

REMOVAL OF ACTINIDES FROM HIGH-LEVEL WASTES GENERATED
IN THE REPROCESSING OF COMMERCIAL FUELS *

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REMOVAL OF ACTINIDES FROM HIGH-LEVEL WASTES GENERATED IN THE
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ABSTRACT

Progress is reported on a technical feasibility study of removing the very long-lived actinides (uranium, neptunium, plutonium, americium, and curium) from high-level wastes generated in the commercial reprocessing of spent nuclear fuels. The study was directed primarily at wastes from the reprocessing of light water reactor (LWR) fuels and specifically to developing satisfactory methods for reducing the actinide content of these wastes to values that would make 1000-year-decayed waste comparable in radiological toxicity to natural uranium ore deposits. Although studies are not complete, results thus far indicate the most promising concept for actinide removal includes both improved recovery of actinides in conventional fuel reprocessing and secondary processing of the high-level wastes. Secondary processing will be necessary for the removal of americium and curium and perhaps some residual plutonium. Laboratory-scale studies of separations methods that appear most promising are reported and conceptual flowsheets are discussed.

The studies comprising this investigation have not yet progressed to the point where it is possible to determine the feasibility (or infeasibility) of chemical ^{process} (flowsheets) for the satisfactory removal of actinides from reprocessing wastes. On the basis of tracer-level studies with synthetic wastes and conceptual material-balance flowsheet studies, the chemistry required for the actinide separations (with the possible exception of plutonium) appears to be theoretically possible. However, a vast amount of work remains to be done

in order to select the best combination of chemical steps and to prove that the desired chemistry can be carried out in workable, integrated processes under high levels of radiation. Our work strongly suggests that recycle methods might be developed for Purex plants which would substantially reduce the quantities of the miscellaneous low- and intermediate-level wastes. The actinide-bearing wastes currently require relatively large-area sites for disposal or storage. Such wastes appear to be amenable to recycle if appropriate chemical reagents or methods are used for treating used solvent and for adjusting the neptunium and plutonium valences in the Purex separation.

INTRODUCTION

Several waste management alternatives presently under study in the United States by the Energy Research and Development Administration (ERDA) depend on treatment of high-level waste to separate (or partition) it into a fission-product fraction containing acceptably low concentrations of long-lived radionuclides and a semipure, concentrated fraction containing the long-lived nuclides [1]. This concept is illustrated in Fig. 1. The fraction depleted in long-lived radionuclides might be disposed of in geologic formations reliably predicted to be stable for a few thousand years. The long-lived fraction might possibly be eliminated by transmutation [2] or disposed of in a manner that takes into consideration the relatively low heat generation rate but very long half-lives of the constituent radionuclides.

