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RECOVERY OF PLUTONIUM AND OTHER TRANSURANIUM ELEMENTS FROM IRRADIATED PLUTONIUM-ALUMINUM ALLOY BY ION EXCHANGE METHODS

R. E. Brooksbank
W. T. McDuffee

OAK RIDGE NATIONAL LABORATORY
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CHEMICAL TECHNOLOGY DIVISION

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MAY 1964

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CONTENTS

Abstract 1
1. Introduction 3
2. Summary 6
3. Flowsheets 14
3.1 Chemical Flowsheet 14
3.2 Equipment Flowsheet 16
4. Procedure 16
4.1 Feed Preparation 16
4.1.1 Description and History of the Fuel 16
4.1.2 Shipping Cask Description and Handling 19
4.1.3 Charging the Fuel to the Dissolver 21
4.1.4 Dissolution of the Fuel 22
4.1.5 Dissolution Rate Studies 25
4.1.6 Feed Clarification and Adjustment 28
4.2 Plutonium Recovery 30
4.2.1 Ion Exchange 30
4.2.2 First Stage Operation 33
4.2.3 Second Stage 35
4.2.4 Handling of the Plutonium Product 36
4.3 Transuranium Element Recovery 38
4.3.1 Feed Preparation 38
4.3.2 Ion Exchange 41
4.3.3 Transuranium Element Product Handling 44
5. Appendix 46
5.1 Detailed Process Data 46
5.2 Examination of Spent Resin 64
6. Conclusions and Recommendations 65
7. Acknowledgement 68
8. References 69

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ABSTRACT

In order to isolate plutonium and other transuranium elements such as americium and curium from the aluminum in highly irradiated Pu-Al alloy fuel elements, an ion exchange process was demonstrated. The americium and curium will be used in the nuclear studies related to the very heavy elements. The report describes the process in detail and gives results from the developmental work and the pilot plant demonstration. The process consisted in dissolving the fuel rods in 6 M HNO_3 -- 0.05 M Hg^{2+} -- 0.03 M F^-, adjusting the valence of the plutonium to four and clarifying the solution by filtration through a sand-bed filter precoated with filter aid, after which the plutonium was recovered by sorption on anion exchange resin, the sorbed plutonium then being scrubbed free of aluminum and fission products with 7 M HNO_3 -- 0.01 M F^-. The partly purified plutonium was then eluted from the resin with 0.7 M HNO_3. Additional purification and decontamination was obtained by repeating the ion exchange step. The plutonium product, meeting radiation specifications, was nearly free of aluminum and had been decontaminated from fission products by a factor of 4.6×10^5. Overall plutonium recovery processing losses of 2.7 and 12.6%, respectively, occurred with fuel elements irradiated to 87 and greater than 97% burnup.

The transuranium elements were then recovered from the aluminum-

bearing, plutonium loading waste effluent solution, which had been collected and stored during the plutonium-recovery phase of the program. The free acid in the raw wastes was removed by evaporation to 140°C, after which the concentrate was diluted with water until the concentration of the aluminum was 2.5 to 2.8 M. At this point, the aluminum nitrate was nearly monobasic. After clarification to remove the aluminum hydroxide that was formed, the rare earths and transuranium elements were sorbed on anion exchange resin at 60°C. Traces of aluminum and light fission products that had sorbed on the resin were scrubbed from it was 8 M $LiNO_3$. The purified rare earth fraction containing the transuranium elements was then eluted from the resin with 0.7 M HNO_3 and evaporated to small volume.

The conditions developed and demonstrated in the recommended flowsheet for the recovery of plutonium and other transuranium elements from irradiated Pu-Al alloy fuel permitted average dissolution rates of the alloy of about 0.8 mg min^{-1} cm^{-2} and resulted in plutonium recoveries greater than 97%. Some reduction in plutonium recovery may be expected, however, as the burnup of the fuel elements approaches 99% because of the destruction or masking of the exchange sites on the resin. In the runs described, resin exposure to the radiation was sufficient in two instances to destroy about 35% of the exchange sites, a rather serious defect of the process.

Transuranium element recoveries greater than 95% were demonstrated operating according to the recommended flowsheet developed during this program. High recovery of the transuranium elements depended on their complete recovery from the aluminum hydroxide cake collected during the clarification step. The cake retained about 35% of the transuranium

elements, which could be recovered by rinsing with dilute nitric acid and returning to the evaporator. High salting strength (8 M NO_3^-) was necessary in the feed and scrub solutions in the ion exchange step.

The process described here offers a useful approach to the recovery of plutonium and other transuranium elements from Pu-Al alloy fuel elements, even those of high burnup. The products, plutonium and transuranium elements, will be nearly free of aluminum and therefore adaptable to subsequent processes for isolating and recovering them.

1. INTRODUCTION

The recovery of plutonium and transuranium elements from highly irradiated Pu-Al alloy was undertaken to provide sufficient quantities for the study of their nuclear properties and for the development and demonstration of processes for their separation and recovery in a high state of purity. This separation takes place in two steps: one that provides aluminum-free plutonium product, and a second one which isolates and recovers the transuranium elements (plus rare earths) from the plutonium-free effluent of the first step. The purpose of the work was to provide a process for the removal of the aluminum to such an extent that it could no longer be regarded as an interference in subsequent processing to follow. This report presents the results of a pilot plant demonstration of a process, featuring anion exchange separation and purification methods, in the recovery of plutonium and transuranium elements from two lots of Pu-Al alloy fuel rods irradiated to greater than 87% burnup.

Specific objectives of the program were to:

1. Recover the plutonium from two lots of highly irradiated Pu-Al alloy fuel rods that would be sufficiently decontaminated from fission products and aluminum for use in further development and in the further production of transuranium elements.
2. Recover the transuranium elements free of aluminum, in a condition suitable for use as raw material in the development of processes for the separation and recovery of highly purified individual elements.
3. Demonstrate on a pilot-plant scale a flowsheet for the recovery of purified plutonium and other transuranium elements as aluminum-free products.

Prominent problems to be solved consisted in dissolving the spent fuel elements, removing the silica from the solution, and contending with the resin degradation resulting from the relatively high concentrations of fission products in fuel elements of up to 99% burnup. Other problems encountered during the recovery of the transuranium elements were: (1) the formation of aluminum hydroxide during evaporation of the effluent from the plutonium recovery step; (2) the removal of aluminum hydroxide from the evaporated effluent; (3) recovery of the transuranium elements from the aluminum hydroxide filter cake; and (4) resin degradation which resulted in increased loss of material. Some of the problems were unusual because of the high burnup of the fuel elements, and others were due to the high salt strengths of the loading and scrubbing solutions.

The process includes dissolution of the alloy in 6 M HNO_3 catalyzed

with 0.05 M Hg^{2+} and 0.03 M F^-, clarification and adjustment of the plutonium valence to four and the acid concentration to 7 M HNO_3 and two stages of anion exchange purification and recovery. The process also includes steps to recover the transuranium elements from the ion exchange loading waste effluent from the plutonium recovery step by removing the free nitric acid by evaporation, adjustment of the acid and aluminum concentration to 0.5 to 1.0 N acid deficient and 2.5 to 2.8 M, respectively, followed by anion exchange sorption of the rare earths and transuranium elements. Additional purification was obtained by scrubbing traces of sorbed aluminum and light fission products from the resin, after which the transuranium elements (and rare earths) were eluted from the resin with 0.7 M HNO_3 and concentrated to small volume by evaporation.

Ion exchange was selected because the technology for plutonium recovery is well known and it had been shown that the transuranium elements also sorb on anion exchangers from neutral, high-salt-strength solutions. As expected, the process worked, though some defects, especially the low loading capacity of the ion exchanger for sorbing the transuranium elements, were noted. The information in this report is presented in the following order: dissolution of the fuel elements, valence adjustment of the plutonium, and its sorption and recovery, free of aluminum. This is followed by a section on the evaporation of the ion exchange effluent from the plutonium recovery step, clarification, and adjustment of the solution prior to the sorption and recovery of the transuranium elements and rare earths, free of both aluminum and plutonium.

2. SUMMARY

Two lots of Pu-Al alloy fuel that had been irradiated to greater than 87% burnup were processed on a pilot plant scale to recover about 700 g of plutonium, rich in the heavier isotopes and pure enough to meet radiation and purity specifications. In addition, about 4 g of transuranium elements was recovered as a purified rare earth fraction that was essentially aluminum-free and suitable as feed material in the further development of separations processes for the recovery of individual transuranium elements.

The process was conducted to demonstrate a flowsheet featuring anion exchange methods that had been developed to recover plutonium and other transuranium elements free of the aluminum originally present. This fuel consisted of two lots of 0.94-in-diam by 5-ft-long plutonium-aluminum alloy rods canned in aluminum; one lot (24 rods) had been irradiated to about 87% burnup and the second (8 rods) to 99%. These were dissolved in 6 M HNO_3--0.05 M Hg^{2+}--0.03 M F^-; dissolved silica was coagulated and removed by digesting the solution with 10 ppm gelatin (followed by plutonium valence adjustment) and filtration. After acid adjustment to 7 M HNO_3, the plutonium was sorbed at 60 to 70°C on 20 to 50 mesh Permutit SK anion exchange resin in the first of two ion exchange purification stages. Residual sorbed aluminum and fission products were scrubbed from the resin with 7 M HNO_3--0.01 M F^- and the plutonium eluted with 0.7 M HNO_3. The plutonium product solution from the first purification stage was adjusted to convert the plutonium valence to four and the acid concentration to 7 M HNO_3, after which the plutonium was sorbed on a second bed of anion exchange resin identical to that in the first. Traces of residual aluminum and

fission products were scrubbed from the resin, first with 7 M HNO_3--0.01 M F^- then with a small rinse with fluoride-free 7 M HNO_3. The finished plutonium product was recovered from the resin by elution with 0.7 M HNO_3.

The transuranium elements were recovered from the aluminum-bearing ion exchange loading waste effluent that had been collected and stored during the plutonium-recovery phase of the program. These were first evaporated to 140°C to remove free acid and to concentrate the aluminum, then diluted with acid and water to adjust the acid concentration to 0.5 to 1.0 N acid deficient and the aluminum to 2.5 to 2.8 M. After filtration through a bed of Ottawa sand precoated with filter aid to remove the aluminum hydroxide, the clarified solution was passed at 0.5 to 1.0 ml min^{-1} cm^{-2} through a 4-in-diam by 40-in-long bed (7.5 liters) of 50 to 100 mesh Dowex 1 X10 anion exchange resin held at 60 to 70°C, where the rare earths and the transuranium elements were sorbed. Residual aluminum and light fission products were scrubbed from the bed with 8 M $LiNO_3$ metered to the resin column at the same rate and temperature as the feed. The sorbed rare earths and transuranium elements were eluted from the resin with 0.7 M HNO_3 and the product solution evaporated to approximately half volume and stored for further processing in the future.

Plutonium was recovered as an aqueous product solution that met radiation specifications. That recovered from the 24-rod lot was decontaminated from fission products by a factor of 6.9×10^5 (Table 2.1) while that from the 8-rod lot averaged 4.6×10^5. The average composition of the plutonium product from each lot of fuel is given in Table 2.2. Overall recoveries totaled 93% (Table 2.3). Ion exchange sorption and scrubbing losses for

Table 2.1. Process Losses and Decontamination Factors in Recovery of Plutonium and Transuranium Elements from Irradiated Plutonium Aluminum Alloy

	Losses (% of Feed)			Gross Decontamination Factor[a]
	Feed Preparation	Ion Exchange		
		Sorption	Scrubbing	
Plutonium Recovery				
24-Rod lot				
First cycle	--	0.9	1.6	7.5×10^3
Second cycle	--	0.1	0.1	93
Overall	--	1.0	1.7	6.9×10^5
8-Rod lot				
First cycle	--	2.1	5.0	1.3×10^3
Second cycle	--	2.7	2.8	170
Overall	--	4.8	7.8	2.3×10^5
Average overall (both lots)		2.9	4.8	4.6×10^5
Transuranium Element Recovery				
Process	27[b]	9.0[b]	4.0[b]	10

[a] Ratio of gross γ's per unit weight in feed to that in product.

[b] Based on raw waste to the evaporator.

Table 2.2. Average Composition of Plutonium Product

Source	Total Weight (g)	Conc. (g/liter)	Counting Rate (cts min^{-1} ml^{-1})		HNO_3 (M)	Isotopic Assay (wt %)				
			Gross α	Gross γ		Mass 238	Mass 239	Mass 240	Mass 241	Mass 242
24-Rod lot	615	8.2	1.5×10^9	1.6×10^5	2.0	--	29.3	51.2	12.8	6.5
8-Rod lot	30.4	0.19	5.0×10^7	1.1×10^5	2.8	1.09	0.35	15.27	7.8	75.49
Average	645.4	6.2[a]	--	--	--	--	--	--	--	--

[a] Weighted average.

Table 2.3. Material Balance: Plutonium Recovery

	Amount of Plutonium (g) From:			
	24-Rod Lot		8-Rod Lot	
Calculated[a] content of rods		576		48
Feed measurements		700		48.3
Difference (gain)		(124)		(0.3)
Recovery process				
Specification product	655		30.4	
Retained product	12		--	
Losses to waste disposal	8		14.4[c]	
Stored waste[b]	3		1.7	
Total accounted for in process	678	678	46.5	46.5
Difference across process		22		1.8
Process balance, recovery/feed, %		96.9		96.2

[a] Based on reactor heat balances.

[b] Later discharged to waste storage after processing for transuranium element recovery.

[c] Includes 9.8 g irreversibly sorbed on first cycle resin bed.

the program averaged 2.9 and 4.8%, respectively, with the lower losses (1.0 and 1.7%) being observed during processing the lower-burnup material present in the 24-rod lot of fuel rods (plutonium content was 5 times that in the 8-rod lot, which averaged 4.8 and 7.8%).

The transuranium-element product fraction of the composition given in Table 2.4 was decontaminated from aluminum by a factor of 70 and from light fission products by a factor of 10 (Table 2.1). Overall recovery of the transuranium elements from the raw plutonium-recovery waste effluent solution averaged 63% (Table 2.5), taking into account the 37% loss that occurred during feed preparation. Ion exchange sorption and scrubbing losses averaged 9 and 4%, respectively, but under the ideal conditions specified in the flowsheet (Sec. 3) averaged 0.2 and 0.4%, coupled with an overall recovery of greater than 95%. High recoveries of the transuranium elements depended on a thorough washing of these from the aluminum hydroxide solids generated in the initial evaporation step and partially removed upon filtration; often, as much as 35 to 40% of the transuranium elements present in the unclarified feed would be retained in the bulky filter cake of these solids. Low sorption and scrubbing losses in the ion exchange step depended on the salt strength of the feed and scrub solutions but were independent of the acid deficiency over the range 0.5 to 1.0 N. At still higher acid deficiencies, colloidal $Al(OH)_3$ occurred in the feed solution in larger amounts and continued to precipitate after filtration and interfering with the mechanical operation of the ion exchange system.

