PNL-4250-3
UC-70

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

PNL--4250-3

DE83 013900

NUCLEAR WASTE MANAGEMENT SEMIANNUAL PROGRESS REPORT OCTOBER 1982 THROUGH MARCH 1983

Compiled by
T. D. Chikalla and J. A. Powell
Nuclear Waste Technology Program

June 1983

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

NOTICE
PORTIONS OF THIS REPORT ARE ILLEGIBLE.
It has been reproduced from the best available copy to permit the broadest possible availability.

Pacific Northwest Laboratory
Richland, Washington

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

CONTENTS

1.0 WASTE STABILIZATION 1.1
1.1 HANFORD WASTE VITRIFICATION PROGRAM 1.1
1.2 IN SITU VITRIFICATION OF HANFORD WASTE 1.19
1.3 IN SITU VITRIFICATION OF TRU WASTE 1.24
1.4 DEFENSE BYPRODUCTS UTILIZATION AND PRODUCTION 1.28
2.0 MATERIALS CHARACTERIZATION CENTER 2.1
3.0 WASTE ISOLATION 3.1
3.1 OFFICE OF NUCLEAR WASTE ISOLATION (ONWI) 3.1
3.1.1 Waste Package Program 3.1
3.1.2 Baselining and Benchmarking Salt Performance Assessment Codes 3.11
3.2 BASALT WASTE ISOLATION PROGRAM (BWIP) 3.20
3.2.1 Modeling Task Force 3.20
3.2.2 Preclosure Performance Assessment 3.21
3.2.3 Development of Eh-pH Sensors 3.22
3.2.4 Solubility Studies 3.26
3.2.5 Hydrothermal Testing of Materials in the 325 Building Hot-Cell Test Facility 3.27
3.2.6 Structural Barrier Materials 3.29
3.2.7 Sorption Studies 3.31
3.2.8 Repository Seal Design Engineering 3.32
3.3 NEVADA NUCLEAR WASTE STORAGE INVESTIGATIONS (NNWSI) 3.33
3.3.1 Waste Package 3.33
3.3.2 Structural Barriers 3.35
3.4 OFFICE OF NUCLEAR WASTE TERMINAL STORAGE INTEGRATION (ONI) 3.36
3.4.1 Simplified System Performance 3.36
3.4.2 RECON Repository Cost Analyses 3.41
3.5 OFFICE OF CRYSTALLINE REPOSITORY DEVELOPMENT (OCRD) 3.42
4.0 LOW-LEVEL WASTE MANAGEMENT 4.1
4.1 SPECIAL WASTE FORM LYSIMETERS - ARID 4.1
4.2 TRANSPORT ASSESSMENT - ARID 4.3
4.3 VERIFICATION INSTRUMENT DEVELOPMENT 4.7
4.4 MOBILITY OF ORGANIC COMPLEXES OF RADIONUCLIDES IN SOILS 4.9
4.5 GROUNDWATER TRANSPORT MODEL DOCUMENTATION AND EVALUATION GUIDELINES 4.11
5.0 REMEDIAL ACTION 5.1
5.1 RADON BARRIER SYSTEMS FOR URANIUM MILL TAILINGS 5.1
5.2 LINER EVALUATION FOR URANIUM MILL TAILINGS 5.12
6.0 SUPPORTING STUDIES 6.1
6.1 HANFORD DEFENSE WASTE STUDIES DEVELOPMENT OF ENGINEERING AND ENVIRONMENTAL INFORMATION 6.1

6.2 HANFORD DEFENSE WASTE STUDIES - PREPARATION OF AN ENVIRONMENTAL IMPACT STATEMENT FOR DISPOSAL OF HANFORD DEFENSE WASTES 6.2

6.3 WASTE MANAGEMENT SYSTEM STUDIES 6.3

6.4 NUCLEAR MATERIALS TRANSPORTATION TECHNOLOGY STUDIES 6.8

6.4.1 Thermal Analysis Methods 6.8

6.4.2 Structural Analysis Methods 6.8

6.4.3 Criticality Studies for Spent Fuel Transportation 6.9

6.4.4 Nuclear Materials Transportation Cost Studies 6.9

6.5 KRYPTON ION IMPLANTATION 6.10

6.6 IODINE-129 FIXATION 6.18

6.7 THERMAL OUTGASSING 6.19

7.0 RECENT WASTE MANAGEMENT PUBLICATIONS 7.1

FIGURES

1.1 Canister Modeling Configuration and Detector Locations 1.4

1.2 B-Plant In-Cell Equipment Configuration 1.8

1.3 Pilot-Scale Radioactive Demonstration Off-Gas System 1.20

1.4 Power, Melt Depth and Run Time 1.21

1.5 Melt Depth, Melt Position and Run Time 1.22

1.6 Off-Gas Temperatures 1.23

1.7 Effect of Temperature on Densification Rate of SrF_2 ..... 1.32

1.8 Effect of Pressure on Densification Rate of SrF_2 ..... 1.32

1.9 Dose Rate Map for Solid Pd (20% ^{107}Pd) in Contact with Tissue 1.36

1.10 Noble Metals Melter 1.38

2.1 MCC-14 Development Task Flowchart 2.2

2.2 Apparatus for Schoepite Solubility Measurements 2.4

2.3 Effect of Particle Size on pH 2.7

2.4 Effect of Particle Size on Leachant Composition 2.8

3.1 Time Dependence for Pressure Generation of Gas Produced by Gamma Irradiation of 70°C Permian Basin Salt Brine No. 1, Saturated NaCl, and Reference Basalt Groundwater 3.6

3.2 Plots of Solution Composition Versus Time for Four Leachates: Deionized Water, Reference Basalt Groundwater, Permian Basin Salt Brine No. 1 (SYN BRINE), and Permian Basin Salt Brine Made from Actual Salt Core 3.8

3.3 Plots of Solution Composition Versus Time for 76-68 Glass and 76-68 Glass Plus Iron or Titanium in Permian Basin Salt Brine No. 1 ..... 3.9

3.4 Plot of Saturation Values for Ca^{2+}, Mg^{2+}, SO_4^{2-} (mg/L Versus Relative Brine Strength at 250°C and 20 MPa 3.10

3.5 East-West Cross-Section of Major Depositional Features of the Permian Within the Palo Duro Basin 3.13

3.6 Major Physiographic Elements of the Texas Panhandle and Major Surface Water Drainage Features 3.14

3.7 Plan View of Hydrologic Benchmark Problem Area, Illustrating Surface Drainage, Outcrops, and Boundary Conditions To Be Considered 3.15

3.8 Cross-Section Views of Area Illustrating Geohydrologic Detail such as Layering (Stratigraphy), Boundary Conditions, Elevations and Geometric Detail 3.16

3.9 Plan Views Illustrating Areal Extent of Each Hydrologic Unit, Dimensions and Hydrologic Parameter Values for Each Unit 3.17

3.10 Isometric View of Initial Finite-Element Grid Used To Simulate Confined-Only Version of the Salt Site-Specific Benchmark Problem 3.19

3.11 PNL-Modified Dickson Autoclave 3.23

3.12 Eh Electrode 3.23

3.13 Fe-H_2O System at 200°C 3.24

3.14 Conceptual Design of pH Electrode 3.25

3.15 Removal by Filtration of Technetium from Oxidizing GR-3 Solution at 60°C for 30 Days in the Presence of 16 to 60 Mesh Umtanum Basalt 3.27

3.16 ^{99}Tc Fluid Concentration Versus Run Time for ^{99}Tc-Doped PNL 76-68 Glass Reacted with Basalt Water and Basalt Plus Basalt Water at 200°C, 30 MPA, and a 10:1 Solution to Solid Ratio 3.29

3.17 Uranium Sorption Kinetics on Cohassett Basalt from Reducing GR-3 Groundwater 3.31

3.18 Sorption of Technetium by Umtanum Basalt from Reducing GR-3 Solution at 60°C 3.32

3.19 Environmental Study Region Associated with the Region 3.43

4.1 Preliminary Engineering Drawing of Special Waste-Form Lysimeter Test Facility 4.2

4.2 Comparison of ^{60}Co/EDTA and Tritium Mobility Under Field Conditions at Hanford 4.4

4.3 Transpiration Experimental Site (Grass) 4.5

4.4 Moisture Distribution at Transpiration Sites 4.6

5.1 Effect of Moisture Content on Radon Attenuation 5.2

5.2 Effects of Vegetation and Rock Covers on Moisture Loss from Soil 5.8

5.3 Effect of Time on Efficiency of 3 1/2-in.-dia Charcoal Canister 5.10

5.4 Effect of Time on Efficiency of 15-in. Charcoal Canisters and PNL Flow-Through System 5.11

5.5 Relative Carbonyl Concentration as a Function of Depth into the Asphalt Liner 5.13

5.6 Comparison of Oxygen and Temperature Effect on Aging 5.14

6.1 1500-cm^2 Disposable Gas Trapping Unit 6.12

6.2 Sputtering Station 6.13

6.3 Four-Chamber Arrangement 6.14

6.4 Release of Krypton at 320°C in 166 Torr of Oxygen from an Ni-Li-Kr Alloy Initially Containing 1.32×10^{-3} Moles Kr/g Sample 6.16

6.5 Particle Size Distributions After Various Grinding Times 6.17

6.6 Krypton Content of $Ni_{0.82}Y_{0.11}Kr_{0.7}$ Alloy Versus Particle Size After Grinding in a Ball Mill 6.18

TABLES

1.1 Calculated Dose Rates for HWVP Melter 1.3

1.2 Decay Heat from Glass in HWVP Melter 1.3

1.3 Heat Transfer and Weight of Filled Canisters 1.4

1.4 Canister Dose Rate 1.4

1.5 Causes of Melter Feed Downtime 1.5

1.6 Predicted Effect of Amperage on the Surface Temperature of Electrode Connector 1.13

1.7 Release Rates for Selected Radionuclides Under Various Operation Conditions, %/hr 1.25

1.8 Initial Site Radionuclide Inventories, Ci 1.25

1.9 Public Doses from Routine Operations 1.26

1.10 Public Doses from Postulated Accidents 1.27

1.11 Nominal Composition of SrF_2 Prepared from $Sr(NO_3)_2$ 1.30

1.12 Results of Hot Pressing SrF_2 ..... 1.31

1.13 Comparative Utilization of Strontium-90 ..... 1.35

2.1 Comparison of MCC-1 Results from the NBS and the MCC 2.7

2.2 Specifications of Approved Testing Materials 2.9

3.1 Ferrous Materials Corrosion in mils/yr 3.3

3.2 Summary of Duplicate-Specimen Penetration Data, mils/yr 3.4

3.3 Irradiation-Corrosion Data Summary 3.4

4.1 Soil Properties 4.7

4.2 Vegetation Cover at Grass: Site Plant Count (Number) per 0.04 m^2 ..... 4.7

5.1 UMTRAP High-Priority Site Climatic Indices 5.5

5.2 Comparison of Tailings Pile Settlement Calculations Made by CSU and PNL 5.6

5.3 Average Radon Flux from FY 1981 Field Test Site - October 1982 ..... 5.9

5.4 Average Radon Flux from Column Tests - October 1982 ..... 5.9

6.1 Krypton Release, Pore Volume of Pores 12 to 800 Å Diameter and Surface Area of Pores Larger than 12 Å During Grinding for the $Ni_{0.82}Y_{0.11}Kr_{0.07}$ Alloy 6.18

1.0 WASTE STABILIZATION

The objective of the Waste Stabilization activities is to develop technology to recover or immobilize and package radioactive wastes from nuclear fuel cycle facilities for useful applications or for ultimate disposal.

1.0 WASTE STABILIZATION

J. M. Latkovich - Program Office Manager

Program Managers: J. E. Minor, V. F. FitzPatrick, G. L. Tingey

Major Contributors: G. E. Backman, S. M. Barnes, M. M. Beary, D. N. Berger, H. T. Blair, D. W. Brite, R. A. Brouns, J. L. Buelt, J. G. Carter, L. A. Chick, C. D. Flowers, R. W. Goles, M. S. Hanson, F. E. Haun, W. O. Heath, G. A. Jensen, D. E. Knowlton, C. A. Knox, D. E. Larson, D. R. Montgomery, K. H. Oma, J. M. Perez, R. D. Peters, D. H. Siemens, D. J. Silviera, S. C. Slate, S. L. Stein, C. L. Timmerman, R. J. Traub, E. J. Wheelwright

1.1 HANFORD WASTE VITRIFICATION PROGRAM (AR-05-10-02 80602)

The objective of the Hanford Waste Vitrification Program (HWVP) is to apply vitrification technology to a selected Hanford waste stream(s), developing the process and equipment to be utilized in a Hanford waste facility.

Background

The Hanford Waste Vitrification Program was initiated at the beginning of FY 1982. During that first year, program planning was based on development and demonstration of a vitrification system that could be operated and maintained in an existing Hanford facility, restricted to crane access for servicing and television for viewing operational and maintenance activities. The target waste stream selected was washed sludge from existing neutralized current acid waste (NCAW). In FY 1983, program scope was altered to develop a vitrification system for the B-Plant Waste Immobilization Annex (BWIA). The BWIA, when constructed, will provide window viewing. A combination of in-cell cranes and manipulators will be used for operations and equipment maintenance. This report reviews work from FY 1982 and the first half of FY 1983.

Summary

Program activities included vitrification process development, remote equipment component development and design, and vitrification system equipment design and fabrication.

Vitrification process equipment development included feed preparation and transfer to the melter, glass melting and pouring, glass canister handling, and melter off-gas decontamination.

A basic melter configuration was conceived and computer-code prediction of temperature and stress profiles generated. Isotherms and equal stress contours were found using mathematical models of the B-Plant melter. The generation of these models allowed design of refractory materials to minimize glass leakage and refractory cracking. Various remote sensing and control instruments for the melter were developed and tested. Development and testing were undertaken on ancillary glass melter hardware and process considerations such as off-center feeding and inverted graphite glass pour funnel to preclude as many process problems as possible. The number of connections to the melter was minimized. A heat pipe-cooled feed nozzle was designed and is under construction.

The following new remotized component systems were designed for use in existing B-Plant cells. (In designing and evaluating the process component systems, the components were designed for crane servicing only, a limitation imposed by the existing B-Plant cells.)

- canister handling system (turntable)
- level detection system
- product sampling system
- canister closure system
- electrode connector
- overflow section heater
- nozzle maintenance system (nozzle reamer)
- melter viewing system
- thermocouple replacement system
- fleed slurry transport system.

Preconceptual design of a vitrification facility intended for operation in a canyon facility was completed. The conceptual design drawing package for that system was completed. The HWVP scope change necessitates changes in all system equipment and permits the addition of feed concentration prior to vitrification. Preconceptual design studies of the vitrification system intended for use in the BWIA are now in progress.

Process Development

As the focus of the HWVP changed from processing existing waste to processing PUREX waste in B-Plant and then in the BWIA, the target waste stream changed, with waste glass development following. Preliminary waste glass designs have been completed for tanked residual liquor and washed neutralized current-acid-wastes. The choice to neutralize and wash the precipitate was made based on SO_4 solubility limits determined for the waste glasses. A variety of waste glasses, including borosilicate, phosphate, and lead were evaluated to find a glass amenable to accepting high quantities of sulfate. Borosilicate glass was chosen as the reference, and the physical, chemical, and electrical characteristics necessary for processing were evaluated.

Development of vitrification equipment included feed preparation and transfer to the melter, glass melting and pouring, glass canister handling, and melter off-gas decontamination.

A basic melter configuration was conceived and computer-code prediction of temperature and stress profiles generated. Isotherms and equal stress contours were found by use of mathematical models of the B-Plant melter. This allowed design of refractory materials to minimize glass leakage and refractory cracking. A variety of remote sensing and control instruments for the melter were developed and tested. Ancillary glass melter hardware and process considerations such as off-center feeding and inverted graphite glass pour funnel were developed and tested to preclude as many process problems as possible. The number of connections to the melter was minimized. A heat pipe-cooled feed nozzle was designed and is under construction.

Plasma torches were chosen to initially start and restart the glass melter. Tests were completed on existing large-scale melters to demonstrate the method's feasibility.

Heat and radiation generated by the waste in the melter was determined in order to support facility considerations, such as cell cooling and shielding as well as melter disposal options. Table 1.1 gives calculated dose rates at various points about the melter assuming a full tank of NCAW glass

TABLE 1.1. Calculated Dose Rates for HWVP Melter

	Dose Rate, Mrem/hr				
	0.5 yr	1.0 yr	3.0 yr	5.0 yr	10.0 yr
1) End shielded by electrodes	1.1E4	3.5E3	4.6E2	1.1E2	2.2E1
2) Sides	4.1E5	9.1E4	9.1E3	3.3E3	1.5E3
3) Above lid	2.4E7	4.6E6	5.1E3	3.1E5	2.1E5
4) Bottom	1.1E6	2.3E5	2.2E4	8.8E3	4.6E3
5) Glass surface	7.5E8	1.5E8	2.0E7	1.2E7	8.8E6
6) Burial box above lid	3.4E4	8.8E3	1.0E3	2.8E2	9.0E1

with a 6-month waste age. (Also included is the dose at the highest rate point around a high-density concrete burial box containing a disposed melter.) Table 1.2 gives the fission product decay heat generated in the melter at various time intervals, again with a full tank of NCAW glass.

Defining a system that can successfully meter liquid waste to the melter continues to be difficult. Various pump concepts have been evaluated and some tested. The low flowrate and high solids content of the liquid wastes eliminates many pump types. Airlifts with no moving parts were tested at PNL and under subcontract with Allied General Nuclear Services. To date, airlifts have not successfully handled the high solids content slurry. A pump test station was constructed, where various mechanical pumps will undergo 1000-hr testing to determine lifetimes and failure modes.

TABLE 1.2. Decay Heat from Glass in HWVP Melter

Decay Period, yr	Decay Heat, kW
0.5	17.4
1.0	6.07
3.0	1.19
5.0	0.506
10.0	0.294

Functional criteria for off-gas processing equipment were defined and issued. Engineering tests of selected off-gas processing equipment were completed.

Preliminary criteria were specified for the waste container system and the canister fill/cool program. A closure process for the B-Plant system was selected. A quality verification strategy for the waste package was drafted. Glass thermal properties were calculated to allow modeling of the temperature history of the waste canister.

Heat transfer and dose rate calculations were also completed for the canisters in support of both facility considerations and product quality verification. Results for various can sizes are shown in Tables 1.3 and 1.4 and Figure 1.1.

Melter operation tests were conducted in PNL engineering-scale ceramic melters with simulated NCAW glass. Early runs confirmed the suitability of the waste glass composition.

A five-day test was completed in which a simulated washed NCAW sludge and frit slurry composition was continuously pumped to the melter. During the 115.3-hr test, ~3512 L (3752 kg) of slurry were processed through the melter. Two hundred twenty kilograms of glass were airlifted in batchwise fashion from the melter into a glass-receiving canister.

TABLE 1.3. Heat Transfer and Weight of Filled Canisters (Neutralized, Washed CAW; 25% Waste Loading from Processing 12% ^{240}Pu)

Canister Diameter, m	Canister Weight, kg	Time from Reactor Discharge, yr	Canister Decay Heat, W	Specific Decay Heat, W/L	Surface Heat Flux, W/cm^2	Wall T due to Convection, °C	Wall T due to Convection & Radiation, °C	Thermal Gradient (Tw-Tc), °C	Center Line T, °C
0.51	1360	2	6156	15.2	0.193	271	135	242	377-513
0.41	850	2	3709	15.2	0.145	232	124	156	280-388
0.30	550	2	2310	15.2	0.121	210	116	88	204-326

TABLE 1.4. Canister Dose Rate

Detector Location (in Fig. 1.1)	Dose Rate, mrem/hr 20 in. D	16 in. D	12 in. D
1	2.6×10^7	1.9×10^7	1.1×10^7
2	1.1×10^8	1.1×10^8	9.7×10^7
3	2.6×10^7	1.9×10^7	1.1×10^7
4	5.3×10^7	6.0×10^7	6.1×10^7
5	3.6×10^7	3.1×10^7	2.6×10^7

Conclusions from the experiment are as follows:

1. The maximum feedrate of 50 L/hr was achieved but not sustained because of foaming of the melt.
2. The dilute slurry formed and sustained a flexible, free-floating cold cap.
3. Foaming of the melt occurred five times during the 5-day test.
4. With off-centered feeding plenum conditions were very stable in that very few off-gas surges occurred.
5. A curved, top-exiting, 4-in. off-gas line remained clear the entire run, and only trace deposits formed on the inside wall.
6. Melter instrumentation gave good indications.
7. A remote in-canister glass sampler that had worked well during pretests failed to obtain a glass sample under actual operating conditions. In later tests the sampler operated correctly.

The major objectives of this experiment were:

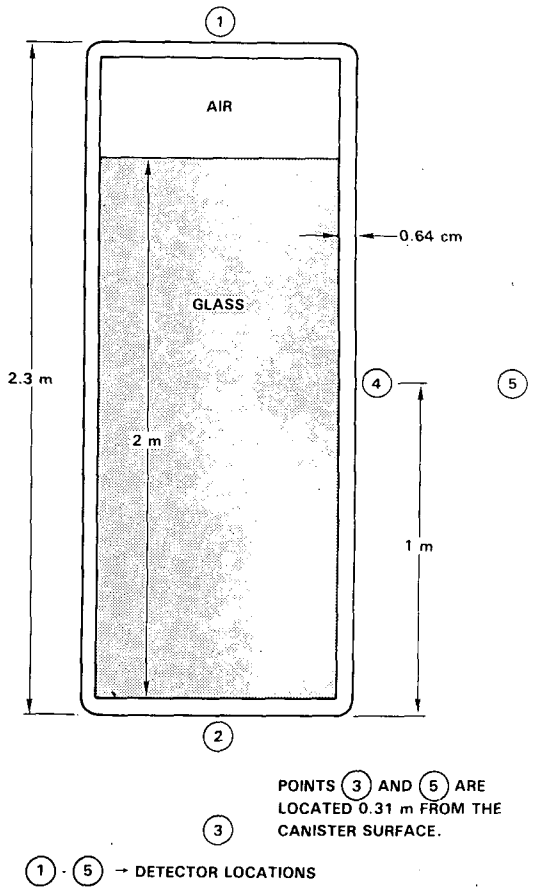


FIGURE 1.1. Canister Modeling Configuration and Detector Locations

1. to achieve and maintain a maximum feedrate of 66 L/hr m^2
2. to define control ranges and processing and monitoring responses
3. to further evaluate the concept of off-centered feeding

4. to continue evaluation of recently installed instruments designed to aid remote melter operation.

A discussion of the attainment of these goals follows.

Maximum Steady-State Production Rate

Downtime in which feed slurry to the melter was stopped accounted for 20.3 hr, giving an on-time efficienty of 82%. Table 1.5 lists the various causes.

Off-Centered Feeding

Slurry was fed into the melter through an off-centered feed port, in which no plugging occurred. With off-centered feeding, a less-than-100% cold cap was maintained, leaving some exposed glass surface, which appears to help maintain a steady melter environment with very few off-gas surges. Another benefit of some exposed glass surface is that the plenum space stays at a higher temperature, aiding in faster evaporation of the slurry and resulting in higher feedrates.

Before the first foaming incident, when the maximum desired feedrate of 50 L/hr had been reached, the melter parameters appeared stable. A cold cap over 70 to 80% of the melt surface was being maintained, with plenum temperatures of 400 to 500°C. During previous melter experiments using a slurry with an oxide loading of 478 g/L, feedrates of 60 L/hr were achieved with feed entering the melter at the centerpoint of the melter. Cold cap coverage during these tests was usually 90 to 95% and plenum temperatures were ~430°C. The slurry processing rate of the melter is largely dependent on the water content of the feed. Therefore, comparing the two slurries, over an hour period the same amount of water was evaporated (~48 L). This comparison suggests that the main advantage with off-centered feeding is greatly diminished off-gas surges; higher processing capacity has not yet been demonstrated.

TABLE 1.5. Causes of Melter Feed Downtime

Cause	Total Time, min.
Foaming in melter	1021
Syringe-type, pulsed feed system	136
Changeover to backup feed pump	28
Removal of Ru sorber bed	18
Install feed sample line	4
Total	1217 min (20.3 hr)

Evaluation of Instruments To Facilitate Melter Remote Operations

Three devices were used during the experiment. A bubbler tube was immersed ~2 in. below the melt level in the glass to indicate the glass/cold cap level and also the melter's general stability as shown by the bubbler's pressure fluctuations. It worked well to indicate the plenum's stability; the few venting surges appeared on the data strip chart as small spikes or unsteadiness in the line. Changes in glass level, however, were not well indicated.

Usually during overfeeding, the rate at which the water from the slurry is evaporated drops and the plenum space temperature gradually drops. The total off-gas flow is inferred by pressure drop across an orifice plate located in the off-gas line just upstream of the venturi. The orifice flow indication remained steady, indicating no discernible drop in the off-gas flow. The plenum temperature was also consistent, though somewhat low at 350 to 400°C. Other

parameters such as the melter resistance and voltage were also stable. If the cold cap had become firmly bridged, the glass temperature would have increased as the cold cap lost contact with the glass. Slurry would have accumulated and have been indicated by a lowered plenum temperature and off-gas flow rate. Evidently the cold cap was not bridged. Had it bridged, it is uncertain that the accumulating cold cap could have been determined via instrumentation only.

The second device was a plenum bubbler designed to indicate formation of foam or an excessive cold cap. The bubbler's open tip was ~4 in. above the normal operating glass level. During normal operating periods the bubbler correctly read the plenum-vacuum pressure of 5 in. H_2O and also indicated minor pressurizations in the melter. When foam developed and rose to cover the tip of the bubbler, the added resistance was immediately indicated on the chart recorder. The movement of the pen was very cyclic, drawing a broad band on the chart paper. As a new bubble formed at the tip of the bubbler rod, the chart pen went up scale. Then as the bubble was released, the pen dropped. The thickness of the foam layer was indicated by the position of the band on the chart paper.

One problem with the bubbler was apparent following a foaming situation. After foaming, some glass was left on the bubbler tube, which slowly ran down the tube and over the opening, causing pressure pulses to be recorded on the chart. These false foaming indications could be verified only by visually observing the inside of the melter.

The third device tested was a pair of glass thermowells positioned equidistant from respective electrodes but at different distances from the feed port. Most of the thermocouples were located near the glass/plenum interface so as to better monitor the condition of the cold cap. The melter was always to be fed at a rate such that one thermowell would indicate colder temperatures than the other. Ideally, the temperatures should indicate that the cold cap had not grown past the thermowell and that some clear glass surface had been maintained. The temperature indicated when the cold cap growth had reached the second thermowell. At this point about 30% of the observable glass surface was left open.

Top-Exiting Off-Gas Line and Ruthenium Adsorber

The off-gas system was modified to include a curved melter/scrubber connection resembling the B-Plant conceptual design and silica gel bed for trapping ruthenium. The new curved pipe operated well through the entire run, and post-run examination revealed only trace deposits in the uninsulated 4-in. pipe.

The silica gel bed gradually developed an excessive pressure drop after 2 days of operation and had to be removed. Examination revealed the bed had collected much dust, from the melter off-gas, with a reddish-brown color resembling feed solids. The high dust level in the bed was due to dirt-laden mist exiting the ejector-venturi scrubber, but this should not be a problem in the full-scale process facility, where this mist would be removed by a demister.

Equipment Remotizing and Testing

The emphasis of the HWVP component development and verification task has been to demonstrate the feasibility of operating and maintaining a liquid-fed ceramic melter system remotely in a canyon facility.

At the outset, it was planned that the vitrification system would be installed in existing cells in B-Plant. In B-Plant cells remote operations are performed using only an overhead crane (with a rotating hook) and an impact wrench. Only periscope viewing is available.

Subsequent studies have shown the prudence of constructing new cells for the ceramic melter system. The currently proposed facility will be an annex to the existing Hanford B-Plant, designated as the B-Plant Waste Immobilization Annex (BWIA). In the BWIA, the ceramic melter system will be maintained remotely using an overhead crane (equipped with a rotating hook) and an impact wrench; however, viewing windows and master-slave manipulators will be provided at selected locations for better viewing and manipulator assistance during maintenance activities, if required. Testing of remote components to support the BWIA is in the early stages.

In designing and evaluating the process component systems, the considerably more demanding restrictions imposed by the B-Plant remote capability were adhered to. The following new component systems were designed for use in existing B-Plant cells:

- canister handling system (turntable)
- level detection system
- product sampling system
- canister closure system
- electrode connector
- overflow section heater
- nozzle maintenance system (nozzle reamer)
- melter viewing system
- thermocouple replacement system
- feed slurry transport system.

Figure 1.2 locates each component system with respect to the proposed melter (the level detection, thermocouple replacement, and feed slurry transport systems are not shown). Each component system is discussed below, where, in general, each system is described and the findings of remotability and operability tests are summarized.

Canister Handling System (Turntable)

In radwaste vitrification, metal canisters are lowered into the hot cell; filled with glass product; cooled, sealed, and transported elsewhere for surface decontamination and storage. In the cell a turntable is used to position an empty canister below the melter pour spout, where molten glass product pours from the melter into the canister. After each canister is filled and allowed to cool, it is capped and removed from the turntable. The turntable consists of a circular rotating rack that suspends four canister holders 90° apart. The canister holders protrude below the rack and into a tank equipped with cooling coils for the controlled solidification of the product. The turntable rotates in 90° increments: an empty canister is lowered into the cell, positioned under the spout, filled, moved through two cooling stations, and then moved into position for removal.

The turntable is rotated by an electric motor turning a cam-operated index drive through a triple-reduction gear box. The output shaft from the cam-operated index drive fits into a socket in the turntable. Rotation slowly accelerates, peaks, and then decreases to a halt as the cam turns to a "dwell" or flat spot. As the cam continues to turn, it engages the cam follower, which turns the output shaft and rotates the turntable to a new position. A limiting mechanism controls the turntable rotation, stopping it, and accurately positioning the canister holders at each of the four stations.

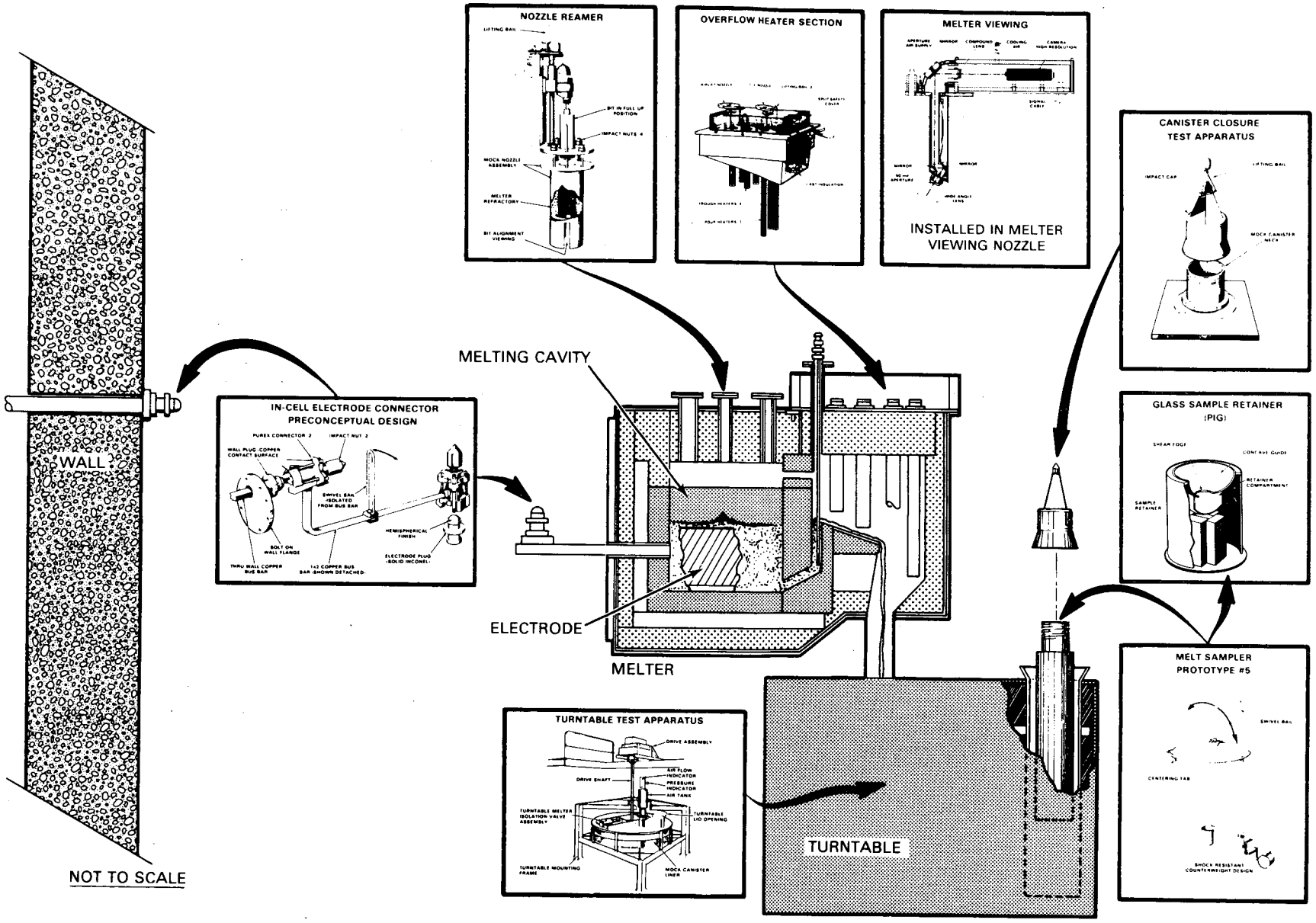


FIGURE 1.2. B-Plant In-Cell Equipment Configuration

If the motor or the cam-operated index drive should fail, the turntable can be rotated by a crane-held impact wrench. In this case, the turntable driveshaft is removed and the socket on the impact wrench is slipped onto a shaft designed for wrench rotation.

When a filled canister in the turntable is to be replaced with an empty one, a turntable port is opened. Before it is opened, however, an isolation valve separating the turntable from the melter pour spout is closed to prevent unrestricted flow of cell air through the open turntable port into the melter. As the isolation valve is directly under the melter, it cannot be directly installed or replaced. A set of rails and a screw jack mechanism fixed to the top of the turntable allow indirect valve operation. A valve gate is mounted on a set of gliders that sit on the rails. Turning a nut with a crane-held impact wrench drives the valve gate along the rail into a position between a flange on the pour spout and one on the turntable port. The wrench-driven mechanism slips the valve gate into and out of a wedge-shaped gap between the melter and the turntable. The valve gate is designed to mate nearly airtight with the flanges of the pour spout and the turntable.

Evaluation of a scaled-down prototype of the turntable indicates that the canister handling system concept is feasible. (The prototype turntable rack had one shortened canister holder and no cooling tank.) Testing was primarily concerned with the remote operation of the isolation valve assembly and turntable drive mechanism, as well as characterization of air inleakage associated with the isolation valve. Tests also evaluated the effect of spilled glass on operation of the isolation valve.

Remote replacement of the isolation valve assembly with the overhead crane was demonstrated. To remove the isolation valve assembly, the impact wrench turns a screw which withdraws the valve gate from the melter flange. When the valve gate is clear of the melter, the lockdown nuts are removed with the remote impact wrench and then the isolation valve assembly, with the use of the crane hook, is lifted off the rail that guides it into position under the melter.

Valve gate sealing was evaluated. In a valve test at ambient temperature, the air inleakage rate was 1.3 scfm, with a vacuum of 3 in. of water. At a temperature of 247°C, which is more typical of actual melter operation, the air inleakage rate was 1.0 scfm with a vacuum of 13 in. of water.

The limiting switch that stops rotation was cycled through 50 rotations of the turntable with negligible misalignment in the turntable. No slop or play was observed in the connection between the turntable driveshaft and the socket connecting the shaft to the turntable (which could cause misalignment).

To evaluate the effect of spilled glass on operation of the isolation valve, 50 mL of glass were poured on the valve gate at both ambient temperature and 240°C. In one test, the glass was allowed to solidify before the valve was closed; in another test, the valve gate was operated while the glass was still molten. In both tests, the glass did not affect either valve operation or the rate of inleakage.

Level Detection System

The proposed level detection system consists of an array of γ-ray detectors mounted in the cell wall plus a transmission source

within the cell. The detector array, transmission source, and the canister being filled are aligned, with the canister placed between the source and the detector array. As the canister is filled, vitrified waste product reduces the intensity of the γ rays that pass through it from the source to the detectors. This source attenuation, detected, establishes whether the canister is filled. The vertical array of detectors allows the presence of glass to be verified at several discrete levels within the canister.

Since the γ-ray source and detector must be aligned in the same horizontal plane, each detector within the array requires a separate source. One way to meet this need is to install a single source spanning the length of the canister (~10 ft). Another method is to mobilize a smaller γ-ray source so that it moves up and down the length of the canister. The source is then visible to each detector periodically rather than continuously. While both methods will work, the second method is much less expensive than a single, canister-length source. The concept of using the vitrified waste product within the canister as the level source was considered but later abandoned since the ^{137}Cs loadings within the waste product are not sufficient to provide a useful detector response. In-cell deployment of the γ-ray detectors was found capable of overcoming this sensitivity problem, but lack of engineering support for this configuration precluded its use.

An NaI γ-ray detector was evaluated for use in the level detection system. The NaI detector is a solid-state γ-ray scintillation device which possesses the highest inherent γ-ray detection sensitivity of all commonly used γ-ray detectors. Consequently, a relatively small NaI detector can replace and outperform much larger γ-ray detection systems. Also, once deployed, the NaI detector requires no physical support or maintenance.

Two γ-ray transmission sources, ^{137}Cs and ^{60}Co, were evaluated for in-cell use. The ability of the proposed level detection system to establish the level of glass within a canister was then assessed for each. Remote installation and replacement of the mobile γ-ray source assembly will be verified in testing. A source-mobilizing mechanism is being designed.

An evaluation of the level detection system under actual canister glass-filling conditions indicates that the current concept is feasible and that ^{60}Co is more versatile than ^{137}Cs as an in-cell transmission source. The evaluation involved an NaI detector, a ^{60}Co source, and an in-can melter. In the experimental configuration, the source and the detector were situated in the same horizontal plane and were confined to move (as a unit) vertically up and down the length of the canister. Canister glass level data were collected by this system under stationary as well as scanning conditions. In both cases, the attenuation of the γ-ray signal produced by the glass was unmistakable and the adequacy of an external transmission source for establishing glass level was unequivocally verified.

Of the two transmission sources considered, ^{60}Co was judged superior because of much lower strength requirements, the absence of feed-related background problems, and compatibility with optional direct waste glass γ-ray detection methods. For B-Plant applications, a ^{60}Co source strength ~10^4 curies is required.

The combination of an encapsulated, enclosed ^{60}Co source and a vertically mobile stage for positioning the source is desirable for several reasons. A vertically mobile

source requires only a single-point source of activity, which could be procured, shipped, deployed, and ultimately disposed of using uncomplicated, standard procedures. Furthermore, in this configuration, the source moves continuously in the vertical direction, allowing the vertical extent of the detector viewing window to be as narrow as desired, thus maintaining resolution of the canister level position. Extending the viewing window horizontally will allow positional tolerances of the detector boreholes within the cell wall to be relaxed without greatly affecting the significance of the counting data. Finally, ^{60}Co is not expected to ever be a major radionuclide component of the slurries processed by the HWVP melter. All of this simplicity, of course, is at the expense of providing a mechanical device within the hot cell. However, this additional complexity is generally not considered a serious drawback.

Testing also indicates that the transmission source level detection approach is well suited to diagnose nonuniform canister filling conditions (known collectively as "roping"). This result stems from the fact that interpretation of the data generated from a transmission source is independent of absolute counting rate. It is the ratio of counting rates (attenuation factor) that directly relates to the uniformity of the canister fill. Consequently, system response functions are quite predictable under all conceivable canister filling conditions.

Product Sampling System

To verify the vitrified waste's composition, a sample is collected and sent to a laboratory for analysis. The sampling system is to obtain a glass sample suitable for analysis without interfering with either the glass-making or canister-filling operations. The product sampling system consists of a device (melt sampler) that is manually placed within the open neck of a canister before the canister is lowered into the hot cell. After the melt sampler is installed in the canister neck, the canister is lowered into the cell and centered under the melter pour spout. When the canister is in place, the pouring operation begins. As the glass stream falls from the pour spout, it enters a sample cup attached to a pivoting arm. The opposite end of the arm is counterweighted in such a way that when the desired weight of glass is collected, the sample cup pivots downward, allowing unobstructed filling of the canister. The counterweight also prevents the arm from pivoting before a sample is obtained. The cup itself pivots on the end of the arm so that as the arm swings downward, the molten glass does not spill. The sampler design incorporates centering tabs which ensure that the sample cup is placed within the narrow path of the stream. After the canister is filled and cooled, the melt sampler is removed from the neck and lowered into a sample retainer that captures the cup and sample. The rest of the melt sampler, including the counterweighted arm assembly, is discarded: only the sample cup is sent to the laboratory.

The tests of the melt sampler demonstrated remote installation, sampling, and sample collection with no complications. To simulate actual operation, the melt sampler was installed under the pour spout of the pilot-scale ceramic melter (PSCM). Five samples weighing from 18.5 to 64.0 g were collected. The fifth sample was recovered after the empty canister in which it had been placed was filled. This dispelled the concern that the sample would spill or otherwise be lost when heat from the glass in the canister

remelted the sample. The pivot point on the cup held the cup upright, preventing sample loss.

The remote installation and removal of the melt sampler were demonstrated using a 12-in.-dia canister neck; the sampler is adaptable to neck diameters of 9 to 16 in.

Finally, the ability to remotely collect the sample cup within the sample retainer was demonstrated repeatedly.

Canister Closure System

After filling and cooling, canisters are capped and later sealed. The closure system, which caps the filled canisters, consists of a threaded cap with lifting bail, an impact nut centered on the bail, and a stop flange that encircles the lower portion of the canister neck. The exterior of the canister neck is threaded, as is the interior of the cap which skirts the canister neck. The in-cell canister capping operation is accomplished by first lowering the cap by crane onto the canister neck. The crane hook is then removed and a crane-operated impact wrench screws the cap onto the canister. The cap is tight when the bottom surface of the cap skirt mates with the upper surface of the stop flange on the canister neck. The seam between the cap and the stop flange may later be welded shut. The impact wrench is removed, and the assembled canister and cap are lifted out of the turntable using the cap-lifting bail.

Evaluation of the prototype system indicated that it functioned well. A prototype cap was lowered onto the canister neck and then screwed on tightly using an impact wrench. Changing the orientation of the cap lead-in threads with respect to the starting threads on the canister neck had little effect on the closure operation; a crane-operated impact wrench accomplished the tightening regardless of the initial cap orientation. The anticipated thermal expansion of the canister neck, caused by heat conducted from the partially cooled product, was simulated by heating the prototype canister neck to various temperatures throughout testing; only above 300°C did screwing the cap on become difficult. Once typical in-cell canister neck temperatures are known, thread tolerances on the cap and/or the canister neck can be designed to accommodate the corresponding thermal expansion.

The possibility of glass being dripped from the spout onto the canister threads was simulated in two tests and was observed to have a negligible effect: using the crane to set the cap onto the canister neck was sufficient to shatter the glass. In the second test, glass was poured onto the stop-flange, covering some threads with a mound of solidified glass. This also failed to obstruct closure: the ratcheting action of the impact wrench tightening the cap vibrated the glass loose.

After remote installation of the cap under simulated cell conditions was demonstrated, the ability of the cap to remain tight during canister removal was evaluated. A load corresponding to the weight of a filled canister (4200 lb) was pulled on the lifting bail with the cap tightened onto the canister neck. The cap was then subjected to transient mechanical shocks, by means of shocks applied to the lifting bail with a 20-oz ball peen hammer, so that each shock applied a torque that would have tended to unscrew the cap. The cap remained tight throughout the shock test.

Electrode Connector

Vitrification within a ceramic melter requires electricity to generate the heat needed to maintain the process bulk temperature at 1150°C, melt raw glass-forming feed components, and evaporate water and other volatiles in the feed slurry. The electrode connector is a remotable jumper that provides an electrical connection between the melter electrodes and a through-wall electrical service line. The service line is connected to a power source located outside the cell wall.

The electrode connector is adapted from proven PUREX jumper technology. A PUREX jumper consists of a pipe, an attached lifting bail, and two PUREX connectors (one at each end of the pipe). PUREX jumpers are used to couple piping within a cell, and they make possible the remote replacement of particular lines from within the network of pipe characterizing most processes. The electrode connector-PUREX jumper concept departs from the standard technology in that electricity rather than fluid, passes through a jumper made of solid copper rather than steel pipe. It also differs from existing remote electrical connection schemes because of its high (2000-A) capacity. What the current design does share with other PUREX jumpers is a history of reliable remote serviceability. In addition, the current electrode connector design is better adapted to remote operations than are previous designs because its contact surfaces are hemispherical rather than flat. The hemispherical shape allows a limited amount of rotation without reducing electrical contact: the perfect translational and rotational alignment of the melter and wall plug contact surfaces is not required.

A field demonstration of the electrode connector verified its ability to safely handle the 2000-A maximum current for which it was designed. The electrode connector was installed in series with the primary electrode power cables on the liquid-fed ceramic melter (LFCM). The LFCM served as an electrical load, enabling the ampacity of the electrode connector to be evaluated. Made of solid copper, the electrode connector was sized for a low resistance (<0.001 Ω), thus minimizing the effect of joule or resistance heating. During the test, the electrode connector handled currents of up to 1000 A with a maximum temperature rise of 10°C attributable to joule heating.

It is estimated from the test data that the surface temperature of the electrode connector would have reached 51°C during 2000-A operation and 40°C during the normal 1500-A operation. Table 1.6 summarizes the predicted effect of amperage on the in-cell surface temperature of the electrode connector (assuming an ambient cell temperature of 40°C).

A megohmmeter was used to verify the electrical isolation between the electrode connector and a plug wall flange. A megohmmeter simulates high-voltage conditions while resistance is measured (frequently electrical isolation that seems adequate at low voltages will break down at high voltages). The

TABLE 1.6. Predicted Effect of Amperage on the Surface Temperature of Electrode Connector

Amperes	Predicted Temperature, °C
1000	50
1250	54
1500	58
1750	63
2000	68
2500	80
3000	92

megohmmeter indicated an infinite resistance across each electrical isolation point, verifying the effectiveness of each insulating component.

The remotability of the electrode connector did not require verification since it incorporates all the essential remotability features of the standard PUREX jumper technology.

Overflow Section Heater

During canister filling, the glass stream that leaves the melting cavity requires external heat to remain molten. Unchecked cooling would affect not only its fluidity, but also the quality of the final solidified product.

The overflow section heater provides a heat source for the glass product stream leaving the melter. It consists of a well-insulated lid equipped with seven coaxial heating elements of silicon carbide. The heaters protrude downward from the lid and are designed to maintain an operating temperature of ~1050°C in the vicinity of the overflow trough and pour spout. The operating temperature is monitored using thermocouples (T/Cs) which enter through a T/C nozzle on the lid of the overflow heater. A split safety cover prevents accidental contact of cell equipment with the exposed electrical connections on the upper surface of the heater lid. Since the heating elements are expected to have shorter service life than the proposed melter, the section is remotely replaceable. The overflow section heater is lowered remotely, via a lifting bail, over the melter overflow section, and is bolted to the melter using remotely tightened impact nuts.

Evaluation of the overflow section heater indicated that further design development is required to provide more protection for the heating elements during remote installation. (Currently there is nothing to prevent the heating element assemblies from contacting the melter or other equipment during remote installation. Such contact, made while lowering the section heater, could easily damage the elements and put the entire overflow section heater out of commission.) While testing indicated that a careful operator could successfully install the section, its installation is far from foolproof. Nevertheless, it is concluded that its remotability is feasible and that required design changes are relatively minor.

As part of the evaluation, it was determined that the vibration caused by remotely tightening the impact bolts was not sufficient to break the heating elements. After 1 min of ratcheting with a standard PUREX wrench, the overflow heater was lifted and the heating elements were inspected. No damage was observed.

An attempt was made to evaluate the air inleakage attributable to the overflow heater lid. During the test, the vacuum source (pump) was measured as 1 in. of water, while a rotometer indicated 1.4 scfm air inleakage. A vacuum gauge on the test assembly showed normal atmospheric pressure within the container box-lid assembly. Since the pump was unable to maintain a vacuum within the container box-lid assembly, the observed 1.4 scfm inleakage should be taken as the maximum air throughput capacity of the pump, rather than the maximum rate of air inleakage through the overflow heater lid. The only relevant conclusion that could be drawn from this test was that the rate of air inleakage

attributable to the overflow heater lid during melter operation will be >1.4 scfm. In near-future testing, the inleakage source and magnitude will be more clearly identified. Based on this information, a design modification can be implemented that would lower the rate of inleakage to an acceptable level. Tentatively, a 10-scfm rate of inleakage at a vacuum of 10 in. of water is considered the maximum acceptable rate of air inleakage through the overflow heater lid.

An attempt was made to heat the overflow section heater assembly to 1050°C to ascertain the structural integrity of the heater section materials during thermal cycling. Although the heating elements were wired in various parallel and series combinations, power was insufficient at the time of testing to reach the 1050°C operating temperature.

When the heaters were wired in parallel, a temperature of 750°C was reached after several days at the maximum power output of the electrical source. The rate of temperature increase was at all times kept below the 40°C/hr value reported as the maximum curing rate by the manufacturer of the cast insulation. After reaching a steady-state temperature of 750°C, the heater assembly was allowed to cool at a rate less than 40°C/hr. When cool, the overflow heater was lifted from the containment box and visually inspected. One small crack in the insulation was noted. No changes were observed in the heating elements, the cages surrounding the elements, the metal liner surrounding the insulation, or the upper portion of the lid.

Nozzle Maintenance System (Nozzle Reamer)

While the solid glass formers and radsalts remain in the melter, the volatile feed components vaporize and leave the melter as off-gas. Along with the exiting gas, entrained feed particles leave the melter through an off-gas nozzle. Some of the entrained particles are deposited on the walls of the nozzle and, with time, build up a tough, ceramic-like coating. Within months, the nozzle may become partially clogged. Other melter nozzles, employed as viewports or as access to instrument sensors, may also become clogged. The function of the nozzle reamer is to remove the blockage from a clogged nozzle so that it can be reconnected for service.

The nozzle reamer is a core-drilling apparatus that is lowered, via a lifting bail, onto an open melter nozzle. A flange on the reamer fits over guide dowels and threaded studs on a flange on the melter nozzle. The reamer is then bolted into place with impact nuts that are tightened remotely. A drill feed system lowers a rotating, diamond-surfaced bit into the nozzle. The bit cuts a core through the blockage and thus unclogs the nozzle. At the point where the rotating bit cuts through the blockage and enters the melting cavity, the drill feed system automatically reverses and lifts the bit back out of the nozzle. When the bit is clear of the nozzle, power to the assembly is disconnected and the reamer is remotely removed from the melter.

An evaluation of a prototype nozzle reamer indicated that the nozzle maintenance concept will work well during in-cell operation. Using a mock nozzle, constructed in accordance with the dimensions and materials as planned for the proposed HWVP melter, the remote installation, operation, and removal of the nozzle reamer were demonstrated. Testing determined that the nozzle reamer is fully remotable.

To simulate a nozzle blockage, a castable ceramic was poured into a form located near the bottom opening of the mock nozzle. Each form consisted of an annular ring 6 in. high. After being poured, the ceramic was allowed to dry for 24 hr and then was cured for an additional 24 hr with heat from two 150-W heat lamps. Alfrax-66® was selected to simulate the melter blockage material because it could be easily cast within the mock nozzle, adhered well, and formed a tough ceramic when cured. How closely the Alfrax-66 simulated the actual blockage material is not known, since necessary information about the properties of that material is not available. Another unestablished variable was the extent of nozzle blockage anticipated. It was assumed that nozzles would be reamed before becoming fully clogged; hence, an annular rather than solid blockage was poured. The inner diameter of the simulated blockage was selected at 5 in., which corrresponds to a 30% nozzle blockage.

An evaluation of the reamer's ability to cut through a simulated 30% melter blockage, located near the bottom opening of a melter nozzle, was successful. Before the test, it was recognized that if the melter was maintained at its normal vacuum during the nozzle-cutting operation, the air drawn through the open nozzle might help remove the drill cuttings (fine powder) from the drill area. To simulate this, a vacuum cleaner hose was centered under the simulated blockage. As the cut was performed, a vacuum was applied. After cutting 1 in. into the blockage, the vacuum was removed and the cut was examined. Virtually all the powdered material had been pulled out of the cutting area. The cutting operation was resumed, and the core bit cut completely through the blockage without incident. When the bit passed the bottom of the nozzle opening, the feed direction reversed and the cored-out blockage material fell from the bit intact, as anticipated.

During the simulation, the eccentricity or wobble associated with the rotating bit was measured several times and was never greater than 1/32 in. off center. A wobble of this small magnitude is normally not visible.

Part of the bottom edge of the nozzle liner material broke off along with the cored-out blockage. The relatively minor damage indicated that the cutting pressure, and thus the feedrate, was acceptable.

