Nuclear Waste Management Quarterly Progress Report July Through September 1979

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

November 1979

**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
JULY THROUGH SEPTEMBER 1979

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

November 1979

Prepared for
the U.S. Department of Energy
under Contract EY-76-C-06-1830

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 Nuclear Waste Management. Only programs in the scope of this document that have significant, reportable results are included. The Commercial High-Level Waste Immobilization Program is reported elsewhere.

These reports are issued on a quarterly basis. The prior one in this series is Nuclear Waste Management Quarterly Progress Report April Through June 1979, PNL-3000-2.

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1. DECONTAMINATION AND DENSIFICATION OF CHOP-LEACH CLADDING RESIDUES

T. M. Lambright - Project Manager

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 develop methods for removal of long half-life transuranics from fuel cladding residues and to consolidate the decontaminated cladding into a dense, chemically unreactive form for efficient storage or burial. The study includes:

- 1. demonstration of a prototype system for combined descaling and densification of unirradiated scrap fuel cladding*
- 2. development of engineering data to provide the basis for scaleup to a production system*
- 3. evaluation of ground-water corrosion tests conducted on samples of alloyed ingots previously produced by the densification process.*

SUMMARY

A prototype system for decontaminating and densifying chop-leach cladding residues was assembled and evaluated by the Pacific Northwest Laboratory. Pilot-scale chemical decontamination equipment was designed to use an HF/Ar gas mixture to remove simulated contamination from scrap cladding pieces. Treated scrap could then be melted in PNL's Inductoslag ingot melting furnace to produce ingots.

As soon as the prototype chemical decontamination equipment became fully operational, the combined decontamination densification system was to be demonstrated. However, tests of the decontamination equipment identified several equipment and process problems that were not completely resolved before the end of the research project. Nevertheless, actual combined operation of the furnace and decontamination equipment was achieved on a small scale with the production of an 8-lb ingot. The decon-

tamination equipment has been dismantled, and the major components have been stored for possible future use.

Successful tests of the Inductoslag furnace showed that it could produce smooth-sided ingots from various alloys of Zircaloy, stainless steel, and Inconel.⁽¹⁻³⁾ The furnace and associated equipment will be left in place for future research efforts.

A two-volume report entitled The Decontamination and Densification Process is being prepared. The report will provide full detail on the operation of both systems.

DECONTAMINATION PROCESS

The Pacific Northwest Laboratory's hull decontamination equipment consists of an HF reactor and washer/rinser unit, HF and Ar gas supply systems, chemical storage and transfer equipment, and control instrumentation, as described in previous reports.^(1,2,4) The prototype equipment, designed to descale fuel cladding hulls, was

dismantled because development work was scheduled to terminate at the end of FY-79. Volume 1 of the final report, entitled The Prototype Cladding Hull Decontamination System, will describe the process and equipment development. A separate report on the development of the decontamination process will also be issued.

Seven tests of the decontamination equipment were previously made to define equipment operating parameters. Autoclaved, unirradiated cladding hull material was used in these tests.⁽⁵⁾ A key problem during the tests was the generation of excessively high temperatures in the HF/Ar gas reactor. The presence of hydrogen gas, generated by the reaction of HF gas with the Zircaloy-4 cladding pieces, allowed zirconium hydrides to form. The zirconium hydrides react with HF gas to release a great amount of heat. Heat released during the last test melted the teflon bellows at the reactor inlet seal. A stainless steel bellows was installed to replace the damaged teflon bellows.

Two modifications were made to alleviate the high temperatures. The HF and Ar gases were mixed more thoroughly at a point further away from the reactor; this ensured that the Zircaloy hulls would not be reacting with essentially undiluted HF gas. In addition, the gas removal tube was extended to allow gas removal near the mouth of the reactor. The extension prevented hydrogen and HF gases from passing through the bed of hulls in the feed chute, minimizing the exothermic reaction of hydrogen fluoride with zirconium hydrides.

An eighth descaling test was completed this reporting period before the equipment was dismantled. About 10 min after HF gas was admitted to the reactor chamber, reactor pressure exceeded the control limit of 10 in. of water. This situation caused

automatic shutoff of HF gas flow and reactor heat. At the same time, two of five reactor zones exceeded the 650°C control temperature. Six minutes later, reactor temperature returned to normal, but reactor pressure could not be lowered even when the argon flow rate was widely varied. Since reactor pressure could not be maintained within limits, total process shutdown was begun.

Later inspection of the pressure control system revealed only one probable source of high reactor pressure. The off-gas filter contained stainless steel wool that appeared to be heavily loaded with zirconium fluoride powder. This possibly could restrict gas flow enough to prevent maintenance of proper reactor pressure. No tubing or valves in the exit gas stream were restricted.

Since this project has been terminated, no additional work will be devoted to decontamination equipment design or development.

DENSIFICATION PROCESS

The Inductoslag ingot melting furnace is the densification portion of the decontamination-densification equipment that was set up to process chop-leach cladding residues. The Inductoslag furnace and the chemical decontamination equipment were set up together on a simulated hot cell floor plan. Although the decontamination equipment has been dismantled and removed, the Inductoslag furnace and associated equipment will be left in place for future research efforts.

An 8-lb ingot was made in the Inductoslag furnace using descaled cladding hull simulations from the final two tests of the decontamination equipment. A significant amount of spitting occurred while the cladding pieces were fed into the furnace's molten metal pool. This was attributed to the release of hydrogen gas that was

absorbed by the cladding pieces during the decontamination process. The ingot is being analyzed for chemistry, hardness, and microstructure.

Volume 2 of the final report Densification by Inductoslag Melting will describe operation of the furnace and its development for melting cladding hulls.

REFERENCES

1. A. M. Platt and J. A. Powell, comps., Nuclear Waste Management Quarterly Progress Report January through March 1978. PNL-2378-1, Pacific Northwest Laboratory, Richland, WA, August 1978.
2. A. M. Platt and J. A. Powell, comps., Nuclear Waste Management Quarterly Progress Report April through June 1978. PNL-2378-2, Pacific Northwest Laboratory, Richland, WA, November 1978.
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December 1978. PNL-2378-4, Pacific Northwest Laboratory, Richland, WA, May 1979.
4. A. M. Platt and J. A. Powell, comps., Nuclear Waste Management Quarterly Progress Report July through September 1978. PNL-2378-3, Pacific Northwest Laboratory, Richland, WA, January 1979.
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2. MONITORING METHODS FOR PARTICULATE AND GASEOUS EFFLUENTS FROM WASTE SOLIDIFICATION PROCESSES

R. W. Goles - Project 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 all analytical off-gas sample work was completed and a draft report characterizing Nuclear Waste Vittrification Project (NWVP) off-gas emissions was written and submitted for approval.

MONITORING RESULTS

The purpose of the program, "Monitoring Methods for Particulate and Gaseous Effluents from Waste Solidification Processes," was to characterize concentrations of vitrification-originated effluent escaping the hot cell through the process off-gas exhaust. First a paper study was conducted to identify undesirable effluent species likely to escape all existing in-cell clean-up devices. Table 2.1 summarizes the findings of this study. After determination of the species to be sampled for, a sampling site was established and a monitoring network developed.

Samples were taken during the second vitrification campaign associated with the NWVP. Subsequent laboratory sample analysis established off-gas emission levels for the efflu-

TABLE 2.1. Important Process Effluent

Gases

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

Semivolatiles

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

Particulates

Fission products, including $^{144}\text{CePr}$
 Activation products
 Heavy elements

ents listed in Table 2.1. Tables 2.2 through 2.7 summarize these results. A complete program summary describing the sampling methods used, the laboratory techniques employed, and the off-gas analytical results obtained appears in Pacific Northwest Laboratory document PNL-3181.

TABLE 2.2. Tritium Emissions

Chemical Form	Sample Type ^(a)	Concentration		Uncertainty, %
		POG-88	Stack ^(b)	
HTO	Scrubber	1.26 $\mu\text{Ci}/\text{m}^3$	1.08 nCi/m^3	5
HTO	^3H - ^{14}C Sampler	1.13 $\mu\text{Ci}/\text{m}^3$	0.96 nCi/m^3	5
HTO	^3H - ^{14}C Sampler	1.23 $\mu\text{Ci}/\text{m}^3$	1.05 nCi/m^3	5
HT	^3H - ^{14}C Sampler	<1 pCi/m^3	---	-
HT	^3H - ^{14}C Sampler	<1 pCi/m^3	---	-
RT	^3H - ^{14}C Sampler	2.9 nCi/m^3	2.5 pCi/m^3	5
RT	^3H - ^{14}C Sampler	6.4 nCi/m^3	5.5 pCi/m^3	5

a. ^3H - ^{14}C samples were taken sequentially.

b. Calculated from flow rate data.

TABLE 2.3. ^{14}C Emissions

¹⁴ C Species	Concentration				Uncertainty, %
Chemical Form	POG-88 ^(a)		Stack ^(b)		Sample 1/Sample 2
	Sample 1	Sample 2	Sample 1	Sample 2	
	(nCi/m ³)		(pCi/m ³)		
CO ₂	70	180	59	150	5/5
CO	41	0.7	35	0.6	10/30
CnHm	31	20	26	17	5/5

a. Samples 1 and 2 were taken sequentially.

b. Calculated from flow rate data.

TABLE 2.4. Iodine Emissions

Chemical Form	Sampling Site	Sampler Type	Concentration	Total		Error, %
				POG-88	Stack	
A11	Room 147	Scrubber/AgZ	1.0 nCi/m^3	1.0 nCi/m^3	(0.86 pCi/m^3) ^(a)	5
I_2	Stack	Resin	0.4 pCi/m^3	(1.2 nCi/m^3) ^(a)	1 pCi/m^3	25
A11 ^(b)	Stack	Charcoal	0.6 pCi/m^3			
A11 ^(b)	Stack	AgZ	<50 fCi/m^3			

a. Calculated from flow rate data.

b. All forms except I_2 .

TABLE 2.5. Semivolatile Emissions

Isotope	Detection Method	Concentration	Error, %
		(POG-88)	
^{79}Se	β^- Counting	$\left\{ \begin{array}{l} < 20 \text{ fCi/m}^3 \\ 1.6 \text{ pCi/m}^3 \end{array} \right.$	8
^{99}Tc			
^{106}Ru	γ Analysis	$\left\{ \begin{array}{l} < 200 \text{ nCi/m}^3 \\ < 50 \text{ nCi/m}^3 \\ < 3 \text{ nCi/m}^3 \\ < 0.1 \text{ nCi/m}^3 \\ < 6 \text{ nCi/m}^3 \\ < 100 \text{ nCi/m}^3 \end{array} \right.$	50
^{125}Sb			
^{126}Sb			
$^{125\text{m}}\text{Te}$			
^{134}Cs			
^{137}Cs			
All		$10^5 \frac{\gamma/\text{sec}}{\text{m}^3}$	

TABLE 2.6. Radionuclide Content of Particulate Matter

Isotope	Concentration, nCi/m^3	Error, %
^{106}Ru	44.	10
^{125}Sb	0.52	10
$^{125\text{m}}\text{Te}$	1.7	15
^{134}Cs	7.8	10
^{137}Cs	52.	10
^{144}Ce	0.82	10
^{154}Eu	0.28	30
^{155}Eu	0.18	15
^{241}Am	0.19	20

TABLE 2.7. Particulate Size Distribution Analysis

Parameter ^(a)	Empirical	Log Normal
Geometric mean diameter	0.13 μm	0.10 μm
Geometric standard deviation	1.51	2.01
Correlation coefficient	-	0.975
Mass mean diameter	-	0.55 μm
Particle concentration	168/ cm^3	-
Particle mass concentration ^(b)	84.7 pg/cm^3	-

a. All parameters relate to undiluted process off-gas before final HEPA filtration.

b. Assumed calcine density - 3 g/cm^3 .

3. TRU WASTE IMMOBILIZATION PROGRAM

C. R. Palmer - Project Manager

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

SUMMARY

Plans were completed for a two-year comparative study of alternative immobilization systems for transuranic (TRU) wastes. The program will compare materials properties, using standardized experimental test procedures, and will consider the alternative processes by means of preconceptual design analysis. A formal comparison model will be developed by applying concepts from utility theory-based decision analysis. In a pilot-scale process test of the in-can melting system, nonradioactive rotary kiln incinerator ash was incorporated in a borosilicate glass matrix.

PROGRAM MANAGEMENT - C. R. Palmer

Planning has been completed for a two-year assessment of alternative TRU waste immobilization systems in which a set of host materials and associated fabrication processes will be characterized for a reference waste. Behavioral properties of the waste forms will be characterized using a battery of standardized testing methods which will address the properties shown below. Process flowsheets and preconceptual designs will be prepared to evaluate the alternative processes. The experimental and conceptual information will be assimilated and evaluated in a multi-attribute decision analysis model to rank the alternative systems by preference.

Waste form properties to be evaluated are as follows:

- Physical Properties
 - Density
 - Porosity
 - Surface area
- Mechanical Properties
 - Compressive strength
 - Impact behavior
- Thermal Properties
 - Sintering behavior
 - Melting behavior
 - Volatility
 - Phase characterization
- Radiation Stability
 - Metamictization
 - Gas generation
 - Stored energy
 - Transmutation
- Chemical Durability
 - Multiparameter leach tests
 - Long-term static leach test
- Compositional Flexibility
 - Sensitivity to waste variation
 - Level of waste loading

DATA ANALYSIS - A. D. Chockie and C. R. Palmer

This task is responsible for management of the program experimental data base and for development and implementation of a utility theory-based decision analysis model. For data base management a computer software

package System 2000⁽¹⁾ is being used. The GLASS data base developed last year has been modified to accept the TRU data class, including nonglass waste forms. All characterization data for both glass and alternatives will be included in GLASS.

A draft plan and technical approach was completed for development of the decision analysis model and was issued for internal review. A Monte Carlo simulation model will be developed that will provide a preference ordering of the alternatives. This ranking will be based upon the relative value associated with different attribute performance levels, the estimates of each system's attribute performance, and the relative importance assigned to each attribute. The method will explicitly incorporate the uncertainties associated with the performance estimates for each immobilization alternative, as well as those arising from the assignment of relative performance level, and attribute importance. Sensitivity analysis will be employed to obtain an improved understanding of the significant factors involved in the preference ordering. Figure 3.1 diagrams the methodology.

WASTE CHARACTERIZATION - G. H. Bryan

This task has concentrated during the last quarter on acquiring a better understanding of the TRU wastes currently generated at facilities of the Rocky Flats Plant and the Los Alamos Scientific Laboratory (LASL). A literature review and three site visits were completed. Ferric hydroxide sludge from liquid waste decontamination and residue from incineration of combustible trash were tentatively selected as the reference wastes, and a sampling and characterization plan was submitted to both laboratories for critique and implementation. All analytical work for

these wastes should be completed by the end of the second quarter, FY-1980.

RADIOACTIVE PRODUCT DEVELOPMENT - G. H. Bryan

Arrangements are being made to obtain a small quantity (3 to 4 kg) of first- and second-stage ferric hydroxide sludge from the liquid waste treatment facility at Rocky Flats. A glove box is being prepared and furnaces installed to fabricate waste form samples from the sludge for microstructural characterization and actinide leaching experiments. It is expected that the sludge will be received during the second quarter of FY-1980.

PROCESS EVALUATION - L. K. Holton and L. L. Petkus

A pilot-scale in-can melting (ICM) experiment was completed during the reporting period. The experiment's chief aim was to evaluate the feasibility of adapting the high-level waste ICM process for treatment of TRU-contaminated dispersible solid wastes. Specific areas of evaluation included behavior of solids during handling and feeding, melting behavior of the ash/additives mixture, and characterization of the ICM off-gas and of the glass product.

A drum of rotary kiln incinerator ash was obtained from the Rocky Flats Plant, and a melter feed batch representing glass 79-160 was prepared using a barrel mixer (chemical compositions of the batch and the glass were reported in Reference 2). The preblended flocculent mixture was manually charged through the airlock of the ICM canister. Table 3.1 summarizes the test conditions.

