
**Nuclear Waste Management
Quarterly Progress Report
October Through December 1977**

**Compiled by
A. M. Platt**

May 1978

Pacific Northwest Laboratory
Richland, Washington 99352
Operated for the
U.S. Department of Energy
by



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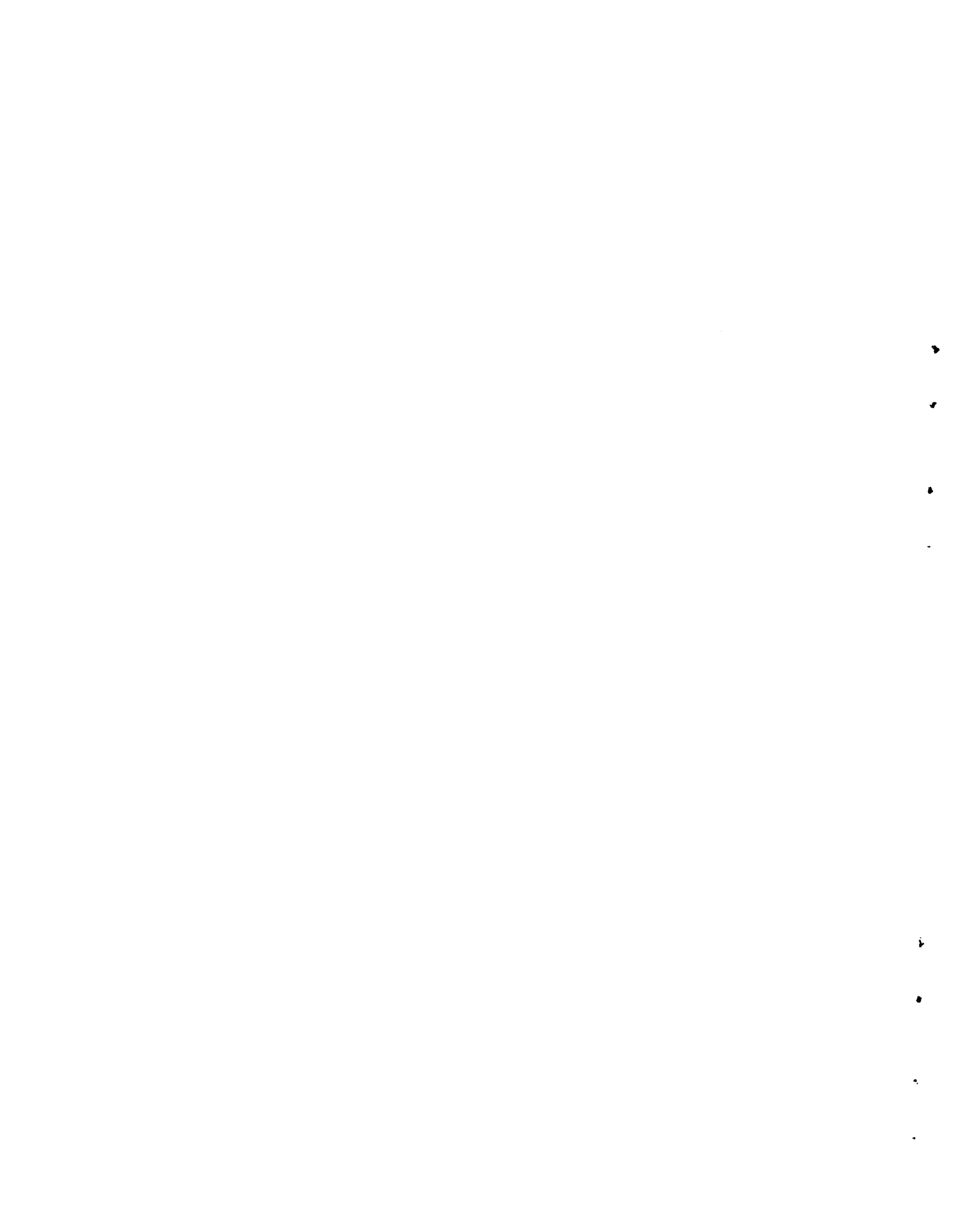
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NUCLEAR WASTE MANAGEMENT
QUARTERLY PROGRESS REPORT
OCTOBER THROUGH DECEMBER 1977

Compiled by
A. M. Platt
Nuclear Waste Technology Program

May 1978

BATTELLE
Pacific Northwest Laboratories
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 Battelle, Pacific Northwest Laboratories. These programs were funded by the Waste Management, Production and Reprocessing Division of ERDA. The Commercial Radioactive Waste Fixation Program is reported elsewhere. These reports are issued on a quarterly basis.

The prior report in this series is Nuclear Waste Management Quarterly Progress Report July Through September 1977, BNWL-2377-3. As of October 1, 1977, Pacific Northwest Laboratories changed the designation for reports from BNWL to PNL. Thus, the next report to be issued in this series will be PNL-2378-1.



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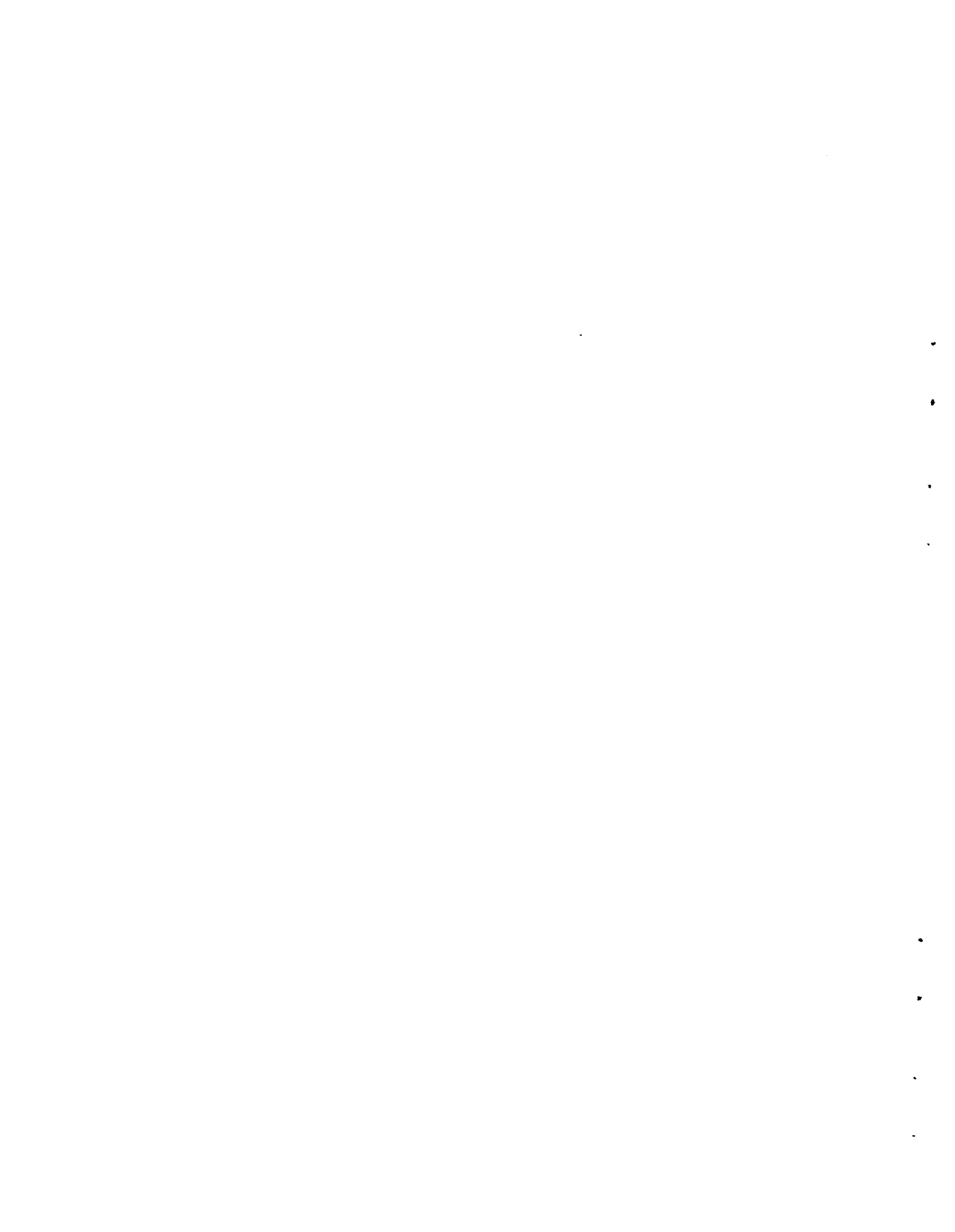
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1.0 DECONTAMINATION AND DENSIFICATION
OF CHOP-LEACH CLADDING RESIDUES

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

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

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

SUMMARY

Equipment for the pilot-scale chemical decontamination prototype^(1,2) is being fabricated. The prototype will process chop-leach cladding hulls at a rate of ~2 kg/hr with an HF-Ar decontamination step followed by an aqueous rinse. The prototype is scheduled to be installed early in CY-78 for nonradioactive demonstration.

The Inductoslag melting furnace,⁽³⁾ which is the melt densification half of the prototype fuel hull decontamination-densification equipment, was installed for nonradioactive testing. An initial test melt of a billet of iron-base alloy CG27 was stopped when a bridge was observed. Subsequent

examination showed that a metal run-out had occurred because not enough flux had melted to form the necessary seal between the crucible wall and the starting billet. The run-out may have contributed to forming the bridge. Additional tests are being planned to determine if the 3 kHz power source will properly start the 10-cm molten pool. Operating at full power, the melting furnace is frequency-limited. A small water leak in one crucible segment also occurred during the test because metal chips in crucible segments blocked water coolant flow, resulting in a melt-through. A water filter will be added at the crucible's coolant inlet line to prevent any accumulation of trash.

The Inductoslag melting furnace will undergo nonradioactive testing with the chemical decontamination prototype in a remotely operated hot cell mockup. Following successful testing, the furnace and chemical decontamination equipment will be installed in a hot cell for demonstration with contaminated chop-leach fuel hulls.

For the densification task, methods and materials for the recasting of Zircaloy are being evaluated. Tests indicate that molten Zircaloy-4 has poor flow characteristics compared to titanium.

DECONTAMINATION PROCESS

Pilot-scale chemical decontamination equipment capable of processing chop-leach hulls at a rate of ~ 2 kg/hr has been described in previous quarterly reports.^(1,2) The HF reactor and aqueous washer components are being fabricated. The chemical makeup and spent liquid holding tanks for this decontamination prototype were designed, and a contract was awarded for fabrication. The equipment is scheduled to be installed with the Inductoslag ingot melting furnace⁽³⁾ early in CY-78 in a simulated hot cell configuration. After nonradioactive demonstration, the decontamination and melt densification equipment will be transferred to a hot cell for test processing of radioactive contaminated fuel hulls. Hot cell equipment layouts were prepared to establish design details for piping and electrical service, instrumentation, equipment controls,

and cell wall penetrations. All equipment and support hardware is being designed to facilitate remote assembly and disassembly. An extension of the 306 Building scrubbed ventilation system is being engineered to remove any HF leakage from the chemical decontamination prototype.

DENSIFICATION PROCESS

The Inductoslag ingot melting furnace,⁽³⁾ which was designed for the melt densification of chop-leach cladding residues, has been installed at the Pacific Northwest Laboratory. The furnace has been undergoing check-out tests prior to planned nonradioactive demonstrations.

Initial testing of the Inductoslag melting furnace showed that a 16-turn induction coil made from flattened 1.3-cm dia (0.5 in.) tubing was required to achieve power sufficient to melt test metals. Operating at maximum power with all capacitance, the equipment was frequency limited. For sustained full-power operation, an additional capacitor may be required.

The furnace was tested using a billet of CG27, which is a high-temperature iron-base alloy (melting point, $\sim 1370^\circ\text{C}$), and calcium fluoride flux (melting point, 1360°C). The furnace chamber was evacuated to ~ 79.9 Pa (0.6 torr) and backfilled with argon to 73.3 kPa (550 torr). Power indications during testing are shown in Table 1.1.

The melt was terminated because a bridge had formed. Examination of the ingot revealed that 2.9 kg

TABLE 1.1. Record of the Initial Test in Inductoslag Furnace Startup

Elapsed Time, min	Power, % of 200 kW	Frequency, % of 3 kHz	Voltage, % of 840	Remarks
0	20	70	70	Power on
4	30	78	60	Increased power, melting
7	40	83	63	
11	45	84		
13	50	84		
15				Power off

(6.5 lb) of metal had been melted, most of which had run down the space between the starting billet and the crucible wall; 0.6 kg (1.4 lb) had dripped onto the catch basin below. Very little if any of the calcium fluoride flux had melted. The melting point of the calcium fluoride was apparently too near the melting point of the billet; not enough flux melted to form a seal between the crucible wall and the starting billet.

The metal run-out was not detected during the 15-min test, but only after the furnace had been opened. The bridge formation, which stopped the test, could have resulted from a run-out occurring before a full molten pool was achieved, thus leaving the bridge in place with insufficient molten metal for coupling.

Control circuitry has been altered within the power source so that a sudden drop in voltage should be indicated at the time a run-out occurs.

There is also some possibility that the 3 kHz-power source does not sufficiently heat the outer edges of the pool. All previous 10-cm-dia (4 in.) ingot melting has been performed with a 10-kHz power source.

Further testing is needed to determine the frequency requirements for properly starting the molten pool.

It was also noted that a small water leak had occurred during the test in one of the crucible segments. Disassembly of the crucible revealed that at least 10 of 24 segments contained metal chips and other trash. The metal chips blocked the water flow, and the crucible segment overheated, melting through to create the water leak. This event is not related to the metal run-out or to a lack of slag melting.

The crucible was disassembled, thoroughly cleaned, and sent to the U.S. Bureau of Mines for repair. A water filter will be installed on the crucible's coolant inlet line to prevent any trash accumulating in the crucible segments. The next test will use a mild steel billet (melting point, $\sim 1500^{\circ}\text{C}$) and calcium fluoride flux.

In ongoing densification work, materials and methods are being evaluated for casting Zircaloy chop-leach cladding material into near-finished reusable shapes. A patented investment molding technique called

Monograf Mold[®] uses a wax pattern with built-up outer layers 0.25 to 0.5 cm (0.1 to 0.2 in.) thick of carbonaceous shell. An advantage of this process is that disposal of the once-through materials could be simplified by burning.⁽⁴⁾ The results of burning crushed and uncrushed Monograf molding material at 950°C are shown in Table 1.2.

Four hours' burning at 950°C appears to be about optimum for the crushed material. It yields a weight reduction of 88% and a volume reduction of 93%, with a resulting residue density of 1.5 g/cm³. Uncrushed material takes about 6.5 hr at 950°C for an optimum weight loss of 95% and a residue density of 0.86 g/cm³. No reasonable explanation is available for the large difference in residue densities between the crushed and uncrushed materials.

Because little is known about molten Zircaloy's fluidity, pure zirconium and Zircaloy-4 were tested in work contracted to the U.S. Bureau of Mines in Albany, Oregon. These tests used the Consumable Electrode Skull melting process. Ten-cm-dia (4 in.) electrodes were melted to a 12.7-cm-dia (5 in.) crucible using ~35 volts at an absolute furnace pressure of ~2.27 Pa (~0.017 torr). Since this process is commonly used in industry for making titanium castings, these test results can be compared with those of similar tests made with titanium. The mold cavity of the spiral test is a 1.3-cm (0.5 in.) half-round groove machined in a horizontal graphite plate. Molten metal flows into the mold cavity from a centrally located down sprue which feeds the cast spiral with a constant metallostatic head.⁽⁵⁾

[®] Patented graphite mold of the Misco Corporation.

TABLE 1.2. Burning Tests of Monograf[®] Molding Material at 950°C

Time, hr	Crushed Material (5-g samples, ~20 mesh)			Uncrushed Material (11- to 20-g samples, max ~2 in.)		
	% Weight Reduction	% Volume Reduction	Residue Density, g/cm ³	% Weight Reduction	% Volume Reduction	Residue Density, g/cm ³
0			0.83			
1	68	58	0.64			
2	82	85	1.09			
4	88	93	1.50	66		0.76
5.5				80		0.72
6				93		0.62
6.5				95		0.86
7	88	93	1.50			
17	90	93	1.45			
24	82	92	1.84			

Table 1.3 shows the results of these tests along with previous results⁽⁵⁾ obtained from pure titanium and the Ti-6 Al-4V alloy. These results show that pure zirconium and Zircaloy-4 are over 60% less fluid than pure titanium and the Ti-6Al-4V alloy. The poor fluidity of Zircaloy-4 will be an important factor in part selection and casting design.

