MASTER

PLUTONIUM ELECTROREFINING AT ROCKY FLATS

THE DOW CHEMICAL COMPANY
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U.S. ATOMIC ENERGY COMMISSION
CONTRACT AT(29-1)-1106

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PLUTONIUM ELECTROREFINING AT ROCKY FLATS

Jack L. Long and Robert D. Schweikhardt

Abstract. Development work at the Rocky Flats Plant in plutonium electrorefining has led to the completion of a practical six-cell facility. In addition, a new laboratory for research and development work on electrorefining and other high temperature processes has been completed. Process developments, including purification, efficiency, and mechanical improvements, are discussed.

An electrorefining process-cost estimate predicts a figure of 9.7 direct man-hours per kilogram of plutonium metal. This is thirty percent less than cost comparisons in aqueous processing.

INTRODUCTION

Historical Background: Plutonium electrorefining work was initiated at Los Alamos Scientific Laboratory, Albuquerque, New Mexico, in 1957. The initial efforts at Rocky Flats started in 1958. The first known collection of high purity plutonium-metal product from electrorefining was at Rocky Flats in 1959, but the metal was in the form of individual globules of about $\frac{1}{8}$ - to $\frac{1}{2}$ -inch diameter. The first massive electrorefined plutonium was obtained from a batch operation at Los Alamos and their patent application was filed in 1961 (1).*

From February through December 1961, efforts at Rocky Flats were aimed at running a large (10-inch diameter) cell which would operate continuously for a period of one week with a minimum throughput of about $\frac{1}{3}$ kilogram (kg) per hour. After development of the cell, ten runs were completed. The electrolyte (for the large cell and the earlier work) was cesium plutonium hexachloride (Cs_2PuCl_6) in mixed alkali chlorides. Because of the ease in preparation (2), the anhydrous Cs_2PuCl_6 was used instead of plutonium trichloride (PuCl_3), plutonium trifluoride (PuF_3), or plutonium tetrafluoride (PuF_4). Additional facilities would have been required also to obtain sufficient anhydrous plutonium halide at Rocky Flats. The cell was operated in an inert atmosphere with stirring of the salt

phase. While some of the techniques required for continuous operation were performed; the overall objective of running continuously at the desired rate for at least one week was not achieved. Many equipment and operational problems were encountered. The most serious were: (a) Limited temperature range for successful operation of the product-removal system; (b) Poor plutonium-metal recovery (less than 70 percent); (c) Extensive chemical recovery requirements; and (d) Glovebox manipulation of all components.

Although operation of the product-removal system was demonstrated, the percentage of failures was greater than could be tolerated for production purposes. The system included a tantalum tube heated by a resistance element. The molten plutonium was withdrawn from the cell by the application of vacuum to a tube end, which opened to a chill mold. The failures were predominantly either tube freeze-up or heating element burn-out.

The poor plutonium-metal recovery had three possible causes: (a) Noninert cell atmosphere from leaks or outgasses; (b) Side reactions in the salt phase, such as $3\text{Cs}_2\text{PuCl}_6 + \text{Pu}^0 \rightarrow 6\text{CsCl} + 4\text{PuCl}_3$; and (c) Plutonium-metal reaction with porous magnesium oxide (MgO) cells. A few runs were made in tantalum cells with improved, but still unsatisfactory plutonium-metal recovery (about 80 percent).

The chemical-recovery problems arose from salt penetration of the MgO cells (making it necessary to pulverize the ceramic fragments before leaching) and from the size of the cell which required about 10 kg of salt per run. Attempts to obtain nonporous MgO cells of 10-inch diameter were unsuccessful. Cell setup and breakout proved difficult in glovebox operations because of the large cell equipment. Two men were required for most of the work.