The ash was processed without significant difficulties. Although ash entrainment in

FIGURE 3.1. Multi-Attribute Decision Analysis Methodology

the melter off-gas had been expected, pulsed-air flowback of the sintered metal filters was required only once during the 10-hr processing period. No bridging of the internal fins nor accumulation in the airlock or feed pipe occurred. The feed could be observed as it fell into the canister through a viewport at the top of the canister. Some separation of ash from glass formers took place, with the low-density ash tending to float in the disengaging section. However, this did not cause phase separation of the vitreous product.

The melting rate of the solids was much higher than those observed in our laboratory melts. Melting adequately accommodated at least 20 kg/h feeding, the maximum feed rate

attempted because of the limited amount of ash available. The carbon was observed to ignite as it fell into the canister, and the combustion flame obscured the melt surface. The exothermic oxidation possibly increased the melting rate.

The reducing environment imposed by the carbon depleted the glass of zinc oxide by reduction to zinc melt, followed by volatilization from the metal and reoxidation above the surface. Because the oxygen supply was inadequate to complete carbon oxidation, a sintered carbonaceous mass formed at the top of the glass phase. The extent of the zinc depletion and the composition of the sintered material will not be known until chemical analyses of the product are complete.

TABLE 3.1. Summary of Test Conditions for the TRU
Incinerator Ash In-Can Melter Experiment

<u>General Information</u>	
Calcine feed system used	Manual charging
Frit feed system used	NA
Off-gas treatment	Filter, condenser, scrubber
<u>Canister Information</u>	
Diameter, cm	20 Sch 40 pipe
Usable length, cm	91
Wall thickness, cm	0.82
Bottom thickness, cm	0.95
Material	304L SS
Fins, no. and dimen., cm	8 (91 x 6.8 x 0.64)
<u>Batch Information</u>	
Waste designation	RFP-TRU ash
Waste form	Ash
Waste loading, wt%	30
Frit type	79-161-PS
Frit form	Powdered glass formers
Additives	None
Batch Preparation	Pre-blended
<u>Operating Parameters</u>	
Time at melt temperature, hr	10.5
Canister surface temperature, °C	1050
Nominal melt temperature, °C	1030-1060
Maximum batch feed rate, kg/hr	20
Melting rate, kg/hr	6
Maximum power consumption, kW	19
Glass cooling rate, °C/hr	
Temp. 1050°C to 550°C	34
Range 550°C to 200°C	12
<u>Product Information</u>	
Product designation	RFP-TRU 79-160-PS
Weight, kg	63
Volume, m ³	0.026
Bulk density, kg/m ³	2400

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1. System 2000 Reference Manual, MRI Systems Corp., Austin, TX, 1974.
2. A. M. Platt and J. A. Powell, comps., Nuclear Waste Management Quarterly Progress Report, April through June 1979, PNL-3000-2, Pacific Northwest Laboratory, August 1979.

4. 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 krypton in solids during ion sputtering.

SUMMARY

Two sputtered deposits of iron containing rubidium metal have been prepared, but neither of these deposits accurately simulates the product expected from decay of ^{85}Kr entrapped in a metal matrix. The experiments suggest three methods for preparing a more representative sample, and work is proceeding with one of the approaches.

High-pressure loadings of two types of glass with krypton have been completed at pressures up to 37 MPa. Measurements of krypton release from some of these samples indicate that only about 0.4% of the krypton will be released in 10 years from a sample held at 420°C during the entire period.

ION IMPLANTATION OF KRYPTON DURING SPUTTER

DEPOSITION - E. D. McClanahan and

G. L. Tingey

Relatively high concentrations of krypton are trapped in metal matrices during sputter deposition.⁽¹⁾ The krypton is effectively contained in the matrix at temperatures expected for storage of the radioactive isotope ^{85}Kr . Concern, however, for the effect of buildup of small quantities of rubidium from decay of the ^{85}Kr has led us to attempt to co-sputter rubidium atoms concurrently with the other metallic components. The fraction

of rubidium required to simulate the decay product of a metal matrix containing ^{85}Kr depends upon the age of the deposit. For example, a 5-at.% krypton-nickel matrix where the ^{85}Kr is 6% of the total krypton content would contain 0.15 at.% rubidium after 10.7 yr. If it is indeed practical to produce rubidium-krypton metallic alloys, krypton release kinetics studied as a function of rubidium content should tell what effect if any the age of stored ^{85}Kr might play.

Properties of the element rubidium (e.g., its high vapor pressure, low melting point, low thermal conductivity, large atom size, and extreme chemical reactivity) made it prudent to design and carry out small-scale experiments of increasing complexity. The steps chosen were first to make a sputter-deposited rubidium-iron alloy, then a rubidium-krypton-iron alloy, and finally a rubidium-krypton-yttrium-iron alloy. The first step was carried out this quarter with only moderate success. An existing sputter-deposition system was modified as shown in Figure 4.1 to produce a rubidium-iron alloy on the 12.7-cm-dia substrate with rubidium content decreasing with the distance from the center. A thermionic discharge was produced in the apparatus at a total krypton pressure of about 0.5 Pa. The substrate and target surfaces were lightly ion etched to

promote adherence of the deposit to the former and facilitate wetting of the latter by the condensed rubidium to achieve maximum cooling. Approximately 1 g of rubidium was distilled into the shallow, 2.54-cm-dia cavity located in the center of the 12.7-cm-dia, mild steel, water-cooled target. This was done in the presence of the gas discharge, which served to heat the interior of the stainless steel tube, thus preventing unwanted condensation of rubidium along its length. After the rubidium was transferred to the target cavity, the stainless steel transfer tube was withdrawn through the push-pull seal until it was clear of the space between the target and substrate.

During transfer of the rubidium, an electrical short in the electron-emitting portion of the apparatus necessitated carrying out the sputter deposition at a reduced power and run duration. From the appearance of the 25- μm thick deposit, it was deduced that an initial layer of almost pure rubidium was laid down as a result of its high vapor pressure, low thermal conductivity, and presumably too high a target input power despite the power reduction caused by the electrical short. Over this layer was a brittle one that broke into bits when the rubidium layer reacted with the laboratory atmosphere when the system was opened. The composition as determined by x-ray fluorescence indicated approximately 2 at.% rubidium.

An alternative scheme for co-sputtering rubidium and iron that avoids working with metallic rubidium was also tested. A multi-component sputtering target was prepared by drilling 12 approximately equally spaced 0.6-cm-dia holes in a 3.8-cm-dia mild steel disk. The holes were filled with a 35 wt% mixture of rubidium chloride and iron powder. A sketch of the experimental arrangement is shown in Figure 4.2. A 4-hr deposition run

using 2-kV krypton ions to bombard the target with a current density of 8.8 mA/cm^2 produced a film 64 μm thick. X-ray fluorescence analysis indicated the presence of rubidium and chlorine in the iron matrix. The deposit surface was distinguished by the presence of depressions, 3 to 4 mm in diameter, directly across from the holes in the target filled with the Rb-Cl-Fe mixture. Such depressions could be explained if the chlorine atoms are ejected from the target as negative ions accelerated to 2 kV by the strong electric field near the target surface. The energetic focused chlorine ions would cause sputtering to take place and essentially remove the film at about the same rate as it was formed. X-ray fluorescence analysis showed the Cl/Rb atomic ratio to be as high as 10 in the depressions. Elsewhere on the substrate the Cl/Rb ratio was one. The 25- μm deposit on the anode shield surrounding the substrate was also analyzed by x-ray fluorescence. Chlorine was not detected at all, and the average rubidium content measured 7.8 at.%. The x-ray diffraction pattern for the Rb-Cl-Fe deposit indicated a highly strained bcc Fe lattice with three weak unidentified diffraction lines. An x-ray diffraction pattern for the deposit on the anode shield has not been obtained.

Although we have successfully sputtered films containing rubidium, it is apparent that these films do not correctly simulate the alloy expected from the decay of entrapped ^{85}Kr . The importance of studying the rubidium effect and the difficulty and long decay time in producing the alloy via ^{85}Kr decay provide sufficient incentive to continue this effort. Our experience during the past quarter suggests the following three directions of experimentation:

1. Design a perforated sputtering target with an internal cavity containing metallic

FIGURE 4.2. Multicomponent Target Sputtering Apparatus

rubidium, the whole of which would be maintained at the temperature required to give the proper rubidium content to the sputter deposit. The matrix atoms would be sputtered while the rubidium vapor effuses through the perforations in the target.

2. Co-sputter RbCl or RbF mixed with Fe powder, but collect for testing only the material ejected beyond the angle where the halide is found.
3. Search for a rubidium compound that could be sputtered without incorporating other elements in the deposit deleterious to testing the rubidium effect.

Experiments are currently planned to prepare a sample using the second of these options.

DISSOLUTION OF KRYPTON IN LOW-DENSITY GLASS -

G. L. Tingey and W. J. Gray

An alternative technique for storing ^{85}Kr is to dissolve or trap the krypton in low-density glass rods at high temperature and

high pressure. The temperature is maintained at the elevated temperature to sinter the glass; this yields a product with very good retention of the krypton.

During this study, we have loaded krypton in two glass types. One was a SiO_2 glass marketed by Corning Glass under the trade name Thirsty Vycor.[®] The second was a glass containing additives that lower the sintering temperature by about 100°C . The sintering characteristics of both glasses have been studied and reported earlier.⁽²⁾ At present, studies have been limited to pressure at or below 37 MPa, with temperatures and sintering times being varied in an attempt to determine the optimum conditions for loading the krypton in the glass. Experiments using Thirsty Vycor have shown that loading of from 6 to 7 cm^3 of $\text{Kr}(\text{STP})/\text{g}$ of glass can be obtained at temperatures as low as 850°C with sintering times of 4 hr or more. Longer heating times do not appear to yield any better products either in krypton content or retention of the krypton. At

900°C, 1 hr is sufficient to obtain the maximum krypton loading and retention.

Only a limited number of experiments have been conducted using the lower temperature sintering glass, but current data show that loadings of from 4 to 6 cm³ of Kr(STP)/g of glass can be obtained when heating to 750°C and sintering at that temperature for 4 hr. The krypton release rate from this material appears to be about the same as observed for the Thirsty Vycor.

For the scoping experiments described above, krypton retention is determined by measuring the rate of release as a function of temperature while heating the sample at a rate of about 200°C/hr. Although these tests

give a good measure of total krypton content and of the temperature at which major release occurs, they do not accurately reflect the low temperature, long-term release characteristics important when considering storage of ⁸⁵Kr. We are, therefore, conducting long-term studies of krypton release at a very low rate. At present, we have measured the rate from Thirsty Vycor at 420°C for periods in excess of 400 hr. The fractional release follows roughly the expression $Kr_R = 1.4 \times 10^{-5} t^{1/2}$, where Kr_R is the fraction of krypton released at time t in hours. If this equation holds for the long term, 0.4% of the krypton would be released over a 10-yr period at 420°C.

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5. 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 treatment of irradiated fuels.

SUMMARY

Statistical analysis was completed on the data from phase III of the iodine-impregnated cement leach test. The four factors most significantly affecting the iodide leach rate were the solubility of the fixation compound in water, concentration of iodide fixation compound in the cement, the addition of bentonite to the cement, and CO₂ pretreatment.

Results from the carbonate-impregnated cement leach test indicate the most significant factors affecting the carbonate leach rate to be CO₂ leachate concentration, the concentration of carbonate fixation compound in the cement, and the addition of barium ion and strontium ion to the cement.

Iodine removal from iodine-loaded, metal-exchanged adsorbents using CO and H₂O resulted in a detectable amount of CO₂ and iodide. The release rate of hydrogen iodide was low, ~4 µg I/cm²/min.

STUDIES ON IODINE LEACHING - K. D. Wiemers

The first two phases in the study of iodine leaching from iodine-impregnated cement involved a parametric investigation of dynamic leaching methods and a screening study of factors suspected of affecting the iodine (as iodate) leach rate.⁽¹⁻³⁾ This report summarizes the results of phase III, a

screening study to determine factors affecting the iodine (as iodide) leach rate. The seven factors screened were solubility of the fixation material, concentration of the iodide in cement, addition of silver ion, barium ion, bentonite, and sodium resinate to the cement, and a carbon dioxide pretreatment procedure. These factors were described in detail in Reference 4.

To statistically screen these factors a 12-trial Plackett-Burman design⁽⁵⁾ was employed. The incremental leach rate,

$$R = \frac{a_n}{A_0} \frac{V}{S} \frac{1}{t_n}$$

where R = incremental leach rate

a_n = quantity of iodide leached during period, t_n

A₀ = initial quantity of iodide present in specimen

V = geometric volume of specimen

S = exposed geometric surface area of specimen

t_n = leach period

was selected as the dependent variable for the statistical analysis because it was independent of prior leach data and normalized for both volume and surface area. To determine the relative importance of the evaluated factors, the absolute value of each factor effect was ranked from largest to smallest. The top four factors were retained for consideration. The relative significance of

these factors as a function of time is given in Figure 5.1. The effects of these factors are described below:

- Increasing the fixation material's solubility in water increased the iodide leach rate. The effect was significant at a 95% confidence level throughout the 15 leach days.
- Curing the specimens in a humid CO_2 environment decreased the iodide leach rate. During this pretreatment, only the top surface was exposed. Further tests exposing the total specimen surface will be conducted to study the effect of CO_2 pretreatment.
- An increased quantity of iodide salt added to the cement raised the iodide leach rate. This effect was observed throughout the 15 leach days, with a 90% confidence level on days 2, 3, and 15.

- Addition of a soluble barium salt to the cement decreased the iodide leach rate during leach days 7 to 12.
- Addition of bentonite to the cement decreased the iodide leach rate most consistently during leach days 7 to 12. During this period the factor reached 90% statistically significant confidence level.
- Addition of a soluble silver salt to the cement decreased the iodide leach rate during the first 9 days of leaching.
- Addition of sodium resinate to the cement caused an occasional increase in the iodide leach rate.

The following comments concern possible explanations for the effects listed above. These comments are largely conjecture as more detailed studies are required to unravel the complexity of interactions in the iodine-cement-leachate system. In these

FIGURE 5.1. Significant Factor Effects for Phase III

comments a positive effect corresponds to an increased iodide leach rate and a negative effect to a decreased iodide leach rate.

The most statistically significant effect was the solubility of the iodide fixation material. The wide range of solubilities selected in this study--from 10.64 to $1.3 \times 10^{-4} \text{ M I}^-$, compared with a difference of 2 orders of magnitude in the iodate leach study^(2,3)--may explain why this factor was effective in the iodide but not the iodate leach study. The positive effect suggests that selection of the fixation material should be limited to the less soluble iodide salts.

The negative effect of curing in a humid CO_2 environment may result from the formation of an impregnated-type carbonate coating. Further studies are necessary to evaluate use of this technique for stabilizing iodine in cement.

The positive effect of iodide concentration in cement between the range of 3 to 10 wt%, g I/g dry cement, agrees with results obtained in the iodate leach study.^(2,3) These results suggest that a compromise between resistance to leaching and amount of fixation material encapsulated may be required.

The negative effect of the addition of barium ion may be explained by the precipitation of sulfate present in the cement which otherwise would be available for exchange with the cation of the fixation compounds; barium sulfate is considerably less soluble than the sulfate salt of the cation of the fixation compounds (see Table 5.1). No explanation is available for the lack of significance of barium ion addition in the iodate-impregnated cement system.⁽³⁾

The negative effect of bentonite may be attributed both to its inert, pore-filling

TABLE 5.1. Solubilities Related to Iodine-Impregnated Cement

Salt	Solubility
BaSO_4	$1.0 \times 10^{-5} \text{ M Ba}^{+2}$
SrSO_4	$5 \times 10^{-4} \text{ M Sr}^{+2}$
PbSO_4	$1.3 \times 10^{-4} \text{ M Pb}^{+2}$
MgSO_4	2.9 M Mg^{+2}
HgSO_4	$2.0 \times 10^{-3} \text{ M Hg}^{+2}$
CaSO_4	$3.0 \times 10^{-3} \text{ M Ca}^{+2}$

characteristics and its effect of increasing the plasticity of uncured cement. The observation of an effect of bentonite at the 5 wt%, g bentonite/g dry cement, level but not at 1 wt% as tested in the iodate leach study^(2,3) more probably indicates an effective concentration threshold rather than a difference in iodide and iodate systems.

