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**Solvent-Extraction Studies of  
Coprocessing Flowsheets—Results  
From Campaigns 1 and 2 of the  
Solvent Extraction Test  
Facility (SETF)**

**MASTER**

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**APPLIED TECHNOLOGY**

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SOLVENT-EXTRACTION STUDIES OF COPROCESSING FLOWSHEETS; - RESULTS FROM  
CAMPAIGNS 1 AND 2 OF THE SOLVENT-EXTRACTION TEST FACILITY (SETF)

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SOLVENT EXTRACTION STUDIES OF COPROCESSING FLOWSHEETS — RESULTS FROM  
CAMPAIGNS 1 AND 2 OF THE SOLVENT EXTRACTION TEST FACILITY (SETF)

E. D. Collins, D. E. Benker, J. E. Bigelow, F. R. Chattin, M. H. Lloyd,  
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ABSTRACT

The Solvent Extraction Test Facility (SETF) was installed in one of the heavily shielded cells of the Transuranium Processing Plant during 1978. This facility contains dissolution, feed preparation, solvent extraction (three 16-stage mixer-settlers), and plutonium product handling equipment, as well as waste solution tanks. Irradiated fuel from the H. B. Robinson-2 Pressurized Water Reactor was processed in the first two campaigns of experimental work, which were completed in 1979. The objective was to test a variety of coprocessing flowsheets. Areas of the solvent extraction process that received special attention included (1) crud formation, particularly in the extraction contactor; (2) uranium and plutonium losses in the extraction raffinate; (3) fission product decontamination; (4) reduction of tetravalent plutonium, particularly by means of hydroxylamine nitrate or nitrous acid; (5) costripping of uranium and plutonium; (6) partial partitioning; and (7) solvent degradation.

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1. INTRODUCTION

The objective of work done in the SETF is to evaluate, at full activity levels, recently developed or improved flowsheets for reprocessing commercial nuclear power reactor fuels. Mixer-settlers were chosen as the type of solvent extraction contactor most appropriate for process chemistry studies. Thus, the central feature of the SETF equipment is a rack containing three banks of 16-stage mixer-settlers. This equipment has a scale of operation in the range of several kilograms of heavy-metal fuels per day. Conceptual design of the SETF was completed in late 1976 and equipment design, fabrication, and installation were accomplished during 1977 and 1978. The facility is located within one of the heavily shielded cells (Cell 5) of Building 7920, the Transuranium Processing Plant (TRU) at the Oak Ridge National Laboratory.

The first two SETF campaigns were made during the periods November–December 1978 and March–June 1979, respectively. The specific objective



during Campaigns 1 and 2 was to test a variety of coprocessing flowsheets for light water reactor (LWR) fuels. During the tests, emphasis was given to the use of hydroxylamine nitrate (HAN) as the reductant for Pu(IV) during the stripping of plutonium.

The irradiated UO<sub>2</sub> fuel used during these campaigns contained 28 kg of heavy metals and had been irradiated to a peak burnup of ~2.7 TJ/kg (~31,000 Mwd/metric ton). This fuel, which is described more precisely in ORNL/TM-6037,<sup>1</sup> was discharged from the H. B. Robinson-2 pressurized water reactor (PWR) on May 6, 1974; thus, it had cooled for ~5 years when the SETF campaigns were made.

Both first-cycle and plutonium second-cycle solvent extraction tests were made during Campaigns 1 and 2. Tests were made using 30% or 15% tri-n-butyl phosphate (TBP) in normal paraffin hydrocarbon (NPH) diluent. The solvent was used on a once-through basis because equipment limitations precluded solvent recycle; however, samples of the waste solvents were obtained for laboratory solvent washing tests. Reductive stripping of plutonium was accomplished using a variety of reducing agents, including HAN, hydrazine-stabilized HAN, ferrous sulfamate, and nitrous acid. Both uranium-plutonium costripping and partial partitioning (separation of part of the uranium from the uranium-plutonium product) methods were tested.

## 2. DESCRIPTION OF EQUIPMENT

The SETF equipment consists of seven process tanks located in the tank pit area of Cell 5 and three equipment racks in the cubicle area, which is a part of Cell 5 that is served by a viewing window and a pair of master-slave manipulators. The featured equipment is the rack containing the mixer-settlers (see Fig. 1). In addition, two racks of accessory equipment were provided. The first rack (shown in Fig. 2) contains a dissolver tank and feed preparation equipment, while the second contains solution transfer equipment, sampling devices, and an ion exchange purification system for plutonium.

The dissolver tank, which has a 12-L capacity, is equipped with a removable, perforated, stainless steel basket. The tank is jacketed for heating and cooling by means of a circulating water system. Off-gas from

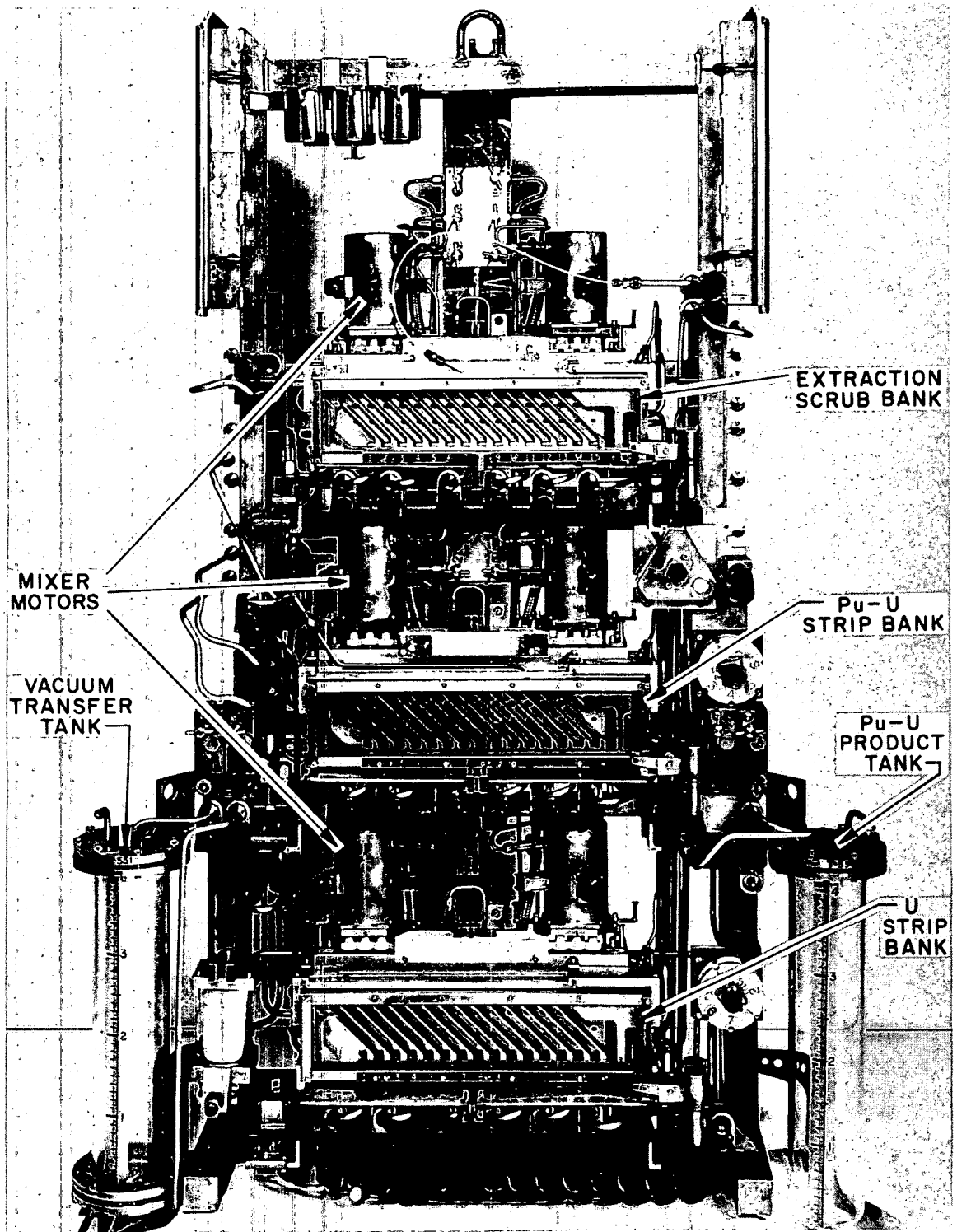


Fig. 1. SETF solvent extraction rack.

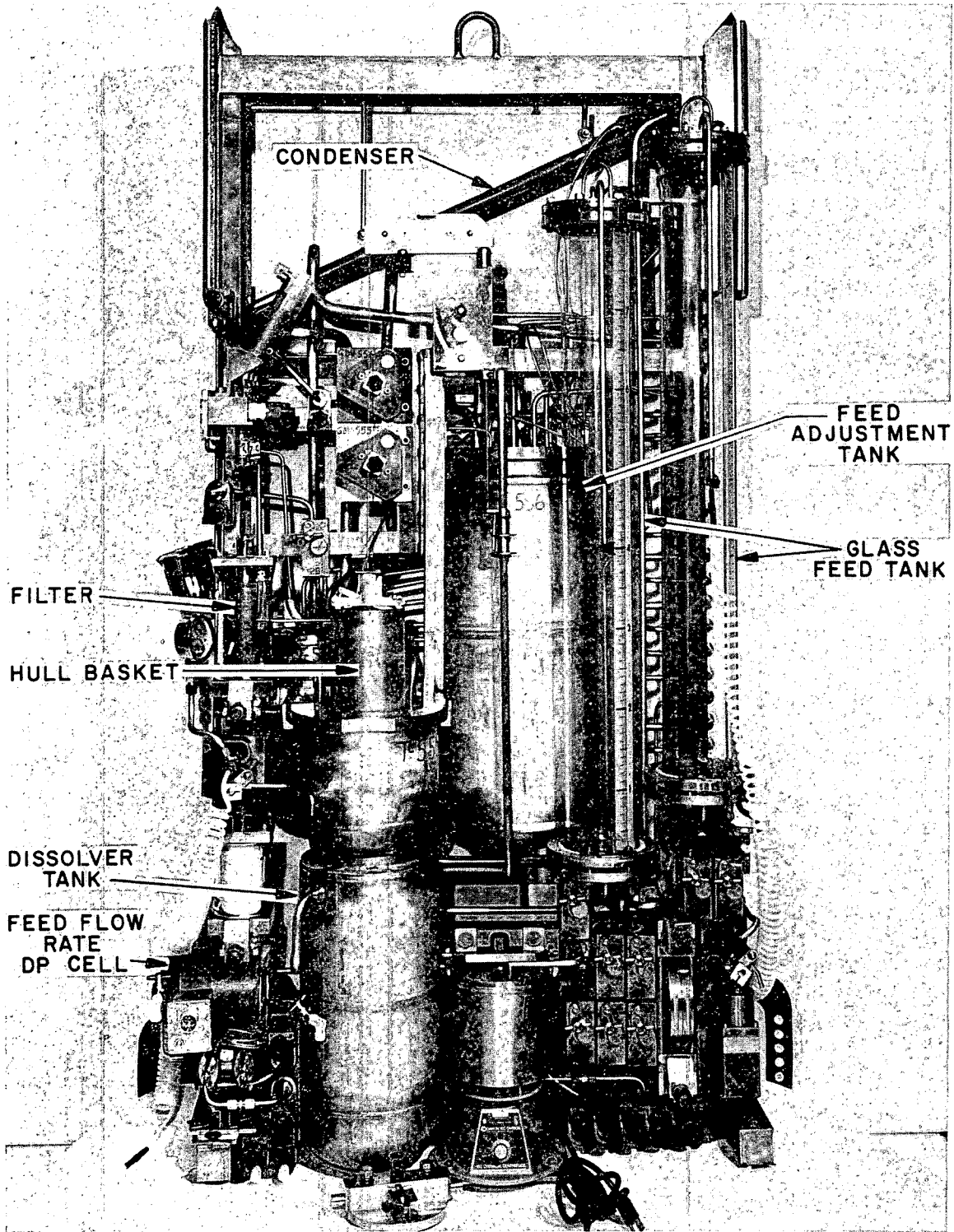


Fig. 2. SETF dissolution and feed preparation rack.

the dissolver can be routed through a condenser for either reflux or removal of condensate. The rack also includes a 20-L, jacketed, feed adjustment tank that is connected to the same off-gas system as the dissolver. Feed solution clarification equipment includes a laboratory centrifuge and several high-efficiency, etched-disc filters. Two calibrated, 8-L glass feed tanks are used with a dual feature enabling one tank to receive clarified solutions while the other is being used to supply feed solution to the mixer-settlers. The feed rate is determined by measuring the pressure drop across a capillary, using an in-cell differential pressure transmitter, and is controlled by regulating the air pressure applied to the feed tank.

The three banks of 16-stage mixer-settlers are vertically stacked onto a single equipment rack to allow gravity flow of the organic solution from one bank to the next. Each mixer-settler is fabricated of stainless steel with a quartz-glass front window, as shown in Fig. 3. These mixer-settlers are of the same geometrical design as those used in previous facilities at ORNL.<sup>2</sup> All of the settling chambers are located on the front side and thus can be observed through the window. Stage volumes are ~80 mL (30 mL for mixers and 50 mL for settling chambers). The organic solvent (light phase) is introduced into the mixer-settler at the right end, as illustrated in Fig. 3, and flows to the left, whereas the aqueous solution (heavy phase) enters at the left side and flows to the right. Mixed phase flow leaving each of the vertical mixing chambers (in the back part of the unit) flows into the corresponding settler (visible in the photo) via the middle ports. The organic solvent floats to the top of each slanted settler and overflows through a port back into the mixing chamber of the adjacent stage at the left. The aqueous solution in each settler sinks to the bottom and underflows through a weir plate back into the mixing chamber of the adjacent stage at the right. The interface levels are regulated by a hydrostatic leg in the aqueous solution outlet line.

Six calibrated head tanks fabricated of glass, which range in volumetric capacity from 2 to 20 L, are located outside the cell bank. An automatic flow control system is associated with each tank. Solvent, scrub, and strip solutions in the tanks are metered to the mixer-settlers.

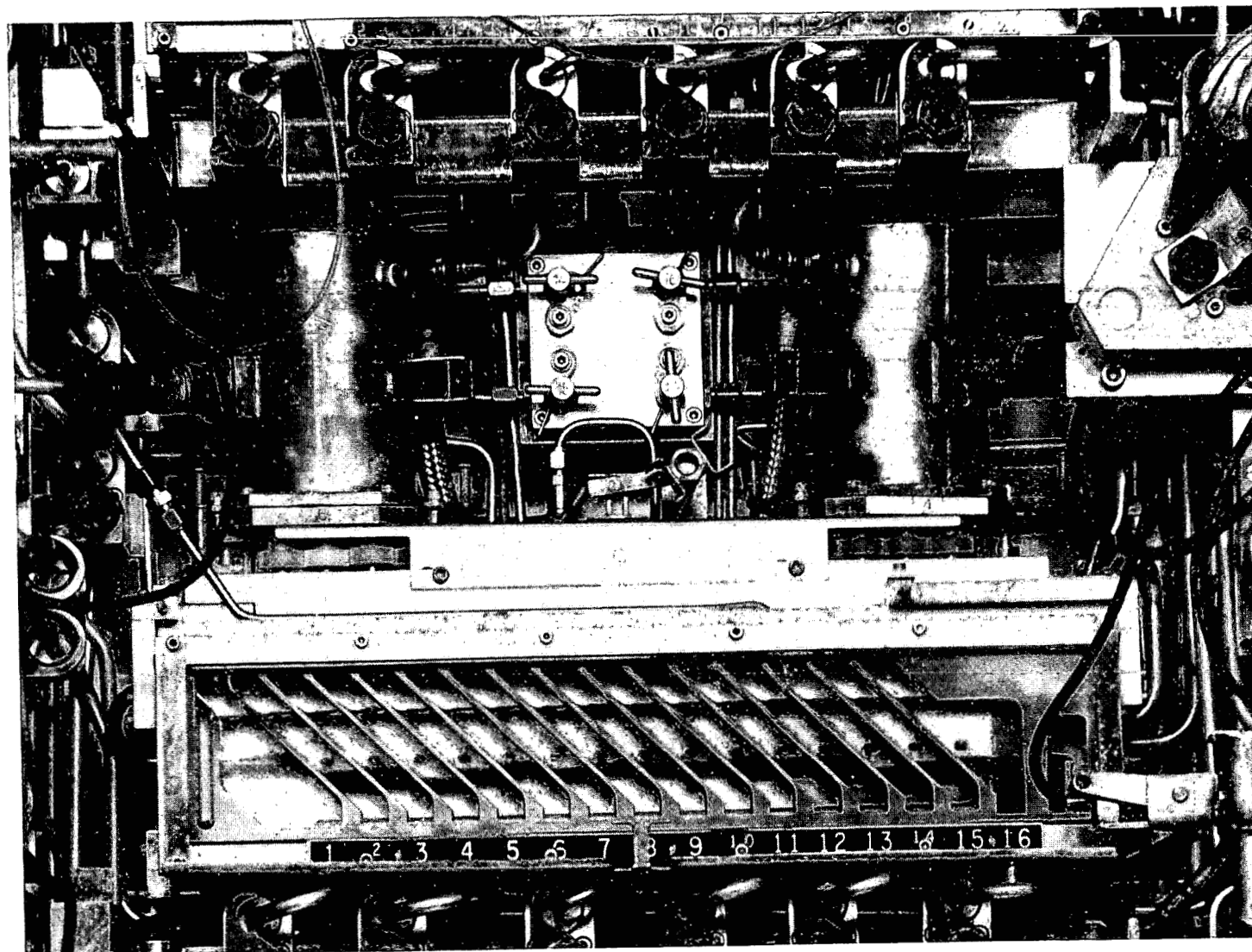


Fig. 3. Front view of SETF mixer-settler.

Effluent streams from the mixer-settlers are routed into catch tanks which have relatively small diameters. The rate of volume increase in the catch tank provides a measure of the effluent stream flow rates. The effluent catch tanks range in capacity from 2 to 100 L.

Samples of flowing streams are obtained by "thieving" with dip tubes into preevacuated bottles. A new dip-tube is used for each sample.

