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Sludge Batch 7 Acceptance Evaluation: Radionuclide Concentrations in Tank 51 SB7 Qualification Sample Prepared at SRNL

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EXECUTIVE SUMMARY

Presented in this report are radionuclide concentrations required as part of the program of qualifying Sludge Batch Seven (SB7) for processing in the Defense Waste Processing Facility (DWPF). The SB7 material is currently in Tank 51 being 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 and is currently being processed as SB6. The radionuclide concentrations were measured or estimated in the Tank 51 SB7 Qualification Sample prepared at Savannah River National Laboratory (SRNL). This sample was prepared from the three liter qualification sample of Tank 51 sludge slurry (HTF-51-10-125) received on September 18, 2010. The sample was delivered to SRNL where it was initially characterized in the Shielded Cells. With consultation from the Liquid Waste Organization, the qualification sample was then modified by several washes and decants, which included addition of Pu from H Canyon and sodium nitrite per the Tank Farm corrosion control program. This final slurry now has a composition 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 SB7 Qualification Sample is part of the work requested in Technical Task Request (TTR) No. HLW-DWPF-TTR-2010-0031. The radionuclides included in this report are needed for the DWPF Radiological Program Evaluation, the DWPF Waste Acceptance Criteria (TSR/WAC) Evaluation, and the DWPF Solid Waste Characterization Program (TTR Task I.2). 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 SB7 will be taken and transferred to SRNL for measurement of these radionuclides.

The results presented in this report are those necessary for DWPF to assess if the Tank 51 SB7 sample prepared at SRNL meets the requirements for the DWPF Radiological Program Evaluation, the DWPF Waste Acceptance Criteria evaluation, and the DWPF Solid Waste Characterization Program. Concentrations are given for thirty-four radionuclides along with total alpha and beta activity. Values for total gamma and total gamma plus beta activities are also calculated.

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LIST OF ABBREVIATIONS

AD	Analytical Development
DWPF	Defense Waste Processing Facility
ICP-MS	Inductively Coupled Plasma Mass Spectroscopy
SB2	Sludge Batch 2
SB6	Sludge Batch 6
SB7	Sludge Batch 7
SRNL	Savannah River National Laboratory
WAC	Waste Acceptance Criteria
TTR	Technical Task Request
WAPS	Waste Acceptance Product Specifications
WCS	Waste Characterization System

1.0 Introduction

Presented in this report are radionuclide concentrations required as part of the program to qualify Sludge Batch 7 (SB7) for processing in the Defense Waste Processing Facility (DWPF). The SB7 material is currently in Tank 51 being 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 and is currently being processed as SB6. The radionuclide concentrations were measured or estimated in the Tank 51 SB7 Qualification Sample prepared at Savannah River National Laboratory (SRNL). This sample was prepared from the three liter sample of Tank 51 sludge slurry (HTF-51-10-125) received by SRNL on September 18, 2010. The sample was delivered to SRNL where it was initially characterized in the Shielded Cells.[1] With consultation of the Liquid Waste Organization, the sample was modified by several washes and decants which included an addition of Pu from Canyon Tank 16.3, and an addition of $NaNO_2$. This final slurry now has a composition 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.[2]

Determining the radionuclide concentrations in this Tank 51 SB7 Qualification Sample is part of the work requested in Technical Task Request (TTR) No. HLW-DWPF-TTR-2010-0031.[3] The work with this qualification sample is covered by a Task Technical and Quality Assurance Plan[4] and an Analytical Study Plan.[5] The radionuclides included in this report are needed for the DWPF Radiological Program Evaluation, the DWPF Waste Acceptance Criteria (TSR/WAC) Evaluation, and the DWPF Solid Waste Characterization Program (TTR Task I.2). The sample is the same as that on which the chemical composition was reported.[2] 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 SB7 will be taken and transferred to SRNL for measurement of these radionuclides.

