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REDUCING THE LONG-TERM HAZARD OF REACTOR WASTE THROUGH ACTINIDE REMOVAL AND DESTRUCTION IN NUCLEAR REACTORS

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ABSTRACT

Public opposition to nuclear power has focused on the long-term risks from reactor waste. In the Purex process used in Europe, this waste is a concentrated nitric acid solution containing all nonvolatile fission products and the actinides Np, Am, and Cm, plus smaller amounts of U and Pu.

Techniques have recently been described which guarantee an absolutely safe containment of this high-active waste (HAW) for about 1000 years. At longer times, the risk to the biosphere is dominated by the actinides. If these actinides are isolated from the rest of the HAW and destroyed through nuclear incineration, the long-term risks of the HAW will be dramatically reduced.

This paper presents a detailed scheme for removing the actinides from the Purex-HAW solution. In principle, the process consists of three different solvent extraction cycles, using HDEHP and TBP in three successive steps. The scheme has been tested on a synthetic HAW solution containing all fission products and actinides (except $Z \geq 96$, Cm) using laboratory-scale mixer-settler batteries. Results from runs on old Purex waste are also presented.

If applied to fresh Purex waste, the process will encounter problems due to radiation damage to the reagents. In practice, this difficulty can be circumvented by using short contact times in the solvent extraction process. Extremely rapid multistage solvent extraction separations can be carried out by the SISAK technique (i.e., batteries of static mixers and special centrifugal separators). This technique is also described.

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INTRODUCTION

One of the obstacles to the further use of nuclear energy is the lack of a firm policy for handling the highly radioactive material which is produced by nuclear reactors. In most countries with sizable nuclear energy programs, reprocessing of the spent reactor fuel elements is considered advantageous for saving resources as well as for economic and safety reasons. Of the waste streams produced in reprocessing, the high-level liquid waste (HLLW) has caused most concern, in spite of official studies in several countries which indicate that this HLLW can be solidified and safely stored in geologic repositories (1-4). However, if the long-lived alpha-active nuclides are removed from the waste and destroyed (by fissioning) in nuclear reactors (5-15), the long-term risk from the solidified HLLW is reduced to a 500-year storage problem. In such a case, geologic deposition may be unnecessary.

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Alpha-Waste Streams

The alpha-waste streams produced in a 30-MWe nuclear fuel cycle are summarized in Table 1. The values are only indicative of volumes and composition, and are based on the experience available in the early 1970s (6b).

The dominating risks are the specific alpha-activity and its availability, and the amount of plutonium. (The latter, especially, has caused considerable public concern.) The table indicates that high values of both appear in many streams of the fuel cycle, in addition to the HLLW. It would thus appear as if a removal of alpha-activity from the HLLW alone would only partly solve the alpha-waste problem. However, in recent years techniques have been developed which reduce the alpha-active waste in mixed oxide fuel (MOX) handling (14), as well as from the reprocessing of high-level solid (9,16) and medium level (6,12,15,17) waste streams by factors of 10 to 100. The actinides removed from these streams end up in the HLLW. Therefore, practically all alpha activity and plutonium not recovered in the main reprocessing will appear in the HLLW.

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TABLE 1

Alpha-waste Streams of a 30-GWe PWR Nuclear Fuel Cycle, Annually Consuming 1000 tonnes of enriched uranium and producing 10 tonnes of plutonium. Decay time is 10 years (See Ref. 6)

Source	Volume[a] (m^3)	Specific activity ($\alpha Ci/m^3$)	Total activity (αCi)	Pu weight (kg)	Radiolysis heat (α,β,γ W/L)
Uranium mining and milling[b]	$3x10^6$(s)	1×10^{-3}	2,370	0	Nil
MOX fuel element fabrication[c]	1100(s)	18[d]	20,000[d]	40	Negligible
	12(l)	380[d]	4,500[d]	9	0.01 - 0.1
Reactor operation[e]	300(s)[f]	17	5,000	0.5	Negligible
Reprocessing[g], HLLW	400(l)	50,000	20×10^6	152[h]	3.5
HLSW	50(s)[i]	1900	95,000	13	0.2
MLW	110(l)[j]	450	50,000	5.5	0.01 - 0.1
LLW	$4x10^4$(l)	0.6×10^{-6}	0.02	0.003	Negligible

[a] Solid (s), liquid waste (ℓ).

[b] Tailings from 6000 t U produced per year from 0.1% U ore; volume increase due to ore treatment is neglected.

[c] Annual production of ∿200 t MOX fuel containing 10 t Pu.

[d] From Pu isotopes only.

[e] 30 reactors at 1000 MWe each.

[f] Ion exchange resins from primary cooling circuit.

[g] Reprocessing, 1000 t heavy metal/year.

[h] Additional 86 kg will grow in from decay of Am-243 (half-life 7400 years).

[i] Mainly cladding hulls compacted to density 6.4; uncompacted average density is ∿1.0.

[j] Mainly neutral salts from cleaning circuits.

Consequently, a removal of alpha-activity from the HLLW dramatically reduces the alpha-waste risk of the whole fuel cycle.

Long-Term Hazard of the HLLW

The decay of the radioactive nuclides in the HLLW leads to a reduction of the radiological hazard with the passage of time. This is commonly illustrated as in Fig. 1. The curves are summations of contributions from individual nuclides (Fig. 2). Reprocessing (Curve II, Fig. 1) and actinide fractionation (Curve III) considerably reduce the toxicity of the waste after about 500 years. The difference between Curves I and II is largely due to a reduction in the plutonium content.

The horizontal lines in Fig. 1 give the hazard indices for (a) 6 metric tons (tonnes) of natural uranium, (b) 1 tonne enriched light water reactor (LWR) fuel, and (c) the amount of uranium destroyed in the reactor operation. Below line (c), the radiological hazard is less than it would be without nuclear reactors. In waste management, it would not be meaningful to try to reach below this level, which we designate here as "safe-as-nature;" it is implicitly assumed that the waste is in as nonleachable (glass, ceramic, etc.) form as the naturally radioactive elements are in the rock. The curve for the spent fuel elements never does reach the "safe-as-nature" line. For the HLLW, it will take about 10 million years, while the actinide-free HLLW will be as "safe-as-nature" after 1000 years. It is interesting to note that all reviewers of the Swedish plan for handling high-level waste have agreed that the waste can be stored in an absolutely safe manner during at least 500 to 1000 years (4).

