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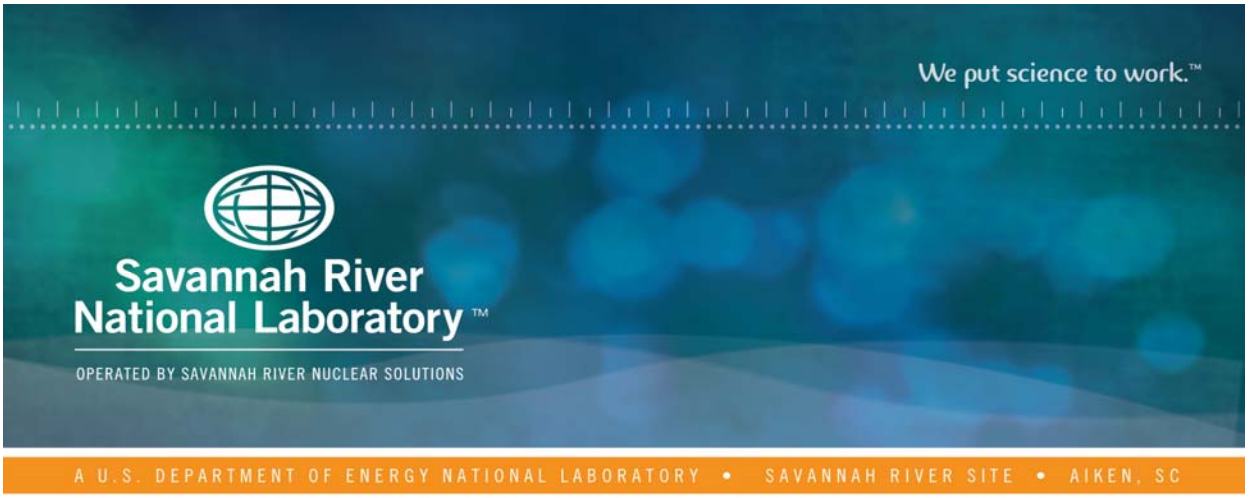

Sample Results from the Interim Salt Disposition Program Macrobatch 8 Tank 21H Qualification MST Solids Sample

T. B. Peters

A. L. Washington, II

February 2015

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REVIEWS AND APPROVALS

AUTHORS:

T. B. Peters, Advanced Characterization and Processing Date

A. L. Washington, II, Advanced Characterization and Processing Date

TECHNICAL REVIEW:

C. A. Nash, Advanced Characterization and Processing Date

APPROVAL:

F. M. Pennebaker, Manager Date
Advanced Characterization and Processing

S. L. Marra, Manager Date
Environmental & Chemical Process Technology Research Programs

E. J. Freed, Manager Date
DWPF/Saltstone Facility Engineering

J. S. Contardi, Manager Date
Tank Farm Engineering

R. E. Edwards, Manager Date
Nuclear Safety and Flowsheet/Process Integration

EXECUTIVE SUMMARY

Savannah River National Laboratory (SRNL) analyzed samples from Tank 21H in support of qualification of Macrobatch (Salt Batch) 8 for Interim Salt Disposition Program (ISDP) processing. As part of this qualification work, SRNL performed an Actinide Removal Process (ARP) and several Extraction, Scrub, Strip (ESS) tests. This document reports characterization of the monosodium titanate (MST) solids from the ARP test. The results of these analyses are reported and are within historical precedent.

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LIST OF ABBREVIATIONS

ARP	Actinide Removal Process
DF	Decontamination Factor
ESS	Extraction, Scrub, Strip
HTF	H-Tank Farm
ICPES	Inductively Coupled Plasma Emission Spectroscopy
ICPMS	Inductively Coupled Plasma Mass Spectroscopy
ISDP	Interim Salt Disposition Program
MST	Monosodium Titanate
NAA	Neutron Activation Analysis
SRNL	Savannah River National Laboratory
SRR	Savannah River Remediation
TTQAP	Task Technical and Quality Assurance Plan
TTR	Technical Task Request
WAPS	Waste Acceptance Product Specifications

1.0 Introduction

This report details the results of the analysis of MST solids recovered from the ARP test. Characterization of the qualification sample, results of the MST removal of strontium and actinides, and results of the ESS tests were previously reported.[1,2] This work was specified in a Technical Task Request (TTR)[3] and in a Task Technical and Quality Assurance Plan (TTQAP).[4] Details for the work are contained in a controlled laboratory notebook.[5]

For this macrobatch, Tank 21H is used as the blend and preparation tank. This material will be transferred to Tank 49H where it will be combined with the heel in Tank 49H prior to processing. In this qualification effort, only samples from Tank 21H have been characterized and treated. The goal for the MST testing during salt batch qualification is to simulate a single MST strike with 0.2 g/L MST.

