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Activated carbon,
Hematite, sodium phosphate
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Neptunium, Loading capacity.

**Identification of Tank 50H Solids Components Responsible for Removal
of Dissolved Actinides (U)**

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SUMMARY

As part of preparations to use the Tank 50H as a receiving tank for the salt supernate solution designed to feed the Saltstone Facility, a follow up study has been completed to identify the Tank 50H solids components responsible for the removal of plutonium and neptunium from synthetic salt solution. Identification of these sorbents will also help determine whether the existence of these absorbents is limited to the Tank 50H or more widespread in the High Level Waste system and thus may pose a greater impact.

The primary results of this batch experimental investigation with synthetic salt solution bearing plutonium, uranium and neptunium are:

- The removal of plutonium and neptunium from synthetic salt solution by Tank 50H solids is attributed to the presence of the old ETF granular activated carbon and transition metal oxides, mostly iron and manganese oxides, in Tank 50H solids. These components of the Tank 50H solids are known active sorbents of both plutonium and neptunium.
- Both activated carbon and iron oxide (hematite), which are components of Tank 50H solids, contribute to the removal of Pu and Np by Tank 50H solids and in combination, they account for about 30% of the Pu and Np removed by Tank 50H solids.
- Nitrated cancrinite, potassium tetraphenylborate and scintillation cocktail in contact with simulant salt solution containing uranium, plutonium and neptunium do not show any measurable tendency to remove or precipitate these actinides from salt solution.
- The two forms of activated carbon investigated in these tests show measurable affinity for plutonium and neptunium.
- The new ETF activated carbon did not precipitate or remove any measurable amount of uranium from the simulant salt solution, while the old ETF granular activated carbon removed less than 10% of the uranium.
- The new ETF activated carbon exhibits lower affinity for plutonium and neptunium than the old ETF granular activated carbon.
- The old ETF granular activated carbon and iron oxide (hematite) exhibit high affinity for both plutonium and neptunium.
- Hematite exhibits greater affinity for plutonium and neptunium than either of the ETF activated carbons.

- Plutonium and neptunium loading capacities onto “neat” old ETF granular activated carbon are, respectively, 3.6 ± 0.22 and 7.9 ± 0.52 μg per gram of old ETF granulated activated carbon.
- Plutonium and neptunium loading capacity onto “neat” new ETF activated carbon are, respectively, 1.30 ± 0.07 and 4.4 ± 0.44 μg per gram of new ETF activated carbon.
- The loading capacity for plutonium and neptunium onto hematite are, respectively, 5.5 ± 0.38 and > 10 μg per gram of hematite.
- Plutonium loading capacity onto the Tank 50H solids (2.01 μg per gram of Tank 50H solids) is about 60% of plutonium loading capacity onto “neat” old ETF granular activated carbon and 40% of plutonium loading capacity onto “neat” hematite.
- Neptunium loading capacity onto the Tank 50H solids (4.48 μg per gram of Tank 50H solids) is about 50% of plutonium loading capacity onto “neat” ETF granular activated carbon and less than 50% of neptunium loading capacity onto “neat” hematite (iron oxide).

Additional testing to verify if other transition metal oxides (MnO_2 and ZnO) are responsible for actinide removal not accounted for by the presence of activated carbon and hematite are recommended.

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1.0 INTRODUCTION

Tank 50H is scheduled to return to HLW Tank Farm service with the capability to receive salt supernate solutions for feed to the Saltstone Facility. However, the presence of two large mounds (“North” and “South” mounds) of solid material¹ in the tank presents a number of issues which have to be dealt with before Tank 50H can be returned to HLW service². Characterizations of the Tank 50H solids^{3,4} indicated the presence of a significant fraction of sodium oxalate, scintillation cocktails, carbon black, cancrinite, sodium phosphate, tetraphenylborate and other yet unidentified materials in the north and south mound solids.

The final fate of decontaminated liquid waste at the Savannah River Site is to grout the liquid supernate for disposal in the Saltstone Facility. Here, the Tank 50H would serve as a staging tank and decontaminated supernate would be routinely transferred from Tank 50H to the Saltstone feed tank for processing. One option under consideration allows keeping the solids in Tank 50H and bringing low-curie salt solution, as planned, into the tank before sending on to Saltstone. An issue with this option is what effect, if any, the oxalate-rich and yet uncharacterized solids in the tank will have on the dissolved uranium and other actinides in the incoming salt solutions. The uncharacterized solids may dissolve and bring about the precipitation and accumulation of actinides such as uranium and plutonium. Since SRS wastes frequently contain enriched uranium and weapons-grade plutonium, accumulation of the uranium and plutonium in a solid phase is a possible criticality safety concern.

Current activities under way to prepare the Tank 50H for service include dissolving the Tank 50H solids with “inhibited” water (0.01 M NaOH) and transferring to the Saltstone Facility for disposal. In these slurry processing activities an attempt will be made to remove all of the solid mounds present in Tank 50H.

In a recent study⁴ to examine the effects of contacting a simulated salt solution spiked with uranium, plutonium and neptunium with Tank 50H solids, it was observed that both plutonium and neptunium were selectively removed from the salt solution in the presence of Tank 50H solids.

In this follow up study an attempt has been made to identify the Tank 50H solids components responsible for removing the actinides mentioned above. Identification of the plutonium-and neptunium-sorbing materials will help to assess whether the material is limited to Tank 50H or occurs widely in the HLW system.

Activated carbon and iron oxides are two of the many materials which have been identified as components of the Tank 50H solids.³ Based on plant history, the presence of activated carbon in the Tank Farm Waste storage system may be limited only to Tank 50H, although other forms of carbon are certainly present in various storage tanks. Activated carbons, it is known, may have been introduced into the Tank 50H as a result of back flushing activities at the ETF carbon beds. In this process, the carbon fines end up as evaporator bottoms which are eventually transferred to the Tank 50H. Iron, on the other hand, exists in various oxidation forms, mostly in the Fe^{3+} and Fe^{2+} oxidation states. The usually amorphous and crystalline (mostly hematite and some magnetite) phases of iron minerals present in the tanks are known neutron “poisons”.

This report responds to Technical Task Request HLE-TTR-2003-068, Rev.1, which includes a task to identify the Tank 50H solids component materials responsible for plutonium and neptunium removal from simulated Tank 50H supernate salt solution. Three types of laboratory tests were performed: (1) separation of Tank 50H solids into insoluble and soluble fractions and testing each fraction for actinide removal efficiency, (2) single component tests with selected Tank 50H components and (3) tests to determine loading curves and the loading capacities for plutonium and neptunium using “neat” commercial materials of the identified actinide sorbents. The word “neat” as used here refers to the fact that the absorbing or precipitating solids which are some of the components of the Tank 50H solids, for example old Effluent Treatment Facility granular activated carbon (old ETF GAC), the new ETF millimeter-size activated carbon rods currently in use at the ETF or hematite (iron oxide), were used as-received from the various commercial sources.

2.0 EXPERIMENTAL

One Tank 50H core sample tube, labeled as HTF-E-03-17, was provided by the plant for this study. A photograph of the sampler and its contents is shown in Figure 1. Based on the orientation of the sample tube, three distinct sample sections with different water content and fluidity (labeled B, C, and D in Figure 1) were observed in the Tank 50H material used in this study. These three sections were blended together in a large polyethylene bottle and air dried in a hood for several hours until a fairly constant weight material was obtained. The resulting air-dried Tank 50H solid sample was used in this study (see Figure 2). The Tank 50H solids components evaluated for actinide (plutonium, neptunium and uranium) removal in the synthetic salt solution included the two forms of the ETF activated carbons, nitrated cancrinite, iron oxide (hematite), potassium tetrphenylborate, liquid scintillation cocktail and the Tank 50H solid and liquid fractions. Sodium phosphate, which is not a component of the Tank 50H solids, was evaluated only as a reference material for uranium removal from the salt solution.

The two forms of activated carbon, described above, were provided by Effluent Treatment Facility (ETF) personnel. The old ETF granular activated carbon (old ETF GAC) with a commercial brand identification name “C-200 GAC” was manufactured by Duratec International and the new ETF activated carbon currently used at the ETF, with a brand name “Nusorb GC-60-15 carbon”, is manufactured by Nucon International, Inc. The iron oxide (hematite) was a Fisher Scientific company commercial product and the nitrated cancrinite was provided by Dr. J. A. Mensah from the University of South Australia. The potassium tetrphenylborate was precipitated from a solution of sodium tetrphenylborate (Alfa Aesar) by addition of potassium nitrate. The resulting solid precipitate was washed and vacuum-dried to constant weight over several days at room temperature. The liquid organic scintillation cocktail (Ultima Gold AB) consisting of linear alkylbenzenes, phenylxylylene, and non-ionic surfactants and other emulsifiers is manufactured by ICN Biomedicals, Inc. of Costa Mesa, California. The anhydrous sodium orthophosphate used in this study was derived from hydrated sodium orthophosphate ($\text{Na}_3\text{PO}_4 \cdot 12\text{H}_2\text{O}$) by heating of the hydrated salt in stainless steel beaker

to above its boiling point for several hours. The resulting solid was then transferred to an oven and maintained at 120 °C for an hour before bringing it to room temperature in a desiccator.

Table 2 summarizes the different Tank 50H solids components³ evaluated for plutonium, uranium and neptunium removal. The x-marks on the table indicate where the presence of a particular component caused the precipitation or loss of that actinide (>10% loss) in the simulant solution bearing uranium, plutonium and neptunium.

The composition of the 5.6 molar sodium salt solution (spiked with near saturation levels of uranium and plutonium) is presented in Table 1. This salt solution is comparable to that expected to pass through the Tank 50H in the near future in support of the low-curie salt program and over the longer term from the Salt Waste Processing Facility. The uranium, plutonium and neptunium concentrations in the simulant used for these studies were stable after preparation. The target concentrations for these actinides in the simulant were, respectively, 10.0, 0.20 and 0.50 mg/L with a 10 % margin of error. Single, pre-test analytical results for uranium, plutonium and neptunium concentrations in the simulant were 10.7, 0.192 and 0.496 mg/L, respectively.

In general, in the single component tests, 5 grams of each Tank 50H solids component were contacted with the simulant solution. The conservative liquid-to-solid ratio (by weight) of 5 used for these tests was based on original estimates of solids in Tank 50H (61,000 gallons) and approximately 300,000 gallons of solution derived from the dissolution of 100,000 gallons of saltcake from Tank 41H. Other liquid-to-solid ratios, usually higher than 5, were used in tests where we had problems performing liquid-solid separations or where the component showed extensive precipitation of the actinides from solution. The tests were performed in 150-mL capped polypropylene bottles. Agitation of the Tank 50 H solids component (neat materials, for example, cancrinite or activated carbon) and simulant was achieved by placing the capped polypropylene bottles in an orbital shaker (Lab-line model C-760) maintained at 175 revolutions per minute. Aliquots were taken from each test bottle every 24 hours for up to four days and analyzed for plutonium and neptunium. After contact, samples were filtered through a 0.45-micron nylon syringe filter disc to remove undissolved solids. The filtrate was acidified with 5.0 M nitric acid and the actinide concentrations determined by inductively coupled plasma mass spectrometry (ICP-MS). All tests were performed at 26 ± 2 °C, which was the bath water temperature in the orbital shaker. Tests with no added solids (simulant solutions only) were featured in each test set as controls for preparation errors, instrumental biases and unexpected removal mechanisms such as sorption with polypropylene bottle walls. The controls (untreated samples) were also subjected to the same separation and acid dilution techniques described above.