In-line, solid-state electronic detectors are used to continually monitor the alpha concentrations (which represent plutonium) in both the pregnant organic (AP) effluent solution from the extraction mixer settler and the aqueous (BP) product stream. The detectors are similar to those which have been used at TRU for many years in various transuranium element production processes. Direct output from the monitors (counts per second) is recorded on time charts and can be calibrated with periodic solution analyses to obtain plutonium concentration-time plots such as that shown in Fig. 4. These plots are useful for following the approach to steady-state operation and for observing process stability.

Also, a special sample loop, which includes flexible inlet and outlet lines, a circulating pump, and an alpha detector, can be used to continuously monitor the plutonium concentration in the organic phase of any selected settler stage. Typical data obtained with this system are included in Fig. 4. During the particular solvent extraction test run illustrated here, the feed stream entered the extraction bank at stage 7 and the aqueous waste (AW) stream exited at stage 16; the plutonium concentration in the organic phase of stage 12 was monitored during the first few hours and the last few hours of the run, and the concentration in stage 10 was monitored during the middle part of the run. Such data can be used to locate the characteristic "wave front" of the plutonium concentration profile within the extraction bank during the various test runs. The "wave front" location is more sensitive to changes in process parameters than is the end stream (AP) concentration and can provide the best signal for application of automatic control to the bank. This has been demonstrated in studies by Mills.<sup>3</sup> Automatic control was not applied in the SETF during Campaigns 1 and 2; however, the position of the "wave front" within the extraction bank was determined in some of the test runs and then was moved to the desired location by adjusting the inlet stream flow rates.

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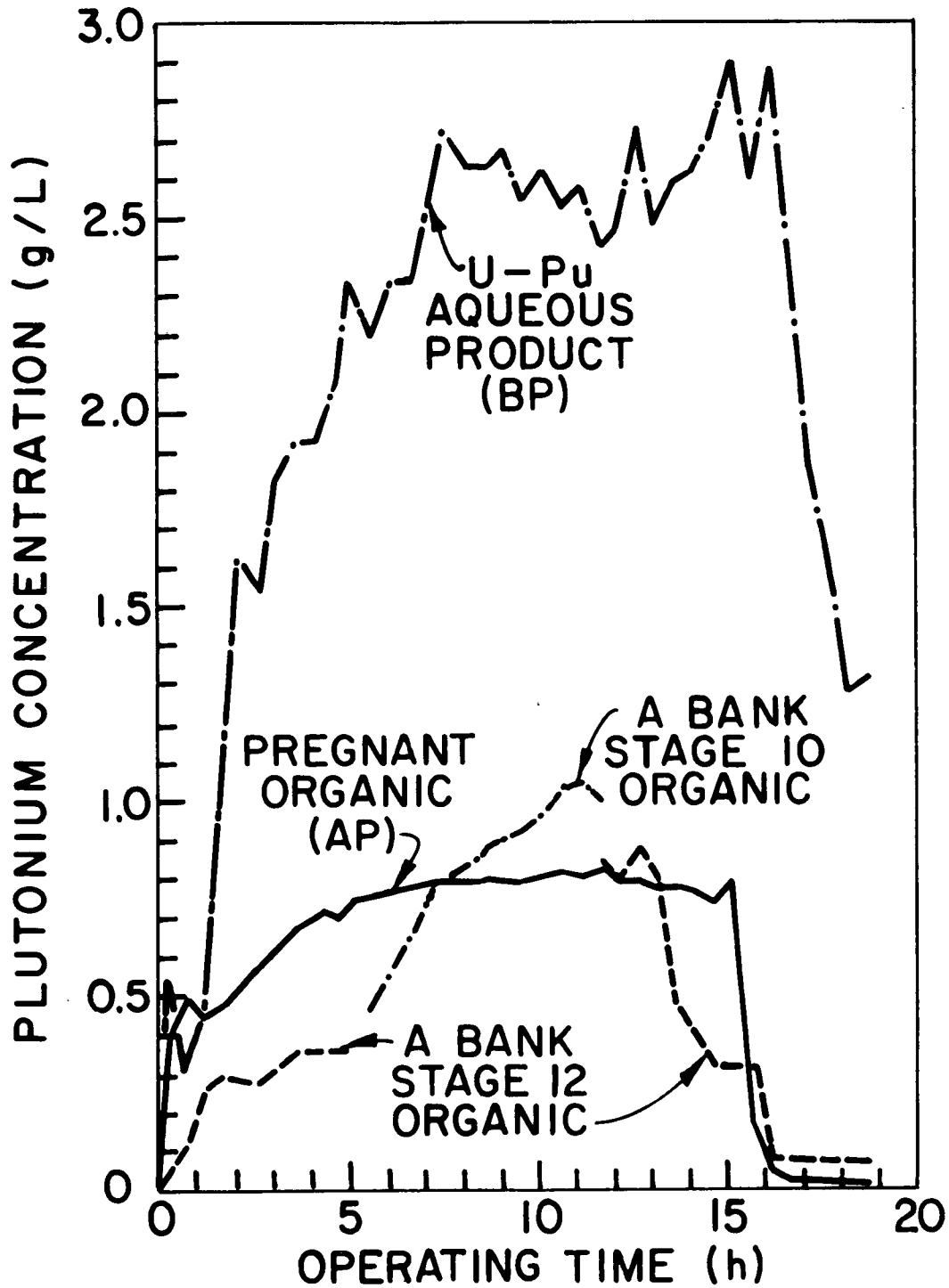


Fig. 4. Variations of plutonium concentrations in process streams.

One of the attributes of the SETF equipment is the ease of modification of the inlet and outlet flow paths to and from the mixer-settlers. This permits a variety of flowsheets to be tested and allows changes to be made, for example, in the number of mixer-settler stages used for a given operation. Flexible polyethylene sections are used in the aqueous solution supply lines, including the feed stream, so that these solutions can be introduced at any desired stage of the mixer-settlers. Polyethylene lines cannot be used for organic solutions, but sections of the rigid lines (jumpers) can easily be replaced to modify the routing.

### 3. OPERATING PROCEDURE

The procedure generally used to prepare for and support solvent extraction tests in the SETF is shown in Fig. 5.

#### 3.1 Disassembly and Shearing of Fuel Rods

Before reaching the SETF, an H. B. Robinson-2 PWR fuel assembly was disassembled at the Idaho National Engineering Laboratory and the Zircaloy-clad fuel rods (which had a diameter of  $\sim 1.0$  cm) were sectioned into lengths of  $\sim 2$  m. The sectioned rods, containing a total of  $\sim 50$  kg of heavy metal, were packaged and shipped to the ORNL High Radiation Level Examination Laboratory (HRLEL). A hand-operated, single-rod, hydraulic shear was used at the HRLEL to cut the rods into lengths of  $\sim 5$  cm. Sheared pieces and dislodged powder containing  $\sim 2$  kg of heavy metal (the desired amount for one dissolver batch) were weighed and then packaged into small cans. Up to five of these small cans were loaded into an aluminum, screwed-top, 12.1-cm-diam shipping container and were transferred to TRU in the HRLEL Dry Storage Carrier. Five shipments were required to transfer the 50 kg of heavy metal.

#### 3.2 Dissolution

The heavy metals contained in the fuel (2 kg per batch) were dissolved by using a variation of a procedure developed at the Savannah River Laboratory (SRL) for IWR fuels.<sup>4</sup> The SETF procedure required  $\sim 8$  to 10 h and consisted of the following steps:



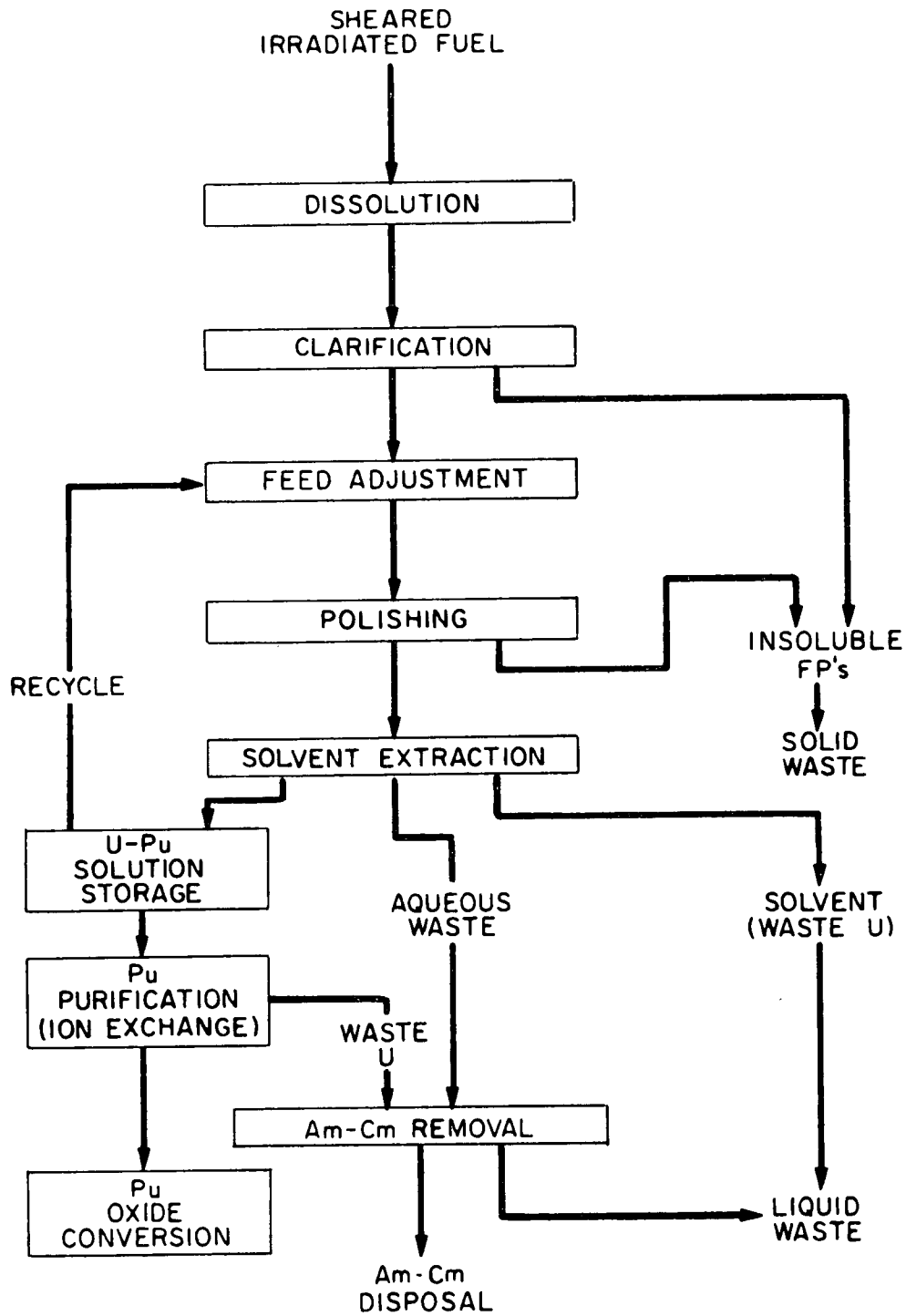


Fig. 5. SETF operating procedure.

1. Approximately 2 L of 3 M HNO<sub>3</sub> was added to the dissolver tank. (This addition was not made if the tank already contained a similar volume of dilute HNO<sub>3</sub> left from rinsing the previous batch of dissolver hulls.)
2. The empty basket was installed in the dissolver tank.
3. The fuel can was opened (with a can opener), and the sheared pieces and dislodged powder were poured into the basket.
4. The tank lid was closed, and the addition of 11 M HNO<sub>3</sub> was begun at a controlled rate of ~2 L/h. The dissolver solution was simultaneously heated to 50°C.
5. The temperature was held at 50°C during the acid addition, which was continued until the dissolver contained 4.5 mol of HNO<sub>3</sub> per mol of uranium. SRL data<sup>4</sup> have shown that the HNO<sub>3</sub> concentration of the solution generally ranges between 3 and 4 M during the acid addition period and that ~60% of the uranium is dissolved.
6. The dissolver solution was then heated to 90°C and held for 2 h to complete the dissolution. Final concentrations were ~350 g/L of uranium and 2.3 M HNO<sub>3</sub>.
7. The solution was cooled and transferred through a filter to the feed adjustment tank. A 1- $\mu$ m- or a 3- $\mu$ m-rated high-efficiency filter was used in some of the tests; however, only a 50- $\mu$ m-rated screening was used in the others.
8. The hulls were rinsed with 2 L of 3 M HNO<sub>3</sub>, and the rinse solution was left in the dissolver tank for inclusion with the next batch. The basket was lifted partially out of the tank, the hulls were allowed to drain until dry, and the dried hulls were poured back into the original can for disposal.

### 3.3 Feed Adjustment

The procedure normally used for feed adjustment consisted of two steps. First, the plutonium was adjusted to the tetravalent state by sparging the solution with N<sub>2</sub>O<sub>3</sub> gas at ambient temperature (~35°C) for 2 to 5 h. [Usually, about 80% of the plutonium was already present as Pu(IV); sparging was continued until more than 95% of the plutonium was Pu(IV).] The second adjustment involved adding the necessary volume of dilute nitric

acid to increase the acidity and reduce the heavy-metal concentration of the feed solution to the desired levels.

### 3.4 Clarification of the Feed Solution

Operational difficulties and inefficient results were obtained during initial clarification attempts with the laboratory centrifuge. Therefore, all feed solution clarifications during Campaigns 1 and 2 were made by means of filtration, using a stainless steel, etched-disc filter (obtained from Vacco Industries, South El Monte, California) which had a pore size of 1  $\mu\text{m}$  and a surface area of  $\sim 150 \text{ cm}^2$ . The filter was precoated with a silica-based filter aid, using  $\sim 1.5 \text{ g}$  of precoat per kg of uranium being processed, in order to achieve a satisfactory filtration rate. Two types of filter aids were used to precoat the filter. First, a layer of coarse material (Johns-Manville Celite 535) was applied, and this was then covered with a finer material (Johns-Manville Standard Super Cel); the weight ratio of coarse to fine material was  $\sim 6$ . Average filtration rates during the clarification were 9 to 37  $\text{mL}\cdot\text{s}^{-1}\cdot\text{m}^{-2}$ . The effectiveness of the clarification was determined by centrifuging a sample of the filtered solution in a clinical batch centrifuge. The feed solution was refiltered when solids were observed in the sample.

### 3.5 Solvent Extraction

The solvent extraction equipment was arranged in a variety of ways, depending on the particular flowsheet being tested. A schematic diagram of a typical arrangement of the equipment is shown in Fig. 6; this particular arrangement provided for coextraction and scrubbing of uranium and plutonium in the A-bank, partial partitioning in the B-bank, and uranium stripping in the C-bank.

When the effluent stream catch tanks became full during a run, the solutions were rapidly transferred by means of the vacuum-pressure transfer tanks, T-551, T-552, and T-553. The plutonium product solution (B-bank aqueous effluent stream) was transferred to the evaporator-storage tank, T-503. The organic waste solution (C-bank organic effluent) was sampled to verify that the plutonium concentration was sufficiently low to permit

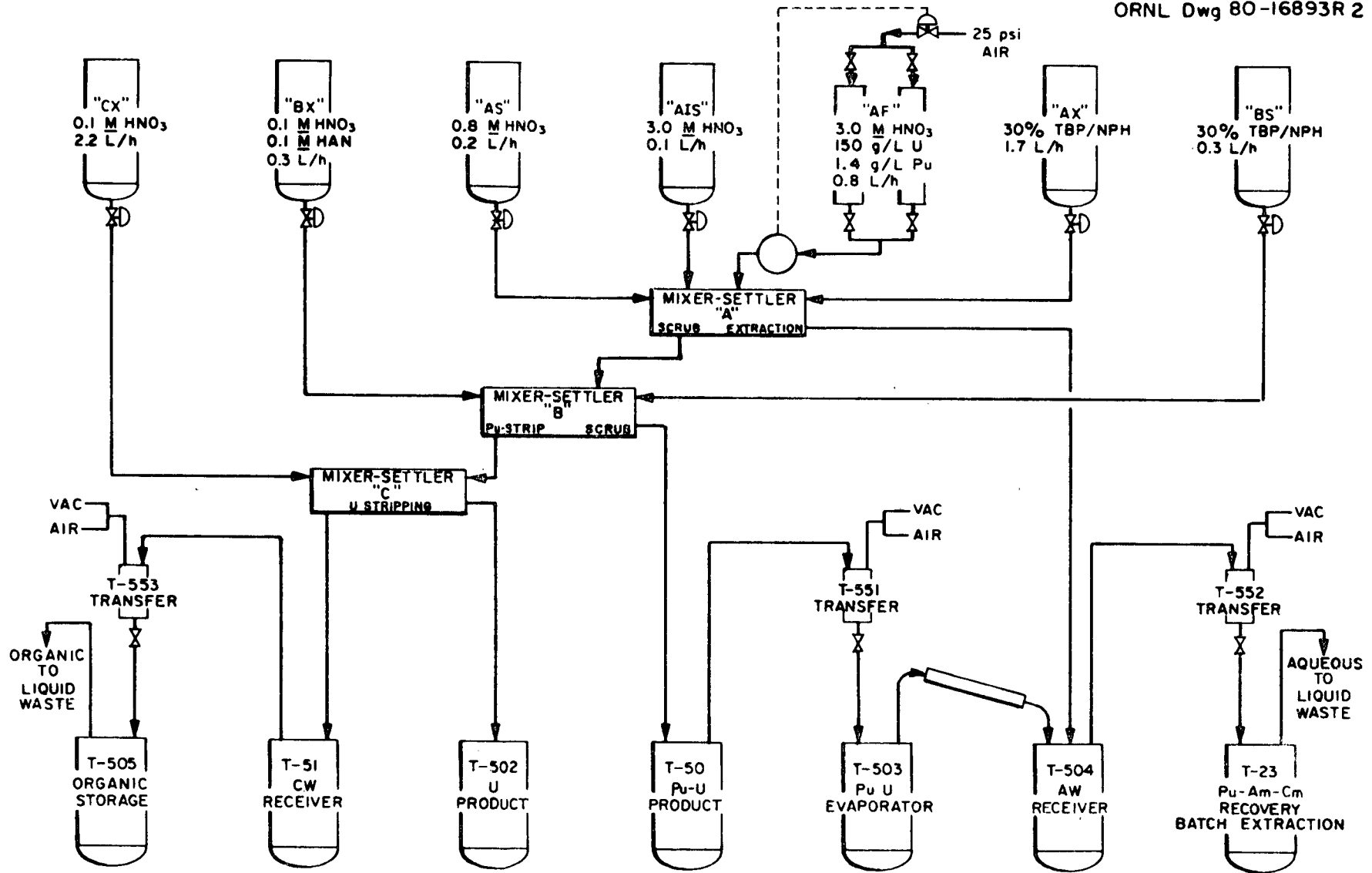


Fig. 6. Schematic flowsheet for a typical solvent extraction test.

retransfer to the ORNL Intermediate Level Waste (ILW) System. The aqueous waste solutions (the A-bank and the C-bank aqueous effluents) were transferred to the transuranium-element processing equipment in TRU for recovery of plutonium, americium, and curium (the long-lived alpha emitters) by means of the Pubex and Cleanex batch extraction processes,<sup>5</sup> which utilize di-2-ethylhexyl phosphoric acid as the extractant. This treatment was necessary to remove excessive amounts of long-lived alpha-emitting isotopes from the waste solutions before they were released to the ORNL ILW System.

