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C-104 High-Level Waste Solids: Washing/Leaching and Solubility Versus Temperature Studies

G. J. Lumetta
D. J. Bates
J. P. Bramson
L. P. Darnell
O. T. Farmer III
S. K. Fiskum
L. R. Greenwood
F. V. Hoopes
C. Z. Soderquist
M. J. Steele
R. T. Steele
M.W. Urie
J. J. Wagner

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1.0 Introduction

This report describes the results of a test conducted by Battelle to assess the effects of inhibited water washing and caustic leaching on the composition of the C-104 HLW solids. The objective of this work was to determine the composition of the C-104 solids remaining after washing with 0.01 M NaOH or leaching with 3 M NaOH. Another objective of this test was to determine the solubility of the C-104 solids as a function of temperature. The work was conducted according to test plan BNFL-TP-29953-8, Rev. 0, *Determination of the Solubility of HLW Sludge Solids*.

2.0 Personnel

The key Battelle personnel and their responsibilities in performing this test are given below.

Staff Member	Responsibilities
G.J. Lumetta	Cognizant scientist. Prepared test plan and designed experiment. Supervised performance of the test. Prepared analytical service request. Interpreted data and reported results.
F.V. Hoopes	Hot cell technician. Performed test.
D.J. Bates	Statistical analysis of data.
M.W. Urie	Managed chemical and radiochemical analytical work.
B.M. Rapko	Technical reviewer.
K.P. Brooks	Task Leader.

3.0 Experimental

Sample Description. The sample used in this test was labeled as C104-GL. The C-104 HLW sample was composited as described in test plan BNFL-29953-031, *C-104 Sample Compositing*. Figure 3.1 summarizes the compositing and sub-sampling scheme. The C-104 sample was received from Hanford's 222-S Laboratory on March 3, 1999. This material was received in 14 glass jars. Figure 3.1 lists the sample numbers along with the mass of material recovered from each jar. The material in the jars was transferred to a stainless steel mixing vessel equipped with a motorized impeller. Before being used, all components of the mixing vessel were rinsed with methanol and then dried at 102°C for 12 h. Materials in the vessel were mixed for 1 h and 20 min before collecting sub-samples. The materials were actively mixed while sub-samples were collected through a 1.9-cm (.75-in.) ball valve located on the bottom of the vessel. The hot-cell temperature during the mixing process was 34°C.

The first three sub-samples (C-104 COMP A, B, and GL) were collected and allowed to settle. After approximately 10 days, the volume of settled solids in these three samples was measured to determine the effectiveness of the sub-sampling technique at collecting samples with representative solids/liquid ratios. The three sub-samples contained 88.9, 89.2, and 89.9 vol% settled solids indicating that the sampling technique provided representative sub-samples.

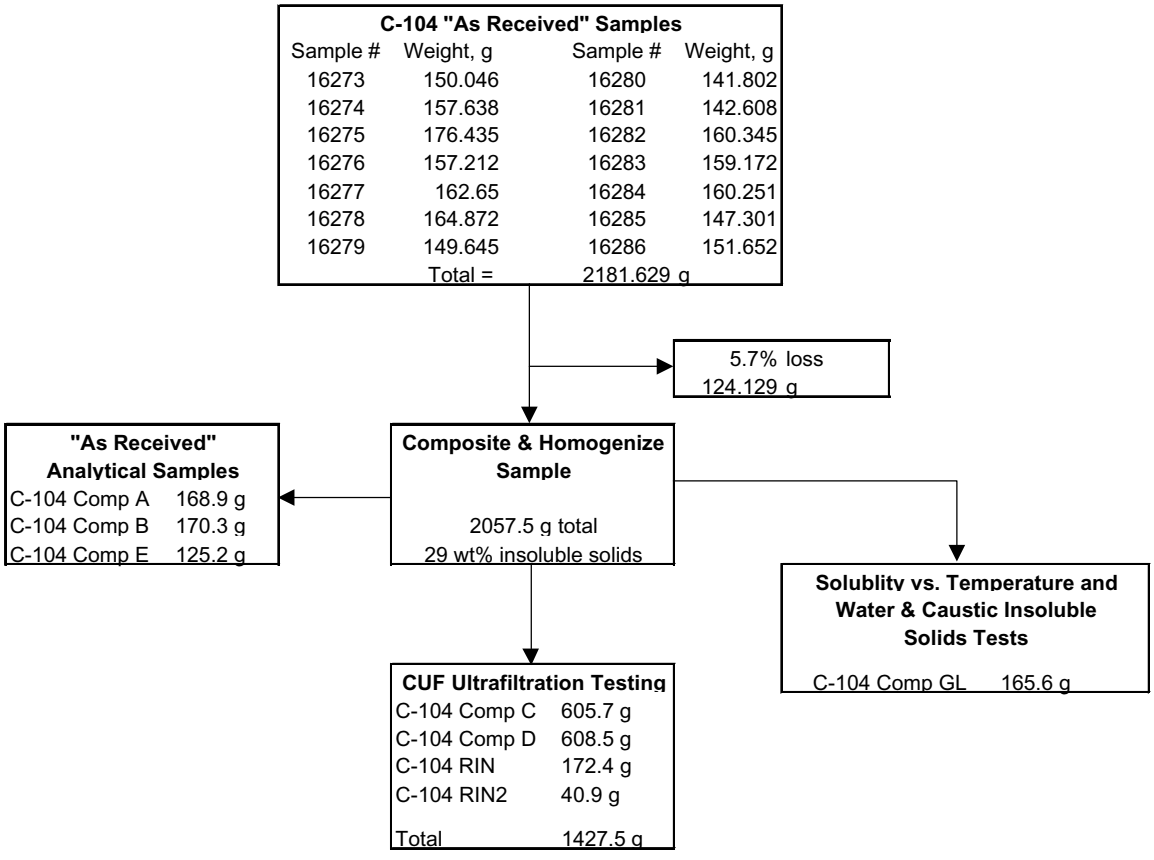

Figure 3.1. Compositing and Sub-Sampling Scheme For the Tank C-104 Sample

Apparatus. The apparatus used consisted of an aluminum heating block placed on a hot plate/stirrer, which was modified so that separate power could be applied to the heating and stirring functions. This allowed for continuous stirring, while the hot plate was powered by a temperature controller. The temperature controller used was a J-KEM Model 270 (J-KEM Electronics, Inc., St. Louis, MO). This temperature controller consists of two separate circuits. One is the temperature control circuit, while the other serves as an over-temperature device, which shuts down the system if a preset temperature is exceeded. The set point for the over-temperature circuit was set at 100°C for this test. A dual K-type thermocouple (model number CASS-116G-12-DUAL, Omega Engineering, Stamford, CT) was used to provide inputs to the temperature controller and over-temperature circuits. Both the J-KEM Model 270 and the dual thermocouple were calibrated before use. The aluminum heating block contained two wells. A vial containing water was placed in one of the wells, with the thermocouple wedged between this vial and the aluminum block. The vessel containing the sample was placed in the other well.

Procedure.[a] Because the stock C-104 HLW sample was very thick and not very fluid, 20 mL of 0.1 M NaOH was added to assist in homogenization. The sample was then placed on a shaker to homogenize immediately before use.

Solubility Versus Temperature. A 31.0459-g aliquot was transferred from C104 GL to a 60-mL high density polyethylene (HDPE) bottle (this bottle also contained a Teflon®-coated magnetic stir bar). Correcting for the 0.1 M NaOH added to fluidize the sample, this corresponded to 27.4 g of the as-received C-104 HLW sample. The sample bottle was sealed, then was heated and stirred at 30 ± 2 °C for 18 h. Two aliquots (4-mL each) were taken for analysis. Each aliquot was immediately filtered through a 0.45-µm nylon syringe filter that had been preheated by immersion in a boiling water bath. The filter was preheated to reduce the possibility of precipitation during the filtration step. The sample was very difficult to filter; less than 1 mL of clarified liquid was obtained from each aliquot. The temperature was increased to 40 ± 2 °C and the sample was stirred for 24 h. The mixture was sampled in the same manner as described above, except that only 2-mL aliquots were used (this actually yielded more liquid sample than when 4-mL aliquots were used, probably because there were less solids present to plug the filter). The temperature was increased to 50 ± 2 °C and the sample was stirred for 21 h. Again, the mixture was sample in the same manner as described above (2-mL aliquots). Because of the small volumes of each of the liquid samples take, only inductively-coupled plasma atomic emission spectroscopy (ICP/AES) analysis was performed (following acid digestion).

Determination of Aqueous-Insoluble Fraction. A 50.8765-g aliquot (44.8 g of as-received C-104 sample) was filtered through a 0.45-µm nylon filter membrane. As was observed in the solubility versus temperature test, the filtration process was relatively slow. The filtered solids were transferred to a 125-mL high density polyethylene (HDPE) bottle (this bottle also contained a Teflon®-coated magnetic stir bar) using a spatula.[b] The residual solids were transferred from the filter to the HDPE bottle using numerous portions of aqueous 0.01 M NaOH. The bottle was filled to capacity with

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[a] See Appendix A for a copy of the test plan and procedural notes.

[b] The wet solids were very sticky.

0.01 M NaOH. The bottle was equipped with a condenser tube, which allowed the system to vent during heating, but minimized evaporation. The mixture was heated and stirred at 85 ± 2 °C for 16.5 h. The test plan indicated that the washing slurry should be cooled prior to filtration, but as per instructions from BNFL, the slurry was filtered while hot. The hot washing slurry was filtered through a pre-weighed 0.45-μm nylon filtration unit. The weight of the filtrate was 100.13 g while the weight of the filtered solids was 41.64 g.

The filtered solids were transferred back into the HDPE bottle using a spatula. Again, the residual solids were transferred from the filter to the HDPE bottle using numerous portions of aqueous 0.01 M NaOH, then the bottle was filled to capacity with 0.01 M NaOH yielding ~123 g of slurry. The mixture was heated and stirred at 85 ± 2 °C for 22.5 h. The washing slurry was again filtered while hot yielding 82.79 g of washing solution and 40.49 g of wet solids. This process was repeated a third time. For the final washing step, the slurry was heated at 85 ± 2 °C for 24 h; 93.11 g of washing liquid was collected and the weight of the wet solids was 48.55 g. A composite sample of the three wash solutions was prepared for analysis.

After the final washing step, the filtered solids were transferred to a pre-weighed glass jar using deionized water. Excess water was evaporated at 80°C, then the solids were dried overnight at 105°C yielding 14.3589 g of dried washed solids.

Determination of Caustic-Insoluble Fraction. A 45.8422-g aliquot (40.4 g of as-received C-104 sample) was filtered through a 0.45-μm nylon filter membrane. The filtered solids were transferred to a 125-mL high density polyethylene (HDPE) bottle (this bottle also contained a Teflon®-coated magnetic stir bar) using a spatula. The residual solids were transferred from the filter to the HDPE bottle using numerous portions of aqueous 3 M NaOH. The bottle was filled to capacity with 3 M NaOH yielding ~140 g of slurry. The bottle was equipped with a condenser tube, which allowed the system to vent during heating, but minimized evaporation. The mixture was heated and stirred at 85 ± 2 °C for 21.5 h. As per instructions from BNFL, the leaching slurry was filtered while hot. The hot slurry was filtered through a pre-weighed 0.45-μm nylon filtration unit. The weight of the filtrate was 98.84 g and the wet solids weighed 41.47 g. A sample of the leaching solution was taken for analysis.

Most of the filtered solids were transferred back into the HDPE bottle using a spatula. Several ~10-mL aliquots of 0.01 M NaOH were used to transfer the remaining filtered solids back into the HDPE bottle. The slurry volume was made to ~100 mL with additional 0.01 M NaOH (total slurry weight ~123 g). The mixture was heated and stirred at 85 ± 2 °C for 21 h. The washing slurry was again filtered while hot yielding 92.45 g of washing solution and 33.35 g of wet solids. The washing process was repeated. For the final washing step, the slurry was heated at 85 ± 2 °C for 22.5 h, 88.31 g of washing liquid was collected, and the weight of the wet solid was 33.92 g. A composite sample of the two wash solutions was prepared for analysis.

After the final washing step, the filtered solids were transferred to a pre-weighed glass jar using deionized water. Excess water was evaporated at 80°C, then the solids were dried overnight at 105°C yielding 7.6051 g of dried leached solids.

4.0 Results

4.1 Solubility Versus Temperature

Tables 1, 2, and 3 present the concentrations of various C-104 waste components at 30, 40, and 50°C, respectively. Two sets of values are presented in each table. The first set of values is the analyte concentrations as determined directly on the aliquots analyzed. In the second set of values, the concentrations have been adjusted for loss in the sample weight that occurred between the time the aliquot was taken and the time the analyses were initiated. These adjustments were made assuming the weight losses were due to evaporation.

Tables 4 and 5 show the changes in the concentrations at 40 and 50°C relative to those at 30°C. Appendix D discusses a graphical analysis of the data, as well as regression results of fitting the component concentrations versus temperature. Based on this data set, only limited conclusions can be drawn. The following discussion will be limited to those analytes for which meaningful conclusions can be drawn.

The regression analysis of the adjusted data indicated statistically significant concentration changes only for Ag, Cd, Cr, Fe, and P (Appendix D). The Ag concentration was below detection limit at 30°C, but appeared to increase when the temperature was raised to 40 and 50°C. Similarly, the Cr concentration increased steadily with increasing temperature up to 50°C. Interestingly, the Cd, Fe, and P concentrations decreased with increasing temperature. The reason for this trend is not clear.

4.2 Dilute Hydroxide Washing

Table 6 presents the concentration of the C-104 components in a composite of the three wash solutions. The composite wash sample was prepared by mixing measured quantities of each wash solution; the relative weight of each wash solution corresponded to the fraction of the total wash solution represented by each. The composite wash solution was weighed immediately before analytical work was begun. The total weight of the sample had decreased 0.2% since the time the composite was first prepared. The concentrations determined were adjusted for this weight loss, assuming the weight loss was due to evaporation. The adjusted concentrations were then multiplied by the total combined weight of the three wash solutions (293.515 g) to yield the quantity of each component present in the wash solutions.

Table 7 presents the results of the analysis of the dilute hydroxide-washed C-104 solids. The solids were solubilized for ICP/AES analysis by KOH and Na_2O_2 fusion methods. Duplicate fusions and ICP/AES analyses were done for each type of fusion. Mean values from these determinations are presented in the table along with the standard deviation from the mean and the relative error. The relative error was obtained by the following formula: %RSD = 100(Std.Dev./Mean). For all the elements determined by ICP/AES the relative error was #10%, indicating good agreement between the duplicate measurements. Except where noted in the table, the mean values from all four measurements were used to determine the quantity of each component in the washed solids.

The Hg concentration was determined on the washed solids by cold vapor atomic absorption spectrophotometry following an oxidative acidic leaching of the solids. The mean Hg concentration was 96 μg/g and good agreement was achieved between duplicates.

TIC/TOC determination was performed using the hot persulfate method. This analysis was performed directly on the washed solids (not on fused material). Good reproducibility (5%) was achieved between duplicate TIC/TOC analyses. To date, no reliable method has been developed to quantify the anions present in Hanford tank solids. Anion (Cl^-, F^-, NO_3^-, SO_4^{2-}, PO_4^{3-}, and $C_2O_4^{2-}$) analysis was done by IC on a solution obtained by leaching the washed solids with deionized water. This in essence yielded the water-soluble anions not completely removed by the washing test. Only small amounts of soluble NO_3^- and perhaps PO_4^{3-} were found in the washed sludge. The low PO_4^{3-} concentration revealed by IC suggests that P found by ICP is indeed due to some water-insoluble P-containing phase(s). The chromatograms suggested interference in the F^- peak by organic anions. Hence, the fluoride values are viewed as unreliable.

Cyanide analysis on the washed solids revealed 13 μg CN^-/g. Reproducibility between duplicate CN^- analyses was good. Ammonia was determined by ion-selective electrode using water-slurries of the solids. Ammonia was not detected (< 9 μg/g) in the dried washed solids; however the value should be treated with caution since the solids were dried at 105°C prior to analysis.

Radiochemical analyses were performed on the solutions prepared by KOH fusion. Cesium-137, ^{241}Am, ^{154}Eu, and ^{155}Eu were determined by gamma spectroscopy. Americium-241 was also determined by alpha spectroscopy following Pu separation , as were ^{238}Pu, $^{239+240}$Pu, ^{242}Cm, and $^{243+244}$Cm. Technetium-99, ^{129}I, ^{235}U, ^{238}U, ^{237}Np, ^{239}Pu, and ^{240}Pu were determined by ICP-MS. Strontium-90 was determined by proportional beta-counting following separation of this isotope.

Agreement between duplicate measurements was good. The values obtained for ^{241}Am by gamma and alpha spectroscopies agreed within 10%. Agreement between the ICP-MS results and the alpha spectroscopic results was also good. The combined activities for ^{239}Pu and ^{240}Pu as determined by ICP-MS were 7.01 μCi/g and the $^{239+240}$Pu value obtained by alpha spectroscopy was 7.07 μCi/g. There was some inconsistency regarding the U analysis. The ICP-MS analysis revealed 54,800 μg/g (^{235}U + ^{238}U), but only 25,550 μg total U was indicated by laser fluorimetry analysis. To be conservative, the higher U value should probably be used. This use of the higher value is supported by the ICP-AES data, which indicated 44500 μg U/g.

Table 8 presents the composition of the dilute hydroxide-washed C-104 solids and the percent of each component removed by dilute hydroxide washing. In addition, the composition of the "untreated" C-104 sample used in this test is presented. These values were obtained by summing the amount of the given component found in the wash solutions (Table 6) and the washed solids (Table 7), then dividing this total by the weight of the C-104 sample used. The washed solids were dominated by Al (15.2 wt%), Zr (5.1 wt%), Fe (4.6 wt%), Na (1.8 wt%), Si (1.3 wt%), and Mn (1.1 wt%). The concentrations of the major radionuclides contained in the washed solids were 17 μCi TRU/g (as indicated by the

total alpha concentration), 7 μCi ^{241}Am/g, 5 μCi ^{239}Pu/g, 784 μCi ^{90}Sr/g, and 44 μCi ^{137}Cs/g, indicating the solids should be treated as HLW.

The wash solutions were stable over a period of ~5.5 months. No precipitates were observed in the solutions after this period of time.

4.3 Caustic Leaching

Table 9 presents the concentration of the C-104 components in the caustic leach solution and in a composite of the two wash solutions. The composite wash sample was prepared by mixing measured quantities of each wash solution; the relative weight of each wash solution corresponded to the fraction of the total wash solution represented by each. The samples were weighed immediately before analytical work was begun. The weight of the leach solution sample had decreased 0.06% and that of the composite wash solution sample had decreased 0.23% since the time the samples were first prepared. The concentrations determined were adjusted for this weight loss, assuming the weight loss was due to evaporation. The adjusted concentrations were then multiplied by the weight of the leach solution (98.8366 g) or the combined weight of the two wash solutions (180.7635 g) to yield the quantity of each component present in the leach and the wash solutions, respectively.

Table 10 presents the results of the analysis of the caustic leached C-104 solids. Analysis of these solids was conducted in the same way as for the dilute hydroxide-washed solids. Generally, excellent agreement between duplicate measurements was obtained for the analytes determined by ICP/AES. The single exception being Mg. Again, the mean values from all four measurements were used to determine the amount of each component in the leached solids, except where noted in the table.

As with the dilute hydroxide-washed solids, the IC results indicated only small amounts of soluble NO_3^- and perhaps PO_4^{3-} were in the leached sludge. The low PO_4^{3-} concentration revealed by IC suggests that P found by ICP is indeed due to some water-insoluble P-containing phase(s). The chromatograms suggested interference in the F^- peak by organic anions. Hence, the fluoride values are viewed as unreliable. TIC/TOC analyses of the leached solids yielded very good reproducibility between duplicates.

Cyanide analysis on the leached solids revealed 23 μg CN^-/g, with good reproducibility between duplicates. Ammonia was determined by ion-selective electrode using water-slurries of the solids. Ammonia was not detected (< 9 μg/g) in the dried leached solids; however the value should be treated with caution since the solids were dried at 105°C prior to analysis.

The relative uncertainties for the radionuclides, except for ^{242}Cm and $^{243+244}$Cm, were less than 10% indicating good reproducibility between duplicates. The values obtained for ^{241}Am by gamma and alpha spectroscopies agreed within 2% indicating good agreement between the two methods. However, the ICP-MS and the alpha spectroscopic results were inconsistent. The combined activities for ^{239}Pu and ^{240}Pu as determined by ICP-MS were 13.0 μCi/g and the $^{239+240}$Pu value obtained by alpha spectroscopy was 26.1 μCi/g. To be conservative, the higher value should probably be used. In contrast, the U value obtained by ICP-MS [96,560 μg/g (^{235}U + ^{238}U)] agreed well with the value of 100,100 μg total U indicated by laser fluorimetry analysis and 90,600 μg U/g determined by ICP-AES (Na_2O_2 fusion prep).

Table 11 presents the composition of the caustic-leached C-104 solids and the percent of each component removed by caustic leaching. In addition, the composition of the "untreated" C-104 sample used in this test is presented. These values were obtained by summing the amount of the given component found in the leaching and washing solutions (Table 9) and the leached solids (Table 10), then dividing this total by the weight of the C-104 sample used. The leached solids were dominated by Th (11.6 wt%), Zr (10.2 wt%), U (10.0 wt%), Fe (8.1 wt%), Na (3.5 wt%), Al (3.4 wt%), Si (2.2 wt%) and Mn (1.9 wt%). The concentrations of the major radionuclides contained in the washed solids were 58 μCi TRU/g (as indicated by the total alpha concentration), 26 μCi ^{241}Am/g, 26 μCi $^{239+240}$Pu/g, 2820 μCi ^{90}Sr/g, and 136 μCi ^{137}Cs/g, indicating the solids should be treated as HLW.

It should be noted that the composition for the original C-104 solid listed in Table 8 should agree with that listed in Table 11. The composition generally agrees, however the Al value obtained from the washing test is much less than that obtained in the leaching test. This was perhaps due to sample inhomogeneity, but a more likely reason is incomplete Al dissolution in the fusion preparations for the washed solids. Significant solids remained when the fused material from the washed solids was taken up in solution for analysis. These solids were suspended by stirring and an aliquot of the resulting suspension was diluted with 2% HCl, yielding a clear solution. However, it is possible that the solids were not suspended in a homogeneous manner. Thorium and U are other key components that do not agree very well.

The caustic leach solution was not stable. Although the solution remained clear after one day, a gel-like material had formed on the bottom of the container after ~20 days. Considerable solids were present after 5.5 months. The wash solutions were stable for ~ 1.5 months, but white solids had formed in the second wash solution after 5.5 months. Interestingly, the first wash solution was clear after 5.5 months. It is not clear why solids formed in the second wash solution, but not the first. It could be due to the lower hydroxide concentration in the second wash solution.

Table 1. C-104 Component Concentrations in Solution at 30°C[a]

	Concentration at 30°C, Unadjusted					Concentration at 30°C, Adjusted[b]				
Analyte	C104-SOL-30-1	C104-SOL-30-2	Mean[c]	Std. Dev.	% RSD	C104-SOL-30-1	C104-SOL-30-2	Mean	Std. Dev.	% RSD
Ag	< 1	< 0.3	< 1	--	--	< 0.3	< 0.2	< 0.3	--	--
Al	(14)	17.9	(16)	(3)	17	(6.3)	9.1	(8)	2	26
Ba	5.02	(1.1)	(3.1)	(2.8)	91	2.27	(0.6)	1.42	1.21	85
Ca	(12)	74.4	(43)	(44)	102	(5)	38.0	21.7	23.1	106
Cd	6.36	5.01	5.69	0.95	17	2.88	2.56	2.72	0.22	8
Co	(2.0)	(1.6)	(1.8)	(0.3)	16	(0.90)	(0.82)	(0.86)	0.06	7
Cr	63.4	50.0	56.7	9.5	17	28.7	25.6	27.1	2.2	8
Cu	6.89	5.72	6.31	0.83	13	3.12	2.92	3.02	0.14	4
Fe	(4.2)	(3.1)	(3.7)	(0.8)	21	(1.9)	(1.6)	(1.7)	0.2	13
K	(560)	452	(506)	(76)	15	(253)	231	242	16	6
La	< 1	< 0.5	< 1	--	--	< 0.5	< 0.3	< 0.5	--	--
Mg	< 4	29.2	29.2	--	--	< 2	14.9	14.9	--	--
Mn	< 0.2	< 0.1	< 0.2	--	--	< 0.1	< 0.1	< 0.1	--	--
Mo	(7.8)	6.63	(7.2)	0.8	11	(3.5)	3.39	(3.5)	0.1	3
Na	72900	59700	66300	9334	14	32967	30514	31741	1734	5
Ni	126	99.6	113	19	17	57.0	50.9	53.9	4.3	8
P	1400	1120	1260	198	16	633	572	603	43	7
Pb	< 2	< 1	< 2	--	--	< 1	< 1	< 1	--	--
Si	405	663	534	182	34	183	339	261	110	42
Ti	< 0.2	(0.22)	(0.22)	--	--	< 0.1	(0.11)	(0.11)	--	--
U	< 83	< 40	< 61	--	--	< 37	< 20	< 37	--	--
Zn	12.1	8.0	10.1	2.9	29	5.5	4.1	4.8	1.0	20
Zr	< 1.0	< 0.5	< 1	--	--	< 0.5	< 0.3	< 1	--	--

(a) Due to lack of sample, only ICP-AES analyses were performed on these samples. Values in parentheses are within 10 times the analytical detection limit, and thus have uncertainties >15%.

(b) Values corrected for mass loss (evaporation) that occurred during interim storage of the samples.

(c) For analytes that were only detected in one duplicate sample, the detected value is given as the mean.

Table 2. C-104 Component Concentrations in Solution at 40°C[a]

	Concentration at 40°C, Unadjusted					Concentration at 40°C, Adjusted[b]				
Analyte	C104-SOL-40-1	C104-SOL-40-2	Mean[c]	Std. Dev.	% RSD	C104-SOL-40-1	C104-SOL-40-2	Mean	Std. Dev.	% RSD
Ag	(0.69)	(0.59)	(0.64)	0.07	11	(0.46)	(0.40)	(0.43)	0.04	10
Al	(3.4)	(4.4)	(3.9)	0.7	18	(2.3)	(3.0)	(2.6)	0.5	19
Ba	(0.34)	< 0.1	< 0.4	--	--	(0.23)	< 0.1	< 0.4	--	--
Ca	43.5	50.6	47.1	5.0	11	29.0	34.1	31.5	3.6	12
Cd	3.16	3.09	3.13	0.05	2	2.10	2.08	2.09	0.02	1
Co	(1.3)	(1.3)	(1.3)	0.0	0	(0.9)	(0.9)	(0.9)	0.0	1
Cr	51.4	48.9	50.2	1.8	4	34.2	33.0	33.6	0.9	3
Cu	4.57	4.46	4.52	0.08	2	3.04	3.01	3.02	0.03	1
Fe	(2.5)	(1.9)	(2.2)	0.4	19	(1.7)	(1.3)	(1.5)	0.3	18
K	382	362	372	14	4	254	244	249	7	3
La	< 0.4	< 0.2	< 0.4	--	--	< 0.2	< 0.2	< 0.2	--	--
Mg	16.4	19.6	18.0	2.3	13	10.9	13.2	12.1	1.6	13
Mn	< 0.1	(0.06)	< 0.1	--	--	< 0.05	(0.04)	< 0.05	--	--
Mo	5.40	5.26	5.33	0.10	2	3.60	3.55	3.57	0.04	1
Na	55500	54000	54750	1061	2	36959	36401	36680	394	1
Ni	79.3	75.4	77.4	2.8	4	52.8	50.8	51.8	1.4	3
P	851	829	840	16	2	567	559	563	6	1
Pb	< 1	< 1	< 1	--	--	< 1	< 0.4	< 1	--	--
Si	507	562	535	39	7	338	379	358	29	8
Ti	< 0.1	< 0.05	< 0.1	--	--	< 0.05	< 0.03	< 0.05	--	--
U	< 29	< 19	< 29	--	--	< 19	< 13	< 19	--	--
Zn	3.45	3.31	3.38	0.10	3	2.30	2.23	2.26	0.05	2
Zr	< 0.4	< 0.2	< 0.4	--	--	< 0.2	< 0.2	< 0.2	--	--

(a) Due to lack of sample, only ICP-AES analyses were performed on these samples. Values in parentheses are within 10 times the analytical detection limit, and thus have uncertainties >15%.