Melter Viewing System

Television viewing of the melter cavity is an interim technique for monitoring foaming and cold-cap bridging within the melting cavity. Viewing the melting cavity is also a supplementary method for measuring the depth of the glass. The camera provides an image of the melting cavity walls and the cold cap at the surface of the molten glass. With additional experience in melter operation, it may be possible to correlate abnormal conditions (such as bridging and foaming) with melter temperatures and off-gas flow rates. This would make television viewing unnecessary.

The melter viewing system consists of: 1) a probe, with lenses and mirrors, that extends through the viewport nozzle into the ceiling area of the melting cavity, and 2) a connected video camera outside the melter. An aperture, at the end of the probe housing, allows light from the melter cavity to be directed into the camera lens. A nonbrowning lens material would be used for radioactive

®Registered trademark of Carborundum Company, Refractories Division, Keasbey, New Jersey.

service. The view through the aperture provides an image of the melting cavity and the cold cap at the surface of the molten glass. An air stream cools the optics within the housing and exits (into the melter cavity) through the aperture.

An evaluation of the melter viewing system indicated that observation of the melt during the feeding operation is feasible. The ruggedness of the melter viewing system was successfully demonstrated when a crane-operated impact wrench was used to fasten the television camera to a flange on a mock viewport (nozzle). With the television operating, the ratcheting, vibrating action of the impact wrench was allowed to continue for 1 min after the bolts were tight, causing a white line to appear on the video screen. The picture returned to normal when the vibrations stopped.

The melter viewing system was also able to take the shock caused by swinging the probe section into the test port as the crane positioned it for mounting in the flange. In addition, the melter viewing equipment was dropped ~3 ft and was not damaged.

To simulate actual operation, the melter viewing system was installed in a side nozzle of the pilot-scale ceramic melter (PSCM). The quality of the television picture correlated with melter temperature above the melt. When the melter was idling, the picture was almost competely vacant of detail. The temperature in the melter plenum then exceeded 900°C. When the melter began processing a fresh feed, the melting cavity walls, viewing port, feed tube, and thermocouple tube were all distinguishable in the television picture. The corresponding melter plenum temperature was 630°C.

When the melter viewing system was initially installed in the PSCM, the picture was faint and distorted into a rounded diamond shape. This distortion was later identified as being caused by the electromagnetic force (emf) of the power cables, which were close to the melter viewing system. The distortion can be avoided during in-cell operation by locating the viewing port away from the power leads. Shielding of the television camera electronics is also advised.

After 14 days of observing the melt within the idling melter, the television picture became clouded by an encrustation of small crystals that limited the view of the melt to a small hole in the center of the picture. During the test, a continuous air flow was maintained through the aperture and the camera temperature was <100°C. The melter viewing system was removed and the encrustation scraped off. The television picture returned to normal: it was round, free of distortion, and had good resolution. When the melter viewing system was reinstalled, it displayed no evidence of encrustation 52 days later, which included both melter feeding and melter idling. The camera temperature throughout this period was ~200°C. An encrustation-free operation of this length of time gives some assurance that the high system temperature is the means of controlling the problem. If, however, the encrustation of the objective head should again pose a problem, one of the following modifications could be implemented: 1) additional air around the viewing aperture, 2) a jet of steam across the encrusted area, 3) a mechanism for removing encrustations without removing the melter viewing system, or 4) electrically heating the encrusted area. Whether any modification is required, and if so, which are implemented, is relegated to future testing.

Thermocouple Replacement System

The function of the thermocouple replacement system is to facilitate replacement of inoperative T/Cs in the melter cavity. This is required as these T/Cs are expected to have a shorter service life than the proposed melter. The system provides a continuous guide path from the gallery to the melter thermowell, eliminating a set of signal-weakening contact points within the cell. An invention disclosure will be filed on the system; further design details are omitted.

The evaluation of the prototype thermocouple replacement system indicated that the concept functioned well. The ability of a crane operator to remove and replace both a T/C jumper and the melter thermowell was successfully demonstrated. In addition, ten long-sheathed T/Cs were inserted through the assembled system and were removed, as would be done in-cell. All tests were repeated several times to establish, with certainty, that T/Cs can be remotely installed and replaced.

Feed Slurry Transport System

The melter feed slurry, composed mainly of glass formers and radioactive particles suspended in water, must be transported through a nozzle on the upper surface of the melter into the melting cavity. Design restrictions associated with this particular application prevent the use of conventional slurry transport systems. The slow feedrate (~50 L/hr), combined with poor rheological properties of the slurry (rapid and irreversible particle sedimentation), causes possible clogging of the feed line to be a major concern. Since the particles within the slurry are highly abrasive, material wear within the transport system is also a problem. Finally, the use of plastics, e.g., for valve seals or pump diaphragms, is prohibited because of their extremely rapid degradation in a radioactive environment.

The proposed slurry transport system consists of a slurry pump, a feed line, a feed line flushing system, a feed nozzle, and all the valves and remote pipe connections associated with the system. An all-metal, positive-displacement pump has been designed, fabricated, and tested for the feed slurry transport application. The slurry pump is air-operated. A sealing system prevents the operating air from being exposed to radioactivity, thus eliminating the air as a possible means of operating-gallery contamination. The pump design avoids close-toleranced moving parts, decreasing the likelihood of failure caused by abrasive wear. Finally, the pump's positive-displacement mechanism allows the low slurry feedrate to be tightly controlled.

Tests indicate that the slurry pump concept is feasible for in-cell service. Simulated melter feed slurries were successfully pumped at a controlled flow rate for over 1500 hr. Future testing will include studies of pump lifetime, sealing system effectiveness, feed line flushing, system configuration, and system remote operability. An invention disclosure will be filed on the system; further design details are omitted.

Design and Fabrication

Preconceptual design of a vitrification facility intended for operation in a canyon facility was completed. The conceptual design drawing package for that system was completed. The HWVP scope change necessitates changes in all system equipment and permits

adding feed concentration prior to vitrification. Preconceptual design studies of the vitrification system intended for use in the BWIA are now in progress.

1.2 IN SITU VITRIFICATION OF HANFORD WASTE (AR-05-10-02 - 80441)

The objective of this program is to develop and evaluate the in situ waste vitrification concept to confirm its feasibility for large-scale use.

Summary

The primary thrust of the FY 1983 efforts is to complete a pilot-scale radioactive demonstration in June 1983. A significant secondary effort is completing the conceptual design of the large-scale system.

Design of the pilot-scale system began in September of 1982 and was completed in mid-November. The initial field test unit located behind the 318 Building was used as a starting point for the design. Significant additions, however, particularly in the cover gas treatment system, were required for containment of volatilized radioactive materials. Fabrication was completed in late January, and the new trailer-mounted pilot-scale system was moved to the test site in February. Installation and site preparation was completed in March, the Operational Acceptance Test (OAT) was conducted March 16 to 18, 1983, and was thorough. Several design and operational difficulties were encountered, and a second OAT will be conducted to verify the adequacy of the corrections. The hot test will be conducted in June, on schedule.

Preparations for the OAT were extensive and rigorous. Supporting documentation has been provided in a timely fashion throughout the design, fabrication, and test cycle. Only minor changes to the Safe Operating Procedure and Run Plan are required, to make the documentation current with the corrective action.

Conceptual design of the large-scale system is proceeding at a satisfactory pace, and remains on schedule.

Pilot-Scale Radioactive Demonstration

Design of the pilot-scale system for the radioactive system began late last fiscal year and was complete in mid-November 1982. The experience gained from tests conducted with the fixed system, located behind the 318 Building, served as a starting point for the design. The design for the radioactive system has three significant changes/improvements over the previous system: 1) improved cover gas system, suitable for radioactive materials, 2) improved electrical system, with a greater range of control, and 3) portability, as the system is now trailer-mounted.

The cover gas system is shown schematically in Figure 1.3. All components downstream of the venturi-ejector scrubber were added to upgrade the system to handle radioactive materials. The Hydro-Sonic® scrubber was added to remove submicron particles and semivolatiles. The separator-condenser-separator-heater were added to dewater and condition the gas stream so as to have minimum impact on the HEPAs.[a] The HEPAs were added to ensure confinement and containment of radionuclides that might pass through the scrubbers. The cover gas system is located

® Registered trademark of Hydro-Sonic Systems, Dallas, Texas.
(a) High-efficiency particulate air filters.

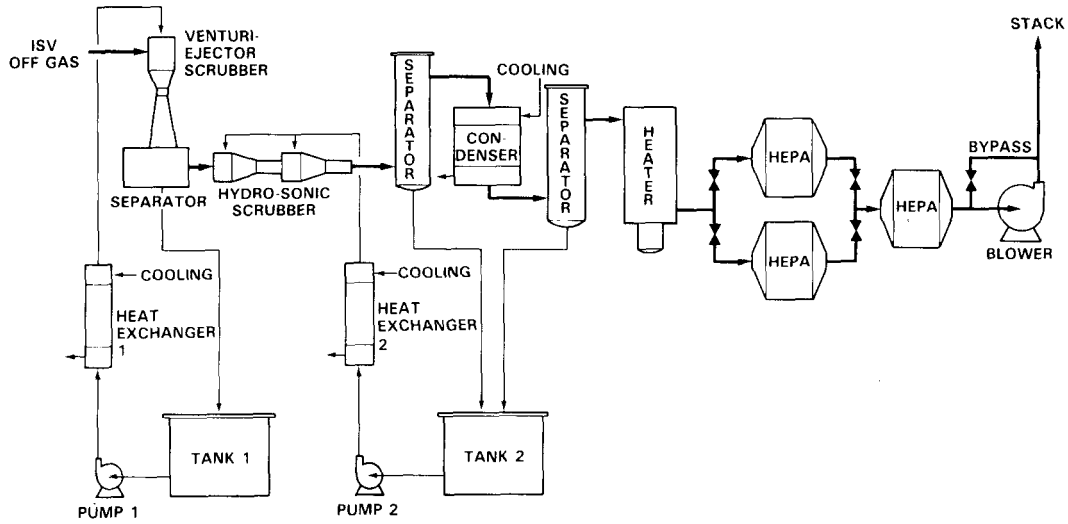


FIGURE 1.3. Pilot-Scale Radioactive Demonstration Off-Gas System

in a large glove box, to protect the operating personnel in the event of a leak or malfunction. The move to a glove box environment is a significant design change.

The electrical system power rating was increased 25% to 500 kW, and the voltage taps were extended down to 250 V, giving a 100% increase in depth capability. The transformer electrical configuration was changed from an open delta to a Scott Tee, allowing more even distribution of the load. The operability of the system was improved and simplified by purchasing more modern equipment.

The cover gas, electrical and instrument systems were mounted in a standard semitrailer, greatly increasing options for future use and improving weather resistance.

The components, the assembled cover gas system, and the semitrailer were obtained by competitive fixed-price procurements. Assembly and installation was performed by plant forces, and fabrication was completed in late January. The trailer was moved to the test site in February, site preparation and acceptance testing were complete in mid-March, and the OAT was conducted March 16-18, 1983.

Preparations for the OAT procedure were extensive and rigorous. Documentation prepared and approved includes:

Site Selection Criteria
Quality Assurance Plan
Safe Operating Procedure
Environmental Evaluation
Safety Evaluation Document
Radiation Work Procedures
Operational Readiness Plan
Run Plan

The site was cleared of surface vegetation and an access road graded. Ground-penetrating radar, magnetometer (ferrous metals), EMI (ground conductivity), and metal (nonferrous) detector surveys of the leveled site indicated that no foreign buried materials, underground piping, cables, or utilities were present. Also a general radiation survey of the site and specific surveys of the excavation and backfill were performed to ensure no spurious radiation was present.

Operator training sessions conducted for all operations staff include:

General Radiation Safety
Step-Off Pad Procedures
Glove Box and Glove Change-Out Procedures
Bag-Out Procedures
Cardiopulmonary Resuscitation (CPR)
Fire Extinguisher Training
Electrical System Operation
Instrument and Control System Operation
Off-Gas System Operation
Sampling System Operation
Emergency Action

The Operational Readiness Review Board conducted an informal walk-through to independently verify the state of preparedness for conducting the OATP. Use of the Management Oversight Risk Tree analysis to corroborate signoffs in the Operational Readiness Plan worked very well.

The OAT provided a system checkout for both the power and off-gas equipment and showed that the integrated system performed adequately. The OAT was completed in 36 hours, when the melt depth reached 5 ft. Power and melt depth plots through the OAT run are provided in Figure 1.4. Total energy consumption was 9920 kWh, with a maximum power input of 410 kW and an average of 275 kW. An additional plot of melt depth as associated with melt position is illustrated in Figure 1.5. This figure shows that the centerline and electrode melting progress at about the same rate. Later in the run the melt edge spreads to include the outer positions to form a melting front that appears to cover the entire 16-ft^2 area between the electrodes.

Problems encountered during and after the run are highlighted below.

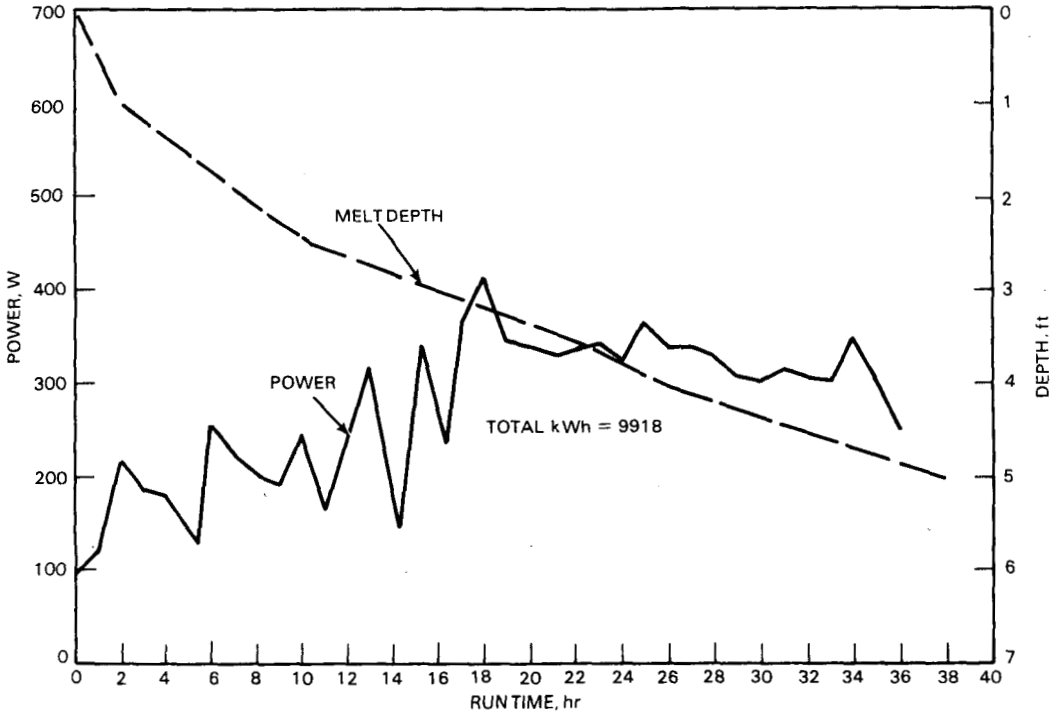


FIGURE 1.4. Power, Melt Depth and Run Time

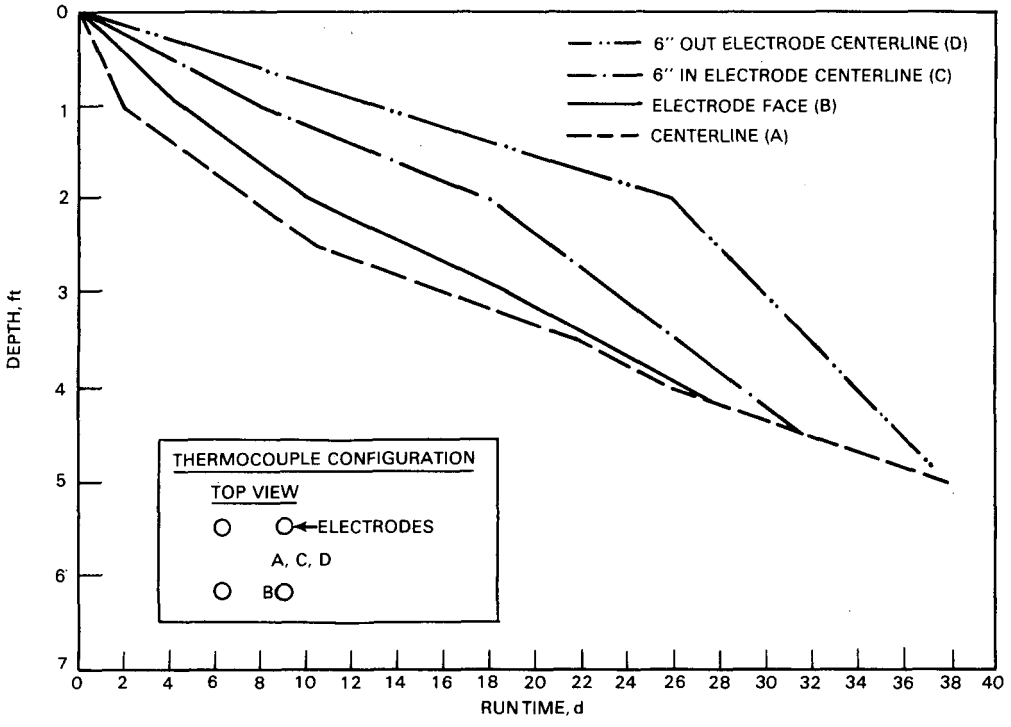


FIGURE 1.5. Melt Depth, Melt Position and Run Time

- Hood vacuum was reduced due to the drying and subsidence of the soil maintaining the seal. A high-temperature skirting will be installed on the hood to assist in maintaining the hood vacuum.
- High temperature and high current caused the circuit breaker to trip, interrupting power to the electrodes. The current limiter has been adjusted to prevent further current trips. A shrouding will be placed around the transformer cooling fan to enhance the air flow across the transformer heat sink, improving its heat transfer to prevent future thermal trips.
- Computer data logging performed well; however, a subprogram nesting error continued to occur. Resolution of this problem is being investigated with assistance from Hewlett-Packard.
- Water was found in the first-stage HEPA filter. The HEPA performed effectively under its increased differential pressure without rupturing. Testing is under way to accurately determine the cause of water buildup. The temperature restraints (<40°C) of the blower did not allow the heater to evaporate all of this moisture. A higher temperature blower is being installed to allow greater evaporation.

The presence of water in the HEPA is a potentially serious problem. In addition to a higher temperature blower, drain lines from the HEPA housings to Tank #2 will be added, and the system will be operated at a lower differential pressure with lower scrub solution flow rates in the venturi and Hydro-Sonic scrubbers.

Examination of the melt after the run indicated a uniform 9-in. subsidence of the surface between the electrodes. This subsidence was caused by the minimal cold cap formed during operation. The surface was not

molten during the run (as it had been in the second pilot-scale field test), but a thin, dull red cold cap formed, causing a maximum hood plenum temperature of 350°C later in the run. The higher plenum temperature assisted with the oxidation of the graphite electrodes above the melt.

The off-gas system for the OAT was well-instrumented for temperatures, pressures, flows, gas composition, and particle-size distribution. Scrub tank samples are currently being analyzed by inductively coupled plasma (ICP) and atomic absorption (AA) for element compositions, as well as for pH, anion concentration, and percent solids. The first-stage HEPA filter used during the run is also being chemically analyzed.

The off-gas flow rate during the OAT averaged 362 scfm ± 1.5% and seemed relatively constant, even though temperatures and vacuum in the off-gas containment hood changed. The first-stage scrubber, a venturi-ejector, was operated with a liquid flow rate averaging 42 gpm at a pressure of 101 psi. The venturi-ejector actually boosted the off-gas flow slightly, having a 2.9-in. water column (wc) net gas pressure gain at the outlet. The second-stage scrubber, a tandem nozzle Hydro-Sonic, was operated with a liquid flow of 23.4 gpm at a pressure of 43.0 psi. The average pressure drop across the Hydro-Sonic scrubber averaged 59.5 in. wc. The unit is designed to scrub efficiently at pressure drops above 30 in. wc.

Figure 1.6 shows off-gas temperatures in the hood, the off-gas line, and at several points in the off-gas system. Temperatures in the hood remained relatively low until about 15 hr into the run, when the cold cap began to thin. Then the plenum temperature climbed to an average of ~320°C, where it remained until the run was completed. The temperature at the inlet of the off-gas system averaged 240°C. Off-gases were effectively cooled during the run, as indicated by the condenser outlet temperature, which averaged 20°C, depending on ambient temperature and off-gas heat load. A heater brought

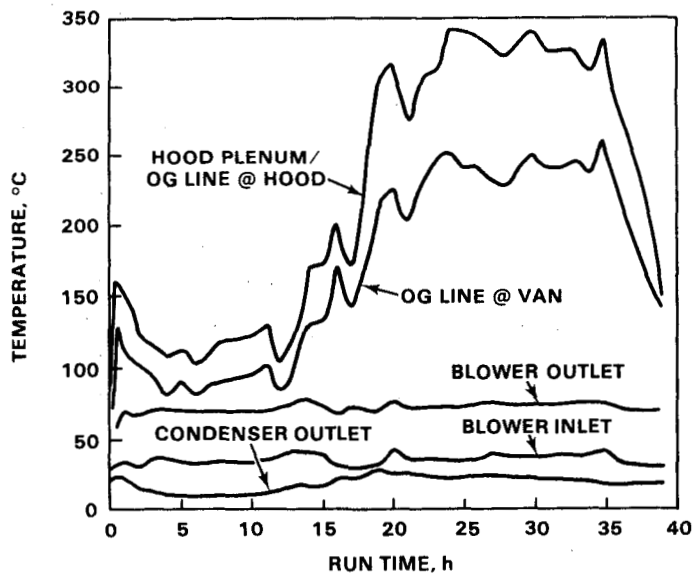


FIGURE 1.6. Off-Gas Temperatures

the off-gases exiting the condenser above the dewpoint before they passed through the HEPA filters and blower. The blower inlet was limited to ≤40°C due to the temperature rating of the blower bearings.

Another OAT will be conducted the first week in May to verify the effectiveness of the corrective actions. The radioactive demonstration will be conducted in June on schedule.

1.3 IN SITU VITRIFICATION OF TRU WASTE (AR-05-15-20 - 80583)

The objective of this program is to determine the generic applicability of the in situ vitrification technology for radioactive waste treatment and disposal.

Summary

A public safety analysis, comparing the pilot-scale (1.2 x 1.2 m) and the large-scale (5 x 5 m) in situ vitrification systems, was performed. The 216 Z 1A site, located in the 200 W Area on the Hanford site, was selected as the reference case. Public dose commitments and the 50-yr dose commitment to the bone (the critical organ) of the maximally-exposed individual were shown to be orders of magnitude below the DOE guidelines.

Public doses were also calculated for potential accident conditions. Six accidental release scenarios were developed, and the two worst cases were examined in detail. All assumptions were conservative. The 50-yr dose commitment to a maximally exposed offsite individual is estimated to be less than 1.0 rem for the large-scale system, less than that for the pilot-scale system. Because of the conservative assumptions used in estimating the doses from postulated accidents, the doses are undoubtedly higher than would actually be received.

Public Safety Analysis

A public safety analysis, comparing the pilot-scale (1.2 x 1.2 m) and the large-scale (5 x 5 m) in situ vitrification systems, was performed. The 216 Z 1A site, located in the 200 W Area of the Hanford Site, was selected as the reference case. Both routine operations and potential accident cases were evaluated. The basis for and the results of the analysis are described below.

Radionuclide Releases

The following assumptions were used to estimate the source term for the 216 Z 1A site:

- Surface area of the contaminated zone is 90 x 30 m.
- The waste site has a 0.3-m layer of uncontaminated overburden.
- The 216 Z 1A waste site inventory is uniformly distributed within the site.
- The entire waste inventory is contained in a 0.6-m layer immediately beneath the overburden layer.
- The full-scale and pilot-scale off-gas systems have equivalent decontamination capabilities.

The rate constants for radionuclide releases, in percent per hour, are given in Table 1.7 for three possible operating conditions. These values, in most cases, represent maximum measured values during pilot-scale tests with nonradioactive simulants. Normal operations, with a cold cap in place, are assumed for 75% of all operating time. Normal operations without a cold cap are assumed for the remaining 25% of the run. The

TABLE 1.7. Release Rates for Selected Radionuclides Under Various Operation Conditions, %/hr

Element	With Cold Cap	No Cold Cap	Gas Venting
Co	0.005	0.005	0.5
Sr	0.0005	0.0005	0.05
Ru	0.0	0.0	0.0
Cs	0.01	0.01	0.5
U & Pu	0.0	0.005	0.5

gas-venting release rates represent the expected release rate of isotopes into the hood for a site containing combustibles. However, since no combustibles are assumed to be in the refernce site, these rates have been used to estimate release rates to the atmosphere following postulated accident events.

The source-term inventory used is given in Table 1.8. For simplicity, the total inventory of the site (column 1) was divided by the number of runs necessary to vitrify the entire site for each case. They were 102 runs for the large-scale system and 1200 runs for the pilot-scale system.

For the routine operations, the total quantity of radionuclides released per run to the off-gas system was calculated using the inventory of Table 1.8 and the rate of Table 1.7 and applying the equation:

$$Q(\text{released}) = Q(\text{initial})\ (1 - e^{-R/100\ t})$$

where R is the rate constant in percent per hour (Table 1.7) and t is time in hours. The release quantity was calculated separately for each operating condition (i.e., with and without cold cap) using the appropriate fraction of total run time, and then was summed to attain the total release quantity. The run times were 24 hr/melt for pilot scale, and 34 hr/melt for full scale.

TABLE 1.8. Initial Site Radionuclide Inventories, Ci

Component	216 Z 1A Inventory	Large-Scale Melt	Pilot-Scale Melt
^{238}Pu	1.8E+1	1.8E-1	1.5E-2
^{239}Pu	8.5E+2	8.3E+0	7.1E-1
^{240}Pu	2.1E+2	2.0E+0	1.8E-1
^{241}Pu	2.5E+3	2.4E+1	2.1E+0
^{242}Pu	7.0E-2	7.0E-4	6.0E-5
^{241}Am	4.0E+1	4.0E-1	3.0E-2
^{90}Sr	7.7E-1	7.5E-3	6.4E-4
^{137}Cs	1.4E-1	1.4E-3	1.2E-4
^{106}Ru	8.9E-3	8.7E-5	7.4E-6
^{60}Co	3.0E-2	2.9E-4	2.5E-5
^{233}U	3.6E-2	3.5E-4	3.0E-5
^{234}U	3.7E-2	3.6E-4	3.1E-5
^{235}U	1.1E-3	1.1E-5	9.0E-7
^{238}U	2.7E-2	2.6E-4	2.2E-5

Off-gas system efficiency was estimated to range from 10^4 for semivolatile nuclides, and 10^5 for "heavy metals," to 10^8 for particulates. For this analysis, the 10^5 value for all nuclides was used, mainly because the volatiles will not contribute for this site (216 Z 1A) and because it is conservative.

Public Dose for Routine Operations

Routine doses were calculated using the DACRIN (Houston et al. 1974) computer program. One-year doses and 50-yr dose commitments to a maximally exposed individual and to the general public from routine operations are given in Table 1.9 for the organs of interest, total body, bone, and lung. The routine doses are all rather small.

The 50-yr dose commitment to the bone (the critical organ) of a maximally exposed individual in the public from one melt, arises almost entirely from released isotopes of plutonium. The dose commitment is 9×10^{-9} rem

TABLE 1.9. Public Doses from Routine Operations

	Maximum Individual, rem		Population, man-rem	
	1-yr Dose	50-yr Dose	1-yr Dose	50-yr Dose
		Large-Scale Operations		
Total body	7E-11	4E-8	3E-6	2E-3
Bone	2E-9	9E-7	7E-5	4E-2
Lung	2E-7	8E-7	7E-3	3E-2
		Pilot-Scale Operations		
Total body	4E-11	3E-8	2E-6	1E-3
Bone	1E-9	6E-7	4E-5	2E-2
Lung	1E-7	5E-7	4E-3	2E-2

for the large scale, and 6×10^{-10} for the pilot scale, assuming a nominal off-gas decontamination factor of 10^5. Thus, for any run, the dose is about a factor of 15 greater for the large scale. However, to do the entire example site, the dose from all runs is 9×10^{-7} rem for the large scale and 7×10^{-7} for the pilot scale.[a] The total dose for the entire site is slightly higher for the large-scale system (about 20%) because, although fewer runs are required, the scale-up predictions are such that any one particular piece of soil is molten for a slightly longer period than for the pilot scale, allowing an incrementally greater release. The doses in either case are extremely small compared with the 500-mrem limit for the maximum individual and the 175-mrem limit for an average individual permitted under DOE guidelines.

Potential Accident Releases

Six scenarios for accidental release were developed to evaluate the potential impacts for the large-scale and pilot-scale systems. These were: 1) a leak in the off-gas line between the hood and the off-gas system, 2) an underground explosion of buried chemicals, 3) a leak of the off-gas cleanup water inside the off-gas trailer, 4) a gas leak inside the off-gas trailer, 5) an accidental release of radionuclides while removing overburden from a site prior to vitrification, and 6) a criticality. Only the first two provide any discrimination between the pilot- and large-scale operations. This is because they are the only ones that cause an increase in dose to the public over routine operations. These two events are described as the following:

- Effluent Line Leak - The run is proceeding normally, with no cold cap. The effluent line between the hood and off-gas system is disrupted such that all the gaseous effluents are released directly to the atmosphere. Power to the electrodes is cut off immediately. The effluents released to the atmosphere decrease exponentially as a cold cap forms over the melt area. The doses reported for this scenario were calculated assuming gas venting was occurring at the time of the accident. If there was no gas venting, the doses

(a) Doses for all runs for the example site are derived by multiplying the dose by the number of runs; 102 for the larger scale system and 1200 for the pilot-scale system.

would decrease by a factor of five for the case of no cold cap, and, because no transuranics are released with a cold cap, by a factor greater than 100 for a melt with a cold cap.

- Explosion - The run is proceeding normally, with no cold cap. An explosion occurs within the hood with sufficient force to dislodge the hood from the off-gas system. Power to the electrodes is cut off immediately. Gaseous effluents are assumed to be released at a rate ten times normal for 5 min following the explosion, and then revert to an exponential decrease from the normal level with a 0.5-hr half time. Gas venting is assumed for this scenario because any substance supplying the energy for an explosion is also assumed to contribute to venting.

Public Doses from Potential Accidents

Doses for the accident scenarios were calculated using the HADLOC computer code and are presented in Table 1.10. The worst accident postulated for an operating ISV system is a chemical explosion destroying the off-gas hood, and thus incapacitating the off-gas system. The 50-yr dose commitment to an off-site individual in this case is conservatively estimated to be less than 1.0 rem for the large-scale system and 0.08 rem for the pilot-scale system. The order of magnitude difference reflects the increased amount of material available for release. This assumes that the melt is undergoing gas venting at the time of the explosion, the weather conditions are unfavorable, and the individual is only 5 miles away for 4 hr. The explosion

TABLE 1.10. Public Doses from Postulated Accidents

	Maximum Individual, rem		Population, man-rem	
	1-yr Dose	50-yr Dose	1-yr Dose	50-yr Dose
A) Explosion				
		Large-Scale Operation		
Total body	1E-3	5E-2	3E+0	1E+2
Bone	2E-2	1E+0	6E+1	3E+3
Lung	1E-1	3E-1	3E+2	7E+2
		Pilot-Scale Operation		
Total body	8E-5	4E-3	2E-1	1E+1
Bone	2E-3	8E-2	5E+0	2E+2
Lung	1E-2	2E-2	3E+1	6E+1
B) Off-Gas Leak				
		Large-Scale Operation		
Total body	4E-4	2E-2	1E+0	6E+1
Bone	1E-2	5E-1	3E+1	1E+3
Lung	5E-2	1E-1	1E+2	3E+2
		Pilot-Scale Operation		
Total body	4E-5	2E-3	1E-1	5E+0
Bone	9E-4	4E-2	2E+0	1E+2
Lung	5E-3	1E-2	1E+1	3E+1

scenario is an upper limit; however, it is not significantly worse than a simple loss of off-gas system.

Because of the conservative assumptions used, the doses estimated from the postulated accidents are undoubtedly higher than would actually be received. The relative magnitudes of the accident doses should be used for comparing the two options rather than estimating the actual effects.

1.4 DEFENSE BYPRODUCTS UTILIZATION AND PRODUCTION (AR-05-15-25 - 80576/ GE-03-47-03 - 80401)

The objective of this program is to identify applications for nuclear byproducts and to develop techniques for recovery of these materials. Emphasis in FY 1983 will focus on recovery and potential applications of fission-product strategic noble metals, development of advanced isotope-fueled power-production devices, and demonstration and deployment of radioisotope-powered lights in the far North.

Summary

The DOE Plan for Recovery and Utilization of Nuclear Byproducts from Defense Wastes, along with an Executive Summary under separate cover, has been submitted to DOE for approval.

The techniques of melt casting and hot pressing of strontium fluoride have been examined to obtain high densities, and thus, higher heat-per-unit volume for byproduct strontium-90 capsules. Both techniques are capable of yielding bulk densities 99% of theoretical. However, the apparently simpler process of melt casting has a further advantage of purifying the power during the process.

High-efficiency thermomechanical generators are considered for coupling with radioisotope-heat sources. Stirling engines appear to have a number of advantages for these systems, and mechanical, thermal, and shielding calculations have been performed for various design concepts. The program goal is to construct a radioisotope-powered convertor for testing and demonstration.

Irradiated nuclear-reactor fuels contain considerable quantities of the strategically important noble metals, palladium, rhodium, and ruthenium. Health physics effects of the small amount of radioactivity associated with these metals are being evaluated and show that the weak beta from ^{107}Pd will exhibit no hazard from an external dose but could be of some concern from an internal dose. Four techniques for recovery of the noble metals from the nuclear waste include: carbon-sorbate adsorption, selective reducing-resin adsorption, precipitation during sugar denitration, and molten-lead extraction.

Department of Energy Plan for Recovery and Utilization of Nuclear Byproducts from Defense Wastes

The DOE Plan for Recovery and Utilization of Nuclear Byproducts from Defense Wastes (DOE-DP-0013, Vol. 2) has been prepared and submitted to DOE for review and comment. A short Executive Summary (DOE-DP-0013, Vol. 1) was also prepared and submitted under separate cover to be used by management, Congress, and others desiring the conclusions and rationale of the plan without the supporting data used in its development. The plan concludes that uniquely useful, intrinsically valuable, and strategically important materials are contained in the fraction of the reprocessed fuel now considered waste. Some of these products have already proven applications while others have high potential

for use in applications such as food technology, agriculture, energy production, public health and safety, medicine, industry, and national defense. The plan also concludes that the removal and use of several of these byproducts will significantly reduce the heat and radiation remaining in the waste and thus facilitate its management and disposal.

The plan outlines DOE's policy and strategy for recovery and development of applications for these nuclear byproducts. It recommends a development plan consistent with cost/benefit principles involving the potential user early in the research phase of the program. The plan recommends that the initial program focus on the following six areas: 1) heat and power sources, 2) food and sludge irradiators, 3) self-powered lighting, 4) strategic noble metal recovery, 5) rare gas supply and application, and 6) supporting studies. The plan also contains detailed supporting information on the current and potential availability of selected byproducts and a description of currently identified applications.

Isotopic Heat Source Development

The major heat-producing isotope from the nuclear byproducts is strontium-90, which along with its short-lived daughter, yttrium-90, yields 0.93 W of heat per gram of the isotope. This isotope has been used to a limited extent for remote power generation using thermoelectric conversion of the heat to power. The strontium is currently removed from the waste at Hanford to reduce the thermal output of the remaining waste and is converted to SrF_2 and encapsulated in double-walled metal capsules and stored under water in the Hanford Waste Encapsulation and Storage Facility (WESF). In the past, the dried SrF_2 powder has been compacted in the metal capsules by cold compaction techniques to a density ranging from about 3.0 to 3.4 g/cm^3. For many byproducts applications, however, it would be advantageous to reduce the volume of the isotope. To achieve this reduction in volume, we have initiated a modest effort to evaluate the feasibility and determine some of the operating parameters for melt casting and hot pressing or extruding SrF_2 to a density near theoretical (4.24 g/cm^3). For these experiments, impurities were specifically added to nonradioactive $Sr(NO_3)_2$ and precipitated as the fluoride to simulate the fission-product composition from WESF. The composition of the powder containing ~5% impurity is shown in Table 1.11.

Melt Casting

Pure SrF_2 melts at 1473°C and vaporizes at 1 atm pressure at 2489°C. Therefore, we conclude that melt casting of the compound has potential for obtaining a higher density solid. Melt casting experiments were performed in a stainless steel vacuum chamber equipped with coils for induction heating. To date, studies have been conducted by: 1) melting and casting the powder directly in the graphite mold, 2) melting in a graphite melting crucible and bottom-casting into a receiving mold for cooling and solidification or 3) melting and casting directly in a tantalum mold. In all three cases, the casting operation was conducted under about 1/3 atm of argon because heating under vacuum caused excessive spattering due to rapid out-gassing.

The impure powder melts at ~1350 to 1400°C, but experiments were conducted from this range up to 1500°C in an attempt to

TABLE 1.11. Nominal Composition[a] of SrF_2 Prepared from $Sr(NO_3)_2$

Component	Weight %
Sr	64.40
F	31.30
Al	0.15
Ba	0.45
Ca	0.15
Cd	0.02
Cr	0.15
Fe	0.22
K	0.12
Mg	0.02
Mn	0.01
Na[b]	1.50
Nd[c]	0.50
Ni	0.15
Pb	0.02
Zr[d]	0.84

(a) Based on composition of feed to precipitation step.
(b) May vary, depending on washing of fluoride precipitate.
(c) Nd used as stand-in for all rare earths.
(d) Zr in $^{90}SrF_2$ will increase with time from the decay process, $^{90}Sr \rightarrow {}^{90}Y \rightarrow {}^{90}Zr$.

optimize the process parameters by decreasing the viscosity and increasing the out-gassing rate. Time at temperature was also varied from 5 to 20 min before cooling and solidification. In most cases, the powder had been vacuum-dried for 24 hr at 275°C prior to baking in air at 1000°C, but two experiments were conducted on powders dried only at 150°C to evaluate the feasibility of eliminating a separate drying step.

The results of this series of tests have led to the following conclusions:

- Melt-casting appears to be a viable option for obtaining high-density solid strontium fluoride in hot cells.
- Physically stable SrF_2 ingots of ~7.5 cm dia by 4.8 cm high have been produced with densities ranging from 3.8 to 4.2 g/cm^3 (90-99% TD).
- Excellent castings were obtained when the molten salt was bottom-cast from a graphite melting crucible into a graphite mold. Bulk densities in excess of 96% of theoretical were achieved with the porosity limited to small pores on the surface of the side wall.
- Significant chemical purification occurred during the casting; sodium and potassium fluorides tend to volatilize and condense on the cold vacuum chamber surface; iron and some other metal ions are reduced by the carbon and deposit as metals on the crucible wall.
- Casting dimensions appear to be controllable and predictable.
- Casting powders dried at 150°C, but not baked, yield relatively high-density solids (~90% TD). Porosity appears to be due to incomplete outgassing of the moisture from the melt. Further out-gassing or bottom-pouring may be required to yield the desired density.
- Cast SrF_2 can be sanded on a wet-belt sander to adjust final dimensions without substantial dust generation.

Other parameters will be examined in future experiments. Castings with a length/diameter ratio of 3 to 4 will be demonstrated and a variety of out-gassing techniques examined. On completion of these studies, adaptation of this technique to hot cell use and for use on waste generated from future PUREX operation should be considered.

Hot Pressing

Another technique that has been used to obtain high-density SrF_2 is hot pressing. It has the advantage over casting of not requiring so high a temperature, but it does require high-pressure equipment. The hot press used in these experiments has a maximum temperature capability of 2200°C and a pressing force of 30 tons in a 25-m^3 hot zone. The experiments examined for this study used a graphite die 2.2 cm inside diameter and 11.5 cm long with pressure applied only from the top. Maximum pressures in the test were limited to 27.5 MPa (4000 psi) by the strength of the graphite die.

A series of experiments was conducted to examine the effect of temperature, pressure, and atmosphere on the resulting solid. The results are shown in Table 1.12. These results show that strontium fluoride can be hot pressed to near theoretical density over a reasonable range of operating conditions. Hot pressing does not significantly purify the powder; thus the specific power achievable from $^{90}SrF_2$ is somewhat lower than cast powders due to both higher impurity content and lower theoretical density. The relationship of pressing time, temperature, and pressure on density are shown graphically in Figures 1.7 and 1.8.

Development of Radioisotope-Powered Thermomechanical Generators (RTMG)

Radioisotope thermoelectric generators (RTG) have been used as remote power sources for both space and terrestrial applications for many years. Most space applications have used ^{238}Pu as the power source, while terrestrial applications generally use ^{90}Sr. As potential applications of remote power expand,

TABLE 1.12. Results of Hot Pressing SrF_2

Run No.	Furnace Temp., °C	Final[a] Die Temp., °C	Pressure, psi	Immersion Density, g/cm^3	Time for Densification, min
141	1000	940	4000	4.26	7.2[b]
142[c]	900	825	4000	4.13	11.2
143	850	780	4000	4.11	17
144	800	730	4000	4.11	20
145	750	700	4000	4.10	31
146	680	637	4000	4.03	125
147	850	780	3000	4.11	19
148	850	780	2500	4.11	19
149	850	815	2000	4.10	45
151[d]	850	740	4000	4.11	17
153[e]	850	780	4000	4.11	26

(a) Or temperature when densification was complete.
(b) For first-stage densification.
(c) A small amount of material extruded into the punch/die gap but did not come out of the die.
(d) Argon atmosphere.
(e) Length/diameter = 2.73.

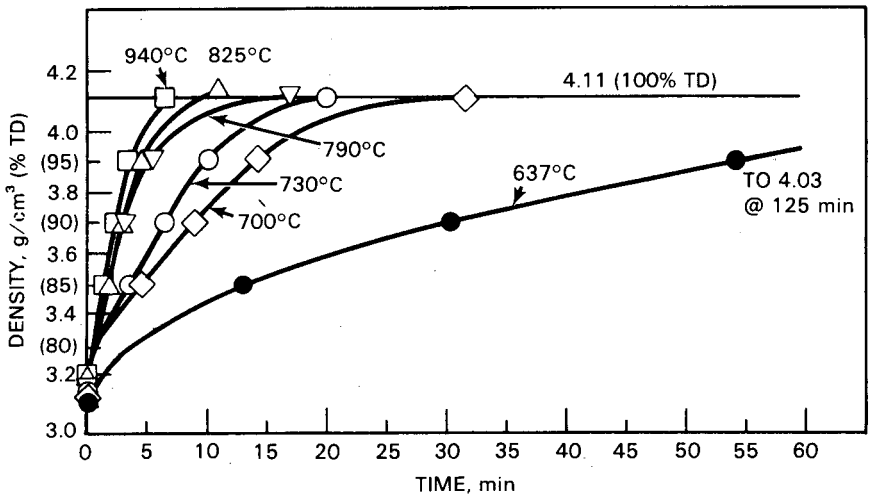


FIGURE 1.7. Effect of Temperature on Densification Rate of SrF_2

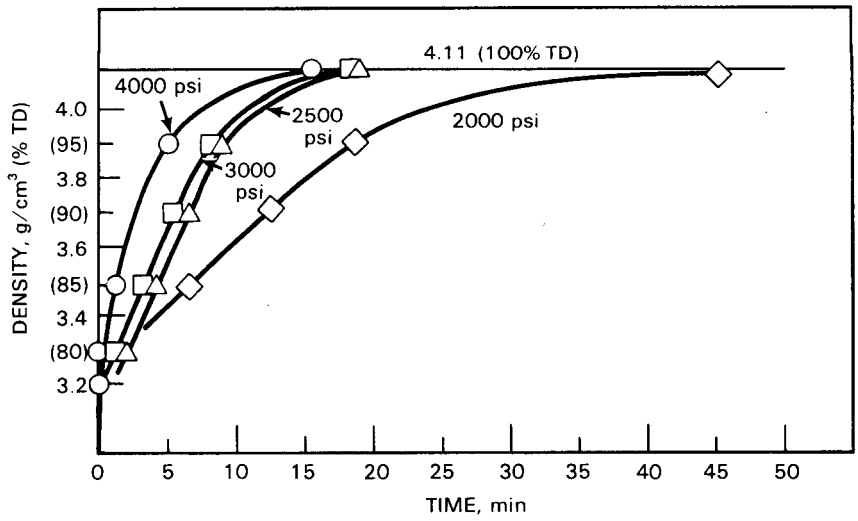


FIGURE 1.8. Effect of Pressure on Densification Rate of SrF_2

there is considerable incentive to develop more efficient conversion systems because of the limited supply of ^{90}Sr and the cost of producing ^{238}Pu. Thermoelectric devices currently available produce power very reliably, but the efficiency is limited to about 8%. On the other hand, thermomechanical conversion devices have been shown to produce power with efficiencies up to 38% and with the potential of even further increases. Table 1.13 illustrates the potential power available from fission-product strontium as a function of conversion efficiency.

One attractive heat engine considered for radioisotope-power conversion is the Stirling engine invented by a Scottish minister in 1818. Stirling engine use over the years has been limited, but development is reaching an advanced state. Free-piston Stirling engines have now been developed and operated for extended periods. One such engine, fueled

TABLE 1.13. Comparative Utilization of Strontium-90

Strontium Source	Amount,[a] kWt	Electrical Power Production, kWe: RTG 8% Efficient	RTMG 20% Efficient	RTMG 30% Efficient	RTMG 40% Efficient
Current B-Plant and WESF					
• $^{90}Sr/Sr$ >0.4	104	8.3	21	31	42
• $^{90}Sr/Sr$ 0.4 to 0.25	220	(b)	44	66	88
Future Hanford PUREX	230	18	46	69	92
Savannah River Plant					
• Present through 2000	620	50	124	186	248
Commercial Waste					
• Accumulated through 1981	2,500	200	500	750	1,000
• Accumulated through 2000	53,000	4,200	11,000	16,000	21,000

(a) As of 7-1-82.
(b) RTG usage generally limited to $\geq$0.4 $^{90}Sr/Sr$.

with radioactive strontium titanate, has operated for over 8 years without maintenance while producing power with an overall efficiency of 17 to 20%. In addition, other Stirling engine designs with conversion efficiencies in the range of 30 to 38% have been demonstrated. These machines have the potential for long-life, maintenance-free operation but currently have not been operated for extended periods.

A major objective of this program is to construct and test a radioisotope-powered converter that will produce power reliably with an efficiency of from 20 to 35%. Recently, contacts have been made with Stirling engine developers to obtain the necessary information to select the most promising system for use with radioisotopes. Heat-source design is also under way, and thermal, mechanical, and shielding calculations have been performed on a variety of systems. The most promising designs use efficient heat-transfer methods such as heat pipes to reduce the temperature difference between the isotope and the engine interface.

Various shielding designs are being evaluated, including the use of tungsten operating at high temperature inside the thermal insulation and the use of vacuum-foil insulation inside the biological shield with a less refractory shield operating at near-ambient temperature.

From these various concepts, an engine will be obtained and coupled to a heat source to provide an isotope-fueled converter for testing and demonstration.

Noble Metal Recovery and Use

Relatively large quantities of palladium, rhodium, and ruthenium are included in the fission products from irradiation of nuclear fuels. The high value and strategic nature of these metals provides considerable incentive to examine the potential for their recovery and ultimate use. Thus, a program was initiated during the summer of 1981 to

examine potential recovery techniques and to determine potential applications.

Current nuclear wastes at Hanford and Savannah River Laboratory are reported to contain about 1400 kg of palladium, 2000 kg of rhodium, and nearly 7000 kg of ruthenium. Significantly larger quantities are present in currently stored commercial spent fuels, with the potential for about 600,000 kg of these metals from commercial fuels between now and the year 2020.

Fission-product palladium is composed primarily of nonradioactive isotopes, with 17% being ^{107}Pd, a 0.035 MeV beta emitter with a half-life ($t_{1/2}$) of 45,000,000 years. Rhodium is almost entirely stable ^{103}Rh, with only a trace of radioactive ^{102m}Rh ($t_{1/2}$ = 206 days) and ^{102}Rh ($t_{1/2}$ = 2.9 years). Fission-product ruthenium contains several stable isotopes plus radioactive ^{103}Ru ($t_{1/2}$ = 39 days) and ^{106}Ru ($t_{1/2}$ = 367 days).

These metals have multiple uses as noble-metal catalysts, corrosion-resistant metallic components, and a variety of other applications. However, the presence of low levels of radioactivity requires consideration of the health effects that could result from utilization of these valuable byproducts. At least three categories of application need to be considered: 1) use in a radiation environment where radioactivity from noble metals is of no additional concern, 2) use in industrial applications where the materials are isolated from the environment and can be adequately controlled, and 3) unlimited application where no control is required. In the latter case, one must be concerned about any potential application which might use the product from recycle or reuse of the material. Thus the hazards involved from its use as jewelry or tooth or body implants, etc., must be evaluated.

Health Physics Assessment

The purpose of this task is to review the uses of reactor-produced noble metals and to estimate the radiation doses to workers and the general population from these uses. Our work has focused on reactor-produced palladium, particularly on the possible occupational and public exposure from ^{107}Pd that could result from uncontrolled use of the metal. The radiation doses considered include doses from objects outside the body, doses from metals inhaled as fumes and aerosols, and doses from surgical implants or dental prostheses.

In considering the dose from radiation sources external to the body, there are three organs or groups of organs for which an external dose is calculated: skin, lens of the eye, and the whole body. For calculation, these organs are assumed to lie at a depth of 70 μm, 0.3 cm and 1.0 cm, respectively. Specific dose limits have been established for these three organ systems: 5 rem, 30 rem and 5 rem per year, respectively. These depths represent the location of the radiation-sensitive tissues; radiation which does not penetrate to these depths is not considered to provide a meaningful dose. For example, the skin consists of two major layers, the dermis and the epidermis (the outermost layer). The epidermis is likewise composed of layers; beginning at the deepest, they are the stratum basale, stratum spinosum, stratum granulosum, stratum lucidum and stratum corneum (Goss 1973, ed.; Jensen 1976). The dose to the skin is significant only if the stratum basale (lying at a depth of 0.07 to 0.12 mm) is irradiated, because it is the source of new skin cells.

Beta particles generally have a short range in tissue, and with certain exceptions,

do not irradiate the whole body. Very low-energy beta particles, such as those of ^{107}Pd, will irradiate the outer layers of the skin, where the radiation dose may be large; but because it does not penetrate to the basal layer, the skin dose is insignificant. Many radionuclides, for example ^{99}Tc, will emit higher energy beta particles, which can penetrate to the basal layers of the skin. In these cases, the radiation dose as a function of depth into the skin must be computed.

Internal dosimetry refers to the calculation of a radiation dose to the organs of the body from radioactive material within the body. The internal radiation dose is a function of the energy of the emitted radiation, the amount of radioactive material in an organ, and the mass of the organ. Internal doses are calculated on an organ-by-organ basis and then can be summed to give a "whole-body" dose. The calculations assume that the radioactivity is uniformly distributed throughout the organ, although the various organs may have different concentrations. The time that the radionuclide will remain in the body is a function of its solubility in body fluids, and whether it is incorporated into tissues, for example, bone. Collectively, the distribution of the material within the body and its eventual excretion is referred to as the metabolism of the radionuclide. All the energy of a beta particle will contribute to the computed radiation dose because the radionuclide is surrounded by living cells. This is in contrast to external dosimetry, in which the beta particle must traverse nonliving skin before irradiating living tissue.

The scientific literature has been searched by on-line computer methods to identify papers that describe environmental concentrations of palladium from industrial processes such as roasting, smelting, and refining of the metal. Once environmental levels of palladium are known, then reasonable estimates of a radiation dose following inhalation of palladium at these levels may be made. Unfortunately, no estimates or determinations of the environmental levels of palladium, either local or in general, appear to have been made.

The calculation of radiation dose from a surgical implant or dental prosthesis is similar to external dosimetry in that the computations are those of an object in contact with the skin. However, the protective layers of dead cells found on the skin do not exist inside the body and may be only one-cell layer thick in the mouth (Bloom and Fawcett 1975). Thus palladium, whose betas will not penetrate dead layers of skin to cause an external dose, will irradiate living cells when used as an implant. The physical characteristics of ^{107}Pd, specifically the low energy of the beta particle and the low specific activity tend to diminish the radiation hazard of pure fission-product palladium when the palladium is contained in products external to the body; however, surgical implants may result in significant radiation doses to the surrounding tissues owing to the intimate contact and long residence times involved.

