**Nuclear Waste Management
Quarterly Progress Report
October Through December 1978**

A. M. Platt
J. A. Powell

March 1979

Prepared for the U.S. Department of Energy
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NUCLEAR WASTE MANAGEMENT
QUARTERLY PROGRESS REPORT
OCTOBER THROUGH DECEMBER 1978

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

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Pacific Northwest Laboratory
Richland, Washington 99352

FOREWORD

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

The prior report in this series is Nuclear Waste Management Quarterly Progress Report July Through September 1978, PNL-2378-3. The next quarterly progress report will have a new series number, PNL-3000.

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1.0 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 groundwater 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 has been assembled in a building operated by the Pacific Northwest Laboratory. Pilot-scale chemical decontamination equipment and an Inductoslag ingot melting furnace are set up on a simulated hot cell floor plan for eventual combined nonradioactive demonstration. Autoclaved, unirradiated scrap cladding pieces will be descaled in the decontamination prototype with HF/Ar followed by an aqueous rinse, and then melted in the induction furnace to produce ingots.

All instrumentation for the chemical decontamination prototype has been calibrated, and standard operating procedures were prepared. Operating tests of the prototype demonstrated successful operation and control of all decontamination equipment subsystems. Problems with the isolation valves between the HF/Ar gas reactor and the aqueous washer/rinser have led to a reconsideration of the valve design.

Corrosion tests are being prepared to determine the corrosion resistance of various zirconium and stainless steel alloy samples taken from ingots produced in previous Inductoslag furnace melting tests.

Reports are being prepared on the development of the chemical decontamination process and the operation of the Inductoslag furnace.

DECONTAMINATION PROCESS

The pilot-scale chemical decontamination equipment shown in Figure 1.1 is designed to "decontaminate" (descal) ~2 kg of chop-leach cladding residues per hour. Operating tests of the prototype equipment have been made to check out concurrent operation of all subsystems, which include an HF gas reactor, the HF reactor furnace, HF and argon gas supply systems, an aqueous washer/rinser, associated process solution tanks and piping, and control instrumentation and safety circuits. The basic components of the system have been described and diagrammed in previous reports.^(1,2) For these operating tests

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FIGURE 1.1. Pilot-Scale Chemical Decontamination Equipment and the Inductoslag Furnace

argon was substituted for the HF/Ar gas mixture, and deionized water was used instead of the aqueous wash solution. Nonradioactive Zircaloy-4 scrap cladding was used for feedstock. These tests demonstrated that the subsystems were controllable within parameters but did not verify the chemical processing aspect of operation.

During the fourth operating test, the gate valves that isolate the reactor from the washer/rinser began leaking. Inspection

revealed that cladding pieces had become trapped in the valves, damaging several valve components. Alternative valve designs are being considered while the damaged valves are being repaired.

Calibration of all instrumentation for the decontamination prototype was completed. Figure 1.2 shows the control panel for the decontamination equipment.

Standard operating procedures were developed to provide a checklist for

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FIGURE 1.2. Control Panel of the Chemical Decontamination Equipment

decontamination equipment startup, operation, and shutdown. A breakdown of potential operating hazards was prepared to inform operators of potential safety problems. A full report is now being prepared on the development of the chemical decontamination process.

DENSIFICATION PROCESS

The Inductoslag ingot melting furnace shown in Figure 1.1 is the densification portion of the decontamination-densification equipment set up for processing chop-leach cladding residues. The furnace is located on a hot cell floor plan with the prototype chemical decontamination equipment, where the two systems will eventually be operated in nonradioactive demonstrations of the combined decontamination-densification process.

REFERENCES

1. A. M. Platt, compiler, Nuclear Waste Management Quarterly Progress Report, January Through March 1978. PNL-2378-1, Pacific Northwest Laboratory, Richland, WA, August 1978.
2. A. M. Platt, compiler, Nuclear Waste Management Quarterly Progress Report,

To date, 21 melts have been made in the Inductoslag furnace. Various alloys of Zircaloy, stainless steel, and Inconel have been cast as described in previous reports.⁽¹⁻³⁾ The furnace has been inactive since these testing heats, awaiting final readiness of the chemical decontamination equipment. A report is now in preparation on the development and operation of the Inductoslag furnace.

In the absence of melting activities, corrosion tests of ingots produced in previous Inductoslag melting heats are being prepared. A modified water bath will be used to determine the corrosion resistance of various zirconium alloy samples taken from the alloyed ingots. The corrosion resistance of the samples is related to the potential leachability of the ingots.

- April Through June 1978. PNL-2378-2, Pacific Northwest Laboratory, Richland, WA, November 1978.
3. A. M. Platt, compiler, Nuclear Waste Management Quarterly Progress Report, July Through September 1978. PNL-2378-3, Pacific Northwest Laboratory, Richland, WA, January 1979.

2.0 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

Quality assurance activities relating to both sampling and laboratory analytical techniques have been pursued during the first quarter of FY 1979. In addition, an alternative sampling location has been established for gaseous radionuclides that may be released in high concentration.

PRE-SAMPLE TESTING

The main objective of this monitoring program is to characterize the gaseous and airborne effluent associated with only one phase of the Nuclear Waste Vittrification Program (NWVP), specifically that of vitrification. However, previtrification activities associated with the generation of high-level liquid waste (HLLW) have provided the opportunity to check out and test the sampling network that will be used during the two future vitrification campaigns. Both particulate and gaseous samples were taken during the initial NWVP spike runs as well as during

actual fuel dissolution campaigns.

Selected samples were taken back to the laboratory for analysis. The analytical work not only served to check out laboratory analytical systems but also formed the basis for estimating emission rates of certain radionuclides during the vitrification process.

The analytical results obtained during pre-sample testing at the stack may be more convenient than in the undiluted process stream. As a result, sampling capabilities at the stack were established to allow for this contingency.

FLOW CALIBRATIONS

Velocity probes located in the vitrification off-gas process line and in the main exhaust stack have been calibrated in flow units. This was done using an on-line mass spectrometer and helium dilution techniques. Since this calibration has been made, any effluent species measured at the stack can be related directly back to its emission rate in the off-gas process line.

3.0 TRU WASTE IMMOBILIZATION PROGRAM

C. R. Palmer - Project Manager

The objectives of this program are to adapt high-level waste immobilization processes to selected low-level transuranic wastes, and to demonstrate that suitable waste forms can be produced routinely for these wastes.

SUMMARY

Laboratory waste form development studies continued during the reporting period. Borosilicate glasses containing up to 70 wt% of various simulated transuranic wastes were fabricated and characterized. Two waste types were selected for initial pilot-scale process tests, and final glasses for these wastes are being developed. Other preparations for the process tests (to be conducted during the fourth quarter of FY-79) are under way.

Sintered ceramic waste forms produced during FY-78 are being characterized. A critical assessment of potential advantages of such a waste form will be reported during the second quarter.

GLASS DEVELOPMENT - C. W. Griffin,
G. B. Mellinger, and L. A. Chick

Laboratory investigation of borosilicate glasses containing simulated low-level transuranic wastes has continued. Initially, 11 compositions (5301-5311 shown in Table 3.1), which incorporated seven different waste types, were examined. The glasses which met visual examination criteria (i.e., homogeneity, crystallinity and porosity) were then characterized with respect to leach resistance (pH 4 and pH 9

water at room temperature, and distilled water at 99 C), viscosity, and electrical resistivity. Following evaluation of these compositions, two waste types and associated glasses were chosen for concentrated study so that representative process tests can begin. One of the selected wastes (designated type 5108) is an incinerator residue which has been acid-leached to recover plutonium, while the other waste (type 5101) represents a mixture of residues typical of a nuclear fuel reprocessing complex. The objective of the continuing experiments is to develop borosilicate glasses which exhibit good physical and chemical properties while maintaining high waste loadings and which remain compatible with existing (high-level) waste vitrification systems. Process compatibility will be examined during the process tests to be conducted later in the fiscal year.

As Table 3.1 implies, the high alumina compositions do not form good glasses at processing temperatures (1050 C), but glasses with lower alumina concentrations were easily melted and were generally homogeneous. While for some waste types the alumina concentration can be acceptably controlled by adjusting the waste loading slightly, the alumina concentration in waste 5101 is sufficiently high to make this approach undesirable. Therefore we are

TABLE 3.1. Chemical Composition of Initial Laboratory Glasses,^(a) mole%

Glass Ident.	5301	5302	5303	5304	5305	5306	5307	5308	5309	5310	5311	5312	5323	5325
Compound														
SiO ₂	46.2	48.8	42.9	56.8	57.4	43.6	56.9	48.9	49.6	52.9	60.8	47.6	45.6	45.9
B ₂ O ₃	9.5	9.7	9.2	8.9	9.2	9.0	9.1	7.2	11.5	11.8	6.9	16.7	9.6	16.0
P ₂ O ₅	0.3	-	-	-	0.5	-	-	-	-	0.5	-	0.7	0.3	0.9
Al ₂ O ₃	12.0	13.6	11.2	5.3	2.5	4.3	5.8	11.2	5.4	2.5	5.8	2.7	9.3	6.0
SO ₃	1.0	1.4	-	0.4	-	-	-	-	-	-	-	-	-	-
K ₂ O	-	-	-	0.3	0.2	-	0.6	-	0.3	0.2	0.6	0.2	-	0.2
Na ₂ O	14.0	14.3	13.7	13.2	13.6	13.3	13.4	10.2	17.5	13.6	13.4	14.7	13.2	14.1
Li ₂ O	-	-	-	-	-	-	-	-	-	-	-	-	6.5	-
MgO	4.3	-	7.1	-	2.0	4.4	3.2	7.1	-	2.0	3.1	2.0	3.4	2.1
CaO	2.4	2.5	4.8	4.5	3.7	12.5	2.4	3.9	5.7	3.7	2.4	4.0	2.5	3.8
BaO	-	-	-	-	0.1	0.2	-	-	-	0.1	-	0.1	-	0.1
TiO ₂	2.6	2.7	2.5	6.6	2.6	6.6	2.5	2.6	6.6	3.9	1.4	2.6	2.7	2.4
Cr ₂ O ₃	1.8	2.6	0.6	-	0.7	-	-	0.6	-	0.7	-	0.7	1.4	0.7
Fe ₂ O ₃	0.1	-	1.1	-	2.4	2.0	2.0	1.1	-	2.4	2.0	2.7	0.1	2.4
MnO ₂	-	-	-	-	0.1	-	-	-	-	0.1	-	0.1	-	0.1
NiO	-	-	2.6	-	0.4	-	-	2.6	-	0.4	-	0.7	-	0.5
CuO	1.5	-	0.1	-	0.2	-	-	-	-	0.2	-	0.2	1.2	0.3
ZnO	4.3	4.4	4.2	4.0	4.1	4.1	4.1	4.6	3.4	4.7	3.6	4.0	4.2	4.2
Nd ₂ O ₃	-	-	-	-	0.2 ^(b)	-	-	-	-	0.2 ^(b)	-	0.2 ^(b)	-	0.2 ^(b)
PbO	-	-	-	-	0.1	-	-	-	-	0.1	-	0.1	-	0.2
Total	100.0													
Waste load ^(g) , wt%	64	45	52	38	63	43	66	38	53	67	63	66	52	63
Visual code(c)	C	B	B	A	A	A	A	C	A	A	A	A	B	A

a. Calculated based upon batch make-up.

b. Stand-in for PuO₂.

c. A - homogeneous, low porosity

B - crystalline, heterogeneous

C - poorly melted, high porosity.

investigating alternative approaches including the use of other network modifiers to reduce the viscosity. Currently glasses containing small quantities of lithia are being studied, of which glass 5323 is an example. It has been observed that addition of more than 5 mole% Li₂O produces glasses of acceptable viscosities, but these glasses are quite heterogeneous and exhibit poor acid leach resistance. Work will continue in an

attempt to improve upon glass 5323.

Table 3.2 includes summary characteristics for this glass.

