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Tank 9H Salt Solution Supernatant Characterization in Support of Potential Operating Strategies of Tank 9H Supernatant Through the Tank Closure Cesium Removal System

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Savannah River National Laboratory®

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EXECUTIVE SUMMARY

Savannah River Remediation (SRR) recently initiated an ion exchange process to remove radioactive cesium from dissolved salt solution in Tank 10H, known as Tank Closure Cesium Removal (TCCR). As part of that ongoing campaign, the salt in Tank 9H will be dissolved and transferred to Tank 10H for processing through the TCCR system, and SRR requested the characterization of the dissolved salt solution in Tank 9H prior to the transfer to Tank 10H.

On July 18, 2019, Savannah River Remediation Engineering (SRR-E) delivered two variable depth Tank 9H salt solution samples, identified as samples HTF-9-19-74 and HTF-9-19-75 (combined total sample volume of about 400 mL) to the Savannah River National Laboratory (SRNL) for characterization. These two Tank 9H variable depth samples (HTF-9-19-74 and HTF-9-19-75 taken at ~180" and 42", respectively) were retrieved from Riser 7 after hydro-lancing activities were complete. Approximately 4200 gallons of water were added to the tank during this activity.

This report presents the analytical characterization results for the Tank 9H salt solution variable depth samples HTF-9-19-74 and HTF-9-19-75 and contains mainly the filtrate analytical results for anions, elements, mass spectral data, total cesium characterization results and select radionuclide results, along with the X-ray diffraction characterization of the solid fractions from the liquid /solid separation of the variable depth salt solutions. Most of these analyses were performed in duplicate with the percent relative standard deviation (%RSD), a measure of the variability between each duplicate data set expressed as a percentage, used to estimate the analytical result quality for each analyte.

The average sodium concentrations in these two variable depth filtrates [9.73 (0.45 %RSD) M for sample HTF-9-19-74 and 9.50 (0.68 %RSD) M for sample HTF-9-19-75] are abnormally high when compared to the average sodium concentration in the Tank Farm supernates which average 5.6 M. These high sodium concentrations in the filtrate account for the high density of the filtrate solutions; densities of these two variable depth Tank 9H filtrate samples are 1.436 ± 0.002 for sample HTF-9-19-74 and 1.433 ± 0.003 g/mL for sample HTF-9-19-75. On the other hand, the total mercury concentrations in these two filtrate samples, which averaged less than 3 mg/L, is in line with expectations.

A liquid/solid separation, designed to generate the filtrate solutions used for the characterizations, also produced off-white colored solid materials. The solid fractions from the solid/liquid separations were found to readily dissolved (based on visual observations) in deionized and distilled (DI) water with simple mechanical agitation of the water/solid mixtures. The resulting two solid crystalline fractions from the variable depth samples contain the same four minerals identified by X-ray diffraction: Gibbsite ($Al(OH)_3$), Nitratine ($NaNO_3$), Sodium nitrite ($NaNO_2$) and Quartz (SiO_2). The laboratory measured water-to-solids fraction mixtures (phase ratios) required for dissolution of these Tank 9H variable depth sample solid fractions were 4.13 mL/gram of sample HTF 9-19-74 solid fraction and 4.11 mL/gram of sample HTF 9-19-75 solid fraction.

A complete dissolution of the solid phases to account for all the elemental constituents of the solids would require acid digestions using aqua regia or peroxide fusion methods instead of DI-water.

A summary of the average analytical results for the analytes, based on one sigma analytical uncertainty, for the Tank 9H variable depth samples are presented in the data tables within this report.

Overall, there are no significant measurable differences in the concentrations of the analytes in the two Tank 9H variable depth samples. The minor analytical differences fall within the analytical uncertainties of the different analytical methods.

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LIST OF ABBREVIATIONS

AD	Analytical Development
CPT	Chemical Processing Technology
DMA	Direct Mercury Analysis
GC/AFS	gas chromatography/atomic fluorescence spectroscopy
IC	Ion Chromatography
ICP-AES	inductively coupled plasma atomic emission spectroscopy
ICP-MS	inductively coupled plasma mass spectrometry
MDA	Minimum Detection Activity
MDL	Minimum Detection Limit
M	mole/Liter
PI	Principal Investigator
RSD	Relative Standard Deviation
SCO	Shielded Cells Operations
SRNL	Savannah River National Laboratory
SRR	Savannah River Remediation
TC	Total Carbon
TCCR	Tank Closure Cesium Removal
TIC	Total Inorganic Carbon
TOC	Total Organic Carbon
UL	Upper Limit
XRD	X-ray diffraction

1.0 Introduction

Savannah River Remediation (SRR) recently initiated an ion exchange process to remove radioactive cesium from dissolved salt solution in Tank 10H, known as Tank Closure Cesium Removal (TCCR). As part of that ongoing campaign, the salt in Tank 9H will be dissolved and transferred to Tank 10H for processing through the TCCR system, and SRR requested the characterization of the dissolved salt solution in Tank 9H prior to the transfer to Tank 10H.

On July 18, 2019, Savannah River Remediation Engineering (SRR-E) delivered two variable depth Tank 9H salt solution samples, identified as samples HTF-9-19-74 and HTF-9-19-75 (combined total sample volume of about 400 mL) to the Savannah River National Laboratory (SRNL) for characterization. These two Tank 9H variable depth samples (HTF-9-19-74 and HTF-9-19-75 taken at ~180" and 42", respectively) were retrieved from Riser 7 after hydro-lancing activities were complete. Approximately 4200 gallons of water were added to the tank during this activity.

This report contains analyses results for the characterization of two Tank 9H variable depth salt solution samples HTF-9-19-74 and HTF-9-19-75. This Tank 9H characterization effort is governed by a Technical Task Request (TTR)[1] and a Task Technical and Quality Assurance Plan (TTQAP).[2]

2.0 Objectives

The primary objectives of this Tank 9H variable depth sample characterization were to analyze, in duplicate, the following for the variable depth Tank 9H samples as summarized in Table 1.

- Densities of the variable depth Tank 9H filtrate samples
- Anions in the supernatant (supernatant diluted with both deionized water and dilute nitric acid prior to submittal for analyses)
- Inductively coupled plasma atomic emission spectroscopy (ICP-AES) elemental suite including Na, K, Al, and other elements
- Total mercury, including methyl and ethyl mercury in the variable depth filtrate samples
- Select radionuclides content in the variable depth sample filtrates including total gamma, total alpha/ beta.
- Inductively coupled plasma mass spectrometry (ICP-MS) elemental suite (Masses 59, 82, 84-114, 116-126, 128, 130, 133-187, 191, 193-196, 198, 203-208, 229-230, 232-252)
- Solids from the liquid/solid separation analyzed by X-ray diffraction for the crystal mineralogy of the solids from the two variable depth samples.

Table 1 Characterization of the Tank 9H salt solution

Analysis	Method	Preparation	Laboratory
Density	Gravimetric/volumetric	Filtrates	SCO
Elementals	ICP-AES	Acid dilutions	AD
Total mercury	DMA	Acid dilutions	AD
Methyl & Ethyl mercury	GC-AFS*	Acid dilutions	AD
Sr-90	Extraction/beta counting	Acid dilutions	AD
Cs-137	Gamma scan	Acid dilutions	AD
Co-60, Ce-144, Eu-154, Ru-106, Sb-125 and Am-241	Cs-removed gamma scan	Acid dilutions	AD
Pu-238, Pu-239/240, and Pu-241	Extraction/alpha PHA & LSC	Acid dilutions	AD
Masses 59, 82, 84-114, 116-126, 128, 130, 133-187, 191, 193-196, 198, 203-208, 229-230, 232-251	ICP-MS**	Acid dilutions	AD
Tc-99	Separation and LSC	Acid dilutions	AD
I-129	I-129 with separation	Cell extraction	SCO/AD
Am/Cm	Am/Cm	Acid dilutions	AD
Total alpha/beta	LSC	Acid dilutions	AD
$Al(OH)_4^{-1}$	Calculated from ICP-AES	n/a	PI
Free hydroxide	Carbonate removal & BT	Water dilutions	AD
CO_3^{2-}	TIC/TOC analyzer	Water dilutions	AD
NO_2^-, NO_3^-, SO_4^{2-}, Cl^-, F^- $C_2O_4^{2-}$, HCO_2^-, PO_4^{3-}, Br^{-1}	IC	Water dilutions	AD
Y-90 and $Ba-137^m$	Calculated	n/a	PI

ICP-AES = inductively coupled plasma atomic emission spectroscopy; DMA = Direct mercury analysis; ICP-MS = inductively coupled plasma mass spectroscopy; LSC = liquid scintillation counting; BT = base titration; IC = ion chromatography, TIC/TOC = total inorganic carbon/total organic carbon and GC-AFS = Gas chromatography atomic fluorescence spectroscopy. * Low level dilutions (ppt) required for these methods. **ICP-MS laboratory dilutions will be optimized for quantification.

3.0 Experimental Setups/Sample description and Preparations/Methodology

3.1 Tank 9H filtrate characterizations

The two "as-received" Tank 9H variable depth samples after opening the tank farm stainless steel sampling container and transferring the contents into separate polymethylpentene beakers, as shown in Figure 1, were filtered to obtain the filtrate and solid fractions as shown in Figure 2, inserts A-D. The liquid solid separation of the salt solutions were attained with the use of 0.45-micron Nalgene® filter membranes (Figures 2, inserts B and D). Laboratory analyses were based on the two Tank 9H salt solution filtrate sample aliquots for samples HTF-9-19-74 and HTF-9-19-75. Aliquots of the filtrate samples were diluted in either nitric acid solution (2.0 M HNO_3) or distilled and deionized (DI) water depending on the analysis requirements before transferring the diluted samples in green shielded bottles out of the SRNL Shielded Cells for analysis by the SRNL Analytical Development (AD) group. The only aliquots of these two Tank 9H variable depth samples which were not diluted prior to submittal for analysis were those for Iodine-129 (I-129) analysis. About one milliliter (~1.44 g) of these "neat" and undiluted filtrates were taken out of the Shielded Cells for characterization to enhance the detection limit for I-129.

3.2 Tank 9H solids fraction (salt crystals) water dissolution and X-ray characterization

The solid fractions, resulting from the liquid-solid separation of the two Tank 9H variable depth samples HTF-9-19-74 and HTF-9-19-75 were air dried in the cell environment for a few hours before sending small quantities (~1.0 grams) of each sample for X-ray analysis. Known quantities of each sample (2.53 grams for sample HTF-9-19-74 solid fraction and 3.10 grams for sample HTF-9-19-75 solid fraction) were dissolved in ordinary laboratory DI-water. The dissolution approach involved dropwise addition of DI-water to beakers containing each solid fraction sample and manual agitation of the container with the mixture followed by evaluation by visual inspection for the dissolution of the solid fraction after addition of DI-water. After the addition of about 1 mL of DI-water to each sample container, if the solid sample was not dissolved the dropwise addition of DI-water was continued until dissolution of the solids was visually observed.

3.3 Format of the Reported Results

The mean results, based on the average of all applicable analytical determinations, are reported in this document, along with the percent relative standard deviation (%RSD). The %RSD provides an indication of the measurement variation between duplicate determinations but is typically not an indicator of analytical accuracy. In general, the one sigma analytical uncertainty as reported by Analytical Development (AD) was 10%, although it was sometimes lower or higher. Specifically, the one sigma analytical uncertainties reported by AD were: a) ~10% for base titration, IC, ICP-AES, ICP-MS, and TIC/TOC analyses; b) ~20% for DMA analyses; and c) ~5% for Cs-137 determined by gamma spectroscopy. As such, only one to two of the leading digits reported for the AD analysis results should be considered significant.

The characterizations which were performed on the Tank 9H variable depth sample filtrates and solid fractions are listed in Table 1. A summary of the analytical methods used in these sample characterizations are presented in Appendix A. Appendix B contains the SRNL Analytical Development Laboratory Information Management System (LIMS) numbers for tracking the analytical data presented in this report. The sample analysis completion dates are tracked in LIMS. Most dilutions were performed in the SRNL Shielded Cells prior to taking representative sample aliquots out of the cells for analyses.

In the Tank 9H variable depth sample filtrate characterization results presented below, values preceded by "<" (less than sign) indicate values were below minimum detection limits (MDLs), and values preceeded by "≤" (less than or equal to sign) indicate that for replicates, at least one of the analysis values was above the instrument detection limit or MDL and at least one of the analysis values was below the detection limit or was an upper limit. Thus, where replicate analyses were both above and below the detection limit, the average of all replicates above and below the detection limit is given and a "≤ "sign precedes the average value. The standard deviations were calculated only for values that were all above the detection limits. The minimum detectable activity (MDA) is defined as the value above which instrument signal can be considered quantitative relative to the signal-to-noise ratio and the upper limit (UL) is defined as activity observed but biased high due to spectral interference or blank contamination. The detection limit (DL) as used in mass spectrometer or Inductively Coupled Plasma–Atomic Emission Spectroscopy (ICP-AES) analyses is equivalent to three times the standard deviation of the blank measurements.

The one sigma percent counting uncertainty for each radionuclide reported in the tables is based on the pooled estimate derived from the individual uncertainties for each replicate measurement for that radionuclide using Microsoft® Excel function, SQRT((SUMSQ(x_i)/n)), where n is the number of replicates and x_i is the individual uncertainty associated with each radionuclide for each analysis. Here it is assumed that the radio-analytical processes, be it counting or other techniques, are of the same precision for each individual measurement.

Occasionally, situations may be encountered where the samples prepared and analyzed in duplicate gave mixed results with one of the duplicate analyses results being less than the minimum detection activity (MDA). In these cases, the reporting of the one sigma percent uncertainty is presented in a slightly different format. In this situation, the individual percent uncertainty associated with each analysis for that radionuclide is reported along with MDA, upper limit (UL) values or the detection limit (DL) values as indicated by the analytical method.

