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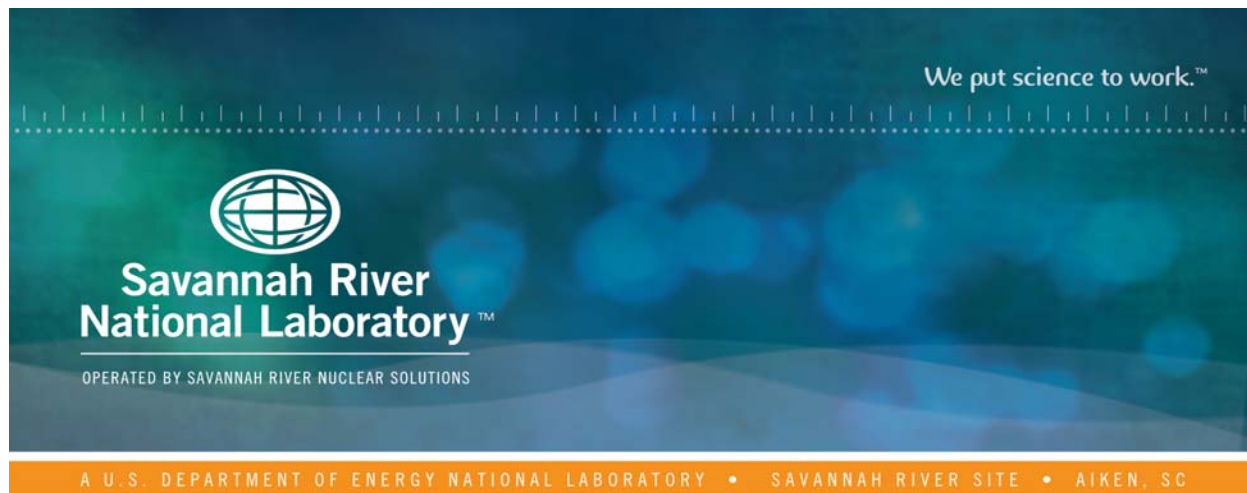
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Sludge Batch 9 (SB9) Acceptance Evaluation: Radionuclide Concentrations in Tank 51 SB9 Qualification Sample Prepared at SRNL

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March 2016

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PREFACE OR ACKNOWLEDGEMENTS

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EXECUTIVE SUMMARY

Presented in this report are radionuclide concentrations required as part of the program of qualifying Sludge Batch 9 (SB9) for processing in the Defense Waste Processing Facility (DWPF). The SB9 material is currently in Tank 51 and has been washed and prepared for transfer to Tank 40. The acceptance evaluation needs to be completed prior to the transfer of the material in Tank 51 to Tank 40. The sludge slurry in Tank 40 has already been qualified for DWPF processing and is currently being processed as Sludge Batch 8 (SB8).ⁱ The radionuclide concentrations were measured or estimated in the Tank 51 SB9 Washed Qualification Sample prepared at Savannah River National Laboratory (SRNL). This sample was prepared from a three liter sample of Tank 51 sludge slurry (HTF-51-15-81) taken on July 23, 2015. The sample was delivered to SRNL where it was initially characterized in the Shielded Cells.ⁱⁱ Under the direction of Savannah River Remediation (SRR) it was then adjusted per the Tank Farm washing strategy as of October 20, 2015.ⁱⁱⁱ This final slurry now has a composition^{iv} expected to be similar to that of the slurry in Tank 51 after final preparations have been made for transfer of that slurry to Tank 40.

Determining the radionuclide concentrations in this Tank 51 SB9 Qualification Sample is part of the work requested in Technical Task Request (TTR) No. U-TTR-S-00009, Rev. 1.^v The work with this qualification sample is covered by a Task Technical and Quality Assurance Plan (TTQAP).^{vi} The radionuclides included in this report are needed for the DWPF Radiological Program Evaluation, the DWPF Waste Acceptance Criteria (WAC) Evaluation, and the DWPF Solid Waste Characterization Program (TTR Task I.B.1). The sample is the same as that on which the chemical composition was reported.^{iv} Radionuclides required to meet the Waste Acceptance Product Specifications (WAPS) (TTR Task III.2) will be measured at a later date after the slurry from Tank 51 has been transferred to Tank 40. Then a sample of the as-processed SB9 will be taken from Tank 40 and transferred to SRNL for measurement of these radionuclides.