The two other glasses shown in Table 3.2 generally meet our laboratory development objectives and incorporate waste 5108 at greater than 60 wt%. Glass 5312 exhibits a lower resistance to leaching under the extreme conditions of the soxhlet test, and minor modifications are being studied to

TABLE 3.2. Characteristics of Selected Laboratory Glasses

Glass No.	5312	5323	5325
Waste Type	<u>5108</u>	<u>5101</u>	<u>5108</u>
Leach resistance, g/cm ² -day			
pH 4, 25°C	4.9 x 10 ⁻⁶	1.2 x 10 ⁻³	3.2 x 10 ⁻⁴
pH 9, 25°C	4.9 x 10 ⁻⁶	3.4 x 10 ⁻⁵	1.9 x 10 ⁻⁵
Soxhlet	2.4 x 10 ⁻⁴	8.8 x 10 ⁻⁵	8.8 x 10 ⁻⁵
Viscosity, Pa·s			
at 950°C	37	64	60
1050°C	11	25	19
1150°C	5	13	8
Electrical resistivity, ohm·m			
at 950°C	0.071	0.056	0.111
1050°C	0.036	0.039	0.063
1150°C	0.022	0.024	0.040

improve performance. Glass 5325 shows a two-fold improvement in the soxhlet leach rate, but the acid and base durabilities have been compromised somewhat. Glass 5312 or its successor will be tested with samples of bulk incinerator ash being procured from Rockwell International at Rocky Flats Plant (~50 kg of ash representative of waste 5108). Any modifications to the glass composition dictated by those tests will be made, and the bulk ash will be vitrified in pilot plant melting equipment. The process demonstration will be designed to determine the extent of potential solids handling and corrosion problems, as well as to provide large-scale product for detailed characterization.

CRYSTALLINE WASTE FORMS - J. M. Rusin

The primary objective of this task during FY-79 is to characterize about 80 sintered ceramic pellets fabricated last year using simulated transuranic waste compositions (a typical incinerator ash, 5102, and the composite waste, 5101). During the reporting period the pellets were visually characterized, and densities were determined. In the second quarter the leach resistance and impact behavior will be characterized. The characterization data and an assessment of the incentives for pursuing development of this waste form will be reported in April 1979.

4.0 KRYPTON SOLIDIFICATION

Garth 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

Krypton has been trapped in a radio-frequency, sputter-deposited sample of SiO_2 . The substrate was rf-biased to 300 V dc, yielding a product containing about 13 mole% Kr. Krypton was released from this sample in the range of 300° to 900°C, with a small quantity of krypton (~1 mole%) remaining after treatment at 1200°C.

ENTRAPMENT OF KRYPTON IN SiO_2 by rf SPUTTERING - G. L. Tingey, E. D. McClanahan, and M. A. Bayne

In this program we have attempted to load several types of glass with relatively large quantities of krypton as a storage technique for ^{85}Kr . It became evident that a radio-frequency sputtering technique had the potential to yield a glass sample with significantly higher krypton content than appeared possible by our dissolution technique. Our interest in this approach was not only to develop another process for loading SiO_2 with ^{85}Kr , but rather to study the properties of a highly loaded glass sample. This approach was encouraged by the work of Wang,⁽¹⁾ who published the results of a study in which he reported sputtering a sample of SiO_2 containing 16.1 at.% Kr as determined by x-ray fluo-

rescence analysis. Accordingly, we have sputtered from a fused silica target bonded to a water-cooled copper backing plate. The substrate was a water-cooled polished copper plate. The target power was 1200 W forward power with a 2900-V dc bias. An rf bias was placed on the substrate and increased slowly up to 300 V and 120 W, at which point krypton uptake was evident. During the overnight run, this bias voltage decreased to 180 V, which undoubtedly decreased the krypton content of the deposit.

The krypton uptake was measured with a Hastings Model ALK-5 mass flowmeter. Since the low flows for this run were near the sensitivity limits of the meter, the measurement was only indicative of the krypton content and cannot be considered quantitative. The sputtering run was continued for about 19 hr, during which time a deposit weighing 1.6 g was generated.

Scanning electron microscopy (SEM) of the deposit shows a dense, featureless material smoother on the free surface than on the side bonded to the substrate. This indicates a smoothing effect probably caused by the substrate bias. The fracture surface was also examined and was characteristic of a glassy (amorphous) deposit. X-ray diffraction measurements indicated that the deposit is amorphous, with a peak half width

of 9.9° (2θ), or a grain size little larger than the SiO_2 molecular size. There was also a shift in the 2θ angle, with the krypton-containing glass at 26.59° compared to 21.1° for a sample containing little or no krypton.

X-ray fluorescence measurements were made, yielding an uncorrected $\text{Kr}/(\text{Kr} + \text{Si})$ peak height ratio of 0.75. This is compared with a ratio of 0.11 for a sample deposited on an unbiased shield adjacent to the substrate.

Krypton release measurements were then made on the samples. The samples were heated, increasing the temperature at a linear rate to 1200°C . Most of the krypton was released in the temperature range between 300° and 900°C , with some residual release occurring even at 1200°C . The total krypton content from the release measure-

ments was 47 cm^3 of Kr (STP)/g of sample, or about 13 mole% Kr.

Using this loading, we have determined that the x-ray fluorescence sensitivity ratio of krypton to silicon is 18.7. Thus the concentration of krypton in the unbiased glass deposited on the shield calculates to be 0.7 mole% Kr. Furthermore, after heating to 1300°C , a krypton-loaded SiO_2 sample still showed an x-ray fluorescence peak with a $\text{Kr}/(\text{Kr} + \text{Si})$ peak height ratio of 0.16, or a krypton content of about 1 mole%.

Apparently a small fraction of the krypton is not released at a significant rate even at 1200°C . A more thorough evaluation is planned for this krypton-loaded, sputter-deposited glass, and another sputtering run will be completed in which the krypton uptake rate will be maximized.

REFERENCES

1. Rong Wang, "A Vitreous Krypton Disilica," Nature, 270:705 (1977).

5.0 CARBON-14 AND IODINE-129 FIXATION

L. L. Burger - Project Manager

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

SUMMARY

Tests were made on the absorption of methyl iodide from an air stream by silver mordenite and by reduced silver mordenite. Both were effective, with the latter showing much greater loading in preliminary observations.

The absorption of hydrogen iodide (HI) in aqueous solutions was briefly studied. Further tests will be conducted to examine the efficiency of scrubbing of dilute HI in hydrogen.

IODINE LOADING - K. D. Wiemers

Characterization of metal-exchanged zeolite iodine loading properties was continued. The objective of this quarter's studies was to introduce iodine in the form of methyl iodide and compare the instantaneous decontamination factor as a function of loading for silver mordenite, Agm, versus reduced silver mordenite, Ag^om.

The loading apparatus consisted of an air supply, needle valves, flowmeters, a manifold, a methyl iodide generator, a thermoregulated reaction vessel, and influent and effluent sampling ports. Methyl iodide was introduced to the manifold from a generator consisting of a bubbler containing methyl iodide submerged in a temperature-controlled bath set at -20°C. The influent and effluent gas streams were

collected using gas sampling bulbs and analyzed by gas chromatographic techniques.

Two runs were made using experimental conditions listed in Table 5.1. Run 1 involved loading of the untreated silver mordenite. In Run 2 the silver mordenite was purged with nitrogen at 300°C followed by 6 hr of 100% hydrogen at 500°C. The hydrogen pretreatment was followed by 45 hr of pre-equilibration with air at 150°C.

The iodine loading characteristics for these two runs are expressed in terms of the instantaneous decontamination factor as a function of loading time (Figures 5.1 and 5.2). The results are summarized in Table 5.2. They indicate that the maximum loading at an instantaneous decontamination factor of 10^3 was 65 times greater for Ag^om than for Agm.

Analysis of the effluent by gas chromatography retention time correlations and gas

TABLE 5.1. Experimental Conditions^(a)

Dry bed wt, g	45.0
Bed diameter, cm	2.5
Bed depth, cm	10.0
Bed temperature, °C	150
Face velocity, m/min	5.1
Average CH ₃ I Influent concentration, g/m ³	1.0

a. Initial starting material for both runs was silver-exchanged (18.2%) mordenite, 1/16 in. extrudate.

FIGURE 5.1. CH_3I Loading onto Agm FIGURE 5.2. CH_3I Loading onto Ag°m

TABLE 5.2. Loading Characteristics

Material	Decontamination Factor	Maximum Loading Capacity, mg I/g Agm
Agm	10^3	0.7
	10^2	45
Ag°m	10^3	45
	10^2	70-80

chromatography-mass spectrometry indicated the presence of dimethyl ether. I_2 was qualitatively detected in the effluent of Run 2 at a loading of between 60 and 80 mg I/g Agm. Irregularities in the DF curves correspond to shut down/start up operations. These are being eliminated with an automated system design.

Future investigations will include further examination of methyl iodide loading mechanisms and measurement of the effectiveness of the reduced form in the presence of influent contaminant gases and in a recycling system.

TRAPPING OF HYDROGEN IODIDE - R. D. Scheele

In the recycle of silver mordenite a hydrogen stream containing about 1.5 vppt (parts per thousand by volume) hydrogen iodide will be generated. An alternative to retrapping the HI in another zeolite is to collect it in an aqueous system for subsequent processing for storage.

To determine relative trapping efficiencies, water and aqueous solutions or slurries of sodium hydroxide, cupric nitrate, lead nitrate, ceric ammonium nitrate-nitric acid, bismuth nitrate-nitric acid, a bismuth (III) oxide-sodium hydroxide slurry and a lead

TABLE 5.3. Relative Trapping Efficiencies for ~1.5 vppt HI-N₂

Scrubbing Solution	Decontamination Factor	Loading [I], M
H ₂ O	2×10^4	0.1
1M NaOH	5×10^4	0.2
0.5M Cu(NO ₃) ₂ + 0.2M HAN	2×10^2	0.2 ^(a)
0.5M Pb(NO ₃) ₂	2×10^3	0.4 ^(a)
0.5M (NH ₄) ₂ Ce(NO ₃) ₆ - 1M HNO ₃	3×10^0	0.07 ^(a)
0.1M Bi(NO ₃) ₃ - 0.5M HNO ₃	1×10^3	0.2 ^(a)
Bi ₂ O ₃ - 1M NaOH	2×10^2	0.08 ^(a)
PbO - 1M NaOH	2×10^1	0.07 ^(a)

a. Calculated based on flow rate measured during 1M NaOH experiment.

oxide-sodium hydroxide slurry have been examined. The results of this study are presented in Table 5.3. Decontamination factors greater than 10^3 for iodine loadings up to 0.2M iodide were realized with 1M sodium hydroxide, water, 0.5M lead nitrate, and 0.1M bismuth-nitrate. Solids formed in the latter two solutions.

Hydrogen iodide-nitrogen (1.5 vppt) at a flow of 1 L/min was passed through three 100-ml gas scrubbing bottles employing coarse cylindrical glass frits for gas dispersion. The bottles were filled with 75 ml of solution/slurry. The primary trap contained the solution/slurry of interest while the secondary and tertiary traps contained 1M sodium hydroxide.

An iodide-specific ion electrode was used for measuring iodide. Total iodide for all aqueous solutions/slurries was calculated from the flow rate and the elapsed time.

The partial pressure of HI above dilute water solutions is extremely low, about 0.1 torr at 10M ($X \sim 0.15$) and 6×10^{-4} torr at 6M ($X \sim 0.10$).⁽¹⁾ This can be extrapolated to about 2×10^{-5} torr at a concentration of about 1M ($\sim 1M$, $X = 0.02$). Thus, at the observed loadings of up to 0.2M HI, the observed decontamination factors are determined by gas scrubbing efficiencies rather than by equilibria.

Further studies of aqueous scrubbing will be carried out.

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6.0 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

Work neared completion on summarizing the geologic and seismic data base and mapping programs. Work continued also on summarizing the geologic investigation of the 270-km-long photolineament in Washington and Oregon.

Costs of managing high-level and transuranic wastes from the LWR fuel cycle have been estimated in preparation for the January meeting of the International Nuclear Fuel Cycle Evaluation Working Group 7 in France. An analysis has been made of the potential radiological consequences of the accidental breach of a hypothetical waste repository, also preparatory to that meeting.

An improved framework has been developed for the systematic analysis of waste management issues by an interdisciplinary (technical and nontechnical) team.

Work was begun also on developing an algorithm to optimize the depth of a geologic repository in terms of cost and safety considerations.