Radiation damage to the anion exchange resin was apparent in both phases of the program. In the plutonium-recovery phase, where the Permutit

Table 2.4. Composition of Transuranium Element Product

Source	Al (M)	Li (M)	HNO_3 (M)	Gross Counting Rate (counts min^{-1} ml^{-1})			Nuclide (dis min^{-1} ml^{-1})				
				α	β	γ	Ce^{144}	Cs^{137}	Ru^{106}	Sr^{90}	Zr^{95}
24-rod lot	0.06	6.4	1.1	1.1×10^9	2.2×10^{10}	2.7×10^9	1.9×10^{11}	4.8×10^7	1.8×10^{10}	5.8×10^7	4.1×10^7
8-rod lot	0.10	2.2	--	4.1×10^9	8.8×10^9	2.1×10^9	5.5×10^{10}	3.4×10^8	1.9×10^{10}	--	1.3×10^8

Table 2.5. Material Balance: Transuranium Element Recovery

	Alpha Counts		Aluminum	
	(counts/min)	(% Raw Feed)	(moles)	(% Raw Feed)
Plutonium Recovery Waste Effluent:				
Raw Waste Feed (in)	7.3×10^{14}	100	1550	100
Prepared Feed (out)				
(a) Discharged to waste	1.6×10^{14}	22	490	32
(b) Sent to ion exchanger	4.6×10^{14}	63	1080	70
Total Prepared Feed	6.2×10^{14}	85	1570	102
Unaccounted for loss (gain)	--	15	--	(2)
Ion Exchange System Losses				
Loading	6.8×10^{13}	9		
Scrubbing	3.0×10^{13}	4		
Piping leak	3.3×10^{13}	5		
Total losses	13.1×10^{13}	18		
Product Recovery				
Recovered product	2.3×10^{14}	31		
Recoverable product	0.6×10^{14}	8		
Total product	2.9×10^{14}	39		
Total accounted for in ion exchange system	4.2×10^{14}	57		

SK resin was used, a steady increase in plutonium losses, coupled with an increasing pressure drop across the resin bed, was observed. The resin was replaced with a fresh lot after receiving an estimated dose of 4×10^8 rads during one period of operation with the 24 fuel rods; losses and the pressure drop across the resin bed then returned to levels observed earlier. The spent resin was black, soft, and had lost almost 35% of its "salt splitting" capacity. A second lot of exposed (4.6×10^7 rads) resin used during the period of recovery of the 8-rod lot of fuel rods, was found to contain sorbed Al, Pu, Ru^{106}, Rh^{106}, and Pd that resisted all efforts at elution even with the strongest reagents, including aqua regia.

The radiation damage to the Dowex 1 X-10 resin used in transuranium element recovery occurred (qualitatively) at a higher rate, requiring renewal of the resin after each lot of feed had been processed. The appearance of the spent resin was similar to that observed in the plutonium-recovery phase of the program in that the resin was oily, tacky, jet black, and retained traces of Al, Ru^{106}, Rh^{106}, and Pd. None of these could be eluted even with aqua regia.

3. FLOWSHEETS

Development and modification of the recovery process continued throughout the program, resulting in a recommended chemical flowsheet that gave good recovery of high-purity products. Only minor modifications to previously existing processing equipment were made to adapt it to the process used, but the two ion-exchange columns were equipment specifically installed for this program.

3.1 Chemical Flowsheet

The chemical flowsheet developed during this program and recommended for future processing of highly irradiated Pu-Al alloy fuels to recover residual plutonium and other transuranium elements is shown in Fig. 3.1.

3.2 Equipment Flowsheet

The equipment in which the work was conducted is located in cell 1, Building 4507. Many of the components served dual purposes during different phases of the program. All equipment was made of stainless steel; process piping was of an all-welded design to minimize corrosion and leakage. The arrangement of the equipment as used in the plutonium-recovery phase is shown schematically in Fig. 3.2 and, as used in transuranium element recovery, in Fig. 3.3.

4. PROCEDURE

The procedure followed in recovering plutonium and the other transuranium elements included feed preparation, ion exchange sorption of plutonium and its recovery, evaporation of the ion exchange effluent and its adjustment, ion exchange sorption of the transuranium elements (and rare earths) and their recovery, and final concentration of the transuranium product for storage. Short descriptions of the fuel, its history, and the shipping cask are included as an introductory background preceding the detailed procedures which follow.

4.1 Feed Preparation

The preparation of feed for the ion exchange system included charging the fuel rods to the dissolver, dissolution of the plutonium-aluminum alloy

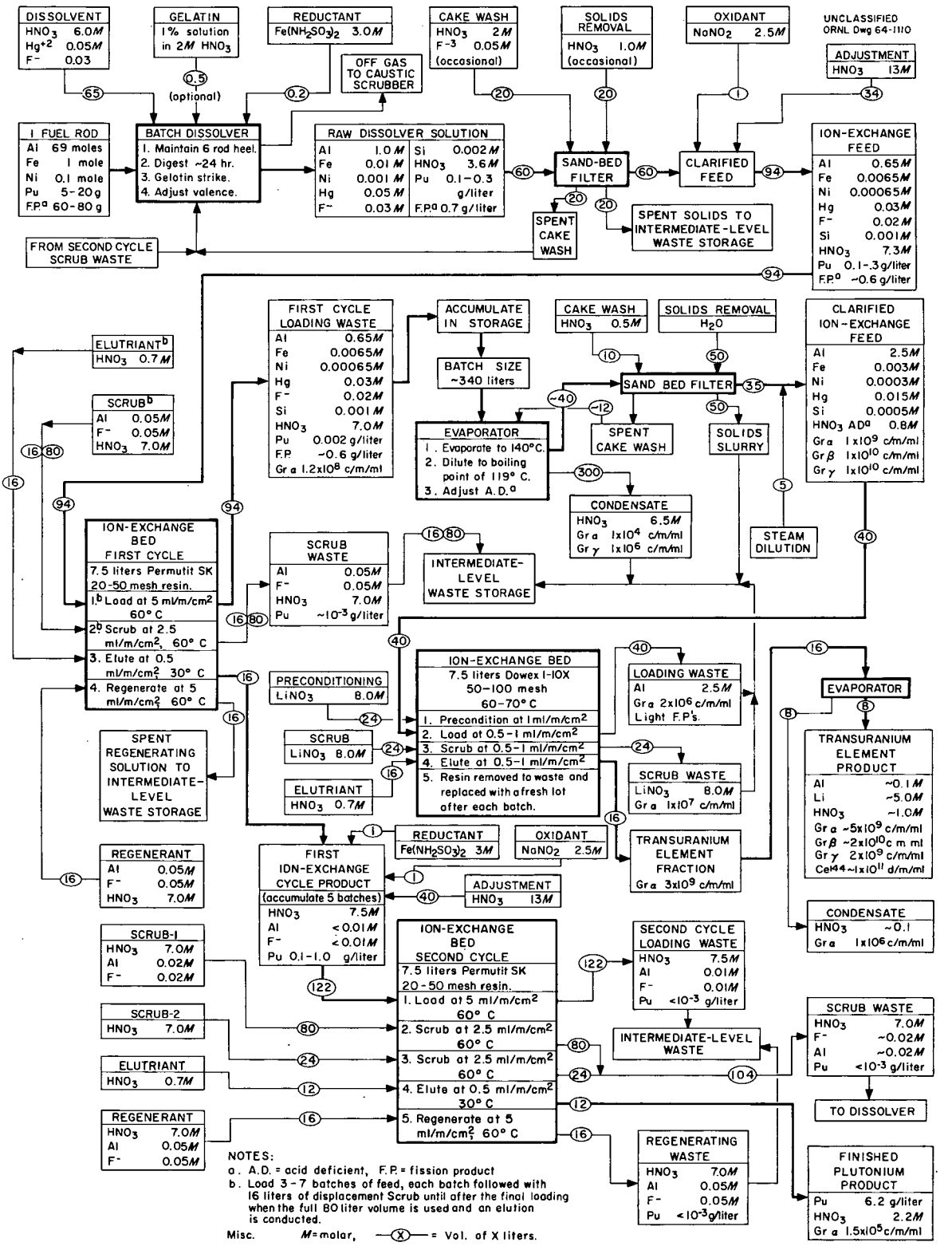

Fig. 3.1. Chemical Flowsheet For Recovery of Plutonium and Transuranium Elements From Irradiated Plutonium-Aluminum Alloy Fuel Rods.

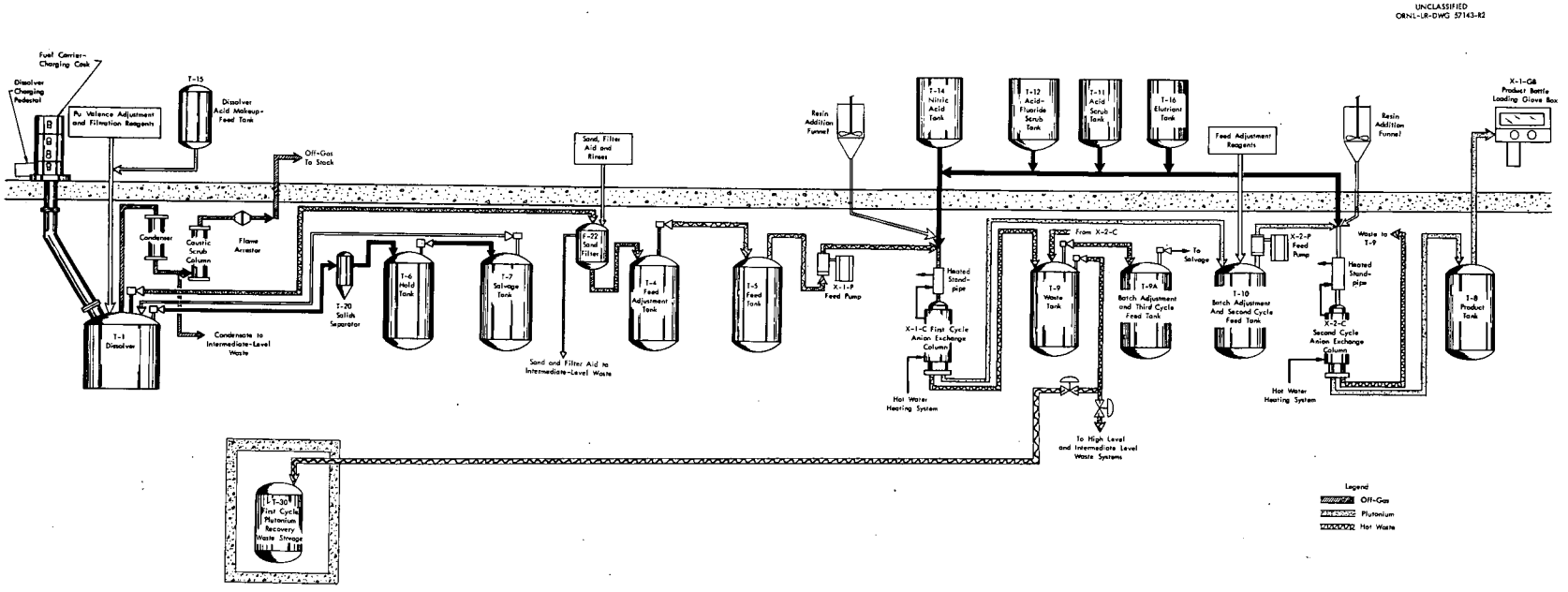

Fig. 3.2. Equipment Flowsheet for Recovery of Plutonium From Irradiated Plutonium-Aluminum Alloy.

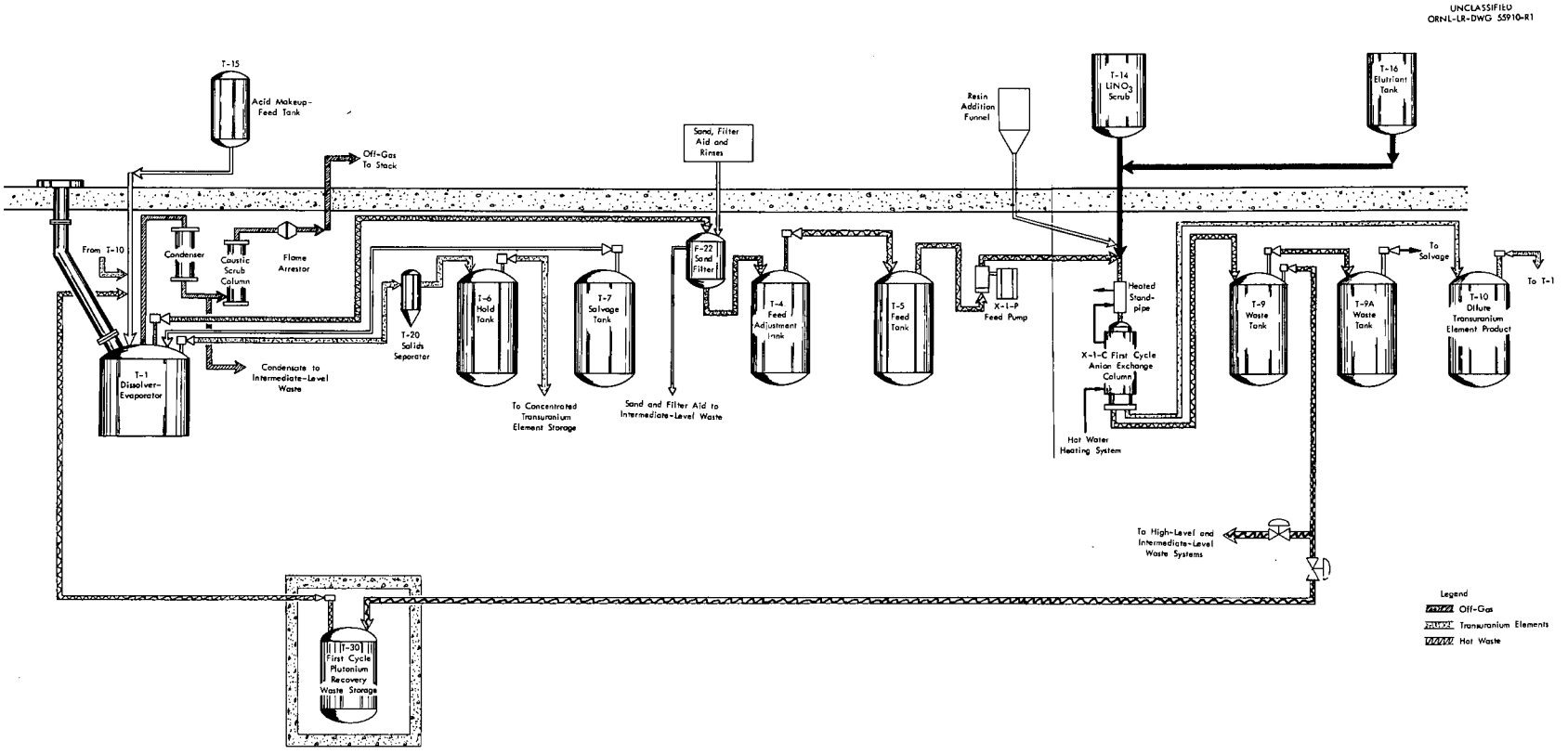

Fig. 3.3. Equipment Flowsheet for Transuranium Element Recovery From Irradiated Plutonium Recovery Wastes.

and the cladding, plutonium valence adjustment, clarification and final adjustment of the acid concentration.

