We put science to work.™

Savannah River National Laboratory™

OPERATED BY SAVANNAH RIVER NUCLEAR SOLUTIONS

A U.S. DEPARTMENT OF ENERGY NATIONAL LABORATORY • SAVANNAH RIVER SITE • AIKEN, SC

Analysis of 2H-Evaporator Scale Wall [HTF-13-82] and Pot Bottom [HTF-13-77] Samples

L. N. Oji

September 2013

SRNL-STI-2013-00352, Rev 1

SRNL.DOE.GOV

DISCLAIMER

Printed in the United States of America

Prepared for
U.S. Department of Energy

Keywords: Tank Farm, Sodium Aluminosilicate, Evaporator

Retention: ***Permanent***

Analysis of 2H-Evaporator Scale Wall [HTF-13-82] and Pot Bottom [HTF-13-77] Samples

L. N. Oji
September 2013

Prepared for the U.S. Department of Energy under contract number DE-AC09-08SR22470.

REVIEWS AND APPROVALS

AUTHOR:

L. N. Oji, Author, Advanced Characterization & Processing Date

TECHNICAL REVIEW: (Reviewed per E7 2.60)

S.H. Reboul, Advanced Characterization & Processing
Date

APPROVALS:

F. M. Pennebaker, Manager, Advanced Characterization & Processing Date

S. L. Marra, Manager, Date
Environmental & Chemical Process Technology Research Programs

D. J. Martin, Manager, SRR PM-E-E-TFE Date

LIST OF REVISIONS		
Revision Number	**Summary of Changes**	**Date**
0	Initial issuance	June 21, 2013
1	Additional analyses as described in section 5 and on Tables 3-6.	July 22, 2013

ACKNOWLEDGEMENTS

The author extends thanks to the multiple members of the SRNL Shielded Cells and AD organization members who processed the samples and provided analytical results, specifically, C. Coleman, R. Sullivan and D. Missimer.

EXECUTIVE SUMMARY

Savannah River Remediation (SRR) is planning to remove a buildup of sodium aluminosilicate scale from the 2H-evaporator pot by loading and soaking the pot with heated 1.5 M nitric acid solution. Sampling and analysis of the scale material has been performed so that uranium and plutonium isotopic analysis can be input into a Nuclear Criticality Safety Assessment (NCSA) for scale removal by chemical cleaning. Historically, since the operation of the Defense Waste Processing Facility (DWPF), silicon in the DWPF recycle stream combines with aluminum in the typical tank farm supernate to form sodium aluminosilicate scale mineral deposits in the 2H-evaporator pot and gravity drain line.

The 2H-evaporator scale samples analyzed by Savannah River National Laboratory (SRNL) came from two different locations within the evaporator pot; the bottom cone sections of the 2H-evaporator pot [Sample HTF-13-77] and the wall 2H-evaporator [sample HTF-13-82].

X-ray diffraction analysis (XRD) confirmed that both the 2H-evaporator pot scale and the wall samples consist of nitrated cancrinite (a crystalline sodium aluminosilicate solid) and clarkeite (a uranium oxy-hydroxide mineral).

On "as received" basis, the bottom pot section scale sample contained an average of 2.59E+00 ± *1.40E-01* wt % total uranium with a U-235 enrichment of 6.12E-01 ± *1.48E-02 %*, while the wall sample contained an average of 4.03E+00 ± *9.79E-01* wt % total uranium with a U-235 enrichment of 6.03E-01% ± *1.66E-02* wt %.

The bottom pot section scale sample analyses results for Pu-238, Pu-239, and Pu-241 are 3.16E-05 ± *5.40E-06* wt %, 3.28E-04 ± *1.45E-05 wt* %, and <8.80E-07 wt %, respectively. The evaporator wall scale samples analysis values for Pu-238, Pu-239, and Pu-241 averages 3.74E-05 ± *6.01E-06* wt %, 4.38E-04 ± *5.08E-05 wt* %, and <1.38E-06 wt %, respectively. The Pu-241 analyses results, as presented, are upper limit values.

For these two evaporator scale samples obtained at two different locations within the evaporator pot the major radioactive components (on a mass basis) in the additional radionuclide analyses were Sr-90, Cs-137 Np-237, Pu-239/240 and Th-232. Small quantities of americium and curium were detected in the blanks used for Am/Cm method for these radionuclides. These trace radionuclide amounts are assumed to come from airborne contamination in the shielded cells drying or digestion oven, which has been replaced. Therefore, the Am/Cm results, as presented, may be higher than the true Am/Cm values for these samples.

These results are provided so that SRR can calculate the equivalent uranium-235 concentrations for the NCSA. Results confirm that the uranium contained in the scale remains depleted with respect to natural uranium. SRNL did not calculate an equivalent U-235 enrichment, which takes into account other fissionable isotopes U-233, Pu-239 and Pu-241. The applicable method for calculation of equivalent U-235 will be determined in the NCSA.

