Phase IV Simulant Testing of Monosodium Titanate Adsorption Kinetics

by
D. T. Hobbs
Westinghouse Savannah River Company
Savannah River Site
Aiken, South Carolina 29808
R. L. Putman

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D. T. Hobbs, 773-A
R. L. Pulmano, 773-24A

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Westinghouse Savannah River Company
Savannah River Technology Center
Aiken, SC 29808
Summary
Testing examined the extent and rate of strontium, plutonium, neptunium, and uranium removal from a 4.5 M Na\(^+\) solution by adsorption onto monosodium titanate (MST) at 0.2 g/L and 0.4 g/L. Previously, Phase III tests determined removal characteristics at both high neptunium and high uranium concentrations or at low neptunium and low uranium concentrations. These tests evaluated removal at a low neptunium concentration (ca. 0.5 mg/L) and high uranium concentration (ca. 10 mg/L), more typical of expected conditions during salt processing.

Results indicated successful decontamination of strontium, plutonium and neptunium from the salt solution at 0.4 g/L MST, but not at 0.2 g/L MST. Strontium and plutonium removal produced equilibrium concentrations below Z-area feed limits at both concentrations of MST although the data indicated that the isotopic distribution of plutonium will serve as an important design consideration. Neptunium removal did not achieve the Z-Area limit at the lower MST concentration. We attribute less neptunium removal in this set of tests compared to the previous Phase III tests to the increase in uranium concentration. Removal rates determined at both MST concentrations provide additional data for sizing continuously stirred tank reactors and batch tanks for the various Salt Disposition Alternative flowsheets.

Introduction
The Salt Disposition Systems Engineering Team identified the adsorption kinetics of actinides and strontium onto monosodium titanate (MST) as a technical risk in several of the processing alternatives selected for additional evaluation in Phase III of their effort. The Flow Sheet Team requested that the Savannah River Technology Center (SRTC) examine the adsorption kinetics of MST for several process alternatives [1].

Previously, Hobbs and Walker studied the adsorption of plutonium and uranium onto MST in alkaline solutions [2]. Results of these tests indicated that MST would remove strontium, uranium and plutonium from simulated In-Tank Precipitation (ITP) waste solutions. Hobbs and Fleischman followed with statistically designed testing to examine temperature and chemical composition [3]. Again, the results clearly indicated that MST would sufficiently remove strontium, uranium and plutonium.

A later study by D.T. Hobbs, M.G. Bronikowski, T.B. Edwards and R.L. Pulmano identified significant parameters affecting sorption including ionic strength, temperature, initial sorbate concentration and MST concentration [4]. Mixing and the presence of sludge solids exhibited minor effects. Sodium tetraphenylborate (NaTPB) did not significantly affect the extent and rate of removal. Removal rates determined at a low MST concentration allowed initial sizing of reactors in various salt alternative flowsheets. Analysis of the results indicated the need to perform additional kinetic testing with simulants at lower neptunium concentrations and with radioactive waste to confirm the results.

As defined in the technical task request [5], this study used a simulated waste solution to measure the extent and rate of strontium, plutonium, neptunium, and uranium removal at 25 °C in the
presence of 0.2 and 0.4 g/L MST. Future tests will use SRS tank waste to confirm that the extent and rates indicated by these results agree with those in actual radioactive waste.

**Experimental**

**Preparation of Treated Salt Solution**

Personnel prepared two separate 2 L batches of synthetic salt solution from reagent grade chemicals and deionized, distilled (DDI) water yielding the composition provided in Table I. Filtering through Whatman #54 filter paper removed undissolved solids. Researchers combined both filtrates and treated with 50.1 grams of MST slurry (7.6 grams MST, Optima Batch #33407) to remove strontium impurities. After mixing at ambient laboratory temperature for 48 hours, they then filtered the MST and salt solution suspension through Whatman #41 filter paper. We repeated the MST treatment using 15 grams of MST slurry (2.3 grams MST, Optima Batch #33407). A third addition combined 15 grams of MST slurry with the simulated waste.

