ORNL/TM-7991
Dist. Category UC-86
(Applied)

Contract No. W-7405-eng-26

ORNL/TM--7991

DE82 014768

Consolidated Fuel Reprocessing Program

SOLVENT EXTRACTION STUDIES OF COPROCESSING FLOWSHEETS — RESULTS FROM CAMPAIGNS 3 AND 4 OF THE SOLVENT EXTRACTION TEST FACILITY (SETF)

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Date Published - May 1982

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ABSTRACT

Experiments on tri-*n*-butyl phosphate solvent extraction of uranium and plutonium at full activity levels (Campaigns 3 and 4) were conducted in the Solvent Extraction Test Facility (SETF), located in one of the heavily shielded cells of the Transuranium Processing Plant. The primary objectives were (1) to demonstrate and evaluate the first two cycles of the Hot Engineering Facility flowsheets (codecontamination and partial partitioning), and (2) to investigate and evaluate the use of HNO_2 as the reductant for tetravalent plutonium during reductive stripping operations. Secondary objectives were to determine the solvent extraction behavior of feed solutions prepared by dissolving fuel from a boiling water reactor (BWR) and to improve the solvent extraction feed clarification.

1. INTRODUCTION

The Solvent Extraction Test Facility (SETF) is located in one of the heavily shielded cells of the Transuranium Processing Plant (TRU) and is used to evaluate, at full activity levels, recently developed or improved flowsheets for reprocessing commercial nuclear power reactor fuels. Emphasis is placed on fast breeder reactor (FBR) fuels. The broad objective of studying uranium-plutonium coprocessing flowsheets, begun during Campaigns 1 and 2 in the SETF,[1] was continued during Campaigns 3 and 4. Specifically, the primary objectives of the work done during Campaigns 3 and 4 were (1) to demonstrate and evaluate the first two cycles of the Hot

Engineering Facility (HEF) flowsheets (codecontamination and partial partitioning), and (2) to investigate and evaluate the use of HNO_2 as the reducing agent for Pu(IV) during the reductive stripping process. Secondary objectives were to compare the solvent extraction behavior of boiling water reactor (BWR) and pressurized water reactor (PWR) fuels and to improve the solvent extraction feed clarification.

Campaigns 3 and 4 were carried out during the period October 1979—July 1980. Prior to that time, emphasis in the program had shifted from the development of methods for reprocessing light water reactor (LWR) fuels to the development of methods for reprocessing fast breeder reactor (FBR) fuels. However, it was recognized that a demonstration facility such as the HEF would probably need the capability for handling both types of fuels. Since no FBR fuels were available during 1979 and 1980, LWR fuels from the H. B. Robinson-2 pressurized water reactor (PWR) and the Dresden-1 BWR were used for the tests made in Campaigns 3 and 4. However, several of the tests were made with a simulated "FBR-type" feed solution formulated from the LWR fuel and recycled plutonium.

The Robinson-2 fuel had been irradiated to a peak burnup of ~2.7 TJ/kg (~31,000 MWd/metric ton) and an average burnup of ~2.4 TJ/kg (~28,000 Mwd/metric ton); it had been discharged from the reactor on May 6, 1974. The Dresden-1 fuel had been irradiated to a burnup of 2.05 TJ/kg (23,780 MWd/metric ton) and had been discharged on September 1, 1975. Since both fuels were discharged ~5 years before they were processed, the ^{95}Zr-Nb fission products had decayed to such an extent that decontamination measurements for these nuclides could not be made.

2. EQUIPMENT AND DESCRIPTION

The SETF equipment items used during Campaigns 3 and 4 for fuel dissolution, feed adjustment, and solvent extraction were the same as those described for Campaigns 1 and 2.[1] Use of precoated, etched-disc filters (with either 1-μm or 3-μm openings) for feed clarification was continued. In addition, two other types of filters were employed in Campaigns 3 and 4. A fritted stainless steel filter, rated at 0.5 μm and manufactured by the Mott Metallurgical Corporation, was used for polishing previously clarified feed solutions during two tests, and a "deep bed" type of filter containing a bed of diatomaceous earth as the filtering medium was used for the primary clarification during several tests. The features of the "deep-bed" filter are compared with those of the etched-disc filter in Fig. 1. Each filter was housed in an ~10-cm-diam glass pipe, ~15 cm long. The diatomaceous earth used to precoat the etched-disc filter was ~5 g of Celite-535 covered by ~1 g of Celite Hi Flo—Super Cel, both manufactured by the Johns Manville Company. About 100 g of Celite-535 and 25 g of Hi Flo—Super Cel were used in the deep-bed filter.

3. OPERATING PROCEDURE

The procedures used for dissolution of the irradiated fuels and for the solvent extraction tests were the same as those previously described.[1] The feed adjustment and clarification procedures were modified to permit (1) clarification of the dissolver solution, and (2) addition of H_2O_2 to the clarified solution during a 1-h digestion period at 95°C prior to the

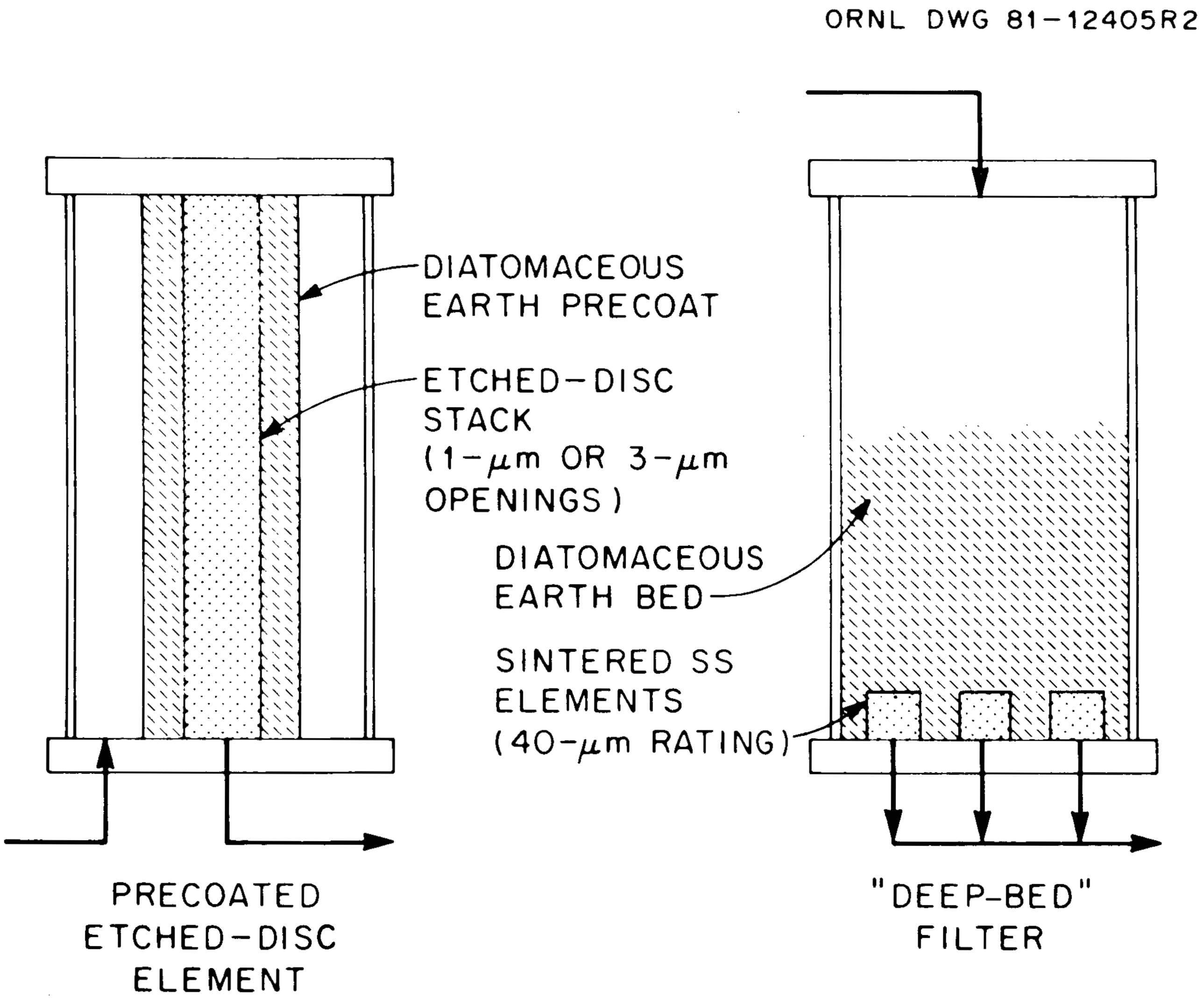

Fig. 1. SETF filters.

regular feed adjustment steps. The purpose of the H_2O_2 addition—digestion step was to enhance dissolution of insoluble colloids that had passed through the filter and, thereby, to prevent or diminish the subsequent formation of colloid-stabilized emulsions (cruds) within the extraction bank mixer-settlers. The inclusion of these modifications had been found to be beneficial during tests made in Campaign 2.[1]

4. EXPERIMENTAL CONDITIONS, RESULTS, AND DISCUSSION

In the HEF codecontamination-cycle flowsheet, uranium and plutonium are separated from the fission products but not from each other, whereas in the partial partitioning-cycle flowsheet, part of the uranium is separated from the plutonium to produce a typical uranium product plus a plutonium-uranium product which is satisfactory for use in FBR core fuel. The codecontamination and partial partitioning cycles of the HEF flowsheets were demonstrated and evaluated while processing a simulated FBR-type feed solution in SETF test 2-4 and an LWR-type feed solution in test 3-1. (The results from test 2-4 are discussed in this report because they are pertinent to the evaluation of the HEF flowsheets.)

The use of HNO_2 as the reducing agent for Pu(IV) during reductive stripping operations was investigated in tests 4-2 and 4-3, and its use in a potentially attractive flowsheet was demonstrated in tests 3-2 (for processing LWR-type feed solutions) and 3-4 (for processing FBR-type feed solutions). The solvent extraction behavior of BWR-type feed solutions was determined in tests 4-1, 4-2, and 4-3. Organic solvents used in tests 3-3 and 4-3 contained 15% (vol) tri-*n*-butyl phosphate (TBP), while the TBP concentration was 30% (vol) in the other tests. In all cases, the organic diluent was normal paraffin hydrocarbon (NPH). Operating conditions and detailed stream analyses for each of the solvent extraction tests are tabulated in the Appendix.

