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**Thermal and Chemical Stability
of Crystalline Silicotitanate Sorbent –
Interim Report**

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**THERMAL AND CHEMICAL STABILITY OF
CRYSTALLINE SILICOTITANATE SORBENT –
INTERIM REPORT**

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ABSTRACT

The Savannah River Site (SRS) is evaluating technologies for removing radioactive cesium (^{137}Cs) from the supernate solutions stored in the high-level waste tanks at the site. Crystalline silicotitanate sorbent (IONSIV IE-911[®], UOP LLC, Mt. Laurel, NJ), which is very effective at removing cesium from high-salt solution, is one of three technologies currently being tested. Because of the extremely high loading of ^{137}Cs that would be expected in 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. Also, even for normal operation, the CST would be exposed to the supernates for long periods of time, up to a year before being removed. Small-scale tests using simulant solutions are in progress to determine the long-term stability of the CST to the solutions at various temperatures.

The cesium loading capacity of the CST drops rapidly as the temperature increases from 25 to 80°C. CST stored in cesium-free simulant solutions for one to seven months at 80°C shows a 19% drop in capacity and a 30% drop in cesium distribution coefficient compared to CST stored at 25°C, when the cesium loading is performed at room temperature. CST exposed to recirculating simulant solution at room temperature in a column test has shown a slow decrease in cesium distribution coefficient, with a drop of 23% after six months, which is a drop in Cs loading capacity of 12%. CST in both the batch and column tests has shown a tendency to become cemented together with sodium aluminosilicate, formed from aluminum, silicon and sodium from the simulant solutions and additional silicon leached from the CST. These clumps have the potential of causing fluid channeling within a column, and of making the CST more difficult to sluice from a column. The CST used in these tests was pretreated at a lower sodium hydroxide concentration than is present in the simulants. Future tests will determine if pretreating the CST at higher sodium hydroxide concentrations reduces the tendency to form clumps.

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), an inorganic ion exchange material with a high affinity for cesium, is the key material in one of the processes being tested. Work conducted at ORNL last year showed that higher temperatures could cause desorption of previously loaded cesium from CST, and that the CST would not reload the cesium when the temperature was reduced following 60 days at the higher temperatures.¹ 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.

Long-term batch and flow-through leaching tests are being conducted to determine any detrimental effect from long-term exposure of the CST to supernate simulant solutions under various conditions. Simulant solutions that represent the average and extremes of composition expected for the SRS supernates during treatment are being used, along with a simplified simulant called high-pH salt solution, that only contains sodium nitrate and sodium hydroxide.

MATERIALS AND METHODS

Most of the CST used for these tests is the commercial, granular form of the sorbent, IONSIV IE-911[®] (UOP LLC, Mt. Laurel, NJ). Lot 999098810005 CST (98-5), which was manufactured in 1998 using a chloride-based process, is being used for all of the test conditions. A few of the test conditions are duplicated using Lot 999099810007 (99-7) CST, which was manufactured in 1999 using a newer, nitrate-based process, and IE-910 (the CST powder which was used to make the 1998 batches of IE-911). The as-received granular CST was contacted with dilute sodium hydroxide to stabilize the pH at ~13 and then backwashed to remove fines prior to use. A standardized procedure for pretreating CST for use with the SRS simulants was developed after these tests were started that uses once-through NaOH at the same concentration as the simulant². Future tests will examine whether the different pretreatment methods affect the results of the stability tests.

Loading Tests

Batch loading tests were performed at temperatures of 25, 30, 35, 40, 45, 50, 60, 70 and 80°C, using 0.1 g of CST and 100 mL of supernate simulant, which contained 50 mg/L Cs and 0.1 $\mu\text{Ci/L}$ ¹³⁷Cs. The CST and simulant were mixed at 120 rpm in Model 3527-5 Environ Shakers (Lab-Line Instrument, Inc., Melrose Park, IL), inside of capped Teflon flasks. The temperature of the samples was maintained within $\pm 0.2^\circ\text{C}$ by a built-in, forced-air convection heater with a temperature controller. The temperatures were checked using a digital thermometer with NIST traceable calibration.

Two samples of the supernate from each flask were filtered through 0.2- $\mu\text{-m}$ -pore size, nylon syringe filters. A 2.00-mL portion of each filtered sample was pipetted into a polyethylene counting tube, capped, centrifuged for about 2 minutes 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 ¹³⁷Cs. Three samples of the starting supernate simulant and three background rates were counted for each series of samples.

Column Leaching Tests

Average concentration supernate simulant and high-pH salt solution are being recirculated through small PVC columns (1.6 cm I.D. by 120 cm tall) containing pretreated 98-5 CST at room temperature. The columns were initially filled with 235 g (220 cm³) of CST, which gave a bed height of 110 cm. The flow rate of the solutions is 6 mL/min. The solutions are pumped through a 0.45- $\mu\text{-m}$ -pore size filter cartridge before entering the column, using a dual-head peristaltic

pump and silicon rubber tubing. Figure 1 shows a picture of the columns, pump and filters. There are pressure gauges on the inlet of the filters. Samples of the solutions are analyzed periodically for dissolved metals, to measure CST leaching and precipitation of simulant components. The solutions are replaced when the concentration of the solutions changes significantly. Samples of the CST are removed from the top, middle and bottom of the column each month and tested for cesium sorption, porosity, surface area, particle size and elemental composition. These tests will continue for 12 months.

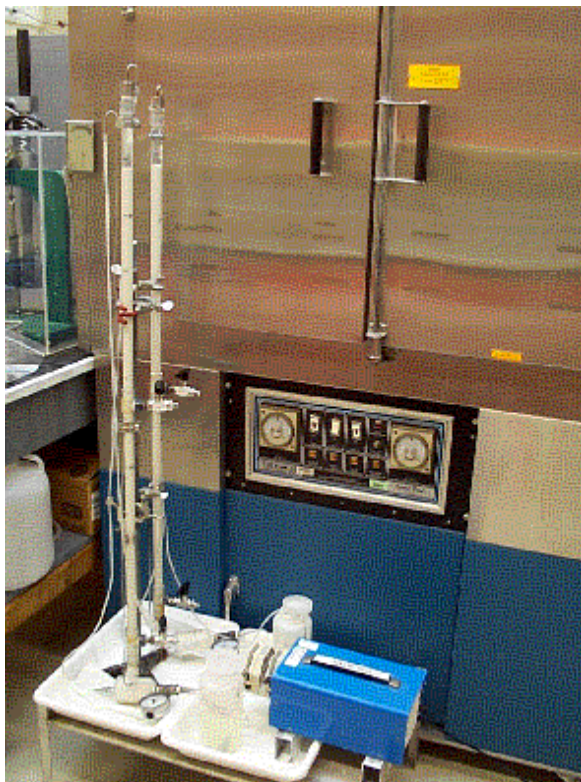


Figure 1. Photograph of column leaching test equipment.

Batch Leaching Tests

Long-term (12 month) batch leaching tests are being conducted using the average, high hydroxide and high nitrate supernate simulants and the high-pH salt solution to determine the effect of temperature and solution composition on the stability of the CST. Teflon bottles containing 16 g of CST and 240 mL of simulant are stored at room temperature and 30, 35, 50 and 80°C. Samples of the solutions are analyzed each month for dissolved metals, to measure CST leaching and/or precipitation of simulant components. Samples of the CST are removed each month and tested for cesium sorption, porosity, surface area, particle size and elemental composition. Lot 98-5 CST is being used for all of these test conditions, with lot 99-7 CST and IE-910 CST powder also tested at 25 and 80°C in average simulant. A room temperature leaching test using average simulant containing 50 mg/L Cs and 98-5 CST, that was started on 6/15/99, is being continued.

