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Kilogram-Scale Electrorefining of Plutonium Metal

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Six-Kilogram Scale Electrorefining of Plutonium Metal

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CONTENTS

PAGE

ABSTRACT.. 1

I. BACKGROUND.. 1

  A. Criticality Constraints in Plutonium Processing...... 1

  B. Why Electrorefine?................................... 2

  C. Plutonium Electrorefining, Principles of Operation... 2

  D. Review of Los Alamos Electrorefining Process......... 4

    1. History.. 4

    2. Equipment and Materials.......................... 4

    3. Operating Procedures............................. 6

    4. Material Balances................................ 7

II. PRESENT STUDIES.. 7

  A. Objectives... 7

  B. Equipment Design..................................... 9

III. EXPERIMENTAL RESULTS.................................. 9

IV. PROCESS IMPLEMENTATION................................. 15

V. CONCLUSIONS... 19

ACKNOWLEDGMENTS 19

REFERENCES... 19

SIX-KILOGRAM SCALE ELECTROREFINING OF PLUTONIUM METAL

by

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ABSTRACT

The electrorefining of metallic plutonium scrap to produce high purity metal has been an established procedure at Los Alamos since 1964. This is a batch process and was limited to 4-kg plutonium because of criticality safety considerations. Improvements in critical mass measurements have permitted us to develop a process for 6-kg plutonium.

The 6-kg process is now operational. The increased size of the process, together with other improvements which have been made, makes plutonium electrorefining the principal industrial tool for processing and purifying metallic plutonium scrap.

I. BACKGROUND

A. Criticality Constraints in Plutonium Processing

The scale of ^{239}Pu processing operations is determined primarily by criticality safety limits. (Criticality safety is defined as the art of avoidance of a nuclear excursion.[1]) These limits depend on many factors such as plutonium mass and concentration, system geometry, neutron moderation, neutron reflection, neutron poisoning, and system interactions. Neutron moderation, that is, the slowing down of neutrons by light elements such as hydrogen, has a dramatic effect on limits. This is demonstrated by comparing batch limits for handling dry PuO_2 powders and aqueous solutions of plutonium nitrate. The Los Alamos limits are

8.8 kg for the oxide and 0.25 kg for nitrate solution. Geometry and neutron poisons also play dominant roles. For example, the 0.25-kg limit for aqueous solutions can be increased to 1.00 kg by using vessels equipped with large diameter stirrer shafts containing boron nitride. Most of the increased limit in this case is due to the annular-vessel shape created by the large stirrer shaft. Thus, neutron moderation and geometry effects are of primary concern to the plutonium-process designer. An inherent advantage of molten salt processing is the absence of water moderation.

B. Why Electrorefine?

Electrorefining is a very attractive operation for applications in which the feed materials are impure metals or alloys and the desired product is pure metal. In this process, impure metal is dissolved at the anode and pure metal is deposited at the cathode. This simple electrolytic conversion of impure-to-pure metal eliminates the many processing steps in conventional plutonium recovery through chemical processing. In addition, electrorefined metal is purer than that produced by chemical conversion of compounds to metal.

Electrorefining has been used extensively for commercial purification of metals in aqueous solutions. For example, many major refineries use aqueous electrolytes for purifying copper, nickel, cobalt, lead, tin, silver, and gold.[2] Active metals, such as alkalis, alkaline earths, lathanides, and actinides, however, cannot be deposited in aqueous solution. These metals require molten salt electrolytes. Molten salt electrorefining processes have been studied extensively for aluminum, lead, plutonium, beryllium, niobium, titanium, vanadium, zirconium, tungsten, molybdenum, uranium, tin, and antimony.[3] In spite of these numerous studies and the potential advantages of electrorefining, aluminum refining is the only process that has found extensive industrial use.[3] Molten salt electrorefining, however, is an ideal processing tool for active metals that have reasonably low melting points, such as aluminum (660°C), plutonium (640°C), and neptunium (637°C).

C. Plutonium Electrorefining, Principles of Operation

In the Los Alamos refining cell (Fig. 1), the magnesia crucible consists of two concentric cylindrical containers. The inner cup contains the impure molten