With a few exceptions, a comparison of select radionuclides measurements from this 2013 2H evaporator scale characterization (pot bottom and wall scale samples) with those measurements for the same radionuclides in the 2010 2H evaporator scale analysis shows that the radionuclide analysis for both years are fairly comparable; the analyses results are about the same order of magnitude.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

CVAA-Hg	Cold Vapor Atomic Absorption for Mercury
DWPF	Defense Waste Processing Facility
HLW	High-Level Waste
ICP-ES	Inductively Coupled Plasma–Atomic Emission Spectroscopy
ICP-MS	Inductively Coupled Plasma – Mass Spectrometry
LSC	Liquid Scintillation Counting
NCSA	Nuclear Criticality Safety Assessment
PHA	Pulse Height Analysis
SRNL	Savannah River National Laboratory
SRR	Savannah River Remediation
TTA	Thenoyltrifluoro-acetone
TTR	Technical Task Request
XRD	X-Ray Diffraction

1.0 Introduction

Savannah River Remediation (SRR) is planning to remove a buildup of sodium aluminosilicate scale from the 2H-evaporator pot by the addition of dilute nitric acid solution.[1] Sampling and analysis of the scale material provides the data needed for SRR to perform a Nuclear Criticality Safety Assessment (NCSA) of scale removal by chemical cleaning. Evaporator scale samples [HTF-13-77] from the evaporator pot bottom and evaporator wall scale samples [HTF-13-82] and [HTF-13-78] were pulled by SRR and delivered to Savannah River National Laboratory (SRNL) in May 2013 for analysis for primary fissile isotopes and non-fissile isotopes of uranium.

The 242-16H evaporator (2H evaporator) system concentrates high level waste consisting primarily of high-level waste (HLW) recycled to the SRS H-Area Tank Farm from the Defense Waste Processing Facility (DWPF). Historically, since the operation of DWPF, silicon in the recycle stream reacts with aluminum in the typical HLW to form sodium aluminosilicate mineral scale deposits in the evaporator pot and gravity drain line. The deposits are primarily nitrated sodium aluminosilicates with smaller amounts of clarkeite, Na((UO2)O(OH)). Nominally, the uranium in the scale is depleted in U-235 because the feed to the evaporator is depleted in U-235. In support of the criticality analysis, SRR obtained samples of evaporator scales from three different locations within the evaporator pot. SRR has requested that SRNL dissolve and analyze these samples for uranium and plutonium isotopic content and major radionuclide components. Crystallographic information for the samples was also requested.

The Technical Task Request (TTR) [1] and the Task Technical and Quality Assurance Plan [2] define the tasks and requirements for the performance of this characterization program by SRNL. In order to meet the SRR schedule needs, an earlier version of this report contained only the analytical results for the uranium isotopes, plutonium isotopes, and crystallographic information for the 2H-evaporator pot and wall scale samples. This revision includes the results of other radiochemical analyses that were performed to gather information useful in calculating the hydrogen generation rate.

Although the TTR for this task did not call for elemental constituents and mercury analyses by Inductively Coupled Plasma – Atomic Emission Spectroscopy (ICP-ES) and Cold Vapor Atomic Absorption for Mercury (CVAA-Hg), respectively, these analyses were included in the characterizations to satisfy environmental compliance requirements for this program.

2.0 Samples

On May 15, 2013, SRR sampled scale materials from three locations in the 2H-evaporator pot and delivered the samples to SRNL. The sample holders included two evaporator wall samples [HTF-13-78 and HTF-13-82] and one evaporator pot bottom sample [HTF-13-77]. The sample holders were opened on May 16, 2013. The evaporator pot bottom sample contained 3.99 grams of material while the wall sample [HTF-13-82] contained 9.81 grams of scale samples, as shown in Figure 1. The evaporator Wall sample holder which was supposed to contain sample HTF-13-78 was empty.

3.0 Experimental Procedure

Three “as-received” portions each of the evaporator pot bottom and wall scale samples (0.249 ± 0.035 g of the material in 100 mL dissolution volume giving an average digestion factor of 401.66 mL/g) were dissolved in the SRNL shielded cells using the sodium peroxide fusion digestion method. Uranium isotope analysis on the dissolved and diluted samples was obtained by using Inductively Coupled Plasma–Mass Spectrometry (ICP-MS). Plutonium isotopes Pu-

238, Pu-239/240 and Pu-241 were measured on the dissolved and diluted samples by thenoyltrifluoroacetone (TTA) separation and alpha Pulse Height Analysis (PHA) and beta Liquid Scintillation Counting (LSC). Small portions (0.06 gram) of the original scale samples were analyzed by X-Ray Diffraction (XRD) to determine the crystalline phases present. Other methods performed to measure the other radioactive and elemental constituents included gamma spectroscopy, Cs-removed gamma spectroscopy, Sr-90 analysis, Am/Cm analysis, Inductively Coupled Plasma – Atomic Emission Spectroscopy (ICP-ES) and Cold Vapor Atomic Absorption for Mercury (CVAA-Hg).

4.0 Results and Discussion

The XRD spectra for the two 2H evaporator scale samples are presented in Figures 2 and 3. XRD analysis confirmed that both the bottom cone section sample and wall sample from the 2H-evaporator consisted of clarkeite (a uranium oxy-hydroxide mineral) and nitrated Cancrinite (a crystalline sodium aluminosilicate solid) [3]. However, looking at the two XRD spectra (around 31^{o} (Two –Theta (degree))) there is an additional crystalline compound that was not positively identified with the XRD library. There appears to be more of this unidentified crystalline material in the wall sample.

Pu-239/240 analytical data was converted from activity units to wt% making the assumption that all activity measured as Pu-239/240 was from Pu-239. This assumption leads to somewhat higher masses of Pu-239 and total Pu. Since it is assumed that all measured Pu-239/240 activity is Pu-239, Pu-240 is not reported. Mass 238 from ICP-MS is acceptable for use as solely the U-238 mass because Pu-238 contributes an insignificant amount to that mass value. The total uranium values provided in the tables are sums of all uranium isotopes determined by ICP-MS.

