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Demonstration of Fuel and Fission Product Recovery from HTGR Reactor Fuel Processing Salt

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K. M. L. Taylor-Pashow
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EXECUTIVE SUMMARY

A molten salt digestion process is being examined to digest the graphite from High Temperature Gas-cooled Reactor (HTGR) fuel pebbles. Once the digestion is complete the salt will need to be processed to recover the fuel kernels, and then treated for final disposal. The proposed plan for (b)(3), (4) The solution will then be neutralized and treated with CST and MST to remove Cs, Sr, and actinides. A series of experiments were performed to examine the proposed process. Results showed successful removal of carbonate(b)(3), (4) with final carbonate concentrations measuring 2.10 – 2.26 mM. After pH adjustment to alkaline conditions, the fuel kernels were readily removed by filtration prior to the MST and CST treatments. The MST and CST treatment processes performed as expected. The Cs removal by CST was comparable to previous CST performance in SRS type high-level waste. While the performance was comparable, deployment of the CST in a column configuration (such as that planned for SRS waste in the Small Column Ion Exchange (SCIX) program) or a multi-strike batch process will likely be necessary to achieve the necessary Cs decontamination. The MST removal of Sr and actinides also compared well with previous data; however, there were some differences which could be explained by the differences in starting concentrations of the sorbates, most notably Sr. The Sr concentrations in these solutions were approximately 20 times the concentration in typical SRS high-level waste. If the actual waste does contain these levels of fission products and this waste is treated in an independent process (versus being introduced into the SRS high-level waste system), increased amounts of MST and/or CST may be necessary in order to meet the waste acceptance criteria.

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LIST OF ABBREVIATIONS

ARP	Actinide Removal Process
CST	crystalline silicotitanate
DDI	distilled deionized
DF	decontamination factors
DOE	Department of Energy
HTGR	High Temperature Gas-cooled Reactors
IC	ion chromatography
ICP-ES	inductively coupled plasma – emission spectroscopy
ICP-MS	inductively coupled plasma – mass spectroscopy
MST	monosodium titanate
PVDF	polyvinylidene fluoride
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
TIC/TOC	total inorganic carbon/total organic carbon
TTQAP	Task Technical and Quality Assurance Plan

1.0 Introduction

The United States Department of Energy (DOE) is considering accepting and processing used nuclear fuel from High Temperature Gas-cooled Reactors (HTGR) in Germany. The fuel is in the form of graphite spheres, called pebbles, each of which contains thousands of small kernels containing uranium and thorium oxides. SRNL is exploring technology for processing production-scale quantities of this reactor fuel involving a molten salt process to remove the graphite, which represents 97-98 wt % of the pebble.[1,2] One aspect of the test program involves the development of a process for recovering the fuel and fission products from the salt. It is expected that the fuel kernels will stay largely intact, but some could be cracked and could release fission products and/or actinides into the salt matrix. It is also known that some fission products will leach from the intact kernels into the salt.

The salt treatment method that was examined involves several steps, (b)(3), (4) [redacted] This was followed by neutralization, filtration to remove the kernels, and finally treatment of the filtrate with crystalline silicotitanate (CST) and monosodium titanate (MST) to remove Cs, Sr, and any soluble actinides. The experimental plan was outlined in a memo[3] prior to starting work, and the work was controlled by a Task Technical and Quality Assurance Plan (TTQAP).[4]

2.0 Experimental Procedure

2.1 Preparation of the Salt Samples

Five salt samples were prepared by adding approximately 50 g of salt generated from a previous HTGR unirradiated fuel pebble digestion demonstration to each of five crucibles.[2] The salt was from the PEB 5 Test of the earlier study. To the crucible were added $CsNO_3$, $Sr(NO_3)_2$ and $NaReO_4$ to represent expected amounts of the fission products Cs, Sr, and Re (Tc surrogate). Unirradiated fuel kernels were also added to each crucible, and sodium hydroxide was added to 2 of the 5 samples. The crucibles were then heated to 600 °C, stirred with a quartz rod, held at temperature for one hour, stirred again, and cooled to room temperature. The composition of each sample is shown in Table 2-1. (b)(3), (4) [redacted]

Table 2-1. Composition of Prepared Salt Samples

(b)(3), (4) [redacted]	(b)(3), (4) [redacted]	(b)(3), (4) [redacted]	(b)(3), (4) [redacted]	(b)(3), (4) [redacted]	(b)(3), (4) [redacted]	(b)(3), (4) [redacted]
[redacted]	[redacted]	[redacted]	[redacted]	[redacted]	[redacted]	(b)(3), (4) [redacted]
(b)(3), (4) [redacted]	[redacted]	[redacted]	[redacted]	[redacted]	[redacted]	[redacted]
[redacted]	[redacted]	[redacted]	[redacted]	[redacted]	[redacted]	[redacted]
[redacted]	[redacted]	[redacted]	[redacted]	[redacted]	[redacted]	[redacted]
[redacted]	[redacted]	[redacted]	[redacted]	[redacted]	[redacted]	[redacted]
[redacted]	[redacted]	[redacted]	[redacted]	[redacted]	[redacted]	[redacted]

2.2 Dissolution of Salt and Recovery of Fuel Kernels

Salt samples 1 and 4 were selected for performing the initial experiments. Each of the samples was dissolved in 200 mL of distilled water. (b)(3), (4) [redacted] A stoichiometric amount of acid was added to react with the Na_2CO_3 along with enough excess to bring the final acid concentration (b)(3), (4) [redacted] After the acid addition was complete, the acidic solution was spiked with $^{239/240}Pu$, ^{237}Np, ^{137}Cs, and Sr. The Cs and Sr isotopes were both targeted to a spiking level of ~30,000 dpm/mL, while the Pu was targeted at 0.5 mg per sample and the Np at 1 mg per sample. A sample was removed at this stage to confirm radioisotope spiking levels.

After spiking with radionuclides sodium hydroxide was added to neutralize the solution, and excess sodium hydroxide was added to bring the final free hydroxide concentration to approximately 2 M. (b)(3), (4) [redacted] Finally, after the sodium hydroxide addition, the mixture was filtered through a 0.45-µm Nylon filter to remove the fuel kernels along with any solids that precipitated during the neutralization.

2.3 Batch Contact Testing

A series of batch contact tests were performed using the dissolved, pH adjusted, and filtered salt solutions described in the previous section. Three 50-mL aliquots of each of the Salt #1 and Salt #4 solutions were transferred to 60-mL polyethylene bottles. One bottle from each set served as a control, while MST and CST were added to the second and third bottles, respectively. The MST was added at a concentration of 0.2 g/L, which is the concentration currently being used at the Actinide Removal Process (ARP) for Sr and actinide removal from SRS high level waste. The CST was added at a concentration of 10 g/L. This concentration was selected to obtain measurable Cs decontamination factors (DF) under the experimental conditions. CST would normally be deployed in a column configuration, but for simplicity of the experiments, batch contact testing was performed to measure the CST performance in this matrix.

After adding the MST and CST, the bottles were shaken in a 25 °C shaker oven at 175 rpm for one week. Samples were removed after 1, 6, 24, and 168 hours of contact. At each sampling event, an aliquot was removed and filtered through a 0.1-µm PVDF syringe filter. The filtrate was then acidified with an equal volume of 5 M nitric acid prior to analysis.

The MST used in these experiments was prepared by Harrell Industries (Lot # 46000619120). The CST (IE-911) used in these experiments was produced by Universal Oil Products (UOP), Lot# 208000056.

2.4 Quality Assurance

Requirements for performing reviews of technical reports and the extent of review are established in manual E7 2.60. SRNL documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2.

