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Solvent Extraction Studies with High-Burnup Fast Flux Test Facility Fuel in the Solvent Extraction Test Facility

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CONSOLIDATED FUEL REPROCESSING PROGRAM

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-9993
Dist. Category UC-86T
(Applied)

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ORNL/TM--9993

TI87 025759

Date Issued — October 1986

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Prepared by the
OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37831
operated by
MARTIN MARIETTA ENERGY SYSTEMS, INC.
for the
U.S. Department of Energy
under
Contract No. DE-AC05-840R21400

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ABSTRACT

A batch of high-burnup fuel from the Fast Flux Test Facility (FFTF) was processed in the Solvent Extraction Test Facility (SETF) during Campaign 9. The fuel had a burnup of ~90 MWd/kg and a cooling time of ~1 year. Two runs were made with this fuel; in the first, the solvent contained 30% tri-n-butyl phosphate (TBP) and partitioning of the uranium and plutonium was effected by reducing the plutonium with hydroxylamine nitrate (HAN); in the second, the solvent contained 10% TBP and a low operating temperature was used in an attempt to partition without reducing the plutonium valence. The plutonium reoxidation problem, which was present in previous runs that used HAN, may have been solved by lowering the temperature and acidity in the partition contactor. An automatic control system was used to maintain high loadings of heavy metals in the coextraction-coscrub contactor in order to increase its efficiency while maintaining low losses of uranium and plutonium to the aqueous raffinate. An in-line photometer system was used to measure the plutonium concentration in an intermediate extraction stage; and based on this data, a computer algorithm determined the appropriate adjustments in the addition rate of the extractant. The control system was successfully demonstrated in a preliminary run with purified uranium. However, a variety of equipment and start up problems prevented an extended demonstration from being accomplished during the runs with the FFTF fuel.

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1. INTRODUCTION

The Solvent Extraction Test Facility is located in one of the heavily shielded hot cells of the Transuranium Processing Plant (TPP) at the Oak Ridge National Laboratory (ORNL).[1] It contains mixer-settler contactors which are used to evaluate solvent extraction flowsheets for the reprocessing of irradiated nuclear reactor fuels. Up to this time, a total of nine experimental campaigns have been completed in the SETF. Results from these tests provide information on heavy metal recoveries, fission product behavior, comparisons of flowsheet options, evaluation of in-line instrumentation, and general operability of the system.

This report describes the work completed in Campaign 9, which is the third SETF campaign in which fuel from the Fast Flux Test Facility was used. In the previous two campaigns,[2,3] fuels were used that had burnups of ~2, ~36, and ~55 MWd/kg and cooling times of 2, 1.3, and 1 year, respectively. In Campaign 9, the fuel had a peak burnup of ~90 MWd/kg and a cooling time of about 1 year (discharged in April 1984). Fuel pin identification numbers are listed in the Appendix.

The processing steps in Campaign 9 included: (1) dissolution of the fuel in nitric acid, (2) clarification of the dissolver solutions by filtration, (3) adjustment of the dissolver solutions to the proper concentrations and plutonium valence for solvent extraction, (4) solvent extraction processing using TBP extractant with partitioning of the uranium and plutonium, (5) purification of the plutonium by nitrate-based anion exchange, and (6) conversion of the plutonium to an oxide form by oxalate precipitation and calcination.

Three solvent extraction experiments were completed; two experiments (Runs 9-2 and 9-3) were made using the irradiated FFTF fuel and one experiment (Run 9-1) was made using unirradiated UO_3 powder as feed. The tests with the irradiated fuel continued the investigation of Purex-type flowsheets that was started in the previous campaigns. The test with the UO_3 was made to demonstrate the process control system that was later used in the runs with the FFTF fuel. The object of the control system was to maximize the loading of heavy metals (uranium and plutonium) in the extraction contactor to increase its efficiency and to improve the decontamination factors (DF) for fission products while still maintaining low losses of heavy metals to the raffinate. The monitored variable was the concentration of plutonium in the organic phase from an intermediate stage in the extraction bank, which was measured using an in-line photometer, and the controlled variable was the addition rate of extractant to the contactor.

2. EQUIPMENT AND OPERATIONAL PROCEDURES

Most of the major equipment items and general operating procedures used in Campaign 9 were similar to those used before and described for previous campaigns. A description of the general layout, equipment and operation of the solvent extraction contactors is given in ref. 1, the fuel dissolution and clarification steps are described in ref. 2, the filtration equipment in refs. 4 and 5, the in-line photometer system in ref. 3, and the plutonium purification and conversion to oxide in refs. 3 and 5. A brief summary of the processing steps is given below.

2.1 FUEL DISSOLUTION

The fuel pins were sheared into ~25-mm-long segments at a separate hot cell facility, the High Radiation Level and Examination Laboratory. The heavy metals in the first batch of ~90 MWd/kg fuel were dissolved by immersing the sheared pins in 7.5 to 8 $\underline{M}$ HNO_3 and heating at 95°C for 4 hours. The volume of acid was sized to make a final solution that was about 350 g/L of heavy metals and 3.5 $\underline{M}$ HNO_3. This procedure was adequate for dissolutions with the ~36 and ~55 MWd/kg fuels; but a significant portion (~10%) of the heavy metals from the ~90 MWd/kg fuel did not dissolve initially and was recovered in a hull leach, in which a larger volume of fresh 8 $\underline{M}$ HNO_3 was used. For the next batch of fuel, the digest time was increased to 6 h and an additional digest was made at 100°C for 2 h; less than 2% of the heavy metals were collected in the subsequent hull leach solution.

2.2 CLARIFICATION AND FEED ADJUSTMENT

The dissolver solution was clarified by pumping it through a deep bed of diatomaceous earth, and the plutonium valence was adjusted by sparging the solution with NO gas while heating at 60°C. After the valence adjustment, the solution acidity and heavy-metal concentrations were adjusted as necessary by addition of the appropriate nitric acid solution.

2.3 SOLVENT EXTRACTION

The solvent extraction contactors consist of three banks of 16-stage mixer-settlers. The settling and mixing chambers hold about 50 and 30 mL, respectively; the total solution flow rate of both organic and aqueous phases during these tests ranged from 1.5 to 3.7 L/h. A water jacket located on the back side of each contactor (adjacent to the mixers) is used to heat or cool the contactor banks to the desired temperatures. However, the inlet lines are not jacketed, so if the temperatures of the inlet solutions are significantly different from that of the contactor there will be a slight temperature variation across the contactor.

The tops of all sixteen settlers are open and accessible using the master-slave manipulators in the hot cell. Samples of either the organic or aqueous phase were taken from any of the settlers by inserting a small dip-tube into the desired phase in any stage and pulling solution into a pre-evacuated bottle; a new dip-tube and bottle was used with each sample.

All reagents for the solvent extraction runs were used on a once-through basis with no recycle of solvent or nitric acid.

2.4 ANION EXCHANGE PURIFICATION AND OXIDE PREPARATION

The plutonium product from the solvent extraction processing was purified by one cycle of nitrate-based anion exchange. The plutonium was then precipitated as Pu(IV) oxalate, filtered, and heated in a furnace to 500°C to make PuO_2.

3. DESCRIPTION OF THE SOLVENT EXTRACTION FLOWSHEETS

The investigation of first-cycle flowsheet options that was begun in Campaigns 7 and 8 was continued. Diagrams and operating conditions for the two types of flowsheets that were used in Campaign 9 with irradiated fuel (Runs 9-2 and 9-3) are shown in Figs. 1 and 2; detailed stream analyses for each run are tabulated in the Appendix.

The layout of the flowsheets, which are similar to the conventional Purex-type arrangement, included (1) coextraction and coscrubbing of the

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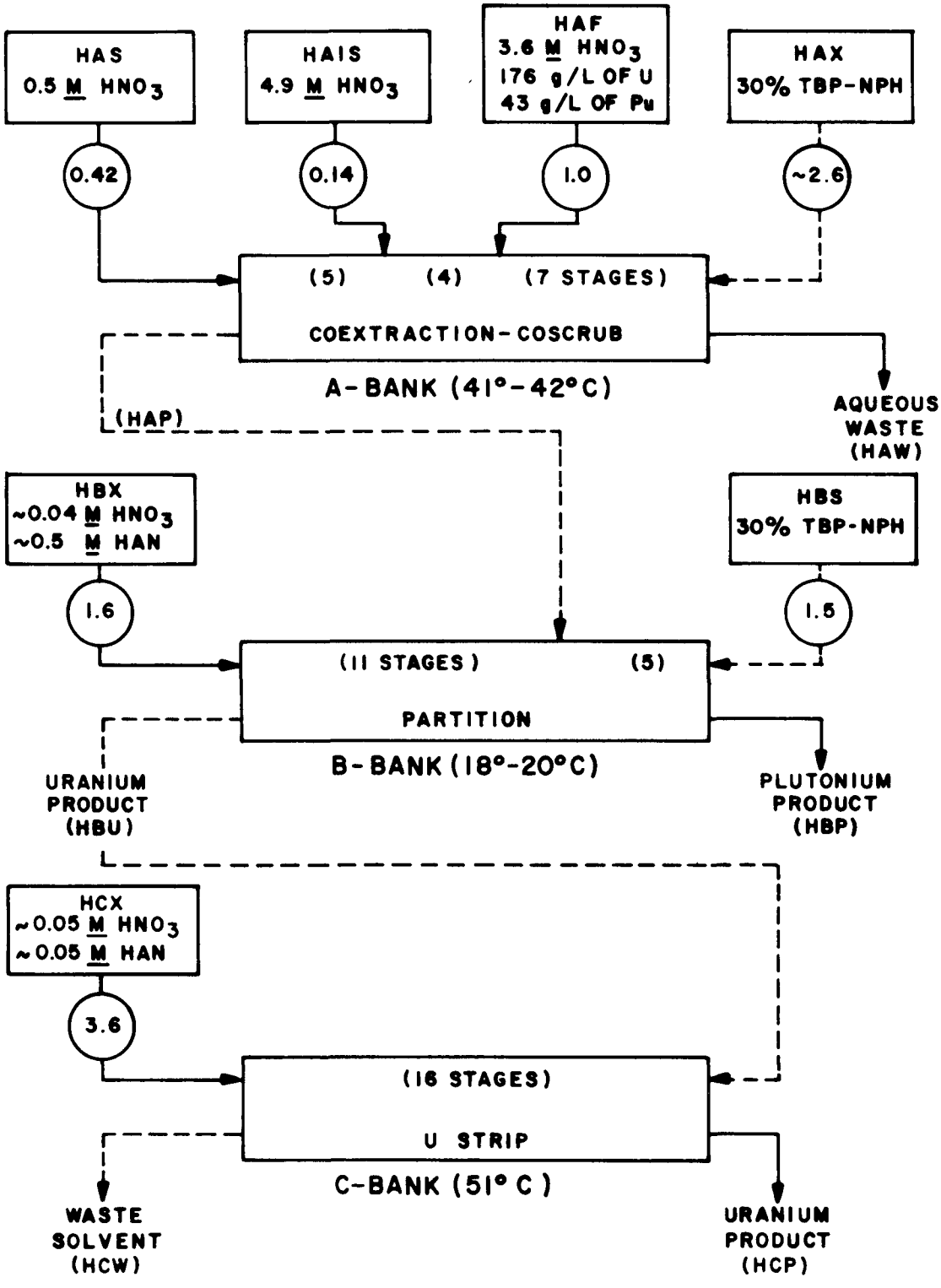

Fig. 1. Solvent extraction flowsheet using the organic backscrub method of partitioning with a chemical reductant (Run 9-2).

