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LA-2981

**LOS ALAMOS SCIENTIFIC LABORATORY
OF THE UNIVERSITY OF CALIFORNIA ○ LOS ALAMOS NEW MEXICO**

**OPERATING INSTRUCTIONS, PROCEDURES, AND EQUIPMENT
FOR THE LOS ALAMOS PLUTONIUM ELECTROREFINING PLANT**

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Printed in USA. Price \$0.75. Available from the
Office of Technical Services
U. S. Department of Commerce
Washington 25, D. C.

LA-2981
UC-25, METALS, CERAMICS,
AND MATERIALS
TID-4500 (24th Ed.)

LOS ALAMOS SCIENTIFIC LABORATORY
OF THE UNIVERSITY OF CALIFORNIA LOS ALAMOS NEW MEXICO

REPORT WRITTEN: September 9, 1963

REPORT DISTRIBUTED: December 18, 1963

OPERATING INSTRUCTIONS, PROCEDURES, AND EQUIPMENT
FOR THE LOS ALAMOS PLUTONIUM ELECTROREFINING PLANT

by

L. J. Mullins
J. A. Leary
A. N. Morgan

This report expresses the opinions of the author or authors and does not necessarily reflect the opinions or views of the Los Alamos Scientific Laboratory.

Contract W-7405-ENG. 36 with the U. S. Atomic Energy Commission





ABSTRACT

The operating instructions, procedures, and equipment for the electrorefining of plutonium on the 3.5 kg. scale are described.

ACKNOWLEDGMENTS

We are indebted to S.D. Stoddard, D.E. Nuckolls, and R.E. Cowan of the Ceramics Section, Group CMB-6, for their assistance in the design of the ceramic components. W.D. McNeese, Group CMB-11, and E.P. Wolff, CMB-7, redesigned the stirrer assembly. J.F. Buchen, CMB-7, designed the circuitry for the automatic back e.m.f. sampling device.



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INTRODUCTION

Fused salt electrorefining processes have been developed for producing high purity, high density plutonium metal on the 500 g. and 3.5 kg. scales. The development of these processes has been described elsewhere.¹⁻⁵ The purpose of this report is to present in detail the operating instructions, procedures, and equipment recommended for use in the Los Alamos electrorefining plant now in construction.

The procedures are designed for personnel on an 8 hour day, 5 day week.

GENERAL SAFETY CONSIDERATIONS

Nuclear Safety

The maximum permissible plutonium content of one electrorefining glove box is 4 kg. (See Appendix B for nuclear criticality memoranda.)

Operational Safety

The procedures outlined in the CMB-11 Health and Safety Booklet⁶ are applicable. The automatic back e.m.f. sampling device is peculiar to the electrorefining process and should be used. This equipment cuts off the electrolyzing current in the event of a stirrer failure, thereby insuring that sodium metal is not generated in excessive amounts.

MATERIAL SPECIFICATIONS

Ceramic Cells

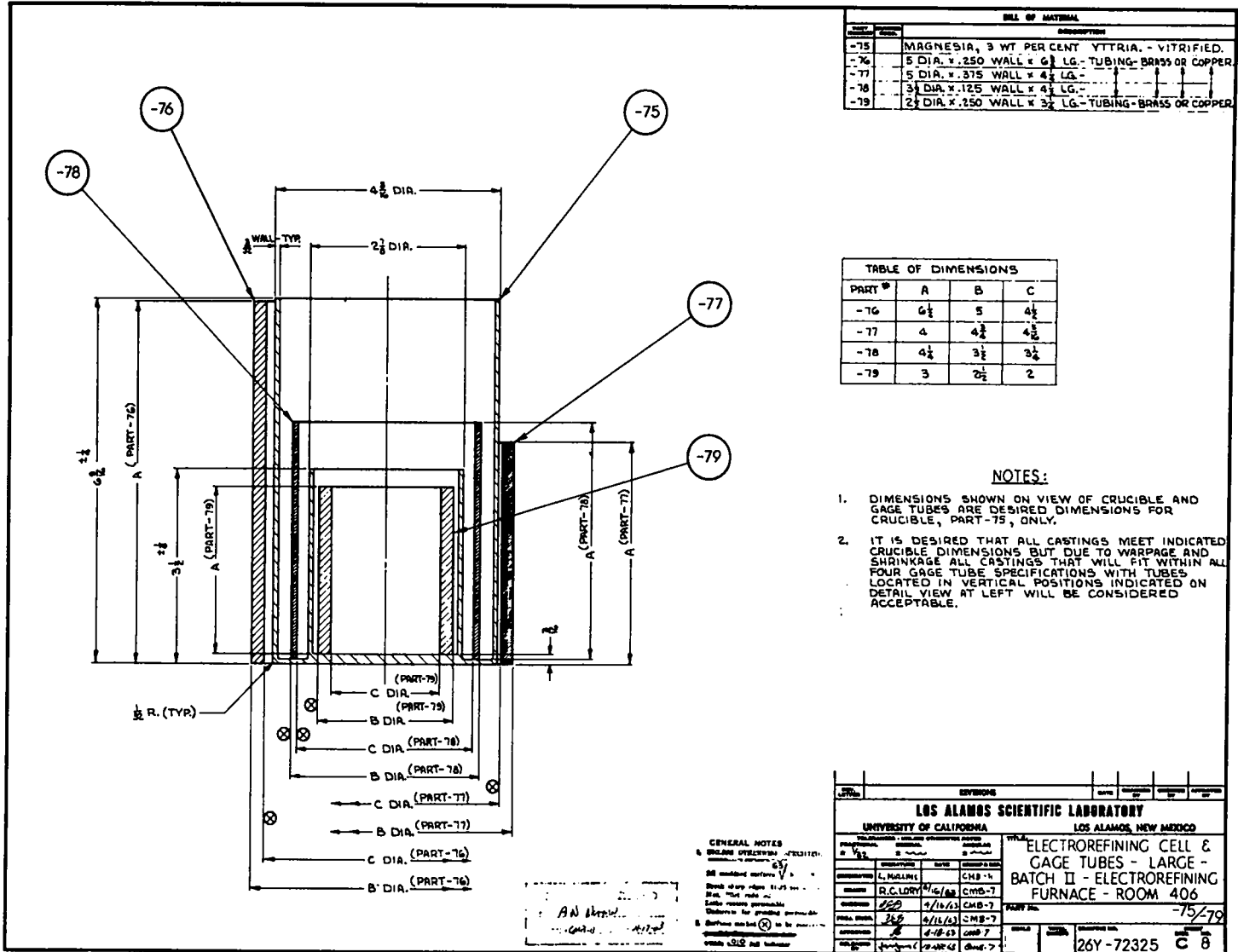
The MgO - 3 w/o Y_2O_3 cell is shown in Fig. 1. Cells must fit the gages shown on this drawing. At present, the cells are fabricated, inspected, and packaged in polyethylene containers by CMB-6. Norton's Blend B MgO and Lindsey's 99.99% Y_2O_3 are used in the cell fabrication.

