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**Prepared for the U.S. Department of Energy
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**Pacific Northwest Laboratory
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HOT SEMIWORKS SUMMARY - RUN PX-18

By

J. L. Murray and R. J. Sloat

March 20, 1957

Semiworks Operation
Chemical Development Operation
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HOT SEMIWORKS - RUN PX-18

1.0 INTRODUCTION

Hot Semiworks Run PX-18 was made to compare HA Column decontamination using HAF prepared by either updraft, or downdraft dissolving. Also determined were the effects on IA Column performance of (1) using Versene washed solvent, and (2) of adding Versene to the IAF and IAS. The Purex plant low-acid flowsheet was used during PX-18.

2.0 SUMMARY

No change in HA Column decontamination was noted which could be directly attributed to the change from updraft to downdraft dissolved feed. However, HA Column decontamination factors were increased by a large factor using either types of feed when the scrub section was operated "loaded." For the most part the periods of "loaded" scrub section and high decontamination factors were accompanied by unstable operation and excessive waste losses.

The use of Versene as an additive to the IAF, IAS (0.0013 M) and IOS (0.001 M) produced an increase in IA Column decontamination. The IAF to IAP decontamination factor increased 60 to 70% to 350 as indicated by the in-line analyses; while a single lab analysis indicated a factor of five improvement. It was not possible to determine whether the increased decontamination resulted from the use of Versene in the organic wash solution or from its introduction in the IAF and IAS streams.

Solvent extraction data are summarized in Table 2.1.

3.0 EQUIPMENT AND FLOWSHEET

No equipment changes were made between Runs PX-17 and PX-18. However, the failure of the IOR pump during PX-17 necessitated the use of all fresh carbonate solution (IOS) in the IO Column. An equipment flowsheet is presented in Figure 3.1 and the operating conditions for PX-18 are listed in Table 3.1.

The Purex Plant low-acid flowsheet was used during Run PX-18. The stream flow ratios for this flowsheet were:

HAF:HAX:HAS:HCX::100:366:67:667
IAF:IAX:IAS:IBS:IBX:ICX::75:376:56:60:30:667
2AF:2AX:2AS:2BX::57.5:14.3:7.2:7.2

Compositions of the entering streams followed the HW No. 3 Flowsheet except for the following changes:

HAF - NaNO_2 = 0
HAS - HNO_3 = 2.3 M
IAF - U = 1.8 M, HNO_3 = 1.0 M
IAS - HNO_3 = 2.0 M

Actual flow rates and stream compositions are given in Table 3.2.

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TABIE 2.1

SOLVENT EXTRACTION SUMMARY DATA

Stream	Losses ⁽¹⁾		Log D.F. (HAF = 0.0) ⁽²⁾
	\$ U	\$ Pu	
HAP			2.4
HAN	1.8	2.8	
HCP			2.4
HCW	0.03	0.06	
IAF			2.1 ⁽⁴⁾
IAP			4.0
IAY	0.04	0.8	
IBP	7 x 10 ⁻³		4.1
IBU		1.0	
ICU			4.6
ICW	0.4		4.9
2AF			4.1
2AF			
2AM ⁽³⁾		0.7	6.4
2BF			
2BW		0.15	6.5

Notes:

- (1) Average for the run. See Figures 5.1, 5.2, 5.3, 5.4 for losses at various periods during the run.
- (2) Data are averages for the run based on HAF. Fission product activity was in general > 80% Zr-Nb. See Table 5.1.
- (3) 2AM & Pu losses were based on 2AF. All other losses were based on HAF.
- (4) A Gamma balance on the columns showed the HAF/IAF df to be 2.4.

