LA-UR -86-840

CONF-860405--21

Received by OSTI

APR 0 7 1986

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LA-UR--86-840

DE86 008727

TITLE A SOLVENT ANODE FOR PLUTONIUM PURIFICATION

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SUBMITTED TO

International Conference on Separations Science and Technology, April 15-17, 1986, New York City, New York

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A SOLVENT ANODE FOR PLUTONIUM PURIFICATION [*]

by

David F. Bowersox, Keith W. Fife and Dana C. Christensen

To be presented at the International Conference on Separations Science and Technology, April 1[illegible]-17, 1986, New York City, New York

Abstract

The purpose of this study is to develop a technique to allow complete oxidation of plutonium from the anode during plutonium electrorefining. This will eliminate the generation of a "spent" anode heel which requires further treatment for recovery. Our approach is to employ a solvent metal in the anode to provide a liquid anode pool throughout electrorefining. We use molten salts and metals in ceramic crucibles at 700°C. Our goal is to produce plutonium metal at 99.9% purity with oxidation and transfer of more than 98% of the impure plutonium feed metal from the anode into the salt and product phases. We have met these criteria in experiments on the 100-1000g scale. We plan to scale our operations to 4 kg of feed plutonium and to optimize the process parameters.

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[*] Work done at the Materials Science and Technology Division of the Los Alamos National Laboratory under the auspices of the United States Department of Energy.

I. Introduction

We define pyrochemical processes as nonaqueous processes which are conducted at elevated temperatures. In our plutonium reprocessing operations, we oxidize scrap metal to oxide, reduce oxide to impure metal, remove excess americium, cast to shape, and electrorefine to a pure product in a series of pyrochemical operations at temperatures between 700° and 900°C. Our research and development efforts are concentrated on improving the efficiencies of these processes, cutting costs, reducing residues and decreasing exposure of personnel to radiation. This paper will be restricted to a discussion of electrorefining and our efforts to develop a solvent anode to significantly increase our yields and reduce the residue volumes for our process.

Since the mid-forties, pyrochemical operations have been employed to reduce uranium and plutonium tetrafluorides to metal.[1,2] Baker's success with these early reductions led Los Alamos to study related pyrochemical process for recovering the actinides from spent reactor fuels. For metal-based fast reactor systems, fuel could be reprocessed without altering the metal state. Studies of electrorefining were initiated to satisfy purity requirements for the Los Alamos Molten Plutonium Reactor Experiment.[3] This work was further expanded in the early sixties to an electrorefining operation in which impure plutonium metal was melted in a ceramic vessel and electrolytically oxidized into a molten salt, transported through the salt and reduced to high purity metal at a cathode[4] With additional research, we developed an integrated method for producing metal from scrap by a series of pyrochemical operations.[5]

II. Plutonium Electrorefining at Los Alamos

A schematic diagram of the Los Alamos Electrorefining Cell is shown in Fig. 1. The cell is formed by two concentric magnesia crucibles joined at the base. In typical operations, 4-6 kg of impure plutonium metal is placed in the inner crucible and an equimolar mixture of sodium chloride-potassium chloride containing 5 w/o (weight per cent) magnesium chloride is added as an oxidant (to produce Pu (III) in the electrolyte prior to passing a current through the cell). The cell is assembled in a stainless steel furnace tube, evacuated and back-filled with argon. The crucibles are heated to 740°C and the stirrer, anode and cathode lowered into the positions shown in the figure. Current (dc) is passed through the cell, with stirring, until the plutonium is no longer the primary ion being formed at the anode. This is indicated by an increase in the static cell potential, which is measured by a back emf sampling unit. At a pre-set potential, the current is automatically terminated and an operator withdraws the hardware from the melt. The heat is turned off and the system cools to room temperature. The ceramic crucible is broken to recover the salt and metal product. The metal product ring, shown in Fig. 2, is mechanically separated from the salt, sampled for analysis and transferred for casting and future use.

The purity of typical product is greater than 99.9% plutonium. The elements less active than plutonium remain in the anode while those more reactive concentrate in the salt. Impurities in the product are usually tungsten, which is introduced from the cell hardware, and americium. Other impurities are present at concentrations of less than 500 ppm (parts per million).