In agreement with results from the iodate leach study, the negative effect of the addition of silver ions to the cement indicates that silver ions may precipitate any free iodide resulting from solubility and/or exchange processes. This effect also supports the iodide solubility effect noted above.

The unexpected positive effect of the air-entraining agent, sodium resinate, may be due to addition of an improper quantity of the reagent. Several samples independent of the statistical screening test were prepared to investigate this area. Leach data are now being analyzed.

Based on the results obtained to date in the leach studies of iodine-impregnated cement, the following recommendations are made for selection of a final fixation form:

1. Within acceptable criteria for thermal stability, cost, and availability, the fixation material should have a low solubility in water.
2. Concentration of the fixation material should be limited to 3 to 5 wt%, g I/g dry cement.

3. Options for increasing the leach stability include:

- CO₂ pretreatment
- silver ion addition to the cement
- bentonite addition to the cement
- barium ion addition to the cement.

Further evaluation of these parameters will include a more detailed study of the effective factors and/or a long-term study using several of the potential terminal fixation forms.

STUDIES ON CARBONATE LEACHING - R. D. Scheele

The leaching study to screen factors suspected of influencing the leach rate of ¹⁴C immobilized as carbonate in cement has been completed. The six factors studied using a Plackett-Burman experimental design⁽⁵⁾ were 1) high initial carbon dioxide concentration in the leachate, 2) solubility of the carbonate fixation form, 3) carbonate fixation form loading in the cement, and 4,5,6) addition of strontium ion, barium ion, or sodium resinate to the cement.

These factors were selected for the following reasons:

1. High initial carbon dioxide in the leachate was found in previous work⁽³⁾ to reduce the leach rate of iodate from iodate-impregnated cement possibly by formation of calcium carbonate on the surface and in the pores of the cement specimens.
2. The solubility of the fixation compound determines the equilibrium carbonate concentration and hence the diffusional driving force.
3. A different loading may change the accessibility to the leach solution.
4. Strontium carbonate has a very low solubility product, 1.1×10^{-10} ; thus strontium ion may inhibit carbonate migration.

5. Barium sulfate has a low solubility product, 1.1×10^{-10} , and may prevent interaction between free sulfate in the cement and the cation of the fixation compound. Sufficient barium was added to precipitate 20% of the normal sulfate content. Barium carbonate also has a low solubility product, 5.1×10^{-9} ; barium ion could also serve as a precipitant for mobilized carbonate.

6. Sodium resinate is an air-entraining agent and may reduce the permeability of the cement by changing the pore structure. Table 5.2 presents the factors and their experimental levels.

The cement specimens were prepared by adding the ¹⁴C-traced strontium and potassium carbonates as solids, strontium and barium nitrate as saturated solutions, and the sodium resinate as 5% solution. The leachate was saturated with CO₂ by the addition of dry ice to the leachate ~1 h before use of the leachate. Beta analysis was used to measure the leached ¹⁴C. Potassium carbonate impregnated cement specimens with no added β activity were leached, and the leachate was counted to determine maximum ⁴⁰K contribution to the measured activities.

Table 5.3 presents the experimental sample and environmental conditions for each trial, cumulative fraction leached normalized to account for cement specimen size variations, the calculated factor effect, and the factor effects ranking for the 12 trials required by the Plackett-Burman design. High initial carbon dioxide concentration in the leachate was the most significant factor causing increased leach rates. The next four "most significant" factors had nearly equal effect levels. High carbonate fixation compound loading increased the leach rate, strontium and barium nitrate additions to the cement reduced the leach rate, and low

TABLE 5.2. Factors Screened in Carbonate Leaching Study

Factor	Experimental Levels
Solubility of carbonate fixation compound	10^{-5} , 8.1 M CO_3^{-2}
Concentration of carbonate fixation compound in cement	0.03, 0.15 g carbonate/g dry cement
CO_2 in leachant	Ambient and saturated
Strontium nitrate addition to cement	0, 0.14 g $\text{Sr}(\text{NO}_3)_2$ /g dry cement
Barium nitrate addition to cement	0, 0.03 g $\text{Ba}(\text{NO}_3)_2$ /g dry cement
Sodium resinate addition to cement	0, 0.01 g sodium resinate/g dry cement

TABLE 5.3. Summary of Carbonate Leaching Experiments

Trial	Solubility, M	CO_2 in Leachant	Addition, g/g Dry Cement				Cumulative Fraction Carbonate Leached After 25 days, cm
			Carbonate	Strontium Nitrate	Sodium Resonate	Barium Nitrate	
1	10^{-5}	Ambient	0.15	0.14	0.01	0.03	2.54E-5
2	10^{-5}	Saturated	0.03	0.14	0.01	0	6.70E-3
3	8.1	Saturated	0.15	0.14	0	0	8.55E-3
4	10^{-5}	Saturated	0.15	0	0	0	1.19E-2
5	10^{-5}	Ambient	0.15	0	0	0.03	2.25E-4
6	10^{-5}	Ambient	0.03	0	0.01	0	5.09E-4
7	8.1	Ambient	0.03	0.14	0	0.03	4.19E-4
8	8.1	Saturated	0.03	0	0.01	0.03	8.86E-3
9	8.1	Ambient	0.15	0.14	0.01	0	1.41E-3
10	10^{-5}	Saturated	0.03	0.14	0	0.03	3.69E-3
11	8.1	Saturated	0.15	0	0.01	0.03	7.79E-3
12	8.1	Ambient	0.03	0	0	0	1.80E-4
Factor Effect ^(a)	-6.93E-4	+7.45E-3	+1.59E-3	-1.44E-3	+5.51E-5	-1.37E-3	
Rank	5	1	2	3	6	4	

a. Calculated using Plackett-Burman Statistical Analysis Techniques⁽⁵⁾

carbonate fixation compound solubility also reduced the leach rate. The effect of sodium resinate was much lower and was considered negligible. Two cement samples were leached with the maximum amount of traced strontium and potassium carbonates added to the cement. No other additions to the cement or leachate were made. The cum-

ulative fractions leached after 25 days were 3.05×10^{-4} and 3.47×10^{-3} cm, respectively. These leach data support the effect of both carbonate fixation compound solubility and concentration in the cement on the carbonate leach rate. Figure 5.2 presents the normalized cumulative fraction of ^{14}C leached from the cement as a function of

FIGURE 5.2. Cumulative Fraction ^{14}C Leached from Cement Versus Time

time for trial 1 and the reference maximum strontium carbonate loading. The normalized, cumulative fraction of ^{14}C leached =

$$\frac{\sum a_n}{A_0} \cdot \frac{V}{S}$$

where a_n = quantity of ^{14}C leached during period, t_n

A_0 = initial quantity of ^{14}C present in specimen

V = geometric volume of specimen

S = exposed geometric surface area of specimen.

Trial 1 had the lowest leach rate of any trial. This comparison in Figure 5.1 is presented to show that by selection of the

carbonate's chemical form and additional additives the leach rate can be reduced substantially.

Regarding the effect of carbon dioxide in the leachate, we made no effort to capture carbon dioxide volatilized from the acidic CO_2 saturated solution. If this loss occurred, the measured factor effect would be greater but would probably not change the screening analysis. We expected a reduced leach rate from a high CO_2 leachant content. The CO_2 present as carbonate in the leachate would precipitate on the surface of the cement as calcium carbonate, which would then block the pores in the cement, reducing

carbonate leaching. However, this did not happen. The increased leach rate could be due to carbonate exchange or to reduced pH. Normally cement buffers the leachant to about pH 10. However, we found some solutions with a pH value lower than 7.3 after 0.7 day contact with the cement sample. The analysis of the pH effect is continuing.

Ag ZEOLITE RECYCLE - K. D. Wiemers,
R. D. Scheele, and L. L. Burger

Several reactions have been examined for removing the iodine from the silver zeolite bed. The use of hydrogen was demonstrated at Idaho National Engineering Laboratory and confirmed by Pacific Northwest Laboratory workers and others. The reaction with N_2O was unsuccessful. No detectable reaction occurred either in bulk experiments or in microscale thermal experiments. Results from ozone experiments are not yet conclusive, and further tests are being made. The use of methane was suggested by B. J. McDonald at the Naval Research Laboratory (Washington, DC).

We are currently examining the reaction of iodine-loaded silver zeolite with $CO + H_2O$ and are making plans to test the reaction with unsaturated hydrocarbons.

For the reaction

$CO + H_2O + 2AgI \rightarrow CO_2 + 2HI + 2Ag$
free energy calculations indicate a vapor pressure of HI of about 6×10^{-4} atm at $300^\circ K$ and rising to $\sim 10^{-1}$ atm at $900^\circ K$. At temperatures above about $800^\circ K$, the vapor pressure of AgI becomes great enough (0.02 torr at $900^\circ K$) that loss on transport of the silver may be significant.

Preliminary experiments have been carried out using both iodine-loaded silver mordenite and pure silver iodide on silica gel and on sodium mordenite. Carbon monoxide and water

were passed over the samples, and the product was examined for hydrogen iodide using water bubblers, for I_2 using NaOH bubblers, and for CO_2 by infrared spectroscopy. The silver iodide-coated materials were prepared by submerging the base supports in a solution of silver iodide in ammonium iodide. The AgI-coated materials were then dried on a hot plate until releases of ammonia and iodine were no longer observed. The materials were then transferred to an oven and dried in air at $500^\circ C$. The two base supports used in these studies were silica gel and sodium mordenite extrudates.

In the first experiment with iodine-loaded silver mordenite at temperatures up to $523^\circ K$, iodine was observed during temperature equilibration in air, but upon exposure to CO and H_2O , no CO_2 or iodine species were detected. In a second experiment using silver iodide on silica gel, CO_2 and H_2O were introduced at $723^\circ K$. Iodide was detected at a rate of $0.45 \mu gI/cm^2 \text{ min}$. Neither iodine nor CO_2 was detected. The third experiment employed silver iodide-coated sodium mordenite. This material was equilibrated in air at $723^\circ K$ for 4 hr. During this period neither iodine nor CO_2 was detected. Immediately after exposure of the material to CO and H_2O , CO_2 and iodide were detected. The average release rate for iodide was $3.8 \mu gI/cm^2 \text{ min}$. No iodine was detected.

Although this is a very low release rate, it is possible that with appropriate temperature and support materials iodine removal by carbon monoxide and water may be feasible. Preliminary thermal analysis experiments were not encouraging. Pure AgI showed no weight change up to the point of AgI volatilization, $\sim 870^\circ K$, when exposed to a stream of $CO + H_2O$. Further tests will be made on the zeolites.

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6. WASTE MANAGEMENT SYSTEM STUDIES

J. V. Robinson - Project Manager

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

SUMMARY

A draft report, A Seismic Data Catalog and Associated Graphical Capabilities, was reviewed by seismologists, and wide distribution was suggested because of the system's unique potential utility in seismic research. Since hearing a presentation on kriging's applications, the Office of Nuclear Waste Isolation (ONWI) is considering further development of the technique for use in geologic site characterization.

Teknekron, Inc., submitted a final report, A Depth Optimization Study for Geologic Isolation of Radioactive Wastes. At DOE-RL's request it is being reviewed by ONWI. Teknekron also has explored and tested the Nominal Group Technique for use in analyzing issues for decision-making in management of radioactive wastes. With certain basic principles adhered to, the technique is found to stimulate effective dialogue that integrates the technical and nontechnical perspectives. A final report has been completed, submitted to DOE-RL, and at their request referred to the Human Affairs Research Center (HARC) for comment.

HARC hosted a workshop in Seattle, "Nuclear Waste Management: Understanding Social Assessments," and is arranging to conduct it also at PNL. Under other tasks several re-

ports were completed. A draft report was completed that analyzes public tradeoffs between different levels of electric power availability and risk to health and safety for different energy technologies. It was found that acceptability was strongly related to the type of technology and only weakly related to perceptions of risk and power availability. Also completed was the report entitled A Comparative Analysis of Network Television News Coverage of Nuclear Power, Coal, and Solar Stories. Under Task 4, four of the five reports were cleared, while the fifth one is undergoing revision. In Task 5, a report has been drafted analyzing conflicts due to discrepancies in perceived and calculated risk. Two reports are undergoing final review for Task 6. One, Attitude-Action Consistency and Social Policy Related to Nuclear Power, concludes that, contrary to common opinion, pronuclear and antinuclear groups show no difference in attitude-action consistency. The second report, designed to aid in the understanding of community action groups, is entitled Community Stress and Social and Technological Change: A Framework for Interpreting the Behavior of Social Movements and Community Action Groups. For Task 7, a report, Nuclear Waste Management and Environmental Mediation: An Exploratory Analysis, was

completed and sent to PNL for review. The report explores the major issues in the use of environmental mediation to resolve aspects of the nuclear waste management controversy.

Throughout FY-1979, PNL provided major assistance to the U.S./DOE delegation to the Nuclear Waste Management Working Group (WG.7) of the International Nuclear Fuel Cycle Evaluation (INFCE). Support was provided through specialized studies that included an analysis of waste management costs; pathway and dose calculations relating to various hypothetical release and consequence scenarios postulated for a salt and a hard rock (granite) repository; the evaluation of waste arisings from two LWR fuel cycles; and a summary of the design and performance considerations required to select an optimum package design for a given repository site.

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

During the reporting period a draft report entitled A Seismic Data Catalog and Associated Graphical Capabilities was reviewed by a group of five non-PNL experts in seismology. As a result, and also based upon PNL's own internal re-evaluation, the section dealing with seismic strain release was deleted and the sections dealing with the graphical display capabilities of the system were expanded. The peer review group suggested we proceed with publication and encouraged wide distribution of the report based upon the system's unique potential utility to those engaged in seismic research. The report will be submitted for clearance by DOE-RL early next quarter. The report was briefly summarized in a previous quarterly progress report, PNL-3000-1.⁽¹⁾

KRIGING* - P. G. Doctor

The feasibility of applying kriging in characterization of a geologic repository was demonstrated under the program as reported in previous quarterly progress reports. During the reporting period, our findings were presented to the Office of Nuclear Waste Isolation. ONWI is now considering further development of the technique for use in their geologic site characterization program, expected to intensify in the next fiscal year.

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

The objective of this study is to develop a simple algorithm relating the safety (occupational and radiological) of a geologic repository for the isolation of high-level radioactive waste to the repository's construction and operating costs. During the past two quarters, Teknekron, Inc., submitted a final draft of the report A Depth Optimization Study for Geologic Isolation of Radioactive Wastes. PNL comments were incorporated and the report was submitted to DOE-RL for clearance. At DOE's request, the report was submitted to ONWI for comment.

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

A study was undertaken by Teknekron to develop and test a methodology for analyzing issues related to management of radioactive wastes. The need for such a methodology

* For background information see Quarterly Progress Reports PNL-2378-3 and 4.

** For technical details see Quarterly Progress Report PNL-3000-1, May 1979.

stems from the complex nature of waste management issues, particularly those relating to permanent disposal in geologic formations. Because of the significant societal implications of applying permanent disposal technology, a host of nontechnical issues must be considered in the deliberations that lead to program decisions. Integrating these nontechnical considerations with the technical factors requires a multidisciplinary perspective and provides a significant challenge to a decision maker.

A review of historical attempts to integrate technical and nontechnical factors in the analysis of radioactive waste management issues revealed little in the way of systematic methodology. Accordingly, a technique was explored that was originally derived to facilitate a group's creativity in program planning. This Nominal Group Technique (NGT) has been widely applied in health, social service education, industry, and government organizations.

The NGT is a systematic approach to generating ideas or solving problems where several disciplines are represented and there is a need to integrate individual perspectives. It is particularly useful for situations where a single discipline would provide insufficient insight or knowledge to arrive at a successful solution to a complex problem. In the NGT, individual judgments are pooled so that the multidisciplinary group can invent or discover satisfactory courses of action. Hence, the process involves a strong iterative orientation to arrive at creative judgments or decisions.

In testing the methodology, the following principles were adopted:

- Group size should be kept small (six people) so that neither the focus on the objectives of the analysis nor the in-

sights derived from the disciplines involved will be lost.

