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DP - 689
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Nuclear Technology - Chemistry
and Chemical Engineering
AEC Research and Development Report

SEPARATION OF NEPTUNIUM AND PLUTONIUM BY ANION EXCHANGE

by

G. A. Burney
Separations Chemistry Division

April 1962

[redacted]

[redacted]

[redacted]

E. I. du Pont de Nemours & Co.
Savannah River Laboratory
Aiken, South Carolina

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FROM: J. E. BEACH JEB

May 11, 1962

DOCUMENT REVIEW [redacted]

Document:	Report DP-689 [redacted]
Title:	Separation of Neptunium and Plutonium by Anion Exchange
Author:	G. A. Burney [redacted]
Contractual Origin:	AT(07-2)-1[redacted]
Present Classification:	Secret [redacted]

Reference: Ryan, J. L., Concentration and Final Purification of Neptunium by Anion Exchange, HW-59193 (Rev.) 1959

No items were noted that, in my opinion, should be called to the attention of the AEC for patent consideration.

The report describes a process for recovering neptunium and plutonium from nitrate solution by adsorption on anion resins. Np and Pu in the IV valence state are adsorbed as anionic complexes. Fission products are removed by washing with 8M nitric acid. Pu is selectively eluted by washing with 8M nitric acid solution which contains ferrous sulfamate and hydrazine to reduce the PuIV to PuIII. Np is then eluted by washing with dilute (0.35M) nitric acid.

This process was previously described in the reference.

Other reagents evaluated for valence adjustment were semicarbazide, aminoguanidine and hydrogen peroxide.

JEB:pa

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EXPLOSIVES DEPARTMENT

June 21, 1963

Mr. T. B. Niland, Chief (2)
Classification and S. S. Accountability
Safety and Technical Services Division
Savannah River Operations Office
U. S. Atomic Energy Commission
Aiken, South Carolina

Dear Mr. Niland:

DECLASSIFICATION REVIEW - DP REPORTS

The following DP reports are recommended for declassification review on the basis of the SRO Classification Guide revisions of May 14, 1963.

DP-531 - Anion Exchange of Neptunium in Nitrate Solutions

This report appears to be unclassified by topic 292.8.3.

DP-689 - Separation of Neptunium and Plutonium by Anion Exchange

This report appears to be unclassified by topic 292.9.3. Topics 292.9.1, 292.9.2, and 292.11.1 have been considered but do not appear applicable.

DP-417 - Recovery of Np-237 and Pu-238 from Irradiated Neptunium Oxide

This report appears to be unclassified by topics 292.9.3, 292.12.2 and 292.12.3. Topic 292.9.2 does not appear applicable.

DP-719 - A Facility for Chemical Separation of Pu-238

This report appears to be unclassified by topics 292.4 and 292.9.3. Topics 292.6 and 292.11.1 have been considered but do not appear applicable, particularly since the equipment described here was a pilot plant operation.

DP-443 - Coextrusion of Powder-Compact Slugs

This report appears to be unclassified by topics 3.2.1 and 3.3.1 of CG-UF-1 and topic 292.12.4 of the SRO Classification Guide.

DP-729 - Chemical Processing of Pu-238

This report appears to be unclassified by topic 292.9.3. Topic 292.9.2 has been considered but does not appear to be applicable. Topic 292.9.1.1 rather than topic 292.9.1 appears to be applicable since no information is disclosed concerning the continuity of operation of the unit processes nor of the process as a whole.

Yours very truly,

S. A. McNeight
Technical Assistant
ATOMIC ENERGY DIVISION

SAMcN:hbw

Form AEC-426(11-61)
AEC Manual Chapter 3202, Appendix 064a

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Author(s) G. A. Burney

Contract No. AT(07-2)-1

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May 17, 1962

Mr. W. P. Overbeck, Director
Savannah River Laboratory
E. I. du Pont de Nemours & Company
Aiken, South Carolina

Attention: Dr. J. W. Morris, Director
Separations & Services Section

Dear Mr. Overbeck:

We have reviewed the following classified document transmitted by your letter of May 15, 1962:

DP-689 - "Separation of Neptunium and Plutonium by Anion Exchange" - by G. A. Burney

This office has no objection to its proposed release. Mr. O'Rear's office was so notified by telephone this date.

Sincerely yours,

Paul J. Hagelston, Director
Technical & Production Division

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July 18, 1963

Mr. W. P. Overbeck, Director
Savannah River Laboratory
E. I. du Pont de Nemours and Company
Aiken, South Carolina

Attention: Dr. J. W. Morris, Director
Separations and Services Section

Dear Mr. Overbeck:

Based on S. A. McNeight's recommendations of June 21, 1963, we have reviewed the following documents for possible declassification:

DP-531TL, letter, C. H. Ice to H. Worthington, 12-6-60
DP-531, "Anion Exchange of Neptunium in Nitrate Solutions", by G. A. Burney

DP-689TL, letter, C. H. Ice to H. Worthington, 6-18-62
DP-689, "Separation of Neptunium and Plutonium by Anion Exchange", by G. A. Burney

We concur in Dr. McNeight's opinion that these documents contain no Restricted Data or other classified information; accordingly, the documents have been declassified without deletions by authority of Thomas B. Niland, Classification Officer, Savannah River Operations Office, U.S. Atomic Energy Commission, July 18, 1963. There are no patent objections in connection with these declassifications.

Please arrange to notify all holders of copies of the documents listed of this declassification action citing the above authority. The copies charged to SROO are being marked accordingly.

Sincerely yours,

Paul J. Hagelston, Director
Safety & Technical Services Division

cc: S. A. McNeight, Technical Assistant
Explosives Department, AED, Wilmington

DP-689

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Dear Mr. Hagelston:

PROPOSED PUBLICATION - DP- 689

Attached for review for suitability for release under the Civilian Application Program are three copies of the following classified report:

Separation of Neptunium and Plutonium by Anion Exchange
by G. A. Burney

Please contact my office if you have any comments. The report will be released when approval is received, but not until after 14 days from the date shown above.

Yours very truly,
Original signed by
W. P. Overbeck
W. P. Overbeck, Director
Savannah River Laboratory

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DP-689, SEPARATION OF NEPTUNIUM AND PLUTONIUM BY ANION EXCHANGE

by G. A. Burney

Quite early in the development of the plant process for recovery of Np^{237} and Pu^{238} from irradiated Np^{237} it was necessary to select the chemical scheme for separating Pu^{238} and Np^{237}. The choice was between the oxidative method in which the plutonium is retained on the anion resin and the oxidized neptunium is allowed to pass through the resin bed, and the reductive method in which both the plutonium and the neptunium are retained, following which the plutonium is reduced and washed from the resin bed.

The reductive method was selected for the plant process because laboratory data and experience in the Chemical Processing Facility showed that this method would give satisfactory performance in the plant. On the other hand, the laboratory data showed that separation by the oxidative method was less efficient, and all attempts to improve the separation were unsuccessful. Experience in the plant has verified the predicted satisfactory performance of the reductive method. Although the separation is quite sensitive to process variables, the recent acquisition of a low crosslinked resin for plant use has permitted very efficient separations without undue restrictions in operating procedures.

The attached report describes the laboratory studies of the reductive method; studies of the oxidative method will be presented in a subsequent report. In future processes both methods of separation should be considered because changes in operating conditions or requirements for the degree of separation could make the oxidative separation the preferred method.

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C. H. Ice

[redacted] C. H. Ice, Research Manager
Separations Chemistry Division

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Author(s) G A Burney

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Division Separations Chemistry

Indicate known prior art

Approval (Introduction, Summary, Cover Letter) JWM 11/26/61 Section Director and Laboratory Director

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SEPARATION OF NEPTUNIUM AND PLUTONIUM BY ANION EXCHANGE

by

Glenn A. Burney

April 1962

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Separations Chemistry Division

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ABSTRACT

The separation of neptunium and plutonium by anion exchange in nitrate solutions was studied under a wide variety of experimental conditions. A method of separation was developed that is adaptable to remote operation in the plant. In the method, Np(IV) and Pu(IV) are absorbed on an anion exchange resin from 8M nitrate solution. The column is washed with 8M HNO_3 to remove contaminants; then the Pu is removed as Pu(III) by washing the column with HNO_3 containing ferrous sulfamate and hydrazine. Finally, the Np is eluted with dilute HNO_3.

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CONTENTS

LIST OF TABLES

LIST OF FIGURES

SEPARATION OF NEPTUNIUM AND PLUTONIUM BY ANION EXCHANGE

INTRODUCTION

At the time this study was initiated, no satisfactory method that could be applied in plant-scale ion exchange processing had been reported for separating neptunium from plutonium. One process that had been described(1) employed a chloride system and was, therefore, incompatible with stainless steel equipment. The objective of the present work was to develop a simple and reliable anion exchange separation that could be used in nitrate solutions, and that was adaptable to remote operation in the plant. The investigation was prompted by the need for such a separation step in the proposed chemical process for the recovery of Pu^{238} and Np^{237} from irradiated Np^{237}.

The work reported here was done at about the same time the anion exchange of Np^{237} and Pu^{239} in nitrate systems was studied at Hanford(2). While the work at Hanford was quite broad in scope, the present report describes the detailed study of the anion exchange of Np^{237} and Pu^{238} that was required in the development of a plant process.

The anion exchange technique of separating neptunium and plutonium that was developed in this study has been successfully applied to the separation of gram quantities of Np^{237} and Pu^{238} in the laboratory(3), and to kilogram quantities in pilot plant equipment(4). The details of the development work and a survey of the effect of variables on the ion exchange process are reported here.

SUMMARY

Satisfactory conditions were found for separating neptunium and plutonium efficiently by anion exchange in nitrate solutions. In the method considered most acceptable for process application, the anionic nitrate complexes of Np(IV) and Pu(IV) are absorbed on an anion exchange column from 8M nitrate solution. The column is then washed with 8M HNO$_3$ to remove fission products and other contaminants. The plutonium is separated from the neptunium by washing the bed with a nitric acid solution containing ferrous sulfamate and hydrazine. This solution reduces Pu(IV) to the weakly absorbed (III) state, while maintaining the neptunium in the strongly absorbed (IV) state. The combination of hydrazine with the ferrous sulfamate decreases the oxidation of the ferrous ion, and results in a minimum of gas formation in the resin column. As the final step in the cycle, the neptunium is eluted from the resin with dilute nitric acid.

With this technique, the fission products in a solution containing neptunium and plutonium were reduced by a factor of about 1000. The neptunium and plutonium were cleanly separated; the neptunium product contained only a few tenths of a per cent of plutonium, and vice versa. The process did not work quite as well with Pu^{238} as it did with Pu^{239}, presumably because alpha radiolysis was greater with Pu^{238}. The most satisfactory anion exchange resin of those investigated was "Dowex" 1-X3, 40-60 mesh. The effects of temperature, flow conditions, compositions of the feed and wash solution, and other important variables were studied.

DISCUSSION

CHEMISTRY RELATED TO SEPARATION OF NEPTUNIUM AND PLUTONIUM

The actinide elements in the quadrivalent state form anionic nitrate complexes that are strongly absorbed by anion exchange resins. The species that is absorbed from concentrated nitrate solutions has been shown to be $M(NO_3)_6^=$ in the case of Pu(IV), Np(IV), U(IV), and Th(IV)(5). The anion exchange absorption of these ions is an attractive method of purifying them from other elements(2-7). Distribution data for the different valence states of neptunium and plutonium are shown in Table I.

TABLE I

Relative Absorbability of Actinides

Isotope	Distribution Ratio(a)
Np(IV)	600
Np(V)	<10
Np(VI)	<25
Pu(III)	<10
Pu(IV)	1000

(a) $\frac{\text{(mg isotope)/(ml of resin)}}{\text{(mg isotope)/(ml of solution)}}$ after one-hour equilibration of actinide in 8M HNO_3 with "Dowex" 1-X4 resin

The distribution ratios show that valence control is necessary for an effective separation of neptunium from plutonium. Two possibilities exist for the control of valence: (1) reduction of plutonium to the weakly absorbed (III) state while maintaining neptunium in the strongly absorbed (IV) state, or (2) oxidation of neptunium to the (V) state while maintaining plutonium in the absorbable (IV) state. Preliminary experiments indicated that the former possibility, the reductive technique, could be most readily developed for process application, and the development of this technique is described here. The details of the oxidative technique will be described in a separate report.