An approximate dose rate map, showing dose rate as a function of depth into tissue from a metal cylinder containing 20 at% ^{107}Pd, is shown in Figure 1.9. The map was prepared using techniques outlined by Brownell (Fitzgerald et al. 1967). The calculations assume an exponential loss of energy by the beta spectrum through the absorbing medium. As can be seen, the dose rate is very high at contact (~2 R/hr) but drops sharply at a distance of 10 to 20 μm. More work needs to be

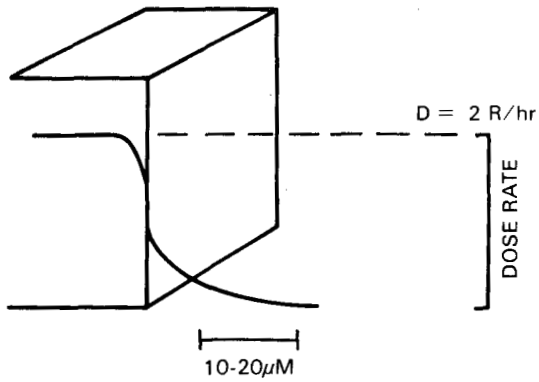


FIGURE 1.9. Dose Rate Map for Solid Pd (20% ^{107}Pd) in Contact with Tissue

done to identify the significance of this analysis relative to the long-term hazard.

One analysis has shown that irradiation due to ^{107}Pd from objects outside the body (i.e., rings, telephone contacts, noble-metal catalysts, etc.) is of no significance because live cells are protected by the thin epidermal layer. However, surgical implants containing fission-product palladium may cause significant doses from long-term exposure directly to body organs. Exposure from ingested dusts or aerosols can not be evaluated adequately without more information on the concentrations expected from processing of the metals or metal ores.

Recovery Methods

Several alternatives exist that might be applicable for the recovery of the strategic noble metals from waste streams. For serious consideration, these methods must be readily adaptable to the present processing or waste-management systems, must have relatively high decontamination factors and specificity for the metal ions recovered, and be able to be implemented at a cost not exceeding the value of the recovered product. Several specific recovery options that appear to meet these criteria include:

- Carbon sorbates: These are activated carbons that have been prepared by treating them with chelating compounds which react specifically with the palladium, rhodium, ruthenium, and technetium. In earlier work at PNL, greater than stoichiometric recoveries of noble metals from HCl solutions were observed. No work has been conducted to date on nitric acid solution.
- Reducing resins: Recently developed reducing resins are available that specifically reduce noble metals. While these have been tested and evaluated in mining operations, they have not been evaluated for nuclear applications; but it appears that the noble metals palladium and rhodium could be recovered at minimal cost in existing processing or waste-handling facilities.
- Precipitation of metals: It has been observed (Buxton and Campbell 1981) that significant quantities of palladium are precipitated with a sludge during sugar denitration of the dissolved fuel. While the existence of palladium, rhodium, and ruthenium in fuel processing sludge has been known for some time, their concentrations have never been determined. Such measurements are currently being planned under this program. If this process shows that the sludge contains relatively high concentrations of the noble metals, standard separation techniques can be used for their recovery at relatively low cost.
- Lead-extraction process: This process involves the extraction of noble metals by extraction into a molten-lead phase during vitrification of the waste product. This

technique has been described previously (Mellinger and Jensen 1982; Tingey 1982).

Recent results from in-can melter tests using the lead-extraction process have shown that near-quantitative separation of lead (and thus noble metals) from the glass occurs when the melt is held at 1100°C for 4 hr and the quantity of carbon reductant is minimized. These results were obtained from a series of experiments using large (1000 g) batches in an in-can melter.

The fabrication of the ceramic melter for continuous recovery of noble metals using lead extraction was completed (Figure 1.10). Ceramic melters are the reference vitrification units for the immobilization of high-level defense and commercial wastes. This small melter will be used to demonstrate that ceramic melters can be modified to provide for noble-metals recovery. Currently, high-level wastes and glass formers are vitrified in the melter box and the vitrified product is overflowed to a canister. In this melter, the simulated high-level waste glass former and lead oxide are vitrified in the melter box. The lead scavenges the noble metals and settles to the bottom of the box. A drain valve is operated intermittently to drain the lead-noble metals mix to a canister. The borosilicate glass, minus the lead and noble metals, is overflowed to a second canister.

References

Bloom, W. and D. W. Fawcett. 1975. A Textbook of Histology. W. B. Saunders Co., Philadelphia, Pennsylvania.

Buxton, S. R. and D. O. Campbell. 1981. U.S. Patent 4290267, "Process for Recovery of Palladium from Nuclear Fuel Reprocessing," September 22, 1981.

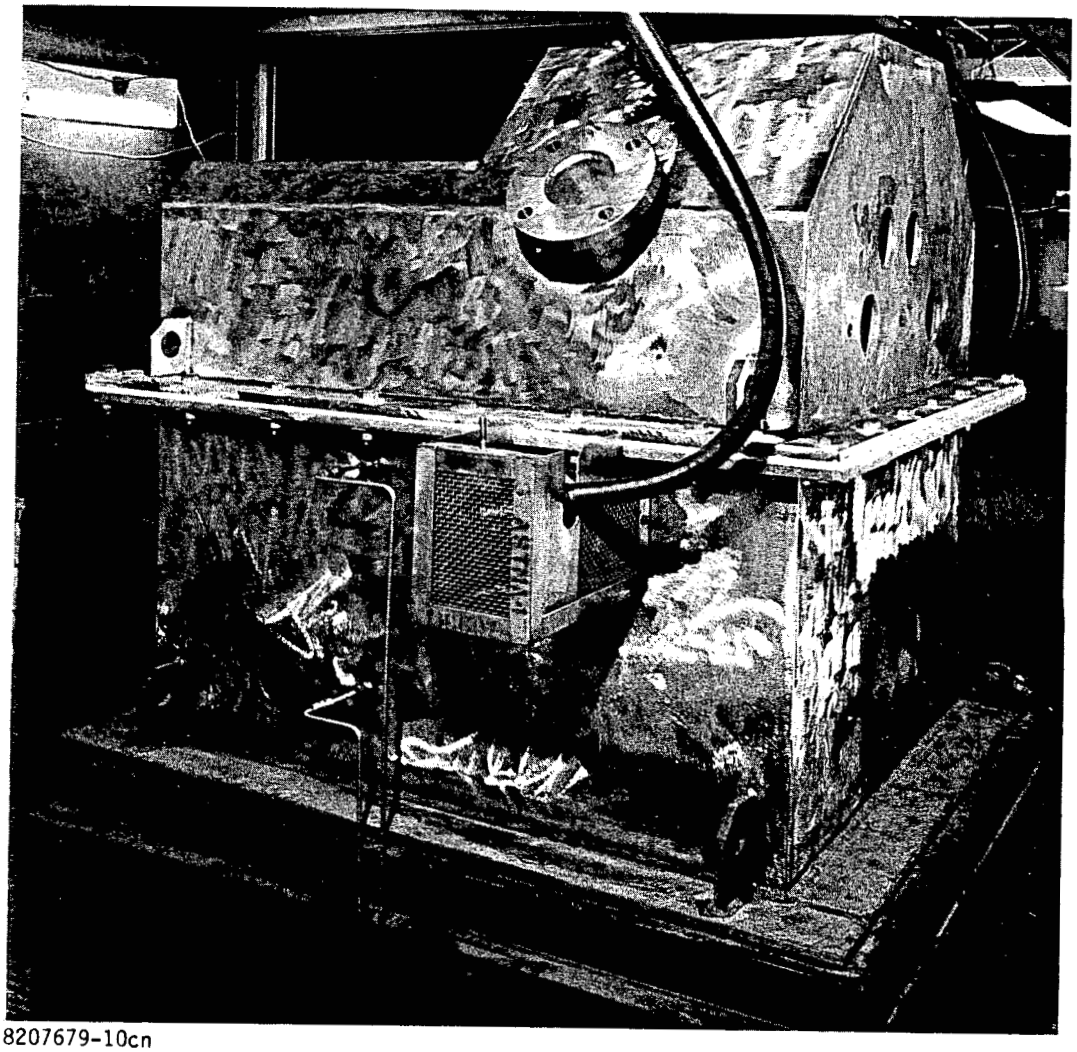
Fitzgerald, J. J., G. L. Brownell, and F. J. Mahoney. 1967. Mathematical Theory of Radiation Dosimetry, Gordon and Breach, New York, New York.

Goss, C. M., ed. 1973. Gray's Anatomy, Lea and Febiger, Philadelphia, Pennsylvania.

Jensen, D. 1976. Principles of Physiology, Appleton-Century-Crofts, New York, New York.

Mellinger, G. B. and G. A. Jensen. 1982. Preliminary Investigation of a Technique to Separate Fission Metals from Fission Product Mixtures. PNL-4116, Pacific Northwest Laboratory, Richland, Washington.

Tingey, G. L. 1982. "Defense Byproducts Utilization and Production Program," Nuclear Waste Management Semiannual Progress Report April 1982 through September 1982. PNL-4250-2, Pacific Northwest Laboratory, Richland, Washington.



8207679-10cn

FIGURE 1.10. Noble Metals Melter

2.0 MATERIALS CHARACTERIZATION CENTER

(AP-05-15-15/AR-05-15-10/DE-60-20-20 - 80145)

J. E. Mendel - Program Manager

Major Contributors: J. L. Daniel, M. D. Merz, D. M. Strachan, R. G. Strickert

SUMMARY

Increased emphasis is being given to the codification of test methods related to waste package performance in geologic repositories. Two new "umbrella" interaction test methods are being defined in conjunction with the repository waste package projects. One test method, designated MCC-14, is devoted to defining the radionuclide release from the waste form in site-specific geologic repository environments; thus there will be an MCC-14 (basalt), MCC-14 (salt) and MCC-14 (tuff). The other interactions test, MCC-105, is a repository-specific corrosion test designed to yield data that can be used in predicting canister and metallic overpack lifetimes in a geologic repository. Workshops were held in November 1982 and February 1983 to obtain input from the projects on MCC-14 and MCC-105, respectively.

Good progress was made in developing MCC reference and test materials. A contract for the manufacture of 500 lb of simulated waste reference glass, ARM-1, to be delivered in May 1983, was awarded to Schott Optical Glass. Four actinide-doped simulated waste glasses, ATM-1, 2, 3 and 4 were prepared for use as MCC-approved testing materials. Preparations were completed for manufacture of a fully radioactive glass. A draft report describing spent fuel that is available as testing material was distributed for comment by the projects.

Codification of waste-form test methods and data continued. At the end of the reporting period MCC-1 (Rev. 1), MCC-3, MCC-12, and MCC-D1 were under review by the Materials Review Board. In addition, an MCC Guidelines for Accuracy and Precision of Test Data was submitted for MRB review.

ADMINISTRATION

The MCC work breakdown structure was consolidated into four technical tasks plus an administration task on October 1, 1982. The reorganization was made to enable the MCC to better meet its increasing commitment to the National Waste Terminal Storage (NWTS) projects. Our reporting format is based on the new breakdown structure.

WASTE MATERIALS INTERACTIONS

MCC-14 Materials Interactions Test Method

To adequately assess the long-term integrity of a nuclear waste package system requires investigation of the reactions between the various components of the waste package (waste form, canister, backfill, and host rock) in contact with a potential groundwater. Although such a system is highly complicated, the various physicochemical mechanisms result in the possible release of radionuclides from the waste package system (source term) to the groundwater for

potential transport to the biosphere. Several test methods are being used by the candidate geologic repository site projects to provide data on such mechanisms as the resultant releases of radionuclides. The purpose of MCC-14 is to codify the test methods in order to reduce and quantify the uncertainty in the data needed for defining the radionuclide source term.

A Workshop on Materials Interactions Testing was held in Seattle on November 18 and 19, 1982, with representatives from the Materials Integration Office (MIO), MCC, and the repository site projects. The objectives were to exchange information on test requirements at each site, applicable methods and procedures, and reference material needs. In addition to the test methods submitted by each site, a set of reference conditions for the experimentally important parameters will be used in a benchmark test for each site. A flowchart of the development of the MCC-14 methods is shown in Figure 2.1. A summary letter report of the Materials Interactions Workshop was issued by the MIO.

Following the workshop, additional documentation of the test methods was submitted to the MCC from the sites. From this information and meetings with liaison personnel, draft outlines of each site test method were prepared. In February a draft MCC-14 test method (basalt) was distributed for review by MCC and BWIP staff. Draft copies of the salt and tuff versions of MCC-14 are being

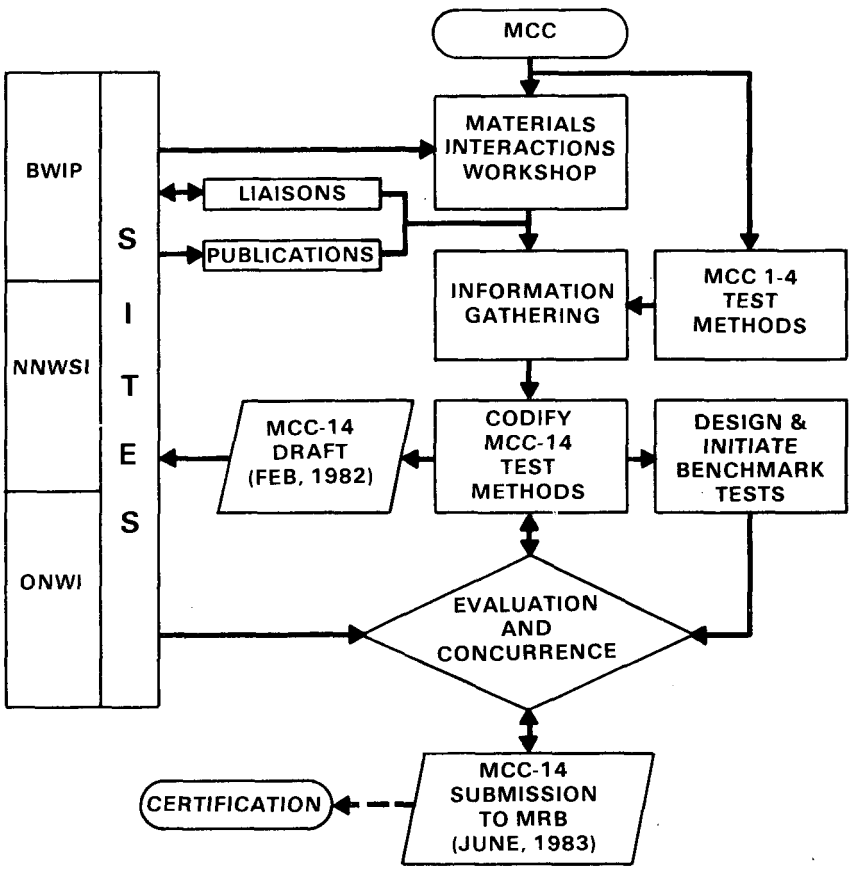


FIGURE 2.1. MCC-14 Development Task Flowchart

completed in March. Preliminary benchmark testing using the procedures is scheduled to begin in the third quarter. Final versions of the MCC-14 test methods are to be submitted to the MRB in June 1983.

The MCC-14 tests use agitated-batch or autoclave methods. When anoxic conditions are needed, the tests are run under an inert gas (argon) atmosphere. Rock and canister material, and synthetic groundwater, are those specified and in use by the individual site projects. Radioactive waste forms are to be supplied by MCC.

One specific concern in the MCC-14 tests was the need to measure the elemental solution concentrations at temperature. However, for temperatures greater than 100°C, this does not currently appear technically feasible. Modification of existing procedures for analyses of quenched solutions, following at-temperature sampling, was considered as a reasonable alternative. The estimation of at-temperature pH values is discussed in the modeling interface section below. Because some radionuclides of interest may exist in colloidal form at temperature and/or following the quenching process, measurement of the radionuclide activity in both filtered and unfiltered aliquots of solutions is proposed.

High-Temperature Solubility Methods

Efforts were begun to design and refine experimental procedures for measuring the equilibrium aqueous solubilities of radioactive nuclear waste components between 25 and 100°C. Procedures include the control and/or measurement of important test parameters (e.g., pH and the concentrations of dissolved constituents). Aqueous speciation reactions at the steady-state pH and concentration values may be determined using the MINTEQ geochemical model. The calculated activities of the aqueous species may then be used to derive solubility products for the test solid phases.

A reaction vessel has been assembled using a design similar to that used by Brush (Ph.D. Thesis, Harvard University, 1980) (Figure 2.2). The pH and temperature of the test solution can be monitored continuously. The solution is agitated with a magnetic stirrer and purged with water-saturated argon gas for removal of dissolved CO_2. Dissolved carbonate forms strong aqueous complexes with many dissolved cationic constituents. The reaction vessel is designed so that aliquots of the solution can be withdrawn periodically and filtered at temperature before being analyzed. Solid reaction products will be characterized by x-ray diffraction, electron microscopy and surface analysis techniques. Applicable experimental procedures from MCC-1, -2, and -3 test methods will also be added as the test method is developed.

The first set of solubility experiments is being conducted with schoepite ($UO_2(OH)_2 \cdot 2H_2O$) and dehydrated schoepite ($UO_2(OH)_2$). The uranium system was selected because:

1. Uranium is present in nuclear reactor wastes.
2. The solutions are not highly radioactive and thus easier to work with during developmental stages.
3. The uranium solubility data will be valuable for assessing maximum concentrations of uranium in groundwaters.

The U(VI) hydroxides were synthesized from the aqueous dissolution of solid UO_3. Concentrations of total dissolved uranium from 0.1-μm and 0.015-μm filtrates were measured by a laser-induced fluorescence technique.

During the 15 days of the first experiment, uranium concentrations up to ~305 mg/L

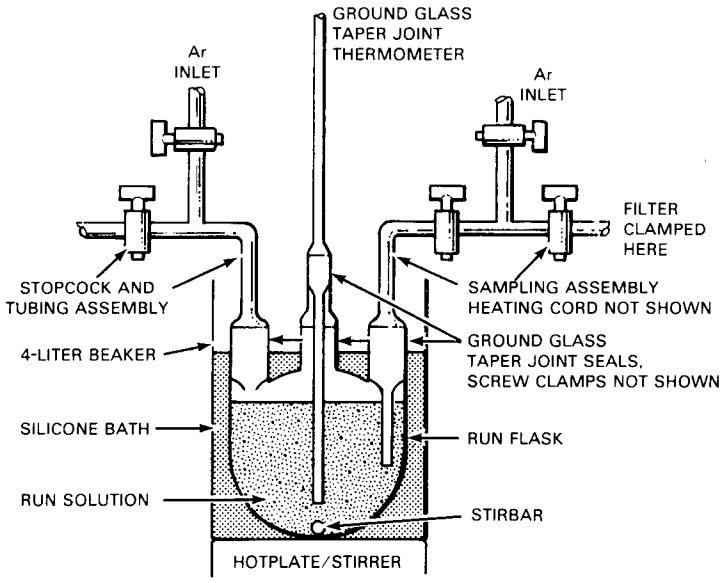


FIGURE 2.2. Apparatus for Schoepite Solubility Measurements

were measured at pH 4 and 25°C. However, small fluctuations (less than a factor of two) in the uranium concentration after this time are being investigated to determine whether a CO_2 leak is present in the system or there are errors in the uranium analysis methods.

Interfacing with Modeling Activities

The MINTEQ geochemical model was used to identify possible equilibrium solubility controls for solution compositions derived from 1-year MCC-1 leach experiments at 90°C using PNL 76-68 borosilicate glass. These modeling calculations also permitted an estimation of the pH at the temperature of the leach experiments.

The modeled solution analyses are those reported by Strachan (1982). The leachant solutions included deionized water (DIW), a silicic acid/sodium bicarbonate solution, and a concentrated K-Mg-Na-Cl brine. Possible solubility controls were calculated using the reported pH, Eh, and temperature values for the constituents Si, Zn, Pb, PO_4, CO_3, and Mo in the solutions. It is important to note that no fewer than 38 different aqueous species are needed to adequately calculate the aqueous speciation for the DIW solutions. Determinatiions of the possible solubility controls for the silicate and brine solutions have been computed and are included in a paper, "Solubility Interpretations of Leach Tests on Nuclear Waste Glass," by D. Strachan and K. Krupka, being presented at the American Chemical Society meeting in March.

The solubility controls for the leachates described above were calculated from pH values measured at room temperature. The reported values may not represent the pH (i.e., the activity of the uncomplexed H^+ species) of the leachates at the test temperature of 90°C, and thus may lead to some ambiguity in the calculated aqueous speciation and predicted solubility controls. Therefore, in-situ pH values were estimated for the DIW and silicate solutions by the method of mass balance of total ionizable hydrogen content (Truesdell and Singers 1974). The pH values

estimated by this method for the various leachates were ~0.9 to 1.0 pH units lower than those measured at room temperature. The calculation of solubility controls is affected significantly by using these estimated pH values. This is especially true for the silicate solids, for which solubilities depend on the speciation of the dissolved silica.

CORROSION

Test Method Development

MCC-101 Static-Immersion Corrosion Test Method. This test method was submitted to the MRB in the previous reporting period and was reviewed by the OTC-MRB (Office of the Chairman of the Materials Review Board) and returned to MCC for revisions, which are under way.

MCC-102 Flowing-Immersion Corrosion Test Method. This test method was submitted to the MRB in the previous reporting period and was reviewed by the OTC-MRB and returned to MCC for revisions, which are under way. Both MCC-101 and 102 are based on National Association of Corrosion Engineers and American Society for Testing and Materials practices.

MCC-103P Stress-Corrosion-Cracking Susceptibility Test Method. This test method was balloted by the full MRB and given approval for publication in the Handbook.

MCC-104S Stress-Corrosion-Cracking Susceptibility Test Method. This test method was submitted to the MRB during the last reporting period and was returned to MCC for revisions to clarify the relations of three separate procedures incorporated in the test method. A revised draft of the method was resubmitted to the MRB. Testing of 1020 steel according to the proposed MCC-104 techniques was started to further substantiate the method. Review of the test method by ASTM was also begun.

MCC-105 Test Method for General Corrosion in Repository Environments. This test method is directed toward obtaining data on metallic structural barriers in the presence of rocks or salt and repository-relevant groundwaters or brine. The method will specify benchmark conditions of temperature, radiation dose rate and oxygen level. The benchmark tests will provide a basis for comparing and evaluating data obtained under more varied conditions.

Workshop on MCC-105, February 3-4, 1982

A workshop for MCC and NWTS Waste Packages investigators from ONWI, BWIP and NNWSI (Office of Nuclear Waste Isolation, Basalt Waste Isolation Project, and Nevada Nuclear Waste Site Investigations) was held to obtain input for MCC-105 Test Method for General Corrosion in Repository Environments. Each site representative presented an overview of the site's test program and described test methods. The MCC offered a strawman MCC-105 consisting of a static corrosion test with benchmark conditions of temperature (90 and 150°C), radiation dose rate (0 and 10^5 R/L) and two oxygen levels, very low and air-equivalent saturation. The sites provided some immediate comments and generally accepted the approach. Further input will be obtained when the MCC provides each site with a draft for review later in the year. The subject of reference and test materials was discussed and there was general agreement

that Approved Test Materials (ATM) were appropriate for MCC-105. The ATMs would be single heats of commercially-available materials such as 1020 steel, TiCode 12 and 304L steel. The primary purpose of using ATMs in this case is to eliminate material variability as a factor in relating various data on corrosion and to establish credibility in a broad range of data taken under conditions other than the benchmark conditions of MCC-105.

Compilation of Corrosion Data

The report State-of-the-Art Report on Corrosion Data Pertaining to Metallic Barriers for Nuclear Waste Repositories, PNL-4474, by M. D. Merz, was issued in October 1982. The report presents a compilation of published corrosion data on metals that have been suggested as canister and overpack materials. The data are categorized according to the solutions used in testing: high-ionic strength solutions (such as seawater and brine) and low-ionic-strength waters (such as basalt and tuff waters).

WASTE FORM CHARACTERIZATION

MCC-1 Static Leach Test Method

Submittal of MCC-1R to the MRB

The MCC-1 Static Leach Test Method was resubmitted to the MRB. This version has been modified for use with highly radioactive waste-form specimens. Basically the changes require the use of leach vessels constructed of inert materials such as gold, platinum, or fused quartz and the use of inert atmospheres to suppress the generation of HNO_3.

Comparison of MCC Data with NBS Data

Accuracy and precision with the MCC-1 test method have been difficult to determine partly because of the lack of a standard on which to base a comparison. To enable such comparison the MCC obtained a sample of glass produced by the National Bureau of Standards (NBS). This glass has basically the same composition as MCC 76-68 glass but with the iron and chromium removed (MCC 1982). The glass is very homogeneous and the composition is accurately known. A set of five specimens of this glass was prepared as prescribed by the MCC-1 test method. An additional set of nine specimens was polished in the same manner as did NBS, using 600 grit SiC powder. All these specimens were leached at 90°C in deionized water as per the MCC-1 test method. The results from these specimens were analyzed statistically. Other than pH, only small statistical differences were found. The NBS had found that zinc was not a well behaved element and yielded data that were not reproducible (MCC 1982).

Table 2.1 compares the data from the set of nine specimens with those from NBS testing. The standard deviations in the MCC data are about a factor of two larger than those from the NBS and the means for the MCC data are on the average about 5% higher than those from the NBS. The 0.27 pH unit difference between the NBS data and the MCC data may also be linked to the high bias. High bias probably resulted from the temperature control, which was ±1°C at the MCC and ±0.04°C at the NBS. It is interesting to note that the pH obtained by the MCC under "air-equilibrated" conditions is higher than that obtained by the NBS under inert atmosphere. This is contrary to what would be predicted

TABLE 2.1. Comparison of MCC-1 Results from the NBS and the MCC

Element	NBS Mean Value, mg/L	NBS Standard Deviation	MCC Mean Value, mg/L	MCC Standard Deviation
B	1.37	0.025	1.32	0.051
Ba	0.0077	0.00025	0.0077	0.00034
Ca	0.138	0.0094	0.147	0.0081
Cs	0.431	0.0081	0.447	0.015
Mo	0.523	0.0093	0.526	0.0189
Na	4.45	0.170	4.84	0.364
Si	5.46	0.102	5.87	0.200
Sr	0.0310	0.0010	0.0320	0.0009
Zn	0.0245	0.00448	0.0310	0.0077
pH	9.34	0.04	9.70	0.02

if CO_2 from the air were a major concern (Barkatt, Barkatt, Pehrsson, Szoke, and Macedo 1981).

MCC-3 - Agitated-Powder Leach Test Method

This test method, plus supplemental material, was resubmitted to the MRB in March. The supplemental material contained results from tests performed with three particle size fractions of powdered MCC 76-68 glass at 90°C in deionized water, which show that the steady-state pH and the concentration of some elements, such as Cs, La, Nd, and Si, are a function of particle size. Particle size fractions of -10 + 40, -100 + 200 and -325 mesh were studied at 90°C with constant, but gentle, agitation. The steady-state pH decreased with increasing particle size (Figure 2.3). As the pH increased, an increase in the steady-state silica concentration was noted (Figure 2.4). Although steady-state silica concentrations were observed, the soluble elements, such as boron and sodium, did not reach steady-state. During the reactions a silica-rich gelatinous material formed, apparently with a composition much like that of the gel layer that normally

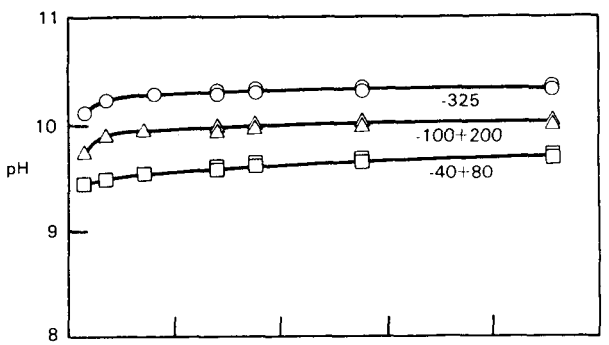


FIGURE 2.3. Effect of Particle Size on pH (MCC-3 Test Method, 76-68 Glass in Deionized Water at 90°C)

forms on this glass. Although the analyses are not yet complete, initial X-ray data indicate iron-zinc silicates with a silicate sheet structure analogous to clay and talc minerals.

REFERENCE MATERIALS AND ANALYTICAL METHODS

MCC-D1 One-Year Leach Test Data for PNL 76-68 Glass

Data package MCC-D1, first submitted on August 2, 1982, was revised to respond to detailed comments from the OTC-MRB. The revised data package was resubmitted to the

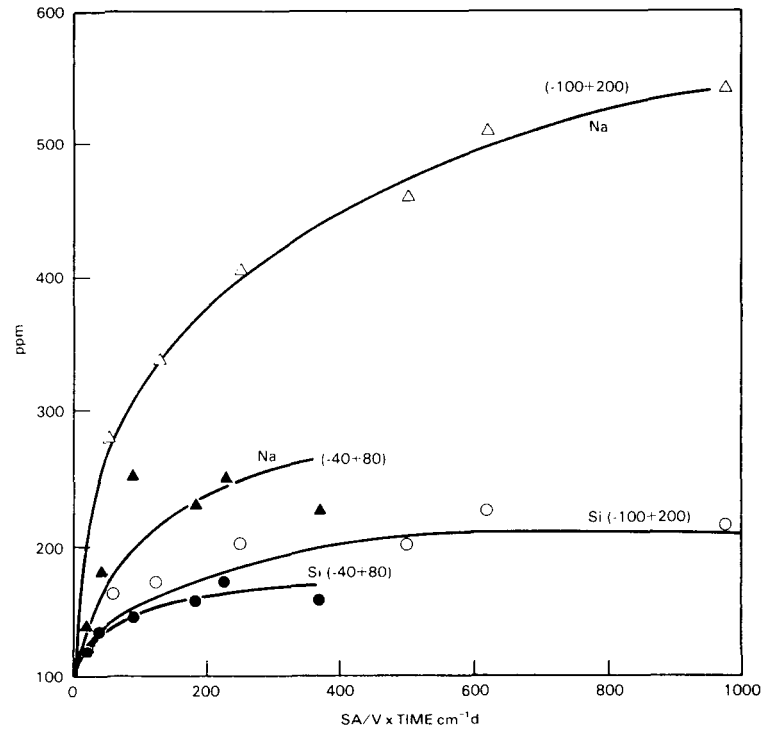


FIGURE 2.4. Effect of Particle Size on Leachant Composition (MCC-3 Test Method, 76-68 Glass in Deionized Water at 90°C)

OTC-MRB on January 20, 1983, and was supplemented with additional requested information on February 24, 1983.

The revised proposed chapter for the Handbook, "MCC Guidelines for Accuracy and Precision of Test Data," was reviewed by the OTC-MRB and returned to MCC with eight pages of comments. The guidelines were revised to accommodate the comments and resubmitted.

Installation of Instruments for Characterization of Radioactive Nuclear Waste Materials

The shielded analytical instruments for analysis of radioactive waste materials have been installed. Cold test runs and calibrations of the SEM with its x-ray fluorescence analyzer and of the inductively coupled plasma emission spectrometer (ICP) have been conducted satisfactorily. Experience with analysis of radioactive particulate specimens on the shielded x-ray diffractometer (XRD) showed that additional specimen transfer protection was desirable; the necessary modifications are under way on the XRD, and were added also to the SEM. The commitment of the ICP and SEM to fully radioactive materials has been delayed by problems in supplying the necessary building services of drains for radioactive liquid waste and ductwork for filtered air ventilation. Meanwhile, the SEM has been used successfully on low-level reactor corrosion product specimens.

Reference Materials

ARM-1

A contract for manufacture of 500 lb of simulated waste reference glass ARM-1 was

awarded to Schott Optical Glass, Duryea, Pennsylvania. Schott has obtained all necessary components for the glass, and has prepared a small test run satisfactorily. Delivery of the glass to the National Bureau of Standards for evaluation is scheduled for May 1983.

The contract with NBS for assistance in obtaining ARM-1 was put in place. NBS representatives have played an active role in selecting the reference glass specifications and in consulting with Schott and the MCC regarding preliminary details of the glass preparation. NBS has also supplied Schott with a laboratory specimen of glass of similar composition and will continue to advise Schott as necessary to assist them in meeting MCC specifications.

ATM-1, 2, 3, 4

Four doped simulated waste glasses have been prepared for use as MCC-approved testing materials. All four were made by adding appropriate quantities of dopants to existing waste glass type MCC 76-68. The specifications of the glasses are given in Table 2.2. Detailed analysis and characterization of these glasses is in progress.

Fully Loaded Radioactive Reference Glass

Preparations have been completed to manufacture about 4000 g of fully loaded radioactive waste glass for use as an MCC-approved testing material. Existing Nuclear Waste Vitrification Project waste solution at PNL will be calcined and converted to glass of a composition based on the Barnwell process. The specific composition will be that being recommended by March 31, 1983, from PNL's glass development work in progress in the Commercial High-Level Waste Program. Cold test runs and calcination of the liquid waste are scheduled for March 1983.

Reference Spent Fuel

A draft of a report discussing criteria for selection of reference spent fuels for use as ATMs and summarizing fuels available for that purpose was completed and is undergoing internal review. It concludes that the H. B. Robinson and Turkey Point fuels are both acceptable representatives of the currently most abundant type of spent fuel, namely, moderate burnup, fragmented UO_2 pellets with low fission-product release. A similar conclusion was reached in informal

TABLE 2.2. Specifications of Approved Testing Materials (All Prepared from MCC 76-68 Simulated Waste Glass)

Designation	Dopant	Dopant Concentration, wt%	Glass Quantity, g
1A	UO_2 (depleted)	4.0	4000
1B	UO_2 (normal)	3.98	430
2	Am_2O_3	0.064	410
3	NpO_2	0.306	440
4	PuO_2	0.062	430

discussions between MCC and local NWTS project representatives.

The selection of a specific fuel for the MCC reference testing material will depend on characterization information available for each spent fuel type, the amount and form of available materials, and the cost of acquisition by MCC. Also of concern are the effects of post-irradiation treatment and storage already experienced by the fuel (size of sectioned pieces, storage atmosphere, present physical condition of available material, etc.). Each NWTS project has been asked to submit a summary of the types of material characterization they consider particularly important in evaluating their planned repository-specific tests.

REFERENCES

Barkatt, A., A. Barkatt, P. E. Pehrsson, P. Szoke and P. B. Macedo. 1981. "Static and Dynamic Tests for the Chemical Durability of Nuclear Waste Glass." Nuclear and Chemical Waste Management 2:151-164.

Bush, L. H. The Solubility of Some Phases in the System UO_3-Na_2O-H_2O in Aqueous Solutions at 60 and 90°C. PhD. thesis, Harvard University, Cambridge, Massachusetts.

Materials Characterization Center. 1982. Workshop on the Leaching Mechanisms of Nuclear Waste Forms, May 19-21, 1982. PNL-4382, Pacific Northwest Laboratory, Richland, Washington.

McVay, G. L. and L. R. Pederson. 1981. "Effect of Gamma Radiation on Glass Leaching." J. Amer. Ceram. Soc. 64(3):154-158.

Palmer, R. A., et al. 1982. Characterization of Reference Materials for the Barrier Materials Test Program. RHO-BW-ST-27P, Rockwell Hanford Operations, Richland, Washington.

Strachan, D. M. 1982. "Results from the Long-Term Use of the MCC-1 Static Leach Test Method." PNL-SA-10762. Pacific Northwest Laboratory, Richland, Washington. (Accepted for publication in Nuclear and Chemical Waste Management.)

Strachan, D. M. 1983. "Effect of Gamma Irradiation on Teflon® PFA and on Simulated Waste Glass Leaching in Teflon PFA," PNL-SA-11092. Pacific Northwest Laboratory, Richland, Washington. (Submitted to J. Amer. Ceram. Soc.)

Truesdell, A. H. and W. Singers. 1974. "The Calculation of Aquifer Chemistry of Hot-Water Geothermal Systems." J. Res. U.S. Geol. Surv. 2:271-278.

®Registered trademark of E. I. du Pont de Nemours Co., Inc., Plastic Products and Resins Department, Wilmington, Delaware.

2.0 NUCLEAR WASTE MATERIALS CHARACTERIZATION CENTER

The objective of the Nuclear Waste Materials Characterization Center (MCC) is to provide a data base that will include information on the components of the waste emplacement package (the spent fuel or processed waste form and the engineered barriers), their interaction with each other, and the effects of their environment. The MCC develops test methods and obtains materials data as necessary and submits these to the Materials Review Board (MRB). After test methods and data have been approved by the MRB, the MCC publishes the approved methods and data in the Nuclear Waste Materials Handbook.

3.0 WASTE ISOLATION

The objective of the Waste Isolation Program at PNL is to support the National Waste Terminal Storage Program's efforts to develop the technology and facilities required for the safe and environmentally acceptable disposal of nuclear waste.

3.0 WASTE ISOLATION

M. R. Kreiter - Program Office Manager

Program Managers: D. J. Bradley, C. R. Cole, L. L. Clark, R. A. Craig, M. R. Kreiter, R. F. McCallum

Major Contributors: L. L. Ames, T. Bander, J. C. Barnard, J. O. Barner, F. W. Bond, M. O. Cloninger, D. G. Coles, M. J. Danielson, R. Ecker, C. A. Geffen, K. S. Goodman, F. N. Hodges, C. Kincaid, W. L. Kuhn, G. L. McVay, B. Napier, G. Petrie, T. B. Powers, S. Simmons, S. C. Sneider, R. E. Westerman, K. M. Yasutake

3.1 OFFICE OF NUCLEAR WASTE ISOLATION (ONWI)

3.1.1 Waste Package Program (DA-30-30-01 - 80579)

The main objective of the Waste Package Program is to help provide the necessary data base on waste package components and multicomponent interactions to qualify a waste package for geologic disposal. Supporting this are the following more specific program objectives: 1) help provide a basis for meeting waste package performance standards; 2) provide input to waste package designers; 3) optimize and select structural barrier materials; 4) develop predictive models for waste package degradation and radionuclide release.

Summary

The following conclusions and observations can be drawn from work conducted during the reporting period:

- After 1 month of salt brine exposure (without gamma irradiation), the corrosion rates of cast ferrous materials (1025 steel; 2 1/2% Cr, 1% Mo steel; and ductile cast iron) in anoxic Permian Basin salt brine at 150°C lie in the range of 0.48 to 0.74 mils/year. These samples show only a magnetite (Fe_3O_4) corrosion product film.
- After 1 month of salt brine exposure (in a gamma irradiation environment) the corrosion rates of 1025 wrought steel; 2 1/2% Cr, 1% Mo steel; and 1025 cast steel lie in the range of 2.9 to 7.0 mils/year. This represents an approximate tenfold increase over the nonirradiated test data. The irradiated environment specimens have corrosion product films that are mixtures of magnetite and anhydrite ($CaSO_4$).
- Slow-strain-rate tests of ductile cast iron and 2 1/2% Cr, 1% Mo cast steel showed that these materials are susceptible to environmentally enhanced cracking in salt brine at 150°C. A 1025 wrought steel, however, shows essentially no difference whether in brine or air tests.
- The effect of ductile iron on glass leaching characteristics in salt brine is less than in basalt or tuff groundwaters.
- Gamma irradiation causes significantly enhanced leaching of glass in the presence of iron due to increased reactions of the iron, resulting in iron-silicate colloid formation.
- The effects of ductile iron on spent fuel leaching in salt brine are not large based upon solution analysis. However, the amount of elements sorbed on the iron may be large, suggesting increased removal of elements from the spent fuel. Analyses of iron sorption are not yet complete, however.

- Gamma radiolysis of Permian Brine results in high generation of gas pressure (>80 atmospheres). This gas is ~70% hydrogen and cannot be explained by current literature or experimentation. It appears to be the result of synergistic effects between brine constituents.
- At 150°C the presence of iron or titanium metal has little effect on the leaching of 76-68 glass in Permian Basin salt brine.
- Tests with Permian Basin salt brine at 250°C indicate decreased concentrations of Ca^{2+}, Mg^{2+}, SO_4^{2-} in solution, apparently a result of precipitation of calcium and magnesium sulfates. The change in solution compositions at 250°C does not change solution pH (measured at 25°C).
- Based on mass-transfer analyses in flowing leach tests, it should not be assumed that the leachate is well mixed next to the waste form.

Program Management

Program administration has focused on the following areas:

- The FY 1983 WPAS and TPP for the Waste Package Program were reviewed, finalized and sent to ONWI on schedule.
- The Waste Package Program for FY 1983 was presented to PNL staff.
- Experiment test procedures being used on WPP waste package interactions testing were formalized and transmitted to S. J. Basham and staff at ONWI.
- The statement of work for the subcontract with Lawrence Livermore National Laboratory (LLNL) was rewritten to reflect the FY 1983 work scope. The subcontract has completed the PNL procurement approval process and was transmitted to DOE-RL for approval.

Liaison activities have included the following:

- WPP staff participated in the Nuclear Waste Materials Characterization Center (MCC) program review.
- A presentation by D. Jackson (LLNL) on his PROTOCOL model was attended by WPP personnel.
- A presentation of the ONWI Program Management System was made by L. Lambert to PNL personnel.
- Mutual waste management research was discussed between Andrea Saltelli, Ispra, Italy, and WPP staff.
- WPP staff (D. Bradley, D. Coles, W. Kuhn, G. McVay, J. Nelson, and R. Westerman) attended, and W. Kuhn and G. McVay gave presentations, at the Materials Research Society annual meeting in Boston.
- A presentation was given to the Materials Department ceramics research section on ONWI-sponsored waste package studies by D. Bradley.
- D. Bradley attended the NWTS/MCC waste package interactions testing workshop in Seattle and presented details of ONWI-sponsored waste package interactions testing procedures.
- A presentation to DOE-RL-BWI staff on waste package studies was given by D. Bradley.
- D. Bradley presented a review of WPP progress to L. Olson and his staff at DOE-RL-BWI.
- WPP staff, coordinated by G. McVay, met with Dr. R. B. Heimann of the Fuel Waste Technology Branch at Whiteshell, Canada.

- D. Bradley attended the NWTS annual information meeting in Las Vegas, Nevada, and toured the Nevada Test Site.

Structural Barriers

General Corrosion Studies. A 150°C general corrosion study on two cast steels (1025 and 2 1/2% Cr, 1% Mo) and ductile cast iron was shut down after 1 month exposure for specimen analysis. The environment was anoxic (~0.2 ppm O_2) brine, designated Permian Basin Salt Brine No. 1, which closely resembles the formulation obtained by direct dissolution, to saturation, of a Permian Basin salt core. A serious problem, however, has been encountered in its use: the autoclave inlet line plugs every few days due to the retrograde solubility of certain brine constituents. To circumvent this operational problem, a new formulation, Permian Basin Salt Brine No. 2, was made, based on the effluent brine composition of an autoclave operating at 150°C, and presumably having the precipitable compounds removed from solution. The major difference between Brines No. 1 and 2 is a reduction in the latter of the Ca^{++} and $SO_4^{=}$ concentrations. Brine No. 2 will be used in the general corrosion study as well as in the irradiation-corrosion tests of steels in anoxic, 150°C brine that have been recently initiated. If plugging persists, the Brine No. 2 will be diluted slightly. Such modifications are not expected to significantly affect the observed corrosion mode or the corrosion kinetics.

The 1-month exposure in Permian Basin Salt Brine No. 1 produced the corrosion rate data shown in Table 3.1.

Interestingly, the corrosion rates observed are about ten times lower than those found by Sandia investigators studying corrosion rates of ductile cast iron in anoxic WIPP Brine A (SNL Monthly Progress Report for High-Level-Waste Interactions - August 1982).

X-ray diffraction analysis of the corrosion product films revealed only the presence of magnetite, Fe_3O_4. One specimen showed a faint trace of anhydrite, $CaSO_4$.

Summary duplicate-specimen data for 6-month exposure analysis of a ductile cast iron and two cast steels in oxic (0.7 ppm O_2) 250°C deionized water are given in Table 3.2. Data from 1 and 3 months are included for comparison.

These corrosion data show a regular, well-behaved diminution in corrosion rates with increased time of exposure. Corrosion products after 6 months exposure consist primarily of Fe_3O_4.

Irradiation-Corrosion Studies. An irradiation-corrosion study was shut down for specimen analysis after 1 month exposure. Investigated were 2 1/2% Cr, 1% Mo cast steel; 1025 wrought steel; and 1025 cast steel in a 150°C Permian Brine No. 2 environment at a ^{60}Co gamma irradiation

TABLE 3.1. Ferrous Materials Corrosion in mils/yr[a]

Temperature	Time	2 1/2% Cr, 1% Mo Steel	1025 Cast Steel	Ductile Cast Iron
150°C	1 month	0.48, 0.49	0.68, 0.61	0.74, 0.59

(a) Data from duplicate specimens presented.

TABLE 3.2. Summary of Duplicate-Specimen Penetration Data, mils/yr

Time, months	Cast Steel 1 1/4% Cr, 1/2% Mo	Cast Steel 2 1/2% Cr, 1% Mo	Cast Ductile Iron
1	0.44, 0.44	0.53, 0.44	0.38, 0.41
3	0.31, 0.29	0.40, 0.38	0.30, 0.31
6	0.25, 0.22	0.27, 0.23	0.23, 0.22

intensity of ~1 x 10^5 rad/hr. The corrosion rates observed are about ten times those obtained under nonirradiated test conditions. The data obtained from duplicate specimens are summarized in Table 3.3.

One specimen of each material type was selected from near the top of the autoclave and one from near the bottom. In Table 3.3, the first entry corresponds to those from the bottom. It can be seen that all the lower-corrosion-rate specimens came from near the bottom of the autoclave. Within groupings, however, the relationship between the alloys remained the same. The variation of corrosion rates with position is most likely due to a temperature gradient within the autoclave. Autoclaves in the radiation facility have consistently run 50°C colder at their inlet (at the bottom) than their control temperature (at the top). A higher heater-element winding density near the autoclave bottom will be tried in an attempt to alleviate this problem.

X-ray diffraction analysis of the surface of the specimens revealed only the presence of magnetite, Fe_3O_4, and anhydrite, $CaSO_4$. The anhydrite and magnetite layers appear to be intergrown. As previously noted, anhydrite was generally not observed in the corrosion product films formed under nonirradiated conditions.

Enviromechanical Studies. Recent efforts have been directed toward slow-strain-rate testing of wrought 1025 steel and construction of a fatigue-crack-growth-rate system for hydrothermal operation.

Previous slow-strain-rate testing indicated that 2 1/2% Cr, 1% Mo cast steel is susceptible to environmentally enhanced cracking in brine at 150°C. The reduction of sample cross-sectional area (upon failure) was diminished in the brine environment, and secondary cracking was observed through the necked region of the specimens, even when tests were conducted at relatively high strain rates (10^{-4}in./in.-sec). Similar tests of ductile cast iron, tested in the as-cast condition, indicated that it was also susceptible to environmental effects and that the ductility was low regardless of the environment.

Slow-strain-rate testing of 1025 steel has begun at 150°C and a strain rate of

TABLE 3.3. Irradiation-Corrosion Data Summary

Material	Penetration, mils/yr
2 1/2% Cr, 1% Mo Cast steel	2.9, 4.9
1025 Wrought steel	3.9, 6.2
1025 Cast steel	4.0, 7.0

10^{-4}in./in.-sec. Specimens with LT orientation have been tested in air and in brine; specimens with TL orientation have been tested only in air. Results of the tests to date indicate that this material is much less susceptible to environmentally enhanced cracking than is cast 2 1/2% Cr, 1% Mo steel and that the ductility of the wrought steel is far superior to that of cast ductile iron. No difference was observed in the behavior of the 1025 steel in air and in brine, and secondary cracking was not found. The corrosion product film was black and coherent. In contrast, the surface of ductile iron exposed to 150°C brine contained discontinuities around graphite nodules, where localized corrosion appeared to be starting.

Source Term Evaluation

Glass Interactions. Previous interactive tests between 76-68 glass and ductile iron in silicate groundwaters at 90°C have shown a large increase in elemental release. This effect is due primarily to the formation of iron-silicate precipitates that scavenge the leachate and prevent saturation effects. Thus the presence of ductile iron significantly enhances (by a factor of five) the elemental leaching of the glass by retarding back reactions.

When these glass/ductile iron interactive tests are conducted in Permian Basin Salt Brine No. 1, a similar (but much reduced) effect occurs; i.e., the glass leaches at a slightly enhanced rate (less than a factor of two) and precipitates form in the leachate. These precipitates are difficult to analyze--both because they are not formed in large quantities and because precipitates also form just from heating a saturated salt brine. Therefore, many types of precipitates are forming but not in large enough quantities to significantly affect glass leaching.

Adding gamma irradiation to the interaction tests enhances the leaching of 76-68 glass in a salt brine environment, particularly when iron is present. This appears due, at least partially, to the accelerated reaction of iron with the brine, which causes more colloids to form and, thus, more scavenging of the leachate, with resultant retardation of back reactions. Presently, the gamma irradiation effects have been evaluated only at 50°C, where enhancement factors of about two are observed. Higher temperature experiments are under way.

Spent Fuel Interactions. Static interactive tests involving spent fuel and ductile iron in 25 and 75°C Permian Basin Salt Brine No. 1 have been conducted to times of 60 days. Similar to the glass results, the effect of ductile iron on spent fuel leaching is not large in salt brine. One exception is technetium, which is reduced about one order of magnitude in the leachate when ductile iron is present. These results are all based on solution analysis and do not include elements sorbed on the ductile iron. The analyses of elements on the iron will be completed during the next month and should allow a more meaningful interpretation.

Currently, a series of experiments has been initiated utilizing unirradiated UO_2 fuel pellets, ductile iron, and oxidized Zircaloy.® These experiments will allow surface analyses to be conducted on all three constituents and will, it is hoped, lead to a better understanding of the interaction processes. The results from these

®Registered trademark of Westinghouse Electric Corporation Specialty Metals Division, Blairsville, Pennsylvania.

experiments should lead to improved design and better understanding of future spent fuel experiments conducted on clad fuel segments.

Colloid Investigations. Many types of colloids are formed in interactive tests conducted in salt brines, complicating the investigation of colloid formation and characterization. The approach is to investigate the formation, sorption, desorption, and stability characteristics of many candidate colloids, such as silica, iron hydroxide, iron silicates, and alkaline earth sulfates in brines, and to compare their behavior to that observed for colloids formed in realistic interaction tests. This understanding should help in interpreting the behavior of the complex matrix of colloids formed in salt brine interaction tests.

Another important aspect of colloids is their size distribution. Smaller colloids will be more mobile than large ones, and thus their size distribution under various conditions should be investigated.

The actinide sorption, desorption, and stability of silica, iron hydroxide, iron silicate, and calcium sulfate colloids in salt brines are now being investigated. Additionally, a hydrodynamic chromatography unit has been ordered that will determine particle size distributions of colloids.

Brine Radiolysis. If brine migrates into the repository during the thermal pulse period, it will be exposed to gamma irradiation. After the canister is breached, brine will be exposed to alpha radiation. Therefore, an understanding of the behavior of brine in both alpha and gamma irradiation fields becomes important. Currently, only results from gamma radiolysis are sufficiently complete to report.

Figure 3.1 compares the pressures resulting from gases generated during gamma

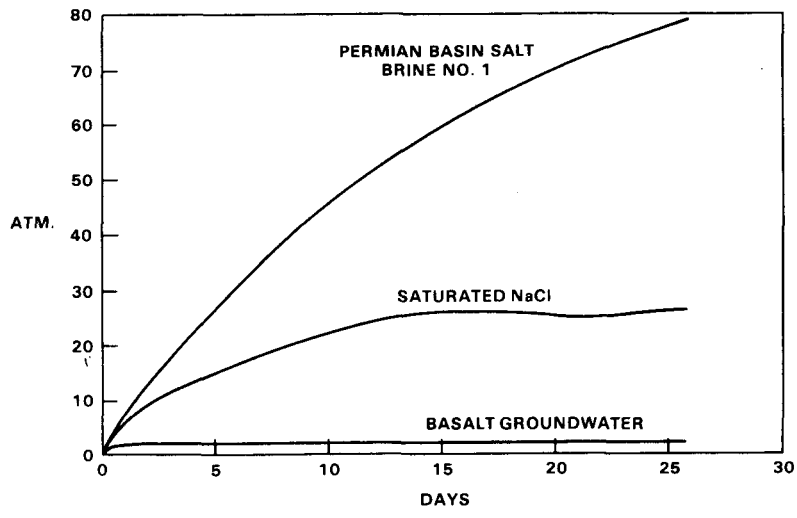


FIGURE 3.1. Time Dependence for Pressure Generation of Gas Produced by Gamma Irradiation of 70°C Permian Basin Salt Brine No. 1, Saturated NaCl, and Reference Basalt Groundwater. The solution volume was 10 mL and the gas phase volume 6 mL. A dose of 10^7 rad/hr was used in a ^{60}Co facility.

radiolysis of Permian Basin Salt Brine No. 1 to those from saturated NaCl solution and basalt groundwater. These results were obtained by irradiating the solutions at ~70°C in fused silica containers (19 mL solution - 6 mL gas phase) under continuous monitoring of pressure. After radiolysis, the gas phase was analyzed with a mass spectrometer. As can be seen from Figure 3.1, the Permian Basin brine generates much more pressure than does saturated NaCl solution or basalt groundwater. The pressures from basalt groundwater and saturated NaCl solution saturate at about 2 and 25 atmospheres, respectively, while the Permian Basin brine has reached 80 atmospheres and is still increasing. The gas phase consists of 75% hydrogen and the rest is primarily oxygen.

It is the saturation or equilibrium pressure that is of interest; it is important in considerations of repository and waste package design and also helps in the understanding of the mechanistic details of solution radiolysis. The time and dose dependence of the pressure buildup is dependent upon the volume available for the gas phase. With less volume, the dose and time required to reach equilibrium pressure is decreased. If there were essentially no volume available for the gas phase, then the equilibrium pressure would be reached very rapidly and with only a small dose. The dose rate has been shown to have no effect on gas generation from 10^7 to 10^4 rad/hr.

