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Analysis of Sludge from Hanford K East Basin Canisters

Contributors:

T. L. Welsh, Statistics and Chemistry
B&W Protec, Inc.

R. B. Baker, In-Basin Experience
DE&S Hanford, Inc.

E. W. Hoppe, Organic Chemistry
A. J. Schmidt, Organic Chemistry
J. Abrefah, Thermo-Gravimetric Analysis
J. M. Tingey, Thermo-Gravimetric Analysis
P. R. Bredt, Physical Properties
G. R. Golcar, Physical Properties
Pacific Northwest National Laboratory

Compiled and Edited by:

B. J. Makenas
DE&S Hanford, Inc.

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ANALYSIS OF SLUDGE FROM HANFORD K EAST BASIN CANISTERS

Contributors:

T. L. Welsh, Statistics and Chemistry
B&W Protec, Inc.

R. B. Baker, In-Basin Experience
DE&S Hanford, Inc.

E. W. Hoppe, Organic Chemistry
A. J. Schmidt, Organic Chemistry
J. Abrefah, Thermo-Gravimetric Analysis
J. M. Tingey, Thermo-Gravimetric Analysis
P. R. Bredt, Physical Properties
G. R. Golcar, Physical Properties
Pacific Northwest National Laboratory

Compiled and Edited By:
B. J. Makenas
DE&S Hanford, Inc.

Date Published
September 1997

Document Title: **ANALYSIS OF SLUDGE FROM HANFORD K EAST BASIN CANISTERS**

Compiled by: Bruce Makenas 9/4/97
B. J. Makenas Date
SNF Characterization Project

Reviewed by: RB Baker 9/8/97
R. B. Baker Date
SNF Characterization Project

Reviewed by: TL Welsh 9/9/97
T. L. Welsh Date
B&W Protec

Reviewed by: 9/9/97
G. R. Golcar Date
Pacific Northwest National Laboratory

Approved by: LL Lawrence for RPO 9/10/97
R. P. Omberg, Manager (Acting) Date
SNF Characterization Project

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ANALYSIS OF SLUDGE FROM HANFORD K EAST BASIN CANISTERS

Contributors:

T. L. Welsh,[2] Statistics and Chemistry
R. B. Baker,[3] In-Basin Sampling Experience
E. W. Hoppe[1] and A. J. Schmidt,[1] Organic Chemistry
J. Abrefah[1] and J. M. Tingey,[1] Thermo-Gravimetric Analysis
P. R. Bredt[1] and G. R. Golcar,[1] Physical Properties

Compiled and Edited By: B. J. Makenas[3]

ABSTRACT

Sludge samples from the canisters in the Hanford K East Basin fuel storage pool have been retrieved and analyzed. Both chemical and physical properties have been determined. The results are to be used to determine the disposition of the bulk of the sludge and to assess the impact of residual sludge on dry storage of the associated intact metallic uranium fuel elements.

This report is a summary and review of the data provided by various laboratories. Although raw chemistry data were originally reported on various bases (compositions for as-settled, centrifuged or dry sludge) this report places all of the data on a common comparable basis. Data were evaluated for internal consistency and consistency with respect to the governing sample analysis plan. Conclusions applicable to sludge disposition and spent fuel storage are drawn where possible.

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[1] Pacific Northwest National Laboratory
[2] B&W Protec, Inc.
[3] Duke Engineering & Services Hanford, Inc.

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AEA	Alpha Energy Analysis
ACL	Analytical Chemistry Laboratory
AR	As-Received
CI	Confidence Interval
CPT	Chemical Process Technology
DCB	decachlorobiphenyl
DFTPP	decafluorotriphenylphosphine
DSC	Digital Scanning Calorimetry
DTA	Differential Thermal Analysis
DQO	Data Quality Objective
EDX	Energy Dispersive X-Ray
EPA	Environmental Protection Agency
GC/ECD	Gas Chromatography/Electron Capture Detector
GC/MS	Gas Chromatography/Mass Spectrometry
GEA (gea)	Gamma Energy Analysis
HLRF	High Level Radiochemistry Facility
HVD	Hot Vacuum Drying
IC	Ion Chromatography
ICP (icp)	Inductively Coupled Plasma
IMS	Isotopic Mass Spectroscopy
IPS	Integrated Process Strategy
KE	K East
LALLS	Low Angle Laser Light Scattering
las	Laser
LL	Lower Limit
LOI	Letter of Instruction
MCO	Multi-Canister Overpack
MS	Mass Spectrometry
NIST	National Institute of Standards and Testing
PCB	Polychlorinated Biphenyls
phos	Phosphorescence
PNNL	Pacific Northwest National Laboratories
PSD	Particle Size Distribution
RCRA	Resource Conservation and Recovery Act
RPD	Relative Percent Difference
RSD	Relative Standard Deviation
SAP	Sampling and Analysis Plan
SEM	Scanning Electron Microscopy
SFBWP	Sand Filter Backwash Pit
SNF	Spent Nuclear Fuel
SVOA	Semivolatile Organic Analysis
TCX	Tetrachloro-m-xylene
TGA	Thermo-Gravimetric Analysis
TIC	Total Inorganic Carbon
TIMS (tims)	Thermal Ionization Mass Spectroscopy
TOC	Total Organic Carbon
TSCA	Toxic Substances Control Act
TWRS	Tank Waste Remediation System
UHP	Ultra High Purity
UL	Upper Limit
U/L	Upper/Lower
PI	Prediction Interval
XRD	X-Ray Diffraction

ANALYSIS OF SLUDGE FROM HANFORD K EAST BASIN CANISTERS

1.0 *SUMMARY AND CONCLUSIONS*

The two Hanford K Basins are water-filled concrete pools which contain over 2000 tons of N Reactor metal fuel elements stored in aluminum or stainless steel canisters. Associated with this Spent Nuclear Fuel (SNF) is an accumulation of particulate layered material which is generally called sludge. Sludge is found on the basin floors, in canisters, and in the basin pits which are used for miscellaneous tasks such as cask handling. In fact, 14 different types of sludge have been tentatively identified depending on which basin, canister type, or pit location that the particular sludge is found. Each type of sludge is a unique nonhomogeneous mixture possibly containing corroded fuel, debris such as windblown sand or insects, rack and canister corrosion products, and/or fission products. All of the various sludges will need to be transported away from the K Basins and disposed. This report addresses the characterization of sludge found in the K East Basin fuel storage canisters.

Measurements of sludge depths (by ultrasonic techniques) in the K East Basin canisters were reported previously (Pitner 1996) and have shown that the canisters are filled with sludge to a depth of 0 to 32 cm with a calculated nominal volume, extrapolated to all canisters, of 3 m^3 (or 7.4 m^3 at the two sigma uncertainty level). Some of the canisters in K East Basin have screened bottoms and slotted sides. Fuel corrosion products (uranium oxides and fission products) found in canisters have, to some extent, mixed with wind blown debris and corrosion products (from canister walls and steel racks) since all of the K East canisters are open-top. Also, any sludge retrieved from the Basin for processing and disposal will, at least initially, contain a significant amount of water. This is because any foreseeable sludge retrieval method will entrain basin water and, perhaps, add additional water to aid the suspension and transport of sludge through pipes and equipment.

The central problem addressed in the current canister sludge characterization effort is designation of a storage method whereby sludge can be stored away from K Basins in a more environmentally acceptable area. Prime alternatives for storage include: (1) transferring sludge to Hanford double shell waste tanks and ultimate disposition along with other tank wastes (with or without pretreatment), (2) processing the sludge into a form (dry or grout) appropriate for solid waste disposal, and (3) storage of sludge in a dedicated purchased Resource Conservation Recovery Act (RCRA) qualified tank. In these cases the chemistry of the sludge must be determined, either to ensure compatibility of sludge with any waste encountered in existing tanks, to ensure that sludge does not contain chemicals which are incompatible with the grout-forming process or interim dry storage or to aid the permitting process for a new tank. It is expected that most of the sludge residing in canisters will be dealt with during or after the movement and repackaging of fuel elements for dry storage.

The second part of the current task is to obtain canister sludge which represents, as closely as possible, the sludge which might accompany fuel elements in an multi-canister overpack (MCO) during dry storage. The actual sludge in the MCO will be that which resides within the element near corrosion sites but canister sludge (with it expected high fuel content) is a valuable stand-in. Especially important here is knowledge of bound water content of sludge which may be a source of oxidizing material or of pressurization from radiolysis.

1.1 SAMPLE GATHERING AND ANALYSIS

A campaign to obtain nine representative samples of sludge from K East canisters has been completed. Canisters were chosen to span a diversity of expected sludge constituents (Welsh 1996). Equipment (Baker 1996; Prescott 1996) was designed and utilized which assured that material, representative of local conditions, was collected from each canister.

Samples were taken from (1) canisters which contained fuel and canisters which did not contain fuel, (2) aluminum and stainless steel canisters, and (3) canisters with damaged fuel and with active in-basin bubbling. Unless otherwise noted chemical concentrations given below are for centrifuged sludge (i.e., with some bulk water removed). The task of renormalizing to as-settled or dry sludge bases is discussed in the accompanying text and appendices. The following is a summary of the chemical and physical property data (Silvers 1997a, 1997b; Miller 1997a, 1997b) obtained from the K East canister samples primarily during hot cell examinations. The total as-settled volume for each of the nine samples ranged from approximately 200 to 500 ml.

1.1.1 Appearance, Chemical Reactivity, Settling Rate, and Density

Sludge color ranged from black (fueled canisters) to reddish brown (nonfueled canisters). Samples generally settled within an hour after agitation (Figure 1.1) in 2 liter water-filled cylinders. After settling, layers were observed in some of the sludge samples and discrete flakes were seen in at least two samples (see Figures 1.2 through Figure 1.4). After settling a number of samples were observed to evolve hydrogen bubbles as shown in Figure 1.5. Some hydrogen was also recovered from shipping containers prior to examination of sludge. Settled densities of sludge typically were in the range of 1.0 to 2.5 g/ml. Densities after centrifuging to remove water were 1.2 to 4.5 g/ml. Dry particle densities were 3.5 to 7.9 g/ml (measured for five samples).

1.1.2 X-Ray Diffraction

X-Ray Diffraction (XRD) provided an identification of crystallographic phases present in sludge. A major goal of this sludge sampling campaign was to ascertain if any metallic uranium, hydride, or metallic zirconium could be identified in the sludge. These species are cause for more concern over pyrophoricity than oxides. Neither uranium metal nor uranium hydride was identified in the homogenized sludge samples. However, some discrete metallic flakes were separated from the bulk of the sludge. The flakes were analyzed and shown to contain detectable amounts of zirconium and zirconium hydride. The majority of the sludge from fueled canisters consisted of uranium oxides and, in some cases, uranium hydrate. Sludge from empty (nonfueled) canisters consisted of primarily iron oxides and iron hydroxides.

1.1.3 Particle Size

Particle size analyses were performed by laser scattering on the upper and lower layers from three primary samples designated "research samples" in the SAP (Welsh 1996). The number frequency distributions of particle diameters centered around means of approximately 1 to 6 μm while volume distributions centered around means of approximately 16 to 320 μm. Ultrasonic treatment tended to break apart many of the large particles (indicating they were originally clusters of smaller particles). Example distributions are shown in Figure 1.6. For diameters greater than those considered in Figure 1.6 similar determinations were made by sieving and weighing the separated fractions. For fueled canisters slightly more than half of the sample mass was found to be greater than 700 μm in diameter. Sieving elucidated at least one instance of ion exchange (ISM) beads being present in the sludge.