Anode Feed Metal

The maximum dimensions of the metal feed cylinders are 2-1/4 in. diameter by 3 in. tall (~ 3.7 kg.). The cylinders are prepared by melting and pouring the metal under vacuum at 640-750°C. The electrorefining process is extremely tolerant of the impurities usually associated with plutonium metal. Thus, large amounts, > 1000 p.p.m., of impurities such as iron, nickel, chromium, and copper are readily separated from plutonium. However, large amounts of impurities which are similar to or more electropositive than plutonium are to be avoided. These latter impurities include rare earths, actinides, and aluminum. Nominal amounts of these impurities, ~ 500 p.p.m., can be tolerated if the 2.10 m/o $PuCl_3$ or PuF_3 electrolyte is used. Details of the separation of impurities from plutonium in the electrorefining process are contained in references 3 and 4.

Electrolyte

The electrolyte feed is a 90 mm. diameter cylinder weighing 1.4 kg; compositions are given below. The alkali halides used in the preparation are analytical grade reagents, and the plutonium halides should contain less than 1000 p.p.m. of metallic impurities.



BILL OF MATERIAL		DESCRIPTION
-75		MAGNESIA, 3 WT PER CENT YTTRIA - VITRIFIED.
-76		5 DIA. X .250 WALL X 6 LG. TUBING - BRASS OR COPPER.
-77		5 DIA. X .375 WALL X 4 LG. TUBING - BRASS OR COPPER.
-78		3 DIA. X .125 WALL X 4 LG. TUBING - BRASS OR COPPER.
-79		2 DIA. X .250 WALL X 3 LG. TUBING - BRASS OR COPPER.

TABLE OF DIMENSIONS			
PART #	A	B	C
-76	6 1/2	5	4 1/2
-77	4	4 1/2	4 1/2
-78	4 1/2	3 1/2	3 1/2
-79	3	2 1/2	2

NOTES:

- DIMENSIONS SHOWN ON VIEW OF CRUCIBLE AND GAGE TUBES ARE DESIRED DIMENSIONS FOR CRUCIBLE, PART-75, ONLY.
- IT IS DESIRED THAT ALL CASTINGS MEET INDICATED CRUCIBLE DIMENSIONS BUT DUE TO WARPAGE AND SHRINKAGE ALL CASTINGS THAT WILL FIT WITHIN ALL FOUR GAGE TUBE SPECIFICATIONS WITH TUBES LOCATED IN VERTICAL POSITIONS INDICATED ON DETAIL VIEW AT LEFT WILL BE CONSIDERED ACCEPTABLE.

UNIVERSITY OF CALIFORNIA
LOS ALAMOS SCIENTIFIC LABORATORY
LOS ALAMOS, NEW MEXICO

ELECTROREFINING CELL & GAGE TUBES - LARGE - BATCH II - ELECTROREFINING FURNACE - ROOM 406

DATE: 1/16/53
BY: R.C. LORRY
CHECKED: 1/16/53
APPROVED: 1/16/53

26Y-72325 C 8

Fig. 1. Electrorefining cell and gage tubes, Drawing No. 26Y-72325, C8

EQUIPMENT SPECIFICATIONS

Electrorefining Furnace Assembly

The furnace assembly is shown in Fig. 2. The materials of construction of various components are given below. Further material of construction details are contained in Drawing 26Y-72325, sheets 1 through 10.