The relatively high gas generation in Permian Basin brine is currently unexplainable based upon the literature and recent experimentation. As seen from Figure 3.1, saturated NaCl solutions yield a maximum pressure of ~25 atmospheres under gamma irradiation. The Permian Basin Salt Brine No. 1 contains ~30 ppm Br^-, which is reported to enhance hydrogen generation. The addition of 30 and 300 ppm Br^- to deionized water results in enhanced hydrogen generation but falls short of explaining the results with Permian Basin salt brine. Therefore, a synergistic effect between constituents in the salt brine or some other trace impurity effect must be causing recombination reactions to be bypassed. The formation of this quantity of hydrogen gas indicates a strongly oxidizing environment in the solution. Analyses of other radiolysis products have not yet been made. Additionally, longer times are needed to reach saturation pressures for the Permian Basin salt brine and 300 ppm Br^- solutions.

Component Compatibility

Multicomponent Testing. Initial interactions tests with Permian Basin salt brine at 150°C were started last fiscal year. These tests were carried out according to the MCC-2 procedure (28 days) and with 76-68 glass and 76-68 glass plus ductile iron or titanium grade 12. The solutions used included both Permian Basin Salt Brine No. 1 and a saturated (at 25°C) brine prepared directly from Permian Basin salt core (G. Friemel Hole No. 1). All tests were carried out with a glass SA/V of 10 m^{-1} and, in tests containing iron or titanium, a glass-to-metal surface area ratio of 1:1. Solution analyses were carried out by ICP.[a]

Figures 3.2 and 3.3 are computer-generated plots of ICP data for boron, molybdenum, cesium, and silicon (without individual data points). Figure 3.2 presents leach data for 76-68 glass in four solutions, where it is seen that dissolution rates for brines are

(a) Inductively coupled argon plasma emission spectroscopy.

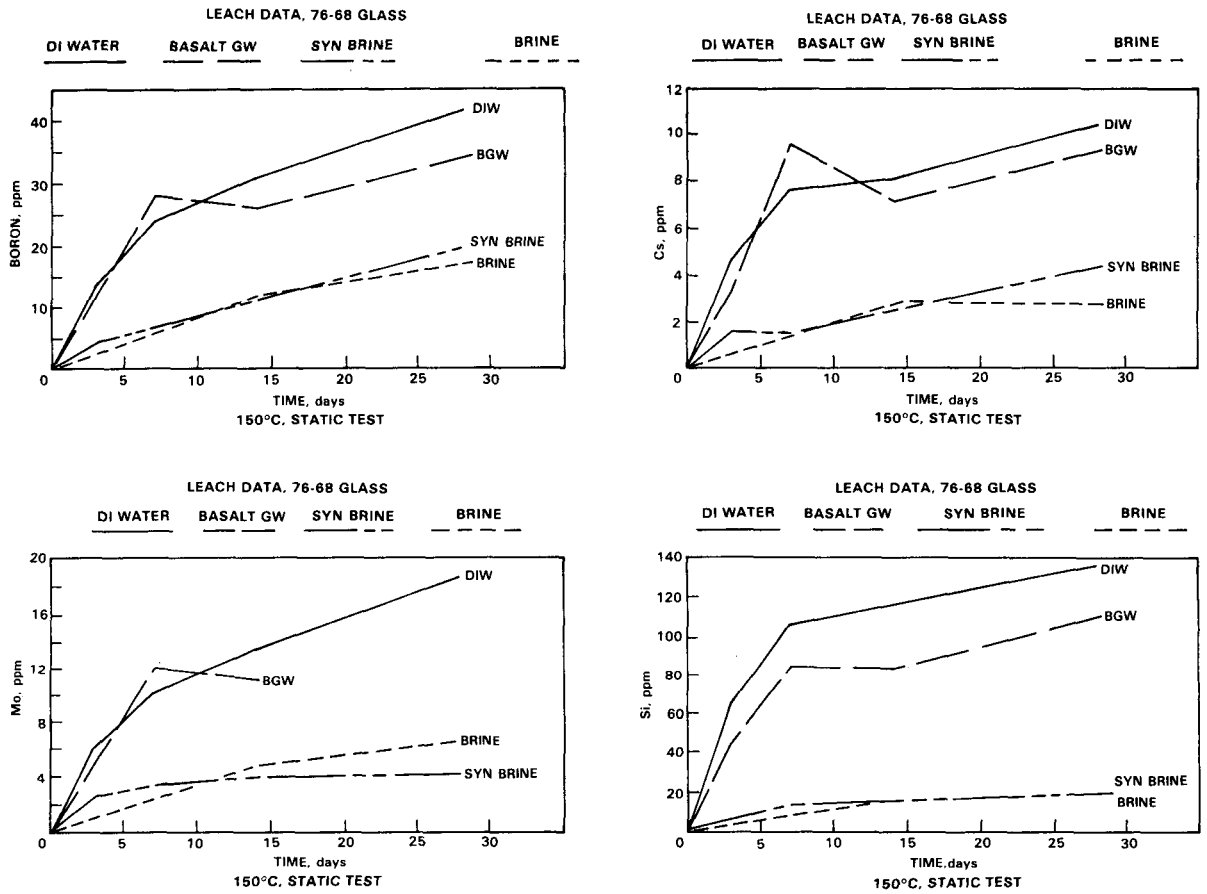


FIGURE 3.2. Plots of Solution Composition Versus Time for Four Leachates: Deionized Water, Reference Basalt Groundwater, Permian Basin Salt Brine No. 1 (SYN BRINE), and Permian Basin Salt Brine Made from Actual Salt Core

considerably lower than for deionized water and low ionic strength basalt groundwater. Gel layer thicknesses, measured by scanning electron microscope, show a similar decrease in thickness for glasses leached in brines. Molybdenum and silicon appear to approach steady-state concentration in brines by 28 days; however, cesium and boron increase in concentration at a relatively uniform rate thereafter. Observed leaching differences between brines and lower ionic strength solutions may be a result of several mechanisms, including changes in solubilities with increasing ionic strength and changes in the solid phases formed in a brine environment. Evidence for the first is provided by Marshall (1980a), who reports an approximate 50% decrease in amorphous silica solubility in 6 M $NaNO_3$ solution at 150°C.

The effects of metals on glass leaching are shown in Figure 3.3. Ductile iron and titanium appear to affect glass leaching in salt brine only slightly. There is some scatter in the brine data because of inherent difficulties in analyzing brines with ICPs. We are now considering the use of x-ray diffraction analysis rather than ICP for brine analyses.

In a salt repository brines likely to reach a waste package will be saturated with solid salts. Their saturation composition

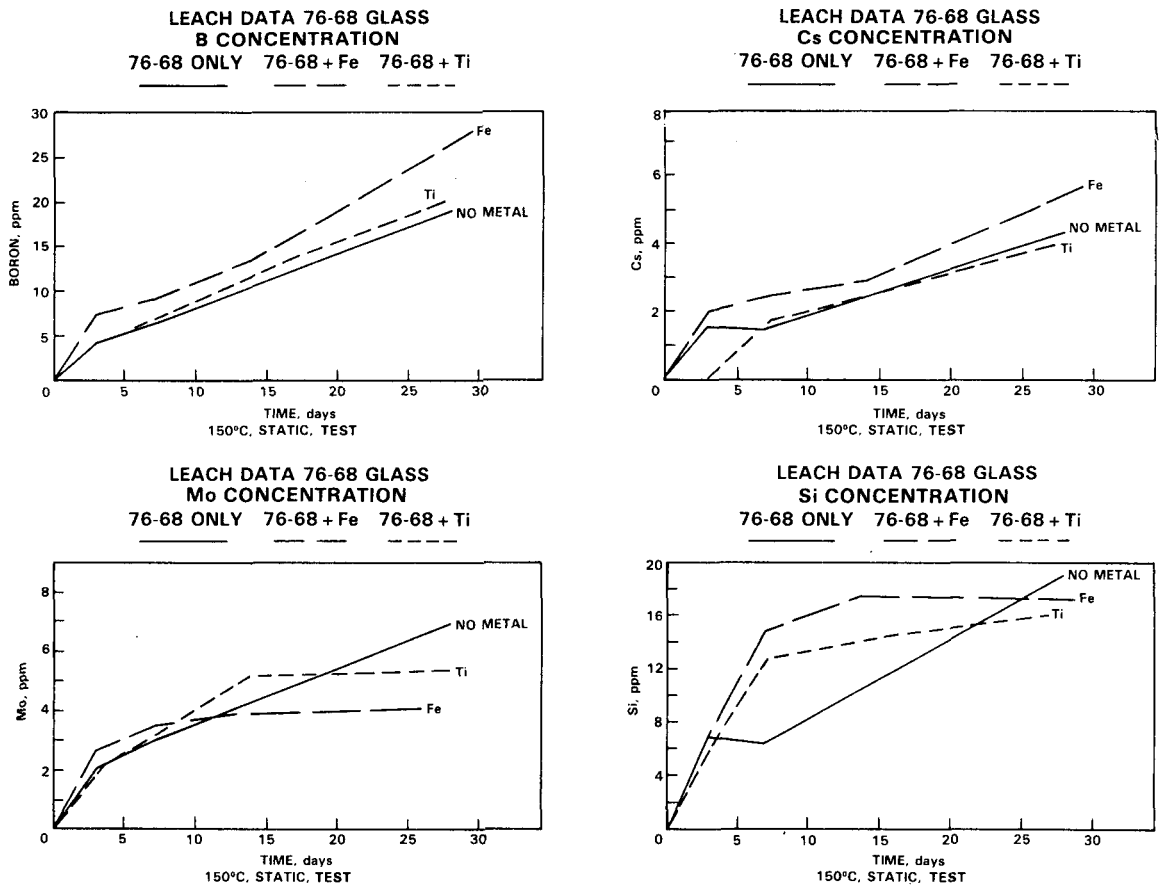


FIGURE 3.3. Plots of Solution Composition Versus Time for 76-68 Glass and 76-68 Glass Plus Iron or Titanium in Permian Basin Salt Brine No. 1

will be very temperature dependent; the alkali chlorides increase in solubility with increasing temperature. Thus it is important to carry out interaction tests with temperature-saturated brines and to take fluid samples at temperature and pressure. Our first attempt, however, to use a Dickson autoclave to sample a brine saturated at 250°C (20 MPa) resulted in a thoroughly clogged sample tube and no usable brine sample. We are now trying to develop sampling techniques to get around this problem.

To at least partly circumvent this problem we began a series of tests with room temperature saturated Permian Basin Salt Brine No. 1 and two diluted brines (50% and 75% of this room temperature saturated salt brine). These tests are being carried out in Dickson autoclaves, and successful liquid sample extraction has been achieved for all three compositions. These data will help greatly in the understanding of temperature-produced variations in brine compositions (e.g., Marshall 1980b) and will provide a data base that aids in evaluating techniques for sampling saturated brines at temperature and pressure.

Initial tests were completed for the 50% salt brine at 150°C and for 50, 75, and 100%

salt brines at 250°C. The 50% brine test produced no change in brine composition, indicating that at 150°C none of the inverse solubilities was exceeded. However, all three tests at 250°C indicate a marked decrease in concentrations of Ca^{2+}, Mg^{2+}, and SO_4^{2-}, probably a result mainly of the precipitation of calcium and magnesium sulfates. The measured saturation values Ca^{2+} Mg^{2+} and SO_4^{2-} for the three brines at 250°C (20 MPa) are plotted in Figure 3.4 as a function of relative brine strength (100% No. 1 brine = 1.0). For these brine compositions the saturation value for Ca^{2+} shows a clear increase with increasing ionic strength, while the saturation concentration of Mg^{2+} passes through a maximum and the saturation concentration of SO_4^{2-} increases to an apparent plateau, although a slight decrease is possible.

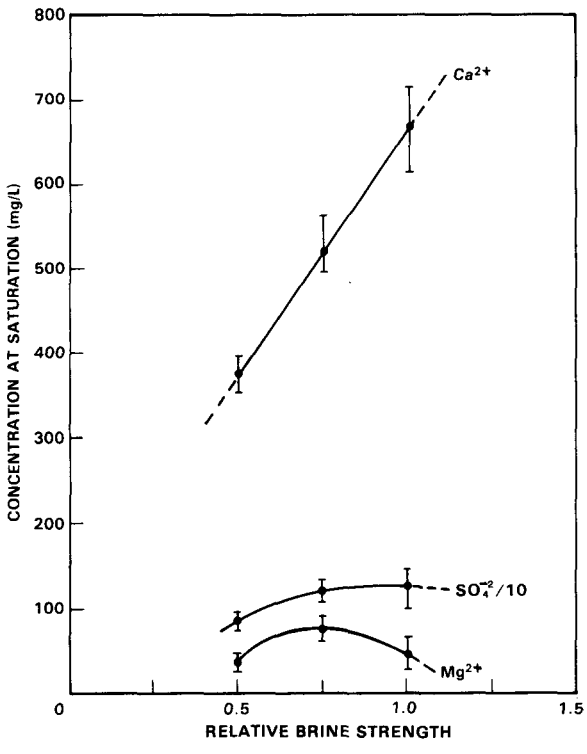


FIGURE 3.4. Plot of Saturation Values for Ca^{2+}, Mg^{2+}, SO_4^{2-} (mg/L) Versus Relative Brine Strength at 250°C and 20 MPa. The brine strength of Permian Basin Salt Brine No. 1, saturated at 25°C, is set arbitrarily equal to unity.

Static autoclave tests are now under way with 76-68 glass and 76-68 glass + ductile iron at 150 and 250°C (20 MPa) in Permian Basin Salt Brine No. 1. Dickson autoclave tests are currently under way with 76-68 glass in this brine at 150 and 250°C and with 75 and 100% brine at 150°C. Preparations are nearly complete for the initiation of tests with uranium-doped 76-68 glass and with UO_2 as preparation for failure tests with actinide-doped 76-68 glass and spent fuel.

Waste Form Release Modeling

The literature on leaching of spent fuel and dissolution of UO_2 is being reviewed to form a basis for a spent fuel release model. Dissolution of UO_2 appears to be controlled by the oxidation of U(IV) to U(VI), which may be caused by oxidizing agents in the solution, by radiolysis of water, and by radiation damage to the UO_2 itself. Some species such as cesium are not homogeneously distributed in the spent fuel but may segregate at grain boundaries, cracks, and fuel cladding interfaces. The model will have to be extended beyond simple dissolution of UO_2 to account for the release of these migrating species.

The results of flowing leach tests, in which the concentration at the exit to a cell holding a sample is measured versus time, are important data to utilize in developing a rate law for the reaction of borosilicate glass waste forms (as well as other waste forms) with leachants. In this task we had been assuming a perfectly mixed

cell for the purpose of interpreting the data, but it appears this may not be appropriate based on an order-of-magnitude estimate of the resistance to mass transfer in "boundary layers" around a sample of glass.

Corrosion Modeling. Our current effort has revolved around the investigation of the basic mechanisms that could be postulated as controlling the rate of corrosion of iron under irradiation conditions. Consideration has also been given to proposing experimental tests that could aid in differentiating the numerous possible mechanisms.

The main concern is the increase of the corrosion rate due to radiolysis of the brine. The effect of radiolysis is to produce, either directly or indirectly, the following:

a. short-lived radical species by direct ionization of the brine constituents
b. intermediate species through interaction of the short-lived species with the brine and each other
c. stable molecular species by various interactions of a, b, and the brine.

Much has been done to quantify the yields of the above species, yet little has been done to interrelate these yields with corrosive effects.

An experimental approach has been put forth that could prove useful in differentiating between a, b, and c. The scheme could also be used to predict dose rate dependencies and possible thresholds.

Waste Package Modeling. Work on a better understanding of brine chemistry has been initiated with a series of meetings to clearly define what can be done in FY 1983. An experimentalist has been identified who should be able to help particularly with a review of the pertinent experimental literature. We reiterated the importance of finding: 1) what pH excursions have in fact been found when brines are heated, and which precipitates are responsible for them, and 2) what activity coefficients have been observed in brines, especially at higher temperatures.

References

Marshall, W. L. 1980(a). "Amorphous Silica Solubilities: I. Behavior in Aqueous Sodium Nitrate Solutions: 25-300°C, 0-6 Nitrate Solutions: 25-300°C, 0-6 Molal." Geochim et Cosmechim Acta, 44:907-913.

Marshall, W. L. 1980(b). "Amorphous Silica Solubilities: III. Activity Coefficient Relations and Predictions of Solubility Behavior in Salt Solutions, 0-350°C." Geochim et Cosmechim Acta, 44:925-931.

3.1.2 Baselining and Benchmarking Salt Performance Assessment Codes (DB-30-30-10 - 80748)

The objective of this program is to provide ONWI with salt site-specific, baselined and benchmarked performance analysis codes to perform the comprehensive analysis required to site, design, and license a repository in salt. Codes to be baselined and benchmarked include SWENT, CFEST, and FE3DGW.

Background

To site, design, and license a repository in salt, ONWI will be required (by NRC regulations as well as good scientific practice) to have salt site-specific baselined and benchmarked hydrologic system performance analysis codes. This report discusses the hydrologic benchmark problem specifications for benchmarking the hydrologic computer codes-[CFEST (flow only), FE3DGW, and SWENT (flow only)].

In searching for an appropriate benchmark problem definition for testing hydrologic computer codes, the following reports and data sources were utilized:

- the Permian Basin data set (ONWI-292)
- Benchmark Problems for Repository Siting Models (NUREG/CR-3097)
- data and report on the initial modeling of the Permian Basin hydrologic systems (ONWI/E512-02900/TR-13).

Any eventual analysis of a salt site like the Permian will most likely require that the upper and lower hydrologic systems and the intervening salt strata be modeled as a single system so that the effects of potential breaching scenarios, such as borehole interconnection, salt dissolution, and faulting, can be studied. The other pertinent considerations are that the parameter variation distributions (such as thickness and conductivity) be well defined and be representative of the kinds of changes that occur in real hydrologic systems.

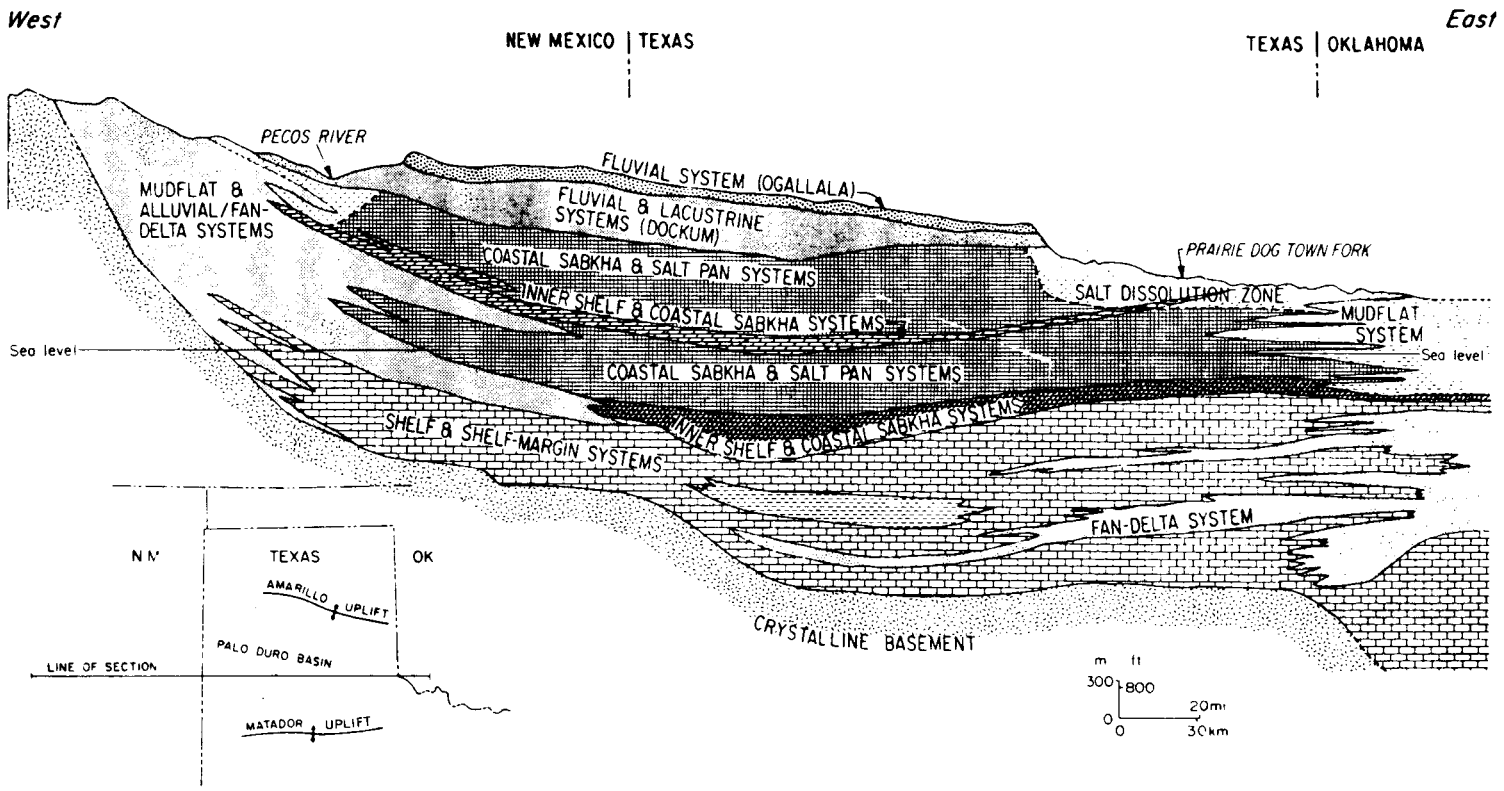
To achieve this degree of realism the Permian Basin data set was studied in detail (ONWI-292 and ONWI/E512-02900/TR-13). This investigation indicated a stratigraphy as shown in Figure 3.5. Our investigation indicates that for purposes of benchmark problem definition two hydrologic units above and two hydrologic units below the salt strata are sufficient. Areal extent was defined from the fact that the two upper hydrologic systems circulate generally along west to east lines. The north-south areal extent of the two upper hydrologic units was defined from essentially the northern caprock escarpment (see Figure 3.6), which acts as a groundwater divide, south to the Matador uplift (see Figure 3.5), which appears to act as a barrier to the north-south communication in the lower hydrologic units.

This uplift can also serve as an arbitrary southern no-flow boundary for the two upper hydrologic units since gradients in the upper units, as stated before, run essentially west-east. The upper and lower hydrologic units both circulate along west-east lines due to the pattern of regional surface drainage systems (see Figure 3.6). The lower hydrologic units are additionally forced into west-east circulation due to the east-west structures (Matador uplift to the south and the Amarillo uplift to the north, Figure 3.5), which are felt to limit north-south communication. Recharge occurs throughout the basin. Estimates for reacharge to the upper units vary from 1.3 cm/yr to 0.44 cm/yr. Recharge to the lower units occurs in the Pecos Mountains to the west, and one would assume slightly higher but similar recharge estimates since the differential in elevation is only ~500 m. The fact that the recharge source for the lower hydrologic units lies in the Pecos Mountains to the west, when considered along with the essentially aquiclude nature of the Permian, explains the under-pressurization of these lower units which ultimately discharge much further east.

To ensure that this benchmark problem is well posed, trial simulations were made and parameters adjusted for a consistent and hydrologically reasonable description. An attempt was also made to keep all parameter values in the range of observations.

Salt Site-Specific Regional Hydrologic (Flow Only) Benchmark Problem Specifications

This benchmark problem is designed to test the ability of groundwater flow codes to model regional groundwater systems for



SOURCE: Bentley (1981)

FIGURE 4-6
EAST-WEST CROSS-SECTION OF THE MAJOR DEPOSITIONAL SYSTEMS IN THE PALO DURO BASIN

FIGURE 3.5. East-West Cross-Section of Major Depositional Features of the Permian Within the Palo Duro Basin. This stratigraphy provided the basis for the hydrologic benchmark problem definition. (from ONWI-292)

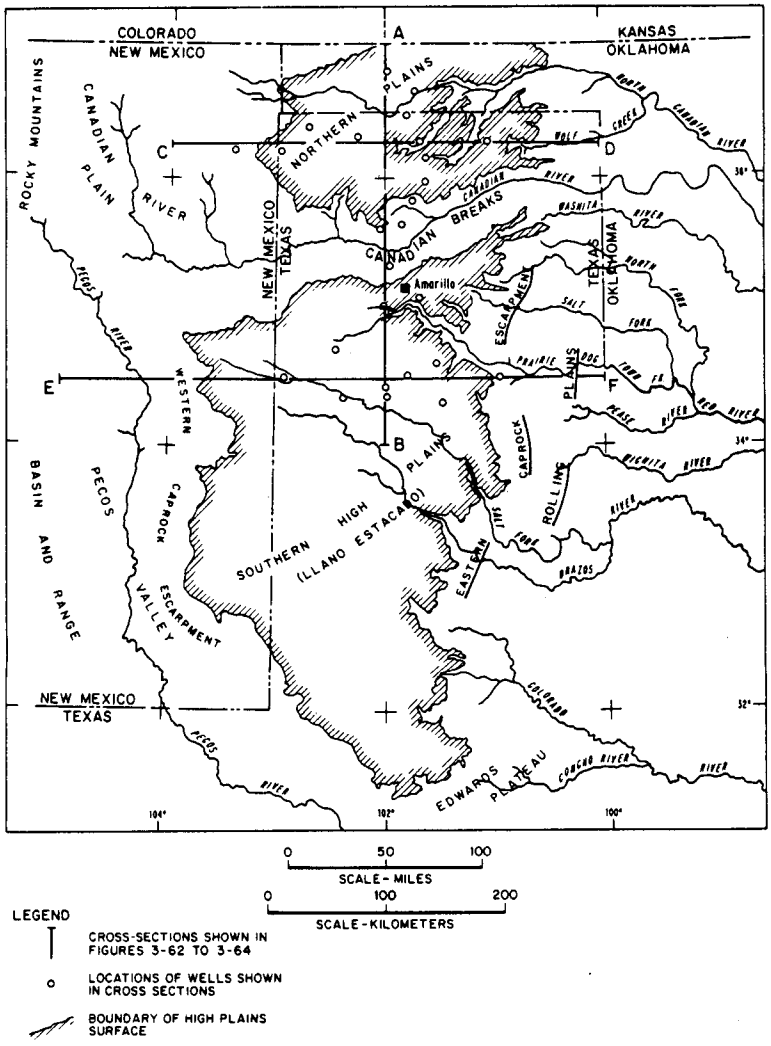


FIGURE 3.6. Major Physiographic Elements of the Texas Panhandle and Major Surface Water Drainage Features (from ONWI-292)

layered sedimentary hydrologic systems typical of bedded salt sites. In formulating this problem an attempt was made to address as many features important to regional modeling and performance assessment needs as can be done in a simplified manner. These features include:

- layering
- anisotropic hydrologic conductivity (i.e., $Kh \neq Kv$
- recharge
- discharge boundaries (rivers)
- outflow boundaries (held head-Dirichlet)
- no-flow boundaries
- interlayer communication (via borehole or fracture zone)
- water table conditions (in some layers or some areas of some layers) or totally confined conditions.

Basic Assumptions. Since this benchmark problem is designed to address flow only, it is assumed:

1. Water in all hydrologic systems is of the same constant density and temperature and that Darcy's law and all other basic assumptions associated with the porous media continuum approach hold.

As this problem is designed to address the saturated flow-only problem, it is additionally assumed:

2. The system being simulated is 100% saturated.

The potentially long time required for water or contaminants to move from a proposed repository at depth to man's environment or to the 10-km boundary addressed in the proposed regulations nullifies most needs to model the hydrologic flow system in a transient manner, so that it is assumed:

3. The hydrologic system is in steady state and the boundary conditions are constant over time so that only the steady-state solution for the hydrologic system need be addressed.

For purposes of this benchmark problem it is additionally assumed:

4. No significant hydrologic flow takes place in the crystalline basement rock.

Specifications for Layering-Scale Geometry

The hydrologic system is assumed to consist of essentially five layers: a bedded salt layer overlain by two aquifer layers and underlain by two additional aquifer units. These five layers degrade into essentially one in the recharge areas to the west where the salt strata truncates. Figure 3.7 depicts a plan view for this idealized benchmark problem that illustrates the surface features (outcrops and rivers), the scale, and the kinds of boundary conditions to be imposed.

Figure 3.8 shows cross-section views of this idealized system to illustrate hydrologic stratigraphy and give a complete description of the geometry. Figure 3.9 provides further plan views showing the areal extent of each hydrologic unit and their associated hydrologic parameters.

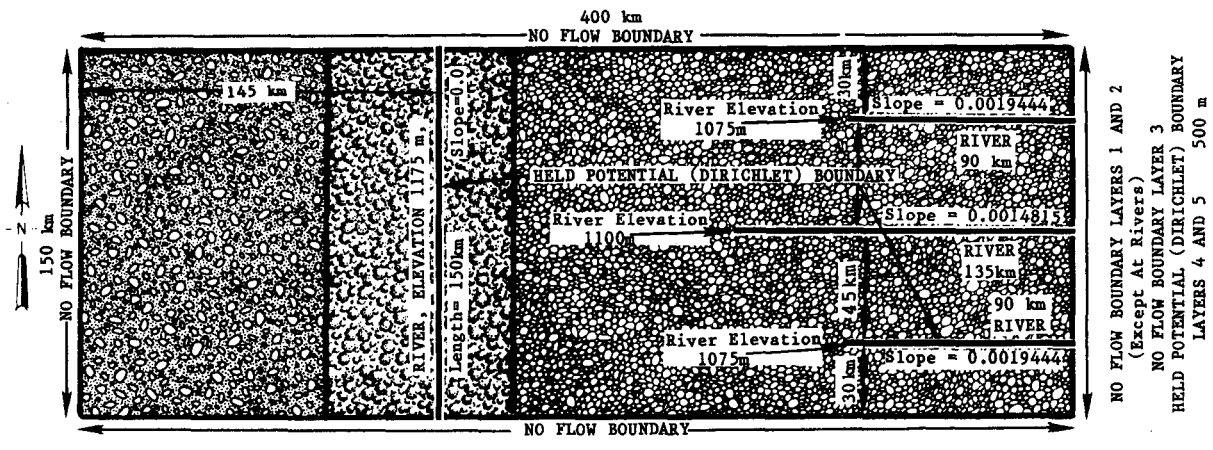


FIGURE 3.7. Plan View of Hydrologic Benchmark Problem Area, Illustrating Surface Drainage, Outcrops, and Boundary Conditions To Be Considered

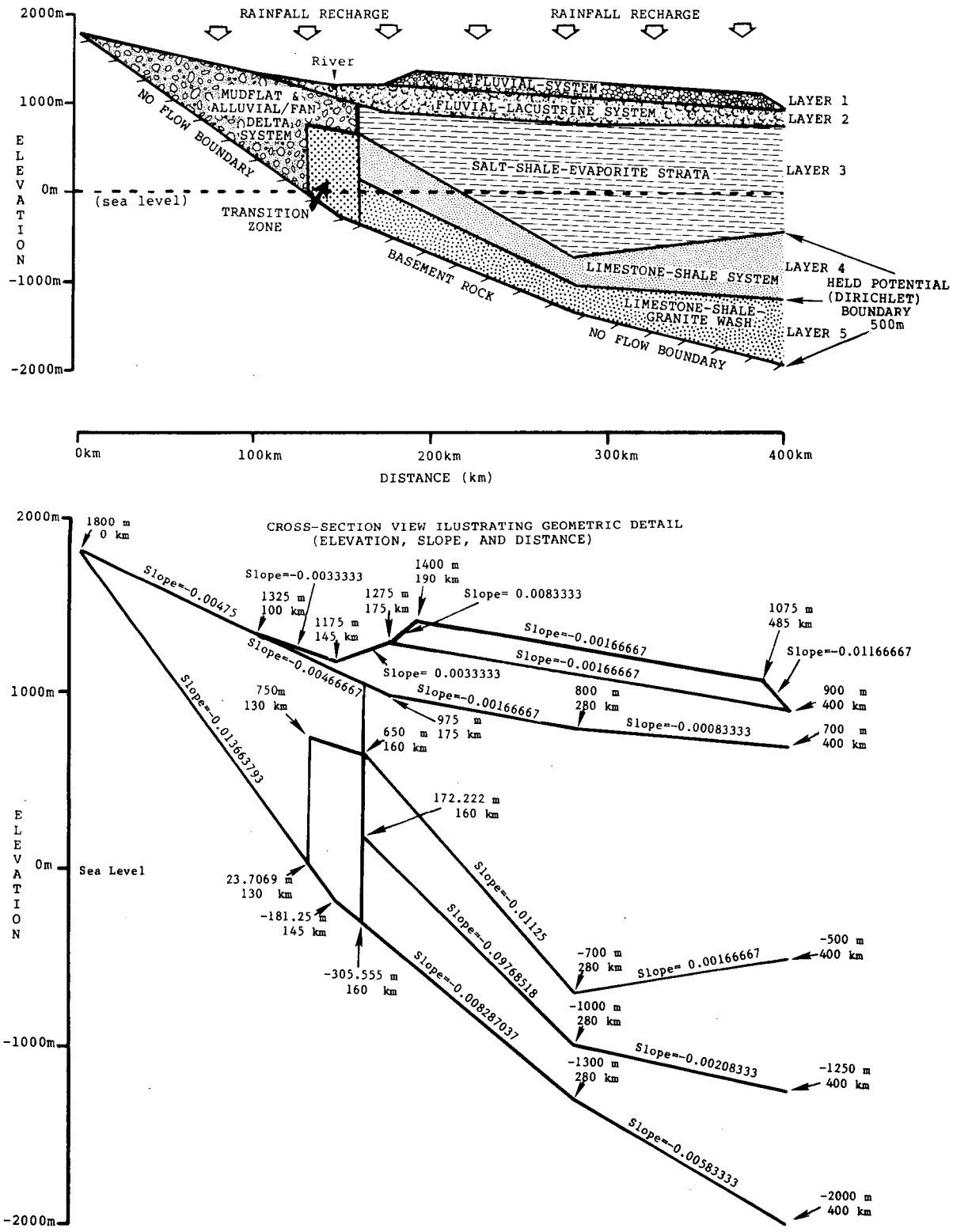


FIGURE 3.8. Cross-Section Views of Area Illustrating Geohydrologic Detail such as Layering (Stratigraphy), Boundary Conditions, Elevations and Geometric Detail

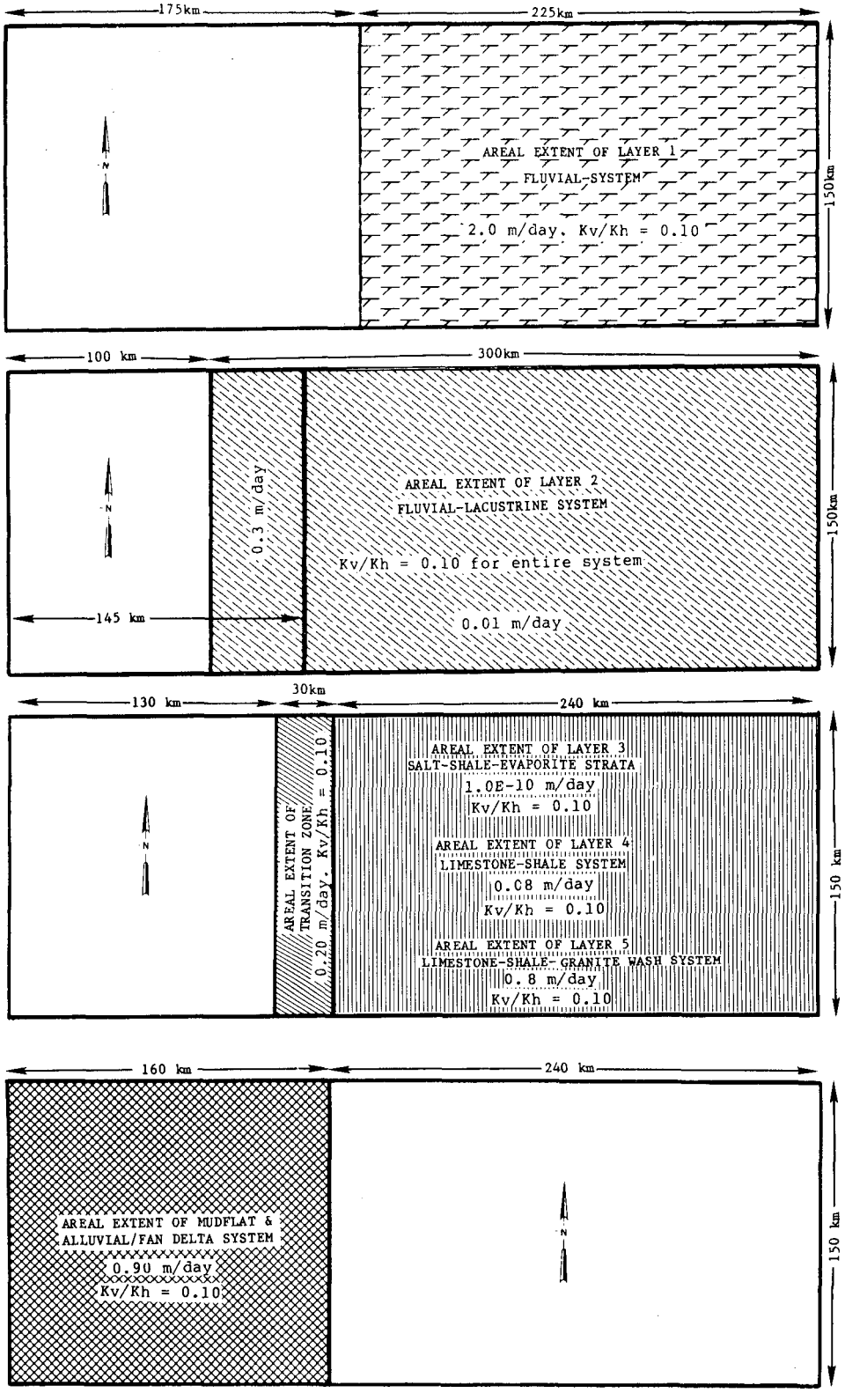


FIGURE 3.9. Plan Views Illustrating Areal Extent of Each Hydrologic Unit, Dimensions and Hydrologic Parameter Values for Each Unit

Specifications for Recharge and Boundary Conditions

Recharge to the groundwater systems occurs at 0.75 cm/yr uniformly over the problem area shown in Figure 3.7. Discharge is to the four river systems shown. These hypothetical rivers are to be treated as held potential (Dirichlet) boundaries. There is one north-south river, 150 km long and located 145 km from the western boundary of the problem area. It has a constant potential of 1175 m. There are three west-east rivers. The centrally located west-east river (75 km from the southern problem boundary) is 135 km long, and its elevation varies from 1100 m at its west end to 900 m along the model boundary to the east. The other two west-east rivers are each 90 km long and located 30 km from the northern and southern problem boundaries, respectively. These two rivers have the same potentials and slopes. They vary in potential from 1075 m at their west end to 900 m at their east end. The elevations for the held potential boundaries are also pictured in Figure 3.7.

Three sides--west, north, and south--of the problem area are no-flow boundaries for all hydrologic units (Figure 3.7). The east side is a no-flow boundary except where the rivers are located for the upper three units--1) fluvial system, 2) fluvial-lacustrine system, and 3) salt-shale-evaporite strata--and a 500-m held potential (Dirichlet) boundary for the lower two units--4) limestone-shale system and 5) limestone-shale-granite wash system.

Specifications For Hydrologic Parameters. Horizontal hydraulic conductivity values for the various hydrologic units as well as the ratio of vertical hydraulic conductivity to the horizontal hydraulic conductivity are as follows:

- Fluvial system (layer 1) 2.0 m/day. Kv/Kh = 0.10.
- Fluvial-lacustrine system (layer 2) 0.01 m/day over the region east of the north-south river and 0.3 m/day over the region west of the north-south river. Kv/Kh = 0.10 for entire system (Figure 3.9).
- Salt-shale-evaporite system (layer 3) 1.0E-10 m/day. Kv/Kh = 0.10.
- Limestone-shale system (layer 4) 0.08 m/day. Kv/Kh = 0.10.
- Limestone-shale-granite wash system (layer 5) 0.08 m/day. Kv/Kh = 0.10.
- Mudflat and alluvial/fan delta system 0.90 m/day. Kv/Kh = 0.10.
- Transition zone 0.20 m/day. Kv/Kh = 0.10.

The hydrologic parameters for the various units are also displayed in Figure 3.9.

Run Specifications. This problem can be run in two ways. It can be assumed that:

- All hydrologic systems are totally confined, with the stated geometry used only to determine the transmissivity distributions for the hydrologic systems. The problem consists of determining the hydraulic head distributions within the various hydrologic units.
- The geometry relates only to potentially saturated hydrologic units and the actual topographic surface may indeed be higher

than illustrated in Figure 3.8. For this case the problem consists of determining the location of the water table surface as well as determining the hydraulic head distributions within the various hydrologic units.

Output Specifications. The output of this hydrologic benchmark problem is the hydraulic head distribution. Contour plots showing areal distribution of hydraulic head for each layer and selected west-east cross-section plots should be produced along with lineprinter listings of hydraulic head distribution.

Initial Benchmarking Runs with the FE3DGW Code

The confined version of the salt site-specific, regional hydrologic flow problem discussed above has been set up, and initial model runs were made to determine appropriate parameter matches. Figure 3.10 shows an isometric view of the three-dimensional grid used to simulate this hydrologic benchmark problem. The problem will eventually be additionally run in an unconfined mode as part of the complete benchmarking of the FE3DGW and CFEST codes.

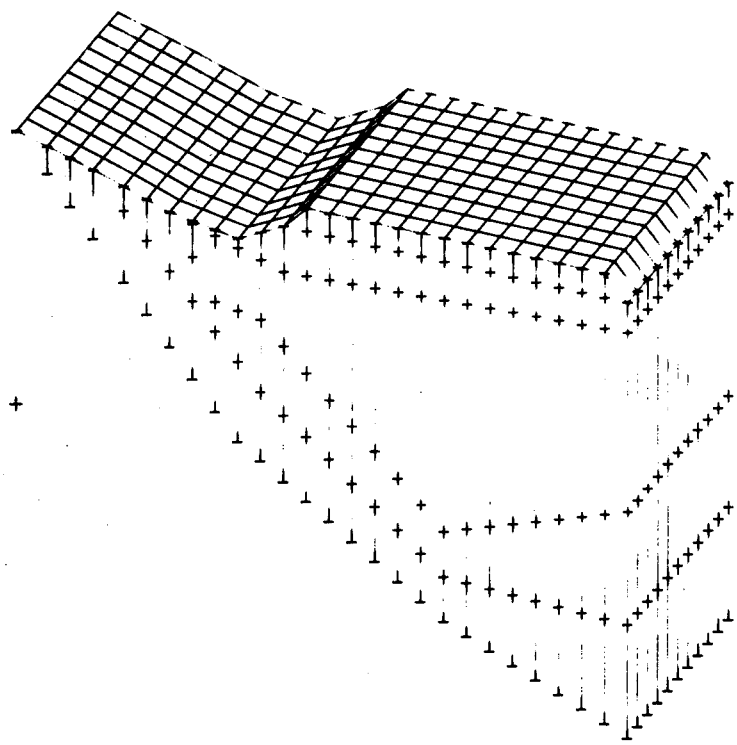


FIGURE 3.10. Isometric View of Initial Finite-Element Grid Used To Simulate Confined-Only Version of the Salt Site-Specific Benchmark Problem

3.2 BASALT WASTE ISOLATION PROJECT (BWIP)

3.2.1 Modeling Task Force (AP-05-15-05 - 80750)

This effort will continue the modeling task force activities initiated in FY 1982. The purposes of the modeling task force are to examine the Rockwell and PNL approaches to modeling the Pasco Basin, resolve differences where possible, and recommend and implement approaches for further resolution of remaining significant differences. The model task force is a mechanism for detailed peer review of the far-field hydrologic modeling.

Technical Progress

As part of the Assessment of Effectiveness of Geologic Isolation Systems (AEGIS), PNL conducted a modeling methodology and demonstration effort using basalt as one demonstration medium. A regional model encompassing the Washington State portion of the Columbia River basalts was used to establish boundary conditions for a Pasco Basin model. The Pasco Basin model was used to predict flow paths and travel times from a hypothetical repository to the river. As a result of this demonstration effort, attention was drawn to differences between PNL's, Rockwell Hanford Operations' and the U.S. Geological Survey's conceptualization of the Pasco Basin basalts.

To resolve those differences, a modeling task force was established in FY 1982. The goals of the task force are to examine the groundwater modeling conducted to date, resolve differences where possible, and recommend and implement approaches for further resolution of remaining differences. More specifically, an attempt to resolve differences will be made by 1) establishing and agreeing upon a standard data set; 2) mutually reviewing the interpretations necessary to achieve a conceptual model; and 3) preparing model input and conducting sensitivity analyses to evaluate the impact of uncertain or unknown parameters.

In FY 1982 the basic differences were clearly identified, a course of action for resolving the differences was established, and some initial modeling studies (sensitivity analyses) were conducted. The basic differences can be summarized as follows:

1. What are the Pasco Basin boundary conditions?
2. What is an acceptable range of hydraulic conductivities for the Saddle Mountain, Wanapum, and Grande Ronde basalts?
3. What is the nature of the vertical movement (permeability) in the Pasco Basin?
4. Is the flow system in the Pasco Basin at steady state?
5. How reliable are potentiometric data obtained during drilling?
6. How reliable are water quality data for determining groundwater flow directions and quantities?

Some of the differences will require additional data before they can be resolved. The task force has acted as a peer review committee for identifying additional data needs and for reviewing proposed field activities.

Differences that can be studied (possibly not completely resolved) with additional modeling (i.e., sensitivity analyses) were the central thrust of the FY 1982 task force effort. PNL has been heavily involved in this effort because we were selected to conduct the modeling sensitivity studies. The first task of FY 1982 was to completely document all the data sources that PNL used in their initial modeling effort. This complete data set was then used by the task force to define a basic revised conceptual model of the Pasco Basin to which all members could

agree. The new model region extends far beyond the Pasco Basin to the southwest, and its major purpose is to help resolve the question of the boundary condition along Rattlesnake Mountain and the Horse Heaven Hills. PNL is currently in the process of collecting the additional data required for this model and implementing it on our computer system. The model selected by the task force for this effort is the USGS/Trescott three-dimensional groundwater flow model. Since we have never used this model, it was necessary for us to obtain it, bring it up on our computer system, and become familiar with it. This task has been accomplished and we are currently running the model with the first data set agreed to by the task force.

Future Activities

Many additional sensitivity runs agreed to by the group will be made later this year to address the boundary condition issue and the other differences discussed above.

3.2.2 Preclosure Performance Assessment (AP-05-15-15 - 80750)

The objective of this program is to develop a preliminary hazards analysis of conceptual waste package and repository designs. The preliminary hazards analysis will identify and assess the consequences of repository operational hazards and provide recommendations for improvement in safety design criteria, design concepts, or operational procedures.

Summary

A series of system flow diagrams for normal repository operations has been completed, as well as a review of federal criteria and regulations relevant to the preclosure safety of a repository. Available literature and the conceptual design document for the nuclear waste repository in basalt were used as background material for these efforts. Potential hazards during repository construction and operation are being evaluated.

System Description

An informal report containing system flow diagrams and descriptions for normal operations of a repository in basalt has been completed in draft form and is being circulated for comment. The report will be incorporated into a final project report in 1984. The system description of the repository includes a discussion of the surface handling and support facilities, access shafts, and subsurface facilities. Major subsystems and equipment and system components that support the operations of the repository phases are identified and discussed. Descriptions and flow diagrams are presented for the following phases:

- initial construction
- mine development
- waste receipt, handling and emplacement
- waste retrieval
- backfill and decommissioning.

Criteria Review and Analysis

An informal report summarizing the federal criteria relevant to the preclosure safety of a repository has been completed in draft form. This report, presently being circulated for comment, will be incorporated into a final project report in 1984. The report includes an analysis of the safety focus of relevant criteria (radiological or nonradiological, public or occupational) and the functional subsystems or repository locations to which they apply. Criteria included in this

analysis were relevant Nuclear Regulatory Commission standards, Occupational Safety and Health Administration and Mine Safety and Health Administration standards, Environmental Protection Agency regulations and relevant Department of Energy orders.

Safety Assessment of Normal Operations

Efforts on this task were recently initiated. A format and approach for the safety analysis methodology to be used were determined. Hazard analysis on repository construction operations has begun.

3.2.3 Development of Eh-pH Sensors (AP-05-15-05 - 80752)

The objective of this work is to develop Eh and pH sensors compatible with the Dickson rocking autoclave. These sensors will provide monitoring capabilities of the oxidation-reduction environment and the hydrogen ion concentration for the hydrothermal waste-barrier-rock interaction studies. Maximum operating limits are 300°C and 300 bars pressure by the close of FY 1983.

Summary

To fit the sensors into the Dickson autoclave, PNL redesigned the autoclave lid. Rockwell Hanford Operations has ordered several of these vessels from the autoclave vendor.

The Eh electrode has been designed and is currently undergoing laboratory evaluation. Preliminary results in a hydrogen environment look very promising. There is a good chance the gold bag of the Dickson autoclave can function as the Eh sensor, which will result in a considerable simplification in Eh design.

A high-temperature, zirconia-based pH electrode is being developed. Zirconia tubes have been procured from the manufacturers and tested for pH response at 90°C. The majority have a good pH response. Other researchers have found pH response to be extremely rare among zirconia tubes, particularly at low temperatures (90°C). Our results are extremely encouraging. Work will continue in evaluating their pH response at elevated temperature and in solving the pressure-seal problems.

Dickson Autoclave

With the original Dickson autoclave, consisting of one autoclave inside another, two sets of pressure seals would be required for the sensors to be inserted so as to penetrate the inner (gold bag) autoclave. PNL has solved this problem by combining the lid for the inner autoclave with that of the outer. Thus only one pressure seal penetration is now needed. This design, which permits the vessels to be operated in a hot cell, is shown in Figure 3.11. Rockwell Hanford Operations has ordered several of these vessels.

Eh Sensor

The Eh sensor will measure the oxidation-reduction potential of the aqueous environment. Solubilities of aqueous compounds are highly dependent upon their oxidation state.

The design for the Eh sensor is shown in Figure 3.12. It is a blowout-proof design for use with the PNL-modified Dickson autoclave. It is uncertain what inert metal is best for Eh measurements. In the laboratory tests, platinum, gold, and titanium are being evaluated. If gold and titanium operate satisfactorily, then the gold bag (autoclave lid is titanium) can be used as the Eh

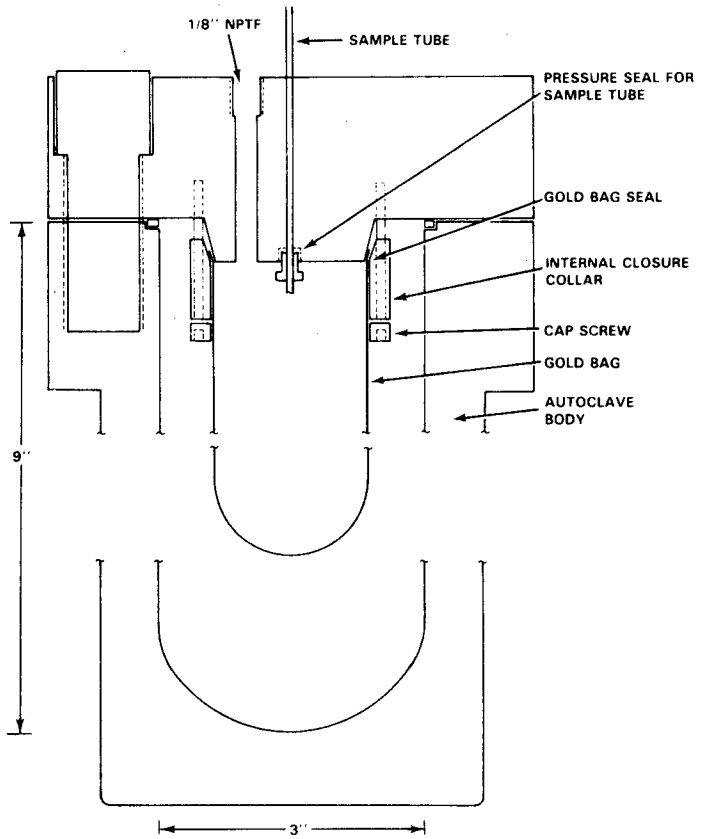


FIGURE 3.11. PNL-Modified Dickson Autoclave

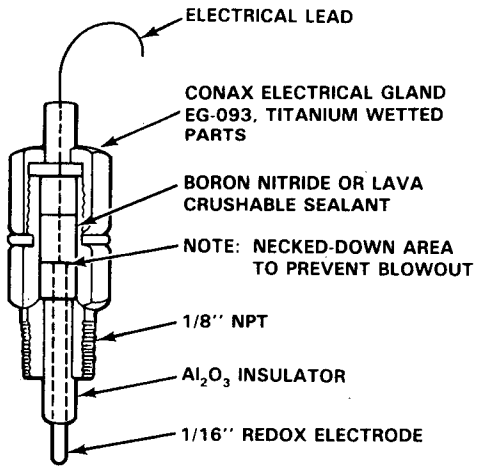


FIGURE 3.12. Eh Electrode

sensor, considerably simplifying sensor design.