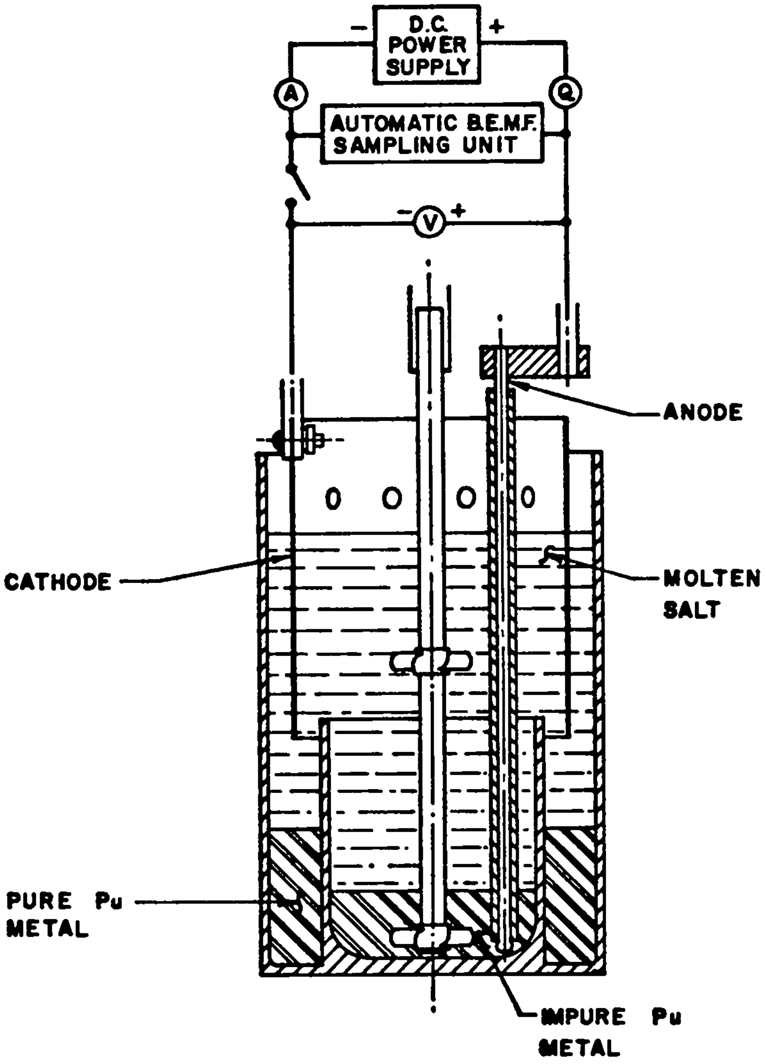

Fig. 1. Los Alamos 4-kg electrorefining cell.

plutonium anode and the outer cup contains the electrolyte. During operation, plutonium is oxidized at the anode and reduced back to metal at the cathode. Therefore, the height of metal in the anode decreases during a run as the height of the product ring or doughnut increases. The cell reactions are simply

$$\text{Anode} \quad - \qquad \text{Pu (impure)} \longrightarrow \text{Pu}^{+3} + 3e^- \quad ; \tag{1}$$

$$\text{Cathode} \quad - \qquad \text{Pu}^{+3} + 3e^- \longrightarrow \text{Pu (pure)} \quad . \tag{2}$$

Therefore, the net cell reaction is

$$\text{Pu (impure)} \longrightarrow \text{Pu (pure)} \quad . \tag{3}$$

Because plutonium is a very active or electropositive metal, most of the impurities remain in the anode. Elements such as americium and cerium, which are more electropositive than plutonium, concentrate in the electrolyte. To achieve these separations, however, the electrorefining cell must operate close to equilibrium. Thus, metal and salt phases must be molten and must be stirred effectively. Under these conditions, we can use standard free energy of formation values to estimate purification factors.[4] To ensure that the cell is indeed operating under equilibrium, the electrolyzing current is interrupted periodically during a run, and the polarization potential or instantaneous back-emf is measured. Electrorefining is terminated automatically when the back-emf exceeds a preset limit. (See Ref. 5 for a discussion of the theory and operation of plutonium electrorefining cells.) Electrorefining will continue as long as a plutonium-rich liquid phase is present at the anode metal-electrolyte interface. The back-emf process control ensures product purity while permitting high anode-dissolution yields. It also permits unattended operation of the electrorefining cell and terminates the operation automatically.

D. Review of Los Alamos Electrorefining Process

1. History. The electrorefining of metallic plutonium scrap to produce high purity metal has been an established procedure at Los Alamos since 1964.[6] During the period 1964 through 1977, 1568 kg of plutonium metal, >99.95% pure, was produced in 653 runs from 1930 kg of metal fabrication scrap.[6] This was a batch operation in which the anode metal feed contained approximately 3 kg of plutonium and the plutonium product weighed 2.4 kg. Optimization of anode feed castings in 1980 permitted an increase to 4 kg anodes with a resultant increase in product throughput.[7] (Note - the criticality limit for this process during the period 1964-1980 was 4 kg plutonium.)

2. Equipment and Materials. The electrorefining equipment is shown in Fig. 2. The electrorefining crucible is contained in a tantalum safety can and a stainless steel loading can. The loading can is contained in a sealed furnace

