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TRACE ELEMENTS RETAINED IN WASHED NUCLEAR FUEL REPROCESSING SOLVENTS

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ABSTRACT

Analysis of purified TBP extractant from solvent extraction processes at Savannah River Plant showed several stable elements and several long-lived radioisotopes. Stable elements Al, Na, Br, Ce, Hg, and Sm are found in trace quantities in the solvent. The only stable metallic element consistently found in the solvent was Al, with a concentration which varies from about 30 ppm to about 150 ppm. The halogens Br and Cl appear to be found in the solvent systems as organo halides. Radionuclides found were principally ^{106}Ru, ^{129}I, ^{3}H, ^{235}U, and ^{239}Pu. The ^{129}I concentration was about 1 ppm in the first solvent extraction cycle of each facility. In the other cycles, ^{129}I concentration varied from about 0.1 to 0.5 ppm. Both ^{129}I and ^{3}H appear to be in the organic solvent as a result of exchange with hydrogen.

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INTRODUCTION

The Savannah River Plant (SRP) has two main chemical separation processes that use tri-n-butyl phosphate (TBP) as an extractant, Purex and HM. (Other processes are used infrequently.) The Purex process uses 30% TBP in normal paraffin hydrocarbons to separate, purify, and recover uranium and plutonium from irradiated natural or depleted uranium reactor targets. The HM process uses both 30% TBP and 7.5% TBP to recover enriched uranium and neptunium from enriched U-Al alloy reactor fuels. Besides the Purex and HM processes, SRP also recovers plutonium, americium, and curium from Pu-Al targets and ^{233}U from thorium targets.

Each process has three extraction cycles. Solvent from each of these six cycles is purified separately for reuse. This purification is performed by contacting the solvent with acidic or alkaline washing solution. In this report, trace elements retained in washed nuclear fuel processed solvents will be discussed.

BACKGROUND

At the Savannah River Plant, as elsewhere throughout the country, the original Purex process encountered periods of poor operation and exhibited abnormal behavior of fission products. The general concepts, equipment, and performance of the original Purex process have been documented at Geneva[1,2] and Brussels.[3] These problems were attributed to the effects of degradation of the organic solvent by both radiolytic and chemical attack. Solvent degradation products promote emulsions which vitally affect the physical performance of the liquid-liquid contactors.

During the first six years of operation[4] at SRP, the complete inventory of solvent in the first cycle had to be discarded on three occasions because of the poor product decontamination and physical problems. As a result of studies of solvent degradation at many sites[5-16] and work on diluents[17-23] which showed that straight-chain compounds had enhanced stability, SRP changed to a normal paraffin fraction (largely C_{12}) for the first solvent extraction cycle[24] of the normal Purex plant.

Orth and Olcott[24] analyzed SRP solvent extraction operations for a period of years before 1963. The analysis considered combinations of plant variables including two different mixer-settler installations that varied widely in contactor size and in solvent residence times and effects of two diluents: 1) ultrasene (from Atlantic Refining Company), which was used in all solvent extraction cycles from plant startup until 1961, and 2) adakane (from Archer-Daniels-Midland Company), which was substituted in the first extraction cycle of the normal Purex plant. Since 1961, only sufficient solvent (diluent or extractant) has been added to the Purex plant first cycle to make up for losses. The change-out half-life due to losses has remained relatively constant at about 40.5 months. With this change-out half-life, there is the possibility of long-term buildup of the degradation products which have lower vapor pressures or lower solubilities. The average life of the HM first cycle solvent, however, was only 24 months at the beginning of the study. With the differences in average age of the solvent, and the different TBP concentrations, the study of solvent impurities was begun to determine if differences could be detected. Gross differences in impurity levels may point to long-term buildup of ligands which may need to be removed by different washing techniques.

SAMPLING AND EXPERIMENTAL PROCEDURE

Process solvent samples from the washed solvent hold tanks of each of the six solvent extraction cycles (Table 1) were taken on a random basis. Samples were stored for up to 6 months to allow any short-lived fission products present to decay.