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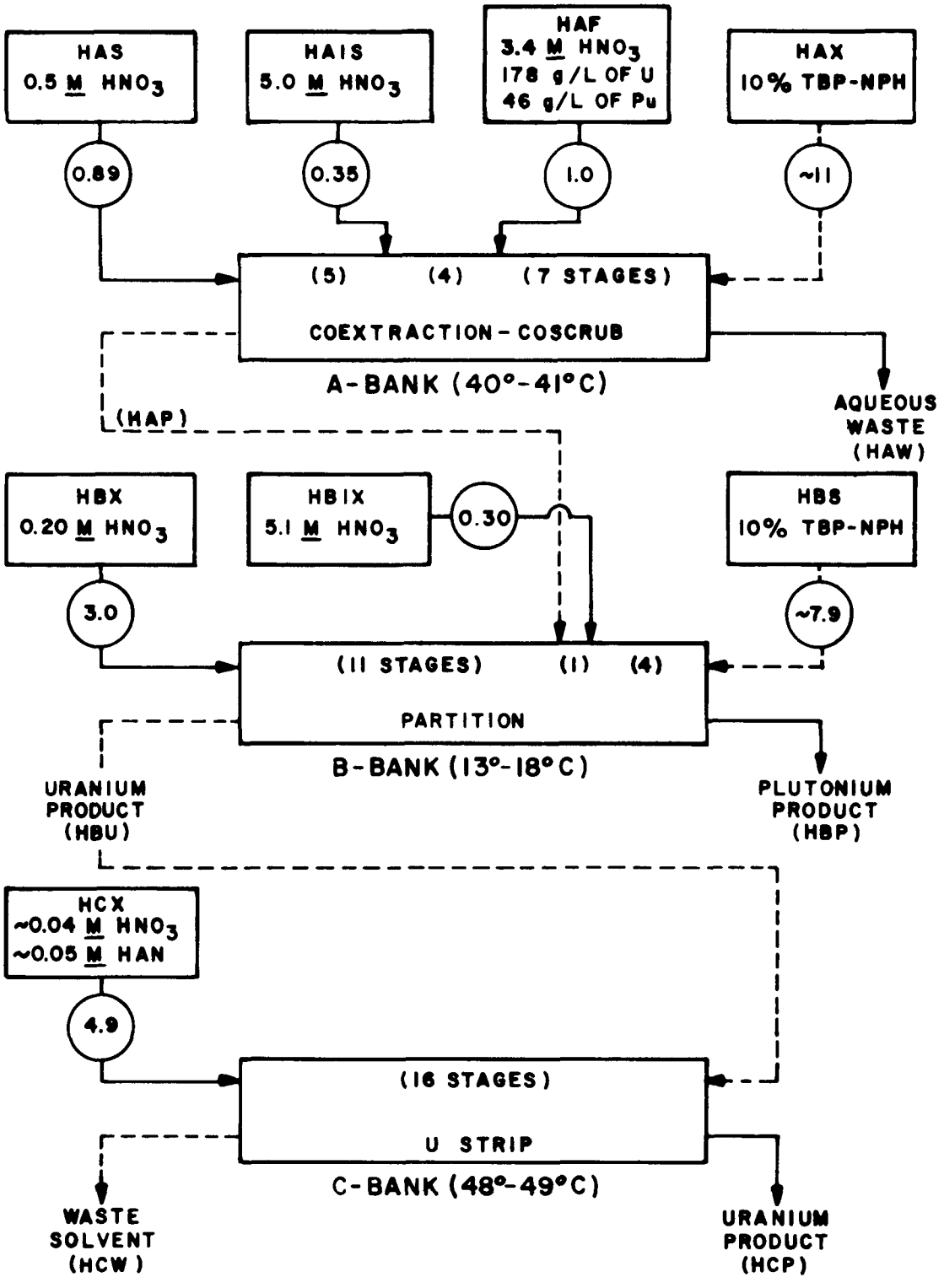

Fig. 2. Solvent extraction flowsheet using the organic backscrub method of partitioning with no chemical reductant (Run 9-3).

heavy metals in A-bank, (2) U-Pu partitioning in B-bank, and (3) stripping of the uranium from the solvent in C-bank. Tables 1 and 2 list the major conditions that have been changed for each of the runs with FFTF fuel (detailed descriptions of Campaigns 7 and 8 are given in refs. 2 and 3).

Table 1. Conditions for the coextraction-coscrub contactor (A-Bank)

Run No.	Fuel burnup (MWd/kg)	No. of stages[a]	Solvent saturation[b] (%)	TBP concentration[c] (%)
7-1	2	3/3/10	~60	30
7-2	2	4/4/8	~65	30
8-1	~36	4/5/7	~50	30
8-3	~55	4/5/7	~90	30
9-2	~90	4/5/7	~90	30
9-3	~90	4/5/7	~80	10

[a]Low-acid scrub/high-acid scrub/extraction stages

[b]Peak saturation.

[c]The diluent for each run was normal paraffin hydrocarbon.

Table 2. Conditions using the organic backscrub method for the total partitioning of U-Pu

Run No.	Strip solution (mol/L)			Temp. (°C)	Stages[a]	Phase ratio (O/A)	
	HNO_3	HAN	N_2H_4			Strip	Scrub
7-1	0.1	0.3	none	40	11/5	2.6	0.80
7-3B	0.1	0.5	none	40	11/5	3.0	0.99
8-1	0.1	0.6	0.1	40	11/5	3.0	0.97
9-2	0.04	0.5	none	18-20	11/5	2.6	0.96
9-3	0.2	none	none	13-18	11/5	6.2	2.4

[a]Strip stages/scrub stages.

The major changes for the first run in Campaign 9 with irradiated fuel (Run 9-2) included using the higher burnup fuel, and a lower operating temperature and strip acidity for the partition bank (B-Bank); A-bank conditions were unchanged. This run was to test automatic control of the extraction bank (see Sect. 4.1, below) and determine if the above changes to the partition bank would mitigate the plutonium reoxidation that has occurred in previous runs.

In the final run with FFTF fuel (Run 9-3), the TBP concentration was lowered from 30 to 10 vol %, the partition bank was operated at a lower operating temperature, and no reductants were added to reduce Pu(IV) to Pu(III). This run measured the effect of using 10 vol % TBP on the fission product decontamination factors (DFs) and U-Pu losses in the A-bank, but the primary objective was to test how well uranium and plutonium could be separated without reducing plutonium.

In most of the previous SETF runs, chemical reductants [hydroxylamine nitrate (HAN), or hydroxylamine nitrate with hydrazine] have been used to change the plutonium valence from Pu(IV) to the relatively inextractable Pu(III) during the partitioning of uranium-plutonium. This reduction step greatly increases the uranium-plutonium separation factor* and makes the partitioning easier; but this step also complicates the flowsheet by requiring additional processing steps to remove excess reductants and to readjust the plutonium valence for the next purification cycle. Furthermore, some of the reductants are highly reactive and may require additional safety controls when used in a processing plant.

If no reductant is used, the separation factor is lower, of course; but some partitioning can still be achieved since the uranium and plutonium extraction coefficients are not identical. The British have measured uranium and plutonium extraction coefficients at various temperatures and

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*Separation factor is defined here as the ratio of uranium extraction coefficient to the plutonium extraction coefficient. The extraction coefficient is defined as the ratio of the concentration of a component in the organic phase (in units of g/L) divided by the concentration in the aqueous phase. The separation factor would then have the following form, $(U_{org}/U_{aq})(Pu_{org}/Pu_{aq})$.

TBP concentrations, and they have suggested that the U(VI)-Pu(IV) separation factor may be high enough to achieve adequate uranium and plutonium separation in a single contactor if the operating temperature and TBP concentration are kept sufficiently low.[6] Run 9-3 was made at a relatively low temperature and TBP concentration (though not as low as the British have considered) in order to measure the amount of U(VI)-Pu(IV) separation that could be achieved in the SETF mixer-settlers and determine if there are any significant operational problems when running at these conditions (e.g., plutonium third phase formation, plutonium polymer formation, poor phase separation, etc.). If the results were encouraging and no problems were encountered, then even lower temperatures and TBP concentrations might be considered.

4. EXPERIMENTAL RESULTS AND DISCUSSIONS OF SOLVENT EXTRACTION TESTS

The FFTF fuel processed in Campaign 9 had a higher burnup than any previous fuel used in the SETF. The concentrations of the major gamma-emitting fission products that were measured in the dissolver solutions for Campaigns 7, 8, and 9 are listed in Table 3. The fission product decontamination factors that are shown later (Sect. 4.2) are based on the dissolver solution values and not on the total activity in the fuel pins. This distinction primarily affects fission products, such as ruthenium, that are relatively insoluble in the high-burnup fuels.

No significant problems were noted with respect to solution pumping, phase separation, formation of solids, or gassing while processing this high-burnup fuel in the SETF mixer-settlers or while using the different TBP concentrations (30 and 10 vol %). However, the length of each test was relatively short (13 to 24 h); thus, any effect that requires a longer initiation period would not have been detected.

4.1 CONTROL OF U-Pu LOADING IN THE COEXTRACTION BANK

The in-line photometer system[3] was used again to help achieve high loadings of uranium and plutonium in the coextraction-coscrub bank (A-Bank) while still maintaining low losses to the aqueous raffinate stream. The

Table 3. Comparison of fission product concentrations of the fast-reactor dissolver solutions processed in the SETF

Radionuclide	Activity [GBq/kg (U + Pu)]			
	Runs 7-1 & 7-2[a]	Run 8-1[b]	Run 8-3[c]	Runs 9-2 & 9-3[d]
^{95}Zr	2	550	3,050	4,600
^{95}Nb	3	601	2,090	6,900
^{106}Ru	180	1,300	1,870	1,100
^{137}Cs	160	2,400	5,030	8,100
^{141}Ce			52	180
^{144}Ce	480	12,400	31,200	50,000
^{154}Eu	0.7	34	150	390
^{155}Eu		390	1,010	1,300

[a]DEA-1 fuel, 2.2 [MWd/kg] (0.2 TJ/kg) burnup and ~2-year cooled.
[b]DE-1-6 fuel, ~36 [MWd/kg] (3.1 TJ/kg) burnup and ~1.3-year cooled.
[c]DE-2-1R fuel, ~55 [MWd/kg] (4.8 TJ/kg) burnup and ~1-year cooled.
[d]DE-4 fuel, ~90 [MWd/kg] (7.8 TJ/kg) burnup and ~1-year cooled.

photometer system measured the plutonium concentration (uranium during Run 9-1) in the organic phase from an intermediate stage in the extraction section where the concentration profiles for the heavy metals were changing rapidly between stages. Based on this measurement, the addition rate of the organic extractant (HAX) was varied to maintain the plutonium (or uranium) concentration at a value that would achieve the desired loadings while also maintaining low aqueous losses. In the previous campaign, these changes in the HAX addition rate were made manually by the operators; in Campaign 9 they were made automatically by a computer-controlled solution addition system. The control algorithm was:[7]

$$M_n = M[1 + K_p E_k + K_d(1.5E_k - E_{k-1} - 0.5E_{k-2})] ,$$

where

M_n = new set point value for HAX flow controller,
M = old set point value for HAX flow controller,
K_p = proportional control constant (0.0005 for Runs 9-1, 9-2, and 9-3),

K_d = derivative control constant (0.005 for Runs 9-1 and the beginning of Run 9-2, 0.01 for the latter part of Run 9-2, and 0.0075 for Run 9-3,

E_k = difference between measured and desired plutonium concentrations,

E_{k-1} = E_k from one minute ago, and

E_{k-2} = E_k from two minutes ago.

The value of M_n was calculated every minute but changes >1% were not allowed. In Runs 9-1 and 9-2, the photometer system monitored the solvent from Stage 13 (the third stage from the HAW exit); in Run 9-3, which used 10% TBP, Stage 12 was sampled.

Most control systems that combine proportional and derivative control modes would also include integral control, to prevent offset of the monitored variable from the desired set point. However, this extraction-scrub system was operated at near saturated conditions; and consequently, minor flowsheet variations could lead to major adjustments in the heavy metal inventory in the extraction section before the outlet stream concentrations from the contactor would be significantly affected. Since the sample point was located in the area where the heavy metal inventory adjustments would be taking place, the absolute value selected for the set point was not critical, nor was a minor offset from the set point. As a result, integral control was not included in order to simplify the development and testing of the control algorithm.