1.1.4 Polychlorinated Biphenyls

Analysis of K East floor and pit sludge had previously indicated Polychlorinated Biphenyls (PCBs) up to approximately 140 ppm by weight for selected settled samples (Schmidt 1997). The PCBs have been found in K East canister sludge at approximately 1.1 ppm, (maximum by weight in settled sludge). Summary Statistics are given in Table 1.1.

Table 1.1. Summary Statistics for Polychlorinated Biphenyl Results from K East Basin Canister Sludge Analyzed by a Gas Chromatography/Electron Capture Detector.

K East Canister Sludge Samples Summary Statistics	Initial Analysis Aroclor 1254 As-Settled Sludge (μg/kg)	Reanalysis Aroclor 1254 As-Settled Sludge (μg/kg)
Minimum concentration observed	60[1] (135)	13
Maximum concentration observed	1110 (1110)	880
Number of observations	9[1] (6)	9
Mean	300[1] (420)	220
Standard deviation	370[1] (410)	300

[1] Results reported as less than a given detection value were used in calculating the summary statistics (e.g., 3 for <3); the numbers in parentheses are the summary statistics when the less than values are deleted from the calculations (see Appendix G and H).

1.1.5 pH and Ion Chromatography

The Ion Chromatography (IC) analyses were performed on water which was in intimate contact with the solid sludge particles and which was recovered from the graduated cylinders used for settling studies.* The pH ranged from 6.06 to 8.39. For ammonia all but one of the sample concentrations were below detection limits. All samples were below detection limit for phosphate and nitrite. Fluoride, chloride, and nitrate ranged up to 0.5, 3.9, and 15.9 μg/ml (of settled sludge) respectively, while sulphate was as high as 6.3 μg/ml.

1.1.6 Metals Detected by Inductively Coupled Plasma

The Inductively Coupled Plasma (ICP) results were all from acid digests of centrifuged solid sludge samples. Concentrations given here are the percent of centrifuged solids. Note that for fueled canisters uranium contents of some samples were significant (occasionally exceeding 60 wt% of centrifuged sludge). Maximum iron concentrations ranged from 3.5 in the fueled canisters to 8.9 wt% in nonfueled canisters. Aluminum concentrations ranged up to 5.6 wt%. Traces of 16 other metals were found in the sludge. The Pb, Cr, and Cd maximum concentrations were 179, 1773, and 64 ppm by weight. Sample 96-01 had significantly higher reported K, Cr, Ni, Na, and P concentrations than the other samples.

1.1.7 Uranium Content

Uranium analyses were performed by three other methods excluding ICP discussed above. In general these confirm the high uranium contents seen for ICP measurements. Sample 96-01 was again anomalous with a reported spread in uranium concentration from essentially 0 to 63 wt% of centrifuged sludge depending on the analysis method.

1.1.8 Plutonium, Americium, Strontium, and Gamma Energy Analysis

Americium-241 values were less than or equal to 337 μCi/g of centrifuged sludge. In general ^{239}Pu/^{240}Pu contents were less than 133 μCi/g. The ^{137}Cs values ranged up to 994 μCi/g and $^{89/90}$Sr up to 1910 μCi/g. The ^{106}Ru/Rh, ^{212}Bi, ^{208}Tl, $^{243/244}$Cm, ^{134}Cs, ^{144}Ce/Pr, ^{94}Nb, and ^{226}Ra were below detection limits in all cases. Cobalt-60 was found less than 1 μCi/g. The ^{154}Eu and ^{155}Eu ranged up to 16.2 and 10.6 μCi/g respectively. These isotopes are being evaluated as accountability tools which might serve (through gamma counting) as markers for fissile material. The ^{237}Np values were either below detection limits or in the range of 10^{-2} μCi/g.

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*Reference Miller 1997a implies that IC water was centrifuged from the sludge. Although this was true for floor sludge analysis, it is not true for the current canister sludge IC.

1.1.9 Radioactivity

Alpha activity was less than 200 µCi/g for all samples and beta values ranged from 14 to 4050 µCi/g. Highest alpha and beta radioactivity values did not necessarily correspond to samples with highest uranium content.

1.1.10 Thermo-Gravimetric Analysis and Percent Water

Thermo-Gravimetric Analysis (TGA) measures weight loss (usually water loss) as a function of increasing temperature. Three types of TGA measurements on the sludge were made as well as a simple percent water determination by non-continuous weighing before and after heating. The three TGA run types were:

1. Continuous temperature increases in 1 atm nitrogen. This is the type of run initially required for TWRS acceptance of sludge.
2. Discrete temperature ramp-and-hold segments in 1 atm helium.
3. Discrete temperature ramp-and-hold segments in vacuum (<0.2 torr) with monitoring of off-gas by mass spectrometry.

The TGA run types 2 and 3 were done to mimic the Integrated Process Strategy (IPS). Canister sludge in the as-settled condition, like floor sludge, contains a large mass fraction of water. Most of this water is removed at low temperatures (≤50 °C) but smaller discrete amounts of bound water are observed to be liberated at higher temperatures. Figure 1.7 gives examples of two different TGA runs.

1.1.11 Carbon Content

Total carbon ranged from 552 to 22800 µg/g of centrifuged sludge. Total inorganic carbon ranged from 251 to 11000 µg/g while organic carbon ranged from 373 to 10500 µg/g. Sample 96-01 was anomalously high in all forms of carbon.

1.2 CONCLUSION

A campaign of sludge sampling, which addresses specific Data Quality Objectives for the Hanford K East Basin canisters, has been completed. Sampling and analyses have been performed in a manner analogous to previous sampling attempts for floor sludge except that receptacles for sludge were not handled above the Basin grating because of dose considerations. K East Basin canister sludge was found to contain significant amounts of uranium and iron as well as numerous minor constituents. The sludge settles quickly in most cases and contains no detectable unoxidized uranium metal or uranium hydride by X-Ray analysis. It has, however, exhibited hydrogen generation in the laboratory.

Future campaigns are addressing K West canisters and may address characterization of the K West Basin floor sludge as well as various pits appended to the Basins.

Figure 1.1. Settling of K East Canister Sludge.

This figure shows the decreasing volume of a sludge sample versus time after agitation in 2 li graduated cylinders. In general sludge tends to settle via a falling interface between liquid and solid rather than by a buildup of solids from the bottom. Sludge Samples 96-09 and 96-11 from nonfueled canisters appeared to settle as two distinct layers with obvious different settling times. These are identified as L1 and L2 in the Figure legend.

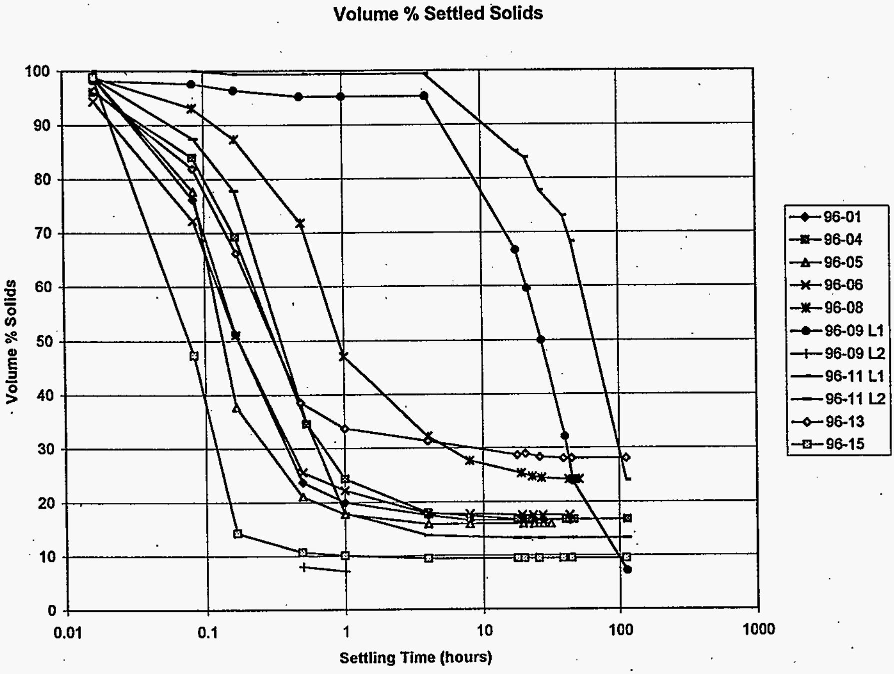

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Figure 1.2. Layers in Sample 96-06 after Settling. This sample was acquired from a fueled canister.

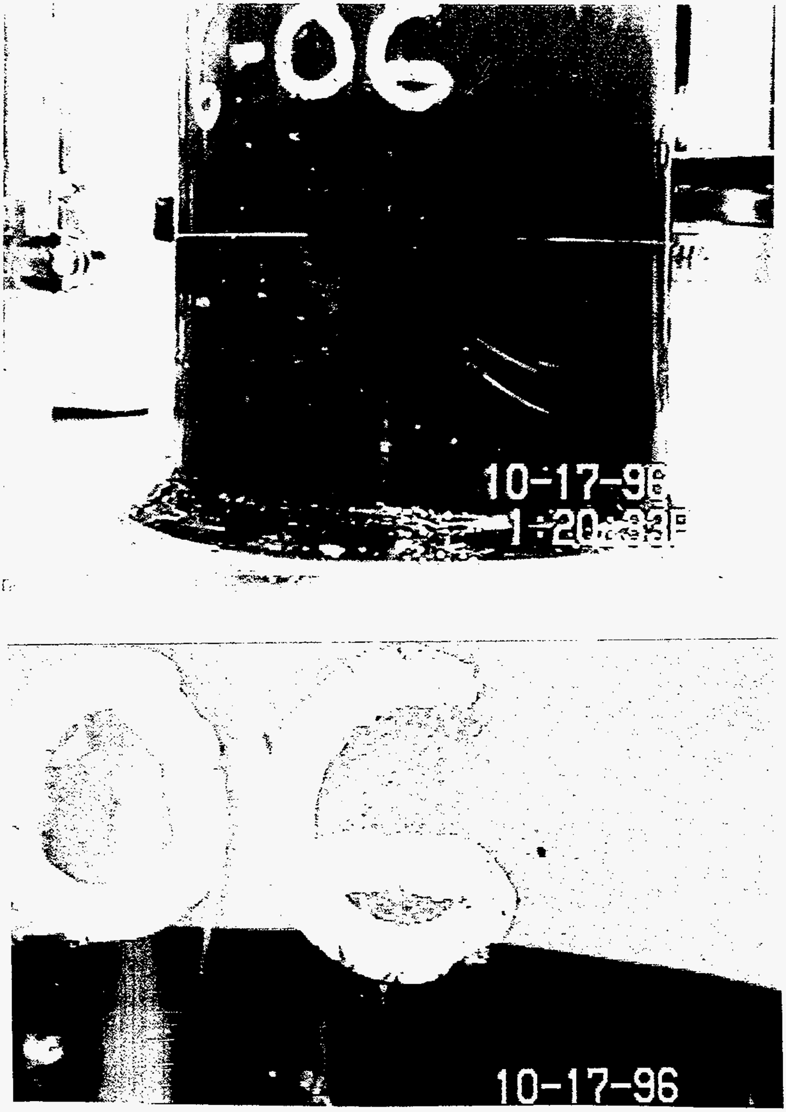
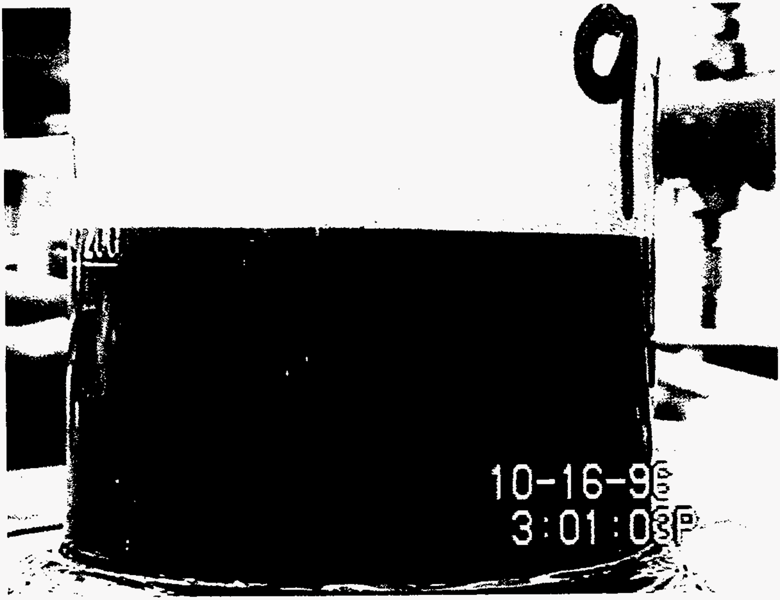


Figure 1.3. Layers in Sample 96-11 after Settling. This sample was acquired from a canister which did not contain fuel elements at the time of sampling.



