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Sample Results from the Interim Salt Disposition Program Macrobatch 6 Tank 21H Qualification Samples

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EXECUTIVE SUMMARY

Savannah River National Laboratory (SRNL) analyzed samples from Tank 21H in support of qualification of Macrobatch (Salt Batch) 6 for the Interim Salt Disposition Project (ISDP). This document reports partial results of the analyses of samples of Tank 21H. No issues with the projected Salt Batch 6 strategy are identified.

A subsequent report will provide analyses related to the Actinide Removal Process (ARP) test and Extraction-Scrub-Strip (ESS) test.

LIST OF ABBREVIATIONS

AA – Atomic Absoption
AD – Analytical Development
AMP – ammonium molybdophosphate
ARP – Actinide Removal Project
CVHg – Cold Vapor Mercury
ESS – extraction, scrub, strip
IC – Ion Chromatography
ICPES – Inductively Coupled Plasma Emission Spectroscopy
ICPMS – Inductively Coupled Plasma Mass Spectroscopy
ISDP – Interim Salt Disposition Program
MST – monosodium titanate
PuTTa – plutonium thenoyl trifluoroacetone scintillation
RSD – relative standard deviation (percent)
SRNL – Savannah River National Laboratory
SRR – Savannah River Remediation
TIC-TOC – Total Inorganic Carbon-Total Organic Carbon
TTQAP - Task Technical and Quality Assurance Plan
TTR – Technical Task Request
WAC – Waste Acceptance Criteria
% RSD – percent relative standard deviation

1.0 Introduction

This report covers the Tank 21H qualification sample results for Macrobatch (Salt Batch) 6 of the Interim Salt Disposition Program (ISDP). A previous document covers initial characterization which includes results for a number of non-radiological analytes.[1] This work was specified by Task Technical Request [2] and by Task Technical and Quality Assurance Plan (TTQAP).[3]

Details for the work are contained in controlled laboratory notebooks.[4]

2.0 Experimental Procedure

Six Tank 21H samples (i.e., dip sample bottles HTF-21-12-96, HTF-21-12-97, HTF-21-12-98, HTF-21-12-99, HTF-21-12-100, and HTF-21-12-101) arrived at SRNL on October 3, 2012. The samples were optically clear, with no visible solids present. Researchers measured the density of each of the solutions. With customer concurrence, the samples were combined and mixed. After combining, duplicate filtered samples (using a 0.45 μm syringe filter) were sent to Analytical Development (AD) for analysis. Samples were not diluted before delivery to AD.

3.0 Results and Discussion

In a previous document,[1] density, Inductively Coupled Plasma Emission Spectroscopy (ICPES), Ion Chromatography (IC) and Free Hydroxide results were reported for the Tank 21H composite. These results are also reported here for completeness (Table 1). Values in parentheses are the relative standard deviation (RSD).

Table 1. Previous Results

Analyte	Result (mg/L)	Analyte	Result (mg/L)
Ag	<1.74	Sb	<36.2
Al	5380 (0.00%)	Si	37.2 (0.21%)
B	45.4 (0.16%)	Sn	<14.7
Ba	<0.97	Sr	<0.15
Be	<0.24	Th	<7.30
Ca	<1.49	Ti	<0.400
Cd	<1.21	U	<45.0
Ce	<7.91	V	<0.52
Cr	45.7 (0.15%)	Zn	5.07 (0.56%)
Cu	0.760 (1.86%)	Zr	<0.56
Fe	2.84 (0.75%)	F^-	<100
Gd	<1.92	Cl^-	100 (2.83%)
K	460 (6.92%)	Br^-	<500
La	<1.36	Formate	330 (1.29%)
Li	14.6 (0.49%)	Nitrite	21800 (7.14%)
Mg	<0.12	Nitrate	138500 (1.53%)
Mn	<0.34	Phosphate	383 (4.25%)
Mo	<11.2	Sulfate	6480 (0.44%)
Na	149000 (0.48%)	Oxalate	377 (0.56%)
Ni	<3.70	TIC	2510 (0.56%)
P	191 (0.00%)	TOC	385 (0.37%)
Pb	<17.6	Free Hydroxide	1.90 M (2.23%)
S	3450 (0.21%)	Density	1.304 (1.05%) g/mL

Values in parentheses are the RSD.

The nickel (Ni) result converted into a concentration of $Ni(OH)_2$ is <5.84 mg/L. The free hydroxide converts to a pH of 14.