Analytical Methods

The CST samples from the leaching tests are rinsed three times with a small amount of deionized water, and then dried at room temperature in a dessicator prior to analysis. Samples of CST were contacted with each of the simulant solutions for one minute and then rinsed and dried using the same procedure as for the batch and column leaching tests. These samples were used as the baseline (zero time) to compare all of the analytical results for the leaching test samples. A portion of four samples from each set of samples was dried to a constant weight at 105°C and then at 400°C for four hours to determine the amount of moisture in the CST. The average weight loss for the CST samples has been 3.84 wt% at 105°C and 13.12 wt% at 400°C. The cesium loading tests used average simulant containing 50 mg/L Cs, and the CST and solution were contacted for seven days at room temperature. Samples were collected the same as for the batch loading tests, except that they were counted on a Model 1282 Compugamma (LKB Wallac) with a sodium iodide detector. This counter has an automatic sample changer, which facilitates analyzing the large number of samples generated from the leaching tests.

The ICP analyses were performed 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. Both simulant solutions and CST samples were digested using nitric and hydrofluoric acid in a microwave oven to solubilize the materials, and then boric acid was added to complex the fluoride ions prior to analysis. The surface area and pore volume of the CST samples was measured using a Gemini III 2375 Surface Area Analyzer instrument. Selected samples of the CST were examined with a scanning electron microscope to identify any changes in the morphology of the particles.

Simulant Solutions

Average, high-nitrate and high-hydroxide supernate simulants and a high-pH salt solution were prepared at ORNL using recipes supplied by SRS³. The compositions are shown in Table 1. The simulants were prepared in large batches (5 L for the high hydroxide and high nitrate, and 20 L for the average and high-pH salt solution) without any cesium. All of these solutions were used for the batch leaching tests, and the average simulant and high-pH salt solution were also used for the column leaching tests. A portion of each solution was spiked with 50 mg/L Cs and 1 μ Ci/L ¹³⁷Cs for use in the cesium-loading tests. Each large batch of simulant was analyzed by ICP for metals and ion chromatography for anions. The spiked batches of simulant were analyzed for cesium by ICP-MS and for ¹³⁷Cs by gamma counting at the ORNL Radioactive Materials Analysis Laboratory. Two large batches (# 4 and 5) of the average simulant have been prepared and used to-date, with only one batch of the remaining solutions used. A total of eight batches (4A-4D and 5A-5D) of spiked (Cs and ¹³⁷Cs) average simulant have been prepared from the two large batches and used for the loading tests of the CST samples from the batch and column leaching tests. The analytical results for the spiked batches of average simulant are shown in Table 2.

Table I. Composition of Simulated Waste Solutions

Component	Concentration (molar)			
	Average	High OH ⁻	High NO ₃ ⁻	High-pH Salt
Na ⁺	5.6	5.6	5.6	5.6
K ⁺	0.015	0.030	0.0041	
OH ⁻	1.91	3.05	1.17	1.91
NO ₃ ⁻	2.14	1.10	2.84	3.69
NO ₂ ⁻	0.52	0.74	0.37	
AlO ₂ ⁻	0.31	0.27	0.32	
CO ₃ ²⁻	0.16	0.17	0.16	
SO ₄ ²⁻	0.15	0.030	0.22	
Cl ⁻	0.025	0.010	0.040	
F ⁻	0.032	0.010	0.050	
PO ₄ ³⁻	0.010	0.008	0.010	
C ₂ O ₄ ²⁻	0.008	0.008	0.008	
SiO ₃ ²⁻	0.004	0.004	0.004	
MoO ₄ ²⁻	0.0002	0.0002	0.0002	

Table 2. Analytical Results For Batches of Simulant Used in Loading Tests.

Batch I.D.	[Cs] (mg/L)	[¹³⁷ Cs] (μCi/L)	Used in Tests:
AVG 4-A	50.0	1.19	Loading tests and 1Month column
HH-1A	48.6	1.11	Loading tests
HN-1A	49.1	1.11	Loading tests
HS-1A	51.8	0.86	Loading tests
AVG 4-B	45.9	0.73	1 Month batch
AVG 4-C	42.0	1.05	2 Month batch
AVG 4-D	47.7	1.19	3 Month batch, 2 & 3 Month column
AVG 5-A	52.4	1.22	4 Month batch and column
AVG 5-B	48.6	1.08	5 Month batch and column
AVG 5-C	57.5	1.24	6 Month batch and column
AVG 5-D	49.6	1.22	7 Month batch and column

RESULTS AND DISCUSSION

Loading Tests

The results of the cesium loading tests are shown in Figure 2. Temperature has a strong effect on the distribution coefficient ($K_d = \text{Cs loading on CST}/[\text{Cs}]$ in simulant) for cesium loading on the CST. The results are calculated based on a dry weight of the CST at 400°C, which was measured

using subsamples of the CST that were not used in the loading tests. The high hydroxide simulant shows the highest K_d , followed by the average and high nitrate simulants, and then by the high-pH salt solution. The prediction of the K_d by the ZAM equilibrium model⁴ (adjusted to a 400°C dry weight) for the high hydroxide simulant at each temperature is shown for comparison. The predictions for temperatures of 45 to 80°C are above the models validated range of 25 to 44 °C. The high hydroxide and average simulants have measured values about 15% lower than the predictions for temperatures up to 35°C, and then the results increasingly diverge from the model at the higher temperatures. The measurements in the high-pH salt solution were about 10% below the model predictions and the high nitrate simulant results matched the predictions for temperatures up to 35°C, and then both diverged at the higher temperatures. All of the solutions had measured K_d s that were a factor of three below the predictions at 80°C. The K_d results for the lot 99-7 CST and the IE-910 powder were 1487 and 1962 L/kg at 24°C and 162 and 323 L/kg at 80°C, respectively. The cesium capacity dropped by about 78% for all of the IE-911 samples and by 65% for the IE-910 samples at 80°C compared to the results at 25°C.

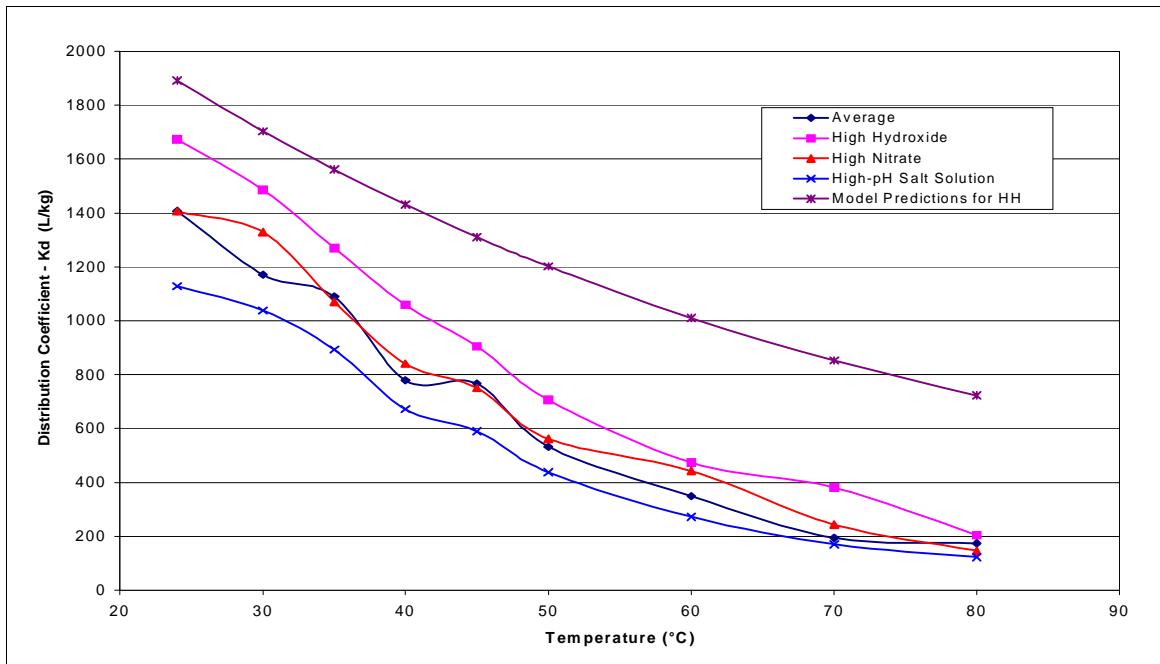


Fig. 2. Results of cesium loading tests using CST and simulant solutions, and comparison with predictions of the ZAM model for the high hydroxide simulant.

