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RECOVERY AND PURIFICATION OF
OF PLUTONIUM AND NEPTUNIUM
FROM IRRADIATED NpO_2 - Al AND NpO_2 -
GRAPHITE TARGETS

SEPTEMBER 4, 1970

MASTER

PREPARED FOR

THE UNITED STATES ATOMIC ENERGY COMMISSION

UNDER CONTRACT AT(45-1)-1830

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FROM IRRADIATED NpO_2 - Al AND NpO_2 - GRAPHITE TARGETS

BY

E. J. Wheelwright

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RECOVERY AND PURIFICATION OF PLUTONIUM AND NEPTUNIUM
FROM IRRADIATED NpO_2 - Al AND NpO_2 - GRAPHITE TARGETS

E. J. Wheelwright

INTRODUCTION

A total of 30 neptunium target elements were irradiated during the summer of 1969. Twelve of the elements were fabricated using aluminum as the matrix material, and 18 were fabricated in graphite pellets. All of the aluminum elements and six of the graphite elements were canned in aluminum, and the remaining graphite elements in Zircaloy-2.

This document reports the results achieved during the recovery and purification of both the Pu and Np from these elements. After the work had been started, a second batch of graphite elements were accepted for processing and data from that batch are included.

SUMMARY AND CONCLUSIONS

The new C-cell dissolver worked very well and was reasonably easy to control. One abnormally active element section was found. This piece started to dissolve at ambient temperature immediately after the lid was sealed and before the off-gas vacuum was adjusted. As a result, a small volume of acid was expelled up the sparge leg. This was recovered and returned to the dissolver. The procedure subsequently was changed so that off-gas vacuum was drawn on the dissolver before metal was charged and the lid closed.

The C-cell dissolving cycle required about eight hours for each two bisected sections. However, about a third of the time was spent in solution makeup and transfer. When properly set up on a production basis, the equivalent of four of this type target elements could be processed per 24 hour day. With elements containing a higher ratio of actinides to aluminum, the actinide processing would be increased proportionally.

The addition of oxygen to the graphite burning process gave good results. None of the pellets was presoaked in an oxidizing agent as had been done in the previous graphite processing campaign. The burning pot was not properly sized so the batch sizes were quite small. However, the process was effective. An

improved pot design would substantially increase the processing rate.

The A-cell ion-exchange columns were designed for a Pm purification process employing displacement chromatography. As a result the L/D ratio for the columns is larger than optimum for the nitric acid anion-exchange process used for the separation and purification of Np and Pu described in this report. The small diameter restricted the solution flow rates and increased the time cycles for each step. The large L/D ratio did increase the effectiveness of the fission product decontamination and this is reflected in the very low F.P. contamination of the product. Previous separations in C-cell usually required two cycles to obtain the necessary F.P. decontamination.

Only qualitative observations can be given concerning the radiolysis problems encountered. The thermal and radiolytic effects from 36 grams of ^{238}Pu absorbed on the resin do create problems of resin bed disruption and resin particle damage. The resin bed disruption results from gas formation and leads to channeling of the absorbed band front. The end result is a decrease in effective resin capacity and a decrease in the effectiveness of the decontamination and partition cycles. Resin bed disruption can be minimized by periodic degassing of the bed. This is quite an effective practice for the glass column used for the Pu purification cycle because most of the resin bed is visible. A stainless steel column is used for the co-absorption cycle, and the degassing must be performed without visual observation of the resin bed. Resin particle damage is readily observable during the Pu purification cycle. After the Pu has been removed, that portion of the resin bed occupied by ^{238}Pu is observed to be much darker than the rest of the resin. Repeated use of the same resin would probably lead to loss of absorption capacity and perhaps to physical entrapment of some Pu within the resin particles. This problem was largely avoided by frequently changing the resin.