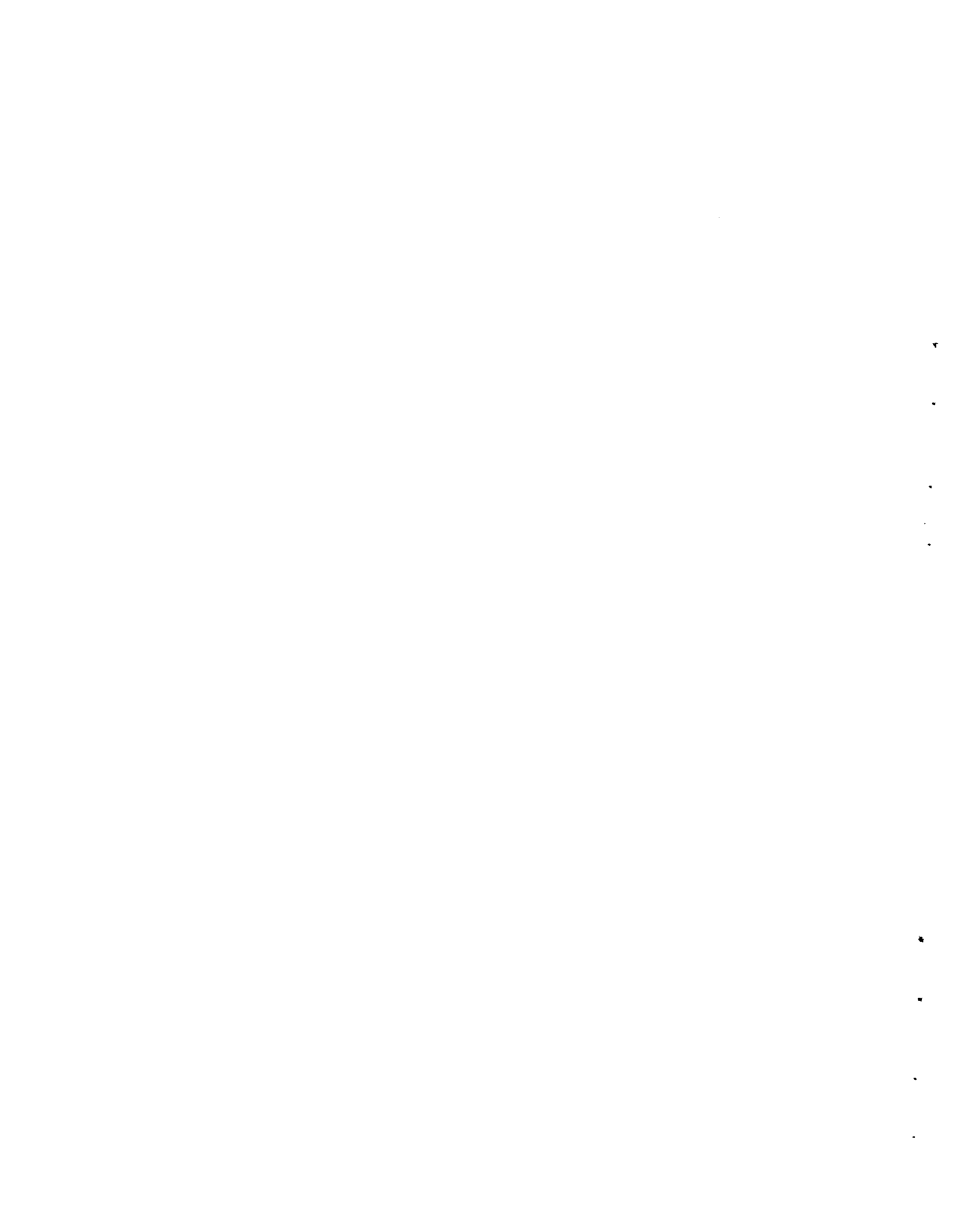
TABLE 1.3. Fluidity Tests

	<u>Length of Spiral, mm</u>
Pure Zircaloy	432
Zircaloy-4	419 and 425
Pure Ti ^(a)	1054 to 1181 (range for 4 tests)
Ti-6 Al-4V ^(a)	1105 and 1143 (2 tests)

a. Data supplied by E. D. Calvert, Albany Metallurgical Research Center, U.S. Bureau of Mines, U.S. Department of the Interior, Albany, Oregon.

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

R. L. Goles - Project Manager

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

SUMMARY

During this reporting period, a laboratory mockup of the calcine process off-gas stream was built and used to assess the absolute collection efficiencies of selected filters. Design of a direct-inlet mass spectrometer (DIMS) has been completed, fabrication started, and components ordered. Also a sampling system has been designed to characterize ^3H - and ^{14}C -bearing compounds in gaseous process streams. Physical and chemical trapping agents required for this differential sampling system have been received and are currently being tested.

PARTICLE GENERATION AND COLLECTION - H. G. Rieck, Jr., and D. C. Hamilton

The particle generator, designed and assembled for measuring filter efficiency, was relocated in laboratory facilities compatible with radioactive tracer studies. A laboratory mock-up system designed to simulate the calcination process off-gas stream was built and used to assess the absolute collection efficiencies of selected filters. The jet mill

assembly was modified, resulting in improved submicron particle generation capabilities.

Figure 2.1 shows a schematic of the apparatus designed and assembled to simulate conditions anticipated in the calcination process off-gas stream. Here submicron particles generated from synthetic calcine are introduced into a 1-1/2-in. ID process line via an 88-mil ID pitot tube. The process line transfers the submicron particulates to a test filter located downstream of this apparatus. A positive displacement pump is used to maintain maximum expected gas velocities within the mock-up process line.

During this quarter a new jet mill assembly has been designed and built as a result of our recent research in particle generation. The new design, more than the previous one, emphasizes grinding through impaction of the particles off chamber walls and off other particles. This has greatly reduced the average size distribution of particles generated by this assembly. A particle size analysis of the complete laboratory generation, delivery, and collection system was conducted using a scanning electron

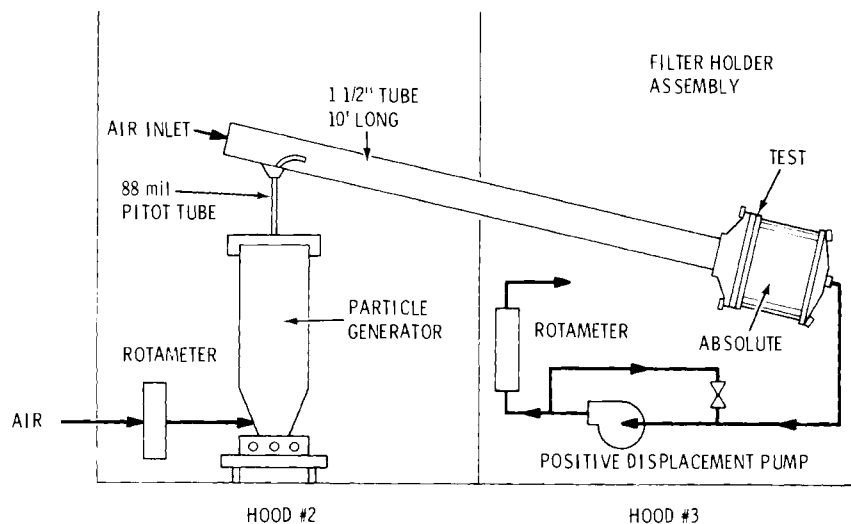


FIGURE 2.1. Simulated Process Line for Filter Efficiency Measurements of Calcine

microscope. Based upon a sample containing 1418 particles, the average particle diameter was 0.13μ , with a geometric deviation of 2.4. There was an excellent qualitative fit to a log-normal distribution, with 39% of the particles counted smaller than 0.1μ in diameter and 94% smaller than 0.5μ .

The laboratory process line mock-up system described above has been utilized in determining filter efficiencies under simulated sampling conditions. In these experiments, synthetic, ball-milled calcine is irradiated in a ^{249}Cf neutron multiplier facility. Induced activity of selective elements present in the calcine material allows "massless" breakthrough of test filters to be assessed with high sensitivity by γ -ray analysis. As of the end of this quarter, three types of filters have been tested under anticipated

sampling conditions. The results of these studies are summarized in Table 2.1.

DIRECT-INLET MASS SPECTROMETRY (DIMS) - D. L. Styris

Application of direct-inlet mass spectrometry for particulate analysis of waste process effluent was discussed in a previous quarterly report.⁽¹⁾ Since that time, design of the direct inlet mass spectrometer (DIMS) has been completed, fabrication started, and components ordered. This design, illustrated in Figure 2.2, includes several unique features which reduce background interference and increase the system's efficiency.

Interferences due to system contamination are minimized by making the entire ultra-high vacuum compatible and reducing the hydrocarbon contamination due to pump back-streaming.

TABLE 2.1. Filter Efficiency Results

Filter Type	Length of Run, hr	Gas Velocity, m/sec	Mass of Deposit, mg	Volume of Air Flow, m ³	Filter Efficiencies Based on Six Peaks (3- ¹⁴⁰ La and 3- ²⁴ Na), %
IPC	2.7	2.78	70	30.8	51.5
0.2- μ m Nuclepore	2.05	2.78	36	23.4	100
5- μ m Millipore	1.5	2.78	72.4	17.1	> 99.9
5- μ m Millipore	1.5	1.41	61.1	8.7	> 99.9

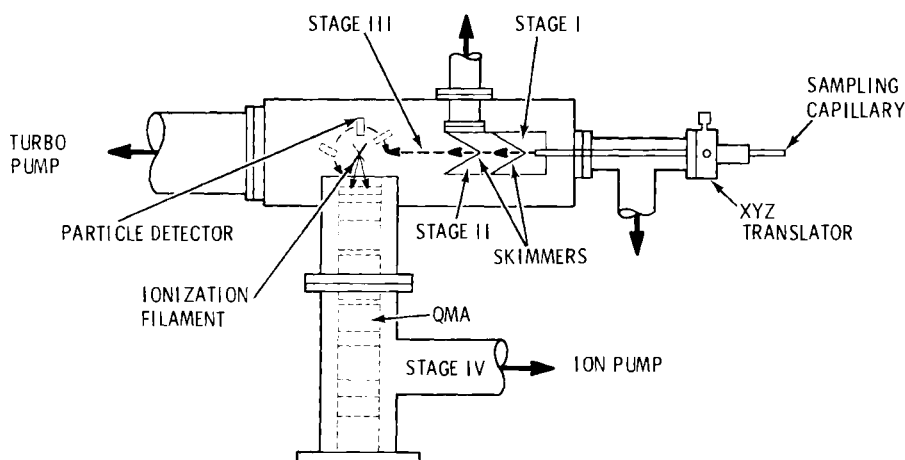


FIGURE 2.2. Schematic of Ultra-High Vacuum DIMS System

This is achieved in this design by incorporating the following features:

1. All-metal construction allows high-temperature bake-out to maximize thermal desorption needed for high-vacuum processing of the system. The skimmer housing is supported within the main vacuum chamber so that it can be vacuum processed with the chamber.
2. The turbomolecular pump used to pump the chamber stage is mated to an isolation gate valve which will allow the pump to run continuously. Since the pump will not be "shutdown," when the system
3. is back-to-air, back-streaming of pump bearing oil will be minimized. Forepumping of the vacuum chamber and quadrupole mass analyzer (QMA) housing is done using standard cryosorption techniques. This eliminates hydrocarbon contamination which would result from mechanically forepumping the system.
4. Bake-out of the vacuum chamber is achieved by internally irradiating the vacuum walls, using internally mounted quartz infrared heaters. This permits rapid bake-out and pulse processing of the system.

5. The inlet capillary tube is metal bellows supported on an x-y-z translator. This facilitates capillary positioning and allows high-vacuum processing of the inlet portion of the system.
6. Water-cooled condensers have been incorporated in the pumping lines of mechanical pumps used to evacuate the skimmer housing stages and back-pump the turbomolecular pump. This condenses back-streaming pump oil without freezing the large quantities of water vapor pumped by these systems. Fast-acting solenoid isolation valves protect the system from contamination during power failure.
7. The mechanical pumping units on the skimmer housing are connected to the housing through UHV all-metal isolation valves. This allows the inlet system to be vacuum-processed prior to operation of the inlet system.

Thermal ionization will take place within the cross-axis ionizer of the Extra Nuclear QMA. It is hoped that this will increase the sensitivity by increasing the ion collection capability. The ionization filament will be contained within the cross-axis ionizer head.

To aid positioning of the system for application to the waste solidification program, the system will be mounted on a portable table having continuous height adjustment capability. This table will support all but the two mechanical forepumps and one mechanical backing pump.

GASEOUS ANALYSIS OF ^3H - AND ^{14}C -
BEARING COMPOUNDS - R. W. Gales

Of the radioactive elements which can be expected to be volatilized during the calcination process, ^3H and ^{14}C form by far the greatest number of chemically different volatile species. As a result, any analysis system designed to characterize an off-gas stream with regard to tritium and ^{14}C will have to address itself to the many chemical identities assumed by these isotopes. A sampling system developed to differentially trap all expected major forms of tritium and ^{14}C , according to chemical class, is illustrated in Figure 2.3. The first trap in this series arrangement consists of a highly efficient desiccator which will selectively remove all tritiated forms of water (HTO , T_2O and DTO). The second trap consists of a CO_2 scrubber which will collect all $^{14}\text{CO}_2$ present in the air stream. The sampled air then passes through a third trap (HT) which catalytically oxidizes all hydrogen components of the gas stream (H_2 , HT, T_2 , DT, D_2 , HD, DT) and adsorbs the oxidized products, in situ. A hopcalite catalyst is then used to oxidize the CO component of the gas stream to CO_2 ; then the fourth trap, containing a CO_2 scrubber, removes this source of ^{14}C , ^{14}CO , from the sample air stream. A platinum catalyst at 600°C is subsequently used to completely oxidize all organic constituents in the sampling stream. The oxidation products, water and carbon dioxide, are

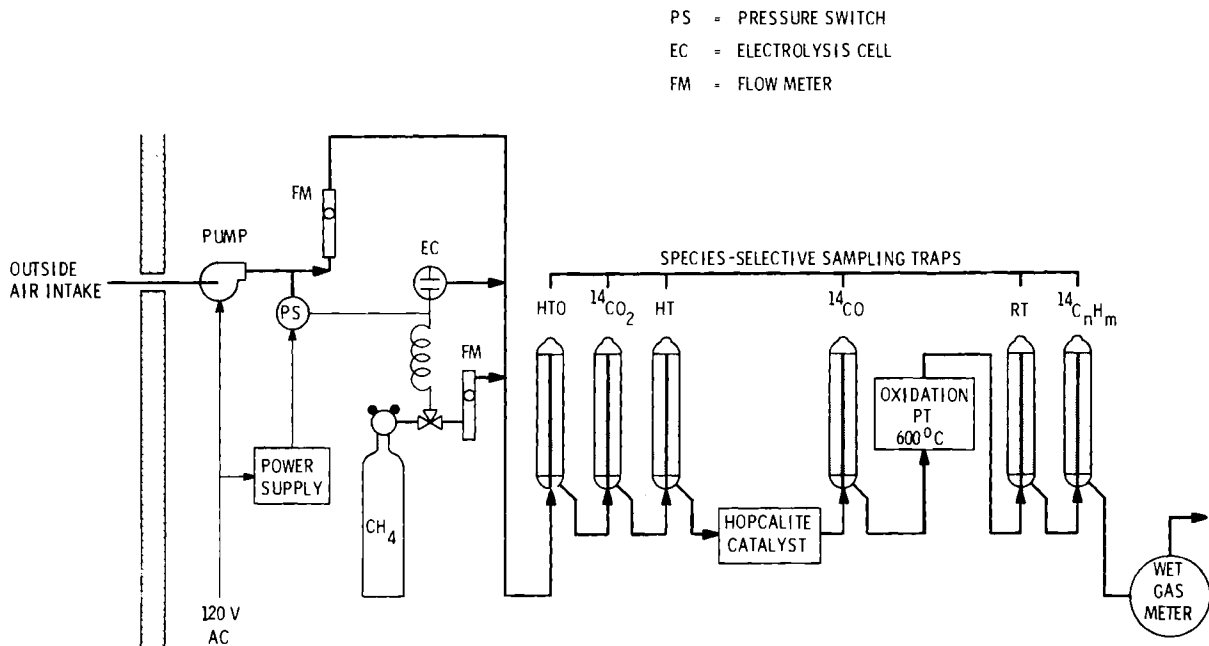
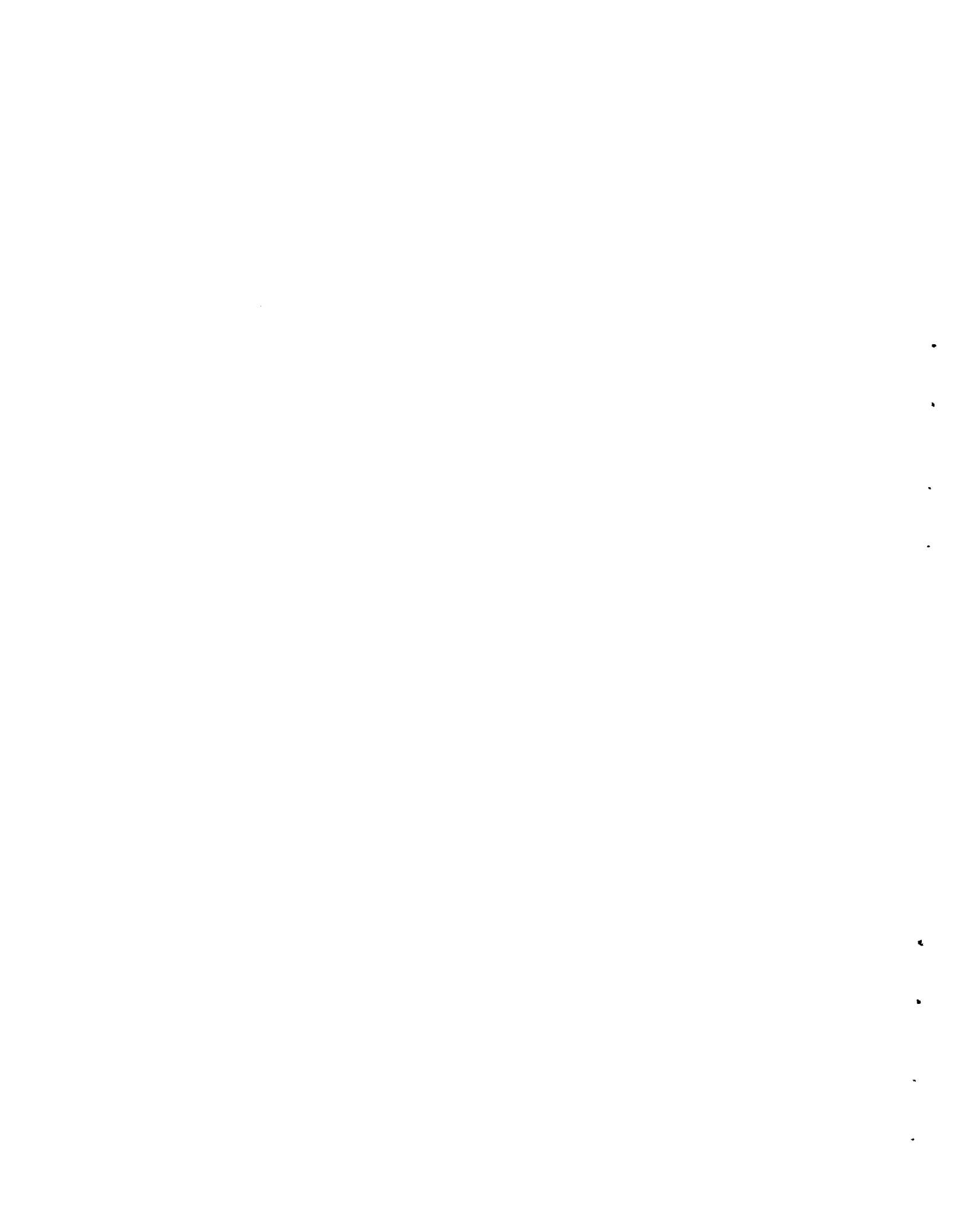


FIGURE 2.3. Gaseous ^3H and ^{14}C Sampling System

separately trapped as previously described. But in this case they are representative of the tritium and ^{14}C contents of organic effluent, respectively.