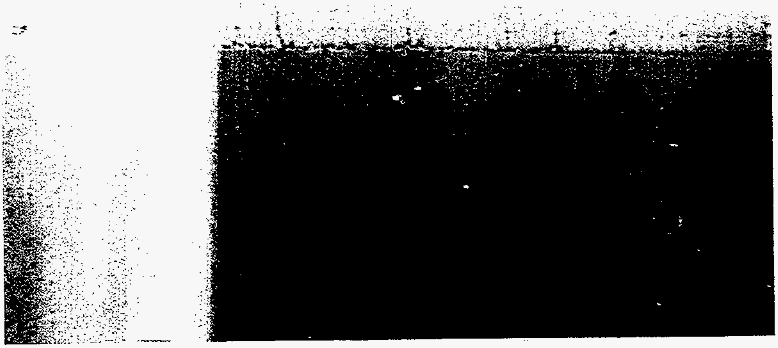


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Figure 1.4. Shiny Flakes Seen at Bottom of Settled Sample 96-04. This sample originated from a fueled canister.

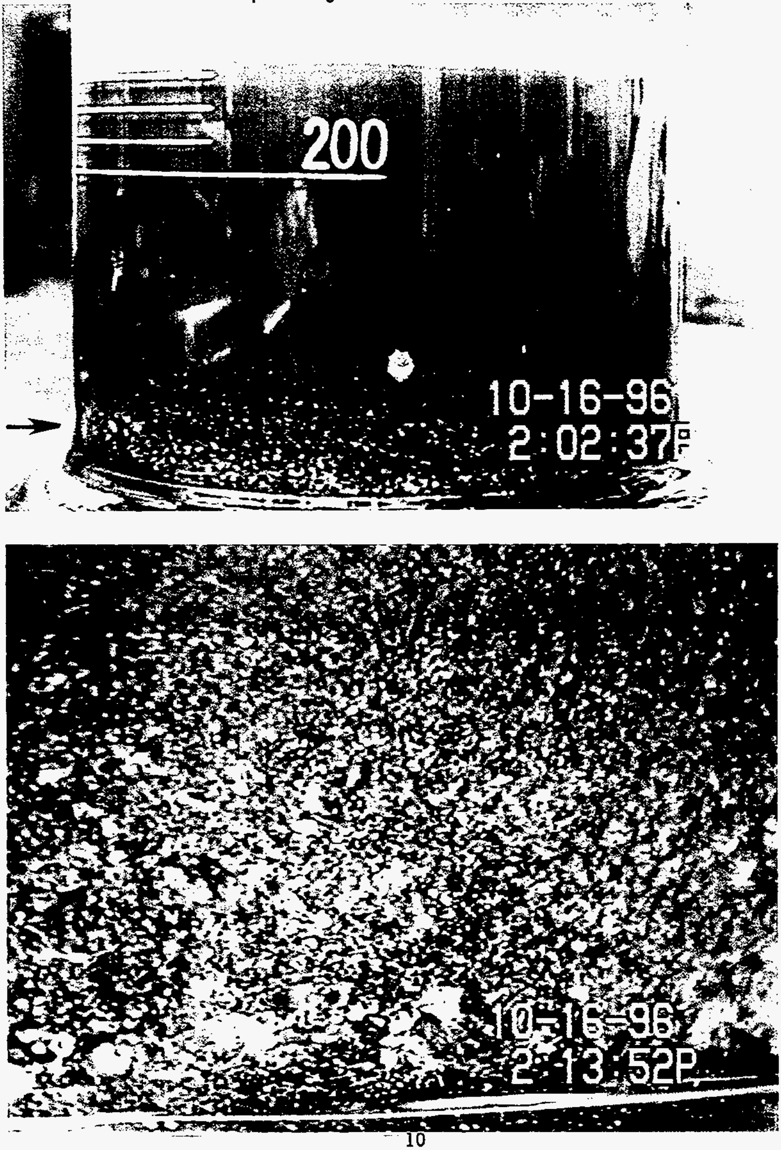

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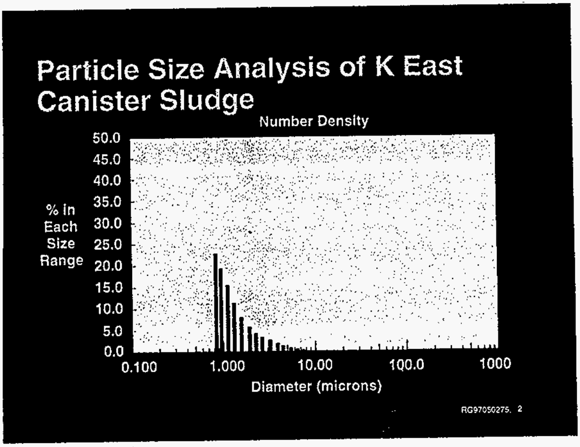
Figure 1.5. A Hydrogen Bubble Emerges from the K East Canister Sludge. Photograph taken in the 325 Building Hot Cell.

Figure 1.6. Example Particle Size Distribution for K East Basin Canister Sludge. The size distribution is shown based both on volume and number of particles for Sample 96-04 U/L without sonification. Particles over 700 μm in diameter are not counted by this laser scattering technique.



Particle Size Analysis of K East Canister Sludge

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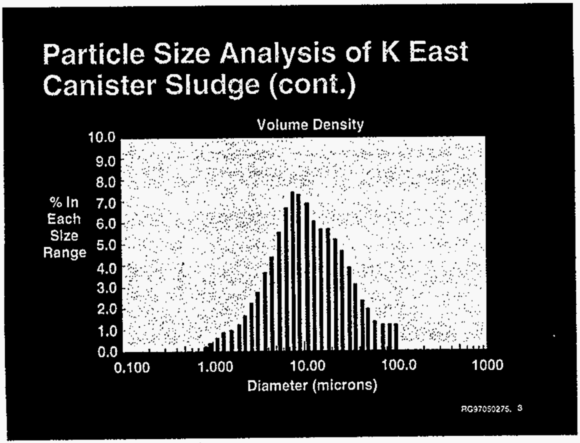
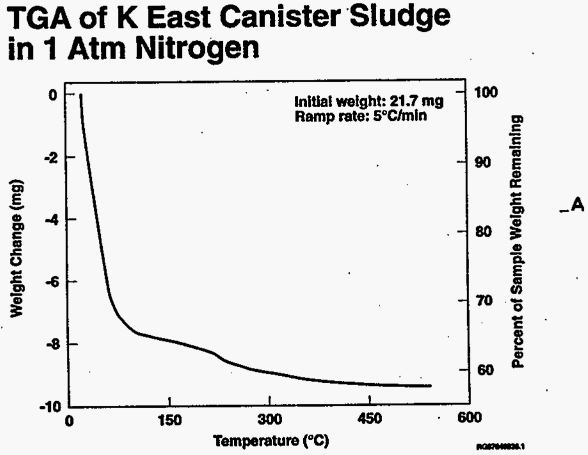


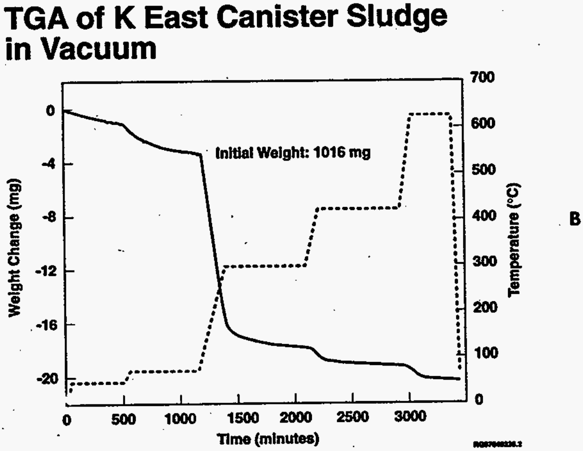
Particle Size Analysis of K East Canister Sludge (cont.)

RG97050275. 3

Figure 1.7. Examples of a Thermo-Gravimetric Analysis Drying Curve for K East Basin Canister Sludge.

(a) with continuous temperature increase in 1 atm nitrogen (300 ml/min) (b) with discrete temperature hold points in vacuum. Note weight loss during initial evacuation at 20 °C is not shown in the vacuum case.


TGA of K East Canister Sludge in 1 Atm Nitrogen


TGA of K East Canister Sludge in Vacuum

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2.0 INTRODUCTION

The Hanford K East Basin is being used to store N Reactor fuel in open top canisters made of aluminum or stainless steel. The sludge in the Hanford K East Basin canisters (as distinguished from sludge on the floor) consists of a mixture of oxidized fuel, sand, fission products from damaged fuel elements, organic material such as insects, paint chips, canister corrosion products, ion exchange beads, and concrete spallation. A recent campaign was completed to sample sludge from the K East Basin fueled and nonfueled canisters. It was performed to facilitate characterization of sludge with attention to those details which would make the resulting data applicable to the needs of the SNF Project Paths Forward for sludge and fuel.

This report supplies a reviewed set of chemical and physical property values for canister sludge and, in addition, calls attention to particular insights about sludge which have been gained during sampling and data reduction. Data Quality Objectives (DQOs) for the sampling effort (Makenas 1996a) indicate that the sampling and subsequent analyses serve the needs of several Spent Nuclear Fuel (SNF) project objectives: storage of the bulk of the sludge away from K Basins (in tanks or as processed solid waste), nuclear materials accountability, sludge recovery and transportation, sludge handling equipment design, sludge simulants, and interim dry storage of fuel with some residual sludge.

The analyses performed on sludge recovered from the K East Basin canisters fall into several classes:

1. Physical properties: Viscosity, particle shape, particle size, settling rate, zeta potential, and wet/dry densities.

2. Radionuclide content: Includes transuranic elements as well as fission products. Techniques employed include Gamma Energy Analysis and Alpha Energy Analysis (GEA and AEA).