A major problem in the reductive technique was the choice of suitable reductants for maintaining the Pu(III) state. Pu(IV) is the stable oxidation state of plutonium in the strong nitric acid or nitrate salt solutions necessary for anion exchange. As an added complication, only a few reductants that are capable of reducing Pu(IV) to Pu(III) are stable in strong nitric acid. The final choice was a combination of ferrous sulfamate and hydrazine. Ferrous ion is a rapid reductant for Pu(IV); in 6 to 8M nitric acid reduction is almost complete in less than five minutes at room temperature. Hydrazine and semicarbazide reduce Pu(IV) only partly and at a very slow rate in 8M nitric acid.

The hydrazine, which is used in combination with ferrous sulfamate, serves as a supplemental reducing agent and reacts with nitrous acid to avoid the rapid, autocatalytic oxidation of Fe(II) and Pu(III). Aqueous hydrazine reacts faster with nitrous acid than does sulfamate ion, and the reaction produces less nitrogen gas. This results in less gas formation in the anion exchange column. The hydrazine does not decrease the slow oxidation of Fe(II) and Pu(III) that occurs in strong nitric acid solutions at 20 to 40°C, as shown by the data in Table II. But, by depressing the rapid autocatalytic oxidation in the resin bed, it significantly reduces the amount of insoluble gas formed in the bed. Because one of the reaction products of hydrazine and nitrous acid is hydrazoic acid, the use of hydrazine in this application was carefully evaluated to establish limits of safety that would avoid explosive HN_3-air mixtures and explosive metal azide formation. This work is reported elsewhere(8).

TABLE II

Oxidation of Fe(II) and Pu(III) in Nitric Acid

The plutonium solutions contained
initially 3 g Pu^{239}(III)/l.

Initial Solution Composition, M			% Fe(II) Oxidized at 40°C			
HNO_3	N_2H_4	$Fe(SO_3NH_2)_2$	1 hr	2 hr	3.3 hr	21 hr
6	-	0.05	15	30	49	100
6	0.05	0.05	21	38	59	100
			% Pu(III) Oxidized at 23°C			
			24 hr	72 hr	168 hr	384 hr
6	-	0.05	0	0	10	40
6	0.05	0.05	0	5	15	30
8	-	0.05	5	10	45	65
8	0.05	0.05	5	10	35	55

EXPERIMENTAL PROCEDURE

The resins used in these studies were "Dowex" 1, "Dowex" 21K, and "Permutit" SK. "Dowex" 1 resin was tested at various crosslinkages including 2, 3, 4, and 8%. The resins were converted to the nitrate form by washing the chloride form received from the vendor with 2M nitric acid. The "Dowex" 1 and "Dowex" 21K resins were supplied as nominal 50-100 mesh beads; the "Permutit" SK was nominally 40-80 mesh. Where other particle size ranges or distributions are specified, the samples were obtained by "wet screening" the commercial resins in the nitrate form.

The experimental studies of the variables affecting the separation of plutonium and neptunium were made in small columns of resin 12.6 inches long by 0.62 cm^2 in diameter. The absorption and elution data are presented in the figures as functions of "bed volumes" of solution. A bed volume is defined as the bulk volume of the settled resin in the bed.

The analyses for Np^{237}, Pu^{238}, and Pu^{239} were made by alpha counting. The neptunium was separated from plutonium or from solutions of high salt content by TTA (thenoyltrifluoroacetone) extraction when required, and any crosscontamination by other alpha emitters was determined by alpha pulse height analysis.

VARIABLES AFFECTING THE SEPARATION

SEQUENCE OF STEPS IN COLUMN OPERATION

Several modifications of a reductive separation scheme were developed and tested; these differed principally in the sequence of steps during the anion exchange cycle. All of the methods relied upon the absorption of Np(IV) on the resin and the washing of Pu(III) from the bed with a nitric acid solution containing ferrous sulfamate and hydrazine. The modifications are described as Methods I, II, and III in Table III. Each method has certain unique advantages which are discussed in the following paragraphs.

In Method I the plutonium in the feed is reduced to weakly absorbed Pu(III) and is washed from the resin before the residual fission products are removed by washing. In the 6M HNO_3 feed solution containing excess reductants, more than 98% of the plutonium is in the (III) state; but, in spite of this, about 50% of the plutonium is absorbed when the feed solution is passed through the bed, presumably because of displacement of the Pu(III)-Pu(IV) equilibrium by the resin in favor of the strongly absorbable (IV) state. The Np(IV) is absorbed from the feed in a concentrated band on the bed, but the plutonium is absorbed rather uniformly over the entire bed of resin. At acid concentrations higher than 6M more of the plutonium is absorbed;

TABLE III

Methods of Column Operation

Step(a)	Stream Composition	Bed Volumes
	Method I	
Feed absorption	6M HNO_3, 0.05M N_2H_4, 0.05M $Fe(NH_2SO_3)_2$, 2 g Np(IV)/1, 0.3 g Pu(III)/1	10
Pu wash	6M HNO_3, 0.05M N_2H_4, 0.5M $Fe(NH_2SO_3)_2$	15
Fission product wash	8M HNO_3, 0.005M KF	20
Np elution	0.35M HNO_3	4
	Method II	
Feed absorption	8M HNO_3, 0.05M $Fe(NH_2SO_3)_2$, 2 g Np(IV)/1, 0.3 g Pu(IV)/1; solution heated to oxidize Fe(II) and produce Pu(IV)	10
Fission product wash	8M HNO_3, 0.005M KF	20
Pu wash	6M HNO_3, 0.05M N_2H_4, 0.05M $Fe(NH_2SO_3)_2$	15
Np elution	0.35M HNO_3	4
	Method III	
Feed absorption	Same as in Method II except for the presence of 5 bed volumes of recycled Pu wash solution	
Fission product wash	8M HNO_3, 0.005M KF	20
Pu wash	5.4M HNO_3, 0.05M N_2H_4, 0.05M $Fe(NH_2SO_3)_2$ (Only 7.5 bed volumes are collected as Pu product. The remaining 5 bed volumes are recycled to subsequent feed batch.)	12.5
Np elution	0.35M HNO_3	4

(a) In each method, the feed absorption step is preceded by a column reconditioning step in which about 3 bed volumes of 8M HNO_3 are passed through the column to remove residual elutriant.

therefore, the lowest acid concentration compatible with maintaining a high saturation of neptunium on the resin is used. The main advantage of Method I is the decreased exposure of the resin to plutonium alpha activity. This is an important advantage if the plutonium being processed contains a large percentage of the highly active Pu^{238} isotope. By diminishing the saturation and the residence time for Pu^{238} on the bed, the useful life of the anion resin is considerably extended. Disadvantages of Method I for process application are:

(1) A large volume of the plutonium product solution results from the 10 bed volumes of feed plus the 15 bed volumes of plutonium wash solution.

(2) The plutonium is not decontaminated from impurities contained in the feed.

In Method II, the Pu(IV) is coabsorbed with Np(IV), and the fission products are washed from the bed before the plutonium is separated. The feed solution is adjusted to 8M HNO_3 to obtain optimum absorption and with 0.05M ferrous sulfamate to reduce the neptunium to Np(IV) and plutonium to Pu(III). The feed is then heated at 55°C for 30 minutes to oxidize Pu(III) to Pu(IV) and the excess Fe(II) to Fe(III). The advantages of Method II are:

(1) The volume of the plutonium product solution is decreased (15 bed volumes compared with 25 bed volumes in Method I).

(2) The plutonium as well as the neptunium is decontaminated from fission products and other impurities.

(3) The feed adjustment step is simplified because less rigid control of the acidity is required.

The disadvantages are:

(1) The exposure and degradation of the resin are increased.

(2) The losses of neptunium and plutonium to the effluents from the absorption and fission product wash steps are increased. These losses are presumably caused by the alpha radiation from Pu^{238}.

In Method III, which is considered most acceptable for process application, Pu(IV) and Np(IV) are coabsorbed as in Method II. The bed is washed with fission product wash solution, and the plutonium is then reduced and washed from the bed. The important difference from Method II is that only about the first 7.5 bed volumes of the plutonium wash effluent are collected as the plutonium product. The remaining 5 bed volumes of effluent are recycled to the subsequent feed batch. This method has the main advantage of improving the separation of the plutonium and neptunium. By decreasing the acidity of the partition wash solution from 6.0 to 5.4M HNO_3, the plutonium is more completely removed from the bed. The neptunium leakage from the bed is also increased at the lower acidity, but this is compensated by recycling the last fraction of the effluent containing most of this neptunium. Typically, the recycle fraction contained less than 10% of the plutonium and less than 1% of the neptunium. The disadvantages of Method III are essentially the same as those listed for Method II. An obvious variation in the recycle concept is to couple it with Method I, with reducing conditions in the feed, followed by the partition wash step in which the last fraction is recycled. The disadvantages of the large volume of plutonium product solution (17.5 bed volumes) and the lack of decontamination for the plutonium made this scheme unattractive for plant use.

From the data shown in Tables IV and V, the performances of Methods I and II may be compared with respect to the efficiency of separating plutonium and neptunium, the losses, and the decontamination from fission products. For these tests five pairs of runs by the two procedures were performed on 20-ml beds of "Dowex" 1-X4, 50-100 mesh resin. The feed for each run was a solution of nitric acid containing 2.3 g Np^{237}/l, 0.5 g Pu^{238}/l, and approximately 5×10^5 γ c/(min)(ml)* of mixed fission products. The resin in the columns was not changed during the course of the five runs. The data in Table IV show that the methods gave essentially equal separation of plutonium and neptunium, but that Method II gave higher losses, particularly of neptunium. Table V shows the improved decontamination of neptunium from fission products in Method II, particularly from Zr-Nb activity. No comparison is given for plutonium decontamination, since in Method I the plutonium receives almost no decontamination. The advantages of using fluoride in the reconditioning and fission product wash solutions are also illustrated in Table V.

Although data were not obtained for the performance of Method III, this method is subject to the lower decontamination from fission products if coupled to Method I and to the higher neptunium and plutonium losses if coupled to Method II. However, the advantage of low cross contamination of both products is maintained when Method III is coupled to either Method I or Method II.

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*Fission product gamma activity in c/(min)(ml) was determined at about 2.5% counting efficiency for Cs^{137}.

TABLE IV

Comparison of Efficiency of Separation and Losses for Methods I and II

Resin: 20 ml "Dowex" 1-X4, 50-100 mesh
Column: 0.62-cm^2 area, 12.6-inch height
Temperature: 23°C
Plutonium Wash: 15 bed volumes of 6.1M HNO_3 - 0.05M N_2H_4 - 0.05 $Fe(SO_3NH_2)_2$
Flow: 2 ml/(min)(cm^2)
Load: 400 mg Np, 70 mg Pu^{238}

	Efficiency of Separation				Losses, %			
	Np in Pu, wt %		Pu in Np, wt %		Np		$Pu^{(a)}$	
Run	Method I	Method II	Method I	Method II	Method I	Method II	Method I	Method II
1	0.28	0.19	0.06	0.14	0.05	1.9	0.5	1.4
2	0.38	0.24	0.09	0.11	0.08	1.0	0.6	1.0
3	0.23	0.27	0.11	0.05	0.06	1.3	0.6	0.7
4	-	-	0.11	0.04	-	-	0.7	0.8
5	0.32	0.25	0.24	0.07	0.06	0.9	1.3	0.7
Avg	0.30	0.24	0.12	0.08	0.07	1.3	0.7	0.9

(a) Loss includes Pu not separated from Np by the plutonium wash.