4.1.1 Test with Uranium (Run 9-1)

Run 9-1 was made with uranium specifically to provide a simple test of the control algorithm. Figure 3 shows a plot of the photometer reading (monitored variable) and the HAX flow rate (controlled variable) vs elapsed time. After the computer control system was started, the uranium concentration in Stage 13 was quickly brought near the set point value (35 g/L), and then it was maintained at 34 to 39 g/L over the next 16 h. The overall performance of the system was very good. Results from six sets of samples taken during this time from (1) the organic product stream (HAP), (2) the solvent at the feed stage, and (3) the aqueous raffinate (HAW) varied from 84.9 to 86.2, 110 to 121, and 0.0002 to 0.0003 g/L of uranium, respectively.

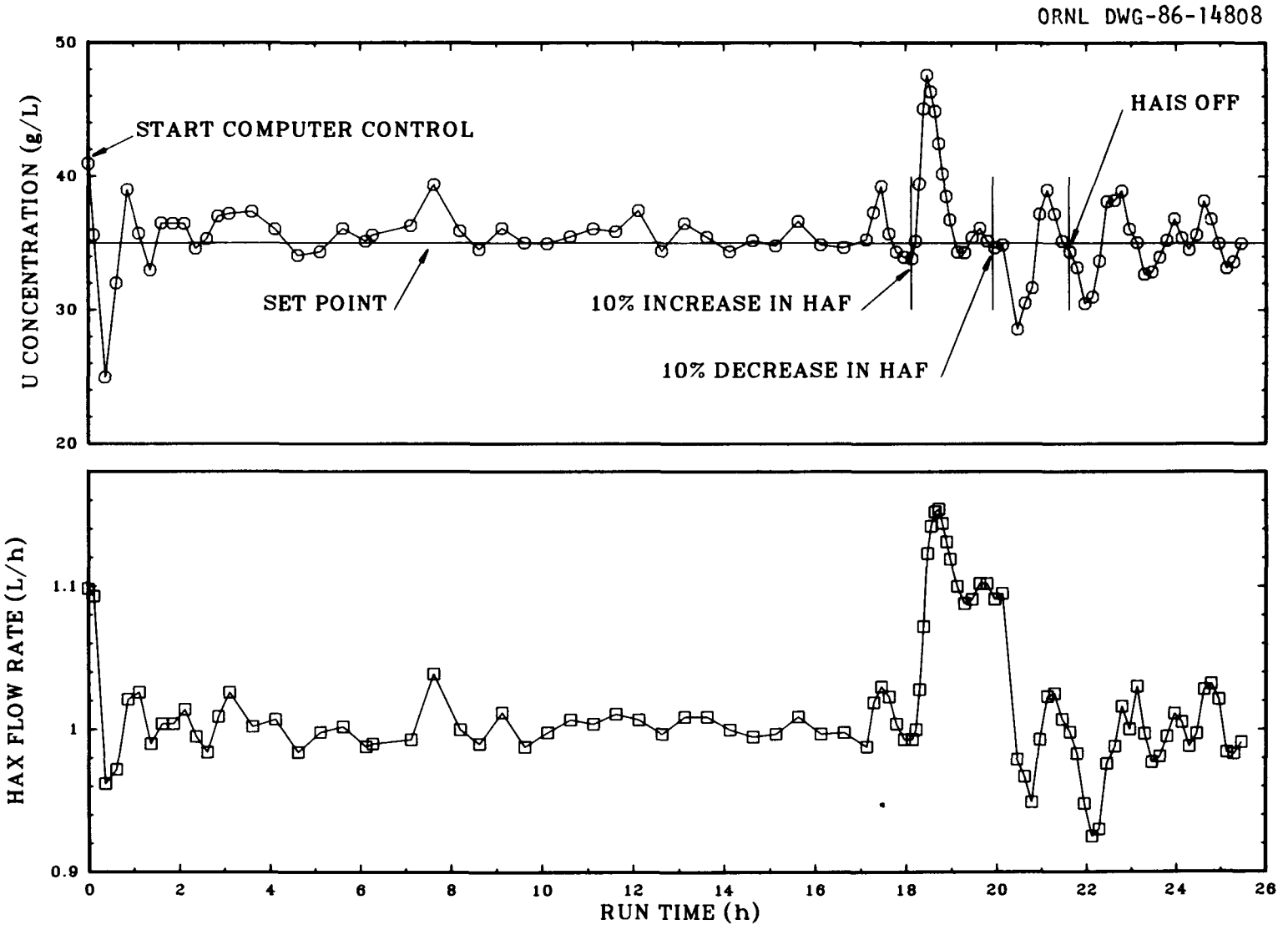

Fig. 3. Concentration readings from the in-line photometer (Stage 13) and the addition rate of the extractant (HAX) for Run 9-1.

After the 18-h point, several perturbations were made to test the ability of the system to respond to upsets. These changes were: (1) 10% increase in the feed rate (HAF), (2) 10% decrease in the HAF back to the original rate, and (3) stopping the HAIS scrub flow to simulate a pump failure. The control system responded properly to each change, but insufficient time was available to determine if the control system would have remained stable after these changes.

4.1.2 Tests with FFTF Fuel

4.1.2.1 Run 9-2

In this run, the plutonium concentration in Stage 13 was the monitored value and the set point was 8 g/L of plutonium. Figure 4 shows a plot of the photometer reading and the HAX flow rate vs run time. During start up of the contactor, the HAX flow rate was manually set at 50 to 60% of the design flow rate (~1 L/h) in order to more quickly bring the contactor to steady state conditions. When plutonium was first detected in the sample loop (Stage 13), the HAX was set to ~80% of the design flow rate and the computer control system was activated. In hindsight this was a mistake because the plutonium inventory was still increasing in the contactor at this time and the control algorithm immediately started decreasing the HAX which was already set at a low value (80% of the design flow rate). This situation led to the large overshoot at the 4-h point. The manual adjustment at 4.1-h was made to help the system recover more quickly.

Then, near the 8-h point we discovered that the feed inlet line had been dislodged from its proper entry position. When this displacement occurred was unknown; but while the wand was out of position, it effectively reduced the number of feed stages and increased the number of scrub stages by two. When the feed inlet was repositioned, a large upset occurred in the 8- to 10-h time period. Also during this upset, a change was made in the K_d coefficient in the control algorithm (from 0.005 to 0.01) at the ~9.5-h point. After the change, unexplained problems with the computer required that the system be kept on manual control for ~15 min until the computer could be restarted.

No further problems were noted after the 11-h point, but unfortunately the HAF solution was consumed at the ~12.8-h point before an extended

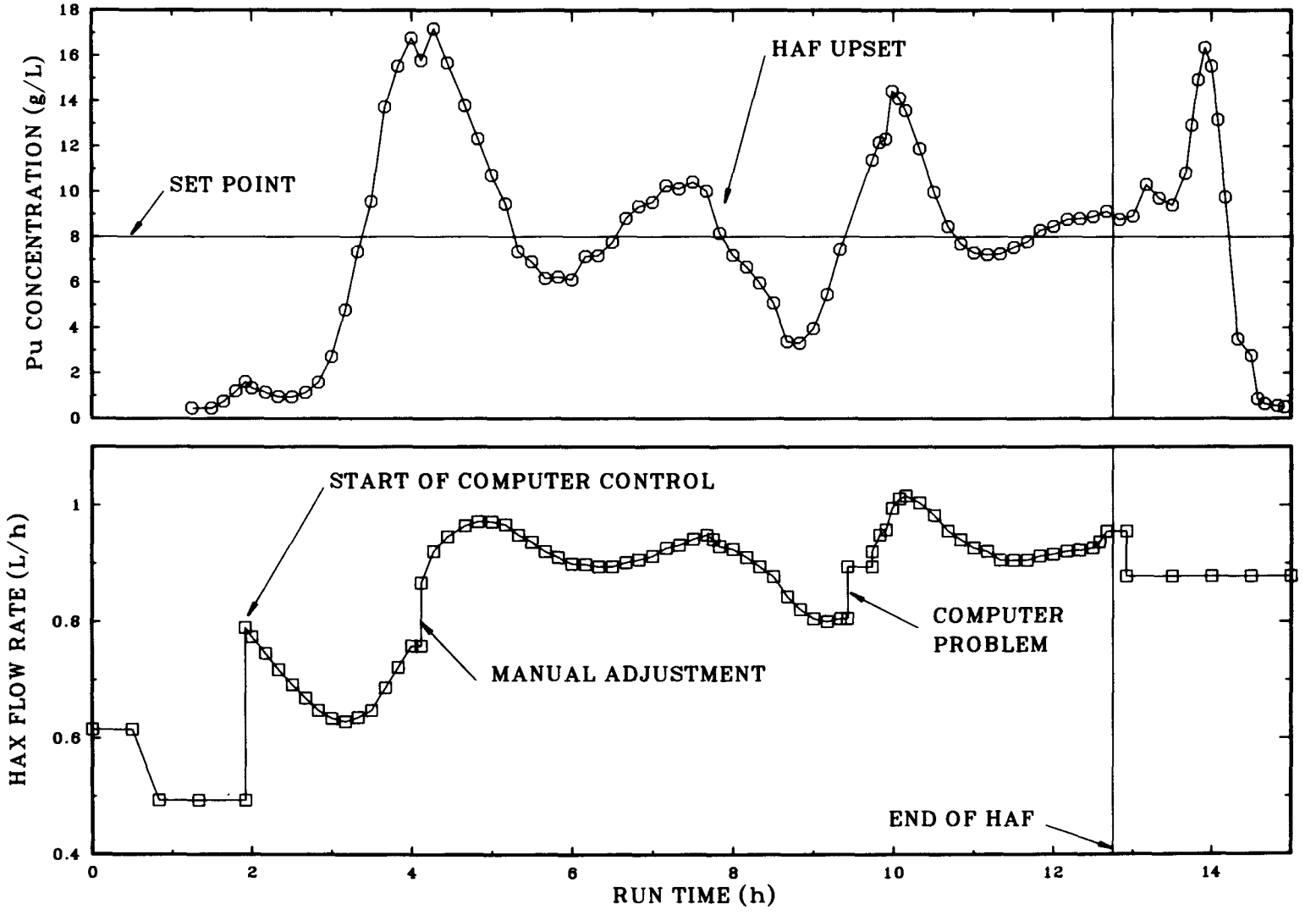

Fig. 4. Concentration readings from the in-line photometer (Stage 13) and the addition rate of the extractant (HAX) for Run 9-2.

demonstration on automatic control could be accomplished. In spite of the problems, the control system appeared to be working properly during most of the run and the variations shown in Fig. 4 for the control stage were not large enough to cause significant fluctuations in the outlet stream concentrations. However, it is difficult to determine the long-term stability of the control system from these results.

4.1.2.2 Run 9-3

In this run the extractant was 10% TBP, and the sampling line to the automatic control system was switched from Stage 13 to Stage 12 (one stage towards the feed inlet) because of the lower heavy-metal concentrations with the lower TBP concentrations. Figure 5 shows a plot of the photometer reading and the HAX flow rate vs run time. To avoid the previous mistake of starting the computer control too early, the system was kept on manual control until the sample stage contained over 8 g/L of plutonium (the set point value). Unfortunately, this concentration was closer to solvent saturation than was expected; and the transition zone for the concentration profile quickly moved past the sample stage towards the raffinate exit, which left the sample stage saturated with heavy metals. In this condition, the heavy metal concentration in the sample stage will change very slowly with changes in the HAX flow rate, unless the transition zone is moved back to this stage. This concentration-time behavior obviously prevented the control system from working properly. Manual adjustments were made to help lower the heavy-metal inventory in the extraction section and bring the transition region back to the sample stage. In addition, the set point was lowered to 6 g/L in order to give a larger control range before reaching saturation (~9 to 10 g/L).

At the 18-h point, the sampling system developed an unknown problem that caused wide fluctuations in the plutonium (and also the uranium) readings. Within 5-min readings the plutonium values varied by as much as ±30%. Apparently the average of these fluctuations was close enough to the actual plutonium concentrations and the control system was sufficiently damped to prevent the contactor from quickly going out of control during the last 6 h of the run. However, it is doubtful that proper control would have been possible over the long term with this type of sampling problem.