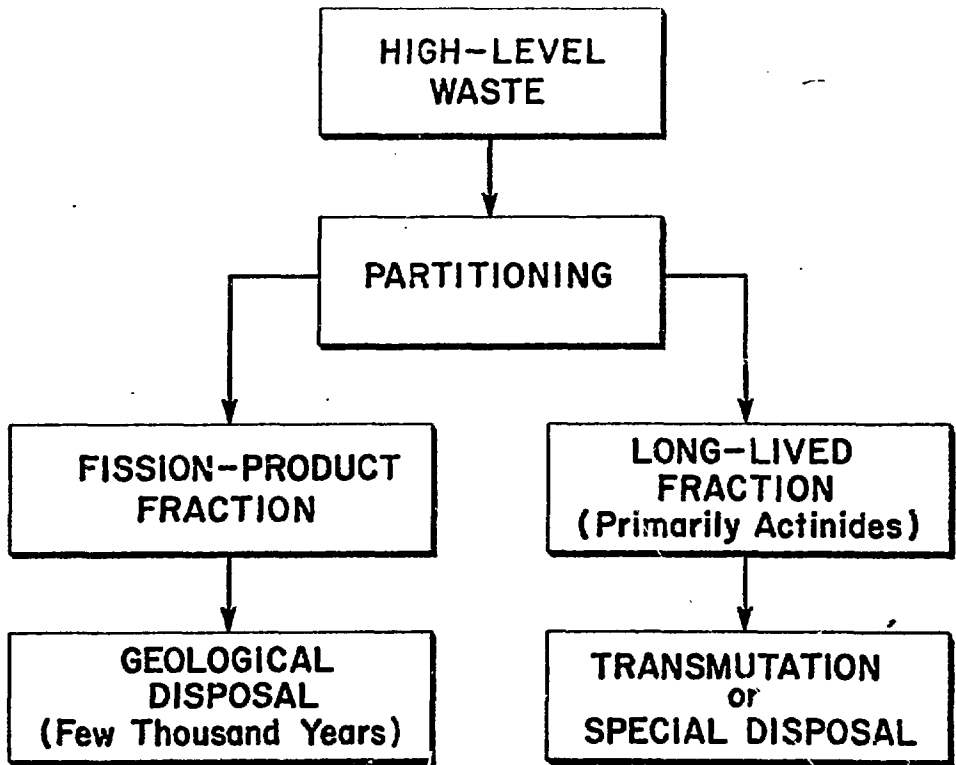


Fig 1. ^{PARTITIONING} ~~FRACTIONATION~~ CONCEPT OF WASTE ~~FRACTIONATION~~ AND MANAGEMENT OF THE WASTES

The present study was limited to the high-level waste from the reprocessing of spent light water reactor (LWR) fuel because this is the only reactor fuel expected to be processed commercially during the next decade. Spent LWR fuels contain several very-long-lived radionuclides — both actinides and fission products — which are discharged to the high-level waste in conventional reprocessing. The actinides are the major contributors to the potential long-term hazard, representing greater than 99% of that for 1000-year-decayed waste. The actinides of concern are uranium, neptunium, plutonium, americium, and curium. The long-lived fission products of concern are ^{99}Tc , ^{93}Zr - ^{93}Nb , ^{135}Cs , and ^{129}I .

Studies of actinide removal reported here are based on reducing the radiological toxicity contribution of actinides in 1000-year-decayed waste to a value that is comparable to the toxicity of the long-lived fission products. It was assumed that 99.9% of ^{129}I , tritium, and noble gases were removed in reprocessing and managed as separate wastes, and that the high-level waste contained only 0.1% of these elements. When actinides and ^{129}I are removed from spent fuels to an extent where the actinide contribution to the hazard potential of high-level waste is about equal to the contribution of the long-lived fission products, the hazard potential of the waste after 1000 year decay is comparable to that of uranium minerals [3-6]. All

comparisons of radiological toxicity (or hazard potentials) in this study are made on the basis of hazard index values. The hazard index is defined as the volume of water required to dilute a unit volume of a radiological mixture to levels for unrestricted water use as defined in the United States Code of Federal Regulations (10 CFR, Part 20).

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In Table 1, the percentages of actinides that are removed in conventional reprocessing of LWR fuels by the Purex process are compared with those percentages which would be required ~~in~~ to reduce the hazard index of 1000-year-decayed, high-level waste to about 5% of the hazard index value of the mineral, pitchblende. The comparison given in Table 1 was made on the basis of LWR fuel with plutonium recycle and having a burnup of 33,000 MWD/metric ton. Additional removal factors from 5 to 1000 would be required and are dependent on the specific actinide element. These removal factors are considerably beyond those normally achieved in high-level radiation, chemical separations facilities. Americium and curium are not recoverable by the Purex process and will require a secondary process for their removal. Recoveries of Am-Cm from specially irradiated targets have ranged from 90-99% in transplutonium element production programs.

Removal of actinides in partitioning to the extent shown in Table 1 appears to be a desirable goal to attain in waste partitioning feasibility evaluations before consideration of further actinide removal. This is because the removal factors required are 5 to 50 times greater than those that have been reported for actinides in processing facilities ^{the existing} at radiation levels. The partitioning goals, if achieved, would reduce the hazard index of 1000-year-old high-level waste by factors of about 60 and about 400 for the wastes from LWRs with no recycle of Pu and with Pu recycle, respectively. Further significant reductions in hazard index would require removal of long-lived fission products (primarily ^{as well as} ^{99}Tc and ^{amounts of the} additional actinides. At the removal percentages shown for partitioning in Table 1, ^{99}Tc and the residual plutonium and americium contribute about equally to the hazard of high-level waste from LWR fuel in which plutonium is recycled.