While the 10-kg cell experiments were in progress at Rocky Flats, Los Alamos workers had demonstrated successful operation of a 3.5-kg cell (3). After ten unsuccessful runs with the large cell, it was decided to adapt the smaller Los Alamos cell to current needs. Equipment was designed and

* See references.

installed in the existing 10-kg cell glovebox for a 3.1-kg cell operation. Plutonium trichloride was obtained from Hanford Facilities (Richland, Washington). The salt required distillation before use for electrorefining. The recovery of a distilled product usually amounted to less than 70 percent, based on weight of PuCl_3 feed.

From February 1961 to October 1963, 89 runs were made. The development period included emphasis on design and fabrication by the Rocky Flats Ceramics Laboratory of two types of MgO stirrers; cell instrumentation for unattended overnight operation; a device for measuring the height of molten-plutonium metal in the anode cup during electrorefining; purchase of MgO double crucibles from a non-Atomic Energy Commission source; successful electrorefining from a melt containing plutonium ions supplied by the reaction between plutonium metal and a salt mixture of sodium chloride-potassium chloride (NaCl-KCl); preparation of high purity metal for the United Kingdom; and an economic evaluation of electrorefining processing at Rocky Flats.

From October 1963 to September 1965, approximately 80 electrorefining runs were made. The major work efforts included continued pure metal supply; design and purchase of a prototype two-piece double crucible; design and operation of a prototype cell; and contributions to the final design of an electrorefining plant. The efforts led to a patent (4) for the *in situ* preparation of PuCl_3 via reaction of plutonium metal with magnesium chloride (MgCl_2).

Recent Developments: A general description of the present batch-electrorefining process follows:

A cast slug of impure plutonium metal, weighing approximately 3 kg, and a 1.3-kg block of NaCl-KCl-MgCl_2 salt are loaded into a ceramic double crucible. The impure metal is in the small inner crucible that is seated concentrically within a larger crucible. The loaded crucible is placed into an electrorefining cell and heated in an argon atmosphere within the cell. When the phases are molten, the inner cup is completely submerged under a cover of the electrolyte salt. Electrodes and a stirrer are lowered into the cell. A DC current is passed between the impure metal anode and a tungsten cylinder cathode, which fits into the annular space between the two crucibles. During electrorefining, most of

the impurities remain in the inner crucible, and pure plutonium is collected in the annular space between the larger and smaller crucibles. At the end of the run, the cell is cooled and the products are separated (refined metal, anode heel, and salt and ceramic residues).

Currently at Rocky Flats, the plutonium electrorefining capability is being expanded. For several years, one 3.1-kg cell had been available for development work; in December 1964, a second cell, a prototype of the production cell, was installed in the same glovebox.

The expanded electrorefining capability, now completed, consists of six production cells located in a new addition to the production building. In addition, there is a laboratory for research and development work on electrorefining and other high temperature processes for plutonium. The balance of this report will describe recent developments in the electrorefining process and equipment, the new facilities, and give a processing cost estimate for plutonium-electrorefining at Rocky Flats.

DISCUSSION

Process Developments:

MAGNESIUM CHLORIDE (MgCl_2) IN SALT – During experiments on the extraction of americium from plutonium metal by molten salts, it was observed that the addition of MgCl_2 to the equimolar NaCl-KCl salt resulted in a more rapid extraction of americium and plutonium ions into the salt (5). This suggested adding MgCl_2 to the electrolyte salt in order to provide the plutonium ions necessary to start electrorefining.

To date, over 50 electrorefining runs have been made using MgCl_2 in equimolar NaCl-KCl as the starting salt. Most of these runs have been made using 2.5 weight percent (wt %) MgCl_2 ; higher concentrations were tried but did not offer any advantage. At the beginning of the process, after the metal and salts have become molten, the MgCl_2 reacts with the impure plutonium metal to form plutonium ions in the electrolyte salt by the exchange reaction: $3\text{Mg}^{++} + 2\text{Pu}^0 \rightarrow 2\text{Pu}^{+++} + 3\text{Mg}^0$. The plutonium ions necessary to start electrorefining are produced within one hour after the system reaches the molten state. Some of the magnesium metal distills off; however,

an increase of about 50 parts per million (ppm) in magnesium content of the electrorefined metal is noted. For most uses of electrorefined metal, the increase is not detrimental because the magnesium content is lowered when the electrorefined plutonium is vacuum-cast. Table I gives comparative data on electrorefining runs made with and without MgCl_2 in the electrolyte.