The TIC and TOC results are in terms of mg/L of carbon. If we assume that the entire TIC result is carbonate, this translates to a carbonate concentration of 0.209 M.

The analytical uncertainty is typically <1% for density measurements. ICPES analytical uncertainty is 10%. The values in the parentheses are the percentage relative standard deviation (RSD). The analytical uncertainty for the IC results is 10%. The analytical uncertainty for the TIC-TOC results is 10%. The analytical uncertainty for the Free Hydroxide result is 3%. The values in the parentheses are the RSD.

3.1 Tank 21H Qualification Analyses

The tank samples were analyzed by Analytical Development (AD) by the listed non-radiological methods (Table 2) and radiological (Table 3) methods. Analyses were performed in duplicate and reported in Tables 4 and 5, respectively. Averages of the individual results, with the percent relative standard deviation (RSD) in parentheses, are reported. Shaded sample results indicate calculated values.

Table 2. Non-Radiological Analyses

Method	Analyte
IC Cations	NH_4^+
TIC	total inorganic carbon
TOC	total organic carbon
AA-As	As
AA-Se	Se
CV-Hg	Hg
HPLC	tetraphenylborate, phenol
SVOA	tributylphosphate
VOA	isopropanol, butanol, isobutanol

Table 3. Radiological Analyses

Method	Analyte
Tritium	^{3}H
^{14}C	^{14}C
gamma scan, Cs-removed	^{60}Co, ^{94}Nb, ^{106}Ru, ^{125}Sb, ^{126}Sn, ^{144}Ce, ^{154}Eu, ^{155}Eu, ^{241}Am, ^{226}Ra,
^{90}Sr	^{90}Sr
^{129}I	^{129}I
gamma scan	^{134}Cs, ^{137}Cs
^{232}U	^{232}U
$^{238-241}Pu$ (filtered and unfiltered) (Plutonium thenoyl trifluoroacetone scintillation)	^{238}Pu, $^{239/40}Pu$, ^{241}Pu
Am/Cm	^{241}Am, ^{243}Am, ^{244}Cm, ^{245}Cm
$^{59/63}Ni$	$^{59/63}Ni$
^{99}Tc	^{99}Tc
$^{147}Pr/^{151}Sm$	$^{147}Pr/^{151}Sm$
ICPMS (Inductively Coupled Plasma Mass Spectroscopy)	isotopes from mass number 81 to 209 and 230 to 252, incl. ^{233}U and above, ^{237}Np, ^{230}Th, ^{232}Th
Liquid Scintillation Counting	total alpha, total beta

3.2 Tank 21H Qualification Results (non-radiological analytes)

The results for the IC-Cations, weight percent insoluble solids, phenol, tetraphenylborate, tributyl phosphate, isopropanol, methanol, isobutanol, butanol, arsenic, mercury, and selenium are listed in Table 4. The analytical uncertainty for all listed analyses is 20%, except for the IC-Cations and wt% insoluble solids, which are 10%. Results shaded in green are calculated results. Values in parentheses are RSD.

Table 4. Miscellaneous Results (mg/L unless otherwise noted)

Analyte	Result
ammonium	<100
wt % insoluble solids	<0.17
phenol	<20
tetraphenylborate	<5
tributylphosphate	<1
isopropanol	<0.25
butanol	<0.25
isobutanol	<0.25
methanol	≤517
As	0.683 (1.24%)
Hg	12.8 (31.4%)
Se	0.224 (1.58%)

Methanol is a calculated value.
Values in parentheses are the RSD.

The oxalate concentration is 377 mg/L, and the formate result is 330 mg/L. The oxalate result is converted to the equivalent carbon result of 103 mg/L. The formate result is converted to the equivalent carbon result of 88.1 mg/L. Subtracting these results from the TOC result gives a remainder of 194 mg/L of carbon. If we assume all of this remainder carbon is in the form of methanol, this gives a calculated methanol result of 517 mg/L. This methanol result should be considered an upper bound as no direct analytical method for methanol exists.

3.3 Tank 21H Qualification Results (radiological analytes)

The results of the radiological analysis are listed in Table 5. The analytical uncertainty for ICPMS samples are 20%. Other analytical methods have varying uncertainties, typically 5-10% and are noted for single sample results.

Results given in italics indicate that one of the sample results was either below detection or quantification limits, in which case the value in the parentheses is the analytical uncertainty. Only quantifiable measured values are reported when available.

