Nuclear Waste Management Quarterly Progress Report January Through March 1978

**A. M. Platt
J. A. Powell**

August 1978

**Prepared for the U.S. Department of Energy
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**Pacific Northwest Laboratory
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NUCLEAR WASTE MANAGEMENT
QUARTERLY PROGRESS REPORT
JANUARY THROUGH MARCH 1978

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

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Pacific Northwest Laboratory
Richland, Washington 99352

FOREWORD

This document is one of a series of technical progress reports designed to report on commercial radioactive waste management programs at Pacific Northwest Laboratory. These programs are funded by the Department of Energy's Office of Waste Management. The Commercial Radioactive Waste Fixation Program is reported elsewhere. These reports are issued on a quarterly basis.

The prior report in this series is Nuclear Waste Management Quarterly Progress Report October Through December 1977, PNL-2377-4.

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1.0 DECONTAMINATION AND DENSIFICATION OF CHOP-LEACH CLADDING RESIDUES

R. L. Dillon, R. S. Kemper, R. R. King

Fuel element cladding residues from the chop-leach head-end process constitute a low-density, high-surface-area metallic waste with a substantial actinide element contamination. The objective of this study is to determine the feasibility of, and to develop methods for, removal of long half-life transuranics from the cladding residues and for consolidation of the zirconium and stainless steel residues. The study involves:

1. *selection and laboratory testing of methods for transuranic removal by*
  - a) *aqueous reagents*
  - b) *molten salt reagents*
  - c) *fluxing during melting*
2. *consolidation of the fuel element hulls by melting and casting Zircaloy, stainless steel, Inconel or alloys of the three*
3. *testing and evaluating the consolidated fuel hulls for long-term waste management*
4. *Absorption and fixation of tritium into the waste ingots*
5. *consolidation of transuranics removed.*

SUMMARY

The approximate distribution of the transuranic elements in a moderately irradiated boiling water reactor (BWR) fuel hull was determined. This was done by chemically leaching the cladding to simulate dissolver treatment and subsequently treating the hull to determine which were the easily removed, the firmly held, and the uniformly distributed transuranic elements and their precursors.

Pilot-scale equipment for chemical decontamination of cladding hulls is being assembled in a configuration simulating the arrangement for remotely-operated hot cell decontamination. The prototype equipment will use an HF/argon decontamination cycle

followed by an aqueous wash to process chop-leach residues at ~20 kg/hr. The decontamination prototype will be operated in conjunction with the Inducto-slag ingot melting furnace, which has been undergoing melting tests while installed in the simulated hot cell configuration.

Simulated Zircaloy cladding residues have been melted in the Inducto-slag furnace to identify the best melting procedures for densification of the residues. Tests to date have established procedures for producing ingot at rates of ~30 to 35 kg/hr and power levels of ~85 to 100 kW. Although ingot soundness and hardness have been acceptable, melting tests are continuing in order to eliminate roughness of the ingot sidewall. The

thirteen individual melts performed with the Inductoslag furnace to date are summarized here.

DECONTAMINATION PROCESS

The approximate distribution of the transuranic (TRU) elements in a moderately irradiated BWR fuel hull was determined by chemically leaching the cladding to similar dissolver treatment and subsequently treating the hull. These processes determined 1) the TRU contamination easily removed from the surface oxide, 2) the contamination not chemically removable from the surface oxide, and 3) the TRU plus precursor elements uniformly distributed in the substrate metal.

Several pieces of BWR fuel cladding from which the fuel was mechanically removed were treated twice with boiling 8M HNO_3 . A treated cladding hull was cut into two pieces, each 3.5 cm long. One piece was leached for six 2-hr periods in a mixture of 0.4M ammonium oxalate, 0.16M ammonium citrate, 0.1M ammonium fluoride, and 0.3M hydrogen peroxide at 90°C. Total alpha analysis of these six solutions indicated the relative rate of leaching as shown in Figure 1.1. The residual hull was completely dissolved, and the six previous leach solutions were combined in a composite sample for detailed analysis.

The second hull piece had its surface oxide completely removed by abrasion and etching, which removed less than 40 μm . The clean metal sample produced was completely dissolved in three samples for detailed analysis.

FIGURE 1.1. Relative Leaching Rates for Six 2-hr Leaches of a Treated BWR Cladding Hull

The results of these analyses are summarized in Table 1.1 for the significant TRU elements. The relatively low levels of contamination in this hull are probably the result of low levels of uranium and thorium contamination of the base metals and the moderate irradiation. Analytical data have not yet been compared with a computer simulation of the exposure.

Pilot-scale equipment for the chemical decontamination of chop-leach cladding residues at a rate of ~ 2 kg/hr has been fabricated and installed on a simulated hot cell floor plan with the Inductoslag ingot melting furnace. The prototype decontamination equipment consists of an HF/argon gas reactor, an aqueous washer, and associated chemical storage tanks as described in previous reports. Lifting rings, foot pads, and additional cross bracing were added to allow for remote manipulation and assembly. The components of the decontamination system were bolted to a platform to hold them in proper spatial orientation.

TABLE 1.1. Significant TRU Elements in a BWR Fuel Hull

Analysis	Surface Leach(a,c)	Oxide Surface and Outer (<40 μ m) Metal Layer(a,b)	Base Metal(a)	Base Metal(c)
Total α	12 nCi/cm ²	3.5 nCi/cm ²	3.4 nCi/cm ²	5.8 nCi/g
Total U	9.8×10^2 ng/cm ²	4.2×10^3 ng/cm ²	4.4×10^2 ng/cm ²	7.5×10^2 ng/g
²³³ U	0	0	0.6 at%	--
Total Th	--	--	1.7×10^2 ng/cm ²	2.9×10^2 ng/g
²²⁸ Th	--	--	5×10^{-5} at%	1.1×10^{-1} nCi/g
Total Pu	30 ng/cm ² 2.7×10^2 nCi/cm ²	1.7×10^2 ng/cm ² 1.9×10^3 nCi/cm ²	11 ng/cm ² 9×10^1 nCi/cm ²	19 ng/g 1.6×10^2 nCi/g
²³⁹ Pu	69 at%	64 at%	69 at%	--
²⁴⁰ Pu	19 at%	20 at%	19 at%	--
²⁴¹ Pu	9 at% 2.6×10^2 nCi/cm ²	10 at% 1.9×10^3 nCi/cm ²	8 at% 8.8×10^1 nCi/cm ²	-- 1.5×10^2 nCi/g
²⁴¹ Am	4.5 nCi/cm ²	1.6 nCi/cm ²	1 nCi/cm ²	1.7 nCi/g

a. All TRU contamination was assumed to be in the base metal or on the interior surface. The unit cm² is for the internal surface area only.

b. This analysis is the leached hull minus the base metal.

c. Inside surface area to mass ratio of this hull was 1.75 cm²/g.

An artist's conception of the equipment in the hot cell layout is shown in Figure 1.2, minus the pipefitting associated with the decontamination unit. A simplified flow diagram of the decontamination system is shown in Figure 1.3. Flow meters and other cell equipment hardware are being installed as the pipefitting work progresses.

The HF and argon gas supply systems were designed, and fabrication is under way. The process control instrumentation was designed, and all components were purchased; control cabinets are scheduled for complete assembly during the next reporting period.

The prototype decontamination equipment and the Inductoslag melt

densification furnace are being set up for combined, nonradioactive demonstrations in a building operated by PNL on the Hanford Reservation. The building's scrubbed ventilation system will be extended to provide ventilation for the melt densification furnace and remove any HF gas resulting from leaks in the chemical decontamination prototype.

A closed circuit television monitoring system is planned for use in the cell. The system will initially be used in hot cell remote assembly and repair operations. Plans include at least one stationary camera and another mobile unit operable by the remote manipulators as suggested in Figure 1.2.

FIGURE 1.2. Fuel Hull Decontamination and Densification Equipment

FIGURE 1.3. Fuel Hull Decontamination Flow Diagram

DENSIFICATION PROCESS

Testing of the Inductoslag ingot melting furnace with simulated Zircaloy cladding residues was continued. The furnace is the densification portion of prototype decontamination/densification system being set up for nonradioactive demonstration. The Inductoslag crucible damaged by a melt-through in one segment of the crucible⁽¹⁾ was repaired, and a water filter was installed to prevent a recurrence of a water channel blockage due to trash in the water system. A second filter was installed on the water coolant line to the induction power source. A temporary supplemental water line was installed until the building water service can be increased. Heretofore, the water supply to the induction power source has been inadequate, resulting in excessive induction coil water temperatures.

The furnace's material feeding system was altered to limit the amount of feed entering the crucible. Precise feed control of the simulated chopped cladding hulls is now achievable. Subsequent minor adjustments can be made to the feed system to accommodate other forms of feed material.

Table 1.2 summarizes the Inductoslag heats made to date. The melting operations from pump-down to power-off on Heats 8 through 13 were performed from a remote position outside the "cell" area designated by painted lines representing the cell walls.

Two television receivers were used to guide operations. The experience gained from these heats has resulted in establishing a melting procedure that can produce ingots at rates of about 30 to 55 kg/hr using power levels of 85 to 110 kW. Energy usage varies from 2 to 5 kWh/kg. Ingot soundness and hardness are excellent. The average Brinell hardness values of 187 are within the ASTM (American Society for Testing Materials) specification of 200 maximum for nuclear-grade Zircaloy and indicate that the material is within acceptable chemical and corrosion resistance standards for Zircaloy-4.

The chief problem with the ingots produced by this melting procedure is the rough surface of the ingot side wall, atypical of Inductoslag-melted ingots. This roughness makes flux removal difficult and is not desirable for leaching resistance during long-term storage. The roughness is caused by metal freezing against the crucible wall without the benefit of a CaF_2 flux layer between the freezing metal and the crucible wall. Future efforts in correcting this problem will focus on:

- raising the induction coil relative to the crucible
- reducing or eliminating crucible taper
- raising the melting position within the crucible
- reducing melting power to a minimum
- reducing the length of the induction coil.

TABLE 1.2. Summary of Inductoslag Melting Furnace
Runs with Zircaloy-4

Heat Number(a)	Melted Weight, Kg	Melt Rate, Kg/hr	Approximate Power, kW	Energy Use kWh/kg	Hardness, BHN(b)	Remarks
1	3	--	100	--	--	Stainless steel charged for equipment checkout; metal runout due to flux not melting; crucible water leak.(1)
2	--	--	85	--	--	Mild steel charged for equipment checkout; magnetic field from the coil pulled excess metal from the feed chute, causing a metal bridge.
3	2.3	10.7	83	7.8	--	USBM(c) expert present; no major problems; heat terminated for examination.
4	10.9	28.4	90	3.2	189	USBM expert present; no major problems; smooth ingot side wall typical of USBM Inductoslag ingots.
5	--	--	82	--	--	Frozen metal ring formed above the melt zone; efforts to melt this ring failed; crucible recoated; segment gaps reduced.
6	14.1	20.7	100	4.9	185-194	Power limited because of hot induction coil cooling water; rough ingot side wall.
7	--	--	--	--	--	Melt stopped because of bridge formation; restarted and ingot pulled away from the bridge.
8	14.1	28.4	100	3.5	187-189	Crucible segment gaps reduced to 0.13-0.25 mm; recoated with ZrO ₂ ; starting stub position at 7 cm below crucible top; power increased to 94 kW when flux began to melt; rough ingot side-wall.
9	15.9	45.3	110	2.4	--	Lost power due to momentary low water pressure in heat exchanger cooling water; regained molten pool within 30 seconds; rough ingot sidewall.
10	15.5	51.8	110	2.1	185-186	Good melt; rough ingot sidewall.
11	17.8	56.5	110-120	1.9-2.1	183-186	120 kW increases the instability of the cone-shaped pool; vapor-degreased hulls used; rough ingot sidewall.
12	19.3	~38.5	~110	~2.9	--	Melting OK; rough ingot sidewall.
13	20.5	47.2	108-110	2.3	187	Melting OK; rough ingot sidewall.

a. Heat numbers one and two are stainless steel and mild steel, respectively.

b. Brinell Hardness Number

c. United States Bureau of Mines

REFERENCES

1. A. M. Platt, compiler, Nuclear Waste Management Quarterly Progress Report October Through December 1977, PNL-2377-4, Battelle, Pacific Northwest Laboratories, Richland, WA, May 1978.

2.0 MONITORING METHODS FOR PARTICULATE AND GASEOUS EFFLUENTS FROM WASTE-SOLIDIFICATION PROCESSES

R. W. Goles - Program Manager

The objective of this program is to develop reliable and accurate techniques for process monitoring and stack emission monitoring for future waste solidification facilities.

SUMMARY

During this reporting period, a search was conducted for a suitable B-cell process off-gas sampling site, an optimum location was established, and all necessary process line modifications were started. Specific sampling methods for selected effluents were identified, and a conceptual design of the sampling network to be used for monitoring the vitrification off-gas stream was established. In addition, particle collection studies, conducted under simulated process line conditions, were continued.