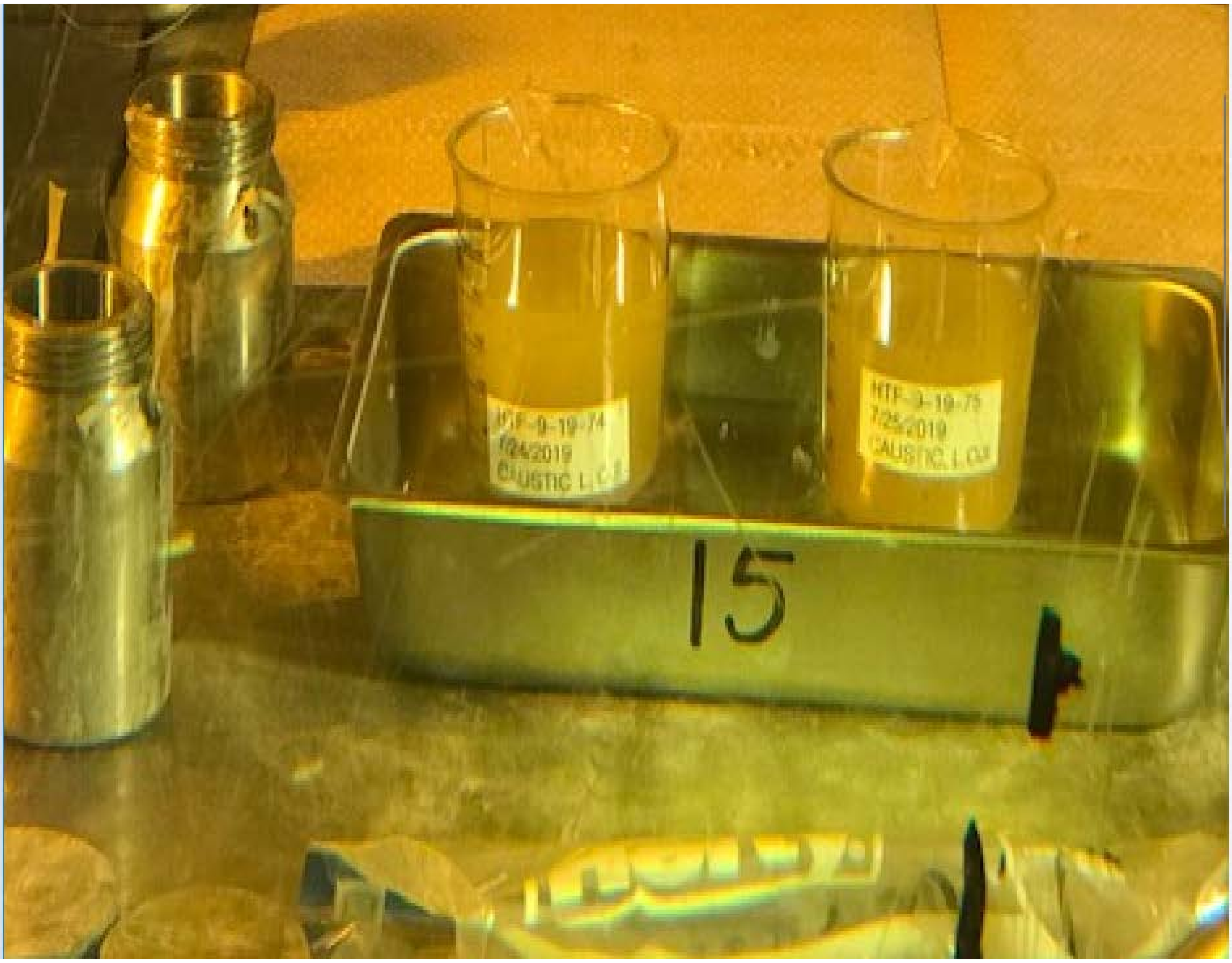
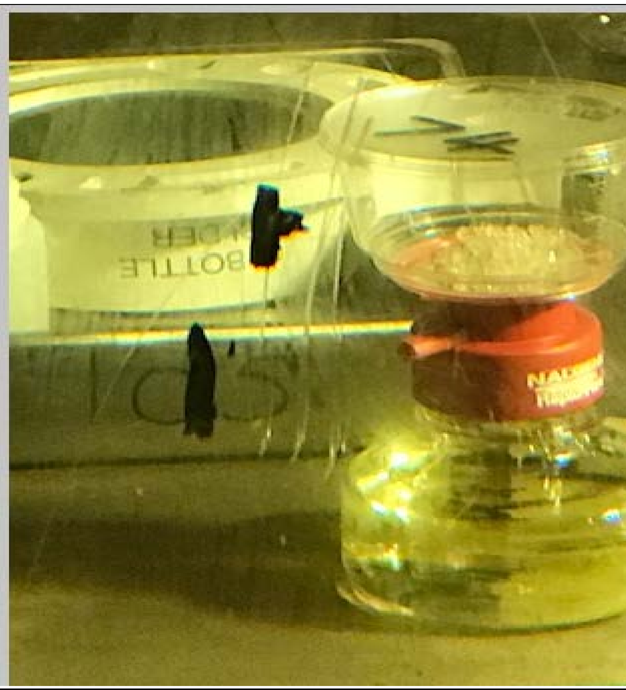
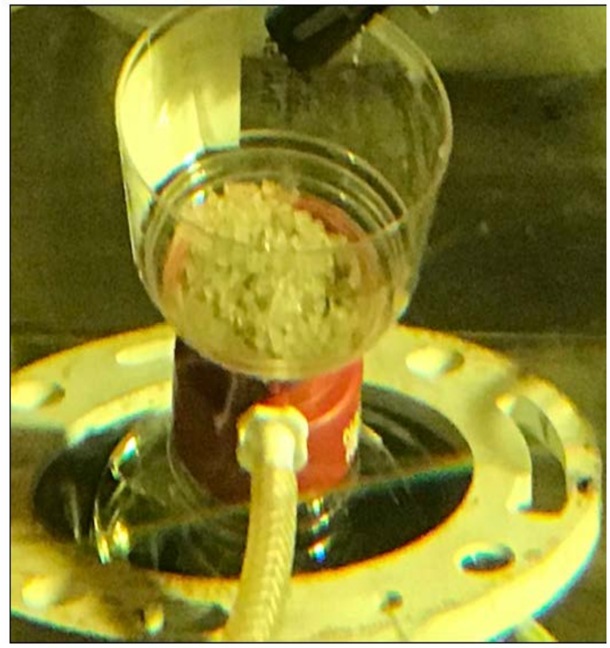


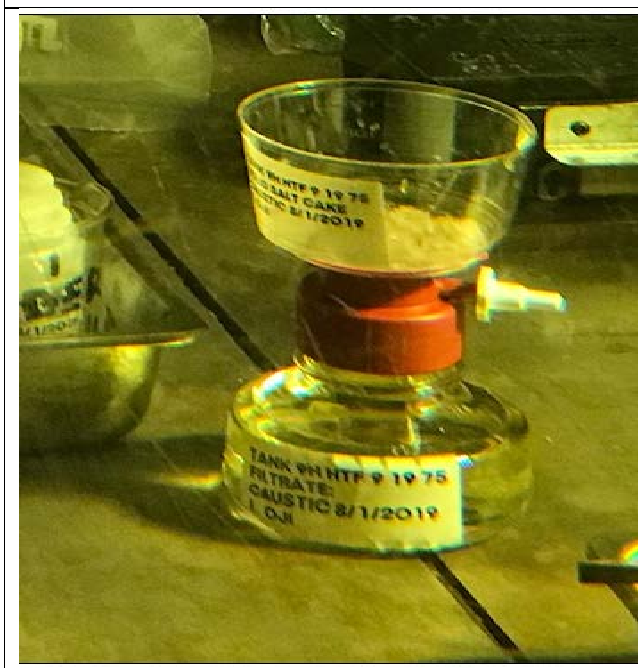
Figure 1 Variable depth Tank 9H salt solution samples HTF-9-19-74 and HTF-9-19-75.



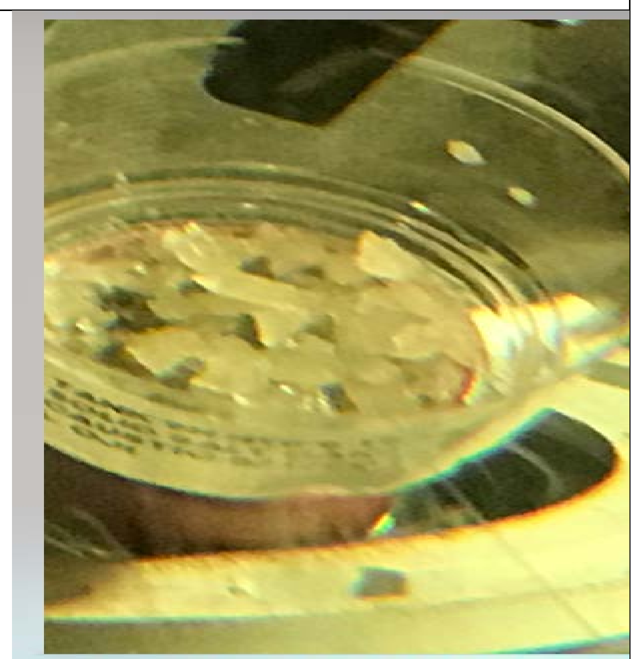
Insert A: Sample HTF-9-19-74 salt crystals and filtrate.



Insert B: Sample HTF-9-19-74 salt crystals



Insert C: Sample HTF-9-19-75 salt crystals and filtrate



Insert D: Sample HTF-9-19-75 salt crystals

Figure 2 Filtrates and crystals for Tank 9H salt solution samples HTF-9-19-74 and HTF-9-19-75

4.0 Results and Discussion

Laboratory analyses were performed on the Tank 9H variable depth samples HTF-9-19-74 and HTF-9-19-75. A combination of routine dissolution/measurement techniques and "tailor-made" digestion/isolation/analysis methods were used to quantify several stable constituents (elements and anions) and select radionuclides. A summary of the customer requested constituents to be analyzed for in the Tank 9H variable depth samples is presented in Table 1.

Most of these analyses were performed in duplicate with the percent relative standard deviation (%RSD), a measure of the variability between each duplicate data set expressed as a percentage, used to estimate the analytical result quality for each analyte. Tank 9H anions and mercury species constituents, elemental constituents, total cesium, select radionuclides and mass spectral suite data (mass 59-252) are presented in Tables 2-6 for sample HTF-9-19-74 and Tables 7-11 for sample HTF-9-19-75, respectively.

The Tank 9H filtrate characterizations with the largest one sigma % measurement uncertainty as shown in some tables, meaning measurements with one sigma % measurement uncertainty above or approaching the expectation of 20% for these methods, were analyses for Sr-90 (Y-90) and Pu-239/240. As presented in Tables 5 (sample HTF-9-19-74) and Table 10 (sample HTF-9-19-75), the one sigma % measurement uncertainties for Sr-90 were 18.1 and 21.1%, respectively. Excluding measurements which involved replicates where at least one of the analysis values was above the instrument detection limit and the other analysis value was below the detection limit or was an upper limit, only the measurement for Pu-239/240 in sample HTF-9-19-75 presented analytical measurement uncertainty with extra ordinarily high uncertainties. The one sigma % measurement uncertainty for this sample Pu-239/240 characterization was 51.2% (Table 10). Some of the analysis results with the largest one sigma % measurement uncertainties also exhibited larger than expected %RSD for these methods, too. For example, the one sigma % measurement uncertainty for Pu-239/240, Table 10, is 51.2 and the corresponding %RSD is 56.4%.

The ICP-AES results for the Tank 9H variable depth samples are presented in Tables 3 and 8. With the analysis result for Mg concentration in Table 3 being the exception, the %RSD for all the analyses were below 10% and thus meet the analytical expectations for this Tank 9H salt solution filtrate sample characterization using the specified analytical methods. The %RSD for the Mg analysis result was 32.1, which is above the expectation of less than 20 %RSD. In the ICP-MS mass data presented in Tables 6 and 11, only the analysis result for mass 88 for sample HTF-9-19-74, Table 6 exceeded the %RSD expectation; the %RSD for mass 88 in this sample was 48.4%.

Both the one sigma % measurement uncertainties and the %RSD for some of these analytes (Sr-90, Pu-239/240 and ICP-MS mass 88) are above analytical expectations because of the low concentrations of these analytes in the Tank 9H filtrate samples. Their diluted concentrations were near the instrument minimum detection limits which leads to large analytical percent uncertainties for these analytes.

4.1 <u>Tank 9H Supernate Characterization for Anions and Mercury Species</u>

The measured densities for these two variable depth Tank 9H filtrate samples (HTF-9-19-74, and HTF-9-19-75), are 1.436 ± 0.002 and 1.433 ± 0.003 g/mL, respectively. The densities for these two Tank 9H variable depth sample filtrates are essentially the same.

The free-OH^{-1} concentrations for sample HTF-9-19-74 (0.66 ± 0.07 molar) and sample HTF-9-19-75 (0.66 ± 0.03 molar) filtrates were used to calculate their corresponding pH values, which are both 13.8, as presented in Tables 2 and 7. The pH result was calculated using the following equation:

$$pH = 14 + \log_{10}\left(OH^{-}\right).$$

To check the anion results, a cation-anion normality balance for samples HTF-9-19-74 and HTF-9-19-75 was performed with the filtrate analytical data on cations and anions. The normal concentrations of cations (mainly Na^+ and K) were summed, as were the anions (NO_3^-, NO_2^-, SO_4^{2-}, HCO_2^{1-}, Cl^-, CO_3^{2-}, PO_4^{3-}, AlO_2^-, $C_2O_4^{2-}$ and free OH^-). The cation sums for the two samples, HTF-9-19-74 and HTF-9-19-75, were 9.76 and 9.53 M, respectively. The anion sums for the two samples were, respectively, 9.69 and 9.18 M.

The differences between the cation and anion molarity values for these samples HTF-9-19-74 and HTF-919-75 were 0.72 % [(9.76-9.69)/((9.76+9.69)/2)]*100 and 3.74% [(9.539.18)/((9.53+9.18)/2)]*100, respectively and are considered to be within the nominal uncertainties (1 sigma) for ICP-ES, IC and free-OH^{-1} analyses, which is about 10%. The small differences can be attributed to analytical uncertainties and analyte detections limitations.