The results presented in this report are those necessary for DWPF to assess if SB9 meets the requirements for the DWPF Radiological Program Evaluation, the DWPF WAC evaluation, and the DWPF Solid Waste Characterization Program. Concentrations are given for thirty-seven radionuclides along with total alpha and beta activity. Values for total gamma and total gamma plus beta activities are also calculated.

Results also indicate that 91% of the Tc-99 that could have been in this sludge batch was removed by chemical processing steps in the SRS Canyons or Tank Farm – it is typical for Tc-99 to partition to the salt phase of the waste. The I-129 found in the sludge was likely retained as a nonvolatile mercury species.

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 - ii. Pareizs, J. M., *Characterization of the As-Received Sludge Batch 9 Qualification Sample (HTF-51-15-81)*, SRNL-STI-2015-00442, Savannah River National Laboratory, Aiken, SC 29808 (2015).
 - iii. Gillam, J. M., *Sludge Batch 9 Washing Spreadsheet: SB9_102015.xlsm*, Savannah River Remediation, Aiken, SC 29808 (2015).
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 - v. Samadi-Dezfouli, A., *Technical Task Request: Sludge Batch 9 Qualification, Confirmatory, and Waste Acceptance Product Specification Samples*, U-TTR-S-00009, Rev. 1, Savannah River Site, Aiken, SC 29808 (2015).
 - vi. Pareizs, J. M., *Task Technical and Quality Assurance Plan for Sludge Batch 9 Qualification, Confirmatory, and Waste Acceptance Product Specification Samples*, SRNL-RP-2015-00120, Rev. 1, Savannah River National Laboratory, Aiken, SC 29808 (2015).

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LIST OF ABBREVIATIONS

AD	Analytical Development
Ci	Curie
DOE	Department of Energy
DWPF	Defense Waste Processing Facility
DSFE	DWPF/Saltstone Facility Engineering
FYSF	Fission Yield Scaling Factor
g	gram
HLW	High Level Waste
HTO	tritiated water
IAEA	International Atomic Energy Agency
ICP-MS	Inductively Coupled Plasma – Mass Spectrometry
L	liter
μCi	micro-Curie
%RSD	Percent Relative Standard Deviation
SB	Sludge Batch
SpA	Specific Activity (Ci/g)
SRNL	Savannah River National Laboratory
SRR	Savannah River Remediation
SRS	Savannah River Site
$t_{1/2}$	half-life
TTQAP	Task Technical and Quality Assurance Plan
TTR	Technical Task Request
WAC	Waste Acceptance Criteria
WAPS	Waste Acceptance Product Specifications
WCS	Waste Characterization System
Wt%	Weight Percent

1.0 Introduction

Presented in this report are radionuclide concentrations required as part of the program of qualifying Sludge Batch 9 (SB9) for processing in the Defense Waste Processing Facility (DWPF). The SB9 material is currently in Tank 51 and has been washed and prepared for transfer to Tank 40. The acceptance evaluation needs to be completed prior to the transfer of the material in Tank 51 to Tank 40. The sludge slurry in Tank 40 has already been qualified for DWPF processing and is currently being processed as Sludge Batch 8 (SB8).¹ The radionuclide concentrations were measured or estimated in the washed Tank 51 SB9 Qualification Sample prepared at Savannah River National Laboratory (SRNL). This sample was prepared from a three liter sample of Tank 51 sludge slurry (HTF-51-15-81) taken on July 23, 2015. The sample was delivered to SRNL where it was initially characterized in the Shielded Cells.² Under the direction of Savannah River Remediation (SRR) it was then modified per the Tank Farm washing strategy as of October 20, 2015.³ This final slurry now has a composition⁴ expected to be similar to that of the slurry in Tank 51 after final preparations have been made for transfer of that slurry to Tank 40.

Determining the radionuclide concentrations in this Tank 51 SB9 Qualification Sample is part of the work requested in Technical Task Request (TTR) No. U-TTR-S-00009, Rev. 1.⁵ The work with this qualification sample is covered by a Task Technical and Quality Assurance Plan (TTQAP).⁶ The radionuclides included in this report are needed for the DWPF Radiological Program Evaluation, the DWPF Waste Acceptance Criteria (WAC) Evaluation, and the DWPF Solid Waste Characterization Program (TTR Task I.B.1). The sample is the same as that on which the chemical composition was reported.⁴ Radionuclides required to meet the Waste Acceptance Product Specifications (TTR Task III.2) will be measured at a later date after the slurry from Tank 51 has been transferred to Tank 40. Then a sample of the as-processed SB9 will be taken from Tank 40 and transferred to SRNL for measurement of these radionuclides.

