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I-33818

DR#0433-2

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ORNL/TM-10266

**OAK RIDGE  
NATIONAL  
LABORATORY**

**MARTIN MARIETTA**

## **Solvent Extraction Studies of 10% TBP Flowsheets in the Solvent Extraction Test Facility Using Irradiated Fuel from the Fast Flux Test Facility**

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OPERATED BY  
MARTIN MARIETTA ENERGY SYSTEMS, INC.  
FOR THE UNITED STATES  
DEPARTMENT OF ENERGY

Printed in the United States of America. Available from  
the U.S. Department of Energy  
Technical Information Center  
P.O. Box 62, Oak Ridge, Tennessee 37830

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ORNL/TM-10266  
Dist. Category UC-86T  
(Applied)

### Consolidated Fuel Reprocessing Program

#### SOLVENT EXTRACTION STUDIES OF 10% TBP FLOWSHEETS IN THE SOLVENT EXTRACTION TEST FACILITY USING IRRADIATED FUEL FROM THE FAST FLUX TEST FACILITY

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ORNL/TM--10266  
TI88 007864

Chemical Technology Division

Date Issued - March 1988

**Notice: This document contains information of a preliminary nature. It is subject to revision or correction and therefore does not represent a final report.**

Prepared for the  
Office of Facilities, Fuel Cycle, and Test Programs  
(AF 20 30 20 0)

Prepared by the  
OAK RIDGE NATIONAL LABORATORY  
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MARTIN MARIETTA ENERGY SYSTEMS, INC.  
for the  
U.S. DEPARTMENT OF ENERGY  
under contract DE-AC05-84OR21400

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# SOLVENT EXTRACTION STUDIES OF 10% TBP FLOWSHEETS USING IRRADIATED FUEL FROM THE FAST FLUX TEST FACILITY

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## ABSTRACT

Two solvent extraction experiments were made in the Solvent Extraction Test Facility (SETF) during Campaign 10 to continue the evaluation of (1) a computer control system for the coextraction-coscrub contactor, and (2) a partitioning technique that separates uranium and plutonium without the aid of chemical reductants. The Fast Flux Test Facility (FFTF) fuel used in this campaign had burnups of ~55 and ~60 (average) MWd/kg. During both experiments, the computer control system successfully maintained stable, efficient operation. The control system used an in-line photometer to monitor the plutonium concentration in the extraction section; and based on this data, it adjusted the addition rate of the extractant to maintain high loadings of heavy metal in the solvent and low raffinate losses. The uranium and plutonium partitioning relied entirely on the differences between the U(VI) and Pu(IV) distribution coefficients (since no reductant was used to adjust the plutonium valence). In order to enhance this difference, the TBP concentration and operating temperature were relatively low in comparison to traditional Purex flowsheets. Final product purities of 99% were achieved for both the uranium and plutonium in one cycle of partitioning.

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## 1. INTRODUCTION

The Solvent Extraction Test Facility (SETF) is located in one of the heavily shielded hot cells of the Transuranium Processing Plant at the Oak Ridge National Laboratory. Mixer-settler contactors were used to evaluate solvent extraction flowsheets for the reprocessing of irradiated, nuclear reactor fuels. A total of nine experimental campaigns have previously been completed in the SETF.<sup>1-7</sup> Results from these tests have provided information on heavy metal recoveries, fission product behavior, flowsheet schemes, in-line instrumentation, and general operability of the system.



This report describes the two solvent extraction experiments that were made for Campaign 10. The work on using a nitric acid-TBP system to partition uranium and plutonium without reducing Pu(IV) to Pu(III),<sup>8</sup> which was started in the previous campaign,<sup>7</sup> was continued in these tests. In order to better achieve this separation, a relatively low TBP concentration was selected for the solvent, 10 vol %, in place of the 20 to 30 vol % that is traditionally used in fuel reprocessing plants. Of course, this lower TBP concentration requires higher solvent flow rates to achieve the same plant throughput; but, if adequate separation can be achieved without a plutonium valence adjustment, plant operation may be greatly simplified. In many plants, the valence adjustment is accomplished by adding chemical reductants, e.g., U(IV) or hydroxylamine (HAN) stabilized with hydrazine. Because of the high concentration of plutonium in breeder fuels (~20% of the heavy metals) relatively large amounts of these reductants are required. Then, after the separation is accomplished, additional process steps must be used to remove any excess chemicals and to readjust the plutonium valence for further processing. Moreover, these chemicals are highly reactive and may require special safety controls for the plant.

The evaluation of an automatic control system for the extraction bank,<sup>6,7</sup> which was started in Campaigns 8 and 9, was also continued in Campaign 10. The objective of the control system was to maintain a high operating efficiency by maximizing the loading of heavy metals (uranium and plutonium) into the organic phase in the extraction contactor while still maintaining low losses of heavy metals to the raffinate. The control system worked by measuring the uranium and plutonium concentration in an intermediate stage in the extraction bank (monitored variable) using an in-line photometer and then varying the addition rate of the extractant (controlled variable) to maintain the plutonium concentration within a desired range that should yield good results.

The fuel used in the first experiment had a burnup of ~55 MWd/kg and had been discharged from the FFTF in May 1983; the fuel for the final run consisted of a mixture of fuel pieces that had burnups of ~2, ~36, ~55, and ~90 MWd/kg (average burnup of ~60 MWd/kg) and cooling times of 5.2, 3.4, 3, and 2 years, respectively.

The processing steps used in this campaign included: (1) dissolution of the fuel in nitric acid ( $\text{HNO}_3$ ), (2) clarification of the dissolver solutions by filtration, (3) adjustment of the dissolver solution to the proper concentrations and plutonium valence for solvent extraction, (4) solvent extraction processing with partitioning of the uranium and plutonium, (5) purification of the plutonium by nitrate-based anion exchange, and finally (6) conversion of the plutonium to an oxide form by oxalate precipitation and calcination.

## 2. EQUIPMENT AND OPERATIONAL PROCEDURES

Most of the major equipment items and general operating procedures used in Campaign 10 were similar to those used before and described for previous campaigns. A description of the general layout and equipment, and operation of the solvent extraction contactors is given in ref. 1; the fuel dissolution is described in refs. 5 and 7; the clarification and feed adjustment steps in ref. 5; the filtration equipment in the refs. 2 and 3; the automatic control system in refs. 7 and 9; the in-line photometer system in ref. 6; and the plutonium purification and conversion to oxide in refs. 3 and 6.

The only major equipment change included the addition of an instream heat exchanger for the product stream from the extraction contactor (which is the feed stream for the partition contactor). The heat exchanger was used to cool the solvent from the extraction bank and minimize temperature variations caused by the different operating temperatures-- $\sim 40^\circ\text{C}$  for the extraction bank vs  $\sim 10^\circ\text{C}$  for the partition bank.

## 3. DESCRIPTION OF THE SOLVENT EXTRACTION FLOWSHEETS

The investigation of first-cycle flowsheet options using 10% TBP, which was begun in Campaign 9 (Run 9-3), was continued. Illustrations of the flowsheets and operating conditions used in Campaign 10 are shown in Figs. 1 and 2; detailed stream analyses for each run are tabulated in the Appendix. Description of the Run 9-3 flowsheet is given in ref. 7.