3. Radiological Properties: Total alpha, beta, and gamma emission rates per volume of sludge.

4. Chemistry: Includes Inductively Coupled Plasma (ICP) spectroscopy, which gives the content of certain metals in sludge and Ion Chromatography (IC) which determines the concentration of certain ions in water which is in intimate contact with sludge. Also included are analyses for semi-volatile organic compounds.

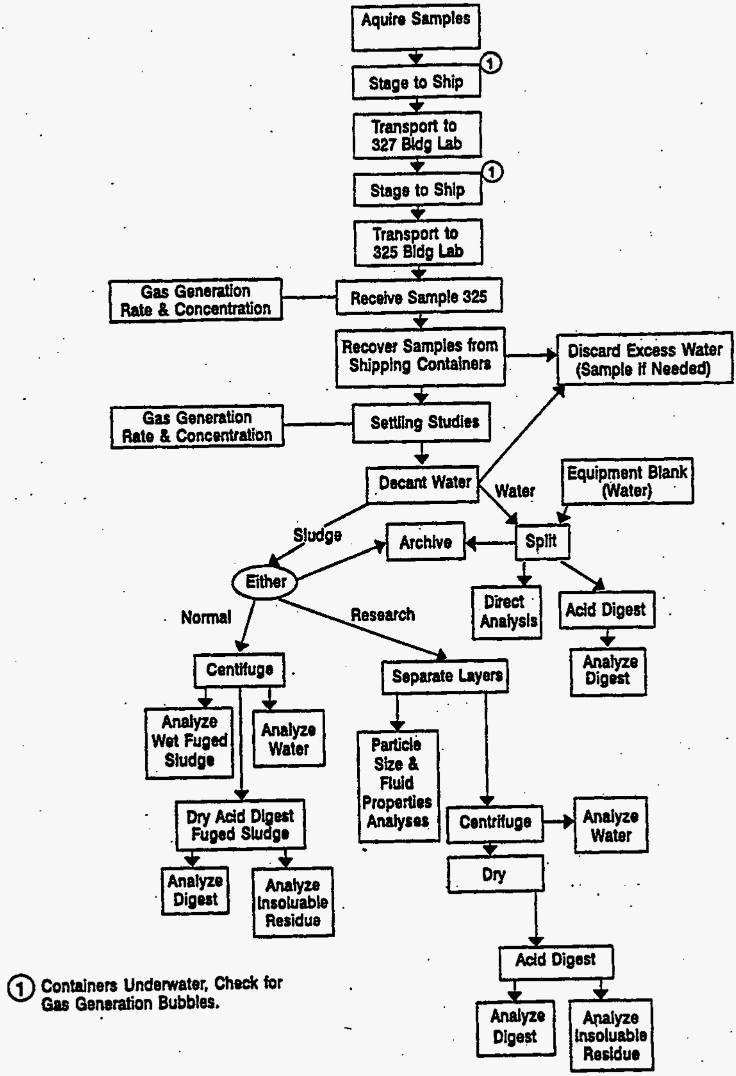
5. Chemical Reactivity Determinations: The TGA gives the percent water in a sample but also highlights specific temperatures where water and other volatiles exit the sludge. The Digital Scanning Calorimetry (DSC) indicates exothermic and endothermic reactions of the sludge in a particular medium. X-Ray diffraction is utilized to give indications of particular crystalline phases. In the case of sludge, potentially pyrophoric compounds such as uranium hydride and uranium metal were specifically targeted for analysis by XRD.

The methodology for choosing particular basin canisters for sampling is covered in reference Makenas 1996b. Nine targeted canisters were chosen with the following parameters in mind (1) damaged fuel or with no fuel, (2) aluminum or stainless steel canister material, and (3) areas of deep or shallow sludge. The capabilities of the sampling equipment used to recover the representative sludge samples are reviewed in Baker 1996 and Prescott 1996. It should be noted that since the sludge was pumped underwater to containers, considerable additional water may have been entrained in the sludge samples as a result of the pumping process. Ultrasonic measurements of sludge depth before and after collection testified to the amounts collected. The target quantities of samples were 200 to 500 ml as per the sampling requirements (Welsh 1996).

Samples were collected from the K East canisters in April 1996. After brief storage in the 327 Building pool, the material was sent to the Pacific Northwest National Laboratory 325 Building hot cells where settling studies were performed along with the decanting of excess water. Subsamples were then sent to 222-S Laboratory for most chemical analyses or retained at 325 Laboratory for other analyses (primarily physical properties and organic analyses). Figure 2.1 is a flowchart of how the samples were processed and analyzed. Note that many of the analyses (primarily chemistry determinations) required special processing steps such as drying at higher temperature. These steps were not performed where they would compromise the data (e.g., physical properties such as viscosity or organic analyses such as PCB determination).

Of the nine samples taken from canisters, three of the samples were split into layers (after settling) with equivalent analyses done separately on each layer (when material quantity permitted) in order to illuminate differences between strata. Two or three layers were identified based on color, texture, etc., in each of these three samples. Throughout this report the designations U, L, and U/L (or M) are used as suffixes to the sample numbers to indicate upper, lower, and transition layers respectively.

Figure 2.1. Flow Chart of Sludge Sample Processing and Analyses.



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3.0 RESULTS, ANALYSIS, AND EXPLANATION OF APPENDICES

The results obtained from this sludge sampling campaign can be divided into inferences derived from the actual sample retrieval, observations made in the laboratory prior to subsampling for analyses, and the data obtained from laboratory analyses.

3.1 IN-BASIN SAMPLING EXPERIENCE

This section provides a summary of the sampling method, sampling locations, and operational observations associated with the sampling of sludge in the K East Basin fuel storage canister.

3.1.1 Overview of the Sampling Method

For each sample, special equipment was used to draw representative samples from the canisters. This equipment was developed especially for sampling high fuel content (i.e., high density, high dose rate, etc.) sludges from the K Basins fuel canisters. The sampling equipment described in the System Design Description (Baker 1996) was used to draw samples of sludge with a volume on the order of 500 ml or more into a 10.3 liter primary sample container (also referred to as a shipping container), Figures 3.1 and 3.2. This equipment was designed to meet a set of functions and requirements consistent with the Sampling and Analysis (SAP) for this characterization effort (Welsh 1996).

The special equipment was designed to handle high radiation dose rate sample material (e.g., 300 R/hr at 2 in.) using the basin pool water as shielding. The sample material was maintained at least 10 ft under the water in the basin pool to assure shielding for the collection team staff. The equipment was developed to collect sludge materials with particle densities as high as those of theoretically dense uranium (approximately 18 cc/g). This requires that a significant quantity of water also be collected in order to keep the particulate matter suspended during collection. A special sludge extraction tube was constructed for insertion into the canisters to acquire the sludge. It was designed to exclude particles greater than 6.4 mm (0.25 in.) while collecting particles smaller than this diameter (see Figure 3.1). The equipment vacuums the sludge material through the extraction tube, and through a flexible hose. It then deposits sludge in the sample container. A vacuum-single-pull system was chosen because it avoids affecting the sample materials physical character, as could occur if sludge were passed through a pump or caught on filters. The equipment was thoroughly tested (Prescott 1996) in a "cold" pool facility (Hanford 305 Building) prior to operation at K East Basin.

Once a sample was drawn into a sample container, the container was monitored in the K East Basin pool for gas generation from the sludge material. The container was then transported to the Hanford 327 Building hot cell facility by cask. The sample containers were then stored in the

327 Building pool and later moved to the nearby 325 Building Analytical Laboratory. In 325 Building hot cells the sludge samples were recovered from the sample containers and analyses started.

Figures 3.3 through 3.7 show selected steps from video footage taken of the sampling in the K East Basin. Appendix A provides some additional background on the sampling equipment as well as the operational sequence. The samples were tracked through the sampling process with chain-of-custody forms to maintain traceability. The K Basin Master Work Plan used for this sampling was MWP-95-005.

3.1.2 Canisters Sampled

The SAP (Welsh 1996) for sampling the sludge from the fuel storage canisters in the K East Basin provides a set of general criteria which were used to select the canisters to be sampled. The SAP was based on the objectives identified in the Data Quality Objectives (Makenas 1996a) for this activity. The primary objective of this characterization was to identify similarities and differences between the characteristics of the canister sludge versus the characteristics of the sludge analyzed from the floor of the main basin and Weasel Pit of the K East Basin (Makenas 1996a).

The sampling of the K East canisters addressed the primary variables of: (1) canister type i.e., Mark I (aluminum or stainless steel material, solid or screen bottom, and possibly slotted barrel walls) or Mark II (stainless steel material, closed bottom), (2) fuel element condition (cladding degradation, reacted fuel, etc.), and (3) canister barrel content i.e., fueled or unfueled (the later canisters should contain material similar to floor sludge). Secondary considerations were the physical location of the canisters in the main basin, depth of sludge in barrels, and their location compared to past floor sludge samples taken.

The depths of sludge were measured in a set of candidate canister barrels in early 1996 (Pitner 1996). This sludge depth data along with (1) visual examinations, recorded on video tape, of the open-top canisters showing the upper ends of the fuel elements (Pitner 1995) and (2) the known parameters of the canisters, were considered in making the final choices of the canister barrels to sample for sludge. Tables 3.1 and 3.2 provide a general summary of the important parameters of the canister barrels selected to be sampled (Makenas 1996b).

It is noted that, as with the floor sludge sampling effort previously completed in the K East Basin, the canisters selected for sampling had to reside below the slots in main basin grating. The sampling equipment was designed to access the pool directly below these slots. This criterion was judged to impose no significant limitation (Baker 1996) since over half the fuel canisters stored in the basin were accessible to the equipment. Equipment to access "off-set" locations would have been significantly more costly and increased operator dosage during sampling, with no apparent significant benefit. The canisters to be sampled were not moved prior to sampling so they would maintain their undisturbed state. Note that some Mark I canisters have holes in the bottom of the canister barrels and some have slots on the sides of the barrels which allow full communication of the sludge in the canisters with the floor sludge or, if moved, loss of sludge from the barrel to the surrounding basin.

Selected for canister sludge sampling (Tables 3.1 and 3.2) were four canisters barrels made of aluminum and five of stainless steel. One canister barrel of each material was chosen which contained no fuel elements. These two acted as references to compare directly to floor sludge analyses made previously (Makenas 1996c). The fueled canisters had varying degrees of fuel conditions from good to very damaged, (see Appendix A). Six of the canister barrels chosen were observed to have gas bubbles being released from them into the basin pool prior to sampling (Table 3.2). This indicates the potential for on-going fuel reaction with water and for significant fuel element damage. The depths of sludge measured by ultrasound in the barrels varied from an apparent 10.2 to over 305 mm (0.4 to over 12 in.).

Figure 3.8 shows the general location of the canister samples compared to the main basin and past sampling of floor sludge. Note that one of the canister samples, Sample 96-15, was taken adjacent to the location of the previous floor sludge sample that was found to have the highest uranium content, KES-O-09 at cubical location 6970 (Makenas 1996c).

As with the sampling of the floor and Weasel Pit at K East Basin, it is noted that the Basin provides a very challenging area for sludge sampling because of (1) the degraded condition of the fuel, canisters and debris (2) the various forms of sludge (3) the nuclear environment and (4) the radiological dose rate of the samples that required their handling under 10 ft of water at all times. During this sampling campaign a few samples from canisters were rejected after sludge collection because of problems with the equipment or difficulties with the sampling of the canisters with highly degraded fuel elements and very heavy deep sludge. Video tapes of the process were reviewed after each sample drawing to help assure that representative samples were obtained. If any significant question was apparent (such as tube clogging), a replacement sample was taken from another canister barrel. The replacement canisters barrels sampled were equivalent to the ones rejected (i.e., no bias was introduced in the selected sample population).

