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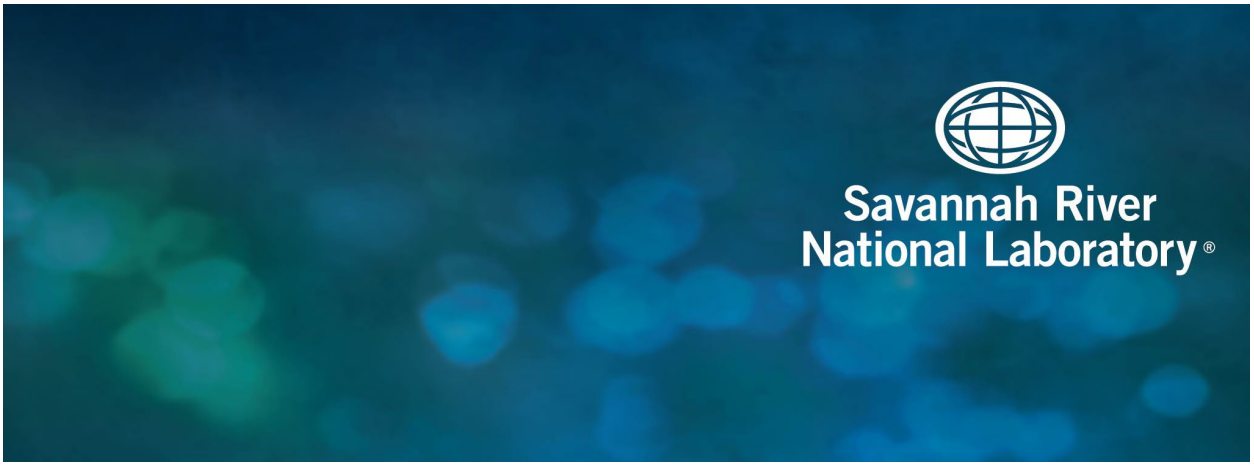


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# Sludge Batch 10 (SB10) Acceptance Evaluation: Radionuclide Concentrations in Tank 51 Washed Qualification Sample

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January 2022

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# EXECUTIVE SUMMARY

Savannah River National Laboratory (SRNL) has been tasked with the radionuclide characterization of the washed Sludge Batch 10 (SB10) qualification sample. The washed SB10 qualification sample is based on SRR Engineering guidance and the sample slurry is expected to be similar in composition to Tank 51 slurry after final preparations for transfer to Tank 40.

Forty-four radionuclides along with total alpha and beta activity have been reported herein. These radionuclide measurements are required for the Defense Waste Processing Facility (DWPF) Radiological Evaluation Program, DWPF Technical Safety Requirements (TSR)/Waste Acceptance Criteria (WAC) Evaluation, and the DWPF Solid Waste Characterization Program.

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# LIST OF ABBREVIATIONS

| | |
|---|---|
| α-PHA | Alpha Pulse Height Analysis |
| DWPF | Defense Waste Processing Facility |
| ICP-MS | Inductively Coupled Plasma Mass Spectrometry |
| LSC | Liquid Scintillation Counting |
| MDL | Method Detection Limit |
| RSD | Relative Standard Deviation |
| n | Sample size |
| SRNL | Savannah River National Laboratory |
| SRR | Savannah River Remediation |
| SaM | Sensing and Metrology |
| SB10 | Sludge Batch 10 |
| SRE | Sodium Reactor Experiment |
| TTQAP | Task Technical and Quality Assurance Plan |
| TSR | Technical Safety Requirements |
| TTR | Technical Task Request |
| WAC | Waste Acceptance Criteria |

## 1.0 Introduction

Savannah River National Laboratory (SRNL) has been tasked with the radionuclide characterization of the washed Sludge Batch 10 (SB10) qualification sample. The as-received SB10 qualification samples from Tank 51, HTF-51-19-114 and HTF-51-20-15, were taken prior to Sodium Reactor Experiment (SRE) additions to Tank 51. The samples were combined and initially characterized by SRNL prior to washing.[1] Material from H Canyon Tanks 16.3 and 16.4 were added during washing. The SRNL-washed, based on Savannah River Remediation (SRR) Engineering guidance, SB10 qualification sample is expected to be similar in composition to Tank 51 slurry after final preparations for transfer to Tank 40.

The radionuclide characterization makes up part of the tasks requested in the Technical Task Request (TTR) and is governed by the Task Technical and Quality Assurance Plan (TTQAP).[2,3] The chemical characterization of the SB10 washed qualification sample will be documented in a separate report. Table 1-1 lists the 36 radionuclides requested in the TTR to be reported for the SRNL-washed SB10 qualification sample. These radionuclides are needed for the Defense Waste Processing Facility (DWPF) Radiological Evaluation Program, DWPF Technical Safety Requirements (TSR)/Waste Acceptance Criteria (WAC) Evaluation, and the DWPF Solid Waste Characterization Program.

