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## Permanganate Treatment Optimization Studies for Strontium and Actinide Removal from High Level Waste Simulants

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### Acronyms and Abbreviations

Ca	Calcium
DF	Decontamination Factor
DWPF	Defense Waste Processing Facility
EDTA	Ethylenediaminetetraacetic Acid
HCOONa	Sodium Formate
HLW	High Level Waste
HS	Heat Source (i.e., mostly $^{238}\text{Pu}$ )
$\text{H}_2\text{O}_2$	Hydrogen Peroxide
M	Moles per Liter

Mn	Manganese
MnO <sub>4</sub> <sup>-</sup>	Permanganate Ion
MST	Monosodium Titanate
Na	Sodium
Np	Neptunium
Pu	Plutonium
RPP	River Protection Program
Sr	Strontium
SRS	Savannah River Site
SRTC	Savannah River Technology Center
U	Uranium
WAC	Waste Acceptance Criterion
WG	Weapons Grade (i.e., mostly <sup>239</sup> Pu)
WPTS	Waste Processing Technology Section
WSRC	Westinghouse Savannah River Company
XRD	X-ray Diffraction

## 1.0 Executive Summary

Approximately 130 million L of High-Level Waste (HLW, the radioactive waste product associated with the dissolution of spent nuclear fuel rods for the recovery of weapons grade material) reside in subsurface tanks awaiting treatment at the Savannah River Site (SRS). The currently proposed designs for the Salt Processing Facility at the SRS include use of monosodium titanate (MST) to remove and concentrate the strontium and actinides (uranium, plutonium and neptunium) from HLW salt solutions. However, the River Protection Program (RPP) at the Hanford Site in Washington State proposes use of permanganate addition for strontium and actinide removal from Hanford HLW.

The MST treatment results in lower solids content than the permanganate treatment according to our calculations from the RPP flowsheet. However, the use of MST for strontium and actinide removal poses a technical risk due to slow removal kinetics particularly for plutonium (e.g., Hobbs et al. 2001 and references therein). Permanganate treatment may offer more rapid removal kinetics than that of MST addition.

To optimize the permanganate treatment for potential use in the SRS application, we studied the addition of permanganate and other agents (such as excess strontium and calcium) that are thought to facilitate high strontium and actinide removal from Hanford waste. If permanganate treatment were selected for treatment of SRS HLW,

exclusion of such agents (such as excess strontium and calcium) from the process if not required would decrease the solids loading of the concentrated waste prior to vitrification of the at the Defense Waste Processing Facility (DWPF).

Our permanganate optimization study examined the effect of calcium concentration, initial manganese (Mn) valence [Mn(II) vs. Mn(VII)], reagent order, reductant concentration, reductant choice (formate vs. peroxide), permanganate concentration, sequential vs. single permanganate addition, seed strontium concentration and ionic strength. Our work yields the following conclusions.

- Addition of dissolved calcium [in the form of soluble calcium nitrate] failed to enhance strontium and actinide removal at 0.013 M Mn concentrations.
- At 0.015 M sodium formate concentration, 0.013 M  $\text{MnO}_4^-$  (permanganate) and 0.01 M Mn(II) nitrate performed similarly with respect to strontium removal. Tests with 0.013 M permanganate (without calcium) proved more effective at actinide removal than 0.01 M Mn(II).
- The order of reagent addition did not significantly influence strontium and actinide removal at initial concentrations of 0.045 M sodium formate and 0.01 M permanganate.
- Studies with variable amounts of sodium formate (0.01 to 0.04 M) at 0.013 M permanganate indicate an optimal reductant concentration as three-fold stoichiometric excess to that of permanganate.
- Tests with peroxide as the reductant demonstrated much higher decontamination factors (DFs) for the actinides than tests with similar amounts of formate (at a three-fold stoichiometric excess to that of permanganate) as a reductant. The waste acceptance criterion (WAC) for dissolved strontium (based on 45 atom % strontium-90, as a worst case scenario), plutonium (assuming heat source (HS) composition, which is mostly plutonium-238) and Np (set at 53 ppb of neptunium-237) were met for all peroxide treatments but not for all of the formate treatments at initial sodium formate levels of 0.045 M and 0.01 M permanganate.
- Removal of strontium and actinides correlated directly with the amount of permanganate ion added. This finding indicates the need for permanganate addition for *both* strontium and actinide removal from our SRS HLW simulant salt solution.
- Sequential addition of permanganate provided greater plutonium and neptunium removal than with single permanganate addition for treatments with 0.045 M formate and a total of 0.01 M permanganate. The test using sequential permanganate addition satisfied the WAC for plutonium [(assuming weapons grade (WG) composition, which is mostly plutonium-239)] while the test with a single, larger permanganate addition failed to meet the requirement.
- Strontium removal proved greatest in the absence of 0.01 M seed strontium addition indicating that seeding with stable strontium does not prove useful in the SRS application. For these tests with 0.001 M or no seed strontium added, the strontium DF remained constant throughout the 168-hour equilibration at sodium formate levels of 0.045 M and 0.01 M permanganate in these treatments.
- After four hours of equilibration, our treatments with 0.01 M strontium nitrate had 1.5 to 2-fold greater actinide removal than treatments with little (0.001 M) or no seed strontium at initial sodium formate levels of 0.045 M.
- Our tests show slightly improved removal of the actinides at low ionic strength (i.e., 4.0 M sodium) than at high ionic strength (5.6 M sodium).
- At initial sodium formate levels of 0.045 M and permanganate levels of 0.01 M, the DF values for uranium and neptunium typically proved highest four hours after the addition of the reagents. Four hours after equilibration, the DFs for uranium and neptunium decreased with time indicating reversible behavior in the removal of uranium and neptunium.

## 2.0 Introduction

The Record of Decision issued on October 17, 2001 identified caustic side solvent extraction as the preferred cesium removal technologies for high-level waste (HLW) treatment at the Savannah River Site. As a pretreatment step for the solvent extraction flowsheet, the incoming salt solution containing small amounts of entrained sludge is

contacted with monosodium titanate (MST) to adsorb strontium (Sr) and actinides (Np, Pu and U).

Testing to design the Hanford Waste Treatment Plant indicate the addition of stable (seed)  $\text{Sr}^{2+}$ ,  $\text{Ca}^{2+}$ , and permanganate show promise for  $\text{Sr}^{2+}$  and actinide removal.<sup>1</sup> This treatment process involves the formation of one or more solids, which precipitate the  $\text{Sr}^{2+}$  [presumably as  $\text{SrCO}_3(s)$ ] and actinides [presumably as a Mn(III, IV) oxide-actinide co-precipitate] in the HLW salt solutions. Studies show that the solids that form in studies with Hanford wastes have good filtration characteristics. Hanford HLW contains considerably more organic compounds [such as formate and ethylenediaminetetraacetic acid (EDTA)] and carbonate ion than SRS waste. Addition of  $\text{Ca}^{2+}$  may benefit  $\text{Sr}^{2+}$  and actinide removal when added prior to the permanganate because  $\text{Ca}^{2+}$  forms strong complexes with EDTA and can displace complexed  $\text{Sr}^{2+}$  ion. These differences in these two waste compositions are significant because permanganate treatment with some RPP wastes appears to require an excess of stable "seed"  $\text{Sr}^{2+}$  (for isotopic dilution of  $\text{Sr}^{2+}$  and Sr precipitation) for Sr removal and a reductant (such as formate ion) for acceptable actinide removal.

The predicted solids loading for MST treatment is lower than that for permanganate treatment according to our calculations from the RPP flowsheet. However, the use of MST 0.4 g  $\text{L}^{-1}$  for Sr and actinide removal poses a technical risk to the current process design due to slow removal kinetics and difficulties in cross-flow filtration (e.g., Hobbs et al. 2001 and references therein). Permanganate treatment may offer faster removal kinetics and improved filtration. Addition of seed  $\text{Sr}^{2+}$  and  $\text{Ca}^{2+}$  would significantly increase the solids loading for permanganate treatment. For a sludge processing rate of 17.5 gallons per minute, 0.01 M  $\text{NaMnO}_4$  (alone) would yield 7.62 lb of solids per hour (as  $\text{MnO}_2$ ). However, if seed  $\text{Sr}^{2+}$  were required for Sr decontamination, a concentration of 0.01 M  $\text{Sr}(\text{NO}_3)_2$  (alone) would yield 9.10 lb of solids per hour (as  $\text{SrO}$ ). If 0.005 M  $\text{Ca}^{2+}$  [as dissolved  $\text{Ca}(\text{NO}_3)_2$ ] were required for Sr decontamination, the  $\text{Ca}^{2+}$  (as  $\text{CaO}$ ) would contribute an additional 2.45 lb of solids per hour (as total Ca). For comparison, 0.4 g MST  $\text{L}^{-1}$  (alone) would yield 4.5 lb of solids per hour.

To achieve optimal removal of the  $\text{Sr}^{2+}$  from SRS waste using permanganate treatment, we must evaluate several aspects prior to consideration for adoption of permanganate into the flowsheet. Our report will focus on the optimization of permanganate treatment with simulant SRS HLW salt solutions. Other Savannah River Technology Center (SRTC) researchers are pursuing other important aspects of permanganate treatment such as the filterability of solids with real and simulant SRS HLW and the removal of Sr and actinides from SRS HLW material. For our simulant studies, we will examine  $\text{Sr}^{2+}$  precipitation in HLW simulants that have low carbonate levels (i.e., near 0.03 M  $\text{CO}_3^{2-}$ ) as opposed to 0.1 M  $\text{CO}_3^{2-}$  as in Hanford wastes and in some SRS wastes.<sup>1,2</sup> We will investigate whether the addition of any potential synergistic precipitating agents (such as  $\text{Ca}^{2+}$ ) is required.

## 2.1 Use of Manganese Oxides for Removal of Radionuclides from Solutions

Manganate solids [the Mn(IV)-dominated oxides] are known for their high surface areas and strong affinities for dissolved cations such as  $\text{Sr}^{2+}$  and actinides (e.g., Pu) in highly alkaline HLW salt solutions. Manganese(III, IV) oxides, which are ubiquitous in the natural environment are noted for their high affinity for actinides and various d-transition metals in oceanic and freshwater systems.<sup>3,4</sup> Additionally, several analytical methods for Pu isolation utilize Mn oxide solids to concentrate or co-precipitate actinides from solutions and from various waste streams.<sup>5,6</sup>

## 3.0 Methods and Materials

We prepared simulant solutions in 4-L batches in addition to several stock solutions of permanganate treatment reagents for two sets of experimental tests. A description of the test methods follows. A third study used non-radioactive simulant in glass beakers to examine color changes because the plastic poly bottles used in our tests with spiked simulants made accurate color observations difficult.

### 3.1 Sample Preparation Description for the First Set of Tests

For these studies, we made a synthetic HLW salt solutions of 1.33 M NaOH, 2.6 M NaNO<sub>3</sub>, 0.43 M NaAl(OH)<sub>4</sub>, 0.34 M NaNO<sub>2</sub>, 0.52 M Na<sub>2</sub>SO<sub>4</sub> and 0.026 M Na<sub>2</sub>CO<sub>3</sub>. We prepared the solution under acidic conditions. A Na<sub>2</sub>CO<sub>3</sub> solution was prepared separately and pretreated with MST to remove the tramp Sr that typically originates from the reagent grade Na<sub>2</sub>CO<sub>3</sub>. The MST was allowed to equilibrate with the Na<sub>2</sub>CO<sub>3</sub> solution for 24 hours. The suspension was then filtered to remove the MST. After the solution was filtered it was added to the acidic starting solution. We then spiked the simulant solution with our target spike levels of 100 ppb stable Sr (with trace <sup>85</sup>Sr), 500 ppb <sup>237</sup>Np, 10,000 ppb <sup>238</sup>U and 100 ppb (or 200 ppb as noted below) weapons grade (WG) <sup>239/240</sup>Pu (6 % <sup>240</sup>Pu). The spike was followed by the addition of the 1.33 M NaOH, 0.43 M NaAl(OH)<sub>4</sub>, 0.34 M NaNO<sub>2</sub>, and 0.52 M Na<sub>2</sub>SO<sub>4</sub>. Without the removal of tramp Sr with MST prior to the addition of our target radionuclides, our solution would have had a six-fold greater Sr concentration than that of our spiked solution. [The measured concentrations for the radionuclides in our simulants are shown in section 8.0 entitled EXPERIMENTAL DATA.]

Our tests with this solution were run in a shaken water bath using 100-mL polybottles with 60 mL of spiked salt simulant solution. The Sr(NO<sub>3</sub>)<sub>2</sub>, Ca(NO<sub>3</sub>)<sub>2</sub>, sodium formate (HCOONa), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) and sodium permanganate (NaMnO<sub>4</sub>) stock solutions were made by dissolving the reagent grade solids in de-ionized distilled water to give the stock solution concentrations listed in **Table 3-1**. The peroxide (H<sub>2</sub>O<sub>2</sub>) solution was used in its most concentrated form (30 wt %). **Table 3-1** also shows that addition of the stock solutions to the salt simulants did not dilute the salt simulants beyond 4 vol %. We also performed simultaneous tests with 0.4 g L<sup>-1</sup> of MST (Lot #33180) for comparison.

A description of the tests and their controls is given in **Table 3-2**. Samples were hand-shaken after each reagent addition and placed in the water bath until the next addition or until sampling. The water bath temperature was maintained at 25 °C. Each reagent addition was 15 minutes apart and sampling began after the last addition, which we refer to as time 0. However, for the peroxide tests, we added the peroxide last and two to three minutes after adding the permanganate solution. Some foaming was observed during this peroxide test. We sampled all of the tests from this first set of experiments at 0, 0.5, 2, 4, 24 and 168 hours except the peroxide tests [and its corresponding MST treatment (Test 11) and control solution], which were sampled at 1, 2, 4, 24 and 168 hours. We filtered the samples with 0.45-μm nylon membrane syringe filters and acidified them (1:1) in 5 M trace metal grade nitric acid (HNO<sub>3</sub>).

**Table 3-1 Stock Solution Concentrations, Target Test Concentrations and the Approximate Volume of Stock Solution added to the Two Sets of Tests.**

Stock Solution Concentrations	Targeted Test Concentrations	Approximate Volume of Stock Solution Added to the Spiked Salt Simulant *
2.88 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.010 M	0.4 mL
	0.010 M	0.2 mL (to 60 mL)
	0.005 M	0.2 mL
	0.001 M	0.04 mL
	0.001 M	0.02 mL (to 60 mL)
0.75 M NaMnO <sub>4</sub>  3.75 M NaMnO <sub>4</sub>	0.002 M	0.3 mL
	0.005 M	0.8 mL
	0.010 M	1.5 mL
	0.0013 M	0.02 mL (to 60 mL)
	0.0065 M	0.1 mL (to 60 mL)
	0.013 M	0.2 mL (to 60 mL)
2.94 M HCOONa	0.009 M	0.4 mL
	0.015 M	0.35 mL (to 60 mL)
	0.020 M	0.4 mL (to 60 mL)
	0.030 M	0.6 mL (to 60 mL)
	0.040 M	0.8 mL (to 60 mL)

	>0.04 M 0.045 M	>0.8 mL (to 60 mL) ** 1.8 mL
2.88 M Ca(NO <sub>3</sub> ) <sub>2</sub>	0.005 M	0.5 mL (to 60 mL)
6 wt % H <sub>2</sub> O <sub>2</sub>	0.045 M	2 mL
30 wt % H <sub>2</sub> O <sub>2</sub>	0.060 M	0.54 mL total (to 60 mL)

\* Small dilutions (4 vol % at most) of the salt solutions occurred due to addition of the reagents. Approximate volumes listed for test conditions are for salt simulant volumes of 115 mL or 60 mL as noted.

\*\* Estimated to be much greater than 0.04 M formate but the concentration is not known. Enough formate was added to change the permanganate solution color from purple to brown. The sodium formate was added in 0.01 M quantities.

**Table 3-2 Experimental Design for the First Set of Tests.**

Test*	Simulant	Addition 1	Addition 2	Addition 3	Addn. 4	Replication
Control	5.6 M Na <sup>+</sup>	-	-	-	-	2
1	5.6 M Na <sup>+</sup>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.01 M Mn(NO <sub>3</sub> ) <sub>2</sub>	-	-	1
2	5.6 M Na <sup>+</sup>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.013 M NaMnO <sub>4</sub>	0.015 M Na Formate	-	1
3	5.6 M Na <sup>+</sup>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.0066 M NaMnO <sub>4</sub>	0.015 M Na Formate	-	1
4	5.6 M Na <sup>+</sup>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.0013 M NaMnO <sub>4</sub>	0.015 M Na Formate	-	1
5	5.6 M Na <sup>+</sup>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.013 M NaMnO <sub>4</sub>	0.020 M Na Formate	-	1
6	5.6 M Na <sup>+</sup>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.013 M NaMnO <sub>4</sub>	0.030 M Na Formate	-	1
7	5.6 M Na <sup>+</sup>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.013 M NaMnO <sub>4</sub>	0.040 M Na Formate	-	1
8	5.6 M Na <sup>+</sup>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.013 M NaMnO <sub>4</sub>	>0.040 M Na Formate	-	1
9	5.6 M Na <sup>+</sup>	0.005 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.013 M NaMnO <sub>4</sub>	0.015 M Na Formate	-	1
10	5.6 M Na <sup>+</sup>	0.001 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.013 M NaMnO <sub>4</sub>	0.015 M Na Formate	-	1
11	5.6 M Na <sup>+</sup>	-	-	MST	-	2**
12	5.6 M Na <sup>+</sup>	0.005 M Ca(NO <sub>3</sub> ) <sub>2</sub>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.013 M NaMnO <sub>4</sub>	0.015 M Na Formate	1
13	5.6 M Na <sup>+</sup>	0.005 M Ca(NO <sub>3</sub> ) <sub>2</sub>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.013 M Mn(NO <sub>3</sub> ) <sub>2</sub>	-	1
14	5.6 M Na <sup>+</sup>	0.005 M Ca(NO <sub>3</sub> ) <sub>2</sub>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.01 M NaMnO <sub>4</sub>	4 x 0.015 M H <sub>2</sub> O <sub>2</sub>	2***

\* These tests were performed to determine the optimal  $MnO_4^-$  concentration, the optimal reductant (as sodium formate or hydrogen peroxide) concentration, whether  $Ca^{2+}$  addition was required, and the optimal seed  $Sr^{2+}$  concentration for Sr and actinide removal (using lower sodium formate concentrations than those tested in **Table 3-3**).

\*\*Test 11 was performed with MST for comparison using target levels of 100 and 200 ppb Pu as noted.

\*\*\*Test 14 was performed with a target Pu concentration of 200 ppb Pu in duplicate.

We sent all 30 minute, 2, 4, 24 and 168-hr samples for analysis by inductively-coupled argon plasma mass spectrometry (ICP-MS) for  $^{238}U$ ,  $^{239/240}Pu$  and  $^{237}Np$ . We also had our samples analyzed by gamma spectroscopy for  $^{85}Sr$  and by triphenyltrifluoroacetone (TTA) scintillation analysis for  $^{239/240}Pu$ . A more description of how these analyses are typically conducted on our HLW simulant solutions is provided elsewhere.

### 3.2 Sample Preparation Description for the Second Set of Tests

Our second set of Mn-containing tests was performed after we had examined the analytical data from the first set of tests. From these initial tests, we determined that higher concentrations of reductant (relative to that of the permanganate concentration) were needed for Sr and actinide removal. Therefore, we performed a second set of tests at higher reductant concentrations.

For these studies, we made a synthetic HLW salt solution of 1.33 M NaOH, 2.6 M  $NaNO_3$ , 0.43 M  $NaAl(OH)_4$ , 0.34 M  $NaNO_2$ , 0.52 M  $Na_2SO_4$  and 0.026 M  $Na_2CO_3$ . We prepared the solution under acid conditions. A  $Na_2CO_3$  solution was prepared separately and pretreated with MST to remove the tramp Sr that typically originates from the reagent grade  $Na_2CO_3$ . The MST was allowed to equilibrate with the  $Na_2CO_3$  solution for 24 hours. The solution was then filtered to remove the MST. After the solution was filtered it was added to the acidic starting solution. We then spiked the simulant solution with our target spike levels of 100 ppb stable Sr (with trace  $^{85}Sr$ ), 500 ppb  $^{37}Np$ , 10,000 ppb  $^{238}U$  and 200 ppb weapons grade (WG)  $^{239/240}Pu$  (6 %  $^{240}Pu$ ) and this step was followed by the addition of the 1.33 M NaOH, 0.43 M  $NaAl(OH)_4$ , 0.34 M  $NaNO_2$ , and 0.52 M  $Na_2SO_4$ . Without the removal of tramp Sr with MST prior to the addition of our target radionuclides, our solution would have had a six-fold greater Sr concentration than that of our spiked solution. We made the lower ionic strength salt simulant solutions (listed in **Table 3-3**) by diluting the spiked solution with de-ionized water as appropriate. [The measured concentrations for the radionuclides in our simulants are shown in section entitled EXPERIMENTAL DATA.]

Our tests were run in a shaken water bath using 125-mL polyethylene bottles with 115 mL of spiked salt simulant solution. A description of the tests with controls is given in **Table 3-3**. The  $Sr(NO_3)_2$ ,  $Ca(NO_3)_2$ ,  $HCOONa$ ,  $H_2O_2$  and  $NaMnO_4$  stock solutions were made by dissolving the reagent grade solids in de-ionized distilled water to give the stock solution concentrations listed in **Table 3-1**. We diluted the hydrogen peroxide ( $H_2O_2$ ) solution to 5 wt % as noted. **Table 3-1** also shows that addition of the stock solutions to the salt simulants did not dilute the salt simulants beyond 4 vol %.

Our controls for the second set of tests were run as singles but analyzed in duplicate. We sampled all of the tests except 25 and 27 at 0.5, 4, 24, 96 and 168 hours. We sampled tests 25 and 27 at 2, 4, 24, 96, and 168 hours. Our controls were samples at 0, 0.5, 4, 24, 96 and 168 hours and we analyzed the 0 and 168 h samplings. All samples filtered with 0.45- $\mu$ m nylon membrane syringe filter and acidified (1:1) in 5 M trace metal grade  $HNO_3$ .

All of 4, 24 and 168-hr samples were analyzed by ICP-MS for stable Sr, U, Pu and Np. We also had the samples analyzed by gamma spectroscopy for  $^{85}Sr$  and by TTA extraction and scintillation analysis for  $^{239/240}Pu$ . The water bath temperature was maintained at 24 °C. All samples were shaken after addition except for the peroxide tests. The samples with peroxide addition foamed after the last reagent (Addition 3 as in Table 3-1) was added. We were unable to sufficiently mix these samples or place them in the water bath until we had performed 2 hours of gently swirling the bottles from time to time. Consequently, we did not pull a 30-minute sample from these peroxide-containing tests. Rather, we sampled the bottles after 2 hours of equilibration outside of the water bath, verified that the foaming had stopped, capped the polyethylene bottles and then placed them in the water bath.

### 3.3 Sample Preparation Description for Use in Visual Observations of Sample Color Changes during Permanganate Treatment with Cold Simulants

To examine the color changes and foaming, we prepared glass beakers with non-radioactive salt simulant (with a chemical composition similar to that used in our first two experimental tests) and performed tests 17, 25 (in duplicate), 26 and 27 (in duplicate). Two peroxide tests were done twice so that we could examine foaming under hand shaken and mechanically stirred conditions.

**Table 3-3 The Second Set of Permanganate Optimization Tests were Performed to Study the Effect of Ionic Strength, Reagent Order, Reductant Choice and Seed Sr Concentration.**

Test*	Simulant	Addition 1	Addition 2	Addition 3	Replication
Control	4.0 M Na <sup>+</sup>	-	-	-	1
Control	4.7 M Na <sup>+</sup>	-	-	-	1
Control	5.6 M Na <sup>+</sup>	-	-	-	1
Control	5.6 M Na <sup>+</sup>	-	-	-	1
15	4.0 M Na <sup>+</sup>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.045 M Na Formate	0.01 M NaMnO <sub>4</sub>	2
16	4.7 M Na <sup>+</sup>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.045 M Na Formate	0.01 M NaMnO <sub>4</sub>	2
17	5.6 M Na <sup>+</sup>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.045 M Na Formate	0.01 M NaMnO <sub>4</sub>	2
18	5.6 M Na <sup>+</sup>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.045 M Na Formate	0.005 M NaMnO <sub>4</sub>	2
19	5.6 M Na <sup>+</sup>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.009 M Na Formate	0.002 M NaMnO <sub>4</sub>	2
20	5.6 M Na <sup>+</sup>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.045 M Na Formate	-	2
21	5.6 M Na <sup>+</sup>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.045 M Na Formate	4 x 0.0025 M NaMnO <sub>4</sub>	2
22	5.6 M Na <sup>+</sup>	-	0.01 M NaMnO <sub>4</sub>	0.045 M Na Formate	2
23	5.6 M Na <sup>+</sup>	0.001 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.01 M NaMnO <sub>4</sub>	0.045 M Na Formate	2
24	5.6 M Na <sup>+</sup>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.01 M NaMnO <sub>4</sub>	0.045 M Na Formate	2
25	5.6 M Na <sup>+</sup>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.01 M NaMnO <sub>4</sub>	0.045 M H <sub>2</sub> O <sub>2</sub>	2
26	5.6 M Na <sup>+</sup>	0.045 M Na Formate	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.01 M NaMnO <sub>4</sub>	2
27	5.6 M Na <sup>+</sup>	0.045 M H <sub>2</sub> O <sub>2</sub>	0.01 M Sr(NO <sub>3</sub> ) <sub>2</sub>	0.01 M NaMnO <sub>4</sub>	2

\* These tests were split into two experimental subsets because of limitations in shaker bath and hood space. A control (or controls) was run in these two subsets when appropriate. For example, the first subset of experiments contained a 5.6 M Na<sup>+</sup> control that applied to tests 18, 19, 20, 21, 22, 23 and 24. The second subset contained three controls (at 4.0, 4.7 and 5.6 M Na<sup>+</sup>) that applied to tests 15, 16, 17, 25, 26 and 27.

## 4.0 Results and Discussion

We will present a summary table of all of the results that shows the DF values that we calculated for the tests after we describe the results of the tests in detail.

### 4.1 Tests with 0.005 M Ca<sup>2+</sup> Addition and Initial Mn Oxidation State: Mn(II)<sup>2+</sup> vs. Mn(VII)O<sub>4</sub><sup>-</sup> at 0.01 M (each) on Sr and Actinide Decontamination

The data presented in this section are from our first set of tests as described in **Section 0**.

#### 4.1.1 Strontium

We used the  $^{85}\text{Sr}$  data to calculate a total Sr solution concentration of  $100 \mu\text{g Sr L}^{-1}$  so that our Sr data from the test with MST addition (and other previous tests, e.g. Hobbs et al. 2001) could be readily compared with Sr data from our permanganate testing. The results of our testing with Sr are shown in **Figure 4-1**. There was little benefit to adding  $0.005 \text{ M Ca}^{2+}$  for improving Sr removal in the Mn(II) and the permanganate  $[\text{MnO}_4^-]$  treatments [using a  $0.015 \text{ M}$  reductant (formate) concentration] as shown in

**Figure 4-1**. This suggests the dissolved Sr in our simulant was not strongly complexed and that our treatment does not require  $\text{Ca}^{2+}$  to disassociate  $\text{Sr}^{2+}$  complexes as with Hanford HLW testing.

There were slight differences between the behavior of  $\text{Mn(II)}^{2+}$  and  $\text{Mn(VII)}\text{O}_4^-$  (permanganate) on Sr removal in the tests. Addition of Mn(II) instead of permanganate was more effective at short equilibration periods (i.e., 30 minutes) than the permanganate treatments. However, we observed that our permanganate treatment without  $0.005 \text{ M Ca}^{2+}$  was more effective at Sr removal after two hours of equilibration (at the  $0.015 \text{ M}$  formate concentration) than in our two Mn(II) tests and the permanganate test with  $0.005 \text{ M Ca}^{2+}$  (**Figure 4-1**). After the 2-hour sampling, the dissolved Sr concentrations in all the tests increased slightly at 4 hours (**Figure 4-1**). After 4 hours, the dissolved Sr concentrations for all of the Mn-containing treatments decreased and the Sr levels were consistently lower than that of the MST addition test (**Figure 4-1**).

The feed solution limit for  $^{90}\text{Sr}$  to the Saltstone (Z-Area) Facility is  $40 \text{ nCi g}^{-1}$ . We converted the activity limit for  $^{90}\text{Sr}$  to two total mass concentration bases at 45 atom %  $^{90}\text{Sr}$  and 5.2 atom %  $^{90}\text{Sr}$  as described in Hobbs et al. (2001). After 30 minutes, the Saltstone process limit or WAC of  $6.8 \mu\text{g Sr L}^{-1}$  for  $^{90}\text{Sr}$  (at 5.2 atom %) was met in all treatments at all sampling times with the exception of the 4-hour equilibration with permanganate and  $0.005 \text{ M Ca}^{2+}$ . The WAC of  $0.79 \mu\text{g Sr L}^{-1}$  (for  $^{90}\text{Sr}$  at 45 atom %) was only met in the Mn(II) treatments (with and without  $\text{Ca}^{2+}$  addition) at 30 minutes of equilibration.

#### 4.1.2 Plutonium

Our tests show that Pu decontamination in the permanganate tests was negatively effected by  $0.005 \text{ M Ca}^{2+}$  addition [at a  $0.015 \text{ M}$  formate concentration] as shown in **Figure 4-2**. This may be due to potential competition of  $\text{Ca}^{2+}$  with Pu for the Mn-oxide solid that was formed upon precipitation. Our Pu DF in this treatment was greatest after 4 hours. After 4 hours, the Pu concentrations increased with time from  $2.4 \mu\text{g Pu L}^{-1}$  to  $14 \mu\text{g L}^{-1}$  (at 168 hours). After 30 minutes, these two permanganate  $[\text{Mn(VII)}]$  treatments met the Saltstone WAC (assuming a total alpha activity based on WG Pu isotopes) of  $42 \mu\text{g L}^{-1}$ . However, these two treatments did not meet the Saltstone WAC for a total alpha activity based on Heat Source (HS) Pu ( $1.7 \mu\text{g L}^{-1}$ ). The Mn(II) treatments (with and without  $\text{Ca}^{2+}$ ) performed similarly to that of the MST, but these treatments were less effective at Pu decontamination than the permanganate tests (**Figure 4-2**).

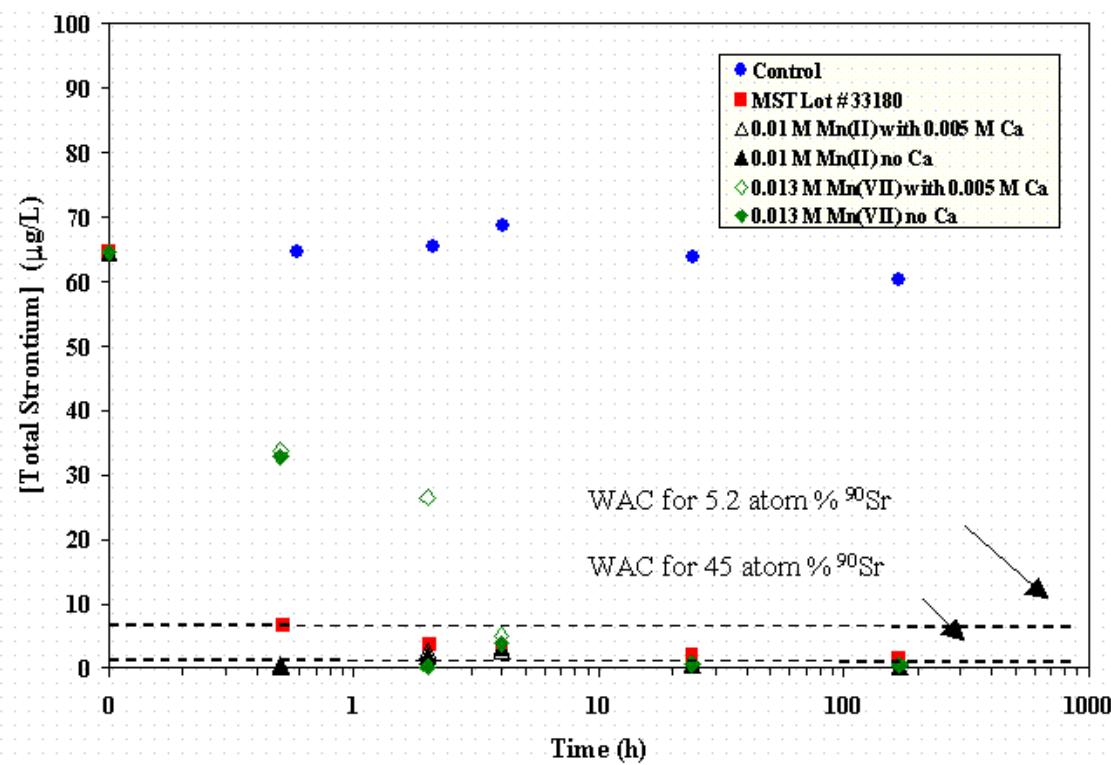
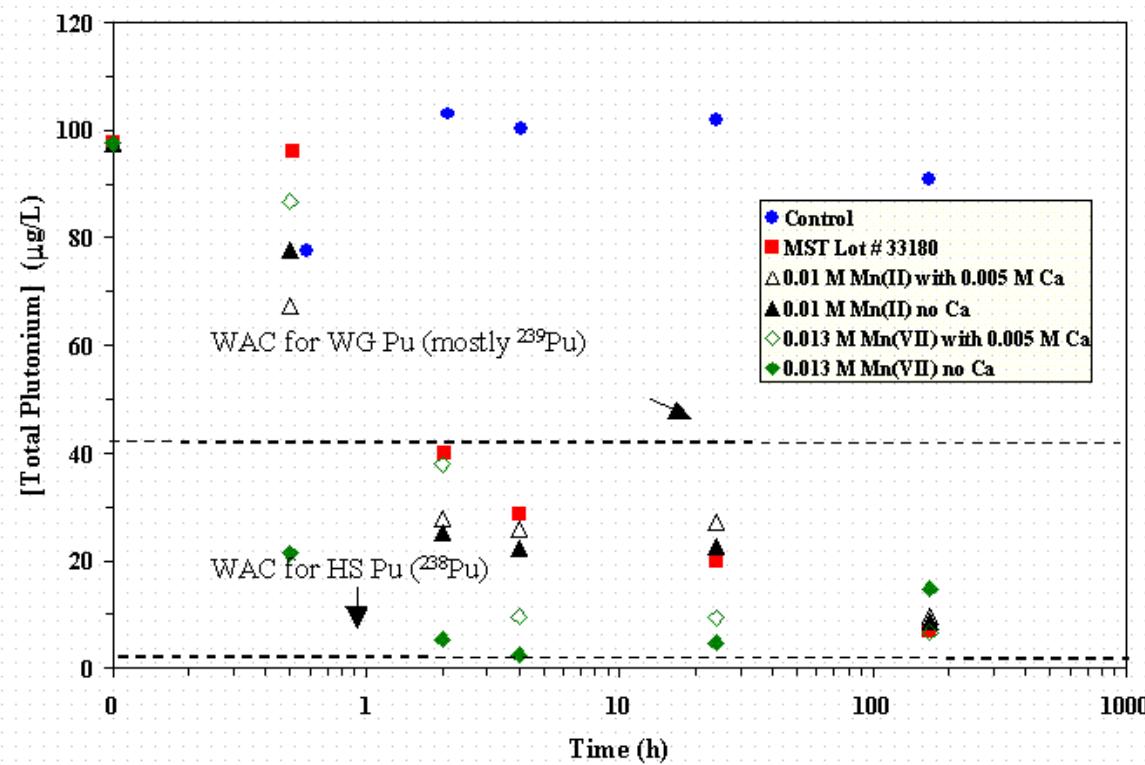


Figure 4-1 Effect of  $\text{Ca}^{2+}$  Addition and Initial Mn Oxidation State on  $\text{Sr}^{2+}$  Removal in SRS HLW Simulant.



**Figure 4-2 Effect of  $\text{Ca}^{2+}$  Addition and Initial Mn Oxidation State on Pu Removal in SRS HLW Simulant PuTTA Data.**

#### 4.1.3 Uranium

The ICP-MS data for U in the control tests indicate that a loss of U from solution (either from precipitation or sorption to container walls) had occurred during the 2- through 24-hour samplings (Figure 4-3). Because of the lower levels measured by ICP-MS, the values for our tests were corrected to account for the lower ICP-MS sensitivity by multiplying the test values by the percent loss of U in the control. The corrected U data for the  $\text{Ca}^{2+}$  addition with Mn(II) or permanganate test are presented in Figure 4-4.

