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**Partitioning of Long-Lived
Nuclides from
Radioactive Waste
—FY 1975 Annual Report**

April 1976

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and Development Administration
under Contract E(45-1):1830**

Battelle

Pacific Northwest Laboratories

BNWL-1926

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PARTITIONING OF LONG-LIVED NUCLIDES FROM
RADIOACTIVE WASTE--FY 1975 ANNUAL REPORT

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SUMMARY

This report summarizes the progress achieved in the waste partitioning program during fiscal year 1975. The work presented is not viewed to be complete, but only intermediate in examining two interdependent questions:

1. Are there incentives for partitioning high-level waste?
2. Is partitioning technically feasible?

To obtain answers to the first question, the incentives for separating and eliminating various elements (but particularly the transuranics) from radioactive waste prior to final geologic storage were investigated. The assumed radionuclide source was all of the high-level U.S. nuclear power economy waste through the Year 2000 (including all tritium, carbon, and iodine from spent fuel and activation products from the cladding). Emplacement of the waste in a non-salt geologic formation surrounded by western U.S. desert subsoil was assumed.

The study required evaluation of numerous parameters concerning the transport of radioactivity from the geologic storage repository to man. These parameters are associated with five major evaluation factors: release pathway between the repository and the biosphere, nuclide retention by the geologic medium, transport agent and its rate, pathways in the biosphere, and consequences to man. The transport assumptions included the following:

- The release pathway of greatest risk from underground disposal is dissolution of the waste and transport to the surface by flowing groundwater.
- Measurements and estimates of the nuclide retention properties of western U.S. desert subsoils were assumed to be applicable for the geologic medium and valid for the chemical species actually present.
- A groundwater velocity of 1 ft/day was assumed. This is ten times greater than that of typical western U.S. desert aquifers.
- Radiation doses were calculated for a "maximum" individual.

Available data were used whenever possible, but many of the study parameters had to be estimated. The values used were either consistent with

current knowledge or were selected to maximize the calculated potential radiation doses. Thus incentives for removing various elements from the waste were greatly increased. Incentives were also greatly overestimated by neglecting all short-term risks and by assuming that elements removed from the waste could be eliminated from the earth without risk.

The conclusion that the incentives for removal of any elements, including the transuranics, from high-level waste are vanishingly small--despite assumptions, many of which tended to maximize them--is based on comparison of predicted potential radiation doses with routine doses from natural sources. The study found that there are feasible conditions of geologic emplacement where the predicted incremental dose to man is calculated to be at least as low as one-tenth of "background." Although "background" varies from 100 to 250 mrem/year for locations in the United States (120 mrem/year was assumed for this study), and consensus standards of allowable incremental dose have not been fully established, we concluded that high-level waste in a glass form can safely be placed in selected geologic media.

The feasibility and methods for reducing the uncertainties in this study should be carefully assessed, and a more general evaluation of partitioning incentives should be made. We recommend that additional work include: 1) the extension of the present migration model to other geologic media, such as faulted monoliths and bedded salt formations, 2) additional laboratory and in situ measurements of the retention of nuclides by various geologic media, 3) estimation of the long-term stability of migrating species, 4) examination of the pathways to man from all steps in the waste management scheme, 5) all wastes from the nuclear fuel cycle, 6) an improved biosphere transport model which includes the long-term accumulation and dispersion of nuclides in the biosphere, and 7) an evaluation of the future value of the waste as a resource.

Only a limited effort was expended in answering the question "Is partitioning technically feasible?" A variety of separation processes with the potential for removing the actinide elements from the fission product waste were examined, and some laboratory testing was performed. A conceptual flowsheet has been devised, but additional

laboratory and hot cell testing is required for verification and optimization. The effect of the actinides associated with the solids found in dissolver solution and in other process streams is a particularly important question requiring resolution. One sample of irradiated oxide fuel has been dissolved in a "typical" nitric acid dissolution, and the residual solids removed and assayed. In this experiment, 0.32 wt% of the oxide fuel was not dissolved and 0.71% of the total amount of plutonium in the fuel remained in the insoluble solids.

INTRODUCTION

The primary objective of this investigation was to determine the effect upon waste management concepts of removing biologically significant long-lived radionuclides (principally but not exclusively actinides) from commercial radioactive wastes and thereby demonstrating the incentives or lack of incentives for partitioning. A second objective was to develop the necessary technology for performing the partitioning processes and thus both determine the technical feasibility and establish a technological basis for pilot-plant or engineering-scale development of the processes.

The second objective provides a contingency in the event that clearly defined incentives for partitioning are determined or that resolution of the primary objective cannot be made.

The radionuclides in the waste have widely different half-lives and biological toxicities. For example, most of the fission products decay to tolerable levels in about 100 years; cesium and strontium decay to low levels in about 1,000 years; but iodine-129, technetium-99, carbon-14, and many of the actinide nuclides persist for hundred of thousands of years. If biologically significant long-lived radionuclides (those having half-lives exceeding 30 to 50 years) are removed from nuclear wastes, the radioactive toxicity will become essentially unimportant in less than 1,000 years. If they are not removed, the wastes can remain significantly radioactively toxic for hundreds of thousands to millions of years.

Incentives for partitioning exist only if both of two conditions are satisfied:

1. The transport rate of the long-lived nuclides from the repository causes an unacceptable risk to man.
2. Acceptable risks result from the extra processing and the ultimate disposal of the long-lived fraction.

The partitioning program was initiated in 1973 with a preliminary overview assessment of the technical feasibility of segregation of high-level

waste constituents. The preliminary study concluded that technology exists to perform separations in the range likely to be needed for the advanced waste management options under study, but that R&D is required to adapt this technology to the specific needs of waste segregation and to develop reliable estimates of costs and benefits as a function of separations duty and separation methods.

The preliminary overview study, which defined R&D needs and proposed an R&D program, was completed in FY 1973 and summarized in BNWL-1776,⁽¹⁾ issued in November 1973. This study was intentionally broad, and input was sought from many people expert in nuclear fuel reprocessing, separations technology, and analytical development. More than 25 people outside PNL participated in the study. They included individual consultants, university people, people from other ERDA sites, and people from industry.

An R&D program at PNL was proposed for FY 1974 to investigate waste segregation, including waste characterization, solids removal and treatment, and development of analytical capability pertinent to partitioning of commercial and ERDA high-level reprocessing wastes. This was later revised to emphasize a study of the feasibility of applying the partitioning concept to Hanford Purex process acid waste to provide early information specific to Hanford waste and was reported in BNWL-1907.⁽²⁾

The four-task program initiated in FY 1975 involved, primarily, experimental development studies which resumed and accelerated the previous study to resolve the several major problems in partitioning identified in the feasibility study.⁽¹⁾ The program was expanded to include consideration of non-high-level wastes and partitioning of solidified waste (e.g., a calcine) such as might be shipped to a central facility for conversion to a storage or disposal form.

Because of the pressing need to answer the question "Should the high-level waste be partitioned?" two new tasks (5 and 6) were added at the start of the Second Quarter of FY 1975, and priorities in funding and available manpower resources were given them. Task 5 was to compare the long-term safety benefits to man from disposed partitioned waste fractions

with the benefits of disposed unpartitioned waste. The objective of Task 6 was the comparison of waste management systems with and without partitioning.

