform and waste volume.

Observations to Date: The work carried out to this point has achieved promising results to the extent that ISOTRON® has been authorized to expand the planned activity to include the fabrication of a prototype version of a commercial device.

This prototype unit includes a carpet-like surface pad which carries out the extraction step.

The extraction pad is connected via tubing and power cable bundle to the electrokinetic separation module (ESM). This ESM recycles the liquid electrolyte (which is circulated from the extraction pad). The ESM apparatus carries out an electrodialysis separation of radionuclides from the electrolyte.

Current project planning calls for this equipment to be operated at the DOE's K-25 Site during the months of October and November 1995.

Approach:

The technology for electrically forcing

Research sponsored by the U.S. Department of Energy's Morgantown Energy Technology Center, under contract DE-AC21-93MC30162 with ISOTRON® Corporation, 13152 Chef Menteur Hwy., New Orleans, LA 70129; telefax: 504-254-5172.

contaminants through concrete has a precedent in work carried out under sponsorships of the U.S. Department of Transportation. In this application, an electrical gradient is used to prevent corrosion of steel reinforcing within bridge decks. The mechanism provides a means to remove de-icing salts from concrete, and at the same time, the intense cathodic polarity applied to the rebar prevents subsequent corrosion.

The technology for electrically forcing radioactive contaminants through concrete was studied by Dr. William Bostick and colleagues during 1993-1994 with positive results, however, the early work was designed to favor use of electroosmotic transport of contaminants.

The first phase of ISOTRON®'s work focused on study of the fundamentals of dissolution (and subsequent "freeing" of these contaminants from the concrete matrix). Only after the contaminants are solubilized within the concrete matrix, can they be transported.

The development of complexants that could selectively solubilize the "target" contaminant metals was seen to be a strategically vital aspect of this program.

For almost ten years, ISOTRON® scientists have worked on electrical migration of radionuclides through soils, groundwater, concrete, and polymeric materials. This experience provided the requisite insight needed to deal with the technological challenges of dissolution kinetics, as well as transport kinetics related to contaminants in concrete.

ISOTRON® scientists are highly focused on this subject of radionuclide desorption from mineral surfaces. This work involves cooperation with recognized experts in USA, including scientists at Oak Ridge National Laboratory and Los Alamos National Laboratory. The ISOTRON® specialists are also working in close collaboration with Russian scientists who are specialized in this field. Over the past four years, this USA/Russian scientific collaboration regarding this electrokinetic technology involved scientists from the Russian Firm "Radon", Russia's largest processor of radioactive waste materials, Russian Institute (VNIPIPT), Russia's Institute of Mining and Metallurgy (located in Moscow), Russian Institute of Agricultural Radio-Ecology (located in Obninsk), and the Russian organization ENERGOPOOL which is a nuclear equipment design organization (located in Moscow). This collaboration also involves Former Nuclear Weapons scientists from the installations Chelyabinsk, Mayak, and Arzamas.

The ISOTRON® staff and collaborators have, in general, approached this program with an emphasis on understanding the sorption mechanisms which are at work within the concrete matrix and which result in the "capture" of contaminants. Conversely, the technology related to ligand enhanced dissolution has emerged as a significant factor in the efficiency of the process. The selective desorption or dissolution of contaminants is recognized as an important consideration in process optimization.

The optimization of the electrolyte design has been approached with the following success criteria:

1. <u>The electrokinetic transport</u> rate of target contaminants (higher is better).

2. <u>Dissolution kinetics</u> – the ISOTRON® studies have shown that the cleanup rate is controlled by the kinetics of complexant aided dissolution. (Theoretical analysis reveals that this extraction process may be either transport controlled or dissolution controlled.)

3. Selectivity for contaminants – removing ions that are not necessary for cleanup is undesirable. Such a condition burdens the process with parasitic power demands, excess electrolyte consumption, and unnecessary waste volume.

4. Environmental (atmospheric) considerations – residual solubilizer components are to be benign.

In general, ISOTRON® scientists have concluded that one of the more effective electrolytes which can be recommended for this application is carbonate.

Project Description:

Development of an electrokinetic extraction process for reversing the mechanics of concrete contamination, whereby the contaminants are selectively desorbed from concrete and migrated out of the concrete, and subsequently collected for disposal.

Results:

Uranium is the target contaminant which was identified for this demonstration at K-25 Site in Oak Ridge. The ELECTROSORB® electrokinetic extraction process has been used to extract uranium from bare concrete floors associated with storage vaults which had been used in various services over a forty-year life span.

The uranium contamination was not uniform. Consequently, removal of uranium was also not uniform. Removal rates range from 0.1 grams per square foot up to a high of 1.1 grams per square foot.

A major success criterion for this process is the efficient recycling of electrolyte.

ISOTRON® scientists have dealt with this problem by means of its electrokinetic separation module. The following illustration describes its function.

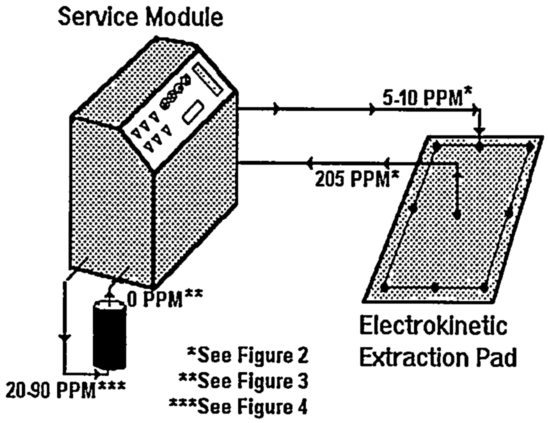

Figure 1

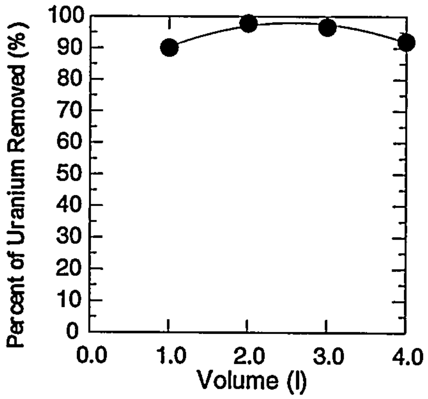

Figure 2

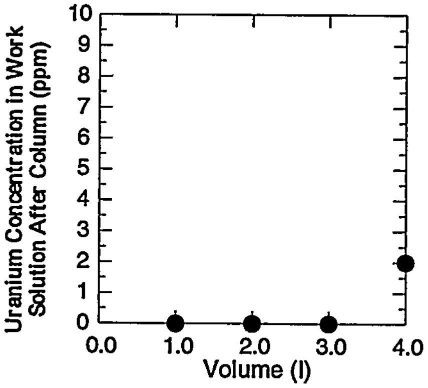

Figure 3

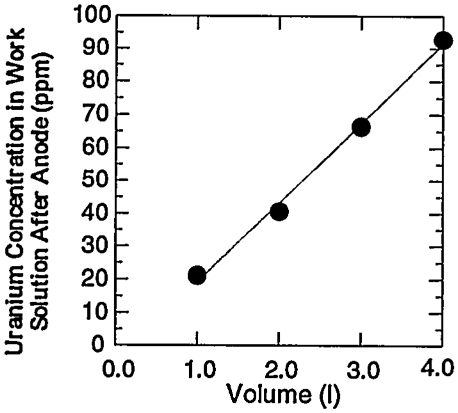

Figure 4

Application:

This application is designed for use in decontamination and decommissioning activities, wherever an extraction pad can be brought into contact with a contaminated surface.

Future Activities:

ISOTRON® staff is preparing for a pilot scale demonstration in October/November 1995.

An independent third party contractor will participate. This contractor will be responsible for the characterization of the demo area prior to decon and after decon. This contractor will gather verification data on power consumption, chemical usage, waste volume, and similar related operational information.

Future work is planned to develop more experience with electrolytes for removing radionuclides such as cesium, strontium, plutonium.

Work is anticipated that will investigate the use of high energy pulsed power to enhance overall process efficiency, and at the same time, a shortened cleanup cycle duration.

Work to simplify and to improve the functional controls and automation of the "ESM" is anticipated.

Acknowledgments:

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PI.3 Decontamination of Process Equipment Using Recyclable Chelating Solvent

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Introduction

The Department of Energy (DOE) is now faced with the task of meeting decontamination and decommissioning obligations at numerous facilities by the year 2019. Due to the tremendous volume of material involved, innovative decontamination technologies are being sought that can reduce the volumes of contaminated waste materials and secondary wastes requiring disposal. With sufficient decontamination, some of the material from DOE facilities could be released as scrap into the commercial sector for recycle, thereby reducing the volume of radioactive waste requiring disposal.

Although recycling may initially prove to be more costly than current disposal practices, rapidly increasing disposal costs are expected to make recycling more and more cost effective (Reference 1). Additionally, recycling is now perceived as the ethical choice in a world where the consequences of replacing resources and throwing away reusable materials are impacting the well-being of the environment.

Current approaches to the decontamination of metals most often involve one of four basic process types: (1) chemical, (2) manual and mechanical, (3) electrochemical, and (4) ultrasonic (Reference 2). "Hard" chemical decontamination solutions, capable of achieving decontamination factors (Df's) of 50 to 100, generally involve reagent concentrations in excess of 5%, tend to physically degrade the surface treated, and generate relatively large volumes of secondary waste. "Soft" chemical decontamination solutions, capable of achieving Df's of 5 to 10, normally consist of reagents at concentrations of 0.1 to 1%, generally leave treated surfaces in a usable condition, and generate relatively low secondary waste volumes.

Under contract to the Department of Energy, the Babcock & Wilcox Company is developing a chemical decontamination process using chelating agents to remove uranium compounds and other actinide species from process equipment. This paper describes the technical approach taken to achieve project objectives, and briefly discusses results obtained to date.

Research sponsored by the U. S. Department of Energy's Morgantown Energy Technology Center, under Contract DE-AC21-93MC30168 with Babcock & Wilcox, a McDermott Company, 1562 Beeson Street, Alliance, OH 44601; telefax (216) 823-0639.

Objective

This work is being performed in response to DOE's request for research on methodologies for the decontamination of process equipment. At the time of project initiation, DOE offered equipment from the gaseous diffusion plant at Oak Ridge, Tennessee, for use in the full scale demonstration of the developed technology. For this reason, the selected technical approach was to develop a process suitable for application to process equipment contaminated in this manner.

Internal components of equipment utilized in the uranium enrichment process were in contact with uranium hexafluoride (UF_6) gas at elevated temperatures over many years of operation. It is suspected that inleakage of air during operations and after plant shutdown have resulted in deposits of other uranium compounds, possibly UO_2F_6 and UF_4. Additionally, occasional outleakage of process gas has left small radioactive deposits on external surfaces. The compressor internals are expected to not only have surface contamination, but, due to long exposures at high temperatures, it is anticipated that the radioactive contaminants have penetrated the grain boundaries of the base metal.

DOE's preliminary evaluation of the potential for recycling the compressor materials was that some equipment parts, such as the compressor base and rotor shaft, might be surface contaminated only, and, as such, releasable for unrestricted use under existing guidelines (eg. NRC Regulatory Guide 1.86, DOE Order 5400.5). Other parts, such as the compressor housing, rotor, and piping, were expected to be bulk contaminated with radionuclides, preventing unrestricted release and making the equipment attractive for reuse within the DOE complex. Requirements of the decontamination technology applied to the compressors were: (1) removal of as much contamination as possible in a cost effective manner, (2) minimization of the volume of secondary waste generated, and (3) minimization of worker exposure. The technical approach and test plan for this program were developed in accordance with this information.

Approach

The use of chelating agents for the chemical cleaning and decontamination of nuclear steam generating equipment is well known. Ethylenediaminetetraacetic acid (EDTA)-based solvents are currently state-of-the-art technology for the dissolution of magnetite (Fe_3O_4), copper, and other metals and metal oxides from the secondary side of nuclear steam generators.

Technologies developed for the decontamination of commercial nuclear power reactors (eg. LOMI and CANDEREM processes) also utilize chelating agents; these processes have been tailored for the removal of metal oxides which are radioactive either from having spent time in-core or from incorporation of radioactive ions in solution into the oxide lattice as it grows on the metal surface. Thus, the vast majority of information available on cleaning and decontamination using chelants does not specifically apply to the removal of actinides.

The approach taken in this program was to build on the chemical cleaning expertise acquired by Babcock & Wilcox during the development and qualification of the Electric Power Research

Institute/Steam Generator Owners Group (EPRI/SGOG) chemical cleaning process (Reference 3). The mechanism of the technology proposed for development was similar to that of the referenced process, and entails the formation of stable complexes between the chelant and contaminant species. When used appropriately, chelating agents can effectively stabilize a metal ion in solution. Stabilization of the metal ion shifts the metal to ion equilibrium and allows chelating solutions to continuously promote dissolution of metals into the solution phase (Reference 4).

Project Description

The goal of this program is to develop a solvent capable of selectively chelating uranium and other actinides. This selectivity is required to eliminate the potential for the chelating capacity of the solvent to be exhausted by metallic species, such as iron, which might be present in overpowering quantities.

A second requirement of the process is the ability to control corrosion of the base metal. In cases where materials are surface contaminated only, and reuse of the equipment is a possibility, corrosion should be controlled at a low rate. However, in cases where contamination has migrated into the grain boundaries of the base metal, a more aggressive solvent is required, to corrode away base metal and expose the radioactive species for removal.

The third requirement of the process is that the spent solvent can be regenerated through destruction of the contaminant-chelant complex, thereby regenerating the chelant for subsequent reuse and reducing the volume of secondary waste generated.

Based on these requirements, a test program was developed that included: laboratory-scale chelant screening tests, to identify solvents capable of solubilizing uranium; refinements tests, in which process application parameters were varied to optimize solvent effectiveness; solvent recycle tests, aimed at development of a process to break the chelant-metal bond and free the chelant for reuse; bench scale testing, involving application of the developed solvent system under dynamic conditions simulating field application; and a pilot plant study, in which the process developed and refined in previous tasks was applied, on a pilot scale, to actual contaminated scrap material. This Phase I activity has been completed. Planned activity for Phase II of this project includes a field demonstration of the developed technology at a DOE site.

Results

Chelant Screening Tests (Task 2.1)

The goal of this task was to perform laboratory screening tests to identify the best solvent for the selective chelation of uranium. As a first part of this task, a literature search was conducted to select organic chelating agents for screening. The criterion for selection was the ability of the chelant to complex the various forms of uranium expected to be present on the compressor surfaces.

Attempts were made to procure several chelating agents described in the literature as forming stable complexes with various forms of uranium. The majority of these compounds were not available for purchase from organic chemical vendors, or were too expensive for practical application to decontamination requirements of the magnitude identified. A third consideration in the selection of candidate chelating agents was the status of EPA approval for utilization of the chemical in the Phase II site demonstration.

Based on these considerations, the screening tests were initiated using three chelating agents that had been previously applied to the chemical cleaning of power generating equipment, namely EDTA (ethylenediaminetetraacetic acid), HEDTA (hydroxyethyl-ethylenediaminetriacetic acid), and DTPA (diethylenetriaminepentaacetic acid). All three of the chelants are known to form strong complexes with uranium.

The purpose of the screening tests was to select two or three solvents for refinement testing, based strictly on their ability to chelate uranium. The deposit material chosen for dissolution testing was uranium dioxide (UO_2), since it was felt to represent one of the least soluble contaminants potentially present.

Each candidate chelant was subjected to several screening tests, run at varying pH's. Other conditions, including chelant concentration, temperature, and exposure time, were held constant throughout initial stages of the screening test program. The tests were conducted in three-liter four-neck reaction flasks. A schematic of a typical solvent screening test flask arrangement is shown in Figure 1. The procedure for testing involved addition of a known quantity of uranium dioxide to the test flask, followed by exposure to solvent for eight hours at 93°C in a controlled temperature water bath.

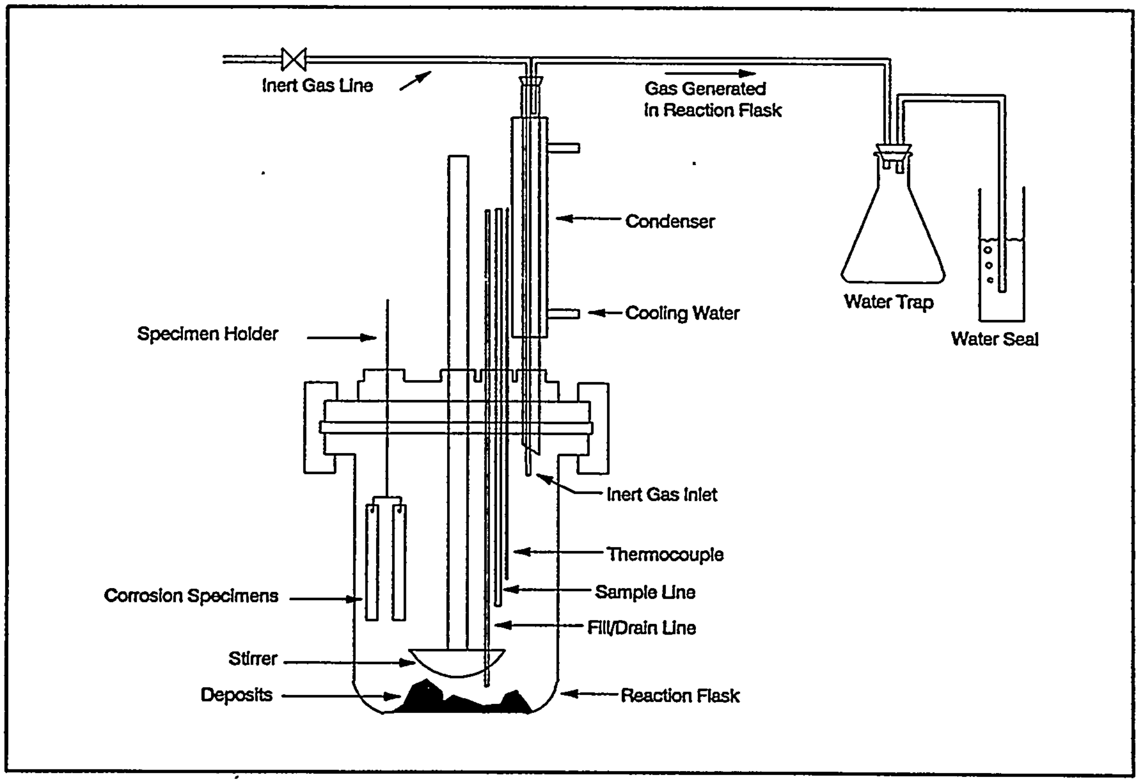

Figure 1 - Typical Screening Test Flask Arrangement

Periodically, samples of the solvent were removed from the test flask for analysis for dissolved uranium and for available chelant. At the conclusion of the test, the entire solution remaining in the test flask was filtered, and the filtered material was dried and weighed to evaluate the completeness of UO_2 dissolution.

Each of the three candidate chelants (EDTA, HEDTA, DTPA) was initially tested in this way at a pH of 5.5, 7.0, and 9.0. The results were similar for all tests; only a small amount of the uranium dioxide added to the test flask (1%-4%) dissolved, generally during the first few hours of exposure. Further tests were then performed at a lower pH (2.5-3.2), achieved by adding citric or formic acid to the solvent formulation. These organic acids have also been used in the chemical cleaning industry successfully for removal of deposits from power generating equipment. These formulations, however, showed no improvement in uranium dissolution.

A decision was subsequently made to test chelants in conjunction with hydrogen peroxide and carbonate salts. The combination of peroxide and carbonate has been applied to the dissolution of uranium compounds in the mining and chemical cleaning industries (Reference 5).

The testing was performed at room temperature, due to the known potential for violent reaction between peroxide and organic materials. The combination of chelant, ammonium carbonate, and hydrogen peroxide at a pH of 9.0 (adjusted with ammonium hydroxide) was found to effect virtually 100% dissolution of the uranium dioxide placed in the test flask, within the first two hours of exposure. Results were equivalent for each of the three tested chelating agents.

This type of solvent appeared to be promising for several reasons:

(1) Since uranium dioxide was dissolved effectively at room temperature, the solvent could be utilized in surface decontamination applications without interference from chelation of iron, which does not occur readily at low temperatures.

(2) Since EDTA is known to be corrosive to metals at higher temperatures, the solvent could be applied in such a way as to remove base metal, thereby releasing contamination trapped within the grain boundaries.

(3) The addition of carbonate and hydrogen peroxide did not complicate the secondary waste stream, since these are not hazardous materials.

Further testing was then performed using an EDTA/ammonium carbonate/ hydrogen peroxide solvent. EDTA was selected as the most favored of the three candidate chelants based on its lower cost and the large information base regarding its application, developed for chemical cleaning applications. Several tests were performed to determine the effect of the chelant on the dissolution process. Results indicated that dissolution was approximately 95% complete using a solvent containing peroxide and carbonate only; dissolution occurred slightly more rapidly, and to 100% completion, with the solvent containing chelant.

The effect of carbonate on the dissolution process was also evaluated. A solvent containing just chelant and peroxide was found to dissolve only 16% of the uranium dioxide placed in the test flask. These results indicated that complexation of the uranium by carbonate was essential to the dissolution process. The effects of chelant and carbonate on uranium dissolution are shown graphically in Figure 2.

These results indicated that a carbonate/peroxide solvent might be effective for removal of light surface uranium contamination. The addition of chelant would be required for decontamination of surfaces characterized by heavier deposits, to stabilize the uranium ions in solution and prevent precipitation from occurring during the cleaning process. Chelant would also be required to effect base metal corrosion for release of bulk contamination.

Solvent Refinement Testing (Task 2.2)

At this stage of screening, it was decided that two solvents would be carried forward to solvent refinement testing. Those selected for development were:

(1) EDTA, ammonium carbonate, and hydrogen peroxide: pH 9.0 with NH_4OH, and

(2) ammonium carbonate and hydrogen peroxide: pH 9.0 with NH_4OH.

The purpose of refinement testing was to vary solvent parameters in an attempt to optimize solvent effectiveness. Parameters investigated included solvent constituent concentrations and application scenarios. The evaluation of solvent performance considered the rate of

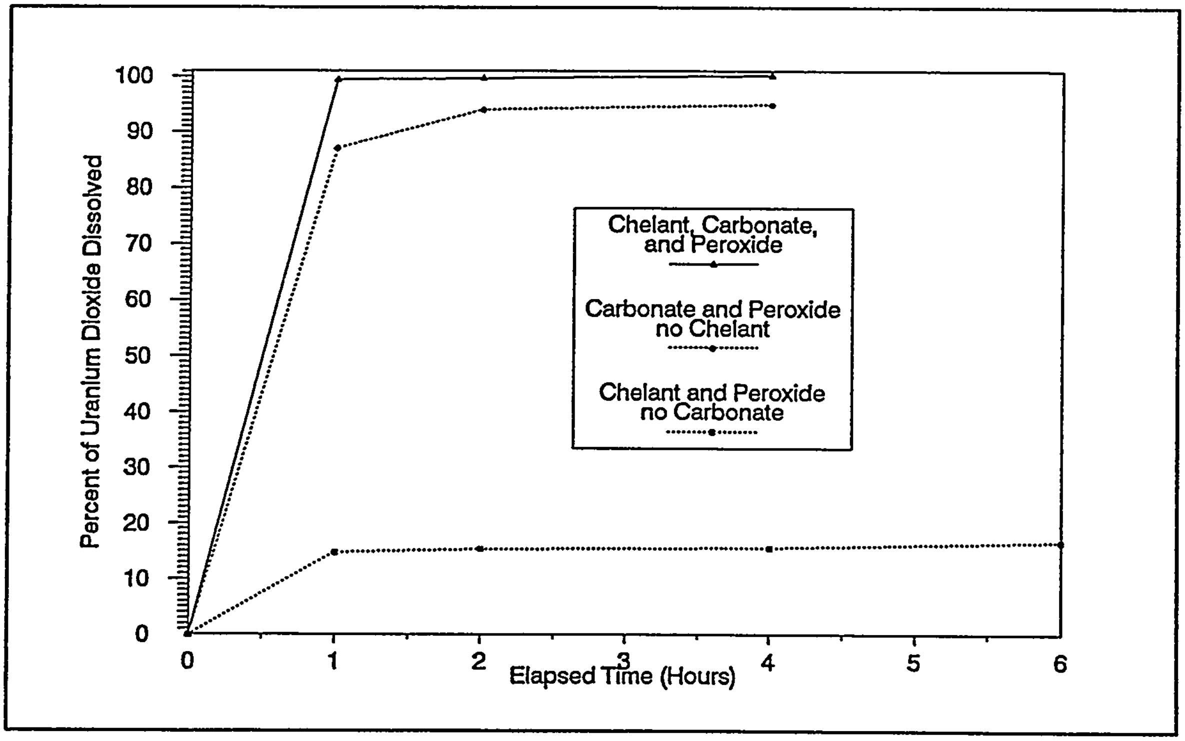

Figure 2 - Effect of Solvent Composition on Uranium Dissolution

uranium dissolution, extent of dissolution, and the amount and type of corrosion to materials of interest.

Refinement tests were run using the same equipment arrangement and utilizing similar procedures as the screening tests.

Since refinement testing was also designed to characterize the corrosivity of both solvents, preweighed specimens of carbon steel, Monel, and nickel were placed in the reaction flask for the estimation of general corrosion by weight loss.

Initial tests were performed to determine the concentration of each constituent required to dissolve uranium dioxide effectively. Testing with varying solvent compositions showed that the basic solvent system was effective over a wide range of constituent concentrations.

However, the importance of tailoring the solvent system to process application conditions was demonstrated. Specifically, the surface area of metal exposed during testing affected uranium dissolution; increased metal content in the system necessitated an increase in the initial H_2O_2 concentration for complete dissolution to occur. These results showed that the metal surface area to solvent volume ratio must be factored into solvent formulation as it is tailored for a specific application. This information is shown graphically in Figure 3.

A secondary part of refinement testing was to evaluate the feasibility of applying the selected solvents in the foam state. Foam decontamination technology is currently being evaluated by DOE, as it has shown waste reduction of 70% over present decontamination methods (Reference 6).

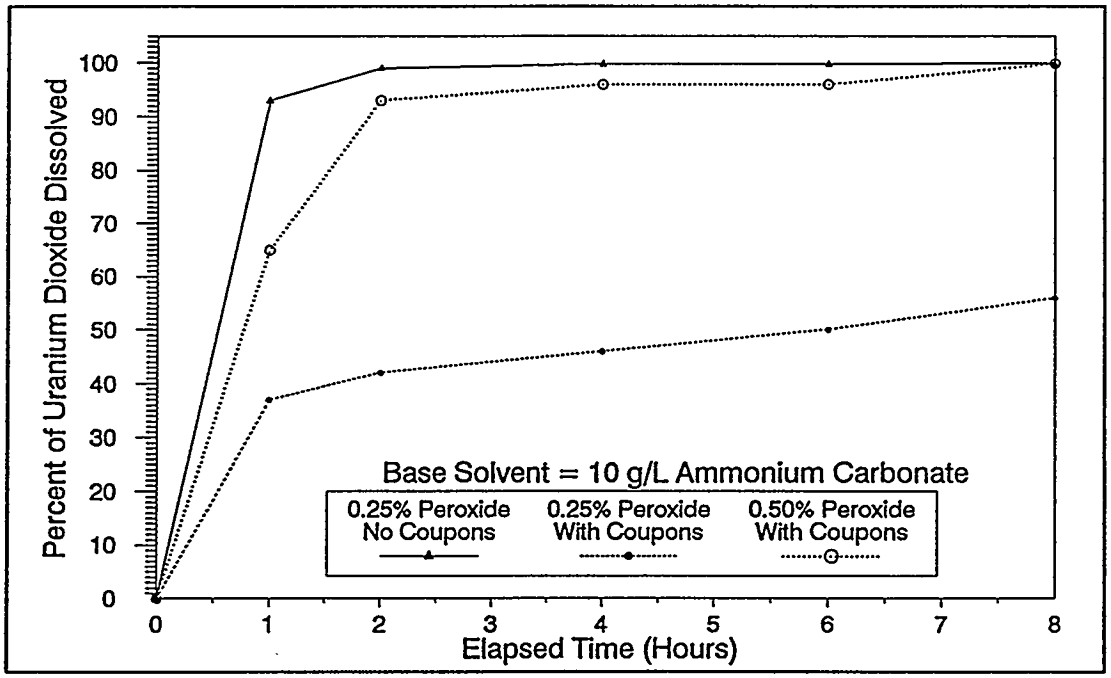

Figure 3.
Effects of Coupons and Peroxide Concentration On Uranium Dissolution

A paste of uranium dioxide was dried onto coupons, which were subsequently exposed to the foamed EDTA/carbonate/ peroxide solvent. The tests showed that UO2 could be dissolved by the solvent in the foam state, although some of the deposit was dissolved after it had dislodged from the coupon and dropped into the condensed foam. Further testing of foamed solvent with more appropriate test samples (material contaminated in service, if available) has been recommended.

In the final phase of refinement testing, process application testing, various application scenarios were simulated to evaluate their effect on dissolution and corrosion. The results of these final tests were used to develop the recommended process for bench scale and pilot scale testing. Based on the results of testing, the following process application scenario was chosen for application in bench- and pilot-scale testing:

- Two hours at room temperature
- Heatup to 93°C and hold for four hours
- Cool to room temperature and inject peroxide
- Two hours at room temperature

The rationale for this application scenario is that readily available uranium will be dissolved during the initial low temperature exposure. The four-hour 93°C exposure will result in corrosion of base metal, thereby releasing contamination trapped in grain boundaries. This released contamination will be dissolved by the final low temperature exposure.

Using this application sequence for the EDTA/carbonate/peroxide solvent, 100% of available UO$_2$ was dissolved and a carbon steel corrosion rate of 1.4 mils in four hours was achieved. These results exceeded the established success criteria for the solvent developed in this task, those being 90% dissolution of uranium present in the system in less than eight hours, and capability of a carbon steel corrosion rate greater than two mils per eight hours. These results are shown graphically in Figure 4.

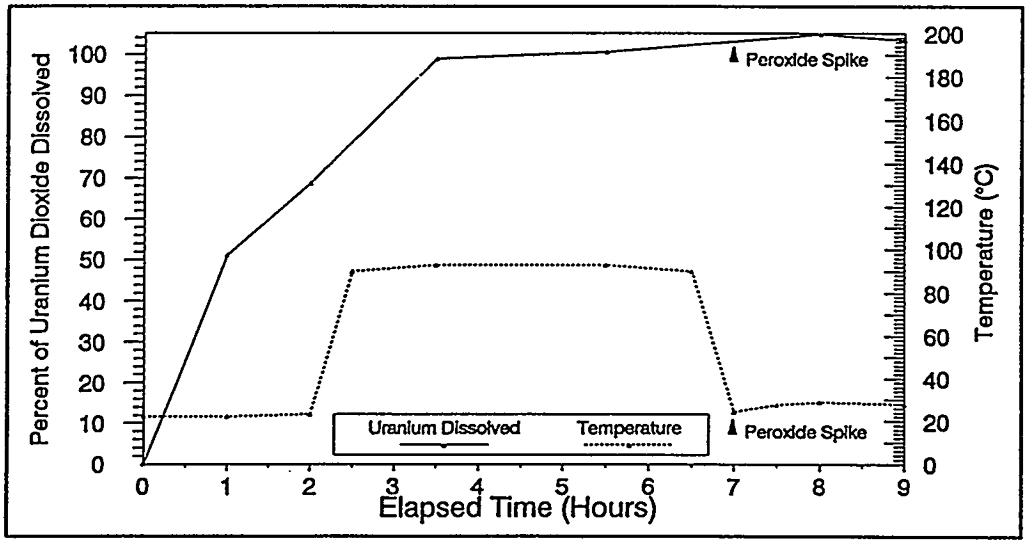

Figure 4
Chelant/Carbonate/Peroxide Solvent
Room Temperature → 93°C → Room Temperature

Solvent Recycle/Contaminant Reduction Testing (Task 3)

As mentioned previously, one requirement of the developed process is recycling of the spent solvent. After the decontamination process has been performed, removed radionuclides will be present as dissolved species in the chelating solution. The objective of this task was to develop a process to break the chelant-contaminant bond, and to precipitate the contaminants as the insoluble metal hydroxides. The end result was to be a filter cake of contaminants and a regenerated chelant solution.

Methods for recycling both the EDTA/carbonate/peroxide and EDTA/ carbonate solvents were evaluated. Spent solvents from Task 2 of the program were used for this testing. Dissolved uranium content of the test solutions before and after treatment was used to evaluate the effectiveness of each recycle process tested.

The EDTA/ carbonate solvent recycle test program resulted in five successful methods for removing uranium from the spent solvent with an efficiency of greater than 99.9%:

1. acidification to pH 1 followed by addition of lime and caustic to pH 12.5
2. pH reduction to 1.0 followed by application of hydrogen-form strong acid cation resin
3. application of carbonate-form strong base anion resin at existing solvent pH
4. application of hydroxide-form strong base anion resin at existing solvent pH
5. precipitation by application of trisodium phosphate after reducing solvent pH to 2.

The process utilizing anion resin in the carbonate form appeared to be most viable, because it removed only uranium from solution. All other processes were acceptable and considered equal.

Only one method was found to be successful with the EDTA/carbonate/ peroxide solvent. This method consisted of reducing solvent pH to less than 2, followed by application of a uranium selective cation resin in the hydrogen form. No precipitation techniques provided acceptable results with the EDTA/carbonate solvent system.

Bench Scale Testing

The purpose of bench scale tests was to evaluate the solvent system developed in the solvent refinement test under dynamic test conditions that simulate the actual field application. The tests were conducted in the B&W Alliance Research Center's low temperature chemical cleaning velocity test apparatus. Ceramic plugs containing uranium dioxide and corrosion coupons of nickel, carbon steel, and Monel were loaded into a sealed reaction chamber. Solvent was circulated through the reaction chamber at a velocity approaching one foot per second. Samples were taken on a periodic basis and were analyzed for dissolved uranium, iron, copper, nickel, and available chelant.

The first phase of the task was development of a suitable binder for making UO_2 test plug that would release uranium slowly throughout the dynamic exposure, in order to simulate the cleaning of scrap material with uranium contamination embedded in base metal grain boundaries. Plugs consisting of various combinations of calcium carbonate, trisodium phosphate, silicon dioxide, and mortar, were evaluated with both solvents. For the first portion of bench scale testing, uranium dioxide was added to a formulation that appeared to produce a plug deteriorating slowly throughout the test exposure.

Results of initial testing indicated that the plugs were more robust than testing without UO_2 in the formulation had indicated, and decomposed at a significantly slower rate than anticipated. Additional testing was performed using smaller plugs made from a formulation with an increased ratio of uranium dioxide to binder.

The application scenario utilized in the bench-scale testing was that qualified in the refinement testing:

- Two hours @ room temperature
- Heatup to 93°C; hold four hours
- Cooldown to room temperature and inject peroxide
- Hold @ room temperature two hours.

Initial testing consisted of two tests using the original plug formulation; one test with each of the two candidate solvents. The test plugs used did not disintegrate to the extent predicted by plug formulation testing. For this reason, some of the encapsulated uranium did not become accessible to the solvents, and, hence, was not dissolved. The EDTA/carbonate/peroxide solvent dissolved 25% of the added uranium; the carbonate/peroxide solvent dissolved about 18%.

As mentioned above, the plugs were then reformulated for additional bench scale testing. Two tests were run with the smaller, modified-formulation plugs; one test with each of the two candidate solvents. The EDTA-containing solvent attacked the reformulated plugs more aggressively than it did the original formulation; 75% of the added uranium was dissolved. The carbonate/peroxide solvent dissolved only 10% of the added uranium. Based on the low dissolved calcium values observed, the revised plug formulation appeared to be very resistant to this solvent.

The corrosion losses on test specimens included in all four tests were uniform, with no localized attack evident. The application of the carbonate/peroxide solvent resulted in negligible corrosion of all three materials; application of the EDTA-based solvent resulted in average corrosion of 0.2 mils for Monel 400, 0.3 mils for Nickel 200, and 0.4 mils for carbon steel. It was thus shown that the addition of chelant is required to effect consistent base metal corrosion, which will be required for release of contamination below the surface of process equipment being decontaminated.

Pilot Plant Study

Upon completion of bench scale testing, a pilot study of the developed process was conducted at B&W's Lynchburg Technology Center. The purpose of this study was to verify operating parameters for the decontamination system to be used in the field demonstration, as well as to demonstrate the success of the process in decontaminating actual contaminated process equipment. The testing was performed in a skid mounted pilot-scale test rig; a schematic of the equipment appears as Figure 5. The study was conducted using scrapped process equipment characterized by contamination felt to be representative of that to be cleaned in the Phase II field demonstration. The contaminated metal consisted of stainless steel pipe brackets and several pieces of steel floor grating.

Three tests runs were made with the pilot system: two with the EDTA/carbonate/ peroxide solvent and one with the carbonate/ peroxide formulation. The application scenario utilized was the same as that applied in bench-scale testing. All stainless steel components subjected to the cleaning solvent showed a decontamination removal efficiency of greater than 99%. Decontamination factors for the carbon steel components subjected to the cleaning solvents were considerably less; however, a large percentage of the carbon steel surface areas subjected to the solvent were covered with a tar-like coating. It is believed that the coating shielded the uranium from the solvent, thereby reducing the effectiveness of the cleaning.

The pilot scale solvent recovery tests were also quite successful. EDTA was recovered from solution by precipitation and the dissolved uranium was reduced by ion exchange to a level between 11 and 144 pCi/L for the four (4) batches of waste solvent treated. This was equivalent to a uranium removal of greater than 99.4% for all of the waste processed. Iron was also removed by the cation exchange column to acceptable discharge levels.

Process application modifications to the test rig required prior to a field demonstration were identified during pilot tests. These include improved cooling capabilities during hydrogen peroxide spikes and control of solvent aeration, to reduce the ferric ion corrosion occurring during process application. In addition, the need for real-time peroxide analysis was also identified.

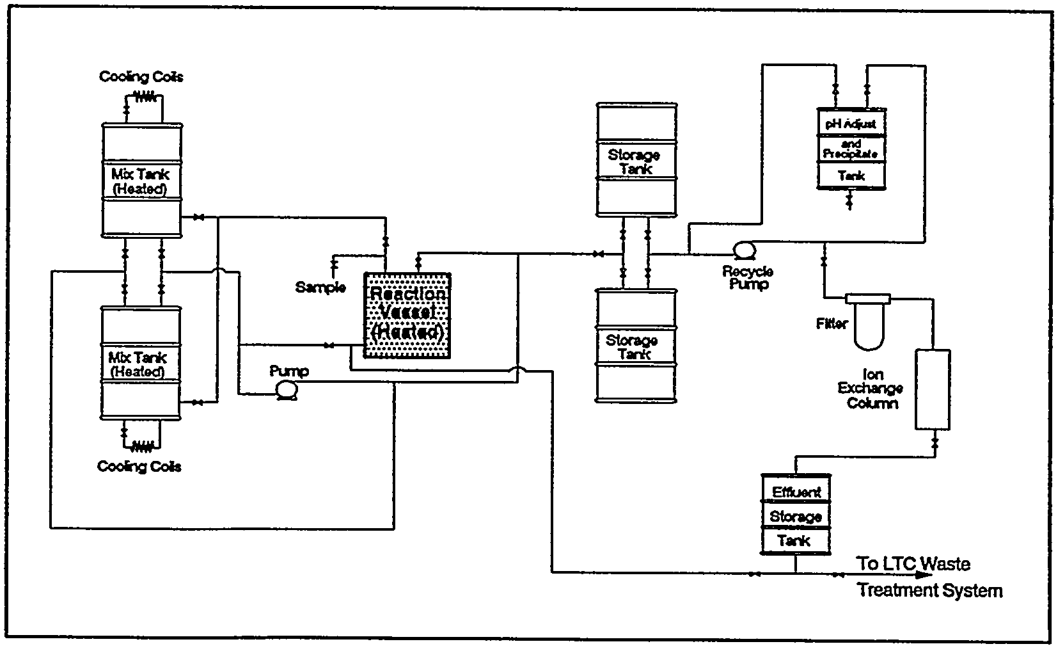

Figure 5 - Schematic of Pilot Scale Test Rig

Application

Based on the results of Phase I testing, two solvent systems are concluded to provide optimum uranium dioxide dissolution:

(1) EDTA, ammonium carbonate, and hydrogen peroxide: pH 9.0 with ammonium hydroxide

(2) Ammonium carbonate and hydrogen peroxide: pH 9.0 with ammonium hydroxide.

Since UO_2 is dissolved effectively by these solvents at room temperature, they can be utilized in surface decontamination applications without interference from chelation of iron, which does not occur readily at low temperatures.

Essentially no corrosion is observed without EDTA being present in the solvent system. The EDTA is required to effect consistent base metal corrosion for release of radioactive material below the surface of equipment requiring decontamination. The chelant also appears to stabilize the dissolved uranium in the presence of competing ions in solution.

From the information generated in Phase I, it is projected that the carbonate/peroxide solvent should be effective for removal of surface uranium contamination with a single low-temperature application. The addition of chelant will be necessary for decontamination of surfaces characterized by uranium contamination buried deeper in metal grain boundaries. In such cases, the presence of EDTA results in the necessary higher base metal corrosion during the 93°C application phase of the decontamination process.

Potential also exists for application of the solvents in the foam state, which would be useful for decontamination of large components. Further evaluation of a foaming decontamination process using these solvents has been proposed as part of Phase II.

Future Activities

Planned activity for Phase II of this program is a full scale demonstration of the developed decontamination and solvent recycle technology. Initially, the site demonstration was to take place at the gaseous diffusion plant at Oak Ridge. However, due to the level of enrichment of the uranium contamination represented, there were criticality concerns associated with the used of aqueous solvent systems for decontamination. For this reason, during the course of Phase I activity, efforts were made to identify a DOE site with a specific need for the developed technology, willing to sponsor a demonstration.

One site that meets these criteria is Fernald, which has accumulated an inventory of nuclear material, low-level radioactive waste, mixed wastes, and contaminated equipment/facilities. As a result of years of uranium metal production, equipment, buildings, soil, and ground water at this site have become contaminated and have the potential to impact public health and the environment.

The uranium contamination levels at Fernald are amenable to support the proposed project. Site conditions there are well characterized and documented to support completion of the test program. In addition, B&W's Nuclear Environmental Services, Inc., which will perform the field cleaning, already has a trained field staff at this site. Prior environmental tests conducted by B&W NESI in cooperation with FERMCO at this site have received excellent support from FERMCO. For these reasons, B&W has recommended the Fernald site as the location for the Phase II demonstration.

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PI.4

Alpha Detection in Pipes Using An Inverting Membrane Scintillator

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Abstract

Characterization of surface alpha emitting contamination inside enclosed spaces such as piping systems presents an interesting radiological measurement challenge. Detection of these alpha particles from the exterior of the pipe is impossible since the alpha particles are completely absorbed by the pipe wall. Traditional survey techniques, using hand-held instruments, simply can not be used effectively inside pipes. Science and Engineering Associates, Inc. is currently developing an enhancement to its Pipe Explorer™ system that will address this challenge.

The Pipe Explorer™ uses a unique sensor deployment method where an inverted tubular membrane is propagated through complex pipe runs via air pressure. The inversion process causes the membrane to fold out against the pipe wall, such that no part of the membrane drags along the pipe wall. This deployment methodology has been successfully demonstrated at several DOE sites to transport specially designed beta and gamma scintillation detectors into pipes ranging in length up to 250 ft.

The measurement methodology under development overcomes the limitations associated with conventional hand-held survey instruments by remotely emplacing an alpha scintillator in direct contact with the interior pipe surface over the entire length to be characterized. This is accomplished by incorporating a suitable scintillator into the otherwise clear membrane material. Alpha particles emitted from the interior pipe surface will intersect the membrane, resulting in the emission of light pulses from the scintillator. A photodetector, towed by the inverting membrane, is used to count these light pulses as a function of distance into the pipe, thereby producing a log of the surface alpha contamination levels. It is anticipated that the resulting system will be able to perform measurements in pipes as small as two inches in diameter, and several hundred feet in length.

This paper presents the design goals of the Pipe Explorer™ alpha detection system, the

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technical approach adopted, and preliminary laboratory test results.

Introduction

The DOE is currently decommissioning and dismantling many of its nuclear materials processing facilities that have been in use for several decades. When dismantling these facilities, the DOE must act conservatively to protect the workers and the general public from inadvertent exposure to residual radioactive materials. The DOE has established policies and guidelines governing the handling of radioactive materials to ensure that an appropriate level of conservatism is consistently applied. The DOE Radiological Control Manual[1] is the primary document that establishes acceptable practices for DOE radiological control activities.

In developing work plans for these decommissioning activities, it is necessary to identify those areas where residual radioactive contamination is likely to occur, and to characterize the actual contamination that will be encountered during the clean-up activities. Traditional health physics instrumentation and procedures can be employed in most circumstances to adequately characterize such radiological contamination.

Most of these facilities, however, have extensive networks of pipes and ducts that have been used to transport radioactive materials, either intentionally or inadvertently. In some instances the isotopes and their abundances are well known, while in other cases little, if any, such information is available. Where accurate information is lacking these systems must be characterized through some kind of measurement process. If the contamination includes isotopes with significant gamma emissions, traditional health physics instrumentation can be employed from the exterior of the pipe or duct work to adequately characterize the residual contamination.

In other cases the processes were such that the residual contamination may be composed of only alpha emitters. Alpha particles are very densely ionizing and thus do not penetrate most materials more than a few tens of microns. Therefore, it is not possible to detect the presence of alpha radiation from the exterior surface of the pipe or duct. So, direct measurements of surface alpha contamination must be made from the interior, where the alpha particles may interact directly with a detector. The very nature of the pipe or duct work restricts access by traditional instrumentation to the proximity of existing openings in the system. This makes it impossible to survey the entire interior surface, which must be accomplished if it is to be adequately characterized.

In addition to the outright cost savings associated with not having to pay for disposal of materials as if they were radioactively contaminated, there are ancillary benefits to conducting these types of characterization efforts. For example, if facility process knowledge indicates that alpha emitting contaminants may be present at hazardous levels inside a piping or duct system and these levels can not be refuted through a radiological survey, then the conservative approach dictates that the worst must be assumed. This means that the workers must wear a personal protective equipment (PPE) ensemble commensurate with the assumed potential for personnel contamination. This adds, unnecessarily, to the cost of performing the work (cost of PPE, support personnel, and lower productivity, increased volumes of secondary rad-waste), as well as increasing the risk from other hazards, such as heat stress, limited mobility and vision, etc. Adequate preliminary characterization of the system would allow a more efficient work plan to be developed, requiring the higher levels of PPE only when and where they are really needed.

Additionally, if adequate characterization methods are available, it is then possible to consider decontamination of the systems in-place. This can result in significant cost savings in those circumstances where extensive excavation is required to extract the system.

The inverting membrane deployment technology employed by the Pipe Explorer™ offers a unique method of carrying radiation detectors into pipes and duct work for the purposes of characterizing residual surface beta and gamma contamination. Incorporation of an alpha measurement capability will greatly enhance the overall utility of the system.

The primary components of the Pipe Explorer™ technology and its function are shown in Figure 1. The heart of the system is an air-tight membrane that is initially spooled inside a canister. The end of the membrane protruding out of the canister is folded over and clamped to the canister outlet, which is inserted into the open end of the pipe to be entered. By increasing the air pressure inside the canister by several pounds per square inch (PSI) the membrane is forced into the pipe, inverting upon itself and unspooling as it does. This process continues until the membrane is completely off the spool. This end of the membrane is sealed and attached to any of several different detectors, which is in turn connected to a tether/signal cable, also spooled inside the canister. As the membrane continues to invert, the detector and tether/signal cable are pulled into the pipe following the membrane. In this manner the detector may be towed into the pipe a distance equivalent to the membrane length. The detector and tether/signal cable are retrieved by reducing the canister pressure and winding the tether/signal cable back onto the spool.

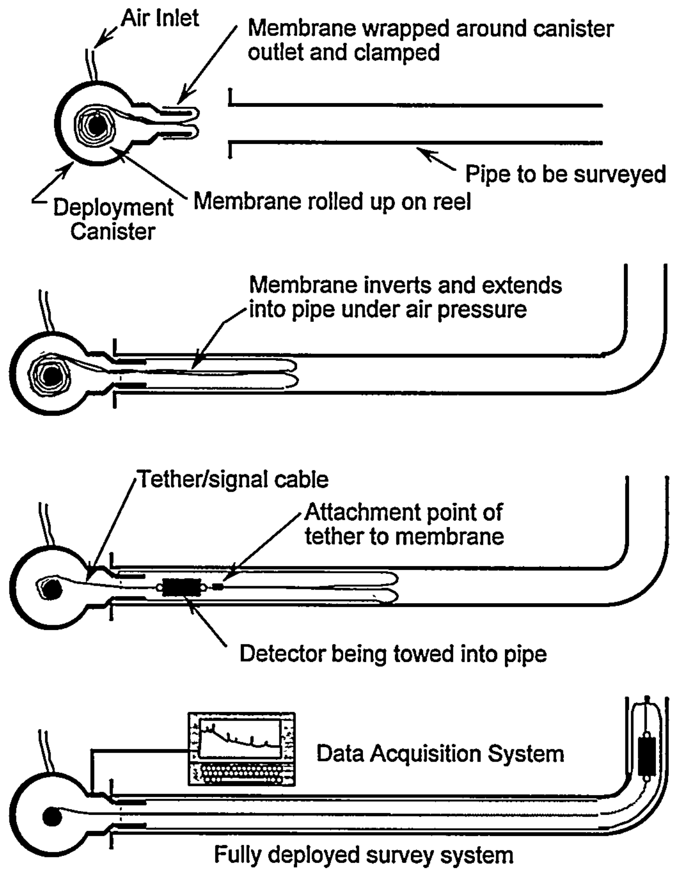

Figure 1. Sketch of the primary components of the Pipe Explorer™ system.

Objectives

The overall objective of this project is to extend the measurement capabilities of the Pipe Explorer™ to include characterization of interior surface alpha contamination of pipes and duct work. The completed enhancements to the system are expected to allow the following:

- Measurements of surface alpha contamination in pipes and ducts up to 250 ft. in length, and in a wide range of pipe diameters, 2-inch to 30-inch,
- Provide low minimum detectable activities for alpha emitters such as uranium, thorium and plutonium,
- Provide these measurements in a cost effective manner,
- Limit the potential for operator contamination to levels significantly lower than available with conventional techniques,
- Protect the detector and other expensive hardware components from potential contamination during the measurement process, and
- Limit the probability of the spread of contamination from the pipe or duct into surrounding areas.

Approach

The general approach taken for this project is to use existing, off-the-shelf, components and technology wherever possible to shorten the development time and reduce the development costs. To accomplish this and meet the performance objectives outlined above, the following technical approach to this measurement problem has been adopted.

A suitable scintillator material will be incorporated into the membrane. When such a membrane is deployed into a pipe it will be in intimate contact with the interior pipe wall, thereby placing the scintillator material into an ideal geometry with the potential surface contamination. Any alpha particles leaving the pipe wall will intersect the membrane and its incorporated scintillator. The interaction of the alpha particle and the scintillator will produce a small light pulse that is measurable by a sensitive photodetector, such as a photomultiplier tube (PMT). The PMT, equipped with a suitable light gathering fixture, will be towed down the pipe by the scintillating membrane as it inverts. This will allow a direct measure of surface alpha activity as a function of distance into the pipe.

The Scintillator - The scintillator must exhibit the following characteristics:

- Have a high light output (large number of photons emitted per alpha particle),
- Exhibit a good spectral match with existing high sensitivity photodetectors,
- Be relatively inexpensive,
- Be easily incorporated into a thermoplastic, such as polyethylene, and
- Must not be a hazardous material.

The high light output and spectral match are attributes that contribute to a high signal/noise ratio, which increases the sensitivity and reduces the minimum detectable activity (MDA) of the system. It is necessary to keep the cost of the membrane as low as possible, because the membrane will have to be disposable. Additionally, because the membrane will likely become contaminated, no hazardous materials can be used in its fabrication that would result in contaminated membrane material being considered a mixed-waste.

The Photodetector - The photodetector must have the following characteristics:

- High sensitivity and high signal/noise ratio,
- Good spectral match to the scintillator,
- Available in a variety of sizes, including one small enough to fit into 2-inch pipes,
- Good temperature characteristics, and
- Moderate tolerance to shock and vibration

The photodetector sensitivity, signal/noise ratio, and spectral match contribute to reducing the MDA of the system. Size of the detector is an important consideration given that many of the piping systems requiring characterization will be 2-inch. In order for the detector package to negotiate 90 degree bends, both the length and the diameter must be minimal. Measurements will likely be conducted in a variety of temperature environments ranging from stable indoor temperatures to the normal temperature extremes found outdoors. The photodetector must be capable of operating over these temperature ranges, as well as exhibiting a low temperature dependence of its response characteristics. And finally, the process of deploying a detector into a piping system requires that it be moderately resistant to the shocks that it will encounter negotiating the various bends and obstructions to be found in pipe systems and duct work.

The Light Gathering Fixture - The light gathering fixture must:

- Exhibit high efficiency in coupling the scintillator emissions to the photodetector,
- Be compact enough to fit into 2-inch pipes.

Due to the geometric arrangement of the scintillator and photodetector, it will be necessary to employ some kind of light gathering fixture. Figure 3 shows the basic arrangement of the scintillator, photodetector, and a light gathering fixture. This light gathering fixture must poses two main attributes. First, it must define a length segment of the pipe in which it gathers photons from scintillation events with a high efficiency, and correspondingly excludes light from adjacent sections of the pipe. Secondly, the fixture must not render the detector package so large as to be unable to negotiate the anticipated bends and obstructions.

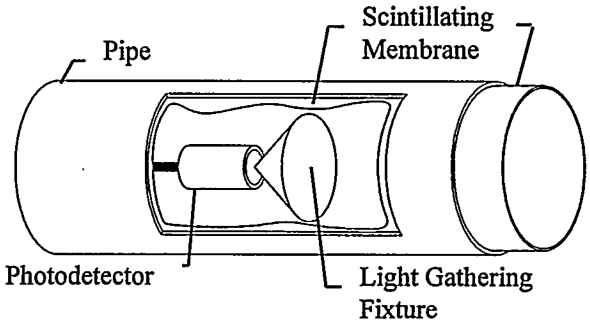

Figure 2. Sketch showing the arrangement of the photodetector, scintillating membrane, and light collection fixture.

Project Description

Two phases are planned for this project. Phase I will include development and laboratory testing of the prototype system. Phase II will include acquisition of membrane materials and field demonstrations. These two phases have been broken down into the following technical tasks. Tables 1 and 2 show the generalized schedule for Phase I and Phase II, respectively.

Phase I - Feasibility Evaluation

Task 1: Evaluate Scintillators/Photodetectors

The first task to be undertaken is to determine the best scintillator/photodetector combination for the alpha detection system from the available scintillators and photodetectors.. The criteria outlined above will be used in this selection process. In general more weight will be given to selection of the scintillator rather than the photodetector, because the performance characteristics exhibit greater variability for the

Table 1
Schedule of Tasks for Phase I
Prototype Design and Evaluation

	Task Description	1995						1996				
		Jul	Aug	Sep	Oct	Nov	Dec	Jan	Feb	Mar	Apr	May
1	Evaluate Scintillators/ Photodetectors	●	━	━	●							
2	Test Scintillator/Photodetector Combination					●	●					
3	Integrate and Test with Pipe Explorer^{TM}							●	━	━	━	●

Table 2
Schedule of Tasks for Phase II
Site Demonstrations

	Task Description	1996					1997								
		Aug	Sep	Oct	Nov	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sept
4	Membrane Acquisition and Testing		●	━	━	━	━	●							
5	Site Demonstrations								●	━	━	━	━	●	

available scintillators than for the photodetectors under consideration. Samples of the scintillator material will be obtained, as well as a photodetector and any support electronics required.

Task 2: Test Scintillator/Photodetector Combination

The next task will focus on an evaluation of the scintillator/photodetector combination selected in the first task. A key element of the evaluation process is the ease with which the scintillator can be joined with the membrane material, and what the physical and scintillation properties of the resulting membrane are. An attempt will be made to fabricate a series of test samples in which various amounts of scintillator are incorporated into a thermoplastic resin. The resulting test membrane samples will be evaluated for their strength properties and scintillation characteristics. In the event that this simple admixing of scintillator and resin does not produce a membrane with acceptable characteristics, a more complex and therefore more expensive, means of incorporating a scintillator rich layer into the membrane will be evaluated.

Task 3: Integrate and Test with Pipe Explorer^{TM}

This will be the final technical task necessary to complete Phase I of the project. The individual components will be integrated with the existing Pipe Explorer^{TM} hardware to form a complete measurement capability. A principal component of this task will be to develop a feasible approach for the light gathering fixture. The approach must allow for measurements in a variety of pipe and duct diameters. It may be necessary to develop more than one type of fixture/photodetector combination to cover the target range of diameters. The tether/signal cabling and associated electronic components will be adapted to integrate the photodetector/light gathering fixture(s). A calibration of the system response to surface alpha activity will be conducted for a variety of different pipe sizes. Several different sizes of the prototype scintillating membrane will be fabricated for use in laboratory tests of the integrated system with a range of pipe diameters. These tests will focus on the overall utility of the alpha measurement capability, and will be conducted using a plated Am-241 alpha source located at various positions along test pipe mock-ups. The results of these laboratory tests will be compiled and interpreted to establish the basic operation parameters of the measurement capability. This information will then be used to evaluate the cost effectiveness of the alpha measurement capability. A Phase I report and DOE review of the technology will occur at this point.

Phase II - Site Demonstrations

Task 4: Membrane Acquisition and Testing

Having identified the appropriate materials and fabrication methodology for the scintillating membrane in Phase I, this task will focus on identifying a manufacturer for the membranes. Once an appropriate manufacturer has been identified, a few standard sizes of the prototype membrane material will be procured in production quantities (e.g., 1,000 foot rolls). Following receipt of the membrane materials they will be evaluated for consistency in their scintillation and strength characteristics. Additional laboratory tests of the performance with the production materials will be performed. MDA levels, and deployment rates predicted from the Phase I testing will be confirmed.

Task 5: Site Demonstrations

Once the production membranes have been adequately tested, and the appropriate operational parameters have been established, the alpha measurement capability of the Pipe Explorer^{TM} will be ready to demonstrate at DOE sites. The initial site selection process will be initiated concurrent with the early stages of Phase I activities. Efforts will be made to locate sites that have an identified need for radiological characterization in either piping systems or duct work, so that the demonstration will serve not only to demonstrate the alpha measurement capability, but also to provide genuinely useful information to the site.

Once the sites for the field demonstrations have been determined, the process of planning the demonstrations may begin. This planning process will involve close coordination with project personnel at the sites to develop detailed test plans. These test plans will include the normal logistical coordination, delineate areas of responsibility, and develop measurement objectives. It is anticipated that two field demonstrations will be conducted at separate DOE sites. It is hoped that each site would posses distinctively different types of pipe systems or duct work with different contaminant situations in which the alpha measurement capabilities of the Pipe Explorer^{TM} could be exercised. A final report will be prepared that includes the measurement results from the field demonstrations as well as cost-benefit analyses.

Accomplishments

Since the start date of July 1995, the following progress has been made toward meeting the technical objectives of this project. Silver activated zinc sulfide, ZnS(Ag), has been selected as the scintillator material. Of the available materials that are characterized for their alpha scintillation properties, ZnS(Ag) exhibits one of the highest light outputs available. It has been widely used for many years as the alpha scintillator of choice, and therefore its behavior is well known. It is a non-toxic, non-hazardous material that is also routinely used as an electronic phosphor. Because of the large volumes of this material produced for the electronics industry, it is available at a reasonable cost (approximately $28/lb.) in large quantities. Moreover the industrial grade of ZnS (without the silver activator) is widely used as a coloring agent in polyethylene plastics. So there is a high degree of confidence that the electronic phosphor form can be easily incorporated into a standard low density polyethylene plastic suitable for blown film production.

Two types of photodetectors were considered for this application, photomultiplier tubes (PMTs) and avalanche photodiodes (APDs). A PMT equipped with a bialkali photocathode has been selected as the photodetector type. This selection was made for several reasons, but sensitivity was among the most significant. By selecting a higher sensitivity scintillator/photodetector combination the design requirements of the light gathering fixture are significantly eased.

In general the PMT is still the most sensitive photodetector available. The spectral response of a bialkali photocathode exhibits an almost ideal match to the emission spectrum of ZnS(Ag), thereby maximizing the overall sensitivity available from the scintillator/photodetector combination. APDs exhibit their highest sensitivity at much longer wavelengths, requiring a scintillator emitting in the red region to maximize their sensitivity. Additionally APDs exhibit significantly higher noise levels than PMTs, which for the low light levels that will be encountered in this application, result in poor signal/noise ratios. Another undesirable feature of APDs is the temperature dependence of both the dark current and the internal gain. While this can be overcome by the use of a thermoelectric cooler, this adds unnecessarily to the complexity and bulk of the detector package. Although the APD is inherently more rugged than the PMT, ruggedized versions of a variety of different sized PMTs are available.

An ultra-compact photosensor package, incorporating a metal channel PMT, with a bialkali photocathode, and support electronics has been purchased and set-up. Laboratory tests to evaluate the performance of the photosensor package have been initiated. This photosensor package is easily capable of being deployed into 2-inch pipe.

Four test samples of a low density polyethylene (LDPE) with varying amounts of ZnS(Ag) incorporated have been prepared. The ZnS(Ag) concentrations range up to what is thought to be the maximum practical abundance that can be achieved and still allow the material to be fabricated into a blown film. Currently, tests are underway to characterize the scintillation and strength properties of these test samples. Figure 3 shows a pulse height distribution of an AM-241 alpha source obtained with the ultra-compact photosensor and one of the test samples. The source activity was approximately 1 microcurie.

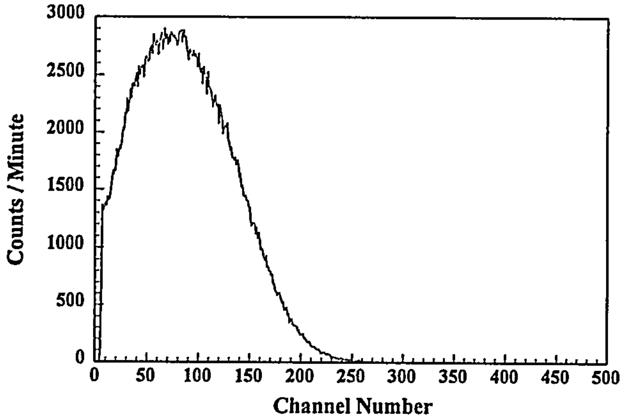

Figure 3. Pulse height distribution of an Am-241 alpha source obtained with ZnS(Ag) in LDPE and an ultra-compact PMT.

Application

As this project is still in the very early stages, no significant activities toward the application of this measurement capability have been made. Once the measurement technology is fully developed and successfully demonstrated, routine application of the measurement capabilities can be made. Most likely, this measurement capability will be utilized in decommissioning and decontamination projects to perform initial characterization measurements, or to verify decontamination activities. Another potential use is to survey air handling duct work for the purposes of identifying zones of plutonium accumulation. As the project progresses, other potential uses of this measurement capability will be identified.

Future Activities

As of this time, good progress is being made toward achieving the technical objectives outlined for this project. No significant obstacles have been identified that would seriously affect the schedule, cost, or technical goals of this project. Future activities identified at this time, generally correspond to completing the scheduled development activities outlined for Phase I of the project. Discussions have been initiated with Rocky Flats concerning the possibility of conducting a field demonstration at this site during Phase II of the project.

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PI.5 Rapid Surface Sampling and Archival Record System

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Introduction

A number of contamination sites exist in this country where the area and volume of material to be remediated is very large, approaching or exceeding 10^6 m^2 and 10^6 m^3. Typically, only a small fraction of this material is actually contaminated. In such cases there is a strong economic motivation to test the material with a sufficient density of measurements to identify which portions are uncontaminated, so extensively they be left in place or be disposed of as uncontaminated waste. Unfortunately, since contamination often varies rapidly from position to position, this procedure can involve upwards of one million measurements per site.

The situation is complicated further in many cases by the difficulties of sampling porous surfaces, such as concrete. For example, on concrete the standard wipe test provides results that are strongly operator- and surface condition-dependent. The results are not usually available real time, because the wipe samples are sent to a remote laboratory for analysis, entailing a delay of days to weeks between sampling and results. Further, the cost of analysis of a clean sample is usually the same as that of a contaminated sample; analysis of clean samples thereby increases the total analysis costs considerably. Another method for surface/sub-surface characterization is to obtain a boring or a drilling. This method also involves the off-site analysis of the drilled material, incorporating the time and economic penalties described above. Other disadvantages of drilling techniques are disfiguration of the surface and distribution of contamination through byproduct dust.

It is for the above reasons that we have chosen to develop thermal sampling methods for characterization of concrete, transite, and contaminated bulk debris. The sampling system we describe has been designed to greatly reduce the economic penalty posed by these difficulties.

Experimental Configuration

Previous work in remediation technology has demonstrated that thermal methods are very effective at extracting semi-volatile organics from porous solid matrices. Therefore, we have developed a thermal sampling head for contamination measurements on concrete surfaces. The overall experimental configuration is illustrated in Figure 1. The concrete sampling head is connected to a "quick-look detector" whose purpose

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is to provide an immediate distinction between contaminated and uncontaminated surfaces. From the quick-look detector the contaminant-laden air is pulled through a sorption tube which provides long term storage of contaminants for detailed analysis. In operation the quick-look indication will be passed along with the sorption tube to indicate which tubes required detailed analysis and to permit avoidance of analysis of uncontaminated samples. It is intended that the sorption tubes will be held in a cassette capable of holding enough tubes for several hours of measurement at a rate on the order of ten measurements per hour. Sample acquisition and analysis will be automated.

Test Methods

The concrete sampler head was tested on concrete "cakes" that had been prepared with contaminated sand. In total, seven concrete cakes were prepared 30 cm in diameter and 3.3 cm thick. One of the concrete cakes was prepared with clean sand and was instrumented with thermocouples to enable us to determine the settings necessary to bring the surface of the concrete to 250°C. The remaining six cakes were prepared using sand that had been contaminated with anthracene, 2,4,5-trichlorobiphenyl, and phenyldodecane. These three compounds represent three typical kinds of environmental pollutants. The contaminated sand was mixed with clean sand, portland cement, water, and poured into cake molds. The concrete cakes were aged in 100% humidity for 7 days, then at 45% humidity for 6 months. The concentrations of the analytes in the resulting cakes are shown in Table 1.

Results

Over a heating period of 4-5 minutes, the concrete sampling head extracts contaminants primarily from the first two millimeter surface layer of concrete. Table 2 shows the average heating rate we achieved over a 100 cm^2 surface over this period. The quick-look detector provided a real time indication of contamination. A typical quick-look indication, corresponding to a total contaminant collection of 10 μg, is shown in Figure 2. Solvent traps were used to trap analyte vapor and water that evolved from the heated cake surface.

In Figure 3 are shown the theoretical amounts of anthracene, 2,4,5-trichlorobiphenyl, and phenyldodecane that were found in the top 1 mm of the concrete cakes. Also shown are the micrograms of anthracene, 2,4,5-trichlorobiphenyl, and phenyldodecane that were found in our solvent traps. For the most part, the 5 minute collection time correlates within a factor of two with the assumed uniform distribution of analytes in the first 1 mm depth. Only anthracene exhibits anomalously low recovery. Loss of anthracene from the concrete surface by evaporation processes are believed to account for the low recovery.

We have also designed and built a bulk sampling oven (see Figure 4) useful for the analysis of dust, sample drillings, and for the initial calibration measurements of equipment. The bulk sampling oven has a removable tray designed to hold loose material. The bulk sampling oven was used to volatilize known amounts of analyte (anthracene, 2,4,5-trichlorobiphenyl, or phenyldodecane). A methylene chloride solution of the analytes was placed in an aluminum pan and allowed to evaporate. The analyte vapors were trapped with almost 100% efficiency on carbon-filled sorbent tubes. A photoionization detector connected to the exit side of the bulk sampling oven was adequately sensitive to 10 micrograms of analyte. Figure 2 shows the response of the quick-look photoionization detector (PID) to 10.3 μg of 2,4,5-trichlorobiphenyl vapor. The PID response to analyte vapor occurred within a matter of 1 minute of inserting the aluminum pan into the oven assembly.

Conclusion

We have determined that the thermal sampling approach gives more accurate results of analyte levels on concrete surfaces when compared to the standard wipe test methods. The use of a quick-look detector eliminates the need to analyze "clean" environmental samples. The sorption tubes have been shown to provide a reliable quantitative means for transfer of analyte to an automated thermal desorption interface to a GC. We will be extending our work to include sampling of steel surfaces in phase II of this program. Models of the extraction process for porous surfaces such as concrete will be developed in the next phase of this program.

Benefits

The RSSAR system will provide DOE with a new tool to dramatically lower the costs associated with building decontamination and decommissioning activities. It will allow surfaces to be rapidly sampled for the presence of semivolatile organic contaminants and will provide the means for quickly identifying clean surfaces. It will archive samples in a form that will easily allow automated laboratory analysis and will automatically associate the sampling data with the lab analytical results.

Future Activities

In Phase II, the team will optimize the concrete sampling head and develop the multi-sample trapping module and will construct a fully integrated lab scale system.

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PI.6 Portable Sensor for Hazardous Waste

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Introduction

We are beginning the second phase of a three and a half year program designed to develop a portable monitor for sensitive hazardous waste detection. The ultimate goal of the program is to develop our concept to the prototype instrument level. Our monitor will be a compact, portable instrument that will allow real-time, *in situ,* monitoring of hazardous wastes. This instrument will be able to provide the means for rapid field screening of hazardous waste sites to map the areas of greatest contamination. Remediation efforts can then focus on these areas. Further, our instrument can show whether cleanup technologies are successful at reducing hazardous materials concentrations below regulated levels, and will provide feedback to allow changes in remediation operations, if necessary, to enhance their efficacy.

Our approach is to excite atomic and molecular fluorescence by the technique of active nitrogen energy transfer (ANET). The active nitrogen is made in a dielectric-barrier (D-B) discharge in nitrogen at atmospheric pressure. Only a few emission lines or bands are excited for each hazardous species, so spectral resolution requirements are greatly simplified over those of other spectroscopic techniques. The dielectric-barrier discharge is compact, 1 to 2 cm in diameter and 1 to 10 cm long. Furthermore, the discharge power requirements are quite modest, so that the unit can be powered by batteries. Thus an instrument based on ANET can readily be made portable.

During the first phase of the program we demonstrated that a variety of hazardous species could be detected by the technique of active nitrogen energy transfer (ANET) excitation of atomic and molecular fluorescence. Species investigated included heavy metals, Hg, Cr, and Se, both chlorinated and non-chlorinated organics, and uranyl compounds. For most of these species we demonstrated sensitivity limits for their detection at parts per billion (ppb) levels.

Our principal goals for this second phase of the program are to develop and breadboard test instrument components and to design a prototype instrument suitable for construction and evaluation in the final phase of the program. A secondary goal is to extend the ANET technology to encompass a greater number of hazardous species, primarily additional heavy metals and radionuclides.

We begin this current phase by surveying several DoE sites to better coordinate the capabilities of our monitor with their specific needs. Following this we will establish procedures for sample handling and analysis, and

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develop systems and software needed to automate these processes. System components and processes being examined include sample collection and introduction, active nitrogen source stability, fluorescence detection and analysis, and measurement, calibration, and test procedures.

The second element of the program is to design a prototype hazardous waste monitor. This design includes not only identifying each of the components to be incorporated into the instrument and determining how they will be integrated into a compact package, but also specifying component technical requirements and their vendors.

At the end of this current phase we will have a detailed set of drawings and specifications for the construction of a first-generation, or alpha-, prototype instrument. This instrument will be built and field tested as part of the third and final phase of the program.

The third, and final, phase of the program will involve fabrication and field testing of the alpha-prototype instrument designed during the program's second phase. We will test and refine the instrument in the laboratory first, then conclude the program with an extensive series of field tests. At the conclusion of the program we will transfer the developed ANET technology to Spectrum Diagnostix (SDx), a subsidiary of Physical Sciences Inc. (PSI). They have commercialized several instruments initially developed at PSI on SBIR programs.

Approach

The technique we chose for detecting hazardous species with high sensitivity was excitation of atomic and molecular fluorescence by active nitrogen energy transfer (ANET). The active nitrogen is made in a dielectric-barrier (D-B) discharge in nitrogen at atmospheric pressure. Figure 1 illustrates our instrument concept schematically. Samples are fed into the D-B discharge region where the active nitrogen is generated, and where it transfers its energy to species in the sample, thereby exciting fluorescence from them. The pertinent fluorescence is then isolated by a small spectrograph or interference filter and is detected by a photomultiplier or diode array placed behind the filter or spectrograph. Signals from the detector are then processed by a computer and a report is generated. The computer control allows both real-time reporting and data storage for subsequent processing or archiving.

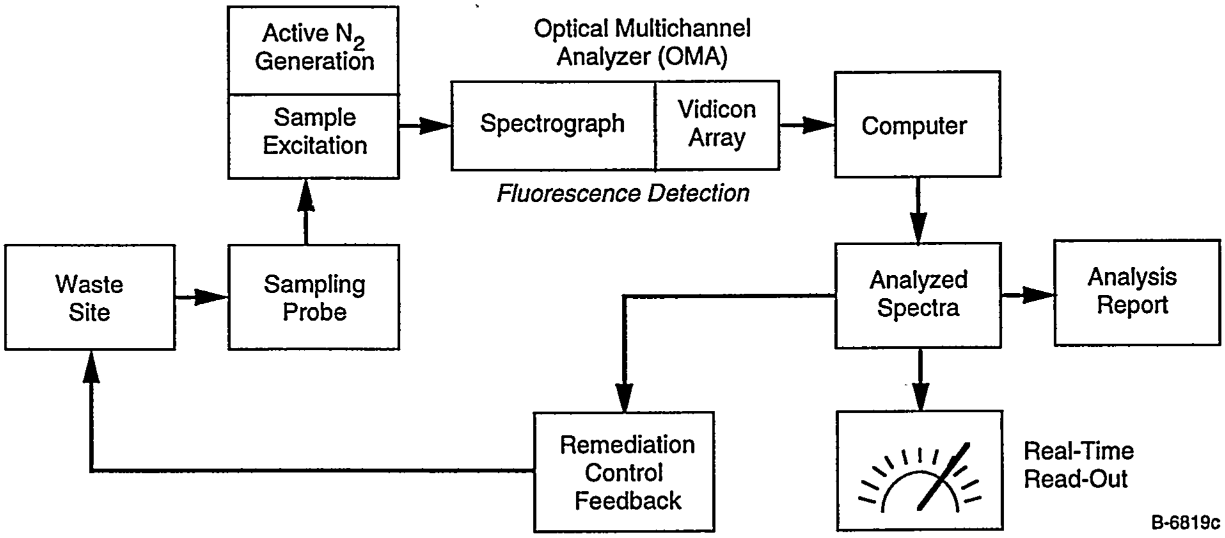

Figure 1. Block Diagram of ANET Analysis System.

ANET excitation generates only a few emission lines or bands for each hazardous species (see Figure 2). Thus, spectral resolution requirements are greatly simplified over those of other spectroscopic techniques. The dielectric-barrier discharge is compact, 1 to 2 cm in diameter and 1 to 10 cm long. Furthermore, the discharge power requirements are quite modest, so that the unit can be powered by batteries. Thus an instrument based on ANET can readily be made portable.

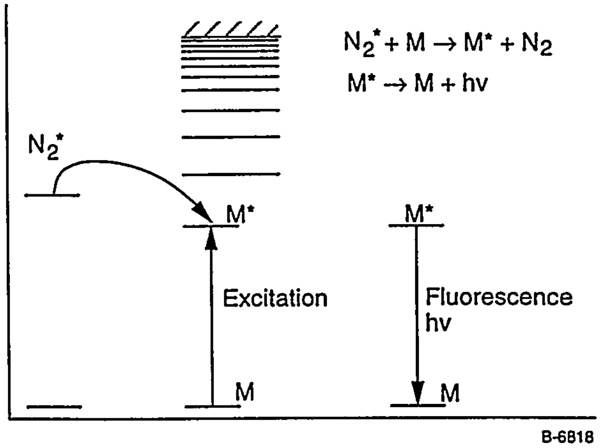

Figure 2. Schematic Descriptin of ANET Excitation Mechanism,

Active Nitrogen Energy Transfer (ANET)

From the beginning of this century, scientists have observed characteristic emissions from atoms and free radicals when atomic and molecular species were added to active nitrogen. A rich literature exists detailing the chemical reactions and energy-transfer processes that occur to excite these emissions.[1-6] Basically, metastable nitrogen molecules (molecules having internal energies of 2 to 10 eV which they cannot easily release via radiation) in the active nitrogen transfer their energy to the various acceptor species. These acceptor species then fluoresce at wavelengths characteristic of the acceptor. Most of these studies relied upon the recombination of nitrogen atoms at relatively low pressures to generate the metastable nitrogen molecules. For analytical applications, this approach has the disadvantage that considerable power is required to dissociate atomic nitrogen. In addition, because these systems operate at pressures on the order of 1 to 10 Torr, a vacuum system is required and sampling under ambient conditions becomes much more difficult.

We conceived and began developing ANET as an alternative to active nitrogen analytical techniques using an atomic-nitrogen recombination source. Our approach is to generate the metastables in a dielectric-barrier discharge operating at atmospheric pressure and total powers of a few Watts. Thus our system requires neither a large power source nor a vacuum system. In addition, we believe that the overall metastable number densities generated in the dielectric-barrier discharge are several orders of magnitude larger than those in the atom-recombination system, which means that ANET has the potential to be several orders of magnitude more sensitive.

Dielectric-Barrier Discharge Technology

A dielectric-barrier discharge is a high voltage a.c. discharge between two electrodes, at least one of which is separated from the discharge region by a dielectric barrier (insulator) such as glass[8-11] (see Figure 3). A typical discharge will run at voltages between 3 and 30 kV at frequencies from line frequency to 100 kHz. Gas pressures are typically an atmosphere and gap spacings are on the order of a few millimeters. In its simplest form, the discharge can be powered by attaching the electrodes to the output of a high voltage, step-up transformer, such as a neon-sign transformer, plugged into a variac.

The dielectric-barrier discharge (also referred to as an ozonizer discharge) is not a

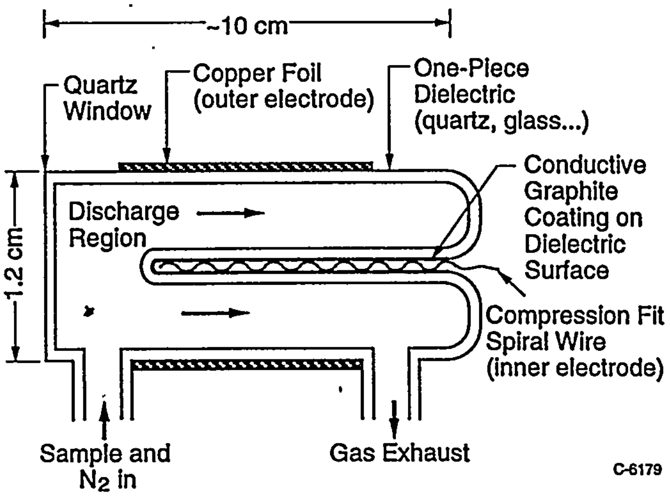

Figure 3. Schematic Diagram of Dielectric-Barrier Discharge Lamp

single discharge in the sense of a continuous arc or glow discharge, but rather a collection of innumerable microdischarges between the dielectric and the other electrode. These microdischarges consist of short duration (typically 10 to 100 ns) current pulses (100 to 1000 A cm^{-2}) localized in roughly cylindrical filaments, typically 100 μm in radius. The mean electron energy in a microdischarge is on the order of 1 to 10 eV which is ideal for metastable generation. At any given instant in time, the microdischarges are distributed uniformly across the face of the dielectric. This uniformity provides a relatively stable excitation throughout the discharge volume.

The Chemistry and Physics of ANET

Dielectric-barrier discharges in pure nitrogen have been shown to be efficient sources of metastable $N_2(A\,^3\Sigma_u^+)$[12-15], even at pressures of one atmosphere, thus providing the active nitrogen source necessary for the selective analyte excitation of ANET. If the analyte species is an atom, such as elemental mercury, the excitation occurs directly. When molecular species are added to the discharge region, the $N_2(A)$ generally reacts with the molecule, producing molecular fragments, which it subsequently excites. For example, $HgCl_2$ is dissociated to $HgCl^*$ which emits around 540 nm, organic molecules are broken down into CN radicals which emit near 388 and 420 nm, and chlorinated organics to CCl molecules emitting at 278 nm.

The chemical processes responsible for exciting fluorescence from species M in a dielectric-barrier discharge are summarized by the following reactions:

$$
\begin{aligned}
N_2 + e^- &\rightarrow N_2^* + e^- &(1)\\
N_2^* + N_2 &\rightarrow N_2 + N_2 &(2)\\
N_2^* + M &\rightarrow M^* + N_2(X) &(3)\\
M^* &\rightarrow M + h\upsilon &(4)\\
M^* + N_2 &\rightarrow M + N_2 &(5)\\
M^* + Q &\rightarrow M + Q' &(6)\\
N_2^* + Q &\rightarrow N_2 + Q', &(7)
\end{aligned}
$$

where, Q represents a species in the discharge that quenches electronic energy in either the N_2^* or the electronically excited analyte fragment.

Although the dielectric-barrier discharge is a pulsed discharge, one can treat it as if it were a continuous discharge if observations are averaged over a number of discharge cycles. Then, because of their short radiative and quenching lifetimes, the excited species in the discharge region are effectively in steady state and we can write

$$
I_{M^*} = k_4[M^*] = k_3[M][N_2^*]/(1 + (k_5/k_4)[N_2] + (k_6/k_4)[Q]). \qquad (8)
$$

Equation (8) shows that for constant metastable number density and total pressure, the fluorescence intensity will be linearly proportional to the additive number density, provided the number densities of any potential quenchers remain constant. This generally will be the case. In previous investigations, the linearity of active-nitrogen excited fluorescence intensity with analyte number density has been demonstrated experimentally to cover four to five orders of magnitude for many species.

The exciting species in active nitrogen responsible for exciting fluorescence from additives generally is considered to be $N_2(A\ ^3\Sigma_u^+)$, although other nitrogen metastables have been invoked from time to time.[1] $N_2(A)$ carries about 6 eV of internal energy. Based on the reaction scheme above, its effective steady-state number density is given by:

$$[N_2^*] = k_1[e^-][N_2]/(k_2[N_2] + k_3[Hg] + k_7[Q]). \qquad (9)$$

The radiative lifetime[16] of $N_2(A)$ is about 2.5s. Thus, the primary mechanism for its deactivation will be quenching rather than radiative decay.

Quenching of either the analyte fluorescence or the nitrogen metastables can be an issue affecting the sensitivity of ANET. Although rate coefficients for quenching $N_2(A\ ^3\Sigma_u^+)$ and many analyte emissions by a number of important species are known[7, 17-19], one should determine the effects of quenching in a D-B discharge *in situ* to ensure that unexpected processes do not complicate the analysis. Our observations related to metastable nitrogen quenching in the D-B discharge[20] are generally consistent with more direct quenching measurements. While our Phase A investigations showed quenching was a problem in our system, we were able to demonstrate that it did not seriously compromise the overall sensitivity of the ANET technique for detecting important hazardous species. Even accounting for quenching effects, our detection sensitivities were at ppb levels or below.

Project Description

Summary of Phase A Results

The primary goals of the first phase of the program were two fold:

- to demonstrate the variety of hazardous species that can be detected by the technique of active nitrogen energy transfer (ANET) excitation of atomic and molecular fluorescence;
- to demonstrate sensitivity limits for detecting several of these species are at parts per billion (ppb) levels.

Species to be investigated included heavy metals, organics and chlorinated organics, and/or transuranic surrogates.

To achieve the stated Phase A goals, we designed a program where we first focused on the ANET technique by studying parameters important to the production and quenching of nitrogen metastables in an atmospheric pressure, dielectric-barrier discharge. These metastables are the primary excitation species in the active nitrogen. The second component of the program was qualitative and quantitative studies on the detection of several hazardous species including heavy metals, both chlorinated- and non-chlorinated-organic molecules, and uranium compounds. The third component of the program was to investigate the effects of adding small quantities of dust to the D-B discharge region. The two issues to be resolved in this part of the investigation were whether the presence of dust in the discharge region would compromise the operation of the D-B discharge and whether species adhering to dust could be detected.

Experimental. Our Phase A program was essentially a laboratory study to prove our concept for monitoring hazardous waste. We demonstrated that a variety of hazardous species that could be detected by ANET and determine the sensitivities at which ANET could detect the various species.

Our apparatus is shown schematically in Figure 4. It consists of a dielectric-barrier discharge lamp, power source, optical multichannel analyzer, and gas handling lines.

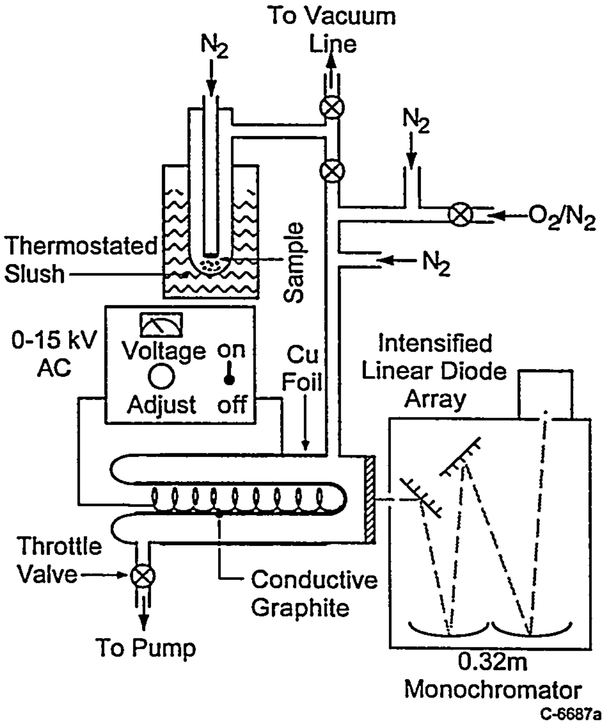

Figure 4. Schematic of Laboratory Apparatus

Most of our lamps were fabricated from a single piece of quartz having a 12 mm o.d. body with an axial protrusion inside the 12 mm tube, thus giving the lamp an annular discharge region. We used several lamps with various protrusion diameters ranging from 3 to 8 mm o.d. The end opposite the protrusion is sealed with a quartz window. The outer electrode is formed by wrapping copper-foil tape around the outside of the lamp body. We painted the inside of the axial protrusion with a layer of conductive graphite or nickel to form the inner electrode. A wire, wrapped loosely with copper turnings to contact the conductive inner surface, was inserted into the protrusion and connected to the power supply. This configuration provides a clean discharge region since all surfaces are quartz.

For most experiments, we powered the discharge with 15 kV neon sign transformer having a center-tap-grounded secondary winding. The voltage to the primary was controlled by a variac operating off standard 60 Hz ac power. For a few measurements we connected the electrodes to the secondary of an automotive ignition coil which was powered with 12 to 15 V square-wave pulses at frequencies between 45 Hz and 3 kHz. This power source has great potential because it lends itself to battery operation.[21]

Samples were placed in a cold finger immersed in a bath that was thermostated for vapor pressure control. A small flow of nitrogen through the cold finger transported the samples into the discharge region. The concentration of the analyte in the discharge region was equal to the product of the compound's vapor pressure in the cold finger and the dilution factor between the N_2 flow through the cold finger and that through the D-B lamp as a whole. The temperature of the thermostated bath generally was quite cold, e.g., slush baths of chloroform (-63 °C) or ethanol (-110°C). At these temperatures the vapor pressures of most species studied were on the order of 10^{-5} Torr or below. The bath temperatures were measured with a thermocouple or thermometer.

We used a Princeton Instruments ST210 Optical Multichannel Analyzer (OMA) system to detect the fluorescence. This system consisted of a 0.32 m monochromator, generally with a 2400 groove mm^{-1} grating and an intensified linear diode array. The lamp was sufficiently bright, that relatively high resolution spectra ($\Delta\lambda \sim 0.1$ nm) could usually be accumulated with good signal to noise in only 10 seconds. Data were stored in a computer for later analysis.

Results from the Phase A Program. The primary activity in Phase A was to characterize the D-B discharge spectroscopically both in the absence of added species, to provide an understanding of the basic operation of the discharge itself, and in the presence of added species. When species were added, we characterized the fluorescence spectrum resulting from their addition, and in some instances, determined how fluorescence intensities characteristic of the additives varied as a function of concentration of the added species.

Our Phase A laboratory study of hazardous species' detection in a D-B discharge in N_2 at atmospheric pressure showed the overall sensitivity for Hg and Se detection to be at sub part-per-billion levels, and that for organic species to be on the order of a few parts per billion. Furthermore, ANET could differentiate between chlorinated and non-chlorinated organic species, could detect several different heavy metals simultaneously, could differentiate elemental mercury from mercury tied up as chloride, and could detect species adhering to particulates. Finally, we observed molecular emission upon adding uranyl compounds to the D-B discharge. We think it most likely that the emission arises from the UO_2 molecule, although emission from UO is also a possibility.

Figure 5 shows the spectral region in the vicinity of the Hg 253.7 nm emission line. The mercury line is prominent in the spectrum, and is surrounded, but well separated from, several bands of the NO($A\ ^2\Sigma^+$ -- $X\ ^2\Pi$) system. These bands are also excited by energy transfer from metastable nitrogen[22]. The NO is formed in the discharge in reactions involving traces of oxygen in the nitrogen.

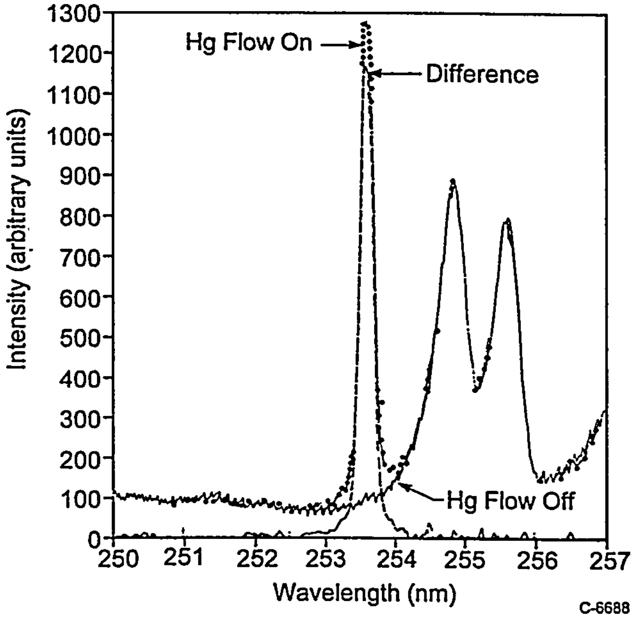

Figure 5. Spectrum of Dielectric-Barrier Disharge in the Absence and Presence of Added Hg

Figure 6 illustrates the linear proportionality between the mercury intensity in the D-B discharge lamp and the mole fraction of Hg added to the lamp. These data, when combined with results of quenching measurements allowed us to estimate a detection sensitivity of 0.07 ppb for Hg in air.

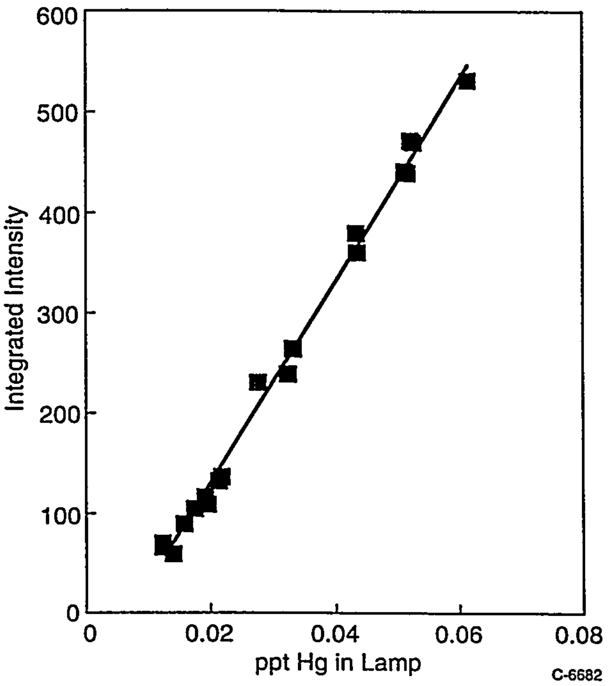

Figure 6. Intensity of Hg 253.7 nm Emission Excited by ANET as a Function of the Mole Fractin of Hg in a D-B Discharge

Measurements with Cr and Se compounds also showed a linear relationship between the intensity of the ANET excited fluorescence and the concentration of the additive in the discharge region. Our results indicate a sensitivity for Se detection in air below 1 ppb. The sensitivity for Cr detection appeared to be somewhat higher, but we think our concentration estimates were erroneous. We will investigate this problem in more detail in the Phase B program.

All hydrocarbons studied can be detected in a D-B lamp in nitrogen at atmospheric pressure by observing emission from the CN(B - X) system at 388 or 420 nm. Figure 7 illustrates this at 388 nm for the case of chloroform addition. Figure 7 shows the spectra generated in the D-B lamp in the absence of any added chloroform, with about 0.05 ppb added chloroform, and the difference between the two spectra, which is a spectrum of just the CN emission. We have observed identical spectra from a number of hydrocarbons including hexane, acetone, methanol, isopropanol, 1,2-dichloroethane, trichloroethylene, 1-chlorobutane, and even bromotrifluoromethane.

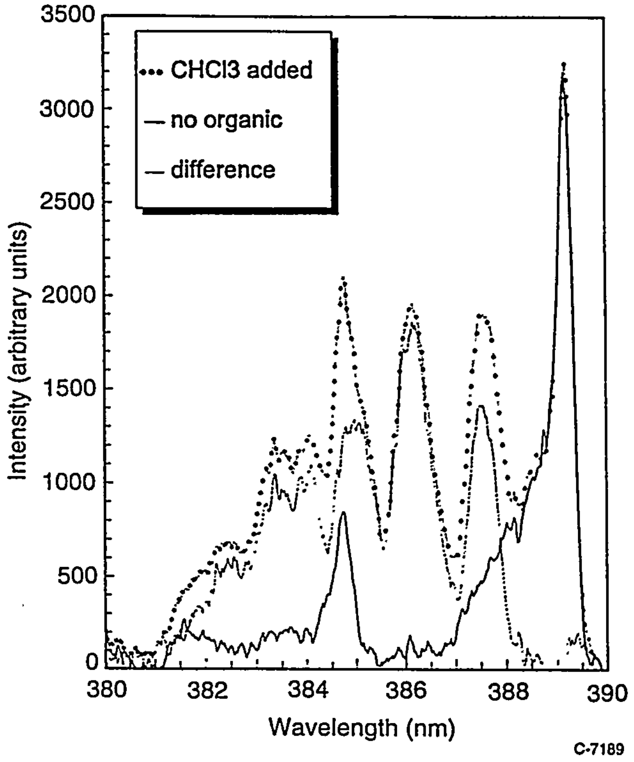

Figure 7. Spectrum of a dielectric-barrier discharge lamp in nitrogen at atmospheric pressure in the absence of any added chloroform, with 0.05 parts per billion of added chloroform, and the difference between the two spectra.

The presence of chlorinated hydrocarbons is signaled by the appearance of CCl emission at 278 nm in addition to the just discussed CN emission. Figure 8 illustrates this in the case of chloroform. Figure 8 shows the spectra generated in the D-B lamp in the absence of any added chloroform, with 0.01 ppb added chloroform, and the difference between the two spectra, which is a spectrum of just the CCl emission. To date, all

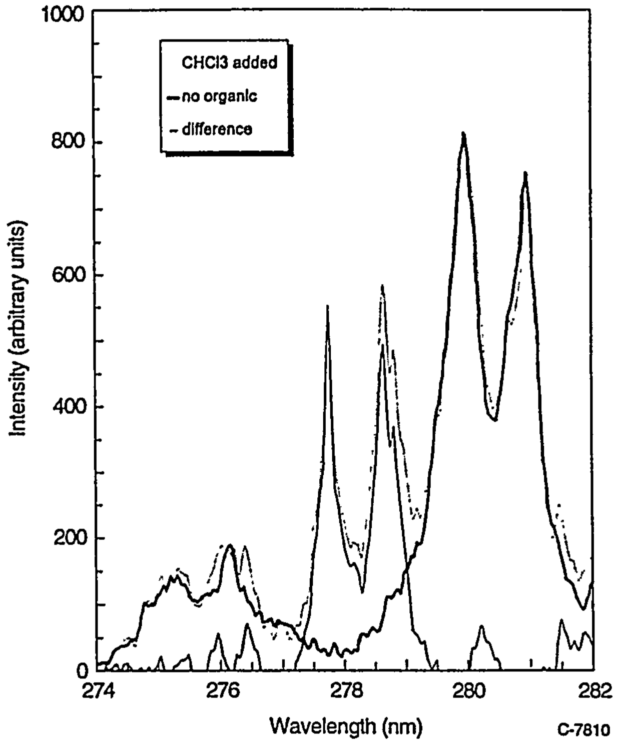

Figure 8. Spectrum of a dielectric-barrier discharge lamp in nitrogen at atmospheric pressure in the absence of any added chloroform, with 0.01 parts per billion of added chloroform, and the difference between the two spectra.

chlorinated hydrocarbons studied have shown this fingerprint.

The relationship between the mole fraction of added hydrocarbon and CN or CCl band intensity was also linear. Our observations on hydrocarbon excitation, when corrected for quenching by air, indicated sensitivities at both wavelengths of about 5 parts per billion.

Figure 9 shows the spectrum resulting from the addition of uranyl acetate to the discharge region along with a background spectrum and the difference between the two. Several of the features in the background spectrum are easily identified as being part of the nitrogen second-positive system. The three rather broad bands appearing at 486, 510, and 534 nm in the spectrum with added uranyl acetate, however, are not readily identifiable. A repeat measurement over the same region using uranyl nitrate in the lamp resulted in a very similar spectrum. We think, therefore, the spectrum in Figure 9 is from uranium oxide, probably UO_2, but possibly UO. Surveying a fairly wide range of wavelengths failed to show atomic line emissions from U. Since most uranium waste is likely to be oxidized, this probably isn't a serious drawback. Unique spectral features identify uranium as being present.