TABLE V

Comparison of Decontamination Performance for Methods I and II

	Run Conditions		Np Decontamination			
			Zr-Nb DF		Ru DF	
Run	0.1M HF in Recondition Wash	0.005M HF in Decontamination Wash	Method I	Method II	Method I	Method II
1	New Resin	Yes	770	2300	4000	5000
2	Yes	No	65	800	690	560
3	Yes	Yes	190	1000	610	540
4	No	Yes	150	3600	2700	2300
5	No	No	18	110	2000	2000

Fe(III)/Fe(II) RATIO IN THE PLUTONIUM WASH SOLUTION

The separation of neptunium and plutonium depends on the reduction of Pu(IV) to Pu(III) with Fe(II). The extent of reduction of Pu(IV) is dependent on the Fe(III)/Fe(II) ratio in solution. In strong nitrate solution and in the presence of anion exchange resin, the equilibrium is shifted in favor of Pu(IV) by nitrate complexing of Pu(IV) and absorption of nitrato-Pu(IV) anions by the resin. Column experiments showed that the efficiency of separating plutonium and neptunium was dependent on the Fe(III)/Fe(II) ratio in the plutonium wash solution. Two series of runs demonstrating the deleterious effect of increasing the Fe(III)/Fe(II) ratio are shown in Figures 1 and 2. Figure 2 shows that the lower crosslinked X3 resin is much less sensitive to the Fe(III)/Fe(II) ratio than is the X4 resin.

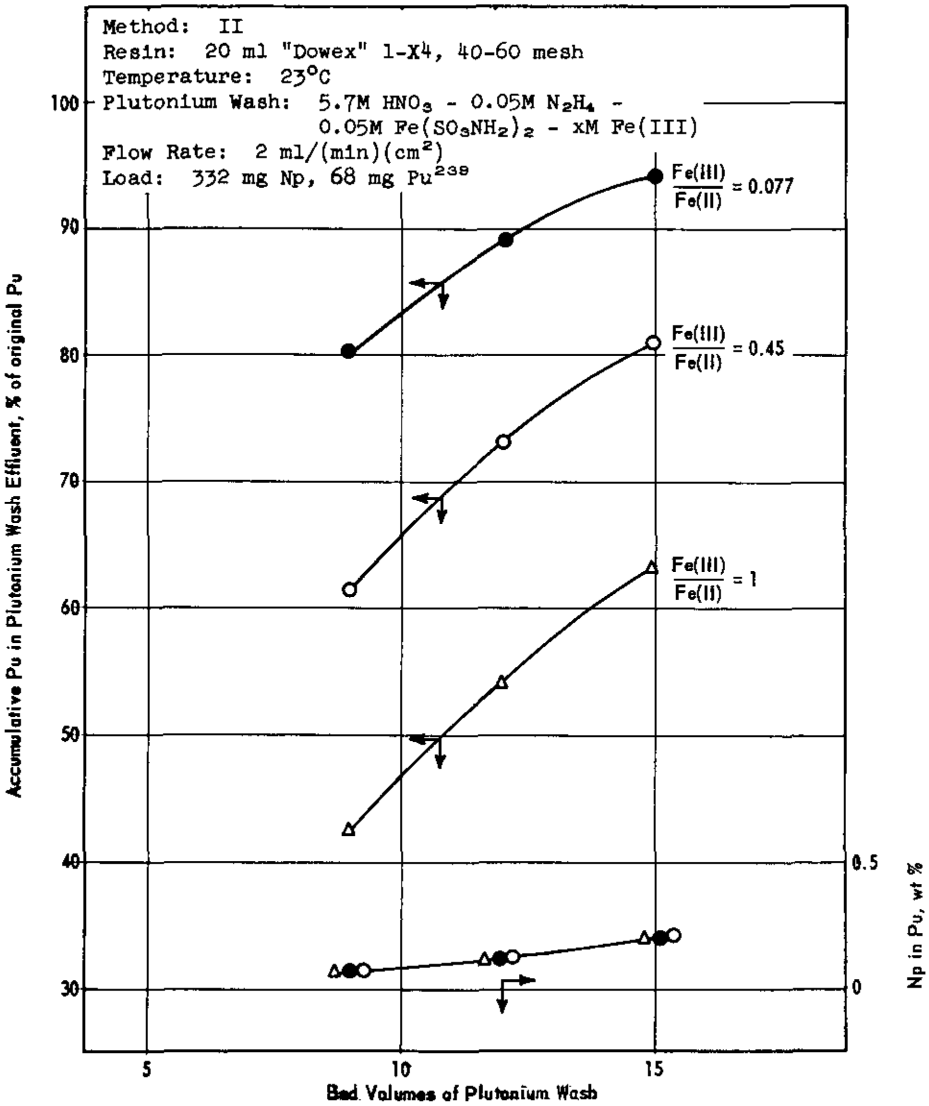


FIG. 1 EFFECT OF Fe(III)/Fe(II) RATIO IN PLUTONIUM WASH SOLUTION WITH "DOWEX" 1-X4 RESIN

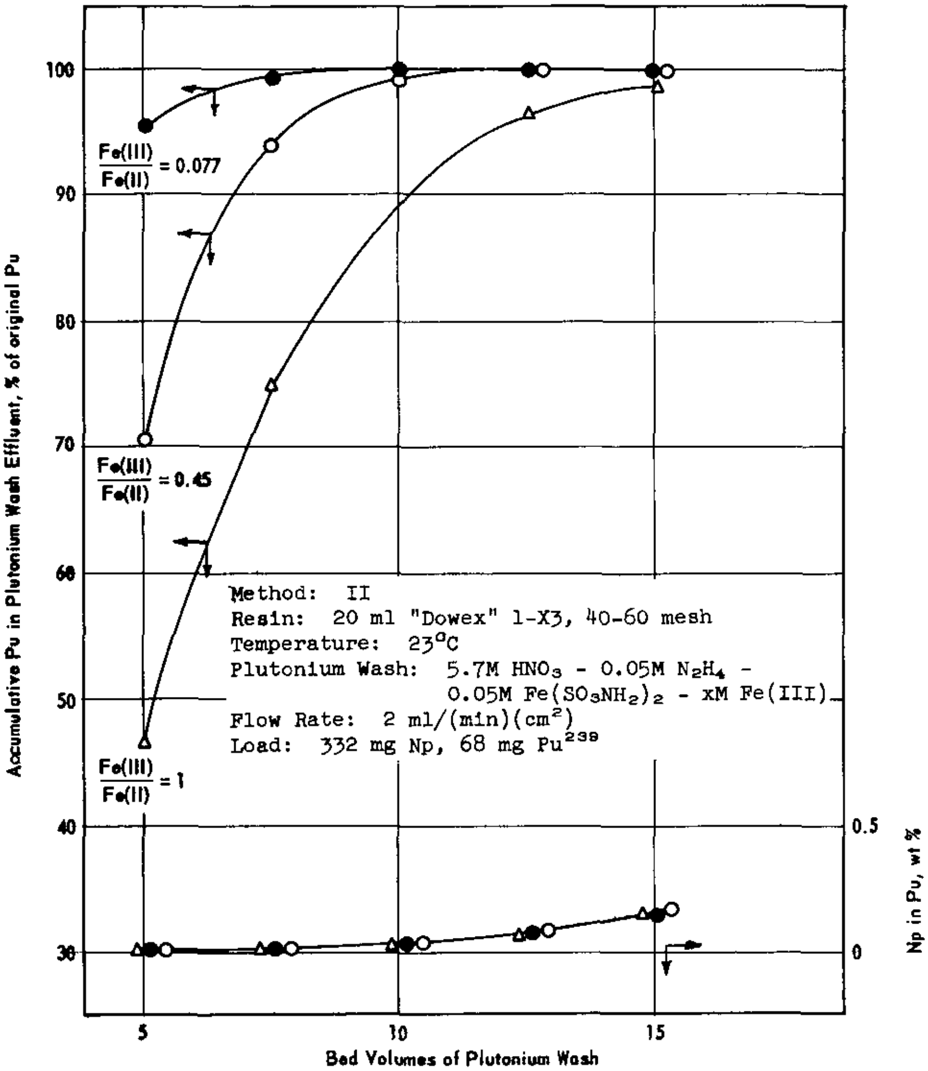


FIG. 2 EFFECT OF Fe(III)/Fe(II) RATIO IN PLUTONIUM WASH SOLUTION WITH "DOWEX" 1-X3 RESIN

Fe(III) in the plutonium wash solution results from several sources: (1) nitric acid oxidation of Fe(II) to Fe(III), (2) oxidation of Fe(II) by Pu(IV), (3) Fe(III) present in the ferrous sulfamate stock solution, and (4) Fe(III) from corrosion of equipment. All of these sources make some contribution, and under extreme conditions any source except (2) could be responsible for inadequate separation of plutonium from neptunium. In the preparation and storage of the plutonium wash solution it is important to hold oxidation of Fe(II) to a minimum. The acid adjustment is completed first, and the solution is cooled before the reductants are added. The rate of oxidation of Fe(II) is more than doubled for each ten degree increase in temperature. After acid adjustment and cooling, the hydrazine is added to scavenge nitrous acid before adding the ferrous sulfamate. Addition of concentrated acid after addition of the reductants is undesirable. The Fe(III) content of the ferrous sulfamate stock must be low, and storage vessels for process solutions must be corrosion resistant.

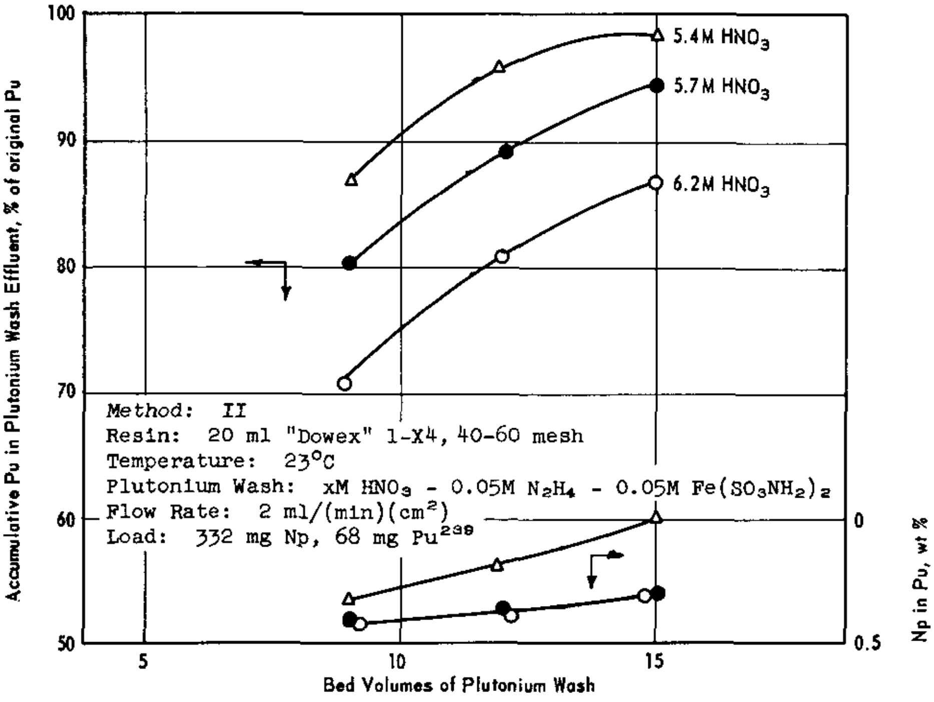
It was shown(8) that the rate of oxidation of Fe(II) in HNO_3 - 0.05M N_2H_4 - 0.05M $Fe(NH_2SO_3)_2$ solutions has approximately second power dependence on the hydrogen ion concentration, and that the rate is not decreased by increasing the concentrations of hydrazine to 0.2M or of ferrous sulfamate to 0.1M. About 10% of the Fe(II) is oxidized at 23°C in 6M HNO_3 - 0.05M N_2H_4 - 0.05M $Fe(NH_2SO_3)_2$ in 8 hours; at 55°C, oxidation is complete in less than 2.5 hours.