Table 2 Corrosion Chemistry Analyses Results for Tank 9H Supernate (filtrate) HTF-9-19-74

Analyte	Analysis-1 M	Analysis-2 M	Average concentration, M	Standard deviation	%RSD N = 2
Fluoride, F^-	<1.17E-01	<1.17E-01	**<1.17E-01**	n/a	n/a
Formate, HCO_2^{1-}	<4.93E-02	<4.94E-02	**<4.93E-02**	n/a	n/a
Chloride, Cl^-	<6.26E-02	<6.27E-02	**<6.26E-02**	n/a	n/a
Nitrite, NO_2^-	1.69E+00	1.65E+00	**1.67E+00**	*2.86E-02*	1.71
Nitrate, NO_3^-	5.29E+00	5.16E+00	**5.23E+00**	*9.46E-02*	1.81
Phosphate, PO_4^{3-}	<2.34E-02	<2.34E-02	**<2.34E-02**	n/a	n/a
Sulfate, SO_4^{2-}	5.52E-02	5.41E-02	**5.47E-02**	*7.47E-04*	1.37
Oxalate, $C_2O_4^{2-}$	<2.52E-02	<2.53E-02	**<2.52E-02**	n/a	n/a
Bromide, Br^{1-}	<2.78E-02	<2.78E-02	**<2.78E-02**	n/a	n/a
Free Hydroxide, OH^-	7.1E-01	61E-01	**6.6E-01**	*7.29E-02*	11.0
Carbonate, CO_3^{2-}	8.05E-01	8.35E-01	**8.20E-01**	*2.15E-02*	2.62
$Al(OH)_4^{-1}$	3.48E-01	3.46E-01	**3.47E-01**	*1.49E-03*	0.43
Total carbon, gC/L	9.97E+00	1.01E+01	**1.00E+01**	8.00E-02	0.84
Inorganic carbon gC/L	9.66E+00	1.00E+01	**9.84E+00**	2.6E-01	2.62
Methyl mercury cation, mg/L	5.34E-01	5.95E-01	**5.64E-01**	*4.32E-02*	*7.66*
Total mercury, mg/L	3.30E+00	3.46E+00	**3.38E+00**	*1.2E-01*	3.42
pH (calculated)	n/a	n/a	**13.82**	n/a	n/a

N = number of replicates.

Table 3 Elemental Analyses of Tank 9H Supernate (Filtrate): Variable Depth Sample HTF-9-19-74

Element	Analysis-1, mg/L	Analysis-2, mg/L	Average; Concentration, mg/L	Average; Concentration, M	%RSD; N = 2
Ag	<1.44E+00	<1.45E+00	**<1.44E+00**	<1.34E-05	n/a
Al	9.40E+03	9.34E+03	**9.37E+03**	**3.47E-01**	0.43
B	<3.95E+00	<3.98E+00	**<3.97E+00**	**<3.67E-04**	n/a
Ba	<9.42E-01	<9.50E-01	**<9.46E-01**	**<6.89E-06**	n/a
Be	<2.02E-01	<2.04E-01	**<2.03E-01**	**<2.25E-05**	n/a
Ca	<7.85E-01	<7.92E-01	**<7.89E-01**	**<1.97E-05**	n/a
Cd	<4.71E-01	<4.75E-01	**<4.73E-01**	**<4.21E-06**	n/a
Ce	<1.10E+01	<1.11E+01	**<1.10E+01**	**<7.86E-05**	n/a
Co	<1.01E+00	<1.02E+00	**<1.01E+00**	**<1.72E-05**	n/a
Cr	1.07E+02	1.09E+02	**1.08E+02**	**2.07E-03**	1.48
Cu	<2.17E+00	<2.19E+00	**<2.18E+00**	**<3.43E-05**	n/a
Fe	<2.54E+00	<2.56E+00	**<2.55E+00**	**<4.56E-05**	n/a
Gd	<9.85E-01	<9.93E-01	**<9.89E-01**	**<6.29E-06**	n/a
K	1.23E+03	1.29E+03	**1.26E+03**	**3.22E-02**	2.87
La	<9.94E-01	<1.00E+00	**<9.98E-01**	**<7.19E-06**	n/a
Li	<1.39E+00	<1.40E+00	**<1.39E+00**	**<2.01E-04**	n/a
Mg	1.80E-01	1.13E-01	**1.46E-01**	**6.02E-06**	32.1
Mn	<5.61E-01	<5.66E-01	**<5.63E-01**	**<1.03E-05**	n/a
Mo	<5.45E+01	<5.50E+01	**<5.48E+01**	**<5.71E-04**	n/a
Na	2.23E+05	2.24E+05	**2.24E+05**	**9.73E+00**	0.45
Ni	<6.96E+00	<7.02E+00	**<6.99E+00**	**<1.19E-04**	n/a
P	3.61E+02	3.76E+02	**3.68E+02**	**1.19E-02**	2.75
Pb	<1.67E+01	<1.68E+01	**<1.68E+01**	**<8.09E-05**	n/a
S	<3.33E+03	<3.36E+03	**<3.35E+03**	**<1.04E-01**	n/a
Sb	<1.57E+01	<1.58E+01	**<1.58E+01**	**<1.30E-04**	n/a
Si	<9.54E+00	<9.62E+00	**<9.58E+00**	**<3.41E-04**	n/a
Sn	<2.45E+01	<2.47E+01	**<2.46E+01**	**<2.07E-04**	n/a
Sr	<1.23E+01	<1.24E+01	**<1.23E+01**	**<1.40E-04**	n/a
Th	<2.03E+01	<2.05E+01	**<2.04E+01**	**<8.80E-05**	n/a
Ti	<2.18E-01	<2.20E-01	**<2.19E-01**	**<4.57E-06**	n/a
U	<5.95E+01	<6.00E+01	**<5.97E+01**	**<2.51E-04**	n/a
V	<3.84E-01	<3.87E-01	**<3.85E-01**	**<7.56E-06**	n/a
Zn	<2.45E+00	<2.47E+00	**<2.46E+00**	**<3.76E-05**	n/a
Zr	<3.75E-01	<3.78E-01	**<3.76E-01**	**<4.13E-06**	n/a

Table 4 Total Cesium* in Tank 9H Variable Depth Sample HTF-9-19-74

Isotope	Average Concentration, mg/L	%RSD**	Isotopic distribution, mass %	Moles	Mole fraction
Cs-133	20.3	0.14	74.1	1.53E-04	7.46E-01
Cs-134	0.005	3.89	0.02	3.51E-08	1.72E-04
Cs-135	2.09	0.27	7.63	1.55E-05	7.57E-02
Cs-137	4.99	2.29	18.2	3.64E-05	1.78E-01
Total Cs	**2.74E+01**	n/a	**100.00**	**2.05E-04**	**1.00E+00**

*Cesium isotopic concentrations are based on ICP-MS data. **%RSD is based on the standard deviation of duplicate analysis.

Table 5 Radiological results for Tank 9H Filtrate: Variable Depth Sample HTF-9-19-74

Analyte	Analysis-1 dpm/mL	Analysis-2 dpm/mL	Average dpm/mL	Average Ci/L of supernate	%RSD N = 2	One sigma % uncertainty
Total alpha	<1.73E+07	<1.54E+07	**<1.64E+07**	**<7.38E-03**	n/a	UL
Total beta	1.25E+09	1.26E+09	**1.26E+09**	**5.66E-01**	0.69	10
Co-60	<3.18E+02	<3.01E+02	**<3.10E+02**	**<1.40E-07**	n/a	MDA
Sr-90	1.29E+05	1.34E+05	**1.31E+05**	**5.92E-05**	2.63	18.1
Y-90	1.29E+05	1.34E+05	**1.31E+05**	**5.92E-05**	2.63	18.1
Tc-99	3.90E+05	3.83E+05	**3.86E+05**	**1.74E-04**	1.20	6.26
Ru-106	<1.97E+03	<2.01E+03	**<1.99E+03**	**<8.97E-07**	n/a	MDA
Sb-125	<1.15E+03	<1.15E+03	**<1.15E+03**	**<5.17E-07**	n/a	MDA
Sb-126	3.34E+03	3.53E+03	**3.43E+03**	**1.55E-06**	3.95	10
Sn-126	3.34E+03	3.53E+03	**3.43E+03**	**1.55E-06**	3.95	10
I-129	<2.55E+01	<2.50E+01	**<2.53E+01**	**<1.14E-08**	n/a	MDA
Cs-134	<1.88E+05	<1.35E+05	**<1.62E+05**	**<7.29E-05**	n/a	MDA
Cs-137	1.06E+09	1.07E+09	**1.06E+09**	**4.78E-01**	0.70	5
Ba-137m	1.00E+09	1.01E+09	**1.01E+09**	**4.53E-01**	0.70	5
Ce-144	<1.95E+03	<1.94E+03	**<1.95E+03**	**<8.77E-07**	n/a	MDA
Eu-152	<7.96E+02	<7.89E+02	**<7.93E+02**	**<3.57E-07**	n/a	MDA
Eu-154	<5.59E+02	<5.60E+02	**<5.60E+02**	**<2.52E-07**	n/a	MDA
Eu-155	<7.40E+02	<8.75E+02	**<8.07E+02**	**<3.64E-07**	n/a	MDA
Th-232†	< 1.12E-03	< 1.10E-03	**< 1.11E-03**	**< 5.00E-13**	n/a	MDA
U-233†	5.48E+01	5.47E+01	**5.47E+01**	**2.47E-08**	0.17	10
U-234†	3.48E+01	3.76E+01	**3.62E+01**	**1.63E-08**	5.43	10
U-235†	8.12E-02	8.27E-02	**8.20E-02**	**3.69E-11**	1.33	10
U-236†	6.01E-01	5.98E-01	**6.00E-01**	**2.70E-10**	0.30	10
Np-237†	4.70E+00	4.35E+00	**4.52E+00**	**2.04E-09**	5.51	10
U-238†	2.87E-01	3.02E-01	**2.94E-01**	**1.33E-10**	3.43	10
Pu-238	1.47E+04	1.48E+04	**1.47E+04**	**6.64E-06**	0.42	6.50
Pu-239/240	1.84E+02	<1.40E+02	**≤1.62E+02**	**≤7.29E-08**	n/a	32.8/MDA
Pu-241	<3.22E+03	<1.82E+03	**<2.52E+03**	**<1.14E-06**	n/a	UL
Am-241	<1.30E+02	<5.19E+01	**<9.11E+01**	**<4.10E-08**	n/a	MDA
Am-242m	<1.33E+02	<2.30E+01	**<7.81E+01**	**<3.52E-08**	n/a	MDA
Am-243	<2.75E+02	<1.41E+02	**<2.08E+02**	**<9.38E-08**	n/a	MDA
Cm-242	<1.10E+02	<1.90E+01	**<6.46E+01**	**<2.91E-08**	n/a	MDA
Cm-243	<9.43E+02	<4.67E+02	**<7.05E+02**	**<3.18E-07**	n/a	MDA
Cm-244	<1.50E+02	<2.69E+01	**<8.86E+01**	**<3.99E-08**	n/a	MDA
Cm-245	<7.75E+02	<3.83E+02	**<5.79E+02**	**<2.61E-07**	n/a	MDA
Cm-247	<8.86E+02	<5.08E+02	**<6.97E+02**	**<3.14E-07**	n/a	MDA
Cf-249	<1.00E+03	<5.40E+02	**<7.71E+02**	**<3.47E-07**	n/a	MDA
Cf-251	<8.86E+02	<4.62E+02	**<6.74E+02**	**<3.04E-07**	n/a	MDA

† Activities for Th-232, U isotopes and Np-237 are calculated from ICP-MS data

As presented in Tables 2 and 7, nitrate and nitrite were the two predominant anions in the two Tank 9H variable depth samples. The nitrate and nitrite concentrations averaged 5.23 M and 1.67 M for sample HTF-9-19-74, and 4.84 M and 1.65 M for sample HTF-9-19-75, respectively. Another anion which measured above instrument detections limit for these two Tank 9H filtrate samples (HTF-9-19-74 and HTF-9-19-75) was sulfate which averaged 5.47E-02 M and 5.18E-02 M, respectively. A measurable quantity of phosphate anion was detected in sample HTF-9-19-75 at 6.54E-03M, however, the phosphate concentration in sample HTF-9-19-74 was below instrument detection limit. The concentration of all other anions measured by IC in these two variable depth samples were below instrument detection limits as shown in Tables 2 and 7. As shown in Tables 2 and 7, the concentrations of total carbon[‡] and inorganic carbon in the two variable Tank 9H filtrates are essential equal, which means there is no measurable organic carbon in either sample.

Total mercury and methyl mercury concentrations in the two Tank 9H filtrates averaged 3.38 and 5.64E-01 mg /L for sample HTF-9-19-74 and 2.39 and 4.74E-01 mg/L for sample HTF-9-19-75, respectively. The combined total mercury value for the variable depth Tank 9H samples averaged less than 3 mg/L.

4.2 Cesium Isotopes from ICP-MS and Isotopic Distributions

Cesium isotopic mass distributions (masses 133, 134, 135 and 137) in the two Tank 9H filtrate samples HTF-9-19-74 and HTF-9-1975 variable depth samples, as shown in Tables 4 and 9, respectively, are essentially identical in magnitude in both average concentrations and average isotopic distributions for the isotopes. It is worth noting that non-cesium elements like Ba and La with atomic masses in the 134-138 regions can bias high the cesium isotopic concentrations if their concentrations in the filtrates are relatively high (assuming the ICP-MS is calibrated for cesium). The combined ICP-MS concentration for Ba, La and Cs-137 with mass 137 averaged 4.96 ± 0.12 mg/L for sample HTF-9-19-74 and 5.08 ± 0.08 mg/L for sample HTF-9-19-75 as shown in Tables 6 and 11. The analytical results for Cs-137 alone by a non-ICP-MS method, which is to say, gamma spectroscopy technique, gives a value of 5.49 ± 0.14 mg/L (1.06E+09 dpm/ mL) for sample HTF-9-19-74 and 5.75 ± 0.04 mg/L (1.11E+09 dpm/mL) for sample HTF 9 19-75. Since the ICP-MS analysis results for mass 137 for the two samples are about equal in magnitude with the gamma scan results for Cs-137, it is safe to conclude that Cs-137 accounts for most of the mass 137 in both samples and the mass contributions from these other elements with mass 137 are negligible. It is also worth noting that the concentration of Ba-138 (major stable barium isotope) is two orders of magnitude lower than mass 137, which makes any contribution from Ba-137 negligible. The one sigma analytical uncertainty for Cs-137 determined by gamma spectroscopy is ~5% and it is 10% for ICP-MS determinations.