There are several advantages to using MgCl_2 to provide plutonium ions to start electrorefining. It eliminates the need to add PuF_3 or PuCl_3 to the starting salt (either of these compounds also act to provide plutonium ions). The facilities needed to produce and maintain pure, dry PuCl_3 or PuF_3 are unnecessary. The complete starting salt may be prepared in a noncontaminated area. The addition of MgCl_2 to the NaCl-KCl mixture prior to casting the starting salt results in a smooth-cast salt block that releases from the quartz preparation mold easier than if the MgCl_2 were not present. The addition of MgCl_2 results in a lower melting electrolyte (about 660°C for NaCl-KCl versus about 650°C for NaCl-KCl , 2.5-wt % MgCl_2).

PURIFICATION — One of the uses of electrorefining is to provide high purity plutonium metal for research work. To meet higher purity requirements, several batches have been double electrorefined to a higher purity than normally possible with a single pass. For example, one of the better double electrorefined products had the following analysis in ppm of the common impurities:

Aluminum	9	Magnesium	10
Americium	8	Silicon	< 2
Carbon	11	Tantalum	<25
Chromium	< 5	Tungsten	<25
Iron	10		

These represent total decontamination factors between the feed metal and the double electrorefined metal ranging from 5 to 85. This approximate range is for the common impurities, except magnesium. There has not been a sufficient number of double electrorefined runs made to establish total decontamination factors for specific impurities and conditions.

Americium is produced in plutonium metal from the beta decay of plutonium 241 (Pu^{241}). The approximate increase in americium content is given by the equation:

$$(\text{Percent Pu}^{241}) (\text{Number of days since americium analysis}) (1.4) = (\text{ppm increase in americium content}).$$

If americium is the primary contaminant to be removed, this may be done by a molten-salt extraction of americium from plutonium metal in electrorefining equipment and with electrorefining salt (5). Molten-salt extraction can be accomplished in a fraction of the time that it takes for a complete electrorefining run. For removing americium from plutonium metal, an extraction followed by electrorefining is about equivalent to two batch extractions. The average decontamination factor (DF) of americium by electrorefining was found to be 4.5 for 25 runs in which the initial americium content of the metal ranged from 36 to 1150 ppm:

$$\text{DF} = \frac{(\text{Impurity concentration in feed})}{(\text{Impurity concentration in product})}$$

TABLE I. Plutonium Electrorefining Runs with and without 2.5 Weight Percent Magnesium Chloride (MgCl_2) in the Starting Equimolar Sodium Chloride-Potassium Chloride (NaCl-KCl) Electrolyte.

Runs with Magnesium Chloride			Runs without Magnesium Chloride		
Run No.	Product Efficiency (percent)	Product Magnesium (parts per million)	Run No.	Product Efficiency (percent)	Product Magnesium (parts per million)
36-C	77	25	52-B	68	< 2
39-C	89	50	53-B	72	< 2
1-A	82	50	57-B	83	< 2
2-A	70	30	59-B	76	< 2
27-B	89	60	64-B	79	2
31-B	81	25	65-B	70	< 2
32-B	73	30	68-B	59	< 2
33-B	84	25	66-B	60	10

Gaseous impurities are generally low in electrorefined metal. Typical values are: less than 3 ppm H_2 ; less than 3 ppm N_2 ; and less than 15 ppm O_2 . These values apply to metal which has been exposed to air for up to several hours. If the electrorefined product could be protected from oxygen (O), it is likely that the O_2 content would drop to less than 3 ppm.