SAMPLING SITE LOCATION -

D. C. Hamilton, H. G. Rieck, Jr.,
and R. W. Goles

A search was made for a suitable off-gas sampling site, based upon the need to obtain representative samples and to locate the site as close to the process hot cell as is physically possible. These conditions were most closely satisfied by establishing the sampling site within Room 11 of the 324 Building. In the piping arrangement, the sampling point will be located along the axis of the 3-in. process line, POG-88,

at an elbow just 6-in. from where the process line enters Room 11. In the process line schematic shown in Figure 2.1, this sampling site lies between the E101 condenser and the preheater, which is designated E104. Since Room 11 is a radiation zone, most sampling equipment will be in Room 147 (directly above Room 11) where the sampled off-gas will be conveyed via a 3/4-in. heated transport tube as depicted in Figure 2.2.

The POG-88 process line will be sampled via a tapered 3/4-in. schedule-40 stainless steel pipe installed along the central axis of the 3-in. off-gas process line. Beyond the sample inlet will be a large orifice ball valve, a filter, and then a transition to a 3/4-in. SS tube. The tubing will be routed to the ceiling of Room 11 and through a proposed 2-in. access hole connecting with Room 147 directly above. A ball valve, a five-port sampling manifold, and a variable flow pumping system will complete the inlet portion of the off-gas sampling line. The sampled off-gas will then return to Room 11 via a 3/4-in. SS tube; there it will be returned to off-gas process line, POG-88, between absolute filter F104 and preheater E104. This

FIGURE 2.1. WSEP Auxiliary Process System

FIGURE 2.2. Process Line Sampling Site

closed sampling system just described is schematically illustrated in Figure 2.2.

SAMPLING NETWORK - R. W. Goles

The sampling systems developed for this phase of the monitoring program are intended to characterize the concentration of selected effluents escaping the process hot cell. As such, these sampling systems are highly sensitive, integrating batch mode devices. The effluents to be sampled during solidification were enumerated in BNWL-2245, which identified effluent species likely to escape the in-cell clean-up devices. These species are:

- Gaseous Effluents

- ^3H as HTO , HT and organics
- ^{14}C as CO , CO_2 and organics
- ^{129}I as I_2 , HOI and organics
- NO_x

- Semivolatile Effluents

- ^{79}Se
- ^{99}Tc
- ^{106}Ru
- ^{125}Sb , ^{126}Sb
- $^{125\text{m}}\text{Te}$, ^{127}Te
- ^{134}Cs , ^{135}Cs , ^{137}Cs

- Particulate Effluents

- Fission products, including $^{144}\text{CePr}$

- Activation products

- Heavy elements.

Where appropriate, several chemical forms of a given isotope will be selectively sampled for in order to extract as much information concerning the process as is possible (see PNL-2377-4). For example, various combinations of sorbing species and catalytic agents will permit selective sampling of three independent chemical forms of each of the following isotopes: ^3H , ^{14}C and ^{129}I . These include: tritiated forms of

water, hydrogen, and organics; ^{14}C -labeled carbon monoxide, carbon dioxide and organics; ^{129}I -labeled iodine, hypoiodous acid, and organics. All the semivolatile isotopes just listed will similarly be sampled for, using various combinations of scrubbing and sorbing agents to ensure quantitative removal of effluent from the sampled process off-gas. Examples of species-selective sorbing agents to be used in this sampling network include: caustic, silica gel, charcoal, ferric oxide, silver zeolite, and various types of molecular sieves.

Particles will be collected in a more or less conventional manner. However, special attention has been given to ensuring that as a representative a sample as possible is collected. Sampling will be accomplished with a 3/4 in. Pitot tube located along the axis of the 3-in. process line, POG-88. The sampled gas and all entrained particles will move along a uniform diameter pipe directly to a commercially available filter of known collection efficiency. This filter will be located as close as physically possible to the process line and will lie along a central axis coincident with the axes of the Pitot tube and the process line itself, thereby ensuring a straight, short shot to the filter. Because erratic changes are expected to occur in process line flow rate, a truly

active control network has been designed to match the sampling rate to the instantaneous process off-gas velocity and thereby ensure continuous, isokinetic sampling conditions.

Figure 2.3 is an abbreviated schematic of the entire sampling system. Details of the various parallel sampling lines appear in Figures 2.4-2.7.

As previously mentioned, most sampling devices of the system are of the integrating batch-mode variety that require subsequent laboratory analysis. However, a Ge(Li) gamma-ray detector will also be deployed as a quasi-real time, general purpose survey instrument analytically applicable to all gaseous, gamma-emitting radionuclides. This analytical subsystem is illustrated in Figure 2.7.

FILTER EVALUATIONS - D. C. Hamilton and H. G. Rieck, Jr.

The testing and evaluation of various commercially available particulate filters continued during this quarter. These studies are concerned mainly with the determination of filter behavioral characteristics and efficiencies as a function of velocity, pore size, particle size, filter type, and loading. The experimental procedures involved in this testing have been detailed in previous reports (PNL-2377-2 to 2377-4). The results of the current tests are summarized in Table 2.1.

FIGURE 2.2. Process Line Sampling Site

closed sampling system just described is schematically illustrated in Figure 2.2.

SAMPLING NETWORK - R. W. Goles

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water, hydrogen, and organics; ^{14}C -labeled carbon monoxide, carbon dioxide and organics; ^{129}I -labeled iodine, hypoiodous acid, and organics. All the semivolatile isotopes just listed will similarly be sampled for, using various combinations of scrubbing and sorbing agents to ensure quantitative removal of effluent from the sampled process off-gas. Examples of species-selective sorbing agents to be used in this sampling network include: caustic, silica gel, charcoal, ferric oxide, silver zeolite, and various types of molecular sieves.

Particles will be collected in a more or less conventional manner. However, special attention has been given to ensuring that as a representative a sample as possible is collected. Sampling will be accomplished with a 3/4 in. Pitot tube located along the axis of the 3-in. process line, POG-88. The sampled gas and all entrained particles will move along a uniform diameter pipe directly to a commercially available filter of known collection efficiency. This filter will be located as close as physically possible to the process line and will lie along a central axis coincident with the axes of the Pitot tube and the process line itself, thereby ensuring a straight, short shot to the filter. Because erratic changes are expected to occur in process line flow rate, a truly

active control network has been designed to match the sampling rate to the instantaneous process off-gas velocity and thereby ensure continuous, isokinetic sampling conditions.

Figure 2.3 is an abbreviated schematic of the entire sampling system. Details of the various parallel sampling lines appear in Figures 2.4-2.7.

As previously mentioned, most sampling devices of the system are of the integrating batch-mode variety that require subsequent laboratory analysis. However, a Ge(Li) gamma-ray detector will also be deployed as a quasi-real time, general purpose survey instrument analytically applicable to all gaseous, gamma-emitting radionuclides. This analytical subsystem is illustrated in Figure 2.7.

FILTER EVALUATIONS - D. C. Hamilton and H. G. Rieck, Jr.

The testing and evaluation of various commercially available particulate filters continued during this quarter. These studies are concerned mainly with the determination of filter behavioral characteristics and efficiencies as a function of velocity, pore size, particle size, filter type, and loading. The experimental procedures involved in this testing have been detailed in previous reports (PNL-2377-2 to 2377-4). The results of the current tests are summarized in Table 2.1.

FIGURE 2.3. Process Line Sampling System

FIGURE 2.4. Multi-Species Sampling Network

FIGURE 2.5. Multi-Stage Sorption Line

FIGURE 2.6. Grab Sample Facility

FIGURE 2.7. On-Line γ -Analysis System

TABLE 2.1. Filter Evaluation Results

Filter Type	Length of Run, hr	Gas Velocity, m/sec	Mass of Deposit, mg	Volume of Air, m ³	Efficiency, %
Gelman (Glass)	1	2.8	2.6	11.4	99.3
Gelman (Glass)	1	1.4	9.6	5.8	99.8
5 μ m Millipore	1.5	2.8	72.4	17.1	>99.9%
5 μ m Millipore	1.5	1.4	61.1	8.7	>99.9

3.0 TRU WASTE IMMOBILIZATION PROGRAM

J. H. Jarrett - Program Manager

The objectives of this program are to determine the degree of immobilization that may be necessary for the acceptable disposal of transuranic waste residues, and to develop process technologies to provide such immobilization.

The scope and objectives of this program were extensively reviewed by Battelle and DOE during this reporting period, and program activities were suspended pending the results of that review. As revised guidance has now been received from the sponsor, the program is presently being defined in

terms of detailed work tasks. The primary thrusts will be to determine the degree of immobilization required for acceptable disposal of transuranic wastes and to develop appropriate process technology to provide such immobilization.

4.0 KRYPTON SOLIDIFICATION

G. L. Tingey - Project Manager

The objective of this program is to develop alternative techniques for storing radioactive krypton in a solid matrix. Two techniques are currently being investigated: 1) dissolution of krypton in low-density glass followed by densification at high temperatures and 2) entrapment of the krypton in solids during ion sputtering.

SUMMARY

Krypton has been loaded in several candidate metals by sputtering with the krypton ions. No clear pattern has as yet emerged from these studies which will allow us to design the proper alloy for maximum trapping of gas. However, several alloys appear to be feasible. Preliminary gas release kinetics show excellent gas retention. Further improvements could also be made by coating the surface with low-gas-containing metal. Scale-up tests appear to be progressing satisfactorily.

Krypton solubility in low-density glass appears to be adequate for storage of ^{85}Kr , but the release rates are rather large. Higher pressure tests are planned for the coming quarter.

KRYPTON ENTRAPMENT DURING SPUTTERING -

M. A. Bayne, E. D. McClanahan,
G. L. Tingey, E. N. Greenwell, and
R. W. Moss

One goal of this program is to maximize the concentration of krypton entrapped in a matrix during sputtering. The higher the loading, the

more economical the process will be, both for the sputtering process and for ultimate storage. Previous work has shown that higher loadings are achieved in metals with a glassy structure than those with crystalline structures.⁽¹⁾ Several materials that were expected to yield the desired glassy structure when sputter-deposited have also been identified.⁽²⁾ Sputter deposits of most of these materials have now been prepared, and the results are presented in Table 4.1. One salient feature is that additions of refractory metal to the target have an unpredictable effect on gas uptake. Uptake increased when tantalum was added to Fe-Si but decreased when added to Fe-Zr. Also, the $\text{Fe}_{.85}\text{Y}_{.13}\text{Me}^{*}.02$ alloy still seems to be the best candidate based on gas uptake although $\text{Fe}_{.26}\text{Zr}_{.73}$ appears promising. The iron alloys tested thus far for gas trapping ability have tended to adhere poorly to some electrode shields. The flaking of this nonadherent material obstructs operation of the sputtering equipment. The next series of alloy development experiments will substitute nickel

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*Me = minor impurity

TABLE 4.1. Summary of Experiments in Developing Amorphous Alloys for Use in ^{85}Kr Storage

Material	Sticking Coefficient 225 eV, atoms/ion	Average Krypton Content,		Net Sputtering Yield 2.5 keV, g/amp-hr	Minimum Gas Release Temp., °C
		cm ³ /g	at. %		
$\text{Fe}_{0.85}\text{Y}_{0.13}\text{Me}^{(a)}_{0.02}$	0.20	31.9	9	5.3	500
$\text{Fe}_{0.95}\text{Y}_{0.03}\text{Me}_{0.02}$	0.028	(b)		(b)	NA
$\text{Fe}_{0.69}\text{Si}_{0.25}\text{Me}_{0.06}$	<0.01	NA ^(c)		5.04	NA
$\text{Fe}_{0.67}\text{Si}_{0.24}\text{Ta}_{0.03}\text{Me}_{0.06}$	0.016	0.64		3.25	NA
$\text{Fe}_{0.67}\text{Si}_{0.24}\text{Cr}_{0.04}\text{Me}_{0.05}$	Experiment aborted: deposit peeled badly				NA
$\text{Fe}_{0.78}\text{Zr}_{0.20}\text{Me}_{0.02}$	0.09	18.5	5.4	5.43	500
$\text{Fe}_{0.26}\text{Zr}_{0.73}\text{Me}_{0.01}$	0.20	22.1	8.1	3.15	(d)
$\text{Fe}_{0.72}\text{Zr}_{0.21}\text{Ta}_{0.05}\text{Me}_{0.02}$	0.075	15.7	4.9	6.08	(d)

a. Me: minor or impurity elements

b. Not calculatable due to experimental problems

c. NA: not applicable

d. Not yet measured

alloys which, in past experience, have exhibited better adherence properties. The nickel/iron material cost increase could easily be offset by steadier equipment operation, and, in addition, the kryptonated nickel alloys will probably have a higher thermal stability and corrosion resistance. Targets are being fabricated.

