ornl

OAK RIDGE NATIONAL LABORATORY

LOCKHEED MARTIN

ORNL/TM-1999/233

Thermal and Chemical Stability of Crystalline Silicotitanate Sorbent

P. A. Taylor
C. H. Mattus

MANAGED AND OPERATED BY
LOCKHEED MARTIN ENERGY RESEARCH CORPORATION
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

ORNL-27 (3-96)

ORNL/TM-1999/233

Chemical Technology Division

THERMAL AND CHEMICAL STABILITY OF CRYSTALLINE SILICOTITANATE SORBENT

P. A. Taylor
C. H. Mattus

Date of Publication: October 1999

Prepared by the
OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37831-6285
managed by
LOCKHEED MARTIN ENERGY RESEARCH CORP.
for the
U.S. DEPARTMENT OF ENERGY
under contract DE-AC05-96OR22464

CONTENTS

LIST OF FIGURES v

LIST OF TABLES v

ABSTRACT 1

1. INTRODUCTION 2

2. MATERIALS AND METHODS 2
   2.1 Crystalline Silicotitanate 2
   2.2 Simulant Solution 3
   2.3 Equipment 3
   2.4 Experimental Methods 6

3. RESULTS 7
   3.1 Loading Tests 7
   3.2 Leaching Tests 10
   3.3 Nonradioactive Leaching Tests 13

4. DISCUSSION 16

5. REFERENCES 17

6. APPENDIXES 19

   Appendix A: ANALYTICAL RESULTS AND CALCULATIONS FOR LOADING TESTS 21

   Appendix B: ANALYTICAL RESULTS AND CALCULATIONS FOR LEACHING TESTS 25

   Appendix C: X-RAY DIFFRACTION PATTERNS FOR CST SAMPLES STORED IN SRS SUPERNATE SIMULANT 31

LIST OF FIGURES

1. Environ Shaker, which was used to mix CST and supernate simulant at various temperatures 4

2. Sample of CST on stainless steel screen 5

3. CST trapped within folded screen 5

4. Oven and stirrer used to contact CST and supernate simulant at 120°C 5

5. Comparison of rates of cesium uptake by CST, using two contacting methods 7

6. Effect of temperature on cesium sorption by CST 9

7. Results of leaching tests 12

8. Leaching of silicon into SRS supernate simulant at various temperatures 15

9. Leaching of titanium into SRS supernate simulant at various temperatures 15

LIST OF TABLES

1. Composition of SRS supernate simulant 3

2. Cesium loading results for CST in supernate simulants 8

3. Results of gamma counting at RMAL 10

4. Results of room-temperature loading tests 11

5. Results of leaching tests 11

6. Volume of supernate simulant in each flask, after 60 days of leaching 12

7. Results of room-temperature reloading tests, following the leaching tests 13

8. Soluble metals in SRS supernate simulant in contact with CST 14

9. Comparison between batch test results and model predictions 16

THERMAL AND CHEMICAL STABILITY OF CRYSTALLINE SILICOTITANATE SORBENT

P. A. Taylor and C. H. Mattus

ABSTRACT

The Savannah River Site (SRS) is evaluating technologies for removing cesium-137 (^{137}Cs) from the supernate solutions stored in the high-level waste tanks at the site. Crystalline silicotitanate sorbent (IONSIV IE-911®, UOP Molecular Sieves, Mt. Laurel, NJ) is very effective for removing cesium from high-salt solution, such as the SRS supernates, and is currently being used at Oak Ridge National Laboratory to remove radioactive cesium from similar solutions. Because of the extremely high loading of ^{137}Cs that would be expected for the large columns of crystalline silicotitanate (CST) that would be used for treating the SRS supernate, any loss of flow or cooling to the columns could result in high temperatures within the column from radiolytic heating. The ability of CST to retain previously loaded cesium while in contact with SRS tank supernates at various temperatures was determined by performing bench scale simulant tests using CST samples that were loaded with stable cesium and radioactive cesium tracer. These results were compared with those obtained from loading tests at the same temperatures.

The loading of cesium on CST decreased as the temperature was increased. At temperatures of 80 and 120°C, the amount of cesium loaded on the CST was reduced by 72 and 83%, respectively, as compared with the loading achieved at room temperature. The CST that was loaded with cesium at room temperature and then heated to higher temperatures leached cesium back into the supernate simulant solution. The leaching rate was about as rapid as the original cesium uptake, and the final concentrations at each temperature tested were approximately the same as those for CST loaded at that temperature.

Following storage at high temperatures (50 to 120°C) in the SRS supernate simulant for 60 days, the CST did not reload any cesium after the temperature was reduced to 22°C. Further work would be needed to determine the full range of storage time and temperature profiles that would impact the loading of cesium onto the CST. X-ray diffraction patterns did not show any significant change in the crystal structure of the CST, so it is unlikely that chemical changes in the bulk CST prevented the cesium from reloading. Chemical analyses of the simulant solutions showed that silicon and smaller amounts of other metals were leaching into the simulant from the CST. Several of the metals increased and then decreased in concentration, which indicates that competing dissolution and precipitation reactions were probably occurring. The leaching and reprecipitation of silicon and other metals from the CST might physically block the pores of the CST, thereby preventing cesium from reloading onto the CST. Based on the amount of aluminum that was lost from the supernate simulant in the nonradioactive leaching tests, silicon loses from the CST could be as high as 4.2 ±0.5 wt %, if all of the silicon that was leached from the CST precipitated as a sodium aluminosilicate compound. UOP has reported that the batch of CST used in these tests contains 1.9 wt % excess silicon trapped in the particles, so the hot supernate simulant is probably mostly leaching this excess silicon from the CST.

1. INTRODUCTION

The Savannah River Site (SRS) is evaluating technologies for removing cesium-137 (^{137}Cs) from the supernate solutions stored in the high-level waste tanks at the site. Crystalline silicotitanate (CST) is currently being used at Oak Ridge National Laboratory (ORNL) to remove ^{137}Cs from liquid low-level waste, which has a composition similar to that of the SRS supernates.[1] Work conducted at ORNL last year showed that higher temperatures could cause leaching of previously loaded cesium from CST, but these tests could not establish whether the leaching was due to equilibrium shifts caused by temperature changes or by degradation of the CST.[2] Because of the extremely high loading of ^{137}Cs that would be expected for large columns of CST used in treating the SRS supernate, any loss of flow or cooling to the columns could result in high temperatures within the column from radiolytic heating.

CST sorbent has been used to remove cesium from a wide range of solutions, including high-salt basic,[1,3,4] high-salt acidic,[4,5] and neutral groundwaters as well as process wastewaters.[6] These results illustrate the chemical stability and cesium affinity of CST; however, chemical stability tests have shown that CST partially converts to zeolite when stored at 95 °C in high-salt, high-pH solutions for 3 months, but is stable when stored at 60°C.[3] This change could result in previously loaded cesium being released back into solution.

The ability of CST to retain previously loaded cesium while in contact with SRS tank supernates at various temperatures was determined by performing bench-scale simulant tests using CST samples that were loaded with stable cesium and radioactive cesium tracer. The results from these tests were compared with those from loading tests conducted at the same temperatures. X-ray diffraction and inductively-coupled plasma emission spectroscopy were used to probe for chemical changes in the CST. This work was targeted to a Technical Task Request[7] from SRS, and the methods for performing the work were initially described in an ORNL Technical Task Plan.[8]

2. MATERIALS AND METHODS

2.1 CRYSTALLINE SILICOTITANATE

The CST used in all of these tests was the commercial, granular form of the sorbent, IONSIV® IE-911 (UOP LLC, Mt. Laurel, NJ). The as-received sorbent is slightly acidic, so dilute sodium hydroxide was used to stabilize its pH at 13 before use. The pretreatment pH matches the pH of wastewaters being treated at ORNL, but is less basic than the supernate simulant (pH = 14) that was used in these tests. The CST was also backwashed with tap water to remove any fines generated during shipping. A sample of CST from UOP lot 999098810005, pretreated as described above, was used in each of the tests in this project. The CST sample was air dried after pretreatment, and then the moisture content of three subsamples was determined by drying at 102 °C until a stable weight was obtained. The pretreated, air-dried material had a moisture content of 6.915 wt % (average of three samples), as compared with a moisture content of 6.924 wt % for the as-received material from the same drum.[9]

2.2 SIMULANT SOLUTION

An average supernate simulant, with a total sodium concentration of 5.6 *M*, was prepared using a recipe from SRS [10] (see Table 1). A small amount of undissolved solids was present in each batch of supernate simulant that was prepared, so the simulant was filtered prior to use. The cesium concentration of the supernate simulant used in these tests was 50 mg/L, which is higher than that typically found in actual SRS supernate solutions. The cesium concentration and CST amounts for these batch tests were chosen so that the equilibrium cesium concentration was about 22 mg/L, which is a typical concentration in the actual SRS supernate. The cesium concentration on the CST in these tests would be about the same as would be expected for CST in a fully loaded column that was treating SRS supernate. The simulant used for the loading and leaching tests was spiked with about 1 μCi of ^{137}Cs per liter.

Table 1. Composition of SRS supernate simulant

Component	Concentration (*M*)
Na^+	5.6
K^+	0.015
Cs^+	0.00038
OH^-	1.91
NO_3^-	2.14
NO_2^-	0.52
AlO_2^-	0.31
CO_3^{2-}	0.16
SO_4^{2-}	0.15
Cl^-	0.025
F^-	0.032
PO_4^{3-}	0.010
$C_2O_4^{2-}$	0.008
SiO_3^{2-}	0.004
MoO_4^{2-}	0.0002

2.3 EQUIPMENT

Samples of CST and supernate simulant were mixed in two Model 3527-5 Environ Shakers (Lab-Line Instrument, Inc., Melrose Park, IL) at temperatures from 21 to 80°C. The samples, which had been placed inside capped Teflon flasks, were swirled at a rotation speed of about 120 rpm. The temperature of the samples was maintained within ±0.2°C by a built-in, forced-air convection heater with a temperature controller. The shakers have a clear plastic lid to retain the heated air. Figure 1 shows one of the shakers, with the lid raised and one Teflon flask mounted in a holder. The temperature inside the shakers was also monitored using VWR Traceable Digital

Fig. 1. Environ Shaker, which was used to mix CST and supernate simulant at various temperatures.

Thermometers (VWR Scientific Products, West Chester, PA) that were factory calibrated on February 22, 1999 using a NIST traceable standard.

The CST loading and leaching tests at 120°C were performed in an older model GCA Corporation Gravity Convection Oven. The oven had a simple on-off temperature controller, so the temperature typically varied within a range of ±2°C over the short term. However, it would drift over longer time periods, so periodic adjustments were required.
Each sample of CST was contained within a piece of 100-mesh stainless steel screen, which was placed in a screened basket on the end of a stainless steel rod. The rods extended through holes in the top of the oven to a Phibbs & Bird (Richmond, VA) Model 7790-400 six-place stirrer. The baskets, containing the CST, were rotated at about 120 rpm within the simulant. Figure 2 shows a sample of CST on a screen, which was then folded twice in each direction to trap the CST (see Fig. 3). Figure 4 shows the oven and stirrer, with the Teflon bottles in a containment pan, and one stirring rod raised to display the screened basket on the end of the rod.

Samples of nonradioactive supernate simulant and CST were placed in capped Teflon bottles, and stored, without mixing, in Model 338F Isotemp ovens (Fisher Scientific Co., Pittsburgh, PA) at 80 and 120°C and in a Model 1350 GM oven (VWR Scientific Products) at 50°C. These ovens had digital temperature controllers that maintained the temperature within ±0.2°C.

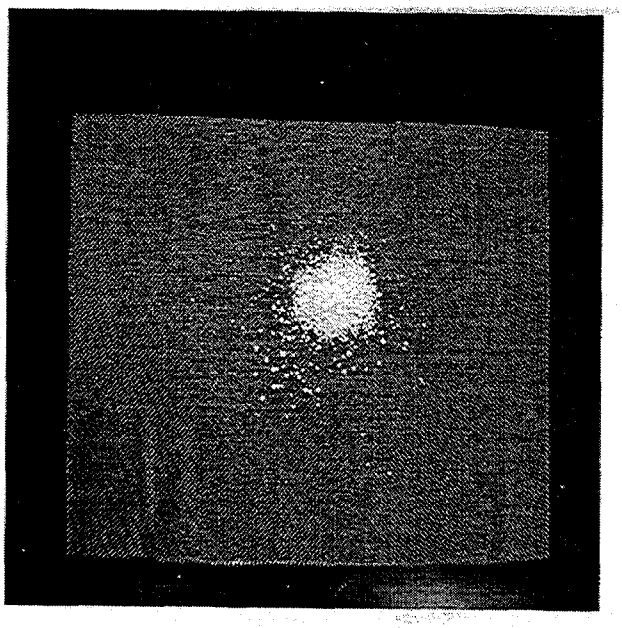


Fig. 2. Sample of CST on stainless steel screen.

