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**Nuclear Waste Management
Semiannual Progress Report
April 1984 Through
September 1984**

**J. L. McElroy
J. A. Powell**

December 1984

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

**Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
by Battelle Memorial Institute**

PNL-4250-6

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PACIFIC NORTHWEST LABORATORY
operated by
BATTELLE
for the
UNITED STATES DEPARTMENT OF ENERGY
under Contract DE-AC06-76RLO 1830

Printed in the United States of America
Available from
National Technical Information Service
United States Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22161

NTIS Price Codes
Microfiche A01

Printed Copy

Pages	Price Codes
001-025	A02
026-050	A03
051-075	A04
076-100	A05
101-125	A06
126-150	A07
151-175	A08
176-200	A09
201-225	A010
226-250	A011
251-275	A012
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NUCLEAR WASTE MANAGEMENT
SEMIANNUAL PROGRESS REPORT
APRIL 1984 THROUGH SEPTEMBER 1984

Compiled by
J. L. McElroy and J. A. Powell
Nuclear Waste Technology Program

December 1984

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

Pacific Northwest Laboratory
Richland, Washington 99352

MASTER

FOREWORD

This document, PNL-4250-6, is one of a series of technical progress reports designed to report semiannually on radioactive waste management programs at the Pacific Northwest Laboratory (PNL). These programs are funded by the Department of Energy's Offices of Nuclear Energy and Defense Programs. Only programs that have significant, reportable results are included in this document.

Technical progress of the Nuclear Waste Treatment Program is reported in an annual report instead of this document.

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1.0 DEFENSE WASTE TECHNOLOGY

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

1.0 DEFENSE WASTE TECHNOLOGY

J. M. Latkovich and C. A. Geffen - Program Office Managers

1.1 HANFORD WASTE VITRIFICATION

(AR-05-10-02 - 80602)

J. E. Minor - Program Manager

The objective of the Hanford Waste Vitrification (HWV) Program is to apply vitrification technology to a selected Hanford waste stream(s), developing a process and equipment to be utilized in a Hanford waste facility. The HWV Program consists of two parts: Hanford Waste Vitrification Plant (HWVP) support and Waste Stream Characterization and Immobilization (WSCI) support. The objective of the WSCI support is the development of technology applicable to the immobilization of high-level waste from the Hanford Site. It will be based on a high-priority waste stream defined in the Hanford Waste Management Plan and by Rockwell Hanford Operations (Rockwell).

Summary

The Hanford Waste Vitrification Plant (HWVP) has the mission of vitrifying those components of all Hanford liquid high-level wastes that require terminal geologic disposal. The HWVP will vitrify pretreated neutralized current acid waste (NCAW). Future process development will center on NCAW from PUREX and other pretreated streams that are identified as requiring geologic disposal.

Remotized component development has included the welding of prototype canister closures; design, installation, and testing of a melter viewing system; jet pump testing for the bulk slurry feed transfer system; evaluation of electrical and instrument connectors; analysis of discharge section thermal mockup test data; and tests performed on the melter nozzle cleaning system.

WSCI generic process development activities during this period have included melter feed rheology studies; melter feed system development; studies of glass foaming; and effluent control system studies.

To provide greater continuity and clarity for the reader, this report combines the results of work in two programs.

Process Development - M. S. Hanson, S. O. Bates, L. A. Chick, D. S. Goldman, H. T. Blair, M. E. Peterson, and J. M. Perez

Waste Glass Development

During the development of HWVP waste glasses, it was observed that the levels of chromium found in the NCAW cause crystal phases to form in the glass. These crystals can cause melt viscosities to become unstable at constant temperatures. The type of crystal phase formed, which affects the degree of the viscosity instability, depends strongly on the thermal history of the glass. These crystals may adversely affect melter performance by causing viscosity instabilities and sludge formation. The full extent of the effects of crystallinity on melter performance is not now known.

Initial development of borosilicate glasses using NCAW showed that these glasses exhibited crystallinity and that their viscosities varied with time when held at constant temperatures. Further testing led to the development of a boron-free calcia silicate glass (HW15, Table 1.1) with stable

TABLE 1.1. Wastes and Glasses Used in HWVP Glass Development

Oxides	HW15 Calcium Glass		HW21 Borosilicate Glass		SRL TDS Frit 165	
	Waste	Glass	Waste	Glass	Waste	Glass
SiO ₂	2.5	50.9	2.5	51.6	11.0	51.4
Na ₂ O	8.4	13.4	8.4	13.4	6.3	11.0
Fe ₂ O ₃	46.3	11.6	46.3	11.6	36.6	10.5
B ₂ O ₃		0.0		7.5		7.1
Li ₂ O		3.8		4.5		5.0
Al ₂ O ₃	17.3	4.3	17.3	4.3	16.2	4.7
Cr ₂ O ₃	4.0	1.0	4.0	1.0	0.5	0.1
MgO	0.5	0.9	0.5	0.9	0.4	0.8
Nd ₂ O ₃	3.4	0.8	3.4	0.8	0.9	0.1
ZrO ₂	3.1	0.8	3.1	0.8	1.2	0.7
MoO ₃	2.4	0.6	2.4	0.6		
CeO ₂	2.0	0.5	2.0	0.5	0.5	0.1
NiO	1.9	0.5	1.9	0.5	2.9	0.9
Cs ₂ O	1.7	0.4	1.7	0.4		
C	1.5	0.4	1.5	0.4		
La ₂ O ₃	1.3	0.3	1.3	0.3	2.0	0.3
SO ₄	1.2	0.3	1.2	0.3	1.0	0.3
MnO ₂	0.9	0.2	0.9	0.2	10.8	3.1
SrO	0.7	0.2	0.7	0.2	0.1	0.0
CaO	0.5	9.1	0.5	0.1	3.3	1.0
CuO	0.5	0.1	0.5	0.1	0.2	
BaO					0.3	0.1
P ₂ O ₅					0.1	
RE ₂ O ₃					4.5	2.3
RuO ₂					0.3	
ZnO					0.9	0.3
TOTAL	99.9	100.0	99.9	100.0	100.0	99.8

viscosities and acceptable crystallinity at 25% waste loading. This glass was used in the WG-2 melter experiment.

Because this was the first time unstable viscosities were observed and since calcia silicate glasses are new to the nuclear waste field, other previously used borosilicate glasses were reexamined. This reexamination showed the previous existence of unstable viscosities and crystallinity, detected only now through improved equipment and techniques. This reexamination indicated also that the time-dependent viscosities were caused by the crystallinity and thermal history of the glasses.

Crystallinity was found to play a major role in the stability of the viscosity. In all glasses where crystals were not observed, the viscosities were stable. In those glasses with crystals, the degree of instability of the viscosities was affected by the type and size of crystals present. Optical evaluation of thin sections and fibers of glass removed during the viscosity tests showed two types of crystals to be present: small, dark cubic crystals, and large, red hexagonal platelet crystals. The hexagonal crystals produce much greater instability in the viscosity than do the cubic crystals.

SEM/EDX elemental analysis was conducted on NCAW borosilicate glasses containing both

cubic and hexagonal crystals. The glass matrix was found to have no detectable levels of chromium, implying that virtually all the chromium resides in the crystals. The analysis showed the cubic crystals to be primarily chromium, with significant amounts of iron and some nickel. Because of the cubic form and the elemental analysis, these cubic crystals are thought to be chromite spinel (FeCr_2O_4) with some nickel and iron substitution. The hexagonal crystals are primarily chromium with some iron but little or no nickel. It is assumed that the hexagonal platelets are a solid solution of chromic oxide and hematite $(\text{Cr,Fe})_2\text{O}_3$. The lack of sites for Ni^{+2} substitution into $(\text{Cr,Fe})_2\text{O}_3$ would explain the reduced nickel.

Reducing the concentration of chromium in the glass decreased the amount of crystals formed. The Cr_2O_3 was removed and replaced with Fe_2O_3 in a Hanford borosilicate glass (HW21) and a Savannah River type of borosilicate glass (SRL 165, Table 1.1). The Cr_2O_3 was then added back into the glasses at levels from 0 to 2 wt%. Following a standard heat treatment, no crystal formation was observed in thin sections of either of the glasses from 0.0 to 0.25 wt% Cr_2O_3 . At 0.5 wt% and above, crystals were evident in both glasses. The Cr_2O_3 concentration in the NCAW waste is about 4.0 to 5.5 wt% (1 to 1.4 wt% in the glass). This can be compared to SRL TDS waste where the Cr_2O_3 concentration is 0.54 wt% in the waste (0.15 wt% in the glass) and no crystal formation was observed. In West Valley glasses with a Cr_2O_3 level of 0.7 wt% in the glass, both cubic spinel and hematite crystals were observed.

Crystallinity can be reduced also by lowering the waste loading in the glass. The crystallinity in glass HW21 was compared at waste loadings of 18, 20, and 25%. At 18% waste loading, no crystals were observed and

the glass had stable viscosities. At 20% waste loading, the glass had some hexagonal platelets and unstable viscosities. At 25% waste loading, the glass contained a large amount of cubic spinel crystals and viscosities were slightly unstable. At 18% waste loading, the Cr_2O_3 concentration in the glass was 0.7 wt%, and in the range where significant crystallinity had been observed. The lack of crystals in the glass indicates that the solubility of the Cr_2O_3 is increased by the reduced amounts of other waste components such as iron.

The thermal history of the glass also was found to be a major factor in crystal formation and thus also to affect the stability of the viscosities. Previous testing of borosilicate glasses suggested that hexagonal platelets were dominant at higher melt temperatures ($>1250^\circ\text{C}$), while the cubic spinel crystals were predominant at lower melt temperatures ($<1200^\circ\text{C}$). Testing was conducted to determine the maximum temperature to which a borosilicate glass (HW21) could be taken without causing hexagonal crystals to form. Glass HW21 was heat-treated with maximum melt temperatures ranging from 1150°C to 1350°C for 1/2 hr. When the glass was heat-treated with maximum temperatures $>1250^\circ\text{C}$, the growth of hexagonal crystals was rapid during subsequent cooling. These hexagonal crystals settled rapidly to the bottom of the melt. This settling, along with the large surface area of the crystals, may account for the unstable viscosities measured. As the crystals settled past the viscometer spindle, the viscosity of the melt would change as the crystal density varied. When maximum melt temperatures were 1200°C or less, only the smaller cubic spinel crystals were present. The cubic crystals were distributed fairly uniformly throughout the melt. Therefore,

when maximum melt temperatures were maintained at 1200°C or less, the viscosities of the borosilicate glasses made with NCAW were stable.

Initial testing of a borosilicate glass using an updated NCAW/CRW (cladding removal waste) blend found the glass viscosity to be only slightly unstable, but a large degree of crystallinity was observed. Initial testing of a calcium silicate glass, also using the updated NCAW/CRW, showed even greater crystallinity than seen in the borosilicate glass, and the viscosity was very unstable. It is assumed that use of the NCAW/CRW worked so poorly in the calcia silicate glass as compared with previous NCAW because the Cr_2O_3 solubility of the calcia silicate glass was being exceeded. This increase in crystallinity is not due to the addition of CRW to the NCAW, but rather to the revised upward estimate of the Cr_2O_3 and Al_2O_3 concentrations in the updated NCAW.

Since it may not be possible to reduce the amount of crystallinity in the waste glass, the effects of crystallinity on melter performance need to be determined. If crystals settle to the bottom of the melter, a sludge could form, possibly reducing the life of the melter or affecting its performance. The rate at which the crystals settle in the melt versus temperature is being evaluated. By using settling velocities along with convection velocities taken from computer modeling of the melter, it can be determined whether settling of the crystals is likely to be a major problem in the melter. Crucibles of an NCAW borosilicate glass (HW21) were given a standard heat treatment to uniformly produce cubic crystals throughout the melt. These crucibles of glass were then held at temperatures of 950, 1050, 1125, and 1200°C for times ranging from 1/2 hr to 169 hr. The crucibles were thin-sectioned and the crystal

settling was measured. Rough preliminary analysis indicates the settling velocity of 8- μm cubic crystals is $<10^{-5}$ cm/sec. Rough estimates from the modeling indicate convection velocities on the order of 5×10^{-2} cm/sec. This suggests that cubic crystals of this size would be swept along in the convection current and not settle to the bottom. Further testing is needed to confirm these estimates. A major factor in the settling velocity is the size of the particles. The settling velocity varies approximately as the square of the particle size, and further melter testing is needed to determine the size range of the crystals produced in the melter.

The redox state of the glasses tested was reducing because the amount of carbon found in the waste was added to the mixed oxides used in blending the glasses. This resulted in the glass being far more reduced than would be necessary to control foaming in the melter. The redox state of the melter glass will not be as reduced because the total organic carbon in the waste stream will be consumed, reducing other compounds in the waste. West Valley Support Program studies have indicated that the foaming tendency of the glass in the melter can be decreased to acceptable levels with carbon additions to reduce the glass redox state to a level corresponding to a $\text{Fe}^{+2}/\text{Fe}^{+3}$ ratio of about 0.2. Other studies have indicated that as the glass becomes more oxidizing, the tendency to form hexagonal crystals is much stronger.

Melter Feed Rheology Studies

Neutralized current acid waste was the base waste used for feed rheology improvement studies in this period. Two different NCAW formulations and two different host glass

compositions were studied. Both wastes were derived from the same fuel, but because of different proposed pretreatment processes, the two had different Na and NO_3^- contents. This required the use of different compounds in preparing simulations of the candidate wastes.

The host glass compositions were designated 82-10 and 84-09. The oxide compositions of the formers as a frit glass and as unreacted chemicals added to the simulated waste are presented in Table 1.2. The SiO_2 was added as either -325 mesh ground powder or a 50 wt% colloidal suspension when the unreacted glass formers were used.

The simulated melter feeds were prepared by combining one part (by weight) waste oxide equivalents with three parts glass-former oxide equivalents. The feeds were prepared at concentrations of 300, 400, and 500 g oxide equivalents per liter. The prepared

slurries were compared on the basis of viscosity, the volume percent of settled solids, and the sediment's resistance to resuspension. The general conclusions are:

- Feeds made with simulated NCAW and unreacted nitrate glass formers have the most desirable rheological properties.
- When nitrate glass formers are used, melter feed concentrated to 400 g total oxide equivalent per liter is possible.
- Feeds made with carbonate glass formers tend more to deposit on surfaces at air interfaces and are more difficult to resuspend than nitrate glass formers, but are more viscous and exhibit less settling of solids.
- Feeds made with hydroxide glass formers form sediments more difficult to suspend than those formed by either the carbonates or nitrates.

TABLE 1.2. Compositions and Unreacted Chemical Equivalents Used To Modify Melter Feed Rheology

82-10 Frit		82-10 Nitrate Equivalents		82-10 Hydroxide Equivalents	
Compound	wt%	Compound	wt%	Compound	wt%
SiO_2	61.7	SiO_2	37.0	SiO_2	45.5
B_2O_3	15.1	$\text{Na}_2\text{B}_4\text{O}_7 \cdot 10\text{H}_2\text{O}$	24.8	$\text{Na}_2\text{B}_4\text{O}_7 \cdot 10\text{H}_2\text{O}$	30.5
Na_2O	14.7	NaNO_3	13.1	NaOH	14.0
Li_2O	7.4	LiNO_3	21.2	LiOH	8.8
MgO	1.1	$\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$	3.9	$\text{Mg}(\text{OH})_2$	1.2

84-09 Frit		84-09 Nitrate Equivalents		84-09 Carbonate Equivalents	
Compound	wt%	Compound	wt%	Compound	wt%
SiO_2	67.0	SiO_2	35.6	SiO_2	52.0
Na_2O	15.0	NaNO_3	21.8	Na_2CO_3	19.9
CaO	12.0	$\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$	27.1	CaCO_3	16.6
Li_2O	5.0	LiNO_3	12.3	Li_2CO_3	9.6
MgO	1.0	$\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$	3.2	$\text{MgCO}_3 \cdot \text{Mg}(\text{OH})_2 \cdot 3\text{H}_2\text{O}$	1.9

- The use of colloidal silica instead of ground silica with nitrate glass formers increases the feed viscosity and settling resistance but does not gel the feed.
- The use of colloidal silica does not improve the properties of feeds made with hydroxide glass formers.
- There is very little difference in the rheology of feeds made with the 82-10 and 84-09 compositions when the nitrate compounds are used.

Also studied during this period were the effects of adding an organic thickener to feeds made with simulated NCAW and ground frit. The thickener was a xanthan gum. The feed concentration used in the testing was 400 g total oxide equivalents per liter. Three levels of gum were evaluated: 0.15, 0.30, and 0.50 wt% of the total oxide loading in the feed. The xanthan gum was effective at the 0.15 wt% level in preventing solids from settling into a sediment difficult to resuspend. Further increases in the gum concentration increased the feed viscosity but did not improve the settling resistance.

Melter Feed System Development

A second-generation air-displacement slurry pump with an activated two-way valve was built and tested for a total of 1167 hr on a feed made with simulated NCAW, ground silica, and nitrate glass-forming compounds. The mechanical parts of the pump were designed to include valve guides for alignment of the seats and a single-valve drive rod as shown in Figure 1.1. Critical wear parts were fabricated from special wear-resistant alloys. A microprocessor controller was custom-built to operate the various solenoid valves and a variable air pressure regulator used to effect the steps in a cycle of the

pump. The controller provided flexibility in varying the operating parameters of the pump. An operating range of 10 to 185 L/hr with a precision of 2% for rates over 15 L/hr was demonstrated using the controller and variable air-pressure regulator. Wear and corrosion during the 1167 hr of operation were negligible. Longer periods of operation will be required before the life of the pump can be predicted from measurements of wear and corrosion.

The second-generation pump was also used on two occasions to support melter demonstrations. During the first, the pump was used to feed the high-bay ceramic melter for 33 hr at an average rate of 13 L/hr. Some drying of feed in the feed nozzle was experienced because each cycle of the pump included a long pause when feed was not flowing to maintain the low flow rate. These pauses were eliminated by adjusting the air-pressure profile during the discharge step in the pump cycle.

During the second melter demonstration, the pump was used to charge 1364 L of a simulated West Valley feed composition, which included nitric acid and ground zeolite, to the pilot-scale ceramic melter (PSCM). Initially, the pump did not function properly, and it was determined that the relative elevations of the feed tank and the melter and the negative pressure in the melter were combining to create a siphon. After a siphon break was installed at the apex of the feed line, the pump operated satisfactorily for 45 hr at an average rate of 30 L/hr. The pump output rate was determined to vary about 15% with inlet head changes of 50 in. to 20 in. It was demonstrated that this can be compensated for by adjusting the pump control parameters.

FIGURE 1.1. Second-Generation Air-Displacement Slurry Pump

Melter Tests

A 50-hr continuous run in the high bay ceramic melter was completed using a feed made with simulated NCAW and host glass 84-09 made from nitrates and ground silica. A total of 550 L of feed were converted to 215 kg of glass. The average feed rate was 11 L/hr. A stable cold cap, covering 70% of the melt surface, was achieved at a feed rate of 15 L/hr. The melter was operating at an average main electrode power of 19 kW. The average melt temperature was 1125°C, and the highest melt temperature recorded during the test was 1200°C. A 100% cold cap coverage resulted whenever the feed rate was increased above 15 L/h. The cold cap was flexible and remained at a thickness of less than 2 in. during the test.

Melt foaming did occur during the test with this waste and glass-former composition. The first occurrence, at 17 hr into the run, was probably due to overfeeding.

During the final 10 hr of the test, foam accumulation was a problem and required lowering the feed rate to maintain stable melter operation. With a cold-cap coverage >80%, a 3- to 5-in. foam layer accumulated beneath the cold cap. At nearly full coverage, the foam rose to the top of the electrodes, about 5 in. above the normal melt level. This required shutting off the electrode power which caused the foam to collapse.

The conclusion drawn from this run was that the simulated NCAW and 84-09 glass composition had melting characteristics similar to those of previous Hanford waste and host-glass compositions tested.

Melter Modeling. The modeling of the PSCM was completed, using the TEMPEST code with the data from the PSCM-9 melter test to verify the predictions of the TEMPEST code

for an operating melter. The model predicted voltages roughly 2.2% above the measured value and temperatures ranging from 1.7 to 5.1% higher than those measured. The high temperatures were expected due to overprediction of the voltage for a constant current input since more joule heat is being generated. Therefore, the predicted melter parameters are considered to be in agreement with the measured value.

Melter Instrumentation. Several different instruments were tested in the pilot-scale ceramic melter. One uses the principle of time-domain reflectometry to detect interfaces between materials of different densities. A probe, made of two high-nickel alloy rods, extends from a port in the top of the melter chamber into the melt. An electronics package generates a high-speed electronic pulse in one of the rods and measures the time required for it to return through the other rod. The time is primarily a function of the distance traveled by the pulse and the dielectric constant of the material between the rods. The device was able to detect changes in the melt level on the order of 1/2 in. and the presence of a second phase on the surface of the melt. However, when liquid feed was charged to the melter, the device was observed to respond to the cold cap building up in the melter rather than to the melt level. This is due partially to the design of the probe and its location in the melter chamber. Other probe designs are being considered.

Another instrument tested used the pneumatic-bubbler technique to determine liquid heads and specific gravities. A single high-nickel alloy rod had three holes bored in it lengthwise. The holes were ported at the 1-in., 8-in. and 20-in. levels with respect to the floor of the melter chamber. Air

pressure at the lowest port gave an indication of the weight factor while the differential pressure between the 1-in. and 8-in. ports was used to determine the specific gravity. The 20-in. port served as a plenum pressure reference and a foam detector. The device worked well for measuring the melt level and could be used to predict the melt level to an accuracy of <0.5 in. The foam detector signaled when the cold cap got excessively high.

