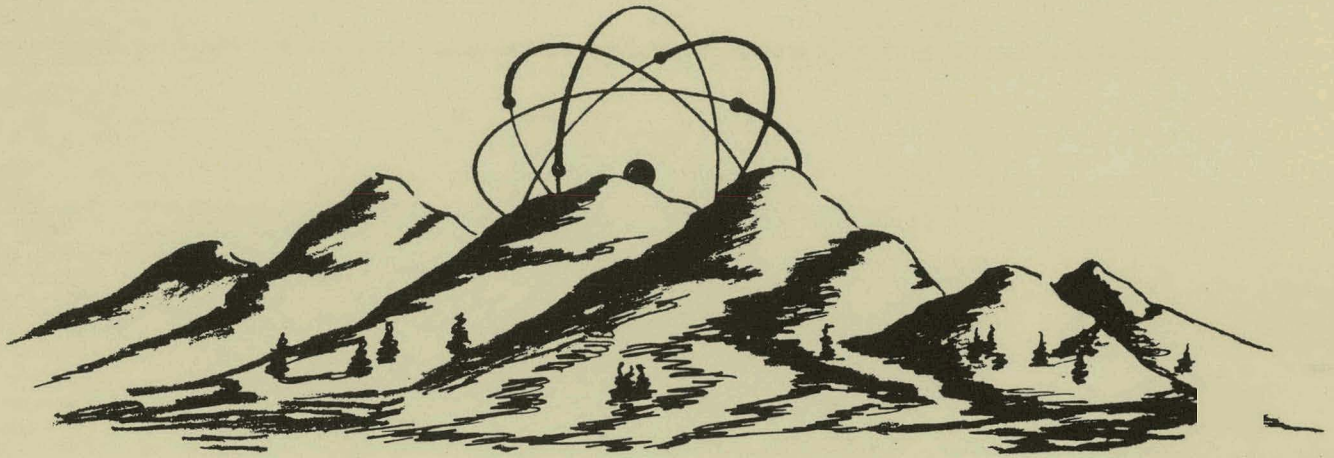


phil
267
12/27/68

MASTER



DISSOLUTION OF PLUTONIUM-BEARING RESIDUES IN A
NITRIC ACID-CALCIUM FLUORIDE SOLVENT SYSTEM

by

G. F. Molen
R. O. Wing

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS



THE DOW CHEMICAL COMPANY
ROCKY FLATS DIVISION
P. O. BOX 888
GOLDEN, COLORADO 80401
U.S. ATOMIC ENERGY COMMISSION
CONTRACT AT(29-1)-1106

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

Printed in USA. Price ~~\$2.~~ Available from the Clearinghouse for Federal
Scientific and Technical Information, National Bureau of Standards,
U. S. Department of Commerce, Springfield, Virginia 22151

22 November 1966

RFP-776
UC-4 CHEMISTRY
TID-4500

CFSI PRICES

LA 3.00; MN .65

DISSOLUTION OF PLUTONIUM-BEARING RESIDUES IN A NITRIC ACID-CALCIUM FLUORIDE SOLVENT SYSTEM

by

G. F. Molen
R. O. Wing

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

THE DOW CHEMICAL COMPANY
ROCKY FLATS DIVISION
P. O. BOX 888
GOLDEN, COLORADO 80401
U.S. ATOMIC ENERGY COMMISSION
CONTRACT AT(29-1)-1106

ACKNOWLEDGMENT

We wish to thank N. E. Moody of the Plutonium Analytical Laboratory and his x-ray fluorescence and radioassay groups for their cooperation in performing the plutonium analyses required for this study. For many very helpful technical discussions we wish to thank J. N. Vance and E. D. Erickson of the Manufacturing Technical Group.

CONTENTS

	Page No.
Acknowledgment.....	ii
Abstract.....	iv
Introduction.....	1
Summary and Conclusions.....	1
Recommendations.....	1
Experimental.....	2
Dissolution Procedure for Single Contact.....	2
Length of the Reflux Period.....	2
Drying Temperature.....	2
Solid to Liquid Ratio.....	2
Adding Additional Calcium Fluoride.....	2
Multiple Contacts.....	2
Four Fresh Acid-Fluoride Contacts.....	2
Two Fresh Acid-Fluoride Contacts.....	3
Solid to Liquid Ratio and Multiple Contacts.....	3
Four Contacts and Reused Acid.....	3
Three Contacts and Reused Acid.....	3
Discussion.....	3
Length of Reflux Period.....	3
Drying Temperature.....	4
Solid to Liquid Ratio.....	5
Adding Additional Calcium Fluoride.....	5
Multiple Contacts.....	6
Four Fresh Acid-Fluoride Contacts.....	6
Two Fresh Acid-Fluoride Contacts.....	8
Solid to Liquid Ratio and Multiple Contacts.....	9
Four Contacts and Reused Acid.....	10
Three Contacts and Reused Acid.....	11
Appendix – Experimental Data.....	13

ABSTRACT

The dissolution of plutonium-bearing residues in a nitric acid-calcium fluoride solvent system is described. The length of the reflux period, the drying temperature of heels, the solid to liquid ratio, the addition of fluoride during dissolution, and the effectiveness of a multiple contact system are evaluated in terms of the effects they have on the dissolution of these residues.

INTRODUCTION

result of fabricating and machining of plutonium metal, many plutonium-bearing residues are generated. These include incinerator ash (ash resulting from the incineration of plutonium contaminated burnables) and glovebox floor sweepings (plutonium containing material that is inadvertently dropped on the floor of a glovebox). To recover the plutonium, the plutonium-bearing materials are dissolved in nitric acid containing calcium fluoride. The plutonium is purified by anion exchange. Many times the material is quite difficult to dissolve in this solvent system. The undissolved residue remaining after an attempted dissolution is called heel. Thus there are two additional types of plutonium-bearing residues which must be considered. They are incinerator ash heel and glovebox floor sweepings heel.

The purpose of this study was to determine the variables affecting the dissolution of these residues. Those considered in this report are the length of the reflux period, the temperature at which the heel was dried, the solid to liquid ratio, the addition of calcium fluoride during the dissolution, and the effectiveness of multiple contacts.

A previous study⁽¹⁾ had indicated that a solid to liquid ratio of 100 g/l was probably optimum and that a calcium fluoride concentration of 0.2M seemed optimum. Consequently, many of the experiments performed in this study used these conditions.

The Rocky Flats production process uses 400 grams of feed material (ash, sweepings or heel) with approximately 4 liters of nitric acid (~12.5M) and approximately 50 grams of calcium fluoride. The solution is boiled for 3 hours. After 3 hours, the solution is cooled for 20 minutes and the supernatant is decanted from the undissolved residue. Another 400 grams of feed material is added to the dissolver pot and additional acid and fluoride are added. The procedure is repeated until five charges have been made. All of the remaining undissolved residue is again contacted without any additional charge being added. This is called a heel run. After the heel run, the undissolved residue is removed, dried and stored until an attempt is made to dissolve it again. Although as many as four attempts have been made to recover plutonium from these heels, some still contain 1-10 percent plutonium. The present study was initiated because of this difficulty of achieving a discardable (<1 percent Pu) heel. Emphasis was placed on studying the effects of multiple contacts.

SUMMARY AND CONCLUSIONS

Several variables were evaluated in terms of the effects they have on the dissolution of plutonium-bearing residues. The parameters considered were the length of the reflux period, the drying temperature of heels, the solid to liquid ratio, the addition of fluoride during dissolution, and the number of contacts.

The length of the reflux period was varied from 15 to 150 minutes. The data from these experiments showed that plutonium dissolution had reached a maximum at a 90 minute reflux period. Longer reflux periods dissolved more non-plutonium solids. This resulted in a higher plutonium concentration in the undissolved residue.

Concentrated nitric acid (15.6M) was also evaluated as a solvent. There were no differences between it and technical grade nitric acid (12.5M) for residue dissolution.

The drying temperature of heels was varied from 350° to 950°C. A drying temperature greater than 650°C drastically reduced the effectiveness of the 12.5M HNO₃-0.2M CaF₂ solvent system.

The solid to liquid ratio was varied from 25 g/l to 250 g/l. A ratio greater than 50 g/l for a single contact was detrimental to the dissolution of the residues.

Calcium fluoride was added during the dissolution in amounts equal to 25, 50 and 100 percent of the original amount added. These additions did not significantly improve the solubility of the residue.

Several systems of multiple contacts were evaluated. They included four fresh acid-fluoride contacts; two fresh acid-fluoride contacts; four contacts, two of which were with "used" acid; and three contacts, one of which was with "used" acid. All of the multiple contact systems evaluated were found to be more effective for dissolution of plutonium-bearing residues than a single contact for the same length of time. A system of three contacts, where the first was with a "used" acid (i.e., the filtrate from a fresh acid contact) and the other two were with fresh acid, was the most successful multiple contact system evaluated. This system resulted in the greatest plutonium recoveries with the lowest undissolved residue concentration while only tripling the filtrate volume.

RECOMMENDATIONS

1. A multiple contact system consisting of three contacts is recommended. The first contact should be with "used" acid for one hour of refluxing; the second contact should be with fresh acid for a reflux period of 90

(1) Butler, F. E., Moseley, J. D., and Molen, G. F., *Recovery of Plutonium from Incinerator Ash*, The Dow Chemical Company, Rocky Flats Division, July 31, 1964, RFP-396.

minutes; and the third contact should be with fresh acid for one hour of refluxing.*

2. An initial solid to liquid ratio of 100 g/l for the three contact system is recommended.**
3. A calcium fluoride concentration of 0.2M for each contact is recommended.
4. The reflux period per contact should never be longer than 90 minutes.
5. When drying heels that have to be reprocessed, the drying temperature should never exceed 650°C.

EXPERIMENTAL

DISSOLUTION PROCEDURE FOR SINGLE CONTACT

The procedure was as follows: Fifty grams of the feed material, 500 ml of the acid and 8.0 grams (0.2M) of calcium fluoride were refluxed in a 500 ml Pyrex Florence flask which rested on a Thermolyne Stirplate that both heated and stirred (magnetically) the solution. The stirring speed (set on high) was held constant throughout all the experiments. Refluxing conditions (110-115°C) were maintained by a water cooled condenser. For residues which contained large amounts of silicon the condenser had to be removed periodically and freed of the deposits of silicon dioxide formed by the hydrolysis of the volatile silicon tetrafluoride.

At the end of the reflux period the hot solution was filtered through a glass porosity (medium) filter and the undissolved residue was washed with 200 ml of 0.5M HNO₃. The undissolved residue was then air dried. The entire residue was accurately weighed, dried at 800°C, reweighed and analyzed for plutonium content by x-ray fluorescence. The filtrate volume was measured, sampled and sent to the laboratory for plutonium analysis.

Length of the Reflux Period

Experiments were performed to determine what effect the length of the reflux period had on the dissolution of the residue. The feed material was incinerator ash heel, glovebox floor sweepings heel, filter sludge, and mixtures of these. These residues contained from 12-14 percent plutonium. The solid to liquid ratio was 100 g/l. The solvent used was 12.5M HNO₃-0.2M CaF₂. The length of the reflux period was varied from 15 to 150 minutes.