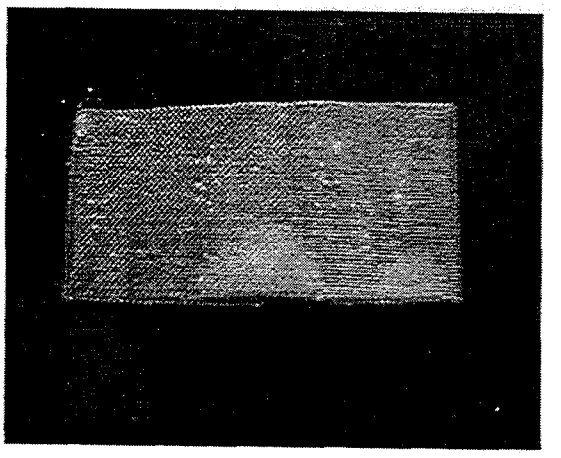


Fig 3. CST trapped within folded screen.

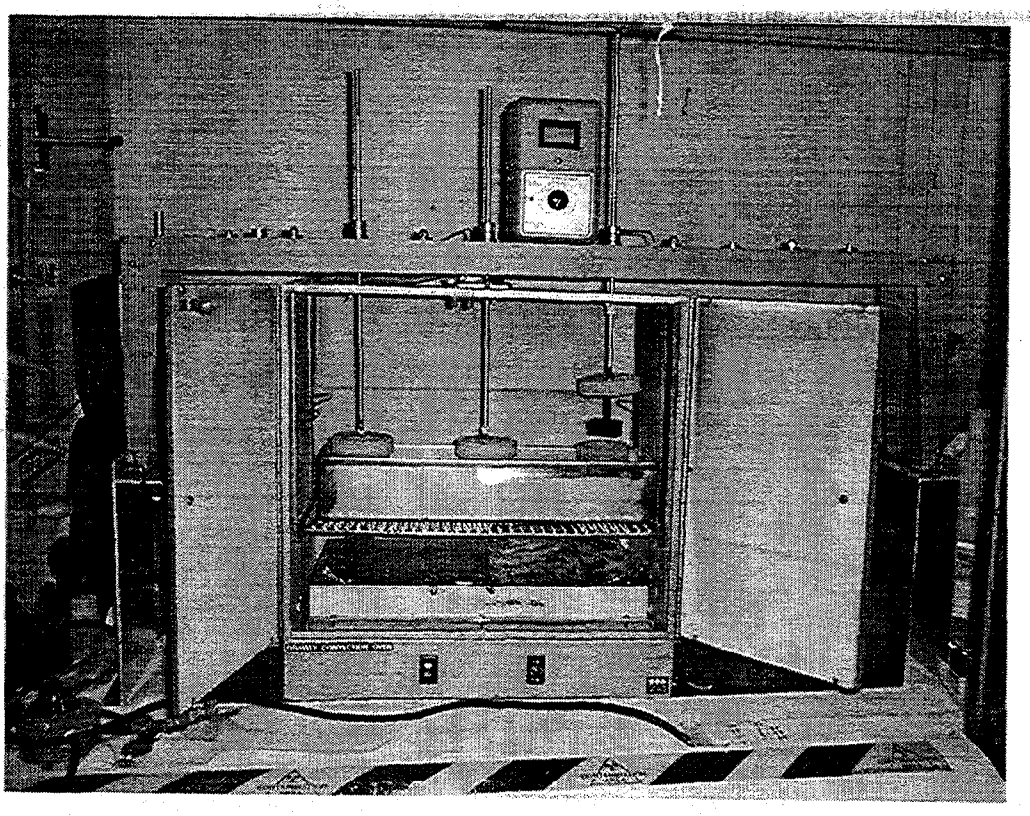


Fig. 4. Oven and stirrer used to contact CST and supernate simulant at 120°C.

2.4 Experimental Methods

Batch loading tests were performed at temperatures of 30, 40, 50, 80, and 120°C, using 0.1 g of CST and 100 mL of supernate simulant. Most of the tests were performed in Teflon flasks with screw-on lids, which were mixed in one of the shakers (described above) for 4 days. The tests at 120°C were conducted in heavy-walled Teflon bottles inside the convection oven. A hole was drilled in the center of each lid for the stirring rod to pass through. Each rod had a basket on the end, which held the folded screen containing the CST. The rod fit snuggly through the hole in the bottle lid, in order to minimize evaporation of the solution; however, the rod could rotate without turning the bottle. At each temperature, two containers with CST and supernate, plus one control sample with only supernate, were used.

The CST and simulant were contacted for 4 days, and then two samples of the supernate from each flask were filtered through 0.2-μ-m-pore size, nylon syringe filters. A 2.00-mL portion of each filtered sample was transferred into a polyethylene counting tube, centrifuged for about 30 s to ensure that no droplets were left on the sides of the tubes, and then gamma counted for 1000 s. The samples were counted at an energy level of 664.7 keV, using a Canberra Industries, Inc. (Meriden, CT) series 90 gamma spectrometer, with a shielded germanium detector, to determine the concentration of ^{137}Cs. Three samples of the starting supernate simulant and three background rates were counted for each series of samples. The net counts, the cesium concentration in each sample, the cesium loading on the CST, and the apparent distribution coefficient (K_d) were calculated as follows:

Net counts = (gross counts) - (average background counts)
[Cs] = (net counts) (50 mg/L Cs) / (average starting solution counts)
Cs loading on CST = (50 - [Cs]) (0.1 L) / (CST weight)
K_d = (Cs loading on CST) / [Cs].

The leaching tests were performed in a similar manner to the loading tests, except that the CST and supernate containers were contacted at room temperature for 4 days to load the CST with cesium. The containers were then heated to 50, 80, or 120°C and stored, while being mixed, for a total of 60 days. Samples of supernate were removed, filtered, counted, and then returned to their original container, after storage times of 1, 2, 4, 16, 29, and 60 days. Following completion of the leaching tests, all of the containers were mixed at room temperature for 4 days and then sampled to determine how much of the cesium previously leached from the CST had been reloaded. For both the loading and leaching tests, one of the samples from a container at each temperature was submitted to the Radioactive Materials Analytical Laboratory (RMAL) at ORNL for analysis by gamma spectroscopy.

Teflon bottles containing nonradioactive supernate simulant (50 mg/L Cs and no ^{137}Cs) and CST (16 g CST in 240 mL of simulant) were stored in ovens at 50, 80, and 120°C. Samples of the supernate were analyzed for dissolved metals by inductively-coupled plasma emission spectroscopy (ICP), and samples of the CST were analyzed by x-ray diffraction to detect any changes in the CST crystal structure after storage times of 1, 7, 21, and 35 days. The x-ray diffraction analysis was performed by the Lockheed Martin Energy System's, Analytical Services Organization. The ICP analyses were performed by C. H. Mattus using a model 61E Trace ICP

from Thermo Jarrell Ash, following standard EPA method SW846-6010B. Standard reference samples from the National Institute of Standards and Technology (NIST) were analyzed along with the samples to verify the accuracy of the results. The results obtained for the reference standards were within 5% of the certified concentrations for Al, Si, and Ti.

3. RESULTS

3.1 LOADING TESTS

A room-temperature loading test was performed using both of the contacting methods (swirling CST and supernate in a flask, and rotating a screened basket of CST through the supernate) to determine the time required to reach equilibrium. Each contacting method resulted in a constant cesium concentration within 4 days (see Fig. 5).

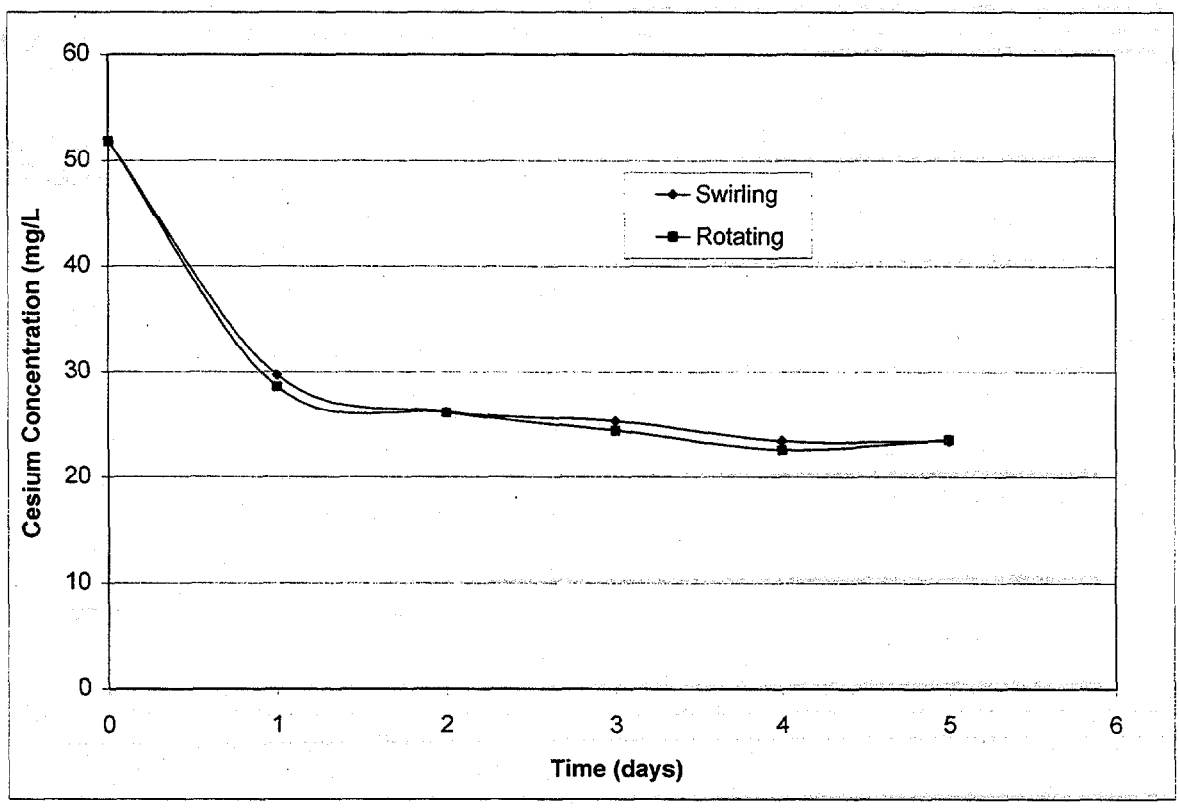


Fig. 5. Comparison of rates of cesium uptake by CST, using two contacting methods.

The first two loading tests were performed with the Environ Shakers, at temperatures of 30.0 and 40.0°C on the built-in temperature controllers. The calibrated digital thermometer gave readings of 28.8 and 40.8°C, respectively, inside the shakers. The results for the loading tests at each of the temperatures are shown in Table 2; a listing of all the counting results and calculated values is presented in Appendix A.

Table 2. Cesium loading results for CST in supernate simulants.

Bottle I.D.	Temp. (°C)	CST weight (g)	Cesium concentration (mg/L) Sample 1	Sample 2	Average	Average K_d (L/Kg)
30-U	30	0	50.9	51.7	51.3	
30-1	30	0.1192	23.1	21.9	22.5	1024
30-2	30	0.1150	25.0	25.9	25.5	837
40-U	40	0	50.7	50.1	50.4	
40-1	40	0.1060	27.3	26.1	26.7	825
40-2	40	0.1060	27.3	25.1	26.2	859
50-U	50	0	52.0	50.9	51.5	
50-1	50	0.1043	27.1	26.9	27.0	815
50-2	50	0.1585	31.0	31.0	31.0	386
80-U	80	0	51.6	51.1	51.3	
80-1	80	0.1395	44.2	43.6	43.9	100
80-2	80	0.1129	41.4	40.2	40.8	200
120-U	120	0	49.2	49.7	49.5	
120-1	120	0.1025	46.0	47.6	46.8	68
120-2	120	0.0985	45.4	46.0	45.7	96

Figure 6 shows a graph of the average results at each temperature for the cesium concentration in the supernate, loading of cesium on the CST, and K_d. For the next set of loading tests, the built-in temperature controllers were set at 50.0 and 80.0°C, and the calibrated digital thermometer gave readings of 50.2 and 80.4°C, respectively. The final loading test was performed using the oven, and the temperature varied between 116 and 125°C during the 4 days of the test. Significant evaporation loss occurred from the solution in the bottles. The amount of loss varied with the position of the bottles (increasing from left to right), which indicates that the temperature within the oven was not consistent. The thermometer probe was located in the center of the oven, so the temperature profile within the oven was not measured. After the first day of the test, a piece of aluminum foil was placed on the right side of the oven. This helped to reduce the evaporation rate of the control bottle, which was located on the right side of the oven.