Table 1. Comparison of Actinide Recoveries in the Conventional Reprocessing of Spent LWR Fuel by the Purex Process with the Recoveries Required to Reduce the Hazard Index of 1000-Year-Decayed, High-Level Waste to About 5% of the Hazard Index of Pitchblende.^a

Actinide Element	% Recovery		Additional Recovery Needed
	Conventional Goal	Partitioning Goal	
U	99.5	99.9	5
Np	(90-95) ^b	95.0	0-2
Pu	99.5	99.99	50
Am-Cm	0	99.9	1000

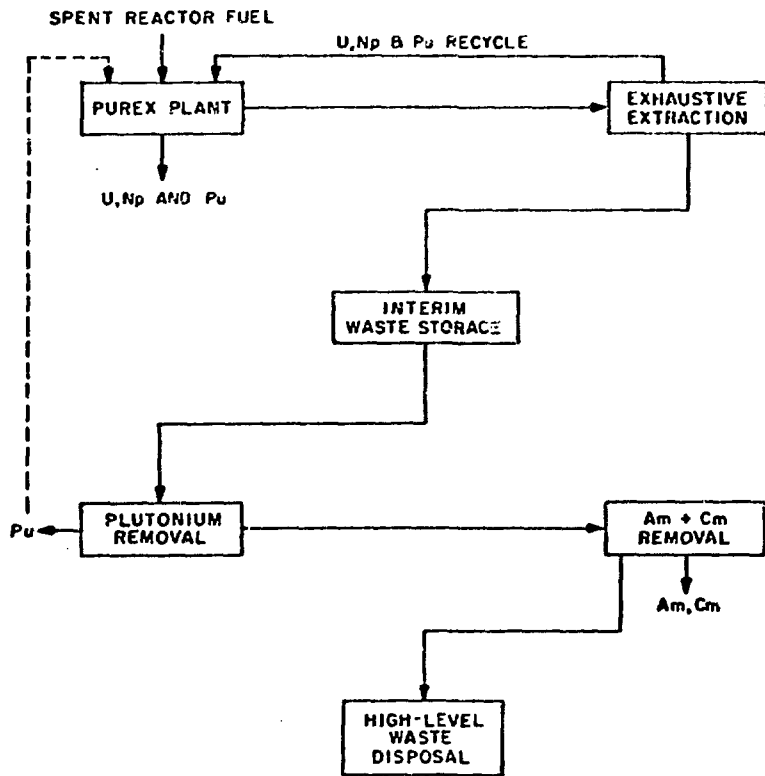
^aBasis of comparison: (1) LWR fuel of the pressurized water reactor type with plutonium recycle (PWR-Pu) having a burnup of 33,000 MWd/metric ton, (2) assumed the high-level waste contained 0.1% of the iodine, tritium, and noble gases and (3) assumed a pitchblende ore containing 70% U.

^bNeptunium is not usually recovered in the reprocessing of power reactor fuels. Recoveries of 90-95% have been made on a special campaign basis.

METHODS FOR REMOVAL OF ACTINIDES

The concept of actinide removal investigated was based on a combination of modified Purex processing and secondary processing of the high-level waste. According to this concept (Fig. 2), the necessary removal of uranium and neptunium and most of the plutonium is accomplished by extra extraction stages in the Purex process and by recycle of the aqueous wastes from the actinide purification cycles. Secondary processing of the high-level waste is used to remove americium, curium, and any residual plutonium. Insoluble residues from fuel dissolution or solids generated in processing would be separated in feed clarification steps and treated, where necessary, to reclaim the actinide values. An important requirement is the capability for removing the actinides without significantly increasing the volumes of high-, low-, and intermediate-level wastes. This requirement prohibits the use of processes which discharge large quantities of chemical reagents in the wastes. If processes that depend on large quantities of reagents are to be used -- and they are probably necessary for removal of americium and curium -- it will be necessary to choose reagents which can be recycled or converted to acceptable chemical wastes that are nonradioactive.