(b) Values corrected for mass loss (evaporation) that occurred during interim storage of the samples.

(c) For analytes that were only detected in one duplicate sample, the detected value is given as the mean. Barium is an exception, where the detected value in one aliquot was greater than the detection limit for the other analyte, For Ba, a conservative value of < 0.4 was used.

Table 3. C-104 Component Concentrations in Solution at 50°C[a]

	Concentration at 50°C, Unadjusted					Concentration at 50°C, Adjusted[b]				
Analyte	C104-SOL-50-1	C104-SOL-50-2	Mean	Std. Dev.	% RSD	C104-SOL-50-1	C104-SOL-50-2	Mean	Std. Dev.	% RSD
Ag	(1.2)	(1.1)	(1.2)	0.1	6	(0.8)	(0.8)	(0.8)	0.0	2
Al	11.1	(4.6)	(7.9)	4.6	59	7.4	(3.4)	(5.4)	2.8	52
Ba	(0.34)	< 0.2	< 0.4	--	--	(0.23)	< 0.1	< 0.3	--	--
Ca	45.8	71.4	58.6	18.1	31	30.7	53.4	55.0	16.1	29
Cd	2.70	2.20	2.45	0.35	14	1.81	1.64	1.73	0.12	7
Co	(1.3)	(1.1)	(1.2)	0.1	12	(0.9)	(0.8)	(0.8)	0.0	4
Cr	63.1	49.9	56.5	9.3	17	42.2	37.3	39.8	3.5	9
Cu	4.41	3.48	3.95	0.66	17	2.95	2.60	2.78	0.25	9
Fe	(1.7)	(1.9)	(1.8)	0.1	8	(1.1)	(1.4)	(1.3)	0.2	16
K	371	300	336	50	15	248	224	236	17	7
La	< 0.4	< 0.4	< 0.4	--	--	< 0.3	< 0.3	< 0.3	--	--
Mg	21.9	28.6	25.3	4.7	19	14.7	21.4	18.0	4.8	26
Mn	(0.10)	< 0.1	< 0.15	--	--	(0.07)	< 0.1	< 0.10	--	--
Mo	5.22	(4.1)	(4.7)	0.8	17	3.49	(3.1)	(3.3)	0.3	9
Na	52100	44900	48500	5091	10	34874	33564	34219	926	3
Ni	75.3	59.6	67.5	11.1	16	50.4	44.6	47.5	4.1	9
P	784	624	704	113	16	525	466	496	41	8
Pb	< 1	< 1	< 1	--	--	< 1	< 1	< 1	--	--
Si	506	678	592	122	21	339	507	423	119	28
Ti	(0.26)	(0.25)	(0.26)	0.01	--	(0.17)	(0.19)	(0.18)	0.01	--
U	< 32	< 14	< 32	--	--	< 22	< 11	< 16	--	--
Zn	6.77	(2.2)	(4.5)	3.2	72	4.53	(1.6)	(3.1)	2.0	66
Zr	< 0.4	< 0.4	< 0.4	--	--	< 0.3	< 0.3	< 0.3	--	--

(a) Due to lack of sample, only ICP-AES analyses were performed on these samples. Values in parentheses are within 10 times the analytical detection limit, and thus have uncertainties >15%.

(b) Values corrected for mass loss (evaporation) that occurred during interim storage of the samples.

Table 4. Unadjusted Concentration Changes Relative to 30°C

Analyte	Pooled %RSD[a]	40°C % Change[b]	40°C Std. Dev.[c]	40°C 90% C.I.[d]	50°C % Change[b]	50°C Std. Dev.[c]	50°C 90% C.I.[d]
Ag	(e)	(e)	--	--	(e)	--	--
Al	37	**-76**	**13**	**± 30**	-51	26	± 60
Ba	--	(e)	--	--	(e)	--	--
Ca	62	9	95	± 224	36	119	± 279
Cd	13	**-45**	**10**	**± 23**	**-57**	**8**	**± 18**
Co	11	**-28**	**12**	**± 27**	**-33**	**11**	**± 25**
Cr	14	-12	17	± 40	0	19	± 45
Cu	12	-28	12	± 29	**-37**	**11**	**± 26**
Fe	17	**-40**	**15**	**± 35**	**-51**	**12**	**± 28**
K	12	-26	13	± 30	**-34**	**12**	**± 28**
La	(e)	(e)	--	--	(e)	--	--
Mg	--	-38	--	--	-14	--	--
Mn	(e)	(e)	--	--	(e)	--	--
Mo	12	-26	12	± 29	**-35**	**11**	**± 26**
Na	10	-17	12	± 28	**-27**	**11**	**± 25**
Ni	14	**-31**	**13**	**± 31**	**-40**	**12**	**± 27**
P	13	**-33**	**12**	**± 29**	**-44**	**10**	**± 24**
Pb	(e)	(e)	--	--	(d)	--	--
Si	23	0	33	± 78	11	37	± 86
Ti	(e)	(e)	--	--	(e)	--	--
U	(e)	(e)	--	--	(e)	--	--
Zn	45	**-66**	**21**	**± 50**	-55	28	± 67
Zr	(e)	(e)	--	--	(e)	--	--

(a) Pooled %RSD is the pooled percent relative standard deviation, obtained as the root mean square of the %RSD values at 30°C, 40°C, and 50°C.

(b) The percent change is given by: $\%Change = 100*(C_T - C_{30})/C_{30}$, where C_T is the average concentration at temperature T (40 or 50°C) and C_{30} is the average concentration at 30°C.

(c) Std.Dev. of % Change is the standard deviation of the % Change values at 40°C and 50°C, both relative to 30°C. It is computed as C_T/C_{30}*Sqrt(2)*%RSD.

(d) 90% two-sided confidence intervals were constructed assuming a statistical t-distribution with 3 degrees of freedom. % Change values larger than their 90% C.I. are considered significant evidence of a change due to temperature. Such values are shown in boldface.

(e) Analyte not detected in solution for at least one temperature.

Table 5. Adjusted Concentration Changes Relative to 30°C

Analyte	Pooled %RSD[a]	40°C % Change[b]	40°C Std. Dev.[c]	40°C 90% C.I.[d]	50°C % Change[b]	50°C Std. Dev.[c]	50°C 90% C.I.[d]
Ag	(e)	(e)	--	--	(e)	--	--
Al	35	**-66**	**17**	**± 40**	-30	35	± 82
Ba	--	(e)	--	--	(e)	--	--
Ca	64	45	131	± 309	153	229	± 538
Cd	6	**-23**	**7**	**± 16**	**-37**	**5**	**± 13**
Co	5	1.1	7	± 16	-1.7	7	± 15
Cr	7	24	12	± 29	**47**	**15**	**± 35**
Cu	6	0.2	8	± 19	-8.0	8	± 18
Fe	16	-15	19	± 44	-27	16	± 39
K	6	2.9	9	± 20	-2.4	8	± 19
La	(e)	(e)	--	--	(e)	--	--
Mg	--	-19	--	--	21	--	--
Mn	(e)	(e)	--	--	(e)	--	--
Mo	6	3.3	8	± 19	-5.2	8	± 18
Na	4	**16**	**6**	**± 14**	7.8	5	± 13
Ni	7	-3.9	9	± 22	-12	9	± 20
P	6	-6.6	8	± 20	**-18**	**7**	**± 17**
Pb	(e)	(e)	--	--	(e)	--	--
Si	30	37	58	± 135	62	68	± 160
Ti	(e)	(e)	--	--	(e)	--	--
U	(e)	(e)	--	--	(e)	--	--
Zn	40	-53	27	± 63	-35	36	± 86
Zr	(e)	(e)	--	--	(e)	--	--

(a) Pooled %RSD is the pooled percent relative standard deviation, obtained as the root mean square of the %RSD values at 30°C, 40°C, and 50°C.

(b) The percent change is given by: $\%Change = 100*(C_T - C_{30})/C_{30}$, where C_T is the average concentration at temperature T (40 or 50°C) and C_{30} is the average concentration at 30°C.

(c) Std.Dev. of % Change is the standard deviation of the % Change values at 40°C and 50°C, both relative to 30°C. It is computed as $C_T/C_{30}*Sqrt(2)*\%RSD$.

(d) 90% two-sided confidence intervals were constructed assuming a statistical t-distribution with 3 degrees of freedom. % Change values larger than their 90% C.I. are considered significant evidence of a change due to temperature. Such values are shown in boldface.

(e) Analyte not detected in solution for at least one temperature.

Table 6. Dilute Hydroxide Washing of C-104 Sludge: Analysis of the Composite Wash Solution[a]

Analyte	Direct	Adjusted[b]	Amount (μCi or μg) in Wash Solutions
Ag	0.654	0.653	192
Al	126	126	36901
Ba	(0.053)	(0.053)	(16)
Ca	11.7	11.7	3426
Cd	(0.52)	(0.52)	(152)
Co	(0.19)	(0.19)	(56)
Cr	12.5	12.5	3661
Cu	0.638	0.637	187
Fe	(0.33)	(0.33)	(97)
Hg	Not Measured	--	--
K	(51)	(51)	(14936)
La	< 0.1	< 0.1	< 26
Mg	4.6	4.5	1335
Mn	< 0.02	< 0.02	< 5
Mo	(0.72)	(0.72)	(211)
Na	10800	10776	3162901
Ni	9.79	9.77	2867
P	107	107	31336
Pb	< 0.2	< 0.2	< 62
Si	112	112	32800
Th	< 3	< 3	< 879
Ti	(0.035)	(0.035)	(10)
U	14.7	14.7	4305
Zn	(0.56)	(0.56)	(164)
Zr	< 0.1	< 0.1	< 26
TOC	775	773	226967
TIC	680	678	199146
Cl^-	150	150	43929
F^- [c]	5000	4989	1464306
NO_3^-	1450	1447	424649
SO_4^{2-}	400	399	117144
PO_4^{3-}	< 250	< 249	< 73215
CN^-	Not Measured	--	--
NH_3	Not Measured	--	--

Table 6. Dilute Hydroxide Washing of C-104 Sludge: Analysis of the Composite Wash Solution (con't)

Analyte	Direct	Adjusted[(b)]	Amount (µCi or µg) in Wash Solutions
^{137}Cs	3.84E+00	3.83E+00	1.12E+03
^{90}Sr	2.91E-03	2.90E-03	8.52E-01
^{99}Tc	1.39E-03	1.38E-03	4.06E-01
^{241}Am(()	< 5E-03	< 5E-03	< 1E+00
^{241}Am(")	Not Measured	--	--
^{154}Eu	< 3E-04	< 3E-04	< 9E-02
^{155}Eu	< 5E-03	< 5E-03	< 1E+00
^{14}C[(e)]	Not Measured	--	--
^{129}I	Not Measured	--	--
^{235}U	Not Measured	--	--
^{238}U	Not Measured	--	--
^{237}Np	Not Measured	--	--
^{238}Pu	Not Measured	--	--
^{239}Pu	Not Measured	--	--
^{240}Pu	Not Measured	--	--
$^{239+240}$Pu	Not Measured	--	--
$^{243+244}$Cm	Not Measured	--	--
^{242}Cm	Not Measured	--	--
Total Alpha	1.16E-04	1.16E-04	3.40E-02

(a) Concentrations for radionuclides are in units of µCi/g; all other components are in units of µg/g. Values in parentheses are within 10 times the analytical detection limit, and thus have uncertainties >15%.

(b) Value adjusted for the 0.2% loss in sample weight that occurred before analysis; this weight loss was assumed to be due to evaporation.

(c) Quantified by IC system as fluoride, but slight retention time peak shift and peak shape suggest significant organic anion interference. It is highly probable that there is little or no fluoride actually present in the sample.

Table 7. Analysis of the C-104 Washed Solids[a]

	KOH Fusion					Na_2O_2 Fusion					Amount (µCi or µg)
Analyte	C104-AQ-8	C104-AQ-8DUP	Mean	Std Dev.	% RSD	C104-AQ-8	C104-AQ-8DUP	Mean	Std Dev.	% RSD	in C104-AQ-8
Ag[b]	990	984	987	4	0	(630)	(720)	(675)	(64)	9	14172
Al	145000	147000	146000	1414	1	163000	152000	157500	7778	5	2178963
Ba	198	192	195	4	2	183	184	184	1	0	2717
Ca[c]	4740	4790	4765	35	1	4830	4931	4881	71	1	69249
Cd	922	872	897	35	4	898	885	892	9	1	12840
Co	< 126	< 125	< 130	--	--	< 48	< 52	< 52	--	--	< 1810
Cr	1550	1500	1525	35	2	1460	1470	1465	7	0	21467
Cu[d]	< 807	< 801	< 810	--	--	(230)	(200)	(215)	(21)	10	(3087)
Fe	47900	46700	47300	849	2	44900	45700	45300	566	1	664817
Hg	95.4	96.8	96.1	1	1	--	--	--	--	--	1380
K	--	--	--	--	--	< 1935	< 2068	< 2070	--	--	< 29694
La[b]	(130)	(130)	(130)	0	0	< 73	(80)	< 85	--	--	(1867)
Mg[d]	< 3529	< 3505	< 3530	--	--	(960)	(970)	(965)	(7)	1	(13856)
Mn	11100	10800	10950	212	2	10500	10500	10500	0	0	153999
Mo	< 81	< 80	< 81	--	--	< 48	< 52	< 52	--	--	< 1158
Na[e]	17964	17484	17724	339	2	--	--	--	--	--	254497
Ni	--	--	--	--	--	2900	2910	2905	7	0	41713
P[d]	2230	2570	2400	240	10	4740	4990	4865	177	4	69856
Pb	1840	1730	1785	78	4	1730	1770	1750	28	2	25379
Si	12900	13200	13050	212	2	12000	12000	12000	0	0	179845
Th[b]	34600	41100	37850	4596	12	9750	18700	14225	6329	44	543484
Ti[b]	2740	2760	2750	14	1	(160)	(180)	(170)	(14)	8	39487
U	26100	25000	25550	778	3	--	--	--	--	--	366870
Zn	(240)	(210)	(225)	21	9	(240)	(230)	(235)	(7)	3	3303
Zr	51300	50500	50900	566	1	--	--	--	--	--	730868
TOC	9900	10600	10250	495	5	--	--	--	--	--	147179
TIC	2560	2380	2470	127	5	--	--	--	--	--	35466
Cl^-	120	< 130	120	--	--	--	--	--	--	--	1723
F^- [h]	2900	2600	2750	212	8	--	--	--	--	--	39487
NO_3^-	1500	1300	1400	141	10	--	--	--	--	--	20102
SO_4^{2-}	570	< 250	< 600	--	--	--	--	--	--	--	< 8615
PO_4^{3-}	640	620	630	--	--	--	--	--	--	--	9046
CN^-	13.4	12.1	12.8	1	7	--	--	--	--	--	183
								--	--	--	
NH_3	< 8.4	< 9.0	< 9.0	--	--	--	--	--	--	--	< 129

Table 7. Analysis of the C-104 Washed Solids (con't)

	KOH Fusion					Na_2O_2 Fusion					Amount (µCi or µg)
Analyte	C104-AQ-8	C104-AQ-8DUP	Mean	Std Dev.	% RSD	C104-AQ-8	C104-AQ-8DUP	Mean	Std Dev.	Rel% Error	in C104-AQ-8
^{137}Cs	4.53E+01	4.27E+01	4.40E+01	1.84E+00	4	--	--	--	--	--	6.32E+02
^{90}Sr	7.78E+02	7.90E+02	7.84E+02	8.49E+00	1	--	--	--	--	--	1.13E+04
^{99}Tc	3.68E-02	3.70E-02	3.69E-02	1.41E-04	0	--	--	--	--	--	5.30E-01
^{241}Am(()	7.07E+00	6.89E+00	6.98E+00	1.27E-01	2	--	--	--	--	--	1.00E+02
^{241}Am(")	7.80E+00	7.21E+00	7.51E+00	4.17E-01	6	--	--	--	--	--	1.08E+02
^{154}Eu	2.24E+00	2.17E+00	2.21E+00	4.95E-02	2	--	--	--	--	--	3.17E+01
^{155}Eu	1.38E+00	1.28E+00	1.33E+00	7.07E-02	5	--	--	--	--	--	1.91E+01
^{14}C	< 7E-03	< 4E-03	< 7E-03	--	--	--	--	--	--	--	< 1E-01
^{129}I	6.64E-04	5.80E-04	6.22E-04	5.94E-05	10	--	--	--	--	--	8.93E-03
^{235}U	8.88E-04	8.38E-04	8.63E-04	3.54E-05	4	--	--	--	--	--	1.24E-02
^{238}U	1.88E-02	1.82E-02	1.85E-02	4.24E-04	2	--	--	--	--	--	2.66E-01
^{237}Np	7.00E-03	6.12E-03	6.56E-03	6.22E-04	9	--	--	--	--	--	9.42E-02
^{238}Pu	7.82E-01	7.98E-01	7.90E-01	1.13E-02	1	--	--	--	--	--	1.13E+01
^{239}Pu	5.24E+00	5.12E+00	5.18E+00	8.49E-02	2	--	--	--	--	--	7.44E+01
^{240}Pu	1.80E+00	1.86E+00	1.83E+00	4.24E-02	2	--	--	--	--	--	2.63E+01
$^{239+240}$Pu	7.11E+00	7.02E+00	7.07E+00	6.36E-02	1	--	--	--	--	--	1.01E+02
$^{243+244}$Cm	1.09E-01	9.63E-02	1.03E-01	8.98E-03	9	--	--	--	--	--	1.47E+00
^{242}Cm	1.24E-02	1.49E-02	1.37E-02	1.77E-03	13	--	--	--	--	--	1.96E-01
Total Alpha	1.67E+01	1.67E+01	1.67E+01	0.00E+00	0	--	--	--	--	--	2.40E+02

(a) Concentrations for radionuclides are in units of µCi/g dry solids; all other components are in units of µg/g dry solids. Values in parentheses are within 10 times the analytical detection limit, and thus have a potential experimental uncertainty >15%. TIC/TOC and cyanide analyses was performed directly on the washed solids. Anion (IC) analysis was done on a water leachate of the washed solids, so this does not accurately represent the anions present in the solids.

(b) Only the mean value from the KOH fusions were used to determine the amount of Ag, La, Th, and Ti in the washed solids.

(c) The Ca values from the Na_2O_2 fusion were corrected for the high process blank.

(d) Only the mean value from the Na_2O_2 fusions were used to determine the amount of Cu, Mg, and P in the washed solids.

(e) The Na values from the KOH fusion were corrected for the high process blank.

(h) Quantified by IC system as fluoride, but slight retention time peak shift and peak shape suggest significant organic anion interference. It is highly probable that there is little or no fluoride actually present in the sample.

Table 8. Concentrations in the Washed and Untreated C-104 Solids and the Relative Amount of Each Component Removed by Dilute Hydroxide Washing

	Washed Solids[a]		Original Sample[c]			
Analyte	µg or µCi/g dry solids	Pseudo 95% C.I. (if %RSDs=10)[b]	µg or µCi/g sample	Pseudo 95% C.I. (if %RSDs=10)[b]	Removed,%[d]	Pseudo 95% C.I. (if %RSDs=10)[c]
Ag	987	± 140r	321	± 45r	1	± 0.3r
Al	151750	± 21476r	49461	± 6885r	2	± 0.4r
Ba	189	± 27r	61	± 9r	1	± 0.14r
Ca	4823	± 682r	1622	± 219r	5	± 1.1r
Cd	894	± 126r	290	± 41r	1	± 0.3r
Co	< 126	--	< 42	--	3	--
Cr	1495	± 211r	561	± 70r	15	± 3r
Cu	215	± 30r	73	± 10r	6	± 1.4r
Fe	46300	± 6549r	14842	± 2099r	0	± 0.004r
Hg	96	± 14r	31	± 4r	--	--
K	< 2068	--	< 996	--	> 33	--
La	130	± 18r	42	± 6r	1	± 0.3r
Mg	965	± 136r	339	± 44r	9	± 2.1r
Mn	10725	± 1517r	3437	± 486r	0	± 0.0008r
Mo	< 81	--	< 31	--	15	--
Na[e]	17724	± 2507r	76281	± 14143r	93	± 25r
Ni	2905	± 411r	995	± 132r	6	± 1.5r
P	4865	± 688r	2259	± 261r	31	± 7r
Pb	1768	± 250r	567	± 80r	0	± 0.06r
Si	12525	± 1773r	4747	± 587r	15	± 4r
Th	37850	± 5353r	12131	± 1716r	0	± 0.04r
Ti	2750	± 389r	882	± 125r	0	± 0.006r
U	25550	± 3613r	8285	± 1158r	1	± 0.3r
Zn	230	± 33r	77	± 10r	5	± 1.1r
Zr	50900	± 7198r	16314	± 2307r	0	± 0.0009r
TOC	10250	± 1450r	8351	± 1115r	61	± 14.6r
TIC	2470	± 349r	5237	± 896r	85	± 22.3r
Cl^-	120	± 17r	1019	± 196r	96	± 26.7r
F^-	2750	± 389r	33567	± 6538r	97	± 27.2r
NO_3^-	1400	± 198r	9927	± 1897r	95	± 26.4r
SO_4^{2-}	< 600	--	2810 > x > 2615	--	93	--
PO_4^{3-}	630	± 89r	202	± 29r	< 89	--
CN^-	13	± 2r	4	± 1r	--	--
NH_3	< 9	--	< 3	--	--	--

Table 8. Concentrations in the Washed and Untreated C-104 Solids and the Relative Amount of Each Component Removed by Dilute Hydroxide Washing (con't)

	Washed Solids[a]		Original Sample[c]			
Analyte	µg or µCi/g dry solids	Pseudo 95% C.I. (if %RSDs=10)[b]	µg or µCi/g sample	Pseudo 95% C.I. (if %RSDs=10)[b]	Removed,%[d]	Pseudo 95% C.I. (if %RSDs=10)[c]
^{137}Cs	4.40E+01	± 6.22E+00r	3.92E+01	± 5.40E+00r	64	± 16r
^{90}Sr	7.84E+02	± 1.11E+02r	2.51E+02	± 3.55E+01r	0.0076	± 0.0019r
^{99}Tc	3.69E-02	± 5.22E-03r	2.09E-02	± 2.47E-03r	43	± 10r
^{241}Am(()	6.98E+00	± 9.87E-01r	2.27E+00	± 3.16E-01r	< 1.4	± 0.4r
^{241}Am(")	7.51E+00	± 1.06E+00r	--	--	--	--
^{154}Eu	2.21E+00	± 3.12E-01r	7.09E-01	± 9.99E-02r	< 0.28	± 0.07r
^{155}Eu	1.33E+00	± 1.88E-01r	4.59E-01	± 6.06E-02r	< 7.1	± 1.7r
^{14}C	< 7E-03	--	< 2E-03	--	--	--
^{129}I	6.22E-04	± 8.80E-05r	1.99E-04	± 2.82E-05r	--	--
^{235}U	8.63E-04	± 1.22E-04r	2.77E-04	± 3.91E-05r	--	--
^{238}U	1.85E-02	± 2.62E-03r	5.93E-03	± 8.39E-04r	--	--
^{237}Np	6.56E-03	± 9.28E-04r	2.10E-03	± 2.97E-04r	--	--
^{238}Pu	7.90E-01	± 1.12E-01r	2.53E-01	± 3.58E-02r	--	--
^{239}Pu	5.18E+00	± 7.33E-01r	1.66E+00	± 2.35E-01r	--	--
^{240}Pu	1.83E+00	± 2.59E-01r	5.87E-01	± 8.29E-02r	--	--
$^{239+240}$Pu	7.07E+00	± 9.99E-01r	2.26E+00	± 3.20E-01r	--	--
$^{243+244}$Cm	1.03E-01	± 1.45E-02r	3.29E-02	± 4.65E-03r	--	--
^{242}Cm	1.37E-02	± 1.93E-03r	4.37E-03	± 6.19E-04r	--	--
Total Alpha	1.67E+01	± 2.36E+00r	5.35E+00	± 7.57E-01r	0.014	± 0.003r

(a) The concentration in the washed solids was determined by summing the quantity found in the washed solids (Table 7) and dividing by the total weight (14.3589 g) of the washed solids (dry basis at 105°C).

(b) Pseudo 95% Confidence Intervals (C.I.) were approximated using propagation of error techniques for the case where the %RSD of all analytical measures used is 10% and all measures are independent The reader can review other potential %RSD values by multiplying the cell value by r, where r is %RSD/10.

(c) The concentration in the as-received sample was determined by summing the quantity found in the wash solution (Table 6) and the washed solids (Table 7) and dividing by the total weight (44.8 g) of sample used. Exceptions to this are cyanide, ammonia, mercury, C-14, I-129, U-235, U-238, Np-237, Pu-238, Pu-239, Pu-240, Pu-239+240, Cm-242, and Cm243+244. For these analytes only that found in the washed solids was included in the calculation (the wash solutions were not analyzed for these).

(d) The percent removed was determined by the following formula: $\%Removed = 100*F_w/(F_w+F_s)$; where F_w is the fraction in the wash solution and F_s is the fraction in the washed solids. The exception is Hg, where only that found in the solids was considered.

(e) The values for Na are not corrected for Na added as NaOH during the washing process.

Table 9. Caustic Leaching of C-104 Sludge: Analysis of the Leaching Solution and the Composite Wash Solution[a]

	Leach Solution C104-OH-3			Composite Wash Solution C104-OH-9		
Analyte	Direct	Adjusted[b]	Amount (µCi or µg)	Direct	Adjusted[c]	Amount (µCi or µg)
Ag	< 1.2	< 1.2	< 116	(0.47)	(0.46)	(84)
Al	45900	45873	4533915	2065	2060	372430
Ba	< 0.8	< 0.8	< 77	(0.05)	(0.05)	(9.2)
Ca	< 20	< 20	< 1931	(4.2)	(4.2)	(757)
Cd	(5.8)	(5.8)	(573)	(0.09)	(0.09)	(16)
Co	< 2	< 2	< 193	< 0.1	< 0.1	< 17
Cr	157	157	15508	19.0	18.9	3418
Cu	(2.4)	(2.4)	(237)	(0.13)	(0.12)	(23)
Fe	(2.6)	(2.6)	(257)	(0.35)	(0.35)	(63)
Hg	Not Measured	--	--	Not Measured	--	--
K	(260)	(260)	(25682)	(13)	(12)	(2254)
La	< 2	< 2	< 193	< 0.1	< 0.1	< 17
Mg	< 8	< 8	< 772	(1.9)	(1.8)	(334)
Mn	< 0.4	< 0.4	< 39	< 0.02	< 0.02	< 3.4
Mo	(4.1)	(4.1)	(405)	(0.28)	(0.27)	(50)
Na	153000	152909	15113049	13550	13519	2443793
Ni	(7.9)	(7.9)	(780)	(0.79)	(0.79)	(142)
P	814	814	80405	32.8	32.7	5916
Pb	(25)	(25)	(2469)	(1.2)	(1.1)	(207)
Si	(72)	(72)	(7112)	65.3	65.2	11777
Th	< 63	< 63	< 6223	< 3.1	< 3.1	< 559
Ti	< 0.4	< 0.4	< 39	(0.027)	(0.026)	(4.8)
U	10.9	10.9	1073	3.42	3.41	617
Zn	(0.6)	(0.6)	(55)	(0.50)	(0.50)	(90)
Zr	< 2	< 2	< 193	< 0.1	< 0.1	< 17
TOC	1300	1299	128412	215	215	38776
TIC	1200	1199	118534	275	274	49597
Cl^-	360	360	35560	< 130	< 130	< 23446
F^- [d]	5500	5497	543280	6600	6585	1190335
NO_3^-	1700	1699	167923	400	399	72141
SO_4^{2-}	500	500	49389	< 250	< 249	< 45088
PO_4^{3-}	1000	999	98778	< 250	< 249	< 45088
CN^-	Not Measured	--	--	Not Measured	--	--
NH_3	Not Measured	--	--	Not Measured	--	--

Table 9. Caustic Leaching of C-104 Sludge: Analysis of the Leaching Solution and the Composite Wash Solution (con't)

	Leach Solution C104-OH-3			Composite Wash Solution C104-OH-9		
Analyte	Direct	Adjusted[(b)]	Amount (µCi or µg)	Direct	Adjusted[(c)]	Amount (µCi or µg)
^{137}Cs	5.69E+00	5.69E+00	5.62E+02	1.49E+00	1.49E+00	2.69E+02
^{90}Sr	7.03E-03	7.03E-03	6.94E-01	1.14E-03	1.14E-03	2.06E-01
^{99}Tc	2.20E-03	2.20E-03	2.17E-01	7.78E-04	7.76E-04	1.40E-01
^{241}Am(()	< 7E-03	< 7E-03	< 7E-01	< 2E-03	< 2E-03	< 4E-01
^{241}Am(")	Not Measured	--	--	Not Measured	--	--
^{154}Eu	< 4E-04	< 4E-04	< 4E-02	< 2E-04	< 2E-04	< 4E-02
^{155}Eu	< 7E-03	< 7E-03	< 7E-01	< 2E-03	< 2E-03	< 4E-01
^{14}C	Not Measured	--	--	Not Measured	--	--
^{129}I	Not Measured	--	--	Not Measured	--	--
^{235}U	Not Measured	--	--	Not Measured	--	--
^{238}U	Not Measured	--	--	Not Measured	--	--
^{237}Np	Not Measured	--	--	Not Measured	--	--
^{238}Pu	Not Measured	--	--	Not Measured	--	--
^{239}Pu	Not Measured	--	--	Not Measured	--	--
^{240}Pu	Not Measured	--	--	Not Measured	--	--
$^{239+240}$Pu	Not Measured	--	--	Not Measured	--	--
$^{243+244}$Cm	Not Measured	--	--	Not Measured	--	--
^{242}Cm	Not Measured	--	--	Not Measured	--	--
Total Alpha	1.77E-04	1.77E-04	1.75E-02	1.03E-04	1.03E-04	1.86E-02

(a) Concentrations for radionuclides are in units of µCi/g; all other components are in units of µg/g. Values in parentheses are within 10 times the analytical detection limit.