2.0 Experimental

2.1 Methods

Three wash/decant cycles were conducted at SRNL to mimic the projected washing in Tank 51.³ After the three washes and decants, an aliquot of the 3-L Tank 51 sample was taken for analyses. The ~400 mL aliquot taken for the radionuclide measurements and other characterization was called “SB9 TK 51 Washed Analytical Subsample” and is the SB9 Qualification Sample prepared at SRNL prior to blending with Tank 40 material.

Data presented in this report represents the measured or estimated radionuclide concentrations obtained from several standard analytical methods performed by SRNL Analytical Development (AD) personnel. These methods were performed on solutions resulting from the dissolutions of the slurry samples. The dissolution methods were an alkali fusion⁷ and an aqua regia digestion.⁸ Two additional preparation schemes were performed to obtain the Am/Cm data and the I-129 data: these methods have been described previously.⁹

Table 2-1 presents the weight percent solids and density measurements for the washed qualification sample prepared at SRNL,⁴ both sludge slurry (henceforth referred to as “slurry”) and supernate. Total and insoluble solids are given only on a slurry basis.

Table 2-1. Weight Percent Solids and Density for Washed Tank 51 SB9 Samples

Sample Name	Wt% Total Solids [%RSD]*	Wt% Soluble Solids [%RSD]*	Wt% Insoluble Solids [%RSD]*	Density (g/mL) [%RSD]*
Slurry	19.59 [0.6]	4.98 [N/A]	14.61 [N/A]	1.15 [4.9]
Supernate	NA	5.83 [0.7]	NA	1.04 [1.2]

* RSD \equiv relative standard deviation; %RSD is defined as the standard deviation of an array, divided by the average of the array, expressed in percent terms.

N/A \equiv not applicable as result is calculated

NA \equiv not applicable to supernate sample

The experimental procedures for specific radionuclides are discussed below. The concentration of tritium in the supernate of the sludge slurry was not determined by using a sample of the supernate that had been separated from the insoluble solids by filtration, as was done for Sludge Batch (SB6). Instead, the tritium concentration in the slurry was determined from the aqua regia digestion leading to a slightly higher detection limit due to the greater dilution used in the aqua regia method. The H-3 concentration on a Ci/gal basis was calculated based on the density of the slurry.

The concentrations and detection limits reported are generally based on three or four replicate samples, though in a couple instances only a single value above the detection limit was measured. Concentrations of H-3, Sr-90, and Pu-241 along with total beta activity are based on analyses by beta counting techniques. The concentrations of Co-60, Eu-154, Eu-155, and Am-241 were measured by Cs removed gamma counting. The concentration of Cs-137 was measured by gamma counting. The results for Tc-99, U-233, U-234, U-235, U-236, Np-237, U-238, Pu-239, and Pu-240, as well as Cm-245 following separation, were determined by Inductively Coupled Plasma Mass Spectroscopy (ICP-MS) that measures the concentration of these radionuclides based on their masses rather than their radioactivity. These concentrations were converted to $\mu\text{Ci/g}$ using their specific activities¹⁰. The results for Pu-238, Am-242m, and Cm-244 along with the total alpha activity were measured by alpha counting.⁹ The concentrations of the following radionuclides: Ru-106, Rh-106, Sb-125, Te-125m, Cs-134, Ce-144, and Pr-144, are reported as method detection limits due to their concentrations being too low to be detected due to their short half-lives and/or the age of the sludge. The concentration of Cm-245 is also reported as method detection limits due to its low concentrations.

Both U-235 fission products, Pm-147 and Sm-151 were measured. The methodology used to obtain the concentration of the high-energy beta Pm-147 and lower-energy beta Sm-151 has been described previously⁹. A detection limit for the Pm-147 is reported due to the low concentration in the aliquots measured.

The concentration reported for C-14 was calculated by DWPF/Saltstone Facility Engineering (DSFE) from the projected concentrations given in the Waste Characterization System (WCS) database for the C-14 in the washed Tank 51 slurry prior to transferring it to Tank 40. This is the method that was agreed upon in the TTQAP for SB9 qualification.⁶

The radionuclide I-129 is a long-lived beta emitting fission product (half-life ($t_{1/2}$) = 1.6×10^7 years) that is in Savannah River Site (SRS) wastes. Four aliquots of wet sludge slurry were prepared as described previously⁹ in the SRNL Shielded Cells, the Y-90 was allowed to decay for nine days, and the samples moved to AD for further preparation and analysis.