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TABLE 2.2

PX-18 RUN CHRONOLOGY

Date	Time	Remarks
3-14-56		Charged 124 eight-inch slugs to dissolver.
3-15-56	000 to 0900	Removed aluminum jackets from slugs.
3-15-56	1120	Started 1st cut Metal Dissolution.
3-17-56	0140	Dissolution complete. 894 pounds of uranium dissolved in 2 updraft and 1 downdraft cuts.
3-18-56	1030	Started HA, HC, 1A Columns on "cold" uranium at 10 T/D rates
	1210	Shutdown 1A Column.
	1310	Started "hot" updraft dissolved HAF to HA Column at 14 T/D rates.
3-19-56	0340	Increased HA-HC Columns to 16 T/D rates.
	0740	Reduced HA-HC Columns to 14 T/D rates.
	1245	Started 1A Column at 14 T/D rates.
	1310	Started IBX-IBS-IC Columns at 12 T/D rates.
	1430	Started 2A-2B Columns at 12 T/D rates.
	2125	Started using downdraft dissolved HAF.
3-20-56	0130	Shutdown IBX-IBS-IC Column flows to level out the
	0215	Started IBX-IBS-IC Column flows.
	0300	Shutdown IBX-IBS-IC Column flows to level out the
	0350	Started IBX-IBS-IC Columns at 10 T/D rates.
	0840	Reduced IBS Column pulse amplitude to 0.6 inch
	1120	Increased HA-HC Columns to 16 T/D rates.
	1345	Shutdown HA-HC Columns.
	1745	Reduced 1A Column to 12 T/D rates to level out the
	1900	Increased 1A Column to 14 T/D rates.
	2215	Started using IOS containing 0.001 M Versene.
3-21-56	0100	Increased IBX-IBS-IC Columns to 12 T/D rates.
	0400	Started using 0.0013 M Versene in IAF and IAS.
	1025	Shutdown 1A Column.
	1555	Shutdown IBX-IBS-IC Columns.
	1640	Started IBX-IBS-IC Columns at 12 T/D rates.
	2315	Shutdown IBX-IBS-IC Columns.
	2345	Shutdown 2A-2B Columns.

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TABLE 3.1

COLUMN OPERATING DATA

Column	Packed Height, Ft.	Column Dia., Sch. 40 Pipe, In.	Packing	% Free Area	Plate Spacing, In.	Interface Location	Amplitude, In.	Frequency, Cycles/Min.	Volume (6) Velocity, Gal/Hr/Ft ²
HA(e)	8	3	S.S. Plates	(1)	(1)	Top	1.07	(2)	688
HA(s)	13	4	S.S. Plates (3)	23	2		0.60		327
HC	10	4	S.S. Nozzle Plates (4)	10	4	Bottom	0.50	100	792
IA(e)	8	3	S.S. Plates	(1)	(1)	Top	1.2	56	670
IA(s)	13	4	S.S. Plates (3)	23	2		0.7		329
IBX	18	4	S.S. Plates	23	2	Top	0.8	40	331
IBS	12	1	S.S. Plates	23	2	Top	0.8 & 0.6	42	835
IC	10	4	S.S. Nozzle Plates (4)	10	4	Bottom	0.5	100	735
2A(e)	8	1-1/2	3/8" Fluorothene Raschig Rings			Bottom	0.8	50	315
2A(s)	8	1-1/2	3/8" Fluorothene Raschig Rings						88
2B	10	1	S.S. Plates 21(5)	2		Top	1.0	40	207
IO	16	5	S.S. Nozzle Plates (4)	10	4	Bottom	0.5	70	496
IF	38	4	Alternate 1' layers of 1" S.S. and 1" Fluorothene Raschig Rings.						

Notes:

- (1) "Graded Cartridge." See HW-43074 RD for details.
- (2) See Figure 5.1; HA frequency ranged from 66 to 98 c/m.
- (3) Anti-backmixing cap between feed tee and scrub section; See HW-39517 RD for details.
- (4) Nozzle depth = 0.035 inch.
- (5) Hole diameter of 2B Column plates = 0.06 inch; plates in other columns have 0.125 inch holes.
- (6) Volume velocities given for periods when HA, HC, IA, IO Columns at 14 T/D rates, and IBX, IBS, IC, 2A, 2B Columns at 12 T/D rates.

TABLE 3.2

FLOW RATES AND COMPOSITIONS OF ENTERING STREAMS

Stream	Composition	Flow, ml/min ⁽⁷⁾	
		Actual	Flowsheet
HAF	U = 1.38 M, HNO ₃ = 2.0 M, Pu = 0.088 g/l	403 ⁽¹⁾⁽⁵⁾	420
HAX	30% TBP in Shell E-2342	1535 ⁽⁵⁾	1537
HAS	HNO ₃ = 2.3 M	287 ⁽¹⁾⁽⁵⁾	280
HGX	HNO ₃ = 0.01 M	2880 ⁽¹⁾⁽⁵⁾	2800
IAF	U = 1.78 M, HNO ₃ = 1.2 M, Pu = 0.11 g/l ⁽²⁾	330	315
IAX	30% TBP in Shell E-2342	1600	1580
IAS	HNO ₃ = 2.0 M ⁽³⁾	237 ⁽¹⁾	236
IRX	HNO ₃ = 0.1 M, Fe ⁺⁺ = 0.06 M	104 ⁽¹⁾⁽⁶⁾	108
IBS	30% TBP in Shell E-2342	212	216
IBXF	U = 0.31 M, HNO ₃ = 0.12 M, Pu = 0.017 g/l	1740	1704
ICX	HNO ₃ = 0.01 M	2360 ⁽¹⁾	2400
2AF	HNO ₃ = 6.2 M, Pu = 0.004 g/l to 0.112 g/l	202	206
2AX	30% TBP in Shell E-2342	52	51.6
2AS	HNO ₃ = 0.5 M	26.9 ⁽¹⁾	25.9
2BX	HNO ₃ = 0.01 M	26.2 ⁽¹⁾	25.9
IOF	30% TBP in Shell E-2342	3250	3270
IOS	0.25 M Na ₂ CO ₃ ⁽⁴⁾	1100	1100