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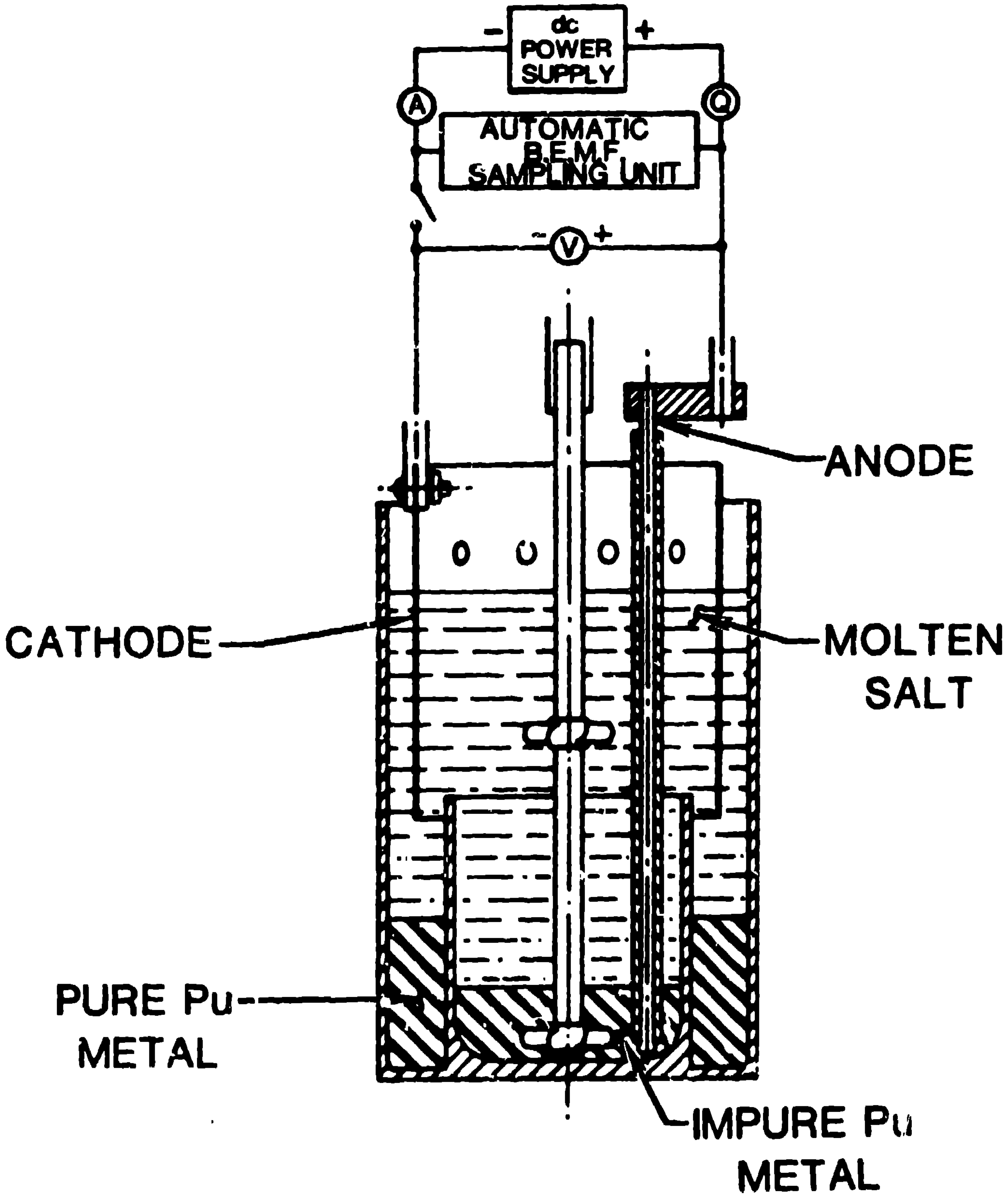


Fig. 1. The (Los Alamos) Electrorefining Cell

[illegible]

Two yields are important in considering the effectiveness of this process. The first, which we will call the oxidation yield, is defined as the per cent of the impure feed which is oxidized and transferred out of the anode. In our present gallium-plutonium system, this yield is about 90%; the remaining 10% of the plutonium forms a solid anode heel containing most of the initial impurities. The second yield, which we designate as the reduction yield, is the per cent of the plutonium in the initial feed recovered as product. In our present system, the reduction yield is typically 70-75%.

The three major residues are the solid anode heel, the electrolyte salt, and the crucible. The anode heel is treated by another pyrochemical process, pyroredox, to recover the remaining plutonium in a form suitable for additional electrorefining.[6] The salt is collected, melted, and contacted with excess calcium to recover residue plutonium.[7] Crucible residues are leached in acid and the plutonium recovered by aqueous methods.[8] The overall recovery of plutonium, considering the residue steps, is satisfactorily high.

Since the inception of the process in 1962, large quantities of extremely pure plutonium have been produced by this process. However, it has a history of somewhat low yields. As described above, the solid anode heel is a major residue. We are now investigating methods of oxidizing and transferring more of the plutonium from the anode compartment to eliminate processing of the spent heel. We propose using a low melting solvent metal in which plutonium is significantly soluble. This report will describe our experiments employing cadmium as the solvent metal in the anode pool along with a discussion of our preliminary results.

III. Approach to Solvent Anode Electrorefining

The behavior of impurities in the electrorefining can be predicted from the free energies of formation of the chlorides. Some of these energies, at 1000°K, are listed in Table 1. For elements with more than one oxidation state, the $-\Delta F^{o}$ for the most stable form of chloride is given. Because plutonium has a quite high value for the energy of formation, most impurity elements remain in the anode pool while plutonium is oxidized. For example, for iron the reaction of interest is

$$3\ \mathrm{Fe(0)}\ (\ell) + 2\ \mathrm{Pu(III)} = 3\ \mathrm{Fe(II)} + 2\ \mathrm{Pu(0)}\ (\ell) \qquad (1)$$

The calculated free energy for this reaction is 96 kcal/mol Pu at 1000°K, which is equivalent to an equilibrium constant of 10^{-21}.[9] The ratio of Fe(II) to Pu(III) is very low because

$$\frac{\mathrm{Fe(II))}^3}{(\mathrm{Fe(0))}^3} = 10^{-21}\ \frac{(\mathrm{Pu(III)}^2}{(\mathrm{Pu(0))}^2} \qquad (2)$$

Assuming that the activity coefficients are within an order magnitude of being equal, and if the metal state of Pu(0) is taken as unity,

$$\frac{\mathrm{Fe(II))}^3}{(\mathrm{Fe(0))}^3} = 10^{-21}(\mathrm{Pu(III))}^2 \qquad (3)$$

and the amount of ferrous ion in the electrolyte is negligible compared to Pu(III). From similar calculations, we predict that most impurities will remain in the anode. These predictions have been verified by our experimental data, as shown by the data for the behavior of impurity elements in electrorefining (Table II). The elements with higher free energy values, such as americium and cerium, will be preferentially oxidized and concentrate in the salt phase.