Two problems were encountered during this work which should be corrected before this facility or a new facility is used on a production basis. The partition cycle must be performed below 30°C to decrease the rate of oxidation of the ferrous sulfamate by nitric acid to an acceptable level. A temperature of 20°C would be better than 30°C . The A-cell columns can be cooled, but the only cooling medium is river water, which is not cold enough in the summer. A properly sized refrigeration unit would solve this problem. The second problem is the time required to get analytical results for process control. The development of an in-line detector for installation on the column effluent line

would greatly aid operational control and prevent premature disposal of waste solution containing significant amounts of Np or Pu.

PROCESSING DETAILS AND ANALYTICAL RESULTS

NpO₂ - A1 Target Elements

The 12 aluminum target elements, number A-1 through A-12, were received in B-cell of 325-A Building and were cut into two equal-length sections with a mechanical powered saw. The center ballast rods were removed and the sectioned elements plus saw cuttings were then transferred into C-cell for dissolution.

The bisected elements were dissolved in 12 dissolver batches in the C-cell 35-liter dissolver. Two bisected sections (equivalent to one full element) were dissolved in five to six hours. Including time required for solution makeup and transfer, dissolution time was usually about eight hours. The processing steps are summarized as follows:

1. A measured 24 liters of 8M HNO₃ plus 33.8 grams of Hg(NO₃)₂·H₂O dissolved in 100 ml of 8M HNO₃ were transferred into the dissolver.
2. One fuel element section was placed in the dissolver basket, lowered into the dissolver, and the dissolver sealed.
3. The vacuum off-gas system was adjusted to continuously sparge the dissolver and draw a negative pressure of one to two inches Hg.
4. The dissolver was heated by electrical resistance elements on the external surface until the solution temperature, monitored by an internal thermocouple, indicated a mildly exothermic reaction. The initiation temperature ranged from 75 to 105°C. The temperature was controlled by reducing the power and circulating cold water through coils in the dissolver.
5. After the exothermic reaction was completed, the dissolver was cooled and opened, the basket was inspected, and a second section of fuel then dissolved in the same solution.

6. Following dissolution of two fuel element sections per batch of dissolver solution, each batch was vacuum transferred to A-cell. The dissolver solution was accumulated in two separate tanks in A-cell because no single tank was large enough to contain the total volume of solution.

The solution in each tank was sampled in duplicate. The analytical results are given in Table I.

The ion-exchange purification process included the following steps:

1. Feed adjustment of both batches to give Pu^{IV} and Np^{IV} at 7.5M HNO_3 .
2. Co-absorption of Pu and Np on a 23 liter bed of Dowex 1, X-3 (30-50 mesh) resin.
3. Co-decontamination with 10.8 bed volumes of 8M HNO_3 containing 0.005M KF at 60°C .
4. Pu partition cycle with 8.3 bed volumes of 5.7M HNO_3 , 0.05M hydrazine , $0.05\text{M ferrous sulfamate}$ at 30°C or less.
5. Np product elution from the resin with 0.35M HNO_3 at 60°C .
6. Pu feed adjustment to Pu^{IV} at 7.5M HNO_3 .
7. Pu absorption on a 4-liter bed of Permutit SK (50-100 mesh) resin.
8. Pu decontamination at 60°C with 10 bed volumes of 8M HNO_3 containing 0.005M KF .
9. Pu product elution with 0.6M HNO_3 at 60°C .

A brief wash with 8M HNO_3 was interposed between steps 3 and 4, 4 and 5, and 8 and 9 to prevent contamination with iron or fluoride. Table II gives material balance results for the first ion exchange run.

A-cell contains three large tanks (Tank 1..... 360 l capacity, Tank 2 360 l capacity, and Tank 3..... 650 l capacity), but Tank 1 is the only one that can be heated or cooled. Both the initial feed adjustment (step No. 1) and the Pu feed adjustment (step No. 6) require extended heating cycles to oxidize Pu^{III} to Pu^{IV} . The run plan specified that the first batch of dissolver solution be collected in Tank 1, the feed adjustment made (step No. 1), and the prepared feed then transferred to Tank 2. The second batch of dissolver solution was then accumulated in Tank 1 and the feed adjustment performed. When the co-absorption step was initiated, the feed in tank 1 was pumped through the resin bed and