Work was being completed on print media and television coverage of nuclear, solar and coal issues; the relationship of human values and attitudes to the nuclear power conflict; DOE's communication process; and public attitudes toward waste disposal facilities. Work was also begun on separate studies of five other nuclear waste management public issues.

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

This work is aimed at providing data and methods to quantitatively address technical factors such as seismic stability that are a concern in licensing and safety analysis procedures for waste repositories. The work has involved developing the data base and computerized capability to manipulate the data for practical applications.

This quarter the data base and display systems have been summarized for use by DOE in repository selection and licensing activities.

Work continued on the summary report of the seismic data base and analysis program and is being reviewed. The geographic data base and analysis program summary will not be completed until next quarter because of priority commitments of staff to other programs.

GEOLOGIC INVESTIGATIONS IN THE NORTHWEST - G. A. Sandness and W. M. Phillips

The purpose of this research program is to identify and evaluate a major photolineament (alignment of topographic, tonal, or textural features seen in aerial photographs or satellite imagery) in the Northwest. Based upon analysis of Landsat satellite imagery, augmented by U-2 and conventional aerial

photographs, a major NE-SW trending photolineament about 270 km long was identified in the states of Washington and Oregon. The photolineament crosses the Columbia River near Portland, Oregon, and continues northeast to a point just east of Omak Lake, Okanogan County, Washington.

Upon selection of the major photolineament, a data base was constructed. Analysis of the data base indicated that the NE-SW trending faults had been previously recognized along two portions of the major photolineament. The two areas, the Bumping Lake region east of Mt. Rainier and the Lake Merwin area southwest of Mt. St. Helens, were selected as sites for geologic field studies.

During the quarter, work continued on a definitive analysis of the geology of the photolineament, utilizing the large amounts of data acquired in the field studies completed last quarter. The thrust of this quarter's effort was to summarize the investigation.

During this quarter, a final report describing the bedrock geology of the Omak Lake portion of the Bumping Lake photolineament was received from Washington State University. The report includes a geologic map of the Omak Lake study area and stereonet depicting joint orientations along the photolineament trace. The report's conclusion is that present evidence indicates that the photolineament is not joint-or-fault-controlled in that area and that alignment of the Columbia River in the region may be fortuitous. However, examination of U-2 photographs from an area slightly to the north of that study area indicates that striking joint orientation anomalies do exist.

Petrographic analysis and compilation of geologic maps and cross-sections are continuing. It is anticipated that the summary of this investigation will be completed during next quarter.

KRIGING - P. G. Doctor

The objective of this project is to evaluate the potential applicability of kriging to the characterization of geologic repositories for the storage of high-level radioactive wastes. Adequate characterization (location, extent, hydrologic regime, etc.) of such repositories is required for site selection and safety assessment.

Kriging is a statistical method for estimating functions that describe spatially distributed phenomena such as groundwater elevation (potential) and depth to basalt. A potential repository must be evaluated from very sparse data. Core samples, which provide the most reliable information, are intrusive and costly; thus the objective is to minimize the number of cores taken. The contribution of kriging to this objective is twofold. First, kriging produces contours of the geologic formation of a potential site with an associated measure of uncertainty. This provides a more complete characterization than can be gained from other contouring methods. Second, given the existing data, kriging can be used to optimize the selection of additional sampling locations. This feature provides a rationale for selecting sample locations and could significantly cut costs of acquiring data.

The most efficient way to evaluate kriging is to experiment with the techniques on representative data sets. The Hanford Wells data base proved to be the most accessible for this purpose. Water elevation tends to be fairly smooth spatially distributed phenomenon, and since the Hanford Wells water elevation data set is fairly extensive, the September 1973 and January 1975 data were used to gain familiarity with the techniques. Compared to

the water elevation data set, the depth-to-basalt measurements seem more characteristic of the type of data used to evaluate potential sites.

The kriging computations are performed using BLUEPACK, a computer package developed by the Paris School of Mines and installed at the DOE-Las Vegas computing facility.

The analysis of the three data sets (two for water elevation and one for basalt contours) was completed and an evaluation written this quarter.

The basic criterion used in this study to evaluate kriging is how realistically it contours the water potential and basalt surfaces. The water potential surface is well known; hand-drawn contour maps, based on substantive knowledge in addition to well data, are considered the standard. The kriged contours based on only the well data are virtually identical to the hand-drawn contours. The kriging standard errors (measure of uncertainty) help quantify the precision of these contours in various places on the reservation. They can be used to help select additional data sampling locations.

The basalt surface is much less well known than is the water potential surface. Consequently, existing top-of-basalt contour maps rely heavily on substantive knowledge. Discontinuities, such as synclines, necessitate a more complex kriging analysis. The kriged contours based on only the well data and the approximate locations of two major synclines showed good agreement with the existing contour maps. The basalt kriging standard errors were much larger than those for the water elevation surface, reflecting the basalt deposit's more complex nature.

The main conclusion of this study is that kriging can produce realistic contours of the geologic structure of a potential repository site. Second, the kriging standard

errors provide a quantification of the precision of the contours, which can be useful evidential information for developing a rationale for site selection. Third, insights gained from kriging analyses can contribute to the substantive knowledge of the geologic phenomena under study.

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

Teknekron completed work on reporting on development of improved methods for assessing nuclear waste management options. This methodology represents an effective technique for systematically analyzing complex waste management issues having societal implications. It could also be employed for analysis of purely technical issues with no obvious societal impacts but with dimensions requiring the attention of multiple technical disciplines for their resolution. The only difference, in principle, in applying the analytical technique for these two cases is the different mix of disciplines selected for the panel of experts.

In any future application of this analytical framework, the following possibilities warrant consideration:

1. Develop a prioritized inventory of waste management issues or problems for which solutions are deemed needed to gain approval of implementation plans and related facilities for managing the various categories of nuclear wastes; this could be done for the entire spectrum of waste categories or for any portion thereof.
2. Analyze all or any portion of the inventory of waste management issues, with the objective of compiling for the decision-maker alternative courses of action for resolving each issue.

Obviously the broad spectrum of applications for the methodology suggested in 1 and 2 above would require careful attention to the makeup of the disciplines to be represented on the panel performing the analyses. However, categories of issues or problems requiring analysis could be compiled in such a way that each category would contain issues sufficiently related and interdependent that their analysis could be undertaken by the same panel.

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

Teknekron, Inc., was notified to proceed on a contract to develop an optimization algorithm which will relate costs and occupational and long-term safety considerations as a function of depth of a repository. Teknekron project staff visited PNL to discuss detailed scope of the project and to obtain whatever cost data from the Draft Environmental Impact Statement: Management of Commercially Generated Radioactive Waste (DOE/EIS-0046D) would be useful to their study.

Teknekron has begun several tasks in parallel in support of the subject study. Source terms are being generated by assuming a mix of LWR spent fuel elements and high-level waste canisters transported to a repository over a period of 5 years, 20 years, and 30 years.

A literature review is in progress to define the generic geologic characteristics to be used in the analysis. To develop the trade-off algorithm, the generic stratigraphies and hydrologic properties are being defined to as far as 4000 m below the surface.

Teknekron associates at the Virginia Polytechnic Institute have commenced work on correlating the costs of exploration, mining,

hoist systems, structural supports, ventilation, air conditioning, and safety systems with the repository depth. They have also started work on relating the construction and operation hazards with the depth of the repository.

Teknekron has commenced evaluation of simple hydrologic and nuclide migration models to be used in their radiological impact analysis. The selected equations will be used to correlate the radiological impact with the depth. Impacts will be converted into costs using current Nuclear Regulatory Commission cost factors. The optimization model will be based upon predicted population dose commitment, which lends itself to conversion into cost factors. Maximum individual dose rate will not be calculated.

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

The Human Affairs Research Center's support program for FY-79 involves seven separate tasks. Task 1 is a workshop entitled: "Nuclear Waste Management: Understanding Social Assessments" to be held February 21 and 22, 1979, at the Battelle Seminar and Study Programs facilities in Seattle. The workshop is for DOE and other Federal agencies and for DOE contractors. Invitations have been mailed out to about 50 individuals who work for the government on nuclear waste management projects. Approximately 30 persons are expected to attend.

Task 2 deals with public perception of risks associated with production of electric power. A consulting agreement with Dr. T. E. Earle, who will be involved in the work, was completed, and work planning and analysis were begun.

Task 3 comprises analysis of television coverage of sources of electric power. It

focuses on comparative analysis of network television news coverage of nuclear power, coal, and solar stories, as summarized below. First, coal news stories (including coal strikes) outnumbered nuclear news stories 463 to 266 from 1972 to 1977. Only 26 news stories mentioned solar power. Coal stories were given the most prominence by the networks, followed closely by nuclear power and followed distantly by solar power. Nuclear power stories, on the average, were slightly antinuclear; coal stories, on the average, were neutral; solar power stories were very prosolar. The cost discussions outnumbered the benefit discussions about four to one both for nuclear power and for coal, whereas for solar, the benefit discussions outnumbered the cost discussions about six to one. Issues discussed by the news stories regarding coal and nuclear power were about the same, although environmental concerns were much more prevalent in the coal stories. In general, the nuclear power news reporting did not seem to be greatly biased, especially when nuclear stories were compared to coal stories.

Task 4 comprises review and completion of FY-78 reports. Materials are nearing completion in time for the February conference (Task 1).

The literature search and analysis of materials was begun for Task 5, which is an analysis of conflicts caused by discrepancies between perceived risk and calculated risk.

Task 6 is a study of ways in which community-level groups deal with environmental problems. This will be done through case study analysis and through analysis of community action literature. Task 6 will also develop a framework for community action group participation in environmental decision-making.

Task 6 is progressing according to plan. A bibliography of studies has been assembled on community-action group involvement in environmental decision-making. Technical review of these materials is presently in progress. Case study materials of facility siting controversies have been assembled and evaluated, and a draft of the results of this work is in preparation. Work has also begun on a limited re-analysis of data (collected in FY-1978) in an effort to examine factors in individuals' participation in action groups which focus upon nuclear power issues.

Work planning for Task 7 was completed in November. This task focuses on the effectiveness of different mediation procedures regarding environmental issues. The literature review was begun in October and will continue through next February. A computerized literature search will be accomplished in January. A semiquantitative method of examining case studies on mediation approaches to environmental issues was found in December and is currently being adapted for use on this project.

7.0 WASTE MANAGEMENT SAFETY STUDIES

W. K. Winegardner - Project Manager

Aid in assuring that waste management and its inherent safety function are given proper emphasis as various alternative fuel cycles become viable; develop assessment methods; identify portions of waste management systems that could benefit by safety design emphasis.

SUMMARY

System definition of a coprocessed UO_2 - PuO_2 fuel cycle was completed. Radiological consequences were calculated for the routine release of ^{85}Kr from a 2000 MT/yr coprocessing facility.

FUEL CYCLE ASSESSMENT - J. W. Voss and S. E. Lyke

A chemical separations facility for the extraction of plutonium denatured with uranium and spiked with fission products was conceptualized to permit definition of radioactive wastes from a coprocessed UO_2 - PuO_2 fuel cycle. This systems definition work will provide the basis for an assessment which will be compared to that previously developed for a once-through fuel cycle.⁽¹⁾ Figure 7.1 shows a schematic of the coprocessed

fuel cycle considered. Fresh UO_2 reactor fuel, enriched to 3 to 4% ^{235}U , is irradiated in a typical light water reactor (LWR) and is subsequently shipped to a fuel coprocessing facility. At this facility, the irradiated fuel is sheared and dissolved. In the chemical separations process, only part of the uranium, fission products and other actinides is separated from the plutonium, in such a way that a pure plutonium stream never exists in the plant. The denatured and spiked stream is converted to an oxide powder form and shipped to a mixed-oxide fuel fabrication facility. Uranium, either natural or depleted ^{235}U , is mixed with the spiked PuO_2 - UO_2 material, blended to attain a fissile enrichment of 3 to 4%, and fabricated into fuel assemblies. This fuel is subsequently recycled to an LWR.

