WASTE MANAGEMENT IN IFR FUEL CYCLE*

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ABSTRACT

The fuel cycle of the Integral Fast Reactor (IFR) has important potential
advantages for the management of high-level wastes. This sodium-cooled,
fast reactor will use metal fuels that are reprocessed by pyrochemical
methods to recover uranium, plutonium, and the minor actinides from spent
core and blanket fuel. More than 99% of all transuranic (TRU) elements
will be recovered and returned to the reactor, where they are efficiently
burned. The pyrochemical processes being developed to treat the high-level
process wastes are capable of producing waste forms with low TRU contents,
which should be easier to dispose of. However, the IFR waste forms present
new licensing issues because they will contain chloride salts and metal
alloys rather than glass or ceramic. These fuel processing and waste
treatment methods can also handle TRU-rich materials recovered from light-
water reactors and offer the possibility of efficiently and productively
consuming these fuel materials in future power reactors.

INTRODUCTION

The Integral Fast Reactor (IFR), a sodium-cooled, metal-fueled fast
reactor, is being developed by the Department of Energy as an advanced
power reactor concept (1). One of its unique features is a close-coupled
fuel cycle in which pyrochemical processes are used to recover actinides
for recycle to the reactor and to convert the high-level wastes to
disposable waste forms. Preparations are underway to demonstrate the fuel
reprocessing and waste treatment methods on a commercial scale in the
existing hot cells associated with EBR-II at the Idaho facilities of
Argonne National Laboratory.

FUEL PROCESSING

The main steps (2) in an IFR fuel recovery plant will be carried out in a
shielded cell with a purified argon atmosphere. Elements of spent core
fuel alloy (U-20 wt % Pu-10% Zr) and blanket fuel (U-10% Zr) are first
separated mechanically from reactor assembly hardware and then chopped and
loaded into steel baskets. These baskets are charged to an electrorefiner,
where the actinides are separated from the fission products. This device
has a steel crucible containing a pool of cadmium metal at 773 K overlain
with a liquid mixture of chloride salts, primarily LiCl-50 wt % KCl-6%
actinide chlorides. A commercial-scale electrorefiner would be about 1 m
in diameter and contain 1000 kg cadmium and 500 kg salt. By immersing the

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chopped fuels in the salt and making them anodic with respect to the cadmium pool, the actinides, zirconium, and noble metal fission products are electrotransported to the cadmium. The alkali metal, alkaline earth, and rare earth fission products are dissolved in the salt as chlorides. The cladding hulls are removed from the electrorefiner and become a waste.

The cadmium pool containing dissolved uranium and TRU elements is then made anodic relative to a steel electrode immersed in the salt to form a dendritic cathode deposit that is nearly pure uranium. This deposit is stripped from the cathode and melted under vacuum to remove adhering salt, and a uranium ingot is prepared for blanket fuel fabrication. Next, a ceramic crucible containing cadmium is immersed in the electrorefiner salt and made cathodic with respect to the cadmium pool. The TRU elements and the uranium remaining in the pool are collected in the cadmium cathode. They are recovered by distilling off the cadmium and used to make core fuel alloy. Small amounts of rare earths accompany the TRU elements, but the amounts are small (estimated decontamination factor is >10) and will not affect fuel performance or reactor operation.

The fission products are allowed to accumulate in the electrorefiner while processing several batches of core and blanket fuel. When the fission product heating becomes too great or the rare earth decontamination becomes too low, the actinide content of the cadmium pool is made as low as possible, and the pool is pumped through a sintered metal filter to remove the noble metal fission products that are insoluble in cadmium, namely Mo, Tc, Ru, and Rh. The filter retains about 10% of the cadmium pool in order to remove the soluble noble metals, namely Pb, Ag, Cd, In, Sn, Sb, Te, and Se. Then, a large fraction (~90%) of the actinides in the salt is transferred to the cadmium pool by adding a controlled amount of lithium metal, a strong reductant. This leaves most (~80%) of the rare earths in the salt along with all of the alkali metal, alkaline earth, and halide fission products. All of the salt is then pumped out of the electrorefiner to the salt treatment process described below.