3.0 Results and Discussion

3.1 Dissolution and Acid Adjustments Salt Samples

The salts were not easily removed from the crucibles after heating and subsequent cooling. For salt sample #1, an aliquot of ultrapure water from a MilliQ Element purification system was added to the top of the crucible, and after sitting for several minutes the water and any dissolved salt was transferred to the bottle. This was repeated twice, with the second time being allowed to sit for ~1.5 hours. At this point, enough salt had dissolved that the remaining salt monolith had loosened from the crucible. The remaining salt was then transferred to the bottle and the crucible rinsed several times to ensure all remaining salt was transferred. The remaining DDI water needed to bring the total to 200 mL was then added to the bottle, and the bottle was gently stirred to dissolve the remaining salt. After 1 hour of stirring the salt appeared completely dissolved.

A similar approach was taken with salt sample #4, however, this salt sample proved more difficult to remove from the crucible. After several soaking attempts in the crucible, the salt monolith could not be loosened from the crucible. In contrast to salt sample #1, which dissolved enough from the outside to loosen from the crucible, salt sample #4 appeared to be preferentially dissolving from the center. Therefore, the entire crucible was submerged in the 200-mL of dissolving DDI water. After soaking for ~3 hours, the salt still could not be loosened from the crucible, and was left to soak overnight. After soaking overnight, much of the salt had dissolved, but there was still some remaining in the crucible that could not be removed, so the soaking was continued. The soaking was continued over a weekend, and on the 4th day (since the start of trying to dissolve) all of the salt had finally dissolved from the crucible.

After dissolving the salt samples in DDI water, concentrated nitric acid was slowly added to each solution over a period of 20-25 minutes. (b)(3), (4). No significant bubbling of the solution was observed until approximately two-thirds to three-fourths of the acid had been added, at which point bubbling was seen throughout the solution (See Figure 3-1). The temperature was monitored throughout the acid addition, and did not exceed 29 °C.

Figure 3-1. Bubbling observed during later portions of acid addition to Salt Sample #1 Solution.

(b)(3), (4)

An additional aliquot of acid was added to each bottle, representing the calculated amount required to reduce the pH from 4 to 1 (increase acid concentration from 0.0001 M to 0.1 M). After this addition the pH was still measuring ~4 for both samples. An additional 3 mL of concentrated nitric acid was then added to salt sample #1, 0.2 mL at a time, while monitoring the pH. The pH did not drop significantly, and was still measuring ~3-4 with pH strips. At this point both solutions were transferred to another rad hood containing a digital pH meter, for more accurate pH measurements.

After transferring the solutions, the pH of salt sample #1 solution was measured to be 3.43. Additional concentrated nitric acid was then added 0.2 mL at a time, while monitoring the pH. An additional 2.8 mL was added to reduce the pH to 1.02. (b)(3), (4) (b)(3), (4) [redacted] After standing for approximately 1 hour (while salt sample #4 was being adjusted), the pH was measured again, and had dropped to 0.94. After stirring for several hours, the final pH was measured to be 0.82. This corresponds to an acid concentration of 0.151 M. A total of 36.65 mL of concentrated nitric acid was added to this sample, which represents a 25% increase in the calculated amount (29.4 mL) of nitric acid needed to adjust the salt solution to a pH of 1.(b)(3), (4) [redacted]

A similar process was performed for salt sample #4 solution.(b)(3), (4) [redacted]

3.2 Radionuclide Spiking and Caustic Adjustment of Salt Solutions

After the acid adjustments were complete, the two solutions were spiked with ^{85}Sr, ^{137}Cs, ^{237}Np, and $^{239/240}$Pu. The target levels and amounts added are shown in Table 3-1. After stirring overnight, a sample was removed and filtered from each bottle and submitted for gamma scan and ICP-MS analyses to determine radionuclide activities. The results from those analyses are shown in Tables 3-2 and 3-3.

Table 3-1. Amounts of Radionuclides Needed for Spiking Salt Solutions

Isotope	Activity or Concentration of Stock	Target Activity[a]	Volume Needed
^{85}Sr	1.28 E+10 dpm/mL	3.0 E+04 dpm/mL	0.6 µL
^{137}Cs	3.31 E+08 dpm/mL	3.0 E+04 dpm/mL	24 µL
^{237}Np	1.1 g/L	1 mg total	909 µL
$^{239/240}$Pu	1.47 g/L	0.5 mg total	341 µL

a) Target activity in final solution (after NaOH addition).

Table 3-2. Measured Radionuclide Concentrations in Salt #1 Solution

Isotope	Measured Activity/Concentration	Calculated Activity/Concentration after NaOH addition	% of Target[a]
^{85}Sr	3.23 E+04 dpm/mL	2.90 E+04 dpm/mL	96.7%
^{137}Cs	9.13 E+04 dpm/mL	8.20 E+04 dpm/mL	273%
^{237}Np	4.51 E+03 µg/L	4.05 E+03 µg/L	107%
$^{239/240}$Pu	2.22 E+03 µg/L	2.00 E+03 µg/L	105%
^{232}Th	2.08 E+02 µg/L	1.87 E+02 µg/L	n/a[b]
^{238}U	2.82 E+03 µg/L	2.53 E+03 µg/L	n/a[b]

a) After correction for dilution with NaOH to reach final solution.
b) Not spiked, amount present due to dissolution of the kernels.

Table 3-3. Measured Radionuclide Concentrations in Salt #4 Solution

Isotope	Measured Activity/Concentration	Calculated Activity/Concentration after NaOH addition	% of Target[a]
^{85}Sr	3.18 E+04 dpm/mL	2.85 E+04 dpm/mL	95.1%
^{137}Cs	8.18 E+04 dpm/mL	7.34 E+04 dpm/mL	245%
^{237}Np	4.42 E+03 µg/L	4.20 E+03 µg/L	113%
$^{239/240}$Pu	2.18 E+03 µg/L	1.95 E+03 µg/L	106%
^{232}Th	1.35 E+02 µg/L	1.21 E+02 µg/L	n/a[b]
^{238}U	1.24 E+03 µg/L	1.11 E+03 µg/L	n/a[b]

a) After correction for dilution with NaOH to reach final solution.
b) Not spiked, amount present due to dissolution of the kernels.

(b)(3), (4) [redacted] The results from the chemical analyses are shown in Table 3-6. These filtered solutions were then used for batch contact testing with MST and CST.

(b)(3), (4) [redacted] The final column indicates the percent of the isotope or element in solution compared to what was calculated, assuming it all remained soluble. The uranium appeared to remain completely soluble, whereas, the thorium precipitated to below detection limit concentrations. Previous work has shown Th has an extremely low solubility in alkaline salt solutions.[5] In the Salt #1 solution, approximately 70% of the Np and Pu precipitated; a similar amount of Pu precipitated from the Salt #4, whereas the Np remained slightly more soluble with only about 60% precipitating. The fission products (Cs, Sr, and Re) remained soluble.

Table 3-4. Measured versus calculated concentrations in final solution from Salt #1.

Isotope	Measured Activity/Concentration	Calculated Activity/Concentration	% of Calculated
^{85}Sr	2.48 E+04 dpm/mL	2.90 E+04 dpm/mL	85.4%
^{137}Cs	8.32 E+04 dpm/mL	8.20 E+04 dpm/mL	101%
Total Re	9.50 E+04 µg/L	9.27 E+04 µg/L	102%
^{237}Np	1.19 E+03 µg/L	4.05 E+03 µg/L	29.5%
^{239}Pu	5.72 E+02 µg/L	1.88 E+03 µg/L	30.5%
^{232}Th	< 1.00 E+01 µg/L	1.87 E+02 µg/L	< 5.34%
^{238}U	2.54 E+03 µg/L	2.53 E+03 µg/L	100%

Table 3-5. Measured versus calculated concentrations in final solution from Salt #4.