Table I
Np-Al Dissolver Solution

	<u>Tank A</u>		<u>Tank B</u>	
	<u>Sample 1</u>	<u>Sample 2</u>	<u>Sample 5</u>	<u>Sample 6</u>
Volume (liters)	147		190	
HNO ₃ concentration (M)	5.6	5.6	5.25	5.26
²³⁸ Pu concentration (g/l)	0.158	0.159	0.111	0.115
total (grams)	23.2	23.4	21.1	21.9
²³⁷ Np concentration (g/l)	1.47	1.51	1.00	1.01
total (grams)	216	222	190	192
¹³⁷ Cs Burnup Analyses (atoms/ml)	1.79x10 ¹⁵	1.78x10 ¹⁵	1.21x10 ¹⁵	1.25x10 ¹⁵
Gamma Energy Analyses				
¹⁴⁴ Ce (dis/min-ml)	8.07x10 ⁸	8.14x10 ⁸	5.84x10 ⁸	5.84x10 ⁸
¹⁴¹ Ce "	4.91x10 ⁷	4.73x10 ⁷	3.43x10 ⁷	3.35x10 ⁷
¹⁰⁶ Ru "	6.59x10 ⁸	6.48x10 ⁸	4.69x10 ⁸	4.90x10 ⁸
¹⁰³ Ru "	1.35x10 ⁸	1.37x10 ⁸	9.75x10 ⁷	9.69x10 ⁷
¹³⁷ Cs "	7.67x10 ⁷	7.63x10 ⁷	5.19x10 ⁷	5.38x10 ⁷
¹³⁴ Cs "	7.89x10 ⁶	7.36x10 ⁶	5.54x10 ⁶	5.96x10 ⁶
⁹⁵ Zr "	3.95x10 ⁸	3.92x10 ⁸	2.86x10 ⁸	2.85x10 ⁸
⁹⁵ Nb "	6.73x10 ⁸	5.89x10 ⁸	4.46x10 ⁸	4.21x10 ⁸
²³³ Pa "	<2.6x10 ⁶	<8.5x10 ⁵	<8.4x10 ⁴	3.34x10 ⁶

Table II
Material Balance - Ion Exchange Run 1

	<u>Solution Volume (liters)</u>	<u>^{238}Pu (grams)</u>	<u>^{237}Np (grams)</u>
<u>Feed Solution</u>			
Batch A	147	23.3	219
Batch B	190	21.4	191
Total	337	44.7	410
<u>Product Solutions</u>			
Np Tank 9	18*	0.082	0.446
Np Tank 8	10	6.78	329
Pu Bottle 1	3.5*	0.038	0.0008
Pu Bottle 2	3.5	31.5	0.78
Pu Bottle 3	2.0	1.15	0.0002
Pu Bottle 4	3.5*	0.062	0.0001
<u>Process Waste Solutions</u>			
Tank 3	620*	1.43	59.1
Tank 2	170	0.887	6.00
Tank 1	346*	0.021	0.066
Tank 3	55	0.43	0.029
<u>Accountability</u>			
grams accounted		42.4	395
% of feed		95	96

Note: The solutions marked with * were discarded to waste.

collected in Tank 3. Tank 1 was then used to accumulate the column effluent waste solution from the last 100 l of the co-absorption cycle and part of the co-decontamination cycle.

All of the ion-exchange columns in A-cell are jacketed and can be heated or cooled. The run plan designated that the resin be removed from the largest column (250 l capacity) and the column be used as a tank to accumulate the effluent solution from the Pu partition cycle (step No. 4), and to prepare the Pu feed for the final Pu purification cycle (step No. 6). This would permit containment of all liquid waste within the cell until the run was completed and analytical results could be obtained on the solution held in each tank. Unfortunately, 20 to 30 l of water were left in the column after the resin was removed and this error was not detected until after the partition solution had been accumulated in the column and part of the acid added to adjust to 7.5M HNO_3 (step No. 6). The Pu analysis of tank 1 waste was known but not the Np analysis. In order to continue with the run, the assumption was made that the Np value would not be greatly different from the Pu value and the waste solution was transferred to a large vault waste tank. Tank 1 was then decontaminated and used to continue the step No. 6 feed adjustment.