<table>
<thead>
<tr>
<th>Table I. Composition of Salt Solution</th>
</tr>
</thead>
<tbody>
<tr>
<td>Component</td>
</tr>
<tr>
<td>NaNO₃</td>
</tr>
<tr>
<td>NaOH</td>
</tr>
<tr>
<td>Na₂SO₄</td>
</tr>
<tr>
<td>Al(NO₃)₃·3H₂O</td>
</tr>
<tr>
<td>NaNO₂</td>
</tr>
<tr>
<td>Na₂CO₃</td>
</tr>
<tr>
<td>Total Na⁺</td>
</tr>
</tbody>
</table>

**Addition of Sorbates and Radiotracers**

Table II contains a summary of the target concentrations of the sorbates and radiotracers for the testing.

<table>
<thead>
<tr>
<th>Table II. Target Concentrations for Salt Solution</th>
</tr>
</thead>
<tbody>
<tr>
<td>Species</td>
</tr>
<tr>
<td>----------------</td>
</tr>
<tr>
<td>Strontium-85</td>
</tr>
<tr>
<td>Total Strontium</td>
</tr>
<tr>
<td>Neptunium-237</td>
</tr>
<tr>
<td>Uranium</td>
</tr>
<tr>
<td>Plutonium</td>
</tr>
</tbody>
</table>

Multiple additions and filtrations occurred before the solution reached the desired concentrations of each sorbate. Personnel added strontium-85 as supplied by New England Nuclear (NEZ0A2, 1.83mCi/mL 8/13/98), and uranium supplied by Mallinckrodt (lot 8640) as uranyl nitrate hexahydrate (UO₂(NO₃)₂·6H₂O). Chemical and Hydrogen Technology Section researchers provided neptunium and plutonium as nitric acid solutions. Testing used two different sources of plutonium (i.e., weapons grade Pu-239/240 and heat source Pu-238) to ensure sufficient activity.
for detection. See appendix 1 for table of initial sorbate concentrations in this test compared with previous tests.

**Adsorption Tests**

Researchers filtered the salt solution and placed 130 mL in to each of six labeled PE bottles (duplicates for each MST concentration, and two controls.) They then randomly placed the bottles in a Lab Line shaking waterbath (Cole-Parmer Catalog #E-01290-20) set to maintain a temperature of 25 °C. Personnel kept the level of the water at or above the liquid level in the sample bottles. Temperature measurements occurred via a thermistor thermometer (Omega® Model # 5831) with probes (Omega® Model #OL-703). After incubating for two days, testing began with an initial sampling of the control bottles, and the addition of the appropriate quantity of MST to provide a MST concentration of 0.2 g/L and 0.4 g/L. Sampling of each bottle occurred in random order at 0.25, 0.5, 0.75, 1.0, 1.5, 2, 4, 8, 24, 96, and 168 hours after the addition of MST.

The sampling method consisted of removing the test bottle from the waterbath, briskly shaking for about 30 seconds to provide a homogeneous suspension, and pulling approximately 5-6 mL of the suspension into a disposable plastic syringe. The researcher then inserted a 0.45 μm disk filter (nylon membrane) onto the syringe, collected about 5 mL of filtrate into a clean PE sample bottle and pipetted 4 mL of the resulting filtrate into a glass vial containing 4 mL of 5M nitric acid. A white precipitate formed immediately upon mixing of the sample and the nitric acid. Personnel then capped the glass sample vial, gently agitated it and then allowed it to stand at ambient laboratory temperature until all solids dissolved. Researchers discarded all excess filtrate. Personnel recapped the test bottle and returned it to the waterbath. The total time outside of the waterbath for sampling did not exceed 3 minutes except on the three-day samples when researchers experienced problems with a leaking pipet.

Figure 1 shows a graph of the recorded waterbath temperatures. Measurements, starting with an incubation period two days before the beginning of the experiment, averaged 25.56 °C with a standard deviation of 0.25 °C.

The Analytical Development Section of SRTC performed the analyses. Strontium-85 activity measurement occurred by gamma pulse height spectroscopy. Personnel determined concentrations of strontium-88, neptunium-237, uranium-238, plutonium-239 and plutonium-240 by ICP-MS (Inductively Coupled Plasma Mass Spectroscopy) analysis. Plutonium-239/240 and plutonium-238 activity determination relied on alpha spectroscopy after chemically separating the plutonium from neptunium.
Figure 1. Waterbath Temperature Profiles

Results and Discussion
Researchers performed duplicate tests at each concentration and for the control. All test parameters remained constant (i.e., temperature, ionic strength and initial sorbate concentration) except for MST concentration. Table III shows the test design with MST concentrations as defined in the task technical and quality assurance plan for this report [6].