Isotope	Measured Activity/Concentration	Calculated Activity/Concentration	% of Calculated
^{85}Sr	2.66 E+04 dpm/mL	2.85 E+04 dpm/mL	93.4%
^{137}Cs	7.62 E+04 dpm/mL	7.34 E+04 dpm/mL	104%
Total Re	1.01 E+05 µg/L	1.01 E+05 µg/L	101%
^{237}Np	1.62 E+03 µg/L	4.20 E+03 µg/L	38.6%
^{239}Pu	5.02 E+02 µg/L	1.95 E+03 µg/L	27.3%
^{232}Th	< 1.00 E+01 µg/L	1.21 E+02 µg/L	< 8.25%
^{238}U	1.11 E+03 µg/L	1.11 E+03 µg/L	100%

Table 3-6. Chemical compositions of final salt solutions.

(b)(3), (4) [redacted]

(b)(3), (4) [redacted]

(b)(3), (4) [redacted]

3.3 Batch Contact Testing with MST and CST

Results from the batch contact tests with MST and CST are summarized in Tables 3-7 through 3-8 and Figures 3-2 through 3-6.

The Cs was successfully removed by CST, but not MST, which is consistent with expectations. MST has been previously shown to have a low affinity for Cs in strongly alkaline solutions. The Cs DFs were fairly consistent in the two different salt samples, reaching a final DF of 16.2 in Salt #1 solution and 14.1 in Salt #4 solution, after 1 week of contact with 10 g/L CST. These DFs are consistent with previous batch testing performed at the same phase ratio.[6] Although the DFs measured were consistent with previous batch contact testing, column deployment of the CST or a multi-strike batch contact process is likely necessary to achieve the required Cs decontamination.

In contrast to Cs, Sr was removed by both MST and CST. At the early time points, Sr was more efficiently removed by MST; however, after 1 week of contact, CST had significantly higher DFs. The difference in particle size between the two materials likely contributed to the kinetic affects, while the higher sorbent concentration of CST (10 g/L) compared to MST (0.2 g/L) led to greater total removal. The Sr DF obtained with MST was somewhat lower than what is typically obtained for SRS high level waste.[7] This difference is likely due to the increased Sr concentration in these salt solutions compared to the Sr concentration in the typical SRS HLW simulant. The Sr concentration in these solutions is approximately 20 times that in the SRS HLW simulant used for testing MST in the 2005 experiments.[7] If the actual waste does contain these levels of Sr and this waste is treated in an independent process (versus being introduced into the SRS high-level waste system), increased amounts of MST may be necessary in order to meet the waste acceptance criteria.

Trends similar to what was observed for Sr were also seen for the actinides, where there was removal by both MST and CST. At the early sampling times the MST generally had higher DFs than CST, but after 1 week of contact the CST removed greater quantities of the actinides. Again this is likely due to the increased sorbent loading in the CST experiments, which can be seen by comparing the distribution constants (K_d) which are shown in Table 3-8 and Figure 3-7. Unlike the DFs the K_d take into consideration the phase ratio used and provide a metric for evaluating the capacity of a material for a particular sorbate when tested at significantly different mass concentrations as in the case of the current testing. The K_d values for Pu, Np, and U removal by MST are much larger than those measured with CST. Previous work has shown that CST has little affinity for the actinides, when added at a similar concentration to MST.[8] The small amount of uptake seen in the earlier work was only observed for the bead form of the CST, not the powdered form, and was attributed to the binder used to prepare the bead of CST.[8] The DFs obtained with MST were fairly consistent with previous experiments.[7] The Pu and Np concentrations in these experiments were approximately 2-3 times greater than those in the 2005 experiments, and the MST was added at half the concentration, which resulted in slightly lower DFs in these experiments.

Table 3-7. Summary of Decontamination Factors (DFs) obtained with MST and CST. Values in parentheses are one sigma uncertainty.

^{137}Cs DF				
Time	**MST – Salt #1**	**MST – Salt #4**	**CST – Salt #1**	**CST – Salt #4**
1 h	1.03 (0.07)	1.01 (0.07)	1.23 (0.09)	1.12 (0.08)
6 h	1.00 (0.07)	0.99 (0.07)	1.70 (0.12)	1.59 (0.11)
24 h	0.96 (0.07)	0.97 (0.07)	5.57 (0.39)	6.68 (0.47)
168 h	1.00 (0.07)	0.94 (0.07)	16.2 (1.14)	14.1 (1.00)
^{85}Sr DF				
Time	**MST – Salt #1**	**MST – Salt #4**	**CST – Salt #1**	**CST – Salt #4**
1 h	2.76 (0.19)	2.38 (0.17)	1.20 (0.08)	1.14 (0.08)
6 h	3.16 (0.22)	2.81 (0.20)	1.61 (0.11)	1.55 (0.11)
24 h	3.53 (0.25)	3.11 (0.22)	7.35 (0.52)	8.79 (0.62)
168 h	4.47 (0.32)	3.60 (0.25)	418 (99.7)	160 (12.8)
168 h (Sr by ICP-MS)	4.36 (1.23)	3.62 (1.02)	164 (46.5)	126 (35.7)
Pu DF				
Time	**MST – Salt #1**	**MST – Salt #4**	**CST – Salt #1**	**CST – Salt #4**
1 h	2.66 (0.75)	2.71 (0.77)	1.13 (0.32)	1.09 (0.31)
6 h	4.34 (1.23)	4.35 (1.23)	1.32 (0.37)	1.28 (0.36)
24 h	6.97 (1.97)	6.56 (1.86)	2.85 (0.81)	2.97 (0.84)
168 h	11.8 (3.35)	10.9 (3.07)	22.5 (6.36)	14.0 (3.95)
Np DF				
Time	**MST – Salt #1**	**MST – Salt #4**	**CST – Salt #1**	**CST – Salt #4**
1 h	2.79 (0.79)	2.24 (0.63)	1.10 (0.31)	1.06 (0.30)
6 h	3.90 (1.10)	2.95 (0.83)	1.23 (0.35)	1.15 (0.32)
24 h	4.47 (1.26)	3.50 (0.99)	1.94 (0.55)	1.74 (0.49)
168 h	4.23 (1.20)	3.66 (1.04)	5.40 (1.53)	3.59 (1.02)
U DF				
Time	**MST – Salt #1**	**MST – Salt #4**	**CST – Salt #1**	**CST – Salt #4**
1 h	1.09 (0.31)	1.11 (0.31)	1.07 (0.30)	1.06 (0.30)
6 h	1.16 (0.33)	1.13 (0.32)	1.14 (0.32)	1.10 (0.31)
24 h	1.19 (0.34)	1.26 (0.36)	1.40 (0.40)	1.43 (0.41)
168 h	1.33 (0.38)	1.35 (0.38)	2.05 (0.58)	1.82 (0.51)

Table 3-8. Summary of Distribution Constants (K_d) obtained with MST and CST.