4.1 Demonstration and Evaluation of HEF Flowsheets

A schematic diagram of the HEF flowsheets for codecontamination and partial partitioning is shown in Fig. 2. Features of the codecontamination flowsheet include the use of a split scrub stream and a split strip stream. The split scrubs, one of high acidity (HAIS) and one of low acidity (HAS), are designed to obtain a high decontamination factor (DF) for both ruthenium and zirconium. The split strip streams are designed to effect complete stripping of plutonium into an aqueous solution (HCIX plus HCX) having a minimum acidity of 0.3 M HNO_3 and to enable complete stripping of uranium by means of an ~0.01 M HNO_3 stream (HCX). Features of the partitioning flowsheet include (1) reduction of plutonium to the trivalent state by means of hydrazine-stabilized hydroxylamine nitrate (HAN) in a continuous plug-flow (nonbackmixing) reactor (simulated by a batch reactor in the SETF), (2) reacidification of the solution to extraction conditions for uranium, and (3) selective extraction of enough uranium to effect the desired partitioning.

4.1.1 Codecontamination cycle

The operating conditions used in the two codecontamination cycle tests are shown in Table 1.

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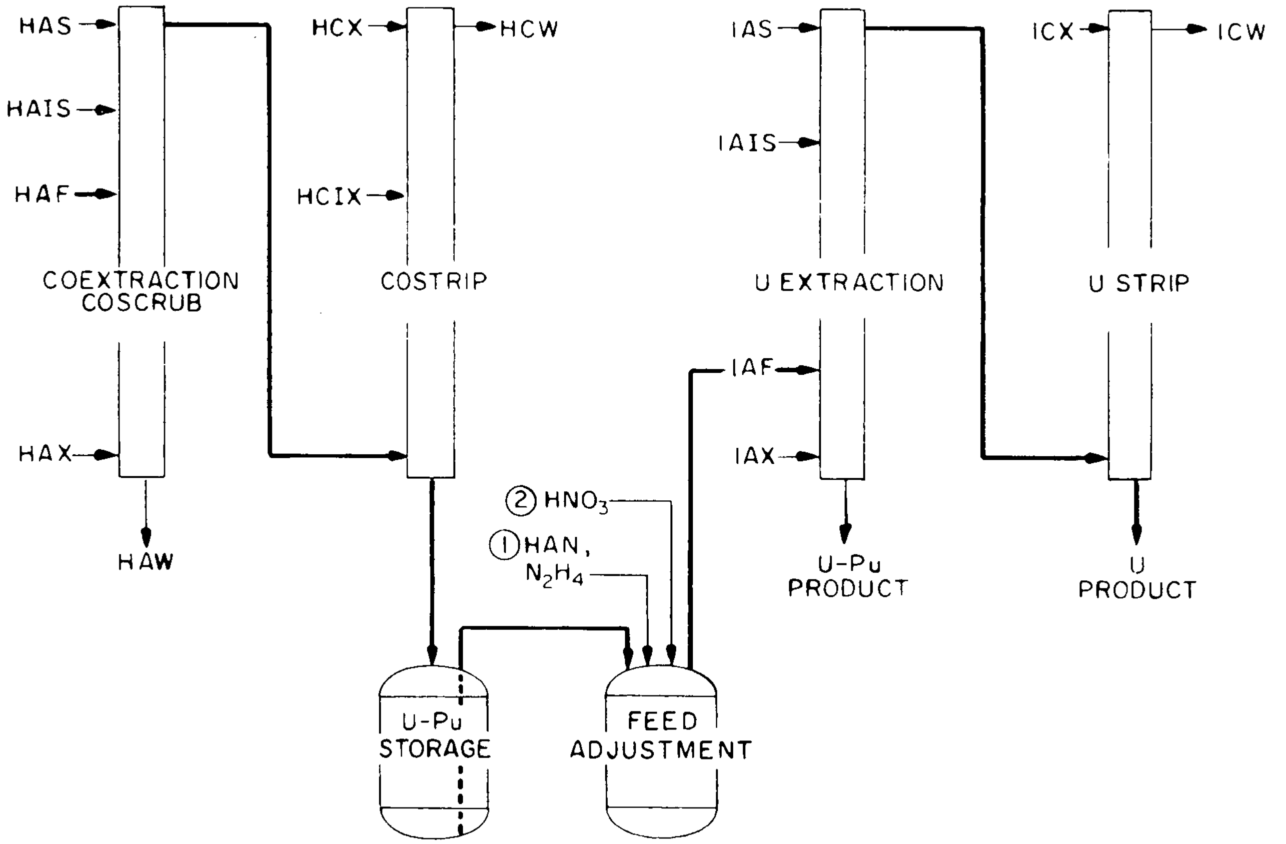

Fig. 2. Flows for codecontamination and partitioning.

Table 1. Conditions used in codecontamination tests

Mixer-settler sections	Number of stages	Temperature (°C)
Extraction	10	a
Scrub/intermediate scrub	3/3	a
Low-acid/high-acid strip	b	50

Stream	Flow ratios Test 2-4	Flow ratios Test 3-1	Solution composition
HAF	1.0	1.0	c
HAX	1.93	2.00	30% TBP in NPH
HAS	0.26	0.23	0.8 M HNO_3
HAIS	0.14	0.13	3.0 M HNO_3
HCX	2.15	2.06	d
HCIX	0.20	0.20	3.1 M HNO_3

[a] The temperature of the extraction/scrub bank was 40°C during test 2-4 and 52°C during test 3-1.

[b] Low-acid/high-acid strip stages were 2/14 during test 2-4 and 6/10 during test 3-1.

[c] HAF composition was 3.3 M HNO_3--144 g/L U--13.5 g/L Pu during test 2-4 and 2.9 M HNO_3--156 g/L U--1.4 g/L Pu during test 3-1.

[d] HCX composition was 0.007 M HNO_3--0.002 M HAN--0.0005 M N_2H_4 during test 2-4 and 0.05 M HNO_3 during test 3-1.

The HAF solution used in test 2-4 was prepared by dissolving LWR fuel containing ~2 kg of heavy metal and then adding plutonium that had been recovered in previous SETF runs. The uranium and plutonium concentrations in the HAF solution were adjusted to 144 g/L and 13.5 g/L, respectively, because these concentrations are similar to those expected during FBR fuel reprocessing. In test 3-1, the HAF solution was formulated only from freshly dissolved LWR fuel and was adjusted to essentially the same total heavy-metal concentration (uranium, 156 g/L; plutonium, 1.4 g/L) as for test 2-4.

Besides the different plutonium concentrations in the feed solutions, one other flowsheet condition differed in the extraction/scrub bank during the two tests. The temperature was 40°C during test 2-4 and 52°C during test 3-1. The higher temperature in test 3-1 apparently improved decontamination from the fission products as shown in Table 2. In both tests, the losses of uranium and plutonium to the raffinate (HAW) stream were very low.

Table 2. Results from codecontamination tests

	Test No.	
	2-4	3-1
Extraction losses, %		
Uranium	<9.7E-4	3.3E-2
Plutonium	6.1E-4	7.1E-3
DFs from:		
^{106}Ru (extraction/scrub)	3.3E3	5.6E3
^{106}Ru (overall)	3.6E3	1.5E4
^{137}Cs (overall)	2.1E5	5.4E5
^{144}Ce (overall)	2.4E5	>6E4

In the two tests, two costripping conditions were different. The low-acidity strip solution (HCX) used in test 3-1 had a higher acid concentration (0.05 <u>M</u> HNO_3) than that in test 2-4 (0.007 <u>M</u> HNO_3) and contained no reductants. Also, six low-acid stripping stages were used in test 3-1, while only two were used in test 2-4. The costripping results, which are illustrated by the uranium and plutonium concentration profiles given in Fig. 3, show that the conditions used in test 2-4 were better than those

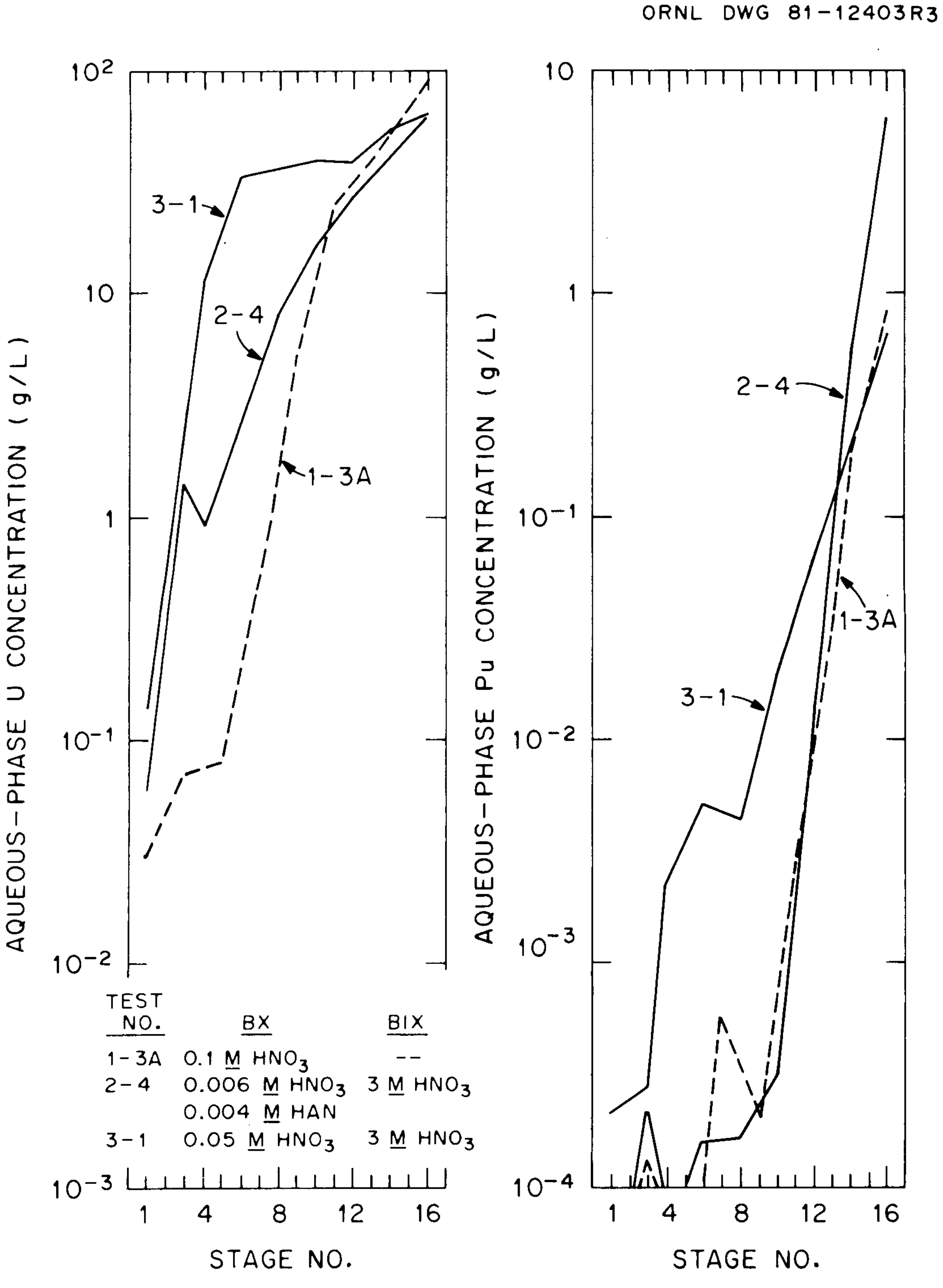

Fig. 3. Uranium and plutonium profiles in the costripping bank.

used in test 3-1 for both uranium and plutonium stripping. The plutonium concentration profile from test 2-4 was similar to that obtained in a previous test (1-3A), in which only a single strip stream (0.10 M HNO_3, without reductant) had been used at a similar flow ratio but at a lower temperature (40°C); however, the uranium stripping in test 2-4 was less effective than that in test 1-3A.