3.1.3 Summary Observations Made During Sludge Sampling

Table 3.3 indicates the dates that samples were taken, which bay of the main basin they were taken in, the final amount of sludge recovered at the laboratory from the sample container, and any general comments by the staff taking the samples.

The sample containers were monitored for release of any gases, which would indicate on-going hydrogen generation from fuel-water reaction in both the K East Basin pool prior to shipment and the 327 Building pool while awaiting transfer to the analytical Laboratory. No significant release of bubbles was noted in either of these locations.

3.2 OBSERVATIONS DURING SETTLING OF SLUDGE SAMPLES

In the 325 Building hot cells the sludge samples were divided into two groups for settling studies due to limitations of cell floor space. Four sludge samples were transferred from metal shipping containers to individual 2 liter graduated cylinders (along with a portion of the accompanying basin water) and allowed to settle in stages during the transfer. The four samples (96-01, 96-05, 96-06, and 96-08) each consisted of 200 to 400 ml of settled sludge. The samples were then resuspended using an air lance (5 minute sparge). Attempts to loft the material using mechanical means such as magnetic stirrers proved unsuccessful. Two samples settled quickly (in approximately 4 hours) to a clear supernate while two remained cloudy for a longer time period. Figure 3.9 illustrates the condition of four of the settling containers during the settling process. In this figure, Samples 96-01 and 96-05 are those that settled quickly while Samples 96-08 and 96-06 remained murky for a longer period. Implied settling rates are discussed later in Section 3.3.

Samples 96-05 and 96-06 were observed to be generating gas bubbles before and after resuspension. Sample 96-08 generated gas bubbles only after several weeks had passed following resuspension. Eruption of bubbles could clearly be seen at the top surface of the sludge but formation of the bubbles generally began near the bottom of the sludge. Eventually, after air sparging, a pocket of gas formed at the bottom of Sample 96-05 (below the sludge). The growing wedge shaped pocket (Figure 3.10) released its gas to the covering water approximately 2 days after resuspension, with resulting dispersion of significant sludge back into the supernate liquid.

In order to further explore the gas generation and release phenomena, a helium lance was used to again resuspend the sludge in Samples 96-05 and 96-06. Bubble formation could be seen before and after resuspension (Figure 1.5). A large bubble began to form on the bottom of the 96-06 sample 7 days after the resuspension. The bubble grew and lifted most of the sludge mass (Figure 3.11) over a period of 5 days. Bubbles were observed to form in the small amount of sludge left at the container bottom and these then would burst into the enlarging gas space below the larger mass of sludge. Subsequently (when the large gas bubble reached about 650 ml) the sludge mass was mechanically probed to release the gas and avoid ejection of material from the top of the graduated cylinder.

A total of five gas samples were taken during the processing described above. These samples were analyzed for composition using mass spectrometry. Gas samples were taken from the space above sludge Samples 96-05 and 96-06 after air sparging and from Sample 96-06 after helium sparging. Two other gas samples were taken from undisturbed shipping containers containing sludge Samples 96-13 and 96-15 (from the second group of five sludge samples). In all cases the gas samples contained significant quantities of only air and hydrogen with hydrogen being the majority constituent in four of the five samples. Trace quantities of fission gasses Xe and Kr were also found presumably released from fuel during the corrosion process which produced the hydrogen. Estimates of the gas generation rate for the 200 to 400 ml settled sludge samples (based on gas collected over a known time period or calculated from initial trapped gas volume) ranged from 0.4 to 1.9 ml/hour. The rate associated with the last and largest trapped gas bubble appears to be somewhat higher (approximately 5 ml/hour).

Subsequently the remaining five sludge samples (second group) were transferred to graduated cylinders and resuspended with helium for settling measurements. Note two of these samples (96-09 and 96-11) were from nonfueled canisters. Samples 96-13 and 96-15 were observed to bubble in a manner similar to the samples in Group 1 but without formation of large trapped bubbles. Additional details on gas generation are given in Appendix B.

3.3 CHEMISTRY AND PHYSICAL PROPERTIES

The raw data from most analyses of K East Basin canister sludge are reported in (Miller 1997a, 1997b) and in (Silvers 1997a, 1997b). Note that the chemical composition values given in Miller 1997a, are for as-centrifuged sludge (i.e., compositional values when stated on a per gram basis, have been normalized to an initial sample mass which includes the water which is removed later by the drying that is routinely done prior to chemical analysis). Conversely this total mass of as-centrifuged sample does not include bulk water removed from as-settled sludge by centrifuging. Chemistry data given in References Silvers 1997a, 1997b are generally for dried or settled sludge and are therefore not directly comparable to Miller 1997a unless weight losses during centrifuging and drying are taken into account. The chemical and radionuclide composition values on a centrifuged sludge basis are given in Appendix C.

Depending on the application of the data, compositional values based on either a wet as-settled mass or a dried mass of sludge may be appropriate. Appendices D and E present the compositional data (initially presented in Appendix C as centrifuged sludge) renormalized to wet (i.e., as-settled) and dry conditions respectively. The renormalization has been performed using the mass loss data for the respective centrifuging and drying steps.

As-centrifuged and as-settled densities for sludge samples are also presented along with the chemistry summaries in Appendices C and D. Five samples, however also were subject to dry particle density measurements (i.e., helium pycnometry). These five samples correspond to various layers from the research samples. Dry particle densities ranged from 4.68 to 7.90 g/cc for four sludge samples from fueled canisters (i.e., layers recovered from 96-04 and 96-06) and 3.49 g/cc for one sample (96-11) from a nonfueled canister.

Appendix F is the statistical analysis of the chemistry results listed in Appendices C through E. Included are an estimate of the analytical variability and box plots showing the distribution of data.

All of the canister samples were analyzed for PCBs and several were subjected to semi-volatile organic analysis (SVOA). Organic compounds identified by SVOA consisted primarily of a few ppm of phthalates. Note that analyses for total carbon, total inorganic carbon, and total organic carbon are presented with the noncarbonacious analyses listed in Appendices C through E. All of the canister samples were found to contain PCBs at a level of 1.1 ppm or less by weight. The primary PCB aroclor detected was 1254. A comparison of the PCB analysis methods with the accepted EPA procedures is discussed in Appendix G. A specific focus on the amount of PCB compounds identified in each sample is provided by Appendix H and a summary is given in Table 1.1.

Appendix I is a comprehensive treatment of physical properties including viscosity, zeta potential, particle size, settling time, and particle shape for layers of the research samples. This appendix also discusses the implications of the physical property measurements on the methodology of choosing sludge simulants and the choice of sludge handling equipment. It should be noted that viscosity measurements were made on as-received sludge as well as on centrifuged sludge and on sludge diluted with water to give a family of data curves. Particle size determinations (Silvers 1997b) were made by an automated laser scattering technique for particle sizes less than 700 μm in diameter and by sieving for larger particle sizes. An example of sieving is shown in Figure 3.12. Ion exchange resin beads were encountered in canister sludge, especially during sieving (Figure 3.13), but not in the greater frequency seen in floor sludge previously. A finding of significance discussed in Appendix I is the effect of ultrasonic application to sludge particles i.e., the mean size of sludge particles decreases with increasing severity of ultrasonic treatment which implies that some large particles may be agglomerates of smaller entities. The flow rate of sludge in the size measurement apparatus was also seen to affect the measured particle size distribution with higher flow rates implying smaller measured diameters.

Also included in Appendix I are data on sludge settling times. Settled sludge varied in macroscopic appearance from reddish brown to black in color but tended more toward black than floor sludge.* Shiny flake-like material was sometimes seen at the bottom of settled samples. Settling times were usually short (less than 1 hour) but some exceptions were noted for certain discrete layers. Figure 1.1 summarizes all of the data for settling.

Appendix J summarizes XRD results. This technique identifies crystalline phases. Those phases observed by this method are listed in Table 3.4. By far the most frequently identified components of sludge from fueled canisters were uranium oxides and occasionally uranium hydrates while nonfueled canisters contained iron oxides. Various uranium oxides produce overlapping X-Ray peaks so that the stoichiometry of the oxides is difficult to determine. It is of significance to note that no uranium based pyrophoric materials such as metallic uranium, or uranium hydride were detected in any homogenized sludge sample. A number of shiny flakes were retrieved from the bulk sludge and analyzed separately by XRD. These flakes proved to be largely zirconium, zirconium hydride, uranium hydrate, or magnetite (Fe_3O_4). Also, analyzed separately were particles that clung to a magnetic stirrer in the sludge. Although uranium hydride is strongly

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*Closeup photography of the sieving operation showed a green to yellow coloration for individual particles possibly indicative of U hydrate and non-stoichiometric oxides.

ferromagnetic, the material on the stirrer consisted of iron compounds in this case. Also analyzed separately was a small amount of material which precipitated from supernate water (originally associated with Sample 96-06) long after settling studies were completed. This thin layer of material proved to be hydrates of U and Ca.

Diffraction methods do not give good identification for cases where compounds are amorphous (noncrystalline) or where very small particles are involved (<0.1 μm approximately). There is virtually no chance, however, that such small undetectable particles, if they originated as uranium metal or as hydrides, would remain unoxidized in a water environment such as the K Basin pools although freshly created fuel particles may be another matter. In the case of previously analyzed floor sludge (Makenas 1996c) significant amounts of uranium were found in elongated amorphous particles. Transmission electron microscopy and electron diffraction, which gave this insight for floor sludge, were not utilized for canister sludge.

Many of chemical determinations, discussed in the preceding sections of this report were done by wet chemistry methods. Therefore, sludge was acid digested for these chemistry analyses such as GEA, ICP, and uranium isotopics. Some residues were left after digestion and these residues also were analyzed by XRD to detect such insoluble compounds as sand. For this study of canister sludge an effort was made to ensure acid digestion methods were the same for all analyses. This may not have been true for previous floor sludge analyses.

Three crystalline compounds of unknown composition (each in a different sample) were found by XRD. Scanning Electron Microscopy (SEM) with energy dispersive X-Ray analysis (EDX) was performed on one of the samples containing an unknown structure in an effort to at least identify elemental constituents (see note in Table 3.4).

Appendix K to M contain the TGA data acquired during sludge drying under nitrogen, helium, and vacuum environments. The runs done under nitrogen atmosphere (Appendix K) were for small samples and the temperature was increased continuously while weight loss was monitored. These were performed at conditions specified by TWRS. Various changes in slope indicate possible water releases from sludge compounds. Thermo-gravimetric analyses runs for small samples (10 to 60 mg) in helium gas are presented in Appendix L. These latter runs were designed to investigate remaining water inventory in sludge associated with fuel in an MCO (during interim dry storage after IPS drying steps).

Canister sludge is the closest approximation to sludge in an MCO that is currently available. The above investigations were supplemented by TGA runs utilizing a vacuum-capable TGA instrument with a mass spectrometer to analyze off-gas. This machine can also handle larger samples than the previously discussed instrument (≈1 g of sludge). Appendix M presents a portion of the weight loss and mass spectrometer data from this latter TGA work and the complete work will also be summarized in a future topical report (Abrefah 1997).

Table 3.1. Summary of Canisters Sampled for Sludge.

