WSRC-TR-2002-00244

Results of Analyses of Tank 37H Criticality Salt Samples (HTK-493 and 494)

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This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-96SR18500 with the U.S. Department of Energy.

Keywords: Tank Farm, Characterization, Criticality

Summary

High Level Waste Division (HLWD) personnel requested Savannah River Technology Center (SRTC) support in analyzing samples of salt cake from Tank 37H as input to a Nuclear Criticality Safety Evaluation (NCSE) in support of dissolution of the salt in the tank.' Two salt core samples (HTK-493 and 494) were pulled from Tank 37H and transferred to the SRTC Shielded Cells Facility on April 2, 2002 for analyses of species that may affect criticality as the salt is dissolved. The core samples were composited and prepared for analysis of the as-received sample and of insoluble solids separated from as-received sample. Results of the analyses are presented herein. Results are given for the as-received salt cake, for insoluble solids separated from the core sample both with and without a correction for soluble solids remaining after dissolution, and for wash water produced by dissolving the soluble salts in inhibited water.

The uranium isotopic enrichment is consistent among the current analyses (10.0 � 10.1 % U-235 to total U for both as-received and insoluble solids) and higher than analyses from samples pulled in 2001 (2.67 % U-235 in the as-received solids and 5.30 % U-235 in the insoluble solids). The total uranium in the as-received sample was ~0.006 wt %. There was ~0.07 wt % total uranium in the dry insoluble solids.

The total plutonium in the as-received sample was ~3E-04 wt %. There was 4E-03 wt % total plutonium in the insoluble solids.

There continues to be significant quantities of neutron poisons (sodium, iron, chromium, manganese and zinc) present in the as-received sample to mitigate the potential for nuclear criticality. The neutron poisons are also present in the insoluble solids, though at a considerably reduced level. Sodium in the as-received sample is present at 17,700 times the equivalent uranium-235. In the insoluble solids, the sodium to equivalent uranium-235 is only ~120 as compared with a desired ratio of 150.

Introduction

The High Level Waste Division (HLWD) plans to modify Tank 37H to be used as a concentrate drop tank for the 241-25H (3H) Evaporator from its current use for salt cake storage. The salt and supernate levels in Tank 37H when the samples were pulled were 252" and 289", respectively. The operating plan for the use of Tank 37H requires that a portion of the salt cake (50" to 100") be removed to provide operating volume for the 3H Evaporator system. A transfer of about 61" of supernate from Tank 37 into Tank 32 was completed prior to collection of the sample so that inhibited water could be added to begin salt dissolution. Additional salt solution transfers and inhibited water additions will be performed to continue to remove salt from the tank. In support of this salt dissolution, an NCSE is needed to assure criticality is not attained. Two salt core samples were taken to analyze the Tank 37H salt cake for criticality concerns by performing High Level Waste (HLW) sample analysis suites 1 and 12 without analysis for sodium chromate. These analyses were performed under task technical and quality assurance plan WSRC-RP-2002-00213.

Discussion

Sampling Method

In order to complete the NCSE required for dissolution of the ~100 inches salt cake, it was desired to have a sample of the insoluble solids representative of the solids from the entire 100-inch depth. For this reason, a novel sampling approach was developed for the current sample. First a caisson was mined into the salt. Water was then used to dissolve salt down to a depth of ~100 inches from the top of the caisson (about 165 inches from the tank bottom). Three days were allowed for the insoluble solids to settle to the bottom of the caisson. Two standard core samples were then pulled from the bottom of the caisson. Sample HTK-493 was pulled from a Tank elevation of 160" or 102" below the salt cake surface in the tank. HTK-494 was pulled from a tank elevation of 150" or 112" below the salt cake surface. This new sampling method also had the advantage of providing more insoluble solids for analysis than previous surface core samples.

Sample Preparation

Upon arrival at the SRTC, the samples were placed in the Shielded Cells Facility and opened. HTK-493 (the first core sample pulled) contained ~50 g of sample, and HTK-494 contained ~10 g of sample (Figure 1). The samples were wet, resembling mud, with HTK-493 appearing to contain somewhat more water than HTK-494. The samples were combined and ground and homogenized using a mortar and pestle (Figure 2). Three aliquots of the as-received sample were pulled for aqua regia dissolution and three were pulled for sodium peroxide fusion dissolution. Approximately 0.5 g of original sample material was dissolved into 250 mL of liquid. The dissolved samples were removed from the Shielded Cells and transferred to the Analytical Development Section (ADS) for analysis for actinides and neutron poisons. Analyses conducted include inductively coupled plasma-emission spectrometry (ICP-ES) for various elemental species including neutron poisons, inductively coupled plasma-mass spectrometry (ICP-MS) for various actinide isotopes, Pu-238/241 analysis for Pu-238 and Pu-241, Tank 50 Rad Screen for total alpha and total beta, gamma scan for gamma emitting radionuclides, and Sr-90 analysis. Additional analyses for arsenic and selenium by atomic absorption (AA) and mercury by cold vapor atomic absorption (CVAA) were requested to satisfy requirements for drain disposal of waste.

Prior to digesting the aliquots with sodium peroxide, those samples were dried. (The sodium peroxide reacts with water before the digestion is completed. Drying is not necessary for samples dissolved using aqua regia.) This drying step allowed a measurement of the weight fraction total solids in the sample.