2.2 Quality Assurance

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 presents the measured or estimated concentrations for thirty seven individual radionuclides and the measured values for the total beta and total alpha activity along with the calculated values for total gamma and total beta plus gamma activities. Four radionuclides with measured values above their method detection limit, Sm-151, Eu-152, Th-232, and Cm-242, are included even though they were not explicitly called out in the TTR.⁵ The concentrations of those radionuclides that could not be measured due to their low concentrations have been estimated from minimum detection limits based on the analytical method used. For all the radionuclides except tritium (H-3), the concentrations are based on the total dried solids from dissolution of three or four replicates of the Tank 51 sludge slurry. Column 2 gives the concentration in units of wt% total dried solids; Column 3 gives the concentrations in units of microcuries (μCi) per gram of total solids in the dried sludge slurry; Column 4 presents the relative percent standard deviations (%RSD); and Column 5 presents the number of replicates analyzed. Finally, Column 6 presents the concentrations of the radionuclides in curies (Ci) per gallon of slurry calculated based on the measured weight percent total solids in the slurry (19.59 wt%), the density of the slurry (1.15 g/mL), and a conversion factor of 3785 mL/gal to convert to liquid gallons. Specific radionuclides will now be discussed.

Essentially all of the tritium in the Tank 51 slurry is present as tritiated water (HTO). Consequently, its concentration in the dried solids cannot be determined because the HTO evaporates during the drying of the slurry. The determination was made on aqua regia digestions prepared in sealed Teflon digestion vessels. The lowest detection limit for the supernate measurements was used to specify the concentration of H-3.

The radionuclides Y-90, Rh-106, Te-125m, Ba-137m, and Pr-144 are in secular equilibrium with their respective parent radionuclides. Thus the activities of Y-90, Rh-106, Te-125m, and Pr-144 are equal to that of their parents. Approximately 5.4% of the Cs-137 decays directly to stable Ba-137; thus the activity of Ba-137m is 94.6% of the activity of the Cs-137.

The reported concentration of C-14 was not determined by proportioning the WCS projection to that measured for SB2 as was done prior to Sludge Batch (SB8). Instead, DSFE provided the value based on their calculations and the WCS projection. For comparison, the previous ratio method is based on the measured concentration of C-14 in SB2 of $8.13 \times 10^{-3} \mu\text{Ci/g}$.¹¹ The projected concentration of C-14 in Sludge Batch (SB2) was $1.40 \times 10^{-6} \text{ Ci/gallon}$.¹¹ The WCS projection for the total curies of C-14 in Tank 51 when the slurry is ready to be transferred to Tank 40 is $8.20 \times 10^{-2} \text{ curies}$.^{12,13} The volume projected to be transferred to Tank 40 is $2.60 \times 10^5 \text{ gallons slurry}$.¹² The projected concentration of C-14 is then $3.15 \times 10^{-7} \text{ Ci/gallon slurry}$. Multiplying the ratio of the projected concentrations in Tank 51 to the projected concentrations in SB2 by the measured concentrations in SB2 gives $1.83 \times 10^{-3} \mu\text{Ci/g}$ for C-14, a value about 5x larger than from the new estimation method.

The results for Tc-99 in the SB9 qualification sample are interesting in that they indicate that most of the Tc-99 that could have been in SB9 has been removed by the processing steps in the Canyons of SRS. As shown below, it is estimated that 91% of the Tc-99 that could have been in SB9 has been removed to the salt tanks or volatilized as a result of SRS canyon processing. This estimate is determined by comparing the maximum amount of Tc-99 that can be predicted to be in SB9 with the measured amount. The

measured concentration in the total dried solids of the SB9 slurry was 1.88×10^{-3} wt% as determined by ICP-MS analysis. This concentration times the specific activity (SpA) of Tc-99 in Ci/g gives the activity concentration shown in Table 3-1.