* This recommendation is based on the equipment and process used in the present Rocky Flats production facility. If a new facility were being installed a three or four stage batch counter-current system would be recommended.

** A solid to liquid ratio of 50 g/l is recommended for a single contact process.

Drying Temperature

Incinerator ash heel was dried at 350°, 450°, 550°, 650°, 750°, 850° and 950°C for 3 hours. This feed material contained -20 percent plutonium. The dissolution procedure was the same as that described except the reflux period was 2 hours.

Solid to Liquid Ratio

In order to determine the effect of the solid to liquid ratio on the dissolution of plutonium-bearing residues, a set of experiments was performed in which the ratio was varied from 25 g/l to 250 g/l. The reflux period was one hour and the feed materials were incinerator ash, incinerator ash heel, glovebox floor sweepings and glovebox floor sweepings heel. The procedure was the same as described above.

Adding Additional Calcium Fluoride

Incinerator ash usually contains 15 to 30 percent silicon. During dissolution the silicon is volatilized as silicon tetrafluoride thus removing fluoride from the solvent. Consequently, experiments were performed to determine if the addition of calcium fluoride during the reflux period improved the dissolution of the residue. Crushed incinerator ash, containing approximately 20 percent plutonium, was the feed material. The reflux period was 2 hours for each sample. An additional amount of calcium fluoride was added to each sample after one hour of refluxing. The amount added was equal to 25, 50 and 100 percent of the initial calcium fluoride concentration of the sample. The procedure was the same as described previously.

MULTIPLE CONTACTS

Four Fresh Acid-Fluoride Contacts

In this group of experiments 50 grams of feed material were contacted with 500 ml of 12.5M HNO₃-0.2M CaF₂ for a given reflux period. At the end of the reflux period the hot solution was filtered, the filtrate volume measured, sampled and the undissolved residue was quantitatively returned to the reaction flask. Five hundred milliliters of 12.5M HNO₃-0.2M CaF₂ were added and the solution was refluxed for the same length of time as before. This procedure was repeated until four contacts of the original feed material had been made. After the fourth contact the solution was filtered and the filtrate volume was measured and sampled. The undissolved residue was air dried, accurately weighed, dried at 800°C, reweighed and the plutonium content determined by x-ray fluorescence. The feed materials used were uncrushed incinerator ash, crushed incinerator ash, incinerator ash heel, glovebox floor sweepings, glovebox floor sweepings

heel, and filter sludge. The reflux periods were 15, 30, 60 and 120 minutes for each of the four contacts.

Two Fresh Acid-Fluoride Contacts

The feed materials used were crushed incinerator ash, incinerator ash heel, glovebox floor sweepings, and glovebox floor sweepings heel. The solid to liquid ratio was 100 g/l for the first contact. The solvent was 12.5M HNO₃-0.2M CaF₂ for each contact. The procedure was the same as that described for the four contact experiments except that only two contacts were used.

Solid to Liquid Ratio and Multiple Contacts

Initial solid to liquid ratios of 100, 150 and 200 g/l were used with a two and a four contact system. The reflux period for both systems was one hour per contact. Only crushed incinerator ash and incinerator ash heel were used for feed materials. The procedure was the same as described for the four contacts.

Four Contacts and Reused Acid

The first two contacts were made with fresh 12.5M HNO₃-0.2M CaF₂. The third and fourth contacts were made with a once-used acid (i.e., the filtrate from a fresh acid contact) and a twice-used acid respectively. The feed materials were the same as those used for the two contact experiments. The initial solid to liquid ratio was 100 g/l and the reflux period was one hour per contact.

Three Contacts and Reused Acid

The first contact was with once-used acid (-12M H⁺) and 0.2M CaF₂ for a one hour reflux period. The second contact was with 12.5M HNO₃-0.2M CaF₂ for 2 hours. An additional

amount of calcium fluoride (0.1-0.2M) was added after the first hour of refluxing. The third contact was with 12.5M HNO₃-0.2M CaF₂ for one hour. The feed materials were the same as those used for the two contact experiments.

DISCUSSION

The effectiveness of the treatments were determined by calculations as follows:

$$1) \% \text{ solid dissolved} = \frac{\text{wt. of feed} - \text{wt. of undissolved residue}}{\text{wt. of feed}} \times 100$$

$$2) \% \text{ plutonium recovered} = \frac{\text{wt. of plutonium in solution}}{\left\{ \begin{array}{l} \text{wt. of plutonium} \\ \text{in solution} \end{array} \right\} + \left\{ \begin{array}{l} \text{wt. of plutonium} \\ \text{in undissolved} \\ \text{residue} \end{array} \right\}} \times 100$$

$$3) \% \text{ plutonium in feed} = \frac{\left\{ \begin{array}{l} \text{wt. of plutonium} \\ \text{in solution} \end{array} \right\} + \left\{ \begin{array}{l} \text{wt. of plutonium} \\ \text{in undissolved residue} \end{array} \right\}}{\text{wt. of feed}} \times 100$$

$$4) \% \text{ plutonium in undissolved residue} = \text{g Pu/g residue analysis} \times 100$$

The experimental data are tabulated in the appendix and are summarized in the following tables and figures.

Length of Reflux Period

Table I and Figure 1 summarize the experiments concerning the effect of the length of the reflux period on the dissolution of plutonium-bearing residues. These data indicate that the optimum reflux period is about 60 minutes. The amount of solid dissolved does not increase appreciably after 60 minutes of refluxing. A large amount (-68 percent) of plutonium is recovered in a 15 minute reflux period. A 60 minute reflux period increased the amount of plutonium recovered by only 20 percent. A 150 minute reflux period increased the amount of plutonium recovered by 25 percent. This increase is even less in terms of the amount of solid dissolved (10 percent).

TABLE I
EFFECT OF LENGTH OF REFLUX PERIOD ON DISSOLUTION OF RESIDUES. SINGLE CONTACT

Solvent: 12.5M HNO₃-0.2M CaF₂ Solid to Liquid Ratio: 100 g/l
Feed Material: Incinerator ash heel, glovebox floor sweepings heel, filter sludge, and mixtures of these

Reflux Period (min.)	No. of Runs	Initial Feed Conc. (% Pu)	Filtrate Conc. (g Pu/l)	Amt of Solid Dissolved (%)	Amt of Plutonium Dissolved (%)	Amt of Plutonium Leached (%)	Amt of Plutonium Recovered (%)	Undissolved Residue Conc. (% Pu)
	5	20.9	14.2	51	57.2	42.8	68.4	9.9
	13	22.5	16.2	54	63.7	36.3	69.3	11.7
45	4	23.1	18.4	56	59.7	40.3	77.6	9.4
60	8	24.6	20.4	59	60.0	40.0	84.4	8.1
90	7	29.6	26.0	55	51.9	48.1	88.2	7.9
120	4	30.2	25.8	62	64.2	35.8	84.7	9.8
150	4	32.3	28.6	64	63.2	36.8	87.5	7.7

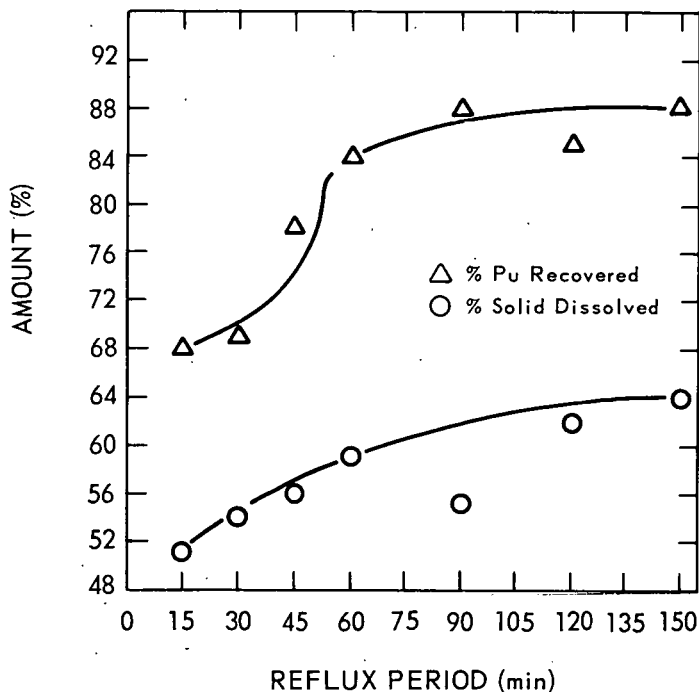


Figure 1. EFFECT OF LENGTH OF REFLUX PERIOD ON DISSOLUTION OF RESIDUES. SINGLE CONTACT.

Solvent: 12.5M HNO_3 -0.2M CaF_2 . Solid to liquid ratio: 100 g/l. Feed material: Incinerator ash heel, glovebox floor sweepings heel, filter sludge, and mixtures of these.

A similar set of experiments was performed using concentrated nitric acid (~15.6M). All other experimental conditions were the same as those above. The data are tabulated in Table II. The more concentrated nitric acid was no more effective for dissolution of these residues than the technical grade nitric acid.

TABLE II

EFFECT OF LENGTH OF REFLUX PERIOD ON DISSOLUTION OF RESIDUES. SINGLE CONTACT. CONCENTRATED NITRIC ACID (~15.6M)

Solvent: 15.6M HNO_3 -0.2M CaF_2 Solid to Liquid Ratio: 100 g/l
Feed Material: Incinerator ash heel, glovebox floor sweepings heel, filter sludge, and mixtures of these

Reflux Period (min.)	No. of Runs	Initial Feed Conc. (% Pu)	Filtrate Conc. (g Pu/l)	Amt of Solid Dissolved (%)	Amt of Pu Rec (%)	Undissolved Residue Conc. (% Pu)
15	4	26.4	17.5	43	62.2	12.0
30	4	20.9	13.6	48	63.5	11.8
45	4	21.7	16.6	49	77.6	13.2
60	8	22.6	16.5	55	73.7	11.4
120	3	18.7	14.8	60	71.5	6.5

Drying Temperature

The data from the experiments to determine the effect of the drying temperature of the heel on dissolution are tabulated in Table III. The average values were used for the graphs in Figure 2. These data clearly show that drying of these heels above 650°C significantly reduces the effectiveness of the 12.5M HNO_3 -0.2M CaF_2 solvent system.