Figure 3.13 is a Pourbaix diagram for the Fe-H_2O systems at 200°C. Evaluation of the Eh electrode should be in an environment similar to that expected in actual rock-water experiments. That environment is considered to be strongly reducing (in the Fe_3O_4 region) within a pH range of 5 to 9.

An experiment has been carried out at a pH of about 5.6 from 100 to 300°C in a hydrogen environment (along line 'B' of Figure 3.13), and measured Eh values and calculated agreed extremely well. Above 150°C, the platinum, gold and titanium electrodes act similarly, which means the gold bag can act as the sensor.

Future experiments will explore the Eh response in hydrogen at a pH of 9. The more oxidizing environments of Fe_2O_3/Fe_3O_4 will be explored at pH 5 and pH 9. The effects of trace amounts of oxygen will also be examined.

pH Sensors

The most important variable in water chemistry is pH. The objective of this program is the development of pH sensors suitable for use in the Dickson rocking autoclave at temperatures to 300°C and 300 bars pressure. A conceptual design is shown in Figure 3.14.

Procedurally, the program requires acquisition of zirconia-based ceramic tubes,

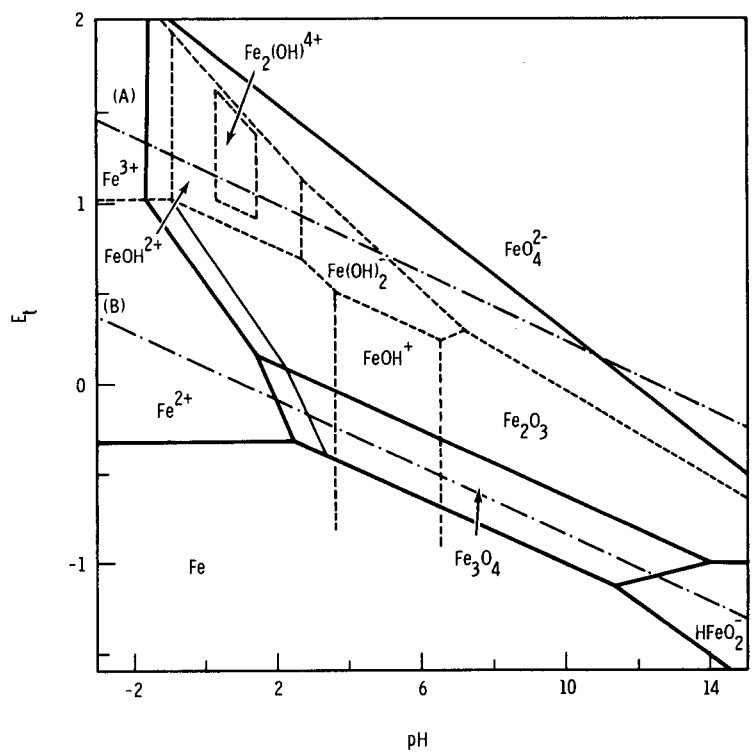


FIGURE 3.13. Fe-H_2O System at 200°C. E_t is an absolute voltage scale which can easily be placed on the standard hydrogen electrode by examining the hydrogen line (B) at standard conditions.

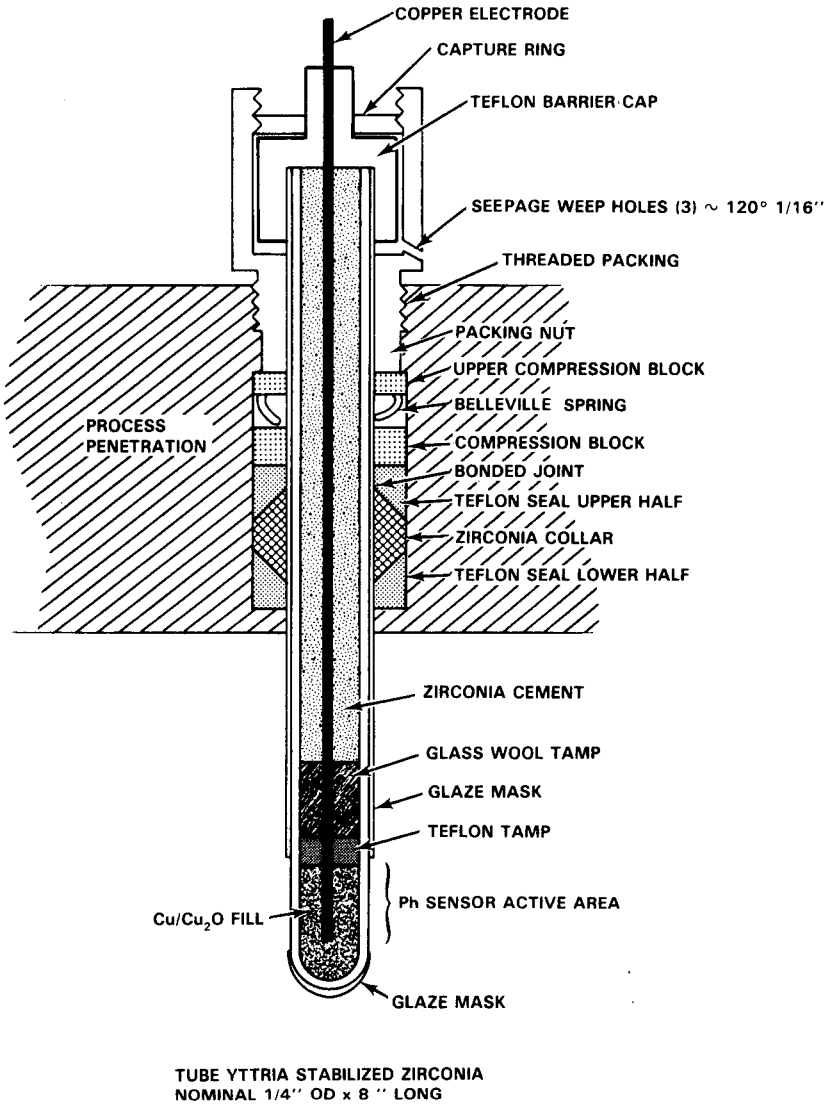


FIGURE 3.14. Conceptual Design of pH Electrode

screening of the material for pH response, fabrication of the tubes into pH electrodes, and certification of electrochemical and mechanical integrity under the conditions of BWIP use. Sixty-two zirconia-yttria tubes have been characterized with respect to "as received" pH response. Of those tested, 60 were found to be functional at temperatures of 90°C. Two sources of material (Coors and Zircoa-Corning) were used (all 8 wt% yttria).

Five tubes were given long-term tests using buffers 4, 7, and 9 at 90°C, and the average Nernstian response was 93.8% with a range from 89.8 to 96.4%. Within this group, one probe utilizing a Cu/Cu_2O internal reference with a chemical etch pretreatment (Zircoa-Corning material) produced the highest response, lowest standard deviation of the measured value (<1 mV), and lowest long-term drift (<10 mV over a 10-day test period). Usable though sluggish response was

observed down to room temperatures. Experience of other pH researchers would generally have projected a high failure rate (no pH response), a sluggish response at temperatures below 90°C, and a characteristic high impedance. In contrast, our material appears to have a low failure rate in the "as received" condition and a characteristically low impedance compared to previously tested materials. Manufacturing procedures are yet to be developed to ensure long-term electrochemical stability and mechanical integrity.

Future work will encompass chemical and thermal treatment to fix the electrochemical activity of the inner reference, the mechanical bonding of component parts, and high-temperature and -pressure verification of pH performance. Validation of performance in projected radiation environments may also be required.

3.2.4 Solubility Studies (AP-05-15-05 - 80752)

The objective of this work is to obtain radionuclide solubility kinetics and equilibria for key radionuclides in the presence of each of the geologic materials that the radionuclide contaminated groundwater might contact. This includes basalt, basalt flow top and secondary minerals from the Umtanum flow and the Middle Sentinel Bluff. Because basalt is a dominant component of the waste package backfill, data from these experiments will complement the basalt-bentonite multicomponent solubility studies to be conducted at Rockwell in FY 1983. Rock materials used in this study will be supplied by the BWIP Waste Package Department.

Summary

Four radionuclides were examined with and without the presence of basalt in concentrations that cross from a removal mechanism of all sorption to one of predominantly precipitation. The flow-through apparatus was modified to use with crushed basalt. A silica and a bentonite colloid were generated and utilized in initial radionuclide migration experiments.

Technical Progress

The solubility-sorption relationships of four radioelements were examined in the presence of basalts in GR-3 synthetic groundwater at 60°C. They included selenite-selenium, uranium, technetium, and carbon. The solubilities were compared to those of both reducing and oxidizing systems without basalts present. Three types of filtration, ranging from none to 15 Å, were used to show the sizes and amounts of any precipitates present in solution.

Figure 3.15 shows technetium removal from solution by filtration after 30 days contact with Umtanum basalt. Without the presence of basalt, the technetium in solution was the same as that initially added (C_I=C), or no technetium was removed from solution. All three filtration results were the same in Figure 3.15 because the technetium was reduced and removed by contact with the Umtanum basalt. There were no precipitates in solution to remove by filtration.

Several precipitates were tentatively identified in the higher concentration radionuclide solutions, including schoepite and uraninite in the oxidized and reduced uranium systems, colloidal, red selenium and black selenium metal in the reduced selenium systems, and black TcO_2 in the reduced technetium systems. There were no apparent precipitates in the carbon system.

The flow-through apparatus required extensive modifications to change from an operational mode for a basalt core to one for crushed basalt. These alterations have

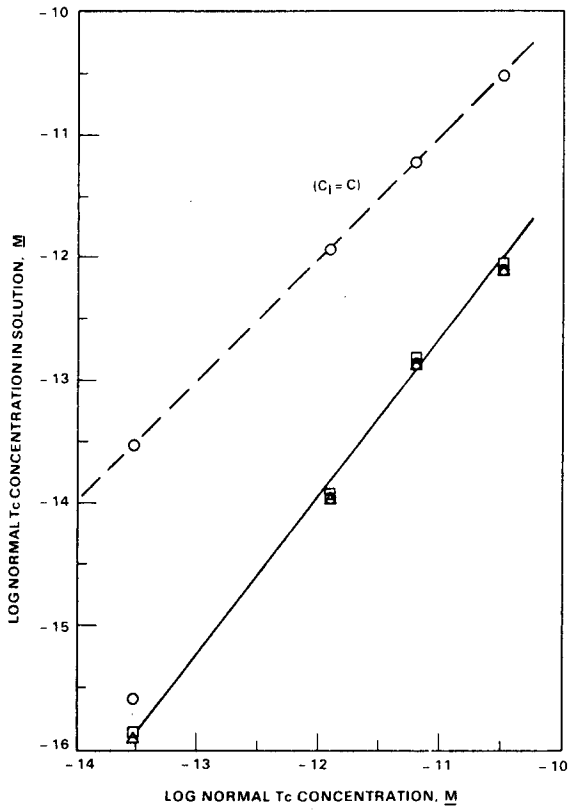


FIGURE 3.15. Removal by Filtration of Technetium from Oxidizing GR-3 Solution at 60°C for 30 Days in the Presence of 16 to 60 Mesh Umtanum Basalt

essentially been completed, and testing at pressure and temperature should get under way soon.

Colloids were prepared from the bentonite backfill and from silica suspensions for use in radionuclide sorption and migration studies. Initially, the radionuclide-loaded colloids in GR-3 synthetic groundwater were contacted with basalts for 30 days to determine the radionuclide distributions between solution, colloid and basalt.

Under oxidizing conditions, the order of sorption on the bentonite colloid was uranium > neptunium > selenite > technetium, and for reducing conditions, neptunium > selenium > technetium. The basalt sorbed considerable amounts of radionuclides on the colloid and in solution in some cases. In decreasing order of specificity, the oxidizing environment resulted in basalt sorption of neptunium > uranium > technetium > selenite, and for the reducing environment, uranium > selenium > technetium > neptunium.

The affinity of the uranium for the silica colloid was even higher under both oxidizing and reducing conditions. Oxidized selenite, selenate and technetium remained in solution, while under reducing conditions only selenite and selenate prefer the basalt to the colloid.

3.2.5 Hydrothermal Testing of Materials in the 325 Building Hot-Cell Test Facility (AP-05-15-05 - 80752)

The objectives of a joint PNL-WHC-BWIP hot-cell hydrothermal testing program are to provide waste-barrier-rock interactions data for quantifying the performance of waste package components in a basalt-repository environment and then evaluate and integrate these data with the BWIP package design effort. The major PNL responsibilities are to provide skills and assistance in the autoclave operation and to conduct the primary interpretation of the hydrothermal data.

Summary

In late FY 1982, a team of Hanford contractors was formed to perform waste-barrier-rock interaction studies using fully-loaded waste forms in a hot-cell test facility. This project is to provide data for quantifying the performance of waste package components in a basalt repository environment. The team of Hanford contractors includes Rockwell Hanford Operations (RHO/BWIP), PNL, and Westinghouse Hanford Company (WHC). BWIP provides program direction, WHC is building the shielded test facility and providing

analytical support, and PNL is providing skills and assistance in running the facility and will conduct the primary interpretation of the testing data.

The facility is currently under construction and is scheduled to begin operation in early FY 1984. Funding for the program began in October 1972. While the facility is being built, PNL is carrying out a series of hydrothermal tests using tracer-doped waste forms. These tests are being run in a PNL hydrothermal testing facility but in conjunction with WHC and BWIP. While providing valuable scientific data, the tests also constitute a "shake-down" for the project. This allows technical problems in the project to be worked out and allows the three groups time to coordinate their activities before the hot-cell testing begins.

Technical Progress

PNL assisted WHC in writing procedures for the operation of rocking autoclaves in the hot-cell testing facility. These procedures were prepared for inclusion in the BWIP basalt operating procedure handbook (RHO-BWI-MA-4).

An informal report, "Preparation and Analysis of 77-260 Tracer Loaded Glass" by C. A. Knox (November 1982), was written describing a glass sample prepared for BWIP during FY 1982. This report describes in detail a glass sample that may be used when we conduct tests on tracer-doped waste forms in the hot-cell testing facility before beginning tests on fully loaded waste forms.

While the hot-cell facility is under construction, we are using the PNL hydrothermal-testing facility to test the behavior of various radionuclides doped into glass when the glass is reacted with basalt water and basalt water plus basalt. Currently two tests are under way on glass doped with ^{99}Tc. They are being run at 200°C and 30 MPa for about 3 months. The WHC team is providing the analytical support for these tests, both for the solutions and for the solids.

Figure 3.16 presents the ^{99}Tc fluid concentration data plotted against run time for both tests--the glass plus water and glass plus water plus basalt. For the glass plus water run, it can be seen that the ^{99}Tc content in the fluid rises rapidly from zero at the beginning of the test to over 40 mg/L after 300 hr. After 1600 hr of run time, the concentration of ^{99}Tc has increased to over 50 mg/L. This amount of ^{99}Tc in the fluid indicates that over 60% of the total amount of ^{99}Tc initially contained in the glass has been repartitioned from the glass into the fluid. Filtration of the fluid samples through both 4000Å and 18Å filters shows no difference in concentration, indicating that the ^{99}Tc is truly in solution (probably as TcO_4^-) and does not exist as a colloid. These results show that under the hydrothermal conditions imposed ^{99}Tc is very soluble in basalt groundwater alone and the glass is not very successful at containing it. However, when an equal amount (equal to the mass of glass) of ground Umtanum basalt is added to the system, ^{99}Tc greatly decreases in solution concentration. Figure 3.16 shows that the concentration rises from zero at the beginning to 2 mg/L after 24 hr and then remains at ~2.5 mg/L after 650 hr of elapsed run time. We suspect that the ^{99}Tc is released to the solution as TcO_4^-. When basalt is present, the ferrous iron it contains reduces the soluble TcO_4^- species to the insoluble TcO_2 compound, thus removing ^{99}Tc from solution. The importance of these observations is that basalt, due to its unique

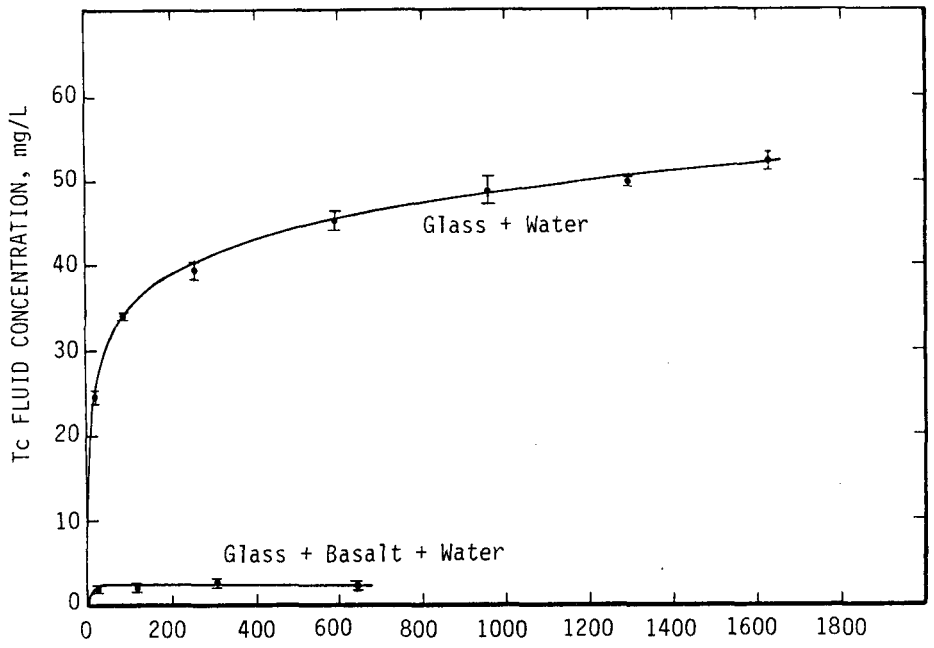


FIGURE 3.16. ^{99}Tc Fluid Concentration Versus Run Time for ^{99}Tc-Doped PNL 76-68 Glass Reacted with Basalt Water and Basalt Plus Basalt Water at 200°C, 30 MPA, and a 10:1 Solution to Solid Ratio

reducing capability, may effectively reduce the mobility of ^{99}Tc after the repository is breached. The potential mobility of this long-lived radionuclide (2.14 x 10^5 year half-life) has been of concern to the nuclear-waste community for some time. A basalt repository may provide a unique control for its mobility.

Future Activities

The pair of ^{99}Tc tests will continue for their planned 3-month duration. We plan also to start tests that include canister material in order to assess whether metallic iron can reduce the ^{99}Tc concentration even further.

A set of tests similar to the ^{99}Tc-doped glass tests is planned except that ^{237}Np-, ^{239}Pu-doped glass will be used. Run duration and conditions will be identical to the ^{99}Tc-doped glass tests. Both the ^{99}Tc- and actinide-doped glass samples have a PNL 76-68 composition.

Other activities include helping BWIP plan the testing activities for FY 1984, characterizing MCC-supplied fully loaded waste forms using a shielded scanning electron microscope, and evaluating data from tests run in FY 1982.

3.2.6 Structural Barrier Materials (AP-05-15-05 - 80752)

The Structural Barrier Materials Task has as its objective the characterization and recommendation of metallic materials for use as structural barrier components in high-level nuclear waste packages for a Hanford basalt repository medium.

Summary

Two major material degradation modes are addressed: general (uniform) corrosion and stress-corrosion cracking. The general corrosion is being determined in autoclave studies with and without the imposition of

gamma irradiation. Resistance to stress-corrosion cracking is being determined by hydrothermal slow-strain-rate studies. Primary emphasis is currently being placed on cast and wrought ferrous materials, as the data obtained to date have indicated that these materials are satisfactory in low-ion-strength basaltic groundwaters.

General Corrosion

In the general corrosion study, specimens of ductile cast iron, 2 1/2% Cr-1% Mo cast steel, and 1 1/4% Cr-1/2% Mo cast steel have been exposed to flowing simulated Hanford basalt groundwater at 150 and 250°C. After 12 months exposure the corrosion rates of all of these materials are <0.25 mil/yr, expressed in terms of linear kinetics. In addition, specimens of 1025 cast steel and 1020 wrought steel have been exposed under the same conditions. After 3 months of exposure, the 1025 cast steel and all but one of the wrought steel specimens have exhibited corrosion rates $\leq$0.27 mil/yr. One wrought steel specimen was heavily etched and pitted, with a metal loss equivalent to 2.8 mil/yr. Additional exposure will reveal whether a susceptibility to nonuniform corrosion exists in this material.

Irradiation-Corrosion

The irradiation-corrosion tests are performed similarly to the general corrosion tests, except that the test autoclave is exposed to ^{60}Co gamma irradiation. Test data are available to 11 months exposure at 250°C and 2 x 10^5 rad/hr on cast ductile iron and 2 1/2% Cr-1% Mo cast steel. The (linearized) corrosion rates were 0.36 mil/yr for the ductile iron and 0.13 mil/yr for the cast steel after this exposure period. Corrosion rates obtained in this study were initially high, compared with data for nonirradiated-specimens; however, the corrosion rates have steadily decreased with time until they are not substantially different from those obtained without irradiation.

In a separate study, the corrosion rates and hydrogen pickups of specimens of titanium alloys Grades 2 and 12 (0.062 in. and 0.032 in. thick, respectively) were determined after a total exposure of 10 months to Hanford basalt groundwater at 250°C and irradiation dose rate intensities of 1 x 10^4 to 1 x 10^6 rad/ hr. The corrosion rates were found to be insignificant, i.e., <1 mm penetration in 1000 years. The hydrogen pickups were substantial, however. Hydrogen pickup was found to increase with radiation dose. At the highest levels of radiation intensity, increases in hydrogen concentration >100 ppm were found in specimens of both alloys. The Grade 12 alloy was more resistant to hydrogen pickup than the Grade 2 (commercial purity) alloy.

Slow-Strain-Rate Tests

The slow-strain-rate behavior of 1020 wrought steel in air has received the major attention in the tests completed to date. The air environment tests have been performed at 150°C and at strain rates of 10^{-4}/sec and 2 x 10^{-7}/sec. The wrought steel has been tested in two textural directions. The air studies are intended to provide "inert environment" elongation, reduction-of-area, fractographic, and tensile strength data for comparison to the same data obtained under hydrothermal groundwater environmental con ditions. (These tests will begin in late April.) A diminution of ductility or tensile

strength, relative to the air environment tests, or fractographic evidence of brittle fracture, would indicate a susceptibility to environmentally enhanced cracking.

3.2.7 Sorption Studies

The objective of this work is to obtain radionuclide sorption-desorption kinetics and equilibria for key radionuclides in the presence of each of the geologic materials that the radionuclide contaminated groundwater might contact. This includes basalt, basalt flow top and secondary minerals from the Umtanum flow and the Middle Sentinel Bluff. Because basalt is a dominant component of the waste package backfill, data from these experiments will complement the basalt-bentonite multicomponent solubility studies to be conducted at Rockwell in FY 1983. Rock materials used in this study will be supplied by the BWIP Waste Package Department.

Technical Progress

Sorption kinetics studies were completed on Cohassett basalt for neptunium, technetium, uranium, selenate-selenium, selenite-selenium and lead. Lead and carbon kinetics also were determined for Umtanum basalt and Umtanum basalt flow top. At least three initial radionuclide concentrations were used under oxidizing and reducing conditions at 60°C in GR-3 synthetic groundwater. A typical kinetic loading curve for uranium on Cohassett basalt is shown in Figure 3.17. Uranium as U(IV) sorbs readily with nearly complete uranium removal from solution under reducing conditions (Eh = -150 mV). Carbon, on the other hand, sorbs very little as HCO_3^- and CO_3^{-2} under any conditions from GR-3 formulation at pH 9.75.

Four basalt columns are now under way at 50 to 80 psig backpressure to avoid gassing due to the HCO_3^- content of GR-3. There are Cohassett and Umtanum basalt columns with radium and a Cohassett basalt column with uranium, all under oxidizing conditions. A single column of Umtanum basalt is being run

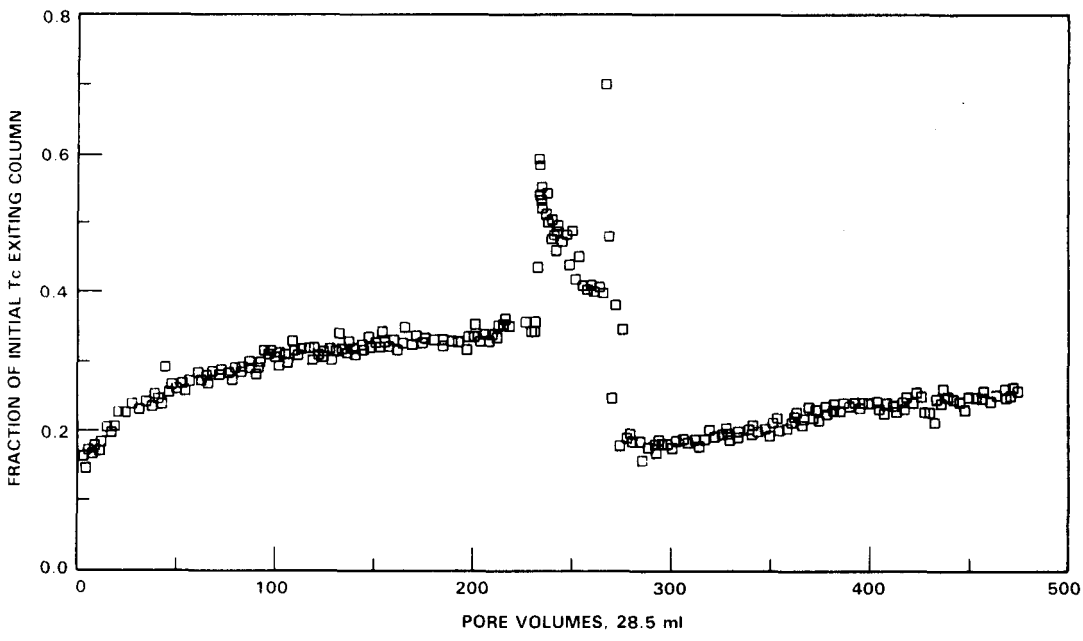


FIGURE 3.17. Uranium Sorption Kinetics on Cohassett Basalt from Reducing GR-3 Groundwater

with technetium under reducing conditions at 80 psig to avoid column gassing due to the hydrazine used to create the reducing environment. The results are shown in Figure 3.18. The fluctuations in C/C_0 were caused by changes in solution Eh over a relatively long period of time. All of these columns are still operating and will be desorbed at some future date with nonradioactive GR-3 synthetic groundwater.

Several static experiments with Cohassett basalt at 150°C were completed, including technetium, plutonium, uranium and neptunium. Equal weights of Umtanum basalt and backfill bentonite, and Umtanum basalt, bentonite and copper were used with technetium and uranium to produce curves of radionuclide in solution versus time and chemical analyses of the major constituents in solution. The radionuclides were usually removed readily from solution by other basalt reactions ongoing at 150°C.

3.2.8 Repository Seal Design Engineering (AP-05-15-05 - 80751)

The FY 1983 objectives of this program are to 1) identify and evaluate the state-of-the-technology related to the degradation of proposed repository sealant materials and 2) develop a preliminary strategy for a statistically valid testing plan and will ensure the adequacy of repository seals.

Technical Progress

Activities on this project were started during the latter part of the reporting period, i.e., February 1983. Literature searches were initiated for pertinent information in the areas of 1) degradation of ancient cementitious materials, 2) degradation phenomena in modern cementitious materials, and 3) extrapolative methods for time-limited data bases.

The review of the literature for degradation in ancient concrete-like materials

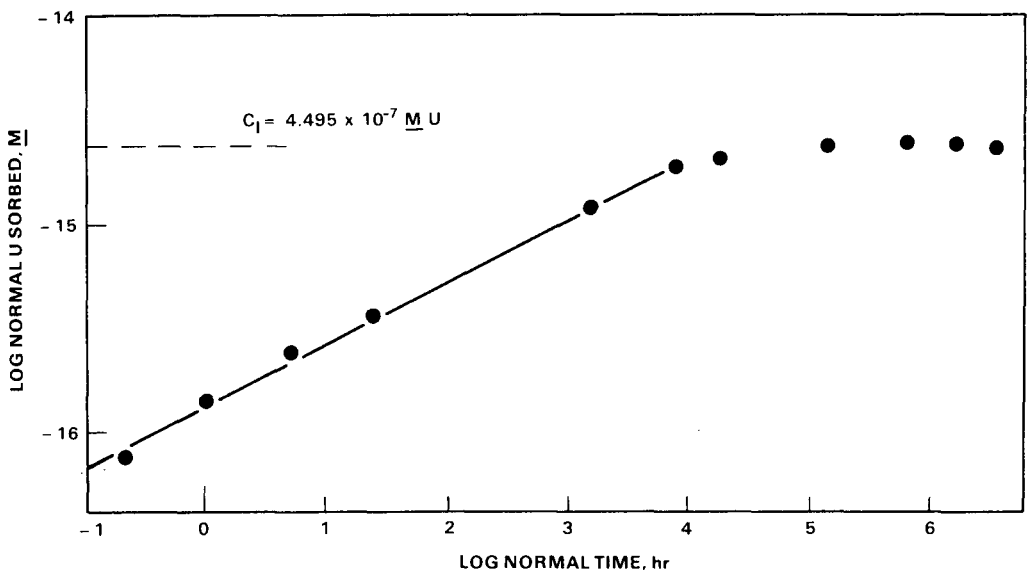


FIGURE 3.18. Sorption of Technetium by Umtanum Basalt from Reducing GR-3 Solution at 60°C

indicates that the major change which has occurred in these materials is carbonation by, primarily, carbon dioxide in the air. In some cases the carbonation of calcium hydroxide in these lime-base materials to calcite was 100% after exposure to air for about 2000 years.

The computer searches of the available literature in the areas of chemical and mechanical degradation of the properties of concrete due to increases in temperature are nearly complete. The key areas of degradation of concrete that are related to exposure to elevated temperatures were determined to be: 1) changes in composition of the original mineral species, 2) changes in relative amounts of the original mineral species, 3) changes in thermal properties of the individual mineral species that are undergoing change, 4) chemical reactions between the original mineral species themselves and other adjacent media, e.g., structural steel, water, or basalt, and 5) volume changes associated with the chemical changes that occur in the original mineral species. All of these changes can affect the strength properties of the concrete, and taken together, they can be expected to affect the permeability of the concrete. The extent of the effect is yet to be determined.

3.3 NEVADA NUCLEAR WASTE STORAGE INVESTIGATIONS (NNWSI) (AP-05-15-10 - 1690/1)

3.3.1 Waste Package

The primary objective of this research is to understand elemental release from the waste form, both with and without the presence of a metal, in a tuff repository environment.

Summary

Research on the Lawrence Livermore National Laboratory contract has been ongoing for about 4 months, and only preliminary results are available. The experiments were designed to evaluate interactive effects between PNL 76-68 waste glass, tuff rock, and 304L stainless steel, the anticipated waste package constituents in a tuff repository. The results to date show that the addition of tuff rock and 304L stainless steel causes no significant effect on elemental release from the 76-68 glass waste form.

Technical Progress

For this summary of current progress, too few analyses have been completed to allow firm conclusions. Such conclusions must await completion of the test matrix and analyses of the leachates, solid samples, and filter cones.

All reported results were obtained by static leach testing following a procedure similar to MCC-1. Presently, only solution analyses (after filtration through 18-Å filter cones) are reported. In many cases only short-term data consisting of two or three data points are available.

Data are reported as Tests 1, 2, or 3. These tests are described as follows:

Test 1. 76-68 glass in tuff groundwater (Well J-13) at a sample surface area-to-solution volume ratio (SA/V) of 10 m^{-1}.

Test 2. 76-68 glass + tuff rock + tuff groundwater with an SA/V = 10 m^{-1} and with monolithic rock. The

rock geometric surface area equals that of the glass, and the actual rock surface area (N_2 BET) equals ~0.25 m^2, which compares to ~2 x 10^{-4} m^2 for the glass sample. Thus the system should be rock dominated.

Test 3. 76-68 glass + tuff rock + 304L SS. This test is identical to Test 2 except for the addition of a piece of monolithic metal with surface area equal to that of the glass sample.

This represents only part of the test matrix. Variations in SA/V ratios and in constituent surface area ratios are included in the complete matrix. The use of powdered rock and glass will be necessary to achieve higher surface area ratios. Planned test times include 3, 7, 14, 28, 56, 91, and 182 days.

Data Summary

The current data can be summarized as follows:

- The effect of metal was not evaluated by itself. Therefore, comments on the effect of metal cannot easily be made. However, the effect of metal plus rock can be commented upon.
- There was a general trend of increasing pH with increasing temperature for 76-68 glass in tuff water. The pH range was from about 8 at 40°C to 9 at 150°C. Both 70 and 90°C were similar at 8.5. The addition of tuff rock and/or 304L SS did not affect pH significantly.
- The more mobile species of sodium, potassium, cesium, molybdenum, and boron behaved similarly with one exception. In general, release from 76-68 glass into tuff water increased with temperature; and addition of rock and/or metal (Tests 2 and 3) resulted in decreased solution concentrations at both 90 and 150°C. Potassium is the exception--the temperature dependence for Test 1 is not strong and the addition of rock and/or metal (Tests 2 and 3) results in enhanced solution concentration. Potassium concentrations are low in the glass and, therefore, perhaps the tuff rock is having a significant effect. Apparently the presence of the rock results in less glass reaction due to silicon additions to the water, as evidenced by the solution concentrations of boron and the other more mobile species.
- In general, the alkaline earth elements decrease in concentration (Test 1) with increasing temperature. This is probably due to pH increases and resulting solubility decreases with increasing temperature. The addition of other constituents (Tests 2 and 3) results in either no effect or in increased solution concentration. The rock composition will have to be determined to resolve the specific alkaline earth behavior. Tuff composition is currently being evaluated.
- The more immobile species such as aluminum, silicon, uranium, and iron are not so easily categorized and should be evaluated individually. Aluminum is present in the glass in such small quantities (actually as an impurity) that its behavior is difficult to follow. Iron has such low concentration in solution that analyses are difficult and have large error bars. In general, all the elements increase with increasing temperature (Test 1) except uranium, which has an inverse solubility and therefore decreases in solution concentration with increasing temperature. The addition of other constituents (Tests 2 and 3) tends to increase solution

concentrations of aluminum and silicon and has little effect on iron and uranium.

Although data are not currently sufficient to warrant detailed analyses or strong conclusions, the presence of either tuff rock or 304L SS does not appear to affect glass leaching detrimentally.

3.3.2 Structural Barriers

The objective of the Structural Barriers Program is the characterization of metals considered potentially useful as structural barrier materials in high-level nuclear waste packages intended for a Nevada tuff repository.

Summary

Two major metal degradation modes are being addressed: general (uniform) corrosion and stress-corrosion-cracking. General corrosion rates are being determined in autoclave studies with and without the imposition of a high-intensity gamma radiation field. Resistance to stress-corrosion cracking is being determined by hydrothermal slow-strain-rate studies. Primary emphasis is currently being placed on 1025 wrought steel, wrought 9% Cr-1% Mo steel, and 304L stainless steel. Investigations of cast steels and cast ductile iron, begun last fiscal year, are being phased out.

General Corrosion

The general corrosion study has, until now, emphasized determining the corrosion rates of 2 1/2% Cr-1% Mo cast steel, 1 1/4% Cr-1/2% Mo cast steel, and ductile cast iron in J-13 well water at 250°C. A layer of crushed tuff rock from the Bullfrog Horizon was placed in the bottom of the autoclave. After 6 months exposure, the ductile cast iron exhibited the highest corrosion rate, viz., 0.54 mil/yr; the two cast steels generally showed <0.20 mil/yr penetration. Magnetite, Fe_3O_4, and nontronite, an iron-aluminum silicate hydrate, were the principal corrosion products formed. This test has been terminated.

The general-corrosion emphasis is now being placed on 1025 wrought steel and 9% Cr-1% Mo wrought steel. (Wrought 304L stainless steel is being procured for eventual evaluation.) The general corrosion study is being conducted at 150°C, with crushed Topopah Spring tuff rock half-filling the autoclave and completely covering half of the sample array. The aqueous medium is flowing air-sparged J-13 well water. The first data from this test will be available in the next reporting period.

Irradiation Corrosion

Two irradiation-corrosion autoclave facilities are currently being set up to test the three candidate structural barrier materials in J-13 well water and crushed Topopah Spring tuff at 105 and 150°C, and ^{60}Co gamma at radiation-intensity levels of ~4 x 10^5 rad/hr. The tests will begin in April 1983.

Slow-Strain-Rate Tests

The slow-strain-rate behavior of 1025 wrought steel has received the major attention in the tests completed to date. These studies are intended to provide "inert environment" elongation, reduction-of-area, fractographic, and tensile-strength data for comparison to the same data obtained under test conditions relevant to a tuff repository. These groundwater tests have begun using 1025 wrought steel, but the data have not yet been

anayzed. Specimens of 304L material, currently on order, will be tested when they arrive on a top-priority basis.

3.4 OFFICE OF NUCLEAR WASTE TERMINAL STORAGE INTEGRATION (ONI)

3.4.1 Simplified System Performance (AP-05-15-15 - 80742)

The objectives of this program are:

1) *Develop and document a simplified simulator containing three linked components: a nuclide source term, the analytical solutions to contaminant transport developed by Dr. T. H. Pigford, and a dose-reaching-the-biosphere calculation.*
2) *Review the first phase results of INTRACOIN and develop explanations for significant inconsistencies among the entries supported by DOE funding.*
3) *Work in a review capacity to provide ONI with comments on the consistency of the field data, the conceptual models, and the conclusions of modeling studies conducted by the NWTS site contractors and submitted to NE22.*
4) *Provide systems analysis of the effect of including TRU waste in a repository.*

Summary

The National Waste Terminal Storage (NWTS) Program is developing performance assessment methods for high-level waste disposal systems; domains of interest include the waste package, the repository, and the far-field surrounding the repository. Studies have been conducted defining the inventory of nuclides to be stored in waste packages within a repository. Models of the waste package, elements of the repository, and far-field have also been further developed. However, the models of specific phenomena for the different domains are still to be coupled to provide an integrated analysis capability. Also, some phenomena have yet to be analyzed for their sensitivity to key parameters.

It is recognized by all involved that coupling of the most complex analyses is precluded by computer hardware constraints. Nevertheless, some form of coupled analysis is necessary to provide gross estimates of system response to alternative source-term and contaminant-migration scenarios. The system response to variation of specific parameters may also be evaluated.

Simplified Simulator

The project has developed a simple model of a high-level waste disposal system. The simplified simulator's purpose is to mathematically describe a disposal system's isolation characteristics. The model, as applicable, can be used in analytical calculations on sites and repositories in support of benchmarking. It can perform bounding-type calculations to show the effect of design variations, the uncertainty in site characteristics, and the importance of modeling or ignoring specific phenomena. Although limited to the simulator's validated range, such a model will provide valuable insight into the response of the waste storage system.

The model can look at a variety of performance measures, a valuable capability because it is uncertain what requirements will be imposed on the isolation system by regulations. Some would use a maximum dose-to-the-individual calculation to provide this final measure; others would use a maximum release criterion. The latter type of analysis removes the variety of assumptions which are required by the dose-to-the-individual calculation. The trade-offs of the dose-to-the-

individual and release criterion approaches can be evaluated using the integrated model.

The computer programs comprising the Simple Code for Predictive Evaluation (SCOPE) system can be separated into three groups:

1. The BAIRN system consists of four programs that account for the transport of radioactive waste in a groundwater system from the repository to the bisophere.
2. Program EXPOSE uses the results of BAIRN to calculate the integrated release of nuclides from the waste in accordance with proposed Environmental Protection Agency (EPA) regulations.
3. The codes PABLM and ALDOS also use the results generated by BAIRN, but they provide an explicit and detailed estimate of the dose to the individual.

In solving for the transport of nuclides, BAIRN specifically uses analytical solutions to the equation for one-dimensional solute transport for up to a three-member decay chain. Analytical solutions for single nuclides are derived by BAIRN 1; 2- and 3-number decay chains are modeled by BAIRN 2 and BAIRN 3, respectively. Principal simplifying assumptions for BAIRN's analytical solutions are:

- Sorption may be represented as in secular equilibrium.
- All elements have infinite solubility.
- Radioactivity is released into groundwater by a band release.
- A uniform one-dimensional flow path is used.

BAIRN 4 approximates chains of four or more nuclides by combining the results from the one-, two- and three-member chain solutions. The results for BAIRN 4 are written into a file that is compatible with exposure and dose codes.

Programs ALDOS and PABLM are used to calculate internal radiation doses to the individual from radionuclides in food products and external radiation doses from radionuclides in the environment. Both programs use the results of BAIRN as well as input from the user for calculations. The results from both programs are similar; they differ mostly in operational details.

The program EXPOSE uses different criteria to estimate the potential hazard to man from the transport of radioactive waste to the biosphere. EXPOSE uses the results of BAIRN 1, BAIRN 2, and BAIRN 3 to sum the curie content released from an aquifer over time. These numbers are compared to criteria developed by EPA for estimating hazard.

The simplified simulator SCOPE is being documented and undergoing verification tests of the simulator components to ensure proper performance of the input, output, and transfer operations of the simulator structure. Other tests will include bounding-type analyses that vary the repository and site characteristics and the model components employed. This is expected to identify conditions under which selected system parameters are important to define system performance. A significant portion of this activity will be devoted to a parametric anaysis of TRU waste placed in four host media: salt, basalt, tuff, and granite.

INTRACOIN

The DOE has sponsored four independent efforts to model test problems defined in the INTRACOIN exercise. Contractors were PNL, INTERA Environmental Consultants, ONWI, and Dr. T. H. Pigford of the Lawrence Berkeley Laboratory. Several differences or inconsistencies in results require explanation.

They could arise from the independent interpretation each investigator lent to the INTRACOIN problems or from the modeling approaches taken.

In INTRACOIN 1, a major difference between the PNL and the Berkeley methods in the boundary condition at the discharge point occurred. PNL used open boundary conditions in which back dispersion from the discharge point is not possible, whereas Berkeley used an infinite boundary and simply sampled the waste stream to measure release. In the latter case, back dispersion is possible. We are examining the effects of open versus infinite boundary conditions relative to mesh description on the INTRACOIN 1 results.

A more fundamental question arose at the INTRACOIN 2 meeting. In a two-dimensional example, significant contamination was found upgradient of the source and also more dispersed to the sides than expected. This was generally ascribed to be the result of a Fickian dispersion model of transport.

Currently, a critical technical issue of groundwater transport modeling is the correctness and adequacy of the classical convection-dispersion equation, when applied to actual field conditions. That fundamental phenomenological equation, based on an extension of Fickian diffusion concepts, is presently the basis of most contaminant transport models. The consensus among groundwater transport researchers is that the classical equation does not always appropriately describe field-scale hydrodynamic dispersion.

Because there is considerable technical concern about the influence that incorporation of non-Fickian field-scale dispersion behavior has on transport predictions, we are conducting an initial study of Fickian versus non-Fickian dispersion to determine their relative influence.

Review of Performance Assessment

This effort is designed to provide technical support for reviewing performance assessment capability at the NWTS sites. Additional effort may address the completeness of performance assessment capabilities at sites and the need for compatible assessment capability. Two efforts are under way. In the first, PNL is reviewing the radiation exposure pathway models for the radionuclides ^{14}C and ^{129}I and the dose factors per unit intake for ^{237}Np. The mechanisms of environmental transport, the fate, and the dosimetry of several radionuclides are objects of scientific debate and of periodic modeling revisions. For ^{14}C our model's present approach for estimating uptake from water by plants results in overestimates of dose, especially when modeling long-term releases from repositories through groundwater. Some investigators have suggested that for ^{129}I, dilution by naturally occurring stable sources, homeostatic limitations on human uptake, and natural removal from the biosphere would significantly reduce potential radiation doses. Present models do not adequately address all of these factors. The International Committee on Radiological Protection (ICRP) has recently increased the factor for human assimilation of ^{237}Np by two orders of magnitude over that previously accepted. Oak Ridge National Laboratory (ORNL) had previously increased the factor for neptunium by one order of magnitude for use in their codes, and this revision was adopted by the EPA. Some experts believe that the ORNL estimate is better than the new ICRP recommendation.

In this effort available ^{14}C models are being reviewed. A more realistic model is being developed and incorporated into the PNL dose codes PABLM and DITTY, based either on the literature review or on a completely new formulation that avoids the weaknesses of the present version.

The literature on ^{129}I in the environment is also being reviewed, and attempts will be made to quantitatively adjust the present model to account for natural dilution, human uptake limitations, and environmental removal if deemed necessary. Appropriate model revisions will be incorporated in the programs PABLM and DITTY.

Finally, the literature is being reviewed on the dose factors for ^{237}Np. Areas of disagreement between the factors selected and ICRP recommendations are being highlighted and the arguments for and against each position given.

In the second part of this effort PNL is reviewing the Nuclear Regulatory Commission's (NRC) performance assessment approach. The NRC is developing, through independent but parallel efforts, a performance assessment method for evaluating geologic disposal of radioactive waste. This method, being developed primarily by the Sandia National Laboratories, will be used to assess the long-term performance of deep geologic repository sites proposed by DOE for licensing. There is a need to compare the DOE's and NRC's methods to determine their compatibility and to define any significant differences that should be addressed now to avoid problems in licensing.

PNL is performing a comparative review of the two methods, focusing on defining differences, potential data collection, and deficiencies in DOE's efforts at characterization of site, waste and repository data that will be required by the NRC method.

Due to the large number of NRC documents, the number of independent DOE sites, and funding limitations, PNL is limiting the review to groundwater flow and transport models, and pathways-to-man and dose (environmental) models used in the NRC method. These two areas make up the bulk of the NRC method, and with the possible exception of NRC's disruptive scenario development, can be considered to be the likely areas of major inconsistencies. Areas not included in this review that may be evaluated at a later time include: disruptive scenario-selection procedures, preclosure accident analyses, geochemical and thermal formulations, and rock mechanics.

TRU

The overall performance of a terminal waste repository depends on the effectiveness of multiple barriers, one of which is the waste package (waste form and other structural barriers). The primary purpose of this effort is to examine the requirements on the effectiveness of the waste-package barrier (specifically for transuranic (TRU) waste). A second purpose is to examine the requirement on the effectiveness of the waste package if no allowance is made for the sorptive properties of the geologic medium through which the waste is transported by groundwater. (This is a test of the requirement that the multiple barriers be independently effective.) The measure of the effectiveness of the waste package is the release time, i.e., the period over which the waste package releases the radionuclides. The measure of the performance of the repository system is that dictated by EPA's regulation 40 CFR 191 (Draft 21).

The EPA regulations provide an isotope-specific basis for determining the maximum allowable radionuclide release to the "accessible environment" in the first 10,000 yr after repository closure. The basis differs depending upon whether the waste is classified as high-level waste (HLW) or TRU waste, both of which were analyzed. Because the regulations and the physical processes are both linear, and therefore additive, this approach gives information applicable for both.

The relationship between EPA's release limits to the environment and the release rates from the waste package has been established via computer simulation of isotope transport through the circulating regional aquifer from the repository cavity to surface waters (the entry point to the accessible environment). A parametric approach was used over various combinations of release rates and other key repository-system performance parameters (including groundwater velocity, transport distance, and net sorptive properties of the media) to determine the release rates from the system over a broad range of conditions. This approach provides "expected" values of release rates as well as an indication of the sensitivity of the accessible rates to variation in (or uncertainty regarding) other repository system performance parameters. The method was applied to transport through materials representative of basalt, tuff, and salt and also through a medium with no sorption. Other assumptions basic to the calculations were:

1. The radionuclide source inventories were the entire HLW and TRU wastes from the processing of 72,000 MTHM of spent fuel as defined in DOE/ET-0028.
2. The migrating nuclides were in linear sorptive equilibrium at all points.
3. The transporting nuclides were dissolved in groundwater in trace concentrations such that there is no inter- or intra-species competition for sorption, nor are solubility limits ever reached.
4. Estimates of the equilibrium sorption coefficients, K_d, were used in most cases; oxidizing conditions were assumed.
5. The groundwater velocity and axial dispersion coefficient were constant for any single parametric calculation.
6. Transverse convection and dispersion were ignored.
7. The release of nuclides from the repository was a constant rate band except for the effects of the radioactive decay (decaying band).

The conclusions from this analysis can be summarized as follows:

- Disallowing (ignoring) sorption places significant demands on the waste form/package/engineered cavity system, requiring release rates as low as 10^{-8}/yr or less from the repository near-field in order to comply with proposed regulation 40 CFR 191 for a groundwater travel time of 1000 yr.
- The requirements imposed on the waste package by 40 CFR 191 do not differ significantly for TRU and HLW waste package, nor are these significantly different among the sorptive media.
- If the sorptive properties of the geologic media are accounted for, a release rate from the repository of 10^{-5}/yr is adequate to ensure compliance with the proposed regulation for all three sorptive reference media for a groundwater travel time of 1000 yr.

3.4.2 RECON Repository Cost Analyses

The objective of this program is to develop a verified repository economics model and data base to evaluate the cost impacts of preliminary repository design trade-offs and to estimate overall waste disposal costs.

Background

The RECON repository economics model was originally developed in 1981 as part of a task to evaluate the economics of repositories and alternative media. In 1982, further development was completed, including calculations of operating requirements and costs as functions of process rates for different repository operations. In addition, many minor improvements were made in the program logic. Preparations were made for a report documenting the RECON model at the end of FY 1982.

Technical Progress

During the first half of FY 1983, the program scope encompassed several tasks: 1) expansion of the data base to run the improved RECON model, 2) development of a model as an interface to facilitate use of the model, and 3) development of equations to model interim waste storage costs.

During the first quarter of FY 1983, the interim storage cost equations were developed based on a draft Monitored Retrievable Storage (MRS) Interim Storage document. In accord with project scope, these equations were determined for just one scenario, a co-located MRS facility at a repository site. The results of this task were transmitted in a letter report to the sponsor in January. During December and January, data requirements for the improved RECON model were developed using data sources provided by the sponsor. Verification runs were made with RECON using this data base to compare the results with hand-calculations made by the sponsor. The verification runs were made in a salt medium for spent fuel and high-level waste repositories.

Costs for the spent fuel repository predicted by the RECON model were $5.5 billion versus costs estimated by the sponsor of $5.4 billion. The RECON estimates for the high-level waste repository were about 12% higher than the sponsor calculations. An attempt to reconcile the differences for the HLW case was not successful because of the widely different accounting categories used. In January, the sponsor requested that an additional table be developed using a consistent accounting category instead of developing additional verification runs for other media. This additional table, using consistent accounting categories, has been developed and is now being debugged.

To improve the user friendliness of the RECON model, a preprocessor was developed which facilitates input of data by the user. The preprocessor is an interactive program with menus describing the different categories of data inputs. The preprocessor allows the user to either create a complete new data base, or to step through the menus changing only selected data inputs. This task has been completed and the preprocessor is now being used to develop data for the program runs.

Additional improvements to the RECON model have also been completed as part of another DOE project. These allow modeling of the borehole in the wall emplacement concept and also co-emplacement of low heat generating wastes with high-level wastes. All of these model improvements have been documented and will be published in the final documentation and Users Manual. This final documentation report, due to be published in April 1983,

will complete the final milestone for this project in FY 1983.

3.5 OFFICE OF CRYSTALLINE REPOSITORY DEVELOPMENT (OCRD)

Northeast Granite Regional Environmental Characterization Studies (DA-30-30-03 - 80459)

Objectives are 1) to prepare an environmental characterization of the Northern Applachian and Adirondack Crystalline Region--comprised of portions of the states of Connecticut, Maine, Massachusetts, New Hampshire, New Jersey, New York, Pennsylvania, Rhode Island, and Vermont and 2) to prepare an analysis for and recommendations of areas within that region that are suitable candidates for more detailed environmental characterization.

Summary

This program has developed an environmental characterization data base for all or parts of nine (9) Northeast and New England states under the sponsorship of the Office of Crystalline Repository Development (OCRD). A draft Environmental Characterization Report (ECR) was transmitted to OCRD, and comments were received for the final draft. Additional tasks scheduled for the latter half of FY 1983 will proceed.

Technical Progress

Program staff supplied a modified proposal to the sponsor in October 1982 for FY 1983 activities. The funding level was set and the effort will continue through July 1983. The project is managed by OCRD's Site Characterization Department and is a component of NWTS's (DOE) repository site selection process as outlined in DOE's Confidence Rulemaking Statement. Major efforts thus far in FY 1983 have been directed toward 1) developing useful site selection/screening tools for OCRD, 2) responding to OCRD and/or DOE comments on the characterization document, and 3) interfacing with the follow-on contractor. The region of the U.S. covered by the project and study is shown in Figure 3.19.