(b) Value adjusted for the 0.06% loss in sample weight that occurred before analysis; this weight loss was assumed to be due to evaporation.

(c) Value adjusted for the 0.23% loss in sample weight that occurred before analysis; this weight loss was assumed to be due to evaporation.

(d) Quantified by IC system as fluoride, but slight retention time peak shift and peak shape suggest significant organic anion interference. It is highly probable that there is little or no fluoride actually present in the sample.

Table 10. Analysis of the C-104 Leached Solids[a]

	KOH Fusion					Na_2O_2 Fusion					Amount (µCi or µg)
Analyte	C104-OH-8	C104-OH-8DUP	Mean	Std Dev.	% RSD	C104-OH-8	C104-OH-8DUP	Mean	Std Dev.	% RSD	in C104-OH-8
Ag[b]	1810	1770	1790	28	2	(790)	(770)	(780)	(14)	2	13613
Al	32600	32600	32600	0	0	36100	35700	35900	283	1	260475
Ba	319	322	321	2	1	359	357	358	1	0	2580
Ca[c]	7260	7090	7175	120	2	8962	9212	9087	177	2	61837
Cd	1590	1560	1575	21	1	1750	1740	1745	7	0	12624
Co[d]	< 62	< 62	< 62	--	--	(60)	(66)	(63)	(4)	--	(479)
Cr	1810	1790	1800	14	1	1980	2000	1990	14	1	14412
Cu[d]	< 396	< 395	< 396	--	--	445	458	452	9	2	3434
Fe	77500	77800	77650	212	0	85300	84800	85050	354	0	618675
Hg	153	164	159	8	5	--	--	--	--	--	1205
K[e]	--	--	--	--	--	< 2056	(2700)	< 3000	--	--	(20534)
La[d]	(210)	(210)	(210)	0	0	(260)	(270)	(265)	(7)	3	(2015)
Mg[d]	< 1731	< 1728	< 1731	--	--	2630	1730	2180	636	29	16579
Mn	17900	17900	17900	0	0	19700	19600	19650	71	0	142786
Mo	< 40	< 40	< 40	--	--	< 51	< 50	< 51	--	--	< 388
Na[f]	34600	35100	34850	354	1	--	--	--	--	--	265038
Ni	--	--	--	--	--	5540	5560	5550	14	0	42208
P[d]	2050	1840	1945	148	8	4650	4730	4690	57	1	35668
Pb	2900	2830	2865	49	2	3190	3250	3220	42	1	23139
Si	22000	22000	22000	0	0	22800	22800	22800	0	0	170354
Th[d]	61800	61600	61700	141	0	114000	119000	116500	3536	3	885994
Ti	359	362	361	2	1	(430)	(430)	(430)	(0)	0	3006
U	101000	99200	100100	1273	1	--	--	--	--	--	761271
Zn	322	316	319	4	1	(340)	(340)	(340)	(0)	0	(2506)
Zr	101000	104000	102500	2121	2	--	--	--	--	--	779523
TOC	17000	16900	16950	71	0	--	--	--	--	--	128906
TIC	6900	6840	6870	42	1	--	--	--	--	--	52475
Cl^-	150	170	160	14	9	--	--	--	--	--	1217
F^- [g]	2900	2800	2850	71	2	--	--	--	--	--	21675
NO_3^-	1200	1300	1250	71	6	--	--	--	--	--	9506
SO_4^{2-}	< 240	< 230	< 240	--	--	--	--	--	--	--	< 1825
PO_4^{3-}	< 240	< 230	< 240	--	--	--	--	--	--	--	< 1825
CN^-	22.0	24.7	23.4	2	8	--	--	--	--	--	178
								--	--	--	
NH_3	< 8.5	< 8.5	< 8.5	--	--	--	--	--	--	--	< 65

Table 10. Analysis of the C-104 Leached Solids (con't)

	KOH Fusion					Na_2O_2 Fusion					Amount (µCi or µg)
Analyte	C104-OH-8	C104-OH-8DUP	Mean	Std Dev.	% RSD	C104-OH-8	C104-OH-8DUP	Mean	Std Dev.	Rel% Error	in C104-OH-8
^{37}Cs	1.36E+02	1.35E+02	1.36E+02	7.07E-01	1	--	--	--	--	--	1.03E+03
^{0}Sr	2.74E+03	2.90E+03	2.82E+03	1.13E+02	4	--	--	--	--	--	2.14E+04
^{9}Tc	5.59E-02	6.30E-02	5.95E-02	5.02E-03	8	--	--	--	--	--	4.52E-01
^{41}Am(()	2.57E+01	2.69E+01	2.63E+01	8.49E-01	3	--	--	--	--	--	2.00E+02
^{41}Am(")	2.48E+01	2.71E+01	2.60E+01	1.63E+00	6	--	--	--	--	--	1.97E+02
^{54}Eu	6.20E+00	7.07E+00	6.64E+00	6.15E-01	9	--	--	--	--	--	5.05E+01
^{55}Eu	3.96E+00	4.46E+00	4.21E+00	3.54E-01	8	--	--	--	--	--	3.20E+01
^{4}C	< 5E-03	< 3E-03	< 5E-03	--	--	--	--	--	--	--	< 3E-02
^{29}I	< 3E-04	< 3E-04	< 3E-04	--	--	--	--	--	--	--	< 2E-03
^{35}U	1.51E-03	1.49E-03	1.50E-03	1.41E-05	1	--	--	--	--	--	1.14E-02
^{38}U	3.28E-02	3.23E-02	3.26E-02	3.54E-04	1	--	--	--	--	--	2.48E-01
^{37}Np	3.28E-02	3.23E-02	3.26E-02	3.54E-04	1	--	--	--	--	--	2.48E-01
^{38}Pu	2.70E+00	2.93E+00	2.82E+00	1.63E-01	6	--	--	--	--	--	2.14E+01
^{39}Pu	9.49E+00	9.74E+00	9.62E+00	1.77E-01	2	--	--	--	--	--	7.31E+01
^{40}Pu	3.45E+00	3.22E+00	3.34E+00	1.63E-01	5	--	--	--	--	--	2.54E+01
$^{39+240}$Pu	2.62E+01	2.59E+01	2.61E+01	2.12E-01	1	--	--	--	--	--	1.98E+02
$^{43+244}$Cm	2.97E-01	4.25E-01	3.61E-01	9.05E-02	25	--	--	--	--	--	2.75E+00
^{42}Cm	8.11E-02	6.54E-02	7.33E-02	1.11E-02	15	--	--	--	--	--	5.57E-01
Total Alpha	5.70E+01	5.98E+01	5.84E+01	1.98E+00	3	--	--	--	--	--	4.44E+02

(a) Concentrations for radionuclides are in units of µCi/g dry solids; all other components are in units of µg/g dry solids. Values in parentheses are within 10 times the analytical detection limit, and thus have a potential experimental uncertainty >15%. TIC/TOC and cyanide analyses was performed directly on the washed solids. Anion (IC) analysis was done on a water leachate of the washed solids, so this does not accurately represent the anions present in the solids.

(b) Only the mean value from the KOH fusions were used to determine the amount of Ag in the washed solids.

(c) The Ca values from the Na_2O_2 fusion were corrected for the high process blank.

(d) Only the mean value from the Na_2O_2 fusions were used to determine the amount of Co, Cu, La, Mg, P, and Th in the washed solids.

(e) Only the single value from the duplicate Na_2O_2 fusion was used to determine the amount of K in the washed solids.

(f) The Na values from the KOH fusion were corrected for the high process blank.

(g) Quantified by IC system as fluoride, but slight retention time peak shift and peak shape suggest significant organic anion interference. It is highly probable that there is little or no fluoride actually present in the sample.

Table 11. Concentrations in the Leached and Untreated Solids and the Relative Amount of Each Component Removed by Caustic Leaching

	Leached Solids[a]		Original Sample[c]			
Analyte	µg or µCi/g dry solids	Pseudo 95% C.I. (if %RSDs=10)[b]	µg or µCi/g sample	Pseudo 95% C.I. (if %RSDs=10)[b]	Removed,%[d]	Pseudo 95% C.I. (if %RSDs=10)[c]
Ag	1790	± 253r	339	± 48r	1	± 0.3r
Al	34250	± 4849r	127892	± 22539r	95	± 24r
Ba	339	± 48r	64	± 9r	3	± 0.7r
Ca	8131	± 1158r	1549	± 218r	4	± 0.9r
Cd	1660	± 235r	327	± 44r	4	± 1.1r
Co	63	± 9r	12	± 2r	30	± 8r
Cr	1895	± 268r	825	± 93r	57	± 11r
Cu	452	± 64r	91	± 12r	7	± 1.6r
Fe	81350	± 11517r	15322	± 2168r	0	± 0.0r
Hg	159	± 22r	30	± 4r	--	--
K	2700	± 382r	1200	± 146r	58	± 13r
La	265	± 37r	50	± 7r	9	± 2.2r
Mg	2180	± 308r	419	± 58r	6	± 1.3r
Mn	18775	± 2658r	3534	± 500r	0	± 0.01r
Mo	< 51	--	< 11	--	> 54	--
Na[d]	34850	± 4929r	441136	± 75795r	99	± 24r
Ni	5550	± 785r	1068	± 148r	2	± 0.5r
P	4690	± 663r	3020	± 418r	71	± 16r
Pb	3043	± 431r	639	± 82r	10	± 2r
Si	22400	± 3168r	4684	± 600r	10	± 2r
Th	116500	± 16476r	21931	± 3102r	1	± 0r
Ti	395	± 56r	75	± 11r	1	± 0.3r
U	100100	± 14156r	18885	± 2665r	0	± 0.05r
Zn	330	± 47r	66	± 9r	5	± 1.1r
Zr	102500	± 14496r	19295	± 2729r	0	± 0.01r
TOC	16950	± 2397r	7329	± 451r	56	± 10r
TIC	6900	± 972r	5461	± 183r	76	± 12r
Cl^-	160	± 23r	910	± 211r	97	± 26r
F^-	2850	± 403r	43448	± 6478r	99	± 21r
NO_3^-	1250	± 177r	6177	± 905r	96	± 20r
SO_4^{2-}	< 240	--	2384 > x > 1223	--	> 51	--
PO_4^{3-}	< 240	--	3606 > x > 2445	--	> 67	--
CN^-	23	± 3r	4	± 1r	--	--
NH_3	< 9	--	< 2	--	--	--

Table 11. Concentrations in the Leached and Untreated Solids and the Relative Amount of Each Component Removed by Caustic Leaching (con't)

Analyte	Leached Solids[a] µg or µCi/g dry solids	Leached Solids[a] Pseudo 95% C.I. (if %RSDs=10)[b]	Original Sample[c] µg or µCi/g sample	Original Sample[c] Pseudo 95% C.I. (if %RSDs=10)[b]	Removed,%[d]	Pseudo 95% C.I. (if %RSDs=10)[c]
^{137}Cs	1.36E+02	± 1.92E+01r	4.61E+01	± 4.75E+00r	45	± 8r
^{90}Sr	2.82E+03	± 3.99E+02r	5.31E+02	± 7.51E+01r	0.004	± 0.001r
^{99}Tc	5.95E-02	± 8.41E-03r	2.00E-02	± 2.04E-03r	44	± 8r
^{241}Am(()	2.63E+01	± 3.72E+00r	4.98E+00	± 7.00E-01r	< 0.5	± 0.1r
^{241}Am(")	2.60E+01	± 3.67E+00r	--	--	--	--
^{154}Eu	6.64E+00	± 9.38E-01r	1.25E+00	± 1.77E-01r	< 0.1	± 0.03r
^{155}Eu	4.21E+00	± 5.95E-01r	8.19E-01	± 1.12E-01r	< 3	± 1r
^{14}C	< 4E-03	--	< 8E-04	--	--	--
^{129}I	< 3E-04	--	< 6E-05	--	--	--
^{235}U	1.50E-03	± 2.12E-04r	2.82E-04	± 3.99E-05r	--	--
^{238}U	3.26E-02	± 4.60E-03r	6.13E-03	± 8.67E-04r	--	--
^{237}Np	3.26E-02	± 4.60E-03r	6.13E-03	± 8.67E-04r	--	--
^{238}Pu	2.82E+00	± 3.98E-01r	5.30E-01	± 7.49E-02r	--	--
^{239}Pu	9.62E+00	± 1.36E+00r	1.81E+00	± 2.56E-01r	--	--
^{240}Pu	3.34E+00	± 4.72E-01r	6.28E-01	± 8.88E-02r	--	--
$^{239+240}$Pu	2.61E+01	± 3.68E+00r	4.90E+00	± 6.93E-01r	--	--
$^{243+244}$Cm	3.61E-01	± 5.11E-02r	6.80E-02	± 9.61E-03r	--	--
^{242}Cm	7.33E-02	± 1.04E-02r	1.38E-02	± 1.95E-03r	--	--
Total Alpha	5.84E+01	± 8.26E+00r	1.10E+01	± 1.55E+00r	0.01	± 0.002r

(a) The concentration in the leached solids was determined by summing the quantity found in the leached solids (Table 10) and dividing by the total weight (7.6051 g) of the leached solids (dry basis at 105°C).

(b) Pseudo 95% Confidence Intervals (C.I.) were approximated using propagation of error techniques for the case where the %RSD of all analytical measures used is 10% and all measures are independent The reader can review other potential %RSD values by multiplying the cell value by r, where r is %RSD/10.

(c) The concentration in the as-received sample was determined by summing the quantity found in the leach and wash solutions (Table 9) and the leached solids (Table 10) and dividing by the total weight (40.4 g) of sample used. Exceptions to this are cyanide, ammonia, mercury, C-14, I-129, U-235, U-238, Np-237, Pu-238, Pu-239, Pu-240, Pu-239+240, Cm-242, and Cm243+244. For these analytes only that found in the leached solids was included in the calculation (the wash solutions were not analyzed for these).

(d) The percent removed was determined by the following formula: $\%Removed = 100*F_w/(F_w+F_s)$; where F_w is the fraction in the wash and leach solutions, and F_s is the fraction in the leached solids.

(e) The values for Na are not corrected for Na added as NaOH during the leaching process.

5.0 Conclusions and Recommendations

The solubility versus temperature test indicated that the concentrations of Ag and Cr increased with increasing temperature and the concentrations of Cd, Fe, and P decreased with increasing temperature. Data for many of the other analytes were scattered to the point that statistically meaningful conclusion could not be drawn. The considerable variability observed for many of the components might have been due to precipitation of these components. It is recommended that the solubility versus temperature test plan be revised for future tests. The revised test should allow for larger sample sizes, immediate acidification of analytical samples (where appropriate), and should describe actions to be taken to minimize sample evaporation during interim storage of samples.

Dilute hydroxide washing largely removed most of the Na salts from the C-104 sludge. Dilute hydroxide washing was largely ineffective at removing Al (2%), Cr, (15%), or P (31%) from the C-104 sludge sample. Cesium-137 (64%) and ^{99}Tc (43%) were appreciably removed by dilute hydroxide washing, whereas the transuranic elements (as represented by the total alpha data) showed little solubility in the washing solutions.

Caustic leaching resulted in significantly better Al removal, with a total of 95% being removed. Improved Cr (57%) and P (71%) removals were also achieved by caustic leaching. Interestingly, caustic leaching did not result in additional ^{137}Cs or ^{99}Tc removal. The leached solids had very high concentrations (~10 wt%) of Th, U, and Zr.

The solutions generated by washing the C-104 solids with 0.01 M NaOH were stable over a period of ~5.5 months. However, the caustic leaching solution was not stable. A gel-like material had formed from the caustic leaching solution after ~20 days and considerable solids were present after 5.5 months.

6.0 References

Brooks, K.P., R.L. Myers, and K.G. Rappe. 1997. *Bench-Scale Enhanced Sludge Washing and Gravity Settling of Hanford Tank C-104 Sludge*, PNNL-11432, Pacific Northwest National Laboratory, Richland, Washington.

Lumetta, G.J., M.J. Wagner, F.V. Hoopes, and R.T. Steele. 1996. *Washing and Caustic Leaching of Hanford Tank C-104 Sludge*, PNNL-11381, Pacific Northwest National Laboratory, Richland, Washington.

Appendix A. Test Plan

Appendix B. Raw Data

Appendix C. Calculations

Appendix D. Statistical Analysis of the Data

Statistical analyses were performed on the data included in this report. In general, simple summary statistics were provided throughout that included estimates of the average (Mean), standard deviation (Std. Dev.) and percent relative standard deviation (%RSD = 100*Std. Dev./Mean) of aliquots. If one or both of the duplicate values were within 10 times the detection limit (values in parentheses in the tables) their mean and standard deviation are also marked in parentheses in the tables. By convention values less than the detection limit ("<") are formatted with 1 significant digit, values within 10 times the detection limit are formatted with 2 significant digits, and all other values are formatted with 3 significant digits.

More detailed statistical analyses included:

- Solubility versus Temperature Study Regression & Modeling Analyses
- Solubility versus Temperature Study Tests for Changes due to Temperature
- Washing and Leaching Studies Estimates of Uncertainty for analyte concentrations in the washed and original untreated solids and the percent removal

For all of the following analyses, it should be kept in mind that all data in each study are taken from one run of the experiment on a single sample. This means that the conclusions may be limited to this particular sample for this particular run. The data provide no information about the additional uncertainty that would result from running different samples or from repeating the experiment on similar samples. The only sources of variability present in these studies are sub-sampling variability and measurement variability. Consequently, the uncertainty statements developed in this report probably underestimate the variability that will be experienced in the real world application of these conclusions.

Solubility vs Temperature Study Regression & Modeling Analyses

The statistical analyses performed here are quantitative assessments of the nature of the relationship between analyte concentrations and temperature. These analyses were performed using the evaporation-adjusted concentrations from Tables 1, 2 and 3. The data were taken from the original Excel spreadsheet and may have additional digits compared to the formatted table values. Only those analytes that had two or fewer of their reported values below the detection limit were used. These statistical analyses were performed using linear and non-linear regression procedures in the Statistical Analysis System (SAS Institute Inc. Cary, North Carolina). Two approaches were used: polynomial regressions that attempt to fit the data without a specified mechanistic model, and a psuedo-Arrhenius model.

Since there are only three temperature points (30, 40, and 50°C), the maximum polynomial regression model that can be fit as a function of temperature is a quadratic. The two concentration values per temperature provide for estimating sub-sampling and measurement uncertainty and for testing the lack-of-fit of the linear regression. The general approach taken was to first fit and test a linear regression, i.e., is a linear regression statistically better than no model. This was followed by a test of the lack-of-fit of the linear regression model, or equivalently in this case, whether adding the quadratic term would be useful in describing the solubility-temperature relationship. It should be noted that no model

can fit this data better than a quadratic model, so if the quadratic model is not significantly better than the linear model, then no other model will be significantly better either.

Table D.1. Linear and Quadratic Polynomial Regression Analyses

Analyte	Estimated Intercept	Estimated Increase per °C	P-value	
			Simple Linear	Quadratic/ Lack-of-Fit
Ag	-0.707	0.0298	0.001	0.188
Al	9.87	-0.115	0.468	0.107
Ba	3.07	-0.0621	0.154	0.373
Ca	-8.82	1.01	0.225	0.983
Cd	4.16	-0.0496	0.002	0.382
Co	0.889	-0.000740	0.705	0.656
Cr	8.18	0.633	0.004	0.946
Cu	3.43	-0.0122	0.203	0.438
Fe	2.42	-0.0231	0.085	0.864
K	254	-0.292	0.687	0.471
Mg	-6.42	0.481	0.143	0.842
Mo	3.79	-0.00893	0.419	0.297
Na	29,256	124	0.353	0.035
Ni	64.0	-0.323	0.108	0.742
P	768	-5.36	0.026	0.681
Si	23.8	8.09	0.123	0.855
Zn	6.77	-0.0848	0.318	0.236

The regression estimates are grayed-out (judged unusable) if: the estimated increase is not significantly different from zero (linear p-value > 0.1) or the lack-of-fit of the linear regression is significant (lack-of-fit p-value < 0.1).

Table D.1 presents the results of the linear and quadratic polynomial regression analyses. Included are the estimates of the intercept and slope for the linear regression. Also included are the probabilities (p-values) for the test of the linear regression and for the test of the lack-of-fit of the linear regression. A significance level of 0.10 was used. Those analytes that have a significant linear regression have a simple linear p-value < 0.10. Those analytes that have a significant lack-of-fit from the linear regression have a lack-of-fit/quadratic p-value < 0.10. Those analytes that did not have a significant linear regression or had a significant lack-of-fit are grayed-out in the table to indicate that their linear regression estimates are not considered useable.

The proposed psuedo-Arrhenius dissolution model has the following form:

$$\text{Concentration} = e^{\text{B-A/Temperature}}$$

or the algebraically equivalent form

$$\ln(\text{Concentration}) = \text{B-A/Temperature}.$$

Although these two forms are algebraically equivalent, the estimates of A and B can be different depending on which form is fit due to the least-squares criterion for fitting being applied in regular space (the first form) or log space (the second form). If there is not much variability in the data the estimates of A and B should be close by either form, if there is large variability in the data the estimates of A and B can be quite different between the two forms. Review of the data did not indicate any particular reason to use the second form, such as increasing variability with increasing concentrations, so the first form was used. This will also make the results more comparable to the polynomial regressions, which were done in regular space.

Table D.2. Dissolution on Kinetics Model [Concentration = exp (B-A/Temperature)]

			Asymptotic 90% Confidence Interval	
Analyte	**Estimated B**	**Estimated A**	**Lower**	**Upper**
Ag	1.98	110	79.1	141
Al	0.317	-49.7	-130	30.6
Ba	-7.17	-226	-687	236
Ca	4.74	50.4	-32.1	133
Cd	-0.109	-33.4	-42.6	-24.2
Co	-0.178	-1.02	-7.80	5.76
Cr	4.25	28.9	17.9	39.9
Cu	0.936	-5.44	-14.4	3.56
Fe	-0.192	-22.5	-43.6	-1.49
K	5.46	-1.25	-10.1	7.61
Mg	4.08	60.8	-19.4	141
Mo	1.16	-3.03	-12.5	6.4
Na	10.6	6.49	-3.80	16.8
Ni	3.70	-8.87	-18.9	1.13
P	5.96	-13.6	-23.2	-3.96
Si	6.77	36.0	-5.37	77.3
Zn	-0.0536	-47.2	-111	16.8

The kinetic model estimates are grayed-out (judged unusable) if: the asymptotic 90% confidence interval of the temperature related parameter A includes zero.

Table D.2 presents the results for the proposed psuedo-Arrhenius dissolution model. Included are estimates of the B and A parameters. Also included are 90% confidence intervals on the temperature related A parameter. Those analytes whose confidence interval on A includes zero (i.e., those for which the lower value is negative and the upper value is positive) are grayed out in the table to indicate that their psuedo-Arrhenius dissolution estimates are not considered useable.

Plots for all analytes assessed are included. The following plotting symbols are used for the data:

- filled diamond—data that was ∃10-times the detection limit

- empty diamond—data that was <10-times the detection limit
- descending triangle—detection limit

The plots also show the linear regression with a solid line, 90% confidence intervals on the mean with dashed lines, and the psuedo-Arrhenius dissolution model with a dotted line. Occasionally, a confidence interval is so wide it goes off the plot.

The aliquot variability is relatively large for some analytes and, along with small sample numbers, leads to "non-significant" regressions for some analytes that may appear to show a relationship. Five of the analytes in Table D.1 had linear p-values <0.1 and quadratic p-values >0.1 and produced useable linear regression equations. These same five analytes showed useable psuedo-Arrhenius dissolution models in Table D.2. Visual comparison of the linear regression model and psuedo-Arrhenius dissolution model indicated they would produce very similar results in most cases. This may be an indication that, even if the psuedo-Arrhenius dissolution model might be better over a larger range of temperatures, the relationships can be closely approximated by a simple straight line over the 30 to 50°C range.

Solubility vs Temperature Study Tests for Changes Due to Temperature

Concentration changes in the Solubility versus Temperature study are expressed as the concentration change at each temperature relative to the concentration at 30°C. This is calculated as $100*(C_T-C_{30})/C_{30,}$ where C_T is the average concentration at temperature = T (40 or 50°C) and C_{30} is the average concentration at 30°C. Table 4 shows these estimates of the change in concentrations for detected analytes for the unadjusted data and Table 5 shows them for the adjusted data.

The following method was used to judge whether the reported changes were significantly different from zero or could instead simply be an artifact of sub-sampling and measurement uncertainty. There is very little data here to estimate the variability at any one temperature with any confidence so a pooled estimate of uncertainty was obtained by pooling the %RSDs at the three temperatures. This assumes the RSDs are relatively constant at each temperature. This result in turn was used as input to standard propagation-of-errors calculations for the variance of the estimation formula $100*(C_T/C_{30})-100$. This results in an estimate of the standard deviation of the % Change as C_T/C_{30}*sqrt(2)*Pooled %RSD. This in turn, was used to generate the range of a two-sided 90% confidence interval using a t-table value of 2.353 for 3 degrees of freedom. Any % Change that is larger than the range indicates the % Change is probably different from zero and is considered strong evidence of a change in concentration. These significant temperature-related changes are bold-faced in Tables 4 and 5.

Washing and Leaching Studies Estimates of Uncertainty for Analyte Concentrations in the Washed and Original Untreated Solids and the Percent Removed

The ability to derive estimates of uncertainty for the values reported in Tables 8 and 11 was even more hampered than it was for the % Change estimates discussed in the previous section. The calculation of the concentrations in Washed or Leached Solids and the

Original Sample were made using a number of sample weights and fraction constituent amounts. Only one of these inputs, namely the solids fraction, had duplicate aliquot data that could be used to estimate sub-sampling and measurement variability. The % Removed calculation in these two tables is even more problematic because of the use of even more terms and because it is the ratio of two other estimates.

To get at least some handle on the uncertainty of these estimates the following approach was taken:

- Treat all weights used in the estimation formulas as constants (without error) under the assumption that their uncertainties are much smaller than the uncertainties in the concentration measurements and can be safely ignored.
- Assume that duplicate aliquots had a common variability.
- Present a "pseudo" 95% confidence interval for at least one value of a %RSD that is assumed to be equal for all measurements that were used in any equation. A %RSD of 10 was chosen as the initial candidate as it appeared to be near the median of %RSDs seen in this study and seems to represent a reasonable starting point. This selected estimate on the uncertainty can be adjusted to determine the effects of other %RSD values by multiplying the "pseudo" 95% confidence interval values by the ratio of any other practicable %RSD divided by 10.

