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Chemical and Radiochemical Analysis of Consolidated Sludge Samples from the K East Basin

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Summary

This report describes results from analyses performed to characterize the consolidated sludge samples collected from the Hanford K East Basin in March and April 1999. The consolidated sludge samples were collected to provide additional material needed for the evaluation and design of systems that will be used to manage the K Basin sludge (i.e., disposition the sludge to T Plant for interim storage).

The analytical results given in this report add to the knowledge on the composition of the K Basin sludge and provide specific information on this sludge necessary to plan and understand subsequent process testing. The following analyses were performed: weight percent (wt%) solids determination; uranium analysis by kinetic phosphorescence; plutonium isotope analysis by extraction chromatographic separation followed by alpha energy analysis (AEA); gross beta analysis; gamma energy analysis (GEA); and metals analysis by inductively coupled plasma atomic emission spectroscopy (ICP-AES).

Contents

Tables

1.0 Introduction

Sludge samples from the Hanford K East Basin were collected by Duke Engineering & Services (DE&S) Hanford, Inc., from March through April 1999 to provide material for characterization and testing activities. The sludge was collected from several locations in the basin and then combined to form "consolidated samples," using the consolidated sludge sampling technique [Pitner 1999[a]]. This sampling method was designed to pull larger sample volumes from the basin. The samples were shipped to the storage pool at the Postirradiation Testing Laboratory (PTL, 327 Building) and transferred to the Pacific Northwest National Laboratory (PNNL) Radiochemical Processing Laboratory (RPL, 325 Building) between May 7 and 13, 1999, where they were recovered for testing and analysis.

This report gives the results of the analyses conducted to characterize the consolidated sludge samples. Section 2.0 describes the samples and preparation for analysis, and Section 3.0 tabulates the data.

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[a] Pitner, A. L. 1999. *K East Basin Sludge/Sampling 1999 Campaigns.* HNF-4746, Rev. 0, Numatec Hanford Corporation, Richland, Washington.

2.0 Sample Description and Preparation

The samples sent to PNNL are described in Table 1. The client sample identification number, designated by DE&S Hanford, is given, along with the corresponding PNNL Analytical Chemistry Laboratory (ACL) sample identification number. Samples KC-2 and KC-3 (canister sludge samples) were combined to form one sample (KC-2/3) to provide adequate sample material for process testing. The entire composite sample, KC-2/3, was wet-sieved using a 250-μm sieve. Approximately 25% of the sample material was retained on the sieve and was designated as P250 (i.e., plus, or greater than, 250 μm). Approximately 75% passed the 250-μm sieve and was designated as M250 (i.e., minus, or less than, 250 μm). This splitting of particle size into these fractions mimics the separation operations planned for retrieval of certain K Basin sludge types. Subsequent analysis of the separate fractions provides a better understanding of how uranium metal is distributed in the sludge.

Table 1. Sample Identification Matrix

Client Sample Identification	ACL Sample Identification	Sample Description
KC-1	00-00781	Canister sludge from highly damaged fuel collected from one sampling location – duplicate samples
KC-1 Dup	00-00782	
KC-2/3 P	00-00783	Canister sludge from moderate and highly damaged fuel at six locations ("P" = Plus 250-μm Split)
KC-2/3 P Dup	00-00784	
KC-2/3 M	00-00785	Canister sludge from moderate and highly damaged fuel at six locations ("M" = Minus 250-μm Split)
KC-2/3 M Dup	00-00786	
KC-4	00-00787	Floor sludge from between slotted canisters containing highly damaged fuel collected from three locations – duplicate samples
KC-4 Dup	00-00788	
KC-5	00-00789	Floor sludge from typical areas of deep sludge collected from three locations away from highly corroded fuel and areas with high concentrations of ion exchange material beads
KC-5 Dup	00-00790	

Weight percent solids determinations were performed on subsamples of each of the settled sludge samples. Samples KC-1 and KC-1 Dup were dried to a constant weight at 60°C. Samples KC-2/3 P, KC-2/3 M, KC-4, and KC-5 and their duplicates were dried at 105°C to constant weight before any subsequent analytical activities. After drying, each sample underwent a caustic fusion procedure (KOH-KNO_3), followed by chemical and radiochemical analyses. The analytical procedures are identified in Table 2. All laboratory data (except weight percent solids) are reported on a dry weight basis. The weight percent solids measurements (Table 3) were later used to calculate concentrations in settled sludge, as indicated in Tables 4 through 8 [see Section 3.0].