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Destruction of Waste Actinides in Nuclear Reactors

The HLLW from 1000 t spent fuel will contain $\leq$ 200 kg Pu (i.e., $\leq$ 2% of the 10 tonnes produced, Table 1), ∿1000 kg U (i.e., 0.1% of the 1000 tonnes spent U fuel), ∿500 kg Np, ∿140 kg Am, 40 kg Cm, and smaller amounts of heavier elements (Table 2). It has been suggested that not all, but only some, of these elements should be

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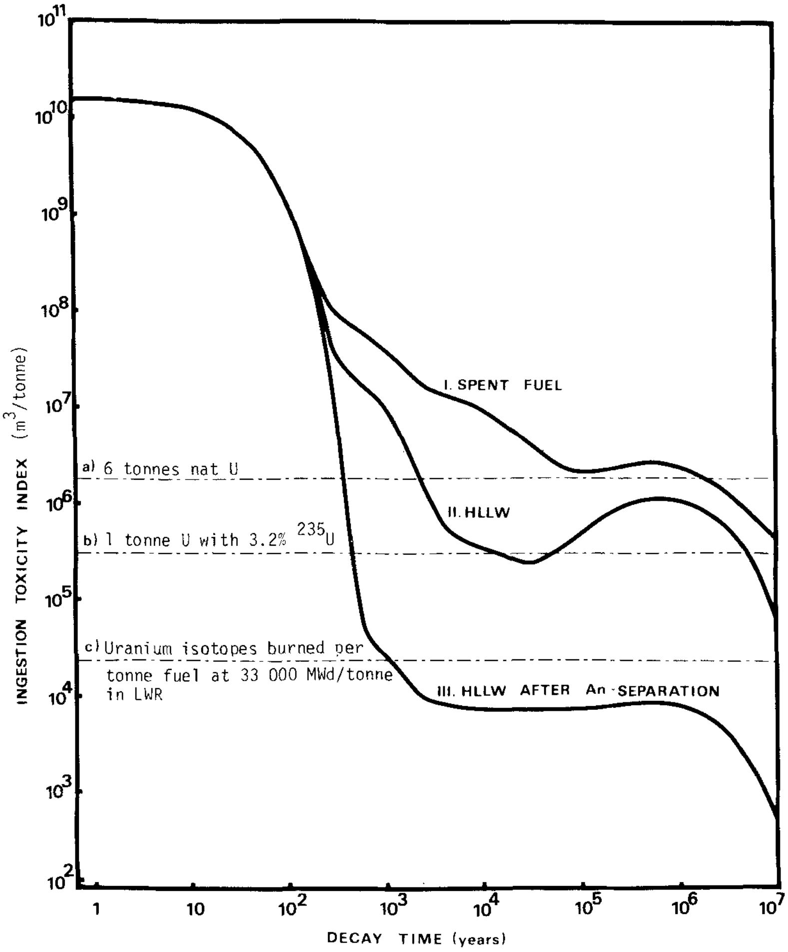

FIGURE 1. Ingestion toxicity index (m^3 water at maximum permissible level MPC_W, per tonne spent fuel element) for spent LWR fuel (curve I), high-level liquid waste from PUREX reprocessing (curve II), and such wastes after actinide removal (curve III). Line (a) is the index for 6 tonnes natural uranium (the amount needed to produce 1 tonne LWR fuel enriched to 3.2% in U-235), (b) is for 1 tonne U enriched to 3.2% in U-235, and (c) index for that amount of uranium isotopes which has been destroyed in the reactor process (33,000 MWd/t U); all lines assume equilibrium with all daughter elements.

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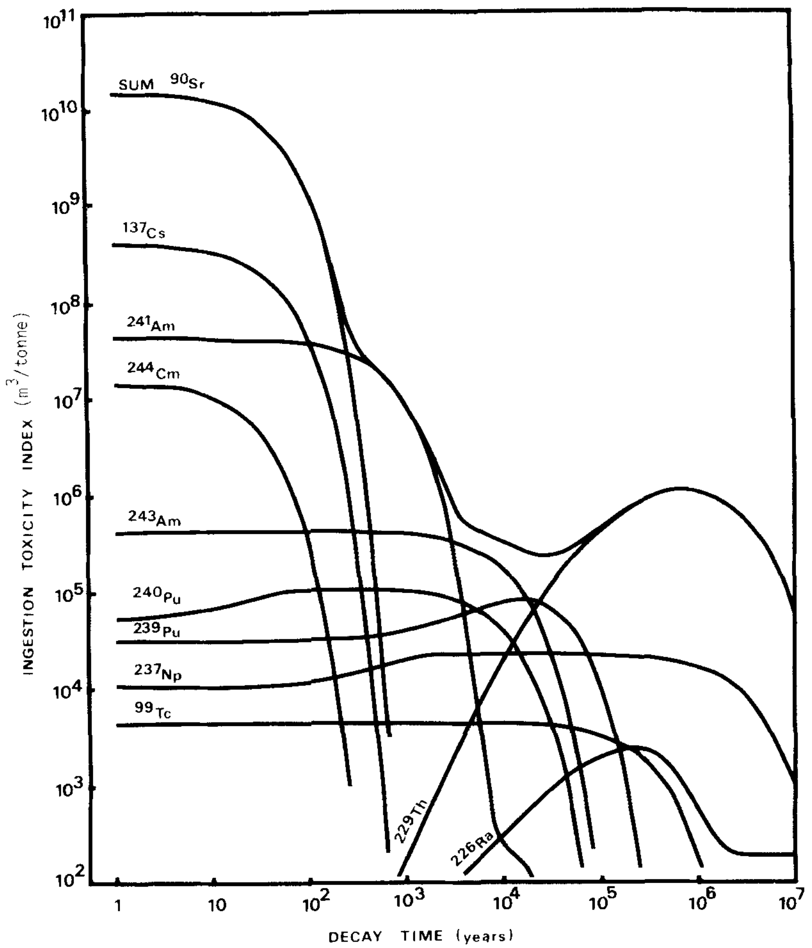