Tables 1 and 2 contain the results for uranium and plutonium radionuclide analyses results. Analyses were performed in triplicate. A measure of uncertainty (±) as reported is the standard deviation of the multiple sample preparation analyses results. On "as- received" basis, the bottom pot section scale sample contained an average of 2.59E+00 ± *1.40E-01* wt % total uranium with a U-235 enrichment of 6.12E-01±*1.48E-02 %*, while the wall sample contained an average of 4.03E+00 ± *9.79E-01* wt % total uranium with a U-235 enrichment of 6.03E-01 ± *1.66E-02* %. The U-235 enrichment was calculated as the U-235 concentration divided by total uranium concentration.

Uranium-233 was the only uranium isotope in the pot scale sample which measured less than the instrument detection limit; averaging <1.88E-04 wt %. One of the triplicate U-233 runs in the wall sample measured less than the instrument detection limits (< 2.00E-04 wt %) while the other two runs were above the instrument detection limits (2.41E-04 and 2.12E-04 wt %).

The bottom pot section scale sample analysis results for Pu-238, Pu-239, and Pu-241 averaged 3.16E-05 ± 5.40E-06 wt %, 3.28E-04 ± 1.45E-05 wt %, and <8.80E-07 wt %, respectively. The Pu-241 analyses results, as presented, are upper limit values.

The evaporator wall scale sample analysis results for Pu-238, Pu-239, and Pu-241 averaged 3.74E-05 ± 6.01E-06 wt %, 4.38E-04 ± 5.08E-05 wt %, and <1.38E-06 wt %, respectively. Again, Pu-241 analyses results, as presented, are upper limit values.

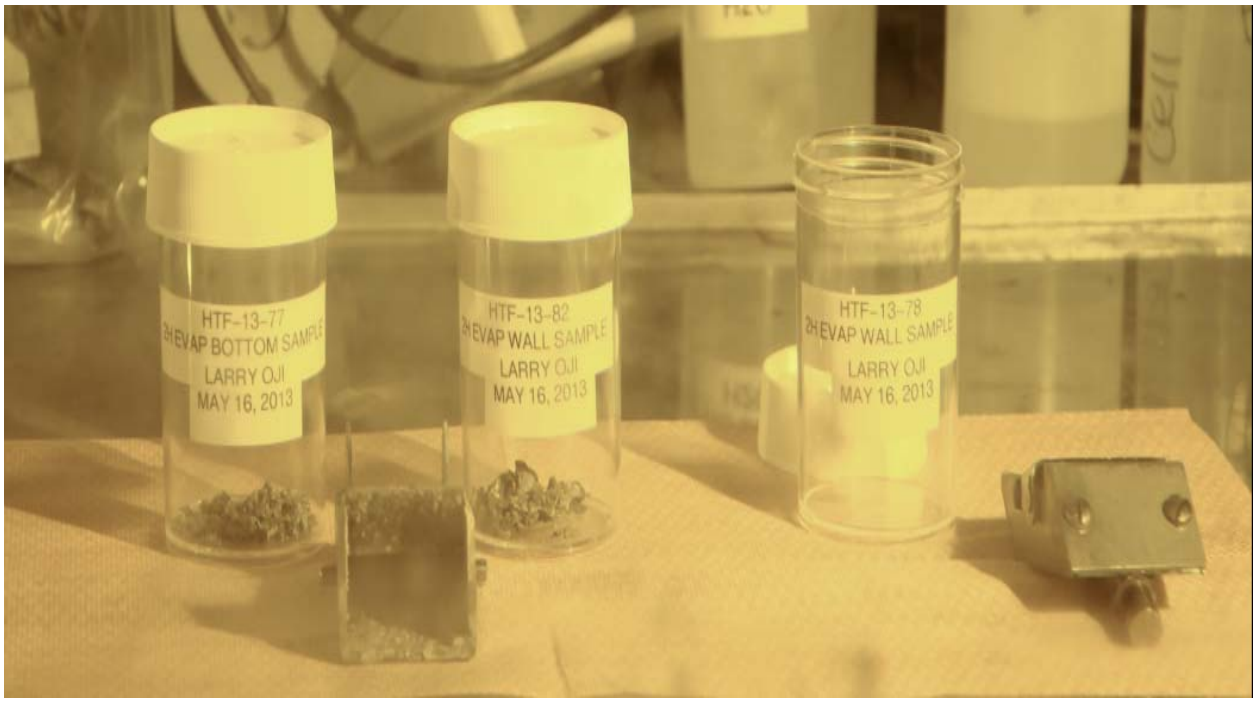

Figure 1 2H-Evaporator Samples; pot bottom sample HTF-13-77 and 2H-evaporator Wall scale samples HTF-13-82 and HTF-13-78. Wall sample HTF-13-78 contained no measurable sample.