4.1.1 Description and History of the Fuel

The fuel received for processing consisted of two lots of 0.94-in-diam by 5-ft-long aluminum-clad rods. One lot, comprising 24 rods, had been irradiated to about 87% burnup and had decayed for about 200 days; the other lot, eight rods, had been irradiated to greater than 99% burnup and decayed for about 100 days.

The 5-ft rods were half-sections of a longer rod that had been made from two lengths of 6% Pu-Al alloy rod co-extruded in an aluminum cladding such that at each end of the canned co-extruded rod a 6-in-long aluminum section was formed. Two sections of alloy rod were joined (forming an aluminum spacer section plug) and the assembly irradiated. Following this and after the decay period, the 10-ft rods were severed within the aluminum spacer section and shipped.

4.1.2 Shipping Cask Description and Handling

Shipments were made in a 10-ton cask (Fig. 4.1) providing 10-in of lead shielding. The cask, which also served as a dissolver-charging component, was equipped with a 24-slot magazine, the slots being in three concentric rows, that was mounted in the cavity and from which the fuel rods could be dropped individually into the dissolver charging chute. The cavity of the cask was equipped for water cooling and could be made either liquid and gas tight or vented into a flash chamber via a relief valve (set at 17 psig) and a glass wool filter. The temperature of the

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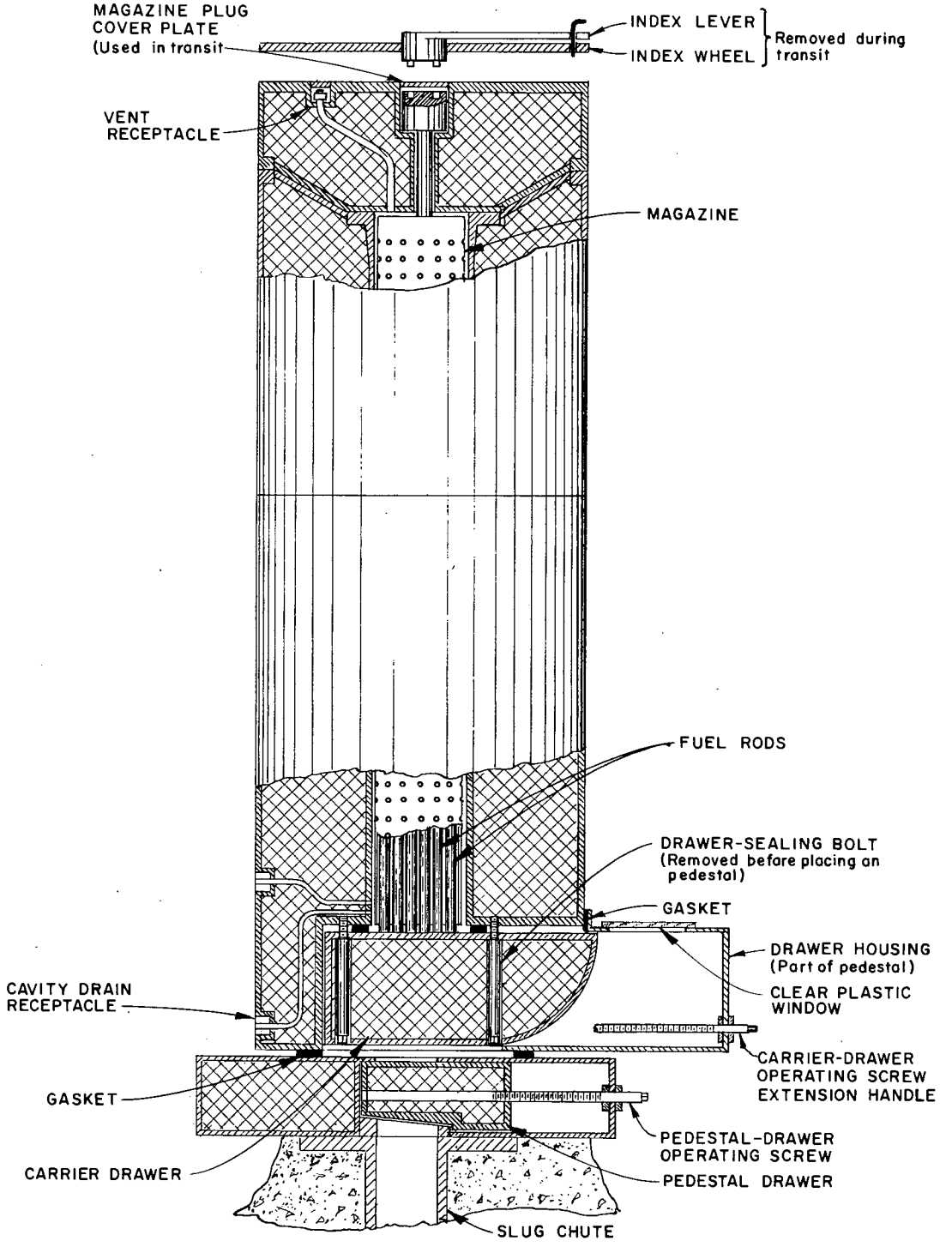

Fig. 4.1. Shipping Cask for Irradiated Plutonium-Aluminum Alloy Fuel in Charging Position on Slug Chute Pedestal.

cask cavity could be measured with the thermocouple mounted in the shielding. During shipment of the fuel rods by trailer truck, the cask rested, drawer-end down, in a cradle that kept the longitudinal axis of the cask at 15° to the horizontal. Water, which filled about 65% of the cavity, partially covered the rods for cooling. Upon arrival here and after inspection, the flash chamber and the relief-valve assemblies were removed, and the cask was moved to the containment area above the cells for temperature observation. After standing 24 hr, the surface temperature of the loaded cask was 7 and 12°, respectively, above ambient temperature (26°C) when loaded with the 24- and 8-rod shipments. The cavity water was then drained (after sampling) to a cell vessel and transferred to the intermediate-level-waste storage system. The cover plates were removed from the drawer and magazine-operating plug, after which the bolts holding the top of the drawer against the gasket in the cavity were removed. The cask was then moved to the slug-chute pedestal and positioned to align the discharge port of the cask with the charging port in the pedestal. A tight seal between the cask and the pedestal was obtained by a soft gasket mounted in a groove in the pedestal plate that pressed against the smooth bottom sides of the cask. The drawer-operating screw in the cask was then engaged with the extension handle, which extends through the housing on the pedestal that encloses the cask drawer when in the withdrawn position, after which the discharge of fuel from the cask could then begin.

4.1.3 Charging the Fuel to the Dissolver

After the cask had been positioned on the slug-chute pedestal and the indexing mechanisms attached, the index arm was moved to place the desired

magazine slot over the discharge port in the cask drawer. The pedestal drawer was then withdrawn by turning the operating screw, moving the cask drawer to point where the discharge port was aligned with the desired magazine slot. The fuel rod dropped by gravity to the charging chute and into the dissolver. Both drawers were closed after the dissolver was charged.

The initial charge of fuel rods to the dissolver was 6 rods. Thereafter, one rod was charged per completed dissolving to maintain a suitable undissolved-metal inventory in the dissolver and obtain a practical dissolution rate.

4.1.4 Dissolution of the Fuel

Dissolution of the fuel rods was conducted in a dissolver (Fig. 4.2) equipped with a down-draft condenser and two sets of coils; the upper coil was for cooling, while the lower one was supplied with either steam or cold water. A later revision of the piping, because of a leak in the lower coil, permitted the use of either set of coils for heating or cooling (Sec. 4.3.3.1). The off-gas leaving the condenser flowed to a 1-in Raschig-ring-filled scrubber into the top of which 1 M NaOH was metered. The non-condensable fraction of the off-gas passed from the scrubber via a flame arrester to the off-gas header serving the facility. The off-gas pressure in the dissolver was controlled between 0 and 5 in. of water gage below cell pressure. Control of the steam flow rate to the coils was maintained by a temperature controller that monitored the vessel temperature.

Condensate from the condenser could be returned to the dissolver or diverted to a collection tank. The initial dissolving in a program began

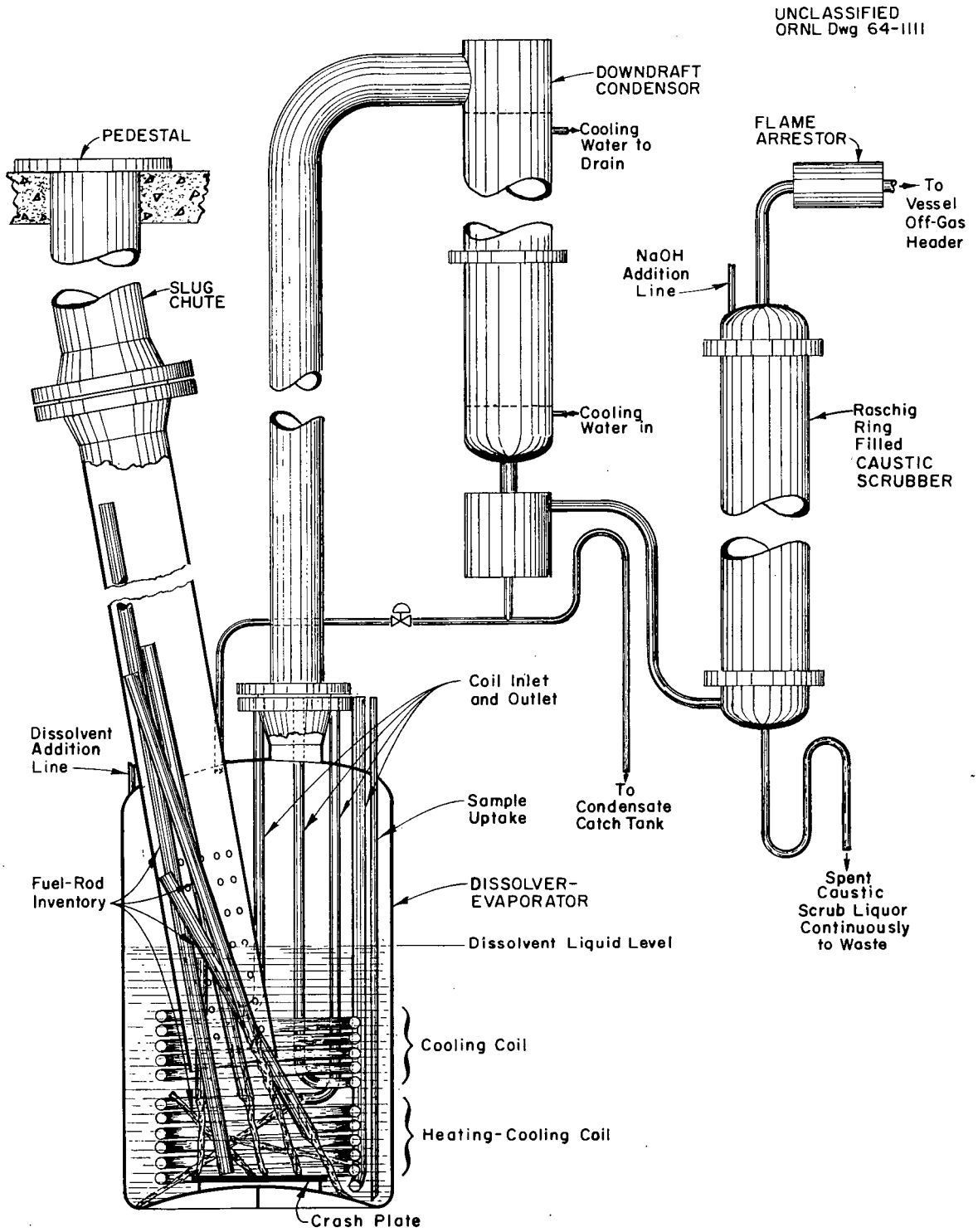

Fig. 4.2. Dissolver-Evaporator System Showing Fuel Rod Inventory at Equilibrium.

with the charging of six fuel rods to the dissolver, which contained 25 liters of 6 M HNO_3. Upon completion of the charging operation, the temperature was raised to 60°C, at which time the addition of 40 liters of 6 M HNO_3 containing 0.082 M Hg^{2+} was begun at 6.5 liters/hr and continued while raising the temperature to 100°C. Dissolution of the aluminum end pieces began at 95 to 100°C and became rapid enough with the six-rod charge to require cooling water in the coils to control the reaction rate. After all of the second lot of the dissolvent had been added and the initial rapid reaction had subsided, the contents of the dissolver were digested at 100 to 102°C for 24 to 32 hr, or until the acid concentration decreased to about 2.5 M HNO_3, at which concentration the dissolution rate was low. The first lot of dissolver solution of a new series usually contained an abnormally high concentration of aluminum along with a low concentration of plutonium; these solutions were transferred from the dissolver earlier than usual because dissolution of the aluminum end pieces consumed the bulk of the HNO_3 before appreciable exposure and dissolution of the Pu-Al alloy could take place. In such cases, after removal of the first dissolver solution, the second dissolving of a series began with the addition of 65 liters of 6.0 M HNO_3--0.05 M Hg^{2+}--0.03 M F^- to the dissolver over a 2-hr period while keeping the temperature as low as possible with cooling water. The temperature was then carefully raised to 95 to 100°C to start the dissolution. After the vigorous reaction began, cooling was required for 0.5 to 1 hr, after which heating was resumed and the temperature kept at 100 to 102 for 24 to 36 hr, or until the metal equivalent to one rod had dissolved.

During all subsequent dissolvings, one fuel rod was charged to the dissolver containing the residue of all rods previously added in the series. Sixty-five liters of 6 M HNO_3--0.05 M Hg^{2+}--0.03 M F^- was then metered to the dissolver at 30 to 40 liters/hr while controlling the temperature with cooling water at 95 to 100°C. After an initial reaction surge that required cooling water, the reaction rate decreased; thereafter, the contents were kept at 100 to 102°C for 24 to 30 hr until the metal equivalent to one rod had dissolved.

After the last rod of a series had been added, dissolvings were continued with fresh dissolvent as before, until no significant amount of plutonium appeared in the dissolver solution.

Vapor evolved during dissolution passed to a downdraft condenser where the condensable components were condensed and returned to the dissolver. The noncondensable fraction of the off-gas passed from a vapor-liquid separator below the condenser to a 6-in-diam by 40-in-long caustic scrubber packed with 0.5-in. Raschig rings. The acid components of the off-gas were sorbed in 1 M NaOH metered at 3 to 4 liters/hr to the caustic scrubber. Spent caustic liquor from the scrubber was continuously discharged to the intermediate-level waste storage system without sampling. The residual, noncondensable, acid-free off-gas passed from the scrubber via a flame arrestor to the off-gas header.

