DP-1649

APPLICABILITY OF HYDROXYLAMINE NITRATE REDUCTANT IN PULSE COLUMN CONTACTORS

DONALD J. REIF

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Savannah River Laboratory
Aiken, SC 29808

PREPARED FOR THE U. S. DEPARTMENT OF ENERGY UNDER CONTRACT DE-AC09-76SR00001

Printed in the United States of America

Available from

National Technical Information Service
U. S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22161

Price: Printed Copy A03; Microfiche A01

725737 ✓

DP-1649

Distribution Category: UC-10

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Approved by: H. D. Harmon
Actinide Technology

Published: May 1983

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ABSTRACT

Uranium and plutonium separations were made from simulated breeder reactor spent fuel dissolver solution with laboratory sized pulse column contactors. Hydroxylamine nitrate (HAN) was used for reduction of plutonium (IV). An integrated extraction-partition system, simulating a breeder fuel reprocessing flowsheet, carried out a partial partition of uranium and plutonium in the second contactor. Tests have shown that acceptable coprocessing can be obtained using HAN as a plutonium reductant. Pulse column performance was stable even though gaseous HAN oxidation products were present in the column. Gas evolution rates up to 0.27 cfm/ft^2 of column cross section were tested and found acceptable.

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APPLICABILITY OF HYDROXYLAMINE NITRATE REDUCTANT IN PULSE COLUMN CONTACTORS

INTRODUCTION

The Hot Experimental Facility (HEF) has been designed at Oak Ridge National Laboratory for nuclear fuel reprocessing.[1] The HEF is a pilot-scale reprocessing facility capable of reprocessing typical breeder reactor and light water reactor (LWR) fuels using a uranium-plutonium fuel cycle. A modified high-acid Purex process will be used to recover fissile elements from the fuel for recycle to breeder reactors. The extraction system provides co-decontamination cycles as well as purification cycles for uranium/plutonium coproduct and uranium product. Safeguarding of fissile materials from diversion is enhanced by never completely separating uranium from plutonium. It would not be practical to control solvent extraction operations well enough to yield the exact plutonium concentration desired in the final product. Therefore the target concentration for the product solution is 25% to 40% plutonium.

The separation of plutonium from uranium is easier if some or all of the plutonium is reduced to the essentially nonextractable Pu(III) form. Hydroxylamine nitrate (HAN) is being considered for plutonium reduction because its oxidation products do not contribute to solid waste volume and no species are produced which adversely affect waste handling processes. The formation of the noncondensable gases, if formed in large amounts, may disrupt pulse column operation. This would not be a problem when treating LWR fuels because plutonium concentrations are low (about 1 to 2% of heavy metals). Much larger amounts of gas would be formed when processing breeder fuels which could contain 10 to 22% plutonium. No information exists to decide if separations involving complete reduction of plutonium with HAN at breeder plutonium levels is practical with pulse column contactors.

This report describes experimental studies of uranium/plutonium separations with simulated breeder level plutonium feeds in laboratory sized pulse column contactors. The specific objective of this program was to determine the feasibility of reducing breeder level plutonium concentrations with HAN in pulse column contactors.

SUMMARY

Coprocessing or partial partitioning of uranium and plutonium was successfully conducted in a pulse column contactor using HAN for plutonium reduction. The pulse column operated in a controllable manner even though a gas phase continuously evolved during operation. Simulated breeder dissolver solutions containing about 10% plutonium were extracted and partitioned in an continuous process involving two pulse column contactors. Plutonium was reduced with HAN in the partition column using a process flowsheet which yielded a coproduct solution containing >25% plutonium (as % of heavy metal). HAN oxidation products produced calculated gas evolution rates up to 0.27 cfm/ft^2 when liquid throughput rates of about 600 gal/hr/ft^2 were used. Although gas phase was visibly present in the contactor, acceptable end-stream compositions were obtained. Low plutonium loss to the organic (BU) stream was found (0.2%) and the aqueous coproduct (BP) stream was 26% plutonium (of heavy metals). No column flooding was observed during the partition run. A foam layer was visible at the column top but no visible aqueous phase exited with the organic overflow. End-stream compositions responded properly to process variable changes during HAN reductions with gas phase present.

Within the scope of this work, nothing indicated that the use of HAN for plutonium reduction in pulse column contactors would be prohibited. Thus we conclude that HAN can be included as a reductant in separation flowsheets for processing spent breeder fuels where pulse column contactors are proposed.

DISCUSSION

Experimental

Program objectives required that large quantities of plutonium be handled in pulse column contactors. Capabilities for use of large amounts of plutonium were available in the Savannah River Laboratory (SRL) but a facility for making solvent extraction runs with pulse column contactors did not exist. Therefore, it was necessary to design and install a laboratory size Pulse Column Facility (PCF) in SRL.

Equipment

Description

The PCF was designed to be housed in a standard SRL double laboratory module. The radioactive portion of the facility is enclosed in a floor-to-ceiling glove box. The glove box contained equipment to carry out solvent extractions, clean solvent for reuse, and concentrate heavy metals for recycle. The facility is illustrated schematically in Figure 1. The solvent extraction apparatus consists of four one-inch (inside diameter) glass columns from six to eight feet long, depending on the column purpose. Three columns are fitted for normal solvent extraction usage; the fourth is fitted with an electrolytic cell for **in-situ** reductions. One column, (Figure 1, Column A), used as the first contactor has five input feed points located along the column length and ends. This column was designed to be used principally as an organic continuous column. The radioactive feed tank, located in the glove box is piped to this column. The second column (B) has four feed points and the third (E) and fourth columns (C) are end fed only. The B, E, and C columns were designed to run aqueous continuous. Each column has two exit streams, one at the top which flows to an intercolumn reservoir, except for the C column, and one at the bottom which flows through a detector loop to aqueous product tanks. The upper exit stream ultimately flows to the organic waste tank. All columns are agitated by an air pulse system. The columns, pulse systems and column phase interface control systems were purchased from Allied General Nuclear Services, Barnwell, SC. The chemical feed system is located in a hood across from the glove box. Except for the radioactive feed and the organic phase feed, feed solutions were transferred from the hood to the columns with metering pumps.

A more detailed description of the SRL PCF can be found in Reference 2.

Procedures

Uranium was determined by colorimetry or by titration by the Davies and Gray method.[3] Plutonium was determined by gross alpha and alpha pulse height analysis and also by gamma spectroscopy with low energy gammas from ^{239}Pu. Hydroxylamine was measured by oxidation with potassium bromate followed by titration of excess bromate by the iodine method. Hydrazine was measured by a colorimetric method involving p-dimethylaminobenzaldehyde.

Diluent was obtained from SRP (a mixture of C_{12} to C_{15}, n-paraffin hydrocarbons with an average molecular weight of about 190. Diluent was mixed with reagent-grade 100 vol % tributylphosphate (TBP) to make 30 $\pm$0.2 vol % TBP solution. The 30% TBP was washed successively with 0.5M Na_2CO_3, 0.1M HNO_3 and water before each use. All other chemicals were reagent-grade and used without further purification.