Trace stable elements (Al, Na, Br, Cl, Hg, and Sm) and ^{129}I were determined by neutron activation analysis with a ^{252}Cf source. Low-level gamma ray spectrometry was used to measure ^{106}Ru. The residual fissile material content of the washed solvent was measured by neutron activation followed by delayed neutron counting. No discrimination between ^{235}U and ^{239}Pu is possible with this technique, and total fissile content is reported as equivalents of ^{235}U. (^{238}U is not measured by this method.)

Tritium was determined by beta-scintillation counting. No correction was made for ^{106}Ru contribution; therefore, only the upper limit values are reported for ^{3}H.

TABLE 1

Process Sampling

Process Sampled	*Cycle Sampled*	*Tank Sampled*
Enriched U/Np (HM Process)	First	H-904
	Second Uranium	H-902
	Second Neptunium	H-906
Depleted U/Pu (Purex Process)	First	F-14.7
	Second Uranium	F-904
	Second Plutonium	F-906

PUREX PROCESS DESCRIPTION

Major operations in the SRP F-Area Purex process are shown schematically in Figure 1. Irradiated target slugs (Al-jacketed U) are received from the reactor areas and charged to the dissolvers. The Al jackets are removed with NaOH and $NaNO_3$, and the dejacketed slugs are dissolved in 50% nitric acid. The resulting solution primarily contains ^{238}U with smaller amounts of ^{235}U, plutonium, and fission, and neutron activation products. This dissolver solution is processed through a feed clarification step to remove the Al activation product Si.

The removal is accomplished by adding gelatin and simmering the solution, thus forming a gelatin-silica polymer, which is removed by centrifugation. The silica is removed to prevent formation of solids, which cause emulsions in the solvent extraction equipment during later processing. The clarified solution is chemically adjusted to ensure the proper acid and uranium concentration. The adjusted solution is then fed to the first solvent extraction cycle where uranium and plutonium are extracted into the solvent in the 1A Centrifugal Contactor. About 95% of the fission products remain in the aqueous phase and are transferred to high-activity waste.

Plutonium is separated from uranium in Mixer-Settler 1B by reduction with a ferrous sulfamate solution that strips it from the solvent. The plutonium is then transferred to the second plutonium solvent extraction cycle. In this cycle, plutonium is further decontaminated and concentrated by extracting it from the aqueous feed into solvent in Mixer-Settler 2A. Plutonium is stripped from the solvent in Mixer-Settler 2B. The resulting aqueous stream is transferred to the plutonium finishing line (JB-Line) where plutonium fluoride is precipitated and reduced by reaction with calcium to produce plutonium metal as a disk or button.