The insoluble solids were separated from the salt cake sample by washing each of 4 10-g aliquots of the as-received sample three times with inhibited water (0.015 NaOH) to dissolve the soluble material. After each wash, the samples were centrifuged to minimize the solids volume, and the clear supernate was decanted. Approximately 30 mL of inhibited water was used for each of the three washes in each of the four centrifuge tubes. The density of the as-received salt was estimated by noting the displacement of the 10 g of salt during the first addition o inhibited water.

After the third wash, the remaining insoluble solids in each of the tubes were dried. Three aliquots each of the dried solids were then dissolved using aqua regia and sodium peroxide fusion digestion. Approximately 0.25 g of dried insoluble solids was dissolved into 250 mL of liquid in each of the dissolutions. The dried insoluble solids were submitted for the same analyses as the as-received sample aliquots. Additionally, a small sample of the dried solids was retained for future x-ray diffraction analysis of crystalline species.

Three samples of the water from each of the three washes were diluted to reduce the dose rate to allow removal from the Shielded Cells. Aliquots of the first two washes were diluted by a factor of about 100. Aliquots of the third wash were diluted by a factor of about 10. These samples were submitted to ADS for the same analyses as the as-received samples. Additionally, those samples were submitted for ion chromatography (IC) for various anions and for titration for free hydroxide, aluminate and carbonate.

If there had been any free liquid sample associated with the original sample, it was planned to dilute and analyze a sample of that material using the same analyses as for the wash water samples. However, on opening the sample, no easily separable free liquid was noted.

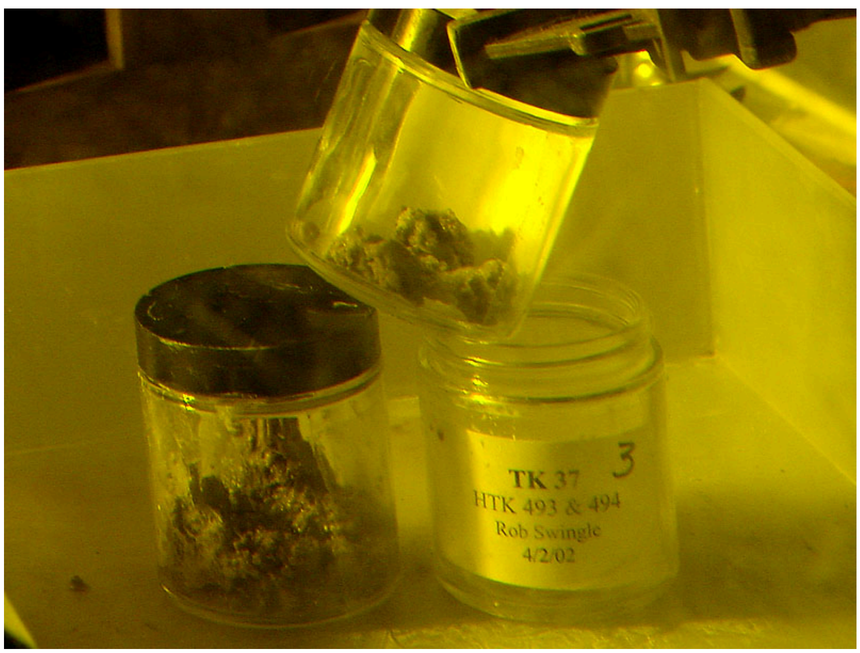

Figure 1. Tank 37H Core Samples (HTK-493 is at Bottom Left, HTK-494 is at Top)