2.10 Extraction of Tank 50H Solids

In tests on Tank 50H solid and liquid fraction interactions with actinides, a sample of air-dried Tank 50H solid (50.14 g) was dissolved in 500 mL of Tank Farm inhibited water. The Tank Farm inhibited water had already been filtered through a 0.45 micron filter prior to use. The resulting slurry of filtered Tank Farm inhibited water and Tank 50H solids was agitated in an orbital shaker set at 175 rpm overnight. After the agitation

of the slurry for 24 hours, a 0.45 micron-filter retainer was used to separate the liquid from the solid fraction. The solid fraction was air dried to constant weight. This leaching of Tank 50H solids was repeated and the average weight of dry solids recovered and dried to constant weight was 25.03 ± 0.04 grams. The weight of the soluble fraction was then determined by difference ($50.14 - 25.03$ g). The solid fraction was analyzed for actinide and total carbon. A 10 ml portion of the Tank 50H liquid fraction (leachate) was mixed with an equal volume of the simulant solution bearing actinides (10 ± 1 mg/L uranium, 0.20 ± 0.02 mg/L plutonium and 0.50 ± 0.05 mg/L neptunium). This sample was prepared in duplicate and the mixture agitated in an orbital shaker for 24 hours. A sample was pulled from the mixture after the 24 hours and analyzed for actinide concentrations. This test was performed with the Tank 50H liquid fraction to determine if the components of Tank 50H solid responsible for actinide removal were in the liquid/soluble phase of Tank 50H solids.

2.2 Plutonium and Neptunium Loading on Sorbents

In the loading curve tests each “neat” solid material under study was mixed with a specific volume of simulant solution at different liquid-to-solid ratios (generally, 14:1, 20:1, 24:1, 30:1, 40:1, 50:1 and 60:1). Seven liquid-to-solid ratio (volume/weight) samples were prepared with the weight of the neat solid test material maintained constant at either 0.25 or 0.5 grams. The eighth sample in each batch of tests was the plain simulant solution used as the control and contained no solids. A twenty-four hour shaking of the mixture in an orbital shaker at room temperature (26 ± 2 °C) was judged sufficient, based on previous experience⁴, for attainment of sorption equilibrium. After 24 hours of shaking, the liquid phase was separated from the solid phase using a 0.45-micron nylon filter. A portion of the liquid fraction was diluted with 5.0 M nitric acid and analyzed for plutonium and neptunium. The control sample was processed similarly including filtration and acid dilution. Apart from runs involving sodium phosphate, the sample solutions were not analyzed for uranium since previous tests showed that none of the “neat” materials were found to remove significant amounts of uranium.

Table 1. Synthetic salt solution composition

| Component | Target concentration | Units |
|---------------------------------|----------------------|-------|
| Total Na | 5.60 | M |
| Free NaOH | 1.33 E+00 | M |
| NaNO ₃ | 2.60E+00 | M |
| NaNO ₂ | 1.64E-01 | M |
| Na ₂ SO ₄ | 5.21E-01 | M |
| Na ₂ CO ₃ | 2.60E-02 | M |
| NaAl(OH) ₄ | 4.29E-01 | M |
| Total U | 10.0 ±1 | mg/L |
| Total Pu | 0.20 ± 0.02 | mg/L |
| Np-237 | 0.50 ± 0.050 | mg/L |



A: Tank 50H sampling tube and contents



B: Tank 50H solids sample from top of sampling tube



C: Tank 50H solids sample from middle of sampling tube



D: Tank 50H solids sample from bottom of sampling tube.

Figure 1 As-received Tank 50H solids.

| | |
|---|--|
|  |  |
| <p>Blend of samples A, B and C: After 72-hour hood exposure</p> | <p>Blend of samples A, B and C: After 264- hours hood exposure</p> |
|  | <p>BLANK</p> |
| <p>Current Tank 50H solids (Left) and previous Tank 50 H solids (Right). Differences in color may be due to differences in aging.</p> | |

Figure 2 Air-dried Tank 50H solids

Table 2 Tank 50H components evaluated for removal of actinides

| Component | Uranium | Plutonium | Neptunium | Comments |
|---|---------|-----------|-----------|-------------------------------------|
| Nitrated Cancrinite | | | | |
| Activated Carbon | | X | X | |
| Iron Oxide (hematite) | | X | X | |
| Sodium phosphate | X | | X | Reference; not a Tank 50H component |
| Potassium tetra phenyl borate | | | | |
| Sodium oxalate | | | | From previous study see reference 4 |
| Scintillation Cocktail | | | | |
| Control (no added component) | | | | |
| Tank 50H solids | | X | X | |
| Tank 50H solid fraction derived by leaching with Plant inhibited water | | X | X | |
| Tank 50H liquid fraction derived by leaching with Plant inhibited water | | | | |

Here X indicates precipitation or loss of that actinide in the presence of that Tank 50H component.

3.0 RESULTS AND DISCUSSION

3.1 Data Presentation

Graphically, the experimental results are presented here in three ways: (1) Changes in actinide concentrations with contact or exposure time in the presence of sorbents, (2) actinide concentration changes at different liquid-to-solid ratios, and (3) loading curves for actinides and strontium in the presence of the precipitating agent (“neat” sorbent) at different liquid-to-solid ratios.

3.2 Physical Properties of Tank 50H Solids

The air-dried Tank 50H solid sample produced the x-ray diffraction pattern shown in Figure 3. The pattern exhibits sharp diffraction peaks and a smooth baseline indicative of a very crystalline material. Three crystalline materials were identified in the air-dried Tank 50H sample: sodium oxalate ($\text{Na}_2\text{C}_2\text{O}_4$), strontium oxalate, and calcium oxalate hydrate ($(\text{COO})_2\text{Ca}\cdot\text{H}_2\text{O}$). An unassigned peak at ($2\Phi = 140$) suggests the presence of additional unidentified crystalline material. The x-ray pattern obtained for the current Tank 50H solids is in fairly good agreement with other similar Tank 50H solids³ (Question marks,?, on XRD spectra indicate unmatched or unidentified peaks).

Aqua regia digestion of air-dried Tank 50H solid was followed by analysis for uranium using inductively-coupled plasma atomic emission spectrometer (ICP-ES). ICP-ES analysis for uranium gave a uranium concentration of 0.217 ± 0.011 mg uranium per gram of Tank 50H solid. Previous characterization of Tank 50H solids gave an average value of 0.365 ± 0.030 mg uranium per gram of Tank 50H solids⁴. Total carbon analysis for air-dried Tank 50H solids gave a value of 135 mg carbon per gram of air-dried Tank 50H solids (13.5 % carbon). This value is in agreement with values reported earlier on Tank 50H solids^{3,4}.

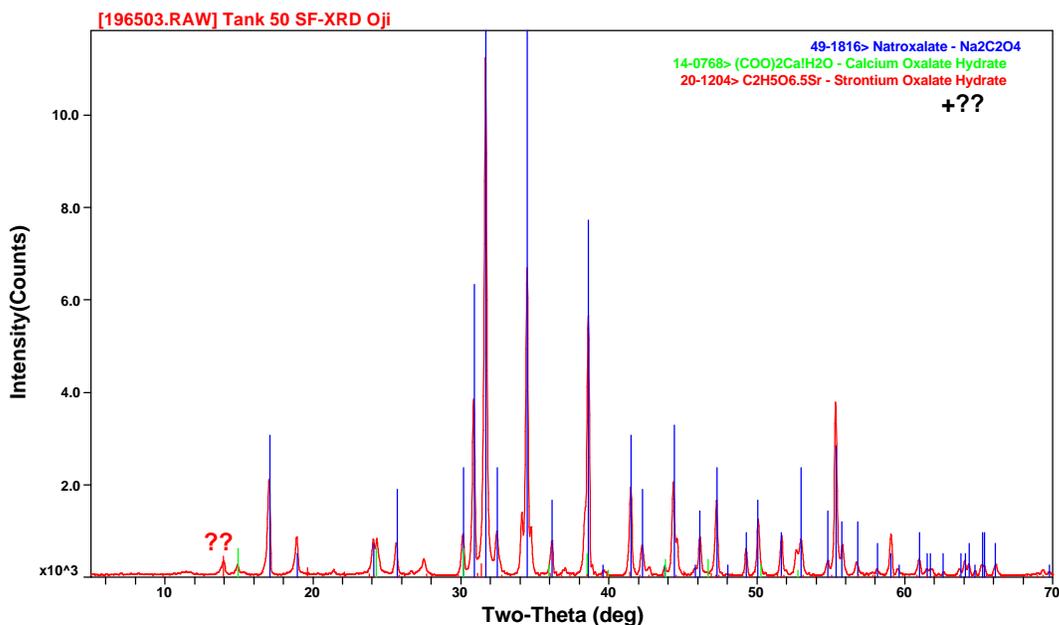


Figure 3. X-Ray Diffraction Powder Pattern of Air-dried Tank 50H Solids .

3.3 Investigation of Tank 50H Solids Interactions with Actinides

The air-dried Tank 50H solids were contacted with salt solution containing uranium, plutonium and neptunium. Plots in Figure 4 show actinide concentrations after 24-hour contact for a range of liquid-to-solid ratios. The selection of 24-hour contact time was based on previous study with Tank 50H solids (reference 4), which showed that this was sufficient contact time for the sorption of actinides onto Tank 50H solids in the stimulant solution.

No significant changes in uranium concentration occurred over the 24-hour contact time when air-dried Tank 50H solids were contacted with the synthetic salt solution. Plutonium and neptunium were removed from the simulant solution in the presence of the

air-dried Tank 50H solids. These results are consistent with earlier study with Tank 50H solids as presented in reference 4.