<table>
<thead>
<tr>
<th>Bottle #</th>
<th>Desired MST conc. (g/L)</th>
<th>Actual MST conc. (g/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.4</td>
<td>0.398</td>
</tr>
<tr>
<td>2</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>3</td>
<td>0.2</td>
<td>0.199</td>
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<tr>
<td>4</td>
<td>0</td>
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<tr>
<td>5</td>
<td>0.2</td>
<td>0.201</td>
</tr>
<tr>
<td>6</td>
<td>0.4</td>
<td>0.401</td>
</tr>
</tbody>
</table>

Control Samples
The experiment used two test bottles (#2 and #4) containing no MST to serve as control tests to correct for removal of sorbates by sorption onto the bottle walls, the filter or by precipitation. The results indicate no significant removal of sorbate in the control samples. For this reason, the results for tests with MST do not require correction for sorption by mechanisms other than that with MST. Figures 2 through 5 compare results with previous test data in which the controls consisted of high and low activity solutions with a salt concentration of 6.0 molarity. With the exception of the last data point in Bottle #4, the sample results for the controls showed little variation. Appendix 2 contains the data from these tests.
The numbers exhibited for the last sample of Bottle #4 appear similar to results in tests containing MST. Though difficult to see in the graphs for plutonium and neptunium (due to the large concentrations in the previous high activity data,) the results in appendices 2 and 4 show that these numbers bear a strong resemblance to the last data points in the test bottles containing 0.2g/L MST. This suggests that the sample did not originate from Bottle #4, but from one of the bottles containing MST. Since the remaining samples did not exhibit a drop in activity and, in particular, the final sample from Bottle #2 remained the same, we eliminated the final sampling point for Bottle #4 from the data set used to quantify experimental error. Only the neptunium data exhibited a standard deviation greater than 4% of the average due to an additional data point with a large error.

Figure 2. Comparison of Strontium Concentration in Controls

![Graph showing comparison of strontium concentration in controls](image-url)
Figure 3. Comparison of Plutonium Concentration in Controls

Figure 4. Comparison of Neptunium Concentration in Controls
Figure 5. Comparison of Uranium Concentration in Controls

Sorption Results
Data from duplicate tests generally exhibited excellent agreement. The difference between duplicate tests and their average values provided a measure of error for the adsorption tests.

Comparison between calculated equilibrium concentrations from the equations developed in the Phase III testing [4] and the experimental results obtained in this study exhibited mixed results (see Table IV). Calculated and experimental values for strontium agreed well at 0.2 g/L MST, but not at 0.4 g/L MST. The plutonium and neptunium results did not agree at either MST concentration. The predicted values were much higher than the measured concentrations. Comparison showed good agreement for uranium at both MST concentrations. We attribute the poor agreement with plutonium to the low correlation coefficient of the Phase III results. For neptunium, we attribute the poor agreement to much higher uranium concentration in the more recent testing. Note that for each case that exhibits poor agreement, the predicted value is greater than the measured value indicating the predictive equations are conservative.

In plots of sorbate concentration versus time, the results exhibited not exponential, but a power decrease in concentration with increasing time agreeing well with results of previous experiments. As discovered in previous experiments, the test data did not fit first or second order kinetic expressions, but with the initial (time = 0) data point removed, the results for each species fit an equation of the following form,

\[ C = at^{-b} \]

with \( C \) = concentration, \( t \) = time, and constants \( a \) and \( b \). Taking the derivative of this expression with respect to time provides a rate equation expressing the change in concentration against the change in time, the necessary information to size a CSTR. Because of the omission of the initial data point, these equations do not provide accurate estimates of concentration and rate and time
before 15 minutes. They also do not predict the initial concentration, because the limit of the calculated concentration as time approaches zero is infinity.