System design and software development is in progress to allow a Hewlett-Packard (HP) 9836C computer and an HP-3497A acquisition system to perform data logging and process control of glass-melter tests. This system will log data on a 14.5-megabyte hard disk. After completion of a test, data files can be written to cartridge tape for long-term data storage. The acquisition and control system will be user-configurable to accept analog and digital inputs and outputs. The user can define signal names, pseudo channels, and complex control strategies, as well as set alarm limits. This is done through the use of menu-driven displays on the HP-9836C computer system. During tests the system will report signals that have exceeded limits, record data, and allow the operator to list or plot test data of his choice.

Software development is in progress for the scanning of the inputs and the writing of outputs. Scanning and storage in the HP-9836C memory of analog inputs/outputs and digital inputs/outputs is functional. Several of the display formats have been implemented, including data lists, data printing, alarm scanning, and alarm display.

A status-list printing routine is under development. This mode allows the user to get a printed list of the channel names defined in the status-list file. The user

will be able to maintain up to ten files on the system, and these files will contain lists of the status channels to be scanned.

The design of the menu-driven user interface was completed. At startup, the user will be shown the main menu for the system, which will provide access paths for any tasks that need to be performed.

A forms package was designed to allow easy configuration of the system. Software development of the forms package is 80% complete, with many of the menus fully functional. The package is designed to allow additional help screens to aid the user. The system configuration software design and data storage, both in HP-9836C memory and on disk, is 90% complete.

Equipment Remotizing and Testing -

D. H. Siemens, D. E. Larson, M. M. Beary, W. O. Heath, and R. F. Klein

Canister Closure Welding

The objective of this activity is to provide the proof concept, preconceptual design, and detailed design of the welding machine and to procure, install, and qualify it. The inertia welding process is the reference process for welding the closures on the HWVP canisters. A schematic of the reference welder arrangement is shown in Figure 1.2.

Canister top flanges and primary and secondary lids were procured and shipped to the welding machine manufacturer, where 14 proof-of-concept welds were made. These proof-of-concept welds consisted of 15 primary lids welded to canister top flanges and four secondary lids welded over previously welded primary lids. The welded parts were returned to PNL for examination and testing.

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FIGURE 1.2. Inertia Welder for HWVP Canisters

Testing of the proof-of-concept welds will consist of some welds being examined for leaks by the helium leak detection method, ultrasonic nondestructive examination, several tensile specimens, corrosion samples, metallography, and canister impact tests. During this report period, all tests have been started. Some welds have been helium

leak checked, some metallography has been performed, tensile samples have been run, ultrasonic examination has been started, and corrosion and impact tests have begun.

Some concern was expressed about the amount of weld upset curl. It was felt that radioactive particles could be trapped in the upset curl and not be removed by canister

decontamination. A program was established to reduce the amount of upset curl and was successfully completed. The weld upset no longer curls but has a clean, smooth, easily decontaminated profile with no reduction in weld quality.

Design of the in-cell canister clamping mechanism began in the latter part of this report period.

Canister Welding Positioning Device

Canister welding and weld inspection are sequential operations involving the use of a positioning device (swing arm) for moving the filled canister into position, for the welding machine to weld the lid onto the canister, and then reposition the sealed canister for weld inspection. The canister is transferred by overhead crane from the turntable to a receptacle or "thimble" attached to an arm restrained at one end by a pivot but free at the other end to be driven in an arc by a motor. The thimble has wedges in the bottom to center the canister for the weld. An electric-motor drive mechanism, under the direct control of an operator, moves the swing arm with the canister on a rail in an arc and under the welder (Figure 1.2). The entire operation of the swing arm occurs in the heavily shielded vitrification cell.

A prototypical swing arm for testing in the Engineering and Development Laboratory of the 324 Building has been fabricated and installed. The drive motor, gear reducer, right-angle drive, and drive wheel of the drive mechanism are attached with four remote nuts that can be removed with an impact wrench. The loosened drive mechanism can then be lifted from the swing arm with a crane.

Another relatively vulnerable component is the spherical thrust bearing, which carries the load at the pivot point of the swing arm. This bearing also is designed for replacement by the cell crane. The bearing is mounted in a plate with four remote nuts. A centering cone, as part of the plate, guides the bearing into place while the plate is lowered into position on the pivot pin by the overhead crane. The spherical thrust bearing will tolerate some misalignment with the pivot pin, all within the precision required by the welder to weld the lid onto the canister.

Should the swing arm fail to operate while the canister is under the welding head, the arm can be moved to a position accessible by the crane with a light cable that is attached to the arm. The cable with bail, when picked up and raised by the crane hook, provides a horizontal travel to the swing arm, which moves it out from under the welding head. The drive wheel is smooth, not a cog; thus a locked drive wheel will slide along the track during retrieval.

Tests are planned to verify that the device is operable remotely and that the swing arm will locate the canister with the precision specified by the inertia welding manufacturer.

Canister Closure Weld Inspection

The primary objective of the work during this period was to build and demonstrate an engineering model of the canister closure weld inspection system. This work is to provide the basic design that can be adapted for use in a remote hot cell environment. In support of this objective, transducers were identified that will survive the hot cell radiation environment, a technique for verifying acceptable system performance while

inspecting closure welds was selected, and data were obtained that will lead to a design for in-cell calibration standards.

An engineering development scanner was designed, assembled, and demonstrated. The scanner, when used with a commercially available ultrasonic test instrument, can reliably detect and record sawcut notches with inner and outer diameter 0.100 in. deep and lying in the weld plane of an actual inertially welded closure. The scanner is acoustically coupled to the canister top by a low-volume flow of water through a narrow gap between the transducer and canister. This coupling technique was demonstrated at surface temperatures of 215°F. The transducers and coupling technique were also evaluated on secondary closure samples. Commercially available transducers were procured that have the same construction as ones procured by AECL in Canada for inservice inspection of their CANDU reactors. These transducers have an operating life of 150 hr in a radiation field of 2×10^6 R/hr gamma plus 30×10^9 neutrons/cm² sec. These transducers also exhibit acceptable acoustic performance. Various inspection verification notch configurations were evaluated. These tests showed that the best acoustic signal to monitor for system performance is the reflection from the underside of the canister top.

In-Melter Viewing

An important tool in operating a liquid-fed ceramic melter is the capability to observe the molten glass in the melter cavity. A television camera, mounted on the melter, that views the melt surface through an arrangement of mirrors and lens is used to make these in-melter observations.

The melter-viewing system has a periscope section that fits into a port in the top of

the melter and projects an image into an industrial TV camera mounted on the outside of the melter (see Figure 1.3). The TV signal is transmitted through a modified Hanford electrical connector and conduit to a control room outside the radiation zone. The lenses in the periscope are protected by a metal tube sealed by a metal cap with a 0.067-in.-dia aperture located at the focal point of the lens. With this arrangement, the field of vision is unrestricted. A continuous flow of air keeps the aperture clear of any glass spatters ejected from the surface of the melt. A steam sweep of the aperture area is provided by a small line on the periscope tip. It removes the solids (from off-gas condensation) that would otherwise accumulate on the relatively cool tip and restrict the view of the melt.

The lenses for the in-melter viewing system consist of a quartz wide-angle lens and cerium-stabilized optical glass for other lenses. Both quartz and cerium-stabilized glass are resistant to radiation damage. Quartz, which is particularly resistant to heat, is the final lens in the periscope.

Since the camera operates in the visible and near-infrared range, a source of light is required in order to produce an image on a TV monitor. The light source within the melting cavity is the surface of the molten glass itself. As this surface becomes covered with crust, consisting of feed materials in various stages of melting, the light source for the camera becomes obscured. The extent of crust, or cold cap, on the molten surface can therefore be judged using the camera simply by noting the dark spots on the TV monitor. Should the entire surface of the melt become covered with cold cap, an unwanted situation, the image of the melting cavity would become completely dark because the cold cap totally obscures the molten glass. This was observed

FIGURE 1.3. Melter Viewing System Components and Cooling Air Flow

both visually and on the TV monitor in the West Valley August 1984 run using the pilot-scale ceramic melter. The TV camera, unambiguously indicates the extent of the cold cap since the molten surface serves as the sole source of light.

The viewing system was handled as though it were in a remote environment where it would be installed in or removed from a melter by an overhead crane.

In a test of its ruggedness, the viewing system was lowered by crane into a simulated melter viewing port, swinging and striking the port as it was positioned. The viewing system was fastened by tightening bolts with

a remotely operated impact wrench. This handling procedure produced no change in the quality of the TV picture.

Tests were conducted on the features designed to keep the 0.067-in. viewing aperture clear of splatters and off-gas condensate. In one test, the air flow through the 0.067-in. viewing aperture effectively deflected a spray of toolmarker ink projected at the lens from an atomizer 2 in. away. No deposits were found on the objective lens with an air flow as low as 1.1 scfm at 5 psi.

In the initial operability test, gas condensing on the end of the periscope appeared

as a fuzziness around the edge of the television picture. Deposition of solids continued on the viewing aperture until vision was extremely limited. To recover the picture, the viewing system had to be removed and the aperture end of the periscope scraped clean of the encrustation. A steam line was installed that projects a flow of steam across the endcap and the 0.067-in. aperture. The steam flow, used only as needed, has succeeded in eliminating the problem caused by gases condensing on the periscope.

Following the addition of steam-cleaning capability, a long-term operation-to-failure test of the melter viewing system began with its installation in an operating melter. Monitoring performed on the melter viewing system depended on the melter operating mode. When the melter was idling, the power to the TV camera and the TV screen was normally off, but switched on daily, and the clarity of the picture was noted. An idling melter requires little attention; thus a daily or once-a-shift check of the system to see if the melter viewing system is operational would not deprive operators of needed information. During glass production, the melter and melter viewing operates continuously.

The melter viewing system was operated continuously during two melter tests: one with a simulated Hanford feed and the other with a simulated West Valley feed. The total installed time for the viewing system was 1400 hr. The picture quality was good throughout this operating period.

Camera temperature, camera coolant air, and air flow through the aperture were measured at regular intervals. No effort was made to control camera temperature by adjusting the coolant air flow or to increase the aperture air flow rate above rates identified as adequate in previous experiments.

The melter viewing system never failed to produce a sharp, clear picture of the melter cavity and molten glass throughout the test period.

Bulk Slurry Feed Transfer System

A test was conducted to determine the feasibility of using a steam-driven jet pump to transfer NCAW feed slurry between two vessels. A Penberthy-Houdaille, Model GH 2-in., 304 stainless steel (SS) jet was used for the tests (Figure 1.4). The jet was installed in a 2-in., 304 SS pipe running between two tanks, one that supplied feed to the jet and one that received the jet discharge. Steam at 90 psi was used to operate the jet.

Tests were performed and measurements taken to evaluate the performance and erosive characteristics of the jet. Two performance characteristics were evaluated, jet dilution and transfer rate. Jet dilution is the condensed steam from the jet operation which mixes with the feed during transfer and increases the volume of the jetted feed. The feed compositions used for these tests are shown in Table 1.3. Some trace components (<0.41 wt%) normally found in NCAW were transmitted. The transfer rate is the time taken to transfer a known volume of feed. The erosive characteristics were evaluated by measuring the jet discharge inside diameter and the steam nozzle outside diameter before and after the test.

The test was designed to maintain as constant a concentration of abrasive solids, i.e., silica, during the test as reasonably possible. To maintain the concentration, jet dilution was removed with a thermosiphon evaporator after the specific gravity of the feed had changed noticeably.

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8406142-1

8406142-4

8406142-2

FIGURE 1.4. Stainless Steel Jet Used for Tests

TABLE 1.3. Jet Test Feed Composition

Compound	wt%
$\text{Fe}(\text{OH})_3$	4.63
$\text{Al}(\text{OH})_3$	1.97
$\text{Na}_2\text{SiO}_3 \cdot 5\text{H}_2\text{O}$	0.65
Diclymium Carbonate	0.51
$\text{ZrO}(\text{NO}_3)_2 \cdot 2\text{H}_2\text{O}$	0.50
SiO_2 (-325 mesh)	14.68
$\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$	11.17
NaNO_3	8.99
LiNO_3	5.04
$\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$	1.32
H_2O	49.44

Nine transfers, totalling 6530 L of feed, were performed during the test. The manufacturer's specification for jet dilution is 1260 lb/hr of operation when water is the jetted solution. The average jet dilution value for the test transfers was 2466 lb/hr. The average feed transfer rate was 236 ± 21 L/hr. The average jet dilution for the test was about twice that specified by the manufacturer. This difference may be due to the greater density and viscosity of the feed used in the test relative to water. There was no noticeable increase in jet dilution from the beginning to the end of the test.

An increase in jet dilution could be explained by erosion of the jet, but this was not observed during the test.

The inside diameter of the jet increased an average of 0.0038 in. along its length, with the greatest increases near the jet intake. The change in the outside diameter of the steam nozzle was negligible. There was no noticeable change in the jet's pumping capacity during the test. Additional tests are recommended to determine long-term effects of erosion on pumping capacity.

Evaluation of Multiple-Pin Electrical and Instrument Connectors

A multipin electrical/instrument connector concept, developed at Savannah River Laboratories (SRL) and based on the proven Hanford remote connector technology, was evaluated. Four different connectors were fabricated, and remotability and electrical/instrument signal performance tests were conducted.

A 40-pin instrument connector was built and mounted on a jumper (Figure 1.5) to allow evaluation of the remote handling and installation of a multipin jumper. The jumper was installed several times without difficulty using remote crane and viewing systems. Initial break-in of the connector was required to achieve good pin/socket alignment between the connector and the wall flange.

A 10-pin connector (Figure 1.6) was built and connected to five thermocouples (T/Cs) to evaluate the transmitting of T/C signals. Thermocouple signals were found to have a mean offset of -0.237°C . Over 400 measurements, temperature reading offset was found to be independent of both the number of times the connector is disconnected and the temperature being measured (from 25 to 1050°C).

FIGURE 1.5. Test Jumper

The instrument connector pins are thus well characterized and are suitable for transmitting signals from remote instruments.

A 20-pin, 90 A/pin connector (Figure 1.7) was built and evaluated for its current-carrying capacity. The pins were found capable of carrying 120 A at a safe pin temperature of 65°C , and thus are capable of operating at 130% of their rated capacity. At 90 A/pin operation, such a connector is predicted to generate 3.3 kW of heat, which may require air cooling.

A closed-circuit TV signal connector (Figure 1.8), using instrument connector pins, was built and tested in conjunction with the melter viewing system. No radio frequency interference problems were encountered even though a coaxial (shielded) pin assembly was not used in the test connector. The quality of the transmitted TV signal observed on a high-resolution black and white monitor was considered acceptable.

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FIGURE 1.6. Test Version of the Multipin Connector

Analysis of Data from Thermal Mockup Test of Discharge Section

Analysis of the data from the thermal mockup test of the discharge section was completed. As a result of discharge mockup testing, lid-mounted heating elements were found capable of providing enough heat to maintain the metal trough and pour spout, as well as the upper portion of the discharge cone, at or above the 1000°C minimum at discharge plenum temperatures of 1050°C. An evaluation of the effect of element failure indicated that of the eight lid heating elements, any one of the four corner elements can fail and have little effect on trough temperatures; but the loss of any one of the four middle elements requires a substantial increase in power of the remaining elements

to maintain trough temperatures. Normal lid element power level varies from 20 kW to 30 kW. An air-cooled glass dam was found to operate at temperatures below 650°C, thus preventing glass migration, at an air flow rate of 10 scfm. Power loss associated with the dam at this flow rate is 3 kW. During testing, the cooling of pour spout temperatures through radiant heat transfer to a cold canister was found to be negligible. Turntable air inleakage above 3.0 scfm was found to dramatically lower pour spout temperatures, requiring increased lid power levels (additional 4 kW at 8.5 scfm) to compensate.

The performance of the lid-heating elements was also evaluated. PNL-designed element contact clamps were found to operate at temperatures substantially higher than achievable using conventional hardware. When

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FIGURE 1.7. 20-Pin/90-Amp Connector Test Apparatus

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FIGURE 1.8. Television Signal Test Connector

element contact surfaces were modified by removing a manufacturer-applied spray coating and reapplying conductive metals using a PNL electroplating technique, contact temperatures were lowered, resulting in anticipated longer element lifetime. It was found that element enclosures designed to minimize through-element inleakage maintained sufficiently low temperatures in the contact environment when left uncooled. When air- or water-cooled, temperatures were not lowered enough to warrant the complexity of remote coolant lines and exhaust.

The inleakage characteristics of two types of discharge lid gasket material were found to fit an orifice-type flow versus pressure drop equation, allowing the calculation of an "orifice" constant for each gasket. At -10 in. H_2O melter vacuum, a 1/8-in.-thick flat gasket leaked at 130 scfh, while a

1-in.-dia. rope gasket leaked at 150 scfh. These leak ratios are well within the arbitrary acceptable leak rate of 300 scfh.

During steady-state operation, the internal temperatures of the discharge section were as shown in Figure 1.9, which also indicates the distribution of element power. Note that element #7 was not operating and that element #8 was operating at ~1 kW, so that most of the heat was distributed from behind the pour spout. At a total power level of 22.57 kW to the lid, trough discharge temperatures were all above 1000°C, with an average pour spout temperature of 1021°C, an average glass path temperature of 1069°C, and through-the-wall temperatures up to 1097°C. The dam centerline temperatures were as shown, with an average air temperature, found to be relatively independent of dam air flow from 4 cfm to 20 cfm, of 260°C.

FIGURE 1.9. Internal Temperature During Steady-State Operation

The cooled edges of the dam were thus cold enough (below 650°C) to stop the migration of glass.

Figure 1.10 is a summary of simple energy balances performed on the discharge mockup for various dam cooling flow rates. For each case, the total input power from the lid elements, the heat loss due to dam cooling, and the remainder considered as wall losses, are given based on the test data. As can be seen, the wall losses at fairly constant plenum and plate temperatures (first four cases) are 22.4 kW $\pm$ 0.3 kW. The total input power required to maintain internal discharge temperatures depends on dam cooling and ranges from about 20 to 30 kW, depending on the extent of cooling. As can be seen, the exterior wall temperatures range from 27°C below to 38°C above the 200°C maximum exterior temperature requirement, with an average exterior temperature of 200°C.

Calculations were made to predict the performance of an actual HWVP discharge section based on the thermal mockup test. As summarized in Table 1.4, for the case shown with

the thermal mockup operating at 25.78 kW, an actual HWVP discharge section is predicted to require 33.85 kW.

Finally, the warpage of the air-cooled glass dam was measured and is shown in Figure 1.11. This warping is considered tolerable provided there is a 3/4-in. gap between the dam and adjacent refractory to avoid damage to the refractory.

Melter Nozzle Cleaning System

Tests were conducted to evaluate the remote operation of a melter nozzle cleaning device to be used for unclogging melter nozzles, particularly the off-gas nozzle.

The nozzle cleaner consists of a core-drilling apparatus that is lowered by crane onto an open melter nozzle. A flange on the nozzle cleaner fits over guide dowels and threaded studs that are on a flange of the melter nozzle. The cleaner is then remotely bolted into place (Figure 1.12). A drill feed mechanism lowers a rotating, diamond-surfaced bit (standard core drill) into the

FIGURE 1.10. Discharge Mockup Energy Balances at Walls for Various Dam Flow Rates

TABLE 1.4. Calculated and Predicted Discharge Section Energy Balances

Heat Transfer Component	Thermal Mockup	HWVP Discharge Section (a)
Lid losses	1.37	1.89
Side wall losses	6.93	9.54
Front wall losses	6.34	8.72
Floor losses	3.20	4.40
Discharge port losses	2.38	3.27
Glass dam losses	3.40 ^(b)	3.40
Discharge port inleakage	1.70 ^(c)	1.7
Discharge lid inleakage	0.46 ^(d)	0.46
	25.78	33.85

(a) Predicted energy balance.

(b) Based on glass dam air flow of 14.5 scfm.

(c) Based on turntable air inleakage flow of 3.0 scfm.

(d) Based on discharge lid inleakage flow of 8.5 scfm.

FIGURE 1.11. Characterization of Dam Warpage

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 mechanism automatically reverses and lifts the bit out of the nozzle. When the bit is clear of the nozzle, power to the assembly is disconnected and the nozzle cleaner is remotely removed from the melter.

The nozzle cleaner was tested at melter temperatures using the thermal mockup to simulate an idling melter plenum. The test setup is shown in Figure 1.12. It was found

that at a melter vacuum of -5 in. H_2O , sufficient air was pulled through the cutting area to cool the bit and remove some of the cutting debris. Water cooling was also found to provide sufficient cooling during ambient (low temperature) testing, but was not tested further since air inleakage cooling was found to be adequate and avoids the complexity of a water-cooling jumper and the possible undesirable effects associated with pouring water in an 800°C+ melter. The rate of inleakage through the cutting area was estimated as 18 scfm in the test apparatus.

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FIGURE 1.12. Melter Nozzle Cleaner

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

V. F. Fitzpatrick - Program Manager

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 major accomplishments in FY 1984 were completion of the design and fabrication of the large-scale system. The large-scale in-situ vitrification (ISV) system was delivered to Hanford on August 30, 1984. Scaled up tenfold over the pilot-scale system, it is housed in three large trailers and is state-of-the-art in off-gas treatment and power and process control.