The most troublesome chemical problems encountered in actinide removal are associated with plutonium and americium-curium. The chief difficulty with plutonium lies in the need to maintain it in an extractable form at very low concentration. Scouting experiments showed that about 0.01% of the plutonium was inextractable when a solution of Pu(IV) in nitric acid was repeatedly extracted with tributyl phosphate (TBP). Interferences from fission products



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 Fig. 2. Conceptual processing sequence for the removal of actinides from LWR reactor fuel.

in the waste may increase the fraction of inextractable plutonium. The[#]major difficulty in americium-curium processing is in effecting the removal of these elements without generating radioactive wastes containing large quantities of chemical reagents. In addition to the problem with inextractability of plutonium from waste solutions, sufficient information has not been obtained relative to the plutonium content and nature of the solids generated in the processing of spent fuels. A considerable amount of work still needs to be done to define the scope of the solids problems and to develop methods for preventing plutonium losses to these solids.

Material-balance flowsheet studies show that actinide removal according to our concept — or any concept — will require extensive recycle. Demonstrating that this recycle can be accomplished is a major undertaking. For example, it will require conclusive proof that none of the steps involved will lead to a buildup of impurities which could cause process failure. Unfortunately, there is no previous experience available concerning the operation of a large-scale radiochemical plant which utilizes extensive recycle of the waste streams.

Most of the laboratory and process flowsheet development studies have been concerned with (1) reprocessing modifications for improved recovery of uranium, neptunium, and plutonium, (2) developing a satisfactory process for recovery of americium and curium, and (3) determination of the nature of solids formation in waste solutions. In addition, a basic study of the behavior of plutonium at very low concentrations in aqueous solutions is being carried out.

Reprocessing Modifications

We prepared a conceptual material-balance flowsheet for a modified Purex process which includes the recycle of nitric acids as well as the aqueous, actinide-bearing waste streams generated in the partitioning and purification cycles for uranium, neptunium, and plutonium. In addition, we carried out some experimental studies on the extractability of neptunium from synthetic wastes and its subsequent partitioning.

In our conceptual flowsheet, the uranium, neptunium, and plutonium are recovered in a primary Purex plant in which the uranium and neptunium concentrations are reduced to the required levels by additional stages of Purex extraction. The Purex process has the capability for accomplishing the required uranium and neptunium recovery and removal of perhaps as much as 99.8% of the plutonium. Our conceptual flowsheet includes the recycle of the nitric acid and actinide values from all of the intermediate- and low-level aqueous waste streams, except those wastes from solvent purification and off-gas treatment. ^{method for recycling these wastes must be developed.} Recycle of the aqueous waste streams is based on using a partitioning reductant other than ferrous sulfamate, which would add sulfate to the high-level waste. The sulfate also interferes with recycle of the actinide values from the partitioning and purification cycle wastes. A considerable amount of experimental work remains to be done on the recycle steps of the flowsheet to determine whether they will lead to an accumulation of impurities which could inhibit recovery of actinides. Also, experimental work needs to be done preparatory to developing a conceptual flowsheet for recycling actinides from the solvent purification wastes.

Laboratory batch extraction tests showed that greater than 98% of the neptunium was removed by two stages of TBP extraction when tracer-level neptunium was maintained as Np(VI). Qualitative rate measurements showed that ferrous nitrate reduces Np(VI) to Np(IV) at a satisfactory rate for partitioning plutonium from neptunium and uranium. The required reduction times, 10 to 20 min, appear to be compatible with the holdup time of production-size solvent extraction columns.