^{137}Cs K_d (mL/g)				
Time	**MST – Salt #1**	**MST – Salt #4**	**CST – Salt #1**	**CST – Salt #4**
1 h	1.47 E+02	2.62 E+01	2.27 E+01	1.20 E+01
6 h	2.41 E+02	n/a	7.01 E+01	5.86 E+01
24 h	n/a	n/a	4.56 E+02	5.67 E+02
168 h	1.24 E+01	n/a	1.51 E+03	1.30 E+03
^{85}Sr K_d (mL/g)				
Time	**MST – Salt #1**	**MST – Salt #4**	**CST – Salt #1**	**CST – Salt #4**
1 h	8.69 E+03	6.83 E+03	1.98 E+01	1.35 E+01
6 h	1.07 E+04	9.00 E+03	6.09 E+01	5.48 E+01
24 h	1.25 E+04	1.04 E+04	6.34 E+02	7.76 E+02
168 h	1.71 E+04	1.29 E+04	4.17 E+04	1.58 E+04
168 h (Sr by ICP-MS)	1.66 E+04	1.30 E+04	1.63 E+04	1.25 E+04
Pu K_d (mL/g)				
Time	**MST – Salt #1**	**MST – Salt #4**	**CST – Salt #1**	**CST – Salt #4**
1 h	8.23 E+03	8.48 E+03	1.30 E+01	9.33 E+00
6 h	1.65 E+04	1.66 E+04	3.15 E+01	2.76 E+01
24 h	2.95 E+04	2.76 E+04	1.85 E+02	1.97 E+02
168 h	5.36 E+04	4.89 E+04	2.15 E+03	1.29 E+03
Np K_d (mL/g)				
Time	**MST – Salt #1**	**MST – Salt #4**	**CST – Salt #1**	**CST – Salt #4**
1 h	8.84 E+03	6.14 E+03	9.93 E+00	6.15 E+00
6 h	1.44 E+04	9.66 E+03	2.33 E+01	1.48 E+01
24 h	1.71 E+04	1.24 E+04	9.35 E+01	7.38 E+01
168 h	1.60 E+04	1.32 E+04	4.41 E+02	2.58 E+02
U K_d (mL/g)				
Time	**MST – Salt #1**	**MST – Salt #4**	**CST – Salt #1**	**CST – Salt #4**
1 h	4.51 E+02	5.39 E+02	6.78 E+00	5.97 E+00
6 h	8.02 E+02	6.65 E+02	1.38 E+01	1.01 E+01
24 h	9.32 E+02	1.29 E+03	4.01 E+01	4.33 E+01
168 h	1.62 E+03	1.75 E+03	1.04 E+02	8.13 E+01