Two conclusions can be drawn from a review of the material balance data given in Table II. The high waste losses, particularly of Np, suggest that the resin capacity for the co-absorption step was not as great for this particular feed solution as had been calculated. The high ratio of Al to Np and the significant amount of energy released from the ^{238}Pu were probably the most significant factors leading to a lower resin capacity. The second conclusion is that the partition step was not completely successful and left too much Pu in the Np as well as too much Np in the Pu product. The partition solution composition is 5.7M HNO_3 , 0.05M hydrazine and 0.05M ferrous sulfamate. The ferrous sulfamate undergoes oxidation by the nitric acid at 25°C and the rate is increased substantially at 35°C. The poor performance of the partition cycle can be attributed to two factors: (a) a significant time lag between preparation of the partition solution and transfer of the last half of the solution into the cell (the partition cycle extended over a 16 hour period), and (b) mild heating of the solution by the centrifugal pump used to transfer the solution into the cell.

Following completion of the first run, each solution was analyzed. Some were sufficiently low in Pu and Np to be discarded to waste and others were recycled through a second purification run. The results are summarized in Table III.

Prior to the second purification run, steps were taken to correct the partition cycle. The centrifugal pump was replaced by a non-heating Lapp pump, provisions were made to cool the partition solution before it entered the cell, and the solution was made up in four separate batches as needed. These precautions improved the performance of the partition cycle in run 2 and subsequent runs. Table IV summarizes the results of run 2. The three Np product fractions were reserved for later loadout. The Pu product fraction, 2-9, was loaded out and returned to ARHCO.

NpO₂ - GRAPHITE TARGET ELEMENTS

Three batches of graphite elements were obtained from DUN. The first shipment included 12 elements irradiated in the "center" of the reactor (designated hereafter as group II), and six elements irradiated in the fringe of the reactor (designated hereafter as group III). The group II elements were numbered GA-21 through GA-26 plus GZ-31 through GZ-36. The group III elements were numbered GZ-41 through GZ-46. The second shipment of graphite elements, discharged at a later date, were designated as group IV. The cans were stripped off at Radiometallurgy and the graphite pellets, segregated in three containers, were transferred into C-cell of 325 Building. A small number of unirradiated Np-graphite pellets was transferred from R. E. Learch for Np recovery and were designated as group V. During the burning and dissolution steps, the groups of pellets were kept separate to maintain their identity as a function of the irradiation history.

The burning was performed on a batch basis in a stainless steel pot approximately six inches ID by 10 inches high. The pot was lowered into a small open-top resistance furnace and the furnace temperature controlled manually. A thermocouple in a well on the side of the pot provided temperature information. The pot lid was sealed on carefully machined flat surfaces and was held in place by simple clamps. The off-gas from the pot was drawn through a water-cooled

Table IIIMaterial Balance - Ion Exchange Run 2

	<u>Solution Volume (liters)</u>	<u>^{238}Pu (grams)</u>	<u>^{237}Np (grams)</u>
<u>Feed Solution</u>			
Accumulation from 1st run	240	40.7	336
<u>Product Solutions</u>			
Np - 2-1	3.0*	0.306	0.927
Np - 2-2	3.5	0.00019	199.2
Np - 2-3	3.5	0.0082	115.2
Np - 2-4	2.0	0.40	3.26
Pu - 2-8	2.5*	0.019	0.00008
Pu - 2-9	3.5	36.05	0.0166
Pu - 2-10	3.0*	0.161	0.00027
<u>Process Waste Solutions</u>			
Tank 3	575*	0.0445	<1.5
" 2	118*	0.00169	<0.10
" 1	245*	0.057	<1.7
" 9	42*	0.0515	<0.17
" 10	40*	0.255	<1.6
" 12	55*	0.227	<1.2
" 13	33.7*	0.0337	<0.15
<u>Accountability</u>			
grams accounted		37.6	325
% of feed		92	97

Note: The solutions marked with * were discarded to waste.