CONCENTRATION OF NITRIC ACID

Plutonium (IV) is rapidly and almost quantitatively reduced to the (III) state in 1 to 10M HNO_3 solution with hydrazine and ferrous sulfamate. However, when Pu(IV) is absorbed on anion exchange resin its reduction and removal from the resin as Pu(III) is relatively slow and inefficient. Advantage is taken of the reduced distribution coefficient of Pu(IV) for the resin at lower acidities to obtain more efficient removal of plutonium. With "Dowex" 1 resin, the elution of Pu(III) with 8M HNO_3 containing hydrazine and ferrous sulfamate is impractically slow and requires a large volume of solution. By using 5.5 to 6.0M HNO_3 containing the reductants, plutonium is removed with 10 to 15 bed volumes of solution. The effect of acid concentration in the plutonium wash solution on the rate of plutonium removal is shown in Figures 3 and 4. The lower nitrate concentration also decreases the distribution coefficient for Np(IV) so that increased leakage of neptunium occurs at lower acidities. This effect is shown by the lower curves in Figures 3 and 4. A comparison of these two figures also illustrates that the X3 resin is less sensitive to changes in acidity than is the X4 resin. It can be seen from Figure 3 that a plutonium wash of 5.4M HNO_3 gave 0.3 wt % Pu in the neptunium, and 0.5 wt % Np in the plutonium when Method II was used. This separation was improved by incorporating the recycle technique of Method III, whereby the neptunium in the plutonium was decreased to 0.1 wt %. With "Dowex" 1-X3 resin only 11 bed volumes of plutonium wash (5.4 to 6.2M HNO_3) were required to yield neptunium containing less than 0.1 wt % Pu, and plutonium containing less than 0.1 wt % Np. The data in this section are from experiments with Pu^{239}; the neptunium content of the plutonium would be approximately doubled with Pu^{238}.

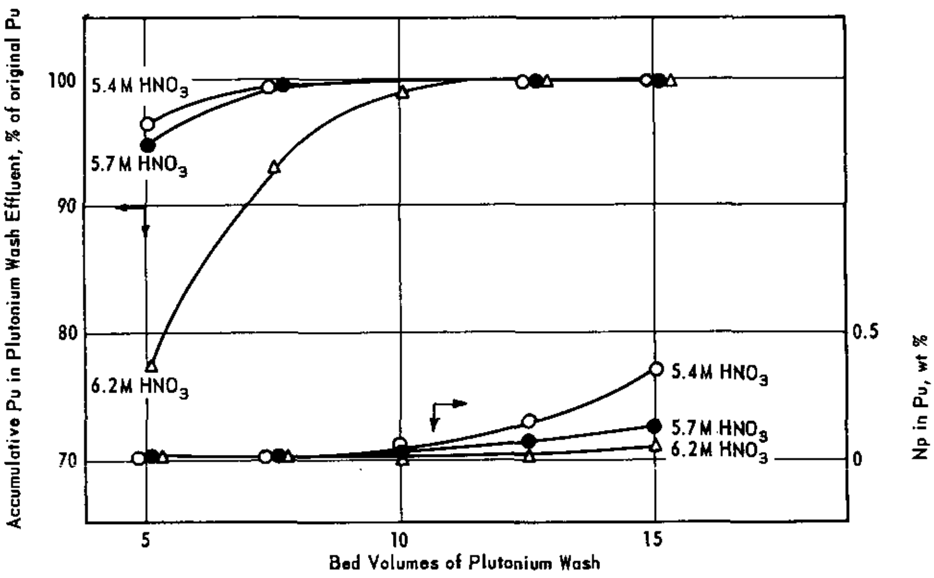
Separation with "Permutit" SK resin required higher acid concentrations to prevent excessive leakage of the neptunium. Typical data are shown in Figure 5.

Separation can also be accomplished with 1M HNO_3 - 4.5M $NaNO_3$ solutions containing ferrous sulfamate alone or in combination with hydrazine. In scouting runs on "Dowex" 1-X3 resin the removal of plutonium with either of these wash solutions was as efficient as with 5.5M HNO_3, but the leakage of neptunium was greater. The neptunium contained less than 0.1 wt % Pu for both systems, but the plutonium product contained 1 wt % Np for the acid-salt wash solution, compared with



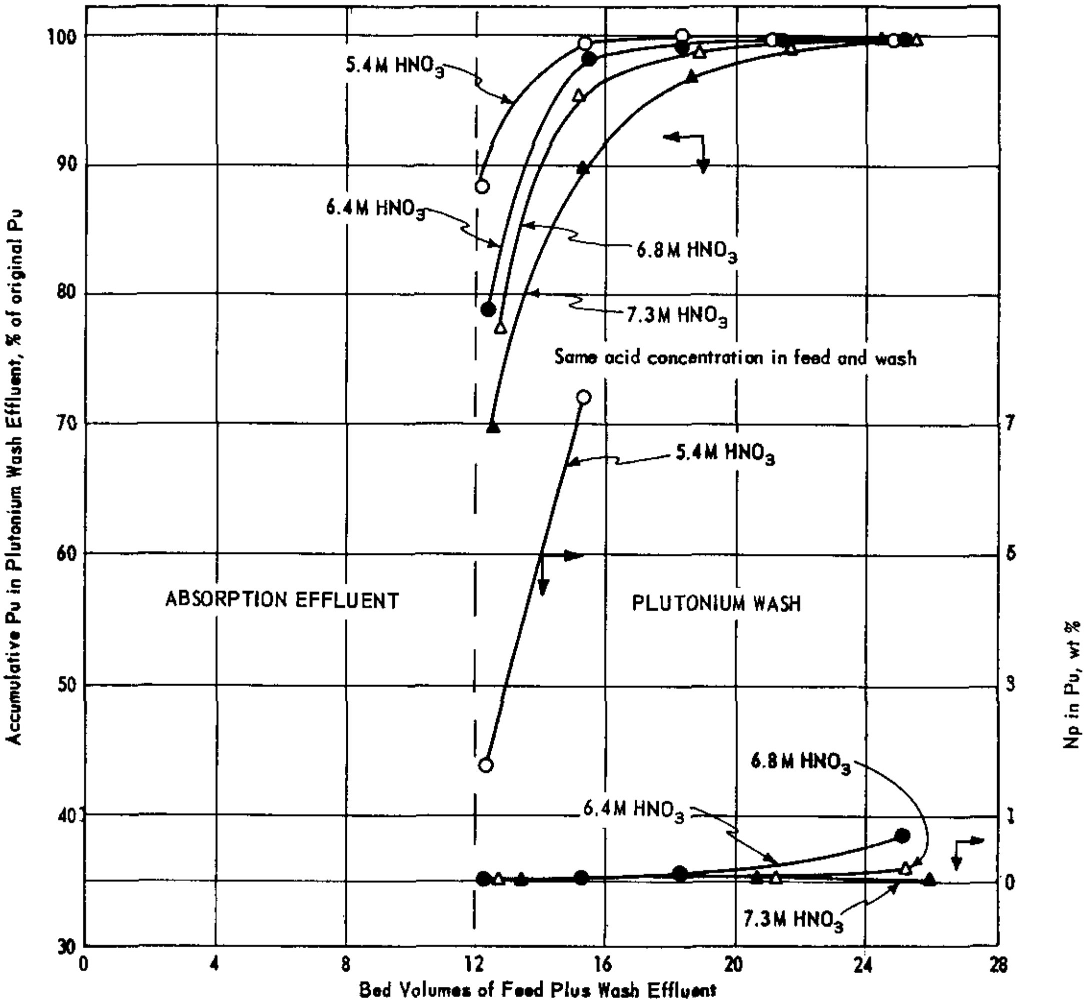
Method: II
Resin: 20 ml "Dowex" 1-X4, 40-60 mesh
Temperature: 23°C
Plutonium Wash: xM HNO_3 - 0.05M N_2H_4 - 0.05M $Fe(SO_3NH_2)_2$
Flow Rate: 2 ml/(min)(cm^2)
Load: 332 mg Np, 68 mg Pu^{239}

FIG. 3 EFFECT OF ACID CONCENTRATION IN PLUTONIUM WASH SOLUTION WITH "DOWEX" 1-X4 RESIN



Method: II
Resin: 20 ml "Dowex" 1-X3, 40-60 mesh
Temperature: 23°C
Plutonium Wash: xM HNO_3 - 0.05M N_2H_4 - 0.05M $Fe(SO_3NH_2)_2$
Flow Rate: 2 ml/(min)(cm^2)
Load: 332 mg Np, 68 mg Pu^{239}

FIG. 4 EFFECT OF ACID CONCENTRATION IN PLUTONIUM WASH SOLUTION WITH "DOWEX" 1-X3 RESIN



Method: I
Resin: 20 ml "Permutit" SK, 40-80 mesh
Temperature: 23°C
Feed: 1.7 mg Np/ml; 0.26 mg Pu/ml
Plutonium Wash: xM HNO_3 - 0.05M N_2H_4 - 0.05M $Fe(SO_3NH_2)_2$
Flow Rate: 1.7 ml/(min)(cm^2)
Load: 410 mg Np, 64 mg Pu^{239}

FIG. 5 EFFECT OF ACID CONCENTRATION IN FEED AND WASH SOLUTIONS WITH "PERMUTIT" SK RESIN

0.2 wt % for the pure acid wash. The increased leakage of neptunium from the bed with the salt system is probably associated with slower diffusion and less efficient reabsorption, or stripping, of the neptunium. In this case, slower flow rates for the wash solution might result in improvement. Similar contamination of the plutonium with neptunium occurs in the HNO_3 system when the flow is increased above 3 ml/(min)(cm^2).

Np/Pu RATIO IN FEED

At a constant total loading of actinides on the resin, the purity of the neptunium product improves as the ratio of absorbed neptunium to absorbed plutonium increases, and the purity of the plutonium product decreases. These effects are illustrated in Figures 6 and 7 for "Dowex" 1-X4 and 1-X3 resins, and the product purities are summarized in Table VI. The effects of the Np/Pu ratio are less pronounced for the X3 resin.