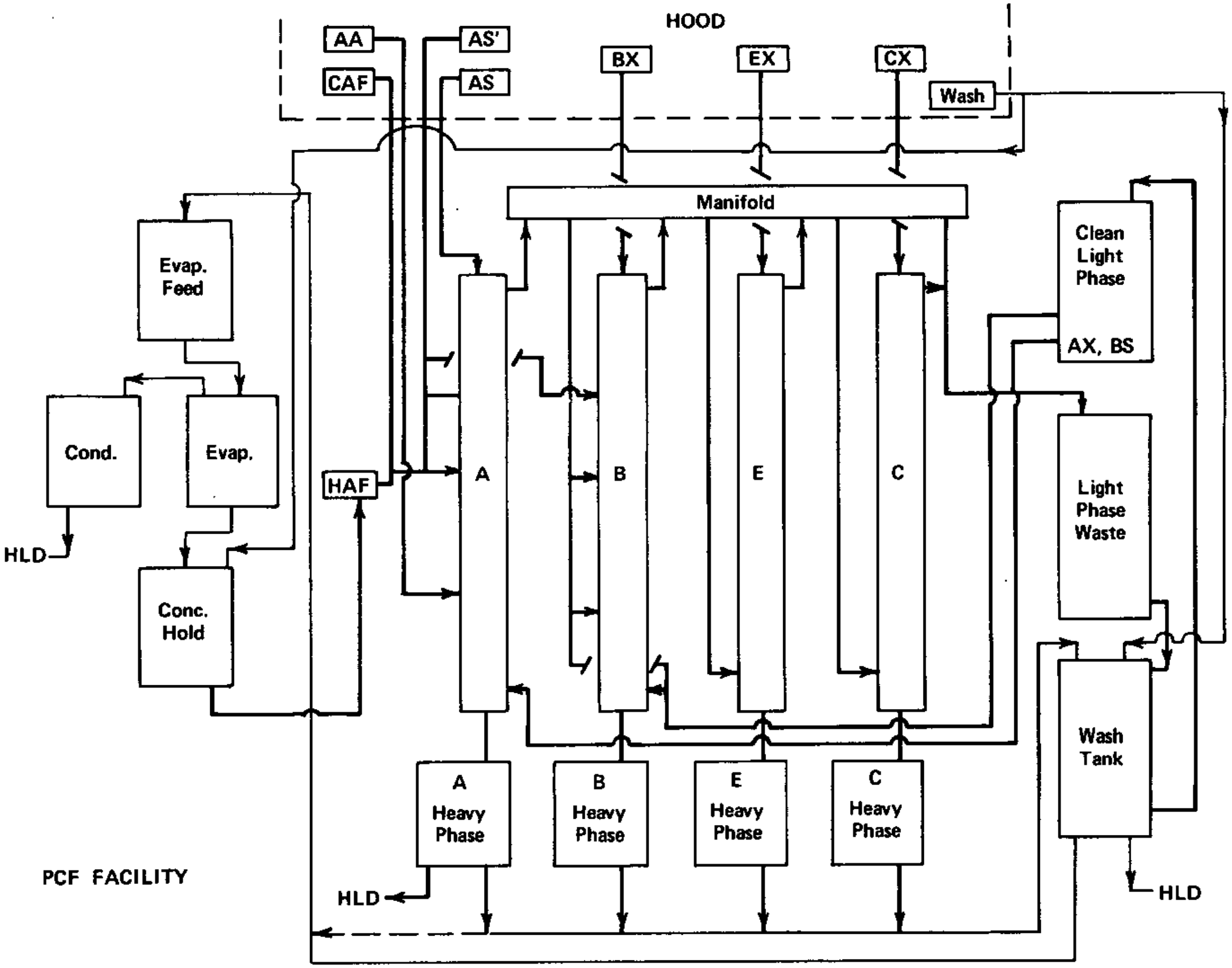


FIGURE 1. PCF Facility

Results

Experimental work was begun with nonradioactive materials to establish equipment capacities and reproducibility. This work was followed by runs with radioactive materials simulating a coprocess flowsheet proposed for breeder fuel reprocessing.

Cold Studies

Column Flooding Tests

Flooding tests were made to define the operating zone for each column. Two types of feeds were used. One representing a nonradioactive startup condition used 30% TBP equilibrated with 3M HNO_3 (organic stream) and 3M HNO_3 (aqueous stream). The upper flooding curves with this system are shown in Figure 2. The second system, representing a simulated radioactive run condition, used 30% TBP saturated with uranyl nitrate in 3M HNO_3 and 3M HNO_3. The flooding curves for this system (Figure 3) shifted toward lower capacity as expected when phase densities become more similar. These tests indicate, with heavy metals present, that the maximum operating capacities for these 1 inch ID columns are 18 L/hr, 25 L/hr, 14 L/hr and 7 L/hr for the A-, B-, C- and E-Columns respectively.

Extraction/Stripping Tests

A series of five runs were made to establish the reproducibility of the over-all system and to estimate the stage height in the extraction column. A two column extraction-stripping (Figure 4) system was used for these tests. The feed (AF) was introduced at one of three points in the extraction column (Figure 4, positions 1, 2 or 3) in order to change the length of the extraction zone. The nonradioactive test conditions and results are given in Table I. The outlet stream compositions are at "steady-state" ("steady-state" is defined as the running period when the instantaneous exit stream density varies less than 0.0025 g/mL). The values in Table I are the average of three samples taken during the "steady-state" period. The results for each sample are given in Table II.

The PCF reproducibility was checked by performing duplicate runs and comparing end stream compositions. Runs PC-10 and 13 (mid-top fed) and Runs PC-11 and 12 (top fed) served to demonstrate the extraction column (A-Column) reproducibility and Runs PC-10 and 13 checked the combined (A- and B-Column) reproducibility. The results, given in Tables III and IV, show that good reproducibility was obtained.