The system is the result of 13 months' intensive effort by a broad spectrum of laboratory staff. Permission to proceed with conceptual design was received mid-July 1983, and the operational hardware was delivered 13 months later. The conceptual design report and all 92 Title II detailed engineering drawings were approved by both PNL and Rockwell Hanford Operations (Rockwell).

A PNL Operational Readiness Review (ORR) Board was selected and approved the Operational Readiness Plan (ORP). This early approval of the ORP will facilitate preparations for the Operational Acceptance Test (OAT) of the large-scale system. A joint Rockwell/PNL site-selection committee issued a report recommending the principal and alternative test sites for the first large-scale radioactive test. The recommendation has been transmitted to DOE-RL for concurrence.

Molybdenum was qualified as the electrode material for initial large-scale testing

based upon long-term pilot-scale testing. Unresolved corrosion problems with graphite electrodes necessitated the selection and qualification of an alternative material for initial testing.

Significant progress was made in developing a depth monitor that uses fiber optics sensing techniques for large-scale field testing. The problem of electrical interference with the transmitter signal has been eliminated.

A document presenting the detailed results of, and conclusions from, the pilot-scale radioactive test (PNL-5240) was submitted for clearance.

Large-Scale System - J. L. Buel, J. G. Carter, C. R. Hymas, S. C. Liikala, K. H. Oma, and C. L. Timmerman

The large-scale ISV system is being developed for testing as one of the complementary technologies for in-place stabilization and disposal of radioactively contaminated soils. The most significant accomplishment in this reporting period for the large-scale system was completion of the fabrication of the large-scale prototype.

The fabrication phase included receipt and inspection of all government-furnished processing equipment and the assembly of the equipment into three processing trailers that house the system. All piping jumpers were fabricated, and the electrical equipment, the instrumentation, and the process control system required for process operation, monitoring, and control were installed. The fabrication phase also included the fabrication of the hood, which captures process effluents from the zone of contaminated soil being vitrified and directs them to the off-gas treatment system.

Procurement of Equipment

All major processing equipment necessary for the fabrication and assembly of the processing trailers has been received, inspected, and delivered to the offsite contractor assembling the trailer. The processing equipment included 45 separate items of effluent treatment equipment such as scrubbers, a condenser, heat exchangers, scrub pumps, and induced draft blowers; electrical equipment, including transformers, a motor control center, and electrical switchgear; instrumentation; and a process control system.

Before shipment to the contractor, all equipment underwent inspection, and many components were subjected to acceptance tests to determine their conformance to specifications. All nonconformances identified during acceptance testing were resolved. One of the more critical pieces of equipment, the tandem nozzle scrubber, was subjected to a performance acceptance test. The performance criterion for the test was that under nominal off-gas flow conditions, the scrubber must remove more than 90% of particles 0.5 μ m in diameter. The test results indicated that the performance criterion was greatly exceeded (97 to 99% removal efficiency). This means that greater than expected particulate removal efficiencies are now anticipated during testing of the large-scale system.

Fabrication and Assembly of the Processing Trailers

A fixed-price competitive bid contract was let for the fabrication and assembly of equipment into three processing trailers. Fabrication and assembly was performed to appropriate National Codes for piping and electrical systems, and a rigorous quality

assurance program was maintained. The three trailers were delivered with the assembled/fabrication processing equipment in August 1984 for final acceptance testing. The completion of this contract concluded the efforts toward a major milestone for the development and testing of the large-scale system.

A preliminary acceptance test was completed at the contractor's facility to ensure compliance with the technical specifications of the contract. The acceptance test required operation of all pieces of equipment in the trailers to check for leaks in the piping and containment module, faulty wiring (e.g., insulation quality, phasing, and motor rotation), and performance of the equipment operating as a system. The findings of the acceptance test included the following:

- The piping system was pressure-tested and determined to be well within the acceptance criteria.
- The requirements for air flow and negative pressure in the containment module were met (i.e., >200 scfm flow at >0.5 in. H₂O negative pressure). Smoke tests revealed no leaks in the containment module.
- All 480 V electrical systems and 240/120 V systems at the distribution panel exceeded megging test requirements (i.e., >1 M Ω to ground over a 60-sec period).
- Both blowers and all but one pump operated appropriately. One seal on a scrubber pump (P-1) failed at high pressure. The O-ring was replaced and a stronger clamp was provided on the pump.
- All but three of the ten valves met the time requirements for operation from fully closed to fully open (<2 sec for on/off valves and <9 sec for control valves). One of the 16-in. and two 12-in. butterfly valves had response times of 8 sec and 4 sec, respectively. Both faults were

corrected by PNL as they were not caused by poor workmanship by the contractor of the trailer.

The off-gas trailer is shown schematically in Figure 1.13. All three trailers are shown schematically in Figure 1.14.

Hood Fabrication

The fabrication work on the stainless steel hood (Figure 1.15) was completed in this reporting period. All that remained was assembly at the contractor's site to verify fit, and the shipment and the assembly at the testing site. The hood was fabricated by an offsite contractor using a design prepared by Rockwell. All internal portions of the hood that may be exposed to corrosive and radioactive gases are composed entirely of 300-series stainless steel. Those portions exposed to $>400^{\circ}\text{C}$ are of 321 or 347 stainless steels, which have stabilizers to resist corrosion at high temperatures. The hood,

12 m on a side, is constructed of numerous panels, 1.2 m by 2.4 m, which can expand independently of one another to prevent panel buckling and warpage.

Pilot-Scale Deep-Depth Test - C. L. Timmerman and K. H. Oma

Pilot-scale Cold Test-3 (PSCT-3) was completed; in this test the effectiveness of using molybdenum electrodes for deeper and longer ISV operation was demonstrated. Analysis of the molybdenum electrodes confirmed that they will be satisfactory for initial large-scale testing. Present predictions indicate operational lifetimes of greater than 1000 hr based on the highest uniform corrosion rates observed in PSCT-3 and in previous tests. Oxidation of molybdenum by air above the ground level is effectively controlled by a thin coating of 316 stainless steel applied by flame spraying. In the subsidence zone, oxidation is controlled by a

FIGURE 1.13. Off-Gas Trailer

FIGURE 1.14. Overhead Views of Process Trailers

natural coating of glass that adheres to the electrodes as the molten glass subsides.

During PSCT-3, power was applied to the electrodes for a period of 107 hr, resulting in a melt depth of 4 m and a vitrified soil mass of about 50 metric tons. Figure 1.16 shows the vitrified block produced. Total energy consumption was 39,600 kW, resulting in an average power consumption of 370 kW. This is much higher than the average powers achieved during the two previous operational cold tests with this pilot-scale system. The formation of stable cold cap greatly reduced heat losses to the off-gas system and contributed to an efficient operation. An

energy-to-mass ratio of 0.8 kWh/kg is estimated.

Several factors contributed to the stable cold cap formation during this test: 1) the startup was achieved using a low power input, which allowed the cold cap to form; 2) glass adhesion to the electrodes provided the additional support needed for the cold cap; 3) the lower cross-sectional area of molybdenum electrodes (one-ninth that of graphite) resulted in less heat transfer to the top region and thus lower temperatures and a more stable cold cap; and 4) the electrode spacing (1.0 m) was closer than for previous tests

FIGURE 1.15. Off-Gas Hood

(1.2 m), reducing the spanning distance and thus strengthening support of the cold cap.

A transmitter/receiver-type depth monitor was successfully tested during PSCT-3. The transmitter, operating at a frequency of 13.56 MHz, was attached to the bottom of one of the electrodes. Fiber-optic depth sensors were positioned at incremental levels along

the electrode and terminated at the transmitter. As the hot glass contacted each sensor, a receiver at the surface detected a change in the signal from the transmitter. The transmission signal was strong throughout the test, providing confidence that the concept will perform well during large-scale ISV.

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FIGURE 1.16. Vitrified Block Produced During Pilot-Scale Cold Test-3

Vitrification Zone Growth Profiles -

K. H. Oma and S. C. Liikala

Melt-profile data were collected during Engineering-Scale Laboratory Test-20 (ESLT-20) and during PSCT-3 to compare with the ISV mathematical model. Both tests used a vertical plan of thermocouples protruding from one side of the melt zone to obtain the data. The pilot-scale melt zone grew faster outward than downward as compared to the engineering-scale melt zone. Several factors may be involved in this: 1) The higher heat conduction of the graphite electrodes used in ESLT-20, as compared to molybdenum electrodes in PSCT-3, may have contributed to the faster melt growth downward. 2) Differences in backfill density and composition could also have affected the melt zone shape. 3) Convective mixing of the molten soil is probably more complete during pilot-scale ISV operation since higher glass temperatures result from the larger melt-zone volume and longer run times. This aids in distributing temperature more uniformly in the pilot-scale system, and thus a more equal growth rate results both sideways and downwards. The molten soil near the edges of the engineering-scale melt zone may be cooler than that near the bottom, resulting in slower outward growth and thus a narrower block.

1.3 DEFENSE BYPRODUCTS PRODUCTION AND UTILIZATION PROGRAM

(AR-05-15-25/80576 - GE-0347-03/80401)

G. L. Tingey - Program Manager

The objective of this program is to identify and develop safe and cost-effective applications for nuclear byproducts, and to optimize the supply of these byproducts, especially those available from Hanford. During FY 1985, this program will focus on developing irradiation for food and other agricultural products in the Northwest,

supporting activities for defense energy sources for a 500-W_e power generator, and developing an adequate supply of safe, cesium-137 sources qualified as irradiation sources.

Summary

Fresh cherries have been irradiated with cobalt-60 gamma at doses up to 200 krad and have been evaluated for quality and by a sensory test panel to determine their suitability for human consumption. Irradiated cherries were also compared to those fumigated by methyl bromide at 7°C. The results of the tests led to the following conclusions:

- Irradiation to a dose of 25 to 30 krad appears to be sufficient to meet quarantine requirements.
- Irradiation of cherries to doses well above those required for quarantine treatment does not significantly degrade the fruit.
- Irradiation reduces the firmness of cherries but has no significant effect on other quality factors.
- Dose levels of up to 60 krad do not significantly affect cherry quality.
- Irradiation up to 200 krad at a temperature of about 25°C does not provide measurable shelf-life extension.
- Irradiation to high doses (>200 krad) at 25°C appears to produce some degradation of cherries.
- There is no apparent advantage to irradiation over low-temperature fumigation. (However, no sensory tests are conducted on fumigated cherries.)

Tritium-powered radioluminescent lights have been tested both statically (by photographic techniques) and by dynamic flight tests near the Hanford Site. The tests were performed to supply the Federal Aviation

Administration (FAA) with data required to evaluate the usefulness of the lights for commercial operation in remote locations. Sufficient photographic data were obtained to permit modeling of the light density at various locations. Evaluations of the flight-test data are not yet completed, but preliminary assessment has shown that the pilots were able to identify, maintain contact with, and use the runway from 1.5 miles or greater during approach and landing.

A strontium-90 fueled electrical power generator is being designed and constructed as a demonstration of high-efficiency radio-isotope power generation. This demonstration will couple a strontium-90 fuel heat source to a free-piston Stirling engine with linear alternator fabricated by Sunpower, Inc. The calculated efficiency of this generator will be 21% to 23%. Advanced generators are anticipated to have an overall efficiency of 35% to 45%.

Irradiation of Northwest Agricultural Products - D. E. Eakin

During the 1984 crop year, two batches of cherries were irradiated for evaluation of quality. Each batch consisted of cherries from three different orchards or packing houses. The cherries were irradiated at about 26°C (78°F) at total doses of 0, 20, 40, 60, 80, 100, and 200 krad. Samples of cherries from the same sources were also fumigated. The fumigation was done at about 7°C (45°F). Following irradiation, the cherries were transported to the U.S. Department of Agriculture (USDA) laboratory in Wenatchee, Washington, and prepared for chemical and physical testing. In addition, samples irradiated at 0, 40, 80, and 200 krad from the second batch were sent to the

Sensory Science Laboratory at Oregon State University for sensory evaluation. The test results are discussed below.

Quality Evaluation

Samples of the cherries sent to Wenatchee were held in cold storage at -1°C (31°F) for periods of 0, 7, 14, and 28 days following irradiation. The quality of the cherries was evaluated immediately after samples were removed from cold storage and also after samples had been held in the ripening room at 20°C (68°F) for 3 days following cold storage. The following tests were conducted on each sample:

- percent decay - percentage of cherries showing decay such as mold
- weight - measured in grams
- condition - subjective visual evaluation on scale of 1 (good) to 4 (poor)
- stem diameter - measured in 1,000ths of an inch with a micrometer
- stem color - subjective visual evaluation on scale of 1 (good) to 4 (poor)
- size - measure of cherry diameter by row number (9, 10, 11, 12, 13 row corresponding to 75/64, 67/64, 61/64, 56/64, and 52/64 of an inch, respectively)
- color - visual evaluation comparing cherries to standard industry color balls (scale of 1 to 5, light to dark)
- 3 FRM - color measurement by three-filter reflectance method
- firmness - measured with pressure tester with readings in percent (100% = 8 oz)
- acid - following pitting and juicing, the acid in the juice measured by titration and expressed as percent malic acid
- pH - the pH of the juice
- soluble solids - measured with refractometer.

The data obtained from these measurements showed some statistically significant differences in color, condition, percent decay, pH, soluble solids, stem color, and stem diameter for individual test periods, but there was no correlation with treatment (dose level or fumigation). Generally there were no significant differences in initial weights or weight losses during ripening nor differences in fruit size with respect to treatment. The acid measurements showed significant differences in individual test periods, but no correlation with dose level. The fumigated cherries generally had the highest acid content of any of the samples. Loss of acid occurs during ripening, and retention of acid

is normally desirable. On this basis only, the fumigated cherries appeared superior to the irradiated ones, but other tests showed no advantage. Sensory tests remain to be done.

Table 1.5 shows cherry firmness scores for both batches of cherries immediately following cold storage and after 3 days of ripening following cold storage. The data show a decline in cherry firmness with increasing dose levels. In nearly all cases, the cherries that received a total dose of 200 krad were rated as least firm. The differences between cherries irradiated to 200 krad and the non-irradiated control were great enough to be considered statistically significant. The

TABLE 1.5. Cherry Firmness (measured with pressure tester;
10 = full scale = 8 oz)

Treatment, krad	Batch 1, Yakima District Storage Time, days				Batch 2, Wenatchee District Storage Time, days			
	0	7	14	28	0	7	14	28
0	6.80	7.04	7.08	6.73	6.49	6.58	6.53	6.83
20	6.73	6.92	6.97	6.44	6.49	6.49	6.30	6.83
40	6.37	6.83	6.99	6.51	6.47	6.37	6.53	6.70
60	6.26	6.66	6.90	6.59	6.26	6.36	6.37	6.60
80	6.30	6.67	6.87	6.46	6.23	6.55	6.27	6.50
100	6.33	6.70	7.00	6.20	6.32	6.40	6.30	6.43
200	6.13	6.43	6.54	6.37	6.04	6.08	6.10	6.37
Fumigated	6.47	6.79	7.10	6.67	6.34	6.54	6.57	6.67
Least significant difference 95% confidence level	0.206	0.156	0.238	0.493	0.298	0.437	0.432	0.352
After 3-day Ripening Period								
0	6.59	6.63	6.79	6.77	6.45	6.38	6.24	6.37
20	6.49	6.67	6.55	6.46	6.20	6.15	5.99	6.22
40	6.43	6.50	6.48	6.65	6.26	6.11	5.93	6.07
60	6.42	6.50	6.41	6.48	6.22	6.15	5.88	6.07
80	6.35	6.50	6.49	6.50	6.00	5.81	5.83	5.87
100	6.30	6.47	6.58	6.42	6.15	5.76	5.67	5.97
200	6.09	6.27	6.00	6.08	5.91	5.84	5.57	5.67
Fumigated	6.60	6.53	6.50	6.25	6.80	6.14	5.93	5.83
Least significant difference 95% confidence level	0.201	0.384	0.273	0.368	0.620	0.281	0.517	0.224

fumigated cherries generally were rated between the lower-dose and the higher-dose irradiated cherries. Although receiving lower firmness ratings, cherries irradiated up to 60 krad generally were not seen as being significantly different from the non-irradiated ones. In the table, a rating of 6 or less would probably represent an unacceptable cherry. Therefore, when starting with cherries softer than normal (as was the case for batch 2), irradiation at the 200-krad level can reduce the firmness to less than the acceptable level. Comparing firmness measurements from batches 1 and 2 shows a great variation in ratings, depending on the source of the cherries. This variation was often greater than the difference caused by irradiation.

Sensory Evaluation

A portion of the cherry samples irradiated at 0, 40, 80, and 200 krad from batch 2 were sent to the Sensory Science Laboratory, Department of Food Science and Technology, Oregon State University in Corvallis, Oregon. These cherries were evaluated by a 20-member panel on the basis of appearance, firmness, and flavor after refrigerated-storage intervals of about 1, 2, 3, and 4 weeks following the irradiation. Table 1.6 shows the mean scores for each separate time of testing across all three attributes. One week after irradiation, there was no statistical difference in appearance ratings. The panel rated the flavor of the 200-krad irradiated sample somewhat less desirable than the others and observed a statistically significant decrease in firmness with dose.

After 2 weeks of storage, a difference in appearance was noted between the samples. The appearance of the control and 40-krad samples was significantly preferred over the

other samples. For firmness, the 80- and 200-krad samples were judged significantly less firm. Flavor declined, with the 200-krad sample being rated significantly lower than the control and 40-krad samples. The 40-krad sample was the only sample not rated significantly lower in palatability than the control after 2 weeks of storage.

After 3 weeks of storage, the panel rated the 80- to 200-krad samples lower than the lower dose samples in all three properties. The 40-krad sample continued to be rated as not significantly different from the control, and there was no significant decrease in rating with storage time.

After 4 weeks of storage, the appearance rating of the 40-krad sample dropped and was significantly lower than the control sample. The control sample was rated significantly more firm than all three irradiated samples. The flavor scores for the control and 40-krad samples dropped after the third week so that they were now judged essentially the same as the other samples. It should be noted, however, that in all cases the cherries were judged desirable, and thus should be considered acceptable for market.

The subjective sensory evaluation tests agree generally with the more quantitative products-quality tests. Based on these results, the following conclusions have been reached:

- Irradiation to a dose of 25 to 30 krad appears to be sufficient to meet quarantine requirements.
- Irradiation of cherries to doses well above those required for quarantine treatment does not significantly degrade the fruit.
- Irradiation reduces the firmness of cherries but has no significant effect on other quality factors.

TABLE 1.6. Cherry Sensory Evaluation

Treatments	Mean Scores of Each Attribute for Discrete Test Times		
	Appearance(a)	Flavor(a)	Firmness(b)
<u>Week 1</u>			
Control	7.10	7.30	6.35
40 krad	6.35	6.75	5.85
80 krad	6.95	6.75	5.10
200 krad	6.35	5.45	4.10
Least significant difference 95% confidence level	0.80	0.98	0.91
<u>Week 2</u>			
Control	7.30	6.50	6.10
40 krad	7.30	6.85	5.85
80 krad	5.65	5.75	4.75
200 krad	5.95	5.25	4.10
Least significant difference 95% confidence level	0.84	1.07	0.93
<u>Week 3</u>			
Control	7.10	7.20	6.10
40 krad	7.25	7.25	5.80
80 krad	5.35	5.50	3.95
200 krad	5.05	5.50	3.90
Least significant difference 95% confidence level	0.73	0.80	0.79
<u>Week 4</u>			
Control	7.35	6.30	5.85
40 krad	5.90	6.25	5.00
80 krad	5.55	5.75	4.50
200 krad	5.50	6.15	4.55
Least significant difference 95% confidence level	0.89	0.98	0.97

(a) Scored by use of nine-point scale (9 = like extremely, 1 = dislike extremely).

(b) Scored by a seven-point firmness scale (7 = just right, 1 = too soft).

- Dose levels of up to 60 krad do not significantly affect cherry quality.
- Irradiation up to 200 krad at a temperature of about 25°C does not provide measurable shelf-life extension.
- Irradiation to high doses (>200 krad) at 25°C appears to produce some degradation of cherries.
- There is no apparent advantage to irradiation over low-temperature fumigation.

(However, no sensory tests are conducted on fumigated cherries.)

To further evaluate the potential of extending the shelf life of cherries, a test is planned for the 1985 crop which will use higher dose levels at lower temperature and/or in inert atmosphere. To provide a more thorough comparison with fumigation, a sensory evaluation test will be conducted on cherries fumigated at low temperatures.

Effectiveness of Disinfestation

During the cherry irradiation studies conducted in 1983, a batch of cherries was purposely infested with cherry fruit-fly larvae by the USDA laboratory in Yakima. The infested cherries were irradiated to nominal doses of 0, 4, 6, 8, 12, and 20 krad in the PNL cobalt-60 facility. The results of these tests after the normal time required for adult emergence are reported in Table 1.7. A substantial reduction in survival rate of pupae and emergence of adults was observed as a result of the irradiation. The table shows the survival rate as a function of gamma dose.