The assigning of "likely element (s)" for any atomic mass in the ICP-MS data is based on the nuclide isotopic abundance, atomic weight and half-lives. Some isotopic masses, for example masses, 127 (iodine), 190 and 192 (osmium), 197 (gold) and 199-202 (Hg), require special method development efforts for their isolation and quantification; these methods are not within the analytical capabilities of the ICP-MS method employed here. The analysis results for these elements are based on other analytical methods as summarized in Appendix A.

4.3 Select Radionuclides, Elemental Analysis and ICP-MS masses 59-252

The measured cesium-137 average activity in the supernate samples HTF-9-19-74 and HTF-9-19-75 filtrates were 1.06E+09 (0.7 %RSD) and 1.11E+09 (2.6 % RSD) dpm/mL, respectively (Tables 5 and 10). The corresponding $Ba-137^m$ activities, calculated as 94.7% of the Cs-137 values, are 1.01E+09 (0.7 %RSD) dpm/mL and 1.05E+09 (2.6 %RSD) dpm/mL, respectively. The other beta-emitting

[‡] Total carbon = inorganic carbon + organic carbon; if inorganic carbon concentration = total carbon, then organic carbon concentration = 0.

radionuclides in the Tank 9H variable depth samples are Sr-90 (1.31E+05 (2.63 % RSD) dpm/mL), Y-90 (1.31E+05 (2.63 %RSD) dpm/mL for sample HTF-9-19-74 and Sr-90 (1.51E+05 (8.85 % RSD) dpm/mL), Y-90 (1.51E+05 (8.85 %RSD) dpm/mL for sample HTF-9-19-75. The measured activities for Co-60 were below instrument detection limits. The only cesium-removed gamma analytes which were detected were Sb-126 and Sn-126 in both filtrate samples. The equilibrium values for these radionuclides averaged 3.43E+03 (3.95 %RSD) dpm/mL for sample HTF-9-19-74 and 3.44E+03 (4.22 %RSD) dpm/mL for sample HTF-9-19-75.

The concentration of Tc-99 in the two filtrates averaged 3.86E+05 (1.2 % RSD) dpm/mL for sample HTF-9-19-74 and 3.66E+05 (3.39 % RSD) dpm/mL for sample HTF-9-19-75. Iodine-129 activities which averaged <2.53E+01 dpm/mL for sample HTF-9-19-74 and <1.91E+01 dpm/mL for sample HTF-9-19-75 were below instrument detection limits. With Am-241 activity in sample HTF-9-19-75 being the exception, all the Am/Cm measurements for the Tank 9H filtrates were below instrument detection limits as shown in Tables 5 and 10. The Am-241 activity in one of the duplicate analyses for sample HTF-9-19-75 was above instrument detection limit (1.32E+02dpm/mL) and the other was below instrument detection limit (<2.11E+02 dpm/mL).

Total alpha and total beta activities in the two Tank 9H filtrates are presented in Tables 5 and 10. The total alpha activity determinations for the two filtrate samples were all less than values (upper limits) because of possible spectral interferences. Total alpha activities averaged <7.38E-03 Ci/L (<1.64E+07 dpm/mL) for sample HTF-9-19-74 and <7.36E-03 Ci/L (<1.63E+07 dpm/mL) for sample HTF-9-19-75. However, a summation of the primary alpha emitting radionuclide[§] concentrations (Pu-238, Pu-239/240, Am-241 and Cm-244 from Tables 5 and 10) gives one a better upper limit estimate for total alpha activities in the two filtrate samples. This summed total alpha activity values for these samples are ≤6.79E-06 Ci/L and ≤6.47E-06 Ci/L for samples HTF-9-19-74 and HTF-9-19-75, respectively. These values, as expected, are about three orders of magnitude lower than the directly measured values for total alpha.

The total beta activity in the two filtrate samples were above instrument detection limits and average 1.26E+09 (0.69 %RSD) dpm/mL for sample HTF-9-19-74 and 1.28E+09 (1.10 %RSD) dpm/mL for sample HTF-9-19-75.

The average uranium enrichment (U-235/total uranium*100) for these samples HTF-9-19-74 and HTF-9-19-75 supernates are 4.1 and 4.2 percent, respectively (Appendix C). The average total uranium concentrations in these supernates are 4.21E-01 and 4.30E-01 mg/L, respectively. Taking into consideration the ICP-MS one sigma analytical uncertainties of about 10%, these measurements (uranium enrichment and total uranium) are quantitively the same in magnitude for these two filtrate samples.

The average Pu-238 to total plutonium ratios by mass for these two Tank 9H variable depth samples are 0.059 for sample HTF-9-19-74 and 0.040 for sample HTF-9-19-75. It is worth noting that these ratios are based on the assumptions that Pu-239 mass constitutes most of the mass reported for Pu-239/240 analysis results as shown in Tables 5 and 10 and where the measured values for Pu-239/240 or Pu-241 are less than detectable values the absolute values are used for these ratio calculations.

The predominant cation concentrations in the Tank 9H filtrate sample HTF-9-19-74 are Na [2.24E+05 (0.45 %RSD) mg/L or 9.73 M], K [1.26E+03 (2.87 %RSD) mg/L or 0.032 M] and Cr [1.08E+02 (1.48 %RSD) mg/L or 0.002 M]. The concentration of these cations in sample HTF-9-19-75 filtrate are Na

[§] Some radionuclide measurements were above instrument detection limits while others were below instrument detection limits or were upper limits, hence, the summed values for total calculated alpha have less than or equal signs (≤).

[2.19E+05 (0.68 %RSD) mg/L or 9.50 M], K [1.27E+03 (0.84 %RSD) mg/L or 0.032 M] and Cr [1.04E+02 (1.75 %RSD) mg/L or 0.002 M]. Aluminum concentration in Tank 9H, which is present as the aluminate anion, averaged 9.37E+03 (0.43 %RSD) mg/L or 0.347 M for sample HTF-9-19-74 and 1.04E+04 (0.49 %RSD) mg/L or 0.386 M for sample HTF-9-19-75.

Table 6 and 11 show the ICP-MS analysis of the two variable depth Tank 9H filtrates for most of the other elements. The last column of each of these two tables shows the assigned likely element (s) with the corresponding masses (m/z). The assigning of "likely element" for any atomic mass in the ICP-MS data is based on the nuclide isotopic abundance, atomic weight and half-lives. Some masses, for example masses, 127 (iodine), 190 and 192 (osmium), 197 (gold) and masses 199-202 (Hg), some of which have been analyzed in previous sections require special method development efforts for their isolation and quantification as presented in Appendix A.

Table 6 Mass Spectral Analyses of Tank 9H Filtrate: Variable Depth Sample HTF-9-19-74

ICP-MS, m/z	Analysis-1, mg/L	Analysis 2, mg/L	Average, mg/L	%RSD; N = 2	Likely element (s)
59	<4.59E-03	<4.53E-03	**<4.56E-03**	n/a	Co
82	1.81E-01	1.81E-01	**1.81E-01**	0.04	Se
84	<2.30E-03	<2.26E-03	**<2.28E-03**	0.99	Sr
85	2.51E+00	2.55E+00	**2.53E+00**	1.09	Rb
86	<2.30E-03	2.43E-03	**≤2.36E-03**	n/a	Sr
87	5.24E+00	5.29E+00	**5.27E+00**	0.63	Rb, Sr
88	8.09E-03	1.65E-02	**1.23E-02**	48.4	Sr
89	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Y
90	1.73E-02	1.81E-02	**1.77E-02**	3.30	Zr
91	1.60E-02	1.59E-02	**1.59E-02**	0.67	Zr
92	2.63E-01	2.58E-01	**2.61E-01**	1.21	Zr, Mo
93	1.84E-02	1.85E-02	**1.85E-02**	0.56	Nb
94	1.73E-01	1.72E-01	**1.73E-01**	0.14	Nb, Mo
95	1.39E+01	1.39E+01	**1.39E+01**	0.01	Mo
96	3.01E-01	3.01E-01	**3.01E-01**	0.15	Ru, Zr, Mo
97	1.29E+01	1.29E+01	**1.29E+01**	0.33	Mo, Tc
98	1.28E+01	1.30E+01	**1.29E+01**	0.99	Ru, Mo, Tc
99	8.81E+00	9.04E+00	**8.93E+00**	1.77	Tc, Ru
100	1.33E+01	1.33E+01	**1.33E+01**	0.09	Ru, Mo
101	7.21E-01	7.36E-01	**7.29E-01**	1.47	Ru
102	6.59E-01	6.21E-01	**6.40E-01**	4.15	Ru, Pd
103	1.78E+00	1.68E+00	**1.73E+00**	4.26	Rh
104	3.29E-01	3.19E-01	**3.24E-01**	2.09	Ru, Pd
105	2.74E-02	2.89E-02	**2.81E-02**	3.81	Pd
106	2.23E-02	2.74E-02	**2.49E-02**	14.5	Pd, Cd
107	1.46E-02	1.74E-02	**1.60E-02**	12.2	Ag
108	< 1.38E-02	< 6.37E-02	**< 3.88E-02**	n/a	Pd, Cd
109	< 9.18E-03	< 1.20E-01	**< 6.47E-02**	n/a	Ag
110	< 6.89E-03	< 6.79E-03	**< 6.84E-03**	n/a	Pd, Cd
111	< 6.89E-03	< 6.79E-03	**< 6.84E-03**	n/a	Cd
112	2.89E-02	2.89E-02	**2.89E-02**	0.19	Sn, Cd
113	< 6.89E-03	< 6.79E-03	**< 6.84E-03**	n/a	In, Cd
114	2.46E-02	2.55E-02	**2.50E-02**	2.77	Sn, Cd
116	3.93E-01	3.96E-01	**3.95E-01**	0.57	Sn, Cd
117	2.73E-01	2.67E-01	**2.70E-01**	1.60	Sn
118	7.88E-01	7.76E-01	**7.82E-01**	1.15	Sn
119	2.41E-01	2.49E-01	**2.45E-01**	2.41	Sn
120	1.05E+00	1.02E+00	**1.03E+00**	1.92	Sn
121	1.48E-02	1.64E-02	**1.56E-02**	7.19	Sb
122	2.74E-01	2.65E-01	**2.70E-01**	2.26	Te, Sn
123	9.54E-03	9.38E-03	**9.46E-03**	1.24	Sb, Te
124	4.05E-01	3.86E-01	**3.95E-01**	3.39	Te, Sn
125	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Sb, Te
126	1.25E+00	1.25E+00	**1.25E+00**	0.06	Te
128	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Te
130	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Te
133	2.03E+01	2.03E+01	**2.03E+01**	0.14	Cs
134	4.58E-03	4.84E-03	**4.71E-03**	3.89	Ba, Cs

Table 6 Continued. Mass Spectral Analyses of Tank 9H Filtrate: Variable Depth Sample HTF-9-19-74

ICP-MS, m/z	Analysis-1, mg/L	Analysis-2, mg/L	Average, mg/L	%RSD, N = 2	Likely element (s)
135	2.09E+00	2.10E+00	**2.09E+00**	0.27	Ba, Cs
136	< 2.30E-03	2.68E-03	**≤2.49E-03**	n/a	Ce, Ba
137	5.08E+00	4.91E+00	**4.99E+00**	2.29	Cs, Ba, La
138	1.66E-02	1.77E-02	**1.72E-02**	4.83	Ba, La, Ce
139	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	La
140	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Ce
141	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Pr
142	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Nd, Ce
143	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Nd., Pm
144	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Nd, Sm, Pm
145	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Nd, Pm
146	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Nd, Sm
147	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Sm, Ti
148	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Nd, Gd, Sm
149	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Sm
150	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Nd, Gd, Sm, Eu
151	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Eu
152	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Gd, Sm, Eu
153	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Eu
154	4.86E-03	4.68E-03	**4.77E-03**	2.57	Gd, Sm, Eu, Dy
155	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Gd
156	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Gd, Dy
157	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Gd, Tb
158	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Gd, Dy, Tb
159	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Tb
160	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Gd, Dy
161	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Dy
162	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Dy, Er
163	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Dy, Ho
164	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Dy, Er
165	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Ho
166	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Er, Ho
167	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Er
168	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Er, Yb
169	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Tm
170	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Er, Yb
171	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Yb
172	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Yb
173	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Yb
174	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Yb, Hf
175	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Lu
176	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Lu, Hf, Yb
177	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Hf
178	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Hf
179	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Hf
180	< 2.30E-03	2.31E-03	**≤ 2.30E-03**	n/a	Hf, W, Ta
181	< 2.30E-03	< 2.26E-03	**< 2.28E-03**	n/a	Ta
182	1.56E-01	1.69E-01	**1.62E-01**	5.98	Hf, W

Table 6 Continued. Mass Spectral Analyses of Tank 9H Filtrate: Variable Depth Sample HTF-9-1974