Table 2. Sample Preparation and Analysis Procedures

Procedure Number	Procedure Title
PNL-ALO-504 Rev. 2	Percent Solids Determination of Soils/Sludges/Solids
PNL-ALO-115 Rev. 1	Solubilization of Metals from Solids Using a KOH-KNO_3 Fusion
PNL-ALO-211 Rev. 0	Determination of Elements by Inductively Coupled Argon Plasma Atomic Emission Spectroscopy
PNL-ALO-4014 Rev. 0	Uranium Analysis by Kinetic Phosphorescence
PNL-ALO-431 Rev. 1	Total Beta Counting and Analysis
PNL-ALO-450 Rev. 1	Gamma Energy Analysis (GEA) and Low-Energy Photon Spectrometry (LEPS)
PNL-ALO-417 Rev. 2	Separation of U, Am/Cm, and Pu and Actinide Screen By Extraction Chromatography
PNL-ALO-420 Rev. 1	Solutions Analysis: Preparation of Alpha Sources By Direct Evaporation

3.0 Analysis Results

The results from the following analyses are given here: weight percent (wt%) solids, uranium concentration by kinetic phosphorescence; plutonium isotope concentration by extraction chromatography followed by alpha energy analysis (AEA); gross beta concentration; gamma energy analysis (GEA); and metals concentration by inductively coupled plasma atomic emission spectroscopy (ICP-AES).

The weight percent solids data are presented in Table 3. For these analyses, duplicate subsamples taken from each consolidated sludge sample were collected, weighed, placed in an oven, dried until constant weight, and recovered for follow-on sample preparation.

Selected consolidated dried sludge samples, prepared by KOH-KNO_3 fusion, were analyzed for uranium. The results are presented in Table 4.

The results of the AEA for plutonium isotopes; the total beta activity analysis; and the GEA for specific radionuclides for fusion-digested dried sludge and the original wet (settled) sludge are given in Tables 5 through 7, respectively.

Samples analyzed by ICP-AES are listed in Table 8. Specific analytes of interest (Ag, Al, Ba, Ca, Cd, Cr, Fe, Pb, Se, and Si) are highlighted. Other analytes are reported for information only. Many of the analytes reported were below the estimated quantitation limit (EQL), which, for Table 8, are within 10 times the instrument's detection limit. Those values are enclosed in brackets [ ]. Some of these less-than-EQL results may have occurred due to inadequate correction for interferences from very high U and Fe concentrations. The results reported below the EQL are for information only.

Table 3. Weight Percent Solids Results

Client Sample Identification	ACL Sample Identification	Wt% Solids	Average Wt% Solids	RPD
KC-1 [(a)]	00-00781	55.04	54.32	2.65
KC-1 Dup[(a)]	00-00782	53.60		
KC-2/3 P	00-00783	53.95	51.06	11.34
KC-2/3 P Dup	00-00784	48.16		
KC-2/3 M	00-00785	60.79	58.94	6.28
KC-2/3 M Dup	00-00786	57.09		
KC-4	00-00787	32.34	32.19	0.93
KC-4 Dup	00-00788	32.04		
KC-5	00-00789	35.49	34.98	2.92
KC-5 Dup	00-00790	34.47		

(a) Wt % solids data for Samples KC-1 and KC-1 Dup were determined following hot cell test instruction TI-30890-10, K East Basin Consolidated Sample KC-1 Small Scale Sieving.
RPD = Relative Percent Difference.