Highlights for the first half of the fiscal year are as follows:

- A new project monitor, Tom Anderson, was appointed in November for the Office of Crystalline Repository Development (OCRD).
- An existing interlaboratory agreement with Battelle's Human Affairs Research Centers was extended for them to continue demographic work for the project.
- Revised schedule of milestones, dates and deliverables after discussions with sponsor in December.
- A comment copy of the Environmental Characterization Report (ECR) was cleared and delivered to sponsor in January.
- Project staff met with OCRD Project Manager in February to review program and identify areas of the ECR that required modification. An updated ECR was transmitted to OCRD in late February.
- PNL staff met in March with OCRD Project Manager and the Project Coordinator for Dames and Moore (who will take over site characterization work in FY 1984) to assess the environmental data base and to discuss procedures for turning over records.

OCRD comments and suggested changes for the ECR were received March 31, 1983, and are being incorporated into a final draft deliverable April 15, 1983. Work on tasks scheduled for the last half of FY 1983 will proceed.

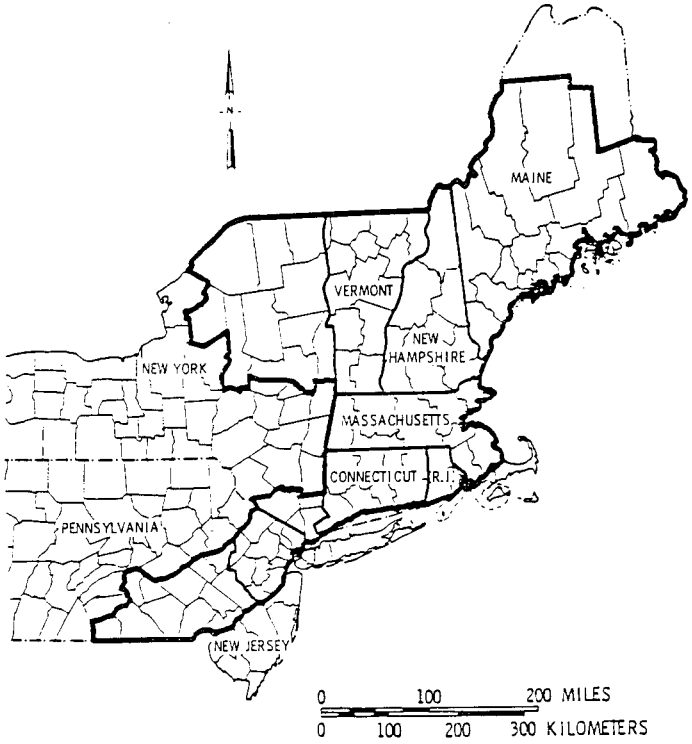


FIGURE 3.19. Environmental Study Region Associated with the Region

4.0 LOW-LEVEL WASTE MANAGEMENT

The objective of the Low-Level Waste Management Program is to develop the technology to dispose of low-level radioactive wastes in a manner that protects the public health and safety.

4.0 LOW-LEVEL WASTE MANAGEMENT

M. R. Kreiter - Program Office Manager
Program Managers: R. L. Brodzinski, T. L. Jones, C. S. Simmons and J. L. Swanson

4.1 SPECIAL WASTE FORM LYSIMETERS - ARID (AP-05-20-15 - 80749)

The objectives are:

1) *To construct and operate a field facility designed to conduct waste form leaching tests that will determine typical source terms generated by commercial solidified low-level waste. Solidification agents tested are cement and polyvinyl styrene.*
2) *To identify through field and laboratory research, the chemical and physical processes that control the concentration of radionuclides in the soil solution surrounding the waste.*
3) *To determine adequate methods for representing source-term boundary conditions in radionuclide transport models.*

Summary

Design of the field test facility is in progress. Final engineering drawings are near completion. Initial cost estimates for construction range from $80K to $130K. The environmental assessment has been completed. Based on the preliminary design, the calculated dose, resulting from exposure to the commercial low-level waste material, is low and meets radiological safety requirements.

Shipment of waste materials from Brookhaven National Laboratory (BNL) will be initiated in late April and is expected to be completed by mid-June. A purchase order for the waste material was completed, and contractual arrangements are being negotiated with BNL. Analytical requirements for waste-form characterization have been specified.

Lysimeter Study

The Special Waste Form Lysimeters Study is a new study at PNL designed to measure the release and migration of radionuclides from solidified commercial low-level waste. A field test facility will be built to contain selected waste forms in specially constructed lysimeters. Data will be gathered that relate laboratory measurements of waste-form characteristics to release and migration rates observed in the field. Results obtained for this arid site facility and those from the humid site located at Savannah River Laboratory (SRL) will be compared. Field leaching tests will be conducted on actual commercial waste forms obtained from Brookhaven National Laboratory. Waste forms tested will include material from power reactor waste streams that have been solidified in cement and Dow polymer. The waste form will be characterized by standard laboratory tests such as MCC-1 and the IAEA leach test to obtain estimates of radionuclide release. These estimates will be compared to leach rates observed in the field. Solubility studies will also be conducted to determine the amount of contaminant available for transport after release.

The field test facility has been planned and preliminary drawings completed. Figure 4.1 shows the cut-away and top view of the test facility. The commercial waste materials are to be contained in 55-gal drums

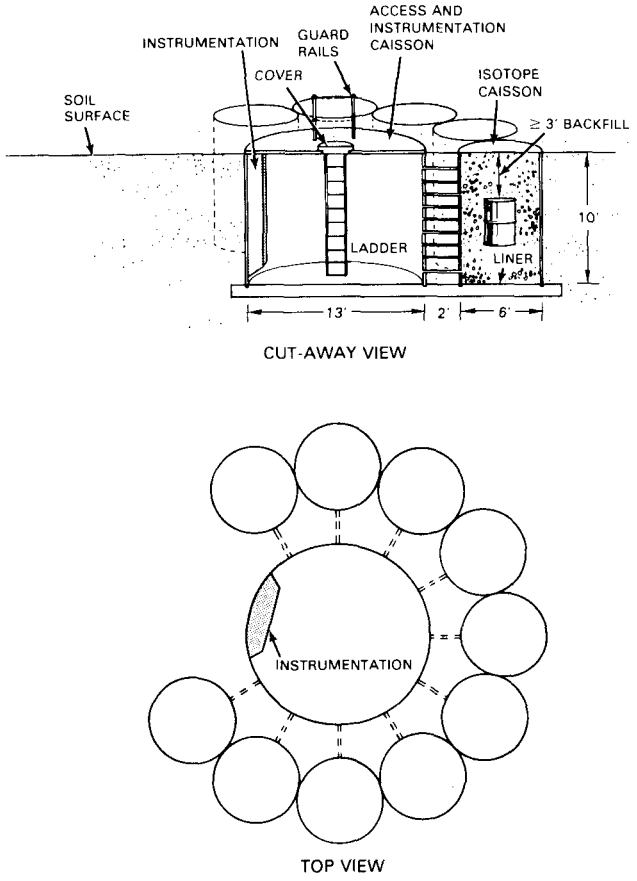


FIGURE 4.1. Preliminary Engineering Drawing of Special Waste-Form Lysimeter Test Facility

placed in closed-bottom lysimeters 10 ft deep and 6 ft in diameter. The lysimeters will be backfilled with soil so that at least 3 ft of backfill material covers the top of the waste drum. The soil material will provide radiological shielding as well as a medium for sorption of soluble radiochemicals that may leach from the waste drums. Suction candles (ceramic cups) placed in the bottom and around the waste drums will be used to periodically collect soil solution for chemical analysis.

A central caisson (Figure 4.1) will provide access to the individual lysimeters for selectively sampling and to instrumentation. Moisture content and temperature of the soil surrounding the waste form will be monitored periodically, and radiological analysis will be performed both in-situ and with selective sampling of the lysimeters. It is anticipated that this will be a long-term (10 year) study, so the instrumentation and lysimeter design will be planned accordingly. Preliminary cost estimates for the facility described in Figure 4.1 range from $80K to $130K. The larger estimate was based, in part, on considerations to place some of the waste forms in lysimeters that extend to depths greater than 10 ft. Due, however, to significantly increased costs for lysimeters 15 to 20 ft deep, it is likely that units 10 ft deep will be used.

Negotiations with staff from BNL have resulted in procurement and pending delivery of waste samples, with completion of the shipment expected by mid-June. Samples will be obtained that are close to those obtained for the SRL lysimeter project. The analytical requirements for the waste forms have been specified to BNL and they are to provide detailed radiochemical information concerning the waste material.

During the remainder of FY 1983, the lysimeter facility will be constructed and the monitoring system installed. Provided there are no unforeseen difficulties in procuring waste samples, they should be installed no later than the first quarter of FY 1984.

After the facility is operational, the site will be closely monitored for any signs of radioactive release from the waste samples. If any release is detected, then soil and soil solution samples will be taken for analysis. Analytical chemistry will include analysis of macro- and trace elements using inductively coupled plasma emission spectroscopy (ICP) and atomic adsorption (AA) techniques. Anions will be analyzed using ion chromatography (IC) and radionuclides by low-level gamma and x-ray counting techniques. This will provide the Low-Level Waste Management Program with direct field results regarding leach and migration rates from actual commercial solidified waste.

In addition to the field monitoring, which is the central purpose of the task, a series of laboratory testing and geochemical exercises is planned. These tests will help predict expected leach rates and help interpret any field results obtained. Specifically, the goals of the laboratory and modeling work will be to identify which mechanisms are controlling the release of radioactivity from the waste form.

4.2 TRANSPORT ASSESSMENT - ARID (AR-05-15-15 - 80033)

The mission of this program is to provide the required technology for use in monitoring radionuclide transport through geohydrologic systems. This technology will assist in the assessment of the safety of buried solid low-level radioactive waste.

Summary

Comparisons were made between laboratory and field observations of the transport of ^{60}Co in Hanford soils. Laboratory column studies indicated that ^{60}Co/EDTA can be highly mobile in Hanford soils. However, the organic complexant, EDTA, apparently dissociates from ^{60}Co in time, greatly reducing the cobalt's mobility. Our field tests confirm that after 3.5 yr there is little evidence of movement of ^{60}Co under the conditions of Hanford soil and climate. These results imply that complexation of ^{60}Co, and perhaps other radionuclides, by EDTA may not be a long-term problem at Hanford low-level waste sites.

Transpiration studies were initiated to investigate the effects that plant water removal has on the water balance at shallow-land burial sites. Water content measurements (by neutron probe) were made at 25 access wells on two selected vegetated sites at Hanford. A complete cover site (short grasses, shallow-rooted) and a sparse cover site (shrubs, deep rooted) were compared. Significant differences in water uptake were observed between the two sites, suggesting that root depth, percent cover, and plant type characteristics are information needed to predict water balance at arid-zone burial sites.

Cobalt-60 Transport Study

A field-scale tracer test using ^{60}Co/EDTA-spiked soil placed in a 1-cm band 60 cm below the soil surface was monitored for 3 yr. During that time, cobalt movement did not exceed 10 cm, as measured by the downwell probe or by destructive sampling. Excavation of the field site revealed some cobalt movement to a depth of 70 cm; the peak concentration, however, remained at its initial depth of 60 cm. Results of the excavation sampling are shown in Figure 4.2. Also shown in Figure 4.2 are the results of a tritium tracer test conducted at an adjacent site. The tritium test was started at the same time as the cobalt test with tritium also being placed at an initial depth of 60 cm. The tritium was sampled in April 1981 (2 yr after tracer placement) while the cobalt was sampled in September 1982 (3.5 yr after placement). The figure shows that the tritium has been transported an average distance of 4 m (peak concentration) while the cobalt peak remains at the original 60 cm. Details of this test and its relationship to our previous laboratory column studies and to work by J. L. Swanson on organic complexant effects on radionuclide mobility were reported by T. L. Jones at the recent Waste Management '83 conference (Jones et al. 1983).

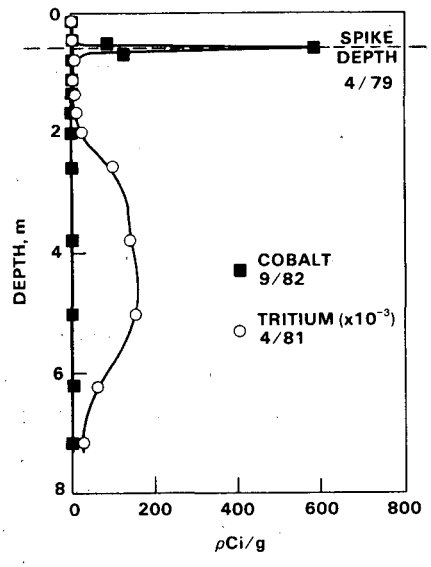


FIGURE 4.2. Comparison of ^{60}Co/EDTA and Tritium Mobility Under Field Conditions at Hanford

It is apparent from these studies that there is a time-dependent effect of complexing agent on ^{60}Co mobility. Both laboratory and field studies confirm that complexed ^{60}Co moves rapidly through Hanford soils when initially added to the soil, but that after 60 to 90 days there is an apparent dissociation of the EDTA complex, resulting in greatly reduced mobility of the ^{60}Co. The implication from our tests is that transport of ^{60}Co and possibly other radionuclides by complexing with EDTA may not be a serious problem at Hanford low-level waste sites.

Field Transpiration Experiments and Simulation Modeling

Effective site selection for low-level waste repositories requires accurate water balance projections for various climatic extremes and surface covers. Mechanisms for water removal from soils can be categorized as evaporation, transpiration, and drainage. Evaporation and drainage have already been reviewed in previous waste management reports, and our efforts during the past 6 months have concentrated on measurement of the transpiration component of the water balance.

In the past, transpiration measurements have seldom been made in plant communities that are not of agronomic importance; thus little data is available from the literature

on water use by native vegetation, particularly for arid-zone sites. In many radwaste management scenarios, burial sites are allowed to revegetate either by design or by passively allowing plants to invade the burial site. Under these conditions, it is important to know the plant characteristics and to measure the transpiration from the cover to accurately assess a complete water balance. Field transpiration experiments have been initiated this year to collect data that will describe the water balance of a typical Hanford area plant community. Data collection has included descriptive data such as plant community structure and soil water status. Vegetation and soil parameters which control transpiration rates also are being monitored. These controlling parameters include the driving forces and pathway resistances to water movement in the soil-plant-atmosphere continuum. Understanding these parameters allows for a broader and more adaptable approach to the modeling of evaporation in the plant communities associated with burial grounds.

Twenty-five neutron access wells were installed on a 5-by-5 grid with 6-m spacing between each well (Figure 4.3). The wells were installed to a depth of 4 m by driving a barrel sampler ahead of the well casing, driving casing flush with the bottom of the barrel sampler, then withdrawing the barrel sampler. This method ensures a tight hole, minimizing void spaces in the surrounding soil and ensuring reliable downwell-neutron probe measurement of soil moisture. Soil samples were subsampled for moisture content, and the remaining material saved for analysis of root density. Soil samples were collected for standard laboratory analysis.

Six neutron access wells were installed in early March on a rectangle, 2 by 7 m, surrounding two large shrubs (Bitterbrush, Purshia tridentata). Access wells were installed to a depth of 4 m, using the same drilling procedure described above.

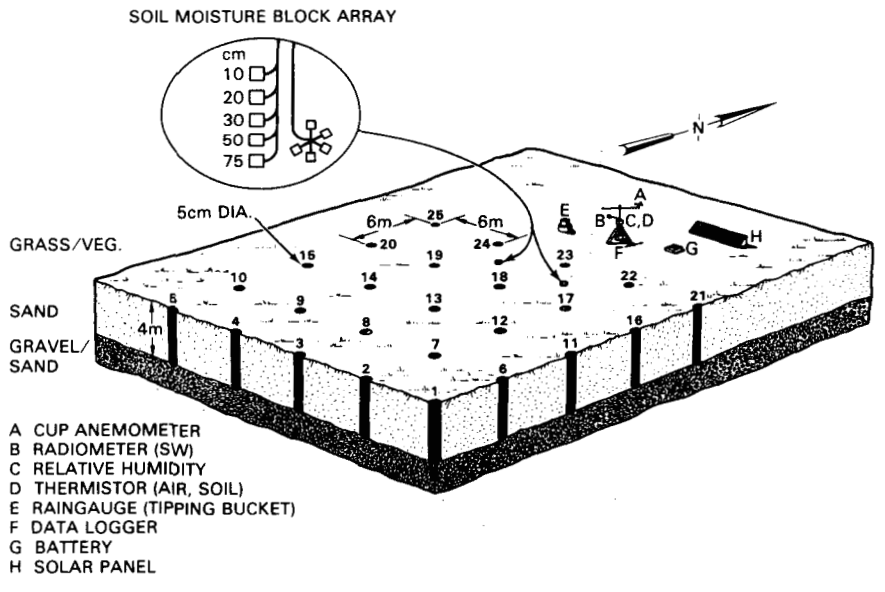


FIGURE 4.3. Transpiration Experimental Site (Grass)

Moisture data are being collected every 2 weeks, with alternate sampling times reaching the full 4-m depth. The soil moisture profile developed as an average of all 25 access wells is presented in Figure 4.4 (curves 1, 2) for data collected during January and April at the grass site. The figure also shows the average water distribution at the brush site (curves 3, 4). Note that the upper profile moisture decreases while the lower increases, indicating redistribution of water downward. The total water in the 4-m profile is ~35 cm at the shrub site and 30 cm at the grass site. Data collected from cores indicate the grass site has twice as many roots as the shrub site. The distribution is quite different, with 100% of the grass roots above 1 m while the shrub site has 20% of the roots located below 1 m. As expected, shrub roots appear to be distributed relatively uniformly with depth.

A composite soil sample collected near the surface at the grass site was analyzed in the laboratory for the parameters indicated in Table 4.1.

Vegetation composition and cover was evaluated by Arid Lands Ecology staff on March 18, 1983. Vegetation was sampled by randomly placing a 0.2 x 0.2-m frame (Daubenmire 1959) at two locations in each 6 x 6-m plot located within the access well grid. Table 4.2 lists the identified species and their respective cover values.

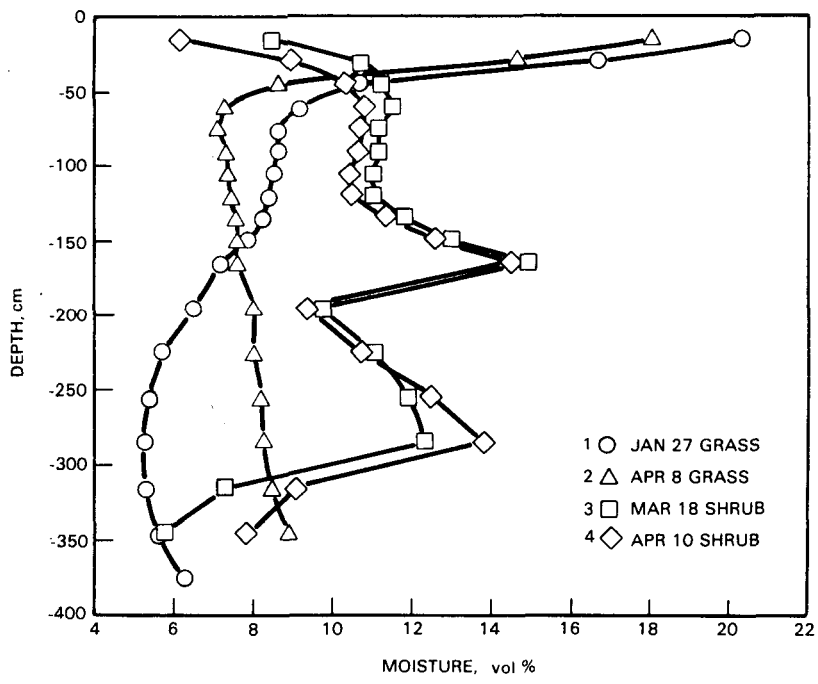


FIGURE 4.4. Moisture Distribution at Transpiration Sites

TABLE 4.1. Soil Properties

Conductivity	180 mmhos
Particle density	2.74 g/cm^3
Permeability	2.49 x 10^{-3} cm/sec
Particle size distribution	80% sand, 17% silt, 3% clay
pH	7.5

References

Daubenmire, R. 1959. "A Canopy-Coverage Method of Vegetation Analysis." Northwest Science 3:43-84.

Jones, T. L., G. W. Gee, J. L. Swanson and R. R. Kirkham. 1983. "A Laboratory and Field Evaluation of the Mobility of Cobalt-60/EDTA." Waste Management '83, ANS-83-01, Tucson, Arizona.

4.3 VERIFICATION INSTRUMENT DEVELOPMENT

(AR-05-15-15 - 80334)

The objective of this research program is to organize, apply, and develop, as required, instrumentation for assaying the radionuclide content of waste packages received by land burial operators, both to verify the data provided by the generator/shipper and to provide the most accurate records possible of buried materials.

Summary

A report titled "Massachusetts Low-Level Radioactive Waste Management Survey" has been used as the model for average United States low-level waste disposal package parameters. From this survey, recommendations are given for the type of containers, the matrix materials, and the radionuclides and their concentrations for fabrication of radioactive waste package standards to be used for assay instrumentation calibration.

A review of all available TRU assay instrumentation technology has been completed, and review of all non-TRU technology is nearly complete. The recommended radioassay system will be given in the final report.

TABLE 4.2. Vegetation Cover at Grass: Site Plant Count (Number) per 0.04 m^2

	Species	Average	Standard Deviation	Range
1.	Litter	61.5	17.7	37.5- 97.5
2.	Moss/lichens	59.6	22.9	0 - 97.5
3.	Poa sanbergii	26.8	15.4	0 - 62.5
4.	Bromus tectorum	34.7	22.3	2.5- 85.0
5.	Draba verna	3.8	4.2	0 - 15.0
6.	Mushrooms	0.2	0.7	0 - 2.5
7.	Chorispora tenella	0.1	0.4	0 - 2.5
8.	Total live plants[a]	125.5	18.9	92.5-175.0
9.	Total vascular plants[b]	65.9	19.6	20.0-115.0
10.	Total cover[a]	187.1	24.3	142.5-237.5

(a) Total live plants = Total cover - Litter.
(b) Total vascular plants = Total cover - Litter - Moss/lichens.
(c) Total cover = 1 through 7.

Waste Receipt Instrumentation Development

The Oak Ridge National Laboratory Program Review Team has directed that standard waste packages should be fabricated based on survey information available from EG&G, Idaho, rather than on field tests at a commercial waste disposal site. This survey information has been provided by EG&G in the form of a final report from Inter/Face Associates, Inc., titled "Massachusetts Low-Level Radioactive Waste Management Survey." The following conclusions are based on data presented in this report.

Those data are extracted from responses to a well prepared and well administered questionnaire from Inter/Face. That this Massachusetts data can be extrapolated to a nationwide average is not immediately obvious. To prepare standard waste packages that will represent actual packages of commercial low-level radioactive waste, it is necessary to know 1) the types of waste containers used, 2) the matrix material composition, and 3) the concentrations of specific radioisotopes present in the waste. This information is available or extractable from the Inter/Face report.

The Inter/Face report generated separate data sets for the years 1979, 1980, and 1981. Although some trends and shifts in disposal practices are apparent, the same conclusions regarding standard preparation were obtained from each data set. The report lists total volume of waste shipped and the number of shipping containers which are identified as Type A and LSA 30- and 55-gal drums, shipping casks with and without disposable liners, and other undefined containers. By subtracting the known volume of drums and casks from the total volume and dividing by the number of other containers, the average volume of "other" containers is found to be greater than 128 ft^3, the volume of a 4 x 4 x 8 ft plywood box. Hence, it is assumed that most "other" containers are 4 x 4 x 8 ft plywood boxes, with a few larger boxes. Therefore, on the average, ~50%, by volume, of the waste is disposed of in plywood boxes, ~33% in 55-gal drums, and ~8% in 30-gal drums, with the balance in shipping casks and other containers. Thus, to represent more than 90% of the waste, standards should be prepared in 4 x 4 x 8 ft plywood boxes, 55-gal drums, and 30-gal drums.

The choice of matrix material is not as clearly defined as the container. Significant fractions of the total waste are described as dry compressible, dry noncompressible, liquid scintillation, slurry/resins/sludges, evaporator concentrates, aqueous liquids, gaseous wastes, and "other." Unfortunately, the "other" category comprises about 60% of the total volume and nonnegligible fractions of the total activity. Consequently, adequate information is not available to simulate the preponderance of waste matrices, and even if it were, it would result in too large a number of permutations for practical considerations. Hence, it is recommended that a matrix material with a density near unity and a matrix material with a density of about 2.5 to simulate cement be used. For lack of better information, the radioactive spikes should be homogeneously distributed throughout the matrix. The low density standards should be fabricated in Ashland Chemical Company water-extended polymer (WEP-662-T), Dow Chemical Company vinyl ester-styrene polymer, General Electric Company Aztech radwaste system polyester monomer, or other similar polymerizable monolith. The higher density standards can be fabricated in the WasteChem VRS™ System asphalt binder, Portland cement, or masonry cement.

A full list of all radioisotopes shipped for disposal is given in the Inter/Face report. Obviously, it would not be prudent to fabricate standards of all 47 nuclides listed. Hence, all radioisotopes falling unequivocally into disposal class A per 10 CFR 61 are disregarded. This classification can be made in most instances based on the small quantities disposed of. The remaining activities are ^{3}H, ^{14}C, ^{55}Fe, ^{60}Co, ^{90}Sr, ^{137}Cs, and transuranics, and these comprise 98.68% of the total activity shipped. At this time, there is no nondestructive analytical technique capable of measuring ^{3}H, ^{14}C, and ^{55}Fe in a sealed waste package. Therefore, standards should be fabricated containing ^{60}Co, ^{90}Sr, ^{137}Cs, and transuranics at concentrations comparable to those in actual commercial waste disposal packages.

A review of all the TRU waste assay technology developed at Los Alamos National Laboratory has been completed. These assay methods and those developed in our laboratories have been evaluated, and a decision has been reached on the appropriate technology for adaptation to a general low-level waste package assay system. Details of this portion of the recommended assay technology will be presented in the final report.

In addition, a document titled The Design and Operation of Nondestructive Solid Waste Measurement Systems at DNPDE has been obtained from the United Kingdom Atomic Energy Authority and has been reviewed to evaluate the design of solid waste management systems used in the United Kingdom to categorize wastes for control purposes.

The final non-TRU assay technology evaluation is also nearly completed, and the details of the entire radiological measurement portion of the package assay system will be given in the final report.

4.4 MOBILITY OF ORGANIC COMPLEXES OF RADIONUCLIDES IN SOILS (AR-05-15-15 - 81034)

The objective of this program is to develop and employ standard procedures for evaluating how and to what extent organic complexants affect the mobility of low-level waste radionuclides in soils.

Summary

Evidence has been obtained for the sorption by some soils of some organic complexant-metal ion complexes themselves. In these systems, therefore, the migration of the undissociated complexes will not occur at the same rate as the migration of water, as had been assumed previously. Other results emphasized even more than previously that all organic complexants should not be considered equally troublesome in waste disposal.

Sorption of Complexes

Another mechanism of importance to radionuclide migration, the sorption by soil of the metal-complexant complex itself, has now been observed in some systems. This observation means that, in these systems, migration of the undissociated complex would not occur at the same rate as the migration of water, as had been assumed previously. This observation is thus very important.

The primary evidence for this phenomenon comes from the different rates of desorption of the radionuclide from the soil by a complexant solution when the radionuclide has been sorbed by the soil in the presence or absence of complexant. Data obtained in the Ni/EDTA/ORNL soil system at an EDTA concentration of 3.5×10^{-6} M and a soil-to-solution ratio of 0.033 g/mL will be used to illustrate this evidence.

When nickel was sorbed onto the soil in the absence of EDTA, and EDTA was then added to the solution, <2% of the nickel was found in solution after 4 and 32 days. When the Ni-EDTA complex was allowed to form before the soil was added, the percentage of nickel remaining in solution dropped rapidly when the soil was added and then remained apparently constant for several weeks (32% of the nickel was found after 4 days and 34% after 21 days). When most of the precomplexed solution was then removed and fresh EDTA solution was added to the soil, the remaining nickel distributed rapidly between the solution and the soil to give the same result (37% of the remaining nickel was found in solution after 3 days and 31% after 11 days). It is concluded that the rapid desorption in the latter case must be due to the fact that the sorbed nickel species is a Ni-EDTA complex, rather than an uncomplexed nickel species as was sorbed in the first case.

Sorption of complexed plant micronutrient species by soils has been observed previously (Norvell and Lindsay 1969; Norvell and Lindsay 1982), but no evidence for this effect with radionuclides has been presented. The distribution coefficient for the Ni-EDTA complex onto the Oak Ridge soil is about 60, which is much higher than the coefficients of <1 calculated for the Fe-EDTA and Ca-EDTA complexes from the data presented by Norvell and Lindsay (1982).

Additional evidence for the sorption of Ni-EDTA complex species by the Oak Ridge soil was obtained in two other ways. In one, the distribution coefficient for nickel measured in precomplexed-type experiments was found to decrease by only a factor of 3 when the EDTA concentration was increased by a factor of 100. A much stronger dependence of distribution coefficient on complexant concentration is expected (and observed in other systems) when the uncomplexed radionuclide is the sorbed species. Another observation that provided evidence for the sorption of Ni-EDTA species was that the disappearance of ^{14}C-labeled EDTA from solution was greater in the presence of nickel than in its absence.

The sorption of complexed radionuclide species observed here must represent a pseudo-equilibrium situation; while the concentrations appear to remain stable over several weeks, the fact that the same situation is <u>not</u> achieved (in this time period) where the radionuclide is sorbed before complexant is added indicates that a true equilibrium position has not been reached. The systems where this sorption occurs thus appear to provide other examples of slow dissociation of the complexes into uncomplexed radionuclide and complexant.

Evidence for the sorption of complexed species has also been obtained in the Co/EDTA/ORNL soil system, and perhaps also in the Ni/EDTA/Savannah River system. Study of this important effect is continuing.

Other Results

Investigations in several other facets of this problem have also yielded important results. These are summarized below:

1. Experimental evidence was obtained to demonstrate the rapid disappearance of citrate from solutions contacting soils; this disappearance had previously been hypothesized.
2. The NTA complex of nickel dissociates more rapidly in a Hanford soil/groundwater system than do the EDTA, DTPA, or HEDTA complexes. NTA-containing wastes would thus appear to be less of a disposal problem than those containing the other complexants.

3. Three commercial decontamination solutions that had been used in decontamination tests on the failed steam generator from the Surrey nuclear power plant exhibited greatly different cobalt-retaining power when the solutions were contacted with soil. This provided added emphasis to the earlier conclusion that all organic complexants should not be considered equally troublesome in waste disposal.

References

Norvell, W. A. and W. L. Lindsay. 1969. "Reactions of EDTA Complexes of Fe, Zn, Mn, and Co with Soils." Soil Sci. Soc. Amer. Proc. 33:86-91.

Norvell, W. A. and W. L. Lindsay. 1982. "Estimation of the Concentration of Fe^{3+} and the $(Fe^{3+})(OH^-)^3$ Ion Product from Equilibria of EDTA in Soil." Soil Sci. Soc. Amer. Proc. 46:710-715.

4.5 GROUNDWATER TRANSPORT MODEL DOCUMENTATION AND EVALUATION GUIDELINES (AR-05-15-15 - 80635)

This program will develop standard guidelines for the documentation and evaluation of groundwater transport models to be applied to low-level waste management.

Summary

Guidelines are being developed to assist potential users of groundwater transport models in selecting appropriate computer codes. Evaluation test cases in the form of analytical solutions and experimental data sets are being identified and recommended to ensure adequate code selection, based on accurate simulation of relevant fundamental physical processes. Code selection depends on identification of the problem objectives, the extent of collectible site-specific data, and development of a conceptual model for the specific hydrologic system. Model evaluation takes into account certain technical issues related to the limitations in modeling capabilities.

Technical Progress

This program, which was originally directed toward transport model documentation guidelines, was recently redirected in October 1982 to address instead model selection by a potential user. The new direction is expected to better meet the modeling needs of the Low-Level Waste Management Program.

A letter report (April 1983) on selection of experimental data sets for use in model evaluation discussed the project's new emphasis. That report pointed out that groundwater transport model selection is primarily a model development problem, depending ultimately on the study objectives of a particular burial site analysis. A major purpose of model selection guidelines will be to overcome inherent limitations of a generic system modeling approach, when applied to site-specific problems. To select an appropriate computer code, which is often incorrectly thought of as the physical model, a user must take the technical responsibility of actually developing a conceptual model for the hydrologic system involved. The user's conceptual model then guides the application of chosen relevant groundwater flow and transport codes. Such codes, once selected, are the numerical implementation of relevant basic equations deemed descriptive of fundamental physical processes active at a shallow-land burial site. To ensure that a correct conceptual model is developed and adequate associated codes are selected, a user must test the completed systems model against

known physical behavior. The known system behavior is expressed by various experimental data sets depicting fundamental physical processes acting in a hydrologic system. Some specific data sets for that purpose were therefore identified.

Before code selection can proceed, three preliminary steps must be completed: 1) definition of problem objectives, 2) collection of all site-specific data, and 3) development of a site-specific conceptual model. Identification of appropriate equations consistent with the conceptual model's complexity then determines the selection of codes. This is in contrast to a generic systems model approach, for which the "model" is essentially already locked into a code. Final code selection is established by performing comparisons against analytical solution and experimental test cases. Failure in this evaluation may require modification of the system conceptualization or possibly a search for more adequate codes.

This study has found that aquifer (saturated zone) and unsaturated zone test cases call for different approaches as a result of having arrived at differing levels of conceptual physical complexity. In particular, aquifer modeling follows well established theory not requiring repeated validation. Unsaturated zone modeling, however, includes many incompletely understood phenomena that must be simulated in detail.

A central thesis of the proposed guidelines is that adequate evaluation test cases, based on both analytical solution and experimental data, must take into account current technical issues on groundwater transport modeling. Those issues express limitations on the current physical theories and modeling capabilities (i.e., numerical methods). Only by keeping those issues in mind can a user validly assess the predictive accuracy of a model and establish credibility in its application.

Many issues first raised in an August 1982 letter report were further discussed with regard to experimental results identified in the April 1983 report. For example, the problems of field-scale macrodispersion and solute migration during unsaturated infiltration were given special emphasis. Those two technical issues are not easily overcome and demand different modeling approaches, depending on the simulation objectives. New thinking on these issues calls for a stochastic approach in many cases, for instance. Such an approach reflects the important influences of field heterogeneity.

To provide a better reference case for estimating infiltration, a new program called INFIL1D was developed for one-dimensional, constant surface flux conditions. The issue of scale-dependent dispersion, for which dispersivity (spreading) increases with solute travel distance, was addressed by preparing the 3-D CFB Borden Landfill tracer experiment as a test case. That experimental data demonstrates the unexpected solute dispersal behavior that can occur even in an apparently homogeneous hydrologic system.

5.0 REMEDIAL ACTION

The objective of the Remedial Action Program at PNL is to support the Department of Energy's Uranium Mill Tailings Remedial Action (UMTRA) Project. The aim of UMTRA is to provide remedial actions at 25 inactive uranium mill sites, and associated contaminated vicinity sites, to stabilize and control the tailings to the extent that radiological and nonradiological hazards from them do not exceed EPA standards for the protection of the public health, safety, and the environment.

5.0 REMEDIAL ACTION

J. V. Robinson - Program Office Manager
Program Managers: J. N. Hartley and J. L. Buelt
Major Contributors: E. G. Baker, S. M. Barnes, P. A. Beedlow, H. D. Freeman, T. E. Gates, G. W. Gee, R. R. Kirkham, K. K. Nielson, V. C. Rogers, S. Tyler

5.1 RADON BARRIER SYSTEMS FOR URANIUM MILL TAILINGS (AH-10-15 - 80379)

The overall objective of this program is to develop radon barrier systems that will meet Environmental Protection Agency standards, minimize the use of remedial action, and remain stable for a very long period, including development of supporting technology and engineering specifications.

Summary

- Radon diffusion coefficients as a function of soil moisture saturation can be predicted for typical soil cover materials within a factor of two. For well graded soils and soils subjected to high compaction, additional work is needed to determine radon diffusion coefficients.
- Measurements on 20 offsite soils adjacent to UMTRAP sites indicate that soils containing less than 30% clay and tested at expected field moisture and compaction conditions will retain limited amounts of water (0.4 saturation ratio) on an average annual basis.
- Detailed unsaturated flow modeling predicts cover moisture contents for long-term periods based on climatic variables, water-retention characteristics of cover material, and estimated plant-cover characteristics. Model predictions generally agree with measured soil cover moistures at the Grand Junction test site.
- In some cases, simplified calculations using a climatic index can be used to make reasonable estimates of expected soil moisture values.
- Based on calculations of expected tailings pile settlement, it was concluded that the asphalt seal should remain intact for a very long time.
- Rock covers used to stabilize radon covers dramatically decrease the loss of water from soil covers. However, the use of rock covers in addition to vegetation may enhance establishment of the vegetation and not increase the water content of the soil.
- The Grand Junction field test site was visited in October. Measurements of radon flux, soil moisture and density, and radon concentration profiles in the columns were completed.
- All radon barrier systems except the uncompacted adobe clay earthen cover reduced the radon flux below the EPA standard of 20 $pCi/m^2 \cdot s$.
- Radon flux measurement systems using various configurations were compared and calibrated using the Bendix "thin" source. Excellent agreement was obtained between the measured and theoretical radon fluxes using the PNL flow-through system.

Earthen Covers

Radon Diffusion in Soils

The present status of radon diffusion information is summarized in Figure 5.1, where nearly 100 diffusion coefficient measurements are represented. The measurements were made using 20 western soils at various moistures and compactions. The curve represents a best-fit correlation with the data. For values of moisture saturation, m equal to 0.4 the correlation gives a radon diffusion coefficient, D of 0.018 cm^2/s, assuming a porosity of 0.4. For sites with these conditions, to meet a 20 pCi $m^{-2}s^{-1}$ radon flux standard the expected cover thicknesses range from 1.4 to 2.4 m (4.6 ft to 7.9 ft) for tailings whose bare-surface radon flux levels range from 100 to 300 pCi $m^{-2}s^{-1}$. If a value of 0.2 is used for m, then the correlation gives a D of 0.036 cm^2/s, for a porosity of 0.4. This value is about one-third of D for air. A cover thickness of 2.5 m (8.2 ft) results from using this D with a bare flux of 100 pCi/m^2s. However, in preparing the samples for the data in Figure 5.1, very little effort was expended in obtaining a good compaction or in using soils with favorable characteristics such as wide particle size distribution. Even so, it is observed that there are data points for D less than 0.01 cm^2/s with m as low as 0.2. Preliminary evidence suggests that increasing the compaction to at least 85% of Proctor results in an overall reduction in D of about 1.5 for well-graded soils.

Long-Term Equilibrium Moisture

The extreme sensitivity of the radon diffusion coefficient to moisture content dictates that the major factors controlling moisture content in soil must be considered in predicting radon attenuation capacities of earthen cover systems. The two major factors that must be accounted for are climatic moisture controls and soil property controls. The seasonal variations in moisture content due to climatic factors (precipitation,

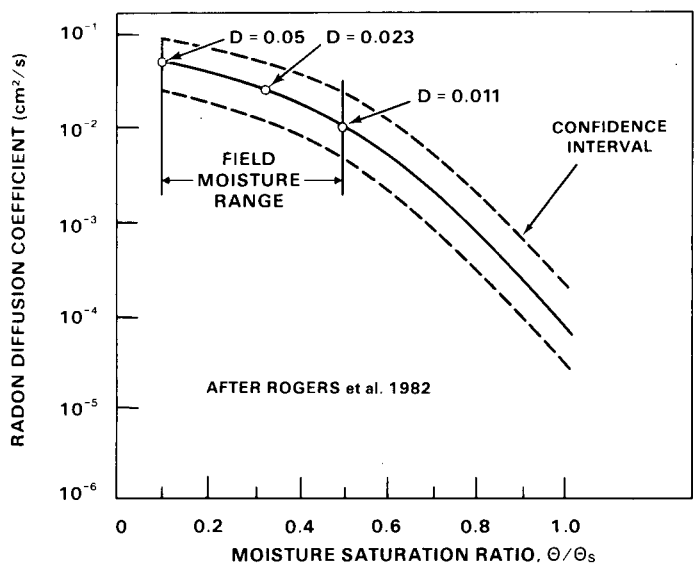


FIGURE 5.1. Effect of Moisture Content on Radon Attenuation

evaporation, plant-water extraction) generally dictate the expected long-term cover water contents.

All but two UMTRAP sites (Canonsburg, Pennsylvania, and Falls City, Texas) can be grouped into three general regions based on climate and geography: the Colorado Plateau, the West Slope of the Rocky Mountains, and the Northern Great Plains.

The Colorado Plateau sites (Grand Junction, Green River, Tuba City, Shiprock, Monument Valley, Mexican Hat, Slick Rock, Ambrosia Lake) have annual precipitation ranging from 15 cm at Monument Valley, Arizona, and Shiprock, New Mexico, to 28 cm at Slick Rock, Colorado. Frequency of rain events varies considerably at these sites, but summers are hot and precipitation is sufficiently low that plant cover is relatively sparse, with water being the major limiting factor for growth. It is expected that soils there become so dry that plants wilt throughout the root zone during much of the year and that estimates of average water contents could be made by determining the soil water contents at plant wilting (typically estimated by a laboratory measurement of the water retained in soil at 15 bars applied pressure). Field measurements made in September-October 1982 at Grand Junction, Shiprock and Monticello yielded water contents of 0.24, 0.14 and 0.18, respectively. Laboratory measurements on these samples yielded moisture saturations at 15 bars of 0.34, 0.14, 0.26.

The West Slope sites (Gunnison, Durango, Rifle, Naturita, Maybell, Salt Lake, Lakeview and Lowman) generally have moderate precipitation, ranging from 28 cm at Gunnison and Rifle to 57 cm at Lowman. No general conclusions can be made about the climate at these sites except that evaporation conditions are sufficiently severe at most that plant-water removal depletes the root zone to wilting conditions at least during the summer months. The water contents at these sites are expected to range between a field capacity and wilting point that can be estimated for most soil types to lie between the water contents held at 0.3 bar and 15 bar applied pressure.

Similar general statements can be made for the Northern Great Plains sites (Riverton, Spook, Bowman and Belfield). It is anticipated that most available cover materials will have sufficient clay content that the water contents held at 0.3 bar applied pressure would approximate the upper limit of stored water and that the lower limit would be equal to or less than the water held at 15 bars applied pressure. Field measurements in October 1982 at Riverton yielded an m of 0.45. The corresponding laboratory measurement at 15 bars yielded a value of 0.24 for m.

A single set of observations of moisture content in a cover for a given time provides only a very limited basis for determining the long-term moisture content, which will ultimately control radon diffusion. Moisture content in cover soils varies in time; hence, a comprehensive analysis requires extensive repeated sets of measurements of water content over a period of years.

Computer modeling provides a useful tool for predicting water movement and storage in soils. Input for a detailed simulation requires climatic variables (precipitation, temperature, humidity) as well as plant extraction factors and soil hydraulic properties.

Simulation of water withdrawal by plants in the top 1.0 m of a soil cover using the UNSAT water flow simulation demonstrated that

plants could significantly affect the moisture saturation level throughout the root zone. Simulations of over 5 years of climatic variations at Grand Junction indicate that soils can dry to saturation levels of less than 0.3 to depths exceeding 1 m. Field data from the 1981 earthen cover test site at Grand Junction substantiate the UNSAT model simulations and further indicate that dry soil placed uncompacted on the pile exhibits little moisture content change with depth (except at the surface). It should be noted that for the 1981 field test, the earthen cover plots were nearly bare of vegetation, and hence are not an ultimate test for the maximum soil drying expected at this site.

As part of the present Technology Development Program, PNL and Rogers Associates Engineering (RAE) are jointly developing semi-empirical engineering techniques for estimating long-term soil moistures at UMTRAP sites. Preliminary efforts have been reported previously. They were based upon over 400 soil samples obtained from undisturbed soils at uranium mining regions in the Western United States. The simple correlation gives long-term soil moistures ranging from $m = 0.2$ to 0.6.

Extension of this work has led to development of an Aridity Index, which is a slight modification of the Thornthwaite Moisture Index. The Aridity Index is the potential evaporation (PE) minus the precipitation (P) times the fractional PE (i.e., PE/100).

The Aridity Index is used in relation to soil texture to estimate the water content of the cover. Soil textural relationships (percent clay content) can aid in estimating expected long-term moisture contents. The relationships can be based on typical water retention-textural characteristics similar to those described in soil literature. Based on preliminary estimations, the relationship indicates that for sites whose Aridity Index is above 15 the moisture saturation will be below 0.4 for all but soils of very high clay content. Table 5.1 shows that six of the nine high-priority UMTRAP sites have Aridity Indices above 15; thus, their expected long-term values for m will be below 0.4. Although these relationships will probably be modified somewhat by compaction, specific clay types and plant cover, a general relationship of this nature appears to be a useful way to estimate expected radon diffusion coefficients for cover soils. Additional work will be needed to determine whether wide ranges in particle size distribution, i.e., large geometric standard deviations (GSDs), will modify the expected relationships.

Long-Term Earthen Cover Stability

Consideration was given to the expected settlement of a tailings pile resulting from the addition of a 3 ft-thick earthen cover. In the UMTRAP Technology Development Report to the TAC (Technical Assistance Contractor), July 1982, Colorado State University (CSU) had provided calculations for the predicted settlement of a theoretical tailings pile (Appendix A). The calculations had assumed a 40 ft-deep impoundment, and a unit weight, γ_c = 125 pcf, for the 10 ft-thick earthen cover. The tailings were composed of 60% sands and 40% slimes. Settlement resulting from adding the 10-ft cover was 9 in., and an additional 8 in. resulted from drainage of the pile. Adjusting the parameters for a 3-ft cover with a unit weight of γ_c = 100 pcf, C. F. Voss had calculated a total settlement of 10.5 in.

TABLE 5.1. UMTRAP High-Priority Site Climatic Indices

Site	(P) Annual Precipitation, cm	(PE) Potential Evaporation, cm	Aridity Index, (PE-P)(PE/100)
Colorado			
Durango	47	58	+6
Grand Junction	21	74	+39
Gunnison	28	55	+15
Rifle (1 & 2)	28	63	+22
New Mexico			
Shiprock	15	62	+29
Pennsylvania			
Canonsburg	93	51	0
Utah			
Salt Lake City	38	71	+23
Wyoming			
Riverton	22	57	+20

Through related work this past summer, it was shown that the settlement predicted by CSU was incorrect; hence, this error would also hold for the settlement calculated by C. F. Voss. Using the simplified unsaturated consolidated approach previously verified, CSU and PNL assumptions and results were compared as shown in Table 5.2

These significant variations between results derive from the following:

1. Using the formulation previously developed, it is shown that H should be the total pile depth. An underestimate of the expected settlement by 50% would result if H is inadvertently used as one-half the pile depth. In addition, if the time rate of settlement of the tailings pile composed of slimes/clays is of concern, the number of drainage faces is the most important condition influencing the settlement. If the pore water is freely flowing from both the upper and lower faces of the pile, the maximum drainage distance is one-half the thickness of the layer. However, utilizing underdrains and a cover on the pile, the maximum distance which pore water must flow is equal to the thickness of the layer. Hence, H should be the total pile depth.
2. Without differing material properties between layers, which would then require the summing of settlements, a layering approach should not be used. Mathematically, this is substantiated since the sum of the logs does not equal the log of the sum. Settlement due to the cover load varies significantly, depending on the approach taken.
3. Since the tailings are drained, a more realistic assumption is to assume that the average pore water pressure (μ_a-μ_w) is somewhat greater than the average water pressure in a pile whose base is at the water table. This is substantiated by the

TABLE 5.2. Comparison of Tailings Pile Settlement Calculations Made by CSU and PNL

	Settlement	Assumptions CSU	Assumptions PNL	Results, in. CSU	Results, in. PNL
Case 1: 60% Sands/40% Slimes					
	Cover load	2 Layers	1 Layer	9	5
Depth of pile - 40 ft		H = 20 ft	H = 40 ft		
Cover thickness - 10 ft	Drainage	$(\mu_a-\mu_w)_o$ = 1 psi	$(\mu_a-\mu_w)_o$ = 16 psi	8	1.8
			Total	17	6.8
Case 2: 100% Slimes					
	Cover load	2 Layers	1 Layer	21	6.9
Depth of pile - 40 ft		H = 20 ft	H = 40 ft		
Cover thickness - 10 ft	Drainage	$(\mu_a-\mu_w)_o$ = 1 psi	$(\mu_a-\mu_w)_o$ = 16 psi	32	6.5
			Total	53	13.4

test data used by CSU. For tailings whose pore water pressure is more than 8 to 10 psi, consolidation due to drainage is very slight (Sherry 1982). The pore water pressure of all drained tailings probably exceeds 10 psi except at the Vitro site at Salt Lake City, where a permanent shallow water table persists in the pile.

Using the consolidation equation previously verified, expected settlement for a 3-ft cover, unit weight γ_c = 125 pcf, will be 2.3 in. If a cover unit weight of γ_c = 100 pcf is used, the expected settlement will be 2.0 in., a significant difference from the 10.5 in. previously reported.

Asphalt Covers

Formulation Analysis

The purpose of this task is to develop and test site-specific asphalt/aggregate formulations for five high-priority sites--Grand Junction, Salt Lake City, Durango, Rifle and Riverton. An RFP was issued for the formulation work, and a subcontract was issued to Petroleum Sciences, Inc., of Spokane, Washington. Samples of typical aggregates were obtained from the five sites. Asphalt emulsions were obtained by Petroleum Sciences from suppliers near the sites.

Petroleum Sciences is currently testing various asphalt/aggregate formulations for each site to see if they meet specifications developed by PNL in FY 1982. PNL will later evaluate the effectiveness (radon diffusion coefficient) of each site-specific admix.

Long-Term Asphalt Stability

With the literature review completed, a final draft report on the physical stability of the asphalt admix seal is being written. Considering the work performed by the University of New Mexico and PNL, along with the settlement calculations (2.0-2.3 in.), the asphalt admix seal would be expected to perform adequately as designed.

Long-Term Stabilization

Revegetation and Rock Covers

The objective of this task is to establish guidelines to aid in designing surface stabilization systems that include rock and vegetation covers. Research has centered on 1) evaluating the interactions between vegetation and radon/biological barriers, 2) determining the effects of surface covers on soil moisture, and 3) examining the effects of rock covers on vegetation establishment and growth.

The first two quarters of FY 1983 have been spent analyzing data collected during the 1982 growing season. Preliminary results of a study conducted at the San Juan Mine in Northwest New Mexico will be described along with a summary of a lysimeter study conducted at Hanford.

The test plot constructed at the San Juan Mine was designed to provide information on the interaction between rock covers and vegetation and on the effects surface covers have on soil moisture.

Preliminary results suggest that rock covers favor the establishment of shrubs and forbs in contrast to soil-only treatments, which favor grass establishment. This suggests that when rock covers are applied to tailings, vegetation consisting of deep-rooted shrubs and forbs can be expected to become established. If shallow-rooted grasses are desired, good-quality soil free of stones should be emplaced. No differences in soil moisture were observed between vegetation and vegetation plus rock treatments.

Lysimeter Study

Simulations of moisture movement through tailings and cover layers (Mayer et al. 1981) suggest that different surface covers lead to different degrees of moisture retention in the covered tailings pile. Evapotranspiration from a vegetated cover in semiarid areas can result in a relatively stable moisture content. A rock cover may increase the moisture content of the tailings pile by significantly reducing evaporation. The purpose of this study was to examine the effects of surface covers on soil water loss. Water loss from lysimeters covered with various types and amounts of rock, vegetation, and rock plus vegetation was compared to loss from bare soil.

Rock size and application rate had little effect on soil water loss. However, vegetation significantly increased water loss relative to bare soil and rock-only cover treatments (Figure 5.2). Vegetation-plus-rock treatments resulted in water loss similar to vegetation-only treatments. Rock covers without vegetation dramatically reduced water loss.

The results of this study support the findings of simulation studies on the effects of rock and vegetative covers on uranium mill tailings (Mayer et al. 1981). Rock covers reduce soil water loss dramatically and could lead to water accumulation and drainage from a waste burial/uranium tailings site.

Rock covers that are revegetated appear to facilitate similar rates of water loss as vegetation covers. Radon barriers (covers) covered with a rock-plus-vegetation cover are

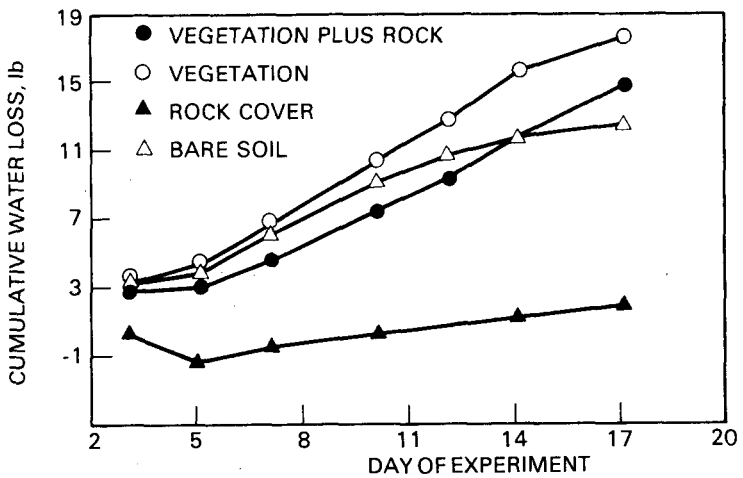


FIGURE 5.2. Effects of Vegetation and Rock Covers on Moisture Loss from Soil

not likely to lead to moisture accumulation higher than that on the surrounding soil unless a water catchment structure is created in the cover.