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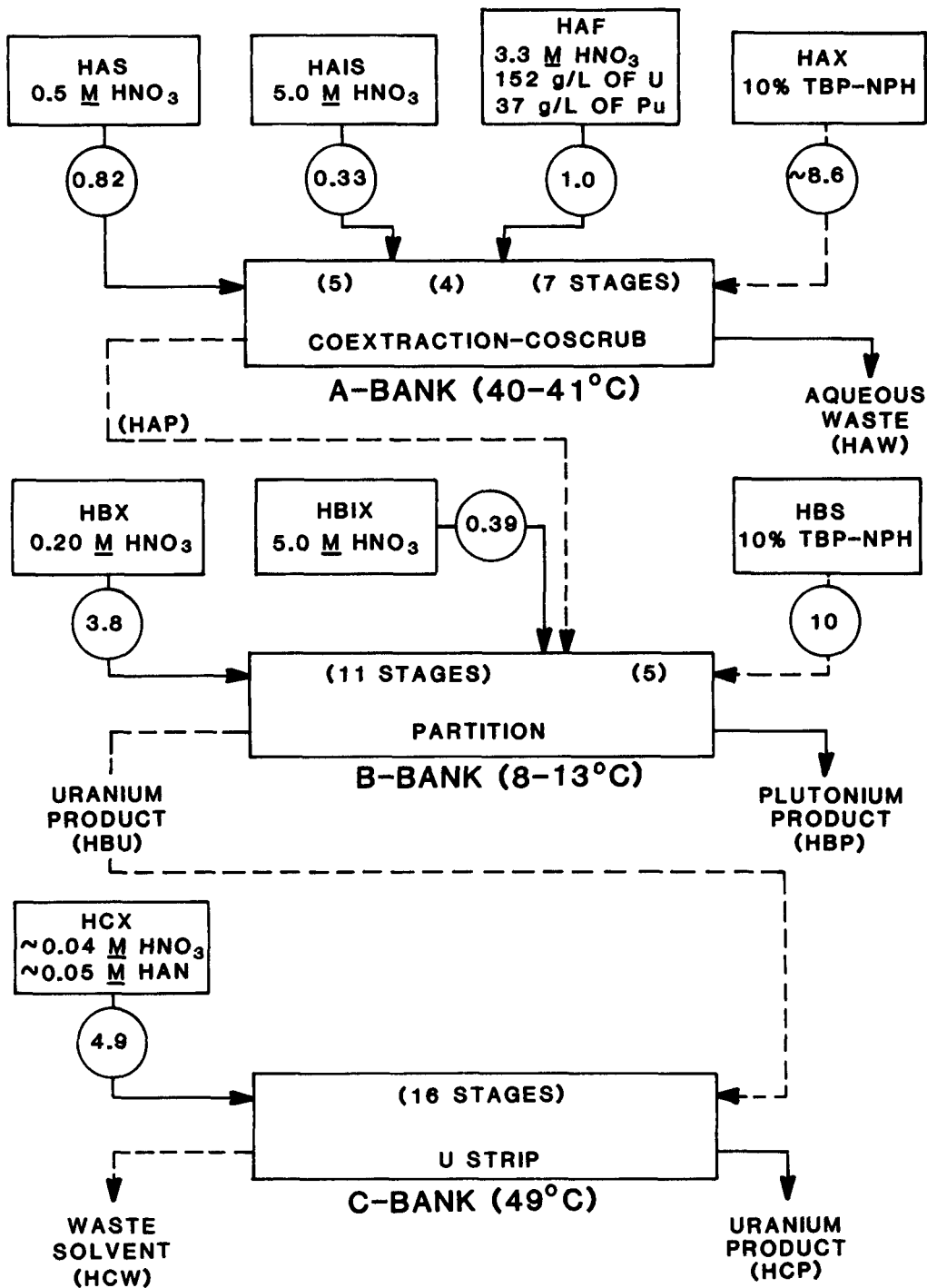


Fig. 1. Flowsheet for Run 10-1.

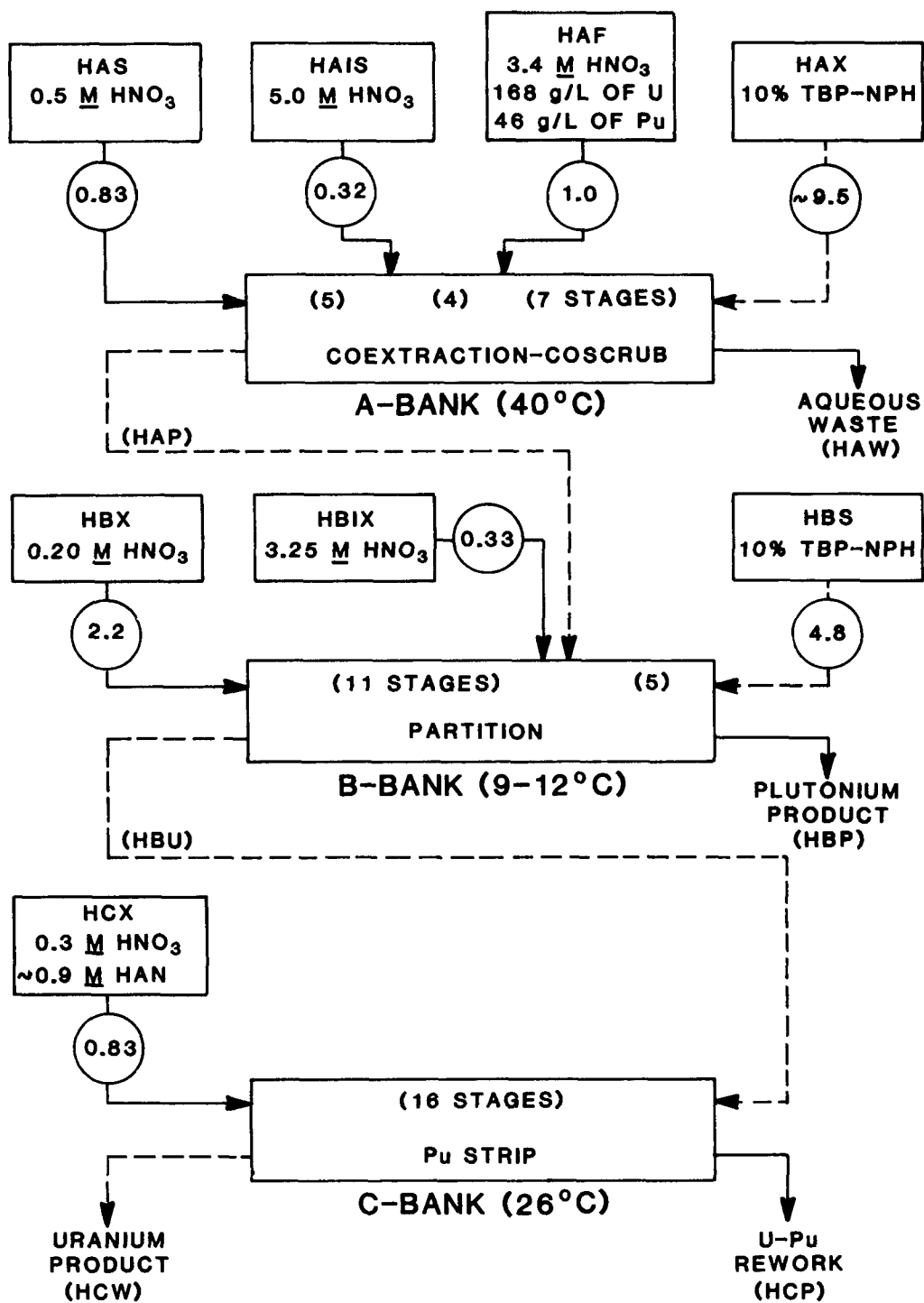


Fig. 2. Flowsheet for Run 10-2.

In each of the three runs (9-3, 10-1, and 10-2) the conditions used in the coextraction-coscrub contactor (A-bank) were similar with respect to the organic solvent, the operating temperature, the acid molarity of the scrubs, the arrangement of the stages, and the algorithm for the automatic control system. The only notable difference was in the type of fuel processed for each run--~90 MWd/kg, ~55 MWd/kg, and ~60 MWd/kg (average) for Runs 9-3, 10-1 and 10-2, respectively.