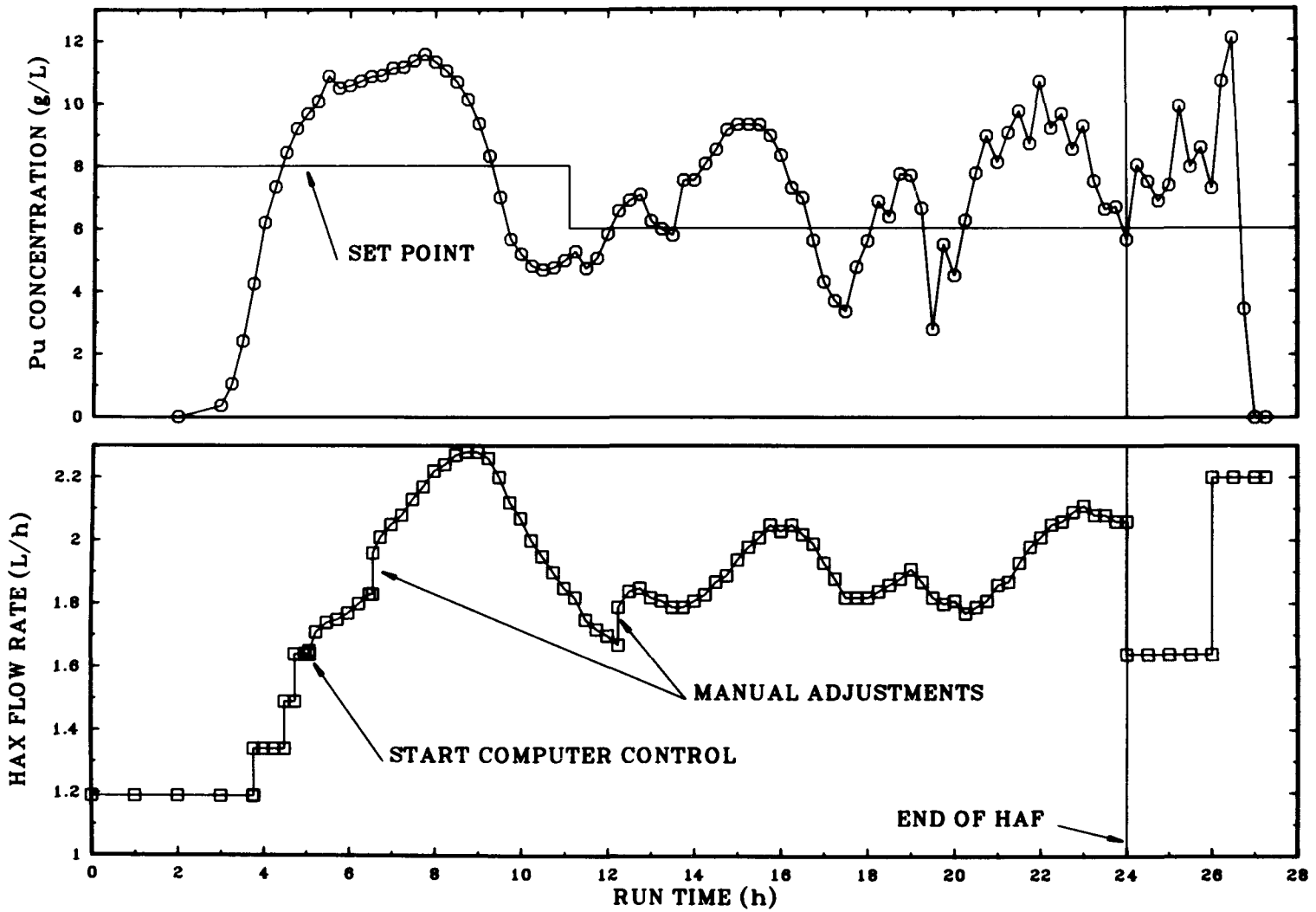

Fig. 5. Concentration readings from the in-line photometer (Stage 12) and the addition rate of the extractant (HAX) for Run 9-3.

4.2 RESULTS FROM THE COEXTRACTION-COSCRUB CONTACTOR

The measured uranium and plutonium losses to the aqueous raffinates (HAW) for Campaign 9 and for the previous runs with FFTF fuel are listed in Table 4. Although the solvent loadings were significantly increased for Runs 8-3, 9-2 and 9-3 (Table 1), and the TBP concentration lowered to 10% in Run 9-3, the raffinate losses of uranium and plutonium have remained low. (The relatively high uranium loss indicated for Run 9-2 may be a sampling or an analysis problem, because it is inconsistent with the plutonium loss and is significantly greater than the amount of uranium measured in the collection tank for the aqueous waste.)

The concentration profiles of uranium and plutonium for Runs 9-2 and 9-3 are shown in Fig. 6. The lower TBP concentration in Run 9-3 led to much lower concentrations of heavy metals in the solvent; however, the degree of solvent saturation (i.e. the fraction of the TBP in the solvent that is complexed with heavy metals) was only slightly lower (~80 vs ~90%). The lower TBP concentration also led to slightly higher Pu/(U+Pu) ratios near the addition point of the low-acid scrub stream (Stage 5). The Pu/(U+Pu) ratio changed from ~0.40 to ~0.50 in the aqueous phase, and from ~0.20 to ~0.27 in the organic phase for Runs 9-2 and 9-3, respectively.

Table 4. Uranium and plutonium losses and fission product decontamination results in coextraction–coscrubbing (A-Bank)

	Run number					
	7-1	7-2	8-1	8-3	9-2	9-3
Extraction losses						
Uranium, %	0.002	0.003	<0.001	0.008	0.12	0.003
Plutonium, %	0.011	0.012	0.02	0.02	0.005	0.05
Fission product DFs						
^{95}Zr	9E1	7E2	5E3	2E4	3E4	2E5
^{95}Nb	2E2	4E3	9E3	7E3	2E4	1E5
^{106}Ru	6E2	3E3	7E3	2E4	3E4	7E4
^{134}Cs					>1E7	>8E6
^{137}Cs	>4E5	>7E5	3E5	>6E6	>5E6	>2E7
^{141}Ce				2E4	>1E5	2E4
^{144}Ce	>1E5	>2E5	>4E5	>7E6	>6E6	>1E7
^{154}Eu	>7E4	>1E4	>2E3	>3E5	>4E5	>7E5
^{155}Eu			>2E4	>4E5	>2E5	>5E5

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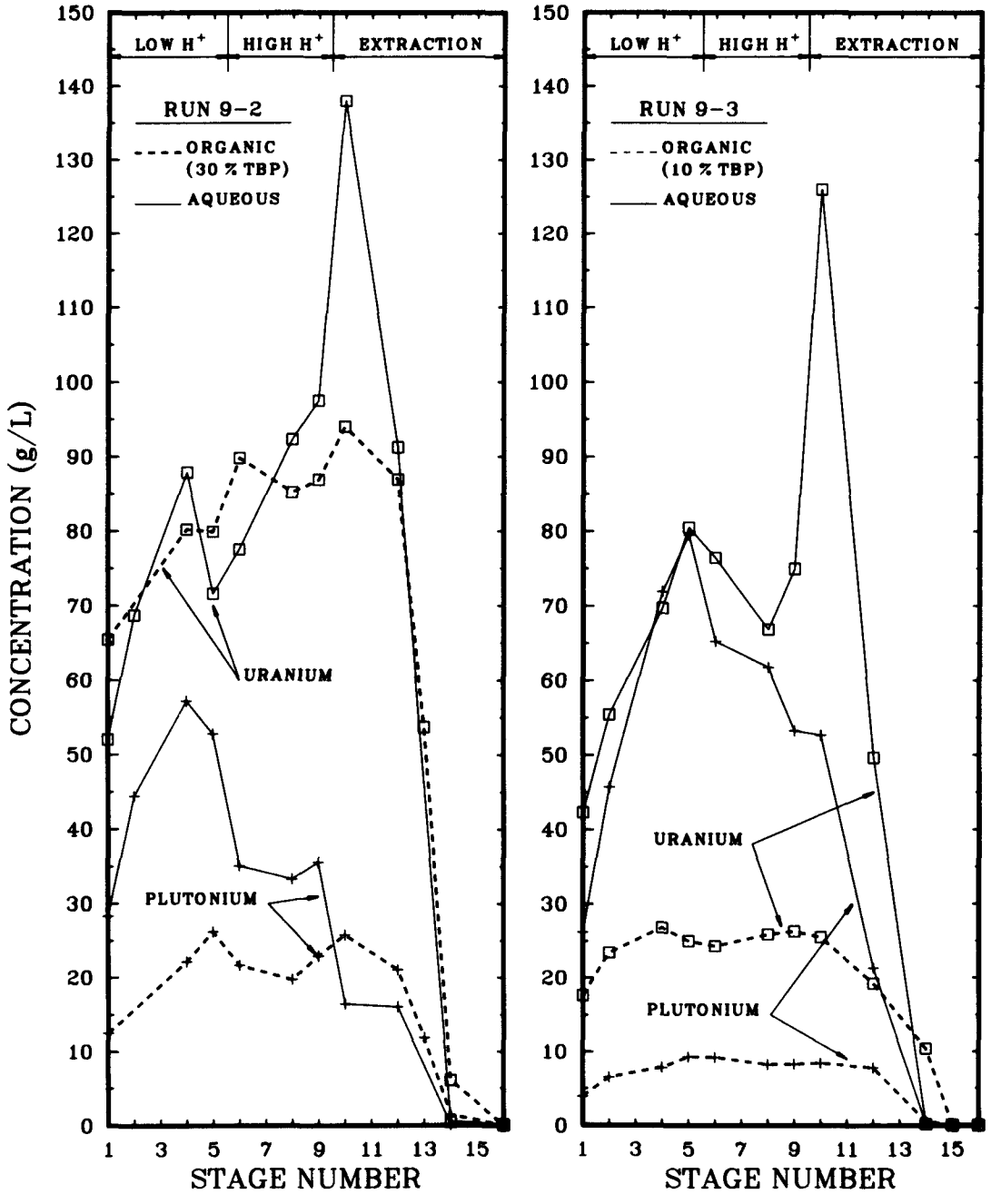

Fig. 6. Concentration profiles for uranium and plutonium in the coextraction-coscrub contactor (A-Bank) for Runs 9-2 and 9-3.

Concentration profiles for ^{95}Zr, ^{95}Nb, and ^{106}Ru, in both the organic and aqueous phases, are shown in Figs. 7, 8, and 9, respectively. In Runs 8-3 (see ref. 3) and 9-2, which used 30% TBP with high solvent loadings, the fission product activity in the organic phase reached a minimum value in the first few scrub stages and very little activity was removed in the remaining stages. Although Run 9-2 had more activity in the feed, because of the higher burnup of the fuel, the activity in the scrub section solvent was about the same magnitude as for Run 8-3. As a result, Run 9-2 showed slightly higher DFs (Table 4) even though the HAP product had similar concentrations of fission products. Apparently, this residual activity is associated with solvent degradation products (solids, chemical complexes, or something similar) that has a different stripping behavior.

Run 9-3, which used 10% TBP, also showed poor removal of fission products in the scrub section; however, the activity level in the solvent was usually at least a factor of ten lower than for the two runs with 30% TBP. This result may indicate that the more dilute TBP solvent was less efficient at collecting (or making) the fission product activity that was difficult to scrub.

4.3 RESULTS FROM PARTITIONING CONTACTOR (B-BANK)

4.3.1 With Reductant (Run 9-2)

In each of the previous SETF campaigns in which partitioning in the first cycle was effected by using the organic backscrub method (Runs 7-1, 7-3B, and 8-1), hydroxylamine nitrate (HAN) was used to reduce plutonium. Although good results have been obtained in each run, a relatively large excess of HAN (HAN/Pu mol ratio of ~4) has been required. The excess was apparently needed to overcome plutonium reoxidation in the scrub section of the contactor. Adding hydrazine in the previous run (8-1) improved the plutonium profile in the scrub section, but excessive consumption of HAN was still evident. Because of the plutonium reoxidation, there was some concern whether this method would remain stable over longer periods than those used in these tests (about 16 h), and whether this method could recover from major upsets.