Figure 4-4 shows that the MST performed similarly to the two Mn(II) treatments with and without  $\text{Ca}^{2+}$  addition. Our permanganate treatments had better U decontamination than the Mn(II) and MST treatments in the absence of  $\text{Ca}^{2+}$ . Our two permanganate-containing tests (with and without  $\text{Ca}^{2+}$  addition) had slight decreases in DF after the 24-hour sampling indicating U uptake was somewhat reversible over time in our simulants. The DF for the two Mn(II) tests remained fairly constant throughout the 168-hour study. There is no WAC limit of concern for U in these tanks because the alpha activity of U in the SRS waste is negligible in comparison to that of Pu and Np.

#### 4.1.4 Neptunium

The ICP-MS data for Np in the control tests indicate that a loss of Np from solution (either from precipitation or sorption to container walls) had occurred during the 2- through 24-hour samplings (Figure 4-5). We corrected these values as discussed for U in Section 4.1.3. The corrected ICP-MS data for Np are shown in Figure 4-6.

Figure 4-6 shows that the MST addition performed slightly better with Np than the two Mn(II) treatments (with and without  $\text{Ca}^{2+}$  addition). Our test with MST continued to remove Np throughout the study whereas the permanganate tests did not. The dissolved Np levels in the permanganate tests increased after the 24-hour sampling. The

dissolved Np levels for our two Mn(II) tests were nearly constant throughout the 30 minute and 24 hour samplings and they decreased slightly after 168 hours.

In general, our two permanganate treatments had faster Np decontamination than the MST treatments and the Mn(II) treatments, particularly in the absence of  $\text{Ca}^{2+}$ . In the absence of  $\text{Ca}^{2+}$ , the permanganate-containing test had a much lower Np concentration (with a DF near 1.6) after 30 minutes (relative to the other treatments) indicating that removal was more rapid with this treatment than with the other treatments. This Np treatment met the Saltstone WAC of  $53 \mu\text{g} \text{ }^{237}\text{Np L}^{-1}$  by 24 hours.

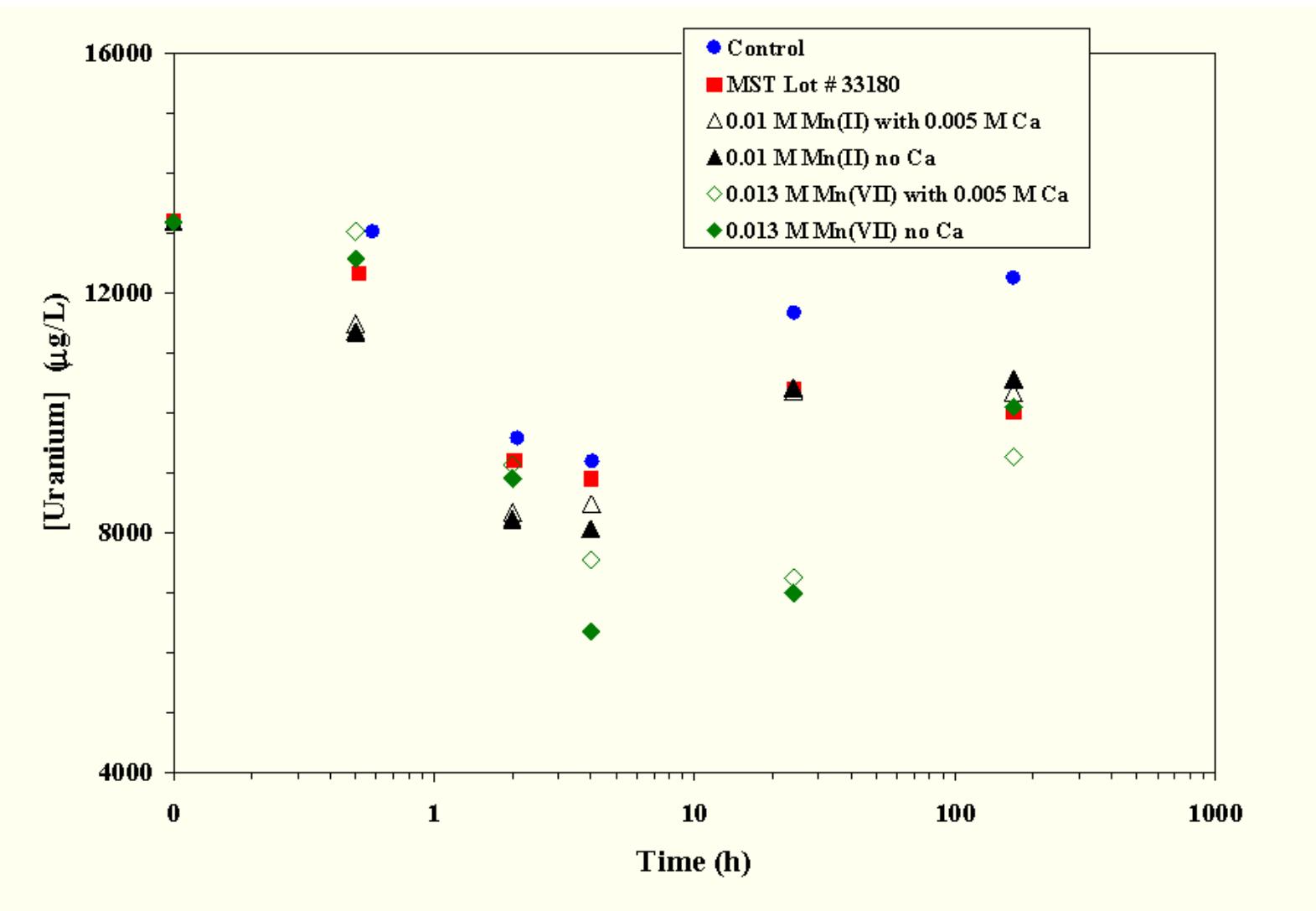


Figure 4-3 Effect of  $\text{Ca}^{2+}$  Addition and Initial Mn Oxidation State on U Removal in SRS HLW Simulant Uncorrected.

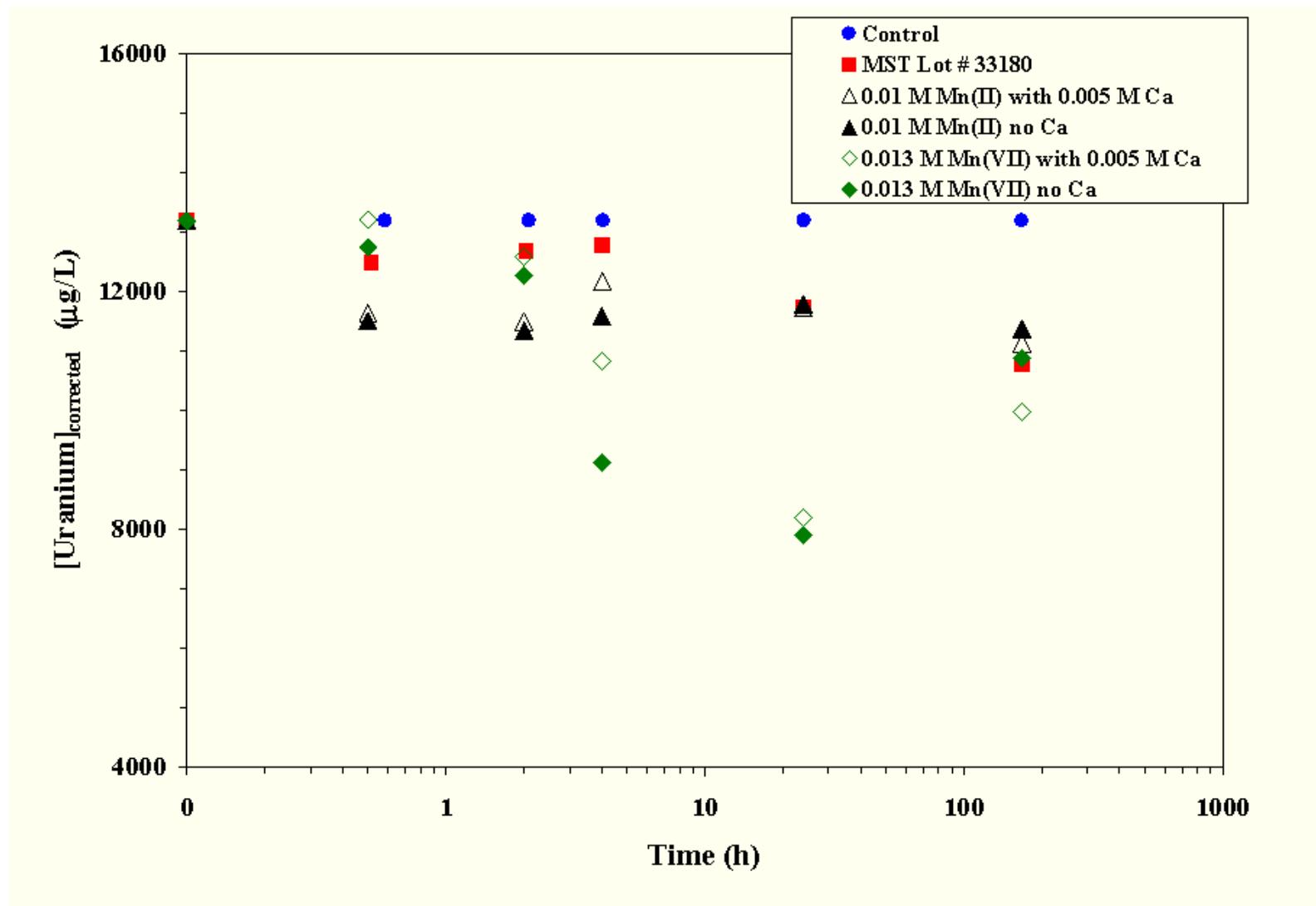
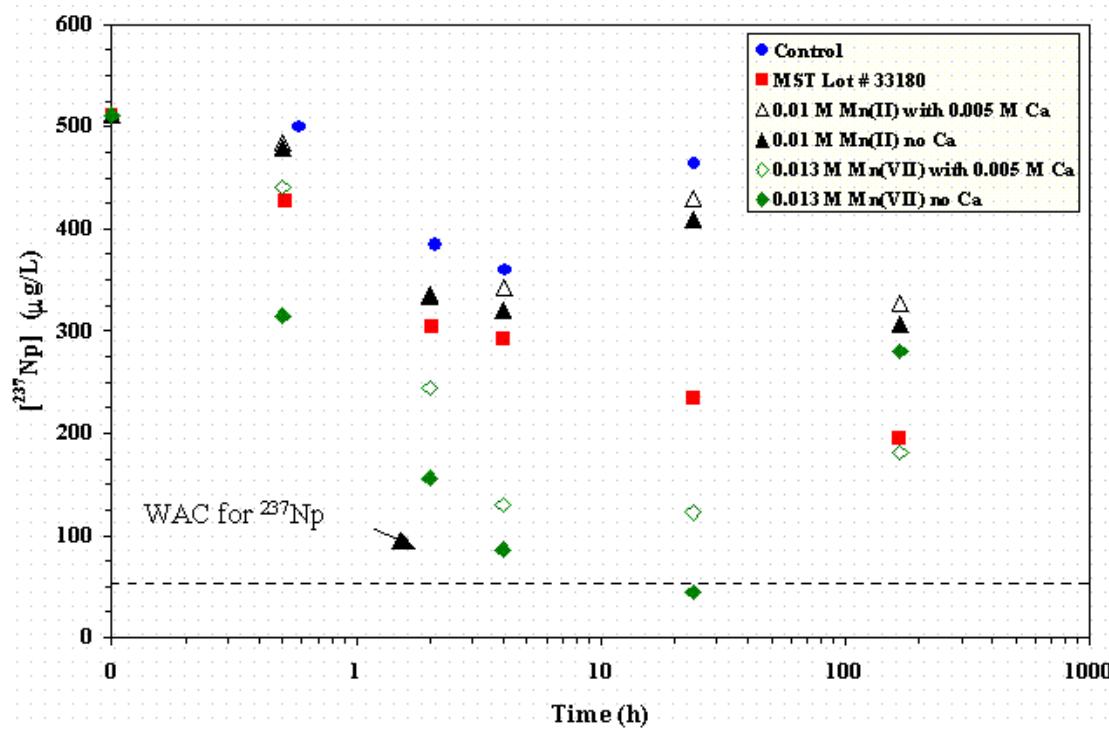
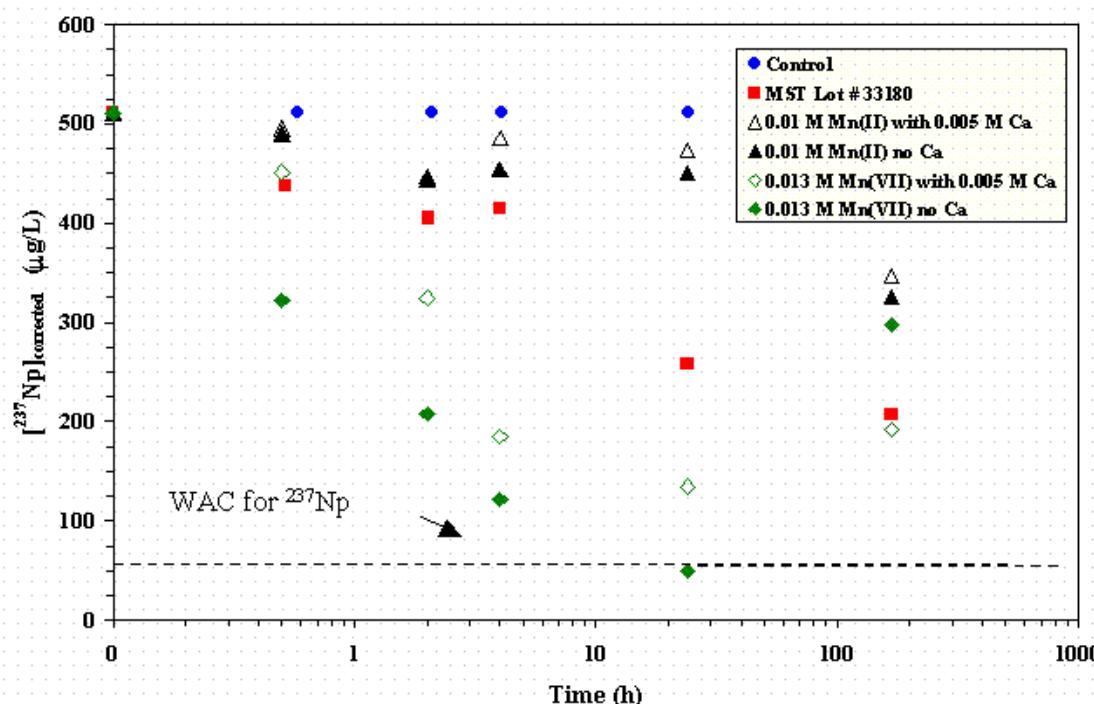


Figure 4-4 Effect of  $\text{Ca}^{2+}$  Addition and Initial Mn Oxidation State on U Removal in SRS HLW Simulant  $\diamond$ Corrected.



**Figure 4-5 Effect of  $\text{Ca}^{2+}$  Addition and Initial Mn Oxidation State on Np Removal in SRS HLW Simulant** Uncorrected.



**Figure 4-6 Effect of  $\text{Ca}^{2+}$  Addition and Initial Mn Oxidation State on Np Removal in SRS HLW Simulant**  $\diamond$  Corrected.

#### 4.2 Use of $\text{H}_2\text{O}_2$ (at $>0.06$ M) as a Reductant with 0.01 M Permanganate Treatment vs. MST Addition (Only)

The data presented in this section are from our first set of tests as described in **Section 0**. The large error in the actinide data for this set of tests is most likely due to the fact that we added slightly different peroxide amounts to insure the Mn(VII) we added became reduced (i.e., until a color change occurred).

##### 4.2.1 Strontium

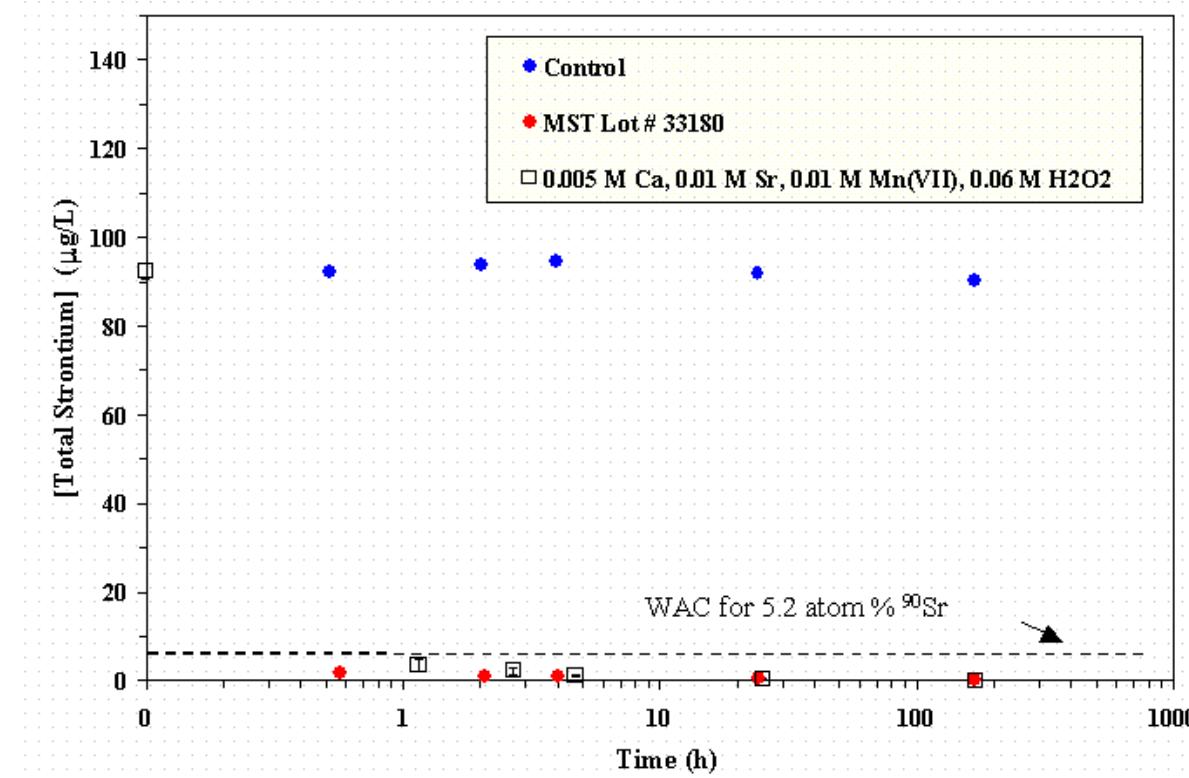
The  $^{85}\text{Sr}$  data for our spiked control solution was used to calculate a total Sr solution concentration of  $100 \mu\text{g Sr L}^{-1}$  so that the Sr data for the test with MST addition could be readily compared with Sr data from our permanganate testing as shown in **Figure 4-7**.

The Saltstone WAC for Sr (at 5.2 atom % of  $^{90}\text{Sr}$ ) of  $6.8 \mu\text{g Sr L}^{-1}$  was met in all of the treatments at all sampling times (**Figure 4-7**). Additionally, all of our tests closely approached the lower WAC for Sr (for 45 atom. % of  $^{90}\text{Sr}$ ) of  $0.79 \mu\text{g Sr L}^{-1}$ . Our data indicate that the MST addition and permanganate treatment (using  $>0.06$  M peroxide, added sequentially) have similar and rapid removal kinetics. The Sr removal under all of these conditions was not reversible in these solutions throughout the 168-hour study.

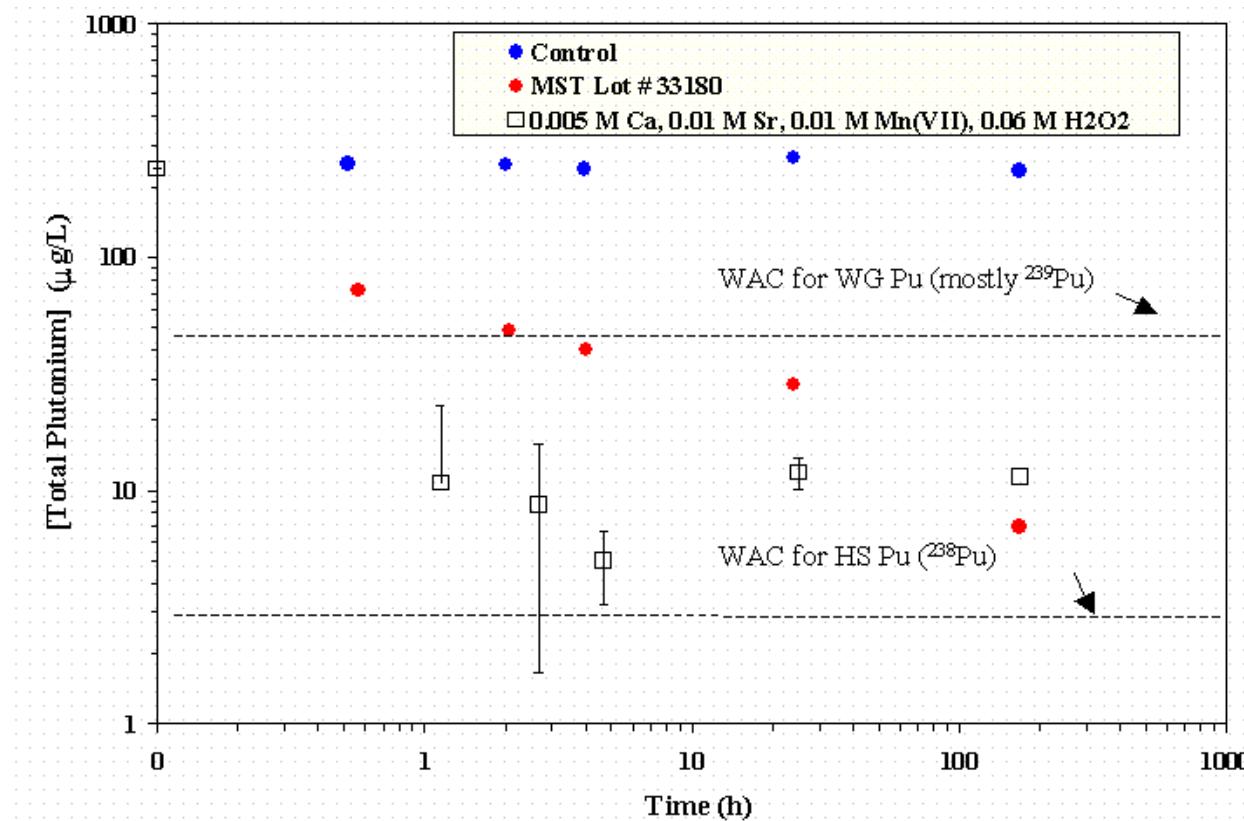
##### 4.2.2 Plutonium

For Pu, our studies show that the permanganate treatment with peroxide as the reductant has faster removal kinetics than that of MST addition (**Figure 4-8**). Our permanganate test with peroxide met the WAC for WG Pu within 1 hour of equilibration whereas the tests with MST addition did not approach the WAC for WG Pu until

after 4 hours of equilibration. These two types of treatment had nearly the same amount of Pu removal after 168 hours and neither treatment met the WAC for HS Pu (**Figure 4-8**).



**Figure 4-7** Permanganate Treatment with Peroxide Versus MST Addition on Sr Removal in SRS HLW Simulant.



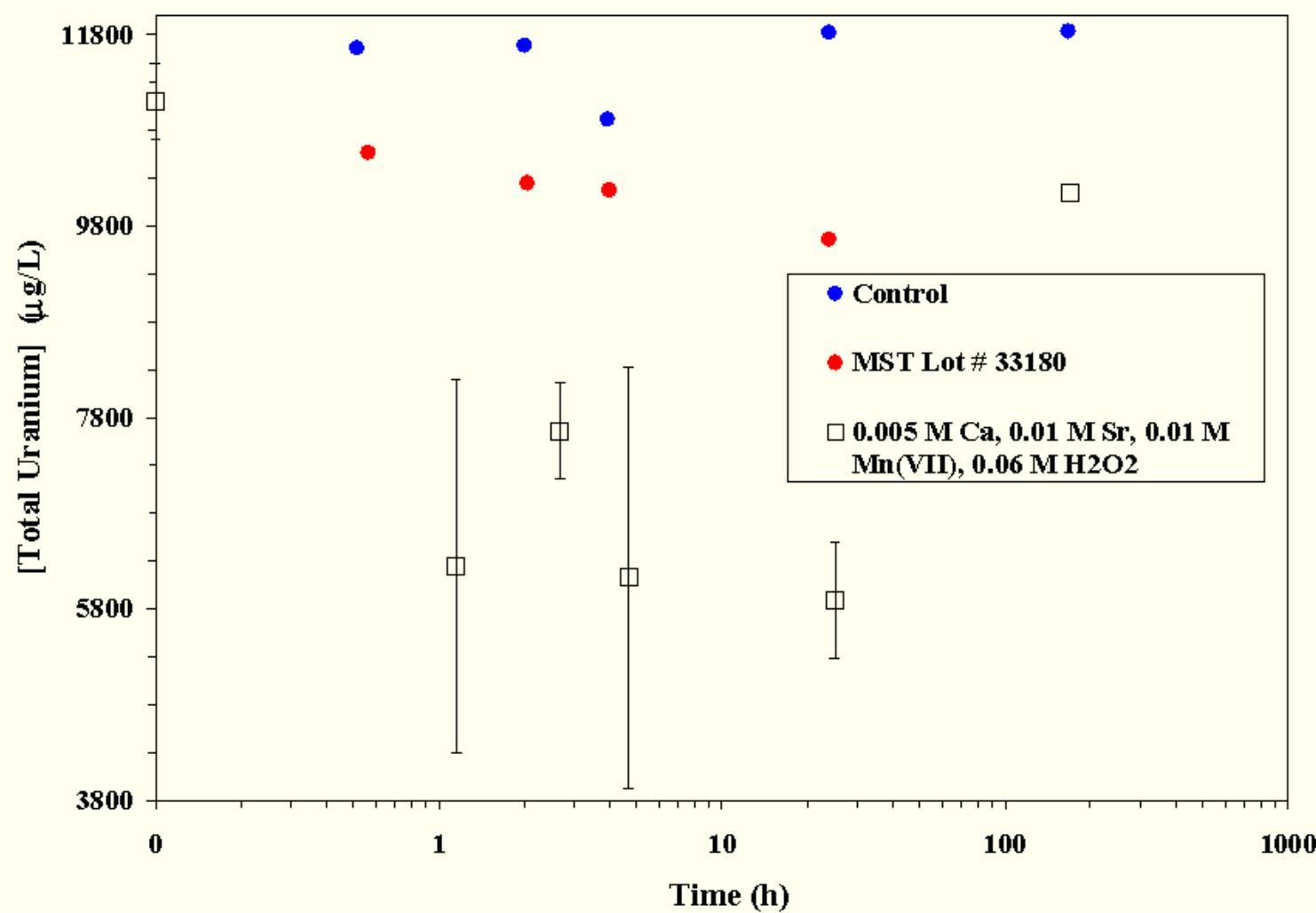
**Figure 4-8 Permanganate Treatment with Peroxide Versus MST Addition on Pu Removal in SRS HLW Simulant.**

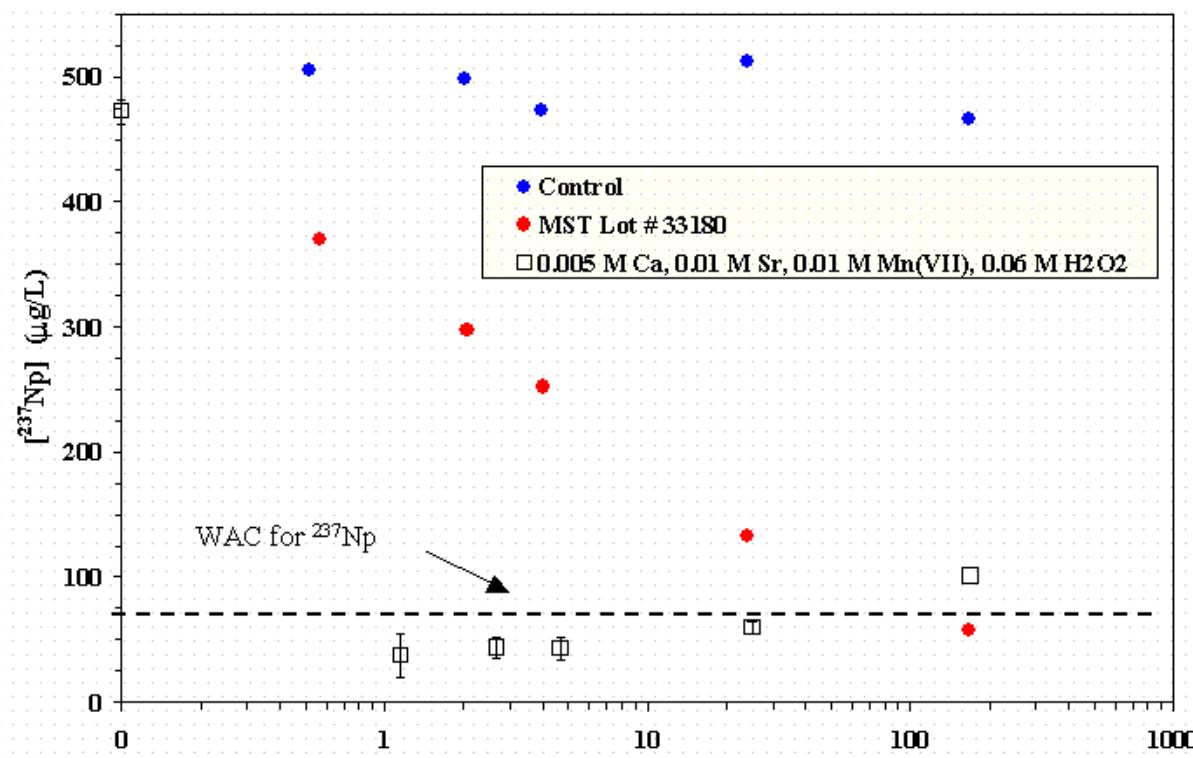
#### 4.2.3 Uranium

At 60 minutes and 24 hours of equilibration, our treatments with permanganate and peroxide (as a reductant) were more effective at U removal than that of MST addition (**Figure 4-9**). For example, nearly half of the U was removed from solution by permanganate treatment and roughly a tenth of the U was removed by MST addition during these 60-minute and 24-hour sampling periods. However, our data indicate removal of U by permanganate treatment was reversible in that after 168 hours, the U levels in solution were comparable to that defined with MST (**Figure 4-9**).

#### 4.2.4 Neptunium

Our ICP-MS data indicate faster Np removal in the permanganate treatment with peroxide as a reductant than with MST addition (**Figure 4-10**). The dissolved Np levels in the permanganate test indicate that the Saltstone WAC of  $53 \mu\text{g Np L}^{-1}$  was met after 60 minutes of equilibration and the levels of Np in this treatment remained lower than the WAC between 60 minutes and 4 hours and then increased with time to roughly  $100 \mu\text{g Np L}^{-1}$  after 168 hours of equilibration. In contrast, the test with MST addition had slower removal kinetics but the Np level in the tests met the WAC after 168 hours of treatment with MST (**Figure 4-10**).





**Figure 4-10 Permanganate Treatment with Peroxide Versus MST Addition on Np Removal in SRS HLW Simulant.**

#### 4.3 Effect of Reagent Order and Reductant Choice with 0.01 M Permanganate

The data presented in this section are from our second set of tests as described in **Section 3.2**.

##### 4.3.1 Strontium

We added several sources of Sr to our HLW simulant, which made our assessment of Sr removal complex. We added 0.01 M of stable seed  $\text{Sr}(\text{NO}_3)_2$  (containing  $^{84, 86, 87, 88}\text{Sr}$ ) to these tests and our simulants contained  $600 \mu\text{g L}^{-1}$  of (tramp) stable Sr. Additionally, our  $^{85}\text{Sr}$  spike solution contained an element of mass 84, which was probably  $^{84}\text{Rb}$  ( $^{84}\text{Rb}$  is the parent isotope or source material used to make the  $^{85}\text{Sr}$  that we used as our spike). This solution was fresh and contained more  $^{84}\text{Rb}$  than the  $^{85}\text{Sr}$  used in our first set of experiments in **Section 3.1**. Therefore, we could not make reliable total (stable) Sr determinations by ICP-MS analyses as in the first study. We made an approximation of Sr decontamination by examining the loss of  $^{85}\text{Sr}$  from our test solutions because this form of Sr was readily traceable.

In our simulant tests,  $^{85}\text{Sr}$  removal was generally unaffected by reagent order (**Figure 4-11**) and all of our treatments resulted in Sr DF values of 6.5 or less. Our permanganate tests with formate as a reductant show that treatment with formate may be slightly faster at Sr removal than treatment with peroxide as a reductant (**Figure 4-11**).

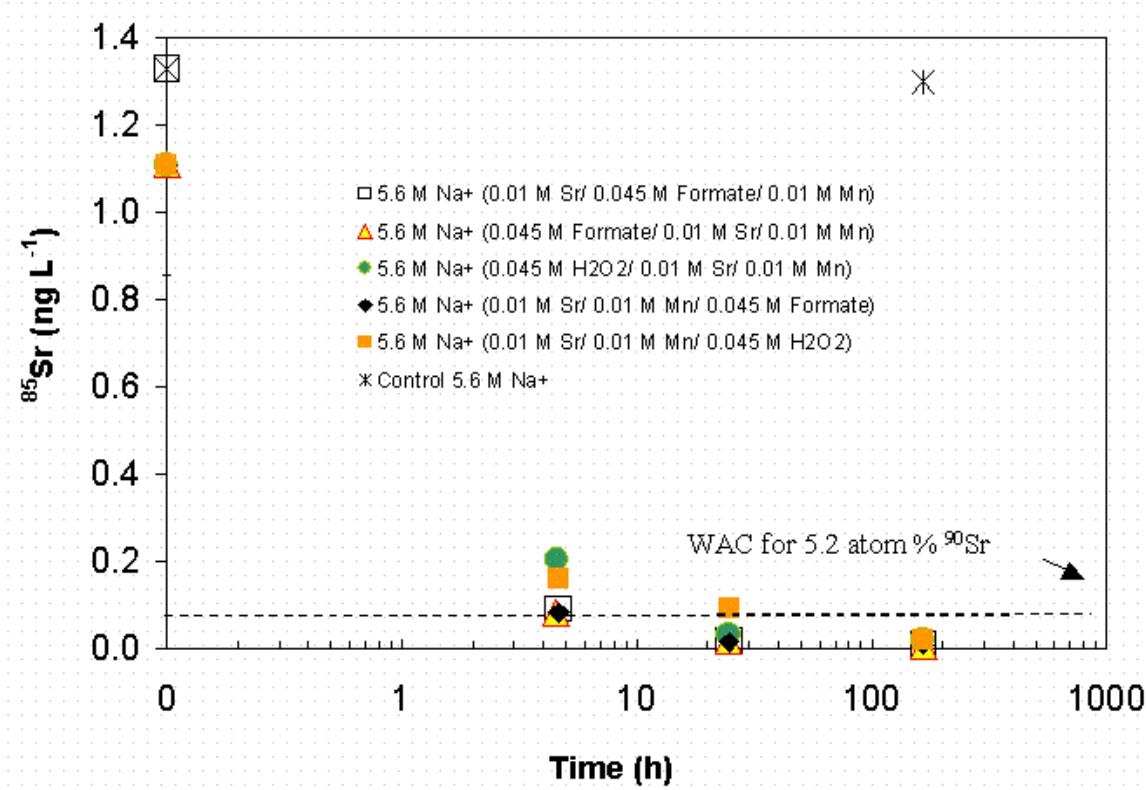
We defined what the Saltstone WAC would be based on a starting Sr level in our salt solutions of  $600 \mu\text{g Sr L}^{-1}$ . To meet the Saltstone WAC (at 5.2 atom %  $^{90}\text{Sr}$ , which is  $6.8 \mu\text{g total Sr L}^{-1}$ ), we would need to have nearly a 100-fold a reduction in the  $^{85}\text{Sr}$  level (of  $1.3 \text{ ng L}^{-1}$ ). To meet the Saltstone limit for 45 atom %  $^{90}\text{Sr}$  (as a worst case

scenario of  $0.79 \mu\text{g}$  total  $\text{Sr L}^{-1}$ ), the  $^{85}\text{Sr}$  level in our solutions would also need to be reduced by nearly 800 fold. Our calculated level for the Saltstone WAC (assuming 5.2 atom %  $^{90}\text{Sr}$ ) is shown as a dashed line on **Figure 4-11**. All of our formate treatments met the WAC for 5.2 atom %  $^{90}\text{Sr}$  after 30 minutes of equilibration. At 4 hours, the  $^{85}\text{Sr}$  levels in the formate treatments approximated the Saltstone WAC for 45 atom %  $^{90}\text{Sr}$  (**Figure 4-11**). In all treatments, Sr removal increased with time indicating removal was irreversible throughout our 168-hour study.