Batch Leaching Tests

After storage for one month at 30, 35, 50 and 80°C, the CST stored in the average, high hydroxide and high nitrate simulants was weakly cemented together. All of the samples stored at 25°C, and the CST in the high-pH salt solution at all temperatures, were still free flowing after one month. The cemented CST could be easily broken up using a plastic spoon. After two months storage the samples stored at 25°C in the average, high hydroxide and high nitrate simulants were also cemented together. During subsequent samplings, the CST that had been previously broken up did not reform into clumps. The CST stored in the high-pH salt solution has not formed any clumps at any storage temperature during the seven months that the tests have

been in progress. A test will be run in the near future to determine if CST that is pretreated using the standard SRS method² will also form clumps during storage in average simulant.

The cesium loading tests using CST samples from the batch leaching tests showed a drop of about 19% in cesium capacity and 30% in distribution coefficient for the samples stored for one month or more at 80°C, compared to samples stored at 25°C, for the average, high hydroxide and high nitrate simulant solutions. The ratio has not changed as the storage time has increased. (Note - All of the cesium loading tests are performed at 25°C using average simulant that initially contains 50 mg/L Cs.) The high-pH salt solution caused less of an effect (see Table 3). These results indicate that there is an initial degradation in the cesium sorption properties of CST as it is contacted with supernate simulants at higher temperatures, sometime during the first month, but no further change after that. Figure 3 shows all of the cesium-loading results for the CST samples taken after five months storage in the simulant solutions. The CST stored at moderate temperatures also shows a drop in K_d compared to the samples stored at 25°C, an average of 15% and 18% reduction at 30 and 35°C, respectively.

Table 3. Average Cesium Distribution Coefficients for CST Samples Stored at 25 and 80°C for 1 to 7 Months

CST	Simulant	Distribution Coefficient (L/kg) ± s.d.		% Change
		Stored at 25°C	Stored at 80°C	
98-5	Average	1130 ± 100	764 ± 100	31.9
98-5	High Hydroxide	1141 ± 100	744 ± 80	34.0
98-5	High Nitrate	1088 ± 130	798 ± 100	26.6
98-5	High-pH Salt	1217 ± 160	968 ± 90	19.5
99-7	Average	1295 ± 160	917 ± 100	27.9

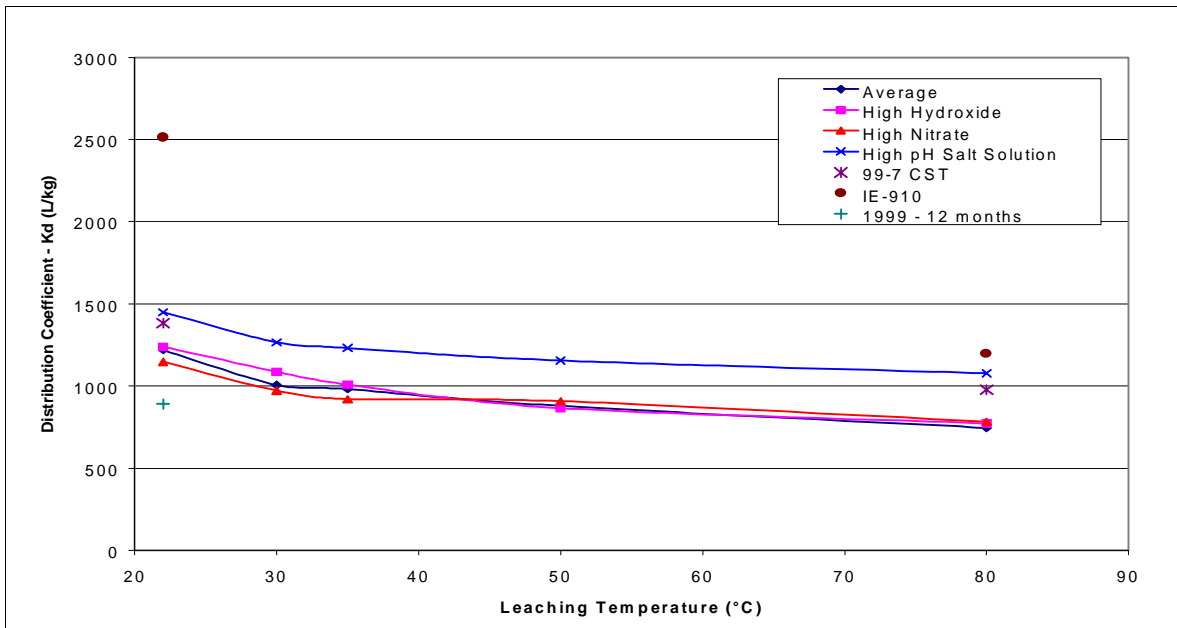


Fig. 3. Cesium Loading Results for CST Samples from the Batch Loading Tests After 5 Months. The 1999 Sample Has Been Exposed for 12 Months.

Figure 4 shows the results for granular CST (lot 98-5 IONSIV IE-911) and IE-910 CST powder stored in average simulant at 25 and 80°C for 0 to 7 months. The graph shows the initial drop in cesium distribution coefficient (K_d) for the samples stored at 80°C for one month. The granular CST does not show any further change in the K_d for storage times up to 7 months. All of the other granular CST samples show the same behavior. In contrast, the IE-910 powder shows a continuing drop in the cesium distribution coefficient as the storage time at 80°C increases. The cesium loading results have been very consistent for all of the CST samples run at one time, but there is significant month-to-month variation in the results. The reason for the variation is not known at this time. A number of possible causes have been examined, including composition of the simulant solutions, cesium-loading temperature and gamma counter efficiency, but none of these correlate with the changes in the cesium loading.