FIGURE 2. Ingestion toxicity index for HLLW containing all Np, Am, and Cm, 0.1% U and 0.5% Pu as a function of time after reprocessing (carried out after 10 years cooling time).

recycled for incineration. Since the main long-term hazard is due to ^{229}Th (Fig. 2), which is a decay product of the main neptunium isotope, ^{237}Np, neptunium must be incinerated. The main short-term hazard is due to the americium isotopes, ^{241}Am and ^{243}Am; thus americium must also be incinerated. Likewise, plutonium must be

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TABLE 2

Composition of HLLW from Purex Reprocessing 1 tonne LWR Fuel of 33,000 MWd/t Burnup, at 30 MW/t U. Original Waste Volume ∿5 m^3 (in 2 M HNO_3)/t U reprocessed (6). Cooling time 10 years.

Component		Weight (kg) in original waste volume	Approximate molar concentration after waste is evaporated to ∿0.5 m^3
H^+			2.0
NO_3^-			3.6
Fission products:	Rb, Cs	2.66	0.045
	Sr, Ba	2.59	0.045
	Y, Ln	8.09	0.140
	Zr, Mo, Tc	8.09	0.167
	Ru, Rh, Pd	3.70	0.070
	Others	1.10	0.015
Total fission products		26.2	0.48
Corrosion products		1.40	0.05
Phosphate		0.9	0.02
Actinides	U, (0.1%)	1.00	0.008
	Np (100%)	0.50	0.004
	Pu (0.5%)	0.05	0.0004
	Am (100%)	0.14	0.0012
	Cm (100%)	0.04	0.0003
Total actinides		1.73	0.0139
Radioactivity from fission products (Ci)		230,000	
Radioactivity from actinides (Ci)		19,000	
Thermal power (W)		1,300	

incinerated, since ^{241}Am is formed by beta-decay of ^{241}Pu. An interesting aspect is that isotopes of curium and the heavier elements have little influence on the ingestion hazard index, except for the period between 10^3 and 10^4 years. It is quite feasible to separate curium and the higher elements from the waste actinide mixture using chemical techniques presently available.

Of the many concepts suggested for nuclear incineration of the actinides, fissioning in light water reactors and fast breeders looks most promising (9,10,12,15). Reactor parameters (spectrum, reactivity, specific power, etc.) will be influenced only slightly by the waste transmutation. However, the buildup of heavier actinides leads to nuclides with considerable neutron emission rates, which will increase handling difficulties. Calculation of the rate of destruction requires data for reaction cross sections, neutron spectrum, actinide content in the fuel pins and their position in the reactor, etc. Although calculations have been made by many authors, the results are uncertain and strongly dependent on the irradiation strategies chosen. For example, the actinides may be irradiated as separated pure elements, or mixed with other actinides or diluting elements, or some elements burned in light water reactors (LWR) (e.g., Np and Pu), and others in fast breeder reactors (FBR) (e.g., Am and Cm), etc.

The simplest approach is to return the separated actinides in bulk to LWRs, whether these are run on a plutonium recycle or a regular enriched uranium scheme. The latter is illustrated in Fig. 3 where it is assumed that the plutonium from the reprocessing plant is not recycled but stored for future use in breeders; this is in line with the nuclear programs of many European countries. The waste actinides are returned to 1/4 of the pressurized water reactors (PWR), which are run on a higher ^{235}U-enrichment to compensate for the capture cross sections of the additional waste actinides. Although the waste actinides can be homogeneously mixed with the uranium fuel, it might be preferable to keep them in separate target pins. Thus ^{237}Np in separate pins would function almost

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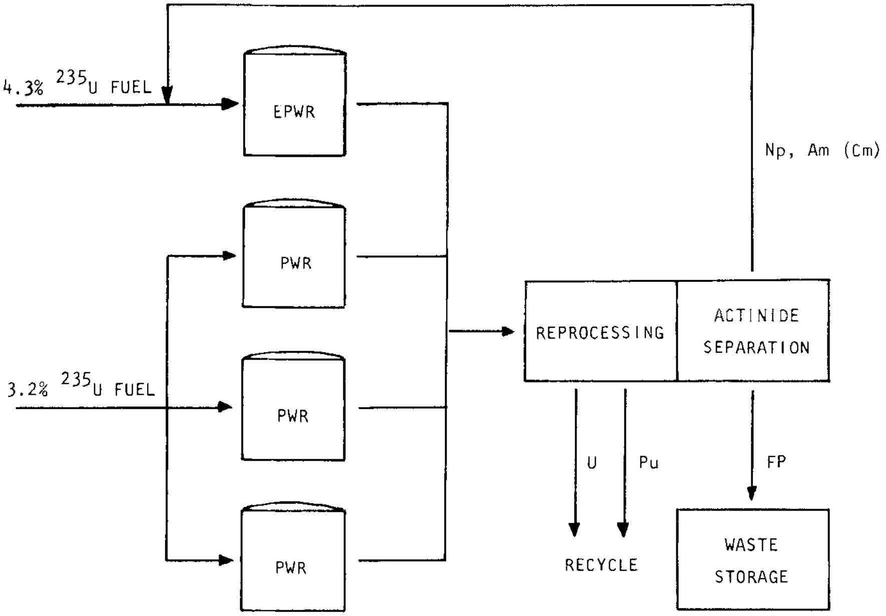

FIGURE 3. Scheme for actinide separation and transmutation.

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exactly as the present boron pins used in LWRs as burnable poison. A limiting factor may be the actinide decay heat in the fuel pins, restricting the actinide content to only a few weight percent.