In Runs 9-3 and 10-1, partitioning was accomplished in B-bank without using a plutonium reductant. The separation relied entirely on the relative differences between U(VI) and Pu(IV) distribution coefficients (defined as the ratio of the organic and aqueous concentrations in units of g/L). This difference is enhanced by using lower TBP concentrations and operating temperatures<sup>8</sup> than have traditionally been used in reprocessing plants. The conditions chosen for Run 9-3, 10% TBP and 15 to 18°C, yielded good results and demonstrated the partitioning technique using 16-stage mixer-settlers. For Run 10-1, the following changes were made in an attempt to improve the overall separation; (1) lowering the temperature further to 8 to 13°C, and (2) decreasing the organic to aqueous phase ratio in the strip section from 6.2 to 4.9 to make a more dilute plutonium product (~10 g/L instead of ~15 g/L). The conditions for the uranium strip contactor remained essentially unchanged for Runs 9-3 and 10-1.

In Run 10-2, both B-bank and C-bank were used for partitioning. The primary goal in that arrangement was to make (1) a plutonium product containing <1 to 2 % uranium (U DF >100), and (2) a uranium product containing <1 ppm plutonium (Pu DF >2E5), which is equivalent to <100 nCi of Pu per gram of U. The uranium product could then be considered non-transuranic, based on current regulations of the federal government, which could greatly simplify subsequent processing or disposal (depending on whether the uranium was designated as a product or waste). The bulk of the separation was accomplished in B-bank using a nonreductant flowsheet similar to Runs 9-3 and 10-1 in order to make the plutonium product (HBP).

Residual plutonium in the solvent from B-bank (HBU) was removed in C-bank with a  $\text{HNO}_3$  solution containing the reductant, HAN. In a reprocessing plant, the solvent from the C-bank (HCW) would be taken to a fourth contactor for recovery of the uranium with a dilute  $\text{HNO}_3$  strip; however, the SETF has only three contactors, so this step was omitted in our demonstration. The aqueous stream from C-bank (HCP) would be recycled back to the feed tanks after treatment to remove HAN and to adjust the plutonium valence. In order to minimize the amount of uranium that was stripped into this rework stream, a relatively large organic to aqueous phase ratio was used and a large excess of HAN (relative to that needed for plutonium reduction) was included to act as an inextractable nitrate salt. It should be noted that the C-bank contactor was considerably oversized for its intended use in this run--a reprocessing plant would probably need only one-half to one-fourth the number of stages used for this run.

#### 4. EXPERIMENTAL RESULTS AND DISCUSSIONS OF SOLVENT EXTRACTION TESTS

##### 4.1 COEXTRACTION-COSCRUB CONTACTOR

The coextraction-coscrub bank was again operated at high loadings of heavy metals by using the in-line photometer and computer control system that was originally developed in Campaigns 8 and 9. The in-line photometer measured the plutonium concentration in the solvent from an intermediate stage in the extraction section (Stage 12), where the heavy metal inventory was changing in response to flowsheet variations. During start-up, the addition rate of the extractant (HAX) was set at ~60% of the design rate in order to more quickly bring the extraction bank to steady state conditions. When plutonium was detected by the photometer, manual adjustments were made to smoothly bring the system near the desired operating range. At that point, the control system was activated, and the control algorithm used the plutonium concentration data to calculate the appropriate changes in the HAX flow rate in order to maintain the plutonium concentration near the desired set point value. All other streams (HAF, HAS, and HAI) were kept as constant as possible. The control constants in the algorithm were not changed from those used in Run 9-3 (see ref. 7 for details).

Plots of the plutonium concentration readings and the HAX flow rate vs elapsed time for Runs 10-1 and 10-2 are shown in Figs. 3 and 4. Run 10-1 was put on automatic control after ~5.5 h of operation. After the initial overshoot, the plutonium concentration showed two relatively symmetric curves until the profile sampling at 19 and 27 h disturbed the contactor. The only difference between the two sampling periods was the phase that was taken--organic phase at 19 h and aqueous phase at 27 h. The sampling of the organic phase had a relatively small impact on the contactor operation, which the control system easily corrected. However, sampling of the aqueous phase caused a much greater perturbation because the concentrations of heavy metals in the aqueous phase are much larger and the aqueous flow rates are lower; consequently, sampling the aqueous phases takes more heavy metals from the contactor which are then replaced more slowly. The control system appeared to respond properly to this perturbation; and in spite of a transient electronic problem that forced a return to manual control for ~15 min during this period, the computer probably would have eventually brought the plutonium concentration back to the set point. Unfortunately, the run ended before this was demonstrated.

For Run 10-2, the set point was lowered from 6 to 5 g of Pu/L to help ensure low cumulative losses to the raffinate. Samples from Run 10-1, had shown that the aqueous losses increased from ~0.04% to ~0.2% when the Stage 12 plutonium concentration increased from ~5 g/L to ~10 g/L. The automatic control system was started after ~4.5 h of operation. Sampling profiles were not taken during this run and no known upset occurred. This run was the first in the SETF in which the control system maintained very stable operation during the entire run.

These tests show that, once the contactor has achieved near steady-state conditions, the control algorithm in its present form can maintain good control for a normally operating mixer-settler contactor. However, whether this system can correct for significant upsets is still unknown.

The overall losses of uranium and plutonium to the aqueous raffinate (HAW) were low for each of the three runs with 10% TBP (Table 1), averaging 0.008% and 0.06% for uranium and plutonium, respectively. These results are reasonably consistent with losses measured for previous runs with 30% TBP, which had averaged ~0.03% and ~0.02% for uranium and plutonium, respectively.