The coprocessing system conceptualized to achieve the denaturing and spiking of the

FIGURE 7.1. Schematic of the Coprocessed UO_2 - PuO_2 Fuel Cycle

recycled plutonium stream is sketched in Figure 7.2. The process uses the concepts developed for the standard PUREX process.^(2,3) Irradiated fuel is chopped into small pieces, dissolved in nitric acid, and filtered of hulls and hardware materials. The dissolved fuel next enters the extraction column, commonly the HA column, where it is mixed with acid and an organic solvent. About 99% of the fission products and actinides are separated from the uranium and plutonium solution. The uranium and plutonium, being in an organic solution, next enter a scrub column, where acid and reductant are added. About 85% of the uranium is separated in this scrub column and is routed to the uranium purification and fluorination section. The remaining uranium and plutonium stream, which is contaminated with fission products and actinides, is concentrated, calcined, and processed through a UO_3 reduction system. The

spent fuel entering the process is estimated to have a Pu/U ratio, by weight, of 2/100, along with a radioactivity of $4E6$ and $6E5$ Ci/MTHM of fission products and actinides, respectively. For each metric ton of heavy metal entering the facility, about 0.15 MTHM, in the form of mixed U/Pu oxide spiked with fission products and actinides, leaves the plant en route to the mixed-oxide fuel fabrication facility. The balance of the heavy metal, about 0.85 MTHM, leaves the facility as UF_6 en route to an enrichment plant. The mixed U/Pu oxide stream has a Pu/U ratio of 15/100 by weight. It is spiked to a radioactivity level of $5E5$ Ci/MTHM fission products, $2.8E6$ Ci/MTHM of plutonium isotopes, mostly ^{241}Pu , and $5.7E3$ Ci/MTHM of other actinides.

Radioactive wastes generated in the co-processing facility are quite similar to predictions for standard reprocessing

FIGURE 7.2. Conceptualized Separations Process for Coprocessed UO_2 - PuO_2 Fuel Cycle (separation assumed at 180 days out-of-reactor)

facilities.⁽⁴⁾ Minor radioisotopic differences are predicted in the various waste streams. An obvious difference would be wastes resulting from the calcination of plutonium solutions which traditionally have been contaminated with only plutonium. In the coprocessing facility, this operation is performed on a plutonium stream contaminated with fission products, which will also be present in the wastes. No significant volume changes are predicted.

The wastes anticipated for the balance of the coprocessed $\text{UO}_2\text{-PuO}_2$ fuel cycle are only slightly different from the conventional recycle fuel cycle. As mentioned previously, the mixed plutonium-uranium oxide powder re-fabricated for reactor fuel is spiked with fission products and actinides. The radioactive wastes from the mixed oxide fuel fabrication facility will be contaminated with fission products and actinides in the coprocessing fuel cycle, as opposed to the conventional cycle in which they are not.

DISPERSION/ACCUMULATION TRADE-OFF ASSESSMENT - P. J. Mellinger

Work continued on methods for providing insights concerning tradeoffs between management by dispersion/dilution and by accumulation/containment. Assessment of noble gas management techniques, with and without ^{85}Kr recovery, is currently being used as

the basis for this work. Radiological consequences were calculated for the routine release of ^{85}Kr from a 2000-MT/yr fuel reprocessing facility capable of servicing about sixty 1000-MWe LWRs. In this set of calculations it is assumed that no attempt was made to recover the krypton. Three population segments were analyzed, and the 80-year cumulative dose was calculated for each as shown in Table 7.1.

Methodology - World Dose Calculation

Similar integrated time periods must be used in the dose calculations for cases in this study to assure that a comparison can be made at the program's conclusion. Therefore the storage scenario must be determined prior to calculating the dose to the world population from the chronic routine release of all ^{85}Kr (no recovery) from coprocessed fuel.

Upon initial startup of the fuel reprocessing plant (FRP), pressurized cylinders containing ^{85}Kr will be filled and placed in storage. This will continue for the 30-year life of the coprocessing plant. Upon shutdown of the coprocessing plant, no more cylinders will enter the storage facility. The facility will be maintained status quo for 20 years, at which time, the first cylinders received will be 50 years old and contain about 4% of their original ^{85}Kr .

TABLE 7.1. The 80-Year Cumulative Dose to Three Population Segments from ^{85}Kr Exposure

Population Segment	Total ^{85}Kr Body Dose	Natural Radiation Background Total-Body Dose
Maximum individual ^(a)	5.7×10^{-3} rem	8.0 rem
50-mile population	1.3×10^3 man-rem	1.6×10^7 man-rem
World population	2.9×10^5 man-rem	5.1×10^{10} man-rem

a. The maximum individual resides at the offsite location of maximum ground-level concentration of ^{85}Kr .

activity. As each cylinder reaches age 50, its contents will be released to the uncontrolled atmosphere. The venting process will then necessarily last for 30 years to assure the 50-year age of the last cylinders to enter storage. The total time of this scenario is 80 years.

Therefore the dose to the world population from the chronic release of all ^{85}Kr will be an 80-year cumulative dose. Kr-85 will be released from the FRP stack for 30 years. At the end of the 30-year period, the atmospheric activity will be a factor of 13.2 times the curies released per year. During the following 50 years, the atmospheric ^{85}Kr concentration will decrease as a result of the radiological decay.

Therefore the total time-integrated concentration of ^{85}Kr in the atmosphere is the integral of the sum of the expressions for concentration during the two time periods (0-30 years and 30-80 years), yielding Equation 1:

$$\text{TIC} = \left[\frac{(1 \text{ Ci/yr})(10^{12} \text{ pCi/Ci})}{3.96 \times 10^{18} \text{ m}^3} \right] \left[\frac{1}{\lambda^2} (\lambda t_1 + e^{-\lambda t_2} - e^{-\lambda \Delta t}) \right] \frac{\text{pCi} \cdot \text{yr}}{\text{m}^3} \text{ per Ci/yr released} \quad (1)$$

where: $3.95 \times 10^{18} \text{ m}^3$ is the volume of the world's atmosphere

t_1 is the time over which the release occurs, 30 years

t_2 is the time over which the dose is calculated, 80 years

$\Delta t = t_2 - t_1$, 50 years.

For the conditions stated above, the expression in the brackets becomes 455 and the time integrated concentration is:

$$\text{TIC} = 2.53 \times 10^{-7} (455) = 1.15 \times 10^{-4} \frac{\text{pCi} \cdot \text{yr}}{\text{m}^3} \text{ per Ci/yr released.}$$

The accumulated dose is then a product of TIC, the annual ^{85}Kr release rate, the world population and the dose conversion factor, presented in Equation 2.

$$\text{Dose} = \left[1.15 \times 10^{-4} \frac{\text{pCi} \cdot \text{yr}}{\text{m}^3} \text{ per Ci/yr} \right] \left[2.1 \times 10^7 \text{ Ci/yr} \right] \left[6.4 \times 10^9 \text{ persons} \right] \left[1.9 \times 10^{-8} \text{ rem/yr per pCi/m}^3 \right] \quad (2)$$

$$\text{Dose} = 2.9 \times 10^5 \text{ man-rem for the 80-year period}$$

This is equivalent to an average of 4.4×10^{-5} rem per person.

Since the 50-mile population dose and the maximum individual dose use more conventional methodology, they are not elaborated upon here.

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8.0 WASTE ISOLATION SAFETY ASSESSMENT PROGRAM

A. Brandstetter - Project Manager

Mark 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 to the National Waste Terminal Storage Program.

SUMMARY

Planning for the Waste Isolation Safety Assessment Program (WISAP) for FY-79 through FY-85 continued during this quarter. A Technology Program Plan (TPP) was prepared for FY-79 and transmitted to DOE and the Office of Nuclear Waste Isolation (ONWI). Expansion of the TPP for FY-80 through FY-85 and preparation of the Work Package Proposal Agreement (WPPA) for FY-79 through FY-85 was begun. The work plan for FY-79 includes continued development of generic methodology and collection of data, but emphasis will shift to four reference geology initial analyses: the Waste Isolation Pilot Plant (WIPP), the Paradox Basin bedded salt, the Columbia Plateau basalt, and a salt dome.

During the first quarter of FY-79, work continued for the Paradox Basin initial

analysis (started in the last quarter of FY-78), and work was started on the other three geologies. Work continued on several topical reports; reports on Hanford evapotranspiration estimates (PNL-2698) and on solution species of Pu-239 (V) (PNL-SA-7027) were issued. Reports on technical progress in FY-77, on the characterization of rock samples, on a three-dimensional, finite-element groundwater flow model, on release scenario analysis methods, on the 1977 workshop on disruptive phenomena, and on the 1977 workshop on transport modeling, were completed and submitted for DOE clearance. Several papers were presented at conferences. A quality assurance (QA) plan was formulated and a QA Manual prepared. Suggestions for validating radionuclide migration models with in situ tests and suggestions for evaluating safety of borehole plugging were formulated.

TASK 1 - RELEASE SCENARIO ANALYSIS -

G. L. Benson, Task Leader and R. A. Craig - Assistant Task Leader

Site Studies

Disruptive release scenarios were developed for the reference site analysis of the Columbia Plateau basalts. A list of data requirements for the analysis of Columbia Plateau basalts was prepared and transmitted to the Basalt Waste Isolation Program (BWIP) of Rockwell Hanford Operations (RHO). The WISAP team developed four potential disruptive release scenarios for the basalt using information developed during the FY-78 program. A meeting was held with BWIP staff in November to discuss the scenarios and related data needs. These scenarios are currently being put in final form for input to Task 3, Release Consequence Analysis, in the next quarter.

Work was begun on developing release scenarios for a salt dome reference site. Meetings concerned with salt dome reference site analysis were held with Law Engineering, the Geologic Program Manager (GPM), and Bechtel National, the Regulatory Project Manager (RPM). These meetings were designed to set the framework for the ensuing safety analysis. Discussions centered around data and methodologies that would be needed and used in the analysis. This work is continuing, with preliminary scenarios slated for input to Task 3 at the end of the next quarter.

Task 1 has been involved in preliminary meetings with Sandia Laboratories for the Nevada Test Site (NTS) and the Waste Isolation Pilot Plant (WIPP), Woodward and Clyde for the Salina Basin, and Bechtel for the Paradox Basin. The task provided input for the parametric studies of the Paradox Basin and the WIPP site.

Model Development

The pilot program for the repository simulation model was exercised to test the overall simulation methodology. This test was performed on data for the Columbia Plateau basalts. The preliminary tests indicated that the methodology developed during FY-78 could be used for the simulation model. However, the pilot program incorporated only a few disruptive phenomena, and hence its current version is not usable for scenario analysis. The pilot program is currently coded in the FLEC computer language and operational on a Digital PDP 11/55 computer system.

A second-generation repository simulation model for the Columbia Plateau basalts has been conceptualized. Logic flow charts have been developed that incorporate much of the FY-78 Task 1 consultant input. The model is scheduled for completion in the next quarter. This version will be designed to run in an interactive mode to facilitate parametric studies by geoscientists. Work has begun also on conceptualization of a repository simulation model for a salt dome reference site.

TASK 2 - WASTE FORM RELEASE RATE ANALYSIS -

D. J. Bradley - Task Leader and
R. P. Turcotte - Assistant Task Leader

Progress at Pacific Northwest Laboratory

Leaching tests on spent fuel and actinide-doped glass using an IAEA procedure continued at PNL. A combination glove box/open-face hood has been installed. Several modifications were done to meet air balance requirements. The hood was connected to off-gas equipment, and the system was checked out and approved. Temperature baths were

ordered, received and installed for the static leach tests on waste glass.

An outline of experiments for static and autoclave testing was worked out for cold and actinide-doped glass. The autoclave tests were started on schedule using gold capsules as pressure vessels. These tests indicate that welding will be required at both ends of the gold vessel because the pressure weld tends to leak.

In preparation for beginning the static leach tests on spent fuel in the next quarter, the leach containers and holders have been fabricated. The leach cabinet for this hot cell work has also been fabricated and will be placed in the hot cell shortly. The spent fuel for these tests has been weighed out and photographed. A survey was completed on release of trace contaminants from various leach test container materials. Polypropylene, polymethylpentene, and teflon were the best materials found.