<u>Item</u>	<u>Part No., Fig. 2</u>	<u>Material</u>
Furnace Tube	11	Stainless Steel, Type 316
Cathode	9	Tungsten
Anode	6	Tungsten
Stirrer	8	MgO - 3 w/o Y ₂ O ₃
Safety Crucible	3	Tantalum
Thermal Spacer	1	MgO
Loading Can	2	Stainless Steel, Type 316
Cell	4	MgO - 3 w/o Y ₂ O ₃
Pre-electrolysis Rod	10	Tungsten

Automatic Back E.M.F. Sampler

The schematic diagram of this equipment is given in Fig. 3.

D.C. Power Supply

The electrical schematic is given in Fig. 4.

Resistance Furnace

The electrorefining cell is heated by a 5 in. I.D. resistance tube furnace, Hevi Duty Electric Co. Type M-5018.

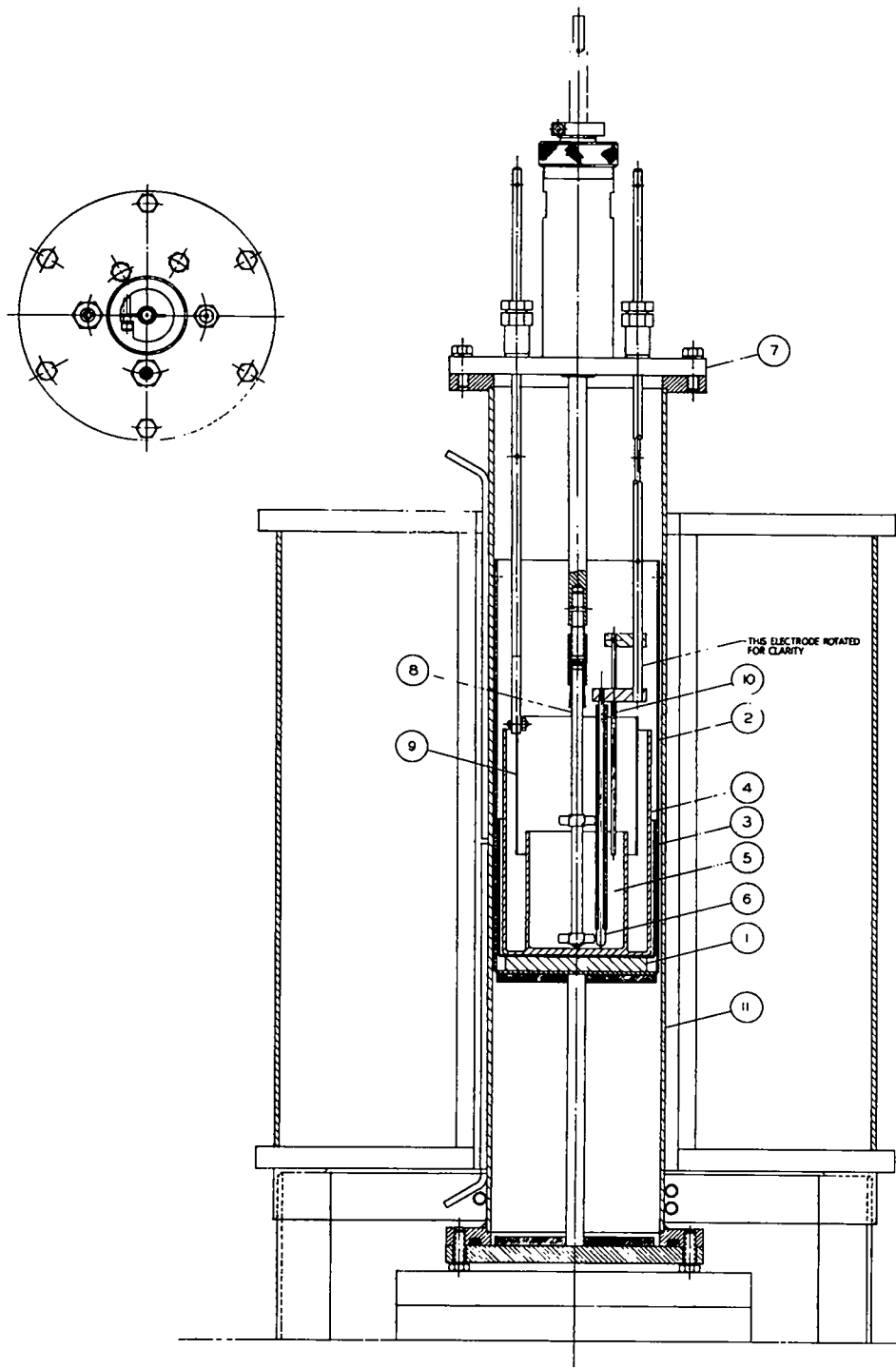
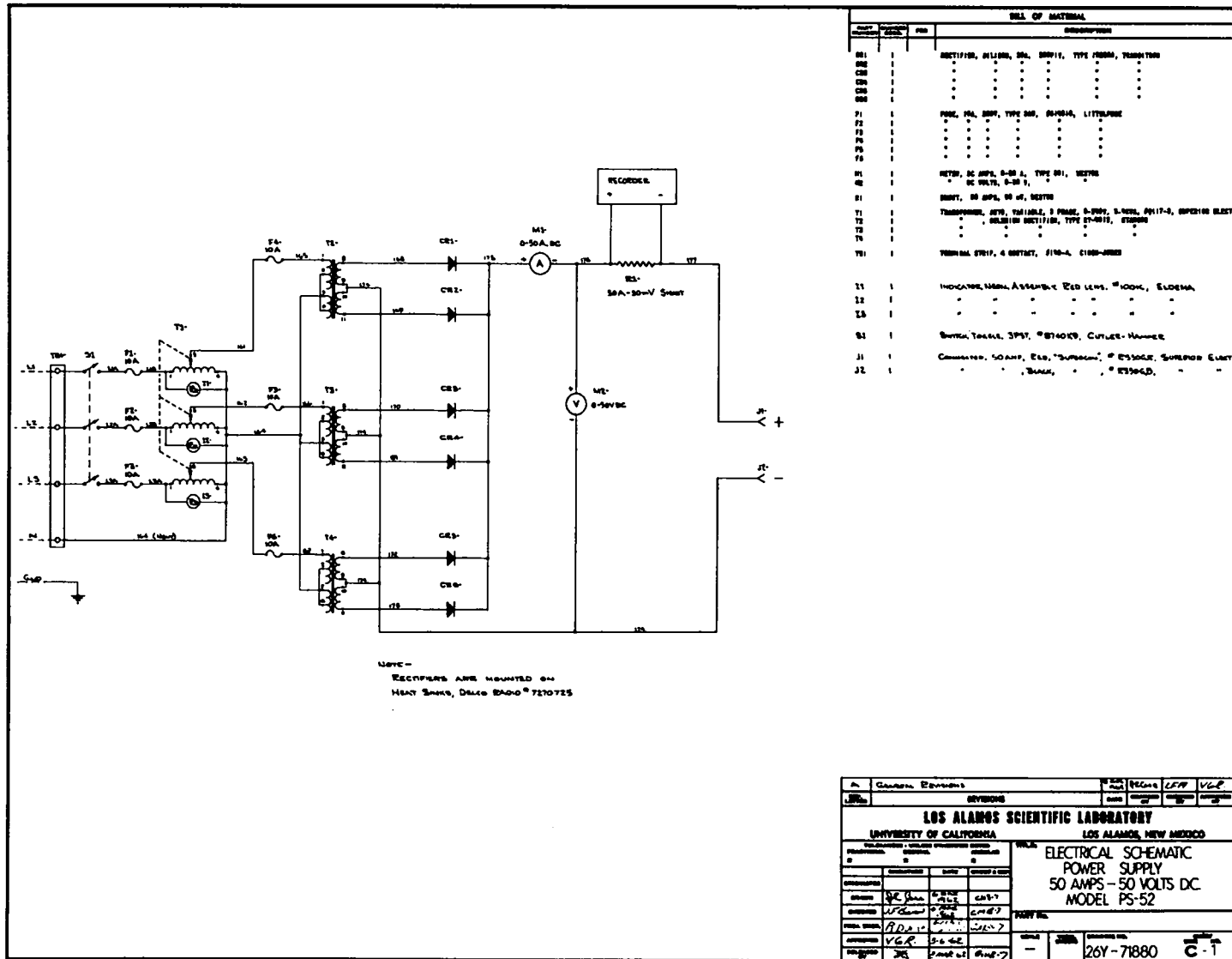