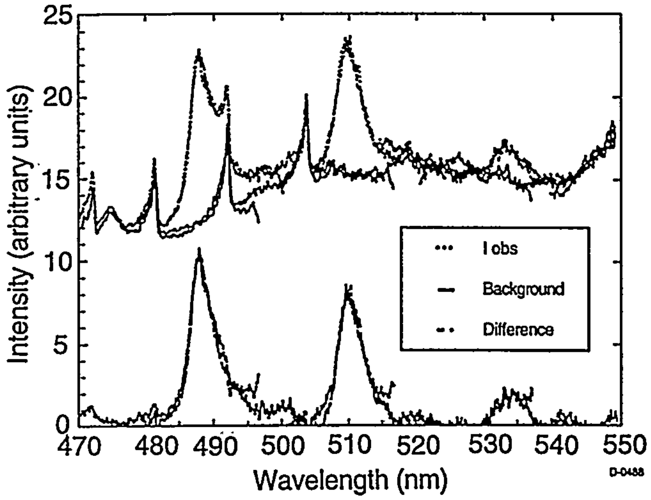

Figure 9. Spectrum of a dielectric-barrier discharge in nitrogen at atmospheric pressure in the absence of any added uranyl acetate, with a small quantity of added uranyl acetate, and the difference between the two spectra. Each spectrum shown is a composite of four separate spectra. We tentatively ascribe the difference bands to the UO_2 molecule.

Objectives of Phase B Program

The work in the first phase of this program successfully demonstrated that ANET technology holds great promise for development into a sensitive, multi-element hazardous species monitor. Our plans for the second phase of the program are to develop a breadboard prototype system and characterize the ANET process under more realistic conditions. During the third phase we will build a working prototype of a commercial instrument and test it in the field.

The Phase B program has three primary components:

- process development
- design engineering
- technology extension.

The purpose of the process development task is to establish the procedures for sample handling and analysis, and to develop systems and software needed to automate these processes. System components and processes to be examined include sample collection and introduction, active nitrogen source stability, fluorescence detection and analysis, and measurement, calibration, and test procedures.

The most crucial issues are sampling, calibration, and testing. The other issues, fluorescence detection and analysis, active nitrogen source stability, and measurement procedures, were explored in some detail in Phase A. Implementing these components into the breadboard system should be straightforward.

The sampling and testing issues, however, will ultimately determine the success or failure of the ANET technology. Designing a system that abstracts samples truly representative of the hazardous environment is crucial. Only if this is accomplished can the true hazardous species concentrations be determined accurately. Therefore, sample entrainment, speciation, calibration and matrix effects are of primary import.

The design engineering task has two elements. The first of these is to identify and specify each of the components to be incorporated into the prototype instrument we shall fabricate as a part of the final phase of the program. Each component needed will be enumerated, and commercial suppliers will be identified. We will investigate the efficacy of in-house construction of some components versus their purchase from commercial OEM suppliers. Final component suppliers will be determined by balancing various factors, including adaptability, quality, size, weight, and cost.

The second element of the design engineering task is to design of a prototype hazardous waste monitor. This design will balance a number of considerations including power management and portability. The location of each component will be studied in relation to the others to identify configurations that promote simple operation and servicing while retaining compactness in the whole instrument. At the end of the program's second phase we will have a detailed set of drawings and specifications for the construction of a first-generation, or alpha-, prototype instrument.

The third component of the program is to demonstrate the applicability of the ANET technology to detecting a number of hazardous species not investigated in Phase A. The activities on this task will involve not only identifying unique finger prints for each of the species to be investigated, but also to determine quantitative limits for their detection. The species we shall investigate in this task include the heavy metals, Sb Be, Cd, Cr, Pb, and Hg, and the radionuclides, Tc, Th, and U.

Results

This section outlines our progress to date on the process development, engineering development, and technology extension tasks.

Process Development tasks

An instrument using ANET to monitor hazardous wastes in real-time will have five essential components:

1. a sampling system for collecting the sample from contaminated material and diluting it with nitrogen;
2. a discharge system for exciting the N_2 metastables which, in turn, excite contaminant fluorescence;
3. a detection system for separating contaminant fluorescence from other features excited either in the discharge or by the N_2 metastables;
4. an in-situ calibration system to ensure the accuracy of the hazardous waste measurements; and
5. a data processing system for controlling the various instrument components, for collecting the data, for converting the raw data into hazardous waste concentrations in the sample, and for reporting and archiving hazardous waste analysis results.

In our developmental work, we are considering each of these issues in some detail before beginning to purchase and assemble components.

Before describing our strategy for each of the design issues, however, we'll first describe schematically the basic principles of the instrument operation. Figure 10 shows a block schematic of the instrument where the component subsystems are identified. The sample is removed from the surface being tested and diluted by a two-state ejector pump system. The diluted sample then flows into the discharge system In the discharge chamber, the N_2 metastables, and subsequently the contaminant fluorescence, are excited. Light from the discharge is dispersed by a small monochromator and detected with a photomultiplier or intensified photodiode array. The detected signals are read and processed by a computer system, and results are displayed and stored. The computer system can also provide feedback signals for active control of remediation processes.

A calibration system will be included that can inject known quantities of contaminant into the diluted sample flow prior to its entry into the discharge cell. In addition, an on-line oxygen monitor may be incorporated, if necessary, so that O_2 levels in the discharge cell can be accurately known. This allows the fluorescence data to be corrected for O_2 quenching. We are developing a more direct monitor of quenching so we may be able to dispense with the O_2 monitor in the final version of the instrument.

Our initial system performance and design goals are as follows:

- ability to detect heavy metals and radionuclides in gas phase and on particulates
- sensitivity range for all species 0.1 to 10 ppbw
- automatic, *in situ* calibration of all contaminant species
- at least six months between repair and maintenance operations

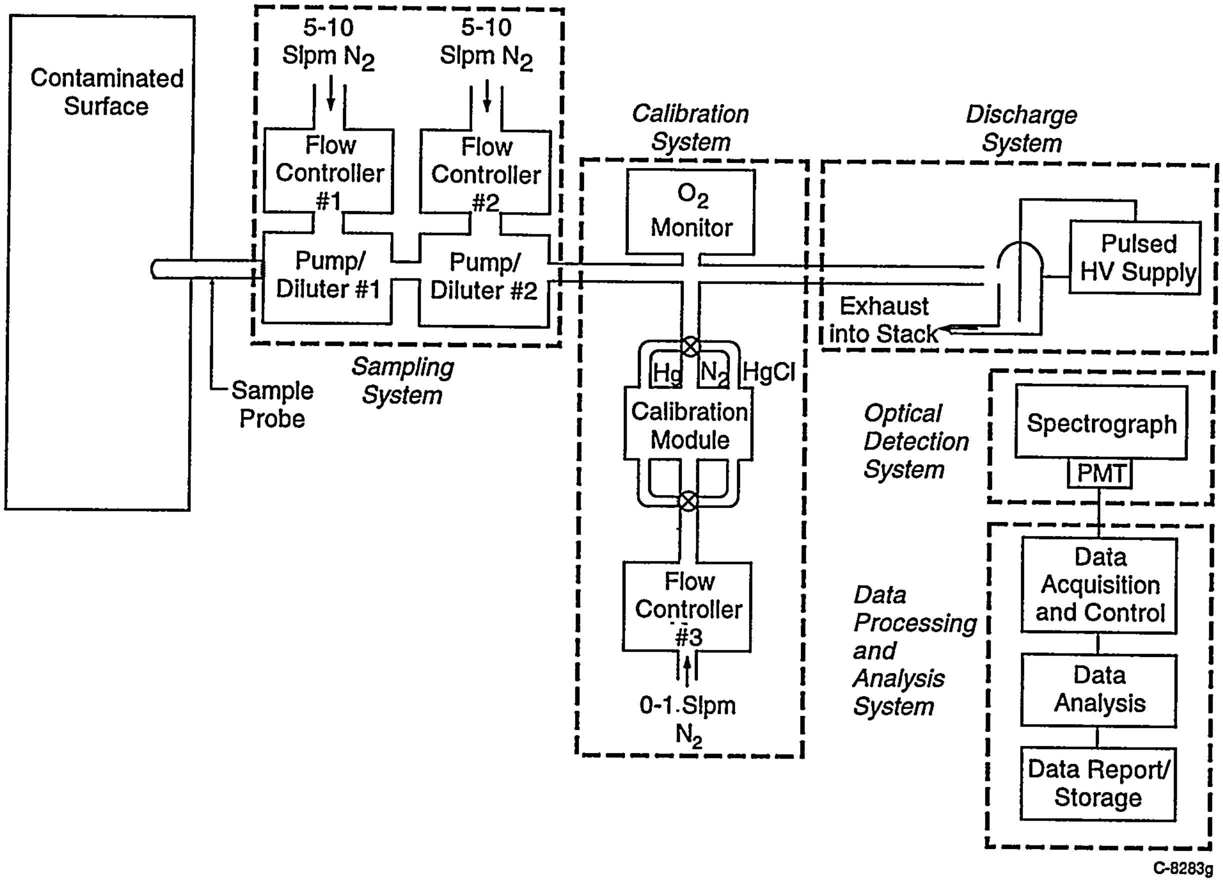

Figure 10. Block Schematic of ANET-Baset Hazardous Waste Monitor

- analysis and display of results in less than one minute
- size, <50x50x100 cm (ca. 10 ft^3)
- weight <75 kg
- power <1.5 kW.

Sampling system. The primary constraint on the sampling system design is that our technique requires a high level of sample dilution, on the order of 1 part in 10^4. In addition, system components must be compatible with species being detected and mechanical simplicity is desirable. These three considerations can be readily satisfied with a gas ejector pumping system. An additional possible constraint might be the necessity of sampling particulates.

Gas-driven ejector pumps can provide the necessary vacuum to draw the sample into the probe. The rate at which gas is drawn into the probe is limited by a small orifice in the probe upstream from the ejector pump. For a pressure drop across the orifice of roughly a factor of two or greater, the rate of flow through the orifice is independent of the ejector-pump flow rate, but is determined only by the pressure and temperature on the high-pressure side of the orifice (critical flow). The required levels of dilution obtain from a balance between the efficiency of the ejector-pump system and the diameter of the critical orifice. In practice, a two-stage ejector-pump system is allows for larger orifices (~100 μm) which reduces the chance of orifice clogging.

Materials compatibility for most species can generally be satisfied with glass, teflon, and sapphire components. We anticipate most components will either be made from or coated with teflon PFA.

Detection system. The detection system will monitor contaminant fluorescence, primarily at ultraviolet wavelengths, and N_2 Herman infrared (HIR) fluorescence between 695 and 710 nm. We showed in the Phase I program that the HIR fluorescence can be related directly to the concentration of N_2 metastables produced in the discharge. Monitoring the HIR emission, therefore, provides a means of compensating for variations in metastable concentrations that might occur with variations in air in the sample. The spectral resolution necessary to separate the atomic fluorescence from neighboring emissions is about 0.4 nm. Commercial interference filters with ultraviolet pass bands having this level of spectral isolation are still in developmental stages and are too expensive to be considered seriously. Spectral resolution requirements in the neear infrared are less stringent, so using an interference filter at 700 nm is a viable option. A spectrometer is necessary to obtain adequate spectral resolution in the ultraviolet, however, and works well to isolate other important wavelengths.

Compact spectrometers using diode array or CCD detectors are adequate for our purposes, but are not cheap. If possible, we would prefer to use a scanning monochromator with photomultiplier detection. Such a system is about an order of magnitude cheaper than a comparably sensitive diode aray or CCD system. In addition, stepper-motor based grating rotation mechanisms have been made sufficiently rugged to with stand the rigors of space flight, so should be able to survive in a relatively hostile field environment.

A compact instrument having a 10 to 15 cm focal length and a standard 1200 g mm^{-1} has a first-order resolution of about 8 nm mm^{-1}. Since aberrations in small focal length spectrometers limit their usefulness to slit widths greater than 100 to 150 μm, the best first-order resolution one can expect is about 1 nm. This is inadequate for atomic fluorescence detection even accounting for the much simpler spectrum generated by ANET. However, adequate resolution is available by monitoring ultraviolet fluorescence in second or third order.

We are considering a system comprised of a 110 mm focal length monochromator with an 1800 g mm^{-1} grating driven by a stepper motor mechanism controlled by the instrument's computer. A computer-controlled filter wheel situated at the monochromator's entrance slit enables switching between filters appropriate to the spectral region observed. The system would collect data from a few discrete wavelengths in a spectral region; then slew to the next and repeat the procedure. A UG-5 glass filter adequately transmits ultraviolet radiation with wavelengths longer than about 225 nm while completely blocking that at wavelengths between 400 and 650 nm. For visible wavelengths, a long-pass colored glass filter effectively blocks second-order uv radiation while transmitting that at longer wavelengths.

A compact photomultiplier assembly can be used to detect fluorescence. Emissions from the discharge extend for a periods of tens of nanoseconds after the initiation of the discharge pulse if they are excited directly in the discharge, but can extend up to 100 to 200 μs if excited by metastable transfer. To enhance discharge emission relative to dark current the photomultiplier's output can be integrated for periods of about 150 μs after the discharge pulse. The data acquisition system would then read the voltage output of the integrator and averages the result with those of several previous pulses.

An alternative approach is to use an intensified photodiode array detector. Such a detector effectively monitors a group of wavelengths simultaneously. Thus one can collect all wavelengths in a given pass band at one time. This approach will increase the speed of data acquisition and may allow several contaminants to be detected simultaneously. In addition such a system can more easily correct for any drift in the discharge system. The disadvantage of a photodiode-array based system is that it is about an order of magnitude more expensive than comparably sensitive photomultiplier-based detection systems.

Discharge system. We expect to use a discharge system similar to that used previously.[12] We described the discharge cell above in presenting our Phase A results. It is powered by applying 15 V pulses to a high-energy coil. The discharge frequency can be controlled by one of the data acquisition and control system's timers. Digital pulses from the computer would trip a gate, which in turn, would open a transistor switch able to sink adequate levels of current at 15 Vdc to the coil's primary. The high-voltage output of the coil's secondary would connect to the D-B discharge lamp.

Calibration system. The calibration system is comprised of two elements: accurately metered sources of contaminant species, and a sensor for O_2 in the sample gas. The contaminant sources allow *in situ* calibration of the monitor, while the O_2 sensor provides a means of compensating for quenching effects. Monitoring the HIR bands will probably suffice for determining the quenching correction. In our first prototype, however, it may be prudent to have an additional support.

The simplest approach for the calibration system would be to use volatile contaminant sources. Accurately controlled vapor release sources can be made from either permeation tubes or capillary diffusion cells. Because their emission rate is temperature dependent, they would be housed in a unit providing independent temperature control for each source cell. An example of a permeation tube is shown in Figure 11. A few drops of a volatile liquid or solid containing a contaminant are placed inside a small cell having a permeable membrane as one of its walls. Cylindrical permeation tubes, for example, can be made from permeable tubing, often teflon or plastic, with the ends sealed by impermeable materials. It is important to have a solid or liquid phase inside the permeation tube so that the vapor pressure inside the source can be maintained at a constant level. This ensures a constant emission rate from the source.

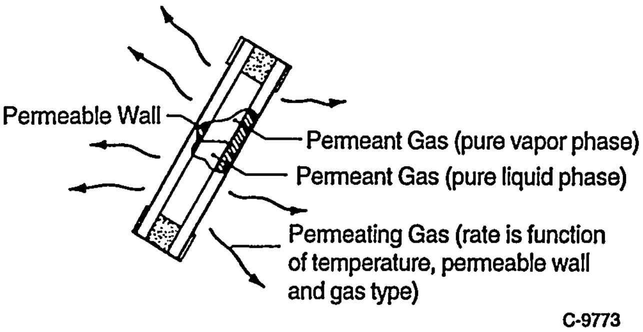

Figure 11. Schematic of Permeation-Tube Calibrator

Figure 12 shows a diffusion cell. A few crystals of volatile material sit in a small vial having a long, thin capillary as the only means of egress for the contaminant vapor. The rate at which the contaminant enters the calibration-gas flow is a function of the rate of diffusion along

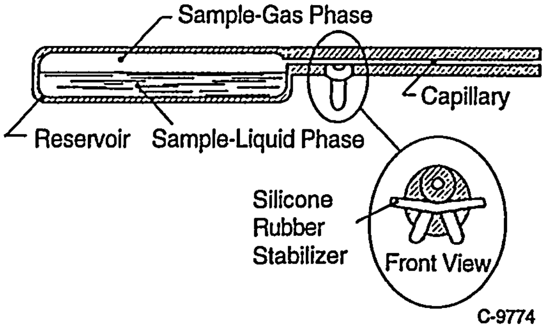

Figure 12. Schematic of Capillary Diffusion Tube Calibrator

and out of the capillary. This rate depends on the capillary's length and diameter, as well as the temperature and physical characteristics of the gas through which the contaminant is diffusing.

Permeation tubes and diffusion cells can be calibrated gravimetrically. That is, the cells can be weighed periodically on a microbalance, and over time the weight loss of material from the cell can be quantified. This approach generally works well only for sources having emission rates greater than about 10 ng min^{-1}. Sources with emission rates slower than this level take too long to calibrate accurately using a gravimetric approach and are usually better calibrated against some form of bench-top analyzer.

Data acquisition and control system. The data acquisition and control system regulates all components of the mercury monitor, collects data from each component, runs automatic system calibrations, converts collected data into mercury or mercuric chloride concentrations, and displays measurement results in real time as well as stores them for later retrieval and archiving. For the current program prototype, we will use a portable computer for this purpose. A commercial instrument would undoubtedly incorporate a single-board computer with operations programmed into EEPROM memory.

We plan to use an IBM-compatible portable PC with Windows-based software. Several data acquisition and control boards would be installed inside the console. Software to monitor and control the boards will most likely be developed using the National Instruments LabWindows package.

Design Engineering Tasks

The purpose of these tasks will be the design of a prototype hazardous waste monitor. This design will balance a number of considerations including power management and portability. The location of each component in relation to the others will be studied to identify configurations that promote simple operation and servicing while retaining compactness in the whole instrument. We will develop our system using AUTOCAD, the industry standard computer-aided design system (CAD) for IBM PC compatibles. This will also allow us to produce standard engineering drawings of the system we shall fabricate in the Phase C program.

A second part of this task is to identify and specify each of the components in the instrument design. In addition, where necessary, commercial suppliers of each component will be identified. We will also investigate the efficacy of in-house construction of some components versus their purchase from commercial OEM suppliers. For example, the major parts of the optical detection system will almost surely be more cost effectively obtained from an OEM supplier. The final component suppliers will be determined by balancing various factors, including adaptability, quality, size, weight, and cost.

Throughout the design phase of the program we will interact extensively with our subsidiary company Spectrum Diagnostix (SDx). They provide instrumentation to the power generation industry for monitoring and controling combustor temperatures and to measure concentrations of ammonia in gas streams. Both of these diagnostics are important in the control of NO_x emissions from combustors.

Technology Extension Tasks

Activities on this task will follow procedures developed during the first phase of the program. The main purpose of this task will be to establish sensitivities for detecting additional hazardous species that were not addressed in the first phase of the program. The list of species includes the heavy metals, Sb, Be, Cd, Cr, Pb, and Hg, and the radionuclides, Tc, U, and Th. Initially, to develop our diagnostic technique, we plan to work with volatile compounds of these species. In later stages of the program we will address explicitly their detection as fine aerosols.

Compounds of the element Tc are very strictly regulated and cannot be handled except in rigorously licensed locations. Because our obtaining such licensing in a timely and cost effective manner is highly improbable, we shall do our work on this species in two stages. The first is to develop a portable experimental system at PSI and test it out on Tc-surrogate compounds such as those containing Mn or Re. Then the facility will be transported off site to a location properly licensed for Tc handling and we will develop Tc detection procedures there.

Conclusions

Our Phase A program indicated that ANET is a very sensitive technique for monitoring heavy metals and chlorinated hydrocarbons. Our approach requires no special sample preparation and can operate continuously. In spite of the fluorescence quenching we have observed, the overall sensitivity is at or below ppb levels, even under less than ideal conditions.

We are now beginning to develop techniques, plans and procedures needed to develop our concept into a real-time monitor of toxic heavy metals and radionuclides at building sites, hazardous waste land fills

Future Activities

The third, and final phase of the program will be to build and field test an alpha-prototype hazardous waste monitor. The fabrication will follow closely the designs developed in the current phase of the program. The alpha-prototype development will be followed by several months of testing and refinement in our laboratories. Finally, the program will conclude with an extensive series of field tests of the instrument.

At the program's conclusion, we will transfer the technology we have developed to Spectrum Diagnostix (SDx), a subsidiary of Physical Sciences Inc. They have commercialized several instruments initially developed at PSI on SBIR programs. Their primary product line currently consists of instrumentation used by the power generation industry but their long-term strategy is to expand into other environmental monitoring and management areas.

Acknowledgements

We appreciate the advice and support of our METC Project Managers, Mr. C. Edward Cristy (Phase I), Mssrs. Scott Renninger and Brian Frankhouser (Phase II). The overall program period of performance runs from October 1, 1993 to April 30, 1997.

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PI.7

Protective Clothing Based on Permselective Membrane and Carbon Adsorption

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OBJECTIVES

The objective of the program is to develop improved protective clothing for use by workers engaged in decommissioning and decontamination of former Department of Energy sites, including those used for atomic weapons research and production. Such sites are contaminated with a variety of hazardous compounds, ranging from asbestos, mercury and other heavy metals, to toxic organic compounds, such as PCB and chlorinated solvents, and radioactive metals and salts. Because of the hazards of exposure to these materials, workers must wear protective garments. These garments, which are made from Saran®, butyl rubber or other impermeable materials, provide excellent protection against particulates, liquids, aerosols, organic vapors and gases, but are impermeable to water vapor. Consequently, humidity and temperature within the suit rise rapidly during use, causing increasing discomfort. Heat stress occurs if the suit is worn for more than brief periods without resting.

The proposed technology concerns a new protective clothing fabric that combines a permselective membrane layer with a sorptive layer. If successfully developed, suits made from this fabric will offer equivalent, or better, protection than current materials, combined with a very high water vapor transmission rate (1,000 $g/m^2 \bullet day$ or more) that will dramatically improve "breathability," comfort, and worker productivity.

BACKGROUND INFORMATION

Over the next three decades, the Department of Energy faces an enormous decontamination and decommissioning task as facilities associated with research, development, and production of atomic weapons are closed. This task is complex and expensive because most sites are contaminated with a variety of hazardous compounds which range from asbestos, mercury and other heavy metals, to toxic organic compounds, such as PCB and chlorinated solvents, and radioactive metals and salts. Because of the hazards of exposure to these materials, workers must wear protective garments. These garments are impermeable to particulates, aerosols, and organic vapors and provide good protection from toxic contaminants. However, the garments are heavy, time consuming to don and remove, and most importantly, are impermeable to water vapor. Since the garments are water vapor impermeable, it is very difficult for body heat to escape. As a result, workers easily become heat stressed and must rest frequently.

Research sponsored by the U.S. Department of Energy's Morgantown Energy Technology Center, Under Contract DE-AC21-93MC30179 with Membrane Technology and Research, Inc., 1360 Willow Road, Menlo Park, CA 94025: (415) 328-2228

These frequent rests can significantly reduce worker productivity. The Heat Stress Limits recommended by the American Conference of Governmental Industrial Hygienists (ACGIH) are useful to demonstrate the productivity to be gained by a water vapor permeable suit (see Figure 1)[1]. Using this figure and making several assumptions (wet bulb globe temperature, WBGT, of 22°C; MTR suit performs like a winter work uniform whereas, conventional protective clothing performs like a water barrier), the MTR water-permeable suit will increase productivity by 25% — a substantial gain.

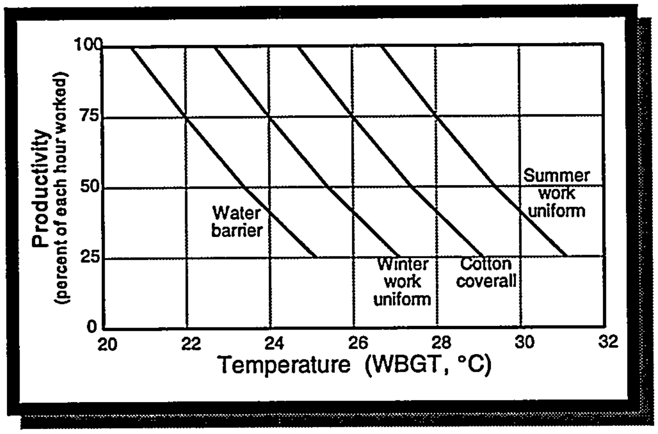

Figure 1. Heat Stress Limits Recommended by ACGIH

PROJECT DESCRIPTION

Membrane Technology and Research, Inc. (MTR) has been developing improved protective clothing that provides protection equivalent to that of current suits, but is water vapor permeable to minimize heat stress, and lighter weight for improved wearer comfort. The innovative feature of our improved fabric (see Figure 2) is an ultrathin, permselective outer membrane that is extremely permeable to water but impermeable to toxic organic compounds. The membrane layer protects the body from all particulate and liquid hazards and gives extended protection against organic vapors. To add a supplementary level of protection, the fabric has a sorptive layer, consisting of a porous membrane containing dispersed adsorbent. This layer increases the protective capacity against organic liquids and vapors and acts as a backup barrier in case the outer membrane is breached by abrasion or wear. The membrane layers are coated onto a conventional nylon fabric that provides mechanical strength. The water vapor transmission rate through the fabric is 600-950 g/m^2•day, compared to protective impermeable butyl rubber suits with transmission rates of 0-10 g/m^2•day, and non-protective porous Tyvek® suits with a transmission rate of 500-1,000 g/m^2•day.

This project is a multi-year, two-phase program to complete development of the fabric and to demonstrate its utility in field trials at the DOE Oak Ridge or Fernald sites. In the first phase of the program, the fabric properties, particularly the chemical resistance, water vapor transmission rate, durability, and flexibility, will be improved by modifying the materials used to form the membrane and the preparation procedure. Production of the fabric will then be scaled up to use commercial-scale production machinery. A small number of prototype suits will be made and a

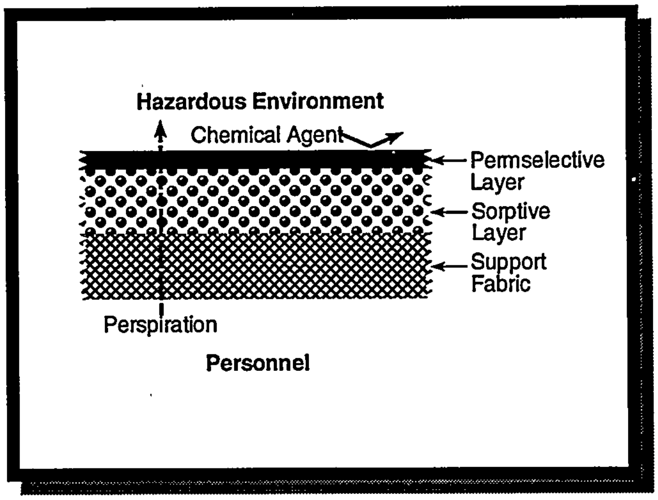

Figure 2. Protective Clothing Concept.

preliminary suit evaluation conducted. In Phase II, 300-400 suits will be produced for a complete laboratory and field demonstration program.

RESULTS

The Phase I project has three major objectives: fabric optimization, commercial-scale fabric production, and prototype suit evaluation. To date, the fabric has been optimized, production of commercial-scale fabric has been completed, and prototype suits have been manufactured. The following sections describe our results.

Fabric Optimization

Fabric optimization involved optimization of the individual layers included in the final protective fabric, and optimization of the way these layers are combined. The layers include the support fabric, the sorbent layer (including the sorbent used, the polymer used, and the ratio of the two), and the permselective layer (including the polymer and post-treatment methods). We also studied the geometry used to combine the individual layers into the final protective fabric. Details of the fabric optimization results were reported previously.^{2}

Commercial-Scale Fabric Production

Commercial-scale fabric production includes scaling up from the 12-inch-wide machines used during fabric optimization to 40-inch-wide machines, and producing at least three to five rolls of 40-inch-wide fabric at least 100 m long.

Scaling up from the small to large machines required a number of trials to ensure that the chosen fabric ran smoothly through our machines with even tension across the fabric, and that the solution used to cast the sorbent layer did not bleed through the membrane.

Modifications made during scale-up include adding new rollers to keep the tension even and prevent wrinkle formation, adjusting the casting solution viscosity and casting conditions to prevent bleed-through, and having the support fabric re-cut to eliminate the fabric edges from snagging in the machines.

Our final production runs were made in conjunction with a commercial laminator. To protect the permselective layer, two layers of the fabric (Figure 2) were laminated, permselective layer to permselective layer. This final fabric structure, pictured in Figure 3, protects the permselective layer on both sides.

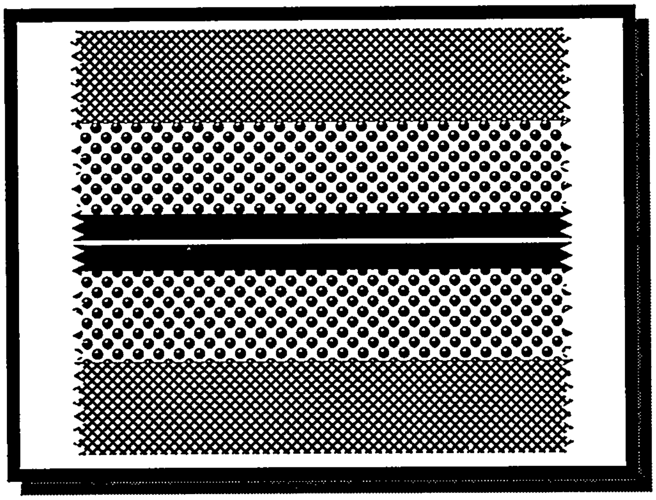

Figure 3. Structure of Protective Clothing Fabric

Prototype Suit Evaluation

To evaluate the prototype suits we will use a sweating mannikin. This approach eliminates many of the variations in other parameters (how each subject sweats in response to heat stress, how each subject is feeling that day, etc.) inherent to actual human tests. A large sample is required to overcome these variations, and because only a limited number (18) of prototype suits were produced, human tests are not practical during this phase. (In Phase II large-scale human tests at an actual DOE site are planned.)

The sweating dummy allows measurement of the heat transfer characteristics of a suit under controlled, reproducible conditions. The heat transfer characteristics include both dry heat transfer (heat conduction through the suit) and heat transfer due to evaporation of sweat. The data from these tests can be used with models (based on human performance data) to predict the productivity of an average worker wearing that suit. Our prototype suits will be tested alongside conventional protective clothing (Saranex-coated Tyvek) to determine the productivity advantage of our suit over current clothing.

To evaluate the economic viability of the MTR fabric, we estimated the paying price for a suit made from MTR fabric. Because the MTR suit allows a worker to actually work a greater fraction of an 8-hour day (because fewer rest breaks are needed than for a worker using a conventional protective suit), the employer should be willing to pay more for the MTR suit than for a conventional suit. Assuming that a worker wearing a conventional suit requires one hour more rest than a worker using the MTR suit, the extra that the employer should be willing to pay for the MTR suit is the cost for one hour of work. This cost includes direct labor costs (the worker's wages) and indirect costs (benefits, supervision, and the cost of equipment used by workers). A conservative estimate includes a worker's direct wages of $15/hour and indirect costs of the same amount, for a total cost of $30/hour. Thus, an employer should be willing to pay over $60 for a suit made from the MTR fabric: $31 (price for a conventional suit) plus $30 in extra productivity (Figure 4). This result suggests that the MTR suit would be economically competitive if it could be manufactured on a large scale (~50,000 suits per year).

FUTURE WORK

Our final step in Phase I is to evaluate the effectiveness of the suits in reducing heat stress, based on results from the sweating mannikin test. If the suits show promise, we will go on to Phase II of the project. In Phase II we will manufacture 300 - 400 suits and perform head-to-head field trials versus the clothing currently used at the field site. If the field trials demonstrate that the suits made from MTR fabric increase productivity, maintain good protection, and are durable, we will, with our manufacturing partner, begin commercial marketing and production of protective clothing made with the MTR fabric.

ACKNOWLEDGEMENT

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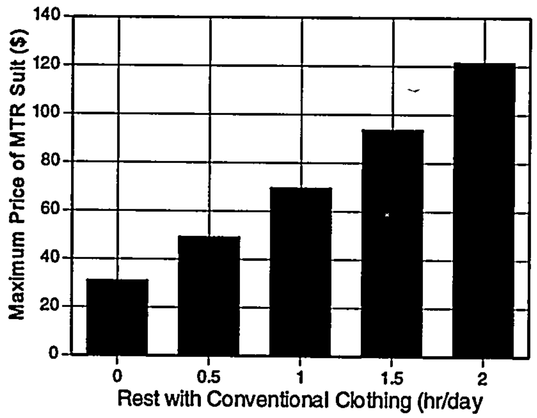

Figure 4. Maximum Price of MTR Suit Due to Higher Productivity

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PI.8 Coherent Laser Vision System

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1.0 Introduction

The Coherent Laser Vision System (CLVS)[1] is being developed to provide precision real-time 3D world views to support site characterization and robotic operations and during facilities Decontamination and Decommissioning. Autonomous or semiautonomous robotic operations requires an accurate, up-to-date 3D world view. Existing technologies for real-time 3D imaging, such as AM laser radar, have limited accuracy at significant ranges and have variability in range estimates caused by lighting or surface shading.

Recent advances in fiber optic component technology and digital processing components have enabled the development of a new 3D vision system based upon a fiber optic FMCW coherent laser radar. The approach includes a compact scanner with no-moving parts capable of randomly addressing all pixels. The system maintains the immunity to lighting and surface shading conditions which is characteristic to coherent laser radar. The random pixel addressability allows concentration of scanning and processing on the active areas of a scene, as is done by the human eye-brain system.

The precision measurement capability of the coherent laser radar (CLR) technology has already been demonstrated in the form of the CLR 3D Mapper (Figure 1-1), of which several copies have been delivered or are under order. The CLVS system, in contrast to the CLR 3D Mapper, will have substantially greater imaging speed with a compact no-moving parts scanner, more suitable for real-time robotic operations.

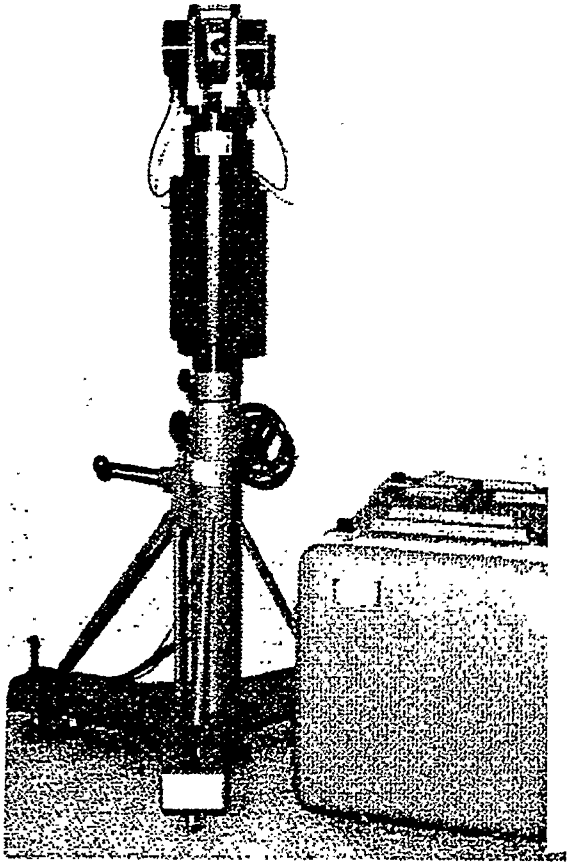

Figure 1-1. CLR Fiber Optic Precision 3D Mapper

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[1] Research sponsored by the U.S. Department of Energy's Morgantown Energy Technology Center, under Contract DE-AR21-94MC31190 with Coleman Research Corporation, 6551 Loisdale Ct., Suite 800, Springfield, VA 22150; telefax: 703-719-9229.

Fiber-optic coherent laser radar (CLR) based systems such as the CLVS are immune to variations in lighting, color, or surface shading, which have plagued the reliability of existing 3D vision systems, while providing substantially superior range resolution. The primarily fiber-optic construction of the CLVS will be more resistant to shock and thermal effects than bulk optic systems, and will be more economical to manufacture and maintain.

The superior accuracy of the CLVS is a direct result of the orders of magnitude greater signal bandwidth involved in the coherent range detection process. The coherent detection applied to optical frequency modulation (FM) is also the source of the CLVS's immunity to amplitude distortions by lighting or surface shading, just as FM radio, unlike AM radio, suppresses noise from lightning bursts. The fiber-optic implementation of the CLVS allows a more sophisticated optical circuitry to be employed than would be practical with bulk optics, and with less cost for assembly and alignment during manufacturing.

2.0 Objectives

The CLVS development program is a progression towards a 256x256 pixel, one frame per second 3D vision system with an rms range accuracy better than 1mm for a 5m working range and a maximum range of approximately 10m. The limitation in working range is a function of the digital processing power which will be employed by the CLVS receiver and is readily extendable in future systems.

The CLVS fieldable prototype is being developed by Coleman Research Corporation (CRC) in a two phase program.

During the current development phase a baseline CLR 3D vision demonstration system is being developed with the following projected performance:

Frame size:	(128 x 128) 3D coordinates
Frame speed:	1 frame per second
Range accuracy:	$\sigma_R = 1\text{mm}$
Scanner:	No-moving-parts

During the second phase, the baseline system will be developed into a fieldable prototype 3D vision system with expanded performance parameters including the ability to scan a (256 x 256) frame of 3D measurements at a one frame per second rate. This enhanced performance will be accomplished by the implementation of "smart receiver" processing algorithms which maximize the use of a priori range information to streamline the real-time CLR range processing computation. The effort will also yield a compact, no moving parts fieldable prototype 3D vision scanner.

The fieldable prototype CLVS will output both range and intensity images on both standard video and digital interfaces.

3.0 Approach

The Coherent Laser Vision System (CLVS) is a fiber optic coupled FMCW coherent laser radar. The radar uses the relatively large tuning range of injection laser diodes to achieve greater precision than available with other techniques. As shown in Figure 3-1, the optical frequency of the laser is swept linearly as a function of time. The laser output is divided and used both as a local oscillator (L.O.) and as the signal to be transmitted. After being time delayed by the round trip transit time to the target, the received signal is mixed with the optical L.O. on a photodetector. The resultant beat frequency is equal to the sweep rate of the optical signal

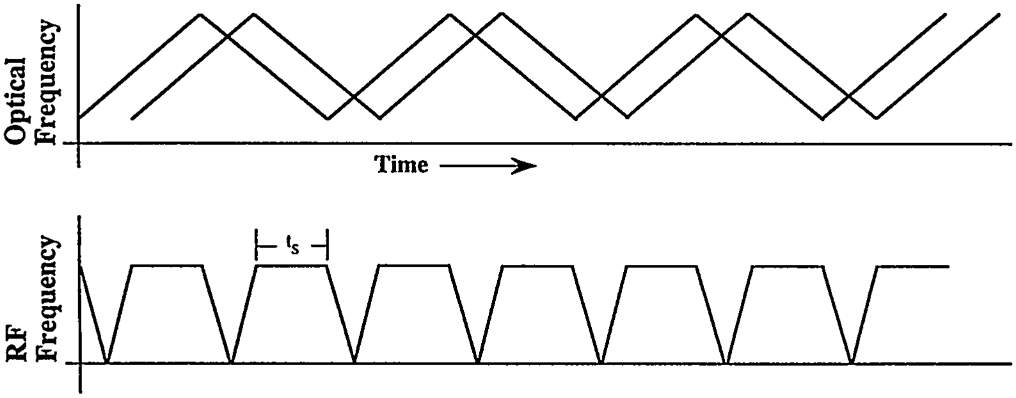


Figure 3-1. Laser Optical Frequency and Heterodyned RF Signal Coherent Laser Radar

multiplied by the time delay between the received signal and the local oscillator. Since this time delay is proportional to target distance, the RF beat frequency is also proportional to target distance.

Due to the short wavelength of the laser, a small wavelength deviation results in a large beat frequency. For example, at an optical wavelength of 1550 nm, a shift in wavelength of only 1 Angstrom (0.1 nm) results in a frequency shift of 12 GHz. This frequency modulation is accomplished by modulating the laser's injection current, thereby thermally tuning the laser wavelength. The basic fiber optic coherent laser radar system optical configuration is shown in Figure 3-2.

The FMCW coherent laser radar's source consists of a 1550 nm single mode diode laser pigtailed to an optical fiber. The laser's optical output is frequency modulated by varying its injection current. In the basic configuration shown, a small portion of the output light is directed into the L.O. path via a power tap. The remainder of the light is focussed by the antenna lens into the range measurement area of interest. Light reflected from a surface in this area is recollected by the lens, directed into the return signal path by the optical circulator by means of a polarization diplexing scheme and mixed with the L.O. light at the second 3dB coupler. By utilizing two detectors and a differential amplifier, common mode rejection of amplitude noise is achieved.

Rms range error is proportional to 1/(Tuning) and proportional to 1/(pixel dwell time)$^{1/2}$. This is true for all radar system, AM or FM as expressed in equation 1.

$$\sigma_R \ \alpha \ \frac{1}{(\Delta f)\ T^{1/2}\ (SNR)^{1/2}} \tag{1}$$

This relationship indicates that, with the coherent FM radar CLVS enjoying a 20 to 50 tuning bandwidth advantage over AM radars of the same power level, it obtains far greater accuracy for a given dwell time. Taking this relationship another way, for a given accuracy level the AM radar must have a dwell time from 400 to 2500 times as long.

For a CLR vision system the capabilities and operation of the laser transmitter subsystem,

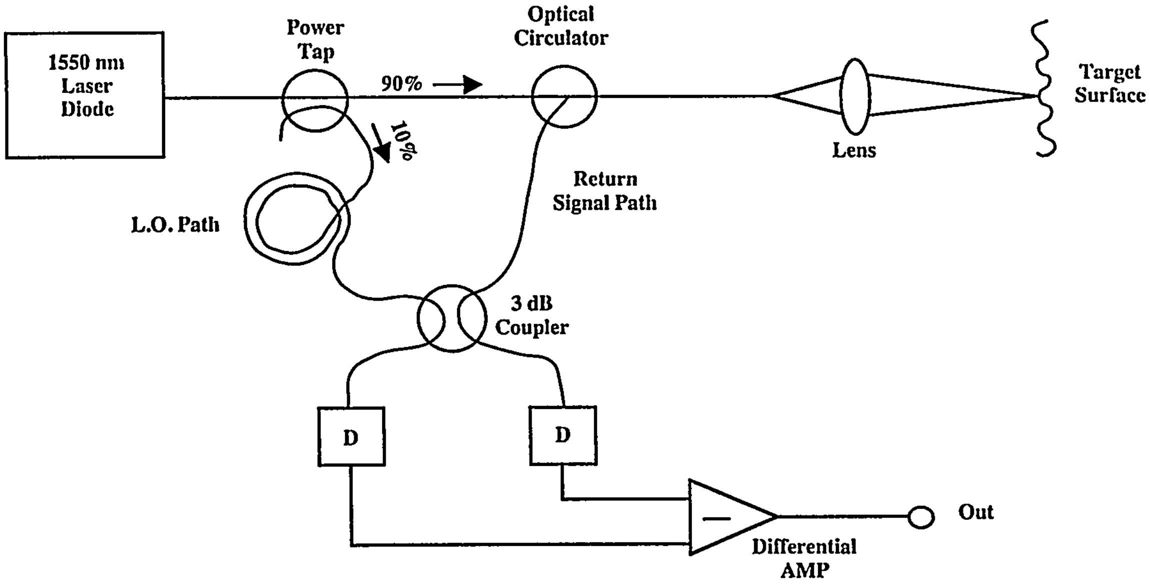


Figure 3-2. Basic Fiber Optic FMCW CLR Configuration

the scanner subsystem, and the receiver-range processor are intertwined and will be discussed as they impact receiver requirements and overall system performance. All optical components considered for the CLVS are fiber coupled, which insures a more rugged, more easily manufacturable system than can be constructed with bulk optics. An overall system block diagram is given in Figure 3-3.

3.1 The laser transmitter

The laser transmitter is based upon a fiber pigtailed laser diode which is frequency modulated by varying the laser drive current. Each linear FM chirp must be accomplished in approximately 50 µsec for a 128 x 128 pixel frame, one frame per second baseline scan rate. Wide FM chirps are desirable because they lead to greater range accuracy (see Eq. 1 above).

3.2 The No-Moving-Parts Scanner

Up to this time the scanner has been a critical and problematic component of laser radar 3D vision systems. In order to move a laser beam over the desired angular field of view at the desired frame rates, a bulk optic based scanner has necessarily involved fairly large optical mirrors moving at substantial angular speeds. The mechanical momentum of the mirrors requires that adjacent pixels be scanned in sequence and limits the scanning rate of the 3D vision system.

Scanning for the CLVS will be accomplished with no-moving parts based upon acousto-optic beam deflection, as shown in Figure 3-4.

With this configuration a pair of Bragg cell acousto-optic beam deflectors position the beam

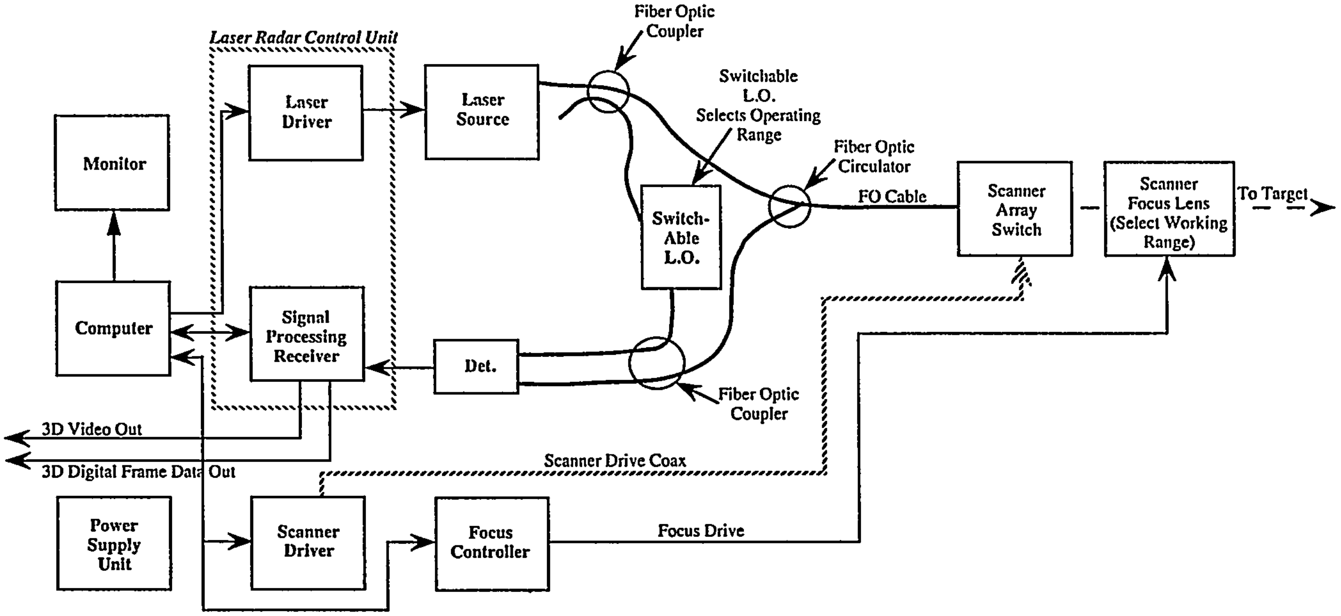


Figure 3-3. Fiber Optic CLR 3D Vision System Block Diagram

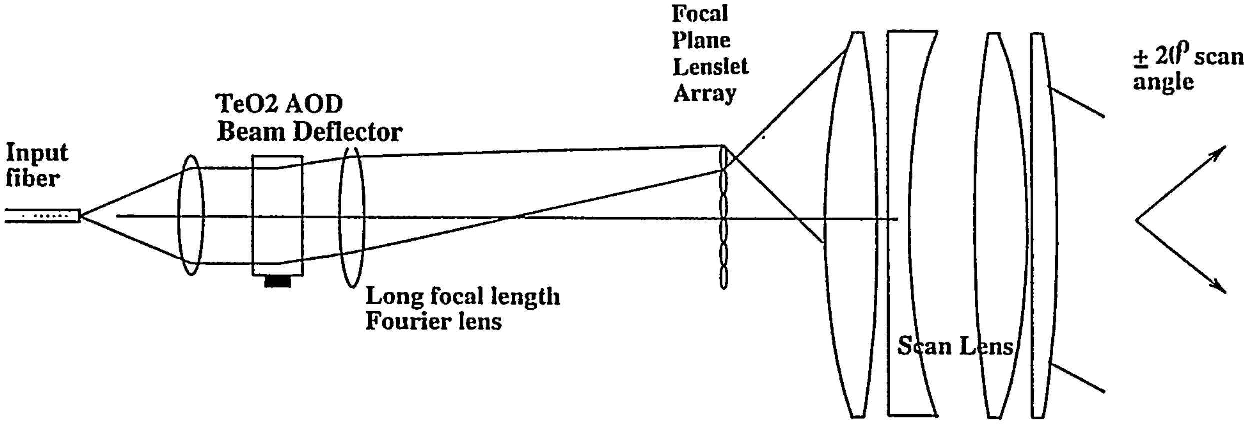


Figure 3-4. Acousto-Optic Scanner Geometry

at selected positions of a rectangular lenslet array. The lenslet array acts like focal plane "film" for a 3D camera, expanding the beam to fill the aperture of the scan lens. By means of this geometry the small beam deflection accomplished by the AO device is magnified to accomplish a practical angular field of view. The inter pixel transition time is limited only by the few microseconds it takes the acoustic signal to clear the Bragg cell, and is independent of pixel position. This will allow for intelligent 3D image acquisition, which concentrates scanning activity on the active, more interesting, parts of the scene, as shown in Figure 3-5.

3.3 The Digital Receiver

A number of companies are now producing high speed digital processing boards which singly, or in a cascaded architecture, can accomplish the 3D CLR receiver processing task. In general, a CLR 3D vision system will process one laser FM chirp for each pixel of an (n x n) frame at the operating frame rate. The processing consists of the detection of the beat frequency of the mixed local oscillator and target return signals. The bandwidth of potential signals is defined by the chirp rate (which determines the frequency offset per meter of target range) and the depth of range of the region being 3D imaged.

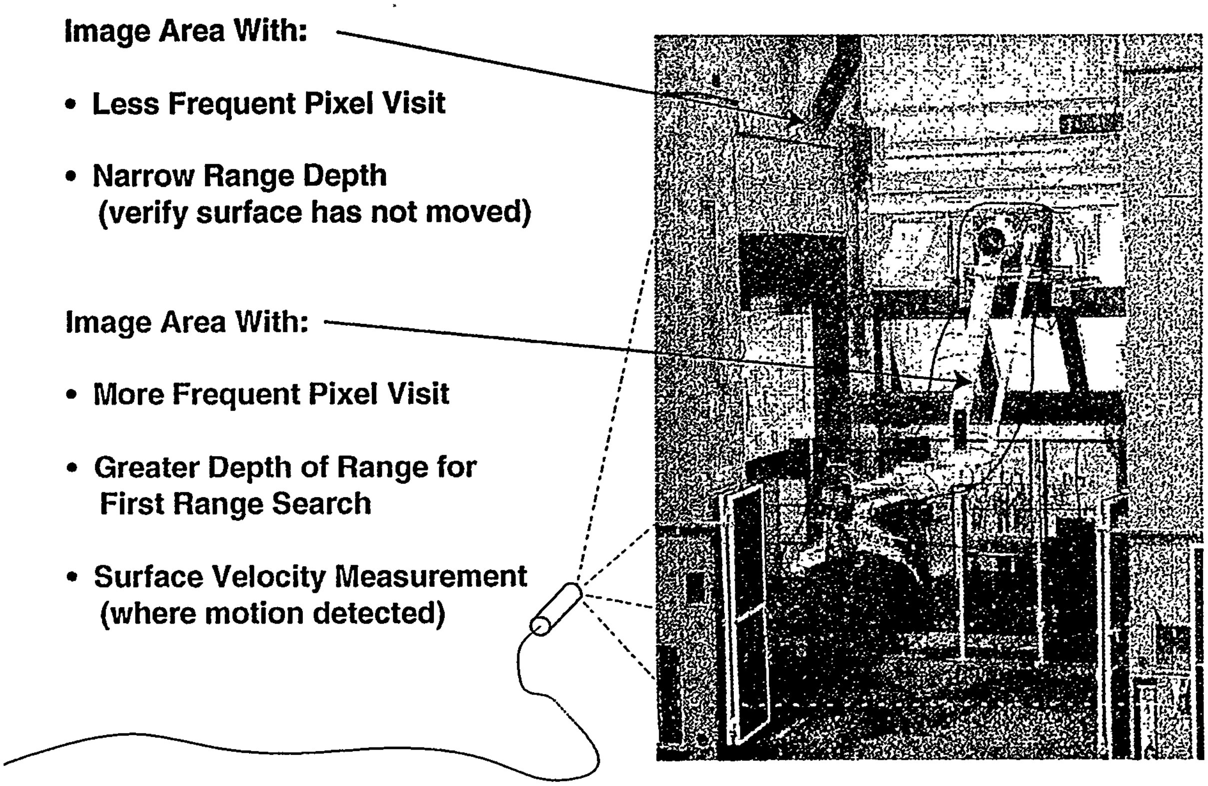


Figure 3-5. CLR 3D Vision, Smart Image Sampling

The bandwidth of potential signal frequencies and the dwell period per pixel define the sample rate and the total number of samples per pixel. This, combined with the total pixel rate, yields the total average rate in samples per second at which the digital receiver must process the detected radar output.

Commercial DSP hardware suppliers can now deliver configurations which will perform these processing steps at the rates required and have already demonstrated this capability on previous (microwave) radar applications.

The receiver configuration of Figure 3-6 computes a new range for every pixel of every frame, using no a priori knowledge of range. It seeks a range detection over the full working range of the 3D vision system as a first and only range estimation step. This would be an acceptable approach if cost were not a consideration. Since cost is a consideration we are motivated to pursue a "smart receiver" approach (see Figure 3-7) which use a priori knowledge to reduce processor load. This approach will be implemented in Phase II of the CLVS development program.

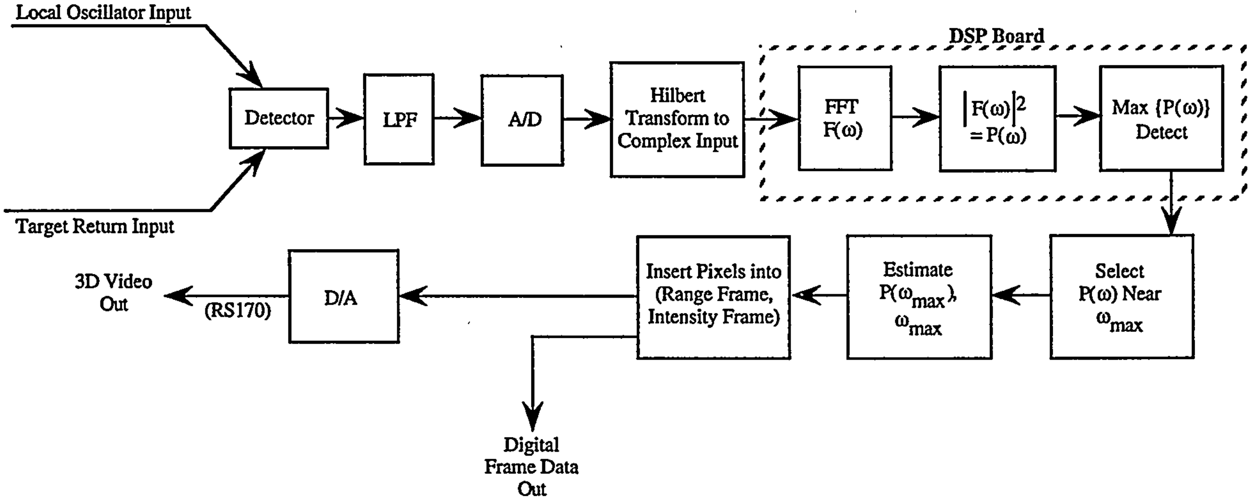


Figure 3-6. CLR 3D Vision Receiver Processor (No "Smart" Receiver Staged Range Detection)

The range for each pixel, along with the known azimuth and elevation for that pixel, constitute a 3D location in "radar coordinates." Radar coordinates will generally be processed into rectangular coordinates by subsequent 3D scene processing hardware which supports robotic operations, 3D scene analysis, or multi-perspective remote viewing.

4.0 Accomplishments

During the current Phase I of the CLVS development program, all subsystems have been designed. Two noteworthy areas of achievement in design and component designation are the laser diode source and the AO beam deflector.

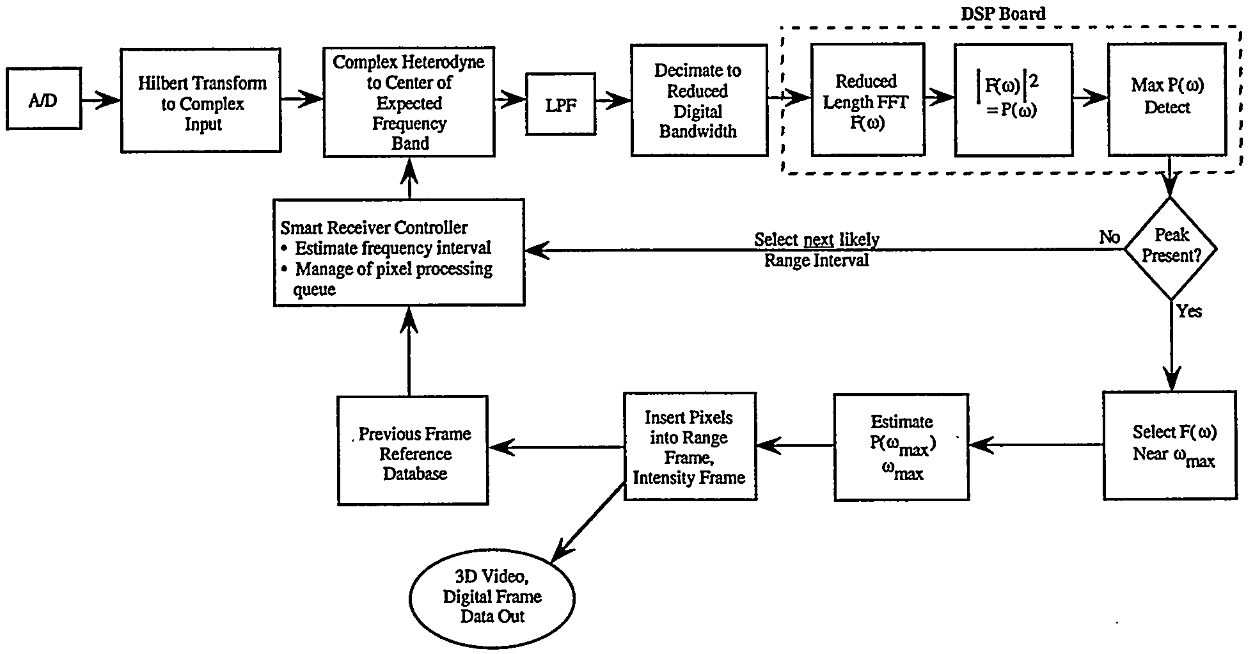


Figure 3-7. CLR 3D Vision "Smart" Receiver Processor

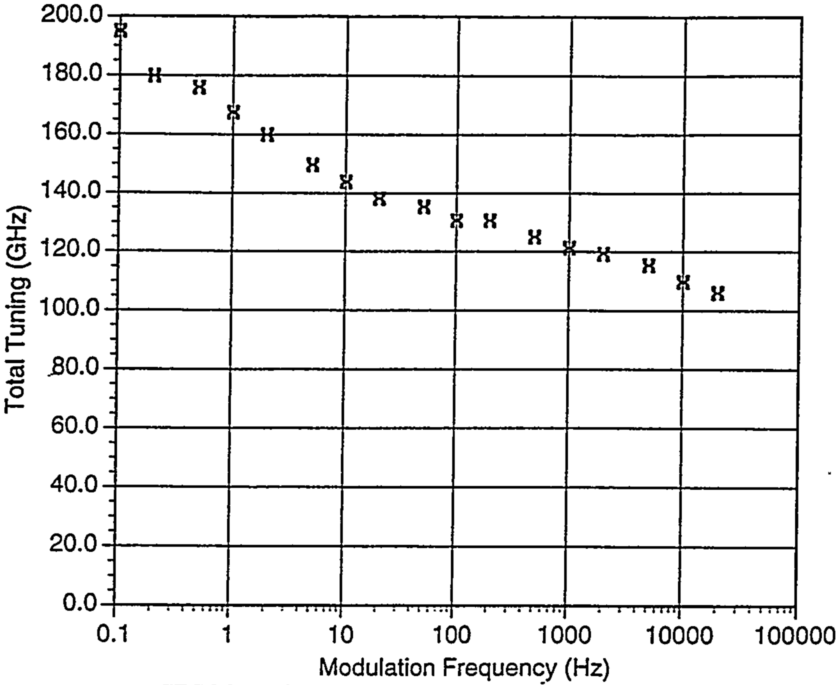
4.1 The Laser Diode Source Improvements

A new laser diode source type has been tested which promises substantially improved CLVS performance over earlier expectations. Laser devices for coherent laser radar (CLR) applications are selected for FM tuning range, maximum tuning speed and minimum linewidth (narrow linewidth means a stable instantaneous laser frequency). The importance of these qualities is as follows:

FM Tuning Range. The range measurement error is proportional to the reciprocal of the laser FM modulation bandwidth Δf (see Figure 4-1). More FM tuning range means more range accuracy.

Tuning Speed. The tuning speed determines how many tuning cycles per second may be accomplished by the laser diode. At least one half tuning cycle must be accomplished during each pixel dwell for a CLR 3D vision system. For example, for a 128 x 128 pixel frame, one frame per second, there must be 16,384 pixels per second and 8192 tuning cycles per second. Generally speaking, the tuning frequency range decreases as the tuning speed increases, because of the thermal inertia of the laser diode. Thus tuning speed and pixels per second for a given diode type are limited by the maximum tuning speed which is possible with the necessary tuning bandwidth for the required CLR vision system accuracy.

Laser Linewidth. The laser linewidth is the bandwidth over which the laser frequency wanders spontaneously. Two parts of the laser waveform separated by more than the "coherence time" from each other will no longer have a stable phase relationship with each other and will not support a high SNR range measurement. This coherence time T_c is the reciprocal of the laser linewidth. A coherent laser radar cannot operate



GEC-Marconi LD4204 #25 Wavelength Tuning Response for tuning from 55-295mA. Data extrapolated from tuning about 200mA +/-5mA. Passive heat sinking at room temperature (no active temperature control).

Figure 4-1. GEC-Marconi LD4204 #25 Wavelength Tuning Response

effectively over greater distances thank $R = c\ T_c/2$ unless a fiber optic delayed reference (local oscillator) signal is used. For the measurements rates of a 3D vision system it is not practical to use a variable delay local oscillator. Thus the narrower the laser linewidth, the longer the depth of range for a CLR vision system.

Figure 4-1 shows tuning range vs. modulation frequency for a new sample laser diode device provided by GEC Marconi. AT 10 kHz, the approximate tuning frequency region for CLVS, a modulation bandwidth of more than 100 GH is possible. This compares with a 20 Ghz bandwidth potential for previously available devices at the eyesafe wavelength of 1.5µm. The linewidth of this new device is 0.5MHz, compared with 5 MHZ for the previously available device. This will ensure no significant coherence length limitation for a depth of range of at least 30M. Range depth will be limited by other factors which as receiver processing capacity and increased spreading loss at long range. With the new laser diode device coherence length will not be a performance limiting factor within the 10M maximum range depth to be considered by CLVS.

4.2 AO Beam Deflector Design

The compressions and rarifications of the AO beam deflector Bragg cell crystal by the acoustic wave change the index of refraction in a spatially periodic fashion, forming a defraction grating. The efficiency with which a single deflected beam is formed depends on the crystal material, orientation, and the amplitude of the acoustic wave. Bragg cells used in optical computation devices are normally not very efficient wasting over 90% of the laser beam energy in a single pass. For the CLVS scanner the laser beam must pass through the Bragg cell twice, once in each direction. Thus, very high

Bragg cell efficiency is imperative for the CLVS scanner. TeO_2 has been selected as the Bragg cell material because of its high efficiency and substantial attention has been paid to achieving high acoustic modulation without overheating the Bragg cell. Many design factors are also considered, including Bragg cell frequency shift, polarization and other effects of the Bragg cell as part of the scanner optical subsystem. The Phase I design is complete and components ordered. AO no-moving parts scanner will be accomplished and integrated as part of a CLVS laboratory demonstration in the Phase I development.

5.0 Applications

The highly accurate, reliable real-time 3D images provided by fielded CLVS systems will provide an improved, dependable world view to plan and guide robotic and remote operations for facility and characterization and facility D&D. In general, the CLVS will overcome the problems of current 3D vision technology - lack of resolution and sensitivity to ambient lighting or surface shading. CLVS will monitor the 3D position of all pixels of a scene simultaneously, thus keeping track of the whole scene, not just one tracked object. The generated 3D image can be overlaid on color video, infrared or other raster-scanned two dimensional images.

CLVS monitoring is intended for scene altering operations such as:

- structure and equipment dismantling
- equipment moving or removal
- waste retrieval
- surface scarafacing
- excavation
- autonomous vehicle operation

A significant fraction of the whole three dimensional scene, thousands of pixels, may change in a few seconds. There is still the need in robotic operations to move precisely while gripping, shoveling or otherwise interacting with scene objects or avoiding collisions even though positions of objects often may not be precisely predictable. Variable lighting conditions and surface shadowing will preclude reliance on 3D vision systems which are sensitive to these environmental factors.

We have developed techniques for accomplishing six degree of freedom (6DOF) end effector guidance using laser radar tracking of a known geometry target. With the high speed scanning possible with CLVS, this 6DOF end effector tracking may be accomplished at high speed.

The simple configuration of the CLVS scanner lends itself to future compact and economical implementations. (See Figure 5-1).

This will lead to many commercial monitoring and sorting applications involving personnel and object recognition. Patient monitoring and 3D surgical guidance are potential medical applications.

The CLVS scanner configuration contains no solid state electronic components and thus is useable in high (10^6 rad) radiation environments. An adaptation of this design is being evaluated as part of the International Thermonuclear Experimental Reactor (ITER) 3D Remote Mapping System (3D-RMS) under subcontract to Oak Ridge National Laboratory (Figure 5-2).

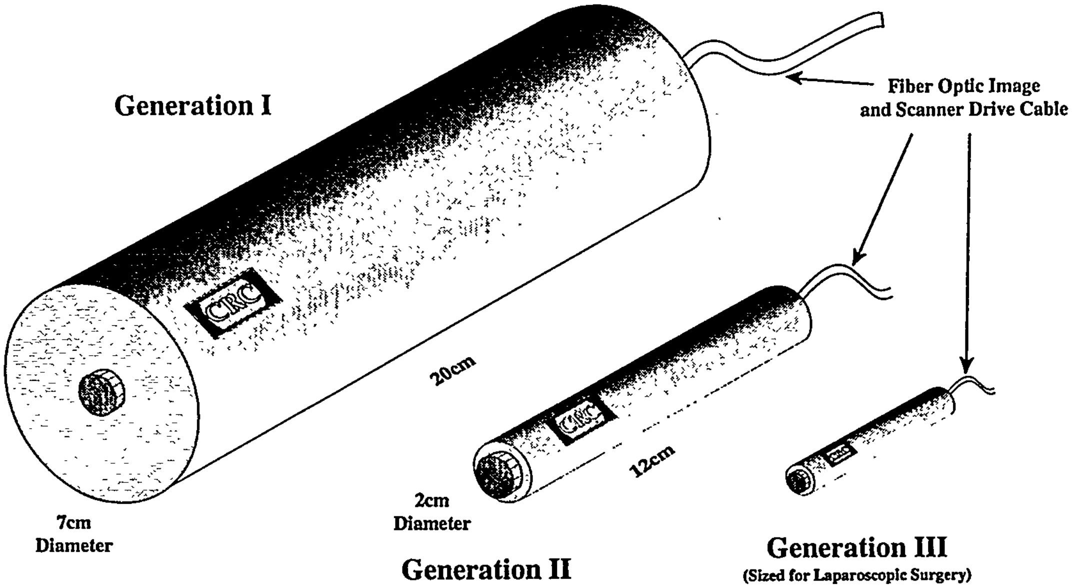


Figure 5-1. Fiber Optic CLR 3D Camera

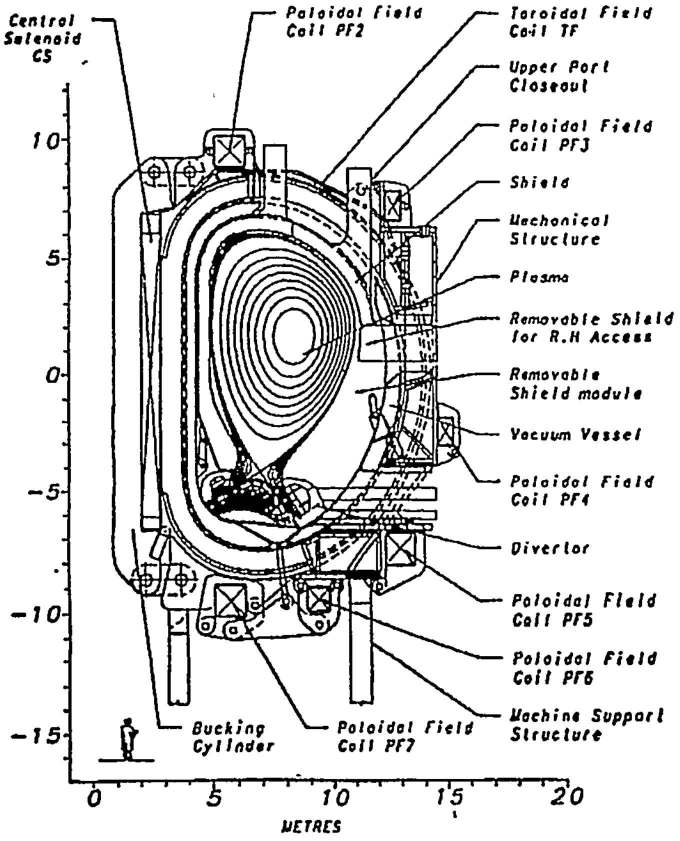


Figure 5-2. ITER Plasma Chamber

6.0 Future Activities

In the second phase of the CLVS development program the CLVS scanner will be implemented as a compact fieldable prototype and the scanning frame density will be increased from 128 x 128 pixels to 256 x 256 pixels. The digital receiver processing will be made "smart" in order to manage the greater number of pixels and to increase the instantaneous depth of range to five meters.

6.1 The "Smart" CLVS Digital Receiver

For each pixel range estimate the "smart" receiver stores and makes use of the range of that pixel and surrounding pixels for the previous frame. The smart receiver also retains knowledge of which pixels have changed recently. By this process the receiver has the ability to center the first round of a staged range search on a range which is likely to be very near the correct range for that pixel. A number of strategies for this search may be pursued which may depend upon the application, the environment, or the portion of the image being processed. For example:

(1) A robot in the center of the working range operates in front of a background in the rear of the working range. In this case, when the robot arm no longer occupies a pixel, the background surface does and the range to that surface defines the second interval to search.

(2) For a CLR 3D vision system on a continuously moving platform, surface points move in range and progress from pixel to pixel in an organized fashion. This organized progression of surface geometry will be approximately predicted, using also the range velocity available directly from each pixel, to determine the most likely range interval for each pixel for each frame.

It should be emphasized that only very approximate predictions of pixel range are necessary to achieve substantial reductions in required processing capacity. For example, if, on the average, pixel range can be predicted within a 1 m interval rather than a 10 m working range, a factor of ten savings in required spectrum analysis processing power is achieved. With this advantage a single board processor will be able to support a (256 x 256) pixel frame per second rate. For the lower density (128 x 128) pixel frame, a higher frame rate than 1/sec may be supportable. The smart receiver coupled with a switched fiber optic scanner which allows random pixel addressability will allow the scanning to visit those pixel locations most frequently which are in the vicinity of the changing parts of the 3D scene. This will allow further increase in receiver efficiency, improved capability to follow changing scenes, or both.

For a "smart" receiver it is understood that, when the CLR 3D vision system is first turned on several frame periods may be required to conclude a search for range for each pixel. This search will be aided by the fact that neighboring pixels will tend to have similar range.

With the use of a fiber scanner it is possible to alter the scanned pixel density or the frame rate programmable and, thus, adjust the processed pixel density to match the predictability of the pixel ranges and the processing capacity of the receiver's DSP hardware. When operating at low density, the 3D video output could continue at high density with (2 x 2) pixel blocks tied to the same range.

6.2 Impact of Expected DSP Hardware Performance Growth on CLVS Performance

DSP hardware manufacturers predict that, within two to four years, the brute force (no smart receiver) processing loads required by one 256 x 256 frame per second over a 10m range, will be manageable by a single, general purpose, DSP board. This expected hardware performance growth will not remove the benefit of a "smart" receiver architecture. The reduction in processing load for a given pixel density and frame rate can always be exploited to improve pixel density, angular aperture or frame rate. In summary, a "smart" receiver should allow the production of a CLR 3D vision system which processes a (256 x 256) pixel frame at one frame per second, for a 10 m depth of range with the same DSP hardware which will be able to process a (128 x 128) pixel frame with 5 m depth of range without a "smart" receiver.

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PI.10 Mixed Waste Treatment Using the ChemChar Thermolytic Detoxification Technique[1]

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Introduction and Need

The diversity of mixed waste matrices contained at Department of Energy sites that require treatment preclude a single, universal treatment technology capable of handling sludges, solids, heterogeneous debris, aqueous and organic liquids and soils. Versatility of the treatment technology, volume reduction and containment of the radioactive component of the mixed waste streams are three criteria to be considered when evaluating potential treatment technologies. Thermal technologies typically offer waste stream versatility and volume reduction, but suffer from potential volatilization of contaminants, most importantly radioactive species, and thus require extensive off-gas treatment trains. The need for a treatment system that can accept a wide variety of waste matrices, achieve regulatory destruction efficiencies for contaminants and produce a clean effluent is highly desirable.

Approach

The ChemChar thermolytic detoxification process being developed under this R&D contract is a thermal, chemically reductive technology that converts the organic portion of a mixed waste stream to an energy-rich synthesis gas while simultaneously absorbing volatile inorganic species (metals and acid gases) on a macroporous, carbon-based char. The latter is mixed with the waste stream prior to entering the reactor. Substoichiometric amounts of oxidant are fed into the top portion of the cylindrical reactor generating a thin, radial thermochemical reaction zone. This zone generates all the necessary heat to promote the highly endothermic reduction of the organic components in the waste in the lower portion of the reactor, producing, principally, hydrogen and carbon monoxide. The solid by-product is a regenerated carbon char that, depending on the inorganic loading, is capable for reuse. The in situ scrubbing of contaminants by the char within the reactor coupled with a char filer for final polishing produce an exceptionally clean synthesis gas effluent suitable for on-site generation of heat, steam or electricity. Despite the elevated temperatures in the thermochemical reaction zone, the reductive nature of the process precludes formation of nitrogen oxides and halogenated organic compound by-products.

Project and Technology Description

The principle objective of this Research Opportunity Announcement program will be to treat two surrogate mixed waste streams in a four inch diameter, continuous feed, adiabatic reactor (Figure 1) to regulatory treatment levels for the organic contaminants and simultaneously absorbing the radioactive surrogate

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Research sponsored by the U.S. Department of Energy's Morgantown Energy Technology Center under contract DE-AR21-95MC31188 with Mirage Systems, 232 Java Drive, Sunnyvale, CA 94089; telefax: 408-734-8845.

species on the char. Extensive sampling of the gaseous effluent will be performed throughout the duration of process runs to ensure destruction of organic contaminants and absorption of inorganic species.

The surrogate mixed wastes to be tested under the program include an aqueous based surrogate containing four soluble Resource Conservation and Recovery Act (RCRA) metals, two radioactive surrogate metals and several RCRA organic compounds (chlorinated and non-chlorinated, see Table 1). The second surrogate mixed waste to be tested is an organic sludge with the same RCRA and surrogate radioactive components as the aqueous waste (see Table 2). The anticipated ten month program has two main project tasks: (i) design and construct a four inch, adiabatic, continuous feed ChemChar reactor at Mirage Systems in Sunnyvale, California; (ii) treatment of the mixed waste surrogates at the University of Missouri, Columbia campus.

Accomplishments

The ChemChar waste treatment process has been tested on the bench scale in two inch diameter, non-adiabatic, batch reactors on a variety wastes at the University of Missouri over the past eight years. These successful, bench-top treatability studies have proven the principle reductive attributes of the process for the conversion of waste materials to a valuable, energy rich, synthesis gas. numerous wastes matrices and contaminants were treated by the ChemChar process including soils, organic tars and sludges, still bottoms, and radioactive metal containing ion exchange resins. Specific contaminants included PCBs, chlorinated benzenes, and volatile RCRA metals. The outcome of these studies consistently resulted in >99.99% destruction and removal efficiency for the organic components and gaseous effluents free of acid gases (hydrochloric acid and nitrogen oxides) and volatile metals (cadmium, mercury, lead). The current program will focus on system optimization and heat management for a four inch diameter reactor for accumulation of data to be used for construction of a pilot system.

Application/Benefits & Future Activities

Successful completion of this program will result in a versatile mixed waste treatment technology ready for pilot plant scale-up. The principle benefit will be the eventual commercialization of a mixed waste treatment system capable of handling a significant volume of the current inventory of wastes across the DOE complex. This recently initiated program will focus initially on the design and construction of a continuous feed, computer controlled ChemChar system, followed by transport of the reactor to the University of Missouri for mixed waste surrogate testing. Pilot scale design and construction is planned for mid-1996.

Acknowledgment

Support for this contract has been provided through Mr. Richard Dennis (the project COR) and METC. The period of performance for this program is 6/95-4/96. The research to be performed at the University of Missouri is headed by Dr. David Larsen (St. Louis) and Dr. Stanley Manahan (Columbia). This program is under the Mixed Waste Focus Area of EM.

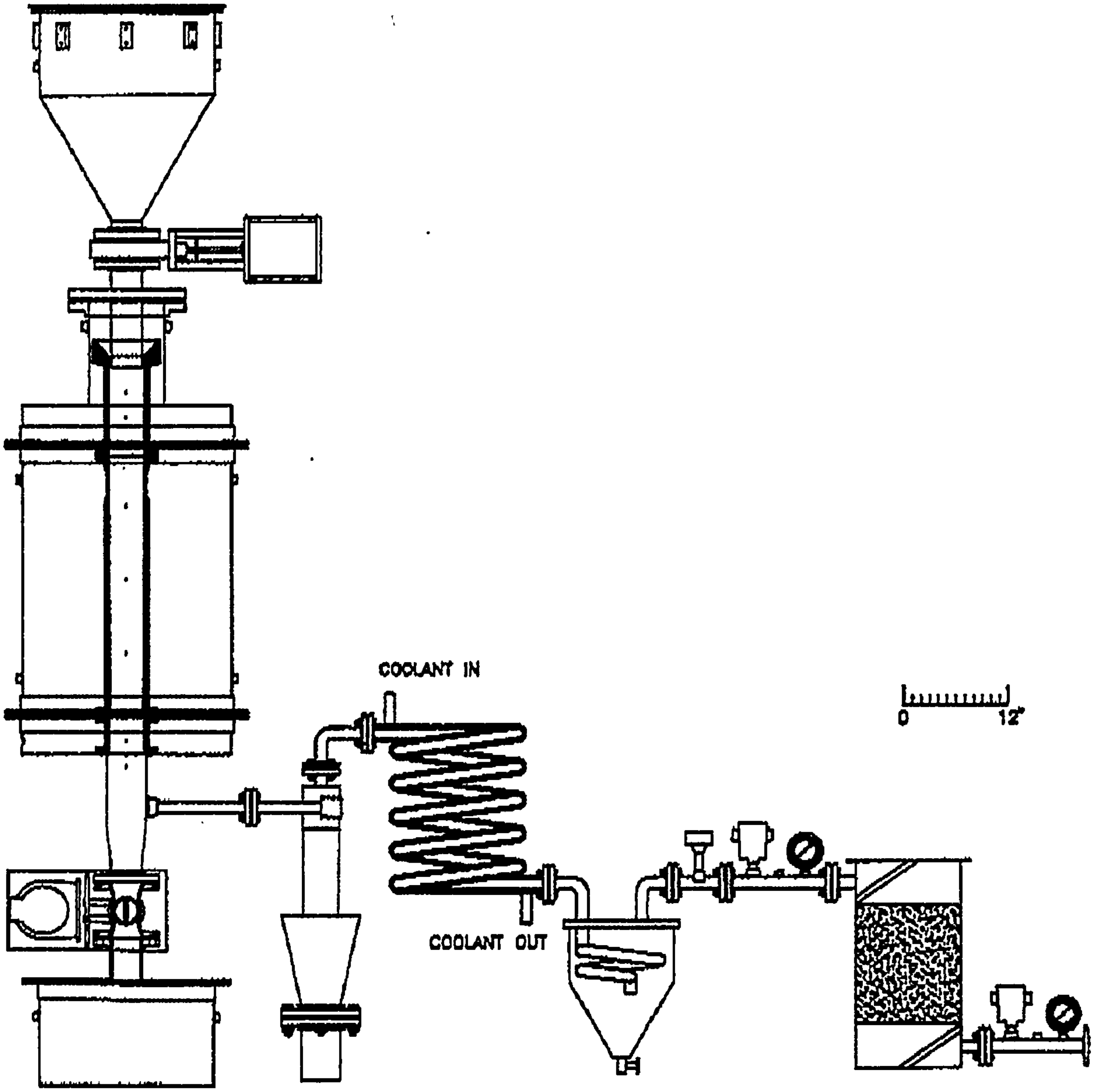

Figure 1. Schematic diagram of the continuous feed ChemChar reactor to be constructed under this program.

Table 1
Composition of the High Organic Sludge Surrogate Mixed Waste

No.	Component Name	Wt %
1	Activated carbon	10
2	Water	20
3	Perlite	10
4	Fe_2O_3	10
5	$CaSO_4*2H_2O$	10
6	Al_2O_3	5
7	Ethylene glycol	14.5
8	Mineral Oil	12
	Total	91.5
RCRA Metals		
9	$CrCl_3*6H_2O$	0.25
10	$NiCl_2*6H_2O$	0.25
11	$PbCl_2$	0.25
12	$CdCl_2*2.5H_2O$	0.25
	Total	1
RCRA Organics		
13	Naphthalene	3
14	1,2-Dichlorobenzene	2
15	Chlorobenzene	2
	Total	7
Radionuclide Surrogates		
16	$CeCl_3$	0.25
17	CsCl	0.25
	Total	0.5

Table 2
Composition of the Aqueous Surrogate Mixed Waste

No.	Component Name	Wt %
1	Activated Carbon	10
2	Water	75
3	$CaCl_2$	3
4	$NaHCO_3$	3
5	$MgSO_4*7H_2O$	3
6	$Na_2HPO_4*7H_2O$	2.5
7	Fly ash (ASTM F)	1
	Total	97.5
RCRA Metals		
8	$Cr(NO_3)_3*9H_2O$	0.25
9	$Ni(NO_3)_2*6H_2O$	0.25
10	$Pb(NO_3)_2$	0.25
11	$Cd(NO_3)_2*4H_2O$	0.25
	Total	1
RCRA Organics		
12	Naphthalene	0.4
13	1,2-Dichlorobenzene	0.3
14	Chlorobenzene	0.3
	Total	1
Radionuclide Surrogate		
15	$CeCl_3$	0.25
16	CsCl	0.25
	Total	0.5

PI.12

Electrodialysis-Ion Exchange for the Separation of Dissolved Salts

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Introduction

The Department of Energy generates and stores a significant quantity of low level, high level, and mixed wastes. As some of the DOE facilities are decontaminated and decommissioned, additional and possibly different forms of wastes will be generated. A significant portion of these wastes are aqueous streams containing acids, bases, and salts, or are wet solids containing inorganic salts. Some of these wastes are quite dilute solutions, whereas others contain large quantities of nitrates either in the form of dissolved salts or acids. Many of the wastes are also contaminated with heavy metals, radioactive products, or organics. Some of these wastes are in storage because a satisfactory treatment and disposal processes have not been developed.

Currently, it is anticipated that these wastes will be solidified for final disposal unless a better process can be demonstrated. Some of the wastes will be combined with cement, others will be solidified by calcination or vitrification. If they are disposed of as a grout or saltcrete, the volume is increased substantially. If calcined or vitrified, some of them produce a product with undesirable integrity. There is considerable interest in developing processes that remove or destroy the nitrate wastes, which DOE is investigating. These processes range from bionitrification to calcination. DOE needs a more economical process than calcination or vitrification and a process that produces a lower volume of highly stable wastes.

Electrodialysis–Ion Exchange (EDIX) is a possible process that should be more cost effective in treating aqueous wastes. EDIX is a process developed by Mr. Phil Grant of *WASTREN* in the 1970s. The process was developed to control the boron and lithium content of the primary coolant in Pressurized Water Reactors. This technique was never commercialized. A small research and development program was conducted at EG&G Rocky Flats in 1992 to determine the suitability of EDIX for processing streams containing low levels of nitric acid and small amounts of nitrate salts (~ 1.5 g/l). The program was exploratory in nature and limited in scope for processing selective waste streams at the Rocky Flats facility for the recovery of nitric acid. It did, however, yield some very promising results that justified conducting additional research and development.

Research sponsored by the U.S. Department of Energy's Morgantown Energy Technology Center, under contract DE-AR21-95MC32112 with *WASTREN, Inc.*, 477 Shoup Avenue, Suite 209, Idaho Falls, Idaho 83402.
Telefax: 208-523-9111.

The purpose of this program, funded by METC, is to develop a process that can process rather dilute nitrate solutions. Thus, the wastes of interest are those generated at Rocky Flats and the contaminated ground water in Richland, Washington. It would be desirable to develop a process that would (1) remove the anions, primarily nitrate from the waste, free of radioactivity, in the form of an acid; (2) yield a stream of high quality water; and (3) produce a small stream containing the cations and radioactive metals. The cation stream might be processed further to separate the radioactive materials from the other cations.

This program will involve studying the suitability of the technology to treat aqueous streams containing low concentrations of organics, heavy metals, radioactive species, nitrates, and sodium. During the study, surrogate solutions typical of some of those in storage or being produced at some of the DOE sites will be evaluated.

The program will consist of a series of bench scale tests to develop the design and optimum operating conditions; two 100-hour runs to better understand the long term operation of the system; the design of a system to process 30 gpm; and the development of a cost estimate for processing 30 gpm in the EDIX system.

Objectives

The technical feasibility of EDIX has been demonstrated on a small scale. EDIX, however, has not been demonstrated as an economical process for treatment of aqueous streams containing various anions and cations. Therefore, the objective of this program is to demonstrate the suitability of the EDIX process for processing surrogate waste streams similar to those generated at some of the DOE sites. Specifically, the program is designed to determine the technical and economic potential of EDIX (1) to produce water meeting discharge criteria from a variety of waste streams containing small quantities of nitrates, chromium, sodium, uranium, and light organics; and (2) to produce waste streams containing the anions and cations in which the volume of these waste streams is less than 1/10th of the volume of the feed stream. During the program operational data will be obtained on the system through two extended runs so that a reliable economic analyses can be developed. These data will be used to design a 30 gpm system for treating one of the solutions tested in the program and to develop a capital and operating cost estimate for the 30 gpm system.

Approach

EDIX is a water/waste water treatment technology that combines ion exchange resins and membranes with the conventional electrodialysis process. The ion exchange resins can be either organic or inorganic and provide the removal mechanism for all ionic forms within a given feed stream. Electrodialysis is a voltage driven process which is used to separate and transport ions to their respective compartments for the separation and concentration of specific salts. The collective EDIX process can be used to split waste salt streams into acids and bases, or concentrate waste salts with substantial volume reduction. The use of ion exchange resins, with continuous regeneration, provide substantial decontamination factors across the cell and can provide very pure effluent water.

The basic cell unit is illustrated in Figure 1. This cell contains three compartments, each separated by a cation or anion permeable membrane. A cation membrane allows passage of cations but not anions. The opposite is true for anion membranes (A). The outer compartments contain a cathode (-) or an anode (+). The center compartment is the deionization section containing mixed-bed ion exchange resins.

The anolyte compartment contains anion resins and the catholyte compartment can be arranged in parallel or in series with similar cell units to provide either a larger flow capacity or greater degree of water purification.

Process Chemistry

The process involves two basic mechanisms: ion exchange by resin and ion migration induced by current flow. The operation of the cell is best described by assuming the feed stream contains sodium (Na^+) and nitrate ions (NO_3^-). The Na^+ ions exchange on the cation resin in the center compartment and displace hydronium (H_3O^+) ions. Under the induced current, the Na^+ ions migrate toward the cathode (-) through the porous resin particles, through the cation membrane, and into the catholyte compartment. In conjunction with the Na^+ transport, hydroxide ions (OH^-) are generated at the cathode from the electrolysis of water. The OH^- combines with the Na^+ to form a concentrated stream of sodium hydroxide (NaOH).

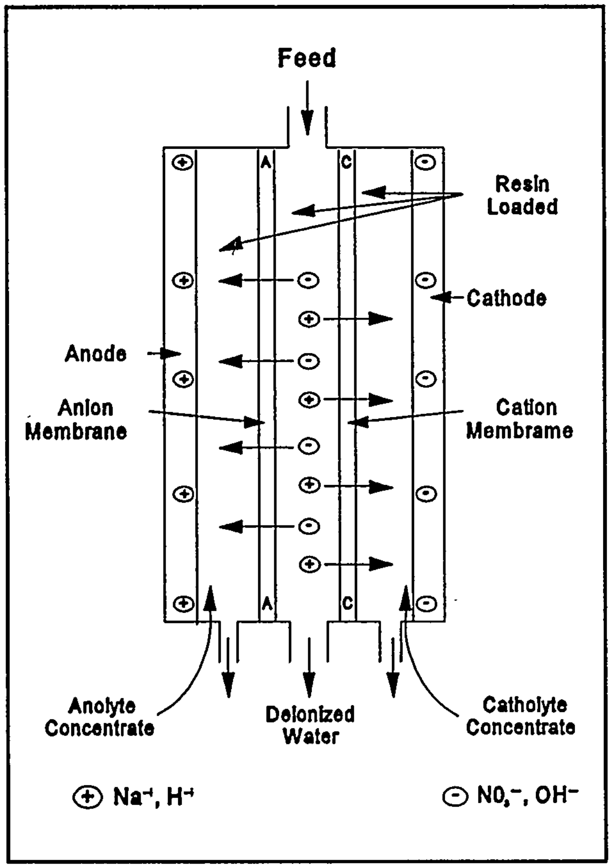

Figure 1. Electrodialysis–Ion Exchange (EDIX) Cell.

Similarly, the NO_3^- exchanges on the anion resin in the center mixed-bed compartment and then migrates with the induced current to the anolyte compartment. H_3O^+ generated from the electrolysis of water combines with the NO_3^- in the anolyte compartment to form nitric acid (HNO_3). If the solution residence time in the center compartment is sufficiently long, complete deionization occurs and all Na^+ and NO_3^- ions are removed from the feed stream.

The mixed-bed resins are constantly regenerated in place. In the feed compartment, Na^+ and NO_3^- exchange on their respective cation and anion resins. The dissociation of water molecules produces OH^- and H_3O^+. These ions displace their similarly charged Na^+ and NO_3^- from the cation and anion resins. The Na^+ and NO_3^- ions migrate to their respective anolyte and catholyte compartments as discussed above. This mechanism is a continuous electrolytical regeneration that precludes the need for regeneration by conventional acid and base techniques.

Another unique feature of this process is the ability to clean the system without disassembly. Cleaning, or purging in this instance, involves reversing the cell polarity similar to the process for conventional electrodialysis. Reversing cell polarity transports the contaminants along with Na^+ and NO_3^- into the feed compartment where they are flushed as unrecovered waste. If the goal is to produce high quality water and the composition of the anolyte and catholyte is unimportant, it may not be necessary to conduct reverse polarity operations.

Equipment Description

EDIX System

The EDIX cell and associated support system are designed to collect and store highly concentrated chemicals. The EDIX system consists of a stack of cells, an electrical rectifier, holding tanks, pumps, flowmeters, pressure gauges, and conductivity meters. The EDIX system is configured in a three-loop flow configuration as shown in Figure 2.

The alkaline and acid streams are circulated through individual tank systems to build up concentrations. The solutions are collected in the acid tank for the acid loop and the alkaline tank for the alkaline loop. These solutions are piped to their respective pumps and pumped through control valves and rotameters to the cell stack. From the cell stack the streams discharge into conductivity pots and drain into their respective holding tanks. Heat builds up in these loops from the pumps and cell voltage. The heat is removed with cooling coils submersed in the acid and alkaline tanks. The feed solution is introduced into the EDIX system via the feed tank. When the cell is energized, the stream will become demineralized through the loss of anions through the anion membrane and the loss of cations through the cation membrane.

The EDIX system is equipped with instrumentation for regulating and monitoring the operating parameters. Each tank is equipped with a conductivity pot for measuring the conductivity and pH level of the stream entering the tank. The tanks have temperature gauges to

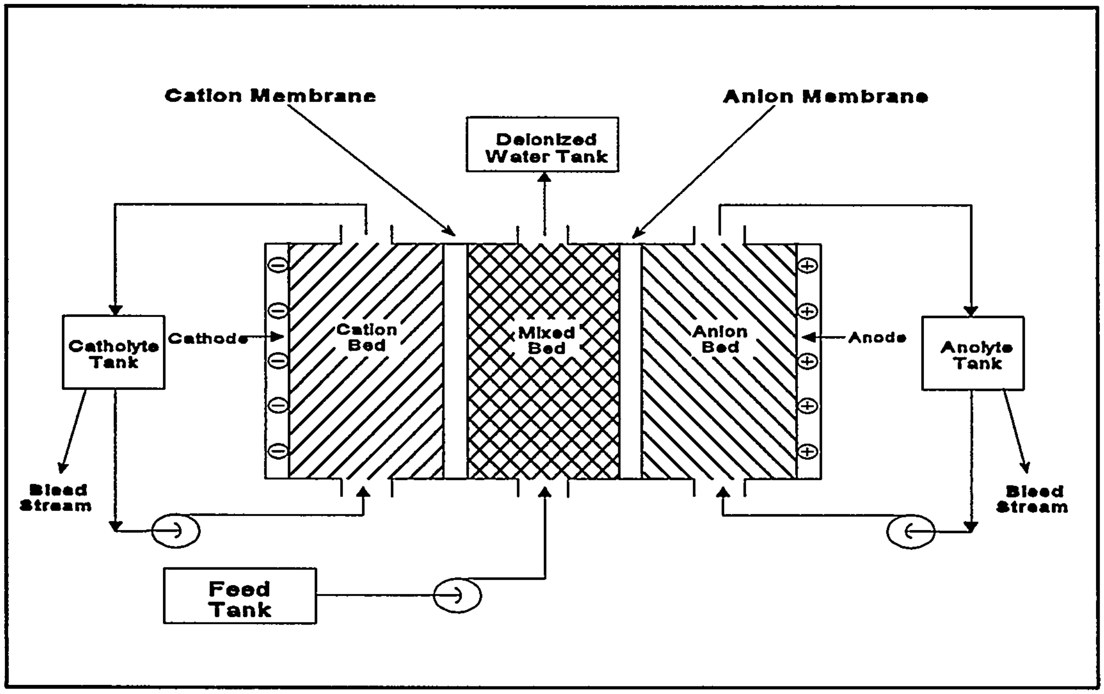

Figure 2. EDIX System—Three-Loop Flow Configuration.

monitor the system temperature. Flowmeters and control valves regulate the flow of solution to the cell stack. Pressure gauges monitor the pressures in the three compartments of the cell. The AC to DC rectifier provides power to the stack of cells. The rectifier is housed in a control box. A 220 V, single-phase, 50-60 cycle AC inlet power source is required for the rectifier. The cell current and cell voltage are recorded on an ammeter and voltmeter.

Cell Stack

The cell stack assembly consists of electrodes and one or more cell pairs. A cell pair is made up of an anode, an anolyte compartment, an anion membrane, a feed compartment, a cation membrane, a catholyte compartment, and a cathode. The cell is arranged in a stack unit, assembled, and compressed by end plates and tie-rods as shown in Figure 3.

The alkaline stream (i.e., NaOH) is concentrated in the catholyte compartment. The acid stream (i.e., HNO_3) is concentrated in the anolyte compartment. The feed stream enters the feed compartment and is deionized. The streams flow through channels; each channel is bounded on both sides by a combination of an

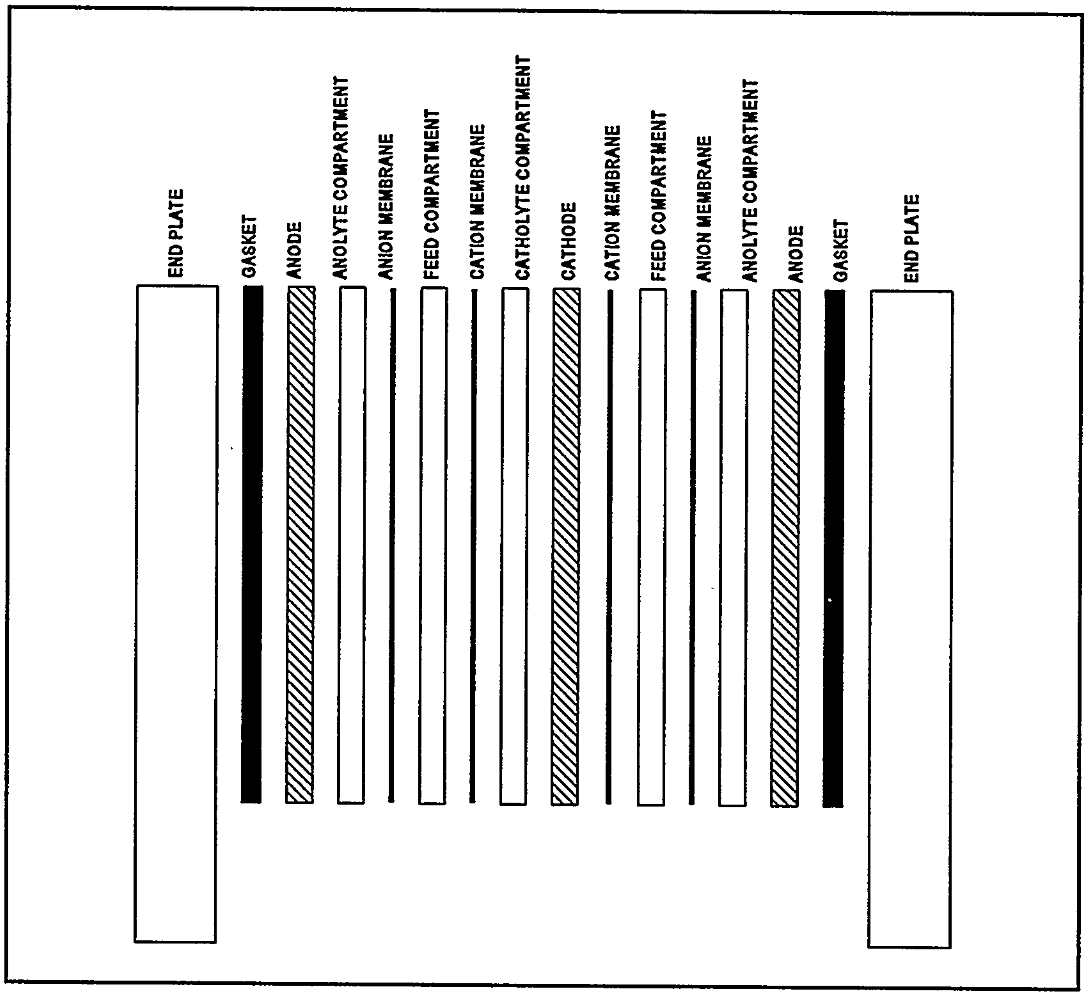

Figure 3. EDIX Cell Stack Configuration.

anion membrane, cation membrane, or electrode. The channels furnish the flow paths, keep the membranes separated, act as gaskets, and contain the ion exchange resins.