2.0 Experimental Procedure

Data presented in this report represent the measured or estimated radionuclide concentrations of the SRNL-washed Tank 51 qualification sample. The concentrations were obtained from several standard analytical methods performed by Analytical Development (AD) personnel within SRNL. Results provided by AD were converted to the requested units using previously published characterization results[2] (see Table 2-1) and specific activities published in Reference 6).

Table 2-1. Weight Percent Solids and Density for SRNL-Washed Tank 51 SB7 Samples

Property	Result
Wt% Total Solids	18.1
Wt% Soluble Solids	5.8
Wt% Insoluble Solids	12.3
Slurry Density (g/mL)	1.15
Supernate Density (g/mL)	1.05

Several sample preparations were conducted in the SRNL Shielded Cells prior to submission to AD for analysis. Sample preparation methods and AD analyses are briefly described below.

Supernate samples were obtained by filtering the slurry. The supernate was diluted with 1 M nitric acid and submitted to AD for beta counting to obtain H-3. The AD result was then converted from a supernate basis to a gallon of slurry basis using insoluble solids and supernate and slurry densities.

Slurry samples were digested by aqua regia. The resulting dissolutions were analyzed by Inductively Coupled Plasma - Mass Spectroscopy (ICP-MS). The results for Tc-99, U-233, U-234, U-235, Np-237, U-238, Pu-239, and Pu-240 were determined from this preparation/analysis.

Slurry samples were dried and digested by alkali fusion with nitric acid uptake. The resulting dissolution solutions were then submitted for several analyses. 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. Concentrations of Sr-90, Pu-241, and total beta activity are based on beta counting. The results for Pu-238 and the total alpha activity were measured by alpha counting. The concentrations of radionuclides Ru-106, Rh-106, Cs-134, Ce-144, and Pr-144 are reported as method detection limits due to their concentrations being too low to be detected because of their short half-lives and the age of the sludge. These limits were determined by Cs removed gamma counting.

The concentration reported for C-14 was provided by Waste Solidification Engineering.[7] The concentration was obtained from the Tank Farm's Waste Characterization System (WCS).

Two additional preparation schemes were performed to obtain I-129, Am-242m, Am-243, Cm-244, and Cm-245. These methods are described in Reference 8.

3.0 Results and Discussion

Table 3-1 presents the measured or estimated concentrations for thirty five individual radionuclides, 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. 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 dissolution of the total dried solids from the SRNL-washed Tank 51 sludge slurry. Column 2 gives the concentrations in units of Curies (Ci) per gram of total dried solids in the sludge slurry. Column 3 presents the concentrations of the radionuclides in curies (Ci) per gallon of slurry calculated from on the measured weight percent total dried solids in the slurry (18.1 wt%), the density of the slurry (1.15 g/mL), and a conversion factor of 3785 mL/gal to convert to liquid gallons. Column 4 presents the concentration of the radionuclides as a weight percent (wt%) of total dried solids. Column 5 provides the percent relative standard deviation (%RSD) and the number of replicates analyzed (n), giving an indication of method precision. 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 could not be determined because the HTO was evaporated during the drying of the slurry. 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% of the Cs-137 decays directly to stable Ba-137; thus the activity of Ba-137m is 94.7% of the activity of the Cs-137.[9]

Table 3-1. Concentrations of Radionuclides in the SRNL-Washed Tank 51 SB7 Qualification Sample