EFFICIENCY – The time to terminate an electrorefining run may generally be calculated using the theoretical electrorefining rate 2.97 grams of plutonium per ampere hour (g Pu/amp-hr), or by measuring the electrical characteristics of the cell during the run (6). In 12 recent electrorefining runs, the actual yield of electrorefined plutonium was within 3 percent of the theoretical electrochemical equivalent weight. Therefore, the end time may be determined based on the initial purity of the feed metal, the desired product weight and purity, the electrorefining current, and an amp-hr calculation. The resistance [and the back electromotive force (emf)] of a cell gradually increases as the plutonium anode is depleted and rapidly increases when complete practical depletion has occurred. Although workable electrical settings for indicating the end points of runs have been used, the optimum values for different electrorefining feeds were not determined.

The above methods for run termination are less effective if the feed metal contains much aluminum and/or carbon. These elements form high melting compounds with plutonium which are less dense than plutonium metal. Therefore, as the anode is depleted, more and more of the metal surface is covered with a solid compound, and eventually part of the anode process will be the dissolution of the solid compound. Different back emf effects have been observed as the liquid-plutonium anode surface becomes covered. These ranged from a rapid rise with more than a thousand grams of metal remaining in the anode to only a gradual rise with an anode essentially depleted of a metallic phase. No systematic investigation of the effect of aluminum and carbon has been made. Based on the limited available run data however, it would appear that interference with run termination can be expected if either or both elements is in the range of 0.05 to 0.1 percent or greater, in the feed metal.

Mechanical Developments:

Several mechanical features of batch electrorefining have been developed during the last two years; the most important of these will be described.

FURNACE – A hinged-top cell was constructed and tested. The cell has a hinged upper section which is cooled with water or Freon refrigerant and a fixed lower section which is resistance-heated and holds the electrorefining crucible. A drawing of the cell is seen in Figure 1. Some of the advantages of this type of cell over a top-loading cell are its compact design, ease of operation (particularly loading, unloading, and cleaning), and the external heating elements (the elements remain uncontaminated for easy replacement and disposal).

CRUCIBLE – The double-crucible design was changed. The former design consisted of a double-cup MgO crucible in which the smaller cup was cemented within the outer cup. The ceramic manufacturers had problems however in getting a good bond of the inner to the outer cup, and it was difficult to test the bond. Several times a cemented double-cup crucible would pass the bond test, but the bond would fail during the electrorefining process. When this happened, the loose inner cup usually broke the ceramic stirrer and the run had to be stopped. Stirrers have been replaced during electrorefining, but this is undesirable. The new crucible design, Figure 2, calls for an inner cup that fits easily into a recess in the bottom of the outer cup. With this design, the inner cup is held in place first by the weight of the anode charge, and later during electrorefining by the slight pressure of the anode rod. The pressure keeps the cup from floating in the product metal as it is collected in the annular space between the two cups. After termination of electrorefining, the anode rod is raised and the product metal collects as a disk in the bottom of the outer crucible. The design has been tested and found satisfactory in 8 electrorefining runs. Since the new design requires one less firing than the cemented double cup and there is no reject loss because of improper cementing, fabrication costs are less. Savings up to 40 percent compared to the cost of cemented crucibles have been realized depending on the manufacturer and the numbers ordered. Shutdowns because of loose cups have been eliminated as has the need for adherence testing of the cemented double crucible. In addition, this type of crucible is easier to load. Placing the loaded inner cup into the outer cup is less difficult than to place the feed slug into the cemented inner cup.

STIRRER – The Rocky Flats Ceramics Laboratory developed a mold and procedure for isostatically pressing MgO-stirrer blades which may represent a substantial savings over slip-casting stirrer blades.

FIGURE 1. Schematic Drawing of Production Prototype Cell.

Legend: (Parts renumbered and some omitted; taken from original Dow Chemical Drawing No. 13517-1, Sheet 1 of 9, Revision B.)

