

WSRC-TR-2001-00533, Rev. 1

**Keywords: Waste processing, cesium
Retention: Permanent**

**SOLVENT EXTRACTION BATCH DISTRIBUTION
COEFFICIENTS WITH SAVANNAH RIVER SITE DISSOLVED
SALT CAKE**

**D. D. Walker
M. H. Beasley
A. D. Coleman
B. H. Croy
C. C. DiPrete
D. P. DiPrete**

March 26, 2002

**Westinghouse Savannah River Company
Savannah River Technology Center
Aiken, SC 29808**



SAVANNAH RIVER SITE

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-96SR18500 with the U.S. Department of Energy.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

This report has been reproduced directly from the best available copy.

Available for sale to the public, in paper, from: U.S. Department of Commerce, National Technical Information Service, 5285 Port Royal Road, Springfield, VA 22161

phone: (800) 553-6847

fax: (703) 605-6900

email: orders@ntis.fedworld.gov

online ordering: <http://www.ntis.gov/support/index.html>

Available electronically at <http://www.osti.gov/bridge>

Available for a processing fee to U.S. Department of Energy and its contractors, in paper, from: U.S. Department of Energy, Office of Scientific and Technical Information, P.O. Box 62, Oak Ridge, TN 37831-0062

phone: (865)576-8401

fax: (865)576-5728

email: reports@adonis.osti.gov

SOLVENT EXTRACTION BATCH DISTRIBUTION COEFFICIENTS WITH SAVANNAH RIVER SITE DISSOLVED SALT CAKE

D. D. Walker, M. H. Beasley, A. D. Coleman, B. H. Croy, C. C. DiPrete,
and D. P. DiPrete

SUMMARY

Researchers characterized high-level waste derived from dissolved salt cake from the Savannah River Site (SRS) tank farm and measured the cesium distribution coefficients (D_{Cs}) for extraction, scrub, and stripping steps of the caustic-side solvent extraction (CSSX) flowsheet. The measurements used two SRS high-level waste samples derived entirely or in part from salt cake. The chemical compositions of both samples are reported. Dissolved salt cake waste contained less Cs-137 and more dianions than is typical of supernate samples. Extraction, scrub, and strip D_{Cs} values for both samples exceeded process requirements and agreed well with model predictions. The results indicate no significant problems processing dissolved salt cake compared to supernate. During the course of testing, researchers observed sorption of cesium on glass sample vials containing strip solutions. The problem was detected in the material balance calculated for each organic/aqueous contact. Methods and recommendations for avoiding this problem are discussed.

INTRODUCTION

A solvent extraction process for removal of cesium from alkaline solutions has been developed utilizing a novel solvent invented at the Oak Ridge National Laboratory (ORNL).¹ This solvent consists of a calix[4]arene-crown-6 extractant dissolved in an inert hydrocarbon matrix. A modifier, an alkylphenoxy alcohol, is added to the solvent to enhance the extraction power of the calixarene and to prevent the formation of a third phase. An additional additive, trioctylamine, is used to improve stripping performance and to mitigate the effects of any surfactants present in the feed stream.

A number of laboratory studies with simulated wastes determined that the solvent system is stable and performs well in the presence of the major chemical components in the SRS high-level waste tanks. However, confirmation of the simulant results with actual high-level waste samples from the SRS tank farms have yielded mixed results relative to cesium removal efficiency. In general the extraction D_{Cs} values equal or exceed the flowsheet requirement of 8.²⁻³ Nevertheless, the experimental values tend to be lower than the values predicted by a model based on waste simulant measurements developed by ORNL researchers.⁴ Previous results with supernate samples are summarized in Table I.

TABLE I. D_{Cs} Values with Actual SRS High-Level Waste*

Tank	D_{Cs} Values (25 °C)						
	Extraction		Scrub	Strip			Ref.
	Model**	Expt.		1	2	3	
13H	8.8	10.1	4.4	1.1	0.25	0.12	3
26F	15	7.7	1.1	0.10	0.23	0.04	3
33F	24	8.8	1.1	0.07	0.04	0.06	3
35H	22	7.9	0.7	0.14	0.17	0.16	3
46F	17	13.8	1.6	0.27	0.10	0.04	3
37F/44H		19.0	1.76	0.20	0.10	0.07	2

* Supernate samples.

**ORNL model, Ref. 4

This work partially fulfills a request from the U. S. Department of Energy, Savannah River Operations Office (C. E. Anderson, Assistant Manager for High-Level Waste, Department of Energy-Savannah River, memorandum to S. Wood, "Fiscal Year (FY) 2001 Annual Operating Plan (AOP) Changes in Support of the Salt Processing Project," dated 10 July 2001). The work complies with the following task plan: D. D. Walker, "Task Technical and Quality Assurance Plan for CSSX Real Waste Batch Tests," WSRC-RP-2001-00772, Rev.0, August 14, 2001.