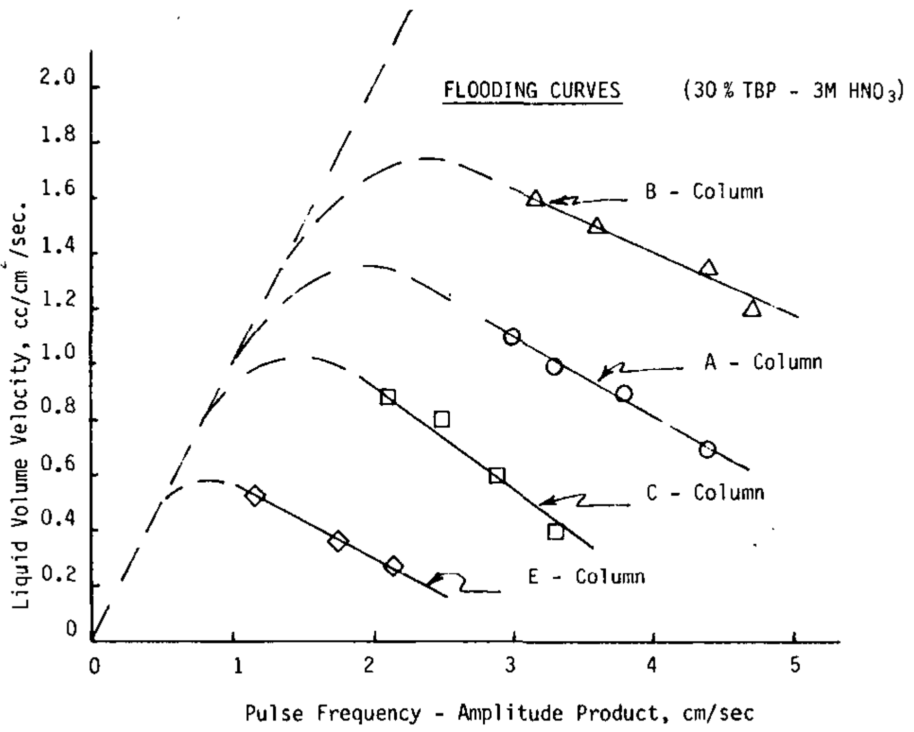


FIGURE 2. PCF Flooding Curves - No Heavy Metals

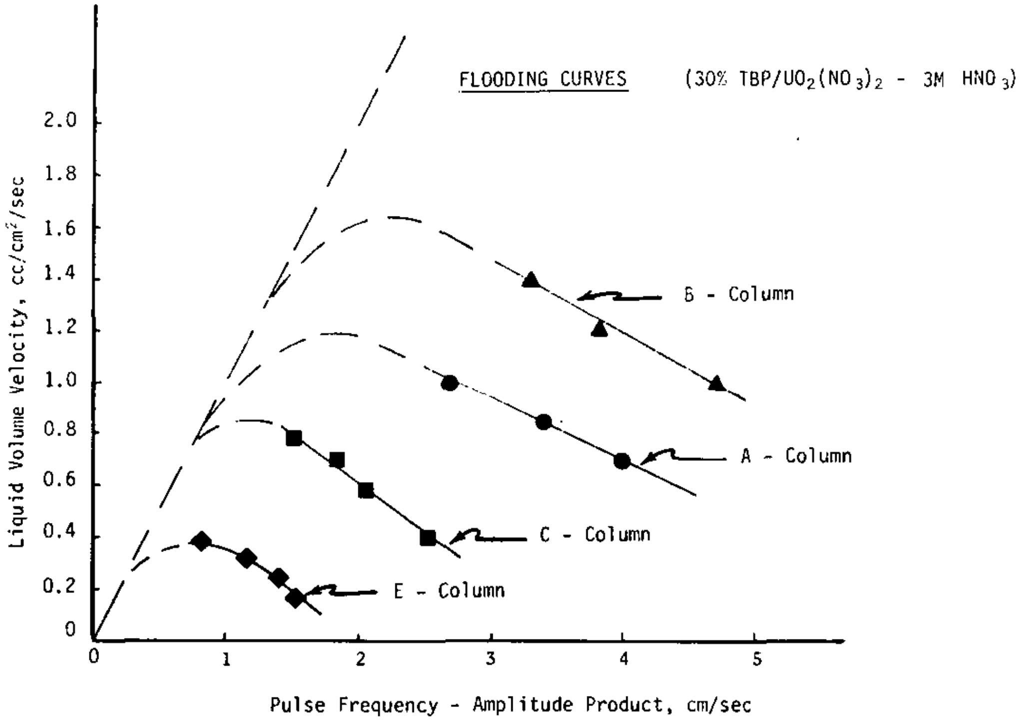
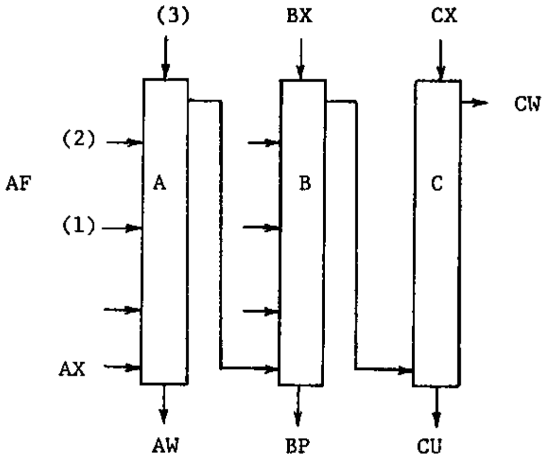


FIGURE 3. PCF Flooding Curves - With Uranyl Nitrate



STREAM	COMPOSITION	REL. FLOW RATE	ABS. FLOW RATE, mL/min.
AF	138g/l U; 3.7M HNO_3	0.64	60
AX	30% TBP	1.00	94.5
BX	0.05M HNO_3	0.92	86.7
CX	0.01M HNO_3	0.92	86.7

FIGURE 4. Nonradioactive Test Flow Diagram

TABLE I

Nonradioactive Test Conditions and Results

				Run No.		
		PC-9	PC-10	PC-11	PC-12	PC-13
Column Temp. °C	A/B	30/30	30/40	30/40	30/50	30/40
Pulse Amplitude, cm	A/B	1.5/1.5	1.5/1.5	1.5/1.5	1.5/1.5	1.5/1.5
Pulse Frequency, c/min	A/B	110/114	110/114	110/114	110/114	110/114
AF Feed Point		Middle	Mid Top	Top	Top	Mid Top
AX Flow Rate, mL/min		94.5	94.5	94.5	94.5	94.5
<u>Flow Ratios</u>						
AF/AX		0.636	0.636	0.636	0.636	0.636
BX/AX		0.917	0.635	0.636	0.636	0.636
<u>Inlet Stream Compositions</u>						
AF Stream, U,g/L		135.5	133.1	139.9	141.4	135.6
HNO_3, M		3.4	3.8	3.7	3.3	3.7
AX Stream, % TBP		30	30	30	30	30
BX Stream, HNO_3, M		0.05	0.05	0.05	0.05	0.05
<u>Outlet Stream Composition</u>						
AP Stream, U, g/L		87.1	86.6	83.7	92.2	86.8
HNO_3, M		0.27	0.25	0.27	0.24	0.28
AW Stream, U, g/L		1.27	0.13	0.006	0.008	0.12
HNO_3, M		3.2	3.4	3.2	3.2	3.4
BU Stream, U, g/L		17.4	14.1	11.8	8.14	13.0
HNO_3, M		0.005	0.005	0.01	0.005	0.005
BP Stream, U, g/L		72.4	79.8	87.9	86.9	88.9
HNO_3, M		0.35	0.35	0.37	0.32	0.35

TABLE II

Nonradioactive Exit Stream Composition

			Run No.				
			PC-9	PC-10	PC-11	PC-12	PC-13
AW Stream,	H^+, M	1	3.17	3.36	3.20	3.18	3.51
		2	3.15	3.37	3.24	3.28	3.40
		3	3.20	3.40	-	-	-
	U, g/L	1	1.12	0.136	0.007	0.007	0.13
		2	1.35	0.103	0.005	0.010	0.10
		3	1.35	0.136	-	-	-
AP Stream,	H^+, M	1	0.27	0.24	0.25	0.25	0.26
		2	0.27	0.26	0.29	0.24	0.30
		3	0.27	0.26	-	-	-
	U, g/L	1	87.74	87.23	83.87	92.17	85.53
		2	88.94	87.23	83.51	92.29	88.06
		3	84.82	85.32	-	-	-
BP Stream,	H^+, M	1	0.35	0.34	0.36	0.34	0.35
		2	0.36	0.36	0.38	0.30	0.36
		3	0.35	0.35	-	-	-
	U, g/L	1	72.68	78.78	89.94	88.30	93.01
		2	72.20	81.22	85.89	85.55	84.68
		3	72.38	79.26	-	-	-
BU Stream,	H^+, M	1	0.004	0.09	0.01	0.005	0.005
		2	0.006	0.004	0.01	0.005	0.005
		3	0.004	0.005	-	-	-
	U, g/L	1	16.92	14.56	12.12	9.86	14.52
		2	18.13	13.67	11.53	6.41	11.47
		3	17.10	14.62	-	-	-

TABLE III

Extraction Column Reproducibility

		Feed Point			
		Top		Mid-Top	
		PC-11	PC-12	PC-10	PC-13
% U	AW	0.005	0.006	0.10	0.08
	AP	96	104	103	102
H^+, M	AW	3.2	3.2	3.4	3.4
	AP	0.27	0.24	0.26	0.28

TABLE IV

Two-Column Combined Reproducibility

		Run No.	
		PC-10	PC-13
% U	AW	0.10	0.08
	AP	103	102
	BU	16.3	13.2
	BP	86.1	88.3
H^+, M	AW	3.4	3.4
	AP	0.26	0.28
	BU	0.005	0.005
	BP	0.36	0.35

An estimation of the number of stages in the extraction column was made by first calculation, using the SEPHIS-MOD4 (Reference 4), of steady-state end-stream compositions for 2, 3, 4 and 5 stages of extraction. The experimental feed compositions and feed rates were used. Then a graph was prepared plotting the log of the calculated uranium concentration of the aqueous raffinate versus the number of extraction stages. Finally the number of stages of extraction for the various feed points were estimated from the graph by interpolation of the aqueous raffinate uranium concentrations observed for each feed point. These results are given in Table V. Based on these data the height equivalent of the theoretical stage (H.E.T.S.) for the extraction column is approximately 1.8 feet.

Radioactive Studies

Hydroxylamine nitrate (HAN) is being considered more frequently as a reductant for plutonium in solvent extraction separations. This is primarily because HAN oxidation products are gaseous and hence do not contribute to solid wastes. A study of HAN reduction kinetics[5] showed two reaction routes depending on the HAN to plutonium ratio.

$$2\ NH_3OH^+ + 2\ Pu^{+4} \longrightarrow 2\ Pu^{+3} + N_2 + 2H_2O + 4H^+ \qquad (1)$$

$$2\ NH_3OH^+ + 4\ Pu^{+4} \longrightarrow 4\ Pu^{+3} + N_2O + H_2O + 6H^+ \qquad (2)$$

Reaction 1 takes place when HAN is present in excess (HAN/Pu >1). Reaction 2 predominates for HAN/Pu <1. Since HAN is inextractable the aqueous phase would be expected to contain small amounts of Pu^{+4} and thus Reaction 1 would predominate during uranium/plutonium partitioning.