4.1.2 Partial partitioning cycle

After each of the codecontamination tests, the aqueous product solution was batch-treated (to simulate the HEF continuous plug-flow reactor) for Pu(IV) reduction followed by acid adjustment to a concentration necessary for efficient uranium extraction. After this treatment, the solution from test 2-4 was divided into two batches and that from test 3-1 was divided into three batches, so that several of the process parameters could be tested in the subsequent selective uranium extraction test runs.

The base-line feed adjustment treatment consisted of (1) addition of the plutonium reductant, HAN, and the stabilizing agent, hydrazine (the HAN/N_2H_4/Pu mole ratio was 4/0.27/1); (2) digestion at 40°C for 2 h; and (3) acidification to 1.3 M HNO_3. In both tests, the portion of plutonium remaining in the tetravalent state after the reduction step and after the acidification step ranged, respectively, from 0.7 to 1.3% (wt) and from 2 to 4% (wt).

The base-line conditions used in the two tests (see Table 3) were designed to enable partitioning of enough uranium to produce a uranium-plutonium product in which the plutonium represented 35% (wt) of the heavy metal (U + Pu). The conditions for the two tests differed because the

Table 3. Conditions used in second cycle (partitioning) tests

	Mixer-settler bank No. of stages Test 2-4	Test 3-1	Temperature (°C)
Scrub/intermediate scrub/extraction bank	2/6/8	4/8/4	25-30
Stripping bank	16	16	50

Stream	Flow ratios Test 2-4	Test 3-1	Solution composition
1AF	1.0	1.0	1.25 $\underline{M}$ HNO_3—48 g/L of heavy metal[a]
1AX	0.37	0.68	30% TBP in NPH
1AS	0.12	0.11	0.1 $\underline{M}$ HNO_3—0.02 $\underline{M}$ HAN—0.01 $\underline{M}$ N_2H_4
1AIS	0.055	0.064	3 $\underline{M}$ HNO_3
1CX	0.34	0.51	0.01 $\underline{M}$ HNO_3

[a] Comprised of 43 g/L of uranium plus 4.5 g/L of plutonium in test 2-4 and 48 g/L of uranium plus 0.46 g/L of plutonium in test 3-1. Feeds for base-line tests also contained HAN and N_2H_4, which were added during the feed adjustment at a HAN/N_2H_4/Pu mole ratio of 4/0.27/1.

plutonium concentration in test 3-1 had to be increased by a factor of 37 (from 0.95 to 35 wt %), whereas that in test 2-4 required a concentration factor (CF) of only 3.7 (from 9.5 to 35 wt %). The results of the two tests (summarized in Table 4) showed that, although the intended plutonium enrichment was excessive, the separations obtained were similar.

Table 4. Results of partitioning cycle tests with synthetic FBR and LWR fuel solutions

Test	Pu in U-Pu product (wt % of heavy metal)	Pu in uranium product Concentration (ppm)	Percent of feed Pu
2-4 (synthetic FBR)	59	6.9	3.7E-4
3-1 (LWR)	58	4.3	3.0E-2

During test 2-4, the effect of increasing the extraction/scrub bank temperature from 26°C to 50°C was to decrease the separation of uranium and plutonium slightly as shown in Table 5. Uranium, plutonium, and HNO_3 concentration profiles during operation at the two temperatures are shown in Fig. 4. The plutonium concentration profile indicates that a small amount of plutonium refluxing occurred while the bank was at 50°C.

Table 5. Results of partitioning at various conditions

Run No.	Conditions	Pu in U-Pu product (wt %)	Pu in uranium product (ppm)
2-4B	Base line[a]	59	6.9
2-4C	Extraction/scrub bank (50°C)	47	11.3
3-1C	Base line[a]	58	4.3
3-1B	No N_2H_4 in feed or scrub	51	27.0
3-1D	3 $\underline{M}$ HNO_3 in feed	~0	b

[a] Described in Table 3.

[b] All plutonium in the feed was reoxidized and extracted along with the uranium. Most of the extracted plutonium refluxed in the scrub section, causing the test to be aborted.

Different feed adjustment conditions were evaluated during test 3-1. Run 3-1B was made without hydrazine in the feed (1AF) or in the scrub (1AS) stream. Following the feed adjustment, the portion of the plutonium remaining in the tetravalent state was the same (4%) as after the base-line feed adjustment (with hydrazine). However, during the mixer-settler operation, the extracted plutonium was not effectively scrubbed and the

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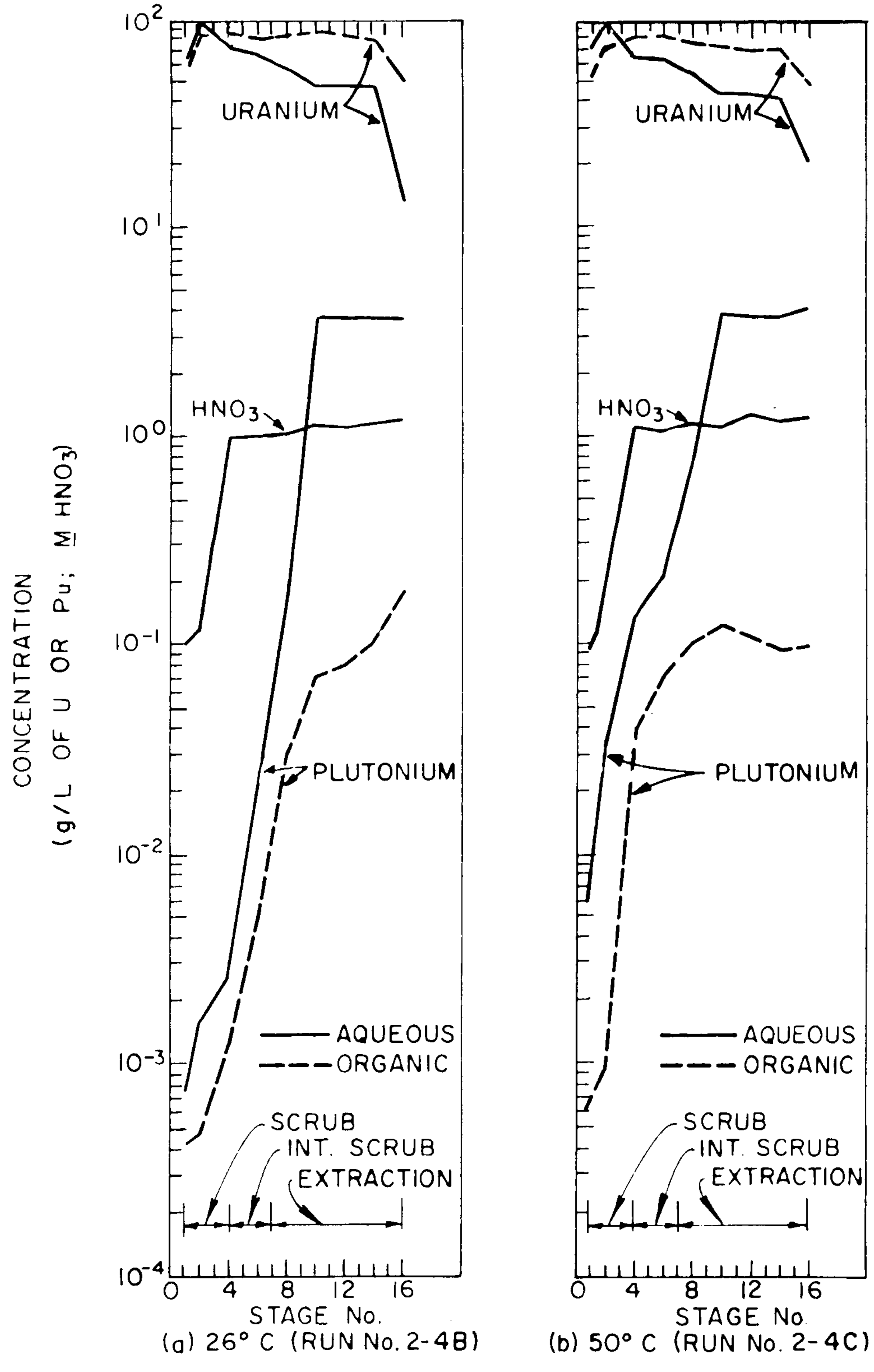

Fig. 4. Concentration profiles during selective uranium extraction test 2-4.

plutonium concentration in the uranium product was significantly increased (from 4 ppm to 27 ppm) as shown in Table 5. The use of hydrazine was beneficial in the selective uranium extraction run (3-1C) because the aqueous-phase acidity in the contactor was ~1.5 M HNO_3. Previous studies[1] have indicated that unstabilized HAN (without hydrazine) can be used effectively for the reduction of Pu(IV) only when the aqueous-phase acidity is kept below 1 M HNO_3, preferably less than 0.5 M HNO_3.

During test run 3-1D, hydrazine-stabilized HAN was used (as in the base-line feed adjustment procedure); however, after the Pu(IV) in the feed batch had been reduced, the solution was acidified to 3 M HNO_3. At this acidity, the reductants were rapidly destroyed (apparently by radiolytically produced nitrous acid) and the Pu(III) was oxidized. The solvent extraction run was aborted before steady state was reached because of extensive refluxing of the plutonium. Before the shutdown, stage samples were taken to substantiate the refluxing, as shown in Fig. 5.