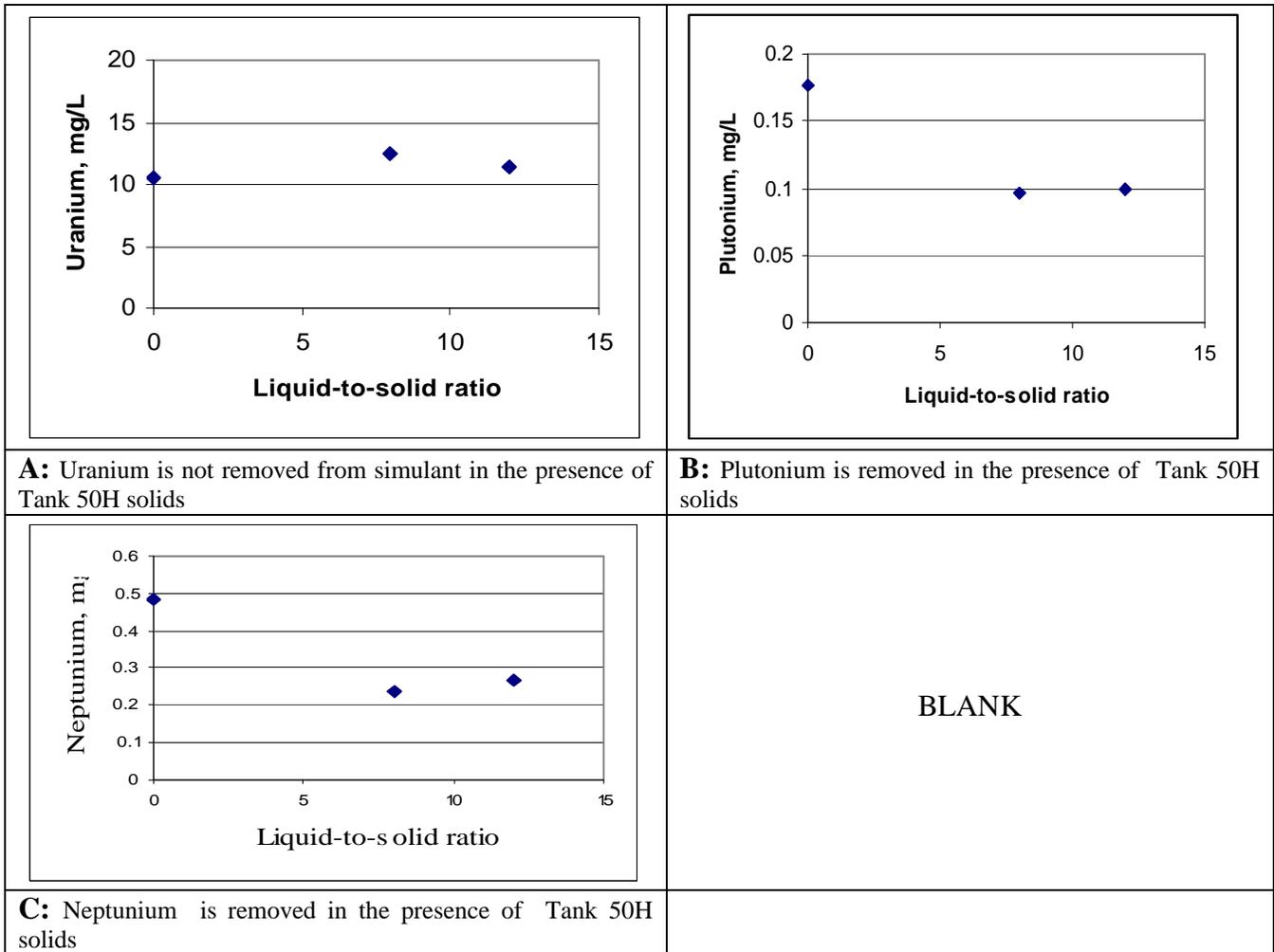


Figure 4. Effects of liquid-to-solid ratio on simulant solution actinide concentration in contact with Tank 50H solids. Plots at liquid-to-solid ratio of zero represent concentration of the actinides before contact.

3.4 Tank 50H Solid and Liquid Fraction Interactions with Actinides

As described earlier, air-dried Tank 50H solids were leached with plant inhibited water, yielding an undissolved solid fraction and a liquid fraction. The solid fraction was air-dried. Analysis of the initial plant inhibited water showed that no actinides or any other radionuclides were present. An equal volume of this Tank 50H liquid fraction and the simulant solution bearing actinides were mixed, agitated and sampled for actinide analysis after 24 hours. Analysis of the filtrate, as summarized in Table 3, shows that there was more uranium, plutonium and neptunium in the Tank 50H soluble fraction than in the original simulant. The uranium, plutonium and neptunium concentrations in the mixture averaged 53.4 ± 12.3 , 0.70 ± 0.18 and 1.85 ± 0.5 mg/L, respectively. Since there

were no radionuclides detected in the plant inhibited water, it can be concluded that the increase in actinide concentrations was due to the presence of Tank 50H soluble fraction.

Based on calculations shown below the actual uranium, plutonium and neptunium contributions from the Tank 50H liquid fraction were, respectively, 97, 1.2 and 3.2 mg/L.

{10 ml of simulant solution + 10 ml of Tank 50H liquid fraction
U, Pu and Np concentration in simulant are, respectively, in 10, 0.2 and 0.5 mg/L

For Np

$$(0.01L)(0.5 \text{ mg/L}) + (0.01L) * X = (1.85 \text{ mg/L}) * 0.02L$$

$$0.01L * X = 0.037\text{mg} - 0.005 \text{ mg}$$

$$X = 3.2 \text{ mg/L}$$

For Pu

$$(0.01L)(0.2 \text{ mg/L}) + (0.01L) * X = (0.7\text{mg/L}) * 0.02L$$

$$0.01L * X = 0.014\text{mg} - 0.002\text{mg}$$

$$X = 1.2 \text{ mg/L}$$

For U

$$(0.01L)(10 \text{ mg/L}) + (0.01L) * X = (53.42\text{mg/L}) * 0.02L$$

$$0.01L * X = 1.068\text{mg} - 0.1\text{mg}$$

$$X = 96.84 \text{ mg/L}$$

Without further characterization and comparing of the Tank 50H liquid fraction and the original air-dried Tank 50H solids, it is not possible to conclude there was a measurable loss of the actinides from the mixture as a result of blending the simulant solution with the Tank 50H liquid fraction. Hence, although the test results so far performed with the Tank 50H solids materials will seem to suggest that the components of the Tank 50H solids responsible for the removal of actinides from solution are in the solid fraction of the Tank 50H solids, one can not say with great certainty that some soluble components like organic complexes in the liquid fraction do not contribute to the removal of actinides.

Aqua regia digestion of the Tank 50H solid fraction in triplicate followed by ICP-ES analysis for uranium gave a uranium concentration of 0.33 ± 0.06 mg uranium per gram of Tank 50H solid fraction. Total carbon analysis for Tank 50H solid fraction gave a value of 144 mg carbon per gram of Tank 50H solids (14.4 % carbon). These values for the uranium and carbon content are consistent with previous characterizations of the Tank 50H solids where these values were, respectively, reported as 0.37 ± 0.03 and 13.5 %.^{3,4}

Table 3 Actinide concentration for Tank 50H liquid fraction

| | U, mg/L | Pu, mg/L | Np, mg/L |
|--|-----------------|-----------------|-----------------|
| Concentration in simulant | 10.0 ± 1.0 | 0.2 ± 0.02 | 0.50 ± 0.05 |
| Measured concentration in the mixture after contact | 53.4 ± 12.3 | 0.70 ± 0.18 | 1.85 ± 0.46 |
| Calculated concentration in Tank 50H liquid fraction | 97 | 1.2 | 3.2 |

Figure 5, inserts A through F, shows the change in actinide concentrations with time for simulant solutions contacted with the Tank 50H solid fraction (undissolved solids from leaching with plant inhibited water). The tests were performed at two simulant-to-solid ratios of 25:1 and 8:1. In Figure 5, the changes in actinide concentration with time are presented in inserts A, C and E and the changes in actinide concentration with variation in liquid-to-solid ratios are presented in Figure 5 inserts B, D and F.

There was no measurable difference in uranium concentrations before and after contacting of the simulant with Tank 50H solid fraction after for more than 72 hours (Figure 5 insert A). The uranium concentration in the post-contact Tank 50H solid fraction averaged 10.6 ± 0.6 mg/L and is not statistically different from the original uranium concentration in the simulant before contact (10.6 ± 1.0 mg/L).

Removal of uranium, even at lower liquid-to-solid ratio was not observed (Figure 5 insert B). Only plutonium and neptunium removed from solution in the presence of Tank 50H solid fraction (Figure 5 inserts C, D, E and F). After 72 hours contact, the soluble plutonium concentration in the mixture had dropped to below instrument detection level (Figure 5 insert C) and the neptunium concentration had dropped to less than 50% of its original concentration in the simulant (Figure 5 insert E). The decrease in plutonium and neptunium concentration with decrease in liquid to solid ratios as shown in Figure 5 inserts D and F, respectively, confirms the existence of a typical sorption phenomenon in the mixture. These observations are similar to the results obtained when the simulant solution was exposed to unleached Tank 50H solids (Figure 4), again confirming that the components responsible for the removal of actinides are in the Tank 50H solid fraction.

3.5 Single Component Tests for Tank 50H Solids

As already described and detailed in Table 2, the identified components of Tank 50H solids are nitrated cancrinite, activated carbon derived from ETF processes, scintillation cocktail, sodium oxalate and potassium tetraphenylborate. The changes in color of the wet Tank 50H solids from bright yellow paste to reddish-brown granules for the dry Tank 50H solids (Figures 1 and 2), suggests that Tank 50H solids contain a chromophore which causes such color changes with exposure to air following the loss of moisture. Based on plant history and chemistry of plant solids like the sludge, one of the possible chromophore candidates for the Tank 50H solids is iron which can exist in several oxidation states. The changes in color of the Tank 50H solids with exposure to air could be attributed to the existence of iron oxides, possibly hematite and magnetite (Fe_3O_4). As a result, it was decided to include hematite (ferric oxide- Fe_2O_3) as one of the test components in this evaluation of components responsible for precipitation of plutonium and neptunium in simulant salt solution. Anhydrous sodium phosphate was also included in the test matrix because it is known to remove uranium.

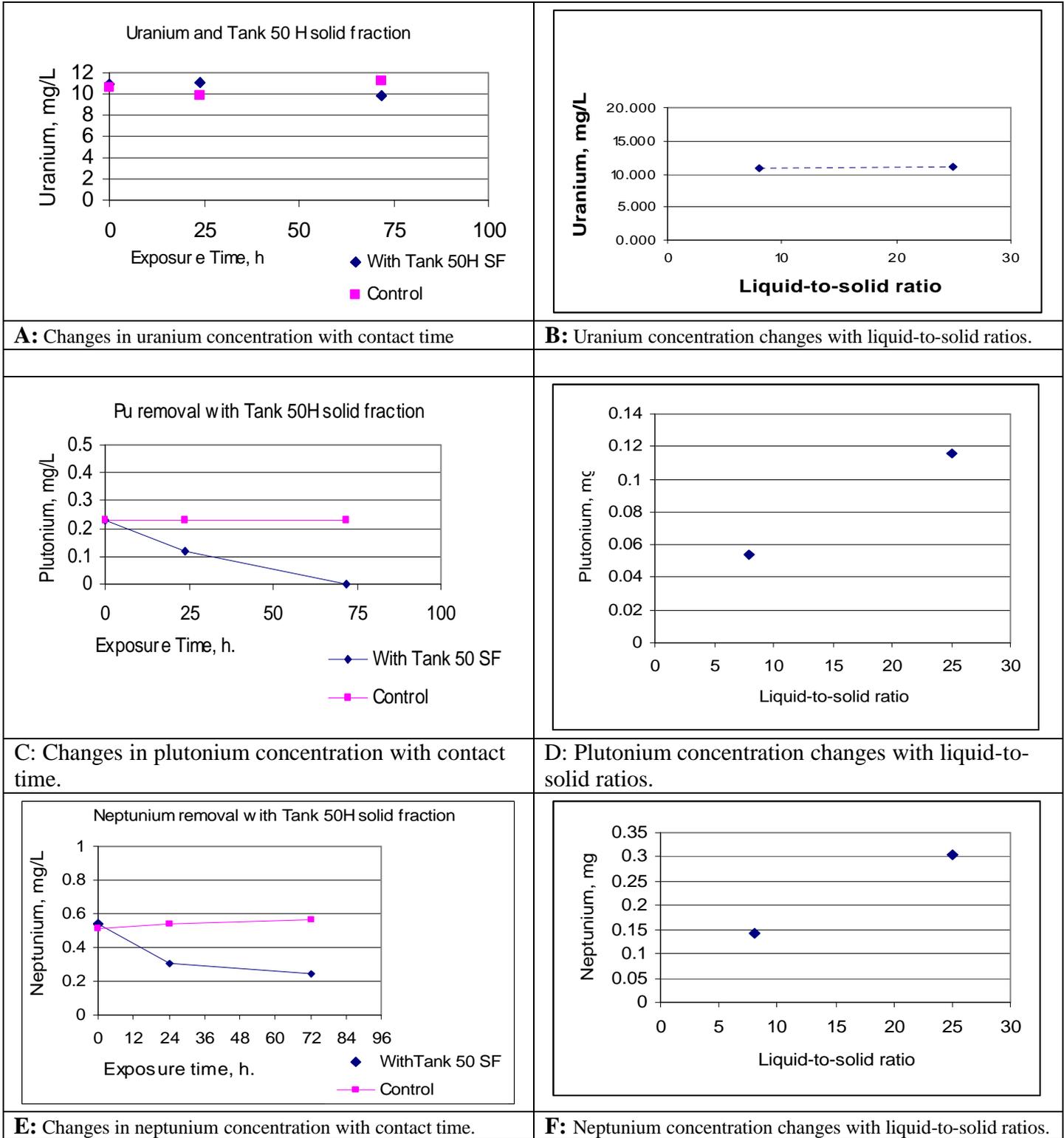


Figure 5. Actinide removal from contact with Tank 50H solid fraction.