#### 4. EXPERIMENTAL CONDITIONS, RESULTS, AND DISCUSSION

During Campaigns 1 and 2, the areas of the coprocessing solvent extraction flowsheets that received particular attention included (1) extraction/scrubbing operations, (2) reduction of tetravalent plutonium, (3) stripping operations, and (4) solvent degradation measurements. The operating conditions for each test run and the resulting outlet stream compositions are tabulated in the Appendix. One test run (2-4) was made to demonstrate the first two cycles (codecontamination and partitioning) of the Hot Engineering Facility (HEF) flowsheet. The results of this test, tabulated in the Appendix, are discussed in detail in a report<sup>6</sup> which covers other demonstrations of the HEF flowsheets.

##### 4.1 Extraction/Scrubbing Operations

The areas of concern in the extraction/scrubbing operations included (1) the formation of cruds in the extraction bank, (2) losses of uranium and plutonium in the extraction bank raffinate stream (AW), and (3) fission product decontamination.

###### 4.1.1 Crud formation

In all first-cycle tests, black interfacial crud formed in the extraction stages of the A-bank mixer-settler, most prominently near the feed inlet. The crud is believed to be an emulsion, stabilized by minute particles of insoluble fission product metals. Significant amounts of ruthenium, molybdenum, and noble metal fission products, which were not soluble in the nitric-acid dissolver solutions, were removed from the SETF feed solutions via filtration. Some of the insoluble particles were apparently

small enough to pass through the feed clarification filter, which had 1- $\mu$ m pores. During plutonium second-cycle runs, in which the feed solutions contained a relatively small concentration of fission products, cruds did not form in the mixer-settlers.

Tests made in laboratory glassware showed that the emulsion-crud formed more readily when the aqueous feed and organic solvent solutions were mixed by a swirling motion (such as probably occurred in the mixer-settlers) rather than when the mixing was accomplished by a more turbulent, axial shaking motion (such as probably occurs in a pulsed column contactor). In the SETF mixer-settlers, the accumulated emulsion-cruds were sticky and not easily dispersed. The cruds were removed periodically from the settling chambers by vacuuming after extensive agitation and flushing.

Crud formation was prevented during small-scale mixer-settler runs at SRL by using feed solutions that had been clarified with the aid of a flocculating agent. Either manganese dioxide or an organic polyamine, Primafloc C-3 (manufactured by the Rohm and Haas Company), was successfully utilized.<sup>7</sup> However, the use of Primafloc C-3 did not prevent the formation of cruds in SETF operations. Manganese dioxide was not tried.

Recently, ~2.5 metric tons of spent fuel from the Point Beach-1 BWR was processed in small-scale (4.5-cm-diam) pulsed columns at the Pacific Northwest Laboratory (PNL).<sup>8</sup> The dissolver solutions were clarified by means of a sintered-metal filter rated at 100  $\mu$ m. Based on SETF experience, a filter of that size would not be expected to retain the insoluble metal particles; even so, no crud accumulation sufficient to cause a shutdown was noted at PNL during the several months of operation. This experience appears to confirm that crud formations similar to those observed in the SETF and SRL mixer-settlers do not pose a serious problem in pulsed column contactors.

A number of methods for the prevention of crud formation were tested during the SETF runs. These included (1) the use of a longer digestion time (6 h) at 90°C during the fuel dissolution, (2) prewashing of the adjusted feed solution with an organic solution (NPH) before clarification, (3) operation of the extraction-scrub bank at a high temperature (50°C), and (4) operation with dilute TBP (15%). None of these methods proved to be successful.

A significant reduction of the crud formation was obtained during one test run (2-4A) in which the dissolver solution was clarified, the heavy-metal concentration was reduced to 150 g/L, and the solution was digested with hydrogen peroxide at 90°C. The objective of this procedure was to dissolve any metal colloids remaining in the clarified feed solution. The peroxide treatment also affected the behavior of ruthenium in the solvent extraction stripping operation, as described in Sect. 4.1.3.

#### 4.1.2 Extraction losses

The losses of uranium and plutonium to the extraction raffinate (AW) streams in the first-cycle runs were within the range of 0.0002 to 0.07% in the Campaign 1 and 2 tests. The median values were 0.004 and 0.007%, respectively, for uranium and plutonium. These values are considered sufficiently low for practical purposes but are somewhat higher than those predicted by SEPHIS<sup>9</sup> code calculations.

The most serious potential for large extraction losses occurs when the heavy metal (uranium and plutonium) concentration in the organic solvent is maintained at a high level (e.g.,  $\geq 75\%$  of saturation) in an effort to obtain high decontamination factors (DFs) from the fission products. Under these conditions, the system is susceptible to upset as a result of slight changes in flow rates or concentrations of the inlet streams which might cause internal refluxing of the heavy metals. This phenomenon was not studied in these experiments, but a series of SEPHIS calculations was performed to illustrate this problem. The results are plotted in Fig. 7.

These calculations were made for different feed flow rates with all other conditions held constant; however, the same effects would result from small reductions in the extractant flow rate. These assumed changes are quite small: a variation of only 3.4% in the intended flow rate (or perhaps only 1.7% each in the feed and extractant flow rates in the "wrong" direction) could cause a serious upset in the concentration profile in the system. It is very difficult in plant-scale equipment to establish and maintain a flow rate with such a high degree of accuracy and precision for an extended period of time.

Thus it appears imperative, when operating a Purex flowsheet at  $\geq 75\%$  of solvent saturation, to monitor the heavy metal concentrations within the

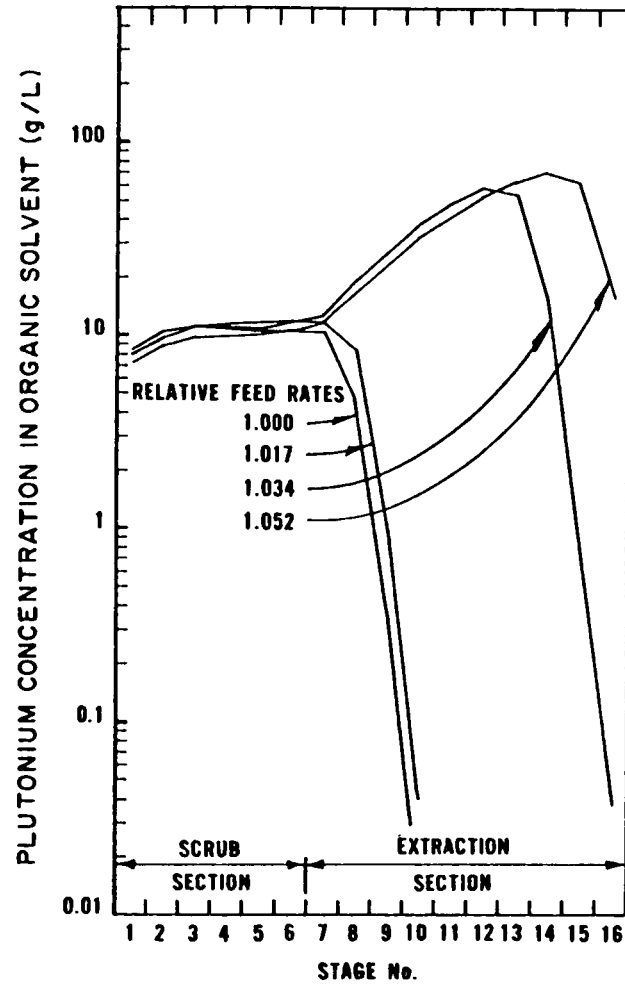
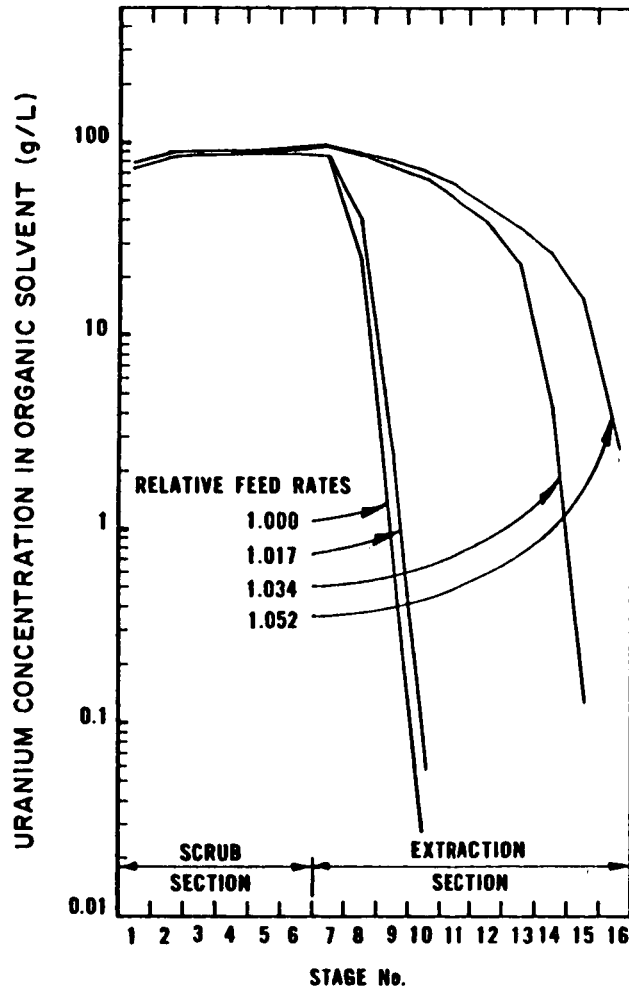


Fig. 7. SEPHIS calculations showing the effect of slight changes in feed flow rate during operations with high heavy-metal concentrations in the solvent.



extraction contactor continuously and to adjust the flow rates as required to keep the concentration "wave front" far enough from the raffinate outlet to maintain acceptably low losses.

In the SETF, plutonium concentration in the solvent is continuously monitored at a suitable location within the extraction section by means of an alpha detector located in the special sampling loop described in Sect. 2. This provides information which allows proper adjustment of the feed and extractant flow rates. An alpha monitor in the AW stream would be useless because the plutonium alpha contribution would normally be overwhelmed by that of the americium and curium. A continuous monitor on a side stream would probably be necessary in any plant system, since it is doubtful that periodic sampling and analysis of the plutonium concentration would provide information on a sufficiently timely basis to maintain proper control of the flow rates.

#### 4.1.3 Fission product decontamination

Fission product behavior in the Purex process is generally well known. However, to reillustrate this behavior, a graph of fission product concentration profiles in the SETF extraction/scrub bank during test 1-3 is shown in Fig. 8. In this test, seven scrub stages and nine extraction stages were used, the feed and scrub solutions were both 3 M HNO<sub>3</sub>, and the operating temperature was 40°C. Cesium extraction was at least two orders of magnitude lower than the other contaminants. Ruthenium and the rare earths were extracted to the same degree, but the rare earths were more effectively removed in the scrub section of the bank. No data were obtained for zirconium and niobium because the <sup>95</sup>Zr-<sup>95</sup>Nb in the fuel had decayed to levels that could not be measured after the long cooling period (4 to 5 years).

Fission product decontamination was not studied per se in the SETF tests; however, measurements were made at a variety of operating conditions (see Tables A-1 and A-6 in the Appendix) to determine the general range of DFs obtained while processing LWR fuel. The values obtained in the first-cycle solvent extraction tests are listed in Table 1. These are combined DFs obtained during the extraction, scrubbing, and plutonium stripping (either partial partitioning or costripping) operations of each test.

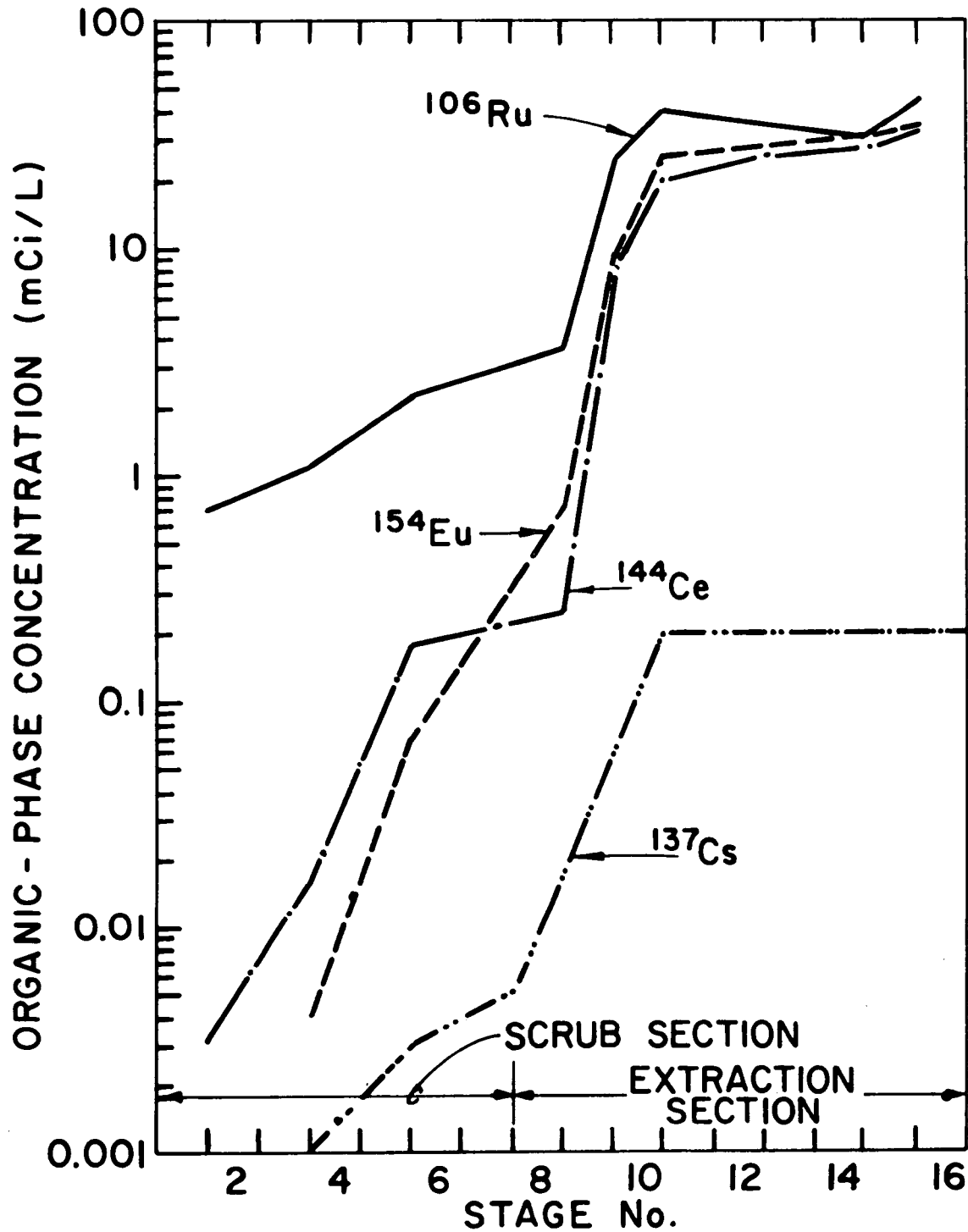


Fig. 8. Fission product concentration profiles in the extraction/scrub bank during test 1-3. All values for  $^{137}\text{Cs}$  are at the limit of detection.

Table 1. Fission product decontamination in first-cycle tests

Test No.	Overall <sup>a</sup> DFs		
	<sup>106</sup> Ru	<sup>137</sup> Cs	<sup>144</sup> Ce
1-1	7.8E3 <sup>b</sup>	2.9E5	>8.9E4
1-2	>4.7E3	6.7E4	4.8E4
1-3	1.0E4	9.3E4	>2.7E4
2-2A	1.4E5	4.9E5	1.5E5
2-2B	1.9E5	3.2E5	2.2E5
2-2C	2.4E5	4.9E5	3.2E5
2-2D	2.4E5	9.1E5	3.7E5
2-2E	3.6E4	8.2E5	>1.3E6
2-4A	3.7E3	2.1E5	2.5E5

<sup>a</sup>Combined DFs obtained during extraction, scrubbing, and stripping operations.

<sup>b</sup>To be read as  $7.8 \times 10^3$ .

In general, the cesium and cerium DFs were in the range of  $10^4$  to  $10^6$ , while the ruthenium DFs ranged from  $10^3$  to  $10^5$ . All of the cesium and cerium decontamination occurred in the extraction/scrub contactor, whereas a small, but significant, portion of the ruthenium decontamination occurred in the strip contactor (see Table 2). This was true in all tests except 2-4A. The lack of decontamination in test 2-4A can be associated with the temperature in the strip contactor ( $\sim 50^\circ\text{C}$ ), which was higher than in the other tests ( $\sim 30^\circ\text{C}$  and  $\sim 40^\circ\text{C}$ ), and with the use of a hydrogen peroxide digestion step used in the feed adjustment procedure for that test (discussed in Sect. 4.1.1). The latter effect has also been observed in subsequent SETF test runs. In the extraction/scrub contactor, the <sup>106</sup>Ru DFs were consistently in the range of  $10^3$  to  $10^4$ .