4.2 Use of HNO_2 for Reductive Stripping of Plutonium

The principle of using HNO_2 for reductive stripping of plutonium was proposed by Bathellier,[2] and a process for its application in costripping plutonium and uranium was devised later by Tsujino et al.[3] The process is based on the principle that the redox potential of Pu(III)/Pu(IV) in the HNO_3—HNO_2 system is such that HNO_2 reduces Pu(IV) to Pu(III) at acidities below ~0.5 M HNO_3. It is well known that HNO_2 oxidizes Pu(III) to Pu(IV) at higher acidities. That property, together with the fact that HNO_2 is continuously generated in HNO_3 solutions containing plutonium (by radiolysis

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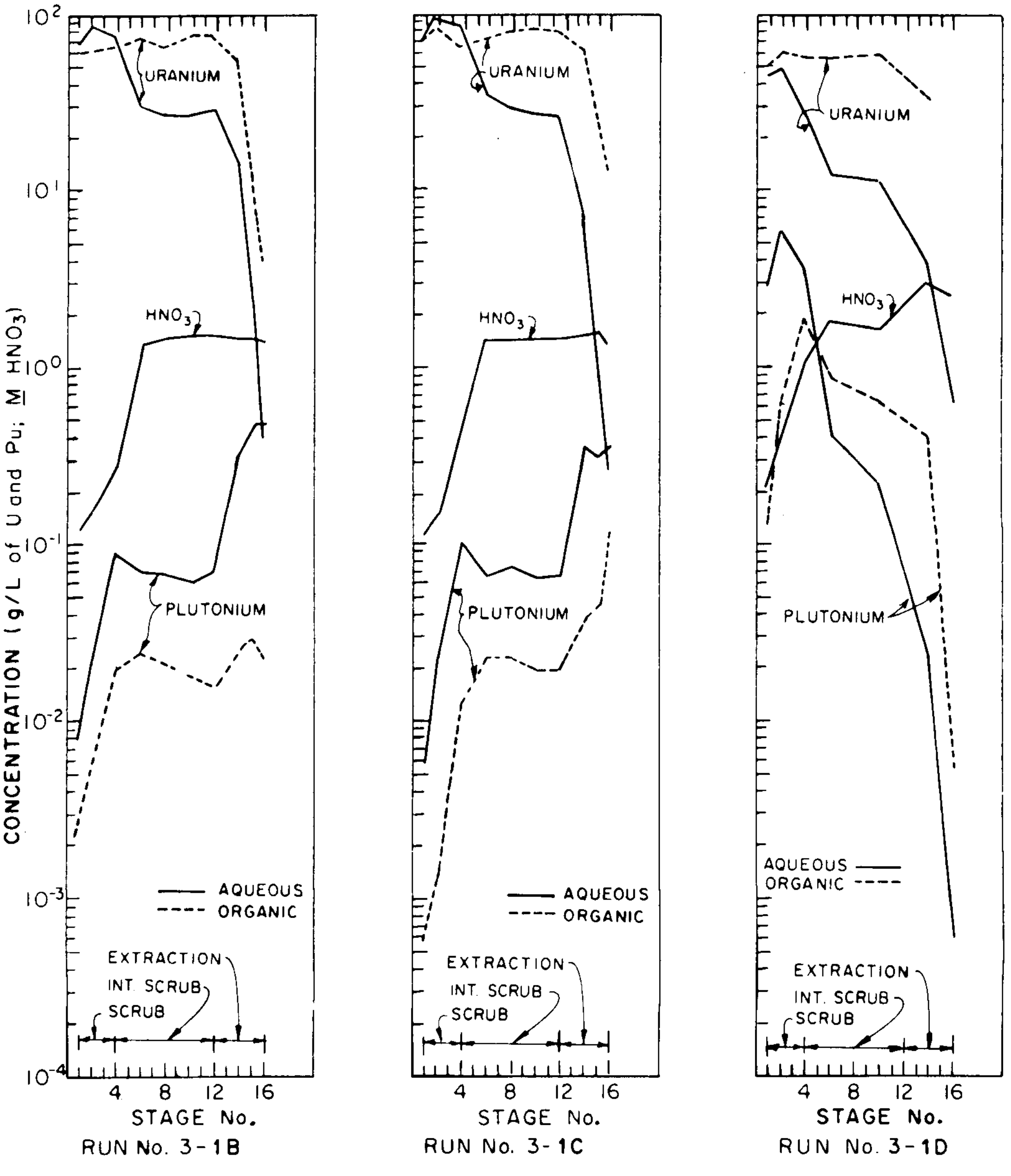

Fig. 5. Concentration profiles during selective uranium extraction test 3-1.

of HNO_3) has indicated the need for holding reductants in HNO_3 solutions containing Pu(III).

The use of HNO_2 as a reducing agent for plutonium in partial partitioning operations was investigated during SETF Campaign 2. The investigation was continued during Campaigns 3 and 4 because the initial results were encouraging (plutonium strip yields of up to 99% were obtained with plutonium concentration factors of 5 to 10) and because the process has several attractive features, such as:

1. When HNO_2 is added to the HNO_3/TBP/Pu system, it introduces no new chemicals that could create hazardous emissions or solids in the waste streams. Added HNO_2 only supplements that already being generated.
2. The HNO_2 can be easily added to the system by dissolving it in the organic inlet stream, since the solubility of HNO_2 in TBP/NPH is relatively high. The $TBP\cdot HNO_2$ addition product has been successfully prepared by bubbling NO gas into nitric acid which was in contact with the organic solvent (TBP/NPH). Nitrous acid is formed and extracted according to the reactions:

$$2NO + HNO_3 + H_2O \rightleftharpoons 3HNO_2, \qquad (1)$$

$$HNO_{2aq} + TBP_{org} \rightleftharpoons TBP\cdot HNO_{2org}. \qquad (2)$$

3. Excess HNO_2 can be easily removed from the waste organic by venting and purging to remove NO gas as the HNO_2 decomposes according to the reverse of reaction (1).

The characteristics previously found to be necessary for satisfactory use of the process do not appear to be difficult and can be described as follows:

1. Stripping works best at lower temperatures (i.e., the use of ambient temperatures of 25-35°C has given satisfactory results).
2. The plutonium reduction rate is relatively slow, and only partial reduction is usually achieved; thus, a complete separation of plutonium from uranium would be difficult. However, a complete separation is not necessary in coprocessing operations.

3. Because of the susceptibility of TBP·HNO_2 to decomposition by venting NO according to the reverse of reaction (1), the TBP·HNO_2/NPH solution must be stored in a closed system. This system may be better suited to column operations (perhaps under a slight positive pressure) than in open mixer-settlers, although it has been used successfully in the SETF mixer-settlers where the organic solution is vented and exposed during residence times of 10 to 20 min. In the SETF tests, the TBP·HNO_2 concentration has been nearly uniform throughout the bank.

4.2.1 Demonstration of potential flowsheets using HNO_2

During Campaign 3, a potentially useful flowsheet which incorporated the use of HNO_2 for reductive strippping of plutonium was examined. This scheme, shown in Fig. 6, utilizes three solvent extraction contactors. In the A-contactor, uranium and plutonium are coextracted and coscrubbed; in the B-contactor, plutonium and uranium are stripped but part of the uranium is reextracted or backscrubbed (a form of partial partitioning); and in the C-contactor, the remaining uranium is stripped. This method features (1) addition of HNO_2 into the B-contactor via dissolution in the organic backscrub stream (BS), and (2) the use of 0.10 M HNO_3 as the strip solution for both the uranium-plutonium product in the B-contactor (BX) and for the uranium product in the C-contactor (CX). The B-contactor is operated at 30°C and the C-contactor at 40°C. In addition to its use for decontamination purposes, the low-acidity scrub stream (AS) in the A-contactor serves to reduce the acidity in the pregnant organic stream (AP/BF) and thus enables maintenance of the maximum aqueous-phase acidity in the B-contactor at $\leq$0.6 M HNO_3.

A series of SETF test runs (3-2A through 3-2D) was made to demonstrate various aspects of the flowsheet. At the conditions used during test run 3-2A (Table 6), the plutonium stripping yield in the B-contactor

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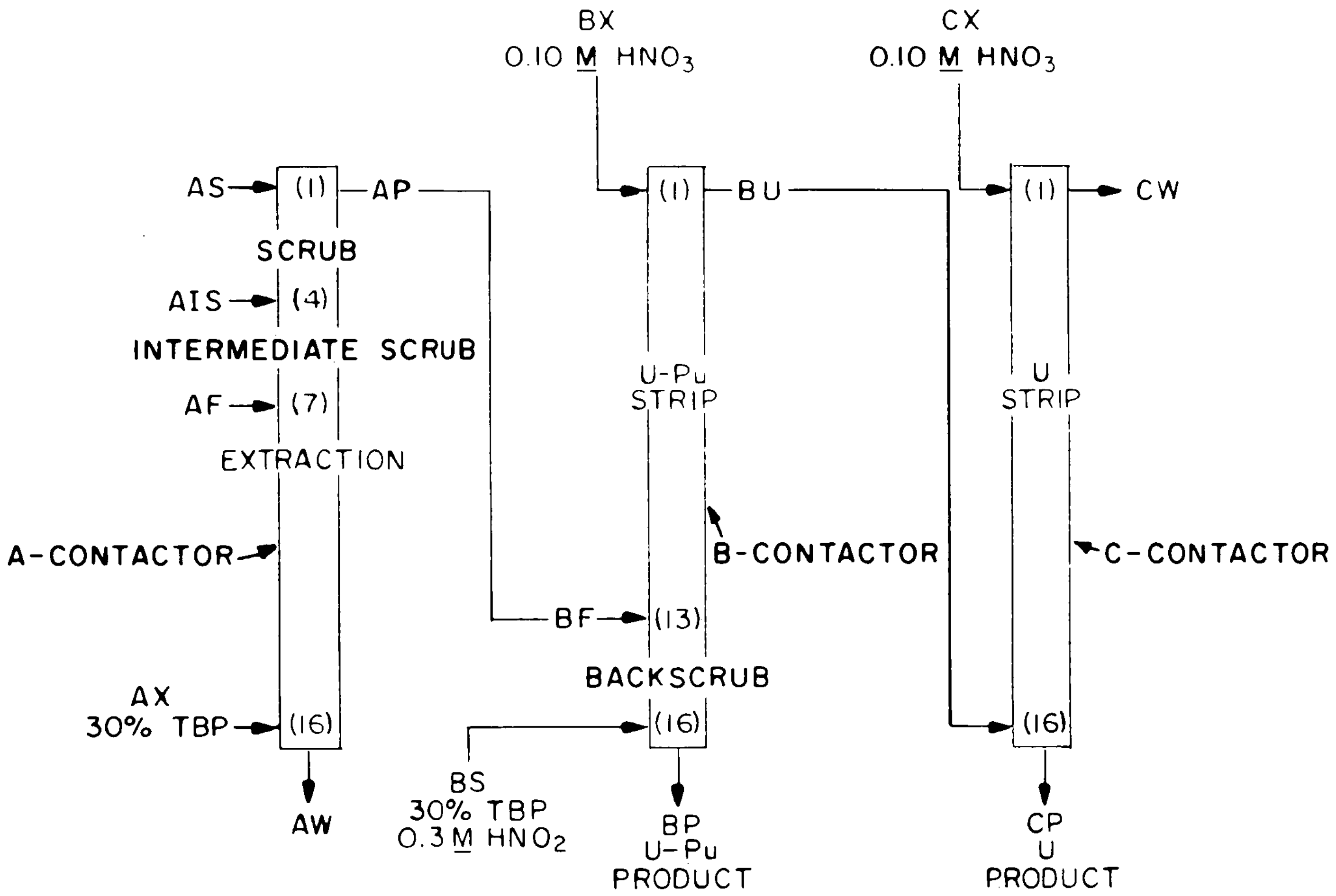

Fig. 6. Coextraction-partial partitioning flowsheet using HNO_2 as the plutonium reductant.