ORNL DWG-86-14812

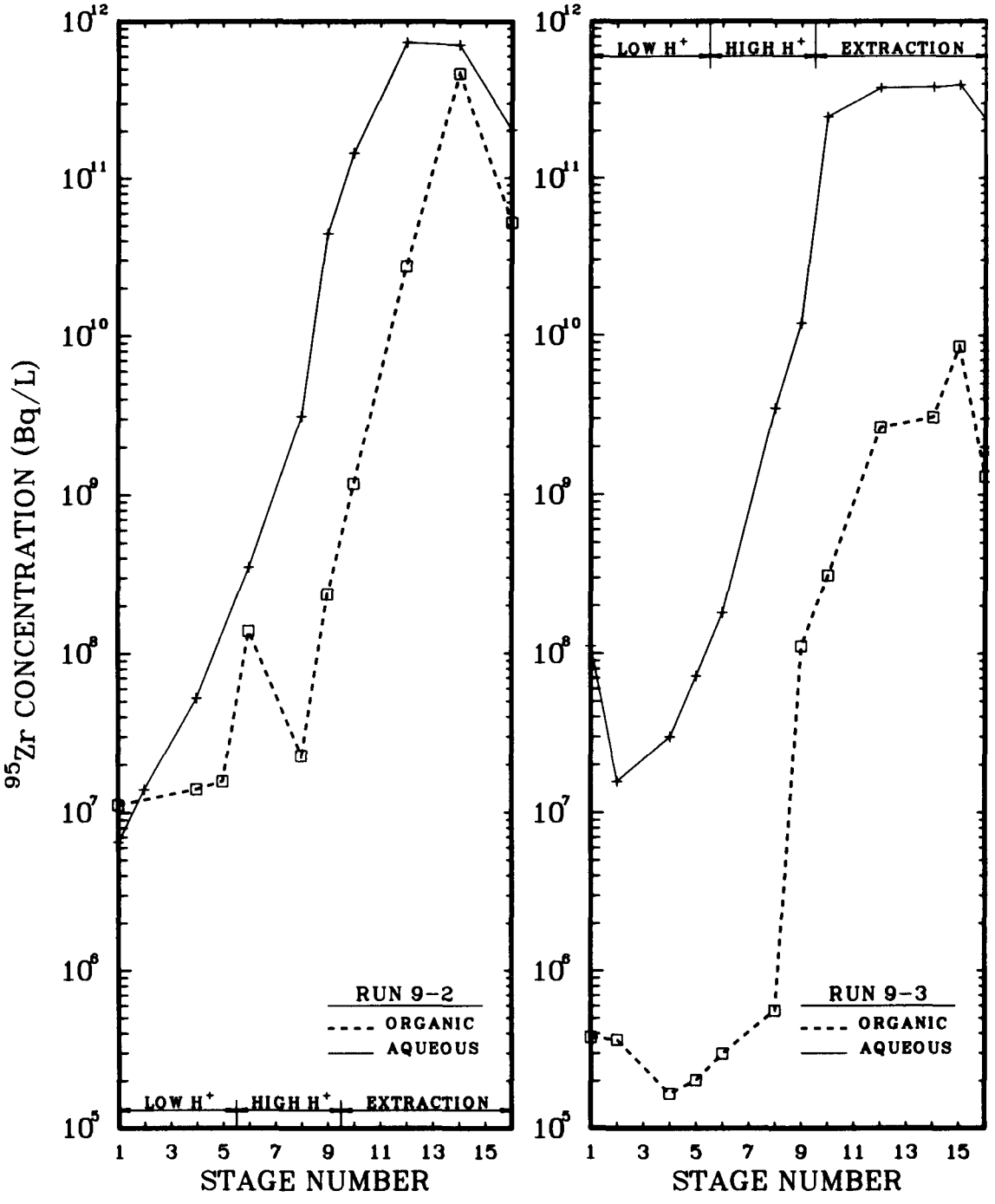

Fig. 7. Concentration profiles for ^{95}Zr in the coextraction-coscrub contactor for Runs 9-2 and 9-3.

ORNL DWG-86-14813

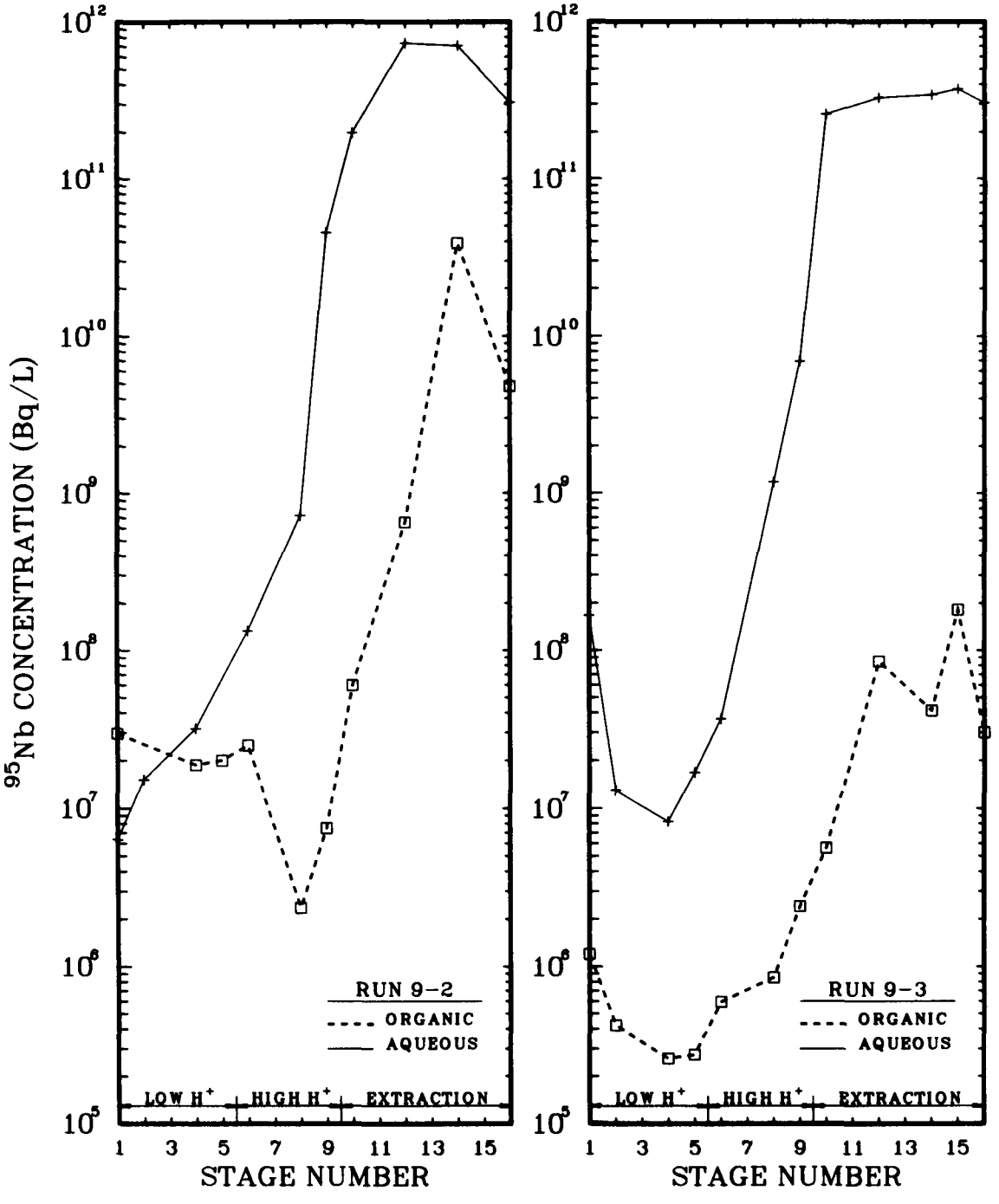

Fig. 8. Concentration profiles for ^{95}Nb in the coextraction-coscrub contactor for Runs 9-2 and 9-3.

ORNL DWG-86-14814

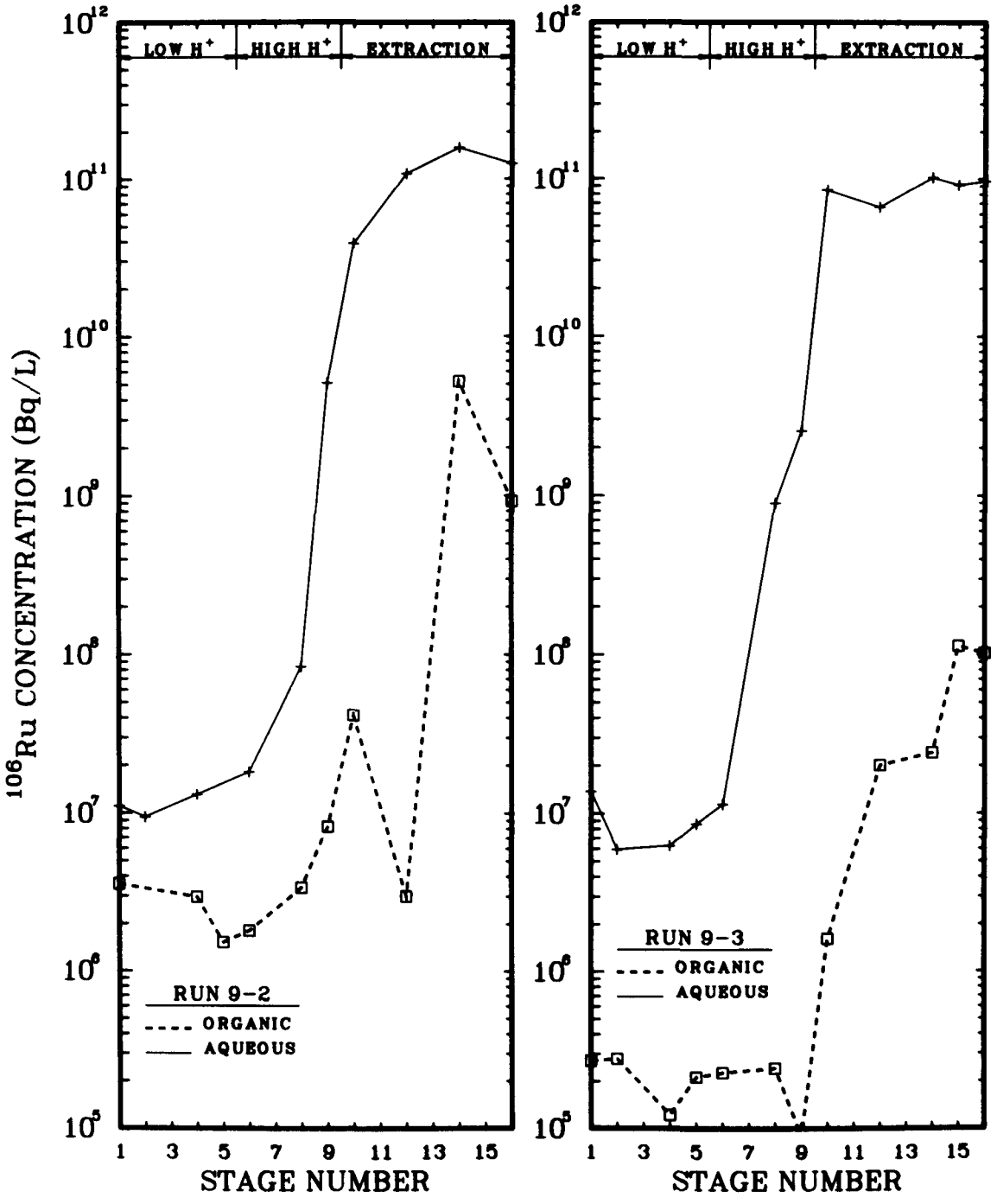

Fig. 9. Concentration profiles for ^{106}Ru in the coextraction-coscrub contactor for Runs 9-2 and 9-3.