The major reaction at the anode, as long as Pu(0) reaches the metal electrode, will be

$$\mathrm{Pu(0)},(\ell)_{\mathrm{impure}} \rightarrow \mathrm{Pu(III)} + 3e^{-}. \qquad (4)$$

TABLE I.

ΔF VALUES AT 1000°K FOR IMPURITY ELEMENTS IN ELECTROREFINING

Element	$-\Delta F^{o}$, kcal/g.atom Cl
Ni	18
Cu	21
Ta	22
Fe	27
Cr	32
Ga	32
Mn	41
Al	46
U	54
Th	59
Pu	59
Ce	66
Am	67

TABLE II.

BEHAVIOR OF IMPURITY ELEMENTS IN ELECTROREFINING

Element	Concentration (ppm) Feed	Concentration (ppm) Product	Element Concentrated in
Ni	750	<10	Anode
Cu	100	2	Anode
Ta	5 000	80	Anode
Fe	25 000	20	Anode
Cr	280	<10	Anode
Ca	10 000	<25	Anode
Mn	70	< 2	Anode
Al	2 700	< 5	Anode
U	275	<20	Anode
Th	160	<10	Anode
Pu	---	---	---
Ce	527	<25	Salt
Am	850	82	Salt

The Pu(III) will be carried through the salt phase to the cathode. The major reaction there, under our operating conditions, will be

$$\mathrm{Pu(III)} + 3e^{-} = \mathrm{Pu(0)},(\ell)_{\mathrm{pure}} \tag{5}$$

and the overall reaction is

$$\mathrm{Pu(0)},(\ell)_{\mathrm{impure}} = \mathrm{Pu(0)},(\ell)_{\mathrm{pure}} \tag{6}$$

The plutonium collects on the cathode and then drips into the annulus between the outer and inner walls to form the product ring.

If a solvent anode pool is employed, reaction (1) is altered to become

$$\mathrm{Pu(0)}_{\mathrm{impure}}\mathrm{Cd(0)},(\ell) = \mathrm{Pu(III)} + \mathrm{Cd(0)},(\ell) + 3\ e^{-}. \tag{7}$$

Transport and the cathode reaction will remain unchanged.

The amount of plutonium in the metal product ring can be estimated from the equilibrium constant for the reaction

$$\mathrm{Am(III)} + \mathrm{Pu(0)},(l) = \mathrm{Pu(III)} + \mathrm{Am(0)},(\ell) \tag{8}$$

and

$$\frac{(\mathrm{Am(0)})}{(\mathrm{Am(III)})} = \frac{1}{K_1}\ \frac{(\mathrm{Pu(0)})}{(\mathrm{Pu(III)})} \tag{9}$$

Under our operating conditions, about 10% of the americium is transferred into the product. Since the (Pu(0)) is approximately unity, we should be able to decrease this ratio by increasing the Pu(III) concentration in the electrolyte.

The composition of the salt phase should not change during electrorefining if the cell potential remains below its decomposition potential. In our regular process, we add $MgCl_2$ to the electrolyte to oxidize some of the feed plutonium prior to electrorefining. This introduces magnesium which can be vaporized out of the system or concentrated as an impurity in the anode. In our solvent anode experiments, substitution of K_3PuCl_6 has eliminated this contaminant. If we recycle the electrolyte, we will already have sufficient Pu(III) in the salt phase to start reducing and collecting plutonium. However, the more active impurities, such as americium, will concentrate in the salt and eventually contaminate the product.

We believe that cadmium transferred into the product by vaporization and condensation in the experiments using NaCl.KCl at 750°C. We found that as much as 15% of the cadmium moved into the product. Lowering the operating temperature to 700°C and employing LiCl.KCl as the electrolyte eliminated most of this problem.

In most of our routine electrorefining, the plutonium has been alloyed with gallium to reduce the density and insure a critically safe operation. From the plutonium-gallium phase diagram, the alloy will be liquid as the electrolysis begins. As it continues, the plutonium concentration decreases, and the concentrations of gallium and the impurities in the anode increase. Finally, the anode becomes pasty and solid as the gallium concentration approaches 16.8 a/o(atomic per cent). As the plutonium concentration continues to decrease, the stirring becomes less and less effective. At about 25 a/o gallium, the anode is completely solid. The principle desired reaction has become

$$\mathrm{Pu(O),(s)} = \mathrm{Pu(III)} + 3e^- \qquad (10)$$

and the potential for plutonium oxidation is

$$E = 0.0291 \ \ln\ (\mathrm{Pu(O)},\ (s))_{\text{anode surface}} \qquad (11)$$

The diffusion of plutonium through the solid anode material to the anode surface becomes much too slow to meet the conditions of high current density that prevail at the anode. Thus, the Pu(O) concentration at the anode approaches zero and competing reactions predominate. Impurity elements will be oxidized, transported to the cathode, reduced and contaminate the metal product. By monitoring the cell potential and terminating the run at a static cell potential of 0.35 to 0.4 v, we maintain high purity product. However, almost 10% of the plutonium feed remains in the solid anode heel.

By using a solvent anode pool, the anode material does not solidify, and plutonium should continue to be oxidized as long as it reaches the anode surface. Our experiments, which show very high oxidation yields, demonstrate the effectiveness of this approach. The high purity of the product indicates that even at very low plutonium concentrations, (less than 1% of the initial feed value), impurity elements are not oxidized into the salt.