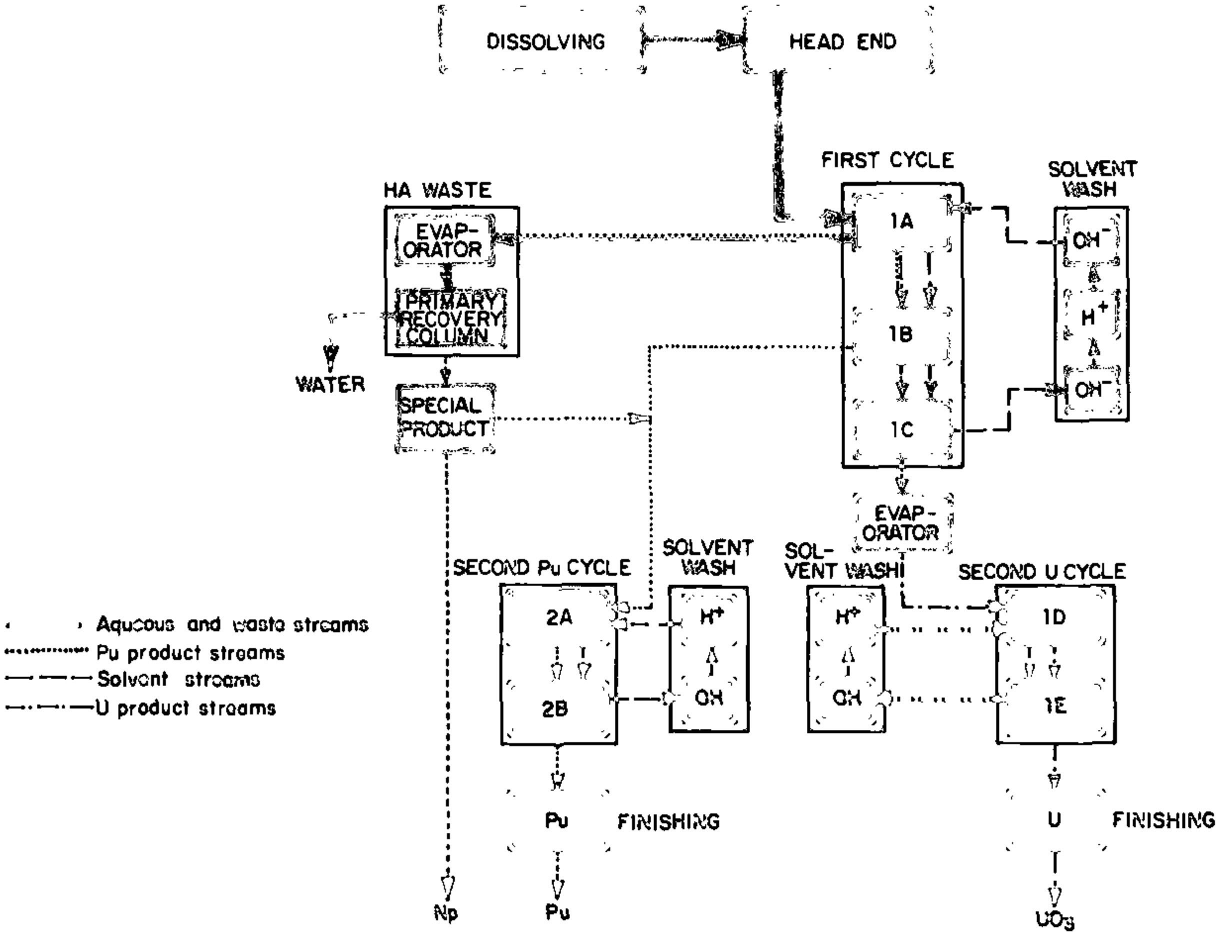

FIGURE 1. Purex Process

Uranium, which remained in the solvent in Mixer-Settler 1B, is stripped from the solvent in Mixer-Settler 1C. The aqueous uranium stream is concentrated in a continuous evaporator before being fed to the second uranium solvent extraction cycle. The concentrate is chemically adjusted, and the uranium is further decontaminated by extracting into solvent in Mixer-Settler 1D and by stripping from the solvent in Mixer-Settler 1E. The aqueous stream leaving Mixer-Settler 1E is transferred to outside uranium facilities (A-Line) where uranium is processed to uranium trioxide.

Each of the three solvent extraction cycles has a separate and continuous solvent system (Figure 1). The solvent recovery system removes radioactivity and solvent degradation products, maintains acceptable quality, and returns washed solvent to the respective extraction cycle. Operating experience has shown that the most efficient washing cycle is alternating alkaline (0.19 to 0.47N Na_2CO_3) and acid (0.03 to 0.12N HNO_3) washes. All solvent is 30 volume % TBP and 70 volume % n-paraffins (normally dodecane), with a density of about 0.8 g/cc.

The first-cycle solvent system is contained completely within the hot and warm canyons because of gamma radiation in the unwashed solvent and aqueous wash solutions. Unwashed solvent leaves Mixer-Settler 1C and flows to the first-stage washer where it is washed with sodium carbonate. After gravity separation from the wash solution, the solvent flows to the second-stage washer, is washed with dilute nitric acid, is separated, and is pumped to the third-stage washer. In this stage, the solvent is washed a second time with sodium carbonate and transferred to first cycle solvent feed tank. If necessary, TBP or diluent is added at this point to maintain proper volume and concentrations. Solvent is fed to first cycle as 1AX and 1BS streams and exits the cycle as the 1CW stream returning to the first-stage washer. When first cycle is down, the solvent may be circulated to maintain quality by transferring from the feed tank to the first-stage washer.