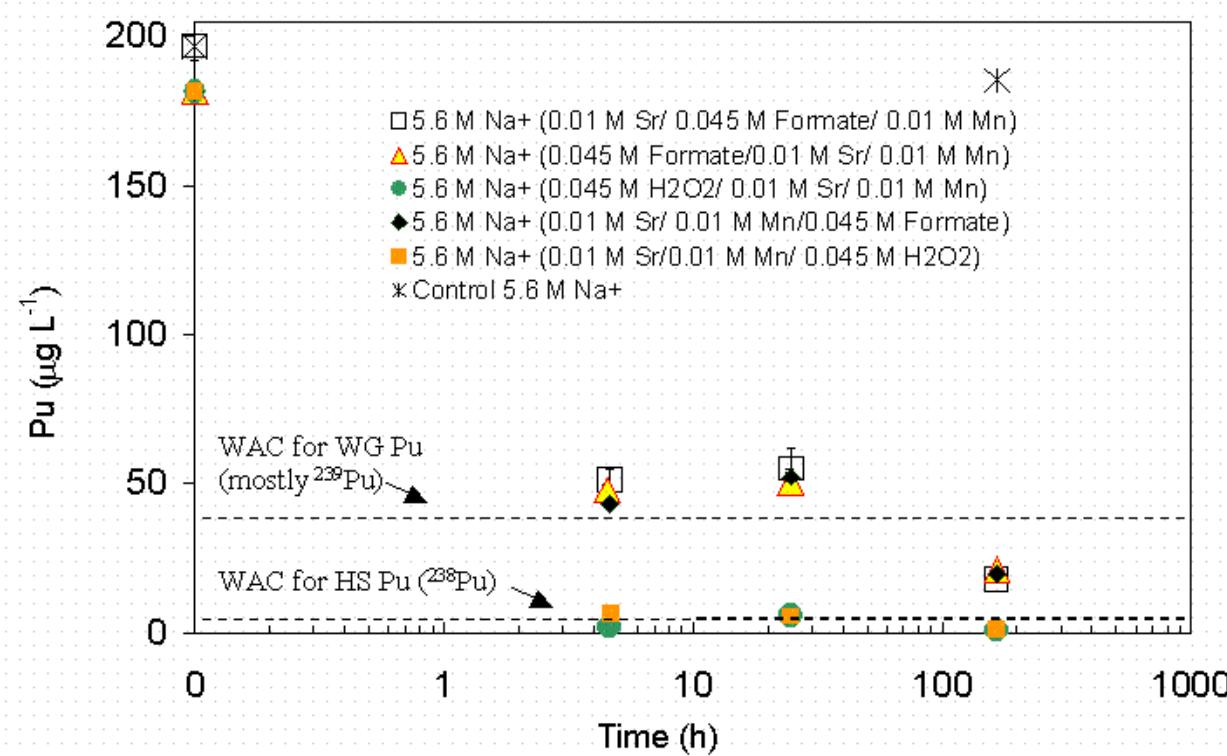
#### 4.3.2 Plutonium

**Figures 4-12** and **4-13** show the results of the Pu-TTA and Pu ICP-MS analyses for our permanganate tests that examined reagent order and reductant choice with permanganate treatment. In general, the two Pu analyses yielded similar data. However, the ICP-MS data for the 24 hour samplings were typically a few  $\mu\text{g L}^{-1}$  lower than that of our Pu-TTA data.

Our tests showed that Pu removal was faster in the salt solutions that received peroxide instead of formate as the reductant at permanganate levels of 0.01 M. The Pu removal was not strongly affected by reagent order (**Figure 4-12** and **Figure 4-13**). The Saltstone WAC for Pu at WG isotopic abundance was met by all treatments with peroxide as the reductant between sampled at 4 and 168 hours whereas the Pu levels with formate only met the WAC after 168 hours of equilibration. Clearly, greater removal occurs in the permanganate tests when peroxide is used as the reductant.



**Figure 4-11 Effect of Reagent Order and Reductant Choice on  $^{85}\text{Sr}$  Removal in SRS HLW Simulant.**



**Figure 4-12 Effect of Reagent Order and Reductant Choice on Pu Removal in SRS HLW Simulant Pu TTA Data.**

#### 4.3.3 Uranium

Our studies with U show that there was an affect of reagent order and reductant choice on U removal from the salt simulants (see **Figure 4-14**). In general, U removal was low when formate was added after the addition of 0.01 M Sr and when formate was added before the addition of permanganate. Our treatments with peroxide first, seed Sr second and permanganate last had a rapid and substantial loss of dissolved U after 30 minutes (**Figure 4-14**) in that nearly half of the initial U level remained in solution. When peroxide was added last, U removal was comparable to tests in which formate was added before or after permanganate (**Figure 4-14**). The levels of U in these solutions (regardless of whether formate or peroxide was added) were similar at 24 hours. The levels of U in all of our tests increased after 168 hours suggesting that U removal was reversible. There is no WAC concern for U in these tanks because the alpha activity of U in the SRS waste was much lower than that of Pu and Np.

The treatments with peroxide added first may have promoted reduction of U(VI) to the less soluble U(IV) species because of the absence of permanganate ion, which is likely to be more competitive for peroxide than U(VI). If this reduction of U(VI) occurs, it appears to be reversible as evidenced by the increase in dissolved U concentration with time.

#### 4.3.4 Neptunium

Our tests show that Np removal was greater when peroxide was added as a reductant than when formate was added as a reductant (**Figure 4-15**). When peroxide was added prior to permanganate, Np removal was greater than when peroxide was added after permanganate at 30 minutes. The peroxide (added) first treatment met the Saltstone WAC of 53  $\mu\text{g Np L}^{-1}$  but only during the first 4 hours. After that period, none of our tests shown in **Figure 4-15** met the Saltstone PL. The treatments with peroxide added may have promoted reduction of Np(V) to the less soluble Np(IV) species (although permanganate might be more competitive for peroxide than Np). If this reduction of Np(V) occurs, it appears to be reversible as evidenced by the increase in dissolved Np concentrations with time.

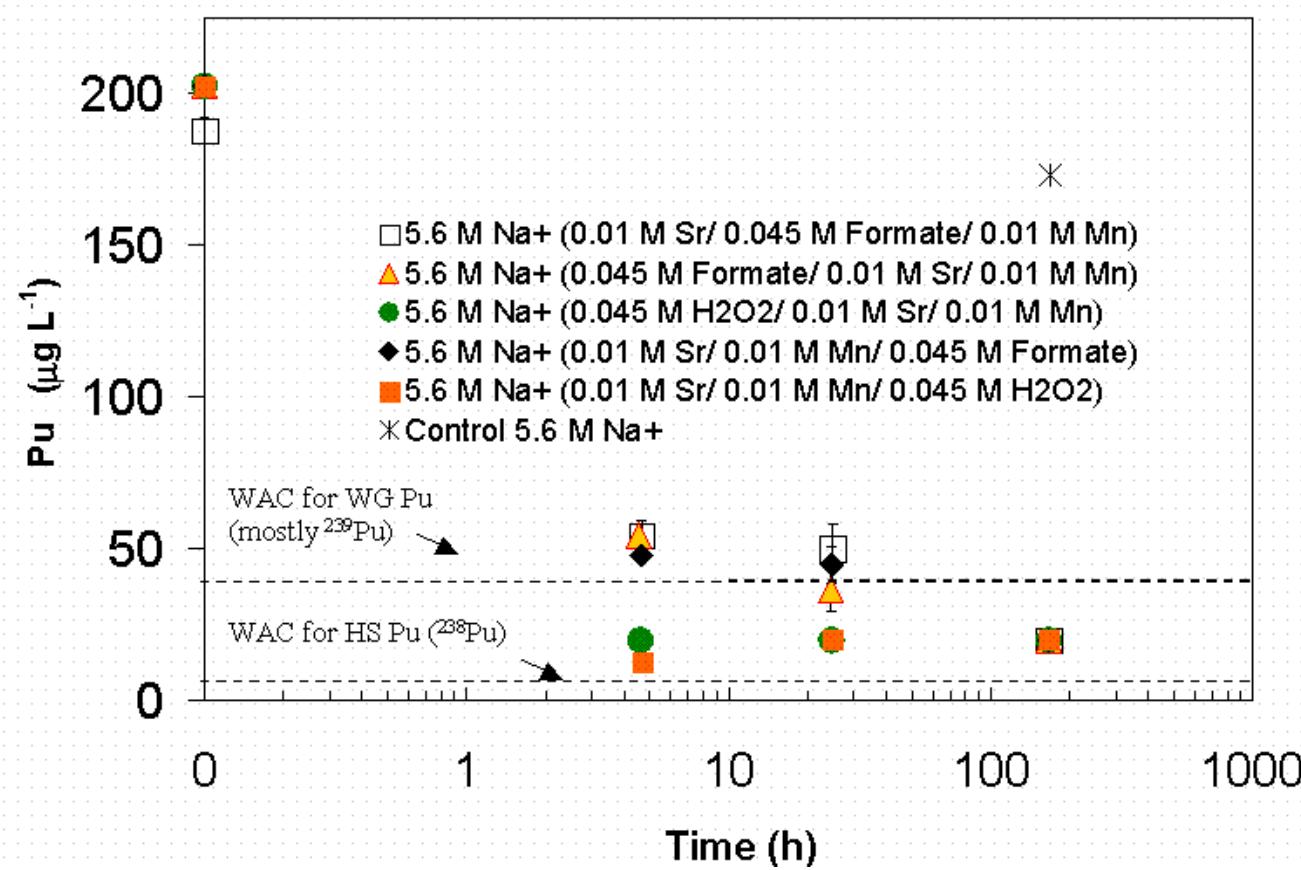
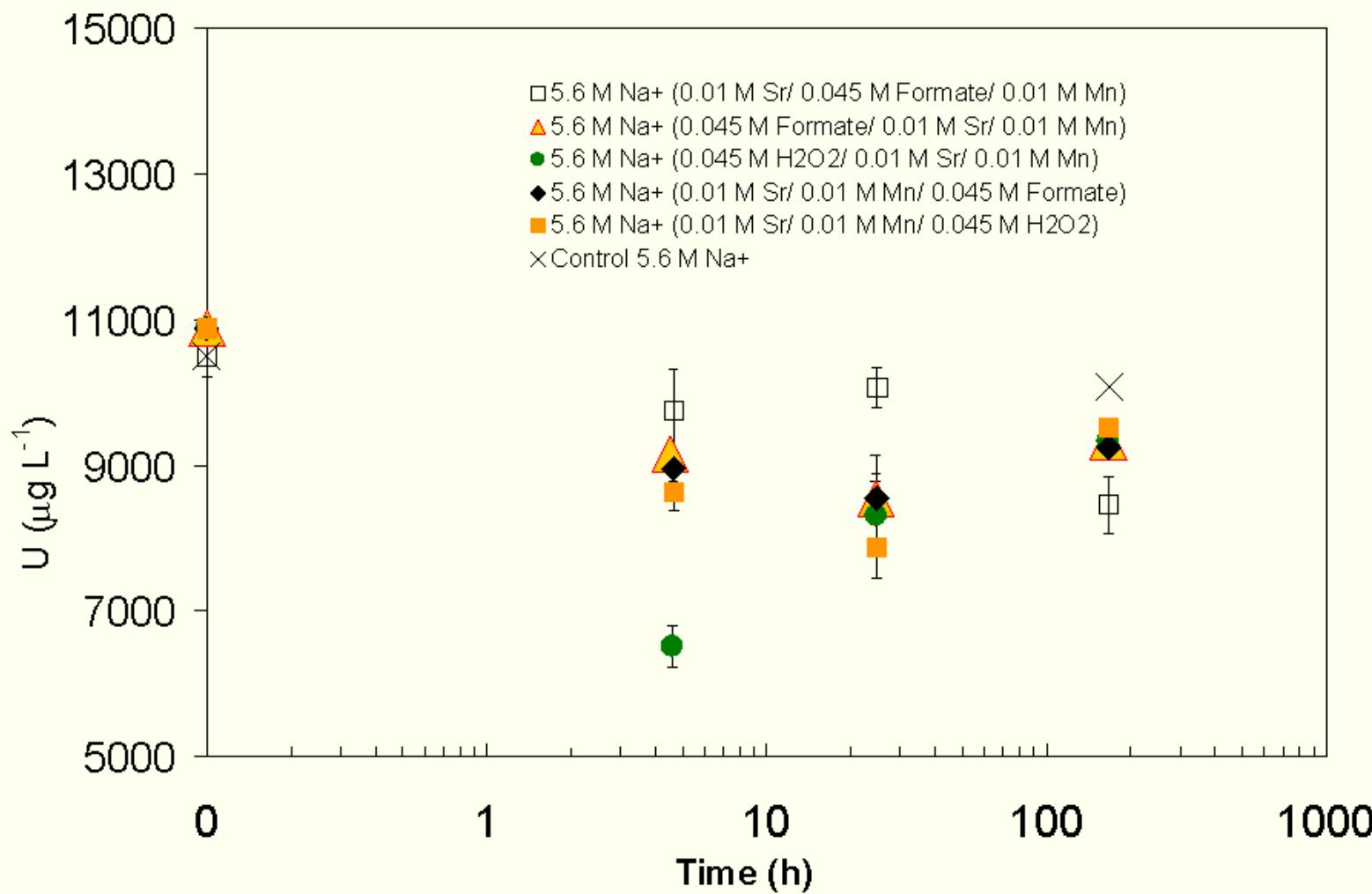


Figure 4-13 Effect of Reagent Order and Reductant Choice on Pu Removal in SRS HLW Simulant ICP-MS Data.



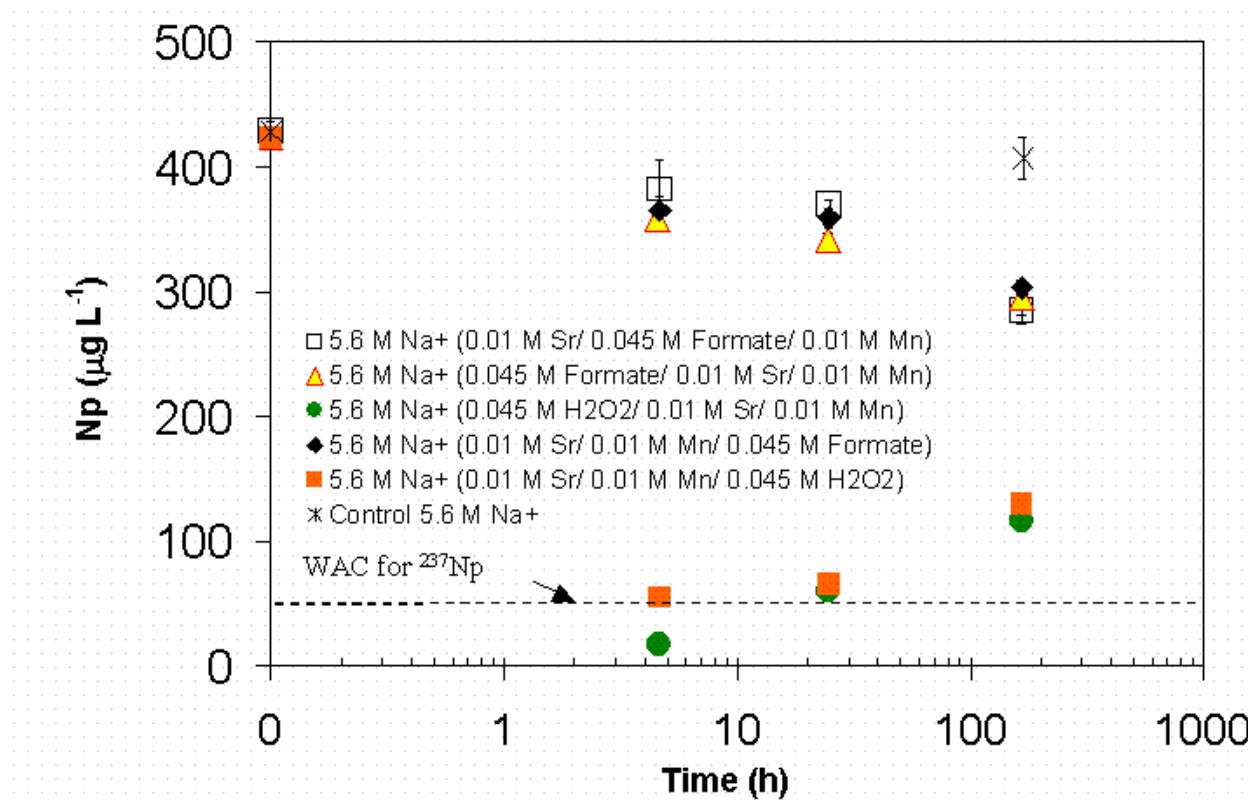
**Figure 4-14 Effect of Reagent Order and Reductant Choice on U Removal in SRS HLW Simulant.**

#### 4.3.5 Stable Strontium

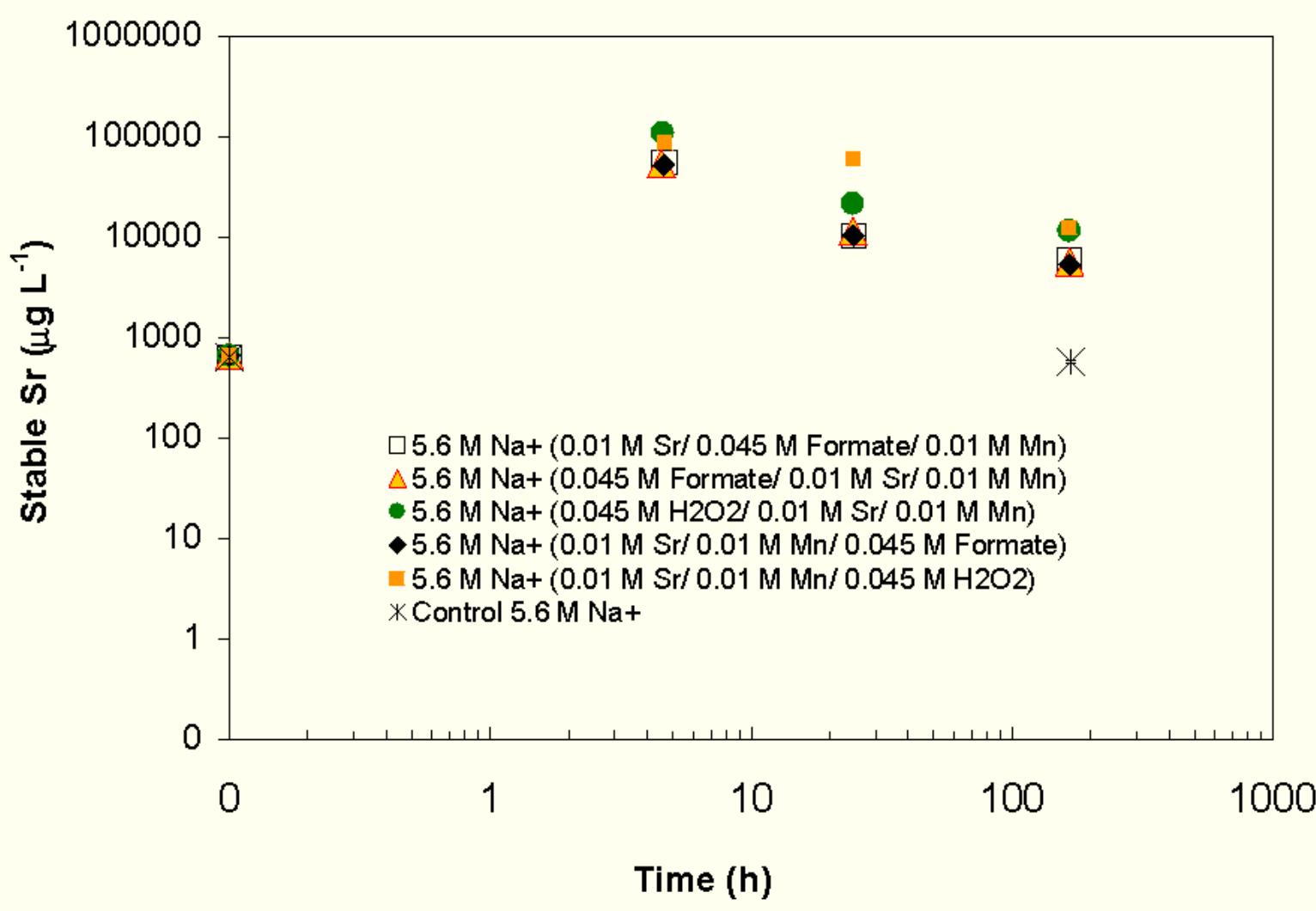
Our  $^{85}\text{Sr}$  spike contained some  $^{84}\text{Rb}$  and the  $^{84}\text{Rb}$  concentration varied from solution to solution. Our  $^{85}\text{Sr}$  spike also contained an isotopic distribution of Sr (assuming the masses of 86, 87 and 89 were due to Sr). These observations complicated our ability to quantify the total (stable) Sr in our samples based on the assumption of a natural distribution of stable isotopes of Sr. Therefore, we only reported the ICP-MS values for elements with masses of 86, 87 and 89 (i.e., the most common Sr isotopes). This assumption includes nearly all of the stable Sr in our samples because  $^{84}\text{Sr}$  is at a low natural abundance of 0.56 atom %.

The concentration of seed Sr (0.01 M Sr) that we originally added was  $\sim 900,000 \mu\text{g Sr L}^{-1}$  and this Sr level far exceeded the levels that we typically observe for dissolved Sr in HLW salt solutions. Therefore, we expected a decrease in the stable Sr concentrations over time due to precipitation and removal of Sr from solution (as in **Figure 4-16**).

The stable Sr concentrations in our tests were higher in the tests that contained peroxide than in the tests with formate (**Figure 4-16**). This suggests that there was an influence of formate on precipitation. The greater rate of Sr removal may be due to decomposition of formate to  $\text{CO}_2$  and  $\text{H}_2$ . Evolution of  $\text{CO}_2$  from formate decomposition may have increased the carbonate concentrations in solution to levels that were higher than those likely to occur in the peroxide treatments and thus favored  $\text{SrCO}_3(s)$  precipitation. [ $\text{SrCO}_3(s)$  precipitation has been observed in studies with Hanford HLW simulant waste]. The low initial levels of carbonate ion (0.026 M) in our salt simulant may have not been high enough to promote  $\text{SrCO}_3(s)$  precipitation. After 168 hours of equilibration, the Sr levels in all of the test solutions were below 20,000  $\mu\text{g L}^{-1}$  indicating a substantial amount of Sr precipitation (as possibly as  $\text{SrCO}_3(s)$  phase) occurred in these treatments.



**Figure 4-15 Effect of Reagent Order and Reductant Choice on Np Removal in SRS HLW Simulant.**



**Figure 4-16 Effect of Reagent Order and Reductant Choice on Stable Sr Concentration in SRS HLW Simulant.**

#### 4.4 Effect of Reductant Concentration at 0.013 M Permanganate Concentration

The data presented in this section are from our first set of tests as described in **Section 0**.

##### 4.4.1 Strontium

The results of our tests that examined the effect of formate concentration at 0.013 M permanganate and 0.01 M Sr concentrations are shown in **Figure 4-17**. Our tests show that Sr removal was generally unaffected by formate concentration. Loss of Sr was slightly greater in the high formate treatments, but these differences were within the error

of the measurements (**Figure 4-17**). Almost all of the tests met the Saltstone WAC for 5.2 atom %  $^{90}\text{Sr}$  and several of the tests with  $>0.04$  M formate met the Saltstone WAC for 45 atom %  $^{90}\text{Sr}$  at 30 minutes, 24 and 168 hours (**Figure 4-17**).

#### 4.4.2 Plutonium

Our tests show that Pu removal was faster in the salt solutions that received high formate concentrations (at permanganate levels of 0.013 M) than with solutions that received lower formate concentrations (**Figure 4-18**). The Saltstone WAC for Pu at WG abundance was met in all of our  $>0.04$  M formate treatments and in all of our 0.04 M formate treatments between 2 and 168 h of equilibration. The dissolved Pu concentrations in the solutions that received  $> 0.04$  M formate decreased after 4 hours indicating that Pu removal was reversible (**Figure 4-18**). The results indicate that reductant (formate) levels should be higher than 0.04 M to meet WACs within the first few hours of equilibration.

#### 4.4.3 Uranium

Our tests show that U removal was faster in the salt solutions that received high formate ( $>0.04$  M) concentrations at permanganate levels of 0.013 M than with solutions that received lower formate concentrations (**Figure 4-19**). Our studies show that U removal was greatest after 24 hours but that U removal was reversible because the U levels increased to levels greater than that of the control U concentrations after 168 hours of equilibration.

#### 4.4.4 Neptunium

Our studies show that Np removal was also influenced by formate concentration. Neptunium removal rates were much greater in the salt solutions that received high formate ( $>0.04$  M) concentrations at permanganate levels of 0.013 M than in salt solutions that received lower formate concentrations at similar permanganate levels (**Figure 4-20**). At lower formate levels, there was a slight correlation of formate concentration with Np removal. Our studies show that Np removal was greatest after 24 hours and this treatment met the Saltstone WAC for  $^{237}\text{Np}$ . The removal of Np in our tests was slightly reversible because the Np levels increased somewhat after 168 hours of equilibration (**Figure 4-20**).

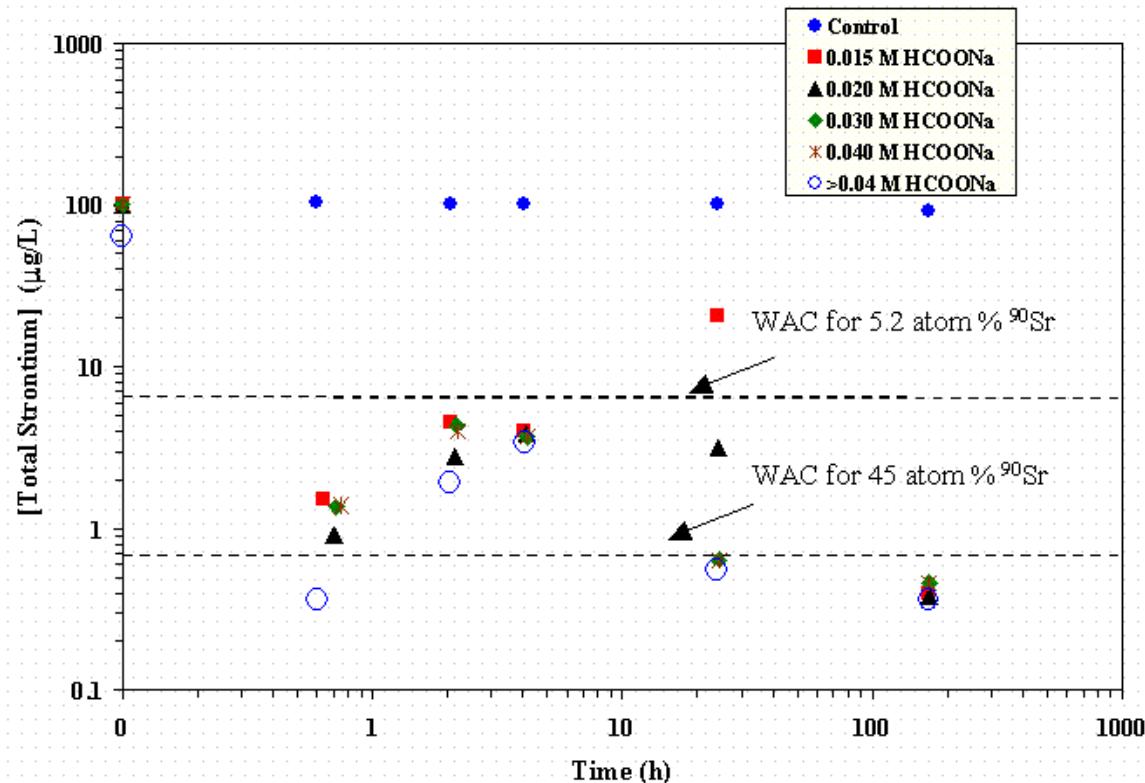
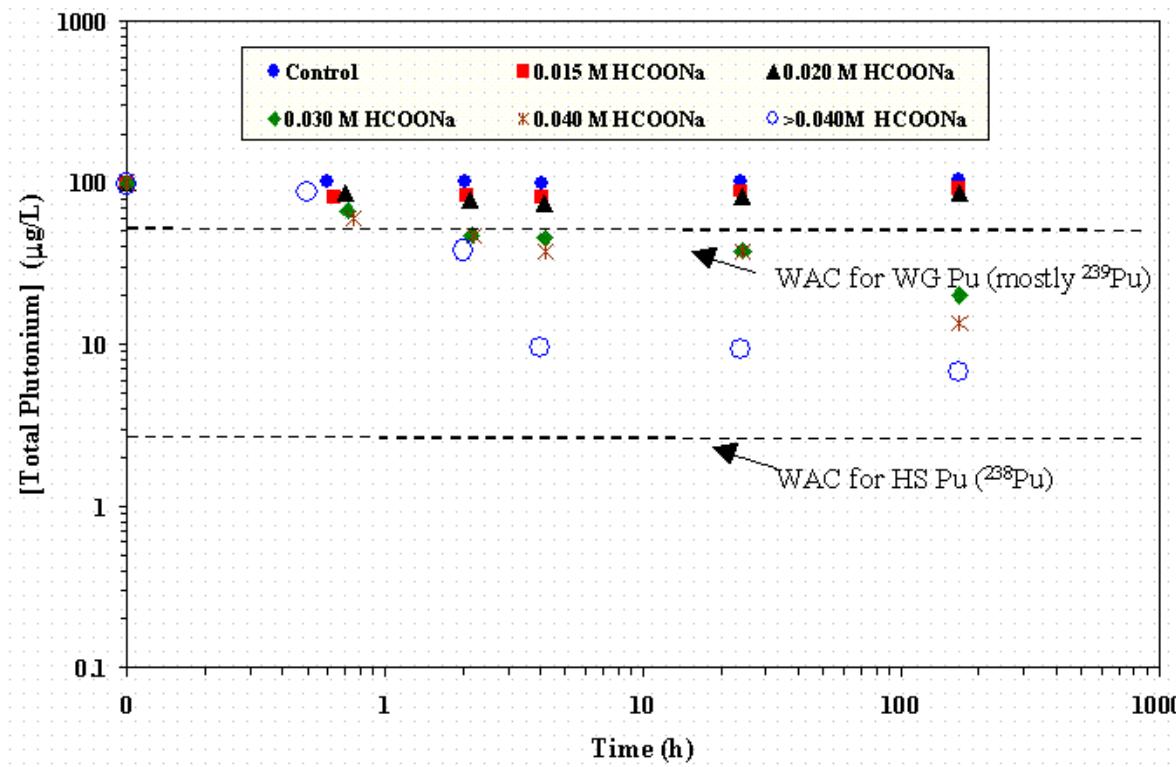


Figure 4-17 Effect of Reductant Concentration with 0.013 M Permanganate on Sr Removal in SRS HLW Simulant ICP-MS Data.



**Figure 4-18 Effect of Reductant Concentration with 0.013 M Permanganate on Pu Removal in SRS HLW Simulant PuTTA Data.**

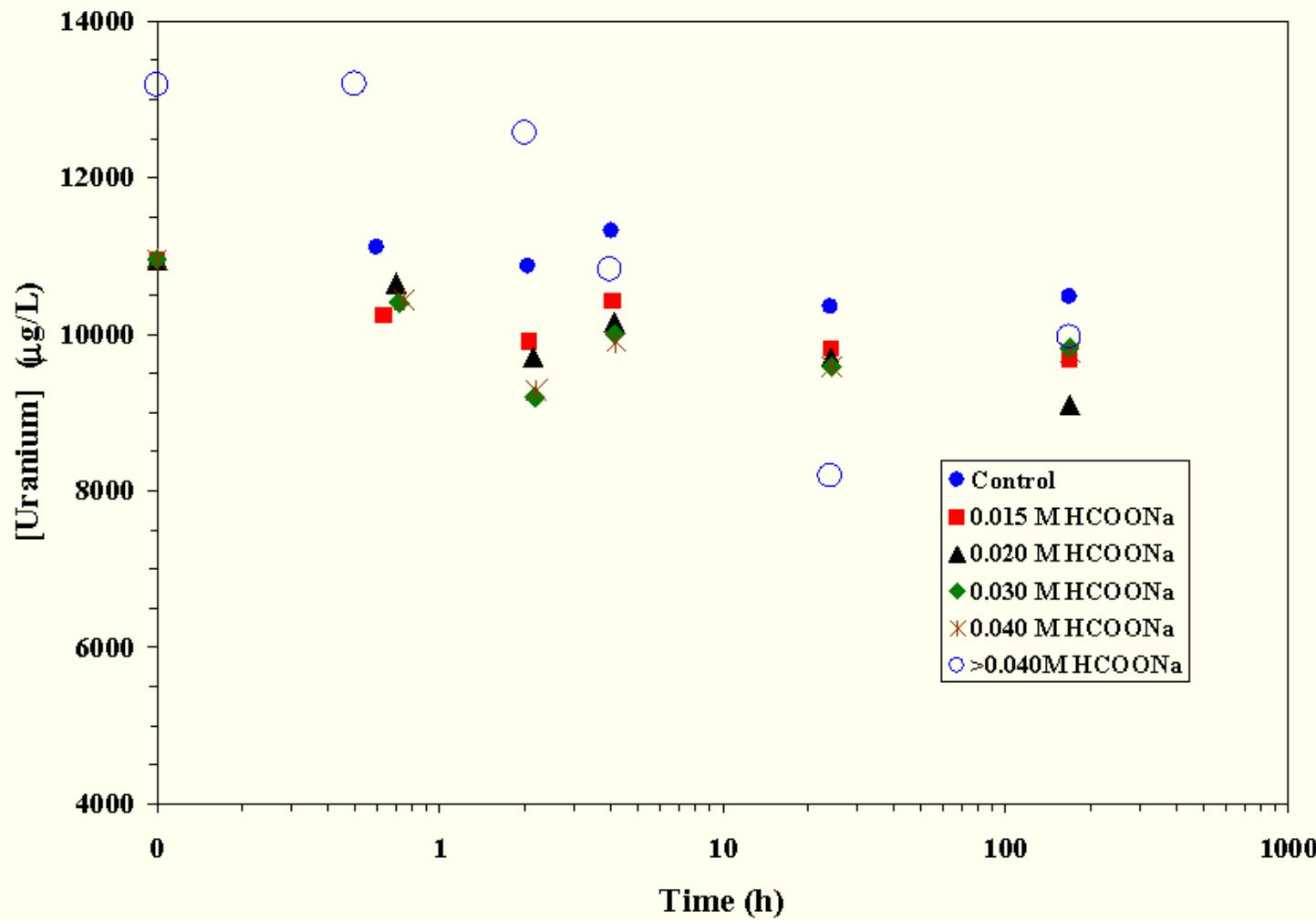
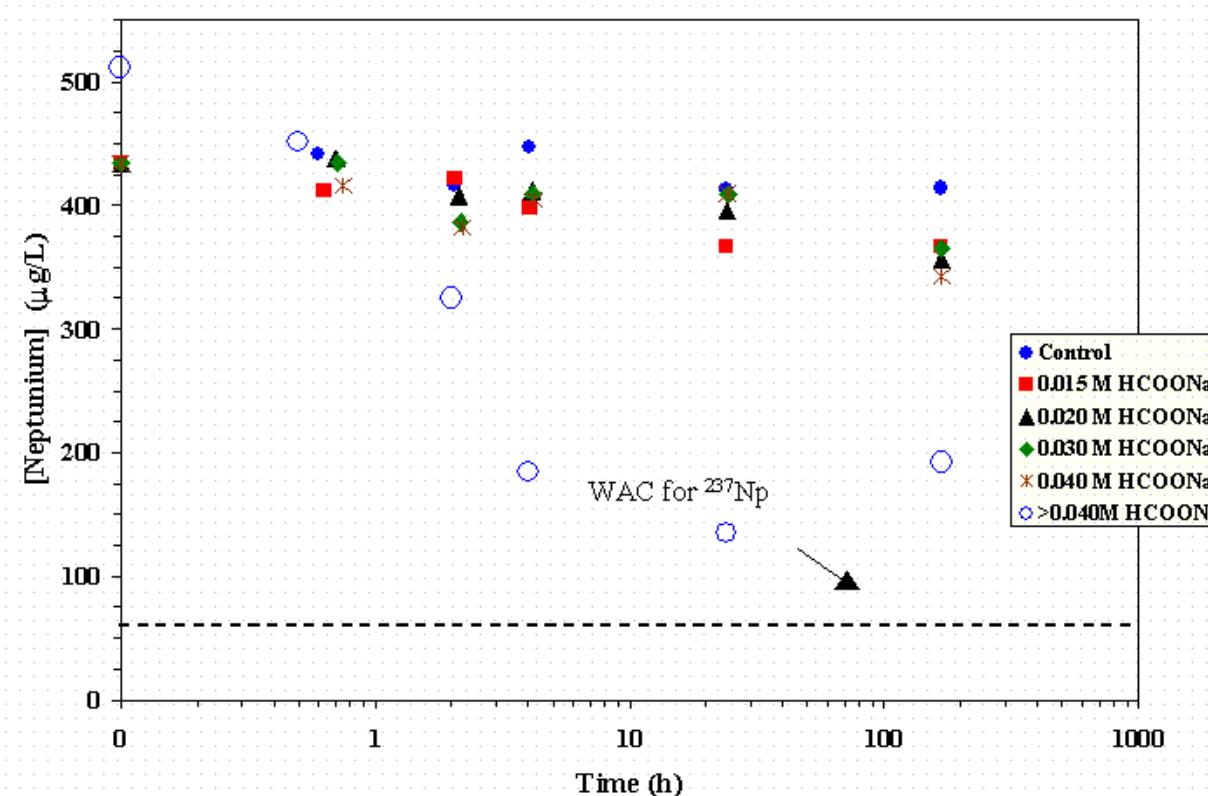


Figure 4-19 Effect of Reductant Concentration with 0.013 M Permanganate on U Removal in SRS HLW Simulant.