Plutonium reduction with HAN has been studied in Light Water Reactor (LWR) reprocessing systems.[6] In LWR systems plutonium is about 1% of the feed heavy metals and, at these low concentrations, produce small volumes of gas when reduced by HAN. Processing breeder or fast flux test facility (FFTF) fuels would deliver plutonium at much higher concentrations (15 to 22% of heavy metals) which would produce much larger gas volumes. These experiments were carried out to establish if pulse column contactors could produce a satisfactory partition while noncondensable gases escaped.

A two column integrated extraction-partition flowsheet was used (Figure 5). The radioactive feed was introduced at the mid-top point of the extraction column (A) to provide a section for scrubbing to reduce the acidity of the extract. Feed compositions and column operating conditions are given in Table VI. The partition column (B) was end-fed. The feed stream flow rate and temperature, the column temperature (top and bottom), the pulse frequency and pulse amplitude where measured every 30 minutes throughout the

TABLE V

Extraction Stages for A-Column Feed Points

Feed Point	Length of Extraction Section, ft	Uranium in AW, g/L	No. of Stages
Top	5.70	0.006	3
Mid-Top	4.15	0.14	2.25
Center	2.85	1.35	1.7

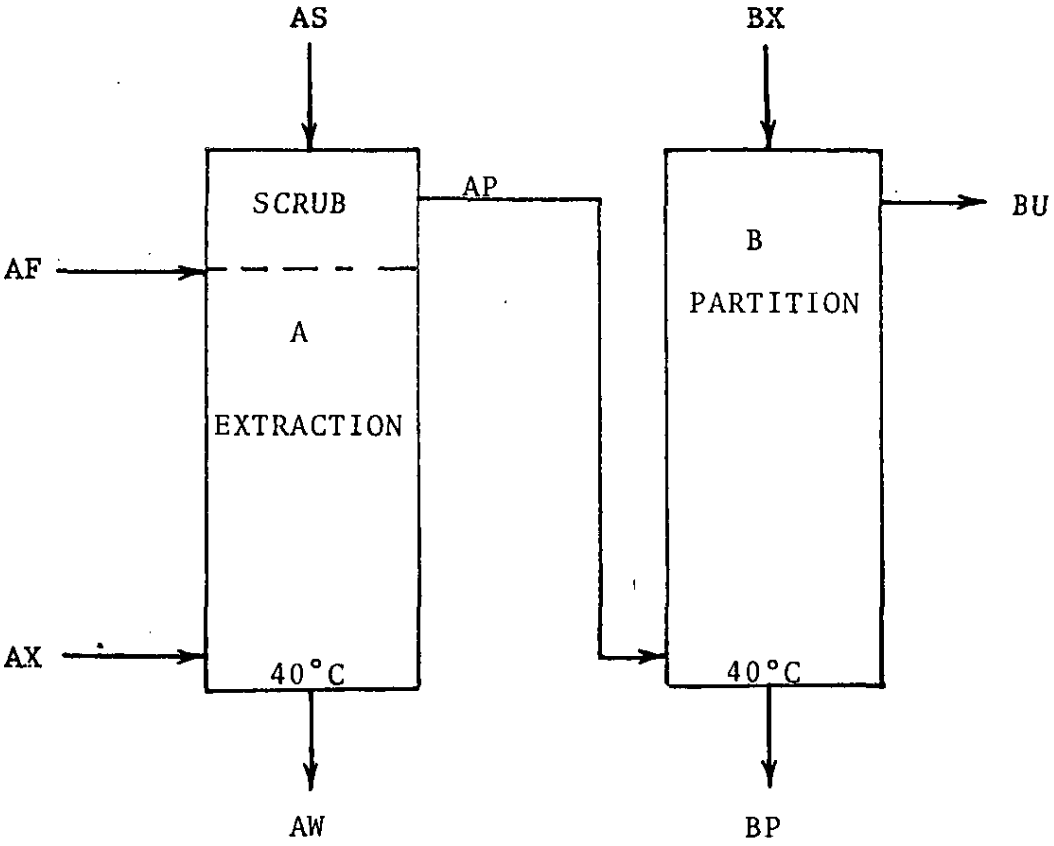


FIGURE 5. Radioactive Test Flow Diagram

TABLE VI

Radioactive Test Conditions and Results

		PC-26	PC-28	PC-30
Column Temperature, °C	A/B	38/41	39/43	38/39
Pulse Amplitude, cm	A/B	1.6/1.7	1.7/1.7	1.7/1.7
Pulse Frequency, c/min	A/B	110/114	110/114	110/114
AX Flow Rate, mL/min		70	80	160
Flow Ratios				
AF/AX		0.571	0.500	0.500
AS/AX		0.205	0.180	0.180
BX/AX		0.286	0.287	0.287
Inlet Stream Compositions				
AF Stream,	U, g/L	129.5	135.1	136.5
	Pu, g/L	14.9	15.25	11.4
	HNO_3, M	2.9	3.1	2.7
AS Stream,	HNO_3, M	0.3	0.3	0.3
AX Stream,	%TBP	30	30	30
BX Stream,	HNO_3, M	0.15	0.05	0.05
	HAN, M	0.077	0.12	0.12
	Hydrazine, M	0.05	0.05	0.05
Outlet Stream Compositions				
AP Stream,	U, g/L	80.2	68.7	75.1
	Pu, g/L	6.37	7.57	5.70
	HNO_3, M	0.048	0.071	0.12
AW Stream,	U, g/L	0.20	0.01	0.01
	Pu, g/L	3.22	0.32	0.015
	HNO_3, M	2.2	2.4	2.1
BU Stream,	U, g/L	53.75	49.0	60.0
	Pu, g/L	0.035	0.011	0.008
	HNO_3, M	0.03	-	0.09
BP Stream,	U, g/L	76.41	73.7	69.5
	Pu, g/L	20.6	26.5	19.8
	HNO_3, M	0.2	-	0.5

run. The AW, BP and BU exit stream density was monitored continuously but read every 2.5 minutes (instantaneous reading). The density readings were plotted as a function of run time as the run progressed. A microcomputer was used to monitor the density meters and operate the X-Y plotter. Exit stream samples (AP, AW, BP, BU) were collected at intervals after the process achieved "steady-state" (based on stream density). The uranium in the partition column organic stream was recovered in a separate two-column stripping run.

Heavy metal solutions from both the extraction/partition run and the stripping run were combined, batch stripped with n-paraffin to remove entrained TBP, and then concentrated by distillation to a volume of about 10 L for reuse. The stripped organic was batch cleaned by successive extractions with aqueous sodium carbonate solution, dilute nitric acid, and then water. The cleaned organic was checked for TBP content and disengagement behavior before returning it to the organic feed tank.

Radioactive feed was prepared by determining the uranium, plutonium, and acid content of the heavy metal concentrate and then making the appropriate adjustments. The goal for radioactive feed was 15 g/L plutonium, 135 g/L uranium in 3.0M nitric acid. The plutonium valence in the concentrate was determined spectrophotometrically and adjusted to Pu (IV) before concentration adjustments were made. Pu (VI) was removed by reduction with hydrogen peroxide. Pu (III) was removed by oxidation with nitrous acid. Nonradioactive chemical feeds were freshly prepared and assayed before each run.

Comparison of HAN Partition Runs

Three HAN partition runs were made where the amount of noncondensable gases produced was increased in each run ranging from 0.14 cfm/ft^2 in Run 26 to 0.27 cfm/ft^2 in Run 30. The gas evolution rates were calculated as shown in Appendix A. Actual measurement of evolved gas was not possible. The end-stream compositions for each run are given in Table VI. The exit stream density is shown in Figures 6, 7, and 8. The BU density was not monitored during the low gas flow run (Run 26). The reason for the variability in the AW and BP densities in Run 26 is unknown. The density of these streams remained stable during intermediate gas flow run (Run 28) and the high gas flow run (Run 30) where oxidation gases were more prevalent, which tends to exclude entrained gas as the cause of the nonuniformity. The content of samples taken during the constant, but highly cyclic, density period were uniform (Table VII) indicating a "steady-state" condition was achieved. The initial portion of the run time (from 0 to about 110 minutes) was conducted using 3M nitric acid as feed to allow feed stream flow rates and temperatures to stabilize before radioactive feed was introduced.