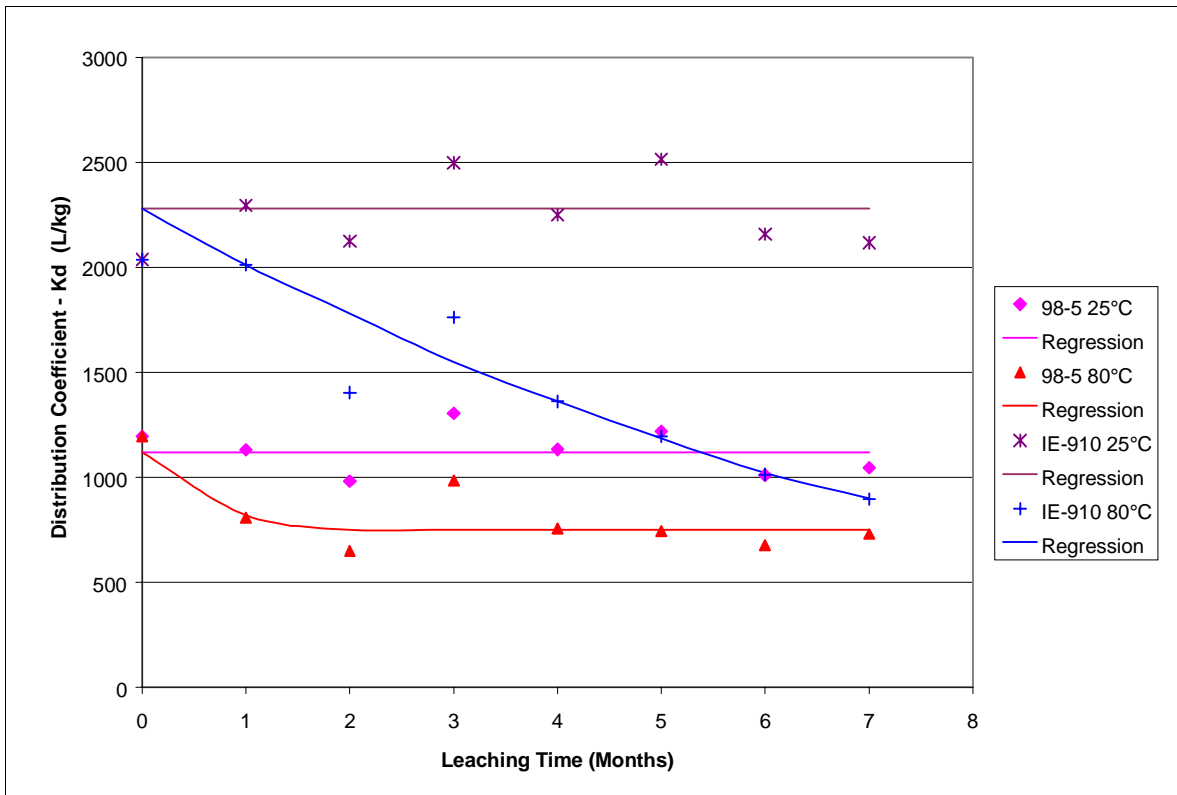


Fig. 4. Results of cesium loading tests for CST samples from batch leaching tests, using 98-5 granular CST and IE-910 powder stored in average simulant at 25 and 80°C.

In order to better understand the results, a cesium loading test was performed on one set of archived samples (98-5 CST stored in high hydroxide simulant at 50°C for 1 to 6 months). The results for this test, and the comparison results for the same samples run during the individual monthly loading tests, are shown in Figure 5. The amount of variation in the results was greatly reduced for the new test, which indicates that most of the variation is caused by experimental effects rather than a change in the CST samples. The results for the new measurements, and the trend of the data for all of the other samples of the granular CST (IONSIV IE-911), suggest that there has not been any change in the cesium loading results during the 7 months that the batch leaching tests have been in progress, after the initial drop during the first month.

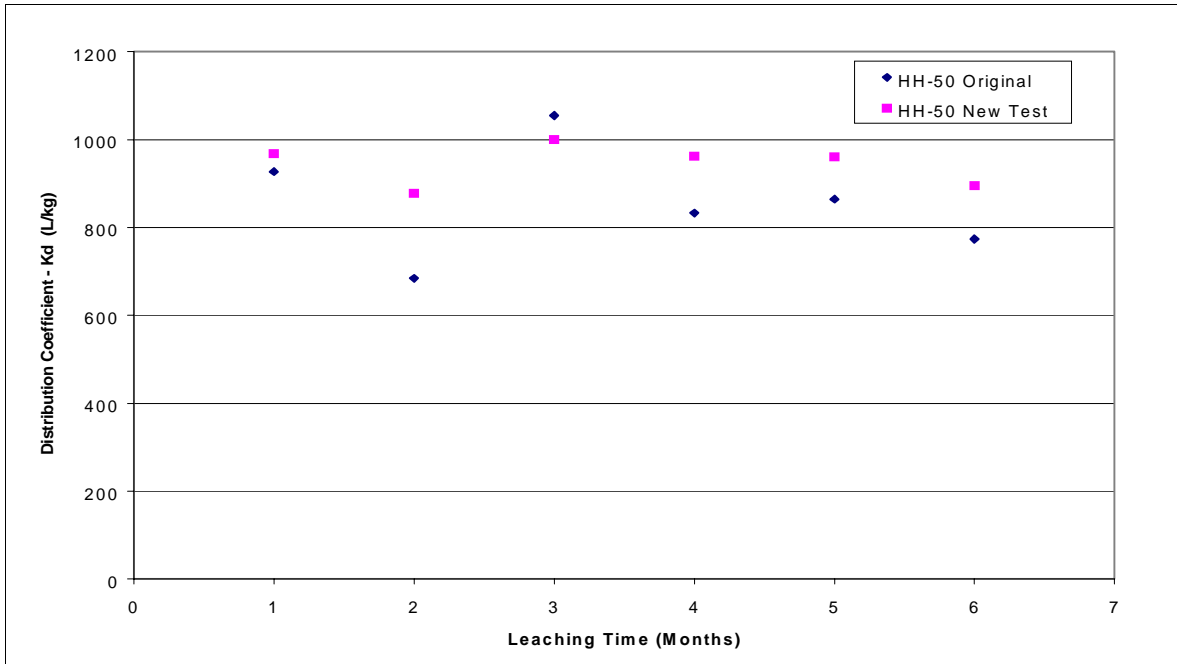


Fig. 5. Comparison of original results with new loading test for CST samples stored in high hydroxide simulant at 50°C for 1 to 6 months.

The IE-910 powder shows an increasing difference in cesium loading for the sample stored at 80°C, compared to that stored at 25°C, in average simulant (see Fig. 4). This would suggest that the powder stored at 80°C is degrading in some way, or being diluted by precipitated sodium aluminosilicate (see p. 12). The granular CST samples have not shown this effect, so it may be related to the high surface area of the powder. The newer, lot 99-7, CST has consistently shown slightly higher cesium loading than the lot 98-5 material. Storage in the high-pH salt solution has had the smallest effect on the cesium loading properties of the CST of any of the simulant solutions.

The surface area, total pore volume and micropore volume (volume of pores <15Å) were measured for selected samples of the CST. Samples from leaching tests conducted in 1999, which used average simulant containing 50 mg/L Cs, were initially measured. These results (see Table 4) suggested that there was a correlation between micropore volume and cesium uptake.

Table 4. Properties of CST Stored in Average Simulant Under Various Conditions.

Storage Conditions	Surface Area (m ² /g)	Total Pore Volume (cm ³ /g)	Micropore Volume (cm ³ /g)	Cesium Kd (L/kg)
133 days at 23°C	30.3	0.047	0.0036	1218
50°C for 86 days, then 23°C for 59 days	25.4	0.041	0.0035	692
80°C for 86 days, then 23°C for 59 days	37.3	0.057	0.0020	644
120°C for 86 days, then 23°C for 59 days	24.8	0.076	0.0000	47

Measurements using a larger number of CST samples from the batch and column leaching tests this year have not shown any correlation between micropore volume, total pore volume or surface area and cesium loading. The results for micropore volume are shown in Figure 6.