These data show a decrease in ability of irradiated fruit-fly larvae to mature and pupate even at the lowest dose tested (3.5 krad). The only adult emergence from treated larvae was a single fly at 10.5 krad. The lack of any adult emergence at lower dose levels leads us to believe that this fly was due to post-treatment contamination of the treated fruit. Also, the lack of adult emergence prevents an estimation of a dosage-mortality curve and prediction of a minimum irradiation dose required to prevent adult emergence. However, the doses required to

prevent 50% and 95% pupation of larvae were 2.7 and 26.4 krad, respectively. During 1984, batches of fruit-fly-infested cherries were irradiated at nominal doses of 0, 0.375, 0.75, 1.5, 3.0, 4.5, 6, and 9 krad. The results from these tests will be available in the spring of 1985.

A limited number of tests conducted to examine control of the codling moth have now been evaluated. These indicate that the irradiation dose required to prevent emergence of an adult codling moth from nondiapausing larvae (in fruit) was 20.7 krad and for diapausing larvae (in fiberboard strips) was 22.5 krad.

Radioluminescent (RL) Light Testing - G. A. Jensen

Tests and evaluations of the prototype light panels, airfield landing lights, and landing-aid systems for tactical, military, and rural airfields were conducted. The primary emphasis of these tests has been to further evaluate the system for arctic and subarctic conditions and for military use; however, potential applications and interest in using RL lighting is not restricted to these conditions. The tests reported here have been a cooperative effort between the State of Alaska Department of Transportation and Public Facilities (DOT&PF), Department of Energy (DOE-HQ), its operation offices at Oak Ridge, Tennessee (DOE-OR) and Richland, Washington (DOE-RL), and contractor personnel at Oak Ridge National Laboratory (ORNL) and PNL. Personnel also participated from the Air Force Engineering and Services Center and the Federal Aviation Administration (FAA) from the Atlantic City Test Center.

RL light panels incorporating the newer tritium tube design developed by ORNL during

TABLE 1.7. Survival Rate of Cherry Fruit Flies as a Function of Irradiation Dose

Dose Received, krad	Number of Surviving Pupae/ 200 Cherries	Number of Emergent Adults/ 200 Cherries
0	218	156
3.5	92	0
5.3	72	0
7.0	53	0
10.5	37	1
17.4	15	0

1983 were used in all the evaluations. Support fixtures in some of the work were those designed for the 1983-84 Alaskan testing. As a result of static and photographic evaluations, modifications were made and a new support fixture was used for flight evaluations near Hanford (Figure 1.17).

Using the older fixtures, the lights were evaluated photographically and visually using Rattlesnake Ridge on the Hanford Arid Lands Ecology site to simulate elevations and flight-pattern locations encountered by pilots during base and final approach. Two runway-lighting configurations and three glide-slope-indicator designs were evaluated using roadways to simulate runways (Figures 1.18, 1.19, and 1.20). Photographs were taken from distances of 0.33, 0.65, 0.77,

84H503-8

FIGURE 1.17. RL Light Support Fixture

FIGURE 1.18. 3200-ft x 80-ft Civilian Airfield Configuration

FIGURE 1.19. Military Airfield Configuration

1.1, 1.4, 1.5, 1.8, and 1.9 miles on the runway centerline and from distances of 0.48, 1.1, 1.4, 1.6, and 1.8 miles at a 25° angle to the centerline to simulate the base leg of the approach. Elevations above the runway were chosen to simulate the elevation the pilot would be above the runway using a 3° approach angle during his descent. Over 100 photographs were taken during these evaluations. One typical photograph taken at a 1.5-mile distance is included (Figure 1.21). Additional work was done using photographic techniques to determine the received-light density at various locations. These data will be used along with photometric measurements of individual panels to model the field-of-light density at different locations. We propose to use this information to

develop a computer simulation that can be used to train pilots and other users of the lights.

Although RL lights have been demonstrated for runway lighting and marking applications, regulatory approval for their use under various parts of the Federal Aviation Regulations (FAR) by the FAA requires specific testing and evaluation. Plans for some of the necessary testing were completed and coordinated with the DOE laboratories, the State of Alaska, and FAA. The specific purpose of the evaluation was to provide a formal initial evaluation of the RL runway-lighting system, which could lead to approval by the FAA for use in nighttime aircraft operations under Part 135 of the FAR. These tests apply only to Part 135 of the FAR; other tests may be

FIGURE 1.20. Visual Approach Slope Indicator

FIGURE 1.21. Civilian Runway--1.5-Mile Distance

required to meet other parts. The results from this test will also aid in developing the lights for military purposes. The goals of this test were:

1. Evaluate the human and other factors affecting the lights and their use. Specifically, the usefulness of the navigation and landing-aid system was evaluated using a specific configuration.
2. Evaluate a procedure or procedures for landing using the lights and suggest modifications in existing criteria used by high-performance aircraft. Most aircraft and pilots using the RL system for civilian aviation will be flying smaller, lower-performance aircraft.

After consideration of several potential sites, a site near the Hanford complex was selected.

The pilot-selection criteria were based on having subjects with air-taxi qualifications and experience. Thus pilots were required to have a minimum of 500 hr flying time, an instrument rating, and a commercial rating. Twenty pilots having these qualifications participated, including FAA flight standards personnel, a NASA test pilot, volunteers from the flight service supplying the aircraft, and several local pilots. During the tests, several visitors from NASA, PNL, DOE-RL, the States of Alaska and Washington, St. Petersburg, Florida, and the U.S. Army visited the site and participated. Cessna 206 and 172 aircraft were chosen for the tests since they are typical aircraft used for air-taxi operations. All tests were conducted with an FAA safety pilot and observer taking notes in the aircraft.

The three-phase test procedure was developed by FAA personnel in conjunction with the PNL and State of Alaska staff. Each phase was completed twice for a left- and right-landing pattern. In Phase I, the subject pilots were flown to a location 1.3 miles from the runway on the centerline, 45° from the centerline, and 90° from the runway centerline, were told the general location of the runway, and asked by the safety pilot if they could clearly see the runway and to point to its location. In Phase II, the pilots were located at a flight path parallel to, and 1.3 miles from, the runway centerline (downwind leg) and given control of the aircraft. The safety pilot then directed the subject pilot where to turn to base, i.e., perpendicular to the extended runway centerline, and then the pilot was directed to complete his approach to the runway and make a low flyover, demonstrating that he had the

required alignment and position for landing. Phase III was conducted by the safety pilot flying the aircraft to a location between 4 and 5 miles from the airport. The subject pilot was then given control of the aircraft and asked to locate the airfield using battery-powered strobes (Figure 1.22) placed about 1 mile north of the runway. The pilot then overflew the runway, located the wind T and identified the direction it was pointing, and made the appropriate downwind, base, and final low approach. Touch-and-go and full-stop landings were not required. Each pilot filled out a questionnaire and added his comments at the end of the test.

The 2100-ft runway was configured as shown in Figure 1.23. Appropriate 1.3-mile markers were identified and specifically placed as needed (Figure 1.24). During the tests, pilot evaluation, video, and photographic documentation were completed and the lights were removed, packaged, and shipped to ORNL.

Although analysis of the data is not complete, preliminary test results are quite favorable. All pilots could identify, maintain contact with, and use the runway from 1.5 miles or greater during approach and landing. Pilots also reported that they could use the lights for runway alignment from distances greater than 2 miles under very dark, clear conditions and between 1.5 and 2 miles under two-thirds to three-quarters moonlight and hazy conditions. Some pilots and observers reported visual contact with the runway at 3 to 3.5 miles along the extended runway centerline and 2 to 2.5 miles when 90° from the runway direction. Visual contact was measured during approaches from east to west or while flying parallel to and south of the runway centerline.

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FIGURE 1.22. Runway Locator Strobes

FIGURE 1.23. Flight Test Runway

FIGURE 1.24. Site of FAA Test

Development of Radioisotope Thermomechanical Generator (RTMG) - E. J. Wheelwright

Highly reliable, maintenance-free, long-life power generators continue to be needed for high-priority military and civilian uses. The military needs include power generators for remote-radar systems, for command, control, communication, and intelligence (C³I) systems and for advanced-weapons systems. Civilian needs include aircraft-navigational and collision-avoidance systems, and both emergency and normal power for remote locations. Strontium-90 produced in nuclear reactors is a uniquely valuable energy source that can meet these needs.

Previous applications of radioisotope power generation have almost exclusively used passive thermoelectric conversion. Although

these systems have high reliability, their efficiency is limited to about 8%. This inherently limits the power production capability using existing radioisotope fuel. Thus power generators are needed with similar high reliability but with much higher conversion efficiency (e.g., 20% to 40%). High-efficiency generators will expand the limited supply of available strontium-90 to meet greater needs for power generation.

Current research along with operational experience suggests that very high reliability can be achieved with high-efficiency radioisotope-powered thermomechanical generators (RTMGs). RTMGs with efficiency of 17% to 25% have been demonstrated to have both long life and high reliability. A demonstration strontium-90 fueled RTMG has been operated in England for the past 8 years without

maintenance and continues to produce power. In addition, a propane-fired (no radioisotope) thermomechanical generator was operated under the North Sea for nearly 3 years before the fuel supply was exhausted. The generator was then recovered and refueled, and it continued to perform at the design level. Conversion efficiencies of 35% to 45% are projected for generators using a 700°C heat source.

In March 1984, authorization was obtained to build and test a 1-kW_e RTMG powered by strontium capsules from the Hanford Waste Encapsulation and Storage Facility (WESF). The mechanical generator selected is a free-piston Stirling engine coupled to a linear alternator for production of electrical power. Following competitive bidding, a

contract was signed with Sunpower, Inc., of Athens, Ohio, to build an engine generator of their "Spike II" design. The generator is to be modified to interface with the isotope heat source being designed at PNL. Several system concepts were prepared and computer-modeled. Heat-flow calculations coupled with engine-performance data show that this RTMG should have a system thermal-conversion efficiency of 21% to 23%. The exact electrical power output will depend on the heat-generation rate of the WESF capsules selected for this initial RTMG demonstration program. Detailed design was initiated in the third quarter of 1984.

A schematic outline of the RTMG is shown in Figure 1.25. The six WESF capsules will be inserted into six cylindrical holes in the

FIGURE 1.25. Schematic of RTMG - Fueled by Six WESF Strontium Capsules

copper cylinder. The copper block will sit on top of the Stirling engine, and the heat from the WESF capsules will be conducted to the engine through the conic-interface surface. MIN-K insulation is used for thermal insulation, and the gamma radiation is shielded by the combination of copper and surrounding lead. Construction of the engine generator and of the heat source will be initiated as soon as detailed design is completed. Design completion is anticipated in November 1984.

1.4 MODELING RADIONUCLIDE RETARDATION MECHANISMS

(AR-05-10-02 - 80813)

J. R. Morrey - Program Manager

The objective of this program was to develop a capability to reliably predict retardation of radionuclides in soils and geological strata. It was structured to accomplish three major tasks: 1) to select and upgrade a baseline geochemical code; 2) to construct a streamlined, efficient geochemical code which would receive initializing information from the baseline code and be coupled to the appropriate aqueous transport code; and 3) to update and expand a selected thermodynamic data base.

Summary

This program, initiated in April 1983, had four technical tasks: 1) to develop an accurate and internally consistent thermodynamic data base; 2) to select and modify a baseline geochemical code; 3) to develop a compact, efficient geochemical subroutine that would couple to transport codes; and 4) to couple the geochemical code to a transport code to simulate transport of radionuclides in ground waters. The plan was to accomplish these objectives over a period of four years.

While technical progress was proceeding well, the program was terminated at the end

of FY 1984 because higher priorities required available funding. Thus the following objectives will not be met: The baseline geochemical code developed will not be verified and documented. The coupled code, operational as a prototype, remains unverified and undocumented. The selection of thermodynamic data for uranium, plutonium, and technetium was completed, but the document was not issued because of program termination.

Accomplishments of the program include the following:

- A baseline geochemical code was selected from among six other choices for further development.
- This code was modified to 1) markedly improve its convergence reliability, 2) include high-ionic-strength capability (though not verified), and 3) include a preprocessor code to help users communicate with the code.
- Thermodynamic data for uranium, plutonium, and technetium were selected and prepared for addition to the data base used by the baseline code.
- A prototype coupled geochemical code was created and was being tested. This code appears to hold unusual promise as an effective tool in predicting radionuclide transport in saturated porous media.

Baseline Geochemical Code

Selection and improvement of a sophisticated baseline geochemical code was required to provide the capability to initially and properly scope site-specific problems. The code MINTEQ (Felmy et al. 1984) was selected after comparing several existing codes: EQ3/EQ6 (Wolery 1979), developed at the Lawrence Berkley National Laboratory; EQUILIB (Morrey et al. 1978), developed at PNL for geothermal applications; PHREEQE (Parkhurst et al.

1980), developed at the U.S. Geochemical Survey; GEOCHEM (Sposito and Mattigod 1980), developed at the University of California at Riverside; and REDEQL.UMD (Harriss and Magnuson 1983), developed for the Environmental Protection Agency at the University of Minnesota. MINTEQ was chosen because it had a full complement of adsorption algorithms, was relatively easily structured, and had associated with it a carefully edited thermodynamic data base. It was developed at PNL by modifying the code MINEQL (Westall et al. 1976), originally developed at the Massachusetts Institute of Technology.

To be easily and reliably used, MINTEQ needed modifications to provide the following: 1) convergence reliability, 2) high-temperature competency, 3) high-ionic-strength competency, 4) conservation of the mass of water, 5) improved ease of use, 6) capability to model closed solid systems with finite quantities of soluble materials, 7) capability to model solid solutions, and 8) capability to create a site-specific data base to be used by the coupled geochemical code in modeling transport phenomena. Items 1, 3, 4, 5, and 6 were addressed before the program was terminated. Modifications substantially enhanced the performance of MINTEQ although not all had been thoroughly verified.

The Newton-Raphson method of convergence used in the original MINTEQ was unreliable unless initial estimates of activities of basic species were close to the equilibrium values. To correct this weakness, three additional methods of convergence were added to make initial estimates no longer necessary. One of these, the Marquardt method (Marquardt 1963), changes the magnitude of the change vector for the basic species by adding a constant to the diagonal of the Jacobian matrix formulated in the Newton-Raphson method. The

second is a back-substitution method on the independent species. It is operational when mass balance is greater than 20%. The third is a deDonder (deDonder 1936) shift algorithm that simultaneously adjusts all components in a chemical reaction. Though still being tested at the time of program termination, these methods had greatly improved convergence performance in the cases analyzed.

A code, PRODEF, was created to help users define problems by interactively responding to questions provided by the code. The code is designed to check logical and chemical errors which otherwise would render a solution meaningless. This will save time and funds in using MINTEQ.

Though high-ionic-strength competency was not scheduled to be achieved until FY 1985, it was added early to the new version of the baseline code (MINTEQ2). Currently a user has the option of designating Debye-Hückel (Westall et al. 1976), Davies (Davies 1963), or Pitzer (Pitzer and Kim 1973) methods of calculating activity coefficients for aqueous species.

With the deDonder (deDonder 1936) algorithm added, MINTEQ2 can now model problems that start with finite quantities of solids. Previously this was not possible with very soluble salts.

Thermodynamic Data Base

Thermodynamic data for important radionuclide aqueous species and solids (e.g., uranium, plutonium, and technetium) were carefully evaluated and made self-consistent. The MINTEQ geochemical code (Felmy, Girvin, and Jenne 1984; Felmy et al. 1984), the SOLUPLLOT code (Bethke 1978), and the MCRT code (T. J. Wolery, not yet documented) were used in this effort.

SOLUPLLOT was used to calculate equilibrium stability fields for predominant aqueous species and solids as a function of pH and redox conditions for a user-specified chemical system at a given temperature. The stability fields were calculated from an input data file that contained the Gibbs free energies of formation (ΔG_f° , 198) and heats of formation (ΔH_f° , 298) for the aqueous species and solids of interest. With the number of aqueous species and solids and their thermodynamic data in this input file altered, SOLUPLLOT graphically analyzed the relative importance and stability relationships among these different constituents.

Coupling Geochemical Code

A prototype streamlined geochemical code (CTMID) was assembled with several features not available in other prototype codes (e.g., MININR, Felmy, Reisenaur et al. 1984; FIESTA, Jennings et al. 1982; and CHMTRN, Miller and Benson 1983). It includes extended Debye-Hückel activity coefficient calculations, adsorption (Langmuir isotherm and the triple-layer model), mass balance on water, and solid-phase precipitation and dissolution on a large enough array of components to promise reasonable modeling capability. It uses a Markov transport model that is easily parameterized in terms of standard ground-water transport models. Scaling laws for this model have been derived.

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1.5 TANK CORROSION (AR-05-15-10 - 80681)

J. R. Divine - Program Manager

The primary objective of this study is to provide the operating personnel of the Hanford waste storage tanks with corrosion data to define acceptable, safe operating specifications for compositions, temperatures, and mixtures.

The secondary objectives are to attain an understanding of the corrosion mechanism in these solutions, to produce corrosion data under nominal storage conditions, to provide a basis for extrapolating from existing chemistries and temperatures, to acquire the data needed for use with new conditions, and to provide surveillance of the corrosion in the tanks.

Summary

The Tank Corrosion program has, over the past several years, examined the corrosion behavior of the tank construction materials in anticipated waste compositions at temperatures from 40°C to the design temperature of about 180°C.

Stress-corrosion cracking (SCC) has not been found to the extent forecast by work at Savannah River Laboratory. It has been observed only in dilute compositions and at high-caustic concentrations at high temperature.

The double-shell slurry compositions had statistically significantly increased corrosion rates with increasing temperature. Future PUREX compositions were statistically unaffected by temperature.

In both waste types the corrosion rate increased with increasing caustic concentration. At this time, temperatures above 140°C (284°F) are not recommended for hydroxide levels above about 7M, a limit that has relaxed slightly from that of earlier results. When the comprehensive data analysis is performed, some further relaxation of this limit is expected.

Background

When the environmental impact statement (EIS) was prepared for Hanford's 241-AN and AW double-shell waste storage tanks, the available data were garnered from the literature and Savannah River and Hanford sources. These data proved inadequate to describe the behavior of the wide range of waste compositions proposed for storage in the new tanks. It was concluded that, though further corrosion-rate development work was needed, the double-shell slurry specified in the EIS could be put in the tanks without additional testing, provided the temperatures were to be kept close to or below the proposed operating values.

Results

Earlier reports in this series described 12-month tests conducted with double-shell slurry and future PUREX wastes at temperatures of 40 to 100°C as well as 4-month tests at temperatures to 180°C. In the lower temperature range, corrosion rates were below the 1 mil/yr (0.001 in./yr) design allowance for all of the double-shell slurry compositions and most future PUREX compositions. The exceptions were dilute hydroxide/nitrate solutions. In these cases, severe corrosion, greater than 2 mil/yr with localized attack, occurred in the most dilute cases, 0.001M OH⁻ and 0.01M NO₂⁻. With slightly higher concentrations, about 0.2M OH⁻ and 0.1M NO₂⁻, SCC occurred. No SCC occurred in the double-shell slurry compositions.

In the higher-temperature tests, 140 and 180°C, the corrosion rates in future PUREX compositions showed, on the average, no significant temperature dependence after 12 months exposure. However, the interaction of temperature and composition was statistically significant, indicating that the effect of temperature is different for some compositions. For example, high-caustic compositions (5 to 9M OH⁻) have significantly higher corrosion rates at 180°C than at 140°C, while the opposite is true for low-caustic compositions (0.1 to 0.2M OH⁻). Other compositions (generally 0.2 to 5M OH⁻) show little difference in corrosion rates between the two temperatures.

The high-temperature, double-shell-slurry data showed significant effects of composition, temperature, and the temperature/composition interaction on corrosion rate. Tentatively, based on separate analyses of individual data sets, the corrosion rate exceeds the design limit for compositions

with 7M or greater hydroxide at temperatures above 140°C. It appears that these limits will relax somewhat after the complete data set is analyzed.

As is illustrated in Figures 1.26 and 1.27, the overall corrosion rates are decreasing with time. It is probable that high rates (>1 mil/yr) will continue to be seen for high-caustic mixtures close to the design temperature of the tanks (~180°C).

The final analysis of the 40 to 180°C double-shell-slurry/fission product data will be completed in November 1984. A formal summary report will be issued.

1.6 CESIUM CHLORIDE COMPATIBILITY (AR-05-15-10 - 80682)

G. H. Bryan - Program Manager

The objective of this study is to evaluate the compatibility of WESF-produced CsCl with 316L stainless steel capsule material under the thermal conditions that would be encountered in a geologic repository. A secondary effort involves a defining of the magnitude, severity, and origin of pitting found on the outer surface of some inner capsules.

Summary

A program was started in FY 1982 to evaluate the compatibility of WESF^(a)-produced cesium chloride (CsCl) with Type 316L stainless steel (SS) under the worst thermal conditions that will be encountered in a geologic repository. The principal objective of the program is compatibility-testing of six standard WESF capsules at a maximum metal/CsCl interface temperature of ~450°C. The capsules are placed vertically in insulated containers and allowed to self-heat to the

(a) The Waste Encapsulation and Storage Facility at the Hanford Site.

FIGURE 1.26. Corrosion Rate Distribution for Mixtures at 140°C

FIGURE 1.27. Corrosion Rate Distribution for Mixtures at 180°C

test temperature, where they are maintained for intervals from 2,200 to 32,000 hr. After thermal aging, the capsules are destructively examined to determine the extent of metal attack by the cesium chloride. The 17,544-hr capsule was examined in FY 1984.