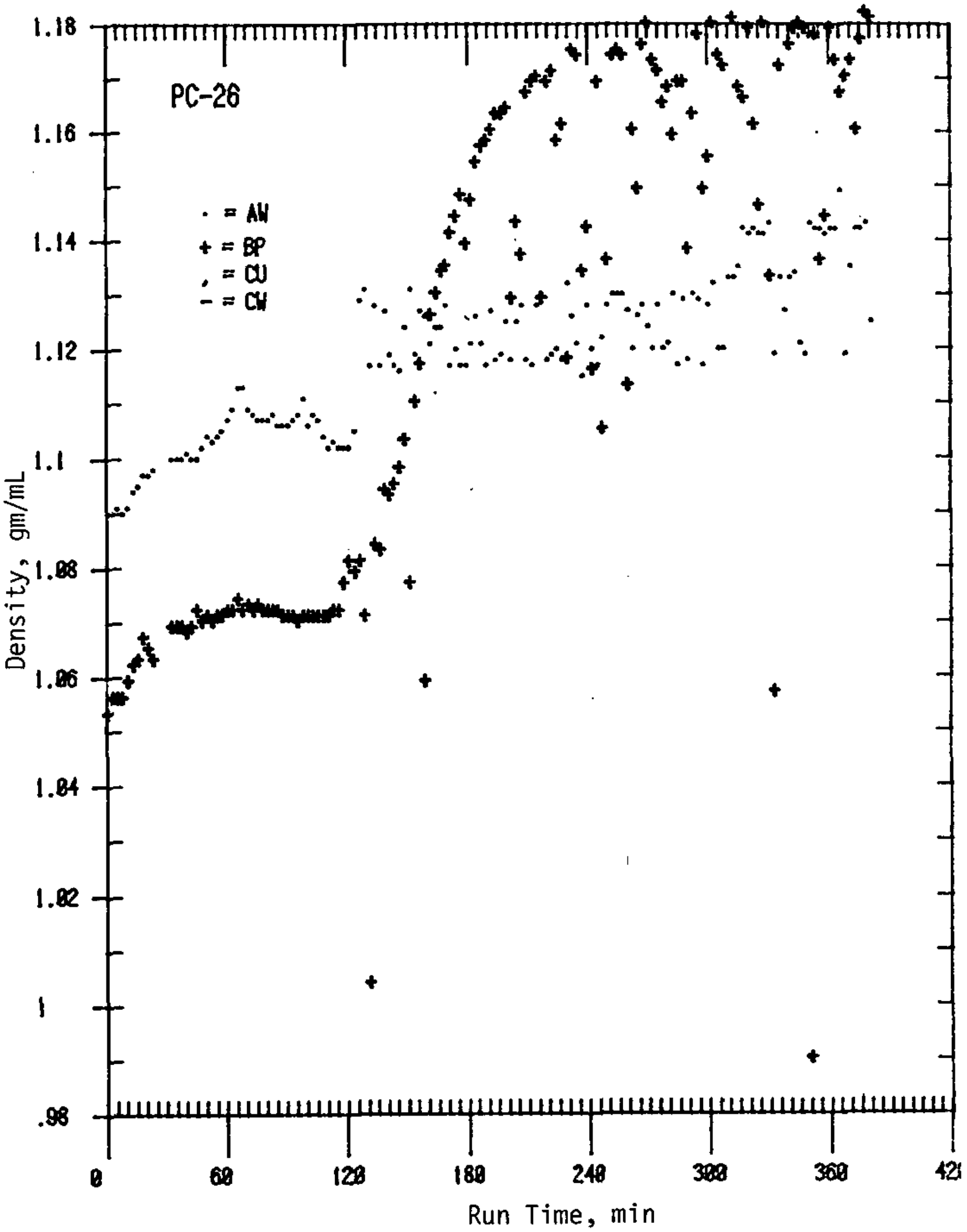


FIGURE 6. Low Gas Flow Exit Stream Density (Run 26)

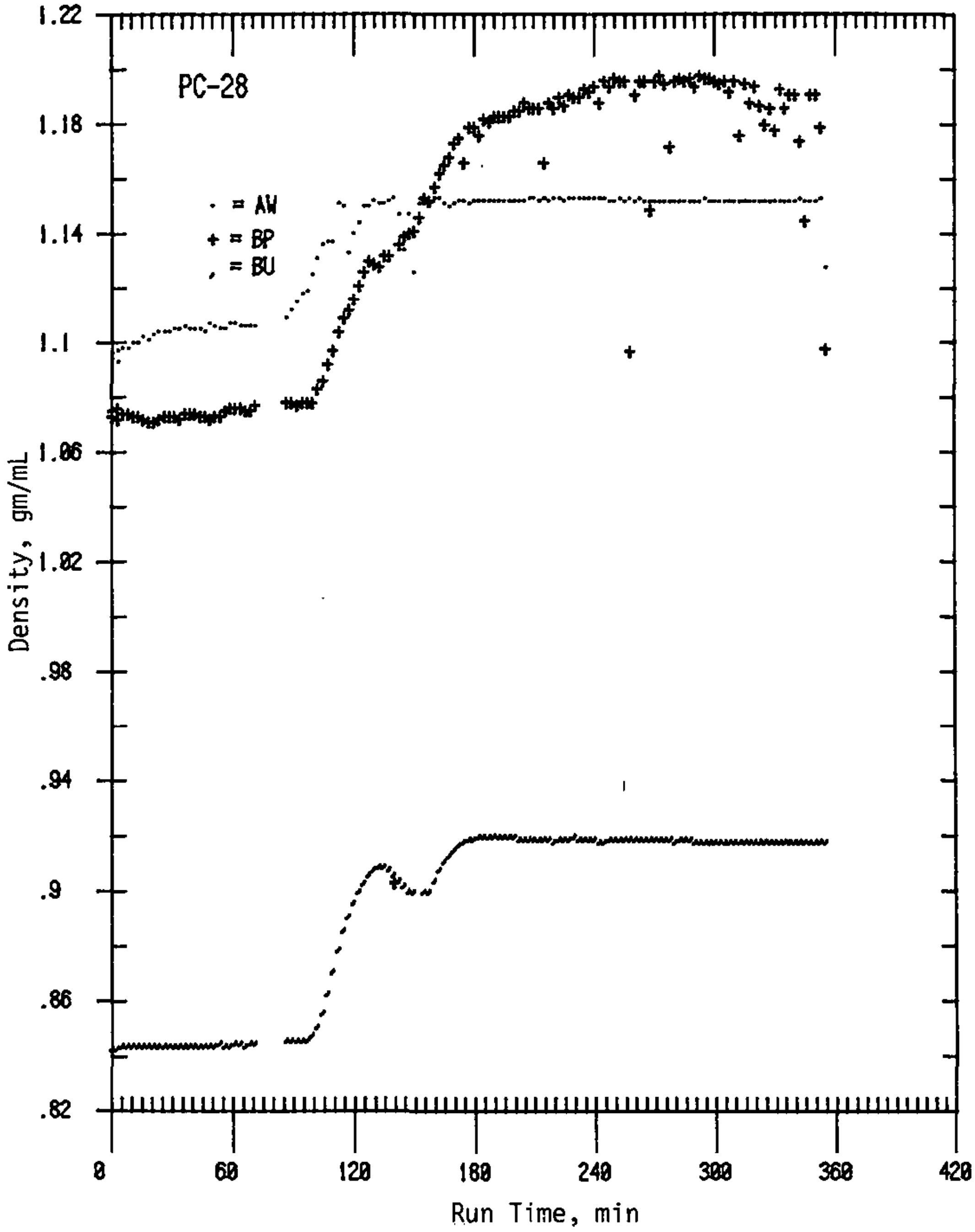


FIGURE 7. Intermediate Gas Flow Exit Stream Density (Run 28)