IV. The Solvent Anode Experimental Results

In a solvent anode system, a metal with a relatively low melting point and with no high melting compounds which form with plutonium is used to form an anode pool. We evaluated a number of possible candidate metals and selected cadmium for our initial experiments. Figure 3 shows the plutonium-cadmium phase diagram.[10] As can be seen by the solidus lines, concentrations of cadmium greater than 85 a/o(atom per cent) will ensure some liquid throughout electrorefining at temperatures of 750°C. The cadmium-plutonium system seems quite promising, even though the vapor pressure of cadmium is quite high at the normal operating temperature of our electrorefining cells.[10] In our initial tests we modified a small-scale (500g) cell and tested it with high purity plutonium as the initial feed. The resulting product is compared in volume with the anode residue in Fig. 4. The ring was 99.9% pure metal, and the oxidation yield was 98.8%. This shows that, with high purity metal in the initial feed, oxidation yields can be very high.

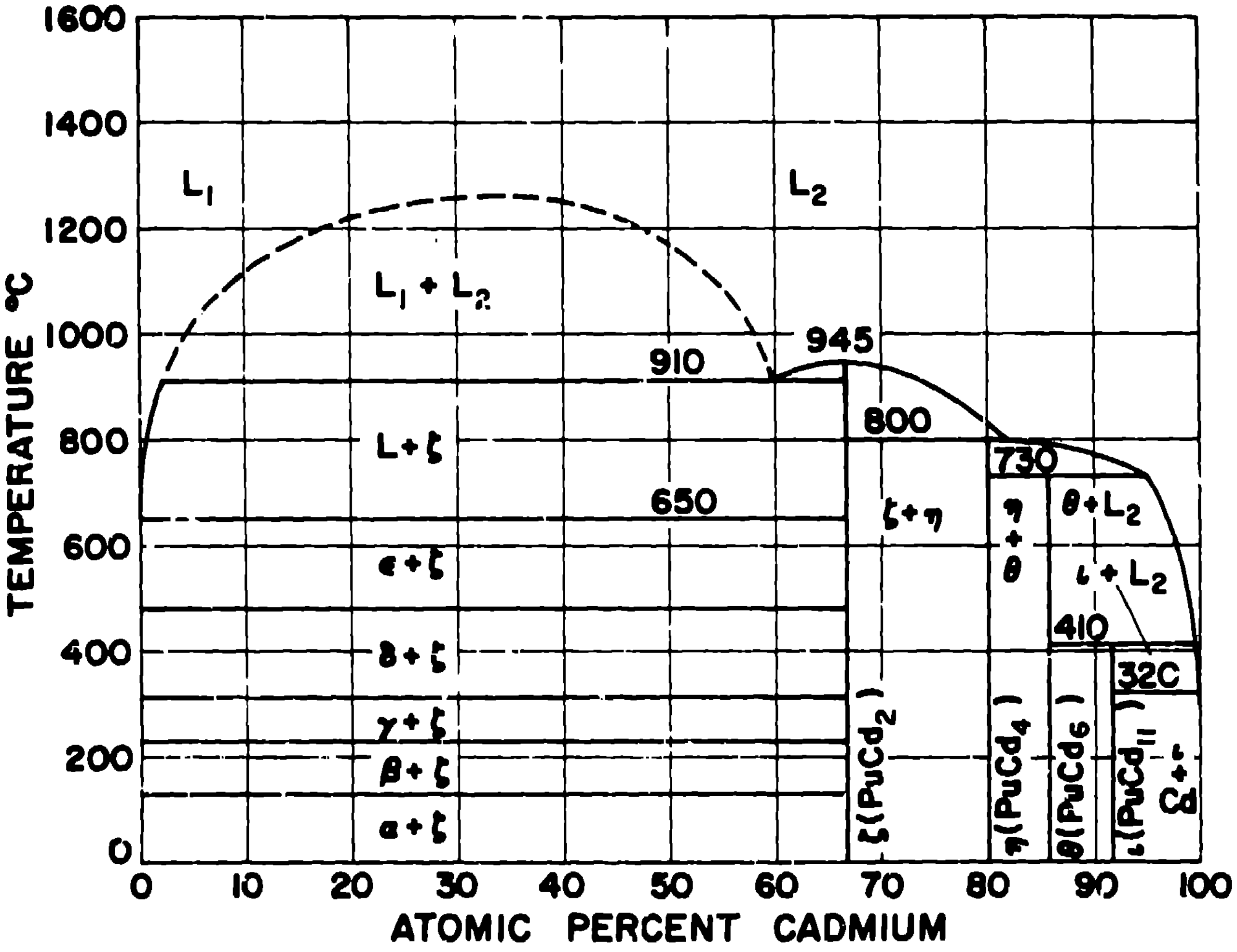


Fig. ~~8~~ 7. Plutonium-cadmium phase diagram. ~~(Redrawn from~~ Ref. 1.)