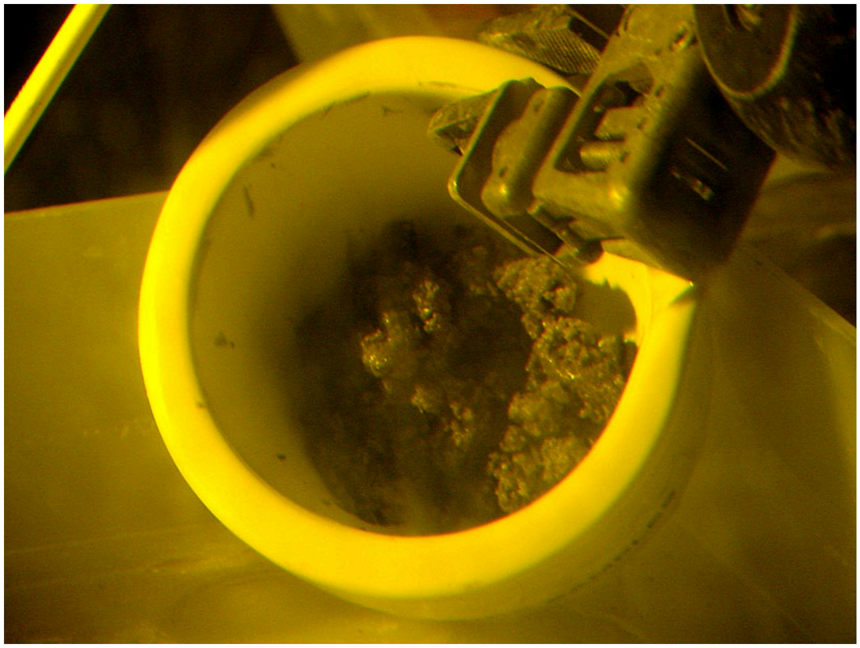

Figure 2. Combined Samples (HTK-493 and HTK-494) in Mortar

Results

Results for as-received solid sample are given in wt % of the wet sample with the exception of certain non-actinide radionuclide results. Results for insoluble solids are given in terms of dried solids. The results are given as an average of three determinations for each digestion method, along with a one-sigma standard deviation. A combined average and standard deviation for both methods are also given. In some cases, a combined average is not appropriate (where analysis of samples from one or the other dissolution method are known or expected to be significantly better than the other or when results are not available for one or the other dissolution method). In those cases, a best value is given instead of the combined average. Standard deviations are not given for values which are below detection limits ("<" values) or for those where multiple determinations resulted in a mixture of actual values and below detection limit values ("≤ " values). In addition to the analytical results reported by ADS, several calculated values are given: total uranium, total plutonium, U-235/total uranium, equivalent U-235, and the ratios of several neutron poison concentrations (Na, Fe, Cr, Mn and Zn) to total uranium. The equivalent U-235 weight percent given in the table is calculated from the weight percentages of the fissile radionuclides with the following equation.

Eq. U-235 = U-235 + 4 * (U-233 + Pu-239 + Pu-241).

Values that are below detection limits are not included in the calculation of total uranium, total plutonium or equivalent U-235.

Results for the composited samples as they were received are given in Table 1. The solids were found to comprise 66.7 $\pm$ 4.3 wt % of the sample. The density of the original material was measured to be 1.6 $\pm$ 0.1 g/mL during the first wash. The results in Table 1 are given in terms of weight of wet sample.

Table 1. Results of Analyses of As-Received Sample (HTK-493 and HTK-494)

	Aqua Regia Dissolution		Peroxide Fusion Digestion		Combined	
Species	Avg.	St. Dev.	Avg.	St. Dev.	Avg.	St. Dev.
ICP-ES (wt %)						
Agα	<2.59E-03	-	na	na	<2.59E-03	-
Al						

	2.33	0.17	2.23	0.25	2.28	0.20
Bχ	<1.56E-03	-	<1.17E-02	-	<1.56E-03	-
Baχ	3.01E-03	4.4E-04	<7.93E-03	-	3.01E-03	4.4E-04
Caε	8.26E-03	1.14E-03	na	na	8.26E-03	1.14E-03
Cd	2.18E-03	7.0E-04	≤ 1.76E-03	-	≤ 1.97E-03	-
Coα	≤ 1.79E-03	-	na	na	≤ 1.79E-03	-
Crβ	4.23E-02	1.5E-03	3.55E-02	3.8E-03	3.82E-02	4.6E-03
Cuε	<1.56E-03	-	na	na	<1.56E-03	-
Feβ	8.66E-02	3.45E-02	7.84E-02	1.26E-02	8.17E-02	1.99E-02
La	<5.17E-03	-	<5.51E-03	-	<5.34E-03	-
Liχ	<1.56E-03	-	<1.80E-02	-	<1.56E-03	-
Mgχ	1.45E-03	1.1E-04	<2.21E-03	-	1.45E-03	1.1E-04
Mnβ ,ϕ	8.28E-03	4.8E-04	≤ 8.64E-04	-	≤ 3.83E-03	-
Moχ	<1.56E-03	-	<4.54E-03	-	<1.56E-03	-
Naα	25.8	0.6	na	na	25.8	0.6
Ni	<4.46E-03	-	<5.64E-03	-	<5.17E-03	-
P	5.78E-02	4.2E-03	<6.1E-02	-	≤ 5.94E-02	-
Pbχ	<2.38E-02	-	<1.33E-02	-	<1.33E-02	-
Siδ	na	na	0.902	0.261	0.902	0.261
Sn	<8.28E-03	-	1.10E-02	1.3E-03	≤ 9.66E-03	-
Srχ	<5.17E-04	-	<3.17E-03	-	<5.17E-04	-
Ti	1.30E-03	2.9E-04	≤ 3.82E-03	-	≤ 2.56E-03	-
Vα	<1.56E-03	-	na	na	<1.56E-03	-
Znε	<1.56E-03	-	na	na	<1.56E-03	-
Zrα	<1.56E-03	-	na	na	<1.56E-03	-
Gamma Scan (dpm/g)						
Cs-137	6.86E08	3.8E07	8.98E08	1.66E08	7.92E08	1.58E08
Tk 50 Rad Screen (dpm/g)						
Alpha	3.01E07	1.36E07	3.44E07	9.4E06	3.22E07	1.07E07
Beta						

