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# Studies of a Liquid Anode for Plutonium Electrorefining*

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## ABSTRACT

We are developing a solvent anode as an alternate method for producing plutonium metal of high purity by an electrorefining process. Our goals are to produce metal of 99.98% purity with an anode residue containing less than 2% of the plutonium in the feed material. If we are successful, we will design and demonstrate a system utilizing semi-continuous and remotely controlled operations.

Establishing a solvent anode method should lead to improved yields and a substantial reduction in the amount of residues generated by the electrorefining process. The new method should be a viable pyrochemical technique for recovering both plutonium and uranium from spent reactor fuel.

Initially, the anode consists of a tantalum rod immersed in a pool of liquid cadmium at 740°C. Impure plutonium or a solid anode residue is in contact with the cadmium. As current passes through the anode, plutonium in the cadmium is oxidized and transfers into the molten salt as tripositive plutonium. More plutonium dissolves into the cadmium and the oxidation continues. The tripositive plutonium is carried through the salt to the cathode, where it is reduced to pure, liquid metal. This metal, which is heavier than the salt, drips from the cathode into an annulus between the anode and cathode compartments and forms a product ring.

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We are studying parameters such as stirring, salt composition, temperature and, in addition, the compatibility of candidate container materials with the reagents and products at elevated temperatures.

## INTRODUCTION

Pyrochemical processes are defined as nonaqueous processes which are conducted at elevated temperatures. Since the early forties, pyrochemical operations have been used to reduce uranium and plutonium tetrafluorides to metal.[1,2] Baker's successes with the bomb reductions led Los Alamos to study pyrochemical processes for processing spent reactor fuels. For the metal-fueled fast reactors, the fuels could be purified without altering their metallic state. Pyrochemical research such as plutonium electrorefining, molten salt extraction and pyroredox processing was initiated at Los Alamos as part of the study for the Los Alamos Molten Plutonium Reactor Experiment.[3]

The pyrochemical recovery of plutonium oxide and scrap at Los Alamos is an integrated process. Feed material is either $PuO_2$ or impure plutonium metal. The oxide is converted to metal by either direct oxide reduction[4] or reduction by the bomb method.[2] If the americium concentration is greater than 1,000 ppm., a molten salt extraction step lowers the americium value to a level satisfactory for producing high purity plutonium by electrorefining.[5] The impure plutonium is then melted and chill-cast into a right cylinder for use as an anode. High-purity plutonium is produced by an electrorefining process utilizing molten salts and liquid metals.[6]

## DESCRIPTION OF PLUTONIUM ELECTROREFINING AT LOS ALAMOS

In our electrorefining cell impure plutonium metal is converted to metal that is purer than metal produced by chemical reduction of plutonium compounds. The process employs tungsten electrodes, ceramic containers, molten salts and liquid metals. Impure metal is oxidized at the anode and transported through the eutectic salt to the cathode. The metal ions are reduced at the cathode to form pure metal. Under the operating conditions at Los Alamos, the product metal is of greater than 99.99 w/o (percent by weight) purity.

A schematic of the Los Alamos Electrorefining Cell is shown in Fig. 1.[6] The cell consists of two magnesia crucibles joined at the base. In a typical run, 4.5-6.0 kg of impure plutonium is placed in the inner crucible and an equimolar mixture of NaCl-KCl that is 10 w/o $MgCl_2$ is added to the system. The cell is assembled, evacuated and back-filled with argon. The crucibles are heated in an argon atmosphere to 740°C and the stirrer, anode and cathode are lowered into position. Both the salt phase and anode are stirred during electrolysis. The electrorefining run is continued until terminated by the back e.m.f. sampling unit. An operator withdraws the anode, cathode and stirrer from the melt and heating is discontinued. After

