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TANK 22 SUPERNATE SAMPLE CHARACTERIZATION FOR SELECT RADIONUCLIDES

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EXECUTIVE SUMMARY

The Savannah River National Laboratory (SRNL) was requested by Savannah River Remediation (SRR), through a Technical Assistance Request (G-TAR-H-00007) to provide additional sample characterization on unfiltered Tank 22 supernate identified as sample HTF-22-18-117, which was delivered to SRNL Shielded Cells on December 14, 2018.

A summary of the average analytical results for this unfiltered Tank 22 supernate liquid follows.

- The density of the unfiltered Tank 22 supernate is 1.008 g/mL, 0.2%RSD*
- The Tank 22 supernate sample was analyzed for a total of 45 radionuclide species, with 15 of these radionuclides showing activity levels above minimum detectable activity of at least one replicate.
- Cesium-137 showed the highest activity at 2.90E+07 dpm/mL, 2.35 %RSD (1.30E-02 Ci/ L) with U-235 showing the lowest measurable activity at an average of 1.66E-01 dpm/mL, 6.26 %RSD (7.49E-11 Ci/L).
- The primary beta emitting radionuclides in the unfiltered Tank 22 supernate sample include Sr-90, Y-90 and Cs-137 at average activity concentrations of 1.10E-05, 1.10E-05 and 1.30E-02 Ci/L, respectively.
- The primary gamma emitting radionuclide is Ba-137^{m} at an average activity concentration of 1.23E-02 Ci/L (note that the activity concentration for Ba-137^{m} was calculated as 94.7% the Cs-137 activity concentration)†.

* Reported to 4 significant figures because this density measurement only differ in the 4th significant place when compared with that of distilled water or filtered Tank 22 supernate.

† Ba-137^{m} is the gamma emitting, short-lived, daughter isotope from the beta decay of Cs-137.

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LIST OF ABBREVIATIONS

AD	Analytical Development
DL	Detection limit
ICP-ES	Inductively coupled plasma emission spectroscopy
ICP-MS	Inductively coupled plasma mass spectrometry
LIMS	Laboratory information management system
MDA	Minimum detectable activity
MDL	Minimum detection limit
RSD	Relative standard deviation
SRNL	Savannah River National Laboratory
SRR	Savannah River Remediation
TAR	Technical Assistance Request
TTQAP	Task Technical and Quality Assurance Plan
UL	Upper limit

1.0 Introduction

The Savannah River National Laboratory (SRNL) was requested by Savannah River Remediation (SRR), through a Technical Assistance Request (TAR) to provide additional sample characterization on the unfiltered Tank 22 supernatant liquid identified as sample HTF-22-18-117, which was delivered to SRNL Shielded Cells on December 14, 2018.

While the original receipt and analysis were governed by a TTR and a Task Technical and Quality Assurance Plan (TTQAP)[2], this additional unfiltered Tank 22 supernate characterization is governed by a TAR[1]. Therefore, the supernate characterization found in this report is considered scoping, although the work did follow the same documentation and procedures identified in the TTQAP. Analysis of this unfiltered Tank 22 supernate were performed in triplicate.

2.0 Objectives

In a previous memo involving the characterization of Tank 22 sample[3] some radionuclide content had already been analyzed and reported for this same Tank 22 supernate sample filtrate. Those analyses were not repeated, and the earlier results for uranium isotopes, Th-232, and Np-237 which were based on filtered Tank 22 supernates, are included in this report. Additionally, the Pu-239, 240, 242, and 244 were also quantified in those earlier filtered samples but were not reported, and those results are included in this report. It is worth noting that the density of the unfiltered Tank 22 supernate solution was essentially the same in magnitude to that of the filtered Tank 22 supernate solution, meaning that there is essentially little or no suspended particles in the unfiltered Tank 22 supernate solution.