A controlled partial vacuum (3 to 4 in., water gage) was maintained in the dissolver at all times. A continuous air purge of 1.5 cfm introduced to the slug chute kept it free of vapor and diluted any explosive radiolytic gases generated in the dissolver to safe concentrations.

4.1.5 Dissolution-Rate Studies

Only 20 in. of the 5-ft length of the fuel rods was exposed to the dissolvent at any time. Previous dissolution studies with inactive 1-in-diam aluminum rods had showed that dissolution progressed along the submerged section of the rod until the thinned section collapsed under its own weight, exposing a new section. The collapses were periodic rather than continuous. The attacked section of the rod assumed the shape of an icicle.

The total dissolution of a single fuel rod was conducted to determine the aluminum-to-plutonium ratios and to obtain specific dissolution-rate data. In this study, the rod was exposed to a single lot of dissolvent until about 50% of the aluminum had dissolved, at which time the spent dissolvent was replaced with a fresh lot and the dissolution resumed and continued until no further aluminum or plutonium was dissolved. The dissolver solution was sampled periodically and analyzed for plutonium and aluminum, from which data the quantity of dissolved materials were determined and plotted as a function of time (Fig. 4.3). Relating these data to the exposed surface of the rod during the time interval gave an average overall specific dissolution rate of about 0.45 mg min^{-1} cm^{-2} for aluminum and 0.005 mg min^{-1} cm^{-2} for plutonium. The aluminum-to-plutonium ratio over the range of this run was 120, compared with the theoretical 64, indicating that all of the plutonium probably had not dissolved.

Similar data were obtained during two normal production (equilibrium) runs, one dissolving material from the 87% burnup lot of fuel and the other from the 99% burnup lot. In these, a fresh rod was charged to the dissolver

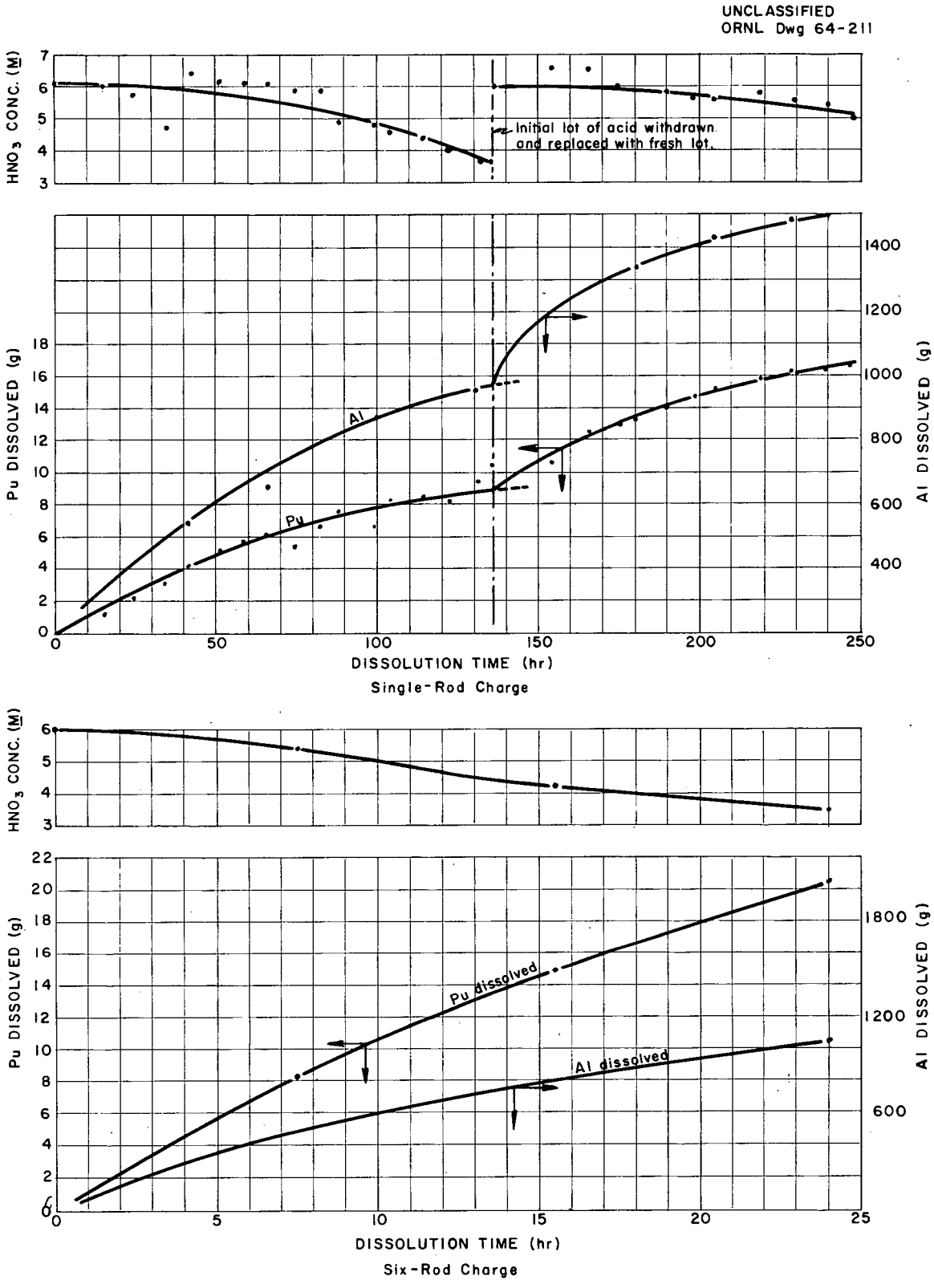

Fig. 4.3. Typical Dissolution Rates.

already containing residual metal from the previous dissolvings of the series. Data from these two runs when related to the surface exposed to the dissolvent as a function of time gave specific dissolution rates for the alloy of 0.8 and 0.7 mg min^{-1} cm^{-2}, respectively. The dissolution rate of the fuel dissolved in this program, although low, agrees well with that predicted from similar data plotted in Fig. 4.4.

4.1.6 Feed Clarification and Adjustment

After the desired amount of plutonium had been dissolved, optionally, sufficient 1% gelatin solution was added to obtain a 10-ppm concentration of gelatin in the dissolver solution, after which the mixture was digested for 1 hr at 80°C to coagulate silica. The plutonium valence was reduced to three by the addition of a reducer, 50% ferrous sulfamate solution (2.9 N), after which the solution was filtered through a 12-in-diam by 1-in-deep bed of Ottawa sand that had previously been coated with filter aid. The valence of the plutonium in the filtrate was adjusted to four by oxidizing with 2.5 M $NaNO_2$ added to it, followed by digestion at 50 to 60°C for 0.5 hr. On occasions, the final valence adjustment was completed prior to filtration, without noticeable effect on the process. It was important, however, to allow sufficient time for the nitrous gases evolved in the reduction of the nitrite to be discharged from the solution prior to passing it to the resin beds; this minimized gas binding and the formation of gas-filled voids within the bed. Such conditions promoted channeling of the solution through the bed, with consequently higher plutonium losses.

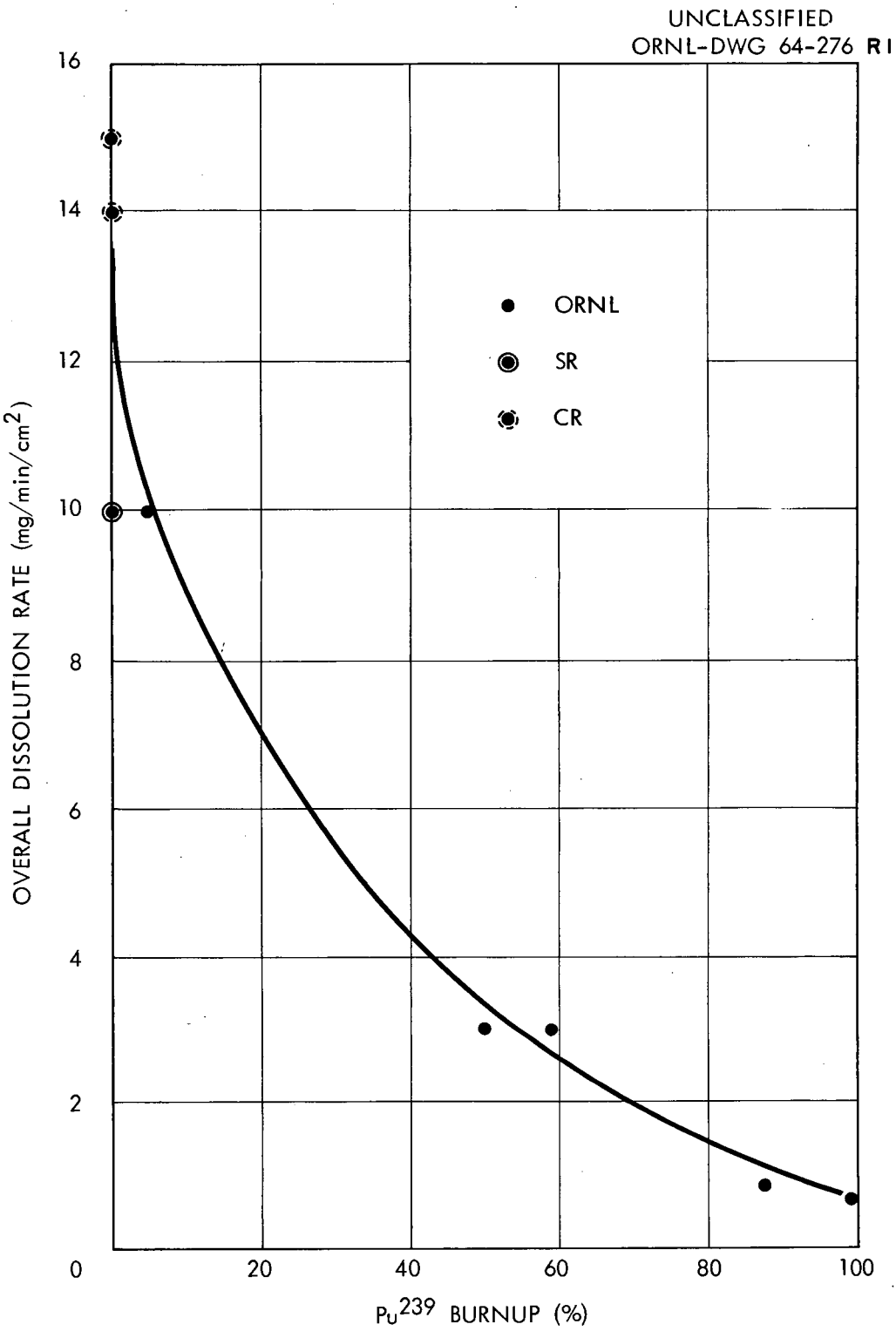

Fig. 4.4 Plant Dissolution Data. Pu-Al alloy, 6.0 M HNO_3 - 0.05 M Hg^{2+} - 0.03 M F^-. Data from Savannah River (1), Chalk River, and Oak Ridge National Laboratory (2, 3, 4, 5, 6).

The acidity of the feed solution was adjusted to 7 M HNO_3 prior to passing the feed to the anion exchange resin. This was done in two ways: In the first four runs, the feed stream was continuously metered to the resin-column head pot, where it was mixed with a metered stream of 13 M HNO_3 such that the net acid concentration was 7 M HNO_3. This procedure was abandoned in favor of a batch adjustment because of gas evolution that occurred when the acidity was increased and it developed that the holdup time in the head pot was insufficient for the acidified stream to discharge itself of the evolved gases.

4.2 Plutonium Recovery

Highly purified plutonium was recovered from the adjusted feed solution in a two-stage process by sorption on chloride-free anion exchange resin. Important steps in each stage included preparation and handling of the resin, charging resin to the system, loading plutonium on the resin, scrubbing and elution of the sorbed plutonium, and storing the finished product.

4.2.1 Ion Exchange

Two stages of ion exchange purification were required to obtain a plutonium product that met radiation specifications. The preparation of chloride-free ion exchange resin and its charging to the columns followed by conversion to the nitrate form were necessary preliminary steps in the operation. Occasional removal of degraded resin from the columns by upflow displacement with 2 M $Al(NO_3)_3$ was a technique developed to permit changing out the resin remotely.

4.2.1.1 Resin Preparation. – The resin used in plutonium recovery consisted of 20- to 40-mesh Permutit SK. Received in the chloride form, it was converted to the nitrate prior to use by digestion in 7 M HNO_3 at room temperature for 2 to 3 hr, after which the spent acid was drained away. The treated resin was washed with demineralized water until free of chlorides, after which it was stored as a resin slurry until charged to the column.

4.2.1.2 Charging Resin to the Column. – The chloride-free resin was charged to the column as a very dilute slurry, dilution water being added to keep the resin particles swept along the piping toward the column during the operation. The slurry was added to a funnel fitted to the resin-charging line and was continuously stirred to keep the resin in suspension. A small stream of water was played over the surface of the resin in the container, sluicing a light slurry of resin to the addition funnel, which was kept half full of liquid at all times. If the slurry in the funnel became too heavy, addition of resin to it was halted. The diluted slurry passed to the top of the heated standpipe and fell through it into the column. Excess water flowed from the bottom of the column through a sealing leg ("jack leg") and diversion valves to the waste collection tanks.

After 7.5 liters (settled volume) of resin had been charged to the column, the addition funnel and lines were flushed thoroughly with water and the funnel assembly removed and the charging line capped.

Before placing the column in service, pressure-drop measurements across the resin bed were determined as a function of flow rate, both with water and with 7 M HNO_3. These data when compared with similar results using

geometrically identical glass columns were useful in determining whether the resin-bed depth was satisfactory and whether the column had been overfilled or not. The 7 M HNO_3 also served to precondition the resin to receive feed. In practice it was found that in removing spent resin from the column (Sec. 4.2.1.3) 0.5 to 1 liter of resin remained. When this happened, the addition of 8 liters of fresh resin would establish a bed that completely filled the column and also established a small-diameter bed extending up into the standpipe.

4.2.1.3 Removing Spent Resin From the Column. — Spent resin, degraded from chemical and radiation exposure, was removed from the column by pumping 16 liters of 2 M $Al(NO_3)_3$ to the bottom of the column below the resin bed via the seal-leg assembly at 15 to 20 liters/hr. The resin bed was displaced upward (more or less intact) to the airlift intake located about at the midpoint of the standpipe. A stream of flush water was continuously added to the top of the standpipe at 30 liters/hr to keep any resin flushed down from the upper section and also to dilute the resin-slurry as it passed to the airlift. The airlift discharged to a waste-collection tank, which was continuously sparged with air to keep the resin suspended. Additional dilution water was also continuously added to the collection tank. The dilute spent-resin slurry was continuously transferred from the collection tank by steam jet to the intermediate waste system, the waste header also being continuously flushed with a stream of water to help keep it swept free of resin. Except for those instances where solids had collected on top of the resin bed during loading operations, effectively reducing the throughput rate and increasing the exposure to radiation and consequent degradation, the technique was simple and worked well.

