**Nuclear Waste Management
Quarterly Progress Report
October Through December 1979**

A. M. Platt
J. A. Powell

April 1980

Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
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NUCLEAR WASTE MANAGEMENT
QUARTERLY PROGRESS REPORT
OCTOBER THROUGH DECEMBER 1979

Compiled by
A. M. Platt and J. A. Powell
Nuclear Waste Technology Program

April 1980

Prepared for
the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Richland, Washington 99352

FOREWORD

This document is one of a series of technical progress reports designed to report on radioactive waste management programs at Pacific Northwest Laboratory. These programs are funded by the Department of Energy's Office of Nuclear Waste Management. Only programs in the scope of this document that have significant, reportable results are included.

These reports are issued quarterly. The prior one in this series is Nuclear Waste Management Quarterly Progress Report July Through September 1979, PNL-3000-3.

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1. HIGH-LEVEL WASTE IMMOBILIZATION

D. E. Larson - Project Manager

The primary objective of the High-Level Waste Immobilization Program (HLWIP) is to develop and demonstrate technology for converting high-level radioactive waste to a glass form and for packaging it for disposal in a repository. This effort includes the development of suitable glass forms for existing waste (Western New York Nuclear Service Center, defense) and future wastes (commercial, alternative fuel cycles). A secondary objective is the application of the process and equipment technology to alternative waste forms (supercalcine, matrices, etc.). Government agencies and commercial industry will be assisted in applying this technology to their specific applications.

The program is a part of the overall National High-Level Waste Long-Term Management Program under the direction of the lead Savannah River office, and it contributes to the 1.3 Fixation area of that program. This function contains two work elements, and the program is structured accordingly.

Work Element 1.3.1 Waste Form Development and Characterization

Work Element 1.3.2 Process and Equipment Development.

SUMMARY

In hydrothermal environment tests, three simulated high-level waste (HLW) glasses, supercalcine, and SYNROC exhibit similar leaching behavior. Releases from the glasses are lowest at temperatures below 250°C (the expected repository temperature range).

Corrosion tests of candidate metallic-container materials in 250°C Hanford ground water are under way, and several polymers that show good resistance to 200°C water in autoclave tests are undergoing gamma irradiation.

Methods to reduce melt-foaming in ceramic melters were investigated. Including monoxides of iron, nickel and manganese in the calcine significantly improved melt quality and reduced foaming. Addition of cornstarch to the melt, which inhibits foaming in laboratory studies, continues to be evaluated.

A successful pilot-scale test of the spray calciner in-can melter process was done using simulated waste representing the combined

contents of the neutralized and Thorex waste tanks at the Western New York Nuclear Service Center. Flexibility of the process for handling high-sodium waste was demonstrated.

WASTE FORM DEVELOPMENT AND CHARACTERIZATION

The goal of this task is to develop glass waste forms and evaluate their properties and to develop and evaluate engineered barriers capable of containing wastes for time periods of up to 1000 years.

Laboratory studies of the 11-component glass were completed, and analysis of data continued. Understanding the effects of crystal phases on properties is a major goal.

Investigation of a newly developed, high-durability waste glass ("Ultraglass") continued with autoclave testing and alteration of the original composition to include defense and commercial simulated wastes. Early results indicate that the glass has an exceptionally low leach rate at 90°C when made with a 28-wt% defense waste loading. Initial studies at higher temperatures

(200°C) show leaching comparable to that of other waste glasses.

Hydrothermal tests of SYNROC-B, a super-calcine, three simulated HLW glasses, and a pyroxene rhyolite obsidian indicate that the ceramics are less temperature-dependent than are the glasses. At temperatures above 250°C the ceramics show lower releases, but below 250°C (where actual repository temperatures are expected to lie) releases from the waste glasses are lowest.

A corrosion screening test of candidate metallic-container materials in 250°C Hanford ground water is under way to provide data on general (bulk) corrosion, stress-corrosion cracking, and the corrosion of weldment heat-affected zones.

A group of polymers that shows good resistance to 200°C water in autoclave tests has received a gamma (^{60}Co) exposure of $5 \times 10^8 \text{R}$. Mechanical testing (tensile, impact, elongation) of these specimens is planned for the next reporting period.

To assess the long-term durability of metals with a view toward their use in containment systems, the durability of metals from archeological objects and of meteorites and native metals was investigated.

Water adsorption properties of bentonite clay in constant humidity atmospheres were determined to be about 14 wt% at 25°C and 0.5 to 0.8 wt% at 85°C.

PROCESS AND EQUIPMENT DEVELOPMENT

This task provides the process and equipment technology to produce glass, glass-ceramics, and glass marbles for high-level waste immobilization. Specific equipment to be developed includes the spray calciner, in-can melter and calcine-fed ceramic melter. Verification tests to confirm that waste processing technology is ready for plant application are part of the effort.

Laboratory tests investigating melt-foaming showed that: 1) silicon carbide does not produce satisfactory results when used at the 0.5% level; 2) use of monoxides of iron, nickel and manganese in the simulated calcine significantly improves the quality of the melt and greatly reduces foaming; 3) when the melt is in contact with K-3 refractory the foaming appears to be reduced; and 4) frit that requires a high melting temperature is not compatible with Savannah River Laboratory calcine when blended at a ratio of 18:7.

Foam prevention studies using larger-scale tests with the experimental ceramic melter showed that a minimum cornstarch addition of ~0.75% to the liquid-fed ceramic melter feed is necessary to effectively prevent foaming. As expected, the carbon dioxide and carbon monoxide offgas concentrations increased as the cornstarch feed was increased, the CO/CO_2 ratio remained very low (<0.007), indicating that complete combustion of cornstarch took place, and the oxide concentration remained high ($>19\%$) because of melter air inleakage.

A test of the spray calciner/in-can melter process was made using simulated waste representing the combined contents of the waste tanks at the Western New York Nuclear Service Center. The test demonstrated the immobilization of combined neutralized sludge, neutralized supernate, and high-thorium acidic waste and showed that the process is flexible enough to handle high-sodium waste liquids.

In a Savannah River-sponsored 5-day run of the spray calciner/ceramic melter system using state defense wastes, foaming continued to limit production rates, and the melter refractory and calciner filters corroded significantly. Equipment to analyze the effluents above the melter surface of the coupled spray calciner/ceramic melter system is being

tested. Determination of the vapor composition is being determined, and methods to control its oxidizing potential are being studied for a better understanding of the corrosion problem.

A draft safety assessment completed for the spray calciner/in-can melting process, identifying 96 accident scenarios and evaluating 13, indicates that postulated accidents in the solidification facility generally lead to inconsequential environmental source terms.

Preparation of the draft of the Spray Calciner/In-Can Melter Manual is about 60% complete and is progressing on schedule toward the September 1980 issue date.

Installation of the Remote Lab-Scale Waste Treatment Facility equipment in 324 Building D-Cell is nearly complete. Some items are being modified somewhat to facilitate remote installation, operation and maintenance. Standard operating procedures, run plans and the Operational Safety Report are complete. An operational readiness review will be conducted by DOE-RL prior to beginning hot operations.

PAPERS

"Leaching and Volatility Results from 11 Component Generic Glass Study." Presented at November 1979 meeting of Materials Research Society.

"Processing of High-Temperature Simulated Waste Glass in a Continuous Ceramic Melter." Presented at November 1979 meeting of Materials Research Society.

"Temperature Dependence for Hydrothermal Reactions of Waste Glasses and Ceramics." Presented at November 1979 meeting of Materials Research Society.

W. J. Gray, "Volatility of Some Potential High-Level Radioactive Waste Forms." To be submitted to Radioactive Waste Management: An International Journal.

R. P. Turcotte, J. W. Wald, and R. P. May, "Devitrification of Nuclear Waste Glasses." Presented at International Symposium on Scientific Basis for Nuclear Waste Management, Boston, MA, November 26-29, 1979.

2. ALTERNATIVE WASTE FORMS

J. M. Rusin - Project Manager

The mission of the Alternative Waste Forms Program is to develop and evaluate selected processes and waste forms as alternatives to the reference borosilicate glass process for immobilizing high-level radioactive liquid wastes. Specific waste-form technologies being developed are glass ceramics, coatings, and matrices that are included in the multibarrier concept.

The program is a part of the overall National High-Level Waste Long-Term Management Program under the direction of the lead Savannah River office and contributes to the 1.3 Fixation area of that program. This function contains two work elements, and the program is structured accordingly:

Work Element 1.3.1 Waste Form Development and Characterization

Work Element 1.3.2 Process and Equipment Development.

SUMMARY

Comparative study tests have been completed on seven alternative waste forms. The most favorable properties were exhibited by sintered supercalcine SPC-5B and waste glass 76-68.

Since a radiation exposure of 3×10^{17} alpha/g, no significant changes in integrity or structure of the titanate phase, $Gd_2Ti_2O_7$, have occurred. This phase is frequently found in waste forms, including glass.

The effect of transmutation reactions, e.g., $^{137}\text{Cs} \rightarrow ^{137}\text{Ba}$, on the structure and properties of waste forms is being investigated. Seven cesium-containing samples have been irradiated, and a search has begun for old ^{90}Sr compounds.

The reaction kinetics of graphite with water/air mixtures has been determined in the temperature range 200° to 300°C. The reaction rates of carbon with water/air mixtures are now being determined in the presence of gamma radiation (1.6 to 3.2×10^5 R/hr). Leach tests on coated spherical particles of supercalcine are also being investigated. The concentration of simulated waste elements

in the leachate for carbon-coated particles was about an order of magnitude less than concentrations in the leachate for the uncoated particles.

Equipment for studying the feasibility of low-temperature (~550°C) application of pyrolytic carbon coatings has been assembled and scoping tests begun.

Simulated zirconia waste pellets, produced in a disc pelletizer at Idaho National Engineering Laboratory (INEL), were received. These pellets were used in tests of a vertical sintering kiln. Three tests of the vertical sintering kiln conducted at 800°C were successfully completed.

About 500 kg of green supercalcine pellets were produced at Ferro-Tech, Inc., with ~5% cornstarch on a dry basis added during pelletization to strengthen pellets. The pin mixer produced pellets of ~20 mesh size, and the disc pelletizer produced pellets from 1 to 25 mm dia. A pin mixer and disc pelletizer have been installed at PNL to fabricate ceramic pellets.

Vacuum casting of 90% lead-10% tin in a marble-filled canister resulted in greater infiltration of the metal between marbles

than did gravity casting. It was decided to use in-can melting of lead shot to encapsulate marbles in a lead matrix. An encapsulation test by this method yielded a high-quality product.

The draft of the Alternative Waste Form Process Preconceptual Design Study has been issued for internal review.

WASTE DEVELOPMENT AND CHARACTERIZATION

The goal of this task is to develop and assess glass ceramic, coated particles, and matrix waste forms for the immobilization of nuclear wastes. An additional goal is to evaluate the radiation stability of crystalline waste forms.

The volatility, leach rates, and mechanical strength of seven alternative waste forms have been determined by means of standard test procedures under the "comparative studies" effort. The waste forms investigated were sintered PW-9 calcine; hot pressed PW-9; PW-9 plus 50% frit; supercalcine SPC-5B; glass 76-68; Portland type II cement plus 10% PW-9; and Portland type II cement plus 10% SPC-5B. The 100% PW-9 samples showed the lowest strength, the highest volatility, and the highest leach rates of any of the materials tested. Sintered SPC-5B supercalcine and the glass 76-68 exhibited the most favorable overall properties.

Two titanate phases, with pyrochlore structures, $Gd_2Ti_2O_7$ and $CaZrTi_2O_7$, are frequently found in waste forms, including glass. The radiation stability of these phases is currently being investigated in cooperation with the Hahn-Meitner Institute, Germany. The $Gd_2Ti_2O_7$ material has been successfully fabricated. After a radiation exposure of 3×10^{17} alpha/g, no significant changes in integrity or structure have occurred.

The effect of transmutation reactions, e.g., $^{137}Cs \rightarrow ^{137}Ba$, on the structure and properties of waste forms, is being investigated. A total of seven waste forms including five glasses, supercalcine SPC-2 and pure pollucite ($CsAlSi_2O_6$), were irradiated to obtain a calculated "useful" conversion of cesium to barium of ~20%. Leaching data are currently being obtained. The x-ray diffraction data obtained to date have been poor because of the strong radiation background. An attempt is being made to examine the pollucite specimen by electron diffraction techniques. Besides the irradiated materials, a search has begun for old ^{90}Sr and ^{137}Cs compounds that can be examined for transmutation effects.

The reaction kinetics of graphite with water/air mixtures has been determined in the temperature range 200° to 300°C. A "Letter to the Editor" describing this work is being published in the first issue of the new Radioactive Waste Management: An International Journal. The reaction rates (leach rates) found were much lower than those obtained for other waste forms or for very stable ceramic materials such as Al_2O_3 , ZrO_2 , SiO_2 , etc. The reaction rates of carbon with water/air mixtures are now being determined in the presence of gamma radiation (1.6 to 3.2×10^5 R/hr). Initial experiments, conducted at 32°C, indicated a higher reaction rate compared with the (extrapolated) nonirradiated data.

Leach tests on coated spherical particles of supercalcine were begun in late FY-1979. The particles had either a SPC-2 or a SPC-4 supercalcine core. These were tested as uncoated spheres, single-coated spheres with either Al_2O_3 or pyrolytic carbon coating, or double-coated spheres with pyrolytic carbon under Al_2O_3 . The preliminary leaching results obtained to date show that

the major elements leached from the uncoated spheres were barium, strontium, silicon and molybdenum. The concentration of these elements in the leachate for the carbon-coated particle was about an order of magnitude less than concentrations in the leachate for the uncoated particle. The particles coated with Al_2O_3 or double-coated with pyrolytic carbon and Al_2O_3 showed even lower leach rates.

The feasibility of low-temperature chemical vapor deposition (CVD) of pyrolytic carbon is being investigated at Battelle Columbus Laboratories (BCL). Two catalysts, nickel tetracarbonyl $[\text{Ni}(\text{CO})_4]$ and cobalt acetylacetonate $[\text{Co}(\text{AcAc})_2]$, are being evaluated. Coating temperatures range between 425° and 625°C. The $\text{Ni}(\text{CO})_4$ appears to increase the carbon deposition rate significantly more than does $\text{Co}(\text{AcAc})_2$.

PROCESS AND EQUIPMENT DEVELOPMENT

The goal of this task is to identify and evaluate processes for the conversion of radioactive wastes to stable, nondispersible forms other than glassy monoliths. These waste forms may include such materials as sintered ceramics, concrete, glass ceramics, cermets, metal-cast waste forms, and others. Specific process demonstrations are being conducted on coated particles and metal matrices.

A program has been started to evaluate use of a prototype remote vertical furnace to sinter waste pellets. This program is to ensure that a waste form sintering furnace is available that is applicable to remote operations and maintenance because commercial furnaces now available are applicable to neither.

Simulated waste pellets have been obtained from INEL. Preliminary tests to evaluate the compatibility of pellet and vertical furnace

components and to establish operating parameters have been completed. Three batches of about 2, 15, and 20 kg each of the simulated zirconia waste pellets were processed through the vertical sintering furnace and exposed to over 800°C for 2 hr in an atmosphere of hydrogen, argon, and nitrogen. Samples from these runs have been returned to INEL for leach tests and other evaluations.

Over 540 kg of supercalcine was shipped to Ferro-Tech, Inc., for testing and production of pellets by disc pelletizing and pin-mixing processes. Test runs there showed that a mix of the supercalcine and 5% cornstarch with a water spray provided agglomerated pellets durable enough to withstand the shipping return to PNL. Four production runs were conducted using the 5% cornstarch/supercalcine mix.

The first run used a pin mixer to agglomerate the powder to a size of ~1 mm. The second run utilized a 91-cm disc pelletizer to attempt to form a pellet size of 1 to 2 mm. This run actually achieved a size range of 1 to 10 mm, with the majority in the 4 to 6 mm size. The third run processed at an attempted size of 10 to 13 mm with the 91-cm disc pelletizer. This size was much easier to control and ranged from 6 to 25 mm, with the majority in the 10 to 13 mm size. The remainder of the material mix was processed with the pin mixer and fed into the 91-cm disc pelletizer for more pellets of 10 to 13 mm. This run provided the most consistent size product from start to finish. It also greatly reduced the dust problem encountered in the runs using only the disc pelletizer. These pellets were more uniform and formed more easily because the pin mixer provided premoistened seed material to the disc, facilitating formation of pellets. All of these agglomeration runs were air-dried at ~150°C and then shipped back to PNL for sintering and coating studies.

A pin mixer has been received for in-house production of calcine and supercalcine pellets. This pin mixer and a previously obtained disc pelletizer are to be installed at PNL for pellet production quality tests.

Comparison of the infiltration of Pb-10Sn alloy through glass marbles under vacuum and air direct casting conditions shows that in both cases all the marbles are surrounded by the alloy. However, the distance the alloy penetrates into the crevices between adjacent contacting marbles is definitely greater under vacuum pouring conditions.

It was decided to use in-can encapsulation (ICE) for the glass-marble/Pb-10Sn alloy integrated demonstration. The ICE process eliminates the need for a separate furnace to contain the molten alloy and connecting hardware between the furnace and canister as required in the direct casting process. Two pilot-scale ICE tests were conducted to evaluate the process. The first test used 8-mm lead shot and the second utilized 2-mm lead shot.

In the ICE process, the marbles are placed inside a container that has a screen on the bottom. This container is then placed inside a canister that is 5 cm larger in diameter. This assembly is placed in a furnace and preheated to 450°C. Lead particles are then fed into the canister where they melt. After the container is full of lead, it is cooled in a

specific manner. The cooling is started at the bottom while heat is applied at the top to keep the lead molten. This special procedure is necessary to prevent the formation of a shrinkage void down the center of the casting or around the marbles. Incorrect cooling can leave voids between the marbles, as evidenced during trial demonstrations. The canisters were then cross-sectioned so that the matrix could be examined. There were no voids or inclusions in the lead, and only occasional small gaps at points of contact with the marble were observed.

The drawings and write-ups for the 11 processes in the Alternative Preconceptual Design Study have been put into final format and were reviewed. Comments from this review and those received from the authors were incorporated and a final draft for internal distribution issued.

PAPERS

- J. M. Rusin, J. W. Wald, and R. O. Lokken, "Alternative Waste Forms: A Comparative Study." Presented at the November 1979 meeting of the Materials Research Society, Boston, MA.
- W. J. Gray, "Reaction of Graphite with Water and Its Implication for Radioactive Waste Storage." Submitted to Radioactive Waste Management: An International Journal.

3. NUCLEAR WASTE MATERIALS CHARACTERIZATION CENTER

R. D. Nelson - Project Manager

The objective of the Materials Characterization Center (MCC) is to ensure that approved materials data are available on waste materials. This objective will be met by 1) developing standard testing procedures and having them approved by the Materials Review Board (MRB), 2) testing waste materials by the approved procedures, and 3) publishing these procedures and MRB-approved data in a Nuclear Waste Materials Handbook.

PROGRAM MANAGEMENT

During the past quarter, the charter of the MCC was approved by the Materials Steering Committee (MSC). In addition, a draft of the program plan, including schedules for waste form and engineered barrier materials test procedures, was prepared and distributed for internal review.