- The group should be composed of both physical and social scientists because of the technical and nontechnical dimensions of the issue to be analyzed; each member should have had extensive exposure to nuclear waste management programs and issues.
- Definitive ground rules and guidelines should be developed before the meeting to govern the structured panel dialogue; constraints on the participants should be minimized, consistent with the need to keep the panel's efforts focused on and aiming towards the objectives while simultaneously stimulating the innovative and creative contributions desired.
- The participants should be exposed at the outset to the assigned (waste management) issue, its relevance to the (waste management) program, and the purpose and objectives of the analysis. The assigned issue (the nominal question) was presented in the following form: *Please list the issues a program manager should resolve to deal with the potential human intrusion of an aquifer potentially contaminated by radioactive waste that has migrated from a deep geologic repository.*

The program manager was described as an individual with any one of the following three roles:

- employed by a government agency, such as the Department of Energy or the Nuclear Regulatory Commission, to develop guidelines and standards for the long-term disposal of nuclear wastes
- employed by a firm with the responsibility to manage a number of deep geologic disposal sites or
- employed by the Department of Energy, the Nuclear Regulatory Commission, or a

private firm responsible for allocating research and development resources regarding the disposal of nuclear wastes.

The two-day panel discussion identified a broad range of sub-issues related to the nominal question, consolidated these into a tractable number of major issues, ranked the major issues, and analyzed the factors potentially leading to resolution of the issue. The exercise resulted in the following general conclusions:

1. The considerations that should receive greatest attention in formulating acceptable solutions to the nominal questions are:
 - a. determination of acceptable levels of risk associated with establishing and operating the deep geologic waste disposal system over the relevant time periods
 - b. establishment of design standards for the performance of the deep geologic waste disposal system
 - c. adequacy of the techniques and data for the physical modeling and analysis of the system for postulated waste release scenarios and
 - d. adequacy of the repository site characterization.
2. Provisions for engineered barriers and contingency planning (to deal with unexpected events in the disposal system) were viewed as moderately important considerations; their relative importance could vary, depending upon how the priorities were established for the items in 1 above.
3. Post-entombment monitoring and long-term institutional control of the repository and its environs were assigned low priority.

The experts unanimously agreed that resolution of considerations 1 a and b must

evolve through an iterative process between the derivation of design standards and the predicted risks associated with the application of those standards. Moreover, it was recognized that the credibility of the predicted risks is a function of appropriately satisfying considerations 1 c and d.

The results from testing the applicability of the NGT for effectively evaluating waste management issues led to the following conclusions about the analytical process:

- Physical and social scientists can effectively and productively engage in a structured dialogue within the NGT framework. This dialogue can result in a systematic identification of considerations that bear on an acceptable solution of the issue and also in consensus on courses of action designed to resolve the complex technical issues having both technical and nontechnical components.
- The size of the group selected for testing the NGT--six panelists plus moderator--appears reasonable in terms of maintaining control and focus of the group on the established objectives, sustaining the interest and participation of all panelists, and being sufficiently representative of critical disciplines to provide a meaningful iterative process for identifying and evaluating both the potential options for resolving issues and the factors on which these options depend.
- An effective and continuous dialogue between social and physical scientists is desirable in deliberating the factors that influence the alternatives for resolving the waste management issues due to the extensive interdependence of these technical and nontechnical factors.
- The extent that the analysis' product is defined in quantitative terms will be influenced chiefly by a) the degree to which

the panel participants are able to arrive at consensus positions on options for resolving the issue and b) the extent of the program manager's ("client" for the product) interaction with or participation in the NGT process.

A final report was completed during the reporting period and submitted to DOE-RL for clearance. At their request the report was referred to HARC for review and comment.

RISK, CONFLICT AND SOCIETAL ACCEPTANCE CONCERNING NUCLEAR WASTE AND ITS DISPOSITION -
S. M. Nealey (HARC)

Task 1, hosting a closed workshop in Seattle entitled "Nuclear Waste Management: Understanding Social Assessments," was completed during the second quarter. At DOE-RL's suggestion, arrangements were completed this reporting period to conduct a similar conference at PNL in October 1979.

The draft report for Task 2 was completed during this quarter. This task was an analysis of public tradeoffs between different levels of electric power availability and risk to health and safety for different energy technologies. An analysis of respondents' judgments of the acceptability of 36 hypothetical energy policies found that acceptability was strongly related to the type of technology and only weakly related to perceptions of risk and power availability. Antinuclear respondents indicated relative acceptance of conservation and solar technology and relative rejection of nuclear and coal technology. Pronuclear respondents accepted all four technologies, but supported nuclear power and coal slightly more than conservation and solar. Antinuclear respondents emphasized risk more than power availability. However, pronuclear respondents

emphasized power availability more than risk, especially when power availability was perceived to be low.

Task 3 was completed in the second quarter by issuance of the report, A Comparative Analysis of Network Television News Coverage of Nuclear Power, Coal, and Solar Stories. Final clearance was given third quarter on four of the five reports that constituted Task 4. It was determined late this quarter that the fifth report needed to be revised to reflect recent organizational changes within DOE. This revision will be accomplished the first quarter of FY-1980.

Under Task 5, an analysis of conflicts due to discrepancies in perceived and calculated risk, a rough draft of the report was completed this quarter. However, it was felt that the report would benefit from revision before submission for clearance. Therefore, it will be submitted as a draft report in the first quarter of FY-1980.

Task 6 has resulted in the issuance of two reports. The first, Attitude-Action Consistency and Social Policy Related to Nuclear Power, received PNL comments this quarter. It will be submitted to DOE-RL for final clearance during the first quarter of FY-1980. The major conclusion drawn in this report was that, contrary to popular opinion, pronuclear groups and antinuclear groups show no difference in attitude-action consistency. The second Task 6 report, Community Stress and Social and Technological Change: A Framework for Interpreting the Behavior of Social Movements and Community Action Groups, was submitted this quarter to PNL for review and comment. This report presents a combination of research conducted on political aspects of community organizations and on social movements. Through analysis of both literatures, a series of propositions were laid out, designed to aid in understanding

of community action groups, including those formed in response to waste repository siting issues. It should be emphasized that, as a first effort in this area, this report sought to present principles of organizational response that were grounded both in theory and in research. A major advantage of this approach is that the propositions about the likely behavior of action organizations are sufficiently general to apply in a variety of community settings and across different issue areas.

The report for Task 7, Nuclear Waste Management and Environmental Mediation: An Exploratory Analysis, was completed this quarter and sent to PNL for review. This report explores the major issues in the use of environmental mediation to resolve aspects of the nuclear energy waste management controversy. The nature of the waste management problem is discussed briefly, and a brief history of siting experiences is provided as background. Then environmental mediation is defined, and experiences with using this technique to resolve an environmental conflict are detailed. Environmental mediation is defined as a form of conflict resolution in which disputing parties voluntarily agree to settle their differences through use of a neutral third party. The applicability of mediation to the waste management conflict is then analyzed. While no conclusions are presented, several questions are raised that need to be resolved before applicability can be ultimately established.

Issues such as problem identification, appropriate participation, appropriate mediators, timing and the proper context for mediation seem most important to resolve. Interwoven with these questions are some particularly difficult issues. Can all participants take part in "good faith" mediation and not use the process of mediation as

a strategy to achieve some other ends? Can an appropriate mediator and funding sources be found that will be viewed as impartial by all disputants? Can value conflicts be separated out or identified in such a way that they do not preclude resolution of issues that are more readily compromised? And can mediation occur in such a context that its results can be implemented by national policymakers as well as by state and local officials? Dialogue is only beginning on all these points, and the appropriateness of mediation deserves much more detailed consideration before any decision is made regarding its usefulness.

INTERNATIONAL NUCLEAR FUEL CYCLE EVALUATION (INFCE)

During the whole of FY-1979, PNL provided major assistance to the U.S./DOE delegation to the Nuclear Waste Management Working Group (WG.7) of the INFCE. This support was provided under the auspices of the Waste Management Systems Studies program. PNL responded to requests for specialized studies of various aspects of the waste management systems related to the seven fuel cycles under investigation by WG.7. These tasks included, *inter alia*, an analysis of waste management costs; pathway and dose calculations relating to various hypothetical release and consequence scenarios postulated for a salt and a hard rock (granite) repository; the evaluation of waste arisings from two LWR fuel cycles; and summary of design and performance considerations required to select an optimum package design for a given repository site.

PNL provided staff assistance at the four WG.7 meetings held in Europe during 1979, and one staff member was assigned to DOE headquarters during the second half of FY-1979

to provide full-time assistance to the U.S. delegation as WG.7's final report approached completion. Descriptions of the major reports prepared by PNL follow.

One objective of the INFCE program is to evaluate and compare the health and safety impacts of different fuel cycles in which all radioactive wastes (except those from mining and milling) are placed in a geologic repository. To assist WG.7 to achieve this objective, PNL examined the radiologic dose to humans from both salt and hard rock geologic repositories containing waste arising as defined for seven reference fuel cycles.

The first report examined the release consequences for a generic waste repository in bedded salt. The top of the salt formation and the top of the repository were assumed to be 250 and 600 m, respectively, below the surface. The hydrogeologic structure above the salt consisted of two aquifers and two aquitards. The aquifers connected to a river 6.2 km from the repository. The regional gradient to the river was 1 m/km in all aquifers.

Hydrologic, transport, and dose models were used to model two release scenarios for each fuel cycle, one without a major disturbance and one in which a major geologic perturbation breached the repository immediately after it was sealed. The purpose of the modeling was to predict the rate of transport of radioactive contaminants from the repository through the geosphere to the biosphere and to determine the potential dose to humans.

Of the many radionuclides in the waste, only ^{129}I and ^{226}Ra arrived at the river in sufficient concentrations for a measurable dose calculation. Radionuclide concentrations in the ground water pose no threat to man because the ground water is a concentrated brine and it is diluted by a factor

of 10^6 to 10^7 upon entering the river.

Another report, prepared in the fourth quarter in cooperation with the Swedes and Canadians, evaluated the long-term behavior of the wastes from each of the seven fuel cycles when placed in a granitic rock or gneiss repository. Currently available hydrologic, leach, transport, and dose models were used to predict the rate of transport of radioactive contaminants from the repository through the geosphere to the biosphere and to estimate the potential dose to humans so that the release consequence impacts of the various fuel cycles could be compared. The scenario considered for release of contaminants from the repository was normal, slow water movement through rock of very low permeability.

The hydrologic model predicted that the water would travel approximately 7100 m to the biosphere in about 11,700 years. The one-dimensional transport model predicted that all discharge rates are small. The maximum individual doses, as calculated by the dose model, are very small and all of similar magnitude. Hence, no differentiations could be made among the seven fuel cycles based on the possible doses to an individual in the very far future.

In another report, prepared during this reporting period, radioactive wastes from the two reference LWR fuel cycle strategies of the INFCE Working Group 7 were described and volume generation rates per gigawatt-year of electricity were projected. In Strategy 1 (LWR once-through) wastes from uranium fuel fabrication, fuel irradiation, and spent fuel packaging were estimated. In Strategy 2 (LWR with U/Pu recycle) wastes from uranium and mixed-oxide fuel fabrication, fuel irradiation, and spent fuel reprocessing were considered.

The projections are based upon data developed for a recent environmental impact statement⁽¹⁻⁶⁾ in which the transuranic wastes (i.e., those wastes containing at least 10 Ci of certain alpha emitters) from fuel cycle activities were characterized. In addition, since the WG.7 assumes that all fuel cycle wastes except mill tailings are placed in a mined geologic repository, the nontransuranic wastes from several activities were included.

It was found that the rate of waste generation on a volume basis is dominated in each cycle by the reactor operations and decommissioning. These activities account for at least 85% of the fuel cycle waste volume (ignoring head-end wastes) when normalized to per unit electrical energy generated. However, at 10 years out of reactor, spent fuel elements in Strategy 1 represent 98% of the fuel cycle activity, but only 4% of the volume. Similarly, the packaged high-level waste, fuel hulls and hardware in Strategy 2 concentrate >95% of the activity in 2% of the waste volume.

As mentioned above, one of the waste forms considered for geologic disposal by INFCE Working Group 7 was spent, unprocessed fuel from the LWR fuel cycle. Since probably no one package design will be optimum for all sites, a report was prepared summarizing the design and performance considerations required to select an optimum package design for a given repository site. Also, the design of a packaging facility oriented to a specific fuel packaging concept was discussed, and the design features common to any packaging facility were enumerated. The packaging options considered vary from simple, single-walled canisters to complex, multi-wall containers. The spent fuel can be packaged "as is" or treated and compacted

to reduce volume. Stabilizer materials discussed include gases, metals, particulates, and nonmetallic solids.

It was concluded that the package option selected is strongly dependent upon the role it must play in a multi-barrier system for ensuring safe isolation of the waste. If the package must contain the waste for as long as 1000 years, then solid glass or alumina container/stabilizer options must be considered. If the spent fuel container is not required to meet any radionuclide control requirements, a simple carbon steel container with a gas stabilizer may be considered. If the option of storage and retrievability for a few years after emplacement (as opposed to disposal) is desired, container designs that enhance heat dissipation must be considered to ensure fuel integrity, and the canister materials will have to be chosen to resist corrosion by the geologic environment of the storage area.

In another report PNL prepared, the costs of managing high-level and transuranic-contaminated radioactive wastes were estimated for two nuclear fuel cycles--LWR with spent fuel disposal and LWR with recycle of uranium and plutonium. Both cycles were assumed to apply to a 100 GWe-yr/yr equilibrium power economy; also, all radioactive wastes except mill tailings were assumed to be disposed to a geologic repository of salt. These and other characteristics of the two cycles are patterned after those adopted for the International Nuclear Fuel Cycle Evaluation (INFCE) effort.

Facility and operating costs were obtained from the recently completed environmental impact statement⁽¹⁻⁶⁾ and a repository design study completed recently for the INFCE effort through the Office of Nuclear Waste Isolation. As several characteristics (e.g.,

facility size, waste production rates) differ between the EIS and INFCE, appropriate scal-

ing factors were used to extrapolate EIS costs to the INFCE cycles.

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5. United States Department of Energy, Technology for Commercial Radioactive Waste Management, Vol. 4: "Transportation, Final Disposal, Decommissioning and Thorium Fuel Cycle Considerations," DOE/ET-0028, May 1979.
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7. WASTE MANAGEMENT SAFETY STUDIES

W. K. Winegardner - Project Manager

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

SUMMARY

Development of a safety assessment approach for evaluating predisposal waste management systems and for applying the approach to selected facilities of two potential nuclear fuel cycles was completed with the submittal of a report for publication clearance. Comparison of the radiological consequences expected from chronic release of all ^{85}Kr from coprocessing activities with those expected from potential krypton recovery, packaging, and storage operations was also completed with the submittal of a report for publication clearance.

FUEL CYCLE ASSESSMENT - P. J. Pelto

Development of a safety assessment approach for the evaluation of predisposal waste management systems and application of the approach to selected facilities in the light water reactor (LWR) once-through fuel cycle and a potential coprocessed UO_2 - PuO_2 fuel cycle was completed with the submittal of a report for publication clearance. The safety assessment approach includes a scoping analysis on pretreatment waste streams and a more detailed analysis on proposed waste management processes. The primary evaluation parameters used in this study include radiation exposures to the public

from radionuclide release from normal operations and potential accidents, occupational radiation exposure from normal operations, and capital and operating costs.

The initial scoping analysis defines safety indices for the pretreatment waste streams based on such factors as radioactivity, volume, gamma power, heat generation, mobility, and toxicity. These indices can provide useful comparisons of waste streams within an individual fuel cycle and between different 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 values can provide be obtained. Building upon the above information, the public and the occupational radiation exposures are either estimated directly from the waste process details or relative safety indices are established. A combination of a preliminary hazards analysis and a generic list of potential accidents is used to assist in the quantification of the radiation exposure to the public from potential accidents.

Capital and operating costs are estimated using the best available data.

The initial application to the once-through and coprocessed UO_2 - PuO_2 fuel cycle waste management operations demonstrates the methodology, provides relative comparisons, and identifies information required for a more comprehensive analysis.

Overall, the waste management aspects of the two fuel cycles examined are much alike. On an individual facility basis, the fuel coprocessing plant has the larger waste management impact. As one would expect, the important waste streams on the basis of safety and cost are the spent fuel for the once-through fuel cycles and solidified high-level waste for the coprocessed UO_2 - PuO_2 fuel cycle.