<table>
<thead>
<tr>
<th>Sorbate</th>
<th>[Sr]</th>
<th>[Pu]</th>
<th>[U]</th>
<th>[Np]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Calculated at 0.2 g/L MST</td>
<td>0.44</td>
<td>4.15</td>
<td>6400</td>
<td>151.7</td>
</tr>
<tr>
<td>Actual at 0.2 g/L MST</td>
<td>0.43</td>
<td>0.79</td>
<td>6901</td>
<td>74.27</td>
</tr>
<tr>
<td>Calculated at 0.4 g/L MST</td>
<td>0.42</td>
<td>4.05</td>
<td>5788</td>
<td>171.2</td>
</tr>
<tr>
<td>Actual at 0.4 g/L MST</td>
<td>0.17</td>
<td>0.54</td>
<td>5359</td>
<td>34.13</td>
</tr>
</tbody>
</table>

Removal Efficiency Criteria

The appropriate isotopic limit for decontaminated salt solution sent to Z-Area provides the criterion for removal of each radionuclide [7]. Uranium does not have a criterion, as such, because of the combination of the low specific activity and solubility of uranium. For example, the calculated alpha activity for uranium assuming 100% U-235 (highest specific activity) is 0.015 nCi/g at the initial uranium concentration tested (8.5 mg/L). This is well below the Z-area total alpha limit of 20 nCi/g indicating uranium removal is not required. The neptunium-237 limit, 0.03 nCi/g, equals approximately 51 µg/L for salt solutions at a density of about 1.206 g/mL (4.5 M Na+).

The Sr-90 limit for feed solution to Z-Area is 40 nCi/g [7]. To convert the activity limit to a total strontium mass concentration basis, we set the Sr-90 content at either 45 or 5.2 atomic %. The higher value derives from the known fission yields of Sr-88 and Sr-90 [8] and correcting for radioactive decay of the Sr-90 (assuming an average waste age of 29 years). The lower value equals that determined in a sample of Tank 51H sludge analyzed at SRTC [9]. The 45 atomic % value provides a reasonable upper limit for Sr-90 since impurities in processing chemicals and process water add some amount of non-radioactive strontium into the HLW. This value does not necessarily bound operations since the age of the waste processed could prove less than 29 years. For 5.2 atomic % Sr-90, the upper limit for strontium becomes 6.5 µg/L, and for 45 atomic % Sr-90, the limit becomes 0.76 µg/L.

Plutonium-containing wastes originate from the production of weapons grade (WG) plutonium (containing primarily Pu-239/240) and heat source (HS) plutonium (containing primarily Pu-238). Z-Area limits the total alpha activity in decontaminated salt solution to 20 nCi/g [5]. We converted this limit to a mass concentration assuming all of the alpha activity comes from plutonium and that the plutonium has either a WG or HS isotopic distribution [9,10]. The resulting mass limit for HS plutonium (1.6-1.8 µg/L) proves much lower than that for WG plutonium (40-44 µg/L) due to the much higher specific activity for HS Pu (14.6 Ci/g) compared to WG plutonium (0.60 Ci/g). Appendix 3 contains a table summarizing the Z-area concentration limits of each sorbate in a 4.5 M Na+ solution. Appendices 4 and 5 contain numerical results from the experiment.
Extent and Rate of Strontium Removal
Strontium data generally exhibited strong agreement between duplicate tests. The difference between the duplicate points exceeded 5% of their average value in only one case at 0.2 g/L. The data at 0.4 g/L exhibited more variation, but with a difference never exceeding 10% of the average value. Figure 6 compares the results at 0.2 g/L MST with previous test data containing the same MST concentration from high and low activity solutions at 4.5M salt concentration. Results indicate a higher equilibrium strontium concentration in the more recent tests compared to those in the Phase III studies. We attribute the higher equilibrium concentration to the higher initial concentrations. Despite these higher final concentrations, the graphs show that within 15 minutes the solution meets Z area feed requirements for 5.2 atomic % strontium-90 feed, and, for the 45 atomic % strontium-90 feed, within 4 hours.