ICP-MS, m/z	Analysis-1, mg/L	Analysis-2, mg/L	Average, mg/L	%RSD, N = 2	Likely element (s)
183	8.30E-02	9.16E-02	**8.73E-02**	6.93	W
184	1.79E-01	1.99E-01	**1.89E-01**	7.43	W
185	2.62E-03	2.77E-03	**2.69E-03**	3.95	Re
186	1.70E-01	1.81E-01	**1.76E-01**	4.60	Os, W
187	8.88E-03	7.74E-03	**8.31E-03**	9.67	Re, Os
188	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Os
189	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Os
191	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Ir
193	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Ir, Pt
194	< 4.59E-03	<4.53E-03	**< 4.56E-03**	n/a	Pt
195	< 4.59E-03	<4.53E-03	**< 4.56E-03**	n/a	Pt
196	6.11E-03	6.61E-03	**6.36E-03**	5.58	Hg, Pt
198	3.07E-01	3.26E-01	**3.17E-01**	4.37	Hg, Pt
203	< 2.30E-03	<2.26E-03	**2.28E-03**	0.99	Tl
204	2.73E-01	2.61E-01	**2.67E-01**	3.30	Pb, Hg
205	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Tl
206	1.63E+00	1.65E+00	**1.64E+00**	0.90	Pb
207	1.41E+00	1.42E+00	**1.42E+00**	0.79	Pb
208	3.43E+00	3.32E+00	**3.38E+00**	2.43	Pb
229	< 2.30E-03	NA	**<2.30E+00**	n/a	Th
230	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Th
232	< 4.59E-03	< 4.53E-03	**< 4.56E-03**	n/a	Th, U
233	2.55E-03	2.54E-03	**2.55E-03**	0.17	U
234	2.51E-03	2.71E-03	**2.61E-03**	5.43	U
235	1.69E-02	1.72E-02	**1.71E-02**	1.33	U
236	4.18E-03	4.17E-03	**4.18E-03**	0.30	U
237	3.00E-03	2.78E-03	**2.89E-03**	5.51	Np
238	3.85E-01	4.04E-01	**3.94E-01**	3.43	U, Pu
239	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Pu
240	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Pu
241	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Pu, Am
242	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Pu, Am
243	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Pu, Cm
244	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Pu, Cm
245	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Cm
246	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Cm
247	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Cm, Bk
248	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Cm
249	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Cf
250	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Cf
251	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Cf
252	< 2.30E-03	<2.26E-03	**< 2.28E-03**	n/a	Cf

4.4 Tank 9H Salt Crystal Characterization and Dissolutions in DI-Water

The XRD analyses spectra on the two crystalline solid fractions (HTF-9-19-74 and HTF-9-19-75) from the two Tank 9H samples are shown in Figure 3. The major X-ray diffraction mineral phases identified in each of the crystals are given in Table 12. The solid crystalline phases of these Tank 9H samples contain the following four minerals in common: gibbsite ($Al(OH)_3$), nitratine ($NaNO_3$), sodium nitrite ($NaNO_2$) and quartz (SiO_2). However, the elemental analysis results for these solids (from water-based dissolution of the solids), as summarized in Table 12, shows that Tank 9H Sample HTF-9-19-74 crystal solids contain extra elements, which include Ca, Cu, Fe and Zn. These elementals were not identified in the other Tank 9H sample HTF-9-19-75. This may indicate that sample HTF-9-19-74 crystals contain extra minerals which may be too small in concentration to be seen in the XRD characterization of this solid sample. A more appropriate approach for accounting for all the elemental constituents of these solid fractions would however involve acid digestions using aqua regia or peroxide fusion methods to ensure that all the mineral contents, including SiO_2, are completely dissolved.

The dissolution of 2.528 grams of sample HTF 9-19-74 solid fraction needed 10.450 grams of DI-water (10.45 mL of DI-water), while 12.753 grams of DI-water was needed for the dissolution of 3.10 grams of sample HTF 9-19-75 solid fraction. These water-to-solid mixtures required for the dissolution of these Tank 9H variable sample solid fractions resulted in a DI-water dissolution phase ratio of 4.13 (10.45 mL/ 2.53 g) mL per gram of solid fraction for sample HTF 9-19-74 and 4.11 (12.75 mL/ 3.10 g) mL per gram of solid fraction for sample HTF 9-19-75. The dissolution of the solid fraction from sample HTF 9-19-74 in DI-water initially produced white precipitates, which eventually dissolved with the addition of more drops of DI-water. These DI-water dissolutions were performed at Shielded Cell's temperature which averaged 26 ± 1 °C and visual observations to confirm DI-water dissolutions of these solid fractions were made through the SRNL Shielded Cell windows.

Table 7 Corrosion Chemistry Analyses Results for Tank 9H Supernate (filtrate) HTF-9-19-75

Analyte	Analysis-1, M	Analysis-2 M	Average concentration, M	Standard deviation	%RSD N = 2
Fluoride, F^-	<1.13E-02	<1.15E-02	**<1.14E-02**	n/a	n/a
Formate, HCO_2^{1-}	<4.78E-03	<4.85E-03	**<4.82E-03**	n/a	n/a
Chloride, Cl^-	<6.07E-03	<6.16E-03	**<6.12E-03**	n/a	n/a
Nitrite, NO_2^-	1.67E+00	1.63E+00	**1.65E+00**	2.33E-02	1.41
Nitrate, NO_3^-	4.90E+00	4.79E+00	**4.84E+00**	7.47E-02	1.54
Phosphate, PO_4^{3-}	6.57E-03	6.51E-03	**6.54E-03**	4.69E-05	0.72
Sulfate, SO_4^{2-}	5.20E-02	5.16E-02	**5.18E-02**	2.74E-04	0.53
Oxalate, $C_2O_4^{2-}$	<2.45E-03	<2.48E-03	**<2.46E-03**	n/a	n/a
Bromide, Br^{1-}	<1.35E-02	<1.37E-02	**<1.36E-02**	n/a	n/a
Free Hydroxide, OH^-	6.80E-01	6.50E-01	**6.60E-01**	2.23E-02	3.41
Carbonate, CO_3^{2-}	7.55E-01	7.59E-01	**7.57E-01**	2.90E-03	0.38
$Al(OH)_4^{-1}$	3.87E-01	3.85E-01	**3.86E-01**	1.88E-03	0.49
Total carbon, gC/L	9.13E+00	9.19E+00	**9.16E+00**	4.38E-02	0.48
Inorganic carbon gC/L	9.06E+00	9.11E+00	**9.08E+00**	3.48E-02	0.38
Methyl mercury cation, mg/L	4.42E-01	5.05E-01	**4.74E-01**	*4.48E-02*	9.46
Total mercury, mg/L	2.35E+00	2.42E+00	**2.39E+00**	5.0E-02	2.17
pH (calculated)	n/a	n/a	**13.82**	n/a	n/a

Table 8 Elemental Analyses of Tank 9H Supernate (Filtrate): Variable Depth Sample HTF-9-19-75

Element	Analysis-1, mg/L	Analysis-2, mg/L	Average; Concentration, mg/L	Average; Concentration, M	%RSD, N = 2
Ag	<1.40E+00	<1.41E+00	**<1.41E+00**	**<1.30E-05**	n/a
Al	1.05E+04	1.04E+04	**1.04E+04**	**3.86E-01**	0.49
B	<1.93E+00	<1.94E+00	**<1.94E+00**	**<1.79E-04**	n/a
Ba	<1.03E+00	<1.03E+00	**<1.03E+00**	**<7.53E-06**	n/a
Be	<1.98E-01	<1.98E-01	**<1.98E-01**	**<2.20E-05**	n/a
Ca	<7.93E-01	<7.95E-01	**<7.94E-01**	**<1.98E-05**	n/a
Cd	<5.27E-01	<5.28E-01	**<5.28E-01**	**<4.69E-06**	n/a
Ce	<1.07E+01	<1.08E+01	**<1.08E+01**	**<7.67E-05**	n/a
Co	<1.15E+00	<1.16E+00	**<1.15E+00**	**<1.96E-05**	n/a
Cr	1.03E+02	1.05E+02	**1.04E+02**	**2.00E-03**	1.75
Cu	<2.13E+00	<2.13E+00	**<2.13E+00**	**<3.35E-05**	n/a
Fe	<2.88E+00	<2.88E+00	**<2.88E+00**	**<5.16E-05**	n/a
Gd	<1.89E+00	<1.90E+00	**<1.90E+00**	**<1.21E-05**	n/a
K	1.26E+03	1.28E+03	**1.27E+03**	**3.24E-02**	0.84
La	<1.12E+00	<1.12E+00	**<1.12E+00**	**<8.07E-06**	n/a
Li	<1.49E+00	<1.50E+00	**<1.50E+00**	**<2.15E-04**	n/a
Mg	<6.15E-02	<6.16E-02	**<6.16E-02**	**<2.53E-06**	n/a
Mn	<5.49E-01	<5.50E-01	**<5.50E-01**	**<1.00E-05**	n/a
Mo	4.99E+01	4.86E+01	**4.93E+01**	**5.13E-04**	1.79
Na	2.18E+05	2.20E+05	**2.19E+05**	**9.50E+00**	0.68
Ni	<4.39E+00	<4.40E+00	**<4.40E+00**	**<7.49E-05**	n/a
P	3.63E+02	3.70E+02	**3.66E+02**	**1.18E-02**	1.38
Pb	<7.07E+01	<7.09E+01	**<7.08E+01**	**<3.42E-04**	n/a
S	1.93E+03	2.44E+03	**2.19E+03**	**6.82E-02**	16.5
Sb	<1.09E+01	<1.09E+01	**<1.09E+01**	**<8.98E-05**	n/a
Si	<3.56E+00	<3.57E+00	**<3.56E+00**	**<1.27E-04**	n/a
Sn	<2.39E+01	<2.40E+01	**<2.40E+01**	**<2.02E-04**	n/a
Sr	<9.07E-02	<9.09E-02	**<9.08E-02**	**<1.04E-06**	n/a
Th	<7.87E+00	<7.88E+00	**<7.87E+00**	**<3.39E-05**	n/a
Ti	<2.14E-01	<2.14E-01	**<2.14E-01**	**<4.46E-06**	n/a
U	<3.36E+01	<3.37E+01	**<3.36E+01**	**<1.41E-04**	n/a
V	<3.76E-01	<3.76E-01	**<3.76E-01**	**<7.38E-06**	n/a
Zn	<1.30E+00	<1.30E+00	**<1.30E+00**	**<1.98E-05**	n/a
Zr	<3.67E-01	<3.68E-01	**<3.67E-01**	**<4.03E-06**	n/a

Table 9 Total cesium* in Tank 9H Variable Depth Sample HTF-9-19-75

Isotope	Average Concentration, mg/L	%RSD	Isotopic distribution, mass %	Moles	Mole fraction
Cs-133	21.0	0.11	74.4	1.58E-04	7.49E-01
Cs-134	0.005	1.29	0.02	3.45E-08	1.63E-04
Cs-135	2.15	0.49	7.61	1.59E-05	7.55E-02
Cs-137	5.08	0.62	18.0	3.71E-05	1.76E-01
Total Cs	**2.82E+01**	n/a	**100.0**	**2.11E-04**	**1.00E+00**

* Cesium isotopic concentrations are based on ICP-MS.

Table 10 Radiological results for Tank 9H Filtrate: Variable Depth Sample HTF-9-19-75

Analyte	Analysis-1 dpm/mL	Analysis-2 dpm/mL	Average dpm/mL	Average Ci/L of supernate	%RSD N = 2	One sigma % uncertainty
Total alpha	<1.68E+07	<1.59E+07	**<1.63E+07**	**<7.36E-03**	n/a	n/a
Total beta	1.29E+09	1.27E+09	**1.28E+09**	**5.74E-01**	1.10	10
Co-60	<2.74E+02	<2.62E+02	**<2.68E+02**	**<1.21E-07**	n/a	MDA
Sr-90	1.41E+05	1.60E+05	**1.51E+05**	**6.80E-05**	8.85	21.1
Y-90	1.41E+05	1.60E+05	**1.51E+05**	**6.80E-05**	8.85	21.1
Tc-99	3.57E+05	3.75E+05	**3.66E+05**	**1.65E-04**	3.39	6.51
Ru-106	<1.87E+03	<1.90E+03	**<1.88E+03**	**<8.48E-07**	n/a	MDA
Sb-125	<1.09E+03	<1.17E+03	**<1.13E+03**	**<5.08E-07**	n/a	MDA
Sb-126	3.41E+03	3.47E+03	**3.44E+03**	**1.55E-06**	4.22	5.0
Sn-126	3.41E+03	3.47E+03	**3.44E+03**	**1.55E-06**	4.22	5.0
I-129	<1.93E+01	<1.88E+01	**<1.91E+01**	**<8.58E-09**	n/a	MDA
Cs-134	<1.35E+05	<1.76E+05	**<1.56E+05**	**<7.01E-05**	n/a	MDA
Cs-137	1.09E+09	1.13E+09	**1.11E+09**	**5.01E-01**	*2.6*	5.0
Ba-137m	1.03E+09	1.07E+09	**1.05E+09**	**4.74E-01**	2.6	5
Ce-144	<1.94E+03	<1.92E+03	**<1.93E+03**	**<8.68E-07**	n/a	MDA
Eu-152	<7.55E+02	<7.69E+02	**<7.62E+02**	**<3.43E-07**	n/a	MDA
Eu-154	<5.33E+02	<5.37E+02	**<5.35E+02**	**<2.41E-07**	n/a	MDA
Eu-155	<7.13E+02	<8.40E+02	**<7.76E+02**	**<3.50E-07**	n/a	MDA
Th-232¥	< 1.09E-03	< 1.08E-03	**< 1.09E-03**	**< 4.90E-13**	n/a	MDA
U-233¥	5.42E+01	5.44E+01	**5.43E+01**	**2.45E-08**	0.23	10
U-234¥	3.70E+01	3.60E+01	**3.65E+01**	**1.64E-08**	1.83	10
U-235¥	8.55E-02	8.75E-02	**8.65E-02**	**3.90E-11**	1.61	10
U-236¥	6.20E-01	6.38E-01	**6.29E-01**	**2.83E-10**	2.03	10
Np-237¥	4.38E+00	4.65E+00	**4.52E+00**	**2.03E-09**	4.11	10
U-238¥	3.04E-01	2.96E-01	**3.00E-01**	**1.35E-10**	2.0	10
Pu-238	1.31E+04	1.46E+04	**1.38E+04**	**6.24E-06**	7.30	6.51
Pu-239/240	3.41E+02	1.47E+02	**2.44E+02**	**1.10E-07**	56.4	51.2
Pu-241	<3.01E+03	<4.10E+03	**<3.56E+03**	**<1.60E-06**	n/a	UL
Am-241	1.32E+02	<2.11E+02	**≤1.71E+02**	**≤7.72E-08**	n/a	88.1/MDA
Am-242m	<4.16E+01	<3.83E+01	**<4.00E+01**	**<1.80E-08**	n/a	MDA
Am-243	<1.50E+02	<1.72E+02	**<1.61E+02**	**<7.26E-08**	n/a	MDA
Cm-242	<3.43E+01	<3.17E+01	**<3.30E+01**	**<1.49E-08**	n/a	MDA
Cm-243	<5.12E+02	<5.46E+02	**<5.29E+02**	**<2.38E-07**	n/a	MDA
Cm-244	<1.14E+02	<8.79E+01	**<1.01E+02**	**<4.55E-08**	n/a	MDA
Cm-245	<4.19E+02	<4.51E+02	**<4.35E+02**	**<1.96E-07**	n/a	MDA
Cm-247	<4.92E+02	<5.37E+02	**<5.15E+02**	**<2.32E-07**	n/a	MDA
Cf-249	<5.40E+02	<5.84E+02	**<5.62E+02**	**<2.53E-07**	n/a	MDA
Cf-251	<5.01E+02	<5.51E+02	**<5.26E+02**	**<2.37E-07**	n/a	MDA