Table 6. Flow ratios and solution compositions during the 3-2 series of partitioning tests

	Test No.				
	3-2A	3-2B	3-2C	3-2D	Solution composition
Feed solution (AF)					
HNO_3, $\underline{M}$	3.2	3.5	3.4	3.9	
U, g/L	153	152	152	18.9	
Pu, g/L	1.28	0.026	1.42	1.54	
AF flow rate, L/h	0.82	0.82	0.81	0.95	
Flow ratios (AF = 1.0)					
AS	0.22	0.23	0.22	0.10	0.8 $\underline{M}$ HNO_3
AIS	0.13	0.14	0.13	0.066	3.0 $\underline{M}$ HNO_3
AX	1.69	1.75	1.72	0.35	30% TBP in NPH
BS	0.70	0.74	0.67	0.15	30% TBP in NPH; 0.25 $\underline{M}$ HNO_2
BX	0.62	0.59	0.59	0.16	0.10 $\underline{M}$ HNO_3
CX	2.38	--	--	--	0.10 $\underline{M}$ HNO_3

was 99.93% and the plutonium content of the BP stream was 5.3% of the heavy metal (U + Pu). This represented a plutonium CF of 6.4. These results (and others) showed that a significant enrichment of the plutonium (CF in the range of 5 to 10) could be obtained while maintaining a sufficient plutonium stripping yield.

Although 0.07% of the plutonium entering the B-contactor during test 3-2A remained with the uranium in the BU stream, 0.068% (97% of the 0.07%) was recovered along with the uranium product (CP) in the C-contactor. A test (3-2B) was then made to demonstrate that, if a significant amount of plutonium was recovered with the uranium in the CP stream during the first-cycle operations, it could be partitioned in the subsequent uranium second-cycle operation by using the same three-contactor flowsheet. The feed for test 3-2B was the uranium product from test 3-2A. A small amount of "extra" plutonium was added to the 3-2B feed. The feed adjustment for test run 3-2B, which included an intercycle evaporation of the feed solution, was done without apparent difficulty or loss of plutonium. The uranium-plutonium product (BP) from the run contained 99.96% of the plutonium and 11.3% of the uranium (CF = 8.3).

Since the plutonium CFs obtained with the flowsheet shown in Fig. 6 are typically in the range of 5 to 10, and a CF of ≥ 30 is needed to prepare FBR core fuel from LWR fuel solutions, the uranium-plutonium product would need to be partitioned further by means of a second cycle of the flowsheet. This was demonstrated by tests 3-2C and 3-2D. In the first cycle (3-2C), the plutonium stripping yield was 99.93% and the CF was 6.7; in the second cycle (3-2D), the stripping yield was 99.97% and the CF was 9.3. Plutonium in the second-cycle U-Pu product was 63% of the heavy metal (U + Pu).

When processing FBR-irradiated fuel solutions (mixed core and blanket), the initial plutonium will represent ~10% of the heavy metal and a CF of only 3 to 4 will be needed. Thus, only one cycle of the flowsheet would be required. Test 3-3 was made to demonstrate these conditions. The feed solution was formulated to contain uranium, plutonium, and fission product elements at concentrations of 98, 7.4, and ~2 g/L, respectively, by combining a batch of dissolver solution (from LWR fuel) with recycle plutonium. The recycled plutonium apparently contained aqueous-soluble TBP-decomposition products which caused the formation of a zirconium-based emulsion in the extraction bank and resulted in operational difficulties during the test run. In an effort to improve operations, the TBP concentration in the solvent was reduced to 15%. Still, numerous shutdowns were required to remove emulsion from the A-contactor. In spite of the operating difficulty, good data were obtained from the B-contactor and the effect of flow ratio in the strip section of that contactor on the plutonium CF was evaluated. Organic/aqueous ratios of 4.7, 5.9, and 7.0 were used in the strip section (an O/A ratio of 1.0 was maintained in the scrub section) to obtain CFs of 2.3, 3.0, and 3.7, respectively; these are in the range required for processing FBR fuels. The yield of stripped plutonium, 99.94%, was not affected significantly by the flow ratio changes.

4.2.2 Effects of process parameters

During Campaign 4 (Tables A-3, A-4, A-5, and A-6), tests were made to elucidate the effects of two of the process parameters: the HNO_2/Pu mole ratio and the residence time of solutions within the uranium-plutonium stripping contactor.

In order to determine the effects of HNO_2/Pu mole ratio, results from several partial partitioning tests were compared (Fig. 7). In these tests, either the reductant was omitted or varying amounts of HNO_2 (Table A-4) were added via the organic backscrub stream. Figure 7 shows the aqueous-phase plutonium concentration profiles within the partial partitioning contactor (a 16-stage mixer-settler containing 13 stripping and 3 scrub stages). A calculated (SEPHIS code) composition profile for Pu(IV) stripping is shown for comparison with the data. This calculated profile indicated that, at the conditions used, most of the plutonium would reflux unless some reduction to Pu(III) was provided. Actual results indicated that, even in tests where no HNO_2 was added, the HNO_2 apparently generated by radiolysis of HNO_3 (0.15 mol/mol plutonium in this case) enabled enough reduction of Pu(IV) to improve the plutonium stripping significantly (the stripping

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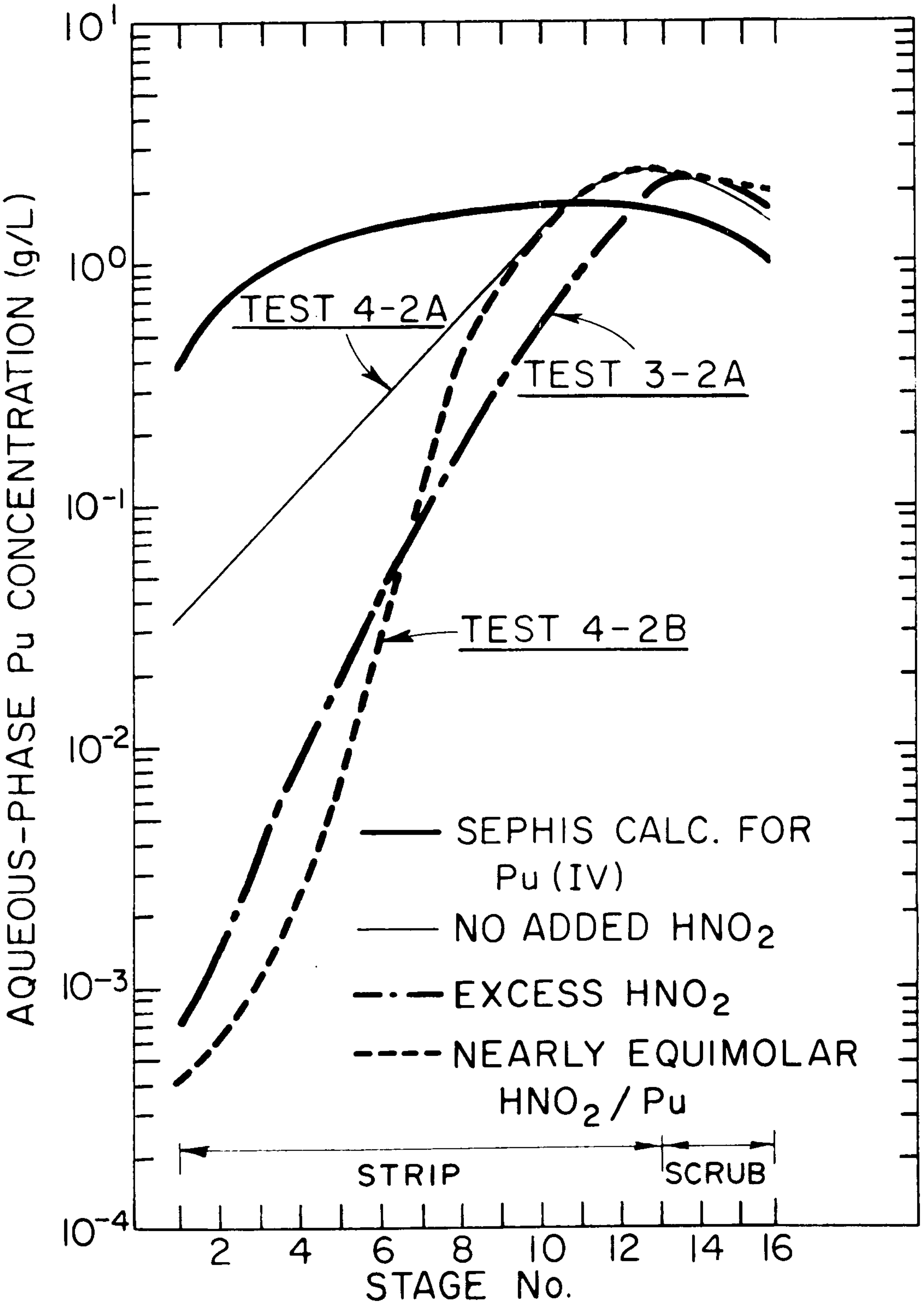

Fig. 7. Effect of HNO_2 concentration in strip contactor.

yield in this run was 97.9%). Moreover, when a relatively small amount of HNO_2 was added (Table A-4) to make the HNO_2 and plutonium approximately equimolar (measured mole ratio of 1.2 in this test), the plutonium stripping was essentially as effective as when a large excess of HNO_2 was used.

The effect of residence time in the mixer-settler on stripping performance was investigated during test 4-3 (Table A-4) while using 15% TBP—NPH as the organic extractant and backscrub solution. The results, summarized in Table 7, show that decreasing the residence time by a factor of ~2

Table 7. Effect of strip-bank residence time on U-Pu stripping

	Residence time per stage in strip bank			
	"Long"[a]		"Short"[b]	
	Mixer[c] (s)	Settler[c] (s)	Mixer[c] (s)	Settler[c] (s)
Total liquid	42	72	21	35
Organic solution[d] (15% TBP/NPH)	32	53	16	26
Aqueous solution[d]	144	240	78	126
HNO_2/Pu mole ratio	40		110	
Stripping performance				
% of Pu stripped	99.81		99.75	
% of U stripped	6.0		5.4	
% Pu in U-Pu product	9.4		11.1	
Pu CF	12		14	

[a] Data from test 4-3A.
[b] Data from test 4-3B.
[c] Based on stage volumes of 30 mL in mixer and 50 mL in settler.
[d] Based on estimated organic/aqueous volume ratio of 2/1 in each stage.

caused very little change in the stripping performance. The plutonium concentration profile, shown in Fig. 8, indicated only a slight degradation of stripping efficiency within the contactor. It is interesting to note that, at the shorter residence time (test 4-3B), the HNO_2 concentration within the contactor was two to three times higher even though the concentration entering the contactor via the backscrub stream was the same. Apparently, the loss of HNO_2 by decomposition and venting of NO was decreased by using a shorter residence time.