Plans for next quarter include holding a workshop on the leaching of waste forms to begin peer review of the MCC leach rate test development program and to obtain responses on its adequacy. Other plans include publication of the program plan and the beginning of experimental work on test methods.

MCC CHARTER

A charter has been written to define the mission, authority, functions, interfaces, documentation, and funding of the MCC, which is an integral part of the Nuclear Waste Materials Characterization Organization sponsored by the Office of Nuclear Waste Management within the Department of Energy. To ensure that this charter was compatible with other waste programs, detailed reviews and discussions were held within PNL and

between the MCC and the Richland Operations Office, Savannah River Lead Office, Office of Nuclear Waste Isolation, the Rocky Flats TRU Waste Program Lead Office, and the MRB Chairman. The charter resulting from these interactions was presented to the MSC in November and has been approved in principle.

MCC PROGRAM PLAN

A program plan is required to provide additional details to the charter, define initial objectives and schedules, provide additional opportunity for coordinating activities with other programs, and provide continuity in the MCC program. The first draft of this plan, which outlines basic test methods for both waste forms and engineered barriers in conjunction with the MCC, has been prepared and is undergoing internal review.

Table 3.1 shows the waste form test methods for which development is planned. Engineered barrier test methods are divided into the canister and overpack methods shown in Table 3.2 and the backfill material methods shown in Table 3.3.

TABLE 3.1. Waste Form Test Methods

Testing Area	Test Method
Chemical durability	Static scoping test
	Dynamic scoping test
	Static long-term test
	Dynamic long-term test
	Autoclave test
Radiation stability	Alpha effects test
Thermal stability	Volatility
	Phase stability
	Gas generation
Mechanical properties	Impact test
	Compressive strength test
Sampling	Specimen description and analysis

TABLE 3.2. Canister and Overpack Test Methods

Testing Area	Test Method
Corrosion	Scoping - uniform corrosion
	Scoping - stress corrosion
	Scoping - nonuniform corrosion
	Long-term with simulated geology
	Radiation-enhanced corrosion
Mechanical and physical properties	Fracture toughness
	Tensile strength
	Creep rupture
Stability	Radiation effects
	Thermal effects

TABLE 3.3. Backfill Test Methods

Testing Area	Test Method
Water interaction	Water permeability
	Ion exchange capacity
Mechanical and physical properties	Plasticity
	Swelling - Shrinkage

4. TRU WASTE IMMOBILIZATION PROGRAM

C. R. Palmer - Project Manager

The objective of this program is to conduct a comparative assessment of alternative immobilization systems for selected transuranic wastes and to recommend the most promising technology for development and demonstration.

SUMMARY

A methodology based upon utility theory was selected to structure the comparison of alternative immobilization systems. The methodology incorporates utilities of system attributes that vary as the performance level varies within each attribute level. It also retains statistical information related to the uncertainties in the importance weights of the evaluation criteria and the uncertainties in performance achievements of the candidate immobilization systems.

Physical and chemical characterization of aqueous waste decontamination sludges and incinerator residues resulted in a set of interim waste characteristics. These data and qualitative characterizations will be amended when experimental support by two subcontractors is completed during the second quarter.

Conceptual screening of several waste form materials and formation processes reduced the original set of alternatives to 13 materials and 22 process unit operations.

DATA ANALYSIS - A. D. Chockie and
G. B. Mellinger

Activities in this task concentrated on two objectives during the quarter. Waste

form data base management progressed through implementation of a computer software package in which a file called GLASS has been established. Experimental data for all TRU glasses were coded for keypunching and storage in GLASS. Currently, new fields are being added to the file to include more generic data on fabrication and characterization to accommodate non-glass waste form data. Fields are also being incorporated to allow storage of experimental compositional analyses, leachate analyses, and other performance data.

The other main objective of the task is the formulation of a quantitative model to structure the comparison of the alternative immobilization systems. This has involved two activities: selection of a decision analysis methodology, including identification of an appropriate computer package to aid the quantitative comparisons, and development of an evaluation criteria hierarchy. Of these activities, only development of the hierarchy remains to be completed. Although a hierarchy has been developed, the lack of uniqueness of such a structure may necessitate further revisions based upon additional information developed during the evaluation of waste forms and processes. Activities in development of the model are discussed below.

Selecting a Decision Analysis Methodology

In examining the state of development of TRU waste immobilization systems and the diversity of options concerning the importance of system selection parameters, it became evident that classical decision analysis methods would not be appropriate. Even though modified to include multiple objectives, classical decision methodology has three basic shortcomings: it uses linear attribute weighting functions; it does not incorporate uncertainty distributions in the estimates of total value and attribute attainment; and it does not facilitate group decision making. The result would be a significant limit placed on the amount or type of information that could be considered, which could possibly lead to erroneous decisions. To avoid this loss of information, it was determined that the decision analysis methodology for the TRU program should incorporate as much distributional information as possible. Basically, the methodology should be able to accommodate:

- system objectives as a continuum of some quantitative measure
- the value dependence upon the degree to which each objective is achieved
- several decision makers' involvement in the assessment.

The objectives selected for measuring the performance of various systems are best represented by detailed quantitative and continuous measures rather than discrete or qualitative measurement units. Each objective can generally be broken down into sub-objectives that have quantitative, detailed, and continuous measures associated with them. In this manner, the performance of a system can be plotted against the probability of attaining some level of performance associated with a measurable subobjective. The

information from such probability distributions of attribute attainment for each system on each subobjective was identified as necessary to the TRU program's decision analysis methodology.

The weights of importance for each attribute will most likely differ over the range of the attribute attainment values. There are certain levels of performance or achievement for the TRU waste systems that will be considered of greater value (utility) than lesser achievement levels. This function of utility for the range of attribute attainment was also specified as a source of information that should be used for analysis of TRU systems.

The third major source of information the selected decision analysis methodology should be able to handle is the ability to assimilate a number of decision makers' inputs concerning the attribute importance weights.

As selected, the methodology will satisfy all requirements discussed above as well as provide an evaluation operation easy to understand and employ. To retain as much statistical information as possible, a computer-assisted model was adapted for the TRU program⁽¹⁾ that can carry the entire distribution of evaluation through the analysis. The methodology, shown in Figure 4.1, uses utility functions to represent the relative value of various levels of attribute performance. These functions replace the linear attribute weighting functions used in classical decision modeling and allow a theoretically more accurate decision model with greater predictive power.

The methodology also allows for explicit incorporation of the uncertainties inherent in TRU immobilization decisions. Uncertainties associated with performance estimates for each immobilization system when measured on each attribute are expressed as probability

FIGURE 4.1. Multi-Attribute Decision Analysis Methodology

density functions. Another source of uncertainty results from the group of decision makers involved in development of the weighting factors of each of the evaluation criteria. The group's values, represented as frequency histograms in this methodology, consist of two key factors: the relative importance the group assigns to each attribute, and the degree of agreement within the group concerning the weights assigned to each attribute.

The important objectives, each system's attributes, and the metrics for measuring attribute performance are identified through interviews and surveys with individuals involved with TRU waste. The input information and the associated uncertainties are assimilated and evaluated in the model to produce a preference ordering of the alternative systems. Sensitivity analysis is employed to improve understanding of the significant factors involved in the preference ordering. This decision analysis technique provides a structured and documented method for objectively comparing those TRU waste immobilization systems identified as feasible alternatives.

Development of an Objectives Hierarchy

The principal time-consuming effort in this task for the first quarter was to develop an evaluation criteria (or objectives) hierarchy. The purpose was to provide a structured method of organizing the many and varied process and product system objectives.⁽²⁾

The TRU study team initially developed a comprehensive list of all the evaluation objectives that might be of concern. This extensive list was modified several times with objectives added, deleted, combined, or revised. Eventually a consensus was reached on an initial process/product objective hier-

archy, but as mentioned previously, development of the hierarchy is evolutionary: the structure is revised as the team members assess the implications of one arrangement versus those of another. The current hierarchy of objectives is shown in Figure 4.2. Efforts are now directed at making minor revisions to the structure based on input both from within and outside the TRU program. These revisions will attempt to include all the goals of the decision makers while still producing a workable hierarchy.

Once a basic hierarchy of objectives was agreed upon, attributes were selected for each lowest-level subobjective. Each attribute is a scale for measuring the degree to which its respective subobjective is met (e.g., rem/yr for occupational dose rate). To be useful, the attribute must be:

- complete--to cover all important aspects of the subobjective
- operational--for meaningful use in the analysis
- separable--to permit simplification of aspects of the evaluation process by breaking it into parts
- nonredundant--to avoid double counting
- minimal--to make the problem dimension as small as possible.

These characteristics reduce to two requirements that were considered when developing the hierarchy and the associated attributes: 1) comprehensive (if by knowing the level of an attribute in a particular situation, one clearly understands the extent to which a process or product achieves the objective), and 2) measurable ("Can the necessary measurement be obtained?"). Eventually this objective-attribute hierarchy will be circulated to other individuals involved with TRU waste outside our project. Their input will be factored in to produce a working list of subobjectives on performance criteria.

FIGURE 4.2. Preliminary Hierarchy of Immobilization System Attributes

WASTE CHARACTERIZATION - G. H. Bryan and
C. R. Palmer

Determination of the chemical and physical properties of process sludges and incinerator residues has been the focus of this quarter's characterization activities. The reference waste for the comparative assessment is a blend of sludge and ash that has been developed using the Rocky Flats Plant (RFP) production facilities as a model. Characteristics of this waste blend continue to be developed through chemical and physical measurements being performed at PNL, RFP and the Los Alamos Scientific Laboratories (LASL). Tables 4.1 and 4.2 summarize characteristics that we are assuming for an interim period. These characteristics will be amended upon

completion of analytical programs at RFP and LASL (expected in February 1980), which will result in determination of our reference waste characteristics.

Review of the available literature regarding characteristics of incinerator residues indicates a wide and seemingly unpredictable variation in chemical composition. The composition depends chiefly on the combustible feed mixture and to a lesser extent on the incinerator design and operating conditions. To establish a better understanding of the variation to be expected in "typical" residues (and thereby determine flexibility requirements for proposed host materials), the nonvolatile inorganic components of several common combustible materials was determined. The results thus far are shown in Table 4.3.

TABLE 4.1. Interim Waste Characteristics for
the TRU Waste Immobilization Cooperative Study

	Process Sludge	Incinerator Ash	Waste Blend
Density (bulk), kg/m ³	1000	200	920
(tap), kg/m ³	1100	350	1020
Particle size distribution, wt%			
>0.8 mm	TBD	38	TBD
0.2 - 0.8 mm		20	
60 - 180 µm		18	
30 - 60 µm		9	
10 - 30 µm		8	
5 - 10 µm		7	
Average plutonium concentration, wt%	0.002	0.2	0.02
nCi/g	10 ³	10 ⁵	10 ⁴
Physical processing characteristics			
Pulverability	Floury	Dusty; perhaps rollable	Dusty
Homogeneity (size, composition)	High	Low	Moderate
Hygroscopicity	Moderate	Low	Moderate
Tramp contaminants	Insignificant	<2 wt% (all under 10 cm ³ , such as small fittings, wire, etc.)	Minor

TBD: to be determined.

TABLE 4.2. Assumed Chemical Composition for Typical Sludge and Ash

Compound	Composition, wt%		
	Process Sludge	Incinerator Ash ^(a)	Waste Blend
Al ₂ O ₃	5	20 ^(b)	7
CaO	8 ^(b)	8	8
Cr ₂ O ₃	—	2	0.2
Fe ₂ O ₃	6	8	6
K ₂ O	3	1	3
MgO	8 ^(b)	4	7
Na ₂ O	15 ^(b)	2	14
NiO	—	1	0.1
SiO ₂	40 ^(b)	35 ^(b)	40
TiO ₂	—	10 ^(b)	1
ZnO	—	2	0.2
C	—	3	0.3
Cl	0.5	0.5	0.5
H ₂ O	15	3	12
	100	100	100

a. Non-leached
b. Highly variable

TABLE 4.3. Chemical Composition of Residues from Oxidation of Combustible Waste Materials^(a)

Material	Neoprene	Hypalon®	Pb-Loaded Neoprene	Paper	Cloth Rags	Polyvinyl Chloride	Polyethylene	Wood	"Latex" Rubber
T, °C	1000	1000	750	1000	1000	600	900	1000	1000
ξ ^(b)	7.5	30	44	0.3	0.3	1.4	0	3.1	1.4
Compound	Composition, wt%								
Al ₂ O ₃	14.4	23.4	1.1	21.0	5.5	0.5		1.3	0.3
BaO				0.1	0.2	8.3			
CaO	0.6	2.3	0.3	12.6	11.5	5.7		11.4	2.5
Fe ₂ O ₃	0.6	0.3	0.1	2.4	2.9	0.2		0.8	0.2
K ₂ O	0.3	0.1		1.1	0.4	0.2		2.3	13.6
MgO	26.0	0.4	0.9	3.8	6.5	0.3		1.8	2.7
NiO	0.1			0.1	0.1				
Na ₂ O	ND ^(c)	ND	ND	ND	ND	ND		11.4	ND
PbO	0.1	0.1	77.8	0.4	0.1				
P ₂ O ₅	ND	ND	ND	ND	ND	ND		57.8	ND
SiO ₂	32.3	28.0	1.7	30.3	29.2	0.6		3.2	4.2
TiO ₂	0.7	35.7	1.5	4.0	26.6			2.4	1.7
ZnO	0.8	0.1	0.6			0.8			34.0
C						73.7		0.4	7.7
S			0.2			0.1			12.8
Other	24.1	9.6	15.8	24.2	17.1	9.7		4.3	20.3
Total	100	100	100	100	100	100		100	100

a. After material had been heated in air for ~3 hr at specified temperature

b. ξ is the weight percent of the material remaining after oxidation.

c. ND - not determined; lack of entry indicates an insignificant concentration.

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An algorithm was developed (called ASHCMP) to allow estimation, using the data from Table 4.3, of the ash composition resulting from incineration of any hypothetical combustible mixture. Ultimately parametric evaluation will be used to characterize incinerator ash expected from both plutonium recovery and volume reduction processes at facilities such as RFP. The immediate effort will be to improve the compositional information stored in ASHCMP through additional quantitative chemical analysis. Improvements will include redetermination of certain anomalous data, as well as performing analyses for other compounds and anions (e.g., halides, phosphates, chromates). We also intend to examine additional combustible materials.

Process flowsheets for the new aqueous waste decontamination facility at RFP were received, and a laboratory simulation of first-, second- and third-stage sludge was completed. This material will be used for experiments in developing nonradioactive waste forms. LASL has agreed to provide kilogram quantities of nonradioactive incinerator ash as it is available.

The plutonium-contaminated sludge and incinerator ash requested from RFP has not yet been received because of difficulties in placing the procurement and in establishing the shipping container requirements. At the end of the quarter it appeared that the contractual problems were resolved, and we are awaiting confirmation from RFP that they are ready to ship these materials. The samples will be used to verify cold waste form characterization work for several host materials.

WASTE FORM DEVELOPMENT AND CHARACTERIZATION

Glass Development - G. B. Mellinger and C. R. Palmer

Leaching measurements and elemental analyses were completed on borosilicate glass samples from the pilot-scale in-can melter process test and were compared to predictions based upon laboratory data. Except for the amorphous uppermost portion of the monolith (mentioned in an earlier progress report as resulting from inadequate oxygen availability to completely oxidize carbon), the glass composition was homogeneous and close to the design composition. While it had appeared that considerable zinc oxide depletion may have resulted from the reducing environment during melting, it was found that the actual loss was less than 5 wt%. The density ranged from 2.64 to 2.68 g/cm³ (2.68 was predicted). Leach rates were from 4×10^{-5} to 1×10^{-4} g/cm²·day using the 24-hr Soxhlet test, and from 1×10^{-4} to 2×10^{-3} g/cm²·day in the 19-hr buffered pH4 static leachant at 25°C. These rates compare with a Soxhlet prediction of about 5×10^{-5} g/cm²·day and an acidic rate of 6×10^{-4} gm/cm²·day from the laboratory experiments. The process test results including detailed product characterization data will be described in detail in a topical report to be published during the second quarter of the fiscal year.

Alternative Product Development - R. O. Lokken and G. B. Mellinger

Screening criteria in the form of process and product objectives and requirements

were formulated and issued for review and comment. These criteria will serve to reduce the set of candidate immobilization systems from about 40 discrete alternatives (including minor process variations) to a smaller set for more detailed evaluation. Subsequently as the quantitative comparison model is developed in Task 2, these criteria will be modified as necessary and the screening will be reviewed to ensure that the candidate set reflects the more objective evaluation criteria embodied in the model.

At this point the alternatives under consideration remain uncoupled according to product, process and secondary treatment categories. This approach will be followed, so long as it remains feasible, to maintain a greater degree of flexibility for the number of alternatives we evaluate. The current concepts under review fall into four material classes: crystalline ceramics, glasses, cements and organics. These classes

have been expanded in Table 4.4. The categories and their expansions will evolve as screening proceeds and as more information is developed for additional concepts. Screening of the alternatives shown will proceed through extensive reviews of the literature, experimental scoping studies of waste form fabrication and performance characteristics using nonradioactive simulations of the reference waste, and preconceptual design analyses of the several process systems.

PROCESS EVALUATIONS - C. L. Timmerman and R. L. Treat

Review of the several process alternatives that could be implemented for the reference waste has led to the creation of a series of generic unit operations (listed below) that encompass most feasible process systems. Initial process evaluations will proceed by

TABLE 4.4. Alternative Immobilization Concepts Under Review for the TRU Waste Immobilization Comparative Study

Host Materials	Process Systems	Secondary Treatments
Crystalline ceramics	Crystalline ceramic processes	Coating processes
<ul style="list-style-type: none"> • Clay additives • Tailored additives • Glass ceramics 	<ul style="list-style-type: none"> • Cold pressing • Sintering • Hot pressing • Extrusion • Pelletization 	<ul style="list-style-type: none"> • Chemical vapor deposition • Catalyzed deposition • Impregnation/polymerization
Glasses	Glass products	
<ul style="list-style-type: none"> • Low-temperature borosilicates • High-temperature aluminosilicates • Phosphates 	<ul style="list-style-type: none"> • Batch melting • Continuous electric melting • Pellet formation • Marble formation 	
Cements	Cement processes	
<ul style="list-style-type: none"> • Hydraulic cements without additives • Hydraulic cements with additives • Thermal/hydraulic castables 	<ul style="list-style-type: none"> • Mixing/casting • Billet pressing 	
Organics	Organics processes	
<ul style="list-style-type: none"> • Bitumen • Polyethylene • Urea formaldehyde • Extensible polymers 	<ul style="list-style-type: none"> • Extrusion • Mixing/casting 	

systematic, sequential analysis of discrete unit operations to determine control parameters, important sensitivities, constraints and advantages. Subsequently, appropriate unit operations will be coupled to represent integrated systems, which will then be the subject of the preconceptual design activity.