Project Description

The purpose of this experimental program is to determine whether EDIX can be used economically to process aqueous waste streams containing small quantities of radioactive materials, nitrates and other anions, heavy metals and other cations, and organics. The goal is to produce a product stream of high purity water containing the organics, an acidic stream free of radioactivity, and a cation stream containing the radioactive materials and other cations.

The process would be considered technically successful if the volume of cation and anion streams are no more than 1/10th of the volume of the feed stream. The overall success will be dependent upon the economics of the process.

The chemistry and fundamentals of processing solutions containing mixtures of dissolved salts, organics, and heavy metals are not well understood. Therefore, a matrix of bench scale tests is planned in which the composition of the feed stream, flow rate, power, and type of resins and membranes are varied. The composition of the feed solution will vary and, in most of the tests, neither radioactive materials or organics will be in the feed stream. The composition of the effluent can also vary. By reducing the throughput and increasing power, removal efficiencies can be attained that meet the desired discharge limits. The experimental program consists of six tasks as described below.

Task 1: Design of the EDIX System and Experimental Program. For the laboratory experiments, a system having a capacity of at least one liter per minute will be required. The system will be required to process a variety of waste streams. However, the concentration of the contaminants (organics, heavy metals, nitrate, and radioactive materials) will be relatively small and in general < 3,000 ppm for any one constituent. The basic hardware will be designed and selected for these conditions. *WASTREN* will review the literature and hold discussions with the resin and membrane suppliers and select the membranes and resins that will have the highest probability of performing as required.

Task 2: Procure, Assemble, and Check Out the System. After the design of the system has been completed, *WASTREN* will purchase the equipment, membranes, resins, and chemicals needed to prepare surrogate solutions. After the equipment is received, it will be assembled into a complete EDIX system. The system will be checked out using deionized water and very dilute salt solutions to determine if there are any leaks; the throughput of the system; and the ability to control flow, pressure drop, voltage, and other operational parameters. Once the system passes these tests it will be ready for testing with the surrogate solutions.

Task 3: Conduct Bench Scale Tests. The experimental program will be divided into three phases. Phase 1 will be to run a series of about ten tests to determine the most suitable resins and membranes, voltages, and flow rates to transfer the anions and cations.

Phase 2 will be to run a series of forty tests in which the composition of the feed stream is varied, with the chief variables being levels of nitrate, sodium, and chromium metals. A proposed test matrix is shown in Table 1.

These experiments are designed to:

a. Determine if the anions and cations are removed selectively or in bulk;

b. Establish the conditions necessary to achieve the desired water quality;

c. Determine what concentration factors can be achieved for both the cation and anion streams;

d. Determine if precipitates are formed in the cation cell because of the high pH and determine what effect, if any, these cations have on overall system performance; and

e. Determine the operating conditions that are needed to preclude the deleterious effects of precipitates in cation cells.

Table 1. Suggested Experimental Program Test Matrix.

Nitrate Level ppm	Chromium Conc, ppm	Sodium Concentration, ppm		
		18	200	2000
30	20	X	X	X
30	200	X	X	X
30	2000	X	X	X
300	20	X	X	X
300	200	X	X	X
300	2000	X	X	X
3000	20	X	X	X
3000	200	X	X	X
3000	2000	X	X	X

Phase 3 will be a series of twelve tests in which some of the solutions tested in Phase 2 will be spiked with various levels of organics and depleted uranium to see if organics adversely affect the process and if the uranium concentration in the discharge streams (product and anion streams) are low enough to label them as non-radioactive.

Task 4: 100-Hour Extended Runs. After the bench scale tests have been completed, two 100-hour continuous campaigns on two different surrogate solutions will be conducted. These campaigns will determine if the performance of the cell changes with time and how frequently it will be necessary to regenerate the resins.

Task 5: Recommend processes for Treating the Catholyte and Anolyte Streams. The results of the 100-hour runs should provide information on the composition of the anolyte and catholyte streams. *WASTREN* will suggest means of processing these streams, but will not conduct any experiments.

Task 6: Design of a 30 gpm system. Based on the results from the 100-hour extended runs, an EDIX system to process 30 gpm of feed solution will be designed. The design will not include any processing schemes for the anolyte and catholyte solutions generated in the EDIX system. The system designs will be used to develop capital and operating costs. The cost of processing 1000 gallons of the desired water quality will be calculated.

WASTREN was authorized to begin work on the project in March 1995. The tentative schedule shown in Figure 4 will require about 20 months to complete. If the entire program is conducted and the schedule met, the program will be completed in November 1996.

The project manager is Dr. Barcoh of *WASTREN*, and the test program will be conducted at the Colorado Minerals Research Institute in Golden, Colorado. Mr. John Litz will be the project engineer for Acta Resources.

Results

The project began in March with the design of the system. Several vendors of electrodialysis cells were contacted and *WASTREN* evaluated the system capabilities and cost. The Electro Syn Cell provided by ElectroSynthesis was selected for the test program. As an electrodialysis cell it has a capacity of three liters per minute when processing a solution containing 200 ppm of sodium, 200 ppm chromium as chromate ($CrO_4^=$), and 300 ppm nitrate. The throughput when the cell contains ion exchange resins and higher concentrations of contaminants will be determined in the experimental program.

Based on discussions with the suppliers of the membrane and ion exchange, the following membranes and ion exchange resins were selected.

Item	Primary	Secondary
Anion Membrane	Tokuyama Sodium Neosepta AMX	Ultrex AMI-7000
Cation Membrane	Tokuyama Sodium Neosepta CMX	Ultrex CMI-7000
Anion Resin	Dowex 21	Reilex HPG
Cation Resin	Dowex G26	Dowex M33

The experimental equipment has been erected and the operational testing is in progress. No data have been generated at the time of publication of this paper.

Applications

The primary focus is on anticipated process performance, cost, and expected advantages and improvements over existing

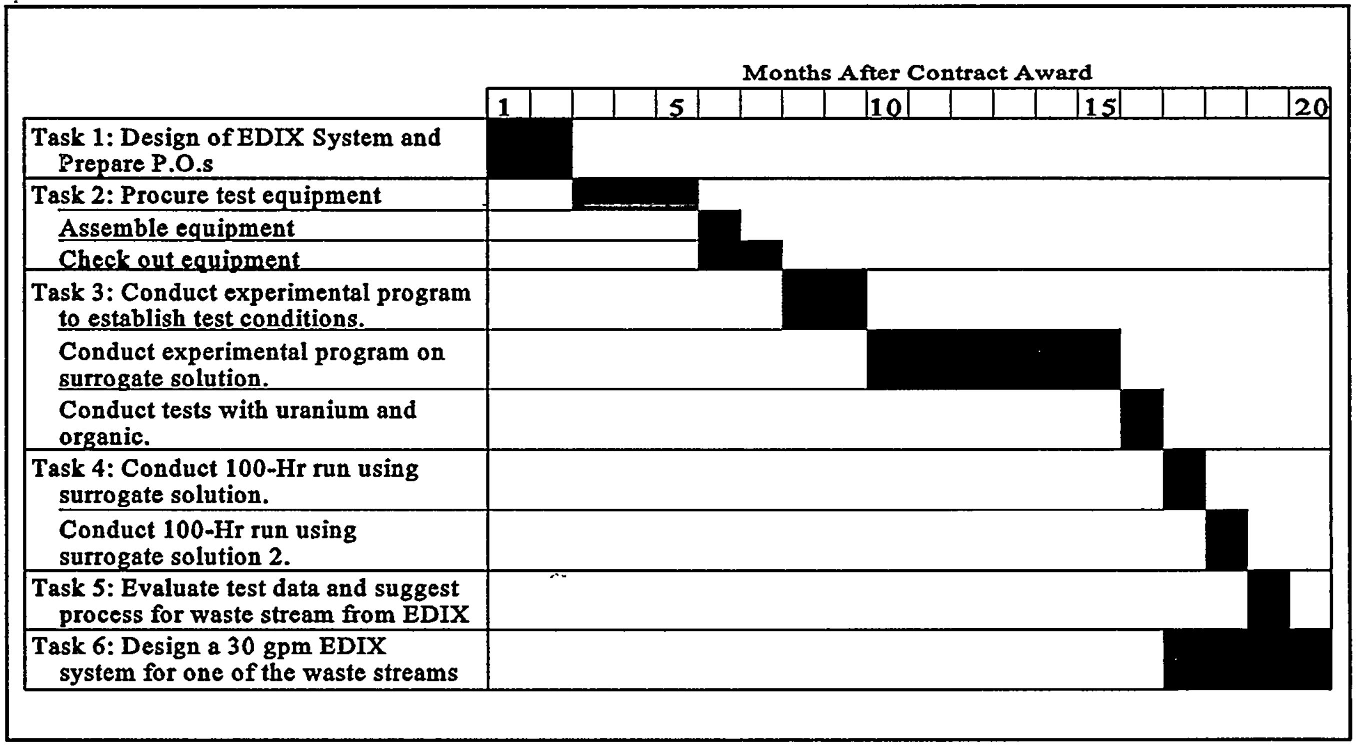

Figure 4. Schedule for EDIX Project.

technologies. EDIX is an emerging technology with only limited development for nitric acid recovery and water purification. Therefore, conclusions on performance and benefits are premature until the test program has been completed.

A major aspect in predicting EDIX operational performance is that each of the conventional ion exchange (IX) and electrodialysis (ED) technologies used in EDIX has extensive performance data and cost information which lends itself to evaluating the combined merits of the proposed technology. Additionally, the preliminary testing of the EDIX technology at Rocky Flats, while limited, also provides a source of empirical data on performance expectations for technology.

The EDIX technology applies two proven and commonly used water treatment technologies, ion exchange and electrodialysis, in a single, self-regeneration process for waste water treatment. The conventional ion exchange process is a relatively low cost ($0.50 - $1.50 per 1000 gallons) technology which can be used for ion substitution and high purity water recovery, but requires resin regeneration and/or resin discard, and has limited ionic capacity. The use of conventional electrodialysis is also low cost ($0.35 - $0.50 per 1000 gallons) and can be used to split salt streams, reduce waste volumes, and recover selective ions as acids and bases. The conventional ED process does not produce a higher effluent quality when compared to IX. The EDIX process benefits from these two conventional processes in that the IX resins are regenerated in place, high purity water can be recovered in a single step, additional flow capacity can be added without major retrofit, and a wider range of chemical/contamination concentrations can be treated than either IX or ED as a stand-alone process.

Another unique part of the EDIX technology is that by combining IX and ED into a single process, the effective range of feed flow rates and stream composition can be increased to include both low concentrations (ppm), high flow input as well as high concentrations, low volume feed streams. The use of IX resins within an EDIX cell provides greater ion retention and more efficiency for ion removal than conventional ED and therefore can produce exceptionally high quality water at a lower cost. The EDIX process should provide cost effective resin and membrane regeneration as part of the process and provides substantial waste volume reduction when compared to conventional technologies like reverse osmosis and membrane filtration systems. The following represents *WASTREN's* judgment on the potential of EDIX:

- Removal of > 99.9% of the ionic sources from the feed stream
- Waste volume reduction factors of up to 200 depending on the composition of the feed stream and the solubility limits of the constituents
- Continuous in-place regeneration of ion exchange resins and selective membrane materials
- Effluent water of high quality to meet all expected recovery and recycle requirements except for dissolved organics where present
- Design flexibility with capacity to treat small to high volume throughput without major design and operational changes
- Concentrated anion and cation waste streams (up to solubility limit of chemical

forms) as a function of input feed concentrations

- Low energy cost (~\$0.60/1000 gal.) and low pressure (< 50 psig) operations
- Longevity of equipment and materials expected to be > ten years based on equipment vendor testing data and proposed life cycle studies
- Potential for small amount of secondary waste generation which could be treated by conventional technologies or possibly could be recycled or returned to feed stream

Cost Comparisons with Existing Technologies

The anticipated technologies with which EDIX would compete include evaporation, reverse osmosis, conventional electrodialysis, ion exchange, and possibly utrafiltration and/or other membrane separation technologies. None provide the single step process proposed in the EDIX technology to produce three separate streams which include clean water (possibly with dissolved organics), a concentrated acid stream, and concentrated base stream containing the radioactive metals.

The anticipated costs for equipment and unit operations are based on *WASTREN's* engineering judgment for the EDIX technology because of the lack of specific performance and operations cost data. However, some assumptions can be made relative to the size of a large scale system and the associated operations. Table 2 shows the relative treatment and capital costs for a variety of techniques processing 72,000 gallons per day.

The major cost savings incentives are associated with volume reduction of the wastes, production of high purity water, and the potential to recycle chemicals for reuse. *WASTREN* believes that the EDIX technology can provide volume reductions up to the solubility limits of the feed salts and bases in the catholyte solution. For highly concentrated feed solutions, the volume reduction can be relatively low. Secondary cost incentives can be associated with water recycle and water management, especially for sites or facilities attempting to reduce plant effluents to near zero. Other potential cost savings incentives could be associated with chemical recovery, lower

Table 2. Technology Treatment with Capital and Operating Costs for a 5 gpm System

Technology	System Costs	Operating Costs
Evaporator	\$2.0M - \$2.5M	\$4.50 - \$6.50 per 1000 gallons
UF/Reverse Osmosis	\$280K - \$350K	\$0.80 - \$0.95 per 1000 gallons
Electrodialysis	\$120K- \$150K	\$0.35 - \$0.50 per 1000 gallons
Ion Exchange with Regeneration System	\$350K	\$0.50 - \$1.50 per 1000 gallons
UF/Nano-Filtration	\$450K	\$1.00 - \$1.50 per 1000 gallons
EDIX (Potential)	\$150K - \$175K	\$0.50 - \$0.60 per 1000 gallons

equipment and operating costs, flexibility in adding system capacity without major system reconfiguration, and operator ease in establishing waste stream process control programs.

Future Activities

The experimental program to determine the optimum conditions for treating the various solutions will begin in October, Task 3 as shown in Figure 4. These tests will require about nine months to complete if the entire test program is completed. Two 100-hour continuous runs will be run during the summer of 1996. During the fall of 1996, the 30 gpm system will be designed and the economics of the process will be calculated.

Acknowledgment

We appreciate the assistance and guidance William Huber, METC Contracting Officer's Representative, has provided to *WASTREN*. This contract began in March 1995 and should be completed in November 1996. *WASTREN* has a contract with Acta Resources of Golden, Colorado, to be responsible for the laboratory work. Mr. John Litz is project manager for Acta Resources. This work is being conducted as part of the Low-Level Mixed Waste Processing, Item 2, Separation of Suspended and Dissolved Solids program of DOE.

PI.13 Decontamination Systems Information and Research Program

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Introduction

The Decontamination Systems Information and Research Program at West Virginia University is a Cooperative Agreement which focuses on research and development associated with hazardous waste remediation problems existing at Department of Energy, Corps of Engineers, and private sector sites. The agreement builds on a unique combination of resources coupling university researchers with small industrial businesses leading towards field tests and large scale demonstration of existing DOE environmental programs.

Objectives

Develop a leading environmental technology research program encompassing the following:

- Evaluation and development of protocols enabling decontamination specialists to systematically evaluate a site and weigh remediation options.
- Identify and initiate development and demonstration of workable systems to fill the gaps in current technologies.
- Train and prepare students to enter into industrial and agency positions in waste and site remediation professions.
- Prepare documentation reporting environmental information allowing DOE to prepare the appropriate level of NEPA documentation for the program.

Approach

The Program is divided into the following three focus areas supporting DOE's needs: 1) In Situ Remediation Process Development, 2) Advanced Product Applications Testing, and 3) Information Systems, Public Policy, Community Outreach, and Economics. These areas fall into the Program structure shown schematically in Figure #1.

The Small Business Support Program works with the Small Business Incubator providing resources for small start-up businesses in the environmental and site remediation fields. Business support includes providing researchers from WVU, and other sources to solve problems or remove barriers to commercialization of remediation processes funded by DOE.

Research Sponsored by the U.S. Department of Energy's Morgantown Energy Technology Center, Under Contract DE-FC21-92MC29467 with the Department of Civil and Environmental Engineering, West Virginia University, Morgantown, WV 26506, (304) 293-3031

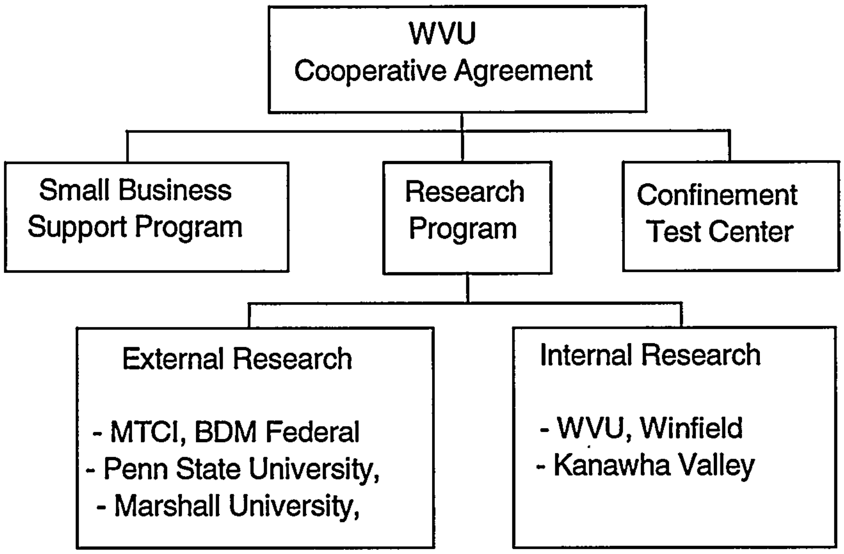

Figure #1 - Decontamination Systems Information and Research Program

The Research Program includes a diversity of projects bringing together a cadre of investigators from both WVU on-campus colleges as well as from regional universities to study a diversity of topics ranging from bioremediation to innovative technology economics.

The Confinement Test Center deals with using existing METC facilities for the testing and demonstration of various barrier and other focus area research projects.

Technology Descriptions

Key research projects for FY 95 include:
1.0 IN SITU REMEDIATION PROCESS DEVELOPMENT

* TASK 1.1 USE OF DRAIN ENHANCED SOIL FLUSHING (DESF) FOR ORGANIC CONTAMINANTS REMOVAL: Enhanced in situ soil flushing using prefabricated vertical drains (PVD) for remediation of contaminated sites where fine-grain soils are present. The research builds on the soil flushing process where remediation of contaminated aquifers using conventional pump-and-treat systems has shown to be ineffective and cost prohibitive. (PI: M. Gabr)

* TASK 1.2 IN SITU BIOREMEDIATION OF CHLORINATED ORGANIC SOLVENTS: This research is evaluating and optimizing the ability of methanotrophic, methanogenic, and other selected bacteria for cost-effective biotransformation of TCE and other volatile organic compounds found at DOE sites. The approach involves in situ bioremediation for destruction of organics in groundwater using microorganisms to carry out the anaerobic reductive dehalogenation of highly chlorinated solvents followed by aerobic degradation of the residual compounds to environmentally acceptable end products. (PI: W. Sack)

* TASK 1.3 MICROBIAL ENRICHMENT FOR ENHANCING IN SITU BIODEGRADATION OF HAZARDOUS ORGANIC WASTES IN SOIL: This research investigates the effectiveness of commercially available microbial inocula for use in bioaugmented remediation of hazardous mixed organic wastes in soil and examining ex-situ remediation and surface composting which is suited to incorporation of a microbial inoculant. Project goals are to obtain soil samples from waste contaminated sites and under laboratory conditions quantify the effectiveness of selected inocula for bioaugmented soil remediation. (PI: A. Sexstone)

* TASK 1.4 CONTAMINANT MOVEMENT PREDICTION IN PERMEABLE SUBSURFACE SOILS: This research project investigates numerical modeling of subsurface flow mechanisms from laboratory data associated with TASK 1.1. Project goals are to develop a numerical model to predict the field-scale groundwater movement using prefabricated vertical drains (PVD) for enhanced soil flushing. (PI: Bilgesu)

* TASK 1.5 DEVELOPMENT OF A STANDARD PROTOCOL AND BARRIER DESIGN MODEL FOR IN SITU FORMED BARRIERS: This project researches chemical grout barriers and circulating air barriers for achieving in situ containment of subsurface contaminants at DOE's Hanford and Savannah River sites. (PI: C. Black)

2.0 ADVANCED PRODUCT APPLICATIONS TESTING

* TASK 2.1 CHEMICAL DESTRUCTION OF CHLORINATED ORGANIC COMPOUNDS: The objective of this research project is to develop chemical treatment procedures to transform chlorinated organic compounds to less or nontoxic materials. Project success has be achieved with the development of a low molecular weight oligomer containing sterically hindered triarylboryl functionalities as an electron carrier for dechlorination. (PI: K. Wang)

* TASK 2.2 CONTINUED DEVELOPMENT OF AN ATMOSPHERIC MONITORING MASS SPECTROMETRY SYSTEM: The objective of this project is the development of a direct atmospheric monitor using tandem mass spectrometry for field determination of volatile organic compounds on real time basis. Field work was begun using the mass spectrometry systems for identifying BTEX at the US Army Corps of Engineer's Winfield site in West Virginia. (PI: F. King)

* TASK 2.3 CAPTURE OF HAZARDOUS WASTE MATERIALS UTILIZING FLUIDIZATION COATING TECHNOLOGY: This research investigates the use of fluidized bed coating technology in the collection, concentration, and stabilization of hazardous inorganic chemical compounds from liquid solution and liquid-solid slurries. (PI: R. Turton)

* TASK 2.4 REMEDIATION OF HAZARDOUS SITES WITH STEAM REFORMING: This project is focused at developing a practical ex-situ method for thermal organic chemical destruction of contaminated soils containing chlorinated wastes without development of dioxin byproducts. The technology uses steam reforming with pulsed combustor technology. The equipment operates under pressurized conditions with the contaminated soil as the fluidized bed. (PI: V. Mansour)

* TASK 2.5 ENVIRONMENTAL POLLUTION CONTROL DEVICES BASED ON NOVEL FORMS OF CARBON: The purpose of this research is to assess the feasibility of using carbon nanofiber devices to remove heavy metal contaminants from aqueous streams. This work addresses the fabrication, behavior, and analytical utility of these novel carbonaceous materials.
(PI: C. Irwin, J. Zondlo)

<u>3.0 INFORMATION SYSTEMS, PUBLIC POLICY, COMMUNITY OUTREACH, AND ECONOMICS</u>

* TASK 3.1 WINFIELD LOCK AND DAM ASSISTANCE PROGRAM: This project supports the United States Army Corps of Engineers remediation of a contaminated industrial site on the Kanawha River at Winfield, West Virginia. West Virginia University's role in the project is to provide community outreach to the citizens of Eleanor, WV, providing technical, logistical and financial assistance in their efforts to understand and evaluate the Corps of Engineers' remediation processes and techniques. (PI: R. Lovett)

* TASK 3.2 A GIS - BASED INFRASTRUCTURE FOR SITE CHARACTERIZATION AND REMEDIATION: The primary goal of this work is to determine an effective Geographic Information System (GIS) based infrastructure for describing, characterizing, and remediating contaminated sites. Objectives involved in this goal include: a) devising and specifying an integrated software and hardware system, b) establishing operational procedures for effective application of the system, and c) establishing methods for effective transfer of technology. (PI: J. Hooper)

* TASK 3.3 SMALL BUSINESS SUPPORT PROGRAM: This project was initiated to address problems with commercialization of innovative site remediation technologies developed by small businesses funded by DOE. Once a problem or opportunity was defined, researchers from WVU, Marshall or other sources would be solicited to work with the small business in a research project to solve the problem or remove the barrier to commercialization of the process. (PI: E. Cook)

* TASK 3.4 APPROACH FOR ASSESSING POTENTIAL VOLUNTARY ENVIRONMENTAL PROTECTION, KANAWHA VALLEY AREA: The objective of this project is to assess the interest and willingness of industry in the Kanawha River Valley, WV to participate in discussions leading toward the voluntary cleanup of contaminated industrial sites using DOE developed technologies. (PI: R. Lovett)

Future Activities

Future activities will involve field demonstration of current projects and a concentration into the following DOE/METC focus areas:

* Contaminant Plume Containment and Remediation.
* Landfill Stabilization.
* Facility Transitioning, Decommissioning & Final Disposition.
* Mixed Waste Characterization.
* Treatment and Disposal.
* High Level Radiation Waste Tank Remediation.
* Crosscutting Innovative Technologies including Characterization, Monitor & Sensor Technology and other Innovative Investment Areas.

Contract Information

Cooperative Agreement Number:
DE-FC21-92MC29467

Contractor:
West Virginia University
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Period of Performance:
October 1, 1992 to September 30, 1997

PI.14 Environmental Management Technology Demonstration and Commercialization

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Introduction

The Energy & Environmental Research Center (EERC), a contract-supported organization focused on technology research, development, demonstration, and commercialization (RDD&C), is entering its second year of a Cooperative Agreement with the U.S. Department of Energy (DOE) Morgantown Energy Technology Center (METC) to facilitate the development, demonstration, and commercialization of innovative environmental management (EM) technologies in support of the activities of DOE's Office of Environmental Science and Technology (EM-50) under DOE's EM Program. This paper reviews the concept and approach of the program under the METC–EERC EM Cooperative Agreement and profiles the role the program is playing in the commercialization of five EM technologies.

Program Concept and Approach

Key components of commercialization activities in the EM program include knowledge of the marketplace, technical expertise, and the ability to forge partnerships with industry and government. In some cases, particularly for the small business technologist, commercialization prospects are hampered by limited technical expertise, testing and demonstration capabilities, capital and, specific to the EM program, knowledge of DOE and EM site needs. Alternatively, commercialization may hinge on the successful resolution of technical barriers. The program under the METC–EERC EM Cooperative Agreement is designed to facilitate the delivery of promising technologies to the marketplace through a dynamic process involving focused technical assistance, partnership brokering, and real-world demonstrations. With respect to resolving technical barriers, the EERC brings a wide range of technical expertise and state-of-the-art facilities for investigating chemical and physical processes. EERC facilities are capable of tests and demonstrations from laboratory to pilot scale and are suitable to conduct tests representing a wide range of physical conditions.

With respect to resolving investment and application barriers, the EERC acts as a broker

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between government and private sector interests in technology development and commercialization. Based on its long experience with government entities, including DOE, the Department of Defense, and the Environmental Protection Agency, the EERC is able to provide guidance in promoting contacts and fostering partnerships with appropriate government agencies and personnel. Alternatively, the EERC's experience in the private sector provides the basis for contacts and partnerships in that venue.

Because of legal and other regulatory constraints, gaining access to field test sites has become a formidable obstacle to the demonstration of promising EM technologies. The EERC's growing family of industrial partners provides the potential for access to a wide variety of technology demonstration sites. For example, the EERC has access to a remediation demonstration site in Alberta, Canada, through a relationship with the Canadian Association of Petroleum Producers, Gulf Canada, and the DOE Jointly Sponsored Research Program.

As a contract RDD&C facility, the EERC has a number of advanced analytical, monitoring, treatment, and processing methods in development, often in conjunction with private sector partners. The METC–EERC EM Cooperative Agreement provides an avenue to evaluate these technologies for EM applications and to work to bring deserving technologies into the EM marketplace.

Technology Commercialization Activities

FY1996 activities are focused on advancing five technologies to the commercialization stage. These technologies cover the gamut of EM Focus Areas and include private sector technologies, technologies already in place at DOE sites, and those under development in-house at the EERC. Most of the technologies are being developed with a commercial partner. FY1996 activities are focused on technical enhancements, the resolution of technical barriers, and field demonstration of prototypes.

Extraction and Analysis of Pollutant Organics from Contaminated Solids Using Off-Line Supercritical Fluid Extraction (SFE) and On-Line SFE/Infrared (IR) Spectroscopy

One of the remediation industry's greatest challenges is obtaining cost-effective, accurate, and precise analyses for organic contaminants. An inexpensive (<$20,000), simple-to-operate field instrument, consisting of SFE coupled with IR, intended for the extraction and analysis of organics at ppm to ppb detection levels is being developed in-house at the EERC. The method, based on SFE with CO_2, is highly accurate, rapid (<30 minutes), and nonpolluting. The unit has applications in several focus areas for characterization and sensing and is expected to be particularly useful as a screening tool.

Field trials using SFE for PAH (polycyclic aromatic hydrocarbon)- and PCB (polychlorinated biphenyl)-contaminated soils showed 1) complete extraction of PAHs and PCBs from field samples in 30 minutes (versus 14 to 18 hours with conventional extraction procedures), 2) SFE instruments performed well on a field generator unit, 3) total solvent use for field SFE was ca. 5 mL per sample, and 4) soil extractions were immediately ready for analysis without further cleanup or concentration.

On-line SFE has been coupled with Fourier transform IR (FT-IR) detection based on a fiber optic interface in a prototype unit. Results of field tests indicate 1) SFE/FT-IR gives good quantitative results for petroleum-contaminated soil in 20 minutes with no organic solvent (compared to a 4-hour lab extraction with 150 mL of Freon-113),
2) detection limits were in the ppm to ppb range for organics in soils, and 3) the interface is easily adaptable to most FT-IR and IR instruments.

An agreement has been reached with Suprex Corporation of Pittsburgh, a major SFE instrument company, to commercialize the fiber optic interface. Activities in the future will focus on the optimization of the interface to meet the conditions of sustained field use and field testing.

Thermal Decomposition of High-Organic-Content Radioactive Wastes

The diverse nature of mixed waste plastics has thwarted application of conventional methods for their reprocessing to yield recyclable chemical feedstocks or safely disposable materials. Because of their unique chemistry, polyethylene terephthalate (PET) and polyvinyl chloride (PVC) pose particular reprocessing challenges. A low-temperature process for thermal decomposition of mixed waste plastics, automotive shredder wastes, and chemical spill residuals has been developed by the EERC in partnership with the American Plastics Council, DOE, and other U.S. companies. This process, which utilizes fluidized-bed technology, will form the basis of a commercially viable recycling process able to accommodate a wide variety of plastics and other high organic-content materials from laboratory and production facilities that handle low-level nuclear materials.

Activities have consisted of pilot-scale testing using a variety of feedstocks and bed materials at temperatures between 480° and 600°C in the EERC 2-kg/hr continuous fluidized-bed reactor (CFBR). Feedstocks used in the tests consisted of postconsumer plastic wastes mixed with radionuclide surrogates selected to represent the following radioactive species: thorium, uranium, plutonium, neptunium, americium, cesium, strontium, ruthenium, rhenium, and iodine.

Results from the pilot-scale tests indicate that the EERC thermal decomposition process can 1) be successfully applied to a wide variety of plastics, including PET and PVC, 2) yield a condensate product with a chlorine content of less than 100 parts per million (ppm) from a feedstock mixture containing up to 10% PVC, 3) concentrate radionuclide surrogates in a dry, particulate solids product, and 4) yield a condensate product with radionuclide surrogate concentrations of less than 5 ppm (the analytical detection limit used in these tests) from a feedstock mixture containing surrogate concentrations of about 3000 ppm.

The EERC has entered into discussions with Los Alamos National Laboratory (LANL) for the performance of bench-scale process evaluation tests with actual radioactive wastes. Under the currently proposed program, LANL will validate EERC surrogate test findings in tests with mixed waste plastics spiked with known quantities of radionuclides and then perform process demonstration tests with actual radioactive waste. Following the LANL tests, pilot-scale surrogate tests at feed rates of up to 100 lb/hr will be performed at the EERC to provide scaleup data. For these tests, analytical detection limits will be lowered to below 100 parts per billion to provide a more definitive indication of process performance.

Fluid-Bed Calcination for Liquid Wastes

The EERC's extensive background and facilities for evaluating fuel properties in FBCs are being applied to bed-sintering problems experienced in the fluid-bed processing of EM liquid wastes at Idaho National Engineering Laboratory (INEL). INEL currently has large volumes (6900 m^3) of liquid wastes, mainly radioactive wastes from tanks, that cannot be processed in INEL's existing calciner because of bed sintering caused by sodium nitrates. The EERC will perform tests using its existing facilities to evaluate denitrification strategies for the Lockheed Martin Idaho Technologies Company (LMITCO) New Waste Calciner Facility. Optimizing the existing facilities at INEL will result in a substantial cost savings in waste processing. This technology would then find application at other sites under the EM tank waste focus area.

The EERC's effort is focused on constituent capture and immobilization and the development of data to determine optimum FBC capabilities to process wastes at INEL other EM sites. Specifically, the EERC will use surrogates to evaluate denitrification using sugar as a reducing agent and silica as a stabilizing agent to produce groutable or vitrifiable materials for disposal. Sodium- bearing liquid waste will be tested alone and in combination with material representing INEL's previously calcined waste to enhance its stability.

Based on the success in tests using surrogates at the EERC, INEL will demonstrate the process in its 12-inch calciner and grouting and vitrification facilities as appropriate.

Centrifugal Membrane Filtration

Large volumes of groundwater contaminated with heavy metals and radionuclides exist at DOE and Department of Defense sites. Conventional actions include the construction of in situ barriers or the use of ex situ treatment technologies such as chemical reduction followed by precipitation and ion exchange. In situ barriers contain the contamination but do not alter its character, and ex situ chemical and ion exchange require the use of hazardous chemicals and may result in secondary waste streams. SpinTek Systems, LP, has developed a uniquely configured centrifugal membrane filtration process to produce a clean, filtered water stream and a low-volume concentrate stream. The process can be applied to contaminated groundwater or to aqueous process or waste streams. Based on previous demonstrations of the SpinTek process, the unit has excellent potential for EM site applications, particularly under the contaminant plume focus area.

The centrifugal membrane filtration unit uses ultrafiltration techniques at pressures of up to 100 psig, in combination with centrifugal force, to remove suspended and dissolved solids to as low as 200 molecular weight. The main components of the unit include membrane filtration disks mounted on a hollow shaft and housed in a stationary pressure vessel. Feed water is introduced into the pressure vessel and flows across the membrane disks that are rotated at high speed.

In order to optimize the application of the technology to EM cleanup needs, the EERC is undertaking six tasks in support of the SpinTek commercialization effort: 1) a literature review; 2) a preliminary verification of process capability; 3) membrane optimization; 4) a process performance evaluation on EM surrogate waste; 5) extended testing on membrane cleaning cycle, scaling, fouling, and corrosion; and 6) preparation of a final assessment report. Testing and evaluation of a small pilot filtration unit is under way.

Laser Cleaning of Contaminated Painted Surfaces

Decontamination through the efficient removal of radionuclide-contaminated paint would greatly facilitate the economical decommissioning of structures and equipment at DOE weapons complex facilities. Conventional technologies, including sandblasting, create a secondary waste stream and erode substrates. Innovative surface abrasion technologies, including the use of CO_2 ice pellets, solve the issue of secondary waste generation but do not reduce the volume of primary waste.

A surface-cleaning technology developed by F2 Associates uses a CO_2 pulse laser with a 2.5-cm^2 area, in combination with robotics, resulting in a remote system that 1) removes contaminated paint from surfaces, 2) does not release hazardous constituents into the atmosphere, 3) reduces primary waste volume, 4) produces no secondary waste stream, and 5) does not erode substrates. The technology is scheduled for demonstration by year-end 1996.

EERC activities, focused on optimizing the technology and developing application parameters, include 1) incorporation of on-line analytical technologies, 2) design for enhanced decontamination, and 3) cost analysis. The incorporation of the on-line analytical capabilities will require researching small real-time radiation detectors and miniature spectrometers that will be used to characterize the emissions from the paint removal on-line instead of sampling after collection. The spectrometer would analyze for lead, chromium, and certain other heavy metals. To aid in defining the applications of these technologies, theoretical calculations will be performed, and existing experimental data related to the volatility and particle-size distribution of hazardous materials from related processes will be reviewed. Finally, a cost analysis algorithm, for use in developing economic profiles to allow a comparison of technology and process options, will be developed to factor in all of the aspects of paint removal under EM conditions, including surface characterization (before and after), cleaning costs, the cost of characterization of waste (if not done on-line), waste disposal costs, and salvage credits.

Summary

The METC–EERC EM Cooperative Agreement is a unique program involving a dynamic approach to bringing technologies into the marketplace. This "hands-on" approach features focused technical support, partnership brokering, and field demonstrations. The EERC provides a platform for the success of this program through its technical expertise, facilities, government and commercial partnerships, and access to field demonstration sites. The EERC, therefore, serves as an effective bridge between government and industry by becoming an active partner in the commercialization process. The search for additional candidate technologies and commercial partners is ongoing.

Acknowledgments

We wish to acknowledge Venkat Venkataraman, our METC Contracting Officer's Representative, and Madhav Ghate, Director of METC's Technology Base Projects Management Division.

PI.15 VAC*TRAX — Thermal Desorption for Mixed Wastes

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Abstract

The patented VAC*TRAX process was designed in response to the need to remove organic constituents from mixed waste, waste that contains both a hazardous (RCRA or TSCA regulated) component and a radioactive component. Separation of the mixed waste into its hazardous and radioactive components allows for ultimate disposal of the material at existing, permitted facilities.

The VAC*TRAX technology consists of a jacketed vacuum dryer followed by a condensing train. Solids are placed in the dryer and indirectly heated to temperatures as high as 260°C, while a strong vacuum (down to 50 mm Hg absolute pressure) is applied to the system and the dryer is purged with a nitrogen carrier gas. The organic contaminants in the solids are thermally desorbed, swept up in the carrier gas and into the condensing train where they are cooled and recovered. The dryer is fitted with a filtration system that keeps the radioactive constituents from migrating to the condensate. As such, the waste is separated into hazardous liquid and radioactive solid components, allowing for disposal of these streams at a permitted incinerator or a radioactive materials landfill, respectively. The VAC*TRAX system is designed to be highly mobile, while minimizing the operational costs with a simple, robust process. These factors allow for treatment of small waste streams at a reasonable cost.

The VAC*TRAX pilot system has proven effective at treating soil, sludge, and assorted debris streams, removing volatile and semi-volatile organic compounds, including polychlorinated biphenyls (PCBs), from the solid matrix. Removal efficiencies typically exceed 99%, and the system has repeatedly proven the ability to remove difficult components, such as PCBs, completely; PCBs were not detected in the product for a number of test runs on PCB contaminated soil and debris. Batch testing has indicated that the system may prove equally effective in treating streams contaminated with elemental mercury. In all cases involving mixed waste, the VAC*TRAX system demonstrated superior containment of the radioactive material within the solid matrix, with negligible partitioning of the material into the condensate.

This paper describes the VAC*TRAX thermal desorption process, as well as results from the pilot testing program. Also, the design and application of the full-scale treatment system is presented. Materials tested to date include spiked soil and debris, power plant trash and sludge contaminated with solvents, PCB contaminated soil, solvent-contaminated uranium mill-tailings, and solvent and PCB-contaminated sludge and trash. Over 70 test runs have been performed using the pilot VAC*TRAX system,

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with more than 80% of the tests using mixed waste as the feed material.

Introduction

The VAC*TRAX system was designed to meet the treatment needs of sites containing small volumes (10-4000 cubic meters) of hazardous and/or mixed waste materials, with the hazardous constituent consisting of organic or volatile contaminants. The process was designed for use on a wide variety of solid matrices, including soils, sludges, and contaminated debris from construction or clean-up operations. Through a Department of Energy (DOE) Program for Research and Development Announcement (PRDA) that is being managed by the Morgantown Energy Technology Center, the VAC*TRAX system has been successfully tested on RCRA and TSCA regulated streams, as well as RCRA mixed waste from the DOE. Rust has additionally proven the usefulness of the VAC*TRAX system on TSCA mixed waste sludge and debris, and batch testing indicates that elemental mercury removal from waste streams can likewise be performed to a high degree of efficiency.

The VAC*TRAX process thermally separates volatile or semi-volatile chemicals from contaminated solids; heating the solids under conditions of high vacuum, the contaminants are swept into a nitrogen carrier gas and conveyed to a series of heat exchangers, where they are condensed and recovered. The solid waste, once loaded into the VAC*TRAX dryer, is completely sealed from the atmosphere. This, combined with a strong vacuum pulled on the system and an aggressive pollution control set-up, provides the control needed for the handling of radioactive materials. The solids are heated indirectly, by hot oil that passes through metal jackets in the dryer, with the metal then heating the solids. Nitrogen purge gas, as well as the high vacuum on the dryer, prevent oxygen accumulation in the system, assuring that thermal desorption, not combustion, is the treatment operation. Based on the low flow-rate of carrier gas required for the system, HEPA filters and activated carbon beds can be economically used at the end of the condensing train, preventing the release of radioactivity and organic contaminants to the air.

The pilot equipment used for testing can achieve hot oil temperatures of 260-290°C and a vacuum down to 50 mm Hg absolute pressure. Agitation of the sample is provided by a heated, paddle-type agitator, and nitrogen carrier gas is also supplied to the system. These conditions provide the driving force required to desorb volatile and semi-volatile chemical compounds, including VOCs, PAHs, PCBs, and even elemental mercury within a reasonable batch-time. The pilot system has handled soil and sludge, as well as such debris as personal protective equipment (PPE), plastic bags, rubber hose, cloth filters, metal filter housings, rags, glass bottles, and various wood scraps. Some size reduction was required for the debris streams, but this was significantly less than what would be required for a continuous process utilizing screw conveyors and the like.

Technology Description - the VAC*TRAX Pilot System

The VAC*TRAX process is shown in Figure 1. As can be seen in the figure, material is loaded into the dryer through a flanged port. In the pilot unit, up to 31 L of material may be charged per batch. After loading, the inlet flange is sealed, the dryer is purged with nitrogen, and a strong vacuum is pulled on the system. The dryer is then heated by a conventional hot oil unit, circulating heated oil through an internal agitator and external jacket on the dryer. The heated dryer, in turn, heats the waste material charged to the system. The waste is heated at a time and temperature appropriate to the matrix and contaminants of concern: removing high concentrations of PCBs from soil would typically require heating the material at 260°C for at least 6-8 hours, while acetone or other volatile contaminants could be removed from the same matrix in less than half this time. After being heated for the required time, the vacuum on the system is reduced, and the product is dropped through a hinged door on the bottom of the dryer into a steel vessel that is fastened to the unit. The unit is then ready to begin another treatment cycle, and the solids from the previous batch may be sampled and/or disposed of, as appropriate.

The nitrogen is heated and introduced directly into the dryer. The desorbed vapors and nitrogen exit the dryer through an upper cupola section attached to the dryer. This cupola section is equipped with filter elements that remove entrained solids (dust) from the exiting gas stream. These solids are retained in the dryer and are returned to the treated product from the filters by sending a short burst of nitrogen through the filters and into the dryer, essentially "blowing back" the filters. The solids can then be emptied as previously mentioned. The gases are then condensed in a system employing three heat exchangers in series. The first heat exchanger rejects heat to ambient temperature, while the second and third exchangers are operated at near 0°C with a chiller system. These units condense the water and volatile contaminants that were removed from the waste in the dryer. The liquids drain to collection traps, where they are removed using double block valves. The remaining gas stream, essentially nitrogen with trace volatile constituents and oxygen, is then passed through a high efficiency particulate air (HEPA) filter to provide absolute removal of all remaining solid particles. The stream then passes through an activated carbon adsorption system, removing residual organic vapors that could not be condensed prior to venting of the stream to the atmosphere. Total hydrocarbon emissions from the unit are extremely low; in runs performed on debris spiked with volatile organic compounds, none of the spiked compounds were found in the process vent after the carbon adsorption system. Particulate (and radioactive) emissions are non-detectible.

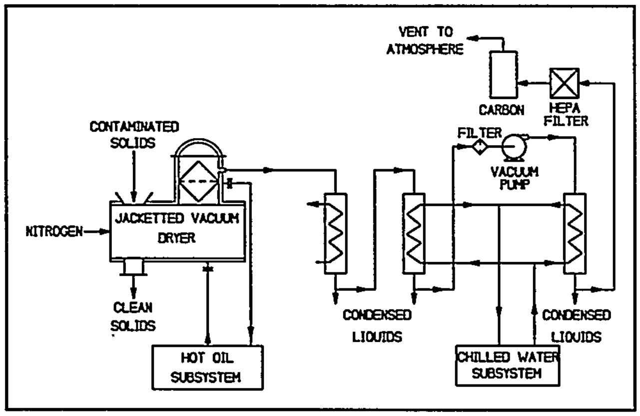

Fig. 1. VAC*TRAX^{SM} Process Flow Diagram

Technology Description - the VAC*TRAX Full-scale System

Based on a market survey, it appears that there is a demand for two sizes of VAC*TRAX systems. These are a small (2-6 drum) unit to handle a large number of low-volume waste streams and a larger (40 drum) unit to handle significant quantities of waste present at a few DOE facilities. At present, the smaller unit is being designed. Rapid mobilization, with minimal cost for installation of utilities and other support facilities are key considerations for the unit. Additionally, the system will be designed to operate within presently existing structures, not requiring that a building be constructed to house this unit. The system is totally self-contained, except for the need for a propane or natural gas fuel source for the hot oil system. The system will be capable of processing waste within a week of arrival on-site, and will be designed for ease of decontamination to decrease demobilization time. These features combine to make the system an economical choice for small waste volumes.

Accomplishments

Over five-hundred hours of testing on spikes, RCRA and TSCA waste, RCRA mixed waste, and TSCA mixed waste have been performed using the VAC*TRAX system. Over 80% of the test runs have been performed on mixed waste. Excellent removal efficiencies have been demonstrated for volatiles, semi-volatiles and PCBs, as shown in Table 1. Even at low temperatures (100°C), the VAC*TRAX system was able to reduce volatiles in a F002 debris waste to below the regulatory level. At 240-260°C, PCBs have been successfully removed from sludge and debris, with starting levels as high as 10% PCBs (100,000 ppm).

In addition to excellent removal efficiencies, the VAC*TRAX system has proven its other main goal, containment of radioactive material within the dryer. For six distinct radioactive waste streams, the condensate recovered from VAC*TRAX testing was found to be non-radioactive based on gross alpha/beta analysis for waste. These results are summarized in Table 2.

Table 1. Contaminant Removal Efficiencies using the VAC*TRAX Process

Contaminant	Solids Matrix	Final Solid Temp (°C)	Time at Temp (min)	Init. Conc.[5] (ppm)	Ending Conc. (ppm)	Removal (%)
Volatiles						
Acetone[1]	Soil/Clay	239	45	1,644	5.98	99.6
Acetone	Debris	101	240	7.51	BQL(0.4)	> 94.5
Carbon Tetrachloride[1]	Soil/Clay	239	45	105	BQL(0.2)	> 99.8
2-hexanone	Debris	101	240	5.93	0.2	96.6
Toluene	Debris	104	360	6.7	0.055	99.2
Tetrachloroethene[1]	Soil/Clay	239	45	118	BQL(0.2)	> 99.8
Tetrachloroethene[2]	Soil/Lint	257	360	1,000	J(0.003)	>99.99997
Tetrachloroethene[2]	Sludge	243	210	272	BQL(1)	> 99.6
Ethylbenzene[1]	Soil/Clay	239	45	630	BQL(0.2)	> 99.97
Ethylbenzene	Debris	104	360	20	0.075	99.6
MIBK	Debris	104	360	15	0.14	96.8
Xylene[1]	Soil/Clay	239	45	2,672	BQL(0.2)	> 99.993
Xylene	Debris	104	360	110	0.092	99.92
Styrene[1]	Soil/Clay	239	45	461	BQL(0.2)	> 99.96
Styrene	Debris	104	360	230	0.14	99.94
Trichlorofluoromethane	Sludge	243	210	2,701	BQL(1)	> 99.92
Semi-volatiles						
Bis(2-ethylhexyl)phthalate[1]	Soil/Clay	239	45	577	1.05	99.8
Pentachlorophenol	Soil/Clay	247	90	581	126	83.2
PCBs						
Arochlor 1242	Soil/Clay	260	600	990	BQL(1)	> 99.90
Arochlor 1242	Soil/Clay	257	240	760	BQL(1)	> 99.87
Arochlor 1254	Sludge	241	1710[4]	107,000	BQL(0.5)	>99.9995
Arochlor 1254	Debris	241	270	500	BQL(0.5)	> 99.90
Arochlor 1260	Debris	241	350	178	BQL(0.5)	>99.7
Elemental Mercury[3]	Soil	252	360	70,700	8,800	87.6

Notes:

1. Test Performed on spike sample
2. Test performed on mixed waste
3. Test performed in a vacuum oven test; the VAC*TRAX system is known to perform significantly better for most contaminants.
4. Test run divided over 4 days of operation
5. All results are converted to a dry weight basis

BQL(A) = Below quantitation limit; A = quantitation limit
J(A) = Estimated value of A, below quantitation limit

Table 2. Containment of Radioactivity within the VAC*TRAX Dryer

Radioactive Constituent		Average Level in Feed	Level in Composited Condensate	% Radioactivity carried to condensate
Co-57	pCi/g	16.91	BQL(0.2)	< 1.2
Co-60	pCi/g	44,000	BQL(0.16)	< 0.00037
Cs-134	pCi/g	25.0	BQL(0.16)	< 0.64
Cs-137	pCi/g	1,195	BQL(0.2)	< 0.017
Sb-125	pCi/g	19.5	BQL(0.48)	< 2.5

Future Expectations

Construction of the small (2 to 6 drum) commercial unit is expected to begin in October of this year. Applications for the required permits will be produced and submitted along a parallel track, thus limiting any unnecessary delays. Demonstration of the full-scale system is expected to first be performed during the fall or winter of 1996. After this, the unit will be used for the remediation of various DOE mixed waste streams. Construction of the large (40 drum) unit will depend on the market demand for the system.

Acknowledgements

A significant portion of the VAC*TRAX demonstration program has been funded by a Program for Research and Development Announcement (PRDA) managed by Bill Huber at the Morgantown Energy Technology Center (METC).

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Session 2

Mixed Waste Characterization, Treatment, and Disposal Focus Area

2.1 Development Studies of a Novel Wet Oxidation Process

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Needs

Many DOE waste streams and remediates contain complex and variable mixtures of organic compounds, toxic metals, and radionuclides. These materials are often dispersed in organic or inorganic matrices, such as personal protective equipment, various sludges, soils, and water. Incineration and similar combustive processes do not appear to be viable options for treatment of these waste streams due to various considerations. There is a need for non-combustion processes with a wide application range to treat the large majority of these waste forms. The non-combustion process should also be safe, effective, cost-competitive, permit-able, and preferrably mobile.

Objectives

The over all objective of the effort described here is to develop a novel catalytic wet oxidation process for the treatment of these multi-component wastes, with the aim of providing a versatile, non-combustion method which will destroy hazardous organic compounds while simultaneously containing and concentrating toxic and radioactive metals for recovery or disposal in a readily stabilized matrix.

Approach

Although the DETOX℠ process had been investigated to a limited extent for application to mixed wastes, there was not sufficient data or experience with the process to determine its full range of application to multicomponent waste forms such as DOE site wastes and remediates.

First, the potential applications of the process needed to be better identified. Following identification of potential applications, the process needed to be demonstrated on waste and remediate types on a practical scale in order that data could be obtained on application range, equipment size, capital and operating costs, effectiveness, safety, reliability, permit-ability, and potential commercial applications of the process. The basic approach for the project was, therfore, to identify the potential range of applications of the process, and then to demonstrate the process on mixed wastes. The effort for the Phases of the project is as follows:

Phase I

The approach for the Phase I effort was to determine the possible range of applications for the DETOX℠ process and to develop a conceptual design for a demonstration unit.

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Phase II

The approach for the Phase II effort was to conduct a demonstration site survey to determine best sites for demonstrating the process, and perform Title II design of a demonstration unit.

Phase III

The approach for the Phase III effort is to prepare a Demonstration Test Plan, fabricate and assemble a demonstration unit, install the demonstration unit at Savannah River Site's TNX facility, and perform shakedown tests to verify that the unit operates properly.

Phase IV

The approach for the Phase IV effort will be to cold test the demonstration unit at Savannah River Site and hot test the demonstration unit at Weldon Spring Site Remedial Action Project.

Project Description

Technology

The DETOX^{SM} process uses a unique combination of metal catalysts to increase the rate of oxidation of organic materials. The process has been patented by Delphi Research, Inc. in the U.S. and several foreign countries. The metal catalysts are in the form of salts dissolved in a dilute acid solution. A typical catalyst composition is 60.% ferric chloride, 3.-4.% hydrochloric acid, 0.13% platinum ions, and 0.13% ruthenium ions in a water solution. The catalyst solution is maintained at 423.-473. K. Wastes are introduced into contact with the solution, where their organic portion is oxidized to carbon dioxide and water. If the organic portion is chlorinated, hydrogen chloride will be produced as a product.

The major reaction in the process is oxidation of organic compounds by ferric iron. Ferric iron is reduced to ferrous state during the reaction. Ferrous iron is in turn re-oxidized to ferric iron by oxygen bubbled into the catalyst solution. Although iron participates in the oxidation reaction, it is returned to its original state and is not consumed, therefore it meets the criteria for a catalyst. Platinum and ruthenium aid in oxidation of organic compounds by iron, and have been designated "cocatalysts."

Many metals will dissolve to an extent in the catalyst solution. This property of the solution can be used to remove toxic and radioactive metals from larger amounts of more inert inorganic material. Since the catalyst solution keeps metals in the ionic state, one does not have to be as concerned with escape of volatile metals such as mercury and cadmium. Many metals can be concentrated in the catalyst solution, if desired, for eventual disposal or recovery. The catalyst solution can be reduced to an iron oxide matrix by boiling off hydrogen chloride and water. The iron oxide matrix is a good form for stabilization by one of a number of methods, preparatory to ultimate disposal of inorganic contaminants.

Project Effort

Phase I. In Phase I, destruction efficiencies were measured for six organic compounds, the fates of representative metals in the catalyst solution were determined, the ability of the catalyst solution to treat contaminated soils was established, and a conceptual design for a field demonstration unit was performed.

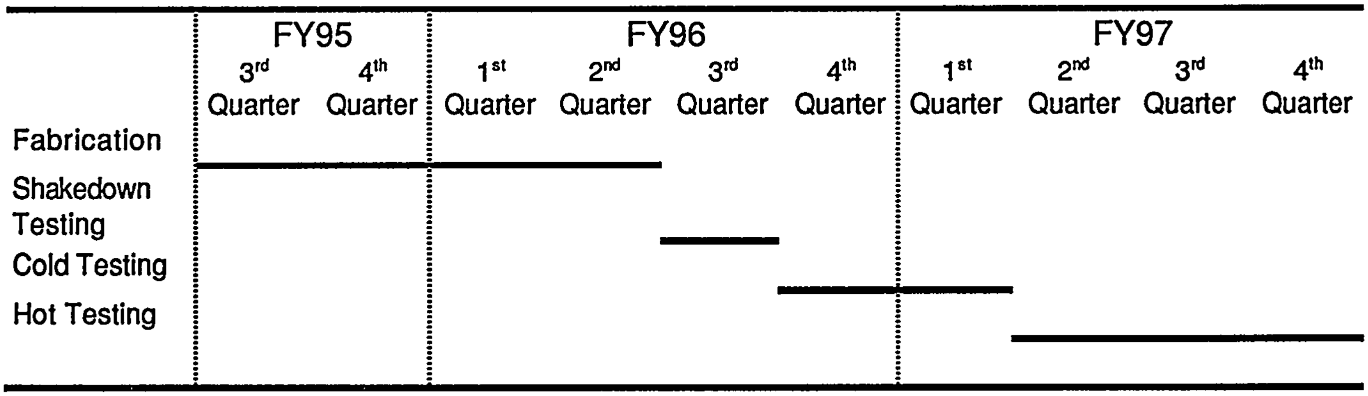

Figure 1: Schedule and Milestone FY95-FY97 Program Schedule

Phase II. In Phase II, a demonstration site selection survey was conducted and detailed engineering design for a modular, skid-mount demonstration unit was performed.

Phase III. Phase III, now ongoing, is preparation of a Demonstration Test Plan (DTP), and fabrication and shakedown testing of the demonstration unit with mineral oil and tributyl phosphate.

Phase IV. Phase IV will be demonstration of the process on a variety of hazardous materials at SRS and on a variety of low-level mixed wastes at WSSRAP.

Results

Phase I

The DETOX^{SM} process was found to be capable of destroying organic compounds with good efficiency, solvating many toxic metals, and removing organic compounds and toxic metals from soils. The results of Phase I have been presented at the 1994 METC meeting.

Phase II

Sites selected for demonstration were Savannah River Site and Weldon Spring Site Remedial Action Project. A Title II design for the demonstration unit was completed. The results of Phase II have been presented at the1994 METC meeting.

Phase III

Demonstration Test Plan. A DTP has been completed following guidance supplied by METC, and has been approved by the Primary Stakeholders in the demonstration effort.

One of the primary goals of the DTP is to establish lists of stakeholders in the demonstration effort. The stakeholders were divided into three categories.

Primary stakeholders were those parties who are directly involved in the demonstration, and included Delphi, JAT, METC, EM-50, SRS, and WSSRAP. RFETS has been added to the list of Primary Stakeholders due to their great involvement in development and potential use of

DETOX^{SM}. Approval of the DTP by the Primary Stakeholders is required.

Secondary Stakeholders in the demonstration are those parties which have a direct interest in the demonstration, those being representatives for EM-30, EM-40, EM-50, and EM-60, demonstration site representatives, the MIxed Waste Focus Area, as well as the EPA and regulatory agencies in pertainent regions and states. Secondary Stakeholders were asked for input to and comments on the DTP.

Extended Stakeholders are those parties which may represent potential users of the technology. Extended Stakeholders include other DOE sites, members of the Mixed Waste Focus Area, DOD waste offices, the Defense Nuclear Safety Board, the National Institute of Occupational Safety and Health, the Western Governer's Association DOIT Committee, the State of New Mexico, the California Water Resources Control Board, and other EPA Regions. The DTP gives the project plan, schedule, and the organization by which the project plan will be accomplished. The DTP is considered to be a living document, and refinements continue to be added with the Primary Stakeholders' approval.

Permit Process and NEPA Documentation. Permitting for the demonstration unit is being pursued for both the demonstration sites. A viable plan has been formulated for each site. Delphi has received a letter from EPA stating that the DETOX^{SM} process unit is considered to be a miscellaneous unit for the pruposes of permitting under RCRA.

After consultation with SRS environmental personnel and representatives of South Carolina Department of Health and Environmental Control, it was determined that the route to pursue for the demonstration at the TNX facility was to obtain a letter of approval from SCDHEC to operate the demonstration unit as a wastewater treatment unit under the clean water act. This approach required preparation of estimates of water effluent for the demonstration unit during the demonstration. The effluent from the demonstration unit will meet the requirements of NPDES after treatment in the TNX facility's Organics Removal Facility (ORF). Estimates of air emissions have also been submitted. The demonstration unit should not require an air permit. A determination has been made that the demonstration at SRS can be conducted under a NEPA Categorical Exclusion (CX). Delphi has supplied information to SRS and filled SRS's Environmental Evaluation Checklist to support the CX.

As WSSRAP is a CERCLA site, no formal permit is required for operating the demonstration unit. However, the substantive requirements of a RCRA Part B RD&D Permit must be met for the demonstration to proceed. Estimates of air emissions from the demonstration unit have been supplied to WSSRAP, and an air dispersion model performed for WSSRAP by Argonne National Laboratory. The results of the air dispersion model indicated that no air permit was required for the demonstration unit if the emission stack height was 16. feet or greater. Delphi is in the process of preparing information on the demonstration for meeting the substantive requirements of RCRA Part B. A determination has been made that the demonstration at WSSRAP can be conducted under a NEPA Categorical Exclusion (CX). No NEPA information is required from Delphi by WSSRAP.

Engineering Design. Final design engineering is being completed and equipment purchase has begun. Several changes have been made in the unit design to improve operation of the unit.

In order to reduce volatile organic compounds in the vent gases, a secondary reaction vessel has been added to the unit. The secondary reaction vessel contains a bubble tower of DETOX^{SM} solution through which product gases from the primary reaction vessel must pass. The height of the DETOX^{SM} solution in the bubble tower is designed to provide sufficient residence time to give four to five "nines" destruction of volatile organic organic compounds in the gases.

More accurate simulations of the demonstration unit overhead system identified a shortcoming of the Title II design. When non-chlorinated compounds are being oxidized in the system, it is desired to operate the reflux condenser at near 100. C to return the majority of HCl in the product gases to the primary reaction vessel. However, at this temperature not enough water could be carried through the reflux condenser with the anticipated product gas flow to remove product water from the DETOX^{SM} reaction vessel. Thus, more gas flow through the reflux condenser was necessary, which involved adding a gas recycle compressor to the system to return vent gas through the reflux condenser. This arrangement worked well in simulations, and by adjusting recirculated gas flow the amount of water removal can be closely controlled.

Consideration of solids rinsing determined that rinse volumes could be reduced significantly by reusing one of the acid rinses and eliminating the water rinses (since the solids would be treated by some stabilization process in any event). The main objective of rinsing is to return the large majority of the residual DETOX^{SM} solution in the solids to the primary reaction vessel, which the acid washes accomplish. Therefore, it was determined that the primary reaction vessel was sufficient to evaporate the water from the acid rinse, and that the evaporator, evaporator condenser, evaporator overhead receiver, and associated equipment could be eliminated from the design. This design change resulted in simplification of the demonstration unit and lowered capital costs.

Modified PFD, P&IDs, heat and material balances, and equipment data sheets were prepared, and the design changes were re-HAZOPed. Tantalum and titanium equipment pieces have now been ordered and detailed design is under way.

Operations and Safety Analysis. One operational concern which needed to be addressed was the identification of conditions where explosive mixtures could be formed in the headspace of the primary reaction vessel. Computer simulations of organics and oxygen in the DETOX^{SM} headspace gases, conducted by Sandia National Laboratories explosives group, had established that there were compositions which could be explosive. However, experiments on actual mixtures needed to be conducted. SNL's explosives group conducted these experiments in a test fixture supplied by Delphi. Mixtures of organic vapor, oxygen gas, and the steam content of a DETOX^{SM} reaction vessel headspace were sparked with an electrical discharge to ignite any flammable or explosive compositions. Organic compounds used were toluene, acetone, and hexane. Only one explosive event was experienced, and for all organics there was no flammability below oxygen concentrations of approximately 5.% by volume. The design parameter for oxygen in the vent gases from the demonstration is approximately 5.% by volume, which translates to less than 1.% by volume in the primary reaction vessel headspace. This leaves a margin for error or upset without generation of any flammable mixture. In addition, a nitrogen inlet line has been added to the primary reaction vessel so that it may be purged of oxygen if excessive levels start to develop.

The University of New Mexico's Chemical and Nuclear Engineering Department is conducting simulations of the demonstration unit to identify operating conditions for the unit and the response of the demonstration unit to potential upset conditions. These studies are on-going as of this report.

Personnel at Argonne National Laboratory are investigating the use of a chemically bonded phosphate ceramic waste form to stabilize the DETOX^{SM} solution residue. Samples of the ceramic have been prepared using simulants of the solution residue loaded with lead and mercury, and ferric phosphate solids which will be produced from tributyl phosphate treatment. The samples show good physical strength and are non-hazardous for lead by the standard EPA Toxicity Characteristic Leaching Procedure. Phosphate ceramic also has very low leach rates for the radionuclide simulants cesium and cerium.

Personnel at SRS are conducting a Process Hazards Review (PHR) to determine the safety requirements for the demonstration. Delphi and JAT have supplied SRS with documentation on the demonstration unit, the demonstration project, and permitting to support the PHR. The PHR is on-going as of this paper.

Benefits

The DETOX^{SM} process is a viable alternative to incineration and similar high temperature or combustive-type processes for the treatment of organic mixed wastes.

Because the process is conducted at relatively low temperatures and pressures, its emissions are more easily controlled than many other thermal treatment processes. This gives advantage not just in operations, but in permitting.

The DETOX^{SM} process can destroy a wide variety of organic materials, making it versatile in application. The DETOX^{SM} solution's ability to solvate toxic and radioactive metals allows seperation of these metals from inert materials in a waste stream. Although some small amount of radioactive material will almost always remain in the inert portion, it will no longer be a mixed waste, and if the input waste was TRU the inerts will in many instances be sufficiently low in radioactivity to be low-level waste only. Careful monitoring of the DETOX^{SM} solution when processing TRU waste can also assure that the process solution residue is also low-level only.

Estimated costs for waste treatment using the process range from \$2.50/kg to \$25.00/kg, depending on the size of the unit and the amount of waste processed. Estimated cost of waste treatment with the 25. kg (dry weight, organics) demonstration unit is \$9.40/kg (dry weight, organic). Process units can be mobile for on-site treatement of wastes.

Future Activities

The demonstration unit will be installed at Savannah River Site's TNX facility, and tested for functionality using mineral oil, tributyl phosphate, and organic solids composed of personal protective equipment (PPE). These tests will establish the suitability of the unit for treatment of hazardous and mixed wastes.

Phase IV is the planned demonstration of the unit on hazardous and mixed wastes . Phase IV demonstration at SRS will use hazardous organic wastes only, to evaluate the performance of the demonstration unit in a non-radioactive setting. The unit will treat an assortment of hazardous wastes including oils contaminated with toxic metals, non-halogenated solvents, halogenated solvents, and contaminated solids. After the hazardous waste experimental campaign, the unit will be cleaned, inspected, serviced, and transported to WSSRAP. This will demonstrate transportability of the unit. At WSSRAP, the unit will be installed, checked out, and used to treat a variety of low level mixed wastes including contaminated ethylene glycol, contaminated oils and solvents, paints and paint sludges, PCB wastes, and tributyl phosphate contaminated with PCBs, uranium, thorium, and mercury. A commercialization assessment will be performed during the demonstration, which will include conceptual design of a baseline commercial process unit and sensitivity analysis for factors affecting performance and cost. Following the demonstration, a determination will be made as to the disposition of the unit and its possible future use at WSSRAP or other sites.

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2.2 Innovative Vitrification for Soil Remediation

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1.0 Introduction[1]

The objective of this DOE demonstration program is to validate the performance and operation of the Vortec Cyclone Melting System (CMS™) for the processing of LLW contaminated soils found at DOE sites. This DOE vitrification demonstration project has successfully progressed through the first two phases. Phase 1 consisted of pilot scale testing with surrogate wastes and the conceptual design of a process plant operating at a generic DOE site. The objective of Phase 2, which is scheduled to be completed the end of FY 95, is to develop a definitive process plant design for the treatment of wastes at a specific DOE facility. During Phase 2, a site specific design was developed for the processing of LLW soils and muds containing TSCA organics and RCRA metal contaminants. Phase 3 will consist of a full scale demonstration at the DOE gaseous diffusion plant located in Paducah, KY. Several DOE sites were evaluated for potential application of the technology. Paducah was selected for the demonstration program because of their urgent waste remediation needs as well as their strong management and cost sharing financial support for the project.

During Phase 2, the basic nitrification process design was modified to meet the specific needs of the new waste streams available at Paducah. The system design developed for Paducah has significantly enhanced the processing capabilities of the Vortec vitrification process. The overall system design now includes the capability to shred entire drums and drum packs containing mud, concrete, plastics and PCB's as well as bulk waste materials. This enhanced processing capability will substantially expand the total DOE waste remediation applications of the technology.

A total of seven (7) soil vitrification trials were conducted during Phase 2 at Vortec's pilot scale vitrification plant located at the University of Pittsburgh Advanced Research Center in Harmarville, PA. The first set of trials used a surrogate soil composition representative of the contaminated soils found at DOE's Hanford site. The Hanford soil was simulated using the composition data supplied by Hanford, and was spiked with Resource Conservation and Recovery Act (RCRA) metals surrogates, an organic contaminant, and surrogate radionuclides. The sampling of the effluent and influent streams taken during the tests confirmed that virtually all of the refractory radionuclides were retained in the glass and would not leach to the environment—as confirmed by both Product Consistency Tests (PCT) and Toxicity Characteristic Leaching Procedure (TCLP) testing. The organic contaminant, anthracene, was destroyed during testing with a Destruction and Removal Efficiency (DRE) of at least 99.99%. Semi-volatile RCRA metal surrogates were captured by the Air Pollution Control (APC) system, and data on the amount and the chemical composition of the particulate were established for use in the APC system design.

A second set of three pilot scale vitrification trials were conducted using surrogate contaminated soil representative of the soil found

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at the DOE-Paducah site. The DOE-Paducah waste streams are much more complex than the soils which were scheduled for remediation at Hanford. The Paducah contaminated soils contain, mud as well as PCB contamination, and construction debris. The surrogate tests performed with the Paducah waste simulants reconfirmed the ability of the vitrified waste to pass TCLP and PCT.

2.0 Program Objectives

The principal objective of the METC/Vortec program is to demonstrate the ability of the Vortec 36-72 ton per day CMS™ to remediate DOE contaminated mixed waste and other waste forms, contaminated with both hazardous materials and low levels of radionuclides and PCB, by producing glass which passes TCLP and PCT.

To convincingly demonstrate the melter capability, a Demonstration Plant will be constructed and operated at DOE-Paducah where there is a need for the remediation of contaminated waste. The following other objectives will be met during the program.

1. Determine the glass chemistry requirements to achieve effective vitrification of contaminated soils found at the Paducah site; that is, given a particular soil (waste), determine how its oxide composition must be modified to produce a vitrified product that will immobilize contaminants over the long-term.
2. Determine expected feedstock soil particle size distribution and the glass flux requirements, so that valid designs and cost estimates can be made for the feedstock preparation system.
3. Determine the Destruction Removal Efficiency (DRE) of the CMS™ for organic contaminants likely to be found in soil from DOE sites requiring remediation.
4. Establish the characterization of the off-gas so that valid designs and cost estimates can be made for the flue gas clean-up system.
5. Establish the cost of a fully integrated soil (waste) vitrification CMS™ with a 36-72 TPD capacity of waste into the CMS™.
6. Conduct start-up, shake-down, and feasibility demonstrations using the fully integrated plant with a capacity of 36-72 TPD constructed at the DOE-Paducah site. This capacity equates to approximately 160-320 barrels/day containing 30% moisture and an average weight of 450 lbs/barrel.
7. Establish the contractual and operating arrangement of Phase 4 for the continued operation of the facility by Vortec to continue remediation at Paducah site and other site's waste streams.

3.0 Background Information

The Department of Energy's goal to clean-up its nuclear complex by the year 2019 requires the development of innovative technologies to convert soils contaminated by hazardous and/or radioactive wastes to forms which can be readily disposed in accordance with current waste disposal methods. These technologies must be able to accomplish this task with minimum public and occupational health risks, with minimum environmental risks, and in a timely and economical manner. Additionally, the technologies must transform the hazardous and/or radioactive waste into a form which is considered non-hazardous; which has long-term stability to prevent migration of radionuclides, and can thus be disposed in an environmentally safe manner; and which satisfies all federal, state, and local emissions regulations. It is imperative that the technology not present any major obstacles to its own safe decontamination and decommissioning. Finally, the final waste form produced must be very stable since some of the materials have very long half-lives that may greatly exceed the capability of institutional controls to protect the environment.

The unique features of the CMS™ technology make it a particularly cost-effective process for the vitrification of soils, sediments, sludges, and other solid wastes containing organic, metallic, and/or radioactive contaminants. Many of the benefits of the CMS™ technology recognized by the glass and hazardous waste management industry would also apply to DOE's ER&WM needs. Benefits with respect to DOE's needs are:

1. The ability of the CMS™ to produce a product which provides for long-term immobilization of heavy metals, toxic inorganics, and radionuclides.

   In numerous pilot scale tests conducted by Vortec, the CMS™ has demonstrated the ability to effectively process RCRA wastes as well as surrogate contaminated soils. Simulated radionuclides and RCRA metals are effectively retained in the glass product and do not leach when tested using both the PCT and TCLP.

2. The CMS™ has demonstrated the ability to effectively oxidize and destroy organic contaminants. Tests performed by Vortec in the U-PARC facility with various carbonaceous materials such as cyanides and other organic contaminants found in most industrial waste, and anthracene and 1, 2- dichlorobenzene as surrogates for organic and PCB contaminates, have validated the organic destruction performance of the CMS™.

3. The CMS™ has demonstrated substantial flexibility with respect to the processing of various types of solid wastes and can accommodate substantial variations of feedstock composition.

   Vortec has completed more than 109 test programs using a variety of materials as feedstocks including the U.S. Environmental Protection Agency (EPA) contaminated soils, flyash, baghouse dust, metal plating sludges, aluminum industry waste, steel industry waste and virgin glass making components. Soils with water content of up to 50 weight percent have been processed into glass products.

4. The CMS™ demonstrated the ability to oxidize and vitrify waste materials introduced as slurries, providing the capability for mixing contaminated or waste oils with various types of hazardous solids, soil wash process sediments, and mill tailings. In addition to contaminated soils. Vortec has demonstrated the ability to vitrify Hanford low level tank waste surrogates with a water content of approximately 70% liquid and 30% solids. In addition, the CMS™ has demonstrated the ability to effectively vitrify a spectrum of metal plating sludges at 60% water content.

5. The CMS™' high temperature process components have water-cooled, steel walls providing for a sealed process which can be operated at negative pressure to prevent leakage of contaminated gases to the atmosphere. These water-cooled components can continue to operate in the event that unusual wear or spalling of refractory occurs until such time as the unit can be safely shut down.

6. The CMS™ small physical size reduces the decommissioning and disposal costs of the process equipment at the end of its useful life. The 36-72 ton/day demonstration unit is being designed to be transportable and modular, thus enabling wastes at several waste sites at a given facility to be serviced.

7. In the processing of substantial quantities of contaminated soils, the life cycle cost of the Vortec CMS™ is lower than other existing vitrification processes. In commercial applications, a 72 TPD CMS™ process unit typically has total processing costs in the range of $50 - $100 per ton of material processed. Radionuclide and PCB contamination increases the per-ton cost somewhat, depending upon the specific activity of the soil and the nature of the PCB contamination. Vortec estimates that the processing costs of low level waste with mixtures of TSCA or RCRA wastes at Paducah will be in the range of $50 to $200 per barrel for the Paducah drummed wastes. In comparison, of the types of wastes to be processed, other competing remediation technologies have estimated

processing costs in the range of $500 to $1000 per barrel for drummed wastes.

Vortec, during the first two phases of this program, has demonstrated the CMS™ technology's ability to remediate surrogate soils. During Phase 2 of the program, the final design and cost estimate for a 36-72 TPD system were also developed. The baseline plant design included a feedstock preparation subsystem combining the waste (average moisture content of 30%) with glass making additives, the CMS™ subsystem, and an air pollution control (APC) subsystem that cleans the flue gas using a venturi scrubber, wet electrostatic precipitator (WESP), and HEPA filters.

3.1 Process Description

The primary components of the basic CMS™ are a counter-rotating vortex (CRV) combustor and a cyclone melter. An artist's rendering of the basic CMS™ concept is shown in Figure 3.1-1. A unique feature of the process is the rapid suspension heating and oxidation of feedstock materials in the CRV combustor prior to the physical and chemical melting processes which occur within the cyclone melter. The use of the Vortec CRV combustor in conjunction with a cyclone melter distinguishes the Vortec combustion and melting technology from other types of cyclone combustion systems. In the CMS™ process, granular glass-forming ingredients and other feedstocks are introduced into the top region of the CRV combustor along with fuel and combustion air. As a result of the intense counter-rotating vortex mixing, it is possible to achieve stable combustion in the presence of large quantities of inert particulate matter (solids-to-gas mass ratios on the order of 1:1). Both convection and radiation heat transfer mechanisms contribute to the rapid heating of the feedstock materials within the CRV combustor. Any organic contaminants in the feedstocks are also effectively oxidized.

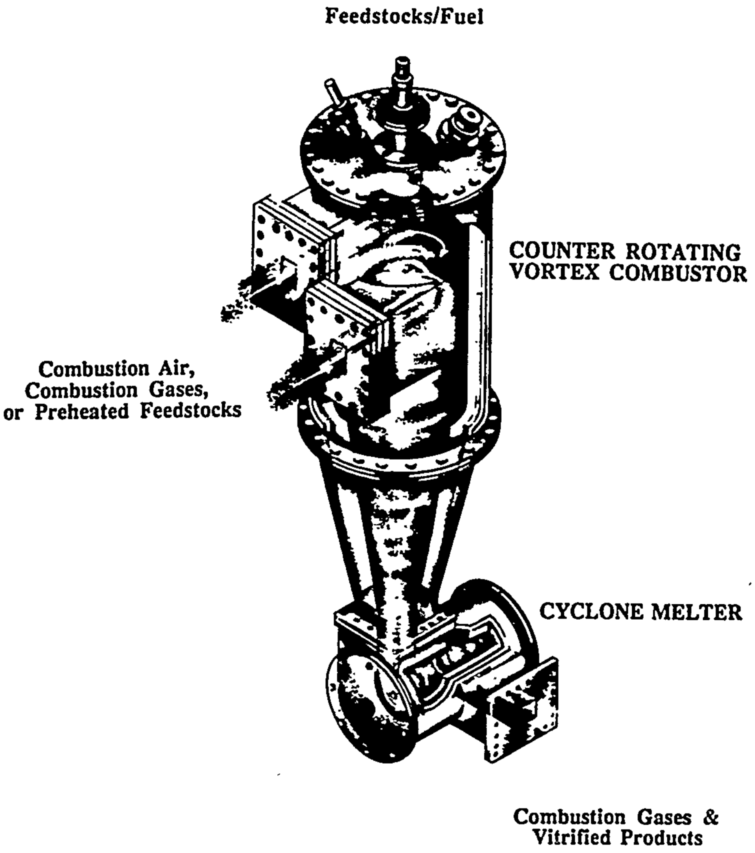

Figure 3.1-1. Artist Rendering of Basic Combustion/Melting System

The melted product formed in the cyclone melter, and the combustion products, exit through the melted material is collected. The vitrified a tangential channel and enter a separator-reservoir (not shown in the figure) where a pool of material exits the reservoir through a bottom or side tap, and the flue gases exhaust to a water quench/electrostatic precipitator assembly or an optional heat recovery unit for combustion air preheating.

The flue gas exiting the separator-reservoir is treated in an air pollution control assembly prior to being exhausted out the stack. As a result of the high thermal efficiency of the Vortec CMS™, the flue gas flow rates are relatively modest. Because the temperature and composition of the vitrified product can be closely controlled, the amount of process fuming (volatile carryover) can also be minimized.

The average gas-solids suspension temperature leaving the CRV combustor is typically on the order of 2000°F to 2700°F, and is a function of the product being vitrified. The process temperatures in the cyclone melter are typically in the range of 2000°F to 3000°F, depending on the melting characteristics of the feedstock being processed. The nitrogen oxide emissions have been found to be substantially lower than those which occur in conventional cyclone combustors. Excess air levels are typically in the range of 5 to 20% depending on the makeup and the nature of the feedstock being processed.

Heat rates demonstrated by the Vortec pilot scale facility typically ranged between 3.5 and 6 million Btu/ton at a glass production rate of 15 TPD. This heat rate is 50% to 80% lower than heat rates for conventional gas-fired glass melting at similar capacity. The energy savings are primarily due to more efficient heating of the glass ingredients in suspension by the products of combustion and lower structural heat losses due to the small physical size of the process components. The CMS™ can also accommodate the use of a variety of fuels, such as oil and coal-derived fuels, and even organic waste materials.

Most of the tests conducted to date have used a dry, shredded, or pulverized feedstock pneumatically transported and injected into the CMS™, and have used natural gas and/or coal as the fuel.

The CMS™ pilot system has demonstrated NO_x emissions of less than 4 pounds per ton of vitrified product, meeting the California emission standard for glass melting furnaces—currently the most stringent in the United States. With natural gas as the primary fuel, the NO_x emissions, calculated as NO_2, have typically been approximately 2 pounds per ton of product. Rapid temperature quenching of the combustion products by the inert solid particles and staged combustion are the primary means of limiting NO_x emissions. Tests conducted for Hanford using a high nitrate concentration tank simulant resulted in no visible plume leaving the pilot plant's stack.

The CMS™ has demonstrated uncontrolled emissions levels of less than 0.5% of feed materials which did not contain low temperature volatiles, such as utility flyash. For materials containing heavy metals and other volatiles, such as MSWI flyash and fiberglass waste, the uncontrolled emissions levels have typically been in the range from 1% to 4%. As part of the METC program, Vortec has demonstrated a procedure for recycling the volatile materials into the glass after the glass has left the high temperature region of the combustor.

3.2 Previous Research Accomplished

After four years of design evolution under various DOE and EPA programs, the CMS™ is completely operational at the U-PARC test facility. Vortec's system has demonstrated the production of glass and the vitrification of a variety of feedstocks, including:

- EPA surrogate soils,
- Spent Pot Liners (K-088) wastes
- Coal fired boiler ash,
- Sewage Sludge ash,
- Auto shredder residue ash
- Municipal solid waste incinerator ash,
- Metal Plating Sludges
- Fiberglass waste with organic contaminants,
- Dusts containing heavy metals and organic materials,
- Electronic industry wastes

4.0 Results

Vortec has successfully completed the verification testing and final baseline plant design required in Phase 2 of this program. Vortec will continue the development, construction, and operation of the CMS™ Demonstration Plant during Phase 3. Vortec believes that the CMS™ technology is at the stage of development that will result in a mature process that is directly applicable to a large number of DOE Environmental Restoration and Waste Management (ER&WM) needs. Vortec is developing the CMS™ technology to commercial readiness, with the intention of economically meeting all public, occupational, and environmental health and safety requirements for remediation technology. Commercial offerings of the CMS™ technology, in plant sizes up to 200 TPD, have been made during the last year.

4.1 Test Program-Phase 2 Results Summary

A total of seven soil vitrification trials were conducted at Vortec's pilot scale vitrification plant located at U-PARC during Phase 2 of the program.

The objectives of the pilot testing were to:

- Demonstrate the effective vitrification of low level waste streams (soil) with the characteristics and compositions found at DOE-Hanford and DOE-Paducah;
- Evaluate the melting performance of the CMS™ with this material, that is, the feedstock composition and viscosity relationship;
- Define the expected range of flue gas emissions;
- Optimize the system operating parameters for the waste by determining the effect of temperature on the melting performance of the CMS™ and on the capture rate of the surrogate contaminants in the vitrified product; and to
- Determine the PCB destruction efficiency of the CMS™.

The first set of trials used a surrogate soil composition representing the contaminated soil found at DOE's Hanford site.

The trials associated with the Hanford waste stream are noted below:

Test No.	Test Date	Surrogate Soil
85	6/8/94	Hanford
86	6/9/94	Hanford
94	10/5/94	Hanford
95	10/6/94	Hanford

The objective of the first two tests, Tests 85 and 86, was to demonstrate the vitrification of soil having the characteristics and composition of the soil found at Hanford (the DOE selected site at that time). These first tests also evaluated the melting performance and expected range of flue gas emissions from the CMS™ when processing the Hanford surrogate soil feedstocks. The surrogate soil feedstock consisted of a synthetic soil spiked with surrogate heavy metal and radionuclide contaminants. During each test, variations in system operating parameters were investigated to search for optimum operating temperature conditions and glass viscosity. Success was measured by the ability of the CMS™ to produce a fully-reacted vitrified product which passes both the Toxicity Characteristic Leaching Procedure (TCLP) test for leaching of the surrogate metal contaminants and the Product Consistency Testing (PCT) for the chemical durability of the vitrified product. Samples of all effluent streams were analyzed to establish the partitioning of the heavy metal and radionuclide surrogates.

The objective of the second two tests, Tests 94 and 95, was to determine the effect of temperature on the melting performance of the CMS™ and to confirm the capture rates of the surrogate heavy metal and radionuclide contaminants in the vitrified product. Analyses of the off-gas were conducted to establish the design specification for the air pollution control system.

A second set of pilot scale vitrification trials were conducted using surrogate contaminated soil representative of the soil found at the DOE-Paducah site. The trials associated with the DOE-Paducah waste stream that have been completed are noted below:

Test No.	Test Date	Surrogate Soil Type
101	3/1/95	Paducah
107	6/21/95	Paducah
108	6/21/95	Paducah

The DOE-Paducah waste streams are much more complex than the soil fines which were scheduled for remediation at Hanford. The test results from the last two tests are still under review, and additional tests planned to establish the performance of the CMS™ when handling the wide variety of materials in storage at the DOE-Paducah site.

The surrogate soil feedstock used consisted of a synthetic soil modeled on the data received from DOE-Paducah from their low level waste inventory. The low level soil was spiked with surrogate heavy metal, radionuclide, and surrogate PCB contaminants. Samples of all effluent streams were analyzed to establish the partitioning of the heavy metal and radionuclide surrogates. Flue gas samples were also analyzed for PCB's.

Glass samples were taken periodically (approximately every 15 minutes) during each test in two forms: glass patty, which was air quenched, and glass cullet, which was water quenched. The glass analysis was conducted on the cullet samples by Corning Engineering Laboratory Services (CELS), and the TCLP testing was conducted by Blue Marsh Laboratories (BML).

From a qualitative standpoint, the glass produced during these tests was consistently dark green in color. Within a single glass sample, only minor color variations could be seen. Composite samples were prepared from the glass cullet samples obtained during steady state operation of the corresponding tests. These composite samples were sent to CELS for chemical composition analysis.

Composite samples were prepared from the glass cullet samples obtained during steady state operation of the corresponding tests. These composite samples were sent to BML for TCLP analysis. The analysis indicated that the TCLP extract contained very little measurable quantities of metals and in all cases were significantly below EPA TCLP limits. PCT test results indicated a Na-normalized leach rate of 0.0032 to 0.015 grams of glass/square-meter/day. The PCT specification for nuclear glasses is a Na-normalized leach rate of no greater than 1.0 grams of glass/square-meter/day.

Mass balances were performed on the surrogate heavy metal and radionuclide contaminants present in the feedstock. The composition of each of the effluent streams were used along with the effluent flow rates to perform elemental mass balances and calculate the partitioning of the contaminants.

Tests 85, 86, 94, and 95 used a surrogate for the soil fines that were to be processed at Hanford. The best data from Hanford indicated that the waste streams contained no organics but did have a full complement of heavy metals along with small amounts of plutonium, cesium, cobalt and uranium. Surrogates for these radionuclides and heavy metals were spiked into soil so that partitioning to the effluent streams could be established.

The best data available from DOE-Paducah during preparations for Test 101, 107, and 108 indicated that the low level waste stream (soil) contained small amounts of organic materials and small amounts of heavy metals, uranium, and plutonium. Since the worst organic material to remediate is the PCB, 1, 2 dichlorobenzene was used as an organic surrogate at a concentration approximately of 1000 PPM, a concentration well beyond what is expected in the actual low-level waste stream. Each of the three tests focused on establishing the DRE for its chemical compound. Test 101 injected the 1, 2 dichlorobenzene as a liquid; Test 107 adsorbed the 1, 2 dichlorobenzene on the surface of granulated carbon particulates to simulate the inclusion of the PCB in a soil matrix; and Test 108 used the same carbon matrix but enriched the air oxygen content to 32%.

The flue gas was sampled for the presence of 1, 2 dichlorobenzene to estimate the destruction removal efficiency (DRE) for the CMS™ pilot plant as it is presently configured without the recuperator. The CMS™ system consistently demonstrated a 99.9% DRE while having a total system gas residence time of less than 1 second. With the addition of a recuperator to the CMS™, a commercial or demonstration system would expect to have a gas residence time in excess of 3 seconds.

In addition, Cerium was included at 500 PPM as a surrogate for uranium or plutonium, and the semi-volatile RCRA metals lead and cadmium were also included. Vortec has shown in the METC and many other tests that approximately 95% to 100% of the non-volatile RCRA metals report to the glass.

As more information is received from the site, additional testing will be required to establish the influence of new contaminants. A good example of this is the influence of plastic drum liners on the flue gas composition. Initial testing at the site indicated that the low level waste stored in barrels contains a plastic liner. Separation of this liner, and any other debris contained in the drum, proved to be difficult to remove and posed a health and safety risk. To avoid this situation, the entire drum will be shredded, and the soil will now contain the shredded plastic liner. This plastic component represents an addition to the feedstock chemistry, and it will have an influence on the off-gas composition. Continued testing during Phase 3 will investigate these and other concerns generated as the multiple waste streams at DOE-Paducah are better defined by the characterization studies being conducted by the site.

4.2 Design Program-Integrated Demonstration Plant

4.2.1 System Requirements

The major system requirements for the Demonstration Plant are as follows:

1. Accept drummed or bulk waste streams with up to 30% moisture. The nominal processing capacity of drummed waste found at Paducah is 160 drums per day. (Note that if the moisture content is greater than 30%, it still can be processed but at reduced capacity). Provisions will be made to enrich the combustion air to approximately 38% oxygen at some later date, thereby allowing for growth in capacity to approximately 320 drums per day.

2. Targeted waste forms are 55 gallon drums of contaminated soils with moisture containing debris such as concrete, tramp metal, wood and plastics. The system will process entire drums, including plastic overpacks, plastic liners and frozen drums. This will minimize health and safety risks and minimize waste characterization analysis costs.

3. The process will be capable of processing waste containing radionuclides, TSCA, and RCRA contaminants.

4. The Demonstration Plant will be transportable and modular allowing use atmultiple DOE sites and/or multiple locations at a single site. Process equipment will be skid-mounted which can be installed on concrete pad foundations.

5. The Demonstration Plant will be capable of processing a wide variety of physical and chemical waste forms throughout the DOE complex. The wastes include soils, sediments, and/or sludges contaminated with hazardous wastes and low-level radioactive wastes. At Paducah, some of the soil contains organic material (solvents, fuels, PCB, plastic liners, etc.). Both volatile (Technetium) and nonvolatile (Uranium, Neptunium, Thorium, and Plutonium) radionuclides may be present in the soil waste stream. The eight heavy metals regulated by 40 CFR 261.24 are also present in the soil. Organic materials that can result in Hazardous Air Pollutants regulated by State of Kentucky 401 KAR 63.022 are also present in selected waste streams. Optional waste streams may include but are not limited to: personnel protective equipment (PPE), HEPA filters, treated scrubber / ESP water particulate (a slip stream is required to control particulate build-up) and spent ion exchange materials.
6. The waste stream will be fed, in dry form, to the melting system with a particle size no greater than minus 30 mesh and a moisture content that enables the material to flow freely. The soils at Paducah, as received, can have a moisture content of 30% and a size distribution well beyond the desired 30 mesh maximum; therefore, drying and grinding processes are included as part of the feed preparation subsystem.
7. The system will produce a glass frit, a chemically stable and reduced volume final waste form, that will pass the Toxicity Characteristic Leaching Procedure. The Air Pollution Control (APC) system will be required to meet DOE/EPA and the State of Kentucky standards for the removal of hazardous material and radionuclides. A venturi

scrubber followed by a single Wet Electrostatic Precipitator (WESP) and HEPA filters are specified to meet the requirements of the site's health, safety, and environmental regulations. This APC system is considered by the State of Kentucky to be the best available technology.

8. The APC process water will have a slip stream to a wastewater treatment process to remove radionuclides and other solids. All solids will be recycled through the CMS™.
9. The vitrified product generated as a result of testing will be disposed on-site or at an approved DOE radioactive waste repository, and will be the responsibility of DOE-Paducah.
10. DOE-Paducah will obtain the permits required to process the waste streams in the CMS™.

4.2.2 Demonstration Plant-System Description

A system flow diagram is shown in Figure 4.2-1. An isometric drawing of the plant arrangement in shown in Figure 4.2-2. The demonstration plant has been designed as a transportable and modular system; that is, the individual, skid mounted components of the process have the capability to be transported by truck, without special permits, to the site, erected, and when operation is complete, dismantled, decontaminated, loaded back onto trucks, and hauled off-site.

As is indicated in the system diagram Figure 4.2-1, contaminated soil is first transported by DOE in 55 gallon drums from the DOE-PGDP storage area to the vitrification facility. There is always at least a three day supply of the material in the storage area. Soil samples collected prior to the 30 day demonstration test will be used to determine the batch composition.

Feed Preparation System

The process of vitrifying the soil begins in the Feed Preparation Subsystem. It consists of:

(1) transportation of drums to the drum shredder for introduction to the feed preparation system, and

(2) a drying, milling, and screening operation to assure that the material is the proper size and moisture content.

At the plant, the drums are emptied into a drum shredder using a conventional fork lift truck with standard drum holding fixture.

To preclude the escape of dust particles when dumping or transporting the soil, all the conveying systems will be designed with anenclosure and operate under negative pressure. In addition, all hoppers and transfer points (dumping points) will also be enclosed and will be under negative pressure. The dust laden air from these devices will pass through a dust collector for particle removal. Solids collected in the dust collector will be transported back into the system. Discharge from the dust collector will pass through a parallel pass HEPA filter system.

The sized and dried soil is transported to a storage silo. Glass making additives are mixed with the soil. Additives (limestone and soda ash) are used to aid in glass forming, obtaining the proper glass properties, or modifying the temperature-viscosity curve. The blending system consists of storage silos and pneumatic feed system for the delivery of the soil and additives to a blend tank. Batch mixing precedes feeding into the Cyclone Melting System.

Cyclone and Melting System

The CMS™ components consist of a counter-rotating vortex (CRV) oxidizer/preheater, a cyclone melter (CM), a separator/reservoir, and a recuperator heat recovery unit.

The prepared feedstock is introduced into the CRV oxidizer/preheater through injectors located at the top of the combustor. Combustion air, which has been heated by waste heat in the recuperator, is mixed with

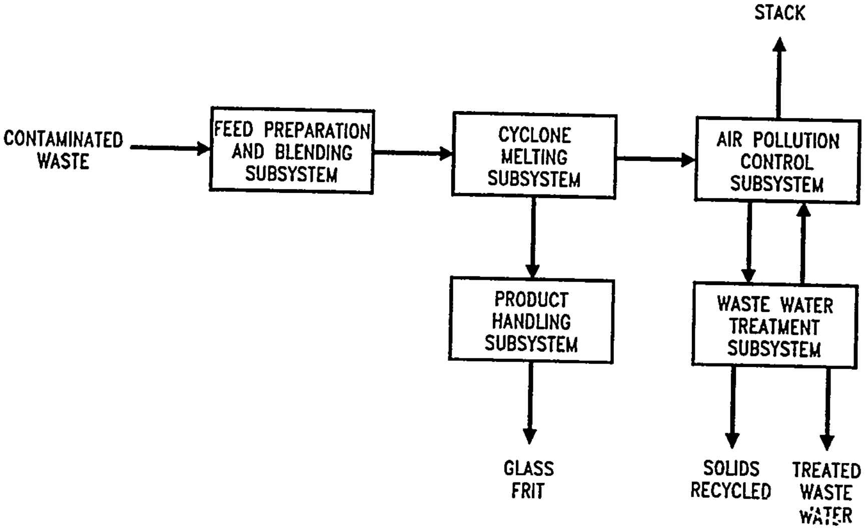

Figure 4.2-1. System Flow Diagram

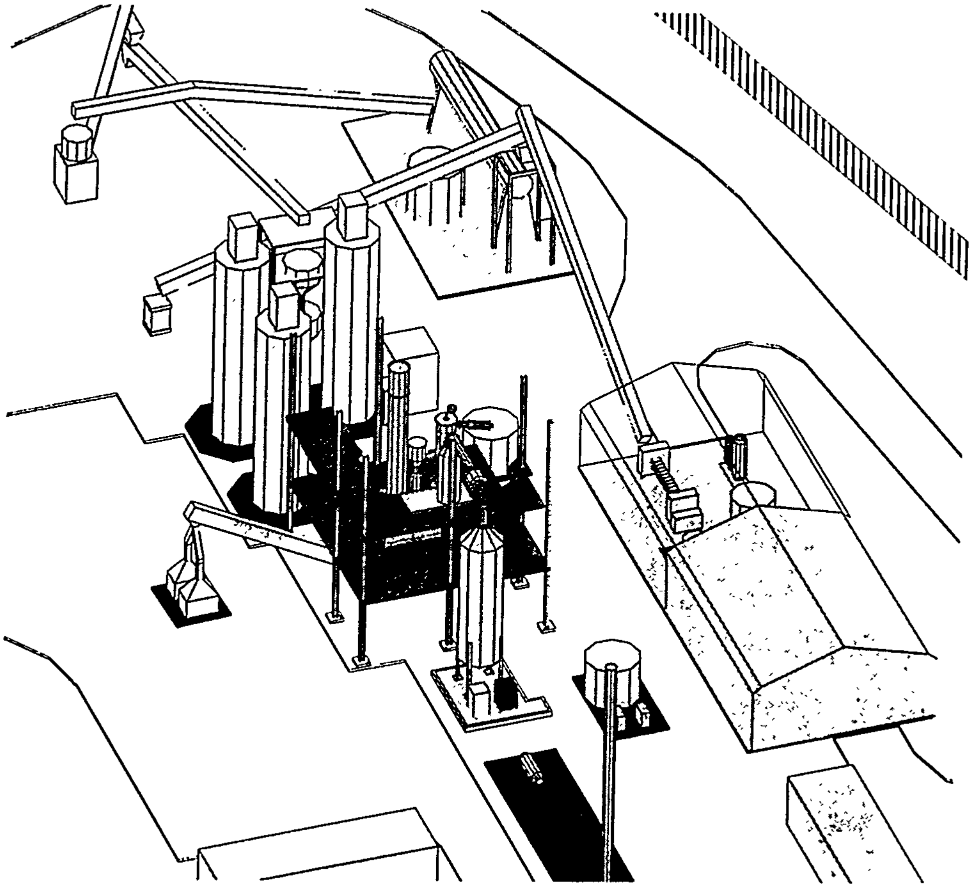

Figure 4.2-2. Isometric Drawing of Plant Arrangement

propane fuel in the inlet arms of the combustor. Auto ignition occurs as the fuelair mixture enters the high temperature region of the combustor, and the resulting combustion products raise the temperature of the feedstock as it enters. Heated feedstock flows through the CRV to the Cyclone Melter, the feedstock reacts in the liquid layer deposited on the walls of the CM, producing the glass. The radionuclides and heavy metals are permanently bonded into the glass matrix. The glass product and the exhaust gases exit the CM through a tangential exit channel and enter a glass/gas separation assembly (separator/reservoir).