Solvent recovery facilities for the second uranium cycle are behind shielded walls in Building 211-F (outside facilities). (Radioactivity in the unwashed solvent from second uranium cycle is low enough to make this practical.) Unwashed solvent from the second uranium cycle leaves Mixer-Settler 1E as the 1EW stream and is transferred continuously to the first-stage alkaline washer. Solvent is washed, separated, and transferred to the second-stage acid washer where it is again washed, separated, and transferred to a storage tank. From the storage tank, it is fed to Mixer-Settler 1D as the 1DX stream. The solvent leaves Mixer-Settler 1D as the 1DU stream and goes to Mixer-Settler 1E.

Part of the solvent recovery system for the second plutonium cycle is in Building 221-F warm canyon, and part of it is behind shielded walls in Building 211-F (outside facilities). Solvent from the plutonium cycle leaves as the 2BW stream. After it flows through a decanter for removal of entrained aqueous solution, it is transferred continuously to the first-stage alkaline washer in the warm canyon. (This washer is inside Building 221-F because plutonium from a process upset could be present in the 2BW. Under these conditions, plutonium could accumulate in the alkaline washer and result in high levels of alpha activity and possibly a nuclear safety hazard.) After the solvent is washed and separated in the first-stage alkaline washer, the solvent is transferred to the second-stage acid washer in Building 211-F where it is again washed and separated. It is then received in a storage tank and fed to second plutonium cycle as the 2AX stream. When the cycle is down, the solvent may be circulated to maintain quality by transferring it from the storage tank to the first stage washer. The aqueous wash solutions in all solvent washers are replaced at regular intervals.

HM PROCESS DESCRIPTION

The HM Process is a modification of the basic Purex process. The two major modifications are use of a lower concentration of TBP (7.5% instead of 30%) and the use of a salting agent [$Al(NO_3)_3$] instead of HNO_3. The lower TBP concentration maintains a lower concentration of ^{235}U in the process for nuclear safety control because enriched ^{235}U is processed. Because the fuel being dissolved is an Al alloy jacketed with Al, total dissolution with Hg^{2+} and HNO_3 is used instead of selective dissolution. Due to this large amount of Al^{3+} in solution, $Al(NO_3)_3$ is used as the salting agent. With these two major modifications, the system is operated very similarly to the basic Purex process. The solvent wash sequence of the HM process first-cycle solvent is carbonate, acid, acid instead of carbonate, acid, carbonate as in the Purex process.

HM-PLANT SOLVENT REPLACEMENT

The Purex plant has used a 30% TBP extractant concentration in the adakane solvent over the last 17 years. Before 1973, the HM-plant first cycle solvent used a variable concentration of TBP extractant; its concentration has ranged from 3.5% to 30% depending on the specific material being processed (see Table 2). Since 1973, the HM plant has used a stable 7.5% TBP extractant concentration. The bulk of the first-cycle diluent at the time this study began had seen less than a three-year exposure to the process; the bulk of the extractant had been in service less than 24 months. The average age of the HM first-cycle solvent then is about 60% of the average age of the Purex first-cycle solvent. The average age of the other two cycles is about the same in both areas.

TABLE 2

Solvent Changes in the HM First Cycle

Dates	*% TBP in Solvent*
10/65 to 11/66	30
2/66 to 3/68	3.5
4/68 to 7/68	30
8/68 to 6/69	3.5
7/69 to 11/69	30
12/69 to 10/72	3.5
1/72 to 2/73	7.5
3/73 to 10/73	3.5
11/73 to present (1978)	7.5

RESULTS AND DISCUSSION

Results are shown in Tables 3 and 4. The stable elements Al, and Na and the long-lived radioisotopes ^{3}H, ^{129}I, and ^{106}Ru were present in the washed solvent from all six solvent extraction cycles. Cl and Br were present in three Purex cycles and the second Np cycle of the HM system; Hg was present in the HM first and second Np cycles and in the Purex second cycle; Sm was present in the first cycle solvents from both Purex and HM.