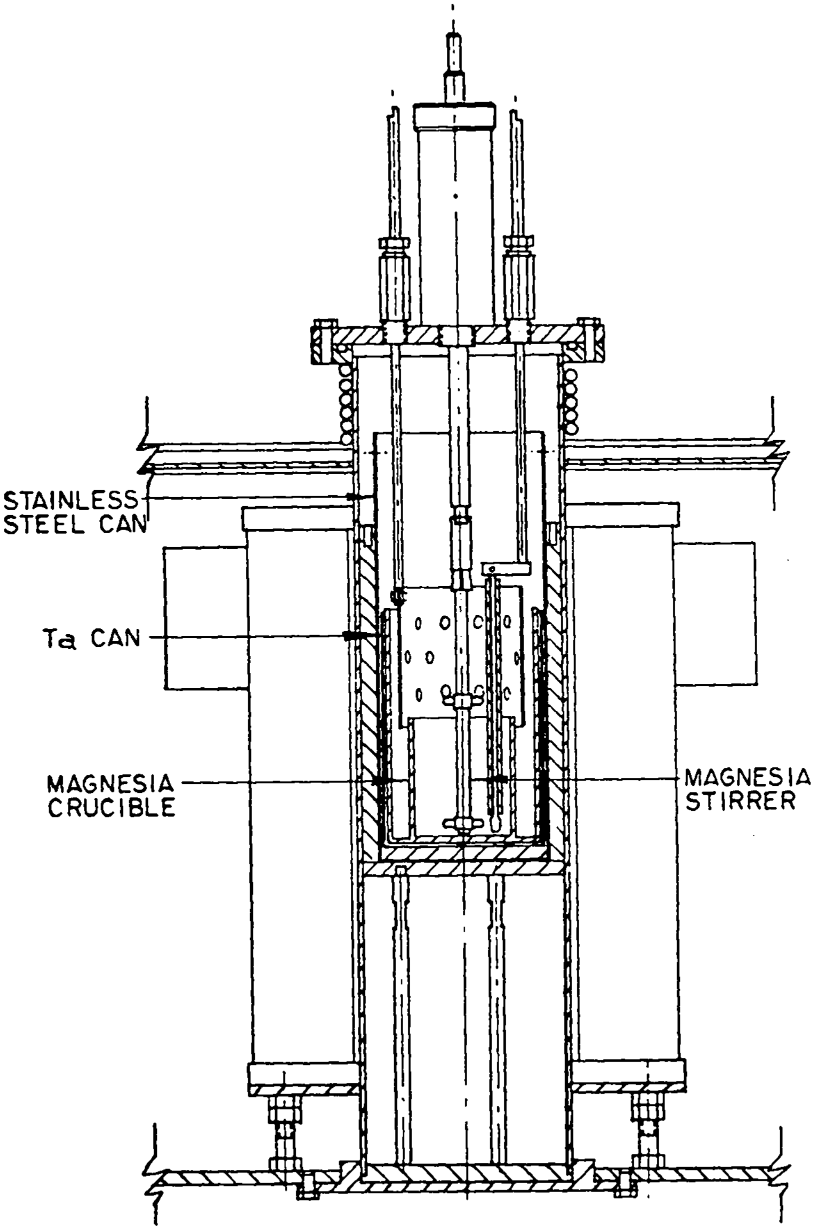

Fig. 2. Electrorefining process equipment, 4-kg scale.

tube and heated by a resistance furnace (Linderberg Model 6015S). The electrorefining crucible, Fig. 1, consists of two magnesia crucibles cemented together. The inner cup contains the impure metal feed. Both the impure metal and the molten salt electrolyte are stirred by a magnesia stirrer. A tungsten rod suspended in the impure metal pool serves as the anode rod. The anode rod is electrically insulated with a magnesia sleeve. A cylindrically shaped sheet of tungsten is suspended in the annular space between the two cups and serves as the cathode.

The magnesia crucible and the magnesia stirrer are highly vitrified bodies and are fabricated commercially.

Raw materials for the anode rod and cathode are purchased commercially and the actual parts are fabricated at Los Alamos.

The sodium chloride and potassium chloride reagents are AR grade and are purchased commercially. The salts are dried overnight under vacuum at ∿150°C and then cast into cylinders. Before use, a hole is drilled in one end of the salt casting. This hole provides a cup for reagent electrolytes.

3. Operating Procedures. The plutonium feed to the process is approximately a 4-kg cylindrical ingot, which is prepared by vacuum casting. The ingot is placed in the inner cup of the crucible. The crucible is loaded into the tantalum safety can that is placed in the stainless steel loading can. The assembly is then lowered into the furnace tube. The 1400-g salt casting is placed, hole up, on top of the inner crucible cup. The PuF_4 reagent is poured into the hole of the salt casting. The cover, containing the stirrer, cathode, and anode rod, is bolted to the furnace tube. A vacuum is pulled on the furnace tube and the assembly is checked for gas tightness. The furnace tube is filled with argon to 4 psig.

The crucible is heated at a rate of 50°C/h to 750°C.

The stirrer, cathode, and anode rods are lowered into position. The actual electrorefining process is accomplished by stirring at 800 rpm and passing a dc current between anode and cathode. The electrorefining is preceded by a pre-equilibration and pre-electrolysis treatment that purifies the electrolyte. As plutonium is dissolved at the anode, the level of metal in the inner cup drops. Plutonium ions are reduced to metal on the cylindrical tungsten cathode. The plutonium drips off the bottom of this cylinder in large globules and collects in the annulus between the small and large crucibles. The progress of a run is monitored automatically by the back-emf sampling unit. This simple device operates by interrupting the dc current periodically and measuring the polarization potential of the chemical cell

$$Pu\ (pure)/NaCl\text{-}KCl\text{-}PuF_3/Pu\ (impure).$$

If the polarization potential exceeds a preset value, the dc current stays off. Thus, the electrorefining is terminated automatically. This measuring device permits unattended operation of the electrorefining process and terminates a run before impurities are dissolved. Upon completion of the electrorefining, the stirrer, anode rod, and cathode cylinder are raised from the melt and heating is discontinued. After cooling and unloading, the metal ring product is isolated from the crucible and salt.

Typical conditions for a 4-kg scale electrorefining are given in Table I. The anode feed weighs 4100 g and contains 4000 g plutonium. The amount of PuF_4 added is determined by the requirements for americium removal. (For a detailed discussion of this point, see Ref. 6). The maximum direct current used is 30 A. The actual current used for any particular run is calculated to permit termination of the run at a convenient time (Note - the units are manned on a 40-h week). For most runs, the current is approximately 20 A.

TABLE I

TYPICAL CONDITIONS FOR ELECTROREFINING
(4-kg SCALE)

Weight of impure Pu, g	4100
Weight of NaCl-KCl, g	1400
Weight of PuF_4, g	120
Temperature, °C	750
Current, A	20
Pu Collection rate, g/h	59

4. Material Balances. A typical material balance for a 4-kg electrorefining run using an impure Pu-1 wt% Ga alloy feed is given in Table II.

II. PRESENT STUDIES

A. Objectives

As a result of criticality studies reported in 1969,[8] the following Los Alamos limits were established for metal casting and other plutonium foundry operations.

(a) 4.5-kg plutonium for alpha phase or unalloyed plutonium, density (d) > 16.5 g/cm^3;

(b) 6.0-kg plutonium for delta phase or, plutonium alloys, d $\leqq 16.5$ g/cm^3.

We recently evaluated these limits for application to electrorefining operations.