Full Sample Number	Shorten Sample Number	K East Cubical Location	Canister Barrel	Number of Fuel Elements in Barrel[1]		Initial Fuel ^{235}U (wt%)	Estimated Average Fuel Burnup (MWd/MTM)	Canister		Canister Barrel has Bottom Holes/Slots?
				Inners	Outers			Type	Barrel Material	
96-01A/1845E	96-01	1845	East	4[2]	5	0.947	0	MKII	SS	No
96-04/2711E	96-04	2711	East	7	7 (2)	0.947	2600	MKI	SS	No
96-05/3128W	96-05	3128	West	6 (2)	6 (3)	0.947	2600	MKII	SS	No
96-06/5465W	96-06	5465	West	6	6 (3)	0.947	1870	MKI	Al	?
96-08/2350E	96-08	2350	East	7	7 (5)	0.947	2220	MKI	Al	?
96-09/4638E	96-09	4638	East	0	0	--	No fuel	MKI	Al	Yes
96-11/6073W	96-11	6073	West	0	0	--	No fuel	MKI	SS	No
96-13/5055W	96-13	5055	West	6	6 (3)	0.947/ 0.711	1680	MKI	Al	?
96-15/6070W	96-15	6070	West	1 (1)	7 (4)	0.947	1580	MKI	Al	?

[1] Number of elements with damage visible from top of barrel are shown in parentheses.
[2] An approximately 1 1/2 long bolt (debris) sitting on top of one inner element, see Appendix A.

Table 3.2. Summary of Observations Made for K East Canisters Sampled for Sludge.

Sample Number	Bubbles from Barrel in Basin ?[1]	Category of Fuel Condition[2]	Qualitative Barrel Condition[2]	Measured Sludge Depth in Barrel in. (cm)	Approximate Volume of Sludge in Sample[3] (ml)	Estimated Volume of Sludge in Canister Barrel Sampled (ml)
96-01	Yes	Good	Very good	2.3 (5.9)	245	1322
96-04	Yes	Poor	Good	1.7 (4.3)	260	758
96-05	No	Very bad	Fair, some corrosion	3.3 (8.5)	230	1655
96-06	Yes	Poor	Very corroded	8.6 (21.7)	260	4312
96-08	Yes	Poor	Fair, corroded	3.4 (8.7)	400	1515
96-09	No	--	Good, slight corrosion	0.4 (1.0)	120	334
96-11	No	--	Very good	0.5 (1.3)	225	418
96-13	Yes	Poor	Very corroded	5.7 (14.4)	435	2858
96-15	Yes	Poor	Fair, some corrosion	>12.1 (>30.8)[4]	170	>6670

[1] Bubbles observed being released from barrel prior to sampling in K East Basin.
[2] See Appendix A.
[3] Initial estimate of sludge recovered at laboratory from primary sample containers.
[4] Ultrasound probe has a maximum depth capability of 12.1 in. (30.8 cm), probe was buried in sludge at this depth.

Table 3.3. Observations Made During Sampling Process.

Sample	Date Sample Collected	K East Basin Bay	Comments
96-01	April 8, 1996	East, NW	Bubbles observed coming from canister barrel during sampling. Sludge moved through flexible sample tube in short bursts.
96-04	April 9, 1996	Center, SE	None.
96-05	April 9, 1996	Center, SE	Large amount of sludge and bubbles rose from canister during sampling.
96-06	April 17, 1996	West, NE	Sludge was observed, appeared white and powdery during backflush after sampling.
96-08	April 10, 1996	Center, NE	Some bubbles from canister barrel at start of sampling. Sludge is dark brown and black at times above canister barrel.
96-09	April 11, 1996	West, SE	Unfueled canister barrel, wire mesh holes on bottom visible. Hard to see sludge transfer through flexible hose.
96-11	April 12, 1996	West, NW	Unfueled canister barrel. Some sludge material observed transferring through flexible tube.
96-13	April 18, 1996	West, NE	Bubbles rose from canister barrel during sampling. White fuel corrosion products observed to be powdery. Backflush showed dark sludge material.
96-15	April 18, 1996	West, NW	Bubbles were released from canister barrel during sampling. Backflushed sludge was dark gray to brown in color (same location as floor sludge same KES-0-09, see reference Makenas 1996c).

Table 3.4. Compounds Identified by X-Ray Diffraction.

Sample Type	Number of Samples* Analyzed	Compound	Number of Observations*
Homogenized sludge (fueled canisters)	10	UO_2, U_4O_9or U_3O_7 $UO_3 \cdot 2H_2O$	10 3
Homogenized sludge (nonfueled canisters)	2	FeO (OH) Fe_2O_3 SiO_2	2 2 1
Acid digest residuals	3	Unknowns SiO_2	2 2
Clinging to magnetic stir rod	1	Fe_3O_4 Fe_2O_3 $FeCr_2O_4$ FeOOH $(Ca,Mn)\ Mn_4\ O_9 \cdot 3H_2O$	1 1 1 1 1
Shiny flakes	2	$ZrH_{1.66}$ ZrH Zr $UO_3 \cdot 2H_2O$ Fe_3O_4 Unknown**	1 1 1 1 1 1
Slow settling compounds in carboy	1	$UO_3 \cdot 2H_2O$ $C_2H_2CaO_4 \cdot 3CaO \cdot 11\ H_2O$	1 1

*Includes duplicates and/or discrete layers.

**XRD sample containing flakes from sludge Sample 96-09 was also analyzed by energy dispersive X-ray analyses in an SEM. Indications of U, Al, Ca, Si, and S were found in addition to the Fe in Fe_3O_4 found previously by XRD.

Figure 3.1. Conceptual Layout:Canister Sludge Sampling Equipment and Extraction Nozzle Detail.

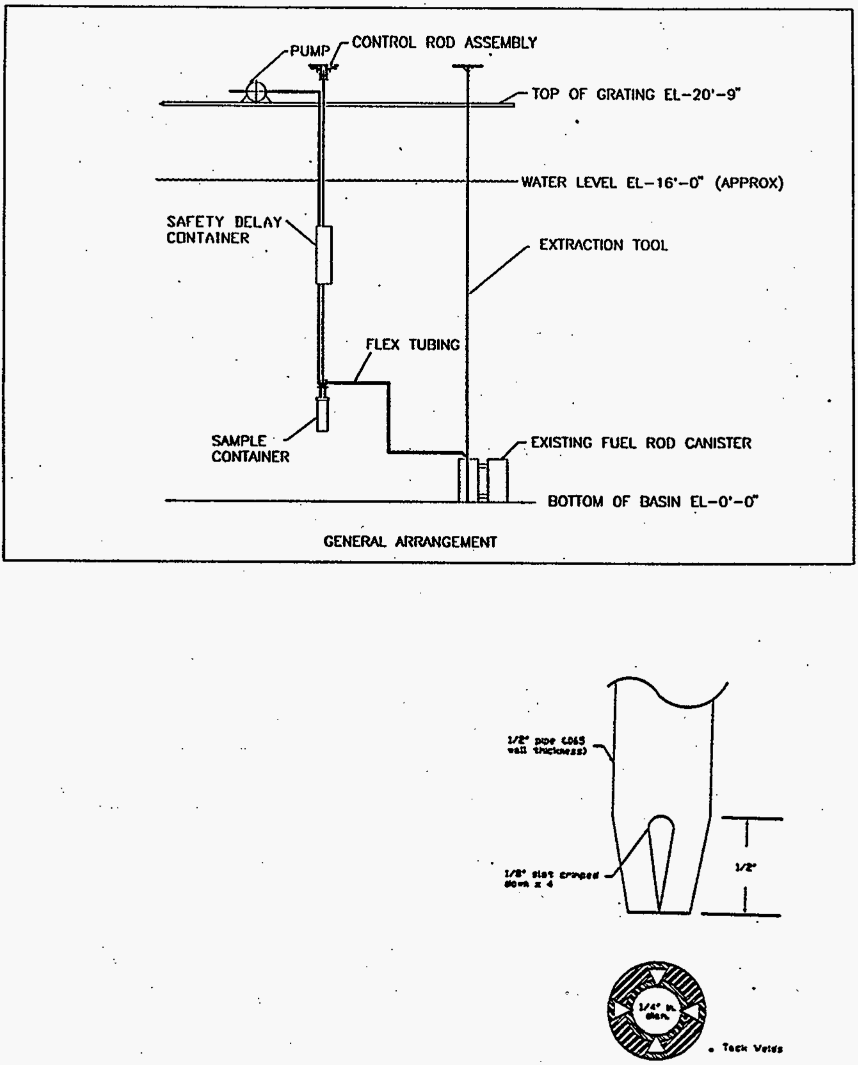

Figure 3.2. Overview of Features of Canister Sludge Sampling Equipment.

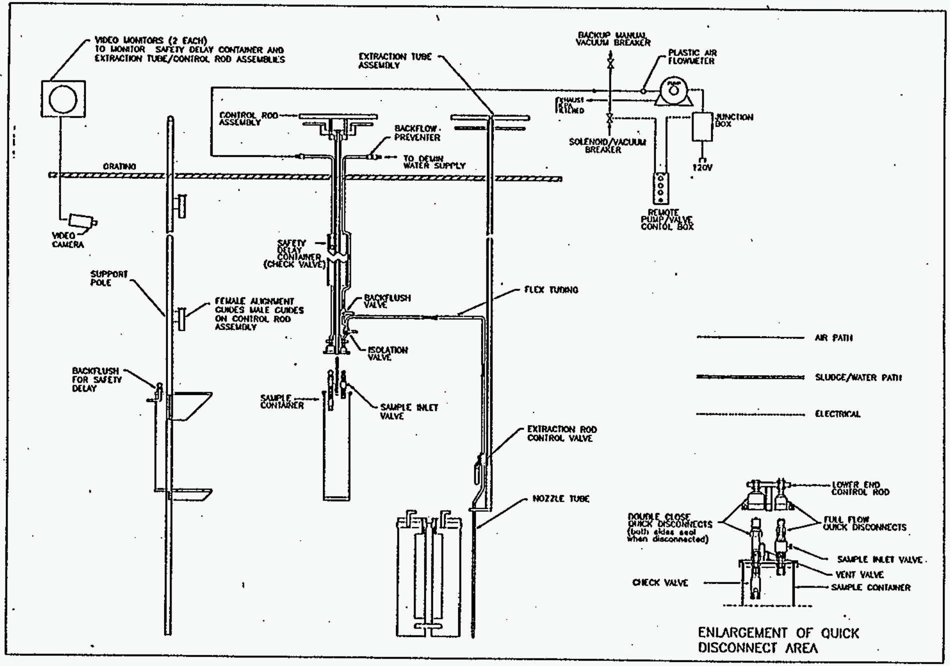

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Figure 3.3. Preparation of Sampling Equipment in the K East Basin. (Item 1 is sample container, Item 2 sample container support pole assembly, Item 3 control rod assembly with safety delay container, Item 4 extraction tube assembly]