The primary functions of the separator/reservoir are to separate the combustion products from the melted material and to provide an interface with a vitrified product handling system. The hot exhaust products exit through an exhaust port which is the interface for the recuperator. The recuperator utilizes the waste heat to preheat combustion air going to the CMS™. Molten glass flows out the separator/reservoir to the Vitrified Product Handling System.

Vitrified Product Handling System

The molten product from the CMS™ will be water quenched to produce a cullet approximately 1/8" in average size. The cullet will be transported by conveyor to B-25 boxes. The B-25 boxes, when full, will be moved to a pick-up area for pick-up and disposal by the DOE-Paducah.

Air Pollution Control System

The Air Pollution Control System will consist of a wet electrostatic precipitator (WESP) system for particulate collection preceded by a venturi scrubber. The scrubber will remove large particulate from the flue gas stream as well as serve the function of reducing the flue gas temperature to protect the APCS components. Other equipment in the APCS consists of an air heater, HEPA filters, induced draft fan, and an exhaust stack.

After removal of small particles in the WESP, the temperature of the off-gas is raised in an off-gas heater prior to entering the HEPA filter for removal of fine particles. Redundant HEPA filters are used to facilitate maintenance. The off-gas exits the HEPA filters and flows from the system through the exhaust stack.

Waste Water Treatment System

The Demonstration Plant also includes a waste water treatment system to remove radionuclides from the process water used in the venturi scrubber and WESP. This system consists of a clarifier, a filter press, sand filter, ion exchange unit, and various pumps and tanks.

Process water from the WESP flows through a wastewater tank, a chemical precipitation tank for chrome removal, and on to a clarifier. The solids from the clarifier, which contain some contaminants not captured in the glass, are dewatered in a filter press and are returned to the Feed Preparation System.

Radionuclides are removed by first filtering the supernate water in a sand bed. The solids are removed periodically from the sand bed by back flushing with the treated water, and the backwash is reintroduced into the clarifier. Radionuclides are removed through ion treatment. The treated effluent is stored in a holding tank for reuse as quench water within the quencher/venturi scrubber.

Instrumentation and Control System

The Instrumentation and Control System consists of the sensors, electronics, instrumentation, computers, and programmable logic controllers (PLC) to control the process in real time, gather data for analysis on system and equipment performance, and monitor process offgas. The control system will be automated to the maximum possible extent. Controllers shall be provided with the capability to be manually operated so that the combustion air blower and cooling water pumps can be operated in case of system failure. The system will be capable of being shut down in emergency situations in a controlled manner using the auxiliary power unit and structured logic. Proven industrial controls and electronics are used. Industrial PLCs enhance reliability. Multiple monitors are capable of being switched to allow individual subsystem processes to be monitored. In addition to collecting data for process and equipment evaluation, the system incorporates a Continuous Emissions Monitoring System for off-gas.

4.2 Operations Description

To demonstrate the effectiveness of the technology, 400 hours of start-up and functional testing are planned, followed by a 30-day period of nearly continuous testing.

The use of oxygen benefits the CMS™ performance by increasing the throughput capacity for a given unit size. This throughput increase is the result of several factors. Gas and solid residence times in the CRV combustor are longer due to the volume reduction of the combustion products. Additionally, a smaller percentage of the fuel thermal energy is carried out of the system by the combustion products, resulting in higher available energy for batch melting and higher process thermal efficiency. Also, the intense flame with a much higher flame temperature due to oxygen combustion (4500 °F compared to 3500 °F for air-natural gas system) allows very steep temperature gradients to be developed and enhances the heat transfer processes in the CRV combustor. This enhanced heat transfer will reduce the residence time required for batch melting as compared to the air-natural gas system. Because of this relaxed residence time requirement, unit throughput can be further increased. The increase in unit throughput also reduces the uncontrolled particulate carryover on a percent basis.

During operations, provisions will be made to enrich the combustion air to approximately 38% oxygen, thereby allowing for growth in capacity to approximately 320 drums per day.

5.0 Schedule

Vortec has completed Phases 1 and 2 of a three phase program to design construct and demonstrate the effectiveness of the CMS™ technology at remediating soils contaminated with both heavy metals and radionuclides. At the conclusion of Phase 2, the ability of the CMS™ to vitrify soils similar to the soil found at selected DOE-Paducah has been demonstrated. The vitrified product passed the TCLP as well as the PCT for leachability of a glass being used to contain radionuclides. In addition, the final design of a 36-72 TPD demonstration plant to process contaminated soil is essentially completed. Phase 3 will carry out the construction of the plant and conduct the 30 days of demonstration testing. Figure 5.0-1 represents the tentative schedule for Phase 3.

ACKNOWLEDGMENT

The authors wish to acknowledge the contributions of DOE-METC COR Mr. Cliff Carpenter. The period of performance of the contract is March 1993 to September 1997.

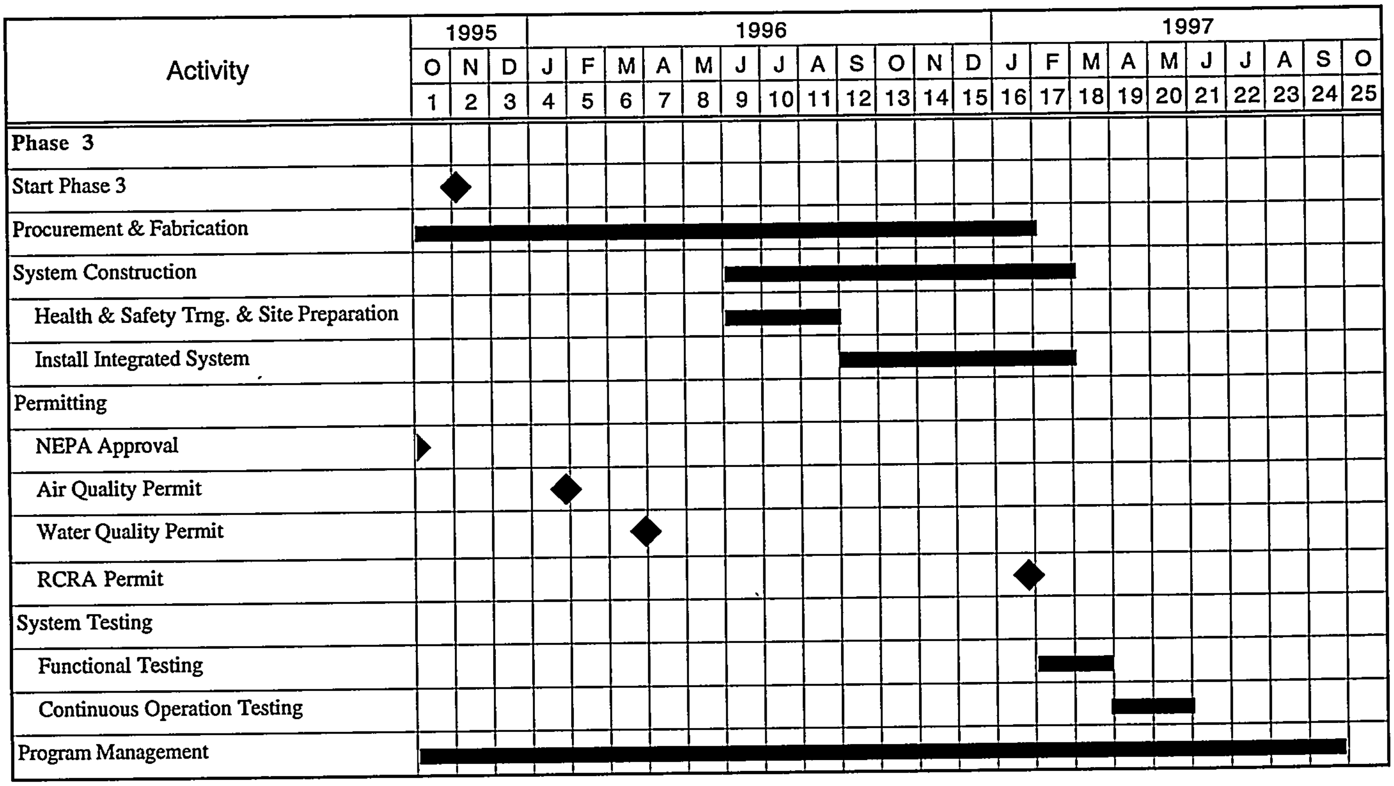

Figure 5.0-1. METC Program Master Phasing Schedule

2.3 Catalytic Extraction Processing of Contaminated Scrap Metal

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Introduction

The U.S. Department of Energy issued a Planned Research and Development Announcement (PRDA) in 1993, with the objective of identifying unique technologies which could be applied to the most hazardous waste streams at DOE sites. The combination of radioactive contamination with additional contamination by hazardous constituents such as those identified by the Resource Conservation and Recovery Act (RCRA) pose an especially challenging problem. Traditional remediation technologies are increasingly becoming less acceptable to stakeholders and regulators because of the risks they pose to public health and safety. Desirable recycling technologies were described by the DOE as: (1) easily installed, operated, and maintained; (2) exhibiting superior environmental performance; (3) protective of worker and public health and safety; (4) readily acceptable to a wide spectrum of evaluators; and (5) economically feasible. Molten Metal Technology, Inc. (MMT) was awarded a contract as a result of the PRDA initiative to demonstrate the applicability of Catalytic Extraction Processing (CEP), MMT's proprietary elemental recycling technology, to DOE's inventory of low level mixed waste. This includes DOE's inventory of radioactively- and RCRA-contaminated scrap metal and other waste forms expected to be generated by the decontamination and decommissioning (D&D) of DOE sites.

Objectives

In conjunction with the DOE, MMT designed a program to evaluate the commercial feasibility of Catalytic Extraction Processing to recycle and safely dispose of targeted portions of DOE's stockpiles of contaminated scrap metal. The contract included the following objectives:

- *Demonstrate recycling of ferrous and non-ferrous metals* -- establish that radioactive scrap metal waste can be decontaminated and converted to high-grade, ferrous and non-ferrous alloys for re-use.
- *Immobilize radionuclides* -- demonstrate that CEP will concentrate the radionuclides in a dense vitreous phase, minimize secondary waste generation, and stabilize and reduce waste volume.
- *Destroy hazardous organic constituents* -- demonstrate the conversion of hazardous organic compounds into valuable product gases, which could be used as chemical feedstocks or as an energy source.
- *Recover volatile heavy metals* -- demonstrate the capability of the gas

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Research sponsored by the U.S. Department of Energy's Morgantown Energy Technology Center, under contract DE-AC21-93MC30171 with Molten Metal Technology, Inc., 51 Sawyer Road, Waltham, MA 02154; telefax: 617-487-7870.

handling train's design to capture and recover volatile heavy metals such as lead and mercury.

- *Establish that CEP is economical for processing contaminated scrap metal from the DOE inventory* -- demonstrate that CEP is a complete processing and recycling technology, and is more cost-effective than competing technologies for processing contaminated scrap metal.

Technology Description

Catalytic Extraction Processing (CEP) is a proprietary technology that allows waste materials of a wide range of chemical and physical forms to be processed leading to decontamination and resource recovery. For processing of radioactive and mixed waste streams, CEP permits both complete destruction of hazardous contaminants and controlled partitioning of radionuclides in a single step. This leads to the potential formation of one or more decontaminated product phases (fuel gas and/or metal alloys), as well as volume reduction and concentration of radionuclides in a stable condensed phase for final disposal. A typical CEP unit consists of a feed preparation system, the Catalytic Processing Unit (CPU) or the reactor holding the molten metal, followed by a gas handling and recovery train. Upon injection into the metal bath, feed materials dissociate into their elemental constituents aided by the catalytic and solvating properties of the liquid metal. Select addition of co-reactants or manipulation of operating conditions, together with the thermodynamically controlled reaction pathways, enables synthesis of products and/or partitioning of compounds into the desired phases.

The CEP process can be conceptually divided into two stages:

- Catalytic dissociation and dissolution: The catalytic effect of the molten metal bath causes complex compounds in the feed to be dissociated into their elements, which readily dissolve in the metal bath. These soluble elements form dissolved intermediates.

- Product synthesis/partitioning: Through the addition of select co-reactants or by controlling operating conditions, the dissolved elemental intermediates can be reacted to form desired products or made to partition to the desired phases. These reactions are driven by thermodynamic forces and solution equilibria.

The solubility of carbon in iron is key to CEP's processing of organic and organo-metallic feeds. Specifically, carbon from the dissociation of organic compounds in the feeds, readily dissolves maintaining a homogeneous reducing environment in the molten metal solvent for a large number of compounds including carbon dioxide, sulfur dioxide, water and a wide range of metal oxides. The reducing strength of dissolved carbon provides CEP the flexibility to recycle organics to synthesis gas, recover metals and inorganics such as halides, sulfur and phosphorus, and separate and concentrate transuranic compounds.

As outlined above, the molten metal bath in the CEP system ensures dissociation of the feed into 'singular' elemental intermediates. The partitioning of feed components and the synthesis of products of value is dependent on manipulating the reaction pathways of the dissolved elemental intermediates. Potential reaction pathways are controlled by reaction thermodynamics. The general thermodynamic characterizations of oxidation/reduction reactions are outlined below. Specific examples of feeds simulating contaminated scrap metal relevant to typical CEP applications are discussed.

The starting point for this discussion is an overview of the Standard Gibbs Free Energy (ΔG°) changes accompanying typical reactions relevant to CEP. The reaction thermodynamics can be summarized graphically with the Gibbs Free Energy change plotted versus temperature, as shown in Figure 1. Such diagrams are used to predict the thermodynamic forces driving separation and recycling of key components in a typical CEP feed stream. Furthermore, these diagrams can be used to determine how operating conditions (temperature, pressure, bath composition) can be varied to synthesize desired products or effect partitioning of components.

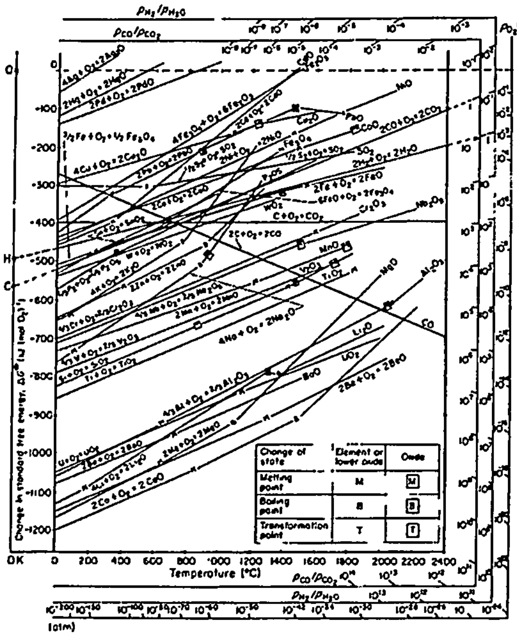

Figure 1
Gibbs Free Energies of Oxidation[i]

These correlations are for pure systems and provide only an approximation of the reaction thermodynamics guiding the process chemistry. Although beyond the scope of this paper, MMT has in-depth understanding of the thermodynamic properties of higher order, non-ideal solution reaction chemistry. This knowledge is drawn upon by MMT for refining the CEP process chemistry and reactor design to meet specific applications.

The fundamental physical chemistry principles of reaction thermodynamics and solution equilibria governing the processing of key feed components relevant to DOE waste streams will be presented. Specifically, processing of organic materials, metals, halides, and transuranics are discussed.

Organic Feed Conversion to Synthesis Gas

When an organic feed is injected into a CEP reactor, it dissociates and dissolves into elemental intermediates on contact with the molten iron solvent. Hydrogen will not dissolve in the metal solvent but will be released as a gas in the product off-gas stream. Addition of oxygen as a co-reactant will lead to the formation of synthesis gas. The production of synthesis gas is due to the effectiveness of the dissolved carbon intermediate as a reducing agent. This is best illustrated by the Gibbs Free Energy diagram (Figure 1). The oxides for which the free energy lines are above that of the formation of CO can be reduced by carbon. Therefore, when oxygen is added to a system containing carbon and iron at 2000°C, three competing reactions take place:

$$2\underline{C} + O_2(g) \rightarrow 2CO(g)$$
$$\underline{C} + O_2(g) \rightarrow CO_2(g)$$
$$2Fe(s) + O_2(g) \rightarrow 2FeO(s)$$

In this system, one can look at Figure 1 to identify that at 2000°C, the Gibbs Free Energy of reaction of carbon to carbon monoxide (-600 kJ/mol O_2) is lower than that involving carbon to carbon dioxide (-400 kJ/mol O_2) and iron to iron oxide (-250 kJ/mol O_2).

Therefore, due to thermodynamic forces CO will be formed preferentially to CO_2 and FeO when oxygen is injected into the carbon-iron melt.

A critical attribute of the CEP system is that a reducing environment is homogeneously maintained and neither carbon dioxide nor steam are formed. This is because in the presence of excess oxygen, wustite (iron oxide) formation is thermodynamically favored over carbon dioxide or steam formation. Therefore, in contrast to open flame organic feed processing systems such as coal gasification, the synthesis gas produced from CEP is of high quality with a CO/CO_2 ratio of 10,000:1 and an H_2/H_2O ratio of 2,000:1.

Metal Recovery

A significant advantage of CEP technology is its ability to recover and recycle metals from organo-metallic feeds. Consider an organo-metallic feed dissociating into its elemental constituents (e.g., C, H, M, where M is the metal) upon injection into the CEP reactor. As outlined above, both the carbon and the metal will readily dissolve in the molten metal solvent. If oxygen is added as a co-reactant, two reaction pathways are possible:

$$2\underline{C} + \underline{M} + O_2(g) \rightarrow 2CO(g) + \underline{M} \quad (A)$$
$$\underline{C} + x\,\underline{M} + y\,O_2(g) \rightarrow M_xO_{2y} + \underline{C} \quad (B)$$

Figure 1 indicates which systems will proceed with formation of carbon monoxide (reaction pathway (A)) and which systems will proceed with formation of the metal oxide (reaction pathway (B)). Specifically, carbon monoxide formation is favored in systems containing metals such as nickel, cobalt and copper whose free energy of oxidation is higher than that of carbon. These metals can be recovered as alloys in the iron solvent. Metals with a free energy of oxidation lower than that of carbon (e.g., aluminum, calcium) will form oxides and partition into the ceramic phase. Certain metals can either be recovered as metal alloys or as oxides in the ceramic phase depending on the operating conditions. For example, magnesium is reduced by carbon at elevated temperatures ($T \geq 1800°C$, $P = 1$ atm). However, at elevated pressures, magnesium oxide reduction by carbon, will not be favored.

Halogen Recovery

Under the Resource Conservation and Recovery Act (RCRA), chlorinated organic compounds of concern include the primary organic hazardous constituents (POHCs) in the waste. Carbon tetrachloride, chloroform, para-dichloro benzene, perchloroethylene, tetrachloroethane, 1,1,1-trichloroethane, trichloroethylene, methyl chloride, polyvinyl chloride (PVC), and polychlorinated biphenyls (PCBs) and all their intermediates and derivatives are among the common organic chlorinated compounds.

CEP technology can recover and recycle halogens from halogenated organic and inorganic feed streams without the formation of undesired by-products (e.g., chlorophenols, dioxins and furans). In order to demonstrate the recovery and recycling of halides in the CEP system, consider the case of a chlorinated organic feed stream. On injection into the molten metal bath, the feed will dissociate into dissolved elemental intermediates. Chlorine can be recovered and recycled as either a metal chloride or hydrogen chloride, or as chlorine gas in the absence of hydrogen. When a ceramic layer is introduced during CEP operation, thermodynamic control predicts predominant retention of chlorine in the ceramic phase under optimized conditions.

The two reaction pathways are:

- *Formation of Metal Chloride*: Calcium, in the form of calcium oxide, added to the molten iron bath will form calcium chloride. The effectiveness of calcium as a chloride scrubbing agent is demonstrated by the Gibbs Free Energy of formation of common chlorides as a function of temperature diagram, shown in Figure 2. Calcium chloride is volatile under CEP operating conditions and will distribute itself in the gas product phase where it can be cooled and separated from the other gaseous components as a condensed solid. However, by manipulating the ceramic phase composition, the metal halide can be prevented from distributing into the gaseous phase.

- *Formation of Hydrogen Chloride*: In the absence of calcium, both hydrogen chloride and ferrous chloride will be formed as the free energy of formation for both compounds is of the same order of magnitude (Figure 2). If the CEP is operated at temperatures of 1500°C, the ferrous chloride will volatilize and be recovered as a condensed solid in the gaseous stream. If the CEP is operated at lower temperatures (1000°C) ferrous chloride can be recovered as a liquid in the vitreous phase.

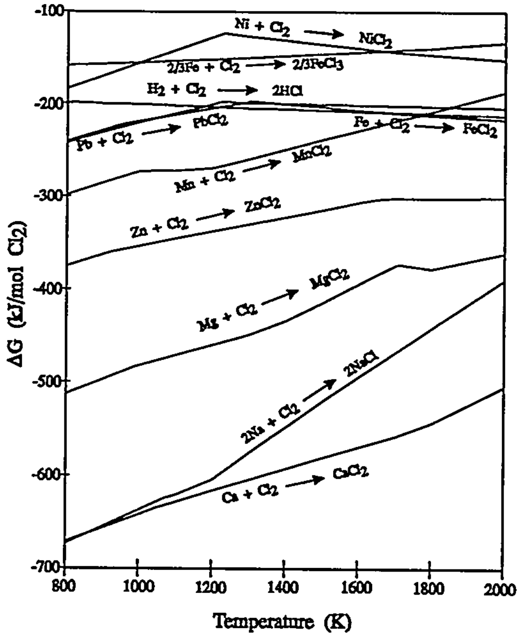

Figure 2
Chloride Free Energy Diagram[ii]

Partitioning of chlorides in a metal/gas/ceramic phase system, however, must take into consideration the effects of mass transfer issues within the ceramic phase. Even though the free energy of reaction can be treated as a true measure of the driving force for the reaction, mass transfer issues can limit the extent of the reaction such that thermodynamic equilibrium is not achieved. In the case of chloride processing with a metal/CaO-Al_2O_3-SiO_2 ceramic phase, the formation of '$CaCl_2$' in the ceramic phase is not only dependent on the activities of the constituents, but also influenced by ceramic phase viscosity, reaction-rate, and residence time of the gas in the ceramic phase. CaO-Al_2O_3-SiO_2 systems are of primary interest to MMT and have been extensively studied and optimized for potential capture of chlorides as $CaCl_2$.

The CEP flexibility in recovering halogens is a major advantage given the technical difficulties and expense associated with processing halogen-containing waste streams in typical open flame combustion processes such as incineration. Halogen containing compounds tend to suppress combustion and are often characterized as non-combustible. Therefore, incineration of these chemicals can result in incomplete combustion leading to formation of highly toxic compounds. The free radical chemistry that occurs during combustion produces radical intermediates which can react to produce dioxins and furans.

Transuranic Recovery

An important advantage of the CEP technology is that it can separate and concentrate radioactive compounds from complex contaminated feeds while also recovering and recycling the non-radioactive components of the feed. This attribute of CEP has important implications in the decontamination of radioactive sites as well as the decommissioning of nuclear weapons and the byproducts of their manufacture. This implies that radioactively-contaminated components from DOE sites can be separated, highly concentrated and immobilized in a small volume that can be safely disposed of. Furthermore, feed components such as reducible metals (e.g., nickel, chromium, manganese) can be recovered as alloys while hazardous organic components can be effectively converted to a stable final form.

Table 1 shows that the Gibbs Free Energies of reduction by carbon of transuranic oxides are positive indicating that the reactions are not thermodynamically favored. It is this oxide stability that provides a mechanism for the partitioning of these radioactive species into the ceramic phase. Data collected from the literature demonstrates the partitioning of transuranic components from the metal to the vitreous phase. Specifically, residual concentrations ranging from 0.05 ppm to 2 ppm of such radioactive components were achieved by others using diffusion of oxidizing, vitreous-forming agents to partition the radioactive components.

Table 1
Free Energies of Reduction of Transuranic Oxides

Reaction			ΔG (kJ/mol) at 2000K
$UO_2(s)+2C(s)$	→	$2CO(g)+U(l)$	167.104
$PuO(s)+C(s)$	→	$CO(g)+Pu(l)$	100.305
$PuO_2(s)+2C(s)$	→	$2CO(g)+Pu(l)$	111.377
$Pu_2O_3(s)+3C(s)$	→	$3CO(g)+2Pu(l)$	463.122
$ThO_2(s)+2C(s)$	→	$2CO(g)+Th(s)$	283.740
$ZrO_2(s)+2C(s)$	→	$2CO(g)+Zr(s)$	156.339

Experimental Feed Conversion Demonstrations

The processing and conversion of organic, organo-metallic and inorganic compounds in mixed waste streams into stable materials has been demonstrated theoretically in the discussion above. In addition to the theoretical analysis, MMT has collected extensive experimental data in bench-scale and pilot plant units and in commercial-scale metallurgical systems. The experimental data validates the theoretical predictions and confirms CEP's capabilities to generate high quality products.

The physical chemistry of CEP, specifically, the solution equilibria and the formation of a 'singular' dissolved intermediate in the reaction pathway, ensures that complete dissociation of feed streams occurs and that product synthesis and recycling can be controlled and manipulated. Therefore, the molten metal, acting as a homogeneous catalyst and solvent with high chemical inertia, renders CEP a highly flexible and robust recycling technology.

Approach

To achieve the objectives set forth in the contract , the scope of work was divided into seven tasks:

1. Design CEP system for recycling contaminated scrap metal
2. Formulate an experimental test plan
3. Experimental testing of surrogate radionuclide partitioning and bulk solids processing
4. Optimization of the vitreous phase
5. Experimental testing of RCRA wastes
6. Preparation of a conceptual design for a CEP plant for processing DOE's contaminated scrap metal
7. Preparation of a final report

Contract-sponsored technical development activities have been carried out at MMT's Recycling Research and Development Facility in Fall River, MA. The facility houses CEP reactor systems including bench- and pilot-scale units, physical models, and a commercial-scale demonstration unit. This state-of-the-art facility is fully permitted by the Commonwealth of Massachusetts for recycling demonstrations using hazardous and non-hazardous materials as CEP feeds. MMT has received recycling certifications from the Massachusetts Department of Environmental Protection for the processing of RCRA and organometallic feeds.

Bench- and pilot-scale experimental units are used to confirm theoretical modeling predictions, establish product quality, determine appropriate materials of construction, assess design parameters and confirm fundamental process chemistry. Many of these systems, including the Advanced Processing Units (APUs) which are appropriate for small commercial-scale operations, are operated round-the-clock for two to three-week experimental campaigns. Physical models are used to study fluid dynamics and transport phenomena, reactor flow patterns, unique reactor configurations, and final design parameters.

The commercial-scale prototype is used primarily for customer and regulatory demonstrations and establishment of commercial design scenarios. MMT has processed a wide range of hazardous, characteristic, and surrogate waste feeds in the commercial prototype. The commercial prototype typically operates round-the-clock during one-week demonstration campaigns, which may include several different feed materials.

The demonstration-scale CPU and gas handling unit is equipped with approximately 2,000 monitored variables and 900 control loops linked into the computerized monitoring and control modules. Numerous sampling ports throughout the gas handling train enable full characterization of the product gas quality. Summa canister sampling is performed to enable third-party analysis down to ppb levels via established EPA methods (TO-14). Mass spectrometer samples are drawn on-line and gas composition measurements are taken every twenty seconds for feed-back process control.

To meet the PRDA objectives, MMT embarked on a parallel theoretical research program and engineering design development to supplement these experimental activities. MMT's proprietary thermodynamic, kinetic, and physical transport models were applied to establish CEP design criteria for optimal processing of scrap metal contaminated with radionuclides and organic compounds. Equilibrium analysis was used to study the homogeneous reactions occurring within the individual phases. Non-equilibrium modeling was used to assess methods to enhance decontamination performance, component separation, and recovery potential using multi-phase processing.

Following theoretical analysis and computer modeling, the physical models were employed to verify predictions of hydrodynamic behavior of the system, including the specialized feeding of bulk solids into the bath. The physical models use water as a surrogate for molten iron, and various oils and emulsions were used to simulate CEP's ceramic layer. The volatility of some feed components in DOE's contaminated scrap metal was modeled using dry ice in various sizes. Hundreds of physical model experiments were performed under the contract, providing valuable data for use in designing bench-scale and pilot plant experiments.

The actual experimental progression was defined by the tasks outlined above. Experiments were designed to verify the CEP system's ability to effect partitioning of radionuclides to the targeted phases. Since inclusion of these radionuclides in the ceramic phase was predicted, concerted efforts were directed toward the optimization of the chemistry of the ceramic phase to provide a stable and durable form for disposal. In parallel, experimental processing of feeds containing RCRA contaminants was conducted to demonstrate CEP's capability to destroy hazardous organic constituents, paving the way for the delisting efforts.

The combined results of MMT's theoretical analysis, modeling studies, and experimental campaigns are currently being integrated to generate design specifications for a commercial CEP plant for processing contaminated scrap metal. The plant design package will include a bulk solid feed subsystem, particulate handling subsystem, gas handling train, a process control plan for automated monitoring and control, and a risk/safety analysis for maximizing operational safety.

Results

Targeted Partitioning to Durable, Stable Form

Previously published studies have demonstrated the ability to partition radioactive components, such as uranium and plutonium, from the metal phase into a vitreous phase via melt refining. Residual concentrations ranging from 0.05 ppm to 2 ppm were achieved using diffusion of oxidizing, vitreous-forming agents to partition the radioactive components. Successful partitioning to less than 10 nCi/g levels (approximately < 0.1 ppm) has been demonstrated for uranium- and plutonium-contaminated metals. CEP offers the potential for superior performance, as the techniques involved in melt refining are completely incorporated and enhanced in CEP technology. Specifically, CEP incorporates active radionuclide partitioning through select co-reactant additions (e.g., oxygen) and enhanced mass transfer (e.g., convection), while melt refining is based upon 'passive' diffusion-based partitioning.

MMT designed a series of bench-scale experiments using radioactive surrogate materials to demonstrate the oxidation and partitioning of radionuclides between the metal and vitreous phases and to identify and optimize the effect of operating conditions on partitioning. The bench-scale experiments were followed by experiments on pilot- and demonstration-scale systems. Hafnium was selected as a radioactive surrogate due to its similarities in thermodynamic behavior (Figure 3) and physical properties (Table 2) to uranium. Zirconium, although similar to uranium in the thermodynamic properties, was not chosen as the primary surrogate material because of its significantly lower density.

The experimental design parameters evaluated were metal system, gas environment and ceramic phase composition. Radioactive surrogate partitioning was measured using neutron activation analysis (NAA) for non-destructive trace analysis. The hafnium LDL was 0.2 ppm in iron and 2 ppm in nickel. X-ray fluorescence (XRF) was used for analysis of the vitreous phase composition. Metal samples were taken at different positions in the metal bath to ensure uniform decontamination. The contract objective was to demonstrate uniform metal decontamination above 98%. In addition, backscattered electron imaging and X-ray analysis were used to investigate the nature of the capture and stabilization of the radionuclide surrogate in the vitreous phase.

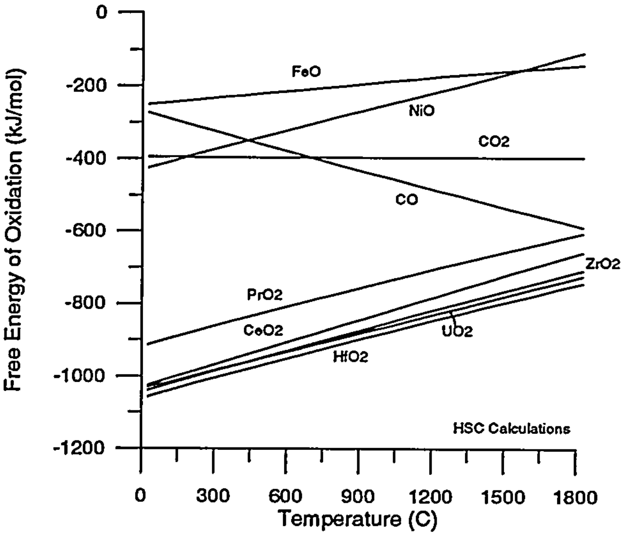

Figure 3
Identification of Radionuclide Surrogate

Table 2
Physical Properties of Uranium and Surrogates

Element	Oxide		Metal	
	m.p. (°C)	Density (g/cm³)	m.p. (°C)	Density (g/cm³)
U	2,878	10.96	1,132	19.05
Hf	2,758	9.68	2,227	13.31
Zr	2,715	5.6	1,852	6.49
Fe	1,369	5.7	1,535	7.86
Ni	1,984	6.67	1,455	8.90

Average decontamination of the metal samples were >99% and exceeded the contract's 98% objective in all experimental trials. The calculated decontamination factors were analytically limited with no radioactive surrogate detected in the metals. Table 3 summarizes the results. V1 refers to aluminosilicate vitreous compositions while V2 refers to borosilicate vitreous compositions.

Table 3
Radionuclide Partitioning

Metal	Vitreous Phase	Decontamination
Iron	V1	≥99.62%
Nickel	V1	≥99.76%
Iron	V2	≥99.08%

Samples taken at different positions in the metal bath demonstrated uniform distribution of any trace amounts of hafnium across the metal (Figure 4 and Figure 5). This indicates that radioisotope transport to and incorporation in the oxide phase is very efficient under CEP operating conditions.

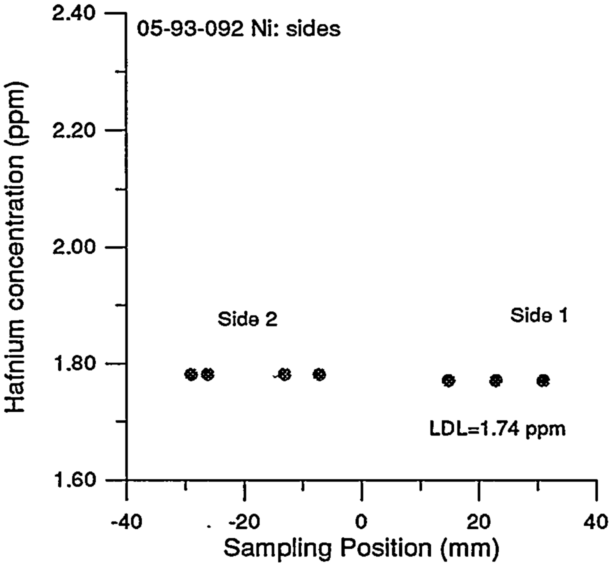

Figure 4
Sampling across Radius of Metal

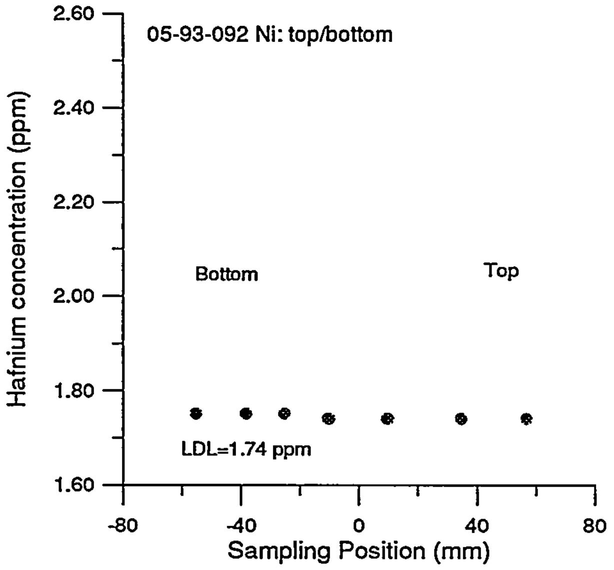

Figure 5
Sampling from Top to Bottom of Metal

Ceramic samples were analyzed using backscatter electron imaging to identify the nature of the radionuclide capture and stabilization in the ceramic phase. This work indicated that operating conditions and vitreous phase composition can affect radionuclide capture and stabilization. Figure 6 shows the backscatter image of a vitreous sample which has separated into hafnium rich and hafnium poor regions. This is in contrast to the vitreous sample shown in Figure 7, where hafnium is uniformly distributed. CEP conditions have been identified to achieve this desired vitreous radionuclide capture and stabilization mechanism.

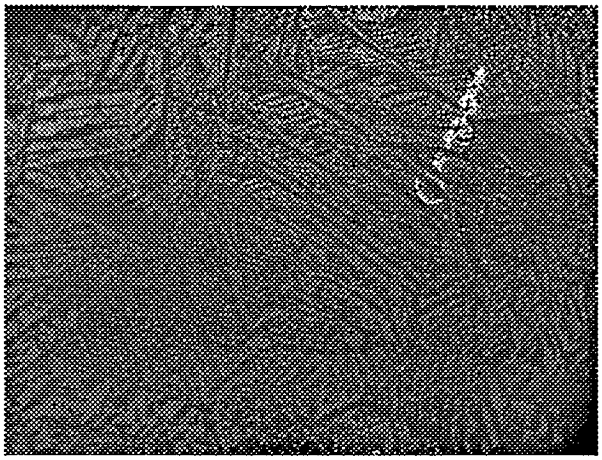

Figure 6
Backscatter Image: Phase Separation Exhibited

Figure 7
Backscatter Image:
No Phase Separation Exhibited

Processing of RCRA Contaminants

The investigation of CEP's conversion and immobilization capabilities for RCRA components of contaminated scrap metal feeds has been well proven during the experimental campaign of this contract. MMT brought to the contract a solid foundation of understanding of the behavior of toxic and halogenated feeds in CEP, and proceeded to optimize the chemistry of the system for the feeds anticipated. Efforts began on bench-scale apparatus, and subsequent prototype trials validated bench-scale results.

CEP has been demonstrated on a range of RCRA-listed wastes as well as characteristic and RCRA-like surrogate material (Table 4) including high-molecular weight aromatics, chlorinated organics, organically-bound nitrogen species (isocyanates), plastics, and organometallics. The technology's environmental performance has been demonstrated during actual waste processing to meet and surpass current and proposed regulatory standards. Specifically, DREs >99.9999% were achieved for principle organic hazardous constituents (POHCs). NO_X and SO_X were not detected in the product gases to detection limits of 3 ppm. Condensed phase non-leachable products, both ceramic and metal phases, of marketable composition were generated. Dioxins were not detected to the targeted regulatory limit of 0.1 ng 2,3,7,8 TCDD TEQ/Nm^3.

Currently the effectiveness of hazardous waste treatment technologies is regulated by the EPA by measuring the concentration of hazardous materials in the after-process wastewater and non-wastewater. All processes have to ensure that organic constituent concentrations in individual wastewater and non-wastewater streams are lower than the regulated limits set by EPA. CEP demonstrations surpass the current limits even when compared with the Best Demonstrated Available Technology (BDAT). CEP was recently approved by EPA (on October 24, 1994) as a non-combustion technical equivalent (BDAT) for eight RCRA-listed isocyanate waste codes (K027) for which incineration had previously been mandated as the commercially available BDAT technology.

The environmental performance of CEP has also been demonstrated on chlorinated waste streams containing some of the most difficult to destroy hazardous constituents. The results of demonstration-scale processing of RCRA-listed waste F024 (chlorinated aliphatics) indicated that hazardous organic constituents in the feed were not detected in ceramic, metal and scrubber water, thereby surpassing the BDAT standards for all effluent streams (Table 5). Destruction Removal Efficiency (DRE) on multiple organic hazardous constituents exceeded 99.9999%, which surpassed the current regulations mandating DREs >99.99% (Table 6). Trace constituents were not detected to the targeted regulatory limit of 0.1 ng/Nm^3 TEQ.

Table 4
Major Feeds Processed at the Demonstration Prototype

	Waste Minimization Performance			Environmental Performance		
Feed	**Product Recovery**	**% of feed Recycled**	**Residual**	**DRE**	**TCLP**	**Off-Gas**
Representative Surrogate Feeds						
Polystyrene/ Graphite	Syngas	99% to syngas	Dust Negligible	≥99.9999%	N/A	PICs < LDL[1] of 1 ppm; CO_2<1% NO, SO_2 < LDL of 100 ppm
Chlorotoluene/ Heavy Organics	Syngas Ceramic	87% to syngas 12% to ceramic	Dust Negligible	≥99.9999%	Passes TCLP	PICs < LDL of 1 ppm; CO_2 < 1% NO, SO_2 < LDL of 100 ppm
Dimethyl Acetamide/ Heavy Organics	Syngas Nitrogen	96% to syngas 3% to nitrogen	Dust Negligible	≥99.9999% (based on THC)	N/A	PICs < LDL of 1 ppm; CO_2 < 1% NO, SO_2 < LDL of 100 ppm[2] NO_X, SO_X < LDL of 1 ppm[3]
Representative Hazardous Waste Feeds						
Industrial Biosolid Waste	Syngas Nitrogen Ceramic Ferroalloy	70% to syngas 8% to nitrogen 20% to ceramic 1% to ferroalloy	Dust Negligible	≥99.9999% (based on THC)	Passes TCLP	PICs < LDL of 1 ppm CO_2 < 1% NO_X, SO_X < LDL of 3 ppm Trace <0.1 ng/Nm^3 TEQ[7]
Surplus Metal/Weapon Componentry	Syngas Ceramic Ferroalloy	25% to syngas 8% to ceramic 63% to ferroalloy	Dust Negligible	N/A	Passes TCLP	PICs < LDL of 1 ppm CO_2 < 1% NO, SO_2 < LDL of 100 ppm[2] Trace <0.1 ng/Nm^3 TEQ[7]
K019/K020[4]/ Chlorobenzene/ Fuel oil	Syngas	76% to syngas 23% to ceramic	Dust Negligible	≥99.9999%	Passes TCLP	PICs < LDL of 1 ppm CO_2 < 1% NO, SO_2 < LDL of 100 ppm[2]
K027[5]	Syngas Ferroalloy (Fe - Ni)	93% to syngas 5% to nitrogen <1% to ceramic <1% to ferroalloy	Dust Negligible	≥99.9999%	Passes TCLP	PICs < LDL of 1 ppm CO_2 < 1% NO, SO_2 < LDL of 100 ppm[2]
F024[6]/ Fuel oil/ Chlorotoluene	Syngas HCl gas	82% to syngas 13% to HCl gas <1% to ceramic	Dust Negligible	≥99.9999%	Passes TCLP	PICs < LDL of 1 ppm CO_2 < 1% NO, SO_X < LDL of 100 ppm[2] Trace <0.1 ng/Nm^3 TEQ[7]

[1] LDL = Lower Detection Limit.

[2] As measured by on-line mass spec. Lower detection limit to 1 ppm was confirmed as in footnote 3.

[3] As measured by third party analytical equipment placed on-line.

[4] EPA listed hazardous waste stream: ethylene dichloride/vinyl chloride heavy ends.

[5] EPA listed hazardous waste stream: toluene diisocyanate distillation residues.

[6] EPA listed hazardous waste stream: chlorinated aliphatic hydrocarbons.

[7] Dioxins and furans non-detectable to the targeted regulatory limit of 0.1 ng/Nm^3 TEQ.

Table 5
CEP Performance in BDAT Constituents Conversion (F024 Processing)

		Non-Wastewater Regulations			Wastewater Regulations	
Constituents	Feed (mg/l)	Ceramic[3] (mg/kg)	Metal[3] (mg/kg)	Non-Wastewater (mg/kg)	Scrubber Water (mg/l)	Wastewater (mg/l)
1,1,2-Trichloroethane	18,000	ND[1] (0.0075)[2]	ND (0.0075)	6.0	ND (0.0015)	0.054
Tetrachloroethene	9,700	ND (0.0075)	ND (0.0075)	6.0	ND (0.0015)	0.056
Chlorobenzene	12,000	ND (0.018)	ND (0.018)	6.0	ND (0.0035)	0.057
1,2-Dichloroethane	6,800	ND (0.0075)	ND (0.0075)	6.0	ND (0.0015)	0.21
1,1,2,2-Tetrachloroethane	11,000	ND (0.005)	ND (0.005)	6.0	ND (0.001)	0.057
Trichloroethene	24,000	ND (0.005)	ND (0.005)	6.0	ND (0.001)	0.054
Xylenes	610	ND (0.005)	ND (0.005)	3.0	ND (0.001)	0.32
1,1,1,2-Tetrachloroethane	1,000	ND (0.025)	ND (0.025)	6.0	ND (0.001)	0.057
Hexachlorobutadiene	3,600	ND (0.025)	ND (0.025)	5.6	ND (0.001)	0.055
Naphthalene	900	ND (0.025)	ND (0.025)	5.6	ND (0.001)	0.059

[1] ND = Not Detected.

[2] (n) where n is the lowest detection limit.

[3] In a commercial facility processing F024, the metal and ceramic would be products, not wastes subject to LDR standards. These data are provided to demonstrate CEP conversion of BDAT constituents.

Table 6
Destruction Removal Efficiency (DRE) for Listed Hazardous Constituents (F024 Processing)

Feed Constituent	Feed Concentration (mg/l)	Off-gas Concentration (ppm)	DRE
1,1,2-Trichloroethane	18,000	ND[1] (0.00037)[2]	≥99.9999%
Chlorobenzene	12,000	ND (0.00037)	≥99.9999%
1,2-Dichloroethane	6,800	ND (0.00044)	≥99.9999%
1,1,2,2-Tetrachloroethane	11,000	ND (0.00029)	≥99.9999%
Trichloroethene	24,000	ND (0.00038)	≥99.9999%

[1] ND=Not Detected

[2] (n) where n is the lowest detection limit.

CEP reaction pathways, combined with the uniform and highly reducing environment maintained over a wide range of operating conditions, preclude the formation of dioxins, furans and other undesirable by-products. Research suggests that all of the following conditions must be present to promote the formation of dioxins: excess free O_2 ($\geq 10^{-3}$ atm partial pressure), the presence of aromatic precursors, sufficient residence time in the temperature range around 300°C (572°F), and the presence of heterogeneous catalytic surfaces (e.g., particulates).[iii,iv,v,vi] These essential conditions for formation of dioxins are either inherently absent or can be minimized through appropriate design and operation of the CEP system.

Extremely low oxygen concentrations in the CEP off-gas will inhibit the formation of dioxins. Oxygen partial pressure is continuously monitored in the prototype demonstration unit and is not detected. Thermodynamic evaluation of CEP reaction scenarios shows that partial pressures of O_2 can approach 10^{-15} atm.

CEP solution chemistry can ensure that all species pass through a dissolved intermediate and preclude the formation of aromatic precursors. Table 4 shows that Destruction and Removal Efficiencies (DREs) are greater than 99.9999% for chlorinated plastics (PVC and polystyrene) and chlorinated solvents.

Monitoring Devices and Control Methods

The development of advanced models and techniques to monitor and control key process parameters is crucial to robust, automated CEP operation. System automation reduces the likelihood of operators' exposure to radioactivity, and allows better control of process variables and a more immediate response to system and feed variabilities. Areas of keen development interest include bath temperature control, bath composition control, containment system monitoring, and bath level, among others. Monitoring these properties through conventional means utilizes intrusive measurements. Carbon concentration and bath temperature can be directly measured using invasive sampling techniques, but these may interrupt the flow of operations and delay opportunities for feedback control. In addition, manual sampling and other conventional methods will increase risks to worker and public health and safety. Therefore, MMT embarked on a technology development and implementation program under this contract to improve the reliability, operability, and degree of automation in CEP systems. The program encompass improving existing and developing new on-line sensing and instrumentation devices, developing sophisticated control algorithms, and the selection and integration of inherently safe equipment and robust operating procedures.

Through the use of its dynamic and steady-state simulation models, detailed theoretical models describing local phenomena, and operational experience garnered from its demonstration-scale unit, MMT identified the key process and design parameters impacting CEP performance. A program was then set up to develop, test, and implement non-intrusive, continuous monitoring of these key parameters, which include bath temperature, carbon concentration, gas composition, ceramic composition, refractory thickness, bath level, bath pressure, dust loading, and headspace temperature. A market search was conducted to identify the most accurate, reliable, and cost-effective sensing devices. Sensing devices currently used in industry have been adapted to CEP systems, and new and novel sensing techniques have been implemented and are undergoing testing on pilot plant and demonstration-scale units to verify their accuracy and reliability. To complement these activities, MMT has developed commercial-quality on-line control modules, providing an easy-to-use operator interface.

Temperature Control. The primary objective of the bath temperature control strategy is to keep the bath molten and above a minimum acceptable temperature set primarily by the liquidus temperature of the bath. Secondary objectives are to minimize changes in the bath temperature and to operate at the optimum temperature to minimize containment wear. The bath temperature is controlled by adjusting the power setting of the bath induction coil power supply. Closed-loop temperature control was successfully demonstrated with the controlled variable being the bath temperature and the controller output the power supply setting. The bath temperature is estimated using embedded thermocouples readings and the CET (Compensated Embedded Thermocouple) temperature model. The CET model is a proprietary software module developed in-house that can predict the bath temperature by solving the governing transient heat diffusion equations. The commercial-quality module has capabilities for digital filtering, sensor diagnosis, automatic rejection of erroneous thermocouple inputs, self-tuning, and automatic refractory thickness correction.

Other techniques for measuring the bath temperature are currently undergoing testing. Specifically, the feasibility of using a two-section (sapphire/quartz) infrared (IR) lightpipe sensor has been recently proven on the demonstration-scale unit. A lightpipe is a solid rod of high IR transmission characteristics, and good chemical stability at high temperatures. One end of the lightpipe rod is in direct contact with the melt, thus providing a path for IR radiation from the molten metal through the lightpipe. The IR emissions are collected by a re-imaging lens or a terminated fiber optic cable at the other end of the rod and carried via a fiber-optic cable to a dual-wavelength pyrometer. During testing, the lightpipe sensor showed immediate response to temperature perturbations and demonstrated stability against thermal cycling.

Delisting

CEP has been demonstrated on a range of RCRA-listed wastes as well as characteristic and RCRA-like surrogate material (Table 4) including high-molecular weight aromatics, chlorinated organics, organically-bound nitrogen species, plastics, and organometallics. CEP's ability to provide complete destruction of the hazardous constituents while maintaining high regulatory integrity has been proven. Due to the unique set of regulations governing the processing of RCRA-listed materials, the disposal of a stable final form containing radionuclides but not exhibiting any RCRA characteristics would benefit from delisting. Delisting would provide increased flexibility in terms of disposal destination and cost.

A delisting petition has been prepared based on experimental data generated from pilot plant and demonstration-scale testing. The delisting petition is an upfront application for a generic delisting of the ceramic phase generated from CEP systems. Five representative mixed waste streams have been selected for delisting:

- Organic sludges: e.g.,ORR MWIR 3090 (F001-F002 solvents, F006 electroplating waste); INEL ID-EGG-158:3
- Inorganic sludges: e.g., ORR MWIR 3004 (F006 electroplating waste); INEL ID-EGG-102:7 (toxic organic and metals with mercury)
- Soils and sediments: e.g., ORR MWIR 3151 (Pb, Hg); INEL ID-EGG-141:990
- Combustible debris: e.g., ORR MWIR 2028 (F003); INEL ID-EGG-114:337
- Scrap metals: e.g., INEL ID-EGG-132:20

The experimental results presented in support of the delisting petition included feed materials characterization, ceramic elemental composition, ceramic hazardous characteristics evaluation, toxicity characteristics leachability procedures (TCLP) testing, and hazardous organic constituents concentration in the ceramic phase.

For a waste to be successfully delisted, the petition must demonstrate that the waste:

- does not meet the criteria for which it was listed
- does not exhibit any hazardous waste characteristics
- does not exhibit any additional factors including additional constituents which may cause the waste to be a hazardous waste

In preparing the delisting petition, MMT has followed the requirements for delisting petitions set forth in 40 CFR § 260.20 and § 260.22, as well as in relevant EPA guidance (such as the guidance manual for petitions to delist hazardous waste (EPA/530-R-93-007, March 1993)). These regulations and guidance specify the administrative information, waste characterization and waste management history, process information, waste sampling and analysis information, and ground water monitoring information that must be included in the delisting petition.

Containment Design

Containment system development has been the focus of an integrated and rigorous effort under this contract. An integrated program aimed at maximizing refractory life and optimizing containment handling for ease of operation has been developed. The program's focus has been on improving the containment system design, optimizing refractory materials selection, and developing a theoretical and empirical understanding of the mechanisms controlling lining degradation.

Refractory wear is the most important physical phenomenon faced in the CEP containment development program, as gradual wear processes will ultimately determine the longevity of CEP units. Therefore, the factors involved in refractory lining wear have been systematically addressed experimentally and theoretically. Containment system design has been found to be the most important component for combating refractory lining deterioration. Much of the initial development work in CEP containment has been focused on improving design methodology. Refractory reliability has shown a steady increase over the course of the contract. This has been due to a combination of improvements to refractory design, layout, heat-up practice, shell design and temperature control. Critical design improvements are being made on an ongoing basis. The results of these important developments are summarized below:

- Developed and implemented crucible-based modular containment system capable of simplified change-out to provide extended life and enable rapid and economical turnaround/replacement of worn components

- Developed application-specific crucible materials compatible with different feed types

- Developed alternative bath chemistry process control strategies based on the type of crucible used

- Thorough investigation of the interaction of refractory systems with halogenated species led to dramatic improvements in containment system longevity

- Developed methods, such as composite coatings and other chemical techniques, to enhance the lifetime of the containment system

- Under development is a model to predict refractory life based on key operating parameters

- Demonstrated compatibility of refractory materials with MMT-developed ceramic phase systems (developed for stabilizing radionuclides)

Future Activities

Bulk Solid Feed Addition

One important area of development under the PRDA contract is the design of a robust system to process a wide variety of bulk solid materials. The capability to process bulk solids such as whole drums or similar-sized materials will increase the range of DOE feeds that can be processed by CEP and eliminate the need for size-reduction feed pre-processing systems. CEP reactor configuration systems are currently under development at MMT for top addition of bulk solid feed, including multi-zone reactors, lances, and other advanced reactor designs. Common to these concepts is the fact that they include multiple zones; a dissolution/volatilization zone and a gas polishing zone. Bulk solids are charged to the dissolution zone (receiving chamber) with a sufficient residence time to ensure complete volatilization of organic constituents and those species which will be volatile under CEP operating regimes. Subsequently, the volatilized gas is directed to the polishing zone (zone 2) to ensure that all portions of the feed proceed through the dissolved elemental intermediate state.

Earlier work on the physical models has established the feasibility of lance feed injection and multi-zone processing. The effect of reactor geometry on feed dispersion, gas bubble dynamics, and residence time were investigated. During hot metal experimentation, materials and methods of construction to prolong lance and baffle life have been identified. Bench-, pilot-, and commercial-scale experiments have shown that top addition of bulk solids using non-optimized CEP reactor configurations is very promising. Optimization of reactor geometry is planned to verify initial modeling and experimental results.

Prior to additional hot metal testing, the physical water models will be utilized where better visualization, flexibility, and controllability of the experiments can be obtained. The first set of experiments will investigate the dependence of the maximum solids loading on conditions in the dissolution/volatilization zone and on the properties of the bulk feed (feed type and size). This characterization of zone 1 will allow the requirements for the second polishing zone to be determined for evaluation. The physical models use water as a surrogate for the molten iron. Feed surrogate materials will also be used to model the behavior of different types of feed materials.

Characterization of the polishing zone will proceed by initially conducting experiments on the water models. The objective will be to determine the parameters affecting the efficiency of the polishing zone. Advanced reactor concepts will be applied to evaluate different methods to enhance the rate of mass and heat transfer which will lead to increasing the efficiency of processing the volatilized gas generated in the first zone. This knowledge will then be applied in the design of the detailed configuration of the polishing chamber.

The experimental data generated by the aforementioned work will provide a valuable basis for designing and conducting hot metal experiments on the pilot plant units. The primary objective will be to characterize the dissolution/volatilization zone for different feed types and sizes, and evaluate the performance of the different configurations developed for the polishing zone. This will include determination of the off-gas composition and generation rate as well as dust loading and composition as a function of key process variables. The data collected will characterize the volatilized gas generated in zone 1 and will determine the requirements for zone 2. In addition, the operability range of the system for materials for which dissolution and volatilization is mass or heat transfer-controlled will be determined experimentally. Specifically, the maximum solids loading will be determined as a function of feed rate, mass of metal in contact with the feed, the dissolution time, and reactor geometry. The maximum power supply capacity which will provide the energy required to dissolve and volatilize the feed materials will be determined. Although limitations exist on the maximum feedable materials (by reactor physical dimensions) and on the rate of volatilization (by the maximum capacity of the GHT), methods will be developed to scale-up the results to commercial-scale systems.

The experimental test program will proceed to assess the performance of the different reactor configurations developed for the polishing zone. Baffle reactors, series reactors, and other advanced multi-zone reactor concepts will be evaluated for their ability to effect a complete conversion of the bulk feed materials while maximizing the lifetime of the equipment. The most appropriate method for bulk solids processing will be determined based on environmental performance criteria and cost-effectiveness.

Acknowledgment

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2.4 Waste Inspection Tomography (WIT)

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Introduction

Waste Inspection Tomography (WIT) provides mobile semi-trailer mounted nondestructive examination (NDE) and assay (NDA) for nuclear waste drum characterization. WIT uses various computed tomography (CT) methods for both NDE and NDA of nuclear waste drums. Low level waste (LLW), transuranic (TRU), and mixed radioactive waste can be inspected and characterized without opening the drums.

With externally transmitted x-ray NDE techniques, WIT has the ability to identify high density waste materials like heavy metals, define drum contents in two- and three-dimensional space, quantify free liquid volumes through density and x-ray attenuation coefficient discrimination, and measure drum wall thickness. With waste emitting gamma-ray NDA techniques, WIT can locate gamma emitting radioactive sources in two- and three-dimensional space, identify gamma emitting isotopic species, identify the external activity levels of emitting gamma-ray sources, correct for waste matrix attenuation, provide internal activity approximations, and provide the data needed for waste classification as LLW or TRU. Objectives

The mobile feature of WIT allows inspection technologies to be brought to the nuclear waste drum storage site without the need to relocate drums for safe, rapid, and cost-effective characterization of regulated nuclear waste. The combination of these WIT characterization modalities provides the inspector with an unprecedented ability to non-invasively characterize the regulated contents of waste drums as large as 110 gallons, weighing up to 1,600 pounds. Any objects that fit within these size and weight restrictions can also be inspected on WIT, such as smaller waste bags and drums that are five and thirty-five gallons.

Background Information

X-ray imaging is an established method for waste container inspection. The technique generally used is real-time radiography (RTR) using a 420 kV radiation source, in which a TV camera is coupled to a two-dimensional, light-producing x-ray detector, such as an image intensifier or a scintillation screen. The camera output provides a TV image that is viewed on a monitor during x-ray exposure which, as an example, can see the motion of a moving liquid surface. However, RTR systems have several

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disadvantages. Area x-ray detectors typically suffer from blooming artifacts that limit spatial resolution. RTR systems have limited contrast discrimination with a true dynamic range of usually less than 14-bits (16,384 gray levels in the image), meaning that contrast in a single exposure is limited. RTR typically does not provide quantitative information, since the data is not in digital form. It limits geometric depth perception because of super-positioning, and it lacks quantitative information including two- and three-dimensional spatial measurements and density measurements. The combination of 420 kV source and a restricted detector dynamic range limits RTR penetrating and discrimination capability for inspecting the denser waste containers including cement-solidified drums, glass, and sludges, which make up nearly half of DOE's inventory of nuclear waste drums.

Emerging technologies for nondestructive evaluation (NDE) of low level, transuranic, and mixed nuclear waste like WIT include high-energy 2 MeV x-ray computed tomography (CT) and digital radiography (DR). In its conventional approach, WIT CT/DR imaging uses a curved linear array of solid-state x-ray detectors. The array is composed of individual, closely aligned detection channels. The channels are separated by thin septa that minimize crosstalk and blooming, while offering superior spatial and contrast resolution with high image quality, compared to real-time approaches. These detectors have enough dynamic range to provide contrast sensitivity of 18-bits (up to 262,144 gray levels). The greater the dynamic range and penetrating radiation, the greater the advantage in examining denser waste forms mentioned above. Thus, the WIT approach can image nearly all dense DOE waste streams with even faster throughput for the lighter waste forms. This cannot be said for the more commonly used RTR x-ray imaging systems.

For WIT CT, x-ray projection data is collected from a thin plane of the object, using a linear detector array on the arc of a curve while the object rotates within a thin x-ray fan beam with spiral-like motion capability. This technique is called third-generation CT. The data are mathematically combined to form a cross-sectional image of the thin, irradiated plane or slice. Slices can be stacked to form a volume rendering of drum content. In WIT DR, the drum is moved vertically in front of the linear detector array while projection data is collected one line at a time. These techniques measure the x-ray attenuation of the waste matrix and drum. The lines are then displayed as a two-dimensional, freeze-frame projection image (like a baggage inspection x-ray) for DR.

Active CT on WIT is similar to the conventional x-ray CT techniques. The difference is that a radioisotopic source and single channel high purity germanium detector are used with a first-generation CT approach. A CT data results in the absolute determination of the attenuation of the drum and its contents.

Two emission imaging techniques are employed on WIT for characterizing materials in waste containers. The first of these is gamma emission tomography, commonly called single-photon emission computed tomography (SPECT). Rather than measuring gamma-rays from an external radiation source, SPECT measures the gamma-ray emission inherent in the radioactive waste emitting from within the drum. In this case, emission from actual nuclear waste within a container can provide three-dimensional volume or slice data of the radioactive sources within the container. SPECT uses large area sodium iodide crystals with a two-dimensional array of photomultiplier tube (PMT) detectors for rapid localization of gamma-ray emissions in two-dimensional space and 3-D with SPECT. These area cameras are called Anger cameras.

The second WIT emission technique uses

an energy sensitive single channel high-purity germanium detector for gamma-ray nuclear spectroscopy. This technique, called nondestructive assay (NDA), can directly identify the emitting isotopic species and the external radioactivity.

Project Description

Bio-Imaging Research, Inc. (BIR), from Lincolnshire, Illinois has completed Phase I over 23 months involving the design, fabrication, factory testing, and evaluation of WIT. BIR has designed the trailer and multiple inspection techniques including DR, CT, SPECT, and area gamma-ray imaging. BIR has also developed the WIT operational software, the computer hardware, and the gantry mechanical systems. Lawrence Livermore National Laboratory (LLNL), as a subcontractor to BIR under a Work-for-Others agreement with BIR, has developed the A&PCT scanning technique and is participating in WIT evaluation.

Phase II, beginning in August, 1995, is a twelve month program for the integration and DOE site demonstration of WIT. Early BIR efforts prior to WIT involved investigating the feasibility of using CT to characterize nuclear waste between 1990 and 1993 under Small Business Innovative Research (SBIR) grants from DOE. WIT is a Program Research and Development Award (PRDA) contract number DE-AC21-93MC30173. The WIT PRDA is funded by Environmental Management's (EM) Office of Technology Development (OTD/EM-50) for the United States Department of Energy (DOE). DOE has in excess of 1,000,000 nuclear waste drums currently stored at nearly 50 sites within the United States that need to be characterized over the next few years before permanent disposition.

The three WIT detection technologies include a linear array of solid-state high-energy x-ray detectors, two area gamma-cameras, and a single high purity germanium detector. The throughput of WIT inspection is dependent on the physics of the drum being inspected. As an example, the lighter the drum weight, the faster the WIT NDE inspection, and the higher the drum radioactivity, the faster the NDA inspection. The reverse is also true where heavier drums with little radioactivity will require longer inspection times. Ideally, WIT is designed to inspect four drums per hour. Throughput extremes could yield inspection times of between one and seven drums per hour for each technology employed.

The WIT linear detector array is curved and consists of 896 individual channels of cadmium tungstate (CdW4) crystals mounted on individual photodiodes with thin septa between channels to eliminate crosstalk, blooming, and in-plane scatter. These detectors have an 18-bit (262,144 gray levels) dynamic range for analog-to-digital conversion. The wide dynamic range is used in WIT to image the variety of material densities and geometries found in DOE waste streams, including combustibles, glass, cement, sludges, and metals that may be present in the same drum. The linear array utilizes a 2 MeV high-energy accelerator as an externally transmitted radiation source using a thin fan beam output with a measured flux of 70 rads per minute at one meter. This high-energy source is needed to penetrate the denser and thicker DOE waste forms like glass logs, sludge, and cemented drums, while allowing for an optimum inspection throughput. The use of energies above 2 MeV is not practical for WIT because of WIT's mobile requirements resulting in weight restrictions for radiation shielding limiting close operator interaction.

The WIT linear detector array and 2 MeV source provide for single pass digital radiography (DR), which yields a freeze frame projection x-ray image (e.g., like a chest x-ray) using an imaging technique similar to x-ray baggage inspection systems with the drum elevating through a stationary horizontal fan beam of x-rays. A single DR drum view at 2 MeV with wide dynamic range can be acquired in less than 30 seconds with only one x-ray technique. Unlike RTR, both high and low density objects can be examined with WIT DR using a single DR image due to the wide dynamic range.

The linear array and 2 MeV source also provide for transmission computed tomography (CT or TCT) with cross-sectional (two-dimensional) slice and volume (three-dimensional) imaging of drum content based on density distribution. WIT TCT slice thickness ranges from 2 mm to 10 mm through the drum. CT images are acquired by simple drum rotation for data collection and drum elevation for slice location. CT is used for waste drum content identification with a spatial resolution of nearly 2 mm and a density sensitivity of nearly 1%. Typical CT reconstructions and DR images have formats of 256 by 256, 512 by 512, or 1024 x 1024 pixels. Individual CT slice scan times for data collection can be as short as eight seconds for a slice or eight seconds for two slices with an optional dual array for imaging lightweight (S.G.>,1) combustible waste. WIT has slower CT scan times (as long as 20 minutes per CT slice) for dense solid glass logs (S.G. 2.7). As a bench mark, 100 slices for a low-density combustible waste filled 55 gallon drum could be acquired and volume rendered in less than ten minutes (with a dual array), whereas a glass log with a density of equal dimensions could take nearly eight hours for 100 slices. A cemented drum (S.G. 2.1) could require a scan time of less than 0.5 hours. Drum wall thickness resolution using WIT TCT is between 0.125 and 0.25 mm. Volume measurements (i.e., for free liquids) of pixels with similar density indications has nearly cubic centimeter resolution with WIT TCT.

WIT's two large area (14" x 17") detectors are each single crystals of sodium iodide in what are typically called Anger cameras. These detectors provide for rapid gamma emission drum area imaging and single photon emission computed tomography (SPECT or ECT) for slice and three-dimensional volume localization of gamma-ray emissions from a drum. Both crystals have a combined total of 110 photomultiplier tubes (PMT) for two dimensional gamma ray emission localization within the waste drum with a spatial resolution of nearly 25 mm over a cross-sectional area of the drum. An emission slice through the drum may also have a thickness of nearly 25 mm. Typical gamma emission projection images and SPECT slice reconstructions have image formats of 32 x 32, 64 by 64, and 128 by 128 pixels. Typical inspection times for a single drum can range from seven minutes to one hour, depending on the emitted gamma ray activity. The higher the activity, the faster the scan time and the reverse is also true.

The single channel, high efficiency, (>100% of that of sodium iodide), high purity, germanium detector (HPGE) uses a 50 mm on a side square collimator and an active source of 1.25 millicuries of Holmium(^{166}Ho). The HPGE detector and ^{166}Ho provide for active and passive computed tomography (A&PCT) with excellent energy sensitivity (of less than 2 kV) for nuclear spectroscopy. A&PCT on WIT are each first-generation CT techniques, each using a single channel energy sensitive HPGe detector. WIT can detect energies between 10 kV and 1.33 MeV using an 8,000 discrete channel multi-channel analyzer (MCA). This detector and source are used to determine an absolute linear attenuation coefficient of the waste drum and matrix. PCT identifies and localizes the radioactivity. The ACT data is used to correct the passive CT data for attenuation caused by the waste matrix and drum itself. The combination of both techniques results in a more accurate nondestructive assay of the waste drum. LLNL has developed the A&PCT techniques. This single detector has a spatial resolution of nearly 50 millimeters over the area with slice thicknesses of nearly 50 mm. Typical A&PCT reconstructions have formats of 14 by 14, 28 by 28, and 42 by 42 pixels. Determination of internal radioactivity approximations for waste drums have thus far shown errors between 5 and 10%. Scan time for a single slice can range from one minute to one hour, depending on the level of radioactivity.

Results

The images included in this paper were taken on-board the trailer and demonstrate CT, SPECT, and A&PCT capabilities. Fused data displays define the waste matrix surrounding gamma emitting sources within the container. WIT can quantify the internal radioactivity which has been corrected for attenuation caused by the waste matrix solely with external measurements without opening the drum.

These Phase I results shown below illustrate the visualization capabilities possible with fusion of transmission and emission images from computed tomography and projection radiographic imaging. The WIT system's combination of inspection technologies lets inspectors choose an appropriate level of characterization for each site or waste container.

Future Activities

The WIT program is managed by the government from the DOE Morgantown Energy Technology Center (METC) in Morgantown, West Virginia. During Phase II, site demonstrations of WIT are planned at LLNL in Livermore, California, Westinghouse Savannah River Company (WSRC) in Aiken, South Carolina, and at the Idaho National Engineering Laboratories (INEL) in Idaho Falls, Idaho. These demonstrations will involve characterizing real waste to validate the system's ability to identify regulated contents and to verify system throughput.

BIR plans to commercialize WIT with the successful conclusion of the PRDA contract. BIR plans to offer drum scanning/characterization services to DOE and other sites requiring mobile capabilities.

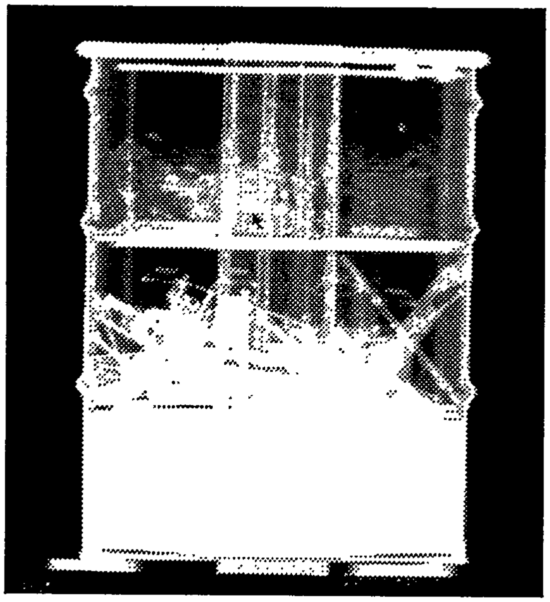


Figure 1 is a 2 MeV transmission DR projection image of a 55 gallon drum phantom. Clothing in the top layer simulates low density combustible waste. Cans of liquid, metal rods and plastic in the mid-section simulate higher density waste. Cement in the bottom layer simulates solidified high-density waste. The 133Ba isotope bottle is visible in the upper left plexiglass tube.

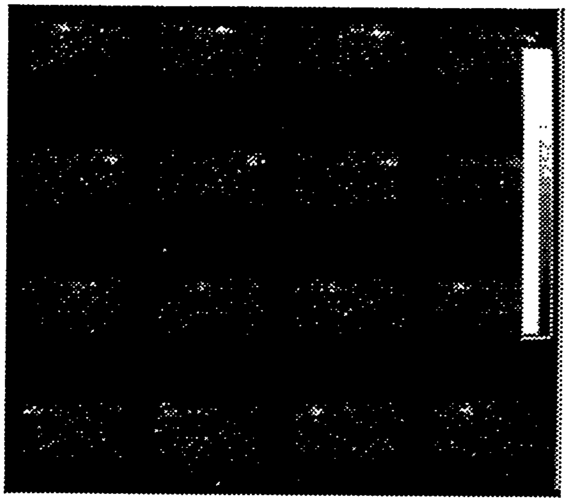


Figure 2 is a series of 16 gamma emission projection images from an Anger camera taken every 22.5 degrees of rotation from a centered radioactive test source. The emission source was 133Ba with a 360 kV peak and 250 microcuries of activity. The window used was 30% and the integration time was one minute for a single view. Shorter data collection times are possible.

Figure 3 is a DR fused image of an emission source in the upper section of the drum The dots are the locations of actual photon counts, which are denser near the emitting source.

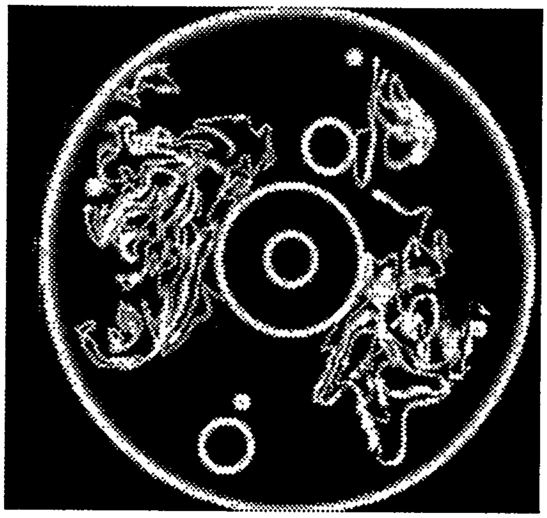


Figure 4 is a 2 MeV transmission CT slice 10 mm thick through the middle level of the drum, with a 1024 x 1024 reconstruction. The Plexiglas tubes are evident, as are acrylic rods and clothing. The steel ball bearing in an aerosol paint can is evident at 1 o'clock.

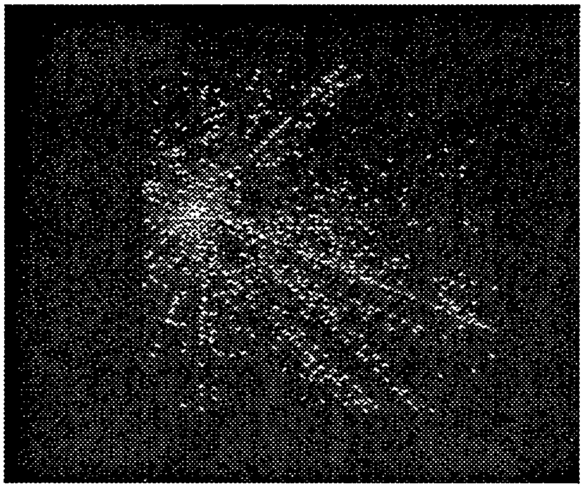


Figure 5 is a SPECT reconstruction 10 mm thick with a 256 x 256 image showing the 133Ba radioactive emission location.

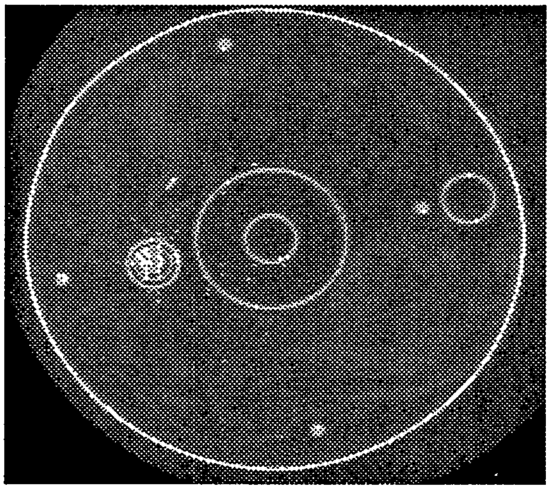


Figure 6 is the data fusion result of combining the transmission CT data with the emission SPECT image.

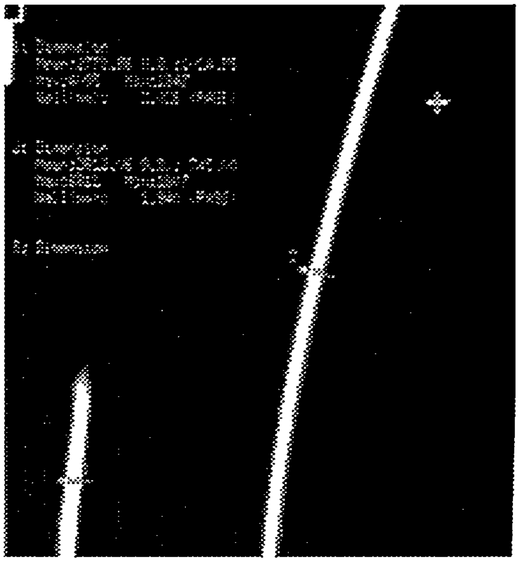


Figure 7 shows a zoomed CT image of a section of a drum wall. Positions of line cursors indicate measurement positions of varied phantom drum wall thicknesses.

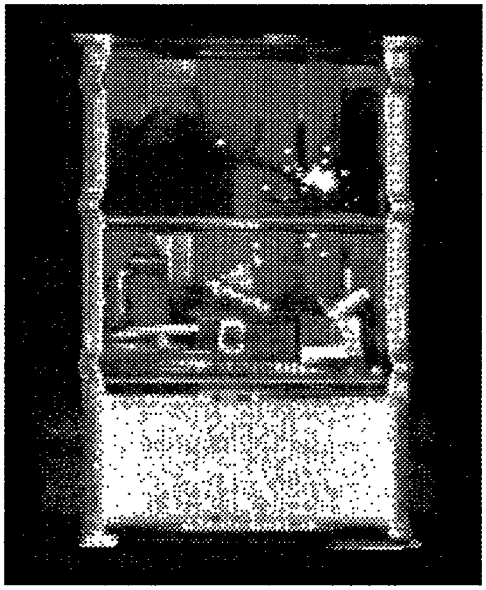
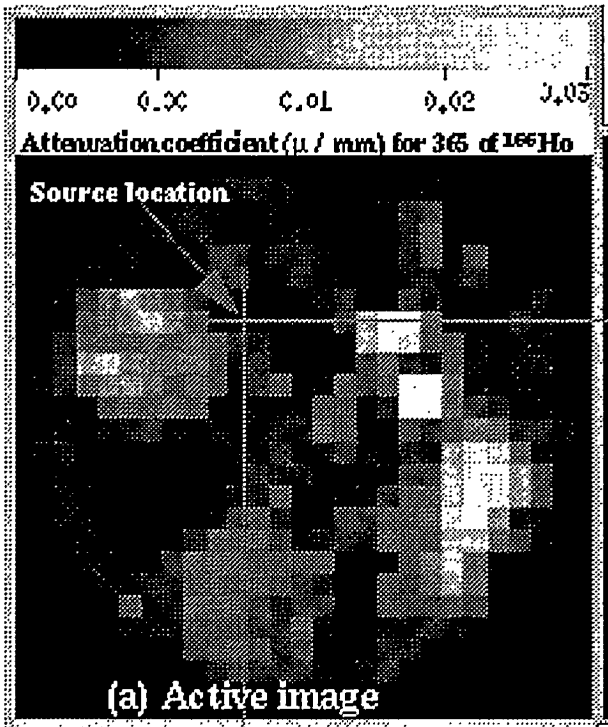
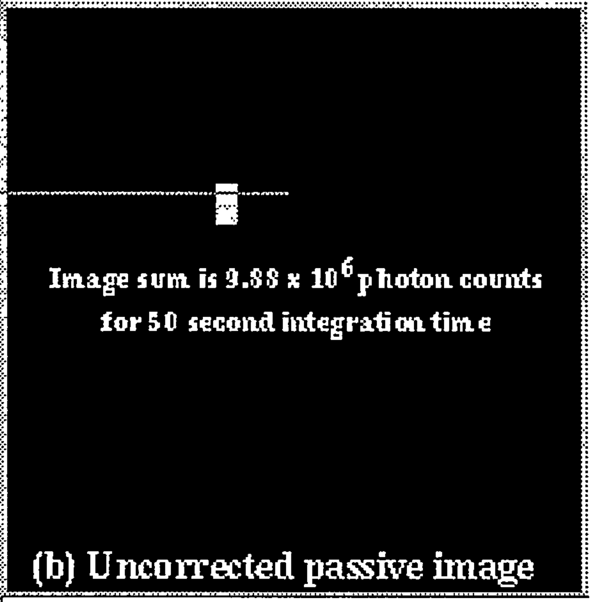


Figure 8 is a volume rendered image with fused SPECT reconstruction. The 133Ba source is clearly visible as a white spot in the upper layer. A transparent bottle with free liquid laying on its side is also evident to the left of the 133Ba source. Aerosol can and liquid level is rendered transparent in the center section of the phantom.

A & PCT Determination of Measured Internal Radioactivity from a Mock 55 Gallon Drum





Midsection of TRU drum with - 65% average attenuation for 356 peak of 133 Ba emission source.

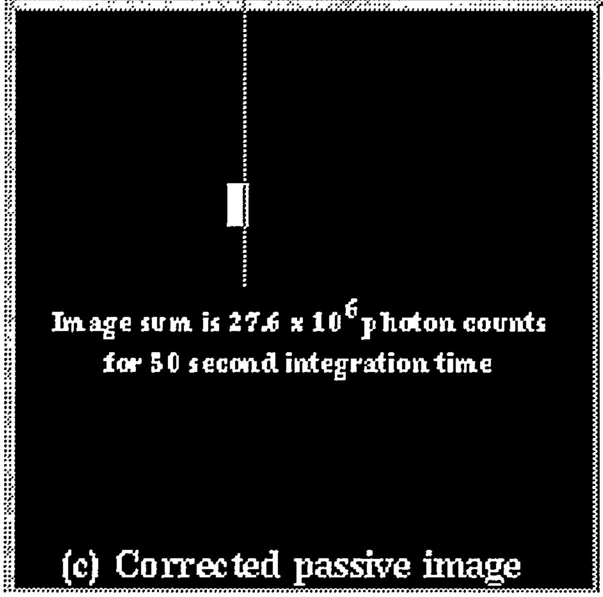


TABLE 1.

	Curies	Photon Counts	Error
Ideal	68.2μCi	26.19 E+6	
Measured	71.9μCi	27.6 E+6	+5.4%

Figure 9 shows the results of an LLNL experiment using active and passive CT to measure internal radioactivity from a mock 55 gallon waste drum.

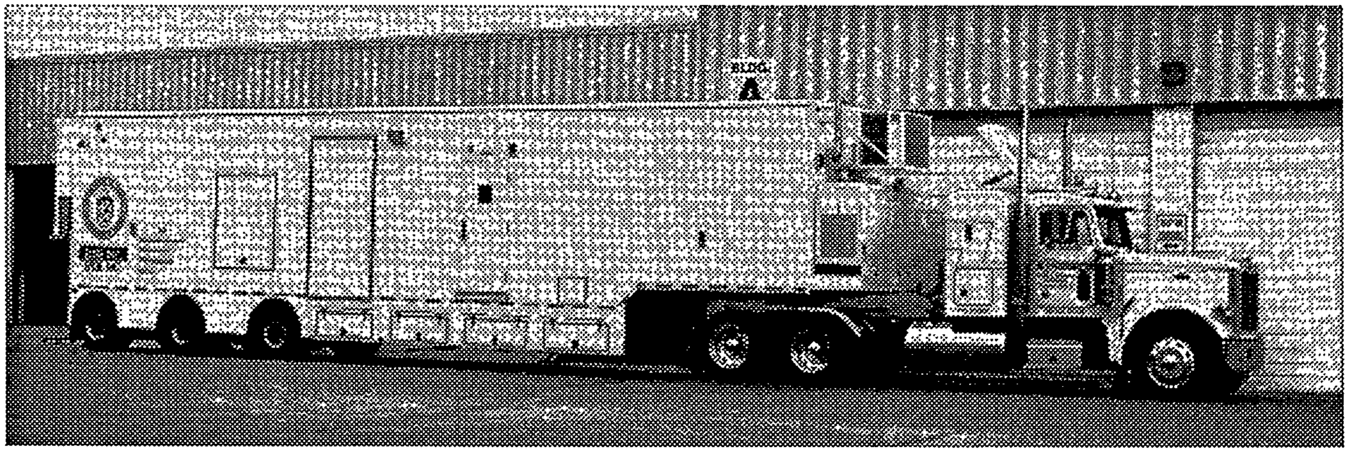


Figure 10 shows trailer which houses the WIT inspection system.

Figure 11 shows the WIT control room and operator's workstation.

Acknowledgments

The primary Bio-Imaging Research (BIR) personnel specifically responsible for the development of WIT are; Dick Bernardi, program manager, Dave Entwistle, project software engineering manager, Ken Hill, mechanical design manager, Jerry Swinford, chief project engineer, Jim Benbow, former project engineer, and Kyung Han, Ph.D., project imaging scientist. Mike Silver, Ph.D.,V.P. of R&D from BIR was instrumental in providing reconstruction software used on WIT. We acknowledge John F. Moore, Ph.D., President of BIR, for his support of the WIT program.

We also acknowledge Calumet Coach as supplier of the WIT semi-trailer, Varian as supplier of the 2 MeV linear accelerator, EG&G Ortek as supplier of the HPGE detector and nuclear spectroscopy software, Technicare as manufacturer of the gamma cameras , and BIR as manufacturer of the high energy x-ray detectors, data acquisition systems, computer systems, mechanical gantry, control systems, operating software, the system design, integration, and testing of WIT . BIR also developed the DR, CT, area gamma imaging, and the SPECT inspection modalities for WIT as well as the volume rendering, data fusion, and quantitative analysis features of WIT including all cine presentations.

We further acknowledge our subcontractor, the nondestructive testing (NDT) group from Lawrence Livermore National Laboratory (LLNL) including Harry Martz, Ph.D., Pat Roberson, Allen Friensehner, Dan Decman, Ph.D., Dave Camp, Ph.D., and Steve Stuckmeyer. Under a "work for others agreement" with BIR, LLNL provided WIT with A&PCT and nuclear spectroscopy information.

We'd like to also acknowledge our government contract program management from DOE METC in Morgantown, WV for their valued contributions including contract officer representatives Curtis Nakaishi, C. Eddie Christy and P. Steven Cooke and contract specialist Mary Spatafore.

2.5 A Robotic End Effector for Inspection of Storage Tanks

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Introduction[1]

The structural integrity of waste storage tanks is of primary importance to the DOE, and is one aspect of the High-Level Waste Tank Remediation focus area. Cracks and/or corrosion damage in the inner tank walls can lead to the release of dangerous substances into the environment. The detection and sizing of corrosion and cracking in steel tank walls through remote non destructive evaluation (NDE) is the primary focus of this work.

Problem

Damaged sites may represent a small proportion of the total inner wall area so, rather than NDE the whole area, more selective and efficient methods of inspection are being developed. The defects of interest can be very small, which implies very accurate deployment of an NDE inspection device. Typically the large manipulators required to access the underground storage tanks (such as the Light Duty Utility Arm) are not designed for such high accuracy.

Solution

Oceaneering Space Systems (OSS) has overcome these conflicting requirements by combining an advanced electro-magnetic NDE technique (Alternating Current Field Measurement, or ACFM) with a compact vision and lighting system that allows an operator to distinguish possible sites of corrosion on the tank walls before approaching the tank wall for a detailed ACFM inspection (figure 1). Uniquely, ACFM is capable of detecting and sizing defects without prior calibration. The ACFM sensor coils are mounted in an array which is driven up and down the end effector's scanning frame, inspecting a 3"x6" area in one pass. ACFM does

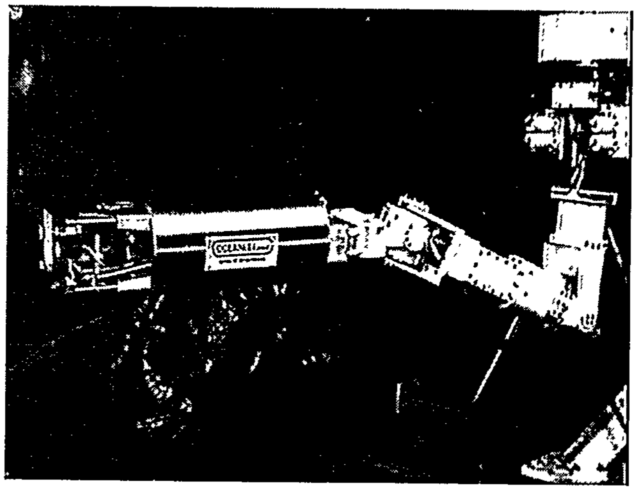

Figure 1. The Robotic Tank Inspection End Effector

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[1] Research sponsored by the U.S. Department of Energy's Morgantown Energy Technology Center, under contract DE-AR21-93MC30363 with Oceaneering Space Systems, 16665 Space Center Blvd, Houston, Texas, 77058; telefax: (713) 488 2027

not require direct electrical contact with the tank wall to detect and size defects, nor is it particularly sensitive to standoff or orientation. In addition ACFM does not require coupling fluid, a drawback of ultrasonic inspection techniques in which the couplant becomes secondary waste.

The ACFM scanning frame is mounted compliantly to allow the manipulator to push the scanning frame up against the tank wall. This guarantees that the array is sufficiently aligned with the wall (figure 2). The video camera is mounted on the centerline of the end effector, thus providing both a primary view for the operator to drive the manipulator around the tank and a direct view of the inspected area within the scanning frame. The operator is provided with both live video and the results of the ACFM inspection on the same monitor. The ACFM appraisal of the wall is presented as a two dimensional false color plot indicating defect position and size.

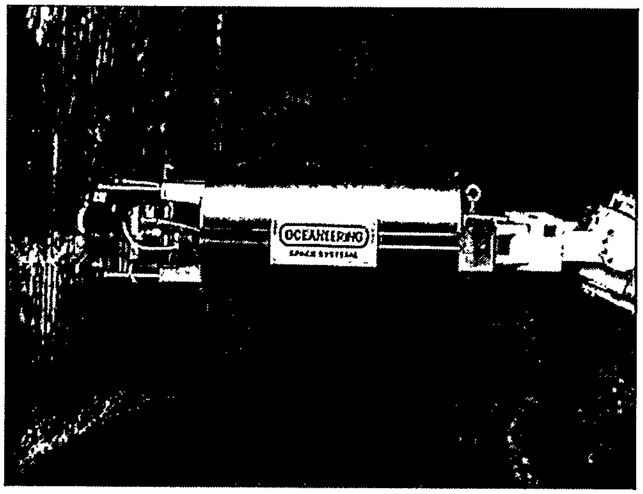

Figure 2. The RTIEE performing an ACFM inspection of the wall

The original Robotic Tank Inspection End Effector (or RTIEE) prototype was designed to contact the wall before inspecting the pitting corrosion. This system was successfully demonstrated at the Idaho National Engineering Laboratory in March 1995, and its performance is described here. Additional work, funded by Morgantown Energy Technology Center (METC), will increase the utility of the inspection system by providing the capability for the end effector to inspect the tank wall while moving past it (a 'fly-by' inspection), and by enabling the system to detect and size cracks as well as corrosion pits on the wall. Following acceptance of this second prototype, DOE will procure a radiation hardened and environmentally protected version of the RTIEE system for use by the Light Duty Utility Arm.

Applications and Benefits

- Monitoring the structural integrity of waste storage tank walls is possible without exposing personnel to a hazardous environment.

- A single compact end-effector combines a tank viewing and lighting system with an NDE technology that can detect and size corrosion pitting, and will soon include detection and sizing of cracking.

- The end-effector is designed to be deployed by the LDUA, or similar manipulator, through small diameter (11") riser access holes in the tank ceiling.

- The end-effector can inspect any conducting material.

- ACFM has several advantages over more traditional NDE techniques: unlike X-Ray ACFM is a benign electromagnetic technique; unlike ultrasonics ACFM does not produce secondary waste in the form of couplant (fluid or gel). In addition ACFM can both detect and size defects, is much less sensitive to standoff and misorientation than eddy current techniques, and works through most coatings and layers.

Alternating Current Field Measurement (ACFM)

ACFM[2] is an electromagnetic NDE technique that has been specifically developed to overcome the shortcomings of eddy current techniques. ACFM combines the ability of the Alternating Current Potential Drop (ACPD) technique to size defects without prior calibration with the ability of eddy current to work without electrical contact. This is achieved by inducing a uniform AC field in the target material and measuring the magnetic fields above the specimen. The uniform current flow is modeled analytically, which makes the field response predictable and allows the characterization and sizing of defects without the use of artificial defect samples to calibrate the system. The use of the uniform field encourages the production of arrays of coils to cover large areas simultaneously even when relatively small defects are targets.

Description of the RTIEE

Figure 1 shows the current RTIEE prototype deployed by an OSS manipulator arm. A mockup waste tank wall can be seen to the left. Five subsystems make up the RTIEE: camera and lights, end effector body, ACFM sensor, software and operator interface, and data transfer system.

The Camera and Lights Subsystem

A CCD camera is mounted on the centerline of the end effector body. A motor driven lens is attached to the camera and a wide angle lens adaptor is attached to the lens. The lens adaptor increases the field of view of the camera from 41.9 degrees to 56.6 degrees and allows the operator to view the back of the scanning frame (useful when seeking contact with the tank wall), and to view a distant object (useful for identifying inspection sites on the tank wall), without any of the scanning frame obstructing the view. Figure 3 shows the camera view when the end effector is pushed up against a wall; the mechanical status indicators can be seen in the four corners of the image.

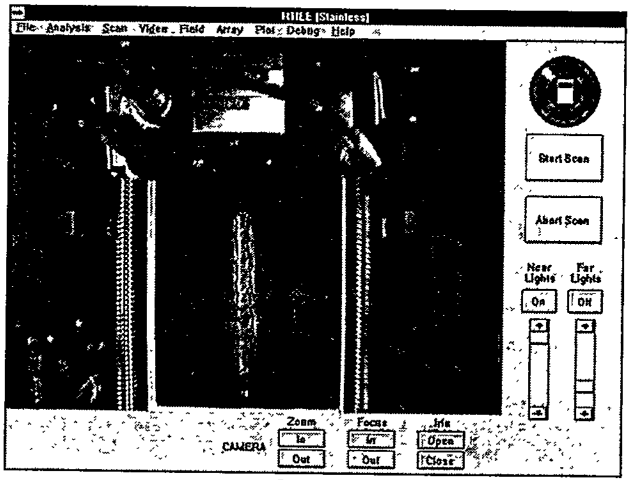

Figure 3. The end effector landed over a weld

The lighting system consists of two sets of lamp arrangements. The near set, mounted on either side of the camera, illuminate the rear of the scanner frame. The far set, mounted on the front face of the scanner frame, illuminate distant objects. The near lamps each have a power rating of 15 watts and the far lamps are rated at 35 watts. The motor driven iris, zoom, and focus functions, and the lighting intensity of all the lamps, are controlled by the operator from the operator interface screen. Local control logic circuitry mounted in the end effector body supports both the camera and lighting functions and the stepper motor used for scanning.

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[2] ACFM has been developed by Technical Software Consultants Limited.

The End Effector Body Subsystem

The body consists of three main areas: the main body (cylindrical shell), the compression rods, and the scanner frame. The main body attaches to a manipulator through a base plate. Four struts protruding from this plate carry the cylindrical shell, and provide a load path into the base. The camera, lens, and sensor electronics are mounted within the main body. Four stainless steel compression rods are mounted in aluminum-bronze housings and are sprung into the housings. These compression rods allow movement of the scanner frame relative to the main body and absorb impact loads as the end effector is pushed up against a tank wall. Each compression rod also has a very stiff spring that prevents the rod from bottoming out. The scanner frame carries the ACFM sensor array. The array is driven up and down the frame on two acme screw drive shafts. The drive shafts are connected to a stepper motor by a timing belt. Two limit switches are attached to the frame and report the two extreme positions of the sensor array carriage. Four status indicators are mounted in the frame. When the protruding push rod of the status indicator is depressed by contact with a wall, a colored strip moves within the indicator window that faces the end effector camera. Finally, the two far lights are mounted facing out in the scanner frame. In figure 4, the far lights, camera lens, and status indicators are clearly seen. The sensor array carriage can be seen in the lower half of the scanner frame and the stepper motor is under the cover protruding from the upper rear of the scanner frame.

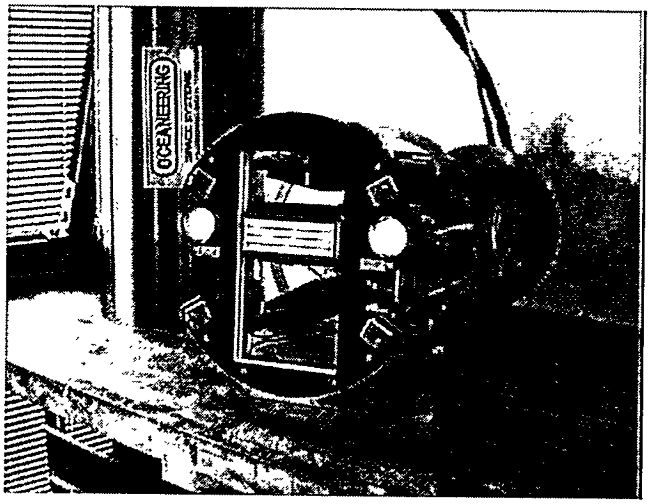

Figure 4. The end effector scanner frame (note the protective body shell has been removed)

The Software and Operator Interface Subsystem

The control and interface software is written in C and developed in the Visual C environment. The software architecture consists of specifically written modules in a robust and hierarchical manner. The operator controls the end effector using a customized graphical user interface (GUI). Figure 2 shows the operator interface screen. Surrounding the video image are the mouse activated menus and functions. The movement of the sensor array can be initiated and halted by clicking the buttons marked Start Scan and Abort. The Save Scan button and the pull-down menus allow the video image to be grabbed and saved to hard disk along with the scan data. These menus and additional buttons also control the analysis of the ACFM inspection data and the recording of the visual data to VHS. Several data display and analysis options are available from the menus, including the detection and sizing algorithms. The near and far light intensity is controlled by the sliders and buttons in the lower right corner of the screen. Controls for the camera lens zoom, focus, and iris are arranged below the inspection windows.

The ACFM Sensor Subsystem

The sensor system consists of two main components: the sensor array, and the data acquisition electronics. The sensor array is made up of 96 Bz coils and 32 Bx coils arranged in four rows (each coil has an outside diameter of .070"). Behind each row is a printed circuit board (pcb) populated with multiplexing and signal processing chips and components. Each pcb is connected via a transition pcb to an umbilical. The umbilical connector and all the boards are mounted inside a carbon steel box which is located in the sensor array carriage. The umbilical connects the array to the data acquisition electronics; the data acquisition electronics are connected to the control computer via a serial line.