Upon completion of the 2 M $Al(NO_3)_3$ displacement of spent resin, the column was flushed with water, and a second displacement with 2 M $Al(NO_3)_3$ was made to effect as complete a removal of spent resin as possible.

4.2.1.4 Changing Types of Resin. – In studies with a resin column mockup, it was found that 7 to 7.5 liters of an 8-liter charge was removed in two displacements. Additional displacements removed insignificant quantities of spent resin.

To effect the complete removal of one type of resin prior to charging another type, and to obtain the minimum cross contamination between the two, it was necessary to partially recharge the column with about 3 to 4 liters of the new type of resin, then fluidize the bed with short bursts of water to the bottom of the bed to mix it thoroughly and then conduct a displacement procedure, removing the mixed resin. The partial charging-mixing-displacement procedure would be repeated several times until only insignificant amounts of the original resin remained, after which a full 7.5-liter charge of the new type of resin would be made.

4.2.2 First Stage Operation

The initial stage of purification began with the sorption of plutonium from the feed followed by scrubbing the loaded bed and elution of the product. Prior to reloading the next batch of feed, the resin was regenerated with 7 M HNO_3.

4.2.2.1 Loading. – The adjusted feed solution was metered at 5 ml min^{-1} cm^{-2} (20 liters/hr) to the resin column via the headpot. The rate was governed by the pressure drop across the resin bed and was steadily

reduced as the program advanced. The solution flowed by gravity to the 0.75-in-diam by 93-in-long standpipe, which was maintained at 60°C by circulating warm water through the jacket. A supplementary electrically heated section was mounted below, though it was rarely used. The required pressure head to drive solution through the 4-in-diam by 40-in-deep (7.5 liters) bed of Permutit SK (20 to 50 mesh) resin was supplied by the height of the solution in the standpipe. The resin bed was maintained at 60°C by circulating hot water in the jacket of the column. The resin bed was supported on a 100-mesh screen reinforced with two layers of 18-mesh screen, while the top of the resin bed was free.

Ion exchange loading wastes were collected in a vessel from which samples of the contents were removed for analyses. Depending on the plutonium content, the loading wastes were discharged to a 900-gal storage tank for later processing to recover the transuranium elements (Sec. 4.3), or transferred to a salvage storage vessel for reworking and plutonium recovery.

4.2.2.2 Scrubbing the Loaded Resin Bed. — Sorbed fission products and residual aluminum were scrubbed from the resin bed with 7 M HNO_3 metered to it at 2.5 ml min^{-1} cm^{-2} until 16 liters (2 bed volumes) of solution had passed. After three to five lots of feed had passed through the resin bed (with the 16-liter scrub following each), the 7 M HNO_3 scrub was extended to 80 liters total volume for more extensive removal of residual fission products and aluminum. After sampling the spent scrub solutions in the collection tank, these were discharged to the intermediate-level waste system or stored for salvage, depending on the plutonium content.

4.2.2.3 Elution. – The plutonium sorbed on the resin bed was eluted with 16 liters of 0.7 M HNO_3 metered to the column at 0.5 ml min^{-1} cm^{-2}, the temperature of the resin bed being maintained at 30°C.

4.2.2.4 Resin Regeneration. – Following an elution, the resin was regenerated at 60°C by metering 16 liters of 7.0 M HNO_3--0.01 M F^---0.01 M Al to the column at 5 ml min^{-1} cm^{-2}. Spent regenerating solution, usually low in plutonium, was discharged to the intermediate-level waste system or stored for salvage, depending on its plutonium content.

Whenever the resin bed was to be idle for any length of time, it was eluted and allowed to stand in 0.7 M HNO_3, with which chemical attack was negligible.

4.2.3 Second Stage

4.2.3.1 Feed Adjustment and Loading. – The plutonium product fractions eluted from the first-stage resin bed were accumulated until a suitable volume was on hand. The composited first cycle product was adjusted, first with ferrous sulfamate at 30 to 40°C, then with $NaNO_2$, and digested for 1 hr to ensure that the plutonium valence was four.

After the digestion at ambient temperature to allow the gases evolved to pass from the solution, the acidity of the feed was adjusted to 7 M HNO_3, and the adjusted solution was metered to a second ion exchange system at conditions identical to those in loading the first-stage resin bed. The second-stage column was identical to the first except that it was not equipped with a head pot.

4.2.3.2 Scrubbing the Loaded Resin Bed. — Residual contaminants sorbed on the resin were removed from the resin with 80 liters of 7 M HNO_3 - 0.01 M F^- metered to the system, held at 60°C, at 2.5 ml min^{-1} cm^{-2}. Scrubbing was continued with 24 liters more of fluoride-free 7 M HNO_3 under the same conditions to displace fluorides from the resin bed. These scrub solutions (which usually contained plutonium at concentrations justifying recovery), after sampling and determining the plutonium content, were stored for later use in preparing dissolvent.

4.2.3.3 Elution and Recovery. — Finished plutonium product was recovered from the resin by elution with 16 liters of 0.7 M HNO_3 metered to the column maintained at 30°C at 0.5 ml min^{-1} cm^{-2}. The first 2 to 4 liters of effluent displaced from the resin bed was routed to the scrubbing waste to obtain highest plutonium concentration in the product fraction; no appreciable loss was incurred by this practice. The finished product solution was collected in one of the smaller (35-liter) cell vessels where it was thoroughly mixed by air sparging and samples removed for analysis.

4.2.3.4 Regeneration of the Resin. — The resin bed was allowed to stand in 0.7 M HNO_3 until the next loading. Before loading again, the resin was regenerated at 60°C by passing 16 liters of 7.0 M HNO_3--0.01 M F^---0.01 M Al through the bed at 5 ml min^{-1} cm^{-2}. The spent regenerant solution was sent to the dissolver for dissolvent preparation or discharged to waste, depending on the plutonium content.

4.2.4 Handling of the Plutonium Product

The finished plutonium product solution, after preliminary results of

analyses were known, was withdrawn into tared polyethylene bottles which, after filling with 10 liters of solution and weighing, were bagged and placed in shipping containers. The bottles were 5 in. in inside diameter, with a minimum wall thickness of 0.125 in; the nominal capacity was 12 liters. The screwed bottle caps were fitted with a 0.125-in disk gasket of neoprene rubber backed with a 0.25-in layer of glass wool fitted under the cap. In storage of the product recovered during the 24-rod program, the cap assembly was pierced with a hypodermic needle to provide a vent for the escape of radiolytic gas. When it was found later that small quantities of the solution escaped via the puncture-vents, each cap assembly was fitted with a positive vent consisting of a hypodermic needle (protruding through the cap, glass wool, and gasket into the gas space above the solution) to which was attached a section of plastic hose leading to a 0.5-in-diam by 3-in-long glass wool filter mounted on the body of the bottle.

The shipping container consisted of a 6-in-diam by 60-in-long stainless steel cylinder fitted with a gas-tight flanged cover. Mounted on the cover was a valved vent line for attachment to an off-gas system for releasing radiolytic gases generated by the plutonium solution during storage and shipment. The stainless steel cylinder was permanently mounted in a 2-ft^2 by 5.5-ft-high plywood box that ensured a safe geometry for any array of containers during storage or shipment.

Immediately prior to shipment, and periodically during storage, the radiolytic gases generated in the product solutions were released to maintain a reasonably low pressure within the steel cylinder.

4.3 Transuranium Element Recovery

The transuranium elements are not sorbed on anion resin at the high acid concentration characteristic of the plutonium recovery process. At zero to 0.2 M HNO_3 in a medium 7 to 8 M in total nitrate, however, the rare earths and transuranium elements are sorbed strongly. The transuranium element recovery process is based on this principle, so that the initial step was to remove the free acid from the plutonium loading waste effluent by evaporation while increasing the aluminum concentration.

4.3.1 Feed Preparation

The preparation of feed consisted in evaporating the first-stage plutonium-loading waste effluent to about 140°C and then adjusting the acid deficiency and aluminum concentrations, after which the aluminum hydroxide formed during the evaporation was removed by filtration.

4.3.1.1 Evaporation of the Waste Solution. — The plutonium-loading waste effluent from the first plutonium recovery stage that had been accumulated and stored during that phase of the program (Sec. 4.2.2.1) was evaporated to 140°C in the dissolver vessel (Fig. 4.2), the piping of which was arranged such that the condensate could be returned to the vessel or diverted to a condensate collection tank. As originally installed, the upper set of coils in the vessel were piped for cooling water only and the lower set for either cooling water or steam. During the runs, however, the lower coil began to leak, making it impossible to conduct the evaporation to 140°C with that coil only. As a result, both coils were modified so that each could be used for heating or cooling, with the added provision

that a 15-psig positive air pressure would automatically be maintained on the coil at all times when steam or water to the coils was turned off. A second modification to the piping included rerouting the coil-discharge line from the process drain (normally nonradioactive) to a waste-collection tank from which samples could be withdrawn. This permitted evaluation of the leakage rate of material from the evaporator into the coil and ensured that no radioactive material found its way into the process drain.

The evaporation of the plutonium-loading waste effluent solution was conducted by transferring 65 liters to the dissolver and evaporating the mixture to small volume at normal pressure. Afterwards, 10- to 30-liter increments of fresh solution were transferred to the dissolver as the evaporation progressed, until 340 to 370 liters had been added, the volume of solution in the evaporator being held between 30 to 65 liters; the minimum volume under evaporation was held to 30 liters to minimize exposure of the heating coils to the vapor. After the last increment had been added, the evaporation was continued until the liquid temperature reached 140°C, at which time the volume of concentrate was about 30 liters, and a portion of the coil was exposed to the vapor. No difficulty in raising the final temperature to 140°C was encountered until the leak occurred in the lower coil, after which sufficient steam leakage into the vessel resulted in pressurizing it. After modifying the piping, mentioned above, the use of steam in both coils was required to reach 140°C, and during most of the last stage of the evaporation the upper coil was exposed to the vapor. The marked increase in the amount of solids in the concentrate observed after the modifications to the coils was made was attributed to the drying

of solids on the heated surfaces exposed to the vapor during the latter stages of the evaporation, coupled with the excessive steam stripping of the HNO_3 from the concentrate that was in equilibrium with the dissolved $Al(NO_3)_3$.

After the concentrated solution had been taken to 140°C, it was diluted with water to a reflux temperature of 119°C (approximately 3.2 to 3.3 M Al). The acid deficiency was about 1 to 1.2 N before the modification to the coils were made, but increased afterward to 1.5 to 1.8 N. While an acid deficiency of 0.5 to 1.0 N was desirable to prevent crystallization of the $Al(NO_3)_3$, at the higher acid deficiency, a colloidal suspension of $Al(OH)_3$ was present that was not removed completely by filtration and continued to precipitate slowly during subsequent ion exchange operations, resulting in plugging the resin bed. Sorption losses, however, apparently were not increased significantly at the higher acid deficiency.

4.3.1.2 Clarification. – The diluted solution was sampled with difficulty because of the high solids content; during about 50% of the program the evaporator sampler was inoperative because of plugging with solids. The solids present in the diluted solution were only partly removed by filtration at 60 to 80°C through a 12-in-diam by 1-in-deep bed of Ottawa sand precoated with filter aid. Filtration was slow and incomplete. Apparently precipitation of finely divided $Al(OH)_3$ continued after the solution had been filtered. Solutions which were adjusted to approximately 0.5 N acid deficient filtered more readily than those of 1 to 1.5 N acid deficiency.

After filtration and sampling, the filtrate was transferred by steam

jet to a feed hold tank. The filter cake after draining as thoroughly as possible was fluidized by backwashing with water and discharged from the filter to the intermediate-level waste storage system.

Material balances completed about the filter in early runs showed that 25 to 40% of the aluminum and transuranium elements were retained in the filter. These materials were recoverable to greater than 95% by washing the cake with two 20-liter volumes of 0.5 M HNO_3 and returning the spent wash to the succeeding lot of waste to be evaporated. Owing to the lack of intermediate storage vessels for holding the spent wash solution for later salvage and piping limitations of the system, only two demonstrations of this procedure were completed.

Routinely, final adjustment of the filtrate was not necessary since the $Al(NO_3)_3$ concentration in the feed was desired at the highest level obtainable (2.5 M minimum). It was difficult to achieve this concentration without the $Al(NO_3)_3$ crystallizing in both the filter and collection tanks as the solution cooled to below 60°C (none of the process vessels other than the evaporator was equipped for heating) because of the dilution with condensed steam that operated the transfer jets during the two transfer operations that were required.

4.3.2 Ion Exchange

Ion exchange operations began with converting the resin to the chloride-free nitrate form, and charging it to the column, after which the resin was then conditioned with 8 M $LiNO_3$. Recovery of the transuranium elements from the feed was then begun followed by scrubbing and elution. The spent resin was removed at the end of each run and replaced with a fresh lot.

4.3.2.1 Resin Preparation. – The resin used in the recovery of the transuranium elements was 50- to 100-mesh Dowex 1 X10. Before use, the resin was freed of chlorides by repeated digestions in an equal volume of 8 M HNO_3 at 25 to 30°C, until no chloride was detected in the acid, washing with demineralized water following each digestion. Complete removal of residual chloride from the resin was attained by successively digesting it with equal volumes of 8 M $LiNO_3$ at 90 to 100°C until a negative test for chlorides was obtained, again washing with demineralized water after each digestion. The chloride-free resin was stored as a water slurry until charged to the column.

4.3.2.2 Charging Resin to the Column. – After conducting repeated resin-removal procedures (Sec. 4.2.1) on the first cycle column to minimize cross contamination with spent residual Permutit SK resin used in the plutonium recovery operation, chloride-free Dowex 1 X10 resin was charged to the column according to the procedure described in Sec. 4.2.1.2. The pressure-drop measurements were then conducted, but, in this case, using water and 8 M $LiNO_3$ instead of water and 7 M HNO_3.

4.3.2.3 Conditioning the Resin. – The Dowex 1 X10 resin was preconditioned for loading transuranium elements by metering 24 liters of 8 M $LiNO_3$ at 5 mil min^{-1} cm^{-2} to the column held at 60 to 70°C.

4.3.2.4 Loading. – The adjusted feed solution was metered to the column held at 60 to 70°C at 0.5 to 1.0 ml min^{-1} cm^{-2}, depending on the pressure drop. At low flow rates (less than 0.5 ml min^{-1} cm^{-2}), the resin that floated in both the 8 M $LiNO_3$ and the feed solution tended to rise

into the standpipe, resulting in a deeper bed, a significant fraction of which then was of smaller cross section area. Consequently, there was a higher pressure drop. While the lower flow rates were desirable from the standpoint of sorption and low losses, practical considerations dictated maintaining the rates high enough to hold the resin bed in the column. A second harmful result of low flow rate, stemming from the smaller effective cross section of that portion of the bed in the standpipe, was that the column plugged rapidly with aluminum hydroxide that continued to deposit from feed solutions after filtration.