As alternative fuel cycles are proposed, the suggested safety assessment approach may provide a ranking of the fuel cycles' waste streams and useful comparisons between various fuel cycles. This information can be used to direct safety research and development on a time and cost effective basis.

DISPERSION/ACCUMULATION TRADEOFF ASSESSMENT - P. J. Mellinger

Comparison of the radiological consequences expected from chronic release of all ^{85}Kr from coprocessing activities with those expected from potential krypton recovery operations was completed with the submittal of a report for publication clearance. The two basic waste management techniques considered are not new among modern industries. The existing technique is to release all the waste gas to the atmosphere, where it is diluted and dispersed. A potential alternative is to collect, concentrate, package and

submit the gas to long-term storage. This study compares the radiation dose commitments to the public and those to the occupationally exposed work force in an attempt to determine whether there would be a significant difference in radiation exposure to the worldwide population between these two alternatives. Table 7.1 presents a summary comparison of the 80-year cumulative dose commitments to the worldwide population from total chronic release as opposed to those from recovery, packaging and storage operations. These doses are compared with and without the mathematical expectation of dose incurred as the result of the probabilistic treatment of accidents. The wide range of occupational doses presented is the result of the evaluation of several design options.

The tradeoffs investigated for the management of noble gas suggest that activities associated with krypton recovery may reduce the worldwide population dose received from total chronic release of this radionuclide (see Table 7.1). Use of one of the more heavily shielded design alternatives for both recovery and storage would lower the worldwide dose from this waste management technique. Because the total exposures from the two alternatives are of comparable magnitude, however, additional investigation is needed to determine the overall merits of each approach. Comparison of worldwide population exposures from chronic releases of ^{85}Kr with occupational exposures to a relatively small number of workers each receiving thousands of times more cumulative dose may be misleading. Moreover, cost studies could show that inordinate amounts of money will be needed to further reduce the already insignificant 80-year cumulative dose of 0.045 mrem/person.

TABLE 7.1. Summary Table of 80-Year Cumulative Radiation
Dose Commitments to the World Population

	<u>No Kr Recovery,</u> <u>man-rem</u>	<u>Kr Recovery, (a)</u> <u>man-rem</u>
<u>Chronic Routine</u>		
Public exposure	2.9×10^5	2.6×10^4
Occupational exposure		3.7×10^5 to 4.2×10^1
Subtotal	2.9×10^5	4.0×10^5 to 2.6×10^4
<u>Accidental Release</u> ^(b)		
Public exposure		1.3×10^4
Occupational exposure		2×10^2 to 1.6×10^{-5}
Total	2.9×10^5	4.1×10^5 to 3.9×10^4

a. Based on 95% recovery and 50 years of interim storage.

b. Mathematical expectation of dose from the analysis of probability and extent of release of 15 accidents selected to bracket the spectrum of anticipated design-basis events.

8. WASTE ISOLATION SAFETY ASSESSMENT PROGRAM

A. Brandstetter - Project Manager

M. A. Harwell - Assistant Project Manager

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

- 1. develop the capabilities needed to assess the post-closure safety of geologic repositories*
- 2. obtain scientifically defensible generic and site-specific data necessary for safety assessments*
- 3. provide, as needed, studies to further support these data and analyses*
- 4. demonstrate the assessment capabilities by performing analyses of reference sites*
- 5. apply the assessment methodology to assist the National Waste Terminal Storage Program in site selection*
- 6. perform repository site analyses responsive to the time schedule and to the level of sophistication required to meet the licensing needs of the National Waste Terminal Storage Program.*

SALT DOME REFERENCE SITE INITIAL ANALYSIS -

M. A. Harwell

During this quarter, a large part of the activities of all WISAP tasks centered on the analysis of a reference site for a salt dome repository. This analysis provided the post-closure safety assessment for the Preliminary Information Report (PIR) being assembled by the Office of Nuclear Waste Isolation (ONWI). The WISAP PIR-related work included gathering of site characterization data from the site project managers and analyzing the various geologic phenomena that potentially could affect the repository's integrity. This analysis indicated that no plausible mechanism from geologic processes could be identified that would effect a breach of the reference repository in a salt dome; this conclusion is limited in that waste- and repository-induced effects were not considered and the near-dome hydrogeology was not adequately known.

The next step in the WISAP PIR work involved analyzing the reference repository for the potential for human intrusion. A human intrusion scenario was identified and quantified, and the consequence analyses were performed. These included dose calculations of direct pathways of radioisotopes to man and computer simulations of geotransport pathways to the biosphere.

The description of the methodology used and the results obtained were collated into a draft report that was transmitted to ONWI, DOE, and PIR subcontractors. This transmittal constituted meeting one of the WISAP control milestones for FY-1979. Following this, the calculations were rechecked and the work presented for numerous internal reviews. During the next quarter the report will be revised, based on the review process, and will be submitted for clearance.

TASK 1 - RELEASE SCENARIO ANALYSIS -

G. L. Benson

As a result of meetings between representatives of WISAP and the Basalt Waste Isolation Program (BWIP) of Rockwell Hanford Operations (RHO), the conceptual report PNL-2892 was revised, cleared by DOE-RL, and subsequently released as a draft report for use and peer review at the Task 1 Release Scenario Workshop held in Seattle during August.

Development of a more efficient program code for the Basalt Repository Simulation Model also began. The format allows simplified updating of documentation so as to reflect the state of the model at any given time and let the user investigate several levels of detail within the model. These levels progress in complexity from a general written description of the use and operation of the model and its components to the actual code programmed into the computer.

A literature search was begun to obtain geologic and hydrologic reports of three potential reference sites for use in a bedded salt repository simulation model.

The Task 1 release scenario workshop was held at the Battelle Seattle Research Center on August 13-15. The first day was used to describe the general methodology and approach of the model. The second day was primarily devoted to presentations by WISAP consultants concerning their contributions to the planning and development of the various sub-models. The final day of the workshop focused primarily on the near-field information needed to bridge the gap between development of the release scenario and analysis of hydrologic transport to guide the development work during FY-1980.

TASK 2 - WASTE FORM RELEASE RATE ANALYSIS -

D. J. Bradley and R. P. Turcotte

Studies are in progress to determine the mechanism and kinetics for UO_2 spent fuel release in aqueous solutions. The materials for this study are 18 single crystal UO_2 , doped UO_2 pellets, and spent fuel samples.

In this quarter, the first series of autoclave experiments were completed, performed at 75 C and 150 C with 200 ppm dissolved oxygen. A high initial dissolution rate was observed for single crystal UO_2 in NaHCO_3 . Both H_2O and NaCl decreased the dissolution rates of UO_2 because of the formation of surface film. Air oxidation of single crystal UO_2 at 285 C led to the formation of UO_{2+x} , and finally U_3O_8 .

Additional results were also reported for the waste glass leaching studies. In these experiments, uranium and americium displayed nearly identical release behavior in WIPP salt brine and bicarbonate solution. It was also reported that uranium and americium release was relatively high in the WIPP brine when compared with deionized H_2O and solutions of NaHCO_3 , CaCl_2 and NaCl , whereas for curium, neptunium, and plutonium, the release was lowest for WIPP brine.

About half the plutonium present in deionized water (pH 6) behaved like monomeric species in extraction tests. Most of the monomeric plutonium was present as Pu(V) rather than $\text{Pu(III)} + \text{(IV)}$. The nonmonomeric species, presumably a Pu(IV) polymer or a very finely divided solid material, was not converted to monomeric species on standing several days in $1\text{M HNO}_3 + 0.01\text{M NaF}$ at room temperature but was converted by digesting in such a solution for several hours at 95 C.

In 0.03M NaHCO_3 leach solutions (pH 9), most of the monomeric plutonium was present as $\text{Pu(III)} + \text{(IV)}$ rather than as $\text{Pu(V)} + \text{(VI)}$ as was found in the deionized water leach solutions. Most of the monomeric neptunium in both the water and the 0.03M NaHCO_3 leach solutions was present as $\text{Np(V)} + \text{(VI)}$. Nonmonomeric neptunium also appeared to be present, and preliminary results indicate that this material was not converted to monomeric species by the treatment that was effective in converting plutonium to monomeric species.

These preliminary results provide insight into the species of radionuclides present in leached glass solutions; however, additional work remains to identify more clearly the species present in these and other leach solutions.

A continuous flow leach test incorporating rock material was developed at Lawrence Livermore Laboratory (LLL); it involves a single-pass leaching system in which distilled water, bicarbonate water, or salt brine is continuously passed over a sample of rock material. The results will be compared with the standard International Atomic Energy Agency (IAEA) leaching method being used by PNL.

The large experimental matrix of autoclave tests has also been completed. Undoped glass beads doped with ^{99}Tc , ^{233}U , ^{237}Np , ^{239}Pu , ^{243}Am , and ^{244}Cm were leached in deionized water, sodium bicarbonate solution, and WIPP B salt brine. Test temperatures were 150°C and 250°C for 2, 4, 8, 16, and 32 days. Solution analyses have been completed for the undoped bead tests, and technetium and curium concentrations have been measured for the doped beads.

Releases of most elements to the salt brine were higher than for the deionized water and the sodium bicarbonate solution.

Silicon, molybdenum, and boron releases, however, were lowest in the brine. It is not clear why this occurred, although it is related to the solution's pH and the high salt content of the brine. A document discussing in detail the results of the solution analyses is being written.

In other experiments this quarter, cement waste forms have been prepared for the determination of plutonium release rates by the static leaching procedure. These specimens, which consisted of 30 wt% ash waste, 30 wt cement and 40 wt water, were prepared using the "cold" Rocky Flats rotary kiln incinerator ash that was spiked with a small quantity of Rocky Flats dual-chamber burner ash to provide a source of plutonium. These samples have a mass of 40 g and contain 128 mCi of plutonium.

TASK 3 - RELEASE CONSEQUENCE ANALYSIS - J. R. Raymond and R. W. Nelson

The Multicomponent Mass Transport (MMT) model has been refined to filter more effectively the statistical fluctuations present in the model's solutions. Technology development work continued with studies on the relationships of sample size or scale relationships to hydraulic conductivity and to stochastics as applied to ground-water hydrology. In this quarter, detailed electrical analog data and Hanford geohydrologic data have been used to test the application of geostatistical methods (kriging) in representing ground-water potential, permeability, and transmissivity.

Also in this quarter, a manuscript entitled "Analysis of a Numerical Solution to the One-Dimensional Stochastic Convention Equation" was drafted for future publication. The paper examines the adequacy of a numerical finite-difference solution scheme

for solving the hyperbolic convection equation that has uncertainty in the velocity field. A one-dimensional analytical solution is the basis for evaluating the numerical approach.

Documentations of the WIPP study and the Paradox test-case analysis were also completed and started through clearance. The first complete drafts of the Paradox sensitivity analysis report and MMT model report have been completed and are in final editing. Model draft reports for FE3DGW6 (the 3-D finite element hydrologic model), VTT (the 2-D finite difference hydrologic model), PATHS (the 2-D analytic hydrologic model), and the data system report all have been completed and are in the final clearance and publication process.

Testing and refinement continued on the extended version of FE3DGW, which solves the coupled equations for flow, heat, and macro-solute transport. Documentation of this code extension has begun. Problems that had arisen with transport flux terms are being handled by a refinement that estimates flux components at Gaussian, as opposed to model, points. These new revisions are being compared to analytical solutions, and work has begun on a mass balance checking and validation scheme.

Work has progressed well on the two-dimensional analytical and numerical solution capability for the stochastic ground-water equations. The coding is well along and debugging is getting under way. Preliminary runs indicated that considerable computing time is involved with the triple vector product evaluation. This probably will require special treatment later to speed up the evaluation process. An analytical solution of the smoothed stochastic equation in two dimensions has also been obtained for use in verifying the two-dimensional solution method.

The geostatistical work to represent potential and hydraulic conductivity variations over a region has been quite successful. It is based upon 144 measurements over the region on a regular grid system. Work is also under way to compare the block average conductance from kriging with the measurements taken on larger grid elements. The variance data obtained from the conductance values are very uniform over the central region due to the uniform measurements grid of the data. A similar uniform pattern of variance for the potential results; again this is due to the regular measurement grid used.

TASK 4 - SORPTION/DESORPTION ANALYSIS -

R. J. Serne and J. F. Relyea

In this quarter, investigations conducted at Argonne National Laboratory (ANL) determined that the adsorption of cesium on basalt depends on the cesium concentration. A nonlinear adsorption isotherm was detected over a range of concentrations from 10^{-13} M to 10^{-7} M Cs. Equilibrium was not reached after 15 and 18 days of sorption. The lack of equilibrium was attributed to diffusion of the nuclide into internal pores in the basalt and subsequent sorption on internal surfaces. It was estimated that internal surfaces were from 10 to 30 times larger than the external surface area, because of basalt porosity.

Investigators at Lawrence Berkeley Laboratory (LBL) have determined cesium adsorption isotherms to be a function of pH and cesium concentrations on a Belle Fourche smectite. Their results show a dependence of $K_d(\text{Cs})$ on the cesium concentration and on pH. The K_d values decrease with increasing cesium concentration or with decreasing pH. The work at LBL shows a decrease in $K_d(\text{Cs})$ of two orders of magnitude when the salt

concentration is increased from $2 \times 10^{-3} \text{ M}$ NaCl to 1.0 M NaCl.

Los Alamos Scientific Laboratories compared the batch and column methods using crushed material (tuff, argillite, and granite). Although residence times for strontium in the column and batch experiments were comparable, the observed K_d values were two or three times lower in the column experiments compared to the batch method. Similar results for cesium on crushed tuff were also observed. Experiments were begun using strontium and cesium in longer columns and with differing flow rates.

Work at Lawrence Livermore Laboratory has shown that column K_d values are lower than batch K_d values determined on the same crushed material (Thirsty Canyon Tuff). The disparity between column and batch methods was less than that with the Yucca Flat Tuff, perhaps because of longer residence time in the Thirsty Canyon Tuff columns.

Oak Ridge National Laboratory (ORNL) has shown that an increase in NaCl concentration reduces europium K_d values on Na-montmorillonite below pH 6; however, above pH 6, $K_d(\text{Eu})$ is increased by an increase in salt concentration.

Work at Pacific Northwest Laboratory (PNL) has shown that neptunium K_d values on the twelve minerals studied are one to two orders of magnitude higher under anoxic conditions than in an ambient atmospheric environment. PNL data also show that tracer ^{237}Pu ($<10^{-10} \text{ M}$ Pu) was adsorbed by teflon centrifuge tubes just as it was by other tube types. To study mineral effects on sorption of plutonium, Pu(VI) was added to vermiculite, biotite, kaolinite, and montmorillonite at pH 4. Vermiculite and biotite showed the largest sorption, even though their surface area and cation exchange capacities were

smaller than that of montmorillonite. Upon desorption, it was found that 20 to 30% of the sorbed plutonium on kaolinite and montmorillonite was exchangeable, but that desorption on biotite and vermiculite was negligible. Mossbauer analysis of the minerals for Fe^{+2} and Fe^{+3} showed no detectable Fe^{+2} in kaolinite or montmorillonite. The ratios of Fe^{+2} to Fe^{+3} in biotite and vermiculite were 1.5 and 0.4, respectively. The Fe^{+2} ion is a strong reductant for oxidized plutonium; the high sorption of Pu(IV) by biotite and vermiculite appears to be due to reduction of Pu(IV) by structural Fe^{+2} and sorption of the reduced plutonium species.

Studies by Rockwell Hanford Operations (RHO) showed that accelerated weathering of basalt by hot water increases sorption for cesium, neptunium, americium, and plutonium. The $\text{Sr}(K_d)$ values were increased by weathering basalt in air, but decreased when weathered under anoxic condition. Weathering of the argillite generally reduced K_d values for strontium and neptunium and increased K_d values for cesium. Americium and plutonium were found to sorb better on argillite weathered in air, but sorption on argillite weathered in nitrogen was less for americium and plutonium than sorption of fresh material. Weathering of granite had no significant effect on the sorption of cesium, americium, or plutonium, but appears to reduce the sorption of strontium and neptunium.

At ORNL technetium sorption on basalt was evaluated as a function of time and temperatures and found to be significantly greater at 22°C and 72°C than 0°C .