Generic unit operations for processing the reference TRU waste are as follows:

1. Drum receiving and handling
2. Milling (ball, rod milling, etc.)
3. Sorting and classifying (screening, pneumatic separation)
4. Blending
5. Material conveying
6. Resistance heating for glass melting
7. Joule-heating for glass melting
8. Glass marble formation
9. Glass pellet formation
10. Powdered waste pelletization (pin mixer, disc pelletizer)
11. Extrusion (twin screw, die)
12. Grout mixing
13. Cold pressing
14. Hot pressing
15. Vacuum casting
16. Sintering
17. Impregnating
18. Coating
19. Offgas processing
20. Recycle and secondary waste processing
21. Packaging
22. Decommissioning

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5. TRU WASTE DECONTAMINATION

R. P. Allen - Project Manager

The objective of this program is to develop electropolishing and related metal cleaning and finishing techniques into an integrated large-scale decontamination system capable of decontaminating large volumes of TRU- and other surface-contaminated solid waste.

SUMMARY

Integrated operation of the disassembly, sectioning and decontamination systems has continued. More than 200 ft² of plutonium-contaminated glove box components were decontaminated from levels much greater than 4×10^6 dis/min 100 cm² to less than TRU levels. Progress in development of vibratory finishing techniques included demonstrating the ability to decontaminate plutonium-contaminated glass and uranium-contaminated molybdenum using steel media and sodium hydroxide. An annular, high-production vibratory finisher was installed in a temporary containment enclosure, and hot testing was initiated with this machine. Tests conducted to evaluate the decontamination capabilities of Freon® (Freon 113) with additives showed that it is effective for grossly contaminated items. Progress in the sectioning studies task included installation and hot testing of a shear/punch for sectioning TRU-contaminated metals and identification of a saw blade for sectioning of TRU-contaminated Plexiglas glove box

panels. Cold testing of a centrifuge for solution processing was begun, tests were conducted on polyelectrolytes and anti-foam compounds, improvements were made in the equipment used for solidifying phosphoric acid, and new techniques were developed for handling vibratory finishing wastes.

PROCESS DEMONSTRATION

Integrated operation of the disassembly, sectioning, and decontamination systems has continued. More than 200 ft² of plutonium-contaminated glove box components from facilities undergoing decommissioning were decontaminated from much greater than 4×10^6 dis/min 100 cm² to less than TRU levels. The decontaminated material included glass (Figure 5.1), Plexiglas, plastic bags, mild steel and stainless steel. The material was disassembled using hand- and remotely-operated pneumatic tools and was sectioned using saws, nibblers, and the plasma arc torch to fit in the 4 ft³ tub-type vibratory finisher. All decontamination was performed in the vibratory finisher using steel ballcone media and sodium hydroxide compound. Typical decontamination cycles in the vibratory finisher were 1 hr.

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® Plexiglas is a registered trade name of Rohm and Haas Co., Independence Mall West, Philadelphia, PA.

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FIGURE 5.1. Plutonium-Contaminated Glass Before Decontamination in a Vibratory Finisher with Metallic Media

DECONTAMINATION STUDIES

Vibratory Finishing Technology

Development of vibratory finishing decontamination techniques continued. Tests conducted in a 4 ft³ tub-type vibratory finisher demonstrated that the use of steel media with sodium hydroxide as the cleaning compound effectively reduces contamination levels on plutonium-contaminated glass and uranium-contaminated molybdenum. The plutonium-contaminated glass came from a process tank removed from a Hanford facility undergoing decommissioning. Contamination levels were reduced from substantially greater than 10⁶ dis/min·100 cm² to 14,000 dis/min·100 cm². The uranium-contaminated molybdenum had been used in the fabrication of fuel for commercial power reactors. The contamination levels were reduced from

60,000 dis/min·100 cm² direct to 5000 dis/min·100 cm and from 6000 dis/min·100 cm² smearable to nonsmearable.

An annular high-production vibratory finisher (Figure 5.2), similar to the model intended for permanent installation in the 231-Z decontamination facility, was installed in a temporary containment enclosure. Testing was begun using both noncontaminated and contaminated material, to determine optimum operation parameters for the annular-type vibratory finisher. These preliminary test results show that the larger size and increased effectiveness of the new machine should substantially increase the amount of material that can be processed per cycle. For example, more than 400 lb of uranium-contaminated molybdenum were processed in one batch during the hot tests. In addition, the machine's automatic unloading/media separation feature will greatly decrease personnel exposure to radioactive material. No operator attention is required for unloading the

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FIGURE 5.2. Annular-Type Vibratory Finisher System for High-Production Decontamination of TRU-Contaminated Material

new annular machine whereas manual unloading of the 4-ft³ tub-type machine requires up to 15 min of constant operator presence at the machine. A portable kitchen dishwasher was successfully used to rinse and dry material processed by the annular vibratory finisher.

During the processing of plutonium-contaminated waste in the 4-ft³ vibratory finisher, the contamination levels of the metal medium (1/4-in. ballcones) began to increase rapidly. This sudden increase in contamination was traced to the processing of plutonium-contaminated aluminum using sodium hydroxide, which reacted chemically with the aluminum and formed a film of contamination on the ballcones. Moreover, inspection of the ballcones disclosed that the inaccessibility of a minute area to other ballcones was preventing removal of the contaminated film by the normal self-cleaning process. The problem was rectified by adding a commercial acidic compound that chemically removed the contaminated film. Once the contamination level of the medium stabilized, sodium hydroxide was once again used as the compound, and the contamination level of the medium remained stable. The problem may be avoided in the future by use of cone-shaped metal media which possess cleaning properties similar to the ballcone's but do not have inaccessible surfaces and should be completely self-cleaning. Also, the use of sodium hydroxide compound should be avoided in decontaminating aluminum in the vibratory finisher.

Ultrasonic Cleaning/Vapor Degreasing

Tests were conducted to evaluate the decontamination capabilities of a commercial vapor degreaser system equipped with an ultrasonic cleaning tank, a spray wand, and

a vapor-cleaning compartment. Representative plutonium-contaminated stainless steel glove box sections and comparable noncontaminated control samples were processed in the vapor degreaser system using Valclene® (Freon 113 with additives) as recommended by program consultants in Freon technology.

Three tests were conducted. The first used Freon 113 contaminated with plutonium from previous work. During this test a set of noncontaminated coupons was immersed in the ultrasonic cleaning tank. These coupons were contaminated to about 2000 dis/min·100 cm². Another set of noncontaminated coupons was spray-washed with the contaminated Freon. These coupons became contaminated to about 6000 dis/min·100 cm². Grossly plutonium-contaminated glove box sections were then immersed in the ultrasonic cleaning tank and spray-washed as before. The contamination levels of these glove box sections were reduced about 60%.

The second test was conducted in noncontaminated Freon 113. As in the first test, a set of noncontaminated coupons was immersed in the ultrasonic tank and a set was spray-washed with Freon. These coupons remained uncontaminated. Grossly plutonium-contaminated glove box sections were then similarly immersed in and spray-washed with this previously uncontaminated Freon. The contamination levels of these glove box sections were reduced about 60%.

The third test was conducted in the same Freon used in the second test. In this third test, noncontaminated coupons were immersed in the ultrasonic cleaning tank and spray-washed with the Freon. These previously noncontaminated coupons became contaminated to about 3500 dis/min·100 cm².

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These tests show that effective decontamination can be achieved using Freon 113 within an ultrasonic cleaner or as a spray for grossly contaminated items. However, Freon does recontaminate, and the achievable decontamination levels are limited by the contamination level of the Freon itself.

SECTIONING STUDIES

Progress in sectioning studies included installation and hot testing of a shear/punch for sectioning TRU-contaminated metals and identification of a saw blade for the rapid sectioning of contaminated Plexiglas glove box panels. Several abrasive saw blades also were evaluated for sectioning TRU-contaminated stainless steel.

A heavy-duty, double-cut hydraulic shear/punch system was procured (Figure 5.3) that

is capable of shearing 9-in.-wide strips of stainless steel 1/4 in. thick or mild steel 3/8 in. thick. The chief advantage of this sectioning device is that it produces no cutting waste. The kerf material is in the form of 1/4-in. strips (Figure 5.4) that can be decontaminated easily either by vibratory finishing or barrel electropolishing. This sectioning technique also has a very low probability of entraining surface contamination in the sheared edge. The only disadvantage of the shear/punch is that the current system is limited to sectioning strips 9 in. wide or less. This means that components must first be reduced to this size using the plasma arc torch or a mechanical cutting technique.

The shear portion of the shear/punch system was installed in the stainless steel facility for sectioning and pretreatment

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FIGURE 5.3. Heavy-Duty, Double-Cut Hydraulic Shear/Punch System Capable of Sectioning 9-in.-Wide Strips of TRU-Contaminated Metals

7909486-15cn

FIGURE 5.4. Mild Steel (3/8 in. thick) Sectioned with Double-Cut Shear/Punch Shown in Figure 5.3. The resulting 1/4-in.-wide V-shaped kerf is in foreground.

studies. The hydraulic unit was installed adjacent to the sectioning/pretreatment facility in a noncontaminated zone for ease of maintenance.

Hot testing of the shear/punch was started by sectioning strips of stainless steel 9 in. wide from plutonium-contaminated glove boxes. Typical cycle times for each cut were 10 sec. Assuming a desirable size of 9 in. by 6 in. for decontamination in the vibratory finisher, 3 linear ft of 9-in. wide strips, or 2.25 ft² of material, could be sectioned per minute using the shear/punch system.

A problem recently encountered in the process demonstration was the difficulty of sectioning plutonium-contaminated Plexiglas

glove box panels. Hot knives, shears, scoring, and breaking were all tried with little success. The cutting rate was slow, and the results were unpredictable. A circular saw blade specifically designed for cutting Plexiglas has now been identified and successfully used to rapidly and controllably section plutonium-contaminated Plexiglas. The unique feature of this blade is its negative rake that permits aggressive cutting action while preventing material from loading onto the teeth of the blade. Typical cutting rates for this blade are 10 to 15 ft/min of 3/8-in. Plexiglas under normal working conditions for contaminated material.

Abrasive saw blades for sectioning stainless steel also were evaluated, but with

disappointing results. Cutting rates were slow, less than 10 in./min for 3/8 in. stainless steel, and the kerf was approximately 3/16-in. wide. This means that waste generation is at least as great as with the plasma torch. Furthermore, the edges produced by abrasive cutting are extremely sharp, creating a severe safety hazard. The advantages, however, of abrasive saw blades are that they can cut through double-wall construction and are very unlikely to entrain contamination.

A diamond saw blade was also evaluated and its use found impractical. The cutting rate on 1/8-in. stainless steel was very slow, only 2 to 3 in./min, and was achieved only with unusual force on the saw.

SOLUTION PROCESSING

Work continued on developing solution processing techniques for spent electrolytes and for the liquid wastes from the vibratory finishing process. Cold testing of a centrifuge was begun, tests were conducted on polyelectrolytes and anti-foam compounds, improvements were made in the equipment used for solidifying phosphoric acid, and new techniques were developed for handling vibratory finishing wastes.

The neutralized spent phosphoric acid electrolyte, the alternative sodium nitrate electrolyte, and the liquid wastes from vibratory finishing all contain a large volume of suspended solids that must be removed during solution processing. Cold testing of a high-volume, automatic centrifuge was begun to remove the suspended solids from these liquid wastes. The complete centrifuge system consists of a control panel, centrifuge, and solid waste receptacle. A predetermined volume of the liquid waste is introduced into the centrifuge bowl, and the liquid is spun out. This leaves the solids, which are fur-

ther compacted and then automatically scraped from the bowl into a waste receptacle.

This centrifuge can effectively remove particles as small as 1 micron. Diluted, neutralized phosphoric acid and spent sodium nitrate electrolytes were both successfully processed by the centrifuge at 55 gal/50 min. Use of this type of centrifuge will allow rapid, effective, and convenient removal and disposal of solids from these liquid wastes.

The effectiveness of the centrifuge for removing solids from various liquid wastes may be increased by the use of polyelectrolytes. Moreover, in some cases, the use of polyelectrolytes alone may be sufficient to remove suspended solids from liquid wastes. Polyelectrolytes, which are polymers of high molecular weights, have a history of success as clarifying agents for water and sewage treatment.

Three polyelectrolytes, all acrylamide copolymers, were tested. Results showed that the settling rates of solids may be accelerated by the use of these polyelectrolytes for dilute, neutralized phosphoric acid electrolytes, sodium nitrate electrolytes, and liquid wastes from vibratory finishing using ceramic media (Figure 5.5). Each polyelectrolyte was effective for a specific waste; none of those tested was universally effective.

The use of some commercially prepared vibratory finishing cleaning solutions has created foaming problems when the liquid waste was processed in the evaporator. Four anti-foam compounds were identified and were tested on several cleaning solutions in an effort to eliminate this problem. The results showed that each of these anti-foam compounds was effective on certain cleaning solutions, but, as with the polyelectrolytes, no universally effective anti-foam compound was found. Until such a universal anti-foam

7912928-23cn

FIGURE 5.5. Effects of Polyelectrolyte on Settling Rate of Solids in Liquid Waste from Vibratory Finisher Decontamination System

compound is identified each new cleaning solution will have to be tested to identify an anti-foam compound prior to its use in the evaporator system.

Improvements were made in the equipment used for the solidifying and neutralizing of phosphoric acid using concentrated sodium hydroxide. The improvements consisted of installing a protective boot around the mixing apparatus and modifying the sample port, the mixing motor mount and the ventilation port (Figure 5.6). These improvements, made primarily to enhance the containment features of the apparatus, will also facilitate the process and thus reduce the amount of operator time.

The initial waste handling systems for the vibratory finishing decontamination process used a settling tank for the immediate collection of wastes from the vibratory

finisher. When ceramic media were used in the vibratory finisher, this system had the disadvantage of concentrating a large volume of radioactively contaminated sludge within the settling tank in a form extremely difficult to remove and properly package.

To facilitate the handling and processing of the ceramic-laden vibratory finishing waste, the sludge tank was replaced with an agitation tank. This consists of a small cylindrical polyethylene tank and a pump with a recirculation loop and a discharge loop. The waste is constantly agitated by pumping it through the recirculation loop, keeping the ceramic particles in suspension. When the tank nears capacity, the liquid containing the suspended solids is pumped to either a centrifuge or a settling tank where the solids are separated in such a manner as to facilitate proper packaging.

7910064-6cn

FIGURE 5.6. Improved Equipment Used for Neutralizing and Solidifying Plutonium-Contaminated Phosphoric Acid Through Controlled Addition of Concentrated Sodium Hydroxide

6. KRYPTON SOLIDIFICATION

G. L. Tingey - Project Manager

The objective of this program is to develop techniques for storing radioactive krypton in a solid matrix. Two techniques being investigated are 1) dissolution of krypton in low-density glass followed by densification at high temperatures, and 2) ion implantation of krypton in sputter-deposited metal matrices.

SUMMARY

A sputtered sample of approximate composition $\text{Ni}_{0.76}\text{Y}_{0.17}\text{Kr}_{0.07}$ has been prepared. The retention of krypton in this sample appears comparable to that of an iron-yttrium alloy, with release occurring at 725 to 900°C. A sputtered sample of iron-yttrium has been prepared containing about 9 at.% krypton and a small amount of rubidium.

ION IMPLANTATION OF KRYPTON DURING SPUTTER DEPOSITION - E. D. McClanahan and G. L. Tingey

Krypton has been successfully trapped in various sputter-deposited metals and metal alloys.⁽¹⁾ The most promising of these are two glassy alloys: $\text{Fe}_{0.79}\text{Y}_{0.12}\text{Kr}_{0.09}$ and $\text{Fe}_{0.76}\text{Zr}_{0.19}\text{Kr}_{0.05}$. Although these two alloys appear to meet the requirements for long-term storage, the potential advantages of nickel-based material for corrosion resistance justified conducting an experiment on a nickel target filled with yttrium plugs.

This experiment was designed to yield an alloy with nominal composition $\text{Ni}_{0.82}\text{Y}_{0.18}$. A system was set up in an attempt to produce a deposit 2.5 mm thick including a 0.13 mm gas-free close-out layer. Minor experimental difficulties necessitated producing the thick deposit in three sequential runs, but a de-

posit was prepared that was 2.1 mm thick, weighing 800 g and containing about 20 liters (STP) of krypton gas. During sectioning of the substrate and deposit, a deposit 0.1 mm thick, containing primarily the close-out layer, buckled and peeled from the rest of the deposit. There was also some evidence of delamination of layers throughout the deposit. Gas release measurements made on the samples were conducted; they show that the glassy deposit contained about 7 at.% krypton or 23 cm^3 of Kr (STP)/g of deposit. The density of the deposit was determined to be 7.04 g/cm^3 . The krypton was retained very effectively with observable release rates occurring only at temperatures above 550°C, and less than 1% of the krypton released below 725°C. In comparing the nickel-yttrium alloy with iron-yttrium, the following observations are made:

1. Nickel-yttrium can be loaded with only ~75% as much krypton as can iron-yttrium.
2. The sputtering rate of nickel-yttrium is ~20% greater.
3. Preliminary data indicate that the gas release temperatures are about equal for the two alloys.

EFFECT OF RUBIDIUM

The long-term stability of any waste form containing ^{85}Kr is complicated by the

increasing concentration of the decay product, ^{85}Rb . In the sputtered deposit, there is the chance that small amounts of rubidium may alter the crystallization of the glassy metal or otherwise affect the properties, which may in turn affect the release behavior. This effect can be tested by sputtering

appropriate quantities of rubidium into the structure during the krypton loading process, thus allowing one to examine the effect without long waits for ^{85}Kr decay.

Preliminary experiments with rubidium were described in a recent quarterly report.⁽²⁾

One technique used a composite target consisting of holes drilled in the iron target that were filled with a mixture of iron powder and rubidium chloride. The drawback of such an approach is that we may be adding Cl^- to the deposit instead of obtaining the pure rubidium desired. It was found, however, that the Cl^- was deposited in a

small area directly opposite the rubidium-chloride-iron powder plugs. The deposit some distance from the plugs was free of Cl^- but had substantial fractions of rubidium, and this area was to be used to examine the rubidium effect. A deposit was prepared from an existing iron-yttrium composite target modified by drilling forty-eight 3.1-mm holes around the circumference in the cylindrical target at the midline. Because of difficulty getting a good adherent deposit, the target power was reduced to 1000 volts and 1 amp, lowering the deposition rate to $5.3 \mu\text{m/hr}$. A deposit $76 \mu\text{m}$ thick has been completed and evaluation is under way. Krypton uptake was about as expected, but preliminary analysis indicates that the rubidium content is significantly lower than planned and no Cl^- was observed. Further evaluations of composition are currently under way, and tests to determine the effect of the rubidium on the krypton retention will follow.

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2. E. D. McClanahan and G. L. Tingey, "Ion Implantation of Krypton During Sputter Deposition," Nuclear Waste Management Quarterly Progress Report, July-September 1979, PNL-3000-3, Pacific Northwest Laboratory, Richland, WA, November 1979, p. 4.1.