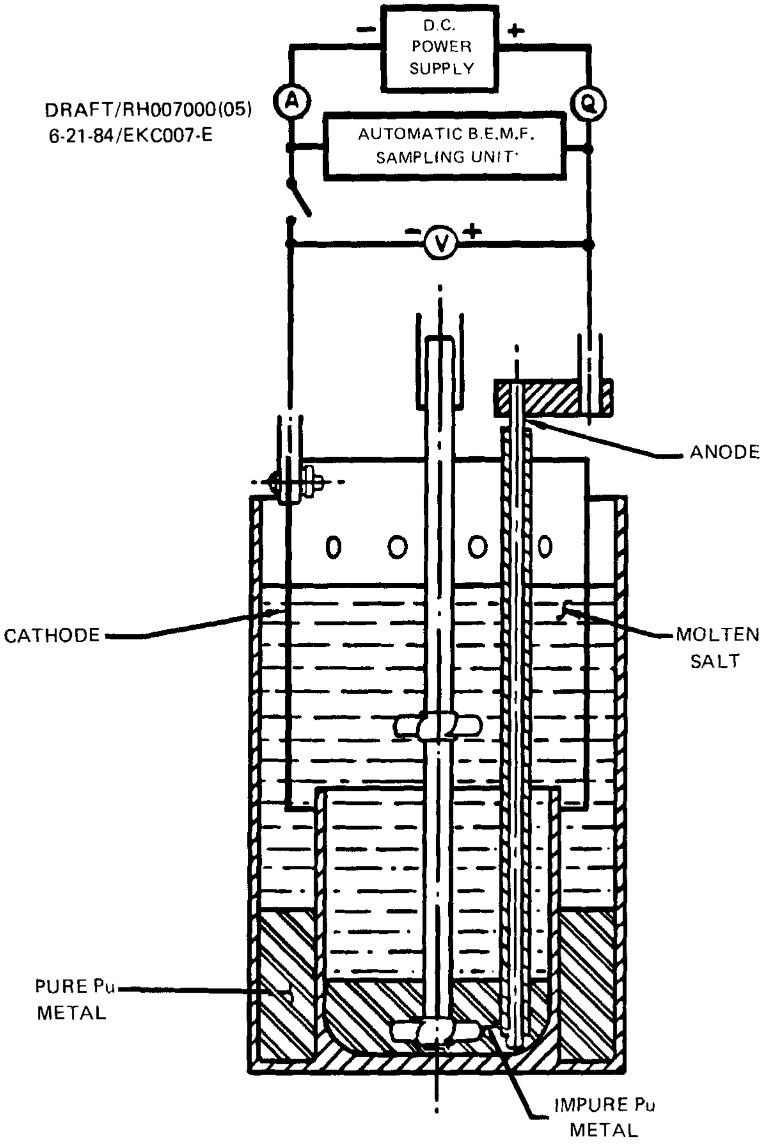

Fig. 1 Los Alamos electrorefining cell.

cooling, the small amount of pyrophoric metal that has condensed on the furnace lid is burned to oxide. The furnace tube is then unloaded and the metal product ring is separated from the crucible and salt.

Three waste streams (an anode heel, the salt and crucible fragments) are produced. Plutonium in the anode heel is recovered by aqueous or pyrochemical methods.[7,8] Calcium is added to the salt, which contains metal beads and plutonium trichloride, to recover the plutonium.[9] Finally, plutonium is recovered from the ceramic crucible fragments by a salt wash.[10]

The purity of the plutonium product for three typical feeds is given in Table 1. Although the product contains less than 100 ppm of detectable metallic impurities, except for tungsten, which is introduced from the electrodes. The product densities are greater than 19.6 g/cm$^3$, which is excellent.

The product yields, where yield is defined as the quantity of plutonium collected in the anode ring multiplied by 100 and divided by the total plutonium in the cast anode ingot, vary from 83% for a Pu·1 w/o Ga alloy to 90% for alpha-Pu on a once-through basis. In the gallium alloy, 10% of the initial plutonium remained in the anode as a heel and 7% in the eutectic salt as uncoalesced metal and tripositive plutonium.