The fission products in SB9 were formed in the SRS reactors by thermal neutron fission of U-235. Based on many experimental studies the relative yields of the fission products are known based on the number of atoms of each fission product isotope formed as a result of fission of 100 atoms of U-235.¹⁴ For a particular sludge batch, values for a fission yield scaling factor (FYSF) can be calculated from each measured concentration of a fission product. The FYSF is simply a proportionality factor that relates the concentration in wt% of an isotope in the dried solids of sludge slurry to its fission yield and mass. For those isotopes that have six important properties, values for the FYSF will be equal for a sludge batch. These properties have been previously discussed.¹⁵ The following equation applies for each fission product.

$$\text{FYSF}_i = \text{wt\%}_i / (\text{FY}_i \times \text{am}_i)$$

Where FYSF_i ≡ the fission yield scaling factor based on isotope
 wt%_i ≡ the weight percent of isotope i in the High Level Waste (HLW) total dried solids
 FY_i ≡ the fission yield of isotope i
 am_i ≡ the atomic mass of isotope i

In SB9 there are 11 fission products that have the six important properties. These isotopes are: Ru-101, Ru-102, Ru-103, La-139, Pr-141, Nd-143 through Nd-146, Sm-147, and Sm-148. All these isotopes were measured by ICP-MS in the total dried solids of the SB9 slurry. The average FYSF for SB9 calculated with these 11 isotopes is 3.35×10^{-5} with a %RSD of 11. After rearrangement of the above equation, the concentration of any fission product can be predicted. For Tc-99 with its half-life of 2.13×10^5 years and fission yield of 6.1%, the predicted wt% for Tc-99 is 2.02×10^{-2} wt%. The measured value is 1.88×10^{-3} wt%, indicating that only 9.3% of the Tc-99 that could have been in this sludge batch was actually still in the sludge slurry. One of the main properties that a fission product must have to be retained in the sludge solids is that it is insoluble in caustic. If it were soluble in caustic it would not precipitate in the sludge solids and would be decanted or washed to the salt tanks in the Tank Farm. Technetium is known to form a pertechnetate anion that is soluble in caustic.¹⁵ This can explain the low concentration of Tc-99 in SB9 and was also previously observed for Sludge Batch (SB3)¹⁵ and SB6¹⁶.

The radionuclide I-129 is a U-235 fission product with a long half-life of 1.6×10^7 years. It was first measured successfully in the SB4 Waste Acceptance Product Specifications (WAPS) studies⁹ and the Sludge Batch (SB5) Acceptance Evaluation.¹⁷ Earlier reported concentrations for sludge batches are based on the WCS database. For I-129 the average measured from four aliquots of the sludge slurry was 5.31×10^{-3} μCi/g of total dried solids or 3.01×10^{-3} wt%. Based on the separations and counting results, this appears to be a reliable result. However, it was further checked by calculating the amount of I-129 that could be present using the FYSF. If this predicted concentration were less than the measured concentration, this would indicate that the measured number was erroneously high. Using the FYSF of 3.35×10^{-5} and the fission yield for I-129 of 0.54%,¹⁴ the predicted concentration of I-129 in SB9 would be 2.3×10^{-3} wt%, assuming no I-129 were lost in processing. The measured value is about 30% higher. If the International Atomic Energy Agency (IAEA) fission yield for I-129 of 0.706% is used,¹⁸ the predicted concentration of I-129 in SB9 would be 3.05×10^{-3} wt%, which is what was measured. Iodine is expected to be retained in HM sludges due to the mercuric nitrate catalyst, which results in nonvolatile mercury species.¹⁹

Table 3-1. Concentrations of Radionuclides in the Tank 51 SB9 Qualification Sample