During a prior examination, it was found that one of the aged test capsules exhibited discoloration and pitting on the outer surface of the inner capsule. The damage was attributed to problems encountered in the electropolishing operation, which is used to decontaminate the inner capsule after it is loaded with the CsCl and welded shut. A study was carried out in FY 1984 to 1) determine whether the pitting was associated with the electropolishing operation, 2) identify acceptable solution(s) to the problem, and 3) establish the effect, if any, on the capsule's long-term integrity.

Another special study performed in FY 1984 was that of examining two capsules from the WESF storage pool to establish the extent of corrosion in storage. This report summarizes program accomplishments in FY 1984.

Thermal Aging Tests - G. H. Bryan

Test capsule No. C-1486 was removed from its insulated container after being held at temperature for 17,544 hr. The time-averaged maximum metal/CsCl interface temperature for the test period was 454°C. The capsule was shipped to ORNL for sectioning and metallographic examination. ORNL completed the metallographic examination of that capsule as well as the 8784-hr test capsule (C-1451), which had been shipped to ORNL in FY 1983. Contrary to expectation, metal attack observed in the 8784-hr capsule was much less than that observed in the capsule tested for 4392 hr as well as the 17,544-hr capsule. The maximum attack in the 8784-hr capsule was

estimated to be 40 μm (0.0016 in.), whereas the maximum attack in the 17,544-hr capsule was estimated to be 260 μm (0.010 in.).

Although it is not unusual in compatibility-testing with radioactive materials to have wide variations in attack with replicate tests, the differences in the level of attack among the 8784-hr, the 17,544-hr and the 4382-hr capsules appear to be greater than variations normally encountered. A plot of the maximum metal attack observed in each test capsule as a function of exposure time (Figure 1.28) shows that the attack rate decreases with time, as expected. Additional data points are needed, however, to determine with more certainty the form of the corrosion rate equation; the data will be generated from capsules already under test.

It should be noted that the original basis for selecting the 450°C test temperature was a projected area/thermal loading limitation in a hypothetical repository.

WESF Pitting Study - J. R. Divine

Pits on the exterior surface of the inner WESF capsule were observed on four 316L SS capsules. These appeared to be associated with the contact points of the rack in the WESF electropolishing unit used for decontamination of the capsules.

The operating parameters for the electropolishing process at WESF were examined for their possible effect on capsule pitting. Within a reasonable range of conditions during normal operations, pitting is not expected. A chloride concentration of 100 ppm, more than believed to be reasonably present in the electropolishing bath, roughened the surface only slightly. Variations in temperature, current density, and time, and in concentrations of dissolved metal and phosphoric acid over the expected ranges were

FIGURE 1.28. Maximum Metal Attack as a Function of Exposure Time

also shown to be unlikely causes of pitting. The most likely cause of the observed pitting is believed to be the deterioration, with use, of the rack, which leads to a nonuniform distribution of current. The presence of point contacts between the capsule supports and the capsule leads to rapid localized dissolution of the capsule and the formation of pits.

The maximum pit depth achieved under worst-case laboratory conditions was about 30% of the capsule wall thickness. Nevertheless, though elimination of pitting is desirable, metallurgical examination and mechanical testing of laboratory-pitted specimens indicate that pitting is harmful only to the extent that it reduces the cross-sectional area of the capsule. The strength and ductility of the pitted specimens are

sufficient to ensure that the integrity of the WESF capsules is not threatened during service because of the pitting. No consideration was taken of any corrosion of the inner capsule surface by the cesium chloride.

Examination of WESF Pool Cell Capsules - G. H. Bryan

Two WESF cesium-chloride capsules were taken from the WESF storage pool and shipped to ORNL for sectioning and examination. Each capsule contained >70 kCi of ^{137}Cs at the time of loading. One capsule had been in the pool for 5 years and the other 8 years at the time of removal. Very little metal attack was observed in the stored capsules. Overall, attack observed in the storage capsules was about the same as that which occurs during

the capsule loading operation. This indicates that the capsules could be stored in a water pool for long periods without suffering significant attack.

1.7 HANFORD GROUT TECHNOLOGY

(AR-05-15-10 and AR-05-10-02 - 80760)

R. L. Treat - Program Manager

The objective of this program is to provide Rockwell Hanford Operations with technical support for disposing grouted low-level wastes in shallow-land sites.

Summary

Regulatory requirements applicable to grout disposal were reviewed. Chemical and radioactive waste species important to the long-term performance of disposed grouts were identified. A plan was prepared for disseminating information to the public on grouting at Hanford. Grout testing plans were prepared. The plans include the use of four large lysimeter test stations that were installed during the reporting period. Artificially aged grouts will also be tested. Permeability measurements of a simulated waste grout indicated that the grout is very impermeable. Analyses of potential water percolation rates at the grout disposal site indicated that 100 to 1,000 years are required for surface water to reach the water table. The shorter travel time would require irrigation of the site or a change to a wetter climate.

A plan for disposal of single-shell tanks by filling with grout was prepared. Chemical analyses of grout formers and a radioactive waste were performed. The analyses of grout formers highlighted the importance of careful analysis prior to use in grouting operations. Grout-flow tests were conducted to aid in the

design of a system for distributing grout in a large trench. A pump study was conducted to aid in selecting a grout pump for Hanford's grouting operation. A one-quarter-scale grout process was installed to test certain design features and to aid in establishing operating specifications.

Regulatory Requirements - S. L. Stein

Regulatory requirements that may be applicable to grouting at Hanford were reviewed and will continue to be reviewed throughout the program. Requirements reviewed include those contained in 1) DOE Order 5820.2 for Management of Radioactive Wastes, 2) Washington State Dangerous Waste Regulations (173-202-WAC), 3) a Memorandum of Understanding (MOU) between DOE and EPA regarding management and disposal of nonradioactive hazardous wastes, and 4) a MOU between DOE and EPA regarding implementation of the Comprehensive Environmental Response Compensation and Liability Act (CERCLA) of 1980. Interpretation of the requirements is continuing.

Performance Assessment - S. L. Stein

Candidate waste streams for grouting were examined in light of applicable regulations and potential grout-performance requirements. This examination revealed that, in protecting the ground water, the following waste components are of primary importance:

radionuclides: ^{99}Tc , ^{129}I , ^{14}C , and

actinides

chemical wastes: nitrates, chromates,

xylene salts, and fluorides.

A broader analysis of candidate waste streams for grouting may result in identifying other chemical wastes that must be controlled.

Public Information Planning - S. L. Stein and
E. M. Toomey

A plan was prepared for disseminating information to the public on grouting wastes at Hanford. The plan will serve as input to the Hanford External Affairs planning effort managed by Rockwell Hanford Operations. The following elements of public information planning were addressed:

- identification of interested parties
- public perception
- questions the public will be likely to ask concerning the grout technology
- recommended points for public interaction
- recommended communications media.

Answers were also provided to the 30 questions identified as likely to be asked by the public.

Grout Testing Plans - R. O. Lokken,
R. L. Treat, D. M. Strachan, and
A. P. Toste

The data required for assessing long-term grout performance were identified. They include grout release rate information as well as data required to determine waste migration rates in Hanford soil. Testing plans include the use of lysimeters, soil columns, and leachability and permeability testing methods. Some grouts will be artificially aged by exposure to elevated temperatures and carbonation. The performance of aged grouts will be compared to that of fresh grouts. The potential effects of organics present in candidate waste streams for grouting are thought to be acceptable. A thorough characterization of organics in grout feed streams is advised, however.

Lysimeter Testing - G. V. Last

Four lysimeter test stations were installed and will be filled with representative Hanford soil and radioactive grouts. They will provide an environment for testing the performance of grouts under realistic disposal conditions. Each lysimeter test station was constructed of 6-ft-dia culvert pipe with one end capped with steel plate. The test stations were buried vertically near an existing lysimeter test site. Soil and radioactive grouts will be added to the test stations during March 1985.

Grout Permeability - D. M. Strachan

Grout permeability measurements were initiated to support assessments of long-term grout performance. A specimen of grout, made from Phosphate Hanford Facilities Waste and a dry mix of 50 wt% Centralia fly ash, 40% type I and II Portland cement, and 10 wt% attapul-gite 150 clay at a mix ratio of 6 lb/gal, was cured for 4 months under 100% relative humidity. It was then placed in the permeability apparatus to check for liquid extraction as a function of pressure and for its permeability. No liquid was extracted even after exerting 110 psi. The permeability was found to be just above the detection limit ($1.8 \times 10^{-11} \text{ m}^2$), indicating that the grout is very impermeable.

Analysis of Hanford Soils - A. E. Reisenauer

Soil characteristics were measured on six distinctly different samples of earth collected at various depths near the proposed grout disposal site. Characteristics measured included particle size, saturated

hydraulic conductivity, and water-retention properties. For study purposes, the soils existed in layers in six different configurations above the water table. Water travel times to the water table were then calculated for each configuration, using water recharge rates of 5.0 and 0.5 cm/yr. For a recharge rate of 5.0 cm/yr, percolating water was calculated to reach the water table in 99 to 149 years. For a 0.5 cm/yr recharge rate, 739 to 982 years were required. These data support the assessment of migration rates of wastes released from grout to the surrounding soil.

Single-Shell Tank Disposal Plan - R. L. Treat

A plan was prepared for the in situ disposal of single-shell tanks by filling the void spaces with waste grout. The plan serves as a contingency for an earlier plan for the demonstration of disposal of tanks in TY Tank Farm by backfilling with gravel. The estimated cost for the grout-filling demonstration is 11 million dollars. This assumed that the six tanks in TY Tank Farm must first be partially filled with gravel to stabilize existing wastes, which would leave 2.7 million gallons of void space in the tanks for grout.

Chemical Analyses - R. O. Lokken and J. L. Swanson

Chemical analyses were performed on grout formers and on actual Phosphate Hanford Facilities Waste (PHFW), a major component of the first waste stream that is scheduled for grouting. An analysis of the composition of a sample of fly ash, one of the grout-forming solids, indicated that it contained a substantial fraction of high-calcium particulates. Follow-up communications with the

supplier revealed that pulverized limestone (calcium carbonate) is handled in the same equipment used to process fly ash. An analysis of a sample of attapulgite clay, another grout-forming solid, revealed that it was so much better as a grout-thickening agent that its use could lead to pumping problems if added in the same proportion as the attapulgite clay tested earlier. These findings demonstrate that careful analyses of grout-forming solids will be required before they are used for grouting operations. Analyses of the radioactive PHFW showed that the waste is nontransuranic, contains primarily activation products, is composed largely of phosphates, citrates, and sulphates, and contains iron as its dominant metallic species.

Grout-Flow Testing - R. L. Treat and R. O. Lokken

Grout-flow tests were conducted to enable resolution of certain operating issues. Major conclusions reached through these tests included: 1) the angle of flow of grout in a large trench will be greater than 3° but less than 30°, 2) the liquid waste and individual solids that comprise the grout-formers blend will not segregate appreciably as the grout flows, and 3) the time required for grout to set increases as a function of flow distance. Additional grout-flow tests are planned to aid the design of a system for distributing grout in a large trench.

Pump Study - D. McCarthy

A comparative study of candidate pumps for grouting was conducted to provide guidance to Rockwell for selecting a pump for the Transportable Grout Facility (TGF). The study focused on progressive-cavity pumps and

reciprocating pumps, primarily of the piston and plunger designs. Twenty-eight pump vendors were contacted for information that would permit making a comparison. It was concluded that the progressive-cavity pump is probably the better choice, so long as assurance is given that the pumping head can be maintained below 350 psi.

Quarter-Scale Grout Process - R. L. Treat,
M. K. W. Chan, and D. McCarthy

A process was installed that can produce simulated grouts at one-quarter the rate that is planned for the Transportable Grout Facility. The TGF is currently under construction for grouting Hanford wastes, and the quarter-scale process will be used to test certain design features and to aid in establishing its operating specifications. The quarter-scale grout process consists of a simulated liquid waste feed system, a grout solids feed system, a grout mixer, a grout pump, and 400 ft of grout transfer line.

1.8 REDUCED WASTE GENERATION -
ADMINISTRATIVE SORTING
(AR-05-20 - 80761)

R. P. Allen - Program Manager

The objective of this program is to provide analytical data for ways in which newly generated waste can be changed from the transuranic (TRU) waste category into the low-level waste (LLW) category.

Summary

A study was conducted to evaluate the technical and economic feasibility of using in situ decontamination techniques to convert glove boxes and other large TRU-contaminated components directly into LLW. The results of the technical evaluation indicate that

in situ decontamination of these types of components to non-TRU levels is technically feasible. Applicable decontamination techniques include electropolishing, hand-scrubbing, chemical washes/sprays, strippable coatings, and Freon® spray-cleaning. The removal of contamination from crevices and other holdup areas remains a problem, but may be solved through further advances in decontamination technology. Moreover, the increase in allowable TRU levels reduces the removal requirement and facilitates measurement of the remaining quantities.

The major emphasis of the study was on a cost/benefit evaluation that included a review and update of previous analyses and evaluations of TRU-waste volume reduction and conversion options (Brown 1982; Chikalla and Powell 1982). The results of the economic evaluation show, for the assumptions used, that there is a definite cost incentive to size-reduce large components and that decontamination of sectioned material has become cost-competitive with the size-reduction options. In situ decontamination appears to be the lowest cost option when based on routine-type operations conducted by well-trained and properly equipped personnel.

Introduction

A study conducted at PNL (Chikalla and Powell 1982) indicated that the size reduction of glove boxes and other large TRU-contaminated components could yield substantial savings in packaging, storage, retrieval, transportation and WIPP-disposal costs for Hanford waste. Decontamination of the sectioned waste to non-TRU levels using

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vibratory finishing (McCoy, Arrowsmith and Allen 1980) was included as part of this study. It was not, however, the lowest-cost treatment option because of the relatively high cost of sectioning material into the small size required for vibratory finishing.

One way of reducing decontamination costs to make it a more cost-competitive option is to eliminate the sectioning operation by using in situ decontamination techniques to convert components directly to non-TRU waste. This approach has been explored in the past, but not used extensively, primarily because of the difficulty of adequately decontaminating and monitoring crevices, gasket seals and other potential contamination holdup areas under the 10 nCi/g definition of TRU waste.

Reconsideration of this direct conversion approach is warranted by two recent developments. One is the tenfold increase in the levels defining TRU waste to >100 nCi/g as specified in DOE Order 5820.2. This change should substantially reduce the extent and cost of decontamination required to achieve non-TRU levels and also help alleviate the component monitoring problem.

The second development is the availability of new and potentially more effective in situ decontamination techniques. These include in situ electropolishing, high-pressure Freon spray-cleaning, and strippable decontamination coatings including a new "self-stripping" formulation.

Based on these considerations, a study was conducted to reassess the technical and economic feasibility of using in situ decontamination techniques to convert components directly into LLW. This report summarizes the results of this study. The first section addresses the technical feasibility of the direct conversion approach. The second section focuses on economic feasibility and in-

cludes cost/benefit evaluations and comparisons with other current and projected TRU waste handling, processing and disposal options.

Technical Evaluation

The following techniques were assessed for their suitability and effectiveness for in situ decontamination applications. Selected experimental studies were conducted for the Freon spray-cleaning and self-strip coating methods to provide evaluation information not otherwise available.

- Hand-scrubbing - Hand-scrubbing and related manual-type decontamination operations are commonly used to remove gross contamination to prepare glove boxes for storage or as a precursor to other decontamination operations. A variety of cleaning/scouring materials and chemical cleaning agents can be employed to match the specific decontamination requirement. Removal of contamination from crevices and other holdup areas can be enhanced by using foaming-type cleaners. Although hand-scrubbing is labor intensive, it can be a cost-effective in situ technique and generate minimal secondary waste if properly employed by well-trained personnel.
- Chemical washes/sprays - Aggressive chemicals applied as washes or sprays have been used both to pretreat and to decontaminate glove boxes in place. Studies conducted at the Savannah River Laboratory (Crawford 1978) have demonstrated that stainless steel can be decontaminated to non-TRU levels using a two-step process that employs alkaline permanganate to extract chromium from the protective oxide film on the stainless, followed by application of oxalic acid to complete the removal of the

modified film. This approach has been further developed (Wobser 1983) by substituting acidic permanganate for the alkaline permanganate and by increasing contact times and temperatures.

- In situ electropolishing - Previous tests (Platt 1979) have shown that in situ electropolishing can readily remove TRU contamination from accessible metallic surfaces. The highly contaminated floor of a heavily used plutonium glove box, for example, was readily reduced from 1100 nCi/g to 0.2 nCi/g using a simple in situ electropolishing device. Since electropolishing is applicable only to metallic surfaces, it must be used in conjunction with other techniques to decontaminate the non-metallic areas. Although electropolishing exhibits good throwing power, decontaminating cracks and other constricted holdup areas also would be a problem.
- High-pressure Freon cleaning - The results of tests conducted to evaluate high-pressure Freon spray-cleaning as an in situ decontamination technique for the conversion of TRU wastes to LLW have been reported (McElroy and Powell 1984). Freon cleaning has been effective in removing smearable contamination for power reactor applications. However, the average decontamination factors for removal of smearable plutonium contamination from representative glove box interior surfaces were less than 10, as compared with 200 for scrubbing with a commercial cleaner. The Freon spray does have a unique ability to penetrate and dislodge contamination in holdup areas, but dispersion and redeposition of the removed contamination is a problem. Work has been initiated by Freon system manufacturers to add complexing agents to the Freon to improve the collection and transport of actinides. If suc-

cessful, this approach could substantially increase the effectiveness of the high-pressure Freon spray technology for in situ operations, and particularly for decontamination of holdup areas.

- Strippable coatings - A number of strip-pable coating formulations have been developed to immobilize and incorporate smearable contamination. This approach has been used successfully for the in situ removal of gross contamination from the inside of glove boxes. The coatings are generally applied using an airless sprayer to minimize labor and exposure. Maintaining the required coating thickness to permit ready stripping after curing is essential; otherwise, removal by scraping is required. Recently, a self-stripping coating was developed that can reduce the time and exposure associated with stripping. When the coating cures, it develops sufficient internal stress to self-spall. The coating fragments can be collected by vacuuming. The cured coating also is water-soluble and can be removed by scrubbing for areas that fail to spall. Decontamination tests using this material indicate good removal of gross smearable contamination and also a modest effect on fixed contamination. No essential difference in decontamination effectiveness was noted for coatings removed by self-stripping as compared with thinner coatings removed by wiping with a wet cloth. The effectiveness of these coatings in removing contamination from holdup areas still requires investigation.

Based on an evaluation of these available in situ decontamination techniques, it is concluded that in situ decontamination of glove boxes and other components to non-TRU levels, particularly under the revised definition of TRU waste, is technically feasible.

Electropolishing techniques have the demonstrated capability to reduce contamination levels even on corroded metallic surfaces to nondetectable levels. Hand scrubbing, chemical washes/sprays, and strippable coatings potentially can clean accessible surfaces to the required levels, although multiple applications may be required. The removal of contamination from holdup areas remains a problem. However, the further development of the high-pressure Freon cleaning technology may provide a solution.

Verifying the in situ decontamination of components to non-TRU levels is potentially more difficult than the actual decontamination operation because of the inaccessibility of the residual contamination. This problem also has been partially alleviated by the increase in allowable TRU levels. Calculations have been performed showing that the amount of contamination remaining in the crevice and gasket regions of a representative 100-nCi/g glove box, assuming decontamination of accessible surfaces to <10 nCi/g, would be readily detectable using a collimated detector through comparison with adjacent cleaned areas.

Economic Evaluation

Cost data have been derived to permit comparison of the in situ TRU waste conversion approach with other handling, processing and disposal options. These comparison options include the following:

1. Direct packaging of glove boxes in the largest standard WIPP-approved container with dimensions 2.8 m (L) x 1.7 m (W) x 1.8 m (H). This decreases waste packaging density from the original 170 kg/m³ to 100 kg/m³.
2. Size reduction of glove boxes for placement in containers 1.7 m (L) x 1.4 m (W) x

0.97 m (H). This increases waste packing density to 350 kg/m³.

3. Additional sectioning to permit disposal in 55-gal drums handled as a six-pack. This increases the waste density to 660 kg/m³.
4. Additional sectioning followed by decontamination to non-TRU levels using vibratory finishing. For each original 2.8 m³ of TRU waste, this produces 0.76 m³ of LLW and 0.11 m³ of secondary TRU waste, including the volume of unprocessed HEPA filters (Chikalla and Powell 1982).

The cost elements considered, based primarily on Hanford data, are as follows:

- containers
- certification
- interim storage
- retrieval
- transportation to WIPP
- WIPP emplacement
- size reduction
- decontamination
- LLW disposal.

The cost data and estimates for the in situ decontamination of glove boxes are based on actual time/cost data for the preparation and manual decontamination of three large glove boxes; a review of previous in situ decontamination operations using other techniques; and discussions with other DOE site and contractor personnel conducting operations.

The cost comparison data are summarized in Table 1.8. Costs are all normalized in terms of \$/m³ of original waste volume. The results show a definite cost incentive to size-reduce large components and that the decontamination of sectioned material is now cost competitive with the size-reduction option for the assumptions used for this evaluation. In situ decontamination appears to

TABLE 1.8. Comparison of Unit Costs^(a) for Various Handling, Processing and Disposal Options^(b) for Representative TRU-Contaminated Components

Cost Element	Case 1	Case 2	Case 3	Case 4	Case 5
Containers	738	170	85	75	
Certification	57	53	209	30	
Storage	1830	463	237	10	
Retrieval	3530	893	459	18	
Transportation ^(c)	1100	300	350	14	
WIPP disposal	1550	431	326	13	
Size reduction	46	388	2080		
Decontamination				2450 ^(d)	1900 ^(e)
LLW Disposal				47	
TOTAL (rounded)	8900	2700	3700	2700	1900

(a) In 1984 \$/m³ of original waste volumes; excludes capital and facility maintenance costs.