4.2.3 Absence of plutonium polymer formation

Since the use of HNO_2 as a reductant for Pu(IV) requires operation of the contactor with aqueous-phase acidities of ~0.5 $\underline{M}$ HNO_3 or less, the use of strip solutions containing acidities as low as 0.10 $\underline{M}$ HNO_3 has been allowed in the SETF. Even when the strip solution was 0.10 $\underline{M}$ HNO_3, the aqueous-phase acidity was usually $\geq$0.25 $\underline{M}$ HNO_3 within the contactor in the region where the plutonium concentration was highest. This was due to the stripping of acid from the pregnant organic stream (AP). The possibility of plutonium polymer formation at these relatively low acidities was recognized, but no evidence of polymer formation (i.e., significant plutonium losses or formation of solids and/or cruds within the strip contactor) was observed during any of the SETF tests. Moreover, following Campaign 3, the mixer-settlers were leached with 8 $\underline{M}$ HNO_3 for several days (including 16 h at 50°C) to dissolve any polymer that may have been deposited. Only 1 mg of plutonium was leached from the stripping contactor, compared with a total of 1.4 kg of plutonium that had been processed in the mixer-settlers.

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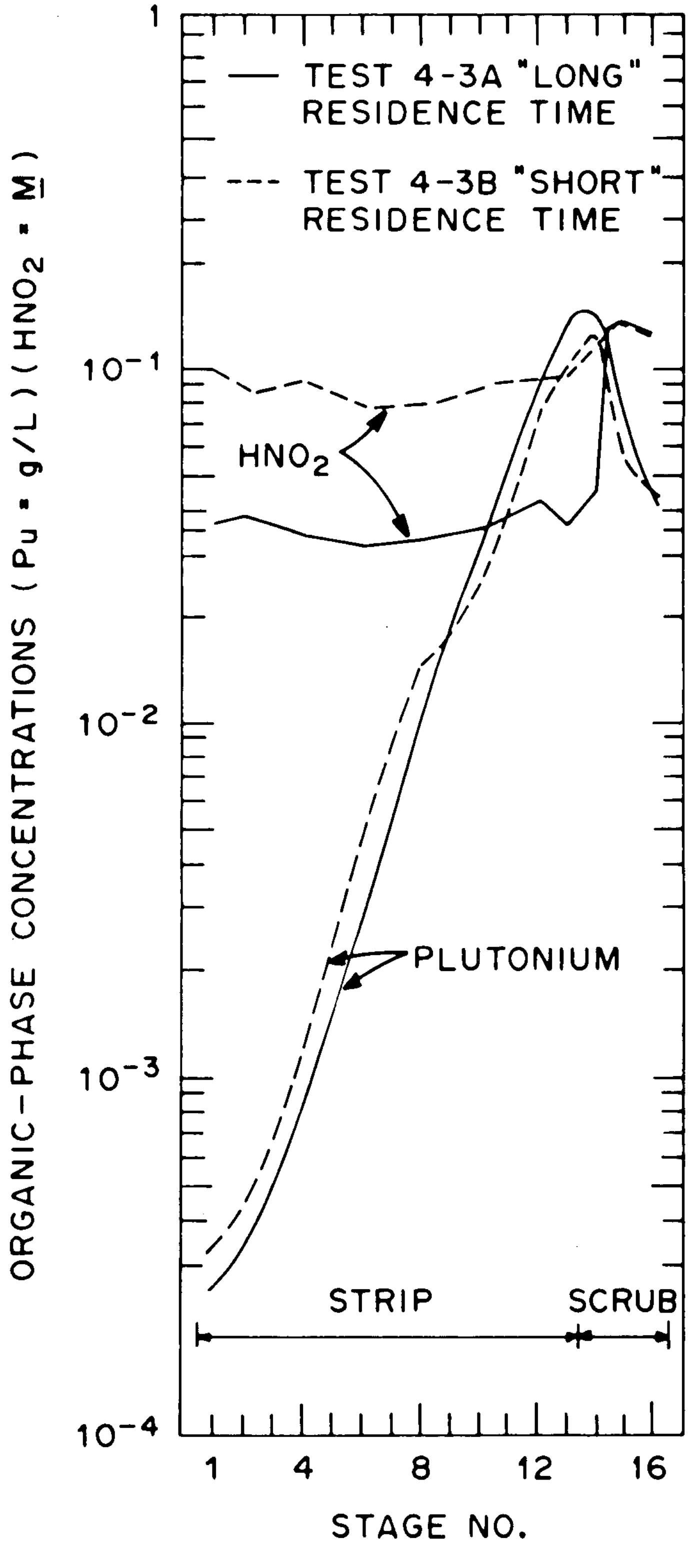

Fig. 8. Effect of strip bank residence time on plutonium and HNO_2 concentrations.

In associated laboratory studies, Lloyd and North[4,5] investigated the possibility of plutonium polymer formation while stripping a 30% TBP—NPH solution that contained 70 g of uranium, 1.5 g of plutonium, and 0.2 mol of HNO_3 per liter (these are concentrations typically obtained when processing LWR fuels). The organic solution was batch-stripped with five successive volumes of HNO_3 solution at 50°C. No indication of plutonium polymer formation was found when using strip solutions with acidities of ≥ 0.05 $\underline{M}$ HNO_3.

4.3 Comparison of Solvent Extraction Behavior of Feed Solutions

The extraction-scrubbing conditions of test 4-1 (Table A-4) were selected to allow comparison of the solvent extraction behavior of a feed solution prepared from the Dresden-1 BWR fuel with that observed in test 3-1 (Table A-1) for feed prepared from H. B. Robinson-2 PWR fuel. No significant differences were found. Uranium and plutonium losses to the AW stream were 0.04% and 0.002%, respectively, for the Dresden fuel as compared with 0.03% and 0.007% for the Robinson fuel. The decontamination factors (DFs) of 3×10^4 for ^{106}Ru, 4×10^6 for ^{137}Cs, and $>2 \times 10^5$ for ^{144}Ce, obtained when processing Dresden fuel, were slightly higher than those obtained when processing Robinson fuel (1.5×10^4 for ^{106}Ru, 5×10^5 for ^{137}Cs, and $>6 \times 10^4$ for ^{144}Ce).

4.4 Feed Clarification Improvements

Methods for preventing the formation of cruds in the extraction bank have been evaluated in previous SETF experiments.[1] Some improvement had been obtained in one test in which the dissolved LWR fuel solution was (1) clarified by means of filtration using a precoated, etched-disc filter (1-μm pore openings); (2) diluted to a heavy-metal concentration of 150 g/L; and (3) digested with H_2O_2 at 90°C. This procedure was employed to enhance the dissolution of any metal colloids in the clarified feed solution. Since the initial trial appeared promising, the procedure was tested further during Campaign 3.

Two modifications were made prior to solvent extraction test run 3-1A. The first modification was the use of a continuous (rather than a batch) addition of H_2O_2 during the digestion step, and the second modification was an additional clarification of the treated feed solution. The latter was accomplished by filtration, using a sintered-metal filter rated at 0.5 μm. The amount of crud formed in the extraction bank during solvent extraction test 3-1A was significantly decreased compared to that observed in previous runs. During later tests, the digestion with H_2O_2 was found to be more effective than the filtration.

During Campaign 4, excellent results were achieved by clarifying the dissolver solutions via filtration using a "deep-bed" type of filter that contained a bed of diatomaceous earth as the filtering medium (see Fig. 1). The filtrations, which were accomplished at up to 5 L/h (in comparison with the 0.5- to 2-L/h rates obtained with the precoated, etched disc filter), yielded filtrates that were lightly colored and free of visible solids. (Samples of the filtrate were centrifuged at high $\underline{g}$ values, and no accumulation of solids was observed.) Only a relatively small formation of crud was observed in the subsequent solvent extraction runs.

5. CONCLUSIONS

The following conclusions have been established from the most significant results of Campaigns 3 and 4 of the SETF:

1. The HEF flowsheet for partial partitioning (by means of reducing the plutonium in the feed solution to the trivalvent state followed by selective extraction of part of the uranium) can only be performed successfully by limiting the maximum acidity of the feed solution. This is because HAN, which is used as the reductant for tetravalent plutonium, becomes less effective as the aqueous-phase acidity is increased. Previous tests indicated that HAN could be used effectively if the aqueous-phase acidity was kept <1 $\underline{M}$ HNO_3 (preferably below 0.5 $\underline{M}$ HNO_3). However, the HEF flowsheet was tested successfully when the maximum aqueous acidity was ~1.5 $\underline{M}$ HNO_3, but a test at 3 $\underline{M}$ HNO_3 failed completely because the trivalent plutonium was rapidly reoxidized.

2. A process in which HNO_2 is added to the organic solvent in the plutonium-uranium strip contactor and serves as the reductant for tetravalent plutonium appeared to be attractive for use in reprocessing irradiated fuels from both FBRs and LWRs. The most attractive feature is that, when HNO_2 is added to the HNO_3/TBP/Pu system, it only supplements the HNO_2 already being generated in the system by radiolysis of HNO_3; thus, it introduces no new chemicals into the system. For successful operation, the aqueous-phase acidity in the plutonium stripping contactor must be kept below ~0.6 $\underline{M}$ HNO_3. The effectiveness of the Pu(IV) reduction is not as good as when HAN is used, but it appears sufficient for partial partitioning operations, especially when processing FBR fuels from which a large degree of separation of uranium from the uranium-plutonium is not required.

3. Strip solutions for plutonium-uranium products, which contain acidities as low as 0.10 $\underline{M}$ HNO_3, apparently can be used without causing significant hydrolysis and polymerization of the plutonium. Since the use of HNO_2 (and the most effective use of HAN) as reductants for tetravalent plutonium require relatively low acidities, strip solutions containing 0.10 $\underline{M}$ HNO_3 were used in many SETF tests, and no evidence of polymer formation was observed.

4. Feed solutions prepared from BWR fuel can be processed without difficulty. The solvent extraction performance in SETF tests was similar to that obtained when using feed prepared from PWR fuel.

5. A "deep-bed" type of filter, containing a bed of diatomaceous earth as the filtering medium, can be used to effectively clarify the solvent extraction feed solutions at faster rates than precoated filters. The used filters, containing insoluble fission product residues mixed with diatomaceous earth (SiO_2), appear to be in a form which is compatible with waste solidification methods (vitrification, cementation, etc.).