Progress at Brookhaven National Laboratory

Delays have been encountered in obtaining both incinerator ash wastes required in the leaching program at the Brookhaven National Laboratory (BNL). The rotary kiln incinerator ash is scheduled to be produced at Rocky Flats early in the next quarter. This ash, although obtained from uncontaminated materials, must be analyzed before shipment from Rocky Flats is authorized. The "hot" dual chamber burner ash has been packaged by Rocky Flats personnel and is scheduled to be shipped to BNL early in the next quarter. To date, waste formulations have used a simulated Rocky Flats ash composition. When the actual incinerator ash is received, the formulations determined with simulated ash will be verified.

Progress at Lawrence Livermore Laboratory

The single-pass flow leach experiment is continuing at Lawrence Livermore Laboratory (LLL). In the past three months the following areas have been emphasized: 1) performing analysis on the backlog of samples that were generated through the ten sampling intervals--308 samples because of "double" samples taken on the tenth sampling day--and 2) beginning the design of a new single-pass leach system in which rock material will be incorporated with the simulated waste glass.

As of December 31, 1978, 171 of the 252 samples collected through the ninth sampling day have been plated on platinum discs, and 133 samples (through the seventh sampling day) have been counted for both neptunium and plutonium. The samples in which brine is the leachant are now being analyzed as a result of the development of a convenient radiochemical procedure for ^{237}Np and ^{239}Pu analysis in the brine samples.

Chemical analysis has begun for the glass bead leachant samples using ion-coupled plasma emission spectroscopy equipment. A test is being conducted to determine the most appropriate technique to use for analysis of the acidified brine samples. The analytical results beyond the third sampling day have not been assembled for reporting at this time. The results of the first three sampling days are reported by Weed et al. (PNL-SA-7196).

A scope camera has been purchased for the ND-60 MCA in order to document the quality of the alpha-spectroscopy analyses. Space has been secured for the new rock-glass leach experiment. It will be in the same room as the current leaching experiment. An eight-channel Gilson Minipuls peristaltic pump has been ordered. It will be evaluated for

incorporation into the new rock-glass leach experiment. A new source of small-bore tubing has been found which should allow us to reduce flow rate significantly. Dye dispersion tests on the cells will be done with the pump. Much thought is also being given to this new leach test to develop useful concepts and designs.

TASK 3 - RELEASE CONSEQUENCE ANALYSIS -

J. R. Raymond - Task Leader and

R. W. Nelson - Assistant Task Leader

Site Studies

Additional data were collected and compiled for the WIPP site. Meetings were held with NUS and Sandia Lab personnel to discuss the WIPP release scenario and the release consequence analysis made with Intera Environmental Consultant's model. WISAP models are being used to analyze the same scenario for comparison. The Intera model was implemented on the PDP 11/45 computer to gain understanding of the methods used for the WIPP study. The model also will be set up on the Univac 1100/44 computer to allow comparison of the two methods. The VTT hydrologic model was implemented for the WIPP site, using the Sandia/Intera data and release scenario. Initial runs were made and the results compared with the output from the Intera model. Both methods computed comparable water travel times from the repositories to the groundwater discharge site.

Meetings were held with personnel from Bechtel National, Inc., and Law Engineering Corporation. Data were obtained on the Louisiana and Texas salt domes preparatory to implementing release scenario and release consequence analyses.

Parametric/sensitivity studies were defined for the Paradox Basin.

Meetings were held with Stone and Webster Engineering Corporation (GPM for the Salina Basin) and associated consultants to discuss hydrologic modeling methods and data needs for the Salina Basin. The GPM is interested in implementing a coarse grid model to assist with basin evaluation and data needs. Methods and data format for this modeling effort will be compatible with the release consequence analysis to be initiated under WISAP next year.

Model Refinement

Development of input data logic and computer coding was completed for coupled solution of pressure, energy and mass transport for the three-dimensional finite element model. Refinements were made in the MMT transport model code and associated support programs.

The closed form mathematical solution for the steady two-dimensional flow system and pathlines was completed for the "front end" of the GETOUT code and written up as an appendix for subsequent use in a document. Specifically obtained were: 1) the potential function, 2) the steady pathline equation, and 3) the equation for the travel time along the pathlines.

Fluid Flow Technology Development

Work was started on the inverse problem and application of kriging and stochastics to hydrologic and transport modeling. Detailed two-dimensional hydrologic fields were generated for the inverse study to evaluate the "sample size" or "sample scale" problem associated with derivation of hydraulic conductivity distributions in heterogeneous media. Tests of "kriging" will also be applied to these fields.

Detailed results were obtained on the significance of dispersion in ground-water contaminant transport. These results show the possibility that the dispersion term, at least for many cases, may be deleted from the transport equation without significant effects on contaminant outflow rates.

Data Base Development

The CIRMIS system was updated to provide more versatility for WISAP studies, and additional work was done on the computer programs to display stratigraphic cross-sections.

A waste/media interaction data base was partially completed; it is to permit users to select Kd values from WISAP Task 4 laboratory sorption-desorption test results. Most of the information from 228 test cases was entered into the data base, including geologic material properties, ground-water properties, Eh, pH, nuclide and Kd values. Files were also developed which contain properties of all isotopes in the isotope table.

Programs were written to calculate radionuclide inventory tables. Procedures were implemented to determine the remaining radionuclides after a set time and to use the sorption-desorption data base to select the Kd values for these radionuclides.

All available stratigraphic data for the WIPP site have been keypunched and entered into the new CIRMIS data base. Additional data can now be added upon receipt.

TASK 4 - SORPTION/DESORPTION ANALYSIS -

R. J. Serne - Task Leader and

J. F. Relyea - Assistant Task Leader

Evaluation of Experimental Methodology

At present investigators from various laboratories use numerous experimental designs

in determining sorption Kd's and/or migration rates for nuclides. Unfortunately, it appears that the resultant data are often dependent not only on the type of solution and geologic material used, but also on the experimental methods. No systematic effort has been expended to resolve the apparent method dependencies, to assess which method(s) yields the most accurate results, or to determine what conversions can be made to compare the results of different methods. Thus a study of the various experimental methods for determining sorption-migration data has been undertaken. The ultimate aim is to form a consensus as to which method(s) should be proposed as standard measurement technique.

During this quarter, comparisons of batch Kd methods with axial filtration, channel chromatography, and flow-through columns have been performed. For selected ground water-rock and pure mineral-simple ground-water systems, results show that a favorable comparison can be obtained with careful experimentation. In flow-through experiments on intact porous materials such as tuff and limestone, migration rates for strontium and cesium were found to vary at most by an order of magnitude from migration rates predicted from batch Kd values on crushed materials. The need for a scaling factor to account for the differences in solid adsorbate material contacted between crushed and intact material should be pursued. Large differences between observed porosity by tritium breakthrough curves and porosities calculated from water saturation content in laboratory columns need further study. Possibly the discrepancies observed so far (maximum about one order of magnitude) between migration in intact cores and predictions from batch Kd's on crushed material are not significant from the standpoint of the overall safety assessment.

Data Bank Acquisition and Synthesis

Work continued on generating data for the generic data bank. Data collected during this report period include K_d 's for iodine, technetium, neptunium, plutonium, americium, strontium, cesium, europium, cerium, nickel, and selenium onto media such as shale, granite, basalt, limestone, tuff, and numerous pure minerals from synthetic salt brines and ground waters. With the large number of combinations of rock type-ground-water type and nuclides, this task of collecting the data bank will continue for at least two more years. As site-specific samples become available from potential repositories, emphasis will shift to them.

The data generated in FY-77 and FY-78 are being entered into a PNL computer information retrieval system to facilitate sorting, organization, and statistical synthesis.

Validation and Mechanism Studies

During this quarter little work was performed on theoretical validation studies such as tabulation of thermodynamic data or incorporation into existing thermodynamic codes. The lack of necessary thermodynamic data on many important secondary minerals is impeding the use of the thermodynamic equilibrium computer models in the quantitative prediction of the fate of radionuclides in the environment. Using other similar minerals for which data are available has allowed semiquantitative predictions to be made on the fate of trace constituents in a basalt regime.

The emphasis of ongoing mechanism studies is to investigate the basic factors and their interactions that control the sorption-desorption of actinides, long-lived fission products, and activation products by geologic media. With knowledge of mechanisms, migra-

tion rates can be predicted from more theoretical and fundamental data for any proposed repository environment. The use of theoretical and fundamental data may cut the costs expended to quantify the migration rates of nuclides at specific sites.

During this quarter Oak Ridge National Laboratory (ORNL) continued studies on the effect of ionic strength and tracer concentration on the adsorption of strontium, barium, cesium, and europium on montmorillonite clay. Systematic studies were made of ion exchange as a function of supporting electrolyte concentration at constant pH. These studies show that the adsorption of strontium, barium and cesium is about as expected from conventional idealized ion-exchange equations. The trivalent ions europium and americium, however, do not conform to idealized equations.

In other studies at ORNL it was found that the redox-sensitive elements neptunium and technetium should not be stable in their mobile form (NpO_2 and TcO_4) under reducing repository conditions. Thus, current assessments using oxidizing K_d values for neptunium and technetium may be overestimating their potential hazard to the public.

At Argonne National Laboratory (ANL) kinetic and partitioning data for americium in fissures of impermeable rock were used as input to a transport model which includes time-dependent sorption (kinetic effects). Laboratory infiltration experiments into fissures were compared with model-predicted behavior. The favorable comparison emphasizes the need to consider reaction kinetics in predicting nuclide migration.

Microautoradiography studies of thin rock sections have been performed at Lawrence Berkeley Laboratory (LBL) and Los Alamos Scientific Laboratory (LASL). These have shown that the active sorption sites in bulk rocks (basalt, quartz, monzonite, argillite,

tuffaceous alluvium and tuff) for actinides are generally the secondary mineral alteration products such as clays, zeolites, etc.

Accelerated weathering studies performed at Rockwell Hanford Operations (RHO) on basalt and argillite increased the sorption tendencies for most isotopes studied (cesium, strontium, plutonium, americium, neptunium). Weathering of granite showed no observable changes in sorption.

Studies conducted thus far at PNL have revealed that: 1) in radioactive waste disposal cribs containing discrete PuO_2 particles, the concentration of soluble plutonium is predictable from a solubility relationship; 2) the solubility of PuO_2 and Pu(IV) hydroxide actually observed under environmental conditions differs from theoretical predictions; 3) Pu(IV) polymer formation can be predicted from pH and total soluble plutonium concentration; 4) Pu(V) rather than Pu(VI) may be the dominant

species in environmental samples in relatively oxidizing conditions; and 5) a potentiostat can be used to control the Eh of geochemical suspensions to aid in determining adsorption coefficients of various oxidation states of multivalent elements such as plutonium, neptunium, and technetium.

Workshop and Subcontracts

A workshop in the FY-78 sorption/desorption studies was held at the Seattle Research Center October 2-5, 1978. PNL staff and the subcontractor staff presented their FY-78 results, participated in discussions evaluating the FY-78 work, and started planning the FY-79 work. Following the workshop, the formulation of scopes for the FY-79 subcontracts was started, considering the results of the workshop and evaluating FY-78 reports submitted by the subcontractors.

REPORTS ISSUED DURING QUARTER

C. R. Cole and S. K. Gupta, A Brief Description of the Three-Dimensional Finite Element Ground-Water Flow Model Adapted for the Waste Isolation Safety Assessment Program. PNL-2652, Pacific Northwest Laboratory.

R. W. Wallace, A Comparison of Evapotranspiration Estimates Using DOE Hanford Climatological Data. PNL-2698, Pacific Northwest Laboratory. October 1978.

D. Rai, R. J. Serne and J. L. Swanson, Solution Species of $^{239}\text{Pu(V)}$ in the Environment. PNL-SA-7027, Pacific Northwest Laboratory. December 1978.

R. J. Serne, D. Rai, J. F. Relyea and L. L. Ames, Review of Progress in Measuring and Understanding Interaction of Waste Radionuclides with Geomedia. PNL-SA-7049, The Materials Research Society, International Symposium on Science Underlying Radioactive Waste Management, Boston, MA. November 29-December 1, 1978.