Figure 7 contains a graph of the data at 0.4 g/L which lacks a direct comparison in the previous Phase III testing. These tests showed similar results in meeting the requirement for 5.2 atomic % strontium-90 feed, but due to the higher concentration of MST, reached the stricter requirement for 45 atomic % strontium-90 more rapidly. After about 24 hours, little change in concentration occurred in all tests indicating that the system attains equilibrium after reaching these limits.

Trendlines generally fit well to the average data at the two MST concentrations providing equations expressing strontium concentration with respect to time. Figure 8 shows the two trendlines and their resulting equations. Because curve fitting required elimination of the initial data point, the equations do not provide an accurate prediction at the limit for 5.2 atomic % strontium-90 feed. In the region where data crosses the limit for 45 atomic % strontium-90 material, however, the equations provide a relatively close fit at 0.4 g/L MST, and a conservative estimate of adsorption rate at the 0.2 g/L MST concentration.

Further examination of the equations shows that they predict a slower rate of adsorption at the higher MST concentration. Initially adsorption rate in the 0.4 g/L MST tests was faster than at the lower MST concentration, as evidenced by the lower strontium concentration at the time of the first sampling. At some point, the concentration of the strontium in solution falls so low that it controls the rate of adsorption rather than the MST concentration. This transition occurs before the first data point, thus explaining the slower rates predicted by the 0.4 g/L MST equation.
Figure 6. Comparison of Strontium Concentration Tests at 0.2 g/L MST

Figure 7. Comparison of Strontium Concentration Tests at 0.4 g/L MST
Figure 8. Strontium Removal Rates

- 0.2 g/L MST
- 0.4 g/L MST
- Equation (0.2 g/L) MST
- Equation (0.4 g/L MST)

Extent and Rate of Plutonium Removal

The plutonium data in each test agrees well with that from its duplicate as shown in Figures 9 and 10. The difference between the duplicate points exceeded 10% of their average value in only one case at each MST concentration for plutonium-238. In the comparison for total plutonium, two points exhibited such large deviations at the lower MST concentration, but only one point did so for 0.4 g/L MST. Figure 9 compares the data to Phase III test results at the same MST concentration for both the high and low activity 4.5M Na + solutions. The previous low activity results appear very erratic while the high activity data forms a steeper curve crossing the results of this experiment. This behavior may arise from the use of less initial plutonium in the current test solution (see Appendix 1).

Figures 9 and 10 also indicate that under both test conditions, plutonium concentrations rapidly meet the Z-area feed specifications for weapons grade plutonium. Assuming the isotopic percentages characteristic of heat source plutonium, solutions with 0.4 g/L MST meet the requirements faster than those with a concentration of 0.2 g/L MST. This evidence indicates adsorption of plutonium can satisfy Z-area requirements, but the isotopic concentration of plutonium will affect process design and specifications.

A graph (see Figure 11) of the average data at each MST concentration against time provides trendlines describing the experimental data. Because data met the conditions for weapons grade plutonium isotopes so rapidly, the trendlines fail to provide information in that region. Closer to the lower limit, the trendlines exhibit a greater deviation from the experimental data, but the derivative of the equation describing the curve should provide a conservative estimate of the adsorption rates. The equations predict that the rate of adsorption at 0.4 g/L MST is approximately twice as fast as the rate at 0.2 g/L MST.
Figure 9. Comparison of Plutonium Concentration Tests at 0.2 g/L MST

Figure 10. Comparison of Plutonium Concentration Tests in 0.4 g/L MST
Extent and rate of Uranium Removal
Figures 12 and 13 provide the graphs of uranium concentration versus time for the two concentrations of MST. Appendices 4 and 5 contain tables of the numerical data. Duplicate tests exhibited close agreement with differences exceeding 10% of the average value in only one set of points.

Figure 12 compares the current data with Phase III test results at the same MST concentration for both the high and low activity 4.5M Na⁺ solutions. The new data forms a curve between the two sets from the previous tests due to an initial uranium concentration approximately halfway between the concentrations for the high and low activity tests.