(b) Case 1 - Direct packaging in large container.
Case 2 - Sectioning for disposal in intermediate-size container.
Case 3 - Sectioning for disposal in 55-gal drums.
Case 4 - Decontamination of sectioned waste.
Case 5 - In situ decontamination of original components.

(c) Truck transport to WIPP from Hanford.

(d) Includes sectioning costs.

(e) Includes component preparation, post-decontamination verification monitoring, and secondary waste handling and disposal costs.

be the lowest cost option based on routine-type operations conducted using well-trained, experienced personnel. A number of factors, however, related both to the uniqueness of each decontamination project and to externally imposed constraints and requirements could alter this conclusion for particular sites or waste conversion applications.

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2.0 NUCLEAR WASTE MATERIALS CHARACTERIZATION CENTER

The objective of the Nuclear Waste Materials Characterization Center (MCC) is to provide an authoritative 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 interactions with each other, and the effects of their environment. The MCC works with waste-form producers and repository developers to standardize test methods and obtain materials data 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.

2.0 NUCLEAR WASTE MATERIALS CHARACTERIZATION CENTER (MCC)
(AR-05-15-10/DB-60-20-20/AR-05-15-30 - 80145)

J. E. Mendel - Program Manager

SUMMARY

Radioactive testing of the MCC-14.4 BWIP^(a) Compliance Test on Radionuclide Release for Determining Acceptance of a Loaded Waste Form was started in September using ATM-4 (Pu-doped 76-68 glass). The test method was also reviewed by the Office of the Chairman of the Materials Review Board (OTC/MRB), and the resulting comments are being incorporated into the test method. The MCC-105.1 Test Method for Long-Term Corrosion in Repository Environments (Basalt Static System) is undergoing final BWIP review before being submitted to the MRB. An approximately 100-page MCC-105.1 Benchmark Test Report has also been prepared and will be submitted to the MRB after BWIP review.

Three new doped-glass Approved Testing Materials (ATMs) were prepared during the reporting period. A sequential scanner was installed on the MCC's shielded ICP to increase the flexibility of the instrument. Characterization of LWR Spent Fuel MCC-Approved Testing Materials - ATM-101, PNL-5109, by J. O. Barner was issued.

The MCC's two original static leach test methods, which have had provisional approval by the MRB for several years, were submitted for full approval. Two data packages of MCC-1 leach data (MCC-D1 and D2) have been presented to the MRB. MCC-D1 was approved by the full MRB and MCC-D2 was approved by the Data Panel of the MRB in this reporting period.

(a) Basalt Waste Isolation Project.

A new test method, MCC-15 Waste/Canister Accident Testing and Analysis, was formulated and is undergoing internal review prior to forwarding to the Transportation Technology Center (TTC) for comments.

The 3-year Leaching Mechanisms Program, in which the activities of six subcontractors were coordinated, finished on schedule. The Final Report of the Defense High-Level Waste Leaching Mechanisms Program, PNL-5157, was issued in August. A 2-day workshop was held in Germantown, Maryland, September 18-19, to wrap up the program.

An MCC Workshop on Nuclear Waste Repository Aging Processes was held in Seattle, Washington, June 12-14.

Draft Statements of Work were prepared describing the MCC's planned "as-requested" activities in support of the BWIP and Office of Nuclear Waste Isolation (ONWI) waste package programs in FY 1985. R. D. Allen was recruited to head a new MCC Task 5, Test Method Submissions, in FY 1985.

WASTE MATERIALS INTERACTIONS -

R. G. Strickert (Task 1 Leader) -
R. L. Erikson, R. W. Fulton, K. M. Krupka,
J. W. Shade, and R. W. Stephens

MCC-14.4 Waste-Form Compliance Test Method Development

The MCC has collected and developed a set of procedures for a materials interactions test method at the request of the BWIP and in support of the BWIP task to develop waste

acceptance requirements and specifications-- The BWIP Compliance Test on Radionuclide Release for Determining Acceptance of a Loaded Waste Form (MCC-14.4). The purpose of this test is to measure steady-state radionuclide concentrations in solutions after hydrothermal interactions of a synthetic basalt ground water, basalt rock, and a radioactive waste-form material.

Measured portions of a crushed and sized waste-form material are combined with crushed and sized reference basalt and immersed in synthetic basalt ground-water solutions. The suspensions are agitated in relatively inert reaction containers at 150°C for up to 3 months. Selected elemental and radio-isotopic analyses are performed on filtered and unfiltered solution aliquots from each container. The de-aeration of the synthetic basalt ground water, loading and sealing of the reaction vessels, and final sampling of the test suspensions are conducted in an inert gas (argon) glove box to eliminate, as much as practicable, the effect of atmospheric oxygen on the solution redox potential.

The steady-state radionuclide concentrations determined in this test method do not, in themselves, indicate any equilibrium solubility-limited reaction, because measured steady-state concentrations include both solution and colloidal species and are not approached from both undersaturated and oversaturated conditions. Furthermore, the test method does not provide procedures for identifying solid (stable or metastable) reaction products.

Following review and concurrence by the BWIP, the MCC-14.4 Compliance Test Method was submitted in July 1984 to the Office of the Chairman/Materials Review Board (OTC/MRB). An appendix to MCC-14.4 identifies the factors that will affect the approval status of

MCC-14.4. These factors include the time necessary to reach steady-state conditions, quench effects, suitability of materials in high-radiation fields, container decontamination, tolerance limits for solution analyses, and benchmark test data for determining the precision and accuracy of the method. An initial review of the test method and related issues was received from the OTC/MRB in August. In September MCC staff met with members of the BWIP staff to discuss appropriate responses to the OTC/MRB review of MCC-14.4. The changes and modifications recommended will be incorporated into a revised MCC-14.4 test method, which is to be resubmitted in December 1984, following review and concurrence by the BWIP.

During the current reporting period, modifications to rotator ovens used in these experiments were completed and the oven hoods were fabricated and installed in the lab. A set of reaction vessel leak tests at 150° and 200°C using Teflon® and aluminum/asbestos gaskets and synthetic (GR-4) basalt ground water were completed for periods up to 1 month. None of the vessels having Teflon seals leaked; however, two of four vessels with aluminum/asbestos seals leaked noticeably (3-30 weight percent loss of liquid over 3 weeks). The aluminum/asbestos seals were badly corroded and coated with a white precipitate. The precipitate was identified as a mixture of poorly crystallized boehmite (gamma-AlOOH) and calcite. Apparently the aluminum corroded in the alkaline GR-4 ground water, and the corroded surface provided nucleation sites for the precipitation of calcite. No precipitation was observed in tests with Teflon seals.

Washing of the Cohasset basalt and the crushed and sized plutonium-doped glass was completed. The results from the ultrasonic

cleaning of the basalt indicate that ultrasonic cleaning was not really necessary to remove the fine material. The procedure adopted involved washing the basalt with distilled water on a 230-mesh sieve. The basalt was dried at 60°C for 24 hr and stored in the argon chamber. The plutonium-doped glass was washed in distilled water in an ultrasonic bath for 5 min, the supernatant was decanted, and the glass was dried.

The first MCC-14.4 waste-form compliance test was initiated in September. Ten titanium pressure vessels were loaded with a 1:1 mixture of ATM-4 (plutonium-doped 76-68 glass) and Cohasset basalt. The GR-4 synthetic basalt ground water was added to obtain a solution-volume-to-solids-mass ratio of 10:1. The vessels were sealed with Teflon gaskets and placed in a rotator oven at 150°C for durations of 3, 6, 9, 12, and 15 weeks. During the first 2 weeks, the vessels were weighed on 3 different days to determine whether there was any leakage; no significant weight changes were observed for any vessels. The first two vessels (duplicates) are to be dismantled and sampled during the first week in October.

Aliquots of the GR-4 ground water made up for the MCC-14.4 test (discussed below) were submitted to two different analysts for ICP/IC analyses. The results from both analysts were in general agreement for all constituents except potassium. The concentrations of calcium and silicon were not within the 5% tolerance limit of their respective concentrations for the theoretical GR-4 composition. The cause of the calcium and silicon discrepancies is being investigated.

In addition to MCC-14.4 testing with plutonium-doped glass, experiments with uranium-doped borosilicate glass (PNL-76-68) have been conducted throughout FY 1984. In some of these tests, conditions (e.g., tempera-

ture, ground-water composition, etc.) have been altered in order to measure the results of such changes on observed elemental solution concentrations.

Analyses from previously completed experiments using uranium-doped glass/basalt suspensions in deionized (DI) water (discussed in the prior report in this series) were compared with results from an MCC-3 glass-only test in DI water. In the glass/basalt test equal amounts of glass and basalt (of the same mesh size) were used; the total SA/V (670 cm^{-1}) was the same in both tests.

The pH values (at room temperature) of the glass/rock suspensions heated to 90°C for up to 91 days ranged from 9.4 to 9.6. Total uranium concentrations measured in 0.45- μm filtrates from the 90°C glass/basalt system were 3 ppm at 90 days. However, uranium concentrations in 1.8- μm filtrates were ~0.5 ppm and appeared to have reached steady state. The difference in measured uranium concentrations in the 0.45- μm and 1.8- μm filtrates suggests that the bulk of the uranium released from the waste glass is in colloidal form.

In other glass/rock interactions tests, various ratios (1:1, 1:10, and 1:100) of uranium-doped glass and Umtanum basalt were added to a synthetic GR-4 ground water at 90° and 150°C. In addition, a rocking autoclave was set up containing a suspension of equal amounts of the uranium-doped glass and basalt in GR-4 ground water at 150°C. These materials interactions tests were designed to test the effects of various conditions pertaining to the MCC-14.4 test method. They have been completed, and the data are currently being analyzed.

Preliminary analyses of solutions from the 150°C rocking autoclave containing uranium-doped glass, basalt and GR-4 synthetic basalt ground water indicate that between 150°C and

25°C, the quench effect on the measured pH (a 0.3 pH unit increase) was similar to that observed in the glass/ground-water test. However, the addition of basalt to the system resulted in lower pH values (for solutions sampled at temperature and measured at room temperature) by 0.5 to 0.7 pH units. In the case of colloidal or dissolved silica, the presence of basalt lowered the pH (to about 9.6), which in turn reduced silica saturation relative to the glass-only experiment.

From the 150°C rocking autoclave experiment, uranium concentrations for nonfiltered aliquots and 0.45- μ m filtrates were about 2-3 ppm and for 1.8- μ m filtrates were 0.5 to 1 ppm. No significant quench effect was observed for uranium. The results suggest that the bulk of the uranium released from the waste glass is in colloidal form (as observed in the 90°C glass/basalt test with deionized water). Further detailed results will be forthcoming in reports to be prepared in FY 1985.

Solubility-Controlled Concentration Data for Radioactive Elements in Nuclear Waste

Accurate thermochemical data for aqueous complexation constants and solubility products are needed to predict the geochemical behavior of nuclear waste elements (e.g., actinides) in surface-water and ground-water systems. The solubility of U(VI) hydroxide is considered to have an important role in constraining the maximum possible concentrations of uranium dissolved in oxidizing to moderately reducing waters (Allard 1982; Wood and Rai 1981; Thompson, Dove and Krupka 1984). Solid U(VI) hydroxide has been identified as an oxidation product in dissolution studies of crystalline UO_2 (Wang 1981) and as a possible precipitate in leaching studies of

irradiated UO_2 fuel (Vandergraff 1980) in air-saturated solutions.

The MCC-3S Agitated Powder Leach Test Method (MCC 1984) was used, with minor modifications, to study the solubilities of U(VI) hydroxide ($\text{UO}_3 \cdot 2\text{H}_2\text{O}$, schoepite). The solubility of U(VI) hydroxide was investigated from both under- and oversaturation at 25°C in carbonate-free, oxidizing aqueous solutions in the pH range of 3 to 11. Details of the experimental techniques, including solution- and solid-phase characterization, are described in Krupka et al. (1984).

The concentrations of dissolved uranium measured in the U(VI) hydroxide solubility experiments are shown in Figure 2.1. The random distribution of uranium concentrations for the first sampling period (solid symbols in Figure 2.1) of the under- and over-saturated experiments indicates that the

FIGURE 2.1. Measured Concentrations of Uranium from 1.8- μ m Filtered Solutions Contacting U(VI) Hydroxide at 25°C. The solid and dashed lines represent the theoretical solubility calculated for schoepite ($\text{UO}_3 \cdot 2\text{H}_2\text{O}$) using the thermodynamic data reported, respectively, by Sylva and Davidson (1979) and Lemire and Tremaine (1980), and by Langmuir (1978).

uranium suspensions had attained solubility-equilibrium conditions within 2 weeks. The increased solubility observed for the U(VI) hydroxide solubility under basic conditions confirmed the amphoteric behavior for dissolved hexavalent uranium. In the near-neutral pH region, the oversaturated experiments (circle symbols) produced significantly higher uranium concentrations than those measured for the undersaturated experiments (square symbols).

Schoepite ($\text{UO}_3 \cdot 2\text{H}_2\text{O}$) was identified in the run products for the undersaturated experiments at $\text{pH} < 11$. At $\text{pH} > 11$, the x-ray diffraction (XRD) pattern for material from the undersaturated experiments could not be identified using computerized search techniques and did not resemble any of the other patterns collected in our study.

In the oversaturated experiments, tetramethylammonium perchlorate was the only solid-reaction product identified from the XRD data. Energy dispersive x-ray analyses using a scanning electron microscope showed that the solid-reaction products from the oversaturated experiments included submicron-sized solid material that contained uranium. The absence of detectable XRD peaks for a U(VI) solid may indicate that the solubility-controlling uranium solid phase is amorphous or that its low mass in these samples relative to that of tetramethylammonium perchlorate precluded its detection by XRD. However, the similarity in the uranium concentrations measured for the acidic and basic solutions suggests that the same uranium solubility control exists for both the under- and oversaturated experiments. The identity of the U(VI)-solubility control in the oversaturated experiments and the $\text{pH} > 11$ undersaturated experiments, as well as the increased scatter observed in the analytical

results for the near-neutral solutions, are still under investigation (see Figure 2.1).

Our experimental results for the solubility of schoepite at 25°C are in good agreement with the solubility calculated for schoepite from the thermodynamic data tabulated in Sylva and Davidson (1979) and Lemire and Tremaine (1980) (solid line in Figure 2.1). Our results also demonstrate the importance of including anionic U(VI) hydroxyl complexes [e.g., $(\text{UO}_2)_3(\text{OH})_7^-$] in speciation/solubility calculations for U(VI) in carbonate-free waters under basic conditions. For example, the solubility calculated for schoepite from the thermodynamic data in Langmuir (1978) (dashed line in Figure 2.1), who does not include an anionic U(VI) hydroxyl complex, is significantly lower than that observed in our experiments at $\text{pH} > 7$. Further discussions on the experimental data and comparisons with theoretical schoepite solubility calculated with the MINTEQ geochemical code (Felmy et al. 1984) from published uranium thermodynamic data are presented in Krupka et al. (1984).

Solubility experiments with U(VI) hydroxide in carbonate-free aqueous solutions at 70°C and pH values of 4, 7, 9, 10, and 12 were also begun in July. These 70°C experiments are designed to approach the solubility of U(VI) hydroxide from undersaturation (via dissolution) and oversaturation (i.e., precipitation). Solutions were taken from these experiments and analyzed for pH and dissolved uranium concentration. Samples of solid material taken from the 70°C under- and oversaturated experiments were submitted for characterization by x-ray diffraction analysis and scanning electron microscopy. Analytical results from the under- and oversaturated experiments are being used for

aqueous speciation and saturation index calculations using the MINTEQ geochemical code; these results will be reported in early FY 1985 and prepared for journal publication.

Details of the solubility study of Np(IV) hydrous oxide in fluoride solutions (discussed in the previous semiannual report) are given in Krupka et al. (1984). The results of the study show that up to 100 ppm fluoride does not have a measurable effect on the solubility of Np(IV) hydrous oxide under reducing, alkaline conditions, contrary to previous observations by Cleveland et al. (1983) and Rees et al. (1983).

CORROSION - M. D. Merz (Task 2 Leader) -
R. Wang and W. E. Anderson

Test methods are being developed and validated for evaluating the corrosion of metallic container and canister materials in nuclear waste repository environments. The methods will be applicable to basalt, salt, tuff and crystalline rock repositories. The MCC program is emphasizing the development of methods that are consistent with the test plans of the waste-package programs for the various repositories and will provide a basis for establishing reproducibility of corrosion tests and statistical statements on the precision of corrosion data.

The MCC began benchmark tests for corrosion in basalt earlier this year and has continued this testing. Tests for 30 and 120 days at 100°C were completed, and the MCC-105.1 Benchmark Test Report was prepared and will be reviewed by the BWIP in October 1984. This test report will provide supportive evidence for obtaining MRB approval of MCC-105.1.

A presentation and publication of MCC activities on corrosion was cleared for the Materials Research Society Meeting in

November 1984. Highlights of the publication, "Test Methods to Predict Long-Term Corrosion of Container Materials in Repositories," are presented below under MCC-105.1.

M. D. Merz and D. W. Shannon presented reviews of corrosion mechanisms at the MCC Workshop on Nuclear Waste Repository Aging Processes to address the importance of aging techniques in corrosion testing.

Test Method Development

The status of MCC corrosion test methods was summarized in the prior report in this series; the status of MCC-101, -102, -103P, -104, and -105.2 remains unchanged from the last report. The status of the other methods is summarized below.

MCC-105 Test Method for Long-Term Corrosion in Repository Environments

Test methods are being developed and codified for basalt, salt and tuff. Six test methods have been identified, MCC-105.1 through MCC-105.6, for application to three types of geologic media. Three of the test methods, two for basalt and one for salt, have been drafted and reviewed by BWIP and ONWI waste-package investigators. Methods for tuff are still in a preliminary stage. The Office of Crystalline Repository Development (OCD) program has not initiated any corrosion testing yet. The MCC has initiated benchmark testing for the basalt static test, MCC-105.1.

MCC-105.1 Basalt Static System. The benchmark tests according to initial drafts of this test method were completed for 30 and 120 days at 100°C. As a result of test results on the 30-day tests, the MCC revised the procedure for loading specimens to ensure

better contact between the crushed basalt + bentonite packing material and the specimen surfaces. The average penetration rate correlated with apparent area of contact; the change in loading procedure is expected to improve the precision of the test method. The data were analyzed for variations among specimens and among vessels. For the 120-day tests, the within-vessel specimen-to-specimen relative standard deviation (RSD) was about 10%, the vessel-to-vessel RSD was about 6%, and the overall specimen-to-specimen RSD was about 11%. The overall average uniform penetration rates for 100°C are shown in Figure 2.2.

MCC-105.3 Salt-Limited Reactants System.

Further development of this salt-system static test is awaiting decisions by ONWI.

The MCC purchased several titanium vessels in anticipation of later development of this test method.

MCC-105.4 Basalt Flowby System. The MCC codified this test method during the previous reporting period. Further development of the test method depends on initiation of experimental activities to prove out the concept of placing corrosion specimens in a packet that limits water access and simulates long-term, postclosure conditions in a basalt repository. The MCC plans to initiate testing in early FY 1985 and to submit the test method to the MRB in FY 1985.

MCC-105.5 Basalt Air-Stream System.

Investigators for the BWIP have requested the MCC to codify a test method wherein the

FIGURE 2.2. Plot of Average Penetration Rate ($\times 10^3$ mm/yr) Versus Time (days) for 100°C Tests. Data points are the average of the penetration rates of all specimens, and the error bar is one standard deviation for individual specimens above and below the average.

corrosion specimens are placed in a slowly refreshed air-stream environment to simulate conditions in the operational period of the basalt repository. Westinghouse Hanford Company investigators have built a test apparatus for performing this type of testing, and the MCC will follow progress of their testing program in FY 1985. The MCC plans to initiate benchmark testing and submit the test method to the MRB in FY 1986.

WASTE-FORM CHARACTERIZATION - S. C. Slate
(Task 3 Leader) - G. R. Robinson, J. W. Shade,
P. A. Scott, J. R. Friley, and W. J. Weber

Major emphasis will be placed in FY 1985 on the application of MCC and other test methods to waste-acceptance specifications. A review of these specifications made this year showed that several MCC test methods are directly usable to demonstrate compliance. The need for two new MCC test methods was also identified in this review. In FY 1985 the MCC will prepare a draft plan on methods that should be employed to collect data that will show that a waste form complies with acceptance specifications.

A summary of progress in each of the major work areas is presented with a short description of the work planned for FY 1985.

MCC-1P AND MCC-2P Static Leach Test Methods

The objectives in this work element were the submission of the two test methods to the MRB for full approval and the collection of data on ARM-1 (Approved Reference Material) glass. MCC-1 was submitted to the MRB early in the year and was returned with comments. These comments were carefully incorporated into the test method and it was returned to the MRB. MCC-2 was submitted to the MRB in August with a report resolving the technical

issues in the appendix. In these submissions the issues on Eh control, quench effects, and applicability to waste-acceptance specifications were resolved by including them as limitations to the test method. The other issues were resolved either by changes in the test method or by presentation of data substantiating the MCC position. An important change in MCC-2 was the elimination of the 190°C test because of excessive leachant losses.