H. C. Weed, D. G. Cowles, D. J. Bradley, R. W. Mensing and J. S. Schweiger, Leaching Characteristics of Actinides from Simulated Reactor Waste Glass. PNL-SA-7196 (also UCRL-81147), The Materials Research Society, International Symposium on Science Underlying Radioactive Waste Management, Boston, MA. November 29-December 1, 1978.

J. A. Stottlemire, G. M. Petrie and M. W. Mullen, Computer Enhanced Release Scenario Analysis for a Nuclear Waste Repository. PNL-SA-7232, The Materials Research Society International Symposium on Science Underlying Radioactive Waste Management, Boston, MA. November 29-December 1, 1978.

C. R. Cole, Panel Discussion on Engineering for Radioactive Waste Repositories in Deep Geologic Formations: Migration Modeling. PNL-SA-7286S, 71st Annual Meeting of the American Institute of Chemical Engineers, Miami, FL. November 12-16, 1978.

9.0 WELL-LOGGING INSTRUMENTATION DEVELOPMENT FOR SHALLOW LAND BURIAL

R. L. Brodzinski - Project Manager

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

SUMMARY

A Phoswich-type detector has been borrowed for evaluation as a ^{90}Sr and transuranic logging system in boreholes and cased wells.

Gas and liquid scintillation spectrometric techniques are being evaluated for determination of tritium by well-logging.

Passive neutron detection techniques for quantitative analysis of plutonium, americium, and curium isotopes in soil have been completed at soil moisture levels of 16%. Experiments have also been completed to obtain the sensitivity and detection limits for measuring these actinides by photon well-logging.

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

A Phoswich-type detector has been borrowed for evaluation as a ^{90}Sr and transuranic logging system in boreholes and cased wells. This counting system has been set up and calibrated for the forthcoming laboratory experiments.

A paper based on earlier work performed in this study, "A Well-Logging Technique for the In Situ Determination of ^{90}Sr ," by R. L. Brodzinski and H. L. Nielson, has been

accepted for publication in the Proceedings of a Symposium on Recent Advances in Nuclear Analytical Methods, April 1-6, 1979, in Honolulu, Hawaii.

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

Various methods for in situ well-logging sampling and analysis of tritium continue to be evaluated for their technical viability, reliability, selectivity, and potential costs. Primary emphasis is being placed on gas and liquid scintillation spectrometric techniques.

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

Calibrations with the passive neutron detection technique for the determination of actinides were completed at soil moisture levels of 16%. Several fundamental relationships were derived relating the neutron-induced activity levels to the source of neutrons, i.e., whether the neutrons originated from spontaneous fission or from (α, n) reactions on light isotopes. These relationships are important in that they allow the

isotopic composition of an unknown transuranic contamination in soils to be determined by this technique.

A paper based on research results of this program entitled "In Situ Quantitative Determination of Transuranic Elements in Areas of High-Level Gamma Radiation," by N. A. Wogman and R. L. Brodzinski, was published in National Bureau of Standards Special Publication 528, Analytical Methods for Safeguards and Accountability Measurements of Special Nuclear Materials. Two additional papers based on this work, "A Technique for the In

Situ Determination of Transuranic Elements in Relatively Inaccessible Locations" and "A Technique for In Situ Moisture Level Determinations in Relatively Inaccessible Matrices," by R. L. Brodzinski, have been accepted for publication in the Proceedings of a Symposium on Recent Advances in Nuclear Analytical Methods, April 1-6, 1979, in Honolulu, Hawaii.

Data acquisition for determination of actinides in soils by photon detection techniques has been completed, and the results are currently being evaluated.

10.0 MONITORING AND PHYSICAL CHARACTERIZATION OF UNSATURATED ZONE TRANSPORT

S. J. Phillips - Project Manager

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

SUMMARY

Laboratory tests of radionuclide transport in unsaturated soil columns were continued. Cobalt, technetium, tritium, strontium and iodine were tested for mobility in Hanford sediments. Breakthrough curves for cobalt and technetium were fitted using a five-parameter curve-fitting routine. Tritium breakthrough curves for unsaturated sediments were found to exhibit a nonlinear, skewed breakthrough pattern that was similar but retarded with respect to the technetium breakthrough curves. At water contents between 44 and 52% saturation, recovery of tritium, cobalt and technetium averaged 98%, and recovery of strontium ranged from 80 to 90% while recovery of iodine was less than 60%, ranging from 10 to 55%. Column sorption of iodine was found to increase with time.

Conductivities for water in unsaturated soil were found to be dependent on soil density. This suggests that laboratory-measured conductivities must be corrected to account for observed density variations between laboratory and field samples.

Preliminary tests of downwell detection of radioisotopes were begun in the laboratory in anticipation of tracer placement in the field. A downwell simulator was

constructed which allowed for monitoring of both water content and radionuclide transport.

Field monitoring continued. Soil drying at the 1.5-m depth in response to upward thermal gradients was observed even though surface moisture increased due to rain and subsequent snow cover. A second weighing lysimeter and associated electronics were purchased, and installation at the field site was begun. This unit will be used to investigate the effects of increased precipitation on water and radionuclide transport.

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

Unsaturated column flow tests continued. Steady flow rates of 6.8 0.1 cm/day were maintained using a flow control system described previously (Platt 1978). Soil columns were leached with salt solutions spiked with one or more tracers. Table 10.1 describes the series of tests conducted and the percent of radioisotope recovery for each test. Recovery was 98 to 99% for cobalt and tritium. Although not shown in Table 10.1, technetium recovery was similar. Percent recovery was between 80 and 90% for strontium and less than 60% for iodine and the high-salt solution. In

TABLE 10.1. Tracer Flow Studies in Soil B at Several Saturation Levels

Column	Solution*	Tracer	Column Saturation, %	Pulse Period, days	Pulse/Pore Volume Ratio, ml/ml	Water Velocity, cm/day	Tracer Recovery, %			
							I	Sr	Co	H
1	Low	I, Sr, Co	44.1	3.00	4.31	6.8	7	--	98	--
	High		45.1	--	--	6.8	11	82	--	--
	High	I	44.9	3.18	4.50	6.9	10	--	--	--
	High	Sr, Co	44.5	2.99	4.17	6.7	--	89	99	--
	High	³ H	44.5	0.95	1.31	6.7	--	--	--	98
2	High	I	50.8	2.52	3.04	6.8	54	--	--	--
	High	Sr, Co	52.1	2.99	3.63	6.8	--	89	98	--
	High	I, Sr, Co	52.5	3.04	3.73	6.8	46	90	90	--

* Low (salt solution) - 0.15M NaNO₃, 0.01M KNO₃, 0.002M Ca(NO₃)₂.

High (salt solution) - 0.15M NaNO₃, 0.15M KNO₃, 0.01M, Ca(NO₃)₂.

the low salt test the percent recovery was near zero for strontium after 12 pore volumes. Subsequent leaching with high salt solution caused an immediate flush of strontium. Strontium concentrations after 2 pore volumes of high salt exceeded initial concentrations by about 10%, and then decreased to zero after 17 additional pore volumes. Further tests with strontium indicated that breakthrough of the strontium pulse occurred after 4 pore volumes, with the peak occurring after 8 pore volumes. A computed column K_d for the high-salt solution on B soil was 0.53, which is somewhat lower than the measured batch K_d (1 ± 0.2) previously reported for strontium in the high-salt solution.

Analysis of breakthrough curves continued. A five-parameter curve-fitting routine was used to obtain the necessary coefficients to describe the dispersion of tracer in unsaturated soil. The convective-dispersive transport equation was modified to account for immobile water fractions. The necessary coefficients generated from the curve-fitting routine are as follows:

$$R_m = 1 + \frac{\rho f K_d}{\theta_m} = \text{retardation factor}$$

where ρ = soil density (g/ml)

f = fraction of soil volume

associated with mobile water

θ_m = mobile water content (ml/ml)

K_d = distribution coefficient (ml/g)

$\bar{\alpha} = \alpha L/q$ = dimensionless mass

transfer

where α = mass transfer coefficient
(1/day)

L = column length

q = volumetric flux (cm/day)

$P = v_m L/D$ = Peclet number

where v_m = average pore water velocity in
dynamic region (cm/day)

D = dispersion coefficient
(cm²/day)

$$\beta = \frac{\theta_m R_m}{\theta R}$$

where $R = 1 + \frac{\rho K_d}{\theta}$

θ = volumetric water content.

Figures 10.1 and 10.2 show the observed and calculated breakthrough curves for cobalt-EDTA in soil B under conditions described for cobalt tracer in column 1 (Table 10.1). Best agreement between observed and calculated breakthrough curves was obtained with the five-parameter curve-fitting routine (Figure 10.2). This was

FIGURE 10.1. Breakthrough Curve for Co-EDTA in Soil B at 46% Saturation Using a Three-Parameter Model. Parameters R , P and pulse are described in text.

FIGURE 10.2. Breakthrough Curve for Co-EDTA in Soil B at 46% Saturation Using a Five-Parameter Model. Parameters are described in text.

also true for the technetium and tritium tracers tested. The fitting routine required estimation of the mobile water fraction, which was set at 0.9. K_d values generated from these data were in relative agreement with those obtained by batch method.

The significance of an error in K_d values at two field water contents is illustrated in Table 10.2. A change in K_d value from 0 to 1 decreases the estimated radionuclide transport velocity by more than an order of magnitude. At a K_d of 100, the nuclide velocity is reduced by more than three orders of magnitude below the rate for a nuclide with a K_d of zero. It should be noted that at flow rates associated with

normal sediment water content (0.07) little movement of water or radionuclide is expected (<0.1 cm/yr) even when the radionuclide is completely mobile. At normal drained sediment water contents (0.07), strontium is estimated to move at the rate of 7.7×10^{-3} cm/yr and 4.4×10^{-3} cm/yr for K_d values of 0.53 and 1, respectively. At these extremely low rates, the error involved in estimating column K_d data from batch K_d data is not significant. At higher soil water contents, produced by irrigation or increased precipitation, appreciable water and nuclide transport can occur. Table 10.2 indicates that for a water content of 0.14 (associated with a water suction head of 40 cm for soil B)

TABLE 10.2. Effect of K_d Variation on Estimated Flow Velocities Under Field Conditions at Hanford Reservation Burial Ground Sites

K_d	Water Content, cm/cm	Bulk Density, g/cm ³	Water Velocity, cm/day	Nuclide Velocity, ^(a) cm/day
0	0.07	1.80	3.1×10^{-4}	3.1×10^{-4}
0.5	0.07	1.80	3.1×10^{-4}	2.2×10^{-5}
1.0	0.07	1.80	3.1×10^{-4}	1.2×10^{-5}
1.5	0.07	1.80	3.1×10^{-4}	7.8×10^{-6}
2.0	0.07	1.80	3.1×10^{-4}	5.9×10^{-6}
5.0	0.07	1.80	3.1×10^{-4}	2.4×10^{-6}
10.0	0.07	1.80	3.1×10^{-4}	1.2×10^{-7}
0	0.14	1.80	1.82	1.8×10^0
0.5	0.14	1.80	1.82	2.4×10^{-1}
1.0	0.14	1.80	1.82	1.3×10^{-1}
1.5	0.14	1.80	1.82	9.0×10^{-2}
2.0	0.14	1.80	1.82	9.0×10^{-2}
5.0	0.14	1.80	1.82	1.4×10^{-2}
10.0	0.14	1.80	1.82	1.4×10^{-2}
100.0	0.14	1.80	1.82	7.0×10^{-4}
450.0	0.14	1.80	1.82	3.1×10^{-4}

a. $V_{\text{nuclide}} = V_{\text{water}} / (1 + K_d \rho/\theta)$.

the nuclide transport drops below 0.2 cm/yr only for nuclides with K_d values above 100. The estimated strontium movement is 84 cm/yr and 47 cm/yr for K_d values of 0.53 and 1, respectively. Under these circumstances K_d errors produce significant errors in transport.

Iodine-125 was sorbed to the soil column and under the imposed test conditions was never recovered from the column even after extensive leaching. Figure 10.3 shows the amount of iodine adsorbed on the soil column compared to that of cobalt and tritium. A time dependence in sorption was observed. The amount retained, after pulse application and flushing, increased to nearly 90% over a period of 60 days. In contrast, the tritium was still easily flushed after 118 days (2% retention).