TASK DISCUSSION

TASK 1 - CHARACTERIZATION AND TREATMENT OF SOLIDS IN HIGH-LEVEL WASTE

Task Objective

Finely divided solids are known to be present in dissolver solution and in other high-activity process streams of fuel reprocessing and waste treatment facilities. These solids include cladding fines, residues from the dissolution of the fuel, and precipitates formed during reprocessing and waste treatment. If small fractions of the plutonium, americium, curium or other actinides present in the irradiated fuel are chemically or physically bound in these solids in unreactive form, they pose a serious problem to the development of a partitioning process; they limit the degree of removal of these long-lived elements from the shorter-lived fission products by any process which relies on the actinides being in ionic or soluble molecular form.

Neither the concentrations of the actinides present in such solids nor the characteristics of the solids have been thoroughly investigated. Both questions must be answered before the technical feasibility of partitioning can be realistically evaluated or the requirements defined for any solids treatment process used in conjunction with a partitioning process. Corrective action might include revision of the initial fuel dissolution flowsheet, secondary treatment of dissolver solution or high-activity acid waste, or separation of the solids followed by separate treatment or disposal.

The objective of this task is to evaluate the effects of the solids on partitioning by determining the amount of plutonium, americium, and curium associated with the solids at various points in the reprocessing of reactor fuels and the treatment of the wastes, and by measuring the volume, particle size, and other characteristics of the solids needed in the development of processes for their treatment and handling. Progress toward the objective was restricted to a single experiment in which a

small quantity of highly irradiated light-water reactor fuel was dissolved and the solid residue analyzed to determine the plutonium, americium, and curium content.

Experimental Work

A small quantity of Yankee reactor fuel from an element known to have been exposed to an average burnup of 39,000 MWd/MT was selected for the experiment. This fuel was considered sufficiently representative of typical LWR fuel to exemplify the dissolution behavior of the plutonium, americium and curium. A part of a fuel rod from this element was cut into sections suitable for dissolution.

The dissolution of the fuel was carried out in a small glass dissolver fitted with an air-cooled condenser, a dip tube for air sparging, and a thermometer well. The outlet of the condenser was connected to a trap and a scrubber containing 2 M NaOH to prevent potential loss of fission-product activity to the hot-cell exhaust system. The dissolver is shown schematically in Figure 1.

FIGURE 1. Schematic Drawing of Oxide-Fuel Dissolution Apparatus

A quantity of fuel pieces totaling 112 g, including cladding, was placed in the dissolver which contained 250 ml of 5.5 M HNO_3 , heated to 90°C, and held at that temperature 4 hr.

Dissolution of UO_2 in HNO_3 proceeds through the reactions:⁽³⁾

and

Both reactions occur, but the first predominates at the acid concentration used here. Dissolution conditions were selected to give a final concentration of 4 to 4.5 M HNO_3 and 1.3 M uranium.

After completion of the dissolution step, the solution was cooled and then filtered through a Teflon[®] filter with a 5 μ pore size. The solids and the cladding hulls were then returned to the dissolver and leached for 4 hr with 250 ml 8 M HNO_3 , again filtered, and the filter exhaustively washed with 2.0 M HNO_3 . The leach solution, washes and initial dissolver solution were combined, diluted to 1.00 ℓ and sampled for chemical analyses. The cladding hulls and the solids were then weighed separately.

The next phase of the experiment was intended to convert the plutonium, americium and curium in the solids to a soluble form which could be analyzed. This was done by first leaching with a solution of HNO_3 -HF followed by fusion of the residue in NaHSO_4 . The HNO_3 -HF leach consisted of heating a weighed portion of the solids in 50 ml 8 M HNO_3 -0.05 M HF for 12 hr at 65°C. The residue was filtered and added to 15 g NaHSO_4 . The NaHSO_4 was heated to give a fluid melt, held at temperature for 1.5 hr, cooled, and dissolved in 2.0 M HNO_3 . The dissolved NaHSO_4 was combined with the HNO_3 -HF leachate, diluted to 250.0 ml, and sampled for analyses.

Results and Conclusions

The total weight of the filtered solids obtained from nitric acid dissolution of the fuel was 290 mg and represented 0.32 wt% of the uranium

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metal present in the irradiated fuel. This is in apparent agreement with results obtained at Oak Ridge National Laboratory (ORNL) with a stainless steel clad mixed-oxide fuel. Under similar dissolution conditions the ORNL workers observed 0.4 to 0.5 wt% solids for fuel with exposures of 25,000 to 50,000 MWd/T.⁽⁴⁾ No attempt was made to identify the constituents of the solids other than the Pu, Am and Cm, but other workers have reported that the principal elements present are Mo, Ru, Pd, Rh, Zr, Tc and Sb.^(4,5)

The analyses of the clarified dissolver solution and the combined solution obtained from the HNO₃-HF leach and the NaHSO₄ fusion are summarized in Table 1. The principal alpha activity found in the "dissolved" solids is plutonium, but the minor amounts of americium and curium indicate a slight tendency for these elements to adhere to the solids.

TABLE 1. Analyses of Clear Dissolver Solution and Solubilized Solids

	Content of Dissolver Solution		Content of Solubilized Solids	
	Amount	% of Total in Fuel	Amount	% of Total in Fuel
Pu	1.00 g	99.3	0.0072 g	0.71
²⁴¹ Am	4.06x10 ¹¹ dis/min	99.9	3.5x10 ⁸ dis/min	0.086
²⁴⁴ Cm	3.28x10 ¹¹ dis/min	99.99	4.19x10 ⁷ dis/min	0.013

Based on the fractions of plutonium, americium and curium found in the nitric acid insoluble residues, it is apparent that treatment or elimination of the solids is required if large decontamination factors are to be obtained in a partitioning process. The actinides in the solids, if left in the aqueous high-level wastes, cannot be extracted by conventional processes and will limit the decontamination factors to 1.7,* 1100 and 8000 for Pu, Am and Cm respectively.

* Assuming 0.5% loss of soluble Pu occurs in the Purex process.

TASK 2 - REMOVAL OF LONG-LIVED NUCLIDES FROM SOLIDIFIED WASTE

Task Objective

The objective of this task is to develop a flowsheet for partitioning starting with a solidified high-level waste, e.g., a calcine, such as might be shipped to a central facility for conversion to a storage or disposal form. The chief aim at the start of the project was to demonstrate a suitable process for getting calcine into a solution that would permit subsequent separation of long-lived nuclides from the waste. Achieving dissolution of the calcine into a nitric acid system appeared to have several advantages, an obvious one being that the resulting solution could probably be used as feed in partitioning flowsheets being developed for other nitric acid waste streams. Thus new technology requirements for processing the solution after dissolution of the calcine could be kept at a minimum.

Experimental Work

The work for this task was divided into three main areas:

1. Laboratory tests with simulated calcine to develop a dissolution process.
2. Demonstration of the calcine dissolution process with actual calcined high-level waste in a hot cell.
3. Hot-cell demonstration of actinide removal from the feed solution resulting from 2.