Table 4. Uranium Analysis Results

Client Sample Identification	ACL Sample Identification	Uranium Concentration, µg/g					
		Dry	Avg.	RPD	Wet	Avg.	RPD
KC-1	00-00781	6.89E+5	6.86E+5	0.87	3.79E+5	3.73E+5	3.49
KC-1 Dup	00-00782	6.83E+5			3.66E+5		
KC-2/3 P	00-00783	3.52E+5	---	---	1.90E+5	---	---
KC-2/3 M	00-00785	6.83E+5	---	---	4.15E+5	---	---
KC-4	00-00787	1.73E+5	1.66E+5	%SD =3.61%	5.60E+4	5.37E+4	%SD =3.61%
KC-4	00-00787 Rep	1.61E+5			5.21E+4		
KC-4 Dup	00-00788	1.65E+5			5.29E+4		
KC-5	00-00789	6.36E+4	---	---	2.26E+4	---	---

Dry = Dry Weight Basis; Wet = Wet (Settled Sludge) Basis.
RPD = Relative Percent Difference.
%SD = Percent Standard Deviation.

Table 5. Plutonium Isotope Analysis Results (µCi/g)

Client Sample Identification	ACL Sample Identification	$^{239+240}$Pu		^{238}Pu		^{236}Pu	
		Dry	Wet	Dry	Wet	Dry	Wet
KC-1	00-00781	1.44E+2	7.93E+1	2.25E+1	1.24E+1	<1.E-1	<6.E-2
KC-1	00-00781 Rep	1.38E+2	7.60E+1	2.09E+1	1.15E+1	<7.E-2	<4.E-2
KC-1 Dup	00-00782	1.43E+2	7.66E+1	2.12E+1	1.14E+1	<7.E-2	<4.E-2
KC-2/3 P	00-00783	9.03E+1	4.87E+1	1.30E+1	7.01E+0	<1.E-1	<5.E-2
KC-2/3 M	00-00785	1.23E+2	7.48E+1	1.74E+1	1.06E+1	<7.E-2	<4.E-2
KC-4	00-00787	3.93E+1	1.27E+1	4.53E+0	1.47E+0	<5.E-1	<2.E-1
KC-4 Dup	00-00788	3.91E+1	1.25E+1	5.28E+0	1.69E+0	<2.E-1	<6.E-2
KC-5	00-00789	1.31E+1	4.65E+0	1.99E+0	7.06E-1	<2.E-2	<7.E-3

Dry = Dry Weight Basis; Wet = Wet (Settled Sludge) Basis.

Table 6. Beta Analysis Results

Client Sample Identification	ACL Sample Identification	Total Beta, µCi/g	
		Dry	Wet
KC-1	00-00781	2.81E+3	1.55E+3
KC-1 Dup	00-00782	2.76E+3	1.48E+3
KC-2/3 P	00-00783	5.13E+3	2.77E+3
KC-2/3 M	00-00785	3.71E+3	2.26E+3
KC-4	00-00787	2.96E+3	9.57E+2
KC-4 Dup	00-00788	2.81E+3	9.00E+2
KC-4 Dup	00-00788 Rep	3.22E+3	1.04E+3
KC-5	00-00789	2.20E+1	7.81E+0

Dry = Dry Weight Basis; Wet = Wet (Settled Sludge) Basis.

Table 7. Gamma Energy Analysis Data (μCi/g)