Recently, SRR requested additional characterization of this Tank 22 supernate sample for the following select radionuclides: plutonium isotopes (Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, Pu-244); curium isotopes (Cm-242, Cm-243, Cm-244, Cm-245, Cm-247, Cm-248); americium isotopes (Am-241, Am-242^{m}, Am-243); californium isotopes (Cf-249, Cf-251); nickel isotopes (Ni-59/63), radium isotope (Ra-226), carbon isotope (C-14); niobium isotope (Nb-94), iodine isotope (I-129), strontium isotope (Sr-90) and chlorine isotope (Cl-36). These isotopes are listed in the TAR[1].

3.0 Experimental Setups/Sample description and Preparations/Methodology

Ordinarily, the characterization of a radioactive liquid sample, such as this unfiltered Tank 22 supernate received in the SRNL Shielded Cell, would require acid and distilled water dilution of the sample followed by the removal of aliquot samples in green shielded cell bottles that are sent to SRNL-AD for analysis. This dilution and shielding reduces exposure to personnel transporting and working with theses sample. However, these dilutions also affect the detection limit of the final analytical result. To reach the detection limits for most of the radioactive species in the TAR, it was necessary to transport small portions (20-30 mL) of the unfiltered Tank 22 supernate, with effective shielding, to the analytical group for special storage and characterization for these radionuclides without any dilutions to achieve lower detection limits.

4.0 Data Quality and Blank Evaluations

Appendix A 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. The SRNL Analytical Development (AD) Group used reagent blanks based on dilute acids, de-ionized water and other test preparation techniques specific to each analytical method used in the sample preparation and characterizations in preparation for analysis.

The inductively coupled plasma mass spectrometry (ICP-MS) results are given for each atomic mass and in most cases each mass number represents only one isotope. An example of an exception is mass 238, since both uranium and plutonium are included in this mass number. However, since the mass contribution of U-238 is significantly greater than that of Pu-238, the 238 signal is used to quantify U-238, not Pu-238. For this reason, Pu-238 was determined by PUTTA (chemical separation coupled with alpha spectroscopy). In cases where ICP-MS and radiochemistry data give similar results for a species (e.g. Tc-99), radiochemistry was typically selected and reported due to better sensitivity and precision.

4.1 Format of the Reported Results

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 triplicate determinations but is typically not an indicator of analytical accuracy. In general, the one sigma analytical uncertainty as reported by Analytical Development was 10%, although it was sometimes lower or higher. Specifically, the one sigma analytical uncertainties reported by AD were: a) ~10% for ICP-MS and b) ~5% for Cs-137 determined by gamma spectroscopy. As such, only two to three of the leading digits reported for the AD analysis results should be considered significant.

In the Tank 22 supernate sample characterization results presented in Table 5-1 in this report, 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 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 a "≤ "sign that precedes the average value. The standard and percent relative 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-ES) 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 a 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 triplicate gave mixed results with one or two of the triplicate analyses results being less than the 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 values or the DL values as indicated by the analytical method.

5.0 Results and Discussion

Analyses were performed on the unfiltered Tank 22 supernate sample aliquots. A combination of routine measurement techniques and "tailor-made" separation/digestion/isolation/analysis methods were used to quantify for select radionuclides as requested by SRR as shown in Table 5-1. Details of the analytical methodologies employed in these characterizations are summarized in Appendix B. The density of the unfiltered Tank 22 supernate is 1.008 g/mL, 0.2%RSD and that of the filtered Tank 22 supernate (filtered with 0.45 µm Nalgene® filter membrane) is 1.003 g/mL, 0.6%RSD

The %RSD for all analytes with measurable minimum detectable activity as summarized in Table 5-1 are less than 10%, with analytical results for Cm-244 being the exception. The %RSD for Cm-244 at 84.3 % with a corresponding one sigma analytical uncertainty of 43.7% is above the analytical uncertainty for radioanalytical methods, which normally range from 5-20% for radionuclide quantified by counting methods (gamma spectroscopy, alpha spectroscopy and liquid scintillation counting). The source of this large variation in Cm-244 result is unknown. Carbon-14 (C-14) analytical uncertainty of 21.9 % is another analytical result which is above the 20% analytical uncertainty expectation.