Of the waste actinides returned to the LWRs, 6 to 7% will be burned up per year, or 20 to 25% in a fuel pin lifetime. At the same time, new actinides are formed in the reactor, so that the overall reduction of waste actinides (Np, Am, Cm) will be less, about 10% in a three-year full-power irradiation cycle (15). After ten cycles, the transuranic inventory will be 1/2 to 1/3 of what it would have been without the partitioning-transmutation cycle (19). Higher values have been given for incineration in FBRs (15).

In a steady-state fuel recycle condition, which may take several decades (depending on the irradiation strategy), the amount of waste actinides will be reduced by a factor of 5 through recycling-transmutation. The ingestion toxicity of the waste will be 100 to 200 times lower than in the case of no partitioning-transmutation for cooling times $\geq 10^3$ years (15), as illustrated in Fig. 1.

Removing the Actinides from the High-Level Liquid Waste

In the Purex process, tributyl phosphate (TBP) dissolved in kerosene is used as an extracting agent in a number of solvent extraction steps for the isolation of uranium and plutonium. In order to remove the actinides from the HLLW obtained in this process, various modifications of the Purex scheme have been applied (8,9, 12,15) or suggested. To a lesser extent other more powerful extractants have been used, such as di-ethylhexyl phosphoric acid (HDEHP) for extraction from moderately acid solutions, or dibutyl-N,N-diethyl carbamyl methylene phosphonate (DBDECMP) from strongly acidic solutions (12).

The problem is far greater when it comes to the separation of the higher actinides (Am, Cm, and transcurium elements) from the lanthanides because of their chemical similarity--both groups having mainly one stable oxidation state (+3) in aqueous solution. More elaborate separation schemes must therefore be used, such as cation exchange from highly salted solutions, anion exchange using organic amines (the Tramex process), extraction with HDEHP in the presence of hydrophilic complex formers, such as diethylenetriamine pentaacetic acid (DTPA) (the Talspeak process), or the use of more exotic chelating reagents, as 5,7-dichloroxine (12).

We shall describe here only the separation scheme developed in our laboratory, which is designated as the "CTH-process." This process has shown, both on the laboratory and small pilot-plant scale, that the lighter actinides (LAn) as well as the heavier actinides (HAn) can be extracted with a high yield (>99%) from Purex HLLW solutions. Through a series of solvent extraction steps using TBP and HDEHP, the lighter (U, Np, and Pu) and heavier (Am and Cm) actinides are obtained without any essential increase in the solidified volume of either the actinide or the remaining actinide-free waste.

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PROCESS DESCRIPTION

Chemical Procedure (19,20)

The overall flow scheme is presented in Fig. 4. In principle it consists of five steps: (1) waste conditioning; (2) extraction and separation of U, Np, and Pu using HDEHP; (3) extraction and separation of Pd, Tc, and Ru using TBP; (4) extraction and separation of Am and Cm using HDEHP; and (5) fixation of the radionuclides in the remaining waste solution.

Waste conditioning. From Table 2 it is seen that the original HLLW, when concentrated to 0.5 m^3/tonne reprocessed heavy metal, contains about 60 g metals per liter of 2 $\underline{M}$ HNO_3 solution. This deep-brown solution contains undissolved material. Before solvent extraction it is therefore diluted to 10 g/L of metal and made 6 $\underline{M}$ in HNO_3. NO_x gas is bubbled into this solution to produce stable valency states and nitrosyl complexes. The resulting solution is clear, stable, and reddish-brown.

Extraction and separation of U, Np, and Pu using HDEHP. The first HDEHP cycle is shown in Fig. 5. The LAn are extracted together with a number of fission product elements, leaving the HAn and Ln in the raffinate. In the second step, Y and the small amounts of HAn and Ln extracted, are stripped together, while the LAn are left in the organic phase. This phase is then successively washed to remove Zr and Nb before the LAn are stripped along with Mo, In, and Fe.

To avoid precipitation of iron in the stripping operation, a mixture of concentrated ammonia and mannitol is added to the organic phase before it is contacted with the ammonium carbonate strip solution. This strip solution is then evaporated to 50% of the original volume, thereby recovering the excess ammonium carbonate in the condensate. It is then acidified with nitric acid and evaporated further. The volume reduction possible in this stage is limited by the solubility of uranyl nitrate. The recovered ammonium carbonate solution is diluted with water and recirculated to the stripping circuit.

In experiments using trace concentrations of Np and Pu, the

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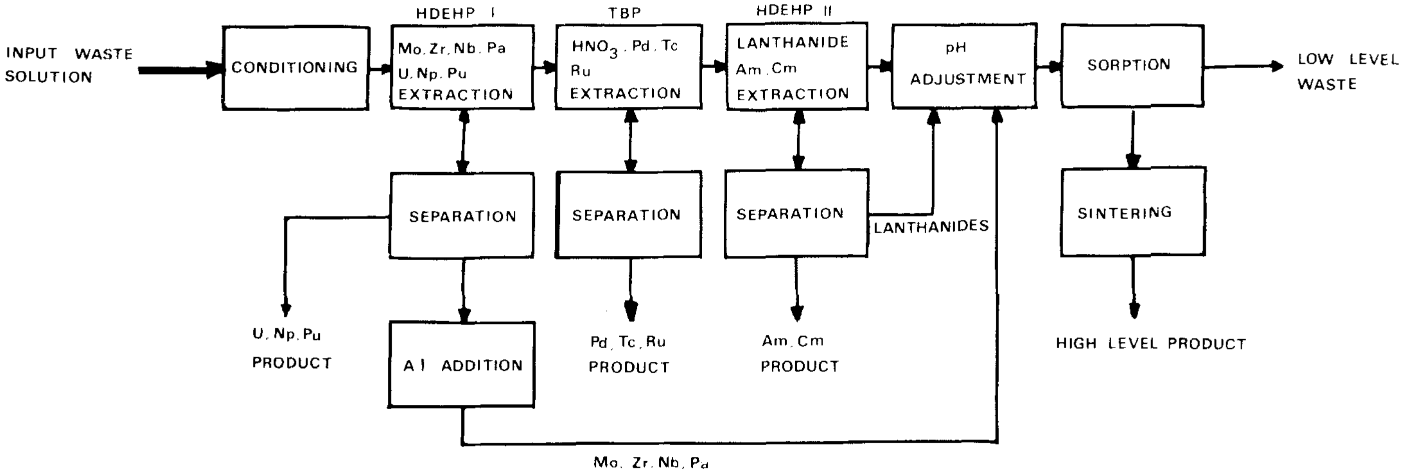

FIGURE 4. Flowsheet for fractionation of high-level liquid waste from Purex reprocessing.