TABLE III

EFFECT OF THE DRYING TEMPERATURE ON DISSOLUTION OF RESIDUES. SINGLE CONTACT

Solvent: 12.5M HNO_3 -0.2M CaF_2
Solid to Liquid Ratio: 100 g/l Feed Material: Incinerator ash heel
Feed Concentration: ~20% plutonium
Length of Drying Period: 3 hours Reflux Period: 2 hours

Drying Temperature (°C)	Amt of Solid Dissolved (%)	Amt of Pu Rec (%)	Undissolved Residue Conc. (% Pu)
350	59	86	2.1
350	60	89	1.8
350	62	86	3.6
Av.	60	87	2.5
450	56	86	1.9
450	54	82	5.4
450	78	87	3.1
Av.	62	85	3.4
550	55	87	1.8
550	54	75	4.0
550	55	76	4.3
Av.	54	79	3.3
650	51	75	3.2
650	52	80	5.0
650	53	77	4.5
Av.	52	77	4.2
750	44	70	8.2
750	42	47	10.0
750	40	49	12.6
Av.	42	55	10.2
850	41	52	6.9
850	40	45	12.8
850	42	44	13.1
Av.	41	47	10.9
950	28	30	13.2
950	31	29	12.5
950	31	28	16.8
Av.	30	28	14.1

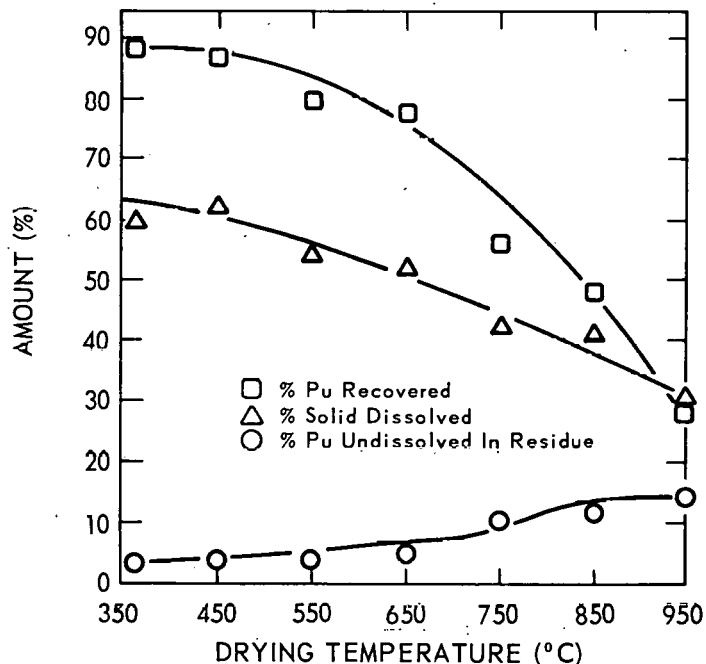


Figure 2. EFFECT OF THE DRYING TEMPERATURE ON DISSOLUTION OF RESIDUES. SINGLE CONTACT.

Solvent: 12.5M HNO_3 -0.2M CaF_2 . Solid to liquid ratio: 100 g/l. Feed material: Incinerator ash heel. Feed concentration: ~20% plutonium. Length of drying period: 3 hours. Reflux period: 2 hours.

Solid to Liquid Ratio

Table IV the data from the experiments on the effect of the solid to liquid ratio on dissolution are tabulated. These average values show that the increase from 50 g/l to 100 g/l has the most significant effect upon the dissolution of these residues. In Figure 3 these average values are graphed. The values tend to level off beyond the 100 g/l ratio. These results show that a solid to liquid ratio less than 100 g/l should be used.

TABLE IV

EFFECT OF SOLID TO LIQUID RATIO ON DISSOLUTION OF RESIDUES. SINGLE CONTACT

Solvent: 12.5M HNO₃-0.2M CaF₂ Reflux Period: One hour

Feed Material	Solid to Liquid Ratio (g/l)	Initial Feed Conc. (% Pu)	Filtrate Conc. (g Pu/l)	Amt of Solid Dissolved (%)	Amt of Pu Rec (%)	Undissolved Residue Conc. (% Pu)
CIA	25	29.0	7.0	89	96.4	4.5
IAH	25	19.1	4.7	67	97.9	0.7
Swps	25	32.6	8.1	86	99.3	1.1
SH	25	38.2	9.4	74	98.7	1.5
Av.	29.7	7.3	79	98.1	2.0	
CIA	50	27.1	11.6	70	85.6	6.5
IAH	50	15.2	7.0	69	91.3	2.7
Swps	50	37.5	18.5	86	98.5	2.6
SH	50	35.6	16.4	69	91.9	7.1
Av.	28.8	13.4	74	91.8	4.7	
CIA	100	26.5	20.1	58	76.1	15.1
IAH	100	13.7	11.7	55	85.4	4.4
Swps	100	26.2	24.3	86	92.5	9.3
SH	100	38.4	24.2	60	63.0	25.2
Av.	26.2	20.1	65	79.2	13.5	
CIA	150	25.3	23.4	48	62.1	18.6
IAH	150	14.1	17.4	51	82.0	5.2
Swps	150	26.7	32.9	84	82.1	19.4
SH	150	38.7	36.5	53	62.7	30.4
Av.	26.2	27.6	59	72.2	18.4	
CIA	200	30.0	37.3	52	62.9	23.6
IAH	200	13.6	22.9	50	84.3	4.3
Swps	200	25.1	40.6	83	80.9	18.1
Av.	22.9	33.6	62	76.0	15.3	
CIA	250	27.3	35.9	45	52.9	23.6
IAH	250	15.1	27.6	39	72.9	6.8
Swps	250	24.6	48.0	84	77.9	21.7
Av.	22.3	37.2	56	67.9	17.4	

CIA - Crushed Incinerator Ash
IAH - Incinerator Ash Heel
Swps - Glovebox Floor Sweepings
SH - Glovebox Floor Sweepings Heel

Effect of Additional Calcium Fluoride

The data from the evaluation of adding fluoride during the dissolution are presented in Table V. The data show that the plutonium concentration in the undissolved residue

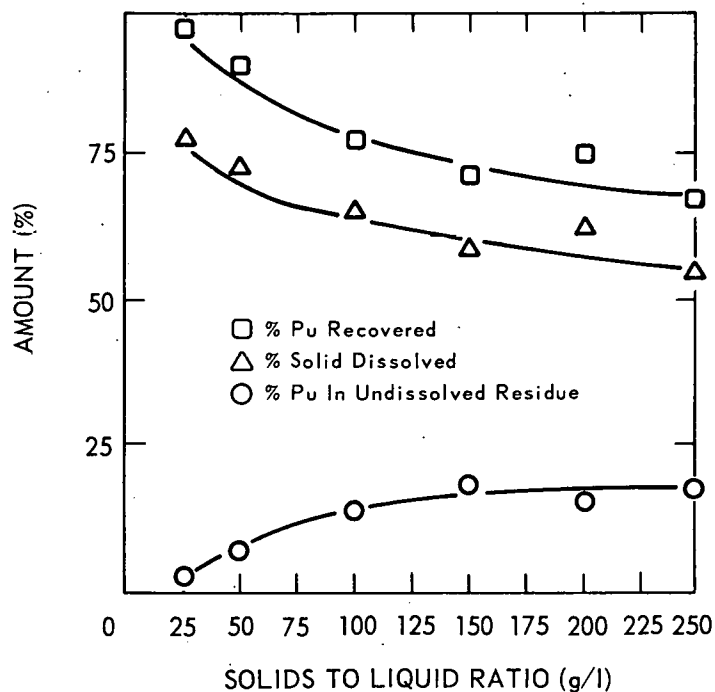


Figure 3. EFFECT OF THE SOLID TO LIQUID RATIO ON DISSOLUTION OF RESIDUES. SINGLE CONTACT.

Solvent: 12.5M HNO₃-0.2M CaF₂. Reflux period: one hour.

decreased 34 percent (2.9-1.9/2.9) with a 25 percent increase in calcium fluoride. A 100 percent increase in calcium fluoride decreased the plutonium concentration in the residue by 55 percent (2.9-1.3/2.9).

TABLE V

EFFECT OF ADDITIONAL CALCIUM FLUORIDE DURING DISSOLUTION. SINGLE CONTACT

Solvent: 12.5M HNO₃-0.2M CaF₂

Solid to Liquid Ratio: 100 g/l Feed Material: Crushed incinerator ash

Feed Concentration: ~20% plutonium Reflux Period: 2 hours

Calcium Fluoride Addition: Added after one hour of refluxing

Initial CaF ₂ Conc. (M)	Amt of Added CaF ₂ (% of Initial Charge)	Amt of Solid Dissolved (%)	Amt of Pu Rec (%)	Undissolved Residue Conc. (% Pu)
0.2	0	64	84.4	2.9
0.2	0	69	83.3	4.1
0.2	0	67	87.7	1.8
Av.		67	85.1	2.9
0.2	25	71	93.5	1.1
0.2	25	83	90.2	2.7
0.2	25	72	89.3	1.0
Av.		75	91.0	1.9
0.2	50	70	90.8	1.9
0.2	50	70	90.6	2.0
0.2	50	71	91.6	1.6
Av.		70	91.0	1.8
0.2	100	72	94.2	1.2
0.2	100	72	94.0	1.2
0.2	100	74	93.6	1.4
Av.		73	93.9	1.3

In general, the data indicate that adding calcium fluoride during the dissolution is important for obtaining a discardable, undissolved residue, but this addition appears to be of only minor importance for increased solid dissolution and plutonium recovery. Since this addition increases the probability of plutonium fluoride complexing, the value of the addition must be compared to the precautions which must be taken for processing the filtrates through anion exchange for plutonium purification. Aluminum nitrate nonahydrate must be added to the ion column feed solutions to complex all of the available fluoride, thus breaking the cationic plutonium fluoride complex, PuF_2^{+2} ; however, this additional aluminum nitrate also tends to increase the probability of these solutions "salting out" in high nitric acid concentrations.

MULTIPLE CONTACTS

Since a significant amount of plutonium can be recovered using a relatively short reflux period, a series of experiments was performed to determine the effect of multiple

contacts using various reflux periods, various solid to liquid ratios and various solvents. The experiments were divided into five groups as follows:

- (1) four fresh acid-fluoride contacts
- (2) two fresh acid-fluoride contacts
- (3) solid to liquid ratio
 - (a) two fresh acid-fluoride contacts
 - (b) four fresh acid-fluoride contacts
- (4) four contacts and reused acid
- (5) three contacts and reused acid.

Four Fresh Acid-Fluoride Contacts

Tables VI and VII contain the average values calculated for each variable from the four fresh acid-fluoride contact experiments.