The Tank 22 supernate was analyzed for a total of 45 radionuclide species. Only 15 of these radionuclides showed a minimum detectable activity (value above which instrument signal can be considered quantitative relative to the signal-to-noise ratio). Cesium-137 showed the highest activity at 2.90E+07 dpm/mL, 2.35 %RSD (1.30E-02 Ci/ L) with U-235 showing the lowest measurable activity at an average of 1.66E-01 dpm/mL, 6.26 %RSD (7.49E-11 Ci/L). As earlier mentioned, Th-232, the uranium isotopes and Np-237 data presented in Table 5-1 were based on the analysis of the filtered Tank 22 supernate as previously reported.[3]

Other detected radionuclides in the unfiltered Tank 22 supernate sample included Sr-90 (2.45E+04 dpm/mL, 5.90 %RSD (1.10E-05 Ci/L)); Y-90 (2.45E+04 dpm/mL, 5.90 %RSD (1.10E-05 Ci/L)); Tc-99 (6.77E+03 dpm/mL, 5.73%RSD (3.05E-06 Ci/L)); C-14 (1.61E+02 dpm/mL, 8.79 %RSD (7.27E-08 Ci/L)); Am-241 (≤ 1.43E+01 dpm/mL, (≤ 6.44E-09 Ci/L)), U-238 (3.72E+00 dpm/mL, 6.49%RSD (1.67E-09 Ci/L)); Am-243 (≤3.10E+00 dpm/mL, (≤ 1.40E-09 Ci/L)).

All the measured plutonium isotopic activities were below instrument/method detection limits and so are reported as less than values. Both Ba-137^{m} and Y-90 activities were calculated as 94.7%[6] of the Cs-137 and 100 % of the Sr-90 activities, respectively.

Thus, the primary beta emitting radionuclides in the Tank 22 supernate sample include Sr-90, Y-90 and Cs-137 at average activity concentrations of 1.10E-05, 1.10E-05 and 1.30E-02 Ci/ L, respectively. The primary gamma emitting radionuclide is Ba-137^{m} at average activity concentration of 1.23E-02 Ci/ L.

Table 5-1 Analysis Results for Tank 22 Supernate Characterization for Select Radionuclides.