Notes:

- (1) Calculated from tank depletion rates; others are from rotameter readings.
- (2) IAF composition included 0.0013 M Versene during last 6 hours of IA Column Operation.
- (3) IAS composition included 0.0013 M Versene during last 6 hours of IA Column Operation.
- (4) IOS composition included 0.001 M Versene from 3-20-56 at 22.5 to end of FX-18.
- (5) All HA-HC Column streams were increased from 14 to 16 T/D rates from 0340 to 0740 on 3-19-56, and from 1120 to 1345 on 3-20-56. The HAF rates during these periods were 468 and 460 ml/min, respectively.
- (6) All IBX-IBS-IC Column rates decreased from 12 to 10 T/D from 0350 on 3-20-56 to 0100 on 3-21-56.
- (7) Actual flows given during periods when HA, HC, IA, IO Columns at 14 T/D rates, and IBX, IBS, IC, 2A, 2B Columns at 12 T/D rates.

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4.0 FEED PREPARATION

4.1 Slug Charging

For Run PX-18, 985 pounds of 8-inch uranium slugs (241 MWD/T, cooled 112 days at the time of charging) were charged to the dissolver. No heel remained in the dissolver from PX-17.

4.2 Jacket Removal

The aluminum slug jackets were removed by the standard procedure of boiling for three hours in a solution initially containing 156 pounds of NaNO_3 and 82 pounds of NaOH . The operation was normal.

4.3 Dissolution

A total of 894 pounds of uranium was dissolved using two different methods. The first 603 pounds of uranium were dissolved in two updraft cuts, followed by a standard downdraft cut which dissolved 291 pounds of uranium. For the first two cuts the acid consumption was 5.44 moles of HNO_3 per mole of uranium dissolved as contrasted with 2.84 moles of HNO_3 per mole of uranium dissolved for the third (downdraft) cut. During the two updraft cuts, cooling water on the dissolver was required to keep the reaction under control during the first half of each cut. Dissolution data are presented in Table 4.1.

4.4 Feed Adjustment

PX-18 dissolver solution was butted to 1.35 molar uranium, 2.0 molar nitric acid in two separate batches. The two updraft cuts comprised the first batch, and the downdraft cut was the second batch. No sodium nitrite was added to either batch of HAF. The HAF was pumped to the HA Column without clarification because of the centrifuge failure and the subsequent plugging of the HAF filter installed to replace the inoperative centrifuges.

5.0 SOLVENT EXTRACTION

5.1 Operation

Between Runs PX-17 and PX-18 all of the columns were flushed with 5 percent nitric acid, filled with their respective "cold" aqueous solutions and the interfaces established with organic. Equilibrium with respect to uranium was established in the HA, HC, and IA Columns by starting up with HAF and IAF made from "cold" uranium, thereby preventing excessive fission product carryover at "hot" startup. Attempts to "load" the HA Column scrub section at 14 T/D rates by varying the frequency were largely unsuccessful because floods developed in the extraction section. Increasing the throughput rate to 16 T/D in the HA and HC Columns caused the scrub section to "load" immediately, and though the operation was not stable the scrub section remained "loaded" when the rates were reduced to 14 T/D four hours later. Changes in HA Column frequency kept

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TABLE 4.1

DISSOLUTION DATA

	Cut Number		
	1	2	3
Source of acid	Fresh	Fresh	Fresh
Initial acid concentration, % ⁽²⁾	60	60	48.5
Weight of 100% HNO ₃ , Lb.	438	438	267
Uranium charged, Lb.	985	--	--
Reflux water addition, Lb.	0	0	55
Reflux addition duration, Hr.	--	--	1
Air addition rate, SCFM	0	0	4.5
Time at boiling, Hr.	10	8	9
Final specific gravity (boiling)	1.75	1.76	1.75
Final vapor temperature, °C.	--	--	98.7
Uranium dissolved, Lb.		603	291
Dissolver heel, Lb.		382	91
Acid consumed ⁽¹⁾		5.44	2.84
Dissolver solution composition			
Uranium M		2.64	2.05
Nitric Acid M		0.12	1.28