Trendlines fit well to the average data at the two MST concentrations providing equations that express uranium concentration with respect to time. Figure 14 shows the two trendlines and their associated equations. The tests with higher MST concentrations achieved equilibrium at lower uranium concentrations, but Z-area requirements place no limit on uranium content.
Figure 12. Comparison of Uranium Tests at 0.2 g/L MST

Figure 13. Comparison of Uranium Concentration Tests at 0.4 g/L MST
Extent and rate of Neptunium Removal

Figures 15 and 16 present plots of the neptunium-237 concentration versus time for both MST concentrations while Appendices 4 and 5 provide the numerical data. The differences between the duplicate data points exceeded 10% of their averages in only two of the sample times for each concentration of MST.

Figure 15 compares the results at 0.2 g/L MST to Phase III test results at the same MST concentration for both the high and low activity 4.5M Na⁺ solutions. In contrast with predictions based upon the previous data for low activity solutions containing similar initial neptunium concentrations, the final neptunium-237 concentration in this experiment exceeded the Z-Area limit in both of the 0.2 g/L MST tests. The significantly higher uranium concentration in this experiment might have caused this behavior due to competition between uranium and neptunium for adsorption sites. The 0.4 g/L MST tests, however, complied with the limit within seven days. Hence, at the uranium and neptunium concentrations of these tests, the process needs MST concentrations larger than 0.2 g/L to meet the Z-area decontamination requirement.

Figure 17 contains a graph of the average data at each MST concentration versus time as well as trend lines describing the data. The curves exhibit excellent agreement with most of the experimental data, but show more deviation for the last two sampling times. The greater uncertainty near the limit results in a conservative estimate of the rate of adsorption.
Figure 15. Comparison of Neptunium Concentration Tests at 0.2 g/L MST

Figure 16. Comparison of Neptunium Concentration Tests at 0.4 g/L MST
Quality Assurance
This work used the following task plan.


This document provides the final deliverable for the work requested in the authorizing task request,

J. R. Fowler, Technical Task Request, HLW-SDT-TTR-99-01.0, revision 0, January 21, 1999

Notebook WSRC-NB-99-00062 (R.L. Pulmano) contains the experimental data obtained from this testing.

Acknowledgements
The authors thank the many people who helped bring this work to completion. M. S. Blume and H. L. Thacker conducted much of the experimental work involved in the adsorption experiments. E. A. Kyser supplied actinide materials. The staff of the Analytical Developmental Section of the SRTC performed the many chemical and radiochemical analyses.

References


Appendix 1.

Initial Sorbate Concentrations
4.5 M Na⁺ Solutions

Initial Concentration (μg/L)

<table>
<thead>
<tr>
<th>Simulant</th>
<th>[Sr]</th>
<th>[Pu]</th>
<th>[U]</th>
<th>[Np]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Phase III High Activity</td>
<td>63.6</td>
<td>168</td>
<td>14800</td>
<td>21000</td>
</tr>
<tr>
<td>Phase III Low Activity</td>
<td>4.96</td>
<td>1.12</td>
<td>1520</td>
<td>340</td>
</tr>
<tr>
<td>Phase IV Low Neptunium</td>
<td>90.0</td>
<td>61.8</td>
<td>9020</td>
<td>423</td>
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</tbody>
</table>
## Appendix 2.
Concentration versus Time Data for Control Samples

<table>
<thead>
<tr>
<th>Bottle 2</th>
<th>Elapsed Time (h)</th>
<th>Total Sr (g/mL)</th>
<th>Pu-239 (g/mL)</th>
<th>Total Pu* (g/mL)</th>
<th>Total U** (g/mL)</th>
<th>Np-237 (g/mL)</th>
</tr>
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<tbody>
<tr>
<td></td>
<td>0.00</td>
<td>8.33E+01</td>
<td>4.35E-04</td>
<td>8.35E+01</td>
<td>9.02E+03</td>
<td>4.06E+02</td>
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<tr>
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<td>0.25</td>
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Average: 9.00E+01
Std. Dev: 2.45E+00
% RSD: 2.74E-02

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Average: 9.00E+01
Std. Dev: 2.96E+00
% RSD: 2.96E-02

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*Total Plutonium based on Pu-239/240 analysis in comparison to which Pu-238 is negligible.
**Total Uranium is sum of U-233, U-234, U-235, U-236, and U-238.
***Data point to be eliminated. See pages 6-7 in report.
Appendix 3.