Sorption of technetium under anoxic condition on unweathered Conasauga shale from 1 M NaCl was considerably less than sorption on basalt. Sorption remains constant over the 5- to 30-day period studied, but seems to be

greater at 72°C than 0°C or 22°C. Uranium added to crushed granite at 72°C is sorbed much more under anoxic conditions than in air.

Workers at ORNL also reported that sorption of Np(V) on climax stock granite shows bright spots on autoradiographs that correspond to FeS₂, iron pyrites, and biotite. These data indicate reduction of Np(V) by Fe⁺² and sorption of the reduced neptunium species.

ACTIVITIES PLANNED FOR THE NEXT REPORTING PERIOD

Work during the next quarter is expected to be a continuation of ongoing efforts.

However, the specific work elements previously conducted under Tasks 1, 2, 3, and 4 will depend on discussions held with ONWI and DOE concerning the FY-1980 Assessment of Effectiveness of Geologic Isolation Systems and Waste-Rock Interaction Technology (AEGIS and WRIT) program plans, as well as acceptance of the proposed work elements by ONWI and DOE. AEGIS will continue the development of release scenario and consequence analysis methodology (former WISAP Tasks 1 and 3). F. Harvey Dove has joined the program to direct future site applications work. Efforts will also be made in support of ONWI and DOE site applications. WRIT will continue the waste form release and the sorption/desorption studies (former WISAP Tasks 2 and 4) and will also assist AEGIS in site applications as requested.

9. WELL-LOGGING INSTRUMENTATION DEVELOPMENT FOR SHALLOW LAND BURIAL

R. L. Brodzinski - Project Manager

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

SUMMARY

Preliminary results have been obtained from experiments on the thermal neutron activation analysis of ^{90}Sr . Physical parameters and experimental techniques have been optimized for maximum sensitivity.

Data reduction from experiments on the phoswich detector measurement of ^{90}Sr is nearly completed, and an open literature publication describing these results is being prepared.

All proof-of-concept experiments on the various subsystems of the tritium logging device were successful.

Data reduction and analysis of the photon detection well-logging experiments for actinide determinations have been continued.

DEVELOPMENT OF LOGGING SYSTEMS CAPABLE OF MEASURING ^{90}Sr - R. L. Brodzinski,

T. M. Tanner, N. A. Wogman, and D. P. Brown

Initial results of thermal neutron activation analysis experiments were inconclusive due to interferences from gamma-ray emitting activation products of impurities in the encapsulated ^{90}Sr source. The intensity of ^{72}Ga and ^{82}Br gamma-rays was sufficient to mask any ^{91}Sr gamma-rays which may have been present. A new source of ^{90}Sr in nitric acid solution was obtained and found to be

satisfactorily free of impurities. A special sample holder was designed and experimentally optimized to minimize the Bremsstrahlung radiation incident on the germanium diode gamma-ray spectrometer. The dimensions of the tapered collimator and absorber were adjusted to maximize the ^{91}Sr signal-to-noise ratio.

Experiments were completed which assessed the ability of a phoswich detector to measure ^{90}Sr from cased and uncased wells. Data reduction and analysis is nearly finished, and a paper entitled "Evaluation of a Phoswich Detector for the In-Situ Analysis of ^{90}Sr " by N. A. Wogman, R. L. Brodzinski, and D. P. Brown is being prepared. It is to be presented at the Institute of Electrical and Electronics Engineers 1979 Nuclear Science Symposium in San Francisco, California, October 17-19, 1979.

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

Laboratory experiments on all stages of the tritium logging system have been successfully completed. Satisfactory methodology has been developed for obtaining tritiated water samples from both liquids and gases by absorption through a semi-permeable membrane and subsequent thermoelectric freezing.

Tritiated water vapor has also been successfully reduced to elemental hydrogen. All subsystems are now ready to be optimized and mated for incorporation into the final logging device.

The efforts of this project were furthered by the work of Professor Clarence Menninga from Calvin College during the summer months. Dr. Menninga executed all the experiments for the tritium monitor and is largely responsi-

ble for having significantly advanced the state-of-the-art in a relatively short time.

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

Data reduction and analysis of the photon detection well-logging experiments for actinide determinations have been continued.

10. MONITORING AND PHYSICAL CHARACTERIZATION OF UNSATURATED ZONE TRANSPORT

S. J. Phillips - Project Manager

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

SUMMARY

A buried waste test facility used in developing systems to monitor shallow land burial of radioactive waste was used to collect continuous data from micrometeorological and hydrogeologic monitoring systems. An operational system is now at this facility collecting data essential to monitoring mass and energy transfer at arid land burial sites. However, additional refinement of monitoring systems is continuing.

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

The objectives of this task for FY-79 were completed during the third quarter.

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

The objectives of this task for FY-79 were completed during the second quarter.

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

Monitoring systems are being developed to evaluate the transport of radionuclides and water (the chief agent in the transport of radionuclides) at arid region radioactive

waste burial sites. For evaluating these monitoring systems under field conditions where they may be employed, a buried waste test facility (BWTF) has been constructed and has been in preliminary phases of operation since October 1979.

In situ hydrogeologic and micrometeorological sensors, data transmission, signal processing, and data collection and recording devices have been installed and made operational. Active and passive logging systems used to measure radionuclide and water migration through geologic media have also been developed and logging operations have begun. This task's basic objective during this reporting period has been collection and computer preprocessing of a complete data set from monitoring systems used at the BWTF.

Data from the BWTF is collected and analyzed primarily as three distinct data sets. These are: 1) micrometeorological data from instruments located above ground level, 2) hydrogeologic data from large caissons installed below ground level, and 3) hydrogeologic data from lysimeters installed below ground surface. Figure 10.1 illustrates the location of the micrometeorological systems and the hydrologic systems within the caissons and the lysimeters.

FIGURE 10.1 General Schematic of Electronic Sensing Devices Installed at the Buried Waste Test Facility

Eleven micrometeorological sensor-sensor arrays are shown in the figure: seven caissons consisting of an inner large access caisson containing instruments for collecting data; two outer caissons containing uncontaminated geologic media; four small caissons containing contaminated geologic media; and two weighing lysimeters. Within the weighing lysimeters and uncontaminated caissons containing geologic media are numerous sensors for monitoring parameters such as temperature, thermodynamic potential, water content, heat dissipation, etc. Typically data from all sensors are collected continuously after being signal processed and dumped to magnetic tape every 2 hr. Uniquely developed data loggers and associated input/output devices are used for this purpose. Data from this system are taken to the laboratory for processing and graphic display using a dedicated small microprocessor-based computer system.

Figure 10.2 illustrates a weighing lysimeter used primarily to determine the mass transfer of meteoric precipitation that enters the site and exits the site in the vapor phase as it is evaporated. At the BWTF one weighing lysimeter is used to monitor ambient conditions and the other to monitor increased precipitation conditions, thus indirectly measuring radionuclide transport. These consist of an outer box which separates the lysimeter from the geologic media, an inner platform scale to measure change of mass over time, an inner box supported on the platform scale containing geologic

media and, within the inner box, hydrogeological monitoring instruments. As water in the partially saturated zone is transported into and from the weighing lysimeter, it is monitored by thermal and thermodynamic water potential devices. If water above the amount that would flow vertically into the geologic media below the lysimeter accumulates in the lysimeter base, it is extracted by water extraction devices and collected and its volume determined and recorded.

Figure 10.3 is an illustration of the system used in the field at the BWTF for logging the concentration of radionuclides (cobalt) over the depth of contaminated caissons. Figure 10.4 is a more detailed drawing of the electronics configuration used with the above system. The relative concentration of ^{60}Co organically complexed with EDTA (an organic sealant) versus depth is logged on a weekly basis to determine the rate, magnitude, and direction of radionuclide transport at the site. The cobalt acts as a specific nuclide, actively transported under given conditions. In addition, because it is organically complexed and has a negligible distribution coefficient (K_d), it acts as a tracer for liquid phase water migration.

Data taken from micrometeorological transducers, hydrogeologic transducers, and weighing lysimeters have been processed in the laboratory by computer techniques. The data have been transferred from magnetic tape to disks for reformulating and subsequent parametric analysis.

FIGURE 10.2. Weighing Lysimeter

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FIGURE 10.3. Generalized Drawing of Downwell Radionuclide Counting System

79F041-2

FIGURE 10.4. Generalized Schematic of Downwell Radionuclide Counting System Electronics

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

J. A. Franz - Project Manager

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

SUMMARY

During this quarter solid crystalline samples have been isolated for Tc(IV) citrate, EDTA and DTPA complexes. Elemental analyses coupled with infrared absorption data are helping in determining complex composition. Electron spin resonance (ESR) equipment difficulties have been resolved, and examination of the complexes will begin. Recent literature has revealed that amperometric titrations can be successfully applied to the determination of the oxidation state of technetium at the trace level.

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

Several breakthroughs have occurred this quarter which will lead to a more complete characterization of Tc citrate and EDTA and DTPA complexes. Among these was the ability to isolate solid crystalline products for the above technetium (IV) complexes. This capability will greatly facilitate purification and characterization of product, important aspects because these complexes are earmarked for soil studies. The isolation of crystalline reaction products was made possible by substituting hydrazine (N_2H_4) for $SnCl_2$ as the reducing agent, followed by the addition of KBr (not required

for EDTA complex) and methanol once the reduction with hydrazine was completed. Large purple, cubic crystals were isolated for the citrate complex, while orange and pale orange crystalline solids were obtained for the EDTA and DTPA analogues, respectively. However, application of the same method (KBr + MeOH addition) was not as successful when $SnCl_2$ was used as the reducing agent; only the EDTA system yielded a crystalline product, and nothing was isolated for the other two ligand systems. Alternative solvent systems will be examined.

The Tc(IV) dicitrate generated by $SnCl_2$ reduction appears to be different from that generated by N_2H_4 reduction. $SnCl_2$ reduction produces a magenta-colored solution, while N_2H_4 reduction yields an intense purple-brown solution under similar reaction conditions. The electronic spectrum of these two dicitrate complexes is presented in Figure 11.1. When N_2H_4 is used as the reducing agent, the 560 nm band of the $SnCl_2$ -produced citrate complex shifts to higher energy (520 nm), with additional absorptions (shoulders) visible at ~370, 295 and 275 nm.

The differences may be due to the participation of $SnCl_2$ as $SnCl_3^-$ in complex formation. This behavior has been observed by Deutsch et al.⁽¹⁾ in the Tc(V) (dimethylglyoxime)₃-O- $SnCl_3^-$ prepared by $SnCl_2$ reduction. X-ray crystallographic data revealed the

FIGURE 11.1. Electronic Spectra of Tc(IV) Dicitrate Complexes in Water

presence of a Tc-O-Sn bond. Efforts are ongoing to define the nature of these two citrate complexes. The crystalline product from N_2H_4 reduction has been submitted for elemental analysis. An infrared absorption spectrum of this Tc(IV) dicitrate complex has been obtained and appears in Figure 11.2 along with infrared spectra of citric acid and two of its salts, $(\text{NH}_4)_2\text{H citrate}$ and $\text{Na}_3\text{ citrate} \cdot \text{H}_2\text{O}$, presented for comparison. The spectra focus on the COO stretching region. The uncoordinated unionized carboxyl stretching band occurs at 1750 to 1700 cm^{-1} , with the lower limit shifting in some cases to 1670 cm^{-1} , as has been observed in $\text{Na}_2\text{H}_2\text{ EDTA}$.⁽²⁾ Upon coordination, the COO stretching band shifts to 1650 – 1590 cm^{-1} ; the ionized free COO^- stretching bands occur at 1630 – 1575 cm^{-1} . In accord with the above, two bands are observed for $(\text{NH}_4)_2\text{H citrate}$, one at 1677 cm^{-1} for the unionized COOH and the second band at 1594 cm^{-1} for the two free ionized COO^- groups. Only one band, as expected, is observed for citric acid (B) at 1695 cm^{-1} . The trisodium salt exhibits the characteristic stretching band for free ionized COO^- groups at 1579 cm^{-1} . In complex D only

one stretching band is observed at $\sim 1619\text{ cm}^{-1}$, indicating that coordination has indeed occurred and that all three carboxyl groups are coordinated as only one band is observed. Efforts are also being directed towards the isolation of a solid Tc(V) citrate complex.

As mentioned earlier, when N_2H_4 is used as the reducing agent, a crystalline orange solid can be isolated for what appears to be a Tc(IV) EDTA complex. Addition of methanol to the orange-brown reaction solution resulted in the precipitation of the orange crystalline product. This sample was submitted for elemental analysis; the results are presented below. Had the sample been recrystallized, better agreement would have occurred between the calculated and observed percentages. A recrystallized sample will be resubmitted for analysis. On the basis of the elemental analysis and infrared spectra data, which will be presented later, the complex analyzes for Tc(IV) EDTA or Tc(IV) EDTA $\cdot 0.5\text{H}_2\text{O}$ are as shown in Table 11.1. The infrared absorption spectra of the COO stretching region in Tc(IV) EDTA, EDTA, and $\text{Na}_4\text{ EDTA} \cdot 2\text{H}_2\text{O}$ are presented in Figure 11.3. It can be seen that complexation has taken place as the carboxyl stretch in

FIGURE 11.2. Infrared Spectra of (a) $(\text{NH}_4)_2\text{H Citrate}$, (b) Citric Acid, (c) $\text{Na}_3 \text{ Citrate} \cdot 2\text{H}_2\text{O}$, and (d) $\text{Tc(IV) Dicitrate Complex}$

TABLE 11.1. Analytical Data on Tc(IV) EDTA

Compound	% Calculated			% Observed		
	C	H	N	C	H	N
$\text{TcC}_{10}\text{H}_{12}\text{O}_8\text{N}_2$	31.01	3.10	7.24	30.28	4.77	7.72
$\text{TcC}_{10}\text{H}_{12}\text{O}_8\text{N}_2 \cdot 0.5 \text{H}_2\text{O}$	30.38	3.29	7.09	30.28	4.77	7.72

the complex is shifted to lower energy, 1611 cm^{-1} , as compared to the free acid at 1697 cm^{-1} . That only one band is observed in the complex argues for the fact that EDTA is coordinating as a hexadentate ligand. Two carbonyl bands would have been observed if EDTA were coordinating as a pentadentate ligand; the second band would have appeared about $1740\text{--}1720 \text{ cm}^{-1}$, as has been observed by Busch and Bailar⁽³⁾ for some Co(III) EDTA complexes. The infrared absorption spectrum of the technetium (IV) DTPA complex will also be examined.

Efforts have been directed towards obtaining complex soil interaction data. Standard solutions of TcO_4 have been examined by liquid scintillation. The solutions analyzed reasonably well for the expected number of counts; however, counting discrepancies arose when Tc(IV) EDTA solutions were examined. These will have to be reexamined again before soil studies are started.

Instrumental difficulties which have hampered ESR studies of the technetium complexes have been resolved. This work will begin shortly.

FIGURE 11.3. Infrared Spectra of (a) EDTA, (b) Tc(IV) EDTA, and (c) $\text{Na}_4\text{EDTA} \cdot 2\text{H}_2\text{O}$

Another promising method for determining the oxidation state of technetium has recently appeared in the literature. This electrochemical method, amperometric titration, has been successfully applied by

Russell and Cash⁽⁴⁾ to the identification of oxidation states of technetium in bone-scanning agents. We plan to incorporate this very sensitive technique, applicable to trace level concentrations, in our studies.

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12. ELECTROPOLISHING FOR SURFACE DECONTAMINATION OF METALS

R. P. Allen - Project Manager

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

SUMMARY

The recently installed hot plasma arc torch system was successfully used to cut 15 ft² of plutonium-contaminated stainless steel into 8-in. squares suitable for processing in the vibratory finisher. Initial tests conducted for Rocky Flats using this material indicate that a vibratory finisher system utilizing metal media and a NaOH flushing solution will be capable of reducing plutonium-contaminated Rocky Flats glove box material to non-TRU levels.

Integrated operation of the disassembly, sectioning, pretreatment and electropolishing systems was successfully demonstrated. A plutonium glove box with 46 ft² of contaminated surface area was disassembled, sectioned using a hand-held plasma arc torch, and converted into non-TRU waste using a combination of electropolishing and vibratory finishing. The decontaminated materials included the stainless steel glove box floor, top and frame; the plastic panels; the plastic glove ports; the rubber gloves; and even the plastic bags. The only components not processed were the filters and the uncontaminated external hardware.