Even though large quantities of pure plutonium have been recovered by the electrorefining process, a number of modifications appear feasible. Almost 12% of the production runs have been terminated before completion, usually because of stirrer failure.[6] Usually, the furnace tube was opened, the anode cathode and stirrer removed, and the metal recast and rerun. A metal stirrer designed to efficiently mix metals of less density would virtually eliminate breakage and increase yields through improved mixing. Replacement of the magnesia crucible with a reusable container would cut crucible costs and reduce the volume of ceramic residues. Recycling the salt eutectic would reduce salt stripping operations and residue volumes. A semi-continuous operation with remotely controlled steps would increase output with reduced exposure of personnel to radiation. Finally, and the topic for this paper, a solvent anode would reduce the amount of plutonium in the anode residue and provide an efficient recovery of the spent anode.

## THEORETICAL CONSIDERATIONS

The behavior of impurity elements in electrorefining, and the free energies at 1000°K, are summarized in Table 2. Because plutonium is a very electropositive metal, most impurities are less reactive and remain in the anode during electrorefining. More electropositive elements such as americum and cerium, concentrate in the electrolyte.

TABLE 1

PURIFICATION OF PLUTONIUM, 10 w/o $PuCl_3$-50.4 w/o KCl-39.6 w/o NaCl ELECTROLYTE

Concentration of Element[b]

| Element[a] | Bomb-Reduced Metal Feed | Bomb-Reduced Metal Product[c] | Pu-Fe Alloy Feed | Pu-Fe Alloy Product[c] | Pu-Ga Alloy Feed | Pu-Ga Alloy Product[c] |
|---|---|---|---|---|---|---|
| Al | 75 | < 5 | 15 | < 5 | 75 | < 5 |
| Mg | 75 | < 5 | 90 | 5 | 75 | < 5 |
| Si | 50 | <10 | 20 | <10 | 50 | <10 |
| Cu | 10 | < 2 | < 2 | < 2 | 50 | < 2 |
| Mn | 7 | < 2 | 7 | < 2 | 7 | < 2 |
| Cr | 40 | <20 | <20 | <10 | 40 | <20 |
| Ni | 110 | <20 | <20 | <10 | 110 | <20 |
| U | 30 | <30 | 30 | <30 | 90 | <30 |
| Th | 124 | <15 | <15 | <15 | 124 | <15 |
| W | <10 | 30 | <10 | 40 | 410 | 28 |
| Am | 143 | 20 | 128 | 24 | 143 | 20 |
| Fe | 280 | 30 | $2.5 \times 10^4$ | 20 | 410 | <20 |
| Ga | <25 | <25 | <25 | <25 | $1.02 \times 10^4$ | <25 |

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[a] Elements not detected in feeds or products include Na, Li, Be, La, Bi, B, Co, Cl, Y, Ca, Pb, Sn, Zn, and Ta.

[b] Parts of impurity per million parts of plutonium, weight basis.

[c] Product density was >19.6 g./cc.

The magnesium chloride initially added to the cell reacts with molten plutonium prior to electrorefining:

$$3/2\ \mathrm{Mg}^{+2} + \mathrm{Pu}^{\circ}(l) \rightarrow \mathrm{Pu}^{+3} + 3/2\ \mathrm{Mg}^{\circ}\ (l). \qquad (1)$$

Since all components are liquids stirred at relatively high temperatures, reactions should occur under near equilibrium conditions. The concentration of an impurity in the product , salt and anode compartments can be expressed by equations such as

$$(\mathrm{Am}^{\circ})\ p = 1/K\ \frac{(\mathrm{Am}^{+3})s}{(\mathrm{Pu}^{+3})s}\ .\ (\mathrm{Pu}^{\circ})p, \qquad (2)$$

where $(\mathrm{Am}^{\circ})p$ and $(\mathrm{Pu}^{\circ})p$ are the concentrations in the product, $(\mathrm{Am}^{+3})s$ and $(\mathrm{Pu}^{+3})s$ are the concentrations in the salt, and $K_1$ is the equilibrium constant that includes corrections for differences between activities and concentrations. If the amount of americium