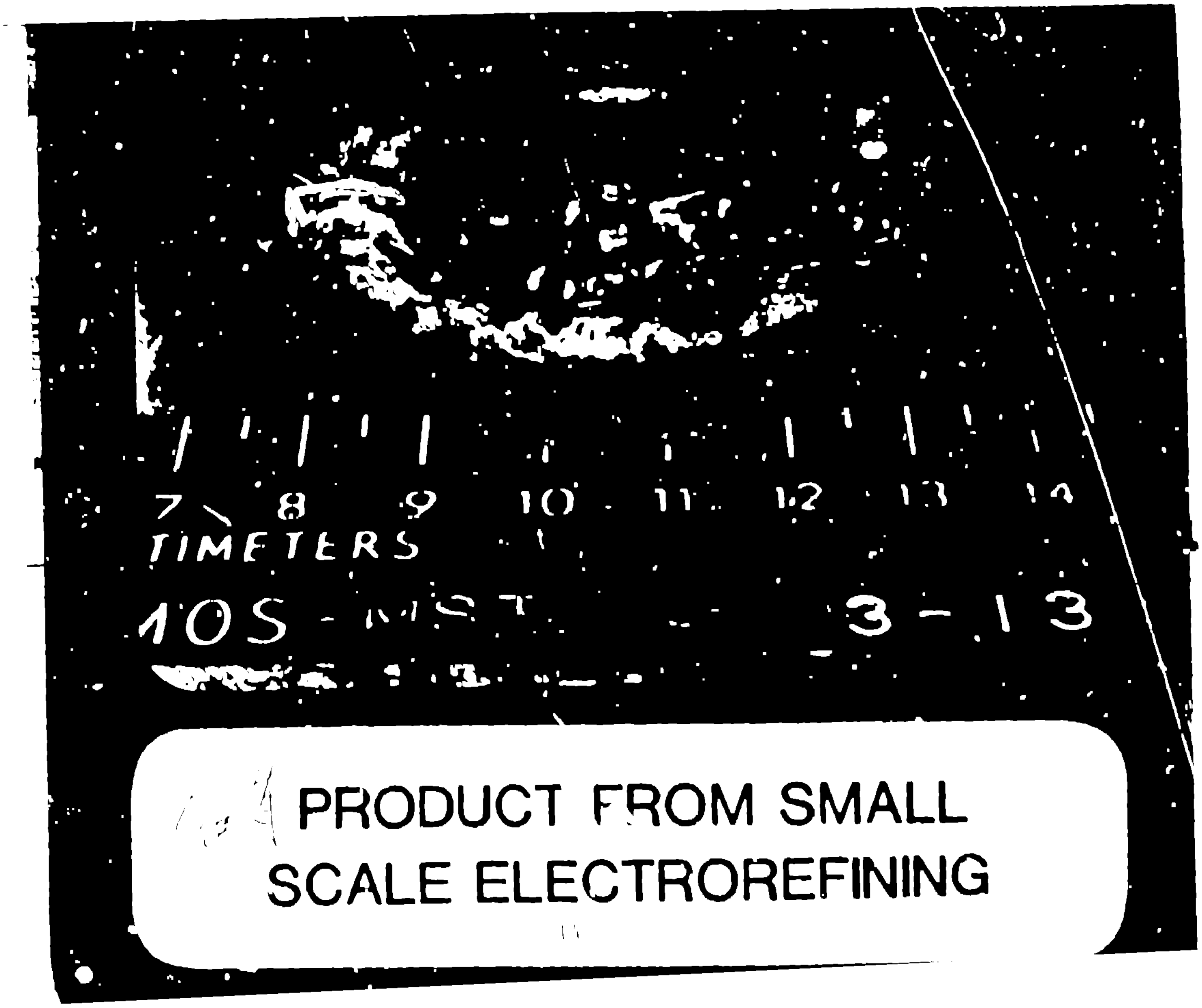


Fig 4 PRODUCT FROM SMALL SCALE ELECTROREFINING

We then conducted experiments using cadmium metal and actual anode residues from the conventional electrorefining process. The product of an experiment is shown in Fig. 5. In the figure, the product metal, (on the bottom as a shiney ring), is 99.9% plutonium of 19.4 g/cc density. The salt, containing blue $PuCl_3$, lies on top of the product ring. Some salt has been broken away to show that the anode cup wall has not been wetted by the cell reagents or products. Over 98% of the plutonium was oxidized and transported from the anode cup. The high oxidation yields and high purity product encouraged us to continue with larger scale operations.

In 6 kg-scale electrorefining cells, we are limited to approximately 1 kg of plutonium as feed because of the volume of cadmium required for a solvent pool that contains no high melting components. Spent anodes from our electrorefining process were first contacted with calcium at 850°C to remove any surface oxides and then added to cadmium in the anode compartment. The Cd:Pu ratio was varied from 1.4:1 to 3.0:1. Oxidation yields increased with increasing ratios. Oxidation yields at 740°C varied from 86 to 90% and reduction yields averaged 80%. Results of a typical run are given in Table III. As shown, the white salt was very low in plutonium and, as a result, could be discarded. The other residues could be treated to recover the remaining plutonium. The product ring contained two phases. The upper, lighter phase contained 15g of cadmium and 55g of plutonium. This was easily separated from the high density, pure plutonium in the lower phase. We oxidized 98.9% of the feed plutonium and collected 79% as high purity product. An additional 18g remained on the cathode surface.