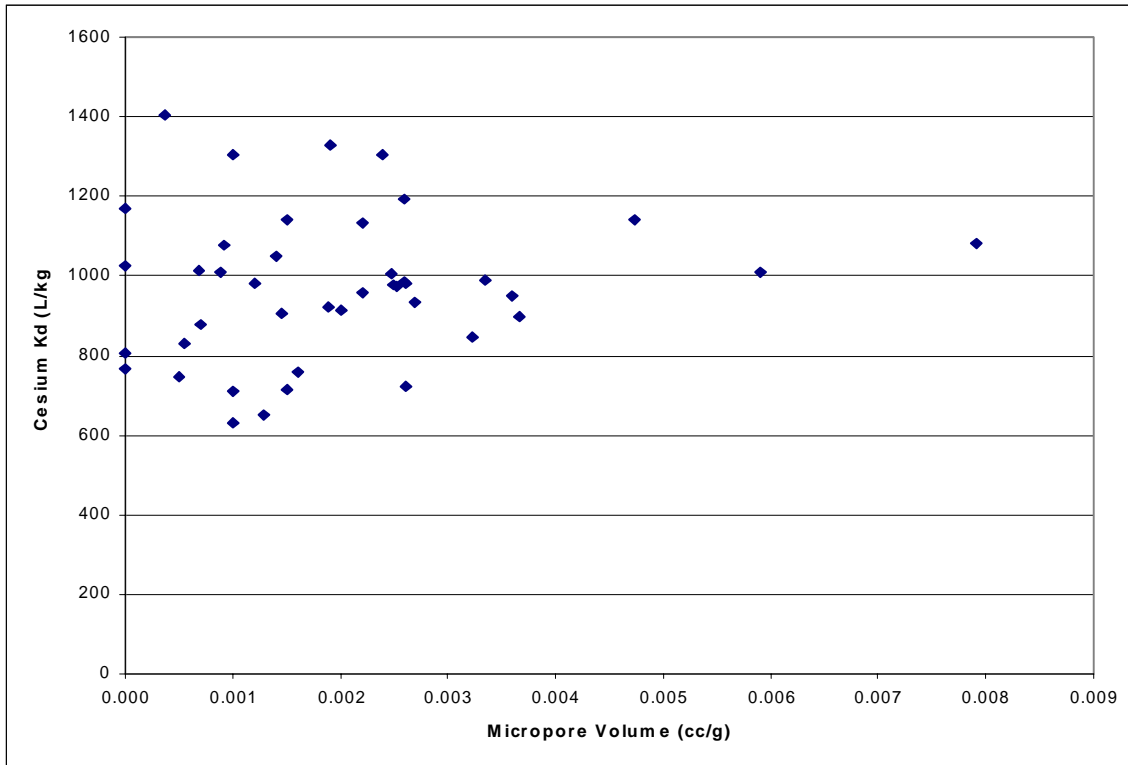


Fig. 6. Comparison of micropore volume and cesium distribution coefficient for CST samples from the batch and column leaching tests.

The photomicrographs of the CST samples stored in average, high hydroxide and high nitrate simulant solutions, particularly those stored at higher temperatures, show precipitates on the surface of the CST particles (see Fig. 7). Energy dispersive X-ray (EDX) spectroscopy showed that the precipitates were composed of sodium, silicon and aluminum. The CST stored in the high-pH salt solution has not shown any precipitates, or other changes in the CST particles. ICP analysis of the simulant solutions from the batch leaching tests has generally shown a slight decrease in the aluminum concentration and a small increase in silicon concentration.

One of the proprietary materials of the CST, called proprietary material 1 (PM-1), has shown some unusual behavior. The results for the average simulant are shown in Figure 8, and the high hydroxide and high nitrate simulants showed similar results. The concentration of PM-1 initially increases, particularly at the lower temperatures, but then usually decreases, presumably as it precipitates. At 80°C the concentration was initially low, but continues to increase over time, approaching the concentration in the 25°C solution after 7 months. In the high-pH salt solution, the PM-1 concentration increased during the first month and then leveled off and remained fairly constant at 150 mg/L at 25°C and 70 mg/L at 80°C. There have not been any significant changes

in the chemical composition of the CST samples from these tests, except for the aluminum concentration, which increased for the first two months and then leveled off, as aluminum from the simulant solutions was incorporated into the IE-911 CST particles. The aluminum concentration in the IE-910 CST powder has continued to increase over time, from 9 g/kg after one month to 21 g/kg after 7 months. The increasing weight of sodium aluminosilicate on the CST powder could be a major cause of the decrease in cesium loading that has been observed.

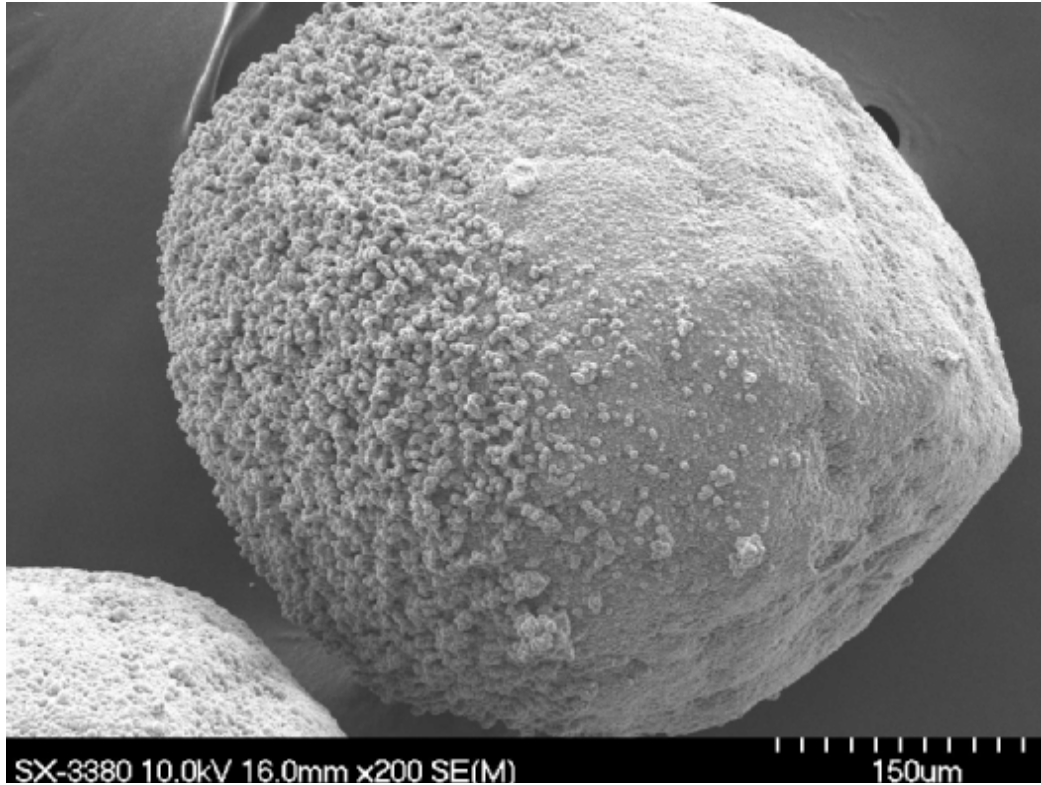


Fig. 7. Photomicrograph of CST stored in high nitrate simulant at 80°C for four months.

Column Leaching Tests

The back pressure for the filter and column combined averaged 4.4 psig for the column with average simulant during the first 10 days, and then the pressure increased to 6.0 psig for the next 20 days. The back pressure for the high-pH salt solution column has remained steady at about 3.5 to 4.7 psig during the seven months that the test has been running. The pressures and flow rates for both columns during the first three months of operation are shown in Figure 9.

The CST in the top of the average simulant column was clumped together when the first sample was taken after one month. The back pressure in the column was reduced after sampling. The pressure increased again during the second month of operation, reaching 8.0 psig when the two-month samples were taken. This time the pressure did not drop after sampling, continuing above 8 psig until the column was backwashed on day 67. The column was backwashed for 2 minutes at a flow rate of 15 mL/min, which suspended some fines above the bed of CST, but was not vigorous enough to remove the fines from the column. The CST throughout the column was

lightly cemented together, and tended to move up the column during backwashing. Light tapping on the column helped breakup the clumps of CST and resettle the bed. The back pressure has remained below 6 psig since the column was backwashed (see Fig 9). Apparently redistributing the fines and breaking up the clumps of CST was sufficient to reduce the back pressure.