The FY-1975 effort was limited to experimental work in Area 1.

Simulated calcines were prepared by pot calcination of a low-sodium synthetic high-level waste prepared with the following chemical substitutions: Fe for Ru, Co for Rh, Ni for Pd, Mo for Tc, K for Rb, rare earths for Y + Am + Cm, and U for Pu + Np. This synthetic high-level waste contains solids; i.e., not all of the constituents completely dissolve during preparation of the waste "solution." In the preparation of this synthetic

waste, molybdenum is added as the tri-oxide, MoO_3 . The light color of the undissolved solids in the synthetic waste indicated that this compound was not completely dissolved even after agitation of the solution at elevated temperature for 24 hr.

Two simulated calcines were prepared: the first by direct calcination of the synthetic waste including undissolved solids, and the second by calcination of filtered synthetic waste solution. Both types were prepared by evaporating synthetic high-level waste to dryness in a stainless steel beaker and then calcining the solids in the stainless beaker at 900°C in a crucible furnace. The cooled calcine was removed from the breaker, pulverized, and blended to a homogeneous mixture.

Acid dissolution, using HNO_3 or HNO_3 -HF, and molten salt fusion were used separately and in combination to dissolve the simulated calcines. The acid dissolution tests were performed in covered glass and plastic (for HNO_3 -HF solution) beakers utilizing Teflon[®]-covered magnetic bar for stirring. The molten salt fusions were conducted in nickel crucibles and the melts dissolved in dilute HNO_3 .

Initial calcine dissolution tests were performed with simulated calcine prepared from unfiltered synthetic waste solution. The results from acid dissolution tests, presented in Table 2, show that digestion with HNO_3 -HF is superior to digestion with HNO_3 alone, but is not adequate for complete dissolution of the calcine.

Table 3 shows results from dissolution tests on the same calcine using molten salt fusions. Caustic-peroxide fusion was the best of the three dissolution methods investigated, yielding 80 to 90% dissolution of the calcine upon contacting the cooled melt with HNO_3 .

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TABLE 2. Dissolution of Simulated Calcine by Acid Digestion

<u>Acid Test No.</u>	<u>Acid</u>	<u>ml Acid/ g Calcine</u>	<u>Temp.</u>	<u>Time, hr</u>	<u>Wt.% Calcine Dissolved</u>
1	15.7 <u>M</u> HNO ₃	10	Boiling	4	28
2	Con HCl	10	Boiling	4	75
3	15.7 <u>M</u> HNO ₃ -0.3 <u>M</u> HF	10	80°C	5	57
4	15.7 <u>M</u> HNO ₃ -0.03 <u>M</u> HF	10	80°C	24	60
5	15.7 <u>M</u> HNO ₃ -0.03 <u>M</u> HF	30	95°C	5	70
6	15.7 <u>M</u> HNO ₃	5	Boiling	2	
	followed by				
	4 <u>M</u> HNO ₃	30	Boiling	1	35
7	15.7 <u>M</u> HNO ₃ -0.003 <u>M</u> HF	5	Boiling	2	
	followed by				
	4 <u>M</u> HNO ₃ -0.01 <u>M</u> HF	35	Boiling	1	55
8	4 <u>M</u> HNO ₃ -0.03 <u>M</u> HF	25	100°C	1	53

TABLE 3. Dissolution of Simulated Calcine by Molten Salt Fusion

<u>Fusion Test No.</u>	<u>Fusion Mixture</u>	<u>Temp., °C</u>	<u>Wt. Ratio Salt/Calcine</u>	<u>Time, hr</u>	<u>Wt.% Calcine Dissolved</u>
1	Sodium hydroxide-sodium peroxide	600	10	2	77
2	Sodium hydroxide-sodium peroxide	600-650	4	2	90*
3	Sodium hydroxide-sodium peroxide	600-650	4	2	87
4	Alkali carbonate	600	10	2	33
5	Sodium-potassium nitrate	600	8	1-1/4	40

* In this test the cooled melt was dissolved in 4 M HNO₃ containing 0.006 M HF.

Although neither the acid digestion tests nor the molten salt fusions resulted in complete dissolution of this calcine, a combination of the two treatments in series resulted in greater overall dissolution. For example, when a caustic-peroxide fusion was performed on the residual solids from acid test No. 8 (Table 2), an overall dissolution of 98% was achieved; when a nitric-hydrofluoric acid digestion was performed on the residual solids from fusion test No. 3 (Table 3), an overall dissolution of 99% was achieved.

The simulated calcine used in the above tests was prepared from unfiltered synthetic waste solution. It was surmised that part of the difficulty in dissolving the simulated calcine may result from refractory molybdenum trioxide or other compounds used to prepare the synthetic waste. That is, the partially refractory nature of the simulated calcine may not be produced by the calcination process, but may be present with some of the starting materials for the synthetic waste.

For this reason, some simulated calcine was prepared from filtered synthetic waste solution. Nitric-hydrofluoric acid digestion tests on this calcine were more successful than those on calcine prepared with unfiltered synthetic waste, as shown by the data in Table 4. In fact, complete dissolution of the simulated calcine was accomplished by using a two-step acid digestion procedure. In this two-step procedure, the calcine was first digested at 100-105°C in 8 M nitric acid for 6 hr. During this treatment 66% of the calcine dissolved. Digestion of the residual solids from this treatment in 8 M HNO_3 -0.15 M HF at 100°C for less than 4 hr resulted in complete dissolution of the solids (see acid tests 20 and 20b in Table 4).

As shown by the data in Table 4, attempts to dissolve the calcine using either a HNO_3 -HF digestion only or a HNO_3 -HF digestion followed by a HNO_3 digestion were not successful in complete dissolution of the calcine. The results indicate that in the HNO_3 -HF treatment, both the HNO_3 and the HF concentrations are important. Eight molar HNO_3 performed better than either concentrated HNO_3 or 4 M HNO_3 when in combination with HF, and 0.15 M HF gave more complete dissolution than did 0.04 M HF in 8 M HNO_3 .

TABLE 4. Dissolution of Simulated Calcine by Acid Digestion

<u>Acid Test No.</u>	<u>Acid</u>	<u>ml Acid/ g Calcine</u>	<u>Temp. °C</u>	<u>Time, hr</u>	<u>Wt.% Calcine Dissolved</u>
9	15.7 <u>M</u> HNO_3 -0.007 <u>M</u> HF	40	80	6	90
10	15.7 <u>M</u> HNO_3 -0.045 <u>M</u> HF	40	80	6	92
11	15.7 <u>M</u> HNO_3 -0.15 <u>M</u> HF	40	80	6	93
12	15.7 <u>M</u> HNO_3	30	80	6	43
13	8 <u>M</u> HNO_3	30	80	6	45
14	4 <u>M</u> HNO_3	30	80	6	42
15	15 <u>M</u> HNO_3 -0.15 <u>M</u> HF	40	100-105	2	87
	followed by				
	4 <u>M</u> HNO_3	30	100-105	3	90-95
16	4 <u>M</u> HNO_3	30	100-105	2	41
	followed by				
	15 <u>M</u> HNO_3 -0.15 <u>M</u> HF	40	100-105	3	90-92
17	8 <u>M</u> HNO_3 -0.15 <u>M</u> HF	40	100-105	3	97
18	8 <u>M</u> HNO_3 -0.15 <u>M</u> HF	40	100-105	3	92
19	4 <u>M</u> HNO_3 -0.15 <u>M</u> HF	40	100-105	3	90
20	8 <u>M</u> HNO_3	20	100-105	6	66
	followed by				
	(a) 8 <u>M</u> HNO_3 -0.04 <u>M</u> HF	40	100	4	98
	(b) 8 <u>M</u> HNO_3 -0.15 <u>M</u> HF	40	100	<4	100