**Figure 4-20 Effect of Reductant Concentration with 0.013 M Permanganate on Np Removal in SRS HLW Simulant.**

#### 4.5 Effect of Permanganate Concentration at 0.015 M Formate Concentration

The data presented in this section are from our first set of tests as described in **Section 0**.

##### 4.5.1 Strontium

The results of our tests that examined the effect of permanganate ion concentration at 0.015 M formate are shown in **Figure 4-21**. Our tests show that Sr removal was related to permanganate concentration between 30 minutes and 4 hours of equilibration and both of these treatments met the Saltstone WAC for 5.2 atom %  $^{90}\text{Sr}$  (of 6.8  $\mu\text{g Sr L}^{-1}$ ). Our tests showed that Sr concentrations increased in the high permanganate treatment (0.013 M) and after 4 hours the Sr levels exceeded the Saltstone WAC for 5.2 atom %  $^{90}\text{Sr}$  (**Figure 4-21**). The test with 0.0013 M permanganate met the Saltstone WAC for 45 atom. %  $^{90}\text{Sr}$  after 168 hours.

##### 4.5.2 Plutonium

Our tests with Pu show that Pu removal was not strongly related to permanganate concentration (**Figure 4-22**) due to the low levels of formate that were added. Treatments with high permanganate levels did not have enough formate to facilitate reduction of Mn(VII) and precipitation of all of the added Mn(VII) as a Mn(III, IV) oxide phases. Our treatment with 0.0013 M and 0.0065 M permanganate and 0.015 M formate almost met the Saltstone WAC for Pu with WG isotopes after 168 hours of equilibration (**Figure 4-22**). The removal of Pu from solution was slow and it continued between 2 and 168 hours indicating that the decontamination process was not reversible during these time periods. Little removal of Pu was observed in the 0.013 M permanganate treatment.

#### 4.5.3 Uranium

Our tests show that U removal was greater than the control and the 0.0013 M permanganate test when the permanganate concentration was 0.065 M and 0.013 M (Figure 4-23). Our studies show that the U concentrations in the 0.0013 M permanganate treatment were often higher than our control (Figure 4-23) suggesting that some loss of U occurred in the control during our tests.

#### 4.5.4 Neptunium

The removal of Np from our solutions was not influenced by permanganate concentration in at 0.015 M formate concentrations and none of our treatments met the Saltstone limit for Np (Figure 4-24).

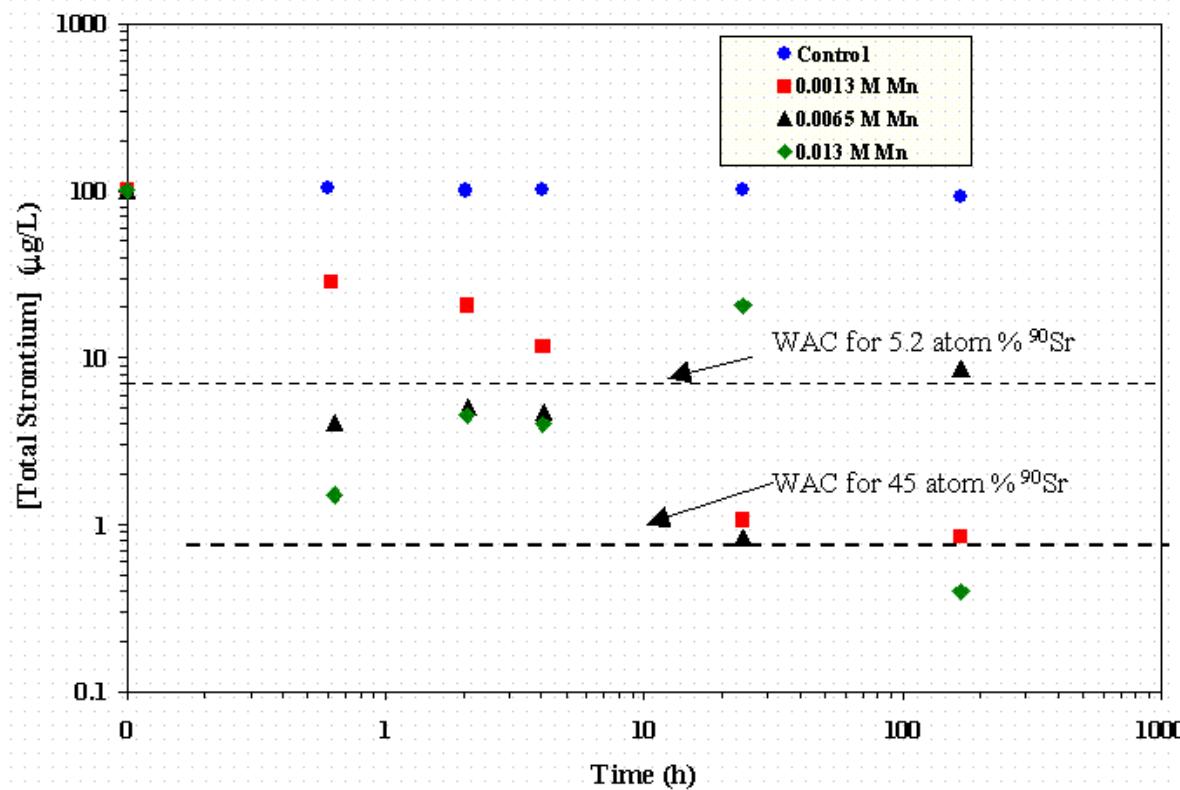


Figure 4-21 Effect of Permanganate Concentration with 0.015 M Formate on Sr Removal in SRS HLW Simulant ICP-MS Data.

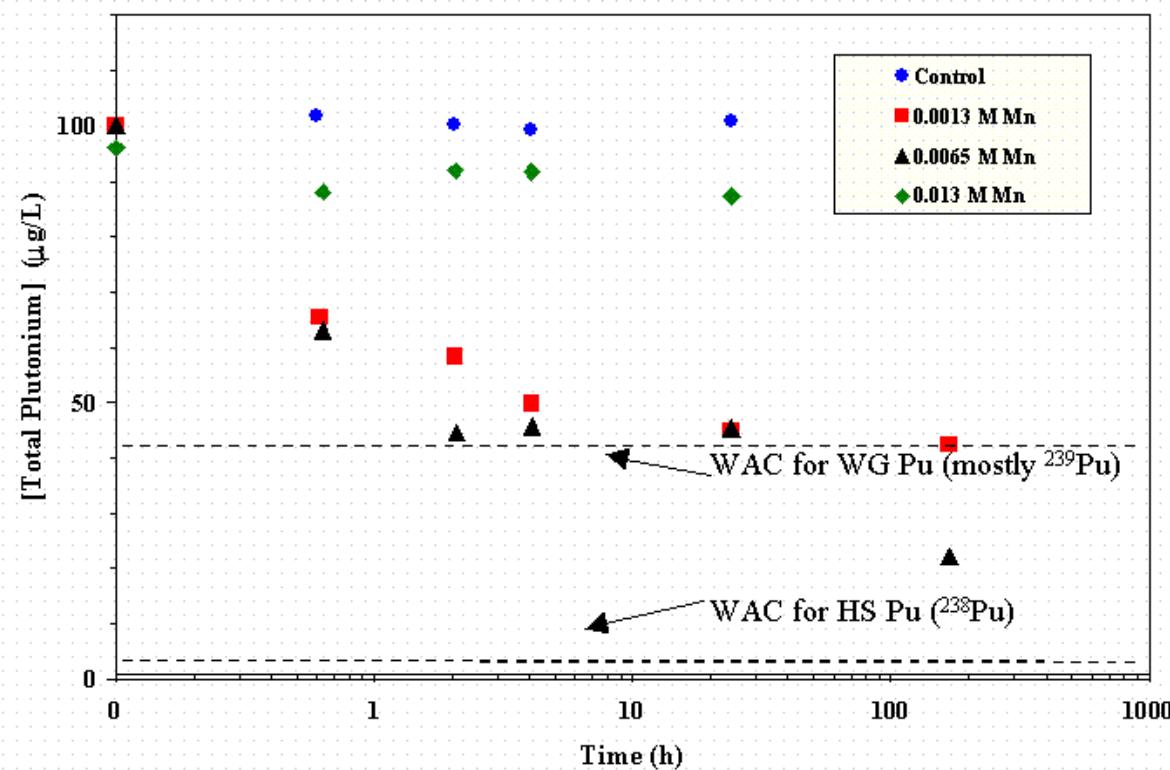


Figure 4-22 Effect of Permanganate Concentration with 0.015 M Formate on Pu Removal in SRS HLW Simulant  $\blacktriangleleft$  PuTTA Data.

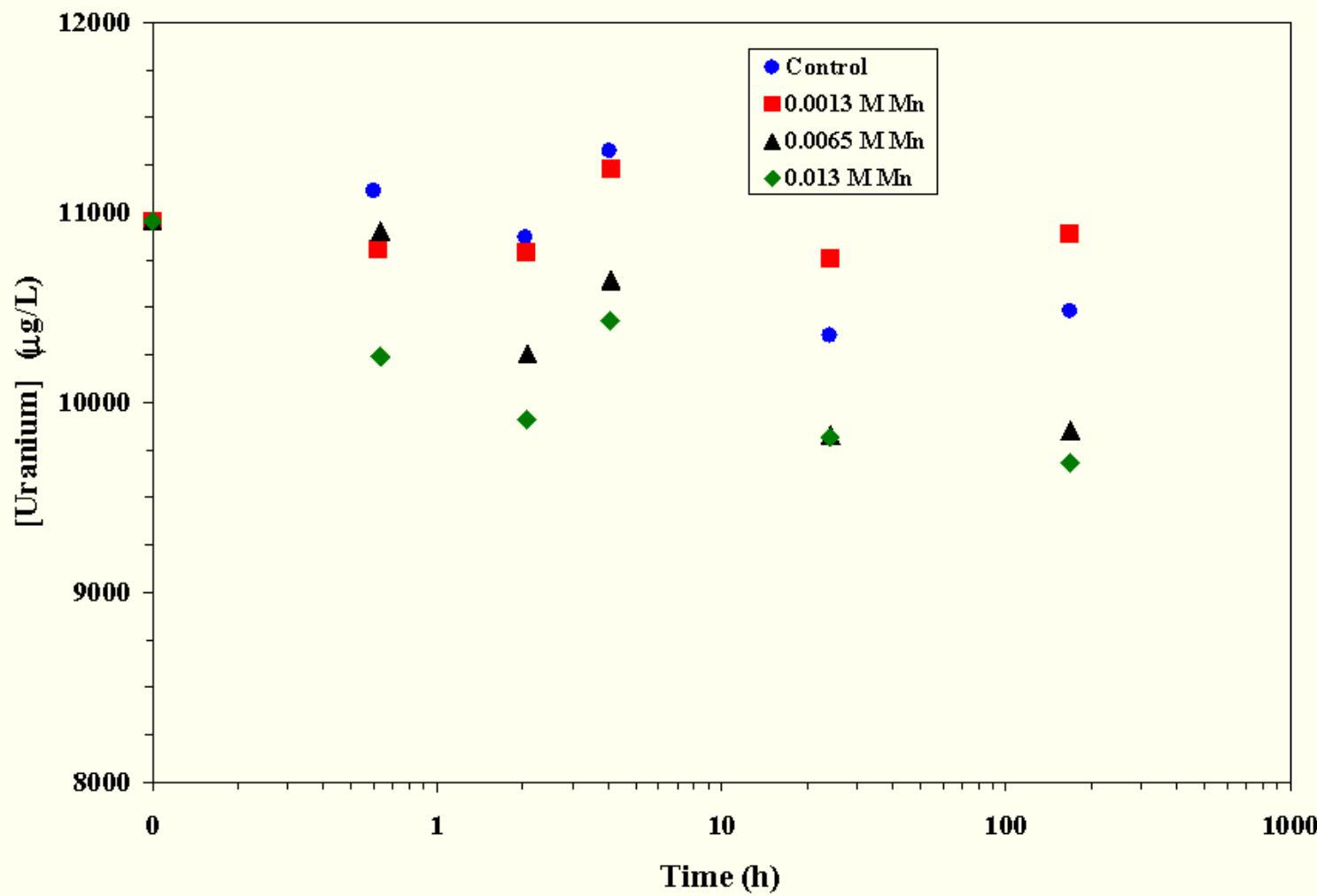
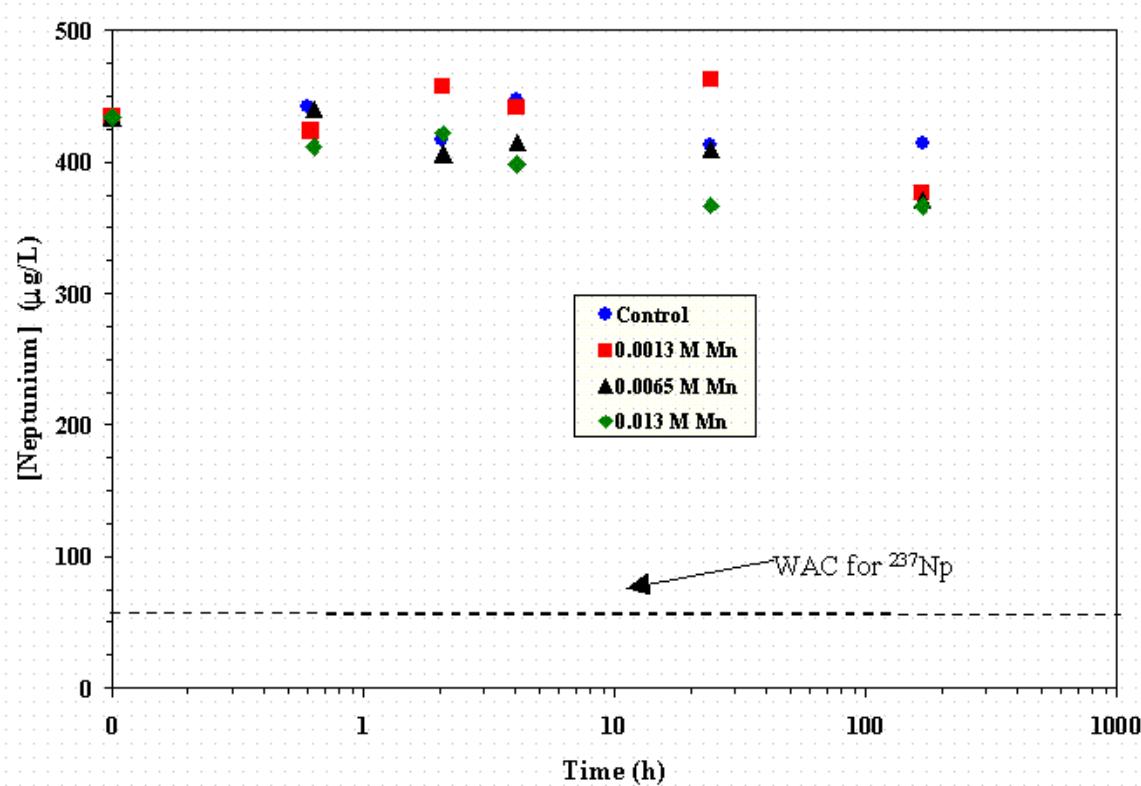


Figure 4-23 Effect of Permanganate Concentration with  $0.015 \text{ M}$  Formate on U Removal in SRS HLW Simulant.



**Figure 4-24 Effect of Permanganate Concentration with 0.015 M Formate on Np Removal in SRS HLW Simulant.**

#### 4.6 Effect of Permanganate Concentration and Permanganate Addition (Sequential vs. Single) at 0.045 M Formate Concentration

The data presented in this section are from our second set of tests as described in Section 0

##### 4.6.1 Strontium

As previously mentioned, we added several sources of Sr to our HLW simulant, which made our assessment of Sr removal based on total Sr complex. We could not make reliable total (stable) Sr determinations by ICP-MS analyses as in our first set of experimental tests as listed in Section 3.1. Therefore, we made an approximation of Sr decontamination by examining the loss of  $^{85}\text{Sr}$  from our test solutions because this form of Sr was readily traceable.

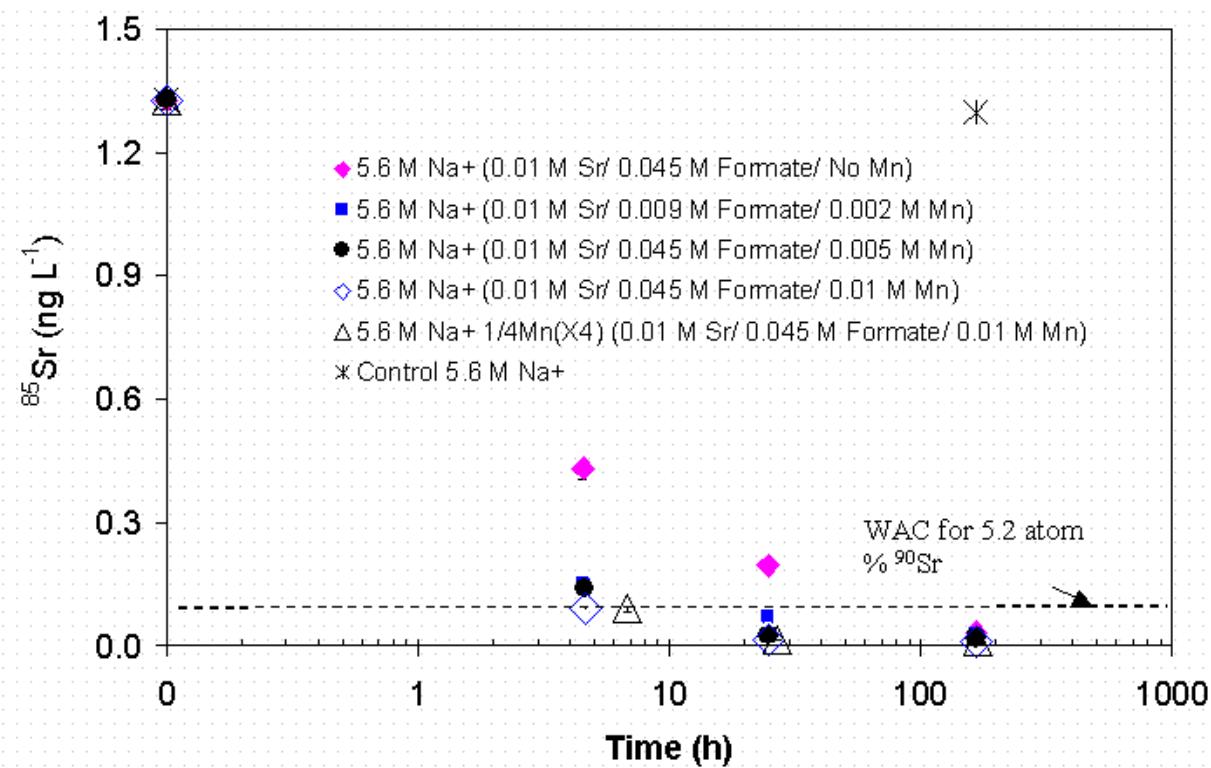
In our simulant tests with formate as the sole reductant,  $^{85}\text{Sr}$  removal was greatest in the treatments with the highest permanganate levels (at 0.01 M permanganate as shown in **Figure 4-25**). Additionally Sr removal was high in these treatments regardless of addition method [i.e., whether the permanganate was added in a single addition or in four smaller (sequential) additions]. These 0.01 M permanganate treatments met the Saltstone WAC for 5.2 atom %  $^{90}\text{Sr}$  and 45 atom %  $^{90}\text{Sr}$  (as a worst case scenario) after 24 hours. Our calculated level for the Saltstone WAC (assuming 5.2 atom %  $^{90}\text{Sr}$ ) is shown as a dashed line on **Figure 4-25**.

##### 4.6.2 Plutonium

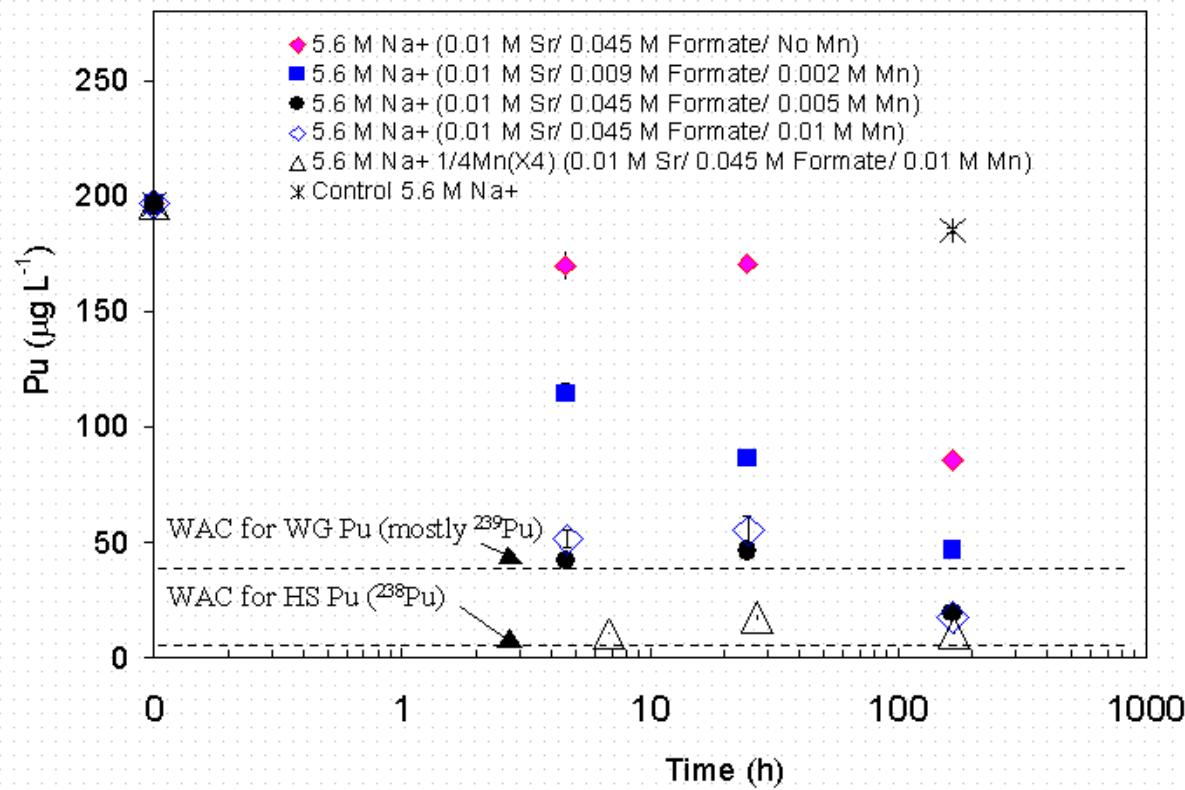
**Figures 4-26 and 4-27** show the results of the Pu-TTA and Pu ICP-MS analyses for the permanganate tests that examined reagent order and reductant choice. In general, our two methods of Pu analyses yielded similar values for dissolved Pu in our tests.

Our tests show that Pu removal was faster in the salt solutions that received sequential additions of permanganate at formate levels of 0.045 M than in salt solutions that received single additions of permanganate at 0.045 M formate concentrations (Figure 4-26 and Figure 4-27). After 4 h of equilibration, our treatments with sequential permanganate addition met the Saltstone limits for Pu at WG isotopic abundance and approached the Saltstone limits for Pu at HS isotopic abundance. Additionally, the low levels of Pu that occurred with sequential permanganate additions persisted after 168 hours of equilibration indicating that the Pu removal process was not reversible during our experimental study period.

Sequential addition may be more effective due to better mixing and several small additions of permanganate may promote the formation of a more crystalline or more ordered Mn oxide phase.



**Figure 4-25 Effect of Permanganate Concentration and Permanganate Addition Method on  $^{85}\text{Sr}$  Removal in SRS HLW Simulant.**



**Figure 4-26 Effect of Permanganate Concentration and Permanganate Addition Method on Pu Removal in SRS HLW Simulant◆Pu TTA Data.**

#### 4.6.3 Uranium

Our studies with U show there was little effect of permanganate concentration and permanganate addition method on U removal from the salt simulants at 0.045 M formate levels (see **Figure 4-28**). In general, U removal was low regardless of treatment condition for all sampling periods. The tests had similar U concentrations to that of the control studies.

#### 4.6.4 Neptunium

Our tests show that Np removal was slightly faster in the salt solutions that received sequential or single additions of permanganate (with a total addition of 0.01 M permanganate) at formate levels of 0.045 M than in salt solutions that received lower levels of permanganate at similar formate concentrations (**Figure 4-29**). Our treatments with sequential permanganate addition proved comparable to single permanganate additions. Our data indicate that Np removal increased with time but none of our treatments met the Saltstone limits for Np during the 168-hour study (**Figure 4-29**).

#### 4.6.5 Stable Strontium

As previously mentioned, the concentration of seed Sr (0.01 M Sr) that we added ( $\sim 900,000 \mu\text{g Sr L}^{-1}$ ) far exceeded dissolved Sr levels that are typically observed in HLW salt solutions. Therefore, we expected to see a substantial decrease in the stable Sr concentrations over time due to precipitation and removal of the seed Sr from solution (as in **Figure 4-30**).

The stable Sr concentrations in our tests were higher in the tests that contained no permanganate than in the tests which did (**Figure 4-30**). Our observations suggest that there may be a synergistic influence of permanganate on Sr precipitation in the presence of formate (as opposed to peroxide) as a reductant. The greater rate of Sr removal in the presence of permanganate may be due to decomposition of formate to  $\text{CO}_2$  and  $\text{H}_2$ , which may be facilitated by a redox reaction with the permanganate ion. Evolution of  $\text{CO}_2$  from formate decomposition may have increased the carbonate levels in solution to levels that were higher than that of the peroxide treatments and thus  $\text{SrCO}_3(s)$  precipitation became favored. The low levels of carbonate ion (0.026 M) in our salt simulant may have been too low to favor rapid (or any)  $\text{SrCO}_3(s)$  precipitation. After 168 hours of equilibration, the Sr levels in all of the test solutions were less than 20,000  $\mu\text{g L}^{-1}$  indicating a substantial amount of Sr precipitation (as possibly a  $\text{SrCO}_3(s)$  or a Sr hydroxide phase) occurred in these treatments (**Figure 4-30**).

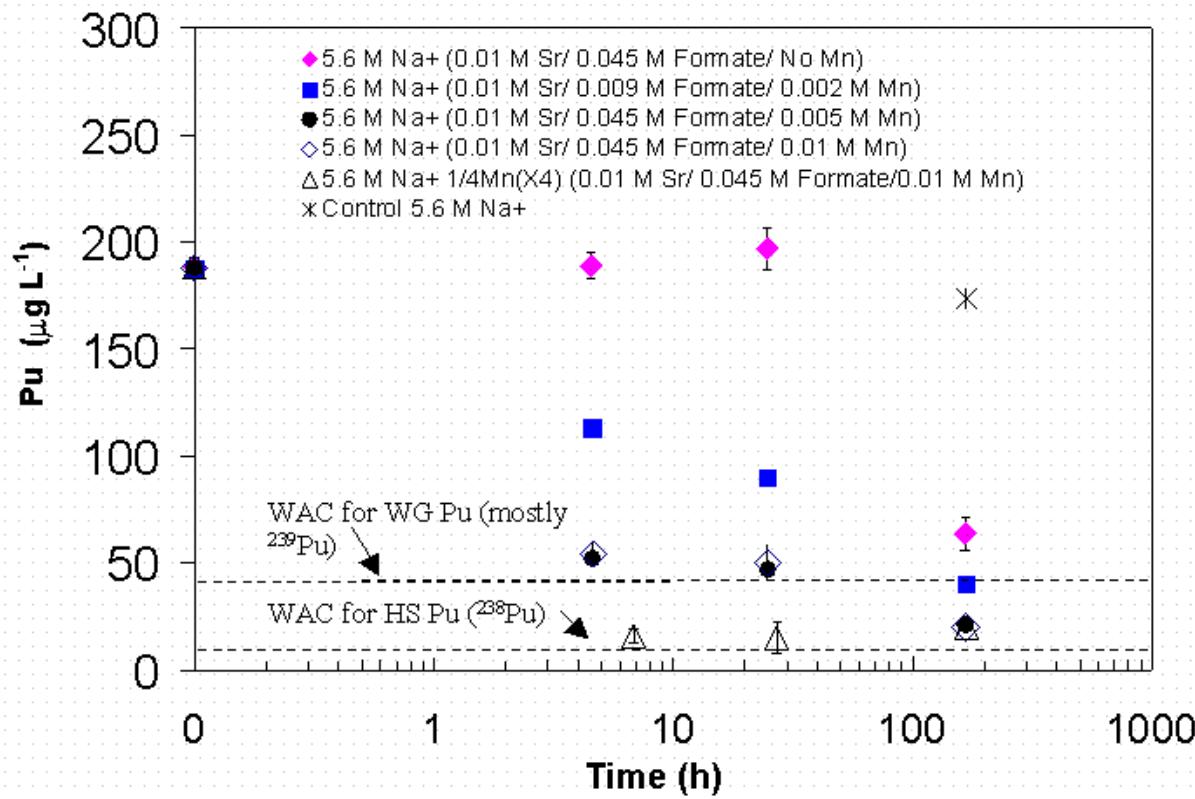
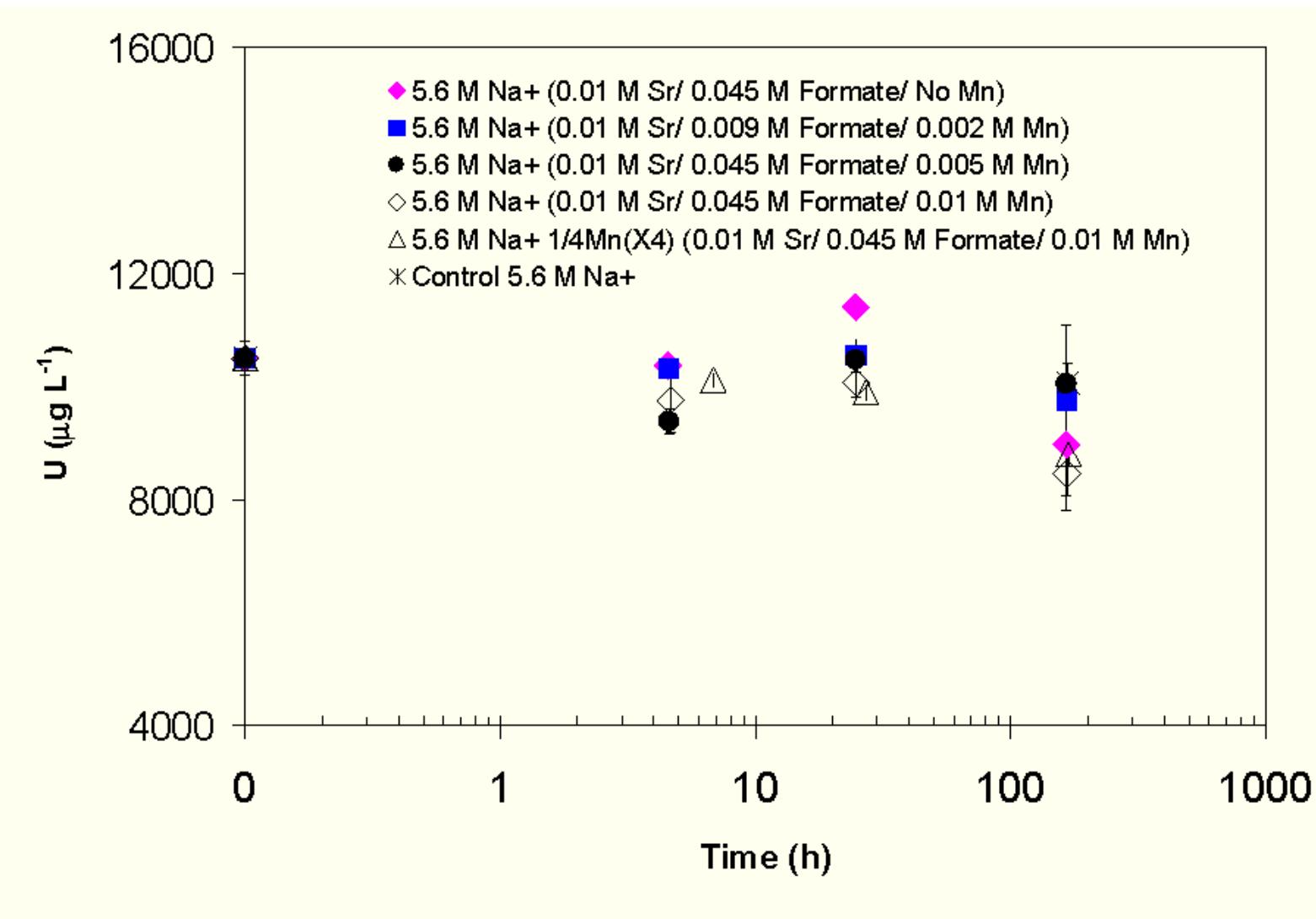
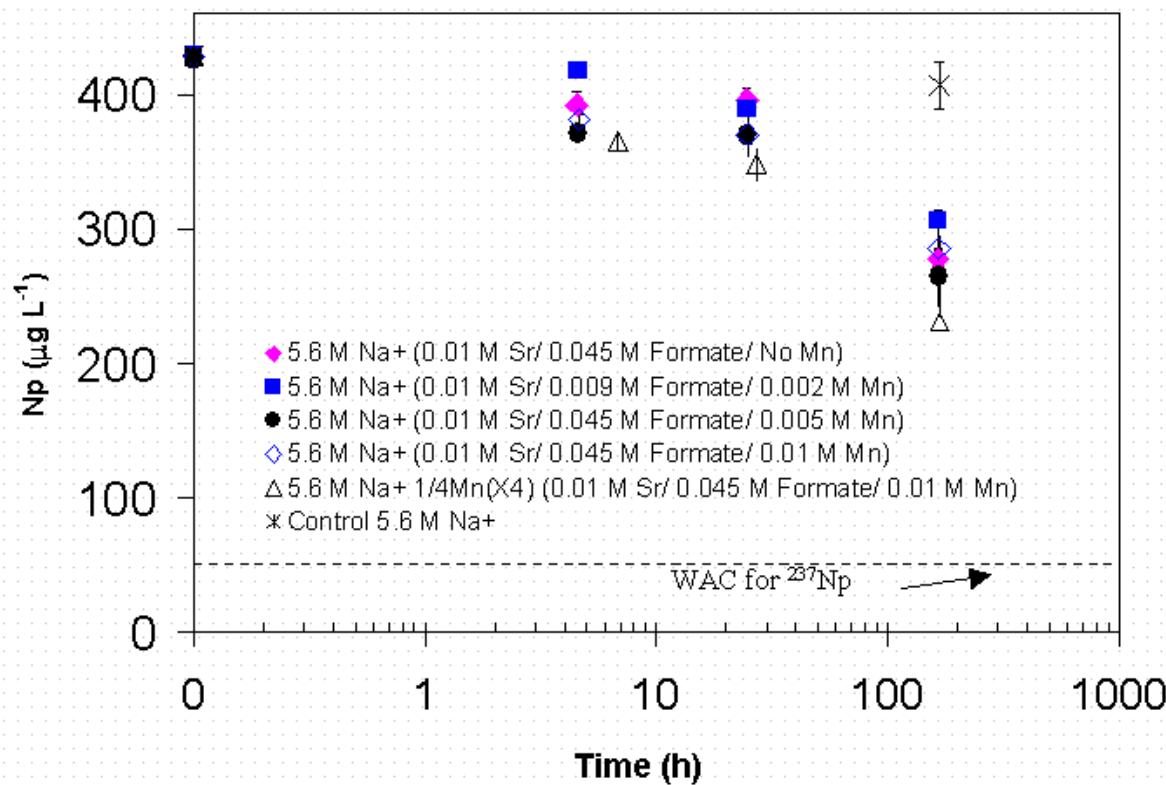