Gas release has been measured on a sample of the krypton-loaded $\text{Fe}_{0.78}\text{Zr}_{0.20}\text{Me}_{0.02}$ and preliminary release kinetics determined. The release rate was too low below 500°C to obtain accurate rates. However, extrapolation of a plot of the log of the release rate versus reciprocal temperature yielded a fractional release at 400°C of $2.0 \times 10^{-8} \text{ min}^{-1}$. At this rate 10% of the krypton would be released in about 10 yr. Further release data are now being determined and the release kinetics analyzed.

Work is continuing to develop techniques and apparatus which will permit us to scale up the process. Further development of the new target arc-limiters was required after the results of initial tests on the sputtering apparatus were analyzed.

Redesign of the grid circuit of the high-voltage tube in the limiter so as to support a drive voltage more proportional to the plate current has completely eliminated the 100-to-200-MHz rf oscillations. Testing at sputtering voltages over 1.8 kV then revealed a tendency of the limiter to briefly turn on immediately after cycling off. This occurred with some new 2-MHz oscillations, which became much worse at the 2.5-kV voltages normally used. Minor changes in the plate and grid circuits have eliminated these effects under load bank testing.

Further testing on a sputtering system will determine whether these changes are sufficient.

During this quarter we have begun the design of a sputtering chamber having twice the gas trapping area of the existing chamber but using most of the existing components (shields, targets, power supplies, etc.). This larger chamber will permit us to test the target current limiters at full output current, to test the concept of segmented target and substrate for product optimization, and to test the whole scheme under increased gas usage conditions ($\sim 30 \text{ cm}^3/\text{min}$ at STP).

Also for scale-up, most of the components for the closed-cycle gas handling and sputtering system for use in a production unit have been received except for the vacuum pump itself. It is now expected in early May.

This quarter a paper entitled "Krypton Entrapment in Pulse Biased Sputter-Deposited Metals" (PNL-SA-6698) has been prepared. It is being presented at the International Conference on Metallurgical Coatings, San Francisco, California, April 3-7, 1978. The abstract of the paper is attached.

KRYPTON DISSOLUTION IN LOW-DENSITY GLASS - W. J. Gray and G. L. Tingey

Early attempts to dissolve krypton in a glass matrix confirmed the reported low solubility of inert gases in glass. Therefore, experiments have been conducted to determine the solubility in low-density glasses. For this work the krypton is loaded

in porous glass rods which are then densified by heating to sintering temperatures while retaining the krypton atmosphere. We have now performed five experiments with krypton loading pressures of 600 to 1000 torr at 450°C . The samples were then sintered at selected temperatures and times to produce sample densification from 6 to 13%. The krypton content generally decreased as the samples were densified while the release temperatures increased. The krypton content in the most highly densified sample was about $1 \times 10^{-3} \text{ cm}^3$ of $\text{Kr(STP)}/\text{g}$ of glass when loaded in a krypton atmosphere at 600 torr. Linear extrapolation to 2000 atm yields a value of 25 cm^3 of $\text{Kr(ST)}/\text{g}$ of glass. Although this krypton loading is not exceptionally high, it would be acceptable for storage of ^{85}Kr . Krypton release measurements for these samples appeared to be quite variable, with significant release occurring below 500°C in all samples examined.

Equipment is now being prepared that will allow krypton loading at pressures up to 33 MPa (5000 psi) and temperatures up to 900°C . Krypton loadings using this equipment are expected to be high enough to allow much better determination of krypton release as a function of temperature. The high-pressure loading will also minimize the uncertainty due to that krypton which is merely adsorbed on surfaces. Finally, it will allow greater confidence in extrapolation of krypton loadings to 20,000 or 30,000 psi.

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KRYPTON ENTRAPMENT IN PULSE BIASED SPUTTER-DEPOSITED METALS

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Abstract

A supported-gas-discharge sputtering system was constructed to investigate krypton entrapment in high-rate, sputter-deposited thick films. Krypton entrapment was studied as a function of substrate and target voltage for nickel, aluminum, titanium and iron deposits. Control of substrate and target voltages was achieved with pulse circuits capable of adjusting the pulse duration and repetition rate. A water-cooled, cylindrical copper substrate 15 cm in diameter was used to collect the sputtered metal from a 9-cm-dia cylindrical target. To observe the immediate effect of changes in sput-

tering parameters on gas trapping, as well as measure the total gas trapped, a sensitive mass flow indicator was installed in the krypton supply of the dynamically pumped system. Data relating krypton content to substrate bias conditions and deposition rate are discussed in light of the theory of Carter, Colligon and Leck on ion absorption in the presence of sputtering. Krypton contents >2 at.% were found in the metals studied, all of which were deposited at rates exceeding 1 nm/sec. Computed krypton concentrations agreed with concentrations in samples that were analyzed chemically.

5.0 CARBON-14 AND IODINE-129 FIXATION

L. L. Burger - Project Manager

The objective of this program is to develop and demonstrate the technology needed to safely and effectively immobilize iodine-129 and carbon-14 released from the treatment of irradiated fuels.

SUMMARY

Studies of iodide oxidation to iodate as a prefixation step have shown that none of the methods is completely satisfactory. However, several show promise. They include the use of ozone, chlorine, concentrated nitric acid, and electrolysis. A survey of alternative fuel cycles indicates that the fixation study program is generally applicable.

The effect of leachant volume and change rate on leaching of iodine compounds from cement has been evaluated, and the investigation of other leaching parameters is under way.

CONVERSION OF IODIDE TO IODATE -

R. D. Scheele

Except for the Iodox process, iodine removal systems will normally result in an iodide as a trapped form. If an iodate is selected as the final fixation form for iodine, a method will be needed to oxidize iodide to iodate. The optimal process for conversion of iodide to iodate would use an oxidant which is compatible with the stainless steel environment of a plant and would introduce no additional waste products. Some of the

methods which appeared promising thermodynamically and/or were discussed in the literature were studied. Table 5.1 presents pertinent oxidation-reduction couples in acid solution.

Kahn and Wahl⁽¹⁾ discuss cerium (IV) oxidation of microquantities of iodide to iodine, iodate, and periodate. They found that iodide ion treated with a hot sulfuric acid-cerium (IV) solution forms iodate as the principal product.

Bray and Liebafsky⁽⁴⁾ and Bray and Caulkins⁽⁵⁾ describe the conditions necessary for conversion of iodide to iodate using hydrogen peroxide. In acid solution, hydrogen peroxide is both an oxidant and reductant; however, according to Bray, the oxidation of iodine to iodate is sufficiently fast to overshadow the reduction of iodate to iodine and ensure nearly quantitative iodate formation.

Merritt⁽⁶⁾ found that ozone quantitatively converted iodide to periodate in alkaline solutions. In carbonate-buffered solutions iodate was the principal product, with some formation of periodate and a white iodine oxide fume which he described as iodine pentoxide.

TABLE 5.1. Oxidation Reduction Couples in Acid Solution

Couple	E°, Volts	Reference
$I^- + 3H_2O \rightarrow IO_3^- + 6H^+ + 6e^-$	-1.085	2
$I^- \rightarrow 1/2 I_{2(aq)} + e^-$		
$1/2 I_{2(aq)} + 3H_2O \rightarrow IO_3^- + 6H + 5e^-$	-0.63	3
$Cl^- \rightarrow 1/2 Cl_2 + e^-$	-1.36	3
$Br^- \rightarrow 1/2 Br_2 + e^-$	-1.09	3
$2H_2O \rightarrow H_2O_2 + 2H^+ + 2e^-$	-1.77	3
$Ce^{+3} \rightarrow Ce^{+4} + e^-$	-1.61	3
$O_2 + H_2O \rightarrow O_3 + 2H^+ + 2e^-$	-2.07	3
$O_2 + 2OH^- \rightarrow O_3 + H_2O + 2e^-$	-1.24	3
$I^- + 6OH^- \rightarrow IO_3^- + 3H_2O + 6e^-$	-0.26	3

Nitric acid oxidation of iodide to iodate in mercuric nitrate-nitric acid solution has been studied by Oak Ridge National Laboratory,⁽⁷⁾ Hanford Engineering Development Laboratory,⁽⁸⁾ and Pacific Northwest Laboratory.⁽⁹⁾ ORNL found rapid conversion of iodide to iodate during reflux of 0.01M I^- in 12 and 14M HNO_3 $Hg(NO_3)_2$ solutions. HEDL found 9 to 16% of the iodine volatilizing during reflux of 6 to 11M HNO_3 , 0.1M $Hg(NO_3)_2$, and 0.01M I^- solutions. The remainder converted to iodate. Also mentioned, but not described, is a volume reduction process in which no iodine is volatilized. Mailen and Tiffany studied the oxidation of iodine with concentrated nitric acid (hyperazeotropic) and concluded that a two-stage oxidation occurred, forming first I^+ then the iodate.

Electrolytic oxidation of iodide to iodate is used on an industrial scale for iodate production.⁽¹⁰⁾ Smith⁽¹¹⁾ describes an electrolytic

technique and cell for oxidation of iodine to iodate using a platinum anode. Mailen and Horner⁽¹²⁾ employed electrolytically oxidized cobalt to trap iodine from a gas stream.

Cerium oxidation would have a unique advantage in that either Ce(III) or Ce(IV) iodate could be used as a storage form. However, our experiments in which we refluxed 0.1M KI solutions of 0.05M and 2M ceric ammonium nitrate were unsuccessful. Very little iodate was formed. We were able in only one case to obtain quantitative formation of iodate using acidic peroxide as the oxidant. In most cases we found the reaction to be two-step, vigorous once started, and difficult to control. The solution foamed, making containment difficult. In addition, the heat generated by the reaction caused rapid evolution of iodine. Even though the reaction was vigorous, complete conversion of iodide to iodate was very slow, evidently due to the competing reduction of iodate by peroxide.

Experiments were performed with ozone (~3 wt% in air) with both acid and alkaline solutions of iodide. White fumes, which were carried out of the reaction vessel by the gas stream, formed in each instance. Their formation and the loss of iodine was least in alkaline solution. Refluxing increased the fume formation. Cotton and Wilkinson⁽¹³⁾ describe the iodine oxide formed by ozone oxidation of iodine as I_4O_9 , and the properties of the white fume observed in our experiments seem to fit I_4O_9 better than I_2O_5 .

Difficulties with ozone oxidation of iodide could possibly be overcome. The I_4O_9 could be trapped or its loss eliminated by redesign of the ozonizer. Also, a periodate could be used as the fixation form, or it could be thermally decomposed to oxygen and iodate.

Chlorine was the only oxidant tried which quantitatively converted iodide to iodate. Bromine oxidation was unsuccessful. Unfortunately, chlorine and its byproducts are not compatible with stainless steel.

We have previously shown that electrolytic oxidation can be used to convert HgI_2 to $Hg(IO_3)_2$ but have not yet examined it for the present application.

^{14}C And ^{129}I in Alternative Fuel Cycles - R. D. Scheele and L. L. Burger

Most proposed alternative fuel cycles would employ oxides or carbides of thorium, uranium, plutonium, or mixtures of them,⁽¹⁴⁾ both as first generation and as recycle fuel. Dis-

solving operations will not differ greatly from those of the Purex process. Head-end steps, if employed, will vary somewhat, but the gaseous products will remain unchanged. Chemical separations will employ either Purex or Thorex flowsheets, modified for nonseparation of plutonium or low fission product decontamination or both.

In each case, most of the carbon-14 is expected to be emitted as CO_2 during the dissolver operation. Reduced forms that may be released during yet unidentified head-end steps can be easily oxidized to CO_2 .

Of the iodine which remains after any head-end treatment, 99% will be evolved from the dissolver.

For these fuel cycles the off-gas treatments leading to trapping of iodine and carbon-14 will employ similar techniques. These include: metal-impregnated zeolites or other silica products, the Iodex process, mercuric nitrate scrubbing, alkali scrubbing for iodine and molecular sieve or scrubbing with an alkali solution or alkali-alkaline earth hydroxide slurry for CO_2 . The fixation and storage problem remains unchanged.

With the HTGR the problem of controlling carbon-14 is different because of the large volumes of CO_2 produced by burning the carbon fuel particle coatings and graphite blocks. The resulting activity level is low, $\sim 1.3 \times 10^{-6}$ Ci $^{14}C/g$ carbon. Snyder and Kaye⁽¹⁵⁾ have described the partitioning of carbon-14 in the treatment of irradiated HTGR fuels and evaluated several modes of release. For the large quantities involved the

most inexpensive and readily available fixation form is desirable. This may be calcium carbonate. Its use for disposal of contaminated CO_2 has been evaluated by Groff.⁽¹⁶⁾

Leachability Studies - K. D. Wiemers

To select an appropriate iodine-129 and carbon-14 fixation material, leachability properties are to be added to the previously collected evaluation parameters: thermal and chemical stability, solubility, cost, and availability.⁽¹⁷⁾ The first leachability properties to be studied are the leach rates of iodate, iodides, and carbonates from cement under dynamic leaching conditions.