As input to the "pseudo" 95% confidence intervals, it was necessary to again use propagation of errors techniques to develop approximate standard deviations. These standard deviations were then multiplied by 2 (close to 1.96 from a standard normal distribution) to give the "pseudo" confidence interval half widths. Note that the use of the standard normal distribution does not account for the minimal amount of data available for these estimates, and provides much smaller confidence intervals than those that would be obtained using a Student's t distribution with only a few degrees of freedom.

For concentrations in Washed or Leached Solids and the Original Sample, the calculations are simple additions of fraction amounts divided by the sum of the corresponding fraction weights. The following propagation-of-error rules were used to develop propagation-of-errors formulas for their standard deviations:

- Variance of a mean is the variance of the measurement/n (the number of values used in the mean)
- The variance of a sum is the sum of the variances
- Constants (sample weights in this case) carry through.

This resulted in a general form for these two concentration estimates as:

$$\text{Std.Dev.} = \text{sqrt}\left(\Sigma_f(\text{var}(f)/n_f)\right)/\text{weights},$$

where f= each fraction used in the calculation of the concentration. Each var(f) term in the propagation-of-errors formula can be replaced, by definition, with $(mean_f*\%RSD)^2$. Since the same %RSD is assumed for all measurements, %RSD can be factored out, resulting in the following general formula:

$$\text{Std.Dev.} = \%RSD*\text{sqrt}\ (\Sigma_f(mean_f^2/n_f))/\text{weights}$$

The actual version of this general formula used for each analyte for each concentration estimate depends on the fractions that were used to calculate it and the number of sub-samples available for each fraction. Certain calculations used the same sample weights in the numerator and denominator. These were cancelled out and removed in the actual error propagation formulas.

For % Removal, the calculations involve 100 times the ratio of two terms, each of which is the sum of fraction amounts. The initial standard propagation-of-errors form of the Std.Dev. for this ratio of two terms is:

$$\text{Std.Dev.} = 100*num/den*\text{sqrt}(var(num)/num^2 + var(den)/den^2)$$

where num = the numerator term, den= the denominator term, and var() is the variance of each.
Both the numerator and denominator also need to have propagation-of-errors applied to them.
Again, each var() term in their propagation-of-errors formula can be replaced, by definition, with $(mean*\%RSD)^2$. Since the same %RSD is assumed for all measurements, %RSD can again be factored out, resulting in the following general formula:

$$\text{Std.Dev.} = 100*\Sigma_f mean_f/\Sigma_d mean_d*\%RSD* \text{sqrt}(\Sigma_f(mean_f^2/n_f)/(\Sigma_f mean_f)^2 + \Sigma_d(mean_d^2/n_d)/(\Sigma_d mean_d)^2)$$

where f = each fraction used in the numerator and d = each fraction used in the denominator. As for the concentration estimates discussed above, the actual version of this general formula used for each analyte depends on the fractions that were used to calculate the numerator and denominator and the number of sub-samples available for each fraction. Certain calculations use variances that have been calculated in prior steps, so these were put directly into the formulas instead of recalculating them.

Appendix A. Test Plan

Work Place Copy

G.J. Lumetta 7/7/99

PNNL Test Plan

Document No.: BNFL-TP-29953-8
Rev. No.: 0

Title: Determination of the Solubility of HLW Sludge Solids

Work Location: RPL/SAL

Page 1 of 21

Author: GJ Lumetta

Effective Date:
Supersedes Date: New

Use Category Identification: Mandatory

Identified Hazards:
__ Radiological
__ Hazardous Materials
__ Physical Hazards
__ Hazardous Environment
__ Other:

Required Reviewers:
X Technical Reviewer
__ Building Manage
__ Radiological Control
__ ES&H
X Quality Engineer
X Other: Client
X Other: Project Manager
X Other: RPL Manager

Are One-Time Modifications Allowed to this Procedure?
x Yes __ No

NOTE: If Yes, then modifications are not anticipated to impact safety. For documentation requirements of a modification see SBMS or the controlling Project QA Plan as appropriate.

On-The Job Training Required? ___ Yes or _x_ No

FOR REVISIONS:

Is retraining to this procedure required? ___ Yes ___ No
Does the OJT package associated with this procedure require revision to reflect procedure changes? __ Yes__ No__ N/A

Approval:

	Signature	Date
Author	G.J. Lumetta	1/13/99
Technical Reviewer	Brian M. Rapko	11-14-99
RPL Manager		1/19/99
Project Manager	D E Kurath	1/14/99
RPG QE		1/15/99
BNFL	PS Thompson BNFL QA:	27/1999

Applicability

This test plan is to be used to determine 1) the aqueous-insoluble fraction of BNFL HLW sludge samples, 2) the caustic-insoluble fraction of BNFL HLW sludge samples, and 3) the effect of temperature on the solubility of solids in the BNFL HLW sludge samples. The work will be conducted in the SAL hot cells. The work will be conducted by Radiochemical Processing Group staff. This work is being done as part of the Technical Support to BNFL for Phase 1B project.

Test Objectives

Justification: This activity supports confirmation of the process sequence, equipment performance and design basis for the HLW entrained solids removal process. BNFL must complete research and testing activities conducted to confirm system design bases before 14 April 1999.

Objective: The purpose of this task is to obtain the information needed in the filtration and washing of the Envelope D material. The specific objective of this test is to determine the relative mass and composition of the water-insoluble solids and of the caustic-insoluble solids (at 85°C) and to determine the components in the liquid portion of the HLW sample at 30, 40, and 50°C and their concentrations.

Definitions

BNFL	British Nuclear Fuels Ltd.
HDPE	High-density polyethylene
HLW	High-level waste
RPL	Radiochemical Processing Laboratory

Emergency Response

In the event of building audible alarms (e.g., fire or criticality) personnel should proceed in accordance with the RPL Building Emergency Procedure. If time permits, ensure that test materials are secured from spilling prior to exiting the area.

Quality Control

Quality assurance for work conducted under this Test Plan is governed by the Standards-Based Management System (SBMS). The quality control for each analysis indicated in Table 1 will be established per Quality Assurance Plan MCS-033. MCS-033 specifies the minimum calibration and verification requirements for analytical systems, as well as batch processing quality control samples to monitor preparations (i.e., blanks, duplicates, matrix spikes, and laboratory control standards).

A work place copy of this document shall be present at the work location. Specific information regarding each test (e.g., sample numbers) will be recorded on the work place copy and kept as project records.

As discussed in the Prerequisites section, calibrated balances must be used in performing this test. Likewise, a calibrated temperature controller is required. The calibration ID, date of calibration, and calibration expiration date must be recorded on the work place copy for each balance used and for the temperature controller.

Measured weights will be recorded on the work place copy at the indicated spot in the work instructions.

Hand written changes or corrections made to the work place copy will be made by means of a single line-out. Such changes or corrections shall be initialed and dated by the staff member making the change and by the cognizant scientist.

Equipment Description

A standard laboratory hot plate/magnetic stirrer will be used for this test. An aluminum heating block will be placed on the hot plate/stirrer to heat the sample. The apparatus will be equipped with two thermocouples. One of the thermocouples will be connected to a temperature controller, while the other will be connected to an over-temperature shut-off device. The latter will be used to ensure the sample is not over heated, which could result in lose of sample.

Prerequisites

Staff performing the work must read and understand the entire test plan prior to beginning work.

The following are items that should be staged prior to start of the test.

- Wide-mouth HDPE bottle; size to be determined (2)
- 30-mL HDPE bottle
- 20-mL HDPE vial (8)
- 30- to 40-mL glass vials (2)
- Hot plate/stirrer
- Aluminum heating block
- Temperature controller with temperature read-out
- Over-temperature shut-off device
- 0.45-µm nylon syringe filters (6)
- 5-mL syringes (6)
- 0.45-µm nylon disposable filter units (8)
- Adjustable 5-mL pipette
- Boiling water bath
- Small plastic bag

The temperature controller shall be calibrated by maintenance services. Record the following information regarding the temperature controller used.

		thermocouples	
Calibration ID:	0 2093	02899	02900
Calibration Date:	1/12/99	1/99	1/99
Expiration Date:	1/2000	1/2001	1/2001

A calibrated balance is required for this test. Record the following information regarding the balance(s) used.

Calibration ID:	360-06-01-016	Calibration ID:	
Calibration Date:	3/2/99	Calibration Date:	

Expiration Date: 8/99 Expiration Date: _____

Before beginning work, a routine performance check should be performed and documented in the space below.

Std. mass	Reading
1	1.0000
10	9.9995
50	49.9993
100	~~99.9972~~ 99.9996 (a)
150 (100+50)	150.0008

M.J. Lunetta 7/19/99

(a) Ran balance through calibration procedure.

Work Instructions

> **Note**
> Where practical, catch pans should be used when working with the tank waste samples, so that they can be recovered if spilled.

Part 1. Solubility Versus Temperature

Started on 7/19/99

1.1. Prepare the sample vials according to the following table. All vials should be HDPE.

Sample ID[a]
C104-SOL-30-1
C104-SOL-30-2
C104-SOL-40-1
C104-SOL-40-2
C104-SOL-50-1
C104-SOL-50-2

(a) The prefix to the sample IDs should be the tank number, e.g. "C106."

1.2. Label a 30-mL HDPE bottle as "C104-SOL-TEST" (_____ = tank number) and place a magnetic stir bar in this bottle. wt. = 15.3308

1.3. Mix the stock HLW sample to give a homogeneous slurry

→ Label = C104 GL

Used a shaker to mix the sample. The sample was very thick → like molasses.

(see next page)

M.J. Lunetta 7/19/99

As noted on the previous page, the sample was very thick. To fluidize a little better, 20mL of 0.1 M NaOH was added. Shook again to homogenize. It was fluid enough to pour at this point.

Note
If the HLW sample does not contain a liquid fraction, then add ~5 g of sludge to 25 mL of 0.1 M NaOH.

1.4. Transfer approximately 25 mL of the homogenized HLW slurry to C104 -SOL-TEST wt. = 46.3767

46.3767
- 15.3308
31.0459 g slurry

1.5. Place C104 -SOL-TEST into an aluminum heating block thermostatted at 30°C

1.6. Stir the contents of C104 -SOL-TEST

1.7. Once the temperature has equilibrated at 30°C, stir the sample for 1 h T = 31°C (A- some reason would not settle back to 30°C)

Start date/time: 7-19-99 3:45
Stop date/time: 7-20-99 10:00 AM

1.8. Preheat two syringe/filter assemblies by placing them in a plastic bag and submersing the plastic bag with the syringe/filters into a boiling water bath

Tare wts

1.9. Withdraw a 4-mL aliquot of the slurry and filter into vial C104 -SOL-30-1 6.5218 7.407

1.10. Withdraw a second 4-mL aliquot of the slurry and filter into vial C104 -SOL-30-2 6.4870 7.3496

1.11. Adjust the temperature of aluminum heating block assembly to 40°C

Unable to filter all 4 mL. Filter plugged.

Gross wts.

1.12. Once the temperature has equilibrated at 40°C, stir the sample for 1 h

Start date/time: 7/20/99 ~13:00
Stop date/time: 7/21/99 13:10

1.13. Preheat two syringe/filter assemblies by placing them in a plastic bag and submersing the plastic bag with the syringe/filters into a boiling water bath

Tare wts

1.14. Withdraw a 4-mL aliquot of the slurry and filter into vial C104 -SOL-40-1 6.5451 7.4937 (4 x 1.2 = 7/21/99)

1.15. Withdraw a second 4-mL aliquot of the slurry and filter into vial C104 -SOL-40-2 6.4873 7.3410 (4 x 1.2 = 7/21/99)

1.16. Adjust the temperature of aluminum heating block assembly to 50°C

1.17. Once the temperature has equilibrated at 50°C, stir the sample for 1 h

Start date/time: 7/21/99 13:30
Stop date/time: 7-22-99 10:30

1.18. Preheat two syringe/filter assemblies by placing them in a plastic bag and submersing the plastic bag with the syringe/filters into a boiling water bath

Tare wt Gross wt.

1.19. Withdraw a 4-mL aliquot of the slurry and filter into vial C104 -SOL-50-1 6.4773 7.6962 (4 x 1.2 = 7/21/99)

42 N.I.L. 7/21/99

1.20. Withdraw a second 2-mL aliquot of the slurry and filter into vial C104 -SOL-50-2 (Tare wt. 6.5736 Gross wt. 7.8534)

1.21. The samples collected during the test are to be submitted for the analyses listed in Table 1. The cognizant scientist will prepare the required ASR.

7/27/99 N.I.L.

Part 2. Determination of Aqueous-Insoluble Fraction

2.1. Homogenize the stock HLW sample by stirring → Put on shaker for ~15 min

2.2 Label a disposable filter unit (0.45-μm nylon) as C104 -AQ-1

2.3 Weigh C104 -AQ-1

Wt. C104 AQ-1 = 64.9439 g (with lid on) (2.3A)
64.9425 (re-weighed after wrench used to loosen top).

Also weigh just the bottom part of the filter unit; i.e., the receiving bottle and cap

Wt. receiving bottle&cap = 41.7325 g (2.3B)

2.4 Connect C104 -AQ-1 to the vacuum line, but do not yet apply vacuum

2.5 Transfer enough of the homogenized HLW sample to give ~25 g solids to the filter funnel of C104 -AQ-1 ~40mL of the slurry.

2.6 Apply vacuum to the filter unit. Disconnect from the vacuum once the liquid has filtered. Filtration was slow. . .

2.7 Place the cap on the top of the filter unit and weigh C104 -AQ-1

Wt. C104 -AQ-1 = 115.8190 g (2.7A)

Carefully remove the funnel part of the apparatus from the receiving bottle, place the cap on the receiving bottle and weigh.

Wt. receiving bottle&cap = 59.2175 g (2.7B)

2.8 Determine the total weight of the sample

Wt. Sample = 2.7A-2.3A = 50.8765 g (2.8A)

Determine the weight of the filtered liquid

Wt. Liquid = 2.7B-2.3B = 17.4850 g (2.8B)

Determine the weight of the filtered solids

Wt. Solids = 2.8A-2.8B = 33.3915 g (2.8C)

2.9 Measure out the appropriate volume of 0.01 M NaOH as instructed by the cognizant scientist into a plastic bottle

↓ Prepared 4/23/99 by GJL.

N.I. 7/27/99

7/27/99

Vol. Used = ~100 mL (2.9A)

2.10 Label an appropriately sized (125-mL) wide-mouthed HDPE bottle as C104 -AQ-2

2.11 Weigh C104 -AQ-2

Wt. C104 -AQ-2 = 31.7939 g (2.11A)

2.12 Slurry the filtered solids using a portion of 0.01 M NaOH (volume = 2.9A ÷ 5); transfer this slurry to C104 -AQ-2 most of the solids transferred with a spatula. Residual slurried in 0.01 M NaOH at transferred to C104-AQ-2.

2.13 Repeat step 2.12 four times to ensure complete transfer of the solids to C104 -AQ-2 Filled to capacity with 0.01 M NaOH

Note The stuff was pretty sticky. stuck together in big clumps. Like silly-putty.

2.14 Weigh C104 -AQ-2

Note Exceeded capacity of calibrated balance. Used different balance. Performance check 100-g weight → 100.1

Wt. C104 -AQ-2 = 169.9 g (2.14A)

Determine the weight of the slurry

Wt. Slurry = 2.14A-2.11A = 138.1 g (2.14B)

2.15 Equip C104 -AQ-2 with a condenser, then place in an aluminum heating block at 85°C

2.16 Stir the sample in C104 -AQ-2 at 85°C for a minimum of 8 hours

Start date/time: 7/27/99 15:45 16.5 h
Stop date/time: 7/28/99 8:15

~~2.17 Allow to cool to ambient temperature~~

~~2.18 Remove the condenser and replace the original cap on _____ AQ-2. Weigh _____ AQ-2.~~

Per instructions from Mike Johnson of BNFL, steps 2.17 and 2.18 were eliminated so that the solutions could be filtered while hot. M.J. Barnett 7/7/99

~~Wt. _____ AQ-2 = _____ g (2.18A)~~

~~Determine mass loss due to evaporation~~

~~Wt. Lost = 2.18A-2.14A = _____ g (2.18B)~~

7/28/99 2.19 Label a disposable filter unit (0.45-μm nylon) as C104 -AQ-3

2.20 Weigh C104 -AQ-3

Wt. C104 AQ-3 = 64.6591 g (2.20A)

Also weigh just the bottom part of the filter unit; i.e., the receiving bottle and cap

Wt. receiving bottle&cap = 41.5884 g (2.20B)

2.21 Connect C104 -AQ-3 to the vacuum line

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7/28/99

2.22 Filter the hot wash slurry Again, filtration was slow.

2.23 Disconnect from the vacuum once the liquid has filtered

2.24 Place the cap on the top of the filter unit and weigh C104-AQ-3

Too heavy to weigh in one piece.

wt. funnel w/ solids = 64.7063 Wt. C104-AQ-3 = 206.4288 g (2.24A) 141.7225 + 64.7063

Carefully remove the funnel part of the apparatus from the receiving bottle, place the cap on the receiving bottle and weigh.

Monitor the clarified solution for precipitate formation.

Wt. receiving bottle&cap = 141.7225 g (2.24B)

see p. 20

2.25 Determine the total weight of the slurry

Wt. Slurry = 2.24A-2.20A = 141.7697 g (2.25A)

Determine the weight of the filtered liquid

Wt. Liquid = 2.24B-2.20B = 100.1341 g (2.25B)

Determine the weight of the filtered solids

Note: solids apparently weigh more than before (see 2.8C)

Wt. Solids = 2.25A-2.25B = 41.6356 g (2.25C)

2.26 Measure out the appropriate volume of 0.01 M NaOH as instructed by the cognizant scientist into a plastic bottle

Vol. Used = ~100 mL (2.26A)

2.28 Weigh C104-AQ-2

Wt. C104-AQ-2 = _____ g (2.28A) Forgot to weigh. 11.6. 7/28/99

2.29 Slurry the filtered solids using a portion of 0.01 M NaOH (volume = 2.26A ÷ 5); transfer this slurry to C104-AQ-2 Transfer made in same manner as before. Solids still very sticky.

2.30 Repeat step 2.29 four times to ensure complete transfer of the solids to C104-AQ-2

2.31 Weigh C104-AQ-2 Filled to near capacity with 0.01 M NaOH.

Wt. C104-AQ-2 = 155.0752 g (2.31A)

Determine the weight of the slurry

Wt. Slurry = 2.31A-2.28A = 123.2813 g (2.31B) 2.11A

2.32 Equip C104-AQ-2 with a condenser, then place in an aluminum heating block at 85°C

7/28/99

2.33 Stir the sample in C104 -AQ-2 at 85°C for a minimum of 8 hours

Start date/time: 7/28/99 10:00
Stop date/time: 7/29/99 8:30

~~2.34 Allow to cool to ambient temperature~~

~~2.35 Remove the condenser and replace the original cap on _____ -AQ-2. Weigh _____ -AQ-2~~

~~Wt. _____ -AQ-2 = _____ g (2.35A)~~

~~Determine mass loss due to evaporation~~

~~Wt. Lost = 2.35A-2.31A = _____ g (2.36B)~~

S.A. Summette 7/7/99 see note on page 7.

2.36 Label a disposable filter unit (0.45-µm nylon) as C104 -AQ-5

2.37 Weigh C104 -AQ-5

Wt. C104 -AQ-5 = 64.7197 g (2.37A)

Also weigh just the bottom part of the filter unit; i.e., the receiving bottle and cap

Wt. receiving bottle&cap = 41.5731 g (2.37B)

2.38 Connect C104 -AQ-5 to the vacuum line

2.39 Filter the hot wash slurry

2.40 Disconnect from the vacuum once the liquid has filtered

2.41 Place the cap on the top of the filter unit and weigh C104 -AQ-5

exceeded capacity of balance. AK.

Wt. C104 -AQ-5 = ~~124.3634~~ g (2.41A)

Carefully remove the funnel part of the apparatus from the receiving bottle, place the cap on the receiving bottle and weigh.

Monitor the clarified liquid for the formation of solids.

Wt. receiving bottle&cap = 124.3634 g (2.41B)

see p. 20

2.42 Determine the total weight of the slurry

Wt. Slurry = 2.41A-2.37A = ~~123.2813~~ g (2.42A)

Can't determine because we didn't get 2.41A. S.A.S. 8/4/99

Determine the weight of the filtered liquid

Wt. Liquid = 2.41B-2.37B = 82.7963 g (2.42B)

Determine the weight of the filtered solids

Wt. Solids = 2.42A-2.42B = 40.4910 g (2.42C)

2.43 Measure out the appropriate volume of 0.01 M NaOH as instructed by the cognizant scientist into a plastic bottle

Vol. Used = ~100 mL (2.43A)

2.44 ~~Label an appropriately sized wide-mouthed HDPE bottle as _____-AQ-2~~ step. not needed. M.J. Smith 7/7/99

2.45 Weigh C104 -AQ-2

Wt. C104 -AQ-2 = ~~159.0003 g~~ (2.45A)

2.46 Slurry the filtered solids using a portion of 0.01 M NaOH (volume = 2.43A + 5); transfer this slurry to C104 -AQ-2

2.47 Repeat step 2.46 four times to ensure complete transfer of the solids to C104 -AQ-2

2.48 Weigh C104 -AQ-2

Wt. C104 -AQ-2 = ~~159.0002 g~~ (2.48A)

Determine the weight of the slurry

Wt. Slurry = 2.48A-~~2.45A~~ 2.11A = 127.2063 g (2.48B)

2.49 Equip C104 -AQ-2 with a condenser, then place in an aluminum heating block at 85°C

2.50 Stir the sample in C104 -AQ-2 at 85°C for a minimum of 8 hours

Start date/time: 7-29-99 10:30
Stop date/time: 7-30-99 10:30

~~2.51 Allow to cool to ambient temperature~~

~~2.52 Remove the condenser and replace the original cap on _____-AQ-2. Weigh _____-AQ-2~~

~~Wt. _____-AQ-2 = _____ g (2.52A)~~

~~Determine mass loss due to evaporation~~

~~Wt. Lost = 2.52A-2.48A = _____ g (2.52B)~~

M.J. Smith 7/7/99 see not on p. 7.

2.53 Label a disposable filter unit (0.45-µm nylon) as C104 -AQ-7

2.54 Weigh C104 -AQ-7

Wt. C104-AQ-7 = ~~63.4990 g~~ 64.8500 (2.54A)
7-30-99

Also weigh just the bottom part of the filter unit; i.e., the receiving bottle and cap

Wt. receiving bottle&cap = ~~40.4567 g~~ 41.6380 g 8-4-99 (2.54B)

2.55 Connect C104 -AQ-7 to the vacuum line

2.56 Filter the hot wash slurry

2.57 Disconnect from the vacuum once the liquid has filtered

2.58 Place the cap on the top of the filter unit and weigh C104 -AQ-7

Combined wt
= 71.7640
+134.7436
206.5076

Wt. C104 - AQ-7 = 71.7640 g (2.58A)

Top half

Carefully remove the funnel part of the apparatus from the receiving bottle, place the cap on the receiving bottle and weigh.

Monitor the clarified liquid for the formation of solids.

Wt. receiving bottle&cap = 134.7436 g (2.58B)

See p. 20

2.59 Determine the total weight of the slurry

Note: this is inconsistent with 2.48B

Wt. Slurry = 2.58A-2.54A = 141.6576 g (2.59A)

Determine the weight of the filtered liquid

Wt. Liquid = 2.58B-2.54B = 93.1056 g (2.59B)

Determine the weight of the filtered solids

Wt. Solids = 2.59A-2.59B = 48.5520 g (2.59C)

2.60 Label a glass vial as C104 -AQ-8

2.61 Dry C104 -AQ-8 at 105°C for a minimum of 1 h

2.62 Cool C104 -AQ-8 to ambient temperature in a desiccator

2.63 Weigh C104 -AQ-8

Wt. C104 -AQ-8 = 128.2633 g (2.63A)

2.64 Using several portions of deionized water, quantitatively transfer the washed solids from the filter membrane to C104 -AQ-8

2.65 Heat C104 -AQ-8 at 80°C to evaporate excess water

2.66 Heat C104 -AQ-8 at 105°C overnight

2.67 Cool C104 -AQ-8 to ambient temperature in a desiccator

2.68 Weigh C104 -AQ-8

Wt. C104 -AQ-8 = 142.6237 g (2.68A)

8-3-99

2.69 Determine the dry weight of the washed solids

Wt. Dry Solids = 2.68A-2.63A = 14.3589 g (2.69A)

N.I. Hunter 8/3/99

2.70 Determine the relative amounts of each wash solution needed to prepare the composite liquid sample

8/4/99 11:10 All solutions were clear.

17.4850 100.1341 82.7503 93.1056

Total Wt. Liquids = 2.8B + 2.25B + 2.42B + 2.59B = 293.5150 g (2.70A)

Wt Fraction AQ-1 = 2.8B/2.70A = 0.0596 (2.70B)

Wt Fraction AQ-3 = 2.25B/2.70A = 0.3412 (2.70C)

Wt Fraction AQ-5 = 2.42B/2.70A = 0.2821 (2.70D)

Wt Fraction AQ-7 = 2.59B/2.70A = 0.3172 (2.70E)

2.71 Label a 20-mL HDPE sample vial as C104 -AQ-9 → Tare wt: 8.1397

2.72 Place C104 -AQ-9 on the balance and tare to 0.000g

2.73 Add the following quantity of the solution in bottle C104 -AQ-1 to C104 -AQ-9

Quantity from C104 -AQ-1 = 10*2.70B = 0.5960 g (2.73A)

Record the weight of C104 -AQ-9

Wt. C104 -AQ-9 = 0.5978 g (2.73B)

2.74 Place C104 -AQ-9 on the balance and tare to 0.000g

2.75 Add the following quantity of the solution in bottle C104 -AQ-3 to C104 -AQ-9

Quantity from C104 -AQ-3 = 10*2.70C = 3.412 g (2.75A)

Record the weight of C104 -AQ-9

Wt. C104 -AQ-9 = 3.4310 g (2.75B)

2.76 Place C104 -AQ-9 on the balance and tare to 0.000g

2.77 Add the following quantity of the solution in bottle C104 -AQ-5 to C104 -AQ-9

Quantity from C104 -AQ-5 = 10*2.70D = 2.821 g (2.77A)

N.I. Hunter 8/4/99

Record the weight of C104 -AQ-9

Wt. C104 -AQ-9 = 2.8756 g (2.77B)

2.78 Place C104 -AQ-9 on the balance and tare to 0.000g

2.79 Add the following quantity of the solution in bottle C104 -AQ-7 to C104 -AQ-9

Quantity from C104 -AQ-7 = 10*2.70E = 3.172 g (2.79A)

Record the weight of C104 -AQ-9

Wt. C104 -AQ-9 = 3.3297 g (2.79B)

Determine the gross weight of C104-AQ-9 → Gross wt. C104-AQ-9 = 18.3751

2.73 The washed solids and the composite wash solution are to be submitted for the analyses listed in Table 1. The cognizant scientist will prepare the required ASR.

8/4/99

Part 3. Determination of Caustic-Insoluble Fraction

3.1. Homogenize the stock HLW sample by stirring

3.2 Label a disposable filter unit (0.45-μm nylon) as C104 -OH-1

3.3 Weigh C104 -OH-1

Wt. C104 -OH-1 = 64.5038 g (3.3A)

Also weigh just the bottom part of the filter unit; i.e., the receiving bottle and cap

Wt. receiving bottle&cap = 41.7462 g (3.3B)

3.4 Connect C104 -OH-1 to the vacuum line, but do not yet apply vacuum

3.5 Transfer enough of the homogenized HLW sample to give ~25 g solids to the filter funnel of C104 -OH-1

3.6 Apply vacuum to the filter unit. Disconnect from the vacuum once the liquid has filtered.

3.7 Place the cap on the top of the filter unit and weigh C104 -OH-1

Wt. C104 -OH-1 = 110.3460 g (3.7A)

Carefully remove the funnel part of the apparatus from the receiving bottle, place the cap on the receiving bottle and weigh.