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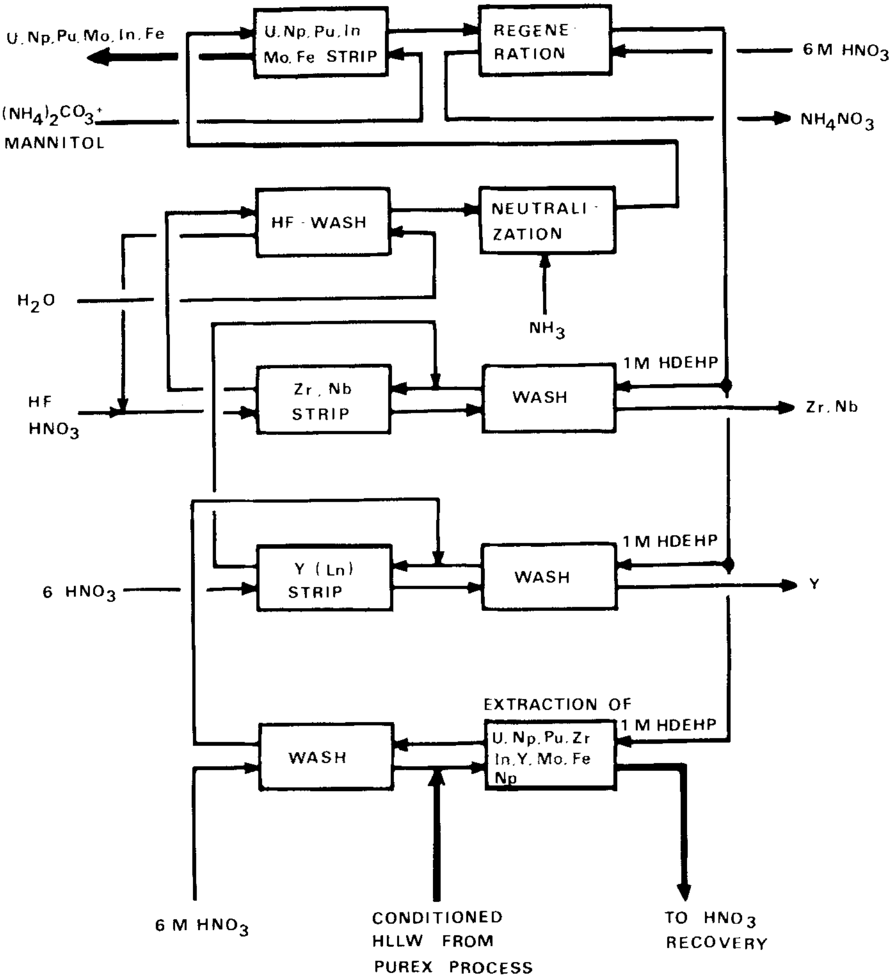

FIGURE 5. Separation of lighter actinides (U, Np, and Pu) from HLLW through HDEHP extraction.

product solution was found to contain 99.99%, 98.9%, and >99.99% of the U, Np, and Pu, respectively, present in the original HLLW fuel solution.

Extraction and separation of Pd, Tc, and Ru using TBP. The second extraction cycle uses TBP (Fig. 6). Its main purpose is to reduce the acidity of the aqueous phase in order to prepare it for a second

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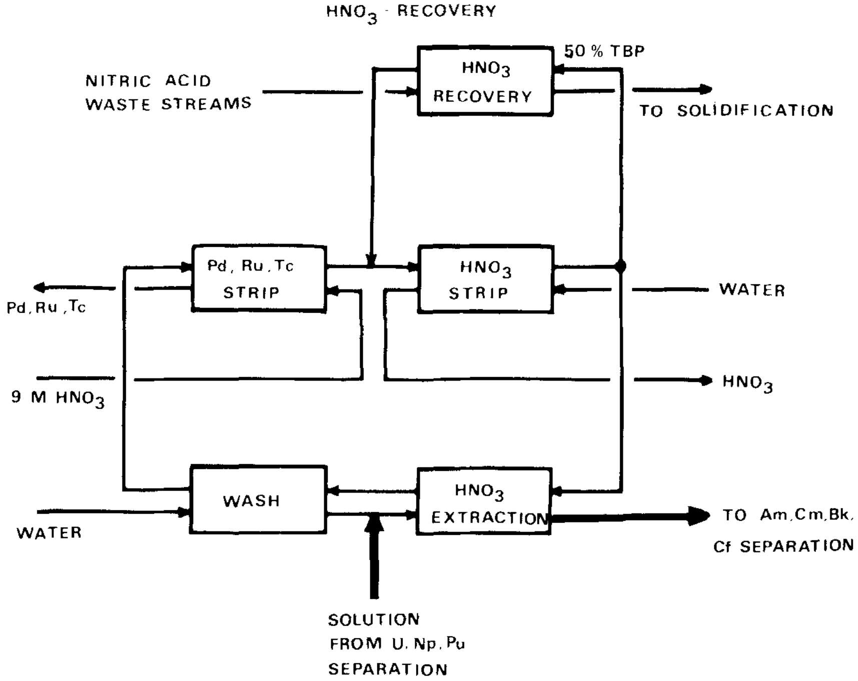

FIGURE 6. TBP extraction step for HNO_3 removal.

low-acidity HDEHP extraction. TBP (dissolved to 50% in kerosene) is very efficient as an extractant for HNO_3; it can be used effectively for precipitating metal hydroxides from nitrate solutions. Together with the HNO_3, TBP also extracts Pd, Ru, and Tc, leaving the HAn, Ln, and the rest of the fission and corrosion products in the 0.1 M HNO_3 aqueous raffinate phase.