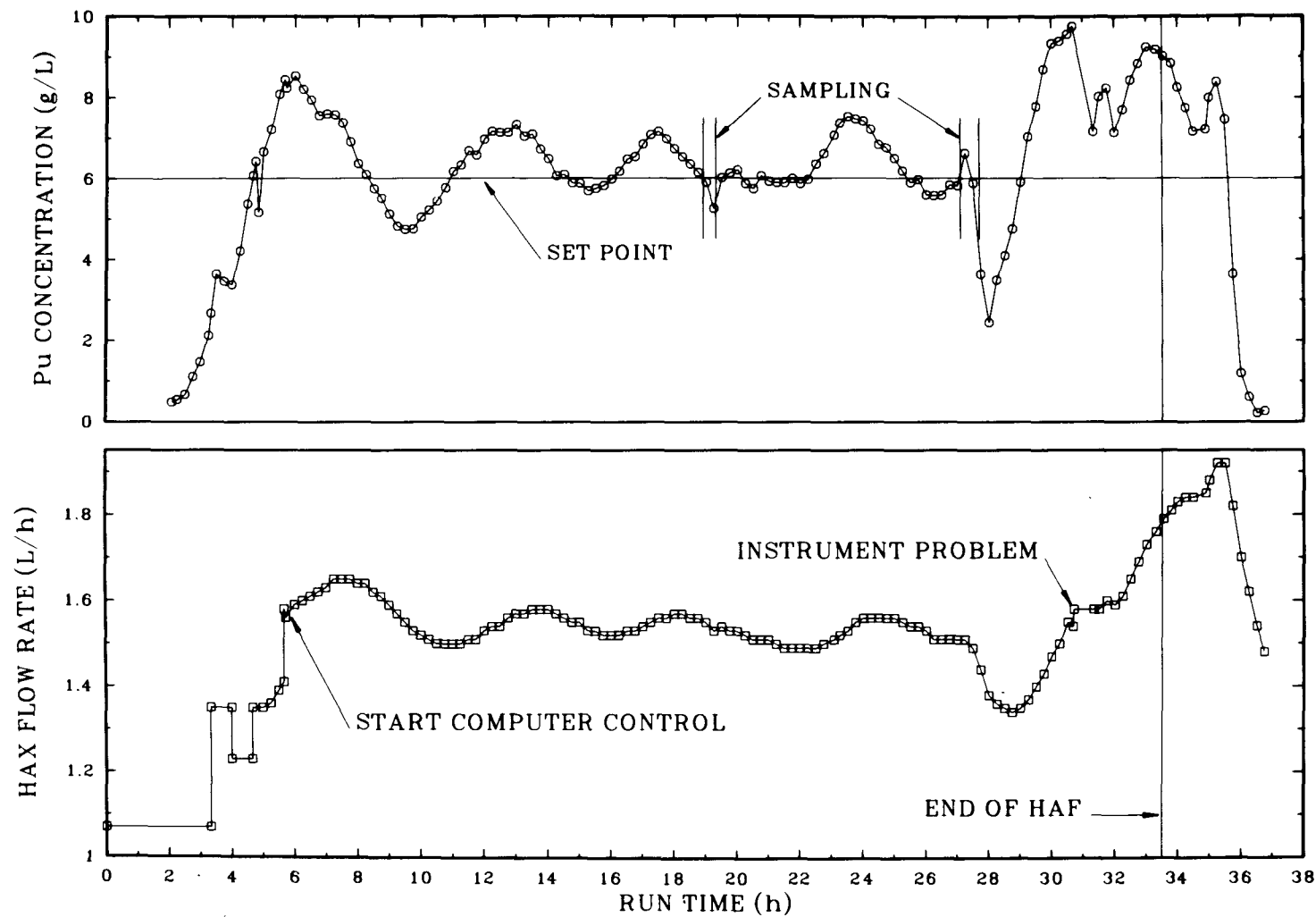


Fig. 3. Concentration readings from the in-line photometer (Stage 12) and the addition rate of the extractant (HAX) for Run 10-1.



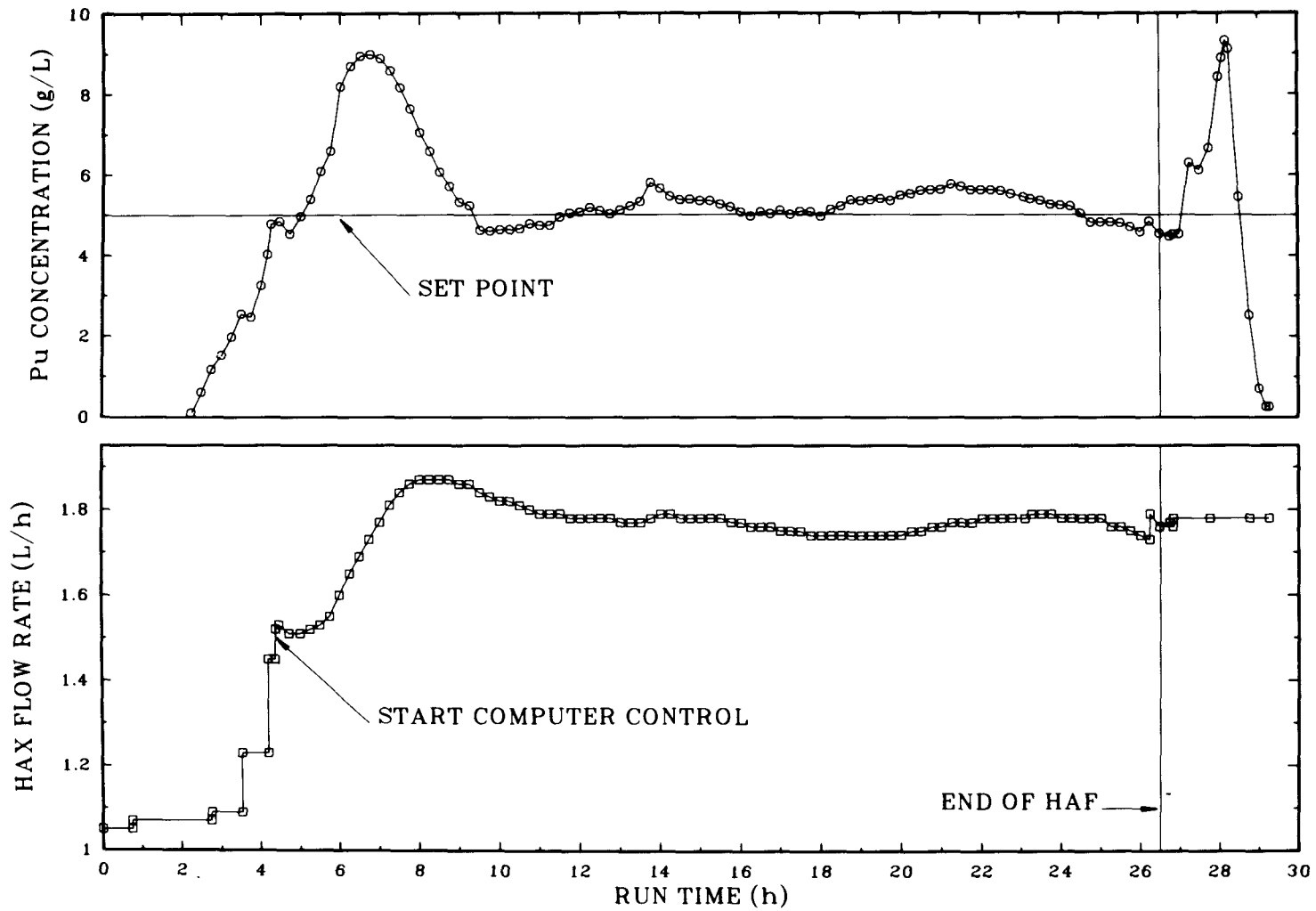


Fig. 4. Concentration readings from the in-line photometer (Stage 12) and the addition rate of the extractant (HAX) for Run 10-2.

Table 1. Distribution of uranium and plutonium in the outlet streams

Stream	Run number		
	9-3	10-1	10-2
Uranium, %			
Aq. raffinate (HAW)	3E-3	0.02	2E-4
Pu product (HBP)	0.42	0.068	0.06
U product <sup>a</sup>	99.6	99.92	95.5
U-Pu rework (HCP)			4.4%
Waste solvent (HCW)	<9E-3	6E-4	
Plutonium, %			
Aq. raffinate (HAW)	0.04	0.11	0.04
Pu product (HBP)	99.8	99.6	99.2
U product <sup>a</sup>	0.18	0.30	0.003
U-Pu rework (HCP)			0.75
Waste solvent (HCW)	1E-4	4E-4	

<sup>a</sup>HCP for Runs 9-3 and 10-1, and HCW stream for Run 10-2.

The concentration profiles for uranium, plutonium, and  $H^+$  for Run 10-1 are shown in Fig. 5. As expected, these results are similar to those measured during Run 9-3, which used a different FFTF fuel (~90 MWd/kg) but essentially the same flowsheet conditions. The peak loading of the solvent occurred in stages 5 through 10 with a solvent loading of 34 to 35 g/L of heavy metals (~80% solvent saturation). After the solvent was treated with the low acid scrub, which caused some of the heavy metals to strip and reflux, the product stream (HAP) contained ~23 g/L (~50% saturation).

The fission product decontamination factors (DFs) that were measured for the coextraction-coscrub contactor (A-bank) in the runs with 10% TBP are listed in Table 2. The only fission product that was consistently