3.6 Nitrated Cancrinite and Activated Carbon Interactions with the Actinides

The data presented here for nitrated cancrinite and the activated carbons are based on the 24-, 48- and 72-hour samples for cancrinite and new ETF activated carbons and 24- and 48-hour samples for the old ETF GAC.

Since cancrinite, an aluminosilicate, is a significant component of the Tank 50H solids, the cancrinite tests were run several times at different liquid-to-solid ratios to ensure that there was no significant removal of actinides within the first 24 hours of contact with simulant solution. The tests with nitrated cancrinite in the 5:1 ratio posed a special problem when separating the liquid from the solid fraction at the end of 24 hours. At this ratio, the resulting mixture formed a thick slurry after mixing and could not be filtered through 0.45 micron filtration discs. A staged filtration technique involving initial filtration with 1.0 micron filter disc followed by 0.45 micron discs was used to overcome this problem. In this initial 24-hour cancrinite test, about 85% of the neptunium and 78% of the plutonium was recovered. Over 95% of the uranium was recovered. The 24-hour 5:1 simulant-to-cancrinite ratio test was repeated three times using staged filtration. Over 95% of the initial neptunium, plutonium and uranium concentrations were recovered in each repeat test. Figure 6 includes the results of the 24-hour repeat tests, along with additional data at other time increments. The additional cancrinite tests used higher simulant-to-cancrinite ratios (25:1). At the higher ratio, filtration was easier and more than 95% of each of the actinides in all cases was recovered. Since the repeat tests and higher ratio tests did not result in any measurable loss of the actinides, we conclude that the apparent loss of neptunium and plutonium in the initial phase of the cancrinite tests was likely due to sampling or analytical errors.

The average uranium, plutonium and neptunium recovered in the control tests were 10.4 ± 0.5 , 0.20 ± 0.03 and 0.51 ± 0.04 mg/L, respectively. The average uranium, plutonium and neptunium recovered in the cancrinite in contact with simulant tests, at 5:1 ratio, were 10.17 ± 0.29 , 0.20 ± 0.01 and 0.51 ± 0.01 , respectively. Figures 6 and 7 summarize the post-contact results for nitrated cancrinite in the actinide-bearing simulant. Since the average actinide recovered after contact is not statistically different from the actinide concentrations in the controls, we conclude that nitrated cancrinite does not actively precipitate uranium, plutonium or neptunium from the synthetic salt solution (Figures 6 and 7).

Two forms of ETF activated carbon were used in these tests. Neither form affected uranium concentration; both forms removed neptunium and plutonium from the simulant salt solution (Figures 8 and 9). At the end of the first 24-hour sampling period, the new ETF activated carbon removed about 72% of the neptunium and more than 95% of the plutonium. Similarly, at the end of 24 hours, the old ETF GAC removed neptunium and plutonium in the simulant below instrument detection level (more than 95%). When the amount of uranium recovered from contacting the new ETF activated carbon with simulant solution is compared (2 sigma) with the uranium in the controls (10.5 ± 0.3), there are no measurable losses of uranium due to the presence of the two types of activated carbon (Figures 8 and 9, inserts A). The average uranium recovered in the new

ETF activated carbon in contact with simulant is 10.4 ± 0.4 and for the old ETF GAC in contact with simulant is 9.62 ± 0.08 . The old ETF GAC removed less than 10% of the uranium in the salt solution.

Based on these test results, it is concluded that nitrated cancrinite in contact with simulant solution containing uranium, plutonium and neptunium does not show any measurable tendency to remove these actinides from solution. On the other hand, the two forms of activated carbon investigated in these tests show measurable affinity to remove plutonium and neptunium from solution. The new ETF activated carbon did not remove any measurable amount of uranium from the simulant solution, while the old ETF GAC removed less than 10% of the uranium. From plant history and process knowledge, the activated carbon in the Tank 50H solids used in this study is the old ETF carbon and not the new ETF activated carbon materials used by ETF.

3.7 Iron Oxide and Sodium Phosphate Interactions with Actinides

Figure 10, inserts A through F and Figure 11 shows the graphical presentation of the results obtained with the exposure of simulant solutions to “neat” hematite for up to 48 hours. In Figure 10, the changes in actinide concentration with time for these actinides are presented in inserts A, C and E and the changes in concentration with variation in liquid-to-solid ratios are presented in Figure 10 inserts B, D and F. The changes in actinide concentration with exposure time were based on a liquid-to-solid (hematite) ratio of 50:1, while changes in actinide concentration with liquid-to-solid (hematite) ratio were based on three ratios: 5:1, 10:1 and 50:1.

Statistically, there were no measurable differences in uranium concentration when the simulant was contacted with hematite for 48 hours (Figure 10 inserts A and B). The uranium recovered from the interaction of the simulant solution with hematite averaged 10 ± 0.4 mg/L compared to an average value of 10.4 ± 0.3 mg/L for the untreated simulant. Precipitation of uranium, even at lower liquid-to-solid ratio (more hematite in contact with simulant), was not observed (Figure 10 insert B).

Plutonium and neptunium concentrations in solution decreased in the presence of hematite (Figure 10 inserts C, D, E and F). Within 24 hours of contacting hematite with the plutonium bearing simulant solution, the plutonium concentration dropped to an equilibrium level of about 0.07 mg/L compared to the initial concentration of 0.2 mg/L. This was more than a 60% drop in plutonium concentration after contact with hematite (Figure 10 insert C). Similarly, the neptunium concentration in the hematite-contacted simulant dropped to less than 50% of its original concentration within 48 hours of contact (Figure 10 insert E). Figure 10, inserts D and F shows the effect of changing the simulant-to-solid ratio. In both overlay plots, as the simulant-to-solid ratios decrease (increase in hematite solid fraction) the amount of plutonium and neptunium removed from the liquid phase increased. This behavior is consistent with typical sorption suggesting that plutonium and neptunium are sorbing onto the hematite (iron oxides).^{5,6}

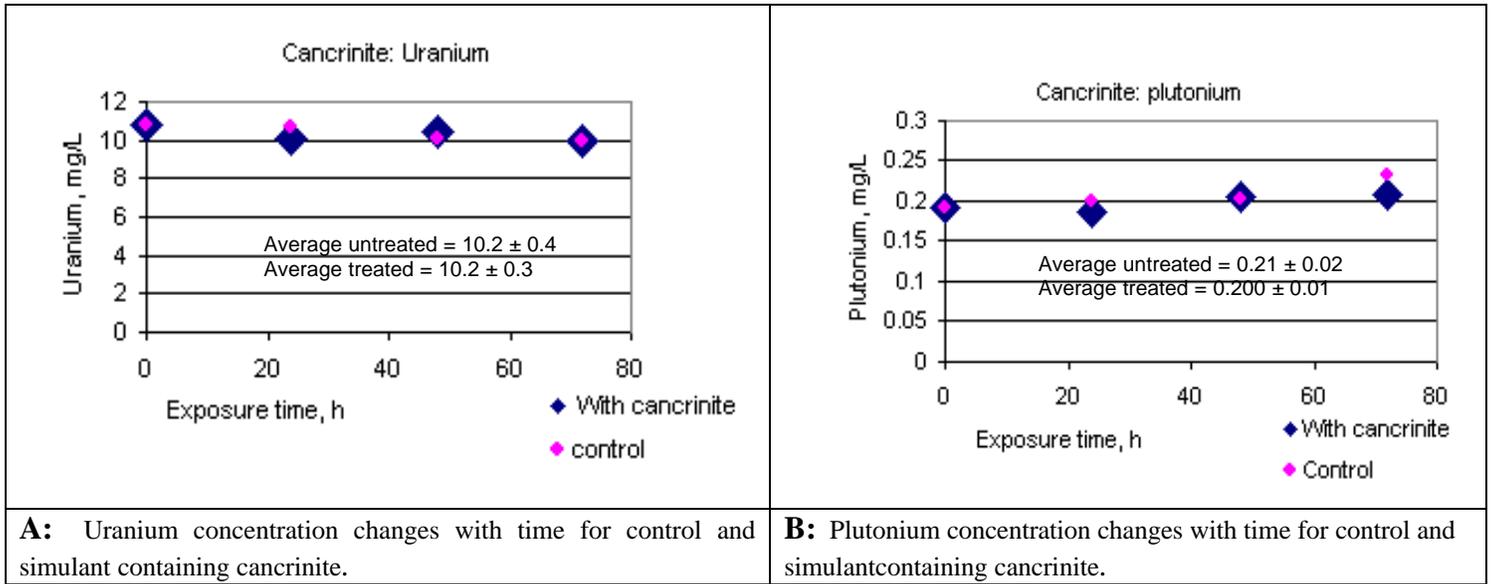


Figure 6 Changes in actinide concentration with time for nitrated cancrinite contacted with synthetic salt solution.

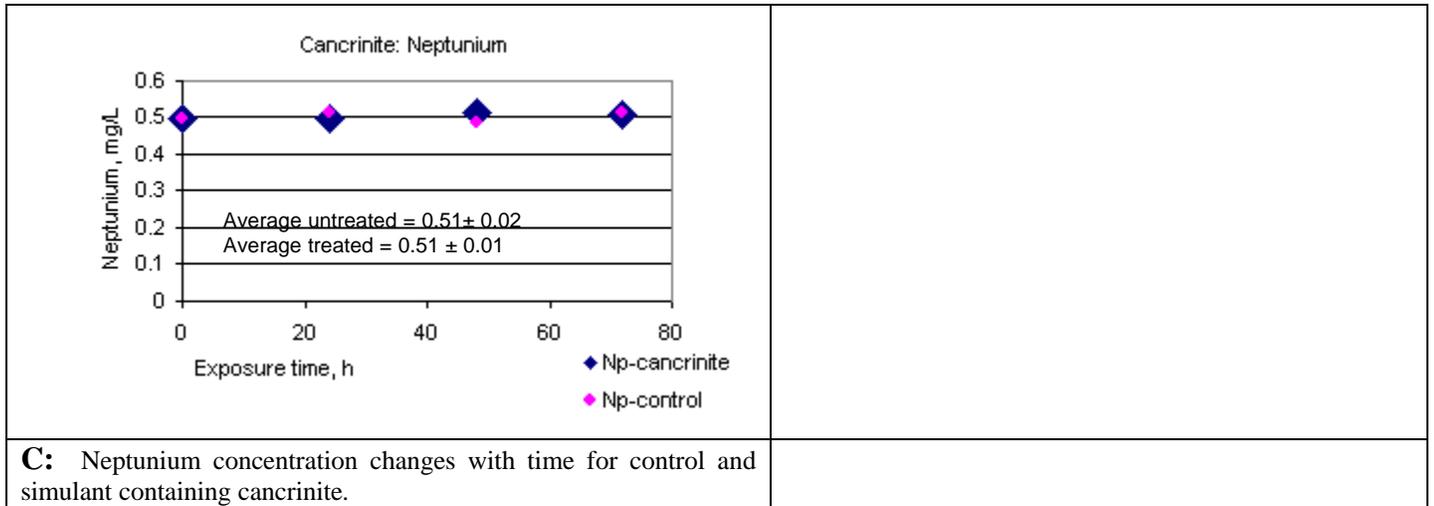


Figure 7 Changes in actinide concentration with time for nitrated cancrinite contacted with synthetic salt solution.