The loading waste from the ion exchange bed was collected in a continuously air-sparged collection tank, samples being withdrawn at intervals during the loading operation to detect losses. The waste was discharged to the intermediate-level waste storage system or reworked depending on the transuranium element content. Because of the developmental nature of the program, as well as the lack of adequate salvage storage capacity, much of the loading waste solution that ordinarily would have been reworked to recover the transuranium element content was discharged from the process to the intermediate level waste storage.

4.3.2.5 Scrubbing the Loaded Resin Bed. — Residual aluminum-bearing solution and sorbed aluminum and light fission products were displaced and scrubbed from the resin with 24 liters of 8 M $LiNO_3$ metered to the column, held at 60 to 70°C, at 0.5 to 1.0 ml min^{-1} cm^{-2}. The spent scrubbing waste flowed to a continuously air-sparged collection tank from which samples were periodically withdrawn to determine the progress of the operation. The spent scrub solution was discharged to the intermediate-level waste

storage system. Because of the incompatability of lithium and aluminum nitrates in mixtures at the concentrations necessary in the feed for this process, reworking these solutions was not generally attempted when significantly high losses occurred.

4.3.2.6 Elution. – After scrubbing the resin almost free of aluminum and light fission products, the sorbed transuranium elements (and rare earths) were eluted from the bed, held at 60 to 70°C, with 16 liters of 0.7 M HNO_3 metered it at 0.5 ml min^{-1} cm^{-2}. A 2- to 4-liter displacement fraction, routed to the spent scrubbing waste collection tank, was taken. Then the product stream was diverted to a continuously air-sparged product-collection tank from which samples were withdrawn at intervals. All the product from an individual lot of fuel rods was collected in a single vessel, so that the progress of the elution of an individual loading of the resin involved taking into account the material recovered from all previous loadings, a practice that decreased the precision of recovery results of any individual run.

4.3.3 Transuranium Element Product Handling

The dilute transuranium element product solution recovered by elution from the ion exchange resin was concentrated by evaporation and stored for future use.

4.3.3.1 Evaporation. – Upon completing the elution of the last lot of transuranium element product from the resin, the dilute product was transferred to the evaporator and concentrated to about 50% of its original volume and transferred to storage.

4.3.3.2 Storage. — After sampling the product concentrate in the evaporator, it was transferred by steam jet to water-jacketed storage tanks located in a separate cell. The evaporator and an intermediate tank in the routing to the storage tanks were flushed twice with three small (5-liter) lots of 0.5 M HNO_3, the spent wash solutions being collected in the storage tanks along with the product solution. The final volume of the stored product including the washes and steam condensate used to operate the transfer jets was about equal to that of the original dilute product before concentration.

5. APPENDIX

Processing data accumulated during the program are tabulated in this section. Results of chemical and physical examinations of the spent resin from the plutonium recovery steps of the process are given also.

5.1 Detailed Process Data

Detailed chemical and radiochemical data relating to dissolution of the individual fuel rods and the feed solutions prepared from these are given in Tables 5.1 and 5.2. Similar data for the spent waste and scrubbing solutions are given in Tables 5.3 and 5.4, respectively. The composition of the second-cycle feed solution is given in Table 5.5; those of the loading and scrubbing waste from the second ion exchange cycle are given in Tables 5.6 and 5.7, respectively.

The composition of the final plutonium product is given in Table 5.8; that of the stored first stage plutonium recovery wastes is given in Table 5.9. The compositions of the evaporator condensate and the ion exchange feed prepared from the plutonium recovery wastes are given in Tables 5.10 and 5.11, respectively; those of the loading and scrubbing wastes collected during transuranium element recovery are given in Tables 5.12 and 5.13. The composition of the transuranium element product fractions are given in Table 5.14; typical alpha-energy analyses are given in Table 5.15. Typical compositions of the condensate collected during the evaporation of the dilute transuranium element product are given in Table 5.16. A detailed material balance based on alpha counting and aluminum analyses is given in Table 5.17.

Table 5.1. Composition of Dissolver Solutions During Plutonium Recovery

Dissolving Batch No.	Volume (liters)	HNO_3 (M)	Al (g/liter)	Counting Rate (counts min^{-1} ml^{-1}) Pu α	Gross α	Gross γ	Dissolution Time (hr)	Remarks
				24-Rod Lot				
1[a]	63	3.6	15	3.1×10^7	1.0×10^8	1.6×10^9	140	1-rod charge
1A[a]	56	5.2	5.8	2.0×10^7	6.7×10^7	1.1×10^9	120	
2	60	2.3	28.5	1.3×10^7	5.1×10^7	7.0×10^8	13	6-rod charge
2A	57	4.0	20.7	4.1×10^7	1.4×10^8	2.3×10^9	24	Residual heel
3	58	2.7	25.6	4.4×10^7	1.4×10^8	2.3×10^9	22	1 additional rod
4	56	3.5	19.0	6.5×10^7	2.0×10^8	3.2×10^9	26	1 additional rod
5	60	4.1	--	4.7×10^7	1.7×10^8	3.1×10^9	24	
6	61	2.6	--	4.3×10^7	1.8×10^8	--	26	
7	71	--	--	--	--	--	24	
8	57	4.8	--	6.3×10^7	--	--	26	
9	53	1.9	--	5.3×10^7	--	--	23	
10	69	5.1	--	6.9×10^7	--	--	6	
11	60	4.8	--	6.2×10^7	--	--	27	
12	59	2.9	--	4.7×10^7	--	--	23	Residual heel + 1 rod
13	63	3.6	--	5.8×10^7	--	--	22	
14	63	2.3	--	6.3×10^7	--	--	17	
15	62	2.6	--	7.3×10^7	--	--	17	
16	60	2.7	--	7.7×10^7	--	--	17	
17	63	4.1	--	8.7×10^7	--	--	18	
18	61	2.1	--	7.1×10^7	--	--	17	
19	61	3.4	--	7.1×10^7	--	--	16	
20	58	5.5	--	9.2×10^7	--	--	26	
21	64	4.6	--	5.5×10^7	--	--	23	
22	61	4.5	--	4.5×10^7	--	--	16	
23	56	2.8	--	5.9×10^7	--	--	23	
24	61	4.7	--	5.6×10^7	--	--	22	
25	54	3.7	--	6.4×10^7	--	--	23	
26	66	3.1	--	8.9×10^7	--	--	47	
27	63	4.3	--	5.8×10^7	--	--	24	Residual metal heel for remaining rods
28	59	3.3	--	4.9×10^7	--	--	33	
29	61	4.0	--	7.2×10^7	--	--	40	
30	63	4.5	32	5.8×10^7	--	--	80	
31	67	5.4	--	3.7×10^7	1.4×10^8	2.7×10^9	53	
32	47	6.2	--	5.3×10^7	--	--	144	
33	61	6.3	--	2.9×10^6	--	--	17	
34	52	6.3	--	1.6×10^7	--	--	47	
35[b]	34	2.7	2.0	8.1×10^6	2.8×10^7	4.9×10^8	26[c]	
36[b]	44	6.7	--	1.2×10^5	3.2×10^5	3.6×10^7	42[d]	
37	46	5.0	--	900	--	--	24	
Average of typical runs 2-31	61	3.7	25.2	5.9×10^7	1.5×10^8	2.4×10^9	26	
				8-Rod Lot				
1	57	0.7	52.8	8.8×10^6	7.9×10^8	4.0×10^9	25	6-rod charge
2	53	2.8	31.8	2.9×10^7	2.2×10^9	1.1×10^{10}	29	Residual metal heel + 1 rod
3	54	3.5	18.7	2.5×10^7	1.6×10^9	8.7×10^9	34	Residual metal heel + 1 rod
4	52	1.9	35.1	2.2×10^7	1.5×10^9	8.2×10^9	30	
5	68	5.5	26.1	2.8×10^7	1.9×10^9	1.1×10^{10}	32	
6	65	4.7	20.6	2.1×10^7	1.3×10^9	7.6×10^{10}	34	
7	60	3.3	45.7	4.0×10^7	2.4×10^9	1.4×10^{10}	42	Residual metal heel
8	65[e]	5.3	23.9	8.2×10^7	1.8×10^9	9.3×10^9	56	
9	65[e]	4.8	16.0	3.7×10^7	9.2×10^8	5.6×10^9	21	
10	47[e]	8.6	15.2	9.0×10^7	3.8×10^8	1.9×10^9	>150	
Average of runs 1-8	59	3.5	31.8	3.2×10^7	1.7×10^9	1.8×10^{10}	35	

[a] Single-rod charged to batch 1 - fresh lot of dissolvent was added to continue with batch 1A after first had been removed.

[b] Residual metal heel dissolved with 3.3 M $NaNO_3$ - 1.8 M NaOH which was later acidified to ≈ 2 M HNO_3 with 9 M HNO_3 and digested at 100-105°C.

[c] Digested alkaline solution 10 hr and the acidified mixture 16 hr.

[d] Digested alkaline solution 8 hr and the acidified mixture 34 hr.

[e] Contains rework material returned to dissolver for cleanout dissolvings.

Table 5.2. Composition of First Cycle Ion Exchange Feed Solution For Plutonium Recovery

Batch No.	HNO_3 (M)	Al (M)	Counting Rate (counts min^{-1} ml^{-1})			Nuclide (counts min^{-1} ml^{-1})			
			Pu α	Gross α	Gross γ	Ru γ	Ce-β	Zr-Nb γ	TRE β
					24-Rod Lot				
1	3.1[a]	0.65	3.0×10^7	1.0×10^8	1.7×10^9				
1A	4.7[a]	--	2.1×10^7	7.6×10^7	1.3×10^9				
2	2.0[a]	1.1	1.5×10^7	6.4×10^7	9.3×10^8				
2A	3.1[a]	--	4.6×10^7	1.5×10^8	2.3×10^9				
3	1.9[a]	--	5.3×10^7	1.6×10^8	2.7×10^9				
4	3.0[a]	--	5.8×10^7	1.8×10^8	2.9×10^9				
5	3.5[a]	--	3.4×10^7	--	2.6×10^9				
6	1.9[a]	--	4.2×10^7	--	--				
7	2.6[a]	--	4.3×10^7	2.0×10^8	3.4×10^9				
8	4.0[a]	--	5.7×10^7	2.5×10^8	4.6×10^9	2.8×10^9 [b]	2.1×10^{10} [b]	5.3×10^8 [b]	2.7×10^9
9	1.4[a]	--	4.9×10^7	2.1×10^8	3.8×10^9				
10	7.6	--	1.8×10^7	8.0×10^7	1.4×10^9				
11	7.0	--	4.2×10^7	--	--				
12	6.3	--	4.1×10^7	1.5×10^8	2.5×10^9				
13	7.0	--	3.8×10^7	1.4×10^8	2.5×10^9				
14	7.8	--	3.7×10^7	1.8×10^8	3.1×10^9				
15	7.0	--	4.1×10^7	1.3×10^8	2.5×10^9				
16	7.0	--	4.0×10^7	1.6×10^8	2.8×10^9	1.2×10^9	1.1×10^9	1.5×10^9	1.6×10^9
17	7.2	--	6.1×10^7	2.2×10^8	3.6×10^9				
18	6.8	--	3.9×10^7	1.3×10^8	2.2×10^9				
19	6.8	--	3.8×10^7	1.4×10^8	2.6×10^9				
20	6.7	--	5.4×10^7	2.2×10^8	3.9×10^9				
21	7.1	--	4.2×10^7	1.4×10^8	2.3×10^9				
22	6.7	--	3.0×10^7	1.0×10^8	1.7×10^9				
23	6.9	--	3.4×10^7	1.4×10^8	2.5×10^9				
24	6.9	--	4.5×10^7	1.7×10^8	2.8×10^9				
25	6.9	--	3.5×10^7	1.4×10^8	2.4×10^9				
26	7.4	--	4.6×10^7	2.0×10^8	3.4×10^9	1.5×10^9	1.4×10^9	2.2×10^9	1.8×10^9
27	7.0	--	4.1×10^7	1.5×10^8	2.6×10^9				
28	6.7	--	3.4×10^7	1.3×10^8	2.3×10^9				
29	7.1	--	3.2×10^7	2.0×10^8	3.9×10^9				
30	7.0	--	3.2×10^7	2.1×10^8	4.5×10^9				
31	7.0	--	3.0×10^7	1.1×10^8	2.0×10^9				
32	6.8	--	3.0×10^7	1.6×10^8	3.0×10^9				
33	6.7	--	6.6×10^6	2.5×10^7	4.5×10^8				
34	7.4	--	1.2×10^7	7.3×10^7	1.7×10^9				
35	7.2	--	3.9×10^6	1.4×10^7	2.4×10^8				
Average	7.3	--	3.6×10^7	1.5×10^8	2.6×10^9	1.4×10^9	1.3×10^9	1.9×10^9	1.7×10^9
					8-Rod Lot				
1	7.0	0.91	3.7×10^6	4.9×10^8	1.7×10^9				
2	7.2	0.65	1.5×10^7	1.1×10^9	5.7×10^9				
3	7.5	0.35	1.4×10^7	8.4×10^8	4.9×10^9				
4	7.2	0.63	1.2×10^7	8.2×10^8	4.9×10^9	2.1×10^{10} [b]	1.0×10^{10} [b]	4.4×10^8 [b]	--
5	6.6	0.66	1.8×10^7	1.3×10^9	6.8×10^9				
6	7.6	0.44	1.2×10^7	1.4×10^9	8.8×10^9				
7	7.7	0.75	1.9×10^7	1.2×10^9	6.3×10^9				
8	8.0	0.75	4.5×10^7	9.9×10^8	5.3×10^9				
9	8.2	0.38	2.6×10^7	6.9×10^8	3.8×10^9				
10	7.9	0.54	6.4×10^7	3.0×10^8	1.5×10^9				
10A	7.2	0.12	9.1×10^6	2.6×10^8	1.4×10^9				
Average	7.5	0.56	2.2×10^7	8.5×10^8	4.6×10^9	2.1×10^{10} [b]	1.0×10^{10} [b]	4.4×10^8 [b]	--

[a] In these runs, the feed stream to the ion exchange beds was continuously mixed with a stream of 13 M HNO_3 calculated to adjust the HNO_3 concentration to 7 M. In all other runs, the adjustment of HNO_3 was conducted on a batch basis.

[b] These values are reported in dis min^{-1} ml^{-1} as Ru^{106}, Ce^{144}, Zr^{95}-Nb^{95}.

[c] Runs 10 through 35.