Conclusions

Dissolution tests conducted with simulated calcine demonstrated that complete dissolution of calcine for subsequent separation of long-lived nuclides from the waste is difficult. Complete dissolution of simulated calcine was demonstrated using a two-step acid digestion procedure. The calcine was first digested at 100°C in 8M nitric acid for 6 hr, and then the dissolution was completed by digestion of the remaining solid material in 8M HNO_3 -0.15M HF at 100°C for 4 hr. Attempts to dissolve the calcine using HNO_3 -HF only or HNO_3 -HF followed by HNO_3 were not successful. Hot cell tests using actual calcined high-level waste were planned but not carried out.

TASK 3 - REMOVAL OF LONG-LIVED NUCLIDES FROM NON-HIGH-LEVEL WASTES

Task Objective

The objectives are to evaluate the applicability of the partitioning concept to non-high-level nuclear wastes and to develop the technology necessary to apply partitioning to these wastes, where desirable, to integrate them into the overall advanced waste management program.

Discussion

Significant amounts of long-lived radionuclides are present in nuclear wastes other than the high-level waste produced in fuel reprocessing. Representative of such wastes are residues from volume reduction of oxidizable wastes; fuel hulls; lower-radioactivity-level liquid wastes from fuel fabrication, fuel reprocessing, and reactor decontamination operations; spent isotopic power sources and wastes involved in decommissioning commercial waste storage sites. From the standpoint of partitioning, consideration of each of these is needed to develop comprehensive advanced waste management programs.

Because of the low priority assigned to Task 3, no reportable work was performed during FY 1975.

TASK 4 - DEVELOPMENT OF LOW-ADDITIVE FLOWSHEETS FOR REMOVING LONG-LIVED NUCLIDES FROM THE FISSION PRODUCTS

Task Objective

The main objective of Task 4 is the conception, development, and demonstration of an integrated flowsheet by which the long-lived nuclides (chiefly, but not exclusively, actinides) can be separated from the short-lived fission products during or after fuel reprocessing, without addition of excessive amounts of inert materials harmful to subsequent waste processing or disposal. Initially, the major concern was the development of methods for the separation of the americium and curium from the fission products, especially the chemically similar lanthanides, since this was considered the most difficult problem to be solved. Prior studies of this separation have emphasized processes [both ion exchange^(6,7) and solvent extraction⁽⁷⁾] which use complexants (e.g., DTPA) to facilitate separation. While there is little doubt that such processes can be made to work, they do involve addition of potentially troublesome additives to the waste (not only the complexant itself but also the cation of the base, usually NaOH or NH_4OH , used to adjust the pH of the complexant solution).

The current work on this task was aimed at finding processes for separating americium and curium from the lanthanides without addition of substitute potentially troublesome additives. Separation of americium and curium from many (but not all) of the lanthanides might be possible using solvent extraction operations involving nitric acid-salted aqueous phases and phosphonate or phosphinate extractants, but definitive experiments were not completed during this reporting period. One advantage these extractants have over some others (e.g., acid organophosphorus compounds) is the ability to extract americium and curium from strongly acid solutions such as the high-level waste stream of the Purex process. A hoped-for but as yet not achieved objective of this study is to find two suitable americium/curium extractants having substantially different separation factors for lanthanides. Separation from all the lanthanides could then be accomplished in two solvent extraction cycles using different solvents to provide separation from different groups of lanthanides. Much testing of possible extractants is required to evaluate this concept.

Investigation of Potential Flowsheets

Results of a literature survey⁽⁸⁻¹²⁾ of the extractability of americium and the lanthanides from nitric acid solution are summarized by the data shown in Figures 2 and 3. Americium distribution coefficients with dibutyl butylphosphonate (DBBP) and one datum point with europium were measured at PNL to supplement the literature data.

FIGURE 2. A Comparison of the Distribution Coefficients of Selected Lanthanide and Actinide Ions with Potential Extractants

FIGURE 3. Distribution Coefficients of Selected Lanthanide and Actinide Ions with DBBP

The data in Figure 2 illustrate a wide variation of extractability of americium with various extractants. They also show significant differences in the relative extractabilities of americium and cerium. The bidentate compound, dibutyl-N, N-diethyl carbamylmethylene phosphonate (DBDECMP), is seen to extract americium very strongly but to give relatively little separation from cerium. The phosphinate, butyl dibutylphosphinate (BDBP), is seen to extract americium quite well, but nothing is known of the separation from cerium. The phosphate, tributyl phosphate (TBP), gives appreciable separation from cerium but does not extract americium sufficiently well except at high HNO_3 concentrations. The phosphonate, dibutyl butylphosphonate (DBBP), extracts americium sufficiently well for a purification process at both low and high HNO_3 concentrations and also gives appreciable separation from cerium. Figure 3 gives additional data on the separation of americium from some other lanthanide elements by DBBP extraction from HNO_3 solutions.

If phosphinates show differences in americium and lanthanide extractabilities comparable to those observed with phosphates and phosphonates, the higher extractant power of the phosphinates would likely make them the most desirable as partitioning agents. Phosphinates are not commercially available, and efforts to obtain samples for testing were not quite complete at the end of the reporting period.

With both TBP and DBBP, the extractability of the lanthanides increases with increasing atomic number. The extensive body of data available for the TBP system^(10,11,13) shows that americium extraction is quite close to that of neodymium, and curium extraction is quite close to that of promethium; thus no separation of neodymium plus promethium from americium plus curium would be achieved with this system. The data for the DBBP system (Figure 3) are less extensive, but the separation factors appear quite comparable. Separation of americium and curium from the next adjacent lanthanides (praseodymium and samarium) could be appreciable, and separation from the remaining lanthanides should be quite extensive by either TBP or DBBP extractants.

The experimental portion of this effort centered around DBBP because of its availability and its apparent superiority as an americium extractant over TBP, the other readily available extractant. Americium distribution coefficients were measured with both 100% and 58% DBBP; these data are shown in Figures 2 and 3. To aid in flowsheet development, HNO_3 distribution coefficients were also measured; these data are compiled in Table 5. Finally, a few tests were completed in a miniature centrifugal contactor.