2.0 Experimental Procedure

Six 200 mL Tank 21H samples (i.e., sample bottles HTF-21-14-126, -127, -128, -130, -131, -132) arrived at SRNL on September 18, 2014. Four of these samples were surface samples and two were obtained approximately 62" from the bottom of the tank (transfer pump suction). The slurry pumps were run for approximately 22 hours, and the samples were pulled approximately 48 hours after the slurry pumps were shutdown. All the samples had the same visual appearance, clear solutions with no solids. While the previous salt batch samples contained solids, the mixing program and timing for this set of samples precluded capturing any of these solids. The density of filtered solution (using a 0.45 μm syringe filter) from each sample was measured and is previously reported.[1] With Savannah River Remediation (SRR) concurrence, the contents of the six sample bottles were then composited and mixed. This composite solution was used for analysis and for the ARP test.

For the ARP test, approximately 500 mL of the ISDP Salt Batch 8 Tank 21H composite material was obtained for testing. Normally, for the salt batch qualification, the material used in the ARP test is decanted. Due to the inability to clearly determine if solids were present or not, and whether they would settle, before use in the ARP test, the 500 mL was filtered through a 0.45 μm filter cup before use.

The composite salt solution was previously measured with a density of 1.257 g/mL at 23 °C. 200 mL each (totaling 400 mL) of the well mixed salt solution was placed into the first and second experiment bottles, while the remainder (100 mL) was placed into the control bottle. Two experiment bottles were used in order to provide enough solution for the later ESS tests and enough MST solids for future analysis. All three bottles had magnetic stir bars added to provide sufficient mixing for batch contact tests. The target

concentration for MST was 0.2 g/L. Personnel added 0.253 g of MST solids in a 16.4 wt% solution from Blue Grass Chemical Specialties MST-2723 to each experiment bottle. This material was an archived batch that has been utilized on all recent salt batches by SRNL. The time was recorded and designated as time 0. Throughout the course of the MST test, agitation and temperature control (via water bath, 25±3 ºC) were provided.

During the experiment, filtrate samples were collected and analyzed as previously reported.[2] When the filtrate results were received, it was found that the decontamination factor (DF) values for experiment #1 were far lower than experiment #2 or historical results. As well, the titanium result from the digested solids from experiment #1 was far lower than experiment #2, indicating that not all of the MST was added to the experiment #1 bottle. As a result, a third experiment and concurrent control were run, under the same conditions, using the same feed, but at a smaller scale, and this third test gave DF results similar to experiment #2 as well as historical precedent.[6, 7] Therefore, characterization from MST only included the results from experiment #2 and experiment #3.

2.1 Analysis of MST Solids

The MST addition and adsorption were performed in three identical contact experiments, one of which (#1) had to be discarded from consideration as previously discussed. The purpose of performing duplicate adsorptions was to provide enough material to test for additional analytes (^{14}C, ^{129}I) and to provide enough material for two ESS tests. After the MST test completed, the MST solids were collected using unwashed filtration in two separate filter cups (one for each of experiment #2 and #3) while the liquid was combined to serve as the feed for the ESS testing. Personnel digested the retained MST solids (aqua regia/microwave) in one filter cup (from experiment #2) for MST solids analysis and retained the solids from the other filter cup (from experiment #3) and sent them to Analytical Development for ^{14}C and ^{129}I analysis. As it is problematic to attempt to isolate only the MST solids, SRNL digested the collected slurry and normalized all the results to titanium, giving a result in “pCi analyte/gram of titanium”. Inductively Coupled Plasma Mass Spectroscopy (ICPMS), Inductively Coupled Plasma Emission Spectroscopy (ICPES), and various radio-counting methods were used for analysis.

The analyses for ^{14}C and ^{129}I were completed using solid samples with other preparations outside of aqua regia/microwave dissolution. The filter from experiment #3 containing both the MST and some salt solution was washed gently with water to remove as much of the salt solution as possible. Assuming the equal distribution of material, the filter was segmented into two sections. One section was utilized for the analysis of ^{129}I, and one section was used for measuring ^{14}C. For ^{129}I, the filter material was digested in a solution of nitric acid to which some potassium iodide was added. The iodide was subsequently precipitated using silver to form silver iodide (AgI). The activity of the ^{129}I was then determined by low energy gamma spectrometry. The AgI precipitate was measured by

Neutron Activation Analysis (NAA) to determine the recovery of the original added iodide.

For the ^{14}C analysis, the filter material was processed using a combustion wet-ashing technique oxidizing the carbon in the sample to carbon dioxide. The carbon dioxide was captured and analyzed by liquid scintillation counting for ^{14}C.