Part	Number Required	Description	Material
1	12	Hex-Nut 1/4-20 UNC.	Stainless Steel
2	Reference	Current Lead Assembly.	
3	2	Collar Set 1/4 Boston No. SC-25.	Commercial Steel
4	2	Electrical Seal Gland Assembly for 1/2-Inch Conductor-Conductor Omitted Conax No. EGT-250-A/-T with 3 Extra Seals of Teflon.	
5	1	Hose (Weatherhead) No. H-138 1/2 ID by 4-Feet 0-Inches Long.	Neoprene
6	4	Female Pipe Conn. (Parker) 6-6 GBTX-B.	Brass
7	4	Male Hose End (Weatherhead) No. 10008-106.	Brass
8	1	Hose (Weatherhead) No. H-138, 1/2 ID by 18 Inches Long.	Neoprene
9	1	Coupling, Standard 1/4 Inches.	Steel
10	1	Tube Assembly, 1-Inch OD by 0.065 Wall, Copper Tubing with Parker No. 16-12 FBTX Male Connector Both Ends, 4-Feet, 0-Inches Overall Length.	Copper
11	2	Head, Screw Cover Complete Conax No. T8 for 1/2-Inch NPT.	Commercial
12	2	Electrical Seal Gland Assembly for 1/4-Inch Conductor-Conductor Omitted Conax No. EGT-250-B-L with 4 Extra Lava Seals.	
13		As Required Norton 33-1 Castable Refractory Cement.	Alundum
14	8	Washer, Flat 1/4-Inch Standard.	Stainless Steel
15	Reference	As Required Thermocouple Extension Wire Chromel-Alumel 20 Gage, Asbestos and Fiberglass Insulated, Minneapolis-Honeywell No. 5W2J36.	Commercial
16	1	Collar, Set 1/2 Boston No. SC-50.	Commercial Steel
17	1	Motor, 1/50 HP, Shunt Wound, 115 V(DC), 1725 RPM-FRAME No. NSH-12, Bodine No. 2150, W/Speed Control, Model SH-12, 5-Foot Cable and Amphenol Connector. Minarik Electric.	Commercial
18	4	Screw, Hex HD 10-24 UNC by 3/4 Inches Long.	Stainless Steel
19	4	Washer, Lock 3/16 Standard.	Stainless Steel
20	2	Screw, Hex HD 1/4-20 UNC by 3/4 Inches Long.	Stainless Steel
21	2	Washer, Lock 1/4 Standard.	Stainless Steel
22	1	Coupling-Lovejoy No. L3 "Unijoint" 5/16-Inch Bore with Set Screws.	Commercial
23	1	O-Ring, 5.100 ID by 0.275 Wall-Parker No. 2-430.	Silicone Rubber (No. S418-6 Duro 60)
24	1	O-Ring 9.475 ID by 0.275-Inch Wide-Parker No. 2-448.	Silicone Rubber (No. S418-6 Duro 60)
25	3	Hinge Clamp Assembly-American Tooling No. ADB-21350.	Commercial
26	12	Washer, Lock 1/8 Standard.	Stainless Steel
27	12	Nut, Hex 3/8-16 UNC.	Stainless Steel
28	1	Sparging Unit No. MBH2084-2 (316LPSS) Pass, Trinity Micro Corporation.	Stainless Steel
29	4 lb	"Fiberfrax" Bulk Ceramic Fiber.	Commercial
30	As Needed	Insulating Brick, Harrison-Walker No. K-20.	Fire Brick

FIGURE 2. New Design Showing Outer (a) and Inner Crucibles (b).

Impurity	Legend	
	Maximum Content (aluminum-oxide cell) (approximations)	Weight Percent (magnesium-oxide cell) (approximations)
Aluminum Oxide (Al_2O_3)	99.65	0.35
Boron (B)	0.002	0.01
Beryllium (Be)	0.0002	0.0015
Calcium Oxide (CaO)	0.10	1.0
		(silicon per calcium)
Chromium (Cr)	0.1	0.1
Copper (Cu)	0.002	0.002
Iron Oxide (Fe_2O_3)	0.10	0.15
Manganese (Mn)	0.002	0.002
Sodium Oxide (Na_2O)	0.08	-
Silicon Oxide (SiO_2)	0.10	0.3 ^a
Titanium (Ti)	0.002	0.002
Carbon (C)	0.01	-
Magnesium Oxide (MgO)	0.10	b

^a In special cases, up to 1.5 percent of silicon oxide (SiO_2) would be acceptable subject to approval by the Dow Chemical Company.