TABLE 2

BEHAVIOR OF IMPURITY ELEMENTS IN ELECTROREFINING

| Element | -ΔF°, kcal./g. atom Cl | Concentration, p.p.m. Feed | Concentration, p.p.m. Product | Element Concentrated in |
|---|---|---|---|---|
| Ni | 18 | 750 | <10 | Anode |
| Cu | 21 | 100 | 2 | Anode |
| Ta | 22 | 5,000 | 80 | Anode |
| Fe | 27 | 25,000 | 20 | Anode |
| Cr | 32 | 280 | <10 | Anode |
| Ca | 32 | 10,000 | <25 | Anode |
| Mn | 41 | 70 | < 2 | Anode |
| Al | 46 | 2,700 | < 5 | Anode |
| U | 54 | 275 | <20 | Anode |
| Th | 59 | 160 | <10 | Anode |
| Pu | 59 | - | - | - |
| Ce | 66 | 527 | <25 | Salt |
| Am | 67 | 850 | 82 | Salt |

---

[a] -ΔF° at 1000°K is tabulated. For elements having a multiplicity of oxidation states, the -ΔF° for the most stable form of the chloride is given.

in the feed material or in the salt is increased, Eq 2 indicates that the americium content of the product also increases. We have empirically determined that the ratio of americium in our feed material to that in the product under our operating conditions is nine; that is,

$$\frac{(\mathrm{Am}^\circ)f}{(\mathrm{Am}^\circ)p} \sim 9, \tag{3}$$

where $(\mathrm{Am}^\circ)f$ is the initial concentration of americium in the feed material. Therefore, to ensure a low americium value in the product, its initial concentration must be low . Other lanthanides and actinides will behave similarly. Although changes may occur if a solvent anode pool is used, the general behavior is not expected to change. For example, addition of a solvent anode metal such as cadmium should not significantly change the ratio of the activities of tripositive americium in the salt phase to the americium metal in the product.

The free energies of most of the impurities are such that the elements will remain in the anode. For example, for iron, the free energy for the reaction:

$$3Fe^\circ(1) + 2Pu^{+3} \rightarrow 3Fe^{+2} + 2Pu^\circ\ (1) \qquad (4)$$

is -96 kcal at 1000°C and the calculated equilibrium constant is $10^{-21}$. Therefore,

$$(Fe^\circ)^3 p = 1/K_2 \ \frac{(Fe^{+2})s^3}{(Pu^{+3})s^2} \cdot (Pu^\circ)p^2 \qquad (5)$$

and (Fe°)p would be very small. Other less electropositive elements behave similarly, and addition of a solvent pool element in the anode should not affect their relative distributions.

The composition of salt in the cell remains constant as long as the potential is kept below the decomposition potential of the eutectic. The composition of the anode changes continuously as the process depletes plutonium. Initially, plutonium is oxidized and transported through the electrolyte to the cathode. The concentrations of the impurities in the anode will increase uniformly as the electrorefining proceeds.

In most of our processing, the plutonium has been alloyed to a composition of 1 w/o gallium to reduce the density. As shown in the Pu-Ga phase diagram shown in Fig. 2, the Pu-Ga mixture is initially a liquid at an electrorefining temperature of 750°C. As the process continues, a solid phase forms at 16.8 a/o (atom percent) gallium. As long as the gallium concentration is below 25 a/o, liquid is present and with efficient stirring plutonium can be oxidized under the cell operating conditions. As the liquid phase decreases, effective stirring becomes more difficult and anode polarization occurs. The diffusion of plutonium in the solid anode becomes much too slow to meet the high current density conditions at the anode and competing reactions will predominate. At this stage, the reaction of interest is

$$Pu^\circ\ (\text{solid anode}) \rightarrow Pu^\circ\ (\text{liquid anode}) \qquad (6)$$

which corresponds to a potential

$$E = 0.0291\ \ln\ (Pu)\ \text{solid anode surface.} \qquad (7)$$

Continued operation after the potential has risen will begin to transfer impurities into the salt and as a result, into the product.