Concentration Limits for Z-area feed (µg/L)
For 4.5 M Na⁺ salt solution
(density = 1.206 g/mL)

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### Appendix 4.

**Concentration versus Time Data at 0.2 g/L MST**

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<td>31.67%</td>
</tr>
<tr>
<td>168.06</td>
<td>0.40%</td>
<td>4.30%</td>
<td>32.21%</td>
<td>2.76%</td>
<td>21.41%</td>
</tr>
</tbody>
</table>

* Total Plutonium based on Pu-239/240 analysis in comparison to which Pu-238 is negligible.
** Total Uranium is sum of U-233, U-234, U-235, U-236, and U-238.
### Appendix 5.

**Concentration versus Time Data at 0.4 g/L MST**

**Bottle 1**

<table>
<thead>
<tr>
<th>Elapsed Time (h)</th>
<th>Total Sr (µg/L)</th>
<th>Pu-238 (µg/L)</th>
<th>Total Pu* (µg/L)</th>
<th>Total U** (µg/L)</th>
<th>Np-237 (µg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.00</td>
<td>9.00E+01</td>
<td>4.35E-04</td>
<td>6.35E+01</td>
<td>9.02E+03</td>
<td>4.06E+02</td>
</tr>
<tr>
<td>0.25</td>
<td>7.53E-01</td>
<td>7.90E-05</td>
<td>1.14E-01</td>
<td>8.60E+03</td>
<td>3.36E+02</td>
</tr>
<tr>
<td>0.50</td>
<td>5.88E-01</td>
<td>6.77E-05</td>
<td>9.31E-01</td>
<td>7.94E+03</td>
<td>3.00E+02</td>
</tr>
<tr>
<td>0.75</td>
<td>4.95E-01</td>
<td>6.18E-05</td>
<td>9.19E-01</td>
<td>7.46E+03</td>
<td>2.72E+02</td>
</tr>
<tr>
<td>0.98</td>
<td>4.77E-01</td>
<td>5.90E-05</td>
<td>8.60E-01</td>
<td>7.72E+03</td>
<td>2.62E+02</td>
</tr>
<tr>
<td>1.14</td>
<td>3.83E-01</td>
<td>3.07E-05</td>
<td>7.62E-01</td>
<td>7.74E+03</td>
<td>2.36E+02</td>
</tr>
<tr>
<td>2.00</td>
<td>3.43E-01</td>
<td>4.76E-05</td>
<td>8.03E-01</td>
<td>8.03E+03</td>
<td>2.46E+02</td>
</tr>
<tr>
<td>3.98</td>
<td>2.73E-01</td>
<td>5.32E-05</td>
<td>5.43E-01</td>
<td>7.19E+03</td>
<td>1.60E+02</td>
</tr>
<tr>
<td>7.98</td>
<td>2.26E-01</td>
<td>3.11E-05</td>
<td>4.55E-01</td>
<td>7.31E+03</td>
<td>1.49E+02</td>
</tr>
<tr>
<td>23.82</td>
<td>1.93E-01</td>
<td>1.88E-05</td>
<td>2.74E+00</td>
<td>6.57E+03</td>
<td>1.02E+02</td>
</tr>
<tr>
<td>71.98</td>
<td>1.58E-01</td>
<td>8.60E-06</td>
<td>1.26E+00</td>
<td>6.11E+03</td>
<td>6.84E+01</td>
</tr>
<tr>
<td>168.02</td>
<td>1.73E-01</td>
<td>4.14E-06</td>
<td>6.03E+00</td>
<td>5.37E+03</td>
<td>3.34E+01</td>
</tr>
</tbody>
</table>