6. ACKNOWLEDGMENTS

The experimental work done in the SETF was performed by the Chemical Technology Division staff of the Transuranium Processing Plant. The extensive analytical work was carried out by Analytical Chemistry Division personnel, primarily those under the supervision of J. H. Cooper and J. M. Peele. Engineering and maintenance services were provided by personnel from General Engineering, the Instrumentation and Controls Division, and the Plant and Equipment Division, under the supervision of S. O. Lewis, A. Shourbaji, E. M. Shuford, and B. R. Scarborough.

7. APPENDIX

Table A-1. Campaign 3 first-cycle tests - extraction/scrub bank conditions and results

	Run No.			
	3-1A	3-2A	3-2C	3-3A, 3-3B, 3-3C
Dates	10/1-2/79	10/25-27/79	11/3-4/79	11/14-21/79
Bank temperature, °C	51-52	49	50	50-52
Number of stages:				
Final scrub/intermediate scrub/extraction	3/3/10	3/3/10	3/3/10	3/3/10
AX stream flow rate, L/h	1.54	1.39	1.40	1.32
Flow ratios:				
AS/AX	0.12	0.13	0.13	0.087
AIS/AX	0.066	0.079	0.073	0.059
AF/AX	0.50	0.59	0.58	0.39
Inlet stream compositions[a]				
AS stream: HNO_3, M	0.8	0.8	0.8	0.8
AIX stream: HNO_3, M	3.0	3.0	3.0	3.0
AX stream: % TBP	30	30	30	15
AF stream:				
HNO_3, M	2.9	3.2	3.4	3.4
U, g/L	156	153	152	98
Pu, g/L	1.4	1.28	1.42	7.4
Am, mg/L	45	42	40	36
Cm, mg/L	3.0	2.7	3.0	1.08
Ru-106, Ci/L	0.89	0.99	0.99	0.34
Sb-125, Ci/L	0.30	0.28	0.41	0.068
Cs-134, Ci/L	c	2.8	2.5	0.89
Cs-137, Ci/L	12.1	11.9	10.3	3.88
Ce-144, Ci/L	1.14	1.03	0.97	0.32
Eu-154, Ci/L	0.51	0.49	0.46	0.145
Np-239, Ci/L	0.0022[b]	0.0021[b]	0.0020[b]	0.00081[b]

Table A-1 (continued)

	Run No.					
	3-1A	3-2A	3-2C	3-3A	3-3B	3-3C
Outlet stream compositions[a]						
AW stream:						
HNO_3, M	c	2.9	3.2	2.8	2.5	2.7
U, mg/L	38	35	<50	<20	23	<20
Pu, mg/L	0.074	0.032	0.047	1.9	c	0.68
Am, mg/L	36	36	33.5	24	27	26
Cm, mg/L	2.3	2.1	2.2	0.75	0.60	0.66
Ru-106, Ci/L	0.65	0.62	0.68	0.24	0.27	0.24
Sb-125, Ci/L	0.081	0.16	<0.06	0.054	<0.06	<0.11
Cs-134, Ci/L	1.7	2.1	1.9	0.69	0.69	0.76
Cs-137, Ci/L	7.4	8.9	6.5	3.1	3.0	3.4
Ce-144, Ci/L	0.76	0.76	0.78	0.25	0.24	0.27
Eu-154, Ci/L	0.43	0.39	0.32	0.12	0.13	0.13
AP stream:						
HNO_3, M	0.05	0.01	0.05	0.03	0.03	0.03
HNO_2, M	0.0004	0.0041	0.0026	c	0.0011	c
U, g/L	79	86	83	33	33	35
Pu, g/L	0.78	0.69	0.68	2.57	2.43	2.43
Ru-106, μCi/L	87	43	489	7.8	<6	<6
Sb-125, μCi/L	<0.6	<0.9	<1.4	<0.8	<0.2	<0.6
Cs-134, μCi/L	<0.11	<0.2	<0.3	2.4	<0.1	0.76
Cs-137, μCi/L	<0.16	<0.5	<0.9	8.1	<0.3	3.0
Ce-144, μCi/L	<6	<6	<3	<5	<6	<6
Eu-154, μCi/L	<14	<6	<0.9	<0.8	<0.2	<0.6
Np-239, μCi/L	830	94	11	84	c	c

[a] On date of run.

[b] Calculated from ^{243}Am concentration in AF.

[c] Not measured.

Table A-2. Campaign 3 first-cycle tests - strip bank conditions and results

	Run No.					
	3-1A	3-2A	3-2C	3-3A	3-3B	3-3C
Dates	10/1-2/79	10/25-27/79	11/3-4/79	11/14-17/79	11/17-19/79	11/20-21/79
Bank temperature, °C	50	27	27	26	27	27
Number of stages:						
Strip/scrub	16[a]/0	13/3	13/3	13/3	13/3	13/3
BX stream flow rate, L/h	1.59	0.51	0.48	0.35	0.28	0.22
Flow ratios:						
AP/BX	0.97	2.72	2.89	3.78	4.88	6.0
BS/BX	--	1.12	1.13	0.91	1.01	1.0
BIX/BX	0.096	--	--	--	--	--
Inlet stream composition						
BX: HNO_3, M	0.05	0.10	0.10	0.10	0.10	0.10
BIX: HNO_3, M	3.0	--	--	--	--	--
BS stream:						
% TBP	--	30	30	15	15	15
HNO_2, M	--	0.29	0.21	0.29	0.29	0.29
AP stream:						
% TBP	30	30	30	15	15	15
HNO_3, M	0.05	0.01	0.05	0.03	0.03	0.03
HNO_2, M	0.0004	0.0041	0.0026	b	0.0011	b
U, g/L	79	86	83	33	33	35
Pu, g/L	0.78	0.69	0.68	2.57	2.43	2.43
Ru-106, µCi/L	87	43	489	7.8	<6	<6
Sb-125, µCi/L	<0.6	<0.9	<1.4	<0.8	<0.2	<0.6
Cs-134, µCi/L	<0.11	<0.2	<0.3	2.4	<0.1	0.76
Cs-137, µCi/L	<0.16	<0.6	<0.9	8.1	<0.3	3.0
Ce-144, µCi/L	<6	<6	<3	<5	<6	<6
Eu-154, µCi/L	<14	<6	<0.9	<0.8	<0.2	<0.6
Np-239, µCi/L	830	94	11	84	b	b

Table A-2 (continued)

	Run No.					
	3-1A	3-2A	3-2C	3-3A	3-3B	3-3C
Outlet stream compositions[c]						
BP stream:						
HNO_3, M	0.38	0.30	0.34	0.33	0.39	0.48
U, g/L	64	35	29	51	43	46
Pu, g/L	0.64	1.97	2.09	9.74	11.6	16.2
Ru-106, μCi/L	27	49	3.2	0.81	<0.81	<0.81
Sb-125, μCi/L	<2	<6	<0.03	<0.2	<0.14	<0.14
Cs-134, μCi/L	3.5	b	2.5	1.9	0.78	0.11
Cs-137, μCi/L	10	11	11	8.0	3.2	0.32
Ce-144, μCi/L	<9	<24	1.2	<0.9	<1.4	<2.2
Eu-154, μCi/L	<2	<6	<0.2	<0.09	<0.11	<0.14
Np-239, μCi/L	844	541	35	7.8	<325	b
BU stream:						
HNO_3, M	0.01	0.03	b	0.02	0.03	0.01
HNO_2, M	<0.0002	0.045	0.047	0.011	0.012	b
U, g/L	0.37	52	55	15	18	21
Pu, g/L	6.4E-5	3.5E-4	3.5E-4	1.3E-3	1.1E-3	1.3E-3
Ru-106, μCi/L	108	28	354	0.38	0.22	0.16
Sb-125, μCi/L	<3	<0.2	b	<0.011	<0.014	<0.016
Cs-134, μCi/L	b	<0.06	b	b	b	0.081
Cs-137, μCi/L	<9	<0.2	b	<0.014	<0.016	0.37
Ce-144, μCi/L	<27	<1	b	<0.9	<0.08	<0.16
Eu-154, μCi/L	<0.3	<0.1	b	<0.017	<0.011	<0.016
Np-239, μCi/L	b	<1	<35	<0.3	<20	<65

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[a] Comprised of six low-acid strip stages and ten high-acid stages.

[b] Not measured.

[c] On date of run.

Table A-3. Campaign 4 first-cycle tests - extraction/scrub bank conditions and results

	Run No.					
	4-1A	4-2A	4-2B	4-3A	4-3B	4-3C
Dates	5/7/80	5/21/80	5/21-22/80	6/4/80	6/5/80	6/6-7/80
Bank temperature, °C	50-51	40	40	41	41-43	40-41
Number of stages:						
Final scrub/intermediate scrub/extraction	3/3/10	3/3/10	3/3/10	3/3/10	3/3/10	3/3/10
AX stream flow rate, L/h	1.41	1.72	1.73	1.39	2.77	1.63
Flow ratios:						
AS/AX	0.134	0.13	0.13	0.065	0.066	0.114
AIS/AX	0.066	0.071	0.070	0.043	0.032	0.056
AF/AX	0.555	0.275	0.283	0.176	0.169	0.315
Inlet stream compositions[a]						
AS stream: HNO_3, M	0.82	0.81	0.81	0.80	0.82	0.82
AIS stream: HNO_3, M	3.1	3.0	3.0	3.6	3.6	3.6
AX stream: % TBP	30	30	30	15	15	15
AF stream:						
HNO_3, M	3.2	3.2	3.2	2.9	2.9	2.7
U, g/L	153	292	292	212	212	103
Pu, g/L	1.3	2.3	2.3	1.46	1.46	0.86
Am, mg/L	51	99	99	61	61	33
Cm, mg/L	4.4	6.2	6.2	2.7	2.7	1.7
Ru-106, Ci/L	0.81	1.27	1.27	1.11	1.11	0.59
Sb-125, Ci/L	0.16	0.30	0.30	0.14	0.14	c
Cs-134, Ci/L	2.3	4.2	4.2	2.32	2.32	1.24
Cs-137, Ci/L	10.8	20.7	20.7	12.4	12.4	6.66
Ce-144, Ci/L	0.62	1.2	1.2	0.74	0.74	0.40
Eu-154, Ci/L	0.49	0.81	0.81	0.49	0.49	0.26
Np-239, Ci/L	0.0025[b]	0.0048[b]	0.0048[b]	0.0031[b]	0.0031[b]	0.0016[b]

Table A-3 (continued)

	Run No.					
	4-1A	4-2A	4-2B	4-3A	4-3B	4-3C
Outlet stream compositions[a]						
AW stream:						
HNO_3, $\underline{M}$	2.4	2.4	2.4	2.3	2.45	3.0
U, mg/L	20	<20	<100	0.6	0.7	<0.1
Pu, mg/L	0.015	0.030	0.021	0.32	0.24	0.59
Am, mg/L	44	28	64	44	48	25
Cm, mg/L	3.1	1.5	3.7	1.7	2.1	1.1
Ru-106, Ci/L	0.57	0.65	0.81	0.49	0.60	<0.08
Sb-125, Ci/L	c	0.16	0.20	c	c	0.077
Cs-134, Ci/L	1.75	2.38	2.23	1.52	1.62	0.82
Cs-137, Ci/L	8.3	11.8	11.0	8.00	8.73	4.35
Ce-144, Ci/L	0.51	0.60	0.065	0.51	0.49	0.24
Eu-154, Ci/L	0.32	0.49	0.43	0.32	0.41	0.18
AP stream:						
HNO_3, $\underline{M}$	0.04	c	0.07	0.04	0.04	0.035
HNO_2, $\underline{M}$	0.008	c	0.0022	0.00043	0.0013	0.0078
U, g/L	84	71	80	33	34	29
Pu, g/L	0.69	0.56	0.55	0.23	0.20	0.176
Ru-106, μCi/L	34	420	327	48	57	81
Sb-125, μCi/L	c	<1	<1.1	c	c	c
Cs-134, μCi/L	c	<0.3	<0.24	<0.09	<0.2	<0.2
Cs-137, μCi/L	0.24	1.3	0.51	0.43	<0.4	<0.2
Ce-144, μCi/L	<2	<3	<2.6	<2	<2	<0.9
Eu-154, μCi/L	<0.6	2.3	<0.9	<0.3	<0.5	<0.2
Np-239, μCi/L	123	218	1240	400	390	202

[a] On date of run.