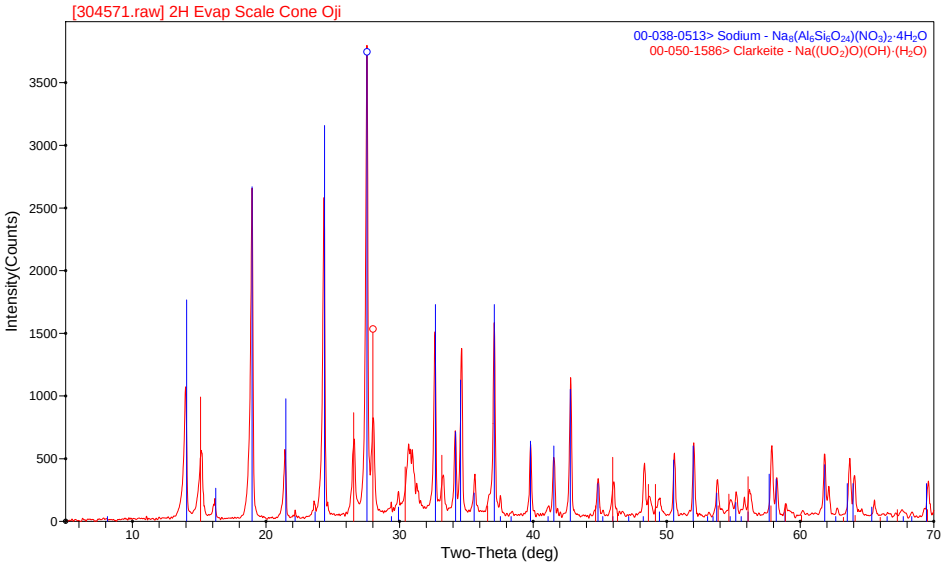

Figure 2 XRD analysis spectra result for 2H-evaporator pot bottom scale sample

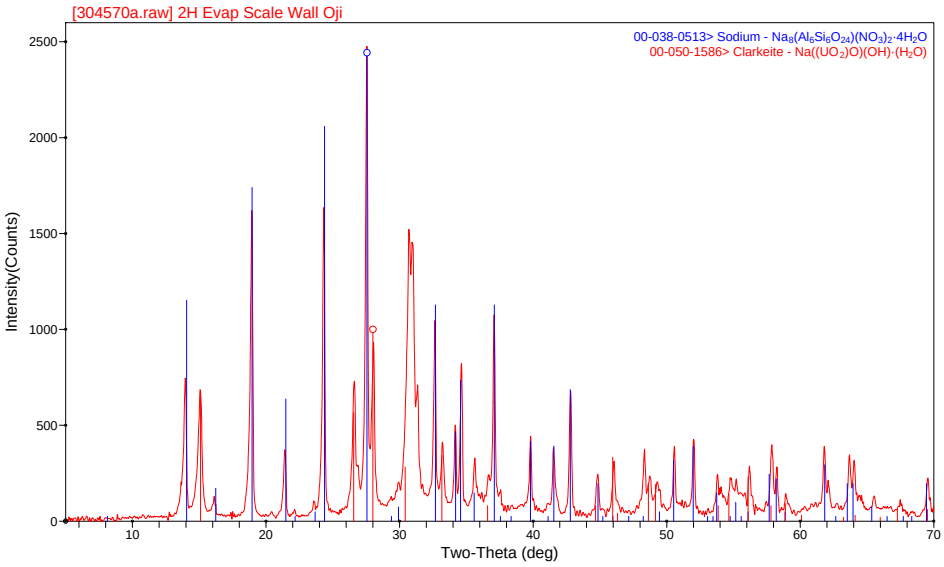

Figure 3 XRD analysis spectra result for 2H-evaporator wall scale sample

Table 1 Analysis of the 2H-Evaporator Pot Bottom Scale, HTF-13-77

Analyte	Method	Prep. 1	Prep. 2	Prep. 3	Average	St.dev.	Units
U-233	ICP-MS	<1.85E-04	<1.81E-04	<1.97E-04	**<1.88E-04**		wt %
U-234	ICP-MS	4.84E-04	4.36E-04	4.45E-04	**4.55E-04**	*2.55E-05*	wt %
U-235	ICP-MS	1.65E-02	1.55E-02	1.54E-02	**1.58E-02**	*6.19E-04*	wt %
U-236	ICP-MS	6.81E-04	7.55E-04	7.43E-04	**7.26E-04**	*4.00E-05*	wt %
U-238	ICP-MS	2.71E+00	2.56E+00	2.43E+00	**2.56E+00**	*1.41E-01*	wt %
U-Total	calculation	2.73E+00	2.58E+00	2.45E+00	**2.59E+00**	*1.40E-01*	wt %
U-235/Total U*100	calculation	6.05E-01	6.02E-01	6.29E-01	**6.12E-01**	*1.48E-02*	%
Pu-238	Pu238/241	3.13E-05	3.71E-05	2.63E-05	**3.16E-05**	*5.40E-06*	wt %
Pu-239*	Pu238/241	3.43E-04	3.14E-04	3.27E-04	**3.28E-04**	*1.45E-05*	wt %
**Pu-241	Pu238/241	<9.40E-07	<8.62E-07	<8.40E-07	**<8.80E-07**		wt %

*It is assumed that all measured Pu-239/240 activity is Pu-239. Thus, Pu-240 not reported.
** Upper limit.

Table 2 Analysis of the 2H-Evaporator Wall Scale, HTF-13-82

Analyte	Method	Prep.1	Prep. 2	Prep. 3	Average	St.dev	Units
U-233	ICP-MS	2.41E-04	<2.00E-04	2.12E-04	**≤2.18E-04**		wt %
U-234	ICP-MS	6.41E-04	4.08E-04	6.75E-04	**5.75E-04**	*1.45E-04*	wt %
U-235	ICP-MS	2.70E-02	1.79E-02	2.77E-02	**2.42E-02**	*5.49E-03*	wt %
U-236	ICP-MS	1.64E-03	9.35E-04	1.33E-03	**1.30E-03**	*3.52E-04*	wt %
U-238	ICP-MS	4.40E+00	2.89E+00	4.71E+00	**4.00E+00**	*9.77E-01*	wt %
U-Total	calculation	4.43E+00	2.91E+00	4.74E+00	**4.03E+00**	*9.79E-01*	wt %
U-235/Total U*100	calculation	6.10E-01	6.15E-01	5.84E-01	**6.03E-01**	*1.66E-02*	%
Pu-238	Pu238/241	4.05E-05	3.05E-05	4.13E-05	**3.74E-05**	*6.01E-06*	wt %
Pu-239*	Pu238/241	4.49E-04	3.83E-04	4.83E-04	**4.38E-04**	*5.08E-05*	wt %
**Pu-241	Pu238/241	<1.71E-06	<9.84E-07	<1.46E-06	**<1.38E-06**		wt %

*It is assumed that all measured Pu-239/240 activity is Pu-239. Thus, Pu-240 not reported.
** Upper limit.