EXPERIMENTAL

Tank 38H and 46F Waste Preparation and Composition

SRS tank farm personnel provided salt cake samples of radioactive waste from Tanks 38H and 46F. The samples originally served for studies of evaporator chemistry.⁵ The Savannah River Technology Center (SRTC) received, stored, and processed the samples in a remotely operated, shielded facility. Table II summarizes source information for each sample.

The Tank 38H samples from the evaporator studies included both solids and solutions. The solutions derived from dissolution of solids in water. Aqueous solutions from the second sampling event (July 2001) were combined and evaporated by circulating ambient temperature argon over the surface. After solids crystallized, the solids and solution from the first sampling event (January 2001) were added, additional water was added, the slurry was stirred to saturate the solution and then filtered (0.45 micron nominal pore size). The resulting solution contained 7.1 M sodium ion (density 1.309 g/mL). Dilution with 1.6 M NaOH solution achieved a final salt concentration of 5.6 M sodium ion (density 1.244 g/mL). Table III lists the composition of this solution.

Table II. Origin of Tank 38H and 46F Salt Cake Samples

	<u>Tank 38H</u>	<u>Tank 46F</u>
Date sampled:	6/26/01 and 7/25/01	7/3/01
Tank Farm identification	HTF-395, HTF-396	(no identification)
Approximate amount:	34 g solids and 90 mL of solution	~40 g of solids dissolved in 240 mL of water

The Tank 46F samples were all solutions derived from dissolution of solids in inhibited water. The solution (240 mL) was evaporated to 100 mL and filtered. The resulting solution contained 4.7 M sodium ion (density 1.167 g/mL). Since the solution was below the desired concentration of 5.6 M Na⁺, it was diluted with Tank 37H/44F composite waste supernate.² Table III lists the composition of the final Tank 46F sample.

Batch Distribution Test Protocol

The protocol for the Cs-137 batch distribution tests included a single extraction step, two scrub steps, and three strip steps (ESS protocol). This differs from previous testing in the number of scrub steps. Previous test results suggested two scrub steps ensured adequate pH adjustment prior to the strip steps.²⁻³ Tests occurred with duplicate samples of each waste solution with targeted organic-to-aqueous (O/A) volume ratios of 0.33 for the extraction step and 5.0 for the scrub and strip steps. We used polypropylene centrifuge tubes (40 mL and 15 mL, Falcon or Corning brand, with lids provided by the manufacturer) for the batch contacts (Figure 1), and borosilicate glass vials for samples and dilutions. Aqueous and organic phase volumes were calculated from the aliquot weights and the solution densities. Table III lists the waste solution densities. The solvent density was 0.826 g/mL and the scrub or strip densities were 1.00. Technicians mixed the phases by shaking 2.5 minutes at ambient cell temperature (25±5 °C) with remote manipulator arms. Centrifuging (~1500 rpm) for 45 minutes separated the phases in the tubes. Using polyethylene eyedroppers, the majority of the organic phase was removed to a clean centrifuge tube. The remaining organic phase and a portion of the aqueous phase were removed and discarded to provide a clean surface for sampling the aqueous phase. Both phases were sampled by 0.5 mL pipette, placed in glass vials, and the vials removed from the shielded facility. In a radiochemical hood, portions (~0.2 mL) of each phase were gravimetrically diluted in new vials. Aqueous samples were diluted (50-fold for Tank 38H, 130-fold for Tank 46F) using 0.2 M nitric acid. Solvent samples were diluted (same dilution as aqueous samples) using 0.5 M modifier dissolved in Isopar[®] L. Diluted samples from Tank 38H were analyzed by routine gamma spectroscopy methods. To avoid a second dilution of the Tank 46F samples (due to the higher Cs-137 activity compared to Tank 38H), the Tank 46F samples were analyzed in a non-routine geometry that placed the sample vials farther from the detector. The non-