Table 5.3. Composition of First Cycle Ion Exchange Loading Waste Solution During Plutonium Recovery

Batch No.	HNO_3 (M)	Counting Rate (counts min^{-1} ml^{-1}) Pu α	Gross α	Gross γ	Remarks
			24-Rod Lot		
1	6.7	4.1×10^3	3.2×10^7	6.6×10^8	
1A	7.8	2.9×10^3	2.8×10^7	6.1×10^8	
2	6.7	2.6×10^3	2.1×10^7	3.9×10^8	
2A	8.3	1.5×10^5	4.9×10^7	1.0×10^9	
3	7.0	9.3×10^5	5.5×10^7	1.2×10^9	
4	7.6	8.1×10^5	6.4×10^7	1.5×10^9	
5	7.4	2.2×10^5	6.6×10^7	1.4×10^9	
6	5.8	3.4×10^5	6.1×10^6	--	
7	5.3	3.3×10^5	9.0×10^7	2.1×10^9	
8	7.9	1.5×10^6	1.1×10^8	2.7×10^9	Reworked as 10A, 10B
9	--	1.5×10^5	7.5×10^7	1.4×10^9	
10	7.0	2.2×10^5	6.5×10^7	1.7×10^9	
10A	5.8	9.0×10^4	7.4×10^5	1.5×10^9	
10B	6.8	1.2×10^5	7.5×10^7	1.8×10^9	
11	6.8	5.2×10^4	--	--	
12	6.5	8.5×10^4	--	--	
13	6.2	1.5×10^5	--	--	
14	7.2	3.4×10^5	1.3×10^8	2.7×10^9	
15	7.0	2.1×10^5	9.6×10^7	2.2×10^9	
16	6.8	3.2×10^5	1.1×10^8	2.6×10^9	
17	7.3	3.1×10^5	--	--	
18	6.9	2.0×10^5	8.3×10^7	1.9×10^9	
19	6.9	2.1×10^5	8.4×10^7	2.1×10^9	
20	5.9	7.7×10^5	1.3×10^8	3.1×10^9	
21	6.9	2.0×10^5	9.1×10^7	2.1×10^9	
22	6.4	1.7×10^5	6.2×10^7	2.9×10^9	
23	6.8	2.3×10^5	9.6×10^7	2.1×10^9	
24	6.1	1.6×10^5	1.0×10^8	2.3×10^9	
25	6.5	3.1×10^5	9.0×10^7	2.1×10^9	Last lot to storage for transuranium element recovery
26	7.0	1.1×10^6	--	--	
27	6.9	1.1×10^6	--	--	
28	6.9	4.7×10^5	9.2×10^7	2.3×10^9	
29	6.6	5.8×10^4	--	--	New first stage resin bed
30	6.4	2.5×10^4	1.4×10^8	3.6×10^9	
31	6.2	2.6×10^5	--	--	
32	6.7	5.5×10^5	7.9×10^7	1.7×10^9	
33	6.7	4.8×10^5	--	--	
34	6.8	2.2×10^5	5.5×10^7	1.2×10^9	
35	--	3.3×10^5	--	--	
Average	6.8	3.4×10^5	7.5×10^7	1.9×10^9	
			8-Rod Lot		
1	6.5	1.3×10^4	3.2×10^8	1.5×10^9	
2	6.9	2.0×10^5	8.6×10^8	4.3×10^9	
3	7.0	1.3×10^5	6.8×10^8	3.9×10^9	
4	7.1	9.3×10^4	6.3×10^8	3.6×10^9	
5	6.1	1.6×10^5	9.7×10^8	5.7×10^9	
6	7.4	2.0×10^5	6.9×10^8	5.1×10^9	
7	7.4	2.4×10^5	1.0×10^9	5.4×10^9	
8	7.4	4.6×10^5	8.5×10^8	4.7×10^9	
9	7.2	6.2×10^5	7.0×10^8	4.0×10^9	
10	7.2	2.3×10^6	2.0×10^8	1.2×10^9	
10A	6.7	5.4×10^5	5.3×10^7	6.6×10^8	
Average	7.0	4.5×10^5	6.3×10^8	3.6×10^9	

Table 5.4. Composition of First Cycle Ion Exchange Scrubbing Waste Solution During Plutonium Recovery

Batch No.	HNO_3 (M)	Counting Rate (counts min^{-1} ml^{-1})			Remarks
		Pu α	Gross α	Gross γ	
			24-Rod Lot		
1	6.8	2.3×10^4	6.0×10^5	5.3×10^7	
1A	7.3	2.2×10^4	7.1×10^4	4.4×10^6	
2	--	1.0×10^4	--	--	
2A	7.5	2.8×10^5	2.1×10^7	6.4×10^8	
3	6.5	5.2×10^5	1.7×10^6	2.8×10^8	
4, 5, 6	7.4	1.6×10^5	--	--	No scrub between 4 and 5
7, 8	7.9	1.5×10^6	--	--	No scrub between 7 and 8
9, 10	7.0	4.5×10^5	--	--	No scrub between 9 and 10
10A, 10B	6.8	1.2×10^5	--	--	
11	7.2	2.1×10^5	2.0×10^7	1.3×10^9	
12	6.8	1.4×10^6	4.9×10^7	1.5×10^9	
13	7.5	1.4×10^5	--	--	
14	7.8	1.2×10^6	5.9×10^7	1.7×10^9	
15, 16	6.8	3.1×10^5	1.5×10^6	4.9×10^8	No scrub between 15 and 16
17	7.2	7.8×10^5	5.6×10^7	1.8×10^9	
18	6.9	5.7×10^5	3.4×10^7	1.1×10^9	
19	7.0	9.3×10^5	1.3×10^7	3.8×10^8	
20	6.9	9.3×10^5	6.5×10^7	2.1×10^9	
21	6.7	5.7×10^5	3.5×10^7	1.1×10^9	
22	6.9	1.8×10^6	8.9×10^6	2.4×10^8	
23	7.6	3.9×10^5	4.3×10^7	1.3×10^9	
24	6.5	3.0×10^5	3.9×10^7	1.2×10^9	
25	7.0	1.1×10^6	1.2×10^7	3.4×10^8	
26, 27, 28	7.1	5.8×10^5	1.5×10^7	4.4×10^8	No scrub between 26,27, and 28
29	6.4	1.6×10^5	7.4×10^7	2.5×10^9	
30	6.8	1.5×10^6	7.1×10^7	2.5×10^9	
31	6.9	1.4×10^5	7.8×10^6	4.2×10^8	
32, 33, 35	7.5	1.5×10^5	--	--	No scrub between 32,33, and 34
Average	7.1	5.7×10^5	3.0×10^7	1.0×10^9	
			8-Rod Lot		
1	6.5	9.2×10^3	7.1×10^7	4.0×10^8	
2	7.2	2.1×10^5	3.5×10^8	2.5×10^9	
3	8.2	1.7×10^5	3.0×10^8	2.0×10^9	
4	7.9	1.6×10^5	2.1×10^8	1.4×10^9	
5	7.0	4.3×10^5	3.4×10^8	2.5×10^9	
6	7.5	2.0×10^5	4.9×10^7	3.2×10^8	
7	7.8	2.5×10^5	4.1×10^8	2.6×10^9	
8, 9	7.5	5.5×10^6	5.9×10^7	3.5×10^8	
10	6.3	3.1×10^6	1.0×10^8	6.3×10^8	
10A	7.0	5.2×10^5	8.7×10^6	1.4×10^8	
Average	7.3	1.1×10^6	1.9×10^8	1.3×10^9	

Table 5.5. Composition of Second Cycle Ion Exchange Feed Solution During Plutonium Recovery

Batch No.	HNO_3 (M)	Counting Rate (counts min^{-1} ml^{-1})		
		Pu α	Gross α	Gross γ
		24-Rod Lot		
1 through 6	8.4	1.5×10^8	1.5×10^8	1.2×10^6
7 through 19	6.9	4.2×10^8	4.2×10^8	5.3×10^6
20 through 28	7.2	2.8×10^8	2.9×10^8	3.5×10^6
29 through 35	7.3	1.8×10^8	1.8×10^8	2.3×10^6
Average	7.5	2.6×10^8	2.6×10^8	3.1×10^6
		8-Rod Lot		
1 through 6	7.4	1.0×10^7	1.1×10^7	2.3×10^6
7 through 9	7.6	2.0×10^7	2.1×10^7	1.5×10^6
10	7.4	3.8×10^7	3.7×10^7	2.5×10^6
10A	7.4	1.3×10^7	1.5×10^7	1.0×10^7
Average	7.5	2.0×10^7	2.1×10^7	4.0×10^6

Table 5.6. Composition of Second Cycle Ion Exchange Loading Waste Solution During Plutonium Recovery

Batch No.	HNO_3 (M)	Counting Rate (counts min^{-1} ml^{-1})		
		Pu α	Gross α	Gross γ
		24-Rod Lot		
1 through 6	8.4	5.2×10^4	7.8×10^5	3.9×10^6
7 through 19	7.3	4.1×10^5	2.3×10^6	2.7×10^6
20 through 28	7.1	3.1×10^5	--	--
29 through 35	6.5	3.1×10^5	1.5×10^6	1.6×10^6
Average	7.3	2.7×10^5	1.5×10^6	2.7×10^6
		8-Rod Lot		
1 through 6	7.7	2.3×10^5	1.6×10^6	4.7×10^6
7 through 9	7.7	1.1×10^6	1.1×10^6	2.2×10^6
10	7.5	9.4×10^5	1.2×10^6	3.4×10^6
10A	7.4	6.2×10^4	4.9×10^6	1.1×10^7
Average	7.6	5.8×10^5	2.2×10^6	5.3×10^6

Table 5.7. Composition of Second Cycle Ion Exchange Scrubbing Waste Solution During Plutonium Recovery

Batch No.	HNO_3 (M)	Counting Rate (counts min^{-1} ml^{-1})		
		Pu α	Gross α	Gross γ
		24-Rod Lot		
1 through 6	7.3	2.5×10^5	3.7×10^5	1.4×10^6
7 through 19	7.1	1.6×10^5	1.7×10^5	1.1×10^6
20 through 28	7.6	1.5×10^5	--	--
29 through 35	7.2	3.7×10^5	--	--
Average	7.3	2.3×10^5	2.7×10^5	1.3×10^6
		8-Rod Lot		
1 through 6	7.8	3.7×10^5	1.3×10^6	3.3×10^6
7 through 9	7.6	9.3×10^5	1.1×10^6	7.0×10^5
10-10A	7.4	5.3×10^5	5.7×10^5	1.7×10^5
Average	7.6	6.1×10^5	9.9×10^5	1.4×10^6

Table 5.8. Composition of Plutonium Product

Bottle No.	Plutonium Total wt. (g)	Plutonium Conc. (g/liter)	Counting Data (counts min^{-1} ml^{-1}) Gross α	Counting Data (counts min^{-1} ml^{-1}) Gross γ	HNO_3 (M)	Sp G (g/liter)	Isotopic Assay (wt %) Mass 238	Mass 239	Mass 240	Mass 241	Mass 242
						24-Rod Lot					
1, 2	106	4.32	7.5×10^8	2.4×10^5	2.2	1.068	--	34.50	48.38	12.07	5.05
3, 4	283	14.10	2.6×10^9	1.7×10^5	2.2	1.087	--	30.45	50.62	12.73	6.21
5, 6	182	9.11	1.7×10^9	1.1×10^5	1.8	1.066	--	25.87	53.25	13.34	7.54
7, 8	84	4.31	--	--	--	1.078	--	23.84	54.23	13.72	8.22
Average[a]	655	7.8	1.4×10^9	1.5×10^5	2.0	1.074	--	29.03	51.54	12.94	6.65
						8-Rod Lot					
1,2,3	8.5	0.30	7.6×10^7	1.6×10^5	2.3	1.071	1.09	0.35	14.55	7.67	76.34
4,5,6	1.9	0.06	1.6×10^7	4.8×10^4	--	1.059	1.07	0.37	14.77	7.59	76.20
7,8,9,10	12.8	0.37	1.0×10^8	210	1.2	1.034	1.11	0.39	15.65	7.93	74.90
11,12,13,14	5.1	0.15	4.3×10^7	5.6×10^4	4.8	1.154	1.06	0.27	15.56	7.78	75.34
15,16,17,19	2.1	0.06	1.6×10^7	2.5×10^5	3.2	1.105	1.06	0.27	15.56	7.78	75.34
Average[b]	30.4	0.19	5.0×10^7	1.1×10^5	2.8	1.086	1.09	0.35	15.27	7.80	75.49

[a] Sp. activity calculated from weighted average, these data = 1.843×10^8 α counts min^{-1} mg^{-1} Pu.

[b] Sp. activity calculated from weighted average, these data = 2.56×10^8 α counts min^{-1} mg^{-1} Pu.

Table 5.9. Composition of Composited Raw Plutonium Recovery Wastes

	24-Rod Lot	8-Rod Lot
Volume (liters)	3030	970
Al (M)	0.35	0.50
HNO_3 (M)	5.2	6.2
Sp. G. (g ml^{-1})	1.26	1.20
Gross α (counts min^{-1} ml^{-1})	4.4×10^7	6.2×10^8
α-pulse (% Gross α)		
5.5 Mev	19	< 2
5.8 Mev	24	56
6.1 Mev	57	42
Pu (g total)	3.1	1.7
Pu conc. (mg $liter^{-1}$)	1	1.7

Table 5.10. Typical Composition of Evaporator Condensate Evolved During Transuranium Element Feed Preparation

Batch Number	Fraction	Gross α (counts min^{-1} ml^{-1})	Gross γ (counts min^{-1} ml^{-1})	HNO_3 (M)	Remarks
			24-Rod Lot		
1	1	1.8×10^4	5.7×10^5	2.7	Evaporation
	3	6800	5.7×10^5	6.9	Evaporation
	5	6800	1.1×10^5	6.3	Evaporation
	9	1.2×10^4	8.3×10^6	6.8	Digestion
	10	1.4×10^4	1.3×10^7	6.6	Digestion
3	1	7800	1.6×10^6	4.0	Evaporation
	3	3000	3.9×10^5	8.8	Evaporation
	6	6200	5.5×10^5	6.6	Evaporation
	10	6.1×10^4	4.2×10^6	6.1	Digestion
	11	2.7×10^4	1.2×10^7	7.1	Digestion
			8-Rod Lot		
9	2	1.7×10^5	1.9×10^6	6.4	Evaporation
	4	1.0×10^5	1.3×10^6	7.4	Evaporation
	9	3.1×10^5	8.9×10^6	6.9	Evaporation
	11	1.6×10^7	8.0×10^7	<0.1	Evaporation
	12	1.4×10^7	7.2×10^7	<0.1	Evaporation
10	1	3.1×10^6	2.4×10^7	1.7	Evaporation
	3	4.5×10^5	9.2×10^6	--	Evaporation
	6	3.2×10^5	4.1×10^6	6.7	Evaporation
	11	7.8×10^6	4.0×10^7	0	Digestion
	14	9.8×10^6	5.5×10^7	0.1	Digestion

Table 5.11. Composition of Ion Exchange System Feed Solution During Transuranium Element Recovery