Analyte	Run-1 dpm/mL	Run-2 dpm/mL	Run-3 dpm/mL	Average dpm/mL	Average Ci/L of supernate	%RSD N = 3	One sigma % uncertainty
C-14	1.74E+02	1.46E+02	1.64E+02	1.61E+02	7.27E-08	8.79	21.9
Cl-36	<1.39E+02	<1.16E+02	< 1.05E+02	<1.20E+02	< 5.41E-08		UL
Ni-59	< 5.55E+01	< 8.87E+01	< 5.73E+01	< 6.72E+01	< 3.03E-08		UL
Ni-63	< 1.58E+02	< 4.05E+01	< 3.15E+01	< 7.67E+01	< 3.45E-08		UL
Co-60	5.42E-01	3.80E-01	< 3.37E-01	≤ 4.2E-01	≤ 1.89E-10		14.3/MDA
Sr-90	2.28E+04	2.53E+04	2.53E+04	2.45E+04	1.10E-05	5.90	16.3
Y-90	2.28E+04	2.53E+04	2.53E+04	2.45E+04	1.10E-05	5.90	16.3
Nb-94	< 1.27E+00	< 2.29E+00	< 1.30E+00	< 1.62E+00	< 7.30E-10		MDA
Tc-99	6.81E+03	6.70E+03	6.79E+03	6.77E+03	3.05E-06	5.73	5.73
Ru-106	<3.26E+00	<9.11E+00	< 8.69E+00	<7.02E+00	< 3.16E-09		MDA
Sb-125	8.92E+00	7.34E+00	8.95E+00	8.94E+00	4.03E-09	0.24	12.1
Sb-126	1.44E+01	1.14E+01	1.40E+01	1.42E+01	6.40E-09	1.99	5.0
Sn-126	1.44E+01	1.14E01	1.40E+01	1.42E+01	6.40E-09	1.99	5.0
I-129	<3.47E+00	<8.62E-01	< 2.96E+00	<2.43E+00	< 1.09E-09		MDA
Cs-134	<5.55E+03	<4.57E+03	< 6.02E+03	<5.38E+03	< 2.42E-06		MDA
Cs-137	2.95E+07	2.92E+07	2.82E+07	2.90E+07	1.30E-02	2.35	5
Ba-137^{m}	2.79E+07	2.76E+07	2.67E+07	2.74E+07	1.23E-02	2.35	5
Ce-144	<4.91E+00	<1.15E+01	<1.17E+01	<9.37E+00	< 4.22E-09		MDA
Eu-152	<7.69E-01	<1.53E+00	< 1.96E+00	<1.42E+00	< 6.40E-10		MDA
Eu-154	<5.18E-01	<9.48E-01	< 1.23E+00	<8.99E-01	< 4.05E-10		MDA
Eu-155	<1.85E+00	<3.13E+01	< 2.49E+01	<1.94E+01	< 8.74E-09		MDA
Ra-226	< 9.03E+00	< 8.46E+00	< 7.85E+00	< 8.45E+00	< 3.81E-09		MDA
Th-232	<4.12E-02	< 4.15E-02	< 4.00E-02	<4.09E-02	< 1.84E-11		MDA
U-233	<2.41E+02	< 2.44E+02	< 2.35E+02	<2.40E+02	< 1.08E-07		MDA
U-234	<1.56E+02	< 1.58E+02	< 1.52E+02	<1.55E+02	< 6.99E-08		MDA
U-235	1.78E-01	1.64E-01	1.57E-01	1.66E-01	7.49E-11	6.26	10
U-236	<1.62E+00	< 1.63E+00	< 1.57E+00	< 1.61E+00	< 7.24E-10		MDA
Np-237	< 1.76E+01	< 1.78E+01	< 1.71E+01	< 1.75E+01	< 7.88E-09		MDA
U-238	3.99E+00	3.65E+00	3.52E+00	3.72E+00	1.67E-09	6.49	10
Np-239	<2.67E+00	<5.96E+00	<6.18E+00	<4.94E+00	< 2.23E-09		MDA
Pu-238	<1.24E+02	<1.11+02	<1.29E+02	<1.21E+02	< 5.45E-08		MDA
Pu-239	< 1.55E+03	< 1.57E+03	< 1.51E+03	< 1.54E+03	< 6.95E-07		MDA
Pu-239/240	<1.15E+02	<6.21E+01	<1.03E+02	<9.34E+01	< 4.21E-08		MDA
Pu-240	< 5.69E+03	< 5.75E+03	< 5.54E+03	< 5.66E+03	< 2.55E-06		MDA
Pu-241	<1.72E+02	<1.72E+02	<1.72E+02	<1.72E+02	< 7.75E-08		MDA
Pu-242	< 9.54E+01	< 9.63E+01	< 9.29E+01	< 9.49E+01	< 4.27E-08		MDA
Pu-244	< 4.43E-01	< 4.48E-01	< 4.32E-01	< 4.41E-01	< 1.99E-10		MDA
Am-241	8.72E+00	<1.68E+01	<1.73E+01	≤ 1.43E+01	≤ 6.44E-09		11/MDA
Am-242^{m}	< 1.08E-01	< 1.08E-01	< 1.08E-01	< 1.08E-01	< 4.86E-11		MDA
Am-243	2.79E+00	<2.09E+00	< 4.41E+00	≤ 3.10E+00	≤ 1.40E-09		32/MDA
Cm-242	< 6.68E-01	< 1.35E+00	< 3.93E+00	< 1.98E+00	< 8.92E-10		MDA
Cm-243	< 6.67E+00	< 7.54E+00	< 1.28E+01	< 9.04E+00	< 4.07E-09		MDA
Cm-244	7.58E+01	2.65E+02	6.24E+01	1.34E+02	6.05E-08	84.3	43.7
Cm-245	< 5.49E+00	< 6.18E+00	< 1.05E+01	< 7.39E+00	< 3.33E-09		MDA
Cm-247	< 6.20E+00	< 8.06E+00	< 1.31E+01	< 9.12E+00	< 4.11E-09		MDA
Cm-248	< 3.44E+00	< 3.11E+01	< 1.83E+00	< 1.21E+01	< 5.45E-09		MDA
Cf-249	< 7.38E+00	< 8.61E+00	< 1.34E+01	< 9.80E+00	< 4.41E-09		MDA
Cf-251	< 6.44E+00	< 7.23E+00	< 1.26E+01	< 8.76E+00	< 3.95E-09		MDA