¥ Activities for Th-232, U isotopes and Np-237 are calculated from ICP-MS data

Table 11 Analyses of Tank 9H Supernate (Filtrate): Variable Depth Sample HTF-9-19-75

ICP-MS, m/z	Analysis-1, mg/L	Analysis-2, mg/L	Average, mg/L	%RSD, N = 2	Likely element (s)
59	< 4.48E-03	< 4.45E-03	**< 4.46E-03**	n/a	Co
82	1.83E-01	1.79E-01	**1.81E-01**	1.48	Se
84	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Sr
85	2.53E+00	2.53E+00	**2.53E+00**	0.22	Rb
86	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Sr
87	5.46E+00	5.48E+00	**5.47E+00**	0.25	Rb, Sr
88	7.40E-03	8.34E-03	**7.87E-03**	8.41	Sr
89	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Y
90	1.95E-02	1.82E-02	**1.88E-02**	4.78	Zr
91	1.66E-02	1.64E-02	**1.65E-02**	0.96	Zr
92	2.65E-01	2.62E-01	**2.64E-01**	0.71	Zr, Mo
93	1.95E-02	1.94E-02	**1.95E-02**	0.49	Nb
94	1.73E-01	1.73E-01	**1.73E-01**	0.23	Nb, Mo
95	1.42E+01	1.43E+01	**1.43E+01**	0.39	Mo
96	3.05E-01	3.02E-01	**3.03E-01**	0.55	Ru, Zr, Mo
97	1.31E+01	1.32E+01	**1.31E+01**	0.47	Mo, Tc
98	1.32E+01	1.31E+01	**1.32E+01**	0.67	Ru, Mo, Tc
99	8.98E+00	8.93E+00	**8.95E+00**	0.40	Tc, Ru
100	1.36E+01	1.36E+01	**1.36E+01**	0.20	Ru, Mo
101	7.46E-01	7.37E-01	**7.41E-01**	0.84	Ru
102	6.95E-01	6.87E-01	**6.91E-01**	0.83	Ru, Pd
103	1.86E+00	1.81E+00	**1.83E+00**	1.90	Rh
104	3.43E-01	3.35E-01	**3.39E-01**	1.86	Ru, Pd
105	7.08E-02	5.99E-02	**6.53E-02**	11.9	Pd
106	5.28E-02	4.37E-02	**4.82E-02**	13.4	Pd, Cd
107	3.11E-02	2.56E-02	**2.84E-02**	13.8	Ag
108	< 1.34E-02	< 1.33E-02	**< 1.34E-02**	n/a	Pd, Cd
109	< 8.96E-03	< 8.90E-03	**< 8.93E-03**	n/a	Ag
110	< 6.72E-03	< 6.67E-03	**< 6.70E-03**	n/a	Pd, Cd
111	< 6.72E-03	< 6.67E-03	**< 6.70E-03**	n/a	Cd
112	2.81E-02	2.94E-02	**2.87E-02**	3.15	Sn, Cd
113	< 6.72E-03	< 6.67E-03	**< 6.70E-03**	n/a	In, Cd
114	2.50E-02	2.53E-02	**2.52E-02**	0.74	Sn, Cd
116	4.11E-01	4.02E-01	**4.07E-01**	1.61	Sn, Cd
117	2.79E-01	2.77E-01	**2.78E-01**	0.46	Sn
118	8.07E-01	7.96E-01	**8.02E-01**	0.97	Sn
119	2.51E-01	2.55E-01	**2.53E-01**	0.90	Sn
120	1.07E+00	1.08E+00	**1.07E+00**	0.71	Sn
121	1.79E-02	1.78E-02	**1.78E-02**	0.26	Sb
122	2.87E-01	2.80E-01	**2.83E-01**	1.76	Te, Sn
123	1.10E-02	1.03E-02	**1.07E-02**	4.49	Sb, Te
124	4.03E-01	4.04E-01	**4.04E-01**	0.23	Te, Sn
125	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Sb, Te
126	1.26E+00	1.24E+00	**1.25E+00**	1.46	Te
128	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Te
130	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Te
133	2.11E+01	2.10E+01	**2.10E+01**	0.11	Cs
134	4.66E-03	4.57E-03	**4.62E-03**	1.29	Cs, Ba

Table 11 continued Analyses of Tank 9H Supernate (Filtrate): Variable Depth Sample HTF-9-19-75

ICP-MS, m/z	Analysis-1, mg/L	Analysis, mg/L	Average, mg/L	%RSD, N = 2	Likely element (s)
135	2.16E+00	2.15E+00	**2.15E+00**	0.49	Ba, Cs
136	< 2.24E-03	< 2.22E-03	**< 2.23E-03**		Ce, Ba
137	5.06E+00	5.10E+00	**5.08E+00**	0.62	Cs, Ba, La
138	1.47E-02	1.70E-02	**1.58E-02**	10.3	Ba, La, Ce
139	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	La
140	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Ce
141	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Pr
142	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Nd, Ce
143	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Nd., Pm
144	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Nd, Sm, Pm
145	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Nd, Pm
146	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Nd, Sm
147	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Sm
148	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Nd, Gd, Sm
149	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Sm
150	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Nd, Gd, Sm, Eu
151	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Eu
152	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Gd, Sm, Eu
153	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Eu
154	5.04E-03	5.16E-03	**5.10E-03**	1.57	Gd, Sm, Eu, Dy
155	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Gd
156	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Gd, Dy
157	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Gd, Tb
158	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Gd, Dy, Tb
159	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Tb
160	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Gd, Dy
161	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Dy
162	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Dy, Er
163	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Dy, Ho
164	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Dy, Er
165	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Ho
166	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Er, Ho
167	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Er
168	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Er, Yb
169	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Tm
170	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Er, Yb
171	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Yb
172	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Yb
173	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Yb
174	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Yb, Hf
175	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Lu
176	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Lu, Hf, Yb
177	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Hf
178	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Hf
179	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Hf
180	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Hf, W, Ta
181	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Ta
182	1.59E-01	1.55E-01	**1.57E-01**	1.97	Hf, W

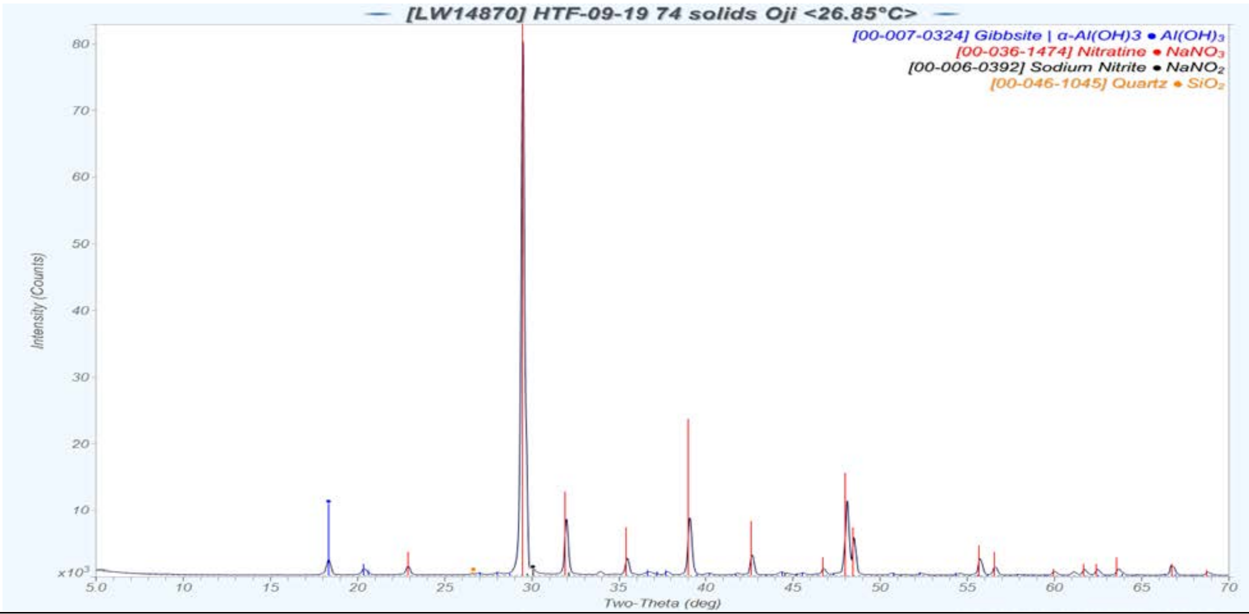
Table 11 Continued. Mass Spectral Analyses of Tank 9H Filtrate: Variable Depth Sample HTF-9-19-75

ICP-MS, m/z	Analysis-1, mg/L	Analysis-2, mg/L	Average, mg/L	%RSD, N = 2	Likely element (s)
183	8.52E-02	8.60E-02	**8.56E-02**	0.66	W
184	1.91E-01	1.89E-01	**1.90E-01**	0.82	W
185	2.71E-03	2.79E-03	**2.75E-03**	1.90	Re
186	1.71E-01	1.77E-01	**1.74E-01**	2.23	Os, W
187	7.14E-03	6.98E-03	**7.06E-03**	1.59	Re, Os
188	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Os
189	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Os
191	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Ir
193	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Ir, Pt
194	< 4.48E-03	< 4.45E-03	**< 4.46E-03**	n/a	Pt
195	< 4.48E-03	< 4.45E-03	**< 4.46E-03**	n/a	Pt
196	6.06E-03	6.99E-03	**6.53E-03**	10.1	Hg, Pt
198	2.56E-01	2.67E-01	**2.62E-01**	2.90	Hg, Pt
203	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Tl
204	2.58E-01	2.49E-01	**2.54E-01**	2.53	Pb, Hg
205	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Tl
206	1.77E+00	1.75E+00	**1.76E+00**	0.98	Pb
207	1.51E+00	1.51E+00	**1.51E+00**	0.22	Pb
208	3.65E+00	3.69E+00	**3.67E+00**	0.77	Pb
229	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Th
230	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Th
232	< 4.48E-03	< 4.45E-03	**< 4.46E-03**	n/a	Th, U
233	2.52E-03	2.53E-03	**2.53E-03**	0.23	U
234	2.66E-03	2.60E-03	**2.63E-03**	1.83	U
235	1.78E-02	1.82E-02	**1.80E-02**	1.61	U
236	4.32E-03	4.44E-03	**4.38E-03**	2.03	U
237	2.80E-03	2.97E-03	**2.89E-03**	4.11	Np
238	4.08E-01	3.96E-01	**4.02E-01**	2.00	U, Pu
239	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Pu
240	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Pu
241	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Pu, Am
242	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Pu, Am
243	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Pu, Cm
244	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Pu, Cm
245	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Cm
246	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Cm
247	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Cm, Bk
248	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Cm
249	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Cf
250	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Cf
251	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Cf
252	< 2.24E-03	< 2.22E-03	**< 2.23E-03**	n/a	Cf,

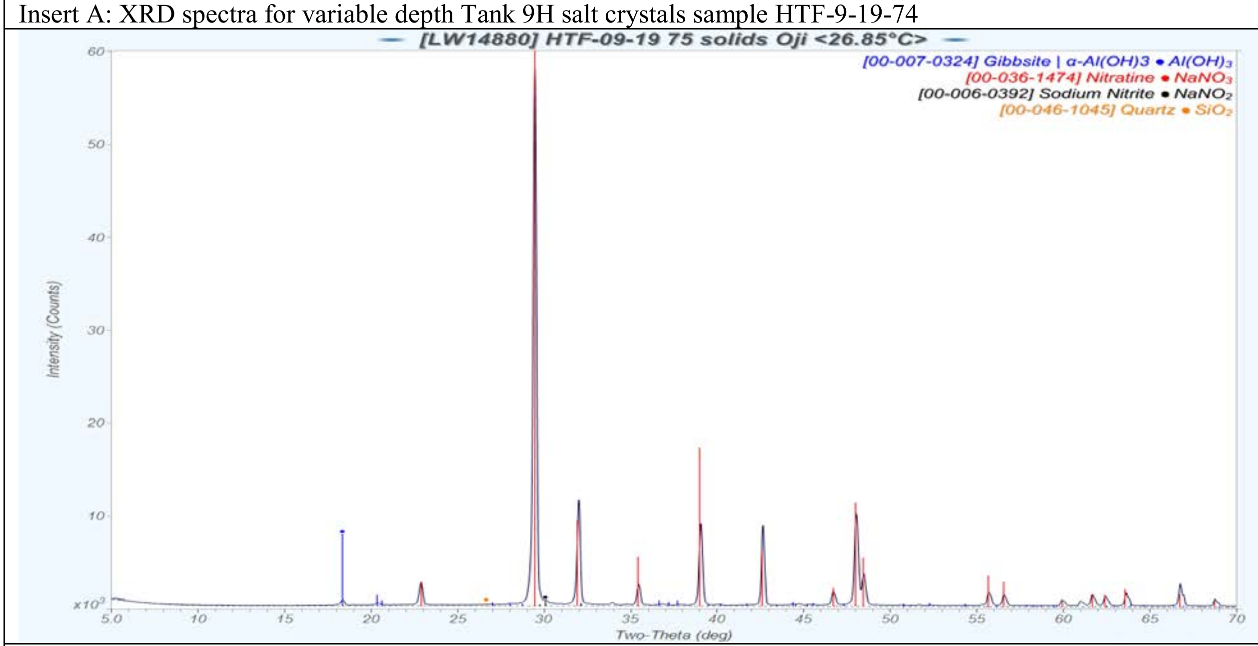
Table 12 Tank 9H dissolved salt crystal elementals and mineralogy