	1.63E09	8E07	1.89E09	2.8E08	1.76E09	2.3E08
Sr-90 (dpm/g)						
Sr-90	3.33E08	4.6E07	3.44E08	8.4E07	3.38E08	6.1E07
Pu-238/241 (wt %)						
Pu-238	5.24E-05	1.8E-06	4.47E-05	5.7E-06	4.86E-05	5.7E-06
Pu-241	6.39E-06	1.33E-06	5.19E-06	6.0E-07	5.79E-06	1.13E-06
ICP-MS (wt %)						
M-232 (Th)	3.96E-05	7.4E-06	5.45E-05	5.6E-06	4.70E-05	1.00E-05
M-233 (U)	1.78E-05	3.8E-06	1.92E-05	5.1E-06	1.85E-05	4.1E-06
M-234 (U)	5.15E-05	8.5E-06	5.58E-05	7.9E-06	5.37E-05	7.7E-06
M-235 (U)	5.65E-04	6.5E-05	6.32E-04	7.2E-05	5.98E-04	7.1E-05
M-236 (U)	2.59E-04	3.7E-05	3.03E-04	2.3E-05	2.81E-04	3.6E-05
M-237 (Np)	1.80E-05	4.4E-06	2.26E-05	3.0E-06	2.03E-05	4.2E-06
M-238 (**U**, Pu)	4.71E-03	6.5E-04	5.24E-03	5.2E-04	4.98E-03	6.0E-04
M-239 (Pu)	2.01E-04	2.0E-05	2.35E-04	1.6E-05	2.18E-04	2.4E-05
M-240 (Pu)	3.43E-05	5.6E-06	3.78E-05	5.3E-06	3.60E-05	5.2E-06
M-241 (Pu, Am)	1.80E-05	3.0E-06	2.27E-05	3.5E-06	2.04E-05	3.9E-06
M-242 (Pu)	<9.78E-06	-	<7.89E-06	-	<8.83E-06	-
M-243 (Am, Cm)	<9.78E-06	-	<7.89E-06	-	<8.83E-06	-
M-244 (Pu, Cm)	<9.78E-06	-	<7.89E-06	-	<8.83E-06	-
M-245 (Cm)	<9.78E-06	-	<7.89E-06	-	<8.83E-06	-
M-246 (Cm)	<9.78E-06	-	<7.89E-06	-	<8.83E-06	-
M-247 (Cm, Bk)	<9.78E-06	-	<7.89E-06	-	<8.83E-06	-
Calculated Values (wt %)						
Equivalent U-235	1.47E-03	1.6E-04	1.67E-03	8E-05	1.57E-03	1.6E-04
Total U	5.60E-03	7.6E-04	6.25E-03	6.3E-04	5.93E-03	7.2E-04
Total Pu	2.94E-04	2.7E-05	3.22E-04	1.7E-05	3.08E-04	2.5E-04
Ratios (Unitless)						
U-235/Total U	0.101	0.003	0.101	0.001	0.101	0.002
Na/Eq. U-235α	1.77E04	1.6E03	na	na	1.77E04	1.6E03

Fe/Eq. U-235β	58.7	29.7	47.2	9.5	51.8	17.4
Cr/Eq. U-235β	27.9	2.3	21.3	1.9	23.9	4.0
Zn/Eq. U-235ε	<1.07	-	na	na	<1.07	-
Mn/Eq. U-235β,ϕ	5.47	3.32	≤ 0.510	-	≤ 2.49	-

α No peroxide fusion result is provided by ADS for these species.

β One of the aqua regia dissolution samples apparently contained a particle that was high in iron (Fe), manganese (Mn), chromium (Cr) and nickel (Ni), perhaps resulting from stainless steel. Because it is not clear that this sample was representative of the salt cake as a whole and because it is conservative from a criticality standpoint to do so, this result was left out of the averages for Fe, Cr, Mn and Ni.

χ One of the two dissolution methods gave a result that was significantly lower than a minimum detection limit given by the other dissolution method. In these cases, the lower result is not considered to be in conflict with the higher minimum detection limit and is considered to be the better number.

δ Peroxide fusion is known to provide a much better dissolution of Si than aqua regia. The peroxide fusion number is therefore considered to be the better number, and no aqua regia number is given.

ε Blank samples run indicate for some trace species that the peroxide fusion dissolutions show some sample contamination or analytical interference. For these samples the aqua regia dissolution result is considered the better number.

ϕ In addition to the one suspect data point for Mn from the aqua regia dissolution, the peroxide fusion results are scattered and therefore suspect.

Results for the washed insoluble solids are given in Tables 2 and 3. Table 2 contains the results for the insoluble solids that have not been corrected for soluble solids that may have been left behind after the third wash. Table 3 contains results for the insoluble solids that have been corrected to remove the soluble solids. The correction factor was calculated by multiplying the amount of water evaporated from each sample by the concentrations from the third wash (Table 4) for that sample. The correction factor was subtracted from the raw result for the insoluble solids given in Table 2. The results for both Tables 2 and 3 are given in terms of dried insoluble solids.

Table 2. Results of Analyses of Insoluble Solids (HTK-493 and HTK-494 � Uncorrected for Soluble Solids)

	Aqua Regia Dissolution		Peroxide Fusion Digestion		Combined	
Species	Avg.	St. Dev.	Avg.	St. Dev.	Avg.	St. Dev.
ICP-ES (wt %)						
Agε	7.78E-03	8.4E-04	na	na	7.78E-03	8.4E-04
Al	26.7	1.5	23.8	0.8	25.2	1.9
Bχ	6.68E-03	1.45E-03	<2.76E-02	-	6.68E-03	1.45E-03
Ba	4.46E-02	6.1E-03	4.56E-02	5.7E-03	4.51E-02	5.3E-03
Caε	0.105	0.009	na	na	0.105	0.009