Since specifications exist for Al as a contaminant in the finished products, the behavior of Al in the solvent extraction cycles (Table 5) is of special interest. In general, Al concentrations of the cycles with 7.5% TBP (first-cycle and second-uranium cycle, HM process) is about 30 to 40 ppm. Thirty % TBP is used in the second Np cycle in the HM process, and its Al concentration is about 140 ppm. Thus, the increase in the Al concentration is roughly equivalent to the increase in TBP concentration. However, in the Purex plant, the concentration of TBP is 30%; but the Al concentration of all three cycles is about 60 to 80 ppm. The Al concentration of the feed to these cycles is only <10 to about 10^3 ppm in contrast to >1M Al in the HM first-cycle feed.

The mechanism of transport of the Al through the cycles is not understood, but since Al is apparently transported through the solvent system, this is a possible source of low-level Al contamination in the final products. A more complete study of the behavior of Al in the solvent system should be attempted. This study should determine both the mechanism of Al retention in the solvent and the washing methods to remove the Al.

TABLE 3

Stable Elements Retained in Washed Process Solvents

			Element, ppm (μg/mL)						
Area	*Cycle*	*Date*	*Al*	*Na*	*Hg*	*Br*	*Cl*	*Sm*	*Mn*
F	First	2-6-76	70	12	-[a]	41 ±4	-	-	-
		4-9-76	59	6	-	42 ±4	-	-	-
		6-11-76	55	9.7	-	1.4	-	-	-
		7-6-76	68	1.5	-	-	-	-	-
		7-28-76	(160?)	5	-	0.6	-	-	-
		8-20-76	(140?)	3.9	-	0.6	-	-	-
		9-15-76	71	10	-	-	-	-	-
		9-15-76	71	5	-	-	-[b]	-	-
		1-7-77	70	6.7	-	-	-	-	-
		1-14-77	49	9.7	-	-	-	-	-
		1-21-77	68	6.9	-	-	-	-	-
			80.1 ±35.6[c] (64.6 ±8.1)[d]	6.95 ±3.12	-	7.78 ±16.68[c] (?)[d]	-[b]	-	-
H	First	10-10-75	33	0.5	4.7	-	-	-	-
		2-11-76	34	0.7	5.7	-	-	-	-
		12-22-76	26	-	-	-	-	1.6	-
		12-29-76	26	2.5	-	-	-	-	-
		1-5-77	18	2.5	-	-	-	1.2	-
		6-2-77	44	2.2	-	-	-	-	0.08
			30.2 ±8.9	1.40 ±1.12[c] (1.68 ±1.00)[d]	1.73 ±2.70 (5.2 ±0.7)[d]	-	-	<2	<0.1
F	Second Uranium	2-25-76	59	2.0	10.2	1.8	36	-[a]	-
		4-7-76	62	2.0	-	1.8	24	1.3	-
		12-17-76	70	2.2	23	2.0	23	-	-
		12-22-76	73	4.7	-	1.6	(-)	-	-
		12-29-76	71	3.5	-	1.5	(-)	-	0.05
		1-05-77	74	3.1	-	1.4	(-)	-	-
		7-21-76	(130)	2.0	-	1.4	25	-	-
		9-15-76	(140)	1.4	3.6	1.5	20	0.02	-
			84.9 ±31.5[c] (68.2 ±6.2)[d]	2.6 ±1.1	4.6 ±8.2	1.6 ±0.2	16.0 ±14.0[c] (25.6 ±6.11)[d]	<1	<0.1
H	Second Uranium	12-10-75	38	1.6	-	-	-	-	-
		3-24-76	41	1.2	-	-	-	-	-
		6-02-77	40	3.4	-	0.67	-	-	-
			39.7 ±1.53	2.1 ±1.2	-	<0.7	-	-	-
F	Second Plutonium	2-18-76	63	5.0	-[a]	1.6	17	-	-
		4-7-76	70	1.5	-	1.4	31	-	-
		7-2-76	(160)	(34?)	-	1.4	24	-	-
		9-15-76	(140)	1.7	-	1.1	21	-	-
		12-22-76	62	2.5	-	1.4	-	-	-
		12-15-76	(-)	2.5	-	1.1	17	-	-
		12-29-76	78	2.3	-	1.8	24	-	-
		1-05-77	65	2.9	-	-	-	-	-
		1-19-77	75	2.3	-	1.4	28	-	-
		1-19-77	(-)	3.3	-	1.2	26	-	-
			71.3 ±50.6[c] (68.8 ±6.6)[d]	5.8 ±10.0[c] (2.7 ±1.0)[c]	- -	1.24 ±0.49[c] (1.38 ±0.23)[d]	18.8 ±10.8[c] (23.5 ±5.0)[d]	-	-
H	Second Plutonium	9-10-75	138	2.9	3.6	-	-	-	-
		2-11-76	130	2.2	2.9	0.7	17.7	-	-
		6-2-77	162	3.8	-	0.6	31	-	0.07
			143 ±17	3.0 ±0.8	2.2 ±1.9[c] (3.2 ±0.5)[d]	0.4 ±0.4[c] (0.6 ±0.07)[d]	16 ±16[c] (24 ±9.4)[d]	-	<0.1

