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Demonstration of the TRUEX Process for Partitioning of Actinides from Actual ICPP Tank Waste Using Centrifugal Contactors in a Shielded Cell Facility

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ABSTRACT

The TRUEX process is being evaluated at the Idaho Chemical Processing Plant (ICPP) for the separation of the actinides from acidic radioactive wastes stored at the ICPP. These efforts have culminated in a recent demonstration of the TRUEX process with actual tank waste. A continuous countercurrent flowsheet test was successfully completed at the ICPP using waste from tank WM-183. This demonstration was performed using 24 stages of 2-cm diameter centrifugal contactors installed in the shielded hot cell at the ICPP Remote Analytical Laboratory. The flowsheet tested consisted of eight extraction stages, five scrub stages, six strip stages, three solvent wash stages, and two acid rinse stages. It was determined that a centrifugal contactor stage in the scrub section was not operational during testing. As a result, the scrub feed (aqueous) solution followed the solvent into the strip section, effectively eliminating the scrub section in the flowsheet.

An overall removal efficiency of 99.97% was obtained for the actinides. As a result, the activity of the actinides was reduced from 457 nCi/g in the feed to 0.12 nCi/g in the aqueous raffinate, which is well below the NRC Class A LLW requirement of 10 nCi/g for non-TRU waste. The 0.04 M HEDPA strip section back-extracted 99.9998% of the actinide activity from the TRUEX solvent. Removal efficiencies of >99.90%, 99.96%, 99.98%, >98.89%, 93.3%, and 89% were obtained for ^{241}Am , ^{238}Pu , ^{239}Pu , ^{235}U , ^{238}U , and ^{99}Tc , respectively. Iron was partially extracted by the TRUEX solvent, resulting in 23% of the Fe exiting in the strip product. Mercury was also extracted by the TRUEX solvent (73%) and stripped from the solvent in the 0.25 M Na_2CO_3 wash section. Only 1.4% of the Hg exited with the high-activity waste strip product.

Data/results of this testing have been independently reviewed by personnel from Argonne National Laboratory.

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Demonstration of the TRUEX Process for Partitioning of Actinides from Actual ICPP Tank Waste Using Centrifugal Contactors in a Shielded Cell Facility

INTRODUCTION

The Idaho Chemical Processing Plant (ICPP), located at the Idaho National Engineering Laboratory (INEL), formerly reprocessed spent nuclear fuel to recover fissionable uranium. The radioactive raffinates from the solvent extraction uranium recovery processes were converted to granular solids (calcine) in a high temperature fluidized bed. During the course of reprocessing, a secondary waste stream, liquid sodium-bearing waste (SBW), was also generated primarily from equipment decontamination between campaigns and solvent wash activities. This SBW cannot be directly calcined due to the high sodium content and has historically been blended with reprocessing raffinates or non-radioactive aluminum nitrate prior to calcination. Fuel reprocessing activities are no longer being performed at the ICPP, thereby eliminating the option of waste blending to deplete the SBW inventory. Currently, approximately 1.5 million gallons of liquid SBW are temporarily stored at the ICPP in large underground stainless-steel tanks.

The United States Environmental Protection Agency and the Idaho Department of Health and Welfare filed a Notice of Noncompliance in 1992 contending some of the underground waste storage tanks do not meet secondary containment requirements as set forth in Title 40, Part 265.13 of the Code of Federal Regulations. As part of a 1995 agreement between the State of Idaho, the Department of Energy, and the Department of Navy, the SBW must be removed from the tanks by 2012.

Several technologies are currently being evaluated for the treatment and final disposition of SBW inventories. These technologies include blending with non-radioactive chemicals, essentially diluting sodium, followed by calcination of the liquid, and radionuclide partitioning followed by immobilization of the resulting high-activity and low-activity waste streams. A recent peer review identified the most promising radionuclide separation technologies for evaluation.¹ The Transuranic Extraction (TRUEX) process, developed by Horwitz and Schulz², was identified as a primary candidate for separation of the actinides from ICPP SBW.

A major emphasis at the ICPP has been directed toward evaluating actinide separation technologies for SBW using the TRUEX process. The active extractant used in the TRUEX process solvent is octyl(phenyl)-N,N-diisobutylcarbamoylmethylphosphine oxide (CMPO). Tributylphosphate (TBP) is added to the solvent as a phase modifier to prevent third phase formation and a paraffinic hydrocarbon is used as a diluent. The process has been effectively demonstrated to remove actinides from acidic SBW simulants and actual SBW solution to well below the 10 nCi/g NRC Class A low-level transuranic waste requirements. Previously, all testing at the INEL has been performed using batch contacts with actual and simulated SBW³, and using small-scale centrifugal contactors (2.0-cm and 5.5-cm rotor diameter) with SBW simulant.⁴ This report summarizes the results of TRUEX flowsheet testing for the separation of actinides from actual SBW solution (WM-183) using 2-cm diameter centrifugal contactors in a shielded cell facility.

PURPOSE AND SCOPE

The purpose of this study was to demonstrate the applicability of the TRUEX process for the treatment of ICPP SBW under continuous, countercurrent conditions using actual ICPP tank waste. Once the applicability of this process to SBW has been established, a comparison of the flowsheet to other technologies currently under development (e.g., other phosphine oxide and phosphonate processes) will be possible. Successful demonstration of the TRUEX process with SBW will also facilitate transfer of the technology from EM-50 to EM-30.

Installation of the 2-cm centrifugal contactors in the ICPP Remote Analytical Laboratory (RAL) shielded hot cell allowed the use of actual SBW in testing of the TRUEX process. Consequently, the scope of this study was to evaluate the TRUEX technology using actual ICPP tank waste. This allowed the removal efficiencies of the actinides to be determined and the effectiveness of the process in reducing the activity of the actinides to below NRC Class A LLW requirements to be evaluated. In addition, the behavior of H^+ , Fe, Hg, and Zr in the process was evaluated.

EQUIPMENT DESCRIPTION

Flowsheet testing was performed using 2-cm diameter centrifugal contactors installed in the CPP-684 RAL shielded hot cell. The 2-cm centrifugal contactors, as shown in Figure 1, consist of 24 stages of 2-cm diameter centrifugal contactors, feed and receiving vessels, feed pumps, and an air purge system for the contactor bearings. The aqueous (waste) and organic feed pumps and feed vessels were located inside the shielded cell. The remaining feed pumps and feed vessels were located outside the cell. All of the feed pump controllers were located outside the cell. Non-radioactive solutions used for the flowsheet testing were pumped to the centrifugal contactors through penetrations in the cell wall.

The centrifugal contactors were designed and fabricated by Argonne National Laboratory (ANL). The centrifugal contactors were designed specifically for operation of the TRUEX process with ICPP SBW. The contactors were modified at the ICPP for remote installation and operation in the RAL hot cell. Specifically, a modified support structure was fabricated for the contactors. This support structure is portable to allow the contactors to be moved out of the way when not in operation, contains leveling screws to adjust for unevenness in the cell floor, and can be disassembled into three sections. It was necessary to design the support structure for disassembly and reassembly so that the structure would fit through the 12 in. by 22 in. glove box access port into the cell and be assembled remotely. The centrifugal contactors were also installed through the access port in groups of four and assembled on the support structure remotely. Lifting bails were installed on each contactor to facilitate remote replacement or inspection of any motor/rotor assemblies. A description of the centrifugal contactors is provided in Table 1.