Wt. receiving bottle&cap = 53.3086 g (3.7B)

N.J. Lumetta 8/4/99

8/4/99

3.8 Determine the total weight of the sample

Wt. Sample = 3.7A-3.3A = 45.8422 g (3.8A)

Determine the weight of the filtered liquid

Wt. Liquid = 3.7B-3.3B = 11.5624 g (3.8B)

Determine the weight of the filtered solids

Wt. Solids = 3.8A-3.8B = 34.2798 g (3.8C)

3.9 Measure out the appropriate volume of 3 M NaOH as instructed by the cognizant scientist into a plastic bottle

Vol. Used = ~100 mL (3.9A)

3.10 Label an appropriately sized wide-mouthed HDPE bottle as C104 -OH-2

3.11 Weigh C104 -OH-2

Wt. C104 -OH-2 = 31.88417 g (3.11A)

3.12 Slurry the filtered solids using a portion of 3 M NaOH (volume = 3.9A ÷ 5); transfer this slurry to C104 -OH-2

3.13 Repeat step 3.12 four times to ensure complete transfer of the solids to C104 -OH-2

3.14 Weigh C104 -OH-2

Exceeded capacity of calibrated balance. Used same balance used in step 2.14

Wt. C104 -OH-2 = 171.8 g (3.14A)

Determine the weight of the slurry

Wt. Slurry = 3.14A-3.11A = 139.9 g (3.14B)

3.15 Equip C104 -OH-2 with a condenser, then place in an aluminum heating block at 85°C

3.16 Stir the sample in C104 -OH-2 at 85°C for a minimum of 8 hours

Start date/time: 8/4/99 11:10
Stop date/time: 8/5/99 8:30 21.5 h

~~3.17 Allow to cool to ambient temperature~~

~~3.18 Remove the condenser and replace the original cap on _____ -OH-2. Weigh _____ -OH-2~~

~~Wt. _____ -OH-2 = _____ g (3.18A)~~

M. J. Smith 7/7/99 see note on p. 7

M. J. Smith 8/5/99

~~Determine mass loss due to evaporation~~

~~Wt. Lost = 3.18A-3.14A = _____ g (3.18B)~~ N.A. Smith 7/7/99 See note on p. 7

8/5/99

3.19 Label a disposable filter unit (0.45-μm nylon) as C104 -OH-3

3.20 Weigh C104 -OH-3

Wt. C104 -OH-3 = 64.3851 g (3.20A)

Also weigh just the bottom part of the filter unit; i.e., the receiving bottle and cap

Wt. receiving bottle&cap = 41.5677 g (3.20B)

3.21 Connect C104 -OH-3 to the vacuum line

3.22 Filter the hot leaching slurry

3.23 Disconnect from the vacuum once the liquid has filtered

3.24 Place the cap on the top of the filter unit and weigh C104 -OH-3

So total wt 140.4043 (3.24B)
64.2410
204.6953

Wt. C104 -OH AQ-3 = 64.2910 g (3.24A) → dust in the filter funnel ...

N.A. Smith 8/5/99

Carefully remove the funnel part of the apparatus from the receiving bottle, place the cap on the receiving bottle and weigh.

Monitor the clarified liquid for solids formation.
8/16/99 8:30 solution clear.
8/26/99 A gel-like material had formed on the bottom of the bottle (See p. 20)

Wt. receiving bottle&cap = 140.4043 g (3.24B)

3.25 Determine the total weight of the slurry

Wt. Slurry = 3.24A-3.20A = 140.3102 g (3.25A)

Determine the weight of the filtered liquid

Wt. Liquid = 3.24B-3.20B = 98.8366 g (3.25B)

Determine the weight of the filtered solids

Wt. Solids = 3.25A-3.25B = 41.4736 g (3.25C)

3.25a Label a 20-mL HDPE sample vial as C104 -OH-3A → Tare wt. = 8.4965

3.25b Transfer ~15 mL of the filtered leachate solution to C104 -OH-3A Gross wt = 26.0674

3.26 Measure out the appropriate volume of 0.01 M NaOH as instructed by the cognizant scientist into a plastic bottle

Vol. Used = ~100 mL (3.26A)

N.A. Smith 8/5/99

8/5/99

3.28 Weigh C104 -OH-2

Wt. C104 -OH-2 = _____ g (3.28A) Did not weigh. M.I.h

3.29 Slurry the filtered solids using a portion of 0.01 M NaOH (volume = 3.26A + 5); transfer this slurry to C104 -OH-2

3.30 Repeat step 3.29 four times to ensure complete transfer of the solids to C104 -OH-2

3.31 Weigh C104 -OH-2

Wt. C104 -OH-2 = 155.1 g (3.31A)

Determine the weight of the slurry

31.9 (3.11A)

Wt. Slurry = 3.31A-3.28A = 123.2 g (3.31B)

3.32 Equip C104 -OH-2 with a condenser, then place in an aluminum heating block at 85°C

3.33 Stir the sample in C104 -OH-2 at 85°C for a minimum of 8 hours

Start date/time: 8-5-99 11:30
Stop date/time: 8/6/99 8:30 21 h

~~3.34 Allow to cool to ambient temperature~~

~~3.35 Remove the condenser and replace the original cap on _____ -OH-2. Weigh _____ -OH-2~~

~~Wt. _____ -OH-2 = _____ g (3.35A)~~

~~Determine mass loss due to evaporation~~

~~Wt. Lost = 3.35A-3.31A = _____ g (3.35B)~~

M.I. 7/7/99 see note on p. 7

3.36 Label a disposable filter unit (0.45-μm nylon) as C104 -OH-5

3.37 Weigh C104 -OH-5

Wt. C104 -OH-5 = 64.6340 g (3.37A)

Also weigh just the bottom part of the filter unit; i.e., the receiving bottle and cap

Wt. receiving bottle&cap = 41.9917 g (3.37B)

3.38 Connect C104 -OH-5 to the vacuum line

3.39 Filter the hot wash slurry

Note: The solids seemed to filter a little faster than when we were doing just the 0.01 M NaOH wash. i.e. caustic leaching perhaps helped filterability. (But filtration was still slow.)

3.40 Disconnect from the vacuum once the liquid has filtered

M.I. 8/6/99

3.41 Place the cap on the top of the filter unit and weigh C104 -OH-5

Total 55.6883 + 134.4445 = 190.4328

Wt. C104 -OH-5 = 55.9883 g (3.41A)

Carefully remove the funnel part of the apparatus from the receiving bottle, place the cap on the receiving bottle and weigh.

TAP only

Monitor the clarified solution for precipitate.

see p. 20

Wt. receiving bottle&cap = 134.4445 g (3.41B)

3.42 Determine the total weight of the slurry

Wt. Slurry = 3.41A-3.37A = 125.7988 g (3.42A)

Determine the weight of the filtered liquid

Wt. Liquid = 3.41B-3.37B = 92.4528 g (3.42B)

Determine the weight of the filtered solids

Wt. Solids = 3.42A-3.42B = 33.3460 g (3.42C)

3.43 Measure out the appropriate volume of 0.01 M NaOH as instructed by the cognizant scientist into a plastic bottle

Vol. Used = ~100 mL (3.43A)

3.45 Weigh C104 -OH-2

Wt. C104 -OH-2 = 160.0 g (3.45A)

3.46 Slurry the filtered solids using a portion of 0.01 M NaOH (volume = 3.43A ÷ 5); transfer this slurry to C104 -OH-2

3.47 Repeat step 3.46 four times to ensure complete transfer of the solids to C104 -OH-2

3.48 Weigh C104 -OH-2

Inadvertently not determined N.I.L. 8/10/99

Wt. ~~C104 -OH-2~~ = _____ g (3.48A)

Determine the weight of the slurry

3.11A

Wt. Slurry = 3.48A-~~3.45~~A = _____ g (3.48B)

3.49 Equip C104 -OH-2 with a condenser, then place in an aluminum heating block at 85°C

3.50 Stir the sample in C104 -OH-2 at 85°C for a minimum of 8 hours

Start date/time: 8-9-99 10:00
Stop date/time: 8/10/99 ~8:30 22.5 h

~~3.51 Allow to cool to ambient temperature~~

~~3.52 Remove the condenser and replace the original cap on _____-OH-2. Weigh _____-OH-2~~

~~Wt. _____-OH-2 = _____ g (3.52A)~~

N.I. [illegible] 7/7/99 see note on p. 7

~~Determine mass loss due to evaporation~~

~~Wt. Lost = 3.52A-3.48A = _____ g (3.52B)~~

3.53 Label a disposable filter unit (0.45-μm nylon) as C104 -OH-7

3.54 Weigh C104 -OH-7

Wt. C104 -OH-7 = 64.7943 g (3.54A)

Also weigh just the bottom part of the filter unit; i.e., the receiving bottle and cap

Wt. receiving bottle&cap = 41.4921 g (3.54B)

3.55 Connect C104 -OH-7 to the vacuum line

3.56 Filter the hot wash slurry

3.57 Disconnect from the vacuum once the liquid has filtered

3.58 Place the cap on the top of the filter unit and weigh C104 -OH-7

Tot 57.2249 / 129.8028 / 187.0277

Wt. C104 -OH-7 = 57.2249 g (3.58A)

Carefully remove the funnel part of the apparatus from the receiving bottle, place the cap on the receiving bottle and weigh. top only

monitor the clarified solution for precipitate.

Wt. receiving bottle&cap = 129.8028 g (3.58B)

see p. 20

3.59 Determine the total weight of the slurry

Wt. Slurry = 3.58A-3.54A = 122.2374 g (3.59A)

Determine the weight of the filtered liquid

Wt. Liquid = 3.58B-3.54B = 88.3107 g (3.59B)

Determine the weight of the filtered solids

Wt. Solids = 3.59A-3.59B = 33.9227 g (3.59C)

3.60 Label a glass vial as C104 -OH-8

3.61 Dry C104 -OH-8 at 105°C for a minimum of 1 h

3.62 Cool C104 -OH-8 to ambient temperature in a desiccator

3.63 Weigh C104 -OH-8

Wt. C104 -OH-8 = 127.4270g (3.63A)

3.64 Using several portions of deionized water, quantitatively transfer the washed solids from the filter membrane to C104 -OH-8

3.65 Heat C104 -OH-8 at 80°C to evaporate excess water

3.66 Heat C104 -OH-8 at 105°C overnight

3.67 Cool C104 -OH-8 to ambient temperature in a desiccator

3.68 Weigh C104 -OH-8

Wt. C104 -OH-8 = 135.032 g (3.68A)

3.69 Determine the dry weight of the washed solids

Wt. Dry Solids = 3.68A-3.63A = 7.6051 g (3.69A)

3.70 Determine the relative amounts of each wash solution needed to prepare the composite liquid sample

92.4526 88.3107

Total Wt. Liquids = 3.42B + 3.59B = 180.7633 g (3.70A)

Wt Fraction OH-5 = 3.42B/3.70A = 0.5115 (3.70B)

Wt Fraction OH-7 = 3.59B/3.70A = 0.4885 (3.70C)

3.71 Label a 20-mL HDPE sample vial as C104 -OH-9 → Tare wt = 8.3703

3.72 Place C104 -AQ-9 on the balance and tare to 0.000g

3.73 Add the following quantity of the solution in bottle C104 -OH-5 to C104 -OH-9

Quantity from C104 -OH-5 = 10*3.70B = 5.115 g (3.73A)

Record the weight of C104 -OH-9

Wt. C104 -OH-9 = 5.1276 g (3.73B)

3.74 Place C104 -AQ-9 on the balance and tare to 0.000g

3.75 Add the following quantity of the solution in bottle C104 -OH-7 to C104 -OH-9

Quantity from C104 -OH-7 = 10*3.70C = 4.885 g (3.75A)

Record the weight of C104 -OH-9

Wt. C104 -OH-9 = 4.8866 g (3.75B)

Determine the gross weight of C104-OH-9 → Gross wt. C104-OH-9 = _____ g

3.76 The washed solids, the leaching solution, and composite wash solution are to be submitted for the analyses listed in Table 1. The cognizant scientist will prepare the required ASR.

END of Work Instructions

Can calculate as follows:

5.1276
4.8866
8.3703 (tare)
18.3845

Forgot to weigh. But analytical work started within 2 days.
M.I. [illegible]
8/13/99

9/29/99 Check for precipitates

M.I. [illegible]

1/18/2000 M.I. [illegible]

C104-AQ-3 → Clear
C104-AQ-5 → Clear
C104-AQ-7 → Clear
→ All still clear (so was C104-AQ-1)

C104-OH-3 → precipitate
C104-OH-5 → Clear
C104-OH-7 → Clear
→ -OH-3 & -OH-7 both had lots of solids. Solids formed a film on the bottom of -OH-7. Interestingly, -OH-5 was still fairly clear.

TABLE 1. Sample Matrix

Sample ID	Acid Digestion	KOH Fusion	Na_2O_2 Fusion	ICP/AES	IC (anions)	TOC	TIC	ICP-MS (^{99}Tc)	ICP-MS (Full)[a]	GEA	^{90}Sr	Total Alpha	Laser Fluorimetry (U)	$^{241}Am/^{243+244}Cm$	^{14}C	Tritium	CVAA Hg	Total CN	Ammonia
SOL-30-1	X			X	X	X	X	X		X	X	X	X						
SOL-30-2	X			X	X	X	X	X		X	X	X	X						
SOL-40-1	X			X	X	X	X	X		X	X	X	X						
SOL-40-2	X			X	X	X	X	X		X	X	X	X						
SOL-50-1	X			X	X	X	X	X		X	X	X	X						
SOL-50-2	X			X	X	X	X	X		X	X	X	X						
AQ-8		X	X	X	X	X	X		X	X	X	X	X	X	X	X	X	X	X
AQ-9	X			X	X	X	X		X	X	X	X	X	X	X	X	X	X	X
OH-3A	X			X	X	X	X		X	X	X	X	X	X	X	X	X	X	X
OH-8		X	X	X	X	X	X		X	X	X	X	X	X	X	X	X	X	X
OH-9	X			X	X	X	X		X	X	X	X	X	X	X	X	X	X	X

(a) Includes Tc-99, I-129, Np-237, U-isotopic, Pu-isotopic

Appendix B. Raw Data

Shielded Analytical Laboratory
SAMPLE CONFIRMATORY WEIGHTS

Tank C104 Core(s) N/A

Project Id: 29953 WP Number: W48486

TI/ASR Number: ASR 5478

Sample Ident.	Sample Weight (g)
C104-SOL-30-1	6.9222
C104-SOL-30-2	6.9279
C104-SOL-40-1	7.1768
C104-SOL-40-2	7.1976
C104-SOL-50-1	7.2932
C104-SOL-50-2	7.5184
C104-AQ-9	18.3523
C104-OH-3A	26.0570
C104-OH-9	18.3618

M&TE: X Cell 2 (360-06-01-016) Other _____

____ Cell 5 (360-06-01-019)

____ Denver (360-06-01-040)

__1__ Mettler AT201 (510-06-01-014)

Analyst: [signature] Date: 8-12-99 Reviewer: [signature] Date: 8/12/99

Battelle PNNL/325 Bldg/RPG/Inorganic Analysis ...
ICPAES Data Report

Project: 29953
Client: G. J. Lumetta

ACL Number(s): 99-2340 through 99-2350 & 99-2346, 99-2349

Client ID: "C104-SOL-30-1" through "C104-OH-9" & "C104-AQ-8", "C104-OH-8"

ASR Number: 5478.01

Total Samples: 11

Procedure: PNL-ALO-211, "Determination of Elements by Inductively Coupled Argon Plasma Atomic Emission Spectrometry" (ICP-AES).

Analyst: JJ Wagner

Analysis Date (Filename): 8-19-99 (A0540 K/Ni), 8-24-99 (A0541 Na/Zr), 8-27-99 (A0542 ALO-128)

See Chemical Measurement Center 98620: ICP-325-405-1 File for Calibration and Maintenance Records.

M&TE Number: ICPAES instrument -- WB73520
Mettler AT400 Balance -- Ser.No. 360-06-01-029

Jerry Wagner 9-14-99
Reviewed by

9-15-99
Concur

9/14/99

Battelle PNNL/325 Bldg/RPG/Inorganic Analysis ...
ICPAES Data Report

Nine radioactive liquid samples, C104-SOL-30-1 through C104-SOL-50-2, C104-AQ-9, C104-OH-3A and C104-OH-9 (ACL# 99-2340 through 99-2345, 99-2347, 99-2348, and 99-2350), were analyzed by ICPAES after preparation by the Sample Receiving and Preparation Laboratory (SRPL). Samples were prepared by SRPL using PNL-ALO-128 acid digestion procedure and plastic vials. Approximately 0.1 ml to 5.0 ml of sample (weighed) was processed and diluted to a final volume of 5 ml, 10 ml, or 20ml. The final volume was calculated by measuring the net weight and dividing by an estimated density. Density of each prepared sample was estimated by weighing a one ml aliquot of each processed sample. Samples received prior to digestion were clear solutions except C104-SOL-30-1 and C104-SOL-30-2. These two samples contained visible solids. The containers in the hot cell also had crystals in the liquids. After digestion all samples were clear and did not require filtering. Although sample C104-OH-3A (ACL# 99-2348) was received as a clear solution it formed a precipitate when acidified. After processing the sample was diluted to 10 ml final volume but some precipitate remained. The sample was then evaporated to near dryness, heated with 0.75 ml of concentrated hydrochloric acid and 0.25 ml of concentrated nitric acid. A precipitate still remained so 5 ml of water and 0.050 ml of concentrated (48%) hydrofluoric acid was added and warmed. After several minutes of continued warming, the precipitate dissolved and the sample was diluted to final volume of about 20 ml. The sample volume (18.7ml) was estimated as above using weight and density measurements.

Two radioactive solid samples each prepared in duplicate, C104-AQ-8 & C104-OH-8 (ACL# 99-2346 & 99-2349), were analyzed by ICPAES after preparation by SAL. Approximately 0.2g aliquots were used to prepare samples using both fusion procedures PNL-ALO-114 (Na_2O_2/Zr), and PNL-ALO-115 (KOH/Ni). After samples were fused they were diluted to a final volume of 100 ml. Samples were diluted an additional 2.01 or 2.03-fold by SAL using 2% v/v HCl because of ALARA radiation dose concerns. Additional dilution up to 25-fold was performed during ICPAES analysis because of high sodium, iron, thorium, uranium and/or zirconium concentration. Both fusion sample preparations required HCl to dissolve the fused samples. All solutions remained soluble after final dilution.

Measurement results reported have been corrected for preparation and analytical dilution. Specific analytes of interest requested by the client include Al, Cr, Fe, Na, Ni, Si, and U. Other required analytes include (table 4.2, page 27 "Analytical Requirements for Filtrate, Washed Solids, and Wash Solutions") Ag, Ba, Ca, Cd, Co, Cu, K, La, Mg, Mn, Mo, Na, Pb, Ti, Zn and Zr. All results reported are in µg/g including liquid samples as requested by the client. Volumes and weights have been recorded on bench sheets and included with final data report. A single element 1,000 ug/ml sodium standard was measured at a frequency of every ten sample measurements and varied from start to end by less than 4% (936, 973, 969, 962, and 945 ug/ml). Worse case bias for sodium is approximately –6%.

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Although not a requested analyte of interest it should be noted that thorium was present in the solid samples at high concentration. Thorium concentration is quite variable between the two fusion prepared samples. For example, in sample C104-AQ-8 (99-2346) thorium concentration between duplicates in the sodium peroxide/Zr fusion is nearly 50% different. There is also a difference in thorium concentration between the two types of fusion. Thorium concentration in sample C104-OH-8 (99-2349) for the sodium peroxide/Zr fusion is about 12 Wt% while the concentration of thorium is about half that for the same sample pair in the potassium/Ni fusion. The large differences may be due to in-homogeneity. Thorium was not detected in the acid digested aqueous samples.

Quality control check-standard results met tolerance requirements for analytes of interest except as noted below. Following is a list of quality control measurement results relative to ICPAES analysis tolerance requirements under MCS-033. Please note the final quality control check measurements at the very end of the run for the aqueous prepared samples (8-27-99 A0542) were somewhat higher than the tolerance limit. It is suspected that the hydrofluoric acid used to dissolve sample C104-OH-9 (ACL# 99-2348) caused the background measurement in the instrument to rise. As a result, the final concentrations in the check standards were typically 11% to 15% too high for many of the analytes measured in the quality control check standards and was particularly high for sodium which was about 47%. A longer clean-out time might have improved the measurement results. The concentration of sodium was very high in the last sample measured (> 500 µg/ml) and likely was the cause of the high residual sodium measured in the check standards. Quality control check standards MCVA and MCVB were analyzed immediately before the sample was measured. All measurements for the check standards at that time were within acceptable tolerance. The three dilutions performed on the sample were in good agreement with each other after adjusting for dilution and the two post-spike sample measurements that followed were also within tolerance limits. Therefore sample concentration results are not likely affected as might be indicated by the results of the final check standard measurements.

Five fold serial dilution:

(Solid samples) Results were generally within tolerance limit of ≤ 10% after correcting for dilution except as follows. Iron, chromium, lead and uranium were also somewhat high in KOH/Ni fusion samples (approximately 11% to 14%). The discrepancy in the KOH/Ni fusion samples may be related to the high aluminum, thorium and uranium concentrations. All three analytes cause interference to the analytes mentioned.

(Aqueous samples) All results were within tolerance limit of ≤ 10% after correcting for dilution except Silicon. Three of the aqueous samples measured approximately 11% to 17%. The discrepancy may be related to the very high concentration of sodium present in the samples.

Duplicate RPD (Relative Percent Difference):

(Solid samples) All analytes of interest were recovered within tolerance limit of ≤ 20% relative percent difference (RPD).

(Aqueous samples) All analytes of interest were recovered within tolerance limit of ≤ 20% relative percent difference (RPD).

Post-Spiked Samples (Group A):

(Solid samples) All analytes of interest were recovered within tolerance of 75% to 125%.

(Aqueous samples) All analytes of interest were recovered within tolerance of 75% to 125%.

Post-Spiked Samples (Group B):

(Solid samples) All analytes of interest were recovered within tolerance of 75% to 125%.

(Aqueous samples) All analytes of interest were recovered within tolerance of 75% to 125%.

Blank Spike:

(Solid samples) A blank spike is not require for fusion prepared samples.

(Aqueous samples) All analytes of interest in the blank spike were recovered within tolerance limit of 80% to 120% except Ag (34%) in sample C104-SOL-30-1 (ACL# 99-2340-BS). Chloride from the sample or from the hydrochloric acid used to prepare the sample using PNL-ALO-128 digestion procedure may have precipitated the silver resulting in low recovery.

Matrix Spiked Sample:

(Solid samples) A matrix spike is not require for fusion prepared samples.

(Aqueous samples) All analytes of interest in the matrix spiked sample C104-AQ-9 (ACL# 99-2347-MS) were recovered within tolerance limit of 75% to 125% except Ag (56%). Chloride from the sample or from the hydrochloric acid used to prepare the sample using PNL-ALO-128 digestion procedure may have precipitated the silver resulting in low recovery.

Quality Control Check Standards:

Concentration of all analytes of interest in the Na/Zr and KOH/Ni fusion prepared analytical runs were within tolerance limit of ± 10% accuracy in the standards: QC_MCVA, QC_MCVB, and QC_SSTMCV. Calibration Blank (ICP98.0) concentration was less than two times IDL

Concentration of all analytes of interest in the aqueous prepared analytical runs was within tolerance limit of ± 10% accuracy in the standards: QC_MCVA, QC_MCVB, and QC_SSTMCV except as follows. Sodium, nickel, and lead were high by 47%, 12% and 14% respectively in QC_MCVA check standard at the end of the run. Concentration of aluminum, sodium, lead and silicon were greater than 2 * IDL in the calibration blank ICP98.0 at the end of the run. Calcium, chromium, iron, manganese and sodium were also high by 11%, 12%, 33%, 15% and 14% respectively in QC_SSTMCV check standard at the end of the run. A suggested reason for the discrepancy is noted earlier (HF acid in the last sample analyzed).

High Calibration Standard Check:

Verification of the high-end calibration concentration for all analytes of interest in the three analytical measurement runs was within tolerance of ± 5% accuracy except for U in the KOH/Ni fusion prepared sample analytical run. Uranium was slightly below the minimum. It was low by 5.9%.

Process Blank:

(Solid samples) All analytes of interest were within tolerance limit of ≤ EQL or < 5% of sample concentration except Na (<7% of sample concentration) in PNL-ALO-115 KOH/Ni fusion prepared samples. Sodium is known to be present in the reagents used to prepare the samples.

No significant blank contribution found for PNL-ALO-114 Na/Zr fusion prepared samples.

(Aqueous samples) All analytes of interest were within tolerance limit of ≤ EQL or < 5% of sample concentration.

Laboratory Control Standard (LCS):

(Solid samples) All analytes of interest at a concentration equal to or greater than EQL were recovered within tolerance limit of 75% to 125% in both fusion prepared LCS standards. SRM-2710 Montana Soil was used for the LCS in both PNL-ALO-114 and PNL-ALO-115 fusion preparations.

(Aqueous samples) No LCS was prepared for PNL-ALO-128 acid digested samples.

Analytes other than those requested by the client are for information only. Please note bracketed values listed in the data report are within ten times instrument detection limit and have a potential uncertainty much greater than 15%.

Comments:

1) "Final Results" have been corrected for all laboratory dilution performed on the sample during processing and analysis unless specifically noted.

2) Detection limits (DL) shown are for acidified water. Detection limits for other matrices may be determined if requested.

3) Routine precision and bias is typically ± 15% or better for samples in dilute, acidified water (e.g. 2% v/v HNO_3 or less) at analyte concentrations greater than ten times detection limit up to the upper calibration level. This also presumes that the total dissolved solids concentration in the sample is less than 5000 µg/mL (0.5 per cent by weight).

4) Absolute precision, bias and detection limits may be determined on each sample if required by the client.

5) The maximum number of significant figures for all ICP measurements is 2.

Battelle PNNL/RPG/Inorganic Analysis ... ICPAES Data Report

	Multiplier=	2.1	41.3	19.9	14.4	9.7
	ALO#=	99-2340-B @1	99-2340 @1	99-2341 @1	99-2342 @1	99-2343 @1
	Client ID=	Process Blank	C104-SOL-30-1	C104-SOL-30-2	C104-SOL-40-1	C104-SOL-40-2
...t. Limit	Run Date=	8/27/99	8/27/99	8/27/99	8/27/99	8/27/99
(ug/mL)	(Analyte)	ug/g	ug/g	ug/g	ug/g	ug/g
0.015	Ag	--	--	--	[0.69]	[0.59]
0.060	Al	--	[14]	17.9	[3.4]	[4.4]
0.080	As	--	--	--	--	--
0.050	B	--	125	112	59.6	58.4
0.010	Ba	--	5.02	[1.1]	[0.34]	--
0.005	Be	--	--	--	--	--
0.100	Bi	[0.38]	--	--	--	--
0.250	Ca	--	[12]	74.4	43.5	50.6
0.015	Cd	--	6.36	5.01	3.16	3.09
0.100	Ce	--	--	--	--	--
0.025	Co	--	[2.0]	[1.6]	[1.3]	[1.3]
0.020	Cr	--	63.4	50.0	51.4	48.9
0.015	Cu	--	6.89	5.72	4.57	4.46
0.050	Dy	--	--	--	--	--
0.100	Eu	--	--	--	--	--
0.025	Fe	--	[4.2]	[3.1]	[2.5]	[1.9]
2.000	K	--	[560]	452	382	362
0.025	La	--	--	--	--	--
0.020	Li	--	27.1	23.6	20.4	19.1
0.100	Mg	--	--	29.2	16.4	19.6
0.005	Mn	--	--	--	--	[0.055]
0.030	Mo	--	[7.8]	6.63	5.40	5.26
0.100	Na	--	72,900	59,700	55,500	54,000
0.100	Nd	--	--	--	--	--
).030	Ni	--	126	99.6	79.3	75.4
).100	P	--	1,400	1,120	851	829
0.060	Pb	--	--	--	--	--
0.300	Pd	--	--	--	--	--
0.300	Rh	--	--	--	--	--
0.075	Ru	--	[5.2]	[4.3]	[3.6]	[3.4]
0.050	Sb	--	--	--	--	--
0.050	Se	--	--	--	--	[0.62]
0.100	Si	--	405	663	507	562
1.000	Sn	--	--	--	[27]	[25]
0.005	Sr	--	--	--	--	--
0.500	Te	--	--	--	--	--
0.800	Th	--	--	--	--	--
0.005	Ti	--	--	[0.22]	[0.18]	[0.19]
0.250	Tl	--	--	--	--	--
2.000	U	--	--	--	--	--
0.015	V	--	--	[0.40]	[0.49]	[0.44]
0.500	W	--	--	--	--	--
0.010	Y	--	--	--	--	--
0.020	Zn	[0.081]	12.1	8.01	3.45	3.31
0.025	Zr	--	--	--	--	--

Note: 1) Overall error greater than 10-times detection limit is estimated to be within +/- 15%.