Fig. 2. Assembly, electrorefining furnace, 3.5 kg. scale.
 Drawing No. 26Y-72524 E1



BILL OF MATERIAL		DESCRIPTION
QTY	UNIT	
001		RECTIFIER, 61120A, 50A, 50V11, TYPE P2000, THERMION
002		"
003		"
004		"
005		"
71		POLE, 50A, 50V, TYPE 505, 50A10A, LITTELWAVE
72		"
73		"
74		"
75		"
81		METER, DC AMP, 0-50 A, TYPE 001, WESTON
82		"
83		"
84		"
71		TRANSFORMER, 475, 240/0-115, 0-115, 0-200, 0-200, PH17-5, SUPERIOR ELECT.
72		"
73		"
74		"
75		"
76		TERMINAL STRIP, 4 CONTACT, J10-4, C100-0002
11		INDICATING METER ASSEMBLY 250 LEV. #100K, ELDEMA
12		"
13		"
81		SWITCH TOWER, 3PST, #874019, CUTLER-HAMMER
31		CONNECTOR, 50AMP, 250, "SUPERIOR" # 875504, SUPERIOR ELECT.
32		"

A. General Experiments		DATE	BY	CHKD	VER										
BY: [Signature]		DATE	BY	CHKD	VER										
LOS ALAMOS SCIENTIFIC LABORATORY															
UNIVERSITY OF CALIFORNIA LOS ALAMOS, NEW MEXICO															
PROJECT: [Blank]			TITLE: ELECTRICAL SCHEMATIC												
SUBJECT: [Blank]			POWER SUPPLY												
MODEL: [Blank]			50 AMPS - 50 VOLTS DC												
DRAWING NO.: [Blank]			MODEL PS-52												
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APPROVED	DATE	BY	CHKD	VER											
[Signature]	5-2-52	[Signature]	[Signature]	[Signature]											
DRAWING NO. 26Y-71880			C-1												

Fig. 4. Electrical schematic, power supply model PS-52, Drawing No. 26Y-71880, C1

Gas Purification Equipment

The "welding-grade" argon gas is purified by passage through a Drierite column and a uranium chip furnace operating at $580 \pm 20^\circ\text{C}$.