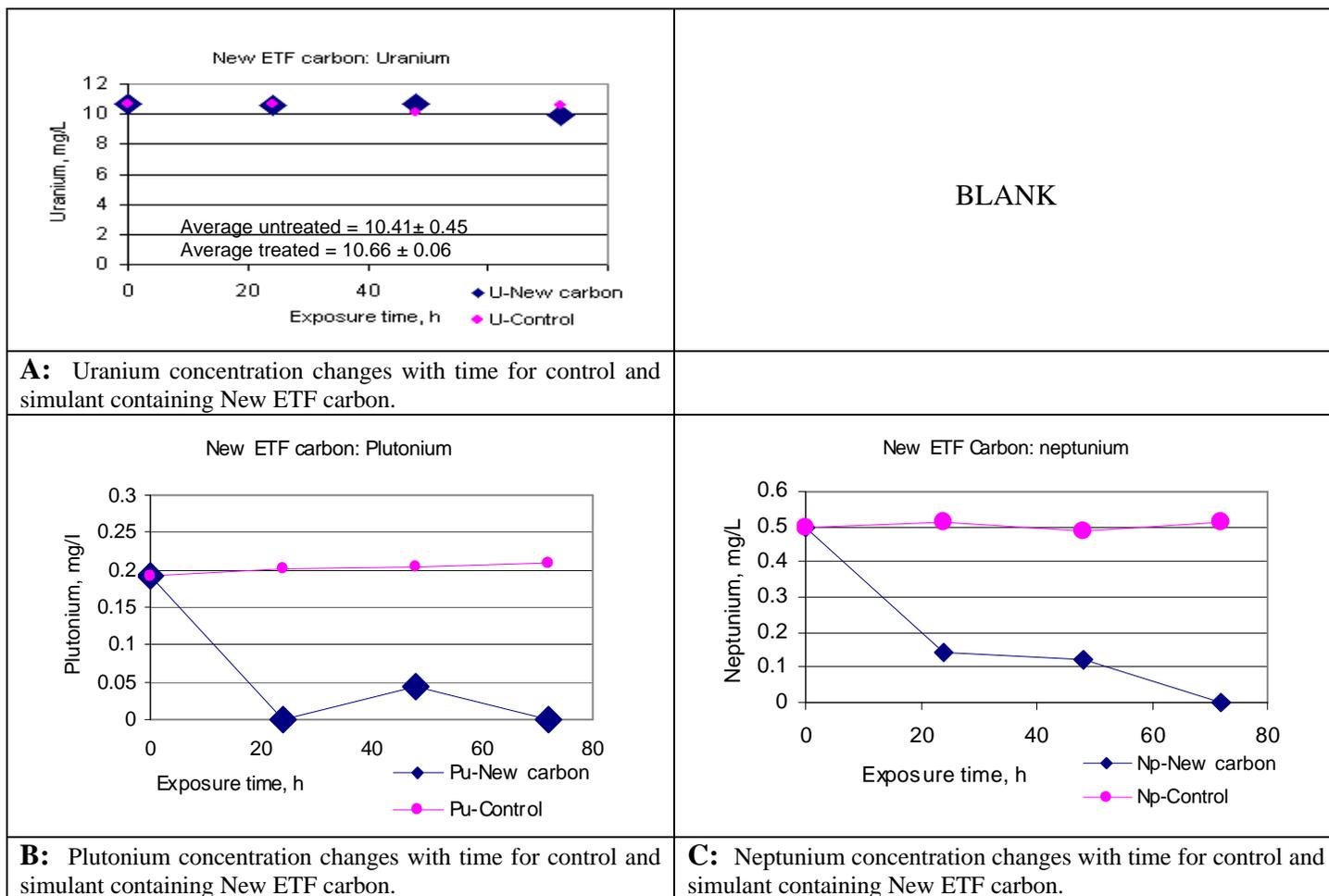


Figure 8 Changes in actinide concentration with time for new ETF activated carbon contacted with synthetic salt solution.

Since none of the materials evaluated showed signs of removing appreciable amounts of uranium from the simulant, a known uranium precipitant (anhydrous sodium phosphate) was included in the test plan to verify the test protocol.

Figure 11 inserts A and B show the results obtained with the exposure of simulant solutions to “neat” anhydrous sodium phosphate for up to 48 hours in comparison to the control runs without sodium phosphate. Sodium phosphate removed both uranium and neptunium from the simulant solution. By the end of the first 24 hours, neptunium removal had reached an equilibrium level of less than 50% of its initial concentration of 0.5 mg/L in the stimulant (Figure 11 insert B). This was not the case for uranium removal. Uranium was still being removed from solution even after 48 hours (Figure 11 insert A). By 48 hours into the test, soluble uranium concentration in the filtrate had dropped to less than 35% of its original simulant concentration of 10 mg/L.

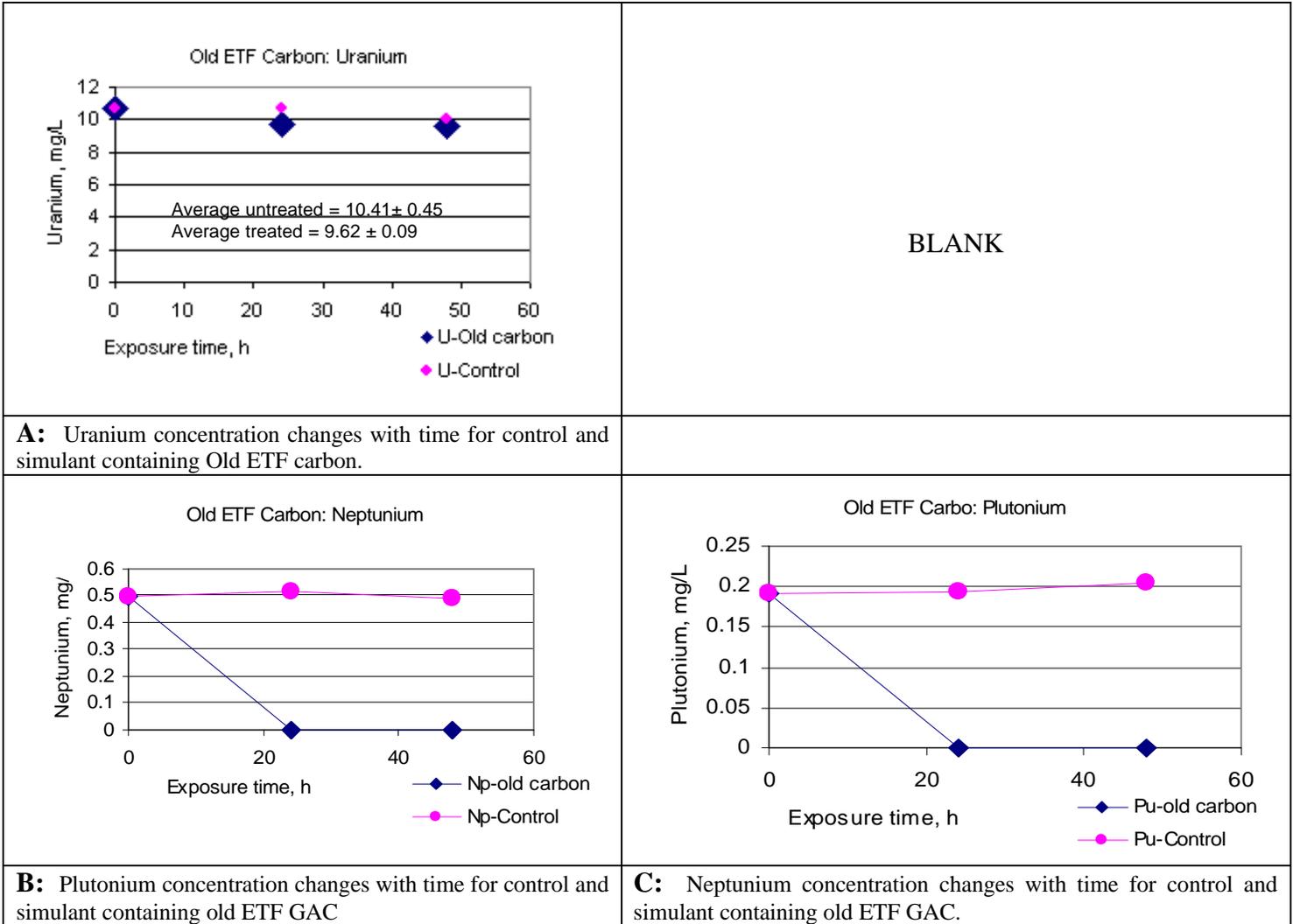


Figure 9 Changes in actinide concentration with time for Old ETF GAC contacted with synthetic salt solution.

3.8 Potassium Tetrphenylborate and Scintillation Cocktail Interactions with Actinides

The interaction of potassium tetrphenylborate (KTPB) with actinides in the synthetic salt solution containing plutonium, neptunium and uranium are summarized in Table 4. The results show that most of the actinides in solution were recovered after contacting the simulant solution with KTPB solids for up to 48 hours. The post contact data for the 24- and 48-hour samples show that the average neptunium concentration of 0.50 ± 0.01 and 0.426 ± 0.003 mg/L, respectively, were not statistically different (2σ) from the neptunium concentration in the control or its initial concentration in the simulant solution of 0.50 ± 0.05 mg/L. The plutonium post contact results with KTPB for the 24 and 48-hour samples were, respectively, 0.188 ± 0.011 , and 0.192 ± 0.011 . These values are not

significantly different from the control values of 0.188 ± 0.01 . Similarly, the 24 and 48-hour sample results for uranium are 10.41 ± 0.07 and 10.39 ± 0.02 mg/L. These values are not statistically different from the observed values for the control of 9.4 ± 0.3 mg/L.

Similarly, no measurable losses of actinides were observed when scintillation cocktail was contacted with the simulant solutions. Most of the actinides were recovered after contact for up to 48 hours, see Table 5. The neptunium concentrations after contact (24- and 48-hour samples) were, respectively, 0.50 ± 0.03 and 0.46 ± 0.01 . These values were within 2• of the of the neptunium concentration in the control (0.51 ± 0.03 mg/L and the original solution of 0.50 ± 0.05 mg/L. The plutonium 24-and 48-hour post- contact results, respectively, 0.196 ± 0.002 and 0.226 ± 0.030 mg/L, were comparable to the plutonium concentration measured in the control of 0.20 ± 0.020 mg/L.

The uranium post-contact results with scintillation cocktails for the 24 and 48-hour samples were, respectively, 10.76 ± 0.05 , and 10.70 ± 0.03 mg/L. These values are not statistically different from the control values of 10.68 ± 0.01 mg/L or the original simulant uranium concentration of 10.0 ± 1.0 mg/L.

