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Review of Actinide and Strontium Loading Data for MST and mMST

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EXECUTIVE SUMMARY

SRNL reviewed the relevant data from MST and mMST fissile loading studies to determine if further studies were required.

With respect to MST, SRNL found that the published results adequately bound the expected conditions that Small Column Ion Exchange (SCIX) process will operate under. The lack of strontium data does not represent an issue as strontium is not relevant to criticality. There is no threat to criticality safety from the lack of strontium loading data.

However, SRNL proposes a single test with MST to ensure that future SCIX operations are conservatively bounded and strontium maximum loading is understood.

With respect to attempts to maximally load mMST, SRNL's knowledge on actinide and strontium loading is limited to uranium behavior. mMST has a very weak affinity for uranium, and even extended contact time at high uranium concentration shows minimal loading onto mMST. This leaves questions about the ability to load plutonium, neptunium and strontium. SRNL proposes to perform two tests with mMST to ensure that questions on plutonium, neptunium, and strontium sorption are answered, as well as ensuring that future mMST operations are conservatively bounded.

LIST OF ABBREVIATIONS

DF – Decontamination Factor
MCU – Modular Caustic Side Solvent Extraction Unit
mMST – modified monosodium titanate
MST - Monosodium Titanate
SCIX – Small Column Ion Exchange
SRNL - Savannah River National Laboratory

1.0 Introduction

As part of the SCIX program (formerly known as the Modular Salt Processing program), SRNL was tasked with performing a number of studies to determine if technology gaps exist in key aspects of the program.[1,2,3] One facet of this work was to review the current literature on maximizing MST and mMST actinide and strontium loading to determine if further experimental studies were required.

2.0 Experimental Procedure

Maximal actinide and strontium loading on MST and mMST is a subject that has not undergone a great deal of scrutiny. Typical studies with MST and mMST have not focused on maximizing loading of actinides and strontium but instead focused on removal after specific times, such as 24 hours. A single document from 2006 is the primary study for maximal MST loading,[4] and a single document from 2008 is the primary study for maximal mMST loading.[5] Finally, a document from 2006 examined Dubinin-Astashov predictions of actinide loading on MST.[6]

SRNL reviewed these documents to determine whether or not a critical gap in the knowledge base exists on the subject of maximal actinide and strontium loading on MST and mMST.

3.0 Results and Discussion

3.1 MST Maximum Loading of Actinides and Strontium

In the 2006 document on MST loading,[4] large volumes of radioisotope-spiked salt simulant were contacted with small masses of MST (8.5L to 0.2g of MST solids) for extended periods of time (~3200 hours) to load the MST to a maximum extent. SRNL tracked the $^{239/40}Pu$, ^{238}U, ^{237}Np, and ^{85}Sr removal over time. At the ~360 hour time, SRNL determined the weight % loading of $^{239/40}Pu$, ^{238}U, ^{237}Np as follows:

Pu: 2.79 ± 0.197 wt %,
U: 14.0 ± 1.04 wt %,
Np: 0.839 ± 0.0178 wt %

These weight % loading values are the highest measured to date. The experiments were designed to provide the best chance scenario to load the MST due to the following factors:

- high liquid:MST solids ratio
- high concentrations of actinides (near their estimated solubility limits)
- extended time periods

At this time, the operating conditions expected to be used for the SCIX program are similar to those used in current MCU operations – low actinide and strontium concentrations, high cesium concentrations, and a sodium concentration in the 5-7M range. Given these conditions, SRNL feels that the previously measured loadings of Pu, U, and Np conservatively bound the loadings that would occur in SCIX operations.

With respect to strontium, SRNL feels that a knowledge gap exists. In the case of Sr, the very low mass concentrations of ^{85}Sr and cold strontium (~100 μg/L) prevented SRNL researchers from determining loadings at maximum extents. Although Sr does not present a criticality safety concern, determination of bounding Sr loading is important for understanding if "hot spots" from Sr-loaded MST are possible.

To predict strontium sorbtion, as well as longer term actinide sorption, SRNL recommends one long term test with MST. SRNL proposes a test in duplicate be done at a high liquid:solid ratio (preferably, the same ratio as used in the previous MST loading work[4]), using simulated waste that is roughly similar in bulk chemical composition to that being fed to MCU i.e., (dissolved saltcake), but with more challenging actinide and strontium concentrations (i.e., dissolved saltcake is too low in actinides and strontium to provide a challenging loading environment). SRNL will allow this test to continue over a long period of time (6 months to 1 year), and will sample the test solution at regular intervals to monitor the actinide and strontium removal. MST loading will be inferred from sorbate depletion in the solution. At the end of the test, the MST will be collected, digested, and analyzed for loading to corroborate the solution analyses.

3.2 mMST Maximum Loading of Uranium

In contrast to the MST study, the single appropriate mMST study only examined the loading of uranium onto mMST. In this study, a simulant was spiked with a high level of depleted uranium and contacted with different levels of mMST and MST for a maximum of 2 weeks. The study found that the maximum uranium loading, after ~336 hours was ~0.043 wt % compared to ~14 wt % for MST (although the experimental conditions were not exactly the same). Clearly, mMST exhibits very poor affinity for uranium and does not load uranium to the same extent as MST. Therefore, SRNL does not feel there is a significant knowledge gap with respect to the maximum loading of uranium onto mMST.

With respect to the other sorbates, mMST exhibits much faster removal of Sr and Pu and generally faster removal of Np compared to MST. However, the loading of these sorbates at extended contact time and high sorbate concentrations has not been studied. Thus, the maximum loading of Pu, Np and Sr cannot be bounded by or inferred by measured results for previous tests.

Therefore, SRNL proposes two mMST tests, under similar conditions as proposed for the MST test. One option is to use mMST at a lesser concentration than the MST, since mMST will probably be used at a lesser concentration normally.

4.0 Conclusions

Previous MST loading studies are under conditions that are conservative compared to the expected operating conditions of SCIX. SRNL recommends determining the bounding loading of Sr onto MST to ensure that estimates of maximum ^{90}Sr loading onto MST are known.

Previous mMST studies with regard to maximum sorbate loading are limited to uranium. Since loading of Pu, Np and Sr are not known at bounding conditions, SRNL recommends performing two tests to measure the loadings at bounding conditions.

5.0 References

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