**Table 1-1. Requested Radionuclides to Report**

| Radionuclides | | | |
|---|---|---|---|
| H-3 | Te-125m | Sm-151 | Np-237 |
| Co-60 | I-129 | Eu-152 | Pu-238 |
| Sr-90 | Cs-134 | Eu-154 | Pu-239 |
| Y-90 | Cs-137 | Eu-155 | Pu-240 |
| Tc-99 | Ba-137m | U-233 | Pu-241 |
| Ru-106 | Ce-144 | U-234 | Am-241 |
| Rh-106 | Pr-144 | U-235 | Am-242m |
| Ag-110m | Pr-144m | U-236 | Cm-244 |
| Sb-125 | Pm-147 | U-238 | Cm-245 |

## 2.0 Experimental Procedure

### 2.1 Methods

The SB10 qualification sample was washed and decanted to mimic sample washing in Tank 51. An aliquot was then taken and used for radionuclide and other measurements. A detailed description of the washing and SRE addition will be provided in the final SB10 qualification report. Table 2-1 gives the weight percent (wt%) solids and density of the washed SB10 sample. Slurry and supernatant aliquots were dried at 110°C until a constant weight was obtained for wt% total dried solids and wt% dissolved solids. For wt% calcined solids, dried slurry samples were heated to 1,100°C and then cooled and weighed. Wt% insoluble solids and soluble solids were calculated from the total dried solids and dissolved solids measurements. Densities were obtained gravimetrically from sample weights in vessels of known volume.

**Table 2-1. SB10 Washed Sample Weight Percent and Density**

| | Value (% RSD) |
|---|---|
| **Supernatant Density (g/mL)** | 1.06 (0.4) |
| **Slurry Density (g/mL)** | 1.10 (1) |
| **Wt% Total Dried Solids** Slurry Basis | 15.0 (0.8) |
| **Wt% Dissolved Solids** Supernatant Basis | 7.4 (3) |
| **Wt% Calcined Solids** Slurry Basis | 11.3 (0.1) |
| **Wt% Insoluble Solids** Slurry Basis | 8.2 (N/A) |
| **Wt% Soluble Solids** Slurry Basis | 6.8 (N/A) |

N/A = not applicable as result is calculated

A subsample (~0.25g) of the SB10 washed sample was digested by peroxide fusion and aqua regia in quadruplicate and transferred to SRNL Sensing and Metrology (SaM) for radionuclide characterization. For the peroxide fusion digestion, this involved the drying and fusing of slurry aliquots with sodium peroxide at 675°C and then dissolving with nitric acid and water. The resulting liquids were then diluted to 100 mL with water. The aqua regia digestions were performed by mixing aliquots of slurry with aqua regia and placing in closed vessels that were then heated at ~110°C for several hours before diluting to 100 mL with water.

The methods Am/Cm and I-129 required specific sample preparation in the SRNL shielded cells prior to being sent to SaM; their detailed preparations have been previously described.[4] From the aqua regia digestion, Tc-99, U-233, U-234, U-235, U-236, U-238, Np-237, Pu-239, and Pu-240 were measured by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) and tritium was measured using Liquid Scintillation Counting (LSC). All other radionuclides were determined from the digested sample from the peroxide fusion. Gamma counting was used to determine the Cs-137 and Cs-134 concentrations. Cs-removed gamma counting was used to determine Co-60, Ru-106, Ag-110m, Sb-125, Ce-144, Eu-152, Eu-154, Eu-155, and Am-241 concentrations. Sr-90, Pm-147/Sm-151, and Pu-238/Pu-241 methods have been described previously.[4] The concentration of the beta emitters Sr-90, Pm-147/Sm-151, and Pu-241 were measured using LSC, while Pu-238 was determined by alpha pulse height analysis (α-PHA). The total alpha and beta counts were determined using LSC.

The radionuclide concentrations given in this report are the average of four replicates. Radionuclide measurements with a "<" (less than symbol) indicates that all four replicates were below the method detection limit (MDL). Only the relative standard deviation (RSD) values of radionuclide measurements in which all four replicates were above the MDL are listed.

## 2.2 Quality Assurance

This work was performed under a TTR.[2] The analysis herein satisfies the Task 1 activity for the radionuclide characterization of the washed sample of the TTQAP associated with this TTR.[3] The TTR identifies the Functional Classification as Safety Class. Thus, this document was reviewed by Design Verification by Document Review. The requirements for performing reviews of technical reports and the extent of review are established in manual E7 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.