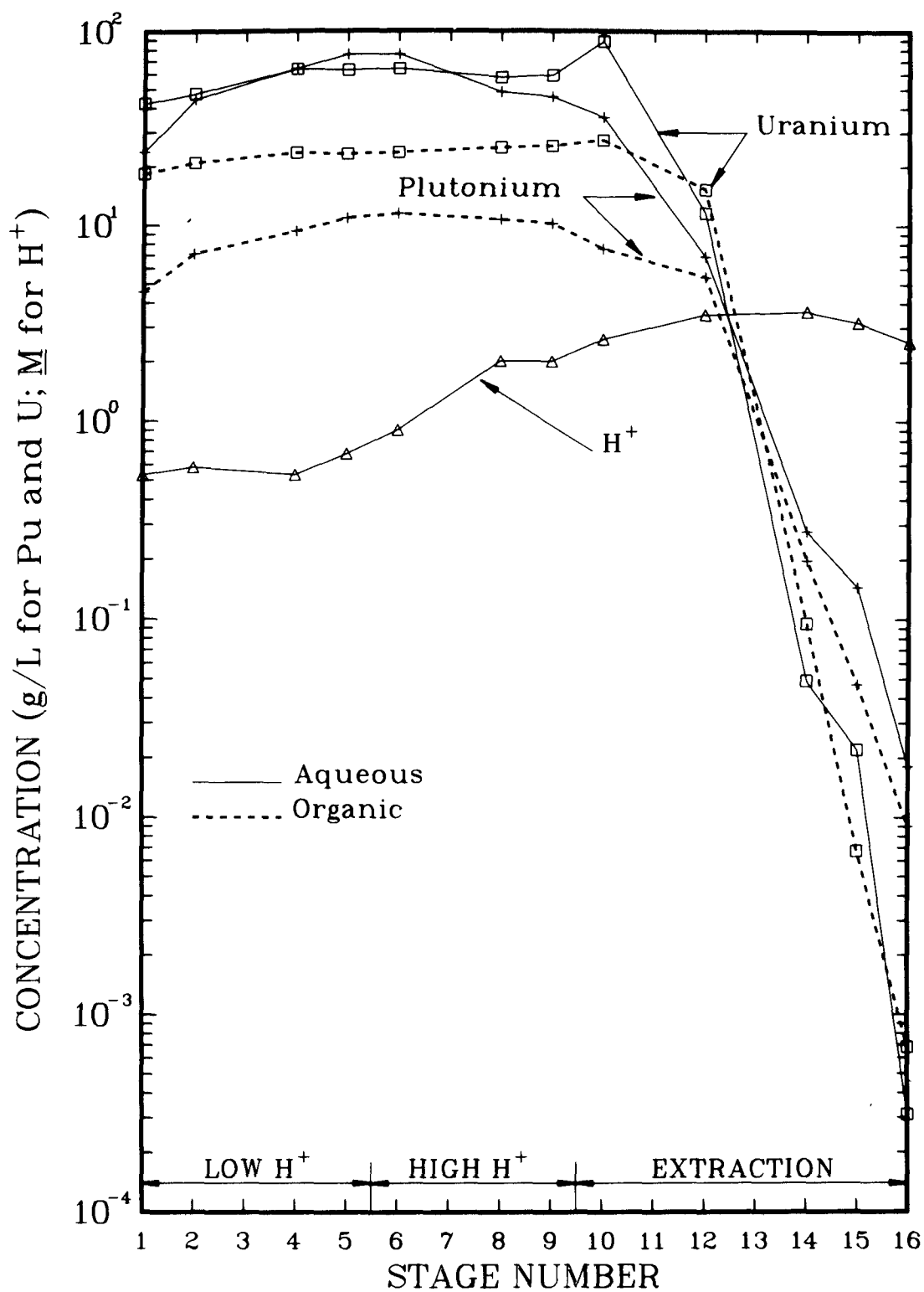


Fig. 5. Concentration profiles for uranium and plutonium in the coextraction-coscrub contactor for Run 10-1.

Table 2. Fission product decontamination results from the  
coextraction-coscrub contactor

Fission Product	Decontamination factors		
	Run 9-3	Run 10-1	Run 10-2
$^{95}\text{Zr}$	2E5		>1E5
$^{95}\text{Nb}$	1E5		4E4
$^{106}\text{Ru}$	7E4	3E4	4E4
$^{137}\text{Cs}$	>2E7	>1E7	>1E7
$^{144}\text{Ce}$	>1E7	>1E7	>5E6
$^{154}\text{Eu}$	>7E5	>2E5	>3E5

detected in the product stream (HAP) was  $^{106}\text{Ru}$ ;  $^{95}\text{Zr}$  was detected only when short-cooled fuel was used (Run 9-3). A comparison of the DFs achieved with 10 vs 30% TBP shows somewhat better results when using 10% TBP— $3 \times 10^4$  to  $7 \times 10^4$  vs  $2 \times 10^4$  to  $3 \times 10^4$  for  $^{106}\text{Ru}$ , and  $2 \times 10^5$  vs  $2 \times 10^4$  to  $3 \times 10^4$  for  $^{95}\text{Zr}$ . Cesium, cerium, and europium were not detected in the product, and the DFs shown were calculated from the limits of detection.

## 4.2 URANIUM-PLUTONIUM STRIP CONTACTORS

### 4.2.1 Nonreductive partition contactor (Runs 9-3 and 10-1)

In Runs 9-3 and 10-1, plutonium was recovered from the solvent and separated from uranium in B-bank (partition bank), while uranium was recovered from the solvent in C-bank (uranium strip bank). Each run was further divided into two parts, "A" and "B", in order to measure the effect of the HBS flow rate on the U and Pu DFs. (Because the sampling in the "B" portion of each run was limited to the stream samples from B-bank, the figures, Tables 1, 2, 4, and 6, and the Appendix only show results for the "A" portion of each run.) The partitioning results are listed in Table 3.

Table 3. Partitioning results for Runs 9-3 and 10-1 using a single partition contactor

	Run number			
	9-3A	9-3B	10-1A	10-1B
Feed solution (HAF)				
Pu, g/g of U	0.255		0.241	
Temperature, °C	13-18		8-13	
HBS flow rate, L/h	1.33	1.62	1.81	1.13
Phase ratio (O/A) <sup>a</sup>				
strip section	6.2	6.8	4.9	3.9
scrub section	2.4	2.9	2.4	1.5
Pu aq. prod. (HBP)				
Pu, g/L	14		8.7	
U, mg/g of Pu	16	2.0	2.7	58
U DF	240	1,900	1,500	72
U org. prod. (HBU)				
U, g/L	9.4		8.0	
Pu, µg/g of U	370	3,300	680	53
Pu DF	680	80	350	4,600
Overall U-Pu separation factor <sup>b</sup>	2E5	2E5	5E5	3E5

<sup>a</sup>Organic to aqueous phase ratio.

<sup>b</sup>Product of the U and Pu DFs.

A comparison of the "A" and "B" results show that relatively small adjustments in the HBS flow rate (factors of 1.2 to 1.6) can change the U and Pu DFs by fairly wide margins (factors of 8 to 20) with no significant change in the overall U-Pu separation (product of the U and Pu DFs). Consequently, minor adjustments in the HBS flow rate can be used to easily regulate the relative purity of the two products. A comparison of Runs 9-3 and 10-1 shows a modest improvement in the overall U-Pu separation (a factor of 1.5 to 2) resulting from lowering the temperature and decreasing the plutonium product concentration.

In comparison to previous runs that had used HAN, the runs without HAN naturally yielded lower U and Pu DFs (10,000 with HAN vs 100 to 1,000 without). However, the omission of HAN did have the advantage of producing a plutonium product in a clean,  $\text{HNO}_3$  solution that did not require treating to remove excess HAN or readjusting the plutonium valence for further processing. Furthermore, the chemical reactions associated with HAN were avoided, along with any concerns with respect to its by-products (gases and  $\text{HNO}_3$ ), kinetics, interferences from competing reactions (such as nitrite or fission products), or plant safety.

Concentration profiles for uranium, plutonium, and  $\text{H}^+$  for Run 10-1 are shown in Fig. 6. (Run 9-3 is shown in ref. 7)

No additional separation of U-Pu occurred in the uranium strip bank (C-bank); essentially all the residual plutonium in the solvent from the partition contactor was stripped with the uranium (Table 4). As a result, the plutonium content in the waste solvents was quite low, ranging from 5 to 8  $\mu\text{g/L}$  (ppb), while the uranium product contained 17 to 22  $\text{mg/L}$  (ppm) plutonium.