TABLE 5. Extraction of HNO_3 by DBBP

<u>Aqueous Phase</u> <u>HNO_3, M</u>	<u>HNO_3 Distribution Coefficient</u>	
	<u>58% DBBP-NPH</u>	<u>100% DBBP</u>
1.3	0.37	
3.0	0.48	
4.4	0.40	
5.5		0.56
6.8	0.31	
8.5		0.45
8.7	0.26	
10.7	0.25	
12.4		0.36
15.7		0.43

The initial centrifugal contactor tests were aimed at maximizing americium recovery and did not include any fission products. Two six-stage contactors were connected in series to provide eight extraction and four scrub stages. The details of contactor design and operation are presented elsewhere.⁽¹⁴⁾ The conditions employed and results obtained in the two runs are presented in Table 6.

TABLE 6. Centrifugal Contactor Testing of Am Extraction

Extractant: 60% DBBP-NPH

Temperature: 40-45°C

<u>Stream</u>	<u>Stage Introduced</u>	<u>Flow, ml/min</u>	<u>HNO₃, M</u>	<u>Am, %</u>
<u>Run 1</u>				
Extractant	12	4.96		
Acid-add	11	0.99	12.0	
Feed	5	1.62	3.0	100
Scrub	1	0.80	3.0	
Organic Product			1.7	85
Aqueous Waste			3.6	30
<u>Run 2</u>				
Extractant	12	4.93		
Acid-add	11	0.38	12.0	
Feed	5	1.65	3.0	100
Scrub	1	0.82	3.0	
Organic Product			1.7	96
Aqueous Waste			1.7	8

From the flow ratios and acid concentrations used in Run 2 and the americium extraction coefficients measured at room temperature (shown in Figure 3), the americium loss in Run 2 should have been less than 1% instead of the 8% measured. Samples taken during Run 2 from the channels connecting some of the stages showed the effective americium extraction coefficients to be about 75% as great as those observed in the batch contacts at room temperature. Since the variation was about the same in the scrub section as in the extraction section, it is likely that this variation was caused by a decrease in the extraction coefficient with increasing temperature rather than by kinetic effects. Using the lower extraction coefficients to calculate the expected loss gives a value of about 5% instead of the 8% measured. This remaining discrepancy was probably the

result of less-than-optimum contactor performance; some of the organic phase samples taken from the connecting channels also contained rather large amounts of aqueous phase. Such behavior certainly reduces the number of theoretical stages achievable.

The americium concentration in the feed for these runs was about 10^{-4} g/l, or about 1000 times lower than that expected in commercial high-level waste solutions. The results of these experiments thus suggest that americium recoveries as high as 99.99% should be attainable from the high-level waste solutions without encountering kinetic problems which could limit the recovery.

The results of this study do suggest there is a good probability that americium and curium can be separated from the bulk of the fission products (including many of the lanthanides) by solvent extraction processes which do not involve the addition of additives (other than HNO_3) to the waste. Whether or not such a process is of significant value depends on factors such as: a) the method chosen for ultimate disposal of the actinide fraction, b) the form chosen for ultimate disposal of the fission product fraction, and c) the process chosen to produce that form.

If the method chosen for ultimate disposal of the actinide fraction does not require that the actinide fraction be completely free of lanthanides, then a process such as this could be of significant value. For example, if the actinide fraction is to be disposed by launching into space, the fact that neodymium and promethium had not been removed might be of little consequence because no radioactive neodymium isotopes would be present in significant amounts and relatively little shielding would be required for the promethium.

If the method chosen for ultimate disposal of the actinide fraction does require that the actinide fraction be free of lanthanides, then one of the separations processes employing complexants would have to be used (based on current knowledge) even though such processes do require adding significant amounts of inert additives. In this instance, the nitric acid-salted extraction process described above serves mainly to prepare a cleaner

feed for a process which will give the required actinide/lanthanide separation, thus lowering the amount of additives required. The value of this reduction depends on the tolerance for the additives by both the waste form chosen for ultimate disposal of the fission product fraction and by the process chosen to reach the ultimate disposal form. For example, a borosilicate glass as the form for ultimate disposal of the fission product fraction can tolerate substantial amounts of sodium (and other additives). The process used to reach the final waste form, however, may not tolerate as much sodium as will the waste form itself; for example, in conversion of liquid waste to solid form by calcination, fluidized bed calcination tolerates sodium much more poorly than does spray or pot calcination.

Other approaches investigated for providing a cleaner feed for an additive-type process which will give high actinide/lanthanide separation include: 1) extraction of zirconium and molybdenum away from americium, curium, and lanthanides with di (2-ethylhexyl) phosphoric acid (HDEHP) from 6 M HNO_3 ,⁽⁷⁾ and 2) extraction of americium, curium, and lanthanides away from other fission products with bidentate organophosphorus compounds from 5-6 M HNO_3 .⁽¹⁵⁾ Neither process gives significant separation of americium and curium from the lanthanides. Thus they do not allow as extensive a decrease in additives as could be obtained with the DBBP process studied here.

Proposed Conceptual Flowsheet

The conceptual flowsheet in Figure 4 is a preliminary effort at integrating a waste partitioning process with the conventional TBP (Purex) process for recovery of purified U, Pu, and Np. The flowsheet features back-cycle of waste streams (including solvent wash solutions) so that all of the waste requiring partitioning is contained in only one stream. The process does add inert chemicals (e.g., sodium and zinc) to the waste stream. But they are in amounts within the composition ranges of some promising glass formulations being studied for immobilizing high-level waste.

FIGURE 4. Conceptual Partitioning Flowsheet

The uranium, plutonium, and neptunium recovery and purification schemes featuring the back-cycle of waste streams are based on those used successfully at the Hanford Purex Plant⁽¹⁶⁾ with relatively low burnup fuel. The higher plutonium and neptunium contents to be encountered in power reactor fuels may necessitate some modifications, but no major perturbation to the concept is anticipated. One such modification included in Figure 4 is the use of nitrogen dioxide instead of sodium nitrite solution for destruction of the holding reductant in the TBP stream to minimize the amount of sodium introduced into the waste. Another modification likely required in processing power reactor fuels would be to use something other than ferrous sulfamate as a plutonium partitioning agent; promising candidates include hydroxylamine nitrate⁽¹⁴⁾ and electrolytic reduction⁽¹⁷⁾ as well as the more commonly considered uranium (IV).

The concept of mixing the spent solvent wash solution with the high-level waste solution so that both of these actinide-bearing waste streams can be partitioned at once has not been demonstrated but is worthy of study. In the conceptual process, the spent solvent wash solutions are acidified and washed with normal paraffin hydrocarbon (NPH) diluent to remove dibutyl phosphate (DBP) so that it will not be recycled back to the main process. The NPH wash solution, which also will contain TBP and will be contaminated with actinides, is burned in an incinerator. The acidified and washed solutions are then combined with the high-level waste solution (HAW), and the combined streams are steam-stripped to remove TBP and NPH prior to digestion in order to convert inextractable forms of plutonium to extractable forms. The digested feed is then contacted with 30% TBP to reduce the residual uranium, plutonium, and neptunium concentrations to the very low levels required for partitioned waste; the 30% TBP extract is routed back to the Purex process.