Radionuclide	Ci/g of Dried Solids	Ci/gal of Sludge Slurry (a)	Wt % of Dried Solids	%RSD, n
H-3	(b)	<1.9E-05	(b)	NA, 4
C-14 (d)	1.27E-09	1.00E-06	2.85E-08	NA
Co-60	3.30E-06	2.60E-03	2.92E-07	6%, 4
Sr-90	1.31E-02	1.03E+01	9.60E-03	3%, 4
Y-90	1.31E-02	1.03E+01	2.41E-06	3%, 4
Tc-99	1.36E-07	1.07E-04	8.02E-04	19%, 4
Ru-106	<6E-07	<5E-04	<2E-08	NA, 4
Rh-106	<6E-07	<5E-04	<2E-14	NA, 4
Sb-125	<2E-07	<2E-04	<2E-08	NA, 4
Te-125m	<2E-07	<2E-04	<1E-09	NA, 4
I-129	1.19E-09	9.41E-07	6.76E-04	11%, 2
Cs-134	<5E-06	<4E-03	<3E-07	NA, 4
Cs-137	7.62E-04	6.01E-01	8.77E-04	3%, 4
Ba-137m	7.22E-04	5.69E-01	1.34E-10	3%, 4
Ce-144	<9E-07	<7E-04	<3E-08	NA, 4
Pr-144	<9E-07	<7E-04	<1E-12	NA, 4
Pm-147	<2E-04	<1E-01	<2E-05	NA, 4
Eu-154	1.34E-05	1.06E-02	4.97E-06	4%, 4
Eu-155	2.90E-06	2.28E-03	6.23E-07	2%, 3
Th-232	9.93E-10	7.82E-07	9.03E-01	2%, 4
U-233	5.47E-08	4.31E-05	5.65E-04	23%, 4
U-234	4.27E-08	3.36E-05	6.83E-04	15%, 3
U-235	6.46E-10	5.09E-07	2.99E-02	3%, 4
U-236	9.20E-10	7.24E-07	1.42E-03	27%, 4
Np-237	1.82E-08	1.43E-05	2.58E-03	8%, 4
U-238	1.71E-08	1.35E-05	5.09E+00	3%, 4
Pu-238	9.73E-05	7.67E-02	5.68E-04	4%, 4
Pu-239	1.05E-05	8.27E-03	1.71E-02	3%, 4
Pu-240	3.68E-06	2.90E-03	1.61E-03	10%, 4
Pu-241	<5E-05	<4E-02	<5E-05	NA, 4
Am-241	3.41E-05	2.69E-02	9.94E-04	5%, 4
Pu-242	<2E-08	<1E-05	<5E-04	NA, 4
Am-242m	2.83E-08	2.23E-05	2.91E-07	32%, 4
Am-243	4.24E-07	3.34E-04	2.13E-04	5%, 4
Cm-244	1.41E-05	1.11E-02	1.74E-05	4%, 4
Cm-245	1.83E-09	1.44E-06	1.07E-06	5%, 4
Total alpha	<7E-04	<6E-01	-	NA, 4
Total beta	3.16E-02	2.49E+01	-	4%, 4
Total Gamma (c)	7.76E-04	6.11E-01	-	NA
Total beta-gamma (c)	3.24E-02	2.55E+01	-	NA

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 18.1 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 leaving virtually no H-3 in the dried solids. The concentration of H-3 was measured in four samples of the supernate of the Tank 51 SB7 sample slurry and converted to Ci/gal.

(c) This is the total activity of the gamma emitters: Co-60, Ba-137m, Eu-154, Eu-155, and Am-241. The Total beta-gamma is the sum of the total beta and total gamma values in the table.

(d) The C-14 concentrations were calculated from a value obtained from the Tank Farm's WCS.

The measured total alpha and total beta values compare reasonably well with the sum of the alpha and beta emitters provided in the table. The total gamma is the sum of: Co-60, Ba-137m, Eu-154, Eu-155, and Am-241 values in the table. The total beta-gamma is the sum of the total beta and total gamma values in the table.

4.0 Conclusions

The results presented in this report are those necessary for DWPF to assess whether the Tank 51 SB7 qualification sample prepared at SRNL meets the requirements for the DWPF Radiological Program Evaluation, the DWPF Waste Acceptance Criteria evaluation, and the DWPF Solid Waste Characterization Program. Concentrations are given for thirty-five radionuclides along with total alpha and beta activity. Values for total gamma and total gamma plus beta activities are also calculated.

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