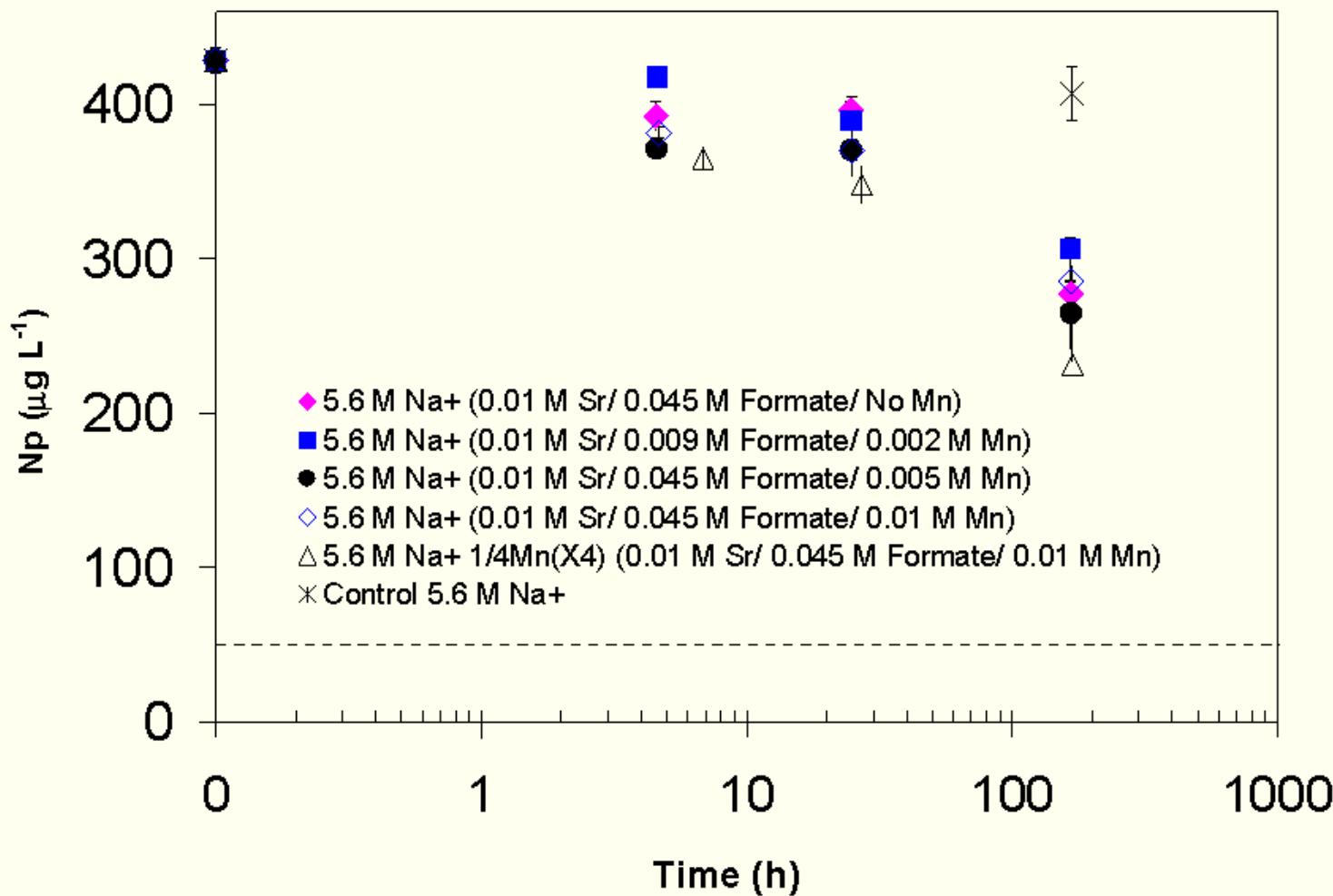
Figure 4-27 Effect of Permanganate Concentration and Permanganate Addition Method on Pu Removal in SRS HLW Simulant ICP-MS Data.



**Figure 4-28 Effect of Permanganate Concentration and Permanganate Addition Method on U Removal in SRS HLW Simulant.**



**Figure 4-29 Effect of Permanganate Concentration and Permanganate Addition Method on Np Removal in SRS HLW Simulant.**



**Figure 4-30 Effect of Permanganate Concentration and Permanganate Addition Method on Stable Sr Concentration in SRS HLW Simulant.**

#### 4.7 Effect of Strontium Seed Concentration at 0.013 M Permanganate and 0.015 M Formate Concentration

The data presented in this section are from our first set of tests as described in **Section 0**.

##### 4.7.1 Strontium

The results of our tests that examined the effect of seed Sr concentration at 0.013 M permanganate and 0.015 M formate on Sr removal are shown in **Figure 4-31**. Our tests show that Sr removal was generally greater at low seed Sr concentrations (**Figure 4-31**). Our results indicate that the Sr levels at all sampling times (except at 24 hours) for the 0.001 M seed Sr test met the Saltstone WAC for Sr at 5.2 atom % <sup>90</sup>Sr and Sr at 45 atom % <sup>90</sup>Sr (**Figure 4-31**). The Sr levels in our tests with higher seed Sr levels of

0.005 M Sr met Saltstone WAC for Sr at 5.2 atom %  $^{90}\text{Sr}$  at 30 minutes, 2, 24 and 168 hours. These data indicate that seed Sr may not be required for Sr removal in our SRS HLW salt simulants. If there was considerable formate decomposition (and production of  $\text{CO}_2$ ), it is likely that the Sr was removed via precipitation as  $\text{SrCO}_3(s)$ . It was also possible that the  $\text{Sr}^{2+}$  was removed from solution by the Mn-oxide phase that precipitated during the study.

#### 4.7.2 Plutonium

Our data for dissolved Pu in the tests that examined the effect of seed Sr concentration at 0.013 M permanganate and 0.015 M formate concentrations are shown in **Figure 4-32**. The data indicate that there was no influence of seed Sr concentration under these conditions (i.e., at low formate levels of 0.015 M). Additionally, the dissolved Pu concentrations in our treatments resembled that of the control throughout the study period (**Figure 4-32**). Decomposition of formate to yield  $\text{CO}_3$  ion and subsequent precipitation of  $\text{SrCO}_3$  may also involve the co-precipitation of Pu with  $\text{SrCO}_3$ .

#### 4.7.3 Uranium

In contrast to our Pu studies with variable seed Sr concentrations (in Section 0), U removal was greater in the presence of high seed Sr. Our observations suggest that during the precipitation of Sr, U may have been incorporated with the precipitating Sr phase and removed from solution (**Figure 4-33**).

#### 4.7.4 Neptunium

Our studies indicate that there was a slight effect of seed Sr concentration on Np removal at 0.013 M permanganate and 0.015 M formate levels (**Figure 4-34**). However, none of these treatments shown had significant Np removal and they did not meet the Saltstone limit for Np.

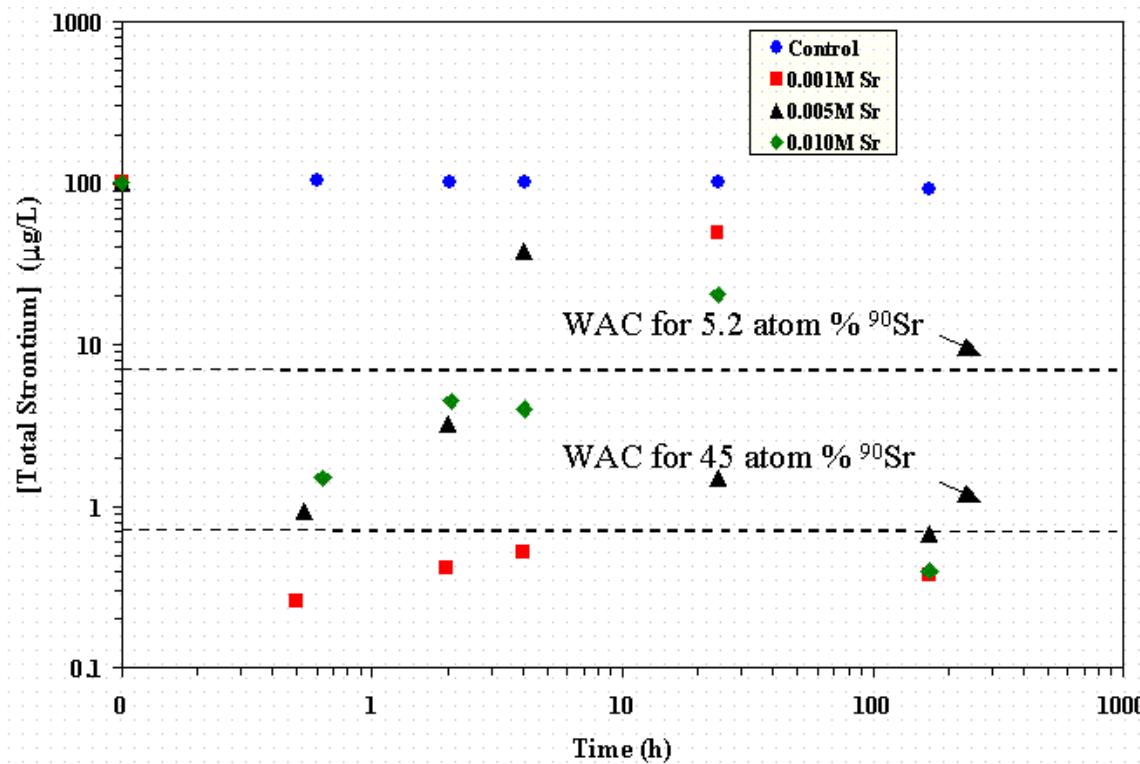
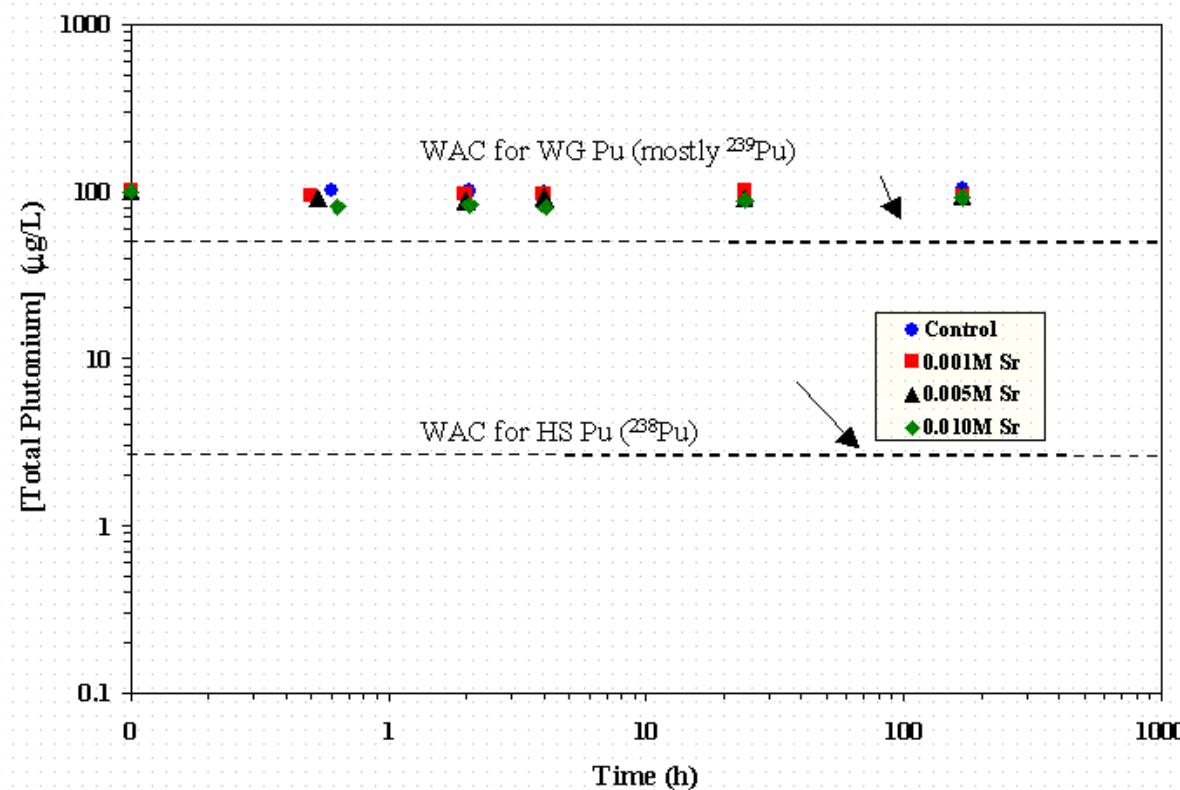


Figure 4-31 Effect of Seed Sr Concentration on Stable Sr Removal at

**0.013 M Permanganate and 0.015 M Formate in SRS HLW Simulant.**

**Figure 4-32 Effect of Seed Sr Concentration on Pu Removal at 0.013 M Permanganate and 0.015 M Formate in SRS HLW Simulant** ◆PuTTA Data.

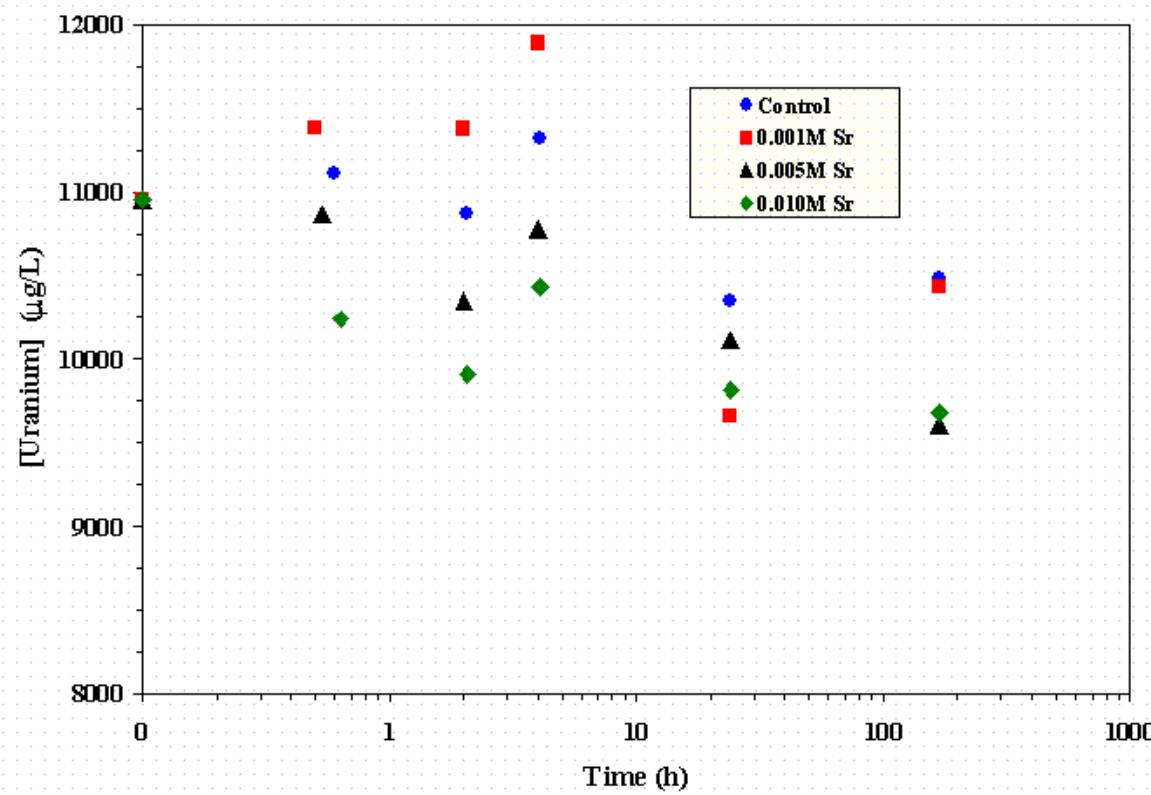
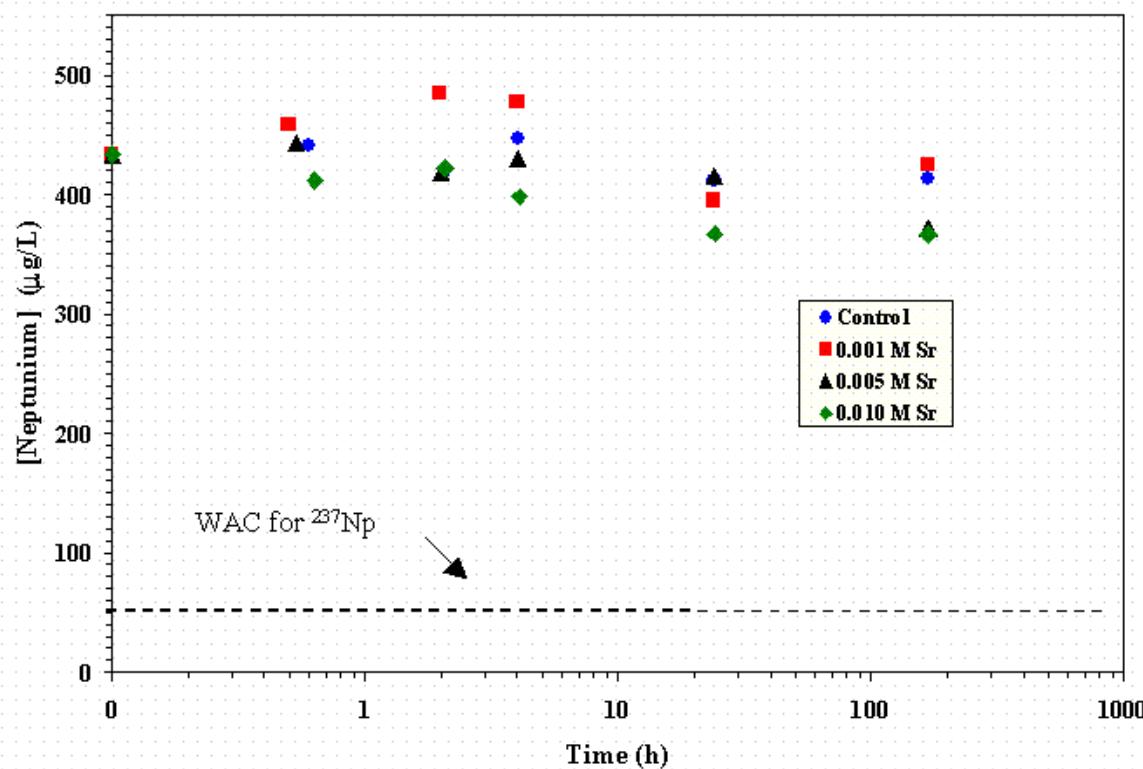


Figure 4-33 Effect of Seed Sr Concentration on U Removal at 0.013 M Permanganate and 0.015 M Formate in SRS HLW Simulant.



**Figure 4-34 Effect of Seed Sr Concentration on Np Removal at 0.013 M Permanganate and 0.015 M formate in SRS HLW Simulant.**

#### 4.8 Effect of Strontium Seed Concentration at 0.01 M Permanganate and 0.045 M Formate Concentrations

The data presented in this section are from our second set of tests as described in Section 3.2.

##### 4.8.1 Strontium

The results of our tests that examined the effect of seed Sr concentration at 0.01 M permanganate treatment and 0.045 M formate are shown in **Figure 4-35**. Our tests show that Sr removal was generally greater at low seed Sr concentration (**Figure 4-35**). Our results indicate that the Sr levels at all sampling times met the Saltstone WAC for Sr at 5.2 atom. %  $^{90}\text{Sr}$  (**Figure 4-35**). The Sr levels in tests with little (0.001 M Sr) or no seed Sr met the Saltstone limits for Sr at 45 atom %  $^{90}\text{Sr}$  at all sampling times. These data indicate that seed Sr was not required for the removal of Sr in our SRS HLW salt simulants.

##### 4.8.2 Plutonium

**Figures 4-36 and 4-37** show the results of the Pu-TTA and Pu ICP-MS analyses for the permanganate tests that examined the influence of seed Sr on Pu removal. In general, the two Pu analyses yielded similar data. However, our ICP-MS data for the 24-hour samplings were typically a few  $\mu\text{g L}^{-1}$  lower than that of our Pu-TTA data.

Our data on Pu for the tests that examined the effect of seed Sr concentration at 0.01 M permanganate and 0.045 M formate are shown in **Figures 4-36 and 4-37**. The data indicate that there was a positive influence of seed Sr concentration on Pu removal at high seed Sr levels. Our treatment with 0.01 M seed Sr was the only one in this set of tests that met the Saltstone limit for WG Pu isotopes and this occurred after one week (based on the Pu-TTA data) or 24 hours (based on the ICP-MS data) of equilibration

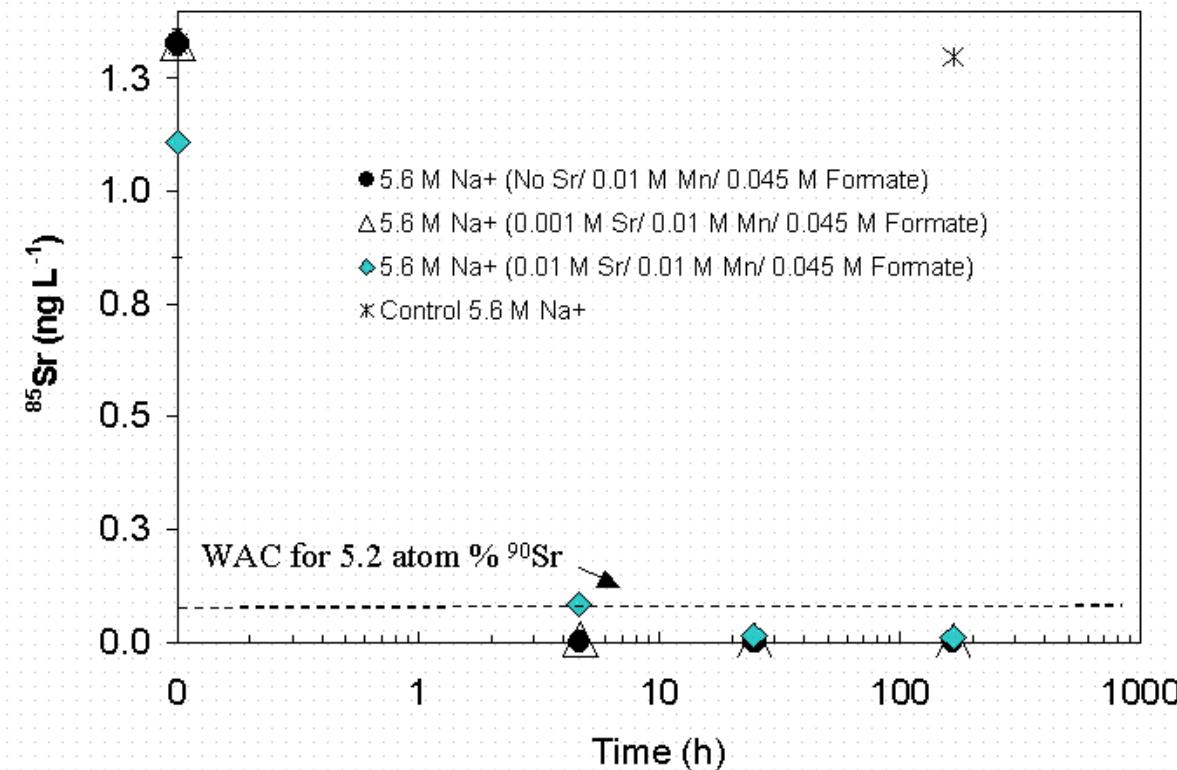
(**Figures 4-36 and 4-37**). Removal of Pu by  $\text{CaCO}_3$  has been observed under near neutral pH conditions. By analogy, Pu may also become structurally incorporated (co-precipitated) in the  $\text{SrCO}_3$  because similarities exist between the ionic radii of the cations occupy in the structures of  $\text{SrCO}_3$  and  $\text{CaCO}_3$ .

#### 4.8.3 Uranium

Our studies show that U removal from solution was greater in the presence of high Sr (0.01 M) and 0.45 M formate suggesting that the precipitating Sr may be incorporating (i.e., co-precipitating with) U and removing it from solution **Figure 4-38**. Removal of U was reversible in this high Sr treatment.

#### 4.8.4 Neptunium

Our studies indicate that there was a slight effect of seed Sr concentration on Np removal (**Figure 4-39**). However, none of these treatments shown had significant Np removal or met the Saltstone limit for Np.



**Figure 4-35 Effect of Seed Sr Concentration at 0.01 M Permanganate and 0.045 M Formate on  $^{85}\text{Sr}$  Removal in SRS HLW Simulant.**

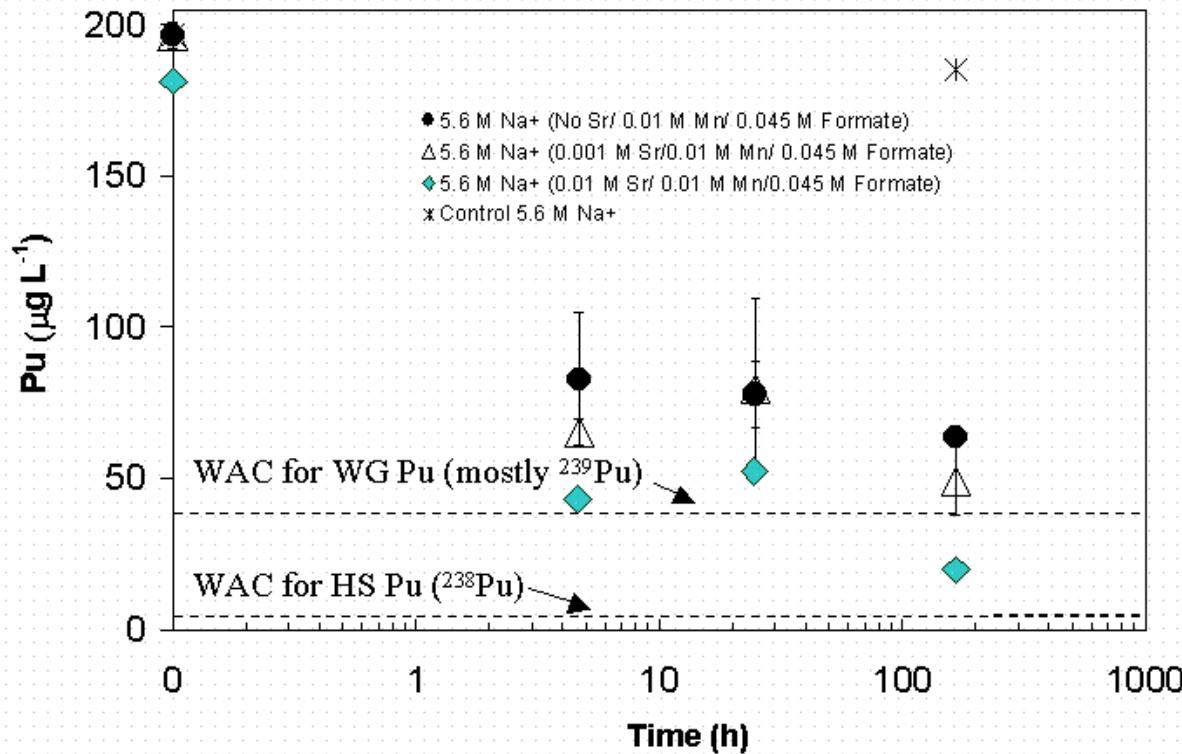


Figure 4-36 Effect of Seed Sr Concentration at 0.01 M Permanganate and 0.045 M Formate on Pu Removal in SRS HLW Simulant◆PuTTA Data.

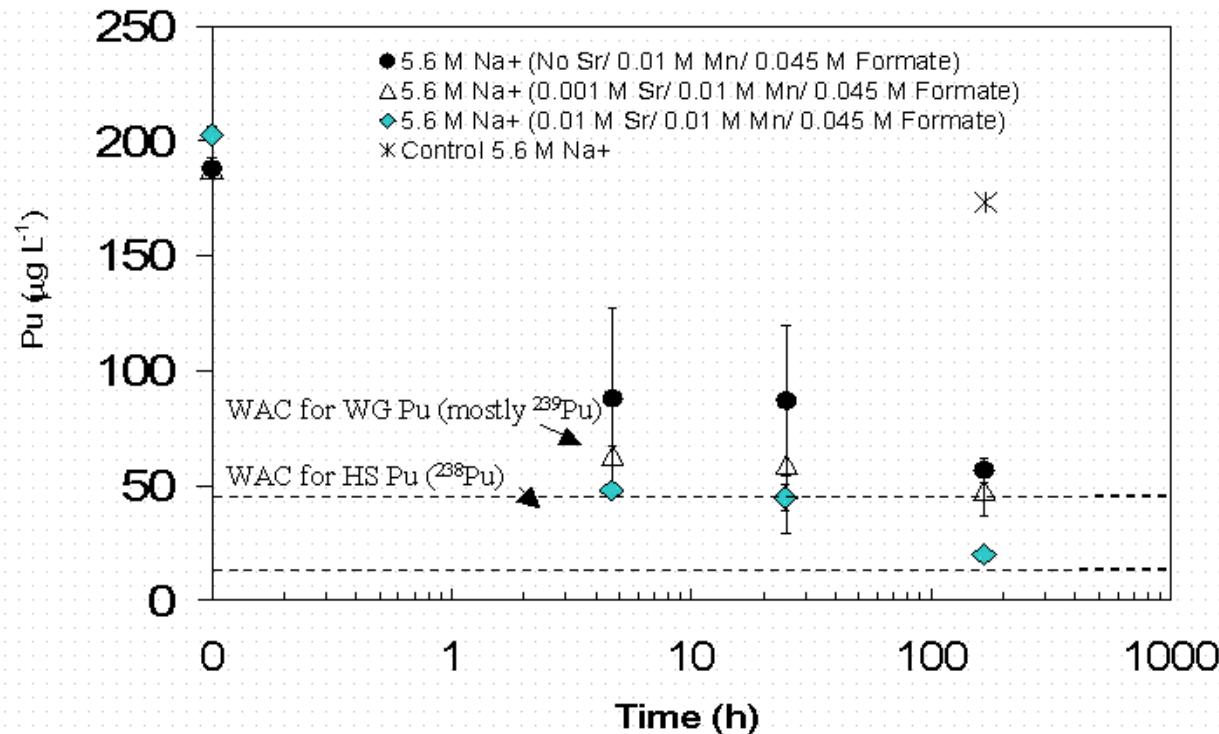


Figure 4-37 Effect of Seed Sr Concentration at 0.01 M Permanganate and 0.045 M Formate on Pu Removal in SRS HLW Simulant ICP-MS Data.

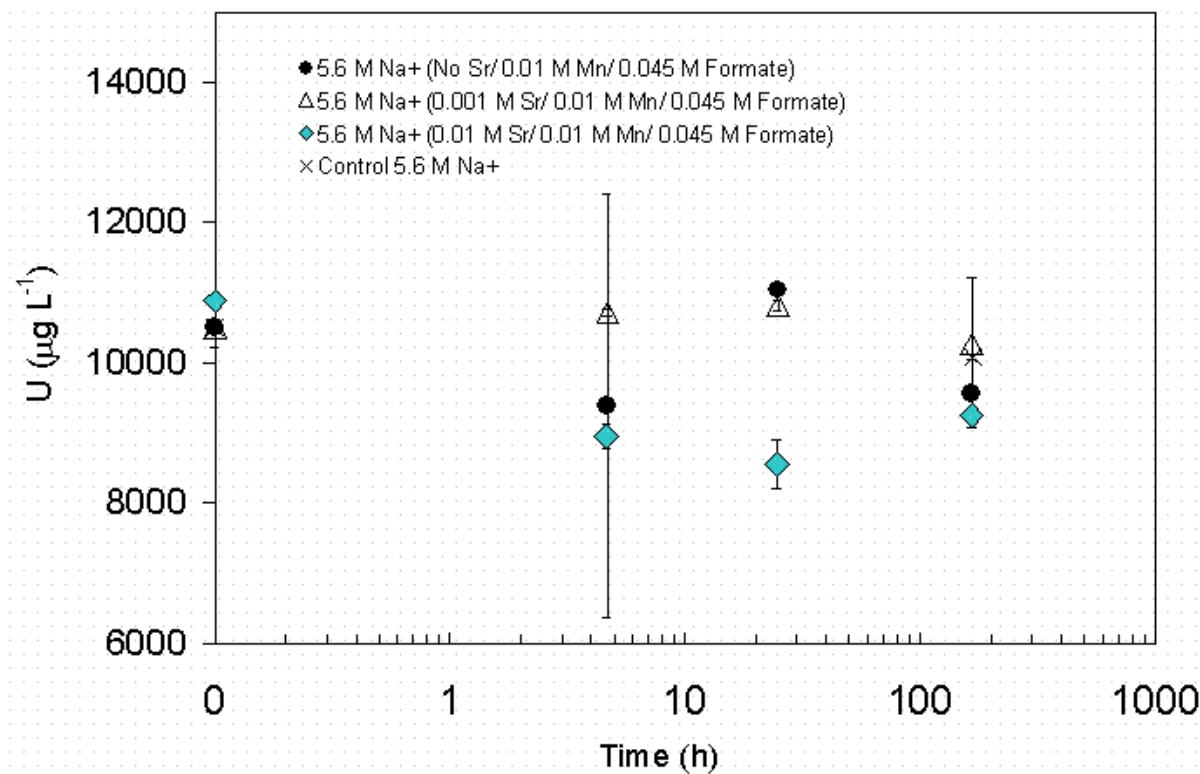


Figure 4-38 Effect of Seed Sr Concentration at 0.01 M Permanganate and 0.045 M Formate on U Removal in SRS HLW Simulant.

#### 4.8.5 Stable Strontium

Our studies show that precipitation of seed Sr was slower for the tests with high seed Sr levels, which was what we had expected (Figure 4-40).