Phase I of this study was statistically designed to evaluate factors which would provide conditions approaching those of dynamic leaching. Dynamic conditions were desired to maximize the leach rate and to allow for interpretation of leach data using mass transport phenomena.⁽¹⁸⁾ A standard leach testing method has been proposed by the International Atomic Energy Agency.⁽¹⁹⁾ However, studies conducted at Brookhaven National Laboratory⁽²⁰⁾ and ORNL⁽²¹⁾ indicate that the leach rate is dependent upon some of the recommended experimental procedures. In particular, the factors chosen to be studied in Phase I were the leachant volume and frequency of leachant renewal. The experimental conditions used were:

- sample: 10% by weight $\text{Sr}(\text{IO}_3)_2$ in Portland type II cement
- water/cement ratio, w/c = 0.37

- during method: 15 days in humid environment
- average sample volume, V: 14.2 cm^3
- average exposed surface area, S = 32.5 cm^2
- leachant: CO_2 -free, redistilled water, pH = 6.5 to 7.3
- total time of experiment: 49 days.

Table 5.2 shows the factors evaluated. Trials 1-8 were run in duplicate. Trial 0 had four samples. Analysis for the iodate ion in the leachant was carried out by reacting an acidified aliquot of the leachant with excess KI and determining the I_3^- ion spectrophotometrically.

The following explains the chosen boundary conditions. The selected leachant renewal frequency for the first 14 days of leaching was between the limits suggested by IAEA (once a day) and ORNL (three times a day). Leach periods of 8 and 16 hr were chosen to adapt to the working day. After 14 days, the frequency of leachant renewal was decreased to provide concentrations of iodate in the leachant within detection limits of the analytical method. The revised frequency of leachant renewal was patterned in such a way as to allow the statistical analysis to continue. The leachant volumes ranged from the minimum volume (250 ml), physically possible to ensure full coverage of the sample, to a volume (550 ml) about five times that recommended by the IAEA.

Leach data from the first 14 days of leaching has been statistically evaluated. The leach rate was calculated as the cumulative fraction

TABLE 5.2. Factors Evaluated in Phase I of Cement Leaching Study

Trial No.	Frequency of Leachant Renewal (per 8-hr period)	Volume of Leachant, ml	
		8-hr Period	16-hr Period
1	1	250	300
2	3	250	300
3	1	450	300
4	3	400	300
5	1	250	550
6	3	250	550
7	1	400	550
8	3	400	550
0	2	325	425

of iodate leached multiplied by volume to surface ratio: $(\sum a_n/A_0)(V/S)$, experienced as a function of t_n .

a_n = iodate lost during nth leach period

A_0 = initial iodate present in specimen

V = volume of specimen, cm^3

S = exposed surface area, cm^2

t_n = duration of leachant renewal period.

The results indicate a positive effect of both the leachant volume and leachant renewal frequency on the leach rate. The leachant volume during the 16-hr leach period had the greatest effect. The effect of leachant volume and leachant renewal frequency is exemplified in Figure 5.1, in which the cumulative fraction of iodate leached multiplied by the volume to surface ratio is plotted as a function of total leach time. The two trials shown are those in which the factors studied were either all maximum or all minimum values, trials 8 and 1 in Table 5.2, respectively. After 14 days of leaching, the leach rate for the set

of minimum conditions was 77% that of the maximum conditions. The leach rates of $\text{Ba}(\text{IO}_3)_2$ obtained by ORNL⁽²²⁾ and the maximum leach rate for $\text{Sr}(\text{IO}_3)_2$ over a 14-day leach period were of the same order of magnitude.

The inflection in the curves at the fourth day and the less significant one at the eleventh day, correspond to the weekends, when the frequency of leachant renewal pattern was interrupted. In future experiments, this discontinuity will be avoided.

With these data, the factors needed to approach a dynamic leaching environment can be estimated. A dynamic leach test will then be initiated to statistically screen 17 variables suspected of influencing the leach rate of iodate from cement. These variables are outlined as follows:

- Cement composition
  - water/cement ratio
  - curing time
  - sodium silicate coating
  - commercial sealant
  - aluminum chloride additive
  - bentonite additive
  - butyl stearate additive

FIGURE 5.1. Combined Effect of Leachant Volume and Frequency of Leachant Renewal on Cumulative Leach Rate

- Fixation material
  - percentage of fixation
  - silver nitrate additive
  - excess cation additive
  - barium nitrate additive
  - variable solubility of fixation material
- Environmental condition
  - temperature
  - initial pH
  - leachant composition
(Ca^{+2} , SO_4^{-2} , and CO_2)

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6.0 INTERNATIONAL PROGRAM COORDINATION

K. M. Harmon - Project Manager

Promote waste management technology exchange between the United States and other nations through the establishment of an international technology coordination center at PNL.

SUMMARY

Collection, review, and summarization of significant documents continued. An annual report (Summary of

National and International Radioactive Waste Management Programs, by K. M. Harmon, PNL-2598) was prepared and issued.

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7.0 WASTE MANAGEMENT SYSTEM STUDIES

J. W. Bartlett - Program Manager

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

SUMMARY

Information contained in our seismic data base was extended, and capability to use the information was improved by developing capability for stereographic projection and color coding the data. Mapping capability was also extended and refined.

A draft report was compiled on results of the knowledge/attitudes survey of Washington residents. The final revision of the report on print media coverage of nuclear power and nuclear waste management was also completed. Analysis of TV coverage and DOE's information program was started.

In work on perceived risk, distribution of the risk perception/risk evaluation questionnaire continued, and preliminary data analysis was begun. Development of survey instruments for evaluating voluntary risk and perceived distribution of risk was started.

DATA BASE DEVELOPMENT FOR NUCLEAR WASTE MANAGEMENT - H. P. Foote and T. N. Bishop

Seismic Data Base

Geophysics can aid geology in the study of seismic risk in four basic ways:

1. alignment of hypocenter locations in three dimensions
2. calculation of recurrence curves and other statistical studies of earthquakes as a function of time, magnitude, and depth
3. measurement of ground acceleration
4. focal plane solutions from first arrivals of earthquakes.

The first two topics lend themselves immediately to a data base. The following briefly summarizes our work on these during the first two quarters:

- Seismic Hypocenter Alignment

The capability to look at earthquake hypocenters in three dimensions has been achieved through the use of stereoplotting. In addition to an earthquake file, additional files of grid lines, faults, continental boundaries, or other surface features can be computed or digitized. The first plot file is then created using any number of previously constructed files. One chooses the desired observation point and focal point of interest (both in spherical coordinates) along with the field of view. A second plot file is created by changing the observation point by roughly five degrees. These plot files can be output on a CRT scope or an electrostatic printer, or a

photographic negative. A paper describing the stereoplotting of seismic information will be submitted for publication.

Capability has been developed to plot stereo pairs on a high-resolution CRT screen. Moving a joystick permits the observation point to be changed quickly and dynamically. In this way, the earthquake data can be rotated so that one gets a three-dimensional look at the data. This is a useful aid in looking for hypocenter lineups along fault zones, with either the worldwide data or with local microseismic data.

An additional tool for graphic display of the earthquake data is a Comtal color image display device. We have overlaid stereo pairs on this device with different colors, one view red and one green. With pairs of red and green cellophane glasses, many people can view the earthquakes in stereo simultaneously. Another use of this device is to color code a third parameter such as depth or magnitude which can then be shown with the epicentral information. With a pseudocolor scale, many gradations of the parameter can be differentiated.

- Seismic Recurrence Plots

Recurrence plots (a histogram of the logarithm of the number of earthquakes as a function of magnitude) can now be plotted for any regions of the world. The selection and histogram programs have been combined so that one can

select a region in time and space, plot a histogram, return to refine the region, and then replot a histogram of the updated file. The program is currently being transferred from the PDP-11/45 to the PDP-11/55 to interface with the plotting programs.

To plot energy release, the equation $\log E = 11.4 + 1.5 M$ is often used, where M is surface wave magnitude. We plan to relate other magnitudes (body wave and local) to surface magnitude through equations derived from our data base and then have the capability to plot seismic energy release maps.

Seismic Data

The following summarizes our seismic data:

1. Worldwide (NOAA), 1938-1977, 130,000 events.
2. Eastern Washington (USGS - University of Washington), 1969-1977, 2000 events.
3. Western Washington (University of Washington), 1970-1974, 1200 events.
4. Pacific Northwest (Woodward-Clyde), 1827-1975, 2150 events (we have discarded 250 duplicate events after a thorough computer and hand search).
5. Canada (Dominion Observatory), 1580-1975, 8200 events.
6. California (California Division of Mines and Geology), 1900-1974, 40,000 events (this combines California Institute of Technology, USGS, and Berkeley data).

7. Rocky Mountain Area (Texas Instruments) - not yet in data base.
8. Offshore Pacific Northwest (Oregon State) - not yet in data base.
9. Dam Locations (Army Corps of Engineers) - not yet in data base (we hope to interface dam information with seismic risk calculations).
Inquiries for additional data are being made.

- DOOP - for use in combining data bases - it finds pairs of possible duplicate events.
- QEDIT - can printout or edit a binary earthquake file.
- XYPLT - can plot two variables from each earthquake from a binary file and make a least squares fit to relate one variable to the other.

Seismic Computer Programs

The following briefly describes our seismic programs at present:

- NEWQK - reads new seismic data from cards or tape into a binary file in the data base.
- SORT - sorts a binary earthquake file by location (Marsden Square), time or magnitude.
- QFILEQ - Creates a File-Q data and index files from sorted binary files.
- SELECT - selects earthquakes from a binary file by location, time, magnitude, depth, or other parameters; output can be a listing, a new binary file, or formatted plotting files containing longitude, latitude, and magnitude or depth.
- HIST - plots histograms or recurrence curves from a binary file.
- HIST2 - plots histograms as a function of two variables.
- ALLQK - selects earthquakes from File-Q files and can also plot histograms or have the same output as SELECT.

MAPPING AND DATA BASE PROGRAMS

In this reporting period, stereo and color mapping programs were completed for display of seismic data in three dimensions. Since about 1963, accurate recordings have become available of seismic events detected by the Worldwide Seismic Network. There are approximately 80,000 events of this type currently in our seismic data base. Their longitude and latitude are accurate to a few tenths of a degree, depths are accurate to 25 km. Since some of these events occur at depths of 700 km, there is a considerable amount of useful three-dimensional information in the data base. For this reason programs were written to display the hypocenter location data in ways that make the three-dimensional nature of the events apparent.

The first program (QK3D) provides an active stereo display on the VS-60 refresh CRT. The operator can control his apparent viewing position continuously with a "joystick" control. Figure 7.1 shows a typical display on this device. Here, the subduction zone beneath and to the north of Japan comes out very clearly in the stereo view.

FIGURE 7.1. VS-60 and Color Display Showing Seismic Activity in the Vicinity of Japan

Two types of viewers were constructed for this display. The first is a reduced version of a standard 9 by 9-in. viewer. This one is made to view images with a 6-in. separation. The second viewer is a pair of prism glasses. They use light refraction through the prisms to bring the two images together.

The second program (CLROK) is basically a mapping program with color coding capability. Figure 7.2 is a picture of the control portion of this program. It allows various types of data files to be selected and plotted in six different colors. The selections are made by light pen on the VS-60 display. The maps are plotted on the Spatial Data Display which appears on the right in Figure 7.1. Its resolution is somewhat less than that of the VS-60 display, but CLROK has color and a much greater

capacity for holding data. Its refresh memory is about 80,000 words, where typically only 3000 to 4000 words are available on the VS-60 scope. The two display devices complement each other nicely with little overlap in their functions.

Stereo pictures may also be plotted on the color display. Here the two images necessary for stereo are plotted in red and green. The image is then viewed with red and green filtered glasses. The display has the advantage of being viewable by several people simultaneously.

So far we have found the color plotting program most useful in presenting color-coded depth maps of seismic data. In these maps, a continuous range of color is used to indicate the depth of a seismic event. The scale can be varied at will so that small variations in depth can be

CODE	FILES	TYPE	OPTIONS
1 WHITE	1 DAI USA **Z	DEPTH	PLOT FILES
2 BLUE	8 *DAI GULF **Z	DEPTH	NEW REGION
3 GRAY	1 DAI POLARIS **Z	POINT	READ INDEX
4 GREEN	1 DAI FAULTS **Z	LINE	ERASE \$DS
5 YELLOW	1 DAI ASTROT **Z	LINE	LIST
6 RED	1 DAI ARSMT **Z	LINE	STOP
7 MAGENTA	1 DAI ASTRA **Z	LINE	
8 DEPTH, MAGNITUDE	1 DAI ASTRA **Z	LINE	
9 STEREO	5 *DAI WPLD **Z	LINE	
	7 *DAI WPLD GRD	LINE	
DEPTH CODE (KMS)	1 DAI STATES **Z	LINE	
BLUE ---- RED	1 DAI DEARSH **Z	DEPTH	
0 0 500 0	1 DAI DEARSH MAG	MAGNITUDE	
	1 DAI DEARSH **Z	DEPTH	
	1 DAI DEARSH MAG	MAGNITUDE	
MAGNITUDE CODE	1 DAI GUEST DEP	DEPTH	
BLUE ---- RED	1 DAI DA-ASA DEP	DEPTH	
0 0 0 0	1 DAI GUEST MAG	MAGNITUDE	
	1 DAI DA-ASA MAG	MAGNITUDE	
VIEWING ANGLES			
THETA = 10.3			
PHI = 11.5			

FIGURE 7.2. VS-60 Screen Showing Control Portion of Color Mapping Program

made to cover the entire color scale. We have recently used this feature in examining some newly acquired micro-seismic data in the San Francisco Bay area.