#### 4.2.2 Nonreductive partition contactor with reductive polish contactor (Run 10-2)

The nonreductive partitioning flowsheet that was used in the first partitioning contactor (B-bank) was similar to the one used in 10-1, except for a change in the phase ratios to yield a more concentrated product. The partitioning results for Run 10-2 are shown on Table 5. The U-Pu separation factor for B-bank (Pu aqueous product and the intermediate

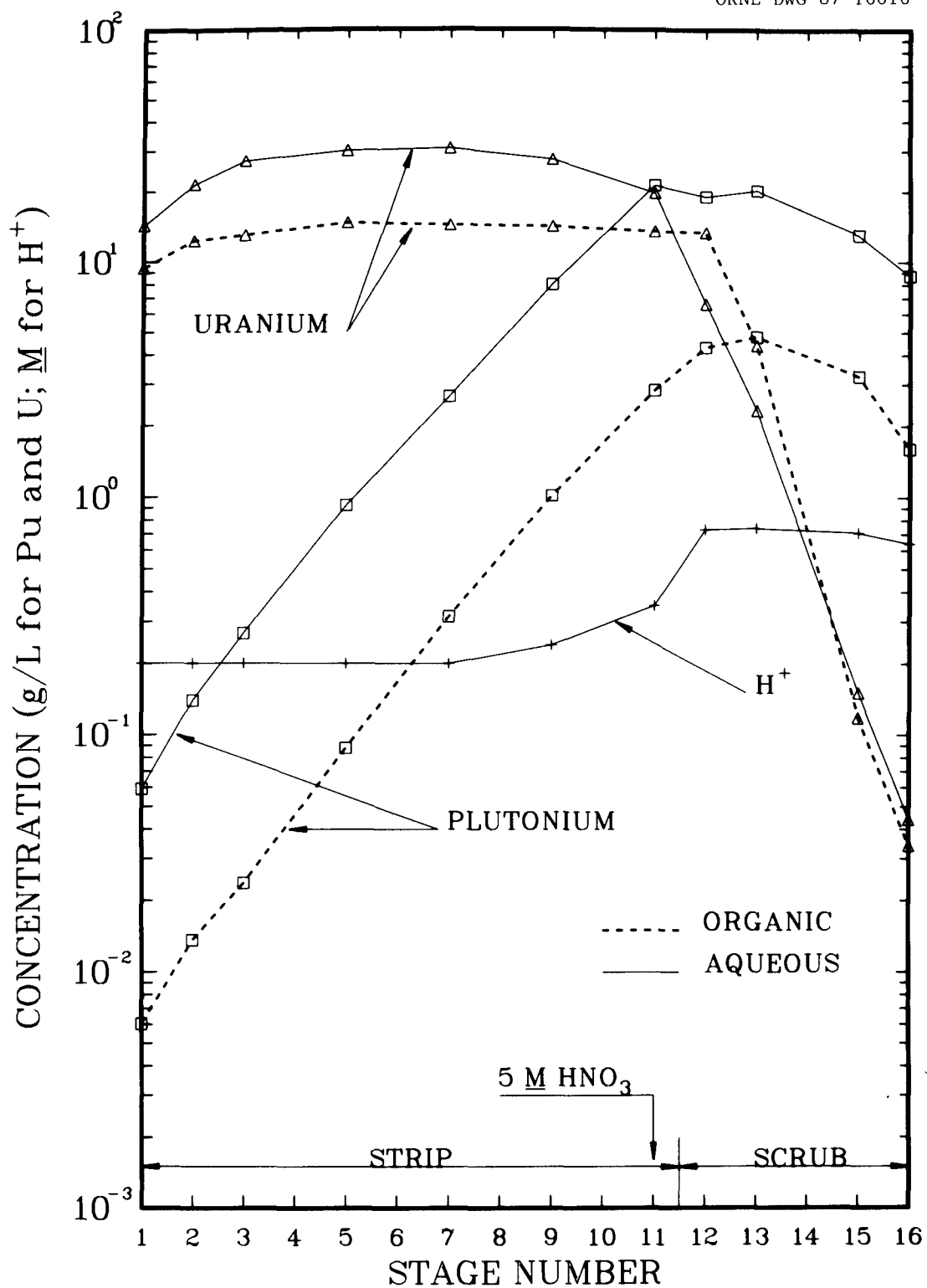


Fig. 6. Concentration profile for uranium, plutonium, and H<sup>+</sup> in the partition contactor for Run 10-1.

Table 4. Results for uranium strip contactor.

	Run number	
	9-3	10-1
U product (HCP)		
U, g/L	38	30
Pu, g/L	0.017	0.022
Pu, $\mu\text{g/g}$ of U	450	750
Waste Solvent (HCW)		
U, mg/L	<3	0.04
Pu, $\mu\text{g/L}$	5	8

U product) was  $2 \times 10^5$  which is slightly lower than the result for Run 10-1 and is probably the result of making a more concentrated product (17 vs 9 g/L) since the operating temperatures were approximately the same. Concentration profiles for uranium, plutonium, and  $\text{H}^+$  for B-bank in Run 10-2 are shown in Fig. 7.

The C-bank contactor lowered the plutonium content in the solvent to a minimum concentration of  $<7 \mu\text{g/g}$  of uranium in about 8 stages. (Whether the 1 ppm limit was actually met is unknown; lower plutonium concentrations could not be measured because of interferences with the uranium in the solvent.) The aqueous rework stream (HCP) from C-bank contained 5% of the uranium and 0.8% of the plutonium used in this run (Table 1). The plutonium DF for the final uranium product (HCW) was  $>4 \times 10^4$  which is similar to that measured for reductive partitioning in B-bank alone (Run 9-2). However, this two step technique still has the advantage of recovering the plutonium product (1) without having to rely on a sensitive chemical reaction and (2) in a clean  $\text{HNO}_3$  solution that requires no further treatments. The HAN that was used in C-bank was included primarily as an inextractable nitrate salt to lessen the amount of uranium stripped, since the amount of plutonium sent to C-bank was quite small.



Table 5. Results for Run 10-2 from the nonreductive partition contactor  
and the reductive polish contactor

	Run number	
	10-2A	10-2B
Feed solution (HAF)		
Pu, g/g of U	0.272	
<u>B-Bank (nonreductive)</u>		
Temperature, °C	9-12	
HBS flow rate, L/h	0.90	0.95
Phase ratio (O/A)		
Strip section	6.6	6.8
Scrub section	1.9	2.1
Pu aq. prod. (HBP)		
Pu, g/L	17	
U, mg/g of Pu	2.4	1.1
U DF	1,600	3,300
Intermediate U prod. (HBU)		
U, g/L	11	
Pu, µg/g of U	1,900	4,600
Pu DF	140	59
U-Pu separation factor <sup>a</sup>	2E5	2E5
<u>C-bank (with reductant, HAN)</u>		
U org. prod. (HCW)		
U, g/L	11	
Pu, µg/g of U	<7	
Pu DF	270	
<u>B-bank and C-bank</u>		
Overall Pu DF	>40,000	
U-Pu separation factor <sup>b</sup>	6E7	

<sup>a</sup>Product of the U and Pu DFs for B-bank.

<sup>b</sup>Product of the U DF for B-bank and the Pu DF for B-bank and C-bank.