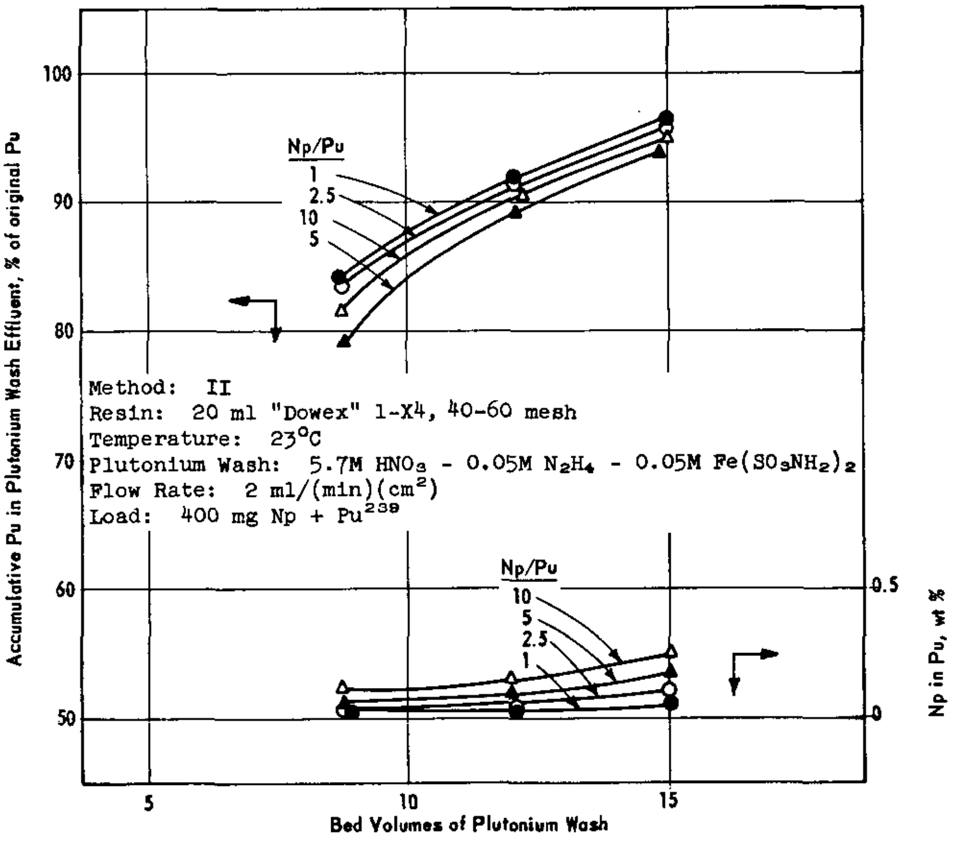
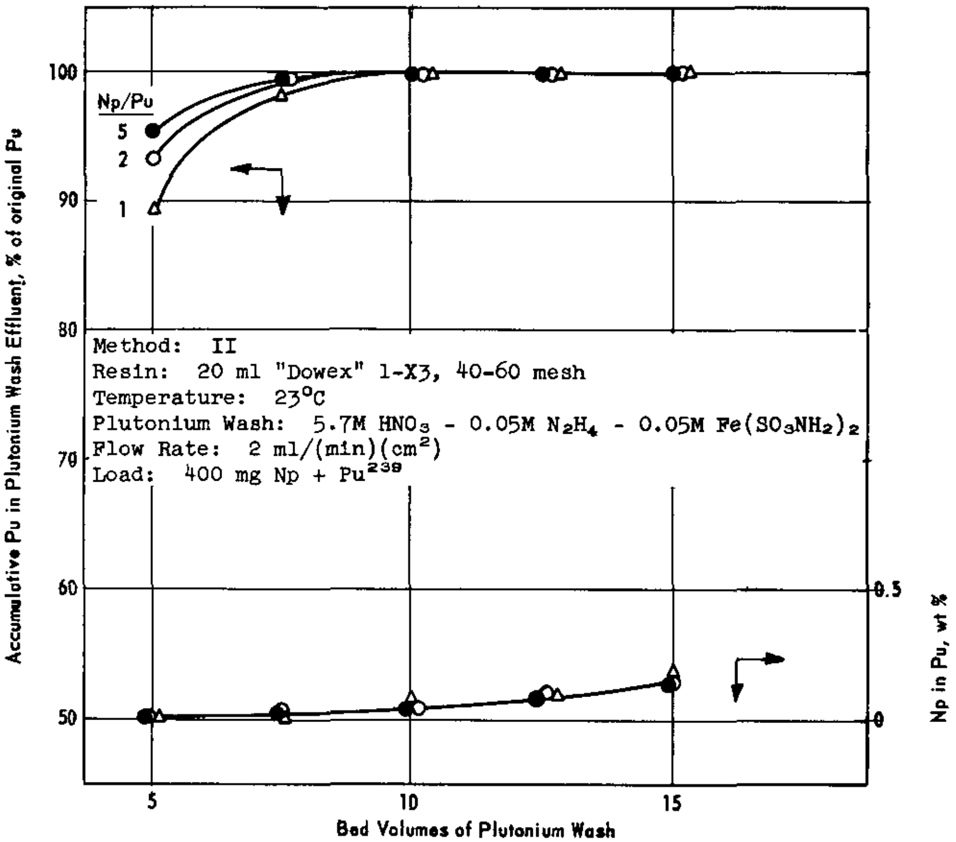


FIG. 6 EFFECT OF Np/Pu RATIO IN FEED WITH "DOWEX" 1-X4 RESIN



Method: II
Resin: 20 ml "Dowex" 1-X3, 40-60 mesh
Temperature: 23°C
Plutonium Wash: 5.7M HNO_3 - 0.05M N_2H_4 - 0.05M $Fe(SO_3NH_2)_2$
Flow Rate: 2 ml/(min)(cm^2)
Load: 400 mg Np + Pu^{239}

FIG. 7 EFFECT OF Np/Pu RATIO IN FEED WITH "DOWEX" 1-X3 RESIN

TABLE VI

Effect of Np/Pu Ratio on Separation

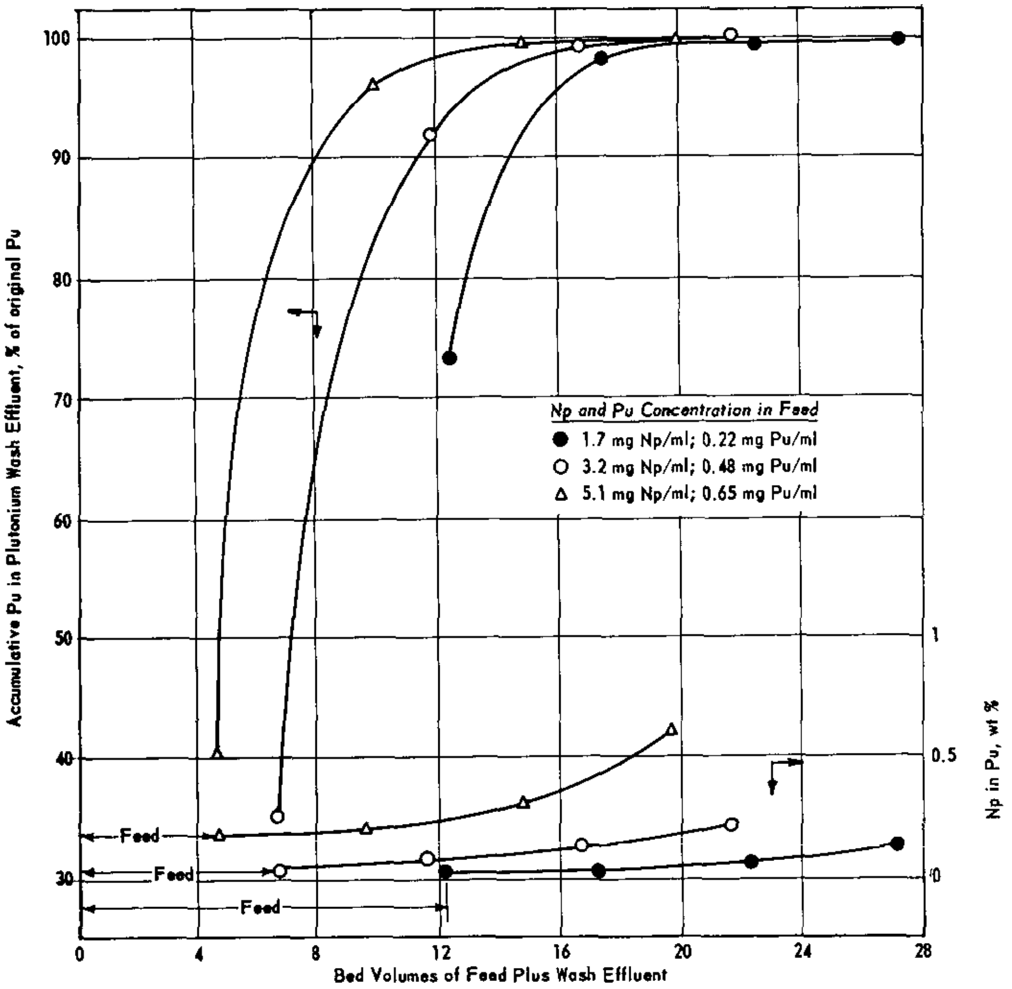
(Conditions as given in Figures 6 and 7)

Resin	Np/Pu Ratio in Feed	Np in Pu, wt %	Pu in Np, wt %
"Dowex" 1-X4, 40-60 mesh, 15 bed volumes of wash	1	0.05	3.9
	2.5	0.09	1.6
	5	0.17	1.1
	10	0.25	0.5
"Dowex" 1-X3, 40-60 mesh, 10 bed volumes of wash	1	0.04	0.1
	2	0.03	0.1
	5	0.03	0.1

The above data were obtained with Pu^{239}. With Pu^{238} the neptunium contamination of the plutonium product would increase more as the Np/Pu ratio is decreased.

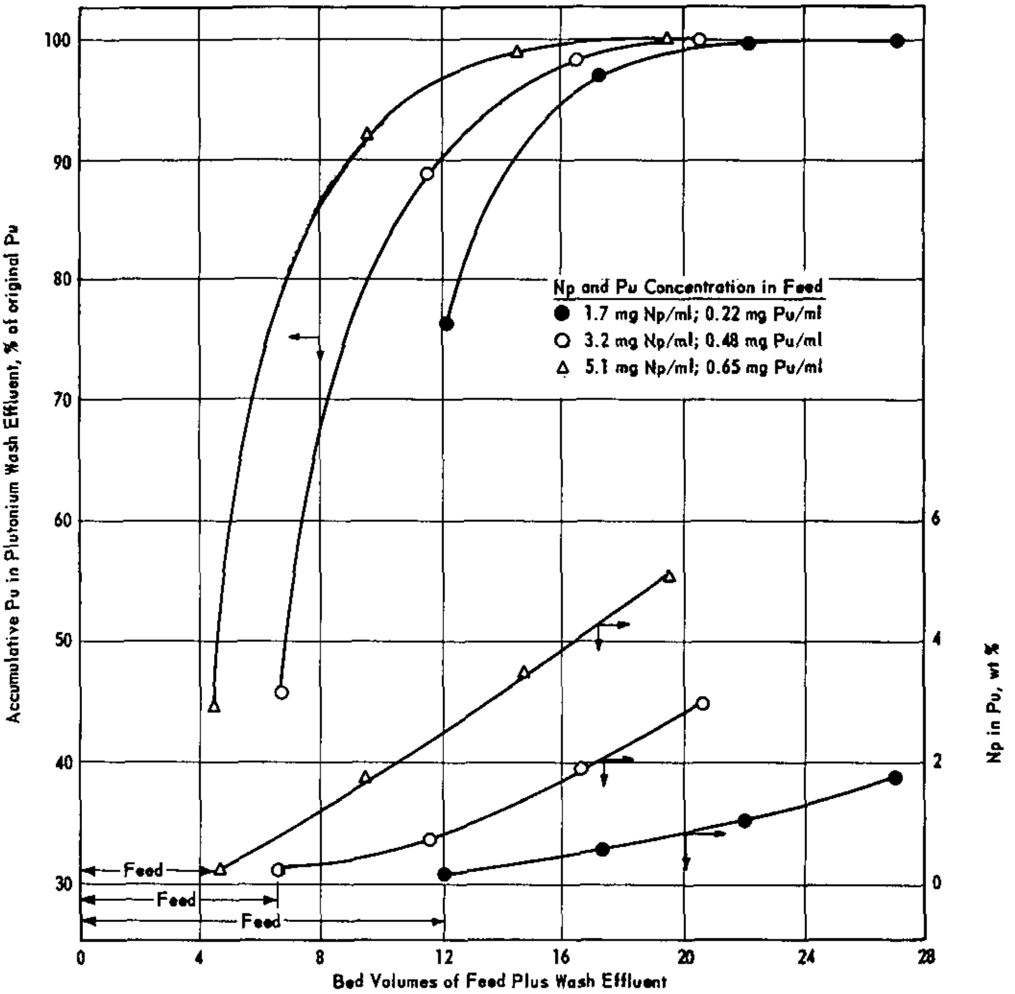
CONCENTRATION OF ACTINIDES IN FEED

Increasing the concentrations of neptunium and plutonium in the feed with all other conditions constant results in lower saturation of the actinides on the resin. This results in more efficient removal of the plutonium from the bed, but also leads to higher crosscontamination of the plutonium with neptunium. Typical results obtained with Method I are shown in Figures 8 and 9 for runs made with "Dowex" 1-X4 resin. The effects of higher concentrations of plutonium and neptunium in the feed can be compensated to some extent by operating at lower flow rates so that a constant mass throughput rate is approximated. The deleterious effect of higher flow rates, particularly on the cross-contamination of plutonium with neptunium. is shown in Figure 9.