TABLE VI

EFFECT OF FOUR FRESH ACID-FLUORIDE CONTACTS AND FOUR REFLUX PERIODS ON PLUTONIUM RECOVERY

Solvent: 12.5M HNO_3 -0.2M CaF_2 for each of four contacts

Solid to Liquid Ratio: 100 g/l for the first contact

Reflux Period (min.)	Feed Material	No. of Runs	Initial Feed Conc. (% Pu)	1st Contact		2nd Contact		3rd Contact		4th Contact		Amt of Solid Dissolved (%)	Undissolved Residue Conc. (% Pu)
				Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)		
15	UIA	3	12.5	7.4	56.2	3.6	87.6	0.9	95.3	0.4	98.2	89	0.9
30	UIA	3	12.1	8.2	62.9	2.8	87.8	0.9	95.4	0.3	98.3	88	0.9
60	UIA	3	17.9	10.3	54.6	6.5	92.1	1.0	97.5	0.3	99.1	90	0.9
120	UIA	3	14.0	10.2	67.0	3.6	92.2	0.8	97.8	0.2	99.5	90	0.4
15	CIA	3	29.3	19.7	62.8	7.3	88.5	1.7	94.7	0.8	97.4	84	4.2
30	CIA	3	39.0	22.5	74.0	6.2	91.0	1.4	95.5	0.9	98.5	88	3.2
60	CIA	3	28.3	22.7	74.9	5.0	92.9	1.3	97.6	0.5	99.2	90	1.0
120	CIA	3	31.9	28.9	82.6	3.8	95.3	0.9	98.6	0.3	99.6	90	0.9
15	IAH	3	16.6	14.3	81.7	1.9	94.2	0.6	98.4	0.2	99.7	77	0.2
30	IAH	3	13.1	11.1	81.0	1.7	93.7	0.6	98.2	0.2	99.7	78	0.1
60	IAH	3	11.3	9.8	84.0	1.3	95.4	0.4	98.8	0.2	99.8	83	0.1
120	IAH	3	13.1	11.7	84.5	1.4	94.7	0.5	98.7	0.2	99.7	78	0.1
15	Swps	3	36.5	26.0	70.4	8.1	92.6	1.7	97.3	0.5	98.8	82	1.6
30	Swps	3	31.5	24.4	75.6	6.1	94.9	1.3	98.7	0.2	99.4	88	1.2
60	Swps	3	33.9	31.7	87.4	3.7	98.2	0.4	99.2	0.2	99.7	96	1.5
120	Swps	3	28.3	28.0	94.2	1.5	99.1	0.2	99.8	0.05	99.9	99	0.8
15	SH	3	26.0	23.2	85.1	2.7	95.5	0.6	97.6	0.3	98.8	78	1.0
30	SH	3	24.5	22.5	85.7	2.4	95.6	0.6	98.1	0.2	99.1	79	0.8
60	SH	3	24.9	22.8	85.7	2.2	96.5	0.5	98.5	0.2	99.3	79	0.6
120	SH	3	28.2	26.6	88.7	2.6	97.8	0.3	99.0	0.1	99.4	80	0.5
15	FS	3	19.7	10.0	49.6	4.4	72.1	2.0	82.1	1.3	88.8	38	3.3
30	FS	3	21.3	11.6	53.7	5.1	77.4	2.1	87.6	1.1	92.7	47	2.4
60	FS	3	20.2	12.8	60.3	4.3	80.8	1.9	90.2	1.1	95.5	55	1.7
120	FS	4	26.5	17.1	57.1	6.9	80.4	3.4	81.7	1.5	96.7	67	1.9

UIA - Uncrushed Incinerator Ash

CIA - Crushed Incinerator Ash

IAH - Incinerator Ash Heel

Swps - Glovebox Floor Sweepings

SH - Glovebox Floor Sweepings Heel

FS - Filter Sludge

TABLE VII

EFFECT OF FOUR FRESH ACID-FLUORIDE CONTACTS AND FOUR REFLUX PERIODS ON PLUTONIUM RECOVERY

Solvent: 12.5M HNO₃-0.2M CaF₂ for each of four contacts

Solid to Liquid Ratio: 100 g/l for the first contact

Reflux Period (min.)	Feed Material	No. of Runs	Initial Feed Conc. (% Pu)	1st Contact		2nd Contact		3rd Contact		4th Contact		Amt of Solid Dissolved (%)	Undissolved Residue Conc. (% Pu)
				Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)		
15	UIA	3	12.5	7.4	56.2	3.6	87.6	0.9	95.3	0.4	98.2	89	0.9
15	CIA	3	29.3	19.7	62.8	7.3	88.5	1.7	94.7	0.8	97.4	84	4.2
15	IAH	3	16.6	14.3	81.7	1.9	94.2	0.6	98.4	0.2	99.7	77	0.2
15	Swps	3	36.5	26.0	70.4	8.1	92.6	1.7	97.3	0.5	98.8	82	1.6
15	SH	3	26.0	23.2	85.1	2.7	95.5	0.6	97.6	0.3	98.8	78	1.0
15	FS	3	19.7	10.0	49.6	4.4	72.1	2.0	82.1	1.3	88.8	38	3.3
Av.			23.9	16.8	67.6	4.7	88.4	1.2	94.2	0.6	97.0	75	1.9
30	UIA	3	12.1	8.2	62.9	2.8	87.8	0.9	95.4	0.3	98.3	88	0.9
30	CIA	3	39.0	22.5	74.0	6.2	91.0	1.4	95.5	0.9	98.5	88	3.2
30	IAH	3	13.1	11.1	81.0	1.7	93.7	0.6	98.2	0.2	99.7	78	0.1
30	Swps	3	31.5	24.4	75.6	6.1	94.9	1.3	98.7	0.2	99.4	88	1.2
30	SH	3	24.5	22.5	85.7	2.4	95.6	0.6	98.1	0.2	99.1	79	0.8
30	FS	3	21.3	11.6	53.7	5.1	77.4	2.1	87.6	1.1	92.7	47	2.4
Av.			23.6	16.7	72.2	4.1	90.1	1.2	95.6	0.5	98.0	78	1.4
60	UIA	3	17.9	10.3	54.6	6.5	92.1	1.0	97.5	0.3	99.1	90	0.9
60	CIA	3	28.3	22.7	74.9	5.0	92.9	1.3	97.6	0.5	99.2	90	1.8
60	IAH	3	11.3	9.8	84.0	1.3	95.4	0.4	98.8	0.2	99.8	83	0.1
60	Swps	3	33.9	31.7	87.4	3.7	98.2	0.4	99.2	0.2	99.7	96	1.5
60	SH	3	24.9	22.8	85.7	2.2	96.5	0.5	98.5	0.2	99.3	79	0.6
60	FS	3	20.2	12.8	60.3	4.3	80.8	1.9	90.2	1.1	95.5	55	1.7
Av.			22.8	18.4	74.5	3.8	92.6	0.9	97.0	0.4	98.8	82	1.1
120	UIA	3	14.0	10.2	67.0	3.6	92.2	0.8	97.8	0.2	99.5	90	0.4
120	CIA	3	31.9	28.9	82.6	3.8	95.3	0.9	98.6	0.3	99.6	90	0.9
120	IAH	3	13.1	11.7	84.5	1.4	94.7	0.5	98.7	0.2	99.7	78	0.1
120	Swps	3	28.3	28.0	94.2	1.5	99.1	0.2	99.8	0.05	99.9	99	0.8
120	SH	3	28.2	26.6	88.7	2.6	97.8	0.3	99.0	0.1	99.4	80	0.5
120	FS	4	26.5	17.1	57.1	6.9	80.4	3.4	81.7	1.5	96.7	67	1.9
Av.			23.7	20.4	79.0	3.3	93.3	1.0	95.9	0.4	99.1	84	0.8

UIA - Uncrushed Incinerator Ash
CIA - Crushed Incinerator Ash
IAH - Incinerator Ash Heel
Swps - Glovebox Floor Sweepings
SH - Glovebox Floor Sweepings Heel
FS - Filter Sludge

In Table VI the data are tabulated by type of feed material and as a function of increasing reflux period. From these data the effectiveness of longer reflux periods for a given type of material can be determined. When making any comparison, care should be taken to properly consider the initial feed concentration for a given reflux period. Another feature of Table VI is that for a given reflux period the variation in dissolution due to material type can easily be compared.

For uncrushed incinerator ash an eightfold increase in the length of the reflux period increased the plutonium recovery after the first contact by 19 percent, after the second contact by 5 percent, after the third contact by 3 percent and after the fourth contact by 1 percent.

The corresponding values for crushed incinerator ash are 31 percent, 8 percent, 4 percent and 2 percent. For incinerator ash heel the values are 3 percent, <1 percent, <1 percent, and <1 percent respectively. The same eightfold

increase resulted in a 34 percent increase in plutonium recovery after the first contact, a 7 percent increase after the second contact, a 2 percent increase after the third contact and a 1 percent increase after the fourth contact for glovebox floor sweepings.

A short reflux period (e.g., 15 minutes) is as effective as a long reflux period (e.g., 2 hours) if a multiple contact system is used for the dissolution. There is a significant difference between short and long reflux periods after the first contact, but this difference has been reduced after the third and fourth contact.

In Table VII the data are tabulated as a function of reflux period and all material types are considered simultaneously. The average values which had been previously calculated are again averaged. The nature of plutonium-bearing residues varies; consequently, it is valid to use these averaged values for predicting dissolution trends.

Since the averaged initial feed concentration is approximately the same (-23-24 percent Pu), a comparison can be made between reflux periods. The cumulative percent plutonium recovered for each contact shows that after the first contact, an eightfold increase in the length of the reflux period results in a 17 percent increase in plutonium recovery. However, after two contacts the eightfold increase only increases plutonium recovery by 6 percent. And, for the third and fourth contacts the increase is only 2 percent. This means that with three or more contacts the value of a longer reflux period is no more effective in dissolving plutonium-bearing residues than the 15 minute reflux period. This is illustrated graphically in Figure 4. Here the cumulative percent plutonium recovered is plotted against the cumulative reflux period.

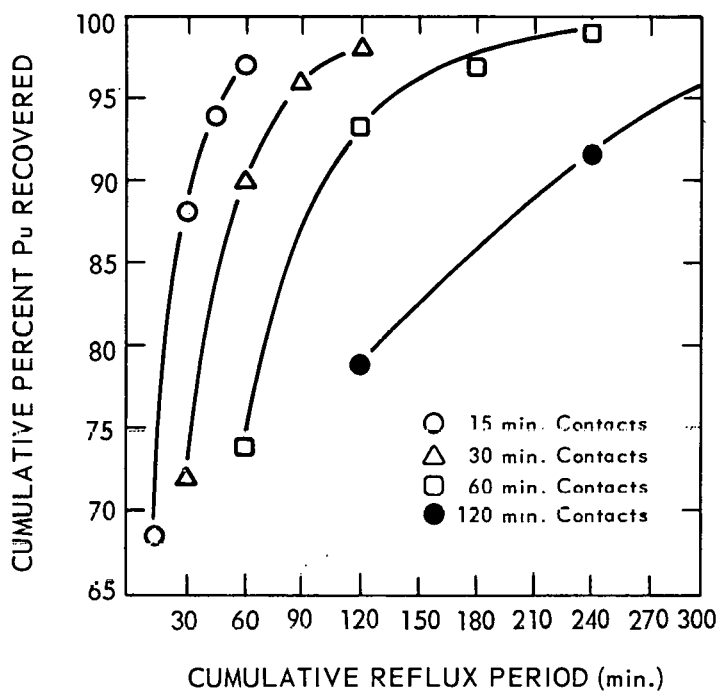


Figure 4. EFFECT OF FOUR FRESH ACID-FLUORIDE CONTACTS AND FOUR REFLUX PERIODS ON PLUTONIUM RECOVERY. Solvent: $12.5M$ HNO_3 - $0.2M$ CaF_2 for each of four contacts. Solid to liquid ratio: 100 g/l for the first contact.