The major gamma emitter in most of the product solutions was not a fission product. It was <sup>239</sup>Np, the decay daughter of <sup>243</sup>Am (see Table 3). At the operating conditions used in the SETF tests, much of the neptunium followed the plutonium through the extraction, scrubbing, and stripping operations. Although the <sup>239</sup>Np decays rapidly (half-life = 2.35 d) after being separated from the <sup>243</sup>Am, it emits a relatively strong gamma (0.28 MeV). Thus, the presence of <sup>239</sup>Np in fresh, first-cycle plutonium product solutions should be considered in the design of shielding for the stripping contactor and product tanks.

Table 2. Ruthenium decontamination in first-cycle tests

Test No.	<sup>106</sup> Ru DFs		Total
	Extraction/scrub	Strip	
1-1	1.1E3 <sup>a</sup>	6.9	7.8E3
1-2	7.0E2	>6.8	>4.7E3
1-3	1.6E3	6.4	1.0E4
2-2A	8.0E3	17.9	1.4E5
2-2B	5.6E3	34.4	1.9E5
2-2C	2.9E3	84.7	2.4E5
2-2D	1.6E4	15.2	2.4E5
2-2E	1.8E3	19.4	3.6E4
2-4A	3.2E3	1.2	3.7E3

<sup>a</sup>To be read as  $1.1 \times 10^3$ .

Table 3. Gamma-emitting components in typical SETF product solutions

Component	Activity level ( $\mu\text{Ci/g Pu}$ )
<sup>106</sup> Ru	141
<sup>110m</sup> Ag	<0.3
<sup>125</sup> Sb	<1.4
<sup>134</sup> Cs	4.6
<sup>137</sup> Cs	16
<sup>144</sup> Ce	<14
<sup>154</sup> Eu	<0.7
<sup>239</sup> Np	1873

## 4.2 Reduction of Tetravalent Plutonium

### 4.2.1 Use of HAN as the plutonium reductant

Flowsheets for the Hot Engineering Facility (HEF) call for the use of hydrazine-stabilized hydroxylamine as the plutonium reductant. Thus, one of the primary objectives of the SETF tests during Campaigns 1 and 2 was to establish the necessary operating conditions for the effective use of HAN and to determine whether stabilization with hydrazine is necessary. Elimination of the need for hydrazine in a reprocessing plant is desirable because of a potential safety hazard — the formation of highly explosive compounds, such as hydrazoic acid and metal azides.

Hydrazine is used as a holding reductant for Pu(III) after the latter is formed by the reaction of Pu(IV) with the HAN. The hydrazine reacts with any nitrous acid that may be generated within the system and thus prevents the reoxidation of Pu(III) by nitrous acid. Theoretically, HAN should also be an efficient nitrous acid scavenger; however, unstabilized HAN has been reported to be inadequate.<sup>10</sup> Contrarily, two SETF experiments showed that HAN can act as a sufficient holding reductant — if the aqueous-phase acidity is kept sufficiently low —  $\sim 1 \text{ M HNO}_3$ , or lower.

The results of the first experiment are shown in Fig. 9. In that experiment,  $\sim 90 \text{ L}$  of a  $1 \text{ M HNO}_3$  solution containing 135 g of plutonium was sparged with  $\text{N}_2\text{O}_3$  for  $\sim 60 \text{ h}$  at 25 to  $30^\circ\text{C}$ . The excess HAN was destroyed progressively, and the Pu(III) was not significantly oxidized until the HAN had been essentially destroyed.

In the second experiment, two solvent extraction test runs were made using unstabilized HAN as the plutonium reductant. In each run, the HAN/plutonium mole ratio was 4, and in one of the test runs (2-3B) a significant amount of nitrous acid was added to the system to adjust the nitrous acid/plutonium mole ratio to  $\sim 1.0$ . Figure 10 shows the plutonium and nitric acid concentration profiles in the stripping contactor. These results indicate that the plutonium stripping effectiveness was not decreased by the presence of the nitrous acid. The plutonium concentration profiles were nearly identical; 99.993% of the plutonium was stripped in each run. The maximum aqueous-phase acidities were 0.53 and  $0.56 \text{ M HNO}_3$ , respectively, in the two runs.

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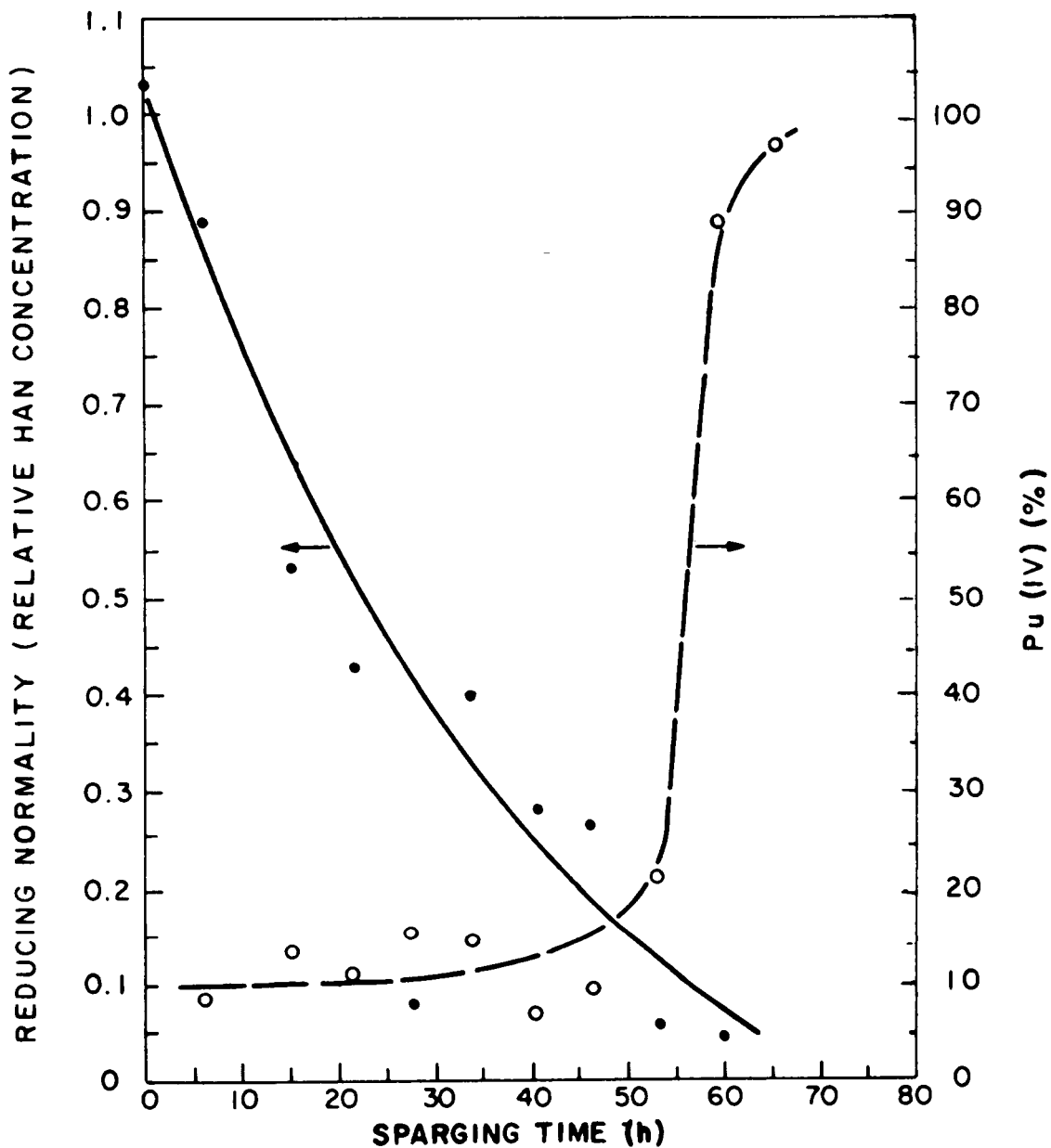


Fig. 9. Destruction of HAN and oxidation of Pu(III) by means of sparging with  $N_2O_3$  at 25 to 30°C.

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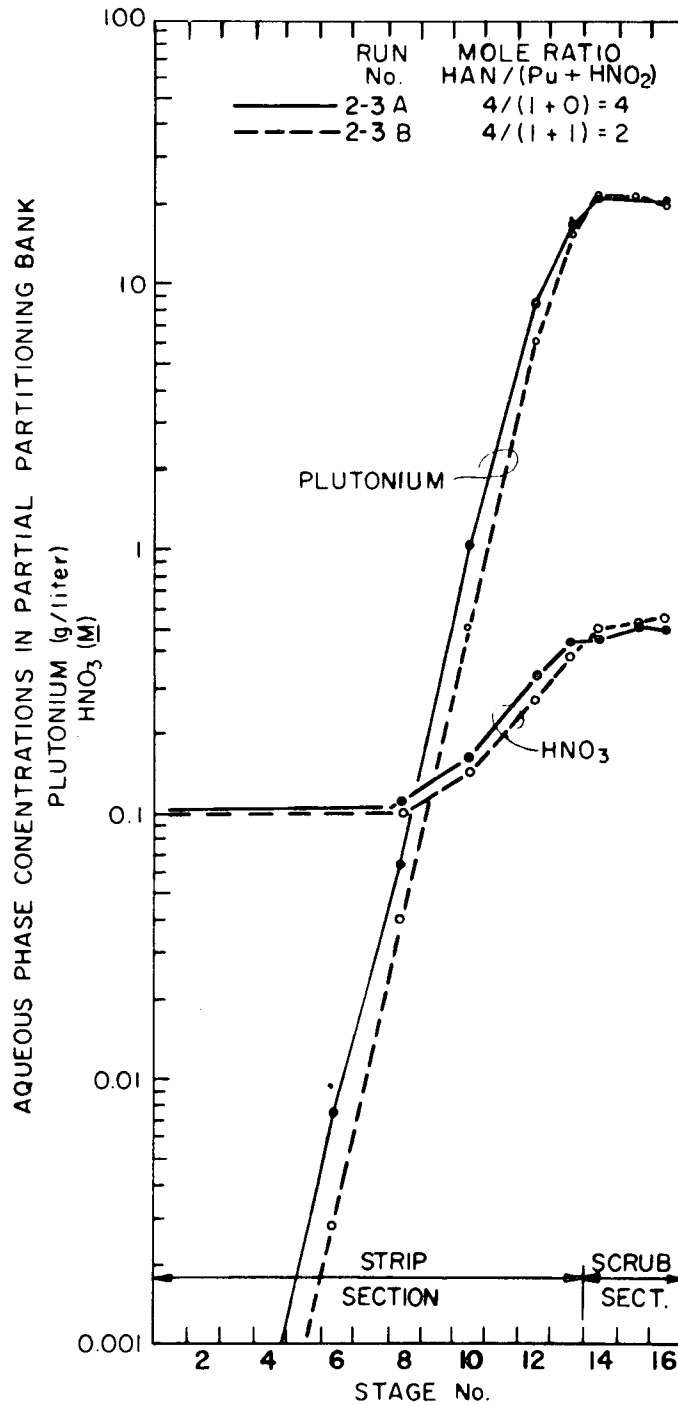


Fig. 10. Effect of nitrous acid on reductive stripping of plutonium using HAN.

The maximum aqueous-phase acidity is probably the key variable in determining whether HAN is a satisfactory holding reductant for Pu(III). In a test at SRL in which HAN did not prevent plutonium reflux (reoxidation and reextraction), the maximum aqueous-phase acidity was 2.3 to 2.5 M.<sup>11</sup>

Barney,<sup>12</sup> at HEDL, has reported that the rate of reduction of plutonium by HAN is inversely proportional to the fourth power of the aqueous acidity. Empirical data from ORNL laboratory experiments associated with SETF operations (shown in Fig. 11) are in fair agreement. Based on the stripping coefficients shown in Fig. 11 and the low aqueous/organic phase ratio (<0.5) normally used in partitioning, a relatively large number of stages would be required to strip 99.9% of the plutonium at acidities greater than ~1.5 M HNO<sub>3</sub>. This is probably because the rate of reduction is slower than the rate of reoxidation at these acidities. Thus, in a countercurrent partitioning contactor, plutonium will be stripped in the region where the aqueous acidity is low enough for HAN to be effective; however, if the aqueous strip solution containing Pu(III) subsequently passes into a region of the contactor where the acidity is high enough to make HAN ineffective, the rate of reoxidation and reextraction will probably exceed the rate of reductive stripping and cause plutonium reflux to occur.

Furthermore, at acidities below ~0.5 M HNO<sub>3</sub>, the redox potential of the plutonium-HNO<sub>3</sub>-HNO<sub>2</sub> system has been reported to be such that nitrous acid causes reduction of Pu(IV) to Pu(III).<sup>13,14</sup> SETF tests, described below, have substantiated this report. Thus, when HAN is being used at an acidity of 0.5 M or lower, the plutonium reduction will not be countered by the presence of nitrous acid. However, the nitrous acid will destroy an equivalent amount of HAN.

The use of unstabilized HAN to accomplish reductive stripping during partial partitioning operations was evaluated extensively during tests 2-1A through 2-1J, which comprised a three-variable factorial design. The following variables were evaluated: (1) maximum aqueous-phase acidity (at levels of 0.3 to 0.5 and 0.75 to 1.1 M HNO<sub>3</sub>), (2) temperature (at 40°C and 50°C), and (3) the presence of excess HAN or excess nitrous acid. When excess HAN was present, the HAN/(Pu + HNO<sub>2</sub>) mole ratio ranged from 5 to 10; when excess HNO<sub>2</sub> was present, the mole ratio was 0.6 to 1.0.



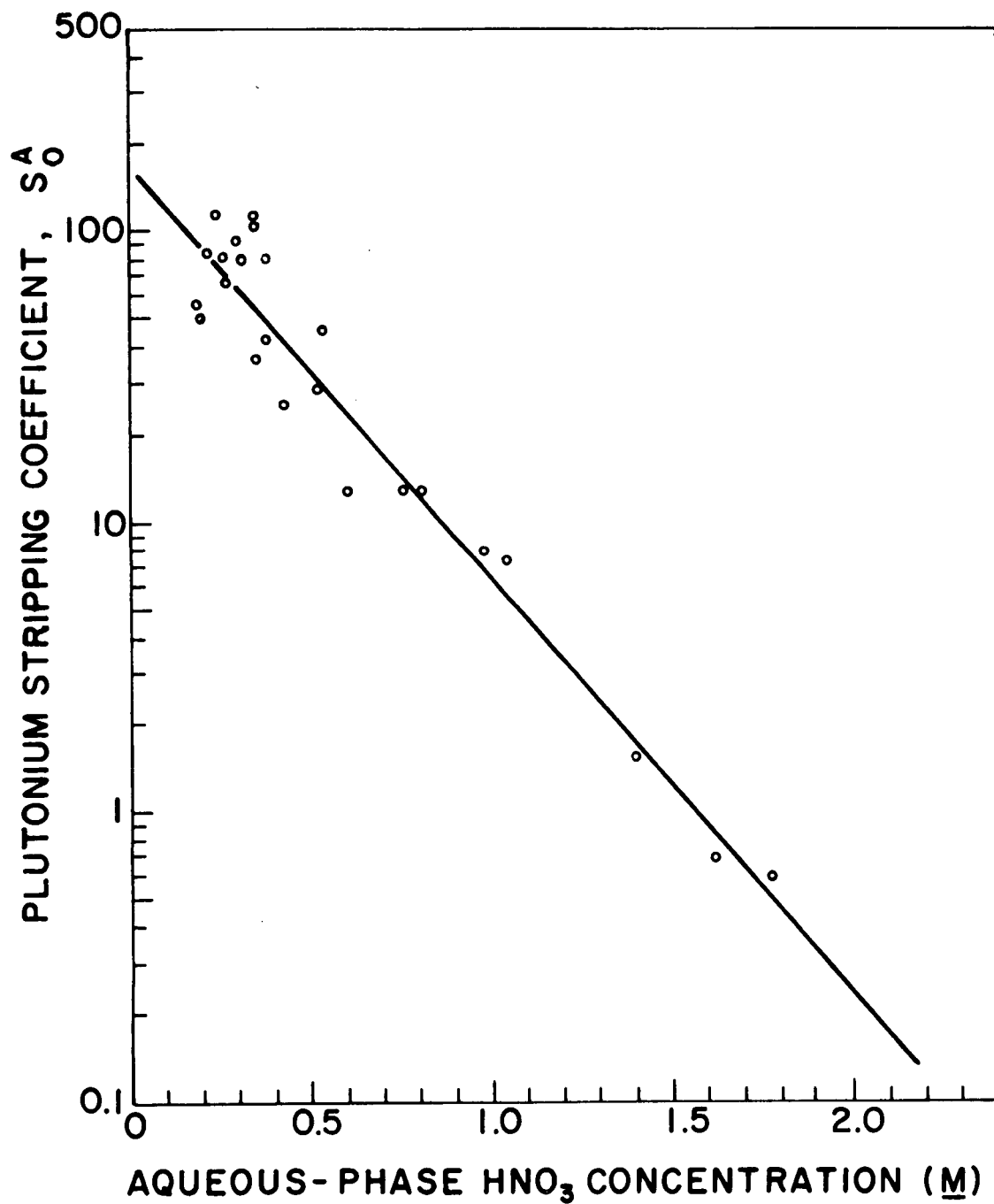


Fig. 11. Effect of aqueous-phase acidity on stripping coefficients when using HAN for reduction of Pu(IV).

The effect of increasing the maximum aqueous-phase acidity up to 1.1 M  $\text{HNO}_3$  appeared deleterious to the yield of stripped plutonium, as shown in Table 4; however, the effect was insignificant when excess HAN was present. Table 5 shows that the effect of increasing the temperature from 40°C to 50°C was small when excess HAN was present and when excess nitrous acid was present at low acidity (<0.5 M  $\text{HNO}_3$ ). Table 6 shows that the effect of changing from excess HAN to excess nitrous acid was deleterious in every case, but the severity of this effect was much greater at the higher acidities.

Overall, the results obtained during Campaigns 1 and 2 and in associated studies show that unstabilized HAN can be used effectively for the reductive stripping of plutonium if the maximum aqueous-phase acidity is maintained below ~1 M  $\text{HNO}_3$  (preferably below 0.5 M  $\text{HNO}_3$ ) and a sufficient excess of HAN is used to react with any nitrous acid that may be present in the system.