[b] Calculated from ^{243}Am concentration in AF.

[c] Not measured.

Table A-4. Campaign 4 first-cycle tests - strip bank conditions and results

	Run No.					
	4-1A	4-2A	4-2B	4-3A	4-3B	4-3C
Dates	5/7/80	5/21/80	5/21-22/80	6/4/80	6/5/80	6/6-7/80
Bank temperature, °C	40-41	24	24	25-26	26	25-26
Number of stages:						
Strip/scrub	13/3	13/3	13/3	13/3	13/3	13/3
BX flow rate, L/h	0.243	0.592	0.601	0.251	0.469	0.276
Flow ratios:						
AP/BX	5.79	2.90	2.88	5.53	5.91	5.90
BS/BX	0.87	1.10	1.10	3.53	3.93	3.88
Inlet stream compositions[a]						
BX stream:						
HNO_3, M	0.09	0.10	0.10	0.10	0.10	0.10
HAN, M	0.06	--	--	--	--	--
BS stream:						
% TBP	30	30	30	15	15	15
HNO_2, M	--	--	0.012	0.10	0.17	0.17
AP stream:						
% TBP	30	30	30	15	15	15
HNO_3, M	0.04	b	0.07	0.04	0.04	0.03
HNO_2, M	0.008	b	0.0022	0.00043	0.0013	0.0078
U, g/L	84	71	80	33	34	29
Pu, g/L	0.69	0.56	0.55	0.23	0.20	0.175
Ru-106, µCi/L	34	420	327	48	57	81
Sb-125, µCi/L	b	<1	<1.1	b	b	b
Cs-134, µCi/L	b	<0.3	<0.3	<0.09	<0.2	<0.2
Cs-137, µCi/L	0.24	1.3	0.51	0.43	<0.4	<0.2
Ce-144, µCi/L	<2	<3	<3	<2	<2	<0.9
Eu-154, µCi/L	<0.6	2.3	<0.9	<0.3	<0.5	<0.2
Np-239, µCi/L	123	218	1240	400	390	202

Table A-4 (continued)

	Run No.					
	4-1A	4-2A	4-2B	4-3A	4-3B	4-3C
Outlet stream compositions[a]						
BP stream:						
HNO_3, M	0.49	0.34	0.32	0.58	0.59	0.50
U, g/L	20.6	9.54	12.7	11.9	10.6	14.5
Pu, g/L	4.36	1.47	1.64	1.24	1.32	1.18
Ru-106, μCi/L	76	560	163	48	178	15
Sb-125, μCi/L	b	b	<1	b	b	b
Cs-134, μCi/L	b	24	4.6	<1	5.7	0.8
Cs-137, μCi/L	7.1	124	24	6.7	31	3.7
Ce-144, μCi/L	<9	<6	<3	<5	<10	<3
Eu-154, μCi/L	1.6	7.3	2.7	0.089	3.0	<0.3
Np-239, μCi/L	837	b	1440	2130	2475	1004
BU stream:						
HNO_3, M	0.01	0.01	0.01	0.03	0.04	0.04
HNO_2, M	<0.0002	0.0006	0.0022	0.037	0.072	0.038
U, g/L	67.9	47.6	49.4	20.8	18.9	17.0
Pu, mg/L	0.32	7.8	0.35	0.26	0.33	0.11
Ru-106, μCi/L	35	190	64	30	24	42
Sb-125, μCi/L	b	0.86	b	b	b	b
Cs-134, μCi/L	b	<0.14	b	b	b	<0.05
Cs-137, μCi/L	<0.3	<0.3	0.19	0.46	<0.07	<0.2
Ce-144, μCi/L	<0.9	<0.8	b	b	<0.4	<0.5
Eu-154, μCi/L	<0.2	<0.5	b	1.6	<0.2	<0.2
Np-239, μCi/L	<1.3	b	b	<6.1	b	<10

[a] On date of run.

[b] Not measured.

Table A-5. Campaign 3 second-cycle tests - extraction/scrub bank conditions and results

	Run No.				
	3-2B	3-2D	3-1B[a]	3-1C[a]	3-1D[a]
Dates	10/31-11/1/79	11/7-8/79	10/4/79	10/4-5/79	10/5-6/79
Bank temperature, °C	50	51	28	27	27
Number of stages:					
Final scrub/intermediate scrub/extraction	3/3/10	3/3/10	5/7/4	5/7/4	5/7/4
AX flow rate, L/h	1.43	0.33	0.74	0.73	0.69
Flow ratios:					
AS/AX	0.13	0.29	0.17	0.17	0.17
AIS/AX	0.077	0.19	0.094	0.094	0.095
AF/AX	0.57	2.87	1.34	1.47	1.27
Inlet stream compositions					
AS stream:					
HNO_3, M	0.8	0.8	0.09[b]	0.09[c]	0.09[c]
AIS stream:					
HNO_3, M	3.0	3.0	3.0	3.0	3.0
AX stream:					
% TBP	30	30	30	30	30
Feed stream:					
HNO_3, M	3.5	3.9	1.3	1.3	3.0
U, g/L	152	18.9	57	48	50
Pu, g/L	0.026	1.54	0.48	0.46	0.42
Ru-106, mCi/L	3.4	0.67	d	d	d
Sb-125, mCi/L	4.1	0.44	d	d	d
Cs-134, mCi/L	0.38	0.064	d	d	d
Cs-137, mCi/L	1.7	0.24	d	d	d
Ce-144, mCi/L	0.16	0.016	d	d	d
Eu-154, mCi/L	0.62	0.011	d	d	d

Table A-5 (continued)

	Run No.				
	3-2B	3-2D	3-1B[a]	3-1C[a]	3-1D[a]
Outlet stream compositions					
Aqueous raffinate stream:					
HNO_3, $\underline{M}$	2.9	3.5	1.4	1.4	2.5
U, mg/L	280	<10	390	260	640
Pu, mg/L	0.0024	0.012	403	356	0.6
Ru-106, mCi/L	8.2	2.2	0.39	0.58	0.61
Sb-125, mCi/L	4.3	0.51	0.46	0.16	0.14
Cs-134, mCi/L	16	4.0	0.91	0.007	0.022
Cs-137, mCi/L	67	16.6	3.7	0.29	0.086
Ce-144, mCi/L	6.1	1.4	0.31	0.016	0.008
Eu-154, mCi/L	2.8	0.68	0.02	0.019	0.003
Organic extract stream:					
HNO_3, $\underline{M}$	0.04	d	0.02	<0.01	<0.01
HNO_2, $\underline{M}$	0.0017	d	d	d	d
U, g/L	81	40	60	71	52
Pu, g/L	0.017	4.3	0.0017	0.0003	0.130
Ru-106, µCi/L	6.5	d	48	6.0	15
Cs-137, µCi/L	0.27	d	0.3	d	0.19
Ce-144, µCi/L	<1.1	d	2.6	d	d

[a] In tests 3-1B, 3-1C, and 3-1D, the selective uranium extraction method for partitioning was used. During test 3-1D, extensive refluxing of plutonium occurred, and the test run was aborted.

[b] AS stream also contained 0.02 $\underline{M}$ HAN.

[c] AS stream also contained 0.02 $\underline{M}$ HAN and 0.01 $\underline{M}$ N_2H_4.

[d] Not measured.

Table A-6. Campaign 3 second-cycle tests — strip bank conditions and results

	Run No.			
	3-1B[a]	3-1C[a]	3-2B[a]	3-2D
Dates	10/4/79	10/4-5/79	10/31-11/1/79	11/7-8/79
Bank temperature, °C	50	49-50	28	27
Number of stages:				
Strip/scrub	16/0	16/0	13/3	13/3
BX flow rate, L/h	0.57	0.55	0.48	0.13
Flow ratios:				
AP/BX	1.30	1.33	2.98	2.50
BS/BX	--	--	1.25	1.10
Inlet stream composition				
BX stream:				
HNO_3, M	0.01	0.01	0.10	0.10
BS stream:				
% TBP	--	--	30	30
HNO_2, M	--	--	0.29	0.21
AP stream:				
% TBP	30	30	30	30
HNO_3, M	0.02	0.01	0.04	b
HNO_2, M	b	b	0.0017	b
U, g/L	60	71	81	40
Pu, g/L	0.0017	0.0003	0.017	4.3
Ru-106, μCi/L	48	6.0	6.5	b
Cs-137, μCi/L	0.3	b	0.27	b
Ce-144, μCi/L	2.6	b	<1.1	b

Table A-6 (continued)

	Run No.			
	3-1B[a]	3-1C[a]	3-2B[a]	3-2D
Outlet stream compositions				
Aqueous product stream:				
HNO_3, M	0.04	0.04	0.33	0.47
U, g/L	64	58	28	6.0
Pu, g/L	0.0018	0.0008	0.050	10.1
Ru-106, μCi/L	4.3	5.4	<11	b
Cs-134, μCi/L	b	<0.11	11	b
Cs-137, μCi/L	0.27	0.27	45	b
Ce-144, μCi/L	<6	b	4.4	b
Eu-154, μCi/L	2.8	b	b	b
Stripped organic stream:				
HNO_3, M	<0.01	<0.01	0.05	b
HNO_2, M	b	b	0.042	0.037
U, g/L	0.37	0.20	51	33
Pu, μg/L	13	2.4	<5	640
Ru-106, μCi/L	8.4	4.6	<81	b
Cs-137, μCi/L	<20	<0.1	<11	b
Ce-144, μCi/L	b	b	18	b

[a] Strip bank used for uranium product stripping.

[b] Not measured.

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