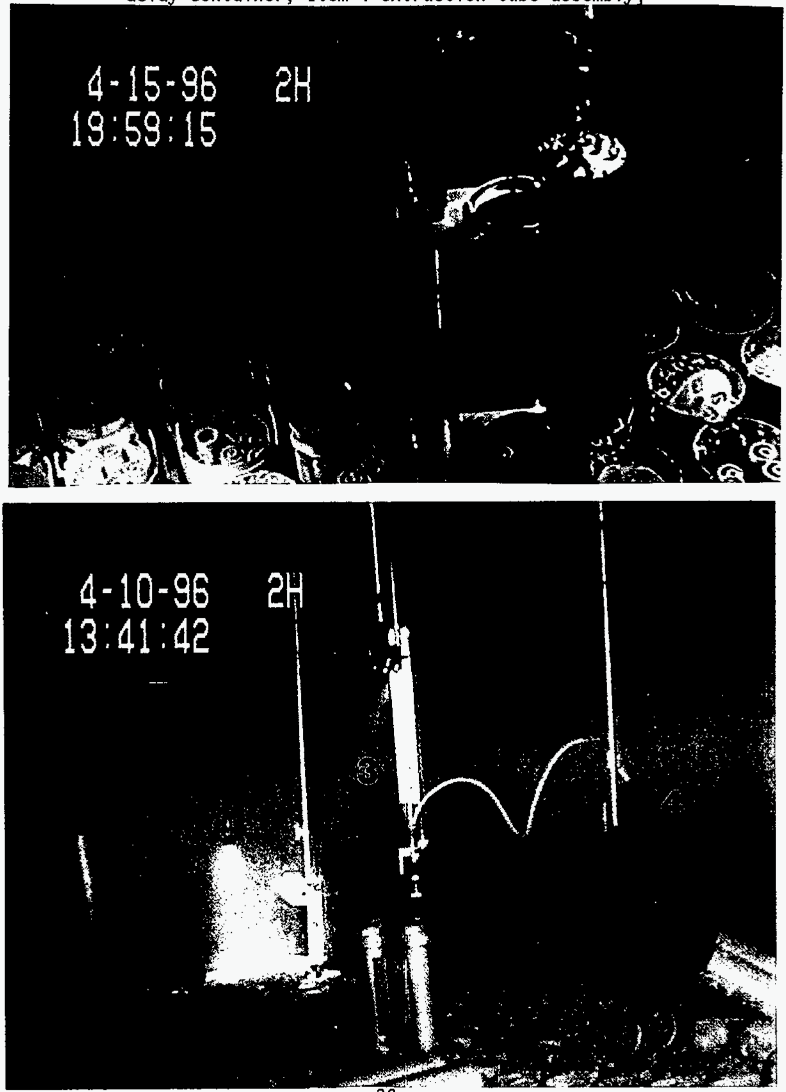

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Figure 3.4. Loading Sample Container on Support Pole and Connecting Control Rod Assembly to Sample Container.

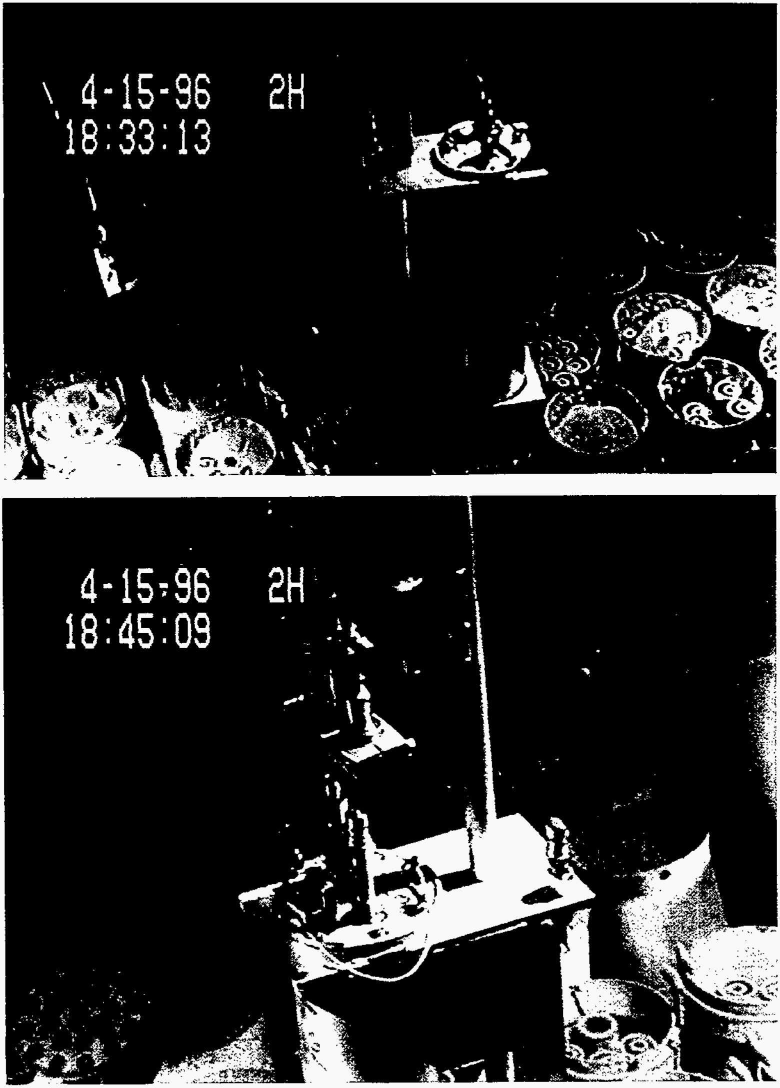

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Figure 3.5. Extraction Tube Nozzle above and Inserted in Fuel Storage Canister Barrel Between Fuel Elements.

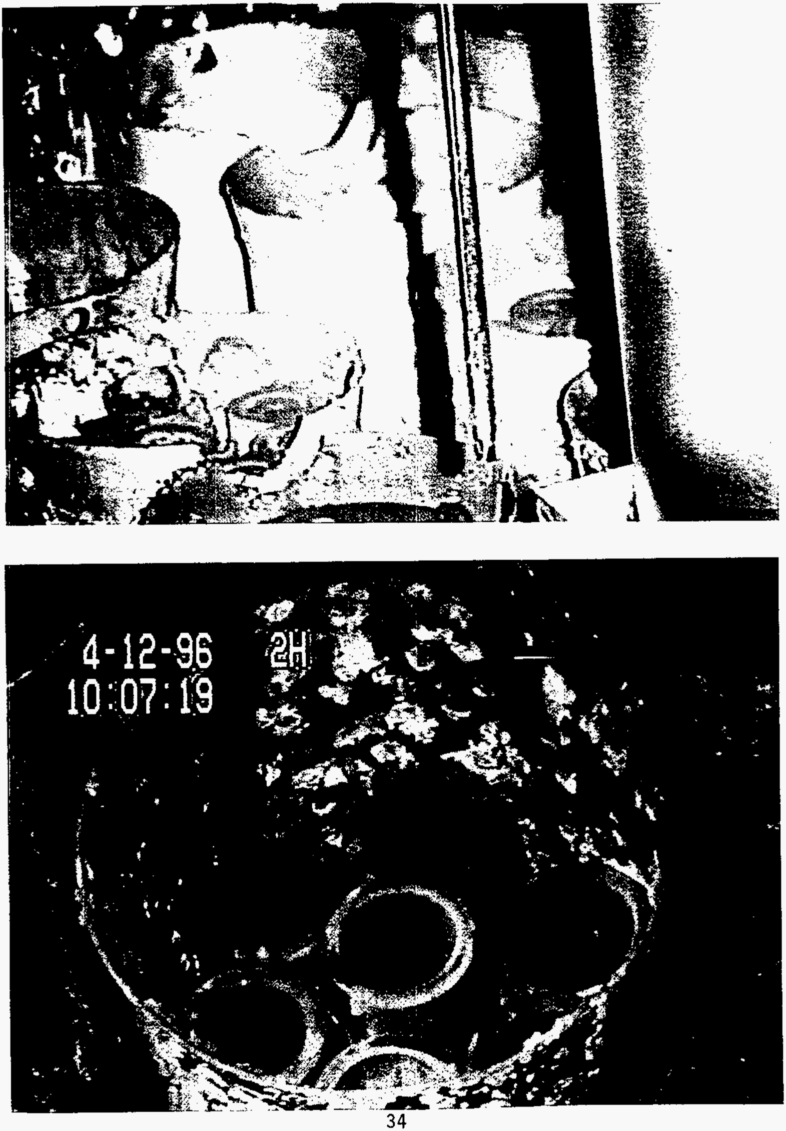

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Figure 3.6. Extraction Tube Drawing Sample of Sludge (96-13) and View of Sludge Being Transferred through Flexible Hose into Sample Container During Sampling.

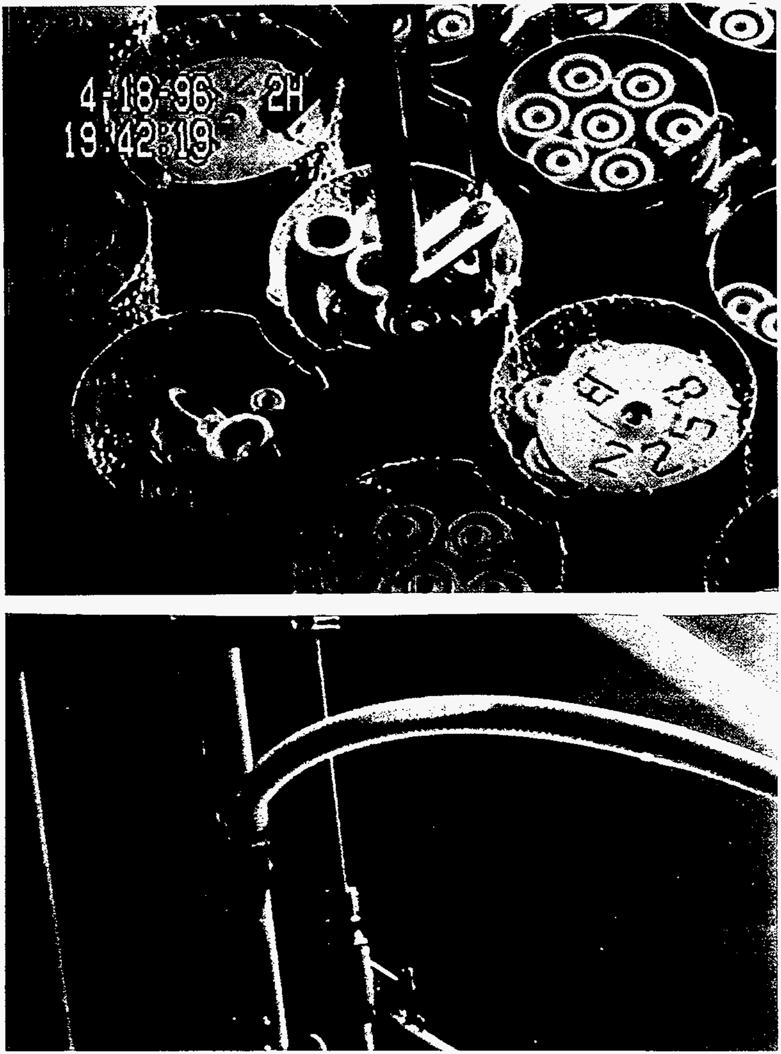

Figure 3.7. Filled Sample Canisters Stored in K East Awaiting Shipment and Sample Containers in Hanford Analytical Hot Cell.

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Figure 3.8. Map Showing Physical Location of Canisters Sampled for Sludge and Locations of Previously taken Floor Sludge Samples. Locations shown without sample numbers are samples for K East floor and Weasel Pit sludge reported previously. Canister Sample 96-15 is from roughly the same location as a floor sludge sample.