Cd	≤ 3.07E-03	-	<4.14E-03	-	≤ 3.61E-03	-
Coα	4.54E-03	1.24E-03	na	na	4.54E-03	1.24E-03
Cr	0.586	0.108	0.524	0.071	0.555	0.089
Cuε	5.88E-03	6.2E-04	na	na	5.88E-03	6.2E-04
Fe	1.14	0.04	1.10	0.19	1.12	0.12
Laα	1.73E-02	3.7E-03	na	na	1.73E-02	3.7E-03
Liχ	<3.34E-03	-	<4.23E-02	-	<3.34E-03	-
Mg	2.42E-02	1.9E-03	≤ 7.03E-03	-	≤ 1.56E-02	-
Mn	0.118	0.023	9.53E-02	1.69E-02	0.107	0.022
Moχ	1.67E-02	3.2E-03	<5.22E-02	-	1.67E-02	3.2E-03
Naα	2.45	0.06	na	na	2.45	0.06
Ni	3.85E-02	7.9E-03	3.66E-02	6.5E-03	3.76E-02	6.6E-03
Pχ	<3.51E-02	-	<1.47E-02	-	<1.47E-02	-
Pb	<4.61E-02	-	<3.14E-02	-	<3.88E-02	-
Siδ	na	na	9.41	1.48	9.41	1.48
Snχ	≤ 2.33E-02	-	<4.43E-2	-	≤ 2.33E-02	-
Sr	4.35E-03	6.9E-04	4.00E-02	2.0E-03	2.22E-02	1.96E-02
Ti	3.68E-02	2.6E-03	4.35E-02	5.2E-03	4.02E-02	5.2E-03
Vα	7.30E-03	1.10E-03	na	na	7.30E-03	1.10E-03
Znε	2.00E-02	3.4E-03	na	na	2.00E-02	3.4E-03
Zrα	1.15E-02	1.4E-03	na	na	1.15E-02	1.4E-03
Gamma Scan (dpm/g)						
Cs-137	7.13E09	6.2E08	6.90E09	8.2E08	7.01E09	6.6E08
Tk 50 Rad Screen (dpm/g)						
Alpha	4.38E08	9.1E07	3.47E08	5.4E07	3.92E08	8.4E07
Beta	1.81E10	2.1E09	1.67E10	1.9E09	1.74E10	2.0E09
Sr-90 (dpm/g)						
Sr-90	5.77E09	5.9E08	4.28E09	6.7E08	5.02E09	1.00E09
Pu-238/241 (wt %)						
Pu-238						

	7.24E-04	1.35E-04	6.29E-04	1.08E-04	6.77E-04	1.21E-04
Pu-241	8.03E-05	1.01E-05	7.14E-05	1.66E-05	7.59E-05	1.33E-05
ICP-MS (wt %)						
M-232 (Th)	5.70E-04	1.00E-04	7.11E-04	1.04E-04	6.41E-04	1.20E-04
M-233 (U)	2.15E-04	5.0E-05	2.32E-04	3.2E-05	2.23E-04	3.9E-05
M-234 (U)	6.22E-04	1.36E-04	6.60E-04	8.3E-05	6.41E-04	1.03E-04
M-235 (U)	6.83E-03	1.61E-03	7.16E03	1.08E-03	7.00E-03	1.24E-03
M-236 (U)	3.19E-03	7.2E-04	3.38E-03	5.3E-04	3.29E-03	5.7E-04
M-237 (Np)	1.92E-04	3.6E-05	2.09E-04	2.9E-05	2.00E-04	3.1E-05
M-238 (**U**, Pu)	5.66E-02	1.24E-02	6.04E-02	8.8E-03	5.85E-02	9.9E-03
M-239 (Pu)	3.00E-03	4.5E-04	3.03E-03	4.8E-04	3.02E-03	4.1E-04
M-240 (Pu)	5.12E-04	9.2E-05	4.97E-04	9.8E-05	5.04E-04	8.6E-05
M-241 (Pu, Am)	2.71E-04	6.7E-05	2.69E-04	4.0E-05	2.70E-04	4.9E-05
M-242 (Pu)	4.24E-05	5.1E-06	4.56E-05	6.6E-06	4.40E-05	5.6E-06
M-243 (Am, Cm)	<8.03E-06	-	<7.90E-06	-	<7.97E-06	-
M-244 (Pu, Cm)	<8.03E-06	-	<7.90E-06	-	<7.97E-06	-
M-245 (Cm)	<8.03E-06	-	<7.90E-06	-	<7.97E-06	-
M-246 (Cm)	<8.03E-06	-	<7.90E-06	-	<7.97E-06	-
M-247 (Cm, Bk)	<8.03E-06	-	<7.90E-06	-	<7.97E-06	-
Calculated Values (wt %)						
Equivalent U-235	2.00E-02	3.6E-03	2.05E-02	3.1E-03	2.03E-02	3.0E-03
Total U	6.75E-02	1.50E-02	7.19E-02	1.05E-02	6.97E-02	1.18E-02
Total Pu	4.32E-03	6.9E-04	4.22E-03	6.9E-04	4.27E-03	6.2E-04
Ratios (Unitless)						
U-235/Total U	0.101	0.002	9.96E-02	7E-04	0.100	0.001
Na/Eq. U-235α	125	22	na	na	125	22
Fe/Eq. U-235	58.2	12.2	55.4	17.5	56.8	13.6
Cr/Eq. U-235	29.2	0.1	25.7	1.9	27.5	2.3
Zn/Eq. U-235ε	1.00	0.08	na	na	1.00	0.08
Mn/Eq. U-235	5.89	0.08	4.66	0.36	5.27	0.71

[α] No peroxide fusion result is provided by ADS for these species.