On the first day of the test, when the temperature was 120 to 125°C, the no. 1 bottle (left side) lost 9 mL of solution, the no. 2 bottle (center) lost 42 mL, and the control bottle lost 53 mL. On the fourth day of the test, when the temperature ranged from 116 to 121°C, bottle no. 1 lost 7 mL, bottle no. 2 lost 5 mL, and the control bottle lost 3 mL. The evaporation loss was replaced with deionized water each day. A small amount of brown coating was observed on the stainless steel stirring rods and baskets after the first day; and the amount increased over time, indicating oxidative corrosion of the stainless steel by the hot supernate simulant. Brown deposits of iron hydroxide were visible on the bottom of each bottle by the fourth day of the test. The ^{137}Cs

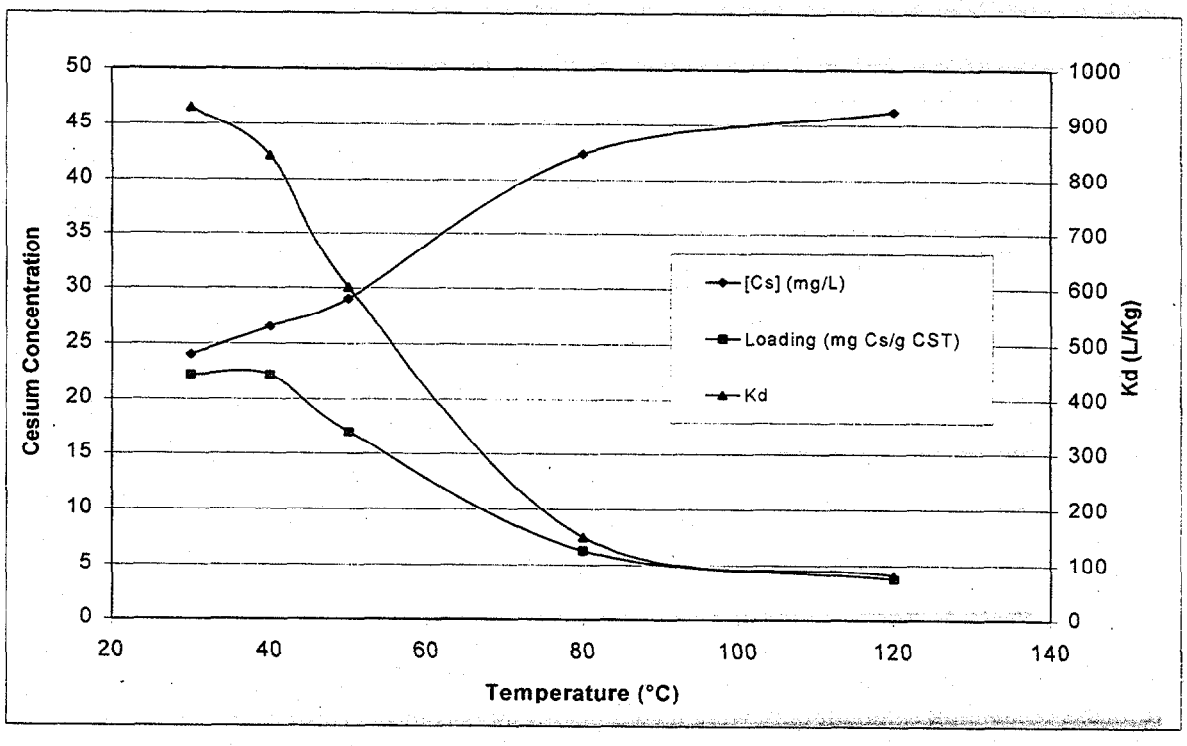


Fig. 6. Effect of temperature on cesium sorption by CST.

concentration measured for the solution in the control bottle was slightly less than the concentration in the starting solution samples, so the iron hydroxide deposits may have removed a small amount of cesium from the solution.

In general, excellent agreement was found between the two samples taken from each container, which indicates that the filtering, pipetting, and gamma counting were very reproducible. However, the agreement between the results for the two containers at each temperature was not as good, particularly at storage temperatures of 50 and 80°C, where the flasks that contained the most CST at each temperature had the higher cesium concentrations. The air-dried CST was difficult to handle because of static charges that caused the particles to jump around on the plastic containers; thus it is possible that some of the CST that was weighed did not get transferred into the Teflon flasks. It is also possible that there were variations within the small samples of CST (0.1 g) that were used for these tests.

A sample of the supernate simulant was analyzed at RMAL by ICP for dissolved metals and by ICP-mass spectrometry for cesium. The concentrations were: Na = >105,300 mg/L, Al = 8430 mg/L, K = 734 mg/L, Si = 94 mg/L, Mo = 19.8 mg/L and Cs = 50.2 mg/L. A sample of the starting solution and a sample from one of the bottles at each temperature were analyzed for ^{137}Cs at RMAL by gamma counting. The counting results, plus the calculated cesium concentrations and K_d results are shown in Table 3. The results from these analyses are similar to those from the in-house gamma counting, except in the case of the sample from bottle 50-1 (which is inconsistent with both the in-house and the other RMAL results).

Table 3. Results of gamma counting at RMAL.

Bottle I.D.	Temp. (°C)	CST (g)	^{137}Cs (Bq/mL)	Cs (mg/L)	K_d (L/kg)
Starting		0	52	50.2	
30-1	30	0.1192	24	23.1	979
40-1	40	0.1060	29	27.9	748
50-1	50	0.1043	27	26.0	888
80-1	80	0.1395	41	42.3	130
120-1	120	0.1025	47	45.2	104

3.2 LEACHING TESTS

Four Teflon flasks that each contained about 0.1 g of CST plus 100 mL of supernate simulant and two flasks containing simulant only were mixed on the shakers at 22°C for 4 days to load the CST with cesium. Also, three Teflon bottles with 100 mL of simulant were positioned in the oven (turned off), and the stirring baskets (two with CST and one without) were placed in the solutions and rotated at about 120 rpm. After 4 days of loading at room temperature, one sample from each of the containers was filtered and counted. The results of the room-temperature loading tests are shown in Table 4. All of the containers with the CST showed very consistent results except for flask L-80-1, which had a higher cesium concentration than the other solutions.

After the room-temperature loading had been completed, the shakers and oven were turned on to heat the samples to the desired leaching temperatures. Two samples from each container were filtered, counted, and then returned to the container after various storage times. The results (average of two samples from each container) are shown in Table 5; a table presenting all of the counting data and calculated values is shown in Appendix B. Figure 7 shows a plot of the average cesium concentrations from both containers at each temperature, and also shows the results from the 4-day loading tests at each temperature for comparison. The loading results are displayed as short line segments so that they will be more visible; however, the data are actually single points at 4 days. The temperature inside the oven typically ranged from 115 to 121°C during the 60 days of the test, with short excursions as low as 112°C and as high as 127°C. The oven thermostat was adjusted several times during the test to keep the temperature within the desired range. The stainless steel stirring rods and baskets continued to corrode, and a thin layer of iron hydroxide was present in the bottom of each bottle at the end of the test. Deionized water was added to each of the bottles at least once a week, as well as just prior to taking each sample, in order to replace evaporation losses. The filtered samples were a light yellow color, probably from soluble chromium compounds that were leached from the stainless steel rods and baskets. The cesium concentration in the control bottle slowly decreased over time, probably as the result of sorption by the iron hydroxide precipitates in the bottles.

Table 4. Results of room-temperature loading tests

Bottle I.D.	CST weight (g)	Temp. (°C)	[Cs] (mg/L)	K_d (L/kg)
L-50-U	0	22	48.7	
L-50-1	0.1043	22	22.3	1196
L-50-2	0.1166	22	20.0	1287
L-80-U	0	22	47.7	
L-80-1	0.0981	22	29.9	688
L-80-2	0.1042	22	23.3	1096
L-120-U	0	22	48.1	
L-120-1	0.0955	22	23.4	1187
L-120-2	0.0977	22	24.3	1080

Table 5. Results of leaching tests

Bottle I.D.	CST weight (g)	Cesium concentration (mg/L), after storage times of					
		1 day	2 days	4 days	16 days	29 days	60 days
L-50-U	0	48.4	49.0	48.1	49.6	50.6	48.6
L-50-1	0.1043	29.3	29.7	29.1	31.8	31.7	30.8
L-50-2	0.1166	27.1	27.8	28.1	29.7	30.5	31.2
L-80-U	0	48.5	49.5	49.3	50.8	52.2	47.9
L-80-1	0.0981	43.6	42.8	43.4	45.4	45.9	41.8
L-80-2	0.1042	40.6	40.3	40.7	42.4	43.3	37.7
L-120-U	0	48.1	47.3	48.1	46.9	43.2	43.7
L-120-1	0.0955	44.2	42.8	43.2	45.2	46.3	44.6
L-120-2	0.0977	44.7	42.7	43.6	46.7	44.2	43.0

The two shakers maintained very stringent temperature control, ranging from 49.9 to 50.1 °C and 79.9 to 80.1 °C on the built-in thermometers, and 47.9 to 49.2 °C and 78.1 to 78.6 °C on the calibrated digital thermometer. The volume of solution in each flask was measured and adjusted to 100 mL with deionized water before the 60-day samples were withdrawn. The volumes before adjustment are shown in Table 6. The evaporation losses from these flasks would cause a slight increase in the soluble cesium concentrations, which could explain the increases seen for the

samples that were taken after 16 and 29 days. The volumes of the solutions were not measured at 16 and 29 days; thus the exact volume losses, and corresponding corrections for the cesium concentration are not known.

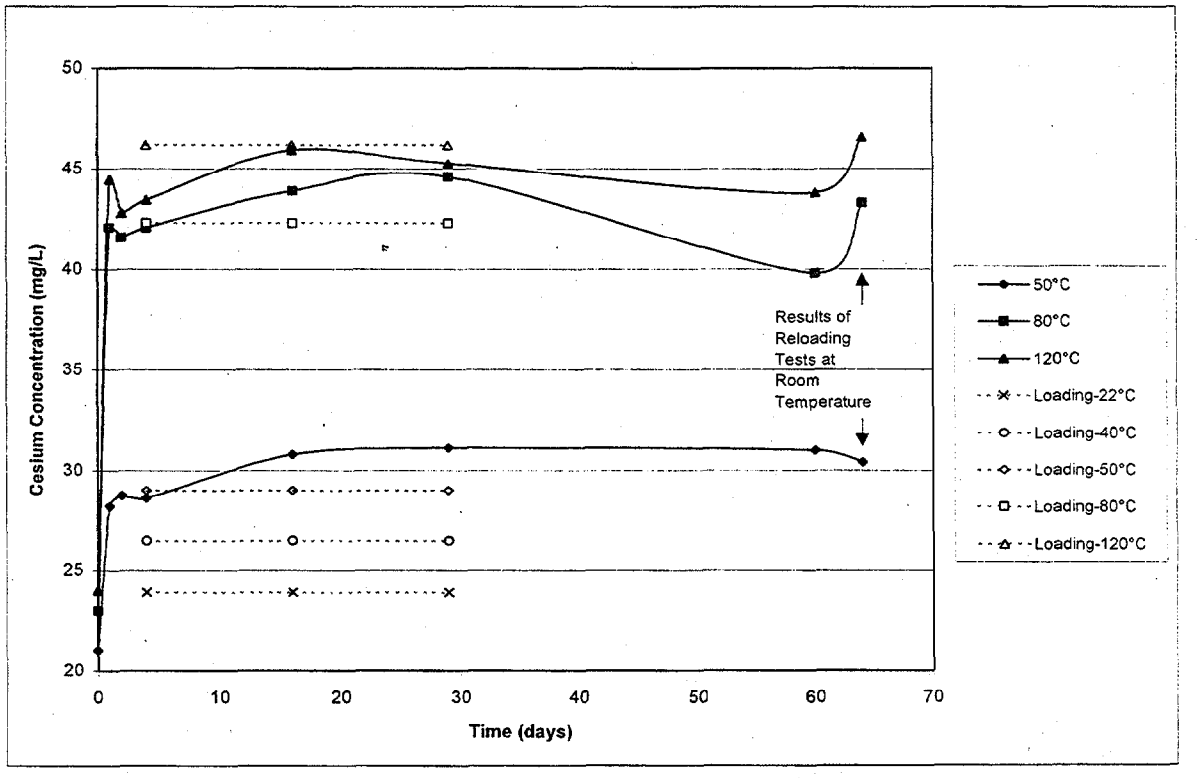


Figure 7. Results of leaching tests. Loading results are shown for comparison.