Table III. Composition of the Tank 38H and Tank 46F Samples

<u>Component</u>	<u>Concentration (molar)</u>		
	<u>Tank 38H</u>	<u>Tk 38H Simulant</u>	<u>Tank 46F</u>
Na ⁺	5.6	5.6	5.9
K ⁺	0.0015	0.0015	0.016
Rb ⁺	<0.002	--	<0.002
Cs ⁺	2.2x10 ⁻⁶	2.2x10 ⁻⁶	7.5x10 ⁻⁵
OH ⁻	0.59	0.70	3.2
NO ₃ ⁻	0.12	0.14	1.28
NO ₂ ⁻	0.060	0.071	0.21
AlO ₂ ⁻	0.011	0.013	0.20
CO ₃ ²⁻	1.59	1.87	0.25
SO ₄ ²⁻	0.35	0.41	0.042
Cl ⁻	0.083	0.10	0.047
F ⁻	<0.1	--	<0.1
PO ₄ ³⁻	0.002	0.0025	0.004
Oxalate	<0.1	--	<0.1
Formate	<0.2	--	<0.2
		<u>Concentration (mg/L)</u>	
As	<0.5	--	0.7
Ba	<3	--	<3
Ca	<1	--	<1
Cd	<2	--	<2
Cr	12	--	26
Hg	4.6	--	3.0
Mg	<1	--	<1
Mo	<3	--	29
Pb	<50	--	<50
Se	0.9	--	1.5
Si	87	--	105
Sr	<1	--	<1
M-99*	0.23	--	3.4
M-235	0.25	--	<0.01
M-236	0.06	--	<0.01
M-237	0.16	--	<0.01
M-238	15.	--	2.6
M-239	<0.01	--	<0.01
		<u>Concentration (d/m/mL)</u>	
Cs-137	1.97E7	1.20E6	6.99E8
Sr-90	7.4E5	--	2.9E5
Tc-99	<1.0E4	--	1.5E5
Pu-238	5.4E5	--	4.8E3
Pu-239/240	7.9E3	--	2 E2
Density (g/mL)	1.243	1.252	1.238
pH	>13	>13	>14

*Results reflect total mass for the indicated mass number. For example, M-238 includes U-238 and Pu-238.

Figure 1. Containers used in batch distribution test protocol

routine geometry was calibrated using radioactivity standards. For subsequent steps in the protocol, the remaining solvent was weighed and the amount of aqueous phase calculated.

One of the two initial Tank 38H samples spilled after the first scrub step. At that time, a third sample was extracted and scrubbed. The third sample and the remaining initial sample were processed together through the second scrub and three strip steps.

During the initial test with Tank 38H waste, poor recoveries of Cs-137 occurred in the three strip stages. Researchers attempted to discover the reason for the poor recovery by performing a test with simulated Tank 38H waste in a radiohood. Table III lists the composition of the simulated Tank 38H waste. The simulant test included new strip solution (1 mM nitric acid) and washing of all centrifuge tubes and glass vials with water, acetone, and ethanol prior to use. The simulant test proved successful and was followed by a repeat test in the Shielded Cells using the remaining Tank 38H waste in one centrifuge tube and Tank 38H simulant in the second tube. New strip solution and washed containers were used. This test proved unsuccessful, yielding low recoveries for cesium in the strip stages for both the waste and simulant. Remaining portions of the aqueous solutions in selected centrifuge tubes and glass vials were removed and the containers washed with 3.0 M nitric acid. Samples and washes were analyzed for Cs-137 by gamma spectroscopy.

A pair of centrifuge tubes containing water were handled and centrifuged simultaneously with the waste samples. Temperatures of the water tubes were recorded periodically during the protocol and were used to indicate the actual sample temperatures. It was noted that centrifuging raised the sample temperatures 3 to 5 °C above the cell ambient

temperature. Temperatures at the time of the eyedropper phase separation were recorded as the temperature for that distribution coefficient.

Analytical Methods

The SRTC Analytical Development Section provided all analyses by the following routine methods. Al, B, Ba, Ca, Cd, Co, Cr, Cu, Fe, La, Li, Mg, Mn, Mo, Na, Ni, P, Pb, Si, Sn, Sr, Ti, V, Zn, and Zr were measured by ICP-ES. Actinide and fission product isotopes were measured by ICP-MS. Common anions (nitrate, nitrite, sulfate, chloride, fluoride, phosphate, oxalate and formate) were measured by ion chromatography. K, Rb, Cs, Se, As, and Hg were measured by atomic adsorption or emission methods. Cs-137 was measured by gamma spectroscopy using a germanium detector. Sr-90 and Tc-99 were separated and quantified by scintillation counting. Pu isotopes were separated and counted on alpha plates.

Scrub and Strip Solutions

Scrub (0.050 M nitric acid) and strip (0.0010 M nitric acid) solutions were prepared by gravimetric dilution of standardized 0.100 M nitric acid (Fisher, Lot #003535-24) and verified correct by ADS standard titration before use.

For the initial tests, two small bottles of the stock strip solution were placed into the shielded facility, one for use with the Tank 38H sample and one for use with the Tank 46F sample. Following the distribution coefficient tests, the three bottles of strip solution (i.e., the stock solution bottle and the two small bottles that were placed in the cells) were titrated a second time. The pH as indicated by pH paper indicated all samples were approximately pH = 3, as expected for 1 mM nitric acid. ADS titration revealed the strip solution in the bottle used for the Tank 38H test was less than 1 mM. Since this concentration is near the detection limit and the amount of solution was limited, a more accurate estimate could not be made. The initial pH, as indicated by the titration curve, was approximately equal for all three solutions (suggesting that the Tank 38H strip solution was not much lower than 1 mM). Gamma spectroscopy of the contents of the Tank 38H strip solution bottle showed 130 d/m/mL of Cs-137. This level of contamination is not unexpected for a non-radioactive solution that has been used in the Shielded Cells. However, it is less than expected for contamination by a drop of high activity waste (One drop, or 0.05 mL, of the Tank 38H waste would contaminate the solution approximately 100X more than observed). This result suggests the solution was not contaminated by the waste used in the test. Additional analyses of the Tank 38H strip solution showed 1 mM nitrate (as expected), 0.6 mM formate, 0.6 mM chloride, and 0.3 mM sulfate, and 12 mM sodium. The concentrations of the anions identified account for only 33% of the sodium concentration. Because of the pH and titration observations, the remainder of the anion balance cannot be due to hydroxide or carbonate. For the Tank 38H repeat test in the Shielded Cells, the strip solution was verified accurate and uncorrupted before and after use.