#### 4.2.2 Use of nitrous acid as the plutonium reductant

SETF tests 2-2A through 2-2E substantiated that nitrous acid could be used as a reductant for tetravalent plutonium, as reported by Bathellier<sup>13</sup> and Tsujino et al.<sup>14</sup> In the SETF tests, plutonium and part of the uranium were stripped from the pregnant organic while using 0.1 M  $\text{HNO}_3$  as the aqueous strip solution and a temperature of ~30°C in the stripping contactor. Nitrous acid was added to the system by dissolving it in the organic backscrub (BS) solution. [In two of the tests, nitrous acid was also dissolved in the extraction solvent (AX) solution.]

Concentrations of >0.3 M  $\text{HNO}_2$  were dissolved into 30% (1.09 M) TBP/NPH; however, the solution had to be stored in closed containers to avoid apparent decomposition of the nitrous acid to gaseous nitric oxide (NO). Some of the nitrous acid was lost from the vented mixer-settlers, but the organic stream (BU) leaving the stripping contactor still contained an excess of nitrous acid (>1 mol per mol of plutonium entering the contactor).

The plutonium concentration profiles in the stripping contactor during test 2-2C are shown in Fig. 12, along with the Pu(IV) concentration profiles calculated by means of the SEPHIS<sup>9</sup> computer code. The stripping effectiveness during test 2-2C was obviously better than that predicted for Pu(IV).

Table 4. Effect of maximum aqueous-phase acidity in the strip bank  
on the yield of stripped plutonium

Conditions of comparison	Stripped plutonium yield (%)	
	Low acidity <sup>a</sup>	High acidity <sup>b</sup>
Excess HAN, 40°C	99.6 (0.28 <u>M</u> HNO <sub>3</sub> )	98.8 (0.78 <u>M</u> HNO <sub>3</sub> )
Excess HAN, 50°C	99.8 (0.30 <u>M</u> HNO <sub>3</sub> )	99.6 (0.74 <u>M</u> HNO <sub>3</sub> )
Excess HNO <sub>2</sub> , 40°C	92.4 (0.49 <u>M</u> HNO <sub>3</sub> )	50.2 (1.1 <u>M</u> HNO <sub>3</sub> )
Excess HNO <sub>2</sub> , 50°C	90.9 (0.49 <u>M</u> HNO <sub>3</sub> )	27.5 (0.94 <u>M</u> HNO <sub>3</sub> )

<sup>a</sup>Less than 0.5 M HNO<sub>3</sub>.

<sup>b</sup>Greater than 0.5 M HNO<sub>3</sub>.

Table 5. Effect of strip bank temperature on the yield of stripped plutonium

Conditions of comparison	Stripped plutonium yield (%)	
	At 40°C	At 50°C
Excess HAN, 0.28-0.30 <u>M</u> HNO <sub>3</sub>	99.6	99.8
Excess HAN, 0.74-0.78 <u>M</u> HNO <sub>3</sub>	98.8	99.6
Excess HNO <sub>2</sub> , 0.49 <u>M</u> HNO <sub>3</sub>	92.4	90.9
Excess HNO <sub>2</sub> , 0.94-1.1 <u>M</u> HNO <sub>3</sub>	50.2	27.5

Table 6. Effect of changing from excess HAN to excess HNO<sub>2</sub> on yield of stripped plutonium

Conditions of comparison	Stripped plutonium yield (%)	
	Excess HAN	Excess HNO <sub>2</sub>
40°C, Low acidity (0.28-0.49 <u>M</u> HNO <sub>3</sub> )	99.6	92.4
50°C, Low acidity (0.30-0.49 <u>M</u> HNO <sub>3</sub> )	99.8	90.9
40°C, High acidity (0.78-1.1 <u>M</u> HNO <sub>3</sub> )	98.8	50.2
50°C, High acidity (0.74-0.94 <u>M</u> HNO <sub>3</sub> )	99.6	27.5

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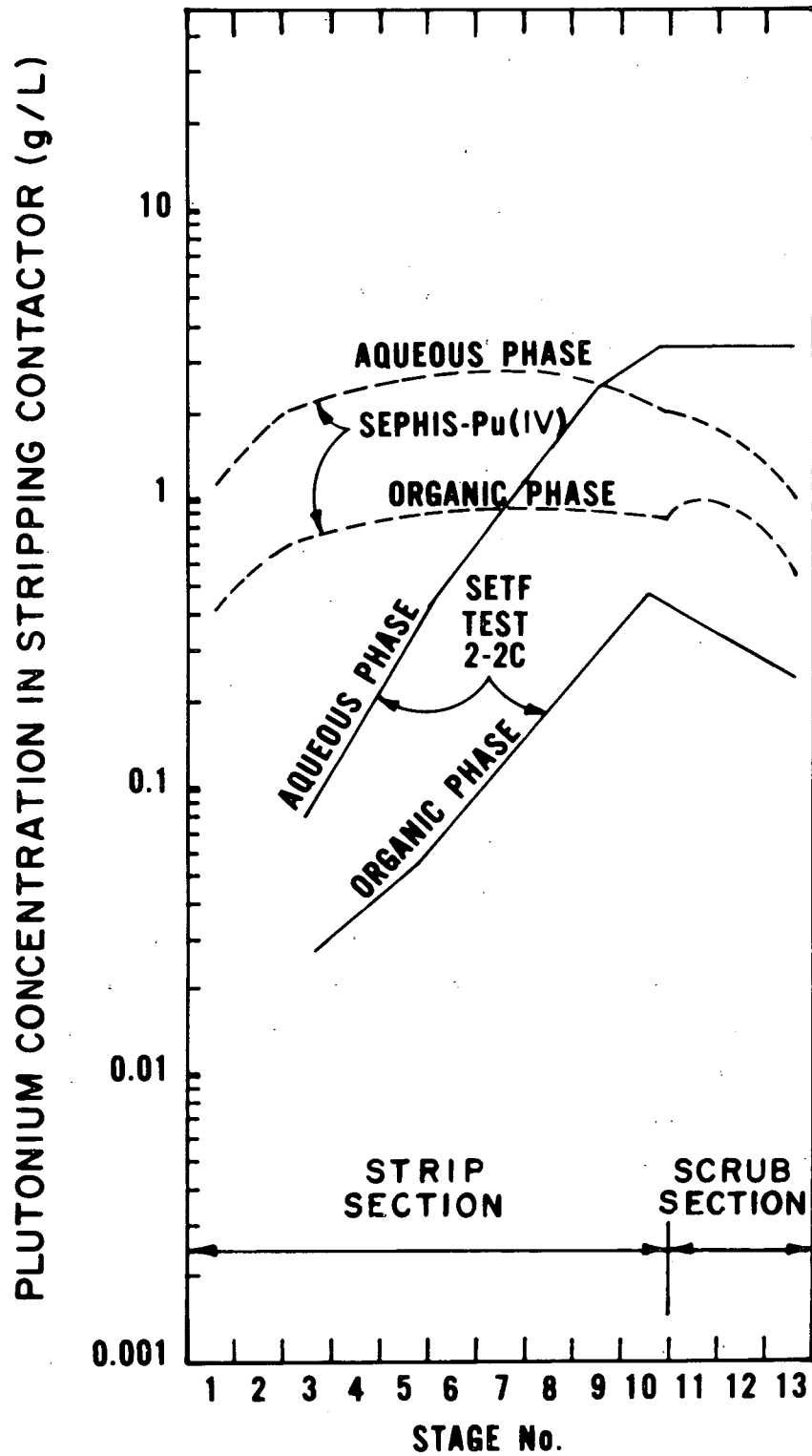


Fig. 12. Comparison of plutonium stripping effectiveness obtained in test 2-2C with SEPHIS calculation of Pu(IV).

Moreover, analysis of a sample of the aqueous stream leaving the contactor (BP) during test 2-2C showed that only 62% of the plutonium was in the tetravalent state. These results imply that a partial reduction of the plutonium had been effected, apparently by means of the nitrous acid.

A plutonium stripping yield of >99% was achieved during test 2-2D while using 13 stripping stages; in addition, a significant uranium-plutonium separation (separation factor of ~1400) was obtained. The stripping yield, equivalent to ~30% per stage, was not as high as that obtained when HAN was used (~65% per stage). Even so, a uranium-plutonium separation process in which nitrous acid is used to effect partial reduction of the plutonium appears to be feasible.

### 4.3 Uranium-Plutonium Stripping Operations

Both costripping and partial partitioning tests were made in the SETF. Costripping operations are those operations in which all of the uranium and plutonium are stripped from the pregnant organic solvent in one contactor, whereas only part of the uranium is stripped with the plutonium in partial partitioning operations.

#### 4.3.1 Costripping

During the costripping tests, plutonium was readily stripped, with or without reduction of the tetravalent plutonium, as shown in Fig. 13. The curve labeled ~Pu(IV) is predominantly IV since HAN and  $N_2H_4$  concentrations were very low. The relatively high flow ratio of the aqueous strip solution to the organic solvent, which is needed for adequate uranium stripping, enabled the Pu(IV) to be stripped with nearly the same effectiveness as observed for Pu(III).

The uranium concentration profiles in the stripping contactor during the costripping tests are shown in Fig. 14. A concentration profile obtained during a uranium stripping test in which the strip solution was 0.01 M  $HNO_3$  is included for comparison. It is readily apparent that the added salting used in the costripping tests, either to provide reductants for the plutonium or to adjust the acidity to a higher level to prevent plutonium hydrolysis, made the uranium stripping less effective.

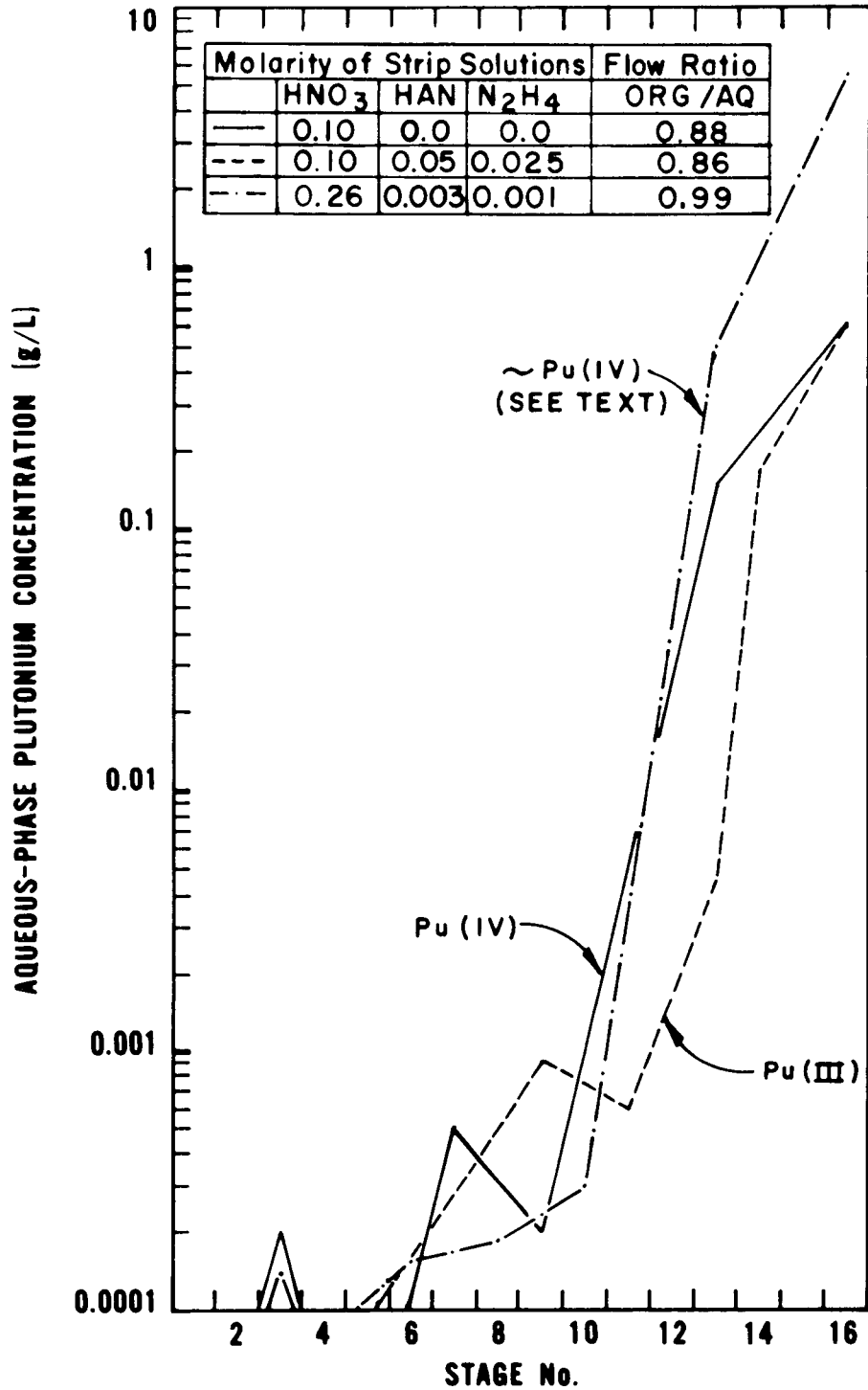


Fig. 13. Plutonium stripping during the costripping tests.



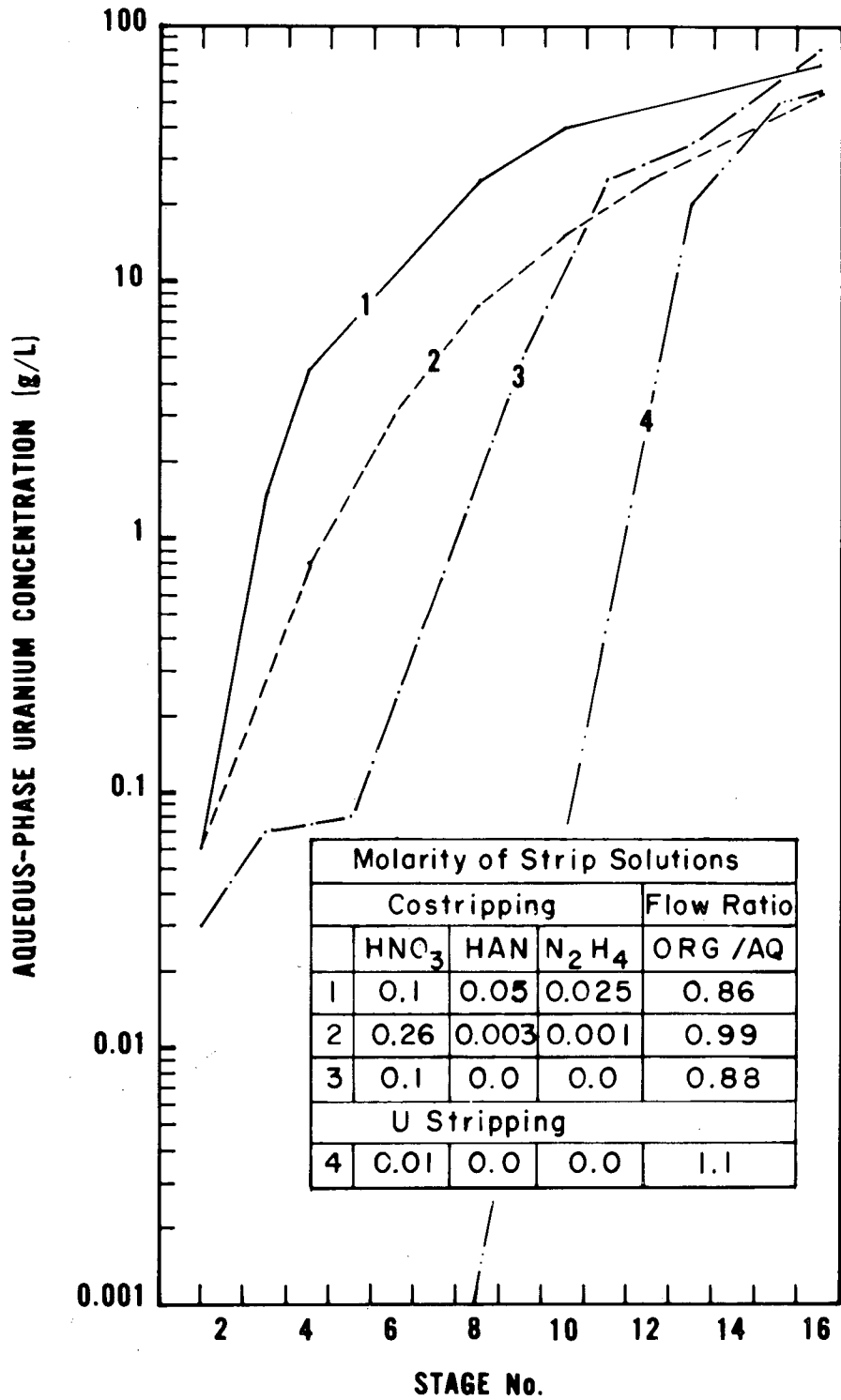


Fig. 14. Uranium stripping during the costripping tests.

Therefore, more stripping stages or a higher flow ratio of aqueous strip to organic solvent must be used to ensure sufficient uranium stripping yields. The latter remedy causes a more dilute product stream to be obtained; thus, intercycle evaporation of the product stream could be required.

#### 4.3.2 Partial partitioning

In partial partitioning operations, the goal is to obtain a plutonium-uranium product having a plutonium/heavy-metal (uranium plus plutonium) ratio of  $\sim 0.25$  since this is the ratio needed for subsequent use of the material as fast breeder core fuel. Because it is not usually possible to operate the partial partitioning system to achieve the exact plutonium/heavy-metal ratio desired, a ratio somewhat higher than 0.25 is sought. Uranium can then be added to dilute the product solution to the desired plutonium/heavy-metal ratio prior to conversion of the material to mixed oxide. During the partial partitioning tests, a goal was arbitrarily set to obtain a product solution having a plutonium/heavy-metal ratio of 0.25 to 0.40. This range was judged to be achievable when processing irradiated LWR fuels in which the plutonium/heavy-metal ratio is  $\sim 0.01$  and from which a relatively large enrichment (a factor of 25 to 40) is required. A narrower range might be specified when processing fast breeder fuels (mixed core and blanket) in which the initial plutonium/heavy-metal ratio is  $\sim 0.1$ .

The portion of the uranium stripped along with the plutonium can be regulated by (1) using inextractable nitrate salting in the aqueous phase; (2) using an organic backscrub stream in the partitioning contactor; or (3) reducing plutonium to the trivalent state in an aqueous solution containing the plutonium and uranium, followed by selective extraction of part of the uranium. All of these methods were tested in the SETF.