If a solvent anode is employed, the plutonium continues to diffuse to the anode surface. Solid formation no longer will limit

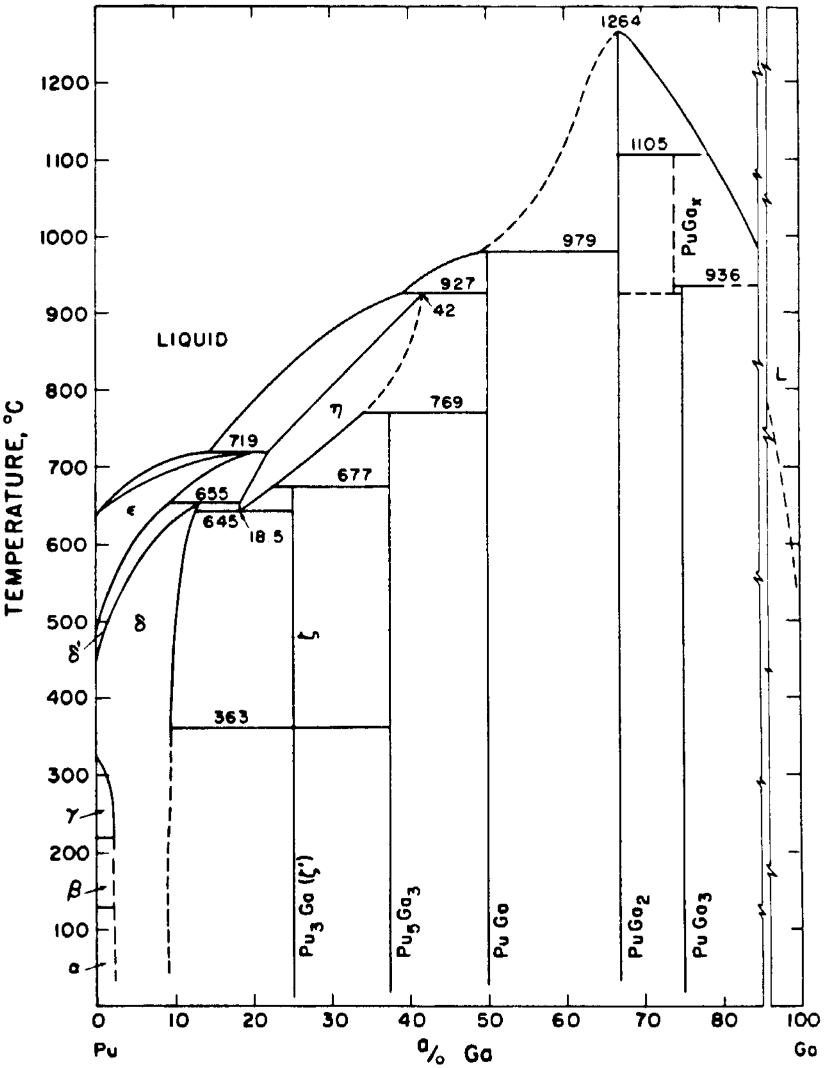

Fig. 2 Pu-Ga phase diagram.

the oxidation of plutonium. However, the solvent element may be oxidized and, with insufficient plutonium available, may be transferred. Tests of transfer conditions will be needed.

In the present process, the transfer of impurities into the salt is monitored by periodically checking the cell potential. At frequent intervals the current is interrupted and the voltage across the electrodes is measured. When the back-EMF reaches a pre-set value, the current is automatically turned off. The electrodes, stirrer and thermocouple well are raised out of the cell and the system is cooled.

Under our operating conditions, which include stirring and current density less than 0.6 amps per square centimeter, the impurity concentrations in the product are below normal spectroscopic detection limits (except for lanthanides, actinides and tungsten, which is introduced from the electrodes.)

The anode residue from our present electrorefining is a solid containing gallium, impurities initially present in the anode feed and 10% of the plutonium feed. The treatment of this residue is discussed in the next section of this report. Approximately 7% of the plutonium feed remains in the eutectic salt, along with those impurities more electropositive than plutonium. We collect these salts and treat them with calcium at high temperature to recover the plutonium. This metal is then recycled through the integrated process.