Solution was fed to the contactors using valveless metering pumps. Surge lines, consisting of 4-inch sections of 1-inch stainless steel tubing, were placed on the outlet of the pumps to dampen the surging flow. Because of the difficulty associated with remote installation, surge lines were not installed on the aqueous and organic feed lines located in cell. Flowrates were adjusted by controlling the pump speed using a ten-turn potentiometer or by manually adjusting the piston stroke length.

Clear, flexible Teflon® or Teflon® lined Tygon® tubing was used for inlet and outlet connections to the feed and receiving vessels. The feed lines were 1/8 in. o.d. tubing and the product lines were 3/8 in. o.d. tubing.

Table 1. Description of the 2-cm centrifugal contactors.

Size	2-cm rotor diameter
Motor	115 Volt, 60 Hz Bodine Model 710
RPM	3,600 rpm (not adjustable)
Material of construction	304L stainless steel
Inlet and outlet ports	3/8 in. o.d. tubing
Configuration	Single stage units which can be configured as desired. Stages are connected using U- tubes.

Figure 1. 2-cm diameter centrifugal contactors installed in the RAL shielded cell

The 2-cm centrifugal contactors do not have provisions for sampling the aqueous and organic solutions exiting individual stages during operation. The aqueous raffinate, strip product, wash effluent, and solvent recycle streams were sampled by routing the solution draining to receiving vessels into sample bottles during the actual flowsheet test. Individual stage samples were taken by draining the contactor stages after shutdown.

An air purge system was connected to the contactor bearing housings. Purge air was required in order to protect the motor body, shaft, and bearings from corrosive process fumes. Air to the bearing housings passed through a rotameter. The air flowrate through the rotameter could be adjusted from zero to five scfh. Air from the rotameter was split to feed each of the 24 contactors. The diameter of the air manifold was large (1/2 in. o.d. tubing), while the diameter of the tubes leading from the manifold to the contactors was small (1/8 in. o.d. tubing), resulting in the air flow to each contactor being approximately equivalent. The offgas from the bearing purge system was vented to the cell.

METHODOLOGY/EXPERIMENTAL PROCEDURE

WM-183 Waste

Sodium-bearing waste, obtained from tank WM-183 in June 1996, was used as feed solution for this testing. The WM-183 waste was filtered prior to testing using a 0.45 micron filter. Currently, approximately 1.5 million gallons of SBW are stored in six tanks. The composition in each tank varies; however, the composition of the solution in tank WM-183 is representative of the solution in all the tanks. The chemical composition of the WM-183 waste, along with the average composition of the current SBW inventory⁵, is shown in Table 2.

Table 2. WM-183 waste and average SBW tank compositions.

Component	WM-183 (M)	Average SBW (M)	Component	WM-183 (M)	Average SBW (M)
Acid (M)	1.72	1.59	SO ₄ (M)	0.066	0.050
Al (M)	0.62	0.64	Zr (M)	<6.56E-03	0.002
B (M)	0.013	0.018	Alpha (nCi/g)	456.8	369.4
Ca (M)	0.042	0.054	²⁴¹ Am (nCi/g)	33.86	90.1
Cl (M)	0.011	0.028	²³⁸ Pu (nCi/g)	327.3	34.7
Cr (M)	0.016	0.003	²³⁹ Pu (nCi/g)	125.9	244.6
Fe (M)	0.051	0.022	U (mg/L)	114	119.2
Hg (M)	0.0023	0.0011	²³⁷ Np (nCi/g)	<6.5 ^a	0.36
K (M)	0.096	0.206	⁹⁹ Tc (Ci/m ³)	0.036	nd ^b
Na (M)	0.77	1.9	¹³⁷ Cs (Ci/m ³)	218	41
NO ₃ (M)	5.24	5.07	⁹⁰ Sr (Ci/m ³)	240 ^a	38
Pb (M)	0.0015	0.0012			
a. Obtained from previous tank characterization (1994)					
b. Not determined					

TRUEX Solvent

The composition of the TRUEX solvent used in these tests was 0.2 *M* CMPO and 1.4 *M* TBP in Isopar L®. The solvent was prepared by the ICPP Quality Control Laboratory and was used previously for flowsheet development studies in the 5.5-cm Centrifugal Contactor Pilot Plant. The purity and composition of the TRUEX solvent were established prior to use in the centrifugal contactors. Impurities in the CMPO, resulting from acid hydrolysis, radiolytic degradation, or residual manufacturing impurities, may hinder the ability to strip the actinides from the TRUEX solvent.⁶ The distribution coefficient for ²⁴¹Am is particularly sensitive to CMPO impurities. Therefore, the distribution coefficient for ²⁴¹Am was evaluated as a function of nitric acid concentration from HNO₃ solutions to evaluate CMPO purity and solvent composition. This method of determining ²⁴¹Am distributions as a function of nitric acid concentration was established as a quality control procedure and was used to test the initial TRUEX solvent and the TRUEX solvent from each flowsheet test.⁷ If the results were comparable to literature values, the solvent was suitable for extraction studies.

Analytical

Samples from testing were analyzed for total alpha, Am, Pu, U, Tc, Hg, Fe, and Zr. Inductively Coupled Plasma Emission Spectroscopy (ICP-ES) was used for Fe and Zr analyses, while atomic fluorescence spectroscopy was used for Hg. Alpha spectroscopy was used for Am and Pu analyses, mass spectroscopy was used for U analyses, and liquid scintillation counting was used for Tc analyses. Neptunium determinations were not performed because of its extremely low activity coupled with the very large Pu activity. The Pu spectrum virtually overwhelms the Np spectrum, making it very difficult to quantitatively determine the Np concentration.

Most organic and aqueous samples were diluted prior to removal from the RAL hot cell due to their intense radioactivity. Aqueous dilutions were made in 3 volume % HNO₃ and organic dilutions were made in unused and freshly prepared TRUEX solvent. Dilutions of 0.1 mLs to 20.1 mLs were needed to bring the samples out of the hot cell. Additional dilutions on some aqueous samples were required for analytical, as opposed to radiological, purposes.

Organic samples could not be analyzed directly; therefore, 2 mL aliquots of each organic dilution were contacted with 10 mLs of 0.25 *M* 1-hydroxyethane 1,1-diphosphonic acid (HEDPA) made up in 0.04 *M* HNO₃ (O/A = 0.2) as part of the analytical preparation of the organic sample. HEDPA has been shown to quantitatively extract Fe, Zr, and the actinides from the TRUEX solvent.⁴ As a comparison, Pu and Am analyses were performed on a few organic samples directly by fusing them in a platinum crucible. Inconsistencies in Pu and Am values were not observed between the direct and indirect (HEDPA strip) organic analyses.