[χ] One of the two dissolution methods gave a result that was significantly lower than a minimum detection limit given by the other dissolution method. In these cases, the lower result is not considered to be in conflict with the higher minimum detection limit and is considered to be the better number.

[δ] Peroxide fusion is known to provide a much better dissolution of Si than aqua regia. The peroxide fusion number is therefore considered to be the better number, and therefore no aqua regia number is given.

[ε] Blank samples run indicate for some trace species that the peroxide fusion dissolutions show some sample contamination or analytical interference. For these samples the aqua regia dissolution result is considered the better number.

Table 3. Results of Analyses of Insoluble Solids (HTK-493 and HTK-494 � Corrected for Soluble Solids)

	Aqua Regia Dissolution		Peroxide Fusion Digestion		Combined	
Species	Avg.	St. Dev.	Avg.	St. Dev.	Avg.	St. Dev.
ICP-ES (wt %)						
Agε	7.78E-03	8.4E-04	na	na	7.78E-03	8.4E-04
Al	26.6	1.5	23.7	0.8	25.2	1.9
Bχ	6.67E-03	1.45E-03	<2.76E-02	-	6.67E-03	1.45E-03
Ba	4.46E-02	6.1E-03	4.56E-02	5.7E-03	4.51E-02	5.3E-03
Caε	0.105	0.009	na	na	0.105	0.009
Cd	≤ 3.07E-03	-	<4.14E-03	-	≤ 3.60E-03	-
Coα	4.54E-03	1.24E-03	na	na	4.54E-03	1.24E-03
Cr	0.585	0.108	0.524	0.072	0.554	0.088
Cuε	5.88E-03	6.2E-04	na	na	5.88E-03	6.2E-04
Fe	1.14	0.04	1.10	0.19	1.12	0.12
Laα	1.73E-02	3.7E-03	na	na	1.73E-02	3.7E-03
Liχ	<3.34E-03	-	<4.23E-03	-	<3.34E-03	-
Mg	2.42E-02	1.9E-03	≤ 7.01E-03	-	≤ 1.56E-02	-
Mn	0.118	0.023	9.53E-02	1.69E-02	0.107	0.022
Moχ	1.67E-02	3.2E-03	<5.22E-02	-	1.67E-02	3.2E-03
Naα	2.34	0.10	na	na	2.34	0.10
Ni	3.85E-02	7.9E-03	3.66E-02	6.5E-03	3.75E-02	6.6E-03
Pχ	<3.49E-02	-	<1.46E-02	-	<1.46E-02	-
Pb						

	<4.61E-02	-	<3.14E-02	-	<3.88E-02	-
Siδ	na	na	9.40	1.48	9.40	1.48
Snχ	≤ 2.33E-02	-	<4.43E-02	-	≤ 2.33E-02	-
Sr	4.35E-03	6.9E-04	4.00E-02	2.0E-03	2.22E-02	1.96E-02
Ti	3.68E-02	2.6E-03	4.35E-02	5.3E-03	4.01E-02	5.2E-03
Vα	7.29E-03	1.10E-03	na	na	7.29E-03	1.10E-03
Znε	2.00E-02	3.4E-03	na	na	2.00E-02	3.4E-03
Zrα	1.15E-02	1.41E-03	na	na	1.15E-02	1.41E-03
Gamma Scan (dpm/g)						
Cs-137	7.08E09	5.9E08	6.85E09	8.0E08	6.96E09	6.4E08
Tk 50 Rad Screen (dpm/g)						
Alpha	4.37E08	9.1E07	3.46E08	5.4E07	3.92E08	8.4E07
Beta	1.80E10	2.1E09	1.66E10	2.0E09	1.73E10	2.0E09
Sr-90 (dpm/g)						
Sr-90	5.77E09	5.9E08	4.28E09	6.7E08	5.02E09	1.00E09
Pu-238/241 (wt %)						
Pu-238	7.24E-04	1.35E-04	6.29E-04	1.08E-04	6.76E-04	1.21E-04
Pu-241	8.03E-05	1.01E-05	7.14E-05	1.67E-05	7.58E-05	1.33E-05
ICP-MS (wt %)						
M-232 (Th)	5.69E-04	1.00E-04	7.11E-04	1.05E-04	6.40E-04	1.20E-04
M-233 (U)	2.14E-04	5.0E-05	2.32E-04	3.2E-05	2.23E-04	3.9E-05
M-234 (U)	6.21E-04	1.36E-04	6.59E-04	8.3E-05	6.40E-04	1.03E-04
M-235 (U)	6.82E-03	1.61E-03	7.15E-03	1.08E-03	6.98E-03	1.24E-03
M-236 (U)	3.19E-03	7.2E-04	3.38E-03	5.3E-04	3.28E-03	5.8E-04
M-237 (Np)	1.92E-04	3.6E-05	2.09E-04	2.9E-05	2.00E-04	3.1E-05
M-238 (**U**, Pu)	5.65E-02	1.24E-02	6.04E-02	8.8E-03	5.84E-02	9.9E-03
M-239 (Pu)	3.00E-03	4.5E-04	3.02E-03	4.8E-04	3.01E-03	4.1E-04
M-240 (Pu)	5.12E-04	9.2E-05	4.96E-04	9.8E-05	5.04E-04	8.6E-05
M-241 (Pu, Am)	2.71E-04	6.7E-05	2.69E-04	4.0E-05	2.70E-04	4.9E-05
M-242 (Pu)	4.24E-05	5.1E-06	4.56E-05	6.6E-06	4.40E-05	5.6E-06