Calculations

Distribution coefficients for cesium (D_{Cs}) were calculated by the following equation.

$$D_{Cs} = \frac{[\text{Cs-137 activity in the organic phase}]}{[\text{Cs-137 activity in the aqueous phase}]}$$

The ambient-temperature distribution coefficients were corrected to 25 °C using the following equation and enthalpy (H) values from Bonnesen, et al.¹

$$D_1 = D_0 \exp\left[\frac{H}{R}\left\{\frac{1}{T_1} - \frac{1}{T_0}\right\}\right]$$

where $R = 0.0083144 \text{ kJ/mol}\cdot\text{°K}$

The enthalpy values used were: extraction, 42.8 kJ/mole; scrub, 61.8 kJ/mole; and strip, 62.5 kJ/mole.

For each set of distribution data, a material balance for Cs-137 was calculated. That is, the activity in the initial solution was compared to sum of the activities in the two final solutions. For cases in which the containers were washed with 3.0 M nitric acid, the results for the acid wash were included to complete the material balance. Distribution coefficients were not calculated for test data in which the material balance did not agree within 15% of the expected total activity. Appendix A lists the measured and calculated results of the experiments.

Cesium Sorption on Glass Vials

The Cs-137 material balances for the strip stages of the Tank 38H waste indicated low recovery of cesium (i.e., the cesium activity found in the organic and aqueous phases after contact summed to only 30 to 60% of the value in the starting organic phase). Repeat analyses of both phases showed poor reproducibility of the analysis on the aqueous strip solutions but good reproducibility with the organic phase. Aqueous samples taken directly from the polypropylene centrifuge tubes provided good material balances (90-105% recovery). An empty glass vial used to transfer one of the aqueous samples and the corresponding polypropylene centrifuge tube were washed with 3.0 M nitric acid. The wash from the empty glass vial contained more activity than the total in aqueous samples removed from the vial, indicating cesium sorbed on the walls. The wash solution from the polypropylene centrifuge tube contained less than 5% of the activity in the aqueous samples removed from it, indicating negligible sorption in these containers.

Cesium sorption in the glass vials occurs slowly over several days. The good strip stage material balances obtained in the simulant test in the radiohood derived from the rapid preparation of the dilutions from the glass sample vials. In the radiohood, dilutions in 0.2 M nitric acid were completed within two hours of placing the samples in the glass vials. In the Shielded Cells tests, the dilutions were prepared two or more days after

placing the samples in the glass vials. In a controlled test, the aqueous strip solution from the Shielded Cells simulant test was placed in a glass vial and sampled within one hour and again after 24 and 48 hours. Figure 2 shows the results. The sample taken within one hour (5.77×10^6 d/m/mL) yielded a recovery of 94%. After 24 hours the activity dropped to 1.46×10^6 d/m/mL, or 64% recovery, and after 48 hours dropped to 1.35×10^6 d/m/mL for 63% recovery.

RESULTS AND DISCUSSION

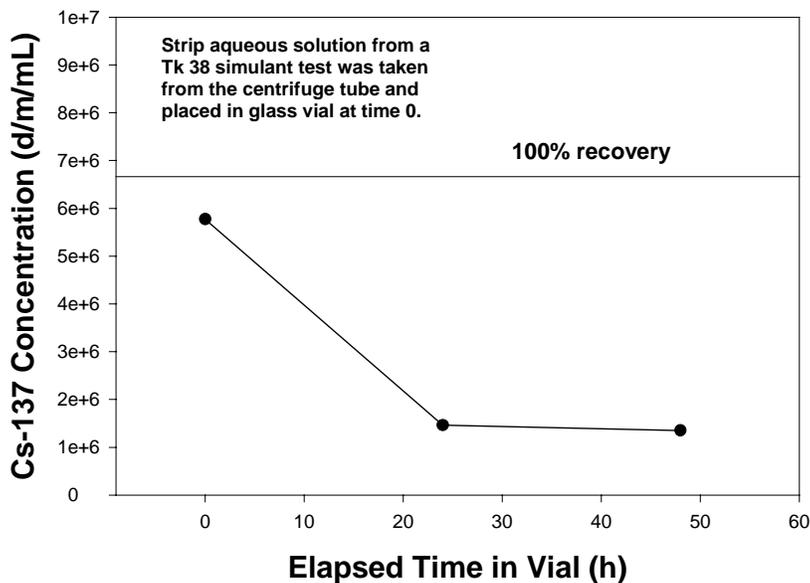
Dissolved Salt Waste Composition

The Tank 38H sample reflects the potential variability of waste compositions that may arise from salt cake dissolution. The Tank 46F sample shows some of the same variability but to a lesser extent because it was diluted with supernate from Tanks 37F and 44H. The following summarize significant differences between the dissolved salt cake samples and average waste.⁶