The waste stream should then require removal of only americium and curium to achieve the actinide partitioning objectives. In the conceptual process, americium and curium are removed from the waste stream by solvent extraction; two cycles are shown in Figure 4 to assure that the required removal (99.9%) can be obtained. The solvent to be used in these americium/

curium extraction cycles is one which will allow the americium/curium fraction to be separated from many of the lanthanides (as well as the other fission products) by extraction, scrubbing, and stripping operations under conditions of varying nitric acid concentrations. This solvent is shown as 60% DBBP-NPH in Figure 4; as discussed earlier, it is likely that a better choice could be made after additional development.

In the PA columns of the conceptual process the americium, curium, and highly extractable fission products are separated from less-well extracted materials by extraction and scrubbing operations employing relatively low HNO_3 concentrations ($\sim 2 \text{ M}$). In the PB columns the americium, curium, and similarly extractable fission products are separated from the more highly extractable materials by stripping into moderately concentrated HNO_3 solution ($\sim 8 \text{ M}$) and scrubbing with fresh solvent. In the PC columns the most highly extractable fission products are stripped out of the solvent phase into dilute HNO_3 . The solvent is washed as required to remove degradation products, and the wash solutions are combined with those from the Purex (TBP) portion of the process.

The fission products contained in the aqueous phases leaving the 3PA and 3PC columns would be sufficiently partitioned from all of the actinides and ready for waste solidification. However, some fission products would be present with the americium and curium in the aqueous phases leaving the 2PB and 3PB columns. With a DBBP extractant there would be essentially no separation of neodymium or promethium and only limited separation of praseodymium or samarium from the americium and curium. To separate the americium and curium from these fission products, a cation-exchange process employing complexants to enhance the separation factors is proposed.

In the cation-exchange process the americium, curium, and residual lanthanides would be loaded onto the resin from dilute ($\sim 0.5 \text{ M}$) HNO_3 and then separated by elution down a sequence of smaller sized resin beds using zinc as a barrier ion and DTPA as the complexant. Details of such a process are presented elsewhere.⁽⁶⁾ The lanthanide-containing solutions would be treated (if necessary) to destroy the complexant and mixed with the other fission product solution for waste solidification. The degraded resin

would be incinerated along with the diluent used to remove the DBP; the residue from this incinerator is the only actinide waste stream of the entire conceptual process.

Evaluation of the Conceptual Flowsheet

The process as outlined in Figure 4 is considered capable of producing a fission product fraction containing little enough sodium for incorporation in a suitable waste-glass composition. Based on the procedures used at the Hanford Purex plant,⁽¹⁶⁾ the Purex process solvent wash solutions would introduce about 100 moles of sodium per metric ton of uranium (MTU), and the sodium nitrite solution added to the HA column for Np valence control would introduce about 5 mole Na/MTU. Adding similar amounts for the similar operations in the partitioning process gives a total of about 210 mole Na/MTU for the solvent extraction portions of the process.

The cation-exchange process described⁽⁶⁾ for recovery of curium from the waste of the Barnwell Nuclear Fuel Plant required use of about 500 mole DTPA/MTU. A process feed purified from fission products except for neodymium and promethium plus small fractions of the adjacent lanthanides is estimated to require only about 100 mole DTPA/MTU. If four moles of NaOH were required for adjusting the pH of the DTPA, the cation-exchange process would introduce about 400 mole Na/MTU, giving an overall total of about 600 mole Na/MTU for the process outlined in Figure 4.

The 600 mole Na/MTU estimated for the outlined process is very near the value of 677 mole Na/MTU calculated for waste from fuel containing 35 kg of fission product oxide per MTU (typical of LWR fuel irradiated to 33,000 MWd/MTU) incorporated into a glass containing 15 wt% Na₂O and 25 wt% fission product oxide, which are appropriate amounts for some glass forms.⁽¹⁷⁾ The amount of sodium introduced in the outlined process could likely be reduced significantly by using strong nitric acid instead of DTPA solution to strip the cation-exchange column and by running the sodium carbonate solution used to wash the solvent to more nearly complete neutralization before it is sent to waste.

Conclusions

Although the conceptual flowsheet is based upon the limited amount of laboratory and literature work completed in FY 1975, the flowsheet does satisfy the main objective of Task 4 if the term "low-additive" is interpreted to mean additives that are acceptable to subsequent waste form processing. The flowsheet is not optimized, comprehensive, nor tested in the laboratory, and such work would be required prior to adoption of the flowsheet.

TASK 5 - EVALUATION OF INCENTIVES FOR PARTITIONING

Task Objective

The purpose of this task is to develop a general method for estimating the incentives for partitioning the radioactive wastes generated by the nuclear power industry and then to apply that method to all the high-level nuclear waste generated through the Year 2000 (including all tritium, carbon, and iodine from spent fuel and activation products from the cladding). The following discussion summarizes an earlier in-depth topical report.⁽¹⁹⁾

Discussion

The general evaluation method defined two conditions which must be simultaneously satisfied in order to establish the existence of partitioning incentives for a given waste type. They are that the risk from unpartitioned waste must be both greater than the publicly acceptable risk and greater than the sum of the risks from all of the partitioned fractions. The publicly acceptable risk is defined as that risk value which is marginally acceptable to the public at large. The risk for a given unpartitioned waste or partitioned waste fraction is the sum over all release pathways of the release probability for each pathway multiplied by the release consequences for that pathway.

The general method can be simplified by considering only that waste management activity which has the greatest risk. This study assumes that the release pathways of greatest risk for partitioned and unpartitioned

waste originate from the waste in its final storage setting. Thus the risks from prestorage processing and transportation are assumed to be insignificant relative to the risk from the stored waste.

Final storage in continental geologic media was assumed rather than seabed or ice sheet disposal. All nuclides partitioned from the waste are assumed to have been completely eliminated by extraterrestrial disposal or transmuted to other nuclides, with negligible risk in either case. This latter assumption clearly increases the likelihood that partitioning incentives will exist and thus tends to bias the analysis in favor of partitioning. The probability of release from the storage site is given the value "1" so that the analysis considers only release consequences.

With foregoing assumptions, the existence conditions for partitioning incentives can be expressed as:

$$C > C_a \quad (1)$$

$$C > C_\ell \quad (2)$$

where: C = consequences from unpartitioned high-level waste released by the leach incident pathway

C_a = publicly acceptable release consequences

C_ℓ = consequences from the low-risk fraction of the partitioned high-level waste.

The consequences of a release of radionuclides from a final storage repository were evaluated using an integrated geosphere-biosphere transport model. The theory of both individual models and the details of the computing scheme for the biosphere model have been reported.⁽²⁰⁻²³⁾ The results from the consequence calculations are reported as 50-year accumulated doses to "maximum" individuals living during various geologic times within the region of influence of the underground terminal storage site. The diet and living habits of these "maximum" individuals are chosen such as to maximize the doses potentially received.

A sensitivity analysis was performed on the important variables over which man has some degree of selection or control when designing a management system for nuclear wastes. The results of these parametric variations are shown in Figures 5, 6, and 7.