The main design problems with this cycle have been to avoid partial extraction of Am and Cm, and internal circulation of Pd. This has been accomplished by the use of a water scrub stage, where Am and Cm are forced out with the raffinate and Pd leaves with the organic extract. The 3 M HNO_3 recovered in the strip battery is evaporated and distilled to produce about 9 M HNO_3, which is mainly reused in the process. Paladium, Ru, and Tc can be recovered by anion exchange from the strip solution. However, the detailed design of this operation has not been completed. Recoveries of these elements are 98% of Pd, 74% of Ru, and 67% of Tc. These yields may be increased at the cost of larger volumes and more dilute solutions.

Extraction and separation of Am and Cm using HDEHP. By contacting this raffinate with HDEHP in the third extraction cycle (Fig. 7),

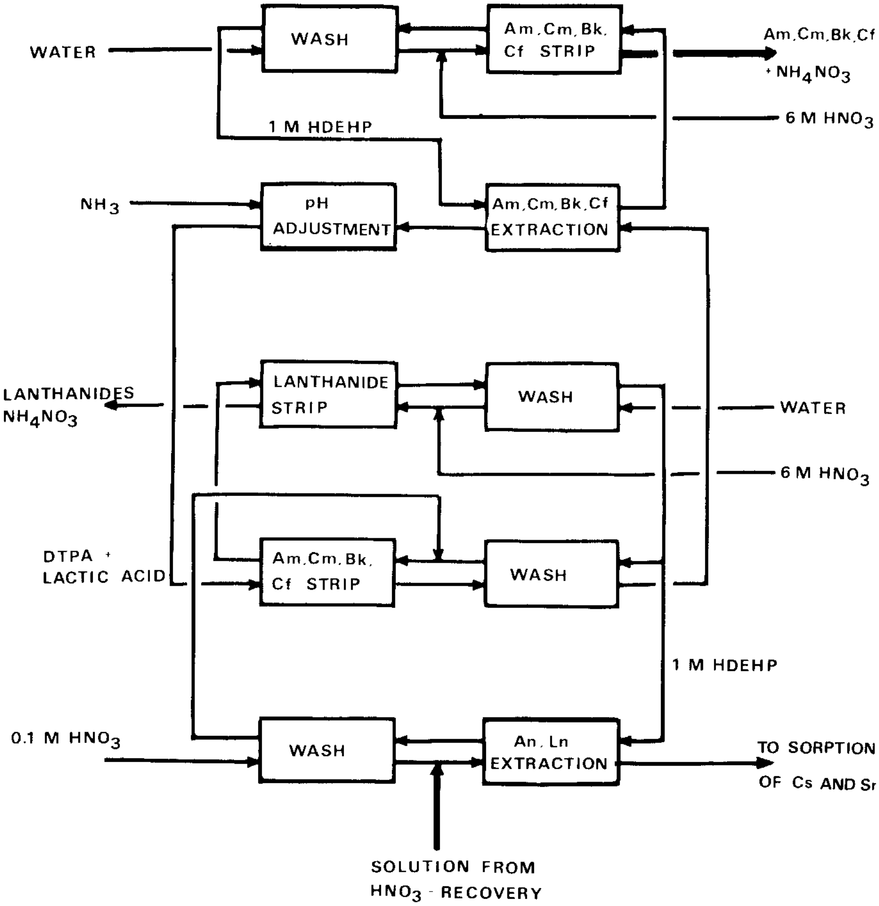

FIGURE 7. Separation of heavier actinides (Am, Cm, Bk, and Cf) from process solution through HDEHP extraction.

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all trivalent An and Ln are extracted together, leaving the rest of the fission products in the aqueous phase. The actinides in the organic phase are then stripped with DTPA and lactic acid, as DTPA forms stronger hydrophilic complexes with the actinides than with the homologous lanthanides at the pH of the lactic acid buffer (pH, 3.3). The HAn are then further purified in a second HDEHP extraction step, after which they are stripped by 6 M HNO_3.

Since the distribution coefficient D for the actinide extraction is 1, the use of a circuit with a high organic-to-aqueous flow ratio allows extraction, and a low ratio, stripping of the actinides, and at the same time permits the recovery and reuse of the DTPA solution. One of the main design problems has been the extraction of ammonia and some lactic acid, which leads to pH shifts in the strip and extraction batteries.

The current design gives a separation factor between Am and Nd of about 8000, which is believed to be sufficient. No attempts to increase this factor further have been made. Recoveries obtained are about 99.8% of Am, and >99.9% of Cm, in experiments using trace concentrations of these elements.

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Fixation of the radionuclides in the remaining waste solution. The actinide-free waste obtained as raffinate from the third extraction cycle contains the Cs and Sr quantitatively (i.e., the main radiation sources of the 10-year-old HAW). These elements can be sorbed on inorganic ion exchangers (e.g., zeolites or titanites), which are fired afterward in order to reduce the leachability of the sorbed elements. This technique, which has been tested in several laboratories, can be used to fix all of the nonvolatile non-actinide waste elements (16,20).

Experiments on HLLW from low-burnup fuel (400 MWd/ton) have shown that practically all of the Cs is sorbed on a mordenite-filled column from the actinide-free HLLW. A second column filled with the zeolite 13-X sorbs practically all the remaining radioactive elements. When dried and sintered under pressure, these zeolites yield insoluble solids.

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Experimental Technique

The basic design data, D-values, and extraction rates have been obtained in batch experiments using a synthetic HLLW containing practically all elements known to be present in real HLLW (21,22). Realistic concentrations have been used for all elements, except radiotracer concentrations for Np, Pu, Am, and Cm.