- Cesium and potassium concentrations are lower in salt cake.
- Hydroxide and nitrate concentrations are lower in salt cake.
- Carbonate and sulfate concentrations are higher in salt cake.

The Tank 38H sample contained an unusually high amount of carbonate ion. This may result from the sampling method that obtained salt cake from near the surface of the salt a deposit. If this layer was ever in contact with air, carbonated salts could have

FIGURE 2. Decrease in Soluble Cs-137 in Glass Vial



accumulated at the surface from the reaction of carbon dioxide in the air with sodium hydroxide in the waste.

Cs-137 Distribution Coefficients

Table IV summarizes the temperature-corrected distribution coefficients for the two waste samples. The table also includes predicted D_{Cs} values for the extraction step and the D_{Cs} values that form the basis for the CSSX baseline flowsheet.⁶ The predicted values derive from an ORNL model based on data from simulated waste tests. The model predicts values at 25 °C. Strip distribution coefficients for the Tank 38H sample were not calculated for tests affected by Cs-137 sorption on glass transfer vials.

Extraction

The extraction distribution coefficients listed in Table IV exceed the values used in the CSSX flowsheet⁶ and agree within 36% to the predicted values. The results indicate that the CSSX process will successfully extract cesium from waste solutions derived from dissolved salt cake. The agreement between real waste and the model in these data is closer than previous comparisons in which they differed by as much as 300%.³ The improvement is likely due to better technique from increased experience in the execution of the ESS protocol in the shielded facility. The remaining discrepancy is not due to the highly unusual composition of the Tank 38H sample.

Usually, high-level waste supernates contain mostly monoanions, such as nitrate, nitrite, chloride, and hydroxide. The Tank 38H waste is predominantly dianions (carbonate and sulfate). Although the model is based on data from simulants containing only monoanions (hydroxide, nitrate, nitrite, and chloride), the agreement is excellent between the model ($D_{\text{predicted}} = 9.6$) and the Tank 38H simulant ($D_{\text{found}} = 10.0 \pm 0.6$, see Appendix A).

TABLE IV. Cs-137 Distribution Coefficients for Tank 38H and Tank 46F Waste

<u>Step</u>	<u>Flowsheet Basis</u>	<u>D_{Cs} Values</u>			
		<u>Tank 38H</u>		<u>Tank 46F</u>	
		<u>Predicted</u>	<u>Measured</u>	<u>Predicted</u>	<u>Measured</u>
Extraction	>8	9.6	13 ± 3	22.8	16.6 ± 0.5
Scrub 1	>0.6		1.7 ± 0.5		1.48 ± 0.36
Scrub 2			1.8 ± 0.9		1.63 ± 0.11
Strip 1	<0.16		0.108 ± 0.031		0.107 ± 0.042
Strip 2			0.073*		0.071 ± 0.011
Strip 3			0.077*		0.055 ± 0.013

*Single data points.

Scrub

The scrub distribution coefficients for both waste samples (1.4 to 1.8) met the requirement of the CSSX flowsheet (>0.6) and are similar to previously measured values for simulants and actual waste (normally 1.0 to 2.0).³⁻⁴

Strip

The strip distribution coefficients for the Tank 38H (0.073 to 0.108) and 46F (0.055 to 0.107) dissolved salt cake compare favorably to the flowsheet basis maximum value of 0.16. The slight downward trend for subsequent strips agrees with previous work with real waste and simulants.³⁻⁴ High values for the strip D_{Cs} values due to alkaline carry-over after a single scrub step reported previously were likely avoided by adding the second scrub step to the ESS protocol.

CONCLUSIONS

Researchers characterized high-level waste solutions derived from dissolved salt cake from the SRS tank farm and measured the cesium distribution coefficients for extraction, scrub, and stripping steps of the CSSX flowsheet. The measurements used two SRS high-level waste samples derived entirely or in part from salt cake. A sample of Tank 38H salt cake provided a waste solution suitable for testing (5.6 M Na^+). A dilute solution of Tank 46F salt cake was combined with concentrated supernate solution from Tank 37H/44F to achieve the desired salt concentration for testing. The chemical compositions of both samples differed significantly from average waste, but the differences were largely predictable from solubility considerations. Extraction, scrub, and strip D_{Cs} values for both samples exceeded process requirements and agreed well with model predictions.