2) Values in brackets [] are within 10-times detection limit with errors likely to exceed 15%.

3) "--" indicate measurement is below detection. Sample detection limit may be found by multiplying "det. limit" (far left column) by "multiplier" (top of each column).

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Det. Limit (ug/mL)	Multiplier= / Run Date= (Analyte)	16.1 / 99-2344 @2 / C104-SOL-50-1 / 8/27/99 ug/g	14.1 / 99-2345 @2 / C104-SOL-50-2 / 8/27/99 ug/g	3.5 / 99-2347 @2 / C104-AQ-9 / 8/27/99 ug/g	78.2 / 99-2348 @10 / C104-OH-3A / 8/27/99 ug/g	3.8 / 99-2350 @2 / C104-OH-9 / 8/27/99 ug/g
0.015	Ag	[1.2]	[1.1]	0.654	–	[0.47]
0.060	Al	11.1	[4.6]	126	45,900	2,080
0.080	As	–	–	–	[22]	[0.75]
0.050	B	53.4	58.5	3.72	[10]	3.85
0.010	Ba	[0.34]	–	[0.053]	–	[0.052]
0.005	Be	–	–	[0.021]	[3.9]	[0.057]
0.100	Bi	–	–	–	–	–
0.250	Ca	45.8	71.4	11.7	–	[5.0]
0.015	Cd	2.70	2.20	[0.52]	[5.8]	[0.087]
0.100	Ce	–	–	–	–	–
0.025	Co	[1.3]	[1.1]	[0.19]	–	–
0.020	Cr	63.1	49.9	12.5	157	19.1
0.015	Cu	4.41	3.48	0.638	[2.4]	[0.12]
0.050	Dy	–	–	–	–	–
0.100	Eu	–	–	–	–	–
0.025	Fe	[1.7]	[1.9]	[0.33]	[2.6]	[0.42]
2.000	K	371	300	[51]	[260]	[12]
0.025	La	–	–	–	–	–
0.020	Li	20.1	15.8	10.6	34.9	6.81
0.100	Mg	21.9	28.6	4.56	–	[1.9]
0.005	Mn	[0.099]	–	–	–	–
0.030	Mo	5.22	[4.1]	[0.72]	[4.1]	[0.27]
0.100	Na	52,100	44,900	10,800	153,000	13,700
0.100	Nd	–	–	–	–	–
0.030	Ni	75.3	59.6	9.79	[7.9]	[0.80]
0.100	P	784	624	107	814	33.0
0.060	Pb	–	–	–	[25]	[1.1]
0.300	Pd	–	–	–	–	–
0.300	Rh	–	–	–	–	–
0.075	Ru	[3.4]	[2.9]	[0.50]	–	–
0.050	Sb	–	–	–	[4.2]	–
0.050	Se	–	[0.72]	–	[14]	[0.66]
0.100	Si	506	678	112	[72]	67.0
1.000	Sn	[17]	–	–	[190]	[11]
0.005	Sr	–	–	–	–	–
0.500	Te	–	–	–	–	–
0.800	Th	–	–	–	–	–
0.005	Ti	[0.26]	[0.25]	[0.035]	–	[0.023]
0.250	Tl	–	–	–	[22]	–
2.000	U	–	–	[14]	–	–
0.015	V	[0.53]	[0.43]	[0.13]	[1.4]	[0.13]
0.500	W	–	–	–	–	–
0.010	Y	–	–	–	–	–
0.020	Zn	6.77	[2.2]	[0.56]	18.0	[0.53]
0.025	Zr	–	–	–	–	–

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Battelle PNNL/RPG/Inorganic Analysis ... ICPAES Data Report

	Multiplier=	3.6									
	ALO#=	99-2350-D @2									
	Client ID=	C104-OH-9									
Det. Limit	Run Date=	8/27/99									
(ug/mL)	(Analyte)	ug/g									
0.015	Ag	[0.46]		–		–		–		–	
0.060	Al	2,050		–		–		–		–	
0.080	As	[0.92]		–		–		–		–	
0.050	B	3.82		–		–		–		–	
0.010	Ba	[0.050]		–		–		–		–	
0.005	Be	[0.056]		–		–		–		–	
0.100	Bi	–		–		–		–		–	
0.250	Ca	[3.4]		–		–		–		–	
0.015	Cd	[0.085]		–		–		–		–	
0.100	Ce	–		–		–		–		–	
0.025	Co	–		–		–		–		–	
0.020	Cr	18.8		–		–		–		–	
0.015	Cu	[0.13]		–		–		–		–	
0.050	Dy	–		–		–		–		–	
0.100	Eu	–		–		–		–		–	
0.025	Fe	[0.28]		–		–		–		–	
2.000	K	[13]		–		–		–		–	
0.025	La	–		–		–		–		–	
0.020	Li	6.69		–		–		–		–	
0.100	Mg	[1.8]		–		–		–		–	
0.005	Mn	[0.018]		–		–		–		–	
0.030	Mo	[0.28]		–		–		–		–	
0.100	Na	13,400		–		–		–		–	
0.100	Nd	–		–		–		–		–	
0.030	Ni	[0.78]		–		–		–		–	
0.100	P	32.6		–		–		–		–	
0.060	Pb	[1.2]		–		–		–		–	
0.300	Pd	–		–		–		–		–	
0.300	Rh	–		–		–		–		–	
0.075	Ru	–		–		–		–		–	
0.050	Sb	[0.21]		–		–		–		–	
0.050	Se	[0.69]		–		–		–		–	
0.100	Si	63.6		–		–		–		–	
1.000	Sn	[9.4]		–		–		–		–	
0.005	Sr	–		–		–		–		–	
0.500	Te	–		–		–		–		–	
0.800	Th	–		–		–		–		–	
0.005	Ti	[0.030]		–		–		–		–	
0.250	Tl	[0.95]		–		–		–		–	
2.000	U	–		–		–		–		–	
0.015	V	[0.15]		–		–		–		–	
0.500	W	–		–		–		–		–	
0.010	Y	–		–		–		–		–	
0.020	Zn	[0.47]		–		–		–		–	
0.025	Zr	–		–		–		–		–	

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Det. Limit (ug/mL)	Multiplier= / ALO#= / Client ID= / Run Date= (Analyte)	1006.8 99-2346-PB-Zr @1 Process Blank 8/24/99 ug/g	967.7 99-2346-Zr @1 C104-AQ-8 8/24/99 ug/g	1034.0 99-2346-D-Zr @1 C104-AQ-8 8/24/99 ug/g	1028.1 99-2349-Zr @1 C104-OH-8 8/24/99 ug/g	1000.0 99-2349-D-Zr @1 C104-OH-8 8/24/99 ug/g
0.100	Ag	--	[630]	[720]	[790]	[770]
0.060	Al	[86]	163,000	152,000	36,100	35,700
0.250	As	--	--	--	--	--
0.050	B	--	--	--	--	--
0.010	Ba	--	183	184	359	357
0.075	Be	--	--	--	--	--
0.100	Bi	--	--	--	--	--
0.250	Ca	[1,800]	6,560	6,780	10,800	11,000
0.020	Cd	--	898	885	1,750	1,740
1.250	Ce	--	--	--	--	[1,300]
0.050	Co	--	--	--	[60]	[66]
0.050	Cr	--	1,460	1,470	1,980	2,000
0.035	Cu	--	[230]	[200]	445	458
0.050	Dy	--	--	--	[76]	[81]
0.100	Eu	--	--	--	--	--
0.025	Fe	376	44,900	45,700	85,300	84,800
2.000	K	[2,300]	--	--	--	[2,700]
0.075	La	--	--	[80]	[260]	[270]
0.050	Li	--	[320]	[320]	518	518
0.100	Mg	--	[960]	[970]	1,730	1,760
0.050	Mn	--	10,500	10,500	19,700	19,600
0.050	Mo	--	--	--	--	--
0.100	Nd	[100]	[150]	[210]	[570]	[610]
0.050	Ni	--	2,900	2,910	5,540	5,560
0.100	P	--	4,740	4,990	4,650	4,730
0.150	Pb	--	1,730	1,770	3,190	3,250
0.750	Pd	--	--	--	--	--
0.300	Rh	--	--	--	--	--
1.100	Ru	--	--	--	--	--
0.150	Sb	--	--	--	--	[170]
0.200	Se	--	--	--	--	--
0.500	Si	--	12,000	12,000	22,800	22,800
0.800	Sn	--	[1,700]	[1,700]	[2,300]	[2,400]
0.050	Sr	--	[120]	[110]	[220]	[220]
0.250	Te	--	--	--	[280]	[310]
1.000	Th	--	9,750	18,700	114,000	119,000
0.050	Ti	--	[160]	[180]	[430]	[430]
0.250	Tl	--	--	--	--	--
2.000	U	--	44,000	45,100	91,100	90,000
0.050	V	--	--	--	[86]	[92]
0.125	W	--	--	--	[240]	[260]
0.015	Y	--	[20]	[32]	[78]	[80]
0.050	Zn	--	[240]	[230]	[340]	[340]

Note: 1) Overall error greater than 10-times detection limit is estimated to be within +/- 15%.
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Battelle PNNL/RPG/Inorganic Analysis ... ICPAES Data Report

Det. Limit (ug/mL)	Multiplier= / ALO#= / Client ID= / Run Date= / (Analyte)	494.6 99-2346-PB-NI Process Blank 8/19/99 ug/g	1008.4 99-2346-NI @1 C104-AQ-8 8/19/99 ug/g	1001.5 99-2346-D-NI @1 C104-AQ-8 8/19/99 ug/g	494.6 99-2349-NI @1 C104-OH-8 8/19/99 ug/g	493.8 99-2349-D-NI @1 C104-OH-8 8/19/99 ug/g
0.025	Ag	–	990	984	1,810	1,770
0.060	Al	[69]	145,000	147,000	32,600	32,600
0.250	As	–	–	–	–	–
0.100	B	–	–	–	–	–
0.010	Ba	[6.4]	198	192	319	322
0.110	Be	–	–	–	–	–
0.250	Bi	–	–	–	–	–
0.250	Ca	–	4,740	4,790	7,260	7,090
0.015	Cd	–	922	872	1,590	1,560
0.275	Ce	–	–	–	[390]	[350]
0.125	Co	–	–	–	–	–
0.020	Cr	–	1,550	1,500	1,810	1,790
0.800	Cu	–	–	–	–	–
0.110	Dy	–	–	–	–	–
0.100	Eu	–	–	–	–	–
0.025	Fe	126	47,900	46,700	77,500	77,800
0.050	La	–	[130]	[130]	[210]	[210]
0.030	Li	–	304	[280]	435	441
3.500	Mg	–	–	–	–	–
0.050	Mn	[120]	11,100	10,800	17,900	17,900
0.080	Mo	–	–	–	–	–
0.150	Na	1,440	20,900	20,400	34,600	35,100
1.000	Nd	–	–	–	–	–
0.100	P	[70]	2,230	2,570	2,050	1,840
0.100	Pb	[62]	1,840	1,730	2,900	2,830
1.500	Pd	–	–	–	–	–
0.400	Rh	–	–	–	–	–
0.650	Ru	–	–	–	–	–
0.250	Sb	–	–	–	–	–
0.350	Se	–	–	–	–	–
0.500	Si	–	12,900	13,200	22,000	22,000
4.500	Sn	–	–	–	–	–
0.015	Sr	–	[88]	[87]	151	151
0.600	Te	–	–	–	–	–
1.000	Th	–	34,600	41,100	61,800	61,600
0.025	Ti	–	[200]	[210]	359	362
0.300	Tl	–	–	–	–	–
2.000	U	–	45,100	44,100	76,300	76,600
0.150	V	–	–	–	–	–
0.500	W	–	–	–	–	–
0.100	Y	–	–	–	–	–
0.050	Zn	–	[240]	[210]	322	316
0.050	Zr	–	51,300	50,500	101,000	104,000

Note: 1) Overall error greater than 10-times detection limit is estimated to be within +/- 15%.

2) Values in brackets [] are within 10-times detection limit with errors likely to exceed 15%.

3) "--" indicate measurement is below detection. Sample detection limit may be found by multiplying "det. limit" (far left column) by "multiplier" (top of each column).

Battelle
Pacific Northwest Laboratories

Project Number

Internal Distribution

329 File
LSO
Mike Urie

Date September 14, 1999

To Gregg Lumetta

From Tom Farmer

Subject ICPMS Analysis of submitted samples
(ALO# 99-2346 through 99-2350)

Pursuant to your request, the samples that you submitted for analysis were analyzed on our radioactively-contained ICPMS for the selected analytes; semiquantitative analysis was necessary on certain isotopes for which a standard was not available (see below). The concentration results for the isotopes of interest are displayed on the attached spreadsheets.

Dilutions of Isotope Products standards for ^{129}I, ^{233}U, ^{237}Np and ^{239}Pu, an Amersham ^{99}Tc standard and an NIST isotopic uranium standard (4321B) were used to generate the calibration curves. Independent standards, from the same vendors, of each analyte were used as the continuing calibration verification (CCV) standards. A spiked sample was also analyzed. The 1% high-purity nitric acid solution used to dilute the standards and samples was used as a reagent blank.

The ^{99}Tc values reported assume that the Ru present is exclusively fission-product Ru, and therefore does not have an isotope at m/z 99; i.e., everything observed at m/z 99 is due to ^{99}Tc. From the appearance of the Ru isotopic abundance, this appears to be a reasonable assumption; the fingerprint exhibited is obviously not natural. Approximate ^{101}Ru concentrations have been provided for your information.

Interference corrections were performed on the following isotopes: ^{129}I (xenon corrected), ^{239}Pu (Uranium hydride corrected). Printouts of the spreadsheet calculations for these corrections have been provided in the data package.

The results are reported in µg analyte /g (ppm) of original sample material for the fusion samples and ng analyte /ml (ppb) of submitted sample for the acid digestion samples. The overall uncertainty of the values is conservatively estimated at ±10%, and is based on the precision between consecutive analytical runs as well as the accuracy of the CCV standard results.

Values for the following isotopes were obtained using responses from related isotopes: ^{236}U (obtained from ^{238}U), and ^{240}Pu (obtained from ^{239}Pu). Because standards were not used and the concentrations of the isotopes were determined indirectly, these results should be considered semiquantitative. Printouts of the spreadsheet calculations are provided in the data package.

If you have any questions regarding this analysis, please give me a call at 372-0700 or James Bramson at 376-0624.

Greg Lumetta Analysis

September 14, 1999

James Bronson 9/20/99

Results are reported in ng analyte/ml of submitted sample.
The uncertainty of the results is estimated at ±10%.

Sample ID	Client ID	ICP/MS Number	Tc-99 ng/ml	•Ru-101 ng/ml
1%HNO3		9a08a1	<0.5	
1%HNO3		9a08a6	<0.5	
1%HNO3		9a08a14	<0.5	
99-2347	C104-AQ-9	9a08a11	83.6	203
99-2347 Dup.	C104-AQ-9	9a08a12	83.7	208
99-2348	C104-OH-3A	9a08a16	149	101
99-2348 Dup.	C104-OH-3A	9a08a17	153	109
99-2348 + spike	C104-OH-3A	9a08a18	236	
Spike Recovery			**87%**	
99-2350	C104-OH-9	9a08a7	48.0	88
99-2350 Dup.	C104-OH-9	9a08a8	46.5	84
2ppb Tc-99 CCV		9a08a4	2.05	
2ppb Tc-99 CCV		9a08a15	1.92	
2ppb Tc-99 CCV		9a08a19	2.00	

•Calculated using response for indium. For information only.

DATA REVIEW

Reviewed by: D.J. Jarmer II

Date: 20Sep99 Pages: 1 of 2

Greg Lumetta Analysis

September 14, 1999

James Bronson 9/20/99

Results are reported in µg analyte/g solid sample.
The uncertainty of the results is estimated at ±10%.

Sample ID	Client ID	ICP/MS Number	Tc-99 µg/g	*Ru-101 µg/g	I-129 µg/g	*U-233 µg/g	*†U-234 µg/g	*U-235 µg/g	*†U-236 µg/g	*U-238 µg/g	*Np-237 µg/g	*Pu-239 µg/g	*†Pu-240 µg/g
1%HNO3		9a09a1	<0.5		<2	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
1%HNO3		9a09a11	<0.5		<2	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
1%HNO3		9a09a20	<0.5		<2	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
99-2346-PB-Ni	Process Blank	9a09a12	<0.5		<2	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
99-2346-Ni	C104-AQ-8	9a09a13	2.17	24	3.81	53.8	2.4±0.7	411	18.9	56000	10±2	84.5	7.92
99-2346-DUP-Ni	C104-AQ-8	9a09a14	2.2±0.3	20	3.3±0.5	50.3	2.8±0.9	388	19.6	54000	8.68	82.6	8.19
99-2349-Ni	C104-OH-8	9a09a16	3.30	42	<2	109	6.1±1.5	698	34.1	97500	17±3	153	15.2
99-2349-DUP-Ni	C104-OH-8	9a09a17	3.7±1.2	43	<2	114	7.1±1.4	688	33.9	96100	18±2	157	14.2
99-2349-Ni + spike	C104-OH-8	9a09a18	56.4		47.7	143				129000	59.9	206	
Spike Recovery			**107%**		**116%**	**86%**				**106%**	**108%**	**107%**	
SRM 2710-Ni	LCS/99-2346-Ni	9a09a19	0.25±0.04	0.2	3.1±1.3	<0.5	<0.5	<0.5	<0.5	24.6	<0.5	0.6±0.2	<0.5
2ppb Tc-99 CCV		9a09a8	2.09										
2.5ppb Tc-99 CCV		9a09a21	2.66										
2.5ppb I-129 CCV		9a09a24			2.41								
5ppb I-129 CCV		9a09a4			4.72								
1ppb U-233, Np-237, Pu-239		9a10a6				1.02					0.936	0.990	
1ppb U-233, Np-237, Pu-239		9a09a24				0.980					1.00	0.905	
100ppb U		9a10a8								100			
100ppb U		9a10a23								109			

*Results are from procedure 9a10a.
†No standard availabe. Results calculated from response of different isotope.
•Calculated using response for indium. For information only.

DATA REVIEW
Reviewed by: C. J. Farmer III
Date: 20sep99 Pages: 1 of 2

J P Brown
9/29/99

Greg Lumetta Analysis

September 14, 1999(revised 9/29/99)

Results are reported in µCi analyte/ml of submitted sample.
The uncertainty of the results is estimated at ±10%.

Sample ID	Client ID	ICP/MS Number	Tc-99 µCi/ml	•Ru-101 ng/ml
1%HNO3		9a08a1	<1E-05	
1%HNO3		9a08a6	<1E-05	
1%HNO3		9a08a14	<1E-05	
99-2347	C104-AQ-9	9a08a11	0.00142	203
99-2347 Dup.	C104-AQ-9	9a08a12	0.00142	208
99-2348	C104-OH-3A	9a08a16	0.00253	101
99-2348 Dup.	C104-OH-3A	9a08a17	0.00259	109
99-2348 + spike	C104-OH-3A	9a08a18	0.00400	
Spike Recovery			**87%**	
99-2350	C104-OH-9	9a08a7	0.000814	88
99-2350 Dup.	C104-OH-9	9a08a8	0.000788	84
CCV results are reported in ng/ml (ppb).				
2ppb Tc-99 CCV		9a08a4		
2ppb Tc-99 CCV		9a08a15		
2ppb Tc-99 CCV		9a08a19		

•Calculated using response for indium. For information only.

DATA REVIEW
Reviewed by: D. Lumetta
Date: 29Sep99 Pages: 1 of 1

Gregg Lumetta Analysis

September 14, 1999(revised 9/29/99)

Results are reported in μCi analyte/g of submitted sample.
The uncertainty of the results is estimated at ±10%.

Sample ID	Client ID	ICP/MS Number	Tc-99 μCi/g	•Ru-101 ng/g
1%HNO3		9a08a1	<1E-05	
1%HNO3		9a08a6	<1E-05	
1%HNO3		9a08a14	<1E-05	
99-2347	C104-AQ-9	9a08a11	0.00138	198
99-2347 Dup.	C104-AQ-9	9a08a12	0.00139	203
99-2348	C104-OH-3A	9a08a16	0.00217	87
99-2348 Dup.	C104-OH-3A	9a08a17	0.00223	94
99-2348 + spike	C104-OH-3A	9a08a18	0.00343	
Spike Recovery			75%	
99-2350	C104-OH-9	9a08a7	0.000785	85
99-2350 Dup.	C104-OH-9	9a08a8	0.000770	82
CCV results are reported in ng/ml (ppb).				
2ppb Tc-99 CCV		9a08a4		
2ppb Tc-99 CCV		9a08a15		
2ppb Tc-99 CCV		9a08a19		

•Calculated using response for indium. For information only.

J C Brown
9/29/99

Greg Lumetta Analysis

September 14, 1999 (revised 9/29/99)

Results are reported in µCi analyte/g solid sample.
The uncertainty of the results is estimated at ±10%.

Sample ID	Client ID	ICP/MS Number	*U-235 µCi/g	*†U-236 µCi/g	*U-238 µCi/g	*Np-237 µCi/g	*Pu-239 µCi/g	*†Pu-240 µCi/g
1%HNO3		9a09a1	<0.000001	<0.00003	<2E-07	<0.0004	<0.03	<0.1
1%HNO3		9a09a11	<0.000001	<0.00003	<2E-07	<0.0004	<0.03	<0.1
1%HNO3		9a09a20	<0.000001	<0.00003	<2E-07	<0.0004	<0.03	<0.1
99-2346-PB-Ni	Process Blank	9a09a12	<0.000001	<0.00003	<2E-07	<0.0004	<0.03	<0.1
99-2346-Ni	C104-AQ-8	9a09a13	0.000888	0.00122	0.0188	0.0070±0.0014	5.24	1.80
99-2346-DUP-Ni	C104-AQ-8	9a09a14	0.000838	0.00127	0.0182	0.00612	5.12	1.86
99-2349-Ni	C104-OH-8	9a09a16	0.00151	0.00221	0.0328	0.012±0.002	9.49	3.45
99-2349-DUP-Ni	C104-OH-8	9a09a17	0.00149	0.00219	0.0323	0.013±0.002	9.74	3.22
99-2349-Ni + spike	C104-OH-8	9a09a18			0.0434	0.0422	12.8	
Spike Recovery					106%	108%		
SRM 2710-Ni	LCS/99-2346-Ni	9a09a19	<0.000001	<0.00003	<2E-07	<0.0004	0.037±0.012	<0.1
CCV results are reported in ng/ml (ppb)								
2ppb Tc-99 CCV		9a09a8						
2.5ppb Tc-99 CCV		9a09a21						
2.5ppb I-129 CCV		9a09a24						
5ppb I-129 CCV		9a09a4						
1ppb U-233, Np-237, Pu-239		9a10a6				0.936	0.990	
1ppb U-233, Np-237, Pu-239		9a09a24				1.00	0.905	
100ppb U		9a10a8			100			
100ppb U		9a10a23			109			

*Results are from procedure 9a10a.
†No standard availabe. Results calculated from response of different isotope.

DATA REVIEW

Reviewed by: O.J. Farmer

Date: 29 Sep 99 1 of 2

J Bowman 9/29/99

Greg Lumetta Analysis

September 14, 1999 (revised 9/29/99)

Results are reported in µCi analyte/g solid sample.
The uncertainty of the results is estimated at ±10%.

Sample ID	Client ID	ICP/MS Number	Tc-99 uCi/g	•Ru-101 µg/g	I-129 µCi/g	*U-233 µCi/g	*†U-234 µCi/g
1%HNO3		9a09a1	<0.01		<0.0003	<0.005	<0.003
1%HNO3		9a09a11	<0.01		<0.0003	<0.005	<0.003
1%HNO3		9a09a20	<0.01		<0.0003	<0.005	<0.003
99-2346-PB-Ni	Process Blank	9a09a12	<0.01		<0.0003	<0.005	<0.003
99-2346-Ni	C104-AQ-8	9a09a13	0.0368	24	0.000664	0.518	0.015±0.004
99-2346-DUP-Ni	C104-AQ-8	9a09a14	0.037±0.005	20	0.00058±0.00009	0.485	0.017±0.006
99-2349-Ni	C104-OH-8	9a09a16	0.0559	42	<0.0003	1.05	0.038±0.009
99-2349-DUP-Ni	C104-OH-8	9a09a17	0.063±0.020	43	<0.0003	1.10	0.044±0.009
99-2349-Ni + spike	C104-OH-8	9a09a18	0.956		0.00831	1.38	
Spike Recovery			107%		116%	86%	
SRM 2710-Ni	LCS/99-2346-Ni	9a09a19	<0.01	0.2	0.00054±0.00023	<0.005	<0.003
CCV results are reported in ng/ml (ppb)							
2ppb Tc-99 CCV		9a09a8	2.09				
2.5ppb Tc-99 CCV		9a09a21	2.66				
2.5ppb I-129 CCV		9a09a24			2.41		
5ppb I-129 CCV		9a09a4			4.72		
1ppb U-233, Np-237, Pu-239		9a10a6				1.02	
1ppb U-233, Np-237, Pu-239		9a09a24				0.980	
100ppb U		9a10a8					
100ppb U		9a10a23					

*Results are from procedure 9a10a.
†No standard availabe. Results calculated from response of different isotope.
•Calculated using response for indium. For information only.

DATA REVIEW
Reviewed by D.J. Lumetta
Date: 29Sep99 Pages: 2 of 2

Battelle Pacific Northwest Laboratory
Radiochemical Processing Group-325 Building
Radioanalytical Applications Team

99-2346
10/4/1999

Client : G. Lumetta

Cognizant Scientist: Sandra K Fiskum Date : 10/4/99

Concur : J R Greenwood Date : 10/4/99

Pu (PNL-ALO-417)
Total Alpha (PNL-ALO-420, PNL-ALO-421)
Sr-90 (PNL-ALO-476)

Measured Activities (µCi/g or /mL)
1-sigma propagated error

ALO ID Client ID	Total alpha Error %	Sr-90 Error %	Pu-238 Error %	Pu-239+240 Error %	Am-241 Error %	Cm242 Error %	Cm-243+244 Error %
99-2346-Ni C104-AQ-8	1.67E+1 3%	7.61E+2 3%	7.38E-1 6%	7.03E+0 4%	7.84E+0 4%	2.91E-2 (a) 24%	1.12E-1 13%
99-2346-Ni Rep C104-AQ-8		7.95E+2 3%	8.25E-1 6%	7.18E+0 4%	7.76E+0 4%	1.24E-2 38%	1.06E-1 13%
RPD		4% average 778	11% 0.782	2% 7.11	1% 7.80	80%	6%
99-2346-Ni duplicate C104-AQ-8	1.67E+1 3%	7.90E+2 3%	7.98E-1 7%	7.02E+0 4%	7.21E+0 4%	1.49E-2 36%	9.63E-2 14%
99-2346-Ni-PB Hot cell blank	5.67E-3 18%	2.56E-1 4%	2.49E-3 6%	5.65E-3 4%	8.27E-4 17%	< 4.E-5	7.71E-4 17%
99-2347 * C104-AQ-9	1.19E-4 13%	**2.98E-3** 12%					
99-2348 * C104-OH-3A	2.06E-4 8%	**8.20E-3** 5%					
99-2349 C104-OH-8	5.70E+1 3%	2.74E+3 3%	2.70E+0 7%	2.62E+1 4%	2.48E+1 4%	8.11E-2 23%	2.97E-1 13%
99-2349-Ni duplicate C104-OH-8	5.98E+1 3%	2.90E+3 3%	2.93E+0 4%	2.59E+1 3%	2.71E+1 5%	6.54E-2 38%	4.25E-1 15%
99-2350 * C104-OH-9	1.31E-4 11%	**1.14E-3** 31%					
99-2350 Replicate C104-OH-9	8.25E-5 15%						
RPD	45%						

(a) This value looks erroneously high compared to the replicate and duplicate.
M.J. Lumetta 10/18/99

Measured Activities (µCi/g or /mL)
1-sigma propagated error

ALO ID Client ID	Total alpha Error %	Sr-90 Error %	Pu-238 Error %	Pu-239+240 Error %	Am-241 Error %	Cm242 Error %	Cm-243+244 Error %
Reagent Blank	< 3. E-6	<1.E-5	< 2. E-5	3.91E-5 38%	4.43E-5 38%	<6.E-6	< 2.E-5
Reagent Spike	104%	101%		109%	95%		
Matrix Spike	76%	91%		113%	96%		

Note: 99-2346 and 99-2349 are reported as µCi/g, the other samples are reported as µCi/mL.