Notes:

- (1) Moles of HNO₃ per mole of uranium dissolved.
 (2) Includes reflux water addition.

the scrub section loaded for ten hours of metastable operation, but then it "unloaded" again with just four hours of updraft dissolved HAF remaining. At 14 T/D rates and a frequency of 88 cycles per minute, the four hours of updraft feed were used to establish the HA Column decontamination factor for that type of feed with stable column operation. Most of the downdraft dissolved HAF was processed with the scrub section "unloaded", under the same operating conditions as the last four hours of updraft feed so their respective decontamination factors could be compared. With six hours of feed remaining the scrub section was "loaded" by increasing the HA Column frequency, and was maintained "loaded" for the last three hours at 16 T/D rates.

IA Column operation at 14 T/D rates was smooth except for a 7-hour period of cyclic flooding in the middle of the run. After the HA and HC Columns were shutdown, the remaining HCP was butted into IAF specifications in a batch to give 17 hours of IAF with the same composition. The IA Column decontamination factor was established during the first 5 hours of operation with this feed, and then 0.001 M Versene was added to the IOS for the remainder of the run. The next 6 hours of IA Column operation compared the established DF against that attained when the IAX used had been washed with IOS containing Versene. The last 6 hours of operation were made with 0.0013 M Versene in the IAF and IAS to note the effect of these additions upon the established decontamination factor.

Unstable operation in the IBS Column during the first 15 hours forced a reduction in the IB-IC Column rates from 12 to 10 T/D. Even then the IBS Column continued to flood or be otherwise upset for an additional 7 hours, until after the IBS pulse amplitude was reduced from 0.8 to 0.6 inch. When the rates were increased to 12 T/D (Purex Plant equivalent rate) because of IBXF tank limitations, the IBS Column operation soon became unstable again and continued that way for most of the rest of the run. See Figure 5-3.

The 2A and 2B Columns operated smoothly at 12 T/D rates for the entire run. The 2AF probably contained some organic during most of the run, as a result of the poor IBS Column operation.

5.2 Data

The principal solvent extraction data obtained from in-line instruments and laboratory analyses are presented graphically in Figures 5-1, 5-2, 5-3, and 5-4. Other laboratory data are given in Table 5-1.

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TABLE 5.1

SOLVENT EXTRACTION RESULTS

Stream	Flow, ml/min PK-18	Flow, ml/min Flowsheet	Pu, g/L	U, g/L	HNO ₃ , g/L	Q _{max} , μc/L	Zr-Nb	Ru	Ce	I-131	Zr	Nb
*HAF (Updraft)	403	420	0.088	328	126	2.71x10 ⁷	77	14	9			
*HAF (Downdraft)	1586	1600	0.021	81.8	6.9	3.38x10 ⁷	88	12				
HAF (Updraft)						2.49x10 ⁴		13	1		71	15
HAF (Downdraft)						2.76x10 ⁴		12	2		66	20
HAW	650	651	1.6x10 ⁻³	3.8	133							
HCP	2890	2843	0.013	41.2	3.1	1.6x10 ⁴						
*HPC		310										
*IAF	330	315	0.11	425	75	3.2x10 ⁵		4			48	48
IAP	1694	1667	0.02	85.2	7	800		6		5	21	68
IAN	510	504	5.7x10 ⁻⁴	0.11	95.4							
IBP	116	113	0.28	0.071	92.6	9040						
*IBXF	1740	1704	0.017	73.2	7.6	292	55	20		25		
IBU	1718	1689	1.9x10 ⁻⁴	71.6	162		90	10				
ICU	2430	2437	1.3x10 ⁻³	52.2	14	67						
*ICU				53.3		951						
*2AF	202	206	.004-.112	0.12	388	3940	92	8		11		
2AP	52	51.6	.012-.85		7.9	90	80	9				
2AN	230	233	10-6.4x10 ⁻³		352							
2BP	26	25.9	.076-2.1			155						
*2BP			2.83	4.3	389	313	77	15		8		
*10F	3250	3270	8.3x10 ⁻⁶	0.38		70-500						
100	3400	3270				23	27	13	1	59		
*100-T						38						

Notes: *Tank Samples, others are stream samples.