Analyte	Client Sample Identification													
	KC-1		KC-1 Dup		KC-2/3 P		KC-2/3 M		KC-4		KC-4 Dup		KC-5	
	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet
Co-60	2.03E-1	1.12E-1	2.15E-1	1.15E-1	4.51E-1	2.43E-1	4.38E-1	2.66E-1	1.07E+0	3.46E-1	1.10E+0	3.52E-1	1.10E+0	3.90E-1
Cs-134	<9.E-2	<5.E-2	<8.E-2	<4.E-2	4.00E-1	2.16E-1	<8.E-2	<5.E-2	<2.E-1	<6.E-2	1.32E-1	4.23E-2	2.61E-2	9.26E-3
Cs-137	4.15E+2	2.28E+2	3.69E+2	1.98E+2	2.04E+3	1.10E+3	4.01E+2	2.44E+2	1.66E+3	5.37E+2	1.69E+3	5.41E+2	1.32E+2	4.68E+1
Eu-152	<9.E-1	<5.E-1	<8.E-1	<4.E-1	<5.E+0	<3.E+0	<9.E-1	<6.E-1	<3.E+0	<1.E+0	<3.E+0	<1.E+0	<4.E-1	<1.E-1
Eu-154	8.73E+0	4.80E+0	8.50E+0	4.56E+0	6.32E+0	3.41E+0	8.84E+0	5.37E+0	2.62E+0	8.47E-1	2.57E+0	8.23E-1	1.11E+0	3.94E-1
Eu-155	2.85E+0	1.57E+0	3.07E+0	1.65E+0	4.49E+0	2.42E+0	3.96E+0	2.41E+0	<2.E+0	<6.E-1	1.69E+0	5.41E-1	4.29E-1	1.52E-1
Am-241	1.23E+2	6.77E+1	1.21E+2	6.49E+1	7.55E+1	4.07E+1	9.63E+1	5.85E+1	2.94E+1	9.51E+0	2.91E+1	9.32E+0	1.31E+1	4.65E+0
Bi-212	<2.E+0	<1.E+0	<2.E+0	<1.E+0	<4.E+0	<2.E+0	<2.E+0	<1.E+0	<2.E+0	<6.E-1	<2.E+0	<6.E-1	<5.E-1	<2.E-1
Tl-208	<7.E-1	<4E-1	<6.E-1	<3.E-1	<3.E+0	<2.E+0	<6.E-1	<4.E-1	<2.E+0	<6.E-1	<2.E+0	<6.E-1	<3.E-1	<1.E-1
Sb-125	<2.E+0	<1.E+0	<1.E+0	<5.E-1	<6.E+0	<3.E+0	<2.E+0	<1.E+0	<4.E+0	<1.E+0	<4.E+0	<1.E+0	<5.E-1	<2.E-1
Ru/Rh-106	<2.E+0	<1.E+0	<2.E+0	<1.E+0	<9.E+0	<5.E+0	<2.E+0	<1.E+0	<6.E+0	<2.E+0	<6.E+0	<2.E+0	<7.E-1	<2.E-1
Ce/Pr-144	<2.E+0	<1.E+0	<2.E+0	<1.E+0	<8.E+0	<4.E+0	<2.E+0	<1.E+0	<5.E+0	<2.E+0	<5.E+0	<2.E+0	<5.E-1	<2.E-1
Nb-95	<7.E-2	<4.E-2	<7.E-2	<4.E-2	<2.E-1	<1.E-1	<7.E-2	<4.E-2	<1.E-1	<3.E-2	<1.E-1	<3.E-2	<3.E-2	<1.E-2
Ra-226	<7.E-1	<4.E-1	<6.E-1	<3.E-1	<4.E+0	<2.E+0	<6.E-1	<4.E-1	<3.E+0	<1.E+0	<3.E+0	<1.E+0	<3.E-1	<1.E-1

Dry = Dry Weight Basis; Wet = Wet (Settled Sludge) Basis.

Table 8. ICP-AES Analysis Data (μg/g)