The ACFM array inspects metal plate by inducing AC fields onto the plate and sensing disturbances in the field with sensor coils that are scanned over the plate. By using a theoretical model of the field and a look-up table, the system can then size any pits it detects. The prototype end effector features two types of solenoids to provide two directions of input field (X and Z) and two orientations of sensor coil (Bx and Bz) to be sensitive to different components of the magnetic field above the plate. The Z field solenoid is wound around the array carriage. The two X field solenoids are mounted on either side of the scanning window.

The variation in the magnetic field caused by pits is very small compared to the general field, so a normalization procedure is used to highlight the pit signal. Experimentation indicates that normalization of the whole scan area is the most effective method. This implies that a complete scan of the 6" by 3" window, on a known good plate, is stored in the software and then subtracted from all scans of similar material. This method has the additional advantage of removing all repeatable field variations caused by the presence of end effector structure around the array. A plate suitable for normalization would be made of the same grade of steel as the tank wall and be approximately the same thickness, it would also be of a large enough area to prevent the plate edges disturbing the field produced by the end effector (currently this is about 14"x14").

The Data Transfer Subsystem

Power and data is transferred to and from the end effector through a single umbilical. This umbilical terminates at a 37 pin connector at the rear of the end effector. The umbilical is 50' long to simulate transmission of power and data over a representative length. Sensor array data is collected by the data acquisition unit and passed to the interpretation software in the computer via the serial line. Provision has been made in the prototype system to drive all supply and signal voltages through 250' of cable if required.

Test Results

The current RTIEE prototype is designed to detect and size corrosion pitting in carbon and stainless steels. Other defects such as cracks and other linear features at welds (lack of penetration, lack of fusion, etc.) will be detectable by the new system currently under development.

As discussed previously, ACFM relies on the imposition of a uniform current field onto the surface of the material to be inspected. This allows the disturbance of the field, caused by a defect, to be modeled analytically; this is strictly true only on materials with a small skin depth.

The smaller the pit that can be confidently characterized, the greater the accuracy of the assessment of corrosion damage on the inner tank wall. There are two aspects to this capability: the first is the ability of the operator or computer to detect the presence of a defect in the magnetic field; the second is the accuracy of the algorithm that sizes the defect based on the amplitude and position of the disturbances in the various fields. The RTIEE design incorporates automatic detection and location algorithms to reduce the level of expertise required by an operator, and to provide the operator with a real time graphical depiction of the condition of the tank wall.

Figure 5 shows the disturbance of the uniform X field as it flows around and into a pit. These experimental results, for carbon steel, closely match the response predicted by the analytical model, and illustrate the peak/trough pair seen by a Bz sensor and the tipped depression seen by the Bx sensor. The ZBz disturbance is not modeled but is such a strong response that the RTIEE uses it to detect pits. The ZBz disturbance is the classic eddy current response consisting of a peak centered over the defect. The RTIEE detection and sizing algorithms use different combinations of field and sensor data depending on the material being inspected. When working on carbon steel, for instance, the ZBz data is first examined for peaks. Having detected a peak, the X field/sensor combinations are examined in the same area to locate their dominant features. If the right features are located then confidence in the presence of a defect increases and the amplitude and position of the dominant features are used to size the defect. The relationship between the disturbance of a field and the diameter and depth of the pit is determined by running the theoretical model for all combinations of pit diameter-to-depth and creating a look up table of results. The model therefore greatly reduces the cost of developing an inspection tool by reducing the number of test plates required to calibrate the system.

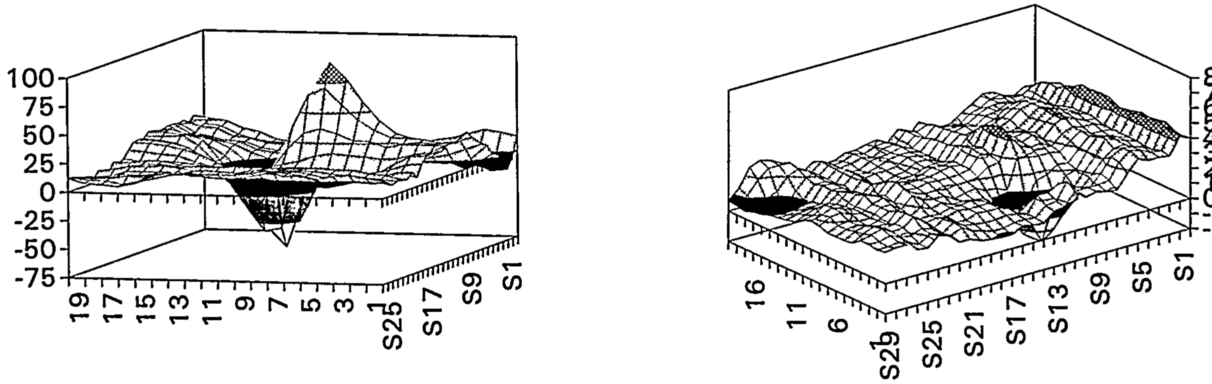

Figure 5. Magnetic fields recorded above a pit in the presence of a uniform field, XBz (left) and XBx (right)

This robust approach to defect detection and sizing is only limited by the practicalities of field deployment. Essentially, small pits produce small field disturbances which decay close to the surface of the steel. Therefore if small defects are to be detected, the sensor head must scan, as a general rule, not more than one pit diameter away from the material surface. The type of steel in the tank wall can also effect results. Stainless steel represents a slightly different challenge than carbon steel because at the inspection field frequency used by the RTIEE (5KHz) the material has a significant skin depth (about .22" as opposed to .005" in carbon steel). This skin depth has the advantage of allowing the RTIEE to detect subsurface features. Results for the inspection of carbon steel by the RTIEE are reported in *Hughes and Gittleman, 1995*. A more thorough discussion of detecting and sizing corrosion pits in stainless and carbon steel can be found in the project's draft final report. The following summarizes the results for the inspection of stainless steel.

Inspection of Stainless Steel

Inspection of stainless steel uses the XBx and XBz fields to detect and size defects. Figure 6 illustrates the screens the operator sees following an inspection of a grid of pits with the XBx and XBz fields illustrated. The depressions in the XBx field, over the larger pits, are clearly seen. This image can be compared to the XBz field for the same inspection that has peak trough pairs over the pits. Figure 7 illustrates the detection and sizing display. The image consists of the detection and sizing display next to a video image of the inspected area. The location, diameter, and depth of each pit detected is listed in the small window. The pit location is recorded in Cartesian co-ordinates relative to the end-effector coordinate system; the upper left corner of the scanning frame corresponds to the origin (0,0). As these images attest, the location algorithm accurately locates the pits, typically within one pit radius of the actual position. Two diameters are shown (horizontal and vertical) to allow the representation of oval features, and the depth measurement is followed by a confidence level that is based on the number of field/sensor combinations that sense the defect. If the detection threshold is set low enough to find even the shallowest .030" pits some spurious indications occur. If the threshold is set slightly higher, only genuine pits are detected and sized.

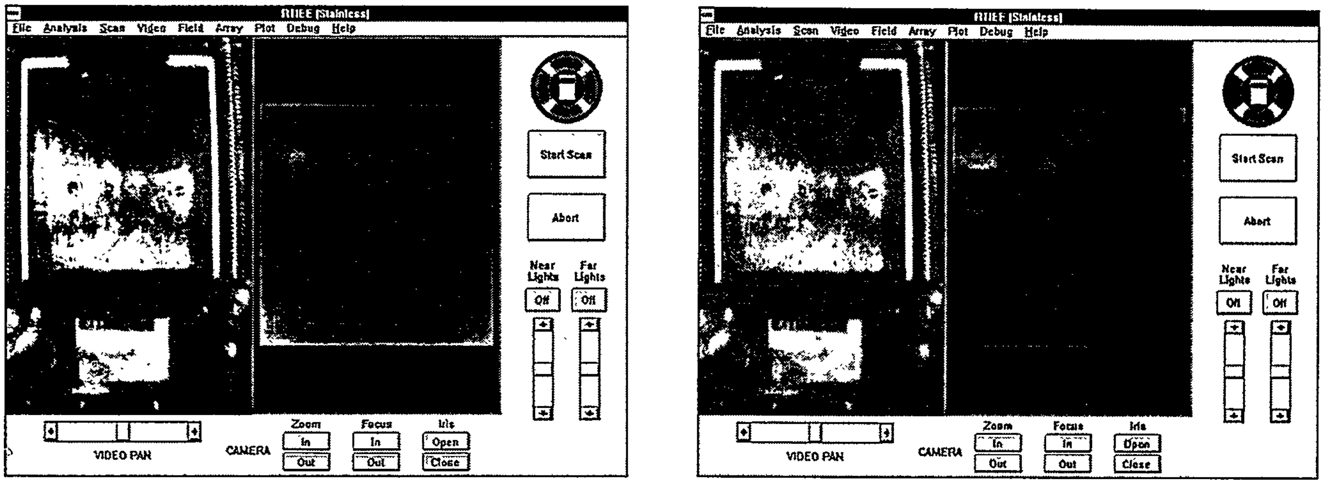

Figure 6. RTIEE results showing the magnetic fields over a grid of pits, XBx (left) and XBz (right)

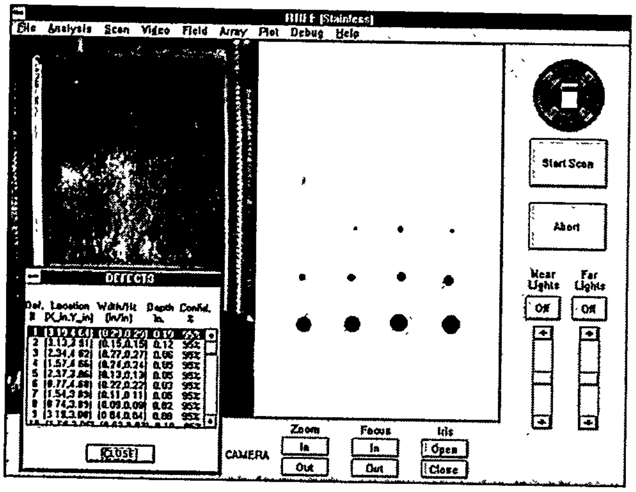

Figure 7. The pit detection and sizing display

The stainless steel pit diameter estimation algorithms perform well, as shown in figure 8. Even though the depth of the pits inspected varied from 12% to 100% of the wall thickness the diameters were typically estimated to within 20% of the actual. If all pits were considered hemispherical then the depth of the features would be estimated as accurately. The depth sizing algorithms, however, attempt to estimate pit depth from data in the inspections fields, as described above. On the larger pits the algorithms are reasonably accurate until the depth of the pit becomes excessive (holes), at which point the current flow at the bottom of the feature is minimal or non existent. Figure 8 illustrates this point with results from ¼" pits and holes. The deeper holes are underestimated but as the holes become shallower the depth estimate becomes more accurate. A real corrosion pit has a depth generally no greater than its diameter and therefore is accurately estimated by these algorithms.

Future Activities

The RTIEE provides a single unit solution for visual and NDE tank inspections performed by robotic systems. It successfully incorporates a vision subsystem, lighting subsystem, fine positioning subsystem, ACFM NDE technology, and data analysis software into one integrated system. The RTIEE system has been successfully demonstrated in a series of manipulator tests using a laboratory waste tank wall mockup. During laboratory tests, operators

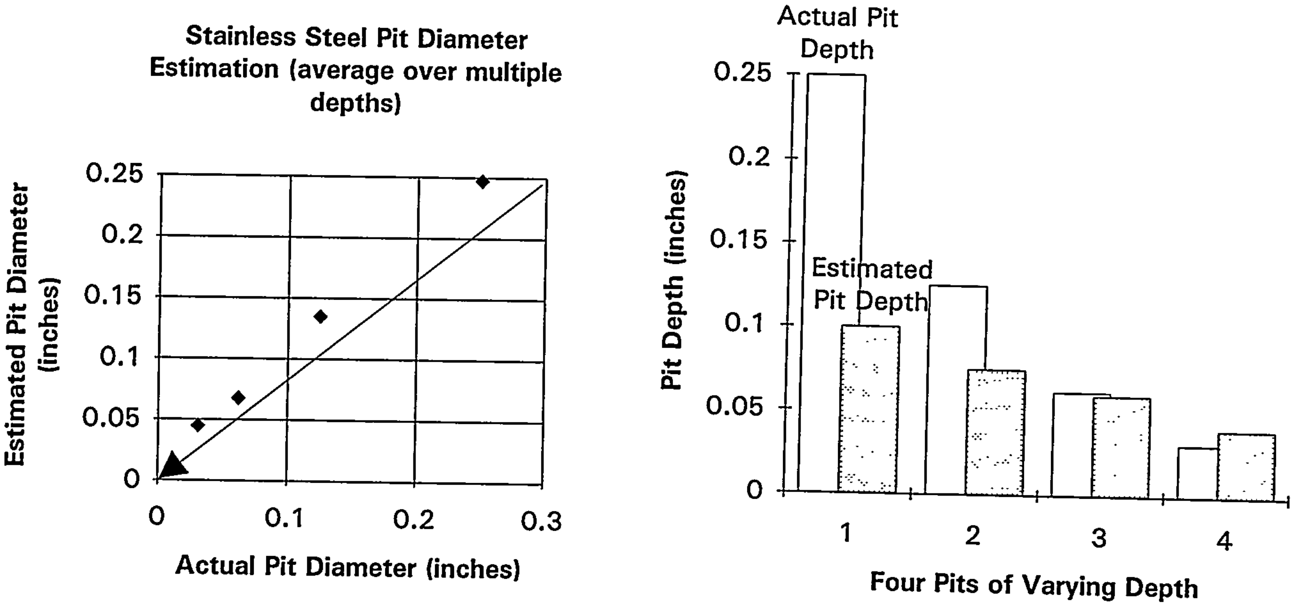

Figure 8. Pit sizing results

used the RTIEE to visually identify an area of potential corrosion attack, and then perform detailed and quantifiable electro-magnetic inspections of that area with the compliant scanner frame in contact with the tank wall. The ACFM sensor and defect characterization software has proved capable of detecting and sizing pits on stainless and carbon steels to a minimum diameter of .030".

The new prototype, under development at OSS, will increase the capability of the RTIEE to include inspection for cracks. In addition, the incorporation of a fly-by inspection mode will greatly increase the area of tank wall that can be inspected in a given time, and this in turn will increase the cost-effectiveness of the end effector system.

Acknowledgments

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Session 3

Decontamination and Decommissioning Focus Area

3.1 Advanced Worker Protection System

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Introduction

The Department of Energy (DOE) is in the process of defining the magnitude and diversity of Decontamination and Decommissioning (D&D) obligations at its numerous sites. The DOE believes that existing technologies are inadequate to solve many challenging problems such as how to decontaminate structures and equipment cost effectively, what to do with materials and wastes generated, and how to adequately protect workers and the environment. Preliminary estimates show a tremendous need for effective use of resources over a relatively long period (over 30 years).

Several technologies are being investigated which can potentially reduce D&D costs while providing appropriate protection to DOE workers. The DOE recognizes that traditional methods used by the EPA in hazardous waste site clean up activities are insufficient to provide the needed protection and worker productivity demanded by DOE D&D programs. As a consequence, new clothing and equipment which can adequately protect workers while providing increases in worker productivity are being sought for implementation at DOE sites.

This project will result in the development of an Advanced Worker Protection System (AWPS). The AWPS will be built around a life support backpack that uses liquid air to provide cooling as well as breathing gas to the worker. The backpack will be combined with advanced protective garments, advanced liquid cooling garment, respirator, communications, and support equipment to provide improved worker protection, simplified system maintenance, and dramatically improve worker productivity through longer duration work cycles.

Phase I of the project has resulted in a full scale prototype Advanced Worker Protection Ensemble (AWPE, everything the worker will wear), with sub-scale support equipment, suitable for integrated testing and preliminary evaluation. Phase II will culminate in a full scale, certified, pre-production AWPS and a site demonstration.

Objectives

The AWPS will establish a revolutionary and distinctive category of Personal Protective Equipment (PPE). The respiratory protection system will provide as a by-product an effective

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form of cooling to relieve heat stress. The AWPS will also include protective clothing which provides consistent protection to the worker commensurate with different types of chemical and radiological hazards. This will be the first such unit to provide respiratory protection, full body skin protection, and full body cooling in one package.

Heat Stress

Current systems for respiratory protection offer limited duration and provide little to no relief from heat stress for extended work periods or in hot environments. When cooling is provided, it must be provided separately and often adds significant weight to the overall worker protective ensemble.

Work activity at DOE sites will vary from relatively low metabolic energy costs at 100 watts (standing) to well over 500 watts (heavy two-handed work). The variability of work tasks have a large bearing on the design of the AWPS cooling system. The cooling can be adjusted independently of the breathing system. However, the more that is understood about the anticipated work loads, the more accurate the design of the AWPS.

DOE D&D will be undertaken in a variety of environmental conditions with a wide range of temperature and relative humidity. The high work loads required for many operations will produce large metabolic energy consumption causing workers to overheat even in low temperature, low humidity conditions. Most protective clothing and equipment by its very nature contributes to worker heat stress. Thus, it is important that clothing ensembles used at DOE sites must be designed to minimize the impact of heat stress. In addition, worker mobility must not be impaired as limitations in worker function also affect increases in heat stress and overall energy expenditures.

Work Hazards

DOE D&D workers face a multitude of hazards at each of the sites.

Contamination Contamination hazards exist at different levels of severity. Based on site visits, the principal contamination hazard to DOE workers is contact with radionuclides or other radioactive contaminants, principally particles. In addition, several sites also involve mixed wastes primarily metals, anions, chlornated hydrocarbons, fuel hydrocarbons, ketones, phthlates, PCB's, explosives, pesticides, alkyl phosphates, complexing agents, and organic acids. While most of the time these contaminants are encountered at relatively low con entrations, some substances have extreme toxic effects at these levels. Much of this contam-nation is found in specific areas at each site, particularly storage tanks, and former processing facilities.

Different contamination hazards will dictate different levels of protection. In those cases involving the most severe hazards, totally encapsulating protective ensembles must be used. More routine work involving self-contained respiratory protection will require protection against well-defined or incidental contamination hazards and a lower level of skin protection. For this reason, the AWPS will encompass two different forms of protective clothing: a vapor-protective totally encapsulating fully-integrated clothing system (EPA Level A); and a liquid splash-protective multi piece protective clothing system (Level B).

Physical The nature of D&D work produces severe physical hazards to DOE workers. Much of the dismantling of structures and removal of waste involves different forms of machinery. Therefore, worker PPE and other equipment must protect workers from the excessive vibration, sharp edges, and rough surfaces found in these environments.

Thermal As determined in site visits, the potential for flame contact and hot surfaces is expected to be at a minimum. Nevertheless, certain operations will involve extensive welding or cutting. These operations provide the potential for worker contact with flame or small amounts of molten metal. To meet these specialized protection needs, additional external protective items may be required.

Deficiencies of Existing Systems

Systems now used in hazardous materials applications have many deficiencies. A Self Contained Breathing Apparatus (SCBA) does not provide sufficient duration for efficient and cost effective hazardous materials operations and does not supply cooling to the subject. Supplied Air Respirators (SAR) have an indefinite duration, but do not supply cooling. They also limit wearer mobility due to the tethered breathing line required for these systems. Some users are employing an air supplied cooling vest. However this system consumes large quantities of compressed air usually requiring an umbilical, and has limited comfort because of the air flow into the suit and the hot air ejected by the vortex mechanism. When in a fully encapsulating or splash suit, without cooling the wearer is not capable of working safely or comfortably for more than 45 minutes even in a relatively cold environment. The worker typically requires a minimum of an hour rest to cool down after each 45 minute work period. In many environments this work period is reduced to twenty minutes.

Many outer protective garments worn today are either uncomfortable and bulky, or they are too expensive for large scale operations. Fully encapsulating suits rarely fit the wearer correctly, creating a more stressful and inefficient work environment. A more form fitting suit that is also less expensive would be more efficient to use in operations the size of DOE D&D work.

In addition, nearly all protective clothing in use today does not provide effective interfaces with other clothing or equipment. End users must use duct tape to seal gloves and boots to garments and to ensure that hoods stay over respirators. This practice defeats the purpose of wearing protective clothing constructed of barrier materials since particulates, liquid, and other contamination can easily penetrate these poorly constructed interfaces. Duct taping is also a time consuming, often two person operation.

Solution

Goals for the proposed AWPS address each of these areas by:

- Providing a high level of respiratory protection consistent with DOE site hazards combined with a cooling garment (to eliminate heat stress and provide for increased worker comfort, productivity, and safety) and protective clothing against both physical and contamination hazards;
- Integrating all clothing and equipment items such that they fit together, provide complete protection to the wearer, and allow ease of donning/doffing, decontamination, and maintenance;
- Replacing several existing protective clothing and equipment systems with two different systems which can be configured in several fashions to acco odate a wide range of operations while minimizing procurement needs, system maintenance, and training; and
- Improving cost efficiency of D&D operations.

AWPS Performance Objectives

The unique features of the AWPS objectives should dictate design for proper apparatus development. The primary source of this information has come from the potential end users of the AWPS, specifically the personnel involved in cleanup at the DOE sites. Site visits have been completed to Fernald, Hanford, and Oak Ridge. Information compiled from the visits is being used to guide the development of the respirator, cooling garment, and outer garment portions of the AWPS, and its associated support equipment.

Based on this information, the Portable Life Support System (PLSS), Liquid Cooling Garment (LCG), and protective clothing used as part of the Advanced Worker Protection System will have the following characteristics:

1. The PLSS will provide at least 2.0 hours of breathing air at an average respiratory consumption rate of 40 liters per minute per National Institute of Occupational Safety and Health (NIOSH) certification standards;

2. The PLSS will operate in any orientation;

3. The PLSS will operate in the pressure demand mode only and provide positive pressure operation at breathing rates up to 120 liters per minute. A normal breathing rate is considered to be 40 liters per minute while a heavy work rate is 100 liters a minute as prescribed by the National Institute for Occupational Safety and Health (NIOSH).

4. The PLSS will provide a clear indication of remaining air supply and provide a low air warning (audible and visual);

5. The overall PLSS weight will be no more than 40 pounds fully charged with liquid air. However, 30 pounds will be used as a design goal;

6. The PLSS outer shell will provide for ease of decontamination;

7. The PLSS will be refillable in a time similar to that of an SCBA high pressure (4500 psig) bottle. The typical fill time for a 4500 psig air bottle of an 60 minute rated SCBA is approximately 5 minutes with a cascade or compressor-based fill system

8. The LCG will weigh less than 6 pounds when fully filled with water;

9. The LCG will provide effective cooling for work rates ranging from 100 to 500 watts for a period of up to 2 hours at the highest work levels;

10. The garments in conjunction with the PLSS will form a complete protective envelope around the wearer. Interfaces between clothing items and the PLSS will maintain protective qualities commensurate with the clothing and PLSS systems separately;

11. The clothing will provide barrier protection consistent with the actual or potential hazards at DOE remediation sites;

12. The clothing will possess adequate strength and durability to prevent loss of suit or system integrity from the physical hazards of the DOE remediation site through the intended number of clothing use cycles;

13. The clothing will afford the wearer less than a 20 percent decrease in mobility, tactility, and time necessary to complete task functions when compared to the same activity without the protective clothing; and

14. The clothing, LCG, and PLSS in combination will extend working times compared to existing PPE ensembles by at least 100%.

Project Description

Hazards associated with the AWPS will be no greater than those associated with breathing apparatus used today. The AWPS is a low pressure system that has passive safeguards to protect against failure of the system and injury to the wearer.

The design of the cooling garment represents a well established technology which has been optimized for increased efficiency, decreased production cost, and decreased weight. Likewise, protective clothing systems will be based on existing, demonstrated materials used in current commercial products. Protective clothing improvements will encompass design enhancements, attention to interfaces, and complete integration with other equipment, all of which contribute to improved worker safety over existing systems.

Hazardous Materials PLSS

The liquid air in the AWPS is contained in the OSS-patented Advanced All-Position Dewar (AAPD). A dewar is essentially a pressure vessel within an outer vessel. A vacuum is drawn between the two vessels to minimize convective heat transfer to the cryogen. It is also designed in such a way as to reduce conductive and radiative heat transfer.

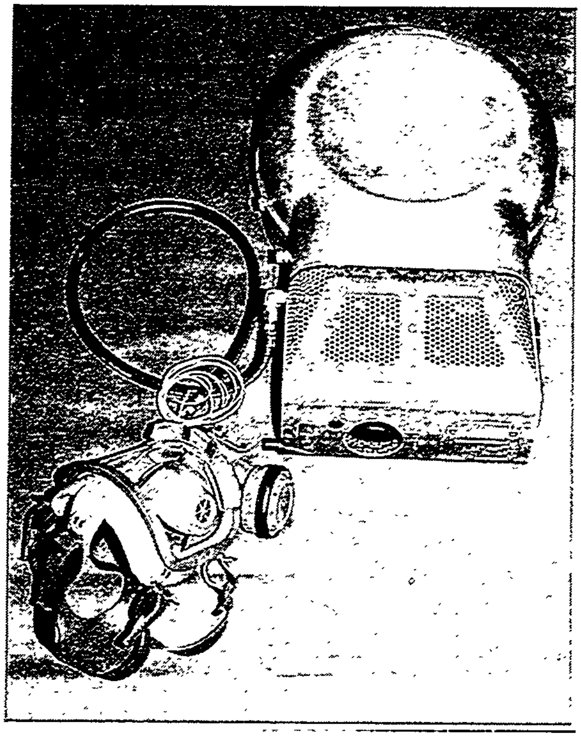

Figure 1. Photo of PLSS

The AAPD uses a pivot mechanism to maintain the vent line in the ullage space above the liquid and a liquid inlet at the lowest point of the Inner Vessel in all orientations. Liquid that is withdrawn from the pivot mechanism goes through the Liquid Hub and Liquid Line into the Vaporizer. In the Vaporizer, the cryogen is warmed to a breathable temperature and the LCG water is chilled to provide body cooling. It is located in the vacuum space between the Inner and Outer Vessel to reduce heat transfer with the ambient environment and increase efficiency.

If, due to a LCG Pump failure, the Vaporizer water freezes, the cryogen would freeze the water and the air leaving the vaporizer gets progressively colder. The Ambient Heat Exchanger (AHX) uses heat from the environment to warm the cryogen to a breathable temperature in the event of a Vaporizer freeze.

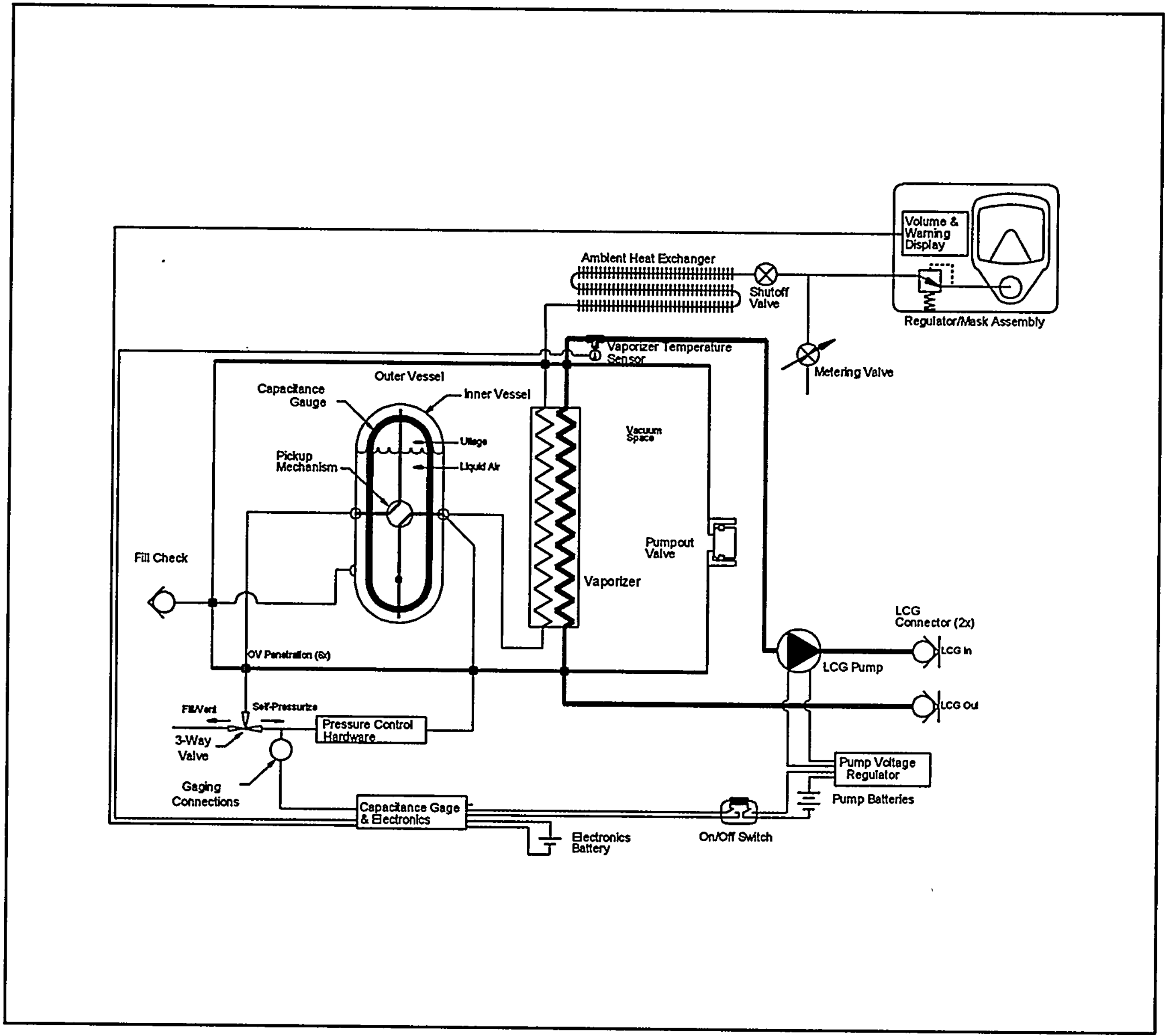

Figure 2. Hazardous Materials PLSS Functional Schematic

A Shutoff Valve after the AHX is used to control the flow to the Face Mask/Regulator and Metering Valve, turning the system on and off. The Face Mask/Regulator used for the Phase 1 AWPS was chosen for its ability to operate at the 60 psig inlet pressure.

The Metering Valve is used to control the cooling rate by adjusting the cryogen flow through the Vaporizer. Higher breathing flow rates result in larger cryogen flows and therefore more heat exchange with the LCG water. The user's breathing rate will be a function of the work rate and the cooling level will track the work rate. However, the cooling due to breathing at a certain metabolic rate is on average ⅔ of the desired level. That is, the heat required to vaporize and warm the cryogen to provide sufficient breathing gas at a given metabolic (breathing) rate is about ⅔ of the metabolic heat produced by the body. Since the liquid cooling garment provides the heat for vaporizing and warming the cryogen, breathing requirements provide about ⅔ of the required heat sink capacity for the LCG. The Metering Valve allows the user to increase cryogen flow through the Vaporizer and therefore his/her cooling rate.

The pressure is maintained in the backpack with a method commonly used in cryogenic dewars known as self-pressurization. Cryogen from the dewar is vaporized and the gas is provided to a Pressure Closing Valve (PCV) that keeps the AAPD at the set pressure. The AAPD also has a Relief Valve to protect against over-pressurization and a Burst Disc provides redundant pressure relief in the event of a relief valve failure.

The 3-Way Valve also in that section of tubing has two positions. In one, it routes the self-pressurization gas to the ullage. In the other, it is used to vent the dewar as it is filled. The AAPD is filled through a cryogen compatible Fill Check Valve to a separate port in the side of the Inner Vessel.

The LCG Pump drives the water through the Vaporizer and the LCG. The Vaporizer is specially designed to protect it from damage that could occur when the water freezes, and for forced de-icing of the vaporizer so that the AWPS can be put back into service quickly if a freeze occurs.

The AWPS also has a Volume and Warning Display mounted on the Face Mask that allows to user to monitor the system. The The display has indicators for system status and remaining duration. System status displays include High and Low pressure indicators and a warning light for pump failure. The duration is marked in 8 divisions (15 minutes/division) When the volume reaches ¼, and audible alarm is set off. For the AWPS, this could still mean 30 minutes of time remains so a "snooze" button has been added. However, when the volume reaches ⅛, the button no longer works and the audible alarm remains on.

The liquid air volume is measured using a Capacitance Gauge integrated into the Pivot Mechanism. The gauge is a pair of parallel plates in a geometry such that the liquid height between them in all orientations is a uniform function of the volume remaining.

Panels in the Bottom Box provide access to the Electronics and Pump Batteries and the Power Switch for the system.

Face-Piece, Regulator, Communications

An InterSpiro face-piece and regulator were modified to incorporate backpack status displays and optional communications. The InterSpiro regulator and face-piece system is used in existing NIOSH certified compressed air SCBAs.

The Phase I AWPS incorporated off-the-shelf communications. The communication system was modified to mount directly to the InterSpiro face piece.

High Performance LCG

The three primary methods the body uses to reject heat (radiation, convection, and evaporation) are all severely impaired in the environment of protective clothing. The result of this heat build-up is heat stress and impaired mental awareness. The High Performance LCG eliminates heat stress by assisting the body in rejecting heat.

The High Performance LCG consists of a body-suit type garment constructed of stretchable material with tubing outside and patches attached to the inside. Heat is conducted away from the subject by circulating chilled water from the vaporizer through the patches that lie against the surface of the skin. The patches provide a large surface area for heat conduction, and are located in various locations such as on the upper torso, thighs, and upper arms. The large area of contact between the patches and the subject's skin will allow a comfortable temperature cooling water to be used.

Many of the cooling suits available on the market today are constructed of small water carrying tubes which are either sewn or woven into a snug fitting suit. The process of attaching and routing the tubes within the suit is time consuming and expensive. The tubing itself is also expensive when compared to the raw materials used in the patch garments. The patch manufacturing process is also much less expensive and less time consuming. Because the patch system has much less flow resistance, and can effectively utilize warmer water, it is inherently a more efficient system to be integrated with the AWPS.

Protective Clothing

Based on the three site visits and the backpack and LCG development, desirable features for both Level A (vapor protective) and Level B (liquid splash protective) suits were determined. Commercially available Level A & B suits were evaluated by the following criteria:

Figure 3. Photo of High Performance LCG

- cost (simplicity);
- mobility in terms of reaching, bending, and kneeling;
- relative comfort during operational use;
- relative comfort during non-operational use, such as before connection of the PLSS/LCG systems;
- ease of donning and doffing;
- ability for wearer to don and doff suit without assistance;
- potential for contamination of wearer during operation;
- ability to prevent contamination of wearer and PLSS during doffing;

- number of sizes needed to achieve appropriate fit of most wearers;
- potential problems with PLSS and LCG interface; and
- overall relative fit.

Level A Suit A review of existing vapor protective suits currently on the market showed that the commercially available Chemfab "Challenge 6400" met the requirements of the AWPS. A training version of the Chemfab suit was obtained and used for prototype testing.

A standard "Challenge 6400" vapor protective suit is reusable with the following characteristics:

1. Integral visor, detachable gloves, and sock-like booties and will be worn over the worker and the PLSS.
2. The hood area of the suit provides ample room for a worker to wear a hard hat, respiratory face mask, and communications set.
3. The back area of the suit has an expanded pouch-like protrusion to accommodate wearing of the PLSS backpack.
4. An inner and outer glove system is used. Outer gloves are Brunswick neoprene gloves with North Silver Shield inner gloves. Gloves are mounted to a hard ring fastened to the end of the suit sleeve. Glove mounting will be accomplished by using a low profile nylon or acetal tie down strap. The glove interface area is covered by a splash shield consisting of the garment material and extending three inches down over the interface area from the forearm area.
5. Booties are fully integrated into the termination of the suit leg. Splash guards consisting of the garment material are provided to prevent liquid accumulation into the outer boots. Bata Shoe HazMax boots will be used as the outer boots.
6. Garment seams (on full production units) are heat sealed and taped on both sides. The garment to visor interface is heat sealed with heavy duty taping on both sides of the seam.
7. The suit closure consists of a gas sealing (on full production units) zipper located on the suit back. The zipper begins at the head region and extends to the upper left thigh region of the suit.
8. Suit sleeves allow the wearer to withdraw his or her hand into the suit interior.
9. The visor consists of 10 mil Teflon FEP sized to provide adequate side-to-side and top-to-bottom vision.

Level B Suit No commercially available Level B suit satisfactorily met the design criteria, there-fore a suit was designed by OSS specifically for the AWPS. A comparative study was conducted to determine the optimum configuration of the project liquid splash protective suit. Three different options were selected and compared in terms of different features and their affect on wearer comfort and job effectiveness:

1. Option 1 involved a two-piece design. The first piece was a standard coverall with full sleeves and attached gloves and sock-like extensions of the trouser legs (booties). The second piece was a hooded cover with integrated visor, half length elasticized sleeves, and a bottom

hem positioned near the waistline of the wearer.

2. Option 2 also involved a two-piece design. Unlike Option 1, the first piece was an bib-style overall with adjustable suspenders and no sleeves. This overall did include booties as in Option 1. The top piece was hooded top with integrated visor, full sleeves, attached gloves, and a bottom hem positioned near the waistline of the wearer.

3. Option 3 was a one piece design similar to the Level A Vapor-Protective Suit but not of a gas-tight construction. This design included an integrated visor, full sleeves with attached gloves, booties, and rear-entry closure provided with a cover flap.

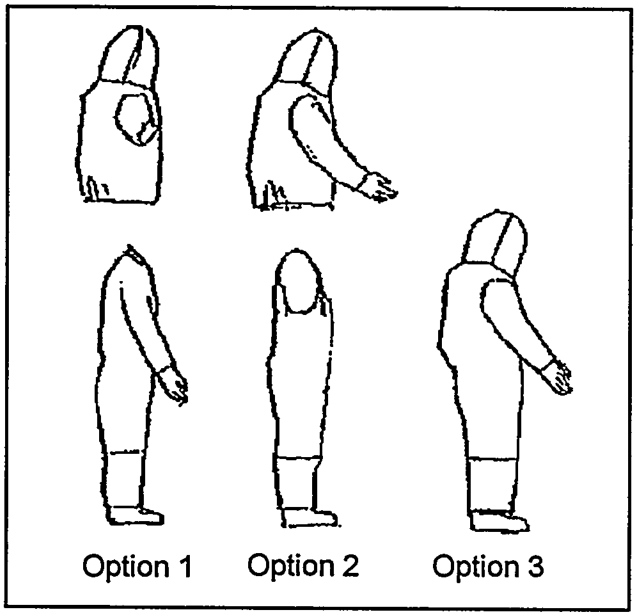

Figure 4. Level B Suit Design Options

Each option used outer boots worn over the booties for providing physical protection to the wearer's feet. Figure 4 illustrates each of the liquid splash suit configurations considered.

A weight between 0.3 and 1 was assigned to each rating area based on OSS's perception of DOE requirements and needs in combination with project objectives. Each characteristics of the considered design configurations was rated using a 1 to 10 scale. A composite score was calculated using the product of the weight for each characteristic and the rating. This composite score was used to rank the three options. Using this system, Option 2 (full hooded top with bib-overall) yielded the highest score, followed by Option 1 (standard overall with short-sleeved hooded top. The encapsulating suit (Option 3) provided the lowest score-primarily because low relative comfort compared with other options and the inability to perform self donning and doffing.

The option 2 suit was fabricated with the following characteristics:

1. It is a two piece garment made of Tychem 9400 (E.I. Dupont deNemours & Company) with coverall and hooded shroud consisting of overalls with suspenders and splash hood with visor and full sleeves with gloves. It will be worn with the PLSS outside the overall but inside the splash hood.

2. The suit hood has a 20 mil PVC visor.

3. Brunswick coated knit neoprene gloves are used with the suit. Gloves are mounted to a light-weight, plastic hard ring fastened to the end of the splash hood sleeve. Glove mounting is accomplished using a delrin or acetal cable tie. The glove interface area is covered by a splash shield consisting of the garment material and extending three inches down over the interface area from the forearm area.

4. Booties are fully integrated into the termination of the suit leg. Splash guards consisting of the garment material are provided to prevent liquid accumulation into the outer boots. Bata Shoe HazMax boots are used as the outer boots.

5. Garment seams are sewn and taped on the outside.

Sub Scale Recharge Station

The sub scale recharge station is a prototype system that uses liquid nitrogen to convert compressed breathing air into subcritical liquid air. The recharge system consists of a heat exchanger with all the necessary monitoring instrumentation to ensure that the system is operating properly. The consumables, breathing air and industrial grade LN_2, are standard items and will be purchased from outside gas suppliers as required for use.

The AWPS Fill System uses cooling coils immersed in a liquid nitrogen bath to chill compressed air to cryogenic temperatures. Air from a high pressure source is reduced to 300 psig and then flows through length of tubing immersed in liquid nitrogen. The liquid air is regulated to 40 psig before entering the AWPS backpack..

Other service equipment included controls for the Vaporizer De-icer, an auxiliary power supply, and a small water reservoir to bleed air bubbles out of the LCG loop, if necessary.

Results of Manned Testing

The Institute for Environmental Research (IER) as Kansas State University was subcontracted to independently evaluate the Advanced Worker Protection System (AWPS) in a series of human subject, controlled environment tests. The principal objectives for testing the AWPS at KSU's Institute for Environmental Research were:

- To provide an independent assessment of the AWPS specifically with the intent of evaluating how the system and its components affect the physiology and perceptions of human subjects wearing this system in both a Level A and B configuration;

- To allow a comparison of the effects on worker endurance for the AWPS liquid air based breathing system with liquid cooling garment versus a traditional, compressed air-based, breathing system with no provision for cooling; and

- To investigate the differences in AWPS performance under different environmental conditions likely to encompass the working situations of end users.

The Institute for Environmental Research at Kansas State University (Manhattan, Kansas) was chosen based on the availability of its highly controlled environmental chambers and their experience with conducting human subject tests involving prototype personal protective equipment.

Test Plan

A two-part test program was designed to achieve these objectives. The first part of this test program involved using a single set of environmental conditions to compare the performance of the AWPS and SCBA-based protective ensembles.

Four different ensembles were evaluated, including:

1. The first ensemble consisted of the prototype PLSS, the prototype LCG, and prototype Level A vapor-protective suit. The protective suit was a ChemFab totally-encapsulating training suit which includes attached Neoprene gloves and booties. Industrial PVC outer boots were worn over the booties. The diaphragm type suit exhaust valve was removed and used for a pass-through for the sensor leads.

2. The second ensemble also consisted of the prototype PLSS and LCG but included a two-piece Level B splash-protective suit. A prototype splash-protective suit was fabricated using the TyChem 9400 material in a two piece design. Sensor leads were passed under the hooded top.

3. The third ensemble used the Level A suit described in the first ensemble with a Interspiro Spiromatic 9030 SCBA. There was no provision of cooling.

4. The fourth ensemble used the Level A suit described in the first ensemble with a Interspiro Spiromatic 9030 SCBA. There was no provision of cooling.

Hard hats and communications sets were worn with each ensemble.

The second part of the testing involved the evaluation of the second ensemble (Level B AWPS) with two different sets of environmental conditions with the intent to determine how human subjects wearing the system would respond to colder and hotter situations.

Test Conditions

The first part of testing was conducted at the following conditions:

Temperature:	85°F (29.4°C)
Relative Humidity:	75%
Air Velocity	< 30 fpm (0.15 m/s)

The environmental conditions chosen for the second part of testing included:

Low temp condition:
45°F (7.2°C)/75% relative humidity
High temperature condition:
100°F (37.8°C) 95% relative humidity

Environmental conditions were maintained using a controlled chamber having the following specifications:

Size:	11.2 ft by 11.2 ft x 9 ft
Temperature:	0 to 140°F (-18 to 60°C)
Humidity Range:	20 to 95% RH at moderate temperatures
Air Velocity:	< 30 fpm (0.18 m/s)

Human Subjects

A total of four different human subjects were used in these evaluations. Test subject ages ranged from 18 to 30 and each was an experienced fire fighter from the local fire department familiar with the use of SCBA. Since only one size of the AWPS was prototyped, all test subjects were of average build and dimensions, able to fit large-sized prototype protective clothing. All test subjects were volunteers in good physical condition as demon-strated in baseline testing (without protective clothing or equipment).

Each test subject wore underpants, a T-shirt, sweat socks, and full body cotton coverall under protective clothing.

Exercise Protocol

Human subjects were exposed to a constant work rate using a tread mill adjusted to a sufficient rate and incline to achieve an average energy expenditure of 400 to 500 kcal per hour. This work rate was used in all tests.

Test Measurements

The following physiological measurements were monitored during each test:

- Core temperature
- Skin temperature
- Heart rate
- Temperature inside the protective garment

In addition, test subjects were asked to rate aspects of their comfort using a qualitative scale.

Test Matrix

All test subjects under went baseline testing in shorts and T-shirt to establish physiological parameters and to become accustomed to the exercise protocol. Each test subject wore each ensemble once. The order of wearing the four different ensembles was randomized for each test subject. Only one subject at a time was tested:

1. A total of twenty (20) tests were undertaken, including the baseline, for comparing AWPS and SCBA-based ensemble testing.
2. Eight (8) additional tests were conducted using the liquid splash-protective ensemble at the two extreme environmental conditions.

Test duration was for a maximum of two hours (once actual exercise started) or when the test subject wished to stop for any reason, or when the test subject's core temperature or heart rate exceeded prescribed exercise levels (39 °C or [220 - subject's age] beats per minute, respectively).

Data Analysis

Each attribute which is measured continuously or discretely during testing was be plotted versus test time showing a comparison of baseline and each ensemble. For the first series of tests, curves of each attribute versus time was shown for the four different ensembles plus baseline. A separate curve was shown for:

- AWPS with Level A suit, PLSS, and LCG;
- AWPS with Level B suit, PLSS, and LCG;
- Level A suit with SCBA;
- Level B suit with SCBA; and
- Baseline condition (no protective clothing or breathing apparatus).

In the second series of tests, splash suit performance was shown for each of the three different environmental conditions using the AWPS with Level B suit, PLSS, and LCG:

- 85 °F and 75% relative humidity;
- 45 °F and 75% relative humidity, and
- 105 °F and 75% relative humidity.

Test participants were asked to rate ensembles and ensemble performance through a questionnaire following the testing.

Test Procedures

Prior to each test, the AWPS backpack was filled with liquid air. This was done using the fill station using compressed breathing air and commercial grade liquid nitrogen as consumables. The pack was then pressurized to operating pressure (60 psig).

Meanwhile, the test subject was getting ready. EKG and skin temperature sensors were attached at various locations on his body. For AWPS tests, the test subject donned the liquid cooling garment (LCG) before continuing with a pair of coveralls, sweat socks, the protective suit pants, and boots. Each piece of clothing was weighed before and after the test. Once suited to this stage, the test subject would proceed to the fill area to don the AWPS or SCBA.

The test subject then returned to the chamber to don the upper portion of the protective garment and begin the test. After returning to the environmental chamber, the test subject began breathing from the mask, donned the hard hat, and donned the upper portion of the protective garment. A final weight was taken, and recorded. This measurement allowed the treadmill to calculate how much work was being done. The treadmill velocity was increased until the caloric expenditure was approximately 7 cal/min. This usually gave a walking speed of ~3 miles/hour. This process was intended to ensure that all test subjects were working at approximately the same rate.

As the test subject began walking, he was asked to rate his thermal comfort from 1 to 9 (1=very cold, 9=very hot). Every five minutes, he was again asked to rate his thermal comfort overall, and to report any distribution of hot or cold across various body parts. The test subject was also requested to indicate any change in operation, status indicator lights, or personal comfort whenever those things happened. The comments were recorded in the log book along with heart rate, core temperature, and LCG temperatures as the test progressed. The test subject was instructed at the beginning of the test and whenever his physiological indicators showed signs of stress that he could and should stop if he experienced headaches, dizziness, nausea, shortness of breath, or other discomfort. Other termination criteria used by the test conductor included heart rate, core temperature, breathing gas supply, and hardware malfunction. During the first two tests, heart rate was limited to 80% of 220-age. It was realized that the work rate expended by the test subjects was fairly intense, and it was decided that this heart rate limit was too conservative. The limit was raised to 100% of 200-age, with the understanding that the test subject was to be carefully monitored if his heart rate went above the 80% level. In addition, the EKG showed fluctuations and irregularities particularly as the test subjects


Figure 5. Manned Test Preparation

became fatigued. It is believed that these artifacts were caused by chest muscle contraction. If the artifact became so severe that the EKG machine could not reliably count the heart rate, the test was terminated. The final test termination criteria was depletion of the air in the pack.

After the test, the subject was weighed and then removed the top of the protective garment and the mask, and filled out an ensemble rating evaluation. Then the backpack was removed and taken to the fill station to vent any remaining cryogen and warm the vaporizer in preparation for the next test. As at the beginning of the test, weights were taken as each type of clothing was removed. Additional comments were solicited from the subjects and recorded in the log book. The LCG and coveralls were laundered and the protective garments were allowed to dry before the next test.

Data Conclusions

There were a number of contributing factors which affected the outcome of some tests, and in some cases, these factors caused tests to be prematurely terminated. Some examples include, blisters developing on the feet of one test subject, headache due to overly tight mask straps, and failure to increase cooling. The latter problem was more an artifact of the testing given the unfamiliarity of the test subjects with PLSS technology. In general, cooling was increased at 15 intervals and at the request of the test subject. This led to variable cooling levels for individual test subject; however, cooling perception is subjective and needed accommodation during these tests.

There were relatively few physiological response differences between the two different types of suits for each test subject. Since the materials used in the construction of each suit were impermeable, the environment immediately next to the test subject's body became rapidly saturated as expelled humid air was released from the breathing mask (of PLSS or SCBA) into the suit. The Level B ensemble should allow for better air exchange with the outside environment compared to the Level A ensemble since it is not of an air-tight design. Relative humidity inside the suit was not measured in any of the tests but no difference between different ensembles on the basis of humidity was perceived. In general, the inner of all test subjects was soaked with perspiration following each test.

Figures 6 and 7 illustrate the dramatic improvements in duration and thermal comfort possible with the AWPS. The two graphs show skin temperatures for the same test subject wearing a level A suit with an SCBA and the AWPS respectively.

Two major observations from these graphs are the overall duration and the rise (or fall) of skin temperatures. The first graph (SCBA) shows a duration of approximately 30 minutes, and all the skin temperatures show an increase. In comparison, the AWPS graph shows a duration of approximately 75 minutes, with a much smaller rise in skin temperatures. In fact, several of the measurements consistently decrease.

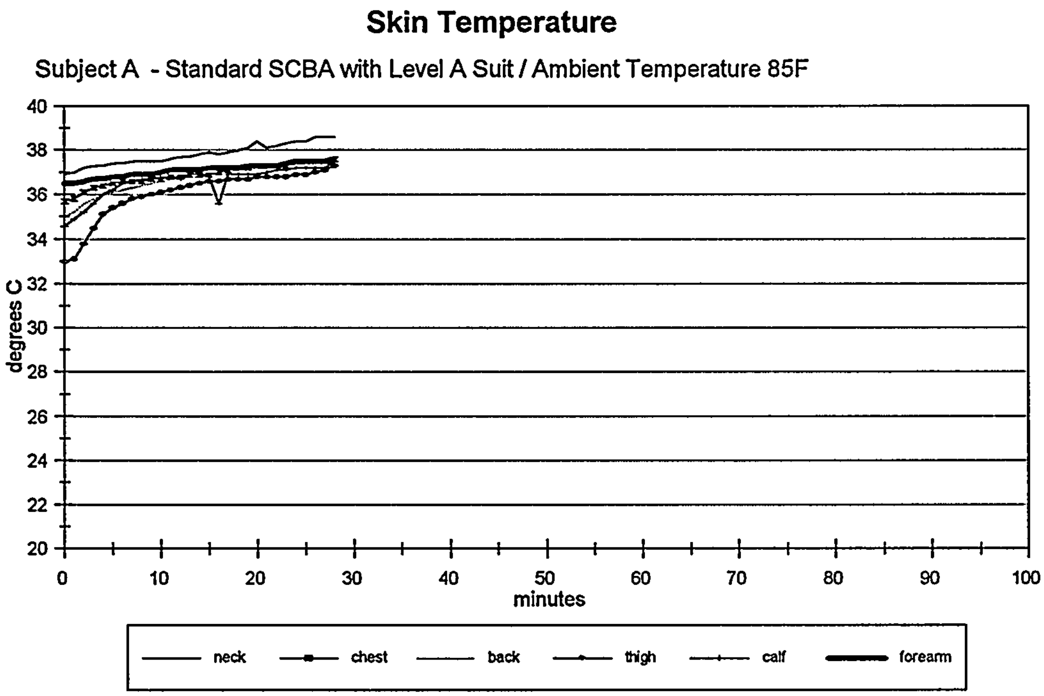

Figure 6. Representative Skin Temperature Graph with Standard SCBA

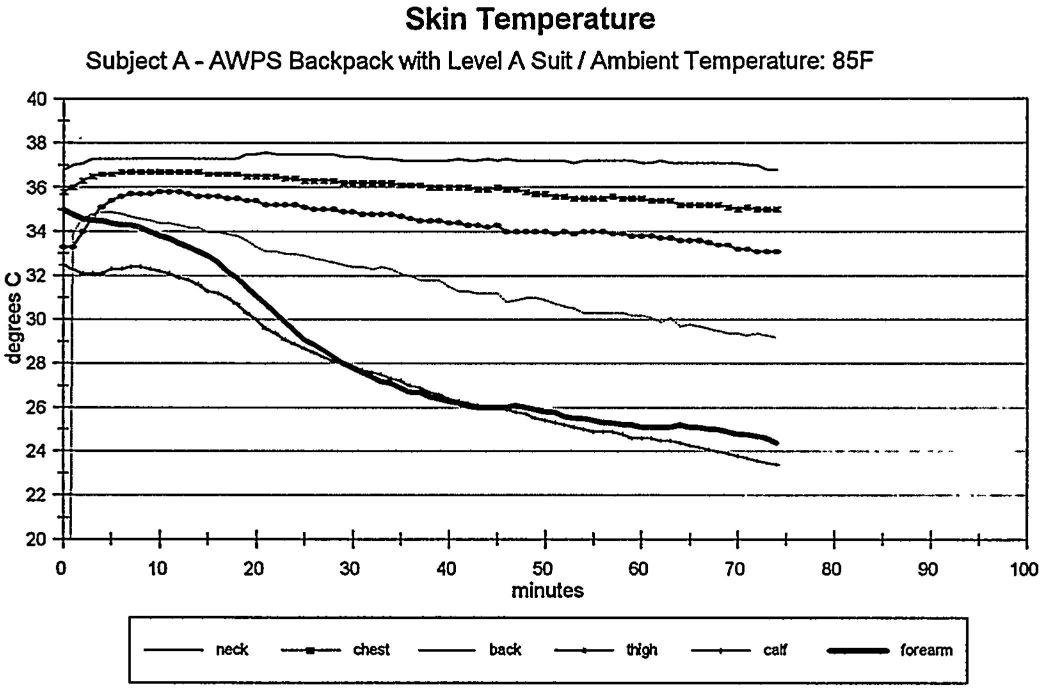

Figure 7. Representative Skin Temperature Graph with AWPS Backpack

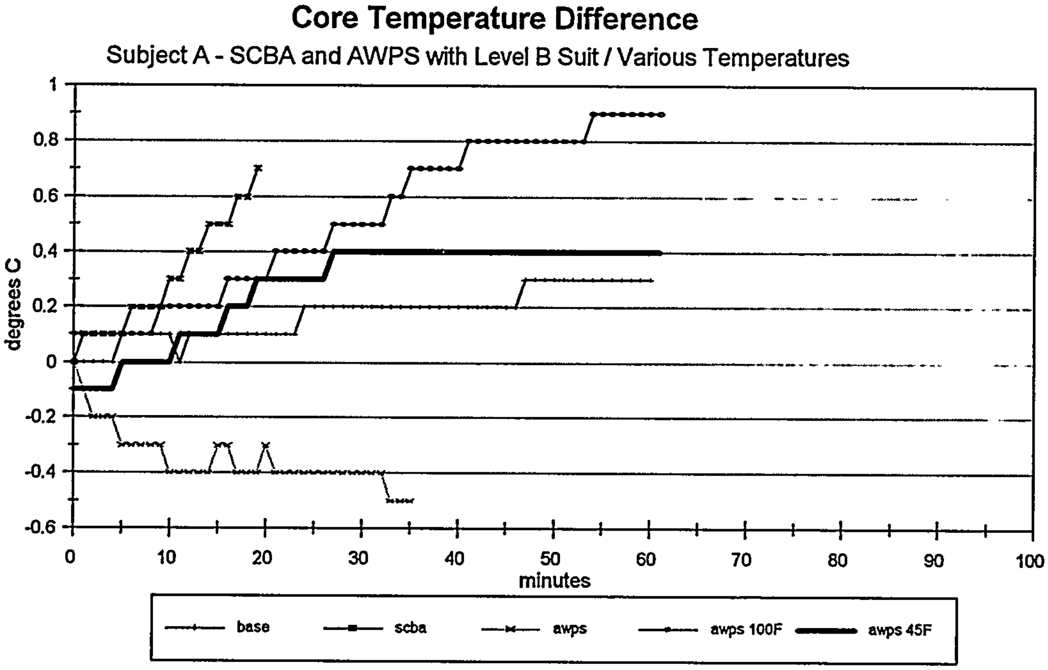

Figure 8. Representative Core Temperature Graph

Skin temperatures are a good indication of perceived thermal comfort. Core temperature is a much better measure of actual thermal condition and shows how much heat the body is losing or storing. People naturally have different core temperatures, and that temperature varies throughout the day. In addition, the important information from core temperature is how it changes. For both of these reasons, core temperature difference is shown in figure 8. The core temperature at the beginning of the test was taken to be zero, and all subsequent temperatures were plotted as a difference. This graph is for a level B suit and includes five different tests, baseline, SCBA and AWPS at 85 °F, and AWPS at 100 °F and 45 °F. Again, the observations to be made from this graph are duration and rate of temperature change.

The baseline test was conducted with the test subject wearing only shorts, a t-shirt, and running shoes. The other tests (on this graph) were conducted with the test subject wearing coveralls, the cooling garment (for the AWPS tests only) the backpack and a level B suit.

Looking at the slopes of the core temperatures, the 85 °F AWPS test actually shows a decreasing core temperature. This test lasted for approximately 35 minutes and was terminated early due to fatigue and sore feet. The baseline and 45 °F AWPS had the smallest core temperature rise. Both tests lasted approx-imately 60 minutes and had similar core temp-erature increases. The 100 °F AWPS had the next fastest core temperature rise (~.9C) and a duration of 65 minutes. While the SCBA (with no cooling) had the steepest core temperature rise of .7 °c in 20 minutes. A good indication of thermal stress is a core temperature rise of 1 °C. The SCBA test would have reached this level at approximately 28.5 minutes, less than half the duration of the 100 °F test which had a 15 °F warmer environment.

Benefit Analysis

Visits to three DOE remediation sites (Fernald, Oak Ridge, and Hanford) yielded the information necessary for determining the life cycle costs for Personal Protective clothing and Equipment (PPE) used in the existing ensembles. This information has been applied in the form of a model which can also determine the equivalent life cycle costs for proposed AWPS configurations. Following is a brief description of the model, and updated costs based on current design and testing data.

Specific life cycle cost information was gathered at each of the sites using an elaborate survey and interview process. This information included:

- The number of missions (sorties) involving PPE;
- The number of individual PPE uses;
- The expected number of uses for each PPE item (service life);
- The number of PPE uses for each maintenance cycle;
- The proportion of PPE use activity involving high, moderate, or low work rates;
- The proportion of PPE use involving Level A or B protection;
- The specific PPE items in use and their purchase costs or sources;
- Consumable and their costs associated with each of the PPE items; and
- Labor costs associated with each of the PPE items.

In some cases exact data was not avai-lable for an item, and estimates were used.

Development of Model

The life cycle cost of PPE is based on costs associated with:

- Purchase;
- Operations;
- Cleaning or decontamination;
- Maintenance and repair;
- Storage; and
- Disposal.

Incumbent in these costs are both labor and material costs. For example, the purchase price for the PPE includes not only the price paid for the item, but the time spent ordering the item, receiving the item, and putting the item into service. Operating costs include both labor and supplies needed to replenish the system. The same is true for maintenance (repair), storage, and disposal costs.

In its simplest form, the total support cost (T) is the sum of each of these costs:

$$T = P + O + C + M + S + D$$

Equation 1

where:

T = Total support cost
P = Purchasing costs
O = Operating costs
C = Cleaning and decontamination costs
M = Maintenance and repair costs
S = Storage costs
D = Disposal costs

For the complex PPE ensembles used in DOE remediation operations, these costs are calculated for each major item of the ensemble--respirator, protective clothing, cooling system, communications device, and any other equipment. In comparing ensembles, only those items which are different need be considered. However, each item must be placed on the same time basis to allow a comparison of the total support cost. For the purposes of this study, the total PPE support costs for a specific job (with a defined amount of actual work) has been chosen as the basis for providing estimates.

Model Variables

Different variables affect each of the individual cost areas in Equation 1. Some costs are based on the number of uses, while others are based on the number of missions involving PPE use or the total number of items in use. In addition, since tasks and consequent PPE item selection vary, the model must account for differences in work activity. For example, heavy work activity will more rapidly consume breathing air supply (for SCBA), increase clothing wear, and create the need for maintenance or disposal. Costs also vary based on the type of worker protection needed, i.e., EPA Level A versus Level B protection. Table 1 lists the different variables involved in determining the costs in each PPE cost area.

Basis for Comparing Different Ensembles

Since the principal utility of the AWPS is for worker protection during Level A and B operations, life cycle costs have been computed for these operations only. Initial visits to the Fernald Environmental Management Project, Hanford, and Oak Ridge DOE sites showed an increase in both Level A and B clothing ensemble uses as decontamination and decommissioning processes fully begin.

For the purposes of this study, a total of 5,000 man hours are assumed to require self-contained breathing air for a specific remediation function. Ninety five percent of this time is assumed to require Level B protection (4,750 man hours), while 5% of that total (250 man hours) are assumed to require Level A protection. Both of these figures are based on the actual number of man hours required to complete the work and not time spent in non-work activities such as donning, doffing, decontamination, and rest periods.

The analysis of life cycle costs is therefore based on the costs needed to affect the indicated number of work hours. A comparison is made between conventional Level A and B systems and the AWPS Level A and B systems.

The current Level A ensemble used in the DOE weapons complex includes:

- A limited use totally encapsulating suit constructed of a lightweight plastic laminate-based material;

Table 1. Model Variables For Each Cost Area

Cost Area	Item Specific?	Model Variables
Purchase	Yes	Number of PPE uses per year Number of uses per PPE item PPE item purchase price PPE purchasing hours Purchasing hourly labor rate
Operations	Yes	Number of PPE uses per year Ratios of low, moderate, and high work level activity Consumable costs Worker labor hours Worker hourly labor rate Indirect labor hours Indirect hourly labor rate
Cleaning and Decontamination	No	Number of PPE uses per year Number of missions per year Consumable costs Proportion of Level A/B use Decontamination labor hours Decontamination hourly labor rate
Maintenance and Repair	Yes	Number of PPE uses per year No. uses per maintenance cycle Ratios of low, moderate, and high work level activity Consumable costs Maintenance labor hours Maintenance hourly labor rate
Storage	Yes	Number of PPE uses per year PPE item purchase cost
Disposal	Yes	Number of PPE uses per year Number of uses per PPE item PPE item purchase cost

- Attached gloves; and
- A 30 minute or 60 minute Self Contained Breathing Apparatus (SCBA).

While other items may be worn with the ensemble such as a over boots, cooling system, communications device, and hard hat, none of these items were considered in estimating total ensemble purchase costs. The suit in this scenario is used only once before requiring disposal. SCBAs have a 5 year service life under heavy use conditions (approximately 1000 uses). The encapsulating suit purchase price is approx-imately $400, while the SCBA costs $2200.

The AWPS Level A ensemble includes:

- A reusable totally encapsulating suit constructed of a durable material;
- Attached gloves;
- An inner cooling garment; and
- The liquid air PLSS.

The suit in this scenario is expected to provide five uses before requiring disposal and costs $3500. The PLSS also is estimated to have a 5 year service life (1000 uses) with a purchase price of__.

The Level B scenario includes the same ensembles but a liquid splash protective suit is substituted for the totally encapsulating suit. For the existing ensemble, this clothing costs $25 and is disposable after a single use. In the AWPS Level B ensemble, the purchase price for the splash suit is $150 and is designed for only one use.

Since each ensemble consists of two separate parts having different service lives, the individual life cycle costs must be calculated for each.

Cost Estimation Method

While some information was obtained during the site visits, the majority of costs have been estimated using related data for EPA remediation activities. Specifically, a series of equations have been developed by Schwope and Renard, "Estimation of the Cost of Using Chemical Protective Clothing,"which define the individual costs and labor requirements of protective clothing as a function of its purchase price. These relationships were used for determining labor requirements for both existing and AWPS ensembles.

Assumptions Despite information gathered on site visits and methods of estimation provided above, a number of assumptions were necessary to calculate life cycle costs. In the first set of calculations, several simplifications of the model were used. These included:

1. All work is conducted at a moderate work load;
2. All operational labor is at a rate of $40/hour (includes overhead);
3. All other labor, including labor for purchasing, storing, and disposal, is at a rate of $25/hour (includes overhead).
4. Decontamination costs are the same for Level A as they are for Level B; and
5. Decontamination labor costs are based on the protective suit only. This include decontamination of gloves, outer boots, and other accessory clothing items but not the respirator. Protection of the breathing apparatus within the suit excludes decon-tamination (but not main-tenance) costs for this item.

Under a moderate work load, the SCBA offers a maximum of 20 minutes useful work time, while the PLSS is assumed to provide 120 minutes useful work time. Table 2 below shows the individual times associated with operational uses of each ensemble (applies to both Level A and B):

Other specific assumptions were required for other costs, particularly purchasing labor consumable for different activities. The following estimates were used:

- Purchasing labor for the existing ensemble is considered to be twice as much for the AWPS due to myriad of choices affecting current clothing purchases. The AWPS purchase labor requirement is 20 hours per year. These costs are based on the case of equipment needs for a 5000 man-hour job as described above.
- Operation consumable vary between ensembles primarily due to the difference between compressed and liquefying air. The AWPS uses 270 cubic feet of compressed air at $0.05/scf and 12 lbs of liquid nitrogen at $0.38/lb for a total con-sumable cost of $16.65. In comparison, the SCBA uses 30 scf of compressed air for a total cost of $1.35.
- Cleaning/decontamination consumable have been previously estimated (by Schwope and Renard) at $50/entry for disposable supplies (e.g., detergents, rags, ground cloths) and $25/entry for reusable supplies, such as buckets, brushes, and decontamination showers
- Maintenance costs are assumed to be the same for AWPS PLSS and the existing ensemble SCBA at $25/maintenance cycle and 10 uses for every maintenance cycle. Likewise, the principal main-tenance for protective clothing is the replacement of the gloves at $25/pair with a total of 20 use and decontamination cycles assumed for each set of gloves.

Table 2. Time Required in Ensemble Operations

Activity	Amount of Time Each Activity (minutes)	
	Current Ensemble	AWPS
PPE donning	15	25
Pre-entry inspection	0	10
Working time	20	120
Decontamination	10	10
Rest time	30	30
TOTAL TIME REQUIRED	75	195
Ratio actual work to total time required	0.27	0.62

Life Cycle Cost Calculations (Base Case)

Table 3 shows the values used for each variable in the model based on the estimates and assumptions above. The AWPS shows a significantly smaller life cycle cost for both Level A and B operations for the 5000 work hours.

Results using the life cycle cost model are presented in Table 4. The overall Life Cycle Cost (LCC) for the existing ensemble was calculated as $1,676,851. For the AWPS, a LCC of $980,886 was obtained.

This represents a difference or savings of nearly $700,000, see figure 9. Note that while the acquisition (purchase) costs of the AWPS hardware are significantly higher than for the existing ensemble, much lower costs were demonstrated for the other expense areas. The majority of the AWPS LCC was the purchase cost which had roughly the same contribution as the operational cost. Over 50% of the LCC for the existing ensemble was due to operational costs.

Table 3. Cost Model Variable Values

Variable	Existing Ensemble	AWPS
Level A suit purchase cost	$400	$3500
Level B suit purchase cost	$25	$150
SCBA purchase cost	$2200	___
Labor rate (except operations)	$25/hour	same
Purchasing hours	20/year	same
Actual work time/use	20 minutes	120 minutes
Support time/use (see Table 2)	55 minutes	75 minutes
Total time/use	75 minutes	195 minutes
Ratio actual to total time	0.27	0.62
Operations labor rate	$40/hour	same
Consumable/use	$1.35	$16.65
Decontamination consumable/mission	$25	$50

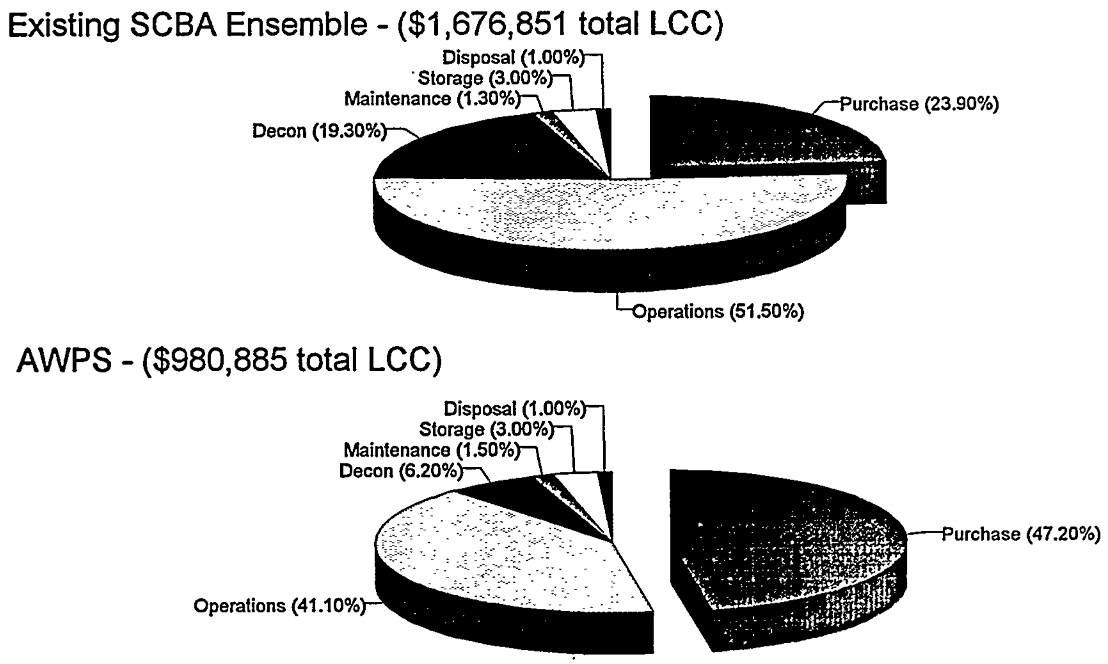

Figure 9. Proportion of Costs of LCC

Table 4. Calculation of Life Cycle Costs (Simple Model - Best Case)

Total # of work hours	5000
Ratio of Level A work	.05
Ratio of Level B work	.95
Total Hours Level A	250
Total hours Level B	4750
Working days/year	260

	Total Time	Actual Time	Support Time	Ratio Act./Tot.	Individ. Uses	Missions Required	Days Required
Op. Time - Existing	75	20	15	0.27	15000	3750	625
Op. Time - AWPS	195	120	35	0.62	2500	625	156

Cost	Variable	Level A				Level B			
		Existing SCBA		AWPS		Existing SCBA		AWPS	
		Suit	SCBA	Suit	PLSS	Suit	SCBA	Suit	PLSS
Purchase Price	I$	$400	$2,200	$____	$____	$25	$2,200	$____	$____
Uses per PPE Item	IU	1	1000	5	1000	1	1000	1	1000
Purchasing Hrs	HRS(P)		20		20		20		20
Purchasing Labor Rate	L$(P)		$25		$25		$25		$25
Purchasing Labor Cost			$1,202		$300		$1,202		$300
Total Purchasing Cost	P		$302,852		$88,738		$98,102		$374,363
PPE Uses	AU		750		125		14250		2375
Support Hours	HRS(O)		.25		0.58		0.25		0.58
Support Labor Rate	L$(O)		$25		$25		$25		$25
Consumables	C(O)		$1.35		$16.65		$1.35		$16.65
Operation Hrs	HRS(W)		1.25		3.25		1.25		3.25
Operation Labor Rate	L$(W)		$40		$40		$40		$40
Total Operation Cost	O		$43,200		$20,154		$820,800		$382,929
# Missions	AM		187.5		31.25		3562.5		593.75
Cons. Cost/Mission	CO		$75		$75		$75		$75
decontamination Hrs	HRSO		.292		0.667		0.102		0.201
Decontamination Labor Rate	L$(C)		$25		$25		$25		$25
Total Decontamination Cost	C		$19,544		$4,427		$303,503		$56,489
# Uses / Maint. Cycle	UMC		20		10		20		10
Consumables	C(M)		$25		$50		$25		$50
Maintenance Hrs	HRS(M)		0.475		1.310		0.131		0.301
Maint. Labor Rate	L$(M)		$25		$25		$25		$25
Total Maint. Cost	M		$1,383		$1,034		$20,154		$13,664
Total Storage Cost	S		$2,925		$2,063		$47,559		$27,253
Total Disposal Cost	D		$975		$688		$15,853		$9,084
LIFE CYCLE COST	T		$370,879		$117,103		$1,305,972		$863,782

Table 4 also provides estimates that show the increase in productivity for the AWPS. Since the assumption has been made that PLSS can provide at least 120 minutes of air and cooling, the actual time on site is six times higher than achieved with a conventional SCBA. Nevertheless, use of the AWPS is considered to require more support time. As a consequence, the best way to examine increases in productivity (separately from the life cycle costs) is to compare actual work times. On that basis the AWPS provides more than a two fold increase in worker productivity. Use of the AWPS allows completion of the 5,000 hour job in 156 days as opposed to 625 days for the existing protection technology.

LCC Summary

The most efficient way to define the feasibility of producing a new system is to determine if the initial cost of the system will benefit the user/industry in the long run. That is to say, the system being developed must give the end user a more productive system that will save time and money. Based on the LCC analysis, we feel the AWPS will provide significant cost benefits to the industry. The sensitivity analysis performed on the key variables shows continued cost benefits, even with the least favorable assumptions.

Future Activities

The performance criteria for the AWPS have been developed and Phase I has demonstrated the ability to meet them. These criteria were developed with both an understanding of the AWPS capabilities combined with the identification of DOE Decontamination and Decommissioning activities protection needs. The unique features of the AWPS will allow it to be used in environments and situations beyond the capability of current systems. Furthermore, a review of current DOE practices has identified several areas of concern which can be rectified with the introduction of the AWPS. The AWPS can reduce DOE inventory needs, reduce selection problems (by providing DOE with a single system that will be suitable for most needs), enhance personnel protection, safety, and provide significant gains in worker productivity.

We have identified, and feel we can meet, all applicable regulatory requirements with regards to the respirator, pressure vessel, protective suits, helmet, and support equipment. Regulatory requirements for DOE use, as well as those which will govern the use of the AWPS once in production, have had a bearing on the design of the AWPS. Compliance with these regulatory and similar requirements will be demonstrated through an extensive battery of testing, and certification of individual components by the appropriate authorities.

Visits to three key DOE remediation sites have yielded the information necessary to perform a cost benefit analysis, with positive results. A model was developed to determine the equivalent life cycle costs of various configurations of the AWPS. This information was used to compare the AWPS with systems currently in use. Using a specific scenario deemed representative of DOE D&D activities, the determination of life cycle costs has shown the AWPS will provide a savings of over $700K (based on a total life cycle cost of $1.7 Million) for a 5000 man hour job when compared to similar costs for existing protective clothing and equipment. This type of scenario is likely to be repeated many times over the 30 year period as the DOE undertakes specific D&D tasks at its various sites which require self-contained breathing air. Through a similar analysis, productivity gains of 100% or better were shown as well as lower potential medical and liability costs through better worker protection. Calculations of life cycle cost also demonstrated the preferred configuration for Level A clothing as being a more durable, reusable product.

The prototype hardware developed in Phase I has served to verify the design approaches for the AWPS, and gathered manned operations and integration information. The testing at KSU will provide the perform-ance data needed to optimize the Phase II hardware, as well as providing independant verification of the AWPS' effectiveness. Technical problems highlighted in the testing,particularly the weight of the backpack, are being solved in an inde-pendent research program nearing con-clusion at OSS. This work, along with Phase I, has brought the technical risk for Phase II of the AWPS project down to a low level.

Independent testing at the KSU's Institute for Environmental Research provided a comprehensive human subject evaluation of the AWPS showing significant increases in human subject endurance and comfort when compared to traditional SCBA-based ensembles. These tests also demonstrated continued performance of the PLSS without any major failure and efficacy of the liquid cooling approach when integrated with chemical protective clothing. The duration that human subjects were able to sustain as compared to the SCBA-based ensembles validated assumptions made in the life cycle cost study were major benefits are realized from increased extended mission performance and increases in productivity.

OSS is continuing the AWPS project into Phase II. The duration of Phase II is planned for 13 months and will consists of seven tasks -

Design Modification and Optimization - incorporate all advances from independent research and Phase I to design pre-production backpack and full scale support hardware for certification and site demonstration

Incorporation of Sizing Requirements into Suit Design - Design next generation of Advanced LCG and Splash Protective Garment (Level B), with multiple sizes. Design modifications to commercially available Vapor Protective Garment (Level A).

Full Scale AWPS Development - Fabrication and in house testing of hardware and garments.

AWPS Use Documentation Manuals - Produce Operator, Service, and Training Manuals.

Certification and Outside Evaluation of System / Components - DOT certification of the Vessel, NIOSH certification of the breathing system, DOT (LANL) evaluation of protective garments.

Production Version AWPS Development Topical Report - Topical report on Phase II tasks and test plan for site demonstration.

DOE Site Demonstration - Field tests at a DOE site (to be determined).

At the completion of Phase II, OSS is committed making the NIOSH certified AWPS available in the commercial market, and continues to invest company resources towards developing that market potential. In addition, OSS continues to support internally funded research and development of other hostile environment life support products related to the AWPS.

Acknowledgements

The AWPS project is supported through the Department of Energy's Morgantown Energy Technology Center, as a cost-sharing research and development contract with Oceaneering Space Systems, and is conducted under the Environmental Management's Decontamination and Decommissioning Focus Area. Oceaneering Space Systems would like to thank Steve Bossart of METC for his technical management of the effort.

3.2 Characterization of Radioactive Contamination Inside Pipes with The Pipe Explorer™ System*

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Introduction

The U.S. Department of Energy's nuclear facility decommissioning program needs to characterize radiological contamination inside piping systems before the pipe can be recycled, remediated, or disposed. Historically, this has been attempted using hand held survey instrumentation, surveying only the accessible exterior portions of pipe systems. Difficulty, or inability of measuring threshold surface contamination values, worker exposure, and physical access constraints have limited the effectiveness of this approach. Science and Engineering associates, Inc. under contract with the DOE Morgantown Energy Technology Center has developed and demonstrated the Pipe Explorer™ system, which uses an inverting membrane to transport various characterization sensors into pipes. The basic process involves inverting (turning inside out) a tubular impermeable membrane under air pressure. A characterization sensor is towed down the interior of the pipe by the membrane.

Advantages of this approach include the capability of deploying through constrictions in the pipe, around 90° bends, vertically up and down, and in slippery conditions. Because the detector is transported inside the membrane (which is inexpensive and disposable), it is protected from contamination, which eliminates cross-contamination. Characterization sensors that have been demonstrated with the system thus far include: gamma detectors, beta detectors, video cameras, and pipe locators. Alpha measurement capability is currently under development.

A remotely operable Pipe Explorer™ system has been developed and demonstrated for use in DOE facilities in the decommissioning stage. The system is capable of deployment in pipes as small as 2-inch-diameter and up to 250 feet long. This paper describes the technology and presents measurement results of a field demonstration conducted with the Pipe Explorer™ system at a DOE site. These measurements identify surface activity levels of U-238 contamination as a function of location in drain lines. Cost savings to the DOE of approximately $1.5 million dollars were realized from this one demonstration.

Research sponsored by the U.S. Department of Energy's Morgantown Energy Technology Center, under contract DE-AC21-93MC30172 with Science and Engineering Associates, Inc. 6100 Uptown Blvd. NE, Albuquerque, NM, 87110; telefax: (505) 881-7420

* Patent Pending

Problem

By their nature, the interiors of pipes and ducts are difficult to access. In many cases, even the exteriors are inaccessible. For example, drainlines are buried or encased in concrete and duct work is often elevated or enclosed. To access these structures for characterizations such as radiological surveys, requires significant effort and cost. These costs are further increased if the characterizations are carried out in a radiological control zone, where greater personal protective measures and support crews are required.

Furthermore, for alpha and beta emitting contaminants, such as U-238 and Pu-239, it is necessary to take unobstructed measurements of contaminated surfaces. Thus, external measurements through pipe walls are inadequate and the only way to gather data is to get an instrument inside of the pipe.

Alternative methods to the Pipe Explorer™ system can be used to transport detectors into pipes, such as pipe crawlers and push rods. However, these methods lead to ambiguous results if there is removable contamination present. With nothing to prevent contamination from getting on the detector there is no way to differentiate between contamination on the pipe wall and contamination on the detector. There are additional limitations associated with these alternative methods. For example, pipe crawlers are typically limited to larger diameter pipes (> 4 inches). They are also cumbersome to operate around elbows and have a difficult time in pipes with slippery surfaces. Push rod methods are limited in length and are often unreliable when trying to get a detector around elbows.

Solution

As a solution to this problem, SEA adapted its inverting membrane technology to transport radiation detectors and other characterization tools into pipes. The system uses an air-tight membrane configured so that when it is pressurized it inverts into a pipe. As it inverts the pressure force on the end of the membrane is adequate to tow a detector around multiple elbows and through several hundred feet of piping. This technology not only provides an effective transportation method for detectors, but it also provides a clean conduit through which the detector can travel.

Technology Description

The primary components of the Pipe Explorer™ technology are illustrated in Figure 1. The heart of the system is an air-tight membrane which is initially spooled inside of a canister. The end of the membrane protruding out of the canister is folded over and attached to a basepipe. When the canister becomes pressurized in this configuration, the air pressure on the membrane causes the membrane to be pulled from the spool. This continues until the membrane is completely off the spool. A characterization tool such as a radiation detector is attached to the end of the membrane and towed into the pipe as the membrane continues to invert. The detector cabling is also towed into the pipe from the spool. To retrieve the system from a pipe, the process is simply reversed, where the cabling, detector, and membrane are wound back onto the spool. The system can thus be used to move a detector freely back and forth through a pipe while the detector output and position are continuously recorded. As a result, the Pipe Explorer™ system provides high resolution analysis of the location of radioactive contamination in pipes.

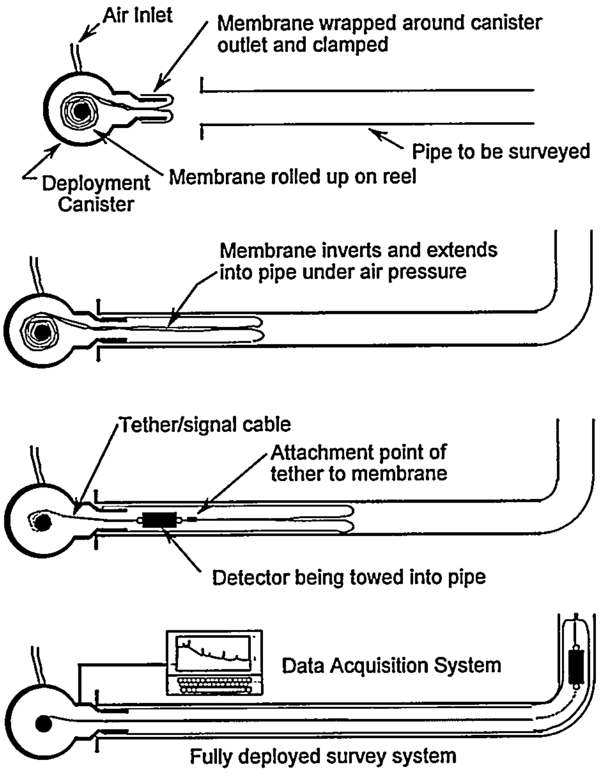

Figure 1. Sequence of membrane and detector deployment with the Pipe Explorer™ system.

The membrane also provides a clean conduit through which the detector travels. This protects both the detector and the workers handling it. Furthermore, measurements are inherently more reliable. A detector transported in any other fashion runs the risk of removable contamination adhering to the sensor, which can cause erroneously high or false positive readings.

The general operating procedure is to first deploy the membrane halfway into the pipe. This is the point where the detector begins to enter the pipe from the deployment canister. At this time data acquisition is initiated. In most cases the detector is deployed out relatively quickly (up to 30-ft/min). More detailed radiological measurements are taken as the detector is retrieved from the pipe at a slower rate.

As the detector is being retrieved, the tether is wound back into the deployment canister. The membrane prevents contamination from contacting the tether. However, as a precautionary measure, two sampling smears are used to swipe the entire surface of the tether and the detector. When the tether is completely retrieved the smears are surveyed with a pancake GM probe to ascertain if any contamination has potentially been transferred into the canister. To date, no contamination of the canister or tether has been noted. Once the detector has been retrieved and the survey completed (the detector can be re-deployed for additional data if needed), the detector is removed from the end of the membrane. The membrane is then fed through a diaphragm to an external reel assembly or manually fed into a disposal drum. The membrane being handled has been inverted. Therefore, the side of the membrane that has been in contact with the contaminated pipe is contained within itself (this is analogous to the way a Hazmat worker removes rubber gloves). The inexpensive membrane (about $0.03/ft) is then disposed. This secondary waste generation is minimal. Several hundred feet of membrane is easily compacted into less than a cubic foot.

Capabilities Summary

The absolute maximum deployable distance of the system is currently limited by the length of cabling and canister size. The current configuration allows for 250-foot deployments. Longer distances may be achievable but no applications to date have required any longer attempts. Practical deployment lengths are limited by elbows in the lines and the diameter of the pipe. Table 1 lists typical results that have been achieved, in laboratory tests, and are used as general guidelines.

The Pipe Explorer™ system has been used to transport several different types of radiological measurement instruments. Table 2 lists these instruments and their descriptions.

SEA currently has two deployment systems available. The first is a fully automated system. With its motorized operation and built in deployment sensors it allows for continual unattended pipe surveys. The second system is a smaller, manually operated system.

Additional uses of the Pipe Explorer™ have been identified and have either been nominally demonstrated or are being integrated with the system. These include;

- Transport of pipe locating beacons
- Transport of video cameras
- Alpha detection methodologies

Table 1. Typical Deployment Lengths and Number of Elbows for Various Pipe Sizes.

Pipe Diameter (inches)	Number of 90° Elbows	Maximum Deployed Distance (feet)
1	0	50
2	2	200
3	4	250
4	4	250

Table 2. Radiological Instruments Used with the Pipe Explorer™ System.

Detector Type	Detection Mode	Notes
Bicron BC-404 Plastic Scintillator 1.25 inch x 1.95 inch	Beta	Large window offers high sensitivity beta detection. Compact package allows transport around 2-inch elbows.
Bicron BC-408 Plastic Scintillator 0.5 inch x 0.5 inch	Beta	Ruggedized packaging good for applications in pipe sizes 3-inches and up.
NaI(Tl) 2-inch x 2-inch crystal size	Gamma	Large crystal provides high sensitivity and good spectral resolution. Larger package size limits applications to pipe sizes greater than 4 inches.
CsI(Na) 1.125-inch x 1.188-inch crystal size	Gamma	Small package allows transport around elbows in 2-inch pipe.

Results

An extensive demonstration of the Pipe Explorer™ was conducted for the DOE Formerly Utilized Sites Remedial Action Program (FUSRAP) at a site in Adrian Michigan. During the 1950's the Bridgeport Brass Company operated a Special Metals Extrusion Plant at the site. This was done under contract with the DOE, then the Atomic Energy Commission. The product of this operation was material for uranium fuel elements for reactors in Hanford, Washington, and the Savannah River Plant in South Carolina. Uranium handled in this operation included depleted, natural, and up to 2.1 percent enriched in U-235. The site is still an active factory where plastic automobile parts, such as door panels and dashboards, are extruded and finished.

During production of the uranium fuel elements, waste material from the extrusion process mixed with oil from the machinery. This mixture subsequently flowed into the oil drainage system contaminating over 1000 feet of buried drain-lines with varying amounts of uranium tainted oil. In order to quantify the extent and degree of this contamination and to conduct post-remediation measurements, the DOE FUSRAP hosted a demonstration of the Pipe Explorer™ system.

SEA conducted surveys at the site on two separate occasions. The first occurred in April 1995 and the second in May 1995. Thirteen surveys were carried out in eight drain-lines. Several lines were surveyed more than once to confirm success of remedial actions. Two Pipe Explorer™ deployment systems were used with 3 different radiological sensors. The first system used during the April demonstration was a manually operated system. Deployment with this system is controlled by a hand crank. Figure 3 shows the system in operation at the site. With this system, the detector is deployed to a specified location where the position of the detector and its output are recorded by the operator. Figure 4 shows data from one of the surveys conducted with the manually operated Pipe Explorer™ system in conjunction with a beta detector. The data was taken prior to any remedial actions. Thus, the drain-line had a substantial amount of thick oily sludge in it (about the consistency of peanut butter). The detector and its tether were successfully deployed and retrieved with none of the oily contamination coming into contact with the detector, tether, or workers. The data in Figure 4 was obtained with a detector designed and calibrated by the DOE Grand Junction Projects Office Radon Laboratory (Reference 1).

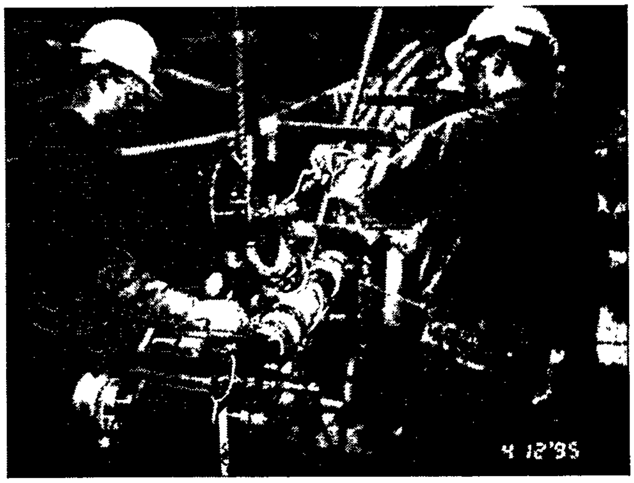

Figure 3. Operation of the Pipe Explorer™ system at the FUSRAP site. The membrane is being retrieved from a drain-line.

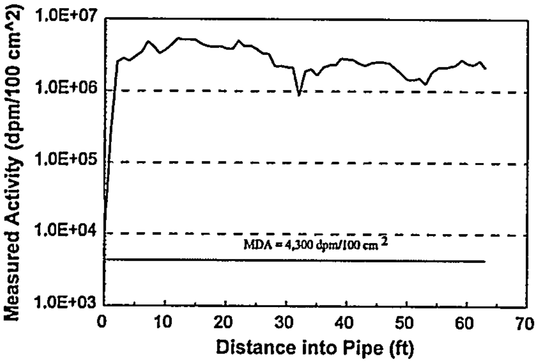

Figure 4. Surface activity measured in a 4-inch drain-line at the FUSRAP site with the manually operated Pipe Explorer™ system. These measurements were taken prior to removal of contaminated sludge from the drain-line.

For the second stage of the demonstration carried out in May 1995, the automated Pipe Explorer™ system was used with a higher sensitivity beta detector. The system canister includes a motorized reel and a deployment distance measurement sensor. Additional sensors in the canister such as a slack indicator, a tension meter, and pressure transducers enable the system to run with minimal operator interaction. All outputs from the sensors are displayed on a control panel. In addition, they are recorded and displayed on a laptop computer acting as a virtual instrument through a LabView® program. The radiological data is also recorded on the laptop so that surface activity as a function of distance into the pipe can be monitored in real time. Figure 5 shows the automated system in use at the FUSRAP site.

A sample of the data obtained with this system is shown in Figure 6. The actual drain-line begins at a distance of 27 feet. Since access to the drain-line was obtained through a deep manhole it was necessary to construct a conduit of this length to guide the membrane to the drain-line entrance. The structure of this data shows the utility of a continuous survey. The data shows a small amount of contamination up to the 40-foot mark in the drain-line. At this point the line intersects another drain-line which had been thoroughly cleaned. After the intersection, however, substantial contamination was encountered. The only exception was a relatively clean section between 90 and 100 feet.

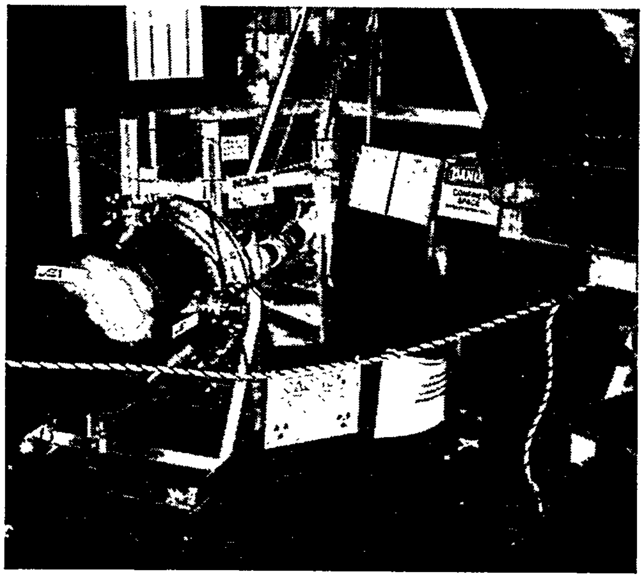

Figure 5. The fully automated Pipe Explorer™ system in use at the FUSRAP site. The deployment canister is on the floor to the left and the operator and control box are on the right. Note that the system is located outside of the radiological control zone.

<u>Confirmation of the Data</u>

Data obtained with the Pipe Explorer™ system at the FUSRAP site was verified with several methods. The first was purely qualitative, where the membrane was visually inspected as it was retrieved from the drain-line. This was useful in such instances as shown in Figure 6 where the data showed significant structure. For example, a large amount of the oily sludge was noted on the portion of the membrane that had traveled 100 to 120 feet into the drain-line. The portion of the membrane around 98 feet had virtually no oil on it, but below 90 feet substantial amounts of the oily sludge were again seen on the membrane.

Another validation method used was to measure the activity of contamination adhering to the membrane as it was being retrieved. Measurements were taken with a conventional pancake GM probe. This data is shown as triangles in Figure 6. The distance accuracy for these measurements is substantially less than the accuracy of the Pipe Explorer™ data (pancake meter data accurate to approximately ±2 feet, Pipe Explorer™ accurate to ±1 inch). Surface activity measured with the Pipe Explorer™ is consistently higher than that measured with the pancake GM probe because the Pipe Explorer™ system measures the contamination in the pipe and the pancake GM probe measures only the contamination that adheres to the external surface of the membrane. Furthermore measurements with the pancake probe are not calibrated for attenuation effects of the membrane, whereas the data obtained by the Pipe Explorer™ system is.

Confirmation of the data was also attempted by pushing a small GM detector into the drain-line. However, contamination adhering to the GM probe assembly tended to obscure the measurement of contamination on the pipe wall.

Detector Calibration

The ideal way to confirm the Pipe Explorer™ system data would have been to excavate a portion of a drain line and have it analyzed. However, the motivation for using the system at the FUSRAP site was to avoid excavating drain-lines. Therefore, confidence in the data was obtained through rigorous calibration of the detector.

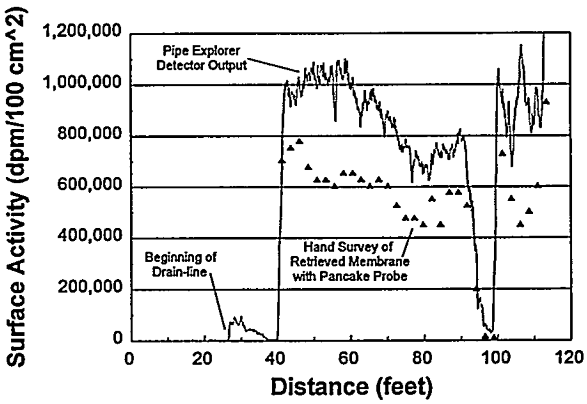

Figure 6. Survey of a drain-line at the FUSRAP site with the automated Pipe Explorer™ system (solid line). Triangular data markers show measurements of contamination on the membrane retrieved from the drain-line.

Detectors used with the Pipe Explorer™ system are specifically calibrated for each use. They are calibrated with an isotope of similar energy of the contaminants that are suspected in a pipe and calibrated in the same measurement geometry. For example, since U-238 was suspected at the FUSRAP site, Sr-90 was used as a calibration source (U-238 is not available in sufficiently high activities for calibrations). The daughter product of Sr-90 (Y-90) emits a beta particle with similar energy as the dominant U-238 daughter product, Pa-234m. The Sr-90 calibration source has an known activity traceable to the National Institute of Standards and Technologies. Using this calibration source results in slightly elevated detection efficiencies because of a lower energy beta emitted by Sr-90 (546 keV max.). This emission is more heavily attenuated by air and the membrane material than the higher energy beta from Y-90, but no effort was made to determine this difference.

The significant added cost of assessing this effect on the calibrations was not deemed necessary, since the error was not considered significant (on the order of 20 percent) and it results in conservative measurements.

The calibrations were carried out to best simulate the measurement conditions that would be encountered at the FUSRAP site, where the detector rests on the bottom of a 4-inch pipe inside of a 4-mil polyethylene membrane. Therefore, all of the calibration measurements were made through a sample of the membrane material in 4-inch pipe. The fundamental procedure used in the calibrations was to move the calibration source to various grid locations surrounding the detector and determine the probe response at each location. The response of the detector to the Sr-90/Y-90 source was integrated over all angular and axial positions to determine detector response to distributed contamination inside of 4-inch pipes. The response of the detector to a check source in a fixed geometry was recorded immediately before and after the detector calibrations. The check source measurement was repeated prior to and after each drain-line survey at the FUSRAP site to verify the detector performance had not changed since the calibrations.