A digitizing program (DIG2) has recently reached an operational phase in its development. This program allows graphical data to be digitized and converted to an angular format. The files it generates are directly usable by the current mapping program. With it we can generate much higher quality map files than previously available from sources such as the Federal Government's World Data Bank II. We have begun digitizing the State of Washington from 1:250,000 maps and have the San Francisco Bay area digitized at 1:125,000. In both cases the average angular error is about 4×10^{-4} degrees. This corresponds to an error of about 150 ft,

which means that most of the original map accuracy has been preserved. Two examples of the mapping capability are given here:

- A partial map of the State of Washington compiled from USGS 1:250,000 maps (Figure 7.3).
- A map of the San Francisco Bay area and California coast compiled from three 1:125,000 maps and six 1:250,000 maps (Figure 7.4).

The Bay area map is overlaid with faults and microseismic events from the California data.

Figures 7.5 and 7.6 show how maps may be combined accurately into one file. Figure 7.5 is a plot of Clear Lake in northern California at a scale of 1:250,000. The upper part of the lake map is from one map, and the lower is from another. The second example is in the Sacramento River Delta area (Figure 7.6). Here the left part of the map is from a

This Figure is an illustration of the capabilities of BNW's digitizing and plotting routines. This map, which was digitized from seven 1:250,000 scale USGS mapsheets, shows the coastlines, lakes, and all non-intermittent rivers and streams in western Washington and Oregon. The output map is a Lambert Conformal Conic projection initially drawn at a scale of 1:1,000,000. The output map was drawn using only one-fourth the resolution available in the digitized files.

FIGURE 7.3. Digitized Map of the Coastline, Lakes and Rivers of Western Washington and Oregon

FIGURE 7.4. A Map of the San Francisco Bay Area and California Coast Compiled from Three 1:125,000 Maps and Six 1:250,000 Maps

FIGURE 7.4. A Map of the San Francisco Bay Area and California Coast Compiled from Three 1:125,000 Maps and Six 1:250,000 Maps

FIGURE 7.5. Composite Map of Clear Lake in Northern California

FIGURE 7.6. Composite Map of Sacramento River Delta Area

1:125,000 scale, and the right side is from a 1:250,000 scale with the composite reproduced at 1:250,000. It is the accurate transformation of the digitized X-Y coordinates to longitudes and latitudes that permit compilation of high-resolution map files for large areas from smaller maps.

NUCLEAR WASTE COMMUNICATIONS -
S. M. Nealey, HARC

The survey of Washington State residents conducted in the fall of 1977 neared completion during this quarter. A comment draft was completed containing the findings on nuclear waste knowledge, attitudes, values, and information sources. These findings will be packaged in two or three technical reports for distribution during the third fiscal quarter.

Regarding resident's knowledge about nuclear power and nuclear wastes, these major findings emerged:

- Pronuclear and antinuclear respondents knew about the same in terms of a total knowledge score, whereas respondents who had not yet formed an opinion on nuclear power had the least knowledge.
- Over 40% of the respondents thought that plutonium is the main fuel in a nuclear reactor.
- A majority of the respondents underestimate the current role that nuclear power plays in production of electricity in the United States.

- A majority of the Washington public knows what reprocessing is.
- About 25% of the Washington public still thinks that a nuclear reactor can explode like an atomic bomb.
- A large majority of the respondents are unaware of the differences between military and commercial wastes.
- A majority of the respondents believe that most commercial wastes are in liquid form, and a plurality believe that liquid wastes cannot be changed to any other form.
- A majority of the respondents were aware of present storage methods and future disposal plans.
- Respondents generally overestimate the amount of wastes and the length of time that most will remain radioactive.

Human values were found to be more closely related to one's nuclear power attitude than was knowledge about nuclear power. Significantly related to one's nuclear power attitude were one's value for: 1) a comfortable life, 2) a world at peace, 3) a world of beauty, 4) family security, 5) freedom, and 6) national security. Of further importance is the fact that the conflict over nuclear power seems to arise partly from pronuclear and antinuclear respondents' opposing opinions on nuclear power's relationship to a world at peace, a world of beauty, family security, and national security.

The final revision of the report on print media coverage of nuclear power and nuclear waste management

has been completed and is being submitted for sponsor clearance. Other than a few specific news sources, the print media has presented a somewhat balanced picture of the nuclear debate. Antinuclear stories were carried slightly more than pronuclear stories. To the extent that the print media influence public opinion about nuclear issues, that influence was judged to be somewhat negative. However, since most people seem to expect a preponderance of "bad news" in the print media, getting a preponderance of bad news about nuclear issues may not have much impact on attitudes.

The analysis of DOE's information program has gotten under way. Interviews were held with members of DOE's Office of Public Affairs in Washington, DC, and Germantown March 21-24. Information was gathered on program objectives, structure, and process of information dissemination. In general, we found that very little information regarding nuclear power is being disseminated by DOE and no information, except an occasional news release, is being disseminated regarding nuclear waste management.

The videotape indexing has been completed regarding network television news coverage of nuclear power, coal, and solar energy. About 20 hr worth of videotapes have been ordered from Vanderbilt University (9 hr nuclear, 10 hr coal, and 1 hr solar). A consultant is being hired to help finalize the television content analysis coding form. Next quarter the content analysis will be carried out on the nuclear news stories.

The report Public Policy Issues in Nuclear Waste Management is nearing completion with the addition of an annotated bibliography of a number of sources in which waste management issues are discussed. After completing this report we hope to make significant progress on the follow-on issues analysis in the next quarter.

PUBLIC PERCEPTION AND EVALUATION OF RISKS - J. A. Hébert and M. K. Lindell, HARC

Distribution of the combined risk perception/risk evaluation ("risk tradeoffs") questionnaire continued this quarter. Completion of the data collection phase has fallen behind the mid-February completion date. This is largely due to difficulties in obtaining satisfactory response rates from environmentalist groups. In some cases we have had to locate substitute groups or mail out a second wave a questionnaires to obtain a sample size large enough for our analyses.

Although the data files are not yet complete, preliminary data analysis has begun. Responses to the open-end items (Part II, Energy Alternatives) are being content analyzed to develop categories for coding. Questionnaires screened thus far indicate that energy technologies are generally evaluated according to three broad traits: safety, performance and cost. Although the responses show little differentiation among classes of cost, there is substantial discrimination among different aspects of performance (feasibility of technology,

availability of fuel supplies, etc.) and side effects (environmental impacts, risk to human health and safety, etc.). Because this result may be an artifact of aggregation, we will need to determine in further analyses whether this level of differentiation holds in the subgroups as well.

Survey instruments are being developed for evaluating voluntary risk and perceived distribution of risk. Draft versions have been prepared and limited pretesting conducted. The "risk evaluation" portion of this questionnaire will use a multitrait-multimethod (MTMM) approach. [The "traits" will be the factors, LTP (long-term public), STP (short-term public), LTO (long-term occupational) and STO (short-term occupational risk)]. The methods will be 1) utility analysis, 2) ratio weighting, and 3) attitude scaling. Utility analysis will make use of recently published global utility procedure. The ratio weighting will follow the same procedure as that used in BNWL-1997, and the attitude scaling will be a significant expansion on the "attitudes toward factors," also from BNWL-1997. The part of the question-

naire dealing with risk perception will address two major issues: 1) perceptions of components of risk and 2) beliefs about the factors studied in the risk evaluation portion of the questionnaire. In the "risk components" portion of the questionnaire, respondents will be asked to rate the relative level of hazard posed by each of a number of energy facilities with respect to several components of risk. These components of risk are being derived from the open-end responses to Section II of the "risk tradeoffs" questionnaire. The section on "beliefs about the factors" will assess the respondents' perceptions of the degree of risk associated with each of the four factors, LTP, STP, LTO and STO. Data on the respondents' beliefs about each factor can be compared with data on the relative importance of these factors to determine which set of variables, beliefs, or values would contribute most to overall public acceptability of nuclear waste processing alternatives. Work on the values stability questionnaire has been terminated due to delays in data collection for the risk perception/risk evaluation questionnaire.

8.0 WASTE MANAGEMENT SAFETY STUDIES

W. K. Winegardner - Project Manager

Evaluate the safety of alternative waste management schemes for viable nuclear fuel cycles up to the final disposal step; develop the methodology necessary to permit the safety assessment; identify portions of waste management systems which could benefit by safety design emphasis.

SUMMARY

An approach was established for initial, comparative safety assessments. The approach includes a scoping analysis on pretreatment waste streams as well as a more detailed analysis on management processes. Data needed for assessment of waste management alternatives within the once-through fuel cycle were assembled.

ASSESSMENT CONCEPTS AND METHODS -
 P. J. Pelto

An initial safety assessment approach for the Waste Management Safety Studies has been established. This approach includes a scoping analysis on pretreatment waste streams and a more detailed analysis on proposed waste management processes. The safety areas of primary concern include radiation exposures to the public from radionuclide releases from normal operations and potential accidents and occupational radiation exposure from normal operations.

The initial scoping analysis defines safety indices based on such factors as activity, volume, gamma power, heat generation, mobility, and

toxicity. These indices can provide useful insights on relative comparisons of waste streams within a fuel cycle and of different fuel cycles.

The more detailed analysis considers the waste streams in conjunction with their conceptual management systems. A preliminary checklist approach is suggested to identify and quantify safety areas of interest. These include processing conditions and inventories, required manpower, degree of containment, intermediate waste form characteristics, degree of remote handling, and radionuclide effluents. For well characterized waste management systems, quantitative information can be obtained. For some waste management systems, qualitative values can provide useful information. Building upon the above information, either the public radiation dose and the occupational radiation exposures are estimated directly from the waste process details or relative safety indices are established. A combination of a preliminary hazard analysis, along with a generic list of potential accidents, is used to assist in the quantification of the radiation exposure to the public from potential accidents.

SYSTEM DEFINITION/ASSESSMENT -

J. W. Voss

Assembly of fuel cycle data needed in assessing waste management system safety, costs, and availability continued. Two alternatives within the once-through fuel cycle are tentatively being considered. The first alternative calls for spent fuel to be discharged from the reactor when its basin is full and sent to an independent spent fuel storage basin where it is stored indefinitely. The second alternative considered is that spent fuel is discharged from the reactor when its basins are full, is shipped to a spent-fuel packaging facility, and then stored in a packaged spent fuel storage facility.

Figures 8.1 and 8.2 summarize tentative waste generation/accumulation characteristics as well as tentative fuel cycle capacities for the

two alternatives as they are assumed to exist for 200 GWe installed capacity.⁽¹⁻⁴⁾ Figures 8.3 through 8.6 show the throughput and inventory assumptions for major reference facilities of the two once-through alternatives. Figure 8.3 shows the reference reactor that will be studied. It is assumed to be a 1000-MW(e) pressurized water reactor (PWR). Standardized reactor designs available will be the basis for the study. Figure 8.4 shows the 3000-MTHM independent spent fuel storage basin to be studied, while Figures 8.5 and 8.6 show the spent fuel packaging and packaged fuel storage facilities, respectively. Each facility definition figure shows the wastes which are in inventory at any one time in a reference year as well as the total annual waste discharge from each facility. The total spent fuel inventory discharge or throughput is also shown in the figures.

THROUGHOUT THIS SERIES OF FIGURES:

* TREATMENT ALTERNATIVES ARE: TRASH - COMPACTION; WET WASTES - CEMENTATION; NCT - PACKAGE; CORE INTERNALS - PACKAGE; OFF-GAS - RELEASE

** WASTE TYPES ARE:

TRASH - COMBUSTIBLE AND COMPACTABLE TRASH
WET WASTES - CONCENTRATED LIQUIDS, PARTICULATE SOLIDS
AND WET WASTES
NCT - NONCOMBUSTIBLE AND NONCOMPACTABLE TRASH WITH
FAILED EQUIPMENT
CORE INTERNALS - SMALL EQUIPMENT AND MATERIALS WHICH
ARE REPLACED ANNUALLY IN CORE

*** RADIONUCLIDES ACCUMULATED IN INVENTORIES AT REACTORS
RANGE FROM 0 TO 5 YRS OLD. THOSE IN STORAGE BASIN AND
DRY CAISSON FACILITIES RANGE FROM 6 TO 18 YRS OLD.