Table 6. Volume of supernate simulant in each flask after 60 days of leaching

Container I.D.	L-50-U	L-50-1	L-50-2	L-80-U	L-80-1	L-80-2
Volume (mL)	99	95	98	97	98	94

After the leaching tests had been completed, the heaters in the oven and shakers were turned off and the samples were mixed at room temperature for 4 days. Two samples from each container were then filtered, pipetted, and counted as before. Analyses of these samples showed that the cesium in the simulant solutions did not reload onto the CST (see Table 7 and Fig. 7). The cesium concentrations were actually a little higher than those for the 60-day leaching samples; however, the control sample concentrations were also higher, so the increase is probably analytical variation.

Table 7. Results of room temperature reloading tests, following the leaching tests

Bottle I.D.	CST weight (g)	[Cs] (mg/L)	Cs loading (mg Cs/g CST)	K_d (L/kg)
L-50-U	0	49.7		
L-50-1	0.1043	30.5	18.7	612
L-50-2	0.1166	30.2	17.0	562
L-80-U	0	49.7		
L-80-1	0.0981	44.4	5.8	129
L-80-2	0.1042	42.2	7.4	177
L-120-U	0	47.6		
L-120-1	0.0955	46.5	3.6	78
L-120-2	0.0977	46.6	3.4	74

3.3 NONRADIOACTIVE LEACHING TESTS

Samples of CST that were stored in SRS average supernate simulant at temperatures of 24, 50, 80, and 120°C for up to 35 days were analyzed by X-ray diffraction to determine if any changes were detectable in the crystal structure of the CST. The analyses were performed by the Lockheed Martin Energy Systems' Analytical Services Organization. The diffraction patterns did not change for any of these samples except the one stored for 35 days at 120°C, which showed a very slight decrease in the peak at a diffraction angle of 28.5°. The diffraction patterns (see Appendix C) matched the library pattern for cesium sodium titanium oxide silicate hydrate

Samples of the supernate simulant were collected from each container, at the same time the CST samples were removed, and analyzed for dissolved metals by ICP. Several metals showed increased concentrations as compared with the original supernate simulant. Table 8 shows the concentrations of the metals that changed significantly from what was present in the original supernate simulant. One of the metals analyzed involves proprietary information from UOP, therefore it is identified only as "Trade Secret Material 2" (TS #2). The concentrations in the starting supernate simulant were as follows: Al = 7855, Pb = <0.06 mg/L, Si = 42 mg/L, Ti = <0.03 mg/L, and TS #2 = 0.6 mg/L. (Note - TS #2 was not purposely added to the supernate simulant, so the concentration measured in the starting solution may be a false positive.) Figures 8 and 9 show graphs of the silicon and titanium concentrations, respectively. Most of the metals that increased in concentration are known to be present in the CST, but the source of the lead is not known. The concentration of several of the metals increased initially and then decreased, which indicates that competing dissolution and precipitation reactions are probably occurring.

The simulant solution contains high concentrations of aluminum, thus most of the silicon that leaches into solution is likely to precipitate as sodium aluminosilicate. The decrease in aluminum can be used as an indication of the amount of silicon that leached from the CST.

Table 8. Soluble metals in SRS supernate simulant in contact with CST

Metal	Temp. (°C)	Soluble metal concentration (mg/L) after storage time of 1 day	7 days	21 days	35 days
Al	24	7819	7585	7656	8063
Al	50	7463	6647	6545	6800
Al	80	6629	6115	6210	6709
Al	120	5836	5836	5097	4803
Pb	24	1.13	1.94	2.64	2.62
Pb	50	1.62	1.16	0.63	0.47
Pb	80	0.94	0.25	0.52	0.65
Pb	120	0.44	2.26	1.93	0.36
Si	24	75	137	198	178
Si	50	121	58	20	16
Si	80	64	14	13	16
Si	120	16	12	11	9
Ti	24	4.64	6.14	7.61	7.53
Ti	50	4.72	3.12	2.01	1.34
Ti	80	2.77	2.91	4.41	5.13
Ti	120	5.61	18.7	14.02	5.68
TS #2	24	5.10	12.0	17.5	21.2
TS #2	50	10.0	14.2	13.7	11.4
TS #2	80	15.0	13.1	12.0	12.0
TS #2	120	23.4	18.7	18.9	14.9

Based on the aluminum losses for the 21-day samples, and assuming an Al:Si ratio of 1 in the precipitated material, the amounts of silicon that could have been leached from the CST and then precipitated are 0.04, 0.32, 0.40, and 0.68 g for the 24, 50, 80, and 120°C samples, respectively. (Note: These numbers were corrected for the 0.01 g of silicon originally present in the 240 mL of supernate simulant.) For the original CST weight of 16 g, the calculated silicon losses amounted to 0.25, 2.0, 2.5, and 4.2 wt % loss of the CST for the 24, 50, 80, and 120°C samples, respectively. The expected error band for the measured Al concentrations of ±5 % results in a range of 3.7 to 4.7 wt % for the maximum calculated CST weight loss of 4.2 wt %. We did not attempt to isolate or identify any aluminosilicate compounds that may have formed, and they did not show up on the x-ray diffraction patterns. It is possible that part of the aluminum precipitated in some other form, so these results represent a maximum leaching of silicon from the CST.

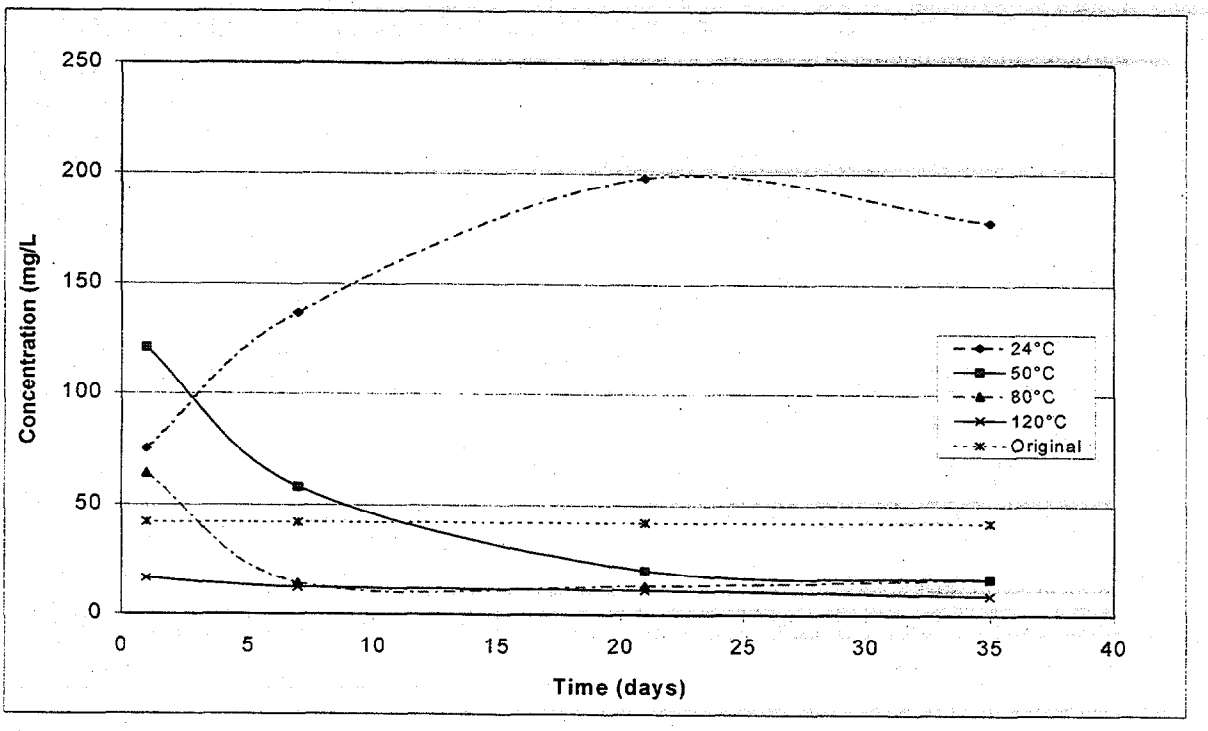


Fig. 8. Leaching of silicon into SRS supernate simulant at various temperatures.

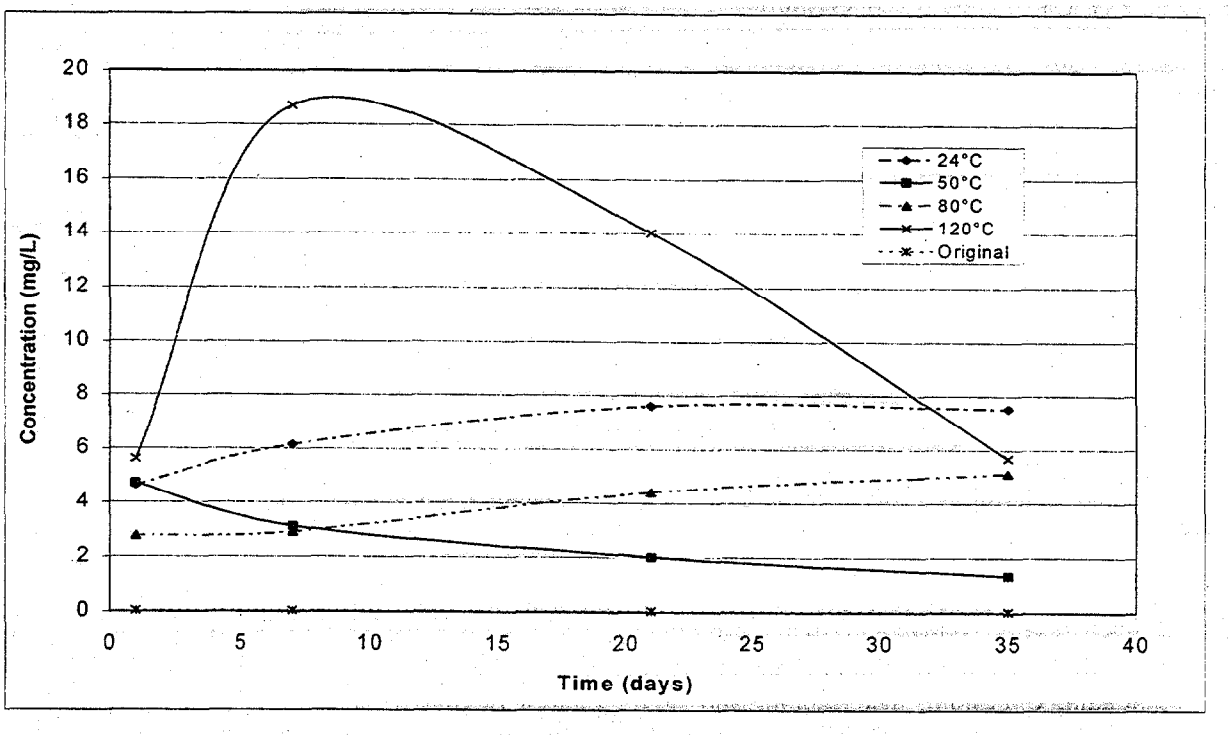


Fig. 9. Leaching of titanium into SRS supernate simulant at various temperatures.

UOP has reported that excess silicon is present in the CST, which is physically trapped in the particles but not chemically reacted to form the silicotitanate.[11] UOP reported that the excess silicon concentration for the lot 999098810005 CST used in these tests was 4.1 wt % as SiO_2 (1.9 wt% Si), which is about half of the maximum calculated silicon loss from the CST. The hot supernate simulant is probably leaching the excess silicon from the CST, but the remaining aluminum loss from the supernate could be caused by either silicon leached from the CST structure or by precipitation of other aluminum containing compounds.

4. DISCUSSION

Zheng, Gu and Anthony have developed an equilibrium model[12] for predicting the distribution of cesium between CST and various salt solutions at temperatures up to 44°C. The model was developed for CST powder, so the binder in the granular CST (IONSIV IE-911) that was used in these tests may affect the results. Table 9 shows a comparison between the average results obtained for the batch tests at each temperature and the results predicted by the model. Most of the tests were performed at temperatures above those where the model has been validated, so it is not surprising that model predictions increasingly diverge from the measured values as the temperature increases.