Several species-specific trapping agents, chemical as well as physical,

have been assembled and will be tested and evaluated in the laboratory to characterize their efficiencies as a function of various expected operating conditions.



3.0 TRU WASTE FIXATION STUDIES

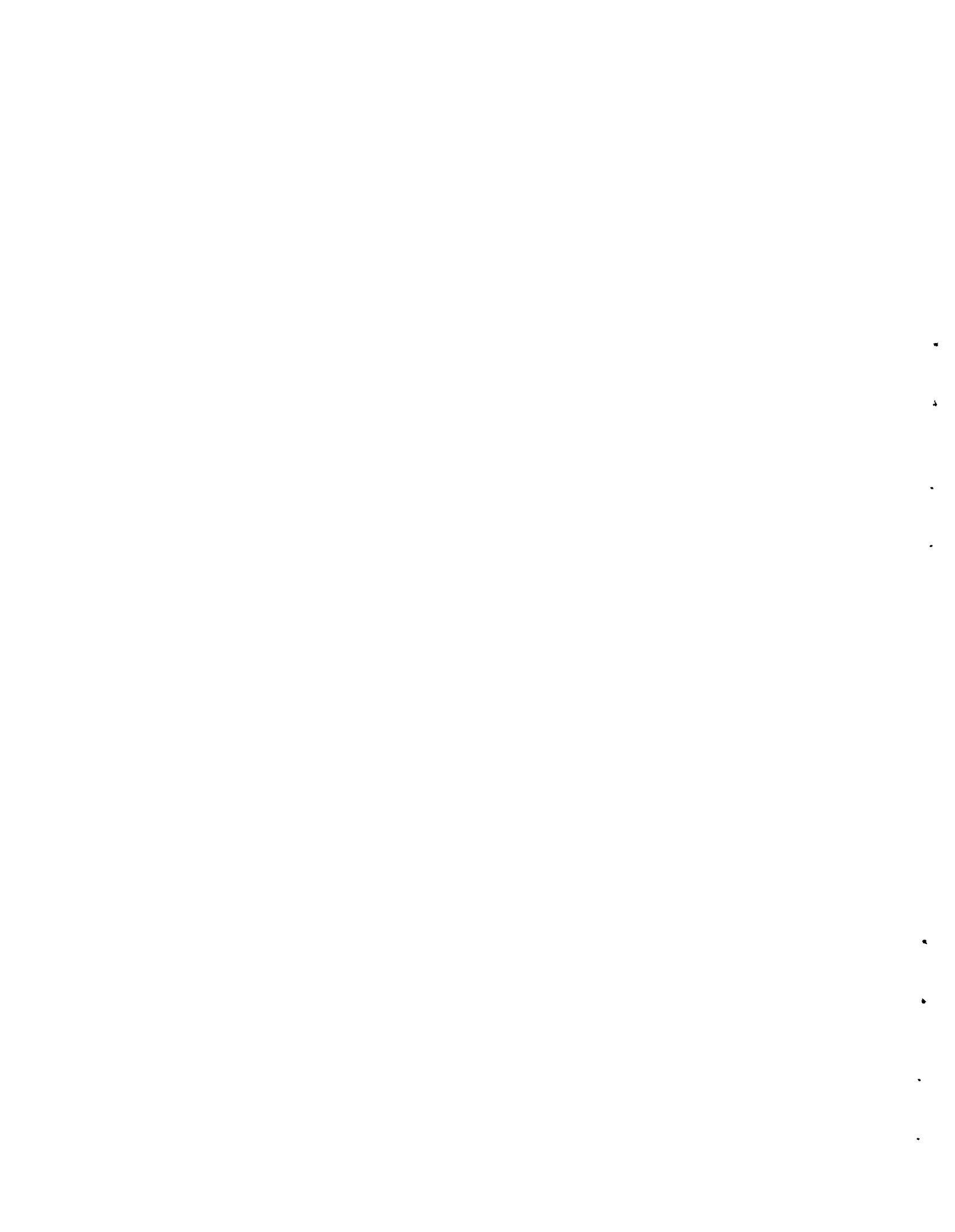
J. H. Jarrett - Program Manager

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

A letter report on TRU Waste Management Criteria Development was issued to DOE Richland Operations on November 11, 1977. The report recommended establishment of a national committee chaired by a DOE-Headquarters staff member and composed of representatives of the various national laboratories involved in TRU waste management programs. The committee would be supported by staff

at PNL, to act as a focal point in guiding the development and adoption of TRU waste form criteria. These viewpoints were reinforced during discussions on December 8, 1977 with D. M. Rohrer of the Nuclear Regulatory Commission.

Additional planning on the program objective will be discussed with the staff of DOE Richland Operations during the next quarter.



4.0 KRYPTON SOLIDIFICATION

G. L. Tingey - Project Manager

The objective of this program is to determine the feasibility of alternative techniques for storing krypton in a solid matrix. Two techniques are currently being investigated: dissolution in a glass matrix and entrapment in solids during high-rate sputtering.

SUMMARY

The technical feasibility of trapping large quantities of krypton in a metal during sputter deposition has previously been demonstrated. Krypton loading of about 4 to 5 at.% is obtained in crystalline metals and over 10% in noncrystalline metals. Other metal alloys have been identified which have potential for yielding an amorphous structure when sputter-deposited and will be tested. Design and testing of equipment for large-scale sputter deposition with radioactive krypton are also reported.

Krypton solubility in partially dense glasses is being determined. The densification of two types of porous glass has been measured. Complete densification of both kinds of glass rods was achieved by heating at 270°C/hr to 800° and 1000°C, respectively, and then holding at the maximum temperature for 2 hr.

KRYPTON ENTRAPMENT DURING SPUTTERING -

E. D. McLanahan, M. A. Bayne,
E. N. Greenwell, and G. L. Tingey

The technical feasibility of trapping large quantities of krypton in a metal which is sputter-deposited

using high-energy Kr^+ ions has been proven for various metal matrices and in particular for the noncrystalline metal alloy $\text{Fe}_{0.87}\text{Y}_{0.13}$.

However, before the process is proven to be applicable as a nuclear waste handling technique for ^{85}Kr , the process and equipment requirements must be much more clearly defined and the economics of the technique evaluated. During the past quarter we have focused our efforts on development of the sputtering equipment and on an evaluation of alternative matrix alloys.

At present, the laboratory sputtering apparatus used is limited to a 7-amp target current. During the past quarter, a new limiter was designed using a control panel and up to four separate paralleled, 7-amp output units. The control panel and two of the output units have now been constructed. The control panel and a first output unit work properly except for a persistent rf oscillation in the tube grid control circuit. This problem is being solved by modifying the grid voltage control circuitry to eliminate the effects of the secondary electron emission.

We have also designed a closed-cycle gas handling and sputtering system for use in a production unit. In this

system all exhaust gases are collected from the vacuum pump and recycled back to the sputtering chamber without allowing any atmospheric leaks. The design of this system is complete, and the major components have been ordered.

Since there are many variables in the sputtering process, we have continued to investigate means of trapping the gas more efficiently. During this quarter, we have investigated the effect of varying the plasma voltages and found that a more simplified electrical circuit could be used. If this same concept proves applicable on the scaled-up prototype unit, it will then be possible to eliminate a power supply from the plasma current circuit. This will reduce the initial capital investment and increase the system's reliability.

To obtain maximum krypton loadings, we have continued investigating various metal alloys. To date we have reported loadings of 4 at.% in crystalline nickel, 3 at.% in crystalline aluminum, 5 at.% in crystalline steel, 5 at.% in crystalline titanium, and 11 at.% in a glassy $\text{Fe}_{0.87}\text{Y}_{0.13}$ alloy containing about 2 at.% impurity ($\text{Fe}_{0.85}\text{Y}_{0.12}\text{Me}_{0.02}$). The higher gas loading in amorphous metals is consistent with other reported data. It is probably due to a larger concentration of trapping sites⁽¹⁾ than is present in crystalline materials. Other suitable alloys are being investigated using the following criteria as guidance:

1. Cost \lesssim \$50/kg
2. Crystallization should occur above 500°C. Since initial gas release usually occurs at or near crystal-

lization temperature, accidental release will be limited to only those instances when the temperature of the krypton storage material exceeds 500°C.

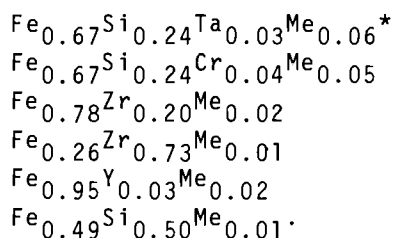
3. Alloys should be, but are not limited to, commercially available ones.
4. Some degree of corrosion resistance should be inherent.

An alloy that generally fits these criteria is "Duriron," an $\text{Fe}_{0.69}\text{Si}_{0.25}\text{Me}_{0.06}$ * alloy used for noncorrosive soil pipe. During this quarter, a target from this material was fabricated and tested. The apparatus and parameters developed in previous depositions were used. Even though Duriron is susceptible to thermal shock, it withstood the application of full sputtering power without cracking. However, a water leak developed in the target, probably between large grains, prematurely ending the experiment. Little or no krypton trapping occurred, and x-ray analysis of the deposit indicated that it was crystalline. This contrasts with the results of Shimada and Kajima,⁽²⁾ who reported obtaining amorphous material of composition $\text{Fe}_{1-x}\text{Si}_x$ ($0.2 \leq x \leq 0.43$) by sputter deposition at much lower rates. Duriron's high impurity content may have been the chief problem in our not obtaining an amorphous product. However, largely because of the low cost of iron-silicon alloys, further experiments with the Fe-Si system are planned.

After the failure of Duriron a literature survey was undertaken to determine those alloys most likely to

* Me = minor or impurity element.

sputter-deposit in the glassy state. The following alloys have been selected for the next series of depositions:



Decay of ^{85}Kr leads to the formation of the fission product, ^{85}Rb . Therefore, for storage of ^{85}Kr for long times, the effect of the rubidium on the alloy is of interest. Possible effects include premature gas release, lowered crystallization temperature, and reduced corrosion resistance of the material. Since the primary candidate material currently is $\text{Fe}_{0.87}\text{Y}_{0.13}$, the initial basis for study will be the Fe-Y-Rb system. The literature indicates that rubidium may cause a decrease in the electrical resistivity⁽³⁾ and a corresponding increase in the thermal conductivity of the alloy with time because electrically conducting rubidium atoms are slowly replacing the insulating krypton atoms. This effect should not change the order of magnitude of the resistivity since the krypton is only ~10% of the alloy to start with and only ~6% of that is ^{85}Kr .

More importantly, it is not known how drastically the presence of about 2% rubidium will affect the crystallization temperature of the alloy. To test this aspect, a sputtering apparatus was designed to make it possible

* Me = minor or impurity element.

to co-sputter rubidium and iron and Fe-Y to obtain nonradioactive material for testing. If this area of experimentation is successful, it may be possible to establish portions of the diagram for the Fe-Rb phase. This is not a simple task in itself, and generally it is not feasible to form these alloys by methods other than co-sputtering.

The sputtering apparatus is now installed in a temporary location during the remodeling of the 231-Z Building. It is anticipated that the sputtering equipment will be installed in the restored laboratory space sometime in May.

DISSOLUTION OF KRYPTON IN GLASS -

G. L. Tingey and W. J. Gray

The solubility of krypton in dense glasses has been shown⁽⁴⁾ to be too low for a viable storage of ^{85}Kr . However, less dense glasses may be of considerable interest if the solubility can be increased significantly without a marked increase in the rate of krypton release. To study this phenomenon we have obtained two types of porous glass rods; both of these have a very open structure with a surface area of about $100 \text{ m}^2/\text{g}$, but they densify at different temperatures. These glasses will be loaded with krypton and then partially densified in an attempt to reduce the release rate of the gas from the sample.

We have measured the densification rate of each of these glasses as a function of temperature using a dilatometer. The length changes are shown as a function of time for the two

types of glass in Figures 4.1 and 4.2, respectively. The heating rate for these measurements was 270°C/hr, and the temperature as a function of time is also shown in the figures.

Krypton loading experiments are now under way. The loadings are performed after the glass samples are thoroughly degassed by heating to 125°C overnight at a pressure of less than 10^{-4} torr. The samples are then partially densified by heating at a

predetermined temperature for the appropriate time. Gas release measurements will then be made to determine the total gas loadings and the release rate as a function of temperature. If these measurements show sufficiently high gas loadings, further experiments will be conducted at krypton pressures of up to 30,000 atm in the high-pressure krypton furnace at Idaho National Engineering Laboratory.

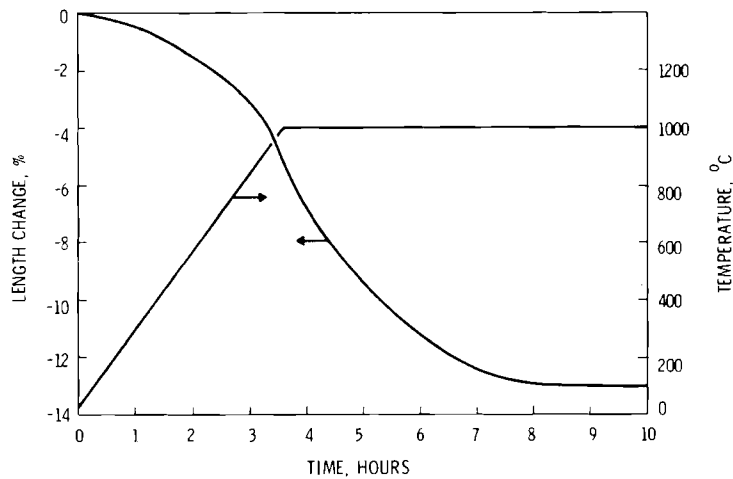


FIGURE 4.1. Densification of High-Temperature Softening Porous Glass Rod

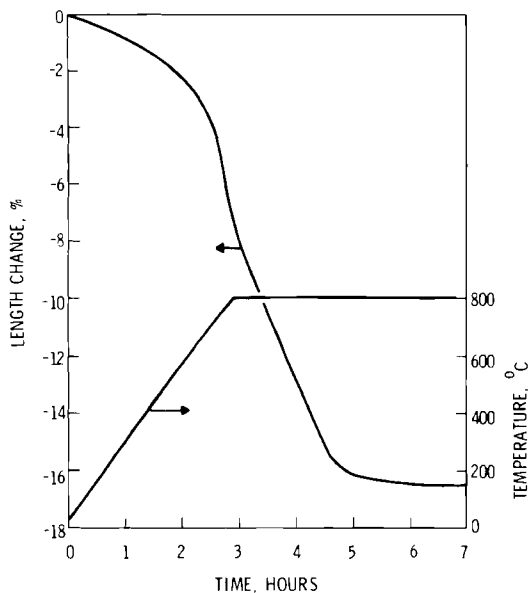


FIGURE 4.2. Densification of Intermediate-Temperature Softening Porous Glass Rod

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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 ^{129}I and ^{14}C released from the treatment of irradiated fuels.

SUMMARY

Cost and availability were added to the list of parameters for choosing fixation materials. It is found that copper and lead meet all three conditions: cost, domestic reliability, and supply. Lowest on the scale are bismuth, silver, and mercury. Attempts to weigh each of these factors along with the physical and chemical properties were made but are generally unsatisfactory due to lack of real criteria for either interim or permanent storage. This problem is currently being addressed.

FIXATION CANDIDATES - R. D. Scheele and K. D. Wiemers

To select the candidates for leaching studies, four parameters have been considered: thermal stability, solubility, cost, and availability. The thermal stability, which includes resistance to oxidation or reaction with water, was evaluated both experimentally, using TGA and DSC techniques, and theoretically, using available thermodynamic data. The latter were summarized in a previous report.⁽¹⁾ The marketing data are reported here in a later section. All are summarized in Table 5.1.

Tables 5.2 through 5.4 rank the candidates in the order of acceptable physical properties. Combining these data with the cost and availability data suggests that the "best" candidates are AgI , $\text{La}(\text{IO}_3)_3$, $\text{Th}(\text{IO}_3)_4$, $\text{Sr}(\text{IO}_3)_2$, and $\text{Ca}(\text{IO}_3)_2$ for iodine and CaCO_3 , and SrCO_3 for ^{14}C . However, it is evident from Tables 5.2-5.4 that if storage conditions are defined on the bases of limited water contact, low temperature, or limited oxygen, then other candidates become very attractive, e.g., copper and lead compounds. Thus it becomes desirable to define transportation and storage limitations, and attention is being given this problem.