Sorption of cesium occurred in glass vials used for holding aqueous strip samples prior to dilution and analysis. Cesium sorption appears to occur to a significant extent only when the samples contain low levels of cesium (such as Tank 38H waste and Tk 38H simulant) in a low acidity or low ionic strength matrix (such as 1 mM nitric acid). No sorption was evident in samples of solvent, concentrated waste, dilutions in 0.2 M nitric acid, or scrub solution (0.05 M nitric acid). We recommend that future testing avoid the use of glass vials for transferring samples. The simplest solution to the problem is to make dilutions directly from the polypropylene centrifuge tubes. We also recommend calculating cesium recoveries or material balances as a diagnostic tool for detecting sorption

FUTURE WORK

We recommend the following additional investigations to improve understanding of the CSSX process chemistry relative to salt cake waste.

- ESS batch tests with waste from older, stagnant tanks will extend the database on salt cake (compared to supernate). The two wastes tested in this study came from active evaporator systems and represent recent tank farm waste.

- Contactor tests using large samples of dissolved salt cake can uncover problems from minor components that batch tests will not reveal. The planned salt cake removal from Tank 37H represents an opportunity for such a test.

ACKNOWLEDGMENTS

The authors appreciate the calculations of extraction D_{Cs} values provided by Dr. Laetitia H. Delmau of ORNL. We also thank the following ORNL researchers for many useful discussions and suggestions in resolving the cesium sorption problem: Dr. Leon N. Klatt, Dr. Bruce A. Moyer, Dr. Laetitia H. Delmau, and Dr. Peter V. Bonnesen.

REFERENCES

1. P. V. Bonnesen, L. H. Delmau, B. A. Moyer, and R. A. Leonard, "A Robust Alkaline-Side CSEX Solvent Suitable for Removing Cesium from Savannah River High Level Waste," *Solvent Extraction and Ion Exchange*, **18**, 1079 (2000).
2. S. G. Campbell, M. W. Geeting, C. W. Kennell, J. D. Law, R. A. Leonard, M. A. Norato, R. A. Pierce, T. A. Todd, D. D. Walker, and W. R. Wilmarth, "Demonstration of Caustic-Side Solvent Extraction with Savannah River Site High Level Waste," WSRC-TR-2001-0023, Rev. 0, April 19, 2001.
3. W. R. Wilmarth, J. T. Mills, V. H. Dukes, M. C. Beasley, A. D. Coleman, C. C. Diprete, and D. P. Diprete, "Caustic-Side Solvent Extraction Batch Distribution Coefficient Measurements for Savannah Site High Level Wastes," WSTRC-TR-2001-00409, Rev. 0, August 24, 2001.
4. L. H. Delmau, T. J. Haverlock, G. Levitskaia, F. V. Sloop, and B. A. Moyer, "Caustic-Side Solvent Extraction Chemical and Physical Properties: Equilibrium Modeling of Distribution Behavior," CERS/SR/SX/018, Rev. 0, April 16, 2001.
5. R. F. Swingle, "Characterization of the Tank 38H Variable Depth and Core Samples," WSRC-RP-2001-00574, Rev. 0, May 2, 2001.
6. R. A. Dimenna, H. H. Elder, J. R. Fowler, R. C. Fowler, M. V. Gregory, T. Hang, R. A. Jacobs, P. K. Paul, J. A. Pike, P. L. Rutland, F. G. Smith III, S. G. Subosits, G. A. Taylor, S. G. Campbell, and F. A. Washburn, "Bases, Assumptions, and Results of the Flowsheet Calculations for the Decision Phase Salt Disposition Alternatives," WSRC-RP-99-00006, Rev. 3, May 2001.