Typical compositions and amounts of the salt and metal waste streams removed from the electrorefiner are listed in Table I. These are based on processing 200 kg of core fuel with a burnup of 100 MWd/kg and 300 kg of blanket fuel with a burnup of 25 MWd/kg. The actinide content of the metal wastes is expected to be very low, but the salt may contain up to 1% of the TRU elements fed to the electrorefiner. The alkali metal, alkaline earth, and halide fission products in the salt are about five times the amounts fed because only 20% of the salt becomes waste following each electrorefining campaign. The NaCl results from the bond sodium in the fuel elements. Similarly, the soluble noble metal content of the metal waste is ten times that fed because only 10% of the cadmium pool is removed. The zirconium in the waste corresponds to that which is soluble in the pool, with the balance being recovered in a separate step.

The cladding hulls are another major waste stream from the electrorefiner, amounting to about 300 kg of stainless steel for the 500 kg of fuel processed. The salt adhering to the hulls will contain about 1 kg of actinides. This salt will be removed by rinsing the hulls with clean salt or vaporizing the actinide-laden salt. The hulls are expected to have a sufficiently low TRU content that if they are melted with the assembly hardware, the steel ingot will be a nonTRU waste.

WASTE TREATMENT

The proposed processes for recovering TRU elements from the salt and metal discharged from the electrorefiner and providing suitable materials for final waste form production are shown in Fig. 1. The electrorefiner salt is first contacted with a molten Cd-U alloy in several countercurrent stages. The uranium in the metal solution exchanges with TRU and rare earth elements in the salt by means of exchange reactions such as

$$U_{Cd} + PuCl_3 = UCl_3 + Pu_{Cd}, \text{ and } U_{Cd} + CeCl_3 = UCl_3 + Ce_{Cd}.$$
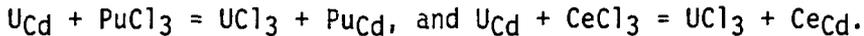
The equilibrium concentrations in the salt and metal phases for all trivalent actinides and rare earths must satisfy the following relationship, which is derived from the equilibrium constant for the exchange reactions,

$$D_U = \alpha_{Pu}D_{Pu} = \alpha_{Am}D_{Am} = \alpha_{Ce}D_{Ce} = \alpha_{Nd}D_{Nd} = \cdots$$

where D = distribution coefficient = y/x,
  y = mol fraction in salt, and
  x = atom fraction in metal.

The term, α, is the ratio of salt- and metal-phase activity coefficients and the standard free energy change of the exchange reaction. Its reciprocal can be thought of as a separation factor (SF), for example, $1/\alpha_{Pu} = SF_{Pu} = D_{Pu}/D_U$. The separation factors for the actinides and several important rare earths have been measured (3,4,5), and the values relative to uranium are listed in Table II. The small values for the actinides and the large values for the rare earths show that actinides are difficult to separate from each other, but they can be easily separated from all rare earths. The separation factors for samarium and europium are very large, >1000, because they form divalent chlorides in this system (4). The factor for yttrium is also large, apparently because the thermodynamic activity of YCl_3 in the salt is very small (5). Although not included in Table II, the separation factors for the alkali metals and alkaline earths are also very large, >10,000. Thus, fission product cesium and strontium will remain completely in the salt during this extraction.

Extraction calculations using these separation factors have shown that adequate TRU recoveries and rare earth separations can be achieved in four to seven equilibrium stages with roughly equal volumes of the metal and salt phases. Some results are shown in Fig. 2, where the fractions of plutonium and rare earths remaining in the product salt are plotted as functions of the metal/salt weight ratio and the number of equilibrium stages. The separation factor of all rare earths relative to uranium was conservatively assumed to be 40. For six stages and metal/salt mass ratio of 5, the plutonium recovery will be about 99.99%, and less than 30% of the rare earths will be carried with the TRU-rich product back to the electrorefiner. The percentages of neptunium, americium, and curium recovered are about 99.99, 99.8, and 99.3%, respectively, for this example. This extraction process results in an amount of uranium transferred into the salt that is equivalent to the amounts of TRU and rare earth elements transferred to the metal. This uranium is eventually lost, but this is a small penalty in the IFR fuel cycle because the uranium is depleted.