## 3.0 Results and Discussion

Table 3-1 lists the measured radionuclide values of the washed SB10 qualification sample. The results reported in units of wt% of total dried solids (column two) and microcuries per gram (μCi/g) of total dried solids (column three) were determined using the 15.0 wt% total dried solids of the slurry given in Table 2-1. Additionally, the specific activities of each individual radionuclide was used for the conversion between the two.[5] The curies per gallon (Ci/gal) of sludge slurry (column five) is calculated using the slurry density of 1.1 g/mL given in Table 2-1. Th-232, Am-243, Cm-242, Cm-246, Cm-247, Cm-248, and Cf-250 concentrations were above their respective MDLs and are reported in addition to the thirty-six radionuclides requested in the TTR. Y-90, Rh-106, Pr-144, and Pr-144m are in secular equilibrium with 100% of their parent radionuclides, and as such, have the same activities. Ba-137m is in secular equilibrium with 94.6% of Cs-137, its parent radionuclide. Te-125m is in secular equilibrium with Sb-125, its parent radionuclide, at 22.9% of its activity.[6] However, it is treated in Table 3-1 as being 100% of the activity as a worst-case scenario, due to the Sb-125 measurement being below its MDL.

Table 3-2 gives the fissile isotope results for each of the four sample aliquots of the SB10 washed qualification sample in μCi/g of total solids.

**Table 3-1. Average Concentrations of Radionuclides in the Tank 51 SB10 Washed Qualification Sample**

| Radionuclide | Wt% in Total Dried Solids | µCi/g in Total Dried Solids | %RSD (n=4) | Ci/gal in Sludge Slurry | Method |
|---|---|---|---|---|---|
| H-3 | N/A* | N/A* | N/A* | 3.89E-05 | Tritium (LSC) |
| Co-60 | 4.12E-09 | 4.66E-02 | 7.8 | 2.91E-05 | Cs-Removed Gamma Counting |
| Sr-90 | 5.27E-03 | 7.18E+03 | 15 | 4.49E+00 | SR90 (LSC) |
| Y-90 | 1.32E-06 | 7.18E+03 | 15 | 4.49E+00 | Calculated (Secular Equilibrium w/Y-90) |
| Tc-99 | 1.90E-03 | 3.22E-01 | 3.3 | 2.01E-04 | ICP-MS |
| Ru-106 | <4.02E-09 | <1.35E-01 | N/A | <8.41E-05 | Cs-Removed Gamma Counting |
| Rh-106 | <3.78E-15 | <1.35E-01 | N/A | <8.41E-05 | Calculated (Secular Equilibrium w/Ru-106) |
| Ag-110m | <5.13E-10 | <2.44E-02 | N/A | <1.52E-05 | Cs-Removed Gamma Counting |
| Sb-125 | <8.70E-09 | <8.98E-02 | N/A | <5.61E-05 | Cs-Removed Gamma Counting |
| Te-125m | <4.98E-10 | <8.98E-02 | N/A | <5.61E-05 | Calculated (Secular Equilibrium w/Sb-125) |
| I-129 | 5.10E-05 | 9.00E-05 | 28 | 5.62E-08 | I-129 with Separation |
| Cs-134 | <1.53E-07 | <1.98E+00 | N/A | <1.24E-03 | Gamma Counting |
| Cs-137 | 2.81E-04 | 2.44E+02 | 1.5 | 1.52E-01 | Gamma Counting |
| Ba-137m | 4.29E-11 | 2.31E+02 | 1.5 | 1.44E-01 | Calculated using 0.946 x Cs-137 activity |
| Ce-144 | <1.47E-08 | <4.68E-01 | N/A | <2.92E-04 | Cs-Removed Gamma Counting |
| Pr-144 | <6.20E-13 | <4.68E-01 | N/A | <2.92E-04 | Calculated (Secular Equilibrium w/Ce-144) |
| Pr-144m | <2.58E-13 | <4.68E-01 | N/A | <2.92E-04 | Calculated (Secular Equilibrium w/Ce-144) |
| Pm-147 | <5.18E-06 | <4.80E+01 | N/A | <3.00E-02 | Pm-147/SM-151 (LSC) |
| Sm-151 | 2.09E-04 | 5.50E+01 | 5.0 | 3.43E-02 | Pm-147/SM-151 (LSC) |
| Eu-152 | <2.53E-08 | <4.37E-02 | N/A | <2.73E-05 | Cs-Removed Gamma Counting |
| Eu-154 | 1.76E-06 | 4.74E+00 | 10 | 2.96E-03 | Cs-Removed Gamma Counting |
| Eu-155 | <6.85E-08 | <3.19E-01 | N/A | <1.99E-04 | Cs-Removed Gamma Counting |
| Th-232 | 1.75E+00 | 1.92E-03 | 2.9 | 1.20E-06 | ICP-MS |
| U-233 | 5.10E-04 | 4.94E-02 | 3.5 | 3.09E-05 | ICP-MS |
| U-234 | 5.93E-04 | 3.70E-02 | 1.1 | 2.31E-05 | ICP-MS |
| U-235 | 2.93E-02 | 6.34E-04 | 3.7 | 3.96E-07 | ICP-MS |
| U-236 | 1.96E-03 | 1.27E-03 | 4.4 | 7.92E-07 | ICP-MS |
| U-238 | 2.43E+00 | 8.16E-03 | 2.9 | 5.10E-06 | ICP-MS |
| Np-237 | 1.11E-03 | 7.82E-03 | 4.2 | 4.89E-06 | ICP-MS |
| Pu-238 | 3.76E-04 | 6.43E+01 | 6.8 | 4.02E-02 | PU238/PU241 (α-PHA) |
| Pu-239 | 1.44E-02 | 8.98E+00 | 3.8 | 5.61E-03 | ICP-MS |
| Pu-240 | 1.15E-03 | 2.63E+00 | 3.6 | 1.64E-03 | ICP-MS |
| Pu-241 | 3.00E-05 | 3.09E+01 | 16 | 1.93E-02 | PU238/PU241 (LSC) |
| Am-241 | 2.83E-04 | 9.72E+00 | 6.0 | 6.07E-03 | Cs-Removed Gamma Counting |
| Am-243 | 9.05E-07 | 1.80E-03 | 4.4 | 1.13E-06 | Am/Cm |
| Am-242m | 1.52E-09 | 1.48E-04 | 11 | 9.25E-08 | Am/Cm |
| Cm-242 | 3.71E-12 | 1.23E-04 | 15 | 7.66E-08 | Am/Cm |
| Cm-243 | 2.38E-09 | 1.23E-03 | 9.4 | 7.66E-07 | Am/Cm |
| Cm-244 | 4.47E-08 | 3.62E-02 | 27 | 2.26E-05 | Am/Cm |
| Cm-245 | 5.27E-09 | 9.05E-06 | 16 | 5.66E-09 | Am/Cm |
| Cm-246 | 6.83E-09 | 2.10E-05 | 16 | 1.31E-08 | Am/Cm |
| Cm-247 | 2.25E-09 | 2.09E-09 | 31 | 1.31E-12 | Am/Cm |
| Cm-248 | 8.11E-08 | 3.45E-06 | 74 | 2.15E-09 | Am/Cm |
| Cf-250 | 3.45E-12 | 3.80E-06 | 25 | 2.37E-09 | Am/Cm |
| Total alpha | N/A | 1.08E+02 | 21 | 6.72E-02 | LSC |
| Total beta | N/A | 1.35E+04 | 1.1 | 8.42E+00 | LSC |
| Total gamma† | N/A | 2.49E+02 | N/A | 1.53E-01 | Calculated |
| Total beta-gamma‡ | N/A | 1.37E+04 | N/A | 8.57E+00 | Calculated |