TABLE II

TYPICAL PLUTONIUM MATERIAL BALANCE, ELECTROREFINING,
IMPURE Pu-1 Wt% Ga FEED, 4-kg SCALE

Plutonium into Run (grams)		
Metal Feed	4000	
PuF_4	91	
Cathode	40	
Total		4131
Plutonium out of Run (grams)		
Pure Pu Product		
Ring	3300	
Cathode	40	
Residues		
Anode	400	
Salt & Crucible	391	
Total		4131

As the result of this evaluation, the 6-kg delta phase plutonium casting limit was adopted for the preparation of anode ingots for electrorefining.[9] Thus, if required, all plutonium electrorefining feeds would be alloyed in the vacuum casting operation to produce anode cylinders having a density $\leqq 16.5 g/cm^3$. Because most scrap metals are plutonium alloys, addition of more alloying elements usually will not be necessary. No mass constraints had to be placed on the pure alpha product ring because of its favorable annular geometry. Thus, the sequence of operations for a typical electrorefining run would be as follows. A maximum of 6 kg of delta-phase plutonium metal would be melted and cast into a cylinder having a diameter of approximately 7.3 cm. Because the casting loss is approximately 5%, the weight of this cylinder would be somewhat less than 5.7 kg. The metal would then be electrorefined. The yield in this process should be ∿85% and the product ring should weigh about 4.8 kg. Although this weight is 0.3 kg higher than the 4.5-kg alpha casting limit, it does not pose a criticality problem because of its annular geometry. Product ring masses do not constitute a criticality constraint in the 6-kg process. As stated above, the only mass constraint is on the anode feed cylinder.

The objectives of the present study were

(a) design and fabricate equipment for the 6-kg process,
(b) demonstrate the process, and
(c) implement the process in production operations.

B. Equipment Design

The basic configuration shown in Fig. 2 was to remain unchanged. The same resistance furnace and furnace tube would be used. [This decision permits us to use the same basic equipment for electrorefining (both 4- and 6-kg scales), oxide reduction, and pyrochemical operations such as molten salt extraction for americium removal.]

The 4-kg crucible geometry shown in Fig. 1 was adopted for the 6-kg process for the following reasons:

(a) use of the same geometry would minimize equipment and procedural changes, and

(b) the annular shape of the product collection zone is geometrically favorable.

Dimensions of the 6-kg crucible are given in Fig. 3. The crucible o.d. is 12.7 cm, which is the same as the diameter of our oxide reduction[7] and molten salt extraction crucibles. The volume of the anode cup is 555 cm^3 and the volume of the annular collection zone is 665 cm^3. The annular volume was designed 20% larger to reduce crucible surface effects on product metal coalescence.

The ceramic stirrer for the 6-kg process is also shown in Fig. 3. Initially, the stirrer had two impellers; however, the first experiment indicated we had a stirring problem so a third impeller was added.

The complete experimental assembly is shown in Fig. 4. Crucible and stirrers for the experiments were fabricated by the Los Alamos ceramic fabrication section, CMB-6.

III. EXPERIMENTAL RESULTS

The results of five experiments with the 6-kg crucible are given in Table III. Plutonium-gallium alloys having a density of <16.5 g/cm^3 were used as feed material in each run. Except for the first experiment, these ingots were relatively pure alloys. Impurities such as carbon, nitrogen, and silicon were at low values; see Table IV for purity data.

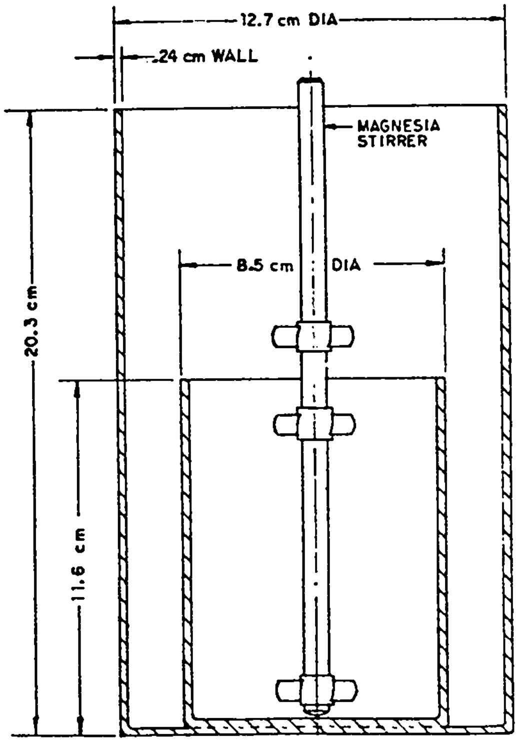

Fig. 3. Six-kilogram electrorefining crucible and stirrer.

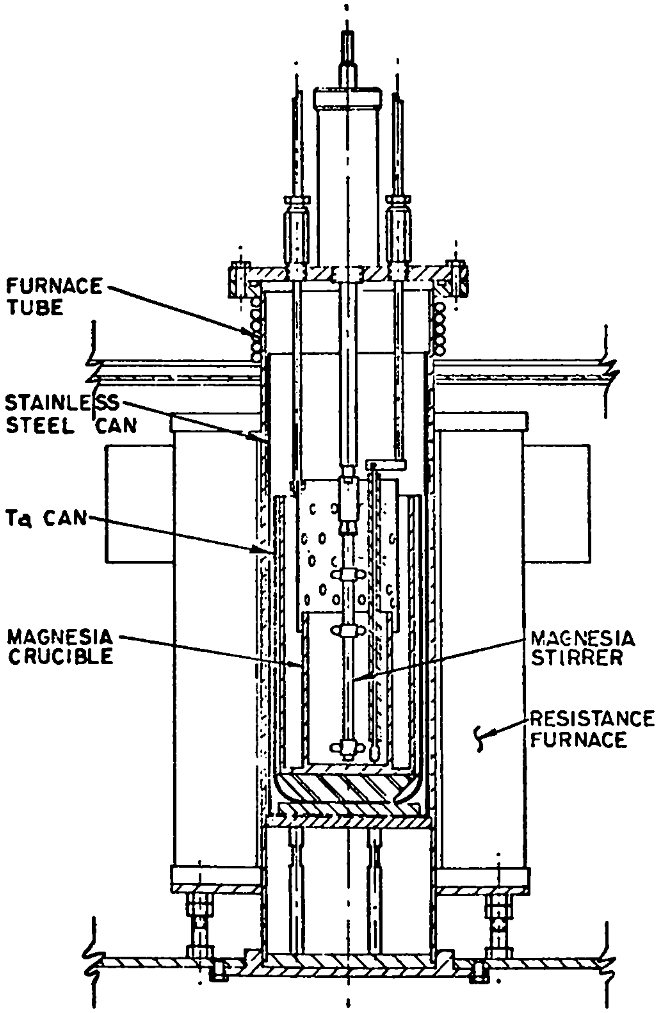

Fig. 4. Electrorefining process, 6-kg scale.