Recovery of Americium and Curium

Several processes for the recovery of americium, curium, and transcurium elements from high-level waste are being evaluated in the Laboratory. These evaluations are being made using tracer-level activity in synthetic wastes. All of the processes involve isolating the chemically similar trivalent lanthanide fission products and trivalent actinides (americium, curium, and transcurium elements) from the other waste elements and then separating the trivalent actinides from the lanthanides by cation exchange chromatography [7,8] or the Talspeak solvent extraction process [9,10]. The processes being studied for the separation of the trivalent actinides and lanthanides (trivalent elements) include: (1) cation exchange; (2) oxalate precipitation coupled with cation exchange; and (3) TBP extraction from solutions heavily salted with aluminum or lithium nitrates. Each of these processes has certain advantages and disadvantages. Additional experimental studies are needed to permit selection of the optimum overall process for removing americium and curium from high-level waste. Involved in this selection is the preparation of detailed flowsheets with recycle or satisfactory handling of every waste stream, including waste solvents and spent resins.

Cation exchange, the method we studied most extensively for removing the trivalent elements, appears promising. In this process, the trivalent elements and certain of the fission products (primarily zirconium and some palladium) are loaded on cation resin from 1 M HNO_3 solutions of waste and the trivalent elements are then eluted selectively with 3.5 M HNO_3 . We have demonstrated greater than 99.9% recovery using americium, europium, cerium, and erbium tracers in synthetic waste. Aside from ion exchange resin, nitric acid is the only chemical reagent that is added in the processing.

Coupling oxalate precipitation with ion exchange may be an attractive alternative to an all-ion-exchange process for removing the trivalent elements. This process has been demonstrated in the laboratory using synthetic wastes. A single oxalate precipitation was used in a first step ^{to} ~~for~~ remove about 95% of the trivalent elements and also to complex the zirconium; then a cation exchange column was used as a final "polishing" step to remove the residual trivalent elements, as well as any solids or colloidal material remaining after the precipitation step. Considerably less ion exchange capacity is required in these operations than in the all-ion-exchange process. Measurements with radiotracers showed greater than 99.9% recovery of the trivalent elements. It is not necessary for the separation of oxalate precipitate in the first step to be quantitative since any residual precipitate will be retained by the ion exchange column and will be dissolved when the trivalent elements are eluted from the resin column with 3.5 M HNO_3 . A question yet to be resolved with regard to the oxalate precipitation step is whether the precipitation and solids removal step is feasible in remote operations.

Tributyl phosphate extraction from heavily salted solutions may be a viable method for the recovery of the trivalent elements [11,12]. If a satisfactory method can be developed for recycling the salting agents, Purex-type technology might be extended to remove^{al of} americium-curium and transcurium elements along with the heavy lanthanides at the time of fuel reprocessing. In nitrate-salted systems, the light lanthanides (about 80 mole % of the lanthanide fission products) can be partitioned from the trivalent actinides and heavy lanthanides. Laboratory studies with synthetic wastes have shown that about 95% of the salting agent, $Al(NO_3)_3$, can be recovered for recycle by precipitating it from 15 M HNO_3 solution. The extra acid required to precipitate the salting agent could then be recovered by distillation and reused.

Ion exchange chromatography appears attractive as a method for separating the trivalent actinides from the lanthanides in both conceptual flowsheet and laboratory studies. Emphasis of the laboratory work has been directed to the use of a hydrogen ion barrier for elution rather than a metallic ion barrier such as zinc, because this would diminish the addition of nonvolatile solids to the waste streams. Laboratory studies show that a barrier ion composition of 80% H^+ --20% Zn^{2+} yields good separations. This composition considerably reduces the quantity of zinc that is required and simplifies the treatment of waste streams from the separations processes.

We have completed conceptual material-balance flowsheets for the recovery of americium-curium by an all-ion-exchange process and for recovery of the trivalent elements by oxalate precipitation coupled with ion exchange. We view these flowsheets as tentative and preliminary since many of the process