^b The percent magnesium-oxide (MgO) content to be greater than 98.0 percent, minus the percent-sintering agent. Sintering-agent content and type also subject to Dow approval.

- NOTES: 1. Apparent material porosity for each assembly shall be less than 0.2 percent, as measured by American Society for Testing Materials procedure C-20-46. Each unit shall be leak-tested by filling with water to $\frac{1}{2}$ inch from top for a period of $\frac{1}{2}$ hour. (Inner crucible to be tested prior to testing of entire double crucible assembly.) Each unit to be certified to meet porosity and leak-check requirements.
2. Unless otherwise specified, these materials shall meet the Atomic Energy Commission high purity oxide, refractory-chemical purity specifications (AEC-SR-101 CORO-1), January 1, 1958.
3. Inner crucible must seat fully in recess without interference.

- Legend
- 1 Temporary Bulkhead.
 - 2 Conveyor.
 - 3 Entry Box.
 - 4 Air Lock.
 - 5 Breakout Box.
 - 6 Storage Box.
 - 7 Control Console.
 - 8 Conveyor Takeup.
 - 9 Charge Preparation.
 - 10 Vacuum Pumps.
 - 11 Hood.
 - 12 Instrument Panel.
 - 13 Vacuum Oven.
 - 14 Power Panel.
 - 15 Argon Purification.
 - 16 Argon System.
 - 17 Electrorefining Cells (6).

FIGURE 3. Layout of New Production Electrorefining Facility. (Scale for above layout is about $\frac{3}{16}$ inches = 1 foot.)

The pressed stirrer has two pairs of pitched blades—one pair to stir the metal phase, and one pair to stir the salt phase. The design has been tested and is satisfactory.

Several modifications were tested on the production prototype and adopted for the production cells. These modifications include a $\frac{1}{2}$ -inch diameter hollow-stirrer shaft of stainless steel, a new stirrer-bearing assembly, a stainless steel crucible holder, and a tantalum safety liner (to fit within the lower section of the cell chamber).

New Facilities:

Previous development work has led to establishment of new facilities at Rocky Flats: one is the six-cell production electrorefining facility; the other is a laboratory for continued electrorefining development and for related high temperature process development.

PRODUCTION FACILITY — Shown in Figure 3 is a layout of the new production electrorefining facility,

now undergoing initial testing. This facility will be operated by the Metal Production Department to supply high purity plutonium metal from regular production-purity plutonium metal or from impure recycle metal.

In addition to the above, a facility is being constructed for the dissolution and ion-column separation of plutonium from chloride residues, primarily chloride residues from electrorefining runs.

The electrorefining charge will be prepared in the charge-preparation box (Figure 3). The ceramic crucible with its charge is loaded into a stainless steel can which acts as a conveyor container and as a holder within the electrorefining cell. The loaded container is removed by the continuous single-track overhead conveyor to the cell to be loaded. After electrorefining, the container is removed from the cell and conveyed to the breakout box, which may be operated with or without an inert atmosphere.

The six-cell facilities will have a nominal capacity of 100 kg of electrorefined plutonium metal per month. Six more cells can be installed in the space provided

Legend

- 1 Take-in Boxes.
- 2 Bag-out Ring.
- 3 Fire Extinguishers.
- 4 Hot-stage Microscope.
- 5 Wet Chemistry Box.
- 6 Spectrophotometer.
- 7 Electrorefining Cells (E-R)
(3.1-kg, E-R).
- 8 Spectrophotometer Furnace Control.
- 9 Box Atmosphere Analyzer.
- 10 Controls (E-R).
- 11 Furnace Power Supply (levitation).
- 12 Cell Atmosphere Analyzer.
- 13 Electrorefining Cells (E-R)
(0.5-kg, E-R).
- 14 Levitation Furnace.
- 15 Furnace Control (levitation).
- 16 Safety Shower.
- 17 Open Hood.