HEDPA will not strip Hg from the TRUEX solvent. Therefore, an additional 1 mL aliquot was taken from the TRUEX dilution and contacted with 5 mLs of 0.25 *M* Na₂CO₃. Sodium carbonate has been shown to quantitatively back extract Hg from the TRUEX solvent.⁴ Mercury analyses of the resulting aqueous solution were conducted by atomic fluorescence spectroscopy.

Gross alpha analyses were performed on all aqueous samples, as well as the aqueous solutions resulting from the HEDPA strips performed on the organic samples. Gross alpha analyses were performed by evaporating 1 mL of each sample on a 52-mm stainless steel counting planchet. The samples were counted by a gas flow proportional counter (Tennelec LB-5100) at a bias of 610 volts. This

voltage is within the recombination region for beta emissions to effectively eliminate crosstalk from the beta constituents. Unfortunately, larger dilutions were required on some of the aqueous samples to minimize beta interferences. Large alpha detection limits were observed in the samples requiring additional dilutions. Beta interferences were minimized/eliminated on some of the aqueous samples (1 Aq Product, 1 Aq Steady State Samples, and the 5 Aq, 6 Aq, 7 Aq, 8 Aq, 12 Aq, and 13 Aq stage samples) by contacting them twice with chlorinated cobalt dicarbollide at an O/A = 3/5. Chlorinated cobalt dicarbollide removes Cs and Sr, the primary beta emitters. This allowed for smaller sample dilutions to be counted; thus, lowering the alpha detection limit.

Americium and plutonium analyses were performed by partitioning these two actinides from one another using extraction chromatography. TEVA Resin® and TRU Resin® columns were placed sequentially and sample aliquots were passed through the columns. Plutonium (and neptunium) was partitioned onto the TEVA Resin® column while americium passes through the TEVA Resin® column and is extracted onto the TRU Resin® column, along with UO_2^{+2} . Plutonium was removed from the TEVA Resin® column with 0.5 M HCl, and americium was removed from the TRU Resin® column with 0.025 M HNO_3 . Uranium was removed from the TRU Resin® column with 0.1 M ammonium oxalate. The sample aliquots were spiked with NIST ^{236}Pu and ^{243}Am prior to separation by extraction chromatography so that analytical yields could be determined. Tracer yields were used to calculate total isotopic activity reported for each sample.

Technetium was chemically separated from the samples and quantified by liquid scintillation counting (LSC). A rigorous radionuclide separation procedure was performed to prevent erroneous LSC results. Sodium bisulfite was first added to a sample aliquot to reduce I^+ , followed by sodium nitrite addition to oxidize I^- to I_2 . Radioiodine was volatilized as I_2 by taking the samples to dryness and bringing them back to volume with 0.1 M HNO_3 . Cesium and strontium were removed by oxidizing the sample aliquots with hydrogen peroxide and passing them through a Dowex 50X cation exchange column. The sample exiting the column was taken to dryness and brought back to solution with 3.0 M H_2SO_4 . Technetium was extracted from sulfuric acid with TBP that was previously pre-equilibrated with unused 3.0 M H_2SO_4 . Aliquots of the TBP were counted using a Packard Tricarb 2500 Liquid Scintillation Spectrometer. Activity values were calibrated using a curve of counting efficiency versus quench. Duplicate Tc samples were analyzed, one being spiked with a known ^{99}Tc activity. The spiked and unspiked samples were processed simultaneously for consistency. Chemical yields were calculated from the spiked samples and used to account for Tc loss during the analytical separation process.

Iron and zirconium analyses were performed by ICP-ES. These analyses were performed at the RAL. A 0.5 mL to 50.5 mL dilution was performed on all aqueous samples which were in turn analyzed with the remotely operated ICP (these samples never left the hot cell). ICP analyses were performed on the aqueous solution resulting from the HEDPA strip of the organic samples, also using the RAL ICP.

TRUEX Flowsheet Demonstration

Based on the results of TRUEX flowsheet development studies performed at the ICPP using a non-radioactive SBW simulant in 2.0 and 5.5-cm diameter centrifugal contactors, a TRUEX flowsheet was recommended for testing in the 2-cm centrifugal contactors using actual ICPP SBW.⁴ This flowsheet consists of eight stages of extraction at an O/A of 0.33, five stages of 0.1 M HNO_3 scrub at an O/A of 1.5, six stages of 0.04 M HEDPA in 0.04 M HNO_3 strip at an O/A of 1.0, three stages of 0.25 M Na_2CO_3 wash at an O/A of 0.67, and two stages of 0.1 M HNO_3 rinse at an O/A of 3.0. There is no benefit for the

Figure 2. Flowsheet for TRUEX demonstration.

fractionation of individual actinides (i.e., Am from Pu) since all the actinides will be disposed of in the high-activity waste glass. Therefore, a gross actinide stripping agent (HEDPA) was used. The resulting flowsheet used for the TRUEX flowsheet testing is shown in Figure 2. It should be noted that this flowsheet was developed for testing with 24 stages of centrifugal contactors, which is expected to be more stages than is required for processing SBW.

The goals of the TRUEX flowsheet testing were to:

- (1) Demonstrate the overall operability of the 2-cm centrifugal contactors in a remote environment with the TRUEX flowsheet.
- (2) Determine the operational time required for the mass transfer in the contactors to reach steady state.
- (3) Determine the concentrations and distribution coefficients of ^{241}Am , Pu, U, ^{237}Np , ^{99}Tc , H^+ , Fe, Hg, and Zr for each stage at steady-state conditions.
- (4) Evaluate the effectiveness of the TRUEX flowsheet in separating the actinides from actual SBW.
- (5) Determine if any precipitate or third phase formation problems exist with this flowsheet.

The time required to reach steady state was evaluated by sampling the aqueous raffinate and strip product streams at 15 minute intervals and determining when component concentrations were no longer changing with time. These data are required in order to verify steady state was achieved prior to shutdown and sampling of each stage.

TRUEX flowsheet testing was performed as follows. The centrifugal contactor motors were started at 3,600 rpm. All aqueous solution flows, except for the actual SBW, were established. Simulant flow, consisting of 1.9 M HNO_3 , 0.6 M Al_2NO_3 , and 0.6 M NaNO_3 , was established for the SBW feed. Thirty minutes after the start of the aqueous feeds, solvent feed flow was established. Once solvent flow was seen exiting stage 24, actual SBW feed (WM-183) was started. Samples were taken from the aqueous

raffinate and strip product streams at intervals of 15, 30, 45, 75, and 90 minutes after the start of WM-183 feed. Level readings on each of the feed tanks were also noted in order to determine actual solution flowrates based on tank depletion rates. Approximately 100 minutes after the start of WM-183 feed, samples were taken of the aqueous raffinate, strip product, Na_2CO_3 wash, HNO_3 rinse, and solvent effluent streams. The centrifugal contactors were then shutdown by simultaneously stopping the feed pumps and contactor motors. Each stage remains approximately at steady-state operating conditions with this type of shutdown. This allowed aqueous and organic samples to be taken from each stage and, therefore, distribution coefficients to be determined for each of the 24 stages.