Elemental/mineral	Sample **HTF-9-19-74**		**Average**	Sample **HTF-9-19-75**		**Average**
	mg/g solid crystal		**mg/g solid crystal**	mg/g solid crystal		**mg/g solid crystal**
	Run-1	Run-2		Run-1	Run-2	
Al	26.1	0.903	**13.50**	0.274	0.274	**0.274**
Ca	0.272	0.007	**0.14**	<0.007	<0.007	**<0.007**
Cu	0.073	0.004	**0.039**	<0.003	<0.003	**<0.003**
Fe	0.229	0.036	**0.13**	<0.034	<0.034	**<0.034**
Na	228	228	**228**	225	225	**225**
Zn	<0.003	0.085	**≤0.044**	<0.003	<0.003	**<0.003**

	Sample **HTF-9-19-74 XRD mineralogy**	Sample **HTF-9-19-75 XRD mineralogy**
Gibbsite ($Al(OH)_3$)	✓	✓
Nitratine ($NaNO_3$)	✓	✓
Sodium nitrite ($NaNO_2$)	✓	✓
Quartz (SiO_2)	✓	✓



Insert A: XRD spectra for variable depth Tank 9H salt crystals sample HTF-9-19-74



Insert B: XRD spectra for variable depth Tank 9H salt crystals sample HTF-9-19-75

Figure 3. XRD spectra for Tank 9H salt crystals-Samples HTF-9-19-74 and HTF-9-19-75

5.0 Conclusions and Recommendations

On July 18, 2019, Savannah River Remediation Engineering (SRR-E) delivered two variable depth Tank 9H salt solution samples, identified as samples HTF-9-19-74 and HTF-9-19-75 (combined total sample volume of about 400 mL) to the Savannah River National Laboratory (SRNL) for characterizations.

This report presents the analytical characterization results for the Tank 9H salt solution variable depth sample HTF-9-19-74 and HTF-9-19-75 filtrates and solids and contains mainly the filtrate analytical results for anions, elementals, mass spectral data, total cesium characterization results and select radionuclide results, along with the XRD and elemental characterization of the solid fractions from the liquid /solid separation of the variable depth salt solutions. Most of these analyses were performed in duplicate with the percent relative standard deviation (%RSD), a measure of the variability between each duplicate data set expressed as a percentage, used to estimate the analytical result quality for each analyte.

The measured average sodium concentrations in these two variable depth filtrates [9.73 (0.45 %RSD) M for sample HTF-9-19-74 and 9.50 (0.68 %RSD) M for sample HTF-9-19-75] are abnormally high when compared to the average sodium concentration in the Tank Farm supernates which average 5.6 M. These high sodium concentrations in the filtrate accounts for the abnormally high density of the filtrate solutions; densities of these two variable depth Tank 9H filtrate samples are 1.436 ± 0.002 for sample HTF-9-19-74 and 1.433 ± 0.003 g/mL for sample HTF-9-19-75. On the other hand, the total mercury concentrations in these two filtrate samples, which averaged less than 3 mg/L, is in line with expectations when compare with Tank 10 analysis result for total mercury [3].

A liquid/solid separation, designed to generate the filtrate solutions used for the characterizations, also produced off-white colored solid materials. The solid fractions from the solid/liquid separations were found to dissolve (based on visual observation) in deionized and distilled water with simple mechanical agitation of the water/solid mixtures. The resulting solid crystalline fractions from the variable depth samples contain the same four minerals identified by X-ray diffraction: gibbsite ($Al(OH)_3$), nitratine ($NaNO_3$), sodium nitrite ($NaNO_2$) and quartz (SiO_2). The laboratory measured water-to-solids fraction mixtures (phase ratios) required for dissolution of these Tank 9H variable depth sample solid fractions were 4.13 mL/gram of sample HTF 9-19-74 solid fraction and 4.11 mL/gram of sample HTF 9-19-75 solid fraction. Bulk dissolution of these solids in the Tank Farm would require the use of inhibited water (0.01 M NaOH and 0.011 M $NaNO_3$) to ensure the dissolution of gibbsite mineral found in the solid fractions.

Elemental analysis results for the Tank 9H solid fraction shows that Tank 9H sample HTF-9-19-74 crystal solids contain extra elements which included Ca, Cu, Fe and Zn. These elements were not identified in the other Tank 9H sample HTF-9-19-75 solids. This may indicate that sample HTF-9-19-74 solids fraction contains extra minerals which may be too small in concentration to be identified by X-ray diffraction characterization of this solid sample phase.

A summary of the average analytical results for other analytes based on one sigma analytical uncertainty for the Tank 9H variable depth samples includes the following.

The measured cesium-137 activity in the supernate samples HTF-9-19-74 and HTF-9-19-75 filtrates averaged 1.06E+09 (0.7 %RSD) and 1.11E+09 (2.6 % RSD) dpm/mL, respectively.

Cesium-137 accounts for most of mass 137 in both samples and the mass contributions from other elements like Ba and La with mass 137 are negligible.

Both Cs-133 and Cs-137 dominate the cesium isotopic mass distribution in both Tank 9H variable depth filtrate samples. The two cesium isotopes, Cs-133 and Cs-137, account for an average of 74% and 18% of the total cesium mass distribution in the two variable depth samples, respectively. Cs-134 and Cs-135 account for the remaining 8% isotopic mass % distribution in each variable depth filtrate sample.

The gamma-emitting radionuclides in the Tank 9H variable depth filtrate sample HTF-9-19-74 are Ba-137^{m} and Co-60 at an average activity concentration of 4.53E-01 (0.70 %RSD) Ci/L and <1.40E-07 (MDA) Ci/L, respectively. The Ba-137^{m} and Co-60 activities for sample HTF-9-19-75 were 4.74E-01 (2.60 %RSD) Ci/L and <1.21E-07 (MDA) Ci/L, respectively.

The primary beta-emitting radionuclides in the Tank 9H variable depth sample filtrates are Sr-90 [5.92E-05 (2.63 %RSD) Ci/L], Y-90 [5.92E-05 (2.63 %RSD) Ci/L] and Cs-137 [4.78E-01 (0.70 %RSD) Ci/L]. for sample HTF-9-19-74. The activities of these beta-emitting radionuclides Sr-90, Y-90 and Cs-137 are 6.80E-05 (8.85 %RSD) Ci/L, 6.80E-05 (8.85 %RSD) Ci/L and 5.01E-01 (2.6 %RSD) Ci/L for sample HTF-9-19-75, respectively.

The total alpha activity for the two filtrate samples were all less than detectable values (upper limits) because of possible spectral interferences. Total alpha activities averaged <7.38E-03 Ci/L (<1.64E+07 dpm/mL) for sample HTF-9-19-74 and <7.36E-03 Ci/L (<1.63E+07 dpm/mL) for sample HTF-9-19-75. However, a summation of the primary alpha emitting radionuclide activities (Pu-238, Pu-239/240, Am-241 and Cm-244) gives one a better upper limit estimate for total alpha activities in the two sample filtrates. These summed total alpha activity values for these samples are ≤6.79E-06 Ci/L and ≤6.47E-06 Ci/L for samples HTF-9-19-74 and HTF-9-19-75, respectively. These values, as expected, are about three orders of magnitude lower than the directly measured values for total alpha.

The total beta in the two filtrate samples were above instrument detection limits and average 5.66E-01Ci/L [1.26E+09 (0.69 %RSD) dpm/mL] for sample HTF-9-19-74 and 5.74E-01Ci/L [1.28E+09 (1.10 %RSD) dpm/mL] for sample HTF-9-19-75.

The average uranium enrichment for these sample filtrates from HTF-9-19-74 and HTF-9-19-75 were 4.1 and 4.2 percent, respectively. The total uranium concentrations in these supernates are 4.21E-01 and 4.30E-01 mg/L, respectively. Taking into consideration the one sigma analytical uncertainty of about 10%, these measurements are quantitively the same in magnitude for these two filtrate samples.

The concentration of Tc-99 in the two filtrates averaged 1.74E-04 Ci/L [3.86E+05 (1.20 % RSD) dpm/mL] for sample HTF-9-19-74 and 1.65E-04 Ci/L [3.66E+05 (3.39 % RSD) dpm/mL] for sample HTF-9-19-75.

Iodine-129 activities which averaged <1.14E-08 Ci/L (<2.53E+01 dpm/mL) for sample HTF-9-19-74 and <8.58E-09 Ci/L (<1.91E+01 dpm/mL) for sample HTF-9-19-75 were all below instrument detection limits.

Except for Am-241 measurements for sample HTF-9-19-75 filtrate, all the Am/Cm measurements for the Tank 9H filtrates were below instrument detection limits. In the Am-241 duplicate measurement results for sample HTF-9-19-75 filtrate, one of the analysis values (1.32E+02 dpm/mL) was above the instrument detection limit and the other was below instrument detection limit (<2.11E+02 dpm/mL).

In both filtrate samples, the only cesium-removed gamma analytes with detections above instrument detection limit were Sb-126 and Sn-126. The equilibrium values for these radionuclides averaged 1.55E-06 Ci/L [3.43E+03 (3.95 %RSD) dpm/mL] for sample HTF-9-19-74 and 1.55E-06 Ci/L [3.44E+03 (4.22 %RSD) dpm/mL] for sample HTF-9-19-75.

The concentrations of total carbon and inorganic carbon in the two variable Tank 9H filtrates are essential equal, which means there is no measurable organic carbon in either of the Tank 9H variable sample filtrates.

Nitrate and nitrite were the two predominant anions in the two Tank 9H variable depth samples. The nitrate and nitrite concentrations averaged 5.23 (1.81 %RSD) M and 1.67 (1.71 %RSD) M for sample HTF-9-19-74, and 4.84 (1.54 %RSD) M and 1.65 (1.41 %RSD) M for sample HTF-9-19-75, respectively.

Another anion with measurement above instrument detection limits for these two Tank 9H filtrate (HTF-9-19-74 and HTF-9-19-75) samples, was sulfate which averaged 5.47E-02 (1.37 %RSD) M and 5.18E-02 (0.53 %RSD) M, respectively.

A measurable quantity of phosphate anion was also detected in sample HTF-9-19-75 at 6.54E-03 (0.72 %RSD) M, however, phosphate concentration in sample HTF-9-19-74 was below the instrument detection limit (<2.34E-02 M). The concentration of other anions besides carbonate, aluminate and free hydroxide, in these two variable depth samples were below instrument detection limits.

Total mercury and methyl mercury concentrations in the two Tank 9H filtrate samples averaged 3.38 and 5.64E-01 mg/L for sample HTF-9-19-74 and 2.38 and 4.74E-01 mg/L for sample HTF-9-19-75, respectively.

Free hydroxide, carbonate and aluminate concentrations averaged 6.60E-01 (11.0 %RSD) M, 8.20E-01 (2.62 %RSD) M and 3.45E-01 (0.43 %RSD) M for sample HTF-9-19-74 and 6.60E-01 (3.41 %RSD) M, 7.57E-01 (0.38 %RSD) M and 3.86E-01 (0.49 %RSD) M for sample HTF-9-19-75, respectively.

Sodium is the predominant cation in the Tank 9H variable depth sample filtrates. Sodium concentration in sample HTF-9-19-74 averaged 2.24E+05 (0.45 %RSD) mg/L or 9.73 M and averaged 2.19E+05 (0.68 %RSD) mg/L or 9.50 M in sample HTF-9-19-75.

Overall, there are no significant measurable differences in the concentrations of the analytes in the two Tank 9H variable depth samples. The minor analytical differences fall within the analytical uncertainties of the different analytical methods.

6.0 QUALITY ASSURANCE

The Task Technical and Quality Assurance Plan details the planned activities and associated quality assurance implementing procedures for the characterization of Tank 9H salt solution (TTQAP, SRNL-RP-2019-00463, Rev. 0, July 11, 2019) [2]. The documents referenced in the TTQAP include the following: L. N. Oji: ELN: L5575-00080-11 (Electronic Notebook (Production); SRNL, Aiken, SC 29808 (2014) and in various AD notebooks contain the analytical data. Other relevant QA documents include the Technical Task Request (X-TTR-00088, Rev 0, June. 13, 2019) [1].

Requirements for performing reviews of technical reports and the extent of review are established in manual E7 2.60. This document, including all calculations was reviewed by Design Verification by Document Review [4, 5]. SRNL documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2.

The TTR requested functional classification ("Safety Class") and the report, calculations and technical memoranda issued in this Tank 9H variable sample characterization effort have received technical review by design verification (E7 Manual, Procedure 2.60, section 5.3). The experimental work, the analyses, and peer check all comply with the customer quality assurance (QA) requirements.

7.0 REFERENCES

1. Fellinger T. L., "Analysis of Tank 9 Salt Solution," X-TTR-H-00088, Rev. 0, June 13, 2019.

2. L. N. Oji, "Task Technical and Quality Assurance Plan for the Analysis of Tank 9H Salt solution Supernatant" SRNL-RP-2019-00463, Rev. 0, July 11, 2019.

3. S. H. Reboul, "Characterization of the March 2017 Tank 10 Surface Sample (Combination of HTF-10-17-30 and HTF-10-17-31) and Variable Depth Sample (Combination of HTF-10-17-32 and HTF-10-17-33)" SRNL-STI-2017-00392, Rev. 0, July 2017.