Table 9. Comparison between batch test results and model predictions

Temp.	Measured Values		Model Predictions	
(°C)	[Cs] (mg/L)	K_d (L/kg)	[Cs] (mg/L)	K_d (L/kg)
30	24.0	930	19.0	1391
40	26.5	842	23.0	1106
50	29.0	601	22.5	941
80	42.3	150	30.0	531
120	46.2	82	39.2	274

Results of the leaching tests show that previously loaded cesium is removed from the CST in the high-salt, high-pH SRS supernate simulant as the temperature increases. The leaching is very rapid, occurring at about the same speed as the original cesium loading, most of which occurs within the first 24 h. The final distribution coefficient for cesium on the CST is also about the same, regardless of whether the CST is initially loaded at a given temperature, or it is loaded at a lower temperature and then temperature raised to the higher level.

After being stored at temperatures of 50 to 120°C in the SRS supernate simulant for 60 days, the CST does not reload any cesium after the temperature has been reduced. Further work would be needed to determine the full range of storage time and temperature profiles that would impact the loading of cesium. The leaching and reprecipitation of silicon and other metals from the CST might physically block the pores of the CST, preventing cesium from reloading onto the CST. The X-ray diffraction patterns did not show any significant change in the crystal structure of the

CST, so it is unlikely that chemical changes in the bulk CST prevented the cesium from reloading. Based on the amount of aluminum that was lost from the supernate simulant in the nonradioactive leaching tests, silicon losses from the CST could be as high as 4.2 wt %, if all of the silicon leached from the CST was precipitated as sodium aluminosilicate.

5. REFERENCES

1. J. F. Walker, Jr., et al., *Cesium Removal Demonstration Utilizing Crystalline Silicotitanate Sorbent for Processing Melton Valley Storage Tank Supernate: Final Report*, ORNL/TM-13503, Oak Ridge National Laboratory, Oak Ridge, TN, March 1998.

2. P. A Taylor, J. D. Hewitt, T. D. Hylton, and T. E. Kent, *Transport and Storage Properties of CST Slurries for the Savannah River Site*, ORNL/TM-13727, Oak Ridge National Laboratory, Oak Ridge, TN, April 1999.

3. J. E Miller and N. E. Brown, *Development and Properties of Crystalline Silicotitanate (CST) Ion Exchangers for Radioactive Waste Applications*, SAND97-0771, Sandia National Laboratories, April 1997.

4. R. G. Anthony, R. G. Dosch, D. Gu, and C. V. Philip, "Use of Silicotitanates for Removing Cesium and Strontium from Defense Waste", *Ind. Eng. Chem. Res.* **33**, 2702–5, (1994).

5. T. A. Todd, K. N. Brewer, D. J. Wood, P. A. Tullock, L. G. Olson, and A. L. Olson, *Evaluation and Testing of Inorganic Ion Exchange Sorbents for the Removal of Cesium-137 From Idaho Chemical Processing Plant Acidic Tank Waste*, Idaho National Engineering and Environmental Laboratory, (in press).

6. D. T. Bostick, S. M. DePaoli, and B. Guo, *Evaluation of Improved Techniques for the Removal of Fission Products from Process Wastewater and Groundwater: FY 1997 Status*, ORNL/TM-13497, Oak Ridge National Laboratory, Oak Ridge, TN, February 1998.

7. R. A. Jacobs, *Technical Task Request HLW-SDT-TTR-99-08.3*, Savannah River Site, Aiken, SC, March 4, 1999.

8. P. A. Taylor, *Technical Task Plan For Determining the Chemical/Thermal Stability of Crystalline Silicotitanate*, EDS-WP-SRS-1, Oak Ridge National Laboratory, Oak Ridge, TN, March 9, 1999.

9. D. T. Bostick and W. V. Steele, *Thermal and Physical Property Determination for Ionsiv® IE-911 Crystalline Silicotitanate and Savannah River Site Waste Simulant Solutions*, ORNL-TM 1999-133, Oak Ridge National Laboratory, Oak Ridge, TN, September 1999.

10. D. D. Walker, *Preparation of Simulated Waste Solutions*, WSRC-TR-99-00116, Rev. 0, Westinghouse Savannah River Company, Aiken, SC, April 21, 1999.

11. J. T. Carter (Savannah River Site) and John Sherman (UOP), Minutes of telephone conversation held on September 14, 1999, HLW-SDT-99-0292, September 15, 1999.

12. Z. Zheng, D. Gu, and R. G. Anthony, "Estimation of Cesium Ion Exchange Distribution Coefficients for Concentrated Electrolyte Solutions When Using Crystalline Silicotitanates," *Ind. Eng. Chem. Res.* **34**, 2142–47 (1995).

6. APPENDICES

Appendix A

ANALYTICAL RESULTS AND CALCULATIONS FOR LOADING TESTS

Results of Loading Tests Using CST Sorbent in SRS Average Supernate Simulant Solution (5.6 M Na+)

Date 4/19/99

Background Count Rate	14	4	8	Average	9	
Starting Solution Count Rate	7948	7595	7710	Average Net	7742	

Sample ID	Storage temperature (°C)	CST weight (g)	Count rate (counts/1000 s/2 mL) Gross	Net	Cs conc. (mg/L)	Cs loading on CST (mg/g)	(meq/g)	Kd (L/kg)	Averages Kd (L/kg)	Cs Conc (mg/L)	Container average Cs Conc (mg/L)	Container average Kd (L/kg)
SRS-30-Ua	30	0	7891	7882	50.9						51.3	
SRS-30-Ub	30	0	8018	8009	51.7							
SRS-30-1a	30	0.1192	3590	3581	23.1	22.5	0.170	975	930	24.0	22.5	1024
SRS-30-1b	30	0.1192	3407	3398	21.9	23.5	0.177	1072				
SRS-30-2a	30	0.1150	3887	3878	25.0	21.7	0.163	866			25.5	837
SRS-30-2b	30	0.1150	4025	4016	25.9	20.9	0.157	807				
SRS-40-Ua	40	0	7861	7852	50.7						50.4	
SRS-40-Ub	40	0	7769	7760	50.1							
SRS-40-1a	40	0.1060	4235	4226	27.3	21.4	0.161	785	842	26.5	26.7	825
SRS-40-1b	40	0.1060	4048	4039	26.1	22.6	0.170	865				
SRS-40-2a	40	0.1060	4237	4228	27.3	21.4	0.161	784			26.2	859
SRS-40-2b	40	0.1060	3898	3889	25.1	23.5	0.177	935				

Results of Loading Tests Using CST Sorbent in SRS Average Supernate Simulant Solution

Date 4/23/99

Background Count Rate	9	4	8	Average	7	
Starting Solution Count Rate	7948	7595	7710	Average Net	7744	

Sample ID	Storage temperature (°C)	CST weight (g)	Count rate (counts/1000 s/2 mL) Gross	Net	Cs conc. (mg/L)	Cs loading on CST (mg/g)	(meq/g)	Kd (L/kg)	Averages Kd (L/kg)	Cs Conc (mg/L)	Container average Cs Conc (mg/L)	Container average Kd (L/kg)
SRS-50-Ua	50	0	8068	8061	52.0						51.5	
SRS-50-Ub	50	0	7888	7881	50.9							
SRS-50-1a	50	0.1043	4204	4197	27.1	22.0	0.165	810	601	29.0	27.0	815
SRS-50-1b	50	0.1043	4181	4174	26.9	22.1	0.166	820				
SRS-50-2a	50	0.1585	4816	4809	31.0	12.0	0.090	385			31.0	386
SRS-50-2b	50	0.1585	4805	4798	31.0	12.0	0.090	387				
SRS-80-Ua	80	0	7992	7985	51.6						51.3	
SRS-80-Ub	80	0	7917	7910	51.1							
SRS-80-1a	80	0.1395	6845	6838	44.2	4.2	0.032	95	150	42.3	43.9	100
SRS-80-1b	80	0.1395	6759	6752	43.6	4.6	0.035	105				
SRS-80-2a	80	0.1129	6417	6410	41.4	7.6	0.057	184			40.8	200
SRS-80-2b	80	0.1129	6230	6223	40.2	8.7	0.065	216				

RMAL Results for Loading Tests Using CST Sorbent in SRS Simulant

Date 4/19, 4/23 & 5/14/99

Sample ID	Storage Temperature (°C)	CST weight (g)	Cs-137 (Bq/mL)	Cs Conc. (mg/L)	Cs Loading on CS (mg/g)	(meq/g)	Kd (L/kg)
SRS-Sim			52	50.0			
SRS-30-1	30	0.1192	24	23.1	22.6	0.170	979
SRS-40-1	40	0.1060	29	27.9	20.9	0.157	748
SRS-50-1	50	0.1043	27	26.0	23.0	0.173	888
SRS-80-1	80	0.1395	44	42.3	5.5	0.041	130
SRS-120-A	120	0.1025	47	45.2	4.7	0.035	104

Results of Loading Tests Using CST Sorbent in SRS Average Supernate Simulant Solution

Date 5/14/99

Background Count Rate	8	15	14	Average	12	
Starting Solution Count Rate	7855	7615		Average Net	7723	

Sample ID	Storage temperature (°C)	CST weight (g)	Count rate (counts/1000 s/2 mL) Gross	Net	Cs conc. (mg/L)	Cs loading on CST (mg/g)	(meq/g)	Kd (L/kg)	Averages Kd (L/kg)	Cs Conc (mg/L)	Container average Cs Conc (mg/L)	Container average Kd (L/kg)
SRS-120-U1	120	0	7607	7600	49.2						49.5	
SRS-120-U2	120	0	7683	7676	49.7							
SRS-120-A1	120	0.1025	7105	7098	46.0	3.9	0.030	86	82	46.2	46.8	68
SRS-120-A2	120	0.1025	7356	7349	47.6	2.4	0.018	50				
SRS-120-B1	120	0.0985	7020	7013	45.4	4.7	0.035	103			45.7	96
SRS-120-B2	120	0.0985	7111	7104	46.0	4.1	0.031	88				

Appendix B

ANALYTICAL RESULTS AND CALCULATIONS FOR LEACHING TESTS

Results of Initial Loading Tests Using CST Sorbent in SRS Average Supernate Simulant Solution (5.6 *M* Na+)

Date 6/21/99

Background Count Rate			2	18	8	Average	9					
Starting Solution Count Rate			10504	10507	10462	Average Net	10482					
Sample ID	Storage temperature (°C)	CST weight (g)	Count rate (counts/1000 s/2 mL) Gross	Net	Cs conc. (mg/L)	Cs loading on CST (mg/g)	(meq/g)	Kd (L/kg)	Averages Kd (L/kg)	Cs Conc (mg/L)		
SRS-L-50-U	22	0	10215	10206	48.7							
SRS-L-50-1	22	0.1043	4674	4665	22.3	26.6	0.200	1196	1242	21.1		
SRS-L-50-2	22	0.1166	4200	4191	20.0	25.7	0.194	1287				
SRS-L-80-U	22	0	10011	10002	47.7							
SRS-L-80-1	22	0.0981	6268	6259	29.9	20.5	0.155	688	892	26.6		
SRS-L-80-2	22	0.1042	4903	4894	23.3	25.6	0.192	1096				
SRS-L-120-U	22	0	10095	10086	48.1							
SRS-L-120-1	22	0.0955	4921	4912	23.4	27.8	0.209	1187	1134	23.9		
SRS-L-120-2	22	0.0977	5109	5100	24.3	26.3	0.198	1080				
							Overall Averages		1089	23.9		