A process in which inextractable nitrate salting is used and the salt is provided by using a concentration of HAN greatly in excess of that required to reduce the plutonium to the trivalent state was developed at SRL.<sup>15</sup> This process was demonstrated in SETF tests 1-1A and 1-1B, as well as in most of the scavenging bank (C bank) operations during Campaign 2. Product solutions having the desired plutonium/heavy-metal ratio can be obtained if a relatively small flow ratio ( $\leq 0.1$ ) of aqueous strip to organic solvent is used. The large excess of HAN remaining in the product

solution must be decomposed under carefully controlled conditions (to avoid the autocatalytic reaction of HAN with  $\text{HNO}_3$ ) before the product solution can be heated.

Results of the SETF tests in which the organic backscrub and selective uranium extraction methods were used to regulate the portion of uranium stripped with the plutonium are summarized in Table 7. These results show that almost any desired enrichment of the plutonium/heavy-metal ratio can be obtained by varying the flow rates. Also, the results indicate that, because of the extreme sensitivity to the flow rates, the desired plutonium/heavy-metal ratio probably could be best maintained by using an in-line monitor to measure the uranium concentration in the product solution. The successful use of an in-line monitor has been demonstrated by Hannaford and Davis,<sup>16</sup> in a recent miniature-mixer-settler experiment at ORNL.

Table 7. Partial partitioning results

Organic/aqueous (O/A) flow ratio	Pu/(U+Pu) ratio		Concentration factor
	Feed	Product	
0.4 <sup>a</sup>	0.9	8.8	9.8
0.9 <sup>a</sup>	0.8	17.4	21.0
1.5 <sup>a</sup>	0.9	62.7	69.7
0.3 <sup>b</sup>	9.7	21.4	2.2
1.2 <sup>a</sup>	8.8	96.9	11.0

<sup>a</sup>O/A flow ratio in backscrub section of uranium-plutonium strip column.

<sup>b</sup>O/A flow ratio in extraction section of selective uranium extraction column.

#### 4.4 Solvent Degradation

A sample of the waste organic solvent from SETF test 1-3 was analyzed for the presence of the TBP decomposition products, dibutyl phosphate (DBP) and monobutyl phosphate (MBP), by means of a standard plutonium retention test. In the standard test, the organic solvent is used to extract plutonium from a stock solution and the plutonium is then stripped exhaustively

with 0.1 M  $\text{HNO}_3$ . The concentration of plutonium retained in the organic solvent is used to indicate the concentrations of DBP and MPB present, since plutonium complexed by these compounds cannot be stripped by means of the 0.1 M  $\text{HNO}_3$ .

When the waste solvent sample was subjected to the retention test, the retained plutonium concentration was  $6.6 \times 10^{-4}$  M. However, during the costripping operation in SETF test 1-3, the concentration of plutonium retained in the solvent was only  $\sim 4 \times 10^{-6}$  M. This is similar to the concentration of plutonium that was retained when fresh, carbonate-washed solvent was subjected to the standard plutonium retention test. Significant amounts of DBP and MPB were apparently present in the waste solvent but were preferentially complexed by uranium during the costripping operation, thus allowing the plutonium to be stripped.

## 5. SUMMARY AND CONCLUSIONS

Two campaigns, each consisting of a series of related solvent extraction experiments, were made in the SETF to test a variety of coprocessing flowsheets, primarily for LWR fuels. The irradiated  $\text{UO}_2$  fuel used in these campaigns had been irradiated in the H. B. Robinson-2 FWR to a peak burnup of  $\sim 2.7$  TJ/kg ( $\sim 31,000$  MWd/metric ton) and had been discharged in 1974. The  $\sim 5$ -year cooling time precluded obtaining data regarding the behavior of the short-lived fission products, particularly  $^{95}\text{Zr}$  and  $^{95}\text{Nb}$ .

Areas of the solvent extraction process that received special attention during these campaigns included (1) crud formation, particularly in the extraction contactor; (2) uranium and plutonium losses in the extraction raffinate; (3) fission product decontamination; (4) reduction of tetravalent plutonium by means of HAN or nitrous acid; (5) costripping of uranium and plutonium; (6) partial partitioning; and (7) solvent degradation.

A black interfacial crud formed in the settlers of the extraction bank during all first-cycle tests. This crud appeared to be an emulsion, stabilized by minute particles of insoluble fission product metals that were not removed during the feed clarification. Several methods were tried to prevent (or at least reduce) the formation, but none was entirely successful.

Some improvement was obtained by digesting the clarified feed solution with hydrogen peroxide at 90°C; this treatment was intended to dissolve any metal colloids remaining in the clarified feed solution.

Losses of uranium and plutonium in the extraction raffinate were within the range of 0.0002 to 0.07%, which is considered sufficiently low for reprocessing purposes. The most serious potential for large losses occurs when operating with  $\geq 75\%$  solvent saturation. Under these conditions, continuous monitoring of the uranium or plutonium concentrations within the extraction contactor would be needed.<sup>16</sup>

Fission product DFs were measured at a variety of operating conditions to determine the general range of values obtained while processing LWR fuel. In general, cesium and cerium DFs ranged from  $10^4$  to  $10^6$ , whereas ruthenium DFs ranged from  $10^3$  to  $10^5$ . While all of the cesium and cerium decontamination occurred in the extraction/scrub contactor, a significant part of the ruthenium decontamination was effected in the strip contactor.

Important objectives of these SETF tests were to establish the necessary operating conditions for the effective use of HAN for reducing tetravalent plutonium and to determine whether stabilization with hydrazine is necessary. The tests and associated studies showed that unstabilized HAN can be used effectively if the aqueous-phase acidity is maintained below  $\sim 1 \text{ M HNO}_3$  (preferably below  $\sim 0.5 \text{ M HNO}_3$ ) and a sufficient excess of HAN is used to react with any nitrous acid that may be present in the system. The use of hydrazine to scavenge nitrous acid is not necessary; thus, the potential problems that accompany the use of hydrazine can be avoided.

Reductive stripping of plutonium by means of nitrous acid appears to be feasible, although not as effective as with HAN. The results showed that suitable yields of stripped plutonium can be obtained if the required degree of uranium-plutonium separation is not extreme.

Several uranium-plutonium costripping tests were performed. The added salting used in the strip solution, either to provide reductants for the plutonium or to obtain a higher acidity (to prevent plutonium hydrolysis), made the uranium stripping less effective than conventional stripping of uranium with  $0.01 \text{ M HNO}_3$  (when only uranium is present). In order to obtain a sufficient uranium stripping yield in costripping operations,

either a relatively large number of strip stages or a relatively high flow ratio of aqueous strip to organic solvent is required. Plutonium stripping occurred readily at the relatively high flow ratios required for uranium stripping, with or without reduction of the tetravalent plutonium.

In partial partitioning operations, the portion of the uranium stripped along with the plutonium can be regulated by (1) using inextractable nitrate salting in the aqueous phase; (2) using an organic backscrub stream in the partitioning contactor; or (3) reducing plutonium to the trivalent state in an aqueous solution containing the plutonium and uranium, followed by selective extraction of part of the uranium. All of these methods were tested in the SETF, but the major emphasis was placed on the use of an organic backscrub. Results showed that almost any desired enrichment of the plutonium/heavy-metal ratio can be obtained by varying the flow rates. Also, because of the extreme sensitivity to flow rates, the desired plutonium/heavy-metal ratio probably could be best obtained by using an in-line monitor to measure the uranium concentration in the product solution.

Plutonium retention tests made on a sample of waste organic solvent from one of the SETF costripping operations indicated that significant amounts of the TBP degradation products were present but did not retard the stripping of plutonium. The degradation products may have been preferentially complexed by the uranium.

## 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 staff, primarily those groups under the supervision of J. H. Cooper and J. M. Peele. Engineering and maintenance services were provided by personnel from UCC-ND 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 1 first-cycle tests — extraction/scrub bank conditions and results

	Run No.		
	1-1	1-2	1-3
Dates	11/19-22/78	11/28-29/78	12/5-7/78
Bank temperature, °C	38-39	39-40	39
Number of stages: scrub/int. scrub/extraction	7/0/9	7/0/9	7/0/9
AX flow rate, L/h	1.573	1.563	1.556
Flow ratios:			
AS/AX	0.108	0.109	0.122
AF/AX	0.261	0.256	0.311
<u>Inlet stream compositions<sup>a</sup></u>			
AS stream:			
HNO <sub>3</sub> , M	1.0	0.50	3.0
AX stream:			
% TBP	30	30	30
HNO <sub>3</sub> , M	~0.45	~0.45	~0.45
AF stream:			
HNO <sub>3</sub> , M	3.0	3.1	3.0
U, g/L	309	301	297
Pu, g/L	3.11	2.73	2.79
Am, mg/L	111	96	123
Cm, mg/L	7	7	7
Ru-106, Ci/L	4.9	5.8	4.8
Cs-137, Ci/L	21.8	23.7	23.6
Ce-144, Ci/L	3.9	4.5	4.8
Eu-154, Ci/L	0.73	0.82	0.99
Np-239, Ci/L	0.0056 <sup>b</sup>	0.0048 <sup>b</sup>	0.0062 <sup>b</sup>

Table A-1 (continued)

	Run No.		
	1-1	1-2	1-3
<u>Outlet stream compositions<sup>a</sup></u>			
AW stream:			
HNO <sub>3</sub> , M	3.5	~4	~4
U, mg/L	<0.1	2	8.5
Pu, mg/L	0.0052	0.0062	<0.05
Am, mg/L	77	75	c
Cm, mg/L	4.5	4.3	c
AP stream:			
HNO <sub>3</sub> , M	0.04	0.03	0.18
HNO <sub>2</sub> , M	0.0008	c	<0.0002
U, g/L	87	76	88
Pu, g/L	0.73	0.51	0.70
Ru-106, µCi/L	1030	1550	766
Sb-125, µCi/L	<2	<5	<2
Cs-134, µCi/L	<2	<1	<0.4
Cs-137, µCi/L	<2	<2	<0.5
Ce-144, µCi/L	<10	<70	<30
Eu-154, µCi/L	<5	<3	<1
Np-239, µCi/L	1200	c	c

<sup>a</sup>On date of run.

<sup>b</sup>Calculated from <sup>243</sup>Am concentration (~25% of total americium) in AF.

<sup>c</sup>Not measured.

Table A-2. Run 1-1 first-cycle tests — strip bank conditions and results

	Run No.			
	1-1A	1-1B	1-1C	1-1D
Dates	11/19-20/78	11/20/78	11/21/78	11/21-22/78
Bank temperature, °C	39	39	38-39	39
Number of stages: strip/scrub	4/0	4/0	4/0	4/0
BX flow rate, L/h	0.25	0.26	0.27	0.26
Flow ratios: AP/BX	5.9	5.8	5.7	6.0
<u>Inlet stream compositions</u>				
BX stream:				
HNO <sub>3</sub> , <u>M</u>	0.06	0.11	0.19	0.22
HAN, <u>M</u>	1.42	0.49	0.03 <sup>a</sup>	b
AP stream:				
% TBP	30	30	30	30
HNO <sub>3</sub> , <u>M</u>	0.04	0.04	0.045	0.05
HNO <sub>2</sub> , <u>M</u>	0.0004	0.0007	0.0009	0.002
U, g/L	91	c	87	87
Pu, g/L	0.75	0.73	0.71	0.73
Ru-106, μCi/L	92	850	1210	1250
Sb-125, μCi/L	<2	<20	<6	<2
Cs-134, μCi/L	<0.3	<3	<4	<2
Cs-137, μCi/L	<0.4	<3	<6	<2
Ce-144, Ci/L	<2	<200	<70	<10
Eu-154, Ci/L	<2	<6	<6	<5
Np-239, Ci/L	c	1000	1200	1400

Table A-2 (continued)

	Run No.			
	1-1A	1-1B	1-1C	1-1D
<u>Outlet stream compositions<sup>d</sup></u>				
BP stream:				
HNO <sub>3</sub> , M	0.31	0.40	0.40	0.40
U, g/L	49	44	28	25
Pu, g/L	4.0	4.4	2.9	4.0
Ru-106, $\mu$ Ci/L	60	612	1070	1200
Sb-125, $\mu$ Ci/L	<3	<6	<6	<8
Cs-134, $\mu$ Ci/L	29	20	41	31
Cs-137, $\mu$ Ci/L	94	71	128	103
Ce-144, $\mu$ Ci/L	9	<60	<80	<70
Eu-154, $\mu$ Ci/L	<5	<3	<3	<4
Np-239, $\mu$ Ci/L	c	9780	6900	14000
BU stream:				
HNO <sub>3</sub> , M	0.01	<0.01	0.01	0.01
U, g/L	83	79	78	80
Pu, g/L	0.005	0.018	0.16	0.012

<sup>a</sup>Also contained 2 M NH<sub>4</sub>NO<sub>3</sub>.

<sup>b</sup>Reductant was 0.03 M ferrous sulfamate. Also contained 2 M NH<sub>4</sub>NO<sub>3</sub>.

<sup>c</sup>Not measured.

<sup>d</sup>On date of run.

Table A-3. Run 1-1 first-cycle tests -- plutonium scavenging bank conditions and results

	Run No.			
	1-1A	1-1B	1-1C	1-1D
Dates	11/19-20/78	11/20/78	11/21/78	11/21-22/78
Bank temperature, °C	38-39	39	38-39	39
Number of stages: strip/scrub	4/0	4/0	4/0	4/0
CX flow rate, L/h	0.23	0.22	0.26	0.27
Flow ratio: BU/CX	6.4	6.9	6.0	5.8
<u>Inlet stream compositions</u>				
CX stream:				
HNO <sub>3</sub> , <u>M</u>	1.9	1.9	1.9	1.9
Ferrous sulfamate, <u>M</u>	0.05	0.05	0.05	0.05
BU stream:				
% TBP	30	30	30	30
HNO <sub>3</sub> , <u>M</u>	0.01	<0.01	0.01	0.01
U, g/L	85	79	91	78
Pu, mg/L	11	18	120	3.4

Table A-3 (continued)

	Run No.			
	1-1A	1-1B	1-1C	1-1D
<u>Outlet stream compositions</u>				
CP stream:				
HNO <sub>3</sub> , M	1.05	1.33	1.35	1.32
U, g/L	41	33	32	36
Pu, mg/L	27	99	840	66
CW stream:				
HNO <sub>3</sub> , M	a	a	0.11	a
U, g/L	76	74	73	74
Pu, mg/L	1.1	3.2	17	0.52

<sup>a</sup>Not measured.

Table A-4. Runs 1-2 and 1-3 first-cycle tests -- strip bank conditions and results

	Run No.			
	1-2A	1-2B	1-3A	1-3B
Dates	11/28-29/78	11/29/78	12/5-6/78	12/6-7/78
Bank temperature, °C	39	39	39	39
Number of stages: strip/scrub	5/3	5/3	16/0	16/0
BX flow rate, L/h	0.23	0.24	1.76	1.82
Flow ratios:				
AP/BX	7.0	6.5	0.88	0.86
BS/BX	1.5	0.41	--	--
<u>Inlet stream composition<sup>a</sup></u>				
BX stream:				
HNO <sub>3</sub> , M	0.20	0.18	0.098	0.10
HAN, M	0.1 <sup>b</sup>	0.1 <sup>b</sup>	--	0.05 <sup>c</sup>
BS stream:				
% TBP	30	30	--	--
AP stream:				
% TBP	30	30	30	30
HNO <sub>3</sub> , M	0.04	0.03	0.15	0.18
HNO <sub>2</sub> , M	0.0002	d	<0.0002	<0.0002
U, g/L	77	76	90	89
Pu, g/L	0.59	0.51	0.76	0.70
Ru-106, µCi/L	1708	1390	833	700
Sb-125, µCi/L	<7	<5	<5	<2
Cs-134, µCi/L	<2	<1	<1	<0.4
Cs-137, µCi/L	<2	<2	<2	<0.5
Ce-144, µCi/L	70	<70	<50	<30
Eu-154, µCi/L	<2	<3	<3	<1

Table A-4 (continued)

	Run No.			
	1-2A	1-2B	1-3A	1-3B
<u>Outlet stream compositions<sup>a</sup></u>				
BP stream:				
HNO <sub>3</sub> , <u>M</u>	0.75	0.57	0.20	0.24
U, g/L	2.3	46	88	76
Pu, g/L	3.8	4.4	0.79	0.68
Ru-106, <u>μCi/L</u>	1620	120	1040	107
Sb-125, <u>μCi/L</u>	13	<20	<4	<4
Cs-134, <u>μCi/L</u>	58	284	<0.7	19
Cs-137, <u>μCi/L</u>	190	916	<0.8	62
Ce-144, <u>μCi/L</u>	<80	150	<50	<50
Eu-154, <u>μCi/L</u>	<3	36	<2	<3
Np-239, <u>μCi/L</u>	66000	d	d	d
BU stream:				
HNO <sub>3</sub> , <u>M</u>	0.01	<0.01	0.08	<0.02
U, g/L	65	67	0.05	0.24
Pu, g/L	0.0041	0.0042	0.00003	0.0004
Ru-106, <u>μCi/L</u>	d	d	d	577
Sb-125, <u>μCi/L</u>	d	d	d	<3
Cs-134, <u>μCi/L</u>	d	d	d	<0.5
Cs-137, <u>μCi/L</u>	d	d	d	<0.5
Ce-144, <u>μCi/L</u>	d	d	d	<40
Eu-154, <u>μCi/L</u>	d	d	d	<1

<sup>a</sup>On date of run.<sup>b</sup>Also contained 0.05 M N<sub>2</sub>H<sub>4</sub>.<sup>c</sup>Also contained 0.02 M N<sub>2</sub>H<sub>4</sub>.<sup>d</sup>Not measured.