FIGURE 8.1. Once-Through Fuel Cycle for Reactor/Basin Mode

FIGURE 8.2. Once-Through Fuel Cycle for Reactor/Packaging Storage Mode

FIGURE 8.3. Reference Reactor

FIGURE 8.4. Reference Independent Spent Fuel Storage Basin

FIGURE 8.5. Reference Spent Fuel Packaging Facility

FIGURE 8.6. Reference Packaged Spent Fuel Storage Facility

REFERENCES

1. Nuclear Assurance Corp., United States LWR Spent Fuel Inventory and Projections, Y/OWI/SUB-77/43500, June 1977.
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3. U.S. ERDA, Alternatives for Managing Waste from Reactors and Post-Fission Operations in the LWR Fuel Cycle, ERDA-7643, May 1976.
4. T. B. Mullarkey et al., A Survey and Evaluation of Handling and Disposing of Solid Low-Level Nuclear Fuel Cycle Wastes, Atomic Ind. Forum Report AIF/NESP-008, October 1976.

9.0 WASTE ISOLATION SAFETY ASSESSMENT PROGRAM

H. C. Burkholder - Project Manager

The objectives of the Waste Isolation Safety Assessment Program (WISAP) are to:

- 1. develop the generic methods needed to assess the long-term safety of geologic isolation repositories*
- 2. obtain the generic and site-specific data necessary to apply the methods*
- 3. demonstrate the use of the methods for a specific site*
- 4. apply the methods to make safety assessments for sites of the National Waste Terminal Storage Program*
- 5. analyze societal acceptance issues and develop methods of communicating assessment results which enhance the rational resolution of those issues.*

SUMMARY

Work during the quarter continued to 1) develop the repository simulation analysis computer code, 2) measure nuclide leach rates for unclad spent fuel and doped glasses, 3) convert, interface, improve, and test hydrologic, contaminant transport, and water-dose and air-dose computer codes, 4) measure the sorption behavior of biologically significant long-lived radionuclides, and 5) prepare reports for a number of societal acceptance-related studies.

TASK 1 - RELEASE SCENARIO ANALYSIS

- J. Greenborg and J. A. Stottlemire

A team of 16 geologic consultants was placed under contract this quarter and has begun developing submodels for the repository simulation computer program. The consultant categories

were discussed in the previous quarterly report. Each consultant was visited in February and briefed on WISAP objectives, work scope, and outputs. A geologic consultant's progress review workshop has been scheduled for June 12 and 13 at the Battelle Seattle Research Center.

A pilot repository simulation model has been formulated. It consists of equations which describe a layered earth model with provisions for the continuous update of the key repository parameters for each layer-thickness, pore pressure, permeability (intergranular and incremental fracture) and state of stress. Potentially disruptive phenomena modeled to date include climatic change, glaciation, meteorites, material accumulation and erosion, and faulting in the basement rocks.

The SIMSCRIPT II.5 simulation language software package has been procured and was made operational on the Hanford UNIVAC 1100 series computer. The repository simulation code will be programmed in this language.

TASK 2 - WASTE FORM RELEASE RATE DATA

- D. J. Bradley and R. P. Turcotte

Leaching studies on actinides in glass and unclad spent fuel continued. An open-face hood/glove box has been ordered for use in further actinide leach tests. A method was developed for producing leach specimens of devitrified waste glass that will be used later in the program. Characterization of the unclad spent fuel continued. Arrangements are being made to obtain clad spent fuel for leaching and autoclave studies. Four autoclaves were ordered and are expected to arrive during the next quarter. Following installation, they will be used to investigate the effect of in-situ repository conditions on leaching behavior.

Preparations to begin studying the leaching behavior of TRU waste forms of four types (cement, bitumen, urea-formaldehyde, and polymer) continued. A large glove box is undergoing some modifications to make it more compatible with its proposed function and to permit connection to existing high-efficiency particulate air (HEPA) filtration facilities. An automatic liquid scintillation counter for analysis of leach solutions has been ordered. Work was completed which investigated the incorporation of

three simulated TRU incinerator ash wastes in Portland type I cement. Final waste selection will be made after communications with personnel at the various DOE waste-generation locations.

A final experimental design was selected for investigating the effects of leachant flow rate and temperature on radionuclide release rates from glass. This design (featuring three groundwater types, three flow rates, and two temperatures) is adequate for a valid statistical analysis for leach rate versus time as affected by temperature, flow rate, and groundwater type. It allows for two degrees of freedom in estimating random variation for comparing the leach rates of three liquids.

TASK 3 - RELEASE CONSEQUENCE ANALYSIS

- J. R. Raymond and J. K. Soldat

Work continued during the quarter to convert, interface, test, and improve computer codes selected for use in the release consequence analysis system.

Work progressed to convert the GETOUT contaminant transport code and the dose models, ARRRG, FOOD, DACRIN, SUBDOSA, and KRONIC, to the new Hanford UNIVAC 1100/44 computer.

Conversion and testing of the PATHS program on the UNIVAC 1100/44 was completed. The batch part of PATHS was modified and tested to allow calculation of the sorption delay that is due to exchange of a single trace constituent with the porous material. The program was further modified to permit

calculation of the flow path length, and this feature was tested. A closely related improvement incorporated into the program allows greater flexibility in getting solutions for several path lines originating at different locations at different times.

Several test cases have been completed using PATHS. These involved steady, two-dimensional flow of water from repositories of different sizes under various flow conditions and with a considerable range of site characteristics. The wide range of conditions covered by the case gave considerable diversity in results. These test cases completely cover the expected range of direct, steady, two-dimensional flow systems from repositories to remote river-type outflow boundaries. Documentation of PATHS was started.

Documentation for the Davis FE hydrologic code was completed. Among the improvements documented were programs to permit retrieval of flow potential and permeability values for the various layers of the three-dimensional element matrix. These are required for impact to the MMT contaminant transport code. Testing of all of the Davis FE model input options such as surface flux, stream flux, nodal pumping stress, and element pumping stress continued. These options are being checked out on a realistic case and compared to an analog model output.

TASK 4 - NUCLIDE TRANSPORT DATA COLLECTION - R. J. Serne

Work has continued on evaluating various sorption-desorption methods. For selected minerals the distribution coefficient value observed for certain nuclides is independent of the method used. For other heterogeneous mineral phases, dependencies on methods used still exist; however, some of the problems have been identified as experimental artifacts rather than true differences. Work will continue in developing guidelines for standard procedures and evaluating the applicability of currently used methods.

Progress in collecting a sorption-desorption data base and evaluating methodology included the incorporation of "controlled samples." These samples include Sentinel Gap basalt, Westerly granite, Oolitic limestone, Climax Stock "granite," Conasauga shale, argillaceous shale, and, from the WIPP site, dolomite and anhydrite. Formulas for synthetic groundwaters were distributed so that all labs can perform distribution coefficient experiments using the same system. Such a round-robin experiment will allow a check on laboratory biases and facilitate experimental comparisons.

A document, Solid Phases and Solution Species of Different Elements in Geologic Environments, has been drafted which includes mineral stability and solution species activity diagrams and cites the thermodynamic data used. Its

purpose is to project the long-term fates of radionuclides for comparison with short-term laboratory experiments. The thermodynamic data are also being incorporated in a computer program which predicts mineral formation and solution activities. The computer program will be used to analyze numerous hypothetical scenarios in order to predict the types of reactions and thermodynamically stable mineral and solution assemblages expected in deep repositories. The computer model will rapidly and efficiently set up potential boundary conditions for formulating laboratory sorption experiments and will systematically illustrate the more significant of the diverse geochemical reactions that could occur in a deep repository.

Preliminary results of plutonium dissolution studies of PuO_2 and $\text{Pu}(\text{OH})_4$ show what appears to be $\text{Pu}(\text{V})$ as the dominant solution species under atmospheric conditions.

Autoradiography techniques are being formulated to attempt to identify which rock minerals or grains are the active adsorbers. Autoradiography will be coupled with scanning electron microprobe work to identify

sorption sites on the individual grain scale.

TASK 5 - WISAP SOCIETAL ACCEPTANCE

- A. H. Schilling (HARC)

Two reports being prepared by other contractors were reviewed. These reports are Resolving Community Conflict in the Nuclear Power Issue: A Report and Annotated Bibliography, and Public Consultation in Public Policy Formation: A State-of-the-Art Report.

Work was completed on a repository impact projection model, and a report entitled A Demographic Model for Assessing the Socioeconomic Impacts of Large-Scale Industrial Development Projects, was completed.

Revisions were prepared for the following reports: 1) The Management of Social and Economic Impacts Anticipated with a Nuclear Waste Repository: A Preliminary Discussion, 2) Compensation as One Response to the Unintended Effects of a Government Program: The Case of Nuclear Waste Management Facilities, 3) State and Local Regulation Relevant to Waste Isolation Facilities, and 4) Land Use Control Considerations in Selected States.

10.0 WELL LOGGING INSTRUMENTATION DEVELOPMENT FOR SHALLOW LAND BURIAL

R. L. Brodzinski - Project Manager

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

SUMMARY

This is a new program as of December 1977; consequently, this first quarter's efforts have dealt largely with program planning and initiation. Calibration of a passive neutron detection technique for quantitative analysis of plutonium oxides and fluorides in dry soil has been completed. An active neutron monitoring system has been designed, assembled, and tested for determination of transuranic concentrations in relatively low-gamma-radiation environments.

DEVELOPMENT OF LOGGING SYSTEMS CAPABLE OF MEASURING ^{90}Sr -

R. L. Brodzinski and H. L. Nielson

Procedures for obtaining a suitable source of ^{90}Sr for testing purposes have been initiated. An evaluation of the necessary electronic instrumentation, Bremsstrahlung radiation detection equipment, and test facilities indicates that suitable systems are available in-house and that no additional equipment purchases will be required for this phase of the

study. Arrangements have also been made to utilize existing facilities for the studies of thermal and fast neutron activation analysis of ^{90}Sr .

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

Efforts to date have been directed toward evaluating and optimizing the technique of scintillation spectrometry for the analysis of tritium from wells in a burial ground.

DEVELOPMENT OF LOGGING SYSTEMS CAPABLE OF MEASURING ACTINIDES -

R. L. Brodzinski and N. A. Wogman

The concept of a passive neutron detection system for the determination of plutonium in areas of high-level gamma radiation was advanced during earlier work for Atlantic Richfield Hanford Company (now Rockwell Hanford Operations). This concept forms the basis for the portion of this research program devoted to developing and calibrating methodology for the determination of transuranics by neutron detection. Even though

the quantitative determination of transuranics by neutron well logging will necessarily be an active system, it must be cross-calibrated with a passive technique in order to accurately correct for the effects of gamma radiation.

The methodology for passive neutron determination of transuranic isotope concentrations is described in detail in an open literature publication entitled "In Situ Quantitative Determination of Transuranic Elements in Areas of High-Level Gamma Radiation," by R. L. Brodzinski and N. A. Wogman and will not be repeated here. This paper will be published in the proceedings of the American Nuclear Society Meeting on Analytical Methods for Safeguard and Accountability Measurement of Special Nuclear Material to be held May 15-17, 1978 in Williamsburg, Virginia. The paper deals primarily with calibrations of the technique in dry soil.

Additional effort will be spent in obtaining calibrations at other moisture levels in order to accurately cross-calibrate the active

neutron well logging monitor. This technology is currently being augmented by a Rockwell Hanford-sponsored program aimed at calibration of the passive system in field situations which have been sampled for laboratory analysis. The passive technique is also expected to be utilized in the near future for in situ determination of plutonium concentrations in several unknown field situations. These field applications are also anticipated to be performed under the auspices of Rockwell Hanford Operations.

A prototype active logging system for the determination of transuranics in a burial ground was constructed and cross-calibrated with the passive technique for plutonium in dry soil. Initial tests indicate that this logging device will be capable of measuring transuranic concentrations of much less than 10 nCi/g from either cased or caseless wells as small as 2 in. in diameter. Future tests planned for this system include calibrations as a function of soil moisture content and gamma radiation intensity.

11.0 MONITORING AND PHYSICAL CHARACTERIZATION OF UNSATURATED ZONE TRANSPORT

S. J. Phillips - Project Manager

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

SUMMARY

Analyses of parameters controlling transport of radionuclides through porous media in low-level waste burial sites are continuing. Effects such as thermodynamic fluid potentials and nonisothermal enhanced fluid migration and media heterogeneity are being analyzed independently in the laboratory. In addition, comprehensive field monitoring studies are being used to study the simultaneous effects of migration controlling parameters. Use of specific devices and transducer systems is being demonstrated in these field monitoring activities.*

LABORATORY ANALYSIS - R. J. Serne, A. C. Campbell, and S. J. Phillips

Laboratory determination of fluid and radionuclide transport parameters in porous media under partially saturated and saturated conditions is currently being performed. To aid in the understanding and quantifying

of the complex factors controlling transport of radioactive waste under field conditions, preliminary laboratory analysis are being made.