The testing on the ARM-1 glass is proceeding without any significant problems. The results are comparable with those from other glasses such as 76-68. The ARM-1 testing will continue into FY 1985, and after completion of the 1-year tests, the data will be collected into two data packages for review by the MRB. Once approved, the data will be distributed with the ARM-1 glass samples for use by investigators to verify the accuracy of their data.

MCC-3P Agitated Powder Leach Test Method

Extensive tests were conducted this year to help resolve several technical issues remaining on this test method. The test method itself was modified to keep it current with the changes made in MCC-1 and MCC-2. This test method was not submitted to the MRB as planned because experimental results were not sufficient to resolve some of the technical issues.

Tests were made using uranium-doped 76-68 glass to evaluate the effects of quenching on the final results. Samples of leachant were taken at 25°C intervals from 150°C to ambient conditions. Some small effects were noted that were judged to be relatively minor. However, as the MCC does not believe it is possible to rule out quench effects for all systems, a warning on quench effects will be

included in the test method's limitations section. A revised version of MCC-3 will be submitted to the MRB in FY 1985.

MCC-3 tests were initiated using ARM-1 glass. The glass samples in three size ranges were placed in deionized water, silicate water, and brine at temperatures of 90°C and 50°C. These tests will continue into FY 1985, and the final results will be compiled in a data package for submission to the MRB.

MCC-4S Low-Flow-Rate Leach Test Method

The MCC decided this year to rewrite the MCC-4 test method in FY 1985 before resubmitting it to the MRB. The test method is technically adequate, but the presentation of the material needs to be clarified. No work beyond this rewrite is planned for this test method.

MCC-7S Method for Preparation of Isothermally-Heat-Treated Forms

The MCC-7 version that was submitted to the MRB was returned with comments. These comments were beneficial in improving the clarity of the test method and were incorporated. The test method will be resubmitted to the MRB early in FY 1985 for full approval.

MCC-13S Recommended Practice for Determining the Microstructure of a Waste Form

A preliminary draft of the MCC-13 recommended practice was prepared. It will be completed in FY 1985 and submitted to the MRB. This recommended practice will be directly useful in testing waste forms to show compliance with waste acceptance specifications.

MCC-15S Waste/Canister Accident Testing and Analysis

MCC-15 was developed this year to supply data required by the Transportation Technology Center (TTC) and by the draft waste-acceptance specifications. The purpose of this method is to subject full-scale prototypic waste canisters to impacts that are required by regulations to simulate transportation and handling accidents. Data are collected on the deformation of the canister and on the damage to the waste form inside the canister. Canister containment integrity is determined by helium leak testing. The test method was drafted this year, and testing was performed to verify the soundness of approach and to provide data to the TTC. The method will be submitted to the MRB early in FY 1985 after it has been reviewed by the potential users of the test method.

Two full-scale canisters were tested this year using the MCC-15 test method. The prototypic Savannah River Laboratory (SRL) canisters were dropped 9 m, with the center of gravity over the bottom corner. The glass was removed from the canisters and sieved into two size ranges: greater than and less than 6 mm. Representative samples of the smaller particles were taken, and the size distribution was determined to 2 microns. The data were found to follow a log-normal distribution and were comparable to earlier data. The amount of respirable fines in one of the canisters was 80 g, which is a factor of about 150 below the proposed maximum limit for transportation of the reference high-level waste canister from SRL.

Thirty-six quarter-scale canisters were filled with SRL glass and tested under different conditions to determine what conditions cause the most damage to the glass and

canister. Canisters were tested at -25°C and 85°C in four orientations: top, bottom, side, and slapdown. The glass from these canisters was removed and sized, and the data is being analyzed.

Development of MCC-15 will continue in FY 1985, and extensive testing of prototypic SRL canisters will be performed. A study is also planned that will use the canister deformation data to establish the ability of the SRL canister to maintain integrity after an impact.

REFERENCE MATERIAL AND HANDBOOK DATA -

J. L. Daniel (Task 4 Leader) - J. O. Barner, H. E. Kjarmo, and G. B. Mellinger

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

A condensed version of Data Package MCC-D1 was published in PNL-3990. MRB approval of the complete data package was received.

MCC-D2 One-Year Leach Test Data for SRL-131 Glass

Data Package MCC-D2 was revised according to recommendations received from the OTC/MRB and was resubmitted. The Data Panel of MRB approved D2 and recommended additional changes. The MCC response to the Data Panel recommendations is in preparation. Data Package D-2 has been published in PNL-3990.

ARM-1 Static Leach Testing

Static leach tests are continuing on the ARM-1 in order to obtain calibration reference data for field applications of test methods MCC-1, -2, and -3. Tests up to 182 days duration have been completed for MCC-1 at 40° and 90°C, and to 91 days for

MCC-2 at 150°C; both are continuing to 1 year. MCC-3 tests have been started at 90° and 150°C, and will continue to 182 days. Results for all ARM-1 tests are scheduled for publication in MCC Handbook Data Packages during FY 1985.

Shielded Analytical Facilities for Characterization of Radioactive Nuclear Waste Materials

The field installation of the sequential scanner module to MCC's shielded ICP spectrometer was completed. Tests show that detection limits obtained on the sequential module are, on the average, about the same as those for the direct (simultaneous) instrument. New computer software was provided for complete operation, control and data management with the integrated instrument. Initial operational problems are software deficiencies that are being corrected with the substitution of an updated version of the computer software and the addition of associated interfaces and hardware. Use of the instrument and testing of all options are continuing.

Reference Materials

The BWIP project now requires that all MCC preparation and characterization of reference materials to be used in BWIP repository research and development (R&D) be conducted under terms of specific written procedures which have received prior BWIP QA approval. Nine such procedures have been written by MCC to cover the sequential tasks involved in production of approved testing materials. Use was made of existing QA procedures wherever possible, but reference materials production is mostly a unique activity requiring

separate written formal procedures. Following iterative discussions with appropriate BWIP and PNL personnel, the final draft of these nine procedures has been submitted to PNL's MCC and QA management for approval. Concurrence by BWIP QA staff is expected by October 31, 1984. The procedures have been written so as to be applicable to all MCC materials preparation and generally will be applied hereafter regardless of which repository project or glass producer has requested the testing material.

ATM Glasses

Three new Approved Testing Materials (ATM) glasses have been completed, two others are in preparation or advanced planning, and one additional ATM glass is scheduled for late FY 1985 (Table 2.1). The detailed characterization of existing glasses, planned for FY 1984, was postponed until FY 1985 due to limited time and funding resulting from the need for rapid delivery of glasses and preparation of QA procedures during FY 1984. In

TABLE 2.1. MCC's Planned Preparation or Acquisition of Approved Testing Materials, April 1984 to September 1985

Identification	Description	Radioactive Isotopes, g/g Glass ^(a)	Status
New Materials			
ATM-1c	NNWSI ^(b) Glass-I (U-doped 76-68 type)	UO ₂ (3.98E-2)	Complete
ATM-8	NNWSI Glass-II (Actinide-doped 76-68 type)	UO ₂ (4.1E-2) NpO ₂ (4.0E-3) PuO ₂ (1.0E-3) Tc ₂ O ₇ (5.0E-3)	Complete
ATM-9	SRL Defense Glass: ONWI, BWIP	UO ₂ (1.84E-3) PuO ₂ (2.50E-3) Am ₂ O ₃ (6.30E-4) NpO ₂ (1.10E-2) Tc ₂ O ₇ (1.00E-2)	Complete
ATM-11	SRL Defense Glass: BWIP	UO ₂ (1.84E-2) PuO ₂ (2.50E-3) Am ₂ O ₃ (6.3E-3) NpO ₂ (1.13E-2) Tc ₂ O ₇ (1.05E-2) Sm ₂ O ₃ (2.00E-2)	Advance Planning Completed
ATM-12	NNWSI Glass III (Actinide-doped "aged" 76-68 type)	UO ₂ (4.00E-2) NpO ₂ (2.30E-3) PuO ₂ (4.00E-4) Am ₂ O ₃ (1.20E-3) Tc ₂ O ₇ (8.40E-3)	Glass in Preparation
ATM-10	West Valley	To be selected	Planned (Late FY 1985)
ATM-102	BWR Spent Fuel	"Typical" Burnup	Planned
ATM-103	Spent Fuel	"Extreme" Type	Planned

(a) Nominal values: analyses in progress.

(b) Nevada Nuclear Waste Storage Investigations.

addition, funding limitations for FY 1985 have forced postponement of several new proposed glass ATMs and elimination of MCC's preparation of special specimen forms from existing glass bars at user request from its base funding. Preparation of ATM-7 also has been postponed. Users may still obtain unscheduled materials or preparations by special arrangement and provision for funding.

Publication of an updated revision of PNL-4955, Approved Reference and Testing Materials for Use in Nuclear Waste Management Research and Development Programs, is planned for December 1984.

ATM Spent Fuel

Characterization of ATM-101 (HB Robinson PWR rods) was conducted on rods subsequently delivered by request to Nevada Nuclear Waste Storage Investigation (NNWSI) and BWIP research programs. In addition, generic computer calculation of the spent fuel composition was conducted using the ORIGEN program. The limited direct analyses available from rods examined to date confirmed the calculated composition satisfactorily. Additional characterization is planned for FY 1985. Meanwhile, the report Characterization of LWR Spent Fuel MCC-Approved Testing Material - ATM-101 (PNL-5109) was issued summarizing all characterization information available to date. Revision and republication of that report is planned as additional data are obtained during FY 1985.

Two additional spent fuel ATMs are included in the acquisition schedule for FY 1985 (Table 2.1). The search for and selection of suitable material for these ATMs has been started.

Foreign Travel: CEC Leach Test Round Robin

J. L. Daniel traveled to Brussels to participate in the third progress review and final report planning meeting for the Static Leach Test Round Robin conducted by the Commission of European Communities (CEC). Emphasis at the meeting was on evaluation of individual data sets, elimination of data "flyers" where deviations from controlled tests were known to have occurred, and selection of formats for data presentation in the final report scheduled for December 1984.

An important requirement for a materials test of this kind is the use of homogeneous identical material by each participating laboratory. The test glass used for the CEC Round Robin was UK-209, but only MCC had analyzed the glass tested. Therefore, CEC accepted MCC's offer to analyze a sample of each laboratory's glass to determine homogeneity. Results of the analysis are shown in Table 2.2. The glass appears to be homogeneous to well within the precision of the leach test. The largest relative deviation (zinc) corresponds to less than 0.1% actual variation in glass composition.

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TABLE 2.2. MCC-Analyzed Composition of the CEC Round Robin Glass
(Glass type UK-209; 11 Participants)

Element	Corresponding Oxide	Mean Oxide Concentration, %	Between Laboratories	
			Standard Deviation	Relative Standard Deviation, %
Al	Al ₂ O ₃	5.22	0.04	0.71
B	B ₂ O ₃	10.88	0.05	0.48
Ba	BaO	0.37	0.00	1.01
Ce	CeO ₂	0.91	0.03	3.12
Cr	Cr ₂ O ₃	0.36	0.00	0.00
Cs	Cs ₂ O	0.50	0.02	3.80
Fe	Fe ₂ O ₃	2.80	0.01	0.45
La	La ₂ O ₃	0.41	0.00	0.93
Li	Li ₂ O	4.09	0.04	0.92
Mg	MgO	5.69	0.05	0.80
Mo	MoO ₃	1.68	0.02	0.90
Na	Na ₂ O	8.25	0.00	0.00
Nd	Nd ₂ O ₃	1.72	0.00	0.00
Ni	NiO	0.33	0.00	0.00
P	PO ₄	0.27	0.01	5.31
Ru	RuO ₂	0.11	0.01	11.08
Si	SiO ₂	53.02	0.17	0.33
Sr	SrO	0.31	0.00	0.89
Zn	ZnO	0.41	0.09	21.54
Zr	ZrO ₂	1.28	0.07	5.48
Total Analyzed		98.61		
Nominal concentration of element oxides not analyzed ^(a)		1.25		
Total Oxides		99.86		

(a) Includes PdO (0.44), Pr₆O₁₁ (0.43), Rb₂O (0.11), SO₄ (0.10), and Y₂O₃ (0.17).

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3.0 SUPPORTING STUDIES

The objectives 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, both domestic and foreign, 3) provide support to DOE's Office of Civilian Radioactive Waste Management, 4) conduct environmental impact studies and analyses, and 5) provide R&D support to the Low-Level Lead Site Program Office at Idaho and to the Consolidated Fuel Reprocessing Program at Oak Ridge.

3.0 SUPPORTING STUDIES

J. V. Robinson and L. T. Lakey - Program Office Managers

3.1 HANFORD DEFENSE WASTE STUDIES PREPARA-
TION OF ENGINEERING AND ENVIRONMENTAL
IMPACT DATA FOR DISPOSAL OF HANFORD
DEFENSE WASTES

(AR-05-10-02 - 80754)

I. C. Nelson - Program Manager

The objective is to assist DOE-RL in developing and assessing methods for the disposal of existing and future Hanford radioactive defense wastes.

Summary

Work performed during this reporting period is summarized as follows:

- review of Rockwell engineering data packages (EDP)
- technical analyses in support of the Hanford Defense Waste Draft EIS (HDW-DEIS)
- preparation of technical bases for environmental impact assessment.

Review of Rockwell Engineering Data Packages

Updated versions of Rockwell's EDPs, received in April 1984, were reviewed. The EDPs provided the technical data base that described defense waste inventories and disposal activities and costs. Impacts assessed in the preliminary draft Environmental Impact Statement (PDEIS) are therefore based on the March 31, 1984, version of the EDPs, with revisions by letter through September 18, 1984.

Technical Analyses in Support of Preliminary
Draft HDW-EIS

Assessments of environmental impacts that could result from waste disposal operations and over the long term (10,000 years and beyond) continued. Analyses included the following:

- impacts from disposal facilities
- operational accidents (radiological) potential
- industrial accident potential
- transportation impacts
- radiological doses to workers and the public during disposal operations
- socioeconomic impacts
- ecological impacts
- disposal costs
- performance of Hanford reference engineered barrier and marker system
- migration potential of radionuclides from disposal sites to ground water
- migration potential of chemicals from waste sites to ground water
- potential for natural and man-induced disturbance of waste disposal systems
- potential for human intrusion (inadvertent) into disposal systems
- probabilistic risk assessment for source releases from disposal systems
- description of the major disposal facilities and their projected environmental impacts.

More than 30 technical issues were resolved in working sessions with DOE-RL and Rockwell staff.

Results of the analyses listed above were prepared as 23 appendices to the PDEIS.

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

(AR-05-10-02 - 80381)

I. C. Nelson - Program Manager

The objective is to assist DOE-RL in the preparation of a draft and a final Environmental Impact Statement for the disposal of existing and future Hanford radioactive defense wastes.

Summary

Work performed during this reporting period is summarized as follows:

- preparation of the preliminary draft EIS (PDEIS)
- DEIS scoping activities and resolution of technical issues
- peer review activities
- review of EPA proposed standard 40 CFR 191.

Preparation of the Preliminary Draft EIS

Concentrated efforts continued throughout this reporting period to prepare a PDEIS that would be suitable for DOE-HQ review. An initial draft was submitted for DOE-RL review on April 20, a second revised draft was submitted on May 15 and a third revised preliminary draft (working papers) on June 8, 1984. After completing its review of the June 8 preliminary draft, DOE-RL requested that PNL: complete remaining analyses, simplify the draft, and initiate a more thorough Hanford Site review. PNL established a priority status for the effort within PNL. PNL revised the PDEIS extensively and initiated a review and revision cycle for each chapter (8) and appendix (23). Progress was

formally charted daily and was reported weekly in project review meetings with DOE-RL and PNL Program Management and Directors.

The PDEIS was submitted to DOE-RL on September 20. Upon DOE-RL's request, copies were submitted also to DOE-HQ, DOE-SRO, DOE-ALO, DOE-IDO, and Rockwell Hanford Operations.

An overview of PDEIS findings was presented to DOE-RL and Rockwell management on September 27.

DEIS Workshops and Technical Issue Resolution

PNL participated in a series of workshops that resolved technical issues addressed in the PDEIS. Of principal concern were issues associated with long-term impacts resulting from disposal of the waste. All issues were successfully resolved and documented.

Peer Review Activities

Peer review of the program progress and the PDEIS was provided by DOE-RL, the PNL Technical Review Committee, the PNL Senior Overview Committee, and Rockwell's Technical Review Committee. Comments were received following each review, and revisions were incorporated as appropriate and as agreed upon in working sessions.

Review of EPA Proposed Standard 40 CFR 191

Review of EPA proposed standard 40 CFR 191 continued. An analysis was performed to determine whether in-place stabilization and disposal of Hanford defense waste would meet the proposed EPA guidelines for radionuclide releases to the accessible environment over a 10,000-year period.

3.3 COVER TECHNOLOGY EVALUATION FOR URANIUM MILL TAILINGS

(AH-10-15 - 80379)

J. N. Hartley - Program Manager

The objective of this program is to monitor the earthen and asphalt radon barriers on the Grand Junction tailings pile to obtain information on the long-term performance of the barriers for use in designing the remedial action of Uranium Mill Tailings Remedial Action (UMTRA) sites.

Summary

- A meeting was held between PNL, Bendix Field Engineering Corporation, and DOE to discuss transfer of the Grand Junction tailings pile monitoring activities to Bendix.
- Data accumulated by dataloggers at Grand Junction during the last 3 years have been compiled and plotted for incorporation into the final topical report to be completed by the end of October 1984.
- Weekly radon flux measurements were made on the earthen covers using large-area activated carbon canisters. The measurements were taken on one location on each of the four earthen covers. Results show that all covers except the uncompacted adobe meet the Environmental Protection Agency (EPA) standard of $20 \text{ pCi m}^{-2} \text{ sec}^{-1}$.
- The final Grand Junction field test monitoring trip was made June 18-19, 1984. The measurements taken on the FY 1980 and FY 1981 test areas included radon flux and neutron moisture. In addition, surface moisture and density measurements were taken on the earthen covers using a nuclear density meter.

Program Management - J. N. Hartley

A meeting was held in Grand Junction among PNL, Bendix, and DOE personnel to discuss transfer of the monitoring activities to Bendix for FY 1985. It was agreed that PNL would transfer to Bendix a CR5 datalogger and the weather station currently located on the test site. DOE requested that the responsibility for the Grand Junction site be transferred to allow Bendix to perform limited monitoring of the site in FY 1985 and beyond.

Data Analysis - H. D. Freeman

The temperature, moisture-block, weather, and alpha-tube data collected over the last 3 years have been compiled and plotted. Plots are presently being incorporated into the final field test topical report to be completed by the end of October 1984.

A model verification was started this month to compare PNL's computer code RND1D with data obtained at Grand Junction during 1983 as part of a study funded by the Nuclear Regulatory Commission. Results of the model verification will be included in the field test monitoring report.

Site Monitoring - H. D. Freeman

Site monitoring included both weekly radon flux measurements on the earthen covers and a 2-week extensive monitoring trip during June 1984.

Weekly radon flux measurements were taken by a subcontractor using a large-area (0.107 m^2) activated carbon canister. A single location on each of the earthen cover

test plots was measured each week. These locations were also measured several times during the June monitoring trip. Results of these measurements are presented in Table 3.1.

The average of all measurements taken is somewhat misleading, as there were a disproportionate number of samples taken during June, when radon flux was high. The second average shown in Table 3.1 represents the once-per-week sampling interval, which would be more representative of the annual average. If the once-per-week average is used, all the covers except the uncompacted adobe meet the EPA standard. These data illustrate the need to take radon flux measurements over a period of time to get a true annual average.

The final monitoring trip to the Grand Junction test site was made June 18-19, 1984. The activities included radon flux measurements, neutron probe moisture measurements, and surface density and moisture measurements. The areas monitored were selected locations on the FY 1980 and FY 1981 test areas, including multilayer, asphalt and earthen covers. Results of the measurements taken using the PNL flow-through system are summarized in Table 3.2.

Table 3.2 shows that the only barrier not meeting the EPA standard of $20 \text{ pCi m}^{-2} \text{ sec}^{-1}$ is the uncompacted adobe. After 4 years of exposure, the FY 1980 asphalt emulsion barrier is still the most effective barrier tested.

TABLE 3.1. Large-Area Activated Carbon Canister Radon Flux Measurements ($\text{pCi m}^{-2} \text{ sec}^{-1}$) on the Grand Junction Earthen Cover Tests April-June 1984

Date	Mancos Shale	Bentonite Clay	Compacted Adobe	Uncompacted Adobe
April 2	1.1	14.4	6.7	36.4
April 9	1.4	12.1	6.5	57.5
April 16	1.2	6.6	6.3	22.3
April 23	2.2	7.7	12.2	40.0
May 1	0.6	7.3	6.6	34.2
May 8	0.8	8.1	7.5	22.5
May 15	0.8	9.5	19.4	39.4
May 22	1.6	13.9	17.0	23.5
June 4	1.6	23.7	23.0	43.0
June 11	1.2	15.0	15.6	33.0
June 19	1.4	22.7	40.9	89.0
June 20	1.6	17.5	41.0	97.0
June 21	1.3	16.7	32.2	85.2
June 25	1.5	18.7	33.2	85.2
June 26	1.8	22.5	35.5	87.1
Average $\pm$ standard deviation (SD)	1.3 ± 0.4	14.4 ± 5.9	20.2 ± 13.2	53.0 ± 27.7
Average using only one measurement per week	1.3 ± 0.5	13.6 ± 6.3	16.4 ± 11.7	44.0 ± 22.8

TABLE 3.2. Summary of Radon Flux Measurements ($\text{pCi m}^{-2} \text{ sec}^{-1}$) Taken at Grand Junction June 18-19, 1984

Barrier	Average	Standard Deviation
FY 1981 Barriers		
15-cm gel mix	18.5	11.9
Asphalt emulsion	2.3	6.2
Mancos shale	1.0	0.9
Bentonite clay	7.4	7.7
Compacted adobe	18.8	13.3
Uncompacted adobe	29.5	25.2
FY 1980 Barriers		
Asphalt emulsion	0.7	1.9
Rubberized asphalt	6.2	3.1

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

M. J. Graham - Program Manager

The primary objective of this program is to conduct waste-form leaching tests in a field facility in order to determine typical source terms generated by commercial solidified low-level waste. The solidification agents to be tested are cement, polyvinyl styrene, and bitumen. Through field and laboratory research, the chemical and physical processes that control the concentrations of radionuclides in the soil solution surrounding the waste in an arid climate will be identified. Methods for representing source-term boundary conditions in radionuclide transport models will be determined.