Benefits

The use of the Pipe Explorer offers many technical benefits. These include;

- 100% gamma and beta surveys of pipe interiors, even in buried pipes.
- 100% alpha surveys of pipe interiors (available soon)
- Detector does not become contaminated
- Removable contamination is not spread along pipe.
- Personnel exposure significantly reduced.
- Immediate results.

Technical benefits such as the ones listed above for the Pipe Explorer^{TM} are usually heralded as the pay-off for a DOE investment in a new technology. However, the primary reason the DOE provides funding for development of environmental technologies is so that economic benefits will result through more expedient and cost effective methods. Substantial cost savings have already been realized from use of the Pipe Explorer^{TM} system at the FUSRAP site demonstration. **These cost savings to the DOE are nearly three times the amount invested in the development of the Pipe Explorer^{TM} system.**

The DOE FUSRAP recognized that the cost of excavating buried drain-lines at the site in Adrian MI would be substantial. Therefore, they developed a methodology to avoid these excavation costs. The plan was to verify that activity levels of contamination in the pipes were below a criteria level of $7x10^5$ dpm/100cm^2 (averaged over the length of the drain-lines). It was determined through a hazard assessment that such levels of contamination posed no threat to the general population. Thus, the drain-lines could be left in place after sealing the contamination with grout. If surface activities were found in excess of the criteria level then the drain-lines were to be flushed and cleaned prior to grouting.

The initial method that was used to characterize the drain-line was to insert a small geiger-mueller (GM) detector directly into the drain lines. This was soon found to be an ineffective method because of the abundance of removable contamination present. The contaminated oil would adhere to the detector, making it difficult to differentiate between measurements of contamination on the pipe walls and contamination directly on the detector. In addition, only limited lengths of the drain-lines could be accessed since in many cases the detector could not be shoved around elbows.

The benefits of using the Pipe Explorer™ over direct insertion of a detector were readily seen in surveys of one of the drain-lines. Data initially obtained with the manually operated Pipe Explorer™ system from this drain-line is shown in Figure 6. Activity levels in the pipe were found in excess of the $7x10^5$ dpm/100cm^2 criteria level. The drain-line was then cleaned and a subsequent survey was conducted. Activity levels were found substantially reduced with the exception of a hot spot near the beginning of the drain-line. A detector manually inserted into the pipe would have come into contact with this hot spot and measurements through the rest of the drain-line would have been inaccurately high. Therefore, the Pipe Explorer™ system provided accurate results showing that the drain-line was within the criteria level. Similar results were obtained in the other seven drain-lines surveyed.

Had accurate data not been available from the Pipe Explorer™ system there would have been no way to assess activity levels in the drain-lines. Therefore, it would have been necessary to excavate them. It is estimated that the costs to excavate the drain-lines would have been on the order of $1.2 million (Ref. 2). However, this estimate neglects the fact that the site is an active automotive parts factory. Therefore, costs associated with plant impacts and relocating factory operations should also be included. Factory personnel have good estimates of these costs from prior experiences of modifications to the plant. Their estimate of these costs are about $0.8 million. The cost savings were diminished somewhat by the expense of cleaning the drain-lines and disposing of the waste generated from the cleaning. This cost is estimated at $0.5 million. Therefore, the net savings is estimated to be;

$1.2 +$0.8 -$0.5 = $1.5 million

It is interesting to note that the cost of surveying excavated drain-lines with the Pipe Explorer™ system was included in the excavation cost estimate. This was done since characterization of waste is necessary prior to disposal. Therefore, whether the drain-lines were left in place or excavated, the FUSRAP remediators identified a need for the Pipe Explorer™ system.

Future Activities

The development of the basic Pipe Explorer™ system which includes gamma and beta detection capability is nearing completion. The final aspect of this phase of development is to demonstrate the system at ORNL during October 1995. Video inspection capability of the system will be demonstrated along with radiological surveys.

After this time the system will be available for service work as an inspection tool. A great deal of interest has already been expressed in using the system at;

- Rocky Flats
- Los Alamos National Laboratory
- Sandia National Laboratory
- Other FUSRAP Sites
- and Argonne National Laboratory

In July of 1995 an the development of an enhancement to the system was funded by the DOE METC. This will enable the system to be used for detecting low levels of alpha emitting contaminants such as Pu-239. This will be accomplished by making the inverting membrane component of the system an alpha sensitive scintillator. A photo-detector, towed through the membrane, much the same way as gamma and beta detectors, will quantify activity levels as a function of length over 100% of the internal surface area (for more information see related paper in these proceedings). After this enhancement is added to the system, complete alpha/beta/gamma surveys will be possible with the Pipe Explorer™ system.

Acknowledgments

SEA wishes to thank Mr. C. Edward Christy of METC for his diligent and enthusiastic support as COR of the Pipe Explorer™ system development. SEA also wishes to thank Mr. Al Tardiff of EM-50 who has helped in finding demonstration sites for the system. We also thank Mr. James D. Kopotic of the FUSRAP for the agreeing to host the demonstration of the Pipe Explorer™ system. Finally we wish to thank Mr. Marty Keller and Mr. Gil Drexel of Bechtel along with the site crew at the FUSRAP site who were all extremely helpful in the successful demonstration of the system.

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3.3 Laser-Based Coatings Removal

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Needs

Over the years as building and equipment surfaces became contaminated with low levels of uranium or plutonium dust, coats of paint were applied to stabilize the contaminants in place. Most of the earlier paint used was lead-based paint. More recently, various non-lead-based paints, such as two-part epoxy, are used. For D&D (decontamination and decommissioning), it is desirable to remove the paints or other coatings rather than having to tear down and dispose of the entire building.

Problems with Other Coatings Removal Technologies

Table 1 gives a summary matrix that compares the various technologies for coatings removal. There are seven important factors to consider in this comparison.

- Waste volume: Radioactive waste storage accounts for ~33% of the cost of D&D. The DOE uses an average number of $300 per cubic foot for storage, disposal, and monitoring. Thus any reduction in waste volume results in a big cost savings. Sand blasting uses about a hundred pounds of sand to remove one pound of coating, and the sand becomes contaminated waste. Since using liquids generally results in radioactive-contaminated liquid wastes, and using chemicals generally results in mixed hazardous waste, it is highly preferred to avoid both liquids and chemicals. Dry ice pellet blasting does not add to the volume. Far-infrared laser light reduces the volume of coatings that contain hydrocarbons, such as lead-based and epoxy paints. We project waste volume reductions of 75% of the original paint volume.

- Cleaning out the surface pores: Only laser light does this effectively.

- Thermal damage to the substrate: Devices like CW (continuous wave) lasers can cause thermal damage,. In fact, CW lasers are available commercially for cutting metals. However, a pulsed-repetition laser can be designed to remove coatings faster than a thermal wave can propagate into the substrate, resulting in no thermal damage. This can increase the resale value of cleaned metal by a factor of nine from ~3¢ per pound for smelter feedstock to ~27¢ per pound for resale and reuse.

- Mechanical damage to the substrate: The chart shows five technologies that cause no mechanical damage. Again, this can increase the resale value of cleaned metal.

- Hazardous chemicals: In closed areas, the operators must wear breathing apparatus when using dry ice pellets or liquefied nitrogen. Otherwise, these technologies do not result in health hazards since the carbon dioxide or nitrogen evaporate into the air. Chemical strippers and strippable coatings can generate mixed hazardous wastes.

- Liquids: Both water blasting and liquid chemical strippers involve liquids that generally require wet-chemistry processing of residuals, such as the sludge from sodium-bicarbonate / air blasting.

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* Research sponsored by the U.S. Department of Energy's Morgantown Energy Technology Center, under Contract DE-AR21-94MC30359 with F2 Associates Inc., 14800 Central Avenue SE; Albuquerque, NM 87123; telfax: 505-271-1437.

Table 1
Comparison of Contaminated Coatings Removal Technologies

	Waste Volume			Cleans out surface pores	No thermal damage	No mechanical damage	No hazardous chemicals	No liquids	Level-D dress
	Increase	Same	Decrease						
Mechanical scabbling		√			√		√	√	
Solid abrasives or air blasting	√				√		√	√	
Dry ice pellet blasting		√			?	√	√	√	CO_2 atmos
Water blasting	√				√				
Liquid nitrogen cryofracture		√			?	√	√	√	N_2 atmos
Wet chemical strippers	√				√	√			?
Dry strippable coatings	√				√	√		√	?
CW lasers		√	√	√			√	√	√
Pulsed-repetition lasers			√	√	√	√	√	√	√

- Level of worker dress: Unless operations are performed robotically, only pulse-repetition laser systems, with the prompt capture of ablated material, enable dress at Level D (as shown in Figure 1). The lowest possible level of dress will keep operations costs down.

Solution:
Pulse-Repetiton Laser Systems

As can be seen in Table 1, pulse-repetiton lasers satisfy all of the desirable criteria. Although this technology does not remove in-depth contamination, such as chemicals that have migrated into concrete, the concept is to first remove the paint and surface contamination, and then determine if any scabbling is even needed.

Technology

General

To avoid substrate thermal damage, the time that each pulse lasts must be very short. With the appropriate pulse length and with laser power densities on target approaching a megawatt per square centimeter, coating material can be ablated faster than heat can propagate into the substrate. For any coatings that have hydrocarbons in them, a carbon-dioxide (CO_2) laser works best, since the far infrared wavelength of the laser light couples very well into hydrocarbon bonds. This "tuned" chemical-bond breaking is more sophisticated than simply putting heat energy on target. Data indicates that binders such as the linseed oil in lead-based paints are reduced to water vapor and carbon

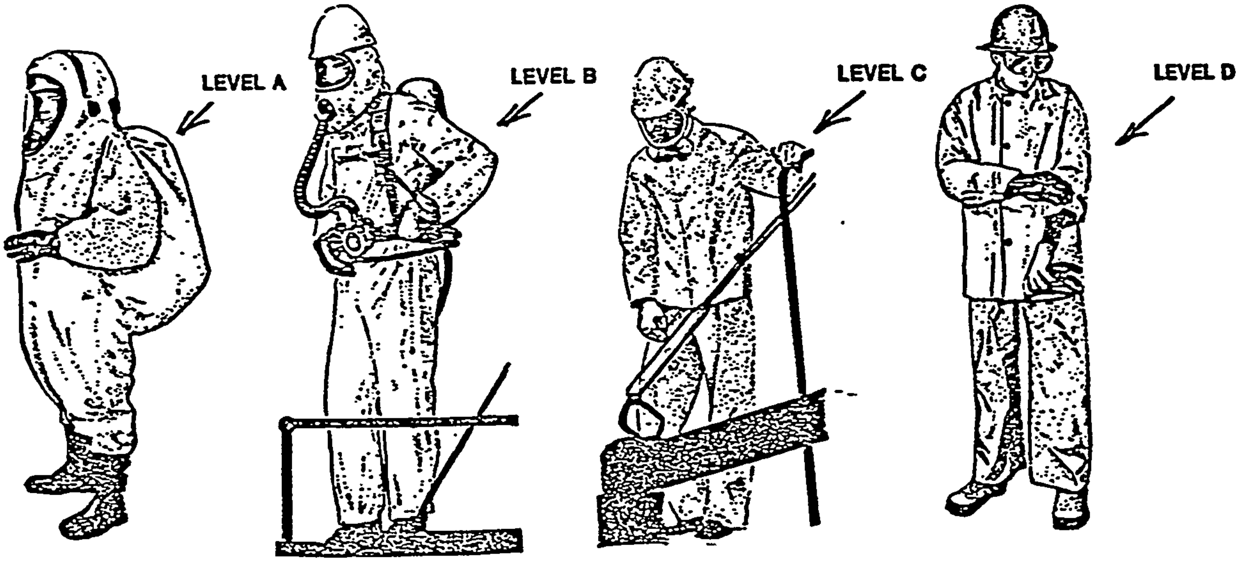
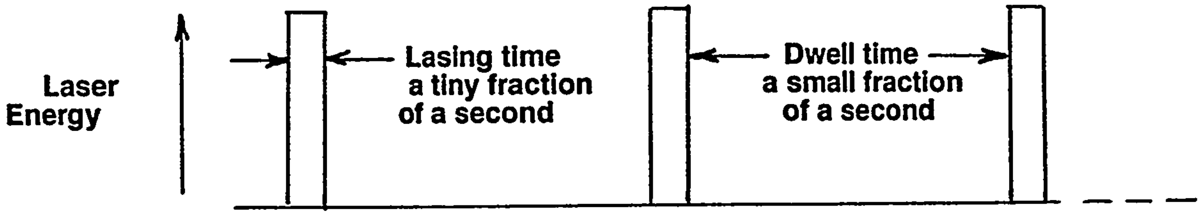
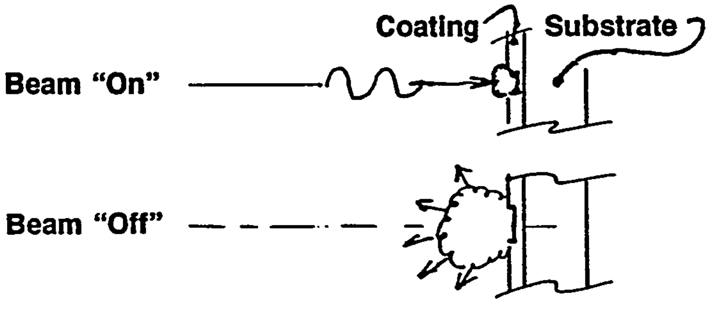


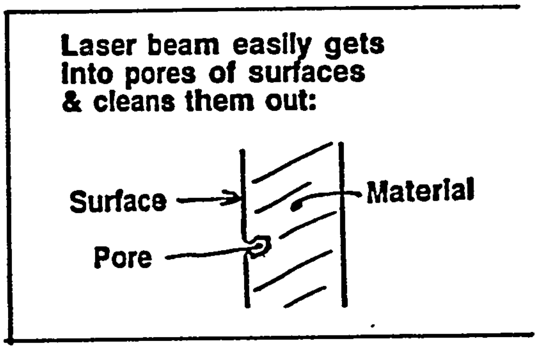
Figure 1
System Design Goals Targeted at Level D Protection During Operation
(Equipment Control Buttons, Switches, and Levers are Designed For Worst Case - Level A).



Lasing time pulse width must be 'just right'; if too short then little stripping, & if too long then excessive substrate heating.

Dwell time must be 'just right'; if too short then there is interference between the next pulse and the debris could from the last pulse; if too long, then a slow process.





Laser beam easily gets into pores of surfaces & cleans them out:

USE SPECIAL VACUUM NOZZLE TO SUCK AWAY ABLATED MATERIAL / DEBRIS CLOUD

Figure 2
Coatings Removal With Pulsed Lasers

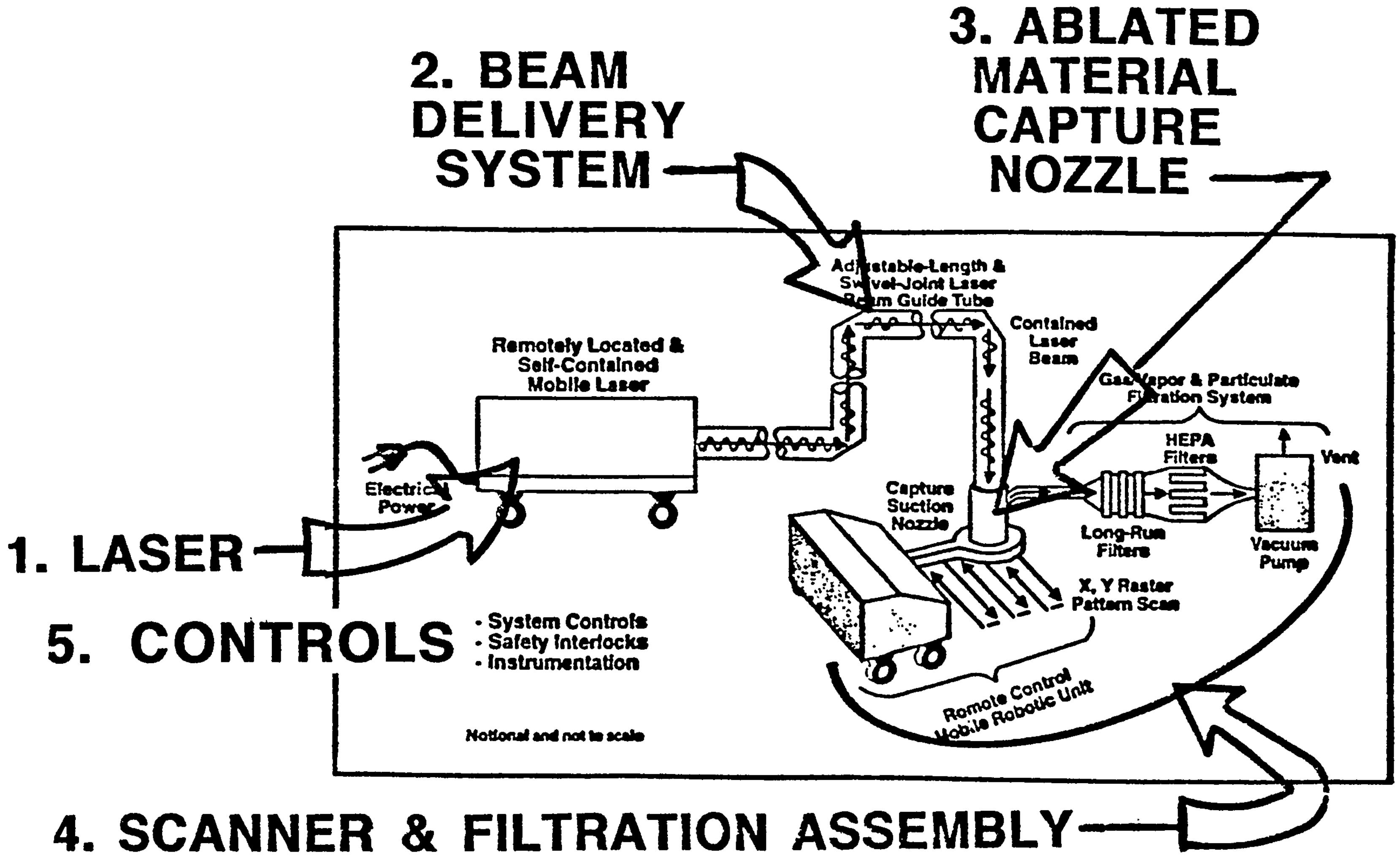


Figure 3
The Five Basic Elements of a Laser-Based Coatings Removal System

dioxide gas when the right power densities and pulse widths are used. For other coatings such as cadmium on steel, a shorter wavelength laser may work more efficiently in terms of the physics, but may not be as cost-effective as other lasers such as efficient CO2-gas lasers.

In a pulse-repetition system, the time between pulses must be long enough to clear (vacuum away) the cloud of ablated material (see Figure 2). Otherwise, the cloud may absorb and / or defocus the next pulse. However, the pulse-repetition rate must also be fast enough, and the spot-size on target must be big enough, to yield reasonable cleaning rates.

Floor and Wall Cleaning

As shown in Figure 3, there are five basic elements to a laser-based cleaning system.

1. Remote laser. This could be located in adjacent room or outside. Nd:YAG-crystal pulse-repetition lasers are commercially available, but not yet with the power for faster cleaning of large surfaces. Also, the near infrared wavelength does not couple into hydrocarbon binders quite as well as that of CO_2 lasers. For our system, we have chosen a high-power pulse-repetition CO_2 laser (see Figure 4). We have found only two suppliers of the such a laser in the world, both in Albuquerque, NM. Tetra Corporation is our supplier. Their laser is transportable, EMI shielded, and weather-proof. It needs only electrical power since it has its own chillers with air heat exchangers.

2. Laser-beam delivery system (BDS). This transports the beam from the laser to a cleaning head. Good fiber optics do not exist

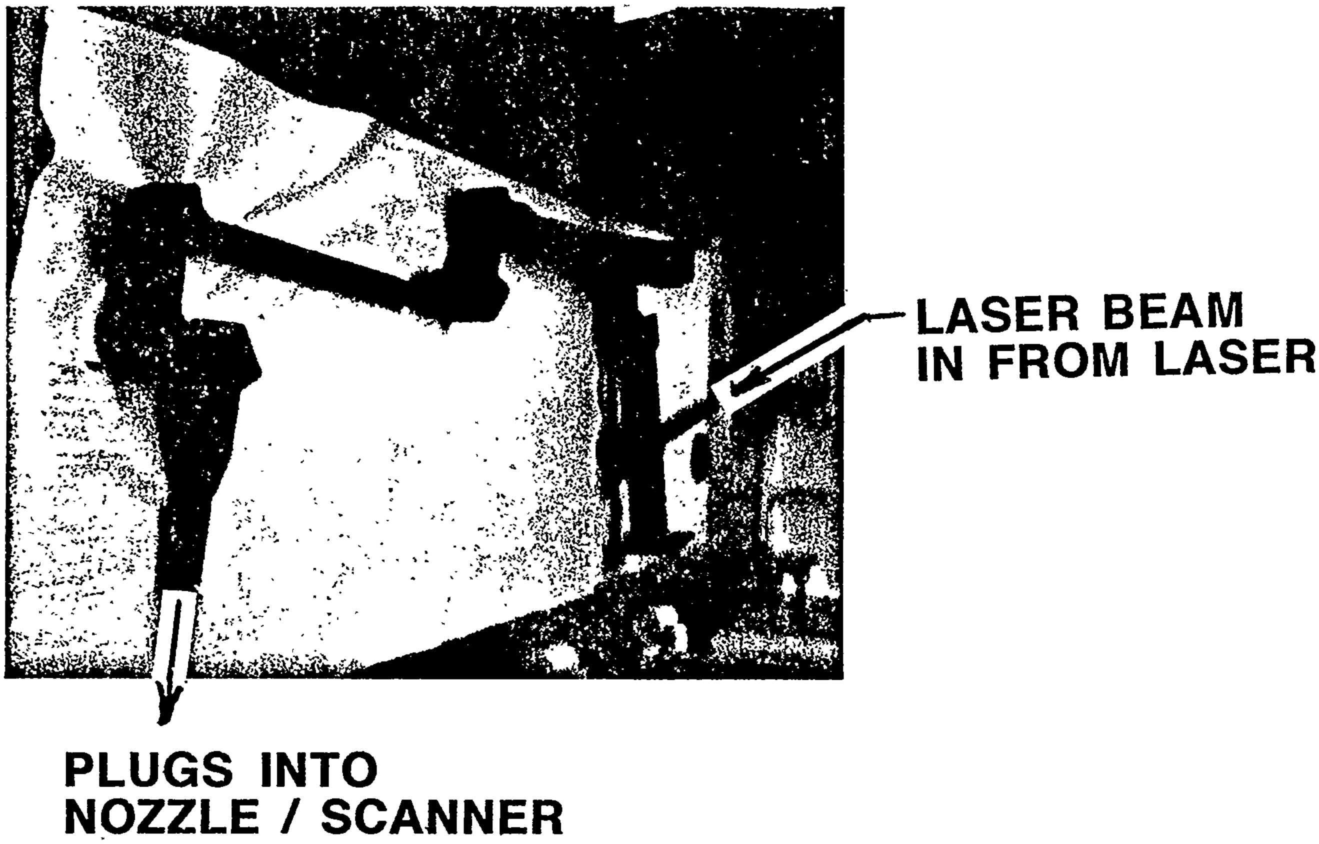


Figure 4
The Laser Beam Delivery System

for the far infrared where CO_2 lasers emit, so CO_2-laser beam delivery is done with rigid beam tubes fitted with corner mirrors in swivel joints, to deliver the beam through "articulating optics" to a cleaning head (see Figure 5). Work at Rutgers University on flexible hollow tubes coated on the inside with artificial-sapphire can also be used for flexible beam delivery from CO_2 lasers up to 2 kW CW, but not yet at the power levels of up to 6 kW average (higher peaks) that we require.

3. A cleaning head. This has optics to deliver the beam on target and promptly capture all ablated particulates, gases and vapors. The cleaning head will be located on a remotely operated scanner attached to the side of a Pentek VAC-PAC (see Figure 6). The scanner will also automatically maintain proper stand-off distance between the nozzle base and surface being cleaned. This will allow air in for dilution and cooling of ablated material, but keep any ablated material from escaping. For less delicate substrates, the 2

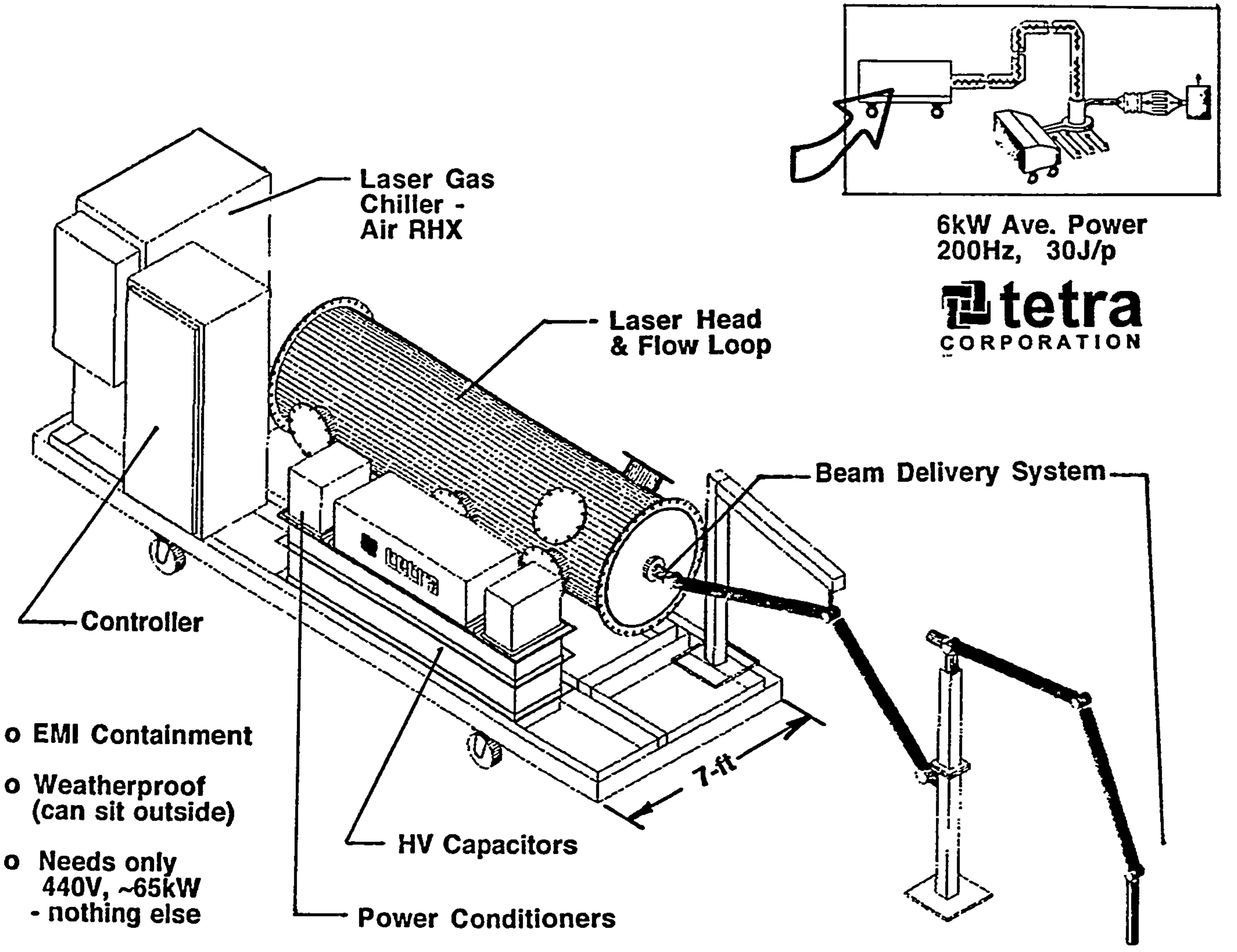


Figure 5
Transportable, Pulsed-Repetition CO_2 Laser

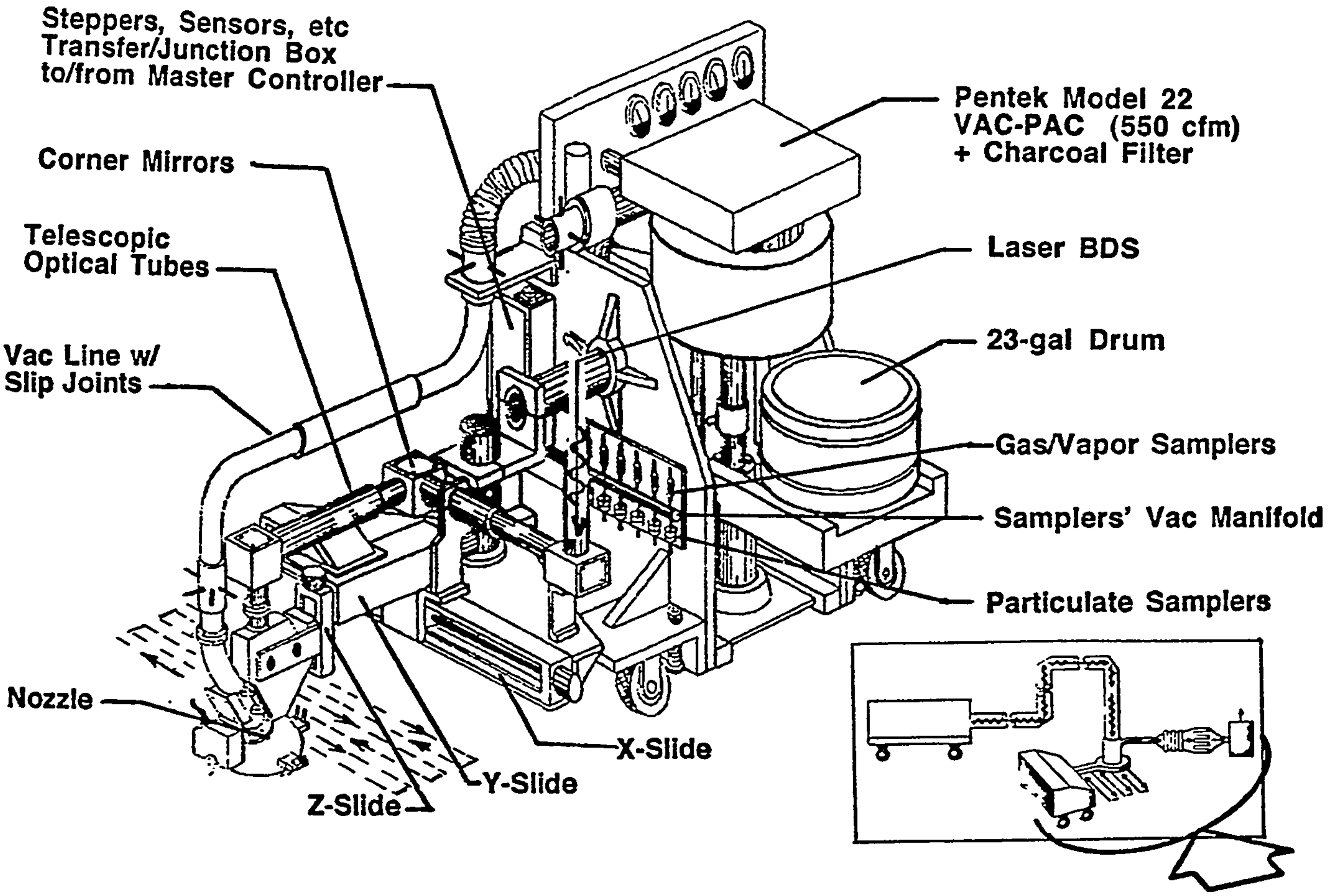


Figure 6
Nozzle, Scanner, and Filtration Assembly

centimeter by 2 centimeter square laser-pulse spots can have a high percentage of overlap during a lateral scan, ~95%. The scanner is designed to eventually be mounted on a MOOSE robot for floor cleaning, and a ROSIE robot for wall cleaning.

4. A filtration system with primary and HEPA particulate filters, and charcoal filters for gases and vapors. The on-line recleanable particulate filters are recleaned with periodic blow-back pulses of air. The system deposits the particulates directly into a 23 or 55 gallon drum for final disposal, thus requiring no further container transfers. These drums can be sealed in-line, with no worker exposure. We are using the Pentek VAC-PAC. With such a filtration system there are minimal residuals requiring any chemical processing.

5. Sensors, safety interlocks, and controls, all interconnected via a 486 computer (see Figure 7). The master controller records data from various places in the system on temperatures, pressures, and flow rates. The data is recorded and is also used in logic trees. For example, a growing pressure differential across a filter would indicate onset of clogging, or a drop to zero would

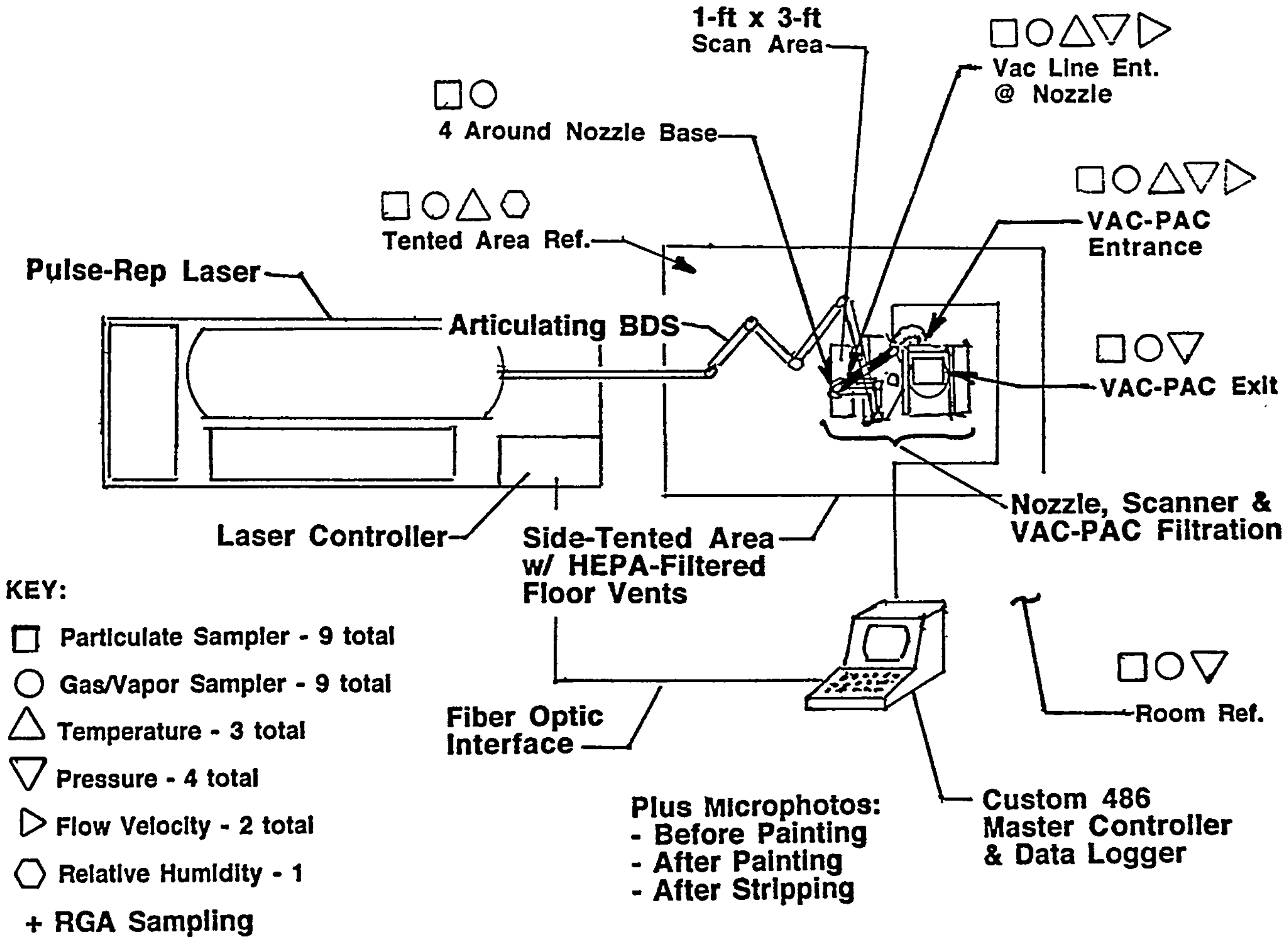


Figure 7
Phase II Test Layout, Instrumentation, and Controls

indicate a filter blow-out, triggering an orderly shut-down procedure. The controller also controls the firing of each pulse of the laser. For Phase II testing, we are also collecting data with gas / vapor and particulate sniffers at various places in the system, and gas / vapor samples using a resdual gas analyzer, plus test-cell relative humidity readings.

In addition, we are working towards adding on-line sensors such as a radioactive sensor, paint-thickness sensor, optical comparitor, and optical spectrometer to analyze the plasma plume (see Table 2). These can be used for feedback and control to the scanner speed and laser pulse rate. The spectrometer can provide an on-line assay of what is going into the drum. Then when a drum is full, the computer would print an integrated assay label to be attached to the drum, so it would never need to be opened again.

Thus, though the laser is an important subsystem, proper <u>system integration</u> is the key for operational performance, cost savings, and acceptability in teerms of the environmental, safety, and health aspects.

Parts Cutting and Cleaning

When a building undergoes D&D, the equipment, pipe, ductwork, etc. is stripped out. Then the floors, walls, ceilings, and girders

Table 2
On-Line Sensors

Phase II System

- Temperature, pressure, pressure differential, flow rate in the vacuum filtration system.
- Gas / vapor samplers, particulate sniffers in various places.
- On-line residual gas analyzer sampling in various places.

Potential Add-Ons

- On-line optical comparitor to tell when coating has been removed.
- On-line paint thickness measurement.
- On-line Spectrometer to look at plasma plume for spectral lines of uranium, plutonium, etc. and to provide an on-line assay of the material going into the VAC-PAC drum.
- On-line radiation monitor to indicate possible migration of contaminants into the substrate.
- Multi-sensor data fusion and correlation would reduce the uncertainty for the controller system logic, as well as controlling the scan rate and the laser pulse-repetition rate.
- On-line assay and label printout for the waste container to reduce the "downstream" assay costs.

would be cleaned. A conceptual design for a three step system to deal with the stripped-out material, using all commercially available subsystems and components, has been completed. As shown in Figure 8, it consists of three major components.

1. Robotic sorting of material in scrap piles, using a gantry robot with dual-arm end effector.
2. Robotic laser cutting of metals, such as longer pieces of pipe or ductwork, to reduce them to workable lengths for cleaning the insides. Laser cutting has the advantage that no physical cutting wheel or saw touches the contaminated material, and there is no need for use of acetylene torches. The laser is located outside of a filtered cutting cell, with a BDS delivering the beam to a pedestal robot.

- Small parts will be cleaned in a filtered glove box. Pre-designs are done.
- Larger parts will be cut before laser cleaning, such as when cleaning the inside of long pipes or duct work. For I-beams and other large parts, the resale value will be maximized if they are cleaned uncut.

Figure 8
Lasers and Robotics For Contaminated Parts Sorting, Cutting, and Cleaning

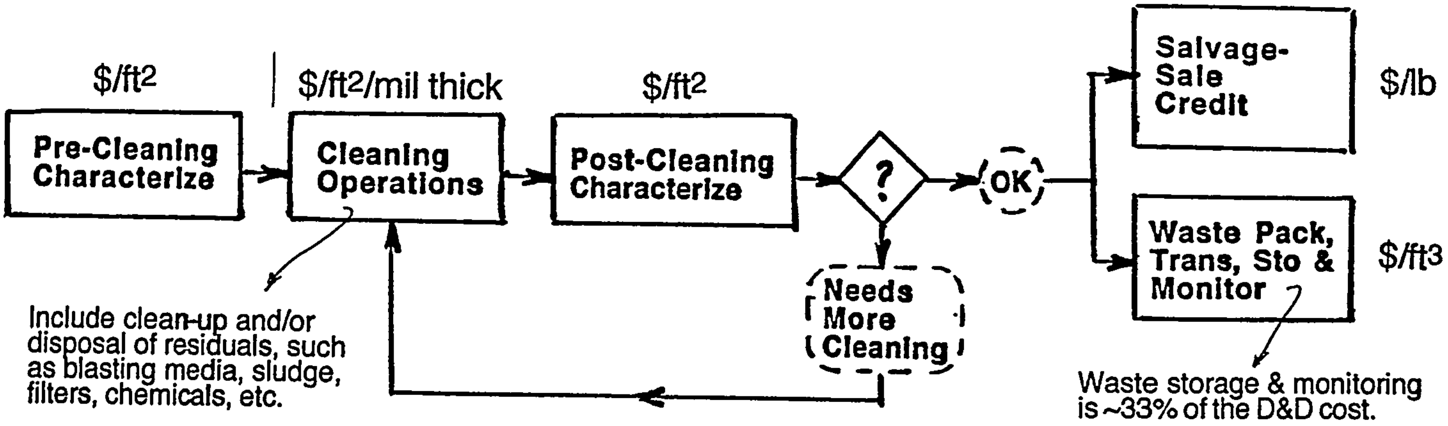
3. Robotic cleaning, using a modified version of the cleaning system described above, including saddle-gantry robot, and filtered cutting cell in addition to the nozzle.

Details were presented at the poster-session of the July 1995 DOE / METC Technology Developers and Users Interface Meeting. For laser cutting, the two worldwide commercially available and commonly used lasers are CW Nd:YAG and CW CO_2. They use electrical power and some cooling water (which can be closed-cycle using an air heat exchanger, eliminating need for water hook-up). Gas purge is used to clean out the kerf for thicker metals. This prevents the melt from clogging the kerf or absorbing and defocusing the beam. For surface-contaminated metals that are to be salvaged, it may also be necessary to first clean a path preceeding the cut so as to avoid having contaminants running into the melt zone.

There is also some interest in exploring the use of the chemical oxygen-iodine laser (COIL) for D&D, which emits in the near infrared close to Nd:YAG. COIL lasers "burn" chemicals (chlorine, hydrogen peroxide, potassium hydroxide, and molecular iodine) to create O2-singlet-delta that transfers energy to iodine that lases. Some scientific experiments funded by the USAF indicate that it may be possible to make large quantities of O2-singlet-delta electrically rather than chemically. The run time needs to be increased substantially; so this technology has awhile to go before being ready for industrial use.

Accomplishments

As reported at earlier METC conferences, in Phase I we demonstrated complete surface and surface-pore cleaning for lead-based paint and two-part epoxy on concrete and metal coupons. The full-scale industrial prototype system that has been under fabrication for several months is now being finished. Most subsystems have completed check-out testing. Half-power (3 kW) tests will be starting in early October 1995. Full-power testing is scheduled to start in early December 1995. PBased on small-scale tests, we predict that the system will be able to remove 100 square feet per hour of 20-mil thick aged lead-based paint. This rate should also apply to radioactive-contaminated lead-based paint since the contamination is usually much less than 1%, and uranium and plutonium are not far from lead in the periodic table.



- Reduced worker exposure, which can result in lower liability insurance costs ($/hr).
- Reduced needs for post-cleaning characterization ($/sq. ft).
- Reduced needs to assay waste-container contents ($/cu. ft).

Figure 9
Cost Comparison Algorithm

Costs and Benefits

A schematic cost algorithm is shown in Figure 9. A cost algorithm like this is needed to compare the different technologies shown in Table 1. For cleaning, most vendors quote dollars per square foot, regardless of thickness. It is not at all clear that credits are presently given for technologies that provide waste volume reduction, salvage value credit, saving insurance costs through reduced worker exposure, or on-line assay to reduce needs for post-cleaning characterization and post-operations drum assay. We will work up cost numbers for laser-based surface cleaning after full-scale testing late this year. FERMCO has done monitored testing to obtain comparative numbers for removing paint from steel using either dry-ice blasting, high-pressure water, plastic pellets, soda blasting, sponge, steel grit, ultra-high-pressure water, or wet ice blasting.

Benefits imbedded in the goals include pore cleaning, waste volume reduction, negligible substrate damage to maximize salvage or recycle value, reduced worker exposure, one-step final containerization, no wet chemistry for cleaning or for processing residuals, and possibly on-line assay. The market for nuclear D&D is quite large, involving both DOE and commercial nuclear facilities. In addition, the market for environmentally-safe non-radioactive lead-based paint removal is huge for ships, bridges, etc. The is also a large market for other applications such as aircraft cleaning. The technology is thus not only "dual use" but "multi-use."

Future Activities

A DOE-funded full-scale prototype system should be ready for laboratory testing by the end of this year. For these tests, we plan to remove lead-based paint from one foot by three feet concrete coupons. We hope to begin Phase III in the spring of 1995. This would involve field tests at a DOE facility to remove radioactive-contaminated paints. Pending funding, we will:

- Expand the test matrix to include two-part epoxy, and repeat all tests for metals.
- Add the other on-line instrumentation described above.
- Integrate the scanner into a MOOSE robot for floor cleaning and a ROSIE robot for wall cleaning.
- Continue development of parts cutting and cleaning systems.

3.4 Concrete Decontamination by Electro-Hydraulic Scabbling

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Abstract

Textron Defense Systems (TDS) is developing an electro-hydraulic device that has the potential for faster, safer, and less expensive scabbling of contaminated concrete surfaces. In the device, shock waves and cavitating bubbles are produced in water by the electric pulses, and the direct and reflected shock waves impinging on the concrete surface result in the crushing and cracking of the concrete. Pulse energy, frequency, and traverse speed control the depth of the scabbling action. Performance thus far has demonstrated the capability of a prototype unit to process a swath 24" wide, up to 3/4" deep at a linear velocity of up to 6 feet per hour, i.e., at a scabbling rate of 12 sq. ft. per hour.

Introduction

Contamination of concrete structures by radionuclides, hazardous metals, and organic substances (including PCB's) occurs at many DOE nuclear weapon sites, as well as at many utility power generation stations. In many instances the contaminants penetrate into the concrete to a significant depth. Removal of the concrete surface layer is considered as the most effective decontamination technology. By scabbling, the mass of concrete is divided into a) contaminated debris (rubble) of relatively small volume, and b) clean bulk concrete structure.

TDS is developing Electro-Hydraulic Scabbling (EHS), a cost-efficient, rapid, controllable concrete scabbling technique based on the Electro-Hydraulic effect (EH).

The EH system delivers strong pulses to the concrete surface by means of powerful shock waves originated by electric discharges and propagated through water between the discharge channel and the concrete. The high impulse pressure, developed at the liquid-solid interface, results in stresses that can controllably deform, crack, or break a whole solid body or spall the surface layer. Accordingly, EH has found applications for crushing and grinding of minerals, drilling of rocks, forming of metals, cleaning of surfaces, and demolition of foundations.

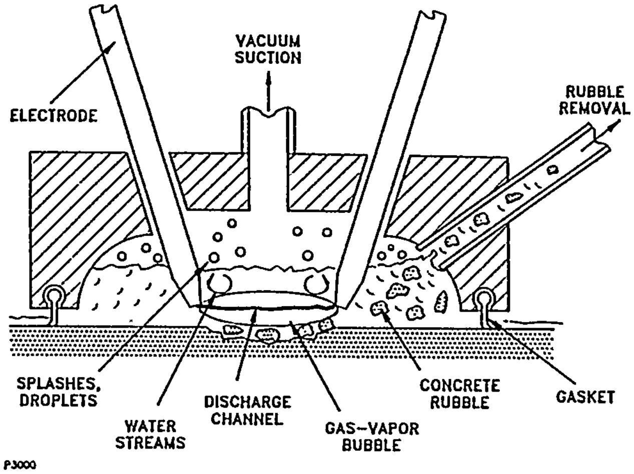


Figure 1

A variety of electromechanical configurations can be used for concrete EHS. In the device shown in Figure 1, the electric discharge takes place between two cylindrical electrodes.

Research sponsored by the U.S. Department of Energy's Morgantown Energy Technology Center, under contract DE-AC21-93MC30164 with Textron Defense Systems, 2385 Revere Beach Parkway, Everett, MA 02149; telefax: 617-381-4160.

EHS System Design Considerations

A block diagram of a "generic" EHS system is shown in Figure 2.

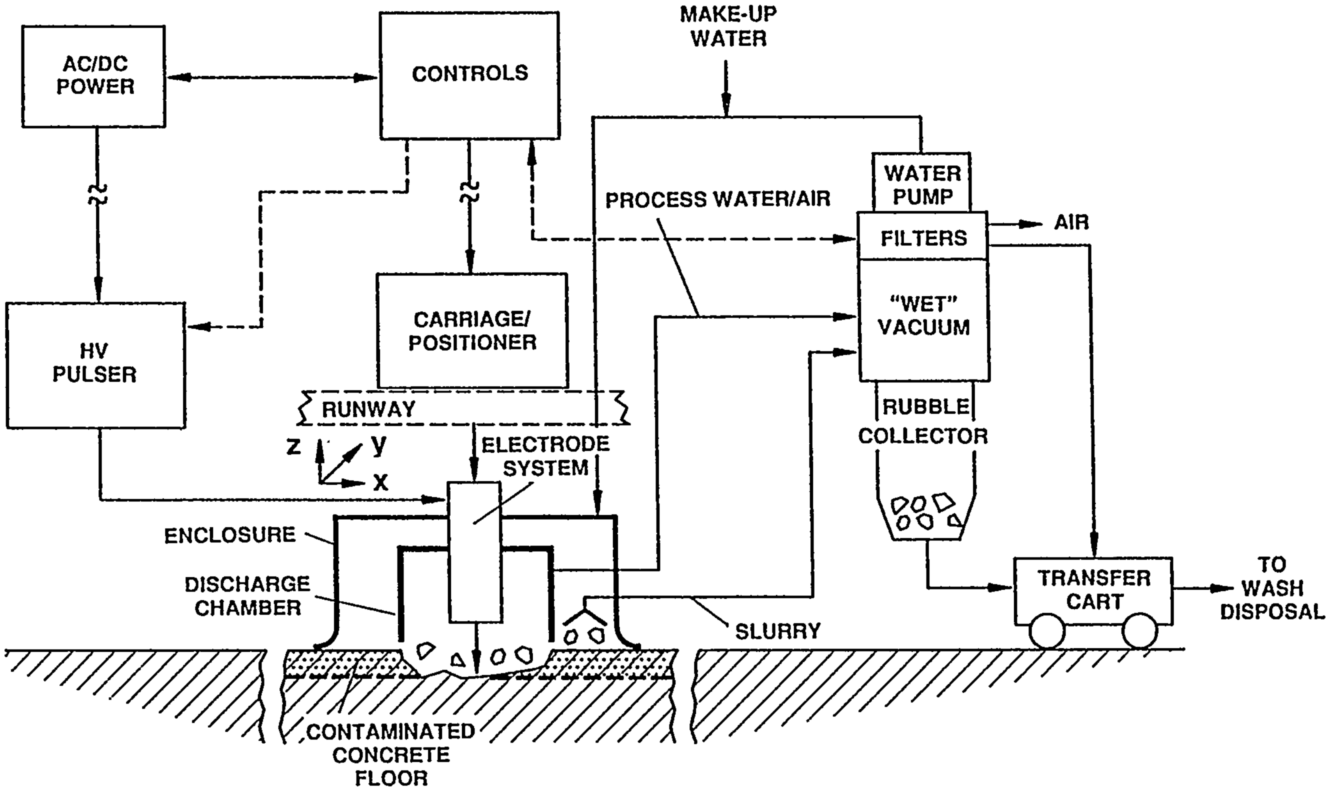


Figure 2

The electric power supply consists of two main units. The first, which can be remote, includes the high voltage DC supply (charger of capacitors) for the electric pulser and the AC supply for auxiliaries - motors, fans, controls. etc. The second unit - Pulse Forming Network (PFN) - should be located close to the EHS tool (module); low inductance of the connecting cables is required to generate short pulses with steep fronts. The DC power supply will operate at 20 to 100 kV, providing pulses of 500 to 4,000 J energy; a spark-gap switch with a function generator controls the repetition rate.

The EH processing module is the central component of the system. It carries single or multiple electrode pairs. Optimization and selection of the electrode configuration (for instance, rod-to-rod (point-to-point) vs. rail-to-rail (line-to-line)) for specific conditions is one of the major design considerations.

To achieve high energy efficiency, it is important to maximize pressure transfer to the concrete surface. Interelectrode gap and distance between electrodes and concrete surface (clearance) are among the variables to be optimized. In most cases, the clearance should be as small as possible, without mechanical damage of electrodes in the event they are "dragged" over the surface. At very high operating voltages, when electric discharge propagates directly through concrete, the electrodes should contact the surface. Electrode materials should be selected to minimize electro-thermo-chemical erosion.

In all designs, a 1 to 4 inch deep water layer should be maintained over the concrete surface. Water provides the media for shock wave propagation and, for short high voltage pulses, acts as an electrical insulation preventing breakdown through the air apart from the surface. It also prevents dust generation and, finally, allows retention and removal of concrete rubble (debris).

The rubble (sludge) is removed either by a solids-tolerant pump or by a wet vacuum type device, and collected in a drum. Wet rubble is separated by a proper filter while filtered process water (plus some make-up fresh water) is returned into a scabbling chamber.

The chamber contains a scabbling module mounted on a positioner. Its principal function is to isolate the process area/volume to prevent the spread of water and contaminated rubble over the surrounding floor. The perimeter of the chamber bottom should have a flexible gasket; the chamber is "sealed" against the concrete surface (e.g., floor) by generating reduced pressure inside the chamber with an appropriate blower. Surface scabbling proceeds in a semi-batch fashion. The X-Y-Z (or X-Z) positioner moves the scabbling module within the sealed chamber; after scabbling of an isolated area is completed, rubble and water are removed (sucked out), the chamber is moved to the next position, and the next scabbling cycle begins. A motorized carriage is used to move the chamber over the concrete surface.

The EHS system also contains a control station that can consist of various degrees of sophistication (from manual to completely computerized) to coordinate functions of scabbling proper, water/rubble flows, and mechanical traverses. These controls should be protected against EM noise generated by electric pulses.

The EHS design should be customized with respect to the type of surface area, its dimensions, orientation (vertical vs. horizontal), and the presence of obstacles or inclusions over/in the concrete construction. Reasonably flat floors, isolated concrete blocks that can be placed into water-filled tanks, and walls and bottoms of tanks themselves are examples of configurations that would require the simplest design of the scabbling module and chamber.

Design and Testing of Laboratory and Prototype EHS Units

Two EHS units were designed, assembled, and tested to demonstrate the technical feasibility of the EHS concept and to generate preliminary engineering data in order to develop system requirements and economic projections.

The first unit, shown schematically in Figure 3, was used to explore a range of operating parameters, as well as several electrode arrangements while scabbling the surface of a 3x2 sq. ft., 6" thick concrete slab immersed in a tank of water.

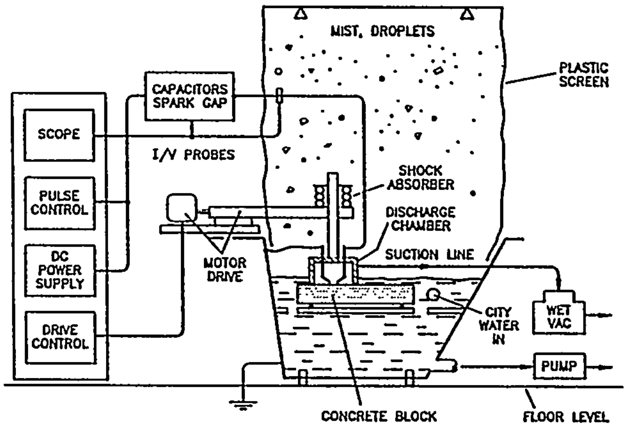
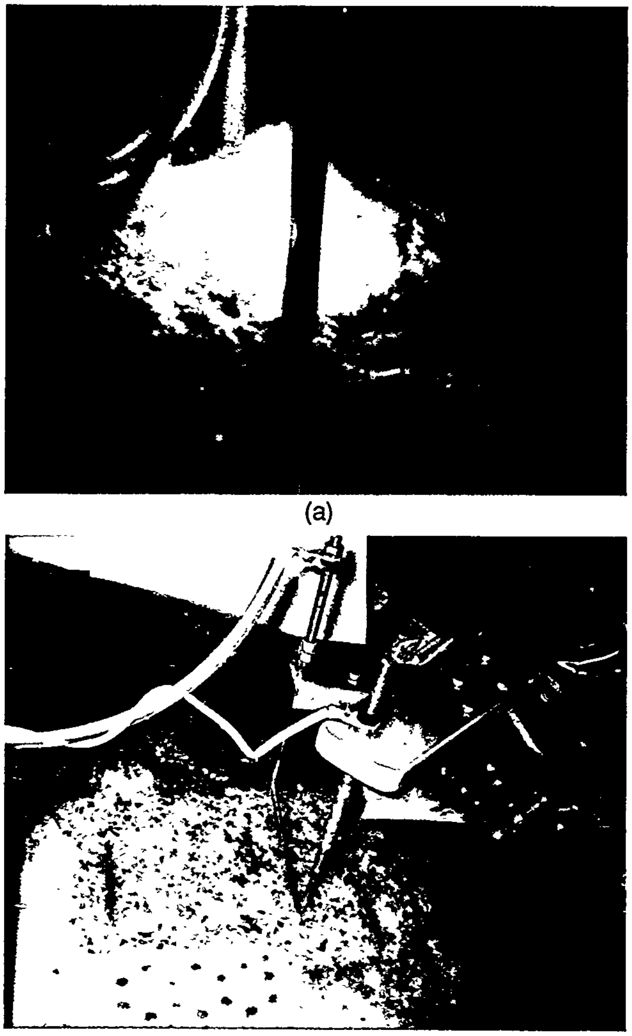


Figure 3

The unit was equipped with a 18 to 25 kV power supply which delivered 800 to 2200 J pulses of 5 to 15 microsecond duration at 0.5 to 3 Hz frequency. About 50% of the stored energy is released in a 6 to 12 mm discharge gap between a single pair of steel rod electrodes. Instantaneous appearances of the EH discharge over concrete surfaces before (a) and after (b) water splash formation are shown in Figure 4.

A range of operating parameters and a summary of the scabbling results are listed in Table 1.



(a)

(b)

Figure 4

Table 1. Range of Operating Conditions and Results for Laboratory Units

Operating Voltage	18-25 kV
Storage Capacitance	3.7-7.4 μF
Pulse Energy	800-2200 J
Operating Frequency	0.5-3.0 Hz
Average DC Power	1.5-4 kW
Electrode Velocity Continuous Traverse	1-12 inch/min
Scabbling Depth	0.2"-1.0"
Scabbling Trace Width	1.5"-3"
Depth of Water Layer	0.2"-1.5"
Concrete Removed, Volume/Pulse	0.4-1.2 cm^3 (0.05-0.25 cu. in.)
Rubble Particle Size	0.1<d>0.75 in.
Concrete Area Processed	10-30 sq in./sq./min
Energy Consumption	400-1500 J/g concrete

The second, larger unit (see Figure 5 schematic) was designed to explore EHS in a configuration applicable to concrete floor decontamination.

After testing several multielectrode configurations operating from either common or individual parallel pulsers, we chose the simplest design with a single pair of wide plate electrodes. A scabbling module with 24"-wide electrodes is shown in Figure 6 (top and bottom views, on a positioner inside the scabbling chamber).

The module is mounted on a motorized positioner traversing the concrete floor in a X-direction by 1" to 2" steps and lifting electrodes (Z-direction) during each step. A scabbling chamber of 3x3x2.5 ft. dimensions is made of fiberglass sheets and is supported by a forklift. The scabbling chamber, together with a top blower/vacuum, rubble collecting drum, diaphragm pump and power supply (25 kV, 8 kW installed DC power) and control cabinet nearby, is shown in Figure 7.

In Figure 8, the appearance of a 2' x 2' floor area scabbled to average 3/8" depth is shown. Operating parameters and characteristic

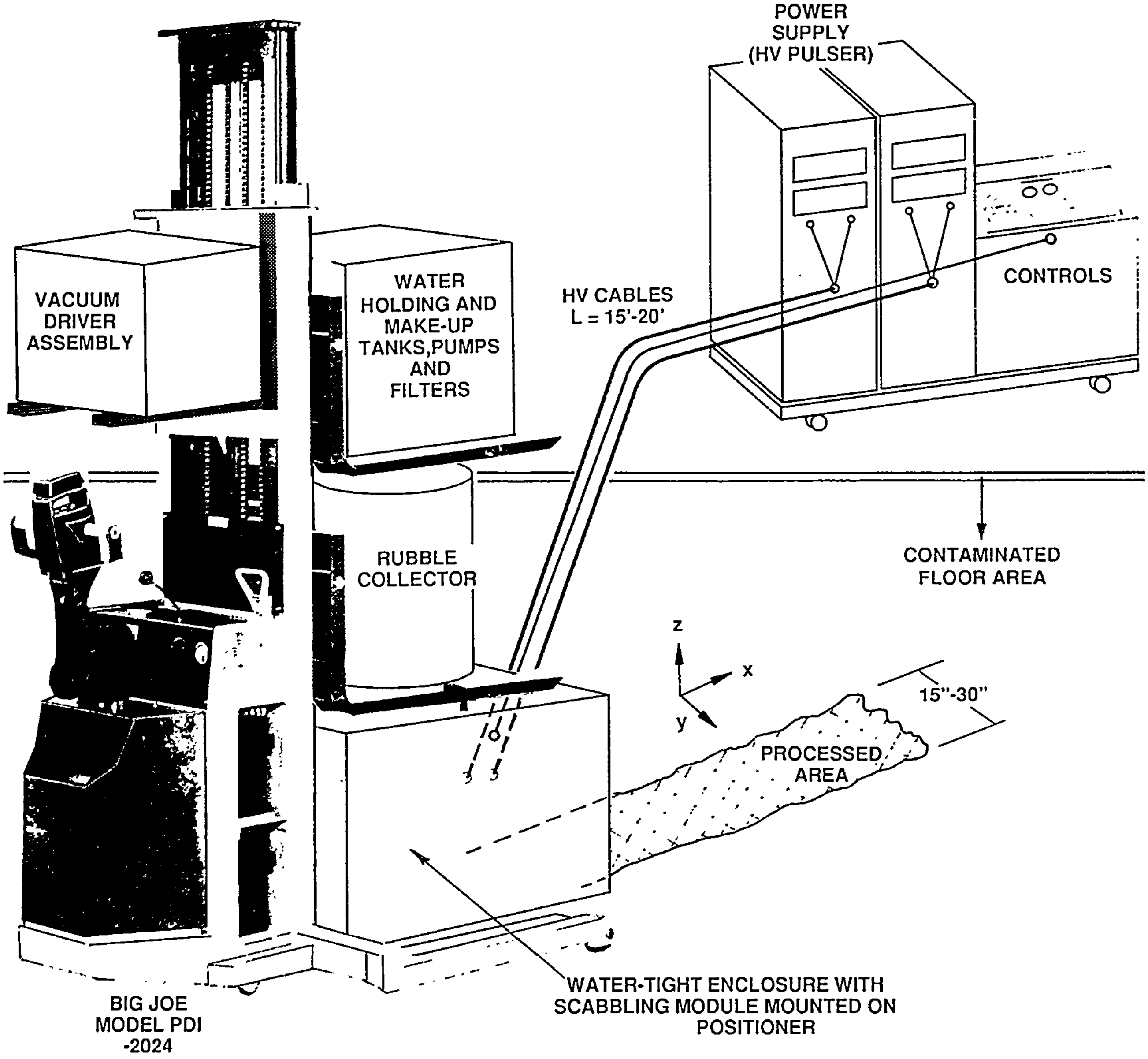
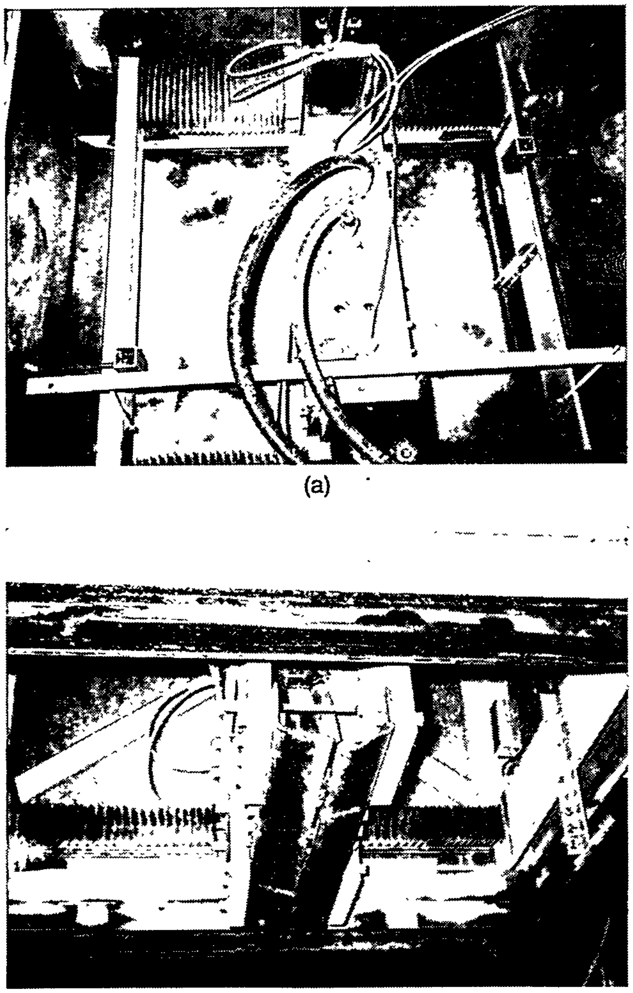

POWER SUPPLY (HV PULSER)
VACUUM DRIVER ASSEMBLY
WATER HOLDING AND MAKE-UP TANKS,PUMPS AND FILTERS
HV CABLES L = 15'-20'
CONTROLS
RUBBLE COLLECTOR
CONTAMINATED FLOOR AREA
z
x
y
15"-30"
PROCESSED AREA
BIG JOE MODEL PDI -2024
WATER-TIGHT ENCLOSURE WITH SCABBLING MODULE MOUNTED ON POSITIONER

Figure 5



(a)

(b)

Figure 6

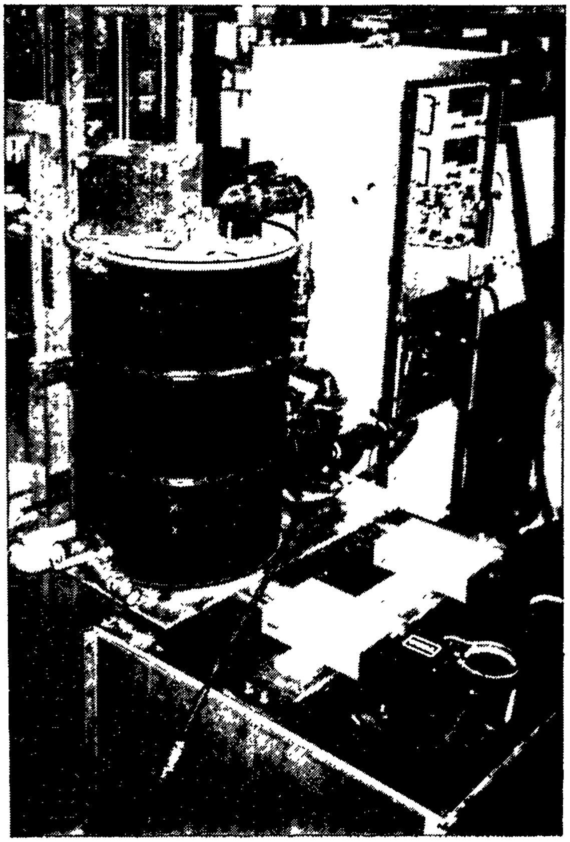


Figure 7

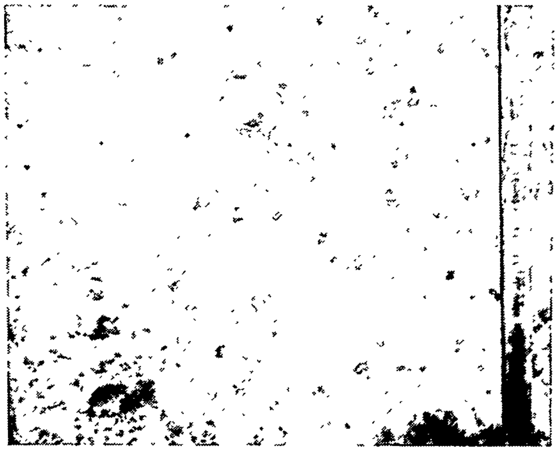


Figure 8

scabbling performance of this prototype unit are listed in Table 2.

Table 2. Range of Operating Conditions and Results for Prototype Units

Operating Voltage	21-28 kV
Storage Capacitance	10-13 µF
Pulse Energy	2.5-3.8 kJ
Operating Frequency	1.2-2.5 Hz
Average DC Power	2.5-6.0 kW
Water Layer Depth	3"-4"
Scabbling Chamber Vacuum	(-4)-(-7) in. H_2O
Module Effective Velocity (stepwise traverse)	1-2 in./min.
Traverse Step Length	1"-2"
Scabbling Trace Width	22"-26"
Scabbling Depth	1/4"-3/4"
Scabbling Rate	10-15 sq.ft./hr.
Energy Consumption (scabbling proper)	0.3-0.5 kWh/sq.ft.

An optional EHS unit employing a HV (120 kV) power supply has been also assembled. According to preliminary experiments, higher voltage and shorter discharge propagates directly through a surface concrete layer and provides two to four times higher scabbling energy efficiency. In addition, the power supply is smaller in size.

Concrete blocks and a floor with metal inclusions (re-bars, pipes) were scabbled without difficulty. In a few experiments, concrete contaminated by depleted uranium or cesium salts was scabbled; substantial reduction of the contaminant concentration in the concrete took place.

Field experiments planned for the near future at the DOE Fernald site should provide data to evaluate decontamination efficiency of the EHS technology. It may also be of interest to explore the capability of the EH technique to destroy organic contaminants in a concrete sludge due to absorption of the intense UV radiation generated by the discharge.

Conclusions

The feasibility of concrete surface scabbling using the electro-hydraulic technique has been demonstrated. Using engineering and design data developed during the preliminary development tests, costs of scabbling contaminated concrete surfaces is estimated to range between 5 and 20 $/m^2 for scabbling depths of between 0.25 and 1.0 inches. Further reduction in these costs can be expected as design improvements are introduced.

Acknowledgements

The development of the EH scabbling process has benefited by the managerial and technical contributions of METC CORs Mary Beth Ashbaugh, Kelly Pearce, and James Longenbach. The project was initiated in October 1993 as part of the technology development focus (EM50) of the DOE Decontamination and Decommissioning Program. The commencement of Phase III to define the operating procedure and performance specifications of a commercial prototype unit at a DOE site is anticipated for November 1995.

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3.5 Advanced Technologies for Decontamination and Conversion of Scrap Metal

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Introduction

The Department of Energy (DOE) accumulated large quantities of radioactive scrap metal (RSM) through historic maintenance activities. The Decontamination and Decommissioning (D&D) of major sites formerly engaged in production of nuclear materials and manufacture of nuclear weapons will generate additional quantities of RSM, as much as 3 million tons of such metal according to a recent study (SC&A, 1995). The recycling of RSM is quickly becoming appreciated as a key strategy in DOE's cleanup of contaminated sites and facilities.

The work reported here has focused on recycle of the concentrated and high-value contaminated scrap metal resource that will arise from cleanup of DOE's gaseous diffusion plants located in Oak Ridge, TN, Paducah, KY, and Portsmouth, OH. An estimate of scrap metal streams from decommissioning of the DOE's gaseous diffusion plants indicates that nickel makes up less than 20 percent of the total expected scrap volume but may carry more than 80 percent of the total value based upon current scrap prices. Because of the abundance and high intrinsic value of the nickel in the DOE's existing scrap metal inventory, the team of Manufacturing Sciences Corporation (MSC), Colorado School of Mines (CSM), and Covofinish Co., Inc. has applied its efforts to the refining and recycle of contaminated nickel and alloying of nickel recovered from DOE scrap with chromium and iron to produce high-quality stainless steel intermediate and end products for restricted reuses.

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[1] Research sponsored by the U.S. Department of Energy's Morgantown Energy Technology Center, under contract DE-AC21-93MC30170 with Manufacturing Sciences Corporation, 804 Kerr Hollow Rd., Oak Ridge, TN 37830; telefax: 423-481-3142.

Critical Recycle Issues

Two general approaches to RSM recycle are evident: 1) release to unrestricted commercial use after first removing contaminants from the surface or refining bulk contaminated metals; and 2) recycle to useful new products that would be restricted in end use. Removal of surface contamination and release is an economical approach in cases where there is no internal contamination and surfaces can be easily accessed. Unrestricted release of bulk-contaminated metals is presently not practiced in the U.S. due to the lack of an accepted standard for unrestricted release of these metals. Expense and difficulty of decontamination and quality assurance associated with unrestricted release make recycling of RSM into products with restricted end use attractive in many cases. Several critical issues surround the choice of recycle approach.

Technology

The DOE inventory of RSM includes some quantity of every engineering metal and alloy in common use. The technologies to produce these metals and alloys from ores and scrap metal feeds are well established and commonly practiced. However, conversion of these metals from their present contaminated state to a state suitable for restricted or unrestricted reuse requires adaptation of established techniques and development of new ones. Development and adaptation of technologies is central to this project and will be treated in more depth in a later section.

Economics

> First Law of Metal Recycle: "Economic viability of metal recycle increases in proportion to intrinsic value of the recycled metal"

> First Law of RSM Recycle: "Economic viability of RSM recycle increases in proportion to cost of disposal"

The market potential for recycling of RSM is the sum of the cost avoidance of disposal and the value of products that may be made from these metals. The comparative benefit of recycle for several metal types is shown in Table 1. The combined effects of intrinsic value of the metal and the cost of disposal are readily shown.

The current commercial price for disposal of radioactively contaminated scrap metals, not including any value that might be recovered, is a minimum of $1.50 per pound. The 1.5 million ton DOE RSM inventory therefore represents a $4+ billion disposal liability. All other sources of RSM, the nuclear utility industry being the largest among these, will create a disposal need of similar magnitude. When viewed as a business opportunity, including products that might be manufactured for restricted use, several businesses of moderate scale should be attracted to provide RSM recycle services and products.

Table 1. Economic Benefit of RSM Recycle

Metal Type	Cost of Commercial Sheet Metal	Cost to Produce RSM Sheet Metal	Cost of Disposal	Benefit of Recycle
Carbon Steel	$0.23	$1.80	$1.50	-$0.07
Stainless Steel	$1.32	$2.20	$1.50	$0.62
Stainless Steel (Ni Feed)	$1.32	$2.32	$1.50	$0.50
Nickel Alloy	$5.17	$2.60	$1.50	$4.07

Worker and Public Exposure

Workers and members of the public may be exposed to radioactivity resulting from contact with metals and contaminants during processing, direct contact with recycled RSM and contact with various effluents from the process. The project described here is actively generating data on actual exposures of workers and assessment of probable public exposures. Such data will be valuable input to future decisions concerning RSM recycle.

Liability

Environmental liability, both present and future, is a concern for both the generator and processor of radioactive scrap metal. Perceptions concerning legal exposure are often cited in decisions to pursue one course of action over another, for example, restricted versus unrestricted recycle. Responsible control of radioactive contaminants and understanding of the best processes to employ to assure metal recycling with minimum radiation exposure has been a key objective in the work reported here.

Current Work

The current work is directed toward development of several key technologies specific to recycle of RSM from gaseous diffusion plant origin. These technologies include 1) refining of contaminated nickel, 2) surface decontamination, 3) manufacturing, 4) detection and quantification of radioactive contaminants, and 5) assessment of worker exposures.

Metal Refining

Two methods of nickel refining are being undertaken. The primary refining approach is proceeding under subcontract with Covofinish Co., Inc. using a patent issued to that company entitled "Method for Removal of Technetium from Radio-Contaminated Metal. The method uses metal displacement reduction to remove Tc-99 from the electrolyte during electrorefining. Encouraging results have been observed in preliminary experiments. The current work will produce a bench-scale electrorefining cell that will identify conditions for later pilot-scale work.

A second approach being investigated is evaporative refining. Nickel contaminated with Tc-99 is being evaporated and collected on a substrate to quantify the degree of separation of the radio-contaminant. A several-order-of-magnitude difference in vapor pressure of the two metals is expected to result in a significant separation.

Surface Decontamination

MSC is pursuing development of chemical methods of surface decontamination as an aid for recycle of RSM. The chemical decontamination methods will be aimed at combinations of chemicals that will remove and prevent redeposition of radio-contaminants.

Gas phase decontamination using chlorine tri-fluoride ($C1F_3$) is being actively developed by Lockheed Martin Energy Systems at the K-25 site. MSC will coordinate its decontamination activities with the Lockheed Martin effort to provide maximum benefit from DOE expenditures.

Manufacturing

A principal activity within the project is development of manufacturing processes to produce prototype stainless steel vitrified waste containers starting from RSM characteristic of that arising from D&D of the gaseous diffusion plants. Manufacturing processes being developed and demonstrated include:

Precleaning of RSM. RSM may require precleaning prior to melting, depending upon the level of contamination from radio nuclides and other foreign material. Reduction of the level of radioactive contamination prior to melting can reduce the level of radioactivity in the final product and can benefit the overall process efficiency by reducing personnel exposure levels from the fabrication operations. It is possible that all operations subsequent to melting could be done with very minimal radiation safety concerns if most of the radio nuclide contamination is removed by precleaning combined with separation in the melt. Removal of other foreign materials before melting will greatly reduce the difficulties in maintaining control of the alloy chemistry of the final product. While it is possible to adjust the chemistry of the melt additions and reactions to achieve the desired alloy chemistry, MSC has demonstrated that stainless steel may be remelted to make new product within the same specification if careful precleaning and melting practices are followed.

Melting. All melting and alloying under this project is performed in an enclosed, gas-tight induction furnace under reduced pressure with an inert gas backfill. Such melting practice has two important advantages: 1) the vacuum or inert atmosphere prevents oxidation of the melt and thereby significantly reduces the secondary radioactive waste generated during the melting process, and 2) melting in a gas-tight vessel allows positive containment of volatile radionuclides such as tritium and cesium and provides the opportunity for treatment of a very low volume of off-gas to remove volatile radioactive constituents if found necessary.

Rolling. In a modem, high-volume commercial operation, stainless steel sheet metal would probably be produced as coiled hot-rolled product from a multiple-stand tandem mill and would be further cold reduced by a second multiple-stand rolling operation or on a reversing mill. Operations of this type produce large tonnage quantities of sheet metal on a commodity basis.

MSC's approach to rolling for metals recycle has been to use a 4-high reversing mill to produce plate or sheet to meet a specific product requirement. In other words, the objective of the rolling operation is different from commercial plate and sheet production in that the end product is not a commodity to be shipped for use in a variety of undefined products, but instead the plate and sheet is produced on a custom basis according to end use. The typical approach is to produce discreet pieces that are tailored in size to minimize waste and internal recycle.

Internal Recycle. A key issue in recycle of RSM is internal recycle of trimmings and other scrap. The overall objective of the processing is maximized process throughput, minimized secondary waste, and virtually complete consumption of the RSM in useful product. Internal recycle of RSM scrap and trimmings will be demonstrated within the project.

Detection and Quantification of Radioactive Contaminants

Work under the first phase of the project highlighted the need to accurately detect and quantify the specific isotopes and level of residual contamination in RSM and products that might be made from recycle of RSM. Determination of radioactive contamination by survey of radiation from the surface of RSM or products is limited in ability to differentiate by isotope and to detect low levels of contamination. Laboratory methods of radioanalytical and radiochemical analysis are expensive and impractical to apply to RSM streams of unknown origin. Methods of field survey, laboratory analysis, knowledge of scrap metal source, process knowledge, and quality assurance are being combined to provide an economical and accurate approach to contaminant characterization.

Worker Exposure Assessment

Personnel exposure data is being gathered from all operations encompassed within the project that involve handling of RSM or secondary waste from processing. The data will contribute to construction of likely worker exposure from large scale RSM recycle activities.

Related Work

Recycle of Stainless Steel to Drums and Boxes

MSC is engaged in demonstration of stainless steel RSM recycle under contract with Westinghouse Savannah River Company. This project includes melting, casting, rolling, and fabrication of boxes and drums in its plant in Oak Ridge, Tennessee. The technologies chosen for demonstration are directly relevant to the economical recycle of contaminated stainless steel on a small scale, dedicated basis and will be used to establish design and operating parameters for a commercially viable plant to recycle RSM.

Recycle of Copper Scrap

FERMCO recently awarded a contract to MSC for an engineering study and demonstration for removal of uranium and other trace surface contaminants from copper scrap to allow unrestricted release of the copper. The demonstration will process about 30 tons of copper and produce performance data and process information that will allow scale up for larger quantities.

New RSM Recycle Plant

MSC and British Nuclear Fuels, a world leader in nuclear fuel cycle service, have formed a joint venture to design and build the world's most advanced recycling plant for radioactive scrap metals. MSC will operate this exciting new facility. Ready to receive shipments of metal in November, 1995, the plant will allow nearly *100% RECYCLE* of scrap metal coming from commercial utilities, DOE sites, and other generators of radioactive waste. All incoming metal will be decontaminated. Metal that can be easily surveyed and verified to be clean will be recycled into the secondary metals market. Most other metal will enter a process of melting, casting, rolling and fabrication to produce useful products such as drums, boxes, and other containers for storage or disposal of radioactive waste and shielding components for research and nuclear industry uses. Scrap from in-house fabrication will be internally recycled. The small amount of metal not suitable for recycle will be melted into maximum density blocks for economical burial.

Summary

Recycle of RSM is unique in its promise of significant positive contribution to the cleanup of DOE's contaminated sites and facilities. RSM recycling offers the possibility of recovering the intrinsic value of the contaminated metals and reduction of future waste by reusing the contaminated metals in place of others that would become contaminated. Reuse of existing infrastructure on DOE sites to do RSM recycling in the future provides the opportunity for productive interim reuse of contaminated facilities and reemployment of displaced workers.

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Acknowledgments

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3.6

Remote Operated Vehicle with Carbon Dioxide Blasting ($(ROVCO_2)$)

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The Remote Operated Vehicle with Carbon Dioxide Blasting ($ROVCO_2$), as shown in a front view in Figure 1, is a six-wheeled remote land vehicle used to decontaminate concrete floors. The remote vehicle has a high pressure Cryogenesis blasting subsystem, Oceaneering Techniologies (OTECH) developed a CO_2 xY Orthogonal Translational End Effector (COYOTEE) subsystem, and a vacuum/filtration and containment subsystem. Figure 2 shows a block diagram with the various subsystems labeled.

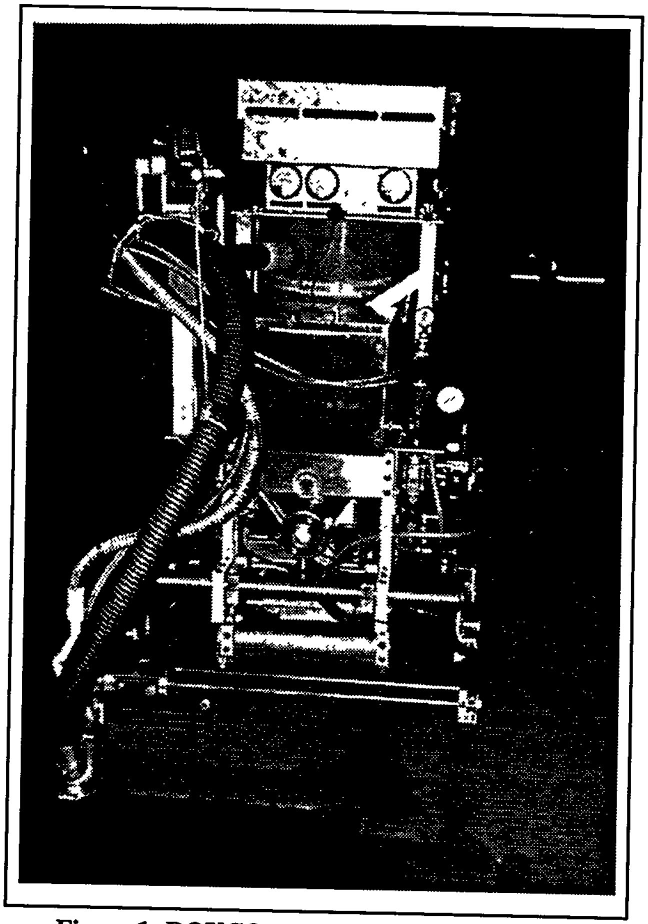

Figure 1: $ROVCO_2$ remote vehicle (front view)

The cryogenesis subsystem performs the actual decontamination work and consists of the dry ice supply unit, the blasting nozzle, the remotely controlled electric and pneumatic valves, and the vacuum work-head. The COYOTEE subsystem positions the blasting work-head within a planar work space and the vacuum subsystem provides filtration and containment of the debris generated by the CO_2 blasting. It employs a High Efficiency Particulate Air (HEPA) filtration unit to separate contaminants for disposal. All of the above systems are attached to the vehicle subsystem via the support structure.

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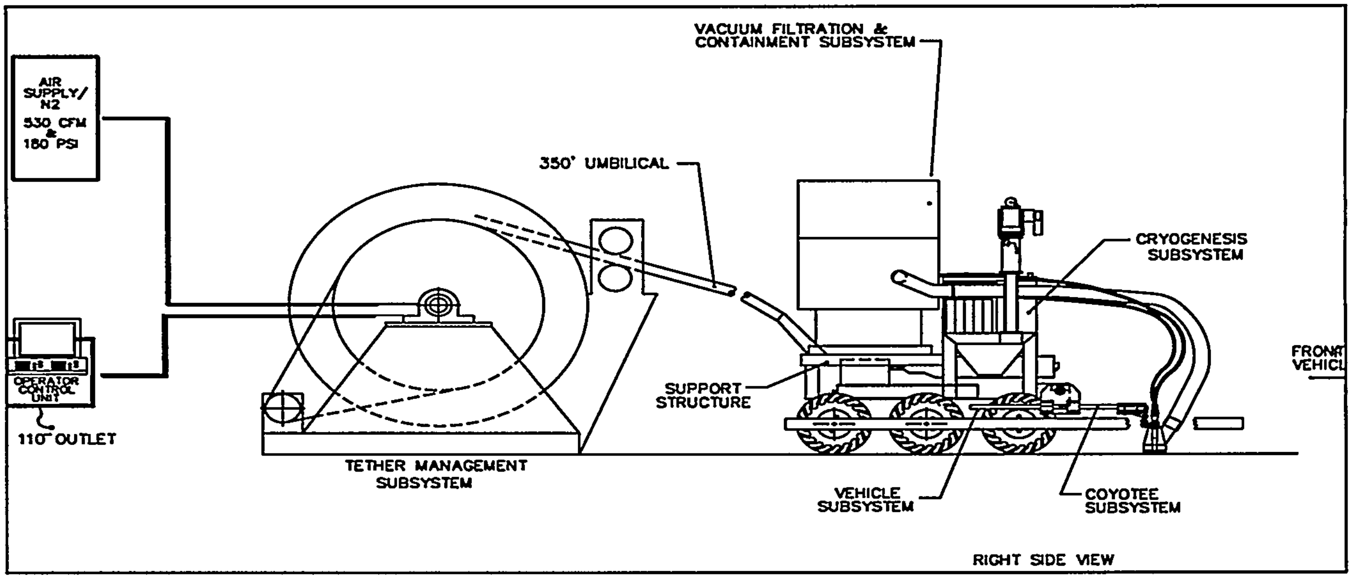

Figure 2: ROVCO$_2$ block diagram showing subsystems

The vehicle subsystem is a six-wheeled, remote controlled vehicle which provides the transport and power required by all of the vehicle-mounted subsystems and equipment. Two camera/light assemblies, one black and white fixed-position camera and one color camera mounted on a pan and tilt unit, provide viewing for navigation, obstacle avoidance, and operations. The vehicle was procured from a commercial vendor.

Separate from the vehicle are the tether management subsystem and the operator control unit. The tether management system provides an electric winch to manage the vehicle's 300 foot umbilical. The umbilical provides command, data, and power transfer between the vehicle and the control subsystem. Additionally, it provides a delivery system for the compressed gas that is used in the blasting system. The operator control unit provides a single operator, integrated controls, automated repetitive functions, video display, and equipment status feedback.

ROVCO$_2$ was developed under a Department of Energy (DOE) program at the Morgantown Energy Technology Center in response to a need at the Oak Ridge K-25 site, and other sites, for concrete floor decontamination. The objectives of the ROVCO$_2$ program were as follows:

- reduction in waste volume,
- faster decontamination of floors,
- improved decontamination effectiveness,
- reduced decontamination costs, and
- reduction in worker exposure to contaminants.

Reduction in Waste Volume

Previous methods of decontaminating included scabbling, shot peen blasting, and chemicals. However, each of these methods had different problems associated with them, especially in waste volume and handling. Scabbling tended to remove a large amount of concrete, shot peen did like wise and created secondary waste, while chemicals, when used in a radioactive environment, created mixed wastes.

ROVCO$_2$ reduced waste volume by using dry ice (drice) pellets as the blasting medium. When the high pressure drice contacts the floor, it subliminates. The impact of the pellet lifts the contaminated material particles off the floor, up into the work-head. These particles are then vacuumed into the collection system and into a drum. The gases pass through a HEPA filter and exhaust. The vacuum system is the Pentek VACPAK, which has been accepted for nuclear use both at DOE sites and power plants. The blasting produces waste material of very fine particles, and no secondary waste material. The small particles compact well into the drum, which can contain eight to twelve hours worth of waste material. The drum can be changed using a maskless procedure, approved at nuclear power plants and it is designed for over packing into 55 gallon drums.

Faster Decontamination

ROVCO$_2$ is intended to operate in an automated blasting mode, requiring only human supervision, providing faster decontamination. This mode of operation allows for higher blasting pressures and more consistent operation, thus speeding cleaning. In addition, because the operator has a supervisory role, they can operate multiple units from a single control area. In all, the system eliminates the need for workers to suit up, reducing worker fatigue.

Improved Decontamination Effectiveness

In addition to reduced waste and faster decontamination, ROVCO$_2$ also provides improved decontamination effectiveness. The nozzle has been designed using Computational Fluid Dynamics (as is used for NASA rocket nozzle design) to produce a maximum pellet velocity of 1100 feet per second. In addition, the containment system engineered to entrain nozzle blast and minimize recontamination of surface. Also, by utilizing the automated features of the vehicle and end effector subsystems, ROVCO$_2$ provides automated, complete blasting coverage of surfaces. This method of decontamination avoids unnecessary overlap or bypassed areas.

Reduced Cost

Another benefit of the ROVCO$_2$ system is its reduced decontamination costs. This reduction is achieved in combination with many of the other benefits. By providing faster decontamination, less overall time is spent on a given area as compared to other methods. Additionally, by minimizing waste production and volume, fewer changeouts are needed, thus increasing the amount of time the unit is operational within the overall time. The automated features of the system allow

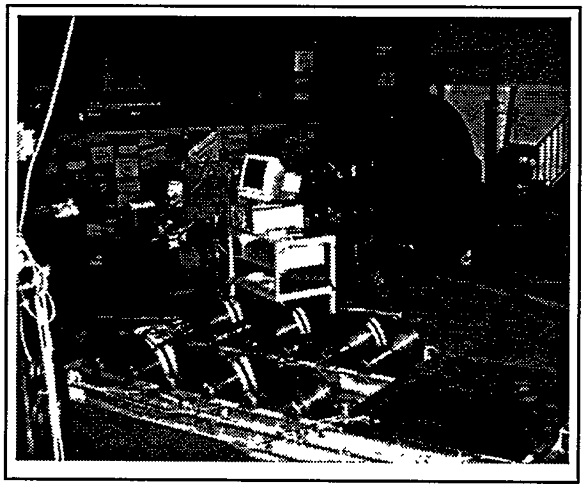

Figure 3: ROVCO$_2$ reliability test stand with Tether Management System winch and Operator Control Unit in the background

large areas to be decontaminated with confidence, minimizing inspection requirements to ensure that the decontamination has been achieved. Finally, the ROVCO$_2$ system has been designed for high reliability and tested (see Figure 3) to ensure it meets its reliability requirements.

Worker Exposure

Finally. as stated earlier, ROVCO$_2$ provides reduction in worker exposure. This reduction is accomplished mainly by allowing for blasting via remote control from up to 100 meters away. Only two operations require human intervention for operation: drum changeout and pellet hopper filling; both of these can be accomplished in low radiation areas by driving the vehicle away from any hot areas. Drum changeout is required once every shift, and hopper filling is required four to six times per shift, however, future fillings can be accomplished by providing a solenoid on the hopper to allow remote filling and thus reducing human intervention even further. Also, the demonstrated high system reliability minimizes worker maintenance exposure. A preventive maintenance schedule will be developed that requires maintenance intervention only when the vehicle is in uncontaminated areas.

Status

At the present time, ROVCO$_2$ is completing its cold testing at Oceaneering in Upper Marlboro, Maryland and is schedule for final system acceptance in October, 1995. Oceaneering is working with DOE to establish an appropriate hot site for initial ROVCO$_2$ operations. Current choices are the K-29 building at the Oak Ridge K-25 site and the Plutonium Finishing Plant at Hanford.

3.7 Mobile Worksystems for Decontamination and Dismantlement

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Introduction

Many DOE nuclear facilities have aged beyond their useful lifetimes. They need to be decommissioned in order to be safe for human presence in the short term, to eventually recover valuable materials they contain, and ultimately to be transitioned to alternative uses or green field conditions. Decontamination and dismantlement are broad classes of activities that will enable these changes to occur. Most of these facilities – uranium enrichment plants, weapons assembly plants, research and production reactors, and fuel recycling facilities – are dormant, though periodic inspection, surveillance and maintenance activities within them are on-going. DOE estimates that there are over 5000 buildings that require deactivation to reduce the costs of performing such work with manual labor. In the long term, 1200 buildings will be decommissioned, and millions of metric tons of metal and concrete will have to be recycled or disposed of. The magnitude of the problem calls for new approaches that are far more cost effective than currently available techniques. This paper describes two technologies that are viable solutions for facility D&D.

Objectives

While facility transitioning is inevitable, there are no specific regulatory drivers that mandate it. Furthermore, there are no release standards for the materials within facilities that represent a significant economic resource, nor are there means to safely dispose of materials that cannot be recycled. The current strategy, therefore, is to make facilities "cheap to keep" while these broader issues are addressed by all stakeholders. In the meantime, all facilities have to be monitored and guarded to minimize environmental, health and security risks.

Deactivation (removal of radiologically active material) and deinventory (removal of nuclear material) are means to reduce the costs and risks associated with facility surveillance and monitoring; health physics, maintenance and security personnel will be able to work around and within facilities with reduced worker protection requirements, thus enabling them to work more effectively and efficiently. Selective equipment removal, in which some part of a

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facility is extricated while minimally disturbing surrounding objects, will thus be particularly relevant in the near term. The motivation for selective equipment removal is to remove highly contaminated components from an area that is otherwise relatively clean thus lowering the overall average level of contamination and reducing worker protection and security requirements. The operation has to be selective because any and everything removed represents a waste stream that must be dealt with.

In the longer term the scope and scale of facility D&D will increase dramatically. The future work agenda will extend beyond isolated equipment removal to complete dismantlement of apparatuses and decontamination of entire buildings. As materials are removed, component decontamination, size reduction and packaging techniques will become critical elements of the overall D&D strategy. Likewise, sensor surveys and mapping will extend beyond surveillance and become the mechanisms to assure that reduction of contamination is in compliance with applicable regulations and that facility closure is possible.

To keep within ALARA standards and minimize worker exposure to radiation and other hazards, these tasks have to be performed with robots and other remote equipment to the maximum extent possible. Remote D&D tasks require manipulators to move objects and position end effectors; the size and extent of most facilities dictates the need for transporters, such as mobile robots and overhead gantries, to move the manipulators. All of these systems need have to have capability, durability, and productivity rivaling human counterparts in order to execute the D&D work agenda in a timely, cost effective manner.

While not new to the nuclear industry, currently available remote equipment is impotent relative to the breadth and magnitude of the work to be performed.Traditional teleoperated systems rely on a human operator to direct the entire task, though most of his effort is spent controlling basic functions and coordinating motions of the robot rather than addressing the work task itself. Furthermore, operator interfaces are cumbersome; the predominant source of information about the work environment is gathered by remote viewing cameras and displayed as conventional (2-D) video images. All told, these factors typically reduce the productivity of remote work to a mere 10% of the equivalent manual laborer.

To approach the efficiency and effectiveness of a human worker, tasks will have to be performed with some degree of automation, in which many aspects of execution are delegated to robot controllers and other software. Automation requires knowledge of workspace geometry; success in the manufacturing domain is attributable to rigging the environment and to *a priori* information about part sizes and locations relative to the robot. However, blueprints and as-built drawings of most DOE facilities do not exist. Enabling technologies for D&D are robotic perception systems that collect data from the remote site and use it to model the geometry of the robot's workspace as it is discovered.

DOE facilities pose unique challenges to the design of an appropriate perception system. Typical facilities are much larger than the robots that will operate in them, which implies that data from multiple sensor viewpoints must be merged to construct a complete model. Complexity of the environment – large numbers of objects, tremendous variation in scale, and complicated arrangements – add to the difficulty. Fortunately, all objects in the facility are man-made, which limits the types and shapes of objects that will be encountered in the model building phase. Further, most objects can be modeled by simple geometric descriptions and can be readily identified by a human observer. A third challenge then is to devise a system that takes maximum advantage of the human's object recognition ability while minimally burdening him.

Approach

DOE's approach to D&D robotics emphasizes modularity: hardware (manipulators, transporters, tools, sensors, control consoles) and software (controllers, perception systems, planners and data processing) can be assembled into systems according to task needs and infrastructure constraints. A key hardware element is the *mobile worksystem*, which provides remote capabilities to operate tools, manipulate and package contaminated objects, and position sensors. In Phase I of this project, an existing teleoperated worksystem (the Remote Work Vehicle – originally developed for accident recovery at Three Mile Island) was upgraded with contemporary computer controllers and tested in mock-ups representative of DOE facilities requiring D&D. Using the results from Phase I, a next generation mobile worksystem, *"Rosie"*, was designed and built.

The most important attribute of the *Rosie* mobile worksystem is its ability to address a variety of relevant D&D problems. Tools will be deployed by a high reach telescoping boom capable of positioning 2000 lbs. throughout a large work envelope. Tools currently envisaged include pipe shears, impact wrenches and grippers for dismantlement; CO_2 blasters, coating sprayers and laser ablation systems for decontamination; scabblers for concrete surfacing; and grapplers for moving heavy objects. The boom is mounted on an omni-directional wheeled locomotor, a configuration that is well-matched to the primarily flat floor world of DOE facilities. *Rosie* is built meet D&D challenges: it can withstand the rigors of heavy work over periods of years, its mobility makes it self-deploying, and its design incorporates spare power and controls for the addition of a variety of tools. In addition, *Rosie* combines dexterity with high strength its power & signal tether provides reliable communications and allows unlimited work duration.