FIXATION CRITERIA - R. D. Scheele and L. L. Burger

The environmental conditions which ^{129}I and ^{14}C fixation compounds will experience during storage are not yet defined. For geologic storage, assuming a surface temperature of 20°C and a normal temperature gradient of $30^\circ\text{C}/\text{km}$, the temperatures will vary from 29 to 65°C .⁽²⁾ If the low-level non-transuranic waste is stored near high-level waste, temperatures are likely to reach no more than 250°C , not necessitating stability at high

TABLE 5.1. Solubility, Thermal Stability, Cost, and Availability Data for Iodate, Iodide, and Carbonate Fixation Compounds

Compound	Solubility in Pure H ₂ O ^(a)	Thermal Stability ^(b)	Cost ^(c)	Availability		
				Domestic ^(d)	World Reserve ^(e)	World Resource ^(f)
Iodates						
Ba(IO ₃) ₂ ·H ₂ O	1.4 × 10 ⁻³	-	1.43 × 10 ²	0.69	0.84	8.4
Ba(IO ₃) ₂	-	490	-	-	-	-
Bi(IO ₃) ₃	-	300	9.26 × 10 ³	0.36	0.49	1.1
Ca(IO ₃) ₂	1.1 × 10 ⁻²	260	7.05	1.02	-	-
Ce(IO ₃) ₃	5.6 × 10 ⁻³	380	1.82 × 10 ⁴	1.77	4.5	22
Ce(IC ₃) ₄	7 × 10 ⁻⁴	330	1.37 × 10 ⁴	-	-	-
CuIO ₃	3.7 × 10 ⁻⁴	-	7.87 × 10 ²	0.90	1.3	5.8
Cu(IO ₃) ₂ ·H ₂ O	5.3 × 10 ⁻³	-	3.95 × 10 ²	-	-	-
Cu(IO ₃) ₂	-	420	-	-	-	-
La(IO ₃) ₃	2.1 × 10 ⁻³	490	2.46 × 10 ⁴	1.77	4.5	22
Pb(IO ₃) ₂	8.6 × 10 ⁻⁵	410	4.20 × 10 ²	0.63	0.70	2.4
Hg ₂ (IO ₃) ₂	1.4 × 10 ⁻⁷	250	-	0.53	0.75	2.7
Hg(IO ₃) ₂	8.6 × 10 ⁻⁵	410	2.91 × 10 ³	-	-	-
Ni(IO ₃) ₂	3 × 10 ⁻²	-	1.15 × 10 ³	0.70	2.1	4.2
AgIO ₃	1.7 × 10 ⁻⁴	430 (decomp. to AgI)	1.34 × 10 ⁵	0.22	0.44	1.7
Sr(IO ₃) ₂	8.7 × 10 ⁻³	stable to 550	15.5	0	2.0	"adequate"
Th(IO ₃) ₄	1.6 × 10 ⁻³	480	3.08 × 10 ³	3.8	110	-
UO ₂ (IO ₃) ₂	4.0 × 10 ⁻³	70	2.98 × 10 ⁴	0.86	0.41	0.71
Zn(IO ₃) ₂	3.4 × 10 ⁻³	410	2.06 × 10 ²	0.36	1.1	6.9
Iodides						
BiI ₃	3.9 × 10 ⁻⁵	sublimes 240	1.42 × 10 ²	0.36	0.49	1.1
CuI	1.0 × 10 ⁻⁶	260	7.87 × 10 ²	0.90	1.3	5.8
PbI ₂	2.4 × 10 ⁻³	400	4.2 × 10 ²	0.63	0.70	2.4
Hg ₂ I ₂	9.7 × 10 ⁻¹⁵	sublimes 106	-	0.53	0.75	2.7
HgI ₂	1.3 × 10 ⁻⁴	sublimes 110	2.91 × 10 ³	-	-	-
AgI	9.1 × 10 ⁻⁹	542	1.34 × 10 ⁵	0.22	0.44	1.7
PdI ₂	-	stable 500	1.10 × 10 ⁶	0.016	2.4	6.9
TlI	2.5 × 10 ⁻⁴	390	6.55 × 10 ³	2.14	1.2	-
Carbonates						
BaCO ₃	7.1 × 10 ⁻⁵	stable to 550	7.96 × 10 ³	0.69	0.84	8.4
CaCO ₃	5.3 × 10 ⁻⁵	stable to 550	3.87 × 10 ²	1.02	-	-
CdCO ₃	2.3 × 10 ⁻⁶	300	1.46 × 10 ⁵	0.39	1.1	28
PbCO ₃	1.8 × 10 ⁻⁷	347°C; 1 atm CO ₂	2.3 × 10 ⁴	0.63	1.25	2.4
SrCO ₃	1.0 × 10 ⁻⁵	stable to 550	8.46 × 10 ²	0	2.0	"adequate"

a. Values are for [I] and [CO₃²⁻] in molarity as calculated from the solubility product, K_{sp}.⁽³⁾

b. Temperature in °C at which a weight loss >0.5% was detected by TGA. Value for PbCO₃ from Reference 4.

c. Operational cost of three 1500-MTU treatment plants. It assumes 4 × 10³ moles I₂ produced per year and 2.19 × 10⁵ moles ¹⁴C produced per year. For further explanation see the section herein on marketing data.

d. Values are the ratio of U.S. supply to demand in the year 2000.

e. Values are the ratio of world reserve as of 1974 to world cumulative demand (1974-2000).

f. Values are the ratio of world resources as of 1974 to world cumulative demand (1974-2000).

TABLE 5.2. Relative Ratings of Iodide Compounds

Solubility in Water, [I]		Iodine Release, °C		Reactivity to Oxygen ($\Delta G_{mo} - \Delta G_{mi}$, kcal mol ⁻¹)				
Decreasing Solubility ↑	Hg ₂ I ₂	9.7 × 10 ⁻¹⁵	Increasing Stability ↑	AgI	542	Decreasing Reactivity ↑	AgI	+21.05
	AgI	9.1 × 10 ⁻⁹		PdI ₂	500		Tl	+13.45
	CuI	1.0 × 10 ⁻⁶		PbI ₂	400		HgI ₂	+4.80
	BiI ₃	3.9 × 10 ⁻⁵		TlI	390		PdI ₂	+0.30
	HgI ₂	1.3 × 10 ⁻⁴		CuI	260		Hg ₂ I ₂	-0.75
	TlI	2.5 × 10 ⁻⁴		BiI ₃	240 sub.		PbI ₂	-1.80
	PbI ₂	2.4 × 10 ⁻³		HgI ₂	110 sub.		BiI ₃	-5.82
			Hg ₂ I ₂	106 sub.	CuI	-14.4		

TABLE 5.3. Relative Ratings of Iodate Compounds

Solubility in Water, [I]		Thermal Stability, °C			
Decreasing Solubility ↑	Hg(IO ₃) ₂	1.4 × 10 ⁻⁷	Increasing Stability ↑	Sr(IO ₃) ₂	stable to 550
	Pb(IO ₃) ₂ /Hg(IO ₃) ₂	8.6 × 10 ⁻⁵		Ba(IO ₃) ₂ /La(IO ₃) ₂	490
	AgIO ₃	1.7 × 10 ⁻⁴		Th(IO ₃) ₄	480
	CuIO ₃	3.7 × 10 ⁻⁴		AgIO ₃	430 decomposes to AgI
	Ce(IO ₃) ₄	7.0 × 10 ⁻⁴		Cu(IO ₃) ₂	420
	Ba(IO ₃) ₂ ·H ₂ O	1.4 × 10 ⁻³		Zn(IO ₃) ₂ /Pb(IO ₃) ₂ /Hg(IO ₃) ₂	410
	Th(IO ₃) ₄	1.6 × 10 ⁻³		Ce(IO ₃) ₃	380
	La(IO ₃) ₂	2.1 × 10 ⁻³		Ce(IO ₃) ₄	330
	Zn(IO ₃) ₂	3.4 × 10 ⁻³		Bi(IO ₃) ₂	300
	UO ₂ (IO ₃) ₂	4.0 × 10 ⁻³		Ca(IO ₃) ₂	260
	Cu(IO ₃) ₂ ·H ₂ O	5.3 × 10 ⁻³		Hg(IO ₃) ₂	250
	Ce(IO ₃) ₃	5.6 × 10 ⁻³		UO ₂ (IO ₃) ₂	70
	Sr(IO ₃) ₂	8.7 × 10 ⁻³			
	Ca(IO ₃) ₂	1.1 × 10 ⁻²			
Ni(IO ₃) ₂	3.0 × 10 ⁻²				

TABLE 5.4. Relative Ratings of Carbonate Compounds

Solubility in Water [CO ₃ ⁻]		Thermal Stability, °C ^(a)			
Decreasing Solubility ↑	PbCO ₃	1.8 × 10 ⁻⁷	Increasing Stability ↑	BaCO ₃	1297
	CdCO ₃	2.3 × 10 ⁻⁶		SrCO ₃	1258
	SrCO ₃	1.0 × 10 ⁻⁵		CaCO ₃	817
	CaCO ₃	5.3 × 10 ⁻⁵		PbCO ₃	347
	BaCO ₃	7.1 × 10 ⁻⁵			

a. Temperature necessary to obtain 1 atm of CO₂.⁽⁴⁾

temperatures for storage. Thus, unless special restrictions are later defined for transportation, stability at high temperatures is not required.

The criteria which the package must meet during transport were discussed in the last quarterly report.⁽⁵⁾ It should be emphasized that due to the specific activities of ^{129}I and ^{14}C diluted with expected quantities of natural carbon from the air, these isotopes are classified as low-specific-activity radioactive materials both in the U.S.⁽²⁾ and internationally.⁽⁶⁾ Requirements are therefore minimal. The packages must be labeled as radioactive material and shipped in a sole-use vehicle.

In addition to the parameters discussed earlier, the factors to be considered in setting the criteria include the following:

- The fixation process should be operable and maintainable in radioactive environment.
- Waste streams should be minor in volume and contain only trace activity.
- The process should be free from explosive or other hazards.
- The environmental pathways of possible release forms must be determined, and the biospheric residence times of these forms must be estimated.
- If storage with HLW is contemplated, then radiation stability can be important.
- The greatest hazards may exist during interim storage or shipping. Therefore, despite the low-specific-activity classification, solubility and thermal stability should be considered for these activities.

IODIDE CONVERSION - R. D. Scheele

If an iodate is chosen as the final fixation form for iodine, it will be necessary to convert iodide solutions, metal iodides, or the HI produced during regeneration of silver zeolite beds to iodate. Several methods reported in the literature have been examined; bromine,⁽⁷⁾ chlorine,⁽⁷⁾ hydrogen peroxide,^(8,9) chlorate,⁽¹⁰⁾ hypochlorite,⁽¹¹⁾ ozone,⁽¹²⁾ permanganate,⁽¹³⁾ and electrolysis^(10,12) have been used for oxidation. Preliminary experiments with these, as well as with Ce(IV), have been inconclusive except for electrolysis and oxidation with chlorine. Ozone has not yet been tried.

MARKETING DATA FOR ^{129}I AND ^{14}C FIXATION MATERIALS - K. D. Wiemers

An evaluation of the cost and availability of candidate ^{129}I and ^{14}C fixation materials was made based on their ratings in three categories:

1. Cost - the cost per kilogram of each metal was used to estimate the actual operational cost for three 1500 MTU treatment plants (see Table 5.1). The cost of the metal was expected to be less than, but relative to, the cost of the actual salt used for fixation. The operational cost data were rated between zero and ten using the value of the reciprocal of the ln of the operational cost.
2. Domestic reliability - this was reflected in the ratio of domestic supply to demand. A ratio value less than one implied that domestic demand could be met only with

reliance on foreign imports. The rating of these data was based on the forecast for the year 2000.

3. Supply - this category was divided into two parts: world reserve as of 1974 and identified world resources as of 1974. These were both compared with the cumulative world demand (1974-2000). Briefly, the Bureau of Mines' definition of reserve is the supply of metal which is economically and politically obtainable, whereas identified resources include known deposits not yet accessible.

Data for the analysis are presented in Table 5.5. To evaluate the ratings three acceptability limits were defined:

- cost per year less than \$1000
- domestic reliability greater than 50%
- satisfactory world supply.

The relative ratings are listed in Table 5.6, with the best candidates for specific qualities shown in the top lines. The two metals that meet all three conditions are lead and copper. At the other end of the scale are bismuth, silver, and mercury. Strontium is inexpensive and in good world supply, but reliance on imports is 100% (mainly from Spain and Mexico). Barium is inexpensive, and a sufficient domestic quantity is forecast to meet demands in the year 2000. However, the world supply is limited. The presence of a large amount of world barium resources indicates that future technological development and perhaps some political changes could lead to an adequate supply. Thorium, thallium, cerium,

and lanthanum are relatively expensive--\$3,090, \$6,550, \$18,200, and \$24,600, per year, respectively. World supply is adequate, and U.S. demands in the year 2000 could be met domestically. Important to note in regard to domestic economics is that the thallium requirements for the treatment plant are more than half the amount of forecasted domestic consumption for the year 2000. Palladium is most expensive (about ten times more than silver), and domestic supply is limited. Similar to thallium, palladium's requirements for the treatment plant approached those of the domestic trade market. World supply of palladium was adequate. Uranium is expensive, and world supply of uranium for enrichment is limited. Domestic and world stockpiles of depleted uranium, however, are adequate to meet projected demands. Cadmium, nickel, and zinc are borderline cases in all three categories. For the ^{14}C fixation candidates, calcium was most acceptable (although this is based on limited information concerning world supply). Strontium's ratings are the same as with ^{129}I candidates. Lead and barium are two of the more expensive minerals for carbonate fixation, and, as mentioned above, barium presents a problem in world supply. Cadmium is most expensive, \$146,000 per year, and a borderline case in domestic and world supply.

Based on the cost and availability ratings, copper and lead are most acceptable, and bismuth, silver, and mercury are least acceptable for ^{129}I fixation. Acceptance of thorium, nickel, zinc, strontium, lead, barium,

TABLE 5.5. Raw Data for Cost and Availability Rating

Metal	Cost \$/kg(a)	Domestic Reliability ^(b)		World Supply	
		Ratio, U.S. Production (yr 2000) to Demand (yr 2000), MT	Ratio, World Reserve (yr 1974) to Demand (yr 1974-2000), MT ^(b,c)	Ratio, World Resource (yr 1974) to Demand (yr 1974-2000), MT ^(b,c)	
Ba	0.27	$\frac{998 \times 10^3}{1438 \times 10^3} = 0.69$	$\frac{9.07 \times 10^7}{1.08 \times 10^8} = 0.84$	$\frac{9.07 \times 10^8}{1.08 \times 10^8} = 8.4$	
Bi	16.67	$\frac{7.26 \times 10^2}{2.04 \times 10^3} = 0.36$	$\frac{5.94 \times 10^4}{1.22 \times 10^5} = 0.49$	$\frac{1.3 \times 10^5}{1.2 \times 10^5} = 1.1$	
Ca	0.04	$\frac{6.62 \times 10^5}{6.50 \times 10^5} = 1.02$	-	-	
Cd	6.00	$\frac{4.54 \times 10^3}{1.15 \times 10^4} = 0.39$	$\frac{7.53 \times 10^5}{6.75 \times 10^5} = 1.1$	$\frac{1.9 \times 10^7}{6.7 \times 10^5} = 28$	
Ce	48.89	$\frac{5.44 \times 10^4}{3.08 \times 10^4} = 1.77$	$\frac{7.01 \times 10^6}{1.55 \times 10^6} = 4.5$	$\frac{3.46 \times 10^7}{1.55 \times 10^6} = 22$	
Cu	1.56	$\frac{3.45 \times 10^6}{3.81 \times 10^6} = 0.90$	$\frac{4.08 \times 10^8}{3.20 \times 10^8} = 1.3$	$\frac{1.9 \times 10^9}{3.2 \times 10^8} = 5.8$	
Pb	0.51	$\frac{8.8 \times 10^5}{1.39 \times 10^6} = 0.63$	$\frac{1.5 \times 10^8}{1.2 \times 10^6} = 1.25$	$\frac{3.0 \times 10^8}{1.2 \times 10^8} = 2.4$	
La	66.67	$\frac{5.44 \times 10^4}{3.08 \times 10^4} = 1.77$	$\frac{7.01 \times 10^6}{1.55 \times 10^6} = 4.5$	$\frac{3.46 \times 10^7}{1.55 \times 10^6} = 22$	
Hg	3.64	$\frac{8.62 \times 10^2}{1.62 \times 10^3} = 0.53$	$\frac{1.7 \times 10^5}{2.3 \times 10^5} = 0.75$	$\frac{6.0 \times 10^5}{2.3 \times 10^5} = 2.7$	
Ni	5.11	$\frac{2.45 \times 10^5}{3.49 \times 10^5} = 0.70$	$\frac{5.4 \times 10^7}{2.6 \times 10^7} = 2.1$	$\frac{1.1 \times 10^8}{2.6 \times 10^7} = 4.2$	
Pd	2600.00	$\frac{0.68}{41.7} = 0.016$	$\frac{6.0 \times 10^3}{2.5 \times 10^3} = 2.4$	$\frac{1.7 \times 10^4}{2.5 \times 10^3} = 6.9$	
Ag	155.56	$\frac{1.56 \times 10^3}{7.15 \times 10^3} = 0.22$	$\frac{1.9 \times 10^5}{4.2 \times 10^5} = 0.44$	$\frac{7.0 \times 10^5}{4.2 \times 10^5} = 1.7$	
Sr	0.04	$\frac{0}{9.07 \times 10^3} = 0$	$\frac{3.2 \times 10^6}{1.6 \times 10^6} = 2.0$	"adequate"	
Tl	16.67	$\frac{1.36}{0.64} = 2.14$	$\frac{2.9 \times 10^2}{2.5 \times 10^2} = 1.2$	-	
Th	6.67	$\frac{9.07 \times 10^2}{2.36 \times 10^2} = 3.8$	$\frac{7.1 \times 10^5}{6.4 \times 10^3} = 110$	-	
U ^(d)	31.56	$\frac{1.73 \times 10^4}{6.3 \times 10^4} = 0.27$	$\frac{1.07 \times 10^6}{2.61 \times 10^6} = 0.41$	$\frac{1.86 \times 10^6}{2.61 \times 10^6} = 0.71$	
Zn	0.82	$\frac{9.98 \times 10^5}{2.77 \times 10^6} = 0.36$	$\frac{2.4 \times 10^8}{2.2 \times 10^8} = 1.1$	$\frac{1.5 \times 10^9}{2.2 \times 10^8} = 6.9$	

a. All costs are for the \$/kg metal except for Ba and Ca. The latter prices were computed as \$/kg metal based on BaSO₄ (1974) and CaCl₂ (1972) prices given in Reference 14. Ce and La are December 1977 prices quoted by Apache Chemicals, Inc. All other prices are 1976 estimates given in Reference 15. Prices of several metals were compared with those listed in References 16 and 17.

b. All statistical forecasts are from Reference 18 except for Ca. The values for Ca represent the quantity of CaCl₂ traded in 1972 as given in Reference 15. Ce and La values are for the total quantity of rare-earth oxides traded. Ce and La comprise 80% of this group.

c. Identified resources are specific bodies of mineral-bearing material whose location, quality, and quantity are known from geologic evidence supported by engineering measurements regarding the demonstrated category, and includes reserves and subeconomic resources. The reserve is that portion of the identified resource from which available mineral or energy commodity can be economically and legally extracted at the time of determination.

d. The price quoted for uranium is derived from the average cost in December 1974 of U₃O₈, the marketable product from the mined ore. The values for domestic demand and production and world cumulative demand are for uranium to be used for enrichment. The estimated domestic demand for depleted uranium in the year 2000 was 3.99 x 10³ MT; 6% of the total domestic demand. About 97% of the demand for uranium to be used for enrichment will be added to the depleted uranium stockpiles providing an abundance of depleted uranium relative to its demand, resources and reserves are determined for natural uranium.