Results of Leaching Tests

Results of Leaching Tests After One Day at Temperature

Date 6/22/99

Background Count Rate			23	22	11	Average	19					
Starting Solution Count Rate			10504	10507	10462	Average Net	10472					
Sample ID	Storage temperature (°C)	CST weight (g)	Count rate (counts/1000 s/2 mL) Gross	Net	Cs conc. (mg/L)	Cs loading on CST (mg/g)	(meq/g)	Kd (L/kg)	Averages Kd (L/kg)	Cs Conc (mg/L)	Container average Cs Conc (mg/L)	Container average Kd (L/kg)
SRS-L-50-Ua	50	0	10060	10041	47.9						48.4	
SRS-L-50-Ub	50	0	10242	10223	48.8							
SRS-L-50-1a	50	0.1043	6210	6191	29.6	19.6	0.147	663	701	28.2	29.3	677
SRS-L-50-1b	50	0.1043	6102	6083	29.0	20.1	0.151	692				
SRS-L-50-2a	50	0.1166	5761	5742	27.4	19.4	0.146	706			27.1	724
SRS-L-50-2b	50	0.1166	5634	5615	26.8	19.9	0.150	742				
SRS-L-80-Ua	80	0	10143	10124	48.3						48.5	
SRS-L-80-Ub	80	0	10193	10174	48.6							
SRS-L-80-1a	80	0.0981	9198	9179	43.8	6.3	0.047	144	187	42.1	43.6	151
SRS-L-80-1b	80	0.0981	9086	9067	43.3	6.8	0.051	158				
SRS-L-80-2a	80	0.1042	8580	8561	40.9	8.8	0.066	214			40.6	223
SRS-L-80-2b	80	0.1042	8460	8441	40.3	9.3	0.070	231				
RS-L-120-Ua	120	0	10102	10083	48.1						48.1	
RS-L-120-Ub	120	0	10088	10069	48.1							
RS-L-120-1a	120	0.0955	9373	9354	44.7	5.6	0.042	125	129	44.5	44.2	137
RS-L-120-1b	120	0.0955	9186	9167	43.8	6.5	0.049	149				
RS-L-120-2a	120	0.0977	9280	9261	44.2	5.9	0.045	134			44.7	122
RS-L-120-2b	120	0.0977	9480	9461	45.2	4.9	0.037	109				

Results of Leaching Tests After Two Days at Temperature

Date 6/23/99

Background Count Rate			8	19	17	Average	15					
Starting Solution Count Rate			10504	10507	10462	Average Net	10476					
Sample ID	Storage temperature (°C)	CST weight (g)	Count rate (counts/1000 s/2 mL) Gross	Net	Cs conc. (mg/L)	Cs loading on CST (mg/g)	(meq/g)	Kd (L/kg)	Averages Kd (L/kg)	Cs Conc (mg/L)	Container average Cs Conc (mg/L)	Container average Kd (L/kg)
SRS-L-50-Ua	50	0	10268	10253	48.9						49.0	
SRS-L-50-Ub	50	0	10298	10283	49.1							
SRS-L-50-1a	50	0.1043	6305	6290	30.0	19.2	0.144	638	671	28.7	29.7	658
SRS-L-50-1b	50	0.1043	6150	6135	29.3	19.9	0.149	678				
SRS-L-50-2a	50	0.1166	5828	5813	27.7	19.1	0.144	688			27.8	683
SRS-L-50-2b	50	0.1166	5866	5851	27.9	18.9	0.142	678				
SRS-L-80-Ua	80	0	10241	10226	48.8						49.5	
SRS-L-80-Ub	80	0	10548	10533	50.3							
SRS-L-80-1a	80	0.0981	9087	9072	43.3	6.8	0.051	158	201	41.6	42.8	171
SRS-L-80-1b	80	0.0981	8888	8873	42.3	7.8	0.059	184				
SRS-L-80-2a	80	0.1042	8530	8515	40.6	9.0	0.068	221			40.3	230
SRS-L-80-2b	80	0.1042	8398	8383	40.0	9.6	0.072	240				
RS-L-120-Ua	120	0	9914	9899	47.2						47.3	
RS-L-120-Ub	120	0	9928	9913	47.3							
RS-L-120-1a	120	0.0955	9014	8999	43.0	7.4	0.056	172	175	42.8	42.8	175
RS-L-120-1b	120	0.0955	8964	8949	42.7	7.6	0.057	179				
RS-L-120-2a	120	0.0977	9147	9132	43.6	6.6	0.049	151			42.7	174
RS-L-120-2b	120	0.0977	8795	8780	41.9	8.3	0.062	198				

Results of Leaching Tests After Four Days at Temperature

Date 6/25/99

Background Count Rate			24	14	12	Average	17					
Starting Solution Count Rate			10504	10507	10462	Average Net	10474					
Sample ID	Storage temperature (°C)	CST weight (g)	Count rate (counts/1000 s/2 mL) Gross	Net	Cs conc. (mg/L)	Cs loading on CST (mg/g)	(meq/g)	Kd (L/kg)	Averages Kd (L/kg)	Cs Conc (mg/L)	Container average Cs Conc (mg/L)	Container average Kd (L/kg)
SRS-L-50-Ua	50	0	10156	10139	48.4						48.1	
SRS-L-50-Ub	50	0	10048	10031	47.9							
SRS-L-50-1a	50	0.1043	6043	6026	28.8	20.4	0.153	708	678	28.6	29.1	688
SRS-L-50-1b	50	0.1043	6188	6171	29.5	19.7	0.148	669				
SRS-L-50-2a	50	0.1166	5830	5813	27.8	19.1	0.144	688			28.1	667
SRS-L-50-2b	50	0.1166	5987	5970	28.5	18.4	0.139	647				
SRS-L-80-Ua	80	0	10371	10354	49.4						49.3	
SRS-L-80-Ub	80	0	10309	10292	49.1							
SRS-L-80-1a	80	0.0981	9109	9092	43.4	6.7	0.051	155	187	42.1	43.4	154
SRS-L-80-1b	80	0.0981	9121	9104	43.5	6.7	0.050	153				
SRS-L-80-2a	80	0.1042	8474	8457	40.4	9.2	0.070	229			40.7	220
SRS-L-80-2b	80	0.1042	8605	8588	41.0	8.6	0.065	211				
RS-L-120-Ua	120	0	9978	9961	47.6						48.1	
RS-L-120-Ub	120	0	10216	10199	48.7							
SRS-L-120-1a	120	0.0955	9054	9037	43.1	7.2	0.054	166	156	43.4	43.2	164
SRS-L-120-1b	120	0.0955	9098	9081	43.4	7.0	0.052	161				
SRS-L-120-2a	120	0.0977	9082	9065	43.3	6.9	0.052	159			43.6	149
SRS-L-120-2b	120	0.0977	9237	9220	44.0	6.1	0.046	139				

Results of Leaching Tests After Sixteen Days at Temperature

Date 7/7/99

Background Count Rate			13	13	50	Average	25					
Starting Solution Count Rate			10478	10307	10342	Average Net	10350					
Sample ID	Storage temperature (°C)	CST weight (g)	Count rate (counts/1000 s/2 mL) Gross	Net	Cs conc. (mg/L)	Cs loading on CST (mg/g)	(meq/g)	Kd (L/kg)	Averages Kd (L/kg)	Cs Conc (mg/L)	Container average Cs Conc (mg/L)	Container average Kd (L/kg)
SRS-L-50-Ua	50	0	10241	10216	49.3						49.6	
SRS-L-50-Ub	50	0	10356	10331	49.9							
SRS-L-50-1a	50	0.1043	6649	6624	32.0	17.3	0.130	539	566	30.8	31.8	547
SRS-L-50-1b	50	0.1043	6581	6556	31.7	17.6	0.132	555				
SRS-L-50-2a	50	0.1166	6125	6100	29.5	17.6	0.133	598			29.7	584
SRS-L-50-2b	50	0.1166	6242	6217	30.0	17.1	0.129	570				
SRS-L-80-Ua	80	0	10659	10634	51.4						50.8	
SRS-L-80-Ub	80	0	10421	10396	50.2							
SRS-L-80-1a	80	0.0981	9486	9461	45.7	4.4	0.033	96	138	43.9	45.4	103
SRS-L-80-1b	80	0.0981	9364	9339	45.1	5.0	0.037	110				
SRS-L-80-2a	80	0.1042	8742	8717	42.1	7.6	0.057	180			42.4	173
SRS-L-80-2b	80	0.1042	8855	8830	42.7	7.0	0.053	165				
RS-L-120-Ua	120	0	9796	9771	47.2						46.9	
RS-L-120-Ub	120	0	9652	9627	46.5							
SRS-L-120-1a	120	0.0955	9484	9459	45.7	4.5	0.034	99	92	45.9	45.2	111
SRS-L-120-1b	120	0.0955	9287	9262	44.7	5.5	0.041	123				
SRS-L-120-2a	120	0.0977	9767	9742	47.1	3.0	0.023	64			46.7	73
SRS-L-120-2b	120	0.0977	9600	9575	46.3	3.8	0.029	83				

Results of Leaching Tests After Twenty Nine Days at Temperature

Date 7/20/1999

Background Count Rate			46	48	32	Average	42					
Starting Solution Count Rate			10410	10459	10233	Average Net	10325					
Sample ID	Storage temperature (°C)	CST weight (g)	Count rate (counts/1000 s/2 mL) Gross	Net	Cs conc. (mg/L)	Cs loading on CST (mg/g)	(meq/g)	Kd (L/kg)	Averages Kd (L/kg)	Cs Conc (mg/L)	Container average Cs Conc (mg/L)	Container average Kd (L/kg)
SRS-L-50-Ua	50	0	10483	10441	50.6						50.6	
SRS-L-50-Ub	50	0	10483	10441	50.6							
SRS-L-50-1a	50	0.1043	6609	6567	31.8	17.4	0.131	549	550	31.1	31.7	553
SRS-L-50-1b	50	0.1043	6571	6529	31.6	17.6	0.133	557				
SRS-L-50-2a	50	0.1166	6429	6387	30.9	16.4	0.123	529			30.5	547
SRS-L-50-2b	50	0.1166	6266	6224	30.1	17.0	0.128	565				
SRS-L-80-Ua	80	0	10669	10627	51.5						52.2	
SRS-L-80-Ub	80	0	10994	10952	53.0							
SRS-L-80-1a	80	0.0981	9543	9501	46.0	4.1	0.031	88	120	44.6	45.9	92
SRS-L-80-1b	80	0.0981	9485	9443	45.7	4.4	0.033	95				
SRS-L-80-2a	80	0.1042	9019	8977	43.5	6.3	0.047	144			43.3	148
SRS-L-80-2b	80	0.1042	8961	8919	43.2	6.5	0.049	151				
RS-L-120-Ua	120	0	9074	9032	43.7						43.2	
RS-L-120-Ub	120	0	8653	8811	42.7							
SRS-L-120-1a	120	0.0955	9574	9532	46.2	4.0	0.030	87	108	45.3	46.3	83
SRS-L-120-1b	120	0.0955	9640	9598	46.5	3.7	0.028	79				
SRS-L-120-2a	120	0.0977	9215	9173	44.4	5.7	0.043	129			44.2	133
SRS-L-120-2b	120	0.0977	9138	9096	44.0	6.1	0.046	138				

Results of Leaching Tests After Sixty Days at Temperature

Date 8/20/1999

Background Count Rate	14	15	10	Average	13
Starting Solution Count Rate	10606	10429	10149	Average Net	10382

Sample ID	Storage temperature (°C)	CST weight (g)	Count rate (counts/1000 s/2 mL) Gross	Net	Cs conc. (mg/L)	Cs loading on CST (mg/g)	(meq/g)	Kd (L/kg)	Averages Kd (L/kg)	Cs Conc (mg/L)	Container average Cs Conc (mg/L)	Container average Kd (L/kg)
SRS-L-50-Ua	50	0	10059	10046	48.4						48.6	
SRS-L-50-Ub	50	0	10134	10121	48.7							
SRS-L-50-1a	50	0.1043	6501	6488	31.2	18.0	0.135	575	559	31.0	30.8	600
SRS-L-50-1b	50	0.1043	6296	6283	30.3	18.9	0.142	625				
SRS-L-50-2a	50	0.1166	6543	6530	31.4	15.9	0.120	506			31.2	517
SRS-L-50-2b	50	0.1166	6435	6422	30.9	16.4	0.123	529				
SRS-L-80-Ua	80	0	9819	9819	47.3						47.9	
SRS-L-80-Ub	80	0	10077	10077	48.5							
SRS-L-80-1a	80	0.0981	9321	9321	44.9	5.2	0.039	116	261	39.8	41.8	207
SRS-L-80-1b	80	0.0981	8034	8034	38.7	11.5	0.087	298				
SRS-L-80-2a	80	0.1042	8168	8168	39.3	10.2	0.077	260			37.7	315
SRS-L-80-2b	80	0.1042	7491	7491	36.1	13.4	0.101	370				
RS-L-120-Ua	120	0	9130	9130	44.0						43.7	
RS-L-120-Ub	120	0	9015	9015	43.4							
RS-L-120-1a	120	0.0955	9196	9196	44.3	6.0	0.045	135	147	43.8	44.6	126
RS-L-120-1b	120	0.0955	9343	9343	45.0	5.2	0.039	116				
RS-L-120-2a	120	0.0977	8984	8984	43.3	6.9	0.052	159			43.0	168
RS-L-120-2b	120	0.0977	8855	8855	42.6	7.5	0.057	176				