Analyses of infiltration and drainage fluid into large one-dimensional sediment columns have been conducted in isotropic, homogeneous media under isothermal conditions. These studies permit evaluation of spatial and temporal changes of fluid wetting front, fluid retention, and hydraulic conductivity-diffusivity. For this work, transient and steady-state cases have been run. Furthermore, tensiometric scanning of these sediment columns was performed to monitor the potential status of fluid during transient cases under partially saturated cases. These analyses assist in simulating meteoric fluid flux through burial grounds. The information derived from this phase of research will be used in conjunction with partially saturated radionuclide transport one-dimensional column studies.

Another important activity involves determining the effect of liquid saturation on radionuclide transport. In evaluating this factor, column studies have been started. Prior to column

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*For relatively detailed information on the background of this program and its tasks, see the prior issue of this progress report, PNL-2377-4.

experiments using partially saturated water conditions, saturated columns of sediment B were used to validate the predicted movement of technetium, strontium, and cesium through the selected sediment. Simulated saturation extract solutions and a similar solution using chloride salts of the major cations were percolated through columns to provide information on these radionuclides and macrocation interactions with the sediment. Operational parameters for the columns are given in Table 11.1.

Technetium was shown to move through the column with no measurable retardation. Both columns exhibited a decrease in effluent concentrations of technetium before it was removed

from the influent. Dispersion and anion exclusion are probable causes. Initial effluent concentrations greater than those in the influent (Column B3) may also be due to anion exclusion. Figure 11.1 shows the normalized movement of ^{95}mTc through the columns of sediment B.

Breakthrough curves for strontium show significant retardation of the radionuclide through sediment B. It was predicted that strontium would move about 250 times slower than water. The maximum effluent activity for the column exposed to a continuous source was recorded at 250 pore volumes, after which the activity level decreased and leveled off. From theoretical calculations assuming ideal

TABLE 11.1. Parameters for Saturated Sediment B Columns

	Column			
	B1	B2	B3	B4
<u>Common Parameters</u>				
Column diameter	←———— 5.4 cm ID —————→			
Particle density	←———— 2.851 ± 0.005, g/cm ³ —————→			
Flow rate	←———— 11.6 cm ³ /hr or 0.51 cm/hr —————→			
<u>Varying Parameters</u>				
Column length, cm	5.8	5.7	5.9	5.8
Soil weight, g	245.6	245.4	245.8	245.6
Soil column volume, cm ³	132.83	130.54	135.12	132.83
Bulk density, g/cm ³	1.85	1.88	1.81	1.85
Porosity	0.35	0.34	0.36	0.35
Pore volume, cm ³	46.62	44.51	49.32	46.52
Influent ^{95m} Tc, cpm/ml	--	54	54	--
Volume influent ^{95m} Tc, ml	--	250	100	--
Influent ⁹⁵ Sr, cpm/ml	--	103-198	109	--
Volume influent ⁸⁵ Sr, ml	--	~12,600	~1600	--
Influent ¹³⁷ Cs, cpm/ml	--	32-125	58	--
Volume influent ¹³⁷ Cs, ml	--	~7700	~1600	--
Influent solution	← Saturation extract →			Chloride Salts of saturation extract

FIGURE 11.1. Movement of ^{95m}Tc Through Columns B2 and B3

ion exchange, the breakthrough curve should be "S" shaped and pass through $C/C_0 = 0.5$ at 250 pore volumes. Obviously, the strontium breakthrough curve is not ideal because some reaction (possibly carbonate precipitation) is occurring to further retard the migration of strontium. Injection of a limited quantity pulse resulted in very little breakthrough. Again, the effluent does not follow ideal ion exchange, suggesting that additional sorption is occurring. Figures 11.2 and 11.3 show the normalized movement of strontium sediment B.

Cesium was not observed in the column effluents because of strong sorption. A breakthrough of 0.5 was not expected until 3000 pore volumes. The columns will be dissected to determine the location of cesium in the columns. From the column profile, sorption data can be calculated and compared with the batch distribution coefficient data.

Macroocation interactions in the columns differed according to the solution used. Both solutions removed the readily soluble salts upon initial contact with the columns. Preliminary observations indicate that soluble salts are not completely removed in the first aliquot of leachate. Figures 11.4 and 11.5 are examples of the observed effluent concentrations for the major cations and anions present in saturation extract solution percolated through column B1.

For column B1 the dissolution of soluble salts kept the effluent high in magnesium for ~ 0.3 pore volumes; calcium for ~ 0.8 pore volumes; sodium for ~ 0.8 pore volumes; potassium for ~ 0.5 pore volumes; chlorine about 3 pore volumes; SO_4 about 4 pore volumes; and HCO_3 about 3 pore volumes. For the first 20 pore volumes an excess over mass present in the percolating influent shows 1 mg K^+ , 5.6 mg Na^+ , 13.5 mg Ca^{+2} , 2.8 mg SO_4^{-2} , and

FIGURE 11.2. Movement of ^{85}Sr Through Column B2, Continuous Spike

FIGURE 11.3. Movement of ^{85}Sr Through Column B3, ~1600 ml Spiked Influent

FIGURE 11.4. Mg^{2+} Concentration in Column B1 Effluent

FIGURE 11.5. Cl^- Concentration in Column B1 Effluent

1.0 mg Cl^- were released from the soil. On the other hand, the Mg^{+2} in the effluent is 2.8 mg less than the influent percolating through the column; HCO_3^- effluent mass was also 8.6 mg lower. For charge balance, an excess of K^+ (0.026 meq), Na^+ (0.243 meq), Ca^{+2} (0.675 meq) is obtained along with a shortage of Mg^{+2} (-0.228 meq) for a net increase in 0.716 meq of cations. For the anions, an excess of SO_4^{-2} (0.058 meq) and Cl^- (0.028 meq) and shortage of HCO_3^- (-0.140 meq) are observed, yielding a net shortage of 0.054 meq of anions. Ideally, the net change in cations and anions should be equal, but with complexation such as CaHCO_3^- , NaSO_4^- , etc., it is difficult to ascertain whether a mass and charge balance discrepancy is present. The excess cations leached from the soil are considerably larger than the amount of soluble salts available in the column (0.13 meq). It appears that

dissolution of calcium carbonate and calcium sulfate in the soil might have been occurring over the 80 hr that the column was run. The decrease in magnesium probably reflects magnesium sorbing onto exchange sites with the consequent release of other cations such as sodium, potassium, and calcium.

The effluent curves for Column B4, percolated with chloride salts, show there was an excess of all the cations found in the 20-pore volumes collected: Mg^{+2} (2.1 mg or 0.17 meq), Ca^{+2} (5.2 mg or 0.26 meq), Na^+ (5.5 mg or 0.24 meq), and K^+ (1.9 mg or 0.049 meq). Only the chloride anion was monitored and was found to lose mass to the soil (3.35 mg or 0.09 meq). A possible explanation is that slightly soluble salts such as calcite (CaCO_3) and gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) are dissolving and the resultant calcium partially replaces ion exchanged K^+ , Na^+ and Mg^{+2} . The readily soluble

salts of Na^+ , Mg^{+2} and K^+ also add to the excess cations observed. The apparent loss of chlorine is unexplainable but may reflect experimental error (the shortage of chlorine is only 4% of the total chlorine percolated through the system).

Although the macrochemistry appears to be more complex than anticipated, once the high concentrations of salts caused by soluble salt dissolution decays (typically within the first pore volume), the small perturbations observed would not greatly affect the radionuclide transport. In the future a few more columns will be run to gain better understanding of the macrochemistry, but immediate activity will switch to tracing radionuclide movement of technetium, iodine, and selenium under unsaturated conditions.

PHYSICAL MODELING OF CONTAMINANT TRANSPORT - L. D. Kannberg and S. J. Phillips

Accelerated testing of the unsaturated flow domain is necessary to determine optimal monitoring systems design and to predict contaminant transport rates. Modeling of one- or two-dimensional flux of contaminants through soils and other geologic materials will permit quantitative rate predictions of radionuclides and fluids transported away from burial facilities. Physical modeling of waste transport systems under controlled laboratory conditions provides effective simulation and prediction capabilities used to assess long-term transport phenomena.

Sets of data for six pore size distributions have been collected from generic sediments. The data were collected at elevated and ambient thermal conditions in order to evaluate nonisothermal fluid retention effects, i.e., viscosity and surface tension. Equilibration time studies were also conducted in order to evaluate effects of hysteresis. Physical three-dimensional columns have been constructed for use in simulating steady-state and transient flow throughout dimensional systems, and a gamma attenuation monitoring apparatus has been developed for determining saturation-potential relationships. This system is illustrated in Figure 11.6.

FIELD MONITORING AND DATA EVALUATION - G. W. Gee and M. D. Campbell

Real-time monitoring of active and/or retired burial facilities is needed to detect and dimensionally evaluate the movement of radiocontaminants. Development and installation of monitoring instruments to evaluate potential and actual contaminant transport in situ will assist in determining the magnitude and hazards. Field collection and data processing of instrument array data is the best method of assessing actual radionuclide transport.

Monitoring transducer systems which measure soil physics and micro-meteorological phenomena are being installed at a test site. These systems have been calibrated and laboratory tested for sensitivity and accuracy.

FIGURE 11.6. Schematic Diagram of Model Monitoring and Analysis System

Capillary Pressure Sensor

A specific transducer system has been developed which uniquely measures a wide range of water potential (capillary pressure) in partially saturated porous media. This system was designed to measure the capillary pressure in sandy sediments over the range from saturation to air-dry water contents (0 to -1000 bars capillary pressure).

This system consists of a heat dissipation probe and associated electronic circuitry. The thermal response of the probe (consisting of a heater, thermal sensor, and porous matrix) is calibrated in terms of the capillary pressure of the matrix. The porous matrix through which the heat is dissipated is soil material confined in a fixed geometry by a wire screen. Calibration is accomplished by draining or drying the sensor to a

fixed dryness (related to the capillary pressure of the porous matrix) and measuring the EMF output of the sensor in response to a timed heat pulse. Figure 11.7 shows the response of three heat dissipation units in ΔEMF (millivolts) as a function of matrix (or capillary) pressure and illustrates the sensitivity of the heat probe in response to different matrix materials. Curve 1 illustrates the sensor response when castone (a dental cement) was used as the dissipation matrix. Curves 2 and 3 illustrate sensor response to a 1:6 castone:soil mix and to soil alone, respectively.

The soil matrix (curve 3) is clearly the most suitable because of the greater sensitivity (greater millivolt output- ΔEMF per unit change in matrix potential (capillary pressure)). The data from curve 3 suggest that a two-point calibration may be possible

FIGURE 11.7. Heat Dissipation Device Performance of Capillary Pressure Versus Δ EMF Output

for soil matrix-type probes. This could greatly speed calibration. Soils or sediments from the test sites may be used in the probes, provided that suitable calibrations are made before field use. These heat

dissipation sensors are insensitive to ambient thermal fluctuations and salt migration. Hence they should prove to be ideal for long-term water migration studies at burial sites under virtually any climatic regime.

12.0 DETECTION AND CHARACTERIZATION OF MOBILE ORGANIC COMPLEXES OF FISSION PRODUCTS

J. A. Franz - Project Manager

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

SUMMARY

A paper, Mobility of Organic Complexes of Some Non-Tru Fission and Activation Products: A Selective Review, has been prepared for public distribution. Two-level factorial experiments were performed which indicate that K_d (soil absorption) of zirconium-tartrate varies linearly with zirconium concentration. Zirconium-gluconate absorption is negatively affected by ligand concentration, with a negative interaction of zirconium concentration. More dilute experiments indicate the same general trends. Carbon-13 NMR* and gel permeation chromatography studies indicate that zirconium-gluconate is a polymer and that zirconium bonds with a carbonyl and an alkyl group at only one end of the tartaric acid molecule. A tetrameric zirconium configuration surrounded by ligand molecules was proposed. A paper summarizing these results was started.

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*Nuclear magnetic resonance spectroscopy

IDENTIFICATION OF NUCLIDES AND COM- PLEXING AGENTS - D. J. Wiggins and J. A. Franz

The PNL report Mobility of Organic Complexes of Some Non-TRU Fission and Activation Products: A Selective Review reviews the literature of the aqueous and soil chemistry of organic complexes of zirconium, niobium, technetium, and nickel.⁽¹⁾

STUDY OF MOBILITY CHARACTERISTICS OF ORGANIC COMPLEXES - D. J. Wiggins and J. A. Franz

Two-level factorial experiments were performed during this quarter to determine the effects of pH and complex concentration on zirconium-tartrate absorption by soil and their interrelation. It was determined that pH was not statistically significant and that K_d varies linearly with complex concentration as depicted in Figure 12.1.