## THE SOLVENT ANODE EXPERIMENTS

The plutonium in the anode residue is currently recovered by aqueous procedures or a pyroredox process developed at Los Alamos.[8] The two major steps in pyroredox are pyrochemical oxidation of plutonium with zinc chloride and subsequent reduction of the plutonium trichloride with calcium metal. The plutonium is then recycled through our integrated cycle.

Other methods proposed for recovering plutonium by pyrochemical means include electrorefining the spent anode as a solid, raising the temperature until the anode is liquid, or adding a liquid solvent in which the anode is partially soluble.[11]

Our survey experiments with cadmium as a liquid solvent have been promising. At 750°C, as shown in the Cd-Pu phase diagram (Fig.3),[12] a liquid phase containing some plutonium is present at all high cadmium concentrations. The reactions in the anode chamber in such systems would be:

$$\mathrm{Pu,\ Cd\ (l)} \rightarrow \mathrm{Pu}^{+3} + 3e^-, \text{ or} \tag{8}$$

$$\mathrm{Pu,\ Cd\ (l)} \rightarrow 3/2\ \mathrm{Cd}^{+2} + 3e^- \text{ and} \tag{9}$$

$$3/2\ \mathrm{Cd}^{+2} + \mathrm{Pu,\ Cd\ (l)} \rightarrow \mathrm{Pu}^{+3} + 3/2\ \mathrm{Cd}^{\circ} \tag{10}$$

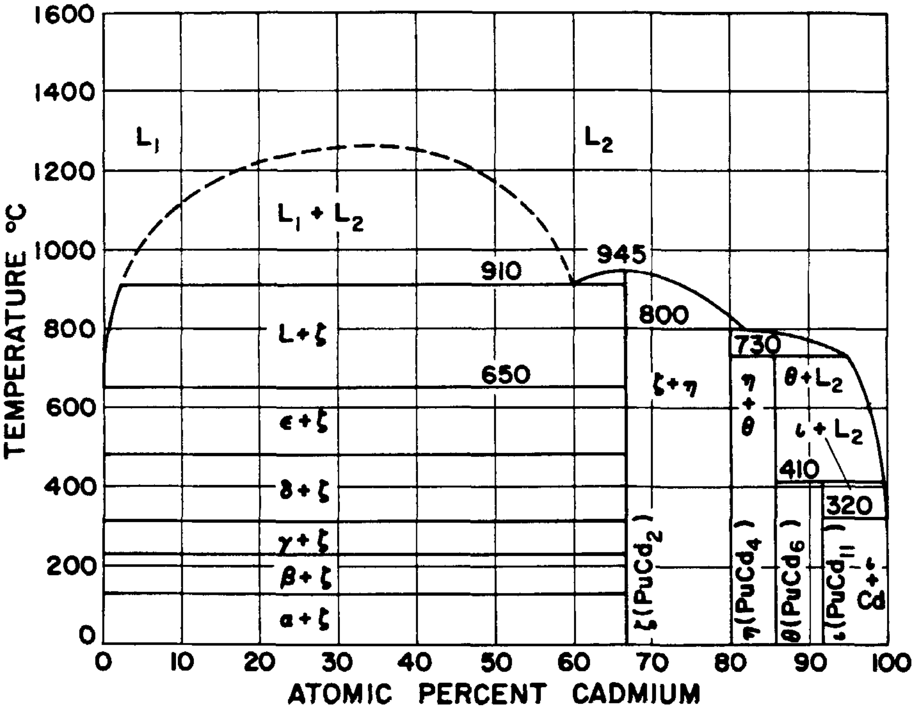

Fig. 3 Plutonium-cadmium phase diagram.

As plutonium is oxidized from the liquid, additional plutonium dissolves. At the cathode, tripositive plutonium is reduced to pure metal; that is,

$$Pu^{+3} + 3e^{-} \rightarrow Pu^{\circ}\ (1) \qquad (11)$$

This metal collects at the bottom of the cathode chamber to form an annular ring. When the process is completed, the cadmium in the final anode residue could be recovered by vacuum distillation. The plutonium in the salt phase, as in our present process, is recovered by salt stripping.