7.0 THERMAL OUTGASSING

L. L. Burger - Project Manager

The objectives of the Thermal Outgassing Program are to:

- 1. develop a method for the safe and effective removal of volatile fission products from irradiated fuels*
- 2. evaluate the use of thermal outgassing, using various sweep gases or vacuum outgassing for the removal of T, ¹⁴C, ¹²⁹I, and ⁸⁵Kr*
- 3. apply the information obtained from both experiment and theory to spent fuel reprocessing, interim and long-term storage, and to reactor safety.*

SUMMARY

The literature survey is nearing completion, and evaluation of the references will begin during the second quarter. Negotiations were successfully concluded to make maximum use of DOE facilities and equipment presently developed by HEDL for studying reactor transients. Heating rates in the range of 300°C/sec, maximum temperatures in excess of 2000°C, and both vacuum and sweep gas modes will be available. Shippingport UO₂ PWR Core-1 blanket fuel has been transferred to DOE and is now available. The approximately 30 pins are 10-1/4 in. long and clad in Zircaloy-2.

THERMAL OUTGASSING EQUIPMENT - L. A. Bray

DOE facilities and equipment developed by HEDL⁽¹⁾ for studying reactor transients (i.e., excursions from normal) will be modified to accommodate thermal outgassing. The

equipment includes a furnace heated by a 100 kVA (maximum power), phase-fired, saturable core reactor (SCR) power supply and monitored using a precision brightness pyrometer. A fast-scanning quadrupole mass spectrometer has been incorporated into the system to analyze and obtain partial pressure of at least four different gaseous species: H₂, N₂(+CO), Kr, and Xe. In addition, the bulk of the released gases and sweep gas will be trapped for analysis by mass spectrometry and gas chromatography. The data will be recorded on a high-speed recorder to further data processing. The physical behavior of the fuel and cladding can be observed via a real-time radiography system and recorded for future analyses.

REFERENCE

1. C. A. Hinman and O. D. Slagle, Ex-Reactor Transient Fission Gas Release Studies, Fuel Pin PNL-2-4, HEDL-TME-77-83, Hanford Engineering Development Laboratory, Richland, WA, May 1978.

8. IODINE-129 FIXATION

L. L. Burger - Program Manager

The objective of this program is to develop technologies that will provide safe, effective removal and packaging of iodine-129 for initial treatment, for interim storage and shipment, and for permanent isolation. The technologies must be related to all phases of the nuclear fuel cycle.

SUMMARY

Studies involving regeneration of iodine-loaded silver-exchanged zeolites with carbon monoxide and water or ozone indicated these methods to be ineffective.

An additional leach study was made evaluating the effect of a humid CO₂ curing environment on the iodide leach resistance of Mg-I₂-impregnated cement. Curing in CO₂ increased the iodide leach resistance by a factor of 10 on the first day of leaching and by a factor of 2 after 39 leach days.

A leach study was performed to determine the effect of CO₂-saturated water on the ¹⁴C leach resistance of carbonate-impregnated cement. It suggested that the low leach resistance of the specimen in this leachate is not due to the independent action of the initial pH or the carbonate concentration of the leachate.

Preparation of iodine-loaded silver adsorbents with known, uniform levels of iodine loading continued. These materials will be used for characterization and regeneration studies.

Aqueous extraction techniques, x-ray fluorescence, and differential scanning calorimetry were used to analyze the free silver content, total silver and iodine, and the chemical form of iodine in silver-exchanged zeolites.

SILVER ZEOLITE REGENERATION - R. D. Scheele

Thermogravimetric analysis (TGA) was employed to evaluate CO + H₂O regeneration of iodine-loaded silver zeolon 900, AgZ-I. The proposed reaction, assuming iodine present as AgI, is

The free energy for this reaction is given in Table 8.1.

Weight loss studies using a gas stream of ~60% carbon monoxide and 40% water were performed on AgI and AgI-coated NaZ.⁽¹⁾ In neither case was a measurable weight loss detected before the temperature at which AgI

TABLE 8.1. Free Energy of Reaction for CO + H₂O Regeneration

<u>T, °K</u>	<u>ΔG_{RX}, kcal/mole AgI^(a)</u>
300	+14
400	+12
500	+12
600	+11
700	+10
800	+ 9

a. G values from I. Barin and O. Knacke, Thermochemical Properties of Inorganic Substances, Springer-Verlag, Berlin and New York, 1973.

begins to vaporize, 870°K. Based on this information and previous studies,⁽¹⁾ CO + H₂O does not appear to be an effective regeneration system.

In the evaluation of ozone as a regenerating agent for iodine-loaded silver zeolites, a bed of AgI-coated NaZ⁽¹⁾ was pretreated with air at the desired reaction temperature and then exposed to 0.14 vol% ozone in air. Reactions were run at 50°C intervals from 250 to 500°C. At 250°C no iodine was observed during the pretreatment period; however, thereafter most of the iodine evolved from the bed was observed during the pretreatment. Ozone was introduced only after iodine evolution was no longer detected at the desired reaction temperature. For analysis of iodine the off-gas was scrubbed with 1M NaOH and 1M KI. The scrub solutions were acidified, KI added if needed, and titrated with Na₂S₂O₃. A total of 17% of the iodine was removed from the bed after several days of pretreatment and exposure to ozone. These results indicate that use of ozone as a regenerating agent would not be practical.

LEACHING STUDIES - R. D. Scheele

Previous studies⁽¹⁾ have indicated an increase in carbon leaching from carbonate-impregnated cement and a decrease in iodine leaching from iodide- or iodate-impregnated cement with CO₂ exposure techniques. These effects were further investigated in two additional leach studies. Interpretation of results in these studies was based on the normalized cumulative fraction of species leached,

$$\frac{\sum a_n}{A_0} \frac{V}{S}, \text{ cm}$$

where a_n = iodide or ¹⁴C leached during leach period, t_n

A_0 = initial iodide or ¹⁴C present in the specimen

V = geometric volume of specimen

S = exposed geometric surface area of specimen.

In the first study, specimens of Portland Type III cement impregnated with magnesium iodide, 0.8 mmol I/g dry cement, were cured for 7 or 19 days in either humid air or humid CO₂ environment. The specimen molds were removed 6 hr after pouring to expose the total surface to curing environment.

Figure 8.1 presents the normalized, cumulative fraction of iodide leached by distilled water as a function of time. Initially the CO₂ pretreatment reduced the amount of iodide leached by a factor of ten. After 7 days leaching, this reduction factor declined to a factor of three. Increased curing time decreased the iodide leach rate slightly.

In the second leach study, specimens of Portland type III cement impregnated with ¹⁴C-traced sodium carbonate (2.5 mmol C/g dry cement) were leached at three initial conditions: HNO₃ pH 3.5; 0.03M Na₂CO₃ pH 11.5; and CO₂-saturated water pH 3.5. The first two leachate conditions were selected to simulate pH and carbonate concentration in CO₂-saturated water.

Figure 8.2 shows the effect of these three leachates on the rate at which ¹⁴C is leached. For comparison, data are included regarding a similar specimen leached in distilled water during a previous study.⁽¹⁾ Both HNO₃ pH 3.5 and 0.03M Na₂CO₃ pH 11.5 decreased the ¹⁴C leach rate slightly as compared to distilled water (Figure 8.2A). However, the ¹⁴C leach rate in CO₂-saturated water was two orders of magnitude greater than that of the specimen leached in distilled water (Figure 8.2B). These results suggest that the effect of CO₂-saturated

FIGURE 8.1. Effect of CO_2 Pretreatment on Iodide Leach Rate from Iodide-Impregnated Cement

water on ^{14}C leach stability of carbonate-impregnated cement is not caused by the independent action of low initial pH or carbonate concentration. The final pH of the pH 3.5 HNO_3 leach solution was about 10, being buffered by the cement. The final pH of the CO_2 -saturated solutions ranged from 5.5 to 8.5 for leach periods of 0.1 to 2 days. The higher acidity may account for the increased leach rate of ^{14}C in CO_2 -saturated water.

CHARACTERIZATION OF IODINE-LOADED SILVER

ADSORBENTS - R. D. Scheele and K. D. Wiemers

Characterization of iodine-loaded, silver-exchanged adsorbents involves both the preparation of iodine-loaded materials and the development of analytical procedures for determination of iodide, elemental iodine, iodate, total iodine, and total and free silver in/on these materials.

During this quarter, silver-exchanged zeolite Type III, AgX, silver-exchanged Zeolon-900,[®] AgZ, and silver-impregnated silicic acid, AC6120, were quantitatively loaded in the presence of elemental iodine at 150°C . The levels of loading ranged from 30 to 270 mgI/g adsorbent* or 14 to 127% of the theoretical loading.** During this preparation, a variation of iodine loading chemistry was observed in terms of the mass transfer zone depth and coloration: a sharp transfer zone was noted for both AgX and AC6120, the transition colors being white to magenta and white to yellow, respectively.

[®] Registered trade name of Norton Co. Chemical Products Process Div., P.O. Box 350, Akron, OH.

* Amount of iodine stabilized at 150°C . Weight of adsorbent was determined after equilibration at 150°C .

** Assuming a 1:1 combination of Ag and I.

FIGURE 8.2. Effect of CO₂-Saturated Leachate on ¹⁴C Leach Rate from Carbon-Impregnated Cement

NO₂ was observed during iodine loading of AC6I20. AgZ had a broad transfer zone, with colors of gray to yellow to purple. These materials are now being evaluated for thermal stability.

Development of analytical procedures for iodine-loaded silver adsorbents included aqueous extraction techniques, x-ray fluorescence, and differential scanning calorimetry (DSC).

Analysis of free silver in AgZ by hot extraction with sodium nitrate followed by gravimetric analysis of the silver as silver chloride gave favorable results. Selectivity for alkali ion-silver ion exchange was Na>K>>Li. Analysis of iodine by sodium thiosulfate or sodium iodide solution extraction was not successful. Decomposition of the zeolite by aqueous hydrofluoride followed by separation of silver and iodine shows some promise, but the method has not been pursued.

Total silver and iodine content of iodine-loaded AgZ was determined by x-ray fluorescence. With this technique results for iodine loading agreed within 1 wt% of values determined gravimetrically during iodine loading.

DSC was used to identify the presence of crystalline AgI by observing its characteristic energy changes: 150°C, a crystal change, and 560°C, the melting point. Figure 8.3 presents the DSC thermograms for AgI-coated NaZ (AgI-NaZ) and iodine-loaded AgZ (AgZ-I). These thermograms show the characteristic AgI changes for AgI-NaZ but not for AgZ-I. The absence of AgI characteristics in AgZ-I may be due to the dispersiveness of the AgI molecules. Normal crystalline properties, including thermal stability, may be changed.

PUBLICATIONS

The report Technical Requirements for the Control of ¹²⁹I in a Fuels Reprocessing Plant, PNL-3186, by L. L. Burger and R. E. Burns, was published in November 1979.

REFERENCE

1. A. M. Platt and J. A. Powell, compilers, Nuclear Waste Management Quarterly Progress Report for July Through September 1979, PNL-3000-3, Pacific Northwest Laboratory, Richland, WA, 1979.

FIGURE 8.3. Comparison of AgZ-I and NaZ-AgI
Differential Scanning Calimetry Thermograms

9. MONITORING AND PHYSICAL CHARACTERIZATION OF UNSATURATED ZONE TRANSPORT

S. J. Phillips - Project Manager

The objective of this project is to develop monitoring instrumentation systems and burial media-waste characterization methods to evaluate the migration of contaminants from disposal sites for low-level solid wastes.

SUMMARY

Laboratory analysis of water migration in partially saturated zone sediments has been completed. Comparative results of several methods to evaluate partially saturated hydraulic conductivity have shown that a reliable simple technique can be used.

A physical model was used to determine the effect of successive applications of water to the ground surface above a cobalt-contaminated soil layer. Leachate from this model was collected and prepared for numerical model evaluation to verify a one-dimensional solute transport model.

Destructive sampling and analysis were made with sediments contaminated by cobalt and tritium from a buried waste test facility. No significant transport of tritium was noted over a 3-month period.

LABORATORY - G. W. Gee and A. C. Campbell

Migration of radionuclides in the partially saturated zone of the ground-water system is controlled by complex physico-chemical interactions. To permit understanding and prediction of radionuclide migration, basic information on water flux as a function of the properties of the geologic media must be compiled. This

task is evaluating water and radionuclide migration on a parametric basis that supports modeling and monitoring tasks of this project.

There have been numerous attempts to obtain reliable estimates of unsaturated flow properties of soil materials. Recent work by Bresler, Russo and Miller⁽¹⁾ suggests that there may be a universal constant that, when expressed as an exponent of the water content, can yield satisfactory results in predicting the functional dependence of hydraulic conductivity on water content. The derived BRM equation is:

$$K(\theta) = K_w \theta^{7.2}$$

where $K(\theta)$ = unsaturated conductivity

K_w = saturated conductivity

$\theta = (\theta - \theta_d) / (\theta_w - \theta_d)$

θ = volumetric water content

θ_d = air dry water content at which $K(\theta)$ approaches zero

θ_w = saturated water content.

This equation was tested on several coarse soils taken from the Hanford Site. Table 9.1 shows the soil characteristics of the materials used in these tests. The BRM equation was tested against the Millington and Quirk (M-Q) method in Reisenauer⁽²⁾ as shown in Figures 9.1 and 9.2. The data appear to be

TABLE 9.1. Characteristics of Test Soil Materials

Soil	Texture			Column Density, g/cm ³	K _{sat} , cm/s	Saturation, g/g
	Sand, %	Silt, %	Clay, %			
R(A)	81.2	15	3.8	1.76	3.5×10^{-4}	0.23
R(B)	91.2	7	1.8	1.79	2.6×10^{-2}	0.17
B	91.7	7	1.3	1.82	9.0×10^{-3}	0.19
L	89.5	9	1.5	1.73	1.7×10^{-2}	0.24

FIGURE 9.1. Comparisons of Millington-Quirk Analysis with Bresler-Russo-Miller Equation

FIGURE 9.2. Comparison of Millington-Quirk Analysis and Bresler-Russo-Miller Equation Using Soil R(B) plus Comparison of Equation with Field Data from Sisson and Gibbs (1979)

in reasonable agreement over the water content range of interest (0.05 to 0.20 g/g H_2O). Data from a field study of infiltration and drainage on the R(B) soil by Sisson and Gibbs⁽³⁾ appear to agree well with the BRM method (Figure 9.2) even though no attempt was made to match the field data at the laboratory column saturation point. Twenty-eight additional laboratory tests were run on the four soils at various column packing densities, ranging from 1.6 to 1.9 g/cm³. In all cases the flow was predictable with the BRM method, with differences in flow being controlled to a large extent by sample porosity and texture. Wetting front advance into dry soil moved predictably with the square root of time in the test columns. Correlation coefficients (R^2), values between predicted and measured values, averaged more than 0.99 for the 32 tests.

This research is an extension of work reported earlier on laboratory methods of unsaturated conductivity.⁽⁴⁾ These data

suggest that the BRM method can be used confidently to predict unsaturated conductivities of sediments typical of the Hanford Site in Washington State. The advantage of this method lies in the simplicity of the data requirements. No detailed water retention characteristics are required. Either field-measured flow characteristics, i.e., field saturation (θ 's) and field-saturated conductivity (K_w), or laboratory-measured θ 's and K_w on either undisturbed cores or columns packed to field densities appear to be all that are needed to predict unsaturated conductivity within an order of magnitude (Figures 9.1-9.2). Considering the natural variability of soils, this agreement appears satisfactory, particularly for coarse materials. Flow predictions at burial sites can be aided by these simple laboratory tests of burial site materials. For more detailed site characterization, particularly for layered soil materials, field tests like those of Sisson and Gibbs⁽³⁾ may be required.

PHYSICAL MODELING - G. W. Gee and
K. O. Schwarzmiller

Physical models are used to evaluate the migration of radionuclides and water. These models are typically large sediment columns used under controlled laboratory conditions that simulate field conditions. These models are used also to evaluate the performance of monitoring transducers that will be used in the field.

A layered soil column was designed and tested to determine flow of water and radio-

nuclide under maximum drainage conditions. The material used was soil B (Table 9.1) packed to an average density of 1.75 g/cm^3 . A 12-cm layer of coarse sand ($>200 \text{ }\mu\text{m}$) was placed at 116 cm in the 176-cm column (Figures 9.3 and 9.4) to produce a layered flow system. Cobalt-60 complexed with ethylene diamine tetracetic acid (EDTA) was used as the radionuclide tracer. This radionuclide complex was chosen because of its low K_d value (<0.1) in soil B; 0.1 mCi was mixed with $4.71 K_d$ soil to produce a soil layer 1 cm thick and was placed at the 60-cm depth.

FIGURE 9.3. Large Soil Column and Associated Monitoring Equipment Used for Evaluating Radionuclide Transport

FIGURE 9.4. Cross-Section of Large Test Soil Column

A total of 29 cm of water was added at various times in increments of 5 and 2.5 cm over a period of 286 days, and the water content and gamma activity were monitored periodically. Samples were taken of the outflow, and gamma activity was determined on the outflow solution. Water content and gamma profiles are shown in Figure 9.5. Figure 9.6 shows the outflow gamma activity with time.

These data are being analyzed at present for transient flow conditions using a one-dimension solute transport model developed by Dr. P. J. Wierenga of New Mexico State University. The apparent immobility of Co-EDTA in the column (Figures 9.5 and 9.6) is somewhat surprising since previous small column studies indicated that Co-EDTA had a K_d in unsaturated sediments of less than 0.1.⁽⁵⁾ However, the data do illustrate that in unsaturated flow regimes solute travel times increase dramatically,

particularly in coarse sediments that seldom attain moisture levels greater than 30% of full saturation during any stage of infiltration or drainage.

FIELD MONITORING - S. J. Phillips, G. W. Gee and K. O. Schwarzmiller

Monitoring systems are being developed to evaluate the transport of radionuclides and water (the chief agent in the transport of radionuclides) at arid-region radioactive waste burial sites. Functional monitoring systems for shallow land burial disposal facilities are necessary to measure ambient environmental conditions and to determine when remedial actions will be necessary.

Monitoring systems and methodologies for measuring radionuclide transport and water migration in the partially saturated zone are currently being tested at a buried waste

FIGURE 9.5. Water Content and Gamma Profiles for Large Column. Numbers by curves are time in days

FIGURE 9.6. Effluent Activity as a Function of Out-flow from Large Column

test facility (BWTF). Destructive samples of sediments contaminated with tritium and cobalt have recently been taken. Laboratory analyses of tritium samples taken from the ground surface to a depth of approximately 7 m have been completed (Figure 9.7). These samples consist of sediments collected horizontally through the walls of buried caissons. The caissons containing contaminated sediments are located in the BWTF and are subjected to ambient and increased precipitation conditions.

Laboratory analyses of tritium have been conducted by two beta liquid scintillation techniques: 1) direct scintillation counting of contaminated sediments with addition of scintillation cocktail and 2) leaching of contaminated sediments with water and subsequent liquid scintillation counting. The results of these analyses have shown that either analytical technique can be used to

determine tritium concentration in soils. However, water leaching is statistically more exact. The results of laboratory counting of contaminated sediments are shown in Figure 9.8. Tritium concentration versus depth is shown above and below 60 cm, where tritium-contaminated sediments were introduced into the caisson in a layer 1 cm thick. The maximum concentration was 7×10^4 pCi g^{-1} , and the concentration is reduced vertically upward and downward. In the 3 months (October to January) between introduction of the contaminant into the caisson and sampling/counting, significant migration of tritium away from its emplacement depth has not occurred. Nor has there been a significant reduction of specific activity. This indicates that tritium migration under both ambient and increased precipitation conditions has not occurred during the initial sampling period.