As shown in Fig. 1, the product metal from the extractor is fed to a metal retort where the cadmium is vaporized and then recycled. The TRU-rich residue is returned to the electrorefiner.

The product salt is fed to a stirred vessel, the salt stripper, where the salt is contacted with a Cd-Li alloy. The lithium reduces essentially all of the uranium and residual TRU elements from the salt, most of the rare earths, but none of the alkali metals and alkaline earths. About 80% of the stripped salt is returned to the electrorefiner, and the rest becomes a waste in order to remove fission product strontium, cesium, and iodine from the process. An estimated typical composition of this treated salt is shown in Table III at the time of discharge (1.5 y) and after various cooling times out to 10,000 y. After about 1000 y, the only fission product activities remaining are Cs-135 and I-129. The actinide content is expected to be very low, resulting in an alpha activity of less than 10 nCi/g, mostly from Am-241. In the laboratory, salts with total actinide contents below 0.1 ppm have been produced by this stripping procedure followed by filtration through a porous metal filter.

The liquid cadmium solution from salt stripping, which contains uranium, rare earths, and trace amounts of TRU elements, is fed to a metal retort along with the filter element containing the zirconium noble metals, and cadmium from the electrorefiner. After the excess cadmium is distilled off, a matrix alloy, for example copper, is added to the metal waste. Thus, the residue is a copper alloy containing the rare earth and noble metal fission products, zirconium, uranium, cadmium, and small amounts of TRU elements. An estimated composition for the metal retort residue without the copper matrix alloy is listed in Table IV with activities from the time of discharge (1.5 y) to 10,000 y. Several long-lived fission products will be dissolved as the metals in the alloy.

IMMOBILIZATION OF HIGH-LEVEL WASTES

The treated salt and metal must be converted to suitable waste forms and sealed in corrosion-resistant containers so that they can be placed in a geologic repository for high-level wastes. The reference form for the metal wastes will be prepared by alloying the fission products, cadmium, and uranium with a corrosion-resistant metal, such as copper, in the waste metal retort. The retorted residue will be densified by compaction or melting and sealed in a steel container.

Several methods for immobilizing and containing the treated salt have been considered. Among these are conversion to glass, salt electrolysis, and dispersion in portland cement. None of these techniques was found to be attractive in initial evaluations. Current efforts are directed toward the sorption of the salt on dry ceramic materials. It seems likely that these materials can be placed in steel containers because the dry salt does not attack iron alloys.

OTHER FISSION PRODUCT WASTES

An IFR fuel cycle facility should have low overall releases of radio-activity, because it should be capable of containing all fission products, including those that are difficult to manage in conventional fuel recovery

processes. The fission gases (Xe, Kr and T) are released into the process cell when the fuel is chopped and then dissolved in the electrorefiner. The process cell atmosphere, which is argon-5% nitrogen, is continuously purified to maintain oxygen and water levels below 100 ppm. The purification system converts tritium to water and traps the water on molecular sieves. Because the volume of this water is small (about 300 L/y for a plant treating 6300 kg/y of actinides with an average burnup of 60 MWd/kg), it can be stored until the tritium has decayed sufficiently to be released. Krypton and xenon can be recovered from the cell gas and concentrated by conventional cryogenic distillation methods. Extensive gas precleaning and drying systems are not required because the feed gas is argon-5% nitrogen with less than 100 ppm total of condensable impurities such as H_2O, O_2, O_3, and CO_2. Iodine and C-14 are not expected to be volatilized in the pyroprocess. Iodine will be contained in the salt as stable iodides, e.g., KI, and carbon will form stable carbides, e.g., ZrC, that should be retained in the metal wastes.