6.0 Conclusions

The Tank 22 supernate sample HTF-22-18-117 has been characterized for select radionuclides as requested in the TAR[1].

A summary of the analytical results for the Tank 22 supernate sample follows.

- The density of the unfiltered Tank 22 supernate is 1.008 g/mL, 0.2%RSD.
- The Tank 22 supernate sample was analyzed for a total of 45 radionuclide species, with 15 of these radionuclides showing activity levels above the minimum detectable activity of at least one replicate (value above/equal instrument signal, which can be considered quantitative relative to the signal-to-noise ratio).
- Cesium-137 showed the highest activity at 2.90E+07 dpm/mL, 2.35 %RSD (1.30E-02 Ci/ L) with U-235 showing the lowest measurable activity at an average of 1.66E-01 dpm/mL, 6.26 %RSD (7.49E-11 Ci/L).
- The primary beta emitting radionuclides in the Tank 22 supernate sample include Sr-90, Y-90 and Cs-137 at average activity concentrations of 1.10E-05, 1.10E-05 and 1.30E-02 Ci/ L, respectively.
- The primary gamma emitting radionuclide is Ba-137^{m} at average activity concentration of 1.230E-02 Ci/ L (note that the activity concentration for Ba-137^{m} was calculated as 94.7%[6] the Cs-137 activity concentration).

7.0 Quality Assurance

Requirements for performing reviews of technical reports and the extent of review are established in Manual E7 Procedure 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.

This characterization of Tank 22 unfiltered supernate was a scoping activity with no specified quality requirements from the customer.

8.0 References

1. “Tank 22 Supernate Sample Characterization,” G-TAR-H-00007, Rev. 0, August 5, 2019.
2. “Task Technical and Quality Assurance Plan for Tank 22 Settled Sludge Core Sample Characterization and Testing,” SRNL-RP-2018-00530, Rev. 0., June 25, 2018.
3. L. N. Oji, J. M. Pareizs, C. L. Trivelpiece, “Tank 22 Composite Core Sludge Sample Characterization Results -Radionuclides, Elementals and Anions,” SRNL-L3100-2019-00024, Rev. 0., June 28, 2019.
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 2000.
6. National Nuclear Data Center: https://www.nndc.bnl.gov/nudat2/getdecayscheme.jsp?nucleus=137BA&dsid=137cs%20bM%20decay&unc=nds