After shutdown, individual stage samples were taken as follows. Approximately 1 mL of solution was drained from each stage in order to remove any aqueous solution trapped in the drain line resulting from startup of the contactors. This prevents any dilution of the aqueous phase in the contactors. The solution from each stage was then drained into individual 60 mL sample bottles. The phases were re-equilibrated by shaking the bottles with a manipulator for several minutes each. Re-equilibration of the phases serves to evaluate distribution coefficients of the individual species under hypothetical conditions of 100% stage efficiency. The re-equilibrated solution from each stage was then poured into a clean separatory funnel, allowed to stand for five to ten minutes, and the aqueous and organic phases were separated.

RESULTS AND DISCUSSION

TRUEX Flowsheet Demonstration

Contactors Operation

Actual solution flowrates were calculated from feed tank depletion rates and are compared to the desired flowrates in Table 3. Precipitate or third phase formation was not observed during testing or after shutdown. Flooding was not observed during testing.

Material balance calculations indicated that the actual strip product flowrate was 4 mL/min higher than the strip feed flowrate and that the aqueous raffinate flowrate was equal to the feed flowrate (indicating no scrub flow). This indicates that one of the scrub section contactor motors was not operating during the test. While preparing the centrifugal contactors for unrelated flowsheet tests after the completion of this testing it was discovered that the stage 13 rotor was not spinning. It is suspected that this was also the case during the TRUEX testing. With an inoperable contactor motor, the aqueous and organic solutions entering the stage are not mixed and the solution level increases until the two phases exit out the light phase (organic) weir. Very little solution overflows out of the heavy phase (aqueous) weir. With stage 13 (scrub feed inlet stage) not operating, the scrub feed would have overflowed with the solvent into the strip section where it would have separated from the solvent and exited with the strip product solution. As a result, the flowsheet test was essentially performed without a scrub section.

Table 3. Flowrates and O/A ratios for TRUEX flowsheet testing.

Section	Phase	Flowrate (mL/min)		O/A Ratio		Total Flow (mL/min)
		Desired	Actual	Desired	Actual	
All	Org.	6.0	5.8	---	---	---
Extraction	Aq.	14.0	13.6	0.33	0.43	19.4
Scrub	Aq.	4.0	4.0 ^a	---	---	---
Strip	Aq.	6.0	5.2	1.0	1.1	11.0
Na ₂ CO ₃ Wash	Aq.	9.0	8.9	0.67	0.65	14.7
Acid Rinse	Aq.	2.0	1.9	3.0	3.1	7.7

a. Scrub feed was carried over to the strip section due to an inoperable contactor stage in the scrub section.

Time to Reach Steady State

The concentrations of gross alpha, ²⁴¹Am, ²³⁸Pu, ²³⁹Pu, H⁺, Fe, and Hg as a function of time (T₀ = start of WM-183 feed) are given in Figures 3 through 5 for the aqueous raffinate and strip product. It was assumed that steady state was reached when the concentration of the components varied by less than the analytical error associated with the sample analyses.

**Aqueous Raffinate Approach to Steady State
(Acid, Fe, and Hg)**

Figure 3. Aqueous raffinate approach to steady state.

**Strip Product Approach to Steady State
(Acid, Fe, and Hg)**

Figure 4. Strip product approach to steady state.

Strip Product Approach to Steady State (Gross Alpha, Am-241, Pu-238, and Pu-239)

Figure 5. Actinide approach to steady state, strip product

Of the non-radioactive components, H^+ and Fe reached steady state within 30 minutes in the raffinate and within 45 minutes in the strip product. Hg reached steady state within 45 minutes in the strip product, but the concentration of Hg immediately prior to shutdown dropped from 145 ppm to 9 ppm. The volume of sample was expended so the solution was not re-analyzed to determine if this change was actual or if it was due to analytical error. With the exception of the Hg concentration at 90 minutes, Hg reached steady-state in the raffinate within 60 minutes.

With the exception of the 90 minute samples, the gross alpha, ^{241}Am , ^{238}Pu , and ^{239}Pu reached steady state in the strip product within 45 minutes. The activities decreased for each of the 90 minute samples by approximately 45%, indicating an error associated with the sample dilution. It should be noted that this concentration fluctuation at 90 minutes did not occur in the raffinate samples for Fe, Hg, or H^+ , in which the samples were analyzed in the RAL hot cell. Therefore, it is expected that steady state was reached within 45 minutes for each of these species.

Concentrations at Shutdown

The concentrations of gross alpha, ^{241}Am , ^{238}Pu , ^{239}Pu , ^{235}U , ^{238}U , ^{99}Tc , H^+ , Fe, and Hg in each stream immediately prior to shutdown are given in Figure 6. Material balances for gross alpha, ^{241}Am , ^{238}Pu , ^{239}Pu , ^{235}U , ^{238}U , ^{99}Tc , H^+ , Fe, and Hg are given in Table 4. The values in Table 4 were normalized to obtain an overall material balance of 100%. Material balance calculations assume all of the scrub feed followed the solvent into the strip section and exited with the strip product. Distribution coefficients were calculated for gross alpha, ^{241}Am , ^{238}Pu , ^{239}Pu , ^{235}U , ^{238}U , H^+ , Fe, and Hg for each of the 24 stages. The resulting distribution coefficients are given in Tables 5 and 6. Data for Zr are not presented since all samples were below the Zr detection limit. ^{237}Np analyses were not performed due to analytical difficulties associated with the low Np activity. A discussion of the behavior of each component follows.

Component	SBW Feed	TRUEX Solvent Feed	LAW Raff.	Scrub Feed	Strip Feed	Strip Product	Na ₂ CO ₃ Feed	Na ₂ CO ₃ Effluent	Acid Rinse Feed	Acid Rinse Effluent	TRUEX Solvent Effluent
	1	2	3	4	5	6	7	8	9	10	11
H ⁺ (M)	1.72	---	1.27	0.1	0.04	0.82	---	---	0.1	nd ^a	nd
HEDPA (M)	---	---	---	---	0.04	0.04	---	---	---	---	---
Na ₂ CO ₃ (M)	---	---	---	---	---	---	0.25	0.25	---	---	---
Fe (M)	5.06E-02	---	3.85E-02	---	---	1.71E-02	---	<4.5E-04	---	<4.5E-04	<2.3E-03
Hg (M)	2.34E-03	---	5.53E-04	---	---	4.34E-05	---	2.25E-03	---	6.12E-06	3.69E-06
Alpha (dps/mL)	2.01E+04	---	5.1	---	---	3.00E+04	---	0.155	---	0.072	<0.22
²³⁸ Pu (dps/mL)	1.44E+04	---	<50	---	---	1.88E+04	---	0.09	---	0.006	nd
²³⁹ Pu (dps/mL)	5.54E+03	---	<44	---	---	7.12E+03	---	0.02	---	0.01	nd
²³⁵ U (ppm)	12.0	---	<0.13	---	---	13.8	---	3.76E-03	---	<6.00E-05	<1.50E-04
²³⁸ U (ppm)	102	---	6.85	---	---	120	---	3.31E-02	---	3.31E-03	7.61E-03
²⁴¹ Am (dps/mL)	1.49E+03	---	<16	---	---	2.12E+03	---	2.7E-03	---	3.00E-03	nd
⁹⁹ Tc (dps/mL)	1.31E+03	---	147	---	---	600	---	1.75E+03	---	3.04	3.18
Flowrate (mL/min)	13.6	5.8	13.6	4.0	5.2	9.2	8.9	8.9	1.9	1.9	5.8

a. Not determined

Figure 6. TRUEX flowsheet test concentrations at shutdown.