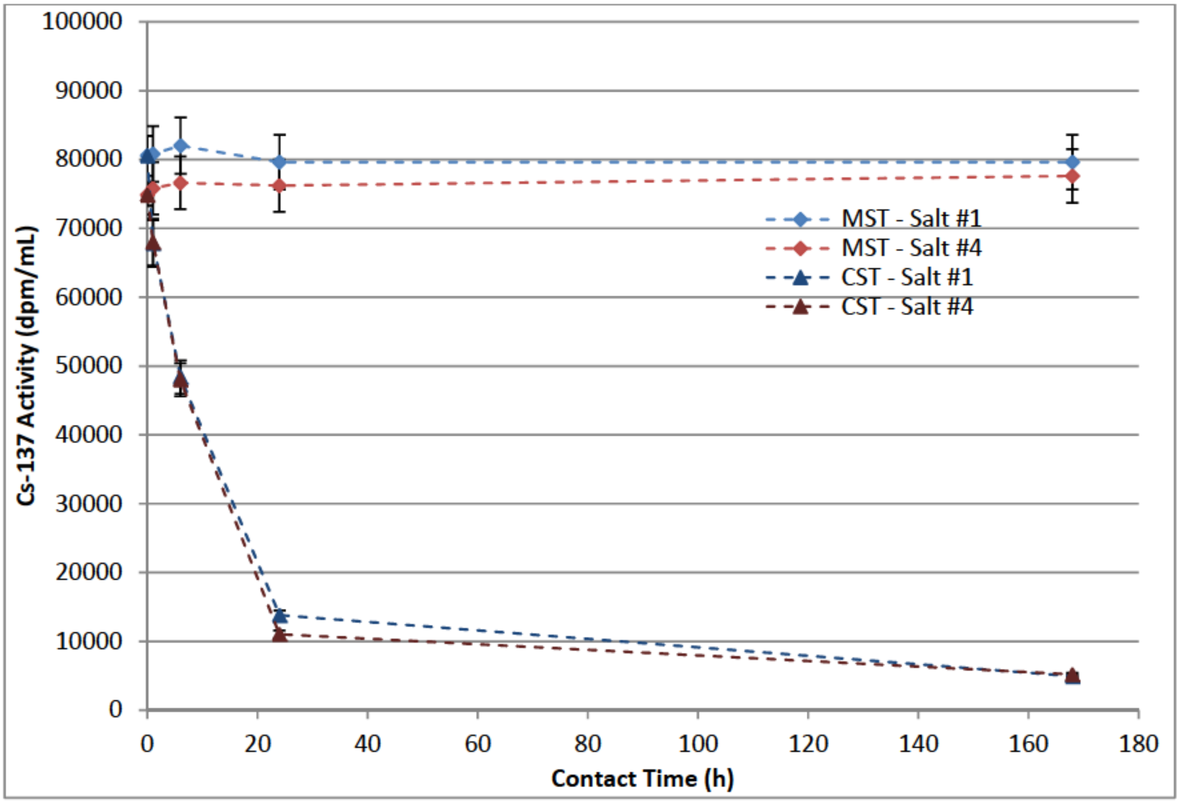

Figure 3-2. ^{137}**Cs activity versus contact time.**

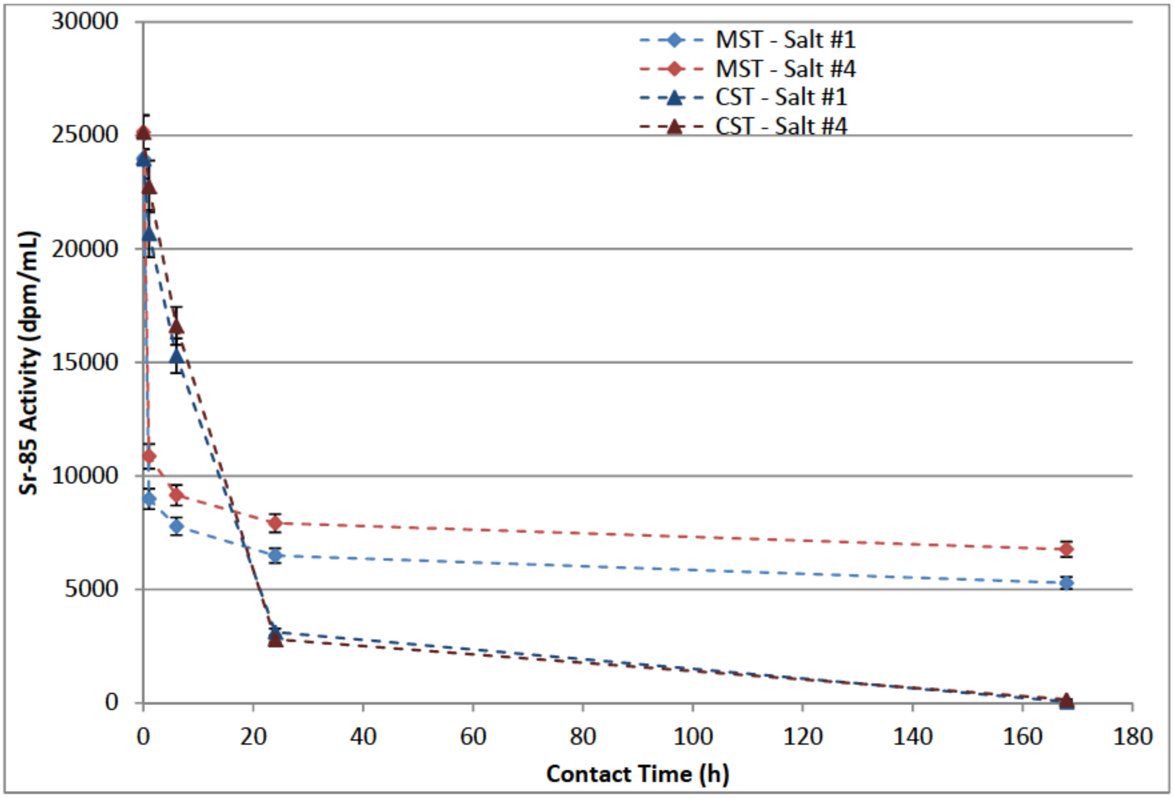

Figure 3-3. ^{85}**Sr activity versus contact time.**

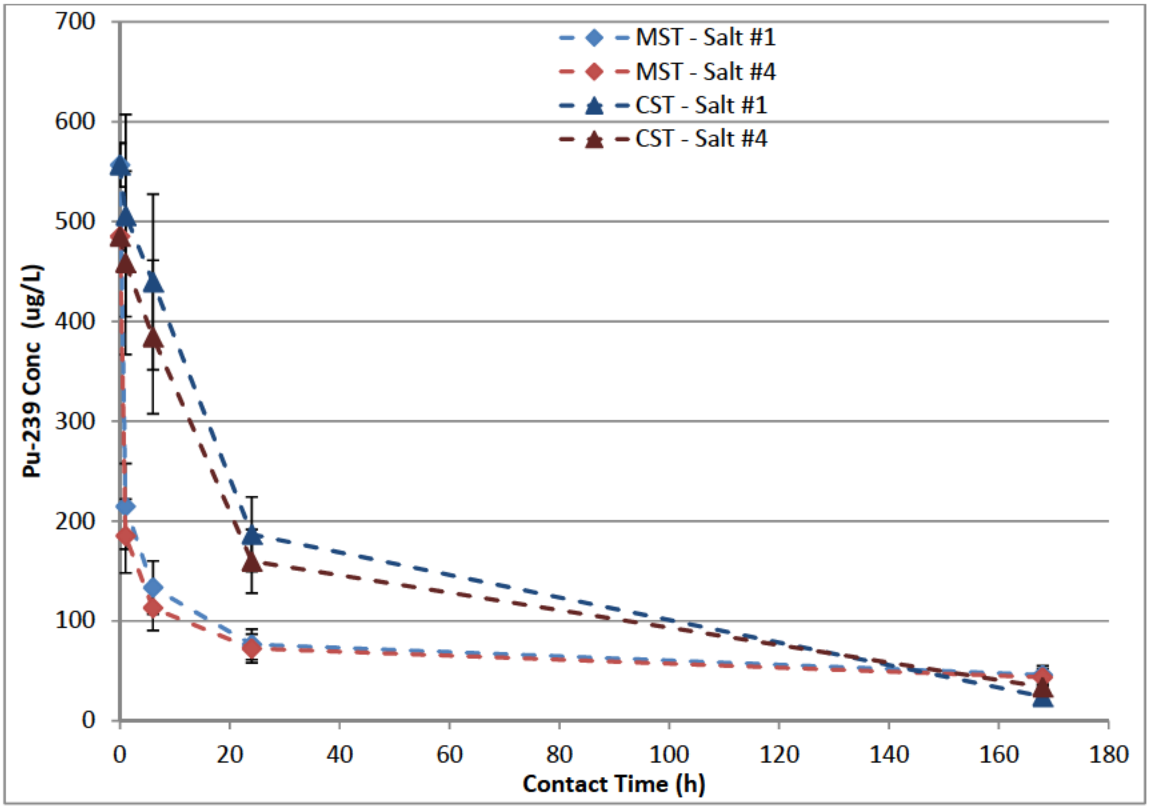

Figure 3-4. ^{239}**Pu concentration versus contact time.**

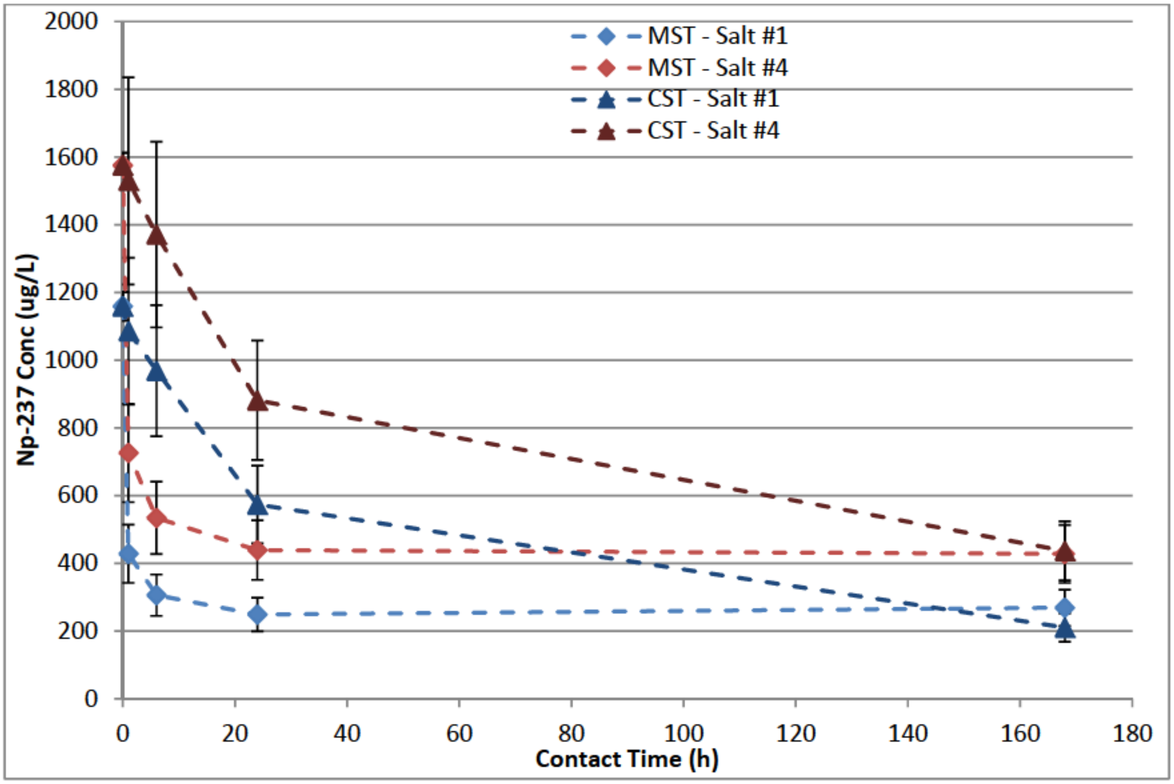

Figure 3-5. ^{237}**Np concentration versus contact time.**

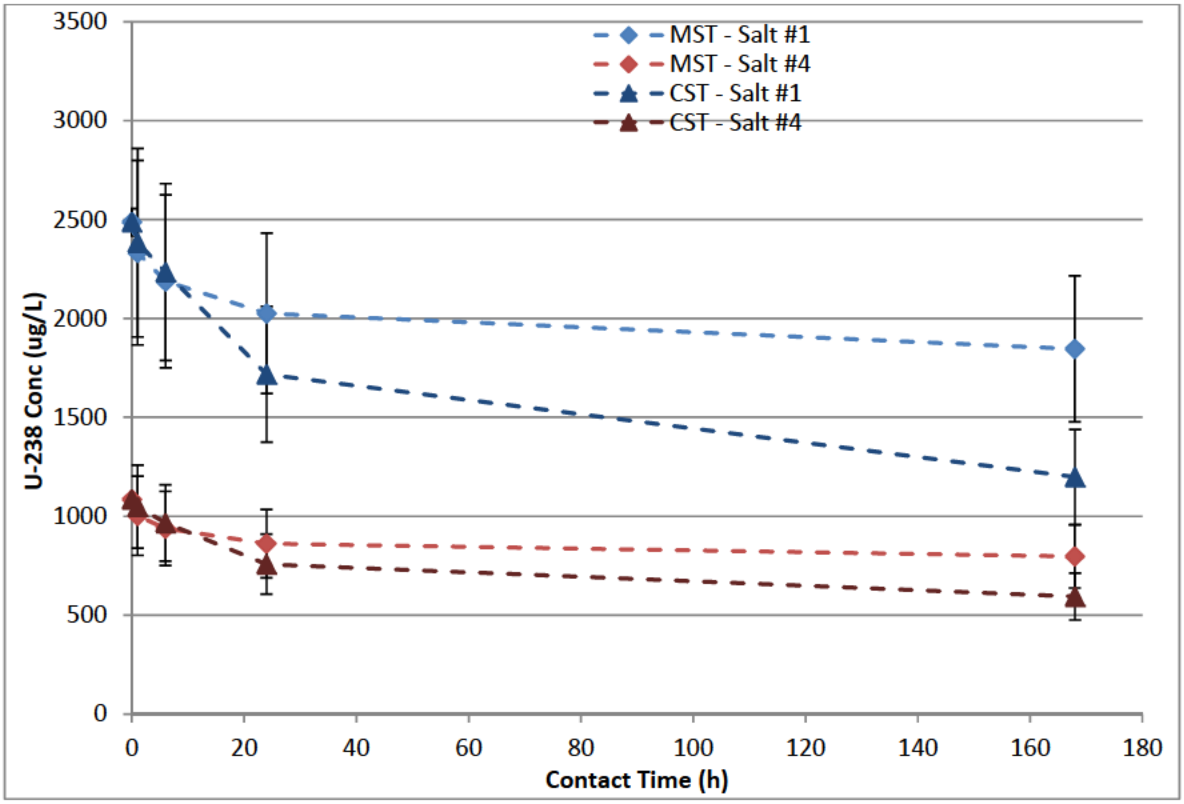

Figure 3-6. ^{238}U concentration versus contact time.

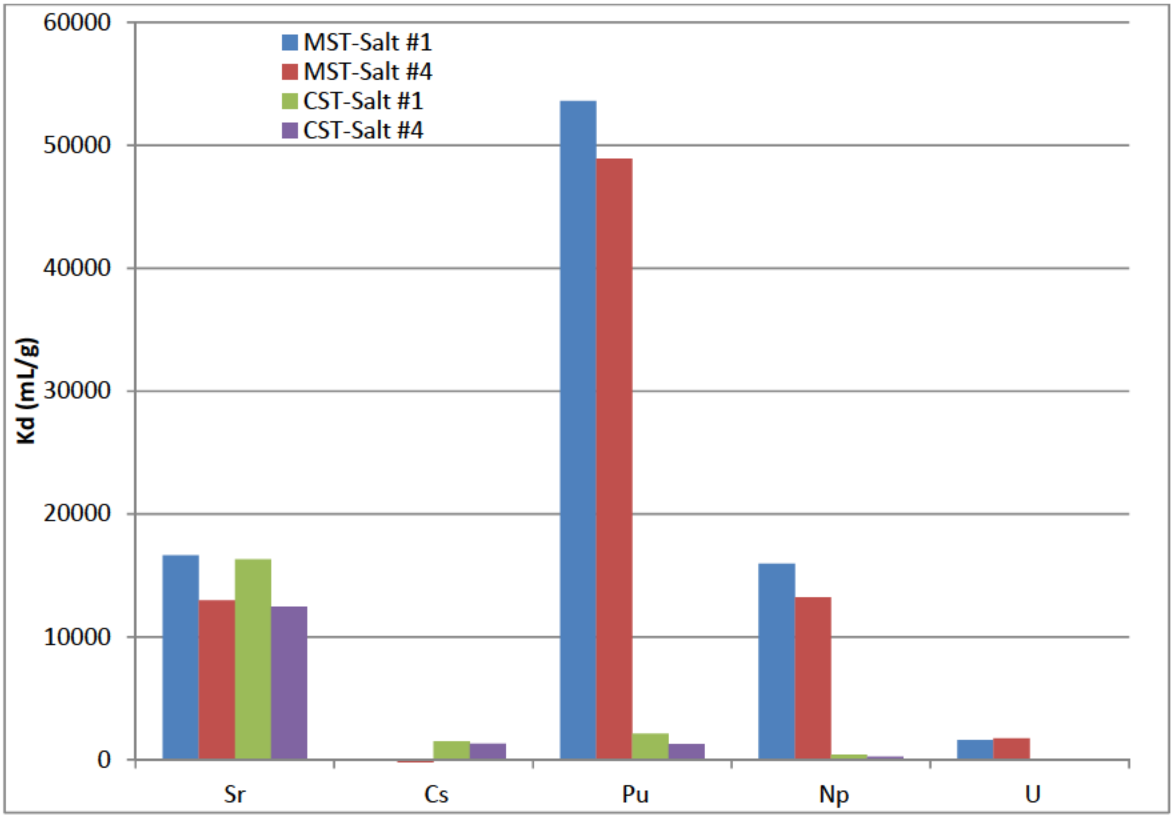

Figure 3-7. Summary of 168-h K_d values.

4.0 Conclusions

While the salt samples were slow to dissolve in water, complete dissolution of the salt monoliths was achieved. (b)(3), (4) The fuel kernels and precipitated actinides were then recovered by filtration.

Tests indicated that the filtrate can be treated with either MST or CST for the removal of Cs and Sr fission products and the remaining soluble actinides (U, Np, Pu). The effectiveness of CST and MST was consistent with previous simulant testing. The MST decontamination factors were somewhat lower than previous work with typical SRS high-level waste simulants, which is likely due to the higher concentrations of Sr and actinides in the salt solutions in the present tests. If the waste is treated in an independent process (versus being introduced into the SRS high-level waste system), increased amounts of MST and/or CST may be necessary in order to meet the waste acceptance criteria. The CST will also likely need to be deployed in either a column configuration or a multi-strike batch contact process in order to achieve the necessary decontamination using reasonable amounts of CST.