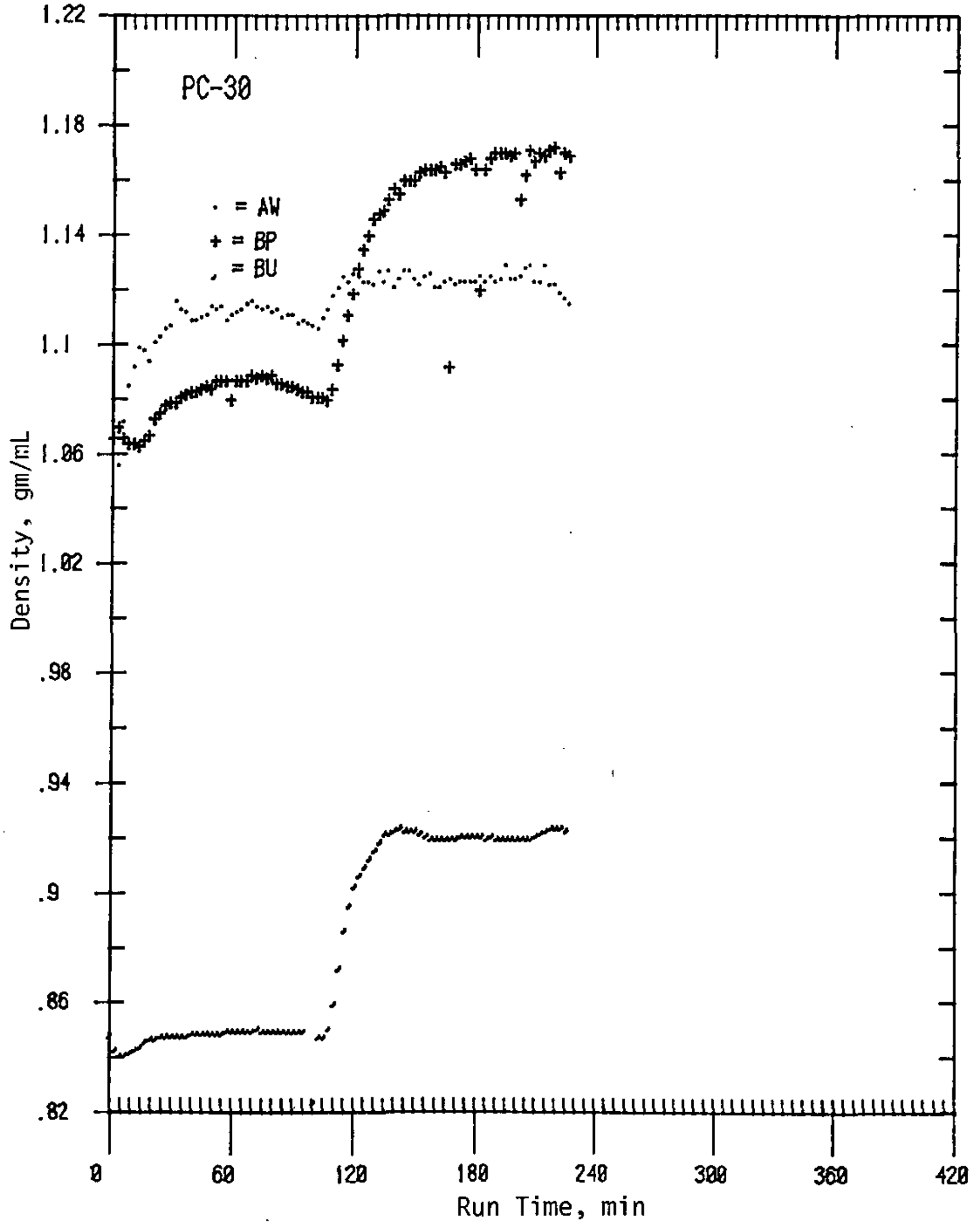


FIGURE 8. High Gas Flow Exit Stream Density (Run 30)

TABLE VII

Low Gas Flow Exit Stream Sample Composition (Run 26)

Stream		Sample Number 1	2	3	Average
AP	H^+, M	0.05	0.05	0.05	0.05
	U, g/L	80.3	80.5	79.6	80.2
	Pu, g/L	-	6.37	-	6.37
AW	H^+, M	2.2	2.2	2.2	2.2
	U, g/L	0.22	0.30	0.25	0.26
	Pu, g/L	2.7	3.6	3.4	3.2
BU	U, g/L	54.7	52.8	53.8	53.8
	Pu, g/L	0.034	0.032	0.039	0.035
BP	H^+, M	0.30	0.30	0.31	0.30
	U, g/L	76.1	76.6	76.6	76.4
	Pu, g/L	22.1	19.9	20.1	20.7

The sharp increase in density clearly indicated radioactive feed start-up.

The visual appearance of the partition columns (shown schematically in Figure 9) was similar in all three runs. A photograph of the Run 28 partition column is shown in Figure 10. A close-up of the Run 30 column upper section is shown in Figure 11. The lower fourth of the column was a dark intense blue color. The next fourth was a green color and the upper half of the column was a straw color. Gas bubbles were visible in the upper three-fourths of the column. No coalesced gas pockets or voids formed within the column or beneath the plates. No cyclic flooding was observed while gas bubbles rose through the column. In all cases the column maintained a stable appearance characteristic of the specific run. A small foam layer was observable at the phase interface and at the top of the organic layer in the upper disengagement section (shown in Figures 12, 13 and 14). The foam layer appeared to be oxidation gases encapsulated with aqueous phase. When the bubbles reached the surface of the organic layer the gases escaped and the aqueous droplet fell back through the organic layer. During the highest throughput run (Run 30) it appeared that some of these falling droplets were entrained toward the organic layer exit but no aqueous was ever detected, via density, in the organic BU stream. No gas bubbles were observed to exit with the organic overflow stream.

Good heavy metal separations with acceptable losses were obtained even though gas evolution occurred in the partition column. As shown in Table VIII, the plutonium losses to the organic stream (BU) were lowest in Runs 28 and 30 even though the gas evolution rate was higher. Process changes were made for Run 28 to increase the plutonium percentage in the BP stream as well as to increase the amount of gas evolved. This was done by reducing the BX stream acidity and increasing the HAN content. These changes produced the desired results even though the gas evolution rate was increased. Run 30 was made at twice the liquid throughput rate of Run 28 but because the plutonium content of the feed (AF) was lower (11.4 g/L versus 15.25 g/L for Run 28) the plutonium content in the product stream did not reach the minimum goal of 25%. The low plutonium loss to the BU stream indicated that the uranium/plutonium partition was accomplished satisfactorily at the highest gas evolution rate.

Conclusions

The partial partitioning of uranium and plutonium, where the plutonium concentration is at breeder level (about 10% of heavy metals), can be successfully accomplished with pulse column contactors with concurrent noncondensable gas evolution. This study has