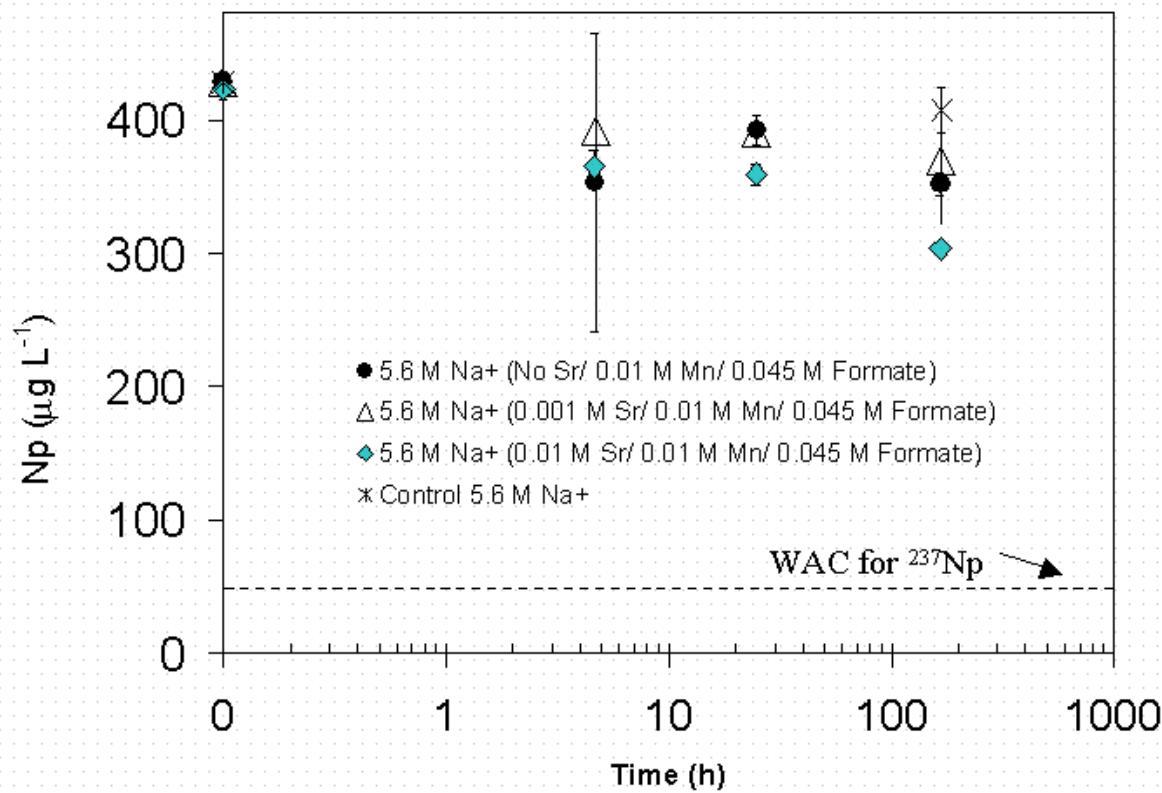
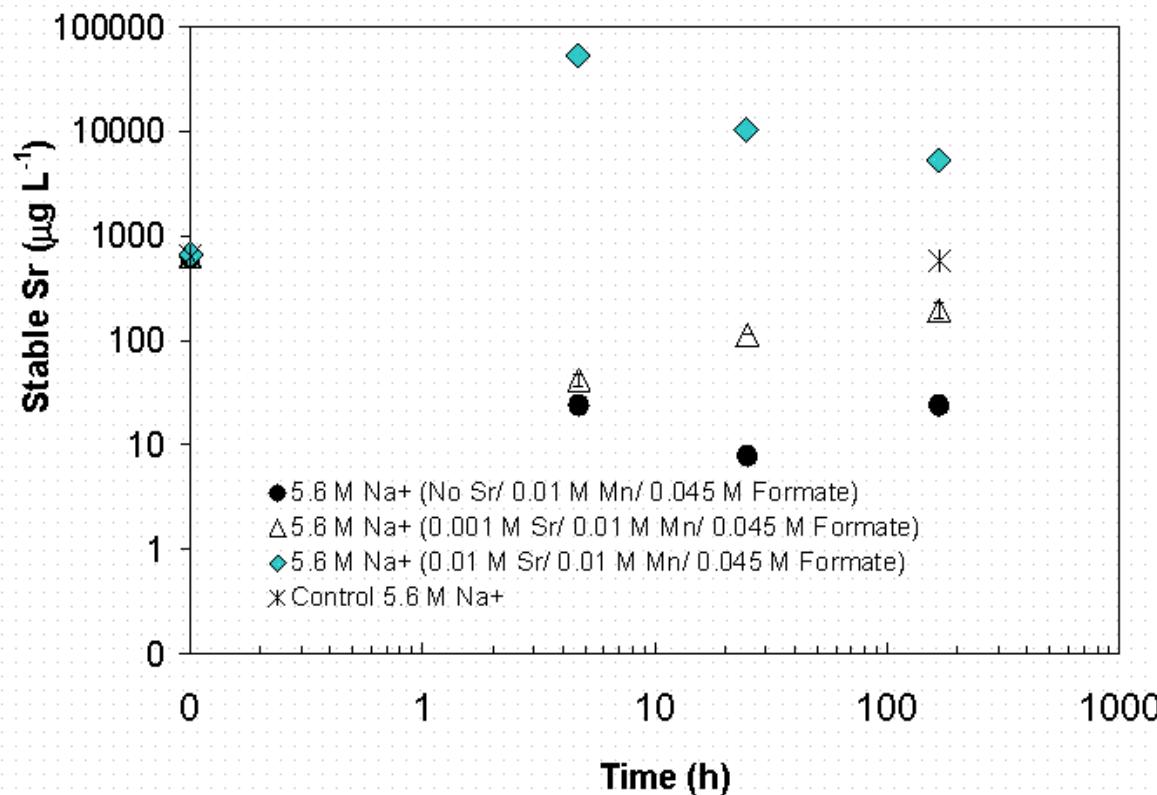


Figure 4-39 Effect of Seed Sr Concentration at 0.01 M Permanganate and 0.045 M Formate on Np Removal in SRS HLW Simulant.



**Figure 4-40 Effect of Seed Sr Concentration at 0.01 M Permanganate and 0.045 M Formate on Stable Sr Concentration in SRS HLW Simulant.**

#### 4.9 Effect of Ionic Strength (i.e., $\text{Na}^+$ Concentration) at 0.01 M Permanganate Concentration and 0.045 M Formate Concentration

The data presented in this section are from the first set of tests as described in Section 2.2.

##### 4.9.1 Strontium

The results of our tests that examined the effect of ionic strength at 0.01 M permanganate treatment and 0.045 M formate are shown in **Figure 4-41**. Our tests show that Sr removal was generally unaffected by ionic strength (**Figure 4-41**). These results are somewhat consistent with our studies at 0.013 M permanganate and 0.015 M formate as previously discussed. Our results indicate that the Sr levels at all sampling times and ionic strengths met the Saltstone WACs for Sr at 5.2 atom %  $^{90}\text{Sr}$  (**Figure 4-41**). Our data indicate that Sr removal was not greatly subject to changes in ionic strength within the study range of 4.0 to 5.6 M  $\text{Na}^+$  and that Sr removal was not reversible during the 168 h period of our study.

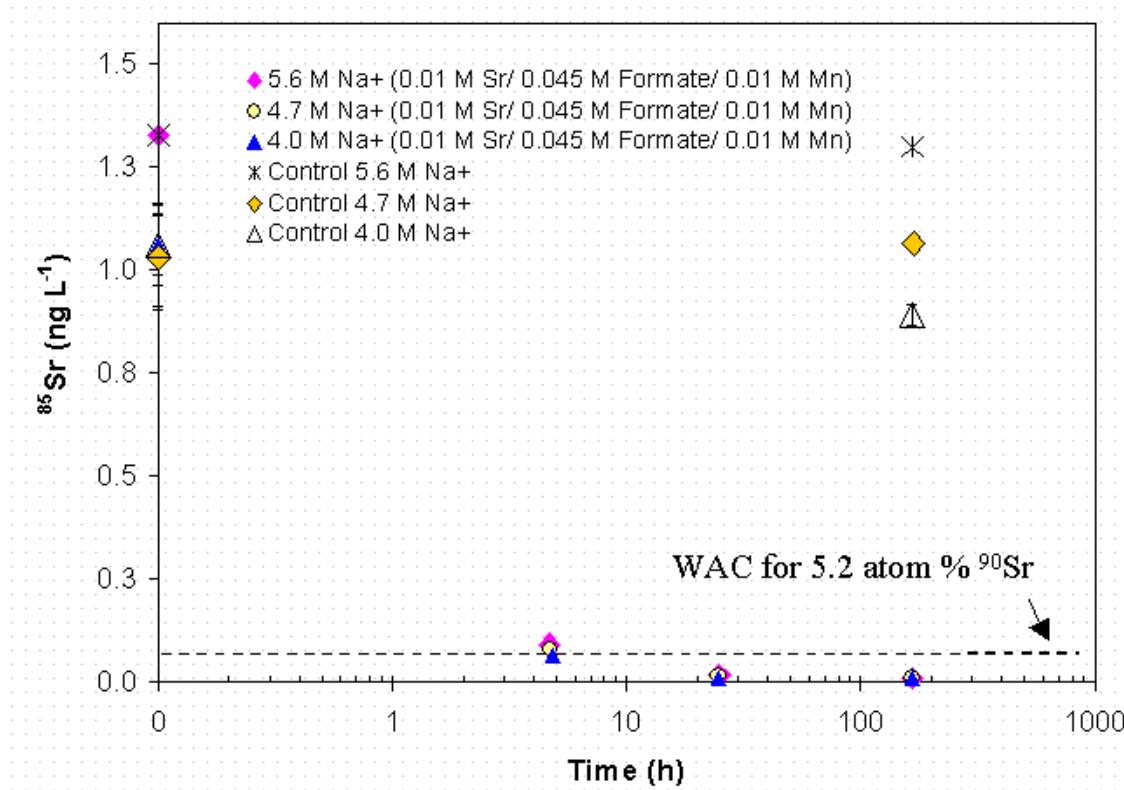
##### 4.9.2 Plutonium

**Figures 4-42 and 4-43** show the results of the Pu-TTA and Pu ICP-MS analyses for the permanganate tests that examined reagent order and reductant choice. In general, the two Pu analyses yielded similar data.

Our data on Pu for the tests that examined the influence of ionic strength at 0.01 M permanganate and 0.045 M formate are shown in **Figures 4-42 and 4-43**. Our data indicate that there was a small influence of ionic strength on Pu removal in that there was greater Pu removal at lower ionic strength than at high ionic strength. Our treatments with 4.0 and 4.7 M  $\text{Na}^+$  met the Saltstone limit for Pu at WG isotopes and this occurred after 4 hours of equilibration (**Figures 4-42 and 4-43**).

#### 4.9.3 Uranium

Our studies show that U removal from solution was greater at lower ionic strength suggesting that there may be some influence of ionic strength on the precipitating Mn-oxide phases in these solutions (**Figure 4-44**).



**Figure 4-41 Effect of Ionic Strength at 0.01 M Permanganate and 0.045 M Formate on  $^{85}\text{Sr}$  Removal in SRS HLW Simulant.**

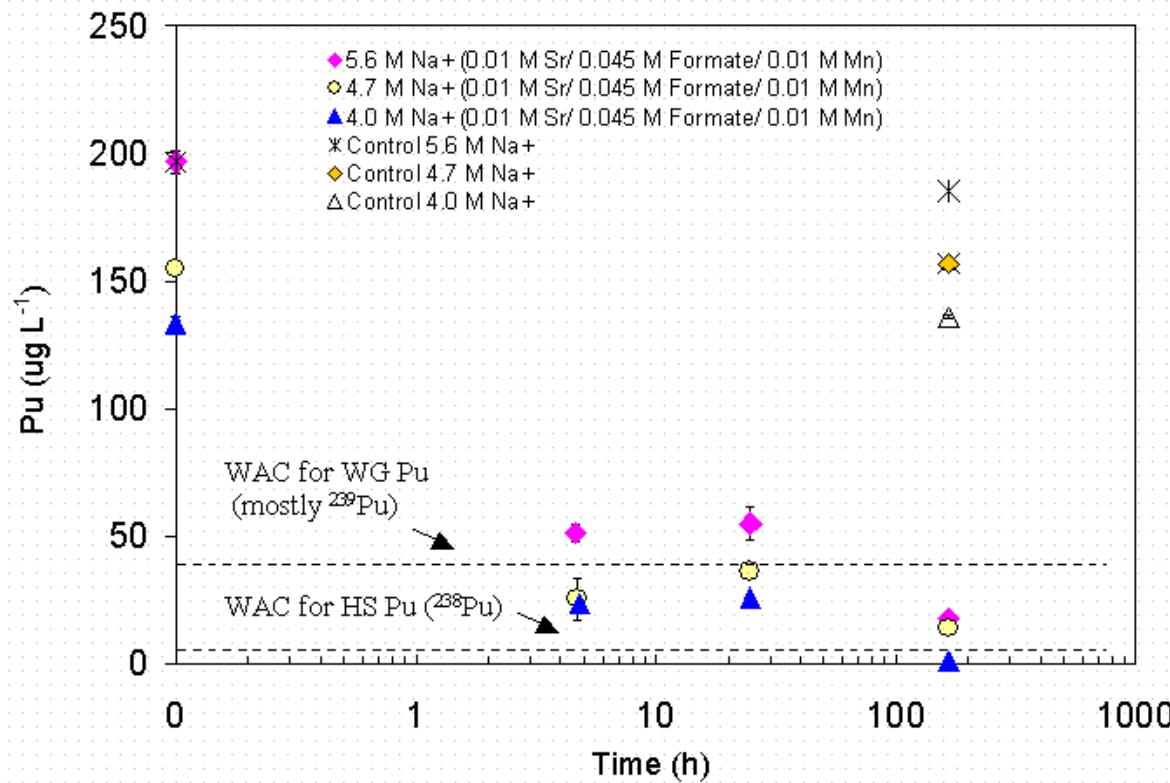


Figure 4-42 Effect of Ionic Strength at 0.01 M Permanganate and 0.045 M Formate on Pu Removal in SRS HLW Simulant. PuTTA Data.

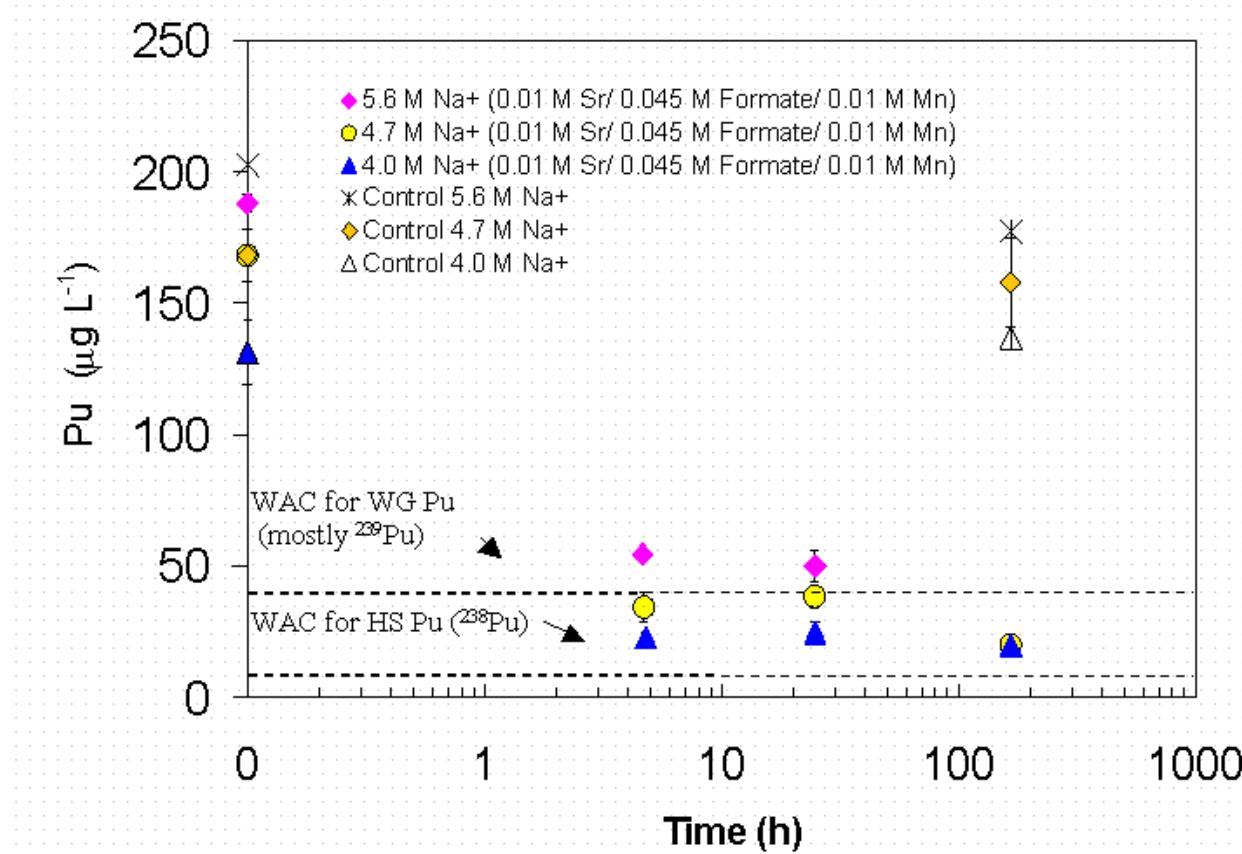
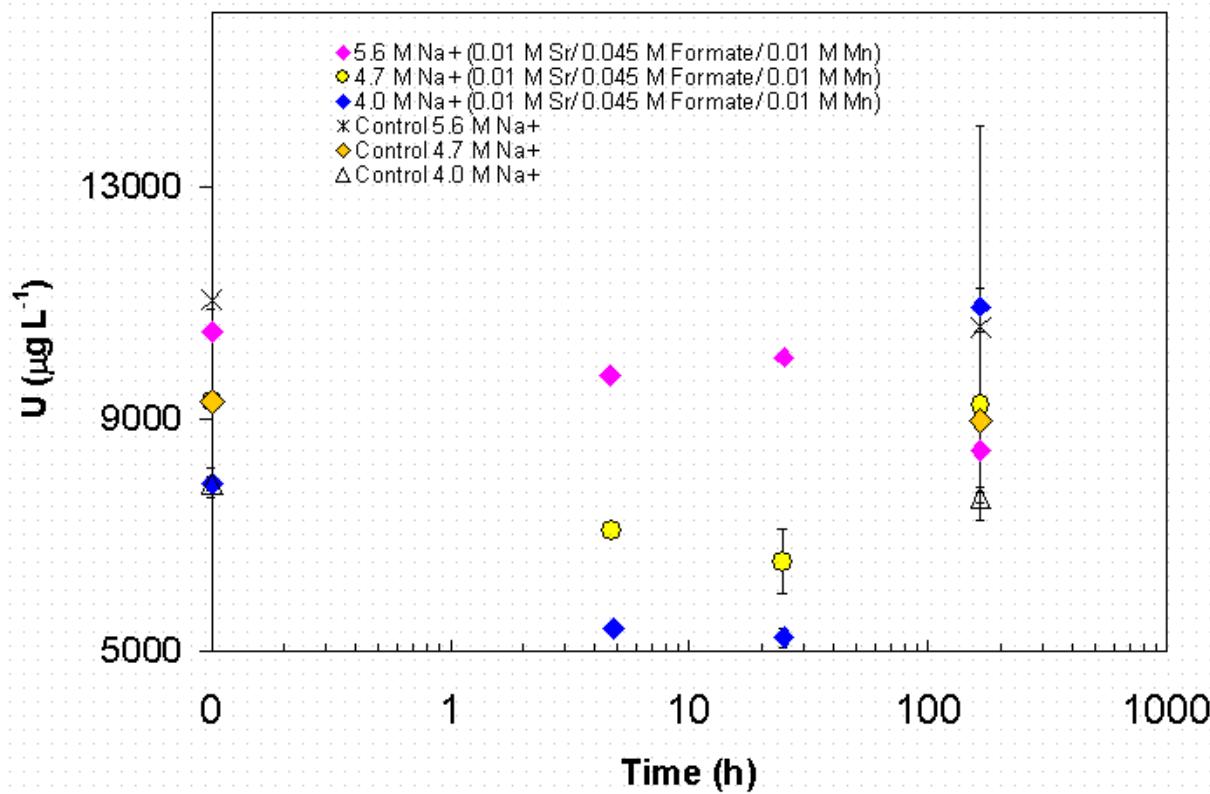


Figure 4-43 Effect of Ionic strength at 0.01 M Permanganate and 0.045 M Formate on Pu Removal in SRS HLW Simulant-ICP-MS Data.



**Figure 4-44 Effect of Ionic Strength at 0.01 M Permanganate and 0.045 M Formate on U Removal in SRS HLW Simulant.**

#### 4.9.4 Neptunium

As in our studies with Pu and U, Np removal from solution was somewhat greater at lower ionic strength (Figure 4-45) than at the higher ionic strength. However, none of these treatments shown had significant Np removal or met the Saltstone limit for Np.

#### 4.9.5 Stable Strontium

Our tests show that precipitation of the seed Sr from the salt solutions was not affected strongly by ionic strength (Figure 4-46).

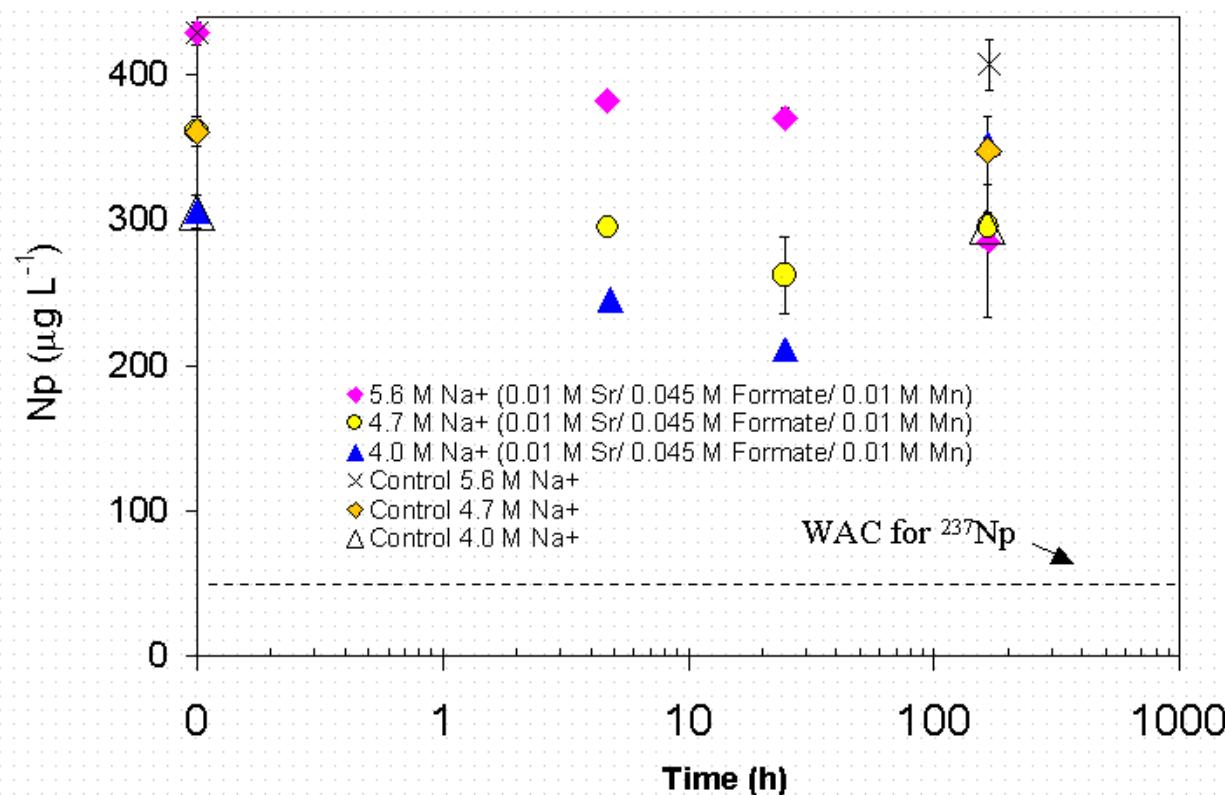
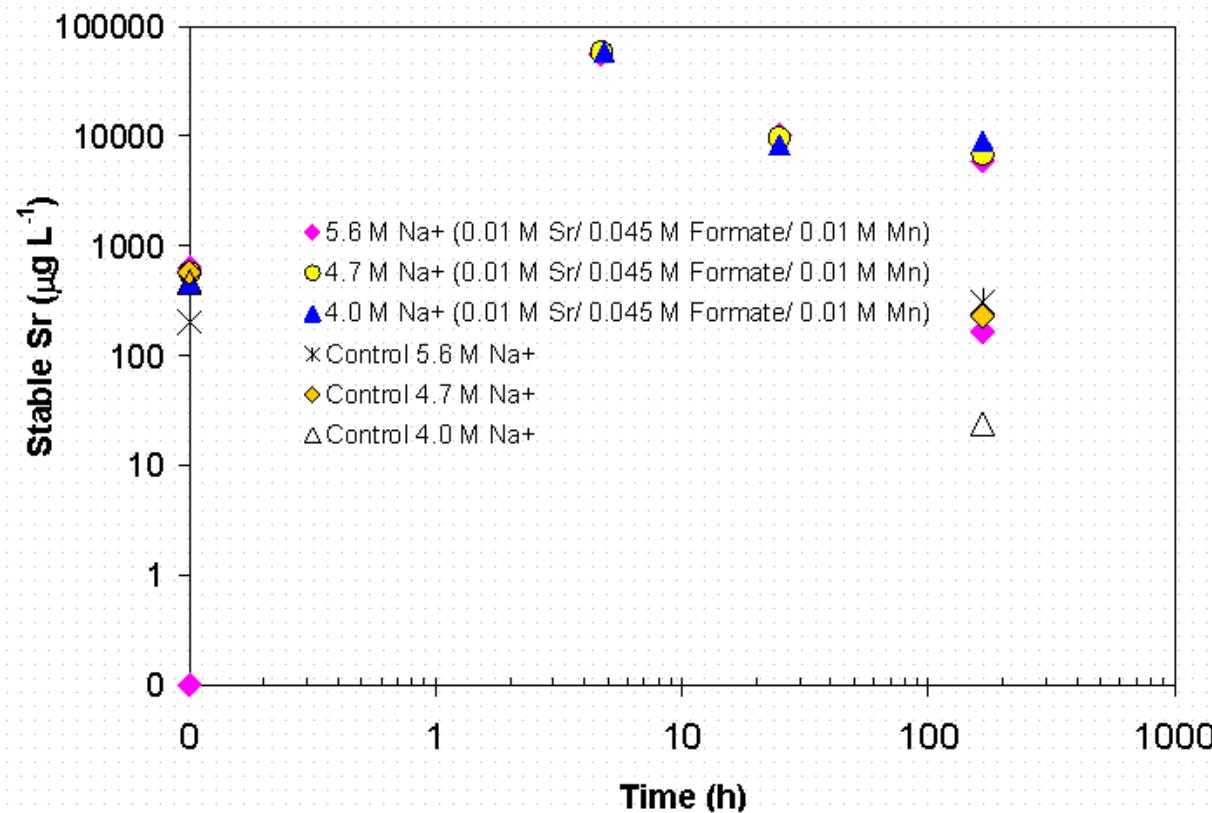


Figure 4-45 Effect of Ionic Strength at 0.01 M Permanganate and 0.045 M Formate on Np Removal in SRS HLW Simulant.



**Figure 4-46 Effect of Ionic Strength at 0.01 M Permanganate and 0.045 M Formate on Stable Sr Concentration in SRS HLW Simulant.**

#### 4.10 Decontamination Factors for All Tests

We tabulated the DF values for the tests as shown in **Table 4-1**. This table is for comparative purposes with MST and with data that has been represented in a similar manner in previous reports [Hobbs et al. (2000) and ref. therein].<sup>1</sup> One must use caution when comparing these data because the DF values are a function of the initial (spiked) Sr or actinide concentration in the salt solution. The starting Pu concentration in the experiments shown here were either 100 or 200 ppb Pu<sup>2</sup> as noted.

For Sr, tests that had DF values of greater than ~15 met the Saltstone WAC for 5.2 atom. <sup>90</sup>Sr whereas the DF values required for meeting the Saltstone WAC for 45 atom. <sup>90</sup>Sr needed to be greater than ~127. For Pu, tests having initial Pu levels of 200 ppb that had DF values of greater than ~5 met the Saltstone WAC for WG Pu whereas the DF values required for meeting the Saltstone WAC for HS Pu needed to be greater than ~120. For Pu, tests having initial Pu levels of 100 ppb that had DF values of greater than ~25 met the Saltstone WAC for WG Pu whereas the DF values required for meeting the Saltstone WAC for HS Pu needed to be greater than ~60. For Np, tests with DF values of ~9.5 or greater met the Saltstone WAC for <sup>237</sup>Np.

Strontium removal was greatest in the treatments without seed Sr addition and our DF values for these two tests far exceeded that for the MST addition test at 4 and 24 hours. For all of the actinides, treatments with 0.01 M sodium permanganate with 0.045 M peroxide as a reductant had the greatest amount of decontamination. The Sr DF values with seed Sr, 0.01 M permanganate and peroxide treatment were fairly good also. For the formate-containing tests, actinide removal was higher when permanganate was added sequentially. Treatments with Mn(II) in the absence of Ca<sup>2+</sup> proved comparable to that of 0.045 formate and 0.01 M permanganate addition.

**Table 4-1 The 4 and 24 hour DF values for Sr and the actinides from our tests with permanganate and MST.**  
**Values shown for Pu are based on Pu-TTA analyses. Average values are shown for tests done in duplicate.**

Test	Method	Decontamination Factor							
		Sr 4 h	Sr 24 h	Pu 4 h	Pu 24 h	U 4 h	U 24 h	Np 4 h	Np 24 h
1	5.6 M Na <sup>+</sup> (0.01 M Sr/ 0.01 M Mn(II))*	16.9	117.1	40.2	20.3	1.4	1.7	4.2	10.4
2	5.6 M Na <sup>+</sup> (0.01 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)*	26.2	3.5	1.1	1.2	1.0	1.1	1.1	1.2
3	5.6 M Na <sup>+</sup> (0.01 M Sr/ 0.0065 M MnO <sub>4</sub> / 0.015 M Formate)*	21.2	120.5	1.8	2.0	1.0	1.1	1.0	1.1
4	5.6 M Na <sup>+</sup> (0.01 M Sr/ 0.0013 M MnO <sub>4</sub> / 0.015 M Formate)*	8.7	94.4	1.5	1.5	1.0	1.0	1.0	0.9
5	5.6 M Na <sup>+</sup> (0.01 M Sr/ 0.013 M MnO <sub>4</sub> / 0.02 M Formate)*	25.8	32.1	1.2	1.3	1.1	1.1	1.1	1.1
6	5.6 M Na <sup>+</sup> (0.01 M Sr/ 0.013 M MnO <sub>4</sub> / 0.03 M Formate)*	27.3	156.8	1.8	2.6	1.1	1.1	1.1	1.1
7	5.6 M Na <sup>+</sup> (0.01 M Sr/ 0.013 M MnO <sub>4</sub> / 0.04 M Formate)*	27.0	156.8	2.2	2.6	1.1	1.1	1.1	1.1
8	5.6 M Na <sup>+</sup> (0.01 M Sr/ 0.013 M MnO <sub>4</sub> / >0.04 M Formate)*	19.1	117.7	4.4	4.3	1.1	1.1	1.1	1.1
9	5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)*	2.6	66.4	1.0	1.1	1.0	1.1	1.0	1.0
10	5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)*	194.2	2.0	0.8	1.0	0.9	1.1	0.9	1.1
11	5.6 M Na <sup>+</sup> (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu*	20.9	32.3	3.4	4.9	1.0	1.1	1.2	2.0
11	5.6 M Na <sup>+</sup> (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	94.6	111.2	4.5	5.0	1.1	1.1	1.5	1.8
12	5.6 M Na <sup>+</sup> (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO <sub>4</sub> / 0.04 M Formate)*	22.7	120.8	3.8	3.6	1.1	1.1	1.1	1.1
13	5.6 M Na <sup>+</sup> (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))*	12.7	95.7	10.2	10.4	1.2	1.6	2.8	3.8
14	5.6 M Na <sup>+</sup> (0.01 M Sr/ 0.01 M MnO <sub>4</sub> / 4 x 0.015 M H <sub>2</sub> O <sub>2</sub> ) with 200 ppb Pu	76.7	198.7	48.0	20.0	1.8	1.9	10.9	7.9
15	4.0 M Na <sup>+</sup> (0.01 M Sr/ 0.045 M Formate/ 0.01 M MnO <sub>4</sub> )	16.8	116.7	5.8	5.1	1.5	1.5	1.2	1.5
16	4.7 M Na <sup>+</sup> (0.01 M Sr/ 0.045 M Formate/ 0.01 M MnO <sub>4</sub> )	13.1	78.6	6.4	4.3	1.3	1.4	1.2	1.4
17	5.6 M Na <sup>+</sup> (0.01 M Sr/ 0.045 M Formate/ 0.01 M MnO <sub>4</sub> )	146.4	852.3	3.8	3.6	1.1	1.0	1.1	1.2
18	5.6 M Na <sup>+</sup> (0.01 M Sr/ 0.045 M Formate/ 0.005 M MnO <sub>4</sub> )	95.1	553.7	4.7	4.2	1.1	1.0	1.2	1.2
19	5.6 M Na <sup>+</sup> (0.01 M Sr/ 0.009 M Formate/ 0.002 M MnO <sub>4</sub> )	87.8	190.8	1.7	2.3	1.0	1.0	1.0	1.1
20	5.6 M Na <sup>+</sup> (0.01 M Sr/ 0.045 M Formate)	30.7	67.9	1.2	1.2	1.0	0.9	1.1	1.1
21	5.6 M Na <sup>+</sup> (0.01 M Sr/ 0.045 M Formate/ 4 x 0.0025 M MnO <sub>4</sub> )	150.3	958.0	19.1	11.4	1.0	1.1	1.2	1.2
22	5.6 M Na <sup>+</sup> (0.01 M MnO <sub>4</sub> / 0.045 M Formate)	4.12E+04	5.70E+04	2.5	2.6	1.2	1.0	1.3	1.1
23	5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.01 M MnO <sub>4</sub> / 0.045 M Formate)	2.23E+03	1.26E+04	3.0	2.6	1.0	1.0	1.1	1.1

24	5.6 M Na <sup>+</sup> (0.01 M Sr/ 0.01 M MnO <sub>4</sub> / 0.045 M Formate)		13.5	78.2	4.2	3.5	1.2	1.3	1.2	1.2
25	5.6 M Na <sup>+</sup> (0.01 M Sr/ 0.01 M MnO <sub>4</sub> / 0.045 M H <sub>2</sub> O <sub>2</sub> )		7.0	11.9	28.9	33.1	1.3	1.4	7.7	6.5
26	5.6 M Na <sup>+</sup> (0.045 M Formate/ 0.01 M Sr/ 0.01 M MnO <sub>4</sub> )		13.4	62.2	3.8	3.6	1.2	1.3	1.2	1.2
27	5.6 M Na <sup>+</sup> (0.045 M H <sub>2</sub> O <sub>2</sub> / 0.01 M Sr/ 0.01 M MnO <sub>4</sub> )		5.5	35.9	94.1	31.4	1.7	1.3	25.4	7.1

\* Applies to salt solutions with a target (spiked) Pu concentration of approximately 100 ppb.

#### 4.11 Color and Foaming Observations with Salt Simulants

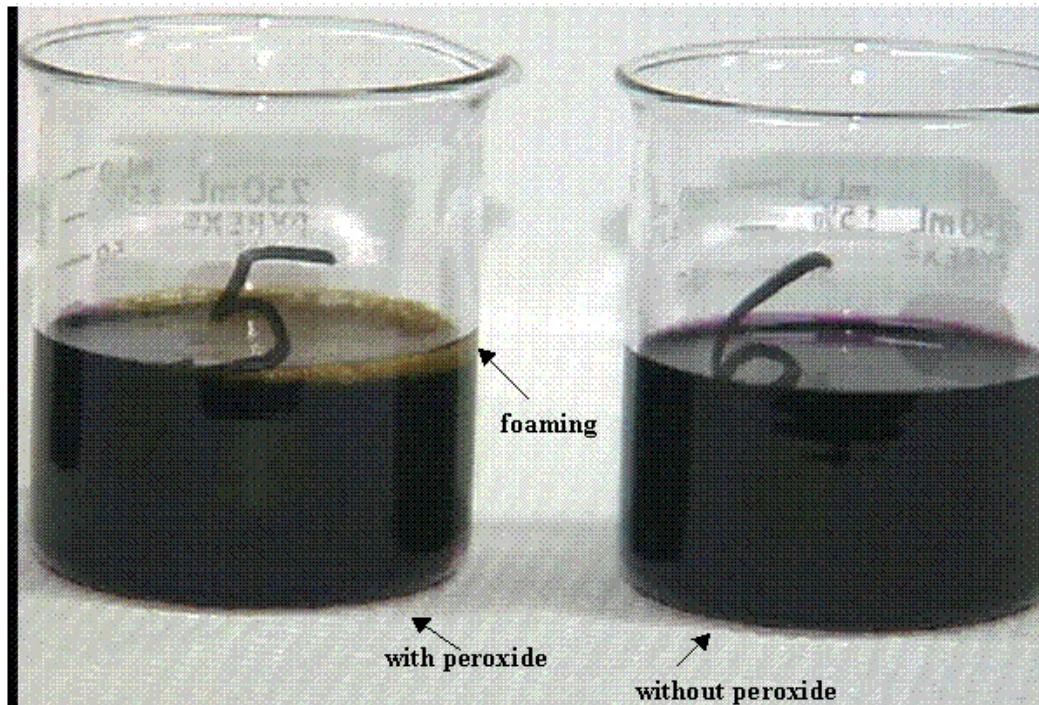
The permanganate treatments underwent two color changes during the first hour of equilibration with the test reagents. Upon permanganate addition in the absence of any reductant, the salt solutions were purple in color. After we added formate as a reductant to the permanganate solutions or permanganate to formate-containing solutions, the solutions changed color from purple to greenish-blue as shown in **Figure 4-47**. [The unfiltered solutions shown in **Figure 4-47** were made with non-radioactive salt solutions in glass beakers so that we could better examine the color changes that we observed (i.e., the samples in the beakers are not from our actual tests with spiked simulant)]. This greenish blue color was accompanied by the generation of brown solids on the bottom of the containers (if not shaken as shown in **Figure 4-47**). These colors persisted for about the first hour of our tests. After 1 hour, the unfiltered suspensions were light or dark brown in color. The filtered solutions from the 30-minute samplings of these tests were a light turquoise in color (prior to acidification). After 4 hours, all of our filtered sample solutions were colorless and clear upon filtration.