A crucial software component of robotic D&D is *Task Space Scene Analysis* (TSSA) in which a computer constructs a geometric model of the region in which the robot is performing its task. With such information available, that automatic control techniques can be used to speed task execution relative to conventional teleoperation. Prior and related ongoing TSSA efforts within DOE emphasize human interaction: approaches in which the operator matches and adjusts the registration of object models to observed data. To complement these, we developed a semi-automatic task space scene analysis system, called *"Artisan"*.

Artisan builds a virtual world for remote D&D operations by constructing 3-D models of a robot's work environment using data from a scanning laser rangefinder. Planar and quadric surface representations of objects are then incorporated into a task space model that can be viewed and analyzed by the operator, accessed by motion planning and robot safeguarding algorithms, and ultimately used by the operator to instruct the robot at a level much higher than teleoperation. All of this occurs with a minimum of human interaction.

Technology Description

Rosie Mobile Worksystem

Rosie (Figure 1) is a omni-directional locomotor with a rotating turret upon which a telescoping boom is mounted. The locomotor is 2 m wide by 2.9 m long and 1.1 m high four-wheel drive/four-wheel steer mechanism that provides and distributes hydraulic and electrical power and computing capabilities for all manipulation, tooling and sensing functions. The locomotor has on-board tether management and is connected via a multi-conductor umbilical to an off-board electrical power distribution unit and control station. The boom is a high-payload, long-reach arm that can carry up to 770 kg with a 6,800 Nm moment load at a distance of 6.1 m from its shoulder joint.

Rosie provides all of the necessary locomotion and heavy lifting capabilities to perform D&D tasks remotely using a baseline of teleoperation. Two forms of power are available at the boom tip for tooling: 20A @ 120 VAC electric and 15 gpm @ 3,000 psi hydraulic. Together with surplus control channels, these allow addition of a wide variety of devices ranging from hand-tools to large demolition and decontamination equipment. *Rosie* is designed to deploy DOE's Arm Work Module (DAWM), a pair of Schilling manipulators on a five degree of freedom positioning platform, at the end of its boom for tasks requiring dexterity. Heavier tools, such as jack hammers, can be substituted for the DAWM to address additional tasks. A list of tasks and tools that Rosie supports is presented in Table 1.

Table 1: *Rosie* tooling attachments for D&D tasks

	Tasks	Targets	Tools
Decontamination	• Surface washdown • Hot spot removal • Surface removal • Sealant application	• Concrete walls & floors • Structural supports	• pressurized water sprayer • CO_2 pellet blaster • scabbler • laser ablation system • coating sprayer
Component Removal & Size Reduction	• Mechanical π disassembly • Drilling • Metal plate cutting • Pipe cutting • Crushing	• Ventilation ducts • Electrical power components • Process tanks, vessels & piping • Waste storage drums • Lighting fixtures & supports • Fuel elements	• hydraulic shear • abrasive grinder/saw • reciprocating saw • grappler • drum handler • impact wrench • concrete crusher
Demolition	• Metal cutting • Concrete cutting • Coring • Drilling • Hammering	• Steel support members • Reinforced concrete • Reactor vessels, tanks, piping & fuel pool liners • Lead shielding • Concrete floors &walls	• high pressure water jet • hydraulic shear • plasma arc cutter • diamond wire saw • jack hammer • pry bar
Debris Collection & Waste Handling	• Grasping • Compaction • Containerization • Container transport	• Dry fines • Liquids • Concrete rubble • Metal scrap • Nuclear material	• gripper • scoop or bucket • vacuum • electromagnet • drum handler

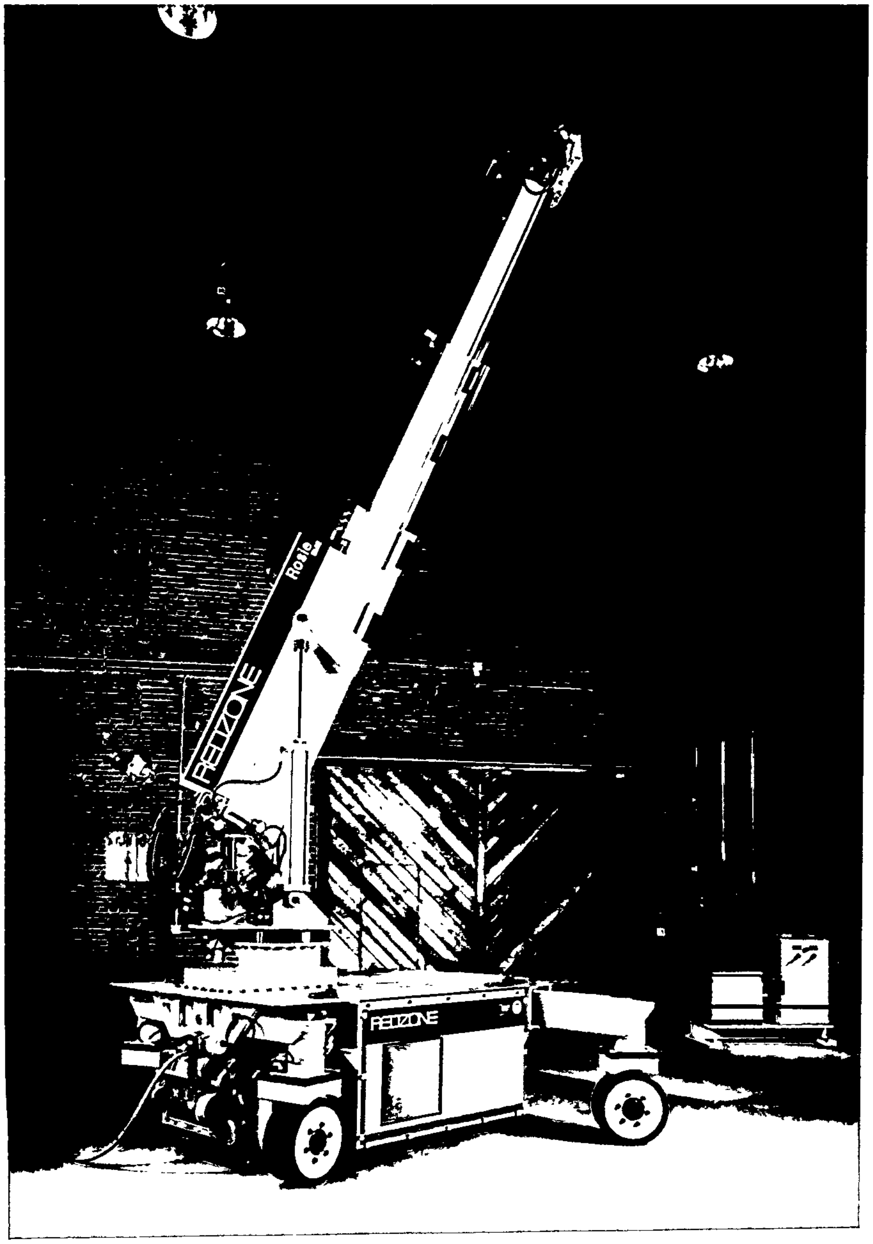

Figure 1: *Rosie* mobile worksystem

Rosie has multiple on-board cameras with pointing and lens control functions for remote viewing and stereo microphones for audio feedback. The worksystem can also operate as a telerobot; because all of *Rosie's* motions are servo-controlled, all movements can be directed by a human operator at a remote console or by another computer. *Rosie's* control and communications architecture is designed to accommodate the addition of sensors connection to additional processors through RTDP-standard interfaces.

Rosie is designed to withstand the same rigors of prolonged operation and occasional abuse experienced by heavy construction equipment, The worksystem is able to survive inadvertent collisions with obstacles while driving or manipulating. As much as possible, all cables and hoses are protected from damage by running them internal to the structure or protecting them in guides. Simplicity has been emphasized and key areas are over-designed to reduce the likelihood of failure. Furthermore, critical components in the electrical and hydraulics subsystems are modular and easily accessed to facilitate replacement. Entire modules, such as the tether reel and the electronics enclosure, can be removed for maintenance.

Artisan Task Space Scene Analysis System

An *Artisan* session begins when the operator instructs the system to acquire several images of the scene. *Artisan* uses a scanning laser range finder which produces both a range image and a reflectance image, both of which are displayed on the workstation screen. Since the field of view of the sensor is usually larger than the area that the operator wishes to work on, he restricts the system's attention to a particular region of interest by drawing a box around it. The operator then indicates what objects *Artisan* should expect to find in that pre-defined object types and sizes. *Artisan* uses these operator-provided "clues" to determine the exact location and dimensions of these objects in the scene. First, it creates a Cartesian mesh from the range data in the region of interest. This mesh is then segmented into planar and quadric patches. Planar patches indicate flat surfaces, such as walls, floors and I-beams, while quadric patches are parts of curved surfaces like pipes, tanks and valves. *Artisan* then automatically tries to match these surface patches to geometric descriptions of the objects that the operator identified earlier and displays the results.

For each object recognized, the operator can either accept or reject what *Artisan* recognized. Accepted objects then appear in a world model at the locations that *Artisan* has calculated. This process of range date collection, processing and user interaction continues until the operator is satisfied with the 3-D model of the robot's workspace. The entire process from range image acquisition through object recognition takes about three minutes on an SGI Indigo workstation.

When Artisan is running, the workstation screen appears like the layout shown in Figure 2. The upper left quadrant contains data from the scanning laser rangefinder displayed as a range image (on the left) and intensity image (on the right). In the range image, distance is encoded by a gray scale; objects in the foreground are dark while objects in the background are light. The lower left screen section shows the ranger data as a segmented mesh; the world model is displayed using a robot simulation package in the lower right. All Artisan functions are accessed through the main menu window in the upper right.

The Artisan software system consists of six modules and described in Table 2 and shown as a block diagram in Figure 3.

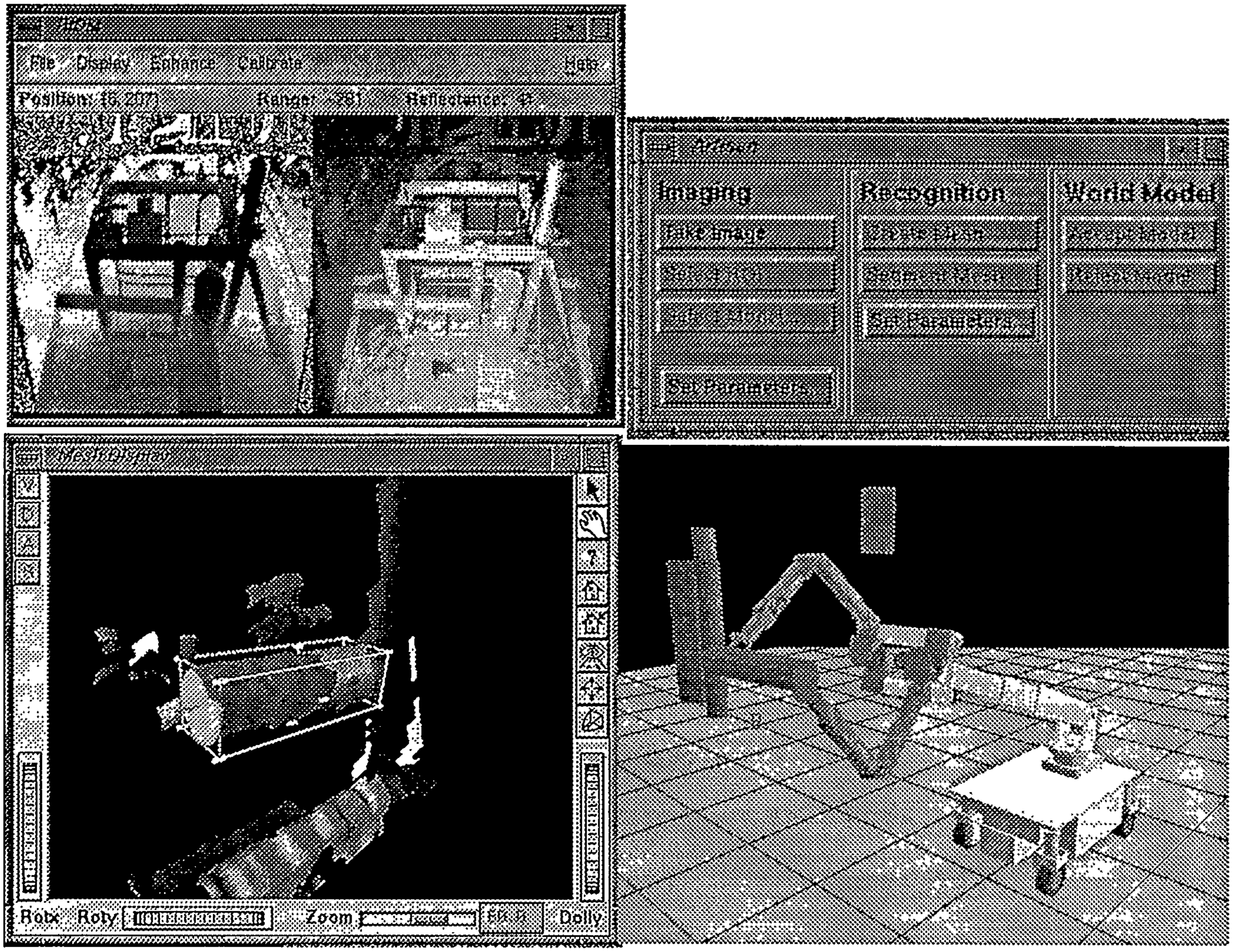

Figure 2: Artisan screen layout

Table 2: Artisan Modules

Artisan Main Menu (AMM)	Allows the operator to acquire images, to choose object models from a library that should be found in a selected region of interest, and to verify object matches
Range Image Module (RIM)	Acquires range/reflectance images; applies temporal and spatial filters to range images; generates meshes from range data
Range Image Display Module (RIDM)	Displays range and reflectance images; allows selection of a region of interest by the user; provides image enhancements for viewing purposes
Mesh Display Module (MDM)	Displays meshes and models generated by other modules; allows the user to resize matched object models and view objects from arbitrary viewpoints
Task Space Scene Analysis Module (TSSA)	Segments mesh data into planar & quadric patches; matches the operator-selected models to mesh patches; resizes models to match mesh data
Presentation & Robot Control Module (PRCM)	Receives object models after the operator has accepted them; performs path planning based on the models and interfaces to a real robot

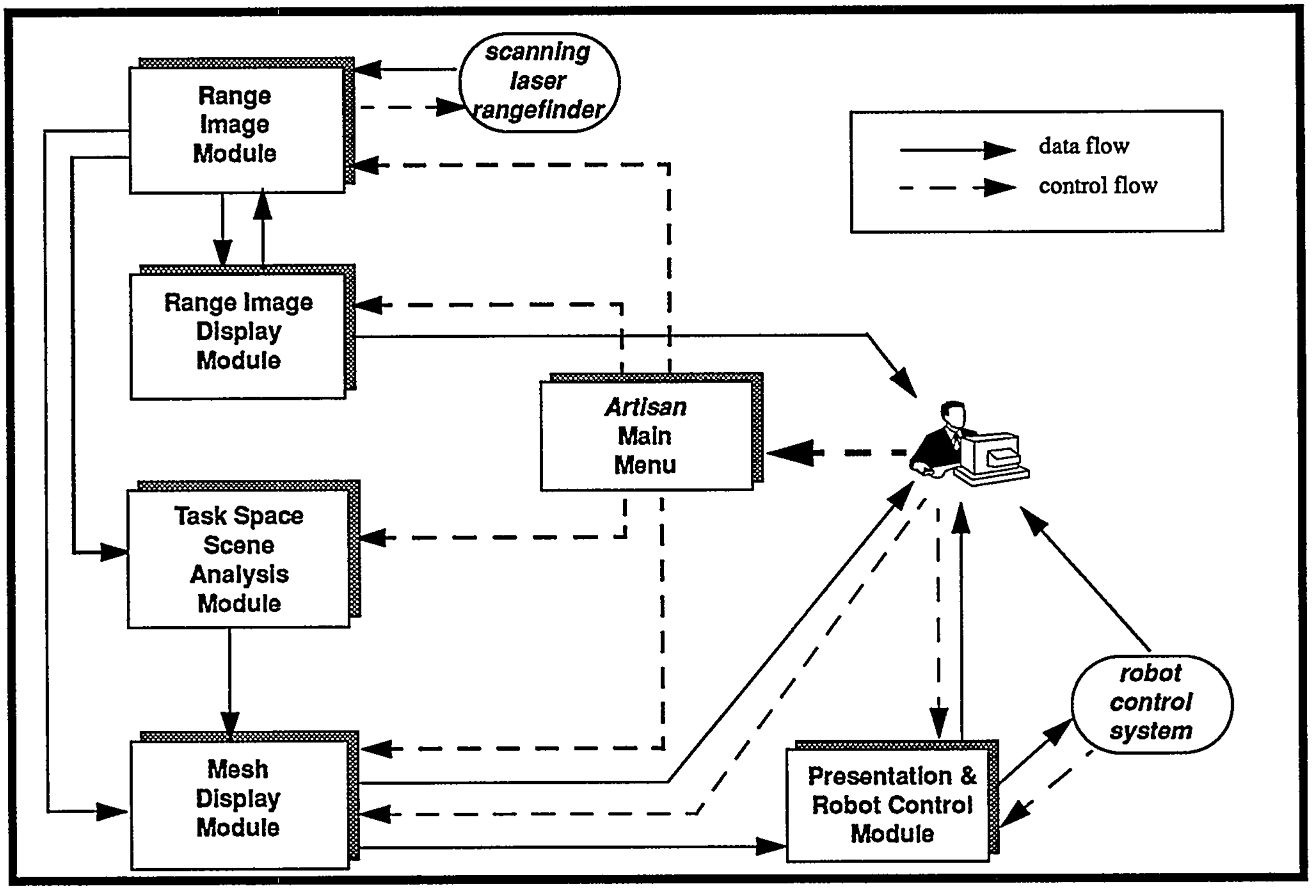

Figure 3: ***Artisan*** **system block diagram**

To develop, test and demonstrate Artisan, we constructed a mock-up containing objects representative of selective equipment removal scenarios (pipes, tanks, conduits, I-beams, etc.) as the target scene. We also developed simplistic graphical programming example to illustrate the viability of the approach for telerobotic selective equipment removal. In this case, an industrial manipulator was situated in front of the mock-up such that it could reach most of the objects in it. The same manipulator was also displayed in the robot workcell simulation using the TeleGRIP package. Objects recognized by *Artisan* then appeared in the workcell at the correct location relative to the simulated manipulator, which was then commanded to follow a trajectory along one of the object's surfaces (to simulate a spraying or washdown decontamination process). The simulated manipulator performed the task successfully, i.e., there were no collisions with other objects and the full path was within the manipulator's workspace, after which the operator instructed the real manipulator to perform the same task by downloading the trajectory from the simulation to the robot's controller. The physical robot then performed the same action.

Benefits

As shown in Figure 4, capable remote equipment is the foundation upon which all remote D&D operations rest. When combined with appropriate tooling and sensors, *Rosie* provides the ability to do a variety of remote work tasks. Building upon such fundamental capabilities, more ambitious goals of cost effectiveness can be achieved. *Artisan* represents a means to help D&D robot operators perform tasks faster and safer by enabling 3-D user interfaces and preview of actions in simulation. The same geometric information enables automatic collision checking and trajectory generation. Once those technologies are integrated to D&D worksystems, the benefits of automation will begin to be realized. The addition of advanced task planning techniques will increase cost effectiveness even further. Table 4 lists several benefits of mobile worksystems like *Rosie*; Table 3 lists benefits of layering on perception systems like Artisan in concert with robotic control techniques.

Future Work

Our efforts in Phase III will build upon the prior phases to validate our technologies in realistic operating scenarios and to create a foundation for application to DOE problems. In collaboration with RTDP we will test and demonstrate *Rosie's* core capabilities in a series of cold tests. Results from these exercises will be used to identify the need for modifications that increase its relevance, responsiveness and appropriateness for near term D&D missions. Likewise *Artisan* will be extensively cold-tested to rigorously and honestly assess its accuracy, cost of implementation, ease of use, and how it can be most effectively be applied to improve overall task performance.

In the Robotics Testing and Assessment Facility at ORNL, DOE is creating some very realistic mock-ups that essentially duplicate several DOE facilities, including the INEL "Rover" fuel processing cells, a Fernald oxidation hearth, and the CP-5 reactor at Argonne. A complete scenario for D&D of one of these facilities will be devised and detailed, and *Rosie* and *Artisan* will be combined with other elements of the D&D Robotics Program to function together as an integrated solution to execute that scenario. The resulting performance data from this full scale exercise will not only prove the viability of remote technologies, but establish both a precedent and an experience base for future hot deployment.

Acknowledgments

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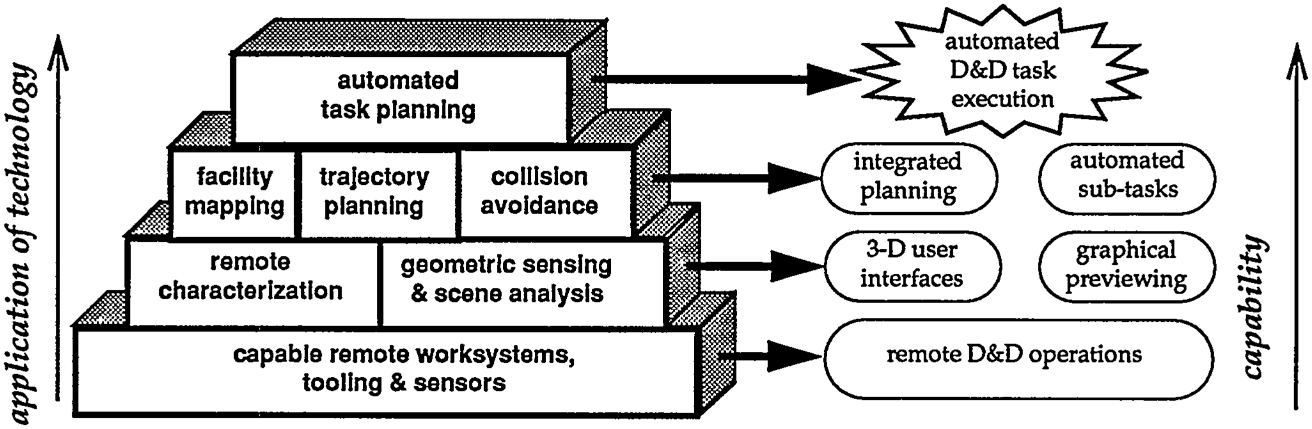

Figure 4: Evolution of D&D Robotics

Table 3: Benefits of *Artisan* and automatic controls

EM Goal	*Artisan*/Automatic Control Features	Benefits Offered
Reduction of Occupational Risks	remote mapping of facilities and structures	No human exposure to hazardous material to generate facility maps and models
	automatic obstacle avoidance and collision-free path planning	Reduced robot operator fatigue
Improved Operations	accurate 3-D model generation	Robot operator interfaces with 3-D graphical displays Development of as-built facility maps and drawings
	3-D models generated using recently gathered data	Facility models can be continually updated as D&D tasks proceed
	automatic motion control	Precise execution of intended motions
	automatic sub-task execution	Operator attention focused on task not robot
Reduction of Environmental Impact	automatic obstacle avoidance and collision-free path planning	Fewer inadvertent collisions with contaminated equipment
Reduced Costs and Time for Remediation	automatic motion control and sub-task execution	Faster execution of operator-directed actions Reduced robot operator fatigue
Minimal Secondary Waste Generation	automatic motion control and sub-task execution	Optimized movement of decontamination equipment
Applicability to Multiple DOE Sites	3-D models generated using recently gathered data	Modeling of facilities in absence of *a priori* information
	standard computing and interfaces	Compatible with D&D robot systems
Feasibility of Implementation	based on commercially available software and sensing technologies	Low risk

Table 4: *Rosie* mobile worksystem features & benefits

EM Goal	*Rosie* Worksystem Features	Benefits Offered
Reduction of Occupational Risks	remote operation of tools and decontamination equipment	Worker radiation exposure truly As Low As Reasonably Achievable
	large work envelope	No need for scaffolding to put workers at elevation
	large payload capacity	No need for complicated rigging to handle heavy objects
Improved Operations	dexterous manipulation	Manipulation of hand tools & objects comparable to human worker
	large payload capacity	Single-worker control of heavy tools
	high reliability electrically powered	Multiple shift per day operation Potential uninterrupted operation
Reduction of Environmental Impact	remote operation	*In situ* containerization of debris No waste stream of contaminated protective clothing
	forceful manipulation	*In situ* size reduction of dismantled components
Reduced Cost and Time for Remediation	remote operation of tools and decontamination equipment	Reduced worker protection requirements No unproductive worker suit-up and suit-down time
	electrically powered	Potential uninterrupted operation
Minimal Secondary Waste Generation	high force generation	*In situ* size reduction of dismantled components
	precise motion control coordinated motion	Optimal positioning decontamination equipment Selective removal of highly contaminated equipment
Applicability to Multiple DOE Sites	generous electrical and hydraulic supply output capacity	Can power wide variety of tools and decontamination equipment
	omni-directional locomotion	High maneuverability in floor facilities
	large work envelope	Floor to ceiling reach
	electrically powered	Compatible with common power sources
	modular design	Reconfigurable to suit D&D application
	standard control computing and interfaces	Compatible with other D&D robot systems
Feasibility of Implementation	based on previously demonstrated mobile worksystem for D&D	Low risk
	modular design emphasizing shelf-available commercial components	Low maintenance costs

3.8 Interactive Computer-Enhanced Remote Viewing System

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Needs

Remediation activities such as decontamination and decommissioning (D&D) typically involve materials and activities hazardous to humans. Robots are an attractive way to conduct such remediation, but for efficiency they need a good three-dimensional (3-D) computer model of the task space where they are to function. This model can be created from engineering plans and architectural drawings and from empirical data gathered by various sensors at the site. The model is used to plan robotic tasks and verify that selected paths are clear of obstacles.

This need for a task space model is most pronounced in the remediation of obsolete production facilities and underground storage tanks. Production facilities at many sites contain compact process machinery and systems that were used to produce weapons grade material. For many such systems, a complex maze of pipes (with potentially dangerous contents) must be removed, and this represents a significant D&D challenge. In an analogous way, the underground storage tanks at sites such as Hanford represent a challenge because of their limited entry and the tumbled profusion of in-tank hardware.

In response to this need, the Interactive Computer-Enhanced Remote Viewing System (ICERVS) is being designed as a software system to:

- Provide a reliable geometric description of a robotic task space, and
- Enable robotic remediation to be conducted more effectively and more economically than with available techniques.

A system such as ICERVS is needed because of the problems discussed below.

Problem

Although the basic technology to meet this need already exists, there are several areas where improvement is needed.

- Interface with Multiple Sensors. Multiple poses and a variety of sensors are necessary to properly characterize a task space. Surface mapping sensors use line-of-sight means, and more than one viewpoint is generally needed to capture sufficient 3-D data. Measurements of the physical, chemical, and radiological properties of and about the site are also needed to ensure worker safety and proper disposition of removed materials. Interfacing with such a wide range of sensors and accepting their data is currently quite difficult.

Research sponsored by the U.S. Department of Energy's Morgantown Energy Technology Center, under Contract DE-AC21-92MC29113 with Mechanical Technology Incorporated, 968 Albany-Shaker Road, Latham, NY 12110; telefax: 518-785-2420.

- Obsolete Drawings. Engineering data and architectural drawings are typically available for production facilities, but these drawings and data have often turned out to be inaccurate or unreliable because they do not incorporate the latest changes made to the site or modifications made during previous remediations.

- Data Conversion. Currently, there is no cost-effective way to convert the empirical sensor data to a surface-based 3-D CAD form used by state-of-the-art robot controllers. Customary triangularization techniques (reference 1) yield far too many polygons, while automated object recognition means are still in their infancy for applications of this complexity. Interactive techniques are handicapped by limited operator perception of the remote site.

- Unstructured Environment. The environment is unstructured and changes during the course of the remediation make the job difficult for robotics; a remediation system cannot control the presence or location of elements in its task space. Because of this unstructured nature, the operator needs a means to visualize the task space from arbitrary viewpoints and to understand the relative locations of the various hazardous elements. This visualization means must also respond in a timely way to changes in the task space geometry during the remediation.

- Model Accuracy. The accuracy of the empirical data can be enhanced by processing. Each set of empirical sensor data contains both random errors (due to, for example, noisy signals) and systematic errors (for example, in the location of the sensor). By combining data from different poses of the same sensor or from different sensors, a more accurate and reliable task space model can be created.

Solution

ICERVS is being designed as a software system that will incorporate versions of the needed improvements and will integrate them into one software module.

- Sensor Interface. ICERVS will accept, store, and display empirical data from multiple sensor types through a common interface for sensors. This interface will accept data from surface mapping sensors, such as laser radars, stereo vision, and structured light systems. It will also accept data from sensors that measure material properties, to include physical (e.g., temperature), chemical (e.g., pH), or radiological properties (e.g., gamma ray spectra). Various display options are available for the property data.

- Updated Drawings. ICERVS will accept architectural drawings of a facility and CAD models of a process site. It will display architectural drawings and provide modeling capabilities for updating 3-D CAD geometry.

- Data Conversion. ICERVS will provide interactive modeling to convert the empirical data to the polygonal form required by robotic controllers. The models will include representations of objects which the operator has perceived in the data as well as of the various waste surfaces in, for example, a storage tank. In addition to creating and editing geometric models, tools will be provided to compare a model with the data it represents. Such models can be exported to and imported from robotic controllers such as IGRIP.

- 3-D Visualization of Unstructured Environment. To expedite the operator's comprehension of the unstructured remote site, ICERVS will provide visualization of the task space, to include the empirical data as well as the geometric models. The ability to accurately display the space from an arbitrary viewpoint and to show the locations of various chemical or radiological hazards will provide the operator with insights not available from remote cameras.

- Model Accuracy. To improve the accuracy of the resulting model, ICERVS provides for the effective combining of sensor data. The overlap in scenes from one sensor at different viewpoints or from different sensors viewing the same area provides redundant information that permits the reduction of random and systematic errors in the combined data.

Technology

ICERVS is a UNIX-based system which operates on a Silicon Graphics computing workstation. It is organized as a client-server system so that an outside client can act as the supervisory controller. Functionally, the system can be described as shown in Figure 1, whose elements are discussed below.

The *common interface for sensors* currently uses an ASCII file format, with the data arranged as (x, y, z, scalar property 1, property 2, ..., property n). This format will be expanded to include array data, as from a spectrometer. Future formats will be made compatible with the format(s) selected by DoE for the D&D activities.

In the *sensor data processing* element, incoming geometric data are normalized to fit within the bounded cube used in the volumetric database. Future versions of ICERVS will provide input processing of sensor data to provide conversion of units (e.g., inches to millimeters) or to accomplish the 3-D registration among different sensor data sets.

In the *volumetric database* element, ICERVS uses octree technology to efficiently store the empirical sensor data. The octree data structure is a hierarchical representation that spatially sorts data for fast display and analysis. It uses less memory than conventional means such as uniform grids or triangle meshes. ICERVS uses the TrueSolid library (sold by Octree Corporation of Cupertino, CA) to provide this functionality.

The *analysis software* contains tools to support visualization and geometric modeling. Visualization is supported by tools that provide rotation to an arbitrary viewpoint, color mapping based on some parameter such as elevation or temperature, and the use of cut planes to view subsets of the data. Modeling tools assist in creating and editing geometric models and in comparing the model with the geometric data it is intended to represent. Additional features will be provided under future work.

The *geometric object database* stores the polygonal representation of the task space in the CAD format currently required by most robot controllers. The database contains geometric models of objects perceived by the operator as well as a triangular mesh representing the floor and other simple surfaces. ICERVS uses SGI Open Inventor software toolkit for functions involving geometric objects.

The *graphical user interface* is X-Windows based and runs on a workstation where an operator would construct a task space model for export to a robotic controller. Images can be rendered in several ways. A preview mode

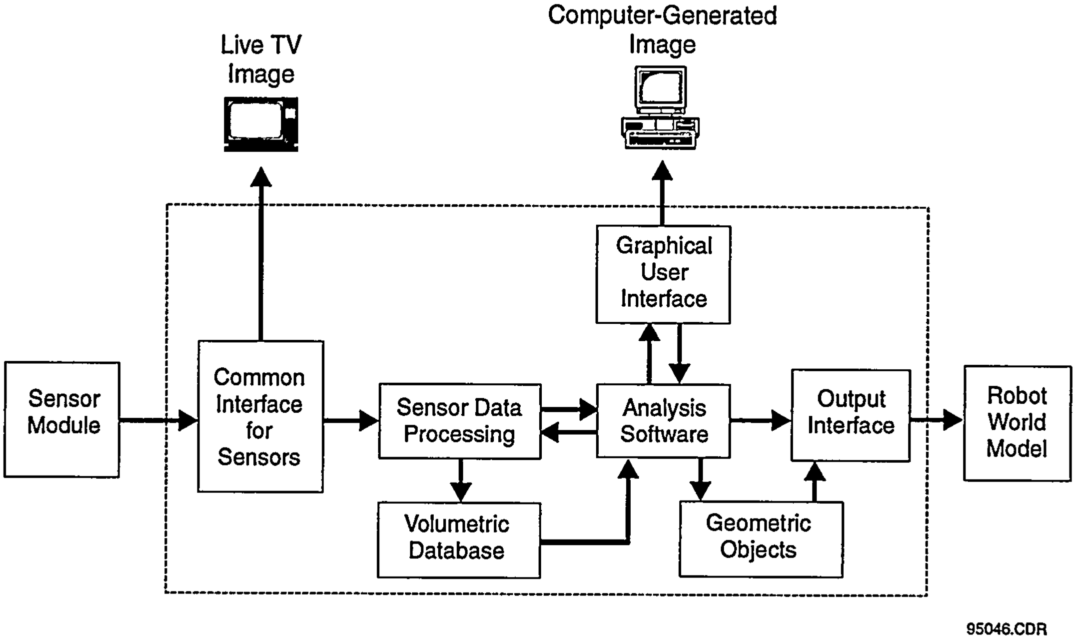


Figure 1. ICERVS Functional Model

provides a high-speed display of sensor data at low resolution. A full fidelity mode yields a high-resolution display containing both sensor data and geometric models, colored to display some property. This is accomplished by "Z buffer" merging (reference 2).

The primary role of the *output interface* is to provide task space model data to robotic controllers. Typically, this data is formatted in files such as IGRIP part files.

If a *remote camera* is provided with an appropriate pan-and-tilt mechanism, then a live TV image provided at the operator's console can be matched by a *computer-generated image* of the same scene. In this way, insights gained from remote cameras can be easily incorporated into the ICERVS model. In the future, such video scenes can be stored directly in the database.

Accomplishments

The development of ICERVS is organized in three phases, as shown in Figure 2, and development is currently more than halfway through the third phase. The objective of the first phase was to demonstrate that a volumetric database could store real sensor data and that the data could be viewed and segments extracted by cut planes. The program was fortunate to have access to data taken by Oak Ridge National Laboratory (ORNL) as they mapped the surfaces in the K-65 tanks at Fernald, OH, both before and after application of a bentonite cap (reference 3). Figure 3a shows a top view of the combined data as well as the location of a pair of cut planes. Figure 3b shows a side view of the data between the cut planes; the profiles of the waste surface and of the top of the cap are clearly visible.

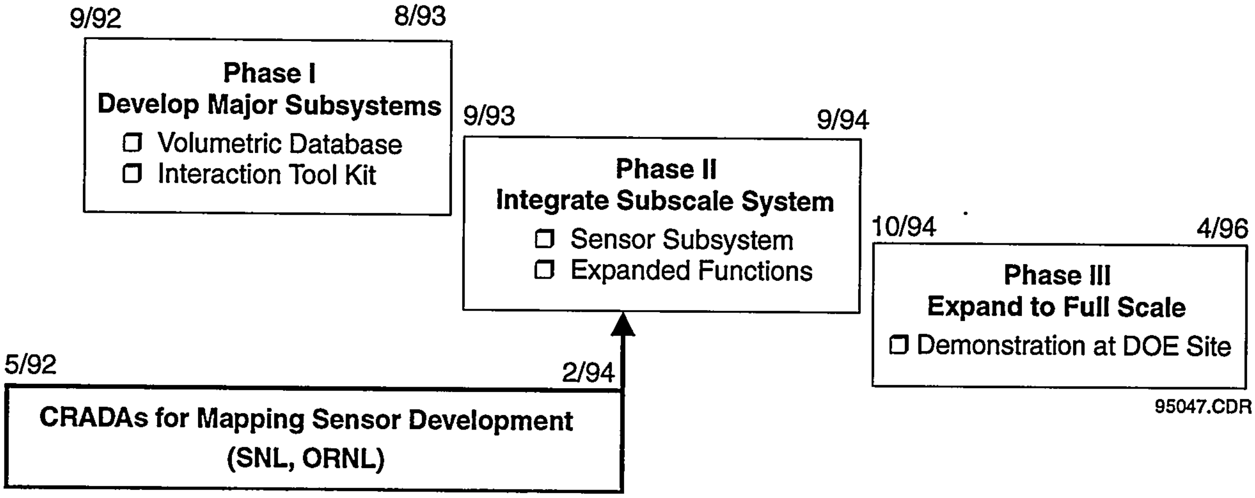
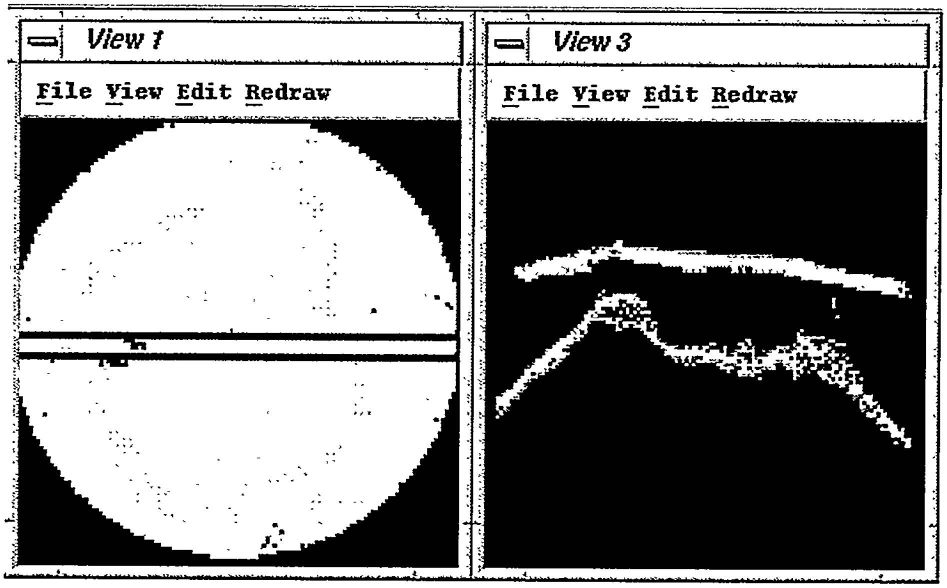


Figure 2. ICERVS Program Structure and Schedule



(a) (b)

Figure 3. 3-D View Fernald Data with Cut Planes

The objective of the second phase was to demonstrate a subscale system and to show the integration with a structured light sensor developed earlier on two CRADA projects. As part of the CRADA, a test facility was constructed to simulate a storage tank, containing a wall segment as well as three pallets of simulated waste (Figure 4). The structured light sensor was mounted above and to the right of the camera that took Figure 4. The data it gathered was transferred to ICERVS and could be viewed there from any viewpoint. It was colored to show elevation. When data are sparse, an interpolated version of the data can be created (Figure 5) to improve viewer comprehension. As part of this phase, simple geometric models were created and fit to objects created in the sensor data.

Feedback from the Phase II demonstration indicated that the user interface was perceived to be slower and more awkward than in other systems. Thus, development of a full-scale system for Phase III included a change in the user interface. The underlying architecture was modified to eliminate keystrokes and make operation simpler. Steps were also taken to improve system response, leading to the creation of the two rendering modes described earlier. The earlier test facility is shown in the preview mode shown in Figure 6, while the comparable full fidelity mode is shown in Figure 7. Because of the richness of the sensor data, the preview mode is quite informative. However, the presence of geometric models and the coloring make the full fidelity model more useful. Two additional (internal) releases of the software are planned by the end of the contract; these will incorporate the additional features described later in this paper under Future Activities.

Benefits

ICERVS can provide a number of benefits when applied to D&D remediation or the remediation of waste storage tanks.

- Better understanding of task space. ICERVS can store, update, and display all of the data pertaining to a specific site. It can provide 3-D visualization of the task space for data analysis and for robotic task and path planning.

- Faster evaluation of site characteristics. By providing one repository for site data, ICERVS eliminates the time spent in seeking out and verifying data. It provides a ready way to compare, interpret, and merge data from different times or from different sensors.

- Safer operation of remote robotics. ICERVS enhances the safety of telerobotic operation by providing more flexible and informative 3-D views of the task space. It enhances the safety of autonomous operations by providing more accurate and current models of the task space.

- Cheaper remediation. ICERVS reduces remediation cost by enabling faster assessment of site characteristics, by facilitating more efficient task and path planning, and by improving efficiency in all modes of robotic operation.

Future Activities

Future work includes the remainder of Phase III and those activities needed to develop ICERVS as a viable product offering.

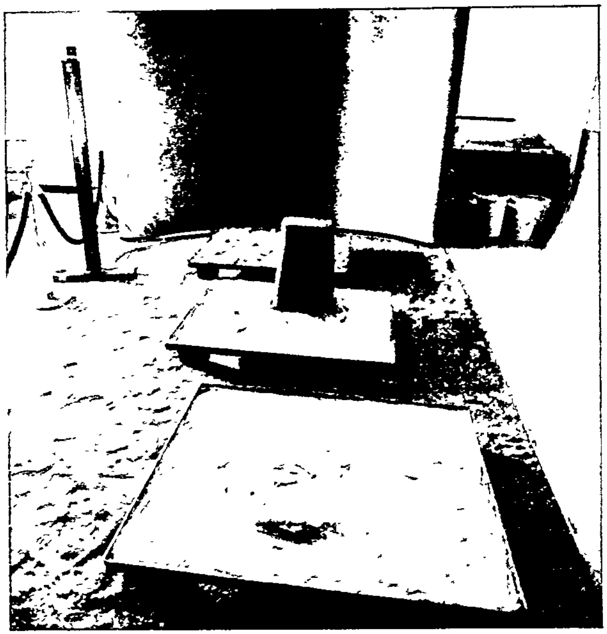


Figure 4. MTI Simulated Storage Tank Facility

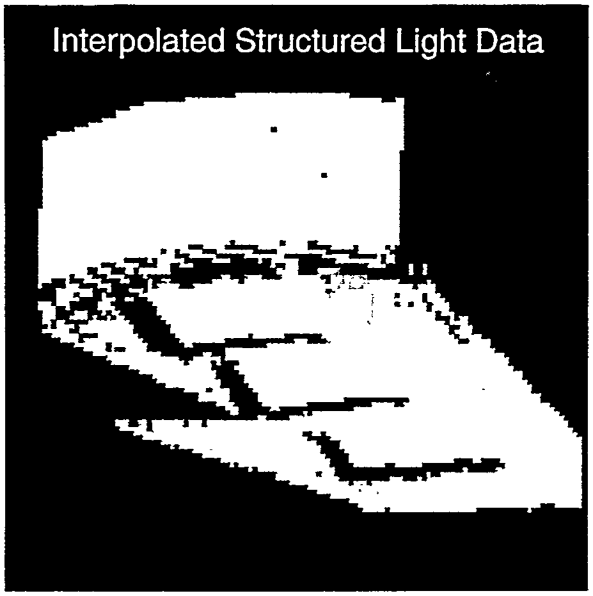


Figure 5. Interpolated Structured Light Data from MTI Simulated Storage Tank Facility

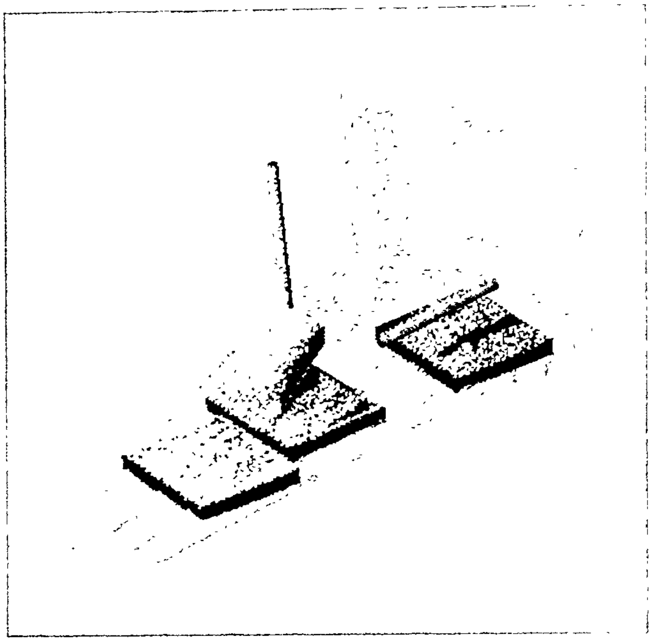


Figure 6. Preview Mode of Display

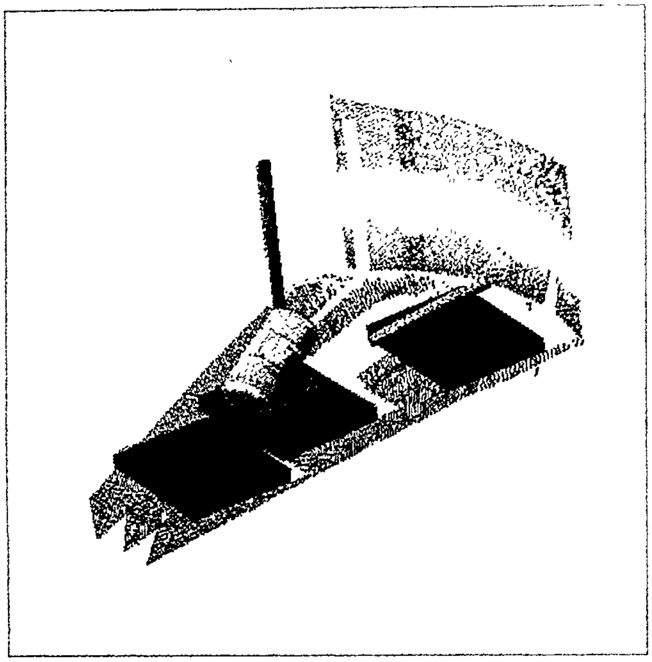


Figure 7. Full Fidelity Mode of Display

Phase III

The additional features to be incorporated during Phase III include:

- The ability to accept and store nonscalar properties. Array data such as that from gamma ray spectrometers or frames of video data are of interest.
- The ability to accept, store, and display 2-D architectural plans.
- A suite of analysis tools, to include software to fit a cylinder to a segment of volumetric data, a connectivity tool to determine the region of points connected to a seed point, and a pipe tracing tool to identify disconnected segments of the same straight pipe.
- The display of live video on the same computer monitor that displays the task space data and models.
- Conversion of output geometric models (CAD files) into IGRIP part file format.

In addition, there are two demonstrations in this phase of the project.

- There will be a full-scale demonstration in a simulated underground storage tank at Hanford, WA. This demonstration will use a structured light system that will be delivered to Hanford by MTI.
- A second demonstration will occur at ORNL in conjunction with the facility mapping program being conducted there as part of the D&D development effort. Sensor data will be provided by a variety of sensors, particularly a laser radar system.

Other

Near-term objectives involve the development of data fusion techniques based on occupancy maps (reference 4). Further, MTI will create a subset of ICERVS to provide desired data visualization means to the MTI structured light system referred to as the Topographical Mapping System, of which a first prototype is being supplied to Hanford. Longer-term objectives include actual deployment of ICERVS on a D&D mission. MTI will pursue the market to ensure that ICERVS has the desired set of features and then assess its effectiveness in beta test sites.

Acknowledgements

It is a pleasure to recognize the support provided by Mr. Vijendra Kothari, the METC Contracting Officer's Representative, and by his predecessor, Mr. Gary Nelkin. The period of performance on this contract will be approximately three and one-half years. Its completion assumed the availability of technology developed under CRADAs with Sandia National Laboratories (SNL) and ORNL. The project has received support from three focus areas, namely, underground storage tanks, D&D, and buried waste.

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3.9 Three Dimensional Characterization and Archiving System

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1.0 Introduction

The Three Dimensional Characterization and Archiving System (3D-ICAS)[1] is being developed as a remote system to perform rapid in situ analysis of hazardous organics and radionuclide contamination on structural materials. Coleman Research and its subcontractors, Thermedics Detection, Inc. (TD) and the University of Idaho (UI) are in the second phase of a three phase program to develop 3D-ICAS to support Decontamination and Decommissioning (D&D) operations. Accurate physical characterization of surfaces and the radioactive and organic is a critical D&D task. Surface characterization includes identification of potentially dangerous inorganic materials, such as asbestos and transite.

Real-time remotely operable characterization instrumentation will significantly advance the analysis capabilities beyond those currently employed. Chemical analysis is a primary area where the characterization process will be improved. Chemical analysis plays a vital role throughout the process of decontamination. Before clean-up operations can begin the site must be characterized with respect to the type and concentration of contaminants, and detailed site mapping must clarify areas of both high and low risk. During remediation activities chemical analysis provides a means to measure progress and to adjust clean-up strategy. Once the clean-up process has been completed the results of chemical

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analysis will verify that the site is in compliance with federal and local regulations.

The development of the field operable in situ analysis capability of the 3D-ICAS will result in a significantly improved capability to perform analyses for trace organic compounds and radionuclides by providing real-time quantitative results on-site. This will greatly improve the effectiveness of Department of Energy (DOE) response to identified sites by streamlining the entire beginning-to-end process from the initial contamination surveys, through monitoring the progress of site restoration efforts, and determining when regulatory standards have been met. The field operable remotely directed analysis instrumentation of 3D-ICAS will have great impact in terms of:

- Improving the quality and efficiency of site clean-up activities
- Reducing health and safety risks to the survey workers
- Reducing the associated cost and time required for remediation
- Reducing waste generation

2.0 The Approach

The 3D-ICAS system robotically conveys a multisensor probe near the surfaces to be inspected. The sensor position and orientation are monitored and controlled using coherent laser radar (CLR) tracking. The CLR also provides 3D facility maps which establish a 3D "world view" within which the robotic sensor system can operate.

The 3D-ICAS fills the need for high speed automated organic analysis by means of a gas chromatographs-mass spectrometer sensor which can process a sample, without direct contact, accomplishing detection and fine grain analysis of regulatory concentrations (EPA 1987 spill cleanup policy: 1μg/ $10cm^2$ for high use interior building surfaces) in approximately one minute. This compares with traditional GC-MS laboratory analysis methods which involves sample preparation and waste generation, and take hours per sample for analysis. The 3D-ICAS GC-MS sensor extracts volatile organics directly from contaminated surfaces without sample removal, then uses multiple stage focusing to accomplish high time resolution insertion into a high speed gas chromatograph. Detection and additional discrimination are provided by a final stage time-of-flight mass spectrometer. This high speed process replaces sample collection and transport and hours of solution preparation before injection into an ordinary GC-MS which typically has a 45 minute run time.

The radionuclide sensors of the 3D-ICAS multisensor probe combines a, ß, and g counting with energy discrimination on the a channel. This sensor combination identifies and quantifies isotopes of specific DOE interest of uranium, plutonium, thorium, technetium, neptunium, and americium to regulatory levels in approximately one minute.

The Molecular vibrational spectrometry (MVS) sensor of the multisensor probe is used to characterize substrate material such as concrete, wood or asbestos. The surface composition information provided by the MVS can be used to provide estimates of the depth of contamination and to optimize the analysis performance of the other contamination detection sensors. In addition, the materials composition information, combined with the surface geometry maps provided by the coherent laser radar, will provide a more complete three dimensional world view to be used to plan and execute robotic D&D operations.

The 3D-ICAS will scan operator designated areas with a full sensor set or a selected subset at a designated sample density. The 3D-ICAS will plan and laser track the sensor trajectory to assure efficient, close sensing without surface contact.

The 3D-ICAS sensor output and contamination analysis along with CLR position information will be available for real-time monitoring immediately after each one minute sample period. After a surface mapping operation is completed, 3D-ICAS will provide three dimensional displays showing contours of detected contaminant concentrations. The 3D-ICAS will further provide permanent measurement data and contaminant level archiving, assuring data integrity and allowing straightforward regulatory review of the characterization process before and after D&D operations.

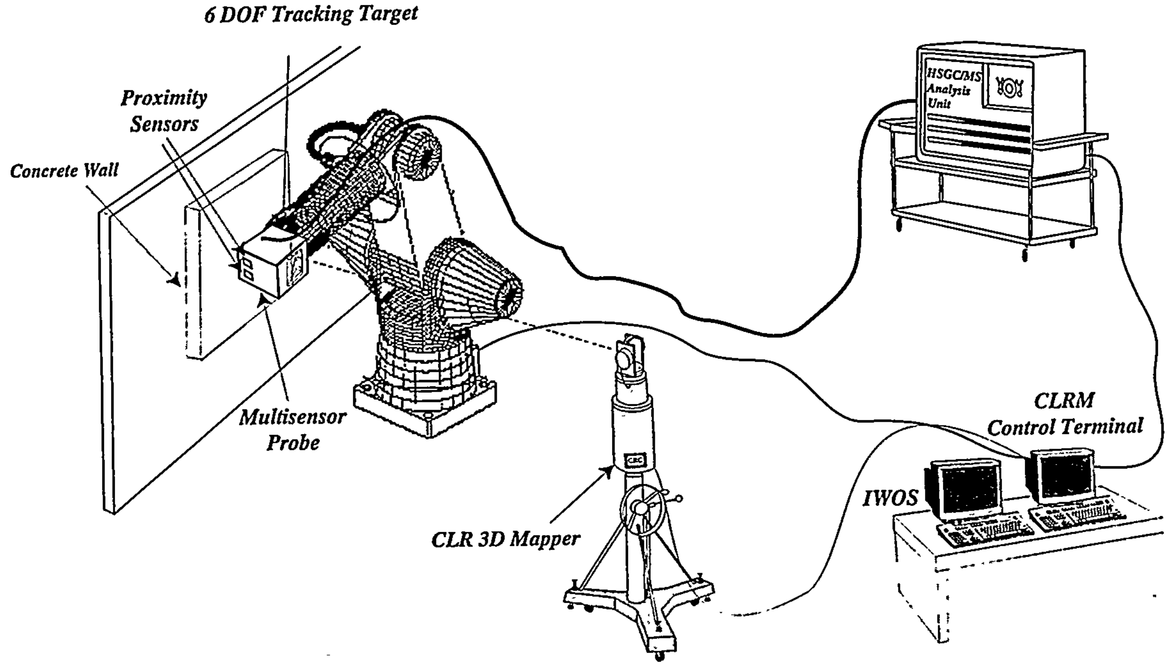


Figure 3-1. 3D-ICAS Phase II Configuration

The 3D-ICAS Phase II demonstration configuration is shown in Figure 3-1.

Figure 3-2 shows the interconnection of the major subsystems. The links are a combination of serial for slow speed command and control, TCP/IP network for file based data file storage and communications, and dedicated analog and digital links for high bandwidth functions such as servo loops, gas chromography, and laser radar transmission and reception.

The Integrated Workstation communications with the TCP/IP network based link to the CLR control computer. This link provides all hardware control functions and database access for the Integrated Workstation.

The CLR computer serves as a focal point for hardware control. Serial links are used between the CLR computer and the Multisensor Probe electronics control computer to provide control and status information. Data from the Multisensor Probe is stored directly on a TCP/IP network drive. Serial links are also used to control the robot arm position. The 3D-ICAS Phase II demonstration multisensor probe is shown in Figure 3-3.

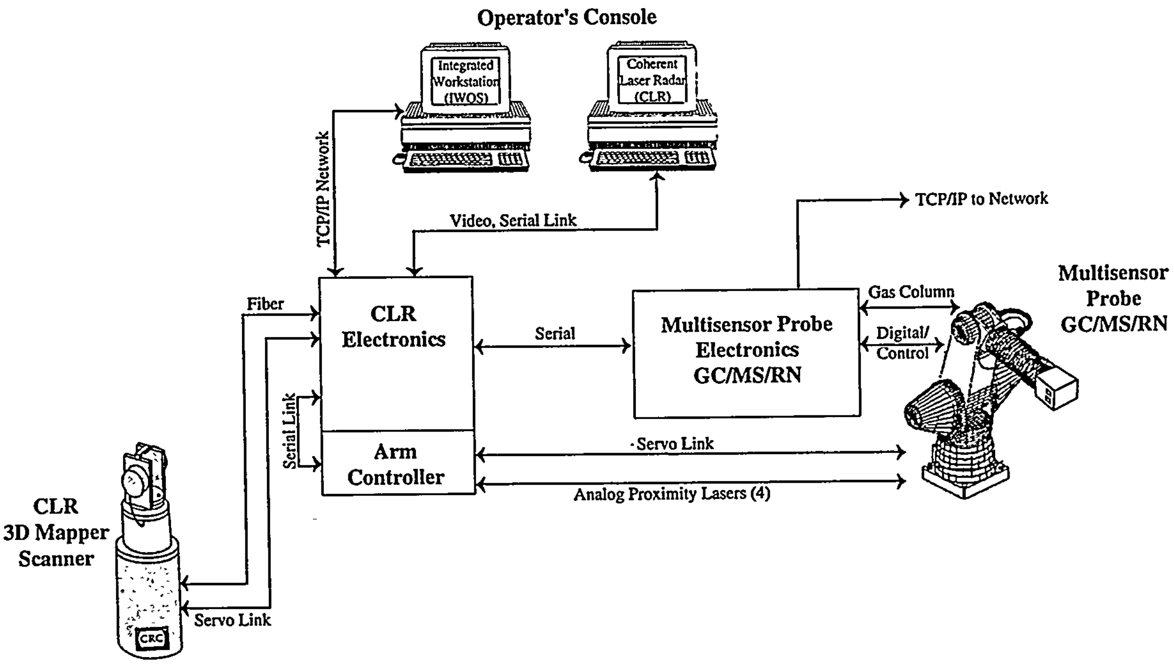


Figure 3-2. 3D-ICAS Phase II Demonstration Interfaces

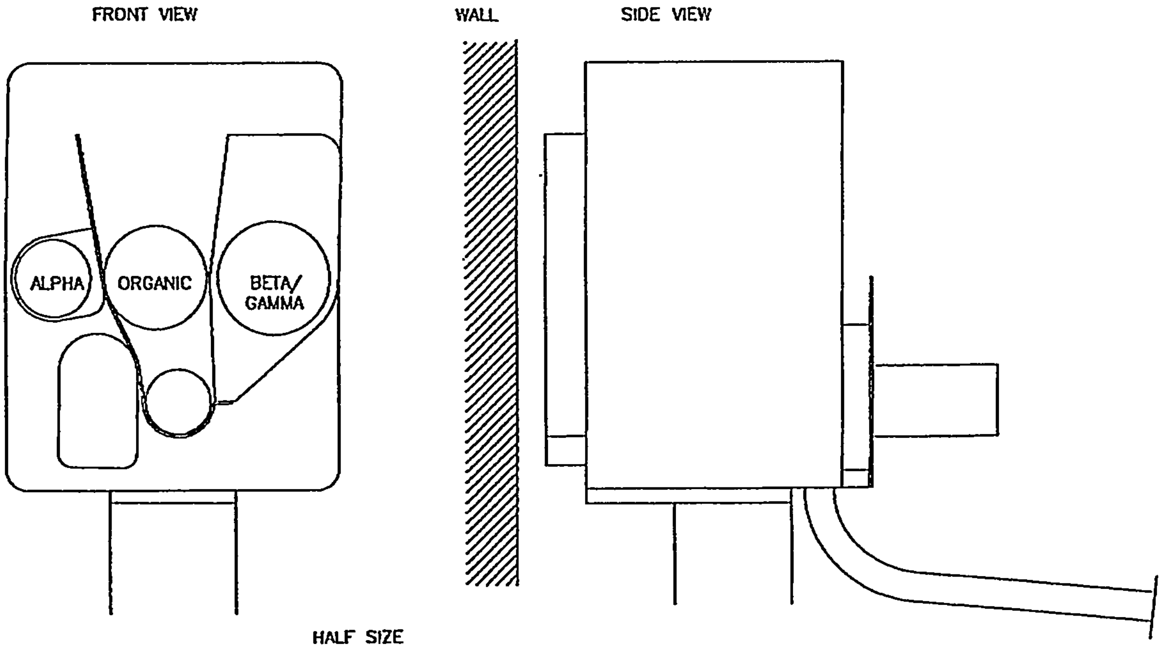


Figure 3-3. Block Diagram of the Phase II Demonstration Multisensor Probe

3.0 The Development Program

The 3D-ICAS is being developed in a three phase 32 month program. The current Phase II includes integration of the GC and MS subsystems, integration of the multisensor probe with the robot arm and the coherent laser radar tracker. The Phase II 3D-ICAS demonstration, involving contamination surface mapping with a CLR guided, robotically maneuvered multisensor probe, will take place in November, 1995.

The Phase III development program will integrate and test the total system in a mobile field prototype and demonstrate the systems at one or more DOE sites.

A substantial part of the Phase II effort is the integration of the 3D CLR mapping and robotic sensor positioning and tracking capability with the sensor data output in a common coordinate frame so that 3D archiving of contamination surveys can take place. This paper will not discuss detailed system integration issues, but will discuss areas of developing technical performance of the GC-MS sensor, the radionuclide sensor, the MVS sensor, and the CLR 3D mapper and 6DOF end effector tracker which are of interest to the 3D-ICAS user.

3.1 The GC-MS Sensor Subsystem

The 3D-ICAS GC subsystem consists of an automated surface sampling, preconcentration and transport subsytem, high speed GC separation capability, (greater than 100 peaks per minute and fast detector electronics. These aspects of the high speed GC technology were successfully demonstrated in Phase I.

The high speed GC system consists of a focusing module and a chromatography module. The focusing module consists of a short length of GC column inside its metallic sheath, coupled to a cooling source. The focusing module is maintained at low temperature while the sample is introduced in a stream of air or carrier gas. At low temperature, the analytes are retained on the stationary phase of the GC column in the focusing module.

After sample focusing, the sample is injected by rapid heating of the focusing module. Sample injection is achieved with tight spatial coherence in less than 50 milliseconds, which helps to produce sharp chromatographic peaks. The GC system uses two sequential focusing stages, to provide additional selectivity and tighter injection onto the chromatograph. After the sample is injected from the second focusing stage, the GC column is then heated through a rapid, tightly-controlled temperature program. Controlled heating rates in excess of 1000 C per second can be achieved. With this technology, the accessible heating rate is no longer a limitation to chromatographic performance. The speed and resolution of the chromatography is determined by processes such as carrier gas flow rate, and molecular diffusion over very short distances, which are inherently fast.

A GC breadboard test system for the current effort was developed to contain all the required elements. The breadboard system provided the cryofocusing elements ("cold spots"), sample switching valves, temperature controller, and analog and digital electronics for high speed temperature programming. Externally-mounted experimental GC columns were interfaced to the breadboard chassis. The high speed temperature programming electronics were modified extensively to accommodate column lengths up to 7 meters, and chromatographic elution times up to 40 seconds.

In Phase II, the high speed GC was integrated with a time-of-flight mass spectrometer and analysis software development to achieve rapid, definitive quantitative measurement of trace level organic contamination in the presence of a complex background, as can be expected in DOE facilities.

A block diagram of the High Speed Gas Chromatograph/Mass Spectrometer Sensor is shown in Figure 3-4.

A time-of-flight mass spectrometer (TOF-MS) system was chosen based on its ability to main-

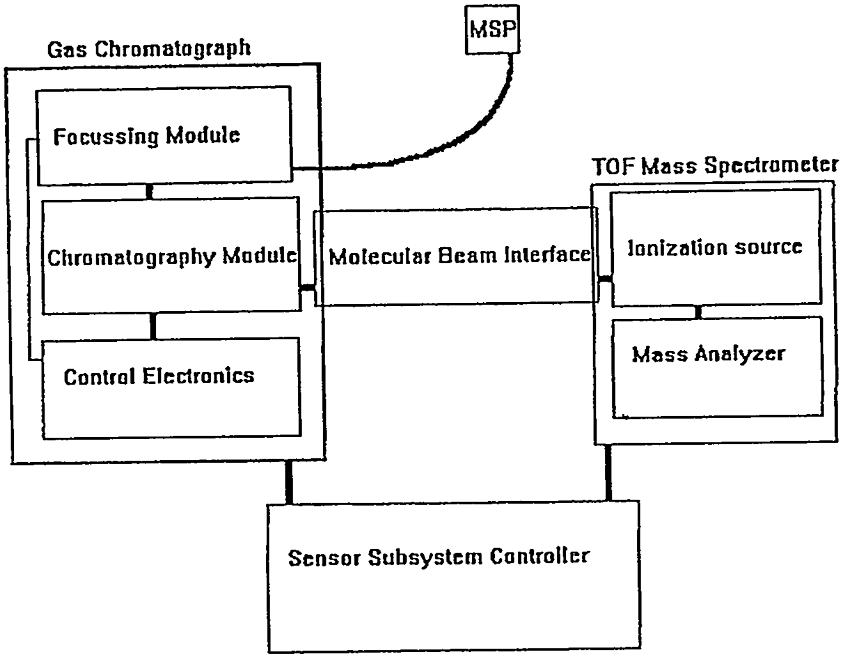


Figure 3-4. Block Diagram of the High Speed Gas Chromatograph/Mass Spectrometer

tain a high data throughput. Early on it was determined that effective High Speed GC-MS would require a detector system capable of 10 millisecond or less response time. The time-of-flight system is the only mass analyzer capable of such a data rate.

The TOF-MS is operationally simple and reliable. All mass spectrometers have some form of interface to the outside world, all require high vacuum chambers, all require ionization source and all require detectors. In general what differentiates mass spectrometers is the technology for mass analysis. For 3D-ICAS, a very fast detector is require to fully utilize the capabilities of the high speed GC technology and hence be capable with the surface contamination mapping function. A time-of-flight mass analyzer was chosen because it can provide full mass spectral scan (to 600 amu) for every ionization event. Other types of mass spectrometers cannot generate full mass spectra at these high rates. The full mass spectra is necessary for identification of the unknown components of a sample through library searching. Additionally, existing TOF technology will allow high repetition scan rates for implementing high duty cycle, high speed operation of the TOF MS detector when coupled to the High Speed GC.

Ionization events were generated by the MS at a 10 KHz repetition rate, 128 events were summed into one data point along the GC axis. This GC sampling period is sufficient to clearly distinguish the GC peak and deconvolute peaks, if necessary. For a GC peak that is 100 milliseconds full wide at half height (FWHM) approximately 1000 full scan mass spectra are recorded across the full width at half maximum. A signal to noise enhancement advantage can be achieved by summing the mass spectra into discrete sampling windows. For the current system's GC peaks on the order of 100 milliseconds wide a data sample period of approximately 10 milliseconds will clearly define the GC peaks. Other mass analysis technologies cannot provide mass spectra at these high repetition rates.

One of the most critical parts of a GC-MS is the interface between the systems. The interface plays the important role of accommodating the pressure drop from the GC column exit to the MS ionization source and enriching the concentration of the analyte in the carrier gas after passing through the interface into the mass spectrometer. The requirements for a GC-MS interface are high transfer efficiency, no impact on the GC separation, no degradation of compounds, no preferential removal of compounds or chemical functional groups. Of all the techniques available the molecular beam interface was chosen as optimal for this fast analysis system.

The directed beam of molecules has an advantage over conventional thermal, diffusive methods for introduction of the molecules into the ionization source. The concentration of molecules within the volume of space that the electron gun molecular beam crosses is close to a maximum. Thus, probability of ionization per unit time is increased, thereby increasing sensitivity.

3.1.1 GC/MS Processing

A block diagram of the GC/MS and radionuclide sensor processor is shown in Figure 3-5. The overall function of these components are to acquire the time-of-flight MS data as a function of GC elution time, control the GC operation, control the radionuclide detectors for data collection and processing. The acquired data from the GC/MS sensor is processed into mass spectra and total ion chromatogram. The GC/MS data are processed through a National Institutes of Standards (NIST) Mass Spectral Database, response factor calibration and sample classification algorithms for further data reduction.

The electronics technology for the HSGC-MS data acquisition and processing were the high speed transient digitizer, high speed data transfer bus and high speed digital signal processors (DSP). The

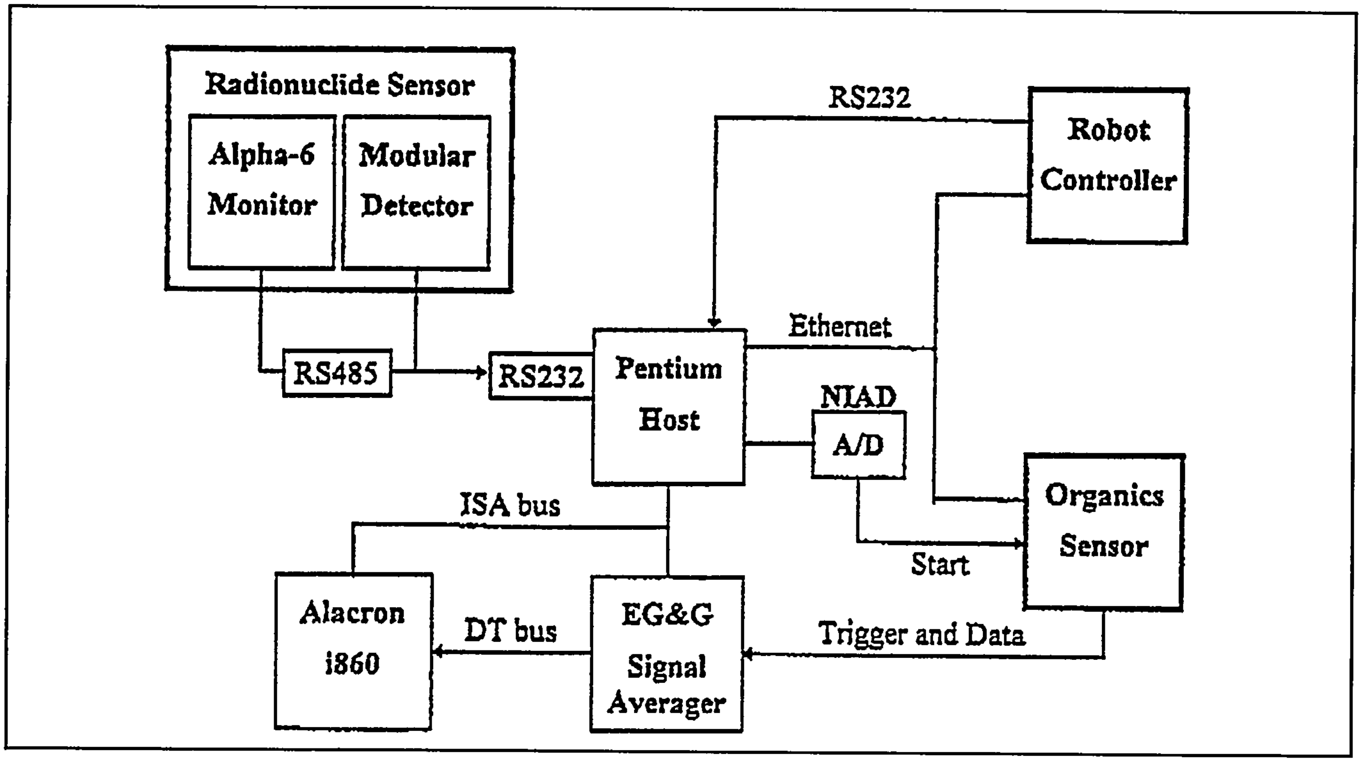


Figure 3-5. Block Diagram of the GC/MS and Radionuclide Sensor

MS events occur on a nanosecond time scale. A mass spectral scale from 35 to 600 amu requires approximately 40 microseconds of data. To achieve a mass spectral resolution of one at 600 amu requires 5 to 10 nanosecond time discrimination.

3.1.2 GC/MS Test Results

The tests were carried out using the breadboard GC-MS to optimize the performance characteristics and demonstrate overall function. The function included sampling from surface, GC separation and MS detection, identification and quantification.

The results of a typical GC/MS ran are shown in Figure 3-6. A 1 µL injection of a mixture of PCBs (arochlors 1221, 1248, and 1254) was used in this test. The chromatographic conditions were as follows: 6 meter DB-5 and helium carrier gas at 100cm/sec. The temperature program was as follows: room temperature to 280C in 17 seconds then hold at 280 C for 5 seconds. The overall cycle time was 30 seconds. About 35 peaks are resolved, the detected compounds list indicates that chlorinated biphenyls compounds have been identified. Shown in Figure 3-7 is a portion of the list of compounds identified through the NIST library search during the analytical run. The list contains the retention time the search confidence level and the name of the compound identified. The list will be expanded to include the chemical classification and the amount of compound detected as the software is completed in the project. It is expected that the detection sensitivity of this instrumentation will meet or exceed the regulatory levels of 1 µg/10 cm2 for high use interior building surfaces (EPA Spill Cleanup Policy, 1987). Figure 3-8 shows the mass spectrum used in the library search for the compound detected at 24.8 seconds. A comparison of Figure 3-7 and 3-8 indicates that the detected compound was identified as trichlorobiphenyl

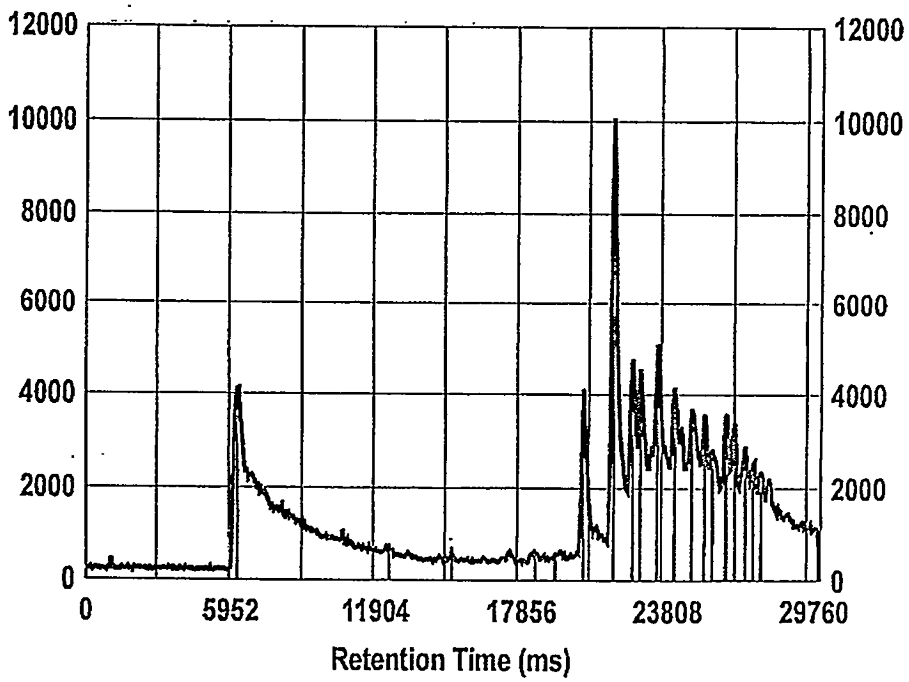


Figure 3-6. Total Ion Chromatogram for a Mixture of PCBs Analyzed using the High Speed Gas Chromatograph/Mass Spectrometer

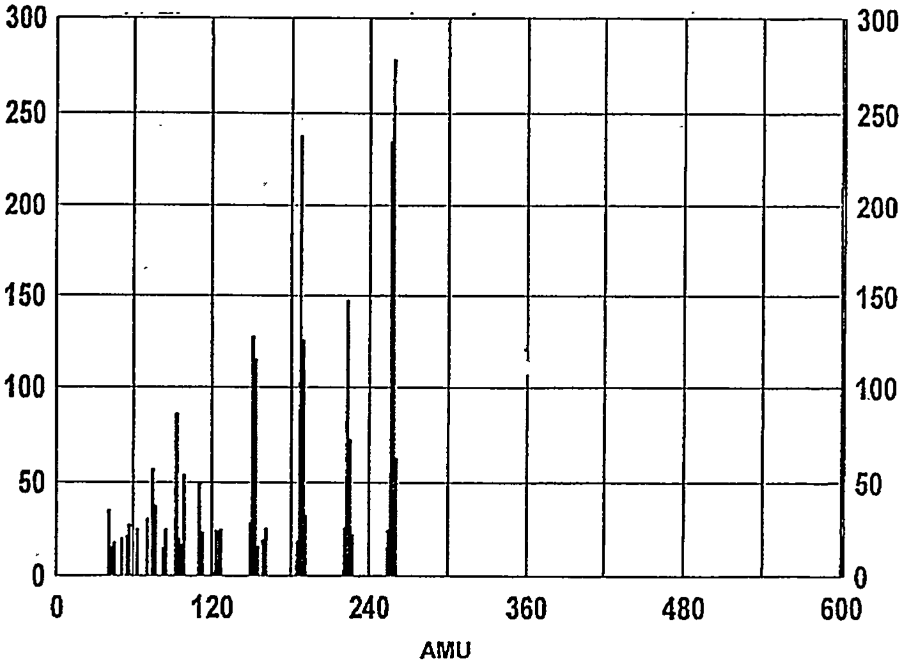
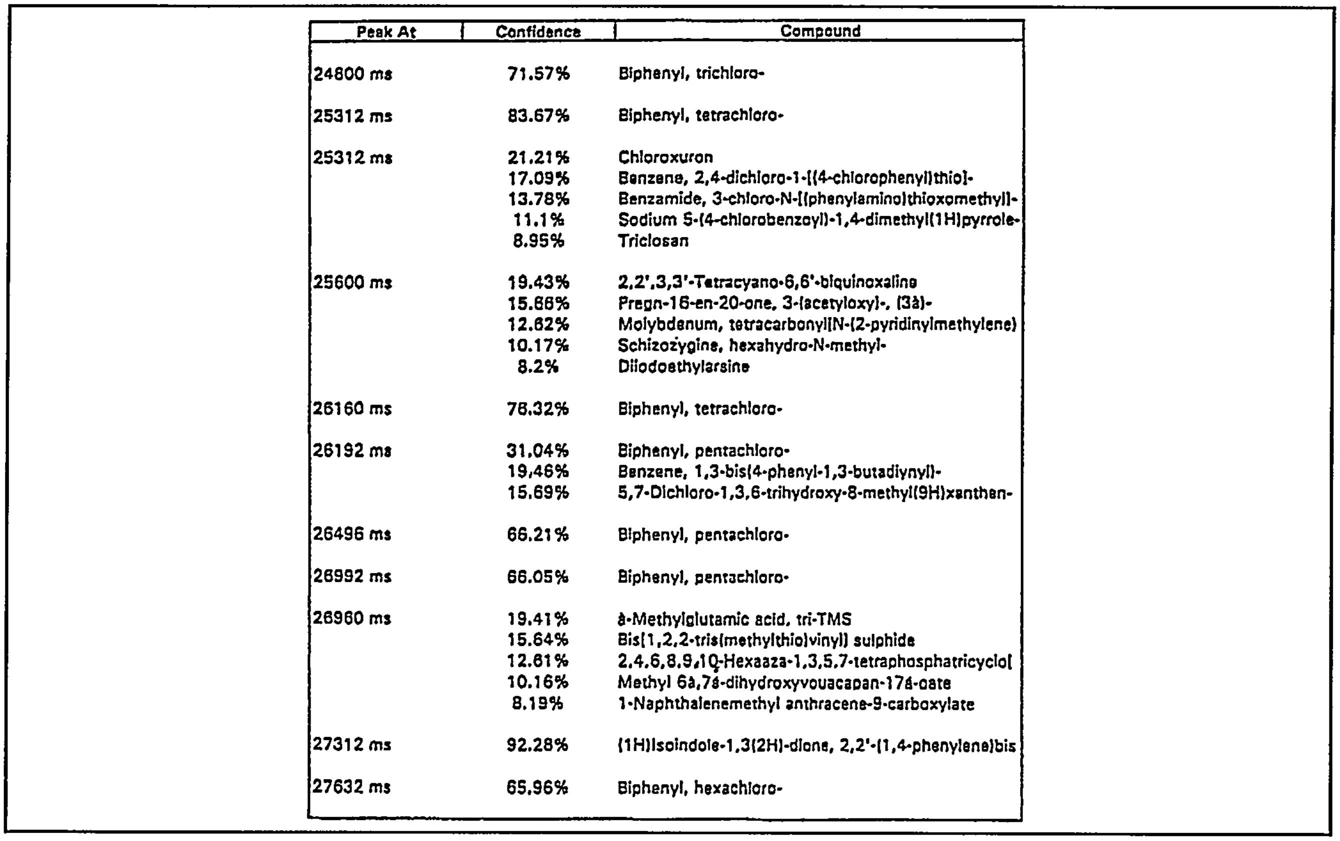


Figure 3-7. Mass Spectrum Measured at 24.800 Seconds



Peak At	Confidence	Compound
24800 ms	71.57%	Biphenyl, trichloro-
25312 ms	83.67%	Biphenyl, tetrachloro-
25312 ms	21.21%	Chloroxuron
	17.09%	Benzene, 2,4-dichloro-1-[(4-chlorophenyl)thio]-
	13.78%	Benzamide, 3-chloro-N-[(phenylamino)thioxomethyl]-
	11.1%	Sodium 5-(4-chlorobenzoyl)-1,4-dimethyl(1H)pyrrole-
	8.95%	Triclosan
25600 ms	19.43%	2,2',3,3'-Tetracyano-6,6'-biquinoxaline
	15.66%	Pregn-16-en-20-one, 3-(acetyloxy)-, (3à)-
	12.62%	Molybdenum, tetracarbonyl[N-(2-pyridinylmethylene)
	10.17%	Schizozygine, hexahydro-N-methyl-
	8.2%	Diiodoethylarsine
26160 ms	76.32%	Biphenyl, tetrachloro-
26192 ms	31.04%	Biphenyl, pentachloro-
	19.46%	Benzene, 1,3-bis(4-phenyl-1,3-butadiynyl)-
	15.69%	5,7-Dichloro-1,3,6-trihydroxy-8-methyl(9H)xanthen-
26496 ms	66.21%	Biphenyl, pentachloro-
26992 ms	66.05%	Biphenyl, pentachloro-
26960 ms	19.41%	à-Methylglutamic acid, tri-TMS
	15.64%	Bis[1,2,2-tris(methylthio)vinyl] sulphide
	12.61%	2,4,6,8,9,10-Hexaaza-1,3,5,7-tetraphosphatricyclo[
	10.16%	Methyl 6à,7á-dihydroxyvouacapan-17á-oate
	8.19%	1-Naphthalenemethyl anthracene-9-carboxylate
27312 ms	92.28%	(1H)Isoindole-1,3(2H)-dione, 2,2'-(1,4-phenylene)bis
27632 ms	65.96%	Biphenyl, hexachloro-

Figure 3-8. A Portion of the HSGC-MS Processed Data

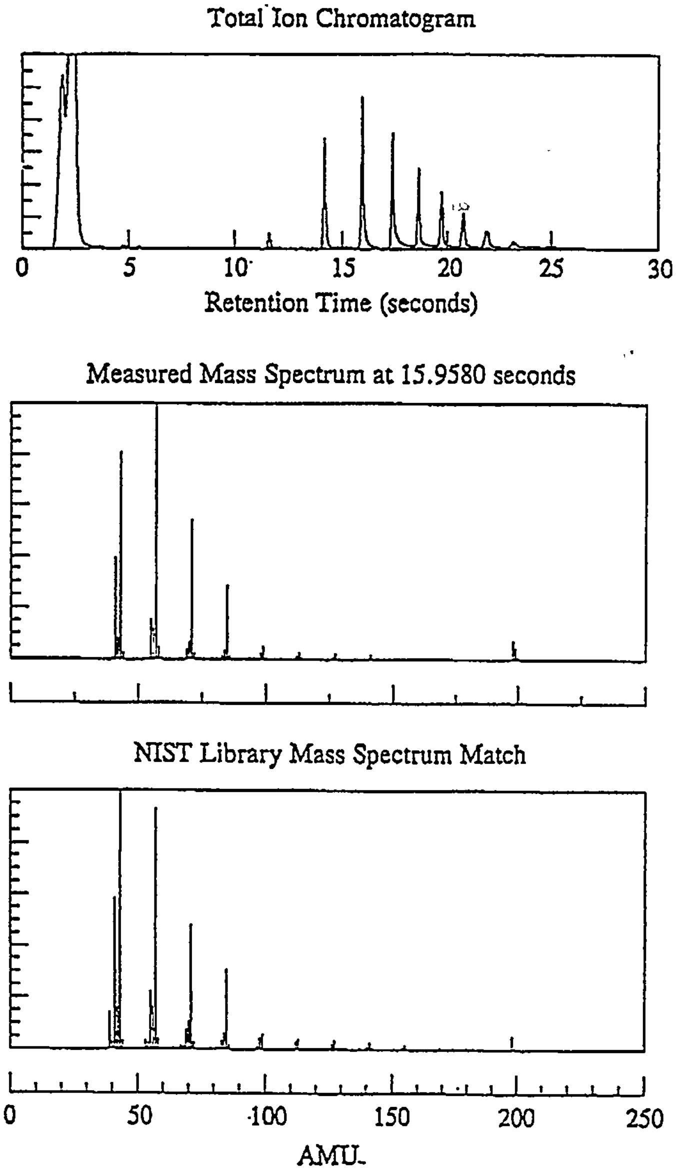


Figure 3-9. HSGC-MS Analysis of Diesel Constituents Showing Total Ion Chromatogram, Mass Spectrum and Library Match

component of the PCB sample. These initial results indicate both hardware and software are properly functioning for the HSGC-MS. These data formats are compatible with the archive and mapper systems. The datafiles will be transferred over hardwire link upon integration with the mapper subsystem.

In a separate experiment, 100 ng sample of diesel range organics (DRO) were analyzed. The data shown in Figure 3-9 indicates a close match between the measured mass spectrum and the library. The implication is that the HSGC-MS is operating under conditions which provide data that are comparable to conventional GC/MS.

The tests indicated that the High Speed GC/MS functions in a manner which allows effective determination of sample constituents in less than 40 seconds.

3.1.3 Summary High Speed GC-MS Analysis

The Phase II objective was to develop the necessary hardware and procedures for high speed GC-MS analysis of the toxic organics, using analog compounds similar to PCBs. The goal was GC separation and mass spectrometer identification in one minute to regulatory limits. The high speed GC-MS sensor is expected to accomplish GC separation, MS identification, MS library database search for identification and quantitation in less than 60 seconds. This capability will satisfy the Phase II success criteria of sampling analysis and data processing for GC/MS. Coupled with the Multisensor probe for automated sample extraction, preconcentration and transport this subsystem has established a new benchmark for speed and performance in organic analysis.

3.2 Radionuclide Sensors

For the majority of radioisotopes of DOE interest two detectors were determined to best meet the functional needs. This was because most isotopes were alpha emitters (Am, Th, Pu, U), and one was a beta emitter (Cs). The 3D-ICAS radionuclide sensor system includes both an Eberline diffused junction silicon detector for alpha emitting isotopes and a Eberline sealed gas proportional detector for beta/gamma emitting isotopes. A multichannel analyzer is integrated with the diffused junction detector for discrimination of the alpha isotopes. This design ensures that all the isotopes of interest will be identified.

The combination of the two detectors will ensure that the DOE specified types of radioactivity will be detected. The specific alpha emitting isotopes U-238, U-235, Pu-239, Pu-242, Am-241 and Th-230 are able to be identified and quantified. Totally beta/gamma activity will be reported as Tc-99. Both Tc-99 and Cs-137 may be present as beta emitters, but Cs is not as important(toxic) as Tc. Although not done in Phase II, the gamma activity can be discriminated from the beta activity by simple mechanical shuttling of metal plate discriminators. A somewhat more complex, variable thickness shutter may be used to provide beta energy level discrimination.

3.2.1 Alpha Detection

The starting point for the alpha detection was Eberline's existing commercial product, the Alpha-6. This device is typically used in air monitoring application for discrimination of low levels of air borne alpha isotopes. The alpha isotopes are discriminated on the basis of pulse height using a multichannel analyzer determined from the penetration depth ('track') of the particle through the silicon substrate. The data are recorded as a function of energy in the multichannel analyzer. The discrimination is based upon "regions of interest" which are collection of channels summed together to give a response for a particular isotope. A 1 inch detector was integrated into the MSP. The associated electronics were integrated into the control software for the sensors.

3.2.2 Beta/Gamma Detection

The starting point for the beta/gamma technology was Eberline's Gas Proportional detector and modular detector board used in various commercial products. The gas (argon-carbon dioxide mixture) undergoes ionization by incident radiation, a charge is collected on the anode which is capacitively coupled to a comparator. Thresholds in the comparator discriminate noise and high energy alpha emission from the beta/gamma response. If gamma radiation is expected a 1/16" plate can be placed in front of the tube to discriminate it from the beta. A 2 inch diameter gas proportional tube was integrated into the Multisensor probe. The associated electronics were integrated into the control software for the sensors.

3.2.3 Experimental Results

Shown in Figure 3-10 is an example of a detector sensitivity testing. The sample is thorium 230 embedded into concrete at Bq/cm2 level (the regulatory level). The data show that within 60 seconds on a medium rough concrete surface sufficient data can be collected to identify the thorium 230. In an-

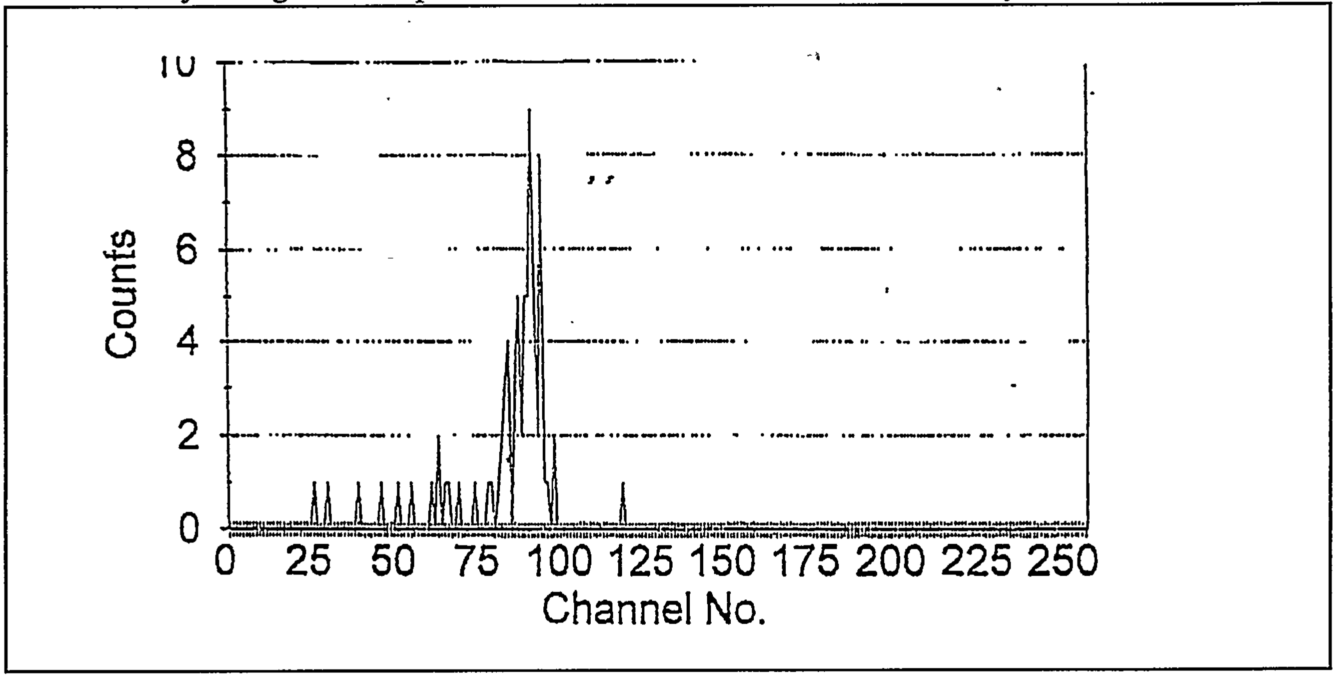


Figure 3-10. Example of Data Measured from 1 Bq/cm^2 of Thorium-230 Embedded on Concrete Surface

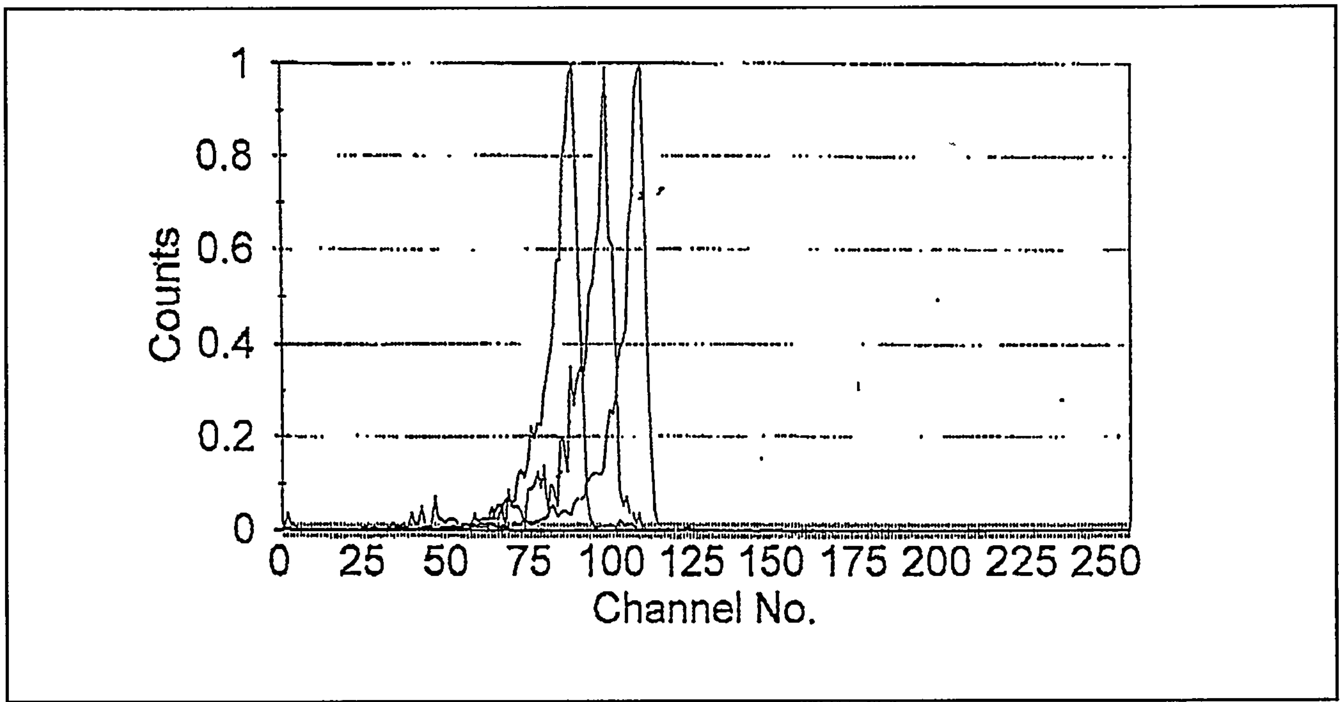


Figure 3-11. Example of Resolution for Discriminating Thorium, Plutonium and Americium using Standard Samples

other test standard samples of thorium, plutonium and americium were analyzed. Shown in Figure 3-11, is the data from this experiment. It is clear that sufficient resolution exists to differentiate these isotopes. No experiments were conducted on mixtures of these isotopes on concrete surfaces due to the difficulty of obtaining long term storage for the samples once the testing was completed. Based on the minor changes in the thorium standard and concrete samples it is expected that this sensor system will be capable of discriminating mixture of these isotopes on concrete.

Shown in Figure 3-12 is an example of the alpha spectrum recorded during a simultaneous measurement of GC/MS and radionuclide data. For this analytical run the sample was a mixture of PCBs and Thorium - 230 alpha standard. The corresponding GC/MS data is shown in Figures 3-6, 3-7 and 3-8 and the results for the PCB sample are described in Section 3-1. The peak of the alpha spectrum occurs at the energy level of 4.7 MeV indicating it is thorium 230. The alpha source is a sample of thorium embedded on a nickel substrate. These source are typically used as check standards through the radiation health industries. The source was 0.5 mm from the detector and the measurement time was 30 seconds. The data presented in Figures 3-6, 3-7, 3-8 and 3-12 indicate that the sensors system can make simultaneous measurements in near-real time.

The sensitivity of the gas proportional tube for beta and gamma detection was determined using 5 different isotopes of widely varying activity (dpm). The data shown in Table 3-1 indicates an acceptable level of sensitivity for several beta emitting isotopes as measured in the background activity of the Eberline facility in New Mexico.

The final column of data is the 10 second minimum detected activity (MDA) at the 95% confidence level for the different isotopes. This data indicate that the gas proportional tube is an effective detector for beta activity from surface as measured in air for short 10 second analysis times.

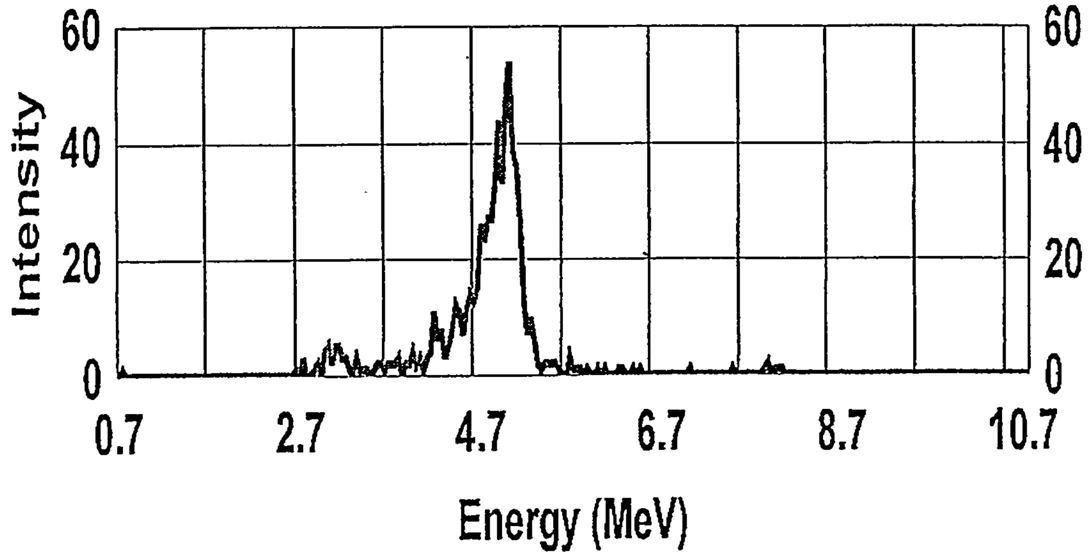


Figure 3-12. Example of Thorium-230 Measured Simultaneously with the HSGC-MS Data

There results indicate that the radionuclide detectors have the necessary sensitivity and discrimination ability to detect the radionuclide of DOE interest.