---

a. Not detected.

b. Detected but only at the minimum level of detection.

c. All data included in average.

d. Suspect data not included in average.

TABLE 4

Long-Lived Radioactive Elements Retained in Washed Process Solvent

Area	*Cycle*	*Date*	^{129}I, *ppm*	$^{239}Pu + ^{235}U$, *ppm*	^{3}T, *dis/(min mL)*	^{106}Ru, *dis/(min mL)*
H	First	10-10-75[a]	1.2 ±0.1	6	$\le 2.6 \times 10^5$	$1.500 \pm 0.089 \times 10^3$
		2-11-76[b]	1.0 ±0.1	9	$\le 5.9 \times 10^5$	$8.100 \pm 0.200 \times 10^3$
		6-02-77	0.9 ±0.07	3	$\le 2.1 \times 10^5$	-[d]
F	First	2-06-76	1.3 ±0.1	10	$\le 7.0 \times 10^5$	$17.30 \pm 0.23 \times 10^3$
		4-09-76	1.1 ±0.1	6	$\le 7.6 \times 10^5$	$18.20 \pm 0.23 \times 10^3$
		9-10-76	-	-	$\le 7.3 \times 10^5$	-
H	Second Uranium	12-10-75[a]	0.08 ±0.01	7	$\le 4.6 \times 10^3$	-
		3-24-76[c]	0.08 ±0.01	10	$\le 5.6 \times 10^3$	$1.60 \pm 0.26 \times 10^2$
		6-02-77	0.08 ±0.02	3	$\le 7.0 \times 10^2$	
F	Second Uranium	2-25-76	0.11 ±0.02	20	$\le 2.0 \times 10^4$	$4.70 \pm 0.39 \times 10^2$
		4-07-76	0.12 ±0.02	20	$\le 2.6 \times 10^4$	$5.12 \pm 0.40 \times 10^2$
		8-25-76	-	-	$\le 4.1 \times 10^4$	-
H	Second Neptunium	9-10-75[a]	0.14 ±0.02	10	$\le 1.6 \times 10^4$	$2.95 \pm 0.38 \times 10^2$
		2-11-76[b]	0.17 ±0.02	3	$\le 1.3 \times 10^5$	$2.59 \pm 0.65 \times 10^2$
		6-02-77	0.12 ±0.03	3	$\le 1.1 \times 10^4$	-
F	Second Plutonium	2-18-76	0.37 ±0.03	8	$\le 2.6 \times 10^4$	$2.62 \pm 0.31 \times 10^2$
		4-07-76	0.42 ±0.04	30	$\le 4.6 \times 10^5$	$2.12 \pm 0.28 \times 10^2$
		8.25-76	-	-	$\le 3.5 \times 10^4$	-

---

a. 3% ^{235}U processed.

b. 39% ^{235}U processed.

c. 60% ^{235}U processed.

d. Not detected.