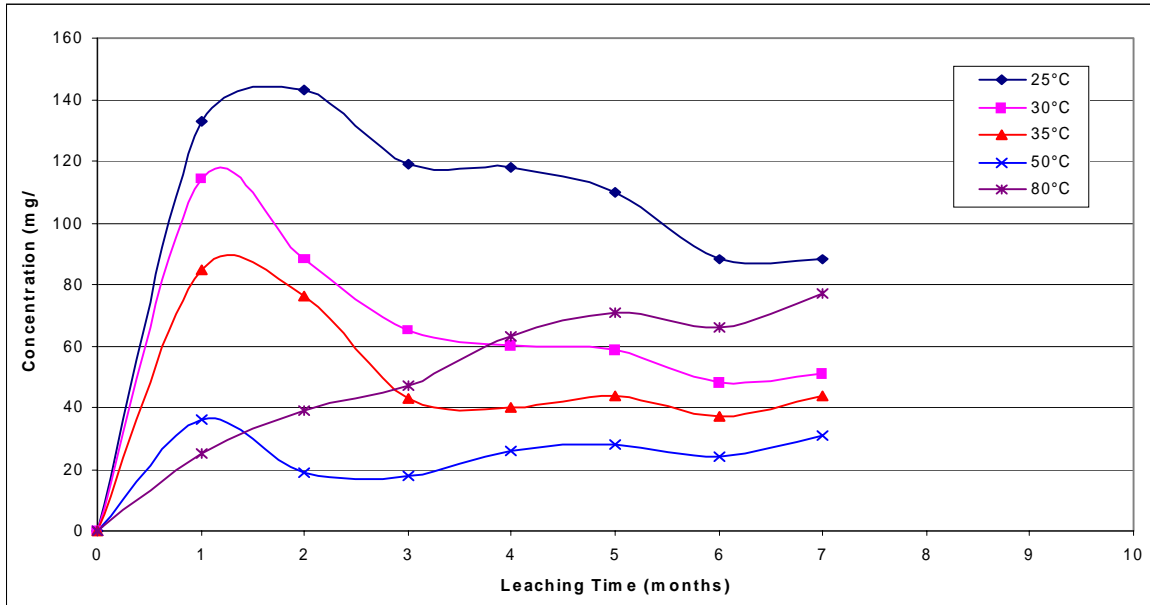


Fig. 8. Concentration of PM-1 in Average Simulant Solutions From Batch Leaching Tests.

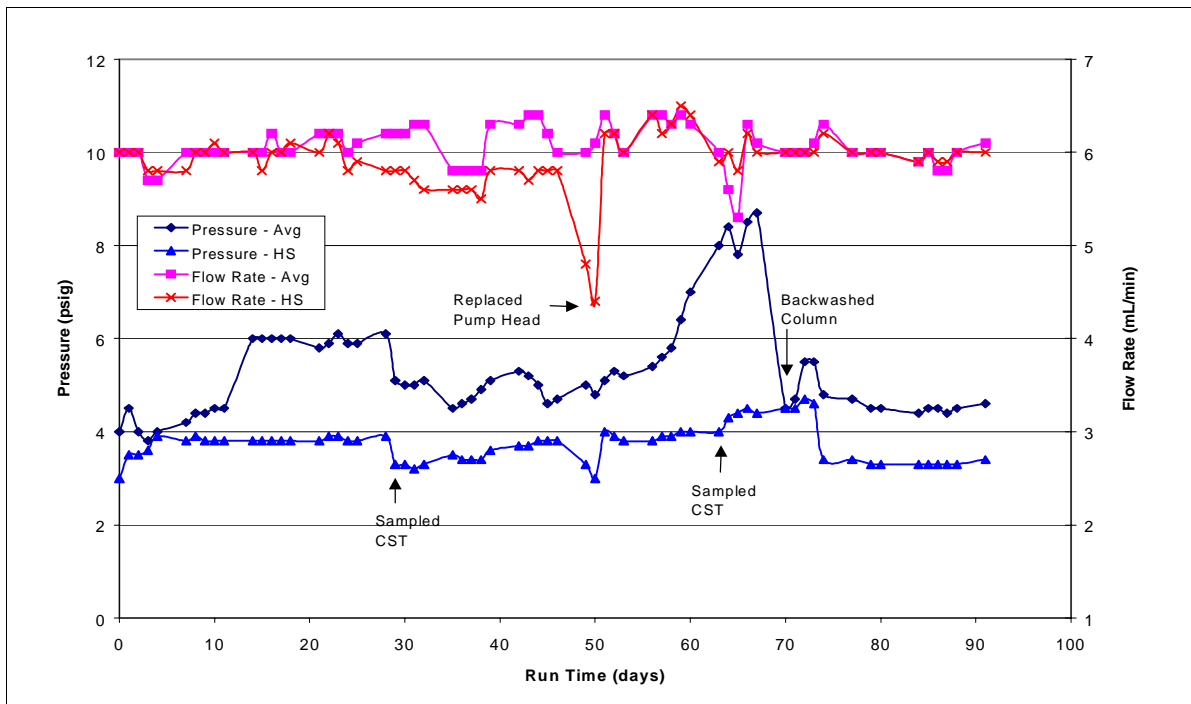


Fig. 9. Flow rate and pressure drop data for column leaching tests.

The cesium distribution coefficients for the CST from the column leaching tests are shown in Figure 10. The data plotted are the average K_d s for the top, middle and bottom CST samples from each column. The CST from the column using average simulant has consistently shown a slightly lower K_d for the sample from the top compared to the middle and bottom samples, while there is no difference in the samples from the column using high-pH salt solution. The results for the two-month samples from both columns are unusually high, and out of line with all of the other results, so these results probably do not represent an actual change in the CST. The CST from the column using the high-pH salt solution has not shown any consistent change in the cesium capacity of the samples, but the samples from the average simulant column show a slight decline over time. The distribution coefficient is 23% lower and the cesium loading on the CST is 12% lower after 7 months exposure to the average simulant. There has been a slow increase in the aluminum concentration of the CST samples from the average simulant column, indicating increasing amounts of sodium aluminosilicate on the CST, which may be the cause of the drop in cesium capacity.

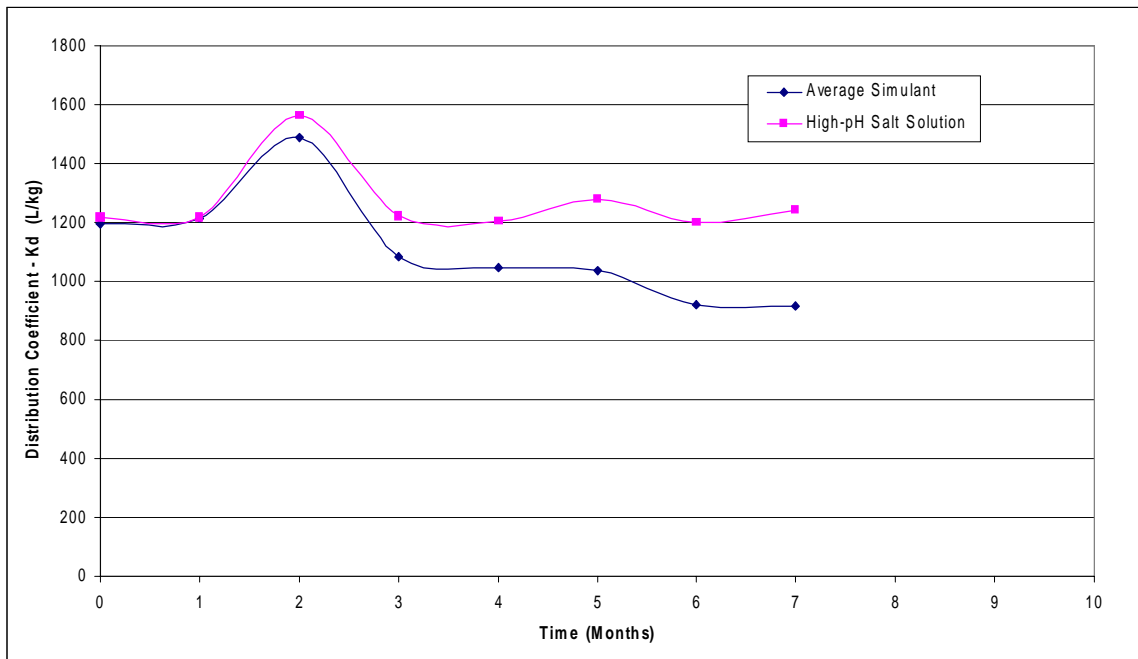


Fig. 10. Cesium distribution coefficients for CST samples from the column leaching tests.