Table 4. Percentage of component in each of the effluent streams for TRUEX flowsheet testing.

Stream	Gross Alpha	²⁴¹ Am	²³⁸ Pu	²³⁹ Pu	²³⁵ U	²³⁸ U	⁹⁹ Tc	Hg	Fe
Aqueous Raffinate	0.025% (0.025%) ^a	<1.07% (<1.10%)	<0.35% (<0.39%)	<0.79% (<0.91%)	<1.1% (<1.4%)	6.7% (7.8%)	11.2% (8.7%)	23.7% (27.0%)	76.1% (76.9%)
Strip Product	101.1% (99.97%)	96.25% (>98.89%)	88.3% (>99.6%)	86.9% (>99.1%)	77.8% (>98.6%)	79.6% (92.2%)	31.0% (23.9%)	1.2% (1.4%)	22.9% (23.1%)
Na ₂ CO ₃ Effluent	4E-4% (4E-4%)	1E-4% (1E-4%)	4E-4% (4E-4%)	2E-4% (2E-04%)	0.02% (0.025%)	0.021% (0.024%)	87.2% (67.3%)	62.8% (79.7%)	<0.58% (<0.52%)
Rinse Effluent	5E-5% (9E-5%)	2E-5% (2E-5%)	6E-6% (6E-6%)	3E-5% (3E-5%)	<7E-5% (<9E-5%)	5E-4% (6E-4%)	0.032% (0.025%)	4E-2% (4E-2%)	<0.13% (<0.13%)
Solvent Effluent	<5E-04% (<5E-4%)	nd ^c	nd	nd	<5E-4% (<6E-4%)	3E-3% (3E-3%)	0.10% (0.08%)	7E-2% (8E-2%)	<1.9% (<1.9%)
Mass Balance ^b	101.1%	97.3%	88.7%	87.7%	78.9%	86.3%	129.6%	87.8%	98.9%

a. Normalized percentage.

b. The mass balance is the amount of a component accounted for based on the sample analysis results.

c. Not determined.

Table 5. Gross alpha, ²⁴¹Am, ²³⁸Pu, ²³⁹Pu, ²³⁵U and ²³⁸U distribution coefficients for TRUEX flowsheet testing.

Stage		D _{gross alpha}	D _{Am-241}	D _{Pu-238}	D _{Pu-239}	D _{U-235}	D _{U-238}
Extraction	1	nd ^a	nd	3.6	41.1	nd	nd
	2	nd	nd	2.0	<2.1	nd	nd
	3	nd	nd	>31.6	nd	nd	nd
	4	nd	<12.2	>17.7	>51.3	nd	nd
	5	102.3	<58.8	<9.5	<20.7	nd	nd
	6	24.3	13.2	10.4	34.0	nd	nd
	7	16.2	59.4	1.1	1.3	nd	nd
	8	671.3	37.6	305	299	790	280
Scrub	9	nd	12.9	716	660	nd	nd
	10	179.9	10.7	5340	7060	nd	nd
	11	179.3	12.0	7300	6910	nd	nd
	12	49667	857	16500	12100	nd	nd
	13	1928	47.6	17800	15300	nd	nd
Strip	14	1.5	6.8	0.091	0.093	22.5	18.7
	15	0.34	1.4	0.012	0.014	1.2	1.2
	16	0.040	nd	0.0014	0.0017	0.02	0.02
	17	0.0044	nd	0.0018	0.010	0.004	0.004
	18	0.012	<0.20	nd	0.13	0.08	0.06
Wash	19	0.022	nd	nd	0.52	0.2	0.8
	20	0.10	nd	0.17	0.87	3.4	35
	21	0.049	nd	0.044	0.91	10.7	64
	22	0.056	nd	0.0070	0.015	24.8	414
Rinse	23	12.7	0.37	9.2	8.5	233	65
	24	87.3	<0.05	60.3	71.4	320	124

a. Not determined.

Table 6. H⁺, Fe, and Hg distribution coefficients for TRUEX flowsheet testing.

Stage		D _H ⁺	D _{Fe}	D _{Hg}
Extraction	1	0.88	<1.3	3.4
	2	0.70	<1.5	1.7
	3	0.72	<11.1	1.0
	4	0.69	<5.5	0.45
	5	0.66	<1.2	1.0
	6	0.70	<5.2	0.44
	7	0.75	<5.1	0.38
	8	0.69	<1.1	0.87
Scrub	9	nd ^a	nd	0.58
	10	0.26	<1.9	0.45
	11	0.85	<1.5	0.39
	12	0.48	<1.5	>54
	13	0.79	<3.7	1.6
Strip	14	0.76	<11.9	10
	15	0.51	nd	59
	16	nd	<4.4	>239
	17	nd	nd	483
	18	nd	nd	1070
	19	nd	nd	1236
Wash	20	nd	nd	0.08
	21	nd	nd	2.3
	22	nd	nd	>7.4
Rinse	23	nd	nd	0.79
	24	nd	<3.0	0.96

a. Not determined.

Nitric Acid. Distribution coefficients of H⁺ in the extraction section ranged from 0.66 to 0.88, resulting in an H⁺ concentration of 1.2 M in the TRUEX solvent exiting the extraction section. The 0.1 M HNO₃ scrub typically will back-extract the HNO₃ from the TRUEX solvent prior to the solvent entering the strip section. H⁺ was not scrubbed from the TRUEX solvent as a result of the inoperable stage in the scrub section, and the H⁺ concentration remained at 1.2 M.

Gross Alpha. The gross alpha activity was reduced from 2.01E+04 dps/mL in the feed to 5.1 dps/mL in the aqueous raffinate immediately prior to shutdown. This corresponds to a removal efficiency of 99.97%. A gross alpha activity of 5.1 dps/mL corresponds to 0.12 nCi/g which is well below the NRC Class A LLW requirement of 10 nCi/g for non-TRU waste. The strip section was very effective in back-extracting the actinides from the TRUEX solvent, removing 99.9998% of the gross alpha activity from the solvent with six strip stages.

Americium. The activity of ²⁴¹Am was reduced from 1.49E+03 dps/mL in the feed to <16 dps/mL in the aqueous raffinate immediately prior to shutdown. A lower detection limit was achieved for the stage 1 aqueous sample (drained from stage 1 after shutdown), yielding an activity of <1.54 dps/mL. This corresponds to a removal efficiency of >99.90%.