Batch K_d measurements for ^{125}I indicated a variable amount of iodine sorption, dependent upon both salt concentration and possibly interacting ion effects (Table 10.3). The measurement error is large enough that there is no significant difference between tests. However, the mean values are largest for the ^{125}I alone and smallest when run with Co-EDTA. These differences appear real, but additional testing will be needed to verify the interaction effect.

Iodide sorption by soils have been observed previously. Walters and Winchester (1971) found a correlation between iodide content and soil organic matter. In contrast, Wildung et al. (1974) reported no statistical correlation between organic matter and iodide sorption. However, they did find a significant correlation between organic matter and methyl iodide. Iodide K_d values ranged from 0.8 to 53 in 22 soils using a 0.01M CaCl_2 solution (Wildung

et al., 1974). Soils with the lowest pH (4 to 6) had the highest K_d 's. The cause of the iodide sorption on soil B is not known at present. The soil is low in organic matter, and the clay mineralogy is such that anions like iodine should be excluded from the exchange sites. A microbial population buildup in time was possible, particularly since both salt solutions are nitrogen sources. Several additional K_d tests with solutions treated with phenol are planned.

Unsaturated Conductivity

Several methods were tested for evaluating unsaturated conductivity in soil B material. The method of Campbell (1974) and the method of Bresler et al. (1978) were compared against measured values. Figure 10.4 shows the comparison of methods. The Bresler method tends to underestimate the measured unsaturated conductivity at low water contents. The Campbell method was shown previously (Platt 1978) to be highly dependent upon the range over which the experimental coefficient, b , was determined. In these tests, the range for the determination of b , the slope of the log-transformed matrix potential versus water content curve, was 10 to 100 cm matrix potential. For soil bulk densities of 1.65 g/cm^3 and 1.80 g/cm^3 , the b values were 1.84 and 2.29, respectively. The two curves for the Campbell equation stay consistently within an order of magnitude and are in reasonable agreement with the experimentally measured values using methods of Rose (1968) and Klute (1972). However, the data suggest that density may significantly affect conductivity in the low water content range. The Campbell equation is based on a capillary model and has no physical meaning when

FIGURE 10.3. Amount of Isotope Retained in Soil After Initial Soil Wetting and Subsequent Nuclide Pulsing

TABLE 10.3. K_d Values for Soil B in Low and High Salt Solutions. Values run in triplicate.

Salt	I-125, ml/g	I-125 Simultaneously Run with Co-60 EDTA, ml/g
Low	18.5 ± 22.7	3.8 ± 3.4
High	3.9 ± 6	-1.6 ± 3.6

water flow occurs as liquid surface film and as a vapor. This is also true of the Bresler method. In addition, the Bresler method is based on infiltration-wetting characteristics and is highly dependent upon soil packing density. Despite the theoretical limitations and approximate nature of the developed relationships, it appears that unsaturated flow characteristics can be predicted with reasonable accuracy using relatively simple methods. Additional sediments, typical of burial ground materials, will need to be tested at several packing densities to assess the influence of texture and density on the predictive capability of the methods under investigation.

PHYSICAL MODELING - M. D. Campbell,
G. W. Gee and K. O. Schwarzmiller

Laboratory testing of wetting front movement and fingering was considered necessary before introduction of the nuclides to the field site. This conclusion was drawn from speculation that fingering in the wetting front could lead to nonuniform nuclide movement, which could result in erroneous conclusions about concentration at a given soil depth. A large column of soil was set up in the lab to test this possibility. Soil spiked with Co-EDTA was placed in a 2.5-cm layer, 60 cm below the soil surface. Water was applied in 5-cm increments and subsequent water content and isotope concentrations monitored. Upon satisfactory

FIGURE 10.4. Calculated and Measured Hydraulic Conductivities

completion of this test, radionuclides will be placed in the small caissons at the field test site to simulate field burial site transport conditions.

FIELD MONITORING - M. D. Campbell,
K. O. Schwarzmiller and G. W. Gee

Field data measurement systems have been described in previous reports (Platt 1978). One hundred six channels of data including air, soil temperatures, and air and soil humidities, wind speed and direction, and net solar radiation are presently being monitored on 2-hr intervals for use in developing appropriate cooperation and soil

water transport models for arid region burial sites. Rain during late summer and fall influenced soil water sensors down to 15 cm, but the sensors at 30 cm did not show moisture increase until after the record snowfall in late November. (Snow fell November 18 and 19, adding about 2.5 cm of water in the snowpack.) This snow drifted somewhat and probably did not deposit average snow on the weighing lysimeter. To make matters worse, no moisture was recorded by the rain gauge because it was emptied of snow by turbulent air. A further complication resulted from wind erosion of the sand from the lysimeter

and from the caissons. Following the melting of the snow, cold weather caused the topsoil to freeze. This resulted in frost heaving of the reference leveling ring and in reduction of the soil bulk density. Moderate redesign of the leveling ring will help resolve most of the resulting error problem, however.

As predicted by models of hydrothermal-linked transport, drying at lower depths in the weighing lysimeter occurred, as shown by resistance blocks. The magnitude of the observed transport will be reported with other data as recently initiated data analyses are completed.

Data analyses began in early December. The first segment of data all collected on printed paper tape and required hand tabulation preceding analyses. The data

logging on digital cassettes began August 21, 1978. These data cassettes are read by a Texas Instrument 733 terminal into a computer for analysis.

Initially, one weighing lysimeter was installed at the field site. Despite earlier difficulties, this weighing lysimeter has provided valuable and unique information about water reception and storage at Hanford. Because this information is essential in defining nuclide burial site characteristics and in modeling site mass-energy budgets, another weighing lysimeter has been procured and site installation design completed. The installation contract was awarded to J. A. Jones Construction Company, and installation has started.

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11.0 DETECTION AND CHARACTERIZATION OF MOBILE ORGANIC COMPLEXES OF FISSION PRODUCTS

J. A. Franz - Project Manager

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

SUMMARY

The sorption characteristics of EDTA and DTPA complexes of zirconium on Hanford soil were examined. The K_d values vary from 4 to 8 over a wide concentration range (10^{-5} to 10^{-8} M), confirming the expected potential mobility of these complexes.

During this quarter effort continued in developing convenient characterization techniques for ^{99}Tc organic complexes. Tc(IV) and Tc(V) complexes were prepared, and ^{13}C , ^1H , and ^{99}Tc FTNMR* characterization of a variety of complexes was attempted. The ^{99}Tc resonance of $\text{Tc(VII)} (\text{NH}_4\text{TcO}_4)$ was detected at 17.90 MHz. Other oxidation states appear to be paramagnetic and were not observable.

IDENTIFICATION OF COMPLEXING AGENTS AND NUCLIDES

Existing data on analysis of waste repositories were augmented by recently available data on inventories of Maxey Flats low-level waste.

* Fourier transform nuclear magnetic resonance.

STUDY OF MOBILITY CHARACTERISTICS OF ORGANIC COMPLEXES

Previous work with tartrate and gluconate complexes of zirconium was supplemented with an examination of the soil absorption behavior of zirconium complexes; ethylenediaminetetraacetic acid (EDTA) and diethylenetriaminepentaacetic acid (DTPA) were used. Solubility determinations for EDTA and DTPA complexes revealed their maximum solubility at room temperature and at pH values of calcareous soil (8.3) to be 5×10^{-4} and 1×10^{-3} M. Batch K_d values were determined in standard fashion by spiking solutions of zirconium nitrate at $\text{pH} < 1.5$ with ^{95}Zr , shaking with MnO_2 to remove ^{95}Nb , and reaction with the desired complex, after which the pH of the medium was increased to ambient levels. The complexes exhibited ^1H NMR spectra essentially identical to those of the uncomplexed ligands. K_d values were calculated from

$$K_d = \frac{[(A_{\text{soln}})/(A_{\text{soil}})] \times [(ml_{\text{soln}})/(g_{\text{soil}})]}{(g_{\text{soil}})}$$

where A_{soln} = activity of Zr in solution,
 A_{soil} = activity of Zr on soil, and
 ml_{soln} = the volume of solution
 contacting a specified
 weight of soil (g soil).

Table 11.1 provides soil Kd values for Zr-EDTA and Zr-DTPA. Figure 11.1 depicts the variation of Kd values over a wide concentration range. The Kd values are approximately independent of concentration range except at 10^{-4} M, which is approaching saturation of the complexes.

It is clear from these results that the strong chelating agents EDTA and DTPA significantly increase the potential mobility of zirconium.

Efforts to achieve convenient methods for characterizing ^{99}Tc oxidation states were partly successful. The main effort centered on detection of ^{99}Tc by FTNMR techniques. Technetium-99 as Tc(VII) is a diamagnetic nucleus with a nuclear spin of 9/2 and associated quadrupole moment. The ^{99}Tc resonance of NH_4TcO_4 was located at 17.90 MHz. Figure 11.2 shows the ^{99}Tc FTNMR resonance from a single 90° pulse of a solution of 99 mg of NH_4TcO_4 in 5 ml of D_2O . The

resonance exhibits a full width at half height ($\Delta\nu_{1/2}$) of 10 Hz. From $T_2^* = \frac{1}{\pi\Delta\nu_{1/2}}$ the relaxation times T_1 and T_2 are 3.2×10^{-2} sec, consistent with a dominant quadrupole relaxation process.

Observations of ^{99}Tc NMR resonances of Tc(V) [as well as Tc(IV)] were attempted. Tc(V) citrate, Tc(IV) citrate and Tc(V) thiocyanate complexes were examined by ^{13}C , ^1H and ^{99}Tc NMR. In every case no observable resonances were detected despite ample concentrations of the nuclide. These results indicate that Tc(V) exists as a paramagnetic species not amenable to this analytical method. Thus the NMR technique appears limited to the (VII) oxidation state. Synthetic methods for technetium complexes are now rather well established. Future work with EDTA-type complexes for different oxidation states of technetium in soil studies is anticipated.

TABLE 11.1. Soil Kd Values for Zr-EDTA and Zr-DTPA

pH	Time, hr	Ligand	Ligand Concentration, moles	Zr Concentration, moles	Soil Quantity, g	Initial counts/min	Final counts/min	Kd
8.0	24	EDTA	1×10^{-4}	1×10^{-4}	4.06	111,200	11,900	10.26
7.2	24	EDTA	1×10^{-5}	1×10^{-5}	3.94	47,800	14,140	3.02
7.4	24	EDTA	1×10^{-6}	1×10^{-6}	4.00	40,800	10,060	3.81
7.0	24	EDTA	1×10^{-7}	1×10^{-7}	4.09	50,400	7,000	7.57
7.6	24	EDTA	1×10^{-8}	1×10^{-8}	3.97	38,400	5,460	7.58
7.5	24	DTPA	1×10^{-4}	1×10^{-4}	4.03	88,600	8,240	12.09
7.6	24	DTPA	1×10^{-5}	1×10^{-5}	4.11	58,000	10,380	5.58
7.8	24	DTPA	1×10^{-6}	1×10^{-6}	4.03	53,400	8,940	6.16
7.5	24	DTPA	1×10^{-7}	1×10^{-7}	4.02	43,200	6,420	7.12
7.2	24	DTPA	1×10^{-8}	1×10^{-8}	4.00	31,200	7,440	3.99

FIGURE 11.1. Variation of K_d Values for Sorption of EDTA (1), DTPA (2), and Gluconate (3) Complexes of Zirconium on Hanford Soil

FIGURE 11.2. ^{99}Tc FTNMR Spectrum of NH_4TcO_4 at 17.90 MHz

12.0 ELECTROPOLISHING FOR SURFACE DECONTAMINATION OF METALS

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

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

SUMMARY

Plutonium operation of the recently completed stainless steel walk-in hood was initiated, and a number of sectioning and pretreatment studies using plutonium-contaminated components were conducted. Various mechanical sectioning techniques were used to cut more than 500 ft² of plutonium-contaminated material into test pieces for subsequent vibratory finishing and electropolishing studies. A heavy-duty shear-type nibbler effectively cut up to 0.25-in. thick stainless steel plate at a rate of 6 ft/min. However, this tool's maneuverability is limited.