Table IVAnalytical Data - Pu Product
Recovered from Aluminum Elements

Volume 3.50 liters

 ^{238}Pu content 10.3 (g/l). 36.05 (grams) ^{237}Np content 0.00475 (g/l) 0.0166 (grams)Gamma Energy Analyses ^{144}Ce (dis/min-ml) $<3.2 \times 10^5$ ^{141}Ce " $<4 \times 10^4$ ^{233}Pa " $<2.1 \times 10^3$ ^{103}Ru " $<3.6 \times 10^3$ ^{106}Ru " $<1 \times 10^3$ ^{137}Cs " $<3 \times 10^2$ ^{95}Zr " $<5.4 \times 10^3$ ^{95}Nb " 9.9×10^4 Isotopic analysis (duplicate samples)

^{238}Pu	90.22 ± 0.08	wt%	90.35	± 0.08	wt%
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^{239}Pu	8.57 ± 0.08	"	8.47	± 0.08	"
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^{240}Pu	1.06 ± 0.02	"	1.05	± 0.02	"
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^{241}Pu	0.124 ± 0.005	"	0.115	± 0.005	"
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^{242}Pu	0.015 ± 0.001	"	0.012	± 0.002	"
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Uncertainties are the 95% confidence level.

 ^{236}Pu $4.3 \times 10^{-5} \pm 0.8$ wt% (based on total Pu)

heat exchanger and then through the C-cell dissolver off-gas system. Pure oxygen was introduced into the pot through a ring near the bottom of the pot. Approximately 10 to 12 pellets could be placed in the pot and burned to metal oxide in each eight hour period. The burning temperature target was 800°C. The reaction time at temperature was about six hours and could be detected by observing the temperature readout record. The burning process is exothermic and once started, the furnace power was decreased to maintain a constant temperature. No attempt was made to determine the oxygen consumption as a function of graphite burned.

The solid residues from the burning step were separately dissolved in a 4-liter stainless steel beaker on a hot plate with boiling concentrated HNO_3 . It was necessary to add a small amount of HF to initiate the reaction. Because of the problem of determining when dissolution was complete, the process was continued for eight hours for each batch. Extra acid was added to maintain a volume of approximately three liters. After cooling, each dissolver solution was filtered through paper and the paper plus residue washed with dilute acid. After each of the four segregated dissolver solutions had been filtered, the filter papers plus enclosed residues were placed in the burning pot, dried, and heated at 800°C for eight hours with a normal oxygen flow. The contents of the pot were then leached in the pot with concentrated HNO_3 plus a trace of HF for eight hours at near-boiling temperature. After cooling and filtering, this leachate was designated as "scrap recovery." The four dissolver solutions and the scrap recovery solution were then each diluted to 3.50 liters in their separate containers and sampled in duplicate. The analyses from these samples are shown in Table V.

The four batches of dissolver solution and the scrap recovery solution were then combined together and adjusted to give Pu^{IV} and Np^{IV} at an acid concentration of 7.5M and a volume of 200 liters. This feed solution was then processed in two separate runs (100 liters of feed for each run) by the ion-exchange process previously described. The material balance results for the two runs are given in Tables VI and VII. The recoveries for these two runs are quite good and the Np product was within specifications for each run. The Pu product, in each run, contained approximately 4 wt% Np and therefore did not meet purity specifications. The inability of this process to reduce the Np contamination in the Pu product