Another consideration which must be given to the data in Table VII is the averaged values for the amount of solid dissolved and to the plutonium concentration in the undissolved residue. If the discard level for these residues is less than one percent plutonium, this latter value becomes very important. The data show that only four contacts of 2 hours each, result in a discardable undissolved residue (heel). However, if the criteria for discard is to be based on both the plutonium concentration in the heel and the cumulative percent plutonium recovered then once again four 15 minute contacts can be considered as effective as

four 2 hour contacts. Longer contacts result in a larger percentage of dissolved solid. This may not always be desirable.

Two Fresh Acid-Fluoride Contacts

Since four fresh acid contacts would produce four times the volume of filtrate presently handled by the plutonium recovery area, two fresh acid-fluoride contacts were evaluated. The data from the previous experiments (Table VII) had shown that two contacts resulted in 88-93 percent of the plutonium being recovered depending on the length of the reflux period. With this in mind, seven two-contact experiments were designed where the reflux period was varied for each contact and calcium fluoride was added during some of the longer reflux periods.

The following combinations were evaluated:

1. The first contact was with fresh $12.5M$ HNO_3 - $0.2M$ CaF_2 for one hour of refluxing and the second contact with fresh $12.5M$ HNO_3 - $0.2M$ CaF_2 for 3 hours of refluxing.
2. The same as condition (1) except that an additional amount of calcium fluoride ($0.2M$) was added after the first and second hour of the reflux period.
3. The same as condition (2) except that the additional amount of calcium fluoride added was $0.1M$.
4. The same as condition (1) except that the first contact was $1/2$ hour and the second contact was for 2 hours.
5. The same as condition (1) except that the first contact was for one hour and the second contact was for 1- $1/2$ hours.
6. The same as condition (5) except that an additional amount of calcium fluoride ($0.1M$) was added halfway through the second reflux period.
7. The same as condition (1) except that the contacts were each one hour.

All of the combinations (see Table VIII) resulted in average plutonium recovery values of 96.0-97.5 percent; however, only condition (2) resulted in a discardable undissolved residue (<1 percent Pu). The amount of solid dissolved varied from 73 percent to 82 percent.

These double contacts will only double the present filtrate volumes. Since filtering capacity is an important consideration in determining the value of a particular scheme for dissolution, two contacts which recover >95 percent of the plutonium while only doubling the filtering volume are extremely attractive.

TABLE VIII

EFFECT OF TWO FRESH ACID-FLUORIDE CONTACTS ON PLUTONIUM RECOVERY

Solvent: 12.5M HNO₃-0.2M CaF₂ for each of two contacts Solid to Liquid Ratio: 100 g/l for the first contact
 Feed Material: Crushed incinerator ash, incinerator ash heel, glovebox floor sweepings, and glovebox floor sweepings heel

Experimental Conditions	No. of Runs	Initial Feed Conc. (% Pu)	1st Contact		2nd Contact		Amt of Solid Dissolved (%)	Undissolved Residue Conc. (% Pu)
			Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)		
(1)	4	27.6	22.5	78.4	5.7	97.5	82	1.1
(2)	4	28.1	22.8	77.1	6.1	97.0	73	0.8
(3)	4	28.2	22.0	74.7	7.1	97.3	80	3.0
(4)	4	25.4	19.4	75.3	6.1	96.3	80	1.8
(5)	4	27.9	22.1	75.8	6.0	96.1	77	2.0
(6)	4	28.0	22.2	76.0	6.2	96.5	76	1.8
(7)	4	24.6	19.7	78.4	5.0	96.0	78	1.8

Conditions (1) - Two Contacts:

First contact with 12.5M HNO₃-0.2M CaF₂ for one hour.
 Second contact with 12.5M HNO₃-0.2M CaF₂ for 3 hours.

Conditions (2) - Two Contacts:

Same as conditions (1) except that an additional amount of CaF₂ (0.2M) was added after the first and second hour of reflux.

Conditions (3) - Two Contacts:

Same as conditions (2) except that the additional amount of CaF₂ added was 0.1M.

Conditions (4) - Two Contacts:

Same as conditions (1) except that the first contact was for 1/2-hour and the second contact was for 2 hours.

Conditions (5) - Two Contacts:

Same as conditions (1) except that the first contact was for one hour and the second contact was for 1-1/2 hours.

Conditions (6) - Two Contacts:

Same as conditions (5) except that an additional amount of CaF₂ (0.1M) was added halfway through the second reflux period.

Conditions (7) - Two Contacts:

Same as conditions (1) except that the contacts were each one hour.

Solid to Liquid Ratio and Multiple Contacts

As a result of the favorable outcome of the multiple contact experiments the effect of increasing the solid to liquid ratio was determined. It was thought that with a higher initial solid to liquid ratio the net effect after repeated contacts would be the same as described previously. The results for two contacts (one hour each) with solid to liquid

ratios of 100, 150 and 200 g/l are tabulated in Table IX. As the ratio was doubled the percent of plutonium recovered decreased by 4 percent which is not considered significant. A higher solid to liquid ratio can be used when a multiple contact system is used for dissolution. Four contacts (one hour each) with increasing solid to liquid ratios were also still effective in recovering plutonium. The data for these experiments are in Table X. The incinerator ash heel used

TABLE IX

THE EFFECT OF SOLID TO LIQUID RATIO UPON THE AMOUNT OF SOLID DISSOLVED, THE AMOUNT OF PLUTONIUM RECOVERED AND THE AMOUNT OF PLUTONIUM IN THE UNDISSOLVED RESIDUE USING TWO ONE-HOUR FRESH ACID-FLUORIDE CONTACTS

Solvent: 12.5M HNO₃-0.2M CaF₂ for each of two contacts Reflux Period: One hour for each contact

Feed Material	Solid to Liquid Ratio (g/l)	Initial Feed Conc. (% Pu)	1st Contact		2nd Contact		Amt of Solid Dissolved (%)	Undissolved Residue Conc. (% Pu)
			Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)		
CIA	100	30.4	22.7	69.2	6.3	90.0	80	7.1
IAH	100	5.8	3.1	51.2	1.0	68.9	35	1.9
Av.		18.1	12.9	60.2	3.6	79.4	58	4.5
CIA	150	30.5	33.3	65.9	10.6	89.7	77	5.6
IAH	150	5.1	4.4	54.2	1.1	68.5	26	2.1
v.		17.8	18.8	60.0	5.8	79.1	52	3.8
CIA	200	29.6	38.1	57.8	16.2	85.7	68	10.4
IAH	200	5.7	6.0	47.9	2.2	67.4	24	2.3
Av.		17.6	22.0	52.8	9.2	76.6	46	6.4

CIA - Crushed Incinerator Ash
 IAH - Incinerator Ash Heel

TABLE X

THE EFFECT OF SOLID TO LIQUID RATIO UPON THE AMOUNT OF SOLID DISSOLVED, THE AMOUNT OF PLUTONIUM RECOVERED AND THE AMOUNT OF PLUTONIUM IN THE UNDISSOLVED RESIDUE USING FOUR ONE-HOUR FRESH ACID-FLUORIDE CONTACTS

Solvent: 12.5M HNO₃-0.2M CaF₂ for each of four contacts Reflux Period: One hour for each contact

Feed Material	Solid to Liquid Ratio (g/l)	Initial Feed Conc. (% Pu)	1st Contact		2nd Contact		3rd Contact		4th Contact		Amt of Solid Dissolved (%)	Undissolved Residue Conc. (% Pu)
			Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)		
CIA	100	29.3	22.6	72.8	5.4	91.3	1.8	97.5	0.4	99.0	90	1.6
IAH	100	5.3	3.2	57.1	0.8	72.6	0.4	81.2	0.4	88.3	70	1.7
Av.		17.3	12.9	65.0	3.1	82.0	1.1	89.4	0.4	93.6	80	1.6
CIA	150	24.0	26.8	69.0	7.4	89.6	2.2	95.6	0.8	98.0	85	1.9
IAH	150	6.0	4.6	47.5	1.5	64.3	0.7	72.3	0.6	78.3	38	2.0
Av.		15.0	15.7	58.2	4.4	77.0	1.4	84.0	0.7	88.2	62	2.0
CIA	200	9.1	13.3	67.5	3.4	86.2	1.1	92.1	0.5	94.7	78	2.0
IAH	200	6.0	6.1	47.0	2.0	63.7	1.0	71.7	0.6	71.1	33	2.0
Av.		7.5	9.7	57.2	2.7	75.0	1.0	81.9	0.6	82.9	56	2.0

CIA - Crushed Incinerator Ash
IAH - Incinerator Ash Heel

for the 150 g/l and 200 g/l ratios was considerably more difficult to dissolve than any ash heel that had been previously used. The amount of solid dissolved after four contacts was the lowest observed. Also the initial feed concentration of the crushed incinerator ash used for the 200 g/l ratio was considerably lower than for the other two ratios. An increased ratio has a rather negligible effect upon the dissolution of these residues particularly when as many as four contacts are used.

Four Contacts and Reused Acid

In an attempt to determine whether the filtrate from a previous dissolution could be used as the acid for another contact, we designed a set of experiments using four contacts except that a once-used acid (i.e., the filtrate from a fresh acid contact) was used for the third contact and a twice-used acid was the solvent for the fourth contact. The results are presented in Table XI. The once-used acids had

TABLE XI

THE EFFECT OF FOUR CONTACTS UPON THE AMOUNT OF SOLID DISSOLVED, THE AMOUNT OF PLUTONIUM RECOVERED, AND THE AMOUNT OF PLUTONIUM IN THE UNDISSOLVED RESIDUE, BY USING A ONCE- AND TWICE-USED [i.e., Pu(NO₃)₄, HNO₃, CaF₂] ACID FOR THE THIRD AND FOURTH CONTACTS RESPECTIVELY

Solvent: 12.5M HNO₃-0.2M CaF₂ for the first two contacts Solid to Liquid Ratio: 100 g/l for the first of four contacts