80A683-1

FIGURE 9.7. Destructive Sampling of Tritium at the Buried Waste Test Facility

FIGURE 9.8. Tritium Concentration Versus Depth at Buried Waste Test Facility (October 1979)

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3. J. B. Sisson and A. G. Gibbs, Application of a Hydraulic Conductivity Scaling to Infiltration-Drainage Field Tests, pp. 1-8. Rockwell Hanford Operations report, RHO-SA-121, 1979.
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5. A. M. Platt and J. A. Powell, compilers, Nuclear Waste Management Quarterly Progress Report, April through June 1979. PNL-3000-1, Pacific Northwest Laboratory, Richland, WA, 1979.

10. WELL-LOGGING INSTRUMENTATION DEVELOPMENT

R. L. Brodzinski - Project Manager

The objective of this research program is to develop appropriate well-logging equipment capable of determining ^{90}Sr , ^3H , and transuranics at sensitivity levels which will provide useful information for the proper operation of a shallow land burial site.

SUMMARY

Preliminary results indicate that the sensitivity for measurement of ^{90}Sr by thermal neutron activation analysis will be much lower than anticipated, due apparently to erroneous literature values of the neutron capture cross section.

Experiments have been completed on the application of a phoswich type detector for the determination of ^{90}Sr from cased and uncased wells, and the results are reported.

Several designs have been considered for the tritium logging device analyzing chamber, and a survey of commercially available systems has been completed.

Data reduction and analysis of the photon detection experiments have been continued, and conceptual designs for an active neutron logging device have been considered for active determinations.

DEVELOPMENT OF LOGGING SYSTEMS CAPABLE OF MEASURING ^{90}Sr - R. L. Brodzinski, T. M. Tanner, N. A. Wogman and D. P. Brown

Thermal neutron activation analyses of ^{90}Sr experiments have been continued.

Preliminary results indicate that the sensitivity for this technique will be much lower than anticipated. This is apparently due to

erroneous values in the literature for the neutron capture cross section of ^{90}Sr .

A phoswich detector system designed primarily for measuring ^{90}Sr is shown in Figure 10.1. It consists of a primary $\text{CaF}_2(\text{Eu})$ beta detector 12.5 cm in diameter by 3.45 mm thick and covered with a 0.006 mm aluminized Mylar® window. A 3.2 mm thick quartz light pipe/absorber is sandwiched between the $\text{CaF}_2(\text{Eu})$ beta detector and a 6.4 cm thick $\text{NaI}(\text{Tl})$ gamma-ray detector. The assembly is housed in stainless steel and coupled to an RCA 8055 photomultiplier tube with an attached low-background tube base. System electronics consist of a high-voltage power supply for the photomultiplier tube, a preamplifier, a pulse shape amplifier/analyzer, and two scaler-timers. A lead shield 10 cm thick was used to eliminate most background events.

In this evaluation, one scaler recorded the pulses from the $\text{CaF}_2(\text{Eu})$ crystal while the other recorded the photons detected by the $\text{NaI}(\text{Tl})$ crystal. In normal phoswich operation, the gamma detector is operated in anticoincidence with the $\text{CaF}_2(\text{Eu})$ detector. However, in this evaluation, both beta- and

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FIGURE 10.1. A Schematic Model of a Low Background Phoswich Detector Designed to Measure ^{90}Sr in the Presence of Gamma Ray Emitting Radionuclides

gamma-rays were analyzed; thus the pulse shape analyzer was electronically optimized for this purpose. The system was adjusted by exposing it to ^{137}Cs and ^{90}Sr (^{90}Y) radiations. A time output oscilloscope was used to observe the amplitude and width of the pulse height distribution time spectrum. This spectrum contained the two time distributions for the beta particle and gamma-ray interactions in their respective detectors [beta in $\text{CaF}_2(\text{Eu})$ and gamma-ray in $\text{NaI}(\text{Tl})$]. By removing one or the other of these sources the time spectrum belonging to each was identified. Time controls were adjusted to maximize the separation of the two time amplitude spectra. Hence, the system was optimized for splitting the beta- and gamma-ray pulses which, as will be seen later, significantly reduces the overall efficiency of the beta detecting crystal.

Sources representing surface contamination were prepared by uniformly distributing known quantities of the appropriate radionuclides on a saran sheet, mounted on a plastic ring 15 cm in diameter for rigidity. Two sets of standards were prepared in this fashion; one

set contained no covering material while the second set was covered lightly with Krylon® to ensure integrity. No difference in counting characteristics was found between these two source configurations. Standards without Krylon coating were also made for the four transuranic radioisotopes ^{238}Pu , ^{239}Pu , ^{241}Am and ^{244}Cm .

Sources representing contamination uniformly distributed in sediments were also prepared for each radionuclide using sieved Hanford sand. Each isotope was spiked into 686 g of sand and was dried and mixed by quartering and coning to insure homogeneity. These sources were then poured into a plastic container 2.5 cm deep by 15 cm dia and covered with saran. Table 10.1 lists the radionuclide sources and their concentrations used in this study.

Aluminum, steel, polyvinyl chloride (PVC), fiberglass, wood, dunite sand, and Ni-CaSO_4 absorbers were prepared in a range of thicknesses varying from 10 to $10,000 \text{ mg/cm}^2$ to study the effects of various well casing and

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TABLE 10.1. Radionuclide Sources Used to Evaluate a Phoswich Detector

Source(a)	Radionuclide	Dis/min
A	^{137}Cs	1.11×10^5
B	^{90}Sr	1.30×10^5
C	^{106}Ru	1.02×10^5
D	^{90}Sr	1.30×10^5
	^{137}Cs	1.11×10^5
E	^{90}Sr	1.30×10^5
	^{106}Ru	1.02×10^5
	^{90}Sr	1.30×10^5
F	^{106}Ru	1.02×10^5
	^{137}Cs	1.11×10^5
G	^{106}Ru	1.02×10^5
	^{137}Cs	1.11×10^5
H	^{239}Pu	9.8×10^4
I	^{238}Pu	1.0×10^5
J	^{241}Am	1.32×10^5
K	^{244}Cm	9.95×10^4

a. Both massless and 2.54 cm thick sand sources 15 cm diameter were made.

grouting materials. The dunite sand and Ni-CaSO_4 absorbers were prepared in the form of plates by "casting" in plaster-of-paris such that the density of the dunite sand varied from 1.72 to 2.83 and the Ni-CaSO_4 from 2.97 to 5.08.

In normal phoswich detector operation, the secondary crystal $[\text{NaI(Tl)}$ in this system] serves as an anticoincidence shield for the primary crystal $[\text{CaF}_2(\text{Eu})$ in this system]. However, in this investigation, both crystal outputs were independently recorded such that a measure of the events in each crystal could be studied from a variety of sources. The effects of various absorber materials on the response of each crystal to specific radionuclides were also measured.

The radioisotopes ^{238}Pu , ^{239}Pu , ^{241}Am , and ^{244}Cm were investigated first to determine whether they would interfere with the detec-

tion of $^{90}\text{Sr}(^{90}\text{Y})$ and to evaluate the use of this phoswich system for the field measurement of transuranic activities. The efficiencies determined for the massless "surface contamination" standards were 0.0059%, 0.0031%, 0.0045%, and 0.0024% in the beta detector and 0.14%, 0.030%, 0.57%, and 0.15% in the gamma-ray detector for ^{238}Pu , ^{239}Pu , ^{241}Am , and ^{244}Cm , respectively. The corresponding efficiencies for $^{90}\text{Sr}(^{90}\text{Y})$ were measured to be 4.2% and 0.12%. Therefore, these transuranics will not interfere with the detection of ^{90}Sr in this phoswich system unless their concentrations are more than 1000 times higher than ^{90}Sr .

Since the transuranic isotopes would not interfere with the detection of ^{90}Sr , major emphasis was placed on the interferences created by ^{137}Cs and ^{106}Ru . As previously mentioned, the phoswich system described here

was electronically adjusted to maximize the separation between signals from the $\text{CaF}_2(\text{Eu})$ and $\text{NaI}(\text{Tl})$ crystals or, in effect, between those from beta particles and those from gamma-rays. Since $^{90}\text{Sr}(^{90}\text{Y})$ is essentially a pure beta emitter, it is not surprising that the "beta" efficiency is 34 times as high as the "gamma" efficiency. This is due primarily to the bremsstrahlung interaction of $^{90}\text{Sr}(^{90}\text{Y})$ beta particles with their environment. Conversely, ^{137}Cs , a high-yield gamma-ray emitter with only a low energy beta emission, was detected with 2.2% efficiency in the $\text{CaF}_2(\text{Eu})$ crystal and 6.4% efficiency in the $\text{NaI}(\text{Tl})$ crystal.

These efficiencies are reversed from those of ^{90}Sr and provide a separation factor of 99 for the measurement of ^{90}Sr in the presence of ^{137}Cs . Ruthenium-106 (rhodium-106), another radioisotope pair normally present in nuclear wastes, yields copious quantities of both high energy beta particles and gamma radiation and was found to be 7.9% efficient in the beta detecting crystal and 2.4% efficient in the gamma ray detecting crystal. Therefore, the separation factor for detection of ^{90}Sr in the presence of ^{106}Ru is only 11, and large quantities of ^{106}Ru could significantly interfere with the measurement of ^{90}Sr .

If the system were to be maximized for beta detection only with the $\text{NaI}(\text{Tl})$ detector in anticoincidence to the $\text{CaF}_2(\text{Eu})$ crystal, ^{90}Sr could be detected at up to 45% efficiency. Figure 10.2 shows the response of the phoswich system to a massless (surface contamination) and a 2.5 cm thick homogeneous sand (contaminated sediments) $^{90}\text{Sr}(^{90}\text{Y})$ source as a function of absorber composition and thickness. The beta curves for both ^{90}Sr sources decrease systematically with increasing absorber thickness independent of the composition. However, the gamma-ray response is widely scattered and uncorrelated. Since this result was not

anticipated, the experiments were repeated at a later date, and each individual data point was reproduced exactly as in the earlier "shotgun" pattern.

To investigate this variable but reproducible gamma-ray response to ^{90}Sr , a series of experiments was conducted on bremsstrahlung production in various absorbers. Energy spectral data were accumulated with a $\text{Ge}(\text{Li})$ spectrometer viewing a 0.4 mCi ^{90}Sr source shielded by dunite, fiberglass, aluminum, wood and PVC absorbers an average of 822 mg/cm^2 thick. A spectrum was also obtained for a slightly thicker stainless steel absorber. The purpose of these experiments was to determine the difference in the shape of the bremsstrahlung spectra due to differences in the atomic number of the absorber material. As can be seen in Figure 10.3, no significant difference in curve shape is seen. The bremsstrahlung production in these various absorber materials is nearly identical. The total number of photons above 40 keV in each of these spectra is also constant within 6%, as shown in Table 10.2. Hence, these data indicate that a uniform response should have been recorded by the $\text{NaI}(\text{Tl})$ crystal of the phoswich assembly; thus the scatter in the gamma-ray data is as yet unexplained.

Table 10.2 also contains information describing the gamma-ray detector response to $^{90}\text{Sr}(^{90}\text{Y})$ as a function of aluminum absorber thickness. The data show that the bremsstrahlung attenuation in aluminum varies by a factor of three across absorber thicknesses from 41 to $10,000 \text{ mg/cm}^2$. These data correspond to the expected attenuation factors integrated over the entire bremsstrahlung spectrum.

The beta crystal response was only four times greater than the gamma-ray crystal response for the thick $^{90}\text{Sr}(^{90}\text{Y})$ sand source, which correlates with the expected attenuation by the bulk sample.

FIGURE 10.2. Beta and Gamma Response of a Phoswich Detector to Massless and 2.5 cm Thick Sand Sources of ^{90}Sr Using Various Absorber Conditions

TABLE 10.2. Bremsstrahlung Creation as a Function of Absorber Type and Thickness from a 0.4 mCi ^{90}Sr Source

Absorber	Thickness, mg/cm ²	Relative Photons, E>40 keV
Polyvinyl- chloride	766	2.45
Dunite	785	2.39
Fiberglass	854	2.42
Aluminum	852	2.55
Wood	855	2.39
Stainless steel	1,180	2.11
Aluminum	41	3.67
Aluminum	82	3.35
Aluminum	165	3.00
Aluminum	432	2.68
Aluminum	618	2.61
Aluminum	858	2.48
Aluminum	1,681	2.36
Aluminum	3,383	1.99
Aluminum	4,241	1.83
Aluminum	5,923	1.58
Aluminum	10,290	1.12

FIGURE 10.3. $^{90}\text{Sr}(^{90}\text{Y})$ Bremsstrahlung Spectra Created by Various Materials of Constant Thickness Analyzed with a Ge(Li) Spectrometer

The beta and gamma response to the ^{137}Cs standard sources is shown in Figure 10.4. Smooth curves were obtained for both sources as a function of absorber thickness for both the beta and gamma crystal outputs. This makes the uncorrelated data obtained from the gamma detector for the ^{90}Sr source even more difficult to explain.

The response curves to the ^{106}Ru sources are shown in Figure 10.5. Again, the curves

are smooth functions of absorber thickness, and they display trends similar to those of ^{137}Cs .

Figures 10.6, 10.7, and 10.8 show the response of the phoswich system to the mixed radioisotopic sources-- ^{90}Sr plus ^{137}Cs , ^{90}Sr plus ^{106}Ru , and ^{90}Sr plus ^{106}Ru plus ^{137}Cs , respectively. The experimental data in these figures coincide precisely with the sum of the individual radioisotopic source data

FIGURE 10.4 Beta and Gamma-Ray Response of a Phoswich Detector to Massless and 2.5 cm Thick Sand Sources of ^{137}Cs Using Various Absorber Conditions

FIGURE 10.5. Beta and Gamma-Ray Response of a Phoswich Detector to Massless and 2.5 cm Thick Sand Sources of ^{106}Ru Using Various Absorber Conditions

FIGURE 10.6. Beta and Gamma-Ray Response of a Phoswich Detector to Massless and 2.5 cm Thick Sand Sources of $^{90}\text{Sr} + ^{137}\text{Cs}$ Using Various Absorber Conditions

FIGURE 10.7. Beta and Gamma-Ray Response of a Phoswich Detector to Massless and 2.5 cm Thick Sand Sources of $^{90}\text{Sr} + ^{106}\text{Ru}$ Using Various Absorber Conditions

FIGURE 10.8. Beta and Gamma-Ray Response of a Phoswich Detector to Massless and 2.5 cm Thick Sand Sources of $^{90}\text{Sr} + ^{106}\text{Ru} + ^{137}\text{Cs}$ Using Various Absorber Conditions

shown in Figures 10.2, 10.4, and 10.5. Although this was not unexpected, it was gratifying to see that the mixed radioisotope curves were not perturbed by the additional energy deposition or dead time losses in the phoswich detector system. Even the mixed gamma-ray response curves are smooth and not adversely affected by the variable ^{90}Sr component, which becomes a negligibly small contribution to the total response.

Since the data obtained from mixing radioisotopes are strictly additive, the curve shapes for complex radioisotopic mixtures can be predicted from individual radioisotopic standards. This is most easily accomplished using computer techniques. The absolute efficiencies for any absorber thickness for a given mixture of radioisotopic sources can be synthetically generated by the computer and compared to data obtained in field environments. In normal operation the quantity

of ^{90}Sr will be measured using simultaneous equations for the various species present based on these data, which indicate that as little as 9 dis/min/cm² of ^{90}Sr as surface contamination can be measured in a 1000 sec counting period in the presence of comparable concentrations of ^{137}Cs and ^{106}Ru .

However, for ^{90}Sr homogeneously mixed in sandy soils typical of the Hanford, Washington, nuclear waste areas the detection limit is 1.5 nCi/cm³ in a 1000 sec counting period if ^{137}Cs and ^{106}Ru are present at a concentration of 0.1 nCi/cm³. This can be compared to a ^{90}Sr detection limit of 0.03 nCi/cm³ under the same conditions utilizing a bremsstrahlung detection technique and an intrinsic germanium diode gamma-ray spectrometer.⁽¹⁾ Hence, although this phoswich system provides adequate sensitivity for measuring ^{90}Sr as a surface contamination, superior techniques are available for measuring ^{90}Sr interstitially mixed in sediments.

An open literature publication describing the results of these experiments entitled Evaluation of a Phoswich Detector for the In-Situ Analysis of ^{90}Sr ," by N. A. Wogman, R. L. Brodzinski, and D. P. Brown will be published in the proceedings of the Institute of Electrical and Electronics Engineers 1979 Nuclear Science Symposium in San Francisco, California, October 17-19, 1979.

DEVELOPMENT OF LOGGING SYSTEMS CAPABLE OF MEASURING TRITIUM - R. L. Brodzinski and C. Menninga

Several designs have been considered for the tritium logging device analyzing chamber, and a survey of commercially available systems has been completed. It has been determined that it is impractical to attempt to modify existing systems for application to this technology and that a unique counting

module will need to be engineered and constructed for use in the tritium logger.

DEVELOPMENT OF LOGGING SYSTEMS CAPABLE OF MEASURING ACTINIDES - H. L. Nielson and R. L. Brodzinski

Data reduction and analysis of the photon detection well-logging experiments for actinide determinations have been continued. Conceptual designs have been considered for an active neutron logging device for actinide determinations.

REFERENCE

1. R. L. Brodzinski and H. L. Nielson, "A Well Logging Technique for the In-Situ Determination of ^{90}Sr ," PNL-SA-7246, 1979. To be published in Nuclear Instruments and Methods.

11. DETECTION AND CHARACTERIZATION OF MOBILE ORGANIC COMPLEXES OF FISSION PRODUCTS

J. A. Franz - Project Manager

The objective of this project is to determine the important mobile volatile organic complexes of fission and activation products found in the storage of low-level wastes in shallow land burial facilities.

SUMMARY

During this quarter, synthetic methods have been established for preparing europium and samarium organic complexes, and techniques have been identified for their detection. Technetium (IV) desorption on Hanford soil by EDTA has been examined; EDTA greatly mobilizes Tc(IV) via complex formation.

STUDY OF MOBILITY CHARACTERISTICS OF ORGANIC COMPLEXES - L. Y. Martin and J. A. Franz

Procedures have been identified for the synthesis of europium and samarium complexes of EDTA and several other chelating complexes. Europium and samarium oxides were ordered. It was determined that plasma emission spectroscopy can analyze europium at $8.5 \times 10^{-9} \text{M}$ concentration limits and samarium at about $7 \times 10^{-8} \text{M}$ levels. This may alleviate much of the need to use radio-tracers and permit the much more rapid cold lab procedures and analyses. Thus synthetic analytical approaches appear firm for the basic soil studies in the nuclide concentration range of 10^{-4} to 10^{-8}M for europium and samarium.