Analyses were made using X-ray fluorescence, radioactive tracers combined with gamma-spectrometry, or atomic absorption photometry, depending upon the element. The results have been expressed as empirical equations with acidity as the independent variable (21,22). Using these equations, three computer programs were written, each permitting the simulation of one of the three circuits. The most promising operating conditions were then selected using these programs.

In parallel with the calculations, the results were verified in a number of small-scale (150 ml) mixer-settler experiments using the synthetic HLLW solution. This provided a feedback for improvements of the computer programs. In general, however, good agreement was found between calculated concentrations and analytical results.

Finally, the whole process was tested by processing 20 L of synthetic HLLW feed using the same type of 150-ml mixer-settlers as in earlier experiments. At the end of the run, the solutions in each settler, as well as all effluents, were sampled and analyzed. As a separate check, 30 ml of HLLW from low-burnup fuel were treated using normal batch techniques and small test tubes. In this case also the analytical results indicated that the process operated as predicted. The small mixer-settlers used are now being modified for remote operations, to permit a demonstration run treating about 20 L of old HLLW (400 MWd/ton).

SEPARATION TECHNIQUES

From laboratory data on the various steps of the Purex separation scheme, one would expect that the HLLW would be completly free of U and Pu, rather than to contain 0.1 to 1% of these elements.

The reason for this default is partly due to the techniques employed: (1) conventional solvent extraction equipment (mixer-settlers and pulse columns) have efficiencies $\leq$ 95% per stage. Even if the theoretical distribution ratio (as measured in the laboratory) of an element between the organic and aqueous phase is >1000, the practical value will be $\leq$20 in each stage of the conventional equipment due to the dispersion of droplets of the opposite phase in each bulk phase. (2) The organic solvent and extractants used undergo radiolysis due to the alpha, beta, and gamma decay of the actinides and fission products, leading to the formation of organic products that react undesirably with the dissolved elements (e.g., the formation of dibutyl phosphoric acid from radiolysis of TBP).

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Both of these effects would be detrimental to the separation scheme described. Since radiation doses >10 Wh/L would cause degradation even in the most radiation-resistant organic compounds, it is desirable to keep the doses below 0.1 Wh/L. The HLLW heat is about 3.5 W/L, while the conditioned feed for the proposed scheme would be lower. It is estimated for the 10-yr-old waste that the radiation doses to the organic phase in the three main extraction cycles will be 0.12 Wh/L, 0.03 Wh/L and 0.4 Wh/L, respectively, during a full solvent cycle. Obviously there is a risk for radiation damage to the chemicals used, and consequently lower yields than estimated above could be expected.

It is therefore desirable to seek a solvent-extraction technique which would minimize these problems. Such a technique is the use of centrifugal extractors. Centrifugal extractors are centrifuges into which two liquid phases continually flow, become mixed, and again separated. The mixer can also be placed outside the inlet to the centrifuge and thus be independent of the centrifuge rotational speed; although this unit acts as an extractor, it is rather a mixer/centrifugal separator (the MEAB-type, below).

The liquid flow centrifuge was invented 101 years ago. However, its introduction for solvent extraction in nuclear technology is rather recent (23-26). The properties of some such centrifuges with a capacity range of interest for reprocessing are summarized in Table 3. The capacity demand for reprocessing HLLW in order to separate out the actinides is ∿0.3 m^3/h (of 10 g metal/L concentration) for a fuel cycle of 1000 tU/year. The larger sized centrifuges (SRL-large and BXP) are better suited to handle the large Purex reprocessing volumes than the much smaller HLLW volumes.

Both the Robatel and the SRL centrifuges are used in presently existing reprocessing plants. The SRL-centrifuges are all single-stage, with a mixing chamber at the inlet; they can be added in batteries to any number of stages. The Robatel BXP is of the same type. The Robatel LX-centrifuges have several separating chambers stacked on top of one another and interspaced with mixing chambers--all on the same axis--so as to make up 4, 8, etc. stages per centrifuge. Both of these types of centrifuges have a stage separation efficiency of <95%; thus at least one of the outgoing separated phases has >5% entrainment of the other phase.

The MEAB centrifuge is unique in that it gives a separation efficiency of 100%; hence each out-going phase has an entrainment content of <0.01% of the opposite phase. It also has an extremely short holdup time (<1 s) and a variable mixing-time. The advantage of these properties is that the kinetics of the solvent extraction process can be used to ultimate advantage--very short (0.1 s) or very long (>20 s) contact times may be employed. The MEAB centrifuge is made of Pd-stabilized titanium, so that it can also be used in HCl and H_2SO_4 solutions, whereas the other centrifuges are made of stainless steel. The MEAB centrifuges have previously been used in nuclear chemistry research (27,28) and for processing spent reactor fuel waste on a small scale (29).

In general, centrifugal extractors offer the following advantages over columns and mixer-settlers: (1) shorter (at least a hundred times) contact time, which reduces radiation damage and makes it possible to use radiation-sensitive chemicals, with otherwise advantageous properties; (2) higher transfer rate, which makes it possible to achieve plant steady-state conditions in less than

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TABLE 3

Commercial Centrifugal Extractors and Mixer-Centrifugal Separators

Manufacturer	Savannah River Lab[a]		Robatel SLPI, Genas, France[b]			MEAB[c]
	SRL-small	SRL-large	LS 324	LX 208N	BXP 320	H-33
Type	Sharples-type separator with inlet mixing chamber, pneumatic interface control		Stacked stages of mixing chambers and disc type separating chambers		Sharples-type with inlet mixing chambers	Baffle separator and external mixer
Stages per unit	1	1	4	8	1	1
Max. capacity, m^3/h[d]	0.15	14	1.6	0.2	6	0.3
Holdup volume, liter	0.2	30	$\sim$10	0.37	17	0.1
Rotational speed, rpm	1800	1700	1700-3400	600-3600	2900	$\leq$16.000
Centrifugal force, *g*	430			37-1300		
Mixing time, s/stage[e]	0.5	0.6	$\sim$0.5		$\sim$1	$\sim$0.1-20
Holdup time, s, total	5	6	22		10	1

[a] E. I. duPont de Nemours, Savannah River Laboratory, Aiken, S.C.; commercialization uncertain (23,24).