The implication to management of uranium tailings sites is that in areas where vegetation is too sparse (extremely arid regions) to stabilize the cover material, combinations of rock plus vegetation will provide additional erosion protection while preventing moisture accumulation. In some areas rock covers may enhance vegetation establishment and growth. The rough surface of the rock cover may benefit seed germination by providing favorable microsites for seeds. Rock covers will reduce evaporation and consequently more water will be available for plant growth.

Field Test Monitoring

Site Visits and Monitoring

The Grand Junction field site was visited during October 18-19, 1982. Activities included measurement of radon fluxes, collection of surface and subsurface soil samples for moisture and bulk-density determinations, neutron and density probe calibration and measurement of radon concentrations in the test columns. Field moisture and density measurements were made using neutron and gamma downwell probes. Site construction consisted of installing two phone lines into the access column, one of which is currently hooked to the CR5 datalogger. Soil temperature and moisture data are accessed from Richland via the phone lines.

Radon flux measurements were made on the 15-cm gel mix plot, half of the asphalt test points, and all of the earthen cover test points. Results are summarized in Table 5.3.

All of the earthen covers show an increase in radon flux over measurements taken in July 1982. This increase is thought to be due to advective transport of radon by water in the top layer of soil, which is more moist in October than in July. The asphalt emulsion barrier shows a net decrease in flux during October, which is attributed to the fact that only half of the area was monitored. The asphalt area next to the multilayer barrier, which in the past has had the highest flux values, was not monitored during this trip.

TABLE 5.3. Average Radon Flux from FY 1981 Field Test Site - October 1982

Radon Barrier	Average October 1982 Radon Fluxes, pCi $m^{-2}s^{-1}$	Change From July 1982, %
Mancos shale	1.7 ± 1.2	+240
Bentonite clay	11.3 ± 7.2	+71
Compacted adobe	9.8 ± 5.4	+172
Adobe	28.4 ± 18.3	+255
Asphalt emulsion	0.9 ± 1.5	-40
Multilayer 15-m gel mix	6.0 ± 5.6	0

Therefore the average flux values were lower during October. The column tests were also monitored during the October trip. Radon flux measurements were made over 12-hr periods, and the results are shown in Table 5.4.

Neutron probe calibrations were completed, and data from column tests and field plots were analyzed. Moisture profile data were prepared for the field test topical report. The data indicate that moisture content changes with time at the field site have been minor except in the top 1 m of cover. Only a slow rate of water movement is apparent in the wetted, compacted soil placed at depth (below 1.8 m) in three of the four earthen cover treatments. Since there is little vegetation cover on the plots, it is expected that several years will elapse before significant drying occurs in the lower layers, primarily because of the very low hydraulic conductivity of the overlying dry adobe clay and the relatively low conductivity of the compacted layers.

TABLE 5.4. Average Radon Flux from Column Tests - October 1982

Column		October 1982 Avg. Radon Flux, pCi $m^{-2}s^{-1}$
EC1	Mancos shale	19.1 ± 7.7
EC2	bentonite clay	15.6 ± 8.4
EC3	compacted adobe	38.8 ± 10.8
EC4	adobe	40.3 ± 8.6
M1	multilayer	4.6 ± 1.4
M2	multilayer	6.4 ± 6.5
A	asphalt emulsion	1.6 ± 0.1

Calibration of Radon Flux Measurement Systems

During the site visit to Grand Junction in October 1982, an opportunity arose to calibrate the radon flux measurement systems. Bendix Field Engineering Corporation had constructed a thin layer of uniformly mixed tailings in a 6-ft-dia tub. Since the source was very thin (~6 cm), it could be assumed that all the radon escaping the tailings particles would diffuse to the surface of the tailings. Therefore, using the radium content emanating power, depth, and bulk density of the tailings, a theoretical radon flux could be calculated. This theoretical flux could be compared with the measured flux to obtain a reasonably accurate calibration of the measurement system. The tailings parameters measured by Bendix were 837 pCi/g of ^{226}Ra, emanating power of 0.19, and bulk density of 1.51 g/cm^3. Using the thin source model, the radon flux from the tub averaged 26.8 pCi $m^{-2}s^{-1}$.

A number of calibration measurements were made using a variety of configurations of activated carbon canisters in addition to the PNL flow-through system. The calibration measurements were made by placing the measurement system on the tailings in the tub, exposing the canister or tent for a specified period, and then counting the activated carbon with a gamma spectroscopy system to determine the measured flux. The depth of the

tailings was measured at each point since a 1- or 2-cm change in depth of the tailings would result in a 20 to 40% change in radon flux.

The activated carbon canisters are made of sections of 10-cm (4 in.) and 38-cm (15 in.) PVC pipe. The covered canisters are the standard systems with a PVC end cap that has a 0.6-cm hole drilled in the top.

The results of the calibration measurements are shown in Figures 5.3 and 5.4. From these figures it can be seen that the radon flux as measured by activated carbon canister depends on the amount of carbon used, the length of the measurement, and the area of the exposed top of the canister. The open-ended activated carbon canisters showed a very interesting trend. For exposure times of 8 hr or less, the measured radon flux exceeded the calculated flux. As the exposure times increased, the radon flux generally decreased, depending on the quantity of carbon used in the canisters. Figure 5.3 shows the ratio of the measured flux divided by the calculated theoretical flux versus time for a series of charcoal densities in 8.9-cm-dia canisters. The general trends indicated by this figure show that higher carbon densities result in higher fluxes. Furthermore, longer exposure times result in lower fluxes. These relationships can be deduced. However, since activated carbon actually acts as a delay bed rather than a permanent collector, there are no data to explain why a 7-hr exposure would result in higher than theoretical fluxes. An in-depth study considering meteorological changes is needed to better understand this phenomenon.

The large (0.107 m^2) activated carbon canisters show the same general trends as the small ones (Figure 5.4). Longer exposure

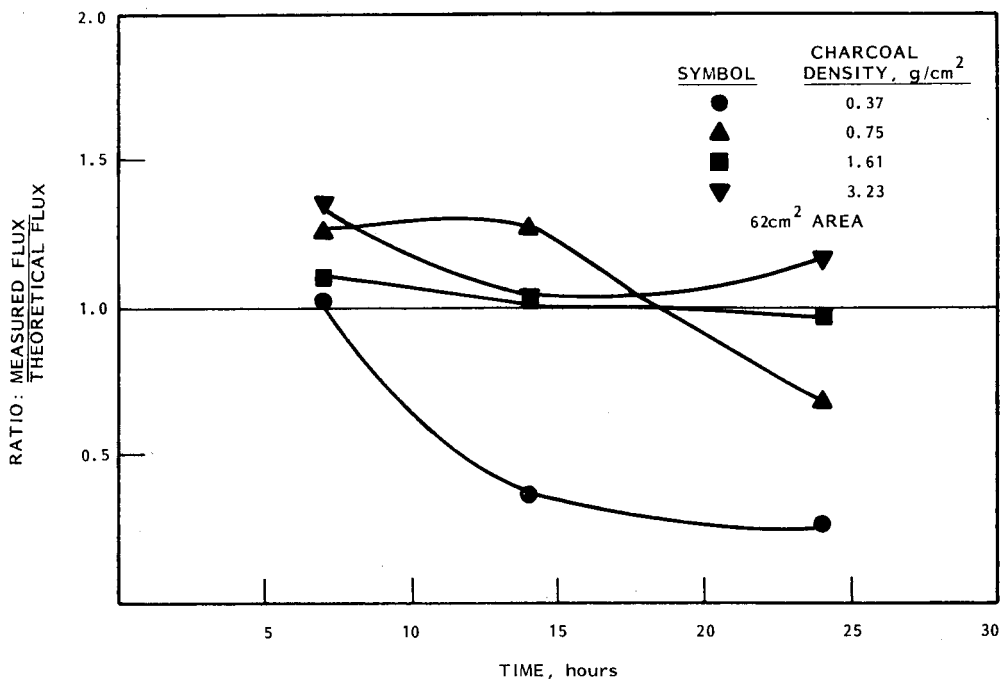


FIGURE 5.3. Effect of Time on Efficiency of 3 1/2-in.-dia Charcoal Canister

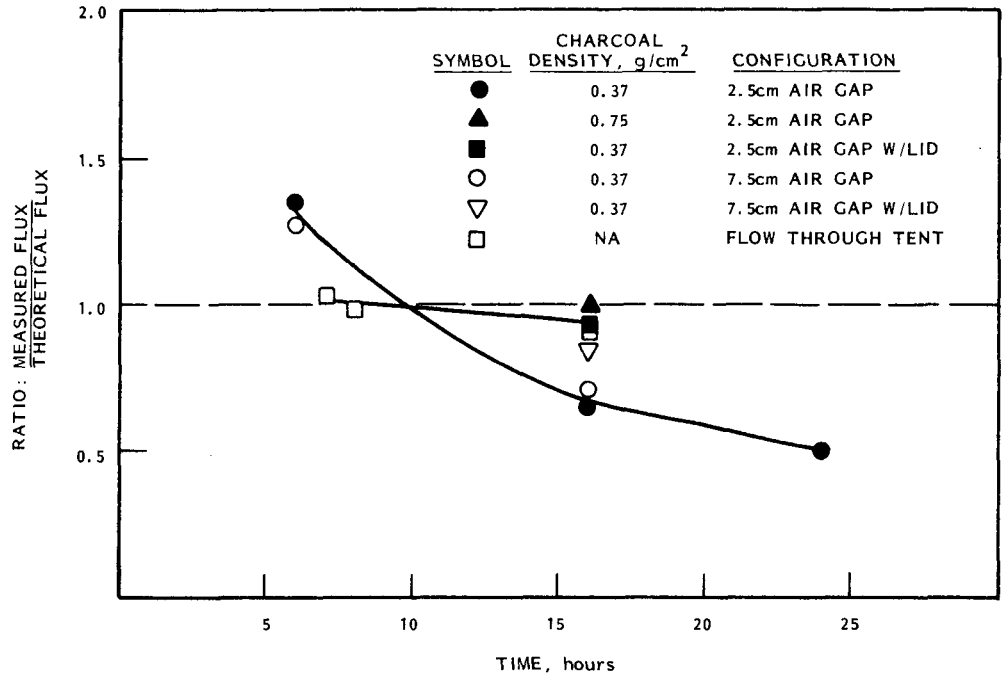


FIGURE 5.4. Effect of Time on Efficiency of 15-in. Charcoal Canisters and PNL Flow-Through System

times result in lower measured fluxes. Short exposure times also resulted in greater than theoretical fluxes.

In addition to charcoal density, the effect of the air gap between the charcoal and tailings and the area of the top exposed to atmosphere was investigated. The air gap seemed to make no significant difference in the measured flux. The slight differences seen between measurements made with a 2.5-cm air gap and a 7.5-cm air gap can be attributed to measurement error (i.e., counting statistics, tailings depth, ^{226}Ra content, and emanating power measurements). Therefore, it is concluded that for radon flux measurements of 16 hr or less, the air gap of the canister is not a significant factor in the resultant flux value.

The PNL flow-through system was also calibrated on the Bendix tailings source; measurements of 7-, 8-, and 16-hr duration were made. Results of these measurements are also shown in Figure 5.4. Excellent agreement between the measured and theoretical fluxes was obtained for the 7- and 8-hr measurements. The measured flux appeared to drop to 93% of theoretical after 16 hr, but more measurements are needed to confirm this drop. From these measurements it appears that the PNL flow-through system is a reasonable method for measuring radon flux. Additional measurements under a variety of conditions are still needed to prove its accuracy in field measurements.

Although these calibration tests indicate that various radon flux measurement techniques can be used to obtain a reasonable estimate of radon flux, caution must be exercised in the length of exposure time of the charcoal in the canister. An in-depth study of the effects of meteorological conditions

as well as time on the response of measurement systems to radon fluxes should be carried out to obtain optimized conditions for radon flux measurements.

Engineering Specifications

Material and Application Specifications

The final goal of this task is a report summarizing the technology for asphalt radon barrier systems. The report will review laboratory and field tests results and will include materials specifications, application procedures, and cost estimates. A draft of the report is essentially complete except for ongoing work. Materials specifications and application procedures are complete except for site-specific asphalt/aggregate formulations and the optimum delay and moisture content for compacting the admix. These are part of Petroleum Sciences' subcontract to be completed by July.

A paper entitled "Cost and Effectiveness of Radon Barrier Systems" was presented at the Fifth Symposium on Uranium Mill Tailings Management in Fort Collins, Colorado, on December 9-10, 1982.

References

Mayer, D. W., P. A. Beedlow and L. L. Cadwell. 1981. Moisture Content Analysis of Covered Uranium Mill Tailings. UMT-0207, PNL-4132, Pacific Northwest Laboratory, Richland, Washington.

Sherry, G. P. 1982. Constitutive Relationships for Unsaturated Uranium Mill Tailings. Colorado State University Masters Thesis.

5.2 LINER EVALUATION FOR URANIUM MILL TAILINGS (AH-05-15 - 80395)

The objective of this program is to evaluate the effectiveness of prospective lining materials for preventing moisture transport of radionuclides and hazardous chemicals from uranium mill tailings over long-term periods.

Summary

The primary emphasis of the activities for the reporting period was on completion of the exposure tests for the final series of asphalt liner samples and analysis of the liner samples from the two series of exposure tests. Results show that the imposed exposure conditions were successful in noticeably accelerating the aging reactions of the 0.7-cm-thick liner material. In addition, preparations have been completed for establishing a subcontract to excavate the liners installed in the field. Results of the clay and soil interaction studies performed in FY 1982 were presented by Relyea (1982) at the Uranium Mill Tailings Management Symposium in Fort Collins, Colorado. Also presented at the symposium were the test and analytical procedures of the asphalt aging studies by Barnes (1982). The decision-tree report (Relyea 1983), which describes a systematic approach for determining the need for and the type of liner at an inactive tailings site, was published in March.

Asphalt Aging Studies

The exposure period for the final series of catalytically airblown asphalt (CAA) liners was completed this last quarter. The

permeability performance of these liners was very close to that observed for the previous series of tests. The average permeability from the most recent series of tests completed in October 1982 was ~2 x 10^{-10} cm/sec as compared with 6 x 10^{-10} cm/sec from the previous set. This similarity in permeability coefficients observed during the accelerated aging tests indicates that a catalytically airblown asphalt membrane will exhibit very stable long-term permeability properties. However, because these liners under various exposure conditions have exhibited nearly identical permeability behavior, another method was used to determine the extent of aging.

To quantify the expected performance lifetime of the asphalt liner, the infrared (IR) absorption data from the two series of aged catalytically airblown asphalt samples have been collected and compiled. A preliminary examination of the data showed increasing carbonyl concentrations with depth into the liner to varying degrees. Since carbonyl concentrations are expected to indicate the extent of aging, the increasing concentration with depth could be explained by dissolution of the carbonyls into the acidic leachate after they form. Figure 5.5 is a logarithmic plot of the relative carbonyl concentrations with penetration into the liner for the same oxygen concentration and temperature conditions but different acidity levels. Near the surface the liner exposed to the lower pH solution appears to have had more of the carbonyl-containing (or oxidized) components of the asphalt leached out.

This conclusion is supported by the presence of an organic film on the surface of the simulated leachate and patches of darkly colored simulated tailings in the exposure columns. Moreover, the oxidized asphalt molecules would be expected to exhibit polar properties and therefore would become increasingly soluble in increasingly polar solvents. Because the pH of the simulated

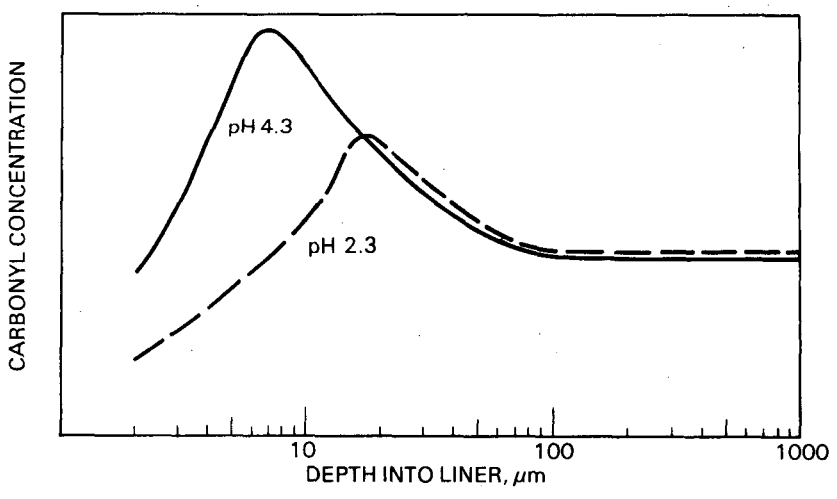


FIGURE 5.5. Relative Carbonyl Concentration as a Function of Depth into the Asphalt Liner

leachate has been controlled by using sulfuric acid, a very polar solvent, increasingly acidic (lower pH) solution would be expected to extract more of the oxidized asphalt from the liner, as has been observed. It is also important to note that the oxidized asphalt leaching observed occurs only in the first ~20 μm of the liner. Therefore this phenomenon is not expected to adversely affect the long-term liner performance.

Beyond the 20-μm depth, increased temperature and concentrations of oxygen were successful in accelerating the rate of carbonyl formation. Figure 5.6 is a plot of four liner samples exposed to ambient and accelerated (low/high) temperatures and oxygen concentrations. The notable difference in concentration of reaction products will be used to determine the rate of

$$r = k(T) \cdot C_{02}^{n}$$

where: r = reaction rate
$k(T)$ = temperature-dependent reaction rate constant
C_{02} = concentration of oxygen, and
n = order of reaction.

By examining the rate of diffusion into the liner and defining the unknowns in this equation from the empirical data collected in these studies, an estimated lifetime of the asphalt liner up to 1000 years can be predicted. The results of these studies will be issued in a technical report next quarter.

Field Studies

Preparations for next quarter's field activities have been completed. A subcontract has tentatively been established for the excavation activities. Planned sampling and analytical activities are as follows:

- repair failed moisture extraction cups for future sampling (if desired)
- obtain three asphalt liner samples for aging analysis
- obtain five clay liner cores for permeability determinations and for chemical and gamma-scan analysis
- cover tailings-filled pit, after sampling, with clean overburden.

Analytical results will be published in the final report to be issued at the end of FY 1983.

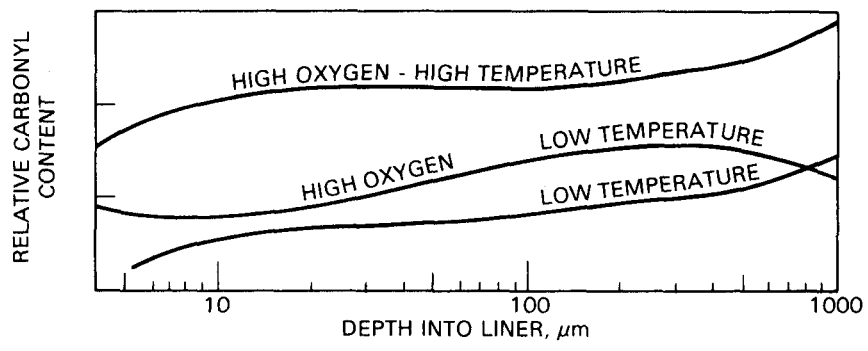


FIGURE 5.6. Comparison of Oxygen and Temperature Effect on Aging

References

Barnes, S. M. and J. L. Buelt. 1982. "Accelerated Aging of Catalytically Air-blown Asphalt Membranes Under Simulated Uranium Mill Tailings Impoundment Conditions." Uranium Mill Tailings Management, p. 437, Colorado State University, Fort Collins, Colorado.

Relyea, J. F. 1983. A Decision Tree Approach to Evaluating Inactive Uranium Processing Sites for Liner Requirements. Pacific Northwest Laboratory, Richland, Washington.

Relyea, J. F. and W. J. Martin. 1982. "Evaluation of Inactive Uranium Mill Tailings Sites for Liner Requirements: Characterization and Interaction of Tailings, Soil, and Liner Materials." Uranium Mill Tailings Management, p. 507, Colorado State University, Fort Collins, Colorado.

6.0 SUPPORTING STUDIES

J. V. Robinson, M. R. Kreiter, and L. T. Lakey - Program Office Managers

Program Managers: S. R. Bierman, M. J. Budden, L. L. Burger, J. R. Friley, E. D. McClanahan, G. W. McNair, I. C. Nelson, J. V. Robinson

Major Contributors: L. L. Clark, R. M. Fleischman, C. A. Geffen, E. N. Greenwell, J. M. Lytle, R. W. McKee, R. W. Moss, J. G. Stephan

6.1 HANFORD DEFENSE WASTE STUDIES DEVELOPMENT OF ENGINEERING AND ENVIRONMENTAL INFORMATION (AR-05-10-02X - 80381)

The objective is to assist DOE-RL in the development and assessment of methodologies for the disposal of existing and future Hanford radioactive defense wastes.

Progress During Reporting Period

Activities during the reporting period were as follows:

- preparation of engineering and environmental analyses required in the preparation of the draft EIS for disposal of certain Hanford defense wastes
- preparation of support documentation.

Preparation of Engineering and Environmental Analyses (E&EA)

The Draft Working Paper Engineering and Environmental Analysis Report for Disposal of Hanford High-Level and Transuranic Wastes was submitted to DOE-RL, as scheduled on December 6, 1982. DOE-RL subsequently reviewed the draft and provided written comments on January 17, 1983. A copy of the draft report was also forwarded by DOE-RL to DOE-HQ to provide information on the thrust and scope of the environmental analyses.

HLW and TRU radioactive wastes analyzed include buried solid wastes (TRU), liquid wastes, tank wastes, and cesium and strontium capsules. Currently stored HLW and TRU and future HLW wastes to be generated by PUREX fuel reprocessing plant operations were considered. Chemical hazards associated with radioactive wastes were qualitatively assessed.

Socioeconomic data, resource commitments and costs associated with disposal of the defense wastes were identified for the period during which disposal operations would occur. Environmental assessments were also made for the post-disposal period.

Environmental analyses include air quality, water quality and use, land use, radiological dose to workers and the general public, nonradiological safety impacts, cost impacts, and socioeconomic impacts that may be associated with disposal options to be considered in the Hanford Defense Waste DEIS.

Support Documentation

Several working sessions were held between PNL and Rockwell Hanford Operations Staff following the review of the draft E&EA report. PNL requested that Rockwell finalize its engineering support documentation that is required to perform environmental analyses. Subsequent working sessions held between DOE-RL, PNL, and Rockwell resulted in changes to the scope of the environmental analyses. Completion of Rockwell's supporting documentation was subsequently postponed until changes in scope are further defined.

Future Activities

Environmental analyses will be conducted based on support documentation to be prepared by Rockwell. Rockwell will finalize support documentation after appropriate PNL review and comment. The resulting data will be used in the assessment of environmental impacts for the Hanford Defense Waste EIS.

6.2 HANFORD DEFENSE WASTE STUDIES - PREPARATION OF AN ENVIRONMENTAL IMPACT STATEMENT FOR DISPOSAL OF HANFORD DEFENSE WASTES (AR-05-10-02X - 80754)

The objective is to assist DOE-RL in the preparation of a Draft and Final Environmental Impact Statement for the disposal of high-level and TRU defense waste stored at Hanford.

Activities During the Reporting Period

Activities were as follows:

- preparation of the Notice of Intent (NOI)
- review of draft regulations
- participation in working sessions with DOE-RL to define the scope of the draft EIS
- interim schedule preparation.

Preparation of the Notice of Intent

Assistance was provided to DOE-RL in finalizing the draft Notice of Intent to announce preparation of the draft EIS in the Federal Register. Publication of the NOI is scheduled for April 1, 1983. Alternatives described in the draft NOI are 1) geologic disposal (the reference disposal for immobilized high-level waste); 2) onsite stabilization and isolation with engineered barriers; and 3) continued storage (no action). Completion of the draft EIS is tentatively scheduled for the end of 1983.

Review of Draft Waste Management Plans and Draft Regulations

A concentrated effort during this reporting period focused on the review of 40 CFR 191 as well as the Environmental Protection Agency's Draft Regulatory Impact Analysis (EPA 520/1-82-024) and Draft Environmental Impact Statement for 40 CFR 191 (EPA 520/1-82-025). Other draft documents reviewed were: The Defense Waste Management Plan (February 1983) and Draft DOE Order 5820 - Radioactive Waste Management. Written inputs were prepared for DOE-RL's review. The purpose of the review was to determine potential impacts of the draft regulations to the disposal of Hanford defense waste sites and preparation of the draft EIS.

DOE-RL/PNL/RHO/Working Sessions

PNL participated, with Rockwell Hanford Operations staff, in intensive work sessions held by DOE-RL to further define the scope of the Draft Hanford Defense Waste EIS.

Interim Schedule

An interim schedule was drafted for preparation of the draft EIS. This schedule assumes that the NOI will be published in April 1983.

Future Activities

Draft chapters for the DEIS will be prepared and submitted to DOE-RL for review planned to begin in May 1983.

6.3. WASTE MANAGEMENT SYSTEM STUDIES (AP-05-20-10 - 80219)

Decision-making in the selection, development, and implementation of waste management systems and technologies requires supporting analyses and independent assessments of alternatives to provide guidance on the most appropriate course of action. To ensure a balanced evaluation, the analyses and assessments must be performed on a systems basis. This program performs such studies.

Repository Cost Model Development (RECON)

The computer model RECON[a] is being used to perform cost trade-off studies which, in turn, may be used to: 1) review repository site programs, 2) provide guidance to site design activities, and 3) investigate system cost reduction strategies, such as those being conducted under another task of the Waste Management System Studies Program (cf., Technical and Economic Evaluations).

The objective of this task in FY 1983 is to broaden and enhance RECON model capabilities to address three current key repository design issues: alternative emplacement methods, co-emplacement of wastes, and receiving and packaging facilities.

The following briefly describes the approach to each of these issues:

- Analysis of Alternative Emplacement Methods
  This task will develop additional logic to describe and develop costs for a repository design in which wastes are emplaced in horizontal boreholes drilled in the rock pillars separating the rooms as in the current BWIP (Basalt Waste Isolation Project) concept.
- Developing Co-emplacement Analysis Capability
  At the beginning of FY 1983, RECON model logic assumed that emplacement rooms are unique to each waste type. This task will develop an additional logic to allow analysis of trade-offs associated with putting multiple wastes in the same room (e.g., low-level TRU in rooms where spent fuel or HLW is emplaced in boreholes in the floor).
- Upgrading Facility Cost-Estimating Capability
  This task will develop the ability to better match receiving and packaging facility capabilities and costs with repository designs and operating scenarios. This will be done by estimating facility construction costs based on general facility design parameters and descriptions. The possibility of adapting the CONCEPT capital cost estimating model to accomplish this will be investigated.

Progress during the reporting period on the tasks is summarized as follows:

- Information on the latest conceptual BWIP design and waste package concepts for basalt was obtained from the BWIP staff. The BWIP conceptual design descriptions were incorporated into the RECON model. Modifications to the program included adding the capability of putting more than one HLW or spent fuel canister in a hole. The room-and-pillar logic already in the model made it possible to simulate the BWIP design without substantial changes. Following these modifications, data were collected from several different reports on basalt repositories in order to model cost estimates for the BWIP design. (Cost estimates were not directly available from the BWIP conceptual design description.)

(a) For background information on the RECON model, the reader is referred to Section 3.4.2 of this report and also to the section entitled "Disposal Charge Analysis" in prior issues of this document.

Cost estimates were prepared for two cases: a case using the design in which holes were drilled in the walls of the room, and a case in which holes were drilled in the floor of the room. As expected, the hole-in-the-wall design was cheaper than the vertical borehole design by about $120 million dollars. The difference in cost is primarily related to mining and rock-handling costs. A letter report describing the results of these two cases was sent to DOE-RL on January 5, 1983.

- Code logical flow diagrams were completed which allow RECON to co-emplace non-heat-generating waste in rooms in the repository excavated for primary, heat-generating waste. The available options encompass the following emplacements:

	Primary Emplacement	Secondary Emplacement	Tertiary Emplacement
1.	Vertical borehole for HLW and spent fuel	TRU in interspersed vertical borehole locations	Option to emplace contact waste in room after primary and secondary emplacement
2.	Vertical bore- for HLW and spent fuel	Contact-handled waste in room after primary emplacement	
3.	Horizontal borehole for HLW and spent fuel	TRU in interspersed horizontal borehole locations	Option to emplace contact waste in room after primary and secondary emplacement
4.	Horizontal borehole for HLW and spent fuel	Contact-handled waste in room after primary emplacement	
5.	Horizontal borehole for HLW and spent fuel	TRU in trench	Option to emplace contact waste in room after primary and secondary emplacement
6.	Room emplacement of HLW and spent fuel	Optional placement of TRU waste or contact waste in same room	

The coding and implementation of the logic flow diagrams resulted in the development of five subroutines which were added to the RECON code to model the co-emplacement of TRU wastes with heat-generating wastes. The subroutines are:

EMPLAC - Controls overall co-emplacement logistics

PPUT - Performs primary heat-generating waste emplacement

SPUT - Places secondary non-heat-generating waste in mined rooms after the primary heat-generating waste has been emplaced

TPUT - Places the third waste type in previously mined rooms that have both primary and secondary emplacements

SSTOR - Mines rooms dedicated to non-heat-generating waste types whenever there is no room for co-emplacement in previously mined primary emplacement rooms

The data input was expanded to include necessary additional information such as the number of secondary waste packages per primary waste package for each waste-type combination.

These new additions to the RECON code were being debugged at the end of March.

Technical and Economic Evaluations

Current studies have shown the importance of considering the entire waste management system in evaluating waste management alternatives. To effectively manage the development of repository concepts, it is essential that DOE have information relating to the impact of repository design, siting and schedule options on total system costs, and the impact of waste form and package design decisions on repository costs. It is, therefore, apparent that a systematic approach is desirable for analyzing alternatives and their impact on the geologic repository requirements. This task is designed to provide information that incorporates all major aspects of the waste management system for program direction and decision-making.

The objective of this task is to provide DOE with information that will assist in the selection of repository sites and waste form criteria and specifications. Information will be developed on a consistent basis to identify the cost impacts of major system design alternatives with the intent of identifying cost-effective options. Information relating to potential safety and radiologic-release trade-offs for system alternatives will also be developed. This information will be used: 1) to assist the sites in developing repository concepts that tend to minimize overall waste management costs and 2) to develop overall waste management systems approaches that are cost-effective in conjunction with commercial waste treatment and storage programs. The RECON model (cf. previous section) will be used to develop the parametric repository cost information necessary for these evaluations.

The major effort on this task during the reporting period was a survey and analysis of system alternatives to be evaluated in detail in FY 1983. The principal concern is with alternatives related to the treatment and disposal of the various TRU wastes that would be produced during spent fuel processing. Accordingly, discussions were held with staff from Barnwell Nuclear Fuels Processing regarding possible treatment alternatives that should be evaluated for the Barnwell plant wastes.

One of our objectives is also to encompass a range of alternatives that would include requirements for NRC licensing as reflected in the most recent draft of 10 CFR 60.

A letter report describing the system alternatives selected for in-depth evaluations was completed and delivered to DOE-RL on March 31. The alternatives include six waste treatment alternative combinations that encompass the range of available treatment options and 22 combined waste management system alternatives. These alternatives are designed to provide answers and perspectives to 13 cost-related questions of interest to DOE that are related to determination of waste acceptance specifications and the cost impact of potential regulatory requirements.

Special Issues Analyses

The Division of Waste Repository Deployment (DWRD) operates with substantial interfaces with other DOE offices, NRC, industry, Congress, and the public. It is important to evaluate the nature of the issues at these interfaces in order to communicate the Division's programs. There is also a need to anticipate and respond to current events. Qualified PNL staff having broad nuclear perspectives and specialized capabilities to conduct supporting analyses and independent

assessments of these interface issues are being utilized in this task on a case-by-case basis.

The objective of this task is to provide analyses of interface issues affecting repository deployment. To date in FY 1983, two issues have been addressed at DOE's request as follows:

- Draft Guidance to the Sites Regarding Waste Acceptance Specifications
  We were asked to prepare draft guidance that DOE-HQ would provide to RL-BWIP, NV-NNWSI, and NPO-ONWI[a] requesting that those sites prepare waste package specifications for commercial HLW, spent fuel, commercial TRU wastes, and Defense Waste Processing Facility wastes. A draft of that guidance was delivered to DOE-RL and DOE-HQ in December.
- Overall Plan for Conducting System Cost Studies for the National Waste Terminal Storage (NWTS) Program
  PNL was requested to prepare an overall plan for conducting waste management system cost studies for the NWTS program, including cost studies related to transportation, storage, waste treatment, and disposal.

Recognizing that it is essential to contact other NWTS contractors to obtain their ideas on required cost studies and information on studies that they are planning, letters were sent to ten contractors with copies to the cognizant field office representatives outlining the objectives and scope of the study and asking for assistance from each of these sites. These contractors were then contacted by telephone, or by visits in the case of Hanford contractors. Some useful information was collected, but, with few exceptions, we found that contractors do not concern themselves with aspects of the waste management system outside their direct purview. As a result, the plan was primarily a PNL effort.

A draft NWTS Program Waste Management System Cost Studies Plan was completed and delivered to DOE-RL on March 29. The plan was divided into two main parts. In the first part, a methodology for developing an integrated waste management system cost model is presented. In the second part, recommended cost studies and related activities, their timing, and their interrelationships are described.

The plan primarily focuses on studies that involve system interactions and relationships among major components of the system--specifically, treatment, transportation, storage, and disposal. Outside of this plan, each repository site program is supporting cost studies related to facility design options but is concerned only to a limited extent with the waste management system interactions external to the repository site. The capabilities and limitations of other system components can have significant system cost impacts that might be mitigated to some extent if the system as a whole is considered.

The primary objective of this cost plan was taken by PNL to be the waste management system cost comparisons required to support the selection of the first repository site. The Nuclear Waste Policy Act of 1982 calls for an April 1, 1987, recommendation by the President on this first repository site.

(a) Richland Operations Office - Basalt Waste Isolation Project, Nevada Operations Office - Nevada Nuclear Waste Storage Investigations, and National Waste Program Office - Office of Nuclear Waste Isolation.

Nuclear Fact Book

In FY 1982, under this program, PNL published the Nuclear Fact Book, PNL-4239, to fill a need for a small, pocket-sized, loose-leaf document containing current, accurate information on topics related to nuclear waste management and directly related topics, e.g., nuclear power. The document has been well received. A need exists to update some of the information contained in the book to reflect significant changes in DOE's waste management program plans or in areas bearing on those plans, e.g., proposed or enacted waste management legislation.

The objective of this task is to update PNL-4239 in a timely fashion to reflect developments significant to the DOE's nuclear waste management program plans. Particular attention was given to those aspects having potential impact on DOE's waste repository deployment plans.

In February we submitted recommended revisions to the book to DOE-RL. We also sent them to the Office of Nuclear Waste Terminal Storage Integration. Comments were received and resultant changes to the new edition made.

In a meeting held in March with RL, it was decided that:

- Future issues of the book will be bound instead of the current loose-leaf version.
- RL/PNL will make distribution to the DOE recipients.
- Arrangements will be made such that copies can be purchased by others through some government printing and distribution channel (e.g., the Government Printing Office).

Public Information Analysis

To facilitate the repository site selection process, it is important that DOE develop an effective means of communicating credibly to the public--especially those near potential sites. Information must be provided that corrects public misconceptions and enables a greater degree of informed participation in program implementation. To meet this responsibility, it is important to clearly define public information needs and to identify communication techniques applicable to the transfer of appropriate technical information to the public. To accomplish this task, DOE will need to identify desired target groups, appropriate message content and the most effective media for credible communication to those target groups.

The objective of this task is to structure and implement a public information program aimed at:

1. increasing public understanding of waste management technology and programs
2. correcting public misconceptions about certain aspects of nuclear waste storage and disposal
3. assisting DOE in successfully interfacing with the communities in potential disposal site areas.

Two primary tasks are scheduled to be completed during FY 1983 to accomplish the desired objectives:

- Task 1 - Development of a document summarizing public information needs in the area of waste management, important target groups for a public information program, and effective communication media and techniques. This information was obtained

from a review of research already conducted by the Human Affairs Research Centers and, as appropriate, by other organizations in the "nontechnical" issues area of repository siting. A letter report for Task 1 was sent to DOE-RL at the end of February.

- Task 2 - This portion of the study will utilize the results of Task 1 to develop for DOE guidance regarding the development and planning that may be required to construct and implement an effective public information program. The letter report on Task 2 neared completion at the end of the reporting period.

6.4 NUCLEAR MATERIALS TRANSPORTATION TECHNOLOGY STUDIES

6.4.1 Thermal Analysis Methods (AR-05-25 - 80746)

The objective of this research is to provide improved analytical methods for thermal analysis of shipping casks.

Summary

An information exchange meeting was hosted in which both structural and thermal codes relevant to spent fuel shipping cask design were discussed. The meeting was attended by government and industrial representatives interested in cask design and licensing.

Thermal Analysis of Casks

In previous reporting periods, several computer codes potentially useful for thermal analysis of spent fuel shipping casks were selected, acquired, and tested for their applicability to thermal cask design. A series of modeling problems of graduating complexity was selected to test the various codes' actual performance and capabilities. Care was taken in selecting the problems so as to allow quantitative comparison of speed and accuracy of the various computer codes and qualitative evaluation of other computer program capabilities.

In the current reporting period, an information exchange meeting was held between government and industry representatives, hosted by the Technology Transportation Center, Sandia National Laboratories. At this meeting, details of the thermal benchmarking effort were presented. Areas included code selection criteria, candidate codes, model problem criteria and selection, and benchmarked code results. An effort was made to obtain industry participation and thus guide future benchmarking and code selection tasks. A follow-on workshop will be held in late FY 1983 or early FY 1984 to present more detailed results of the benchmarking effort and to exchange results with industry.

6.4.2 Structural Analysis Methods (AR-05-25 - 80747)

The objective of this research is to provide a survey of analytical structural methods applicable for the design and licensing of spent fuel shipping casks. Structural computer code benchmarking is being used in the performance of this survey.

Summary

An information exchange meeting was hosted, and was attended by government and industrial representatives, in which structural and thermal codes relevant to spent fuel shipping cask design were discussed.

Structural Analysis of Casks

In previous reporting periods for this project, several computer codes were identified as potentially useful for structural analysis of spent fuel shipping casks and were acquired. These codes were available to the public either as source forms or on a fee-for-use basis. The codes were benchmarked using simple cask-like models. A series of germane structural response parameters was selected and was tabulated so that the codes could be compared in a consistent way.

During this reporting period an Industrial/Government Joint Thermal and Structural Codes Information Exchange meeting was held. Information on the structural code benchmarking and on a similar thermal code benchmarking activity was presented. The information exchange meeting was hosted by the Transportation Technology Center, Sandia Laboratories, Albuquerque, New Mexico.

6.4.3 Criticality Studies for Spent Fuel Transportation (AR-05-25 - 80263)

The objective of this research is to obtain experimental criticality data in support of the design and safe operation of nuclear fuel transportation systems.

The document Existing Experimental Criticality Data Applicable to Nuclear Fuel Transportation Systems and Near Term Data Needs was issued as TTC-0295/PNL-4118. The data documented in this report will serve as the initial step in developing a firm and complete data base for verifying calculational models used in the criticality safety analyses of transportation systems. An experimental program is planned to provide those missing data considered most beneficial in completing the data base. The initial series of experiments is scheduled to begin in FY 1984 and involves fuel shipping arrangements in which neutron flux traps are created. These experiments are currently being designed in coordination with Sandia Laboratories staff and the Chairman of a Nuclear Energy Agency Work Group assessing the reliability of different criticality calculational techniques.

6.4.4 Nuclear Materials Transportation Cost Studies (AR-05-25 - 80744)

The objective of this research program is to develop the methodologies and data required for estimating shipping costs for all major categories of defense and commercial nuclear waste, and waste packaging alternatives.

Logistics analyses supplied to the nuclear waste management programs of the U.S. Department of Energy, through the Transportation Technology Center (TTC) at Sandia National Laboratories, are used to predict nuclear waste material flow, transportation packaging demands, shipping and receiving rates, and transportation-related costs for alternative strategies. Key to these analyses are the reference shipping costs for various waste materials being developed at the Pacific Northwest Laboratory (PNL).

PNL conducted research for the TTC to develop techniques for determining the costs of shipping irradiated reactor fuel. Information was collected from a truck carrier licensed to serve the 48 contiguous states, and from various rail freight tariff guides (as of January 1982). This collection of information supplied the basis for developing a set of formulas to predict truck, rail, or truck heavy-haul shipping costs. A representative example of these formulas is given

below for the total cost associated with transporting irradiated reactor fuel by truck.

$$\left[\left(AA \cdot \frac{LOAD}{100}\right) + \left(AB \cdot \frac{EMPT}{100}\right) + (DHC \cdot DIS2) + (SVC \cdot DIS3)\right] \cdot (1 + FS) + (AEC \cdot DIS4) + (SEC \cdot DIS1) + OWC + DET$$

where

AA = Value for specified distance traveled with full cask, $/cwt

AB = Value for specified distance traveled with empty cask, $/cwt

LOAD = Weight of full cask, lb

EMPT = Weight of empty cask, lb

SEC = Special equipment charge, $/mile

AEC = Charge for each armed escort, $/man-mile

DHC = Deadhead charge[a] for special equipment, $/mile

SVC = Charge for each separate escort vehicle (if requested), $/mile (each vehicle carries 2 escorts)

FS = Fuel use surcharge

OWC = Applicable overweight charge for each state traversed

DET = Personnel and equipment detention charge

DIS1 = Specified distance traveled with loaded cask, miles

DIS2 = Specified distance from terminal location of special equipment to point of loading, miles

DIS3 = Total distance traveled by separate escort vehicle(s) from terminal location until subsequent return to terminal, miles

DIS4 = Sum of the number of armed escorts (provided by carrier) times the distance traveled with a loaded cask by each escort, man-miles

The formulas reflect only the costs to transport the spent fuel. No cask leasing or purchase costs are included.

In addition to development of the various cost methodologies, a set of data was formulated to represent a hypothetical transport of spent fuel with each transport mode. The results from these data allowed a baseline comparison among the transport modes. The actual fee charged by a truck, rail, or truck heavy-haul carrier is based on the variables listed above and cannot be determined until a contract is signed. A formal report relating the results of this study is expected to be published in June of this year.

Research to predict the costs of transporting other forms of nuclear waste (remote- and contact-handled transuranics, high-level, and low-level) is in progress. Current and conceptual shipping container/cask designs, shipping systems, shipping modes, and combinations of origins and destinations have been investigated for these forms. An applicable set of formulas for each particular waste form will be developed for use in waste disposal cost and logistic studies.

6.5 KRYPTON ION IMPLANTATION (AS-25-05 - 86116)

The objective of this program is to develop the process of implanting krypton in a sputter-deposited metal matrix for ultimate storage of krypton-85. Pilot-scale demonstration of the process, using nonradioactive krypton, and small laboratory-scale tests using krypton-85 will be conducted.

(a) Cost incurred for moving equipment and personnel from their domicile to the shipment origin.

Summary

At the start of FY 1983, a primary objective was the design, construction, and demonstration of a full-scale, remotely-operable sputtering chamber for trapping krypton in a metal matrix. Full-scale is interpreted as a system compatible with a large fuel reprocessing plant, i.e., AGNS. At mid-year, the effort was redirected toward a system suitable for a smaller fuel reprocessing operation, i.e., the Breeder Reprocessing Experimental Test (BRET). The chamber design has been revised such that the diameter and connections will serve both BRET and an AGNS installation; capacity can be adjusted by increasing or reducing the chamber length.

The outer dimensions and connection details of the new disposable chamber have been established as well as the general arrangement of the internal electrodes. However, the final geometry and electrode material selection depends on the successful deposition of a full-term deposit 1 cm thick, using an experimental demountable chamber of similar design. Testing is under way and progress to date using the demountable system with a 1500-cm^2 substrate area indicates that 88% of the design pumping capacity can be achieved using 84% of the input design power. The thickest deposit achieved to date with krypton-loaded nickel-lanthanum is 0.1 cm.

More tests are under way to learn whether modifying the current limiters, pressure control system, and anode geometry and operating procedures will increase the reliability of pressure control and the overall system and whether internal stress in the krypton-loaded deposit will limit the deposit thickness. The detailed mechanical designs have been completed for a remotely operable sputtering station and the service connector assembly. A preliminary layout for a four-chamber arrangement has been made.

Environmental considerations dictate that krypton release be minimized in the storage of ^{85}Kr-containing deposits. Also safety considerations dictate that the release of ^{85}Kr be studied under conditions that could occur during storage, handling, or transportation. Included in these studies are the release of krypton from a Ni-La-Kr alloy in 0.66 torr O_2 and at 320°C and in water at room temperature and the release of krypton from a Ni-Y-Kr alloy during grinding at room temperature.

During oxidation a small amount of krypton is released from the surface of the Ni-La-Kr alloy, but after a short time the alloy becomes passive to further oxidation. After this, the krypton release rate decreases to 20% of the rate before oxidation. Thus the deposit is stable and poses no sizable release hazards if exposed to air.

The release of krypton from the Ni-La-Kr alloy in water at room temperature is greater than in dry oxygen. Based on nearly a year of tests, the estimated 10-year release amounts to 1.76% of the contained krypton. Krypton release from the Ni-La-Kr alloy in water at 90°C is also being measured, and these results will be reported later.

During grinding, krypton is released from the closed pores of the Ni-Y-Kr alloy as they are opened. Our results indicate that krypton is contained predominantly in pores smaller than 12 Å in diameter. Also the krypton content of the alloy decreases rapidly as the particle size falls below 10 μm. This information is consistent with a void size of ~20 Å observed in an electrolytically thinned Ni-Ti-Kr alloy with transmission electron microscopy.

Equipment Development and Testing

The design requirements for fabrication and demonstration of a remotely operable sputtering station for ion-implanting ^{85}Kr in a sputtered Ni-La alloy have been changed to match the krypton output of a smaller fuel reprocessing operation, i.e., the Breeder Reprocessing Experimental Test (BRET). The disposable chamber design has been revised such that the inner substrate diameter of 25 cm and the external electrode and water cooling connections will serve both BRET and on an AGNS installation; capacity can be adjusted by increasing or reducing the chamber length. The design will allow the substrate area to be varied from 1000 to 3000 cm^2 by changing the length of the cylindrical portion of the chamber from 0 to 25 cm. Radiation shielding calculations were completed for the ^{85}Kr-filled disposal trapping unit for the BRET-size facility. A lead thickness of 5.7 cm will be required.

A less expensive unshielded 1500-cm^2 substrate surface area trapping unit (which could be disassembled and modified as necessary) was designed and fabricated and is being used to establish the final internal design of the shielded disposable trapping unit. A sketch of the demountable unit is shown in Figure 6.1. Tests using this unit have demonstrated a pumping speed of 2.2 L/hr at STP with a Ni-La target input power of 21.6 kW for a period just under 1 hr. To meet the design requirements for this size unit, it must pump 2.5 L/hr at STP for about 100 hr with an input target power of 24 kW.

Based on the performance of the test unit, it was decided to proceed with the design of the shielded sputtering station to delay the internal electrode design until a full-term (1 cm thick) deposit could be made. The detailed mechanical design including the service connector assembly has been completed. A reduced assembly drawing is shown in Figure 6.2. A preliminary layout of a

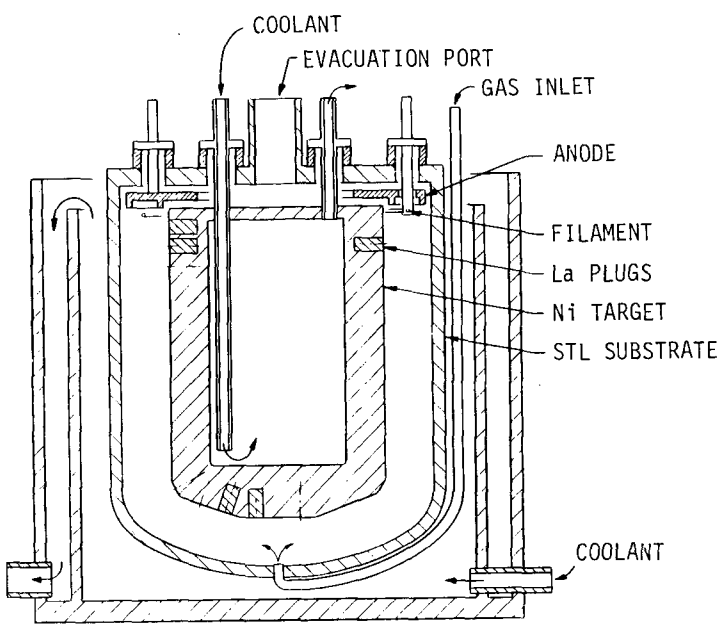
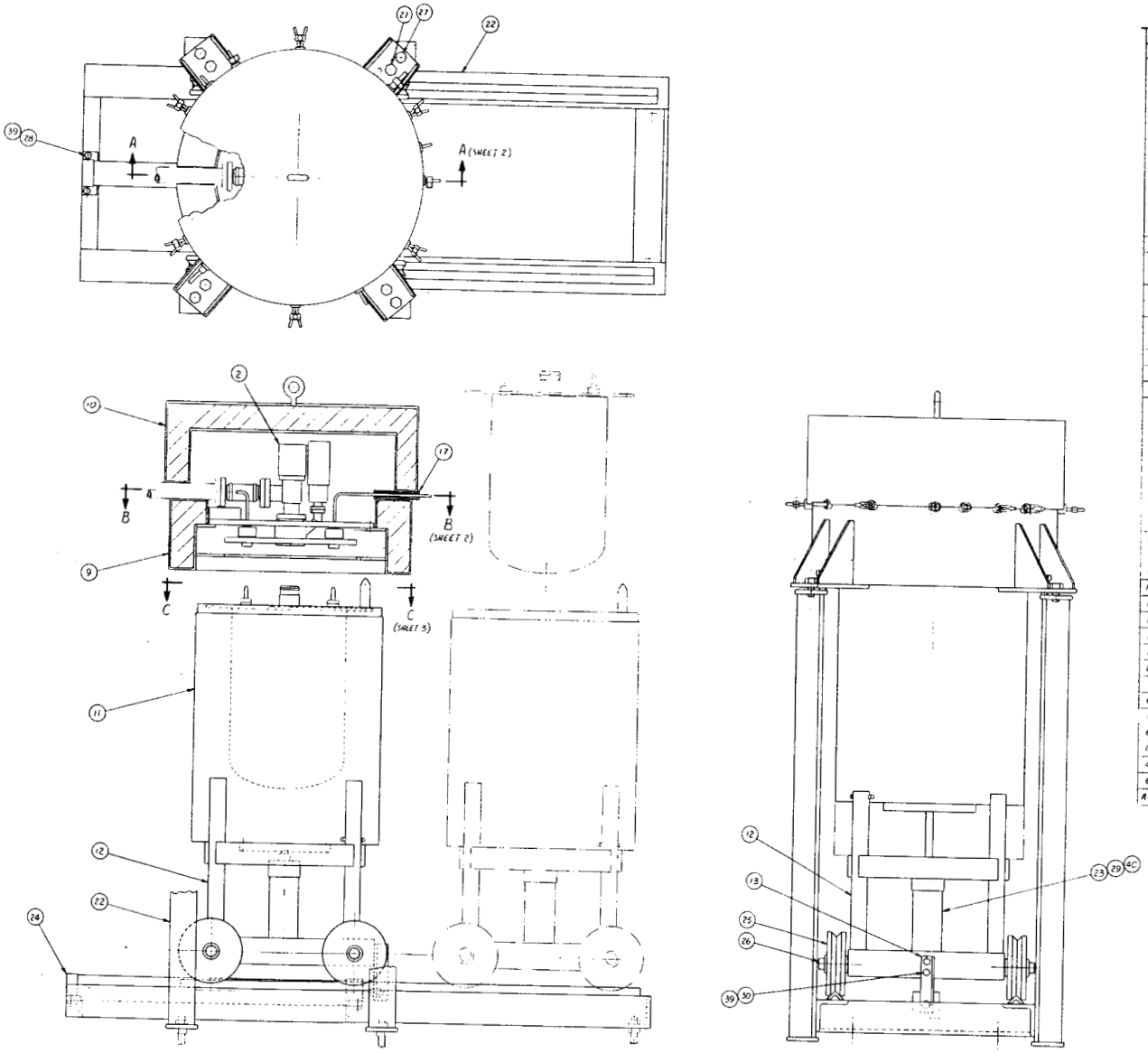


FIGURE 6.1. 1500-cm^2 Disposable Gas Trapping Unit



QTY	PN	DESCRIPTION
	1	ASSEMBLY
1	2	SERVICE CONNECTOR ASSEMBLY
1	3	DISPOSABLE GAS TRAPPING UNIT
1	4	COOLING CAN
1	5	SHIELD
1	6	SHIELD
1	7	GUIDE
1	8	PLATE
1	9	HOLDER
1	10	LID
1	11	LASK
1	12	CART FRAME
1	13	TONGUE
1	14	GUIDE PLATE
5	15	INSULATOR
2	16	
6	17	
6	18	CONNECTOR
1	19	
1	20	
2	21	
1	22	FRAME
1	23	HYDRAULIC CYLINDER MODEL NO J62 B 2 1/2 BORE x 5" STROKE, OPERATING PRESS 300 PSI, SINGLE ACTING, NON CUSHIONED 5/8 DIA STD ROD, STYLE #6 STD ROD END
1	24	HYDRAULIC CYLINDER, MODEL NO J72 B 2" BORE x 24" STROKE, OPERATING PRESS 300 PSI DOUBLE ACTING CUSHION BOTH ENDS, 5/8 ⌀ STD ROD STYLE #6 STD ROD END
4	25	WHEEL, SANTON V-GROOVE 6 DIA x 2" W, 2 3/16 HUB LENGTH AXLE SIZE 3/4 DIA 87°-7/8 WIDE GROOVE STL
4	26	SHOULDER SCR, 3/4 DIA x 2 1/4 L, SOC HD
3	27	CAP SCR 5/8 11UNC 2A x 1 1/4 L HEX HD
4	28	5/8 16UNC 2A x 1 1/4 L
10	29	5/16 18UNC 2A x 1" L
6	30	3/8 16UNC 2A x 1" L
8	31	1/4 20UNC 2A x 5/8 L SLOTTED FL HD
2	32	1/4 20UNC 2A x 2" L SLOTTED RD HD
2	33	1/4 20UNC 2A x 1 3/4 L, SLOTTED RD HD
8	34	1/4 20UNC 2A x 1/2 L HEX HD
1	35	3/8 16UNC 2A x 3/4 L HEX HD
6	36	#8-32UNC 2A x 3/4 L SCH
1	37	GASKET 953 5070
6	38	HEX NUT #8 32UNC-2B
6	39	3/8-16UNC-2B
4	40	5/16 18UNC 2B
6	41	PLAIN WASHER 5/16 NOMINAL
AR	42	TUBING 1/4 OD x .035 WALL

FIGURE 6.2. Sputtering Station

four-chamber arrangement also has been made, as shown in the reduced, unfinished drawing, Figure 6.3.