Battelle
Pacific Northwest Laboratories

Page _____ of _____

ENGINEERING WORKSHEET

Prepared By: M.J. Lumetta	Date: 10/1/99	Project:

Subject: C104 Test : Conversion of Solution ~~Lot~~ Radiation data to uCi/g

Density data for ASR 5478 - G. Lumetta

Acid Digestion in SRPL (lab 525)

Sample	Volume,ml	Wt, g.	Density,g/ml
99-2340	0.1	0.1201	1.201
99-2341	0.2	0.2316	1.158
99-2342	0.3	0.3403	1.134
99-2343	0.4	0.4533	1.133
99-2344	0.5	0.5653	1.131
99-2345	0.6	0.6618	1.103
99-2347	2.5	2.5586	1.023
99-2348	5	5.8278	1.166
99-2350	2.5	2.5903	1.036
99-2350Dup	2.5	2.5593	1.024

Obtained from Lewy Greenwood on 9/29/99
M.J. Lumetta

C104-AQ-9

Total α : $(1.19 \times 10^{-4}$ uCi/mL$) / (1.023$ g/mL$) = 1.16 \times 10^{-4}$ uCi/g

Sr-90 : ~~$(5.54 \times 10^{-3}$~~ $(2.98 \times 10^{-3}$ uCi/mL$) / (1.023$ g/mL$) =$ ~~5.54×10^{-3} uCi/g~~ 2.91×10^{-3} uCi/g *

Cs-137 : $(3.93$ uCi/mL$) / (1.023$ g/mL$) = 3.84$ uCi/g

Am-241(γ) : $(< 0.005$ uCi/mL$) / (1.023$ g/mL$) = < 0.005$ uCi/g

C109-OH-3A

Total α : $(2.06 \times 10^{-4}$ uCi/mL$) / (1.166$ g/mL$) = 1.77 \times 10^{-4}$ uCi/g

Sr-90 : ~~$(1.11 \times 10^{-2}$~~ $(8.20 \times 10^{-3}$ uCi/mL$) / (1.166$ g/mL$) =$ ~~1.11×10^{-2} uCi/g~~ 7.03×10^{-3} uCi/g *

Cs-137 : ~~$(2.93$~~ $(6.62$ uCi/mL$) / (1.166$ g/mL$) = 5.69$ uCi/g

Am-241(γ) : $(< 0.007$ uCi/mL$) / (1.166$ g/mL$) = < 0.006$ uCi/g

C104-OH-9

Total α : $[(1.31 \times 10^{-4} + 8.25 \times 10^{-5}) / 2]$ uCi/mL $/ (1.036$ g/mL$) = 1.03 \times 10^{-4}$ uCi/g

Sr-90 : ~~$(3.27 \times 10^{-3}$~~ $(1.14 \times 10^{-3}$ uCi/mL$) / (1.036$ g/mL$) =$ ~~2.15×10^{-3} uCi/g~~ 1.10×10^{-3} uCi/g *

Cs-137 : $(1.54 \times 10^{0}$ uCi/mL$) / (1.036$ g/mL$) = 1.49$ uCi/g

* Corrections made by M.J. Lumetta 10/4/99
based on updated information from
Sandy Fiskum

Battelle Pacific Northwest Laboratory
Radiochemical Processing Group-325 Building
Radioanalytical Applications Team

99-2346
9/21/99

Client : G. Lumetta

Cognizant Scientist: L R Greenwood Date : 9/23/99

Concur : T Trang-Le Date : 9/21/99

Gamma Energy Analysis (PNL-ALO-450)

Measured Activities (µCi/g)

ALO ID Client ID	Co-60 Error %	Nb-94 Error %	Sb-125 Error %	Cs-134 Error %	Cs-137 Error %	Eu-154 Error %	Eu-155 Error %	Am-241 Error %
99-2346-Ni PB Hotcell Blank	<5.E-3	<4.E-3	<2.E-2	1.13E-2 14%	1.74E-1 3%	<2.E-2	<2.E-2	<2.E-2
99-2346-Ni C104-AQ-8	2.72E-1 4%	1.32E-1 7%	2.48E-1 31%	<3.E-2	4.53E+1 2%	2.24E+0 2%	1.38E+0 6%	7.07E+0 10%
99-2346-Ni duplicate C104-AQ-8	2.68E-1 3%	1.01E-1 7%	2.70E-1 20%	<3.E-2	4.27E+1 2%	2.17E+0 2%	1.28E+0 6%	6.89E+0 10%
RPD	1%	27%	8%		6%	3%	8%	3%
99-2347* C104-AQ-9	4.26E-3 3%	<2.E-4	<8.E-3	<3.E-4	3.93E+0 2%	<3.E-4	<5.E-3	<5.E-3
99-2348* C104-OH-3A	4.44E-3 3%	<3.E-4	<1.E-2	<3.E-4	6.63E+0 2%	<4.E-4	<7.E-3	<7.E-3
99-2349 C104-OH-8	1.07E+0 2%	2.03E-1 15%	6.54E-1 17%	<5.E-2	1.36E+2 2%	6.20E+0 2%	3.96E+0 4%	2.57E+1 4%
99-2349-Ni duplicate C104-OH-8	1.05E+0 2%	2.71E-1 10%	7.73E-1 13%	<5.E-2	1.35E+2 2%	7.07E+0 2%	4.46E+0 4%	2.69E+1 4%
RPD	2%	29%	17%		1%	13%	12%	5%
99-2350* C104-OH-9	7.72E-4 5%	<7.E-5	<3.E-3	1.40E-4 25%	1.54E+0 2%	<2.E-4	<2.E-3	<2.E-3

Note: * samples are reported in uCi/ml.

Battelle PNNL/RPG/Inorganic Analysis --- IC Report

WO/Project: W48486(W51311)/29953
Client: G. Lumetta

ACL Numbers: 99-02346 through 99-02350
ASR Number 5478

Procedure: PNL-ALO-212, "Determination of Inorganic Anions by Ion Chromatography"
Analyst: MJ Steele **Analysis Date:** September 16-20, 1999

M&TE: IC system (WD25214); Mettler AT400 Balance (360-06-01-031) See Chemical Measurement Center 98620 RIDS for IC File for Calibration, Standards Preparations, and Maintenance Records.

Analyst: MJ Steele

Approval: Michael W Lee **Date** 10-25-99

Notes:

1) "Final Results" have been corrected for all dilution performed on the sample during processing or analysis.
2) The low calibration standards are defined as the estimated quantitation limit (EQL) for the reported results and assume non-complex aqueous matrices. Actual detection limits or quantitation limits for specific sample matrices may be determined, if requested.
3) Routine precision and bias is typically ± 15% or better for non-complex aqueous samples that are free of interference and have similar concentrations as the measured anions.

Final Results:

The samples were analyzed by ion chromatography (IC) for inorganic anions as specified in ASR 5478. The liquid samples were diluted at the IC workstation up to 2000-fold to ensure that all anions were within the calibration range, and the solids samples were diluted an additional 10-fold following leaching per procedure ALO-103. The anion results are presented in the table below.

Lab ID	Solid Sample ID	Solids Dil Fctr	F ug/g	Cl ug/g	NO_2 ug/g	NO_3 ug/g	PO_4 ug/g	SO_4 ug/g
99-2346 PB	Solids Process Blank	46.5	< 12	< 12	< 24	< 24	< 24	50
99-2346	C104-AQ-8	45.4	2,900*	120	320	1,500	640	570
99-2346 Dup	C104-AQ-8 Dup	48.7	2,600*	< 130	< 250	1,300	620	< 250
	RPD(%)		11%	n/a	n/a	14%	3%	n/a
99-2349	C104-OH-8	46.2	2,900*	150	< 240	1,200	< 240	< 240
99-2349 Dup	C104-OH-8 Dup	45.8	2,800*	170	380	1,300	< 230	< 230
	RPD(%)		4%	13%	n/a	8%	n/a	n/a
Lab ID	Liquid Sample ID	Liquid Dil Fctr	F ug/ml	Cl ug/ml	NO_2 ug/ml	NO_3 ug/ml	PO_4 ug/ml	SO_4 ug/ml
99-2347	C104-AQ-9	1	5,300*	150	2,600	1,500	< 250	400
99-2347 Rep	C104-AQ-9 Rep	1	4,700*	< 130	2,600	1,400	< 250	400
	RPD(%)		12%	n/a	0%	7%	n/a	0%
99-2348	C104-OH-3A	1	5,500*	360	3,600	1,700	1,000	500
99-2350	C104-OH-9	1	6,600*	< 130	500	400	< 250	< 250

RPD = Relative Percent Difference (between sample and duplicate/replicate)

* = Quantified by IC system as fluoride; however, slight retention time peak shift and peak shape suggest significant organic anion interference. High probability that little or no fluoride is actually present in the samples.

Q.C. Comments:

Following are results of quality control checks performed during IC analyses. In general, quality control checks met the requirements of the governing QA Plan.

Working Blank Spike/Process Blank Spike: Process Blank Spike recoveries ranged from 99% to 113%, well within the acceptance criteria of 80% to 120%.

Matrix Spiked Sample: No matrix spike was performed on the samples submitted under this ASR. However, matrix spikes processed and analyzed with this batch of sample had recoveries within the acceptance criteria of 75% to 125%, except for nitrate which produced over range condition due to the high nitrate concentrations in the samples.

Duplicate: Except for one oxalate duplicate which demonstrated an RPR of 22%, the RPDs which ranged from 3% to 14% which is within the acceptance criteria of 20%.

System Blank/Processing Blanks: Over 20 system blanks were process during the analysis of the sample. With the exception of only 3 nitrate values and 1 sulfate value, no anions were detected above reportable concentrations in the system blanks or in the processing/dilution blank.

Quality Control Calibration Verification Check Standards: Over 20 mid-range verification standards were analyzed throughout the analysis run. Except for a few case the reported results for all analytes of interest were recovered within the acceptance criteria of ±10% for the verification standard. For the few failures, no recoveries exceeded ±15%.

Battelle PNNL/RPG/Inorganic Analysis ---Hg Report

WO/Project: W48486/29953
Client: G. Lumetta

ACL Numbers: 99-02346 and 99-02350
ASR Number 5478.01

Procedure: PNNL-ALO-131, "Mercury Digestion"
PNNL-ALO-201, "Mercury Analysis"

Analyst: J. J. Wagner

Digestion Date: October 21, 1999 **Analysis Date:** October 27, 1999

M&TE: Hg system (WD14126); Mettler AT400 Balance (360-06-01-029) See Chemical Measurement Center 98620 RIDS for Hg File for Calibration, Standards Preparations, and Maintenance Records.

Analyst: Jerry Wagner

Approval: _____ **Date** 11-5-99

Final Results:

The samples were analyzed by cold vapor atomic absorption spectrophotometry for inorganic mercury as specified in ASR 5478.01. The solids samples were diluted an additional 250 to 500-fold following sample digestion per procedure ALO-131. The mercury concentration results are presented in the table below.

Battelle PNNL/RPG/Inorganic Analysis ---Hg Report

Lab ID	Solid Sample ID	Solids Grams	Solids Dig Fctr	Solids Anal Fctr	Hg ug/g
99-2346 PB	Solids Process Blank	0.2084	120.0	1	<0.024
99-2346	C104-AQ-8	0.2102	118.9	250	95.4
99-2346 Dup	C104-AQ-8 Dup	0.2138	116.9	250	96.8
	RPD(%)				1.5%
99-2349	C104-OH-8	0.2077	120.4	500	153
99-2349 Dup	C104-OH-8 Dup	0.2018	123.9	500	164
	RPD(%)				6.9%

RPD = Relative Percent Difference (between sample and duplicate/replicate)
"Sample weight" used for the process blank is an average weight of the samples.

Notes:

1) "Final Results" have been corrected for all dilution performed on the sample during processing or analysis.
2) The low calibration standard is defined as the estimated quantitation limit (EQL) for the reported results and assumes non-complex aqueous matrices. Actual detection limits or quantitation limits for specific sample matrices may be determined, if requested.
3) Routine precision and bias is typically ± 15% or better for non-complex aqueous samples that are free of interference.

Q.C. Comments:

Following are results of quality control checks performed during Hg analyses. In general, quality control checks met the requirements of the governing QA Plan.

Working Blank Spike/Process Blank Spike: Process Blank Spike recovery is 112%, well within the acceptance criteria of 80% to 120%.

Matrix Spiked Sample: A matrix spike was prepared for the samples submitted under this ASR. However, the concentration of the matrix spike processed and analyzed with this batch of sample was too low in concentration relative to the high concentration of mercury in the samples measured. As a result, matrix spike recovery could not be assessed.

Duplicate: All RPDs were within the acceptance criteria of 20%.

System Blank/Processing Blanks: A system blank was process during the analysis of the sample. All reportable sample concentrations were many times greater than that measured in the system blank or in the processing/dilution blank.

Quality Control Calibration Verification Check Standards: Over 4 mid-range verification standards were analyzed throughout the analysis run. All were within the acceptance criteria of 80% to 120% recovery for the verification standard.

Battelle, Pacific Northwest National Laboratory
Richland, WA
Radiochemical Processing Group

filename 99-2346
11/15/99

Client: Lumetta

Cognizant Scientist: T D Freeman 11-15-99

Concur: C. Soderquist 11-15-99

Procedure: PNL-ALO-4014

Uranium Analysis by Kinetic Phosphorescence

Sample	Lab Number	Units	Uranium Concentration,	± 1s
Process Blk	99-2346PB NI	**µg/g**	**1.60E+0**	**± 2%**
C104-AQ-8	99-2346Ni	**µg/g**	**2.61E+4**	**± 4%**
C104-AQ-8	99-2346Ni DUP	**µg/g**	**2.50E+4**	**± 4%**
	RPD		**4%**	
C104-AQ-9	99-2347	**µg/mL**	**1.47E+1**	**± 4%**
C104-OH-3A	99-2348	**µg/mL**	**1.05E+1**	**± 4%**
C104-OH-3A	99-2348-Rep	**µg/mL**	**1.08E+1**	**± 3%**
C104-OH-3A	99-2348-Rep	**µg/mL**	**1.13E+1**	**± 3%**
C104-OH-8	99-2349	**µg/g**	**1.01E+5**	**± 4%**
C104-OH-8	99-2349 DUP NI	**µg/g**	**9.92E+4**	**± 4%**
	RPD		**2%**	
C104-OH-9	99-2350	**µg/mL**	**3.42E+0**	**± 4%**

	Standard	Observed	Expected	Yield
Before Run	Rec-282-e2	1.05E-2	1.00E-2	**1.050**
	Rec-282-d2	1.06E-1	1.00E-1	**1.060**
	Blank	<2.E-5		
After Run	Rec-282-e2	1.07E-2	1.00E-2	**1.070**
	Rec-282-d2	1.06E-1	1.00E-1	**1.060**
	Blank	<2.E-5		

Battelle PNNL/RPG/Inorganic Analysis --- TOC/TIC Report

Client: G. Lumetta
ACL Numbers: 99-2346 to 99-2350
Analyst: MJ Steele
Charge Code/Project: W48486/ 29953
ASR Number: 5478
Analysis Date: 12/09/99 and 01/07/00

Procedure: PNL-ALO-381, "Direct Determination of TC, TOC, and TIC in Radioactive Sludges and Liquids by Hot Persulfate Method"
M&TE: Carbon System (WA92040); Balance (360-06-01-023).

Final Results:

Liquids		TIC	TIC RPD	TOC	TOC RPD	TC	TC RPD
Lab Number	Sample ID	ug C/ml	(%)	ug C/ml	(%)	ug C/ml	(%)
00-2347	C104-AQ-9	610		790		1,400	
99-2347 Rep	C104-AQ-9 Rep	750	21	760	4	1,500	7
99-2348	C104-OH-3A	1,200		1,400		2,600	
99-2348 Rep	C104-OH-3A Rep	1,200	0	1,200	15	2,400	8
99-2350	C104-OH-9	270		210		480	
99-2350 Rep	C104-OH-9 Rep	280	4	220	5	500	4
99-2350 MS	C104-OH-9 MS Rec.	101%		97%		100%	
Results on a per gram dry weight basis							
Solids		TIC	TIC RPD	TOC	TOC RPD	TC	TC RPD
Lab Number	Sample ID	ugC/g	(%)	ugC/g	(%)	ugC/g	(%)
99-2346	C104-AQ-8	2,560		9,900		12,460	
99-2346 Dup	C104-AQ-8 Dup	2,380	7	10,600	7	12,980	4
99-2346 MS	C014-AQ-8 MS	95%		87%		92%	
99-2349	C104-OH-8	6,900		17,000		23,900	
99-2349 Dup	C104-OH-8 Dup	6,840	1	16,900	0	23,740	1

RPD = Relative Percent Difference (between sample and duplicate/replicate)

The analysis of the subject samples submitted under ASR 5478 was performed by the hot persulfate wet oxidation method. The hot persulfate method uses acid decomposition for TIC and acidic potassium persulfate oxidation at 92-95°C for TOC, all on the same sample, with TC being the sum of the TIC and TOC.

The table above shows the results, rounded to two to three significant figures. The raw data bench sheets and calculation work sheets showing all calculations are attached. All sample results are corrected for average percent recovery of system calibration standards and are also corrected for contribution from the instrument calibration blanks.

Q.C. Comments:

The TIC standard is calcium carbonate and TOC standard is α-Glucose (the certificates of purity are attached). The standard materials were used in solid form for system calibration standards as well as matrix spikes. TIC and TOC percent recovery are determined using the appropriate standard (i.e., calcium carbonate for TIC or glucose for TOC).

The QC for the methods involves calibration blanks, system calibration standards, sample duplicates, and one matrix spike per matrix type.

Calibration Standards: The QC system calibration standards for the 12/09/99 and 01/07/00 analysis runs were all within acceptance criteria, with the average recoveries being 100.2% and 98.4% for TIC and 99.7% and 98.1% for TOC, respectively.

Calibration Blanks: The six calibration blanks run at the beginning and end of the analysis runs were acceptable. The standard deviation calculated from the calibration blanks is less than the estimated method detection limit for both TIC and TOC.

Duplicates: The relative percent differences (RPD) between duplicates are within the acceptance criteria of 20%, except for the TIC for liquid sample 99-2347 (C104-AQ-9). At an RPD of 21%, this sample is only slightly outside the acceptance criteria, and the poor RPD is most likely due to the small sample size (i.e., 0.2 ml) used for the sample. The duplicate was analyzed using a 0.9 ml sample size and is considered to provide the most accurate results.

Matrix Spike: The accuracy of the carbon measurements can be estimated by the recovery results from the matrix spike. The matrix spike for liquid sample run (i.e., 99-2350, C104-OH-9 MS) recovered at 101% for TIC and 97% for TIC, well within the 75% to 125% recovery acceptance criteria. The matrix spike for the solids sample run (i.e., 99-2346, C104-AQ-8 MS) recovered at 95% for TIC and 87% for TOC.

General Comments:

- The reported "Final Results" have been corrected for all dilution performed on the sample during processing or analysis.
- Routine precision and bias are typically ±15% or better for non-complex samples that are free of interferences.
- The estimated quantitation limit (EQL) is defined as 5 times the MDL. Results less than 5 times the MDL have higher uncertainties, and RPDs are not calculated for any results less than 5 times the MDL.
- Some results may be reported as less than ("<") values. These less than values represent the sample MDL (method detection limit), which is the system MDL adjusted for the volume of sample used for the analysis. The system MDL is based on the attached pooled historical blank data. The evaluation and calculation of the system MDL is included in the data package.

Report Prepared by: _____ **Date** 1-17-00

Review/Approval by: _____ **Date** 1/20/00

Archive Information:

Files: ASR 5478 Lumetta.doc	ASR 5478 5536 5571 Liq+Solids.xls
	ASR 5478 5626 Liq+Solids.xls

Battelle
Putting Technology To Work

Project No. 29953

Internal Distribution

Date October 28, 1999

To G. Lumetta

From M. Urie

Subject Cyanide Results for Samples C104-AQ-8 and C104-OH-8

ALO#	Client ID	Sample (µg CN / g)	Duplicate (µg CN / g)	RPD (%)	Spike Rec (%)
99-2346	C104-AQ-8	13.4	12.1	10	
99-2349	C104-OH-8	22.0	24.7	12	
99-2346 spike	C104-AQ-8 spike	---	---	---	93

The CN results for two C104 tank samples analyzed on September 2, 1999 per ASR 5478 are reported in the table above. The sample aliquots were weighed in the Shielded Analytical Laboratory and delivered, ready for distillation, to Laboratory 400 in the Radiochemical Processing Laboratory. The samples were distilled with the addition of sulfamic acid to ensure there would be no interference if nitrates were present in the sample. The samples were analyzed using a Lachat QuickChem AE Autoanalyzer (WC36517). The reporting limit is estimated to be 0.2 mg/kg.

An independent calibration check solution run at the beginning and end of each analysis batch gave an average recovery of 110%. Both samples were prepared and analyzed in duplicate. In addition, a matrix spike sample of C106-OH-8 was prepared. The spike recovery at 93% was within the control limits (±15%). The solid laboratory control standard (ERA-LSC) analyzed at 221 ug/g, well within the certified advisory range of 77 to 301 ug/g.

The relative percent difference (RPD) between the samples and duplicated for both samples was well within the acceptance criteria of 20%. The initial analysis of the samples was performed with no analytical dilutions, which resulted in the absorbences being above the highest calibration standard. The samples were diluted 3-fold and reanalyzed. Although the initial analyses were over range, the results correspond very well with the final results.

All sample preparation sheets, standard preparation information, and analytical data are included with this report.

C Soderquist
Concur

1-20-00
Date

`Memo File: CN ASR 5478 Lumetta.doc Spreadsheet File: CN ASR 5478 Lumetta.xls

CYANIDE ANALYSIS REPORT

File: 99090201.RS
Procedure: PNL-ALO-287 & -289
M&TE: WC36517, Latchat QuickChem AE Autoanalyzer
WD04501, Mettler AT400 Balance

Client: Lumetta/ ASR #5478

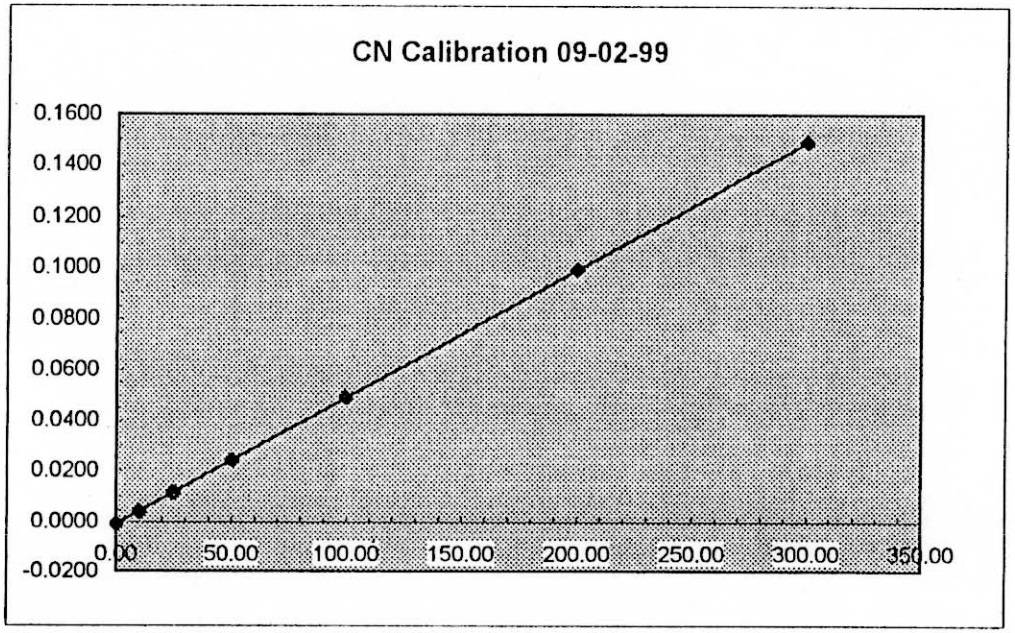
Analyst: PK Berry
Calibration File: 99090201
Date: September 2, 1999

Lab ID	Client ID	Sample Mass (g)	% Solids	Hot-Cell Dilution Factor	Sample Volume mL	Final Digest Volume mL	Additional Dilut. Factor	Absorbance	ppb CN in Aliquot	CN in Sample		Expected Value	Dupl & or Rep RPD	% Rec
Mid-ICV, 125ppb					6	6	1	0.0659	133.63	133.63	ug/L	124.87		107%
Low-ICV, 30ppb					6	6	1	0.0153	32.31	32.31	ug/L	30.01		108%
ICB					6	6	1	-0.0004	0.87	< 2.0	ug/L			
BL-2346					6	6	1	-0.0003	1.07	< 2.0	ug/L			
BS-2346					6	6	1	0.0842	170.28	170.28	ug/L	166.167		102%
99-2346	C104-AQ-8	0.2164	100		6	6	1	0.2360	474.24	13.15	mg/kg			
99-2346-DUP	C104-AQ-8	0.2915	100		6	6	1	0.2880	578.36	11.90	mg/kg		10%	
99-2346-MS	C104-AQ-8	0.2202	100		6	6	1	0.2988	599.99	16.35	mg/kg	648.73		92%
99-2349	C104-OH-8	0.2088	100		6	6	1	0.4081	818.85	23.53	mg/kg			
99-2349-DUP	C104-OH-8	0.1904	100		6	6	1	0.4059	814.44	25.67	mg/kg		9%	
LCSS		0.051	100		6	6	10	0.0931	188.10	221.29	mg/kg	177.00		125%
Distilled Cal Std					6	6	1	0.1037	209.32	209.32	ug/L	200.17		105%
Mid-CCV, 125ppb					6	6	1	0.0693	140.44	140.44	ug/L	124.87		112%
Low-CCV, 30ppb					6	6	1	0.0162	34.11	34.11	ug/L	30.01		114%
CCB					6	6	1	-0.0004	0.87	< 2.0	ug/L			
5ppb, Quant chk					6	6	1	0.0021	5.88	5.88	ug/L	5.01		117%

LCSS sample is an ERA Certified Reference Material cyanide standard in soil; Lot # 218,Barcode #95760, Exp. Date 10-17-99.
Blank Spike was a 0.1 aliquot of a 9.97ppm CPI Standard Stock Dilution #2 (prep date 6/99) added to 6mL 0.25M NaOH, analyzed distilled.
MS was a 0.1 aliquot of a 9.97ppm CPI Standard Stock Dilution #2 (prep date 6/99) added to solid sample matrix, plus 5.5 mL water. Analyzed distilled.
Non-homogeneous sample matrix resulted in differences in duplicate results.