Feed Material	Exp. Cond.	Initial Feed Conc. (% Pu)	1st Contact		2nd Contact		3rd Contact		4th Contact		Amt of Solid Dissolved (%)	Undissolved Residue Conc. (% Pu)	Used Acid Conc. (g/l)	
			Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)			Once-Used Acid	Twice-Used Acid
CIA	(1)	28.6	23.0	73.3	4.8	94.3	6.8	96.2	7.4	97.0	78	2.5	6.3	7.2
IAH	(1)	12.8	11.7	85.3	1.5	95.4	1.8	98.3	2.0	99.2	72	0.2	1.4	1.9
Swps	(1)	30.8	27.2	84.5	5.1	97.5	4.6	98.7	5.3	99.2	81	1.2	4.2	5.1
SH	(1)	35.8	29.6	77.8	7.6	94.5	8.6	97.9	8.5	98.7	61	0.9	7.4	8.2
Av.		27.0	22.9	80.2	4.8	95.4	5.4	97.8	5.8	98.5	73	1.2	4.8	5.6
CIA	(2)	26.5	21.4	73.5	4.0	96.1	6.3	96.8	6.8	97.1	83	2.4	6.1	6.7
IAH	(2)	13.1	11.6	85.0	1.4	94.5	1.9	97.4	2.3	98.5	74	0.6	1.5	2.2
Swps	(2)	32.1	30.7	84.4	5.1	96.9	5.7	98.8	6.7	99.6	90	0.8	5.1	6.4
SH	(2)	36.7	33.8	81.5	6.7	97.9	7.4	98.5	8.2	99.2	73	0.8	7.2	8.0
Av.		27.1	24.4	81.1	4.3	96.4	5.3	97.9	6.0	98.6	80	1.2	5.0	5.8

CIA - Crushed Incinerator Ash
IAH - Incinerator Ash Heel
Swps - Glovebox Floor Sweepings
SH - Glovebox Floor Sweepings Heel

Conditions (1) - Four One-Hour Contacts:

First contact with 12.5M HNO₃-0.2M CaF₂
Second contact with 12.5M HNO₃-0.2M CaF₂
Third contact with once-used acid (~12M H⁺) and 0.2M CaF₂
Fourth contact with twice-used acid (~12M H⁺) and 0.2M CaF₂

Conditions (2) - Same as above except the CaF₂ added for the third and fourth contacts was only 0.1M.

a plutonium concentration of 1.4-7.4 g/l prior to reuse. The once-used acids had a plutonium concentration of 1.9-8.2

g/l prior to reuse. The hydrogen ion concentration of the used acids was 11-12M. The averaged results indicated that a four contact system could be used.

The plutonium recovery averaged 98 percent with solid dissolution varying from 73-80 percent. The plutonium concentration in the undissolved residue was 1.2 percent. Prior to the third and fourth contact (i.e., the ones with used acids) the plutonium recovery was 95-96 percent. It is the additional 2-3 percent that becomes very important if the criteria for discard is the cumulative percent plutonium recovered as well as the plutonium concentration in the undissolved residue.

Three Contacts and Reused Acid

The data for three contacts and reused acid are tabulated in Table XII. The used acid had a plutonium concentration from 0.3-1.2 g/l for conditions (2). Based on the averages calculated from the data 73-79 percent of the plutonium was recovered after the first contact, 96-98 percent after the second contact and >99 percent after the third contact. The plutonium concentration in the undissolved residue was less than one percent. The solid dissolution varied from 81-84 percent.

TABLE XII

THE EFFECT OF THREE CONTACTS UPON THE AMOUNT OF SOLID DISSOLVED, THE AMOUNT OF PLUTONIUM RECOVERED AND THE AMOUNT OF PLUTONIUM IN THE UNDISSOLVED RESIDUE BY USING A ONCE-USED [i.e., $\text{Pu}(\text{NO}_3)_4$, HNO_3 , CaF_2] ACID FOR THE FIRST CONTACT

Solvent: 12.5M HNO_3 -0.2M CaF_2 for the second and third contacts Solid to Liquid Ratio: 100 g/l for the first contact

Feed Material	Exp. Cond.	Initial Feed Conc. (% Pu)	1st Contact		2nd Contact		3rd Contact		Amt of Solid Dissolved (%)	Undissolved Residue Conc. (% Pu)	Once-Used Acid Conc. (g Pu/l)
			Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)			
CIA	(1)	26.6	21.0	62.9	8.5	94.3	1.2	98.8	87	1.4	0.9
IAH	(1)	17.8	16.3	84.9	2.5	98.0	0.3	99.7	78	0.2	1.2
Swps	(1)	32.5	31.4	85.6	4.5	99.1	0.3	99.9	86	<0.1	0.3
SH	(1)	35.9	34.0	80.9	6.4	98.6	0.4	99.7	74	0.3	0.3
Av.		28.2	25.7	78.6	5.5	97.5	0.6	99.5	81	0.5	0.7
CIA	(2)	27.0	19.1	59.9	8.5	91.4	1.5	97.4	84	2.4	0.2
IAH	(2)	14.3	13.8	74.9	3.0	95.5	0.6	99.9	75	<0.1	1.5
Swps	(2)	29.9	30.3	86.7	3.7	98.9	0.3	99.9	96	0.2	0.6
SH	(2)	45.0	36.5	70.6	12.7	99.0	0.2	99.6	81	0.6	0.3
Av.		29.0	24.9	73.0	7.0	96.2	0.6	99.2	84	0.8	0.6

CIA - Crushed Incinerator Ash
IAH - Incinerator Ash Heel
Swps - Glovebox Floor Sweepings
SH - Glovebox Floor Sweepings Heel

Conditions (1) - Three Contacts:

First contact with once-used acid (~12M H^+) and 0.2M CaF_2 for one hour.

Second contact with 12.5M HNO_3 -0.2M CaF_2 for 2 hours and an additional amount of CaF_2 (0.2M) was added after the first hour of reflux.

Third contact with 12.5M HNO_3 -0.2M CaF_2 for one hour.

Conditions (2) - Same as above except that in the second contact only 0.1M CaF_2 was added after the first hour of reflux.

APPENDIX —
EXPERIMENTAL DATA

TABLE IA

EFFECT OF LENGTH OF REFLUX PERIOD ON DISSOLUTION OF RESIDUES. SINGLE CONTACT

Solvent: 12.5M HNO₃-0.2M CaF₂ Solid to Liquid Ratio: 100 g/l

Reflux Period (min.)	Feed Material	Initial Feed Conc. (% Pu)	Filtrate Conc. (g Pu/l)	Amt of Solid Dissolved (%)	Amt of Pu Rec (%)	Undissolved Residue Conc. (% Pu)
15	IAH	31.8	20.9	29	65.7	13.8
15	IAH	15.0	3.0	59	20.0	17.9
15	SH	10.2	10.1	62	99.0	0.2
15	SH	19.8	16.5	62	83.3	5.9
15	SH	27.7	20.5	44	74.0	11.5
Av.		20.9	14.2	51	68.4	9.9
30	IAH	33.3	23.9	31	71.7	13.2
30	IAH	28.2	21.0	36	74.4	10.6
30	IAH	34.1	26.0	48	76.2	14.2
30	IAH	27.7	20.6	36	74.3	11.5
30	IAH	13.7	3.6	65	26.3	21.0
30	IAH	12.3	7.7	79	62.6	18.4
30	IAH	13.5	4.4	62	32.6	20.8
30	SH	14.2	12.1	64	85.2	3.0
30	SH	14.1	12.1	76	85.8	4.0
30	SH	14.7	11.4	59	77.6	5.5
30	SH	27.3	23.5	52	86.1	6.3
30	SH	27.2	20.6	50	75.7	10.5
30	SH	32.4	23.4	44	72.2	14.1
Av.		22.5	16.2	54	69.3	11.7
45	IAH	37.9	30.1	38	79.4	12.2
45	IAH	12.4	6.7	69	54.0	15.9
45	SH	15.0	13.9	68	92.6	1.9
45	SH	27.2	23.0	48	84.6	7.4
Av.		23.1	18.4	56	77.6	9.4
60	IAH	31.5	25.3	37	80.3	9.7
60	IAH	30.4	24.9	42	81.8	9.5
60	IAH	14.9	11.9	75	79.9	10.1
60	IAH	14.1	13.8	80	97.9	0.9
60	SH	13.6	11.6	62	85.3	4.7
60	SH	13.4	12.2	69	91.0	2.6
60	SH	43.9	38.2	57	87.0	11.8
60	SH	35.2	25.3	48	71.8	15.8
Av.		24.6	20.4	59	84.4	8.1
90	IAH	33.6	28.6	46	85.2	8.4
90	IAH	29.4	25.0	50	85.0	8.2
90	SH	29.3	25.5	37	87.0	6.2
90	SH	24.8	20.7	48	83.5	6.2
90	M	24.1	22.5	38	93.4	2.6
90	M	22.2	20.6	82	92.8	3.7
90	FS	43.5	39.3	85	90.3	20.0
Av.		29.6	26.0	55	88.2	7.9
120	IAH	29.9	22.0	42	73.6	13.0
120	SH	24.9	19.0	30	76.3	8.4
120	M	26.0	24.5	83	94.2	3.9
120	FS	39.8	37.7	92	94.7	14.0
Av.		30.2	25.8	62	84.7	9.8
150	IAH	30.2	22.6	37	74.8	10.8
150	SH	29.2	25.1	48	86.0	6.8
150	M	22.9	21.4	84	93.3	4.0
150	FS	47.0	45.1	88	96.0	9.1
Av.		32.3	28.6	64	87.5	7.7

Incinerator Ash Heel

Glovebox Floor Sweepings Heel

FS - Filter Sludge

M - Mixture of the Above

TABLE IIA
EFFECT OF LENGTH OF REFLUX PERIOD ON DISSOLUTION OF RESIDUES. SINGLE CONTACT.
CONCENTRATED NITRIC ACID (-15.6M)

Solvent: 15.6M HNO₃-0.2M CaF₂ Solid to Liquid Ratio: 100 g/l

Reflux Period (min.)	Feed Material	Initial Feed Conc. (% Pu)	Filtrate Conc. (g Pu/l)	Amt of Solid Dissolved (%)	Amt of Pu Rec (%)	Undissolved Residue Conc. (% Pu)
15	IAH	39.3	29.3	28	74.4	13.5
15	IAH	19.9	5.7	36	28.6	14.3
15	SH	18.6	11.6	52	62.4	11.5
15	SH	28.0	23.3	57	83.2	8.8
Av.		26.4	17.5	43	62.2	12.0
30	IAH	26.1	16.0	25	61.3	12.2
30	IAH	15.9	6.7	68	42.1	17.0
30	SH	13.3	9.7	60	72.9	8.0
30	SH	28.3	22.0	41	77.7	10.0
Av.		20.9	13.6	48	63.5	11.8
45	IAH	32.9	22.4	14	68.1	11.2
45	IAH	11.9	10.8	80	90.8	4.1
45	SH	17.0	9.6	60	56.5	16.1
45	SH	25.1	23.8	42	94.8	21.2
Av.		21.7	16.6	49	77.6	13.2
60	IAH	34.8	24.2	6	69.5	10.8
60	IAH	14.5	10.6	86	73.1	22.1
60	SH	15.6	10.2	64	65.4	9.5
60	SH	29.3	22.9	45	78.2	11.0
60	IAH	11.0	10.7	88	97.3	1.5
60	IAH	29.3	18.2	42	62.1	17.6
60	SH	15.0	9.6	63	64.0	8.2
60	SH	31.7	25.3	48	79.8	10.4
Av.		22.6	16.5	55	73.7	11.4
120	M	22.9	21.3	86	93.0	5.9
120	FS	9.3	3.6	61	39.2	8.2
120	IAH	23.8	19.6	32	82.3	5.4
Av.		18.7	14.8	60	71.5	6.5