The desorption of technetium as TcO_2 on Hanford soil EDTA has been examined for the concentration range 10^{-5} to 10^{-8}M . The

effect of EDTA concentration on the value of K_d was also investigated by using three EDTA concentrations (EDTA/TC = 1:1, 5:1, and 10:1) for each concentration of technetium. Water/soil ratios were 30:1; TcO_4^- was reduced by hydrazine in the presence of the soil; shaken for three days; EDTA was added and the contents were shaken for 10 days. Necessary precautions were taken to exclude air throughout the desorption studies. pH was not controlled. The mobilization of Tc(IV) by EDTA via complex formation is easily observed in the decreased values of K_d . These K_d values are presented in Table 11.1, and their graphical presentations appear in Figures 11.1-11.3. Examination of Figures 11.1-11.3 indicates that K_d would approach zero at $\leq 10^{-9} \text{M}$ concentrations. The most significant reductions in K_d as the EDTA concentration is varied occur for the higher technetium concentrations (10^{-5} , 10^{-6}M). This set of experiments will be repeated to reexamine the discrepancies associated with the 10^{-5}M concentration system; the concentration range will be expanded to include 10^{-9}M .

Technetium (IV) desorption by citrate and DTPA on Hanford soil and soil studies on Tc (EDTA and citrate) complexes will begin shortly.

Niobium and zirconium desorption studies on Hanford soil are nearing completion.

TABLE 11.1. K_d Sorption Coefficients for $^{99}\text{Tc(IV)}/\text{EDTA}$ System

Tc Concentration, moles/l	K_d				pH	
	Control No EDTA	EDTA Tc = 1	EDTA Tc = 5	EDTA Tc = 10	Before EDTA Addition	pH Final
10^{-5}	2947.3	1022.0	374.7	281.4	10.4	10.1
10^{-6}	847.2	1217.1	647.6	317.2	9.6	9.3
10^{-7}	45.7	79.0	67.0	31.9	8.7	8.3
10^{-8}	1.5	1.5	3.0	3.2	8.3	7.9

FIGURE 11.1. K_d Sorption Coefficients Versus Concentration for ^{99}Tc (EDTA/Tc=1:1)

FIGURE 11.2. K_d Sorption Coefficients Versus Concentration for ^{99}Tc (EDTA/Tc=5:1)

FIGURE 11.3. K_d Sorption Coefficients Versus Concentration for ^{99}Tc (EDTA/Tc=10:1)

12. INTERNATIONAL PROGRAM SUPPORT

K. M. Harmon - Project Manager

The objectives of this program are to provide required assistance to DOE-Nuclear Energy Waste (NEW) in coordinating and promoting the exchange of waste management technical information with other countries and in evaluating the results of international cooperative activities; assist in disseminating information derived from the international program; and prepare special studies.

SUMMARY

DOE-NEW has intensified the department's efforts to exchange technical information with other nations and to join in bilateral and multilateral cooperative test programs.

In support of this program, PNL's International Program Support Office has undertaken a coordinating role and is helping DOE-NEW assemble an integrated international program plan. Collection, review and summarizing of significant documents has continued.

13. WASTE MANAGEMENT SYSTEM STUDIES

J. V. Robinson - Project Manager

The objectives of this program are to evaluate alternative systems for treatment and disposal of radioactive wastes and to develop information required to ensure effective communication with the public.

SUMMARY

A report, Release Consequence Analysis for a Hypothetical Geologic Radioactive Waste Repository in Hard Rock, was completed and delivered to the International Atomic Energy Agency (IAEA).

A report entitled A Seismic Data Catalog and Associated Graphical Capabilities was published.

The Human Affairs Research Center (HARC) conducted a workshop at Pacific Northwest Laboratory entitled "Nuclear Waste Management, Understanding Social Assessments." Agreement was reached with the Department of Energy's Richland Operations Office (RL) that HARC's work under this program this fiscal year would consist of six discrete tasks.

A draft report, prepared by Teknekron, Inc., entitled Nuclear Waste Management Issues: A Multidisciplinary Evaluation Framework, was submitted for clearance. Another draft report, A Depth Optimization Study for Geologic Isolation of Radioactive Wastes, also prepared by Teknekron, Inc., was nearly ready for submission to DOE-RL at the end of the quarter.

The Light Water Reactor section of a Waste Management Databook was 90% complete by the end of the quarter.

INTERNATIONAL NUCLEAR FUEL CYCLE EVALUATION (INFCE)

As a result of the last meeting of WG.7 of INFCE in September 1979, the Department of Energy requested Pacific Northwest Laboratory to complete and deliver by December a final report on the granite repository consequences analysis. During November we modified the initial draft on the basis of comments received from the delegates to WG.7, including the co-author country (Sweden) and the participant country (Canada). Copies of the revised draft were delivered to Sweden and Canada.

Comments by the Swedes and the Canadians were received and incorporated into the report entitled Release Consequence Analysis for a Hypothetical Geologic Radioactive Waste Repository in Hard Rock, December 1979 (INFCE/DEP/WG.7/21). The camera-ready mats will be sent directly to the IAEA and copies of the report distributed (DOE Richland Operations Office, Headquarters, Swedes, Canadians, etc.) early in January 1980.

A SEISMIC DATA CATALOG AND ASSOCIATED GRAPHICAL CAPABILITIES - T. N. Bishop, H. P. Foote and S. C. Blair

A report entitled A Seismic Data Catalog and Associated Graphical Capabilities,

PNL-2893, incorporating the comments and suggestions of a peer review group, was cleared by DOE-RL and published.

SOCIAL PERCEPTIONS, CONFLICT, AND INSTITUTIONAL ISSUES RELATED TO NUCLEAR WASTE MANAGEMENT - S. M. Nealey (HARC)

HARC conducted a workshop at PNL entitled "Nuclear Waste Management, Understanding Social Assessments" on October 23 and 24, 1979. Twenty-seven PNL staff members and three DOE-RL representatives participated. It is planned that a shorter version will be conducted in the future for the PNL Directors' Council.

Effort on this element of the program proceeded at a minimal level pending resolution of FY-1980 tasks to be pursued. Such resolution was achieved during a meeting at DOE Richland Operations Office on December 20. As a result, six tasks as described below were approved.

Task 1. Development of a nuclear waste management social science research program plan directed at nontechnical issues in nuclear waste management.

Task 2. Survey of perceived risks and analysis of domains of social conflict associated with nuclear waste management.

Task 3. Analysis of factors contributing to the preservation of information about nuclear waste management over long time periods.

Task 4. Identification of remaining technical problems in nuclear waste management to serve as a basis for later analysis of the linkage between technical problems and nontechnical waste management issues.

Task 5. Consultation with experts on the applicability of environmental mediation techniques in the siting of nuclear waste facilities.

Task 6. Revision and issuance of topical reports completed in draft form at the end of FY-1979.

During the first quarter, work on Task 2 began. This task involves combining survey efforts that in the past have been separate. Existing questionnaire materials were reviewed with the aim to reduce respondent time required and integrate content from the several relevant content pools.

Work on Task 5 was also begun with the solicitation of reviews of the draft report on environmental mediation. S. M. Nealey attended a conference in Palo Alto on December 6-7 hosted by Resolve. The conference was concerned with developing mediation strategies for nuclear waste management decision making. We anticipate that John Busterud, Resolve's president, will be one of the consultants involved with the completion of the report to result from Task 5.

Activities in Task 6 involved solicitation of internal and external reviews of four reports completed in draft form during FY-1979. These draft reports, listed below, are undergoing technical review and will be revised during the second quarter of FY-1980.

M. K. Lindell and T. C. Earle, Public Attitudes Toward Risk Tradeoffs in Energy Policy Choices. B-HARC-411-025.

M. K. Lindell, R. W. Perry, and M. R. Greene, Attitude-Action Consistency and Social Policy Related to Nuclear Power. B-HARC-411-030.

M. K. Lindell, T. C. Earle, and R. W. Perry, Social Issues and Energy Alternatives: The Context of Conflict Over Nuclear Waste. B-HARC-411-033.

R. W. Perry, C. R. Schuller, M. K. Lindell, M. R. Greene, T. C. Earle, and J. T. Walsh, Community Stress and Social and Technological Change: A Framework for

Interpreting the Behavior of Social
Movements and Community Action Groups.

ASSESSING NUCLEAR WASTE MANAGEMENT OPTIONS* -
R. G. Bradley (Teknekron, Inc.)

A draft report entitled Nuclear Waste Management Issues: A Multidisciplinary Evaluation Framework (PNL-3078), incorporating comments by HARC, was submitted to DOE-RL for clearance.

WASTE REPOSITORY COST/SAFETY OPTIMIZATION -
M. Thadani (Teknekron, Inc.)

A draft report entitled A Depth Optimization Study for Geologic Isolation of Radioactive Wastes, PNL-3079, also prepared under contract by Teknekron, Inc., was reviewed by DOE-RL in September. At their suggestion, the report was sent to the Office of Nuclear Waste Isolation for comments. ONWI's comments were received in November. ONWI expressed concern regarding the "limiting" assumptions used by Teknekron to support the specific conclusions reached in the report. Teknekron made changes in the report that satisfied ONWI's concerns. Those revisions are being incorporated into the final report, which will be sent to DOE-RL early in January.

WASTE MANAGEMENT DATABOOK - R. W. McKee

Capabilities to perform an integrated analysis of nuclear waste management systems for once-through and reprocessing cycles were developed and applied by PNL during preparation of DOE's generic environmental impact statement on managing commercially generated radioactive wastes (CWMS).

* For background information see Quarterly Progress Report PNL-3000-3.

The CWMS integrated system model includes waste generation, waste treatment, transportation, waste storage and isolation, socio-economics and facility decommissioning subsystems. In addition to the two-volume CWMS, three reports, comprising nine volumes, provide summaries of the support information presented in the Statement.

The bases for the waste management technology and facility descriptions and environmental impacts contained in the reports were developed by experts in over 20 areas of waste management technology and environmental analysis from eleven DOE contractors and laboratories. These experts were appointed as Task Leaders and were assigned responsibility for selecting the particular alternatives to be evaluated and the reference process or concept for each alternative in their field of expertise and for developing facility descriptions for approximately 80 individual waste management alternatives. The facility descriptions, coupled with detailed waste arisings and projected power growth impact, provided the bases for estimating impacts and costs for individual as well as integrated waste management facilities.

The CWMS and its supporting reports thus contain the most current and complete data available concerning the characteristics, generation rates, and projected cumulative amounts of the various post-fission radioactive wastes from the LWR fuel cycle. As such, the CWMS is a principal reference source of consistent, reconciled input data for the various studies and systems analyses of the LWR cycle being performed by lead contractors.

To facilitate the utilization of the large amount of information on waste arisings and related data contained in the CWMS and its support documents, an effort was begun in

FY-1979 to consolidate them into a Waste Management Databook. The purpose is to develop a compendium that is readily updateable as new and/or more accurate data become available and that provides ready access to the more commonly used information.

Another sub-task begun the last quarter of FY-1979 was the development of a draft data section for the high-temperature gas reactor (HTGR) fuel cycle, similar in format and depth to the LWR fuel cycles data described above. These HTGR data may also be incorporated into the Waste Management Databook. It should be noted here that the objective is not to perform any system analysis of the fuel cycle(s) per se, but rather to characterize the waste quantities and types arising from them.

PNL is aware that Oak Ridge National Laboratory (ORNL) was recently assigned responsibility by the Waste Products Division of DOE's Office of Nuclear Waste Management (NEW/P) to coordinate the development of a Level I data base for integrated waste management system studies. The committee has

been advised of PNL's Databook project, and no duplication of effort is envisioned. Moreover, representation on the Steering Committee should facilitate the proper incorporation of the CWMS data into the ORNL work-product.

ORNL's scope includes a specific requirement to obtain and maintain a base of waste management systems cost data consistent with the cost data contained in the CWMS. PNL plans to include in the Waste Management Databook a chapter on costs that should facilitate ORNL's assignment.

About 90% of the LWR fuel cycle data were compiled, and work continued on editing the data. Review of the HTGR data prepared by North Carolina State University has been deferred, pending discussions with ORNL staff involved in the Phase I effort on Integrated Waste Management Systems Studies.

P. Craig, DOE-RL, and J. Robinson, PNL, attended the Steering Committee meeting of the ORNL Integrated Data Base project held December 6, 1979, in Rockville, Maryland.

14. WASTE MANAGEMENT SAFETY STUDIES

W. K. Winegardner - Project Manager

The objectives of this program are to aid in ensuring that waste management and its inherent safety function are given proper emphasis as various alternative fuel cycles become viable; develop assessment methods; and identify portions of waste management systems that could benefit by safety design emphasis.

SUMMARY

The two reports summarized in the previous quarterly⁽¹⁾ were revised as a result of comments obtained during publication clearance. They have been cleared and will be published as: 1) P. J. Pelto and J. W. Voss, Comparative Techniques for Nuclear Fuel Cycle Waste Management Systems, PNL-3153, and 2) P. J. Mellinger, G. R. Hoenes, L. W. Brackenbush, J. Greenborg, ⁸⁵Kr Management Trade-Offs: A Perspective to Total Radiation Dose Commitment, PNL-3176.

PNL-3153 describes a safety assessment approach for evaluating predisposal waste

management systems and applying the approach to selected facilities of two potential nuclear fuel cycles. PNL-3176 compares the radiological consequences expected from chronic release of all ⁸⁵Kr from coprocessing activities with those expected from potential krypton recovery, packaging, and storage operations.

REFERENCE

1. A. M. Platt and J. A. Powell, comps., Nuclear Waste Management Quarterly Progress Report, July through September 1979. PNL-3000-3, Pacific Northwest Laboratory, Richland, WA, February 1980.

15. ASSESSMENT OF EFFECTIVENESS OF GEOLOGIC ISOLATION SYSTEMS

A. Brandstetter - Project Manager

The objectives of the Assessment of Effectiveness of Geologic Isolation Systems (AEGIS) Program are to:

1. *develop the capabilities needed to assess the post-closure safety of geologic repositories*
2. *demonstrate the assessment capabilities by performing analysis of reference sites*
3. *apply the assessment methodology to assist the National Waste Terminal Storage Program in site selection and*
4. *perform repository site analyses responsive to the time schedule and to the level of sophistication required to meet the licensing needs of the National Waste Terminal Storage Program.*

SUMMARY

Starting with FY-1980 (October 1, 1979), the Waste Isolation Safety Assessment Program (WISAP) was terminated and divided into two separate programs: the Assessment of Effectiveness of Geologic Isolation Systems (AEGIS) Program and the Waste/Rock Interaction Technology (WRIT) Program. The AEGIS Program will continue the scenario and consequence analysis methodology development work of WISAP and will assist ONWI (Office of Nuclear Waste Isolation) and DOE in site applications. The WRIT Program will continue the sorption/desorption and waste form leach studies of WISAP, will support AEGIS in the development of geochemical reaction and waste leaching models, and will provide related data as needed for site applications.

During the first quarter of FY-1980, developmental work in AEGIS concentrated on improvements of the release scenario model, development of a coupled ground-water flow/energy transport/solute transport finite-element model, various aspects of uncertainty

and geostatistics research related to the ground-water flow and contaminant transport equations, and documentation of scenario, ground-water flow, contaminant transport, and radiation dose models. Site applications included revisions and presentations of the salt dome reference site initial analysis (RSIA) performed in FY-1979 and initiation of a basalt RSIA.

METHODOLOGY DEVELOPMENT - M. A. Harwell, J. T. Zellmer, R. W. Nelson, and J. K. Soldat

Issues Analysis

The issues analysis task began. Work included the first drafting of the aspects of incorporating organic phenomena into the AEGIS methodology. Work continued on monitoring the regulatory aspects of nuclear waste management. Work began on the white paper on problems of human intrusion with bedded salt and on the white paper concerning the use of streamtube networks for simulating multi-dimensional transport.

Scenario Methodology Development

Model Development

Development and implementation of the basalt release scenario model and its documentation continued. A draft of the user's manual was completed; it includes the logic path and assumptions used in developing the model. The manual now provides sufficient information for the non-computer-oriented geoscientist to understand the simulation procedure. The manual gives a detailed outline and explanation of the operations within each of the eleven submodels.

The scenario methodology development team considered several improvements to the model. Most important among these were: 1) modifications to the hydrologic system characterization, 2) continued development of the layered earth model (LEM), and 3) methods of reducing computer run time. Modifications to the model generally reflected increased understanding of the hydrologic and geologic systems and increased availability of data.

An editing program designed to facilitate data entry into the model is now operational. This program allows the user to develop logic flow diagrams and to enter and modify the data and parameters used in the simulation. With a visual display screen and interactive light pen, the user can easily modify the simulation run parameters to suit his particular needs or to test ideas and data. The output program is being modified and updated in response to model development and output needs. A "test bed" program designed to test and debug the individual submodels is complete and ready for preliminary testing.

All scenario submodels were reviewed internally and are being sent to AEGIS consul-

tants for further review prior to testing of the submodels with actual data. Personnel from AEGIS who are involved with release scenario development, site applications, transport methodology, geochemistry, and statistics met with consultants and U.S. Geological Survey scientists to discuss data needed for model development and define future development directions.

Statistics Package

Work on the statistical package for the model also continued. This included the direct application of statistical procedures to simple, discrete events, such as meteorite impact. The statisticians assisted the scenario staff in developing ways to organize input data gathered from multiple sources. Also, the scenario model was changed to reduce the number of dimensions in the response space and make the Monte Carlo statistical package more tractable.

Transport Methodology Development

Model Refinement

The transport methodology development staff began and is progressing well on generalizing the capability for the generation of the finite-element mesh of the CFEST (Coupled Flow, Energy, and Solute Transport) model so that axially symmetric meshes can be automatically merged into rectangular regional grids. Development of the automatic mesh generation code for the two-well cases has been completed. Input has been greatly simplified for generating both node locations and element description. A user's manual, including example runs, is being prepared.

Inverse Problem

Work continued on the sample size aspects of the inverse problem for determining hydraulic conductivity. The transformation devised during the previous quarter to obtain the geometry factors was exact only when the diagonals of the flow net element were of equal length. When the diagonals are not equal, then the transformation is only approximate. We derived an expression for the error that occurs in the geometry factors when the flow net element diagonals are unequal.

Simulation of Macroscopic Dispersion

Previous work raised very important questions on the validity and utility of the widely applied equations for convective and dispersive transport of contaminants. In particular, that work indicated that the small-scale dispersive parameters verified in laboratory studies were of only minor importance in field transport situations. Instead, natural heterogeneities in field situations cause the large observed variations in contaminant concentrations. Work this quarter has centered around the three questions relating to the following uncertainty aspects:

1. Under what probability density distributions of subsurface velocity variation can the traditional dispersive-type parameters be realistically used?
2. To what extent are the perturbation approaches, as widely used in other disciplines, applicable in subsurface hydrology?
3. If perturbation approaches should be found inappropriate for subsurface hydrologic systems by our current detailed investigation, what other approaches are available to handle the larger uncertainties?

The results derived point out that the convective-dispersion equation does not have to be introduced into descriptions of macroscopic transport because the phenomenon can be simulated by an ensemble average of strictly convective flow. A new version of the random walk method appears promising to use for direct simulation of transport. For such a method it is not necessary to introduce a dispersion parameter because a distribution of velocity variations is used directly. Work in this area appears most promising and is being continued.