Method: I
Resin: 20 ml "Dowex" 1-X4, 50-100 mesh
Temperature: 23°C
Feed and Wash: 6M HNO_3 - 0.1M N_2H_4 - 0.05M $Fe(SO_3NH_2)_2$
Flow Rate: 2 ml/(min)(cm^2)
Load: 420 mg Np, 58 mg Pu^{238}

FIG. 8 EFFECT OF CONCENTRATION OF Np AND Pu IN FEED AT LOW FLOW



Method: I
Resin: 20 ml "Dowex" 1-X4, 50-100 mesh
Temperature: 23°C
Feed and Wash: 6M HNO_3 - 0.1M N_2H_4 - 0.05M $Fe(SO_3NH_2)_2$
Flow Rate: 4 ml/(min)(cm^2)
Load: 420 mg Np, 58 mg Pu^{239}

FIG. 9 EFFECT OF CONCENTRATION OF Np AND Pu IN FEED AT HIGH FLOW

The effects of higher concentrations in the feed when "Permutit" SK resin is used are more pronounced because of the slower diffusion rates of the complex anions in the "Permutit" resin. In runs comparable to those in Figure 8, the neptunium contamination in the plutonium product was three to four times higher with "Permutit" SK than with "Dowex" 1-X4.

REAGENTS FOR VALENCE ADJUSTMENT

Ferrous sulfamate combined with hydrazine was the most satisfactory reductant-stabilizer found. Other reagents that were tested alone or in combinations were semicarbazide, aminoguanidine, and hydrogen peroxide.

Hydrazine, semicarbazide, and aminoguanidine, when used in combination with ferrous sulfamate, all serve the useful purpose of suppressing gas formation in the resin bed, presumably by scavenging nitrous acid that is formed and preventing it from reacting with sulfamate ion to form nitrogen. Hydrazine and aminoguanidine were slightly more effective than semicarbazide in this respect; hydrazine was preferred because of its lower cost. Hydrazine from three sources was tested and found to be equally effective: (1) as a 3M hydrazine nitrate solution, pH 5 to 6, purchased from Fairmount Chemical Company; (2) as a basic solution of 35% hydrazine in water purchased from Betz Laboratories under the trade name "Hyzeen"; and (3) as reagent-grade hydrazine sulfate.

Hydrogen peroxide at a concentration of 0.1M in 6 to 8M nitric acid reduces Pu(IV) to the extent of about 90% in less than one hour, and reduces neptunium essentially completely to Np(IV). In spite of this, separation of Np and Pu absorbed on anion resin bed using this wash solution was completely ineffective. Apparently, the resin phase shifts the equilibrium so strongly in favor of Pu(IV) that the peroxide reduction proceeds to only a small extent.

PLUTONIUM ALPHA ACTIVITY

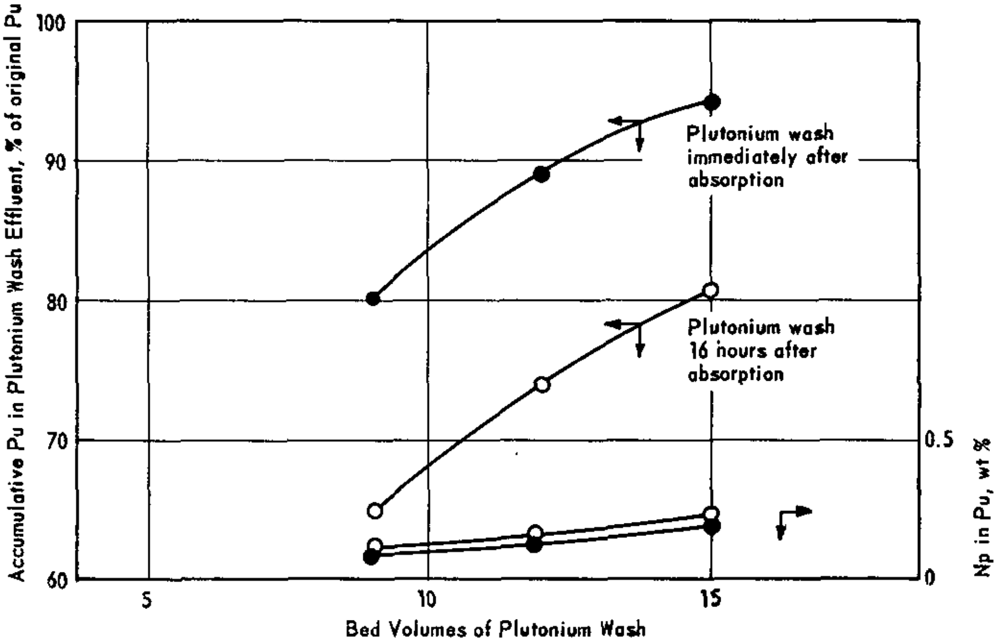
In general, because of increased oxidation due to alpha radiolysis, losses are higher when separating Pu^{238} from neptunium than for separating Pu^{239} and neptunium. This effect is particularly pronounced when Method II is used. Coabsorption of the neptunium and Pu^{238} in that method results in a relatively high saturation of Pu^{238} on the resin, and no reductants are present in the feed and fission product wash steps to overcome the effects of radiolysis products. In a typical cycle, involving the absorption of 16 mg of Np^{237} and 3.5 mg of Pu^{238} per ml of resin and the washing with 20 bed volumes of 8M HNO_3 - 0.005M KF prior to separation, the average neptunium loss to the effluents is 1.5 to 2.0% and the Pu^{238} loss is about 1%. Under the same conditions except substituting Pu^{239}, the neptunium loss is about 0.5% and the Pu^{239} loss is about 0.1%. That the increased neptunium loss in the presence of Pu^{238} is predominantly due to oxidation was shown by adding 0.02M H_2O_2 as a reductant to the decontamination wash solution. This lowered the loss from 1.5 to 0.7%. Most of the neptunium loss in the absence of Pu^{238} is due to complexing of the neptunium by fluoride in the wash solution. Addition of H_2O_2 to the wash results in higher plutonium losses, however, because there is some reduction to Pu(III).

In Method I, the effects of alpha radiation are less because only about one-half of the plutonium in the feed is absorbed, and this is washed from the bed in the subsequent step. With either Pu^{238} or Pu^{239} the neptunium loss is usually less than 0.1%, and the plutonium loss is dependent on variables other than the isotopic content.

TIME BETWEEN ABSORPTION AND PLUTONIUM REMOVAL

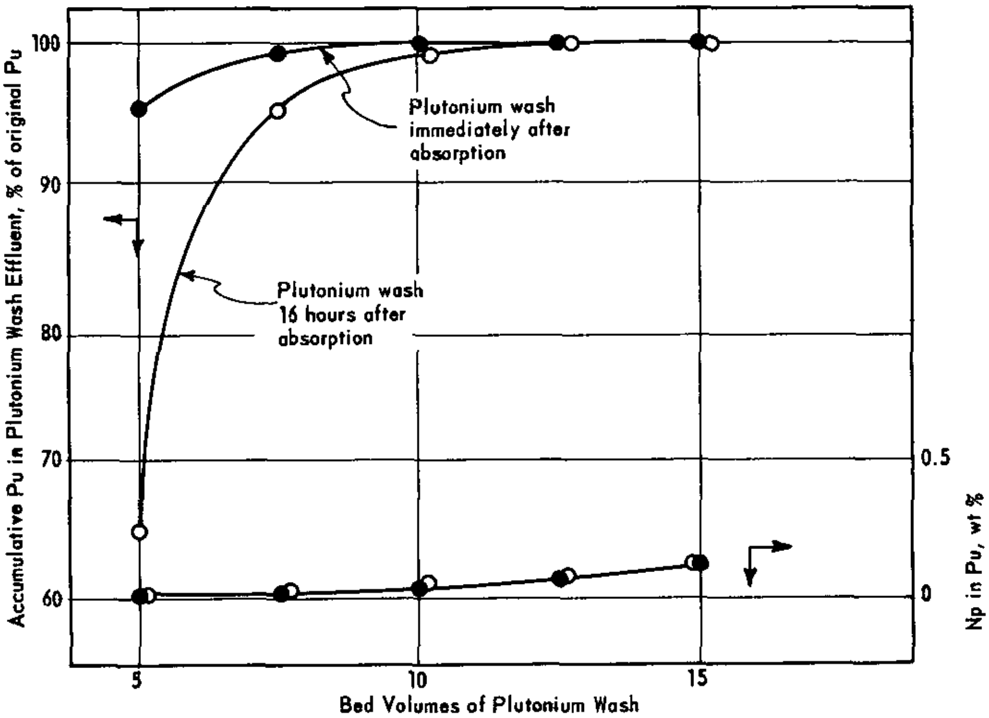
As indicated in the discussion above, primary considerations when Pu^{238} is being separated are the effects of the alpha radiation on the valence of plutonium and neptunium and on the resin. These factors make it necessary to minimize the time that Pu^{238} remains on the resin. In a typical cycle where several grams of Pu^{238} are absorbed per liter of "Dowex" 1 resin, the resin is visibly darkened after one cycle of exposure lasting 10 to 12 hours. After an exposure of approximately $350\,\frac{(\text{grams Pu}^{238})(\text{hours exposure})}{(\text{liters "Dowex" 1 resin})}$ under static conditions, the resin beads begin to form agglomerates, from which it is difficult to elute plutonium and neptunium. Under flow conditions, the agglomeration occurs at a dose of about 1000 g-hr/l.

In the processing of Pu^{239}, alpha radiolysis is no longer the predominant consideration governing exposure time. The main effect is the diffusion of the anionic nitrate species into the resin beads, resulting in less effective separation. The effect is more pronounced for higher crosslinked resins. The effect of a 16-hour delay between absorption of Pu^{239} and the plutonium wash is shown in Figures 10 and 11 for X3 and X4 resins.



Method: II
Resin: 20 ml "Dowex" 1-X4, 40-60 mesh
Temperature: 23°C
Plutonium Wash: 5.7M HNO_3 - 0.05M N_2H_4 - 0.05M $Fe(SO_3NH_2)_2$
Flow Rate: 2 ml/(min)(cm^2)
Load: 332 mg Np, 68 mg Pu^{239}

FIG. 10 EFFECT OF RESIDENCE TIME OF PLUTONIUM ON "DOWEX" 1-X4 RESIN



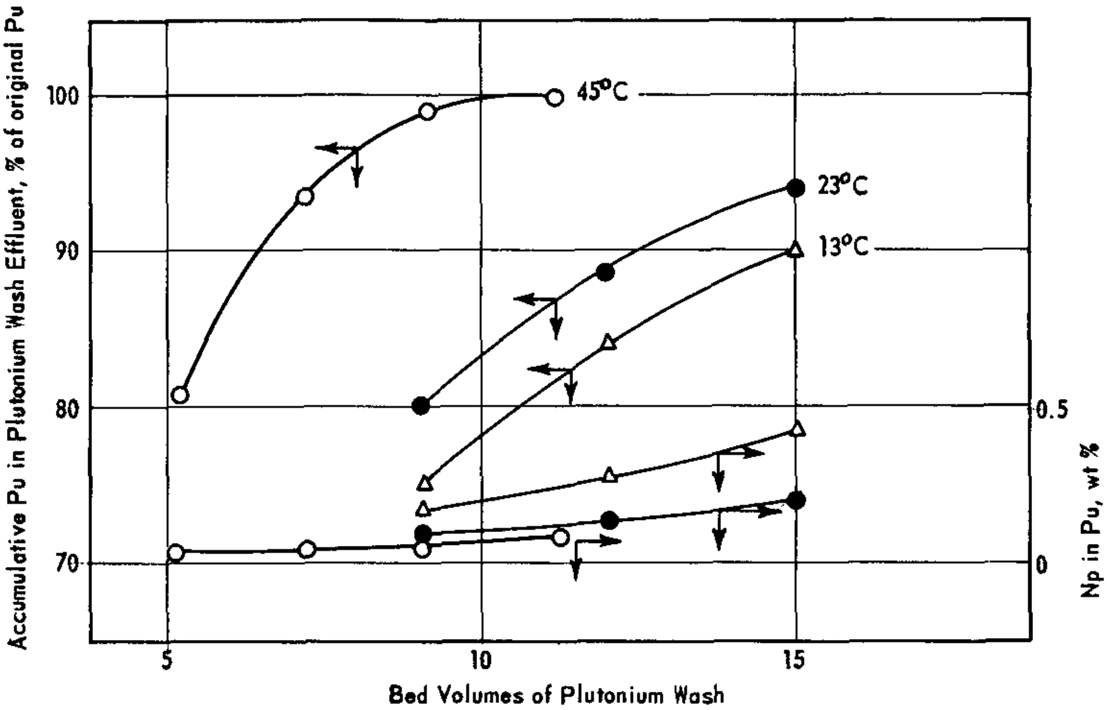
Method: II
Resin: 20 ml "Dowex" 1-X3, 40-60 mesh
Temperature: 23°C
Plutonium Wash: 5.7M HNO_3 - 0.05M N_2H_4 - 0.05M $Fe(SO_3NH_2)_2$
Flow Rate: 2 ml/(min)(cm^2)
Load: 332 mg Np, 68 mg Pu^{239}

FIG. 11 EFFECT OF RESIDENCE TIME OF PLUTONIUM ON "DOWEX" 1-X3 RESIN

TEMPERATURE

The separation of neptunium and plutonium is improved at elevated temperatures. The improvement results from two effects: (1) the distribution coefficient of the Pu(IV) anionic nitrate complex is decreased at higher temperatures; and (2) the diffusion rates in the resin phase are increased.