We used a ceramic stirrer with two impellers in the first run and obtained a product ring weighing 4.3 kg from a feed ingot containing 5.6 kg of plutonium. The electrorefining residues consisted of an anode containing 1.03 kg plutonium and a slag and crucible residue containing 0.40 kg plutonium. The large anode residue was the result of early termination of the run by electrode polarization. We thought this was caused by inadequate stirring. To improve the stirring, we used a three-bladed stirrer in the second run. The product yield in this run was exceptional. A 5.3-kg product ring was obtained from an alloy feed containing 5.89 kg of plutonium. The higher yield in this run was probably the result of both improved stirring and the use of a purer alloy feed. The three-bladed

TABLE III. SUMMARY OF 6-kg SCALE ELECTROREFINING EXPERIMENTS

Experimental Conditions

Feed Ingots -- Pu-Ga alloys, density <16.5 g Pu/cm^3

Temperature -- 800°C

Electrolyte -- (NaCl-KCl) plus PuF_4 or $MgCl_2$

Current -- 15- to 30-A dc

		Materials Into Run (kg)					
Run No.	Impeller Type	Metal	Pu	PuF_4, Pu	$MgCl_2$	Product Ring (kg Pu)	Product Yield (%)[a]
1	Double	5.755	5.625	0.091	0.000	4.289	76.2
2	Triple	5.950	5.890	0.035	0.000	5.314	90.2
3	Triple	5.816	5.700	0.000	0.066	5.103	89.5
4	Triple	5.425	5.370	0.000	0.066	4.739	88.2
5	Single Ta	5.513	5.380	0.035	0.000	4.703	87.4

[a] Product yield = $\frac{\text{g Pu in Product Ring}}{\text{g Pu in Metal Feed}} \times 100$.

TABLE IV
COMPARISON OF FEED AND PRODUCT PURITY

	Concentration, g Element per 10^6 g Plutonium									
	Run 1		Run 2		Run 3		Run 4		Run 5	
Element[a]	Feed	Prod.	Feed	Prod.	Feed	Prod.	Feed	Prod.	Feed	Prod.
Be	<1	<1	<1	<1	5	<1	2	<1	250	5
B	2	<1	<1	<1	<1	<1	50	<1	15	<1
Mg	300	<1	10	20	<1	5	<1	2	15	10
Al	110	<5	10	45	1500	<5	110	<5	100	<5
Si	40	<5	210	<5	75	<5	220	<5	130	15
Cr	80	<5	25	<5	35	<5	50	<5	185	<5
Mn	25	1	30	<1	2	<1	8	<1	25	1
Fe	530	20	200	35	200	5	46	<5	560	60
Ni	190	<5	100	<5	1500	<5	200	<5	180	6
Cu	5	<1	10	<1	5	<1	5	<1	10	5
Zr	<100	<100	>500	<100	<100	<100		<100	<100	<100
Mo	10	<3	25	<2	<3	<3		<3	5	<3
Ag	<1	2	2	<1	<1	5	<1	<1	<1	<1
Ta	<100	<100			<100	<100		<100	<100	<20
W	<10	100			50	100		<10	100	200
Pb	10	<5		<5	5	<5	10	<5	7	<5
C	310	25	30	10	150	10	160	5	380	65
Am	468	148	164	42	217	31	742	90	494	130
U	90				45	7		<5	55	20
Th	<10	<10		10	10	<10		<10	10	<10
Ga, wt%	1.10	0.0110	1.02	0.0014	1.01	0.0016	1.05	0.0005	1.27	0.0008
Pu, wt%	98.74	99.95	98.9	99.97	98.70	99.97	98.88	99.99	98.70	99.94

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[a] Elements below limits of detectability in both feeds and products include Li, Na, K, Ca, Ti, U, Co, Zn, Rb, Sr, Y, Nb, Ca, Sn, Ba, La, Hf, Re, and Bi.

ceramic stirrer was also used in runs 3 and 4, and rings weighing 5.1 and 4.7 kg were obtained. The ring from run 3 is shown in Fig. 5. Anhydrous magnesium chloride was used in place of PuF_4 in runs 3 and 4. Its use affected neither product yields nor product purity. The yields from the three runs using the three-bladed ceramic stirrer were 90.2, 89.5, and 88.2%. Product purity in all cases was good. We also investigated the use of tantalum stirrers because of the uncertainty of obtaining ceramic stirrers commercially. From early work,[10] we knew that bare tantalum stirrers immersed in the anode pool were dissolved during electrorefining operations. Protection of anodic metallic parts by ceramic insulators, however, permits the use of tantalum and tungsten. For example, the insulated tungsten anode rod of the 4-kg refining cell is not attacked. Protecting the impellers of metallic stirrers, however, is difficult. Conventional