Extraction distribution coefficients for ^{241}Am ranged from <12 to 59. The strip section was very effective in back-extracting the ^{241}Am from the TRUEX solvent, removing >99.99%. As a result of the inoperable contactor stage in the scrub section, acid was not scrubbed from the solvent and the distribution coefficients on the first two strip stages were greater than one (6.8 and 1.4).

Plutonium. The activity of ^{238}Pu was reduced from $1.44\text{E}+04$ dps/mL in the feed to <50 dps/mL in the aqueous raffinate immediately prior to shutdown. An improved detection limit was achieved for the stage 1 aqueous sample (drained from stage 1 after shutdown), yielding an activity of 5.1 dps/mL. This corresponds to a removal efficiency of 99.96%.

The activity of ^{239}Pu was reduced from $5.54\text{E}+03$ dps/mL in the feed to <44 dps/mL in the aqueous raffinate immediately prior to shutdown. An improved detection limit was achieved for the stage 1 aqueous sample (drained from stage 1 after shutdown), yielding an activity of 1.1 dps/mL. This corresponds to a removal efficiency of 99.98%.

Distribution coefficients for Pu in the extraction section varied considerably. However, most of the Pu was extracted in the first extraction contact, resulting in Pu activities near the detection limit. Also, residual Pu activity from previous testing is expected to be present in the contactors, resulting in contamination of the stage samples. As a result, distribution coefficients for stages 1 through 7 are expected to have a large uncertainty associated with them. The distribution coefficients for ^{238}Pu and ^{239}Pu on the first extraction stage (stage 8) were approximately 300. Strip distribution coefficients were <0.1. As a result, >99.99% of the Pu extracted by the TRUEX solvent was back-extracted with the 6 strip stages.

Uranium. The activity of ^{235}U was reduced from 12 ppm in the feed to <0.13 ppm in the aqueous raffinate immediately prior to shutdown. This corresponds to a removal efficiency of >98.89%. The activity of ^{238}U was reduced from 102 ppm in the feed to 6.85 ppm in the aqueous raffinate immediately prior to shutdown. This corresponds to a removal efficiency of 93.3%. Much higher removal efficiencies were expected for U. However, residual U activity from previous testing in the centrifugal contactors is expected to be present in the contactors, resulting in contamination of the raffinate sample.

The distribution coefficients for ^{235}U and ^{238}U on the first extraction stage (stage 8) were 790 and 280, respectively. As a result of the inoperable contactor stage in the scrub section, acid was not scrubbed from the solvent and the distribution coefficients on the first two strip stages were greater than 1.0. The distribution coefficients dropped to less than 0.1 on the remaining strip stages since the acid was stripped from the solvent on the first two strip stages. As a result, >99.9% of the U extracted by the TRUEX solvent was back-extracted with the 6 strip stages.

Technetium. The activity of ^{99}Tc was reduced from 1,310 dps/mL in the feed to 147 dps/mL in the aqueous raffinate immediately prior to shutdown. This corresponds to a removal efficiency of 89%. Distribution coefficients were not determined for ^{99}Tc on each of the stages. However, an average ^{99}Tc extraction distribution coefficient of 2.3 on each of the eight extraction stages results in a removal efficiency of 89%. The strip section was moderately effective in back-extracting the ^{99}Tc from the TRUEX solvent, with 26% of the extracted ^{99}Tc exiting in the strip product stream. The remainder of the ^{99}Tc (74%) was stripped from the solvent in the Na_2CO_3 wash section, preventing any buildup of ^{99}Tc in the TRUEX solvent.

It is important to note that the ^{99}Tc activities in ICPP tank wastes are anticipated to be below NRC Class A LLW requirements (0.3 Ci/m^3). Technetium removal is of concern due to its mobility, as TcO_4^- , in the environment. It is therefore advantageous to be able to fractionate ^{99}Tc from the wastes.

Iron. Twenty three percent of the Fe was extracted by the TRUEX solvent and exited with the strip product. The HEDPA strip was effective in back-extracting the Fe from the solvent, resulting in < 1.9% of the Fe remaining in the solvent effluent. Solvent sample analyses were below the Fe detection limit so distribution coefficients could not be calculated directly. Using solvent concentrations obtained from material balances, extraction distributions ranging from 0.3 to 0.75 were obtained.

Results from previous flowsheet testing with simulated SBW indicated that < 1% of the Fe exits with the strip product. Fe is typically back-extracted from the TRUEX solvent in the scrub section. However, no Fe was scrubbed from the solvent due to the inoperable contactor stage in the scrub section.

Mercury. The TRUEX solvent extracted 73% of the Hg from the WM-183 waste. Extraction distribution coefficients ranged from 0.4 to 3.4. The HEDPA strip was ineffective in back-extracting Hg from the solvent, resulting in only 1.4% of the Hg exiting with the strip product (high-activity waste fraction). However, the three stages of 0.25 M Na₂CO₃ wash removed 99.9% of the Hg from the TRUEX solvent. As a result, 98.6% of the Hg in the WM-183 feed was partitioned between the low-activity waste Na₂CO₃ wash effluent and raffinate streams.

Comparison With Results Obtained Using SBW Simulant

Prior to performing the TRUEX flowsheet testing with actual waste solution, testing was performed in the same equipment using simulated SBW spiked with ²⁴¹Am, ²⁰³Hg, and ⁹⁵Zr. The composition of the simulated SBW is given in Table 7. The testing performed with the simulant was identical to the testing performed with actual waste except stage samples were not taken at the completion of the test. Results obtained using simulated SBW are compared in Table 8 with results obtained using actual waste solution.

Results obtained using ²⁴¹Am and ²⁰³Hg tracers spiked into the SBW simulant compare very well with ²⁴¹Am and Hg results with actual waste solution, indicating that the simulant is representative of WM-183 waste solution. Results obtained for non-radioactive Zr and spiked ⁹⁵Zr in the SBW simulant are drastically different. Only 11% of the non-radioactive Zr was extracted as compared to essentially all of the ⁹⁵Zr. Results for Zr with the actual WM-183 waste were below the analytical detection limit so it is not known how the actual Zr data will compare to the simulant data. Additional testing should be performed in order to evaluate the behavior of Zr with actual waste solution and determine if the non-radioactive Zr or the ⁹⁵Zr in the SBW simulant is more representative of Zr behavior in actual waste.

Table 7. Composition of Simulated SBW

Component	<i>M</i>	Component	<i>M</i>
Acid (H ⁺)	1.26	K	1.38E-01
Al	5.63E-01	Mn	1.42E-02
B	1.40E-02	Mo	1.49E-03
Cd	2.05E-06	Na	1.17
Ca	3.41E-02	NO ₃	4.46
Ce	3.63E-04	Ni	1.63E-03
Cl	3.52E-02	Pb	9.51E-04
Cs	7.52E-05	PO ₄	<9.18E-03
Cr	5.63E-03	Sr	5.15E-03
F	9.66E-02	SO ₄	3.86E-02
Fe	2.40E-02	Zr	3.86E-04
Hg	1.80E-03	⁹⁵ Zr (dps/mL)	79.7
²⁰³ Hg (dps/mL)	16.3	²⁴¹ Am (dps/mL)	1.090

Table 8. Comparison of results obtained with simulant and actual waste.