Appendix A: Tank 22 unfiltered supernate characterization AD Tracking Numbers*

Analytes	Method (s)	SRNL AD Tracking Number (LIMS):
C-14	Carbon-14-method	LW14768-LW147771
Cl-36	Cl-36-method	LW14768-LW147771
Ni59/63	Ni59/63-method	LW14768-LW147771
Co-60	Cs-removed gamma spect.	LW14793- LW14795
Sr-90	Sr90-method	LW14768-LW14771
Nb-94	Nb-94 method	LW14768-LW14771
Tc-99	LSC	LW14768-LW147771
Ru-106	Cs-removed gamma spect.	LW14793- LW14795
Sb-125	Cs-removed gamma spect.	LW14793- LW14795
Sb-126	Cs-removed gamma spect.	LW14793- LW14795
Sn-126	Cs-removed gamma spect.	LW14793- LW14795
I-129	I-129 method	LW14768- LW14771
Cs-134	GAMMA SPEC	LW14768- LW14771
Cs-137	GAMMA SPEC	LW14768- LW14771
Ce-144	Cs-removed gamma spect.	LW14793- LW14795
Eu-152	Cs-removed gamma spect.	LW14793- LW14795
Eu-154	Cs-removed gamma spect.	LW14793- LW14795
Eu-155	Cs-removed gamma spect.	LW14793- LW14795
Ra-226	Ra-226 method	LW14768- LW14771
Th-232	ICP-MS	LW12306- LW12308
U-233	ICP-MS	LW12306- LW12308
U-234	ICP-MS	LW12306- LW12308
U-235	ICP-MS	LW12306- LW12308
U-236	ICP-MS	LW12306- LW12308
U-238	ICP-MS	LW12306- LW12308
NP-237	ICP-MS	LW12306- LW12308
Np-239	Cs-removed gamma spect.	LW14793- LW14795
Pu-238	Pu-238/241	LW14768-LW14771
Pu-239	ICP-MS	LW12306- LW12308
Pu-239/ Pu-240	Pu-TTA	LW14768-LW14771
Pu-240	ICP-MS	LW12306- LW12308
Pu-241	Pu-238/241	LW14768-LW14771
Pu-242	ICP-MS	LW12306- LW12308
Pu-244	ICP-MS	LW12306- LW12308
Am-241	Cs-removed gamma, Am/Cm	LW14793- LW14795, LW14768- LW14771
Am-243	Cs-removed gamma, Am/Cm	LW14793- LW14795, LW14768- LW14771
Cm-243	Am/Cm	LW14768- LW14771
Cm-244	Am/Cm	LW14768-LW14771
Cm-245	Am/Cm	LW14768-LW14771
Cm-247	Am/Cm	LW14768-LW14771
Cm-248	Am/Cm	LW14768-LW14771
Cm-249	Am/Cm	LW14768-LW14771
Cm-252	Am/Cm	LW14768-LW14771

*Project: IDs: LW-AD-PROJ-190807-5, Project: ID: LW-AD-PROJ-190807-7 and Project: ID: LW-AD-PROJ-181115-8

Appendix B: Summary of Analytical Methods

Inductively Coupled Plasma – Mass Spectroscopy (ICP-MS)

Samples are diluted as necessary to bring analytes 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.

Sr-90

Aliquots of Tank 22 supernate samples 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.

Am-241 (Cs-removed gamma analysis)

Aliquots of Tank 22 samples 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 listed above. Aliquots of Tank 22 supernate were analyzed for Co-60 and Am-241. Sb-125, SB-126 and Sn-126were also measured from this analysis.

Pu-238, 239/240, 241

Aliquots of Tank 22 supernate samples 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 Brand name for one of Eichrom's resins). The Pu-containing extracts were measured by liquid scintillation analysis to determine Pu-241 concentrations. Laboratory reagent blanks and a Pu-238 standard were run as controls.

Pu-239, 240, 242, 244

These actinides were present at very low levels in Tank 22, as a result, the plutonium isotopes detection limits from the direct ICP-MS analysis were reported. No radiochemical separations were carried out.

Am-242m, 243, Cm-243, 244, 245, 247, 248, Cf-249, 251

Aliquot of Tank 22 supernate samples were digested using a sodium peroxide fusion. The americium, curium and californium species were extracted from aliquots of peroxide fusion using a CMPO/tributyl phosphate commercial resin based solid phase extractant and purified further with a commercial resin using HDEHP bound to a solid phase extractant support. 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, 244, and Cm-248 concentrations were measured using passivated, implanted, planar silicon (PIPS) alpha spectrometers. Am-241 quantities had been measured from the cesium removed gamma analyses, Am, Cm, and Cf results were traced with the measured Am-241 recoveries. from the sample matrix. Laboratory reagent blanks were also run as controls.