*These lab results are believed to be non-representative. From gamma balance, IAP gamma = $1.5 \times 10^5 \mu\text{c/l}$, and IBP gamma = $860 \mu\text{c/l}$.

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FIGURE 5.1

HA-HC CYCLE DATA

FIGURE 5.2

1A COLUMN DATA

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FIGURE 5.3

1BX-1BS-1C COLUMN DATA

- 1AF/1BU In-line DF
- 1AF/1BU Lab. DF
- ▲ 1AF/1BXP Lab. DF
- ▼ 1AF/1BXP Lab. DF
- 1AF/1BP Lab. DF
- 1AF/1CU Lab. DF

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FIGURE 5.4

2A-2B COLUMN DATA

5.3 Discussion

Codecontamination Cycle. HA Column decontamination factors of 2.5 (dF) were achieved with both updraft and downdraft dissolved HAF during portions of PX-18 with stable column operation (unloaded scrub section). During portions of the run when the HA Column scrub section was operated "loaded" the dF's were 2.7 to 3.0 with both types of feed. Poor dF's during the startup period caused an overall dF of 2.4. The decontamination factor was apparently unaffected by the change from updraft to downdraft dissolved feed. In general the HA Column waste losses were high during periods of "loaded" scrub section operation, and low during periods of stable operation. Uranium losses ranged from 0.1 percent to 6.1 percent and plutonium losses from 0.1 percent to 9.8 percent. HAF to HCP dF's were slightly lower than HA Column dF's, possibly due to an occasional aqueous carryover from the HA Column that could not be sampled in the HAF.

Partition Cycle. On Figure 5.2 it can be seen that both the laboratory and in-line IA Column DF's increased when 0.0013 M Versene was added to the IAF and IAS. No immediate change in decontamination was observed when 0.001 M Versene was added to the IOS prior to the additions to the IAF and IAS. However, effects of changes in solvent treatment procedure would take longer to produce changes in the performance of the primary "A" type columns. Consequently it is not known whether the Versene addition to the IAF and IAS or the Versene in the IOS produced the increased IA Column decontamination. (IAF to IAP in-line DF increased from 25 to 37; IAF to IAP lab (one analysis) DF increased from 70 to 350.)

IAW waste losses were low except during the 7-hour period of cyclic flooding shown on Figure 5.2. During steady state periods U losses were 0.002 percent to 0.2 percent and Pu losses were 0.02 percent to 3 percent. The average arithmetic decontamination factor across the IB Column was 5.3 for the IBU and 1.6 for the IBP. The average uranium DF for the run was 4.86×10^4 (dF = 4.9).

Final Pu Cycle. On Figure 5.4, the 2A and 2B Column laboratory and in-line data are in good agreement. These columns achieved an average dF of 2.4, producing an overall plutonium dF of 6.5 for the run. Pu losses in the 2A Column averaged 0.7 percent discounting one result of 4.7 percent in an organic 2AW sample. The 0.7 percent Pu loss is not considered excessive since the Hot Semiworks 2A Column extraction section is only 8 feet in length.

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6.0 SOLVENT RECOVERY

In-line and laboratory gamma analyses for the solvent recovery system are shown on Figure 6.1. IOO-L and IOO-T laboratory gammas are lower than their respective in-line gammas by a factor of 10-100. In the case of the IOO-T this very poor agreement between the two sets of data might be due in part to the fact that a high gamma cell background made it impossible to detect less than $300 \mu\text{c}/\text{l}$ on the in-line monitor. The average IOF/IOO DF was 12 ($\text{dF} = 1.1$), and the amount of HAF uranium and plutonium in the IOF was 0.9 percent and 0.08 percent, respectively. The addition of 0.001 M Versene to the IOS had no pronounced effect on IO Column decontamination.

7.0 ACID RECOVERY

During PX-18 no major effort was made to accumulate acid recovery data because of the emphasis placed on the solvent extraction portion of the process and because of the absence of sufficiently long periods of operation at uniform conditions. Neither supplemental NaNO_2 nor water was added to the No. 1 Acid Concentrator.

The IWW gamma averaged $8 \times 10^7 \mu\text{c}/\text{l}$, the 2WW $3 \times 10^6 \mu\text{c}/\text{l}$, and the recovered acid $1.8 \times 10^3 \mu\text{c}/\text{l}$ during PX-18. The concentration of the recovered acid was 9 molar.

FIGURE 6.1

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SOLVENT RECOVERY DATA

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