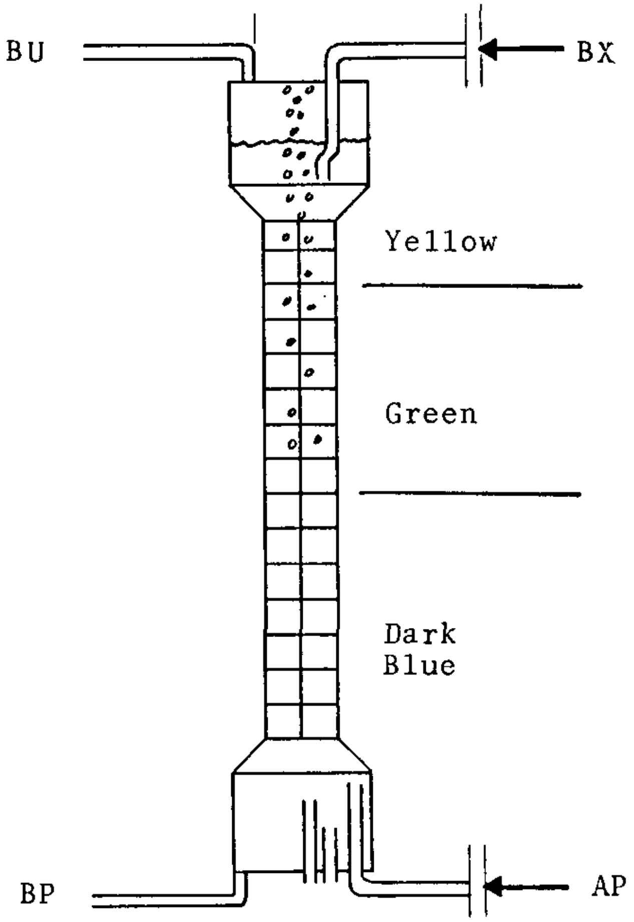


FIGURE 9. Schematic Partition Column Appearance

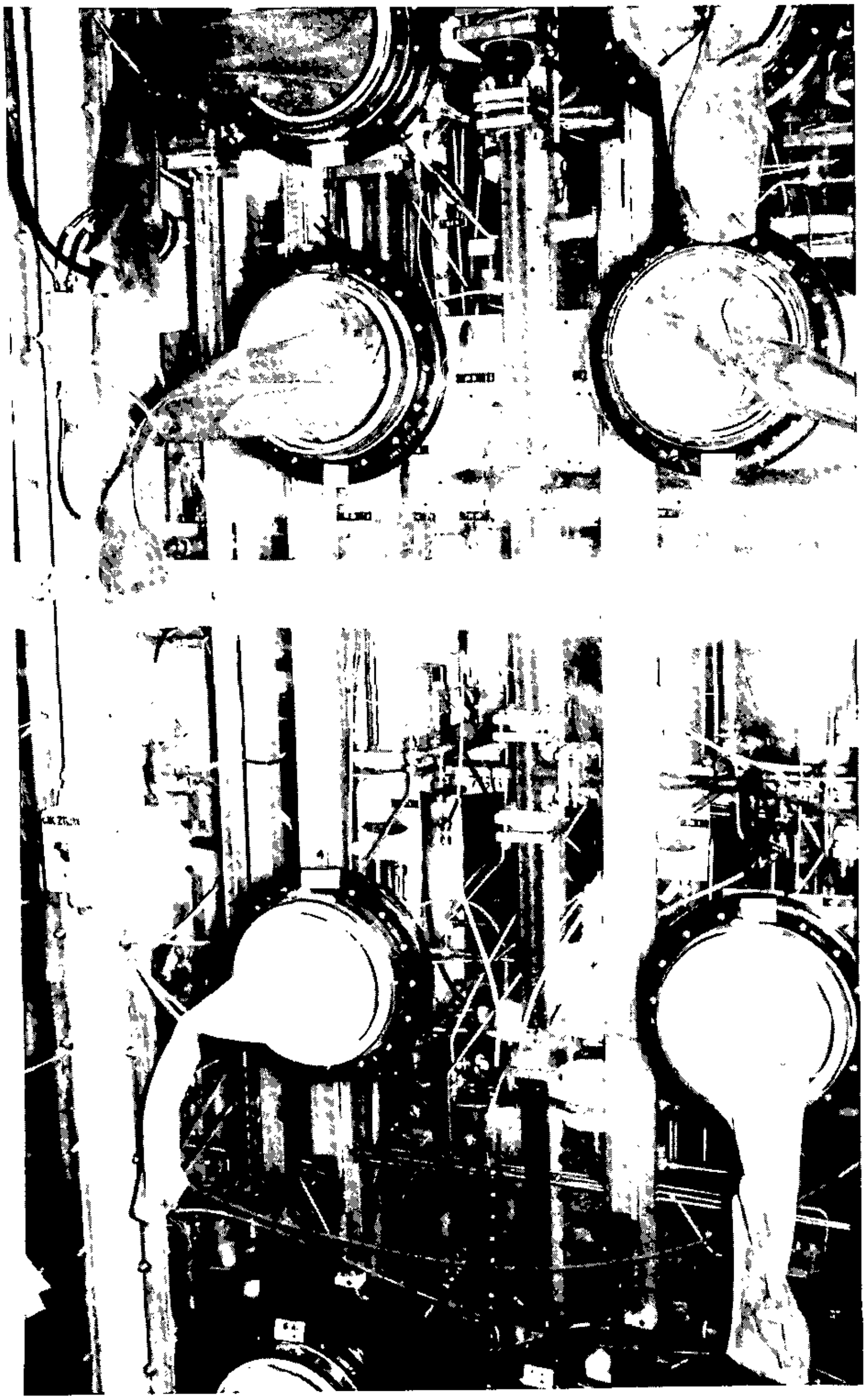


FIGURE 10. Intermediate Gas Flow Partition Column (Run 28)

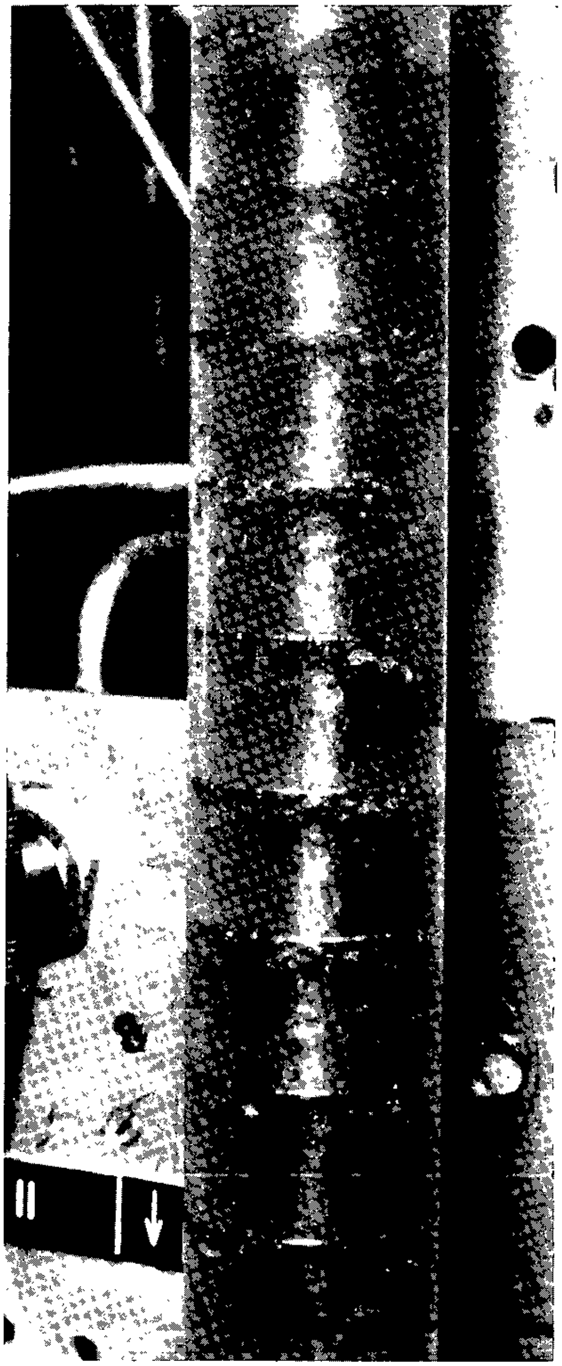


FIGURE 11. High Gas Flow Partition Column - Upper Section (Run 30)