After addition of peroxide to salt solutions with permanganate or after the addition of permanganate to salt solutions with peroxide, the samples began to foam (as shown in **Figure 4-48**). The foaming was concurrent with the solutions turning brown. The reaction was nearly instantaneous and it was evident that brown Mn oxide solids formed rapidly in the treatments. No foaming was observed with tests that had formate as a reductant. The foaming is likely from the rapid liberation of oxygen gas from the peroxide during reaction with permanganate ion.

The color changes that we observed during our testing are representative of a change in the Mn oxidation state in our treatments. Manganese(VII) is typically purple in aqueous solutions and dissolved Mn(VI) and Mn(V) are typically blue-green in color. In solution, Mn(IV) and Mn(III) are not highly soluble and they form brownish-black solids.



**Figure 4-47 Photograph of a Salt Solution in a Glass Beaker  
Containing Permanganate after Addition of 0.01 M Seed Sr, 0.01 M  
Permanganate and 0.045 M Formate. This Photo was taken Prior to Mixing.**



**Figure 4-48 Photograph of Salt Solutions in Glass Beakers Containing Recently Added Permanganate with Peroxide as a Reductant (Note Foaming, at Left) and Without Added Peroxide (at Right).**

## 5.0 Conclusions

The testing indicates that permanganate treatment offers a rapid and high degree of Sr and actinide decontamination under certain conditions in our salt simulants. Our findings indicate that peroxide (more so than formate ion) greatly facilitated actinide removal in the permanganate studies at permanganate concentrations of 0.1 M. The use of peroxide addition with permanganate should be explored for waste treatment because it offers faster Sr and actinide removal than MST addition (at 0.4 g L<sup>-1</sup>) or formate with permanganate. Peroxide in the hydrogen form would not contribute to the Na<sup>+</sup> content of the waste. Our studies indicate that the permanganate concentrations may need to be as high as 0.01 M and reductant concentrations should be in three-fold stoichiometric excess.

However, the effect of temperature on permanganate treatment in the presence of peroxide has yet to be evaluated. Higher treatment temperatures may pose a complication to waste treatment in that it may increase process control difficulty, which would lower actinide removal. An increase in temperature may promote peroxide decomposition and result in greater foaming. Studies should be conducted to examine whether there is a temperature effect on Sr<sup>2+</sup> and actinide removal.

Suggested tests consist of sequential addition of peroxide with and without seed Sr (Sr may not be required when peroxide is the reductant but this has not been evaluated in our current study). Other tests that determine ways to minimize foaming during mixing of the permanganate and peroxide should also be explored. Our study indicates that 0.005 M Ca<sup>2+</sup> addition is not required for Sr and actinide removal using formate (0.015 M) as the reductant.

## 6.0 Quality Assurance

The following documents govern the work reported in this document.

- D. T. Hobbs, T. B. Peters, M. J. Barnes, M. C. Duff and K. M. Marshall, "Task Technical and Quality Assurance Plan for FY01 Strontium and Actinide Removal Testing," WSRC-RP-2001-00188, Rev. 1, July 31, 2001.
- Savannah River Site Salt Processing Project: FY 2002 Research and Development Program Plan, PNNL-13707, Rev. 1, December 2001.
- Notebooks WSRC-NB-2001-00161 (M. C. Duff), WSRC-NB-2000-00120 (D. T. Hobbs) and WSRC-NB-2001-00011 (D. T. Hobbs) contain the experimental data obtained from this work.

## 7.0 Acknowledgements

The authors appreciate H. D. Harmon (PNNL), P. C. Suggs (DOE-SR), T. B. Peters (SRTC), W. L. Tamosaitis (SRTC), and D. D. Walker (SRTC) for their assistance, support and insight. B. H. Croy, N. E. Gregory, M. S. Bloom, and H. L. Thacker (all of SRTC) are acknowledged for their assistance in the preparation of the permanganate samples. D. P. Diprete, C. C. Diprete, M. A. Malek, P. R. Woller and W. T. Boyce provided excellent analytical support and C. Pierce assisted with the health physics aspects of this work.

## 8.0 Experimental Data

**All data in this section have been corrected for dilution. Data from the first set of experiments:**

<u>Description</u>	<u>Average Time (h)</u>	<u>Sr (ug/L)</u>	<u>Stable</u>	
			<u>Sr std dev</u>	<u>Sr DF</u>
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	0.0	64.5	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	0.5	32.8	NA	1.96
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	2.0	0.3	NA	243.52
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	4.0	3.8	NA	16.87
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	24.0	0.6	NA	117.07
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	168.0	0.4	NA	152.91
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.0	100.2	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.6	0.3	NA	389.81
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	2.0	4.5	NA	22.04
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	4.0	3.8	NA	26.25
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	24.0	28.5	NA	3.51
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	168.2	0.4	NA	240.12
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	0.0	100.2	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	0.6	4.1	NA	24.40
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	2.1	5.0	NA	19.89
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	4.1	4.7	NA	21.22
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	24.1	0.8	NA	120.54
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	168.3	8.7	NA	11.58
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	0.0	100.2	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	0.6	28.3	NA	3.54
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	2.1	20.5	NA	4.88
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	4.1	11.5	NA	8.68
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	24.1	1.1	NA	94.37
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	168.3	0.8	NA	118.43
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	0.0	100.2	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	0.7	0.9	NA	109.69
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	2.1	2.8	NA	35.69
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	4.1	3.9	NA	25.83
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	24.2	3.1	NA	32.05
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	168.4	0.4	NA	264.15
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	0.0	100.2	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	0.7	1.4	NA	73.22
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	2.2	4.3	NA	23.23
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	4.2	3.7	NA	27.29
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	24.3	0.6	NA	156.79
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	168.4	0.5	NA	215.66
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	0.0	100.2	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	0.8	1.4	NA	72.03
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	2.2	4.0	NA	24.92
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	4.2	3.7	NA	26.95
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	24.3	0.6	NA	156.79
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	168.5	0.5	NA	218.71
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	0.0	64.5	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	0.5	0.4	NA	176.20
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	2.0	1.9	NA	33.79
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	4.0	3.4	NA	19.09
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	24.0	0.5	NA	117.70
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	168.0	0.4	NA	177.97
5.6 M Na+ (0.005 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.0	100.2	NA	1.00
5.6 M Na+ (0.005 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.5	0.9	NA	107.62
5.6 M Na+ (0.005 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	2.0	3.2	NA	30.90

5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	4.0	38.1	NA	2.63
5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	24.0	1.5	NA	66.36
5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	168.0	0.7	NA	148.19
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	0.0	100.2	NA	1.00
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	0.5	0.3	NA	389.64
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	2.0	0.4	NA	242.17
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	4.0	0.5	NA	194.22
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	24.0	49.3	NA	2.03
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	168.0	0.4	NA	266.78

**Data from the first set of experiments:**

<u>Description</u>	<u>Average Time (h)</u>	<u>Sr (ug/L)</u>	<u>Stable</u>	
			<u>Sr std dev</u>	<u>Sr DF</u>
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	0.0	64.5	NA	1.00
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	0.5	6.6	NA	9.80
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	2.0	3.6	NA	17.95
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	4.0	3.1	NA	20.88
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	24.0	2.0	NA	32.27
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	168.0	1.5	NA	43.09
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	0.0	93.3	NA	1.00
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	0.6	1.7	NA	55.86
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	2.0	1.0	NA	94.55
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	4.0	0.8	NA	111.22
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	24.0	0.6	NA	150.53
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	168.0	0.3	NA	304.85
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	0.0	64.5	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	0.5	0.5	NA	124.23
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	2.0	2.7	NA	23.90
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	4.0	2.8	NA	22.73
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	24.0	0.5	NA	120.79
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	168.0	0.4	NA	181.59
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	0.0	64.5	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	0.5	33.7	NA	1.91
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	2.0	26.4	NA	2.44
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	4.0	5.1	NA	12.66
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	24.0	0.7	NA	95.65
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	168.0	0.6	NA	106.72
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	0.0	92.4	0.0	1.00
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	1.1	3.5	1.8	26.56
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	2.7	2.2	1.2	41.10
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	4.7	1.2	0.7	76.72
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	25.1	0.5	0.2	198.66
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	169.5	0.1	0.1	783.31
5.6 M Na+ Control with 200 ppb Pu	0.0	92.6	NA	1.00
5.6 M Na+ Control with 200 ppb Pu	0.5	92.1	NA	1.01
5.6 M Na+ Control with 200 ppb Pu	2.0	93.7	NA	0.99
5.6 M Na+ Control with 200 ppb Pu	4.0	94.4	NA	0.98
5.6 M Na+ Control with 200 ppb Pu	24.0	91.7	NA	1.01
5.6 M Na+ Control with 200 ppb Pu	168.0	90.2	NA	1.03
5.6 M Na+ Control with 100 ppb Pu	0.0	64.5	NA	1.00
5.6 M Na+ Control with 100 ppb Pu	0.6	64.6	NA	1.00
5.6 M Na+ Control with 100 ppb Pu	2.1	65.5	NA	0.98
5.6 M Na+ Control with 100 ppb Pu	4.1	68.7	NA	0.94
5.6 M Na+ Control with 100 ppb Pu	24.2	63.8	NA	1.01
5.6 M Na+ Control with 100 ppb Pu	168.0	60.3	NA	1.07

**Data from the first set of experiments:**

**PuTTA Data**

Description	Average Time (h)	Pu (ug/L)	Pu (ug/L) std dev	Pu DE
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	0.0	97.5	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	0.5	21.4	NA	4.55
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	2.0	5.4	NA	18.02
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	4.0	2.4	NA	40.22
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	24.0	4.8	NA	20.27
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	168.0	14.8	NA	6.59
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.0	100.0	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.6	78.2	NA	1.28
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	2.0	80.8	NA	1.24
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	4.0	77.7	NA	1.29
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	24.0	85.6	NA	1.17
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	168.2	86.3	NA	1.16
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	0.0	100.0	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	0.6	63.0	NA	1.59
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	2.1	44.7	NA	2.24
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	4.1	45.7	NA	2.19
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	24.1	45.3	NA	2.21
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	168.3	22.1	NA	4.53
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	0.0	100.0	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	0.6	65.5	NA	1.53
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	2.1	58.3	NA	1.72
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	4.1	49.8	NA	2.01
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	24.1	44.8	NA	2.23
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	168.3	42.2	NA	2.37
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	0.0	100.0	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	0.7	85.8	NA	1.17
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	2.1	78.6	NA	1.27
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	4.1	74.4	NA	1.34
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	24.2	82.4	NA	1.21
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	168.4	85.5	NA	1.17
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	0.0	100.0	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	0.7	67.2	NA	1.49
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	2.2	47.3	NA	2.11
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	4.2	45.3	NA	2.21
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	24.3	37.8	NA	2.65
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	168.4	20.2	NA	4.96
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	0.0	100.0	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	0.8	60.7	NA	1.65
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	2.2	46.4	NA	2.15
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	4.2	37.3	NA	2.68
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	24.3	37.8	NA	2.65
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	168.5	13.7	NA	7.33
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	0.0	97.5	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	0.5	77.6	NA	1.26
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	2.0	25.1	NA	3.88
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	4.0	22.3	NA	4.37
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	24.0	22.7	NA	4.29
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	168.0	8.7	NA	11.27
5.6 M Na+ (0.005 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.0	100.0	NA	1.00
5.6 M Na+ (0.005 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.5	91.2	NA	1.10

5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	2.0	87.7	NA	1.14
5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	4.0	92.3	NA	1.08
5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	24.0	90.9	NA	1.10
5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	168.0	94.0	NA	1.06
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	0.0	100.0	NA	1.00
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	0.5	93.8	NA	0.97
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	2.0	97.0	NA	0.98
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	4.0	95.8	NA	0.93
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	24.0	100.4	NA	1.00
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	168.0	94.0	NA	1333.91

**Data from the first set of experiments:**

**PuTTA Data****Description**

	<b>Average Time (h)</b>	<b>Pu (ug/L)</b>	<b>Pu (ug/L) std dev</b>	<b>Pu DF</b>
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	0.0	97.5	NA	1.00
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	0.5	96.1	NA	1.01
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	2.0	39.9	NA	2.44
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	4.0	28.6	NA	3.41
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	24.0	19.8	NA	4.93
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	168.0	6.8	NA	14.42
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	0.0	219.5	NA	1.00
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	0.6	67.6	NA	3.25
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	2.0	48.6	NA	4.51
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	4.0	44.0	NA	4.99
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	24.0	19.4	NA	11.32
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	168.0	6.8	NA	32.33
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	0.0	97.5	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	0.5	67.2	NA	1.45
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	2.0	27.7	NA	3.52
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	4.0	25.9	NA	3.76
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	24.0	27.1	NA	3.60
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	168.0	9.7	NA	10.07
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	0.0	97.5	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	0.5	86.8	NA	1300.31
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	2.0	37.9	NA	1300.31
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	4.0	9.6	NA	1300.31
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	24.0	9.4	NA	1300.31
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	168.0	6.7	NA	1300.31
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	0.0	218.0	0.00	1.00
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	1.1	6.3	0.14	34.69
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	2.7	6.4	0.49	33.96
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	4.7	6.9	0.41	31.67
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	25.1	7.2	1.15	30.40
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	169.5	8.7	0.58	24.94
5.6 M Na+ Control with 200 ppb Pu	0.0	216.9	NA	1.00
5.6 M Na+ Control with 200 ppb Pu	0.5	215.7	NA	1.01
5.6 M Na+ Control with 200 ppb Pu	2.0	261.6	NA	0.83
5.6 M Na+ Control with 200 ppb Pu	4.0	220.8	NA	0.98
5.6 M Na+ Control with 200 ppb Pu	24.0	191.4	NA	1.13
5.6 M Na+ Control with 200 ppb Pu	168.0	227.1	NA	0.96
5.6 M Na+ Control with 100 ppb Pu	0.0	97.5	NA	1.00
5.6 M Na+ Control with 100 ppb Pu	0.6	77.6	NA	1.26
5.6 M Na+ Control with 100 ppb Pu	2.1	103.0	NA	0.95
5.6 M Na+ Control with 100 ppb Pu	4.1	100.3	NA	0.97
5.6 M Na+ Control with 100 ppb Pu	24.2	101.8	NA	0.96
5.6 M Na+ Control with 100 ppb Pu	168.0	90.7	NA	1.07

**Data from the first set of experiments:**

## ICP-MS Data

Description	Average Time (h)	Pu (ug/L)	Pu (ug/L) std dev	Pu DE
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	0.0	130.2	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	0.5	31.6	NA	4.12
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	2.0	0.0	NA	1735.33
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	4.0	0.0	NA	1735.33
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	24.0	0.0	NA	1735.33
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	168.0	29.6	NA	4.40
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.0	96.1	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.6	84.4	NA	1.14
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	2.0	100.7	NA	0.95
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	4.0	86.6	NA	1.11
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	24.0	81.2	NA	1.18
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	168.2	96.5	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	0.0	96.1	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	0.6	66.2	NA	1.45
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	2.1	68.2	NA	1.41
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	4.1	53.6	NA	1.79
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	24.1	47.1	NA	2.04
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	168.3	27.7	NA	3.47
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	0.0	96.1	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	0.6	68.0	NA	1.41
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	2.1	70.2	NA	1.37
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	4.1	62.9	NA	1.53
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	24.1	62.6	NA	1.53
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	168.3	48.6	NA	1.98
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	0.0	96.1	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	0.7	79.8	NA	1.20
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	2.1	88.2	NA	1.09
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	4.1	79.3	NA	1.21
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	24.2	76.8	NA	1.25
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	168.4	87.3	NA	1.10
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	0.0	96.1	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	0.7	72.8	NA	1.32
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	2.2	48.1	NA	2.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	4.2	54.6	NA	1.76
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	24.3	37.5	NA	2.56
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	168.4	26.1	NA	3.68
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	0.0	96.1	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	0.8	64.3	NA	1.49
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	2.2	54.3	NA	1.77
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	4.2	44.5	NA	2.16
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	24.3	37.5	NA	2.56
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	168.5	20.5	NA	4.69
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	0.0	130.2	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	0.5	94.7	NA	1.37
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	2.0	40.7	NA	3.20
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	4.0	32.0	NA	4.07
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	24.0	41.5	NA	3.14
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	168.0	0.0	NA	1735.33
5.6 M Na+ (0.005 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.0	96.1	NA	1.00
5.6 M Na+ (0.005 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.5	98.6	NA	0.97

5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	2.0	98.0	NA	0.98
5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	4.0	93.9	NA	1.02
5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	24.0	89.1	NA	1.08
5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	168.0	97.9	NA	0.98
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	0.0	96.1	NA	1.00
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	0.5	96.8	NA	0.99
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	2.0	104.5	NA	0.92
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	4.0	114.3	NA	0.84
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	24.0	98.4	NA	0.98
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	168.0	110.4	NA	0.87

**Data from the first set of experiments:**

## ICP-MS Data

Description	Average Time (h)	Pu (ug/L)	Pu (ug/L) std dev	Pu DE
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	0.0	130.2	NA	0.00
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	0.5	62.1	NA	2.10
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	2.0	65.1	NA	2.00
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	4.0	38.8	NA	3.35
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	24.0	38.1	NA	3.41
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	168.0	0.0	NA	1735.33
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	0.0	228.5	NA	1.00
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	0.6	71.6	NA	3.19
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	2.0	48.3	NA	4.73
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	4.0	39.8	NA	5.74
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	24.0	28.5	NA	8.02
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	168.0	7.0	NA	32.74
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO <sub>4</sub> / 0.04 M Formate)	0.0	130.2	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO <sub>4</sub> / 0.04 M Formate)	0.5	80.2	NA	1.62
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO <sub>4</sub> / 0.04 M Formate)	2.0	40.7	NA	3.20
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO <sub>4</sub> / 0.04 M Formate)	4.0	38.8	NA	3.35
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO <sub>4</sub> / 0.04 M Formate)	24.0	50.2	NA	2.59
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO <sub>4</sub> / 0.04 M Formate)	168.0	20.6	NA	6.33
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	0.0	130.2	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	0.5	118.6	NA	1.10
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	2.0	56.9	NA	2.29
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	4.0	0.0	NA	1735.33
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	24.0	16.2	NA	8.03
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	168.0	0.0	NA	1735.33
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO <sub>4</sub> / 4 x 0.015 M H <sub>2</sub> O <sub>2</sub> ) with 200 ppb	0.0	238.0	0.00	1.00
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO <sub>4</sub> / 4 x 0.015 M H <sub>2</sub> O <sub>2</sub> ) with 200 ppb	1.1	10.7	0.06	22.16
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO <sub>4</sub> / 4 x 0.015 M H <sub>2</sub> O <sub>2</sub> ) with 200 ppb	2.7	8.7	12.28	27.42
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO <sub>4</sub> / 4 x 0.015 M H <sub>2</sub> O <sub>2</sub> ) with 200 ppb	4.7	5.0	7.01	47.98
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO <sub>4</sub> / 4 x 0.015 M H <sub>2</sub> O <sub>2</sub> ) with 200 ppb	25.1	11.9	1.74	19.98
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO <sub>4</sub> / 4 x 0.015 M H <sub>2</sub> O <sub>2</sub> ) with 200 ppb	169.5	11.4	1.84	20.91
5.6 M Na+ Control with 200 ppb Pu	0.0	238.4	NA	1.00
5.6 M Na+ Control with 200 ppb Pu	0.5	251.4	NA	0.95
5.6 M Na+ Control with 200 ppb Pu	2.0	248.5	NA	0.96
5.6 M Na+ Control with 200 ppb Pu	4.0	238.1	NA	1.00
5.6 M Na+ Control with 200 ppb Pu	24.0	267.0	NA	0.89
5.6 M Na+ Control with 200 ppb Pu	168.0	234.7	NA	1.02
5.6 M Na+ Control with 100 ppb Pu	0.0	130.2	NA	1.00
5.6 M Na+ Control with 100 ppb Pu	0.6	128.6	NA	1.01
5.6 M Na+ Control with 100 ppb Pu	2.1	96.0	NA	1.36
5.6 M Na+ Control with 100 ppb Pu	4.1	114.0	NA	1.14
5.6 M Na+ Control with 100 ppb Pu	24.2	111.5	NA	1.17
5.6 M Na+ Control with 100 ppb Pu	168.0	151.9	NA	0.86

Data from the first set of experiments:

## ICP-MS Data

## Description

	Average Time (h)	U (ug/L)	U (ug/L) std dev	U DE
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	0.0	13182	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	0.5	12736	NA	1.04
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	2.0	12267	NA	1.07
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	4.0	9121	NA	1.45
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	24.0	7899	NA	1.67
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	168.0	10872	NA	1.21
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.0	10955	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.6	10307	NA	1.06
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	2.0	10115	NA	1.08
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	4.0	10483	NA	1.05
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	24.0	9836	NA	1.11
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	168.2	9670	NA	1.13
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	0.0	10955	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	0.6	10901	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	2.1	10258	NA	1.07
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	4.1	10644	NA	1.03
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	24.1	9827	NA	1.11
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	168.3	9851	NA	1.11
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	0.0	10955	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	0.6	10803	NA	1.01
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	2.1	10787	NA	1.02
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	4.1	11229	NA	0.98
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	24.1	10755	NA	1.02
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	168.3	10891	NA	1.01
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	0.0	10955	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	0.7	10653	NA	1.03
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	2.1	9709	NA	1.13
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	4.1	10150	NA	1.08
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	24.2	9699	NA	1.13
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	168.4	9104	NA	1.20
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	0.0	10955	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	0.7	10402	NA	1.05
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	2.2	9194	NA	1.19
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	4.2	10002	NA	1.10
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	24.3	9577	NA	1.14
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	168.4	9820	NA	1.12
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	0.0	10955	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	0.8	10439	NA	1.05
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	2.2	9281	NA	1.18
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	4.2	9902	NA	1.11
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	24.3	9577	NA	1.14
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	168.5	9767	NA	1.12
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	0.0	13182	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	0.5	11499	NA	1.15
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	2.0	11338	NA	1.16
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	4.0	11577	NA	1.14
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	24.0	11775	NA	1.12
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	168.0	11370	NA	1.16
5.6 M Na+ (0.005 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.0	10955	NA	1.00
5.6 M Na+ (0.005 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.5	10865	NA	1.01

5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	2.0	10346	NA	1.06
5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	4.0	10776	NA	1.02
5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	24.0	10114	NA	1.08
5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	168.0	9605	NA	1.14
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	0.0	10955	NA	1.00
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	0.5	11382	NA	0.96
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	2.0	11376	NA	0.96
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	4.0	11888	NA	0.92
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	24.0	9654	NA	1.13
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	168.0	10434	NA	1.05

**Data from the first set of experiments:**

<u>Description</u>	<u>Average Time (h)</u>	<u>U (ug/L)</u>	<u>U (ug/L) std dev</u>	<u>U DF</u>
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	0.0	13182	NA	1.00
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	0.5	12488	NA	1.06
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	2.0	12672	NA	1.04
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	4.0	12771	NA	1.03
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	24.0	11740	NA	1.12
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	168.0	10766	NA	1.22
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	0.0	10999	NA	1.00
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	0.6	10562	NA	1.04
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	2.0	10245	NA	1.07
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	4.0	10173	NA	1.08
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	24.0	9657	NA	1.14
5.6 M Na+ (MST Lot #33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	168.0	8077	NA	1.36
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	0.0	13182	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	0.5	11635	NA	1.13
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	2.0	11484	NA	1.15
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	4.0	12165	NA	1.08
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	24.0	11717	NA	1.12
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	168.0	11121	NA	1.19
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	0.0	13182	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	0.5	13197	NA	1.00
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	2.0	12570	NA	1.05
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	4.0	10824	NA	1.22
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	24.0	8187	NA	1.61
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	168.0	9964	NA	1.32
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	0.0	11100	0.00	1.00
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	1.1	6243	397.42	1.78
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	2.7	7662	1951.88	1.45
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	4.7	6132	506.15	1.81
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	25.1	5890	2202.27	1.88
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	168.0	10144	604.89	1.09
5.6 M Na+ Control with 200 ppb Pu	0.0	11164.0	NA	1.00
5.6 M Na+ Control with 200 ppb Pu	0.5	11656.9	NA	0.96
5.6 M Na+ Control with 200 ppb Pu	2.0	11683.0	NA	0.96
5.6 M Na+ Control with 200 ppb Pu	4.0	10908.9	NA	1.02
5.6 M Na+ Control with 200 ppb Pu	24.0	11818.4	NA	0.94
5.6 M Na+ Control with 200 ppb Pu	168.0	11833.1	NA	0.94
5.6 M Na+ Control with 100 ppb Pu	0.0	13182	NA	1.00
5.6 M Na+ Control with 100 ppb Pu	0.6	13010	NA	1.01
5.6 M Na+ Control with 100 ppb Pu	2.1	9566	NA	1.38
5.6 M Na+ Control with 100 ppb Pu	4.1	9180	NA	1.44
5.6 M Na+ Control with 100 ppb Pu	24.2	11660	NA	1.13
5.6 M Na+ Control with 100 ppb Pu	168.0	12245	NA	1.08

**Data from the first set of experiments:**

<u>Description</u>	<u>Average Time (h)</u>	<u>Np</u>	
		<u>(ug/L)</u>	<u>std dev</u>
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	0.0	511.1	NA
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	0.5	322.6	NA
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	2.0	207.7	NA
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	4.0	122.1	NA
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	24.0	49.4	NA
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn(II))	168.0	297.3	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.0	434.0	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.6	413.0	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	2.0	410.3	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	4.0	404.0	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	24.0	358.4	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	168.2	360.3	NA
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	0.0	434.0	NA
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	0.6	440.1	NA
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	2.1	406.1	NA
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	4.1	415.2	NA
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	24.1	409.6	NA
5.6 M Na+ (0.01 M Sr/ 0.0065 M MnO4/ 0.015 M Formate)	168.3	371.1	NA
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	0.0	434.0	NA
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	0.6	423.6	NA
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	2.1	457.0	NA
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	4.1	441.9	NA
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	24.1	462.6	NA
5.6 M Na+ (0.01 M Sr/ 0.0013 M MnO4/ 0.015 M Formate)	168.3	376.3	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	0.0	434.0	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	0.7	438.2	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	2.1	407.3	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	4.1	411.5	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	24.2	395.7	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.02 M Formate)	168.4	356.8	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	0.0	434.0	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	0.7	434.1	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	2.2	386.2	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	4.2	409.5	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	24.3	409.2	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.03 M Formate)	168.4	364.8	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	0.0	434.0	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	0.8	416.1	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	2.2	382.4	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	4.2	404.6	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	24.3	409.2	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ 0.04 M Formate)	168.5	342.7	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	0.0	511.1	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	0.5	489.9	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	2.0	444.6	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	4.0	454.4	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	24.0	451.0	NA
5.6 M Na+ (0.01 M Sr/ 0.013 M MnO4/ >0.04 M Formate)	168.0	325.6	NA
5.6 M Na+ (0.005 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.0	434.0	NA
5.6 M Na+ (0.005 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	0.5	443.7	NA
5.6 M Na+ (0.005 M Sr/ 0.013 M MnO4/ 0.015 M Formate)	2.0	418.0	NA

5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	4.0	429.6	NA	1.01
5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	24.0	415.5	NA	1.04
5.6 M Na <sup>+</sup> (0.005 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	168.0	371.8	NA	1.17
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	0.0	434.0	NA	1.00
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	0.5	458.9	NA	0.95
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	2.0	485.4	NA	0.89
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	4.0	477.6	NA	0.91
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	24.0	395.0	NA	1.10
5.6 M Na <sup>+</sup> (0.001 M Sr/ 0.013 M MnO <sub>4</sub> / 0.015 M Formate)	168.0	425.4	NA	1.02

**Data from the first set of experiments:**

<u>Description</u>	<u>Average Time (h)</u>	<u>Np</u>	
		<u>(ug/L)</u>	<u>std dev</u>
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	0.0	511.1	NA
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	0.5	437.3	NA
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	2.0	404.7	NA
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	4.0	414.6	NA
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	24.0	258.3	NA
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 100 ppb Pu	168.0	206.7	NA
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	0.0	460.7	NA
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	0.6	369.7	NA
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	2.0	297.5	NA
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	4.0	251.9	NA
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	24.0	132.8	NA
5.6 M Na+ (MST Lot#33180 at 0.4 g L <sup>-1</sup> ) with 200 ppb Pu	168.0	57.0	NA
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	0.0	511.1	NA
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	0.5	495.7	NA
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	2.0	447.3	NA
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	4.0	485.6	NA
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	24.0	473.2	NA
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M MnO4/ 0.04 M Formate)	168.0	346.8	NA
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	0.0	511.1	NA
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	0.5	451.2	NA
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	2.0	324.8	NA
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	4.0	184.6	NA
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	24.0	134.9	NA
5.6 M Na+ (0.01 M Sr/ 0.005 M Ca/ 0.01 M Mn(II))	168.0	192.2	NA
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	0.0	472.0	0.00
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	1.1	37.3	10.03
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	2.7	43.5	17.11
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	4.7	43.3	8.36
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	25.1	59.8	8.94
5.6 M Na+ (0.01 M Sr/ 0.01 M MnO4/ 4 x 0.015 M H2O2) with 200 ppb Pu	169.5	100.7	5.01
5.6 M Na+ Control with 200 ppb Pu	0.0	481.4	NA
5.6 M Na+ Control with 200 ppb Pu	0.5	504.5	NA
5.6 M Na+ Control with 200 ppb Pu	2.0	498.1	NA
5.6 M Na+ Control with 200 ppb Pu	4.0	473.4	NA
5.6 M Na+ Control with 200 ppb Pu	24.0	512.3	NA
5.6 M Na+ Control with 200 ppb Pu	168.0	466.4	NA
5.6 M Na+ Control with 100 ppb Pu	0.0	511.1	NA
5.6 M Na+ Control with 100 ppb Pu	0.6	499.4	NA
5.6 M Na+ Control with 100 ppb Pu	2.1	384.0	NA
5.6 M Na+ Control with 100 ppb Pu	4.1	360.0	NA
5.6 M Na+ Control with 100 ppb Pu	24.2	463.9	NA
5.6 M Na+ Control with 100 ppb Pu	168.0	481.6	NA

**First half of the data from the second set of experiments:**

<b>Description</b>	<b>Average Time (h)</b>	<b>Sr-85 ng/L</b>	<b>Sr-85 std dev</b>	<b>Sr-85 DF</b>
Control 5.6 M Na+	0.1	1.327	0.023	1.00
Control 5.6 M Na+	168.0	1.298	0.003	1.02
Control 5.6 M Na+	0.1	1.327	0.023	1.00
5.6 M Na+ (0.01 M Sr/0.045 M Formate/No Mn)	4.5	0.432	0.027	30.73
5.6 M Na+ (0.01 M Sr/0.045 M Formate/No Mn)	24.7	0.195	0.009	67.88
5.6 M Na+ (0.01 M Sr/0.045 M Formate/No Mn)	166.1	0.033	0.004	404.74
Control 5.6 M Na+	0.1	1.327	0.023	1.00
5.6 M Na+ (0.01 M Sr/0.009 M Formate/0.002 M Mn)	4.6	0.151	0.012	87.83
5.6 M Na+ (0.01 M Sr/0.009 M Formate/0.002 M Mn)	24.8	0.070	0.005	190.85
5.6 M Na+ (0.01 M Sr/0.009 M Formate/0.002 M Mn)	166.2	0.026	0.000	508.71
Control 5.6 M Na+	0.1	1.327	0.023	1.00
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.005 M Mn)	4.6	0.139	0.001	95.05
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.005 M Mn)	24.8	0.024	0.002	553.72
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.005 M Mn)	166.2	0.018	0.001	759.15
Control 5.6 M Na+	0.1	1.327	0.023	1.00
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.01 M Mn)	4.6	0.091	0.002	146.36
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.01 M Mn)	24.9	0.016	0.002	852.31
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.01 M Mn)	166.2	0.010	0.001	1358.78
Control 5.6 M Na+	0.1	1.327	0.023	1.00
5.6 M Na+ 1/4Mn(X4) (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	6.8	0.088	0.004	150.32
5.6 M Na+ 1/4Mn(X4) (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	27.1	0.014	0.000	957.99
5.6 M Na+ 1/4Mn(X4) (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	168.4	0.010	0.001	1281.60
Control 5.6 M Na+	0.1	1.327	0.023	1.00
5.6 M Na+ (No Sr/0.01 M Mn/0.045 M Formate)	4.7	0.000	0.000	41236.64
5.6 M Na+ (No Sr/0.01 M Mn/0.045 M Formate)	24.9	0.000	0.000	56976.45
5.6 M Na+ (No Sr/0.01 M Mn/0.045 M Formate)	166.3	0.000	0.000	90688.69
Control 5.6 M Na+	0.1	1.327	0.023	1.00
5.6 M Na+ (0.001 M Sr/0.01 M Mn/0.045 M Formate)	4.7	0.006	0.001	2228.01
5.6 M Na+ (0.001 M Sr/0.01 M Mn/0.045 M Formate)	24.9	0.001	0.000	12624.86
5.6 M Na+ (0.001 M Sr/0.01 M Mn/0.045 M Formate)	166.4	0.002	0.000	5867.43

**First half of the data from the second set of experiments:**

**PuTTA Data**

<b>Description</b>	<b>Average Time (h)</b>	<b>Pu (ug/L)</b>		
		<b>Pu (ug/L)</b>	<b>Standard Deviation</b>	<b>Pu DE</b>
Control 5.6 M Na+	0.1	196.6	4.5	1.0
Control 5.6 M Na+	166.1	185.2	0.9	1.1
Control 5.6 M Na+	0.1	196.6	4.5	1.0
5.6 M Na+ (0.01 M Sr/0.045 M Formate/No Mn)	4.5	170.0	6.3	1.2
5.6 M Na+ (0.01 M Sr/0.045 M Formate/No Mn)	24.7	170.7	3.6	1.2
5.6 M Na+ (0.01 M Sr/0.045 M Formate/No Mn)	166.1	85.5	1.6	2.3
Control 5.6 M Na+	0.1	196.6	4.5	1.0
5.6 M Na+ (0.01 M Sr/0.009 M Formate/0.002 M Mn)	4.6	114.6	4.2	1.7
5.6 M Na+ (0.01 M Sr/0.009 M Formate/0.002 M Mn)	24.8	86.3	1.7	2.3
5.6 M Na+ (0.01 M Sr/0.009 M Formate/0.002 M Mn)	166.2	46.6	0.1	4.2
Control 5.6 M Na+	0.1	196.6	4.5	1.0
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.005 M Mn)	4.6	42.2	1.0	4.7
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.005 M Mn)	24.8	46.3	0.2	4.2
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.005 M Mn)	166.2	19.2	0.2	10.2
Control 5.6 M Na+	0.1	196.6	4.5	1.0
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.01 M Mn)	4.6	51.3	3.7	3.8
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.01 M Mn)	24.9	54.9	6.7	3.6
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.01 M Mn)	166.2	17.6	0.5	11.2
Control 5.6 M Na+	0.1	196.6	4.5	1.0
5.6 M Na+ (1/4Mn(X4)) (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	6.8	10.3	0.6	19.1
5.6 M Na+ (1/4Mn(X4)) (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	27.1	17.3	1.3	11.4
5.6 M Na+ (1/4Mn(X4)) (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	168.4	10.5	0.6	18.8
Control 5.6 M Na+	0.1	196.6	4.5	1.0
5.6 M Na+ (No Sr/0.01 M Mn/0.045 M Formate)	4.7	82.7	22.3	2.5
5.6 M Na+ (No Sr/0.01 M Mn/0.045 M Formate)	24.9	77.7	11.0	2.6
5.6 M Na+ (No Sr/0.01 M Mn/0.045 M Formate)	166.3	63.7	2.5	3.1
Control 5.6 M Na+	0.1	196.6	4.5	1.0
5.6 M Na+ (0.001 M Sr/0.01 M Mn/0.045 M Formate)	4.7	65.5	2.6	3.0
5.6 M Na+ (0.001 M Sr/0.01 M Mn/0.045 M Formate)	24.9	79.6	25.5	2.6
5.6 M Na+ (0.001 M Sr/0.01 M Mn/0.045 M Formate)	166.4	49.3	3.6	4.0