Calibration Solution: CPI prepared solutions
Expiration: 06/18/99

1.0000 Correlation Coefficient
0.000499403 SLOPE
-0.000835843 INTERCEPT



Calibration Data

true	ppb CN	Abs	Calc Stds	% rec for std
0	0.00	-0.0007	0.27	NA
10	10.02	0.0041	9.88	99%
25	25.02	0.0116	24.90	100%
50	50.05	0.0241	49.93	100%
100	99.76	0.0490	99.79	100%
200	200.17	0.0992	200.31	100%
300	300.10	0.1490	300.03	100%

Definitions

99-xxxx = sample
99-xxxxD = duplicate
99-xxxxS = spiked sample
LCSS = laboratory control standard, solid
LCDW = laboratory control standard, liquid
PBW = process water blank
BS = spiked process water blank
PBS = process soil blank
ICV = initial calibration verification
CCV = continuing calibration verification
ICB = initial calibration blank
CCB = continuing calibration blank
CRDL= contract required detection limit
IDL = instrument detection limit

CYANIDE ANALYSIS REPORT

File: 99090202.RS
Procedure: PNL-ALO-287 & -289
M&TE: WC36517, Latchat QuickChem AE Autoanalyzer
WD04501, Mettler AT400 Balance

Client: Lumetta/ ASR #5478

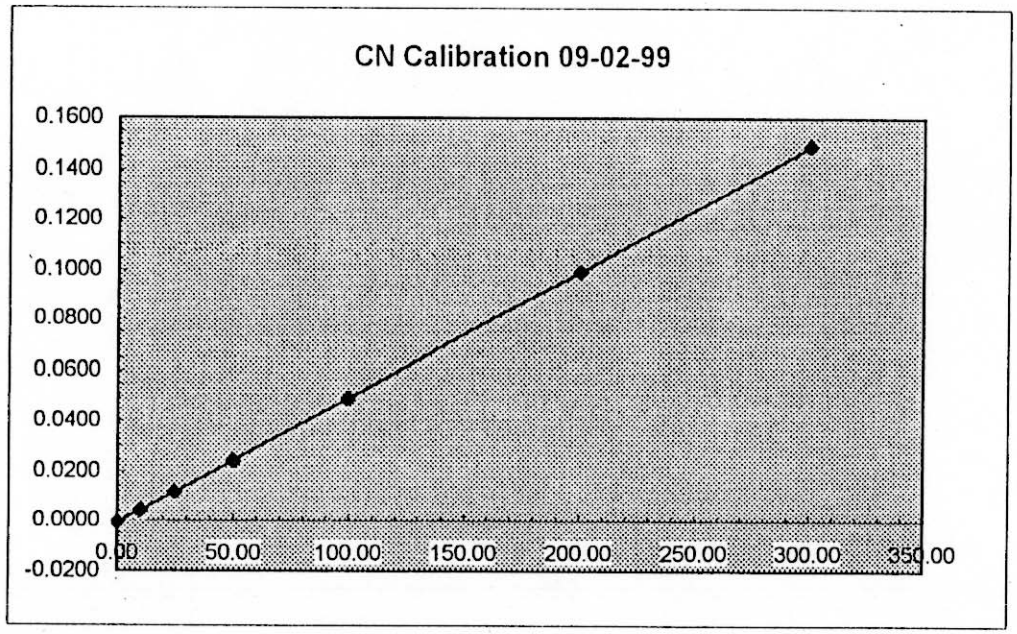
Analyst: PK Berry
Calibration File: 99090201
Date: September 2, 1999

Lab ID	Client ID	Sample Mass (g)	% Solids	Hot-Cell Dilution Factor	Sample Volume mL	Final Digest Volume mL	Additional Dilut Factor	Absorbance	ppb CN in Aliquot	CN in Sample		Expected Value	Dupl.& or Rep RPD	% Rec
Mid-ICV, 125ppb					6	6	1	0.0656	133.03	133.03	ug/L	124.87		107%
Low-ICV, 30ppb					6	6	1	0.0155	32.71	32.71	ug/L	30.01		109%
ICB					6	6	1	-0.0001	1.47	< 2.0	ug/L			
99-2346	C104-AQ-8	0.2164	100		6	6	3	0.0793	160.46	13.35	mg/kg			
99-2346-DUP	C104-AQ-8	0.2915	100		6	6	3	0.0968	195.51	12.07	mg/kg		10%	
99-2346-MS	C104-AQ-8	0.2202	100		6	6	3	0.1004	202.71	16.57	mg/kg	218.67		93%
99-2349	C104-OH-8	0.2088	100		6	6	3	0.1264	254.78	21.96	mg/kg			
99-2349-DUP	C104-OH-8	0.1904	100		6	6	3	0.1296	261.18	24.69	mg/kg		12%	
Mid-CCV, 125ppb					6	6	1	0.0689	139.64	139.64	ug/L	124.87		112%
Low-CCV, 30ppb					6	6	1	0.0161	33.91	33.91	ug/L	30.01		113%
CCB					6	6	1	-0.0002	1.27	< 2.0	ug/L	5.01		
5ppb, Quant chk					6	6	1	0.0020	5.68	5.68	ug/L	5.01		113%

LCSS sample is an ERA Certified Reference Material cyanide standard in soil; Lot # 218,Barcode #95760, Exp. Date 10-17-99.
Blank Spike was a 0.1 aliquot of a 9.97ppm CPI Standard Stock Dilution #2 (prep date 6/99) added to 6mL 0.25M NaOH, analyzed distilled.
MS was a 0.1 aliquot of a 9.97ppm CPI Standard Stock Dilution #2 (prep date 6/99) added to solid sample matrix, plus 5.5 mL water. Analyzed distilled.
Non-homogeneous sample matrix resulted in differences in duplicate results.

Calibration Solution: CPI prepared solutions
Expiration: 06/18/99

1.0000 Correlation Coefficient
0.000499403 SLOPE
-0.000835843 INTERCEPT



Calibration Data

true	ppb CN	Abs	Calc Stds	% rec for std
0	0.00	-0.0007	0.27	NA
10	10.02	0.0041	9.88	99%
25	25.02	0.0116	24.90	100%
50	50.05	0.0241	49.93	100%
100	99.76	0.0490	99.79	100%
200	200.17	0.0992	200.31	100%
300	300.10	0.1490	300.03	100%

Definitions

99-xxxx = sample
99-xxxxD = duplicate
99-xxxxS = spiked sample
LCSS = laboratory control standard, solid
LCDW = laboratory control standard, liquid
PBW = process water blank
BS = spiked process water blank

PBS = process soil blank
ICV = initial calibration verification
CCV = continuing calibration verification
ICB = initial calibration blank
CCB = continuing calibration blank
CRDL= contract required detection limit
IDL = instrument detection limit

Battelle, Pacific Northwest National Laboratory
Richland, WA
Radiochemical Processing Group

filename 99-2346
2/24/00

Client: Lumetta

Cognizant Scientist: C. Soderquist 2-24-00

Concur: J.R. Greenwood 2-25-00

Sample	Lab Number	Ammonia Concentration, ug/g	± 1σ
Process Blk	99-2346 PB	**<8.0**	
C104-AQ-8	99-2346	**<8.4**	
C104-AQ-8	99-2346 DUP	**<9.0**	
C104-OH-8	99-2349	**<8.5**	
C104-OH-8	99-2349 DUP	**<8.5**	

Battelle, Pacific Northwest National Laboratory
Richland, WA
Radiochemical Processing Group

filename 99-2346
3/13/2000

Client: Lumetta

Cognizant Scientist: M. K. Fisher 3/13/00

Concur: L D Greenwood 3-13-00

PNL-ALO-482

Sample	Lab Number	C-14 uCi/g	± 1σ	MDA* uCi/g
C104-AQ-8	99-2346	**6.28E-3**	**± 4%**	**<7.E-3**
C104-AQ-8	99-2346 DUP	**2.87E-3**	**± 5%**	**<4.E-3**
C104-OH-8	99-2349	**3.85E-3**	**± 5%**	**<5.E-3**
C104-OH-8	99-2349 DUP	**2.46E-3**	**± 5%**	**<3.E-3**
	Blank 1	**<3.E-4**		
	Blank 2	**1.58E-2**	**± 3%**	
	Blank 3	**6.32E-3**	**± 4%**	
	Sample spike	**87%**		
	Blank spike	**94%**		

*Note: Two of the blanks (2 and 3) analyzed with the samples had more activity than the samples indicating sample contamination as a result of carryover of the C-14 spike run just previous to these blanks. This tailing or memory effect appears to be variable and undefined at this time. Samples were run after a blank was run and thus the carryover of the spike is less severe than that shown for the blank. It is recommended that the listed MDA values be reported. These MDA's are calculated as a function of the obtained sample activity (found activity + 3 times the 1-s error).

Appendix C. Calculations

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ENGINEERING WORKSHEET

Prepared By: M.J. Smith | Date: 8/13/99 | Project:

Title/Subject: C104 Test Data Wrap-Up

Sample Weights

Vial Tare wt.	Initial Gross wt.	Initial Sample wt.	8/12/99 Gross wt.	8/12/99 Sample wt.	Sample ID
6.5218	7.4072	0.8854	6.9222	0.4004	C104-SOL-30-1
6.4870	7.3496	0.8626	6.9279	0.4409	-2
6.5451	7.4937	0.9486	7.1768	0.6317	-40-1
6.4873	7.5410	1.0537	7.1976	0.7103	-2
6.4773	7.6962	1.2189	7.2932	0.8159	-50-1
6.5265	7.8534	1.3269	7.5184	0.9919	-2
8.1397	18.3751	10.2354	18.3523	10.2126	C104-AD-9
8.4965	26.0674	17.5709	26.0570	17.5605	C104-DH-34
8.3703	18.3845	10.0142	18.3618	9.9915	C104-DH-9

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ENGINEERING WORKSHEET

Prepared By: M.J. Lumetta	Date: 8/3/99	Project:

Title/Subject: C-104 Test: Determination of "Water-Insoluble" Solids

Dean Kurath wanted to know the amount of water-insoluble solids present in the C-104 sludge sample. We should be able to get this information from Part 2 of this test.

The original sample provided (C-104 GL) was reported to contain ~~165.6~~ 149.5 g of material. 8/3/99 M.J.L.

We added 20 mL of 0.1 M NaOH to mobilize the material.

(20 mL)(1.005 g/mL) = 20.1 g 0.1 M NaOH added.
↳ ρ of 0.1 M NaOH

So the weight of the slurry = ~~165.6~~ 149.5 + 20.1 = ~~185.7~~ 169.6 g M.J.L. 8/3/99

50.8765 g of this slurry was used in the washing test (2.8A)
This yielded 14.3589 g of dried washed solids (2.69A)

$$(50.8765\text{ g slurry})\left(\frac{\overset{149.5}{\cancel{165.6}}\text{ g C104 sludge}}{\underset{169.6}{\cancel{185.7}}\text{ g slurry}}\right) = \overset{49.8}{\cancel{45.4}}\text{ g C104 sludge used in washing test}$$ M.J.L. 8/3/99

$$\therefore\quad 100\,\frac{14.4\text{ g dried solids}}{\underset{49.8}{\cancel{45.4}}\text{ g C104 sludge}} = \overset{32.1}{\cancel{31.7}}\text{ wt \% "insoluble" solids in the C-104 sludge}$$ M.J.L. 8/3/99

9/24/99 M.J.L.

Caustic Leaching Test: 45.8422 g slurry used (3.8A)

$$(45.8422\text{ g slurry})\left(\frac{149.5\text{ g C104}}{169.6\text{ g slurry}}\right) = 40.4\text{ g C104 sample used}$$

9/29/99

Solubility Versus Temp Test: 31.0459 g slurry ~~used~~ used (see step 1.4 in test plan)

$$(31.0459\text{ g slurry})\left(\frac{149.5\text{ g C104}}{169.6\text{ g slurry}}\right) = 27.3665\text{ g C-104 sample used.}$$

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ENGINEERING WORKSHEET

Prepared By: S.J. Tumatto	Date: 9/24/99	Project:

Subject: C-104 Test: Example Calculations

Dilute Hydroxide Washing

Aluminum

Sample C104-AQ-9 → 126 mg/g found

Correcting for sample evaporation:

$$(126\ mg/g)\left(\frac{10.2126\ g}{10.2354\ g}\right) = 125.7^{V}\ mg/g$$ (basically no change)

Total weight of wash solutions = 293.5150 g (2.70A)

Thus, $(125.7^{V}\ mg/g)(293.515\ g) = 36901$ mg Al in the wash solutions

Sample C104-AQ-8 → 163000 mg/g, 152000 mg/g } Na_2O_2 fusions
145000 mg/g, 147000 mg/g } KOH fusions

⇒ Mean 151750 mg/g
Std. Dev. 8057 mg/g

$$\text{Relative Error} = 100\left(\frac{8057}{151750}\right) = 5.3\%$$

Wt. of dried washed solids = 14.3589 g (2.69A)

Thus, $(151750\ mg/g)(14.3589\ g) = 2178963$ mg Al in the washed solids

~~2178963~~ 2178963 + 36901 = 2215864 mg Al in the C-104 sample

$$\frac{2215864\ mg\ Al}{44.8\ g\ sample} = 49461\ mg\ Al/g\ C\text{-}104\ sample$$

$$100\left(\frac{36901}{2215864}\right) = 1.7\%\ Al\ removed\ by\ washing$$

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ENGINEERING WORKSHEET

Prepared By: M.J. Lumetta	Date: 9/24/99	Project:

Title/Subject: C-104 Test : Example Calculations

Caustic Leaching

40.4 g C-104 sample used
7.6051 g dried leached solids
98.8366 g leach solution
180.7635 g wash solution

Sample C104-04-3A → 45900 mg/g Al

Correcting for sample evaporation: $(45900)\left(\frac{17.5605}{17.5709}\right) = 45873$ mg/g MJL 9/24/99

(45873 mg/g)(98.8366 g) = 4533931 mg Al in leach solution

Sample C104-04-9 → (2080 + 2050)/2 = 2065 mg/g

Correcting for sample evap.: $(2065)\left(\frac{9.9915}{10.0142}\right) = 2060$ mg/g

(2060 mg/g)(180.7635 g) = 372373 mg Al in the wash solution

Sample C104-04-8

36,100 } Na_2O_2 fusion
35,700 }
32 600 } KOH fusion
32 600 }

⇒ Mean 34250 mg/g
Std. Dev. 1912 mg/g

Rel. error = $100\left(\frac{1912}{34250}\right) = 5.6\%$

(34250 mg/g)(7.6051 g solids) = 260475 mg Al in leached solids

4533931 + 372373 + 260475 = 5166779 mg Al in C-104 sample

$\frac{5166779 \text{ mg}}{40.4 \text{ g}} = 127,890$ mg Al/g C-104 sample (note that this does not agree with that obtained in the washing test.)

$100\left(\frac{4533931 + 372373}{5166779}\right) = 95\%$ Al removed.

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ENGINEERING WORKSHEET

Prepared By:	Date:	Project:
M.I. Lumetta	12/8/99	BNFL 29953

Title/Subject: C104 Test: Comparison of Laser Fluorimetry U data + ICP-MS U data

Washed Solids

Laser Fluorimetry → 25550 mg/g

ICP-MS → ^{235}U 8.63×10^{-4} mCi/g ÷ 2.2×10^{-6} Ci/g → 392 mg ^{235}U/g

^{238}U 1.85×10^{-2} mCi/g ÷ 3.4×10^{-7} Ci/g → 54,412 mg ^{238}U/g

54 800 mg U/g

Laser fluorimetry result ~ 1/2 less than ICP-MS

Leached Solids

Laser Fluor: 100100 mg/g

ICP-MS: ^{235}U 0.0015 mCi/g ÷ 2.2×10^{-6} Ci/g = 682 mg/g

^{238}U 0.0326 mCi/g ÷ 3.4×10^{-7} Ci/g = 95882 mg/g

96560 mg/g

Laser Fluorimetry result within 4% of ICP-MS result.

Appendix D. Statistical Analysis of the Data

Statistical analyses were performed on the data included in this report. In general, simple summary statistics were provided throughout that included estimates of the average (Mean), standard deviation (Std. Dev.) and percent relative standard deviation (%RSD = 100*Std. Dev./Mean) of aliquots. If one or both of the duplicate values were within 10 times the detection limit (values in parentheses in the tables) their mean and standard deviation are also marked in parentheses in the tables. By convention values less than the detection limit ("<") are formatted with 1 significant digit, values within 10 times the detection limit are formatted with 2 significant digits, and all other values are formatted with 3 significant digits.

More detailed statistical analyses included:

- Solubility versus Temperature Study Regression & Modeling Analyses
- Solubility versus Temperature Study Tests for Changes due to Temperature
- Washing and Leaching Studies Estimates of Uncertainty for analyte concentrations in the washed and original untreated solids and the percent removal

For all of the following analyses, it should be kept in mind that all data in each study are taken from one run of the experiment on a single sample. This means that the conclusions may be limited to this particular sample for this particular run. The data provide no information about the additional uncertainty that would result from running different samples or from repeating the experiment on similar samples. The only sources of variability present in these studies are sub-sampling variability and measurement variability. Consequently, the uncertainty statements developed in this report probably underestimate the variability that will be experienced in the real world application of these conclusions.

Solubility vs Temperature Study Regression & Modeling Analyses

The statistical analyses performed here are quantitative assessments of the nature of the relationship between analyte concentrations and temperature. These analyses were performed using the evaporation-adjusted concentrations from Tables 1, 2 and 3. The data were taken from the original Excel spreadsheet and may have additional digits compared to the formatted table values. Only those analytes that had two or fewer of their reported values below the detection limit were used. These statistical analyses were performed using linear and non-linear regression procedures in the Statistical Analysis System (SAS Institute Inc. Cary, North Carolina). Two approaches were used: polynomial regressions that attempt to fit the data without a specified mechanistic model, and a psuedo-Arrhenius model.

Since there are only three temperature points (30, 40, and 50°C), the maximum polynomial regression model that can be fit as a function of temperature is a quadratic. The two concentration values per temperature provide for estimating sub-sampling and measurement uncertainty and for testing the lack-of-fit of the linear regression. The general approach taken was to first fit and test a linear regression, i.e., is a linear regression statistically better than no model. This was followed by a test of the lack-of-fit of the linear regression model, or equivalently in this case, whether adding the quadratic term would be useful in describing the solubility-temperature relationship. It should be noted that no model

can fit this data better than a quadratic model, so if the quadratic model is not significantly better than the linear model, then no other model will be significantly better either.

Table D.1. Linear and Quadratic Polynomial Regression Analyses

Analyte	Estimated Intercept	Estimated Increase per °C	P-value	
			Simple Linear	Quadratic/ Lack-of-Fit
Ag	-0.707	0.0298	0.001	0.188
Al	9.87	-0.115	0.468	0.107
Ba	3.07	-0.0621	0.154	0.373
Ca	-8.82	1.01	0.225	0.983
Cd	4.16	-0.0496	0.002	0.382
Co	0.889	-0.000740	0.705	0.656
Cr	8.18	0.633	0.004	0.946
Cu	3.43	-0.0122	0.203	0.438
Fe	2.42	-0.0231	0.085	0.864
K	254	-0.292	0.687	0.471
Mg	-6.42	0.481	0.143	0.842
Mo	3.79	-0.00893	0.419	0.297
Na	29,256	124	0.353	0.035
Ni	64.0	-0.323	0.108	0.742
P	768	-5.36	0.026	0.681
Si	23.8	8.09	0.123	0.855
Zn	6.77	-0.0848	0.318	0.236

The regression estimates are grayed-out (judged unusable) if: the estimated increase is not significantly different from zero (linear p-value > 0.1) or the lack-of-fit of the linear regression is significant (lack-of-fit p-value < 0.1).

Table D.1 presents the results of the linear and quadratic polynomial regression analyses. Included are the estimates of the intercept and slope for the linear regression. Also included are the probabilities (p-values) for the test of the linear regression and for the test of the lack-of-fit of the linear regression. A significance level of 0.10 was used. Those analytes that have a significant linear regression have a simple linear p-value < 0.10. Those analytes that have a significant lack-of-fit from the linear regression have a lack-of-fit/quadratic p-value < 0.10. Those analytes that did not have a significant linear regression or had a significant lack-of-fit are grayed-out in the table to indicate that their linear regression estimates are not considered useable.

The proposed psuedo-Arrhenius dissolution model has the following form:

$$\text{Concentration} = e^{\text{B-A/Temperature}}$$

or the algebraically equivalent form

$$\ln(\text{Concentration}) = \text{B-A/Temperature}.$$

Although these two forms are algebraically equivalent, the estimates of A and B can be different depending on which form is fit due to the least-squares criterion for fitting being applied in regular space (the first form) or log space (the second form). If there is not much variability in the data the estimates of A and B should be close by either form, if there is large variability in the data the estimates of A and B can be quite different between the two forms. Review of the data did not indicate any particular reason to use the second form, such as increasing variability with increasing concentrations, so the first form was used. This will also make the results more comparable to the polynomial regressions, which were done in regular space.

Table D.2. Dissolution on Kinetics Model [Concentration = exp (B-A/Temperature)]

			Asymptotic 90% Confidence Interval	
Analyte	Estimated B	Estimated A	Lower	Upper
Ag	1.98	110	79.1	141
Al	0.317	-49.7	-130	30.6
Ba	-7.17	-226	-687	236
Ca	4.74	50.4	-32.1	133
Cd	-0.109	-33.4	-42.6	-24.2
Co	-0.178	-1.02	-7.80	5.76
Cr	4.25	28.9	17.9	39.9
Cu	0.936	-5.44	-14.4	3.56
Fe	-0.192	-22.5	-43.6	-1.49
K	5.46	-1.25	-10.1	7.61
Mg	4.08	60.8	-19.4	141
Mo	1.16	-3.03	-12.5	6.4
Na	10.6	6.49	-3.80	16.8
Ni	3.70	-8.87	-18.9	1.13
P	5.96	-13.6	-23.2	-3.96
Si	6.77	36.0	-5.37	77.3
Zn	-0.0536	-47.2	-111	16.8

The kinetic model estimates are grayed-out (judged unusable) if: the asymptotic 90% confidence interval of the temperature related parameter A includes zero.

Table D.2 presents the results for the proposed psuedo-Arrhenius dissolution model. Included are estimates of the B and A parameters. Also included are 90% confidence intervals on the temperature related A parameter. Those analytes whose confidence interval on A includes zero (i.e., those for which the lower value is negative and the upper value is positive) are grayed out in the table to indicate that their psuedo-Arrhenius dissolution estimates are not considered useable.

Plots for all analytes assessed are included. The following plotting symbols are used for the data:

- filled diamond—data that was ∃10-times the detection limit

- empty diamond—data that was <10-times the detection limit
- descending triangle—detection limit

The plots also show the linear regression with a solid line, 90% confidence intervals on the mean with dashed lines, and the psuedo-Arrhenius dissolution model with a dotted line. Occasionally, a confidence interval is so wide it goes off the plot.

The aliquot variability is relatively large for some analytes and, along with small sample numbers, leads to "non-significant" regressions for some analytes that may appear to show a relationship. Five of the analytes in Table D.1 had linear p-values <0.1 and quadratic p-values >0.1 and produced useable linear regression equations. These same five analytes showed useable psuedo-Arrhenius dissolution models in Table D.2. Visual comparison of the linear regression model and psuedo-Arrhenius dissolution model indicated they would produce very similar results in most cases. This may be an indication that, even if the psuedo-Arrhenius dissolution model might be better over a larger range of temperatures, the relationships can be closely approximated by a simple straight line over the 30 to 50°C range.

Solubility vs Temperature Study Tests for Changes Due to Temperature

Concentration changes in the Solubility versus Temperature study are expressed as the concentration change at each temperature relative to the concentration at 30°C. This is calculated as $100*(C_T-C_{30})/C_{30}$, where C_T is the average concentration at temperature = T (40 or 50°C) and C_{30} is the average concentration at 30°C. Table 4 shows these estimates of the change in concentrations for detected analytes for the unadjusted data and Table 5 shows them for the adjusted data.

The following method was used to judge whether the reported changes were significantly different from zero or could instead simply be an artifact of sub-sampling and measurement uncertainty. There is very little data here to estimate the variability at any one temperature with any confidence so a pooled estimate of uncertainty was obtained by pooling the %RSDs at the three temperatures. This assumes the RSDs are relatively constant at each temperature. This result in turn was used as input to standard propagation-of-errors calculations for the variance of the estimation formula $100*(C_T/C_{30})-100$. This results in an estimate of the standard deviation of the % Change as C_T/C_{30}*sqrt(2)*Pooled %RSD. This in turn, was used to generate the range of a two-sided 90% confidence interval using a t-table value of 2.353 for 3 degrees of freedom. Any % Change that is larger than the range indicates the % Change is probably different from zero and is considered strong evidence of a change in concentration. These significant temperature-related changes are bold-faced in Tables 4 and 5.

Washing and Leaching Studies Estimates of Uncertainty for Analyte Concentrations in the Washed and Original Untreated Solids and the Percent Removed

The ability to derive estimates of uncertainty for the values reported in Tables 8 and 11 was even more hampered than it was for the % Change estimates discussed in the previous section. The calculation of the concentrations in Washed or Leached Solids and the

Original Sample were made using a number of sample weights and fraction constituent amounts. Only one of these inputs, namely the solids fraction, had duplicate aliquot data that could be used to estimate sub-sampling and measurement variability. The % Removed calculation in these two tables is even more problematic because of the use of even more terms and because it is the ratio of two other estimates.

To get at least some handle on the uncertainty of these estimates the following approach was taken:

- Treat all weights used in the estimation formulas as constants (without error) under the assumption that their uncertainties are much smaller than the uncertainties in the concentration measurements and can be safely ignored.
- Assume that duplicate aliquots had a common variability.
- Present a "pseudo" 95% confidence interval for at least one value of a %RSD that is assumed to be equal for all measurements that were used in any equation. A %RSD of 10 was chosen as the initial candidate as it appeared to be near the median of %RSDs seen in this study and seems to represent a reasonable starting point. This selected estimate on the uncertainty can be adjusted to determine the effects of other %RSD values by multiplying the "pseudo" 95% confidence interval values by the ratio of any other practicable %RSD divided by 10.

As input to the "pseudo" 95% confidence intervals, it was necessary to again use propagation of errors techniques to develop approximate standard deviations. These standard deviations were then multiplied by 2 (close to 1.96 from a standard normal distribution) to give the "pseudo" confidence interval half widths. Note that the use of the standard normal distribution does not account for the minimal amount of data available for these estimates, and provides much smaller confidence intervals than those that would be obtained using a Student's t distribution with only a few degrees of freedom.

For concentrations in Washed or Leached Solids and the Original Sample, the calculations are simple additions of fraction amounts divided by the sum of the corresponding fraction weights. The following propagation-of-error rules were used to develop propagation-of-errors formulas for their standard deviations:

- Variance of a mean is the variance of the measurement/n (the number of values used in the mean)
- The variance of a sum is the sum of the variances
- Constants (sample weights in this case) carry through.

This resulted in a general form for these two concentration estimates as:

$$\text{Std.Dev.} = \text{sqrt}\left(\Sigma_f(\text{var}(f)/n_f)\right)/\text{weights},$$

where f= each fraction used in the calculation of the concentration. Each var(f) term in the propagation-of-errors formula can be replaced, by definition, with $(mean_f*\%RSD)^2$. Since the same %RSD is assumed for all measurements, %RSD can be factored out, resulting in the following general formula:

$$\text{Std.Dev.} = \%RSD*\text{sqrt}\ (\Sigma_f(mean_f^2/n_f))/\text{weights}$$

The actual version of this general formula used for each analyte for each concentration estimate depends on the fractions that were used to calculate it and the number of sub-samples available for each fraction. Certain calculations used the same sample weights in the numerator and denominator. These were cancelled out and removed in the actual error propagation formulas.

For % Removal, the calculations involve 100 times the ratio of two terms, each of which is the sum of fraction amounts. The initial standard propagation-of-errors form of the Std.Dev. for this ratio of two terms is:

$$\text{Std.Dev.} = 100*num/den*\text{sqrt}(var(num)/num^2 + var(den)/den^2)$$

where num = the numerator term, den= the denominator term, and var() is the variance of each.
Both the numerator and denominator also need to have propagation-of-errors applied to them.
Again, each var() term in their propagation-of-errors formula can be replaced, by definition, with $(mean*\%RSD)^2$. Since the same %RSD is assumed for all measurements, %RSD can again be factored out, resulting in the following general formula:

$$\text{Std.Dev.} = 100*\Sigma_f mean_f/\Sigma_d mean_d*\%RSD* \text{sqrt}(\Sigma_f(mean_f^2/n_f)/(\Sigma_f mean_f)^2 + \Sigma_d(mean_d^2/n_d)/(\Sigma_d mean_d)^2)$$

where f = each fraction used in the numerator and d = each fraction used in the denominator. As for the concentration estimates discussed above, the actual version of this general formula used for each analyte depends on the fractions that were used to calculate the numerator and denominator and the number of sub-samples available for each fraction. Certain calculations use variances that have been calculated in prior steps, so these were put directly into the formulas instead of recalculating them.