Initial screening experiments were performed to determine the important variables of zirconium-gluconate absorption by soil. Ligand concentration and complex concentration

FIGURE 12.1. Effect of Ligand:Zr Ratio and Quantity of Complex on Absorption of Zr-gluconate by Soil

were determined as the most statistically significant variables of the six variables studied. Then two-level factorial experiments were performed to determine the effects of the ligand:zirconium ratio and the quantity of complex on the absorption of zirconium-gluconate by soil. The following conclusions were made:

- Ligand concentration has a negative effect on soil absorption (K_d).
- Ligand:zirconium ratio and quantity of complex interact, negatively affecting soil absorption (K_d).

- Quantity of complex does not independently affect soil absorption (K_d).
- The soil absorption values (K_d) are linearly related.

These results are summarized in Figure 12.2.

The experimental results of soil absorption of more dilute zirconium-gluconate and zirconium-tartrate complexes indicate the same general trend as found in the previous experiments that used higher concentrations: K_d increases as zirconium-tartrate complex concentration decreases, and K_d remains constant as zirconium-gluconate concentration decreases.

Several experiments on the absorption of uncomplexed zirconium were performed. Results ($K_d \approx 489$) agree with the results of other investigators.

Carbon-13 NMR studies were performed to determine the structure of the involved complexes. Carbon-13 NMR results indicate that high-molecular-weight polymer formation is occurring, especially for zirconium-gluconate (1:1), where a solid polymer was formed at very high concentrations of zirconium and gluconic acid (1:4). Polymer formation is also indicated by the long data acquisition times necessary for obtaining spectra. This may be due to dipolar broadening caused by the size and shape of the molecule restricting the isotropic molecular motion so that we are seeing only a small concentration of uncomplexed unpolymerized ligand in the zirconium-gluconate system. Although the zirconium-gluconate NMR spectrum (Figure 12.3) does not

FIGURE 12.2. Zirconium-tartrate Absorption by Hanford Soil

FIGURE 12.3. ^{13}C NMR of 0.5M Zirconium-gluconate at pH 5.4 Where Only Uncomplexed Gluconic Acid is Detected

indicate the presence of uncomplexed zirconium-gluconate, the 1:1 zirconium-tartrate spectrum indicates the presence of about 50% uncomplexed carbonyl and alkyl groups (Figure 12.4). It appears that all of the tartaric acid does complex with the zirconium uniformly at one end of the molecule:

Gel permeation chromatography was performed to determine the size and/or molecular weight of the involved complexes. The high-pressure gel permeation chromatography (HPGPC) indicates that zirconium-gluconate and zirconium-tartrate each forms one polymer having a molecular weight of 9000 to 10,000, using polyethylene glycol as a standard. Since an additional molecule is introduced into the system, dodecyl sulfate as a surfactant, additional experiments using a different surfactant will be performed next quarter.

The HPGPC and NMR results indicate the formation of the following type of polymer, where each zirconium molecule is attached to a tartaric acid molecule and the zirconiums form the inner core of the polymer:

This tetrameric zirconium structure is particularly plausible considering the proposed tetrameric structure of uncomplexed zirconium.⁽³⁾ As in the uncomplexed zirconium solution, when pH is raised, polymerization may occur by olation, thereby increasing the size of the inner zirconium core.

A paper summarizing these results was started during this quarter and will be submitted to the Journal of Environmental Science and Technology.

An experiment was performed to determine the resolubilization of oven-dried hydrous zirconium by tartaric acid. A precipitate was obtained having an entirely different character from that of hydrous zirconium. Elemental analyses indicate that:

FIGURE 12.4. ^{13}C NMR of 0.5M Zirconium-Tartrate (1:1) at pH 7.0

- 12.65% of the zirconium is resolubilized by tartaric acid
- 1.18% of the tartaric acid forms an insoluble complex with the zirconium.

Therefore it appears that, even though zirconium may precipitate onto the soil, it can be resolubilized by a concentrated pocket of decomposing

organic matter which could contain compounds having a structure similar to that of tartaric acid.

A procedure for preparing technetium complexes of the stability of organic complexes of technetium⁽⁴⁾ has been chosen, and $^{99\text{m}}\text{Tc}$ has been ordered for studies to be performed during the next quarter.

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13.0 ELECTROPOLISHING FOR SURFACE DECONTAMINATION OF METALS

R. P. Allen and H. W. Arrowsmith - Project Managers

The objective of this program is to develop electropolishing as a large-scale decontamination technique for radioactive metal surfaces. Major program activities include 1) the establishment and operation of a demonstration decontamination facility and 2) laboratory studies to understand and optimize the decontamination process.

SUMMARY

The electropolishing facility has been prepared for a test campaign to demonstrate the decontamination of 46 m^2 (500 ft^2) of plutonium-contaminated metallic waste. The prepolished stainless steel surfaces were readily cleaned using dilute phosphoric acid and swab techniques. The tanks were decontaminated by electropolishing using reverse polarity.

A large contact in situ device has been developed and successfully used to electropolish over 2700 cm^2 (3 ft^2) of stainless steel sheet. This device uses high-current densities but normal electropolishing voltages and provides complete containment of the electrolyte.

The in situ decontamination of the inside of a 6-m-long (20 ft) section of piping using a movable 0.6-m (2 ft) cathode was demonstrated on the corrosion test loop at the Hanford N-Reactor. Radiation levels in the less contaminated portions of the pipe were reduced about 4 R/hr, and more than 40 R/hr in the more heavily contaminated portions.

In other studies, the versatility of electropolishing was demonstrated by the open pit electropolishing of the bridge crane to be used in the pretreatment/sectioning facility. Also, vibratory finishing techniques using ceramic media were shown to remove a significant fraction of the recast layer on sectioned material.

Preparations for the April 18-19 public information meeting have included mailing over 600 circulars, writing a report on progress and applications of electropolishing as a decontamination technique, and updating a videotape describing the electropolishing program.

DECONTAMINATION STUDIES

The electropolishing facility has been completely decontaminated and fresh electrolyte and rinse solutions added to prepare for a test campaign to demonstrate the decontamination of 46 m^2 (500 ft^2) of mechanically sectioned, plutonium-contaminated metallic waste. The electropolishing and rinse tanks were decontaminated from over $1 \text{ M dis/min} \cdot 100 \text{ cm}^2$ to near

background by electropolishing using the contaminated acid and reverse polarity such that the tank walls were the anode. The electrolyte was then removed and the tanks rinsed and refilled. The prepolished stainless steel surfaces on the tank exteriors and adjacent room walls were readily cleaned to background by conventional swab cleaning techniques using dilute phosphoric acid.

Prior to cleanup of the electropolishing facility for further decontamination studies, it was used to decontaminate product-receiving canisters and lids. The lids had alpha contamination levels varying from 5000 to 60,000 dis/min·100 cm². After the initial run, one lid was decontaminated to background and two others showed only low-level contamination. These two lids were disassembled and further electropolished in a titanium basket, where all pieces but one were reduced to background. A pasty residue sealed in a crack prevented total decontamination of the remaining piece. The external surfaces of the product-receiving canisters were also successfully decontaminated. However, some contamination remained on the internal surfaces, again due to a nonconductive residue inside a crack. This illustrates the need for adequate pretreatment methods and also for proper design of components to facilitate decontamination by electropolishing.

IN SITU DECONTAMINATION TECHNIQUES

Development is continuing on in situ electropolishing techniques for

use in decontaminating surfaces that either cannot be transported to, or immersed in, a conventional electropolishing cell. A large contact in situ device (Figures 13.1 and 13.2) was developed which can rapidly electropolish defined surface areas using high-current densities but normal electropolishing voltages. This device was used successfully to electropolish over 2700 cm² (3 ft²) of stainless steel sheet (Figure 13.3), while providing complete containment of the electrolyte. Possible applications of this device include decontaminating storage tank walls, fuel pool walls, hot cell surfaces, and any other large, near-planar contaminated surface.

Special electropolishing techniques have been developed for the in situ decontamination of the inside of long pipes. This important application of in situ electropolishing techniques was demonstrated on the corrosion test loop at the Hanford N-Reactor in a test sponsored by United Nuclear Industries, Inc.

The corrosion test loop, which had been in contact with the primary coolant for about 15 yr, is a 6-m-long (20 ft) section of 6.9-cm-ID steel pipe. The inside of this pipe was electropolished 0.6 m (2 ft) at a time, using a movable cathode (Figure 13.4) that consisted of a 0.6-m-long, 4.8-cm-OD (2 ft long, 1-7/8 in. OD), stainless steel pipe with nylon insulators at each end. The electrolyte was pumped through the cathode into the pipe, which was isolated from the primary coolant system by valves, and returned to the external electrolyte reservoir through a drain

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FIGURE 13.1. Schematic of Large-Contact, In Situ Technique Capable of Electropolishing Defined Surface Areas While Containing the Electrolyte

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FIGURE 13.2. Large-Contact, In Situ Device Built According to Principle Shown in Figure 13.1 and Capable of Electropolishing 2700 cm² at a Time

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FIGURE 13.3. Electropolished Area Produced Using the Contact Device Shown in Figure 13.2

FIGURE 13.4. Schematic of the N-Reactor Corrosion Test Loop with Electropolishing Equipment Installed

line at each end of the loop. The electrolyte was supplied to the cathode through PVC pipe 6 m long (20 ft) that also served to move the cathode and, in addition, contained the electrical cable to the cathode. A standard reactor process tube end cap was modified to allow free passage of the plastic pipe while ensuring containment of the electrolyte.

Each 0.6-m (2 ft) section of pipe was electropolished for 20 min at a current of about 1076 A/m^2 (100 A/ft^2). The hydrogen generated by the electropolishing process was flushed out of the pipe by the circulating (15 l/min) electrolyte and was vented from the external electrolyte reservoir. The measured hydrogen level in the working area was always well below a

hazardous concentration. At the conclusion of the electropolishing runs, the electrolyte was removed and the pipe rinsed by pumping water through it. The limited test time did not permit a more adequate internal spray rinse.

The radiation levels inside the pipe before and after electropolishing were measured at 30.5-cm (1 ft) intervals using a Geiger Muller tube. The electropolishing treatment reduced the average radiation levels in the less contaminated portion of the pipe by about 4 R/hr. The readings near pipe fittings, where pockets of contamination had collected, were reduced by more than 40 R/hr. Electrolyte containing corrosion inhibitors was used to minimize corrosion of the

pipe by the phosphoric acid. Ultrasonic wall thickness measurements and pressure tests at the conclusion of the decontamination study verified the integrity of the pipe and permitted its return to service.

PREPOLISHING APPLICATIONS

The stainless steel walls and all the equipment for the walk-in hood for the pretreatment facility have been prepolished to facilitate the maintenance of a low-background contamination level. This included the prepolishing of two 5.8-m-long (19 ft) bridge rails and the 3.0-m-long (10 ft), 182-kg (400 lb) traversing beam for the bridge crane. Innovative techniques were needed to electropolish components of this size. The rails were prepolished using an

improvised electropolishing tank cut from 20.3-cm-dia (8 in.) acid-resistant PVC pipe. The traversing beam for the bridge crane was prepolished in an open pit, 4.3 by 1.2 by 0.9 m (14 by 4 by 3 ft) (Figure 13.5), lined with sand, concrete curing blankets, plywood, and multiple layers of plastic and then filled with phosphoric acid electrolyte. The ability to successfully electropolish this large beam using this rudimentary electropolishing system illustrates the inherent versatility and adaptability of electropolishing as a technique for prepolishing and decontaminating surfaces.

PRETREATMENT STUDIES

Construction work on the pretreatment facility (Figure 13.6) is almost complete. The prepolished stainless

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FIGURE 13.5. Open-Pit Electropolishing of Traversing Beam for Bridge Crane

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FIGURE 13.6. Cut-Away View of Stainless Steel Walk-In Hood Located in Sectioning/Pretreatment Facility

steel floors, walls, and airlock of the walk-in hood have been installed, and the external closure welds have been completed. In addition, the special doors with inflatable gaskets and both manipulators have been installed.

Testing continues on the 113-2 (4 ft³) vibratory finisher obtained for use in the pretreatment facility. A combination of low-wear-rate ceramic media and caustic cleaning compound was found to remove paint and grease adequately with minimum metal removal while generating significantly less secondary waste than higher-wear-rate plastic and ceramic media. Studies using high-cutting-rate ceramic media did demonstrate, however, the ability of vibratory finishing to remove a significant fraction of the potentially contaminated recast layer on plasma-arc-sectioned stainless steel.

Visits with vendors of vibratory finishing equipment have revealed

that the vibratory finisher may have a new application. They routinely use vibratory finishing to remove flashings from die-molded plastic and rubber parts. Softer parts are processed at sub-zero temperatures. This would indicate that similar processes could be used to decontaminate selected nonmetallic components such as Plexiglas® panels from glove boxes.

PUBLIC INFORMATION MEETING

Preparations are continuing for the public information meeting and tour to be held April 18-19. Over 600 circulars have been mailed. These describe the meetings and contain general information on electro-polishing as a decontamination technique. A report discussing the progress and applications of electro-polishing as a decontamination technique and a videotape are also being prepared for use in the meeting.

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