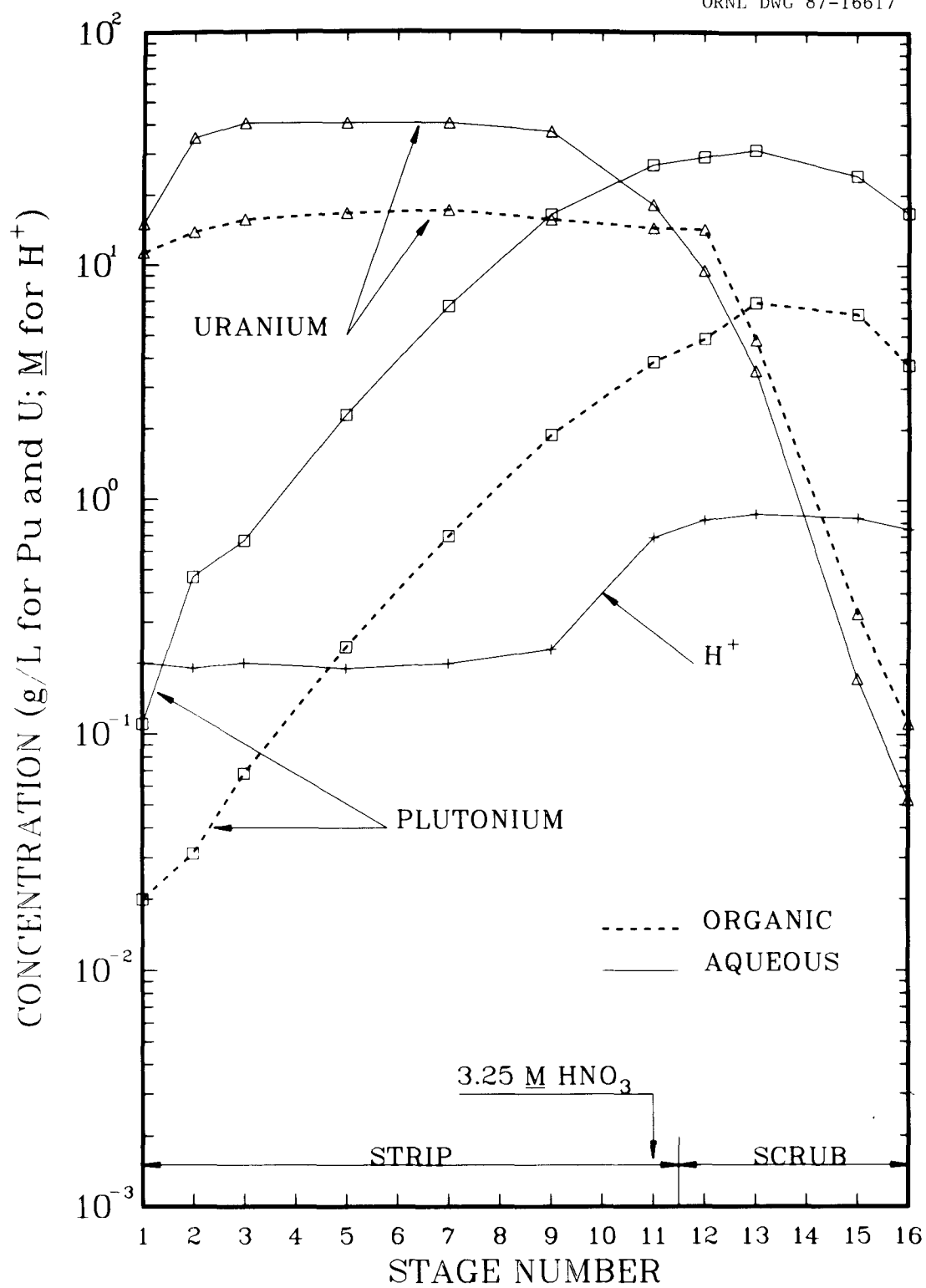


Fig. 7. Concentration profile for uranium, plutonium, and H<sup>+</sup> in the partition contactor for Run 10-2.

## 5. PLUTONIUM PURIFICATION AND OXIDE CONVERSION RESULTS

The aqueous plutonium product solutions that were recovered from the solvent extraction processing were each purified by one cycle of anion exchange and then converted to the oxide form by an oxalate precipitation-calcination step. Table 6 lists the activity levels of the major gamma-emitting isotopes that were measured in the final plutonium oxide product and the overall DF values achieved by the combined processing steps of solvent extraction, anion exchange, and oxalate precipitation, which were made in the high-activity hot cells. The oxide products contained a total of 399 g of plutonium, which represents ~79% of the plutonium originally measured in the dissolver solutions.

Table 6. Radioactivity levels of fission product radionuclides in the plutonium oxide products and the overall DF values achieved

Fission product radionuclide	Radioactivity level in product (MBq/kg Pu)		Overall DF <sup>a</sup>	
	Batch 1	Batch 2	Batch 1	Batch 2
<sup>95</sup> Zr		2		1E5
<sup>106</sup> Ru	46	<2	1E5	>2E6
<sup>125</sup> Sb	6	4	3E5	
<sup>137</sup> Cs	<0.4	1	>7E7	2E7
<sup>144</sup> Ce	<0.6	<3	>5E7	>1E7
<sup>154</sup> Eu	<1	<0.7	>4E5	>1E6

<sup>a</sup>The overall DF is defined as the ratio of the radionuclide concentration (kBq/g Pu basis) in the fuel dissolver solution to its concentration in the PuO<sub>2</sub> product.

## 6. SUMMARY AND CONCLUSIONS

The more significant results and conclusions regarding the solvent extraction flowsheet tests conducted with irradiated FFTF fuel and the in-line photometer system are described below.

The performance of the coextraction-coscrub contactor with 10% TBP in place of 30% TBP has been as good, if not slightly better, with respect to U-Pu losses to the raffinate, fission product DFs, and the physical operation of the mixer-settlers. However, the throughput rate of heavy metals was somewhat lower. An increase in the solvent flow rate by approximately a factor of three is required to compensate for lower solvent capacity with 10% TBP. The SETF process equipment could not entirely handle this increase in flow rate. Similarly, other facilities, which were designed to use 30% TBP, might have to reduce their throughput rates if their solvent transfer, storage, and purification systems cannot handle the higher solvent flow necessary with 10% TBP.

The in-line photometer has continued to yield excellent real-time data on the heavy-metal concentrations in the extraction system. Although the existing out-of-cell electronics were not as reliable as desired, the sampling technique appears to be sound. The computer control system, which uses the data from the in-line photometer (input variable) to determine the appropriate changes in the solvent addition rate (output variable), was able to maintain steady, efficient control of the extraction contactor with no major process upsets during normal operations. However, the length of each test was relatively short because of safeguard restrictions that limited the amount of feed for each run; as a result, a systematic study of the control characteristic of the system was not possible.

Partitioning in 16-stage mixer-settlers without a reductant yielded product purities of at least 99% for both uranium and plutonium (U and Pu DFs in the range of 100 to 1,000). Although previous results using HAN reductant have typically given product purities of 99.99% (DFs of 10,000), the nonreductant system was much simpler. In addition, the design and operation of a large reprocessing plant may be further simplified if the potential safety concerns associated with HAN are eliminated from the flowsheet. As a result, if the product specifications are not too extreme, the nonreductive flowsheet may still be an attractive method to consider.

## 7. 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 under the supervision of J. L. Botts and J. M. Peele. Engineering and maintenance services were provided by personnel from the General Engineering Division, the Instrumentation and Controls Division, and Plant Equipment Division, under the supervision of S. O. Lewis, E. M. Shuford, and B. R. Scarborough. Also, D. D. McCue gave invaluable assistance in the operation of the in-line photometer and computer control system.