5.0 References

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Appendix A. Additional MST/CST Sorption Data

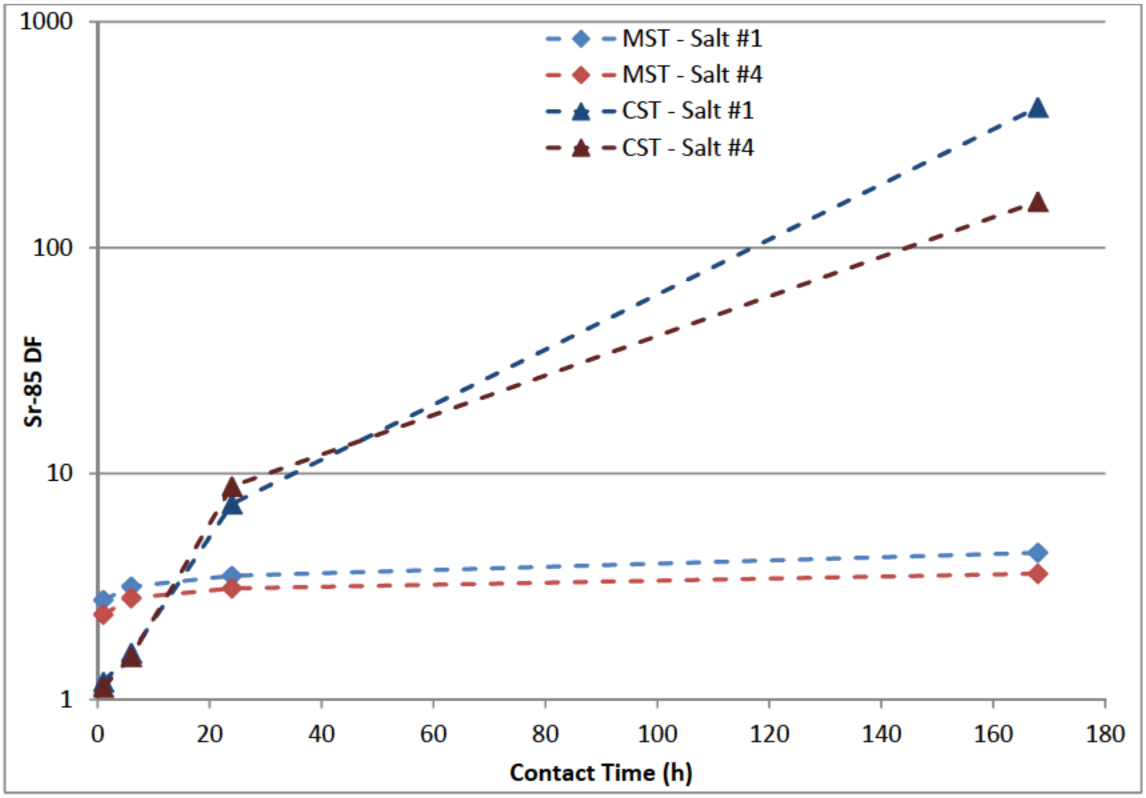

Figure A-1. ^{85}Sr DF versus contact time.

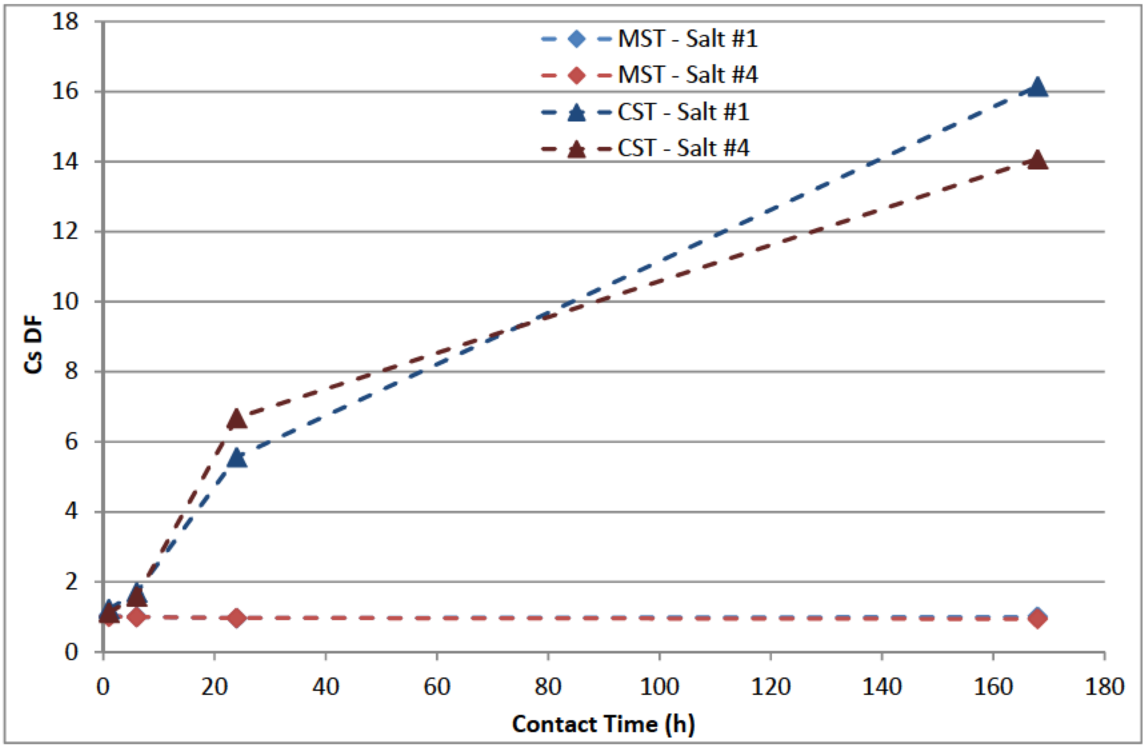

Figure A-2. ^{137}Cs DF versus contact time.

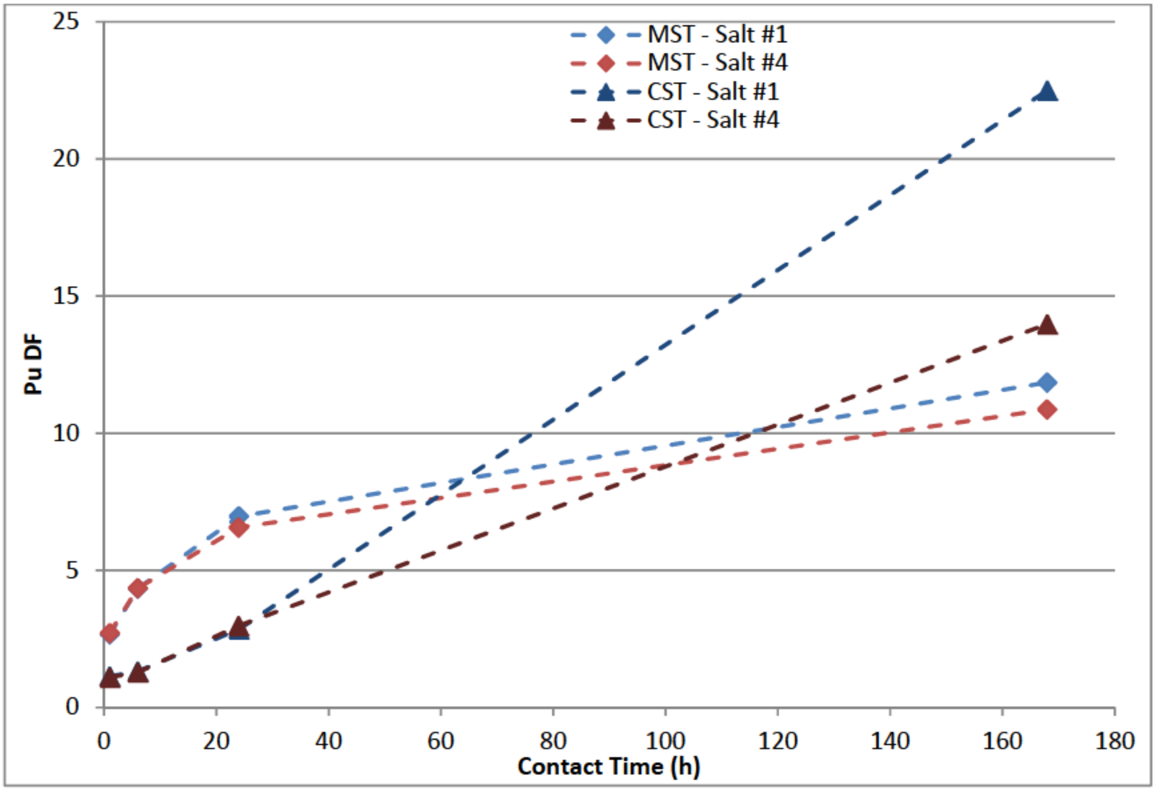

Figure A-3. Pu DF versus contact time.