TABLE 5.6. Relative Ratings of Cost and Availability

	Operational Cost	Domestic Reliability	Supply	
			Reserve	Resource
Decreasing Cost ↑ ↓	Ca	U(a)	U(a)	U(a)
	Sr	Th	Th	Sr/Cd
	Ba	Tl	La/Ce	La/Ce
	Zn	La/Ce	Pd	Ba
	Cu(II)	Ca	Ni	Pd/Zn
	Pb	Cu	Sr	Cu
	Cu(I)	Ba/Ni	Cu	Ni
	Ni	Pb	Pb	Hg
	Cd	Hg	Tl	Pb
	Hg	Cd	Cd/Zn	Ag
	Th	Bi/Zn	Ba	Bi
	Tl	Ag	Hg	
	Bi	Pd	Bi	
	Ce(IV)	Sr	Ag	
	Ce(III)			
	La			
	U			
	Ag			
Pd				

a. Based on present and projected availability of depleted uranium.

and cadmium on the bases of cost and availability depends upon the weighted value of domestic reliability, world supply, and operational cost. For ^{14}C calcium is best rated and cadmium

and lead lowest rated. However, the actual range is not so great as to exclude any of the ^{14}C candidates on the bases of cost and availability.

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6.0 INTERNATIONAL PROGRAM COORDINATION

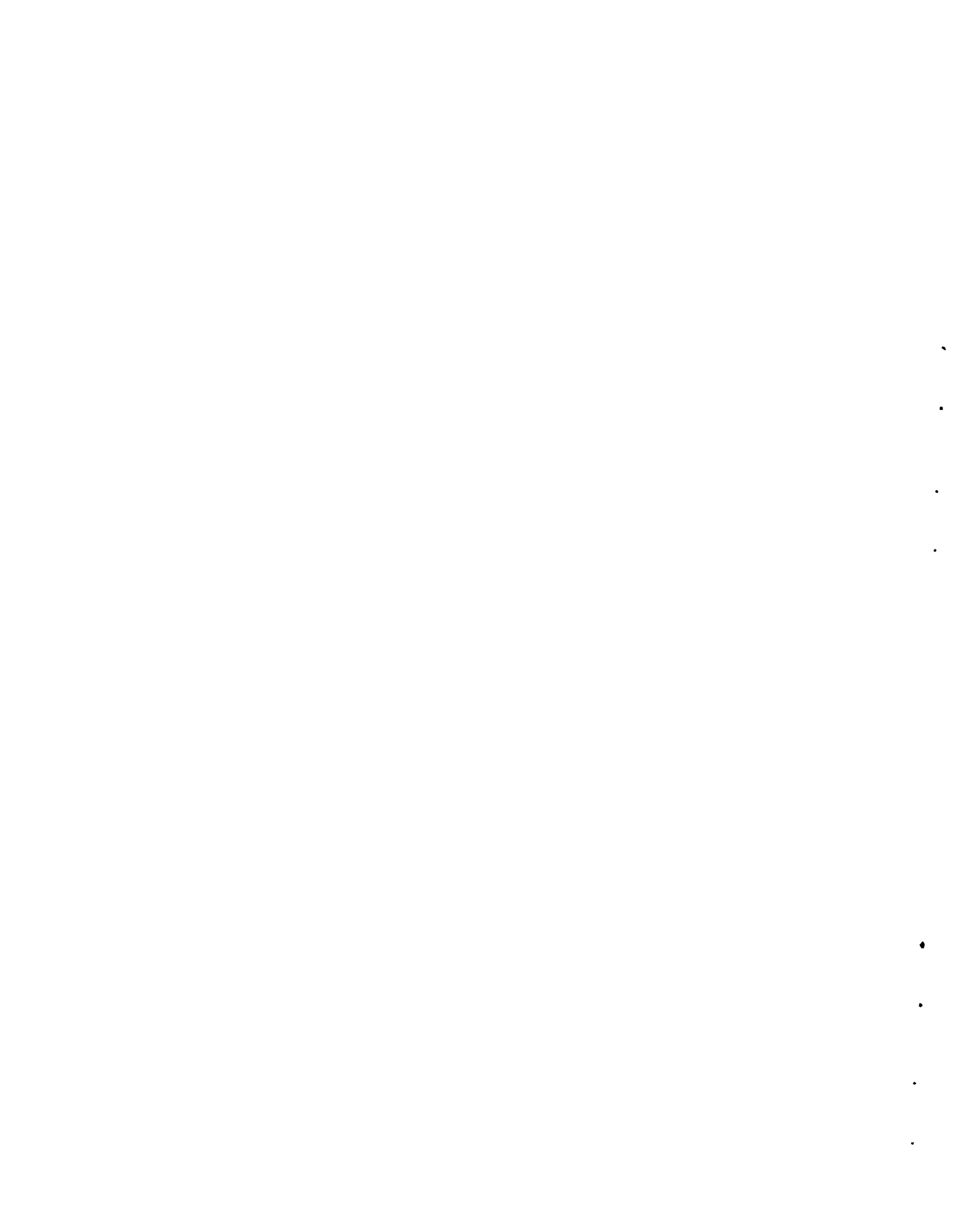
K. M. Harmon - Project Manager

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

SUMMARY

Collection and review of significant documents continued. Summaries

were prepared concerning management programs in Belgium, Italy, and Japan.



7.0 WASTE MANAGEMENT SYSTEM STUDIES

J. W. Bartlett - Program Manager

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

SUMMARY

Three major activities were under way in the first quarter.

In work to develop a data base for evaluating candidate waste repository sites, the computerized data base was expanded, and the computer capability to manipulate and display the data was refined.

In work on nuclear waste communications, analysis and write-up of data from the knowledge/attitudes survey continued. Work on print media information analysis neared completion, and similar analysis for electronic media information was started. Work on issues analysis continued.

In work on public perception and evaluation of risk, questionnaires were distributed to consultants who will gather data in six states. Development of additional data-gathering instruments continued.

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

This work is aimed ultimately 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.

Present work is developing the data base and computerized capability to manipulate the data for practical applications.

Seismic Data Base - T. N. Bishop

During this quarter, more data (approximately 8000 earthquakes) were obtained from the Canadian Dominion Observatory. In addition, a data set from the Rocky Mountain area (about 500 earthquakes) was being sent us by Texas Instruments. Figure 7.1 displays the location of our six seismic data bases at present.

The existing seismic programs were made more efficient, and work began on transferring selected groups of earthquakes for plotting on the associated peripheral graphic devices of the PDP-11/55.

Mapping Programs - H. P. Foote

A detailed project plan document was prepared. It describes the tasks, objectives, and schedule for the project.

Programs were written to evaluate the new display devices on the

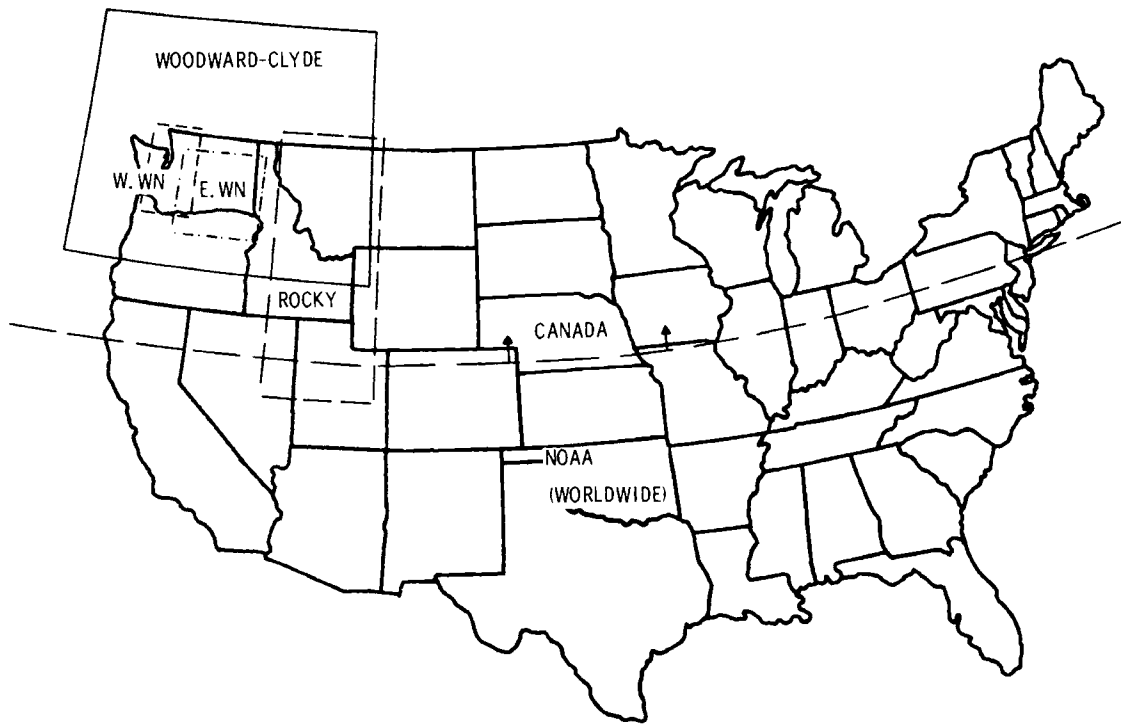


FIGURE 7.1. Location of Seismic Data Bases as of December 1977.

PDP-11/55. The vector plotting accuracy, stability, and capacity of the VS-60 refresh display scope was measured. The tests showed that it was capable of displaying stereo pairs for true three-dimensional plotting of earthquakes. It can refresh 3000 to 4000 vectors without flicker. This allows plotting of maps at a medium detail level. The spatial and color resolution of the spatial data system color display was checked along with its data transfer rate and random access capability. The color quality has proven excellent and the resolution adequate for most mapping applications. The writing rate will allow vector plotting faster than we had expected. This means that most line drawings can be completed on this device in under a half-minute.

A program was written to plot earthquakes coded by color on a LANDSAT image stored on the SDA display. The color was used to denote either the depth of the seismic event or its magnitude. The main purpose of this program was to test whether color-coded information could be overlaid on a gray-scale image. The test was successful, and we expect to employ this technique in our future mapping output displays.

Data Collection - T. N. Bishop and H. P. Foote

In addition to the new seismic data previously mentioned, we acquired a large mapping data base from Lawrence Berkeley Laboratory. This data base consists of approximately 4.8 million

points defining coastal boundaries, islands, lakes, rivers, and political boundaries for the world at a map scale of about 1:2,000,000. It was originally digitized by the CIA and has recently been made available to the Department of Energy laboratories. We have made changes as necessary in the data structure to optimize it for our own computer system. It can now be referenced by our mapping programs and used in conjunction with our other map data bases. For example, it could be used as a base map for plotting seismic events and volcanos.

NUCLEAR WASTE COMMUNICATIONS -

S. M. Nealey, HARC

Data analysis and write-up are halfway to completion on the knowledge/attitude survey of Washington State residents. Initial analyses indicate that human values and the perceived relationship of nuclear power to these values are much more strongly related to attitudes about nuclear power than is knowledge about it. A draft report will be finished next quarter.

The content analysis of printed media coverage of nuclear power issues is nearing completion. The write-up has been redrafted and is presently undergoing internal review before submission for DOE review and clearance.

Work plans have been formulated for the content analysis of the handling of nuclear power issues and events by television networks. Videotape reproductions of relevant portions of the three major network television news broadcasts will be

collected during the second quarter, television coverage will be analyzed the third quarter, and a report will be written the fourth quarter. The content analysis rating form used for the analysis of print media will be used for the analysis of television media in order to provide comparable data. Television coverage of coal and solar technologies will also be analyzed to serve as a reference point for television coverage of nuclear power.

This quarter we have identified a research specialist who will undertake next quarter the analysis of the DOE nuclear information program. Final work plans for this task will be drawn up in January 1978.

Follow-on work on the issues analysis continued. We are currently characterizing the sources from which the major nuclear power issues have come. The work plan for the issues domain analysis has been drawn up, and a final report will be issued by the end of June 1978.

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

During this quarter, distribution of the combined risk perception/risk evaluation questionnaire began. Packets of surveys were sent to consultants in six states: Massachusetts, Florida, Illinois, Texas, Colorado and California. These consultants have arranged to distribute the questionnaires to about 20 citizens' groups. The range of these groups' orientations on dimensions such as

environmental concern, economic development, and political activism will enable us to study differences among groups in the relationship between perception and evaluation of technological risks. The data collection phase will be completed by mid-February.

During this quarter we have progressed in developing questionnaires for investigating some further questions on the public's perception and evaluation of risk. We have completed a draft version of the instrument for the risk perception task and are in the process of revising it. On the values stability task (Risk Evaluation II) we have been more successful than we anticipated. Cross-tabulation of the demographic data from the questionnaire reported in BNWL-1997 indicates that most respondents from the groups we intended to

resurvey are uniquely identifiable from responses to three variables: sex, age and education. If we can obtain permission from these groups to survey their members, we will be able to conduct our analyses at both the group and individual levels rather than only at the group level. The ability to conduct analyses at both levels will allow us to separate the effects of changes in the values of individuals from effects of changes in the composition of the membership of the group.

The instrument for Risk Evaluation I is still being developed. This task will seek to determine whether the apparent differences in acceptability between voluntary versus involuntary risk can be extended to occupational (presumably voluntary) versus public (presumably involuntary) exposure.

8.0 WASTE MANAGEMENT SAFETY ASSESSMENT

W. K. Winegardner - Project Manager

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

SUMMARY

The three-report set (BNWL-2144, 2145, and 2146) was published. It describes and presents user instructions for PNL-developed computer programs for drawing fault trees and for assessing trees in terms of risk. A draft systems definition of the once-through fuel cycle was prepared. This draft is the initial effort at assembling pertinent fuel cycle data needed for upcoming evaluations.

ASSESSMENT CONCEPTS AND METHODS -

P. J. Pelto

The three-report set to describe and present user instructions for PNL-developed computer programs for drawing fault trees and for assessing these trees in terms of risk was published. The reports were abstracted in the previous quarterly progress report.⁽¹⁾ Report numbers and authors are repeated below:

- J. L. Carter, ACORN: A Computer Program for Plotting Fault Trees, BNWL-2144, November 1977.
- P. J. Pelto and W. L. Purcell, MFAULT, A Computer Program for Analyzing Fault Trees, BNWL-2145, November 1977.

- G. D. Seybold, RAFT: A Computer Program for Fault Tree Risk Calculations, BNWL-2146, November 1977.

Work began on developing methods (indices) for use in upcoming public/occupational safety studies of alternative waste management schemes for viable fuel cycles.

SYSTEM DEFINITION/ASSESSMENT -

J. W. Voss

A draft systems definition of the once-through fuel cycle was prepared. This draft is the initial effort to assemble pertinent fuel cycle data needed for assessing the safety, cost, and availability of waste management systems. The components of the fuel cycle considered were the reactor, the spent fuel packaging facility, and the spent fuel storage facility. As-generated waste quantities for these facilities were tabulated. Several different low-level waste treatment options were considered, yielding as-treated waste volumes for the reference facilities. A logistics scheme imposed on the system yielded transportation requirements for the fuel cycle. The treatment of each major fuel cycle component is abstracted below.

Reactor

A generic Light Water Reactor (LWR) was considered in this study. As-generated waste quantities were tabulated as available in recent literature.^(2,3) Projections of installed generating capacity and spent fuel availability were made on the basis of present utility plans.

Spent Fuel Packaging

Spent fuel was considered as being packaged in a helium-filled, stainless steel canister. Failed fuel was considered as being overpacked with a second canister. As-generated waste quantities were estimated on the basis of current facility concepts.

Spent Fuel Storage

Packaged spent fuel was considered as being stored in one of four storage concepts: 1) the sealed storage cask, 2) the drywell concept, 3) the air-cooled vault, and 4) the water basin concept. As-generated waste quantities were estimated on the basis of current facility concepts.

Low-Level Waste Treatment

Several different waste treatment options were considered in this study, as listed in Table 8.1. In each case, noncombustible-noncompacted trash and failed equipment were considered to be packaged with minimum treatment.

TABLE 8.1. Alternative Low-Level Waste Treatments(a)

<u>Alternative</u>	<u>Trash Treatment</u>	<u>Wet Waste Treatment</u>
1A	Minimum	Dewater
1B	Minimum	Cement
1C	Minimum	Bitumen
1D	Minimum	Urea-formaldehyde
2A	Compaction	Dewater
2B	Compaction	Cement
2C	Compaction	Bitumen
2D	Compaction	Urea-formaldehyde
3A	Controlled air combustion	Cement
3B	Controlled air combustion	Bitumen
3C	Controlled air combustion	Urea-formaldehyde
4A	Molten salt combustion	Cement
4B	Molten salt combustion	Bitumen
4C	Molten salt combustion	Urea-formaldehyde
5A	Acid digestion	Cement
5B	Acid digestion	Bitumen
5C	Acid digestion	Urea-formaldehyde

a. In every case, noncombustible-noncompactable trash and failed equipment is packaged with minimum treatment.

Logistics

The logistics scheme was based on the location of existing and planned power plants. The spent fuel packaging and storage facilities were assumed to be located in southeastern Washington State. Four burial grounds for low-level waste were assumed (southeastern, midwestern, northwestern, and western U.S.). Packaged low-level wastes were assumed to be shipped



to the nearest operating burial ground. Logistics parameters calcu-

lated included Ci-miles for shipment of low-level wastes and MTHM-miles for shipment of spent fuel.

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9.0 WASTE ISOLATION SAFETY ASSESSMENT PROGRAM

H. C. Burkholder - Project Manager

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

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

SUMMARY

Work during the quarter focused on writing reports about FY-77 work, getting contracts in place for FY-78 work, and continuing the technical activities started in FY-77. Six reports are planned to document the FY-77 work: 1) PNL-2642, Waste Isolation Safety Assessment Program - Technical Progress for FY-77; 2) PNL-2643, Safety Assessment Methods for Geologic Nuclear Waste Repository Systems - Release Scenario Analysis; 3) PNL-2644, Safety Assessment Methods for Geologic Nuclear Waste Repository Systems - Release Consequence Analysis; 4) PNL-2645, WISAP Workshop on Potentially Disruptive Phenomena for Nuclear Waste Repositories; 5) PNL-2646, WISAP Workshop on Release Consequence Analysis Methods for Nuclear Waste Repositories;

and 6) PNL-2647, WISAP Workshop on Geochemical Interaction Data for Geologic Media.

TASK 1 - RELEASE SCENARIO ANALYSIS - J. Greenborg

Work in this task is divided into three subtasks for FY-78: 1) characterization of potentially disruptive phenomena, 2) development of the repository simulation analysis computer code, and 3) development of a geologic/geophysical data base.

A team of 16 geologic consultants was organized during the quarter to provide quantitative descriptions of the potentially disruptive phenomena for nuclear waste repository systems, and the contracting process was initiated. The consultant categories were:

- climatology
- meteorites

- glaciology
- volcanology
- seismology
- man-caused
- structural geology
 - gulf interior
 - Pacific Northwest
 - Southwest
- subsurface hydrology
 - gulf interior
 - Pacific Northwest
 - Southwest
- surface hydrology
- geomorphology
 - coastal
 - Pacific Northwest
 - Southwest.