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## APPENDIX





Table A-1. Campaign 10 first-cycle tests — extraction scrub bank conditions and results

	Run No.	
	10-1	10-2
Dates	3/19-20/86	4/30-5/1/86
Bank temperature, °C	40-41	40
Number of stages		
Final scrub/ inter. scrub/extraction	5/4/7	5/4/7
HAX stream flow rate, L/h	1.55 <sup>a</sup>	1.76 <sup>b</sup>
Flow ratios		
HAS/HAX	0.0955	0.0875
HAIS/HAX	0.038	0.034
HAF/HAX	0.116	0.105
Inlet stream compositions		
HAS stream, HNO <sub>3</sub> , mol/L	0.49	0.48
HAIS stream, HNO <sub>3</sub> , mol/L	5.05	5.0
HAX stream, % TBP	10±0.1%	10±0.1%
HAF stream		
HNO <sub>3</sub> , mol/L	3.3	3.4
U, g/L	152	168
Pu, g/L	37	45.7
<sup>241</sup> Am, g/L	0.269	0.328
<sup>242</sup> Cm, mg/L	0.20	0.50
<sup>95</sup> Zr, GBq/L		11.5
<sup>95</sup> Nb, GBq/L	0.6	8.59
<sup>106</sup> Ru, GBq/L	177	119
<sup>125</sup> Sb, GBq/L	71	<0.9
<sup>134</sup> Cs, GBq/L	135	321
<sup>137</sup> Cs, GBq/L	979	1210
<sup>144</sup> Ce, GBq/L	1200	1870
<sup>154</sup> Eu, GBq/L	21.0	31.9

Table A-1 (continued)

	Run No.	
	10-1	10-2
Outlet stream compositions		
HAW stream		
HNO <sub>3</sub> , mol/L	2.52	3.39
U, mg/L	11.3	0.11
Pu, mg/L	18.2	8.9
<sup>241</sup> Am, g/L	0.136	0.190
<sup>242</sup> Cm, mg/L	0.095	0.25
<sup>95</sup> Zr, GBq/L		7.84
<sup>95</sup> Nb, GBq/L	<0.3	3.20
<sup>106</sup> Ru, GBq/L	92.7	64.4
<sup>125</sup> Sb, GBq/L	35.8	<5
<sup>134</sup> Cs, GBq/L	70.5	154
<sup>137</sup> Cs, GBq/L	482	595
<sup>144</sup> Ce, GBq/L	590	926
<sup>154</sup> Eu, GBq/L	9.73	19.0
HAP stream		
HNO <sub>3</sub> , mol/L	0.04	
U, g/L	18.8	16.3
Pu, g/L	4.13	4.52
<sup>95</sup> Zr, kBq/L		<10
<sup>95</sup> Nb, kBq/L	<10	24
<sup>106</sup> Ru, kBq/L	740	302
<sup>125</sup> Sb, kBq/L	<10	<10
<sup>134</sup> Cs, kBq/L	<10	<10
<sup>137</sup> Cs, kBq/L	<10	<10
<sup>144</sup> Ce, kBq/L	<10	<30
<sup>154</sup> Eu, kBq/L	<10	<10

<sup>a</sup> Average flow rate; the HAX varied from 1.34 to 1.65 L/h during the run.

<sup>b</sup> Average flow rate; the HAX varied from 1.74 to 1.87 L/h during the run.

Table A-2. Campaign 10 first-cycle test-conditions and results  
for B-bank contactor

	Run No.	
	10-1	10-2
Dates	3/19-20/86	4/30-5/1/86
Bank temperature, °C	8-13	9-12
Number of stages		
Strip/scrub	11/5	11/5
HBX stream flow rate, L/h	0.682	0.401
Flow ratios		
HAP/HBX	2.27 <sup>a</sup>	4.39 <sup>b</sup>
HBIX/HBX	0.103	0.152
HBS/HBX	2.65	2.24
Inlet stream compositions		
HBX stream		
HNO <sub>3</sub> , mol/L	0.20	0.20
HBIX stream		
HNO <sub>3</sub> , mol/L	5.0	3.25
HBS stream, % TBP	10±0.1%	10±0.1%
HAP stream		
HNO <sub>3</sub> , mol/L	0.04	
U, g/L	18.8	16.3
Pu, g/L	4.13	4.52
<sup>95</sup> Zr, kBq/L		<10
<sup>95</sup> Nb, kBq/L	<10	<4
<sup>106</sup> Ru, kBq/L	740	302
<sup>125</sup> Sb, kBq/L	<10	<10
<sup>134</sup> Cs, kBq/L	<10	<10
<sup>137</sup> Cs, kBq/L	<10	<10
<sup>144</sup> Ce, kBq/L	<10	<30
<sup>154</sup> Eu, kBq/L	<10	<10

Table A-2 (continued)

	Run No.	
	10-1	10-2
Outlet stream compositions		
HBP stream		
HNO <sub>3</sub> , mol/L	0.66	0.74
U, g/L	0.0235	0.0406
Pu, g/L	8.69	17.3
<sup>95</sup> Zr, kBq/L		<10
<sup>95</sup> Nb, kBq/L	<20	4
<sup>106</sup> Ru, kBq/L	<100	<40
<sup>125</sup> Sb, kBq/L	<50	<10
<sup>134</sup> Cs, kBq/L	<20	<10
<sup>137</sup> Cs, kBq/L	<20	32
<sup>144</sup> Ce, kBq/L	<200	<70
<sup>154</sup> Eu, kBq/L	<50	<10
HBU stream		
HNO <sub>3</sub> , mol/L		<0.01
U, g/L	8.03	10.8
Pu, mg/L	5.45	20.9
<sup>95</sup> Zr, kBq/L		<10
<sup>95</sup> Nb, kBq/L	<10	10
<sup>106</sup> Ru, kBq/L	353	215
<sup>125</sup> Sb, kBq/L	<10	<10
<sup>134</sup> Cs, kBq/L	<10	<10
<sup>137</sup> Cs, kBq/L	<10	<10
<sup>144</sup> Ce, kBq/L	<20	30
<sup>154</sup> Eu, kBq/L	<10	<10

<sup>a</sup> Average ratio; the HAP flow rate varied from 1.34 to 1.65 during this run.

<sup>b</sup> Average ratio; the HAP flow rate varied from 1.74 to 1.87 during this run.

Table A-3. Campaign 10 first-cycle tests—conditions and results  
for C-bank contactor

	Run No.	
	10-1 <sup>a</sup>	10-2 <sup>b</sup>
Dates	3/19-20/86	4/30-5/1/86
Bank temperature, °C	49	26
Number of stages		
Strip	16	16
HCX stream flow rate, L/h	0.879	0.153
Flow ratios		
HBU/HCX	3.82	17.4
Inlet stream compositions		
HCX stream		
HNO <sub>3</sub> , mol/L	0.04	0.3
HAN, mol/L	0.05	0.9
HBU stream		
HNO <sub>3</sub> , mol/L		<0.01
U, g/L	8.03	10.8
Pu, mg/L	5.45	20.9
<sup>95</sup> Zr, kBq/L		<10
<sup>95</sup> Nb, kBq/L	<10	10
<sup>106</sup> Ru, kBq/L	353	215
<sup>125</sup> Sb, kBq/L	<10	<10
<sup>134</sup> Cs, kBq/L	<10	<10
<sup>137</sup> Cs, kBq/L	<10	<10
<sup>144</sup> Ce, kBq/L	<20	30
<sup>154</sup> Eu, kBq/L	<10	<10

Table A-3 (continued)

	Run No.	
	10-1	10-2
Outlet stream compositions		
HCW stream		
U, g/L	4.5E-5	10.7
Pu, $\mu$ g/L	7.8	<80
$^{95}\text{Zr}$ , MBq/L		<10
$^{95}\text{Nb}$ , MBq/L	<10	<10
$^{106}\text{Ru}$ , MBq/L	239	138
$^{125}\text{Sb}$ , MBq/L	<10	<10
$^{134}\text{Cs}$ , MBq/L	<10	<10
$^{137}\text{Cs}$ , MBq/L	<10	<10
$^{144}\text{Ce}$ , MBq/L	<10	20
$^{154}\text{Eu}$ , MBq/L	<10	<10
HCP stream		
$\text{HNO}_3$ , mol/L	0.07	0.17
U, g/L	29.6	8.5
Pu, g/L	0.0222	0.393
$^{95}\text{Zr}$ , kBq/L		16
$^{95}\text{Nb}$ , kBq/L	<10	24
$^{106}\text{Ru}$ , kBq/L	163	189
$^{125}\text{Sb}$ , kBq/L	<10	<10
$^{134}\text{Cs}$ , kBq/L	<10	18
$^{137}\text{Cs}$ , kBq/L	80	108
$^{144}\text{Ce}$ , kBq/L	100	<40
$^{154}\text{Eu}$ , kBq/L	<10	<10

<sup>a</sup>C-bank was used as uranium strip contactor.

<sup>b</sup>C-bank was used to strip residual plutonium from uranium product.

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