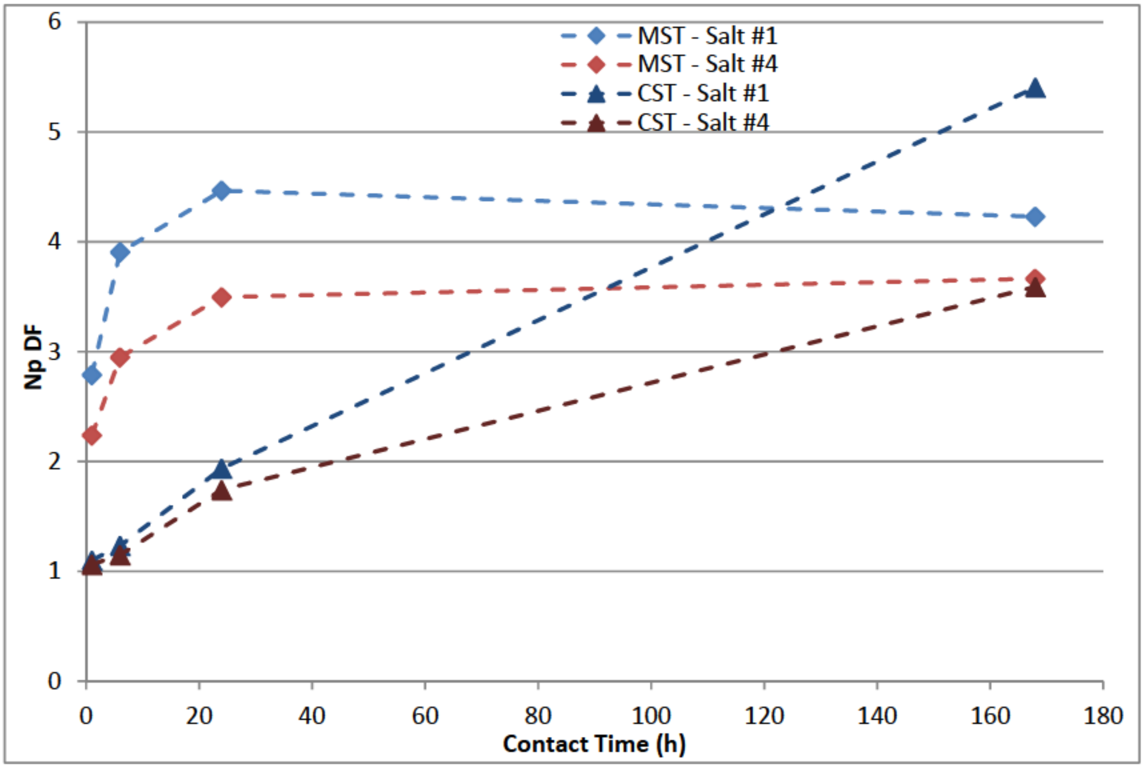

Figure A-4. Np DF versus contact time.

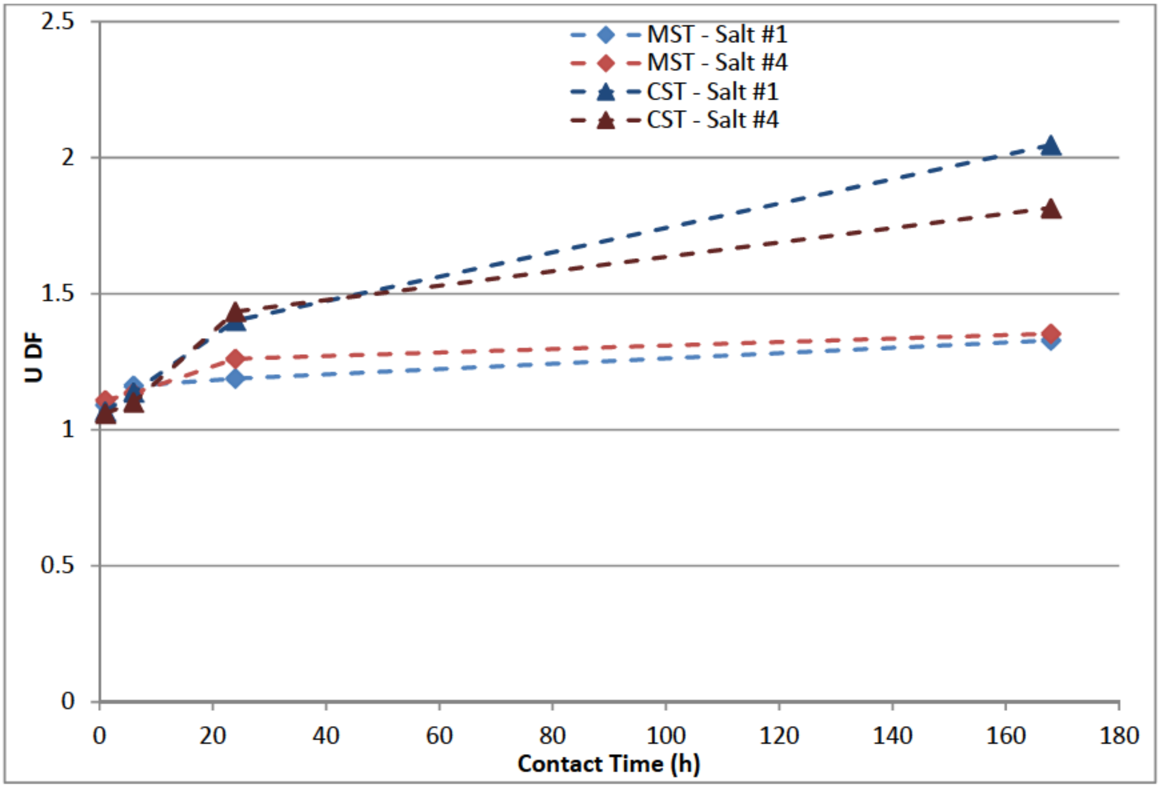

Figure A-5. U DF versus contact time.

Table A-1. Summary of Re Decontamination Factors Obtained with MST and CST.

Time	MST – Salt #1	MST – Salt #4	CST – Salt #1	CST – Salt #4
1 h	1.01 (0.28)	1.01 (0.29)	1.01 (0.28)	1.01 (0.29)
6 h	1.01 (0.29)	1.02 (0.29)	1.01 (0.28)	1.01 (0.29)
24 h	0.97 (0.28)	1.00 (0.28)	0.96 (0.27)	1.00 (0.28)
168 h	1.01 (0.29)	1.01 (0.29)	1.01 (0.29)	1.01 (0.28)

Distribution:

S. L. Marra, 773-A
T. B. Brown, 773-A
D. H. McGuire, 999-W
S. D. Fink, 773-A
C. C. Herman, 773-A
E. N. Hoffman, 999-W
F. M. Pennebaker, 773-42A
W. R. Wilmarth, 773-A

Records Administration (EDWS)

K. M. L. Taylor-Pashow, 773-A
D. T. Hobbs, 773-A
C. A. Nash, 773-42A
W. F. Bates, 703-H
K. M. Fox, 999-W
R. A. Pierce, 773-A