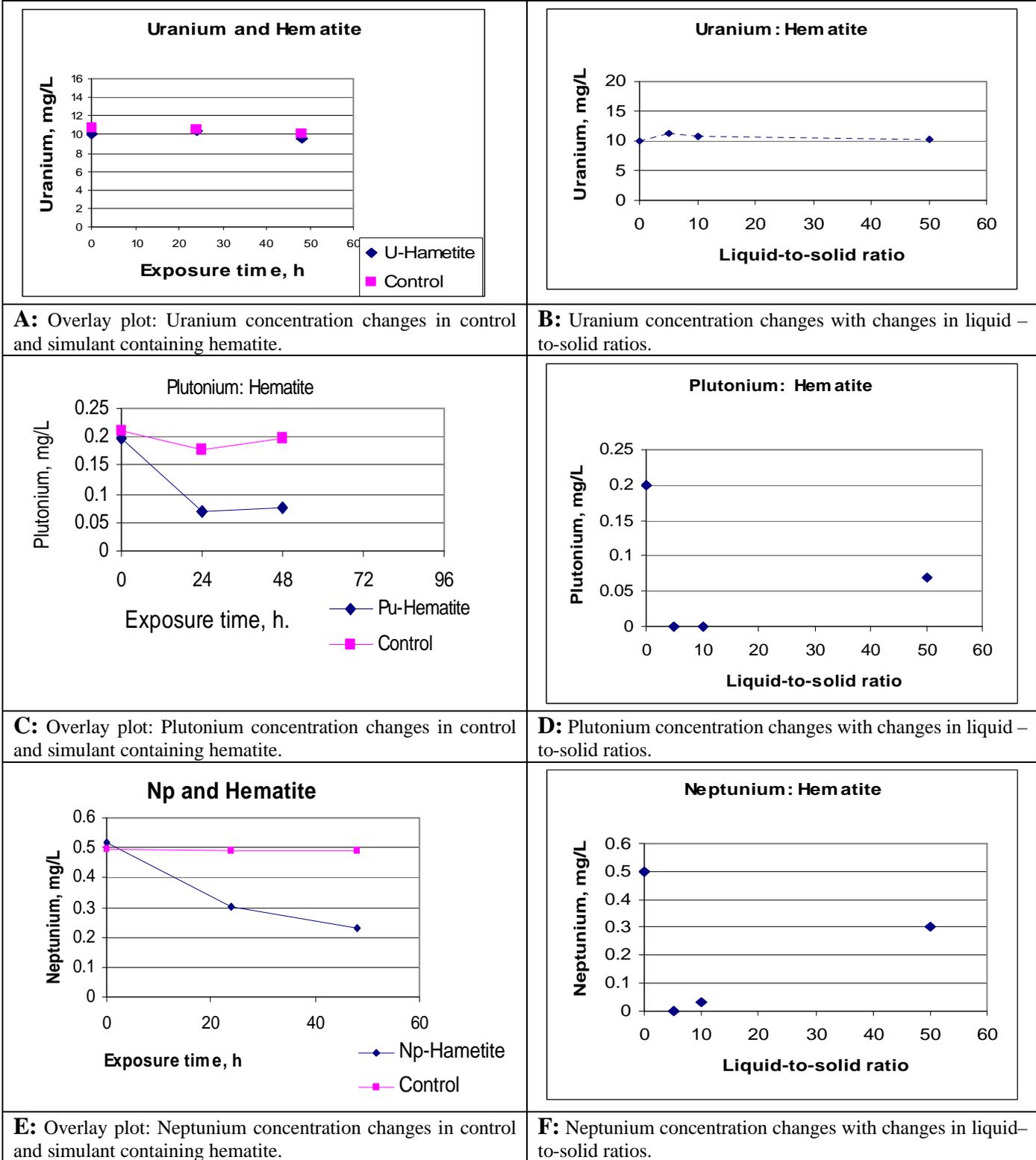


Figure 10 Changes in actinide concentration with time for hematite contacted with synthetic salt solution

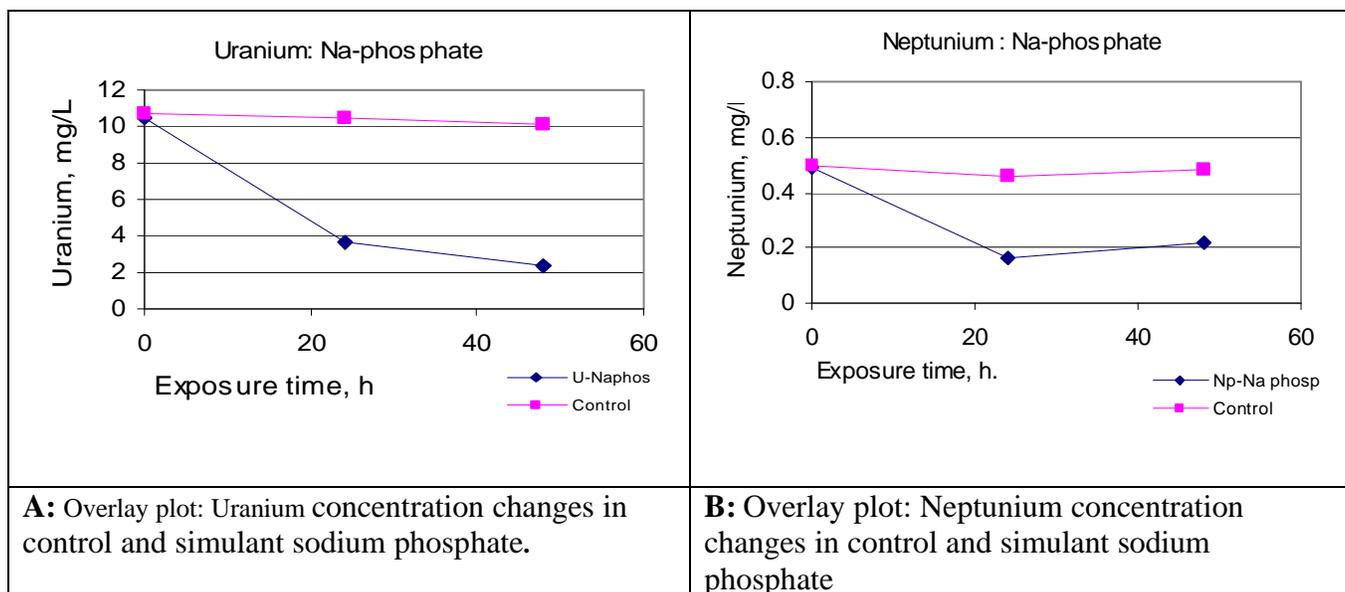


Figure 11 Changes in actinide concentration with time for sodium phosphate contacted with synthetic salt solution.

Table 4 Data summary for the interaction of actinides with potassium tetraphenylborate

| | Initial concentration mg/L | After Contact mg/L | Comments |
|------------------|----------------------------|--------------------|---------------------------------|
| Neptunium | | | No measurable loss of neptunium |
| Control | 0.5 ± 0.05 | 0.49 ± 0.030 | |
| 24-hour contact | 0.5 ± 0.05 | 0.50 ± 0.020 | |
| 48-hour contact | 0.5 ± 0.05 | 0.43 ± 0.030 | Within 2• |
| Plutonium | | | No measurable loss of plutonium |
| Control | 0.2 ± 0.02 | 0.19 ± 0.010 | |
| 24-hour contact | 0.2 ± 0.02 | 0.19 ± 0.010 | |
| 48-hour contact | 0.2 ± 0.02 | 0.19 ± 0.010 | |
| Uranium | | | No measurable loss of uranium |
| Control | 10 ± 1.0 | 10.4 ± 0.30 | |
| 24-hour contact | 10 ± 1.0 | 10.4 ± 0.070 | |
| 48-hour contact | 10 ± 1.0 | 9.44 ± 0.280 | |

Table 5 Data summary for the interaction of actinides with scintillation cocktail

| | Initial concentration mg/L | After Contact mg/L | Comments |
|------------------|-----------------------------------|---------------------------|---------------------------------|
| Neptunium | | | No measurable loss of neptunium |
| Control | 0.5 ± 0.05 | 0.52 ± 0.030 | |
| 24-hour contact | 0.5 ± 0.05 | 0.49 ± 0.020 | |
| 48-hour contact | 0.5 ± 0.05 | 0.46 ± 0.010 | Within 2• |
| Plutonium | | | No measurable loss of plutonium |
| Control | 0.2 ± 0.02 | 0.2 ± 0.02 | |
| 24-hour contact | 0.2 ± 0.02 | 0.2 ± 0.02 | |
| 48-hour contact | 0.2 ± 0.02 | 0.23 ± 0.030 | |
| Uranium | | | No measurable loss of uranium |
| Control | 10 ± 1.0 | 10.7 ± 0.010 | |
| 24-hour contact | 10 ± 1.0 | 10.8 ± 0.010 | |
| 48-hour contact | 10 ± 1.0 | 10.7 ± 0.030 | |

3.9 Plutonium and Neptunium Sorption Summary

The loading results for plutonium and neptunium onto old ETF GAC, Tank 50H solids, Tank 50H solid fraction and hematite, at a liquid -to-solid ratio of 5:1, are summarized in Table 6. The old ETF GAC and Tank 50H solids loadings were run in duplicate.

The calculated average plutonium and neptunium sorbed per gram of Tank 50H solids, respectively 0.024 and 0.056 mg are just about half the amounts for plutonium and neptunium sorption onto the old ETF GAC, which are respectively, 0.04 and 0.1 mg per gram of old ETF carbon. The sorption of these actinides from the simulant solution onto hematite is 0.1 mg plutonium per gram of hematite and 0.25 mg neptunium per gram of hematite at this liquid-to solid ratio.

Since the Tank 50H solids contain, respectively, 13.5 ± 0.5 % old ETF granular activated carbon and 0.4% iron or 1.26% iron oxide (Fe/Fe₂O₃)(See ICP-ES analysis for Tank 50H solids in reference 3), the amount of these actinides removed by these sorbents can be calculated based on their percent content in Tank 50H solids. Here we have assumed that the removal of these actinides from solution is purely by sorption and not by any other removal mechanisms such as precipitation.

If the old ETF GAC and hematite are the only two components of the Tank 50H solids responsible for the removal of all the plutonium and neptunium, and if one calculates the amounts of plutonium and neptunium removed per gram of each sorbent and add them together, the result should equal the amount of each of these actinides absorbed per gram of Tank 50H solids. The plutonium and neptunium absorbed per gram of Tank 50H solids, average respectively, 0.024 and 0.056 mg as summarized in Table 6.

A typical calculation of the amount of plutonium removal contribution by these sorbents as components of the Tank 50H solids, using 13.5% carbon content for Tank 50H solids and 1.26 % hematite (reference 3) and the neat sorbent values in Table 6 (0.1 mg Pu/ g hematite and 0.04 mg Pu/ g old ETF GAC) follows:

Plutonium:

$(0.1 \text{ mg Pu/ g hematite})(0.0126 \text{ g hematite/g Tank 50H solids}) + (0.04 \text{ mg Pu/g old ETF GAC})(0.135 \text{ g old ETF GAC/g Tank 50H solids}) = \underline{0.0067 \text{ mg Pu/ gram Tank 50H solids.}}$
 The percent Pu absorbed as a result of the combined presence of old ETF GAC and hematite in Tank 50H solids is therefore about 28% $(100 - [(((0.024 - 0.0067))/0.024) * 100])$.

Similarly for neptunium, $(0.25 \text{ mg Np/ g hematite})(0.0126 \text{ g hematite/g Tank 50H solids}) + (0.1 \text{ mg Np/g old ETF carbon})(0.135 \text{ g old ETF carbon/g Tank 50H solids}) = \underline{0.0166 \text{ mg Np/ gram Tank 50H solids.}}$ The percent Np sorbed as a result of the combined presence of old ETF activated carbon and hematite in Tank 50H solids is also about 30% $(100 - [(((0.056 - 0.0166))/0.056) * 100])$.

Hence, these results of the single components tests account for only 30% of the observed plutonium and neptunium sorbed by the Tank 50H solids. The assumption here is that the chemical and physical properties of the “neat” components evaluated are not significantly different from the similar components in the Tank 50H solids. Since these “neat” components do not account for the other 70% plutonium and neptunium removal from the synthetic salt solution by the Tank 50H components, other components of the Tank 50H solids, not evaluated here, may also be responsible for actinide removal.