Table A-5. Runs 1-2 and 1-3 first-cycle tests - plutonium scavenging bank conditions and results

	Run No.			
	1-2A	1-2B	1-3A	1-3B
Dates	11/28-29/78	11/29/78	12/5-6/78	12/6-7/78
Bank temperature, °C	38-39	39	39	39
Number of stages: strip/scrub	4/0	4/0	16/0	16/0
CX flow rate, L/h	0.34	0.28	0.22	0.19
Flow ratio: BU/CX	5.8	5.9	7.0	8.2
<u>Inlet stream compositions</u>				
CX stream:				
HNO <sub>3</sub> , <u>M</u>	2.0	2.0	2.1	2.1
Ferrous sulfamate, <u>M</u>	0.05	0.05	0.05	0.05
BU stream:				
% TBP	30	30	30	30
HNO <sub>3</sub> , <u>M</u>	0.01	<0.01	0.08	<0.01
U, g/L	60	67	0.05	0.24
Pu, mg/L	2.3	2.1	0.017	0.44

Table A-5 (continued)

	Run No.			
	1-2A	1-2B	1-3A	1-3B
<u>Outlet stream compositions</u>				
CP stream:				
HNO <sub>3</sub> , M	0.78	1.11	a	a
U, g/L	26	26	<0.05	0.42
Pu, mg/L	19	22	0.49	0.003
CW stream:				
HNO <sub>3</sub> , M	a	0.16	a	a
U, g/L	61	63	0.04	0.16
Pu, mg/L	1.1	0.37	0.020	0.005

<sup>a</sup>Not measured.

Table A-6. Runs 2-2 and 2-4 first-cycle tests — extraction/scrub bank conditions and results

	Run No.					
	2-2A	2-2B	2-2C	2-2D	2-2E	2-4A
Dates	5/7-8/79	5/9-10/79	5/11/79	5/13/79	5/15-16/79	6/18-19/79
Bank temperature, °C	40	40	40	50-51	40	39
Number of stages: scrub/int. scrub/extrac.	3/3/10	3/3/10	3/3/10	3/3/10	3/3/10	3/3/10
AX flow rate, L/h	1.651	1.628	1.648	1.650	1.678	1.516
Flow ratios:						
AS/AX	0.119	0.116	0.116	0.116	0.066	0.133
AIS/AX	0.065	0.066	0.067	0.056	0.033	0.075
AF/AX	0.297	0.273	0.251	0.267	0.139	0.518
<u>Inlet stream compositions<sup>a</sup></u>						
AS stream:						
HNO <sub>3</sub> , M	0.79	0.79	0.80	0.80	0.80	0.80
AIX stream:						
HNO <sub>3</sub> , M	2.95	2.95	3.05	3.05	3.08	3.05
AX stream:						
% TBP	30	30 <sup>b</sup>	30 <sup>c</sup>	30	15	30
AF stream:						
HNO <sub>3</sub> , M	3.2	3.3	3.1	3.1	3.2	3.3
U, g/L	298	284	299	313	318	144
Pu, g/L	2.71	2.80	2.63	2.63	2.91	13.5
Am, mg/L	99	114	117	103	103	22
Cm, mg/L	5.5	6.8	6.3	6.6	6.1	2.2
Ru-106, Ci/L	4.2	3.5	3.0	3.2	2.8	1.0
Sb-125, Ci/L	0.54	0.53	0.51	0.54	0.55	0.24
Cs-134, Ci/L	6.7	7.3	6.8	7.2	6.5	2.5
Cs-137, Ci/L	25.1	26.0	25.1	26.1	24.0	9.9
Ce-144, Ci/L	3.3	3.2	3.2	3.3	3.0	1.2
Eu-154, Ci/L	0.97	1.1	1.0	1.1	0.95	0.39
Np-239, Ci/L	0.0037 <sup>d</sup>	0.0043 <sup>d</sup>	0.0044 <sup>d</sup>	0.0039 <sup>d</sup>	0.0037 <sup>d</sup>	0.0008 <sup>d</sup>

Table A-6 (continued)

	Run No.					
	2-2A	2-2B	2-2C	2-2D	2-2E	2-4A
<u>Outlet stream compositions<sup>a</sup></u>						
AW stream:						
HNO <sub>3</sub> , M	2.6	2.7	3.0	2.1	e	2.9
U, mg/L	e	37	60	e	56	<1
Pu, mg/L	0.22	0.10	1.0	0.37	0.21	0.059
Am, mg/L	61	65	67	0.99	28	36
Cm, mg/L	3.0	4.2	3.7	0.06	1.6	1.5
Ru-106, Ci/L	1.4	1.9	1.5	1.7	e	0.70
Sb-125, Ci/L	0.27	0.35	0.27	0.38	e	e
Cs-134, Ci/L	3.7	4.2	4.0	4.4	e	1.8
Cs-137, Ci/L	13.7	15.4	15.0	16.2	e	7.2
Ce-144, Ci/L	1.9	2.2	1.9	2.0	e	0.90
Eu-154, Ci/L	0.51	0.65	0.65	0.76	e	0.24
AP stream:						
HNO <sub>3</sub> , M	0.03	0.07	0.06	0.025	0.008	0.12
HNO <sub>2</sub> , M	<0.0002	0.013	0.023	0.0009	0.0016	<0.001
U, g/L	90	72	81	91	42	83
Pu, g/L	0.77	0.69	0.68	0.78	0.32	9.51
Ru-106, μCi/L	149	154	268	60.3	167	220
Sb-125, μCi/L	<2	e	4.6	e	1.0	e
Cs-134, μCi/L	<0.5	e	e	e	<0.08	e
Cs-137, μCi/L	0.93	e	e	0.51	<0.6	1.1
Ce-144, μCi/L	<9	2.4	e	e	<0.8	e
Eu-154, μCi/L	<2	<0.8	e	e	<0.2	e
Np-239, μCi/L	3	e	e	e	380	910

<sup>a</sup>On date of run.

<sup>b</sup>Also contained 0.27 M HNO<sub>2</sub>.

<sup>c</sup>Also contained 0.33 M HNO<sub>2</sub>.

<sup>d</sup>Calculated from <sup>243</sup>Am concentration (~25% of total americium) in AF.

<sup>e</sup>Not measured.

Table A-7. Runs 2-2 and 2-4 first-cycle tests — strip bank conditions and results

	Run No.					
	2-2A	2-2B	2-2C	2-2D	2-2E	2-4A
Dates	5/7-8/79	5/9-10/79	5/11/79	5/13/79	5/15-16/79	6/18-19/79
Bank temperature, °C	28	28	28	28	27-28	51
Number of stages: strip/scrub	5/3	10/3	10/3	13/3	13/3	16 <sup>a</sup> /0
BX flow rate, L/h	0.31	0.30	0.40	0.40	0.40	1.69
Flow ratios:						
AP/BX	5.4	5.5	4.1	4.1	4.2	0.90
BS/BX	0.93	1.0	1.2	1.1	1.2	--
BIX/BX	--	--	--	--	--	0.093
<u>Inlet stream compositions<sup>b</sup></u>						
BS stream:						
HNO <sub>3</sub> , $\bar{M}$	0.10	0.10	0.10	0.10	0.10	0.006 <sup>c</sup>
BIX stream:						
HNO <sub>3</sub> , $\bar{M}$	--	--	--	--	--	3.0
BS stream:						
% TBP	30	30	30	30	15	--
HNO <sub>2</sub> , $\bar{M}$	0.30	0.27	0.33	0.33	0.16	--
HNO <sub>3</sub> , $\bar{M}$	0.31	0.33	d	d	0.1	--
AP stream:						
% TBP	30	30	30	30	5	30
HNO <sub>3</sub> , $\bar{M}$	0.03	0.07	0.06	0.025	0.008	0.12
HNO <sub>2</sub> , $\bar{M}$	<0.0002	0.013	0.023	0.0009	0.0016	<0.001
U, g/L	90	72	81	91	42	83
Pu, g/L	0.77	0.69	0.68	0.79	0.32	9.6
Ru-106, $\mu\text{Ci/L}$	149	154	268	60	167	220
Sb-125, $\mu\text{Ci/L}$	<2	d	d	d	d	d
Cs-134, $\mu\text{Ci/L}$	<0.5	d	d	d	d	d
Cs-137, $\mu\text{Ci/L}$	0.93	d	d	0.51	0.6	1.1
Ce-144, $\mu\text{Ci/L}$	<9	2.4	d	d	<0.8	d
Eu-154, $\mu\text{Ci/L}$	<2	<0.8	d	d	<0.2	d
Np-239, $\mu\text{Ci/L}$	d	d	d	d	380	910

Table A-7 (continued)

	Run No.					
	2-2A	2-2B	2-2C	2-2D	2-2E	2-4A
<u>Outlet stream compositions<sup>b</sup></u>						
BP stream:						
HNO <sub>3</sub> , <u>M</u>	0.37	0.43	0.34	0.34	0.26	0.38
U, g/L	47	36	34	34	72	62
Pu, g/L	2.4	3.7	2.9	3.2	1.3	6.1
Ru-106, $\mu$ Ci/L	26	24	13.5	16	35	120
Sb-125, $\mu$ Ci/L	<3	<5	2.7	d	0.76	d
Cs-134, $\mu$ Ci/L	12	28	13	9.7	3.7	5.9
Cs-137, $\mu$ Ci/L	45	108	57	35	13	21.1
Ce-144, $\mu$ Ci/L	19	19	11	11	<1	2.2
Eu-154, $\mu$ Ci/L	<1	5.4	d	d	0.9	d
Np-239, $\mu$ Ci/L	160	<140	d	d	2600	910
BU stream:						
HNO <sub>3</sub> , <u>M</u>	0.02	0.05	0.055	0.027	d	d
HNO <sub>2</sub> , <u>M</u>	0.02	0.023	0.089	0.0035	0.014	<0.0002
U, g/L	65	58	56	~70	22	0.001
Pu, g/L	0.21	0.020	0.008	0.0047	0.0002	0.0016
Ru-106, $\mu$ Ci/L	112	120	173	60	d	88
Sb-125, $\mu$ Ci/L	<2	d	d	d	d	d
Cs-134, $\mu$ Ci/L	<0.1	d	d	d	d	d
Cs-137, $\mu$ Ci/L	0.2	d	d	d	d	0.81
Ce-144, $\mu$ Ci/L	<1	1.9	1.6	1.2	d	<0.5
Eu-154, $\mu$ Ci/L	<0.5	d	d	d	d	d
Np-239, $\mu$ Ci/L	<20	d	d	d	d	d

<sup>a</sup>Comprised of 2 low-acid strip stages and 14 high-acid stages.

<sup>b</sup>On date of run.

<sup>c</sup>Also contained 0.0035 M HAN.

<sup>d</sup>Not measured.

Table A-8. Run 2-2 first-cycle tests — plutonium scavenging bank conditions and results

	Run No.				
	2-2A	2-2B	2-2C	2-2D	2-2E
Dates	5/7-8/79	5/9-10/79	5/11/79	5/13/79	5/15-16/79
Bank temperature, °C	40	40	40	38-39	39-40
Number of stages: strip/scrub	16/0	16/0	16/0	16/0	16/0
CX flow rate, L/h	0.20	0.22	0.22	0.20	0.21
Flow ratio: BU/CX	9.7	8.8	9.6	10.6	10.3
<u>Inlet stream compositions</u>					
CX stream:					
HNO <sub>3</sub> , M	0.21	0.21	0.21	0.21	0.21
HAN, M	~1.5	~1.5	~1.5	~1.5	~1.5
BU stream:					
% TBP	30	30	30	30	15
HNO <sub>3</sub> , M	0.021	0.05	0.05	0.027	a
HNO <sub>2</sub> , M	0.022	0.023	0.089	0.0035	0.013
U, g/L	65	58	56	a	22
Pu, mg/L	210	20	8	4.7	0.23
Ru-106, µCi/L	112	120	173	60	a
Cs-134, µCi/L	<0.1	a	a	a	a
Cs-137, µCi/L	0.19	a	a	a	a
Ce-144, µCi/L	<1	1.9	1.6	1.2	a
Eu-154, µCi/L	<0.5	a	a	a	a

Table A-8 (continued)

	Run No.				
	2-2A	2-2B	2-2C	2-2D	2-2E
<u>Outlet stream compositions</u>					
CP stream:					
HNO <sub>3</sub> , <u>M</u>	0.7	0.63	0.68	0.48	0.23
Reducing, <u>N</u>	3.2	2.8	3.2	2.8	4.0
U, g/L	19	15	15	a	9.8
Pu, mg/L	2000	220	73	54	1.2
Ru-106, <u>μCi/L</u>	92	27	44	24	52
Cs-134, <u>μCi/L</u>	<0.9	<1	0.13	a	a
Cs-137, <u>μCi/L</u>	3.5	1.4	0.22	0.81	0.54
Ce-144, <u>μCi/L</u>	5.1	3.5	<1	3.0	a
Eu-154, <u>μCi/L</u>	1.9	a	a	a	a
CW stream:					
HNO <sub>3</sub> , <u>M</u>	0.06	0.04	0.05	0.04	a
HNO <sub>2</sub> , <u>M</u>	<0.0002	0.0004	<0.0002	0.0003	0.0002
U, g/L	63	57	51	68	18
Pu, mg/L	2.7	0.27	1.5	0.35	<0.2
Ru-106, <u>μCi/L</u>	94	104	230	60	a
Cs-134, <u>μCi/L</u>	1.0	a	a	a	a
Cs-137, <u>μCi/L</u>	3.5	<0.5	a	a	a
Ce-144, <u>μCi/L</u>	5.1	a	2.4	1.2	a
Eu-154, <u>μCi/L</u>	0.8	a	a	a	a

<sup>a</sup>Not measured.



Table A-9. Run 2-1 second-cycle tests — extraction/scrub bank conditions and results

	Run No.								
	2-1A	2-1B	2-1C	2-1D	2-1E	2-1F	2-1G	2-1H	2-1J
Dates	3/26-27/79	3/27/79	3/27/79	3/27-28/79	3/28/79	3/29/79	3/29/79	3/30/79	3/30/79
Bank temperature, °C	40	40	39	41	40	41	42	40	40
Number of stages: scrub/int. scrub/extrac.	3/3/10	3/3/10	6/0/10	6/09/10	3/3/10	3/3/10	3/3/10	6/0/10	6/0/10
AX flow rate, L/h	1.12	1.03	0.96	1.00	1.01	1.07	1.07	1.04	1.07
Flow ratios:									
AS/AX	0.11	0.12	0.17	0.19	0.12	0.12	0.12	0.19	0.14
AIS/AX	0.059	0.064	--	--	0.077	0.067	0.068	--	--
AF/AX	0.94	1.02	1.17	1.11	1.10	0.95	0.97	0.98	0.97
<u>Inlet stream compositions</u>									
AS stream:									
HNO <sub>3</sub> , M	0.8	0.8	3.0	3.0	0.8	0.8	0.8	3.0	3.0
AIX stream:									
HNO <sub>3</sub> , M	3.0	3.0	--	--	3.0	3.0	3.0	--	--
AX stream:									
% TBP	30	30	30	30	30	30	30	30	30
HNO <sub>2</sub> , M	a	a	a	a	a	0.48	0.48	0.48	0.48
AF stream:									
HNO <sub>3</sub> , M	3.5	3.5	3.5	3.5	3.5	3.5	3.5	3.5	3.5
U, g/L	78.0	78.0	78.0	78.0	78.0	78.0	78.0	78.0	78.0
Pu, g/L	2.1	2.1	2.1	2.1	2.1	2.1	2.1	2.1	2.1
Ru-106, µCi/L	7700	7700	7700	7700	7700	7700	7700	7700	7700
Sb-125, µCi/L	105	105	105	105	105	105	105	105	105
Cs-134, µCi/L	78.4	78.4	78.4	78.4	78.4	78.4	78.4	78.4	78.4
Cs-137, µCi/L	289	289	289	289	289	289	289	289	289

Table A-9 (continued)

	Run No.								
	2-1A	2-1B	2-1C	2-1D	2-1E	2-1F	2-1G	2-1H	2-1J
<u>Outlet stream compositions<sup>b</sup></u>									
AP stream:									
HNO <sub>3</sub> , M	0.05	0.03	0.15	0.15	0.02	0.13	0.13	0.28	0.24
HNO <sub>2</sub> , M	0.0005	0.001	<0.002	0.0004	0.0003	0.043	0.035	0.045	0.040
U, g/L	69	84	92	87	87	73	73	74	80
Pu, g/L	1.5	1.2	0.90	0.65	0.68	1.9	1.8	1.9	1.5
Ru-106, μCi/L	48	16	4.6	8.7	10.3	24	17.5	a	38
Cs-134, μCi/L	<0.3	<0.5	<0.1	<0.1	<0.1	<0.2	<0.1	<0.8	<0.5
CS-137, μCi/L	<0.5	<0.5	<0.2	<0.2	<0.2	<0.2	<0.2	<0.8	<0.2

<sup>a</sup>Not measured.<sup>b</sup>AW stream not sampled.