FIGURE 4. Layout of New Pure-Metals Laboratory for Electrorefining and Related Studies.

in the existing loop (Figure 3). A second loop of 12 cells can also be installed in the building space now available. This would bring the total number of cells to 24. The actual capacity would depend on the criticality batch limit (now set at 3.5 kg per cell), the purification desired, and the practical utilization of the equipment yet to be determined.

Starting with off-specification plutonium metal, the production facility will produce refined plutonium of less than 200 ppm total detectable impurities (if the americium-241 content of the feed is more than 225 ppm, a slagging run prior to electrorefining may be required to make metal with less than 200 ppm total impurities). Plutonium metal with less than 100 ppm total detectable impurities should be possible in the production facility by double electrorefining and careful handling.

LABORATORY FACILITY — Figure 4 shows the layout of the new laboratory for plutonium electrorefining and related studies.

The central glovebox line in Figure 4 is an argon-atmosphere line. The argon from this line is continuously filtered and sent to purification equipment located in another room. There the moisture and oxygen are removed and the argon is recycled to the glove line at a controlled negative pressure.

There are six cells (four large and two small) within the inert-atmosphere line. The four large cells will have a criticality batch limit of 3.1 kg of plutonium. The cells are similar to the production cells (see Figure 1) and will be used to develop and optimize production-scale electrorefining and pyrometallurgical processes and to make nonroutine electrorefining runs, if necessary. The two small cells (less than 500-g Pu) will be used to make small-scale initial investigations of electrorefining and related processes. These cells have high temperature glass in the upper sections which will permit observation during the experiment.

In addition to the six cells, the laboratory will have the following equipment and capabilities:

- (a) The Cary-14 Spectrophotometer will give information on the states present in various molten-salt systems by measurements of absorption spectra.
- (b) The Unitron-HTL high temperature microscope will permit observation and photographing of

radioactive specimens at temperatures up to 1500°C and magnifications to 1000X.

- (c) The Vacuum Industries, Series 2100, Model-1806 levitation furnace is contained in a glovebox to permit use of radioactive materials. The unit can suspend and melt a small charge of material by electromagnetic energy. The operation can be conducted in vacuum or in certain atmospheres so that purification by outgassing or gas-liquid reactions may be accomplished on molten materials without contamination from a crucible.

COST ESTIMATES

An estimate of 17.3 direct man-hours per kilogram of plutonium was previously made for electrorefining process costs. A revised process-cost estimate based on the six-cell production facility now available and the expansion data for a 24-cell facility are given as follows:

Assumptions:

- a. Services and supplies for electrorefining will cost no less than services and supplies for aqueous processing and no more than services and supplies for foundry operations.
- b. Six electrorefining cells will not be required to process more than 42 kg of plutonium feed per a 5-day week.

Basis:

- a. Feed charge will be 3.5 kg per run. This has been approved by the Nuclear Safety Group.
- b. Run failures will be 10 percent or less. This percentage was experienced when the objective of the runs was the production of electrorefined metal.
- c. Six cells will process 42 kg of feed per a 5-day week and will provide 34.3 kg of electrorefined product. The ratio of product-to-feed is based on 17 runs. The same runs also provided the basis for breaking down 7.7 kg of plutonium cell residue to 4.4-kg plutonium in anode residues and 3.3-kg plutonium in salt and ceramics.