Table V

Np-Graphite Dissolver Solution Analytical Data

	Group II		Group III		Group IV	
Volume (liters)	3.50		3.50		3.50	
HNO ₃ conc (M)	2.75	2.80	4.16	4.16	6.01	6.04
²³⁸ Pu conc (g/l)	3.58	3.67	0.414	0.428	1.42	1.43
²³⁸ Pu total (grams)	12.5	12.8	1.45	1.50	4.97	5.01
²³⁷ Np conc (g/l)	114.2	118.5	60.7	58.2	40.7	39.8
²³⁷ Np total (grams)	400	415	213	204	142	139
¹³⁷ Cs Burnup (atoms/ml)	1.48×10^{16}	1.44×10^{16}	3.52×10^{14}	3.40×10^{14}	9.55×10^{15}	9.16×10^{15}
²³⁶ Pu (wt%, based on total Pu)	$3.2^{+0.4} \times 10^{-5}$	$3.0^{+0.5} \times 10^{-5}$	$2.0^{+0.4} \times 10^{-5}$	$1.7^{+0.5} \times 10^{-5}$	$3.1^{+0.9} \times 10^{-5}$	$3.5^{+0.6} \times 10^{-5}$
Pu isotopics (wt%)						
238	$96.93^{+0.02}$	$96.92^{+0.01}$	$99.134^{+0.009}$	$99.144^{+0.008}$	$96.68^{+0.02}$	$96.68^{+0.01}$
239	$2.97^{+0.02}$	$2.97^{+0.01}$	$0.847^{+0.009}$	$0.840^{+0.008}$	$3.17^{+0.02}$	$3.16^{+0.02}$
240	$0.094^{+0.002}$	$0.093^{+0.01}$	$0.019^{+0.002}$	$0.016^{+0.002}$	$0.146^{+0.002}$	$0.148^{+0.002}$
241	$0.006^{+0.006}$	$0.010^{+0.01}$	<0.007	<0.007	$0.011^{+0.001}$	$0.011^{+0.001}$
242	$<0.007^{+0.007}$	$<0.007^{+0.007}$	<0.007	<0.007	<0.007	<0.007

Note: The Pu isotopic data are at the 95% confidence level.

Table V (cont.)
Np-Graphite Dissolver Solution Analytical Data

	<u>Group II</u>		<u>Group III</u>		<u>Group IV</u>	
<u>Gamma Energy Analyses</u>						
¹⁴⁴ Ce (dis/min-ml)	8.99x10 ⁹	9.76x10 ⁹	2.46x10 ⁸	2.44x10 ⁸	7.14x10 ⁹	7.36x10 ⁹
¹⁴¹ Ce "	1.32x10 ⁸	1.37x10 ⁸	3.53x10 ⁶	3.60x10 ⁶	4.72x10 ⁹	4.60x10 ⁴
¹⁰⁶ Ru "	3.37x10 ⁸	3.35x10 ⁸	1.29x10 ⁷	1.59x10 ⁷	7.70x10 ⁸	7.58x10 ⁸
¹⁰³ Ru "	<6.19x10 ⁶	<1.06x10 ⁷	<5.39x10 ⁵	1.17x10 ⁶	1.59x10 ⁹	1.59x10 ⁹
¹³⁷ Cs "	5.61x10 ⁸	6.20x10 ⁸	1.54x10 ⁷	1.46x10 ⁷	4.31x10 ⁸	3.93x10 ⁸
¹³⁴ Cs "	1.15x10 ⁷	1.28x10 ⁷	<1.83x10 ⁵	<3.27x10 ⁴	<1.65x10 ⁵	<1.63x10 ⁵
⁹⁵ Zr "	2.38x10 ⁹	2.63x10 ⁹	6.63x10 ⁷	6.55x10 ⁷	1.11x10 ¹⁰	1.09x10 ¹⁰
⁹⁵ Nb "	2.21x10 ⁹	2.43x10 ⁹	2.81x10 ⁸	1.27x10 ⁸	1.51x10 ¹⁰	1.47x10 ¹⁰
²³³ Pa "		1.58x10 ⁸	8.73x10 ⁷	8.71x10 ⁷	1.28x10 ⁸	