**First half of the data from the second set of experiments:**

## ICP-MS Data

Description	Pu ( $\mu$ g/L)			
	Average Time (h)	Pu ( $\mu$ g/L)	Standard Deviation	DE
Control 5.6 M Na <sup>+</sup>	0.0	188.0	27.0	1.0
Control 5.6 M Na <sup>+</sup>	168.0	173.3	11.8	1.1
Control 5.6 M Na <sup>+</sup>	0.0	188.0	4.5	1.0
5.6 M Na <sup>+</sup> (0.01 M Sr/0.045 M Formate/No Mn)	4.5	188.9	6.1	1.0
5.6 M Na <sup>+</sup> (0.01 M Sr/0.045 M Formate/No Mn)	24.7	197.0	9.9	1.0
5.6 M Na <sup>+</sup> (0.01 M Sr/0.045 M Formate/No Mn)	166.1	63.7	7.9	3.0
Control 5.6 M Na <sup>+</sup>	0.0	188.0	4.5	1.0
5.6 M Na <sup>+</sup> (0.01 M Sr/0.009 M Formate/0.002 M Mn)	4.6	113.3	0.6	1.7
5.6 M Na <sup>+</sup> (0.01 M Sr/0.009 M Formate/0.002 M Mn)	24.8	90.0	0.0	2.1
5.6 M Na <sup>+</sup> (0.01 M Sr/0.009 M Formate/0.002 M Mn)	166.2	40.4	0.6	4.7
Control 5.6 M Na <sup>+</sup>	0.0	188.0	4.5	1.0
5.6 M Na <sup>+</sup> (0.01 M Sr/0.045 M Formate/0.005 M Mn)	4.6	52.2	3.0	3.6
5.6 M Na <sup>+</sup> (0.01 M Sr/0.045 M Formate/0.005 M Mn)	24.8	47.0	1.4	4.0
5.6 M Na <sup>+</sup> (0.01 M Sr/0.045 M Formate/0.005 M Mn)	166.2	21.1	2.9	9.0
Control 5.6 M Na <sup>+</sup>	0.0	188.0	4.5	1.0
5.6 M Na <sup>+</sup> (0.01 M Sr/0.045 M Formate/0.01 M Mn)	4.6	54.4	5.1	3.5
5.6 M Na <sup>+</sup> (0.01 M Sr/0.045 M Formate/0.01 M Mn)	24.9	50.0	8.5	3.8
5.6 M Na <sup>+</sup> (0.01 M Sr/0.045 M Formate/0.01 M Mn)	166.2	20.0	0.0	9.4
Control 5.6 M Na <sup>+</sup>	0.0	188.0	4.5	1.0
5.6 M Na <sup>+</sup> (1/4Mn(X4)) (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	6.8	15.9	3.0	12.0
5.6 M Na <sup>+</sup> (1/4Mn(X4)) (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	27.1	15.0	7.1	14.1
5.6 M Na <sup>+</sup> (1/4Mn(X4)) (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	168.4	20.0	0.0	9.4
Control 5.6 M Na <sup>+</sup>	0.0	188.0	4.5	1.0
5.6 M Na <sup>+</sup> (No Sr/0.01 M Mn/0.045 M Formate)	4.7	88.0	39.6	2.4
5.6 M Na <sup>+</sup> (No Sr/0.01 M Mn/0.045 M Formate)	24.9	87.0	32.5	2.3
5.6 M Na <sup>+</sup> (No Sr/0.01 M Mn/0.045 M Formate)	166.3	56.7	5.1	3.3
Control 5.6 M Na <sup>+</sup>	0.0	188.0	4.5	1.0
5.6 M Na <sup>+</sup> (0.001 M Sr/0.01 M Mn/0.045 M Formate)	4.7	63.0	4.2	3.0
5.6 M Na <sup>+</sup> (0.001 M Sr/0.01 M Mn/0.045 M Formate)	24.9	59.0	29.7	3.6
5.6 M Na <sup>+</sup> (0.001 M Sr/0.01 M Mn/0.045 M Formate)	166.4	47.9	11.2	4.0

First half of the data from the second set of experiments:

**ICP-MS Data**

<b>Description</b>	<b>Average Time (h)</b>	<b>U (ug/L)</b>		
		<b>U (ug/L)</b>	<b>Standard Deviation</b>	<b>U DF</b>
Control 5.6 M Na+	0.0	10499	288.9	1.0
Control 5.6 M Na+	168.0	10075	335.1	1.0
Control 5.6 M Na+	0.0	10499	288.9	1.0
5.6 M Na+ (0.01 M Sr/0.045 M Formate/No Mn)	4.5	10370	212.3	1.0
5.6 M Na+ (0.01 M Sr/0.045 M Formate/No Mn)	24.7	11409	123.0	0.9
5.6 M Na+ (0.01 M Sr/0.045 M Formate/No Mn)	166.1	8972	1164.4	1.2
Control 5.6 M Na+	0.0	10499	288.9	1.0
5.6 M Na+ (0.01 M Sr/0.009 M Formate/0.002 M Mn)	4.6	10317	159.8	1.0
5.6 M Na+ (0.01 M Sr/0.009 M Formate/0.002 M Mn)	24.8	10548	299.8	1.0
5.6 M Na+ (0.01 M Sr/0.009 M Formate/0.002 M Mn)	166.2	9762	104.9	1.1
Control 5.6 M Na+	0.0	10499	288.9	1.0
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.005 M Mn)	4.6	9375	219.7	1.1
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.005 M Mn)	24.8	10468	5.7	1.0
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.005 M Mn)	166.2	10045	1036.5	1.1
Control 5.6 M Na+	0.0	10499	288.9	1.0
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.01 M Mn)	4.6	9755	564.1	1.1
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.01 M Mn)	24.9	10069	270.1	1.0
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.01 M Mn)	166.2	8458	394.4	1.2
Control 5.6 M Na+	0.0	10499	288.9	1.0
5.6 M Na+ 1/4Mn(X4) (0.01 M Sr/ 0.045 M Formate/ 0.01 M Mn)	6.8	10117	134.4	1.0
5.6 M Na+ 1/4Mn(X4) (0.01 M Sr/ 0.045 M Formate/ 0.01 M Mn)	27.1	9896	149.9	1.1
5.6 M Na+ 1/4Mn(X4) (0.01 M Sr/ 0.045 M Formate/ 0.01 M Mn)	168.4	8802	207.4	1.2
Control 5.6 M Na+	0.0	10499	288.9	1.0
5.6 M Na+ (No Sr/0.01 M Mn/0.045 M Formate)	4.7	9381	3016.5	1.2
5.6 M Na+ (No Sr/0.01 M Mn/0.045 M Formate)	24.9	11038	48.1	1.0
5.6 M Na+ (No Sr/0.01 M Mn/0.045 M Formate)	166.3	9563	488.1	1.1
Control 5.6 M Na+	0.0	10499	288.9	1.0
5.6 M Na+ (0.001 M Sr/0.01 M Mn/0.045 M Formate)	4.7	10717	46.7	1.0
5.6 M Na+ (0.001 M Sr/0.01 M Mn/0.045 M Formate)	24.9	10814	76.4	1.0
5.6 M Na+ (0.001 M Sr/0.01 M Mn/0.045 M Formate)	166.4	10265	946.7	1.0

**First half of the data from the second set of experiments:**

## ICP-MS Data

Description	Average Time (h)	Np (ug/L)		
		Np (ug/L)	Standard Deviation	Np DF
Control 5.6 M Na+	0.0	428.4	8.0	1.0
Control 5.6 M Na+	168.0	406.9	17.4	1.3
Control 5.6 M Na+	0.0	428.4	8.0	1.0
5.6 M Na+ (0.01 M Sr/0.045 M Formate/No Mn)	4.5	392.1	9.5	1.1
5.6 M Na+ (0.01 M Sr/0.045 M Formate/No Mn)	24.7	396.0	5.7	1.1
5.6 M Na+ (0.01 M Sr/0.045 M Formate/No Mn)	166.1	277.3	36.0	1.6
Control 5.6 M Na+	0.0	428.4	8.0	1.0
5.6 M Na+ (0.01 M Sr/0.009 M Formate/0.002 M Mn)	4.6	417.7	5.0	1.0
5.6 M Na+ (0.01 M Sr/0.009 M Formate/0.002 M Mn)	24.8	389.0	15.6	1.1
5.6 M Na+ (0.01 M Sr/0.009 M Formate/0.002 M Mn)	166.2	306.4	7.2	1.4
Control 5.6 M Na+	0.0	428.4	8.0	1.0
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.005 M Mn)	4.6	370.7	2.6	1.2
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.005 M Mn)	24.8	370.0	17.0	1.2
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.005 M Mn)	167.3	264.3	31.1	1.6
Control 5.6 M Na+	0.0	428.4	8.0	1.0
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.01 M Mn)	4.6	381.5	23.3	1.1
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.01 M Mn)	24.9	370.0	2.8	1.2
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.01 M Mn)	166.2	285.2	10.5	1.5
Control 5.6 M Na+	0.0	428.4	8.0	1.0
5.6 M Na+ 1/4Mn(X4) (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	6.8	365.0	12.7	1.2
5.6 M Na+ 1/4Mn(X4) (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	27.1	348.0	0.0	1.2
5.6 M Na+ 1/4Mn(X4) (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	168.4	231.1	9.7	1.9
Control 5.6 M Na+	0.0	428.4	8.0	1.0
5.6 M Na+ (No Sr/0.01 M Mn/0.045 M Formate)	4.7	353.0	111.7	1.3
5.6 M Na+ (No Sr/0.01 M Mn/0.045 M Formate)	24.9	392.0	11.3	1.1
5.6 M Na+ (No Sr/0.01 M Mn/0.045 M Formate)	166.3	351.5	8.1	1.2
Control 5.6 M Na+	0.0	428.4	8.0	1.0
5.6 M Na+ (0.001 M Sr/0.01 M Mn/0.045 M Formate)	4.7	391.0	7.1	1.1
5.6 M Na+ (0.001 M Sr/0.01 M Mn/0.045 M Formate)	24.9	390.0	8.5	1.1
5.6 M Na+ (0.001 M Sr/0.01 M Mn/0.045 M Formate)	166.4	369.0	48.0	1.2

**First half of the data from the second set of experiments:**

**ICPMS Data**

<b>Description</b>	<b>Stable</b>		
	<b>Average</b>	<b>Sr</b>	
	<b>Time (h)</b>	<b>ug/L</b>	<b>std dev</b>
Control 5.6 M Na+	0.0	633	10.3
Control 5.6 M Na+	168.0	575	25.5
Control 5.6 M Na+	0.0	633	4.5
5.6 M Na+ (0.01 M Sr/0.045 M Formate/No Mn)	4.5	107126	14614.5
5.6 M Na+ (0.01 M Sr/0.045 M Formate/No Mn)	24.7	115496	7545.9
5.6 M Na+ (0.01 M Sr/0.045 M Formate/No Mn)	166.1	18980	3516.9
Control 5.6 M Na+	0.0	633	4.5
5.6 M Na+ (0.01 M Sr/0.009 M Formate/0.002 M Mn)	4.6	53648	79.2
5.6 M Na+ (0.01 M Sr/0.009 M Formate/0.002 M Mn)	24.8	49155	6714.4
5.6 M Na+ (0.01 M Sr/0.009 M Formate/0.002 M Mn)	166.2	13138	472.4
Control 5.6 M Na+	0.0	633	4.5
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.005 M Mn)	4.6	60771	855.6
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.005 M Mn)	24.8	16838	965.2
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.005 M Mn)	166.2	9720	469.4
Control 5.6 M Na+	0.0	633	4.5
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.01 M Mn)	4.6	56105	4062.5
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.01 M Mn)	24.9	10333	2328.9
5.6 M Na+ (0.01 M Sr/0.045 M Formate/0.01 M Mn)	166.2	5922	1068.7
Control 5.6 M Na+	0.0	633	4.5
5.6 M Na+ 1/4Mn(X4) (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	6.8	68173	8571.5
5.6 M Na+ 1/4Mn(X4) (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	27.1	10685	1879.3
5.6 M Na+ 1/4Mn(X4) (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	168.4	6495	115.8
Control 5.6 M Na+	0.0	633	4.5
5.6 M Na+ (No Sr/0.01 M Mn/0.045 M Formate)	4.7	24	0.0
5.6 M Na+ (No Sr/0.01 M Mn/0.045 M Formate)	24.9	8	0.5
5.6 M Na+ (No Sr/0.01 M Mn/0.045 M Formate)	166.3	24	0.0
Control 5.6 M Na+	0.0	633	4.5
5.6 M Na+ (0.001 M Sr/0.01 M Mn/0.045 M Formate)	4.7	42	5.7
5.6 M Na+ (0.001 M Sr/0.01 M Mn/0.045 M Formate)	24.9	113	0.4
5.6 M Na+ (0.001 M Sr/0.01 M Mn/0.045 M Formate)	166.4	193	34.1

**Second half of the data from the second set of experiments:**

<u>Description</u>	<u>Average Time (h)</u>	<u>Sr-85 ng/L</u>	<u>Sr-85 std dev</u>	<u>Sr-85 DF</u>
Control 5.6 M Na+	0.0	1.108	0.253	1.03
5.6 M Na+ (0.045 M Formate/0.01 M Sr/ 0.01 M Mn)	4.5	0.083	0.001	13.38
5.6 M Na+ (0.045 M Formate/0.01 M Sr/ 0.01 M Mn)	24.6	0.018	0.000	62.24
5.6 M Na+ (0.045 M Formate/0.01 M Sr/ 0.01 M Mn)	166.1	0.005	0.004	298.00
Control 5.6 M Na+	0.0	1.108	0.253	1.03
5.6 M Na+ (0.045 M H2O2/ 0.01 M Sr/ 0.01 M Mn)	4.6	0.203	0.000	5.46
5.6 M Na+ (0.045 M H2O2/ 0.01 M Sr/ 0.01 M Mn)	24.6	0.031	0.003	35.94
5.6 M Na+ (0.045 M H2O2/ 0.01 M Sr/ 0.01 M Mn)	166.1	0.018	0.000	61.63
Control 5.6 M Na+	0.0	1.108	0.253	1.03
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/0.045 M Formate)	4.6	0.082	0.002	13.45
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/0.045 M Formate)	24.7	0.014	0.001	78.21
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/0.045 M Formate)	166.1	0.009	0.000	123.82
Control 5.6 M Na+	0.0	1.108	0.253	1.03
5.6 M Na+ (0.01 M Sr/0.01 M Mn/ 0.045 M H2O2)	4.7	0.159	0.001	6.95
5.6 M Na+ (0.01 M Sr/0.01 M Mn/ 0.045 M H2O2)	24.7	0.093	0.000	11.95
5.6 M Na+ (0.01 M Sr/0.01 M Mn/ 0.045 M H2O2)	166.1	0.019	0.001	59.89
Control 4.7 M Na+	0.0	1.025	0.113	1.01
4.7 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	4.7	0.078	0.001	13.10
4.7 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	24.7	0.013	0.003	78.58
4.7 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	166.1	0.009	0.001	120.71
Control 4.0 M Na+	0.0	1.059	0.073	1.00
4.0 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	4.8	0.063	0.003	16.84
4.0 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	24.8	0.009	0.001	116.67
4.0 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	166.1	0.007	0.001	163.77
Control 4.7 M Na+	0.0	1.030	0.129	1.00
Control 4.7 M Na+	166.1	1.064	0.005	0.96
Control 4.0 M Na+	0.0	1.060	0.098	1.00
Control 4.0 M Na+	166.0	0.890	0.024	1.19
Control 5.6 M Na+	0.0	1.110	0.243	1.00
Control 5.6 M Na+	166.0	1.241	0.009	0.89

**Second half of the data from the second set of experiments:**

**PuTTA Data**

<b>Description</b>	<b>Average Time (h)</b>	<b>Pu (ug/L)</b>		
		<b>Pu (ug/L)</b>	<b>Standard Deviation</b>	<b>Pu DE</b>
Control 5.6 M Na+	0.00	181.40	2.68	1.0
5.6 M Na+ (0.045 M Formate/0.01 M Sr/ 0.01 M Mn)	4.52	47.78	1.79	3.8
5.6 M Na+ (0.045 M Formate/0.01 M Sr/ 0.01 M Mn)	24.58	50.31	4.29	3.6
5.6 M Na+ (0.045 M Formate/0.01 M Sr/ 0.01 M Mn)	166.13	21.17	0.27	8.6
Control 5.6 M Na+	0.00	181.40	2.68	1.0
5.6 M Na+ (0.045 M H2O2/ 0.01 M Sr/ 0.01 M Mn)	4.60	1.93	0.03	94.1
5.6 M Na+ (0.045 M H2O2/ 0.01 M Sr/ 0.01 M Mn)	24.62	5.80	0.59	31.4
5.6 M Na+ (0.045 M H2O2/ 0.01 M Sr/ 0.01 M Mn)	166.10	0.68	0.06	266.4
Control 5.6 M Na+	0.00	181.40	2.68	1.0
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/ 0.045 M Formate)	4.64	43.17	0.63	4.2
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/ 0.045 M Formate)	24.65	52.14	1.52	3.5
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/ 0.045 M Formate)	166.08	19.72	2.32	9.3
Control 5.6 M Na+	0.00	181.40	2.68	1.0
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/ 0.045 M H2O2)	4.67	6.28	0.10	28.9
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/ 0.045 M H2O2)	24.69	5.48	0.12	33.1
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/ 0.045 M H2O2)	166.07	1.13	0.04	160.9
Control 4.7 M Na+	0.00	154.85	0.89	1.0
4.7 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	4.71	25.47	8.13	6.4
4.7 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	24.73	36.15	0.72	4.3
4.7 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	166.05	13.91	0.36	11.1
Control 4.0 M Na+	0.00	133.36	2.68	1.0
4.0 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	4.81	23.64	0.01	5.8
4.0 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	24.84	26.04	1.25	5.1
4.0 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	166.05	1.02	0.03	130.9
Control 4.7 M Na+	0.00	154.85	0.89	1.0
Control 4.7 M Na+	166.05	156.75	1.79	1.0
Control 4.0 M Na+	0.00	133.36	2.68	1.0
Control 4.0 M Na+	166.05	135.89	0.89	1.0
Control 5.6 M Na+	0.00	181.40	2.68	1.0
Control 5.6 M Na+	166.07	185.82	8.94	1.0

**Second half of the data from the second set of experiments:**

## ICP-MS Data

<u>Description</u>	<u>Average Time (h)</u>	Pu (ug/L)		
		<u>Pu (ug/L)</u>	<u>Standard Deviation</u>	<u>Pu DF</u>
Control 5.6 M Na+	0.1	203	3	1.0
5.6 M Na+ (0.045 M Formate/0.01 M Sr/ 0.01 M Mn)	4.5	54	1	3.7
5.6 M Na+ (0.045 M Formate/0.01 M Sr/ 0.01 M Mn)	24.6	36	7	5.7
5.6 M Na+ (0.045 M Formate/0.01 M Sr/ 0.01 M Mn)	166.1	20	0	10.1
Control 5.6 M Na+	0.1	203	3	1.0
5.6 M Na+ (0.045 M H2O2/ 0.01 M Sr/ 0.01 M Mn)	4.6	20	0	10.1
5.6 M Na+ (0.045 M H2O2/ 0.01 M Sr/ 0.01 M Mn)	24.6	20	0	10.1
5.6 M Na+ (0.045 M H2O2/ 0.01 M Sr/ 0.01 M Mn)	166.1	20	0	10.1
Control 5.6 M Na+	0.1	203	3	1.0
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/0.045 M Formate)	4.6	48	1	4.2
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/0.045 M Formate)	24.7	45	6	4.6
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/0.045 M Formate)	166.1	20	0	10.1
Control 5.6 M Na+	0.1	203	3	1.0
5.6 M Na+ (0.01 M Sr/0.01 M Mn/ 0.045 M H2O2)	4.7	13	1	16.1
5.6 M Na+ (0.01 M Sr/0.01 M Mn/ 0.045 M H2O2)	24.7	20	0	10.1
5.6 M Na+ (0.01 M Sr/0.01 M Mn/ 0.045 M H2O2)	166.1	20	0	10.1
Control 4.7 M Na+	0.1	168	10	1.0
4.7 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	4.7	34	6	5.0
4.7 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	24.7	38	4	4.4
4.7 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	166.1	20	0	8.4
Control 4.0 M Na+	0.1	131	12	1.0
4.0 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	4.8	23	2	5.8
4.0 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	24.8	25	4	5.4
4.0 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	166.1	20	0	6.6
Control 4.7 M Na+	0.1	168	10	1.0
Control 4.7 M Na+	166.1	158	17	1.1
Control 4.0 M Na+	0.1	131	12	1.0
Control 4.0 M Na+	166.0	137	4	1.0
Control 5.6 M Na+	0.1	203	3	1.0
Control 5.6 M Na+	166.0	177	8	1.1

**Second half of the data from the second set of experiments:**

**ICP-MS Data**

<b>Description</b>	<b>Average Time (h)</b>	<b>U (ug/L)</b>		
		<b>U (ug/L)</b>	<b>Standard Deviation</b>	<b>U DE</b>
Control 5.6 M Na+	0.0	10880	24	1.0
5.6 M Na+ (0.045 M Formate/0.01 M Sr/ 0.01 M Mn)	4.5	9154	141	1.2
5.6 M Na+ (0.045 M Formate/0.01 M Sr/ 0.01 M Mn)	24.6	8538	600	1.3
5.6 M Na+ (0.045 M Formate/0.01 M Sr/ 0.01 M Mn)	166.1	9331	58	1.2
Control 5.6 M Na+	0.0	10880	24	1.0
5.6 M Na+ (0.045 M H2O2/ 0.01 M Sr/ 0.01 M Mn)	4.6	6513	292	1.7
5.6 M Na+ (0.045 M H2O2/ 0.01 M Sr/ 0.01 M Mn)	24.6	8308	472	1.3
5.6 M Na+ (0.045 M H2O2/ 0.01 M Sr/ 0.01 M Mn)	166.1	9325	154	1.2
Control 5.6 M Na+	0.0	10880	24	1.0
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/ 0.045 M Formate)	4.6	8952	164	1.2
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/ 0.045 M Formate)	24.7	8545	344	1.3
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/ 0.045 M Formate)	166.1	9244	88	1.2
Control 5.6 M Na+	0.0	10880	24	1.0
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/ 0.045 M H2O2)	4.7	8638	263	1.3
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/ 0.045 M H2O2)	24.7	7879	423	1.4
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/ 0.045 M H2O2)	166.1	9513	57	1.1
Control 4.7 M Na+	0.0	9291	24	1.0
4.7 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	4.7	7073	72	1.3
4.7 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	24.7	6552	554	1.4
4.7 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	166.1	9250	1997	1.0
Control 4.0 M Na+	0.0	7896	110	1.0
4.0 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	4.8	5411	33	1.5
4.0 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	24.8	5245	168	1.5
4.0 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	166.1	10939	3108	0.8
Control 4.7 M Na+	0.0	9291	24	1.0
Control 4.7 M Na+	166.1	8968	285	1.0
Control 4.0 M Na+	0.0	7896	254	1.0
Control 4.0 M Na+	166.0	7659	84	1.0
Control 5.6 M Na+	0.0	11055	154	1.0
Control 5.6 M Na+	166.0	10589	84	1.0

**Second half of the data from the second set of experiments:**

**ICP-MS Data**

<b>Description</b>	<b>Average Time (h)</b>	<b>Np (ug/L)</b>		
		<b>Np (ug/L)</b>	<b>Standard Deviation</b>	<b>Np DF</b>
Control 5.6 M Na+	0.0	422.8	7.6	1.0
5.6 M Na+ (0.045 M Formate/0.01 M Sr/ 0.01 M Mn)	4.5	358.5	3.1	1.2
5.6 M Na+ (0.045 M Formate/0.01 M Sr/ 0.01 M Mn)	24.6	341.7	5.5	1.2
5.6 M Na+ (0.045 M Formate/0.01 M Sr/ 0.01 M Mn)	166.1	294.9	14.3	1.4
Control 5.6 M Na+	0.0	422.8	7.6	1.0
5.6 M Na+ (0.045 M H2O2/ 0.01 M Sr/ 0.01 M Mn)	4.6	17.1	4.2	25.4
5.6 M Na+ (0.045 M H2O2/ 0.01 M Sr/ 0.01 M Mn)	24.6	60.0	2.5	7.1
5.6 M Na+ (0.045 M H2O2/ 0.01 M Sr/ 0.01 M Mn)	166.1	116.3	0.9	3.6
Control 5.6 M Na+	0.0	422.8	7.6	1.0
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/0.045 M Formate)	4.6	365.1	11.7	1.2
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/0.045 M Formate)	24.7	359.2	7.9	1.2
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/0.045 M Formate)	166.1	303.3	3.0	1.4
Control 5.6 M Na+	0.0	422.8	7.6	1.0
5.6 M Na+ (0.01 M Sr/0.01 M Mn/ 0.045 M H2O2)	4.7	55.0	5.2	7.7
5.6 M Na+ (0.01 M Sr/0.01 M Mn/ 0.045 M H2O2)	24.7	65.4	1.7	6.5
5.6 M Na+ (0.01 M Sr/0.01 M Mn/ 0.045 M H2O2)	166.1	129.9	8.3	3.3
Control 4.7 M Na+	0.0	360.7	10.3	1.0
4.7 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	4.7	294.8	6.6	1.2
4.7 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	24.7	261.8	26.3	1.4
4.7 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	166.1	295.4	61.7	1.2
Control 4.0 M Na+	0.0	305.6	11.7	1.0
4.0 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	4.8	245.3	4.7	1.2
4.0 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	24.8	210.9	16.1	1.5
4.0 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	166.1	352.4	93.7	0.9
Control 4.0 M Na+	0.0	360.7	10.3	1.0
Control 4.0 M Na+	166.1	347.5	23.8	0.9
Control 4.7 M Na+	0.0	305.6	11.7	1.0
Control 4.7 M Na+	166.0	295.9	9.6	1.2
Control 5.6 M Na+	0.0	422.8	7.6	1.0
Control 5.6 M Na+	166.0	398.7	1.7	1.1

**Second half of the data from the second set of experiments:**

**ICPMS Data**

<u>Description</u>	<b>Stable</b>		
	<b>Average Time (h)</b>	<b>Sr ug/L</b>	<b>std dev</b>
Control 5.6 M Na+	0.1	660	2.3
5.6 M Na+ (0.045 M Formate/0.01 M Sr/ 0.01 M Mn)	4.5	54118	950.5
5.6 M Na+ (0.045 M Formate/0.01 M Sr/ 0.01 M Mn)	24.6	11583	687.8
5.6 M Na+ (0.045 M Formate/0.01 M Sr/ 0.01 M Mn)	166.1	5659	12.0
Control 5.6 M Na+	0.1	660	2.3
5.6 M Na+ (0.045 M H2O2/ 0.01 M Sr/ 0.01 M Mn)	4.6	107911	4401.3
5.6 M Na+ (0.045 M H2O2/ 0.01 M Sr/ 0.01 M Mn)	24.6	21453	297.4
5.6 M Na+ (0.045 M H2O2/ 0.01 M Sr/ 0.01 M Mn)	166.1	11507	52.5
Control 5.6 M Na+	0.1	660	2.3
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/0.045 M Formate)	4.6	52406	1026.8
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/0.045 M Formate)	24.7	10352	1211.1
5.6 M Na+ (0.01 M Sr/ 0.01 M Mn/0.045 M Formate)	166.1	5302	104.1
Control 5.6 M Na+	0.1	660	2.3
5.6 M Na+ (0.01 M Sr/0.01 M Mn/ 0.045 M H2O2)	4.7	86016	2034.7
5.6 M Na+ (0.01 M Sr/0.01 M Mn/ 0.045 M H2O2)	24.7	60583	17.0
5.6 M Na+ (0.01 M Sr/0.01 M Mn/ 0.045 M H2O2)	166.1	12404	412.1
Control 4.7 M Na+	0.1	572	3.4
4.7 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	4.7	58403	2472.0
4.7 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	24.7	9704	194.8
4.7 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	166.1	6844	1816.2
Control 4.0 M Na+	0.1	474	7.3
4.0 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	4.8	58445	2482.7
4.0 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	24.8	8329	574.9
4.0 M Na+ (0.01 M Sr/ 0.045 M Formate/0.01 M Mn)	166.1	9118	2570.1
Control 4.7 M Na+	0.1	572	3.4
Control 4.7 M Na+	166.1	231	6.2
Control 4.0 M Na+	0.1	474	7.3
Control 4.0 M Na+	166.0	24	0.0
Control 5.6 M Na+	0.1	203	3.4
Control 5.6 M Na+	166.0	314	17.6

## 9.0 References

1. Rutland, P. L. (1998). MST Alpha Removal and Hg Removal for Salt Team Phase 3 Evaluation. HLE-TAR-98062, Rev. 0, Savannah River Site, July 15, 1998.
2. Hobbs, D. T., Blume, M. S. and Thacker, H. L. (2000). Phase V Simulant Testing of Monosodium Titanate Adsorption Kinetics. WSRC-TR-2000-00142, May 24, 2000.
3. Peretrukin, V. F., Silin, V. I., Kareta, A. V., Gelis, A., V., Shilov, V. P., German, K. E., Firsova, E. V., Maslennikov, A. G. and Trushina, V. E. (1998). Purification of Alkaline Solutions and Wastes from Actinides and Technetium by Co-precipitation with Some Carriers using the Method of Appearing Reagents: Final report, Report PNNL-11988, UC-2030, Pacific Northwest National Laboratory, Sept. 1998.
4. Orth, R. J., Zacher, A. H., Schmidt, A. J., Elmore, M. R., Elliott, K. R., Neuenschwander, G. G. and Gano, S. R. (1995). Removal of Strontium and Transuranics from Hanford Tank Waste via Addition of Metal Cations and Chemical Oxidant - FY 1995 Test Results. Report PNL-10766, UC-721, Pacific Northwest National

Laboratory, September 1995.

5. Hallen, R. T., Bryan, R. T. and Hoopes, F. V. (2000). Development of an Alternative Treatment Scheme for Sr/TRU Removal: Permanganate Treatment of AN-107 Waste. Report PNWD-3047, BNFL-RPT-025 Rev. 0, Pacific Northwest National Laboratory, July 2000.
6. Poirier, M. (2002). Task Technical and Quality Assurance Plan for the Filtration Test using Actual Savannah River Waste Treated with Strontium and Permanganate. WSRC-RP-2001-01020, Rev. 0, Savannah River Site, October 23, 2001
7. Barnes, M. J., Hobbs, D. T., Duff, M. C. and Fink, S. D. (2002). Permanganate Reduction of Savannah River Site Actual Waste Samples for Strontium and Actinide Removal. WSRC-TR-2002-00048, Rev. 0, Savannah River Site, January 17, 2002
8. Nash, C., et. al. (2000). Investigation of Varied Strontium-transuranic Precipitation Chemistries for Crossflow Filtration, BNF-003-98-0171, April 18, 2000.
9. Pike, J. A., Drumm, M. D., Subosits, S. G., Statton, J. L. and Washburn, F. A. (2001). Feed Basis for Processing Relatively Low Radioactivity Waste Tanks. WSRC-TR-2001-00559, Rev. 0.
10. Characterization data for some of the SRS supernate tank wastes indicates that carbonate concentrations may be as high as 0.29 M CO<sub>3</sub><sup>2-</sup> as noted in Pike et al. (2001).
11. Wilmarth, W. R., Nash, C. A., Rosencrance, S. W., DiPrete, D. P. and DiPrete, C. C. (1999). Transuranium Removal from Hanford AN-107 Simulants using Sodium Permanganate and Calcium. Report BNF-98-003-0160, Savannah River Site, September 22, 1999.
12. Krot, N. N., Bessonov, A. A., Gelis, A. V., Shilov, V. P., Perminov, V. P. and Astafurova, L. N. (1998). Coprecipitation of transuranium elements from alkaline solution by the method of appearing agents. I. Coprecipitation of Pu(VI, V) with manganese dioxide. Radiochem. 40, 347-352.
13. Means, J. L., Crerar, D. A., Borcsik, M. P., and Duguid, J. O. (1978). Adsorption of Co and selected actinides by Mn and Fe oxides in soils and sediments. Geochim. Cosmochim. Acta 42, 1763-1773.
14. Duff, M. C., Hunter, D. B., Triay, I. R., Bertsch, P. M., Reed, D. T., Sutton, S. R., Shea-McCarthy, G., Kitten, J., Eng, R., Chipera, S. J. and Vaniman, D. T. (1999). Mineral associations and average oxidation states of sorbed Pu on tuff. Environ. Sci. Technol. 33, 2163-2169.
15. Dyer, A., Pillinger, M., Newton, J., Harjula, R., Moller, T. and Amin, S. (2000). Sorption behavior of radionuclides on crystalline synchrtrin tunnel manganese oxides. Chem. Mater. 12, 3798-3804.
16. Kuznetsov, V. A. and Genralova, V. A. (2000). Sorption of 90Sr and 137Cs on silica and iron, manganese, titanium, and aluminum hydroxides. Radiochem. 42, 166-169.
17. Crespo, M. T., Gascon, J. L. and Acena, M. L. (1993). Techniques and analytical methods in the determination of uranium, thorium, plutonium, americium and radium by adsorption on manganese dioxide. Sci.Tot.Environ. 130, 383-391.
18. El-Naggar, I. M., El-Asby, M. A., Abdel-Hamid, M. M. and Aly, H. F. (1993). Sorption behavior of uranium and thorium on cryptomelane-type hydrous manganese dioxide from aqueous solution. Solv. Extract. Ion Exchange. 11, 521-540.
19. The first mass concentration value was calculated from known fission yields for 88Sr and 90Sr with a decay correction for 90Sr that assumes a waste age of 29 years. If the waste were younger than 29 years, this value would not be bounding. However, we believe it to be a reasonable estimate because there is a likelihood that the waste contains stable Sr from process chemicals and process water. Such additional (stable) Sr would lower the average Ci content of Sr in the waste. The second mass concentration basis is a lower value, which was based on a determination of 90Sr content in a Tank 51H sludge sample. We will refer to these two values throughout the report.
20. We observed similar phenomena when spiking HLW salt simulant solutions that were prepared under basic conditions with Sr and U. After adding our U spike (which followed our Sr spike), we observed a pronounced loss of U from our solutions. This behavior occurred during the initial basic (i.e., high pH) preparation of the salt solutions for this study, which is why we chose to prepare and spike our salt solutions under acidic conditions. By using an acidic preparation approach, we could introduce the Sr and actinides under conditions that favored under-saturation with respect to any potential Sr or actinides phases. [We originally selected a basic solution preparation because this method of preparation is less labor intensive and was thought to yield the same results as an acidic preparation method of the same solution].
21. Meece, D. E. and Benninger, L. K. (1993). The coprecipitation of Pu and other radionuclides with CaCO<sub>3</sub>. Geochim. Cosmochim. Acta. 57, 1447-1458.
22. We observed similar phenomena when spiking HLW salt simulant solutions that were prepared under basic conditions with Sr and U. After adding our U spike (which followed our Sr spike), we observed a pronounced loss of U from our solutions. This behavior occurred during the preparation of the salt solutions for this study, which is why we chose to prepare and spike our salt solutions under acidic conditions.