- d. The anode residue processing cost is based on converting the residue to oxide (0.67 direct man-hours per kg Pu) and converting the oxide to metal via the production process (12.5 direct man-hours per kg Pu). The burning and chemistry processing costs are based on actual production experience.
- e. The salt and ceramic processing costs are based on 55.4 man-hours for processing salts and ceramics containing 3.3-kg plutonium from 12 runs through the chloride line, plus the cost (10.0 direct man-hours per kg Pu) of peroxide precipitation, fluorination, reduction, and button breakout.
- f. The cost of casting anode feed is 1.14 man-hours per kg assuming that only one 3.5-kg casting is poured during one furnace cycle.
- g. Five men per day during a 5-day work week can operate six cells around the clock and also cast the required salt charges. This is an extrapolation of man-hours required for a one-cell operation.

Calculations:

The cost given is less than our previous estimate (17.3 direct man-hours per kg) because the feed charge has been increased from 3.1 to 3.5 kg, the cost of PuCl_3 preparation and distillation has been eliminated, and the cost of cell operation plus salt-casting has been reduced.

Operation	Man-Hours Per Week
Cell operation and salt casting	$5 \times 8 \times 5 = 200.0$
Metal casting	$42 \times 1.14 = 47.9$
Salt and ceramic processing	$55.4 + 33 = 88.4$
Anode-residue processing	$4.4 \times 13.2 = 58.1$

(Plutonium metal processed per week = 42-kg)

$$\begin{aligned}
 \text{Cost} &= \frac{200 + 47.9 + 88.4 + 58.1}{42} \\
 &= \frac{394.4}{42} \\
 &= 9.4 \text{ Direct man-hours per kilogram plutonium.}
 \end{aligned}$$

If the plutonium-metal feed contains certain impurities (i.e. in excess of 750 ppm of carbon or aluminum), the preceding estimated processing cost may be low because these elements reduce the amount of anode depletion at which a given product purity is attainable. If americium in the feed amounts to several hundred ppm or more, additional processing costs may be incurred to provide product metal containing less than 200 ppm total impurities.

Comparisons:

For comparison, the estimated aqueous-processing cost for oxide is 12.5 direct man-hours per kg plutonium or 13.4 direct man-hours per kg plutonium, if the burning cost is included (based on production figures obtained when this report was written). The 13.4 direct man-hours per kg-plutonium figure would drop to about 12.1 direct man-hours per kg plutonium, if the existing aqueous facilities were run at maximum capacity.

It should be pointed out that the cost of processing such things as washables, burnables, some percentage of laboratory wastes, plus sand, slag, and crucible, is not included in either the aqueous cost or in the electrorefining cost. Therefore, some additional cost would have to be added. The added cost would be proportional to the load. However, the added cost of sand, slag, and crucible processing is proportionately much less for electrorefining than for aqueous processing. Based on a 3-wt % plutonium loss to sand, slag, and crucible to be recovered at 60 direct man-hours per kg plutonium, the processing costs are increased by 0.3 direct man-hours per kg plutonium for electrorefining and 1.8 direct man-hours per kg plutonium for aqueous. The total cost for electrorefining is then 9.7 direct man-hours per kg plutonium and for aqueous is 13.9 direct man-hours per kg plutonium.

The given cost comparisons between aqueous processing and electrorefining are stated in terms of direct labor man-hours. The ratio of actual unit-dollar costs for these operations could differ from the direct man-hour ratios, if the proportions of indirect labor and overhead burdens to direct labor costs are different.

CONCLUSIONS

Development work at Rocky Flats has resulted in several contributions to the batch electrorefining process. These include a hinge-top cell design, a two-piece ceramic crucible, and an electrolyte that does not require the addition of a plutonium halide. These contributions have been included in the new production facility capable of producing over 100 kg per month of electrorefined plutonium.

It is estimated that the production electrorefining facility will process plutonium-metal feed for 9.7 direct man-hours per kg. This is 4.2 direct man-hours (30 percent) less than the lowest aqueous processing cost for comparable feed.

In addition to the production facility, a new laboratory facility has been completed which will be used for electrorefining development work and to develop new high temperature processes.

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