Uncertainty Analysis

Progress on the stochastic-deterministic model for simulating contaminant transport continued on the two-dimensional, finite-difference solution. Specifically, numerical testing of the methods used is under way, using an analytical solution for the expectation and variance of the contaminant concentration obtained when specified for stochastic components of the velocity field and initial conditions. Debugging of the two-dimensional uncertainty transport code has progressed so that it now duplicates the results of the one-dimensional code when running a one-dimensional problem. Testing is in progress on a two-dimensional problem with a flow field containing reversals in flow direction. This permits many code paths to be exercised that are inactive for one-dimensional problems. Initial comparisons of the expectation and variance have been made between the finite difference and an analytical solution.

Geostatistics

The work with geostatistical methods to determine the variance on experimental data

entered the third phase by using potential data obtained from the Bordon field site in Canada. The Kriging proprietary computer package is being used to represent the ground-water potential, obtain the variance, and help the Canadians decide where additional locations for field measurements would be most beneficial. When the Bordon data analysis is completed, three experimental data sets will be available for use in the stochastic-deterministic transport model being developed. The results coming from experimental data sets through the use of Kriging are individual variances and covariances for the hydraulic conductivity, gradient, and effective porosity. However, the stochastic-deterministic transport model requires combining the individual variances and covariances to provide the input velocity and the variance of the velocity. To circumvent this joint distribution problem, a general perturbation theory approach was applied to provide expressions for the mean, variance, and covariance of soil properties. Specifically, the uncertainties in hydraulic conductivity and porosity are used to obtain the potentials and then to compute flow velocities.

Dose Methodology Development

Dose methodology development during FY-1980 is limited to the completion of dose model documentation. During this quarter, reports were in preparation on the ARRRG, FOOD, and PABLM computer codes, which analyze the radiological doses resulting from aquatic pathways and the ingestion of food.

SITE APPLICATIONS - F. H. Dove, C. R. Cole, J. T. Zellmer, and J. K. Soldat

Salt Dome Reference Site Initial Analysis

The salt dome reference site initial analysis performed in FY-1979, and documented in PNL-2955,⁽¹⁾ was presented to many internal reviewers, including all AEGIS staff, NWTPO and line management at PNL, and staff of DOE-RL, ONWI, DOE-RLC, and WIPP (Waste Isolation Pilot Plant). Based on feedback from these presentations, a list of assumptions was prepared, and the draft document was revised to more clearly emphasize the assumptions. Follow-up calculations were performed on the radiation dose burdens to operators of the solution mine. Preliminary results indicated that direct radiation doses to the mine workers may not be high enough to cause early detection of the salt contamination; early detection would prevent significant health effects on the population consuming the salt. The population and maximum individual doses were recalculated by using revised input data on solubility limits, salt consumption, and salt dome utilization. Another set of calculations showed the effects on doses of having an inventory of reprocessed high-level waste (HLW) rather than of spent fuel.

Basalt Reference File Initial Analysis

A preliminary schedule was established for preparing the basalt reference site initial analysis (RSIA) in FY-1980. One important consideration has been the inclusion of ONWI/Rockwell review cycles of working papers prepared at timely junctures in

the assessment. An agreement was drafted to coordinate the AEGIS basalt RSIA with the BWIP site characterization report (SCR). This will ensure that the analysis will use the best available data and staff expertise from both organizations.

Applicable data and interpretations of the regional hydrology and geology are being collected and evaluated to establish a conceptual model that is common to both the release scenario and consequence analysis. A three-level conceptual system proposed by Rockwell⁽²⁾ is being considered (initially) that groups the stratigraphy of the Columbia Plateau basalt into shallow, intermediate, and deep ground-water flow zones. Available data are most abundant at present for the shallow zone and least abundant at present for the deep zone.

The coarse-grid model of the Columbia Plateau basalt developed by the U.S. Geological Survey (USGS) was discussed with USGS staff in Tacoma, Washington. The USGS

model uses the finite-difference code developed by Trescott and others.⁽³⁾ This simulates a three-layered aquifer configuration that generally conforms to the basalt stratigraphy. The layers are not necessarily the same as those proposed by Rockwell. A potentiometric surface for the upper layer (Wanapum) has been mapped by the Washington State Department of Ecology. Because of a lack of data describing the deeper hydrologic systems, the USGS synthesizes potentiometric surfaces for the lower aquifers to be essentially similar to that of the Wanapum. Most ground water is believed to flow in the rubbly zones between massive portions of the individual basalt layers. Regional ground-water flow tends to be southwesterly across the modeled region. Recharge of the regional aquifers is predominantly in the northern, northeastern, and eastern portions of the Columbia Plateau basalt, and these aquifers eventually discharge into the Columbia and Snake Rivers.

REFERENCES

1. M. A. Harwell et al. Reference Site Initial Assessment for a Salt Dome Repository. PNL-2955, Pacific Northwest Laboratory, Richland, WA, 1979.
2. R. A. Deju and K. R. Fecht. Preliminary Description of Hydrologic Characteristics and Contaminant Transport Potential of Rocks in the Pasco Basin, South Central Washington. Information Report, RHO-BWI-LD-20. Rockwell Hanford Operations, Richland, WA, 1979.
3. P. C. Trescott et al. "Finite-Difference Model for Aquifer Simulation in Two Dimensions with Results of Numerical Experiments." U.S. Geological Survey, Techniques of Water Research Investigations of the USGS, Book 7, Chapter C1, 1976.

16. WASTE/ROCK INTERACTIONS TECHNOLOGY

R. J. Serne - Project Manager

The objectives of the Waste/Rock Interactions Technology (WRIT) Program are to:

- 1. identify and characterize mechanisms of waste form radionuclide release and subsequent geochemical interactions with engineered barriers and natural geologic media, e.g., leaching, solubility, and sorption*
- 2. collect data and develop and evaluate the effectiveness (develop a perspective on applications/limitations of each model) of models that predict leaching, sorption and release processes in the repository and surrounding media*
- 3. obtain, through verification studies and documentation, the most acceptable test methodologies for sorption, leaching, solubility limitations, and waste package-geomedia interaction studies*
- 4. support repository site characterization and licensing specifically through data generation, analysis, and predictive model formulation.*

PROGRAM MANAGEMENT - R. J. Serne

A technology program plan for WRIT was prepared and discussed with ONWI. Final scope, milestones and budgets were agreed upon and officially accepted by ONWI. As a result of the Quality Assurance (QA) audit of WISAP/WRIT performed by ONWI, a new, improved

QA plan was prepared that emphasizes proper control and documentation.

Draft FY-1979 technical annual progress reports from WISAP Task 4 (Sorption-Desorption Analysis) subcontractors were reviewed and comments made. These reports will be published in the third workshop proceedings.

WASTE PACKAGE INTERACTIONS - NUCLIDE RELEASE STUDIES - D. J. Bradley

Waste Form-Solution Interactions

To allow emphasis on new areas of interest, it was decided to do fewer solution analyses. The IAEA leach tests on waste

glass will be analyzed every four rather than every two months. Examination of our leach data to 500 days indicates that trends have stabilized and that longer times between solution analyses will not affect the data's usefulness. The schedule of solution analyses for spent fuel has been changed to permit more emphasis on a particular sample and less on triplicate analyses.

The objective of the continuous-flow leach study by Lawrence Livermore Laboratory (LLL) in cooperation with Pacific Northwest Laboratory (PNL) is to compare the leach rates of simulated high-level waste glass using two different leach methods. PNL uses a modification of the standard International Atomic Energy Agency (IAEA) leaching method while LLL uses a new single-pass system in which fresh leachant solution moves continuously over the sample.

Leach rates for doped plutonium-neptunium glass (76-68) determined by the IAEA method and by continuous-flow tests at 25°C agree closely. At 25°C, flow rates covering the

range (7 to 216 m/yr) show little effect on leach rates. However, at 75°C the effect of flow rate on leach rate is measurable. At the higher flow rate, leach rates increase by one order of magnitude.

It is thought that the oxygen content of simulated ground waters might influence the rate of dissolution of nuclear wastes incorporated into glasses. To test this theory, a study was done at Argonne National Laboratory (ANL) and comparison studies are under way at PNL, both using the same solutions, doped glass beads, and containers. At PNL two parallel sets of static waste-glass leaching measurements have been started using 76-68 glass and normal atmospheric conditions. At ANL oxygen has been excluded from the solutions and the surrounding environment. The results of ANL's anoxic leach tests at both 23° and 75°C reveal that sodium bicarbonate solutions leach all three actinides more effectively than do water or brine. There is a negative temperature effect in the leaching of plutonium by NaHCO_3 and very little temperature effect for leaching the plutonium by water or brine.

Waste Package Interactions

An autoclave experiment was conducted at PNL, designed partly to estimate the leaching to expect in future waste-package experiments and to identify any experimental design changes that might be required.

The waste package consisted of a stainless steel minicanister about 2-1/2 in. long by 1/2 in. ID filled with UO_2 -doped 76-68 glass. A pellet of glass 1/4 in. thick and surrounded by a rim from the canister tube was cut from the tube after casting. To simulate a fracture, the glass was exposed at one open end of the canister and also by cutting a vertical slit through the canister wall parallel

to the long axis. A small pellet was machined from a sample of NTS Climax Stock granite (quartz monzonite) and crushed to chips ~2 to 5 mm in diameter for use as back-fill material. The waste package was placed in a 1-gal autoclave and covered with 1275 ml of deionized water, resulting in a water-to-sample mass ratio of ~4:1. The package was run at 200°C and 1800 psi for 30 days using argon as an overpressure medium.

It appeared that a slow leak developed in the system's seal so that most of the water escaped. Only about 125 ml of water remained at the end of the 30 days, and signs of oxidation were apparent in the upper part of the package. The granite pellet was discolored at the point of contact with the glass pellet, and the latter exhibited brown discoloration, particularly in fractures and at the glass-metal interface. The remaining solution was analyzed by inductive coupled plasma spectroscopy (ICP). The final solution was significantly enriched in Na, B, Ca, K, Si, Mo, and Ni, and there was a slight enrichment in Ba, Al, Cr, Fe, Sr, and Cs. The initial pH was 5.7, and the final pH was 7.5, the direction of change expected in a silicate system.

TRU Leaching Studies

Cement TRU waste forms have been prepared at Brookhaven National Laboratory (BNL) for the determination of plutonium release rates by the static leaching procedure. Subsequent liquid scintillation counting (LSC) analysis of the leachant showed a fractional activity release of 1.6×10^{-8} for the cement specimen and 6.9×10^{-7} fractional release for the ash after 33 days. These correspond to fractional activity release rates of $4.9 \times 10^{-10} \text{ day}^{-1}$ and $2.1 \times 10^{-8} \text{ day}^{-1}$ for cement and ash, respectively.

The other TRU waste forms--urea-formaldehyde, bitumen, and water-extensible polymers--are being fabricated so that the remaining leach tests can begin.

Surface Science Studies

The Physical Electronics 550 system was received and installed at PNL in the past quarter. The computer-controlled surface analytical system presently includes auger electron spectroscopy (AES) and electron spectroscopy for chemical analysis (ESCA). Delivery of the secondary ion mass spectroscopy (SIMS) unit, which will complete the system, is expected shortly. A Kevex® Alpha-X x-ray spectrometer also has been ordered. The Kevex unit includes an alpha source (alpha particles produce ions in the target that give off x-rays upon decay) and a windowless x-ray detector as well as graphical capability and microprocessor. The windowless x-ray detector is sensitive to all elements of $Z \geq 6$. Alpha-X spectroscopy is a complementary tool to AES/ESCA/SIMS and, for certain elements, gives quantitative bulk concentrations.

The first major project to be undertaken on the 550 system is the development of a data base of solid state analyses for 76-68 glass leached under various conditions and the relation of the solid state analyses to ICP solution data. To date, analyses using ESCA have been performed on the glass beads leached in 50°C deionized water. Several important features were noted. Sodium, silicon, molybdenum, and calcium are depleted at the surface, and thus are leached from the glass at a higher rate than are

other glass constituents. In contrast, neodymium, iron, cerium, zirconium, titanium, and zinc are enriched at the surface, and thus are leached from the glass at a higher rate than are other glass constituents. Figures were constructed to demonstrate the buildup and depletion processes of selected elements as a function of leaching time. The main point to be observed is that, at 50°C in deionized water, for relatively short leaching times, buildup or depletion is reasonably linear with time. Successive experiments with higher temperatures and/or longer leaching times will determine if this trend continues.

Spent Fuel Special Studies

Autoclave leaching studies are in progress on single-crystal UO_2 with deionized water and NaHCO_3 and NaCl solutions containing 200 ppm oxygen. The leaching rate trend versus water type at 75°C for UO_2 is $\text{NaHCO}_3 > \text{H}_2\text{O} = \text{NaCl}$. At 150°C, the leaching rate versus water type is $\text{NaHCO}_3 > \text{H}_2\text{O} > \text{NaCl}$. Note that the rates at both temperatures in NaHCO_3 are essentially the same.

Electrochemical oxidation and leaching of single crystals of UO_2 in H_2O , NaHCO_3 and NaCl solutions are in progress. So far the highest leaching rate was found for the NaHCO_3 solution, which is consistent with the above autoclave experiment results. The surfaces of the UO_2 samples are being analyzed for comparison with those surfaces resulting from the autoclave and static leaching experiments. We plan next to leach UO_2 pellets (crushed crystalline UO_2 hot-pressed together) as these will be more applicable to actual fuel pellets.

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GEOLOGIC MEDIA - NUCLIDE INTERACTION STUDIES - J. F. Relyea

Technical Coordination

During the first quarter of FY-1980 trips were made to Lawrence Berkeley Laboratory, Lawrence Livermore Laboratory, Los Alamos Scientific Laboratories, Argonne National Laboratory, and Oak Ridge National Laboratory (LBL, LLL, LASL, ANL, ORNL), where work packages were discussed for each subcontractor. Work plans for the Georgia Institute of Technology and Rockwell Hanford Operations (GIT, RHQ) were discussed by phone. Work packages for GIT, LASL, LLL, and LBL have been prepared and await appropriate signatures. The work plan for PNL internal WRIT Task 3 investigations has been incorporated into the technology program plan (TPP) for submission to ONWI.

The third WISAP Task 4 contractor information meeting (FY-1979 predecessor of WRIT Tasks 3, 4, and 5) was held in Seattle from October 14 to October 17. Twenty-nine presentations were made. Eighty-three people attended, representing nine national laboratories, three universities, four government agencies, four private companies, and three foreign countries. Discussion tapes from the meeting have been transcribed and are being edited to include with annual reports from each subcontractor in the proceedings.

Evaluation of Experimental Methodology

Earlier investigations conducted at ANL have shown that cesium sorption on Sentinel Gap basalt decreases with increased cesium solution concentrations but increases with time from 15 to 28 days. Similar effects have now been observed with cesium on oolitic limestone. The observed K_d (Cs) values on

limestone decrease from about 100 ml/g with $[Cs] \approx 10^{-8} M$ to about 10 ml/g at $[Cs] \approx 10^{-7} M$. The decrease in K_d (Cs) to 100 ml/g occurs at a lower cesium concentration for sorption on the limestone than on basalt ($K_d [Cs] \approx 500$ ml/g on basalt at $[Cs] \approx 3 \times 10^{-11} M$ and $K_d [Cs] \approx 165$ ml/g at $[Cs] \approx 7 \times 10^{-7} M$), and is much more abrupt. Sorption K_d values for strontium (from $10^{-11} M$ to $10^{-6} M$ on oolitic limestone) were not dependent on time or tracer strontium concentrations. The behavior of strontium may be caused by excess Ca^{+2} ($1.2 \times 10^{-3} M$) in the synthetic ground water. The dependence of K_d values on tracer concentrations will require that transport models either use a K_d that varies with tracer concentration or else take the conservative view and use a minimum K_d value. The latter choice may be desirable for a PIR (preliminary information report)-type computer run but not for licensing. This is because "realistic" K_d values for "realistic" nuclide concentrations may be much higher than the so-called conservative case.

Column experiments using crushed rock (Climax Stock granite and argillite) continued at LASL for comparison of batch and flow-through column measurements of K_d . Flow rates (pore velocities) in the columns were 48 m/yr (0.54 cm/hr) in argillite and 32 m/yr (0.36 cm/hr) in granite. Particle sizes used were 180 to 250 μm for the argillite and 35 to 106 μm for the granite. All four columns were loaded with a spike of tracer, using upward flow controlled by a syringe pump.

LASL researchers have observed that the tendency of column sorption ratios (for materials with low K_d values) to be smaller than batch values could be due partly to the shorter residence times. Perhaps in batch experiments lasting weeks or months, diffusion into the rock particles contributes

significantly to sorption in cases of low K_d values. Effort is being made to reconcile the differences between batch and column methods for measuring K_d values. This reconciliation is needed to obtain confidence in measured values; to permit comparison of the K_d s measured by the different methods; and to understand the mechanisms responsible before applying laboratory results to in situ experiments.

Data from ORNL show that sorption of divalent cations is favored over monovalent cations with an increase in temperature. Sorption of Sr^{+2} on a Na^+ -saturated illite increases with temperature while Cs^+ sorption on a Ca^{+2} montmorillonite decreases with temperature. Sorption of Sr^{+2} on a Ca^{+2} -saturated montmorillonite is essentially unaffected by temperature. This implies that sorption of the highly charged cations of americium and plutonium should increase with temperature. It implies also that sorption of monovalent radionuclides in calcium-dominated ground waters should decrease with increasing temperature.

Sorption of neptunium under reduced oxygen levels was studied at PNL. Comparison with previous PNL data shows that $K_d(\text{Np})$ apparently increases in an anoxic environment; however, the neptunium concentration in the influent solution varies by about five orders of magnitude in the two experiments ($\sim 10^{-11}\text{M}$ in the anoxic experiment and $\sim 10^{-6}\text{M}$ in the ambient atmosphere experiment). Data show that $K_d(\text{Np})$ is generally higher in a nearly saturated NaCl brine (5.13N NaCl), which is contrary to the normal ion-exchange behavior of a cation. Unlike plutonium, there was no significant sorption of neptunium on blank or sample tube walls (<2%).

Neptunium and technetium appear to have higher K_d values in brine solutions than expected from ion-exchange theory. The mecha-

nism behind this behavior is presently unknown but might be a bonus in a salt environment. Investigations in WRIT Task 4 may provide insight as to the mechanism of this behavior and allow generalization to other radionuclides.

Mechanism Studies

LLL investigators have completed a batch K_d study of 15 sandstones with the sorption of ^{137}Cs , ^{141}Ce , ^{125}Sb , ^{75}Se , ^{85}Sr , and ^{237}Pu from brine. Observed K_d values for strontium varied from -0.39 ml/g to 0.01 ml/g, indicating that the sandstones do not adsorb strontium from brine. Values for cesium, antimony, and selenium were 0 to 8 ml/g, 0 to 14 ml/g, and 0 to 63 ml/g, respectively. The cesium and plutonium K_d values were 20 to 1800 ml/g and 60 to 9000 ml/g, respectively.

The weathering of basalt, granite, and argillite and its effects on nuclide sorption are being studied at RHO. Investigators at RHO have weathered these three rocks in a Soxhlet extractor (refluxing hot distilled water to leach samples) using either air or a nitrogen atmosphere. Generally weathering had little effect on K_d values for granite. Weathering of argillite in a nitrogen atmosphere seemed to reduce sorption of all nuclides except cesium. Weathered basalt (both in air and nitrogen) gave higher K_d values than those observed on fresh basalt, especially for plutonium and americium. Only with argillite does weathering appear to adversely affect sorption for those nuclides studied.