FIGURE 5. The Effect of Path Length on the Bone Dose Versus Time Curve

FIGURE 6. The Effect of Leach Rate on the Bone Dose
 Versus Time Curve

FIGURE 7. The Effect of the Time of Initial Release from the Disposal Site on the 50-Year Accumulated Dose

Figure 5 shows the effect of path length on the incremental bone dose versus time curve. Results are shown for 0, 0.1, 1, and 10-mile (16 km) path lengths. The background and NCRP dose limit lines are superimposed for comparison. For a zero path length (a disposal site located in the surface water body itself), all nuclides discharge simultaneously with a bone dose of 600,000 times background, controlled by ^{90}Sr . For a path length of 0.1 miles (0.16 km), incremental doses are at or above background over most of the 100,000-year period required for complete discharge of all nuclides to the biosphere. The largest dose results from ^{90}Sr at 250 years (17,000 times background). At a 1-mile path length the bone doses are above background over large time periods, but the incremental doses are never greater than the ^{14}C dose (50 times background) which occurs between 250 and 500 years after Year 2000. For the 10-mile path length, only the bone dose from ^{14}C (50 times background) at 1500 years and the bone dose from ^{237}Np (2 times background) at 14,600 years are above background. Thus as the distance between the terminal storage site and the biosphere increases, the nuclides discharge to the surface water body over longer time periods and the incremental doses decrease. In particular, the highest incremental dose level was reduced by a factor of 12,000 (600,000/50) by increasing the path length from 0 miles to 10 miles. These results confirm that isolation of the waste from the biosphere can greatly reduce consequences of waste released from containment at the terminal storage site.

Figure 6 shows the effect of leach rate on the incremental bone dose versus time curve. The leach-rate values shown are 0.3, 0.03, 0.003, and 0.0003%/year. As before, the background and the NCRP radiation limit lines are superimposed for comparison. The 0.3%/year curve in Figure 6 (the base case) is exactly the same case as the 10-mile curve in Figure 5. When the leach rate is decreased an order of magnitude to 0.03%/year, the ^{14}C and ^{237}Np bone doses are decreased an order of magnitude. The ^{14}C dose is about five times background. However, the ^{14}C now discharges to the biosphere over a 3333-year period (1/0.0003) instead of the 333-year period for the base case. Thus, although the incremental dose to a "maximum"

individual is reduced by a factor of 10, there are presumably 10 times as many such "maximum" individuals receiving that incremental dose compared with the base case. For the 0.003%/year leach rate, the incremental doses are always below background, and the ^{14}C dose is spread out over a 33,333-year period. The results for the 0.0003%/year leach rate show further redistribution of the doses so that more individuals get lower doses. However, because of radioactive decay the total integrated doses over all time decrease as the leach rate decreases.

An interesting and important phenomenon can be illustrated by examining the bone dose results for the uranium peak at 2.07×10^6 years. As the leach rate is decreased from 0.3 to 0.003%/year, no change is observed in the incremental dose, which is controlled by ^{226}Ra . This happens because the ^{226}Ra is created predominately by decay of the precursors ^{234}U and ^{238}Pu after they have left the final storage site. Because of the differences in the migration rates of radium, uranium, and plutonium, the ^{226}Ra discharge to the biosphere is spread over large time periods and is thus unaffected by leach rate until the leach rate is 0.0003%/year or less (i.e., until the leach time begins to approach the migration time for uranium). Thus, decreasing the leachability of the waste form greatly reduces the consequences of single nuclide releases from the site but does not reduce the consequences from chain members created after their precursors left the disposal site unless the leach time approaches the migration time of those nuclides through the geosphere.

Figure 7 shows the effect of the time of initial release after Year 2000 on the controlling doses to critical organs. These results show that increased canister integrity and disposal site stability cannot reduce the incremental doses below background unless containment at the disposal site can be reasonably assured for periods of greater than 1,000,000 years. Although the ^{14}C and ^{99}Tc doses disappear if the release can be delayed by 100,000 years and 1,000,000 years, respectively, the ^{129}I and ^{237}Np doses remain at above background levels. However, the results shown in Figure 7 assume a 10-mile (16 km) path length. As the path length decreases, the importance of canister integrity and site stability increases.

The combined effects of the three waste management control variables shown in Figures 5, 6, and 7 on the incentives for partitioning can best be shown with three-dimensional plots such as Figure 8. The three variables (path length, leach time, and the time of initial release after Year 2000) are plotted on the three axes. Thus an increase in the magnitude of any of the parameters results in greater isolation of the waste from man's environment. A surface can be located in this three-dimensional space which corresponds to any assumed incremental dose level. Figure 8 shows such a surface with background as the assumed incremental dose level. Combinations of path length, leach time, and time of initial release which define points located inside or under the surface (i.e., between the surface and the origin of axes) have incremental doses above background; combinations located outside the control surface have incremental doses below background.

In plots of these waste management control surfaces, the incremental dose need be greater than the assumed criterion (e.g., background) at only one time during the entire time the waste discharges to the biosphere to cause that point to be inside the surface.

The topographic features of the waste management control surface can be conveniently named according to which nuclides cause them. The ^{90}Sr spire occurs at small path lengths and small times of initial release and reflects the large leach times (small leach rates) required to lower the controlling ^{90}Sr dose to background. The ^{226}Ra ridge occurs at intermediate path lengths and initial release times and denotes the region of the surface where the dose is controlled by the ^{226}Ra created predominately by decay of ^{238}Pu and ^{234}U after they have been released from the disposal site. The ^{14}C ledge and ^{129}I plateau are named in similar fashion.

Figure 8 (or similar graphs) can be used together with the two existence conditions (inequalities 1 and 2) to determine the partitioning incentives for the separation of various elements from the waste. As discussed previously, two things must be known: 1) the publicly acceptable release consequences and 2) the waste management control variables for the final storage system. Thus the question of "how safe is safe enough?" must

FIGURE 8. Waste Management Control Surface for Incremental Background Dose with No Partitioning

be answered and the terminal storage site, container, and waste form must be selected. Using any value (arbitrary or otherwise) for the publicly acceptable release consequences, a graph similar to Figure 8 is drawn. Then using the given values of the control variables, the point characteristic of the chosen waste management system is located on the graph. If that point is above the control surface, the release consequences are below the publicly acceptable consequences, inequality 1 is not satisfied, and no partitioning incentive exists. If that point is below the control surface, inequality 1 is satisfied, and an incentive to partition something from the waste exists if inequality 2 is also satisfied. What must be separated from the waste and eliminated depends upon where the system characterization point for the chosen system is located under the surface. Therefore, the objective of the partitioning alternative is to separate selected elements from the waste until inequality 1 is no longer satisfied.

This method can be illustrated by assuming that incremental doses of the order of natural background are the publicly acceptable consequences and the waste management system characterization point is located at a path length of 1 mile (1.6 km), a leach time of 333 years, and an initial release time of 0 years after Year 2000. Since this hypothetical point is below the surface in Figure 8, inequality 1 is satisfied and incentives for partitioning will exist for separation and elimination of nuclides which lower the release consequences (satisfy inequality 2).

If 99% (100 DF) of the plutonium and uranium (^{238}Pu and ^{234}U) are removed from the waste at the Year 2000, the ^{226}Ra ridge on the waste management control surface disappears as shown in Figure 9. The ^{14}C ledge now becomes much wider in the path length-leach time plane. As shown in Figure 9, elimination of 99% of the plutonium and uranium moves the control surface down toward the characterization point. Thus incentives for partitioning of plutonium and uranium exist for the hypothetical situation.