Ni-59, Ni-63

Aliquots of Tank 22 supernates were spiked with an elemental nickel carrier. The nickel species were extracted from the matrix using dimethylglyoxime (DMG) based extractant. Ni-59 concentrations were measured using low energy photon/x-ray, thin-windowed, semi-planar high purity germanium spectrometers. Ni-63 concentrations were measured by liquid scintillation analysis. Elemental nickel carrier yields were measured by ICP-ES and were used to correct the radioactive nickel species' analyses for any nickel losses from the radiochemical separations. Reagent blanks, a Ni-63 standard and a Ni-59 standard were run as controls.

Ra-226

Tank 22 supernate sample aliquots were digested using a PF. Each replicate was prepared in duplicate with the duplicate containing a Ra-224 tracer. The Ra-226 was extracted from the matrix using a combination of resin decontamination and ion exchange. The purified Ra-226 was sealed in polypropylene tubes and stored for several daughter Rn-222 half-lives. The Ra-226 progeny daughter isotope Pb-214 was then analyzed for using a high purity germanium well gamma ray spectrophotometer and results were corrected spike Ra-226 recoveries. A simulant blank spiked with Ra-226 was run through the process to serve as a calibration standard. A simulated blank sample spiked with Ra-226 was run through the process to serve as a control standard.

I-129

Tank 22 supernate samples were dissolved in concentrated acid with an added KI carrier. A matrix blank and matrix blank containing an I-129 spike were also prepared using sodalite. The samples were rendered caustic and decontaminated with strikes with crystalline silicotitanate (CST) and monosodium titanate (MST) followed by a filtration step. The samples were then acidified and treated with Actinide and AMP resins to facilitate removal of interfering isotopes. Sodium sulfite was added to the material to reduce the iodine. Silver nitrate was added to the solution to precipitate the iodine as AgI, which was separated via filtration. The filtrate is analyzed for I-129 content using low energy photon/x-ray, thin-windowed, semi-planar, high purity germanium spectrometers. Elemental iodine yields were measured by neutron activation analysis and were used to correct the I-129 analyses for any iodine losses from the radiochemical separation.

C-14

Tank 22 supernate sample was added to a mixture of sodium hydroxide, and sodium carbonate/sodium hydroxide. A series of oxidation and reduction steps designed to liberate C-14 containing carbon dioxide were carried out, which selectively trapped the C-14 in a basic solution. The basic solutions were acidified and the C-14 containing carbon dioxide was captured in Carbosorb E and measured by liquid scintillation analysis. A laboratory blank, a C-14 calibration standard and a C-14 control standard were also run through the process.

Nb-94

Aliquots of Tank 22 supernate were spiked with a radioactive Nb-95 tracer, and then purified using Triskem Industry's Zr Resin. The purified aliquots were analyzed by high purity germanium spectrometers to measure Nb-94 and Nb-95. The niobium chemical recoveries were determined from the Nb-95 tracer measurement. The Nb-94 values were corrected with the Nb-95 recoveries. Reagent blank and laboratory reagent blanks were run as controls.

Cl-36

Sub-samples of Tank 22 supernate sample were weighed and then digested in concentrated acid. The dissolutions were subjected to numerous resin-based decontamination steps. Chlorine was then separated from the non-volatile components of the matrix via AgCl precipitation. The precipitate was then counted using a gas flow proportional counter (GFPC) analysis. The AgCl precipitate was then activated by neutron activation analysis to determine Cl losses during the processes, and to correct Cl-36 results for those losses. The HCl used to initially digest the samples was used to trace Cl-36 throughout the processes.

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