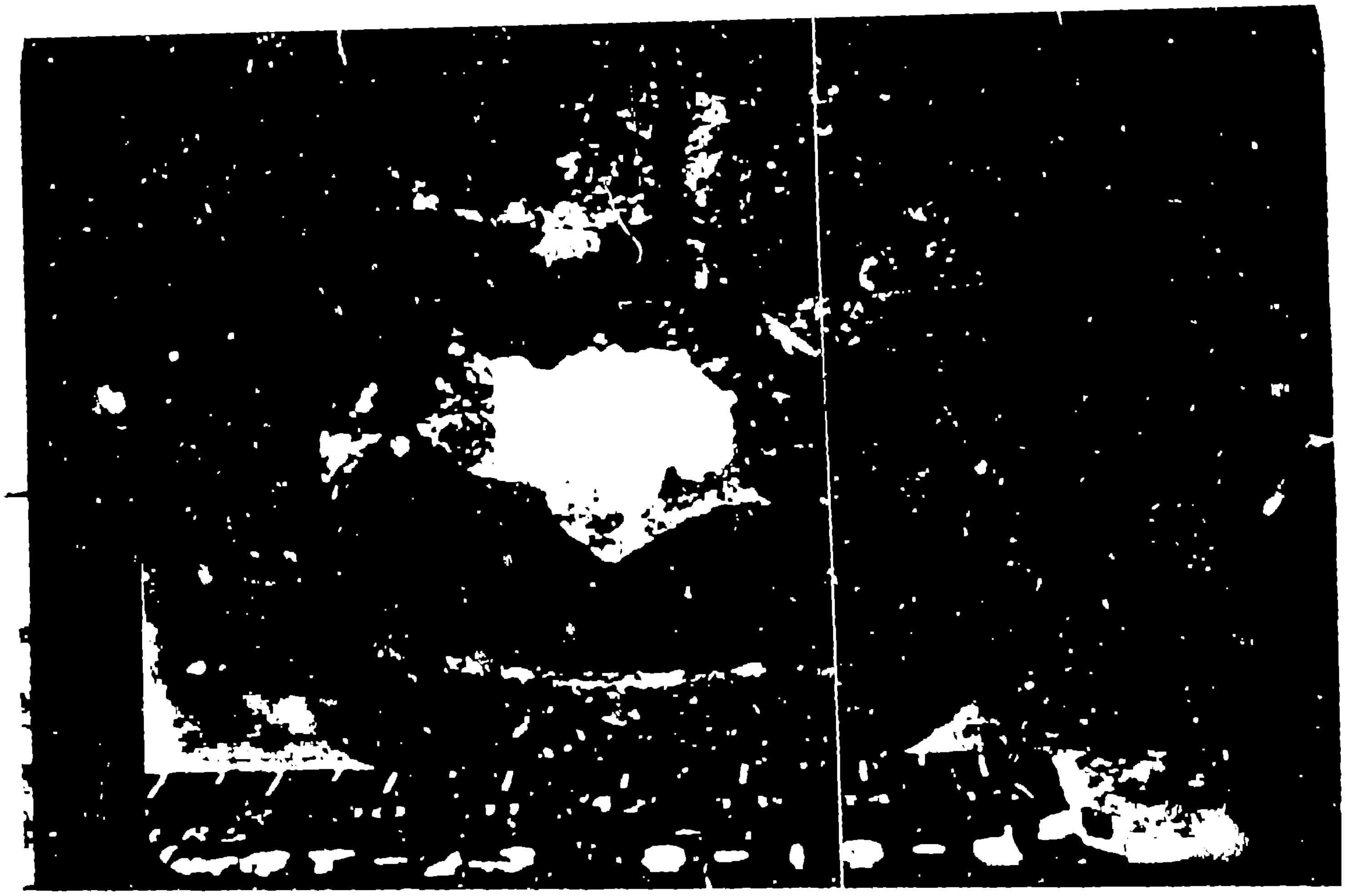


Fig. 4 Small Scale Solvent [illegible] Experiment

TABLE III.

SUMMARY OF EARLY ELECTROREFINING EXPERIMENT
(ER 27R) AT 750°C

Operating Conditions

725–750°C for 24h with stirring
6 amps, 1.2v
End point potential: 0.4v

Initial Composition	
Pu	544g
NaCl.KCl	2350g
Cd	1500g
$MgCl_2$	75g

Residues	
Cathode	18g Pu
Black salt	23g Pu
White salt	10g Pu
Crucible shards	2g Pu
Anode heel	6g Pu

Product	
Cd (top ring)	15g Cd, 55 g Pu
Product ring	430g Pu, density 19.5g/cm^3

Oxidation Yield	98.9%
Reduction Yield	79.0%

A spent cadmium anode and a product ring from a similar run are shown in Fig. 6. The lower metallic ring, which was 99.9% plutonium, contained 100 ppm cadmium. The upper ring was removed and recycled in a subsequent experiment. The electrolyte formed two phases; a lower black salt which contained almost all the plutonium in the salt, and a light-colored upper layer. The lighter salt and crucible could be discarded; the plutonium in the black phase was recovered pyrochemically.

Several steps were taken to improve our procedures and to further define the experimental parameters. We replaced the ceramic stirrer used on conventional electrorefining with an electrically-isolated tantalum stirrer designed for efficient mixing. We substituted a LiCl.KCl eutectic with a lower melting point for the NaCl.KCl and lowered our operating temperature to 700°C. At this temperature, the cadmium vapor pressure is significantly lower, and the second layer of product did not form. Finally, we replaced $MgCl_2$, which had been added as an oxidant, with K_3PuCl_6 to provide initial Pu(III) in the electrolyte. This substitution reduces the Mg present in the anode compartment and the anode heel.

The results of three experiments with these modifications are summarized in Table IV. In each run, 400g of K_3PuCl_6 was added to the eutectic salt. The system was maintained at 700°C and the stirring rate adjusted to 200-250 rpm. The current was set at 8-10 amps, and the voltage was 1.2v. The runs terminated when the back emf reached 0.35-0.4v. A 1950g cast cylinder of cadmium was combined with the plutonium and placed in the anode compartment. The 2000g of eutectic salt was added to the crucible. In all three cases, the oxidation yield was greater than 95% and the metal product was of satisfactory purity