Analyte	Client Sample Identification													
	KC-1		KC-1 Dup		KC-2/3 P		KC-2/3 M		KC-4		KC-4 Dup		KC-5	
	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet
Ag	[78]	[43]	[73]	[39]	[51]	[28]	[75]	[46]	[32]	[10]	[32]	[10]	--	--
Al	19,400	10,700	21,400	11,500	135,000	72,800	19,200	11,700	68,700	22,200	67,700	21,700	153,000	54,300
B	--	--	--	--	[110]	[60]	[54]	[33]	[120]	[39]	[76]	[24]	[93]	[33]
Ba	[60]	[33]	[75]	[40]	135	[70]	[83]	[50]	230	74	227	73	152	54
Be	--	--	--	--	[14]	[8]	[10]	[6]	[66]	[21]	[65]	[21]	[38]	[13]
Ca	[1200]	[660]	[1,300]	[700]	[2,300]	[1,240]	[960]	[580]	10,400	3,360	10,300	3,300	4,810	1,707
Cd	[16]	[9]	--	--	[24]	[13]	[15]	[9]	176	57	174	56	[120]	[43]
Ce	[820]	[450]	[800]	[430]	[450]	[240]	[790]	[480]	[280]	[90]	[280]	[90]	--	--
Co	[56]	[31]	[52]	[28]	--	--	[60]	[36]	--	--	--	--	--	--
Cr	[77]	[42]	[79]	[42]	[190]	[100]	[90]	[55]	707	230	698	224	680	240
Cu	303	167	289	155	[230]	[124]	325	198	669	216	696	223	807	286
Dy	[130]	[72]	[130]	[70]	[76]	[40]	[130]	[79]	--	--	--	--	--	--
Fe	3,470	1,910	3,300	1,770	29,100	15,700	14,300	8,690	241,000	77,900	244,000	78,200	161,000	57,100
La	[370]	[204]	[360]	[190]	[200]	[108]	[360]	[220]	[120]	[39]	[120]	[38]	[56]	[20]
Mg	[250]	[138]	[150]	[80]	[880]	[475]	[300]	[180]	3,110	1,006	3,490	1120	1,770	630
Mn	[160]	[88]	[140]	[75]	[330]	[180]	[230]	[140]	785	254	758	243	649	230
Mo	[250]	[138]	[250]	[134]	[160]	[86]	[230]	[140]	[85]	[27]	[84]	[27]	[78]	[28]
Na	2,440	1,340	2,300	1,230	3,010	1,620	2,160	1,310	3,470	1,120	3,720	1190	3,740	1,330
Nd	[930]	[512]	[900]	[480]	[510]	[275]	[910]	[550]	[300]	[97]	[290]	[93]	[140]	[50]
P	--	--	--	--	[250]	[135]	--	--	[740]	[240]	[700]	[224]	[610]	[220]
Pb	[300]	[165]	[260]	[140]	[300]	[160]	[290]	[175]	[410]	[130]	[410]	[131]	[370]	[130]
Se	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Si	[1,700]	[930]	[1,500]	[800]	19,400	10,470	[2,900]	[1,760]	50,100	16,200	48,100	15,400	54,600	19,400
Sn	[3,500]	[1930]	[3,300]	[1770]	[2,200]	[1,190]	[3,500]	[2,130]	[3,500]	[1,130]	[3,500]	[1,120]	[2,100]	[750]
Sr	[47]	[26]	[48]	[26]	[47]	[25]	[43]	[26]	[91]	[29]	[89]	[29]	[56]	[20]
Th	[4,400]	[2,420]	[4,400]	[2360]	[2,400]	[1,300]	[4,400]	[2700]	[1,300]	[420]	[1,200]	[380]	--	--
Ti	[98]	[54]	[93]	[50]	767	414	[150]	[90]	1,150	372	1,200	380	2,110	750
U	692,000	380,900	689,000	369,000	354,000	200,000	692,200	421,000	181,000	58,500	176,000	56,400	64,700	23,000
V	[120]	[66]	[110]	[60]	[100]	[54]	[120]	[73]	[84]	[27]	[85]	[27]	[65]	[23]
Y	[62]	[34]	[59]	[32]	--	--	[62]	[38]	--	--	--	--	--	--
Zn	--	--	--	--	[170]	[90]	[97]	[59]	1,170	378	1,180	378	740	260
Zr	[150]	[80]	[140]	[75]	[130]	[70]	[130]	[80]	[440]	[140]	[420]	[135]	[190]	[67]

Bracketed values listed in the above table are within 10 times the instrument detection limit, and have a potential uncertainty greater than 15%.
Measurements reported other than analytes of interest are for information only.
Dry = Dry Weight Basis; Wet = Wet (Settled Sludge) Basis.