Batch No.	Al (M)	HNO_3 (N AD[a])	Gross (counts min^{-1} ml^{-1})			Nuclide (dis min^{-1} ml^{-1})				
			(Alpha)	(Beta)	(Gamma)	Ce^{144}	Cs^{137}	Ru^{106}	Sr^{90}	Zr^{95}
					24-Rod Lot					
1	2.4	0.08[b]	3.0×10^{8}	--	7.3×10^{9}	--	--	--	--	--
2	2.3	0.32	2.8×10^{8}	1.0×10^{10}	7.6×10^{9}	4.3×10^{10}	1.5×10^{10}	4.9×10^{10}	5.8×10^{9}	8.5×10^{7}
3	2.0	0.31	2.3×10^{8}	8.0×10^{9}	6.4×10^{9}	2.9×10^{10}	9.0×10^{9}	3.7×10^{10}	3.7×10^{9}	6.4×10^{7}
4	2.9	1.24	3.2×10^{8}	7.1×10^{9}	5.6×10^{9}	2.9×10^{10}	1.1×10^{10}	2.8×10^{10}	3.6×10^{9}	5.5×10^{7}
5	2.4	0.60	2.6×10^{8}	8.5×10^{9}	5.6×10^{9}	2.3×10^{10}	--	--	--	--
6	2.8	1.04	2.9×10^{8}	1.0×10^{10}	8.4×10^{9}	3.2×10^{10}	--	--	--	--
7				Returned to Run 8						
8	2.6	0.48	2.5×10^{8}	1.0×10^{10}	7.7×10^{9}	2.7×10^{10}	--	--	--	--
Average	2.5	0.60	2.8×10^{8}	8.9×10^{9}	6.9×10^{9}	3.1×10^{10}	1.2×10^{10}	3.8×10^{10}	4.4×10^{9}	6.8×10^{7}
					8-Rod Lot					
9	2.0	1.14	3.5×10^{9}	1.5×10^{10}	1.9×10^{10}	4.6×10^{10}	--	--	--	--
10A	3.0	1.77	3.9×10^{9}	1.5×10^{10}	2.1×10^{10}	5.2×10^{10}	--	--	--	--
10B	2.5	1.64	3.4×10^{9}	1.4×10^{10}	1.9×10^{10}	4.4×10^{10}	--	--	--	--
11	2.6	0.38	2.9×10^{9}	9.4×10^{9}	1.4×10^{10}	3.0×10^{10}	--	--	--	--
Average	2.5	1.23	3.4×10^{9}	1.3×10^{10}	1.8×10^{10}	4.3×10^{10}	--	--	--	--

[a] AD = Acid deficiant.

[b] Acid.

Table 5.12. Composition of Ion Exchange Loading Waste Solution During Transuranium Element Recovery

Batch No.	Al (M)	Gross (counts min^{-1} ml^{-1})			Nuclide (dis min^{-1} ml^{-1})				
		(Alpha)	(Beta)	(Gamma)	Ce^{144}	Cs^{137}	Ru^{106}	Sr^{90}	Zr^{95}
					24-Rod Lot				
1	2.0	4.9×10^{7}	1.6×10^{9}	8.8×10^{9}	3.3×10^{8}	8.7×10^{9}	9.9×10^{8}	3.7×10^{9}	4.7×10^{7}
2	2.1	1.7×10^{8}	3.5×10^{9}	4.2×10^{9}	1.4×10^{10}	8.8×10^{9}	2.7×10^{9}	3.5×10^{9}	6.3×10^{7}
3	2.3	1.9×10^{8}	3.4×10^{9}	--	--	--	--	--	--
4	--	4.9×10^{5}	1.8×10^{9}	4.8×10^{9}	1.8×10^{6}	--	--	--	--
5	1.7	7.9×10^{7}	1.9×10^{9}	4.2×10^{9}	5.2×10^{7}	--	--	--	--
6	--	8.3×10^{5}	1.8×10^{9}	4.4×10^{9}	2.7×10^{7}	--	--	--	--
7				Returned to Run 8					
8	--	6.4×10^{5}	1.8×10^{9}	3.8×10^{9}	7.0×10^{7}	--	--	--	--
Average	2.0	7.0×10^{7}	2.3×10^{9}	4.2×10^{9}	2.4×10^{9}	8.8×10^{9}	1.8×10^{9}	3.6×10^{9}	5.5×10^{7}
					8-Rod Lot				
9	--	2.4×10^{8}	1.0×10^{10}	2.5×10^{9}	$\sim 5.0 \times 10^{8}$	--	--	--	--
10A	--	7.4×10^{7}	--	--	--	--	--	--	--
10B	--	7.7×10^{7}	2.3×10^{9}	6.8×10^{9}	--	--	--	--	--
11	--	5.7×10^{6}	1.8×10^{9}	7.5×10^{9}	6.8×10^{7}	--	--	--	--
Average	--	9.9×10^{7}	4.7×10^{9}	5.6×10^{9}	2.8×10^{8}	--	--	--	--

Table 5.13. Composition of Ion Exchange Scrubbing Waste Solution During Transuranium Element Recovery

Batch No.	Gross (counts min^{-1} ml^{-1})		
	Alpha	Beta	Gamma
		24-Rod Lot	
1	2.6×10^8	--	1.3×10^9
2	7.1×10^8	--	1.0×10^9
3	6.1×10^7	1.7×10^9	--
4	2.8×10^5	5.8×10^8	1.1×10^9
5	2.5×10^8	2.3×10^9	1.1×10^9
6	1.6×10^6	5.2×10^8	8.9×10^8
7		Returned to Run 8	
8	1.3×10^6	7.7×10^8	4.5×10^9
Average	1.8×10^8	1.2×10^9	1.6×10^9
		8-Rod Lot	
9	2.1×10^8	8.5×10^8	2.4×10^8
10A	--	--	--
10B	1.8×10^8	5.7×10^9	1.6×10^9
11	4.8×10^7	1.2×10^9	3.9×10^9
Average	1.5×10^8	2.6×10^9	1.9×10^9

Table 5.14. Composition of Transuranium Element Product

	Al (M)	Li (M)	HNO_3 (M)	Gross (counts min^{-1} ml^{-1})			Nuclide (dis min^{-1} ml^{-1})				
				(Alpha)	(Beta)	(Gamma)	Ce^{144}	Cs^{137}	Ru^{106}	Sr^{90}	Zr^{95}
						24-Rod Lot					
Dilute	0.035	2.0	0.13	3.3×10^8	6.3×10^9	7.6×10^8	4.4×10^{10}	--	--	--	--
Concentrate	0.060	6.4	1.10	1.1×10^9	2.2×10^{10}	2.7×10^9	1.9×10^{11}	4.8×10^7	1.8×10^{10}	5.8×10^7	4.1×10^7
						8-Rod Lot					
Dilute	0.06	1.1	--	1.9×10^9	3.5×10^9	6.7×10^8	2.7×10^{10}	3.5×10^8	5.6×10^7	--	$< 2 \times 10^7$
Concentrate	0.10	2.2	--	4.1×10^9	8.8×10^9	2.1×10^9	5.5×10^{10}	3.4×10^8	1.9×10^{10}	--	1.3×10^8

Table 5.15. Alpha Energy Analysis of Transuranium Element Product

	Gross Alpha (counts min^{-1} ml^{-1})	Alpha Pulse (% gross alpha) 5.5 (Mev)	5.8 (Mev)	6.1 (Mev)
		24-Rod Lot		
Dilute	3.3×10^8	28	18	54
Concen-trated	1.1×10^9	24	26	50
		8-Rod Lot		
Dilute	1.9×10^9	--	67	33
Concen-trated	4.1×10^9	--	65	35

Table 5.16. Typical Composition of Condensate Evolved During Transuranium Element Product Evaporation

Fraction	Gross α (counts min^{-1} ml^{-1})	Gross γ (counts min^{-1} ml^{-1})	HNO_3 (M)	Remarks
		24-Rod Lot		
1	6.1×10^6	1.5×10^7	--	
3	6.4×10^4	2.4×10^5	--	
4	4.9×10^4	2.7×10^5	--	
5	4.8×10^5	1.2×10^6	--	
6	1.7×10^5	4.8×10^5	--	
		8-Rod Lot		
12	5.7×10^7	4.2×10^7	< 0.1	
13	2.3×10^6	3.4×10^6	0.1	
14	--	2.8×10^6	--	

Table 5.17. Material Balance: Transuranium Element Recovery Based on Alpha Counting and Aluminum Analyses

Stream	Alpha Total (counts min^{-1})		Alpha Total (% of Raw Feed)	Aluminum (Moles)	Aluminum (% of Raw Feed)
		24-Rod Lot			
Raw Feed: IN		1.3×10^{14}	100	1061	100
Prepared Feed: OUT					
Discharged to waste	3.2×10^{13}		25	220	21
Processed to ion exchange	9.7×10^{13}		75	863	81
Total accounted for: (feed preparation)	1.3×10^{14}	1.3×10^{14}	100	1083	102
Ion exchange losses					
Loading	2.6×10^{13}		20		
Scrubbing	1.9×10^{13}		15		
Total losses	4.5×10^{13}	4.5×10^{13}	35		
Product recovery					
Recovered	3.8×10^{13}		29		
Recoverable	3.5×10^{12}		3		
Total product	4.2×10^{13}	4.2×10^{13}	32		
Total accounted for: (ion exchange)		8.7×10^{13}	67		
		8-Rod Lot			
Raw Feed: IN		6.0×10^{14}	100	485	100
Prepared Feed: OUT					
Discharged to waste	1.3×10^{14}		22	270	56
Processed to ion exchange	3.6×10^{14}		60	220	45
Total accounted for: (feed preparation)	4.9×10^{14}	4.9×10^{14}	82	490	101
Ion exchange losses					
Loading	4.2×10^{13}		7		
Scrubbing	1.1×10^{13}		2		
Leakage to sump[a]	3.3×10^{13}		5		
Total losses	8.6×10^{13}	8.6×10^{13}	14		
Product recovery					
Recovered	1.9×10^{14}		32		
Recoverable	5.7×10^{13}		10		
Total product	2.5×10^{14}	2.5×10^{14}	42		
Total accounted for: (ion exchange)	3.4×10^{14}	3.4×10^{14}	56		

[a] Including material left on incompletely eluted resin.

5.2 Examination of Spent Resin

After three months service in the first stage column during the processing of the 24-rod lot of fuel rods, the charge of Permutit SK resin had received an estimated dose of 4×10^8 rads. Upon removal and examination of the resin it was observed to be soft and jet black but remained essentially as discrete beads; however, there was some evidence of fines that indicated a small fraction of the resin beads had shattered.

During the recovery of plutonium from the eight-rod lot of fuel, the resin was removed from the column after a month of service and examined. The estimated radiation dose received during this interval was 4.6×10^7 rads. This resin also was jet black and sticky (it compacted and aggregated under pressure). Prior to removal, the resin had been exhaustively eluted with 0.7 M to 3 M HNO_3 containing from 0.01 to 0.1 M F^- and with 0.7 M to 3 M HNO_3 containing 0.1 M Fe $(NH_2SO_3)_2$ or $NH_2OH.H_2SO_4$.

After removal from the column, samples from the top and bottom of the resin bed were examined in the laboratory. The resin from each location was exhaustively eluted with 0.7 to 7 M HNO_3 containing 0.01 to 0.1 M F^-, with reducing agents, and finally, with aqua regia. In all cases, plutonium at about 0.1 mg/ml (in the eluant), and aluminum at about 0.1 mg/ml, was eluted along with fission products, indicating these to be tightly bonded to the resin. Samples of resin from each location were then ashed and the residue dissolved as completely as possible in aqua regia - a small portion of the ash did not dissolve, even on repeated fuming to dryness. The solution of the ash and the residual solids were examined for plutonium, aluminum, and other ionic material using radiochemical and spectrographic procedures. Results were as follows:

	Component Found (g/liter of resin)		
	Top of Resin Bed[a]	Bottom of Resin Bed	Residual Solids from Ash Dissolution
Pu	3.2	0.07	---
Al	0.5	--	Moderate
Pd	0.14	--	Strong
Rh	0.03	--	Weak
Fe, Cr	Trace	--	Weak
Zr	< 0.2	--	---
Nb	< 2.0	--	---
Rare Earths	<1.0	--	---
Cs	--	--	Faint trace

[a] Estimated to comprise about 20 to 30% of the bed.

6. CONCLUSIONS AND RECOMMENDATIONS

From the results and experience obtained in this program, it is concluded that:

1. Adequate dissolution rates of highly radiated Pu-Al alloy can be obtained with 6 M HNO_3 catalyzed with 0.05 M Hg and 0.03 M F^- to make the method practical for pilot-plant-scale operation.

2. Highly purified plutonium, decontaminated from fission products, can be recovered with low losses by anion exchange methods. However, a preliminary feed preparation step in which the bulk of the aluminum was removed (for example, by dissolution in NaOH followed by centrifugation

of the plutonium hydrates and dissolution of these in the minimum amounts of HNO_3) and the plutonium concentration increased would result in feed solutions better suited for processing the highest burnup material because of the lower unit losses obtained when processing solutions of higher plutonium concentration.

3. Transuranium elements (and rare earths) can be separated and recovered from the plutonium-loading waste effluent in good yield and nearly free of aluminum. The low loading capacity of the anion resin, however, imposes some limitation on the general applicability of the method.

Difficulties encountered during the program lead to the recommendation that:

1. Improved methods for solids removal and subsequent handling should be developed that will permit good separation with rapid throughput. In addition, an improved method of discharging the solids to waste in the minimum volume of waste solution without plugging process lines would increase recoveries and improve the mechanical operation of the system. Solids-free feed solutions are required for efficient operation of the ion exchange system.

2. The ion exchange columns should be equipped with an efficient resin removal system that permits complete removal of spent resin to a unit where total dissolution of the resin can be conducted, both for its ultimate disposal and for the recovery of irreversably sorbed valuable material. This requirement becomes more important with extremely high burnup material where resin degradation is severe.

3. An improved evaporator should be designed that permits the concentration of solution to about 20% of the working volume of the vessel without exposing the heat transfer surfaces. Such a design would result in the formation of fewer solids in the concentrate, easier, more rapid clarification of concentrated solutions, lower losses of valuable material, less material for rework, increased system capacity, and decreased volumes of waste solution.

4. A resin hold-down device should be included in the resin column design. Because of the high densities of the feed and scrub solutions necessary in the transuranium element recovery process, the ion exchange resin tends to float. In the present program, it was necessary to operate at minimum flowrates such that the downflow rate in the standpipe exceeded the rate of rise of the resin in the solution. Flexibility of operation was sacrificed as a consequence, since at below these minimum rates resin rose in the column and standpipe, resulting in an effectively longer column of smaller diameter with increased pressure drop. The need in this case was also increased when solutions that "gas" (typical at these radiation levels) are handled. Gas binding of the resin bed was a serious operational problem and often resulted in partially turning the bed over, with increased loss of valuable material. For this reason also, sensing devices for measuring the pressure drop across the resin bed should not be of the pneumatic type that discharge gas into the column.

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