The ICP results for aluminum, silicon and proprietary material 1 (PM-1) in the column feed solutions are shown in Figures 11 and 12 (note that different y-axis scales are used). For the average simulant column the silicon concentration slowly increases and then decreases along with the aluminum concentration, indicating the precipitation of sodium aluminosilicate. The concentrations were restored when the feed solution was changed, and the aluminum concentration has remained constant since that time. The silicon concentration has continued to change, but has not shown any consistent trend, so this may just be analytical variation. For the column with high-pH salt solution, the silicon concentration increased during the first ten weeks, but dropped back to the starting level when the feed solution was changed. As with the results for the average simulant column, there has been considerable scatter in the silicon concentrations. The analysis of the starting solution (0 weeks) showed 500 mg/L Si, even though there was not

any silicon in the high-pH salt solution, again indicating considerable analytical variation. For both solutions the PM-1 material rapidly leached from the CST, up to an apparent solubility limit, each time new feed solution was introduced.

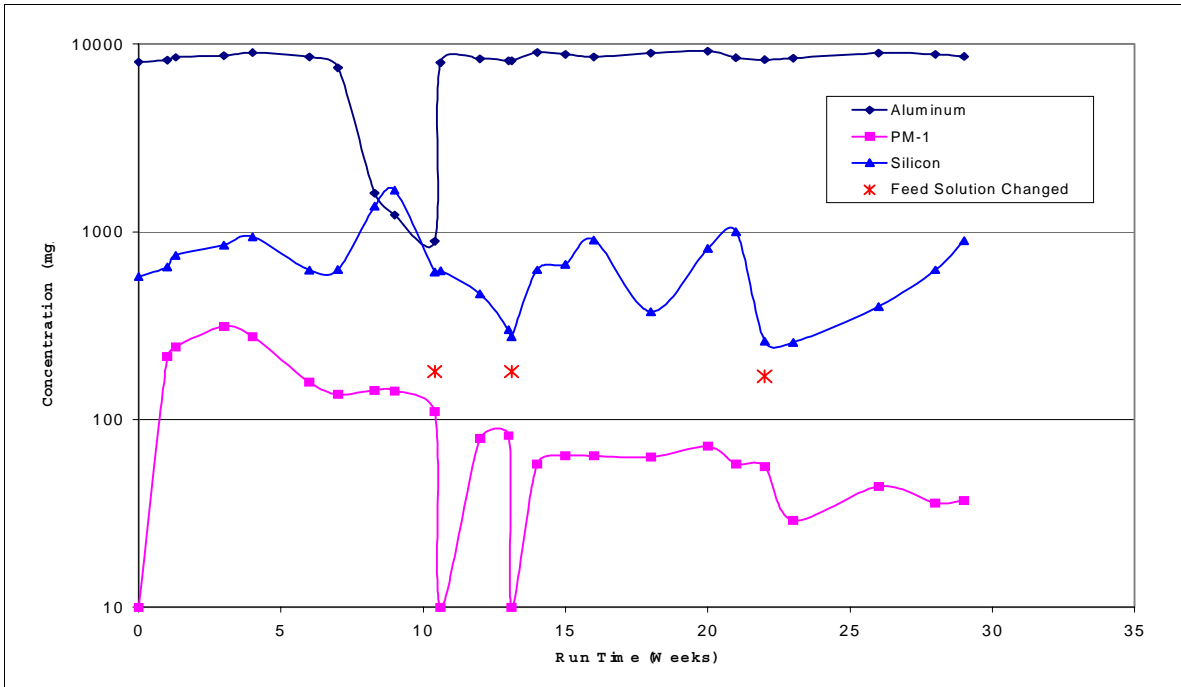


Fig. 11. Metal concentrations in the average simulant from the column leaching test.

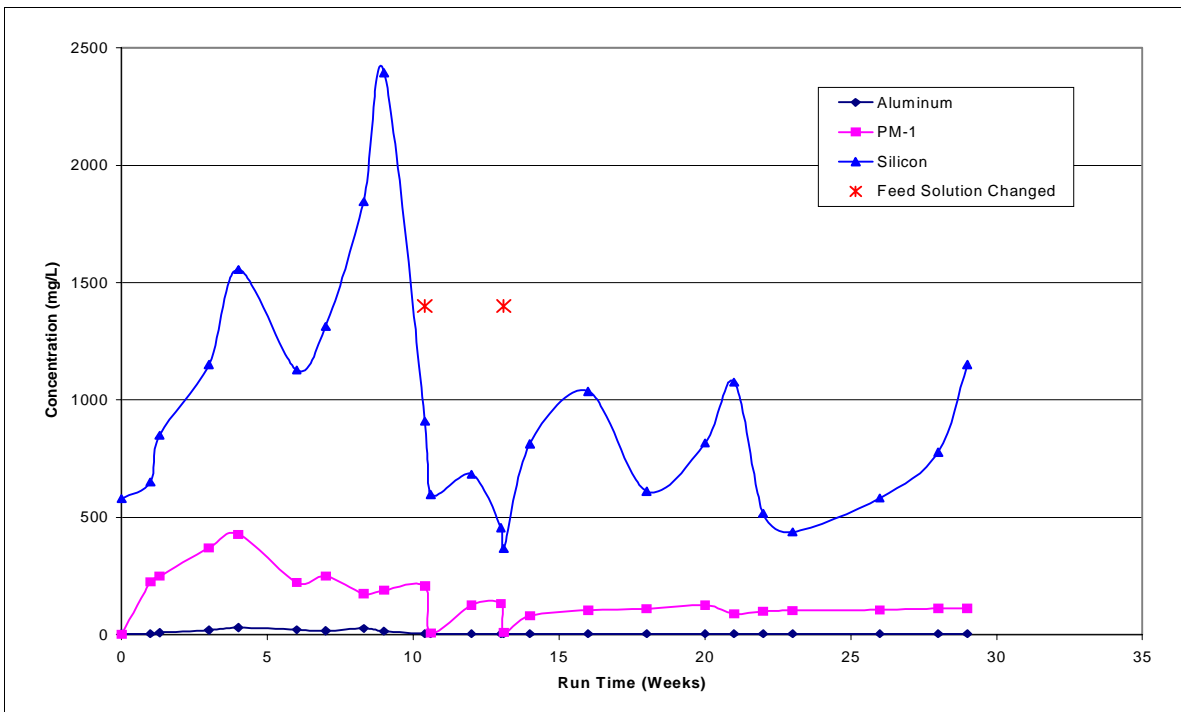


Fig. 12. Metal concentrations in the high-pH salt solution from the column leaching test.

Photomicrographs show that smaller CST particles and fragments tend to collect in the upper part of the columns. The first sample from the top of the column using average simulant, after one month, showed clumps of CST particles and fines cemented together (see Fig. 13).

Precipitates of sodium aluminosilicate have continued to slowly form on the inside wall of the silicon rubber tubing leading from the filter to the top of the average simulant column. Apparently the surface of the tubing acts as an initiation point for the precipitation. This section of tubing has been changed twice during the six months that the test has been running. There has been no indication of solids collecting in the filter, on the walls of the column or in the feed tank. The same precipitation process is probably causing the clumping of the CST particles in the column. Additional analyses will be performed in the future to identify the cause(s) of the clumping.