5.0 Additional Radionuclide and Elemental Analysis

Tables 3 and 4 contain the results for the additional radionuclides analyzed for the individual 2H-evaporator pot scale samples (wall and pot bottom scales). For these two evaporator scale samples obtained at two different locations within the evaporator pot the major radioactive components (measured in wt %) in the additional radionuclide analyses were Sr-90, Cs-137, Np-237, Pu-239/240 and Th-232. In the analysis for Th-232 by ICP-MS it is assumed that the contribution by U-232 was negligible. Small quantities of americium and curium were detected in the blanks used for Am/Cm method for these radionuclides. These trace radionuclide amounts are assumed to come from airborne contamination in the shielded cell digestion or drying oven, which has been replaced. Therefore, the Am/Cm results presented in Tables 3 and 4 are definitely higher than the true expected values for the sample Am/Cm results.

Tables 5 and 6 contain the elemental constituents for the 2H evaporator wall scale sample and the 2H evaporator bottom pot sample, respectively. Tables 7 shows a compiled list of select radionuclides measurements from these 2013 2H evaporator measurements (pot bottom and wall samples) and the measurements for the same radionuclides in the 2010 2H evaporator analysis

(pot bottom and wall samples) [5]. In general, with comparative analyses values for Co-60, Eu-154 and Th-232 being the exceptions, radionuclide analysis for both years are quantitatively comparable; the analyses results are about the same order of magnitude.

The U-235 enrichments for both years remain depleted relative to natural uranium. The measurement results for Co-60 and Eu-154 show that there was less Co-60 and Eu-154 in the 2010 2H evaporator scale samples than in this current 2013 evaporator scale analyses. The difference in Eu-154 is more pronounced and is about an order of magnitude higher in 2010 measurement than in 2013. The analytical results for Th-232 in 2013 for the scale samples were measurably less than the values obtained in 2010 for Th-232 in the 2H-evaporator scale samples.

Note that the reporting units in Table 7 were chosen to be consistent with those of 2010 report, for ease of comparison. As such, the reporting units for the first group of radionuclides (U-233 through Pu-241) are wt % while the reporting units for the second group (Co-60 through Cm-244) are in dpm/g (unit conversions of the results given in Tables 3 and 4 yielded the 2013 dpm/g values identified in Table 7).

Table 3 Additional Analysis of the 2H-Evaporator Wall Scale Sample, HTF-13-82

Analyte	Method	Prep. 1	Prep. 2	Prep. 3	Average	St.Dev.	Units
Co-60	Cs-rem. γ	1.78E-10	1.58E-10	3.78E-10	**2.38E-10**	*1.22E-10*	Wt %
Sr-90	Sr-90	1.12E-03	7.43E-04	1.08E-03	**9.81E-04**	*2.07E-04*	Wt %
Y-90	Sr-90	2.81E-07	1.86E-07	2.71E-07	**2.46E-07**	*5.19E-08*	Wt %
Sb-125	Cs-rem. γ	<1.30E-09	1.23E-09	1.71E-09	**≤ 1.41E-09**		Wt %
Cs-137	γ scan	4.70E-05	4.55E-05	4.27E-05	**4.51E-05**	*2.19E-06*	Wt %
Ba-137m	γ scan	7.19E-12	6.96E-12	6.53E-12	**6.89E-12**	*3.35E-13*	Wt %
Ce-144	Cs-rem. γ	<2.85E-09	<2.23E-09	<2.75E-09	**<2.61E-09**		Wt %
Eu-154	Cs-rem. γ	7.89E-09	7.14E-09	1.37E-08	**9.56E-09**	*3.56E-09*	Wt %
Th-232	ICP-MS	3.28E-03	2.07E-03	4.35E-03	**3.23E-03**	*1.14E-03*	Wt %
Np-237	ICP-MS	3.69E-04	2.69E-04	4.26E-04	**3.55E-04**	*7.90E-05*	Wt %
*Pu-239/240	Pu238/241	4.49E-04	3.83E-04	4.83E-04	**4.38E-04**	*5.08E-05*	Wt %
♣Am-241	Am/Cm	4.37E-06	2.28E-06	5.25E-06	**3.97E-06**	*1.52E-06*	Wt %
♣Am-242m	Am/Cm	<1.82E-09	<2.73E-09	<1.77E-09	**<2.10E-09**		Wt %
♣Cm-242	Am/Cm	<4.43E-12	<6.62E-12	<4.29E-12	**<5.11E-12**		Wt %
♣Am-243	Am/Cm	<1.49E-06	<9.54E-07	<1.04E-06	**<1.16E-06**		Wt %
♣Cm-243	Am/Cm	<1.88E-08	<1.54E-08	<1.50E-08	**<1.64E-08**		Wt %
♣Cm-244	Am/Cm	1.04E-07	6.46E-08	1.12E-07	**9.35E-08**	*2.54E-08*	Wt %

* Assumes all measured Pu-239/240 activity is Pu-239. Thus, Pu-240 not reported

♣Small amounts of theses radionuclides were detected in the blanks; possibly from cell airborne contamination. These blank values influenced the detection limits. Therefore, the values reported here may be higher than the true Am/Cm values for these samples.