Development of a second generation repository simulation computer code was begun. It was decided to use the SIMSCRIPT II.5, a simulation language software package, and procurement procedures were initiated.

TASK 2 - WASTE FORM RELEASE RATE DATA - D. J. Bradley

Work in this task in FY-78 is divided into three subtasks: 1) spent unprocessed fuel, 2) doped-glass waste forms, and 3) low-level TRU waste forms.

Spent fuel leaching studies started during the quarter, using the same procedures, leach solutions, and sampling techniques that were used in the doped-glass leaching studies started in FY-77. A detailed chemical and physical characterization of the spent fuel was started.

The doped-glass leaching studies were continued. Solution samples from FY-77 work were started through

the analysis procedure. Experiments were begun to determine the effects of temperature and leachant flow rate.

Studies of the leaching behavior of TRU incinerator ash solidified in concrete, bitumen, urea-formaldehyde, and polymer were initiated. The initial work focused on the manufacture and characterization of the various waste forms.

TASK 3 - RELEASE CONSEQUENCE ANALYSIS - J. R. Raymond

Work in FY-78 is focusing on conversion and interfacing of the computer codes selected in FY-77 for use in the WISAP release consequence system. These codes already exist in usable forms, and the thrust of the work here is aimed at getting them operational on the new Hanford computer and at increasing their usefulness. All converted codes will be subjected to a series of test problems; and documents describing the codes and the results of the tests will be written.

During the quarter the conversion and testing of the DAVIS FE model (a three-dimensional finite element hydrologic code) was completed on the PDP 11/45 computer. Work to interface the DAVIS FE and MMT models (a three-dimensional contaminant transport code) was initiated.

Conversion of PATHS, GETOUT, ARRRG, FOOD, DACRIN, KRONIC, and SUBDOSA to the Univac 1100/44 computer was started. Several improvements in the PATHS model (a two-dimensional hydrologic code) and the GETOUT model (a one-dimensional

contaminant transport code) were made simultaneous with the conversion. Work to interface these two codes will begin later in the year. All of the dose codes were slightly modified to use common data libraries.

TASK 4 - TRANSPORT DATA - R. J. Serne

Comments on draft FY-77 progress reports were incorporated into final documents. Program changes suggested during the September 1977 workshop were considered and subsequent adjustments made in plans.

Prior to selecting a standard procedure for the experimental determination of K_d , efforts continued in assessing the applicability and developing time and cost estimates for five methodologies: batch, flow-through column, axial filtration, channel chromatography, and high-pressure intact core. Preliminary findings from analysis of the experiments performed in FY-77 follow. The batch method is proving the least costly method for measuring the affinity of nuclides for rock particles and wafers. The accuracy of the batch method and effects of hydrodynamic dispersion need further study. The channel chromatograph can be used only for nuclides with low affinity for sorption. Flow-through column experiments often yield non-chromatographic effluent curves which complicate the interpretation of data. Because some experimenters observe ideal behavior, these anomalous effluent curves need more study to ascertain whether improper experimental techniques or kinetic-dominated

scaling effects could be the problem. High-pressure intact core K_d experiments will be costly and may also suffer from scaling effects.

The compilation of K_d values for the nuclides plutonium, americium, neptunium, iodine, technetium, strontium, cesium, europium, ruthenium, cerium, uranium, antimony, and cobalt in the presence of various rocks, minerals, and groundwaters continued. By the end of FY-78 enough data should be available to make preliminary estimates of the migration potential of long-lived radionuclides in hypothetical environments. Current plans are that by the end of FY-79 the generic data bank will be complete and attention will switch to site-specific studies and sorption mechanism studies. The adaptive learning network statistical analyses continued on PNL-generated K_d data to provide numerical predictor equations which account for the dependency of K_d on rock and groundwater characteristics.

Compilation of thermodynamic data on mineral stability and solution speciation of long-lived nuclides continued. The thermodynamic data will be used to construct diagrams relating the expected long-time (geologic ages) fate of radionuclides to the results of short-term laboratory tests.

Work also continued on experimental verification of PuO_2 and $\text{Pu}(\text{OH})_4$ solubilities in natural waters at various pH's. The valence state speciation of the resultant soluble plutonium is also being determined to develop a fundamental understanding of sorption-desorption processes.

Work was started to document the Seattle workshop and assemble FY-77 technical progress reports into a final document complete with executive summary.

TASK 5 - SOCIETAL ACCEPTANCE -

A. H. Schilling (HARC)

A review paper, Results of Research on the Social Impacts of Nuclear Waste Isolation Being Conducted by the Battelle Human Affairs Research Centers, was completed; it is intended chiefly for internal use by HARC researchers in waste management. Also, the following papers were completed in draft form and submitted to the Office of Waste Isolation for review and comment:

- Restriction of Surface and Sub-surface Property Rights in Selected States, a review of the legal issues in acquisition of property rights needed for a nuclear waste repository
- Chronology and Analysis of Major Events Surrounding the Proposed Radioactive Waste Disposal Facility at Lyons, Kansas - 1954 to 1972
- Case Study of Public Acceptance of High-Level Nuclear Waste Management at the Hanford Atomic Works, 1943 to 1977.

Working draft reports were prepared, reviewed, and written as discussion papers. These reports deal with the following areas:

- Compensation as One Response to the Unintended Effects of a Government Program: The Case of Nuclear Waste Management Facilities. This paper analyzes the issues involved

in providing compensation for the costs of developing nuclear waste repositories. The paper treats the social costs of repository development (e.g., impacts on the community at large rather than those involved in the relocation of particular families and businesses) and discusses a state and local regulatory framework for repository development.

- A Demographic Model for Assessing the Socioeconomic Impacts of Large-Scale Industrial Development Projects. This is an analysis of the ability of state and local officials to manage adverse socioeconomic impacts that would accompany repository development. A discussion draft paper, "The Management of Social and Economic Impacts Anticipated with the Development of a Nuclear Waste Repository: A Preliminary Discussion," has been prepared. The paper briefly reviews potential social and economic impacts from repository development, describes existing Federal programs that can be used to mitigate such impacts, analyzes the process of obtaining Federal assistance under such programs, describes planning strategies available to state and local officials with management responsibilities, and finally provides an overview of the planning process available for impact management.
- Public Issues in Nuclear Waste Management. This paper reviews the literature on waste management in order to identify the existing public issues. Clustering techniques are applied to define

categories and to provide some discussion of relationships among various issues.

- The Management of Social and Economic Impacts Anticipated with a Nuclear Waste Repository: A Preliminary Discussion. This model,

available now, projects both population changes and the resulting increases in demand for social services attributable to the development of a repository.

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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

Analyses of parameters controlling the transport of radionuclides through partially unsaturated geohydrologic shallow land burial media are continuing. Sediment-radionuclide chemical reactions and kinetics have been determined from macrocations, strontium, cesium, and technetium. Liquid and vapor phase flux parameters have also initially been determined. Isothermal and nonisothermal flux determinations have been made for evaluating temperature dependence of water flow and determining energy balance functions in porous media.

BACKGROUND

Radioactive contaminants have been disposed to shallow land disposal sites in considerable quantities since the advent of plutonium production. Sometimes these materials have been transported into the hydrologic system, to become potential risks within the aquatic and terrestrial ecosystems. The objective of this program is to monitor liquid and vapor phase fluid movement and the transport of radionuclides and as-

sociated waste materials within the unsaturated hydrologic domain. To accomplish this objective, available instrumentation methods are being evaluated, optimum monitoring systems defined, and new data collection and analysis systems developed applicable to transport monitoring.

Specific tasks contributing to the overall program objectives are listed as follows:

Evaluation of Monitoring Systems

This task, to identify optimum systems for use at various burial facilities in monitoring radionuclides, liquid and vapor phase fluids, and other waste constituents, was completed in FY-1977.

Laboratory Analysis

This task is to review and develop laboratory techniques for determining unsaturated porous media flow and nuclide adsorption. Characterization of flow parameters by laboratory methods is required when predictive models and field evaluations of the flow system are made. Laboratory analysis is the only cost effective preliminary procedure for site selection and optimization of site engineering. This task will also develop new methods for, and conduct laboratory

determinations of, unsaturated flow conditions potentially encountered in various ecological provinces where burial of radioactive solid waste is practiced. The data and computational techniques developed by this task will be formulated into a working data evaluation package for use by investigators at other national laboratories' solid waste burial facilities.

Physical Modeling of Contaminant Transport

This task is to complement laboratory analyses. Physical modeling of the unsaturated zone beneath solid waste burial sites will allow investigators to assess the applicability and operational integrity of monitoring instrumentation. Also, the spatial distribution of contaminants with time can accurately be determined under controlled environmental conditions.

The cases to be modeled will depend on the complexity of flow systems found at waste burial facilities. Physical models will permit accelerated testing of the unsaturated flow domain beyond those conditions encountered in the field. The data obtained from these models will be used to optimize design of field monitoring instrumentation and to verify field monitoring tests.

Field Monitoring and Data Evaluation

The primary objective of this task is to monitor and evaluate fluid and radioactive transport in the unsaturated system at an active burial facility or a number of burial facilities in differing climatic zones. A comprehensive suite of monitoring instrumentation networks and data

collection systems needs to be specified for each site to be evaluated. Monitoring networks must be installed under conditions set forth by previous tasks and tasks evaluated under cooperating programs. Data collection frequencies and recording modes will be evaluated to define the input needed for hydrologic modeling of the unsaturated system. Collection and evaluation of data from these systems during this program will define the criteria which may be applied in selecting repositories for solid waste and liquid waste.

LABORATORY ANALYSIS - R. J. Serne, D. Rai and S. J. Phillips

Preliminary characterizations of sediment physico-chemical properties affecting the transport of radionuclides under partially saturated groundwater conditions are being conducted. Analyses extended to cover evaluation of transport parameters in the partially saturated geohydrologic system require development of advanced methodologies and analytical apparatus. Two generic sediment types obtained from an arid site have in part been analyzed for the following variables: cation exchange capacity; saturation extract; mineralogy; ability to sorb technetium, cesium, and strontium; and hydraulic conductivity-diffusivity and retentivity of partially saturated soils.

Table 10.1 lists sediment chemical characteristics measured. All parameters are needed as input values to an existing computer code which models the chemistry (ion exchange,

TABLE 10.1. Sediment Characteristics

Characteristic	Value	
	Sediment A	Sediment B
Saturated paste pH	8.83	8.23
Moisture % at saturation	23.8 ± 0.7	16.6 ± 0.4
Saturated paste composition, ppm		
Na ⁺	149	9
K ⁺	1.8	5.6
Ca ⁺⁺	25	37
Mg ⁺⁺	4.4	5.6
Cl ⁻	30.4	0.8
SO ₄ ⁼	185	7.5
NO ₃ ⁻	2.7	6.4
CO ₃ ⁼	10	5.0
HCO ₃ ⁻	179	133.1
Cation exchange capacity, meq/100 g	12.0 ± 0.1	5.0 ± 0.1
Distribution on exchange, meq/100 g		
Ca ⁺⁺	6.6 (55%)	3.5 (70%)
Mg ⁺⁺	3.8 (32%)	1.0 (20%)
K ⁺	0.2 (1%)	0.4 (8%)
Na ⁺	1.4 (12%)	0.1 (2%)
Calcium carbonate content, %	1.38 ± 0.17	0.07 ± 0.02
Organic carbon content, %	0.14 ± 0.0	0.10 ± 0.00
Particle size distribution, %		
Sand	45	87
Silt	44	9.3
Clay	11	3.7
Clay mineralogy	Smectite (poorly crystallized), minor amount of mica, trace of kaolinite	

complex formation and precipitation-dissolution) controlling the macroconstituents (calcium, magnesium, sodium, potassium and anions) in solutions percolating through sediments. Routson and Serne have modeled the movement of strontium and cesium radionuclides in selected sediment as a function of the macrocations found in solution.⁽¹⁾ Thus for the selected media the macrocation reactions are modeled and the fates of trace waste

radionuclides are predicted from empirical relations dependent upon the macroconcentrations. This task is being performed to learn whether similar techniques can be developed and used to evaluate and to model the movement of radionuclides under partially saturated conditions from shallow land burial sites.

Screening experiments were performed using the radionuclides technetium, strontium, and cesium. Six

synthetic groundwaters were prepared for contact with each sediment. The first solution contacting each sediment had the composition of the saturation extract. This composition represents the groundwater which would first occur when water saturates the arid sediments under study and begins to percolate. The second solution, a 5 to 1 dilution of the saturated extract, represents a scenario where a large amount of water contacts the arid sediment, dissolves all the evaporates, and percolates through the sediment. The third solution was five times as concentrated as the saturation extract. If unsaturated conditions prevail in the sediment, percolating waters could dissolve large amounts of evaporites and build up salt concentrations greater than the saturated extract. The fourth through sixth solutions are similar to the first three solutions except that the anion contents were satisfied by using

chloride. The fourth through sixth solutions were used to simplify the experiments by minimizing complexation of the cations with bicarbonates-carbonates and sulfates. The intention was to simplify the chemical reactions occurring and allow cation competition effects to be observed and studied. For the concentrated solutions representing five times the saturation extract, precipitates formed, lowering the total salt concentration. Table 10.2 shows the measured concentrations of the synthetic groundwaters.

The distribution coefficients (K_d) for technetium, strontium and cesium for the various solutions contacting sediments "A" and "B" were determined in triplicate for two contact times, 8.5 days and 21.5 days. The pH's of the solutions were measured at these times also. Table 10.3 lists the average values for the K_d 's and pH's.

The negative K_d values for technetium are an artifact of the experimentation. They represent cases in which

TABLE 10.2. Composition of Solutions Contacting Sediments

<u>Solution</u>	<u>Na</u>	<u>K</u>	<u>Ca</u>	<u>Mg</u>	<u>HCO₃</u>	<u>CO₃</u>	<u>SO₄</u>	<u>Cl</u>	<u>NO₃</u>
ppm									
Sediment A									
1	113	1.2	15	3.6	80	10	200	8	2
2	17	0.6	9	1.8	31	0	39	2	0.5
3	550	6.2	49	12.6	232	50	950	34	13
4	138	1.3	30	5.1	---	--	---	271	---
5	26	0.4	11	2.1	---	--	---	57	---
6	760	6.7	126	18.3	---	--	---	1485	---
Sediment B									
1	5.8	3.7	4.2	3.4	32	8	7	0.5	6
2	1.0	1.1	4.1	1.3	16	0	4.5	<0.5	1
3	23	17	6.4	11.3	80	24	36.5	0.8	32
4	7.4	1.2	38	5.6	---	--	---	99	---
5	1.8	0.6	8	1.3	---	--	---	21	---
6	46	6.2	185	22.6	---	--	---	474	---

TABLE 10.3. Sediment K_d and pH Values

Solution	$K_{d_{Tc}}$		$K_{d_{Sr}}$		$K_{d_{Cs}}$		pH	
	8.5 days	21.5 days	8.5 days	21.5 days	8.5 days	21.5 days	8.5 days	21.5 days
Sediment A								
1	-0.38 ± 1.51	-2.77 ± 0.23	133 ± 13	112 ± 1	5307 ± 100	13,525 ± 4,741	8.13 ± 0.11	8.26 ± 0.03
2	-0.88 ± 0.52	-1.13 ± 2.38	238 ± 21	181 ± 12	3377 ± 1311	6,432 ± 1,219	7.95 ± 0.09	8.07 ± 0.03
3	-1.36 ± 0.71	-0.04 ± 0.21	40 ± 3	38 ± 8	3730 ± 1387	7,298 ± 1,098	8.60 ± 0.01	8.52 ± 0.11
4	0.41 ± 0.34	0.57 ± 0.17	86 ± 8	74 ± 3	8832 ± 3417	8,217 ± 2,380	7.56 ± 0.02	7.78 ± 0.03
5	-2.34 ± 0.98	0.54 ± 0.65	222 ± 28	207 ± 18	5995 ± 3632	6,033 ± 1,534	7.72 ± 0.02	7.90 ± 0.02
6	-0.81 ± 0.39	-0.52 ± 1.20	17 ± 1	21 ± 3	4233 ± 1321	11,522 ± 2,154	7.65 ± 0.03	7.75 ± 0.03
Sediment B								
1	0.54 ± 1.61	0.07 ± 0.32	129 ± 15	114 ± 9	2042 ± 401	1,505 ± 320	7.50 ± 0.09	8.14 ± 0.02
2	-0.02 ± 0.99	-1.62 ± 0.60	170 ± 27	132 ± 11	1388 ± 226	1,094 ± 295	7.46 ± 0.04	8.00 ± 0.02
3	-0.85 ± 0.32	-0.31 ± 0.30	31 ± 3	38 ± 1	2291 ± 819	3,016 ± 722	7.25 ± 0.05	8.25 ± 0.08
4	1.27 ± 1.67	0.06 ± 0.68	47 ± 4	39 ± 2	3405 ± 1215	5,805 ± 2,109	8.08 ± 0.01	7.63 ± 0.11
5	-0.96 ± 0.37	0.52 ± 0.51	36 ± 2	37 ± 1	5708 ± 2635	3,045 ± 377	7.91 ± 0.01	7.57 ± 0.05
6	-0.09 ± 0.76	0.38 ± 1.23	9 ± 3	12 ± 2	3215 ± 470	3,253 ± 80	8.31 ± 0.06	7.44 ± 0.02

the average activity in blank tubes (without sediments) is less than the activity in the sediment suspension effluents. Negative K_d values occur only in systems where sediment sorption is extremely low. In this circumstance, counting statistics becomes very important because the effluent and influent activities are nearly equal. In the calculation of K_d the effluent is subtracted from the influent, and similar values can lead to a negative difference. For transport calculations, a negative K_d value should be equated to zero and thus the radionuclide should travel at the same velocity as the water. The very low technetium adsorption occurs because the technetium species which is stable in the Eh-pH field common for shallow land burial sites is TcO_4^- anion. Sediments low in organic content commonly have very low anion exchange capacity and allow anions to percolate relatively unimpeded. It is possible that some of the negative K_d values for tech-

netium are caused by anion exclusion by the inner surfaces of the clay minerals present in the sediments. This possibility will be investigated using flow-through column experiments. The data in Table 10.3 show no relationship between different solution compositions, pH's or times and the $K_{d_{Tc}}$. There is also no significant difference in $K_{d_{Tc}}$ for the two sediments studied.