Table 7 shows a summary of selected transition metal cations that are components of the Tank 50H solids (reference 3). From literature,^{7,8,9} these transition metal cations in their oxide forms are known efficient absorbents of plutonium and neptunium. Manganese oxide is known to have significantly higher affinity for plutonium and neptunium than does hematite.^{5,10} Thus, the manganese present in the Tank 50H solids, may responsible for the sorption of plutonium and neptunium not accounted for by the activated carbon and hematite.

Table 6 Plutonium and neptunium removed from synthetic salt solution per gram of sorbent after 24 hours contact.

| | Old ETF ¹ GAC | Tank 50H ² solids | Tank 50H <u>solid</u> ³ <u>Fraction*</u> | Hematite ⁴ | Liquid-to- solid ratio |
|--------------|-----------------------------|---------------------------------|--|-----------------------|---------------------------|
| Units | mg Pu/ g sorbent | mg Pu/ g sorbent | mg Pu/ g sorbent | mg Pu/ g sorbent | |
| Pu | 0.04 | 0.022 | 0.020 | 0.10 | 5:1 |
| Pu | 0.04 | 0.025 | 0.020 | NA | 5:1 |
| | | | | | |
| Units | mg Np/ g sorbent | mg Np/ g sorbent | mg Np/ g sorbent | mg Np/ g sorbent | |
| Np | 0.10 | 0.054 | 0.050 | 0.25 | 5:1 |
| Np | 0.10 | 0.059 | 0.050 | NA | 5:1 |
| | | | | | |

* Calculated by graphing the 8:1 and 25:1 liquid-to-solid ratio data and determining what the 5:1 ratio would be based on the generated linear functions ($Y_{Np} = 0.0072 \text{ ratio} + 0.01141$ and $Y_{Pu} = 0.003 \text{ ratio} + 0.005$).

1. From Figure 9
2. From Figure 4
3. From Figure 5
4. From figure 10

Table 7 Transition metal cation and Activated Carbon content in Tank 50H solids*

| Transition metal cation or Activated carbon | mg/g Tank 50H solids | Equivalent metal oxide | mg metal oxide /g Tank 50H solids | % metal oxide |
|---|-------------------------|--------------------------------|---|---------------|
| Fe | 4.40 | Fe ₂ O ₃ | 12.60 | 1.26 |
| Mn | 0.24 | MnO ₂ | 0.620 | 0.0620 |
| Zn | 0.325 | ZnO | 0.448 | 0.0484 |
| Cr | 0.062 | Cr ₂ O ₃ | 0.181 | 0.0180 |
| Ti | 0.07 | TiO | 0.117 | 0.0117 |
| | | | | |
| Old ETF GAC | 0.135 | - | - | 13.5 |

* From reference 3.

3.10 Plutonium and Neptunium Loading Curves with “Neat” ETF Carbons, Hematite, and Sodium phosphate.

Loading curves for plutonium, neptunium and uranium were calculated for various “neat” sorbents using results from equilibrium tests over a range of simulant-to-solid ratios. Since none of the single components identified in the Tank 50H solids showed measurable uranium sorption, the only uranium loading curve obtained was for the control (sodium phosphate).

The loading curves presented here are based on a minimum of six different simulant-to-solid ratios. The concentration of each actinide on the solid phase was calculated from the difference between the initial and final solution concentration. The solids were not analyzed. Figures 12, Figure 13, and Figure 14 show neptunium-237 and plutonium-239 loading curves, respectively, for the old ETF granular activated carbon, the new ETF activated carbon, and hematite after 24 hours exposure to the simulant solution at room temperature. Figure 15, inserts A and B show, respectively, the loading curves on sodium phosphate for neptunium-237 and uranium after 24 hours of exposure to the simulated salt solution bearing these actinides.

Some of the loading curves presented in the graphs, for example Figure 14 insert A for loading of neptunium onto hematite, do not have sufficient data points to clearly characterize the maximum loading area of the curve, which is the plateau region of the Langmuir isotherm. The sorption equilibrium for these actinides under these conditions may not have been attained after 24 hours of contacting them with the sorbents. To estimate what the maximum actinide loading values are at the plateau region of the curves a second degree polynomial fitting was used for all the loading data. The use of a second degree polynomial fitting function for the loading curve data applies only up to the inflection point of the curve and not beyond that point and only if that point falls within the data set. CAUTION! These equations may not be used to extrapolate to values outside the data set.

Thus, a second degree polynomial regression of the data, Figures 12 through 15 provides the following equations for estimating maximum loading of plutonium-239, neptunium-237, and uranium (sodium phosphate only) on these precipitants:

Plutonium-239:

$$y = -0.0518x^2 + 16.029x + 7.3876 \text{ for new ETF activated carbon}$$

$$y = -9E-05x^2 + 0.0352x \text{ for old ETF GAC}$$

$$y = -0.0004x^2 + 0.0959x \text{ for hematite}$$

Neptunium-237:

$$y = -3E-05x^2 + 0.0212x - 0.0247 \text{ for new ETF activated carbon}$$

$$y = -6E-05x^2 + 0.044x \text{ for old ETF GAC}$$

Projected value, $y = -2E-05x^2 + 0.0344x - 0.1176$, is outside of data set for hematite (see Figure 14 insert A).

$y = -37.53x^2 + 31.807x$ for sodium phosphate

Uranium:

$y = -4.183x^2 + 55.584x$ for sodium phosphate,

where y is loading capacity in units of μg actinide per gram of “neat” sorbent material and x is the final actinide concentration in $\mu\text{g/L}$. By equating the first derivatives of each loading capacity equation above to zero and solving for x values, the maximum loading capacities (y-values) for plutonium and neptunium were determined from the absorption data.

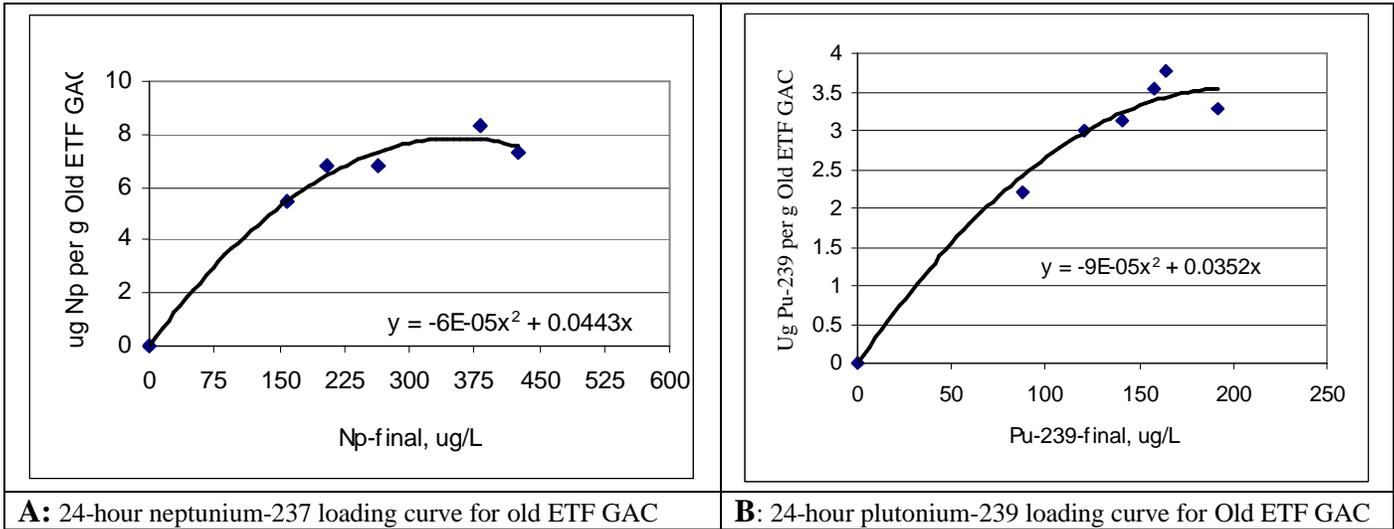


Figure 12 Neptunium and plutonium loading curves for Old ETF Activated carbon.

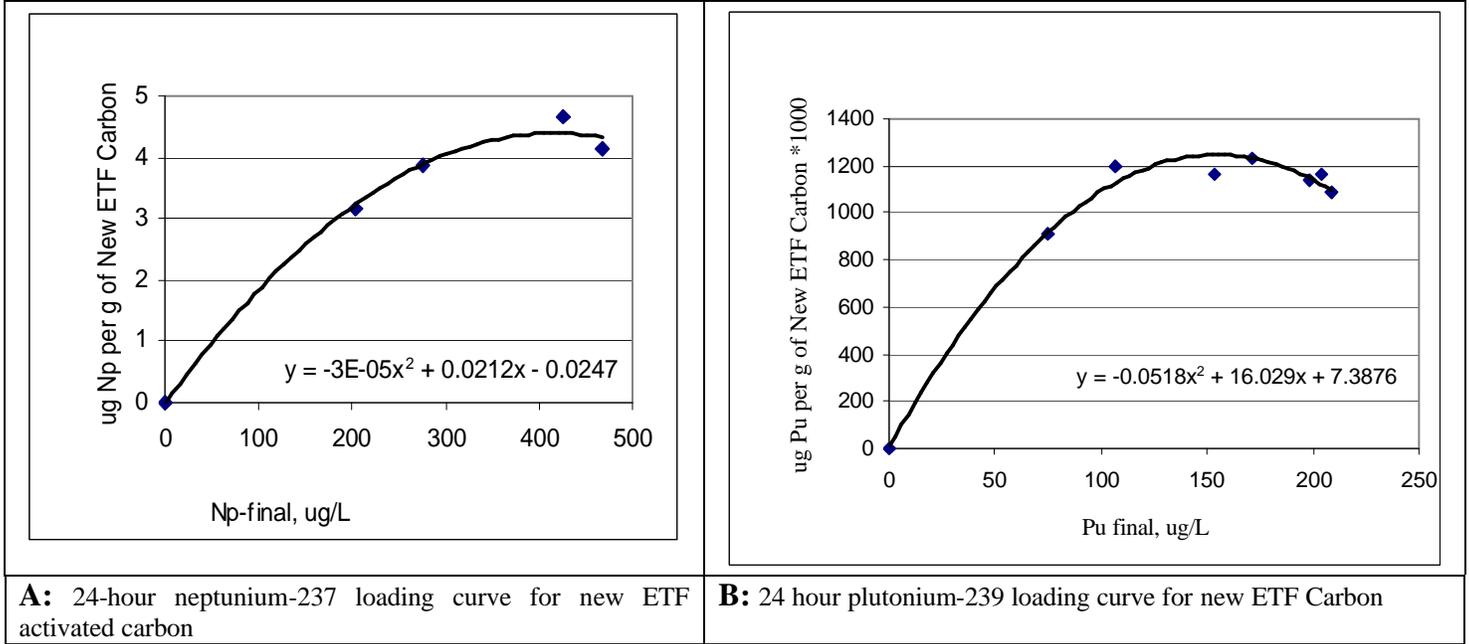


Figure 13 Neptunium and plutonium loading curves for New ETF activated carbon.