^{90}Y is calculated as equal to the ^{90}Sr result. ^{106}Rh is calculated as equal to the ^{106}Ru result. The ^{125m}Te is calculated as equal to the ^{125}Sb result.^{137m}Ba is calculated as 94.7% of the ^{137}Cs result (as seen in Table 5).[5] ^{144}Pr is calculated as equal to the ^{144}Ce result. The ^{135}Cs result assumes that all of mass 135 from the ICPMS result is ^{135}Cs. The ^{232}Th result assumes that all of mass 232 from the ICPMS result is ^{232}Th. The Total Alpha value is calculated by adding all the alpha results together and treating the less-than results as real values. Thus, this value is biased high. Total gamma is calculated as the sum of the ^{137}Cs, ^{134}Cs, ^{135}Cs, ^{60}Co, ^{94}Nb, ^{106}Ru, ^{125}Sb, ^{126}Sn, ^{144}Ce, ^{144}Pr, ^{154}Eu, ^{155}Eu, and ^{226}Ra. The ^{238}Pu, $^{239/40}$Pu, and ^{241}Pu results are from radiocounting, while the other Pu results are from ICPMS.

Table 5. Radiological Results of Tank 21H Analyses for Macrobatch 6 (pCi/mL unless otherwise noted)

Analyte	Average Result	Analyte	Average Result
^{3}H	<9.13E+02	^{154}Eu	<1.38E+01
^{14}C	<5.45E+03	^{155}Eu	<6.62E+01
^{59}Ni	<2.17E+01	^{226}Ra	<1.04E+03
^{63}Ni	<2.53E+01	^{232}Th	<2.20E-03
^{60}Co	<4.12E+00	^{232}U	2.91E+00 (44.4%)
^{90}Sr	1.76E+05 (21.2%)	^{233}U	<1.94E+02
^{90}Y	1.76E+05 (21.2%)	^{234}U	<1.25E+02
^{94}Nb	<9.95E+00	^{235}U	3.33E-01 (17.5%)
^{99}Tc	2.17E+04 (3.81%)	^{236}U	<1.29E+00
^{106}Ru	<1.81E+02	^{238}U	6.57E+00 (7.60%)
^{106}Rh	<1.81E+02	^{237}Np	<2.82E+01
^{125}Sb	<1.48E+02	^{238}Pu (total)	1.08E+04 (6.17%)
^{125m}Te	<1.48E+02	$^{239/40}$Pu	9.90E+02 (5.14%)
^{126}Sn	2.26E+02 (3.37%)	^{241}Pu	<3.52E+03
^{129}I	1.79E+01 (4.44%)	^{242}Pu	<7.64E+02
^{134}Cs	<6.39E+03	^{244}Pu	<3.54E+00
^{135}Cs	3.66E+02 (10.9%)	^{241}Am	*6.17E+01 (14.9%)*
^{137}Cs	5.76E+07 (0.00%)	^{243}Am	<2.70E+00
^{137m}Ba	5.45E+07 (0.00%)	^{244}Cm	*6.89E+01 (15.9%)*
^{144}Ce	<1.69E+02	^{245}Cm	<7.20E+00
^{144}Pr	<1.69E+02	Total Alpha	<1.26E+04
^{147}Pm	<4.77E+01	Total Beta	7.58E+07 (2.94%)
^{151}Sm	<2.34E+01	Total Gamma	5.76E+07

Results shaded in green are calculated values.
Values in parentheses are the RSD.
Values in italics are single results.

4.0 Conclusions

Results of the analyses of the Tank 21H samples from this report in conjunction with the findings of the previous report,[1] indicates that the material does not display any unusual characteristics nor pose any concerns for processing or the batching strategy.

5.0 References

[1] T. B. Peters, S. D. Fink, "Results of Initial Analyses of the Macrobatch 6 Tank 21H Qualification Samples", SRNL-STI-2012-00685, November 2012.

[2] S. E. Campbell, "Qualification of ISDP Salt Batch 6", HLW-DWPF-TTR-2012-0012, Rev. 1, October 18, 2012.

[3] T. B. Peters and S. D. Fink, "Task Technical and Quality Assurance Plan for ISDP Salt Batch 6 Sample Qualification", SRNL-RP-2012-00625, Rev.0, October 11, 2012.

[4] T. B. Peters, "ISDP6", SRNL-NB-2012-00107, October 25, 2012.

[5] http://www.nndc.bnl.gov, E. Browne, J. K. Tuli Citation: Nuclear Data Sheets 108, 2173 (2007)

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