In Campaign 9, two changes were made in an attempt to lessen the reoxidation, (1) the operating temperature of the partition contactor was lowered from 40 to ~20°C, and (2) the acidity of the aqueous strip (HBX) was lowered from 0.1 to 0.04 M HNO_3.

Previous SETF runs were made at 40°C to better optimize the reaction kinetics of the HAN reduction; unfortunately, this temperature also increases the rate of Pu(III) oxidation in the solvent.[8] Based on the results from the previous runs, the relatively long residence times in the SETF mixer-settlers coupled with the 40°C operating temperature apparently makes the Pu(III) reoxidation a more serious problem than the HAN reduction. Therefore, the temperature was lowered in Run 9-2 to help provide a better balance between HAN reduction and Pu(III) reoxidation. An additional benefit of the lower temperature was that the uranium was relatively more extractable and therefore easier to separate from the plutonium.

Lowering the acid concentration in the partition contactor is another way of minimizing Pu(III) reoxidation;[9] and, in contrast to the temperature change, the lower acidity also significantly improves the HAN reduction since the reaction rate is inversely proportional to the fourth power of the acid concentration.[10] Aqueous strips of less than 0.1 M nitric acid had not been used before because of a concern for the possibility of forming plutonium polymer. However, a costripping flowsheet used in Runs 7-2 and 8-3 with a 0.02 M acid strip yielded no apparent polymer problems (the HAN reaction with plutonium generates the additional acid to prevent polymerization). Based on this experience, the ~0.04 M nitric acid strip was considered safe for use in Run 9-2.

The results from Run 9-2 were quite good, not only did the run yield satisfactory product solutions (Table 5) of similar quality to the previous runs, but relatively little plutonium reoxidation was indicated in the contactor.

Figures 10 and 11 show the concentration profiles for uranium and plutonium in the three runs. The plutonium extraction coefficients (ratio of the plutonium concentration in the organic phase to the aqueous phase) in the scrub section for Runs 7-3B, 8-1, and 9-2 were, 0.2 to 0.4, 0.1 to 0.2, and 0.03 to 0.08, respectively. Because the aqueous concentrations were reasonably constant in the scrub sections, the varying extraction

ORNL DWG 86-14815

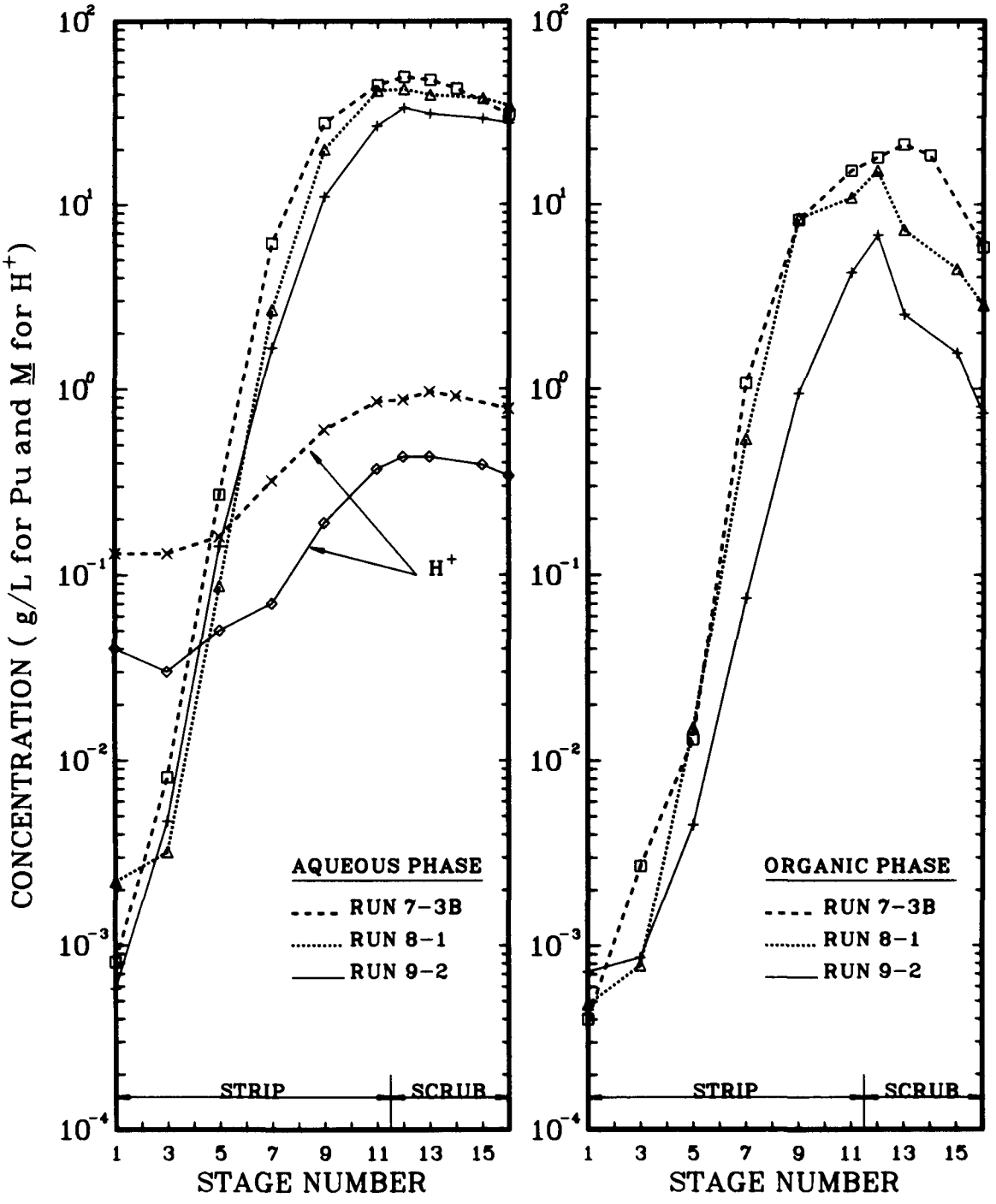

Fig. 10. Comparison of the concentration profiles for plutonium in the partition contactor for Runs 7-3B, 8-1, and 9-2.

ORNL DWG 86-14816

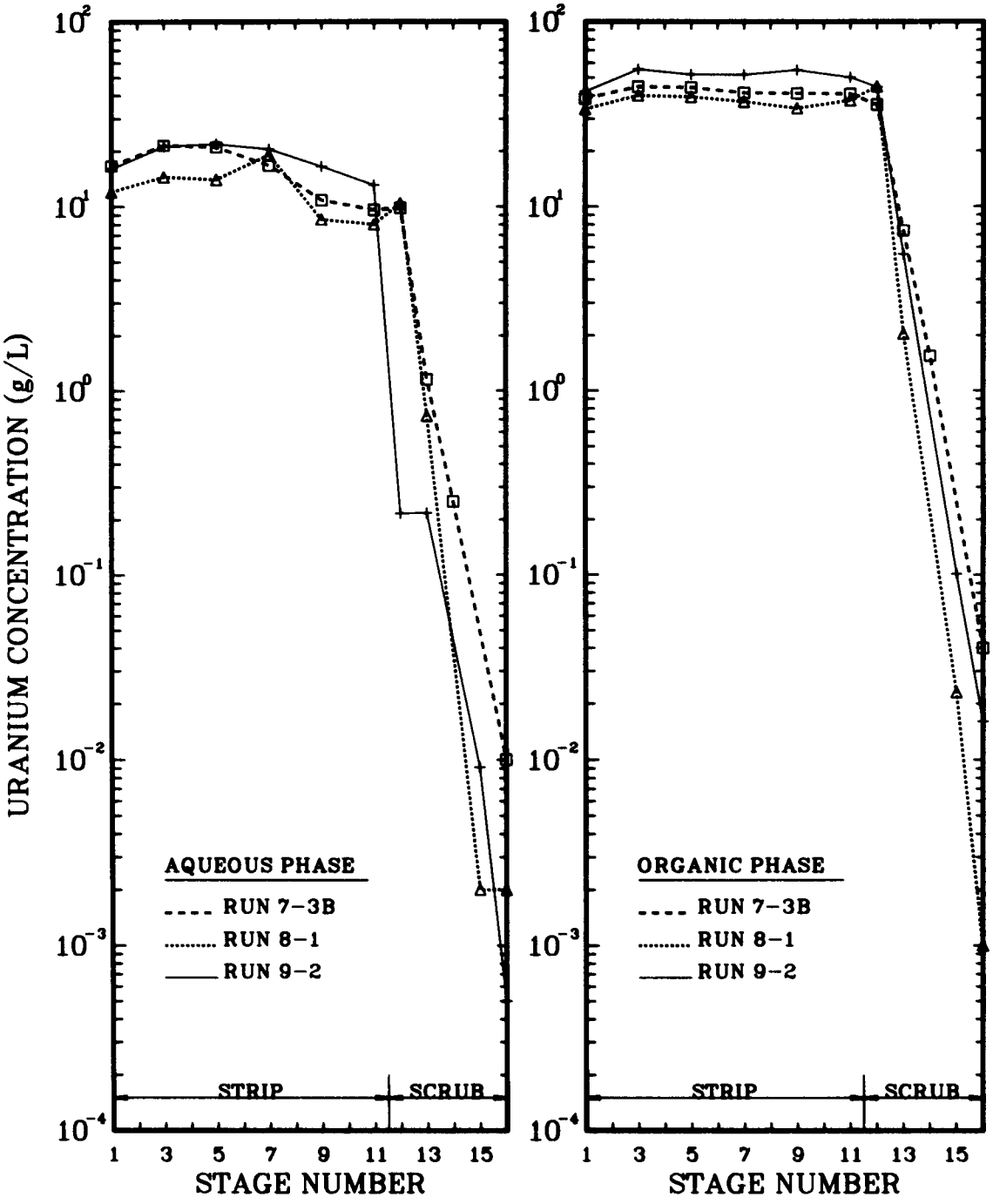

Fig. 11. Comparison of the concentration profiles for uranium in the partition contactor for Runs 7-3B, 8-1, and 9-2.

Table 5. Results of tests with total partitioning

Stream	Run number			
	7-3B[a]	8-1	9-2	9-3
Feed solution (HAF)				
Pu, g/g U	0.268	0.328	0.243	0.255
Pu aqueous product (HBP)				
Pu, g/L	30.3	33.4	27	14
U, μg/g Pu	180	<60	33	2000 to 1.6E4
U DF	2E4	>5E4	1E5	1900 to 240
U organic product (HBU)				
U, g/L	39	34	42	9.4
Pu, μg/g U	23	15	13	3300 to 370
Pu DF	1E4	2E4	2E4	80 to 680

[a]Second cycle run using 7-2 Pu product plus depleted uranium.

coefficients for the three runs are probably the result of the scrub sections containing differing amounts of the more extractable Pu(IV). Furthermore, the higher the extraction coefficients, the more Pu(IV) that was probably present and the more reoxidation that was occurring. The low extraction coefficients for Run 9-2 are close to the values expected for Pu(III) and so are an indication that very little Pu(IV) was present and that little reoxidation was occurring in that run. The addition of hydrazine for Run 8-1 was useful in lessening the plutonium reoxidation that was noted during Run 7-3B (as indicated by the lower extraction coefficients), but that addition was not as effective as lowering the acid concentration and the temperature for Run 9-2. Even though hydrazine is able to scavenge nitrous acid from the aqueous phase [which probably helps lessen reoxidation since nitrous acid is a catalyst for Pu(III) reoxidation], it apparently is unable to completely stop the reoxidation reaction from occurring.

In addition to lowering the extraction coefficients, the changes for 9-2 also significantly lowered acid production in the partition bank.

Table 6 shows an overall acid balance and the amount of free acid that was produced in the partition contactor during these runs. Because the accepted stoichiometry has only 2 mol of H^+ produced per mol of Pu(IV) reduced,[10] the relatively large output of acid in the previous runs was considered an indication that a vicious circle of reduction-reoxidation-reduction was taking place, and consequently consuming excessive amounts of HAN. (This may also explain why the HAN-to-plutonium mol ratio had to be increased from ~2 for Runs 7-1 and 7-3A to ~4 for Run 7-3B in order to achieve good plutonium recoveries.) The lowering of the acid production is another indication that plutonium reoxidation (and excessive HAN consumption) was less a problem after the changes were made for Run 9-2.