STATUS

The chemical feasibility of the electrorefining process has been established in laboratory experiments with uranium, plutonium, and inactive fission product elements. A prototype electrorefiner that approaches commercial scale has been operating successfully with uranium for three years, demonstrating the engineering feasibility of this key process. A commercial-scale (20 kg actinides per batch) electrorefiner is being designed and fabricated for the planned fuel cycle demonstration in the EBR-II facilities. This equipment is expected to be in operation with irradiated fuel within a year (6).

The chemistry of the key waste salt treatment steps, extraction and stripping, has been sufficiently well established that engineering-scale process equipment is being assembled for testing within the next few months. The salt extractor concept being pursued is similar to centrifugal contactors developed for aqueous-organic extractions (7). The salt stripper is a stirred vessel with the capability for charging and discharging molten salt and cadmium. This apparatus and a metal retort, which is in the initial stages of development, will be tested to obtain information for the equipment design in the next phase of the IFR fuel cycle demonstration.

CONCLUSIONS

The IFR and its fuel cycle have several features that seem to be advantageous for managing high-level waste An important feature is the recovery of more than 99% of all TRU elements by the electrorefiner and the salt extractor followed by the efficient burning of the TRU elements by the reactor. Heavy, non-fissionable isotopes of the minor actinides (Np, Am, and Cm) do not build up in the fuel cycle because of the hard neutron flux of the metal-fueled fast reactor. Thus, the process wastes should have low alpha activities, which should make their disposal easier.

Because all TRU elements can be burned efficiently, these elements can be recovered from light-water reactor fuels and then consumed in IFRs. Some of the aqueous and pyrochemical processes proposed for treating LWR fuels,

produce the TRU elements mixed with the rare earth fission products. Such material can be introduced into an IFR fuel process and the rare earths separated to produce a satisfactory fuel material.

The release of certain radionuclides from conventional reprocessing plants (or the difficulty and cost of their containment) was one reason for the decision in this country to forego fuel reprocessing and bury spent fuel directly. An IFR fuel reprocessing plant should have lower radioactive releases than conventional plants. The troublesome fission products (Kr, T, I, and C-14), can be well contained, and thus, the plant should be more acceptable.

However, the principal IFR high-level wastes, chloride salts and metals, are different from the glass and ceramic waste forms presently being considered, and as such will present unique problems in the licensing process. Despite their low alpha activities, both the salt and metal will require long-term confinement because of their long-lived fission product contents. Despite its very low alpha activity, the treated salt, which contains Cs-135 and I-129, presents particularly difficult licensing problems because it is water soluble. A waste form and engineering barrier system that adequately contains the cesium and iodine must be developed. The metal wastes will contain Zr-93, Tc-99, Pd-107, Sn-126, and small but significant amounts of TRU elements. Compared to the containment of these fission products in glass in an oxidized form, they may be better contained as metals dissolved in a corrosion-resistant alloy.

There is much work still to be done to develop a practical and economical IFR fuel cycle. Reprocessing spent fuel from EBR-II and treating the wastes will demonstrate process feasibility and will provide information for the design of improved process equipment. Converting these wastes to suitable waste forms and showing that these waste forms can be licensed for geologic disposal will also be undertaken. These efforts are worthwhile because the IFR concept has the potential to use our nuclear resources more effectively and produce wastes with lower long-term risks.

REFERENCES

1. Y. I. Chang, "The Integral Fast Rector," Nuclear Technology 88, 129-138 (1989).

2. M. J. Steindler et al., "Chemical Technology Division Annual Technical Report 1987," Argonne National Laboratory Report ANL-88-19 (May 1988), pp 92-98.

3. M. J. Steindler et al., "Chemical Technology Division Annual Technical Report 1989," Argonne National Laboratory Report ANL-90/11 (March 1989), pp 105-106.

4. J. P. Ackerman and J. L. Settle, to be published in J. of the Less Common Metals.

5. H. Miyashiro et al., "Development of Pyrometallurgical Partitioning," First OECD/NEA Information Exchange Meeting on Separation and Transmutation of Actinides and Fission Products, November 6-8, 1990, Mito, Japan.

6. M. J. Lineberry and R. D. Phipps, "Preparations for the IFR Fuel Cycle Demonstration," Trans. Am. Nucl. Soc. 60, 170 (1989).