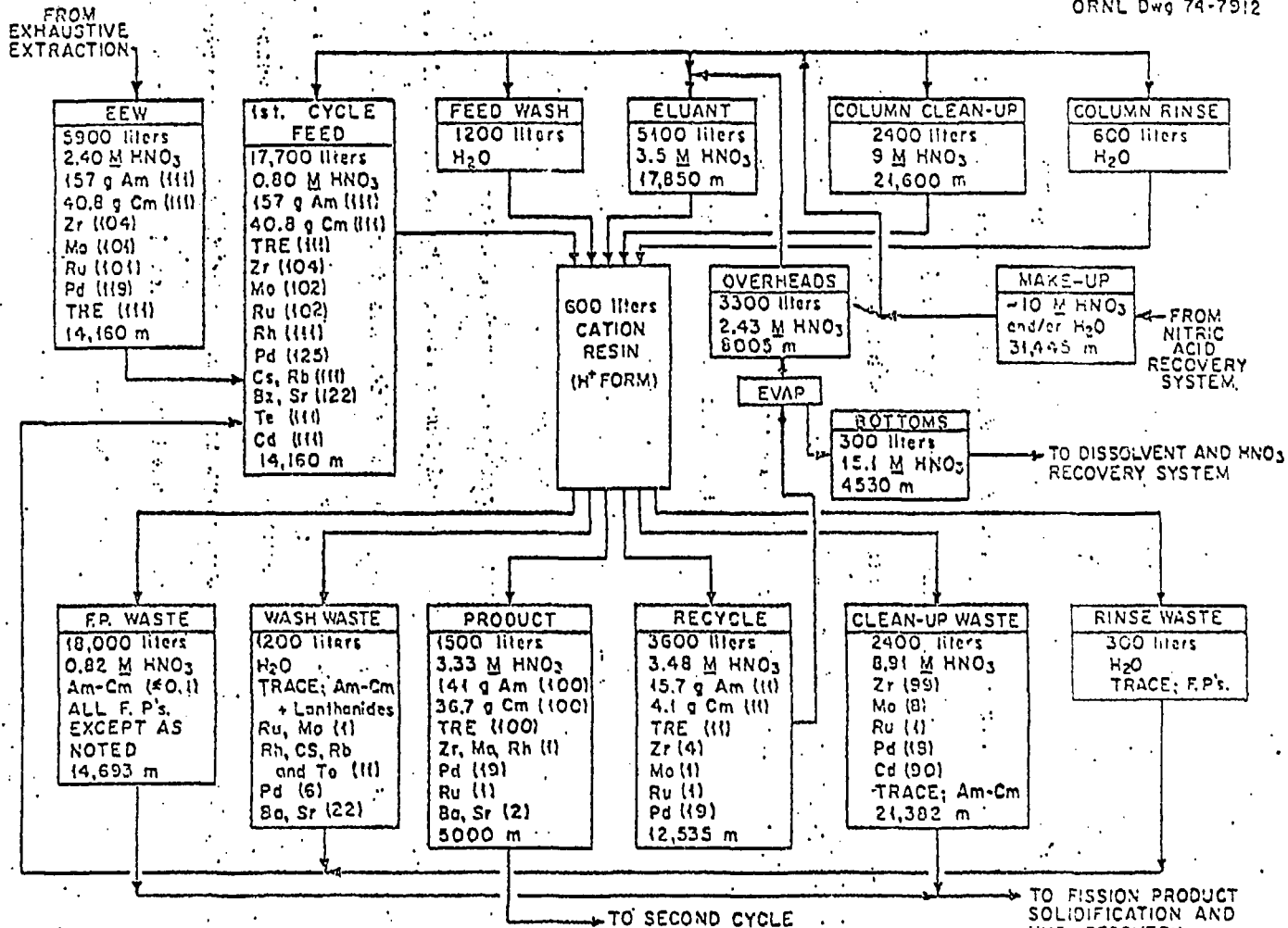
steps require experimental verification at full activity level and in regard to performance of recycle concepts and minor fission product behavior. A great deal of work remains to be done to confirm the overall flowsheets. The conceptual flowsheet for the all-ion-exchange process for removing the trivalent actinides and lanthanides is shown in Fig. 3. Material balances are based on the high-level waste from reprocessing 1 metric ton of LWR fuel irradiated to a burnup of 33,000 MWd/metric ton. It was assumed that the residual uranium, neptunium, and plutonium had been removed by extra extraction stages with TBP leaving the aqueous raffinate (EEW) containing the americium and curium.