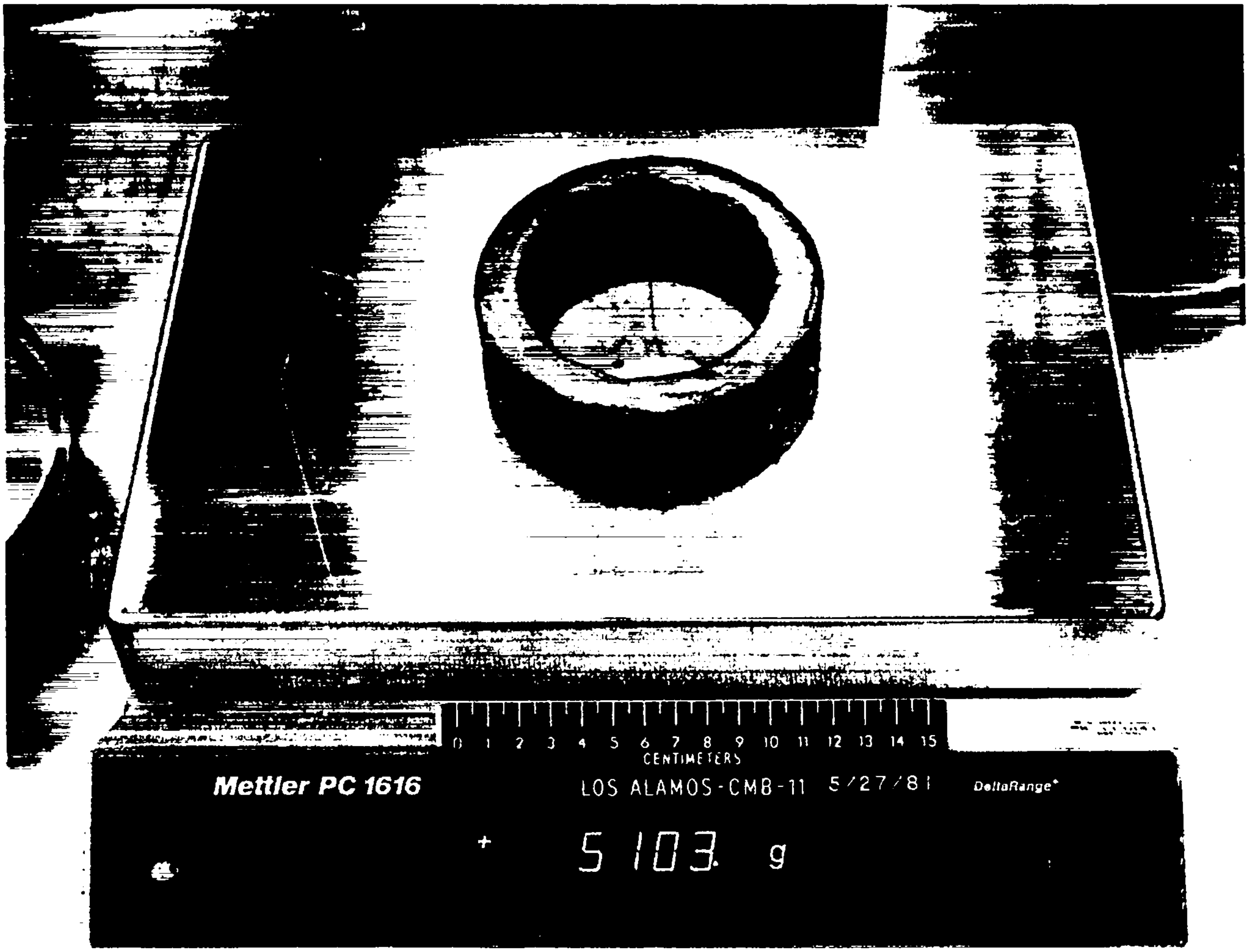

Fig. 5. Plutonium product ring, run 3.

ceramic coatings provide limited protection at best. Another possible alternative is to keep the metal impeller immersed in the anode. The stirrer shown in Fig. 6 was designed with this in mind. The Ta-10 wt% W shaft is protected by a vitrified magnesia sleeve. The single tantalum impeller and nut are immersed in the anode. Experiments with 6-kg crucibles using mercury to simulate plutonium metal and water to simulate the molten salt demonstrated that excellent mixing of each phase was achieved at relatively low speed, 400 rpm. This stirrer was used in run 5, Table III, with excellent results. An 87.4% yield of pure product, 99.94% plutonium, was obtained from a feed containing 98.70% plutonium. Back-emf measurements made during run 5 confirmed the results of the mercury stirring experiments; that is, mixing was good.

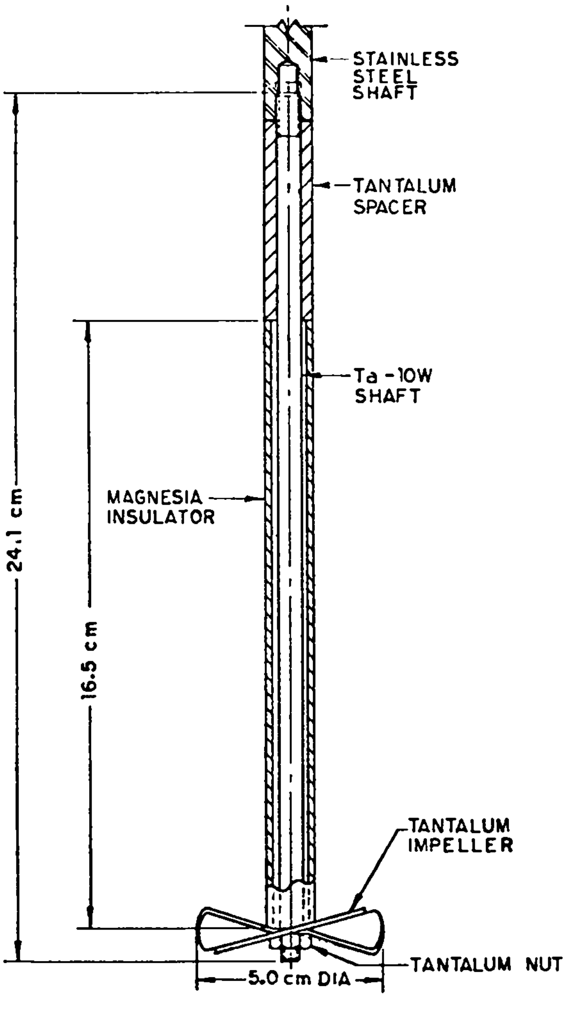

Fig. 6. Tantalum electrorefining stirrer.