ELECTROLYTE PREPARATION

Although most of the experimental work has been done with an all-chloride electrolyte, NaCl-KCl-PuCl₃, both PuF₃ and PuF₄ have been shown to be satisfactory substitutes for PuCl₃^{3,4} when used in magnesia ceramics. From a practical standpoint, the fluoride salts are superior to PuCl₃ because they are not hygroscopic and are more generally available in laboratories that produce plutonium metal. For these reasons, only the preparation of the NaCl-KCl-PuF₃ electrolyte is presented below. (PuF₃ is preferred to PuF₄ because PuO₂ is less likely to be present as an impurity in the trifluoride salt.)

Two different concentrations of PuF₃, 2.10 m/o and 0.49 m/o (2.14 w/o), may be used in the NaCl-KCl solvent. The higher concentration is used for runs in which the metal feed contains more than 100 p.p.m. of Am or rare earths and a high purity product is desired. The lower concentration is used for most of the routine processing runs. The procedures given below are for the 0.49 m/o PuF₃ electrolyte. Details for the higher concentration are given in Appendix A, together with the recipes for the all-chloride electrolytes. The PuF₃ is prepared by fluorinating plutonium peroxide in a H₂-HF atmosphere. Thirty grams of the salt are added to the NaCl-KCl casting when the electrorefining cell is loaded.

NaCl-KCl Castings

A 90 mm. diameter salt casting is prepared by melting 609 g. of NaCl and 776 g. of KCl analytical reagent grade salts in a quartz tube (approximately 15 g. of salt is lost in the casting procedure). The salts are dried prior to fusion for at least 24 hr. at 110°C in a vacuum drying oven and are then thoroughly mixed. The mixed salts are placed in a 90 mm. I.D. quartz tube in a vacuum furnace and heated to 300°C at a rate of 50°/hr. and then to the melting point at a rate of ~ 400°/hr. The molten salt is held under vacuum at ~ 700°C for 1 hr. The charge is then allowed to cool to room temperature. Nickel or stainless steel thermocouple protective tubes may be used; however, nickel is preferred. After the casting is unloaded from the quartz tube, it is weighed and stored in a sealed polyethylene container until ready for use. The quartz tube is stored in a dry atmosphere.

OPERATING PROCEDURE

Inspection of Equipment

The stirrer, cathode, and anode should be inspected visually for mechanical defects. Use a resistance meter to check the integrity of the cathode and anode insulators.

Loading of Furnace (see Fig. 2)

Place MgO plate (1) in stainless steel can (2).

Place tantalum safety crucible (3) in stainless steel can.

Place magnesia cell (4) carefully in tantalum safety crucible.

Place metal feed cylinder carefully in anode cup (5).

Place NaCl-KCl casting in magnesia cell.

Place PuF_3 in cavity of NaCl-KCl casting; do not put PuF_3 in direct contact with metal.

Lower stainless steel container into stainless steel furnace tube with handle provided for this purpose.

Rotate container to favor positioning of anode rod (6) in center of anode cup.

Seal furnace tube by placing head flange (7) provided with stirrer (8), anode rod, cathode (9), and pre-electrolysis rod (10) on furnace tube.

Bolt down flange, and connect gas fitting to inert gas line.

Apply vacuum to furnace tube, and check for vacuum tightness; the pressure in the furnace tube should be less than or equal to 100 microns.

Flush system three times with argon and leave system under vacuum.

Turn on uranium chip furnace.

Furnace Heating Cycle

Turn on furnace tube cooling water; negative pressure circulating water is used.

Turn on cell resistance furnace. The cam provided on the controller provides a heating rate of 50°C/hr .

Turn on temperature recorder; furnace tube and chip furnace temperatures are recorded.

When furnace tube has reached 200°C , flush system three times with argon and leave system at a positive gage pressure of 2-3 p.s.i. The pressure regulator on the argon supply should be set to maintain this pressure.

When the electrorefining cell has reached a temperature of 740°C , set the controller to maintain this constant temperature. The controller thermocouple is positioned in the thermowell welded on the outside of the furnace tube. This thermocouple is calibrated monthly against another chromel-alumel thermocouple immersed in the electrolyte. For electrorefining at 740°C , the controller thermocouple is usually set at 800°C .

Loosen the stirrer packing gland and lower the stirrer (8) to the position shown in Fig. 2 (i.e., stirrer is positioned less than $1/16$ in. off bottom of cell.)

Lower cathode cylinder (9) to position shown in Fig. 2 (bottom rim of cathode is positioned $1/4$ in. below top rim of anode cup.) Place locking pin in position.

Lower anode rod (6) to bottom of anode cup with anode rod near center of anode cup as shown in Fig. 2; rotate anode rod to side of anode cup; tighten packing nut.

Lower pre-electrolysis rod (10) to position shown in Fig. 2; place locking pin in position.

Pre-electrolysis

Connect pre-electrolysis rod to negative terminal of D.C. power supply.

Connect anode rod to positive terminal of D.C. power supply.

Turn on stirrer. It should rotate at ~ 1000 r.p.m.

Turn on current-voltage recorder.

Turn on D.C. power supply and adjust current to 2 amp. This should require a potential* of between 0.2 and 0.4 volts.

After passing current for a few minutes, check and record the back e.m.f. (back e.m.f. should be 0.0-0.1 volts.)