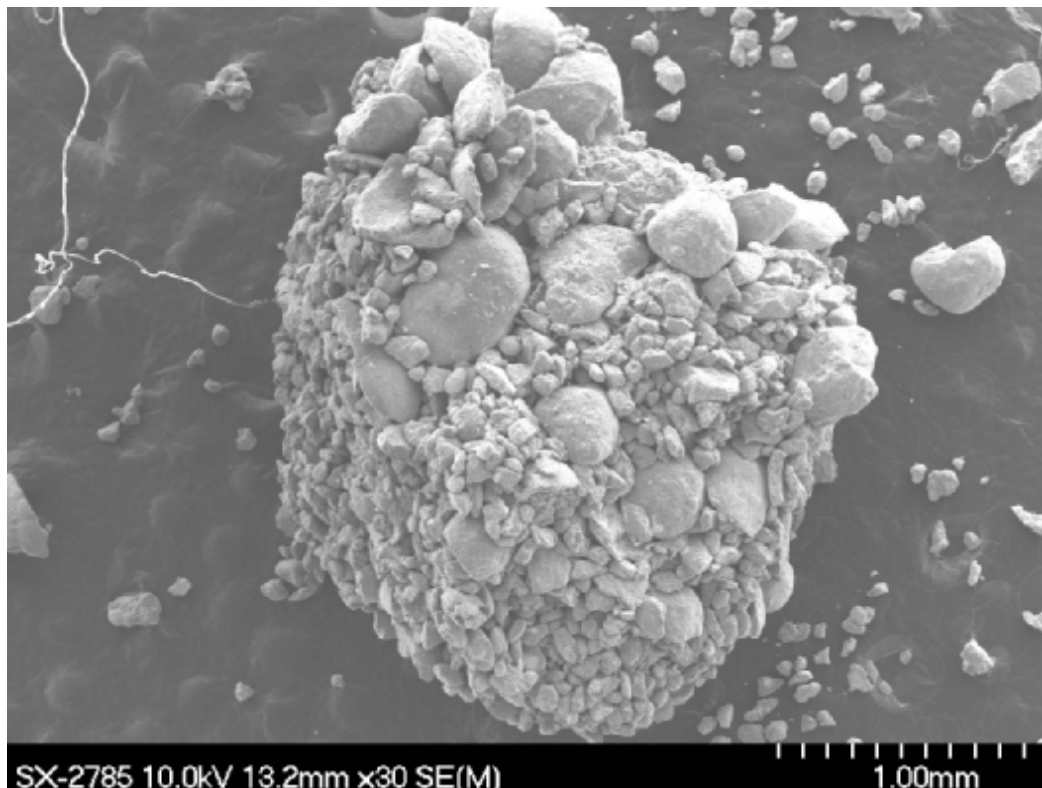


Fig. 13. SEM Photomicrograph of CST Clump That was Formed at the Top of the Column Using Average Simulant.

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Appendix 1. Results of Cesium Loading Tests at Various Temperatures.

Loading Temp. (°C)	Distribution Coefficient – Kd (L/kg) in each simulant solution					
	Average	High Hydroxide	High Nitrate	High-pH Salt Solution	Avg, 99-7	Avg, IE-910
24	1407	1674	1406	1128	1487	1962
30	1171	1485	1329	1038		
35	1090	1270	1071	893		
40	779	1059	841	672		
45	767	906	750	590		
50	533	706	561	438		
60	350	474	443	273		
70	195	381	243	170		
80	174	204	146	122	132	323

Loading Temp. (°C)	Cesium Loading (mg Cs/g CST) in each simulant solution					
	Average	High Hydroxide	High Nitrate	High-pH Salt Solution	Avg, 99-7	Avg, IE-910
24	27.2	29.4	27.2	24.4	28.0	31.6
30	25.0	27.9	26.4	23.0		
35	24.0	25.9	23.8	21.6		
40	19.8	23.6	20.8	18.2		
45	19.8	21.8	19.5	16.7		
50	15.6	18.7	16.2	13.6		
60	11.5	14.3	13.7	9.5		
70	7.1	12.2	8.6	6.3		
80	6.4	7.4	5.6	4.6	6.2	10.9

**Appendix 2. Cesium Distribution Coefficient and Loading Results
For CST Samples from the Column Leaching Tests.**

Time (Months)	Distribution Coefficient - Kd (L/kg)	
	Average Simulant	High-pH Salt Solution
0	1194	1217
1	1214	1220
2	1487	1565
3	1084	1221
4	1047	1204
5	1037	1279
6	921	1199
7	916	1242

Time (Months)	Cesium Loading (mg Cs/g CST)	
	Average Simulant	High-pH Salt Solution
0	25.1	25.6
1	23.9	24.0
2	28.2	28.9
3	24.2	25.7
4	23.9	26.2
5	23.9	26.4
6	22.4	25.7
7	22.3	26.1

Appendix 3. Distribution Coefficients for CST Samples from Batch Leaching Tests

Sample I.D.	Cesium Distribution Coefficient - K_d (L/kg)						
	1 Month	2 Months	3 Months	4 Months	5 Months	6 Months	7 months
AVG-25	1130	980	1305	1133	1219	1012	1048
AVG-30	989	880	1201	963	1006	834	804
AVG-35	935	924	1133	936	981	845	825
AVG-50	933	749	1041	818	878	773	744
AVG-80	807	650	985	757	744	679	732
HH-25	1139	982	1304	1192	1238	979	1117
HH-30	1089	880	1258	1005	1087	903	1057
HH-35	1005	958	1206	973	1009	870	993
HH-50	927	871	1055	833	865	774	863
HH-80	765	712	913	736	772	616	699
HN-25	1083	1077	1335	1048	1147	873	948
HN-30	1010	881	1133	902	972	742	865
HN-35	951	747	1081	855	920	772	842
HN-50	859	816	1063	830	906	720	804
HN-80	847	747	1025	714	783	701	775
HS-25	1072	1012	1470	1055	1449	1206	1257
HS-30	631		1462	1068	1266	1138	1106
HS-35	1023	951	1290	1049	1232	1021	1097
HS-50	973	860	1241	1008	1155	979	1000
HS-80	900	904	1125	979	1076	858	936
910-AVG-25	2295	2124	2497	2250	2514	2160	2122
910-AVG-80	2011	1403	1761	1361	1196	1014	899
99-7-AVG-25	1169	1011	1627	1288	1382	1282	1291
99-7-AVG-80	828	879	1142	958	977	809	832
1999-AVG-25	721	922	1027	863	889	762	739

Appendix 4. Cesium Loading Results for CST Samples from Batch Leaching Tests

Sample I.D.	Cesium Loading (mg Cs/g CST)						
	1 Month	2 Months	3 Months	4 Months	5 Months	6 Months	7 months
AVG-25	21.9	22.9	27.1	24.9	25.7	23.4	23.4
AVG-30	20.5	21.5	25.9	23.0	23.3	21.0	20.6
AVG-35	19.5	22.2	24.9	22.5	23.0	21.3	20.8
AVG-50	19.6	19.6	24.0	21.0	21.7	20.0	19.7
AVG-80	18.0	18.0	23.2	20.0	19.7	18.5	19.2
HH-25	21.9	23.0	26.9	25.5	25.9	22.5	24.7
HH-30	21.6	21.5	26.2	23.5	24.2	22.1	23.2
HH-35	20.8	22.5	25.7	23.0	23.4	21.6	22.4
HH-50	19.6	21.6	24.0	21.2	21.4	20.1	21.3
HH-80	17.6	19.0	22.3	19.7	20.1	17.5	18.6
HN-25	21.2	24.0	27.1	24.0	25.0	21.4	22.5
HN-30	20.9	21.5	25.2	22.0	23.0	19.5	21.2
HN-35	20.0	19.7	24.5	21.4	22.3	19.9	21.1
HN-50	18.8	20.7	24.3	21.1	22.0	19.3	20.4
HN-80	18.4	19.7	23.8	19.3	20.3	19.0	20.1
HS-25	21.5	23.3	28.5	24.0	28.0	25.0	26.1
HS-30	15.5		28.4	24.2	26.1	24.2	24.7
HS-35	21.0	22.7	26.8	24.0	25.9	23.6	24.6
HS-50	20.4	21.2	26.2	23.4	25.0	23.0	23.4
HS-80	19.7	22.1	25.0	23.2	24.2	21.3	21.7
910-AVG-25	29.0	32.5	47.7	33.3	34.1	32.6	33.1
910-AVG-80	28.0	27.2	30.4	27.2	25.6	23.8	21.6
99-7-AVG-25	22.5	23.5	29.7	26.5	27.4	26.4	26.3
99-7-AVG-80	18.7	21.6	25.3	22.8	22.9	20.4	21.1
1999-AVG-25	17.1	22.6	24.3	21.8	22.1	19.7	19.8