Table A-10. Run 2-1 second-cycle tests -- strip bank conditions and results

	Run No.								
	2-1A	2-1B	2-1C	2-1D	2-1E	2-1F	2-1G	2-1H	2-1J
Dates	3/26-27/79	3/27/79	3/27/79	3/27-28/79	3/28/79	3/29/79	3/29/79	3/30/79	3/30/79
Bank temperature, °C	41	50	42	51	40	41	40	40	51
Number of stages: strip/scrub	5/3	5/3	5/3	5/3	5/3	5/3	5/3	5/3	5/3
BX flow rate, L/h	0.23	0.23	0.23	0.23	0.23	0.24	0.25	0.24	0.25
Flow ratios:									
AP/BX	4.8	4.5	4.3	4.3	4.4	4.4	4.3	4.3	4.3
BS/BX	0.83	0.82	0.79	0.81	0.80	0.82	0.82	0.80	0.82
<u>Inlet stream compositions</u>									
BS stream:									
HNO <sub>3</sub> , M	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10
HAN, M	0.13	0.13	0.13	0.13	0.13	0.13	0.13	0.13	0.13
BS stream:									
% TBP	30	30	30	30	30	30	30	30	30
AP stream:									
% TBP	30	30	30	30	30	30	30	30	30
HNO <sub>3</sub> , M	0.05	0.03	0.15	0.15	0.02	0.13	0.13	0.28	0.24
HNO <sub>2</sub> , M	0.0005	0.001	<0.002	0.0004	0.0003	0.043	0.035	0.045	0.040
U, g/L	69	84	92	87	87	73	73	74	80
Pu, g/L	1.5	1.2	0.90	0.65	0.68	1.9	1.8	1.9	1.5
Ru-106, μCi/L	48	16	4.6	8.7	10.3	24	17.5	a	38
Cs-134, μCi/L	<0.3	<0.5	<0.1	<0.1	<0.1	<0.2	<0.1	<0.8	<0.5
Cs-137, μCi/L	<0.5	<0.5	<0.2	<0.2	<0.2	<0.2	<0.2	<0.8	<0.2

Table A-10 (continued)

	Run No.								
	2-1A	2-1B	2-1C	2-1D	2-1E	2-1F	2-1G	2-1H	2-1J
<u>Outlet stream compositions</u>									
BP stream:									
HNO <sub>3</sub> , M	0.47	0.30	0.78	0.74	0.28	0.49	0.49	0.96	0.94
Reducing, N	0.1	0.22	0.21	0.24	0.38	<0.01	<0.01	<0.05	<0.01
U, g/L	11	38	27	34	40	27	27	6.7	9.3
Pu, g/L	9.4	4.9	4.4	2.6	2.8	7.0	6.1	3.1	2.1
Pu(IV), %	13	2.2	1.8	1.3	0.6	72	83	96	92
Ru-106, µCi/L	54	13.5	8.1	11.4	9.5	40	53	21.6	30
Cs-134, µCi/L	<2	3.0	0.48	0.40	1.1	0.81	<0.5	<0.8	<1
Cs-137, µCi/L	8.1	10.4	1.6	1.0	3.2	2.2	1.6	3.2	3.0
BU stream:									
HNO <sub>3</sub> , M	0.03	<0.01	0.01	<0.01	<0.01	0.04	0.05	0.05	0.04
HNO <sub>2</sub> , M	a	a	a	a	a	0.010	0.031	0.029	0.029
U, g/L	57.5	61	69	68	64	57	59	62	62
Pu, mg/L	1.95	0.6	8.2	1.7	1.3	100	130	760	1300
Ru-106, µCi/L	a	14.4	9.5	7.6	7.6	18.5	16.4	10.0	24
Cs-134, µCi/L	a	<0.08	<0.1	<0.05	<0.1	<0.2	<0.1	<0.08	<0.5
Cs-137, µCi/L	a	<0.1	<0.2	<0.1	<0.2	<0.2	<0.2	<0.1	<0.8

<sup>a</sup>Not measured.

Table A-11. Run 2-1 second-cycle tests - plutonium scavenging bank conditions and results

	Run No.								
	2-1A	2-1B	2-1C	2-1D	2-1E	2-1F	2-1G	2-1H	2-1J
Dates	3/26-27/79	3/27/79	3/27/79	3/27-28/79	3/28/79	3/29/79	3/29/79	3/30/79	3/30/79
Bank temperature, °C	40	39-41	38-39	40-41	39-40	40	42	40	42
Number of stages: strip/scrub	16/0	16/0	16/0	16/0	16/0	16/0	16/0	16/0	16/0
CX flow rate, L/h	0.12	0.13	0.13	0.13	0.13	0.14	0.14	0.13	0.14
Flow ratio: BU/CX	10.9	9.4	8.8	8.6	9.2	9.1	9.2	9.5	9.1
<u>Inlet stream compositions</u>									
CX stream:									
HNO <sub>3</sub> , M	0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20
HAN, M	1.52	1.52	1.52	1.52	1.52	1.52	1.52	1.52	1.52
BU stream:									
% TBP	30	30	30	30	30	30	30	30	30
HNO <sub>3</sub> , M	0.03	<0.01	0.01	<0.01	<0.01	0.04	0.05	0.05	0.04
HNO <sub>2</sub> , M	a	a	a	a	a	0.010	0.031	0.029	0.029
U, g/L	57.5	61	69	68	64	57	59	62	62
Pu, mg/L	1.95	0.64	8.2	1.7	1.3	100	130	760	1300
Ru-106, µCi/L	a	14.4	9.5	7.6	7.6	18.5	16.4	10	24
Cs-134, µCi/L	a	<0.08	<0.1	<0.05	<0.1	<0.2	<0.1	<0.08	<0.5
Cs-137, µCi/L	a	<0.1	<0.2	<0.1	<0.2	<0.2	<0.2	<0.1	<0.8

Table A-11 (continued)

	Run No.								
	2-1A	2-1B	2-1C	2-1D	2-1E	2-1F	2-1G	2-1H	2-1J
<u>Outlet stream compositions</u>									
CP stream:									
HNO <sub>3</sub> , M	0.08	0.06	0.11	0.09	0.08	0.52	0.47	0.51	0.55
U, g/L	13	35	22	20	18	16	16	17	20
Pu, mg/L	280	11	100	11	9.8	1140	1180	6800	8400
Ru-106, µCi/L	a	a	a	a	a	16.2	30.3	18.9	<0.8
Cs-134, µCi/L	a	a	a	a	a	<0.3	0.65	<0.2	<0.2
Cs-137, µCi/L	a	a	a	a	a	1.4	2.4	<0.5	<1
Ce-144, µCi/L	a	a	a	a	a	a	a	a	8.1
CW stream:									
U, g/L	55	54	65	63	62	59	55	61	59
Pu, mg/L	2.0	0.25	0.081	0.13	0.079	0.37	0.35	1.1	1.2
Ru-106, µCi/L	a	a	a	a	a	14.6	13.5	9.2	12.4
Cs-134, µCi/L	a	a	a	a	a	<0.1	<0.1	<0.08	<0.5
Cs-137, µCi/L	a	a	a	a	a	<0.2	<0.2	<0.1	<0.5

<sup>a</sup>Not measured.

Table A-12. Run 2-3 second-cycle tests — extraction/scrub bank conditions and results

	Run No.		
	2-3A	2-3B	2-3C
Dates	5/29/79	5/30/79	5/30-31/79
Bank temperature, °C	41-42	42	40
Number of stages, scrub/int. scrub/extrac.	4/2/10	4/2/10	4/2/10
AX flow rate, L/h	0.810	0.781	0.816
Flow ratios:			
AS/AX	0.15	0.15	0.15
AIS/AX	0.10	0.11	0.075
AF/AX	1.08	1.08	0.95
<u>Inlet stream compositions</u>			
AS stream:			
HNO <sub>3</sub> , M	0.80	0.80	0.80
AIX stream:			
HNO <sub>3</sub> , M	3.0	3.0	3.0
AX stream:			
% TBP	30	30	30
AF stream:			
HNO <sub>3</sub> , M	3.2	3.2	3.2
U, g/L	72	72	72
Pu, g/L	6.5	6.5	6.5
Ru-106, mCi/L	3.45	3.45	3.45
Sb-125, mCi/L	0.30	0.30	0.30
Cs-134, mCi/L	0.24	0.24	0.24
Cs-137, mCi/L	0.88	0.88	0.88
Ce-144, mCi/L	0.17	0.17	0.17
Eu-154, mCi/L	0.050	0.050	0.050

Table A-12 (continued)

	Run No.		
	2-3A	2-3B	2-3C
<u>Outlet stream compositions</u>			
AW stream:			
HNO <sub>3</sub> , M	2.9	a	a
U, mg/L	0.69	0.60	a
Pu, mg/L	0.0076	0.033	0.010
Ru-106, mCi/L	a	3.51	3.62
Sb-125, mCi/L	0.39	0.30	0.27
Cs-134, mCi/L	0.35	0.27	0.33
Cs-137, mCi/L	1.22	1.05	1.19
Ce-144, mCi/L	0.16	0.22	0.16
Eu-154, mCi/L	0.054	0.11	0.065
AP stream:			
HNO <sub>3</sub> , M	a	0.05	0.07
HNO <sub>2</sub> , M	0.0017	0.0020	0.0012
U, g/L	65	70	62
Pu, g/L	6.3	6.3	7.5
Ru-106, μCi/L	24.3	9.2	10.8
Cs-134, μCi/L	a	<0.5	a
Cs-137, μCi/L	a	<0.5	a

<sup>a</sup>Not measured.



Table A-13. Run 2-3 second-cycle tests -- strip bank conditions and results

	Run No.		
	2-3A	2-3B	2-3C
Dates	5/29/79	5/30/79	5/29-30/79
Bank temperature, °C	41	42	27
Number of stages: strip/scrub	13/3	13/3	13/3
BX flow rate, L/h	0.26	0.25	0.26
Flow ratios:			
AP/BX	3.2	3.1	3.1
BS/BX	1.2	1.2	1.6
<u>Inlet stream compositions</u>			
BX stream:			
HNO <sub>3</sub> , M	0.095	0.095	0.13
HAN, M	0.36	0.36	--
BS stream:			
% TBP	30	30	30
HNO <sub>2</sub> , M	--	0.080	0.47
AP stream:			
% TBP	30	30	30
HNO <sub>3</sub> , M	a	0.05	0.07
HNO <sub>2</sub> , M	0.0017	0.0020	0.0012
U, g/L	65	70	62
Pu, g/L	6.3	6.3	7.5
Ru-106, µCi/L	24.3	9.2	10.8
Cs-134, µCi/L	a	<0.5	a
Cs-137, µCi/L	a	<0.5	a

Table A-13 (continued)

	Run No.		
	2-3A	2-3B	2-3C
<u>Outlet stream compositions</u>			
BP stream:			
HNO <sub>3</sub> , <u>M</u>	0.51	0.56	0.67
Reducing, <u>N</u>	1.08	0.42	--
U, g/L	0.66	0.74	7.3
Pu, g/L	21	20	20
Pu(IV), %	0.4	0.9	76
Ru-106, $\mu$ Ci/L	a	8.1	10.8
Cs-134, $\mu$ Ci/L	a	a	0.35
Cs-137, $\mu$ Ci/L	a	1.1	<1
BU stream:			
HNO <sub>3</sub> , <u>M</u>	a	a	0.03
HNO <sub>2</sub> , <u>M</u>	a	~0.001	0.026
U, g/L	47	52	37
Pu, mg/L	0.5	0.3	10
Ru-106, $\mu$ Ci/L	13	a	24
Cs-134, $\mu$ Ci/L	0.32	0.4	a
Cs-137, $\mu$ Ci/L	3.5	a	<0.8
Cs-144, $\mu$ Ci/L	a	a	1.1

<sup>a</sup>Not measured.

Table A-14. Run 2-3 second-cycle tests — plutonium scavenging bank conditions and results

	Run No.		
	2-3A	2-3B	2-3C
Dates	5/29/79	5/30/79	5/30-31/79
Bank temperature, °C	41	42	27
Number of stages: strip/scrub	16/0	16/0	16/0
CX flow rate, L/h	0.12	0.13	0.16
Flow ratio: BU/CX	9.3	8.2	7.6
<u>Inlet stream compositions</u>			
CX stream:			
HNO <sub>3</sub> , M	0.19	0.19	0.39
HAN, M	1.5	1.5	--
BU stream:			
% TBP	30	30	30
HNO <sub>3</sub> , M	a	a	0.03
HNO <sub>2</sub> , M	a	~0.001	0.026
U, g/L	47	50	37
Pu, mg/L	0.5	0.31	10
Ru-106, µCi/L	12.7	a	24
Cs-134, µCi/L	a	a	0.4
Cs-137, µCi/L	0.32	a	a
Ce-144, µCi/L	3.5	a	1.1

Table A-14 (continued)

	Run No.		
	2-3A	2-3B	2-3C
<u>Outlet stream compositions</u>			
CP stream:			
HNO <sub>3</sub> , $\bar{M}$	a	0.05	0.42
U, g/L	13	8.4	32
Pu, mg/L	0.30	0.30	30
Ru-106, $\mu\text{Ci/L}$	14.6	11.4	9.2
Cs-134, $\mu\text{Ci/L}$	0.46	0.43	0.22
Cs-137, $\mu\text{Ci/L}$	1.4	0.24	a
Ce-144, $\mu\text{Ci/L}$	1.5	2.4	1.4
CW stream:			
HNO <sub>3</sub> , $\bar{M}$	a	a	a
HNO <sub>2</sub> , $\bar{M}$	a	a	0.01
U, g/L	50	50	34
Pu, mg/L	0.27	a	0.031
Ru-106, $\mu\text{Ci/L}$	16.2	7.0	38
Cs-137, $\mu\text{Ci/L}$	0.49	0.27	a
Cs-144, $\mu\text{Ci/L}$	a	<0.5	a

<sup>a</sup>Not measured.

Table A-15. Run 2-4 second-cycle tests — extraction/scrub bank conditions and results

	Run No.	
	2-4B	2-4C
Dates	6/20/79	6/22/79
Bank temperature, °C	26-28	52
Number of stages: scrub/int. scrub/extraction	2/6/8	2/6/8
AX flow rate, L/h	0.36	0.35
Flow ratios:		
AS/AX	0.35	0.35
AIS/AX	0.16	0.16
AF/AX	2.96	2.73
<u>Inlet stream compositions</u>		
AS stream:		
HNO <sub>3</sub> , <u>M</u>	0.09	0.09
HAN, <u>M</u>	0.02	0.02
N <sub>2</sub> H <sub>4</sub> , <u>M</u>	0.01	0.01
AIX stream:		
HNO <sub>3</sub> , <u>M</u>	3.0	3.0
AX stream:		
% TBP	30	30
AF stream:		
HNO <sub>3</sub> , <u>M</u>	1.2	1.3
Reducing, <u>N</u>	0.44	0.35
U, g/L	46	48
Pu, g/L	4.7	4.6
Ru-106, μCi/L	180	180
Ce-144, μCi/L	0.85	0.88

Table A-15 (continued)

	Run No.	
	2-4B	2-4C
<u>Outlet stream compositions</u>		
U-Pu product (aqueous):		
HNO <sub>3</sub> , M	1.2	1.3
U, g/L	14	23
Pu, mg/L	3.75	4.3
Ru-106, $\mu$ Ci/L	a	5.7
Cs-137, $\mu$ Ci/L	a	0.19
U product (organic):		
HNO <sub>3</sub> , M	<0.01	<0.01
U, g/L	62	55
Pu, mg/L	0.43	0.62
Ru-106, $\mu$ Ci/L	23	8.2
Cs-137, $\mu$ Ci/L	a	<0.2

<sup>a</sup>Not measured.

Table A-16. Run 2-4 second-cycle tests - strip bank conditions and results

	Run No.	
	2-4B	2-4C
Dates	6/21-22/79	6/22/79
Bank temperature, °C	51	50-51
Number of stages: strip/scrub	16/0	16/0
BX flow rate, L/h	0.36	0.35
Flow ratios: AP/BX	1.11	1.17
<u>Inlet stream compositions</u>		
BX stream:		
HNO <sub>3</sub> , <u>M</u>	0.012	0.012
AP stream:		
% TBP	30	30
HNO <sub>3</sub> , <u>M</u>	<0.01	<0.01
U, g/L	62	55
Pu, mg/L	0.43	0.62
Ru-106, μCi/L	23	8.2
Cs-137, μCi/L	a	<0.2

Table A-16 (continued)

	Run No.	
	2-4B	2-4C
<u>Outlet stream compositions</u>		
BP stream:		
HNO <sub>3</sub> , M	0.02	0.02
U, g/L	69	65
Pu, mg/L	<0.55	0.22
Ru-106, $\mu$ Ci/L	6.8	8.1
Cs-134, $\mu$ Ci/L	a	0.49
Cs-137, $\mu$ Ci/L	1.35	1.05
Ce-144, $\mu$ Ci/L	2.43	a
BU stream:		
U, mg/L	120	<0.1
Pu, mg/L	0.002	0.019
Ru-106, $\mu$ Ci/L	11.4	11.9
Cs-134, $\mu$ Ci/L	<0.2	0.2
Cs-137, $\mu$ Ci/L	<0.3	<0.9
Ce-144, $\mu$ Ci/L	<0.8	a

<sup>a</sup>Not measured.



## 8. REFERENCES

1. J. H. Goode and R. G. Stacy, Head-End Reprocessing Studies with H. B. Robinson-2 Fuel, ORNL/TM-6037 (June 1978).
2. W. T. McDuffee, V. C. A. Vaughen, F. A. Kappelmann, and W. R. Whitson, Recovery of Multigram Quantities of Curium-242 and Curium-244 in the Curium Recovery Facility, ORNL-4241 (December 1969).
3. A. L. Mills, Meas. Control 1, 87-89 (March 1968).
4. Savannah River Quarterly Report, Light Water Reactor Fuel Recycle, October-December 1976, DP-ST-LWR-76-1-4 (1977).
5. J. E. Bigelow, E. D. Collins, and L. J. King, "The CLEANEX Process: A Versatile Solvent Extraction Process for Recovery and Purification of Lanthanides, Americium, and Curium," Actinides Separations, ACS Symp. Series 117, pp. 147-56, ed. by J. D. Navratil and W. W. Schulz, American Chemical Society, Washington, D.C. (1980).
6. E. D. Collins et al., Solvent Extraction Studies of Coprocessing Flow-sheets - Results from Campaigns 3 and 4 of the Solvent Extraction Test Facility (SETF), ORNL-7991 (May 1982).
7. M. J. Plodinic, Clarification of Light Water Reactor Dissolver Solutions, DP-1516 (December 1978).
8. E. J. Wheelwright et al., Technical Summary Nuclear Waste Vitrification Project, PNL-3038 (May 1979).
9. A. D. Mitchell, A Comparison Between SEPHIS-MOD 4 and Previous Models of the Purex Solvent Extraction System, ORNL/TM-6565 (February 1979).
10. G. L. Richardson and J. L. Swanson, Plutonium Partitioning in the Purex Process with Hydrazine-Stabilized Hydroxylamine Nitrate, HEDL-TME-75-31 (1975).
11. M. C. Thompson, G. A. Burney, and J. M. McKibben, Recent Savannah River Experience and Development with Actinide Separations, DP-MS-78-64 (April 1979).
12. G. S. Barney, "A Kinetic Study of the Reaction of Plutonium(IV) with Hydroxylamine," J. Inorg. Nucl. Chem. **38**, 1677 (1976).
13. A. Bathellier, "The Use of Nitrous Acid as a Backwashing Agent for Actinide-Bearing Solvents," Proc. Int. Conf. Solvent Extr. Chem. Met., UKAEA (September 1965).
14. T. Tsujino, T. Aochi, and T. Hoshino, "Reduction Stripping of Plutonium Loaded on TBP with Addition of Nitrous Acid," J. Nucl. Sci. Technol. **13(6)**, 321-26 (June 1976).

15. M. S. Okamoto, Coprocessing Solvent Extraction Studies, DP-MS-77-76 (March 1978).
16. B. A. Hannaford and G. D. Davis, Plutonium Flowsheet Development in Miniature Mixer-Settlers, ORNL/TM-7408 (May 1981).



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