The effect of temperature on the separation was studied with a jacketed resin bed by circulating water of the desired temperature through the annular jacket. The temperature of the plutonium wash solution in the feed vessel was maintained at 23°C to limit oxidation of the ferrous ion. Runs were made with "Dowex" 1-X4 at 14, 23, and 45°C. The results are shown in Figure 12. It can be seen that with the X4 resin an increase in temperature significantly improved the removal of plutonium, but did not seriously increase the rate of leakage of neptunium from the bed.



Method: II
Resin: 20 ml "Dowex" 1-X4, 40-60 mesh
Plutonium Wash: 5.7M HNO_3 - 0.05M N_2H_4 - 0.05M $Fe(SO_3NH_2)_2$
Flow Rate: 2 ml/(min)(cm^2)
Load: 332 mg Np, 68 mg Pu^{239}

FIG. 12 EFFECT OF TEMPERATURE

TYPE OF ANION EXCHANGE RESIN

Of the resins tested "Dowex" 1-X3 and "Dowex" 1-X2 gave optimum performance in the separation of neptunium and plutonium. Other resins are satisfactory and conditions were devised that allowed acceptable performance with "Dowex" 1-X4, "Dowex" 21K, and "Permutit" SK resins.

The two properties of resins that affect the separation of plutonium and neptunium most significantly are the degree of crosslinkage and the particle size. To some extent higher crosslinkage in the resin can be compensated by the use of a smaller particle size range. For example, when 30-40 mesh resin beads are used, "Dowex" 1-X3 is satisfactory. The same size range of X4 resin gives poorer separation, but the use of 50-60 mesh or 60-80 mesh resin improves the separation. This effect of particle size is shown in Table VII. For these tests, particle size ranges were screened from a single batch of "Dowex" 1-X4 resin.

TABLE VII

Effect of Particle Size on Separation

Method: II
Resin: "Dowex" 1-X4
Column: 0.62-cm^2 area, 12.6-inch height
Plutonium Wash: 15 bed volumes of 5.8M HNO_3 - 0.05M $Fe(SO_3NH_2)_2$ - 0.05M N_2H_4
Flow: 2 ml/(min)(cm^2)

Mesh Range	Pu^{239} in Np, wt %	Np in Pu^{239}, wt %
30-40	2.2	0.1
40-50	1.7	0.1
50-60	1.1	0.1

The efficiency of separation is very sensitive to the degree of crosslinkage of the polymer, particularly in the 2 to 4% range. The general insensitivity of the separation to process variables when using X3 resin as compared with X4 resin has been referred to in a number of other sections of this report. The lower crosslinked resins give better separation, because of improved diffusion rates, and allow the use of coarser particles with the attendant decrease in pressure drop across the bed. These gains must be balanced against the less desirable physical properties of the lower crosslinked resins: the greater shrinking and swelling, the lowered physical strength, and the decreased radiation resistance. The effect of crosslinkage on the separation was difficult to evaluate quantitatively because different lots of "Dowex" resin of the same divinylbenzene content apparently varied significantly in the actual amount of crosslinkage. Typical curves shown in Figure 13 may be compared for samples of "Dowex" 1 that were nominally 2, 3, 4, and 8% crosslinked. All of the resin samples were 40-60 mesh, with the exception of the X8 resin which was 50-100 mesh. The crosscontamination of the products in the runs shown are summarized in Table VIII.

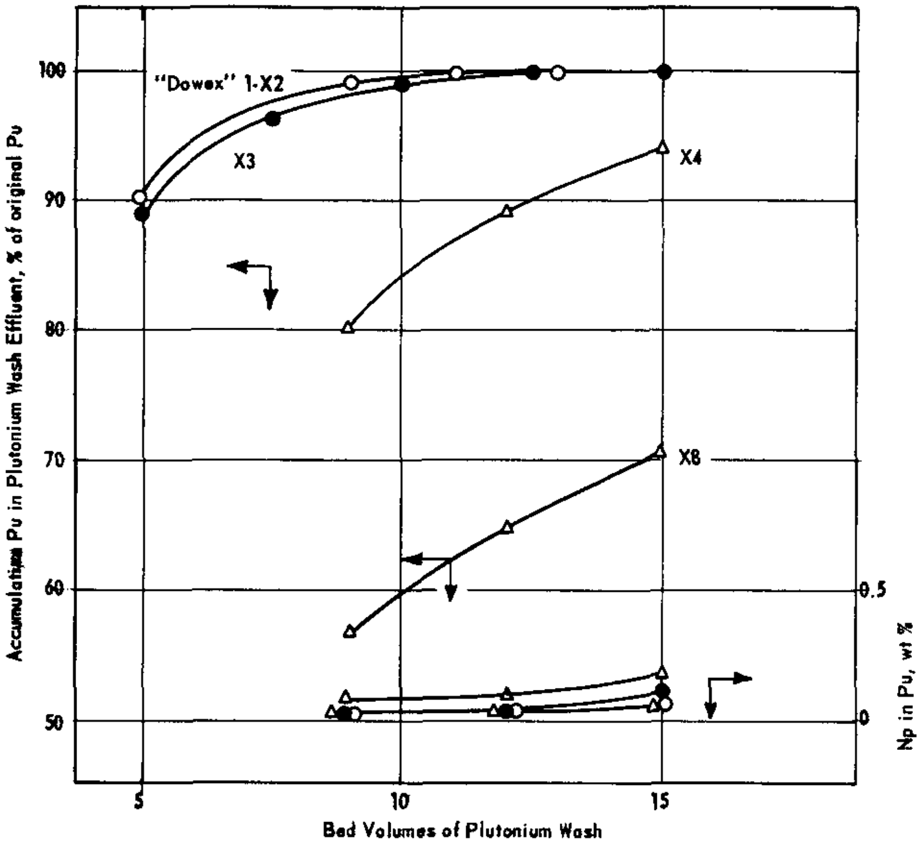
TABLE VIII

Effect of Crosslinkage of Resin

(Details of the experimental runs as given in Figure 13)

Crosslinkage	Mesh Range	Pu^{239} in Np, wt %	Np in Pu^{239}, wt %
X2	40-60	0.02	0.1
X3	40-60	0.01	0.1
X4	40-60	1.1	0.2
X8	50-100	7.0	0.1

Excellent separation under conditions comparable to those used for "Dowex" 1-X3 was obtained with "Dowex" 21K resin. The decontamination from fission products with this resin, however, was not as good as with the "Dowex" 1 resins.



Method: II
Resin Bed: 0.62-cm^2 area, 12.6-inch height
Temperature: 23°C
Plutonium Wash: 5.7M HNO_3 - 0.05M N_2H_4 - 0.05M $Fe(SO_3NH_2)_2$
Flow Rate: 2 ml/(min)(cm^2)
Load: 320 mg Np^{237}, 70 mg Pu^{239}

FIG. 13 EFFECT OF RESIN CROSSLINKAGE

"Permutit" SK resin has the advantage of greater stability to alpha and gamma radiation. The diffusion rates are lower in this resin than in the "Dowex" resins; but, as discussed in the sections on the effects of acid concentration and flow conditions, satisfactory separation can be obtained at decreased flow rates. The decontamination from fission products with "Permutit" resin is less than with "Dowex" 1 resin. The relative abilities of "Dowex" 1-X4 and "Permutit" SK resins to remove fission products from product streams were tested as follows. Typical process concentrations of tracer solutions of Ru^{103}, Zr^{95}-Nb^{95}, Cs^{137}, and Ce^{144} were individually added to separate solutions of 8M HNO_3, and the solutions were made 0.05M in $Fe(NH_2SO_3)_2$ and heated at 55°C for 30 minutes. No actinide elements were used in these tests. Each adjusted solution was divided into two portions and fed to 5-ml beds of fresh "Permutit" SK and "Dowex" 1-X4. After the absorption step, the resin was washed with 30 bed volumes of 0.005M HF in 8M HNO_3, then eluted with 6 bed volumes of 0.35M HNO_3.

The activity in the eluate was compared with the activity in the feed to obtain a "decontamination factor". The results in Table IX show that the "Dowex" resin gave higher decontamination factors by factors of approximately 2 for Zr^{95}-Nb^{95}, 4 for Ru^{103}, and 100 for Ce^{144}. Similar tests were used to show that the "Dowex" 1 resin was superior in decontamination performance to "Dowex" 21K, "Dowex" 2-X4, "Dowex" 3, and "Duolite" A101D.

TABLE IX

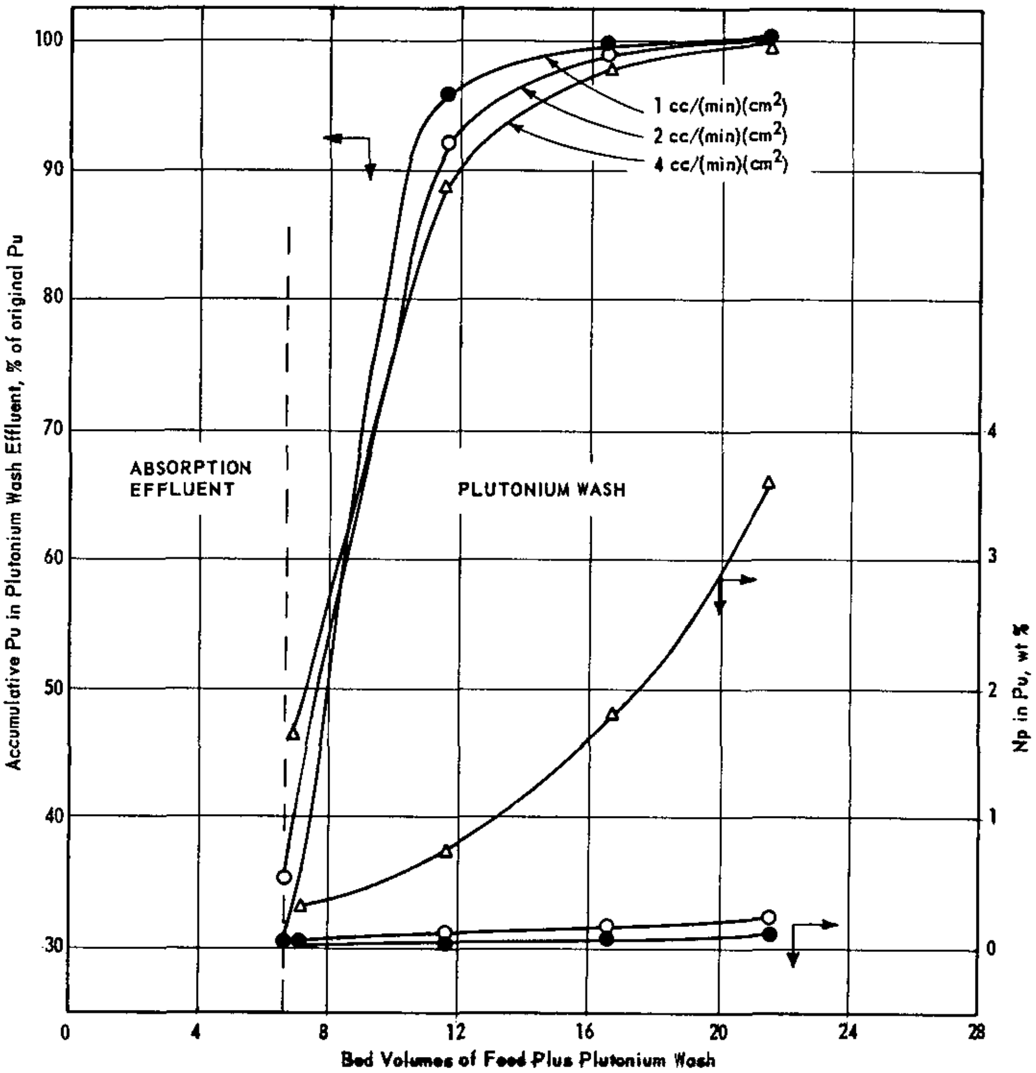
Removal of Fission Products by Anion Exchange

Nuclide	Conditions	Decontamination Factors "Permutit" SK	Decontamination Factors "Dowex" 1-X4
Ru^{103}	Fresh resin	700	2,700
Zr^{95}-Nb^{95}	Fresh resin	2,000	3,800
	Used resin	1,000	1,500
Cs^{137}	Fresh resin	>5,000	>5,000
Ce^{144}	Fresh resin	200	20,000

FLOW CONDITIONS

The effect of flow conditions on separation is interrelated with several variables including the method of operation; the type, particle size, and crosslinkage of the resin; and the total actinide load. When the flow is too rapid, the crosscontamination of both the neptunium and plutonium products increases.