**Bottle 6**

<table>
<thead>
<tr>
<th>Elapsed Time (h)</th>
<th>Total Sr (µg/L)</th>
<th>Pu-238 (µg/L)</th>
<th>Total Pu* (µg/L)</th>
<th>Total U** (µg/L)</th>
<th>Np-237 (µg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.00</td>
<td>9.00E+01</td>
<td>4.35E-04</td>
<td>6.35E+01</td>
<td>9.02E+03</td>
<td>4.06E+02</td>
</tr>
<tr>
<td>0.30</td>
<td>8.99E-01</td>
<td>8.72E-05</td>
<td>1.27E+01</td>
<td>8.38E+03</td>
<td>3.37E+02</td>
</tr>
<tr>
<td>0.55</td>
<td>5.80E-01</td>
<td>6.65E-05</td>
<td>9.66E+00</td>
<td>7.75E+03</td>
<td>3.00E+02</td>
</tr>
<tr>
<td>0.78</td>
<td>5.01E-01</td>
<td>6.04E-05</td>
<td>8.70E+00</td>
<td>7.76E+03</td>
<td>3.00E+02</td>
</tr>
<tr>
<td>1.03</td>
<td>4.47E-01</td>
<td>5.81E-05</td>
<td>8.06E+00</td>
<td>7.89E+03</td>
<td>2.96E+02</td>
</tr>
<tr>
<td>1.55</td>
<td>3.83E-01</td>
<td>5.13E-05</td>
<td>7.50E+00</td>
<td>7.70E+03</td>
<td>2.21E+02</td>
</tr>
<tr>
<td>2.07</td>
<td>3.23E-01</td>
<td>4.52E-05</td>
<td>6.54E+00</td>
<td>7.24E+03</td>
<td>2.29E+02</td>
</tr>
<tr>
<td>4.07</td>
<td>2.75E-01</td>
<td>3.86E-05</td>
<td>5.64E+00</td>
<td>7.36E+03</td>
<td>1.77E+02</td>
</tr>
<tr>
<td>8.10</td>
<td>2.43E-01</td>
<td>3.02E-05</td>
<td>4.39E+00</td>
<td>7.21E+03</td>
<td>1.49E+02</td>
</tr>
<tr>
<td>24.13</td>
<td>1.70E-01</td>
<td>1.74E-05</td>
<td>2.54E+00</td>
<td>6.50E+03</td>
<td>9.07E+01</td>
</tr>
<tr>
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<td>1.62E-01</td>
<td>1.07E-05</td>
<td>1.56E+00</td>
<td>5.65E+03</td>
<td>5.03E+01</td>
</tr>
<tr>
<td>168.08</td>
<td>1.73E-01</td>
<td>3.64E-06</td>
<td>4.84E-01</td>
<td>5.35E+03</td>
<td>3.48E+01</td>
</tr>
</tbody>
</table>

**Average Data 0.4 g/L MST**

<table>
<thead>
<tr>
<th>Elapsed Time (h)</th>
<th>Total Sr (µg/L)</th>
<th>Pu-238 (µg/L)</th>
<th>Total Pu* (µg/L)</th>
<th>Total U** (µg/L)</th>
<th>Np-237 (µg/L)</th>
<th>% error against averages</th>
<th>Total Sr (µg/L)</th>
<th>Pu-238 (µg/L)</th>
<th>Total U** (µg/L)</th>
<th>Np-237 (µg/L)</th>
<th>% error against averages</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.00</td>
<td>9.00E+01</td>
<td>4.35E-04</td>
<td>6.35E+01</td>
<td>9.02E+03</td>
<td>4.06E+02</td>
<td>0.0%</td>
<td>0.00%</td>
<td>0.00%</td>
<td>0.00%</td>
<td>0.00%</td>
<td>0.00%</td>
</tr>
<tr>
<td>0.25</td>
<td>7.53E-01</td>
<td>7.90E-05</td>
<td>1.14E-01</td>
<td>8.60E+03</td>
<td>3.36E+02</td>
<td>0.0%</td>
<td>0.00%</td>
<td>0.00%</td>
<td>0.00%</td>
<td>0.00%</td>
<td>0.00%</td>
</tr>
<tr>
<td>0.50</td>
<td>5.88E-01</td>
<td>6.77E-05</td>
<td>9.31E-01</td>
<td>7.94E+03</td>
<td>3.00E+02</td>
<td>0.0%</td>
<td>0.00%</td>
<td>0.00%</td>
<td>0.00%</td>
<td>0.00%</td>
<td>0.00%</td>
</tr>
</tbody>
</table>

* Total Plutonium based on Pu-239/240 analysis in comparison to which Pu-238 is negligible.
** Total Uranium is sum of U-233, U-234, U-235, U-236, and U-238.
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