[b] Robatel et Mulatier, Genas, France (25).

[c] MEAB Metallextraktion, V. Frolunda, Sweden (26).

[d] Normal conditions may be 1/2 maximum capacity.

[e] When not given in literature assumed to be 1/10 of holdup time per stage.

an hour (as compared to days for other equipment); (3) a smaller holdup volume, which reduces inventory cost and plant size as a whole, as well as criticality risks.

Multistage MEAB Centrifuges

Since multistage MEAB centrifugal extractors are the latest addition of such extractors for reprocessing and may offer some advantage over other centrifugal extractors, their use will be described somewhat further. For the past seven years, they have been used in 3- or 4-stage SISAK batteries (28,30). Initially these centrifugal extractors had the characteristics indicated in Table 3. More recently, however, the SISAK batteries employ a smaller version of the extractor (capacity: 0.2 m^3/h; holdup time: 0.3 s)(31). The transfer rate through these batteries is <6 s; for this purpose static mixers of the Kenics type (32) are used. The SISAK system as shown in Figs. 8 and 9 is designed for

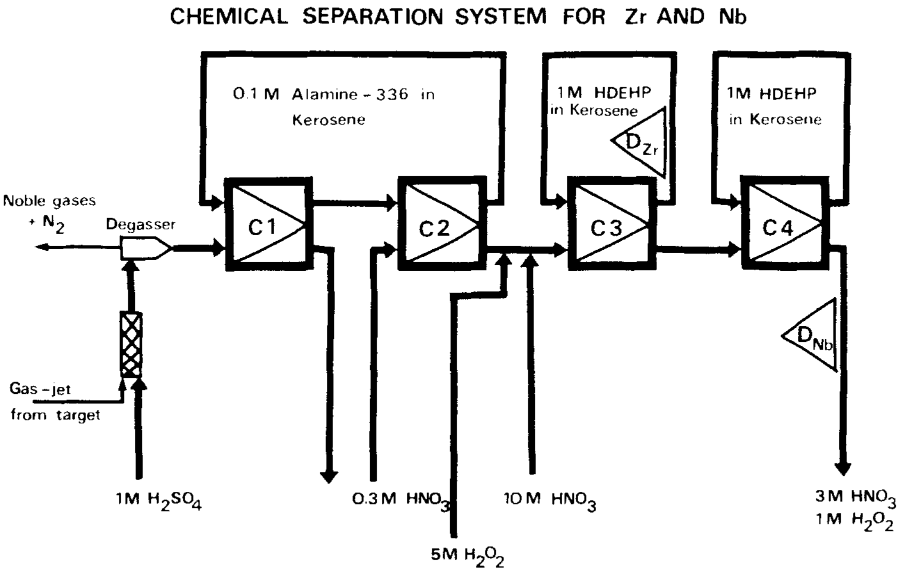

FIGURE 8. Rapid separation of short-lived zirconium and niobium isotopes in a four-stage MEAB centrifuge battery.

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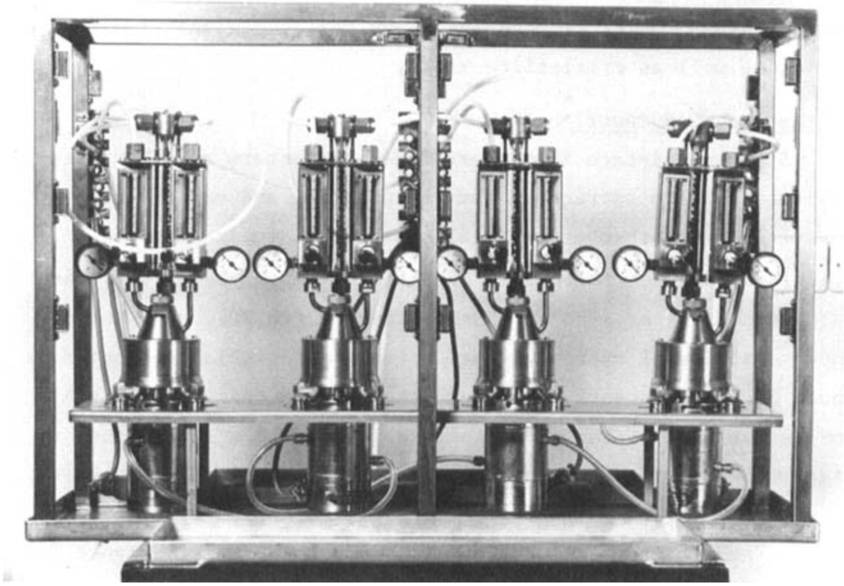

FIGURE 9. Four-stage battery of MEAB H-10 centrifugal separators and Kenics static mixers.

rapid chemical separation of zirconium and niobium isotopes from irradiated material (33,36). It uses four solvent extraction steps. The SISAK system has been used by nuclear chemists to isolate and to determine detailed decay schemes for a dozen very short-lived fission products.

CONCLUSIONS

Because the long-term hazard of the high-active waste is considered to be a serious obstacle to the peaceful use of nuclear energy, it is meaningful to attempt to reduce this hazard through the extraction of the actinides from the waste and their subsequent nuclear incineration. On the basis of the results presented in this paper, we believe that the chemical separation problem can be solved. Moreover, contrary to a recent report (9), the problem appears to

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be capable of solution within a reasonable time-frame and at a modest expenditure of effort.

The use of centrifugal contactors can reduce plant size considerably; we estimate this reduction to be about a factor of three as compared with plants utilizing mixer-settlers or pulsed columns, although plant-size reductions of as much as a factor of 10 to 20 have been estimated (33). Similarly, the costs associated with the operation of a facility using centrifugal extractors are estimated to be about a factor of five less than the operation of a facility with more conventional equipment (33).

It should also be noted that considerable experience in the use of centrifugal extractors has already been gained (35). In addition, acceptance of the concept of fractionating the actinides from the HLLW for their subsequent nuclear incineration has already been announced by France (36).

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