7. R. A. Leonard, G. J. Bernstein, A. A. Ziegler, and R. H. Pelto, "Annular Centrifugal Contactors for Solvent Extraction," Separation Sci. Technol. 15, 925-943 (1980).

DISCLAIMER

TABLE I.
Salt and Metal Discharged from Electrorefiner

(from processing 500 kg of actinides
with total energy output of 75 MWy.)

Salt	Weight of Chloride, kg	Metal	Weight of Metal, kg
LiCl-KCl-NaCl	440	Cd	100
RbCl-CsCl	25	Zr	1
$SrCl_2$-$BaCl_2$	14	Noble Metals	11
(Rare Earth) Cl_3	17		
UCl_3	0.7	U	~0
(TRU) Cl_3	3.3	TRU	~0
	500		111

TABLE II.
Separation Factors of Actinides
and Rare Earths Relative to Uranium

Salt: LiCl-KCl
Metal: Cd
Temperature: 773 K

U	1.0	La	60
Np	2.0	Ce	48
Pu	1.9	Nd	43
Am	3.1	Sm	>1000
Cm	3.8	Eu	>1000
		Y	>1000

TABLE III
Estimated Composition of Treated Waste Salt

(500 kg actinides processed)

Component	Weight of Salt, kg	Cooling =	Activity, Ci				
			1.5 y	10 y	100 y	1000 y	10,000 y
LiCl-KCl-NaCl	90		0	0	0	0	0
RbCl-CsCl	4.9		$2.1 \cdot 10^5$	$1.6 \cdot 10^5$	$2.0 \cdot 10^4$	1.6	1.6
$SrCl_2$-$BaCl_2$	2.8		$7.3 \cdot 10^4$	$5.9 \cdot 10^4$	$6.4 \cdot 10^3$	$1.5 \cdot 10^{-6}$	0
YCl_3	0.5		$2.0 \cdot 10^3$	0	0	0	0
$SmCl_2$	1.3		$3.9 \cdot 10^3$	$3.7 \cdot 10^3$	$1.9 \cdot 10^3$	2.3	0
EuCl-2	0.2		$1.4 \cdot 10^4$	$1.4 \cdot 10^3$	$2.9 \cdot 10^1$	0	0
I-129	0		$4.5 \cdot 10^{-2}$	$4.5 \cdot 10^{-2}$	$4.5 \cdot 10^{-2}$	$4.5 \cdot 10^{-2}$	$4.5 \cdot 10^{-2}$
Total β-γ Activity			$3.0 \cdot 10^5$	$2.2 \cdot 10^5$	$2.8 \cdot 10^4$	3.9	1.6
Total α Activity			$\sim 2 \cdot 10^{-2}$	$\sim 3 \cdot 10^{-3}$	$\sim 2 \cdot 10^{-4}$	$\sim 4 \cdot 10^{-5}$	$\sim 3 \cdot 10^{-6}$
Decay Power, W			940	610	70	<1	<1

TABLE IV
Estimated Composition of Treated Waste Metal

(500 kg actinides processed)

Component	Weight of Metal, kg	Cooling =	Activity, Ci				
			1.5 y	10 y	100 y	1000 y	10,000 y
Cadmium	35		0	0	0	0	0
Zirconium	3		$1.0 \cdot 10^4$	2.3	2.3	2.3	2.3
Short-Lived Noble Metals	11		$6.3 \cdot 10^5$	$3.7 \cdot 10^3$	$<2 \cdot 10^{-7}$	0	0
Rare Earths	8		$5.6 \cdot 10^5$	$1.6 \cdot 10^4$	$<3 \cdot 10^{-7}$	0	0
Se-79	0		0.30	0.30	0.30	0.30	0.27
Tc-99	0		14	14	14	14	14
Pd-107	0		0.2	0.2	0.2	0.2	0.2
Sn-126	0		1.3	1.3	1.3	1.3	1.3
Total β-γ Activity			$1.2 \cdot 10^6$	$2.0 \cdot 10^4$	18.1	18.1	18.0
U - α Activity	7		$8 \cdot 10^{-3}$	$8 \cdot 10^{-3}$	$8 \cdot 10^{-3}$	$8 \cdot 10^{-3}$	$8 \cdot 10^{-3}$
TRU - α Activity	~0.006		~20	~5	~3	~1	0.3
Decay Power, W			4800	20	<1	<1	<1