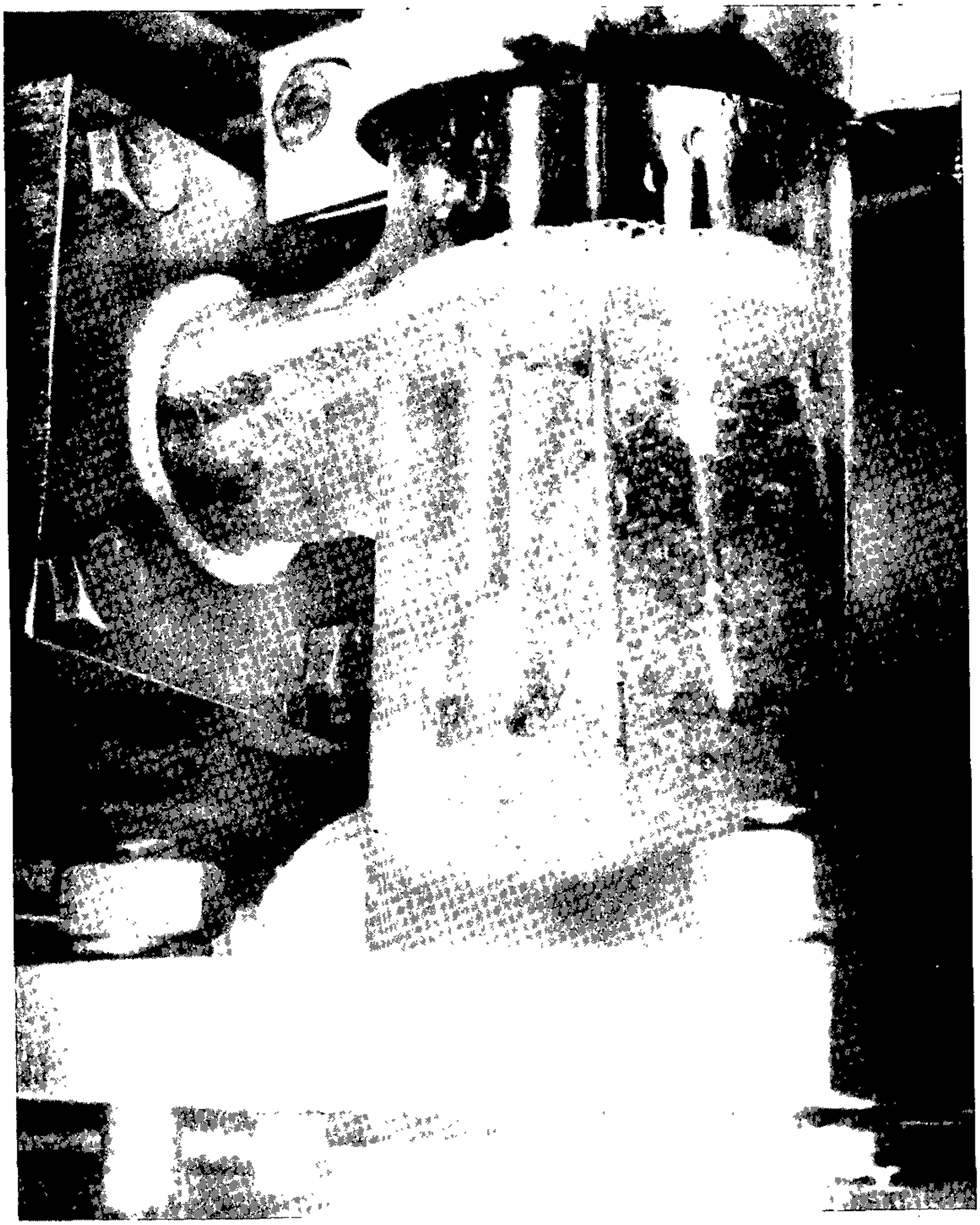


FIGURE 12. High Gas Flow Upper Disengaging Section 1 (Run 30)

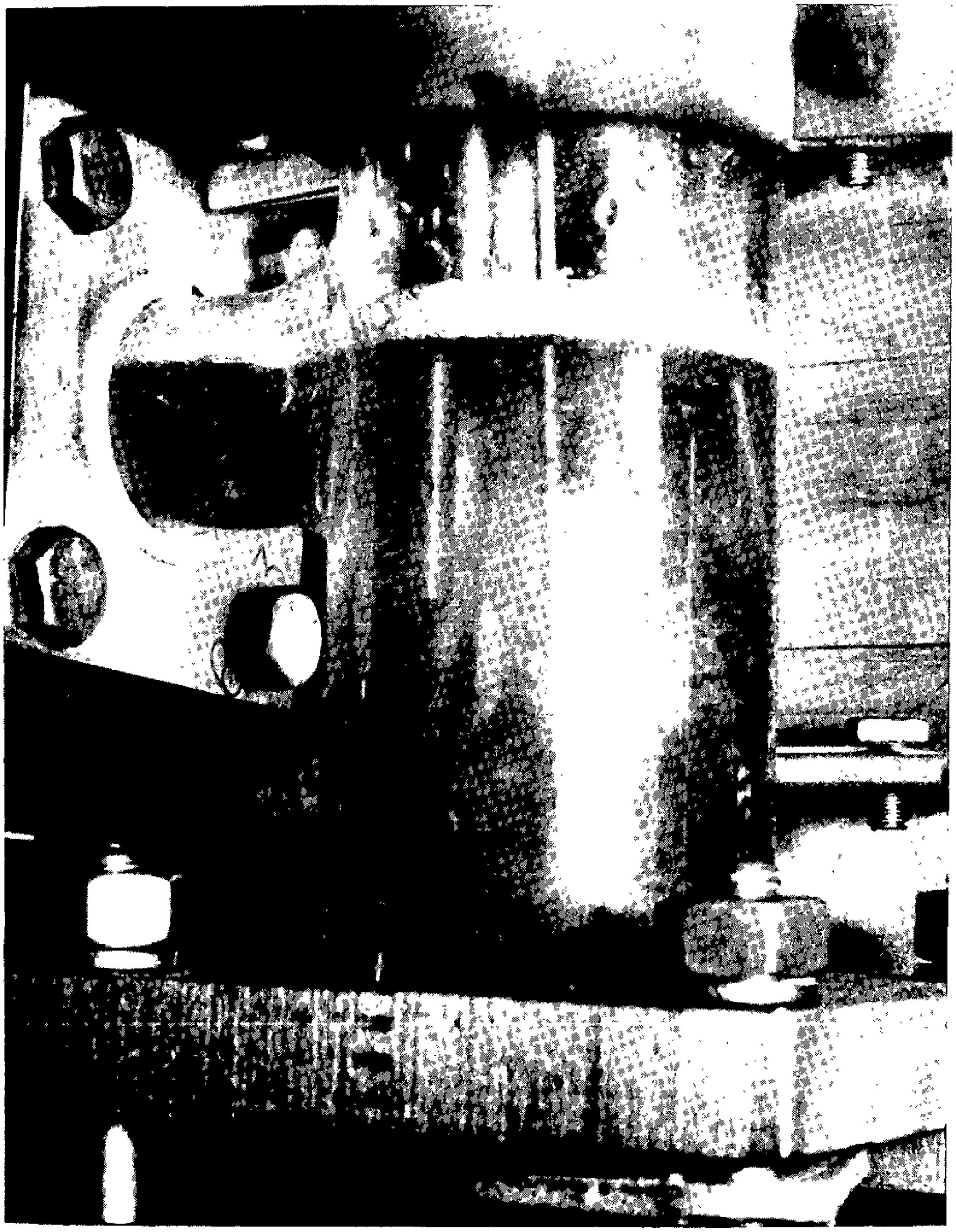


FIGURE 13. High Gas Flow Upper Disengaging Section 2 (Run 30)

FIGURE 14. High Gas Flow Upper Disengaging Section 3 (Run 30)

TABLE VIII

Partition Column Plutonium Results

	Run Number		
	PC-26	PC-28	PC-30
% Pu lost to BU*	0.63	0.13	0.18
% Pu in Product Stream (BP)	21	26	22
Calculated gas flow rate, mL/min	21	29	43
cfm/ft^2	0.14	0.19	0.27

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* Based on Pu fed to B-Column in the AP stream.

shown that pulse column operation was acceptable and process reaction to process changes were normal for gas evolution rates up to 0.27 cfm/ft^2. Higher rates were not studied. Thus, a HAN flowsheet can be selected to reduce solid waste volume for reprocessing breeder spent fuels with pulse column contactors.

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APPENDIX A

Calculation of Off-Gas Evolution Rate

$$\frac{\text{AP Pu Concentration (g/L) x AX Feed Rate (L/min)}}{\text{239 g Pu/mole}} = \text{moles Pu/min}$$

mole Pu/min ÷ 2 = moles HAN consumed/mole Pu reduced/min

moles HAN consumed/min x 22800 mL/mole = mL gas evolved/min

For Run 26

$$\frac{6.37\ (0.07)}{239} = 1.86 \times 10^{-3} \text{ mole Pu/min}$$

$$\frac{1.86 \times 10^{-3}}{2} \times 22800 = 21.3 \text{ mL gas evolved/min}$$