Table 6. Acid balance and estimated production (mol ratios) in partition contactor

Run number	Input		Output	Increase
	HAN/N_2H_4/Pu	H^+/Pu	H^+/Pu	H^+/Pu
7-3B	4.1/--/1	1.64	6.48	4.8
8-1	4.3/0.7/1	1.53	6.60	5.0
9-2	4.4/--/1	0.87	3.68	2.8

Overall, the results from this run have provided a higher degree of confidence in the ability of this type of flowsheet to successfully partition breeder fuels using HAN than the previous runs did. In addition, Run 9-2 has demonstrated that hydrazine is unnecessary when partitioning with HAN.

4.3.2 Without Reductant (Run 9-3)

The uranium, plutonium, and acid concentration profiles for Run 9-3 are shown in Fig. 12. The measured separation factors for uranium and plutonium were ~4, which was lower than expected, based on the British U-Pu(IV) extraction data.[6] As a result, the plutonium loss to the solvent

ORNL DWG 86-14817

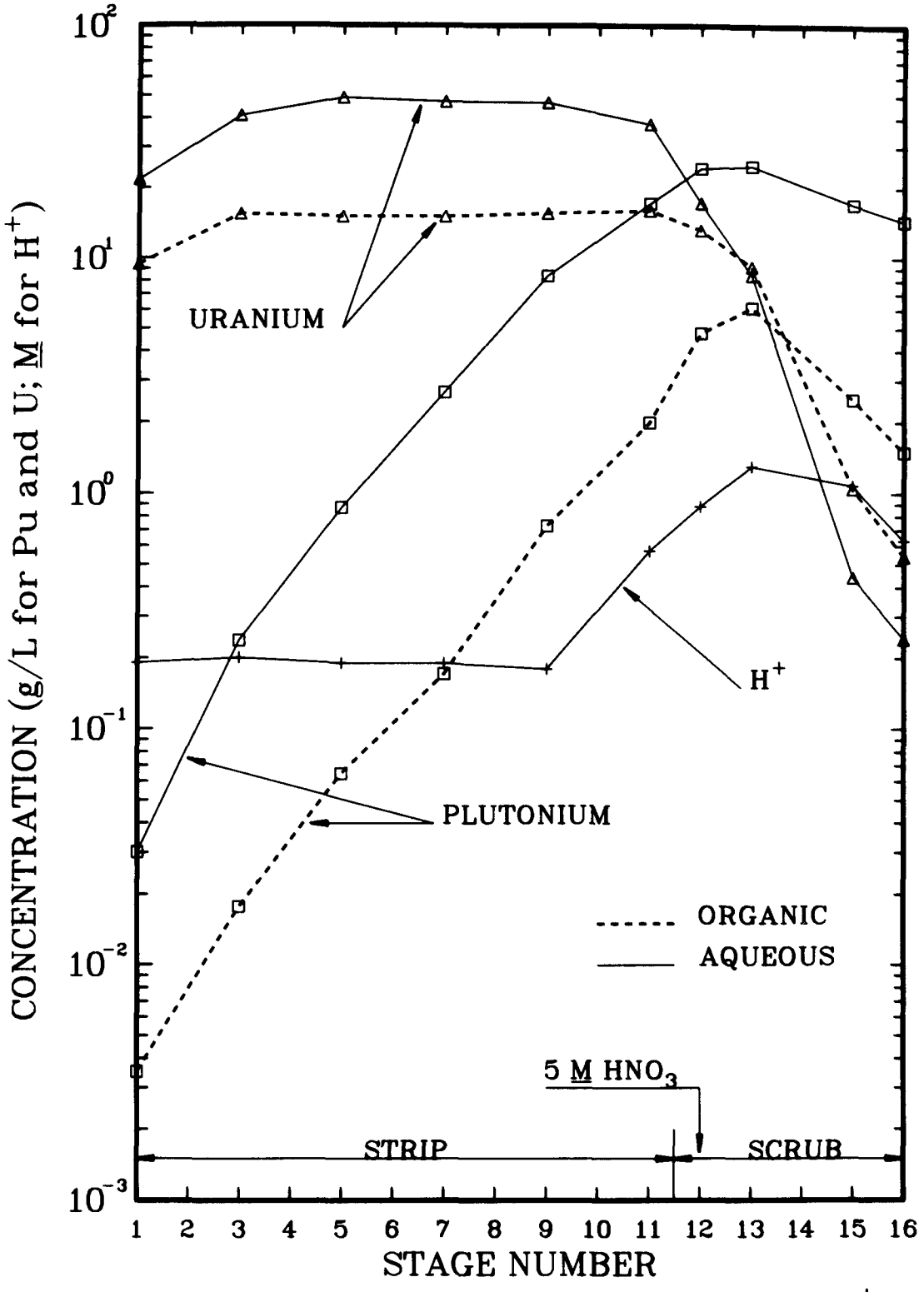

Fig. 12. Concentration profile for uranium, plutonium, and H^+ in the the partition contactor for Run 9-3.

(HBU) was unexpectedly high when the run was first started, ~3000 μg of Pu/g of U (Table 5). To correct for this, the addition rate of the organic backscrub stream (HBS) was lowered by ~20% and consequently the results at the end of the run were more acceptable, ~400 μg of Pu/g of U in the uranium product.

The lowering of the backscrub addition rate naturally allowed more uranium to be carried along with the plutonium product. At the final conditions, the heavy metals in the plutonium product contained about 1.6% uranium. However, this value is not considered excessive because a plutonium product from a reprocessing plant would eventually have to be mixed with uranium in order to recycle it back to another reactor. Even more uranium probably could be allowed into the plutonium before the uranium mass would have a serious impact on the size and throughput of the plutonium purification equipment.

Overall, the results from this test were encouraging. A reasonable separation was achieved and no operational problems were encountered at the lower (13 to 18°C) temperatures and TBP (10%) concentrations.

4.4 RESULTS FROM THE URANIUM STRIP CONTACTOR

Table 7 shows the uranium and plutonium contents in the inlet and effluent streams for the uranium strip contactor (C-Bank). As expected, any plutonium left in the solvent from the partition bank was stripped and collected in the aqueous uranium product. As a result, the plutonium content in the waste solvent streams were quite low, ranging from 3 to 40 μg/L. The uranium content in the waste solvent was typically <1 mg/L, which is close to the detection limit for the uranium analyses.

4.5 FISSION PRODUCT BEHAVIOR IN PARTITION AND STRIP CONTACTORS

The material balances for ^{95}Zr, ^{95}Nb, and ^{106}Ru tended to be poor for the partition and the strip contactors, ranging from 50 to 300% recovery from a contactor for Runs 8-1, 9-2, and 9-3. This situation may have been caused by solids in the solutions, which would have made taking representative samples difficult, or by cross-contamination with the feed or raffinate samples, which had a ratio of $\sim 10^7$ times as much activity and which were handled in the same analytical hot cells and laboratories.

Table 7. Results from uranium strip contactor

	Run number		
	8-1	9-2	9-3
Feed (HBU)			
U, g/L	34	42	9.4
Pu, mg/L	0.5	0.5	3.5
Uranium product (HCP)			
U, g/L	37	48	38
Pu, mg/L	0.4	0.2	17
Waste solvent (HCW)			
U, mg/L	<1	0.4	<1
Pu, µg/L	40	10	2.9

The relative distribution instead of DFs of these fission products are shown in Table 8 for the three outlet streams — plutonium product (HBP), uranium product (HCP), and waste solvent (HCW). The bulk of the ruthenium tended to remain in the waste solvent stream; apparently it formed a strong complex in the solvent that was not easily removed during uranium or plutonium stripping. On the other hand, the zirconium and niobium tended to distribute more equally over all three outlet streams (with the exception of Run 8-1), and the amounts measured in each stream varied significantly in the three runs. This lack of consistency may indicate that zirconium and niobium were in an insoluble form that may either be hard to sample or may have tended to collect at the interface and was randomly carried in either phase. (The high recovery of zirconium in the HBP indicated for Run 8-1 may, in part, be the result of a sample contamination problem; the HBP had about three times as much zirconium as was measured in the HAP).

Table 9 lists the overall fission product DFs as calculated from the feed (HAF) and the final product streams (HBP for plutonium and HCP for uranium). These numbers represent the total fission product purification acheived in the first cycle from the combined process steps of coextraction, coscrubbing, and stripping.

Table 8. Distribution (%) of ^{95}Zr, ^{95}Nb, and ^{106}Ru in the outlet streams from the strip banks

Fission product nuclides	Run number		
	8-1[a]	9-2[a]	9-3[b]
^{95}Zr			
Pu product (HBP)	97	28	53
U product (HBU)	0.8	41	19
Waste solvent (HCW)	2.6	31	28
^{95}Nb			
Pu product (HBP)	40	15	14
U product (HBU)	21	58	42
Waste solvent (HCW)	39	27	44
^{106}Ru			
Pu product (HBP)	16	<11	<15
U product (HBU)	7	16	22
Waste solvent (HCW)	77	73	63

[a]Solvent was 30% TBP.
[b]Solvent was 10% TBP.

Table 9. Fission product decontamination factors for the final plutonium and uranium product streams

Fission product nuclide	Run number		
	8-1	9-2	9-3
DFs for plutonium			
^{95}Zr	2E3	2E5	2E5
^{95}Nb	1E4	3E5	9E5
^{106}Ru	2E4	>4E5	>8E5
^{137}Cs	7E5	2E7	8E6
^{144}Ce	>4E5	>2E7	1E7
^{154}Eu	>3E3	>1E6	>2E6
DFs for uranium			
^{95}Zr	2E5	2E5	7E5
^{95}Nb	2E4	8E4	3E5
^{106}Ru	5E4	3E5	5E5
^{137}Cs	1E7	7E6	1E7
^{144}Ce	6E6	>3E7	>5E7
^{154}Eu	8E4	>1E6	>2E6

5. RESULTS OF PLUTONIUM PURIFICATION AND CONVERSION TO OXIDE

The aqueous plutonium product solutions recovered from 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 10 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. Because all of the equipment for these process steps was located in the same high-activity hot cells, relatively little additional fission product purification was measured after the initial solvent extraction step. Any further purification achieved apparently was lost by recontamination from the hot cell equipment. The oxide products contained a total of 355 g of plutonium, which represents ~82% of the plutonium originally measured in the dissolver solutions.

Table 10. Radioactivity levels from 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[a]	
	Batch 1[b]	Batch 2[c]	Batch 1	Batch 2
^{95}Zr	170	107	2E5	1E5
^{95}Nb	340	340	1E5	5E4
^{106}Ru	110	160	6E4	3E4
^{125}Sb	<20	<3	>3E5	>1E6
^{137}Cs	<6	<1	>7E6	>4E7
^{144}Ce	<40	<9	>8E6	>3E7
^{154}Eu	<10	<0.7	>2E5	>3E6

[a]The overall DF is defined as the ratio of the radionuclide concentration (MBq/kg Pu basis) in the fuel dissolver solution to its concentration in the PuO_2 product.

[b]Includes only plutonium from Run 9-2.

[c]Includes only plutonium from Run 9-3.

6. SUMMARY AND CONCLUSIONS

The most significant results and conclusions regarding the tests conducted with high-burnup irradiated FFTF fuel and the in-line photometer system are as follows:

1. The use of the high-burnup fuel or the lower TBP concentrations and temperatures did not adversely affect the operation of the mixer-settlers. The physical operation was good with regard to solution pumping, phase separations, or solid accumulations and not noticeably different from previous runs. However, the duration of each test was relatively short (13 to 24 h); so any problems that require a longer time to develop would not have been detected in our tests.