The strontium sorption data show that high sorption occurs on both sediments for the saturation extract solutions. The first three solutions, which include anions capable of forming soluble complexes with strontium, show greater sorption than the three chloride solutions. This has been observed for other sediments and may reflect strontium exchange with calcium in precipitates such as $CaCO_3$. It is also known that strontium sorption increases with increasing solution pH. The simulated saturation extracts averaged 0.3 to 0.9 units pH larger than the chloride solutions.

This might be part of the cause for the greater sorption of strontium. For both sediments the K_d for strontium shows significant cation competition effects. The sorption of strontium decreases as the cation content of solutions increases, as would be expected from simple ion exchange theory. Sediment A, with more fine material and a larger cation exchange capacity (see Table 10.1), shows higher K_d for strontium, which would be expected from ion exchange principles. Strontium sorption appears to be complete after 8 days and from other experiments on Hanford sediments has been found to reach steady state within 48 hr.

The cesium sorption data show very high sorption for both sediments for all the solutions studied. There appear to be no significant cation-competition effects occurring between the solutions. All solutions show very high sorption. For sediment B the simulated saturation extract solutions show lower K_d values for cesium than do synthetic solutions containing only chloride. This suggests possible formation of soluble complexes. Cesium is not known to form any strong complexes with bicarbonate, carbonate or sulfate anions; therefore the observed differences cannot readily be explained. The very large sorption of cesium and a lack of ion exchange-like cation competition suggest that the clay material (especially the mica) may be preferentially fixing the cesium. The reversibility of the cesium sorption should be investigated because illitic

clays are known to irreversibly fix cesium so that it desorbs very slowly and only partially.

The screening experiments suggest:

- Anions will move as rapidly as water through the Hanford sediments under shallow land burial conditions.
- Strontium sorption is controlled by ion exchange-like reactions and can be modelled by the techniques described by Routson and Serne.⁽¹⁾
- Cesium is very strongly bound at the conditions studied and probably will not be transported more than a few feet before decaying to undetectable levels. It also appears that the methods described by Routson and Serne⁽¹⁾ should not be used to generate Kd_{CS} data needed for transport models.

From the radionuclide sorption screening experiments, one can estimate that:

- Elements such as technetium, iodine, and antimony might be mobile (move at the speed of the carrier water).
- Strontium would be retarded significantly (move about 250 times slower than water).
- Cesium could be extremely retarded (move about 3000 times slower than water).
- Rare earths such as europium, cerium, praseodymium, and samarium would be retarded significantly by precipitation reactions; i.e., at alkaline pH's rare earths are very insoluble.

Isothermal and nonisothermal effects of gas and liquid flux through

sediments are being evaluated to quantitatively define mass and energy balance relationships within burial site media.

Studies to date have shown that the thermal conductivity of both sediments studied (A and B) varied linearly with water content over the measurement range. Table 10.4 shows these relationships.

The thermal conductivity relationships for sediments A and B are nearly parallel. This suggests that even for widely different textural materials the thermal conductivity can be predicted over at least part of the water content range by similar relationships if the dry thermal conductivity is known. Further testing will be required to evaluate whether or not similar relations hold for other sediments.

Table 10.5 shows the temperature dependence of thermal conductivity for sediment B as a function of water content.

These data should be considered tentative, and further testing is needed. However, if these results are valid, the present theory of the effects of vapor transport on thermal conductivity (Philip and DeVries, 1957)⁽²⁾ will require modification. Some recent field studies suggest that the thermal conductivity of partially saturated sediments is not as

TABLE 10.5. Temperature Dependence of Thermal Conductivity of Sediment B

Water Content, cm ³ /cm ³	Thermal Conductivity, cal (cm °C) ⁻¹ x 10 ⁻⁴		
	12°C	26°C	36°C
0.0344	8.3	7.7	7.6
0.0525	9.8	9.2	7.7
0.0947	17.5	--	13.4
Oven dry (0)	5.8	5.6	--

sensitive to temperature as was originally supposed (Kimball et al., 1976).⁽³⁾ Effects of hysteresis, sampling error, and other complications all contribute to the difficulty in measurement and to the apparent insensitivity of thermal conductivity to temperature.

The results from several tests of capillary models for conductivity in unsaturated soils indicate that it is very sensitive to such things as water retention characteristics and matching factor values. Campbell's model (1974), which uses a log-log relationship of the sediment water characteristic, is highly sensitive to the range over which the computation is made. Table 10.6 shows how the slope parameter affects the calculated conductivity.

Data from Table 10.6 suggest that care must be used in selecting the range of water contents over which to compute values; i.e., the exponential term b must be evaluated over a

TABLE 10.4. Thermal Conductivity Versus Water Content

Soil	Texture, %			Relationship	Water Content Range θ, %	Linear Correlation R ²
	Sediment	Silt	Clay			
A	45	44	11	kt = -0.426 + 0.968 θv	4-40	0.996
B	87	9	4	kt = 5.64 + 0.979 θv	0-30	0.964

TABLE 10.6. B Sediment Unsaturated Conductivity^(a)

Tension h, cm	Water Content, cm/cm	Water Conductivity, cm/min	
		b = 1.74 ^(b)	b = 9.87 ^(c)
10.5	0.273	11.0 x 10 ²	3.2 x 10 ⁴
13.5	0.249	5.9 x 10 ²	---
16.5	0.231	3.7 x 10 ²	6.7 x 10 ⁶
19.5	0.198	1.4 x 10 ²	---
22.5	0.188	9.6 x 10 ³	---
25.5	0.178	6.8 x 10 ³	---
28.5	0.153	2.5 x 10 ³	5.5 x 10 ⁻¹⁰
100	0.071	1.8 x 10 ⁻⁵	---
500	0.050	1.9 x 10 ⁻⁶	7.7 x 10 ⁻²¹
5,000	0.046	1.0 x 10 ⁻⁶	---
10,000	0.045	9.4 x 10 ⁻⁷	---
15,000	0.041	4.9 x 10 ⁻⁷	2.0 x 10 ⁻²³

a. Calculated after method of Campbell (1974) where $k(\theta) = k_s (\theta/\theta_s)^{2b+3}$ and $k_s = 1.09$ cm/min; $\theta_s = 0.39$.⁽⁴⁾

b. Valid in range of $h = 10-40$ cm.

c. Valid in range of $h = 100-15,000$ cm.

limited range of water content. Also the extension of wet range data to dry range is questioned. Further testing will be necessary to establish the ranges over which computed conductivities are valid for given sediments.

Several analytical expressions for water characteristics have been developed. None seem entirely satisfactory, due primarily to the complicated pore geometries of natural sediments. Sediment water characteristics of the form

$$h(s) = a [e^{b(s-1)} - 1]$$

where

s = degree of saturation - θ/θ_s

a, b = empirical constants

θ_s = water content at saturation

are currently being tested to determine the appropriateness for use in describing water characteristics of sediments A and B.

PHYSICAL MODELING OF CONTAMINANT

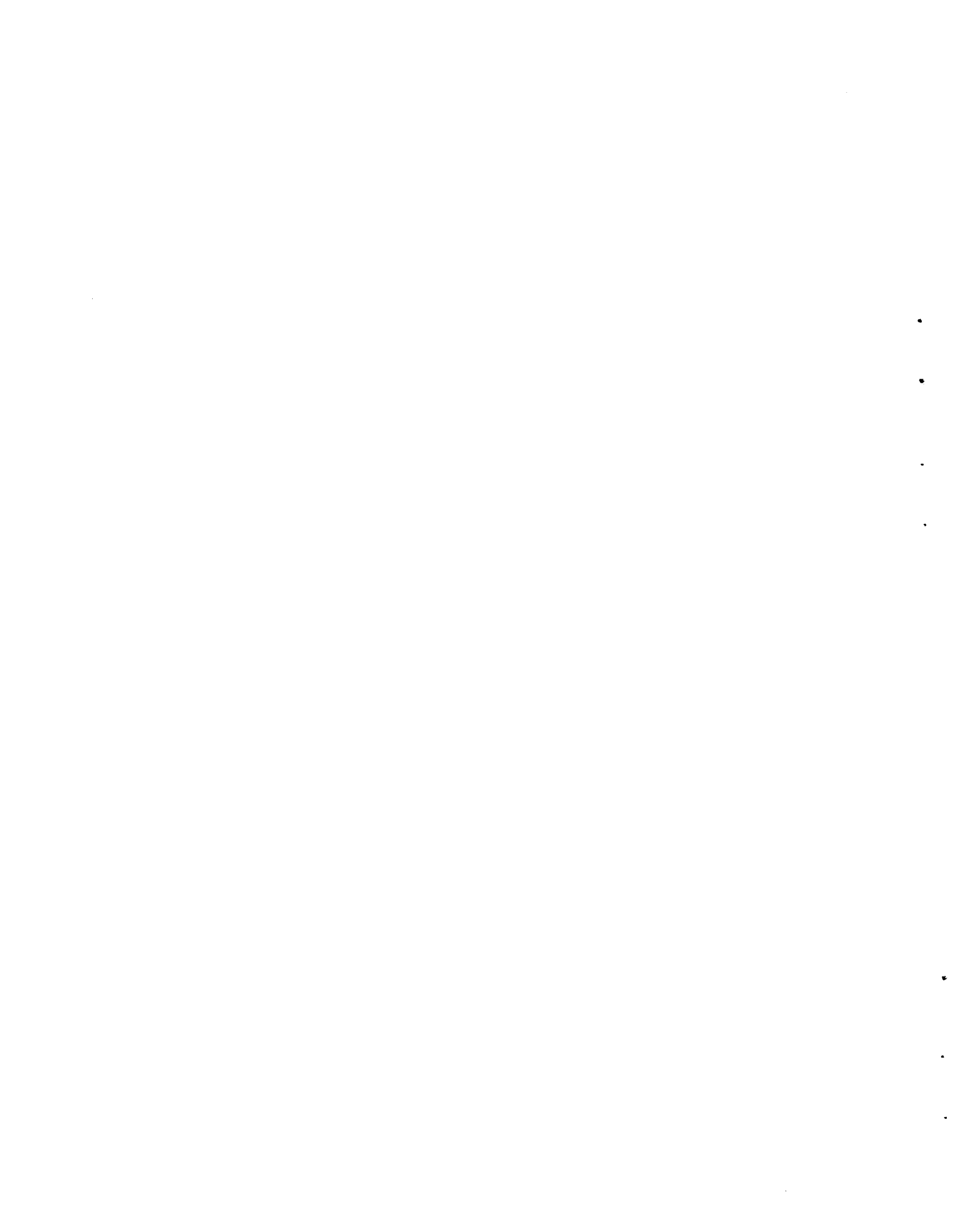
TRANSPORT - L. D. Kannberg and

S. J. Phillips

One-dimensional physical modeling of fluid flux has been started using short-column test cases. Gamma ray attenuation analysis of density and fluid content has been used employing a collimated 0.25-Ci ¹³⁷Cs source and NaI (Tl) multichannel analyzer. This system allows maximum stability of counting over intermediate and long-term infiltration and drainage conditions.

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11.0 DETECTION AND CHARACTERIZATION OF MOBILE ORGANIC
COMPLEXES OF FISSION PROJECTS

J. A. Franz - Project Manager

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

SUMMARY

A first draft was completed of a PNL Report on the results of the review of the aqueous and soil chemistry and the stability data of zirconium, nickel, technetium and niobium organic complexes. Soil column absorption studies of nickel gluconate indicate that flow rate and concentration significantly affect absorption. Batch test tube studies of zirconium-tartrate absorption were performed to determine the significant variables by statistical experimental design. These results indicate that concentration of zirconium is the only significant variable.

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

A first draft of a PNL report on the results of our review of the chemistry and the stability data of zirconium, nickel, technetium, and niobium complexes was completed during this quarter. This work reviews sources of complexing agents and their structures, discusses mechanisms of the formation of complexes

and the relevant coordination chemistry and soil behavior of each element, and recommends future methods for containment of radioactive isotopes.

Based on the available information gathered in this review, zirconium, niobium, technetium, and nickel are all mobile in the presence of organic complexing agents to some extent, given sufficient organic materials and assuming a formation mechanism. The following predictions are based on available data or on our experimental results:

- Zirconium, niobium, and nickel organic complexes all are absorbed less by soil than are the uncomplexed species.
- Synthetic organic chelating agents and α -hydroxycarboxylic acids form significantly stable complexes with zirconium, depending on the pH of the aqueous medium. This can result in high mobility in soil systems.
- Niobium complexes form most efficiently with salts of synthetic organic chelating agents and α -hydroxycarboxylic acids but hydrolyze when excess chelate is not present.

- Nickel-DTPA and nickel-EDTA complexes are more mobile in soil systems than are uncomplexed nickel species. We predict that other synthetic organic chelating complexes of nickel would also be more mobile.
- Technetium organic complexes are potentially as mobile in aqueous soil systems as is uncomplexed technetium.

The properties of complexes formed in the soil environment are what determine the need for general procedures to contain these radionuclides. Procedures that both minimize the formation and decrease the mobility of complexes are indicated:

- Construct physical barriers to rainwater percolation.
- Curtail the burial of degradable organic material in the presence of fission and activation products.
- Curtail the burial of synthetic organic chelating agents in the presence of fission and activation products. This would significantly decrease movement of all radionuclides.
- Reduce organic complexing ability of nickel, zirconium, niobium and, potentially, technetium by treatment of waste with alkali, followed by firing (to eliminate organic complexing agents and form anhydrous zirconium and niobium oxides).

Further studies needed to clarify the role of complexing agents in mobilizing zirconium, niobium, nickel, and technetium include:

- the role of polymerization and hydrolysis of zirconium and

niobium in affecting compound structure, stability, and complex charge and the resultant absorption by soil

- the mechanism of precipitate solubilization and soil mineral replacement in mobilizing zirconium and niobium hydrous oxides
- the stability and water solubility of mixed metal-mixed ligand complexes, especially those involving zirconium and niobium
- the stability of technetium complexes at high pH with time, and the resultant absorption by soil.

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

Soil absorption of nickel-gluconate was studied; elution curves are depicted in Figure 11.1. The experiments were done at three different flow rates (1 ml/min, 2 ml/min, 0.1 ml/min) and with two different concentrations of complex (1×10^{-4} moles and 1×10^{-6} moles). These experiments indicate that flow rate and complex concentration could have significant effects on absorption of nuclides by soil. Given a slow enough flow rate and a small enough concentration, all of the nickel-gluconate could be absorbed.

To assess the amount of soil required to absorb an organically bound nuclide and to compare the absorption of an inorganic nuclide to the organic nuclide, one determines K_D values. Two general experimental methods are used in determining K_D values: column or batch. The

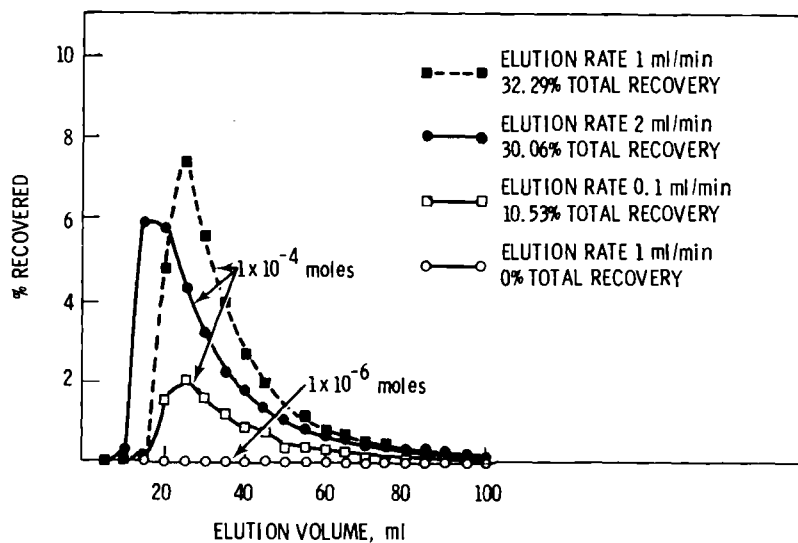


FIGURE 11.1. 1:1 Nickel-Gluconate Elution by Distilled Water,

column method employs a soil-packed column. A water solution containing the nuclide is passed at a constant flow through the soil in the column. The effluent is monitored. The batch method involves shaking the nuclide solution and soil until equilibrium is attained. The supernatant is monitored. The column experiment washes away any water-soluble soil ions which are vitally present in the batch experiment. The batch method allows colloidal nuclide to remain in the sampled supernatant which might be filtered out by soil in the column experiment.

The use of K_d values in predicting the behavior of organically bound nuclides is more complicated than in predicting that of inorganic ones. The organic molecule can bond very strongly with the nuclide to form a variety of charged and uncharged complexed species. Uncharged and anionically charged species will not be exchanged onto the anionically

charged soil sites. Even though other mechanisms may be involved in soil-nuclide interaction, the decrease in the cationic exchange mechanism will cause some organically bound nuclides to elute directly through the soil whereas other uncomplexed and complexed species will be absorbed. The K_d value assumes a uniform absorption or nonabsorption behavior of a nuclide from which larger amounts of soil and/or nuclide can be extrapolated; thus unaltered K_d values of organically bound nuclides having nonequilibrium may not be valid. These problems can be circumvented by analyzing these variables and incorporating them into the K_d value for each complex.

Hence, during this quarter we designed a statistical experimental strategy for determining the most important variables for a given organically bound nuclide and for determining a K_d value. To complement the flow studies we decided to use

the batch method with a centrifuging step at the end of equilibrium.

Screening experiments are the first step in statistical experimental strategy. Therefore we performed screening experiments to determine the important variables of zirconium-tartrate absorption by soil. The pH and complex concentrations were determined as the most statistically significant variables of the six variables studied. From these experiments our major conclusions are:

- Zirconium-tartrate absorption is not affected by the presence of excess ligand.
- Zirconium-tartrate absorption does not reflect the presence of a discrete nonabsorbable complex species.
- Preliminarily it appears that zirconium-tartrate is absorbed more strongly by soil at pH 8.0 than at pH 6.0.
- Zirconium-tartrate absorption as reflected in K_d values is greater at a concentration of 10^{-5} than

at 10^{-4} moles. Further study is needed to determine whether this phenomenon may be inherent in the calculation of K_d .

- Filtering of the supernatant in these batch studies does not change the experimental results. Zirconium-tartrate apparently does not form a filterable noncentrifugable colloid.

During this quarter, additional studies of the aqueous chemistry of zirconium revealed that not only do alkali precipitation and acid solubilization change zirconium's complexing ability, but that drying in the presence of concentrated acid also does so. The latter procedure can also make zirconium insoluble. This information makes it more evident that the studies in the literature on controlled solutions of zirconium where zirconium appears to be in an unpolymerized, unhydrolyzed state are inadequate to allow prediction of zirconium's behavior under conditions of uncontrolled waste burial in soil.