FIGURE 9. Waste Management Control Surface for Incremental Background Dose with 99% Elimination of Uranium, Plutonium, Carbon, Technetium, Iodine and Neptunium

Figure 9 also shows the effect of sequentially removing and eliminating 99% of the carbon (^{14}C), technetium (^{99}Tc), iodine (^{129}I), and neptunium (^{237}Np), after the uranium and plutonium have previously been removed. It should be noted that the carbon and iodine are currently reprocessor off-gas products and hence would not be included in high-level waste by current definitions. However, the significant doses from both carbon and iodine suggest that they might be candidates for isolation from reprocessor off-gas streams.

The 99% removal of carbon and technetium significantly reduce the height of the control surface above the characterization point, and hence partitioning incentives exist for these elements. Iodine removal changes the surface at all path lengths up to 100 miles for initial release times of 1,000,000 years and greater but causes no change in the control surface at small initial release times because neptunium is present at the same dose level. However, iodine removal makes possible the reduction in the height of the control surface when neptunium is subsequently removed. After having eliminated uranium, plutonium, carbon, technetium, iodine, and neptunium, the characterization point is above the control surface for the assumed publicly acceptable incremental dose, and incentives for additional partitioning do not exist for this hypothetical situation. Now, ^{126}Sn controls the incremental dose for path lengths greater than 0.1 mile and initial release times up to 100,000 years.

The previous discussion considered the partitioning incentives for a hypothetical waste management system deliberately chosen to maximize those incentives. Consideration now is given to waste management systems chosen to make the partitioning incentives marginal. If the final storage site is separated from the biosphere by at least 5 miles of western U. S. desert soil or equivalent, the waste form has a leach time of at least 15,000 years (1 ft diam x 10 ft long monolithic borosilicate glass waste form with a surface flux of slightly less than 10^{-5} g/cm²/day), the waste containment fails at the Year 2000, and the publicly acceptable consequence to an individual is an incremental background dose or higher, the system

characterization point would be located on Figure 8 about at the intersection of the carbon ledge and the radium ridge. Thus no partitioning incentives would exist for this scenario.

Likewise, if the publicly acceptable incremental dose is one-tenth of background (which has the effect of raising the control surface shown in Figure 8 by an order of magnitude in the region where the carbon ledge and radium ridge intersect), the migration path consists of at least 5 miles of western U. S. desert subsoil or equivalent, the leach time is at least 150,000 years (surface flux of slightly less than 10^{-6} g/cm²/day), and no credit is taken for the integrity of the container or the stability of the site from water penetration, no partitioning incentives exist.

These scenarios seem quite reasonable. It seems very likely that a terminal storage site in a nonsalt formation can be found in the United States where the path from the disposal site to the biosphere is 5 miles or more of western U. S. desert soil or equivalent. Leach rates as low as 10^{-7} g/cm²/day have been measured for sample borosilicate glass waste forms, so that the assumed range of 10^{-5} to 10^{-6} g/cm²/day seems conservative. Much evidence exists to show that current containers can last 100 years or more and that almost any reasonable site could be expected to be stable from water penetration for 1000 years or more. Thus taking no credit for canister integrity or the water penetration stability of the site seems very conservative. The assumption that the publicly acceptable incremental consequences are one tenth of background also seems conservative. Thus, if the assumptions made in developing and applying the method for this study are valid, incentives for partitioning high-level waste do not exist.

The results of this study are applicable to the evaluation of incentives for conversion of high-level calcine into glass. Calcine is very soluble in water and has a large surface to volume ratio, so that the leach time might be less than 1 year and would probably not be greater than 100 years. Thus calcine waste forms would be below the waste management control surface in Figure 8 for an incremental background dose (and much below a similar surface for a one-tenth of background incremental dose) even if

the release does not occur for 10,000,000 years. Thus, if the leach incident is the highest risk pathway to man from high-level waste, there is an incentive to convert calcine into glass.

Conclusions

In simplest form, the results obtained from the work in Task 5 can be summarized in three statements:

1. If an incremental background dose (6000 mrem for a 50-year accumulated dose) is the publicly acceptable release consequence, 5 miles or more of western U. S. desert subsoil separate the final storage site from the biosphere, the leach time is 15,000 years or more (monolithic waste form with leach rate of slightly less than 10^{-5} g/cm²/day), and the initial release occurs in the Year 2000 or later, there is no incentive to partition anything from high-level waste.
2. If one-tenth of background is the publicly acceptable release consequence, the path length is 5 miles or more, the leach time is 150,000 years (monolithic waste form with a leach rate of slightly less than 10^{-6} g/cm²/day), and the initial release occurs in the Year 2000 or later, there are no partitioning incentives for high-level waste.
3. If the leach incident is the highest risk pathway to man from the high-level waste, there is an incentive to convert high-level calcine into glass.

TASK 6 - COMPARISON OF WASTE MANAGEMENT SYSTEMS WITH AND WITHOUT PARTITIONING

Task Objective

The objective of this task is to characterize and compare waste management systems with and without partitioning in order to assess the economic and safety benefits and liabilities of the partitioning operations. The primary thrust has been to identify and evaluate the options for waste form, packaging, and disposal that are introduced by partitioning. The evaluation

is dependent upon results from Tasks 1, 4, and 5. Factors such as practical limits of transmutation or space disposal, the impact of actinide removal on packaging, potential degradation, and disposal of the long-lived fraction require evaluation.

Work was not initiated on Task 6 during the reporting period. Input must be available from Tasks 1, 4, and 5 before significant results can be achieved in Task 6.

PROPOSED FUTURE WORK

1. The role of solids in relation to partitioning, plutonium accountability, and to waste management in general remains, for the most part, poorly defined. The results from examination of the actinide content of the residual solids from dissolution of one single batch of irradiated oxide fuel clearly demonstrate that dissolver solution solids contain a significant amount of plutonium and other transuranic elements. It is vital to examine additional batches of irradiated fuel in order to evaluate the composition of the solids (including the actinide content) and other properties of interest to reprocessing and waste management, as a function of fuel fabrication method and irradiation history.
2. A general evaluation of partitioning incentives should be made which removes or justifies the assumptions made for this study and which includes an evaluation of the waste as a mineral resource. To accomplish this task, the general study must consider:
 - all wastes from the nuclear fuel cycle
 - all steps in the waste management scheme for each waste both with and without partitioning
 - all pathways to man from each step.
3. In support of Recommendation 2, further work should:
 - complete development of the nuclide migration model for a particulate medium with salt present and extend the present model to describe the faulted monolith type of geologic formation
 - measure in the laboratory and in situ the sorption equilibrium constants for nuclides in the various geologic media with and without the presence of salt and develop a better fundamental understanding of actinide migration phenomena

- improve the biosphere transport model to include the long-term (>100 years) accumulation and dispersion of nuclides in the biosphere.
4. Because the present analysis shows incentives for partitioning are highly unlikely, further work developing specific partitioning processes should not be pursued at the expense of the first three items.

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