Pass 2 amp. for 1 hr.; shut off current; shut off stirrer.

Disconnect and raise the pre-electrolysis rod.

Connect negative terminal of D.C. power supply to cathode rod.

Electrorefining

Turn on stirrer.

Pass current at a value which will permit termination of the electrorefining at a convenient time.** Resistance ($\frac{\text{volts}}{\text{amps}}$) should be ~ 0.15 ohms.

Turn on automatic back e.m.f. sampler. Set sampler dial at 0.3 volts.

After several minutes operation, measure and record back e.m.f. (back e.m.f. should be 0.0-0.2 volts.)

*The potentials are measured across the lead wires carrying current from the D.C. power supply to the electrorefining cell.

**Example of current calculation:

Assume: charge of 3.2 kg. Pu⁰
electrolysis to terminate 46 hr. later

$$\text{Current} = \frac{3200}{2.97} \frac{\text{g.}}{\text{g/amp.hr.}} \frac{1}{46 \text{ hr.}} = 23.4 \text{ amp.}$$

Proper operation of the equipment should be checked periodically (at least twice) during the normal 8 hr. working day. The check should include:

1) Observation of the argon gas flow meter. (If more than 0.5 cubic feet of gas is being used per hour, the stirrer packing gland should be tightened.)

2) Measurement and recording of back e.m.f.; record measurement on current voltage recorder chart.

3) Visual inspection of stirrer operation; stirrer should rotate at ~ 1000 r.p.m.

The electrorefining is terminated automatically by the back e.m.f. sampler.

Symptoms of Electrorefining Failure

Back e.m.f. equal to or less than 0.00 volts, and resistance less than 0.08 ohms.

This condition can be due to either a broken anode cup or to a "shorting-out" of anode and cathode. Observance of the operating instructions will eliminate the latter possibility. If anode cup is broken, stop the run.

Back e.m.f. greater than 0.2 volts, and resistance greater than 0.3 ohms.

This condition is due to improper positioning of the anode rod, i.e., not positioning it in the anode metal. Observance of the operating instruction will eliminate this failure.

Back e.m.f. greater than 0.2 volts, and resistance less than 0.08

ohms.

This condition is due to improper positioning of the anode rod; i.e., not placing it in the anode metal and "shorting it out" against the cathode. Observance of the operating instructions will eliminate this failure.

Temperature Cycling

Upon termination of the electrorefining, stop the stirrer.

Disconnect anode and cathode leads.

Agitate the product metal by gently lowering cylindrical cathode to bottom of cell, then raising to its electrolysis position. Repeat operation approximately fifty times.

Raise stirrer out of salt; insert locking pin.

Raise anode rod out of salt; insert locking pin.

Raise cathode cylinder out of salt; insert locking pin.

Turn on program controller to "terminal heating cycle." (The cell temperature will be raised to 800°C and held at this temperature for 2 hr. The cell will then be cooled to 400°C at a rate of 50°/hr. The furnace will then be shut off automatically.)

Unloading

Do not unload until cell temperature is less than 50°C.

Carefully unbolt and remove furnace head.

Remove stainless steel can from furnace tube with handle.

Remove cell from safety crucible, and record cell total weight.

Break crucible and salt away from product ring and anode residue.

Record weights of product and anode residue.

Sample ring by breaking off a wedge with hydraulic press.

Storage and Care of Equipment

Store the ceramic stirrer in an inert, dry atmosphere immediately after unloading the furnace.

Remove salt deposits from furnace tube with vacuum cleaner.

Keep furnace tube under vacuum when not in use.

If cathode is to be used for next run, store in inert, dry atmosphere.

ANALYTICAL DATA REQUIRED

Feed Materials

Metal: Submit one top sample of alpha feeds and a top and a bottom sample of alloy feeds for plutonium and alloying agent analyses.

Products

Metal Ring: Submit one sample for spectrochemical, chemical, radiochemical, and gas analyses. Submit one sample for density determination.

Salt: Submit one sample for radiochemical analyses.

Typical analytical request forms are shown on the following pages.

REQUEST FOR ANALYSIS
TO
ANALYTICAL LABORATORY
CMB-1

Sample No: Metal Feed Date Submitted: _____

Description: _____

Remarks: _____

Submitted by: _____ Phone: _____

ANALYSIS

Constituent(s) for which analysis is requested	Method of analysis requested	Expected Percent	Percent by Analysis
Pu	Chemical		
Alloying Agent (e.g. Fe)	Chemical		

If spectrochemical analysis is requested, indicate one of the following types.

SPECTRO QUAL: Estimates of percentages of elements are determined. This type may be requested for determination of (1) presence or absence of elements, (2) major and minor constituents, (3) general survey, (4) identification of unknowns and (5) approximate analysis of miscellaneous materials.

SPECTRO QUANT: A more accurate analysis based on reference standards which must be carefully prepared for each kind of material. Please consult Spectroscopic Section, Phone 7-4986, before requesting. Method is most applicable to analysis for elements below 1% level.

Remarks: _____

Analyst: _____ Date Reported: _____

Form No. 122

REQUEST FOR ANALYSIS
TO
ANALYTICAL LABORATORY
CMB-1

Sample No: NaCl-KCl-PuF₃ Date Submitted: _____

Description: _____

Remarks: _____

Submitted by: _____ Phone: _____

ANALYSIS

Constituent(s) for which analysis is requested	Method of analysis requested	Expected Percent	Percent by Analysis
Pu	Radio-Chemical	2	
Am	" "		

If spectrochemical analysis is requested, indicate one of the following types.