RMAL Results For Leaching Test Samples

Sample ID	Storage temperature (°C)	Storage time (days)	CST Weight (g)	Cs-137 (Bq/mL)	Cs Conc. (mg/L)	Cs Loading on CS (mg/g)	(m eq/g)	Kd (L/kg)
SRS-L-U		0	0	67	50			
SRS-L-50-1	50	29	0.1043	43	32.1	17.2	0.129	343
SRS-L-80-2	80	16	0.1042	55	41.0	8.6	0.065	268
SRS-L-120-1	120	60	0.0955	61	45.5	4.7	0.035	114

Results of Reloading Tests Following 60-day Leaching Tests

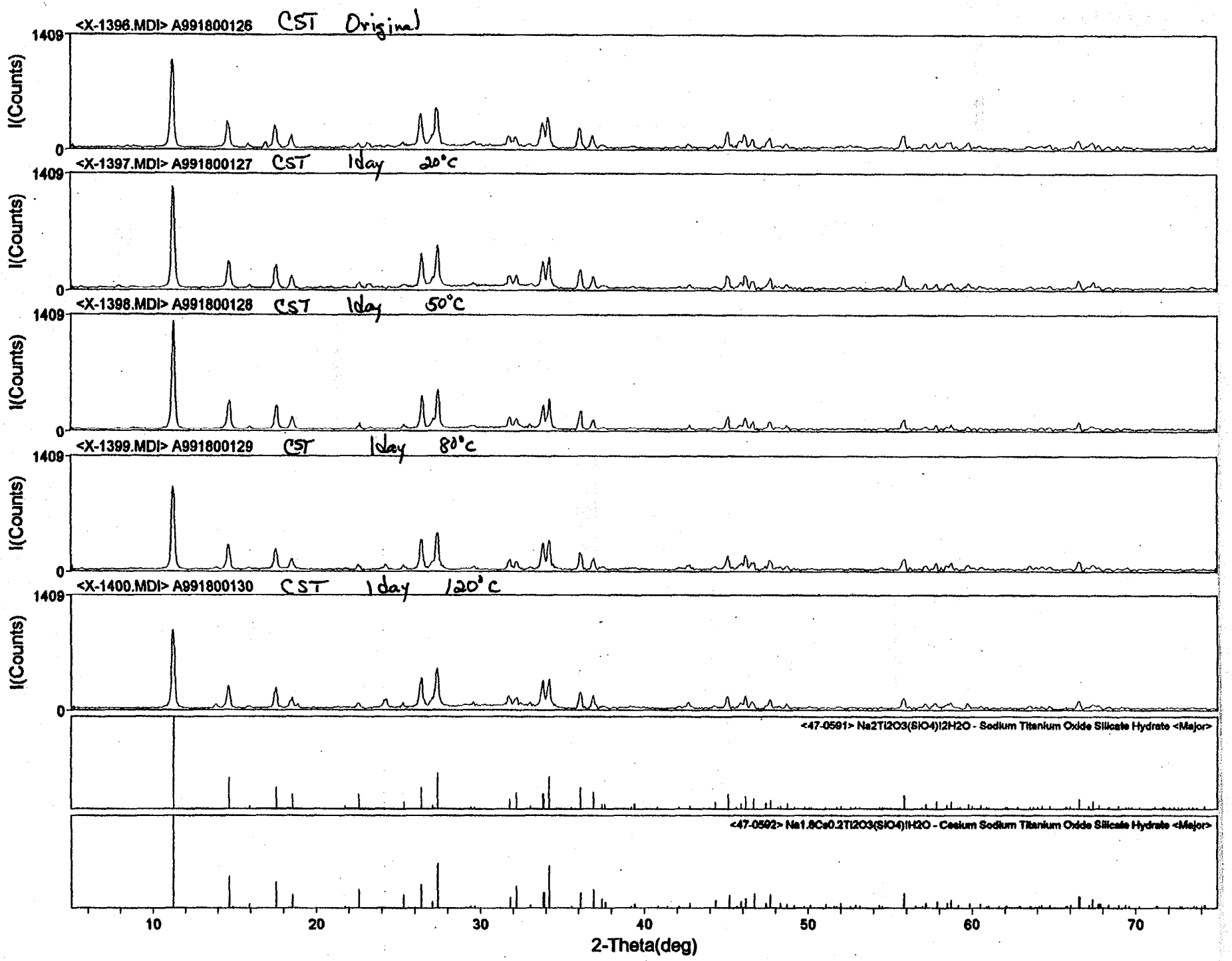
Date 8/24/1999

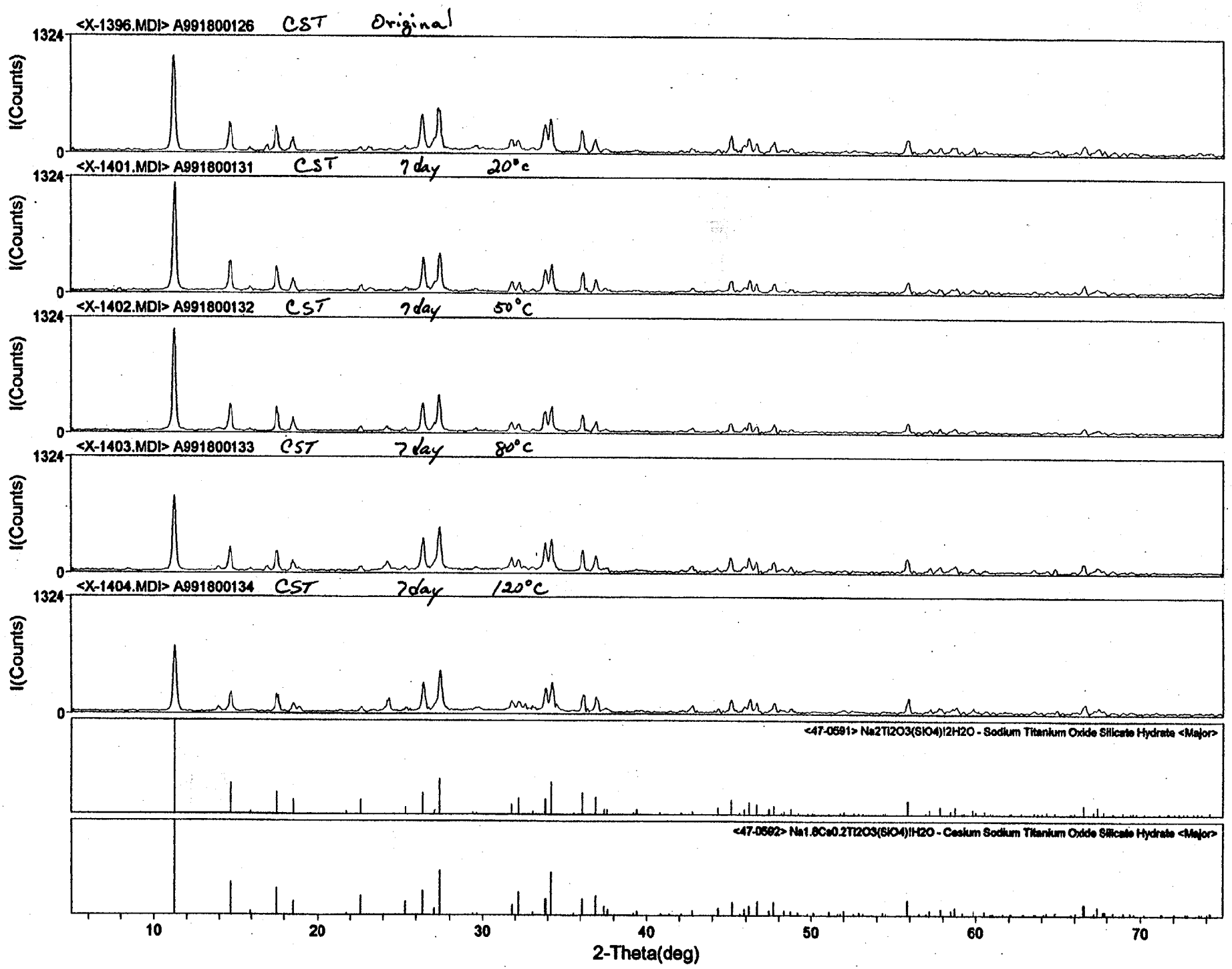
Background Count Rate	16	10	11	Average	12
Starting Solution Count Rate	10318	10312	10149	Average Net	10247

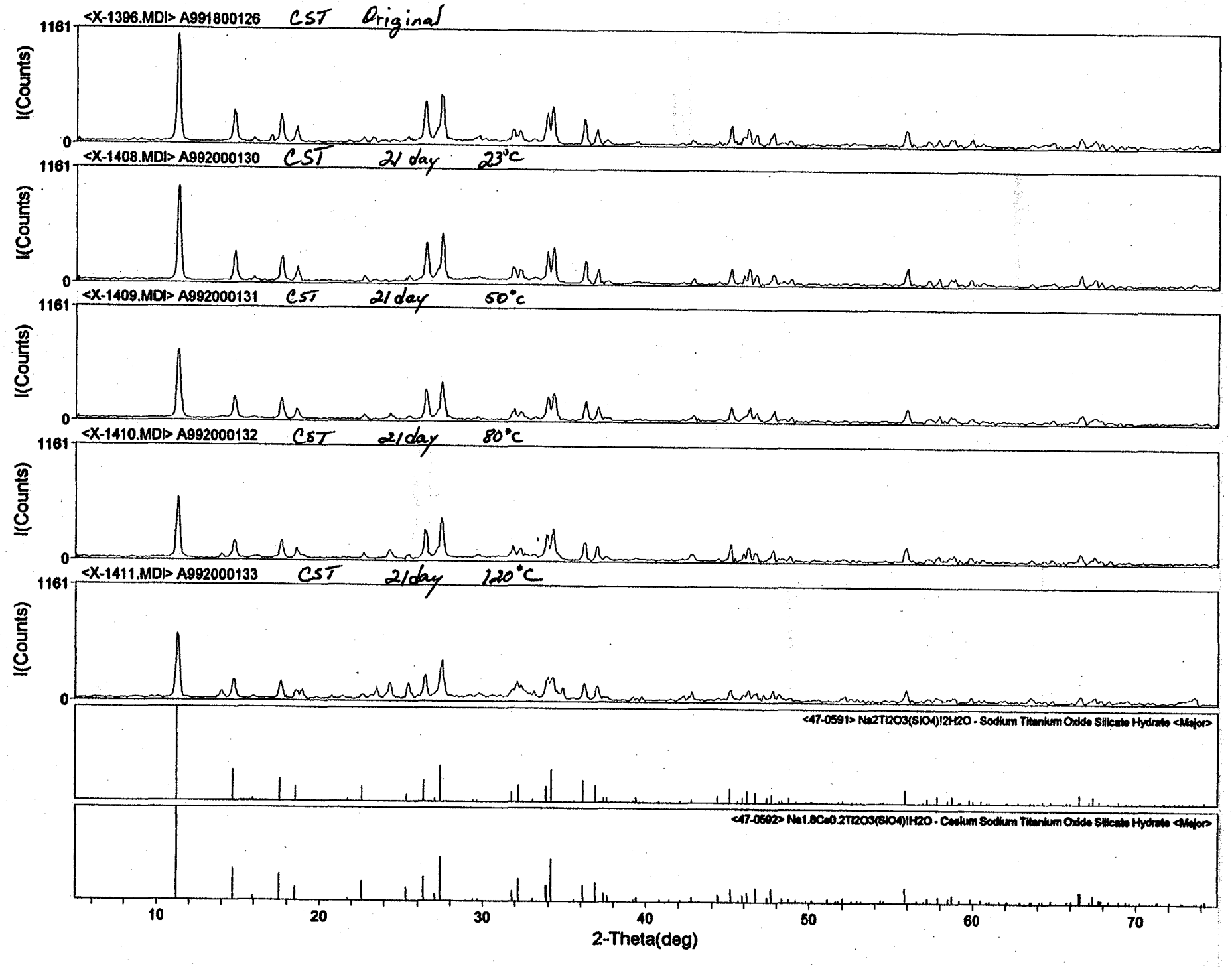
Sample ID	Storage temperature (°C)	CST weight (g)	Count rate (counts/1000 s/2 mL) Gross	Net	Cs conc. (mg/L)	Cs loading on CST (mg/g)	(meq/g)	Kd (L/kg)	Averages Kd (L/kg)	Cs Conc (mg/L)	Container average Cs Conc (mg/L)	Container average Kd (L/kg)
SRS-L-50-Ua	23	0	10236	10224	49.9						49.7	
SRS-L-50-Ub	23	0	10120	10108	49.4							
SRS-L-50-1a	23	0.1043	6226	6214	30.4	18.8	0.142	619	587	30.4	30.5	612
SRS-L-50-1b	23	0.1043	6282	6270	30.7	18.6	0.140	605				
SRS-L-50-2a	23	0.1166	6180	6168	30.2	17.0	0.128	564			30.2	562
SRS-L-50-2b	23	0.1166	6204	6192	30.3	16.9	0.127	559				
SRS-L-80-Ua	23	0	10212	10200	49.8						49.7	
SRS-L-80-Ub	23	0	10172	10160	49.6							
SRS-L-80-1a	23	0.0981	9049	9037	44.2	6.0	0.045	135	153	43.3	44.4	129
SRS-L-80-1b	23	0.0981	9143	9131	44.6	5.5	0.041	123				
SRS-L-80-2a	23	0.1042	8619	8607	42.1	7.6	0.057	181			42.2	177
SRS-L-80-2b	23	0.1042	8690	8678	42.4	7.3	0.055	172				
RS-L-120-Ua	23	0	9842	9830	48.0						47.6	
RS-L-120-Ub	23	0	9669	9657	47.2							
RS-L-120-1a	23	0.0955	9586	9574	46.8	3.4	0.025	72	76	46.6	46.5	78
RS-L-120-1b	23	0.0955	9492	9480	46.3	3.9	0.029	83				
RS-L-120-2a	23	0.0977	9568	9556	46.7	3.4	0.026	73			46.6	74
RS-L-120-2b	23	0.0977	9551	9539	46.6	3.5	0.026	75				