Formation of Solids in Waste Solutions

Studies are being carried out to identify the solids that are present in waste and, ultimately, to determine their actinide content. In most of our work, we have used synthetic waste solutions; however, we have recently obtained solid residues from the dissolution of irradiated LWR fuel (burnup, 33,000 MWd/metric ton). Synthetic waste solutions contain a small amount of solids when they are prepared. These solids consist primarily of zirconium, tin, silver, halides, and some selenium.

On aging the waste solutions, small amounts of solids containing predominantly zirconium and molybdenum continue to precipitate. Small quantities of colloidal material are also formed during the aging and can be detected by a Tyndall beam. Although analyses of the residues from fuel dissolution are incomplete, qualitative spectrographic analysis indicates that such residues consist mainly of technetium, ruthenium, palladium, silicon, and rhodium, plus small amounts of zirconium, molybdenum, nickel, and iron.

The retention of plutonium by synthetic waste solids is discussed in Sect. 8.4.



Note: Nos. in () are %'s of a unit fuel charge.
 m = moles of HNO₃

Fig 3

Fig. 1. CONCEPTUAL MATERIAL BALANCE FLOW SHEET FOR THE
 Fig. 2. Americium-curium removal: feed preparation; first-cycle
 ion-exchange using only HNO₃.

FOR ION-EXCHANGE REMOVAL OF AMERICIUM-CURIUM AND
 THE RARE EARTHS FROM HIGH-LEVEL WASTE.

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Plutonium Behavior in Synthetic Wastes

It is anticipated that high plutonium recoveries (99.99%) during reactor fuel reprocessing will be difficult to achieve due to the formation of inextractable plutonium species. Therefore, studies were initiated to investigate reactions that can result in plutonium losses and to determine the basic mechanism involved.

Since plutonium will be recovered from spent reactor fuels by the Purex process, in which Pu(IV) is extracted into TBP from HNO_3 feed solution, experiments were designed to determine the stability of Pu(IV) in HNO_3 and in synthetic fission product solutions at various temperatures and acidities of interest. Some of these conditions are conducive to hydrolysis and precipitation of several of the fission products, and efforts were made to determine the effects of this behavior on plutonium extractability.

To date, we have identified three principal problem areas that can result in plutonium losses prior to or during Purex processing. These include: (1) decreasing plutonium distribution coefficients with successive TBP extraction stages, (2) Pu(IV) interaction with the ruthenium-rhodium-palladium components of synthetic feed, and (3) plutonium losses associated with precipitates of zirconium and zirconium-molybdenum. The basic mechanisms involved in these reactions have not, as yet, been determined.

It was found that a small but significant amount of plutonium does not extract into 30% TBP--dodecane from a 3 M HNO_3 feed solution. This effect involved ~0.01% of the plutonium present in the feed solution and resulted

in a decrease in plutonium distribution coefficients from 17.0 to 0.2 in seven consecutive extraction stages. The initial plutonium was highly purified, and spectrophotometric analysis indicated that it was composed entirely of ionic Pu(IV); however, the limits of detection for other species are well above 0.01%. The nature of this inextractable plutonium has not yet been identified; identification will be difficult because of the very low concentration.

A second type of reaction which has been observed involves Pu(IV) instability when synthetic feed solutions are aged or heated. This instability was found to be due primarily to interaction with the ruthenium component, although similar effects were observed to a smaller degree with rhodium and palladium. At room temperature, Pu(IV) is not stable in the presence of ruthenium, rhodium, and palladium at acid concentrations of 2 M or less. As much as 50% of the Pu(IV) can convert to other species in about 10 days. When such solutions are heated, the change occurs very rapidly and greater than 90% of the Pu(IV) can be converted at 80°C even in fairly concentrated acid solution (3 to 7 M). In preliminary efforts to identify the species to which the Pu(IV) is converted, it was determined spectrophotometrically that the Pu(IV) was oxidized to Pu(VI); HDEHP and TBP extraction behavior also indicated that the species was Pu(VI). The principal ^{effect} ~~effort~~ of the ruthenium-plutonium interaction on plutonium extraction behavior in the TBP-HNO₃ system was the reduction of distribution coefficients by a factor of 2 to 3, although in some experiments small third-phase losses occurred due to solids formation.