2. The computer control system for maintaining high loadings of heavy metals in the coextraction-coscrub contactor was successfully demonstrated in a test using uranium only. The control system kept the uranium loadings high (>110 g/L) while maintaining low losses (~0.3 mg/L) to the raffinate during a 16-h test period. In addition, the system continued satisfactory control during ±10% upset in the feed rate. The tests with the FFTF fuel were plagued with a series of equipment and start up problems that prevented an extended test on automatic control from being accomplished. However, the control system appeared to be working properly, and a successful demonstration might have been achieved if safeguards restrictions had not limited the amount of feed material that could be used for the tests.

3. The higher fuel burnup and the lower TBP concentrations did not adversly affect the uranium and plutonium losses to the aqueous raffinate from the coextraction-coscrub contactor, which remained acceptably low and similar to the results from previous runs.

4. With high loadings of heavy metals in the coextraction–coscrub contactor, the fission product concentrations reached minimum levels that nitric acid scrubbing could not remove. The nature of this residual activity is unknown but could be associated with such things as, solvent degradation products, solids, or chemical complexes.

Although the run with 10% TBP showed the same fission product leveling in the scrub section, the activity concentrations were a factor of ~10 lower than in the runs with 30% TBP, which may indicate that this residual activity does not collect as easily in the more dilute TBP solutions.

5. The plutonium reoxidation problem that was noticed in previous partitioning runs using HAN or HAN with hydrazine, was not evident in this campaign which used a lower temperature and acid concentration for the partitioning contactor. The use of hydrazine for stabilizing Pu(III) is not necessary and may not be adequate when high plutonium concentrations are present.

6. The partial partitioning of uranium and plutonium without using a plutonium reductant yielded acceptable products. Because the amount of U-Pu separation is limited when no reductant is used, the goal of this type of flowsheet was to have very low plutonium concentrations in the uranium product but allow some of the uranium to strip with the plutonium, yielding a product that might contain as much as a few per cent uranium. The complete separation of uranium from the plutonium is not considered necessary, since the plutonium will probably be mixed with uranium before it is recycled to another reactor.

7. The dissolution of the ~90 MWd/kg fuel required a longer dissolution period and a higher temperature than was necessary for the ~36 and ~55 MWd/kg fuels.

7. ACKNOWLEDGMENTS

The experimental work 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, and D. O. Campbell and D. J. Crouse assisted in the selection of flowsheet conditions.

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APPENDIX

Table A-1. Campaign 9 — fuel pin identification numbers

Run 9-2	Run 9-3
N2C 236	N2D 120
N2C 259	N2D 330
N2C 563	N2D 353
N2C 577	N2E 004
N2C 615	N2E 436
N2C 699	N2E 764
N2C 741	N2E 225[a]
N2C 990	N2N 167[a]

[a]The center sections of these pins (~20% of the fuel in the pin) were retained by the Chemical Development Section for fuel characterization studies.

Table A-2. Campaign 9 first-cycle tests — coextraction/coscrub (A-Bank) conditions and results

	Run number		
	9-1	9-2	9-3
Dates	3/21-22/85	3/27/85	5/8-9/85
Bank temperature, °C	40-41	41-42	40-41
Number of stages:			
final scrub/intermediate scrub/extraction	5/4/7	5/4/7	5/4/7
HAX stream flow rate, L/h	1.03	0.92[a]	1.8[b]
Flow ratios:			
HAS/HAX	0.147	0.166	0.083
HAIS/HAX	0.046	0.053	0.033
HAF/HAX	0.34	0.39	0.093
Inlet stream compositions:			
HAS stream, HNO_3, mol/L	0.49	0.50	0.49
HAIS stream, HNO_3, mol/L	5.0	4.9	5.0
HAX stream, % TBP	30.0±0.5	30.0±0.5	10.0±0.5
HAF stream			
HNO_3, mol/L	3.4	3.6	3.4
U, g/L	270	176	178
Pu, g/L		42.8	45.5
^{241}Am, g/L		0.32	0.37
^{242}Cm, mg/L		6.7	6.3
^{95}Zr, GBq/L		1080	642
^{95}Nb, GBq/L		1600	773
^{106}Ru, GBq/L		287	219
^{125}Sb, GBq/L		207	192
^{134}Cs, GBq/L		846	943
^{137}Cs, GBq/L		1720	1980
^{141}Ce, GBq/L		46.4	14.7
^{144}Ce, GBq/L		11200	10500
^{154}Eu, GBq/L		93.3	83.5
^{155}Eu, GBq/L		272	293

Table A-2 (continued)

	Run number		
	9-1	9-2	9-3
Outlet stream compositions:			
HAW stream			
HNO_3, mol/L	3.0	2.9	3.1
U, mg/L	0.2	130	3
Pu, mg/L		1.1	8.4
^{241}Am, mg/L		206	166
^{242}Cm, mg/L		4.22	2.90
^{95}Zr, GBq/L		389	264
^{95}Nb, GBq/L		614	305
^{106}Ru, GBq/L		127	80.5
^{125}Sb, GBq/L		122	80.7
^{134}Cs, GBq/L		542	394
^{137}Cs, GBq/L		1040	892
^{141}Ce, GBq/L		24.3	6.98
^{144}Ce, GBq/L		7190	4550
^{154}Eu, GBq/L		48.4	38.8
^{155}Eu, GBq/L		166	139
HAP stream			
HNO_3, mol/L	0.02	0.03	<0.02
HNO_2, mol/L		0.0015	
U, g/L	85.3	65.4	19.0
Pu, g/L		12.9	3.8
^{95}Zr, MBq/L		10.3	0.293
^{95}Nb, MBq/L		25.9	0.666
^{106}Ru, MBq/L		3.0	0.269
^{125}Sb, MBq/L		<0.3	<0.02
^{134}Cs, MBq/L		<0.03	<0.01
^{137}Cs, MBq/L		<0.1	<0.01
^{141}Ce, MBq/L		<0.1	0.057
^{144}Ce, MBq/L		<0.6	<0.08
^{154}Eu, MBq/L		<0.07	<0.01
^{154}Eu, MBq/L		<0.4	<0.05

[a]Average flow rate, the HAX varied from 0.63 to 1.0 L/h during the run.

[b]Average flow rate, the HAX varied from 1.7 to 2.3 L/h during the run.

Table A-3. Campaign 9 first-cycle tests — conditions and results for B-bank contactor

	Run number	
	9-2	9-3
Dates	3/27/85	5/8-9/85
Bank temperature, °C	18-20	13-18
Number of stages:		
strip/scrub	11/5	
strip/acid add'n/scrub		11/1/4
HBX stream flow rate, L/h	0.562	0.505
Flow ratios:		
HAP/HBX	1.64[a]	3.56[b]
HBS/HBX	0.96	2.64[c]
HBIX/HBX		0.099
Inlet stream compositions:		
HBX stream		
HNO_3, mol/L	0.04	0.20
HAN,[d] mol/L	0.50	
HBS stream, % TBP	30±0.5	10±0.5
HAP stream		
HNO_3, mol/L	0.03	<0.02
HNO_2, mol/L	0.0015	
U, g/L	65.4	19.0
Pu, g/L	12.9	3.8
^{95}Zr, MBq/L	10.3	0.293
^{95}Nb, MBq/L	25.9	0.666
^{106}Ru, MBq/L	3.0	0.269
^{125}Sb, MBq/L	<0.3	<0.02
^{134}Cs, MBq/L	<0.03	<0.01
^{137}Cs, MBq/L	<0.1	<0.01
^{141}Ce, MBq/L	<0.1	0.057
^{144}Ce, MBq/L	<0.6	<0.08
^{154}Eu, MBq/L	<0.07	<0.01
^{155}Eu, MBq/L	<0.4	<0.05

Table A-3 (continued)

	Run number	
	9-2	9-3
Outlet stream compositions:		
HBP stream		
HNO_3, mol/L	0.39	0.65
U, mg/L	0.9	240
Pu, g/L	27.0	14.4
^{95}Zr, MBq/L	3.19	0.858
^{95}Nb, MBq/L	3.43	0.275
^{106}Ru, MBq/L	<0.4	<0.09
^{125}Sb, MBq/L	<0.09	<0.03
^{134}Cs, MBq/L	<0.03	<0.02
^{137}Cs, MBq/L	<0.06	0.081
^{141}Ce, MBq/L	0.31	0.172
^{144}Ce, MBq/L	<0.3	0.261
^{154}Eu, MBq/L	<0.6	<0.02
^{155}Eu, MBq/L	<0.4	<0.08
HBU stream		
HNO_3, mol/L	<0.01	
U, g/L	42.0	9.45
Pu, mg/L	0.53	3.5
^{95}Zr, MBq/L	5.56	0.107
^{95}Nb, MBq/L	11.7	0.283
^{106}Ru, MBq/L	2.26	0.175
^{125}Sb, MBq/L	<0.2	<0.01
^{134}Cs, MBq/L	<0.02	<0.01
^{137}Cs, MBq/L	<0.07	<0.01
^{141}Ce, MBq/L	<0.07	<0.01
^{144}Ce, MBq/L	<0.4	<0.03
^{154}Eu, MBq/L	<0.06	<0.01
^{155}Eu, MBq/L	<0.1	<0.02

[a]Average ratio, the HAP flow rate varied from 0.63 to 1.0 L/h during this run.

[b]Average ratio, the HAP flow rate varied from 1.7 to 2.3 L/h during this run.

[c]Average ratio, the HBS flow rate varied from 1.3 to 1.6 during this run.

[d]Hydroxylamine nitrate.

Table A-4. Campaign 9 first-cycle tests — uranium strip bank conditions and results

	Run number	
	9-2	9-3
Dates	3/27/85	5/8-9/85
Bank temperature, °C	51	48-49
Number of stages	16	16
HCX stream flow rate, L/h	1.31	0.824
Flow ratios:		
HBU/HCX	1.11	3.80
Inlet stream compositions:		
HCX stream		
HNO_3, mol/L	0.05	0.04
HAN, mol/L	0.05	0.05
HBU stream		
HNO_3, mol/L	<0.01	
U, g/L	42.0	9.45
Pu, mg/L	0.5	3.5
^{95}Zr, MBq/L	5.56	0.107
^{95}Nb, MBq/L	11.7	0.283
^{106}Ru, MBq/L	2.26	0.175
^{125}Sb, MBq/L	<0.2	<0.01
^{134}Cs, MBq/L	<0.02	<0.01
^{137}Cs, MBq/L	<0.07	<0.01
^{141}Ce, MBq/L	<0.07	<0.01
^{144}Ce, MBq/L	<0.4	<0.03
^{154}Eu, MBq/L	<0.06	<0.01
^{155}Eu, MBq/L	<0.1	<0.02

Table A-4 (continued)

	Run number	
	9-2	9-3
Outlet stream compositions:		
HCW stream		
HNO_3, mol/L	<0.01	
U, mg/L	0.40	<1
Pu, mg/L	0.0098	0.0029
^{95}Zr, MBq/L	1.33	0.080
^{95}Nb, MBq/L	2.34	0.159
^{106}Ru, MBq/L	1.20	0.067
^{125}Sb, MBq/L	<0.07	<0.01
^{134}Cs, MBq/L	<0.02	<0.01
^{134}Cs, MBq/L	<0.02	<0.01
^{141}Ce, MBq/L	<0.02	<0.01
^{144}Ce, MBq/L	<0.1	<0.02
^{154}Eu, MBq/L	<0.03	<0.01
^{155}Eu, MBq/L	<0.03	<0.01
HCP stream		
HNO_3	0.06	0.06
U, g/L	48.5	38.1
Pu, mg/L	0.22	17
^{95}Zr, MBq/L	2.01	0.201
^{95}Nb, MBq/L	5.64	0.566
^{106}Ru, MBq/L	0.3	0.091
^{125}Sb, MBq/L	<0.04	<0.01
^{134}Cs, MBq/L	0.036	<0.01
^{137}Cs, MBq/L	0.069	0.044
^{141}Ce, MBq/L	<0.01	<0.01
^{144}Ce, MBq/L	0.1	<0.05
^{154}Eu, MBq/L	<0.02	<0.01
^{155}Eu, MBq/L	0.07	<0.03

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