IV. PROCESS IMPLEMENTATION

The success of the first 6-kg experiments led to the immediate decision to incorporate the process into production operations. First, items such as crucibles and ceramic stirrers were ordered from commerical suppliers. Concurrent with this procurement, production support equipment such as gloveboxes and electrical control equipment was designed to accommodate the larger process. Fig. 7 shows the redesigned, compact control console that contains all of the electrorefining support equipment. This console contains a microprogrammer, a back-emf sampling unit, a strip-chart recorder, an ac furnace power supply, and a dc power supply. Fig. 8 shows a typical glovebox with all support equipment. A furnace well, below the glovebox on the left, houses the resistance furnace, furnace tube, and other processing hardware. The electrical control console is below the glovebox, on the right. The stirring motor and drive units are outside the glovebox. Keeping the equipment out of the actual box provides a clean and clutter-free work area within the glovebox.

After the procurement, fabrication, and installation of all the redesigned equipment, the new 6-kg scale electrorefining process was put into operation. From the start-up date, a total of 256 runs have been made.

Of the 256 runs, we experienced 41 failures resulting from equipment breakdown. Some of the problems included

loss of facility power,
loss of cooling water pumps,
failure of control console components,
broken ceramic stirrers, and
broken cathode shields.

We completed 215 successful runs or 84% of those attempted. The plutonium metal from the aborted runs was recast into anode cylinders for new runs.

The feed metal to the process was from a wide variety of sources. Consequently, the quality of the feed metal was very inconsistent. The average size feed ingot cast for the operation was 5645 g of bulk weight. The average purity of this material was 98.5%, or 5560 g of the ingot was plutonium.

The average product yield during the start-up period was 71%. After complete equipment tests, training of operators, and establishment of operating procedures, the product yield has increased so that approximately 82% of the feed plutonium now results in product. A typical material balance for recent operations is given in Table V. The plutonium feed materials consist of 5.56 kg

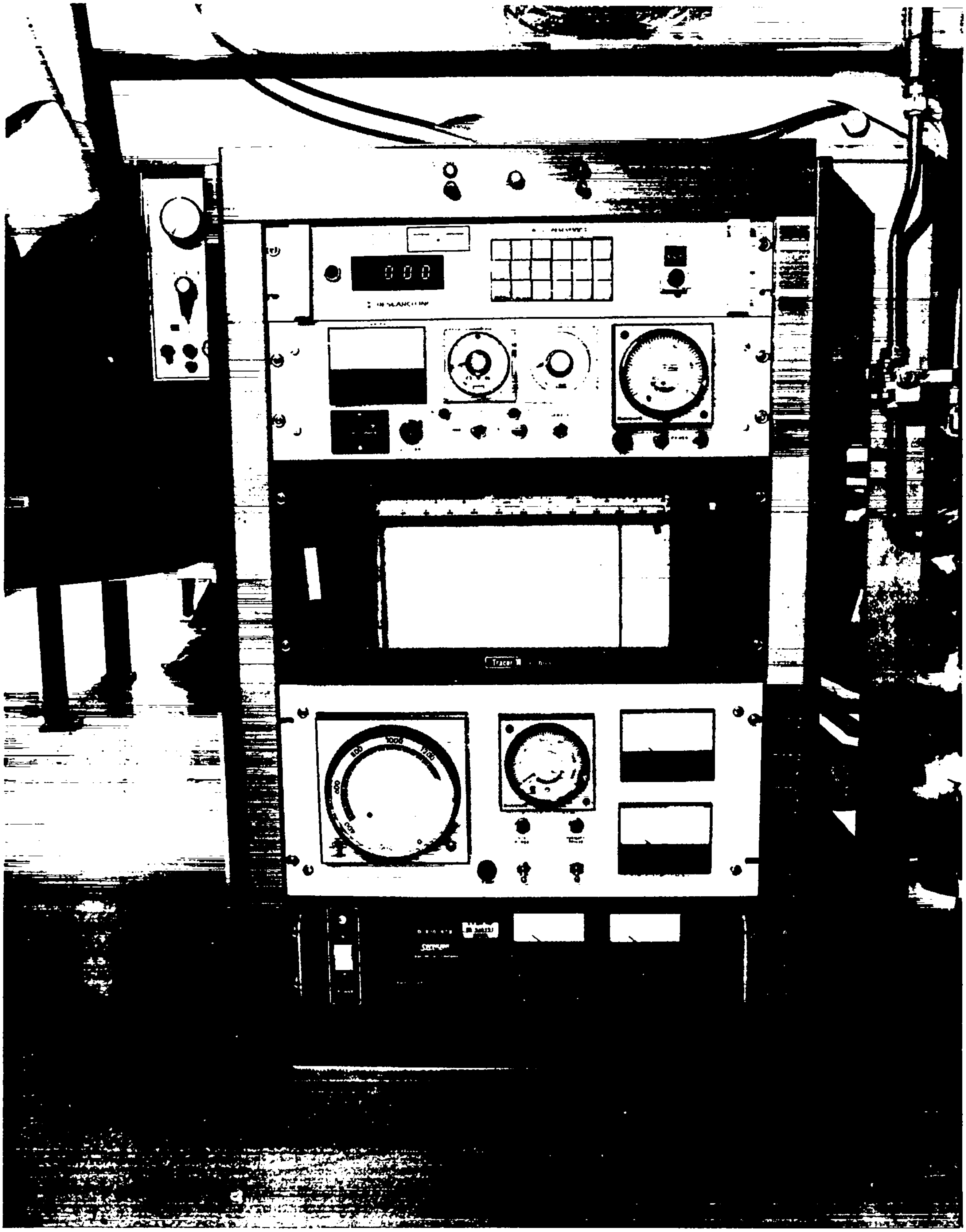

Fig. 7. Electrorefining console.