IAH - Incinerator Ash Heel
SH - Glovebox Floor Sweepings Heel
FS - Filter Sludge
M - Mixture of the Above

TABLE IIIA
THE EFFECT OF SOLID TO LIQUID RATIO UPON THE AMOUNT OF SOLID DISSOLVED, THE AMOUNT OF PLUTONIUM RECOVERED AND THE AMOUNT OF PLUTONIUM IN THE UNDISSOLVED RESIDUE

Solvent: 12.5M HNO₃-0.2M CaF₂ Reflux Period: One hour

Feed Material	Solid to Liquid Ratio (g/l)	Initial Feed Conc. (% Pu)	Filtrate Conc. (g Pu/l)	Amt of Solid Dissolved (%)	Amt of Pu Rec (%)	Undissolved Residue Conc. (% Pu)
CIA	25	29.0	7.0	89	96.4	4.5
CIA	50	27.1	11.6	70	85.6	6.5
CIA	100	26.5	20.1	58	76.1	15.1
CIA	150	25.3	23.4	48	62.1	18.6
CIA	200	30.0	37.3	52	62.9	23.6
CIA	250	27.3	35.9	45	52.9	23.6
IAH	25	19.1	4.7	67	97.9	0.7
IAH	50	15.2	7.0	69	91.3	2.7
IAH	100	13.7	11.7	55	85.4	4.4
IAH	150	14.1	17.4	51	82.0	5.2
IAH	200	13.6	22.9	50	84.3	4.3
IAH	250	15.1	27.6	39	72.9	6.8
Swps	25	32.6	8.1	86	99.3	1.1
Swps	50	37.5	18.5	86	98.5	2.6
Swps	100	26.2	24.3	86	92.5	9.3
Swps	150	26.7	32.9	84	82.1	19.4
Swps	200	25.1	40.6	83	80.9	18.1
Swps	250	24.6	48.0	84	77.9	21.7
SH	25	38.2	9.4	74	98.7	1.5
SH	50	35.6	16.4	69	91.9	7.1
SH	100	38.4	24.2	60	63.0	25.2
SH	150	38.7	36.5	53	62.7	30.4
SH	200	-	-	-	-	-
SH	250	-	-	-	-	-

CIA - Crushed Incinerator Ash
IAH - Incinerator Ash Heel
Swps - Glovebox Floor Sweepings
SH - Glovebox Floor Sweepings Heel

TABLE IVA

THE EFFECT OF FOUR FRESH ACID-FLUORIDE CONTACTS UPON THE AMOUNT OF SOLID DISSOLVED, THE AMOUNT OF PLUTONIUM RECOVERED AND THE AMOUNT OF PLUTONIUM IN THE UNDISSOLVED RESIDUE

Solvent: 12.5M HNO₃-0.2M CaF₂ for each of four contacts Solid to Liquid Ratio: 100 g/l for the first contact
Feed Material: Uncrushed incinerator ash

Reflux Period (min.)	Initial Feed Conc. (% Pu)	1st Contact		2nd Contact		3rd Contact		4th Contact		Amt of Solid Dissolved (%)	Undissolved Residue Conc. (% Pu)
		Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)		
15	12.8	8.1	60.1	3.5	88.2	0.9	95.4	0.4	98.3	89	0.8
15	12.6	7.3	55.3	3.9	87.4	1.0	95.4	0.4	98.2	88	0.9
15	12.0	6.9	53.2	3.3	86.8	0.8	95.2	0.4	98.2	89	1.1
Av.	12.5	7.4	56.2	3.6	87.6	0.9	95.3	0.4	98.2	89	0.9
30	11.2	6.8	57.5	3.0	85.6	1.0	94.2	0.4	97.8	88	1.0
30	10.8	6.9	59.1	3.0	87.0	0.9	95.1	0.3	98.4	88	1.0
30	14.2	10.9	72.1	2.6	90.7	0.9	96.9	0.2	98.6	89	0.8
Av.	12.1	8.2	62.9	2.8	87.8	0.9	95.4	0.3	98.3	88	0.9
60	19.4	11.3	55.6	6.8	91.6	1.1	97.1	0.3	98.9	90	1.0
60	18.0	10.3	54.1	6.9	93.7	0.8	98.0	0.2	99.2	90	0.8
60	16.4	9.3	54.2	5.9	90.9	1.1	97.3	0.3	99.1	90	0.9
Av.	17.9	10.3	54.6	6.5	92.1	1.0	97.5	0.3	99.1	90	0.9
120	14.4	10.3	65.7	3.8	91.8	0.9	98.1	0.2	99.6	89	0.4
120	13.4	9.9	67.0	3.5	93.1	0.6	97.4	0.3	99.6	90	0.4
120	14.2	10.3	68.2	3.4	91.8	0.8	97.9	0.2	99.4	90	0.4
Av.	14.0	10.2	67.0	3.6	92.2	0.8	97.8	0.2	99.5	90	0.4

TABLE VA

THE EFFECT OF FOUR FRESH ACID-FLUORIDE CONTACTS UPON THE AMOUNT OF SOLID DISSOLVED, THE AMOUNT OF PLUTONIUM RECOVERED AND THE AMOUNT OF PLUTONIUM IN THE UNDISSOLVED RESIDUE

Solvent: 12.5M HNO₃-0.2M CaF₂ for each of four contacts Solid to Liquid Ratio: 100 g/l for the first contact
Feed Material: Crushed incinerator ash

Reflux Period (min.)	Initial Feed Conc. (% Pu)	1st Contact		2nd Contact		3rd Contact		4th Contact		Amt of Solid Dissolved (%)	Undissolved Residue Conc. (% Pu)
		Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)		
15	29.2	19.3	59.7	7.8	87.3	2.0	94.5	0.9	97.5	85	4.1
15	29.6	20.7	66.4	6.8	90.1	1.4	94.9	0.7	97.2	84	4.7
15	29.2	19.1	62.3	7.2	88.2	1.8	94.8	0.8	97.6	84	3.8
Av.	29.3	19.7	62.8	7.3	88.5	1.7	94.7	0.8	97.4	84	4.2
30	31.4	23.2	70.5	6.5	91.7	1.5	96.7	0.7	98.8	88	2.8
30	31.0	23.2	71.7	6.2	91.9	1.3	96.1	0.7	98.3	88	3.8
30	29.4	21.0	69.1	6.0	89.3	1.3	93.8	1.4	98.5	87	2.9
Av.	30.9	22.5	70.4	6.2	91.0	1.4	95.5	0.9	98.5	88	3.2
60	28.8	22.3	73.0	5.4	91.9	1.5	97.2	0.6	99.1	89	1.9
60	28.0	23.0	75.8	4.9	93.6	1.2	97.9	0.4	99.3	90	1.7
	28.0	22.9	75.9	4.8	93.2	1.3	97.8	0.4	99.2	90	1.7
	28.3	22.7	74.9	5.0	92.9	1.3	97.6	0.5	99.2	90	1.8
120	27.6	23.6	78.6	4.5	94.5	1.0	98.1	0.3	99.3	90	0.9
120	22.8	20.0	80.4	3.1	94.2	1.0	98.3	0.3	99.7	88	0.2
120	45.2	43.2	88.7	3.9	97.3	0.9	99.3	0.2	99.7	92	1.5
Av.	31.9	28.9	82.6	3.8	95.3	0.9	98.6	0.3	99.6	90	0.9

TABLE VIA

THE EFFECT OF FOUR FRESH ACID-FLUORIDE CONTACTS UPON THE AMOUNT OF SOLID DISSOLVED, THE AMOUNT OF PLUTONIUM RECOVERED AND THE AMOUNT OF PLUTONIUM IN THE UNDISSOLVED RESIDUE

Solvent: 12.5M HNO₃-0.2M CaF₂ for each of four contacts Solid to Liquid Ratio: 100 g/l for the first contact
Feed Material: Incinerator ash heel

Reflux Period (min.)	Initial Feed Conc. (% Pu)	1st Contact		2nd Contact		3rd Contact		4th Contact		Amt of Solid Dissolved (%)	Undissolved Residue Conc. (% Pu)
		Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)		
15	22.0	20.5	89.0	1.5	96.3	0.6	99.0	0.2	99.9	78	0.1
15	14.6	12.1	79.2	2.1	94.3	0.6	98.4	0.2	99.7	77	0.1
15	13.2	10.4	76.8	2.0	91.9	0.7	97.9	0.3	99.4	75	0.2
Av.	16.6	14.3	81.7	1.9	94.2	0.6	98.4	0.2	99.7	77	0.2
30	12.8	10.8	81.1	1.7	93.6	0.6	98.3	0.2	99.8	79	0.1
30	13.6	11.4	79.2	1.8	93.8	0.7	98.2	0.2	99.7	77	0.1
30	13.0	11.2	82.8	1.5	93.6	0.6	98.2	0.2	99.7	77	0.1
Av.	13.1	11.1	81.0	1.7	93.7	0.6	98.2	0.3	99.7	78	0.1
60	13.4	11.6	84.9	1.5	95.4	0.5	98.4	0.2	99.9	78	0.1
60	8.0	7.0	84.9	0.9	97.3	0.2	99.8	0.1	99.8	94	0.1
60	12.4	10.8	82.1	1.4	93.4	0.5	98.2	0.2	99.8	77	0.1
Av.	11.3	9.8	84.0	1.3	95.4	0.4	98.8	0.2	99.8	83	0.1
120	13.0	11.5	82.7	1.5	93.4	0.6	98.0	0.2	99.5	77	0.2
120	13.2	11.8	84.7	1.4	95.3	0.6	99.8	0.2	99.8	78	0.1
120	13.2	11.8	86.2	1.3	95.3	0.4	98.3	0.1	99.8	79	0.1
Av.	13.1	11.7	84.5	1.4	94.7	0.5	98.7	0.2	99.7	78	0.1

TABLE VIIA

THE EFFECT OF FOUR FRESH ACID-FLUORIDE CONTACTS UPON THE AMOUNT OF SOLID DISSOLVED, THE AMOUNT OF PLUTONIUM RECOVERED AND THE AMOUNT OF PLUTONIUM IN THE UNDISSOLVED RESIDUE

Solvent: 12.5M HNO₃-0.2M CaF₂ for each of four contacts Solid to Liquid Ratio: 100 g/l for the first contact
Feed Material: Glovebox floor sweepings