N/A = not applicable

*Drying the slurry sample would drive off the Tritium (H-3) due to it being mainly present as HTO. The concentration was measured in the slurry with a value of 9.34E-03 µCi/g and converted to Ci/gal using the slurry density. Only the one measurement of four replicates that was above the MDL for H-3 is reported in the table.

† Total activity of reported gamma emitters: Co-60, Ru-106, Rh-106, Sb-125, Te-125m, Cs-134, Ba-137m, Ce-144, Pr-144, Eu-152, Eu-154, Eu-155, and Am-241. The MDL value was used in the calculations for radionuclides that had concentrations below the MDL.

‡ Total activity from beta and gamma.

**Table 3-2. Replicate Activities of Fissile Radionuclides for the SB10 Washed Qualification Sample in µCi/g of Dried Total Solids**

| Radionuclide | Repl. 1 | Repl. 2 | Repl. 3 | Repl. 4 | Reported Average | %RSD |
|---|---|---|---|---|---|---|
| U-233 | 5.11E-02 | 4.89E-02 | 5.04E-02 | 4.72E-02 | 4.94E-02 | 3.48% |
| U-235 | 6.57E-04 | 6.19E-04 | 6.49E-04 | 6.09E-04 | 6.34E-04 | 3.67% |
| Pu-239 | 9.40E+00 | 8.83E+00 | 9.06E+00 | 8.61E+00 | 8.98E+00 | 3.76% |
| Pu-241 | 3.72E+01 | 3.18E+01 | 2.88E+01 | 2.59E+01 | 3.09E+01 | 15.66% |

## 4.0 Conclusions

The results for forty-four radionuclides along with total alpha and beta activity have been reported herein. These radionuclide measurements are required for the DWPF Radiological Evaluation Program, DWPF TSR/WAC Evaluation, the DWPF Solid Waste Characterization Program and the Tank 51 flammability calculations.

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