SPECTRO QUAL: Estimates of percentages of elements are determined. This type may be requested for determination of (1) presence or absence of elements, (2) major and minor constituents, (3) general survey, (4) identification of unknowns and (5) approximate analysis of miscellaneous materials.

SPECTRO QUANT: A more accurate analysis based on reference standards which must be carefully prepared for each kind of material. Please consult Spectroscopic Section, Phone 7-4986, before requesting. Method is most applicable to analysis for elements below 1% level.

Remarks: _____

Analyst: _____ Date Reported: _____

Form No. 122

To: CMB-1, Attn. _____ Submitted by _____

From: CMB-11 _____ Date _____

Sample Nos. _____ Weight, grams _____

Description Metal Product _____

ANALYSIS REQUESTED		ANALYSIS REQUESTED	
<input checked="" type="checkbox"/> Al		<input checked="" type="checkbox"/> Li	
<input checked="" type="checkbox"/> B		<input checked="" type="checkbox"/> Mg	
<input checked="" type="checkbox"/> Be		<input checked="" type="checkbox"/> Mn	
<input checked="" type="checkbox"/> Bi		<input checked="" type="checkbox"/> N ₂	
<input checked="" type="checkbox"/> C		<input checked="" type="checkbox"/> Na	
<input checked="" type="checkbox"/> Ca		<input checked="" type="checkbox"/> Ni	
<input checked="" type="checkbox"/> Ce		<input checked="" type="checkbox"/> O	<input checked="" type="checkbox"/> Exact Value
<input checked="" type="checkbox"/> Co		<input checked="" type="checkbox"/> Pb	
<input checked="" type="checkbox"/> Cr		<input checked="" type="checkbox"/> Pu	
<input checked="" type="checkbox"/> Cu		<input checked="" type="checkbox"/> Si	
<input type="checkbox"/> F		<input checked="" type="checkbox"/> Sn	
<input checked="" type="checkbox"/> Fe		<input checked="" type="checkbox"/> Ta	
<input checked="" type="checkbox"/> H		<input checked="" type="checkbox"/> Th	
<input checked="" type="checkbox"/> La		<input type="checkbox"/> V	
<input checked="" type="checkbox"/> Remarks		<input checked="" type="checkbox"/> Zn	
Following analyses are		<input checked="" type="checkbox"/> Zr	
also requested:			
Y			
U			
W			
Am			

APPENDIX A

ELECTROLYTE RECIPES

Since both PuCl_3 and PuF_3 can be used in the equimolar NaCl-KCl solvent, recipes for the preparation of 2.10 m/o and 0.49 m/o Pu halide castings are given in Table A-1. The castings can be made in a glove box by combining the weights of salt shown in Table A-1, or the halide salts can be added to a NaCl-KCl casting. The latter procedure is recommended.

TABLE A-1
Composition of Electrolyte for 3.5 kg. Scale

Pu Halide	Pu Halide Salt	Composition of Electrolyte						g. of Salt		
		PuCl ₃ or PuF ₃		KCl		NaCl		Pu Halide Salt	KCl	NaCl
		m/o	w/o	m/o	w/o	m/o	w/o			
PuCl ₃	PuCl ₃	2.10	10	48.95	50.44	48.95	39.56	140	706	554
PuCl ₃	NaCl-PuCl ₃	2.10	10	48.95	50.44	48.95	39.56	186	706	508
PuCl ₃	PuCl ₃	0.49	2.50	49.75	54.65	49.75	42.85	35	765	600
PuCl ₃	NaCl-PuCl ₃	0.49	2.50	49.75	54.65	49.75	42.85	46	765	589
PuF ₃	PuF ₃	2.10	8.72	48.95	51.17	48.95	40.11	122	716	561
PuF ₃	PuF ₃	0.49	2.14	49.75	54.86	49.75	43.00	30	768	602

APPENDIX B

CRITICALITY MEMORANDA

To: H. C. Paxton, N-2

April 14, 1961

From: L. J. Mullins, CMB-11

Request for Criticality Approval of 5 kg. Batch Electrorefining Equipment

CMB-11

Description

Electrorefining Cell --- A sketch of the alumina cell to be used is appended (Drawing No. 26Y-72288).

Equipment Setup --- A sketch of the assembled equipment is appended. Also attached are photographs of the equipment and product ring from small scale runs.

Procedure --- 4-5 kg. of alpha or delta phase plutonium in the shape of a cylinder will be loaded into the inner chamber of the electrorefining cell. On top of this is placed a 4 in. diameter cylinder of (NaCl-KCl-PuCl₃) weighing about 1.6 kg. and containing 150-175 g. of PuCl₃. The cell is loaded into the 4-1/2" I.D. stainless steel furnace and provided with a helium atmosphere. The charge is then heated to 700°C at which time a tungsten cylindrical cathode (2-3/4" diameter) is lowered into the molten salt, the bottom of the cylinder being 3-1/2" from the bottom of the cell. A 1/4" diameter tantalum stirring rod equipped with a 1-1/2" and a 1" diameter stirring blade is lowered into the molten metal. The stirrer turns at 1700 rpm. D.C. current is passed through the cell which results in the dissolution of plutonium metal at the anode (the inner chamber) and the plating out