Reflux Period (min.)	Initial Feed Conc. (% Pu)	1st Contact		2nd Contact		3rd Contact		4th Contact		Amt of Solid Dissolved (%)	Undissolved Residue Conc. (% Pu)
		Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)		
15	35.0	26.8	76.0	7.0	96.0	0.5	97.4	0.4	98.5	85	2.2
15	39.2	26.8	67.9	8.6	89.8	2.8	96.9	0.7	99.0	80	1.6
15	35.4	24.3	67.2	8.6	93.1	1.9	97.7	0.5	98.9	81	1.1
Av.	36.3	26.0	70.4	8.1	92.6	1.7	97.3	0.5	98.8	82	1.6
30	33.8	26.2	77.5	6.3	95.8	1.0	98.8	0.3	99.4	88	1.4
30	28.0	21.9	75.7	5.8	95.7	0.9	98.6	0.2	99.3	87	1.3
30	32.6	25.0	73.6	6.3	93.3	1.9	98.8	0.2	99.4	90	1.2
Av.	31.5	24.4	75.6	6.1	94.9	1.3	98.7	0.2	99.4	88	1.2
60	33.4	34.2	86.8	3.7	97.6	0.4	98.8	0.2	99.4	94	1.6
60	35.4	31.0	86.8	4.1	98.5	0.4	99.6	0.1	99.8	96	1.6
60	33.0	29.8	88.7	3.3	98.4	0.3	99.3	0.2	99.8	97	1.6
Av.	33.9	31.7	87.4	3.7	98.2	0.4	99.2	0.2	99.7	96	1.6
120	21.8	21.6	95.8	0.8	99.2	0.2	99.8	0.03	99.9	99	1.3
120	30.4	30.2	94.1	1.6	98.9	0.2	99.7	0.06	99.9	99	0.8
120	32.8	32.1	92.6	2.2	99.3	0.2	99.8	0.05	99.9	99	0.3
Av.	28.3	28.0	94.2	1.5	99.1	0.2	99.8	0.05	99.9	99	0.8

TABLE VIII

THE EFFECT OF FOUR FRESH ACID-FLUORIDE CONTACTS UPON THE AMOUNT OF SOLID DISSOLVED, THE AMOUNT OF PLUTONIUM RECOVERED AND THE AMOUNT OF PLUTONIUM IN THE UNDISSOLVED RESIDUE

Solvent: 12.5M HNO₃-0.2M CaF₂ for each of four contacts Solid to Liquid Ratio: 100 g/l for the first contact
Feed Material: Glovebox floor sweepings heel

Reflux Period (min.)	Initial Feed Conc. (% Pu)	1st Contact		2nd Contact		3rd Contact		4th Contact		Amt of Solid Dissolved (%)	Undissolved Residue Conc. (% Pu)
		Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)		
15	26.8	23.8	84.7	2.9	95.7	0.6	98.1	0.2	98.9	79	1.0
15	27.0	23.8	84.3	2.9	95.3	0.7	97.8	0.3	99.0	77	0.9
15	24.2	22.1	86.2	2.2	95.4	0.4	97.0	0.4	98.6	78	1.1
Av.	26.0	23.2	85.1	2.7	95.5	0.6	97.6	0.3	98.8	78	1.0
30	24.0	21.8	85.0	2.4	95.3	0.6	98.0	0.3	99.2	80	0.8
30	25.2	23.5	85.6	2.5	95.7	0.6	98.3	0.2	99.0	78	0.8
30	24.2	22.1	86.4	2.2	95.8	0.5	97.9	0.2	99.0	78	0.8
Av.	24.5	22.5	85.7	2.4	95.6	0.6	98.1	0.2	99.1	79	0.8
60	26.0	22.8	82.3	2.3	96.7	0.5	98.5	0.2	99.3	80	0.6
60	24.0	22.3	86.7	2.3	95.9	0.6	98.3	0.2	99.2	78	0.6
60	24.8	23.4	88.2	2.1	96.9	0.5	98.7	0.2	99.4	80	0.5
Av.	24.9	22.8	85.7	2.2	96.5	0.5	98.5	0.2	99.3	79	0.6
120	24.6	23.7	89.8	1.9	97.8	0.3	99.0	0.1	99.4	81	0.5
120	24.0	22.7	88.0	2.0	96.5	0.4	98.2	0.1	98.8	81	1.1
120	36.0	33.5	88.4	3.9	99.2	0.2	99.7	0.1	99.9	77	0.1
Av.	28.2	26.6	88.7	2.6	97.8	0.3	99.0	0.1	99.4	80	0.5

TABLE IXA

THE EFFECT OF FOUR FRESH ACID-FLUORIDE CONTACTS UPON THE AMOUNT OF SOLID DISSOLVED, THE AMOUNT OF PLUTONIUM RECOVERED AND THE AMOUNT OF PLUTONIUM IN THE UNDISSOLVED RESIDUE

Solvent: 12.5M HNO₃-0.2M CaF₂ for each of four contacts Solid to Liquid Ratio: 100 g/l for the first contact
Feed Material: Filter sludge

Reflux Period (min.)	Initial Feed Conc. (% Pu)	1st Contact		2nd Contact		3rd Contact		4th Contact		Amt of Solid Dissolved (%)	Undissolved Residue Conc. (% Pu)
		Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)		
15	19.4	10.0	50.2	4.4	73.2	2.0	83.3	1.2	89.6	37	3.0
15	19.0	10.0	51.6	4.1	73.7	1.6	81.7	1.3	88.4	39	3.4
15	20.6	10.0	47.1	4.7	69.5	2.4	81.3	1.5	88.5	37	3.5
Av.	19.7	10.0	49.6	4.4	72.1	2.0	82.1	1.3	88.8	38	3.3
30	21.8	11.7	52.8	5.2	76.6	2.5	88.0	1.2	93.6	40	2.1
30	20.2	11.5	55.1	4.4	76.6	1.8	85.7	1.0	90.8	46	2.7
30	21.8	11.7	53.2	5.6	79.0	2.2	89.1	1.0	93.6	47	2.4
Av.	21.3	11.6	53.7	5.1	77.4	2.1	87.6	1.1	92.7	47	2.4
60	17.6	11.3	60.8	3.3	79.6	1.7	89.1	0.9	94.0	55	2.0
60	21.2	13.0	59.5	4.6	81.0	2.2	91.0	1.0	95.6	55	1.8
60	22.8	14.2	60.5	4.9	81.8	1.9	90.4	1.5	96.8	54	1.4
	20.2	12.8	60.3	4.3	80.8	1.9	90.2	1.1	95.5	55	1.7
120	30.0	16.8	54.7	7.6	80.8	4.1	93.3	1.1	96.9	67	1.8
120	28.6	16.9	57.3	6.8	80.4	3.1	90.9	1.3	95.4	67	1.8
120	28.0	18.1	62.8	5.9	83.6	2.6	92.9	1.3	97.1	66	1.9
120	29.4	16.5	53.7	7.2	77.6	3.6	89.8	2.3	97.3	67	2.0
Av.	26.5	17.1	57.1	6.9	80.4	3.4	81.7	1.5	96.7	67	1.9

TABLE XA

EFFECT OF TWO FRESH ACID-FLUORIDE CONTACTS ON PLUTONIUM RECOVERY

Solvent: 12.5M HNO₃-0.2M CaF₂ for each of two contacts Solid to Liquid Ratio: 100 g/l for the first contact

Feed Material	Experimental Conditions	Initial Feed Conc. (% Pu)	1st Contact		2nd Contact		Amt of Solid Dissolved (%)	Undissolved Residue Conc. (% Pu)
			Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)	Filtrate Conc. (g Pu/l)	Amt of Pu Rec (%)		
CIA	(1)	26.2	20.4	71.6	6.0	94.9	84	2.6
IAH	(1)	15.9	13.1	84.0	2.4	96.2	72	1.2
Swps	(1)	32.2	27.8	82.4	5.7	99.6	98	0.2
SH	(1)	36.2	28.6	75.5	8.7	99.3	75	0.5
Av.		27.6	22.5	78.4	5.7	97.5	82	1.1
CIA	(2)	26.7	20.3	70.5	7.4	97.1	76	1.5
IAH	(2)	15.9	14.3	84.5	2.4	99.0	72	0.2
Swps	(2)	36.6	30.2	78.2	6.4	93.9	77	0.8
SH	(2)	33.2	26.6	75.2	8.2	98.1	68	0.7
Av.		28.1	22.8	77.1	6.1	97.0	73	0.8
CIA	(3)	28.1	21.6	70.0	7.3	95.6	83	3.1
IAH	(3)	13.6	11.6	82.9	2.2	98.7	75	2.6
Swps	(3)	33.0	28.3	80.8	3.7	97.0	92	5.3
SH	(3)	30.3	26.3	65.2	13.1	98.0	69	1.2
Av.		28.2	22.0	74.7	7.1	97.3	80	3.0
CIA	(4)	25.8	19.2	67.9	6.6	93.4	76	3.8
IAH	(4)	16.2	12.7	76.8	2.9	94.9	71	1.6
Swps	(4)	21.7	22.4	97.5	0.3	98.9	94	0.8
SH	(4)	37.9	30.1	59.1	14.7	98.0	80	1.0
Av.		25.4	19.4	75.3	6.1	96.3	80	1.8
CIA	(5)	27.0	20.3	68.7	6.4	91.8	74	3.9
IAH	(5)	14.9	12.7	81.7	2.0	95.3	68	1.5
Swps	(5)	31.9	28.4	84.3	4.8	99.2	93	1.2
SH	(5)	36.8	26.9	68.5	10.7	98.1	72	1.3
Av.		27.9	22.1	75.8	6.0	96.1	77	2.0
CIA	(6)	27.4	20.9	69.4	6.6	93.3	78	3.7
IAH	(6)	15.8	13.2	81.4	2.4	96.2	70	1.2
Swps	(6)	30.5	27.3	85.6	4.1	99.0	86	0.9
SH	(6)	38.4	27.4	67.7	11.5	97.6	70	1.2
Av.		28.0	22.2	76.0	6.2	96.5	76	1.8
CIA	(7)	27.5	21.0	72.3	5.5	92.5	76	3.7
IAH	(7)	14.9	12.9	82.9	1.7	93.8	67	1.7
Swps	(7)	20.3	19.7	92.0	1.6	99.3	93	0.6
SH	(7)	35.8	25.3	66.4	11.1	98.4	74	1.1
Av.		24.6	19.7	78.4	5.0	96.0	78	1.8

CIA - Crushed Incinerator Ash
IAH - Incinerator Ash Heel
Swps - Glovebox Floor Sweepings
SH - Glovebox Floor Sweepings Heel

Conditions (1) - Two Contacts:

First contact with 12.5M HNO₃-0.2M CaF₂ for one hour.
Second contact with 12.5M HNO₃-0.2M CaF₂ for 3 hours.

Conditions (2) - Two Contacts:

Same as conditions (1) except that an additional amount of CaF₂ (0.2M) was added after the first and second hour of reflux.

Conditions (3) - Two Contacts:

Same as conditions (2) except that the additional amount of CaF₂ added was 0.1M.

Conditions (4) - Two Contacts:

Same as conditions (1) except that the first contact was for 1/2 hour and the second contact was for 2 hours.

Conditions (5) - Two Contacts:

Same as conditions (1) except that the first contact was for one hour and the second contact was for 1-1/2 hours.

Conditions (6) - Two Contacts:

Same as conditions (5) except that an additional amount of CaF₂ (0.1M) was added halfway through the second reflux period.

Conditions (7) - Two Contacts:

Same as conditions (1) except that the contacts were each one hour.