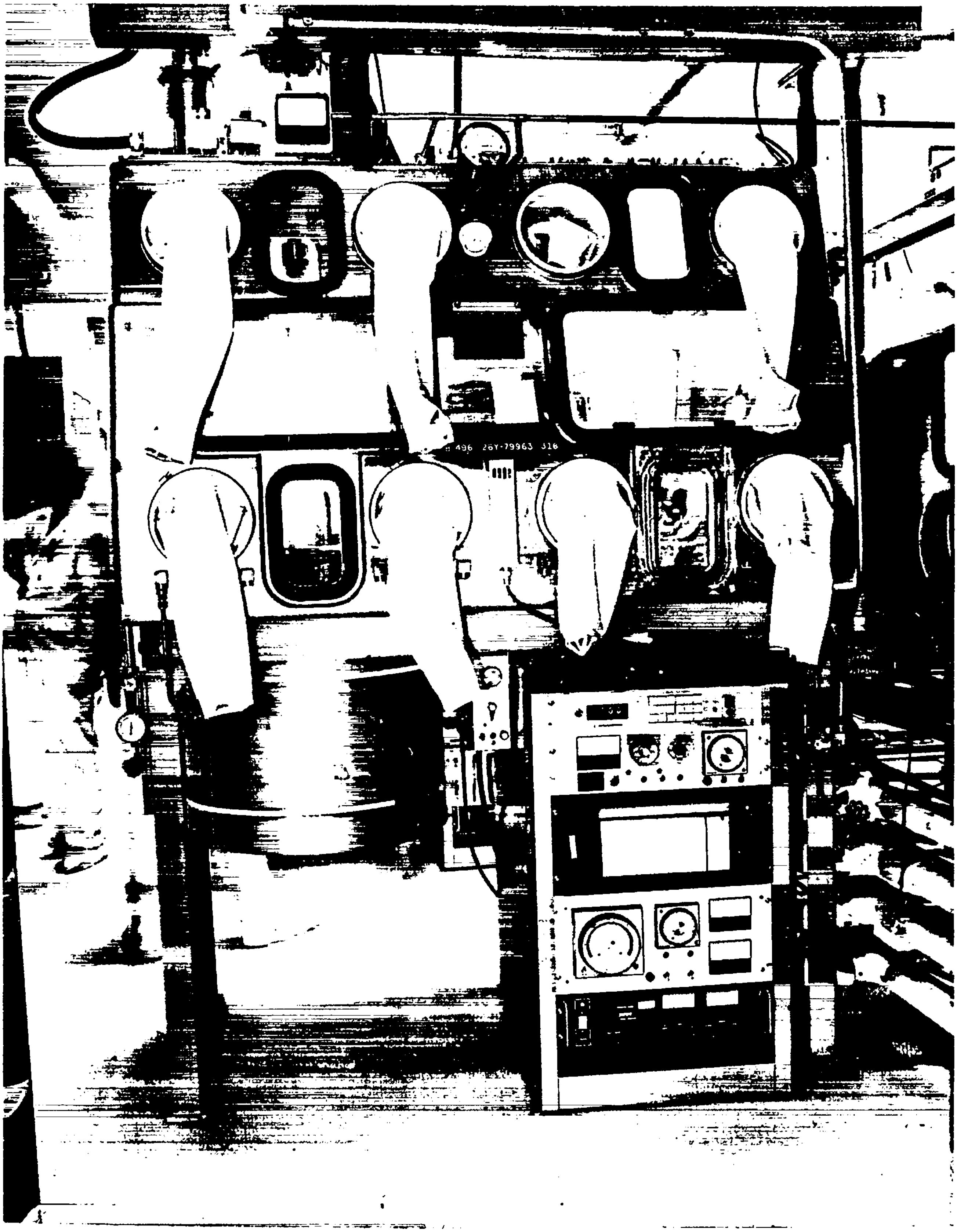

Fig. 8. Electrorefining glovebox.

of plutonium in the feed cylinder and 0.04 kg on the cathode from an earlier run. The material out of the run consists of a 4.56-kg plutonium product ring, 0.04 kg on the cathode, and 1.00 kg in residues. The average purity of this product is 99.96% pure. A total of 791 kg of this pure product has been produced since the process went into the production sequence. Table VI compares purity of the anode casting feed with the electrorefined product. The table again demonstrates the excellent purification that can be achieved in the electrorefining process.

TABLE V

TYPICAL PLUTONIUM MATERIAL BALANCE, ELECTROREFINING, IMPURE Pu-1 WT% Ga FEED, 6-kg SCALE

Plutonium Into Run[a] (grams)		
Metal Feed	5560	
Cathode	40	
Total		5600
Plutonium Out of Run (grams)		
Pure Pu Product		
Ring	4560	
Cathode	40	
Residues		
Anode	600	
Salt & Crucible	400	
Total		5600

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[a] Sixty-five grams of $MgCl_2$ are added to the NaCl-KCl salt casting.

TABLE VI

COMPARISON OF PURITIES OF ANODE CASTING FEED AND METAL PRODUCT

	g Element/10^6 g Sample	
Element	Feed(JAM 2113)	Prod(ERP8-14)
Be	<1	<1
B	200	<1
C	810	5
Na	70	15
Mg	10	2
Al	0.19%	<5
Si	290	<5
Ca	0.10%	<3
Cr	330	<5
Mn	10	1
Fe	0.11%	10
Ni	330	<5
Cu	3	<1
Zn	<5	<5
Ga	1.40%	35
Zr	<100	<100
Mo	50	<3
Ag	<1	<1
Cd	<10	<10
Pb	10	<5
Am	322	56
Pu	97.70%	99.98%

V. CONCLUSIONS

Plutonium electrorefining has been an effective processing tool for recycling metallic scrap since 1964. It produces a high purity metal product with a minimum investment in labor. The recent development and implementation of the 6-kg process permit us to almost double our product output with no compromise in yields or product quality.

ACKNOWLEDGMENTS

The development of the 6-kg electrorefining process would not have been possible without the advice, cooperation, and criticality evaluations provided by D. R. Smith and T. McLaughlin of H-6, Los Alamos National Laboratory. These contributions were an essential part of the process development studies.

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