Summary

The Special Waste-Form Lysimeters - Arid Task was begun in FY 1983 for the long-term field evaluation of solidified low-level waste forms intended for shallow-land burial. A similar task is ongoing at the Savannah River Laboratory (SRL) for a humid climate. A field lysimeter facility, consisting of ten lysimeters around a central instrument cais-

son, has been constructed in which to perform the field testing. Waste samples have been emplaced in the facility, which became operational at the end of the second quarter of FY 1984. Two bitumen waste forms were added to the facility in September 1984.

All laboratory-scale solidified samples for the leach tests have been prepared by Brookhaven National Laboratory (BNL). The leach tests are proceeding on schedule. PNL has completed the organic analyses of three of the four liquid-waste samples.

The geochemical computer code used in this program has been selected and deficiencies in the thermodynamic data base identified. Cobalt was added to the thermodynamic data base for the geochemical computer code.

Baseline data on the lysimeter soils have been collected and published in a topical report. These data include water-retention characteristics, hydraulic conductivities, and particle-size distributions for soils collected from each of the ten lysimeters.

Lysimeter Study

The Special Waste-Form Lysimeters Study is designed to quantify the release and migration of radionuclides from solidified commercial low-level waste. A field test facility has been built to contain selected waste forms in specially constructed lysimeters. Solidified low-level waste samples from commercial power reactors have been emplaced in the facility (Table 3.3). The waste forms are being characterized by standard laboratory tests such as MCC-1 and the IAEA leach test to obtain estimates of radionuclide release. Solubility studies are also being conducted to determine the amount of contaminant available for transport after release.

TABLE 3.3. Waste Forms Obtained for the Special Waste-Form
Lysimeters - Arid Program

<u>Waste Stream</u>	<u>Reactor</u>	<u>Solidifying Agent</u>
Boric acid concentrate waste	PWR ^(a)	Masonry cement
Boric acid concentrate waste	PWR	Bitumen
Evaporator-concentrate (regenerative) waste	BWR ^(b)	Portland Type III cement
Evaporator-concentrate (regenerative) waste and ion-exchange-resin waste	BWR	Portland Type III cement
Evaporator-concentrate (regenerative) waste and ion-exchange-resin waste	BWR	Vinyl ester-styrene

(a) Pressurized water reactor.

(b) Boiling water reactor.

Data collected from this research will be used to relate laboratory measurements of waste-form characteristics to the release and migration rates observed in the field. Comparisons can be made between results obtained from this arid site facility and those from the humid site at SRL.

The field lysimeter facility is designed to accommodate ten waste forms. Ten closed-bottom lysimeters, 3 m deep by 1.8 m dia, are arranged around a central instrument caisson, 4 m deep by 4 m dia (Figure 3.1). The central caisson houses all data and sample collection equipment while providing access to sampling ports in each lysimeter for selective sampling of the soil and waste forms. Leachate from each closed-bottom lysimeter drains to the central caisson, where it can be collected, measured, and sampled.

Suction candles (ceramic cups) placed in the bottom and around the waste will be used to periodically collect soil water samples for chemical analysis. Moisture content and soil temperature are being monitored at 12-hr

intervals. The data are sent over a phone line to a computer; a backup cassette tape data-storage system is also in place. Radiological analyses will be performed in situ and through selective sampling. These will provide the Low-Level Waste Management Program with direct field results regarding leach and migration rates from actual commercial solidified waste.

PNL has completed the organic analyses of the liquid fraction of the boric acid concentrate waste (from a PWR) that was solidified with masonry cement. A variety of organic chelating agents were identified in the hydrophilic organic fraction of the sample. Two of them, ethylene-diaminetetraacetic acid (EDTA) and citric acid, are typically used in the decontamination operations at nuclear facilities. The organic analyses of the BWR waste streams (evaporator-concentrate waste and evaporator-concentrate waste with ion-exchange-resin waste) and the solid residue from the PWR waste stream (boric-acid concentrate waste) are also complete. The PWR

FIGURE 3.1. Conceptual Drawing of the Field Lysimeter Facility

solid residue contains chelating agents similar to those found in the liquid fraction of the sample. The organic contents of the BWR waste stream are qualitatively different. The major components are dimethyl phenols and their phosphate esters. Such compounds are used as plasticizers and hydraulic fluids.

In addition to the field monitoring, which is the central purpose of the task, a series of laboratory testing and geochemical modeling exercises is planned. These tests will

help to predict expected leach rates and help to interpret the field data. The ion speciation-solubility and mass transfer geochemical code MINTEQA2 will be used to model the waste stream, the waste form, and any column effluent and batch solutions. The geochemical modeling efforts are being focused on the organics to determine the effects of these organics on the behavior of radionuclides in the environment. Specifically, the goals of the laboratory and modeling work will be to

identify the mechanisms that are controlling the release of radioactivity from the waste form.

3.5 TRANSPORT ASSESSMENT - ARID

(AR-05-15-15 - 80033)

G. W. Gee - Program Manager

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

Field Data Collection

Field sites at Hanford were monitored through September. Moisture storage changes from June through September 1984 indicated that as much as 2 cm drainage occurred below the 3.5-m depth at the 300 Area grass site. Monitoring of field sites will continue for the next 3 years in order to document moisture profile changes below vegetated and nonvegetated (bare) soil surfaces at Hanford. This follow-on work will be supported by Hanford Defense Waste programs.

Transpiration Modeling

The water balance modeling effort was completed and incorporated in the final report Arid Site Water Balance: Evapotranspiration Modeling and Measurement (PNL-5177).

Program Overview

All milestones have been met on this program, and it has been officially completed. The major objectives of the program have been realized in the following ways:

- A field test facility was established for arid site water balance and radionuclide transport studies. This facility consists

of both deep (7.6 m) closed-bottom lysimeter for radionuclide and water transport studies and shallow (1.5 m) weighing lysimeters for evapotranspiration and drainage studies.

- Basic data sets have been collected on the physical and hydrologic properties necessary to characterize a low-level waste site in an arid environment. The data sets include particle-size analysis, water-retention characteristics, thermal properties, and vapor-transport parameters. These data also include 5 years of record of water storage changes and climate variables, and 2 years of record of drainage and evapotranspiration at both bare and vegetated sites.
- Extensive testing of monitoring systems for unsaturated water flow has revealed some limitations in the use of conventional sensors (e.g., resistance blocks, tensiometers, thermocouple psychrometers, neutron probes). Neutron probes, properly calibrated and maintained, appear to be the most reliable instruments for monitoring soil-moisture changes over long periods at arid sites. However, moisture measurement alone cannot be used to predict water flow rates except under special circumstances.
- Field and laboratory measurements of radionuclide migration in unsaturated Hanford soil indicated that mobility or complexation with organic complexants (e.g., EDTA) is time-dependent and does not appear to depend significantly on the degree of saturation of the soil water. In contrast, tritium moves both in the vapor and liquid phases in unsaturated soil and is not retarded to any measurable extent. We have determined that tritium is an acceptable tracer for unsaturated water flow (vapor and liquid transport).

- Model simulations of water flow at arid sites confirm our observations that significant drainage can occur on either bare or vegetated soils under arid climate conditions. Precipitation distribution (e.g., fall/winter versus spring/summer) and soil and plant characteristics play major roles in determining the amounts of drainage that can be expected at arid sites.

Publications

The following publications document the above observations. These publications represent contributions that were supported either entirely or in part by the Transport Assessment - Arid Program from FY 1978 through FY 1984.

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3.6 KRYPTON-85 RISK HEALTH IMPACTS ASSESSMENT

P. J. Mellinger - Program Manager

The objective of this study is to develop documentation on the need, or lack thereof, for ^{85}Kr recovery and storage as opposed to routine release of the radionuclide.

Two alternative methods of capturing ^{85}Kr , cryogenic distillation and fluorocarbon absorption, and one method of immobilizing the captured gas, ion implantation/sputtering, were theoretically incorporated into a representative fuel reprocessing plant (FRP). The Barnwell Nuclear Fuel Plant was assumed for this study even though there are no known plans to start up this facility.

Work-force and population radiation doses were calculated and analyzed for operating

and maintaining the two capture options for immobilization. Those doses were then compared to the public doses from routine release of ^{85}Kr .

Given the uncertainties in the models used to generate lifetime (70-yr) risk numbers, the differences in total risks to the two exposed populations cannot be considered significant. The differences in the numbers are too small to be meaningful: 0.02 to 0.027 radiation-induced fatal cancers expected in the occupational work force over 30 years of exposure due to ^{85}Kr recovery as opposed to 0.017 fatal cancers in the general population over the same time period due to total routine release of ^{85}Kr . It is possible that no risk would be incurred from either release or capture of the ^{85}Kr . There is certainly no reason to conclude that risks from ^{85}Kr routinely released to the environment are greater than those that would result from routinely capturing and immobilizing the radionuclide.

In addition to the krypton study, this program examined also the adequacy of radioiodine control and monitoring at nuclear fuel reprocessing plants. The conclusion of this study is that, with available techniques, inline, real-time monitoring for ^{129}I in FRP gas streams is currently not feasible. Moisture, chemicals, and other radioactive fission products interfere with in-plant measurements. Samples collected over several days must be taken to a laboratory for ^{129}I analysis. Measurement techniques currently in use or under investigation include neutron activation analysis, scintillation counting, mass spectroscopy, and gas chromatography coupled with electron capture detection.

This program has been concluded.

3.7 KRYPTON ION IMPLANTATION

E. D. McClanahan - Program Manager

The objective of this program is to develop the process of implanting krypton in a sputter-deposited metal matrix for the 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.

Summary

The design for a disposable krypton trapping unit for BRET^(a)-scale application was completed. This unit was designed to be capable of being connected remotely to a station providing the necessary utilities for its operation. The same station will also accept a larger module suitable for commercial-scale operation. The BRET-scale trapping unit equipped with a target whose copper-to-yttrium atom ratio is 4.1, can pump 1.5 L of krypton STP per hour operated at a total input power of 18 kW.

A major milestone to be completed this year was to choose a metal matrix for implanting ⁸⁵Kr for 200-Ci-scale testing in FY 1985. The copper-yttrium alloy system was chosen; the other candidate systems were copper-lanthanum, nickel-yttrium and nickel-lanthanum. The copper-yttrium system is believed to be superior because of its mechanical stability and adhesiveness and the relatively low vapor pressure of copper as compared to nickel. The bulk krypton release for the krypton-loaded copper-yttrium alloy occurs above 600°C, which means that its thermal stability is comparable to or exceeds the other candidate alloys. The 18 at.% yttrium alloy can achieve a krypton loading of 8 to 9 at.% compared to 8 to 12 at.% krypton for nickel-lanthanum alloys. The

higher sputtering yield for copper and its lower cost partially compensate for the higher krypton content and lower cost of lanthanum in the nickel-lanthanum alloy.

Equipment Development - E. D. McClanahan and E. N. Greenwell

One goal for FY 1984 was to complete the design of the disposable gas trapping unit for BRET-scale application. The preliminary design was tested on experimental demountable systems with substrate areas of 1000 to 1500 cm². The pumping performance of these tests met the design expectations, but pump life was about 40 hr. While the trapping life has not been specified for BRET operation, a design goal for commercial-scale operation was previously set at ~130 hr (Parsons 1981). Internal electrical shorts were the primary cause of cessation of pumping in the demountable test units. These were not unexpected because the dimensional tolerances for the electrodes and their spacings are critical. For example, the gaps between the filament support posts and the anode and their adjacent structures are about a half-centimeter. The process by its nature requires that the inside diameter of the growing film on the substrate decrease by 2 cm and that the target decrease in diameter by more than 2 cm.

To maintain the desired trapping rate, it is essential that all internal electrodes, including the anode, filament, target and substrate, remain electrically isolated from each other for the life of the trapping unit. Internal shorting of the electrodes usually occurs by one of three mechanisms: 1) sputter-deposited material becomes unbonded by a combination of poor adherence and high residual stress, 2) shorting of ceramic insulators because of film buildup, or 3) the deposit

(a) Breeder Reprocessing Engineering Test.

builds up on the anode or filament post sufficiently to bridge the gap between it and the adjacent structure.

Unbonding due to poor adherence or to high intrinsic compressive stress can be prevented by the choice of the material for the structure, of the cleaning procedure and, most importantly, of the trapping alloy system, which is discussed in the next section.

Breakdown of the insulator surfaces by buildup of a continuous metallic film is prevented by shielding the ceramic surface from the line-of-sight access to sputtered metal atoms.

Test operation of the demountable trapping units showed that the shape and position of the electrodes could not prevent excessive buildup on the anode and filament supports; therefore, the design effort was focused on minimizing the deposition on these components by increasing their temperature. The temperature of filament posts is controlled by choice of the proper cross-sectional area utilizing I^2R heating. The anode temperature can be increased by decreasing its surface area.

Tests using the 1000-cm² demountable trapping unit with both nickel-yttrium and copper-yttrium composite plug-type targets gave the following results: 1) of the configurations tested, no operating parameters were found that would enable the anode to run sufficiently hot to prevent excessive buildup on its surface, 2) a configuration was found that permitted only 4% of the total amount deposited on the substrate to be deposited on the anode, and 3) the latter configuration may require active control on the part of the operating program to restore the discharge following an electrical fault in the plasma discharge circuit. It was not possible to determine whether the plasma instability was a result of poor pressure control or too

small an anode. In any event, a compromise must be made in the size and configuration of the anode and the desire for passive plasma discharge control.

Figure 3.2 is a drawing of the design for the BRET-scale disposable trapping unit. This trapping unit is designed so that it can be connected by remote control to a radiation-shielded station providing the necessary utilities for its operation. The same station will also accept a larger trapping module suitable for commercial-scale operation. The target is a copper-yttrium alloy containing 18 to 21 at.% yttrium. The anode and filament supports are made of tantalum and the substrate is stainless steel. This unit as designed should pump 1.57 L/hr of krypton at STP operating at 2400 V, 5.5 A on the target and 270 V, 10 A on the substrate. The total power dissipated in the trapping unit is 18.1 kW, which equates to a pumping efficiency of 0.087 L/kWh.

Supporting Research - R. W. Knoll

The goal of this task has been to perform krypton ion-implantation experiments in a small bench-scale apparatus, and then to characterize the resulting deposits and select the alloy most suitable for krypton entrapment in the large-scale systems. This work was motivated by the previous year's experience in attempting to produce thick deposits of nickel-lanthanum-krypton alloys. Those deposits spalled apart due to the material's brittleness and high intrinsic compressive stress when thickness exceeded 1 to 2 mm. In the first half of this year, either thick or thin deposits of nickel-yttrium-krypton, copper-yttrium-krypton, copper-lanthanum-krypton (and nickel-lanthanum-krypton at elevated substrate

FIGURE 3.2. BRET-Scale Disposable Gas Trapping Unit

temperature) were made in the cylindrical bench-scale apparatus using a substrate bias of -225 V, as described in the previous semi-annual progress report (McClanahan et al. 1984). Cylindrical sputtering targets containing an axially graded distribution of the rare earth alloy were used in those experiments to produce deposits containing a composition spectrum. Thus materials characteristics such as krypton content versus rare earth content for a given substrate-bias voltage could be obtained in a single experiment.

During the second half of FY 1984, characterization of the above deposits continued, and several more copper-yttrium-krypton deposits were made in order to explore that alloy system in greater detail. Data from these deposits showed that alloy system to have the least desirable krypton-entrapment

characteristics. Its mechanical stability was poor, partly because of equipment problems that occurred during the run, but also because of frequent plasma-arcing during deposition (probably related to the presence of lanthanum metal). Measurements of krypton content compared to lanthanum content indicated a maximum of only ~ 5 at.% krypton for lanthanum concentrations > 9 at.%. While better mechanical stability might have been obtained under more ideal deposition conditions, and while a greater krypton content might be attained at a substrate bias other than -225 V, copper-lanthanum-krypton alloys appear inferior to the other alloy systems as krypton-entrapment matrix materials.

On the other hand, the copper-yttrium-krypton system exhibited some very favorable characteristics; thus additional experiments

were performed on that material in the second half of FY 1984. Experimental results obtained earlier in FY 1984 had indicated that copper-yttrium alloys might be preferable to nickel-yttrium or nickel-lanthanum alloys as krypton-entrapment matrix materials for several reasons. First, the mechanical stability of a thick (approx. 3 mm) copper-yttrium-krypton deposit (Run GT129) was much better than that of nickel-lanthanum-krypton and at least as good as that of nickel-yttrium-krypton. Second, the vapor pressure of copper is considerably greater than that of nickel at a given temperature; therefore the copper alloy has less tendency to collect on and to short out the anode during deposition. However, the thermal stability of the copper-base alloys was unknown, and the krypton content of the GT129 copper-yttrium-krypton deposit was found to be significantly lower than that of nickel-yttrium-krypton or nickel-lanthanum-krypton deposits containing rare earth concentrations between 5 and 15 at.%. Experiments were therefore conducted on copper-yttrium-krypton deposits to measure thermal stability and to determine whether krypton content could be increased.

Description of the new results requires a brief review of the GT129 copper-yttrium-krypton experiment performed earlier using the bench-scale deposition apparatus. In that work, the substrate was biased at -225 V during deposition (i.e., the Kr^+ ion bombardment energy was about 225 eV), and a copper-yttrium target with an axially graded yttrium content was used. The yttrium content of the resulting deposit varied from ~17 at.% to 3 at.% between the top and bottom of the substrate cylinder, and krypton content varied with yttrium content as shown by the GT129 data points in Figure 3.3. The increasing slope of the krypton versus yttrium curve in

the alloy composition range of ~5 to 14 at.% yttrium suggested that krypton content might be increased to ~10 at.% by increasing yttrium content to ~20 at.%. The scatter in the data in the 14 to 17 at.% yttrium range produced uncertainty about the direction in which the curve was headed. Thus an experiment was performed (Run GT137) using a uniform copper-yttrium target designed to produce a deposit containing 18 to 19 at.% yttrium. By monitoring the inflow rate of krypton during that experiment, the krypton content of the deposit was estimated to remain in the 6 at.% range if a substrate bias of -225 V (as in GT129) was used. This result showed that the krypton versus yttrium curve for -225 V bias actually levels off above yttrium concentrations of ~15 at.%. Krypton content, however, could be increased to ~8 at.% by increasing substrate bias to -270 V, as indicated by the GT137 data point in Figure 3.3.

To further investigate the influence of substrate bias on krypton content, another copper-yttrium-krypton deposit (Run GT142) was produced using a -270 V substrate bias with the graded copper-yttrium target of experiment GT129. As shown in Figure 3.3, for yttrium concentrations ≥ 4 at.% the entrapped krypton concentration was considerably higher in the deposit biased at -270 V. Although the GT142 data extended only to ~14 at.% yttrium, that curve can be extrapolated to the GT137 data point at 18 at.% yttrium, as indicated by the dashed line. The principal results of these experiments are that 1) copper-yttrium-krypton deposits containing ~8 at.% krypton are attainable if a substrate bias of -270 V and a yttrium concentration of 18 to 20 at.% are used and 2) compared with the nickel-lanthanum-krypton and nickel-yttrium-krypton

FIGURE 3.3. Krypton Content Versus Yttrium Content at Various Substrate Bias Voltages for Sputter-Deposited Cu-Y-Kr Alloys

alloy systems, a higher substrate bias and/or a higher rare earth concentration is required in the copper-yttrium-krypton alloy to entrap a given concentration of krypton.

Thermal stability measurements were performed on copper-yttrium-krypton specimens of four different compositions to determine crystallization temperatures and the temperature range in which bulk release of the entrapped krypton occurs. The specimens were from the thick GT129 deposit, and their compositions ranged from $\text{Cu}_{88}\text{Y}_{8.5}\text{Kr}_{3.5}$ to $\text{Cu}_{79}\text{Y}_{15}\text{Kr}_6$. Measurements were performed using differential scanning calorimetry over a temperature range of 20 to 700°C and at a scan rate of 20°C/min. Thermal gravimetric analysis was also performed on each specimen over the same temperature range. Results of these experiments were very encouraging, as

the amorphous deposits crystallized in the temperature range of 330 to 350°C, and bulk krypton release occurred above 600°C. Therefore the thermal stability of the copper-yttrium-krypton deposits during rapid heating is on par with that of the nickel-yttrium-krypton and nickel-lanthanum-krypton deposits.

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4.0 RECENT WASTE MANAGEMENT PUBLICATIONS

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