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SALT STRIPPING

SALT CONTAINMENT

WASTE SALT

Alkali Metals – Na, Rb, Cs

Alkaline Earths – Sr, Ba

Rare Earths – Y, La, Ce, ...

Halides – Br, I

Actinides – U, TRU (<1%)
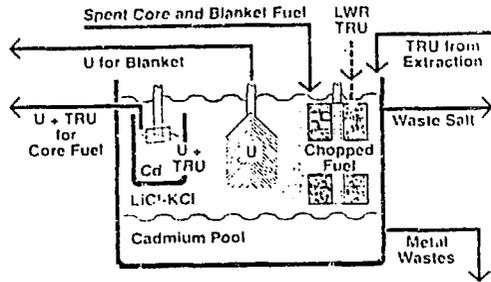
ELECTROREFINER 500°C

EXTRACTION

BENEFITS OF IFR CYCLE FOR HLW DISPOSAL

- Burns all actinides
- TRU in repository reduced >99%
- Separation of heat-producing elements for possible long-term storage and use
- Captures all fission products, including Kr, T, I, and C-14
- Long-lived Tc, Zr, Pd, and Sn in metal alloy

→ METAL WASTES →

Cadmium

Cladding Hulls

Zr

Noble Metals – Mo, Tc, Ru, ...
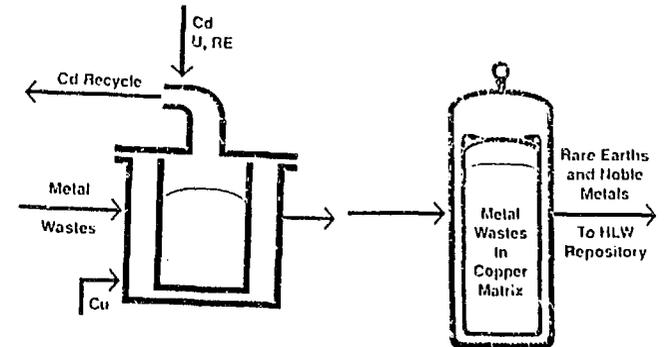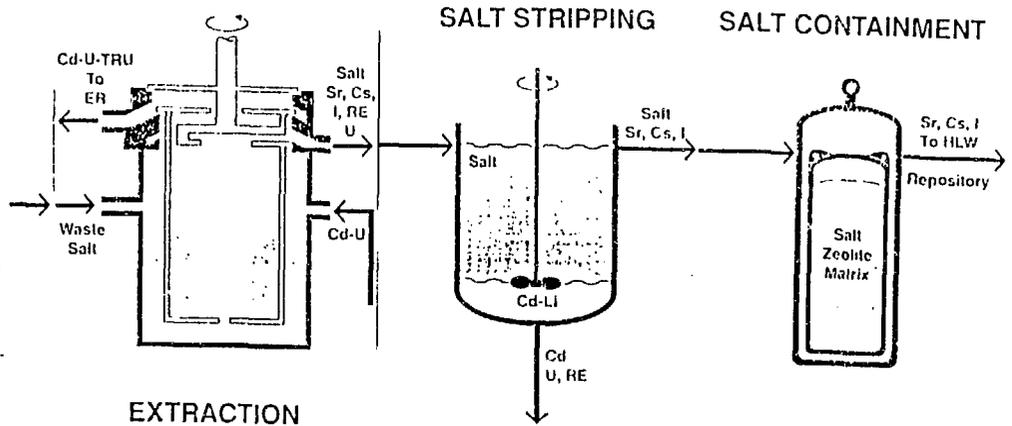
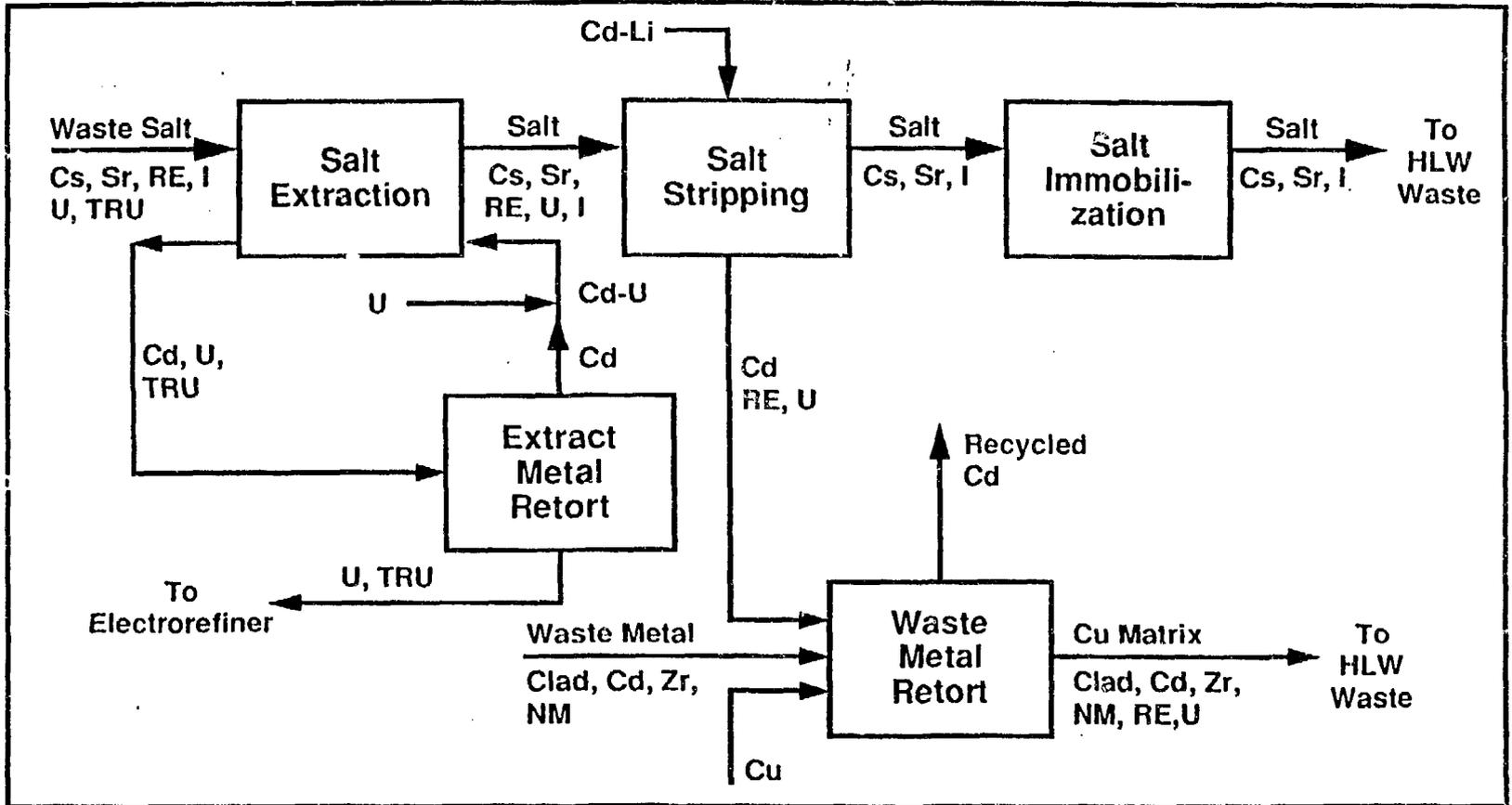**METAL WASTE RETORT** **METAL CONTAINMENT**