Use of an anti-spatter compound during sectioning successfully minimized airborne contamination and surface contamination of the sectioning facility. Additional work is

required, however, to better contain and handle the waste from the plasma arc torch cutting operation.

Hot testing of the vacuum evaporator system continued with the successful purification to reasonable levels of 250 gal of water representing a variety of plutonium-contaminated vibratory finisher, rinse, and cleaning solutions.

Substantial progress also has been made in further developing in situ decontamination methods. In a related project, the interior of a 5000-gal tank was almost completely decontaminated from the outside using a magnetically coupled contact in situ electropolishing device.

PROCESS DEMONSTRATION

Integrated operation of the disassembly, sectioning, pretreatment and electropolishing systems was successfully demonstrated. A plutonium glove box with 46 ft² of contaminated surface area (Figure 12.1) was disassembled, sectioned using a hand-held plasma arc torch, and converted into non-TRU waste using a combination of electropolishing and vibratory finishing. The decontaminated materials (Figure 12.2) included the stainless steel glove box floor, top, and frame; the plastic panels; the plastic glove ports;

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FIGURE 12.1. Plutonium-Contaminated Glove Box Being Sectioned by Use of Hand-Held Plasma Arc Torch

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FIGURE 12.2. Representative Decontaminated Material from Plutonium-Contaminated Glove Box Shown in Figure 12.1

the rubber gloves; and even the plastic bags. The only components not processed were the filters and the uncontaminated external hardware.

The glove box was about 24 in. long, 39 in. wide and 26 in. high. The floor, top and frame were constructed of 3/16-in. stainless steel; side panels were constructed of 1/4-in. Plexiglas.[®] The contents of the box included a ram and die set, several dies, and miscellaneous steel plates.

All noncontaminated external hardware was removed from the glove box before placing it in the sectioning facility. Final disassembly was performed inside the sectioning facility. Pneumatic wrenches, screwdrivers and impact hammers were used extensively to speed disassembly.

Once the nonmetallic components were removed, the remaining metallic structure was sectioned using a hand-held plasma arc torch. Most of the 32 ft² of stainless steel were sectioned into squares of approximately 8 in. for processing in the vibratory finisher. Three large sections of stainless steel, about 6 ft² each, were sectioned for direct immersion electropolishing.

During sectioning, an anti-spatter compound was used to control the spread of contamination and to reduce the tendency of the material ejected from the cut to stick onto the surfaces of the sectioning facility. No airborne contamination above ambient levels and only slight surface contamination due to the sectioning operation was detected. However, equipment must be developed to control the buildup of smoke in the sectioning facility and to better contain the material ejected from the cut.

The Plexiglas, representing about 14 ft² of the total contaminated surface area, was

sectioned by scoring and breaking into approximately 8-in. squares for decontamination by vibratory finishing. The sectioning of Plexiglas proved to be the major problem area for this demonstration. No technique has yet been identified for rapidly sectioning large Plexiglas panels into predictable sections. The gloves, gaskets, bagout bags, and miscellaneous plastic sheets were sectioned by shearing.

The stainless steel, Plexiglas, gaskets, gloves and miscellaneous sections were decontaminated simultaneously in the vibratory finisher. There was no need to segregate the material by type or size. The vibratory-finished material was then rinsed using a high-pressure water spray. Process times were typically 1 hr. All material was less than 10 nCi/g after decontamination.

The three large stainless steel sections were decontaminated to background levels by immersion electropolishing at 15 min/section.

DECONTAMINATION STUDIES

Vibratory Finishing

The evaluation of steel vibratory finishing media is continuing. Tests conducted in cooperation with Rocky Flats showed that the combination of steel media and sodium hydroxide compound is effective in reducing TRU contamination levels on stainless steel, Plexiglas, Neoprene[®] gaskets and Hypalon glove sections to less than 10 nCi/g and generally nonsmearable in 30 to 60 min. This demonstrates the ability to simultaneously decontaminate the principal metallic and nonmetallic materials from a typical

[®] Registered trademark of Rohm and Haas Co., Independence Mall West, Philadelphia, PA.

[®] Registered trademarks of the E.I. Du Pont de Nemours Co., 1007 Market St. Wilmington, DE.

glove box. Wear rates for the processed stainless steel were from 0.01 wt%/hr to 0.02 wt%/hr.

The steel medium used during these tests was 1/4-in. ballcones (Figure 12.3), a commonly used burnishing medium. The unique shape of this type of medium permits good liquid drainage through the vibrating tub, good medium-to-part contact for flat surfaces, and access to recessed surfaces on parts with complex geometries.

Several basic commercial burnishing compounds were evaluated during these tests. While all the compounds produced good burnishing results, not all achieved good decontamination; but the reason is not yet known. In addition, the commercial compounds have the disadvantage of producing foam in the evaporator system used for purification and concentration of the aqueous waste. Dilute 10% sodium hydroxide proved to be a very successful compound for use with metal media (prior tests showed it useful also in combination with ceramic media). Sodium hydroxide effects good decontamination and provides adequate rust inhibition necessary for preserving the steel media. Additionally, it is compatible with existing waste treatment systems at both Hanford and Rocky Flats.

During these tests, evaluation of the effectiveness of vibratory finishing with steel

media as a decontamination technique for copper and lead were performed. TRU-contaminated copper and lead sections were processed with the other materials mentioned above. The levels of fixed contamination on the lead and copper increased during vibratory finishing, although smearable contamination was reduced to nondetectable levels. Further tests will be conducted to determine whether the fixed contamination levels on these softer materials can be successfully reduced by changes in the vibratory finishing operating parameters.

Abrasive Blasting

Dry abrasive blasting techniques are used in the metal finishing industry to remove dirt and scale from surfaces requiring further manufacturing operations. The ability of dry abrasive blasting to remove plutonium contamination from stainless steel surfaces was evaluated by blasting the surface of representative glove box sections for 1 min using 16-mesh garnet sand propelled by 80-psi air pressure. The results of these tests are summarized in Table 12.1. These data show that most of the plutonium contamination was removed by this treatment. Moreover, decontamination was accomplished with only minimal removal (~ 0.25 wt%) of the base material.

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FIGURE 12.3. Steel Vibratory Finishing Media

TABLE 12.1. Effect of Dry Abrasive Blasting on the Plutonium Contamination Level of Stainless Steel Surfaces

Sample No.	Side 1 dis/min/cm ²		Side 2 dis/min/cm ²	
	Before	After	Before	After
1	6,100	400	3,000	400
2	4,400	400	2,800	500
3	>6,200	400	>3,300	300
4	5,300	300	3,600	400
5	4,400	400	2,000	300
6	7,600	500	3,800	400
7	5,300	300	3,300	500
8	6,200	300	6,400	300
9	9,800	500	6,300	400
10	6,400	400	3,500	300
11	>17,400	600	>17,400	700

However, these tests also disclosed that the minimum attainable contamination level was a definite function of the contamination level of the abrasive medium. This means that the abrasive must be changed frequently, resulting in excessive secondary waste generation, or else a method must be developed to separate contamination from the abrasive medium. Other disadvantages of dry abrasive blasting for plutonium-contaminated materials are that 1) it is difficult to ensure containment of the contaminated dust and 2) the process is labor-intensive, requiring continual attention by the operator.

In Situ Electropolishing

Work is progressing on development of in situ electropolishing devices. The ability to decontaminate large areas using this technique was demonstrated by reducing radiation levels on a 5000-gal liquid radioactive waste tank contaminated with corrosion products, mainly ⁶⁰Co, from 20 mr/hr to background with no detectable smearable contamination. This was accomplished using only 55 gal of elec-

trolyte. Moreover, 85% of the interior tank surfaces were decontaminated from outside the tank.

The tank, Figure 12.4, was cylindrically shaped, 24 ft long and 6 ft in dia, and built of 3/8-in. stainless steel. Access was gained through a single manhole located at the top center of the tank. A perforated sparger line, about 4 in. in dia and 24 ft long, ran through the tank. The tank had been used for many years to collect primary coolant from a power reactor during maintenance operations.

This decontamination was accomplished in situ using a series of electropolishing devices. Electrolyte was supplied to these devices by a small pump mounted in the sump of the tank. The electrolyte was allowed to flow from the devices, was collected in the bottom of the tank, and then recirculated through the devices.

Most of the decontamination, 85%, was done using a large swab device. This device had a cathode area of 60 in. and could decontaminate 2 ft²/min. It was magnetically coupled to a remote operating device that permitted manipulation from outside the tank.

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FIGURE 12.4. 5000-Gal Liquid Radioactive Waste Tank Decontaminated by Use of In Situ Electropolishing Techniques

A long cane-shaped, pumped-stream device was then used to decontaminate corners and small recessed areas. This device had a 1-in.-dia, disc-shaped cathode and could decontaminate $0.1 \text{ ft}^2/\text{min}$. Personnel entry was required for the use of this device.

The interior of the sparger line was decontaminated by inserting a perforated cathode, 2.5 in. in dia and 4 in. long, having a flexible seal at each end. The electrolyte was forced through the perforations in

the cathode, making the device function as a pumped-stream device. The exterior of the sparger line was decontaminated using a swab device. This device was built to fit over the pipe and was 3 in. long. It could decontaminate 1 linear foot of pipe/min.

Finally, small areas requiring additional decontamination were decontaminated using a small hand-held swab device. This device had a cathode area of 16 in.^2 and could decontaminate $0.5 \text{ in.}^2/\text{min}$.

SECTIONING STUDIES

Plasma Torch System

Hot testing of the plasma arc torch, installed in the stainless steel sectioning/pretreatment facility (Figure 12.1), has continued. This system was used successfully to cut 15 ft² of 1/4-in. TRU-contaminated stainless steel into 8-in. square sections for processing in the vibratory finisher. Additionally, the plasma arc torch was used to cut 32 ft² of 3/16-in. stainless steel from a TRU-contaminated glove box into sections for processing in the vibratory finisher and the electropolishing system. No airborne contamination above ambient levels and little surface contamination of the sectioning/pretreatment facility were observed due to the plasma arc torch cutting operation. However, several problem areas were identified that will require additional development. The smoke from the cutting process builds up in the sectioning facility, obscuring vision and rapidly plugging HEPA filters. Several approaches are under evaluation to mitigate this accumulation of smoke. A method to contain the material ejected from the cut must also be developed.

Tube Sectioning Studies

Equipment capable of rapidly cutting tubing and pipe into lengths suitable for processing in the vibratory finisher or electropolishing systems has been obtained for evaluation. This equipment uses an air-driven cutter (Figure 12.5) to section the rotating tubing with no waste and minimal probability of entrainment of surface contamination. The unit illustrated can accommodate tubing or pipe from 3/4 to

5 1/4 in. in dia and with wall thicknesses up to 3/16 in. Typical sectioning times range from 9 sec for small tubing to 7 sec for the larger pipe.

SOLUTION TREATMENT STUDIES

Hot testing of the dual-effect vacuum evaporator system has demonstrated its effectiveness in purifying aqueous rinse and pretreatment solutions. Using only a single distillation, we have successfully purified to releasable levels 250 gal of water representing a variety of plutonium-contaminated vibratory finisher, rinse, and cleaning solutions. In addition, 300 gal of water contaminated with β/γ fission products was purified to releasable levels using a single distillation. Adequate purification of these foaming solutions was achieved through control of system variables and addition of defoaming agents.

ELECTROPOLISHING TECHNOLOGY

Studies are in progress to investigate the effectiveness of barrel electropolishing techniques in reducing contamination levels and eliminating the need for racking. A small enclosure has been designed for permanent installation in the electropolishing facility to permit tests of the barrel system using plutonium-contaminated materials. Tests using uranium-contaminated materials were reported earlier.⁽¹⁾

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FIGURE 12.5. Air-Driven Pipe Cutter for Rapid Sectioning of Tubing and Pipe

13. DECONTAMINATION AND DECOMMISSIONING OF HANFORD FACILITIES - TECHNOLOGIES

H. W. Arrowsmith - Project Manager

The objective of this project is to conceive, develop and test advanced D&D technology applicable to the Hanford decommissioning effort. In addition to meeting the local need for improved D&D technology, this work will help provide a firm technological base for the D&D of retired DOE and commercial facilities at other sites.

SUMMARY

The intrinsic germanium x-ray spectrometer has been found to be the most efficient and sensitive system for surveying large areas of transuranic contaminated surfaces. Efforts are currently in progress to assemble and calibrate a completely portable field survey instrument utilizing germanium diode spectroscopy.

There are no outstanding differences among the concrete specimens of Hanford type concrete studied. The differences observed are small, and none can be attributed specifically to the history of the specimen.

To improve the concrete spaller system operation and the surface removal rate, a longer track was added and motors with control circuitry were incorporated to automatically position the drill. The improved equipment was tested at 100-F Area.

The ability of the arc saw to split pipe was demonstrated. Enough cuts have been made to demonstrate a low blade-wear trend.

A new cost/benefit task was established to develop and analyze unit cost data for the various D&D technologies.

Processing of material removed from the reference alpha facility was begun. The material was decontaminated to <10 nCi/g TRU using vibratory finishing techniques.

A 5000-gal liquid radioactive waste tank was successfully decontaminated using in situ electropolishing techniques. The tank decontamination was accomplished using only 50 gal of electrolyte, with 85% of the interior surfaces decontaminated from outside the tank.

Two fixatives were selected for field testing: a polyvinyl alcohol and a polybutyl dispersion. The polyvinyl alcohol forms a clear, tough film easily removed by vibratory finishing. The polybutyl dispersion forms a durable, strippable film.

INSTRUMENTATION DEVELOPMENT -

R. L. Brodzinski and N. A. Wogman

The objective of this research has been to develop sensitive instrumentation specifically for detecting low-level transuranic contamination on surfaces. The sensitivities, detection limits, and time and cost estimates for surveying facilities undergoing D&D efforts with a portable, omnidirectional, planar, 19 cm² intrinsic germanium diode were reported earlier based on previous work in this program.⁽¹⁾ This report summarizes the sensitivities and detection limits obtained utilizing a phoswich type detector as the survey instrument and compares these results to those of the germanium diode.

A phoswich detector system that employs a $\text{CaF}_2(\text{Eu})$ crystal, 3.45 mm thick and 12.5 cm in diameter, coupled to a 6.4 cm thick $\text{NaI}(\text{Ti})$ crystal was evaluated for its ability to measure ^{238}Pu , ^{239}Pu , ^{241}Am , and ^{244}Cm as surface contamination under a variety of overburdens. The experimental details and a full description of the instrumentation have been given elsewhere⁽²⁾ and will not be repeated here. The detection limits for plutonium and americium in soils are given in Table 13.1 for this phoswich system and for the germanium spectrometer system evaluated earlier. Also given in the table is the plutonium detection limit for another phoswich system that was designed primarily for the measurement of transuranic activities. The phoswich used in these studies was designed primarily for the detection of ^{90}Sr beta particles. As can be seen from the data, the LASL phoswich is indeed more sensitive to transuranic activities by a factor of 5. However, the intrinsic germanium spectrometer survey system developed earlier in this program is a factor of 6 more sensitive than the LASL phoswich and a factor of 25 more sensitive for the detection of plutonium than the phoswich used in these studies. The germanium spectrometer is a factor of 1700 more sensitive than the phoswich for the detection of ^{241}Am .

In conclusion, the intrinsic germanium x-ray spectrometer has been found to be the most efficient and sensitive system for surveying large areas of transuranic-contaminated surfaces. Efforts are currently in

progress to assemble and calibrate a completely portable field survey instrument utilizing germanium diode spectroscopy.

CONCRETE PROPERTIES - G. H. Beeman

Work during this fiscal year was directed at identifying a correlation between accelerated aging tests of Hanford type concrete and existing Hanford structures. The ultimate goal is the ability to predict the probable condition of structures at Hanford after a lifetime of 100 years or more.

Numerous investigations in other laboratories have shown that variations in water to cement ratio, conditions of cure, extent of mixing, and the amount of air entrainment all cause differences in the details of microstructure of concrete and cement paste. In addition, concrete microstructure normally changes slowly and continually for a period of years after initial set has occurred. However, relatively little attention has been given to studying or predicting very long-term behavior based on short-term examination. The purpose of this program was to evaluate the microstructure of several existing test and field samples of Hanford concrete and to correlate microstructural data with the physical test measurements on hand from other programs and tasks.