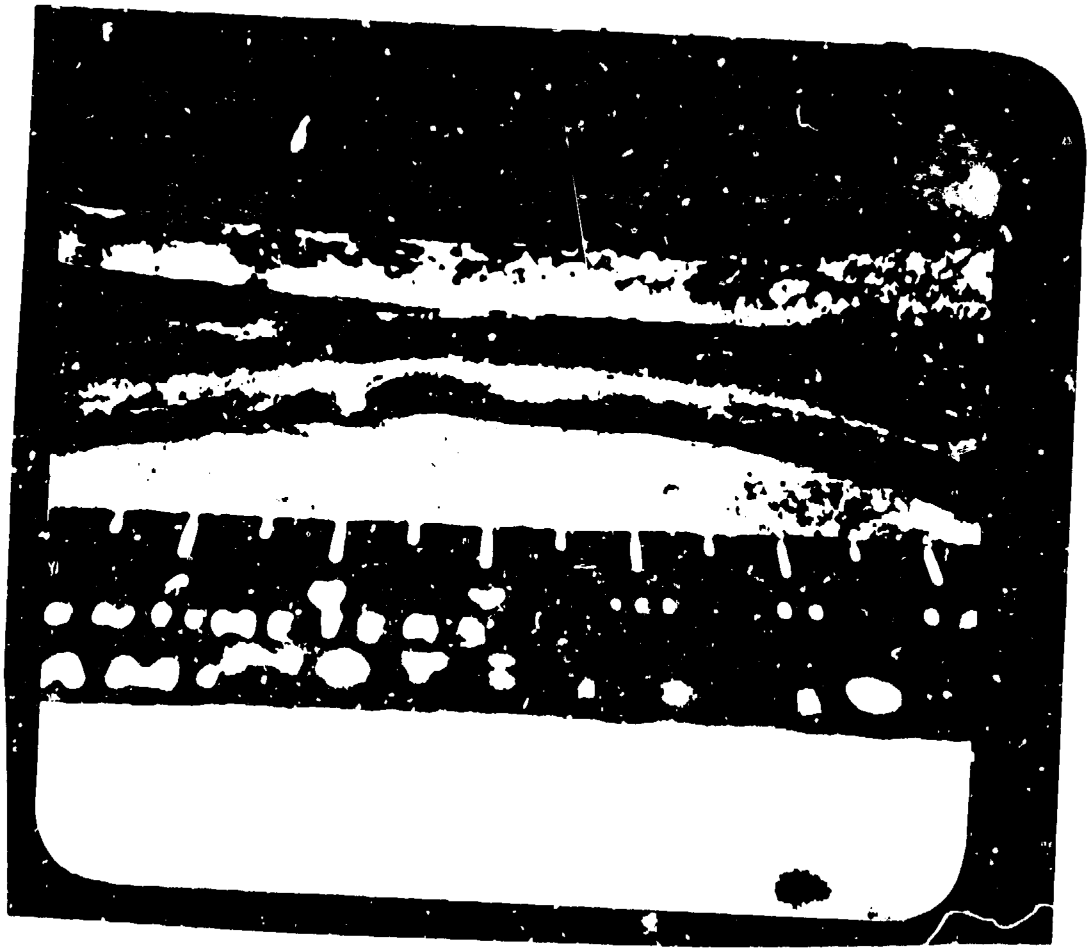


Fig 9 Conduct ring with cadmium transfer

TABLE IV.

SUMMARY OF MODIFIED ELECTROREFINING EXPERIMENTS, 700°C

Operation Conditions
Temperature 700°C
Stirring rate 200 - 250 rpm   400g K_3PuCl_6 in eutectic salt
Current 8 - 10 amps, 1.2v
End Point Potential 0.4v

ER	33	34	35
Initial Composition			
Pu, feed (g)	650	665	642
Cd (g)	1950	1950	1950
Eutectic (g)	2100[a]	2100[a]	2000[b]
Results			
Oxidation Yield	95	99	99
Reduction Yield	72	72	66
Product density, g/cc	19.2	19.3	19.7

[a] $\mathrm{LiCl.KCl}$

[b] $\mathrm{NaCl.KCl}$

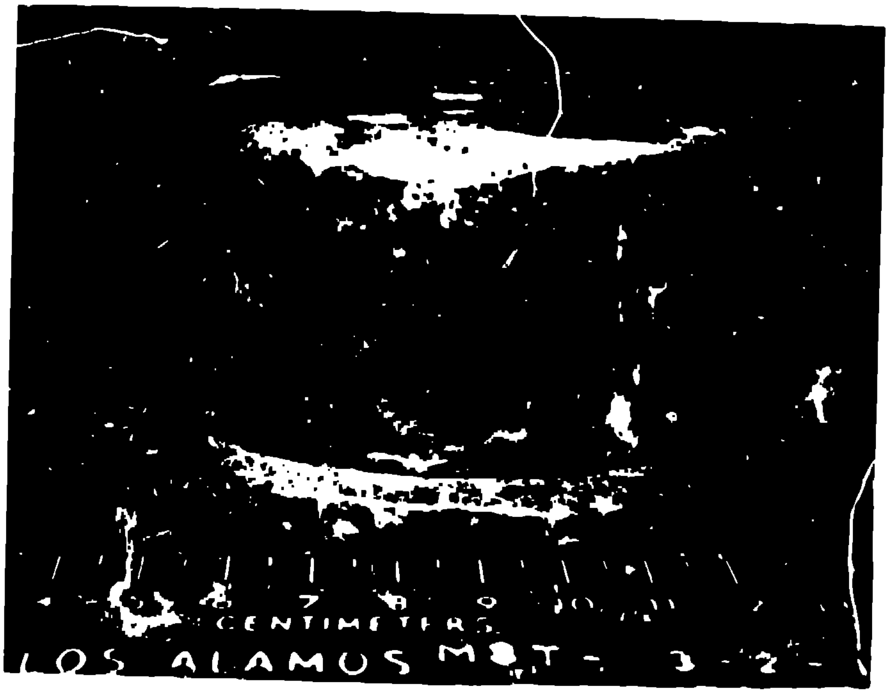
(99.9%). The reduction yield, however, was only about 70%. Figure 7 shows the product metal and the spent anode from ER 35. Figure 8 is the product ring from a different angle. The spent anode contains less than 1.3% (8g) of the initial plutonium. The product is high purity plutonium with an unusually high cadmium concentration (3000 ppm).