of metal at the cathode. The metal drips into the annulus of the cell. When dissolution of the metal at the anode is complete the current is turned off, the cell is heated to 800°C for 1 hr. and then allowed to cool to room temperature. After the furnace is unloaded, the cell is broken apart and the salt and metal are then stored in separate containers. A 10-20 g. sample is cut from the product ring which is then transferred to the casting operation. Prior to re-melting and re-casting, however, the ring is pickled successively in containers of nitric acid, water and ethyl alcohol in order to remove any adhering salt. (if this pickling procedure is considered to be unsafe, the ring could be cleaned mechanically.) The product (ca. 5 kg.) is melted in a 3 in. diameter tantalum pouring crucible and poured into molds of the desired shape, usually 1 in. diameter rods.

L. J. Mullins

LJM:mb

To : L. J. Mullins, CMB-11

May 2, 1961

From : H. C. Paxton and W. R. Stratton, N-2

Subject: NUCLEAR SAFETY-PU ELECTROREFINING EQUIPMENT

Symbol : N-2-4069

Based on Rocky Flats measurements of Pu reflected by 2-1/2"-thick KCl and intermixed with the salt, a limit of 5 kg. total Pu will be safe in your electrorefining cell (drawing 26Y-72288; process described in your memo of 4/14/61). This quantity of material would remain somewhat subcritical even under the extreme condition that the cell be immersed in water. But, of course, there is no double-batching allowance.

Pickling of the resulting annulus in a 1-liter beaker will be satisfactory (immersion in large volumes of hydrogenous fluids should be avoided). It is suggested that crucible charges for casting into 1"-diam. rods be limited to 4-1/2 kg. α -Pu subject to review of the specific casting setup.

/s/
H. C. Paxton

/s/
W. R. Stratton

HCP:WRS:ml

cc: Roy Reider, H-3
Thru B. L. Moore, W-DO
R. J. Bard, CMB-8
Elizabeth Plassmann, W-7
H. C. Paxton (file)
W. R. Stratton (file)

H. C. Paxton, N-2

September 27, 1962

J. A. Leary, CMB-11

Criticality limitations in the 5 kilogram scale plutonium electrorefining process.

CMB-11

The electrorefining process now is being operated routinely in CMB-11. Generally the total amount of plutonium in each electrorefining batch is between 3500 and 4000 grams.

We are now considering a change in composition of the electrolyte for a series of experiments. The approved system contained 100 grams of plutonium (as 140 g. PuCl_3) dissolved in 1260 grams of an equimolar mixture of sodium chloride and potassium chloride. We would like to change this composition by adding the plutonium as PuF_3 instead of PuCl_3 . This would give a total fluoride concentration of 1.25 gram-atoms (124 g. PuF_3) in 1384 grams of total electrolyte. The specific gravities of these salt systems are not known. However, it is estimated that changing the plutonium salt will not significantly alter the total specific gravity of the molten electrolyte.

Your approval is requested to make this procedural change.

JAL:mb
cc/ J.A. Leary
CMB-11 file

J. A. Leary

To : J. A. Leary, CMB-11

October 3, 1962

From : D. R. Smith, W. R. Stratton, N-2

Subject: MODIFICATION OF PLUTONIUM ELECTROREFINING PROCESS

Symbol : N-2-7567

The proposed change in the electrorefining process, as described in your memo to H. C. Paxton of September 27, 1962, has been considered with respect to criticality safety. The proposed substitution of fluorine for chlorine on an atom for atom basis (approximately 24 gm. fluorine for approximately 40 gm. chlorine) would be expected to result in a slight reduction of the reactivity of the system, and thus enhances the criticality safety of the process.

/s/
D. R. Smith

/s/
W. R. Stratton

DRS:WRS:af

CC: Wm. J. Maraman, CMB-11
W. R. Stratton, N-2
D. R. Smith, N-2
N-2 file

To : J. A. Leary, CMB-11

October 18, 1962

From : D. R. Smith, W. R. Stratton, N-2

Subject: CALCULATIONS OF PLUTONIUM ELECTROREFINING CELL

Symbol : N-2-7581

Three DTK calculations have been performed to provide guidance for evaluating the reactivity of the electrorefining process.

The first calculation provides the critical radius for a sphere of Pu^{239} , reflected by about 1/8" of material approximating Alumina, reflected by one inch of salt. The salt composition in this problem is 10 weight % PuCl_3 , 50.4 weight % KCl , 39.6 weight % NaCl . The critical radius obtained is 4.554 cm, or a critical mass of 7.7 kg.

The second problem is identical to the first, with the addition of 20 cm water outside the salt to evaluate the effect of external reflection. The critical radius is 4.25 cm, and critical mass 6.28 kg.

The third problem is a repeat of the first, with PuF_3 replacing PuCl_3 in the salt. This resulted in a reduction of critical radius by 0.1%, which is negligible.

We conclude that an adequate margin of safety is inherent in operations using up to 4 kg. of Pu in the anode cup.

/s/

D. R. Smith

/s/

W. R. Stratton

DRS:WRS:af

CC: Wm. J. Maraman, CMB-11

L. J. Mullins, CMB-11

Roy Reider, H-3

W. R. Stratton, N-2

D. R. Smith, N-2

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