APPENDIX A
Measured and Calculated Results

Tank 38 H Sample					
		Initial Test in Shielded Cells			
Step		Sample #			
		1	2	3	
Extraction					
Experimental data	Temp (deg C)	23.6	23.6	25.9	
	Aqueous activity (d/m/mL)	3.68E+06	3.60E+06	3.58E+06	
	Organic activity (d/m/mL)	4.42E+07	4.47E+07	4.46E+07	
	D-value	12.0	12.4	12.5	
	D-value corrected to 25 deg C	11.1	11.4	13.1	
Cs-137 material balance	Initial (d/m)	5.91E+08	5.91E+08	5.91E+08	
	Final (d/m)	5.52E+08	5.55E+08	5.53E+08	
	% recovered	93	94	94	
Scrub #1					
Experimental data	Temp (deg C)	27.6	25.2	26.5	
	Aqueous activity (d/m/mL)	2.77E+07	2.67E+07	3.48E+07	
	Organic activity (d/m/mL)	3.78E+07	3.91E+07	3.83E+07	
	D-value	1.36	1.46	1.10	
	D-value corrected to 25 deg C	1.69	1.49	1.25	
Cs-137 material balance	Initial (d/m)	4.42E+08	4.47E+08	4.46E+08	
	Final (d/m)	4.33E+08	4.44E+08	4.53E+08	
	% recovered	98	99	101	
Scrub #2					
Experimental data	Temp (deg C)	21.3		21.3	
	Aqueous activity (d/m/mL)	1.89E+07		1.97E+07	
	Organic activity (d/m/mL)	3.46E+07		3.40E+07	
	D-value	1.83		1.73	
	D-value corrected to 25 deg C	1.34		1.26	
Cs-137 material balance	Initial (d/m)	3.78E+08		3.83E+08	
	Final (d/m)	3.84E+08		3.79E+08	
	% recovered	102		99	
Strip #1*					
Experimental data	Temp (deg C)	24.3		23.1	
	Aqueous activity (d/m/mL)	1.20E+08		1.89E+07	
	Organic activity (d/m/mL)	1.10E+07		1.05E+07	
	D-value	0.0917			
	D-value corrected to 25 deg C	0.086	Error in material balance.		
Cs-137 material balance	Initial (d/m)	3.46E+08		3.40E+08	
	Final (d/m)	3.50E+08		1.43E+08	
	% recovered	101		42	
Strip #2					
Experimental data	Temp (deg C)	24.8		24.2	
	Aqueous activity (d/m/mL)	4.46E+06		5.49E+06	
	Organic activity (d/m/mL)	3.07E+06		2.81E+06	
	D-value				
	D-value corrected to 25 deg C	Error in material balance.			
Cs-137 material balance	Initial (d/m)	1.10E+08		1.05E+08	
	Final (d/m)	3.96E+07		3.91E+07	
	% recovered	36		37	
Strip #3					
Experimental data	Temp (deg C)	26.5		25.3	
	Aqueous activity (d/m/mL)	1.31E+06		1.24E+06	
	Organic activity (d/m/mL)	7.65E+05		6.46E+05	
	D-value				
	D-value corrected to 25 deg C	Error in material balance.			
Cs-137 material balance	Initial (d/m)	3.07E+07		2.81E+07	
	Final (d/m)	1.03E+07		8.94E+06	
	% recovered	33		32	
* Sufficient solution was available in the centrifuge tube to obtain the Sample #1 result.					

Tank 38H Repeat Tests						
Step		Simulant in Radiohood		Shielded Cells Repeat		
		1	2	Simulant	Tk 38H	
Extraction						
Experimental data	Temp (deg C)	23.8	21.8	15.9	16.2	
	Aqueous activity (d/m/mL)	2.55E+05	2.28E+05	1.68E+05	1.85E+06	
	Organic activity (d/m/mL)	2.58E+06	2.79E+06	3.08E+06	5.21E+07	
	D-value	10.1	12.2	18.3	28.2	
D-value corrected to 25 deg C		9.4	10.1	10.6	16.7	
Cs-137 material balance	Initial (d/m)	3.60E+07	3.60E+07	3.60E+07	6.21E+08	
	Final (d/m)	3.35E+07	3.47E+07	3.58E+07	5.77E+08	
	% recovered	93	97	100	93	
Scrub #1						
Experimental data	Temp (deg C)	19.6	19.6	11.9	12.3	
	Aqueous activity (d/m/mL)	1.26E+06	1.16E+06	5.11E+05	7.31E+06	
	Organic activity (d/m/mL)	2.68E+06	2.68E+06	3.04E+06	5.05E+07	
	D-value	2.13	2.31	5.9	6.9	
D-value corrected to 25 deg C		1.34	1.46	1.9	2.3	
Cs-137 material balance	Initial (d/m)	2.58E+07	2.79E+07	3.08E+07	5.21E+08	
	Final (d/m)	2.93E+07	2.91E+07	3.14E+07	5.20E+08	
	% recovered	114	104	102	100	
Scrub #2						
Experimental data	Temp (deg C)	21.1	20.3	15.1	14.1	
	Aqueous activity (d/m/mL)	1.02E+06	1.07E+06	4.12E+05	6.69E+06	
	Organic activity (d/m/mL)	2.33E+06	2.38E+06	2.87E+06	4.87E+07	
	D-value	2.28	2.22	7.0	7.3	
D-value corrected to 25 deg C		1.64	1.49	3.0	2.8	
Cs-137 material balance	Initial (d/m)	2.68E+07	2.68E+07	3.04E+07	5.05E+08	
	Final (d/m)	2.53E+07	2.59E+07	2.95E+07	5.00E+08	
	% recovered	95	97	97	99	
Strip #1						
Experimental data	Temp (deg C)	22.1	20.4	17.6	16.1	
	Aqueous activity (d/m/mL)	5.68E+06	6.20E+06	5.77E+06	9.82E+07	
	Organic activity (d/m/mL)	1.14E+06	9.99E+05	1.55E+06	2.77E+07	
	D-value	0.201	0.161	0.269	0.282	
D-value corrected to 25 deg C		0.157	0.109	0.141	0.130	
Cs-137 material balance	Initial (d/m)	2.33E+07	2.38E+07	2.87E+07	4.87E+08	
	Final (d/m)	2.28E+07	2.24E+07	2.70E+07	4.73E+08	
	% recovered	98	94	94	97	
Strip #2						
Experimental data	Temp (deg C)	21.8	20.2	16.3	16.5	
	Aqueous activity (d/m/mL)	3.35E+06	2.84E+06	4.28E+06	7.78E+07	
	Organic activity (d/m/mL)	3.68E+05	3.44E+05	5.87E+05	1.19E+07	
	D-value	0.110	0.121	0.137	0.153	
D-value corrected to 25 deg C		0.084	0.080	0.064	0.073	
Cs-137 material balance	Initial (d/m)	1.14E+07	9.99E+06	1.55E+07	2.77E+08	
	Final (d/m)	1.04E+07	9.12E+06	1.44E+07	2.75E+08	
	% recovered	91	91	93	99	
Strip #3						
Experimental data	Temp (deg C)	23	21.7	20.2	18.3	
	Aqueous activity (d/m/mL)	1.19E+06	1.03E+06	3.58E+05	3.46E+07	
	Organic activity (d/m/mL)	9.85E+04	8.73E+04	1.76E+05	4.74E+06	
	D-value	0.083	0.085		0.137	
D-value corrected to 25 deg C		0.070	0.064		0.077	
Cs-137 material balance	Initial (d/m)	3.68E+06	3.44E+06	5.87E+06	1.19E+08	
	Final (d/m)	3.37E+06	2.93E+06	2.48E+06	1.17E+08	
	% recovered	91	85	42	98	