With Method I, in which plutonium is present in the (III) state in the feed, the amount of plutonium absorbed on the resin increases as feed flow through the column is decreased. This effect is shown by the initial points on the curves in Figure 14. With either "Dowex" 1-X4, 50-100 mesh resin or with "Permutit" SK, 40-80 mesh resin this effect is compensated by the more efficient removal of the plutonium with a slow flow for the plutonium wash. In addition, the slower flow elutes less neptunium, so that the plutonium product contains less neptunium. These effects are illustrated in Figure 14 for "Dowex" and in Figure 15 for "Permutit". For an average loading of 16 mg of Np and 3.5 mg of Pu per ml of resin, a maximum flow of 2 ml/(min)(cm^2) is recommended when using "Dowex" 1-X4, 50-100 mesh resin and 1.7 ml/(min)(cm^2) when using "Permutit" SK, 40-80 mesh resin.



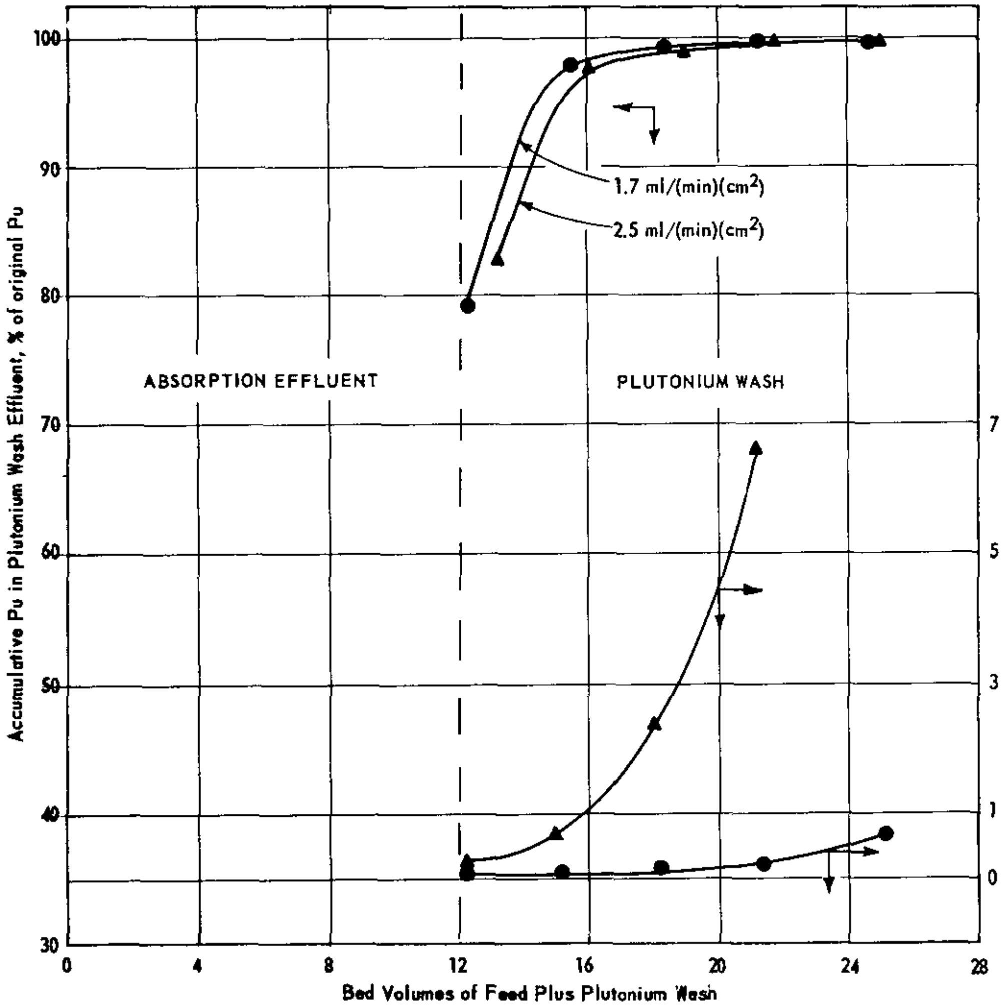
Resin: 20 ml "Dowex" 1-X4, 50-100 mesh

Temperature: 23°C

Feed and Wash: 6M HNO_3 - 0.1M N_2H_4 - 0.05M $Fe(SO_3NH_2)_2$
3.2 mg Np/ml - 0.48 mg Pu/ml

Load: 420 mg Np, 62 mg Pu^{239}

FIG. 14 EFFECT OF FLOW RATE WITH METHOD I AND "DOWEX" 1-X4 RESIN



Resin: 20 ml "Permutit" SK, 40-80 mesh
Temperature: 23°C
Feed and Wash: 6.4M HNO_3 - 0.05M N_2H_4 - 0.05M $Fe(SO_3NH_2)_2$
Load: 410 mg Np, 62 mg Pu^{239}

FIG. 15 EFFECT OF FLOW RATE WITH METHOD I AND "PERMUTIT" SK RESIN

With Method II, all the plutonium is absorbed; thus, more efficient plutonium wash conditions are required. With "Dowex" 1-X4, 40-60 mesh resin the plutonium wash flow must be reduced to about 1.3 ml/(min)(cm^2) to attain 99% removal of the plutonium with 15 bed volumes of 5.7M HNO_3 - 0.05M N_2H_4 - 0.05M $Fe(NH_2SO_3)_2$. The data are given in Figure 16. Reducing the resin crosslinkage to 3% eliminates much of the sensitivity toward flows, as shown in Figure 17. By using "Dowex" 1-X3 resin in the recycle technique, Method III, a flow of 3 ml/(min)(cm^2) of plutonium wash solution can be tolerated.

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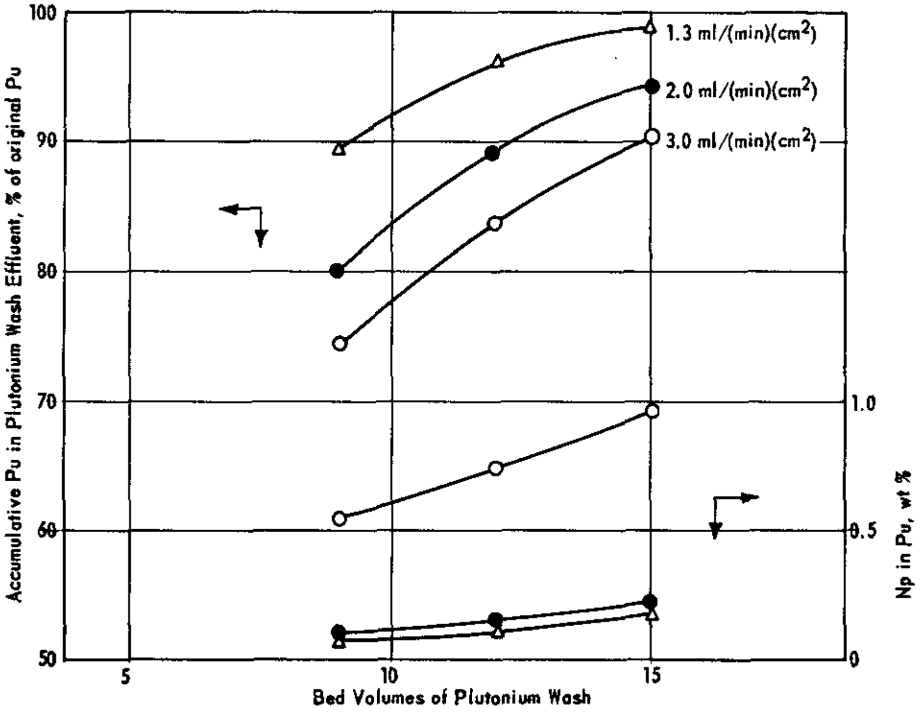
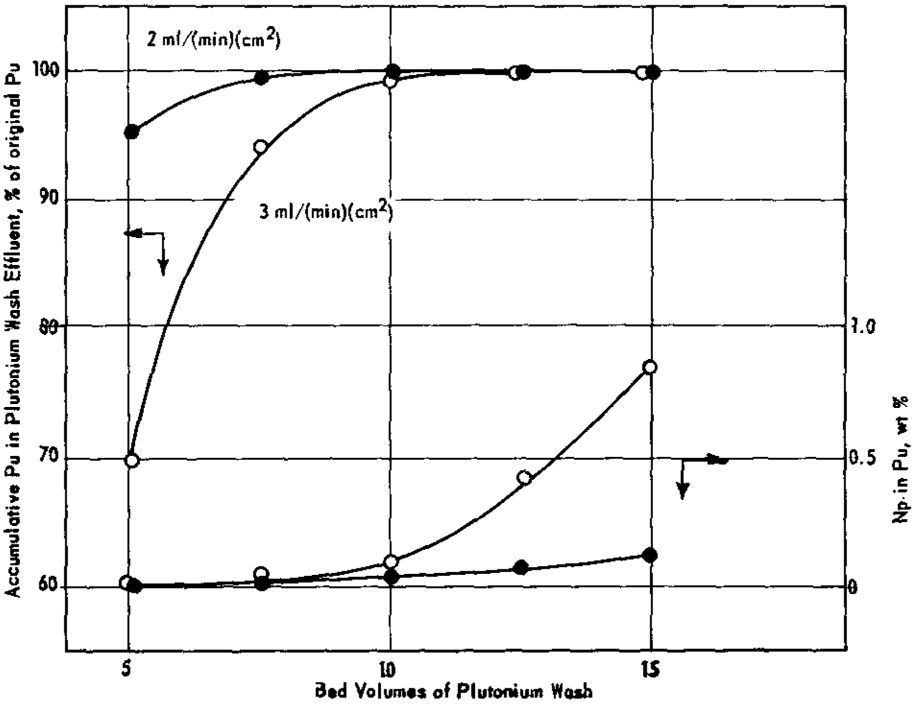


Resin: 20 ml "Dowex" 1-X4, 40-60 mesh
Temperature: 23°C
Plutonium Wash: 5.7M HNO_3 - 0.05M N_2H_4 - 0.05M $Fe(SO_3NH_2)_2$
Load: 332 mg Np, 68 mg Pu^{239}

FIG. 16 EFFECT OF FLOW RATE WITH METHOD II AND "DOWEX" 1-X4 RESIN



Resin: 20 ml "Dowex" 1-X3, 40-60 mesh
Temperature: 23°C
Plutonium Wash: 5.7M HNO_3 - 0.05M N_2H_4 - 0.05M $Fe(SO_3NH_2)_2$
Load: 332 mg Np, 68 mg Pu^{239}

FIG. 17 EFFECT OF FLOW RATE WITH METHOD II AND "DOWEX" 1-X3 RESIN

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