TABLE 5

Trace Elements Aluminum and Sodium in Process Solvent

Area	*Cycle*	*Solvent*	*Salting Agent*	*Average Al, ppm*	*Average Na, ppm*
F	First	30% TBP	HNO_3	64.6 ±8.1	6.95 ±3.12[a]
	Second Uranium			68.2 ±6.2	2.6 ±1.1
	Second Plutonium			68.8 ±6.6	2.7 ±1.0
H	Second Neptunium			143.0 ±17	3.0 ±0.8
				86.2 ±12.1	2.8 ±1.2
H	First	7.5%	$Al(NO_3)_3$	30.2 ±8.9	1.4 ±1.1
	Second Uranium		HNO_3	39.5 ±2.1	2.1 ±1.2
				34.8 ±9.1	1.8 ±1.6

---

a. This solvent is high in Na due to entrainment of Na_2CO_3; the other solvents are washed with HNO_3 before transfer to the solvent hold tanks.

With the exception of the Purex first cycle, the same trend found with Al is also found with Na; i.e., the Na content of the 30% TBP solvent is about twice that of the 7.5% TBP solvent. The Purex first-cycle solvent is an exception because the last washer contains an Na_2CO_3 solution; sodium carbonate is entrained in this solvent. In the other solvent wash system, the solvent is washed with Na_2CO_3 and then HNO_3 before storage for recycle into the solvent extraction system. The entrained Na_2CO_3 is washed out during the acid wash. Sodium, however, is not a problem as an impurity in the final products.

Mercury is used as a catalyst to aid in Al dissolution in the HM process. The presence of Hg in the first cycle HM solvent was therefore to be expected. However, the Hg is apparently stripped with the Np because Hg is found in the second Np cycle solvent but not in the second U cycle solvent. In the Purex system, however, Hg was found only in the second U cycle solvent. Hg was found in only three of the eight samples pulled.

The behavior of tritium is also of interest in the various cycles. Although no correction was made for possible ^{106}Ru contribution to the beta scintillation counting, the ^{106}Ru concentration was typically two orders of magnitude less than the measured ^{3}H concentrations except for the second uranium cycles. Therefore, even though only upper limit values are given, the values cannot be more than 10% high for the first-cycle solvent of both processes nor more than 10% for the second Np and Pu cycles. For the second uranium cycles, the error may be as high as 25%.

The overall mechanism of transport of tritium through the cycles appears to be entrainment. Most of the entrained tritium is then removed during the separation of the transuranium actinide from uranium. This mechanism would account for the lower amounts of tritium in the second uranium cycles of both systems (Table 4).

An attempt was also made to determine if the tritium remaining in the washed solvents was basically entrained tritium or exchanged tritium. Solvent samples were counted for ^{3}H before and after a nitric acid wash; results are shown in Table 6. Because very little of the ^{3}H beta activity could not be removed by successive washing, the tritium must be assumed to be retained in the solvent as a result of hydrogen exchange.

Halogens in the cycles are of interest from two standpoints: 1) they are stainless steel corrosion agents, and 2) ^{129}I is a long-lived fission product of environmental interest. The three halogens (Cl, Br, I) detected in the system may be associated with organic molecules and not simple entrained halogen. The first cycles of both processes appear almost devoid of Br and Cl. This is perhaps a result of the higher radiation of the first cycles leading to exchange of halogens in the natural process solvents. The second cycles do appear to contain both Br and Cl, with Cl about a factor of 10 more concentrated than Br.

The isotope ^{129}I has its largest concentration in the first cycles, as expected. In general, the second-cycle concentrations are about a factor of 2 to 10 less than the first cycles.

Fissile isotopes, ^{239}Pu and ^{235}U, were uniformally low at 3 to 30 ppm.

The radioisotope ^{106}Ru followed the expected trend with the first cycles being a factor of 10 more concentrated than the second cycles.

TABLE 6

Tritium Washable from Process Solvents

		^{3}H, *dis/(min mL)*[a]		*Nonexchangeable*
Area	*Cycle*	*Before Wash*	*After Wash*	^{3}H, *%*
F	First	1.78×10^{5}	1.72×10^{5}	96.6
H	First	1.03×10^{5}	9.39×10^{4}	91.2
F	Second Uranium	5.58×10^{3}	5.33×10^{3}	95.5
H	Second Uranium	4.20×10^{2}	2.34×10^{2}	55.7
F	Second Plutonium	2.33×10^{4}	2.19×10^{4}	94.0
H	Second Neptunium	2.41×10^{4}	1.60×10^{4}	66.4

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[a] Average of a minimum of eight samples.

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