V. Discussion and Conclusions

The reduction yields of 70-80% are lower than we would like for this process. We believe that the reduction yield can be increased by increasing the initial Pu(III) concentration in the electrolyte. We also plan to vary some of the operating parameters. The cadmium-containing anode could be reused until the impurity buildup becomes high enough to contaminate the product or decrease the yields. At that point, most of the cadmium could be recovered by distillation, recast and recycled. The residue remaining after distillation would either be discarded or, if the plutonium content were above the discard level, treated by alternate means.

We do not recycle the electrolyte at this time; however, there are no major obstacles to instituting such a step in the process. Reuse would be limited by buildup of americium and its transfer into the product. When necessary, the plutonium and americium in the salt can be separated and recovered by pyrochemical reduction. The salt could then the reused or discarded.

We are successfully using cadmium as our solvent element, even though it exhibits a high vapor pressure at the temperatures of interest. Other metals with relatively low melting points may prove more effective, provided that plutonium is at least somewhat soluble and that no high-melting alloys or compounds are formed in the composition range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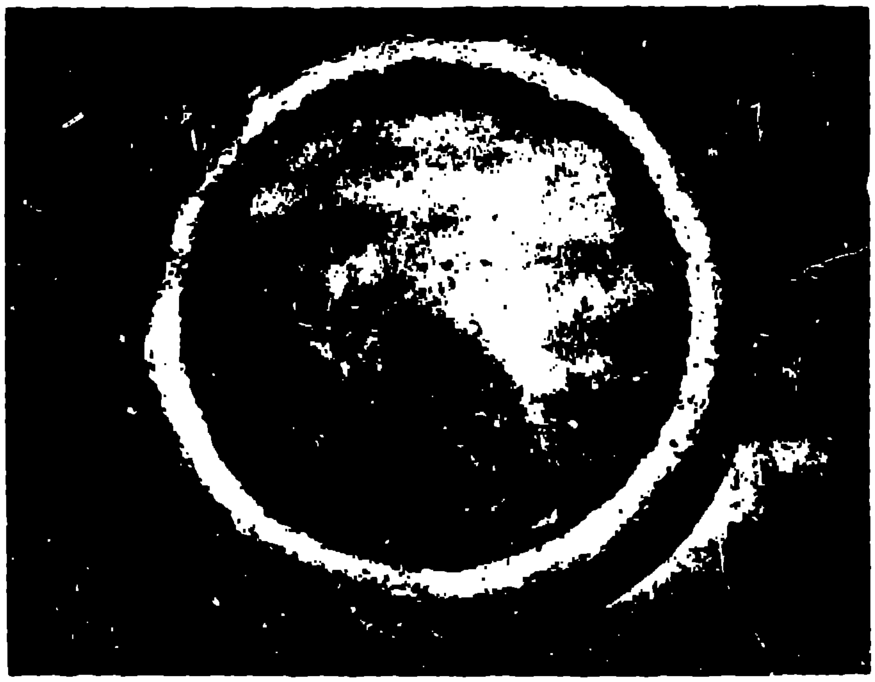

Fig. 7 [illegible] A [illegible] ring from
A pendant ring from a [illegible]
[illegible],

of interest. The volume of cadmium required is too large for effective use of our present electrorefining cell with larger quantities of plutonium. We plan to redesign the cell. We may attempt to operate at lower temperatures and collect the plutonium product on a metal cathode.[11, 12] In such a system, the solid product could be withdrawn and separated. Additional electrodes and impure feed could be added to the cell for continuous processing. When impurity build-up occurred, the reagents could be withdrawn and recycled or discarded.

These experiments utilizing a cadmium solvent pool for the anode in electrorefining plutonium have been very successful. High oxidation yields, averaging 99%, demonstrate that the plutonium is indeed being oxidized to depletion in the anode under the operating conditions. The product metal is of quality at least equal to that obtained in our regular process. The spent anode no longer needs to be processed to recover 10% of the initial feed, and recycle of the reagents appears to be feasible. Reduction yields are lower than desirable, but we plan to change conditions to improve them.

More experiments are needed to define optimum stirring rates, ranges of effective initial Pu(III) concentrations and an optimum cell potential for current termination. We hope to develop reference electrodes suitable for examining electrode reactions and identifying cell potentials. Recycle of the electrolyte and the spent anode should be demonstrated. We plan to consider several cell designs and to test a semi-continuous cell. Other solvent metals and electrolytes may be tested. At lower temperatures, for example, plutonium could be collected as a solid on the cathode, removed, and treated separately. If mercury were the solvent, electrorefining could be carried out in Pyrex equipment at 300°C.

We are continuing this research program. The study is challenging, and the potential rewards are large. We plan to present more data as our study progresses.

VI. Acknowledgements

The authors wish to thank C. Brown and P. C. Lopez for the experimental work reported in this paper. We also acknowledge the ideas, advice and encouragement that we have received from J. G. Reavis of Los Alamos, L. J. Mullins of TRUECO, M. C. Coops and J. B. Knighton of Lawrence Livermore National Laboratory, and M. J. Steindler, L. Burris and W. E. Miller of Argonne National Laboratory.

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