Radionuclide	Wt% in Total Dried Solids	μCi/g in Total Dried Solids	%RSD	Replicates	Ci/gal in Sludge Slurry (a)
H-3	(b)	(b)	N/A	N/A	<3.5E-05
C-14 (c)	8.64E-09	3.85E-04	N/A	N/A	3.28E-07
Co-60	2.85E-08	3.23E-01	1.1	3	2.75E-04
Sr-90	3.34E-03	4.56E+03	8.0	3	3.89E+00
Y-90	8.38E-07	4.56E+03	8.0	3	3.89E+00
Tc-99	1.88E-03	3.19E-01	1.6	3	2.72E-04
Ru-106	<3.7E-09	<1.2E-01	N/A	N/A	<1.1E-04
Rh-106	<3.5E-15	<1.2E-01	N/A	N/A	<1.1E-04
Sb-125	<4.9E-09	<5.1E-02	N/A	N/A	<4.3E-05
Te-125m	<2.8E-10	<5.1E-02	N/A	N/A	<4.3E-05
I-129	3.01E-03	5.31E-03	8.3	4	4.52E-06
Cs-134	<2.4E-08	<3.1E-01	N/A	N/A	<2.6E-04
Cs-137	1.30E-03	1.13E+03	2.9	3	9.64E-01
Ba-137m	1.99E-10	1.07E+03	2.9	3	9.12E-01
Ce-144	<1.2E-08	<3.7E-01	N/A	N/A	<3.1E-04
Pr-144	<4.8E-13	<3.7E-01	N/A	N/A	<3.1E-04
Pm-147	<6.4E-06	<5.9E+01	N/A	N/A	<5.1E-02
Sm-151	3.29E-04	8.65E+01	5.1	3	7.37E-02
Eu-152	5.09E-08	8.81E-02	12	3	7.51E-05
Eu-154	1.79E-06	4.84E+00	6.5	3	4.13E-03
Eu-155	1.45E-07	6.74E-01	35	3	5.75E-04
Th-232	8.01E-01	8.79E-04	0.73	4	7.50E-07
U-233	7.67E-04	7.43E-02	2.6	4	6.33E-05
U-234	8.57E-04	5.36E-02	3.7	4	4.57E-05
U-235	4.45E-02	9.62E-04	1.3	4	8.20E-07
U-236	2.35E-03	1.52E-03	0.67	4	1.30E-06
Np-237	2.92E-03	2.06E-02	1.2	4	1.75E-05
U-238	3.05E+00	1.02E-02	1.2	4	8.73E-06
Pu-238	7.97E-04	1.36E+02	17	3	1.16E-01
Pu-239	6.78E-03	4.21E+00	1.4	4	3.59E-03
Pu-240	7.03E-04	1.60E+00	1.2	4	1.37E-03
Pu-241	3.02E-05	3.11E+01	18	3	2.65E-02
Am-241	3.39E-04	1.16E+01	4.2	3	9.92E-03
Am-242m	1.97E-05	1.91E+00	N/A	1	1.63E-03
Cm-242	4.79E-08	1.58E+00	N/A	1	1.35E-03
Cm-244	5.02E-06	4.06E+00	10	3	3.46E-03
Cm-245	<3.0E-07	<5.1E-04	N/A	N/A	<4.4E-07
Total alpha	N/A	1.97E+02	1.4	3	1.68E-01
Total beta	N/A	1.18E+04	1.6	3	1.00E+01
Total gamma(d)	N/A	1.09E+03	N/A	N/A	9.28E-01
Total beta-gamma	N/A	1.29E+04	N/A	N/A	1.10E+01

N/A ≡ not applicable.

(a) This was calculated for all the radionuclides except H-3 using the weight percent total solids in the slurry of 19.59 and a slurry density of 1.15 g/mL.

(b) Most of the H-3 in the slurry is present as HTO; thus, drying the slurry sample would drive off most of the H-3. The concentration of H-3 was measured in four slurry samples. The result was <8.04E-03 μCi/g slurry. This was converted to Ci/gal slurry using slurry density.

(c) Calculated value.¹²

(d) The total activity of the 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.

Table 3-2 provides the replicate measurements made for the fissile radionuclides in order for DWPF to ensure the fissile content of the glass in a canister is below the 897 g/m³ DOE requirement.⁵

Table 3-2 Replicate Activities of Fissile Radionuclides for the SB9 Qualification Sample in $\mu\text{Ci/g}$ of Total Dried Solids*

Radionuclide	Repl. 1	Repl. 2	Repl. 3	Repl. 4	Reported	%RSD**
U-233	7.51E-02	7.40E-02	7.63E-02	7.17E-02	7.43E-02	2.6
U-235	9.59E-04	9.59E-04	9.49E-04	9.78E-04	9.62E-04	1.3
Pu-239	4.14E+00	4.24E+00	4.20E+00	4.28E+00	4.21E+00	1.4
Pu-241 [‡]	2.53E+01	3.15E+01	3.66E+01	N/A	3.11E+01	18

* ICP-MS data unless specified otherwise

[‡] Pu-238/-241 method.

**Values in the %RSD column are relative to the true calculated averages of the quantities in the table, while the average values reported have been rounded off to a reasonable number of significant figures.

4.0 Conclusions

The results presented in this report are those necessary for DWPF to assess if SB9 meets the requirements for the DWPF Radiological Program Evaluation, the DWPF WAC evaluation, and the DWPF Solid Waste Characterization Program. Concentrations are given for thirty-seven radionuclides along with total alpha and beta activity. Values for total gamma and total gamma plus beta activities are also calculated.

Results also indicate that 91% of the Tc-99 that could have been in this sludge batch was removed by chemical processing steps in the SRS Canyons or Tank Farm – it is typical for Tc-99 to partition to the salt phase of the waste. The I-129 found in the sludge was likely retained as a nonvolatile mercury species.

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