Table V (cont.)
Np-Graphite Dissolver Solution Analytical Data

	<u>Unirradiated from Learch</u>		<u>Scrap Recovery</u>	
Volume (liters)	3.50		3.05	
HNO ₃ conc (M)	6.03	6.09	0.84	0.86
²³⁸ Pu conc (g/l)	0.148	0.144	0.0144	0.0139
²³⁸ Pu total (grams)	0.52	0.50	0.05	0.05
²³⁷ Np conc (g/l)	7.90	7.97	0.549	0.540
²³⁷ Np total (grams)	27.7	27.9	1.92	1.89
¹³⁷ Cs Burnup (atoms/ml)				
²³⁶ Pu (wt%)	3.2 [±] .9x10 ⁻⁵		2.4 [±] .7x10 ⁻⁵	
Pu isotopics (wt%)				
238	88.28 [±] 0.05	88.31 [±] 0.05	93.39 [±] 0.03	93.41 [±] 0.03
239	9.84 [±] 0.05	9.81 [±] 0.05	5.85 [±] 0.03	5.85 [±] 0.03
240	1.51 [±] 0.01	1.51 [±] 0.001	0.625 [±] 0.006	0.617 [±] 0.009
241	0.295 [±] 0.002	0.292 [±] 0.002	0.108 [±] 0.003	0.103 [±] 0.003
242	0.078 [±] 0.002	0.077 [±] 0.002	0.030 [±] 0.003	0.026 [±] 0.003

Note: The Pu isotopic data are at the 95% confidence level.

Table VI
Material Balance Results - Ion Exchange Run 3

	<u>Solution Volume (liters)</u>	<u>^{238}Pu (grams)</u>	<u>^{237}Np (grams)</u>
<u>Feed Solution</u>	100	9.85	393
<u>Product Solutions</u>			
Np - 3-1	3.50	0.0137	311.2
Np - 3-2	3.50	0.72	58.5
Np - 3-3	3.00	0.136	2.42
Pu - 3-1	3.50*	0.0015	0.00118
Pu - 3-2	3.00	9.51	0.489
Pu - 3-3	2.00*	0.007	0.0018
<u>Process Waste Solutions</u>			
Tank 3	456*	0.059	<14.6
Tank 2	321*	0.133	< 1.76
<u>Accountability</u>			
grams accounted		10.58	389
% of feed		107	99

Note: The solutions marked with * were discarded to waste.

Table VII
Material Balance Results - Ion Exchange Run 4

	<u>Solution Volume (liters)</u>	<u>^{238}Pu (grams)</u>	<u>^{237}Np (grams)</u>
<u>Feed Solution</u>	100	9.85	393
<u>Product Solutions</u>			
Np - 4-1	3.50	0.0007	48.7
Np - 4-2	3.50	0.201	252.7
Np - 4-3	3.50	0.109	29.2
Np - 4-4	3.50	0.033	16.8
Np - 4-5	3.50	0.021	11.0
Np - 4-6	3.50	0.013	4.03
Np - 4-7	3.50*	0.008	0.20
Np - 4-8	3.50*	0.001	0.003
Pu - 4-1	3.50*	0.0021	0.0002
Pu - 4-2	3.00	8.76	0.268
Pu - 4-3	3.00*	0.012	0.0003
<u>Process Waste Solutions</u>			
Tank 3	431*	0.093	<12.5
Tank 2	320*	0.123	< 2.8
Tank 1	312*	0.052	< 1.2
<u>Accountability</u>			
grams accounted		9.42	379
% of feed		96	97

Note: The solutions marked * were discarded to waste.

below about 4 wt% in one cycle perhaps represents a limit of process itself or of the equipment employed for the process.

A comparison of Table VII with Table VI shows that the Np product elution for run 4 required twice the volume of solution that run 3 required. The eluant composition and flowrate were the same for the two runs. The reason for the extended elution for run 4 is not known, but the longer elution did not cause problems with the recovery or purity of the product.

The ^{238}Pu accountability for run 3, given in Table VI, is greater than 100%. Part of the reason for this high number is the broad uncertainty range in the analytical results and part results from not dividing the feed solution for runs 3 and 4 into two exactly equal volumes.

The radiochemical impurities in each of the Np product fractions from runs 2, 3, and 4 are given in Table VIII. The Np product solutions were combined, thermally concentrated, air sparged at 90°C to ensure oxidation to Np^{V} or Np^{VI} , and loaded out. Analytical data on the combined solution as loaded out, are given in Table IX.