Further experimentation with the 1500-cm^2 test unit has resulted in deposits of krypton-loaded Ni-La of up to 0.1 cm thick. The chief factors necessary for achieving reliable, continuous operation are: 1) absence of electrical shorts between electrodes as a result of deposit buildup, nonadherent deposited material, or movement of electrodes due to thermal expansion or warpage or a combination of both; 2) properly protected power supplies that can return quickly to the

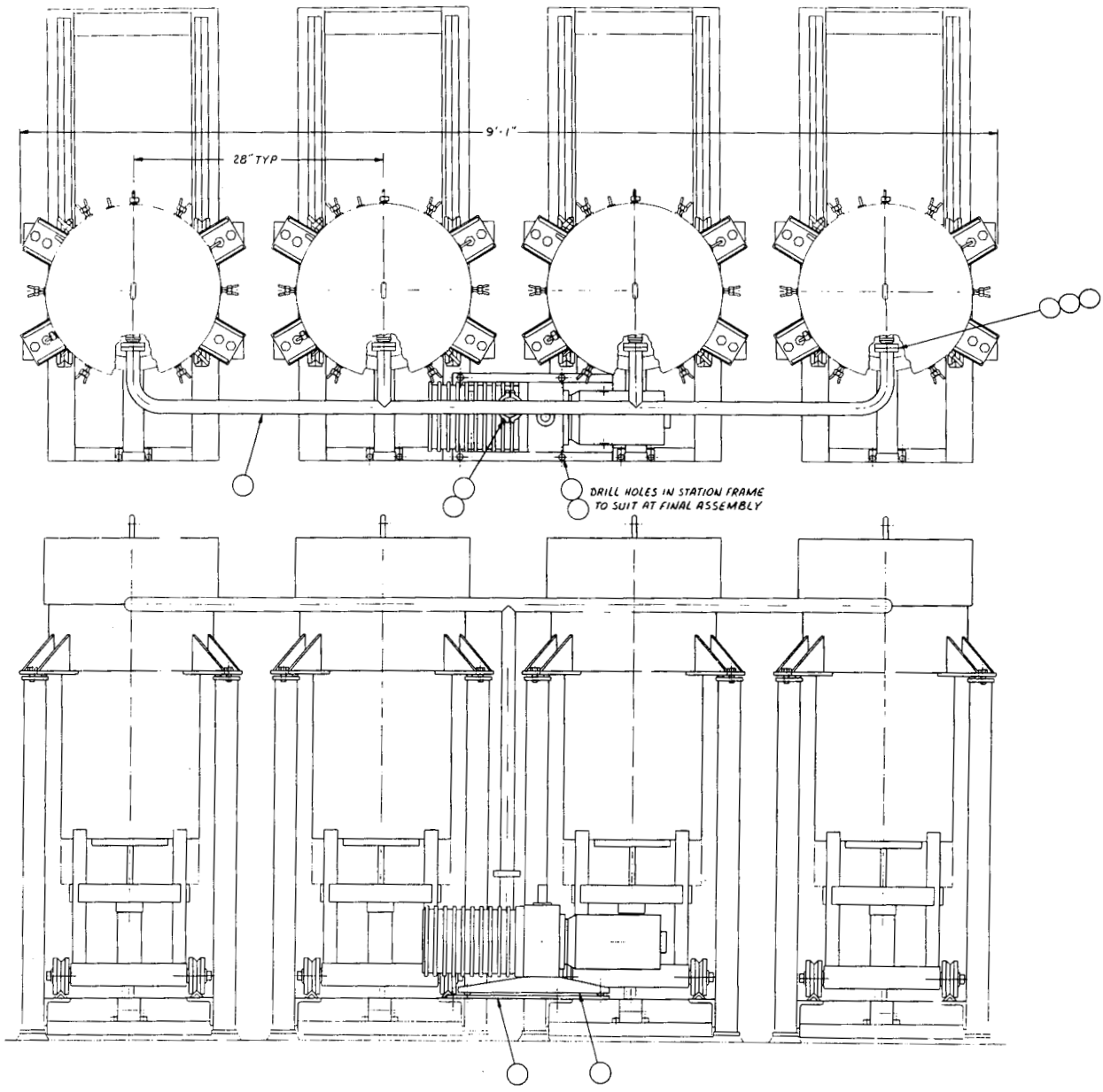


FIGURE 6.3. Four-Chamber Arrangement

desired current settings after faults are cleared; 3) adequate control of gas pressure; and 4) temperature control of all electrodes. Failure to meet these requirements will result in a malfunction of the unit. Two weak points that prohibit reliable operation of the present unit are the failure of the target power supply limiter to operate properly above 2100 V and the lack of dimensional stability of the anode-filament configuration.

Since spurious rf oscillations appearing when the target potential exceeds 2100 V have prevented full power operation, the power supply/process controller network was improved in several ways. The system was modified to stop sputter deposition if the gas trapping rate falls below a preset level, provide faster recovery from electrode arcing, and improve power supply control. Additional filtering was added to the filament and plasma current signal inputs to improve the computer reading accuracy. A new output transistor was successfully tested in the substrate limiter on a dummy load bank. Also a newly designed 100-A, air-cooled plasma limiter was load-bank tested. An adjustable load bank with an integral shorting relay for 2500-V operation was designed and fabricated to permit long-term pulsed testing of combinations of high-voltage power supply limiters at the 25-kW level. An order has been placed for a commercial target power supply with a current cutback circuit of comparable size. This unit is reported to successfully power a sputtering target for a plasma isotope separation process.

Test runs with the latest anode configuration indicate that the anode temperature during full-power operation is insufficient to achieve significant deposit vaporization but high enough for deposit melting. Tests with anodes constructed of tantalum or molybdenum show that either material is suitably stable. It must be shown, however, that the buildup of liquid nickel-lanthanum will not cause an electrical short between the anode and adjacent structures.

In March, the pumping station, power supply, and control console were relocated from the 231-Z Building at the north end of the Hanford Reservation to the Research Technology Laboratory (RTL) near Richland. Experiments will continue as soon as the equipment is operable using a newly completed steel substrate.

Kinetics of Krypton Release

Environmental considerations dictate that krypton release be minimized in the storage of ^{85}Kr-containing deposits. It is also desirable that the release of ^{85}Kr be understood in the event of nonstandard conditions during storage or transportation. With these goals in mind, studies involving the release behavior of krypton sputter-deposited nickel-lanthanum alloy in O_2 at 320°C, in water at room temperature, and in water at 90°C have been added to the study of the isothermal release rate measurements in the range 250 to 450°C. Also added to these studies was the effect of grinding on krypton release and determination of particle porosity and krypton content measured as a function of the particle size. The grinding study was performed on the nickel-yttrium alloy because it required a large amount of material. The purpose of the studies is to attempt to understand how krypton is distributed in the solid and by what mechanisms it is released.

Release Behavior in Oxygen

The test to determine the krypton release behavior of the Ni-La-Kr alloy during continuous exposure to atmospheric concentrations of oxygen has continued for about 11 months. These results are shown in Figure 6.4. During that time, three different release-rate regimes have been observed. In each regime, the total krypton released is proportional to the square root of time, indicating that diffusion is the rate-controlling mechanism for krypton release. The first regime lasted 26 days, and the krypton released was ~220% as great as would be expected in vacuum (dashed line in Figure 6.4) without oxygen exposure. The second regime continued for another 115 days, and the krypton released was ~90% as great as would be expected in vacuum without oxygen exposure. The third regime has continued for 190 days and is still continuing, and the krypton released was about 20% as great as would be expected in vacuum without oxygen exposure.

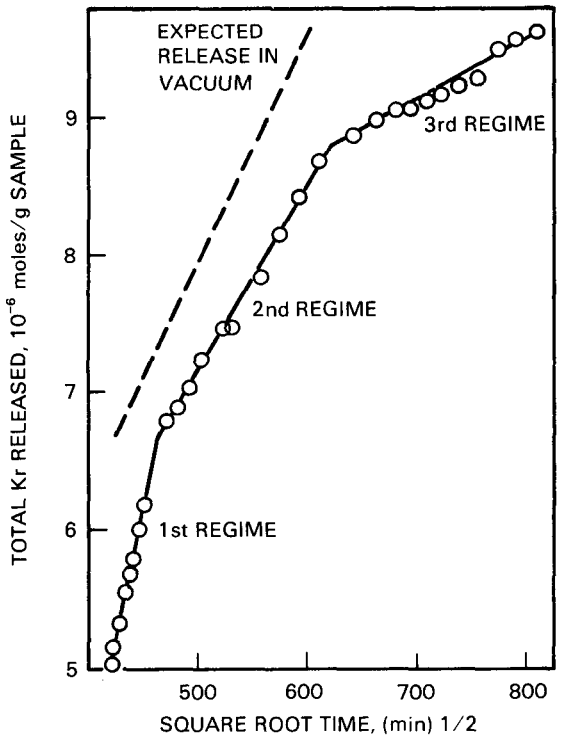


FIGURE 6.4. Release of Krypton at 320°C in 166 Torr of Oxygen from an Ni-Li-Kr Alloy Initially Containing 1.32 x 10^{-3} Moles Kr/g Sample

The overall release was much less than if no oxygen exposure had occurred because of the markedly decreased krypton release rate during the last 190 days of exposure. This is true even though it is apparent from these results that oxidation of the alloy causes the release of krypton during the first 141 days of exposure.

It is proposed that the mechanisms for release of krypton during the three regimes are as follows:

1. During the first 26 days, the krypton release rate was controlled by the oxidation rate or the diffusion of oxygen through the oxide layer already formed on the metal surfaces. After 26 days, the outer surface of the alloy became resistant to further oxidation, and the krypton release rate decreased.
2. During the second regime, oxidation was apparently still continuing because the krypton release rate was still 4.5 times as great as in the third regime. The krypton release rate in the second regime was apparently still controlled by the rate of oxidation, which may have been controlled by the diffusion of oxygen into pores with restricted openings to the surface or by a part of the material that oxidizes more slowly but for a longer time than the other part.
3. In the third regime, the alloy has apparently become resistant to further oxidation, and the rate-controlling mechanism for krypton release has become the diffusion of krypton through the oxide layer.

Evidently the rate of diffusion through the oxide layer is only about 20% of the rate through the unoxidized metal because the release rate is only about 20% of the rate of the unoxidized metal.

The results of this test are consistent with those of a test reported earlier (Tingey 1982). In that test a sample was intermittently exposed to oxygen for 4 hr at 320°C, and it was concluded that an oxide layer forms on the alloy that is resistant to diffusion of both oxygen and krypton and leads to a krypton release one-sixth that of the expected release with no oxygen exposure. The tests reported here show that continuous exposure of the sample to oxygen eventually results in a decreased release rate of about the same magnitude as the 4-hr exposures did, and that the Ni-La-Kr alloy eventually becomes resistant to further oxidation.

The results of this test show that the Ni-La-Kr alloy is stable in oxygen and that the krypton release rate decreases by a factor of five after a stable oxide film forms on the surface.

Release Behavior in Water

The study of krypton release behavior of the Ni-La-Kr alloy in water has shown that water does not greatly increase the krypton release rate. This study is still continuing; but for 300 days in room temperature water, the release is consistent with a diffusion mechanism releasing only about 1.8% of its total krypton after 10 years.

Grinding Studies

During grinding, krypton was released from the Ni-Y-Kr alloy as fracture lines extended through closed pores containing krypton, thus opening them and releasing the krypton. Particle size distributions after grinding are shown in Figure 6.5. As seen in Table 6.1, open porosity increased and krypton was released during grinding. However, the krypton released was greater than could have been contained in the pores that were opened. This is illustrated by using the liquid density to calculate the maximum volume of krypton that could be contained in pores, which is 0.578 L Kr (STP) per mL pore volume. The amount of krypton released during grinding was far greater than the amount that could have been contained in the 12 to 800 Å pores that were measured (see Table 6.1). Then a large volume of pores either >800 Å or <12 Å must have been opened, or possibly pores were destroyed during grinding.

It is proposed that pores in the Ni-Y-Kr alloy were mostly <12 Å in diameter because: 1) it appears the volume of pores >800 Å was

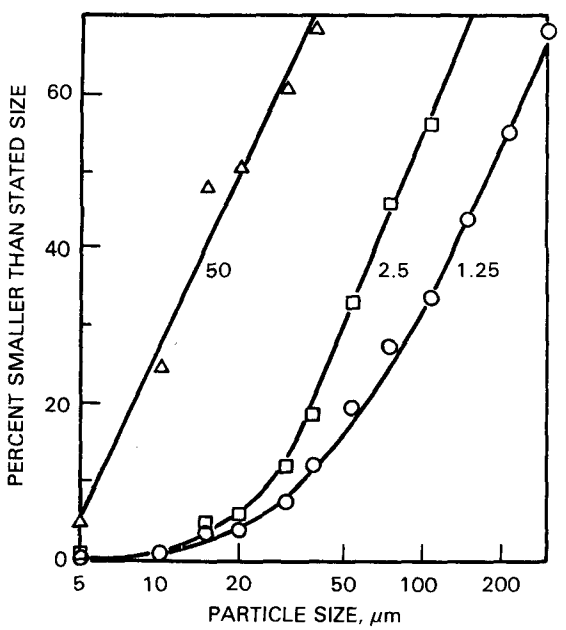


FIGURE 6.5. Particle Size Distributions After Various Grinding Times (shown in minutes on lines)

TABLE 6.1. Krypton Release, Pore Volume of Pores 12 to 800 Å Diameter and Surface Area of Pores Larger than 12 Å During Grinding for the $Ni_{0.82}Y_{0.11}Kr_{0.07}$ Alloy

Grinding Time, min	Increase in Open Pore Volume, μL/g Sample	Surface Area, m^2/g Sample	L Kr (STP) / mL Pores	% of Contained Kr Released
1.25	0.17	0.107	1.1	0.8
2.5 0.17	0.108	1.9	1.4	
5 0.15	0.109	2.2	1.5	
20 0.46	0.189	7.5	15.4	
50 0.60	0.263	24.0	64.4	

small because pores were mostly 400 Å and smaller and 2) it appears that pores were not destroyed because if pores were destroyed during grinding, surface area would increase at a greater rate than porosity. But, as seen in Table 6.1, surface area and porosity increased at comparable rates. Thus the porosity and release measurements lead to the conclusion that more than 95% of the krypton is contained in pores less than 12 Å in diameter, but that there are significant pores of larger diameter that may affect release mechanisms. In other work a void size of ~20 Å was observed in an electrolytically thinned Ni-Ti-Kr alloy with transmission electron microscopy. To test the effect of particle size and porosity on release rate, release under isothermal conditions will be measured on several particle-size fractions.

Measurement of the krypton content of various particle-size fractions of the Ni-Y-Kr alloy after grinding is shown in Figure 6.6. The krypton content is maximum in particles ~80 μm in diameter and is greater than in the bulk alloy before grinding. The decrease in krypton content with particle size in particles <80 μm is explained by the release of krypton during grinding as fracture lines include pores, thus opening them and releasing the krypton. Additional work is needed

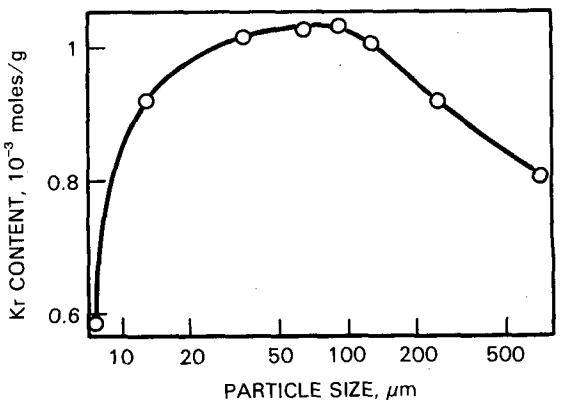


FIGURE 6.6. Krypton Content of $Ni_{0.82}Y_{0.11}Kr_{0.7}$ Alloy Versus Particle Size After Grinding in a Ball Mill. Dashed line represents krypton of the bulk alloy before grinding.

to explain the decrease in krypton content in particles >80 μm.

Reference

Tingey, G. L. 1982. "Krypton Implantation." Nuclear Waste Management Semiannual Progress Report, October 1982 Through March 1983. PNL-4250-2, Pacific Northwest Laboratory, Richland, Washington, pp. 3.1-3.7.

6.6 IODINE-129 FIXATION (AS-25-05 - 87128)

The objective of this program is to develop technologies that will provide safe and 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.

The iodine fixation program was completed during this report period. Work consisted of analysis of experimental data and the preparation of reports. The following papers and reports were written during the period:

PNL-4489, Methyl Iodide Sorption by Reduced Silver Mordenite, R. D. Scheele, L. L. Burger, and C. L. Matsuzaki

PNL-4490, Recycle of Iodine-Loaded Silver Mordenite by Hydrogen Reduction, L. L. Burger and R. D. Scheele.

PNL-SA-10896, "Leach Resistance of Iodine Compounds in Portland Cement," R. D. Scheele, L. L. Burger, and K. D. Wiemers. Am. Chem. Soc. 185th National Meeting, Seattle, Washington, 1983.

PNL-4689, The Status of Radioiodine Control for Nuclear Fuel Reprocessing Plants, L. L. Burger and R. D. Scheele.

The last-named report is the final one of the program. It summarizes in capsule form the concepts for capture of airborne iodine and for fixation and disposal. Capture processes are judged with respect to:

- compatibility with overall airborne waste treatment
- amenability of capture form to treatment for storage or disposal
- other advantages and disadvantages
- recommended studies or demonstrations.

Fixation and disposal are more difficult to evaluate because of lack of criteria. Several concepts appear to give adequate control for a few hundred to a few thousand years. It is suggested that adequate dispersal of ^{129}I is the best long-range option for iodine control.

6.7 THERMAL OUTGASSING (AR-05-15-05 - 80246)

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 fuel*
2. *evaluate the use of thermal outgassing using various sweep gases or vacuum outgassing for the removal of* ^{14}C, ^{129}I, ^{85}Kr *and* ^{3}H
3. *apply the information obtained from both experiment and theory to spent fuel reprocessing, interim and long-term storage, and to reactor safety.*

The work during the period consisted of analysis of data and preparation of reports. A report, PNL-4487, Thermal Outgassing of Irradiated Nuclear Fuel - A Literature Review was written by L. G. Morgan, L. L. Burger, L. A. Bray, and C. L. Matsuzaki. The final report, PNL-4488, Removal of Fission Products by Thermal Outgassing was written by L. A. Bray, L. L. Burger, and L. G. Morgan.

In this study the thermal release of fission gases from irradiated nuclear fuels was achieved with the use of a sweep-gas. Vacuum outgassing was partially successful, but the sweep-gas technique besides being more effective was found easier to employ. The following were demonstrated:

- Complete release of krypton is obtained.
- Complete release of tritium from both the fuel and cladding is obtained.
- The addition of 6% H_2 to the helium sweep-gas results in greatly improved tritium release as a function of time but hinders the krypton release.
- Most of the iodine and cesium are released when the inert gas is used. When 6% H_2 is present, the cesium remains in the fuel. Additional data are required to explain the behavior of iodine, cesium, and other semivolatile fission products.
- Under some conditions, the cladding fractured in the heat treatment; thus chopping the fuel may not be necessary. Further investigation of cladding fracturing and interaction with UO_2 could lead to an alternative head-end process.

- The dramatic effect of the presence of hydrogen in the helium sweep-gas indicates that the composition of the atmosphere around the heated fuel is very important to fission gas and semivolatile release. Observations such as the appearance of iodine as I_2 suggest that experiments using traces of other gases such as O_2 in the sweep gas would be instructive. Other inert gases such as argon and N_2 need to be studied.
- The heat treatment did not significantly affect the dissolution properties of the fuel in nitric acid.
- An engineering evaluation of the process as a head-end step for fuel reprocessing was carried out and showed that thermal sweep-gas out-gassing is technically feasible and cost-competitive.

The program ends with this report.

6.0 SUPPORTING STUDIES

The objective of the Waste Management Supporting Studies are to do the following: 1) focus technical analysis on the independent assessment of the information base upon which to make decisions to implement waste management systems and technologies of interest, 2) review on-going technology base activities, 3) provide support to DOE's Nuclear Waste Policy Act Project Office, and 4) conduct environmental impact studies and analysis of public and institutional issues.

7.0 RECENT WASTE MANAGEMENT PUBLICATIONS

7.0 RECENT WASTE MANAGEMENT PUBLICATIONS

Allen, M. D. and L. G. Morgan. 1982. "Comments on the Note by L. S. Nelson: 'Unusually High (Oxidizer/Pu) Ratios in the Macro-Residues from Plutonium Droplet Combustion.'" High Temperature Science, 15:321-326.

Allen, R. P., L. K. Fetrow and M. W. McCoy. 1982. "An Integrated Decontamination System for Surface-Contaminated TRU Waste." In The Treatment and Handling of Radioactive Wastes, A. G. Blasewitz, J. M. Davis and M. R. Smith, eds., pp. 98-102. Battelle Press, Columbus and Richland.

Andrews, W. B. (PNL) and R. H. Jones (Hazardous Material Systems). 1982. "Transportation Packaging for Processed Transuranic Waste." In The Treatment and Handling of Radioactive Wastes, A. G. Blasewitz, J. M. Davis and M. R. Smith, eds., pp. 595-600. Battelle Press, Columbus and Richland.

Baker, E. G., H. D. Freeman and J. N. Hartley. "Cost and Effectiveness of Radon Barrier Systems." Uranium Mill Tailings Management, pp. 383-396. Proceedings of the Fifth Symposium on Uranium Mill Tailings, December 9-10, 1982. Colorado State University, Fort Collins, Colorado.

Barnes, S. M. and J. L. Buelt. "Accelerated Aging of Catalytically Airblown Asphalt Membranes Under Simulated Uranium Mill Tailings Impoundment Conditions." Uranium Mill Tailings Management, pp. 433-440. Proceedings of the Fifth Symposium on Uranium Mill Tailings, December 9-10, 1982. Colorado State University, Fort Collins, Colorado.

Barnes, S. M., G. J. Sevigny and R. W. Goles. 1982. "Corrosion Experience with a Slurry-Fed Ceramic Melter." In The Treatment and Handling of Radioactive Wastes, A. G. Blasewitz, J. M. Davis and M. R. Smith, eds., pp. 203-207. Battelle Press, Columbus and Richland.

Beedlow, P. A., L. L. Cadwell and M. C. McShane. "A Systematic Approach to Designing Surface Covers for Inactive Uranium Mill Tailings." Uranium Mill Tailings Management, pp. 397-408. Proceedings of the Fifth Symposium on Uranium Mill Tailings, December 9-10, 1982. Colorado State University, Fort Collins, Colorado.

Beedlow, P. A. and M. C. McShane. 1982. Revegetation/Rock Cover for Stabilization of Inactive Uranium Mill Tailings Disposal Sites. PNL-4328, UMT-0210.

Bjorklund, W. J. and R. A. Brouns. 1982. "Operating Characteristics of a Direct Liquid-Fed Vitrification Process." In The Treatment and Handling of Radioactive Wastes, A. G. Blasewitz, J. M. Davis and M. R. Smith, eds., pp. 197-202. Battelle Press, Columbus and Richland.

Bjorklund, W. J., R. A. Brouns, R. D. Dierks, R. W. Goles et al. 1982. Defense Waste Vitrification Studies During FY-1981. PNL-4366.

Bradley, D. J., D. G. Coles, F. N. Hodges, G. L. McVay and R. E. Westerman. 1982. Nuclear Waste Package Materials Testing Report: Basaltic and Tuffaceous Environments. PNL-4452.

Brodzinski, R. L. and W. K. Hensley. 1982. "Instrumentation for the Verification of the Radionuclide Content of Waste Packages." In Proceedings of the Fourth Annual Participants Information Meeting DOE Low-Level Waste. ORNL/NFW/82/18, pp. 619-626.

Brodzinski, R. L. and W. K. Hensley. 1982. "Well Logging Instrumentation for the In Situ Determination of ^{90}Sr, Tritium and Transuranics." In Proceedings of the Fourth Annual Participants Information Meeting DOE Low-Level Waste Management. ORNL/NFW/82/ 18, pp. 437-455.

Buckwalter, C. Q. and L. R. Pederson. 1982. "Inhibition of Nuclear Waste Glass Leaching by Chemisorption." J. of Am. Cer. Soc., 65a:431-436.

Buckwalter, C. Q., L. R. Pederson and G. L. McVay. 1982. "The Effects of Surface Area to Solution Volume Ratio and Surface Roughness on Glass Leaching." Journal of Non-Crystalline Solids, 49:(1-3):397.

Buelt, J. L. 1982. "The Feasibility of Incinerating and Vitrifying Organic Resins in a Single Step." In The Treatment and Handling of Radioactive Wastes, A. G. Blasewitz, J. M. Davis and M. R. Smith, eds., pp. 504-509. Battelle Press, Columbus and Richland.

Burger, L. L. and R. D. Scheele. 1982. Recycle of Iodine-Loaded Silver Mordenite by Hydrogen Reduction. PNL-4490.

Burkholder, H. C. 1982. "Engineered Components for Spent Fuel Radioactive Waste Isolation Systems--Are They Technically Justified?" In Scientific Basis for Nuclear Waste Management V, W. Lutz, ed., pp. 619-628. Proceedings of the Materials Research Society Fifth International Symposium on the Scientific Basis for Nuclear Waste Management, held June 7-10, 1982, in Berlin, Germany. Elsevier Science Publishing Company.

Carrell, J. R., L. K. Holton and D. H. Siemens. 1982. "High-Level Waste Solidification System for the Western New York Nuclear Service Center." In The Treatment and Handling of Radioactive Wastes, A. G. Blasewitz, J. M. Davis and M. R. Smith, eds., pp. 547-552. Battelle Press, Columbus and Richland.

Chick, L. A. and R. P. Turcotte. 1982. Glass Leaching Performance. PNL-4576.

Cline, J. R., D. A. Cataldo, W. E. Skiens and F. G. Burton. 1982. "Biobarriers Used for Shallow Burial Ground Stabilization." Nuclear Technology, 58:150-153.

Coles, D. G., R. W. Mensing, J. Rego, H. C. Weed and R. W. Buddemeir. 1982. A Leaching Study of PNL-76-68 Glass Beads Using the LLL Continuous-Flow Method and the PNL Modified Method: A Final Report. UCRL-19492 (Rev. 1).

Daling, P. M. and R. L. Engel. 1983. Analysis of Near-Term Transportation Hardware Requirements and Transportion Costs. PNL-4575.

Danielson, M. "An Evaluation of the Three-Point Method to Measure Corrosion Rates." Corrosion, 38:580-586.

Department of Energy. 1982. Low-Level Radioactive-Waste-Management Handbook Series: Methods to Decrease Low-Level-Waste Generation. DOE/LLW-13TB.

Department of Energy, Richland Operations Office. January 1983. Spent Fuel Storage Requirements (an update of DOE-RL-82-1). DOE/RL-83-1.

Dove, F. H., C. R. Cole and M. G. Foley. 1982. AEGIS Technology Demonstration for a Nuclear Waste Repository in Basalt. Assessment of Effectiveness of Geologic Waste Isolation Systems. PNL-3632.

Elmore, M. R., C. T. Li and J. N. Hartley. 1982. "Interim Stabilization of Uranium Mill Tailings." Uranium Mill Tailings Management, p. 91, Colorado State University, Fort Collins, Colorado.

Engel, R. L. and P. M. Daling. 1982. "Defense TRU Waste Transportation and Receiving Simulation." In The Treatment and Handling of Radioactive Wastes, A. G. Blasewitz, J. M. Davis and M. R. Smith, eds., pp. 601-605. Battelle Press, Columbus and Richland.

Engel, R. L. and M. K. White. 1982. Fiscal Implications of a 1-Mill/kWh Waste Management Fee. PNL-4513.

Fahey, A. and W. J. Weber. 1982. "The Crystal Structure and Stoichiometry of the $Ca_{2+x}Nd_{8-x}(SiO_4)_6O_{2-1/2}$ x System." The Rare Earths in Modern Science and Technology, 3:341-344.

Fetrow, L. K., R. P. Allen and M. W. McCoy. 1982. "Sectioning Techniques for Plutonium Contaminated Glove Boxes." In The Treatment and Handling of Radioactive Wastes, A. G. Blasewitz, J. M. Davis, and M. R. Smith, eds., pp. 103-108. Battelle Press, Columbus and Richland.

Gallucci, R. H. V. 1982. "A Risk Assessment for the Transportation of Radioactive Zeolite Liners." In The Treatment and Handling of Radioactive Wastes, A. G. Blasewitz, J. M. Davis and M. R. Smith, eds., pp. 510-514. Battelle Press, Columbus and Richland.

Gilbert, E. R. and A. B. Johnson, Jr. 1982. "Assessment of the LWR Fuel Inventory for Dry Storage." In Proceedings of the ANS Topical Meeting - LWR Spent Fuel Storage, held in Savannah, Georgia, September 1982.

Goles, R. W. and G. J. Sevigny. "Off-Gas Characteristics of Liquid-Fed Joule-Heated Ceramic Melters." CONF-820833, Proceedings of the DOE Nuclear Air Cleaning Conference, held in Denver, Colorado, August 1, 1982.

Gray, W. J. 1982. "A Study of the Oxidation of Graphite in Liquid Water for Radioactive Waste Storage Applications." Radioactive Waste Management and the Nuclear Fuel Cycle, 3(2):137-149.

Guenther, R. J. 1983. Results of Simulated Abnormal Heating Events for Full-Length Nuclear Fuel Rods. PNL-4555.

Harmon, K. M. and L. T. Lakey. 1982. "A Survey of Terminal Waste Storage Programs Throughout the World." In Proceedings of the 1982 National Waste Terminal Storage Program Information Meeting, DOE/NWTS-30, pp. 131-136. Office of NWTS Integration, Battelle Project Management Division, Columbus, Ohio.

Hartley, J. N. et al. 1982. "Uranium Mill Tailings Remedial Action Project (UMTRAP)--Cover and Liner Technology Development Project." In Management of Wastes from Uranium Mining and Milling, IAEA-SM-262. Proceedings of an International Symposium jointly organized by the International Atomic Energy Agency and the OECD Nuclear Energy Agency, held in Albuquerque, New Mexico, May 10-14, 1982. International Atomic Energy Agency, Vienna.

Hartley, J. N., G. W. Gee, E. G. Baker and H. D. Freeman. 1983. 1981 Radon Barrier Field Test at Grand Junction Uranium Mill Tailings Pile. DOE/UMT0213, PNL-4539.

Hodges, F. N., J. H. Westsik, Jr. and L. A. Bray. 1982. "Development of a Backfill for Containment of High-Level Nuclear Waste." In Scientific Basis for Nuclear Waste Management V, W. Lutze, ed., pp. 641-648. Proceedings of the Materials Research Society Fifth International Symposium on the Scientific Basis for Nuclear Waste Management, held in Berlin, Germany, June 7-10, 1982. Elsevier Science Publishing Co., New York, New York.

Holton, L. K. and D. E. Larson. 1982. "Evaluation of Process Alternatives for Solidification of the West Valley High-Level Liquid Wastes." In The Treatment and Handling of Radioactive Wastes, A. G. Blasewitz, J. M. Davis and M. R. Smith, eds., pp. 537-541. Battelle Press, Columbus and Richland.

International Atomic Energy Agency. 1982. Storage of Water Reactor Spent Fuel in Water Pools--Survey of World Experience. (A. B. Johnson, Jr., et al.) IAEA Technical Report Series No. 218, Vienna.

Johnson, A. B., Jr. and W. J. Bailey. 1982. "Assessment of Interim Storage of Spent High-Burnup LWR Fuel." American Nuclear Society Transactions, 40:150-152.

Johnson, A. B., Jr. and W. J. Bailey. 1982. "Current Experience with Fuel Handling and Reconstitution: Relationship to Rod Consolidation," PNL-SA-10341, August. In Proceedings of the ANS Topical Meeting on LWR Spent Fuel Storage, held in Savannah, Georgia, September 1982.

Johnson, A. B., Jr., W. J. Bailey, E. R. Gilbert and J. T. Mayer. 1982. "Materials Behavior in Interim Storage of Spent Fuel." Transactions of American Nuclear Society, 43:314-315.

Johnson, A. B., Jr., P. A. Pankaskie and E. R. Gilbert. 1982. "Spent Fuel Behavior in Dry Storage." In Proceedings of the OECD Specialist Workshop on Technology for the Dry Storage of Spent Fuel Elements, held in Madrid, Spain, May 11-13, 1982.

Jones, T. L., G. W. Gee, J. L. Swanson and R. R. Kirkham. 1983. "A Laboratory and Field Evaluation of the Mobility of Cobalt-60/EDTA." Waste Management '83, Tucson, Arizona.

Kennedy, W. E., Jr. and B. A. Napier. 1982. "Allowable Residual Contamination Levels: Transuranic Advanced Disposal Systems for Defense Waste." Transactions of the American Nuclear Society, 43:99-101.

Kennedy, W. E., Jr., B. A Napier and J. K. Soldat. 1982. "Transuranic Advanced Disposal Systems: Preliminary ^{239}Pu Waste-Disposal Criteria for Hanford". In Proceedings of the Alpha-Contaminated Waste Management Workshop (CONF-820845) held in Gaithersburg, Maryland, August 10-13, 1982. Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Kircher, J. F. and D. J. Bradley. "NWTS Waste Package Design and Materials Testing." In Scientific Basis for Nuclear Waste Management VI, D. G. Brookins, ed., pp. 383-390. Symposium held November 1982 in Boston, Massachusetts. Elsevier Science Publishing Co., New York, New York

Kirkham, R. R., P. A. Beedlow and G. W. Gee. "Effect of Rock Cover on Multilayer Radon Barrier Effectiveness." Uranium Mill Tailings Management, pp. 409-420. Proceedings of the Fifth Symposium on Uranium Mill Tailings, December 9-10, 1982. Colorado State University, Fort Collins, Colorado.

Kuhn, W. L. and R. D. Peters. "Leach Models for a Commercial Nuclear Waste Glass." In Scientific Basis for Nuclear Waste Management VI, D. G. Brookins, ed., pp. 167-174. Proceedings of the Materials Research Society Symposium held November 1982 in Boston, Massachusetts. Elsevier Science Publishing Co., New York, New York.

Lakey, L. T. 1982. "Status of Foreign Practices for the Management of Alpha-Contaminated Radioactive Wastes." In Proceedings of the Alpha-Contaminated Waste Management Workshop (CONF-820845) held in Gaithersburg, Maryland, August 10, 1982. Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Lakey, L. T. and K. M. Harmon. 1983. International Fuel Cycle and Waste Management Technology Exchange Activities Sponsored by the United States Department of Energy--FY-1982 Evaluation Report. PNL-4637.

Langstaff, D. C., W. J. Bailey, A. B. Johnson, Jr., M. P. Landow, V. Pasupathi and R. W. Klingensmith. 1982. Examination of Stainless Steel-Clad Connecticut Yankee Fuel Assembly S004 After Storage in Borated Water. PNL-3828.

Lokken, R. O., L. A. Chick and L. E. Thomas. Development and Characterization of Basalt Glass Ceramics for the Immobilization of Transuranic Wastes. PNL-4136.

Ludwick, J. D. 1982. A Postulated Licensing Schedule for an Independent Spent Fuel Storage Installation. PNL-4498.

Materials Characterization Center. 1982. Workshop on the Leaching Mechanisms of Nuclear Waste Forms, May 19-21, 1982. PNL-4382.

Mayer, D. W. and G. W. Gee. 1983. Multidimensional Simulation of Radon Diffusion Through Earthen Covers. DOE/UMT-0212. PNL-4458.

McCoy, M. W., R. P. Allen and L. K. Fetrow. "Vibratory Finishing for Decontamination--Pilot Scale Operation." In The Treatment and Handling of Radioactive Wastes, A. G. Blasewitz, J. M. Davis, M. R. Smith, pp. 109-114. Battelle Press, Columbus and Richland.

McElroy, J. L., W. J. Bjorklund and W. F. Bonner. "Waste Vitrification: A Historical Perspective." In The Treatment and Handling of Radioactive Wastes, A. G. Blasewitz, J. M. Davis and M. R. Smith, eds, pp. 171-177. Battelle Press, Columbus and Richland.

McVay, G. L. and C. Q. Buckwalter. 1983. "The Effect of Iron on Waste Glass Leaching." J. of Am. Cer. Soc., 66(3):170-174.

McVey, L. A., R. L. Brodzinski and T. M. Tanner. 1983. "In Situ Neutron Activation Analysis of and the Neutron Capture Cross Section for ^{90}Sr." J. Radioanal. Chem., 76:131.

Mendel, J. E. 1982. "The Scientific Basis for Long-Term Prediction of Waste-Form Performance Under Repository Conditions." In Scientific Basis for Nuclear Waste Management VI, D. G. Brookins, ed., pp. 1-8. Proceedings of the Materials Research Society Symposium held November 1982 in Boston, Massachusetts. Elsevier Science Publishing Co., New York, New York.

Mendel, J. E. 1982. "Waste Glasses, Requirements and Characteristics." In The Treatment and Handling of Radioactive Wastes, A. G. Blasewitz, J. M. Davis and M. R. Smith, eds., pp. 178-183. Battelle Press, Columbus and Richland.

Merrill, E. T. and J. F. Fletcher. 1983. Economics of At-Reactor Spent Fuel Storage Alternatives. PNL-4517.

Merz, M. D. 1982. Materials Characterization Center State-of-the-Art Report on Corrosion Data Pertaining to Metallic Barriers for Nuclear Waste Repositories. PNL-4474.

Morgan, L. G., L. L. Burger, L. A. Bray and C. L. Matsuzaki. Thermal Outgassing of Irradiated Nuclear Fuel - A Literature Review. PNL-4487.

Napier, B. A. and W. E. Kennedy, Jr. 1982. "How Clean Is Clean Enough?: Allowable Residual Contamination Levels." Transactions of the American Nuclear Society, 43:98-99.

Nielson, K. K. et al. 1982. "Radon Emanation Characteristics of Uranium Mill Tailings." Uranium Mill Tailings Management, p. 355. Colorado State University, Fort Collins, Colorado.

Oma, K. H., R. K. Farnsworth and J. M. Rusin. 1982. In Situ Vitrification: Application Analysis for Stabilization of Transuranic Waste. PNL-4442.

Pederson, L. R. 1982. "Are Neutral Sodium Atoms Produced on Glass Surfaces by Electron Bombardment?" Surface Science, 119:L307-L313.

Pederson, L. R., C. Q. Buckwalter, G. L. McVay and B. L. Riddle. "Glass Surface Area to Solution Volume Ratio and Its

Implications to Accelerated Leach Testing." In Scientific Basis for Nuclear Waste Management VI, D. G. Brookins, ed., pp. 47-54. Proceedings of the Materials Research Society Symposium held November 1982 in Boston, Massachusetts. Elsevier Science Publishing Co., New York, New York.

Perez, J. M., Jr. 1983. Design and Performance of Feed Delivery Systems for Simulated Radioactive Waste Slurries. PNL-4423.

Pitman, S. G. 1982. Investigation of Environmentally-Assisted Fracture of Metallic Nuclear Waste Package Structural Barrier Materials in a Simulated Basalt Repository Environment. PNL-4379.

Rasmussen, D. E. et al. 1982. Comparison of Cask and Drywell Storage Concepts for a Monitored Retrievable Storage System. PNL-4450.

Relyea, J. F. 1983. A Decision Tree Approach to Evaluating Inactive Uranium Processing Sites for Liner Requirements. PNL-4436.

Relyea, J. F. and W. J. Martin. "Evaluation of Inactive Uranium Mill Tailings Sites for Liner Requirements: Characterization and Interaction of Tailings, Soil and Liner Materials." Uranium Mill Tailings Management, pp. 507-520. Proceedings of the Fifth Symposium on Uranium Mill Tailings, December 9-10, 1982, Colorado State University, Fort Collins, Colorado.

Ross, W. A., J. H. Westsik, Jr., F. P. Roberts and C. O. Harvey. "Comparison of the Leachability of Three TRU Cement Waste Forms." In Scientific Basis for Nuclear Waste Management VI, D. G. Brookins, ed., pp. 175-182. Proceedings of the Materials Research Society Symposium held November 1982 in Boston, Massachusetts. Elsevier Science Publishing Co., New York, New York.

Scheele, R. D. and L. L. Burger. 1982. Selection of a Carbon-14 Fixation Form. PNL-4447.

Shade, J. W. 1982. "Release Kinetics and Alteration of Waste Glass in Crystalline Rock Systems." Chemical Geology, 36(1-2):103-121.

Smith, R. I. and J. F. Nesbitt. 1982. MRS/IS Facility Co-Located with a Repository: Preconceptual Design and Life Cycle Cost Estimates. PNL-4625.

Strachan, D. M. "Results from a One-Year Leach Test: Long-Term Use of MCC-1." In Scientific Basis for Nuclear Waste Management V, W. Lutze, ed., pp. 181-192. Proceedings of the Materials Research Society Fifth International Symposium on the Scientific Basis for Nuclear Waste Management, held June 7-10, 1982, in Berlin, Germany. Elsevier Science Publishing Co., New York, New York.

Templeton, W. L. 1983. "Lessons from Radioactive Waste Disposal Applied to Other Pollutants." Chapter 20, Volume 3 of Radioactive Wastes and the Ocean, P. Kilho Park et al., eds. John Wiley and Sons, New York, New York.

Timmerman, C. L. "Economics of Transuranic Waste Immobilization and Disposal." In The Treatment and Handling of Radioactive Wastes, A. G. Blasewitz, J. M. Davis and M. R. Smith, eds., pp. 19-29. Battelle Press, Columbus and Richland.

Treat, R. L., K. H. Oma and S. C. Slate. "High-Level Waste Forms: Glass Marbles and Thermal Spray Coatings." In The Treatment and Handling of Radioactive Wastes, A. G. Blasewitz, J. M. Davis and M. R. Smith, eds., pp. 331-335. Battelle Press, Columbus and Richland.

Turcotte, R. P., J. W. Wald, F. P. Roberts and J. M. Rusin. 1982. "Radiation Damage in Nuclear Waste Ceramics." J. Am. Cer. Soc., 65(12):589-593.

Wald, J. W. and P. Offermann. "A Study of Radiation Effects on Curium-Doped $Gd_2Ti_2O_7$ (pyrochlore) and $CaZrTi_2O_7$ (zirconolite)." In Scientific Basis for Nuclear Waste Management V, W. Lutze, ed., pp. 369-378. Proceedings of the Materials Research Society Fifth International Symposium on the Scientific Basis for Nuclear Waste Management, held June 7-10, 1982, in Berlin, Germany. Elsevier Science Publishing Co., New York, New York.

Wald, J. W., J. W. Shade, R. O. Lokken and J. M. Rusin. 1982. "A Comparative Evaluation of Selected Alternative Nuclear Waste Forms." Radioactive Waste Management and the Nuclear Fuel Cycle, 3(2):215-239.

Weber, W. J. "A Review of the Current Status of Radiation Effects in Solid Nuclear Waste Forms." In Scientific Basis for Nuclear Waste Management VI, D. G. Brookins, ed., pp. 407-414. Proceedings of the Materials Research Society Symposium held November 1982 in Boston, Massachusetts. Elsevier Science Publishing Co., New York, New York.

Weber, W. J. and F. P. Roberts. "A Review of Radiation Effects in Solid Nuclear Waste Forms." Nuclear Technology, 60:178-198.

Westerman, R. E., S. G. Pitman and J. L. Nelson. 1982. General Corrosion, Irradiation Corrosion and Environmental-Mechanical Evaluation of Nuclear Waste Package Structural Barrier Materials. PNL-4364.

Westsik, J. H., Jr., C. O. Harvey, F. P. Roberts, W. A. Ross and R. E. Thornhill. "Comparison of 200 Liter and 40 Milliliter Leach Tests." In Scientific Basis for Nuclear Waste Management VI, D. G. Brookins, ed., pp. 243-250. Proceedings of the Materials Research Society Symposium held November 1982 in Boston, Massachusetts. Elsevier Science Publishing Co., New York, New York.

DISTRIBUTION

No. of Copies

OFFSITE

27 DOE Technical Information Center

2 Nuclear Regulatory Commission
7915 Eastern Avenue
Silver Springs, MD 20910
ATTN: R. E. Cunningham
J. Roberts

R. D. Smith
Division of Waste Management
Nuclear Regulatory Commission
Washington, DC 20555

D. M. Rohrer
Office of Inspection and Enforcement
Nuclear Regulatory Commission
MS359 EW/W
Washington, DC 20555

Materials Section Leader
High-Level Waste Licensing Branch
Nuclear Regulatory Commission
Washington, DC 20555

W. F. Holcomb
Environmental Protection Agency
Office of Radiation Programs
Washington, DC 20460

W. E. Mott
DOE Division of Environmental Control Technology
Washington, DC 20545

4 DOE Terminal Waste Disposal and Remedial Action
GTN
Washington, DC 20545
ATTN: J. E. Baublitz, NE-24
F. E. Coffman, NE-20
J. R. Coleman, NE-25
D. J. McGoff, NE-23

3 DOE Office of Defense Waste and Byproducts Management
GTN
Washington, DC 20545
ATTN: J. E. Dieckhoner
J. J. Jicha
G. K. Oertel

No. of Copies

J. W. Bennett
Geologic Repository Division
DOE Nuclear Waste Policy Act Project Office
S-10, Forrestal
Washington, DC 20585

3 Geologic Repository Division
DOE Nuclear Waste Policy Act Project Office
GTN
Washington, DC 20545
ATTN: C. R. Cooley, S-10/NE-22
C. H. George, S-10/NE-22
R. Stein

W. W. Ballard, Jr.
DOE Office of Spent Fuel Management and Reprocessing Systems
NE-40, GTN
Washington, DC 20545

M. J. Lawrence
Storage Projects
DOE Nuclear Waste Policy Act Project Office
S-10, Forrestal
Washington, DC 20585

S. A. Mann
DOE Chicago Operations Office
9800 South Cass Avenue
Argonne, IL 60439

J. O. Neff
DOE National Waste Program Office
505 King Avenue
Columbus, OH 43201

J. D. Hamric
DOE Idaho Operations Office
550 2nd Street
Idaho Falls, ID 83401

Office of the Assistant Manager for Energy Research and Development
DOE Oak Ridge Operations Office
P.O. Box E
Oak Ridge, TN 37830

3 Oak Ridge National Laboratory
P.O. Box Y
Oak Ridge, TN 37830
ATTN: W. D. Burch
L. J. Mezga
T. H. Row

No. of
Copies

D. T. Oakley
Los Alamos Scientific Laboratory
P.O. Box 1663
Los Alamos, NM 87544

3 DOE Savannah River Operations Office
P.O. Box A
Aiken, SC 29801
ATTN: H. L. Brandt
E. S. Goldberg
T. B. Hindman

2 Exxon Nuclear Idaho
P.O. Box 2800
Idaho Falls, ID 83401
ATTN: D. L. Condotta
File Copy

G. B. Levin
EG&G Idaho
P.O. Box 1625
Idaho Falls, ID 83415

S. G. Harbinson
DOE San Francisco Operations
1333 Broadway
Oakland, CA 94612

R. Y. Lowrey
DOE Albuquerque Operations Office
P.O. Box 5400
Albuquerque, NM 87185

2 Allied-General Nuclear Services
P.O. Box 847
Barnwell, SC 29812
ATTN: J. A. Buckham
A. Williams

2 Argonne National Laboratory
9700 South Cass Avenue
Argonne, IL 60439
ATTN: J. H. Kittel
M. J. Steindler/L. E. Trevorrow

7 Battelle Memorial Institute
Office of Nuclear Waste Isolation
505 King Avenue
Columbus, OH 43201
ATTN: S. H. Basham
W. Carbiener
N. E. Carter
S. Goldsmith
J. F. Kircher
W. J. Madia
B. Rawles

No. of
Copies

K. V. Gilbert
Rockwell International
Rocky Flats Plant
P.O. Box 464
Golden, CO 80401

3 E. I. du Pont de Nemours Company
Savannah River Laboratory
Aiken, SC 29808
ATTN: J. L. Crandall
E. J. Hennelly
S. Mirshak

R. Williams
Electric Power Research Institute
3412 Hillview Avenue
P.O. Box 10412
Palo Alto, CA 94304

R. K. Brown
Westinghouse Electric Corporation
Advanced Energy Systems Div.
WIPP Library
P.O. Box 40039
Albuquerque, NM 87196

J. L. Knabenschuh
Nuclear Fuels Services, Inc.
P.O. Box 191
West Valley, NY 14171

2 Bechtel National, Inc.
Box 3965
San Francisco, CA 94119
ATTN: J. L. Jardine
W. A. Freeby

J. L. Larocca, Chairman
Energy Research and Development
Authority
Empire State Plaza
Albany, NY 12223

2 Sandia Laboratories
Albuquerque, NM 87185
ATTN: D. R. Anderson
W. Weart

J. W. Bartlett
The Analytic Sciences Corp.
6 Jacob Way
Reading, MA 01867

R. G. Post
College of Engineering
University of Arizona
Tucson, AZ 85721

No. of
Copies

L. L. Hench
Dept. of Materials Science and Engineering
University of Florida
Gainesville, FL 32611

F. K. Pittman
3508 Sagecrest Terrace
Ft. Worth, TX 76109

ONSITE

9 DOE Richland Operations Office

E. A. Bracken
P. F. X. Dunigan, Jr.
O. J. Elgert
R. D. Izatt
O. L. Olson
H. E. Ransom
J. J. Schreiber
M. W. Shupe
J. D. White

9 Rockwell Hanford Operations

R. A. Deju
K. A. Gasper
R. J. Gimera
H. E. McGuire
J. W. Patterson
J. H. Roecker
M. J. Smith
D. D. Wodrich
File Copy

UNC United Nuclear Industries

T. E. Dabrowski

Westinghouse Hanford Company

A. G. Blasewitz

No. of
Copies

48 Pacific Northwest Laboratory

W. F. Bonner
D. J. Bradley
R. L. Brodzinski
L. L. Burger/R. D. Scheele/L. A. Bray
T. D. Chikalla
L. L. Clark
R. A. Craig/C. R. Cole
R. L. Dillon
D. W. Dragnich
V. F. FitzPatrick/R. A. Brouns/J. L. Buelt
R. M. Fleischman
W. A. Glass
C. R. Hann
J. N. Hartley/H. D. Freeman
J. H. Jarrett
T. L. Jones/G. W. Gee
M. R. Kreiter
L. T. Lakey
J. M. Latkovich
R. C. Liikala
R. P. Marshall/W. R. Wiley/D. B. Cearlock
J. L. McElroy
R. W. McKee
J. E. Mendel/M. D. Merz
J. E. Minor/D. H. Siemens
K. S. Murthy/R. F. McCallum
I. C. Nelson/J. G. Stephan
R. E. Nightingale
D. E. Olesen
A. M. Platt
J. A. Powell
J. V. Robinson
J. L. Swanson/E. J. Wheelwright
G. L. Tingey/G. A. Jensen
R. P. Turcotte/G. L. McVay
C. M. Unruh/E. C. Watson
H. H. Van Tuyl
B. E. Vaughan
R. E. Westerman
R. D. Widrig
L. D. Williams
Technical Information (5)
Publishing Coordination (2)