Table 3-1. Indicates an Acceptable Level of Sensitivity for Several Beta Emitting Isotopes

Isotope	Activity (dpm)	Half Life (Yrs.)	Current Activity	Gross Count Rate
SrY-90	6790	28.6	5131	1080
Ba-133	1410000	10.5	1311427	47300
	0		7	
Tc-99	16320	213000	16320	4800
Pm-147	19296	2.6234	8435	977
C-14	4099.2	5730	4097	328

3.3 The Molecular Vibrational (MVS) Sensor

The sensor for identification of base materials of interest to DOE (concrete, transite, asbestos, wood, and other organics) is a combination of an IR reflection sensor and a Raman scattering sensor.

During Phase II it has been demonstrated that the MVS probe should be based primarily on an extended near-infrared (ENIR) spectrometer equipped with a heavy metal fluoride (HMF) fiber-optic probe and secondarily on a 785-nm Raman spectrometer equipped with a silica fiber-optic probe. High quality spectra have been obtained from most materials investigated within 1 minute for both probes.

In an extensive test of a number of Raman spectrometers and laser wavelengths, it was shown that excitation at a wavelength of 785 nm gave the optimal combination of fluorescence rejection, minimal sample heating, portability and low data acquisition times to achieve a given signal-to-noise ratio (SNR). The Raman probe arrangement has been modified from the type studied in Phase I[1], in which the probe-head investigated incorporated 6 collection fibers around one excitation fiber (the so-called 6-around-1 arrangement) with no optical filtering. The optimum probe configuration was found to be a single input fiber with a notch filter mounted at the end so that no Raman scattered radiation from the silica fiber reached the sample. The radiation scattered from the sample is then passed through a second optical filter to remove the Rayleigh-scattered light (at 785 nm) and pass all the longer-wavelength (Raman scattered) radiation which is then focused into a single output fiber. The second filter prevents the Rayleigh-scattered radiation from giving rise to Raman scattering in the output fiber. A commercial spectrometer and probe head manufactured by Kaiser Optical Systems, Inc. were identified as being optimal for the measurement of the Raman spectra of solid samples with a low Raman cross-section (such as asbestos). This instrument is now in operation at the UI and will be incorporated into the Phase III multisensor probe.

Two types of software for classification and/or identification of the samples have been studied. In the first, commonly known as spectral searching, the measured spectrum is compared directly to a library of reference spectra and the absolute difference between these spectra is calculated and summed over all wavelengths. The reference spectrum yielding the smallest sum is the best match to the spectrum of the unknown. This approach proves to be most useful when the spectrum of the unknown contains several narrow bands that are easily distinguished from the spectral baseline. In this case, an automated baseline correction routine that has been developed at UI can be applied prior to the application of the spectral searching program. For many of the samples, however, the spectra are not of the ideal form for spectral searching. For such samples, other approaches must be applied. Two such techniques were investigated: a relatively well-known algorithm known as principal components analysis (PCA), and a new form of spectral classification involving the use of self-organized mapping (SOM) neural networks. The latter approach has proved to be more robust than either spectral searching or PCA and will form the basis of the classification software in the final field version of the MVS probe.

The results obtained with the silica fiber-optic probe analysis of the materials of DOE interest indicated that NIR diffuse reflectance (DR) spectrometry with a silica fiber-optic probe can be used to analyze all materials except those that totally absorb NIR radiation. It was also shown that NIR DR spectrometry could be used to distinguish between the five major types of asbestos. However, the spectral features that are of importance for distinguishing asbestos from analogous samples such as bricks and concretes are so weak that they can be lost in the baseline noise if the corresponding measurements are made using detectors operating at temperatures appropriate for field measurements. For such samples, access to the much stronger fundamental C-H and O-H stretching modes would be highly beneficial. For this reason, we investigated the feasibility of extending the wavelength range of fiber-optic DR measurements down to about 2500 cm^{-1}.

Experimentally, we have shown that, for samples for which ENIR DR spectra do not contain enough useful information to permit rapid identification, Raman spectrometry often does.

Thus ENIR DR and Raman spectrometry often prove to be highly complementary techniques.

3.3.1 Extended Near Infrared Spectrometry

An experimental fiber-optic probe for DR measurements using heavy metal fluoride fibers and incorporating a thermoelectrically-cooled mercury cadmium telluride detector was purchased from Galileo Electro-Optics Corp. and interfaced to a ATI/Mattson Genesis Fourier transform spectrometer configured to cover the spectral region between 1 and 4 μm. The combination of this spectrometer and the heavy metal fiber DR probe enabled high quality spectra to be measured when the probe head was in direct contact with the sample. Typical spectra measured in a time of 45 seconds with this instrument are shown in Figure 3-13.

For a fieldable ENIR probe, direct surface contact is undesirable because of possible probe contamination. Therefore a second probe design was tested which employed a lens to focus on surfaces spaced a 3mm distance. With this design there is a small change in relative band intensities in the observed DR spectra. This effect can be accommodated by including appropriate reference spectra in the database for spectral identification or through a software correction. The approach for this compensation will be determined in the remainder of Phase II.

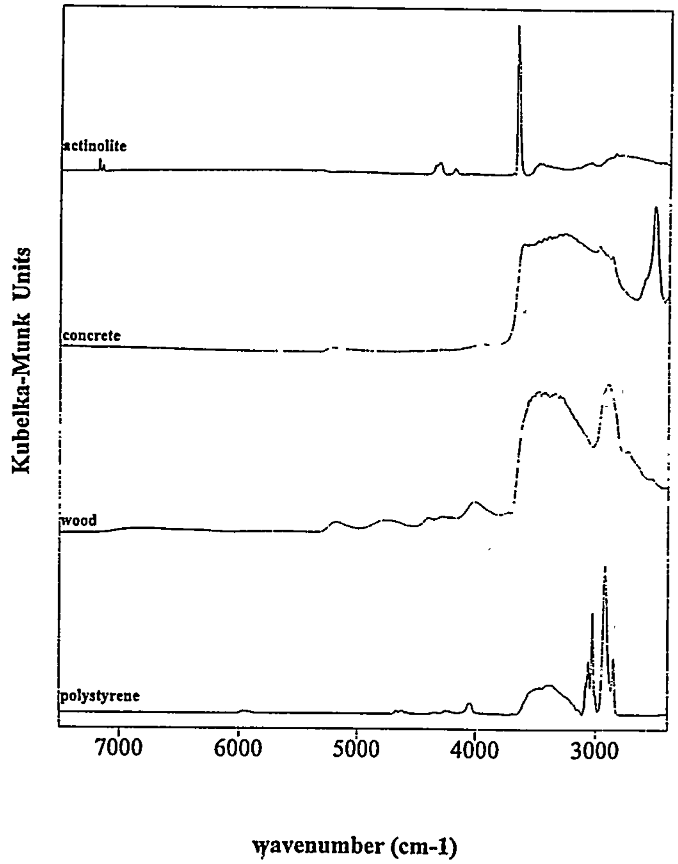


Figure 3-13. ENIR DR Spectra of Actinolite, Concrete, Wood and Polystyrene Foam Measured in the Contact Mode

3.3.2 Raman Spectrometry

Most CCD-Raman spectra were acquired on a Renishaw Raman spectrometer utilizing a microscope as the sampling device. The 785-nm diode laser provided a maximum power of 2 mW at the sample. Excitation with gas lasers emitting at 632.8 and 514.5 nm was also tested but fluorescence overwhelmed the Raman spectrum features for most materials of interest. Different lenses allowed standoff distances between 1 and 10 mm to be tested. Single- and multiple-scan accumulation spectra were collected at approximately 6-cm^{-1} resolution using CCD detector charge integration times from 1 to 120 seconds. Band positions are accurate to within 2 cm^{-1}. The results obtained with this spectrometer without the fiber-optic probe were sufficiently encouraging that we investigated other commercially available Raman spectrometers to which fiber-optic probes had been interfaced. Of these instruments, the one yielding spectra of solid samples with by far the highest SNR was the Kaiser Optical Holoprobe Raman spectrometer.

The Kaiser Holoprobe is a state-of-the art NIR Raman spectrometer optimized for use with 785-nm excitation. The system comprises a 50-mW external-cavity-stabilized diode laser providing a maximum laser power at the sample of 20 mW, a transmission grating, a CCD detector, and a state-of-the-art fiber-optic probe packaged in a smaller volume than the Renishaw spectrometer. This system offered a useable Raman shift wavenumber range of 50 - 3500 cm-1 with 785-nm excitation.

3.3.3 Tests for the Appropriate Excitation Wavelength

When the 632.8-nm and the 514.5-nm laser lines were used for excitation, the spectra are so severely affected by molecular and/or atomic luminescence that no Raman bands can be observed above the background. Thus the wavelength for the excitation laser for the MVS Raman probe had to be in the near infrared; in practice the only two wavelengths between which a choice had to be made were 1064 and 785 nm (see Table 3-2). The 785 nm excitation was selected because of the greater strength of the Raman effect and shorter sample time required at that wavelength.

Table 3-2. Wavelength for the Excitation Laser for the MVS Raman

Material	**Intersample Variability**				**Signal-to-Noise Ratio (degraded by fluorescence and sample heating)**			
	514.5	632.8	785	1064	514.5	632.8	785	1064
Concrete	-	1	4	1	-	VP	M	VP
Bricks	-	1	1	3	-	VP	VG	G
Asbestos	1	1	3	3	VP	VP	G	M-P
Transite	-	1	1	3	-	P	P	M-P
Porcelain	-	-	1	2	-	-	VG	G
Woods	1	1	1	1	VP	P	P	VG
Asphalt	-	1	1	1	-	VP	VP	VP
Polymers	4	3	2	2	G-M	G	G	G
Organics	-	3	2	1	-	M	G	G

3.3.4 Probe-head Arrangements

The fiber-optic probe design for Raman spectrometry has changed significantly from the 6-around-1 arrangement tested in Phase I to the filtered probe-head now used.

The filtered probe-head arrangement prevents any Raman scattering from the fiber-optic cables from obscuring the Raman signal from the sample. The Kaiser Optical System[3] Raman probe, has been identified as optimal for the 3D-ICAS application based on the signal throughput of the fiber-probe, the low Raman cross-sections of some of the samples of interest in this project (especially, asbestos and concrete), the wavenumber range ($\Delta\nu$ = 50-1000 cm^{-1}) needed to identify DOE materials of interest.

A recent study sponsored by the DOE at Westinghouse-Hanford[4] has shown for non-reflective non-fluorescent samples that a 6-around-1 probe-head with a flat-tip termination can yield up to an order of magnitude more signal than the filtered probe-head arrangement. It should be noted, however, that the filtered probe that was used for this study had only a 10% throughput. The probe head selected by UI for the 3D-ICAS has a throughput of approximately 50%; thus, all else being equal, there is only factor of two difference between using the Kaiser probe and the Westinghouse tested design.

The reduced silica fiber scattering background for highly reflective materials more than compensates for this small reduction in throughput efficiency.

It has been shown both at the UI and in other DOE funded efforts[5,6] that if the background radiation caused by the Raman spectrum of silica in the input fiber is not filtered out the Raman signal from the sample can be significantly degraded or completely obscured. In Figure 3-14, the Raman spectrum of balsa wood measured through a 2-meter unfiltered 6-around-1 probe-head is presented. A comparison of the resultant balsa spectrum (Figure 3-14c) produced by subtraction of the silica background from the original probe-recorded spectrum of balsa with the balsa spectrum recorded in the macro-chamber of the spectrometer (Figure 3-14d) shows that the SNR has been significantly degraded when unfiltered fiber-optic probe-heads are used. This is especially obvious in the wavenumber range below 1000 cm-1 where spurious intensities for vibrational bands are observed between 200 and 1000 cm^{-1}. (It should be noted that Balsa wood is not a particularly weak Raman scatterer.)

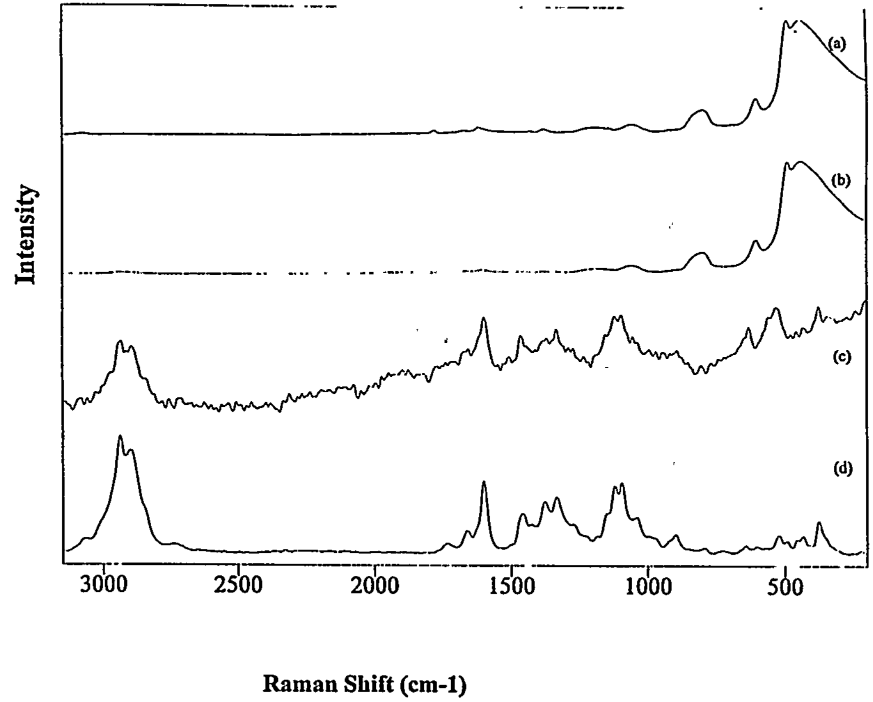
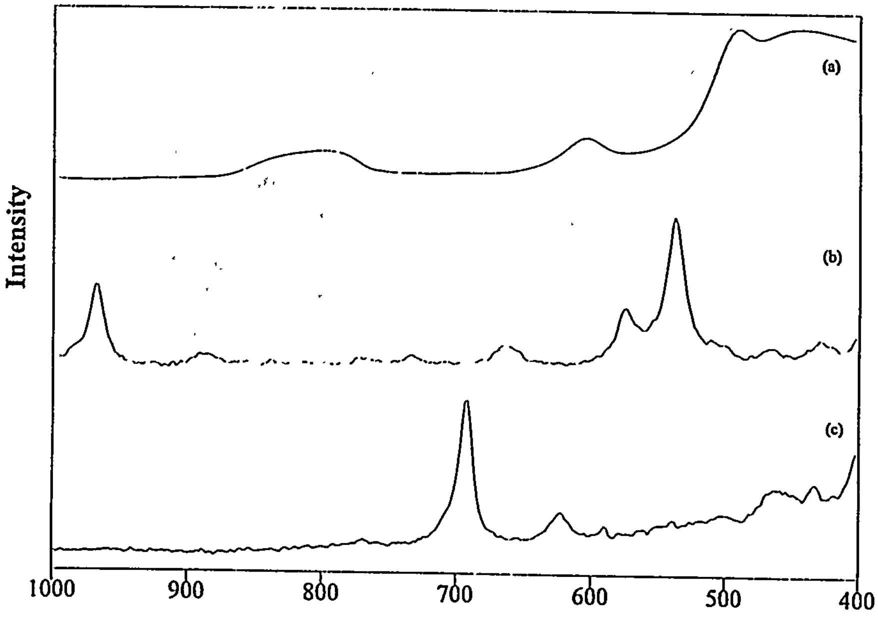


Figure 3-14. Typical Raman Spectroscopic Performance of an Unfiltered Focused Fiber-Optic Probe

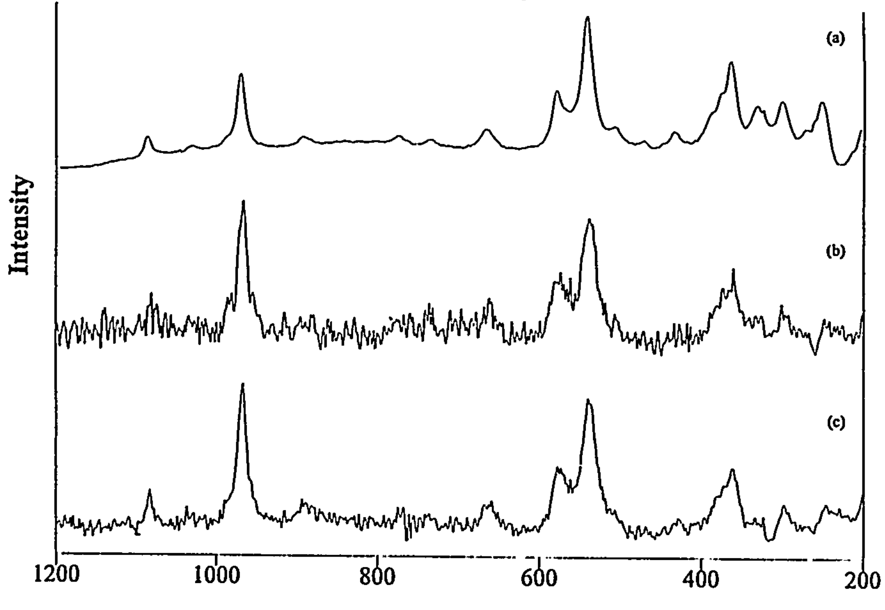
In Figure 3-15 the Raman spectra of two asbestos minerals, chrysotile and crocidolite, are presented as well as the Raman silica background from the 2 meter 6-around-1 Raman probe. It can be seen that the silica background will obscure the strong vibrational bands of crocidolite (Figure 3-15b) and that the chrysotile bands may also be lost in the background. The asbestos minerals are very weak Raman scatterers (at least one order of magnitude weaker than the sample of balsa wood shown in Figure 3-14). Thus even though spectra measured with the 6-around-1 fiber-probe have not been obtained for the asbestos minerals it appears clear that the noise associated with silica background which is left in the resultant spectrum after background subtraction will totally obscure any asbestos signal from being observed.



Raman Shift (cm-1)

Figure 3-15. Raman Spectra in the Wavenumber Range

In contrast, the filtered probe-head design of Kaiser has been used to obtain Raman spectra of crocidolite within two minutes using only 5 mW of laser power; the signal level of these spectra is sufficient to allow the mineral to be identified, as shown in Figures 3-16b and 3-16c. All the intense features



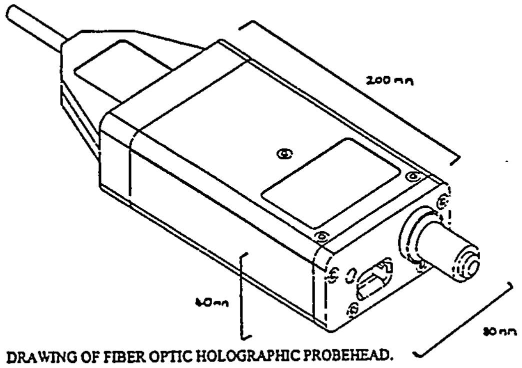
Raman Shift (cm-1)

Figure 3-16. Raman Spectra of the Asbestos Mineral Crocidolite Recorded

in the reference spectrum (Figure 3-16, measured in about an hour) can be seen with a SNR of greater than 3 in spectra measured in 2 minutes with the Kaiser fiber-optic probe.

The schematic of the Kaiser holographic probe (Figure 3-17) shows that the probe-head comprises (i) a laser bandpass filter incorporated in the probe-head case after the radiation has emerged form the illumination fiber and (ii) holographic Rayleigh notch filters which prevent reflected laser radiation from entering the collection fiber. These two components satisfy the silica background rejection requirement demonstrated above. A third important component is an emission indicator built into the probe head. This indicator should allow an untrained operator of the Raman spectroscopic probe to verify if the laser operation on a regular basis without the need for expensive test equipment or extensive sensor down-time.

In conclusion since a number of materials of interest in this project (most importantly asbestos) possess crucial vibrational information in the region 800-50 cm-1, the chisel-tip probe approach pioneered at Dow Chemicals9 and the flat-tip approach5 proved to be unsuitable for the DOE's specific needs. They have, therefore, not been adopted because both methods fail to meet the sample-dictated criteria necessary for routine analysis. The results of Phase II have shown that the Kaiser Holoprobe Raman fiber-optic probe is the best available design for studies of weakly Raman scattering solid materials, especially when the wavenumber range below 800 cm-1 is important (as it is for asbestos).



DRAWING OF FIBER OPTIC HOLOGRAPHIC PROBEHEAD.

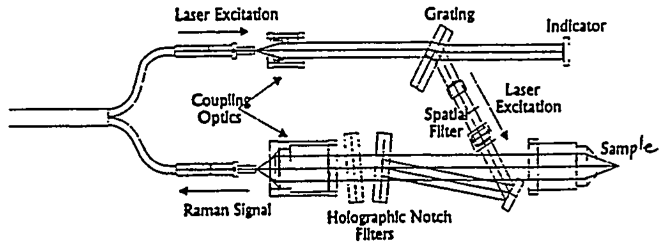


Figure 3-17. Diagrammatic Representation of the Kaiser Holographic Probe-Head

3.3.5 Software for Identification and Classification

Two types of software for classification and/or identification of the samples have been studied, spectral searching and self-organized mapping (SOM) neural networks. In spectral searching, the measured spectrum is compared directly to a library of reference spectra; the absolute difference between the spectrum of the unknown and each reference spectrum is calculated at each wavelength and summed over all wavelengths. The reference spectrum yielding the smallest sum is the best match to the spectrum of the unknown. Spectral searching proves to be most useful when the spectrum of the unknown contains several narrow bands that are easily distinguished from the spectral baseline. In this case, an automated baseline correction routine that has been developed at UI can be applied prior to the application of the spectral searching program.

Unfortunately, the spectra of many of the samples encountered in this project are not of the ideal form for spectral searching. For such samples two other techniques were investigated: principal components analysis (PCA), and self-organized mapping (SOM) neural networks. For both techniques, the dimensionality of the input data is reduced from a fairly large number (the number of data points in each spectrum, often several hundred) to a much smaller number (usually two). In each case, a two-dimensional plot is produced in which it is hoped that samples of a given type will form a cluster in a given region of the plot. In PCA, for example, the ordinates of the plot are the scores of the first two principal components (PC1 and PC2). When the ENIR spectra of a number of samples of brick, concrete, asbestos and polymers are subjected to the PCA, quite poor clustering results (Figure 3-18). With the use of SOM neural networks, on the other hand, most samples of a given type fell within well-defined regions of the map (Figure 3-19). Neural computing with self-organizing mapping has proved to be more robust than either spectral searching or PCA and will form the basis of the classification software in the final field version of the MVS probe, using both Raman and ENIR reflectance data.

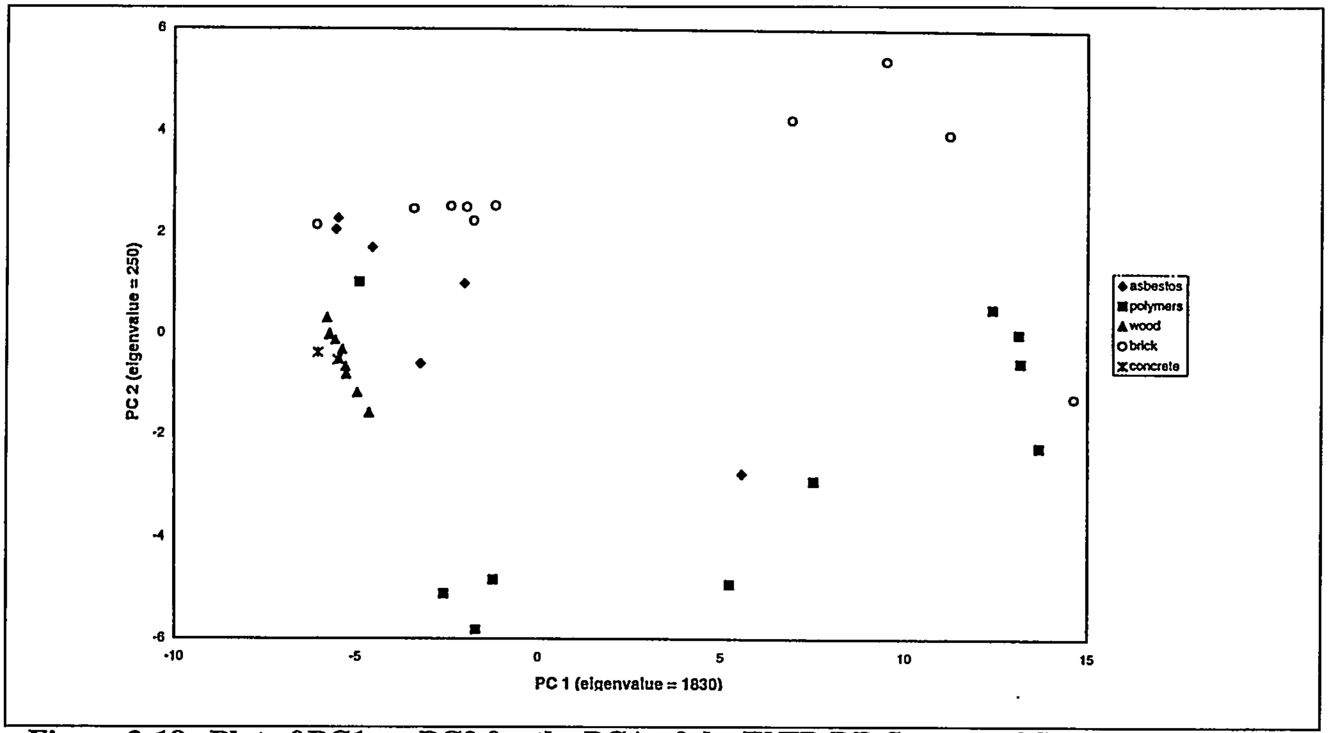


Figure 3-18. Plot of PC1 vs. PC2 for the PCA of the ENIR DR Spectra of Several Samples of Asbestos, Polymers, Wood, Brick and Concrete

3.4 3D-ICAS System Integration

The 3D-ICAS system uses its coherent laser radar (CLR) to obtain 3D facility maps, then uses

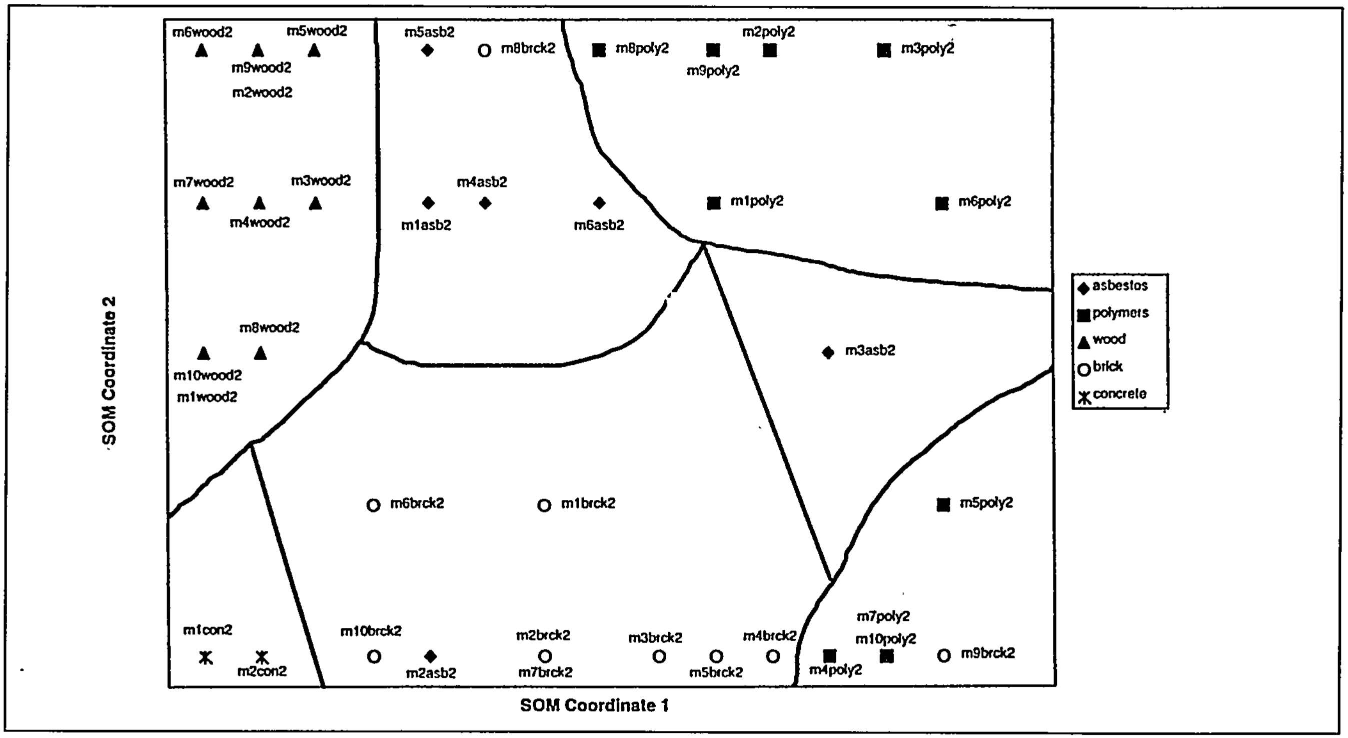


Figure 3-19. Plot of Coordinate 1 vs. Coordinate 2 for a SOM

the CLR to guide a robot arm borne multisensor probe along surfaces for contamination detection, 3D mapping and archiving. A substantial portion of the development effort has been devoted to obtaining the integrated functionality of these subsystems. We will describe some of these subsystems and their interaction briefly. We will then provide performance examples of the 3D-ICAS "eyes" the CLR 3D Mapper.

3.4.1 Robot Arm

Robot arm control and manipulation is an important part of the Phase II work. Integration of control of the robot arm shows advancement towards a fieldable system. During Phase II a model 465A robot arm from CRS Robotics (CRS) was selected, programmed and used to manipulate the sensor probe in a limited area.

The CRS 465A robot arm has about the same reach as a human's arm with more flexibility. The arm is not operated at full speed. The CRS C500 controller is programmed in the Robot Applied Programming Language, RAPL II, a language developed by CRS which is a derivative of BASIC and C. In Phase III the robot arm will be attached to a mobile platform allowing for full working space access.

3.4.2 Multisensor Probe

The Phase II multisensor probe comprises the GC sensor head, two radionuclide sensors, one for alpha particles and one for beta/gamma emissions, and four proximity sensors. Three tetrahedron tracking targets are mounted on the outside of the probe. The probe is cube 6x6x6 inches and weighs 6 lbs.

The Phase III probe will be essentially the same with three modifications. First, several tracking targets will be added to provide complete tracking capability. Second, the MVS sensor will be integrated with the other sensors on the probe. Third, proximity sensors will be selected and mounted in such a way that curved surfaces can be handled.

3.4.3 Proximity Sensor

The Phase II system used commercial proximity sensors mounted at the four corners of the face of the probe. These proximity sensors provide a capability to move the probe up to a flat surface and maintain a standoff distance of 1 mm or more.

3.4.4 Robot Arm Platform

The Phase II system was not required to provide mobility for either the mapper or the robot arm. Portability was provided by mounting each unit on a wheeled tripod and moving them manually. For the Phase III demonstration the 3D-ICAS will take advantage of DOE developed mobility platforms for both the 3D mapper and the sensor robot arm.

3.4.5 Control Strategy

The multisensor probe moves to each point specified in the survey list in the order in which they appear in the list generated by the integrated workstation. Movement between points is along a standard sequence of steps. First, the multisensor probe moves from the rest position along a straight line to a position near the next survey point, rotating along the way so that the face of the probe is nearly parallel to the surface at the end of the segment. Second, the probe moves toward the surface until the proximity sensors detect the surface. The final approach is accomplished in small steps under control of the proximity sensors. The sensor system is commanded to acquire the contaminant sample and initiate processing. Then the probe moves along a straight line to the arm's rest position.

When the probe has settled at each point, the CLR tracker is activated and a scan of the tracking target is made. The scan data are processed to provide an accurate estimate of the probe's location and orientation.

The Phase II probe control strategy moves the probe up the desired spot and all contaminant sensing is done with the probe in one place. The surface being sampled was assumed in Phase II to be flat over the entire face of the probe so that moving the probe up to the surface with the face of the probe parallel to the surface was an acceptable approach strategy. In Phase III a more robust approach strategy will be implemented. First, one will be able to move the probe up to a single spot, or move each of the four sensor heads in sequence to the same spot. This will allow the probe to make contaminant measurements at the same spot. It also permits the system to survey curved surfaces such as pipes. Finally, with the sampling sequenced properly, i.e. with the MVS and RN samples taken first, the GC sampling can be suppressed if the MVS indicates that organic contaminant levels are so high that the GC sensor could be contaminated. In this case the high sensitivity of the GC-MS is not needed.

3.4.6 End Effector Tracking Function

CRC has developed an efficient approach for the CLR for tracking location and orientation of a robot end effector. Analysis, simulation, and laboratory experiments demonstrate the effectiveness of this technique. The tracking system has been integrated with the 3D-ICAS Phase II system demonstration. The tracker provides 6-DOF (X, Y, Z location and Yaw, Pitch, Roll orientation) tracking using a symmetric tetrahedron target.

3.4.7 Tracking Targets

The tracking target is a symmetric tetrahedron consisting of an equilateral triangle base and three identical faces: Figure 3-20. Reflective paint is applied to the entire target except along the edges between faces.

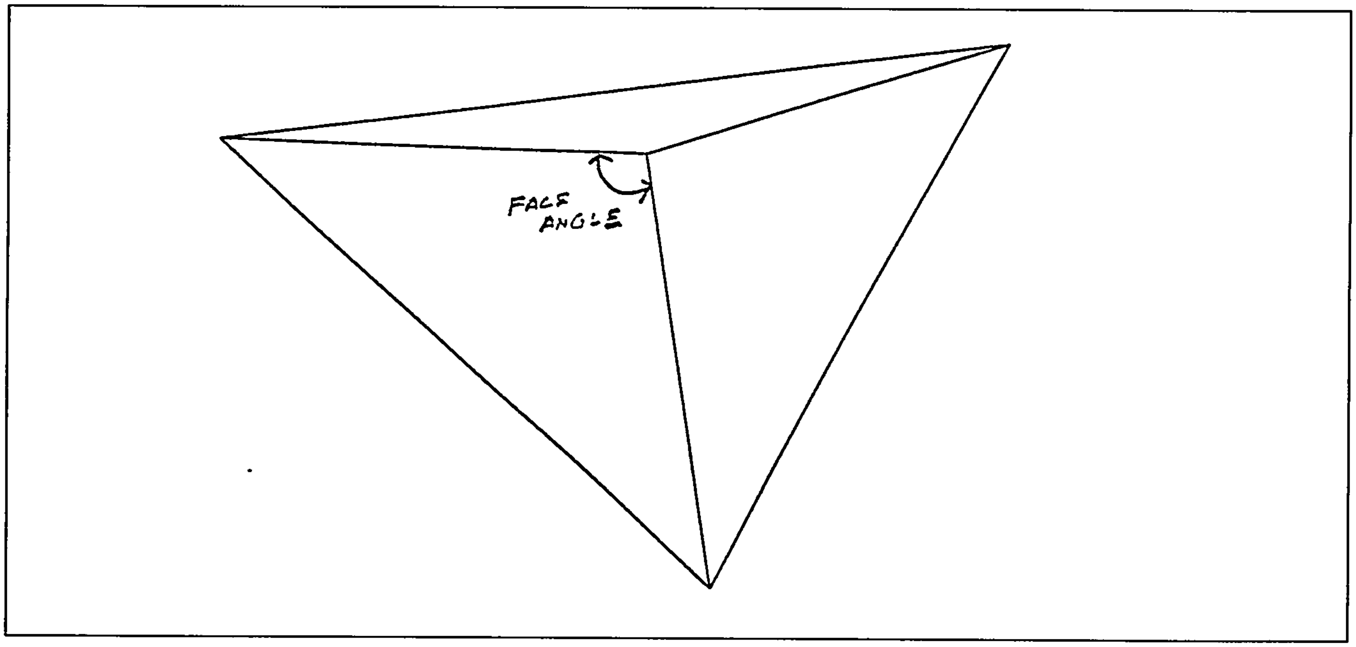


Figure 3-20. Tetrahedon Tracking Target

Target size was constrained by the size of the probe and the face angle was selected considering location accuracy, orientation accuracy, and the number of targets required for full visibility. Targets with 110 degree face angle were specified for the Phase II demonstration.

3.4.8 Position Estimation Algorithm

The technique consists of scanning the CLR beam in a circular pattern around the apex of the target. The center and radius of the scan are preset so that the scan encircles the apex and remains on the target for the entire scan. Since the approximate location and orientation of the target are known, the desired scan pattern is easily determined. The CLR returns range, azimuth, elevation, and signal quality for each measurement point during the scan.

The first step of the position estimation algorithm is to estimate the three planes that best fit the three data sets independently. The intersection of the three planes determines the location of the apex of the target. The vectors normal to each plane are immediately available. These vectors provide the target's orientation. The second step is to fit the independent normal vectors by a set of normal vectors constrained in such a way that they correspond to a tetrahedron target with the specified face angle. The location of the apex is recomputed after the best fit orientation is determined. This estimate of location and orientation will be used as the starting point for a steepest descent optimization in Phase III.

3.4.9 Target Tracking Results

Two targets were fabricated by a commercial machine shop. Target 1 was specified with a 90 degree face angle and target 2 with a 110 degree face angle. Tests of the targets showed that the angles between the faces ranged from 88.60 degrees to 91.31 degrees whereas they should have been 90.00 degrees. Corresponding angles for target 2 were much more consistent, showing a spread of 0.03 degrees, but they were over a degree different from the design value. Each target was evaluated by taking ten concentric scans with different radii, estimating the location of the apex and orientation, and computing the root-sum-squared standard deviations. Table 3-3 summarizes the results.

Table 3-3. Root-sum-squared Standard Deviation (rsssd) from Ten Scans

	Target 1	Target 2
Design face angle	90 degrees	110 degrees
Apex estimate rsssd	0.97mm	0.63mm
Direction angle Estimate rsssd	1.45 degrees	1.0 degrees

The results indicate that the flatter target gives better results and estimate accuracy on the order of 0.6 millimeters and 1.0 degrees should be possible.

3.4.10 Database Function

The database capability developed for the 3D-ICAS system serves as a repository for all of the data collected by the system. As the data are gathered, the system automatically sorts, keys and archives it for easy retrieval and display by the Integrated Workstation (IWOS). The data can also be made available for external clients as a server in an overall facility contamination management system.

Data are recorded for each survey point. The location of the sensor head is recorded precisely in facility coordinates along with the entire data structure from all of the sensors. The data structure includes raw data from the radionuclide (RN) and High-Speed Gas Chromatograph/Mass Spectrograph (HSGC/MS) sensors. Data from the Molecular Vibrational Spectrometer (MVS) will be incorporated during Phase III in accordance with the three phase development schedule. The data structure also includes processed assignments for four organic compounds provided by the HSGC/MS. Finally the database holds any specific images generated from the raw 3D mapper data.

International Business Machine's (IBM) industrial database DB2 was selected for use in the 3D-ICAS system after surveying the leading large-scale commercial database vendors.

A primary consideration in the selection was the ability to directly support structured query language (SQL) in multi-platforms. SQL is used in the 3D-ICAS access and storage of the data. This allows future expansion of the system to provide the database information to other users in a networked environment. DB2 also runs on a wide variety of computers including workstations, PCS, and mainframes.

In 3D-ICAS DB2 will be run on the laser radar control computer. Using the preemptive multitasking capabilities of OS/2, the database will run in a separate session to ensure data integrity and responsiveness. This configuration will be carried over to Phase III.

3.4.11 Data Archiving and Retrieval

The database is setup as a set of tables to store the following information (see Table 3-4):

Queries can be made on all of the fields. The defined primary key fields used for sorting are the Facility Identification and Date, although any data field may be selected and sorted.

Raw data from the Molecular Vibrational Spectrometer will be integrated into the database function during Phase III.

Table 3-4. Archival Data

Identification
- Facility Identification (Agency, Plant, Building, Room)
- Date, time

Mapping
- Location of fiducial marks in room coordinate frame
- 3D mapper data in room coordinates
- Any generated facility scenes
- Bounding box of coordinates for each 3D mapping run

HSGC/MS
- 3D coordinates of each sample point (in room coordinates)
- Gas chromatogram for each point
- Mass spectrogram for each peak in each gas chromatogram
- Mass of each of five compounds at each point (PCB, fuel, volatile organics, explosives, other)

RN
- 3D coordinates of each sample point (in room coordinates)
- Alpha spectrum
- Beta-Gamma count
- Activity level of each of six isotopes (U235, U238, Pu239, Pu242, Am241, Th230)

3.4.12 Data Display: Facility Scene

A facility scene image is generated from the 3D facility frame mapper data by selecting the area to display and an optical viewpoint and converting range from the viewpoint into a rendered image. The facility scene is rendered in black and white so that subsequent contamination overlays in color will be highlighted.

3.4.13 Data Display: Contaminants

When a facility scene is displayed on the IWOS, the database can be queried for related survey information. The data can be overlaid on the facility scene as scatter contour plots or displayed in one of several different graphical or list windows.

Contaminant concentration levels are presented as thresholded scatter diagrams rather than continuous line contour plots because the interpolation ingerent in constructing continuous contours could produce an erroneous picture of the contamination, especially when the survey points are widely separated. Each contaminant is displayed in its own window. The window contains the gray scale facility scene overlayed with contamination levels in color. An asterisk is placed at each location where a measurement was made. The color of the asterisk depends on the concentration of the contaminant. Green, yellow, red, and blue indicate no, light, medium, and heavy contamination respectively. Default thresholds for defining the four contamination levels are provided; the user may change the thresholds.

More detailed displays of contamination data are keyed from the facility scenes with contamination overlays on them. For any contamination display, the operator can select a location to get detailed information about with the mouse. Radionuclide activity levels are displayed in tabular form. Gas chromatograms are displayed in graphical form. Masses of the five organic compounds are displayed as a bar graph. Secondary displays consisting of mass spectrograms are obtained by mousing on desired peaks in the gas chromatogram; mass spectrograms are displayed as graphs and also in tabular form. An example of these displays is shown in Figure 3-21. Displays of data from the Molecular Vibrational Spectrometer will be integrated into the display function during Phase III.

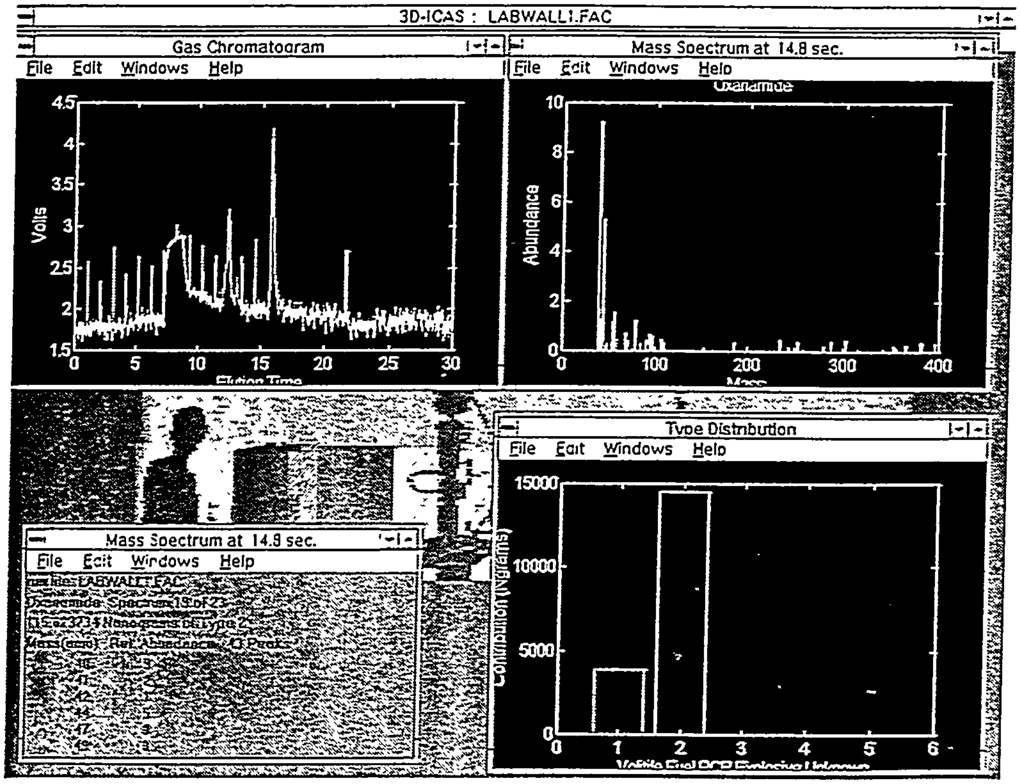


Figure 3-21. Facility Scene Contaminant Data Display

3.5 The CLR 3D Mapper

The CLR 3D Mapper, which serves as the robotic eyes for 3D-ICAS, has continued to be refined with other program support during the 3D-ICAS Phase II development effort. A CLR 3D Mapper to be dedicated to 3D-ICAS is being completed during Phase II. The accuracy of the CLR 3D Mapper is critical to the development of reliable facility maps and for accurate robotic sensor end effector tracking and guidance. An example of the mapping accuracy of the CLR 3D Mapper is shown in Figure 3-22, which shows a 3D image of a dime taken at four meters range.

4.0 Application and Benefits

The completed 3D-ICAS will be directly applicable as a system to DOE facility characterization decontamination and decommissioning. The sensor subsystems of 3D-ICAS represent an advance in portable real-time chemical analysis for chemical constituents and dangerous materials of interest to DOE. The CLR 3D mapping subsystem, developed outside of the 3D-ICAS program, can perform as a critical subsystem on a wide range of DOE robotic applications and I industrial metrology applications.

Benefits of 3D-ICAS High Speed Contaminant/Base Material Analysis for Overall Decontamination and Decommissioning Operations

3D-ICAS site chemical analysis is expected to improve the technological capabilities of current chemical analysis methods by combining state-of-the-art sample preparation techniques, High Speed GC, time-of-flight mass spectrometry (TOF-MS), molecular vibrational spectrometry, and radionuclide

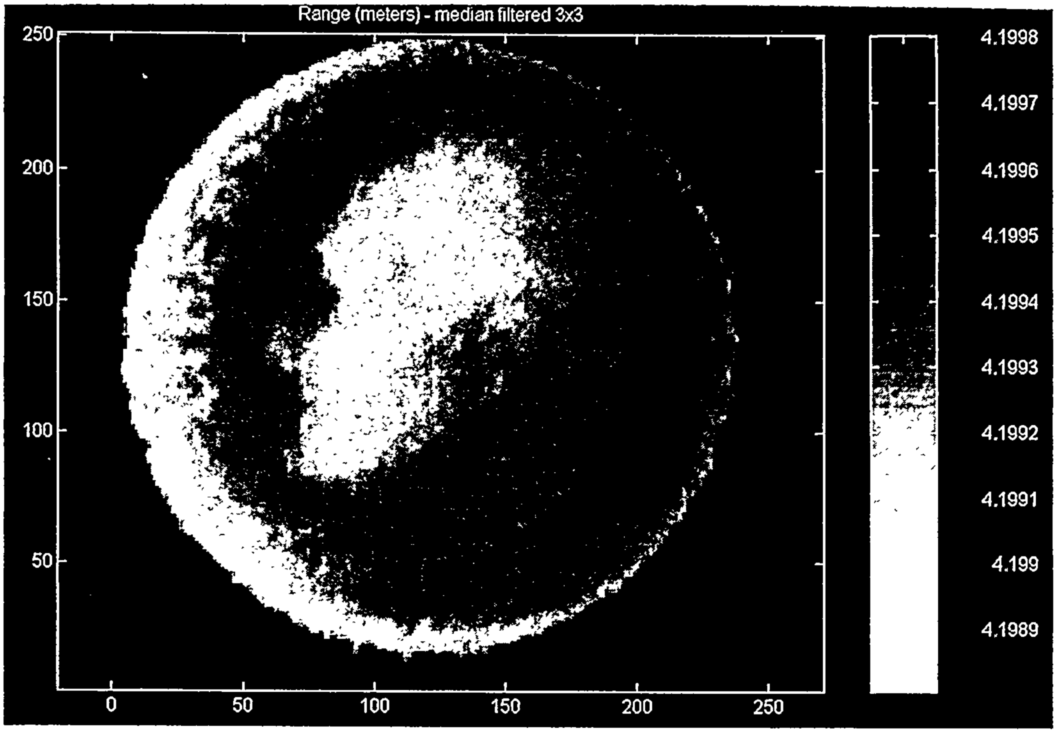


Figure 3-22. 3D Image of a Dime taken at Four Meters

analysis into a system that can be operated at a field site to provide real-time, reliable qualitative and quantitative results. The availability of this technology will positively impact DOE clean-up operations in the following ways:

- Improved Performance (faster, more accurate)
- Cost Reduction
- Reduction of Health Risks
- Reduction of Environmental Risk
- Improved Operations
- Reduced Time for Remediation
- Waste Minimization
- Supporting Overall Institutional Goals

Improved Performance: The quality of analytical data generated by 3D-ICAS chemical analysis instrumentation will equal or exceed that currently being generated by conventional laboratories. Sample automation and elimination of the extensive sample handling associated with off-site storage and analysis will improve the reliability of the data as well as significantly reduce sample preparation time. High speed GC techniques will produce chromatographic peaks more than 100 times sharper than conventional separations and improve the detector performance for a given mass of analyte. High speed GC will also reduce the separation time from 20-45 min for a typical run down to 100 seconds or less, resulting in faster analysis times. The Radionuclide analysis may not provide more sensitivity than conventional laboratory analysis, but the direct real-time field results will certainly produce the required data expeditiously. Molecular vibrational spectrometric analysis will identify inorganic and organic surface materials and moderate (down to low parts per million) levels of contaminations, at high spatial resolution, in sample times as short as five seconds.

Cost Reduction: The proposed technology will reduce the overall cost of clean-up operations. Costs for chemical analysis will be reduced because the whole process from sample preparation to data management can be automated and performed on-site. Operational cost benefits will result from the availability of real-time analysis. Manpower and equipment costs would be reduced through the use of the robot operated chemical instrumentation due to improved operational efficiency.

Reduction of Health Risks: The automation of sample preparation steps and actual site mapping would eliminate sample handling by laboratory personnel. Further, survey worker exposure to hazardous locations would be significantly reduced or eliminated. Insurance premiums alone will be reduced (it should be cheaper to insure robots than workers).

Reduction of Environmental Risks: On-site, real-time analysis with the robot operated analyzer would provide improved site monitoring during decontamination. Using teal-time analysis, engineers could monitor and respond to problems in less time than is possible with conventional chemical analysis.

Improved Operations: The robot operated analysis instrumentation would improve the capabilities of DOE response in each of the three phases of remedial efforts by providing a more efficient, streamlined beginning-to-end operational capability. During the survey state worker exposure is reduced; and mapping and characterization efforts proceed efficiently with the aid of real-time, remote controlled mapping and analysis. For example, when mapping out the contaminated area, engineers could review analytical results immediately and direct more intensive efforts where they are needed most. More accurate and detailed characterization of the waste site would concentrate restoration efforts to contaminated areas and eliminate needless efforts on areas that are in compliance or pose minimum risk. Once decontamination efforts are begun, removed materials and remaining site locations could be continually monitored to measure the progress of clean-up operations. Remediation operations would proceed faster because field engineers could have near real-time results indicating the status with respect to regulatory requirements.

Waste Minimization: Real-time monitoring of decontamination operations would have a direct impact on the amount of waste generated during remediation. Procedures could be halted as soon as compliance is achieved, preventing excess of removal of otherwise clean material. More detailed mapping and characterization of site would identify areas that are in compliance and do not require decontamination. The analyses would be able to classify materials as they are removed thus optimizing the handling requirements to be consistent with their hazard level.

CLR 3D Mapper Advantages over all Other Current Technologies

Immunity to Ambient Light and Surface Shading: The FM CLR is immune to ambient lighting conditions in an exactly analogous way that FM radio is immune to the background amplitude bursts in lightning. The other methods depend upon amplitude detection.

Lightweight Radiation Resistant Fiber Optic Implementation: The high sensitivity of the FM CLR detection process allows the use of a remote scanner at the end of an optical fiber with no solid state electronics. Because of the high efficiency of the optical fibers, the remote scanner could, in principle, be kilometers from the integrated workstation.

Benefits of the CLR 3D Mapper Performance Advantage: The fast CLR 3D Mapping capability will not impede or interrupt D&D operations. Immunity to lighting conditions means that other D&D operations will not have to be disrupted while lighting is controlled for 3D mapping operations. A more accurate 3D mapping capability allows robotic operations to proceed with less supervision for characterization, decontamination, and dismantling operations. Characterization benefits from the high accuracy CLR 3D mapping are:

- Allows clear visualization of surface and more accurate planning and execution of surface contaminant mapping operations
- Allows the sensor probe to follow a surface at close proximity without collision
- Supports object shape and texture determination and better contaminant penetration modeling

5.0 Future Developments

The Phase III 3D-ICAS development is aimed at demonstrating an fieldable contamination 3D mapping, analysis and archiving system to support D&D operations. Highlights of the 3D-ICAS system capability and performance improvements to be accomplished in Phase III are as follows:

- Integration of the multisensor probe and supporting robot arm 3D-ICAS sensor subsystem) with a DOD provided mobility platform
- Integration of the CLR 3D Mapper subsystem with a DOE provided mobility platform
- Reduction in size and weight and field hardening of sensor subsystems and analysis hardware
- Improvement in speed of the CLR guided 6DOF multisensor probe end effector tracking and control
- Increase in flexibility of sensor route planning to allow a variety of mapping coverages for each sensor component
- Refinement of contaminant detection and analysis software for all sensor components
- Refinement of system control, display, and other human interface software
- Adaptation of 3D-ICAS system mission to evolving DOE site cleanup requirements

Beyond the 3D-ICAS development effort the sensor subsystem components may be adapted to other DOE applications. For example the sensor components may be applied to a floor survey robot such as MAX developed by Savannah River Technology Center. The MVS sensor may be applied to identify mixed waste chemical components stored in glass or transparent plastic containers.

The CLR 3D mapping technology will be extended in performance by other government and industrial development applications to both higher speed and greater accuracy for robotics and metrology applications. A radiation hardened CLR 3D Mapper is being designed as a Remote Metrology Subsystem for the International Thermonuclear Reactor Subsystem. This development will take advantage of the compact no-moving-parts scanner technology development by CRC now being sponsored by METC under contract DE-AC21-94MC31190.

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3.10 BOA: Pipe-Asbestos Insulation Removal Robot System

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Abstract

The *BOA* system is a mobile pipe-external robotic crawler used to remotely strip and bag asbestos-containing lagging and insulation materials (ACLIM) from various diameter pipes in (primarily) industrial installations. Steam and process lines within the DOE weapons complex warrant the use of a remote device due to the high labor costs and high level of radioactive contamination, making manual removal extremely costly and highly inefficient. Currently targeted facilities for demonstration and remediation are Fernald in Ohio and Oak Ridge in Tennessee.

Overview

The two-phase program has progressed past Phase I with a proof-of-concept prototype development and testing scope, and is currently in Phase II. As part of the current scope, a complete regulatory, market and cost/benefit study has been completed. Current efforts are targeted towards the design of a prototype system to abate steam and process lines in the 4 to 8-inch diameter range at a DOE facility by October 1996. In the first-phase effort completed in December 1994, we developed and tested a proof-of-concept prototype system using preliminary locomotion and removal systems, with fiberglass insulation as a surrogate material (see Figure 1) [4].

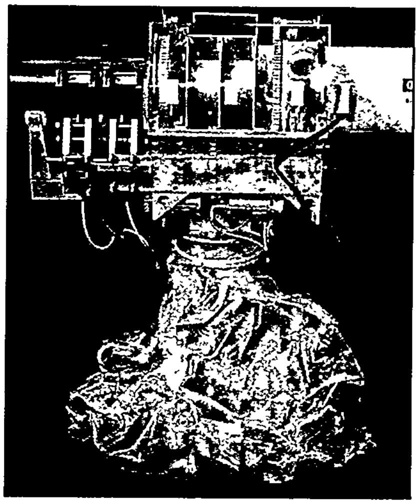

Figure 1. BOA Phase I Prototype Robot

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[1] Research sponsored by the U.S. Department of Energy's Morgantown Energy Technology Center, under contract DE-AR21-93MC30362 with Carnegie Mellon University, CMU-FRC 201, 5000 Forbes Ave., Pittsburgh, PA 15213; 412-268-6884, - 5895 FAX

Preliminary Experimental Results

It was determined that such a self-propelled, negative-pressure mini-containment system could meet EPA and OSHA mandated fiber-count levels during abatement operations, and that automated removal operations on piping could achieve a high removal rate. Using a mechanical cutting method (circular diamond-grit coated blade), we were able to achieve a net abatement rate of 4 ft./hr., which we knew we had to improve on to make the system more cost-effective. Compressing the material off the pipe once cut, was not sufficient to guarantee removal 100 percent of the time without some form of human assistance. This result lead us to the realization that a truly reliable and omni-directional cutting system was needed. The use of fiberglass as a surrogate was changed to Calcium Silicate (CalSil), since it was termed more akin to asbestos-containing material (ACM) in the field. This change made in-situ compression of the ACLIM unrealistic and the need for water-assisted/misted cutting and size reduction necessary, further aiding to reduce loose fiber emanation.

Based on these main and other secondary results, the DOE review panel decided to continue the project into Phase II. A revised statement of work for Phase II called for improvements and refinement to the design of the robotic removal head and locomotor system, further guided by a regulatory analysis and a market study and cost/benefit analysis to determine regulatory and performance requirements, market size and commercial potential of such systems for the DOE and within the abatement contractor industry.

Current Efforts and Results

The overall study clearly highlighted guidelines in the areas of regulatory compliance and certification, potential market sizes in the DOE and industry, as well as overall performance requirements and system-cost boundaries in order to be competitive and achieve substantial savings in the thermal insulation abatement market segment.

Regulatory Analysis

As part of the regulatory analysis, we charted a 'certification' path for any alternative abatement method proposed to EPA and OSHA. Even though OSHA/EPA do not certify equipment for use in abatement jobs, they do specify system performance in terms of allowable exposure limits (which aids somewhat in system design), work practices (process of using abatement techniques and equipment) and approval processes (permitting, notification, etc.). From a design stand-point, we will have to ensure we meet the fiber-emissions level regulations, which currently lie at 0.1 fibers/cc - as spelled out in 40 CFR Part 61 [3]. These restrictions imply the use of static and dynamic seals, positive airflow at all times, proper wetting and fiber-sealing and a proper deployment procedure to avoid any fiber release. The 'certification' process that BOA will have to go through, involves the drafting of a technical performance report by an on-site industrial hygienist or project designer with P.E. license which is then submitted to the DC-office of OSHA for review and acceptance - a process spelled out in 29 CFR 1926.1101 (g) (6) [2]. Local, state, and regional EPA and OSHA officials are kept abreast of the development and are invited to view the deployment and check for compliance on top of the required independent air monitoring. A full timeline and a list of deliverables and names within EPA and OSHA have been drafted for implementation during Phase II.

Market Study

A thorough review of thermal insulation systems and the asbestos abatement industry within the DOE and industry was conducted [1]. It was determined that the DOE has about 2 million linear feet of total piping (1.5M indoors, 0.5M outdoors) of medium bore-size (4 to 8 in. DIA.) in need of abatement, collected in the six major sites (Savannah River, Hanford, INEL, Oak Ridge, Rocky Flats, Fernald). A breakdown by site and indoors/outdoors is given in Table 1 below.

Table 1. DOE Pipe Footage Breakdown

DOE SITE	Outdoor	Indoor	TOTAL
Savannah River	110,000	562,000	672,000
Hanford	100,000	300,000	400,000
INEL	60,000	189,000	249,000
Oak Ridge	30,000	184,600	214,000
Rocky Flats	60,000	186,000	246,000
Fernald	70,000	48,700	118,700
TOTAL	430,000	1,460,300	1,890,300

The industrial market size was determined to be about 33.5 million linear feet each year over the next 10 years [1]. We believe that a BOA-like system, attacking only a portion of that market (4 to 8 inch diameter piping) currently abated with glovebags (22 percent) and then only in more sizeable installations where clearances are available for the robot to work on pipes, would be applicable to up to 0.5 million linear feet total within the DOE and about 1.5 million linear feet a year within the industrial market segment.

Cost/Benefit Analysis

Based on the potential performance of a robot abating at a rate of 40 linear feet per hour, compared with about 3 to 6 feet in DOE/Industry, with associated per-foot abatement costs ranging between $25 and $150 for Industry/DOE, it was determined that substantial savings could be realized with the use of such a robot system [5]. Overall abatement costs could decrease between 25 and 50 percent, depending on whether the system replaces a current glovebag or full-containment method. Overall savings were thus computed to lie between $10 million and $15 million for DOE, which does not even count savings due to reduced radiation exposure, work-crew reduction and insurance savings, overall worker safety, and potential litigation cost savings. Potential unit sales to DOE (and/or its M&Os and subcontractors) and commercial asbestos abatement contractors were estimated to be between 150 and 300 units over the next 7 years, depending on the size of the contractor and job, as well as the final production cost of the system.

Based on the study period at the beginning of Phase II, we also developed a new cutting method to allow more reliable rapid cutting and ease the waste transport. A new operational scenario reflecting the guidelines and lessons learned from the study itself is detailed below.

Operational Scenario

The BOA system consists of a robotic on-pipe locomotion and removal head sized for different pipe diameters, remotely controlled by a single operator from a button-box and inter-connected to off-board logistics support systems (see Figure 2). These off-board systems consist of the positioner that allows the system to be positioned on and off the pipe and around obstacles, the control and computer box to monitor and control all systems, the remote HEPA vacuum and bagging station, and the water-based pressure-washer system.

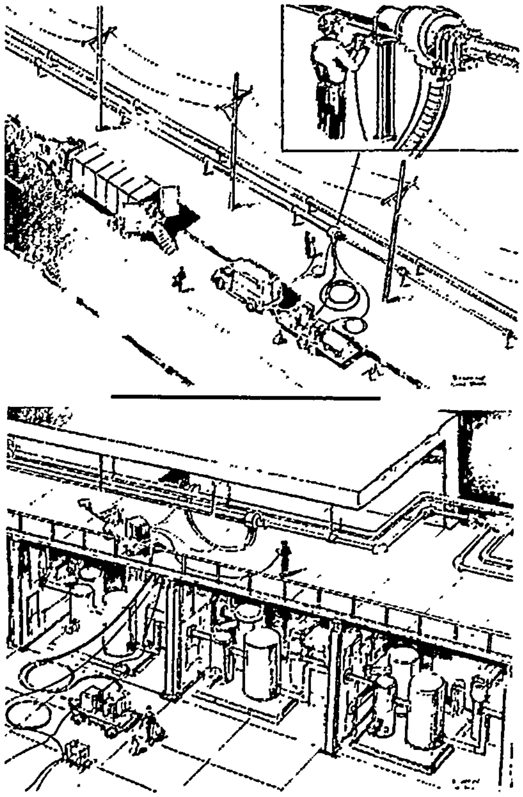

Figure 2. BOA Deployment Concepts

BOA is projected to be able to abate straight sections of pipe at a rate of 40 ft./hr. using a hybrid endmill/water-jet cutting system which can handle all forms of ACLIM, including aluminum lagging, steel bands and wires, wire-mesh and screws (cut by the endmill), and any form of insulation material such as the simulant CalSil (cut by the water-jet). A picture of lagging and insulation samples we will need to deal with is shown in Figure 3.

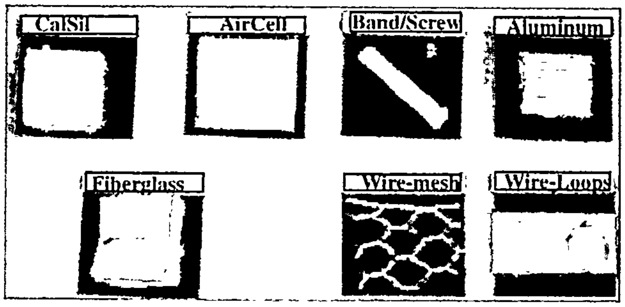

Figure 3. Lagging and Insulation Material

The robot can get past hangers unassisted, sealing the insulation left around the hanger (=/- 6in.) for manual post-removal. In the case of obstacles such as valves, junctions, bends, tees, etc., the robot is emplaced around the obstacle using the work positioner and letting the robot self-start behind the obstacle. Once a section of pipe has been cleared, the locomotor clamps onto the pipe and inches along the pipe using a triple tripod clamping mechanism connected by guide-rails and linear electric actuators. The diced-up insulation blocks are roughly 2 inches on edge, and are water-blasted into the waste-chute which leads into the vacuum hose connected to the HEPA vacuum via a water-separation and waste-bagging unit. The water is separated and re-used, while the waste material is bagged into standard 6-mil poly bags by the second operator. Bagging operations can occur as far away as 500 feet from the actual pipe abatement location.

Competing Technologies

The BOA system is unique in that it represents a new class of abatement technology that is currently not available, namely a self-locomoting negative pressure mini-enclosure for automated pipe-insulation abatement. The only 'mechanized' solutions for pipe insulation abatement contractors consist of a re-usable glovebag and a remoted vacuum filtering and bagging system as shown in Figure 4.

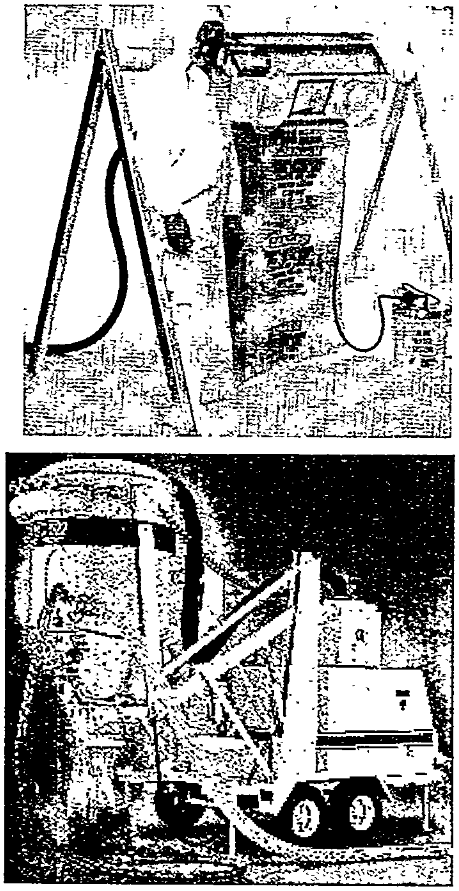

Figure 4. 'Competing' Technologies

Ongoing Work

We are currently in the design phase of the prototype system, which we intend to present to a DOE review panel in November 1995. Current plans are to build and test the robot system and carry out an acceptance test at CMU at the end of July 1996. Upon successful completion, DOE will build a full-scale partial cold-test replica of the designated final test site, where we intend to perform a full-scale cold-demo by October 1996, and thereafter a full-scale asbestos abatement field trial at the chosen DOE site location.

Acknowledgments

We would like to acknowledge the assistance of the METC COR, Vijendra Kothari, and Dr. Linton Yarbrough at DOE HQ for their assistance during the current project. Furthermore, we wish to acknowledge participation from many people at various sites for their assistance in gathering the data required for our study and their support in finalizing the field trial site. Carnegie Mellon University will develop the system and collaborate with an industrial partner to commercialize the technology at the conclusion of the program in 1996.

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[6] Site Survey Reports from several sites: Oak Ridge, Fernald, INEL, Savannah River, Rocky Flats, and Hanford.

3.11 An Intelligent Inspection and Survey Robot

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Introduction

Large quantities of mixed and low-level radioactive waste contained in 55-, 85-, and 110-gallon steel drums are stored at Department of Energy (DOE) warehouses located throughout the United States. The steel drums are placed on pallets and stacked on top of one another, forming a column of drums ranging in heights of one to four drums and up to 16 feet high. The columns of drums are aligned in rows forming an aisle approximately three feet wide between the rows of drums. Tens of thousands of drums are stored in these warehouses throughout the DOE complex.

ARIES (*A*utonomous *R*obotic *I*nspection *E*xperimental *S*ystem) [Ref. 1], is under development for the DOE* to survey and inspect these drums. The robot will navigate through the aisles and perform an inspection operation, typically performed by a human operator, making decisions about the condition of the drums and maintaining a database of pertinent information about each drum.

Objectives

The overall objective is to replace a weekly human inspection of drums using modern mobile robot, computer, and mechanical technology.

The integrity of each drum in storage is visually inspected weekly, according to regulations under the Resource Conservation and Recovery Act [Ref. 2], to determine if a drum has degraded to the condition that it should have its contents repacked into a new drum container. Currently, inspectors periodically roam the warehouses noting and reporting drum degradation. Typically the inspectors look for rust areas, streaks indicating leaks, dents, bulges, and tilting of the drums. These indicators identify *suspect* drums. The drums are stacked such that the side seam of the drum is in full view of the inspector. Empirical data has shown that a drum shows its first signs of degradation along this welded bead. A bar-code label is used to identify each drum. The bar-code is located to either side of the seam, in full view of the inspector. If the drum has degraded to the point that it warrants attention, the inspector identifies the drum using a

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Research sponsored by the U.S. Department of Energy's Morgantown Energy Technology Center under contract DE-AC21-92MC29115 with South Carolina Universities Research and Education Foundation (SCUREF), Strom Thurmond Institute, Clemson, SC 29634-5701; phone: 803-656-0953; telefax: 803-656-0958.

bar-code reader so that the drum can be retrieved by a fork lift and transported to the re-packing area.

Approach

The first phase of this three-phase project was a task-oriented, proof-of-principle phase in which demonstrations and reports were provided as the deliverables. The second phase was a technology integration effort to develop a single, commercializable prototype mobile robot capable of meeting many of the demands of the mission of environmental compliance and clean-up of DOE sites. During the third phase the prototype will demonstrated and evaluated in a DOE warehouse storage facility then productized for commercial availability.

An industrial partner, Cybermotion,[1] manufacturer of the Navmaster line of mobile robots was selected to join the university research and development team. This selection was based on the fact that the Cybermotion system is robust and has proven records in the areas of autonomous monitoring and security systems. Also, these systems are currently in use in other DOE and DOD applications.

Project Description

The Vehicle. Cybermotion, Inc. has developed a new 6-wheeled version of their Navmaster series of mobile robots that is the base mobile vehicle for ARIES. This new version consists of an improved and enhanced mobile platform (Model K3A)[Ref. 3] and a new subturret (Figure 1) that will permit turning around in a three-foot aisle. It has a capable ultrasonic imaging system used in navigation and collision avoidance and an automatic docking/charging system. Enhancements to the robot sonar system and a new lidar system have improved its ability to navigate in the drum aisles. Drum-referencing algorithms and camera-positioning algorithms have been included in the primitive instruction set for the new robot.

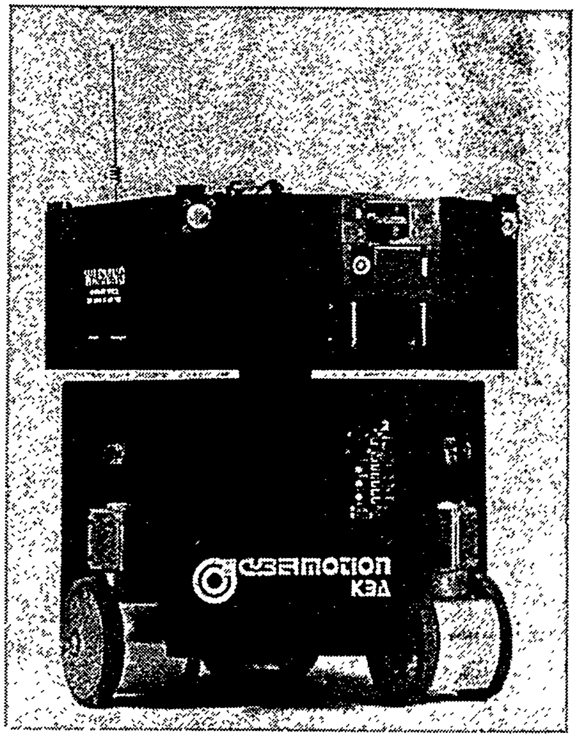

Figure 1
New Robot Base and Subturret

Computers and Control: Computer systems and control enhancements have been made at the University of South Carolina (USC). The computer systems (Figure 2) consist of an onboard system and an off-board supervisory system. The onboard computer system, housed in the robot sub-turret, provides control of the inspection processes and manages other onboard activities. The onboard computer is a VMEbus system using a MIPS R3000 proc-

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[1] Cybermotion, Inc., 115 Sheraton Dr., Salem, WV. (703) 562-7626.

essor board running the VxWorks[2] real-time operating system. The drum navigational algorithms, developed earlier in the project by USC and Cybermotion [Ref. 4], have been enhanced to accommodate the new robot.

Standard UNIX workstations are used for the offboard supervisory computers. The software is written to be portable across most UNIX systems and currently runs on Silicon Graphics, DEC, HP, and Sun workstations. Silicon Graphics systems are used for development purposes and some three-dimensional features require this system for acceptable performance. Provisions have been made for alternative representations in other systems. The offboard system provides three primary functions: *(i)* functional compatibility with the PC-based software provided by Cybermotion for control and programming of the basic robot, *(ii)* programming tools for creating the mission program, and *(iii)* the ability to monitor and control the robot during the inspection process. An assembler for the path language of the robot has been provided. This assembler operates in the DOS and UNIX environments.

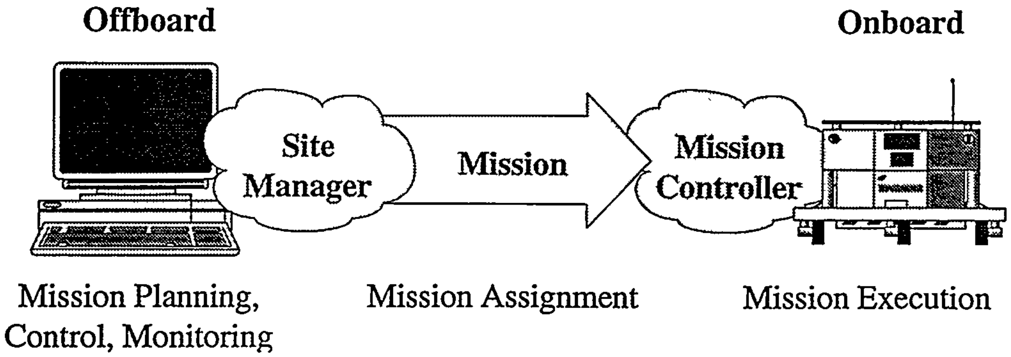

Figure 2.
Computer Systems for ARIES

Offboard computers networked via wireless Ethernet with onboard computers provide the high-level planning, monitoring, reporting, and general supervision of ARIES. Multiple control and monitoring stations may be employed. Planning the inspection task (the *mission*) begins with the implementation of a world representation of the robot's environment. A path planner automatically generates robot path programs for user-specified paths, based on the site description contained in the robot world. The mission program, used to control the inspection process, is down-loaded from the offboard system to the onboard computer where it is executed. The offboard systems may be used to monitor and control the system during the inspection process.

Mechanical Systems: A camera positioning system (CPS), designed by USC and Cybermotion, capable of performing survey and inspection of drums in the warehouse has been fabricated by Cybermotion. The CPS consists of four separate inspection modules, one for each drum in a four-high stack. Each module includes a camera, barcode scanner, and strobe lighting. For the

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[2] Wind River Systems, 1010 Atlantic Ave., Alameda, CA 94501.

drum inspection process, at each stack of drums the CPS extends up and folds out to deploy the four inspection modules at various heights on the drums required by the vision system. Two "photo" positions are required for each drum. These positions are determined by requirements of the computer vision inspection system and are programmed as a table in the primitive instruction set. The CPS is retracted to its more compact position (Figure 3) for traveling in the warehouse en route to inspection assignments and between drum aisles. It has an overall height of approximately eight feet in this collapsed position.

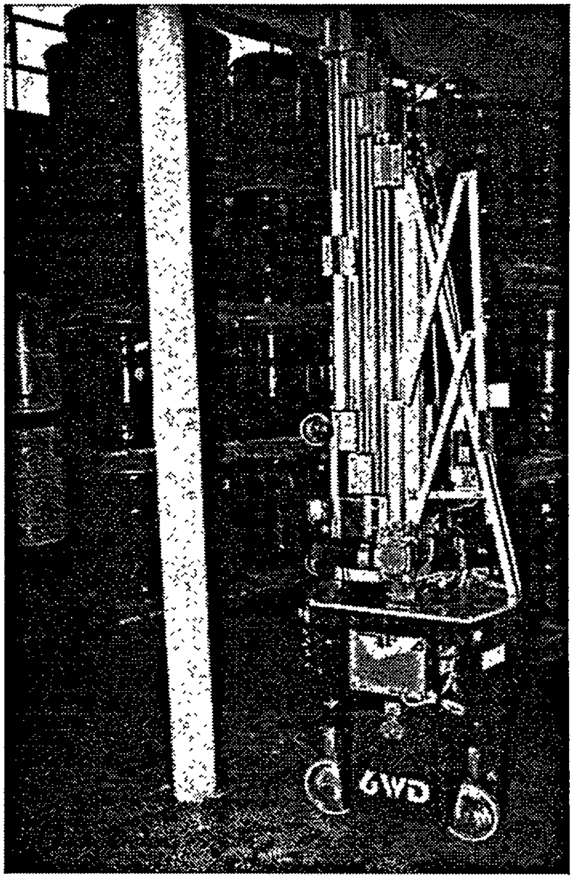

Figure 3
ARIES in Lowest Position

During inspection assignments in the various aisles of the warehouse, ARIES will encounter drums of three different sizes (55-, 85-, and 110-gallon capacities) stacked up to four high. The system will determine the size of each drum stack and position the CPS accordingly to the required two positions per drum. The upper position for 55-gallon drums is shown in Figure 4.

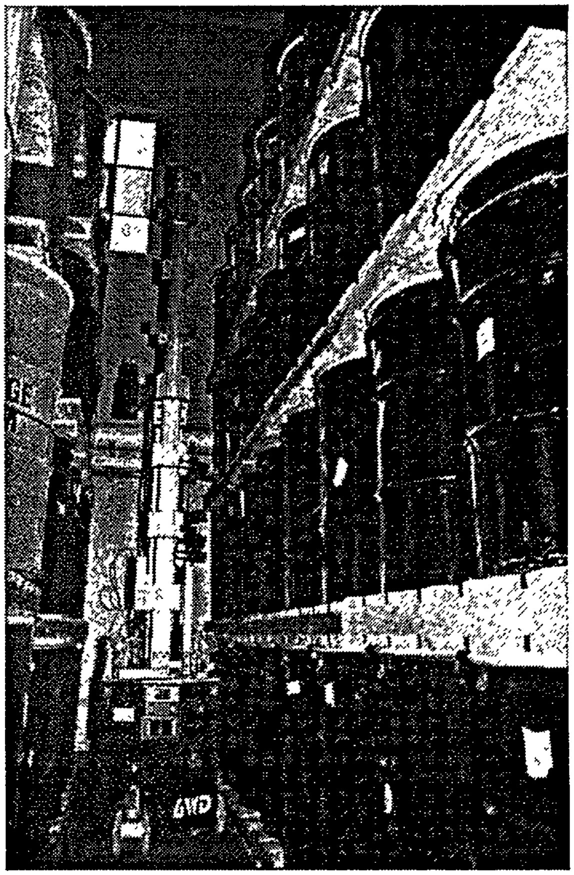

Figure 4
ARIES in 55-Gallon Drum Position

Vision System. The application payload of the CPS includes computer vision and bar-code scanner modules developed at Clemson University (CU). The vision system [Ref. 5] is used to analyze the drums' external and visible conditions and to determine their structural integrity. The overall function of the vision module is to locate *suspect* drums and to report these conditions. Once drums have been located by the robot's navigation system, visual assessment of drum condition is primarily an autonomous assessment of visible and quantifiable surface characteristics.

The visible surface blemishes which indicate probably drum failure are rust patches of approximately 0.5 x 0.5 inch size and paint blisters indicating internal surface rust. Basic sensor requirements for image delivery and image acquisition led to the specification of a color camera. Lighting for image acquisition has been carefully considered. The variability in site ambient lighting and power limitations of the mobile robot requires the use of a strobe-based image acquisition system. This ensures the on-robot illumination dominates ambient lighting and allows a reduced camera aperture and consequently increases imaging depth of field. A modular vision acquisition and analysis system is used. A learning algorithm is provided which adjusts the algorithm parameters according to information given by a human tutor.

Detail Survey & Inspection: A supplemental prototype dexterity package has been developed at CU to provide more sophisticated manipulations for more detailed inspection of the waste containers. A camera mounted on the end of a six degree-of-freedom revolute manipulator can be used in a telerobotic mode for close detailed inspection of the drum surface. The bar-code can be located and read using an integral camera and scanner. An additional sampling tool can be used to obtain surface samples of suspect areas and returned for operator analysis. This module will be demonstrated as a separate component during the Phase 2 demonstration at USC, but there are no plans for further development or testing at a DOE warehouse site since this is not a requirement in the current inspection process.

Radiation Hardening Study: A study has been conducted to determine the requirements for fabricating a radiation-hardened version of the mobile inspector [Ref. 6]. Requirements for the low-level warehouse drum inspections do not dictate radiation hardening, since the radiation levels of the inspection environment are low. However, future uses of such a mobile system in other applications may require a radiation-hardened system.

Accomplishments

Current mobile robot, computers, and mechanical technologies have been integrated to produce a practical commercial mobile robot system to be employed in a complex environment as a proposed solution to a vital National problem. The current ARIES prototype will be tested and evaluated at a DOE warehouse storage site and a commercial mobile system will be delivered to the DOE. Additional systems will be available from Cybermotion, Inc.

At the conclusion of Phase 2 a report will be available that includes plans and costs associated with the development of a radiation-hardened version of ARIES for applications that require high radiation considerations, such as decontamination and decommissioning operations.

Benefits

The mobile robot inspector, ARIES, is designed to relieve the warehouse inspector of the tedious and mundane task of inspecting warehouse-stored drums. Cost savings, reduced worker radiation exposure, improved documentation, improved quality with inspection consistency, as well as minimized disruptions to daily warehouse operations, are some of the anticipated benefits from an autonomous inspection.

A commercial mobile system will be available to the DOE's environmental management program for use in drum storage inspection. It is anticipated that the system will be applicable to other DOE applications such as decontamination and decommissioning the nuclear sites.

Future Activities

A prototype demonstration will be conducted at USC on November 30, 1995. During 2QFY96 testing and demonstrations will be scheduled at a drum storage warehouse at the DOE Fernald site. Other test sites are under consideration.

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ARIES is a collaborative effort between Clemson University (CU), University of South Carolina (USC), and Cybermotion, Inc. The team members are: Joe Byrd (Principal Investigator and Project Manger), Jerry Hudgins, Bob Pettus, and Dave Rocheleau of USC; Darren Dawson, Fred Sias, and Bob Schalkoff of CU; Ed Hamilton and Angela Harrington of SCUREF; and, John Holland and Ken Kennedy, and staff of Cybermotion, Inc. Also, 10 graduate students at USC and CU have been invaluable team members. Cybermotion is under subcontract.

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3.13 Houdini: Reconfigurable In-Tank Robot

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Introduction/Needs

RedZone Robotics, Inc. and Carnegie Mellon University (CMU) are developing a tethered mobile robot, Houdini, to work inside waste storage tanks in support of the Department of Energy's Environmental Restoration and Waste Management (EM) Program. This project is funded by the DOE's Environmental Management Office of Technology Development through the Morgantown Energy Technology Center (METC). Our goal is to develop technology that is useful for in-tank operations throughout the DOE's EM program.

The first application of the Houdini system is to support the waste retrieval action planned for the final remediation of the Fernald site's waste silos. RedZone and CMU have discussed potential applications for the system with personnel from several other DOE sites, and have found that the system would be widely useful in the DOE complex for tasks both inside and outside of waste storage tanks. We are tailoring the first implementation of the Houdini system to the specific needs of the Fernald silo remediation. The Fernald application-specific design constraints are primarily interface issues and should not interfere with the utility of the system at other sites.

In addition, DOE personnel at the Oak Ridge National Laboratories (ORNL) have expressed a strong interest in the Houdini system. They have a target application scheduled for mid-1996. This program represents a unique opportunity to develop a new technology that has immediate application in two CERCLA cleanup actions; the proposed applications at Fernald and ORNL support Federal Facility compliance agreements.

Objectives/Problem

The primary application for the Houdini system is to support the final remediation of Silos 1, 2, & 3 at the Fernald Site. Houdini will perform essential missions in support of this final remediation effort that will retrieve waste from the tanks and vitrify it for long term storage.

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The CRU4 area at the Fernald site contains 4 above-ground, concrete waste silos. All four domed waste silos are 24.4 meters (80 feet) in diameter, 11 meters (36 feet) high at the center of the dome, with 8.3 meter (27 foot) high vertical walls. Four 0.51 meter (20 inch) diameter manways are evenly distributed around each tank dome at 15 feet from the side walls, at a slant of 17 degrees from horizontal. A fifth 20 inch manway near the center of the dome will be eliminated when a 6 foot opening is created to support the remediation activity.

Some structural deterioration has been noted in the walls and domes of the silos, resulting in load restrictions. No detectable load is allowed on the 6.1 meter (20 foot) diameter center section of the dome. On other dome areas, the maximum live load is limited to 700 pounds including personnel and gear.

Waste material in Silos 1 and 2 is described as K-65 material and has the consistency of toothpaste. The waste is covered with a 0.3 meter (12 inch) thick layer of Bentonite clay to reduce radon emissions from the waste. Material in Silo 3 is a light, dry metal oxide powder similar in consistency to talcum powder. The Silo 3 waste may be compacted near the bottom of the silo. In addition, each silo contains pipes, wrenches, sample bottles, gloves, and other debris that has fallen into the tanks over the years.

The fourth silo is identical to the other three, but was never used for waste storage. It will be used as an uncontaminated mock-up facility to fully test all procedures prior to remediation of other silos. Silo 4 may be partially filled with a surrogate waste material to support these tests.

To enable the waste retrieval operations in light of the dome load restrictions, a superstructure has been constructed over Silo 4. An equipment room situated over the center of the dome is supported by the superstructure. Seals will be installed between the equipment room and the silo dome and a six foot diameter opening will be made in the silo dome directly under the equipment room. Doors in the equipment room floor will provide access to the tank. Controlled entry points, rails, winches, equipment carts, and other mechanisms in the equipment room will support the deployment and retrieval of equipment into the silo.

The primary retrieval method for Silo 1 and 2 waste will be hydraulic removal. A sluicing pump will be lowered into the tank from the equipment room. Water will be added to the waste material and the liquefied waste will be pumped out of the tank. In Silo 3, pneumatic conveyance will be used to retrieve the waste material. The methods will remove the bulk of the waste materials from the tanks, leaving only debris and a waste heel to be removed by other means.

Approach/Solution

The Houdini system will be used for heel and debris removal from the tanks, during and after the bulk material removal by sluicing and pneumatic conveyance. In Silo 4, Houdini's capabilities will be fully tested in an uncontaminated environment. The silo will be partially filled with surrogate waste material and debris that approximates the waste properties in Silos 1 and 2. During bulk material removal, Houdini will be deployed to remove debris material that interferes with the sluicing operation. Using a shearing tool and gripper, Houdini would be deployed to gather debris, size reduce the debris as necessary, and load the debris into a tram bucket for retrieval from the tank. After the bulk waste removal action is complete, Houdini will be deployed to perform waste heel removal and debris collection. To perform the heel removal, a small sluicing pump will be lowered into the tank. Houdini will use its gripper to deploy a water spray nozzle. Water spray will be used to mobilize the heel material and wash it toward the sluicing pump for removal. The Houdini plow blade will be equipped with squeegees on the sides and bottom and will be used to push slurried waste material toward the sluicing pump. The edge of the plow blade can be used to push waste material away from the side wall of the tank to clean the edges of the tank floor. Houdini can also be used to wash down or apply a spray coating to the tank side walls for decontamination.

In Silos 1 and 2, Houdini will be used to support waste retrieval for final remediation in the same modes as described above for the Silo 4 functional test. The radioactive waste in Silos 1 and 2 will be primarily retrieved by sluicing, after which, the waste will be vitrified for long term storage.

In Silo 3, Houdini will support waste retrieval for final remediation in conjunction with the bulk material removal by pneumatic conveyance. Specifically, Houdini will perform debris collection, size reduction, and removal during and after bulk pneumatic conveyance, and perform heel removal by deploying a pneumatic vacuum hose. In addition, Houdini could be used to plow waste material to a central point for pneumatic retrieval. Houdini could also deploy tools to assist in removing compacted waste material from the tank floor.

Project Description/Technology

System Overview

The Houdini system consists of five main components and their subsystems; the vehicle, deployment system, PDCU, control consoles, and tooling.

Vehicle. The vehicle is a hydraulically-powered, track-driven, folding frame machine similar to a small bulldozer. The vehicle can fold to fit through a 0.57 meter (22.5 inch) diameter opening for deployment, and is equipped with a plow blade and a manipulator arm. The plow blade also folds for deployment and can be height-adjusted for plowing various materials at various rates. The manipulator is a Schilling Titan class hydraulic dexterous manipulator, which can deploy a variety of tooling for performing work inside a tank. The vehicle tether is attached to the rear of the folding frame assembly. The tether termination will support the full weight of the vehicle and tooling to enable deployment and retrieval. Two camera and light assemblies provide visual feedback for remote operation. One camera and light unit is mounted on the forearm of the manipulator. The camera is aimed by orienting the manipulator. The second camera unit includes a pan and tilt unit and is mounted on a mast at the manipulator shoulder. A microphone will provide audio feedback to the operator. Navigation system sensors will be installed on the vehicle as part of the navigation system interface.

Deployment System. The deployment system is designed to interface with the superstructure and equipment room above the Fernald waste silos. It is an important part of the Houdini system. First, it provides a convenient way to remotely manage and store the 150 feet of tether that is the lifeline of the Houdini vehicle. Second, it provides the lifting force that is needed to lower and raise the vehicle into and from the tank. Third, it provides a "docking area" where the vehicle can be secured during storage or transport, and lastly, it provides an area where spare parts or tools can be stored when not in use.

Tether Reel. The tether reel is a "spool" looking device that is 48 inches in diameter and 30 inches wide. A flange at each end contains the tether on the drum. Payout of the tether is controlled by a mechanical level-wind system that ensure that the tether does not cross over on itself and tangle. The tether reel is driven by a hydraulic motor with a "power-off" brake in case of hydraulic power loss. The hydraulic motor is sized to allow a pull force of 2000 lb tangent to the reel surface. A payout sensor is used to monitor the length of tether that has been reeled out. The sensor shall also indicate ends of travel (i.e., when the tether is completely in or out). A means of manual retrieval is necessary in case of hydraulic power failure. This is accomplished with an external, battery powered, hydraulic supply and manually operated valve.

Tether. The tether is used to lower and raise the vehicle in the tank and provides control signals and electric and hydraulic power to the vehicle. The tether will be a custom fabrication that includes:

- Strain-relief termination
- Hydraulic supply and return lines
- Shielded-twisted pairs for control and feedback signals
- Mini-coax lines for camera signals
- Shielded-twisted pairs for navigation system signals
- Conductors for power to onboard valving
- Kevlar braid for structural support (to carry the weight of the vehicle)
- Abrasion resistant coating

Power Distribution and Control Unit. An environmentally sealed and temperature controlled power distribution and control unit (PDCU) will be installed on the superstructure, outside the equipment room. The PDCU includes the electric transformers and distribution/conditioning equipment, the control system, and tether and control system interface connectors. A separate enclosure will house the electrically-powered hydraulic power supply.

Control Console. The operator console provides the operator interface to the Houdini system. The console includes joysticks, switches, a master manipulator, and video monitors for controlling system functions and monitoring system operation.

Navigation System Interface. An interface will be provided to the Position and Orientation Tracking System (POTS) which has been developed at ORNL. ORNL has agreed to make POTS available to this program as Government-Furnished Equipment. POTS will provide accurate feedback on the vehicle's position and orientation inside a tank to enable more efficient and robust controls.

Control Trailer. A control trailer will house the operator control console. The trailer will be a simple mobile, industrial trailer that provides heated and air conditioned real estate for the operator. Tie downs will be provided for securing the trailer against high wind. The trailer will be structurally capable of being moved with the control console inside. The trailer will require 110 volt site power.

Suitcase Controller. A hard-wired suitcase controller will be available to perform system checkout, local operations and provide for emergency operations in the case of console/control computer or telemetry failures between the control center and the deployment system. Switches, buttons, and a single remote viewing monitor will provide for simple operations from the suitcase controller.

Tooling. Specialized tooling will be provided to enable the use of the Houdini system in support of the Fernald waste retrieval plans. The bottom and sides of the plow will be equipped with squeegee blades to provide efficient mobilization of the waste slurry on the floor of the tank. A gripper, shear, and scoop will be provided for deployment from the manipulator. For sluicing operations, Fernald will provide a small pump for heel removal operations; RedZone will equip the pump with a water line, hose reel, and spray nozzle that can be deployed by the Houdini manipulator to spray-wash waste material toward the sluicing pump. For vacuum retrieval operations, Fernald has agreed to provide a hose grip that will attach to the manipulator and enable the deployment of a pneumatic vacuum hose.

Application/Benefits

As a technology for supporting the DOE's EM program and in comparison or collaboration with other competing technologies, Houdini provides many benefits. The Houdini system is designed to eliminate or reduce potential public and operational health risks associated with work on DOE tanks. The system provides fully-contained remote operation, reducing the risk of spreading contamination outside of the tanks. Because of Houdini's similarity to bulldozers and backhoes from the construction industry, it provides simple, intuitive, and efficient waste handling techniques. Houdini's transportation, installation, deployment, and removal operations are simple due to its compact size. Houdini's simplicity and operational capability lead to cost efficiency with respect to development, operation, and maintenance.

Evaluating the merits of the Houdini system for these applications requires comparing it to competing technologies. In comparison to mobile robot systems that are currently available, Houdini's folding frame technology provides a substantially larger work platform which can fit through existing tank openings. As a larger platform, Houdini is more powerful, more efficient, and more capable than other, smaller mobile systems.

Several non-robotic retrieval methods are being considered for use in DOE tanks. These technologies, such as sluicing, pumping, and pneumatic conveyance, are appropriate or preferred technologies for some of the tanks in the complex. As it will at Fernald, Houdini could assist in the application of these retrieval and conveyance methods. In addition, the current design could be applied for use in Oak Ridge's north and south tank farms.

Depending on specific work tasks and application sites, Houdini can be deployed to either complement or replace a long-reach manipulator (LRM) system. Used in conjunction with LRMs, Houdini provides additional or enhanced capabilities inside a tank. In tasks where Houdini is useful instead of LRMs, Houdini will be simpler and less expensive to deploy, operate, retrieve, and decontaminate than LRMs.

Future Activities/Applications

In addition to the primary application at Fernald, other applications for Houdini have been identified in support of tank waste retrieval operations in the DOE and private sector. Also, several tasks outside of tanks have been identified for which Houdini would be useful.

Oak Ridge North and South Tank Farms

The north and south tank farms at Oak Ridge National Laboratory have a total of 16 domed, cylindrical, single-shelled, underground storage tanks made of Gunite (similar to concrete), ranging in diameter from 20 to 50 feet and equipped with 24 inch diameter manway penetrations. These tanks were used to store laboratory waste and are expected to contain a wide variety of materials, with estimated radiation levels of 1 to 100 R. During 1983-1984 the tanks were emptied through a sluicing method, leaving a heel of up to several feet thick at the bottom. The heel waste must be removed to prevent the migration of waste material out of the tanks. ORNL is under a Federal Facility Compliance Agreement to complete a CERCLA treatability study on the Gunite and associated tanks. The baseline plan for this study includes the evaluation of both vehicle- and arm-based retrieval systems. The current plan at ORNL is to evaluate the Houdini vehicle during this study, and if successful, Houdini might be selected for the final remediation retrieval action.

Other DOE Tank Applications

The Houdini system is useful in a variety of other DOE tank waste retrieval operations. Houdini could be deployed in a tank prior to the major removal action to collect additional information about the waste content and tank interior.

In support of other in-tank work systems, such as long reach manipulators, Houdini could be used to deploy cameras, lights, and sensor systems. The mobile deployment of such monitoring equipment will provide viewing and data gathering flexibility that cannot be achieved by mounting such equipment on fixed masts or on a long reach manipulator.

The long reach manipulator (LRM) systems that are being developed for tank waste retrieval will require a variety of tools to accomplish their tasks. The Houdini crawler could serve as a mobile tool carrier for the LRM, carrying several tools and making them available at the most appropriate location inside the tank.

In support of final tank decontamination and decommissioning, Houdini could deploy tools to scarify internal tank surfaces.

Commercial Tank Applications

Periodic cleaning and inspection of storage tanks in petro-chemical industries are becoming common maintenance procedures. It is likely that these procedures will be required by law in the US in the next few years. We have been in contact with several service providers in the petro-chemical industry, who have expressed an interest in the Houdini system in support of these operations.

Non Tank Applications

Alternate uses currently envisioned for this system include indoor as well as outdoor tasks. In support of buried-waste excavation programs, Houdini could perform fine excavation and monitoring to assist a larger remote excavator, perform fine excavation to isolate and extract specific objects, and assist removing a drum in one piece. In support of decontamination and dismantling programs, Houdini could be used as a small platform to gain access through tight areas for selective equipment removal and could lend assistance to larger worksystems as a tool-carrier platform, size-reduction system, or waste packaging system. In support of surveillance and monitoring operations, it could perform such functions as monitoring drum storage areas and decommissioned processing areas requiring access to tight corridors. Removal of the tether is possible through the use of a gas-engine or batteries and the interface of a radio telemetry system.

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