Fig. 1. Treatment of salt and metal discharged from electrorefiner (RE = rare earth, TRU = transuranics, NM = noble metal, HLW = high-level waste)
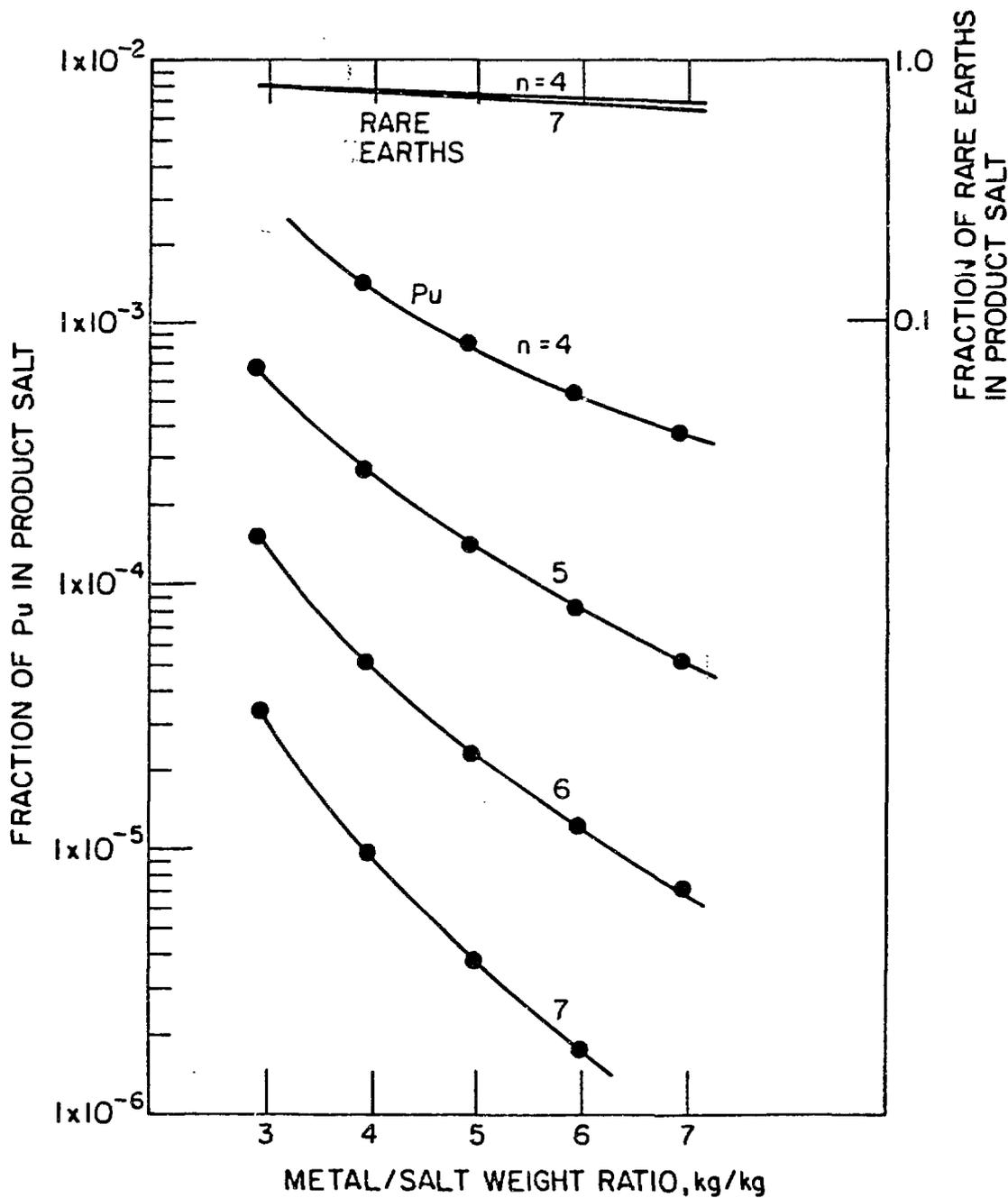
Fig. 2. Separation of plutonium from rare earths by salt-metal extraction
(n = number of equilibrium stages)