Since plutonium losses can result from the formation of solids in feed or waste solutions, the hydrolytic and precipitation behavior of zirconyl nitrate is being studied at various conditions of aging and heating. At 60 to 80°C, there is a consistent trend in the dependence of zirconium hydrolytic behavior on HNO_3 concentration, and this trend was not altered by the presence of $\text{Pu}(\text{NO}_3)_4$. At low acid concentrations (1 M or less), colloidal solutions of zirconia are formed. These colloidal solutions are very stable, and formation appears to be irreversible in that the particles do not readily dissolve or ~~disperse~~ disperse even in concentrated acid solutions. Hydrolysis of $\text{ZrO}(\text{NO}_3)_2$ in more-concentrated acid medium (> 2 M), results in the formation of zirconia precipitates instead of colloidal solutions. Maximum precipitation occurs at 2 M HNO_3 , and the amount of precipitate that forms decreases with increasing acid concentration. A small amount of precipitate is formed even in 6 M HNO_3 .

The effect of zirconium hydrolysis on plutonium-TBP extraction behavior was evaluated in two experiments. In the first, $\text{Pu}(\text{NO}_3)_4$ was heated at 80°C with $\text{ZrO}(\text{NO}_3)_2$ in 3 M HNO_3 until precipitation occurred. Some plutonium is undoubtedly associated with the zirconia precipitate, but the amount is too small to be detected by gross alpha analysis of the feed solution. The effect on plutonium extraction behavior is also quite small, although plutonium losses are a factor of 3 greater than for solutions of $\text{Pu}(\text{IV})$ alone after five extraction stages.

In the second experiment, 1 M HNO₃ containing both Pu(NO₃)₄ and ZrO(NO₃)₂ was heated at 80°C. This gave rise to a major plutonium loss mechanism in which 2% of the plutonium became completely inextractable in TBP from 3 M HNO₃.

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- [1] "High-Level Radioactive Waste Management Alternatives," WASH-1297, U.S. Atomic Energy Commission (1974).
- [2] H. C. Claiborne, "Neutron-Induced Transmutation of High-Level Radioactive Wastes," ORNL-TM-3964, Oak Ridge National Laboratory (1972).
- [3] H. C. Claiborne, "Effect of Actinide Removal on the Long-Term Hazard of High-Level Waste," ORNL-TM-4724 (1975).
- [4] W. D. Bond and R. E. Leuze, "Feasibility Studies of the Partitioning of Commercial High-Level Wastes Generated in Spent Nuclear Fuel Reprocessing: Annual Progress Report for FY-1974," ORNL-5012 (1975).
- [5] W. D. Bond, H. C. Claiborne, and R. E. Leuze, Nucl. Technol., 24 (1974) 362.
- [6] J. O. Blomeke, J. P. Nichols, and W. C. McClain, Phys. Today, 26, (1973) 36.
- [7] E. J. Wheelwright and F. P. Roberts, "The Use of Alternating DTPA and NTA Cation-Exchange Flowsheets for the Simultaneous Recovery and Purification of Pm, Am, and Cm," BNWL-1072, Battelle-Northwest Laboratories (1968).
- [8] J. A. Kelley, "Ion Exchange Process for Separating Americium and Curium from Irradiated Plutonium," DP-1308, E. I. DuPont deNemours and Co. (1972).
- [9] B. Weaver and F. A. Kappelmann, "Talspeak: A New Method of Separating Americium and Curium from Lanthanides by Extraction from an Aqueous Solution of Aminopolyacetic Acid Complex with a Monoacidic Phosphate or Phosphonate," ORNL-3559, Oak Ridge National Laboratory (1964).
- [10] B. Weaver and F. A. Kappelmann, J. Inorg. Nucl. Chem., 30, (1968) 263.
- [11] M. C. Thompson, "Distribution of Selected Lanthanides and Actinides Between 30% TBP in n-Paraffin and Various Metal Nitrate Solutions," DP-1336, E. I. DuPont deNemours and Co. (1973).
- [12] J. N. McKibben et al., Partitioning of Light Lanthanides from Actinides by Solvent Extraction with TBP, DP-1361, E. I. DuPont deNemours and Co. (1973).