Table 4 Additional Analysis of the 2H-Evaporator Pot Bottom Scale Sample, HTF-13-77

Analyte	Method	Prep. 1	Prep. 2	Prep. 3	Average	St.Dev.	Units
Co-60	Cs-rem. γ	<1.27E-10	<1.13E-10	<1.18E-10	**<1.19E-10**		Wt %
Sr-90	Sr-90	5.98E-04	6.11E-04	5.52E-04	**5.87E-04**	*3.12E-05*	Wt %
Y-90	Sr-90	1.50E-07	1.53E-07	1.38E-07	**1.47E-07**	*7.82E-09*	Wt %
Sb-125	Cs-rem. γ	<1.25E-09	<9.56E-10	<1.14E-09	**<1.12E-09**		Wt %
Cs-137	γ scan	5.93E-05	5.93E-05	6.11E-05	**5.99E-05**	*1.05E-06*	Wt %
Ba-137m	γ scan	9.07E-12	9.07E-12	9.35E-12	**9.16E-12**	*1.60E-13*	Wt %
Ce-144	Cs-rem. γ	<2.05E-09	<1.69E-09	<1.85E-09	**<1.86E-09**		Wt%
Eu-154	Cs-rem. γ	5.06E-09	6.16E-09	4.24E-09	**5.15E-09**	*9.63E-10*	Wt%
Th-232	ICP-MS	1.00E-03	1.09E-03	1.12E-03	**1.07E-03**	*6.13E-05*	Wt%
Np-237	ICP-MS	2.47E-04	2.02E-04	2.94E-04	**2.48E-04**	*4.57E-05*	Wt %
*Pu-239/240	Pu238/241	3.43E-04	3.14E-04	3.27E-04	**3.28E-04**	*1.45E-05*	Wt %
♣Am-241	Am/Cm	1.52E-06	1.69E-06	1.59E-06	**1.60E-06**	*8.61E-08*	Wt %
♣Am-242m	Am/Cm	<9.83E-10	<9.78E-10	<1.38E-09	**<1.11E-09**		Wt %
♣Cm-242	Am/Cm	<2.40E-12	<2.38E-12	<3.34E-12	**<2.71E-12**		Wt %
♣Am-243	Am/Cm	<1.45E-06	<9.61E-07	<6.22E-07	**<1.01E-06**		Wt %
♣Cm-243	Am/Cm	<1.58E-08	<6.67E-09	<9.16E-09	**<1.05E-08**		Wt %
♣Cm-244	Am/Cm	6.18E-08	7.13E-08	7.91E-08	**7.07E-08**	*8.64E-09*	Wt %

* Assumes all measured Pu-239/240 activity is Pu-239. Thus, Pu-240 not reported

♣Small amounts of theses radionuclides were detected in the blanks; possibly from cell airborne contamination. These blank values influenced the detection limit. Therefore, the values reported here may be higher than the true Am/Cm values for these samples.

Table 5 Elemental Constituents of Wall 2H Evaporator Scale Sample

Component	2H EVAP SCALE W_ES 1	2H EVAP SCALE W_ES 2	2H EVAP SCALE W_ES 3	Average	Stdev	Units
Ag	<1.37E-02	<1.40E-02	<1.40E-02	**<1.39E-02**		*Wt %*
Al	1.13E+01	8.61E+00	1.00E+01	**9.97E+00**	*1.35E+00*	*Wt %*
B	5.74E-03	3.86E-03	4.51E-03	**4.70E-03**	*9.55E-04*	*Wt %*
Ba	1.82E-03	1.86E-03	1.94E-03	**1.87E-03**	*6.11E-05*	*Wt %*
Be	<9.49E-04	<9.72E-04	<9.72E-04	**<9.64E-04**		*Wt %*
Ca	1.92E-01	1.81E-01	1.72E-01	**1.82E-01**	*1.00E-02*	*Wt %*
Cd	1.13E-03	1.31E-03	1.16E-03	**1.20E-03**	*9.64E-05*	*Wt %*
Ce	<6.47E-02	<6.63E-02	<6.63E-02	**<6.58E-02**		*Wt %*
Co	<1.46E-03	<1.50E-03	<1.50E-03	**<1.49E-03**		*Wt %*
Cr	6.28E-03	9.68E-03	6.76E-03	**7.57E-03**	*1.84E-03*	*Wt %*
Cu	<7.75E-03	<7.94E-03	<7.94E-03	**<7.88E-03**		*Wt %*
Fe	1.15E-01	1.44E-01	1.74E-01	**1.44E-01**	*2.95E-02*	*Wt %*
Gd	<5.21E-02	<5.34E-02	<5.34E-02	**<5.30E-02**		*Wt %*
Hg	9.36E+00	8.62E+00	1.08E+01	**9.59E+00**	*1.11E+00*	*Wt %*
K	<1.88E-02	<1.93E-02	<1.93E-02	**<1.91E-02**		*Wt %*
La	<9.96E-03	<1.02E-02	<1.02E-02	**<1.01E-02**		*Wt %*
Li	1.11E-03	1.04E-03	1.34E-03	**1.16E-03**	*1.57E-04*	*Wt %*
Mg	2.84E-03	3.60E-03	1.18E-03	**2.54E-03**	*1.24E-03*	*Wt %*
Mn	6.06E-03	5.73E-03	1.25E-02	**8.10E-03**	*3.82E-03*	*Wt %*
Mo	<7.77E-03	<7.96E-03	<7.96E-03	**<7.90E-03**		*Wt %*
Na	1.29E+01	1.02E+01	1.17E+01	**1.16E+01**	*1.35E+00*	*Wt %*
Ni	1.37E-02	1.63E-02	2.04E-02	**1.68E-02**	*3.38E-03*	*Wt %*
P	<9.60E-03	<9.84E-03	<9.84E-03	**<9.76E-03**		*Wt %*
Pb	<6.47E-03	<6.62E-03	<6.62E-03	**<6.57E-03**		*Wt %*
S	<4.74E-01	<4.86E-01	<4.86E-01	**<4.82E-01**		*Wt %*
Sb	<2.72E-02	<2.79E-02	<3.52E-02	**<3.01E-02**		*Wt %*
Sn	<2.86E-02	<2.93E-02	<2.93E-02	**<2.91E-02**		*Wt %*
Sr	6.85E-03	7.03E-03	7.49E-03	**7.12E-03**	*3.30E-04*	*Wt %*
Th	<5.17E-02	<5.30E-02	<5.30E-02	**<5.26E-02**		*Wt %*
Ti	<4.59E-03	<4.70E-03	<4.70E-03	**<4.66E-03**		*Wt %*
U	5.10E+00	5.13E+00	5.65E+00	**5.29E+00**	*3.09E-01*	*Wt %*
V	<4.98E-03	<5.10E-03	<5.10E-03	**<5.06E-03**		*Wt %*
Zn	2.26E-03	3.19E-03	1.54E-03	**2.33E-03**	*8.27E-04*	*Wt %*
Zr	<3.87E-03	<3.97E-03	<3.97E-03	**<3.94E-03**		*Wt %*