Tank 46F Sample						
	Step		Sample	Sample	Average	Error
			1	2		
Extraction						
	Experimental data	Temp (deg C)	22.6	22.6		
		Aqueous activity (d/m/mL)	9.20E+07	9.54E+07		
		Organic activity (d/m/mL)	1.79E+09	1.78E+09		
		D-value	19.5	18.7		
	D-value corrected to 25 deg C		16.9	16.2	16.6	0.5
	Cs-137 material balance	Initial (d/m)	2.10E+10	2.10E+10		
		Final (d/m)	2.07E+10	2.07E+10		
Scrub #1						
	Experimental data	Temp (deg C)	25.8	23.4		
		Aqueous activity (d/m/mL)	1.03E+09	1.06E+09		
		Organic activity (d/m/mL)	1.67E+09	1.48E+09		
		D-value	1.62	1.40		
	D-value corrected to 25 deg C		1.73	1.22	1.48	0.36
	Cs-137 material balance	Initial (d/m)	1.79E+10	1.78E+10		
		Final (d/m)	1.88E+10	1.69E+10		
Scrub #2						
	Experimental data	Temp (deg C)	27.2	25.7		
		Aqueous activity (d/m/mL)	1.02E+09	8.20E+08		
		Organic activity (d/m/mL)	1.32E+09	1.32E+09		
		D-value	1.29	1.61		
	D-value corrected to 25 deg C		1.55	1.71	1.63	0.11
	Cs-137 material balance	Initial (d/m)	1.67E+10	1.48E+10		
		Final (d/m)	1.52E+10	1.48E+10		
Strip #1						
	Experimental data	Temp (deg C)	29.1	23.3		
		Aqueous activity (d/m/mL)	4.01E+09	4.62E+09		
		Organic activity (d/m/mL)	3.88E+08	4.13E+08		
		D-value	0.097	0.089		
	D-value corrected to 25 deg C		0.136	0.077	0.107	0.042
	Cs-137 material balance	Initial (d/m)	1.32E+10	1.32E+10		
		Final (d/m)	1.19E+10	1.34E+10		
Strip #2						
	Experimental data	Temp (deg C)	27.4	24.7		
		Aqueous activity (d/m/mL)	1.62E+09	1.51E+09		
		Organic activity (d/m/mL)	1.04E+08	9.75E+07		
		D-value	0.064	0.065		
	D-value corrected to 25 deg C		0.079	0.063	0.071	0.011
	Cs-137 material balance	Initial (d/m)	3.88E+09	4.13E+09		
		Final (d/m)	4.28E+09	4.00E+09		
Strip #3						
	Experimental data	Temp (deg C)	27.5	25.3		
		Aqueous activity (d/m/mL)	4.02E+08	3.87E+08		
		Organic activity (d/m/mL)	2.09E+07	1.75E+07		
		D-value	0.052	0.045		
	D-value corrected to 25 deg C		0.064	0.046	0.055	0.013
	Cs-137 material balance	Initial (d/m)	1.04E+09	9.75E+08		
		Final (d/m)	1.01E+09	9.49E+08		