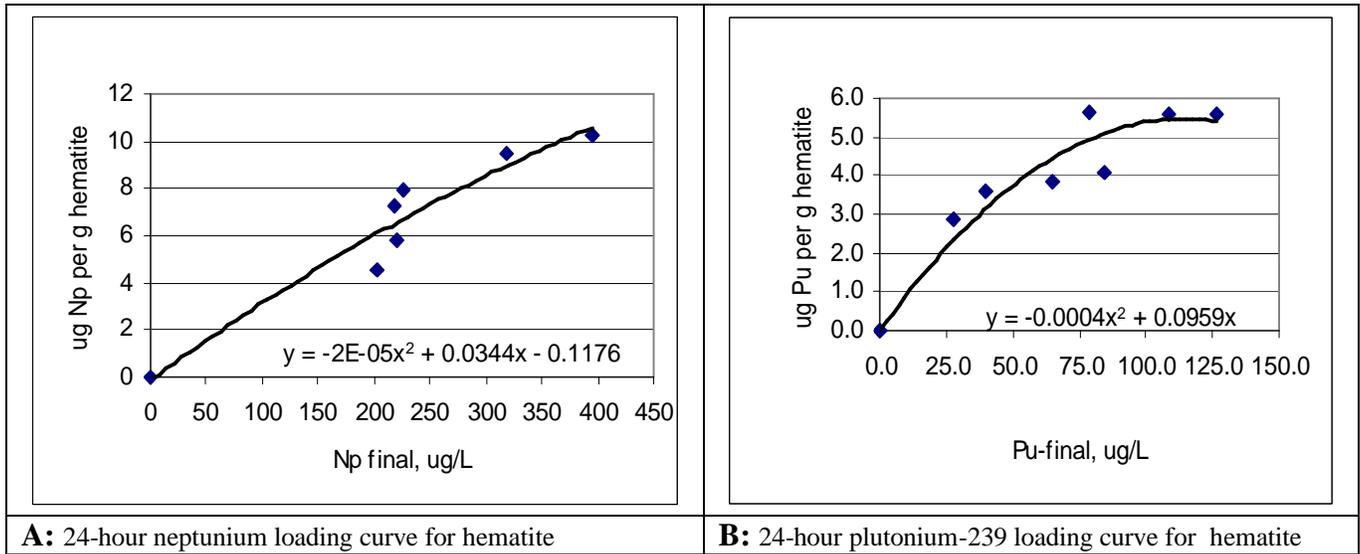


Figure 14 Neptunium and plutonium loading curves for hematite.

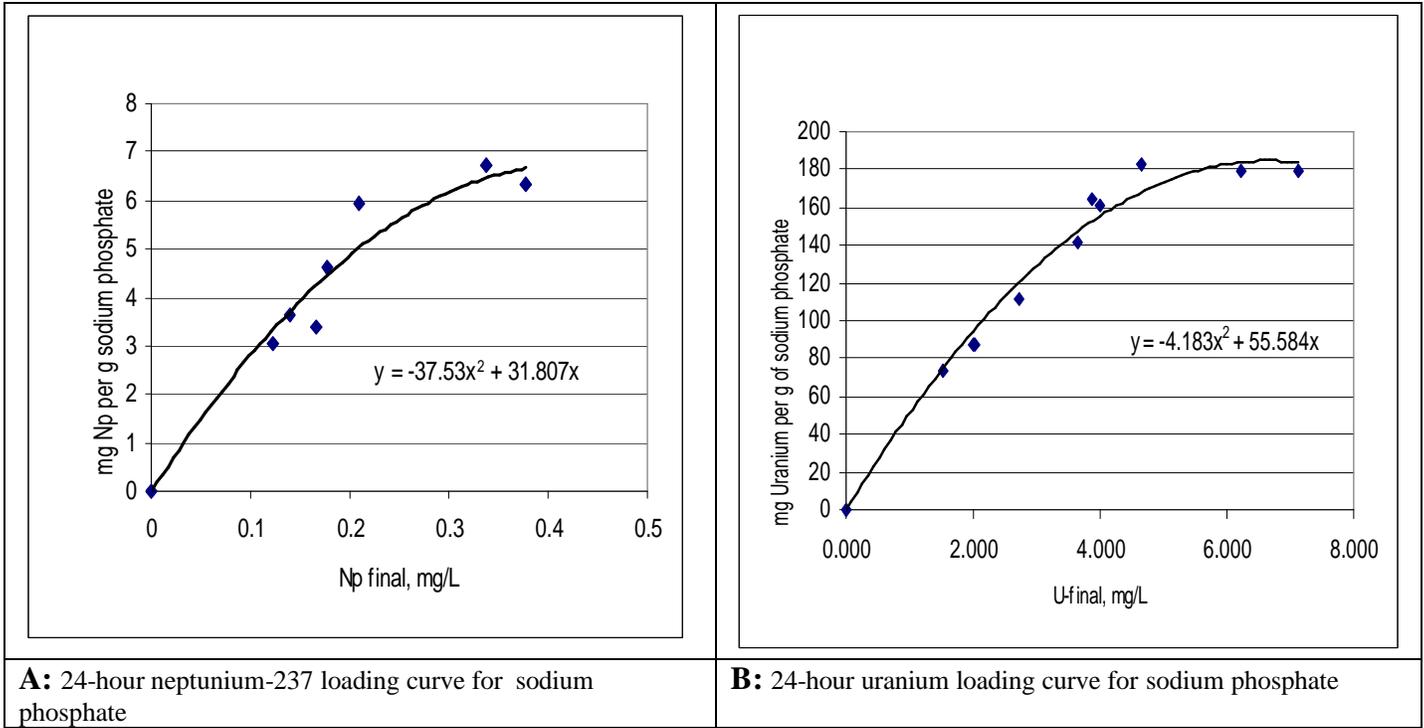


Figure 15 Neptunium and uranium loading curves for sodium phosphate.

Table 8 shows a summary of loading data for plutonium, neptunium and uranium on, respectively, Tank 50H solids⁴, “neat” old and new ETF carbons, “neat” hematite and sodium phosphate. Based on these loading curve data, “neat” hematite and “neat” old ETF activated carbon have the highest affinity for both plutonium and neptunium at, respectively, 5.5 ± 0.38 and $> 10 \mu\text{g}$ per gram hematite and 7.9 ± 0.52 and $3.6 \pm 0.22 \mu\text{g}$ per gram old ETF activated carbon. The new ETF carbon with a loading of $1.3 \pm 0.070 \mu\text{g}$ plutonium and $4.4 \pm 0.44 \mu\text{g}$ neptunium per gram, exhibits lower affinity for these actinides than the old ETF granulated activated carbon. Sodium phosphate (used here as a reference sorbent for uranium) as expected exhibited a very high affinity for uranium. Its affinity for neptunium was comparable to that of the old ETF activated carbon, although it showed no measurable affinity for plutonium. Both uranium and neptunium were readily absorbed in the presence of anhydrous sodium phosphate. The loading capacity values were, respectively, 184.6 ± 18.5 and $6.8 \pm 0.6 \mu\text{g}$ per gram of anhydrous sodium phosphate.

Therefore, the relative order for loading plutonium and neptunium onto these sorbents is iron oxide (hematite) > old ETF GAC > New ETF activated carbon >> Tank 50H solids.

Loading capacity information provides a basis for estimating the maximum volume of Tank Farm supernates that can be transferred into the Tank 50H before accumulating sufficient quantities of plutonium to pose a nuclear criticality safety concern. Conversely, the information could be used to determine how much of the Tank solids could be left behind and not present a safety concern due to the accumulation of plutonium and

neptunium. The loading capacities for plutonium and neptunium with Tank 50H solids contacted with synthetic salt solutions bearing soluble levels of these actinides are, respectively, 2.01 and 4.48 μg per gram of Tank 50H solids.⁴ But for plutonium and neptunium loading capacities onto “neat” hematite, these Tank 50H solids loading capacities for plutonium and neptunium are within 40-50% loading capacities for plutonium and neptunium onto “neat” ETF GAC as summarized in Table 8.

This result also supports the finding that an additional component in the 50H solids, besides activated carbon and iron oxides, contributes to the observed sorption of plutonium and neptunium.

Table 8 Loading curve summary

| Solids | Maximum loading $\mu\text{g/g}$ solid | Maximum loading $\mu\text{g/g}$ solid | Maximum loading $\mu\text{g/g}$ solid |
|------------------------------|--|--|--|
| | | | |
| | Pu | Np | U |
| Tank 50H solids* | 2.01 | 4.48 | NA |
| | | | |
| Old ETF GAC | 3.6 ± 0.22 | 7.9 ± 0.52 | NA |
| New ETF Carbon | 1.3 ± 0.070 | 4.4 ± 0.44 | NA |
| | | | |
| Iron Oxide (hematite) | 5.5 ± 0.38 | >10.0 | NA |
| | | | |
| Sodium phosphate (anhyd.) | Not measured | > 6.0 | 184.6 ± 18.5 |

NA = No measurable absorption of uranium.

- See reference 4.

4.0 CONCLUSIONS

Based on the batch experiments performed in this study the following conclusions are made:

- The removal of plutonium and neptunium from the synthetic salt solution by Tank 50H solids is due largely to the presence of the old ETF granular activated carbon and transition metal oxides, likely iron and manganese oxides, in Tank 50H solids. These components of the Tank 50H solids are known active sorbents of both plutonium and neptunium.
- Both activated carbon and iron oxide (hematite), which are components of Tank 50H solids, contribute to the removal of Pu and Np by Tank 50H solids and in combination, they account for about 30% of the Pu and Np removed by Tank 50H solids.

- Nitrated cancrinite in contact with simulant salt solution containing uranium, plutonium and neptunium does not show any measurable tendency to remove these actinides from salt solution.
- The two forms of activated carbon investigated in these tests show measurable affinity for plutonium and neptunium.
- The new ETF activated carbon did not remove any measurable amount of uranium from the simulant salt solution, while the old ETF carbon removed less than 10% of the uranium.
- The new ETF activated carbon exhibits lower affinity for plutonium and neptunium than the old ETF carbon.
- The old ETF granular activated carbon and iron oxide (hematite) exhibit high affinity for both plutonium and neptunium.
- Hematite exhibits greater affinity for plutonium and neptunium than the ETF carbons.
- Both potassium tetrphenylborate and scintillation cocktail did not show any measurable interaction or removal of uranium, plutonium and neptunium from the synthetic salt solution.
- Sodium phosphate (used here as a reference sorbent for uranium), as expected, exhibited high affinity for uranium and neptunium, but failed to show any measurable affinity for plutonium.
- Plutonium and neptunium loading capacity onto “Neat” old ETF granular activated carbon are, respectively, 3.6 ± 0.22 and 7.9 ± 0.52 μg per gram of old ETF granulated activated carbon.
- Plutonium and neptunium loading capacity onto “Neat” new ETF activated carbon are, respectively, 1.3 ± 0.070 and 4.4 ± 0.44 μg per gram of new ETF activated carbon.
- The loading capacity for plutonium and neptunium onto hematite are, respectively, 5.5 ± 0.38 and > 10 μg per gram of hematite.
- Plutonium loading capacity onto the Tank 50H solids (2.01 μg per gram of Tank 50H solids) is about 60% of plutonium loading capacity onto “neat” ETF granular activated carbon and 40% of plutonium loading capacity onto “neat” hematite.
- Neptunium loading capacity onto the Tank 50H solids (4.48 μg per gram of Tank 50H solids) is about 50% of plutonium loading capacity onto “neat” ETF granular activated carbon and less than 50% of neptunium loading capacity onto “neat” hematite (iron oxide).

Additional testing to verify if other transition metal oxides (MnO_2 and ZnO) are responsible for actinide removal not accounted for by the presence of activated carbon and hematite is recommended.

5.0 QUALITY ASSURANCE

This study fulfills the activity defined in Task Technical and Quality Assurance Plan: "Identification of Tank 50H Solids Components Responsible for Removal of Dissolved Actinides" WSRC-RP-2003-0388, Rev.0, April 17, 2003. Data obtained from this study reside as records in WSRC-LB-2001-00179.

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