In survey experiments employing a 500g electrorefining cell, high purity plutonium and cadmium metal were placed in the anode cup. In this system, the temperature of the melt was estimated from the temperature of the furnace tube. In one experiment, the cell temperature did not remain at 750°C throughout the experiment and dendritic plutonium deposited on the cathode. This caused a low yield. How-

TABLE 3

Small Scale Experiment: High Purity Plutonium Electrorefining with the Solvent Anode, 700°C

| | Material, g | | | |
|---|---|---|---|---|
| | Pu (Metal) | Cd | NaCl-KCL | $MgCl_2$ |
| Feed | 85.5 | 105.6 | 244.3 | 7 |
| Product | 65.7 | — | — | — |
| Salt (NaCl-KCl) | 17.0 | — | 200.0 | — |
| Anode Heel | 1.0 | 90.9 | — | — |
| Dissolution | 98.8% | | | |
| Collection | 74%[a] | | | |

[a] Ring was not coalesced due to lower operating temperature. Dendrites formed on cathode.

TABLE 4

Small Scale Experiment: Spent Anode Feed
Electrorefining with the Solvent Anode, 740°C

| | Material, g | | | |
|---|---|---|---|---|
| | Pu (Anode) | Cd | NaCl-KCL | $MgCl_2$ |
| Feed | 419 (468) | 853 | 1191 | 45.2 |
| Product | 262.8 | — | — | — |
| Salt | 84.3 | — | 1053 | — |
| Anode Heel | 58.3 | 833 | — | — |
| Cathode | 31.5 | — | — | — |
| Crucible | 6.5 | — | — | — |
| Dissolution | 86.0% | | | |
| Collection | 81.5% | | | |
| Density | 19.54 g/$cm^3$ | | | |

ever, 98.8% of the plutonium was oxidized and transported from the anode cup. Experiments such as this demonstrated the feasibility of the cadmium solvent anode for plutonium processing.

In experiments in full-scale electrorefining cells, spent anodes from the conventional electrorefining process were treated with calcium to remove any oxides (polishing) and then combined with cadmium to form an anode. Results from a typical experiment are tabulated in Table 4. From 86 to 90% of the plutonium dissolved at 740°C and yields averaged 80%. The product was metallic, high density plutonium of 99.99% purity. The cadmium was 100 ppm in this product. The Cd to Pu ratio in the anode chamber was varied from 1.4

to 3.0 in these runs and yields increased at the higher ratios. Less than 4% of the cadmium was vaporized. The electrolyte divided into a black salt, which contained most of the Pu left in the salt phase, and a lighter salt which formed an upper layer. The lighter salt and crucible were discarded; the plutonium in the black salt was reduced, condensed and recycled through the electrorefining process.

This process combined with conventional electrorefining would recover more than 98% of the feed plutonium. When the Cd-rich spent anode was used for a second electrorefining run, both yield and dissolution decreased. A large amount of black salt was formed. The Cd-rich anode heel was treated with $ZnCl_2$ to oxidize plutonium into the salt phase and form a Zn-Cd button which was free of plutonium. Efforts to process the salt by calcium reduction were unsuccessful. Perhaps aqueous chloride ion exchange is the best method for recovering this plutonium.

In summary, the recovery of spent anodes from the conventional electrorefining process appears to be a viable alternative to the pyroredox process. By combining these processes, more than 99% of the plutonium in the initial electrorefining feed is recovered in two steps as high purity plutonium metal. The remainder is either in residues below our discard limits or can be recycled through an aqueous step.

We plan to study this process employing relatively pure plutonium in the cadmium anode and to measure the effects of variables such as temperature, current density and stirring rate. Our initial tests used a NaCl-KCl eutectic, but we may consider other systems, such as the lower-melting LiCl-KCl eutectic. At lower temperatures, the plutonium could be collected as dendrites on a spinning cathode. Cathodes could be removed, and additional cathodes and feed material added during processing to form a semi-continuous system.

The study of these alternatives to our electrorefining process could lead to changes throughout our integrated pyrochemical system. We may alter the molten salt extraction step (to remove americium) and eliminate a casting step. This work is in progress and results will be reported when available.

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