Table 6 Elemental Constituents of Pot Bottom 2H Evaporator Scale Sample

Component	2H EVAP SCALE C_ES 1	2H EVAP SCALE C_ES 2	2H EVAP SCALE C_ES 3	Average	Stdev	Units
Ag	<1.40E-02	<1.41E-02	<1.41E-02	**<1.41E-02**		*Wt%*
Al	1.35E+01	1.26E+01	1.38E+01	**1.33E+01**	*6.24E-01*	*Wt%*
B	4.03E-03	4.38E-03	2.41E-03	**3.61E-03**	*1.05E-03*	*Wt%*
Ba	1.08E-03	8.13E-04	1.43E-03	**1.11E-03**	*3.09E-04*	*Wt%*
Be	<9.72E-04	<9.76E-04	<9.76E-04	**<9.75E-04**		*Wt%*
Ca	1.17E-01	7.72E-02	1.21E-01	**1.05E-01**	*2.42E-02*	*Wt%*
Cd	<1.16E-03	<1.16E-03	<1.16E-03	**<1.16E-03**		*Wt%*
Ce	<6.63E-02	<6.66E-02	<6.66E-02	**<6.65E-02**		*Wt%*
Co	<1.50E-03	<1.50E-03	<1.50E-03	**<1.50E-03**		*Wt%*
Cr	4.12E-03	2.53E-03	3.51E-03	**3.39E-03**	*8.02E-04*	*Wt%*
Cu	<7.94E-03	<7.97E-03	<7.97E-03	**<7.96E-03**		*Wt%*
Fe	3.35E-02	3.98E-02	7.69E-02	**5.01E-02**	*2.35E-02*	*Wt%*
Gd	<5.34E-02	<5.36E-02	<5.36E-02	**<5.35E-02**		*Wt%*
Hg	5.01E+00	7.29E+00	4.70E+00	**5.67E+00**	*1.41E+00*	*Wt%*
K	<1.93E-02	<1.93E-02	<1.93E-02	**<1.93E-02**		*Wt%*
La	<1.02E-02	<1.02E-02	<1.02E-02	**<1.02E-02**		*Wt%*
Li	8.74E-04	9.27E-04	7.40E-04	**8.47E-04**	*9.64E-05*	*Wt%*
Mg	3.48E-04	5.10E-03	6.91E-04	**2.05E-03**	*2.65E-03*	*Wt%*
Mn	1.02E-03	1.11E-03	1.69E-03	**1.27E-03**	*3.64E-04*	*Wt%*
Mo	<7.96E-03	<7.99E-03	<7.99E-03	**<7.98E-03**		*Wt%*
Na	1.52E+01	1.41E+01	1.55E+01	**1.49E+01**	*7.37E-01*	*Wt%*
Ni	<8.79E-02	<8.82E-02	<8.82E-02	**<8.81E-02**		*Wt%*
P	<9.84E-03	<9.88E-03	<9.88E-03	**<9.87E-03**		*Wt%*
Pb	<7.10E-03	<6.67E-03	<6.65E-03	**<6.81E-03**		*Wt%*
S	<4.86E-01	<4.88E-01	<4.88E-01	**<4.87E-01**		*Wt%*
Sb	<2.79E-02	<2.80E-02	<2.80E-02	**<2.80E-02**		*Wt%*
Sn	<2.93E-02	<2.94E-02	<2.94E-02	**<2.94E-02**		*Wt%*
Sr	3.49E-03	2.60E-03	3.57E-03	**3.22E-03**	*5.38E-04*	*Wt%*
Th	<5.30E-02	<5.32E-02	<5.32E-02	**<5.31E-02**		*Wt%*
Ti	<4.70E-03	<4.72E-03	<4.72E-03	**<4.71E-03**		*Wt%*
U	3.23E+00	1.93E+00	2.84E+00	**2.67E+00**	*6.67E-01*	*Wt%*
V	<5.10E-03	<5.12E-03	<5.12E-03	**<5.11E-03**		*Wt%*
Zn	1.32E-03	1.33E-03	1.88E-03	**1.51E-03**	*3.20E-04*	*Wt%*
Zr	<3.97E-03	<3.98E-03	<3.98E-03	**<3.98E-03**		*Wt%*