4 "Technical Reviews", Manual E7, Procedure 2.60, Revision 17, August 25, 2016.

5 "Savannah River National Laboratory Technical Report Design Check Guidelines", WSRC-IM-2002-00011, Revision 2, August 2004.

Appendix A: Summary of Analytical Methods

Inductively Coupled Plasma–Atomic Emission Spectroscopy (ICP-ES)

Samples are diluted as necessary to bring analytes within the instrument range. A scandium internal standard is added to all samples after dilution at a concentration of 2 mg/L. The instrument is calibrated daily with a blank and two standards: 5 and 10 mg/L NIST traceable multi-element standards in dilute acid. Background and internal standard correction were applied to the results.

Ion Chromatography for Anions (IC-Anions)

For IC Anions, samples were diluted with a carbonate/bicarbonate diluent as necessary to bring analytes to within instrument calibration. A 3-point calibration curve is run daily on the instrument with concentrations of 10, 25 and 50 µg/mL.

Total mercury was analyzed by DMA.

With direct mercury analysis (DMA) method for total mercury analysis, controlled heating in an oxygenated decomposition furnace is used to liberate mercury from solid and aqueous samples in the instrument. The sample is dried and then thermally and chemically decomposed within the decomposition furnace. The decomposition products are carried by flowing oxygen to the catalytic section of the furnace. With the completion of oxidation, halogens and nitrogen/sulfur oxides are trapped. The remaining decomposition products are then carried to an amalgamator that selectively traps mercury. After the system is flushed with oxygen to remove any remaining gases or decomposition products, the amalgamator is rapidly heated, releasing mercury vapor. Flowing oxygen carries the mercury vapor through absorbance cells positioned in the light path of a single wavelength atomic absorption spectrophotometer. Absorbance (peak height or peak area) is measured at 253.7 nm as a function of mercury concentration.

The typical working range for this method is 0.05 - 600 ng. The mercury vapor is first carried through a long pathlength absorbance cell and then a short pathlength absorbance cell. (The lengths of the first cell and the second cell are in a ratio of 10:1 or another appropriate ratio.). The same quantity of mercury is measured twice, using two different sensitivities, resulting in a dynamic range that spans at least four orders of magnitude. The instrument detection limit (IDL) for this method is 0.01 ng of total mercury.

Gas chromatography/atomic fluorescence spectroscopy: Methyl and ethyl mercury analysis

Methylmercury and ethylmercury are analytically separated and quantified from aqueous samples by purge and trap (P&T) gas chromatography (GC) cold-vapor atomic fluorescence spectroscopy (CVAFS). The methyl- or ethylmercury species are first derivatized using sodium tetraethylborate or sodium tetrapropylborate, respectively, to induce volatility prior to sample purge using nitrogen. The purged vapor enters a GC module where the various mercury species are separated isothermally prior to ballistic pyrolysis to convert all mercury species to fully reduced elemental mercury. The ground-state mercury travels to the CVAFS for detection.

Inductively Coupled Plasma – Mass Spectroscopy (ICP-MS)

Samples are diluted as necessary to bring analyte concentrations within the instrument range. An internal standard with bismuth and indium is added to all samples after dilution. The instrument is calibrated daily with a blank and a minimum of four calibration standards that are NIST traceable multi-element standards in dilute acid. Background and internal standard correction were applied to the results.

Gross Alpha/Gross Beta

Aliquots of the Tank 9H supernatant filtrates were added to liquid scintillation cocktail and analyzed for gross alpha and gross beta activity using liquid scintillation analysis. Alpha/beta spillover was determined for each aliquot analyzed, and subsequently used for accurately determining alpha and beta activity, via the addition of a known amount of plutonium to an identical aliquot of each sample.

Cs-137, Cs-134

Aliquots of the Tank 9H supernatant filtrates were analyzed by coaxial high purity germanium gamma-ray spectrophotometers to measure Cs-137 and Cs-134. Laboratory reagent blanks were run as controls.

Sr-90

Aliquots of the Tank 9H supernatant filtrate were spiked with an elemental strontium carrier. The strontium species were extracted from the matrix using a crown-ether-based solid phase extractant. Sr-90 concentrations were measured by liquid scintillation analysis. Elemental strontium carrier yields were measured by neutron activation analysis and were used to correct the Sr-90 analyses for any strontium losses from the radiochemical separations. Laboratory reagent blanks and a Sr-90 standard were run as controls.

Co-60, Am-241 (Cs-removed gamma analysis)

Aliquots of the Tank 9H supernatant filtrate were subjected to a Cs-removal process utilizing Bio Rad AMP-1 resin. The Cs-removed solutions were analyzed by coaxial high purity germanium spectrophotometers to measure the gamma-emitting radionuclides. Aliquots of the Tank 9H supernatant filtrate were also analyzed for Co-60 and Am-241 following the same protocols; no additional preparations were required.

Pu-238, 239/240, 241

Aliquots of the Tank 9H supernatant filtrates were spiked with Pu-236 tracer. The plutonium was extracted from the matrix using thenoyltrifluoroacetone (TTA) following a series of oxidation-state adjustments. The TTA extracts were mounted on stainless steel counting plates and counted for Pu-238 and Pu-239/240 using passivated, implanted, planar silicon (PIPs) detectors. Each separation was traced based on the Pu-236 recovery. Aliquots of sample were also subjected to Cs-removal with Bio-Rad Ammonium Molybdophosphate (AMP) resin and extracted using TEVA columns (TEVA is a brand name for one of Eichrom's resins). The Pu-containing extracts were measured by liquid scintillation analysis to determine Pu-241 concentrations. Shielded cell reagent blank and laboratory reagent blanks and a Pu-238 standard were run as controls.

Am-242m, 243, Cm-243, 244, 245, 247, Cf-249, 251

Americium, curium and californium species were extracted from aliquots of the Tank 9H supernatant filtrate using a CMPO/tributyl phosphate commercial resin based solid phase extractant and purified further with a proprietary commercial resin called HDEHP based solid phase extractant. Am-241, 243, Cm-243, 245, 247, Cf-249 and 251 concentrations were measured using low energy photon/x-ray, thin-windowed, semi-planar high purity germanium spectrometers. Am-242m, Cm-242, and 244 concentrations were measured using passivated, implanted, planar silicon (PIPS) alpha spectrometers. Cm-245, 247 and 248 ratios to Am-241 were measured using ICP-MS and were applied to the previously quantified Am-241. Am-241 quantities had been measured from the cesium removed gamma analyses, Am, Cm, and Cf results were traced with the Am-241 present in the sample matrix. Shielded cell reagent blank, ARG, and laboratory reagent blanks were also run as controls.

Tc-99

Aliquots of Tank 9H filtrate were oxidized and spiked with Tc-99^{m} that had been extracted from molybdenum which had been activated in SRNL's Cf-252 neutron activation analysis facility. The technetium species were extracted from the matrix using an Aliquat-336 based solid phase extractant.

Tc-99 concentrations were measured by liquid scintillation analysis. Tc-99^{m} yields were measured with a NaI-well gamma spectrometer and were used to correct the Tc-99 analyses for any technetium losses from the radiochemical separations.

Densities:

Density measurements were conducted at a temperature of ~26 °C. This temperature was governed by the Shielded Cells conditions at the time of the measurements. Densities were measured using weight calibrated balances and 2.0 mL volume-calibrated glass test tubes. Three individual Tank 9H supernate filtrate aliquots were utilized in the measurements. The supernatant was generated as a filtrate by passing slurry through a 0.45 µm filtration membrane (note that this generation method was utilized for all of the supernatant analyses and not just those used for determining density). The density of a deionized water standard was determined along with the slurry and supernatant determinations, to demonstrate measurement accuracy.

Elemental Analysis of Supernatant:

In preparation for the elemental analyses (prior to AD submittal), two supernatant aliquots filtrates were each diluted by a factor of ~21 (on a volume basis), using ~3 M HNO_3. The use of the ~3 M HNO_3 diluent was considered beneficial for minimizing loss of constituents through sorption to the walls of the sample submittal vessels and through potential precipitation reactions. An applicable "acid blank" and a multielement standard were submitted along with the acidified/diluted supernatant, for quality assurance purposes. ICP-AES and DMA were performed on the acidified/diluted supernatant aliquots, to quantify routine elemental constituents, and mercury, respectively. Acid diluted supernatant aliquots filtrates were diluted in the Shielded Cells by a factor of ~80 for methyl mercury and ethylmercury analysis by gas chromatography/atomic fluorescence spectroscopy.

Anions in the Supernatant:

In preparation for the anion analyses (prior to AD submittal), two supernatant aliquots were each diluted by a factor of ~21 (on a volume basis), using de-ionized water. IC was performed on the diluted supernatant aliquots, to quantify bromide, chloride, fluoride, formate, nitrate, nitrite, oxalate, phosphate, and sulfate. Total inorganic carbon (TIC) analyses were performed to quantify carbonate, and base titration analyses were performed to quantify free hydroxide. Aluminate was quantified based on the ICP-AES supernatant aluminum concentration, assuming 100% of the aluminum was present as aluminate. Dilution-correction of the results was performed prior to reporting.

Select Radioisotopes in the Supernatant filtrate:

The same acid dilution method that was used for the supernatant elemental analyses was utilized for preparing the supernatant aliquots for the select radioisotope analyses (two supernatant filtrate aliquots acidified and diluted using ~3.0 M nitric acid plus a ~3.0 M acid blank for quality assurance purposes). Gamma spectroscopy was performed on the acidified/diluted supernatant aliquots to quantify Cs-137, Cs-134. Cesium removed gamma and ICP-MS was performed to quantify for Th-232, U-233, U-234, U-235, U-236, U-238 and Np-237 using the acid diluted supernatant filtrates. Dilution correction of the results was performed prior to reporting.

Appendix B: Tank 9H Characterization; AD Tracking Numbers*

Analytes	Method (s)	SRNL AD Tracking Number (LIMS):
Anions	IC	LW14453, LW14454, LW14521, LW14522
Free-OH	Free-OH	LW14453, LW14454, LW14524, LW14525
TIC/TOC	TIC/TOC	LW14456, LW14457, LW14524, LW14525
Elemental	ICP-ES	LW14502, LW14503, LW14431, LW14432
Select Elements (Ag, Th)	ICP-MS	LW14505, LW14506, LW14432, LW14434
Hg	DMA	LW14515, LW14516, LW14444, LW14445
MeHg and Ethyl mercury	GC-AFS	LW14529, LW14530, LW14447, LW14448
Total Alpha	Rad Screen (LSC)	LW14450, LW14451, LW14518, LW14519
Non-volatile Beta	Rad Screen (LSC)	LW14450, LW14451, LW14518, LW14519
Sr-90/Y-90	Sr90	LW14442, LW14443, LW14513, LW14514
Tc-99	LSC	LW14438, LW14439, LW14509, LW14510
I-129	I-129	LW14442, LW14443, LW14527, LW14528
Pu-238	Pu-238/241	LW14436, LW14437, LW14507, LW14508
Pu-241	Pu-238/241	LW14436, LW14437, LW14507, LW14508
Pu-239/ Pu-240	Pu-TTA	LW14436, LW14437, LW14507, LW14508
Cs-134	GAMMA SPEC	LW14438, LW14439, LW14509, LW14510
Cs-137	GAMMA SPEC	LW14438, LW14439, LW14509, LW14510
Ba-137^m	Calculated value	LW14438, LW14439, LW14509, LW14510
U-233	ICP-MS	LW14505, LW14506, LW14432, LW14434
U-234	ICP-MS	LW14505, LW14506, LW14432, LW14434
U-235	ICP-MS	LW14505, LW14506, LW14432, LW14434
U-236	ICP-MS	LW14505, LW14506, LW14432, LW14434
U-238	ICP-MS	LW14505, LW14506, LW14432, LW14434
Np-237	ICP-MS	LW14505, LW14506, LW14432, LW14434
Co-60	GAMMA SPEC Cs REMOVED	LW14440, LW14441, LW 14511, LW 14512
Sb-126	GAMMA SPEC Cs REMOVED	LW14440, LW14441, LW 14511, LW 14512
Ce-144	GAMMA SPEC Cs REMOVED	LW14440, LW14441, LW 14511, LW 14512
Pr-144	GAMMA SPEC Cs REMOVED	LW14440, LW14441, LW 14511, LW 14512
Eu-154	GAMMA SPEC Cs REMOVED	LW14440, LW14441, LW 14511, LW 14512
Eu-155	GAMMA SPEC Cs REMOVED	LW14440, LW14441, LW 14511, LW 14512
Am-241	GAMMA SPEC Cs REMOVED	LW14440, LW14441, LW 14511, LW 14512
Cm-244	Am/Cm	LW 14442, LW 14443, LW 14513, LW 14514

*Project: IDs: LW-AD-PROJ-190722-1 and LW-AD-PROJ-190725-3

Appendix C: Tank 9H Uranium Characterization.

Tank 9H Sample HTF-9-19-74

Uranium Isotopes	Run-1, mg/L	Run-2, mg/L	Average, mg/L	%RSD
U-233	2.55E-03	2.54E-03	2.55E-03	0.17
U-234	2.51E-03	2.71E-03	2.61E-03	5.43
U-235	1.69E-02	1.72E-02	1.71E-02	1.33
U-236	4.18E-03	4.17E-03	4.18E-03	0.30
U-238	3.85E-01	4.04E-01	3.94E-01	3.43
Total Uranium			**4.21E-01**	
Uranium-235 % enrichment			4.1	

Tank 9H Sample HTF-9-19-75

Uranium Isotopes	Run-1, mg/L	Run-2, mg/L	Average, mg/L	%RSD
U-233	2.52E-03	2.53E-03	**2.53E-03**	0.23
U-234	2.66E-03	2.60E-03	**2.63E-03**	1.83
U-235	1.78E-02	1.82E-02	**1.80E-02**	1.61
U-236	4.32E-03	4.44E-03	**4.38E-03**	2.03
U-238	4.08E-01	3.96E-01	**4.02E-01**	2.00
Total Uranium			**4.30E-01**	
Uranium-235 wt % enrichment			**4.2**	

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