The Pu product solutions from runs 3 and 4 were combined together and reprocessed by ion exchange. The process was similar to that used previously except that both absorption cycles were carried out on the 4-liter column. The material balance results are given in Table X and the product purity data in Table XI.

Table VIII

Radiochemical Impurities in Np Product Fractions

Fraction No.	2-2	2-3	2-4	3-1	3-2	3-3	4-1	4-2	4-3	4-4	4-5	4-6
Volume (liters)	3.50	3.50	2.00	3.50	3.50	3.00	3.50	3.50	3.50	3.50	3.50	3.50
²³⁷ Np Content (grams)	199	115	3.3	311	58.5	2.4	47.8	253	29.2	16.8	11.0	4.0
Radiochemical Impurities (μ Ci/gram Np)												
¹⁴⁴ Ce	--	0.7	35	0.7	5.4	16	2.2	1.2	4.0	8.2	3.7	1.0
¹⁴¹ Ce	--	--	--	0.5	6.2	5.6	--	--	--	--	--	--
¹⁰³ Ru	0.2	0.3	16	0.04	0.5	6.2	1.3	0.8	3.0	3.0	3.7	13
¹⁰⁶ Ru	0.8	1.2	61	0.08	0.8	7.9	0.8	0.5	2.7	2.0	4.5	12
¹³⁷ Cs	--	--	--	0.03	0.09	0.8	--	--	--	--	--	--
⁹⁵ Zr	--	--	--	0.05	0.02	1.1	--	--	--	--	--	--
⁹⁵ Nb	2.5	4.4	90	1.4	1.3	7.3	44	3.1	11	16	19	19
²³³ Pa	--	68	--	27	53	50	19	20	20	18	17	6.5

Note: The blank spaces were reported at insignificantly low values.

Table IX
Neptunium Product From All Runs
As Returned to ARHCO

Solution Volume (liters)	26.75
^{238}Pu Content (grams)	1.68
^{237}Np Content (grams)	1052
Wt% Pu Impurity	0.16
Radiochemical Impurities ($\mu\text{Ci/gram Np}$)	
^{144}Ce	0.009
^{141}Ce	1.07
^{103}Ru	0.3
^{106}Ru	1.0
^{137}Cs	0.1
^{95}Zr	--
^{95}Nb	1.2
^{233}Pa	38

Table X
Material Balance Results - Ion Exchange Run 5

	<u>Solution Volume (liters)</u>	<u>^{238}Pu (grams)</u>	<u>^{237}Np (grams)</u>
<u>Feed Solution</u>	14	18.3	0.76
<u>Product Solutions</u>			
Pu-5-1	3.50*	0.021	not determined
Pu-5-2	3.00	17.01	0.0014
Pu-5-3	3.50*	0.092	not determined
<u>Process Waste Solution</u>			
Tank 2	319*	1.09	" "
<u>Accountability</u>			
grams accounted		18.2	
% of fuel		99.5	

Note: The solutions marked with * were discarded to waste.

Table XI

²³⁸Pu Product Recovered from Graphite Elements
Analytical Results as Sampled in A-Cell

Volume (liters)		3.00
²³⁸ Pu content	5.67 (g/l)	17.01 grams
²³⁷ Np content	0.000456 (g/l)	0.00136 grams
%Np impurity	0.008 wt%	

Gamma Energy Analyses

¹⁴⁴ Ce (dis/min-ml)		$< 1.79 \times 10^4$
¹⁴¹ Ce	"	$< 1.37 \times 10^2$
¹⁰³ Ru	"	$< 2.98 \times 10^2$
¹⁰⁶ Ru	"	$< 6.53 \times 10^3$
¹³⁴ Cs	"	$< 5.07 \times 10^2$
¹³⁷ Cs	"	$< 2.42 \times 10^3$
⁹⁵ Zr	"	$< 1.78 \times 10^4$
⁹⁵ Nb	"	3.46×10^5

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