Table 7 Comparison of select average radionuclides concentrations for 2010 and 2013 2H evaporator scale characterizations

Radionuclide	Bottom Pot sample-2013	Wall sample-2013	Bottom Pot sample-2010	Wall sample-2010
U-233, wt%	<1.88E-04	≤ 2.18E-04	<1.10E-03	<1.07E-03
U-234,wt%	4.55E-04	5.75E-04	<5.48E-04	≤ 5.76E-04
U-235, wt%	1.58E-02	2.42E-02	7.17E-03	8.84E-03
U-236, wt%	7.26E-04	1.30E-03	≤ 5.92E-04	5.89E-04
U-238, wt%	2.56E+00	4.00E+00	1.23E+00	1.45E+00
Total U, wt%	2.59E+00	4.03E+00	1.24E+00	1.46E+00
U-235/Total U, (%)	6.12E-01	6.03E-01	5.8E-01	6.1E-01
Pu-238, wt%	3.16E-05	3.74E-05	7.02-05	8.15E-05
*Pu-239, wt%	3.28E-04	4.38E-04	5.88E-04	6.88E-04
**Pu-241, wt%	<8.88E-07	<1.38E-06	2.44E-06	2.85E-06
Co-60, dpm/g	<3.00E+03	5.98E+03	1.92E+04	2.28E+04
Sr-90, dpm/g	1.78E+09	2.97E+09	1.40E+09	1.42E+09
Cs-137, dpm/g	1.16E+08	8.70E+07	1.06E+08	8.38E+07
Eu-154, dpm/g	3.09E+04	5.73E+04	2.03E+05	2.09E+05
Th-232, dpm/g	2.61E+00	7.87E+00	1.73E+01	1.69E+01
Np-237, dpm/g	3.88E+03	5.55E+03	≤ 2.26E+04	7.73E+03
Am-241, dpm/g	1.22E+05	3.02E+05	8.89E+05	1.20E+06
Cm-242, dpm/g	<1.99E+02	<3.75E+02	8.53E+02	1.30E+03
Cm-244, dpm/g	1.27E+05	1.68E+05	9.57E+05	1.19E+06

*Assumes all measured Pu-239/240 activity is Pu-239
** Upper limit

6.0 Conclusions

The 2H Evaporator scale samples from the wall and bottom sections of the 2H-evaporator pot [bottom pot scale sample (HTF-13-77) and wall sample (HTF-13-82)] are composed of nitrated cancrinite (a crystalline sodium aluminosilicate solid) and clarkeite (a uranium oxy-hydroxide mineral).

On "as-received" basis, the bottom pot scale sample contained an average of 2.59E+00 ± *1.40E-01* wt % total uranium with a U-235 enrichment of 6.12E-01 % ± *1.48E-02*, while the wall sample contained an average of 4.03E+00 ± *9.79E-01* wt % total uranium with a U-235 enrichment of 6.03E-01% ± *1.66E-02* wt %. These results confirm that the uranium contained in the scale remains depleted with respect to natural uranium.

With a few exceptions, a comparison of select radionuclides measurements from this 2013 2H evaporator scale characterization (pot bottom and wall scale samples) with those measurements for the same radionuclides in the 2010 2H evaporator scale analysis shows that the radionuclide analysis for both years are quantitatively comparable; the analyses results are about the same order of magnitude. The U-235 enrichments for both years remain depleted relative to natural uranium. There seems to be less Th-232 in the 2013 analysis results for the 2H scale evaporator samples (pot bottom and wall scale samples) in comparison to the 2010 2H scale evaporator samples analysis.

Because all the 2013 scale samples from the 2H evaporator so far analyzed contained a significant amount of elemental mercury, it is recommended that analysis for mercury be included in future Technical Task Requests on evaporator sample analysis at SRNL to track mercury accumulation in the evaporator scales as well as help satisfy environmental compliance requirements for this program.

7.0 Quality Assurance

This work was performed in accordance with the requirements of the task technical request [1]. The experimental data are for these analyses are contained in Laboratory Notebook SRNL-NB-2013-00026 and various Analytical Development (AD) notebooks.

Requirements for performing reviews of technical reports and the extent of review are established in manual E7 2.60. SRNL documents the extent and type of review using the SRNL Technical Report Design Checklist contained in WSRC-IM-2002-00011, Rev. 2.

8.0 References

1. H. Bui, "2H Evaporator Scale Sampling Analysis," X-TTR-H-00024, Rev. 0, May 8, 2013.

2. L N. Oji, "Task Technical and Quality Assurance Plan for 2H Evaporator Scale Sample Analysis and Dissolution Studies" SRNL-RP-2013-00289, Rev. 0, May 2013.

3. J. C. Buhl. F. Stief, M. Fechtelkord, T. M. Gesing, U. Taphorn, and C. Taake, "Synthesis, X-ray diffraction and MAS NMR characteristics of nitrate cancrinite $Na_{7.6}[AlSiO4]_6(NO3)_{1.6}(H2O)_2$," Journal of Alloys and Compounds, 305, 93-102, (2000).

4. K. B. Martin and C. J. Martino, "The 2H Evaporator Pot & Gravity Drain Line Scale Nuclear Criticality Analysis Suite," WSRC-STI-2008-00185, Rev. 0, April 21,

5. C. J. Martino, "Analysis of 2H-Evaporator Scale Samples (HTF-10-116 and HTF-10-117)," SRNL-TR-2010-00264, Rev.1, November 4, 2010.