2.2 Quality Assurance

Requirements for performing reviews of technical reports and the extent of review are established in Manual E7, Procedure 2.60. SRNL documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2. The Quality Assurance requirements of RW-0333P were applied to those activities identified as waste form related to the analysis of the Salt Batch 8 qualification samples (i.e., MST sludge solids and Waste Acceptance Product Specification (WAPS) radionuclides). Personnel qualified to RW-0333P performed and reviewed this work.

3.0 Results

As there were few sludge solids in the feed material after the initial filtering, the solids digestion data reflects the MST solids, and whatever adsorbs to the MST, as well as entrained/interstitial salt solution.

Most actinides and strontium adsorb to MST and the analysis of the MST provides relevant data for those species. Additionally, elements including Eu, Co, and Am, have the potential to adsorb depending on the alkalinity of the solution and solubility.[8,9]

Many species, such as cesium, tin, and technetium are not expected to adsorb to MST. The results for these radioisotopes are likely from residual feed solution that remains as interstitial liquid. This is confirmed by noting that the ^{137}Cs:^{126}Sn and ^{137}Cs:^{99}Tc ratios are similar to that of the feed solution. Therefore, the values reported in Table 1 should all be considered upper bounds.

Results for these elements encompass the MST solids, non-visible solids present in the filtered solution, and the material from interstitial or entrained salt solution due to hydration in the absence of washing during the experiment. As there are no experimental data for many of these analytes (all except Sr, Pu, Np, Co, Eu, and Am which are directly adsorbed by MST) as to whether or not they adsorb to MST under our conditions without a washing step, SRNL cannot conclusively determine if the real values for an analyte result are from MST solids adsorption or interstitial liquid entrainment.

All results were single results as specified in the TTQAP. However, there is a blank generated to ensure instrument calibration. The blank does not show any cross contamination from any analyte.

Values in parentheses are the analytical uncertainty. The total alpha result is from a cesium-removed sample. This was determined to be a more appropriate analysis as it reduces the interference from the beta emitters (this has not been done for previous salt batches). The total beta sample did not have the cesium removed.

Table 1. Tank 21H MST Solids Radiological Results

Analyte	Result (pCi per gram of Ti)	Analyte	Result (pCi per gram of Ti)
^{3}H	<5.69E+04	^{235}U	1.67E+03 (20%)
^{14}C	<9.22E+04	^{237}Np	<5.57E+03
^{60}Co	<1.28E+04	^{238}U	3.33E+04 (20%)
^{90}Sr	1.65E+08 (24%)	^{238}Pu	3.56E+07 (6.2%)
^{94}Nb	<1.27E+04	$^{239/40}Pu$	6.50E+05 (9.9%)
^{99}Tc	9.30E+06 (7.1%)	^{241}Pu	1.41E+07 (15%)
^{106}Ru	<3.83E+05	^{241}Am	<4.93E+04
^{125}Sb	<3.12E+05	^{242}Pu	<3.78E+08
^{126}Sb	1.13E+05 (8.4%)	^{242m}Am	<1.53E+04
^{126}Sn	1.13E+05 (8.4%)	^{242}Cm	<6.00E+02
^{129}I	<7.20E+05	^{243}Am	<7.59E+03
^{134}Cs	<3.54E+06	^{243}Cm	<2.36E+04
^{137}Cs	3.45E+10 (5.0%)	^{244}Pu	<1.75E+06
^{144}Ce	<3.65E+05	^{244}Cm	2.55E+04 (16%)
^{147}Pm	<3.86E+07	^{245}Cm	<1.95E+04
^{151}Sm	<2.72E+07	^{247}Cm	<2.85E+04
^{154}Eu	<4.09E+04	^{249}Cf	<2.68E+04
^{155}Eu	<1.34E+05	^{251}Cf	<2.33E+04
^{226}Ra	<1.39E+06	Total Alpha (Cs-removed)	4.89E+07 (10%)
^{233}U	1.07E+05 (20%)	Total Beta	3.96E+10 (10%)
^{234}U	4.39E+05 (20%)		

Generally speaking, the results from the Salt Batch 8 MST solids tend to be higher than from the Salt Batch 7 MST solids, with some exceptions, such as elements that tend to remain as sludge at high pH (Am, Cm, etc). These higher values in the Salt Batch 7 MST solids may be a reflection of the Salt Batch 8 initial filtration removing some sludge fines.

4.0 Conclusions

Analysis of the MST solids sample indicates that the material does not display any unusual characteristics. In conjunction with the previous reports,[1,2] the macrobatch 8 solution in Tank 21H, when combined with the existing Tank 49H heel is acceptable for processing in the ISDP process.

5.0 References

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