Studies at PNL have shown that more than one mechanism may be responsible for the adsorption of ^{239}Pu . The iron content of minerals studied correlated with the observed disappearance of plutonium from solution. Analyses of plutonium oxidation states showed

that 90% of the plutonium in solution was an oxidized form [Pu(V) or Pu(VI)]. When washed with a 1.0N NaCl solution, only 5 to 32% of the adsorbed plutonium could be removed. Batch adsorption tests with plutonium in air and in ozone have shown reduced sorption of plutonium in the oxidizing ozone atmosphere. These results indicate that only a small fraction of plutonium is adsorbed by ion exchange and that ferrous iron in the minerals may reduce Pu(VI) to a strongly adsorbed form that is not easily exchanged.

NUCLIDE SOLUTION CHEMISTRY - D. Rai

Technical Coordination

To start the solution chemistry task, various national laboratories and a few other funding agencies were contacted to identify ongoing research and possible subcontractors.

Plutonium and Neptunium Species in Oxic Environments

Rai et al. (1978) showed that solutions at pH 3.5 to 8 in equilibrium with $\text{Pu}(\text{OH})_4$ and PuO_2 contained Pu(V).⁽¹⁾ The concentration and the nature of the plutonium species in equilibrium with PuO_2 and $\text{Pu}(\text{OH})_4$ have been periodically monitored. For over 600 days the pH of the solutions was observed to decrease with time (presumably due to radiolysis of water). However, the concentrations of plutonium in solution versus pH followed the empirical PuO_2 and $\text{Pu}(\text{OH})_4$ solubility lines. The pH appears to affect the nature of species in solution. At a pH of <3 all the solutions contacting $\text{Pu}(\text{OH})_4$ show that the oxidation state of plutonium in solution is [Pu(III) or Pu(V)] as compared to solutions of pH values >3, which show plutonyl [Pu(V and VI)] to be the dominant species.

Other studies done at PNL on leaching of plutonium/neptunium-doped glass beads show the predominant neptunium species in solution to be $\text{Np}(\text{V and/or VI})$ and the predominant plutonium species $\text{Pu}(\text{V and/or VI})$ (except in the case of NaHCO_3 leach solutions). Sodium bicarbonate solutions contain only about 30% Pu(V and/or VI). Why oxidation states of plutonium in NaHCO_3 water differ from those of all other solutions is not known, but since NaHCO_3 leach solutions have high pH values (9.3) compared with the others (~7 pH) the differences may relate to pH. Further studies are being planned to investigate these differences in oxidation states.

Effect of Fe^{2+} , pH, and Eh on Technetium Behavior

Francis and Bondietti (ORNL), while studying the sorption of TcO_4^- by geologic materials, observed that the loss of TcO_4^- from solution appears to be due to the reduced oxidation state. Their results showed that Eh was the most important variable controlling the technetium concentration. Losses of TcO_4^- from basalt suspensions were observed at ~0.240 volts higher than those predicted from the thermodynamic data. The TcO_4^- was observed to be stable in the presence of 100 μM Fe^{2+} , pH 5, and Eh 0.4 volts. As the pH was increased to 6.5 and greater, the loss of TcO_4^- from solution was directly proportional to the loss of Fe^{2+} . Data indicated that TcO_4^- was reduced by Fe^{2+} as the pH was increased and Tc^{+4} was isomorphically substituted into the lattice of $\text{Fe}(\text{OH})_3$. The ability of Fe(II)-rich rocks to reduce trace amounts of TcO_4^- , NpO_2^+ , UO_2^{2+} , PuO_2^+ and PuO_2^{2+} is under investigation. The results with technetium suggest that certain rocks and possibly certain reducing ground waters may

significantly increase the adsorption of elements such as technetium, neptunium, uranium and plutonium as compared with adsorption under oxidizing conditions.

PREDICTIVE MODEL DEVELOPMENT - W. L. Kuhn

For this task, begun this quarter, definition was the major activity. The scope will ultimately include accumulation and management of data developed by WRIT, reduction of data to empirical models, and development of predictive mechanistic models of radionuclide release from a repository to the "far field," the "source term" for geotransport calculations. These activities have been separated from the experimental tasks where the responsibilities would otherwise lie, and are collected here to provide a central contact with those using information developed by WRIT. For FY-1980 the major contact is expected to be with the AEGIS program (Assessment of the Effectiveness of Geologic Isolation Systems).

Data Banks and Data Reduction

Transcription of leaching data to computer storage was begun. Development of a software package specific to the format of WRIT Task 2 leach data was begun to establish a preliminary data bank on leaching. When complete, the software will provide for tabulation, plotting, and certain simple regression sub-routines such as

$$(\text{fraction leached}) = at^{1/2} + bt,$$

where t is time and a and b depend on radionuclide and experiment. This functional form has no real mechanistic basis but is a convenient form for preliminary data reduction.

Geotransport Sorption Kinetics

Geotransport codes, for example, as used by AEGIS, assume sorption equilibrium between ground water and geomedium. It has been suggested recently (Levine 1979) that sorption kinetics may be interesting to consider as well.⁽²⁾ To determine when this might be true, a simple study was done based on the "Thomas" solution (Sherwood et al. 1975) for breakthrough chromatography.⁽³⁾ The problem studied was a step change in radionuclide concentration at the inlet to a sorbing column, where sorption and desorption occur at finite rates. Decay and dispersion were ignored for simplicity.

The results show that the dimensionless group

$$\frac{(\text{sorption rate constant})(\text{groundwater travel time})}{(\text{sorbent porosity})}$$

must be ≈ 1000 for kinetics to be important. Sorption this slow would not be evident during ordinary laboratory experiments, so that laboratory sorption coefficients can be taken to be defensibly conservative.

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D. J. Bradley, C. O. Harvey and R. P. Turcotte, Leaching of Actinides and Technetium from Simulated High-Level Waste Glass. PNL-3152, Pacific Northwest Laboratory, Richland, WA, 1979.

D. J. Bradley, Chmn., Proceedings of the Task 2 Workshop Waste Isolation Safety Assessment Program. PNL-SA-7992, Pacific Northwest Laboratory, Richland, WA, 1979.

PLANS FOR NEXT QUARTER

Program Management

Next quarter long-range FY-1981-1986 plans will be prepared in DOE format to facilitate long-range budget requests. All WRIT FY-1980 subcontracts will be submitted for DOE-RL processing. Preparation and execution of a new QA plan will continue.

Waste Package Interactions - Nuclide Release Studies

IAEA, static and autoclave leaching experiments on spent fuel, doped glass, and TRU waste forms will continue. Waste package system nuclide release studies will be given increased emphasis. Solid state and surface analyses of waste forms, host rock and canister material from leaching and systems nuclide release studies will be performed. Special studies on UO_2 crystals and pellets will continue to aid in the interpretation of spent fuel leaching results. WRIT leaching methods will be documented in a draft topical report.

Geologic Media-Nuclide Interactions Studies

A critical review of current adsorption test methods will be prepared as a topical report. Methodology studies will continue

on generic rock-ground-water systems to provide objective data to facilitate selection of candidate standard sorption tests. Mechanism studies on generic rock, minerals, and ground-water systems will be continued to elucidate the physicochemical processes controlling nuclide sorption-desorption. Experiments on intact cores and fractured cores will be given increased emphasis.

Nuclide Solution Chemistry Studies

Work will continue to identify probable subcontractors and to delineate specific short- and long-term task goals and work packages. Measurements of solution oxidation states of actinides and of the solubility of actinide hydroxides and oxides will continue.

Predictive Model Development

Activities planned next quarter include final debugging of the software on the sorption data information retrieval system; installation of sorption and leaching data into computer memory; documentation of the status of adaptive learning network empirical sorption models; and preparation of a work statement for a subcontract to review existing codes and to modify for WRIT needs a chemical equilibrium thermodynamic computer code, to be selected.

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17. SPENT FUEL AND FUEL POOL INTEGRITY PROGRAM

A. B. Johnson, Jr. - Project Manager

The objectives of this program are to develop and conduct a surveillance program for nuclear fuel stored in spent fuel pools to determine whether degradation of cladding or fixtures is occurring; also to examine selected spent fuel pool components for evidence of degradation; monitor similar studies going on in other countries; and cooperate in international information exchanges.

SPENT FUEL ACQUISITIONS

The world's oldest Zircaloy-clad fuel was discharged from the Shippingport reactor in 1959. Since examination the fuel has been stored at the Expended Core Facility (ECF) at Idaho Falls. The custodian of the fuel, the Naval Reactors Branch, DOE, has agreed to transfer fifteen rods from the bundle (0551) to the Spent Fuel and Fuel Pool Component Integrity program for examination after 20 years in storage. Also to be transferred is a complete bundle (074) discharged in 1963. Characteristics of the assemblies appear in Table 14.1.

Rods from Bundle 0551 were examined soon after discharge from the reactor. Mr. Richard Atherton of Bettis Atomic Power Laboratories is helping identify the documentation related to the Shippingport fuel

fabrication, reactor exposure, and post-irradiation examinations.

Plans are being developed to examine selected rods from bundles 0551 and 074. The bundles will then be placed in storage, with periodic surveillance and other hot cell examination after 5 years.

POOL COMPONENT EXAMINATIONS AND MECHANISM STUDIES

A section of spent fuel pool piping was received from the Three Mile Island No. 1 reactor. The pipe had a through-wall crack which propagated by an intergranular mode. Initial investigations have been summarized.

Slow strain rate testing is under way to further investigate the cracking mechanism.

TABLE 17.1. Characteristics of Shippingport Fuel
To Be Examined

Item (a)	Operation (b)	Depletion, MWd/MT	Years of Pool Storage at Shippingport and ECF (c)
15 fuel rods from Blanket Bundle 0551	5,806 EFPH in PWR Core 1 Seed 1	About 4,000 average	19
Blanket Bundle 074 (previously designated backup for MELBA)	27,780 EFPH in PWR Core 1 Seeds 1, 2, 3, 4	18,000 average	15

- a. The blanket bundles consist of 120 fuel rods on an 11 by 11 square array. The rods are 10-1/4 in. long and have sealed end caps at both ends, which are welded together to form tube sheets at the ends of the bundles. Seven bundles are stacked axially to form a fuel assembly in the reactor. The rods have Zircaloy-2 tubing with an OD of 0.411 in. nominal and contain annealed about 25 fuel pellets. The fuel pellets are right circular cylinders of sintered UO_2 .
- b. Average rod surface temperature during PWR core 1 operation was about 490°F to 545°F.
- c. Water pit temperature at Shippingport and ECF before 1973 was 60 to 80°F. Since 1973 the water pit temperature at ECF has been 45 to 60°F.

18. SYSTEMS STUDY ON ENGINEERED BARRIERS

M. O. Cloninger - Project Manager

The objectives of the Systems Study on Engineered Barriers Program are:

1. *to roughly determine what incentives exist for providing highly durable and effective engineered barriers for the isolation of radioactive waste in a deep geologic repository, based on what effect, if any, the engineered barrier may have on the ultimate potential dose to humans, and indicate the degree of effectiveness that may be required to provide maximum benefit*
2. *based on present barriers concepts and knowledge, to determine whether current technology and barrier concepts meet the requirements implied by the successful completion of the first objective and perform a cost/benefit analysis for those concepts that show promise.*

SUMMARY

The analysis of engineered barrier incentives for the reference salt site was essentially completed and verifies the initial study conclusions. An evaluation of flow-field dispersion effects was started to determine how such effects may alter the conclusions about waste package release rates. A task was begun at Science Applications, Inc. (SAI), to determine potential barriers performance based on currently available data and theory. SAI has prepared a draft interim report on their first-generation model analysis, to be completed and issued next quarter.

CONSEQUENCE ANALYSIS

The release consequence calculations for the reference salt site case analysis were essentially completed. The results tend to verify the initial conclusions from the FY-1979 work.⁽¹⁾

Several simplifying assumptions were made in order to perform the analysis. The conclusions are based on the validity of the

assumptions and are limited to the reference salt site repository system and any real systems whose properties are within or near the range of the reference site parameters and constants.

Assumptions

1. The radionuclide source is one-fifth of the U.S. nuclear power economy spent fuel accumulated through the year 2000 as defined in CWM/GEIS.⁽²⁾
2. The release pathways with the greatest risk to man originate from the waste after ultimate disposal.
3. A nonsalt geologic medium similar in sorption characteristics to WIPP (Waste Isolation Pilot Plant) site Magenta/Culebra dolomite characterizes the geology beyond the repository itself. The ground water leaving the repository is the same as WIPP Brine B.
4. The highest risk pathway from the site to the biosphere is the leach incident/ground-water transport pathway.
5. All nuclides are soluble in ground water, and the dissolved nuclides do not change speciation during migration.

6. The nuclides are dissolved in the ground water in trace concentrations.
7. The migrating nuclides are in sorption equilibrium at all points.
8. Estimates of the sorption equilibrium constants are used in most cases.
9. Transverse convection and dispersion are neglected.
10. The water velocity and the axial dispersion coefficients are constant for any single parametric calculation.
11. The release of nuclides from the repository confines is a constant rate band (fraction per year) term, except for the effect of radioactive decay (decaying band release).
12. Doses are calculated for a "maximum" individual.
13. For all dose calculations, the discharging nuclides accumulate in the biosphere for 70 years; the maximum individual is exposed to that accumulation during those 70 years, and the nuclides are subsequently dispersed into the biosphere with no dose after the exposure period.
14. Man's dietary and living habits will remain constant throughout geologic time.
15. If the maximum individual is adequately protected, all individuals are adequately protected (this tends to ignore any "statistical" consequences due to integrated population doses based on a linear consequences approach).
16. A millirem dose received by a person living in the year 2051 is of equivalent value in all respects to a millirem dose received by a person living in the year 1,002,051 (10^6 years later). There is no value discounting with time of dose occurrence.

Conclusions

Even in view of the potentially large uncertainty band inherent in this type of analysis, these results indicate apparent trends that do not appear to be overly sensitive to a reasonable variation in geohydrologic parameters:

1. Increasing geologic isolation (longer path length or slower ground-water velocity between the repository site and man's environment*) decreases the dose from single nuclides and generally decreases the dose from nuclides in actinide decay chains.
2. An increase in the effectiveness of the barrier system to limit nuclide release rates into the ground water directly decreases the dose from single nuclides. But increased effectiveness does not decrease the dose from nuclides created by decay after their precursors have left the site unless the release rate becomes so very small that the spread of the nuclides in time and space due to lower release rates is greater than that due to the different sorption characteristics of the decay chain members. A product of one of these actinide decay chains, ^{226}Ra , generally dominates the potential dose from the repository; however, even for a geologic site with fairly good geohydrologic characteristics

* Greater sorption capacity of the surrounding geologic media also increases isolation and is perhaps the most important factor in isolation. Sorption capacity effect was not investigated as a parameter for the reference salt site analysis, but it will be analyzed to a limited degree as other reference sites evaluations (granite, basalt and shale) are completed.

(low potential for ground-water transport, good sorption, long pathways to the biosphere), there is an apparent ALARA incentive to provide a low-leach waste form or other engineered system that will limit the release rate of radionuclides into the ground water in the case of the general leach incident scenario. There is a stronger incentive to provide an effective release rate limiting barrier if an intrusion by water well penetration very near the repository is a reasonably probable event.

3. Increasing container longevity (up to about 10,000 years or so) does not reduce the potential dose by delaying the leach initiation for the general leach incident scenario (of course, release delay at the repository can postpone the radiation release to the biosphere, but assumption 16 negates this potential benefit). However, a container life of up to a few hundred years can provide substantial benefit in the water well intrusion scenario (and probably most other human intrusion scenarios). Beyond 1000 years, any event delay that might be provided by a durable container can actually increase the potential dose from the intrusion well scenario due to buildup of ^{226}Ra .
4. Only the delay in event initiation that could reasonably be provided by a very stable, dry geology (10^5 years or more) can significantly reduce the potential dose from either scenario once the few hundred year-lived container has been provided.

Results from investigation of the flow-field dispersion effects may modify these tentative conclusions that pertain to incentives for low release rate.

BARRIERS PERFORMANCE ASSESSMENT

A program was initiated by ONWI through a contract to Science Applications, Inc. (SAI), of La Jolla, California, in December 1978. The objective was to estimate the long-term in situ performance of current engineered barrier ("waste package") concepts in order to evaluate the concepts against the results of the PNL work. This SAI program is being continued in FY-1980 via subcontract from the PNL program in order to facilitate coordination of these two closely related efforts.

Interim Follow-on Studies

Analysis of additional variations of package design concepts suggested by the results of the FY-1979 performance assessment study was completed. Times of initial and final release to the geosphere were assessed for several design variations using the existing performance assessment model "BARRIER." Of particular interest was the use of cast solid stabilizers in the canister (Concept E) to aid in resisting crushing forces in soft rock media. Most of the follow-on design assessment work pertained to package design concepts B1 and E in salt and shale media. Also, additional barrier materials and the use of solid canister stabilizers in concepts other than Concept E were investigated.

Cost estimate results indicated substantial cost associated with the heavy sleeve design concept, B1. This was mainly due to the large amount of corrosion-resistant material needed on the large-diameter sleeve. The solid stabilizer design (E) was found more cost-effective because of the relatively smaller amount of material applied to a smaller diameter.

Packages for protection of spent fuel elements emplaced in geologic media can apparently be developed that sufficiently delay release of radioactivity from the package to the geosphere. Additional delay does not appear warranted.

A draft of the Task 3.1 report was completed near the end of the quarter and submitted to PNL for comment. The final report will be completed next quarter. Follow-on studies involving assessment model improvements and cost/benefit studies are expected to commence next quarter.

Program Support

Scoping studies of soft rock stresses were completed. It was found that salt creep rates would be very high, based on models and experiments of project Salt Vault. At 195°C and 1.8 KSI overburden pressure, a closure rate of 50% in 15 hr for Lyons salt and 50% in 20 hr for WIPP salt was found. Therefore, in salt media it is best to assume that full overburden stress is imposed on the package at time = 0. Improved models have been determined for loose backfill and backfill/sleeve combination. One result of these studies is the conclusion that the pressure-volume function (and hence the bulk modulus) currently in BARRIER may not be realistic, representing a sand that is too stiff. An alternative soft sand model was developed. Fairly simple analytical relationships allow straightforward calculation of the wall thickness required to avoid yield of the metal sleeve. Calculation can also be made easily for successive failures of inner shells (e.g., overpack and canister). The expressions are in terms of bulk modulus, shear modulus, and yield stress, and they calculate actual yield radiations rather than relying on ASME code criteria. The possibility of including the creep rate

(as opposed to assuming initial full overburden pressure) is being explored to accommodate some shale media where creep would be significant but slow enough to make rate important in the results. The external pressure buildup in time would form a boundary condition for the new stress profile model. As a result of the scoping calculations, significant revisions to the failure subroutine of BARRIER are anticipated. The changes should bring substantial improvement of the cost simulation, eliminating much of the previous conservatism and allowing a more realistic modeling of barrier performance.

SUMMARY REPORT PROGRESS

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