Stream	²⁴¹ Am		²⁰³ Hg		Zr		⁹⁵ Zr
	Simulant	WM-183	Simulant	WM-183	Simulant	WM-183	Simulant
Aqueous Raffinate	<0.25%	<1.1%	47.1%	23.7%	93.3%	nd ^a	<0.6%
Strip Product	101.3%	96.3%	1.0%	1.2%	11.4%	nd	96.4%
Na ₂ CO ₃ Effluent	<0.1%	1E-4%	51.8%	62.8%	1.7%	nd	<0.2%
Rinse Effluent	<0.02%	2E-5%	<0.13%	0.04%	nd	nd	<0.04%
Solvent Effluent	<0.07%	nd	7.8%	0.07%	1.8%	nd	<0.12%
Mass Balance ^b	101.3%	97.3%	107.7%	87.8%	107.4%	nd	96.4%

a. Not determined

b. The mass balance is the amount of a component accounted for based on the sample analysis results.

CONCLUSIONS AND RECOMMENDATIONS

Conclusions

An overall removal efficiency of 99.97% was obtained for the actinides. As a result, the activity of the actinides was reduced from 457 nCi/g in the feed to 0.12 nCi/g in the aqueous raffinate, which is well below the NRC Class A LLW requirement of 10 nCi/g for non-TRU waste. Removal efficiencies of >99.9%, 99.96%, 99.98%, >98.89%, 93.3%, and 89% were obtained for ^{241}Am , ^{238}Pu , ^{239}Pu , ^{235}U , ^{238}U , and ^{99}Tc , respectively.

The 0.04 M HEDPA strip section was very effective in back-extracting the actinides from the TRUEX solvent. Six strip stages removed 99.9998% of the actinide activity from the solvent.

Nitric acid was not scrubbed from the TRUEX solvent with the six scrub stages due to the scrub feed stage not operating. As a result, all of the scrub feed overflowed with the solvent into the strip section, effectively eliminating the scrub section.

Iron was partially extracted by the TRUEX solvent, resulting in 23% of the Fe exiting in the strip product. The inoperable contactor stage in the scrub section prevented the Fe from being scrubbed from the solvent. Mercury was also extracted by the TRUEX solvent (73%) and stripped from the solvent in the 0.25 M Na_2CO_3 wash section. Only 1.4% of the Hg exited with the high-activity waste strip product.

Flooding, precipitate formation, or third phase formation were not observed during the flowsheet testing.

Centrifugal contactor flowsheet test results obtained using ^{241}Am and ^{203}Hg tracers spiked into the SBW simulant compare very well with ^{241}Am and Hg results with actual waste solution, indicating that the simulant is representative of WM-183 waste solution.

Recommendations

The TRUEX process for the separation of actinides from ICPP SBW is now sufficiently developed to allow transfer of the technology from the EM-50 Tanks Focus Area to EM-30. EM-30 testing of the flowsheet with actual waste to verify scrub operational parameters and to optimize the flowsheet is recommended.

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Appendix A
Experimental Data

Table A-1. Approach to steady state samples.

Description	Sample Label	Alpha (dps/mL)	Am-241 (dps/mL)	Pu-238 (dps/mL)	Pu-239 (dps/mL)	H+ (M)	Fe (M)	Zr (M)	Hg (M)
Approach	1aq-15	nd	nd	nd	nd	1.357	3.05E-02	< 6.56E-03	4.39E-05
To	1aq-30	nd	nd	nd	nd	1.303	3.87E-02	< 6.56E-03	0.000274
Steady	1aq-45	nd	nd	nd	nd	1.328	3.89E-02	< 6.56E-03	0.000276
State	1aq-60	nd	nd	nd	nd	1.301	3.96E-02	< 6.56E-03	0.000429
Samples	1aq-75	nd	nd	nd	nd	1.294	3.98E-02	< 6.56E-03	0.000434
	1aq-90	nd	nd	nd	nd	1.296	3.92E-02	< 6.56E-03	0.000226
	14aq-15	3.98E+04	2.08E+03	3.09E+04	1.17E+04	0.706	9.14E-03	< 7.15E-04	0.000434
	14aq-30	2.60E+04	1.39E+03	1.81E+04	6.81E+03	0.602	1.49E-02	< 7.15E-04	0.000548
	14aq-45	2.90E+04	1.70E+03	2.02E+04	7.88E+03	0.764	1.54E-02	< 7.15E-04	0.000693
	14aq-60	3.01E+04	2.23E+03	2.48E+04	8.90E+03	0.788	1.60E-02	< 7.15E-04	0.000703
	14aq-75	2.88E+04	1.98E+03	2.13E+04	7.92E+03	0.818	1.60E-02	< 7.15E-04	0.000723
	14aq-90	1.79E+04	1.13E+03	1.11E+04	4.20E+03	0.837	1.65E-02	< 7.15E-04	0.000723

Table A-2. Effluent samples taken just prior to shutdown.

Description	Sample Label	Alpha (dps/mL)	Am-241 (dps/mL)	Pu-238 (dps/mL)	Pu-239 (dps/mL)	Tc-99 (dps/ml)	U-235 (ppm)	U-238 (ppm)	H+ (M)	Fe (M)	Zr (M)	Hg (M)
FEED Sample	WM-183	2.01E+04	1.49E+03	1.44E+04	5.54E+03	1.31E+03	12	102	1.722	5.06E-02	< 6.56E-03	0.002338
Product	1aq-Prod	5.1	< 1.60E+01	< 5.00E+01	< 4.40E+01	147	0.133	6.85	1.27	3.85E-02	< 6.56E-03	0.000553
Samples	14aq-Prod	3.00E+04	2.12E+03	1.88E+04	7.12E+03	600	13.8	120	0.82	1.71E-02	< 1.36E-03	4.34E-05
Prior	20aq-Prod	0.115	2.70E-03	0.09	0.0201	1.75E+03	3.76E-03	0.0331	nd	< 4.53E-04	< 7.10E-04	0.002248
To	23aq-Prod	0.072	3.00E-03	0.0059	0.011	3.04	< 6.00E-05	3.31E-03	nd	< 4.53E-04	< 7.10E-04	6.12E-06
Shutdown	24or-Prod	< 2.22E-01	nd	nd	nd	3.18	< 1.50E-04	7.61E-03	nd	< 2.26E-03	< 3.55E-03	3.69E-06

Table A-3. Sample data for the individual contactor stages after phase re-equilibration.