M-243 (Am, Cm)	<8.03E-06	-	<7.90E-06	-	<7.97E-06	-
M-244 (Pu, Cm)	<8.03E-06	-	<7.90E-06	-	<7.97E-06	-
M-245 (Cm)	<8.03E-06	-	<7.90E-06	-	<7.97E-06	-
M-246 (Cm)	<8.03E-06	-	<7.90E-06	-	<7.97E-06	-
M-247 (Cm, Bk)	<8.03E-06	-	<7.90E-06	-	<7.97E-06	-
Calculated Values (wt %)						
Equivalent U-235	2.00E-02	3.6E-03	2.05E-02	3.2E-03	2.02E-02	3.1E-03
Total U	6.74E-02	1.49E-02	7.18E-02	1.05E-02	6.96E-02	1.18E-02
Total Pu	4.32E-03	6.8E-04	4.22E-03	7.0E-04	4.27E-03	6.2E-04
Ratios (Unitless)						
U-235/Total U	0.101	0.002	9.96E-02	7E-04	0.100	0.001
Na/Eq. U-235α	119	18	na	na	119	18
Fe/Eq. U-235	58.2	12.2	55.5	17.2	56.8	13.6
Cr/Eq. U-235	29.2	0.1	25.7	1.9	27.5	2.3
Zn/Eq. U-235ε	1.00	0.08	na	na	1.00	0.08
Mn/Eq. U-235	5.89	0.08	4.66	0.36	5.27	0.71

$^{\alpha}$ No peroxide fusion result is provided by ADS for these species.

$^{\chi}$ One of the two dissolution methods gave a result that was significantly lower than a minimum detection limit given by the other dissolution method. In these cases, the lower result is not considered to be in conflict with the higher minimum detection limit and is considered to be the better number.

$^{\delta}$ Peroxide fusion is known to provide a much better dissolution of Si than aqua regia. The peroxide fusion number is therefore considered to be the better number, and therefore no aqua regia number is given.

$^{\varepsilon}$ Blank samples run indicate for some trace species that the peroxide fusion dissolutions show some sample contamination or analytical interference. For these samples the aqua regia dissolution result is considered the better number.

Table 4 gives the results for analyses of each of the three wash waters. As with the solids samples, the results are given as the average of three determinations. Since the three washes are expected to be different, no combined averages are given. No dissolutions were performed on the washes, only dilutions. The results of the wash water analyses are not always intuitive. The concentrations of many species decrease with later washes as would be expected as more material is dissolved away. However, the concentrations of some species increase with the later washes. This may be the result of pH effects. Some species are not as soluble in highly caustic solutions as they are in more neutral solutions, as is the case in going from the first to the third washes. Some of the aberrations are due to the increasing sensitivity of the instrumentation (ICP-ES and ICP-MS) in more dilute solutions. With the concentrations of aluminum and silicon measured, we expect a significant portion of the insoluble solids to be aluminum silicates. These aluminum silicates are chemically related to zeolites that have been used in the past in cesium removal columns in the Tank Farm. We believe it possible that this material might behave as an ionexchange material for some species, most notably Cs and Pu. As the chemistry of the wash water changes going from the 1st to the 3rd washes, we may be dissolving more of the aluminum silicates, thereby releasing more of the cesium.

Table 4. Results of Analyses of Wash Water Dissolved Solids (HTK-493 and HTK-494)

	First Wash		Second Wash		Third Wash	
Species	Avg.	St. Dev.	Avg.	St. Dev.	Avg.	St. Dev.
ICP-ES (mg/L)						
Ag	<5.11	-	≤ 0.647	-	<4.99E-02	-
Al	794	78	49.7	2.7	193	18
B	<3.06	-	≤ 0.340	-	8.98E-02	2.63E-02
Ba	<3.06	-	<0.306	-	0.309	0.183
Ca	≤ 4.97	-	≤ 0.904	-	0.795	0.351
Cd	2.55	0.23	≤ 0.237	-	≤ 2.99E-02	-
Co	≤ 5.02	-	≤ 0.409	-	<2.99E-02	-
Cr	≤ 11.6	-	1.63	0.13	3.92	2.63
Cu	<3.06	-	<0.306	-	<2.99E-02	-
Fe	≤ 5.62	-	0.646	0.061	7.79	4.26
La	<10.2	-	<1.02	-	≤ 0.113	-
Li	<3.06	-	<0.306	-	<2.99E-02	-
Mg	<1.02	-	<0.102	-	0.183	0.097
Mn	<1.02	-	<0.102	-	0.725	0.625
Mo	≤ 4.37	-	≤ 0.474	-	0.106	0.021
Na	7.28E04	4.1E03	5.93E03	1.75E03	1.01E03	200
Ni	<9.19	-	<0.951		≤ 0.316	-
P	184	10	13.4	1.5	1.39	0.25
Pb	<47	-	<4.69	-	<0.459	-
Si	88.7	6.8	6.17	4.26	156	15
Sn	43.3	9.6	4.34	1.24	0.376	0.183
Sr	<1.02	-	<0.102	-	3.33E-02	1.52E-02
Ti	1.40	0.49	≤ 0.205	-	0.459	0.320
V	≤ 3.30	-	≤ 0.477	-	≤ 3.99E-02	-