Since concrete is inherently a heterogeneous system, the comparison and evaluation of concretes requires a visual "averaging" of observations of sufficient samples to distinguish any subtle differences among the

TABLE 13.1. Detection Limits for Measuring Plutonium and Americium in Soils with Various Detectors. Counting Time = 1000 sec.

	$\text{CaF}_2(\text{Eu})/\text{NaI}(\text{Ti})$ Phoswich ⁽²⁾	Intrinsic Germanium Diode ⁽¹⁾	$\text{NaI}(\text{Ti})/\text{CsI}(\text{Na})$ Phoswich ⁽³⁾
^{239}Pu	5500 pCi/g	220 pCi/g	1200 pCi/g
^{241}Am	3800 pCi/g	2.2 pCi/g	

normal variations in the heterogeneous mixtures. To that end, we have extended our investigation to bring as many samples and viewpoints to bear on the conclusions as were practical within the time and funding constraints. We have utilized the diverse experience and expertise in two significantly different types of analytical laboratories and have included eleven sources of Type II Hanford concrete, each with different histories of age, stress, and temperature.

The primary conclusion reached in the study of these concrete specimens is that there are no outstanding differences. The differences that are observed are small, and none can be attributed specifically to the history of the specimen; in other words, the differences are greater within the heterogeneous nature of the concrete specimen itself than between specimens in this series. The one possible exception is the extent of microcracking occurring through the cement paste, but even here the differences are marginal and need further investigation for evaluation of significance (Figure 13.1).

All the concretes are holding up very well; there is no noticeable effect of time or stress in any case. The reinforcing bar appears stable and almost entirely unreactive. The only rebar reactions are apparently oxidation (probably prior to fabrication of the concrete structure) and a thin, stable enriched calcium reaction layer, probably formed on initial contact with the calcium hydroxide or carbonate in the cement paste.

No specimens were available of the rebar in existing building structures, e.g., in Purex or 221T. Therefore, the extent and rate of reaction between cement paste and the rebar surface, if any exists, could not be determined as a factor of age. This particular question bears further investigation.

CONCRETE DECONTAMINATION - J. M. Halter and R. G. Sullivan

This task's objective is development of equipment and techniques for removal or decontamination of contaminated concrete. Removal methods currently under investigation include a rock splitter, a water cannon, and use of a very high pressure water jet. This task also will maintain awareness of developments in dry ice blasting technology that have possible applications in D&D operations.

A major problem associated with nuclear facility decontamination and decommissioning is how to economically demolish and dispose of contaminated concrete. To help solve this problem, equipment is being developed at PNL to remove contaminated concrete surfaces. It is being designed to reduce the quantity of contaminated rubble that must be specially handled and buried, and to provide a convenient method for cleaning contaminated areas.

The concrete spaller is the primary removal method under investigation. The spaller is a tool that exerts radial pressure with expanding split wedges on the sides of a shallow cylindrical hole drilled into the concrete surface, causing the surface to spall. Powered by a hydraulic cylinder, the spaller tool is capable of removing an 8-in.-dia section of concrete surface each time it is operated.

The spaller and an air-powered drill are mounted on a portable platform, which can be positioned by a fork lift along the surface to be removed. The spaller and platform system configuration are shown in Figure 13.2. When the system was tested in November 1978, it performed very well, removing concrete surface at about 100 ft²/hr.

To improve the system operation and surface removal rate, a longer track was added to increase the drill operating range, and

B18230

200X

B18231

300X

B18232

3,000X

B18233

10,000X

FIGURE 13.1. Purex Building Samples Showing Radial Microcracking Through Air Bubbles and Pockets of Needle-like Ettringite Structures in the Cement Paste Area

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FIGURE 13.2. Concrete Spaller and Air-Powered Drill Mounted on Portable Platform

motors with control circuitry were incorporated to automatically position the drill. The improved equipment was again tested at 100 F Area on concrete typical of that found in contaminated facilities. However, the removal rate decreased from the previous test to about 60 ft² surface removed per hour because of unexpected equipment problems. One problem was that the control system could not sense contact with reinforcing rods, and the drilling continued without further progress. Another problem was that the vibration from the drill caused the limit switches to give false position readings to the control system, halting operation. To resume operation after each stoppage, the operator had to intervene with manual override, thus reducing the surface removal rate.

The concrete spaller was used in September to decontaminate a section of floor in the 303-C Building. In the spaller's first evaluation in a plutonium-contaminated environment, a floor surface was removed quickly without spread of contamination.

The use of very high pressure water streams was investigated as a possible technique for removing the surface layer of contaminated concrete. Flow Industries of Kent, Washington, which manufactures high-pressure equipment, demonstrated that the surface of a piece of typical Hanford concrete could be removed to a depth of 1/4 to 3/8 in. at a rate of 6 ft²/min. With a modest development program, it is felt that this technique could be adapted into a machine that could remove the surface of walls, floors and ceilings of a facility and, with an air mover system incorporated next to the nozzles, collect into tanks the rubble that is generated.

One gallon of water is used to remove about 2 ft² of surface. It is thought

that recontamination of the previously cleaned surface and driving the contamination further into the surface are not a problem. However, removing the water from the contaminated rubble may be difficult to do economically.

ARC SAW EVALUATION - G. S. Allison

The objective of this task is to evaluate and demonstrate application of the arc saw to D&D activities.

The arc saw's ability to split pipe has been amply demonstrated. A sufficient number of cuts have been made to demonstrate also a reasonably low blade wear trend. Additional work is required, however, to establish the optimum power parameters and to develop the remote handling techniques required to feed an actual installation. Figure 13.3, a schematic of the PNL arc saw, indicates the size of an arc saw installation for splitting contaminated piping for decontamination.

A problem has been encountered in splitting both sides of mild steel pipe simultaneously when the saw is in the splitting mode. As the blade enters the lower side, the arc becomes unstable and shuts off the power supply, apparently with the shorting protection trip. This does not occur with stainless pipe, which has been successfully split by cutting both sides simultaneously. One possible cause for this problem is an adverse effect of the magnetic loop of the mild steel pipe on the arc.

COST/BENEFIT - L. D. Perrigo

The objective of this task is to acquire, assemble, analyze and evaluate unit cost data for the various D&D technologies.

A new cost/benefit task was established to accomplish this objective.

FIGURE 13.3. PNL Arc Saw, Indicating the Size of Installation for Splitting Contaminated Piping

ELECTROPOLISHING/VIBRATORY FINISHING APPLICATIONS - H. W. Arrowsmith and M. W. McCoy

The objective of this task is to demonstrate the applicability of the highly successful electropolishing and vibratory finishing decontamination processes to field D&D operations.

Processing of material removed from the reference alpha facility began. Several 55-gal drums of material were successfully decontaminated to <10 nCi/g TRU.

The first drums of material received from the reference alpha facility for processing contained stainless steel frame members and Plexiglas® panels from a large process hood, predominantly Plexiglas. Various fixatives had been applied to the material before disassembly in the facility. The material was processed in the vibratory finisher using a steel medium and a dilute, 10%, sodium hydroxide solution as the compound. The medium used was 1/4-in. ballcones. This medium's unique shape allows decontamination of a wide variety of shapes and sizes of material, thus providing a field unit of sufficient versatility to eliminate most presorting.

Work is progressing on development of in situ electropolishing devices. Several devices were built and successfully hot tested on a 5000-gal liquid radioactive waste tank contaminated with corrosion products, chiefly ^{60}Co . Radiation levels were reduced from 20 mrem/hr to background with no detectable smearable contamination. This was achieved using only 55 gal of electrolyte. Moreover, 85% of the interior tank surface was decontaminated from outside the tank.

Three electrolyte-supplied swabs and two pumped-stream devices were used for this decontamination. A small hand-held swab device was used for spot decontamination. This device has a cathode area of 16 in.^2 and could decontaminate $0.5\text{ ft}^2/\text{min}$ at a current density of 5 A/in.^2 . A large swab device (Figure 13.4) with a magnetic coupler capable of remote decontamination was used to remotely decontaminate 85% of the internal tank surface. This device had a cathode area of 60 in.^2 and could decontaminate $2\text{ ft}^2/\text{min}$ at a current density of 4 A/in.^2 . The final swab device was built to decontaminate the external surfaces of a 4-in.-dia, perforated sparger line 24-ft long, which ran through the waste tank. The cathode was shaped to fit over the pipe and was 3 in. long. This device could decontaminate 1 linear ft of pipe/min at a current density of 5 A/in.^2 .

The first pumped-stream device was hand-held and was used to decontaminate corners and other inaccessible surfaces. The cathode was a 1-in.-dia disk. This device could decontaminate $0.1\text{ ft}^2/\text{min}$ at a current density of 1.5 A/in.^2 . The final device was also of the pumped-stream type. It was used to decontaminate the internal surfaces of the 24-ft-long tank sparger line. The cathode was a cylinder 2.5 in. in diameter by 4 in. long with flexible seals which allowed the device to negotiate complex pipe configurations. This device can decontaminate 16 linear in. of pipe/min at a current density of 3 A/in.^2 .

One objective of this year's work was to compare processing and facility costs, materials balance factors and safety considerations for central, modular and portable TRU electropolishing decontamination facilities. Based on the excellent results obtained with vibratory finishing, it now appears that vibratory finishing is more suitable than

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FIGURE 13.4. Swab Type In Situ Electropolishing Device with Remote Operation Capabilities

electropolishing for field decontamination operations. A central electropolishing facility would provide the capability for additional decontamination if required.

Our studies and experience indicate that approximately 40% of the equipment and other nonstructural material removed from typical TRU facilities undergoing D&D is nonmetallic (plastic, rubber, gloves). Additionally, the material is painted and has accumulations of grease and dirt. Vibratory finishing can decontaminate all this material as well as remove the paint, grease and dirt. It produces a decontaminated product <10 nCi/g with little or no smearable contamination and in an ideal condition for electropolishing should additional decontamination be desired.

A conceptual design of a vibratory finisher decontamination system for field D&D operations will be completed in FY-1980.

FIXATIVES - M. W. McCoy and H. W. Arrowsmith

The objective of this task is to identify, develop and demonstrate improved contamination-stabilizing coatings for use in D&D operations.

Studies were begun to evaluate the effectiveness of various substances in fixing smearable surface contamination to facilitate the handling of contaminated material. Initially, five substances were tested for ease of application, drying time, durability, method of removal, and compatibility with selected decontaminants. Other characteristics evaluated included flammability, odor, and safety hazards.

Two of the five substances tested were selected for field tests: a polyvinyl alcohol and a polybutyl dispersion, both commercially available. The polyvinyl alcohol forms a tough, clear, very durable film.

A light film, 1 mill, is sufficient to fix contamination but can be removed with a caustic solution and is readily removed by the vibratory finisher. The polyvinyl alcohol, therefore, is ideally suited for application on material to be decontaminated. The polybutyl dispersion forms a resilient, durable, translucent film. A light film, 1 mill, will fix contamination. Films of 1 mill or more can be stripped intact, making the polybutyl dispersion, therefore, best suited for application on fixed items such as structures that will not be removed for further decontamination.

A pressure sprayer (Figure 13.5) was developed, capable of quickly applying either fixative over large areas within minimal disturbance to the contamination. The sprayer consists of a pressurized reservoir, a connecting hose, and a wand equipped with a specially fabricated spray nozzle. Only the wand and hose enter the contaminated area.

These fixatives were successfully demonstrated during operations in a transuranic-contaminated facility. The facility had plutonium contamination levels of 4×10^6 dis/min 100 cm² on horizontal surfaces. Entries made before the use of the fixatives resulted in a contamination level of 40,000 dis/min 100 cm² on personnel's protective suits. Similar entries made after applying the polyvinyl alcohol to the work area resulted in no detectable contamination on personnel's protective suits.

During a subsequent entry into this same facility, the entire floor area was sprayed with the polybutyl dispersion. Entries made after this application resulted in contamination levels of only 600 dis/min 100 cm² to personnel's shoe covers.

Tests showed that the strippable fixative, the polybutyl dispersion, also possesses decontamination properties. Strippable coatings are especially effective for removing loose surface contamination. The use of strippable fixatives for decontamination was demonstrated in a TRU-contaminated facility where one application and removal of the fixative reduced contamination levels from 4×10^6 dis/min 100 cm² to 10,000 dis/min 100 cm².

TECHNOLOGY ASSESSMENT - R. P. Allen and H. W. Arrowsmith

The objectives of this task are: 1) to assess the present state of decontamination technology, 2) to identify critical technology development needs, and 3) to maintain a strong interface with other Hanford contractors engaged in D&D activities.

Discussions were held with Rockwell Hanford Operations' staff to set priorities for the decontamination of the material removed from 233S Building during their D&D demonstration project. The disposal of liquid wastes from that project was also discussed.

FIGURE 13.5. Spray Apparatus for Application of Fixatives to Large Areas

Discussions were held with personnel from the Mound Laboratory concerning the D&D project at that facility. Specific topics included concrete decontamination, fixatives and decontamination of large tanks in situ. Discussions were also held with personnel from Idaho National Engineering Laboratory and Los Alamos Scientific Laboratory on D&D projects.

A seminar conducted by Health Physics Systems was attended by members of the staff. The use of freon for decontamination of clothing, respirators, small tools, and electric motors was discussed.

A consulting agreement was established with Roto Finish, a leading manufacturer of vibratory finishing equipment.

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Cycle and Materials
Office of Nuclear Safety Materials
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Room 562
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Assistant Director for Radioactive
Waste Management Branch
NRC Division of Materials and
Fuel Cycle Facility Licensing
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Washington, DC 20545

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Washington, DC 20545

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Washington, DC 20545

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Washington, DC 20545

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Washington, DC 20545

R. D. Walton
DOE Office of Nuclear Waste Management
NEW, B-107, HQ
Washington, DC 20545

J. Neff, Program Manager
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Rockwell International
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A light film, 1 mill, is sufficient to fix contamination but can be removed with a caustic solution and is readily removed by the vibratory finisher. The polyvinyl alcohol, therefore, is ideally suited for application on material to be decontaminated. The polybutyl dispersion forms a resilient, durable, translucent film. A light film, 1 mill, will fix contamination. Films of 1 mill or more can be stripped intact, making the polybutyl dispersion, therefore, best suited for application on fixed items such as structures that will not be removed for further decontamination.

A pressure sprayer (Figure 13.5) was developed, capable of quickly applying either fixative over large areas within minimal disturbance to the contamination. The sprayer consists of a pressurized reservoir, a connecting hose, and a wand equipped with a specially fabricated spray nozzle. Only the wand and hose enter the contaminated area.

These fixatives were successfully demonstrated during operations in a transuranic-contaminated facility. The facility had plutonium contamination levels of 4×10^6 dis/min 100 cm^2 on horizontal surfaces. Entries made before the use of the fixatives resulted in a contamination level of 40,000 dis/min 100 cm^2 on personnel's protective suits. Similar entries made after applying the polyvinyl alcohol to the work area resulted in no detectable contamination on personnel's protective suits.

During a subsequent entry into this same facility, the entire floor area was sprayed with the polybutyl dispersion. Entries made after this application resulted in contamination levels of only 600 dis/min 100 cm^2 to personnel's shoe covers.

Tests showed that the strippable fixative, the polybutyl dispersion, also possesses decontamination properties. Strippable coatings are especially effective for removing loose surface contamination. The use of strippable fixatives for decontamination was demonstrated in a TRU-contaminated facility where one application and removal of the fixative reduced contamination levels from 4×10^6 dis/min 100 cm^2 to 10,000 dis/min 100 cm^2 .

TECHNOLOGY ASSESSMENT - R. P. Allen and H. W. Arrowsmith

The objectives of this task are: 1) to assess the present state of decontamination technology, 2) to identify critical technology development needs, and 3) to maintain a strong interface with other Hanford contractors engaged in D&D activities.

Discussions were held with Rockwell Hanford Operations' staff to set priorities for the decontamination of the material removed from 233S Building during their D&D demonstration project. The disposal of liquid wastes from that project was also discussed.

FIGURE 13.5. Spray Apparatus for Application of Fixatives to Large Areas