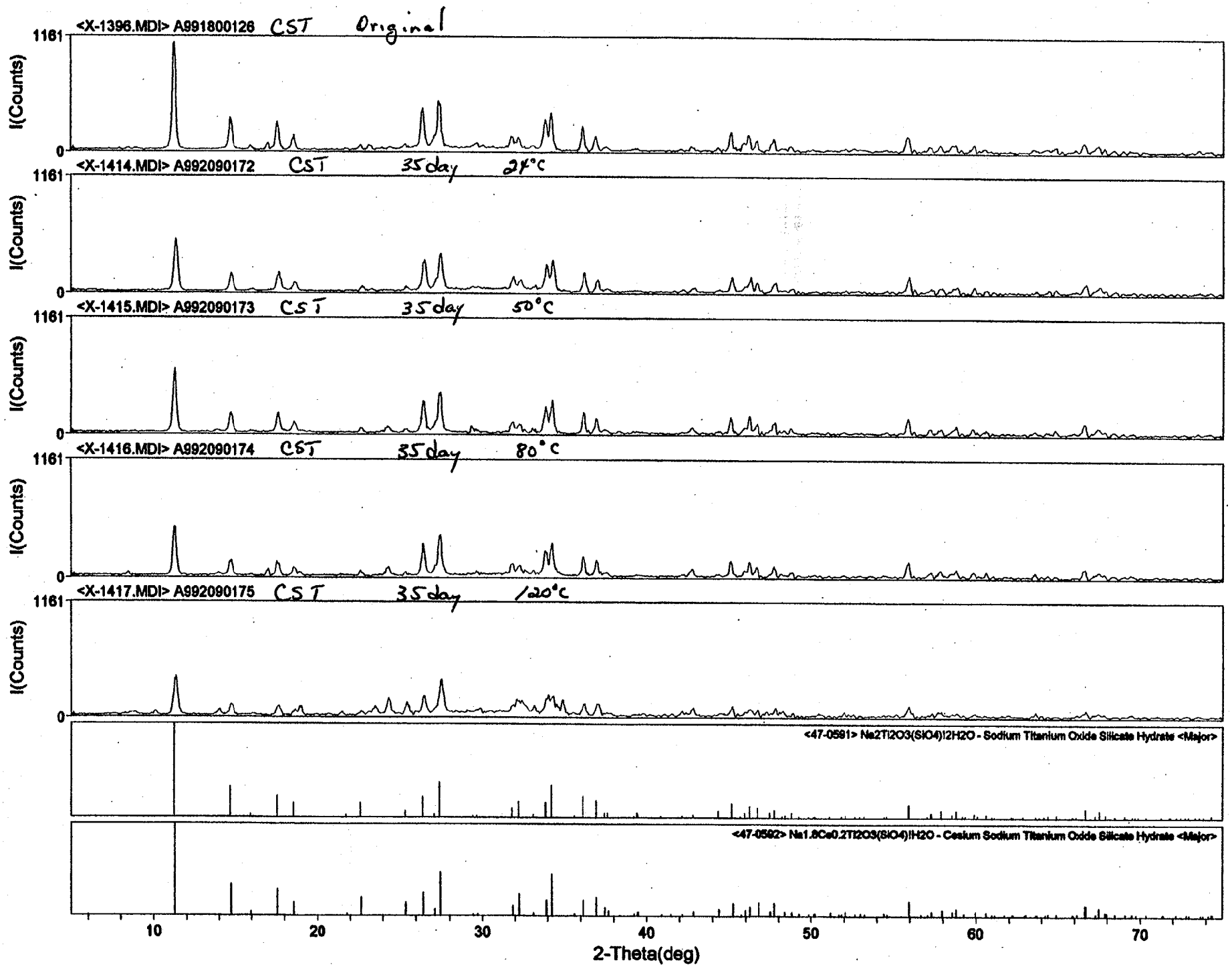
Appendix C

X-RAY DIFFRACTION PATTERNS FOR CST SAMPLES, STORED IN SRS SUPERNATE SIMULANT


<X-1396.MDI> A991800126 CST Original
<X-1397.MDI> A991800127 CST 1day 20°C
<X-1398.MDI> A991800128 CST 1day 50°C
<X-1399.MDI> A991800129 CST 1day 80°C
<X-1400.MDI> A991800130 CST 1day 120°C
I(Counts)
1409
0
<47-0591> Na2Ti2O3(SiO4)I2H2O - Sodium Titanium Oxide Silicate Hydrate <Major>
<47-0592> Na1.8Cs0.2Ti2O3(SiO4)IH2O - Cesium Sodium Titanium Oxide Silicate Hydrate <Major>
10
20
30
40
50
60
70
2-Theta(deg)


<X-1396.MDI> A991800126 CST Original
<X-1401.MDI> A991800131 CST 7 day 20°c
<X-1402.MDI> A991800132 CST 7 day 50°C
<X-1403.MDI> A991800133 CST 7 day 80°c
<X-1404.MDI> A991800134 CST 7day 120°C
I(Counts)
1324
0
<47-0591> Na2Ti2O3(SiO4)I2H2O - Sodium Titanium Oxide Silicate Hydrate <Major>
<47-0592> Na1.8Cs0.2Ti2O3(SiO4)IH2O - Cesium Sodium Titanium Oxide Silicate Hydrate <Major>
10
20
30
40
50
60
70
2-Theta(deg)


<X-1396.MDI> A991800126 CST Original
<X-1408.MDI> A992000130 CST 21 day 23°C
<X-1409.MDI> A992000131 CST 21 day 50°C
<X-1410.MDI> A992000132 CST 21 day 80°C
<X-1411.MDI> A992000133 CST 21 day 120°C
I(Counts)
1161
0
<47-0591> Na2Ti2O3(SiO4)!2H2O - Sodium Titanium Oxide Silicate Hydrate <Major>
<47-0592> Na1.8Ca0.2Ti2O3(SiO4)!H2O - Cesium Sodium Titanium Oxide Silicate Hydrate <Major>
10
20
30
40
50
60
70
2-Theta(deg)


<X-1396.MDI> A991800126 CST Original
<X-1414.MDI> A992090172 CST 35day 24°C
<X-1415.MDI> A992090173 CST 35day 50°C
<X-1416.MDI> A992090174 CST 35day 80°C
<X-1417.MDI> A992090175 CST 35day 120°C
1161
0
I(Counts)
<47-0591> Na2Ti2O3(SiO4)!2H2O - Sodium Titanium Oxide Silicate Hydrate <Major>
<47-0592> Na1.8Cs0.2Ti2O3(SiO4)!H2O - Cesium Sodium Titanium Oxide Silicate Hydrate <Major>
10
20
30
40
50
60
70
2-Theta(deg)

INTERNAL DISTRIBUTION

1. R T Jubin
2. T. E. Kent
3. D. D. Lee
4. C. H. Mattus
5. C. P. McGinnis
6. L E McNeese
7. S. M. Robinson
8-12. P. A. Taylor
13. T. D. Welch,
14. ORNL Central Research Library
15. Laboratory Records - RC
16. Laboratory Records, OSTI

EXTERNAL DISTRIBUTION

17. Jeffrey L Barnes, Westinghouse Savannah River Company, PO Box 616, 704-3N, Room S162, Aiken, SC 29808
18. Joe T Carter, Westinghouse Savannah River Company, PO Box 616, 704-3N, Room S151, Aiken, SC 29808
19. Samuel D Fink, Westinghouse Savannah River Company, PO Box 616, 773-A, Room B112, Aiken, SC 29808
20. Roger L Gilchrist, Pacific Northwest National Laboratory, PO Box 999, MS:K9-91, Richland, WA 99352
21. TS Gutmann, US Department of Energy, Savannah River Operations Office, PO Box A, Aiken, SC 29802
22. JO Honeyman, Lockheed Martin Hanford Corporation, PO Box 1500, MS: G3-21, Richland, WA 99352
23. R. A. Jacobs, Westinghouse Savannah River Company, PO Box 616, 704-3N, Room S252, Aiken, SC 29808
24. Jim Jewett, Numatec Hanford Corporation, P. O. Box 1970, Richland, WA 99352
25. R. A Kirkbride, Numatec Hanford Corporation, PO Box 1970, MS: H5-27 Richland, WA 99352
26. C. S. Louie, US Department of Energy, Richland Operations Office, PO Box 550, MSIN: B4-55, Richland, WA 99352
27. J. P. Morin, Westinghouse Savannah River Company, PO Box 616, Savannah River Technology Center, 703-H, Aiken, SC 29808
28. J. R. Noble-Dial, US Department of Energy, Oak Ridge Operations Office, PO Box 2001, Oak Ridge, TN 37831-8620
29. Arlin Olson, Idaho National Engineering and Environmental Laboratory, Building 637, MS-5218, Idaho Falls, ID 83415-5218
30. L. M. Papouchado, Westinghouse Savannah River Company, PO Box 616, 773-A, Room A-263, Aiken, SC 29808
31. Reid Peterson, Westinghouse Savannah River Company, PO Box 616, 773-A, Room B-132, Aiken, SC 29808

32. S. F. Piccolo, Westinghouse Savannah River Company, PO Box 616, 704-3N, Room S152, Aiken, SC 29808
33. Michael Poirier, Westinghouse Savannah River Company, PO Box 616, 679-T, Room 2A6, Aiken, SC 29808
34. J. M. Reynolds II, US Department of Energy, Savannah River Operations Office, PO Box A, Building 704-196N, Room S441, Aiken, SC 29802
35. Jim Rindfleisch, Idaho National Engineering and Environmental Laboratory, Building 637, MS-5218, Idaho Falls, ID 83415-5218
36. K. J. Rueter, Westinghouse Savannah River Company, PO Box 616, 706-S, Room 103, Aiken, SC 29808
37. W. W. Schulz, 12704 Sandia Ridge Place NE, Albuquerque, NM 87111
38. Patricia C Suggs, US Department of Energy, Savannah River Operations Office, PO Box A, Building 704-196N, Room S431, Aiken, SC 29802
39. J. L. Swanson, 1318 Cottonwood, Richland, WA 99352
40. W. L. Tamosaitis, Westinghouse Savannah River Company, PO Box 616, 773-A, Room A-231, Aiken, SC 29808
41. Larry Tavlarides, Syracus University, Dept. of Chemical Engineering & Materials Science 334 Hinds Hall, Syracuse, NY 13244-1190
42. T. A. Todd, Idaho National Engineering and Environmental Laboratory, Building 637, MS-5218, Idaho Falls, ID 83415-5218
43. George Vandegrift, Argonne National Laboratory, Building 205, 9700 South Cass Avenue, Argonne, IL 60439
44. J. H. Westsik, Pacific Northwest National Laboratory, PO Box 999, MS:K9-91, Richland, WA 99352

45-53. Tanks Focus Area Technical Team, c/o BJ Williams, Pacific Northwest National Laboratory, PO Box 999, MSIN K9-69, Richland, WA 99352

54. Tanks Focus Area Field Lead, c/o TP Pietrok, US Department of Energy, Richland Operations Office, PO Box 550, MS: K8-50, Richland, WA 99352