The vibratory finisher was installed in the sectioning/pretreatment facility and was evaluated using plutonium-contaminated material and both ceramic media and a low-wear-rate steel medium. These tests demonstrated vibratory finishing's ability with either type of media to rapidly remove gross plutonium contamination and leave an essentially nonsmearable surface ideal for subsequent handling and final decontamination by electropolishing. Although the steel media do not decontaminate to as low a level as the ceramic media, they have the substantial advantage of producing no secondary waste because of media wear. These tests also demonstrated the ability of vibratory

finishing to remove plutonium contamination from nonmetallic (epoxy-coated) surfaces.

A contact-type in situ electropolishing device was developed and used to decontaminate a 5-ft² area inside a plutonium-contaminated glove box. The electropolishing treatment reduced the contamination level of the stainless steel floor from 1100 nCi/g to 0.2 nCi/g. In comparison, efforts to decontaminate the floor using conventional scrubbing techniques could not reduce the level below 470 nCi/g.

The relative resistance of electropolished surfaces and other representative surface finishes to contamination in a typical nuclear service environment was evaluated. The contamination level of electropolished stainless steel exposed in a BWR fuel transfer channel during refueling operations was substantially lower than that of the other surface finishes tested, demonstrating the value of prepolishing surfaces intended for nuclear service.

DECONTAMINATION STUDIES

Cleanup operations from the nitric acid evaporation occurrence have been completed, and the electropolishing facility is back in full operation. Additional safety circuits have been added, and all sensors, including

those for air flow, tank temperature, leakage, and tank overflow, are now inter-connected in such a way that failure of any one system shuts off total power and water to the entire electropolishing facility. All equipment needed for handling future out-of-bounds or emergency situations has been assembled and stored in special canisters adjacent to the electropolishing room. In addition, detailed emergency procedures have been prepared and included with the standard operating procedures for the decontamination facilities to cover any abnormal operating conditions.

PRETREATMENT STUDIES

The recently completed sectioning/pretreatment facility (Figure 12.1) is now in full operation using plutonium-contaminated components. The facility appears to be functioning

as designed. Any airborne contamination generated during operations with heavily contaminated components is rapidly reduced to background levels by the facility ventilation system. The contamination that settles on the polished stainless steel walls and floor is readily removed, when required, by hand rubbing with damp rags.

Various sectioning techniques have been used to cut more than 500 ft² of plutonium-contaminated pipe, ducting and glove box components into pieces suitable for further processing in the vibratory finisher and/or the electropolishing system. The stainless and mild steel pipe (4-, 5-, and 8-in. dia) was cut into 1.5-ft lengths using clamp-on type mechanical saws as shown in Figure 12.2. The ducting and thinner glove box components (≤ 0.125 -in. thick) were sectioned using a punch and die

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FIGURE 12.1. Stainless Steel Walk-In Hood Used for Sectioning and Pretreatment Studies

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FIGURE 12.2. Clamp-On Type Mechanical Saw Used to Section Pipe and Ducting Inside Walk-In Hood Shown in Figure 12.1. Note the use of polished stainless steel on walls and floor to facilitate decontamination of the Walk-In Hood.

type nibbler. The thicker glove box components were sectioned using a new heavy-duty shear-type nibbler that can cut stainless steel 0.25 in. thick at speeds up to 6 ft/min. This nibbler was also used to cut some of the 8-in.-dia pipe longitudinally to permit evaluation of this geometry in the vibratory finisher. Although the nibblers are fast and produce a waste form that can be decontaminated easily, they are difficult to maneuver and are restricted primarily to straight cuts in relatively planar material.

The vibratory finisher and associated sludge tank were installed in the sectioning/pretreatment facility for further evaluation tests using beta-gamma-contaminated items from N Reactor and plutonium-contami-

nated glove box pieces from Hanford and Rocky Flats. Most of these tests were conducted using a low-wear-rate steel burnishing medium rather than the ceramic-type deburring media used in the previous⁽¹⁾ vibratory finisher studies. The results of these tests were very encouraging. The steel medium was very effective as a pretreatment technique in rapidly removing grease, rust, and both latex and enamel type paint. In addition, its burnishing action was almost as effective as the ceramic media's cutting action in removing most of the gross surface contamination. More importantly, the surfaces were cleaned and decontaminated with essentially no secondary waste generation from wear of the steel medium.

The beta-gamma-contaminated mild steel fuel spacers, process tube end caps, and process tube inserts were tested by vibratory finishing in the steel medium for a total of 10 hr using a commercial liquid compound. This test (Table 12.1) showed that average surface contamination levels of the components were substantially reduced and that most of this reduction occurred during the first hour of processing. Moreover, even after only 1 hr of processing, all components were either nonsmearable or had such low levels of smearable contamination (<1000 counts/min) as to substantially facilitate subsequent handling and final decontamination operations.

The effect of free abrasive on the decontamination capabilities of the steel medium was investigated by adding 1 kg of AlO_2 abrasive at the end of the 10-hr run and processing for an additional hour. The fixed contamination levels were further reduced by about 60%, but the resulting secondary waste was an extremely fine powder very difficult to handle. It should be noted that although the steel medium did not generate any secondary waste, it did become slightly contaminated (Table 12.1). However, this contamination was reduced to

insignificant levels by the same self-cleaning action observed previously for the ceramic-type media.

The first test of the vibratory finisher with plutonium-contaminated material was conducted using four pieces of stainless steel from Hanford glove boxes and two pieces of stainless steel (coated on one side with epoxy usually difficult to decontaminate) from Rocky Flats glove boxes. The results (Table 12.2) show that the vibratory finisher with the steel medium is also very effective in removing plutonium contamination. As in the case of beta/gamma-contaminated material, most of the decontamination occurred within the first hour of processing and included complete elimination of the smearable contamination. Moreover, the decontamination process was equally effective for the epoxy-coated surfaces, illustrating the ability of vibratory finishing to decontaminate selected non-metallic transuranic waste.

A second plutonium test of the vibratory finisher and the steel medium was conducted using 19 stainless steel Hanford glove box pieces to verify the initial test results and to obtain decontamination rate data for shorter time periods. The results are shown in Figure 12.3 for 60 min of processing in

TABLE 12.1. Decontamination of Beta-Gamma-Contaminated Components Using Vibratory Finisher with Steel Medium

Component	Fixed Contamination, mrad/hr							
	Initial		After 1 hr		After 2 hr		After 10 hr	
	Outside	Inside	Outside	Inside	Outside	Inside	Outside	Inside
Fuel spacer	800	10,000	20	100	25	75	15	70
Fuel spacer	300	1,500	35	45	25	25	20	80
End cap	200	5,000	20	700	10	100	10	120
End cap	200	1,000	10	30	10	28	8	15
Insert	500	-	300	-	250	-	200	-
Insert	150	-	100	-	130	-	100	-
Steel medium			2,000 counts/min		1,000 counts/min		400 counts/min	

TABLE 12.2. Decontamination of Plutonium-Contaminated Glove Box Sections Using Vibratory Finisher with Steel Medium

Material/Source	Surface Contamination, dis/min 100 cm ²					
	Initial		After 1 hr		After 10 hr	
	Contact	Smear	Contact	Smear	Contact	Smear
Stainless steel/Hanford	62,000	38,000	3,800	Nondetectable	2,500	Nondetectable
Stainless steel/Hanford	88,000	25,000	8,800	Nondetectable	2,500	Nondetectable
Stainless steel/Hanford	69,000	19,000	3,800	Nondetectable	2,500	Nondetectable
Stainless steel/Hanford	750,000	94,000	7,500	Nondetectable	7,500	Nondetectable
Epoxy/Rocky Flats	125,000	50,000	5,600	Nondetectable	Background	Nondetectable
Stainless steel/ Rocky Flats	69,000	13,000	10,000	Nondetectable	3,800	Nondetectable
Epoxy/Rocky Flats	250,000	3,000	11,000	Nondetectable	700	Nondetectable
Stainless steel/ Rocky Flats	75,000	13,000	2,500	Nondetectable	15,000	Nondetectable

FIGURE 12.3. Contamination Level Versus Time for Plutonium-Contaminated Glove Box Pieces Vibratory Finished for 60 min in Steel Medium Followed by 60 min in Ceramic Medium

the steel medium followed by an additional 60 min of processing in a fast-cutting ceramic medium. The average contamination level of the 19 samples dropped from 270,000 dis/min·100 cm² to 26,000 dis/min·100 cm² in the first 6 min of processing

in the steel medium, a decrease of 90% in contamination level. The samples also were essentially nonsmearable after this brief treatment in the steel medium. The contamination level continued to decrease with

additional processing time, but at a substantially lower rate. After 60 min of processing in the steel medium, the fixed contamination level was 11,000 dis/min·100 cm², representing removal of 96% of the initial contamination. Subsequent processing of the same samples in the fast-cutting ceramic medium (Figure 12.3) reduced the contamination level by an additional 2% to 4500 dis/min·100 cm², with most of this decrease occurring during the first 15 min of vibratory finishing.

As a final comparison test, an additional 17 stainless steel glove box pieces were processed for 1 hr in the fast-cutting ceramic medium. Eleven of these test pieces, from Rocky Flats glove boxes, had one side coated with epoxy. Pretreatment in the vibratory finisher reduced the average contamination level of the stainless steel surfaces from 250,000 dis/min·100 cm² to 3400 dis/min·100 cm² and that of the epoxy-coated surfaces from 196,000 dis/min·100 cm² to 1400 dis/min·100 cm². As before, the vibratory-finished surfaces were essentially nonsmearable.

In summary, these tests show that vibratory finishing techniques using either ceramic or steel media are capable of rapidly removing gross plutonium contamination from both metallic and selected nonmetallic (epoxy) surfaces. Although the ceramic media can decontaminate to lower levels, the steel media have the substantial advantage of producing no secondary waste because of media wear. Both types of media also produce surfaces essentially nonsmearable and ideal for subsequent handling and final decontamination by electropolishing.

IN SITU DECONTAMINATION TECHNIQUES

The contact-type in situ electropolishing device described in a previous report⁽²⁾

has been further developed into a small, portable system and used to decontaminate a 5-ft² area on the floor of a heavily-contaminated plutonium glove box. The initial reading on the glove box floor as measured using a silicon-surface-barrier alpha detector⁽³⁾ was 900,000,000 dis/min·100 cm². Electropolishing using the contact in situ device (Figure 12.4) reduced the contamination level to only 200,000 dis/min·100 cm². This represents a change from 1100 nCi/g to 0.2 nCi/g for the 0.48-cm-thick stainless steel plate. For comparison, previous decontamination efforts using conventional scrubbing techniques could reduce the contamination level only to 400,000,000 dis/min·100 cm².

PREPOLISHING APPLICATIONS

Previous studies have shown that electropolished surfaces are more easily decontaminated using conventional decontamination methods than are metal surfaces with standard as-received finishes. This has been attributed to the removal by the electropolishing process of the microscopic surface imperfections capable of entrapping and retaining contamination.⁽¹⁾ Contamination tests conducted in a BWR fuel transfer channel during refueling operations have now confirmed that electropolished surfaces also are more resistant to contamination by radionuclides than are other surface finishes under identical service conditions. The results of tests comparing 18 electropolished 304 stainless steel samples with equivalent samples representing standard surface finishes are presented in Table 12.3. The electropolished surfaces were substantially less contaminated by the refueling operation than were the other finishes. Subsequent treatment by a high-pressure water spray reduced the levels

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FIGURE 12.4. Contact-Type In Situ Electropolishing Device Used to Decontaminate 5 ft² Area on the Floor of a Plutonium Glove Box

TABLE 12.3. Comparison of Contamination Levels for Electropolished and As-Received Surface Finishes Exposed in a BWR Fuel Transfer Channel During Refueling

Surface Finish	Contamination Level, counts/min	
	After Exposure During Refueling	After Decontamination with High-Pressure Water Spray
Electropolished	3,000-6,000	1,500
Superfinish	40,000	20,000
2B sheet	40,000	20,000
No. 1 finish bead blasted	15,000	7,500

even further. It produced a final contamination level for the electropolished surfaces lower than the minimum achievable levels for the other finishes by a factor of 5 to 13.

This further demonstrates the value of pre-polishing surfaces that will be contaminated in their normal service environment.

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