Zn	<3.06	-	<0.306	-	7.98E-02	4.56E-02
Zr	3.68	0.60	≤ 0.477	-	≤ 0.103	-
Gamma Scan (dpm/mL)						
Cs-137	2.53E07	8E05	5.59E06	1.94E06	4.74E07	3.19E07
Tk 50 Rad Screen (dpm/mL)						
Alpha	4.54E05	9.0E04	4.22E04	1.68E04	6.22E05	2.68E05
Beta	5.96E07	2.9E06	7.63E06	2.03E06	8.32E07	1.00E07
Pu-238/241 (µ g/L)						
Pu-238	1.58	0.04	6.26E-02	2.5E-03	2.55	1.71
Pu-241	0.187	0.014	<9.90E-03	-	0.300	0.197
ICP-MS (µ g/L)						
M-232 (Th)	<6.13	-	<6.12	-	5.42	3.51
M-233 (U)	≤ 6.46	-	<6.12	-	2.53	0.72
M-234 (U)	19.4	2.1	<6.12	-	8.58	2.34
M-235 (U)	184	10	43.4	8.0	95.8	25.6
M-236 (U)	87.8	7.2	22.7	4.2	45.5	11.5
M-237 (Np)	15.0	0.5	<6.12	-	≤ 1.16	-
M-238 (**U**, Pu)	1.50E03	1.1E02	377	59	705	186
M-239 (Pu)	7.84	0.71	<6.12	-	21.1	18.1
M-240 (Pu)	<6.13	-	<6.12	-	3.66	2.98
M-241 (Pu, Am)	<6.13	-	<6.12	-	1.90	1.41
M-242 (Pu)	<6.13	-	<6.12	-	<0.599	-
M-243 (Am, Cm)	<6.13	-	<6.12	-	<0.599	-
M-244 (Pu, Cm)	<6.13	-	<6.12	-	<0.599	-
M-245 (Cm)	<6.13	-	<6.12	-	<0.599	-
M-246 (Cm)	<6.13	-	<6.12	-	<0.599	-
M-247 (Cm, Bk)	<6.13	-	<6.12	-	<0.599	-
Calculated Values (µ g/L)						
Equivalent U-235	233	24	43.4	8.0	191	96.5
Total U	1.80E03	140	443	71	857	226

Total Pu	8.46	2.78	6.26E-02	2.5E-03	29.5	24.4
Ratios (Unitless)						
U-235/Total U	0.102	0.002	9.78E-02	2.7E-03	0.112	0.001
Na/Eq. U-235	3.13E05	1.6E04	1.37E05	2.9E04	6.80E03	4.58E03
Fe/Eq. U-235	≤ 24.8	-	15.2	3.1	40.6	3.6
Cr/Eq. U-235	≤ 51.5	-	38.9	10.4	19.4	3.6
Zn/Eq. U-235	<13.2	-	<7.23	-	0.460	0.254
Mn/Eq. U-235	<4.41	-	<2.41	-	3.31	1.44
Wet Chemistry Results (M)						
AlO_2^-	<0.204	-	<0.204	-	<0.020	-
CO_3^{-2}	<0.204	-	<0.204	-	<0.020	-
Free OH^-	<0.204	-	<0.204	-	<0.020	-
IC-Anions (M)						
Cl^-	<5.76E-03	-	<5.75E-03	-	<5.63E-04	-
F^-	<1.08E-02	-	<1.07E-02	-	<1.03E-03	-
CHO_2^-	<2.27E-02	-	<2.27E-02	-	<2.22E-03	-
NO_3^-	0.263	0.134	≤ 1.64E-02	-	1.72E-03	9E-05
NO_2^-	<2.22E-02	-	<2.22E-02	-	<2.17E-03	-
$C_2O_4^{-2}$	<1.16E-02	-	<1.16E-02	-	<1.13E-03	-
PO_4^{-3}	<1.08E-02	-	<1.07E-02	-	<1.05E-03	-
SO_4^{-2}	0.249	0.013	1.45E-02	4.8E-03	9.70E-04	4.81E-04

Quality Assurance

All analyses were conducted by ADS according to their routine operating and quality assurance procedures. Shielded Cells Operations personnel conducted all sample preparation work according to written instructions provided by Waste Processing Technology personnel. All results were recorded in laboratory notebook WSRC-NB-2002-00086, along with several notebooks kept by ADS personnel.

Acknowledgments

The author gratefully acknowledges the assistance of D. J. Wheeler, A. L. Williams, C. Coleman, C. Conley, W. Boyce, M. Malek, F. Pennebaker, W. Smith, D. Diprete, C. Diprete and M. Whitaker in the completion of this work.

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