Description	Sample Label	Gross Alpha (dps/mL)	Am-241 (dps/mL)	Pu-238 (dps/mL)	Pu-239 (dps/mL)	H+ (M)	Fe (M)	Zr (M)	Hg (M)
Extraction	1or	25	<1.08E+01	1.83E+01	4.40E+0	1.08E+00	<4.16E-02	<6.52E-02	1.15E-03
	1aq	nd	<1.54E+00	5.08E+00	1.07E+00	1.23E+00	3.15E-02	<6.56E-03	3.39E-04
	2or	33	nd	6.90E+01	7.28E+00	1.19E+00	<5.19E-02	<8.15E-02	1.50E-03
	2aq	nd	1.21E+00	3.51E+01	1.07E+01	1.70E+00	3.56E-02	<6.56E-03	8.77E-04
	3or	37	<1.33E+01	4.80E+01	2.51E+02	1.30E+00	<4.55E-01	<7.10E-01	1.20E-03
	3aq	nd	<1.16E+00	<1.52E+00	nd	1.80E+00	4.10E-02	<6.56E-03	1.21E-03
	4or	56.5	<1.28E+01	1.73E+01	1.54E+01	1.24E+00	<2.29E-01	<3.60E-01	6.13E-04
	4aq	nd	1.05E+00	<9.80E-01	<3.00E-01	1.79E+00	4.20E-02	<6.56E-03	1.36E-03
	5or	133	1.27E+02	<4.16E+01	<2.87E+01	1.18E+00	<5.19E-02	<8.15E-02	1.70E-03
	5aq	1.3	2.16E+00	4.39E+00	1.39E+00	1.78E+00	4.43E-02	<6.56E-03	1.65E-03
	6or	197	5.82E+01	5.92E+01	5.55E+01	1.27E+00	<2.29E-01	<3.60E-01	7.48E-04
	6aq	8.1	4.40E+00	5.67E+00	1.63E+00	1.82E+00	4.42E-02	<6.56E-03	1.71E-03
	7or	635	2.34E+02	1.19E+02	5.52E+01	1.30E+00	<2.29E-01	<3.60E-01	7.83E-04
	7aq	39.1	3.94E+00	1.10E+02	4.16E+01	1.75E+00	4.52E-02	<6.56E-03	2.06E-03
	8or	3.97E+04	3.36E+03	1.97E+04	7.24E+03	1.22E+00	<5.19E-02	<8.15E-02	1.94E-03
	8aq	59.2	8.94E+01	6.45E+01	2.42E+01	1.76E+00	4.60E-02	<6.56E-03	2.23E-03
Scrub	9or	7.06E+04	3.48E+03	3.58E+04	1.32E+04	1.15E+00	<4.16E-02	<6.56E-02	1.45E-03
	9aq	nd	2.70E+02	5.00E+01	2.00E+01	nd	<9.20E-02	<1.44E-01	2.49E-03
	10or	7.32E+04	3.69E+03	3.58E+04	1.32E+04	7.38E-01	<4.16E-02	<6.52E-02	1.40E-03
	10aq	406.9	3.44E+02	6.70E+00	1.87E+00	2.80E+00	2.18E-02	<8.20E-03	3.09E-03
	11or	8.57E+04	4.50E+03	5.04E+04	1.86E+04	1.60E+00	<4.16E-02	<6.52E-02	1.30E-03
	11aq	478	3.75E+02	6.90E+00	2.69E+00	1.88E+00	2.83E-02	<8.20E-03	3.31E-03
	12or	5.96E+04	3.17E+03	2.81E+04	1.05E+04	1.49E+00	<4.16E-02	<6.52E-02	1.25E-03
	12aq	1.2	3.70E+00	1.70E+00	8.65E-01	3.13E+00	2.83E-02	<8.20E-03	2.29E-05
	13or	1.11E+05	7.57E+03	5.41E+04	2.06E+04	1.19E+00	<5.19E-02	<8.15E-02	1.80E-03
	13aq	57.6	1.59E+02	3.03E+00	1.35E+00	1.51E+00	1.41E-02	<8.20E-03	1.14E-03
Strip	14or	2.99E+04	1.64E+04	1.68E+03	6.40E+02	7.60E-01	<2.29E-01	<3.60E-01	6.58E-04
	14aq	1.95E+04	2.41E+03	1.85E+04	6.86E+03	1.00E+00	1.93E-02	<8.20E-03	6.28E-05
	15or	1.38E+04	7.38E+03	2.69E+02	1.08E+02	2.74E-01	<1.15E-01	<1.81E-01	1.11E-03
	15aq	4.04E+04	5.11E+03	2.16E+04	7.99E+03	5.39E-01	<5.23E-03	<8.20E-03	1.89E-05
	16or	2.99E+02	1.87E+02	1.90E+00	8.50E-01	nd	<2.26E-03	<3.55E-03	4.54E-03
	16aq	7.48E+03	nd	1.36E+03	5.01E+02	nd	5.20E-04	<7.14E-04	1.89E-05
	17or	1.01E+00	2.60E-01	1.50E-01	2.90E-01	nd	<2.26E-03	<3.55E-03	4.34E-03
	17aq	2.28E+02	nd	8.34E+01	2.88E+01	2.08E-01	<4.55E-04	<7.14E-04	8.97E-06
	18or	2.15E-01	<1.44E-01	nd	2.89E-01	nd	<2.26E-03	<3.55E-03	4.54E-03
	18aq	18.17	7.15E-01	1.27E+01	2.22E+00	1.61E-01	<4.55E-04	<7.14E-04	4.24E-06
	19or	0.223	nd	nd	3.89E-01	nd	<2.26E-03	<3.55E-03	4.44E-03
	19aq	9.96	3.30E-01	6.90E+00	7.50E-01	1.86E-01	<4.55E-04	<7.14E-04	3.59E-06
Na2CO3	20or	0.408	nd	5.80E-01	3.79E-01	nd	<2.26E-03	<3.55E-03	1.81E-05
Wash	20aq	3.89	7.30E-02	3.37E+00	4.35E-01	nd	<4.55E-04	<7.14E-04	2.28E-04
	21or	0.357	nd	2.87E-01	7.20E-01	nd	<2.26E-03	<3.55E-03	6.73E-06
	21aq	7.31	1.09E-01	6.47E+00	7.90E-01	nd	<4.55E-04	<7.14E-04	2.87E-06
	22or	0.572	nd	4.80E-02	1.30E-02	nd	<2.28E-03	<3.57E-03	4.59E-06
	22aq	10.14	3.60E-02	6.87E+00	8.80E-01	nd	<4.55E-04	<7.14E-04	6.23E-07
	23or	15.22	1.20E-01	9.10E+00	1.10E+00	nd	<2.26E-03	<3.55E-03	3.94E-06
Acid	23aq	1.198	3.27E-01	9.90E-01	1.30E-01	nd	<4.55E-04	<7.14E-04	4.97E-06
Rinse	24or	7.07	<6.00E-02	4.70E+00	6.50E-01	nd	<2.26E-03	<3.55E-03	4.98E-06
	24aq	0.081	1.25E+00	7.80E-02	9.10E-03	nd	7.60E-04	<7.14E-04	5.18E-06