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

SUMMARY

A vibratory finisher has been obtained for the pretreatment facility to remove paint, grease, corrosion layers and gross contamination from components before electropolishing. Testing of the vibratory finisher has shown promising results; e.g., some items pretreated in this way were decontaminated to the extent that they did not require electropolishing. An important advantage of the vibratory finisher is that the contamination removed by the abrasive action is continuously washed out, leaving the equipment and media clean and relatively uncontaminated.

Other progress in the pretreatment facility includes installation of the glove-box line. Construction is under way on the stainless steel walk-in hood, and a pair of master-slave manipulators and two high-pressure water/solvent spray systems have been received.

Two types of plasma arc cutting torches have been obtained: a larger, 400-A, water-injected torch and a smaller, 100-A, dry torch. Initial plasma arc sectioning studies

will use the small torch because of indications that 1) the thickness of the recast layer for plasma arc cut metal can be minimized by using the small torch and 2) the low heat input and reduced cutting width will minimize metal loss.

DEMONSTRATION FACILITY

Modifications have continued to improve the effectiveness of the demonstration facility. Following the failure of a rotary seal pump and a sealless magnetically coupled pump for agitation/filtration of the electrolyte, a third type of acid pump has been evaluated. Special features of this 189-ℓ/min (50-gal/min) pump include a better type of pump motor coupling to reduce alignment problems and acid-resistant alloy seals that can be pressurized and flushed with electrolyte or water to reduce wear and eliminate leakage. This pump has been operating successfully in the electropolishing facility for three months.

Two sizes of a bearingless submersible pump also were obtained for evaluation. The larger pump (83 ℓ/min

or 22 gal/min) will be tested for service with the electropolishing facility. The smaller pump (11.4 l/min, or 3 gal/min) will be evaluated for in situ decontamination applications.

PRETREATMENT FACILITY

Although work had been delayed due to a boilermaker's strike, the glove-box line has been installed in the pretreatment facility. It consists of three glove boxes for bead blasting, degreasing, and disassembly interconnected by a conveyor system, which also leads to the sectioning area and the electropolishing facility.

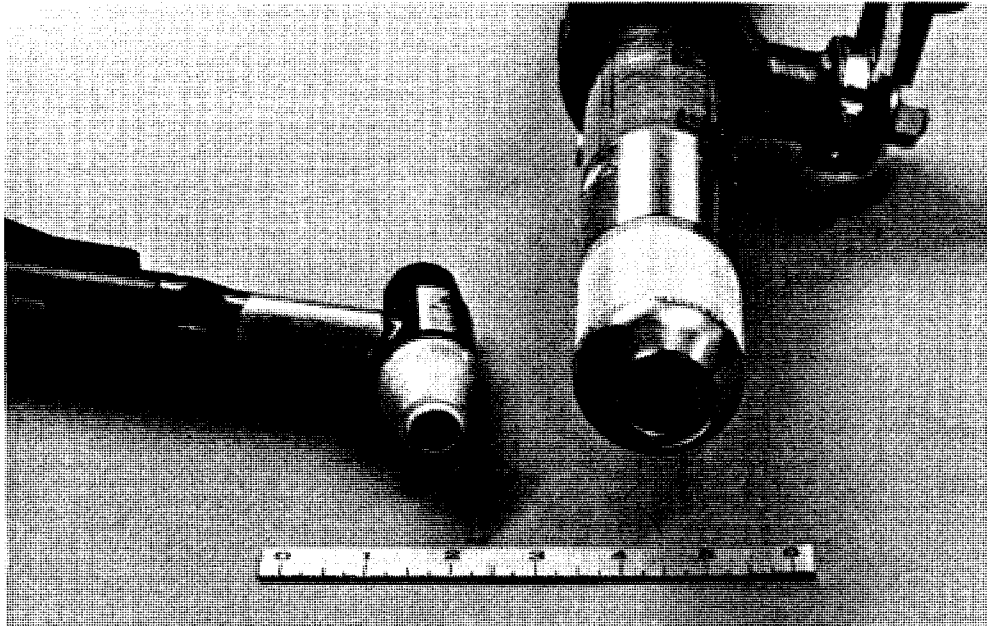
Construction is also under way on the stainless steel walk-in hood for the sectioning/pretreatment facility. Drawings have been received from the designers for the door, which will have inflatable gaskets. An overhead crane for this facility is also on order.

A pair of master-slave manipulators has been acquired to minimize personnel exposure during the disassembly, sectioning, and pretreatment operations. These units are capable of lifting 45.5 kg (100 lb) each and have a special hooking device (load hook) to facilitate hoisting operations.

Two high-pressure water/solvent spray systems have been received capable of delivering a 12-l/min (3.2 gal/min) stream through an adjustable nozzle at a pressure of 3.5 MN/m^2 (500 psi). One of these systems has been used to pretreat some highly contaminated piping for the electropolishing facility. The other one has been used on a portable cart in

various areas of the laboratory to test its effectiveness, for example, in removing difficult oil and grease from vacuum pumps. The results have been highly successful. These systems have the capability to spray detergents, detergent/water solutions, or water rinses to meet whatever pretreatment needs are required.

Two different types of plasma arc torches have been obtained, and a plasma arc cutting test facility is being installed for use in sectioning studies and to provide test material for other pretreatment studies. The larger, 400-A, water-injected torch is capable of cutting thicker materials at faster speeds (up to 508 cm/min or 200 in./min) than is the smaller, 100-A, dry torch, which cuts 0.64 cm (0.25 in.) stainless steel at 102 cm/min (40 in./min) (Figure 12.1). However, studies have shown that the thickness of the recast layer for plasma arc cut metal can be minimized by using the small torch (small nozzle orifice and low heat input) and operating at the maximum cutting speed for each metal thickness. In addition, a wafer muffle can be added to the small torch to help contain dross and contamination and to reduce noise and ultraviolet radiation from the cutting operation. Also important is that the low heat input and reduced cutting width of the small torch will minimize metal loss (secondary waste). Calculations show that metal loss using the 100-A torch should be less than 2 wt% for sectioned plates 61 by 61 cm (2 by 2 ft) or larger. Consequently, initial plasma arc sectioning studies will use the small torch. It will be mounted on a heavy-duty



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FIGURE 12.1. Plasma Arc Torches to be Used in the Pretreatment Facility: 100-A Dry Torch (Left) and a 400-A Water-Injected Torch (Right)

machine carriage capable of moving a torch at controlled speeds in a straight line up to 508 cm/min (200 in./min).

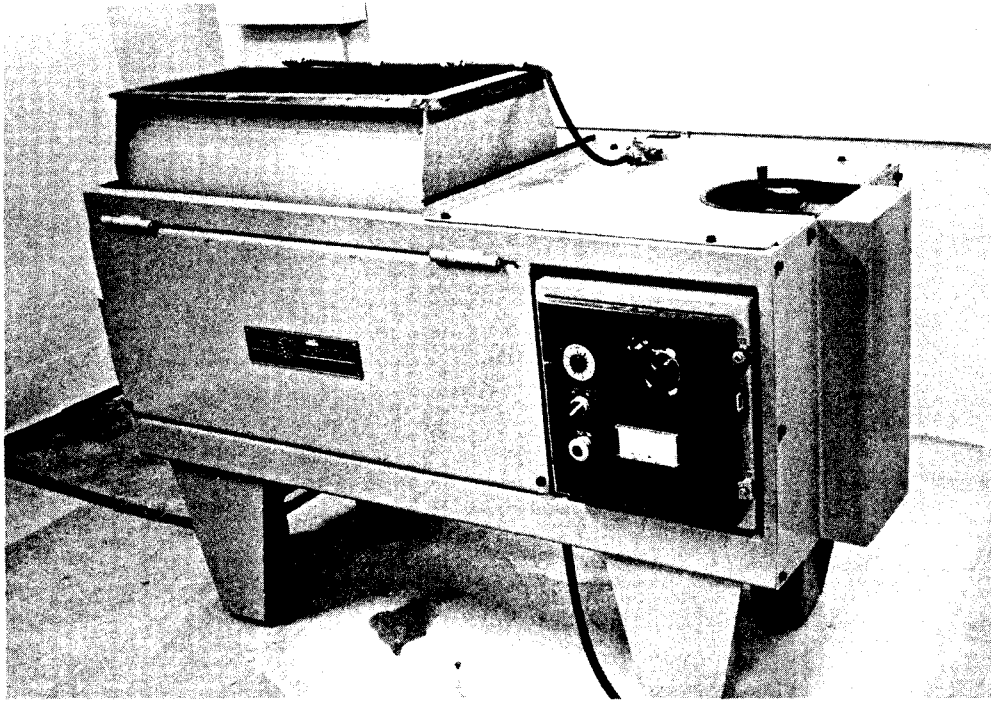
PRETREATMENT STUDIES

A special pretreatment technique is under evaluation for use in the decontamination program. Mass finishing, commonly called vibratory finishing, has been used for improving surface finishes, for removing burrs, sharp edges, and heat treat scale, and now for removing radioactive contamination from metallic surfaces. Vibratory finishing techniques to remove radioactive contamination from metallic surfaces rely on the motion between the part to be cleaned and the media of abrasive material. The variables in vibratory finishing are:

vibratory equipment, media, compounds, flushing solution, and time cycles.

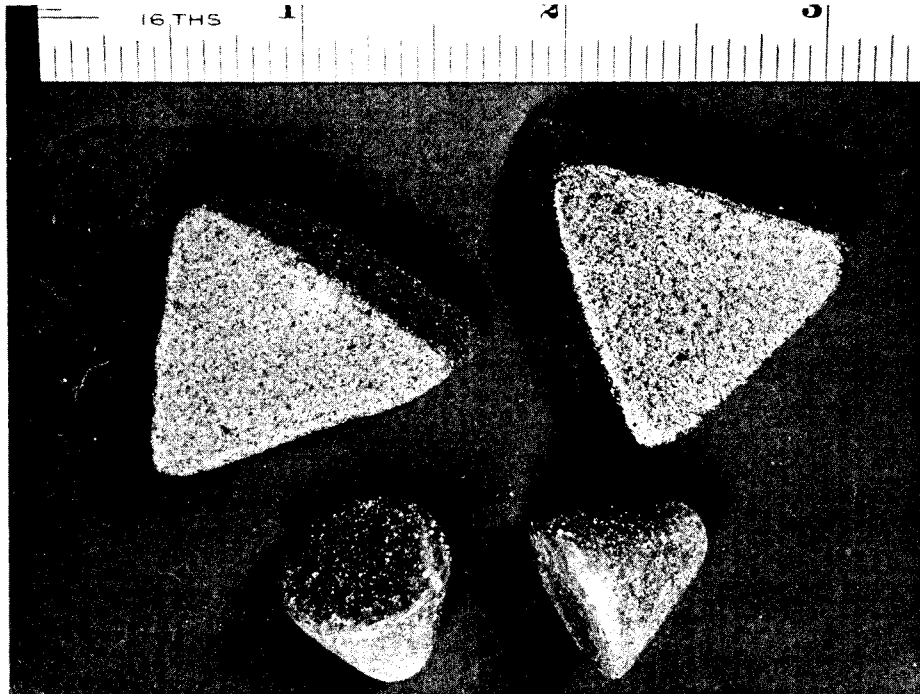
A vibratory finisher with a 113-dm³ (4 ft³) tub capacity has been obtained for the pretreatment facility to remove paint, grease, corrosion layers and gross contamination from components before electropolishing (Figure 12.2). Tests are also under way to determine the effectiveness of vibratory finishing in removing dross and the recast layer from plasma arc torch cut material. Testing is being conducted in a ventilated greenhouse, 3.66 by 3.66 by 2.44 m (12 by 12 by 8 ft), to prevent the possible spread of radioactive contamination.

A variety of noncontaminated metal specimens were tested in the vibratory finisher, which contained a conical-type plastic media (Figure 12.3). Operating at a machine speed



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FIGURE 12.2. Vibratory Finisher for Pretreatment Studies



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FIGURE 12.3. Conical-Shaped Plastic Media (Below) and Triangular-Shaped Ceramic Media (Above) for Use in Vibratory Finisher

(vibrational frequency) of 1300 rpm and using a chemical cleaning compound, the test unit successfully removed rust and even epoxy-type paint from irregularly shaped objects [pipe support bracket, 5-cm (2 in.) OD pipe, 2.5-cm (1 in.) hexagonal nut] in less than 10 hr processing time. Tests with rectangular stainless steel samples of varying length-to-width ratios showed that although the sharp corners and edges are rounded by the abrasion process, the actual amount of metal removed is very small (less than 0.5 wt% for 31 hr of processing). The wear rate for the plastic media was only 0.6 wt%/hr.

Other pretreatment tests were conducted using beta/gamma-contaminated carbon steel components from N-Reactor and a medium-cutting-rate, triangular-shaped ceramic abrasive with 2.86-cm (1.1 in.) sides (Figure 12.3). The vibratory tub containing the media and the parts was continuously flushed with 15.4 ℓ /hr (4 gal/hr) of filtered, recirculated water.

The initial test load consisted of 16 contaminated carbon steel pipe clamps from N-Reactor with a total weight of 28 kg (62 lb). These were processed in the vibratory finisher containing 57 dm³ (2 ft³) of the ceramic media. The initial radiation level of the clamps averaged about 100 mR/hr, with most of the contamination associated with a heavy rust layer. Four hours of vibratory finishing were sufficient to remove this corrosion layer and leave a clean, bright surface ready for electropolishing. In addition, this pretreat-

ment reduced the average radiation level to about 1 mR/hr, with higher readings in a few areas that were inaccessible to this size media.

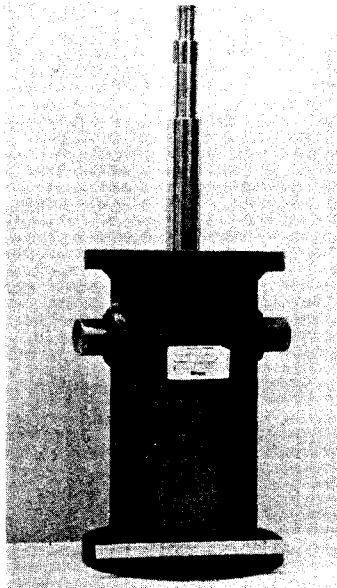
The second test consisted of 150 contaminated carbon steel rings, 7.6-cm OD by 5.1-cm ID by 1-cm thick (3-in. OD by 2-in. ID by 0.4-in. thick), processed in the same 57 dm³ (2 ft³) of ceramic media. The rings weighed a total of 30 kg (66 lb) and had an initial radiation reading of 3 to 6 mR/hr. This was reduced to less than 0.5 mR/hr by 4 hr of processing. As before, most of the remaining contamination was in small, inaccessible grooves. This is not a problem, however, as media can be obtained in a variety of sizes and configurations. Also, it should be noted that the amount of metal processed in these tests was only about one-half of the normal working capacity of the 113-dm³ (4 ft³) vibratory finisher.

These tests show that vibratory finishing can successfully remove gross beta/gamma contamination as well as rust and other surface material to produce surfaces that are clean and ready for final decontamination by electropolishing. Furthermore, even after processing more than 14 dm³ (0.5 ft³) of contaminated, rusted metal parts in 57 dm³ (2 ft³) of media, the media and tub walls were found to be essentially uncontaminated. The same abrasive action that removes the contamination also keeps these surfaces clean. The contamination, rust, paint, spent abrasive, etc., are continuously washed out of the vibratory finisher into a waste container where they can be collected

and concentrated for disposal. Thus, in addition to preparing surfaces for final decontamination by electropolishing, the use of vibratory finishing as a pretreatment technique will help extend solution life by minimizing contamination and other surface residue introduced into the electropolishing bath.

RELATED PROJECTS

A carbon steel pneumatic cylinder and piston assembly, 30.5-cm (12 in.) diameter by 61-cm (24 in.) high, (Figure 12.4) used to actuate a steam valve at the Hanford N-Reactor was decontaminated on an emergency basis for United Nuclear Industries, Inc. This permitted the rebuilding of this critical component at a noncontaminated machine shop and averted a delay in reactor startup.

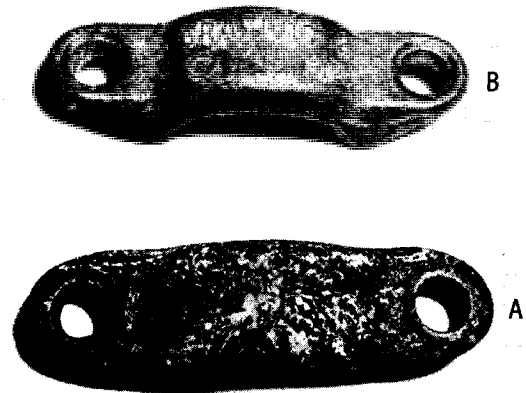


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FIGURE 12.4. Decontaminated Carbon Steel Pneumatic Cylinder and Piston Assembly

Mild steel clamps, many of which were covered with rust and corrosion, were also received for decontamination from N-Reactor. Two approaches were tried. Some of the clamps were pretreated by overnight immersion in a rust-removal solution, given 2 to 3 hr of ultrasonic treatment, and then electropolished. Because of the heavy rust layer, electropolishing of many of the clamps had to be repeated, which required high currents for long periods.

The second approach involved placing the clamps in the vibratory finisher, using a ceramic media and a rust-inhibitor compound. Initial radiation levels of the clamps averaged about 100 mR/hr, with most of the contamination tied up in the heavy rust layer. After 4 hr of processing in the vibratory finisher, the clamps were bright, with all the rust and paint removed (Figure 12.5). In addition, radiation had been reduced to about 1 mR/hr. In fact, the clamps had been decontaminated to the extent that electropolishing was not required.



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FIGURE 12.5. Mild Steel Clamps (A) Before and (B) After Decontamination in Vibratory Finisher

Electropolishing was successfully used to decontaminate a plutonium-contaminated, carbon steel vessel for the Rockwell Hanford Operations Company. Electropolishing rapidly reduced the surface contamination level from 1,000,000 dis/min/100 cm² to background for most of the component. Some areas, however, had readings in excess of 10,000 dis/min/100 cm² even

after 60 min of electropolishing. This was traced to incomplete paint removal and illustrates the need for effective pretreatment procedures to completely depaint and degrease the metal surfaces before electropolishing. Decontamination was completed using mechanical depainting and spot electropolishing procedures.



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