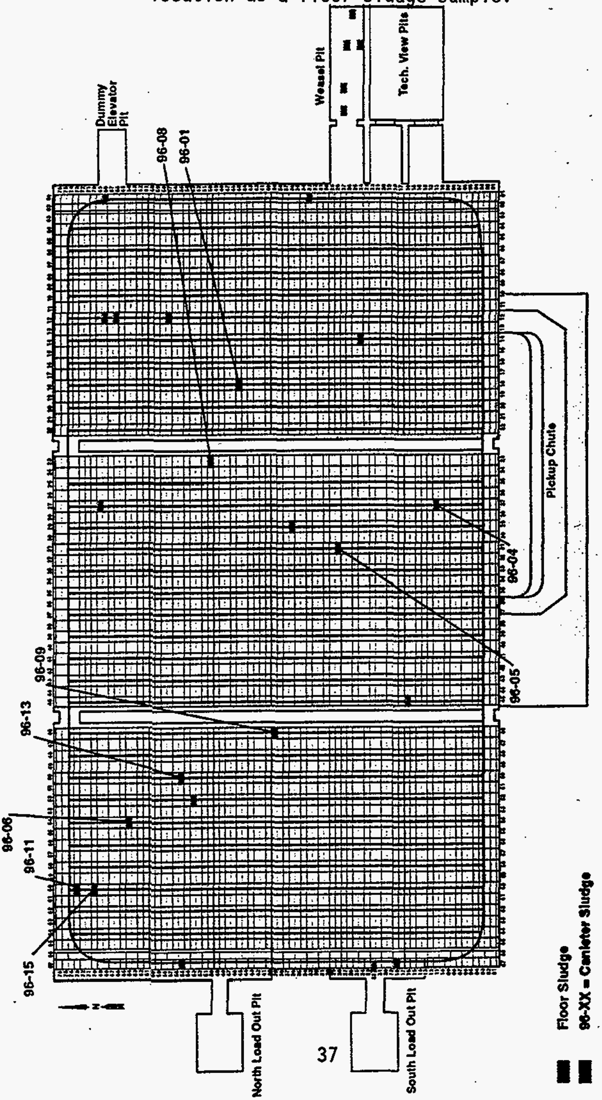

Figure 3.9. Four K East Basin Canister Sludge Samples 96-01, 96-05, 96-06, and 96-08 (Left to Right) Settling in Graduated Cylinders Hours after Agitation. (Note only the two samples on the left have settled completely.)

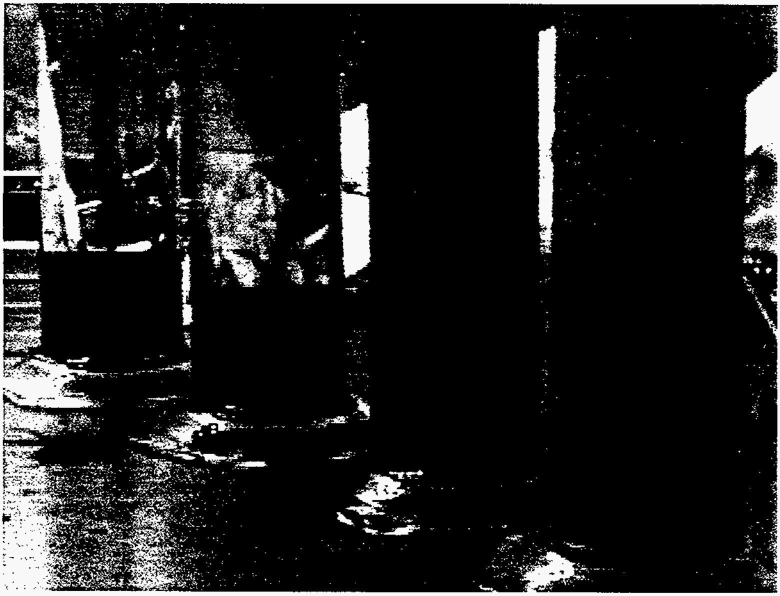

Figure 3.10. A Wedged Shaped Bubble Forming in Sample 96-05 after Sparging with Air.

Figure 3.11. Sample 96-06 after Sparging with Helium (a) Basin Water (b) Sludge Mass (c) Generated Gas and (d) Residual Sludge. Major graduations indicate 200 ml.

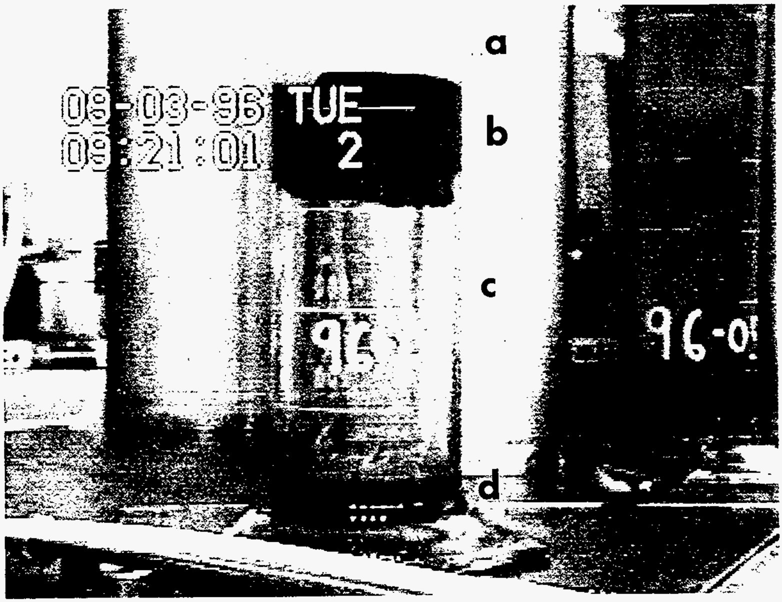

Figure 3.12. Particles Sieved from the Lowest Layer of Sample 96-06. Tyler Size 14 sieve openings are 1.18 mm (1180 μm).

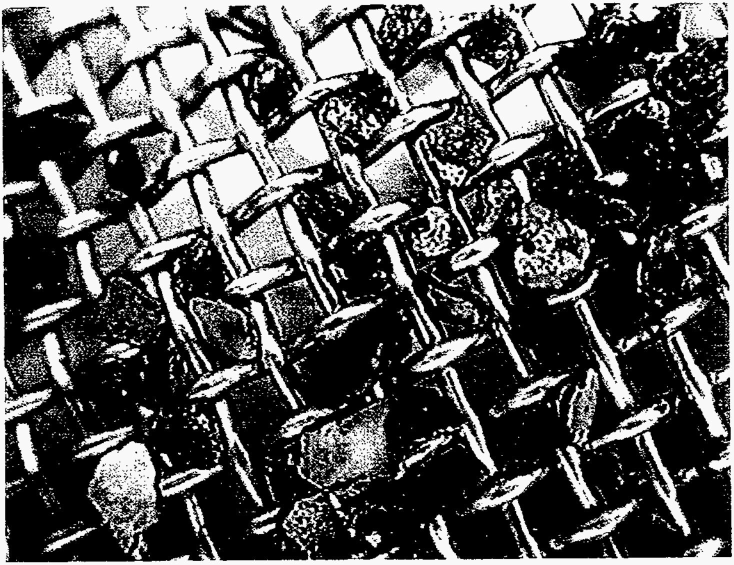

Figure 3.13. Ion Exchange (approximately 1 mm in diameter) Beads Recovered from the Lowest Layer of Sample 96-11.

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4.0 DISCUSSION

4.1 DISCUSSION OF SLUDGE CHEMISTRY

Tables 4.1 to 4.4 are summaries of the representative constituents of K East canister sludge. These values were calculated from the chemistry data listed in Appendices C, D, and E. The reader is cautioned that straight numerical averaging of the data in the appendices may not be representative of homogenous mixing of all K East Basin canister sludge since data were taken from canisters having different depths of sludge and the data presented here-in are not volume weighted.* Table 4.5 compares the most recent data with previous data from the K East floor and Weasel Pit (Makenas 1996c). The maximum fissile and fission product analyte concentrations from the current canister sludge campaign generally exceed those of the prior analyses. The reader will also find that in some cases the same type analyses were performed on portions of the same samples at both 222-S and 325 Building laboratories. In the case of total uranium, for example, four different values are available for some sludge samples. The "upper limit" columns in Tables 4.1 to 4.4 are calculated for 95% confidence that 95% of the data lie below the stated calculated value. These calculated limit concentrations may exceed what is physically plausible (e.g., greater than 100%).

The tables in this section and in the appendices do not include values for caustic demand published in Miller 1997a. Subsequent to the publication of this reference it was found that these results may have been biased low with respect to the actual caustic demand. Additions of hydroxide during the testing procedure apparently react with sludge at an unexpectedly slow rate. Evaluations of this technique and alternative procedures are continuing.

Chemistry determinations made during the campaign for K East canister sludge are not all-inclusive, with respect to the periodic table, as noted in the DQO document covering this effort (Makenas 1996a). Some process knowledge was used to justify particular analyses. For example, mercury was not determined because there was no reason to suspect mercury presence, and the cost and time for this separate analysis was significant. Cyanide was not found in floor sludge and there was no reason to suspect its presence in canister sludge.

Achieving a mass balance for the sludge analyses would be very difficult if not impossible. Sludge consists of the chemical constituents that have been analyzed (listed in the appendices). It also contains water (which is determined by TGA and other methods) from sludge which has had some chance to dry and to therefore lose water. The sludge was handled in two different hot cells and several different hoods all of which, from experience, offer a

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*A similar caution can be stated for particle size measurements. This is especially true since the size measurements are performed on separate layers of unequal thickness which in turn come from canisters containing unequal depths of sludge.

chance for sludge to dry. Finally sludge includes small residuals which could not be acid digested for chemistry analysis. For a mass balance one would also have to make an assumption on the valence state of specifically targeted elements in order to compute the weight of oxygen, hydrogen etc., which are combined with the target species. Organic compounds such as insect protein are too numerous to ever be totally identified. However, total organic carbon does provide some information in this vein.

As pointed out in the summary of this report, Sample 96-01 yielded very different chemistry results when compared to other samples. No apparent explanation for this was found, however the assumption is made that there is an inhomogeneity or dilution problem. Appendix O is a summary of sludge chemistry similar to the tables in this section except that some data from 222-S Laboratory for Sample 96-01 were not considered.

Some of the analytes chosen for this sludge analysis campaign had the specific goal of confirming that certain marker isotopes could serve as an accountability tool to track special nuclear materials (SNM) such as plutonium. Chief among these were the europium isotopes. The acceptability of these isotopes for accountability purposes remains to be determined. Regression analysis for these analytes versus plutonium concentrations is given in Appendix N. Concentrations of these marker isotopes are higher in canisters than those seen in floor sludge and thus, the statistical correlations have been extended to a greater range.

4.2 DISCUSSION OF BUBBLING SLUDGE

The color of the K East canister sludge does not in general resemble the reddish brown coloration of most previous K East basin iron-bearing floor sludge. The blacker color is probably an indication of the higher uranium content based on chemistry results from two K East floor sludge samples which had 10 to 20 wt% uranium (calculated from as-centrifuged weight) and which had an atypical (relative to other floor sludge) dark coloration. Since the collected gas from canister sludge was hydrogen with traces of fission gas, the supposition is that the uranium (and/or hydride) may be reacting with water to form uranium oxide and hydrogen. Oxygen gas which would be a marker for radiolysis, was not found but conceivably could be consumed by corrosion reactions. Calculations of the potential for radiolysis do not support the amounts of hydrogen generation seen here. The presence of uranium in metallic or hydride form has not been confirmed by X-ray diffraction studies and total uranium content has been shown to be quite high. Further oxidation of low stoichiometry oxides may also play a role in bubble formation. If reactions with metallic uranium and uranium hydride are the chief sources of hydrogen generation, it is possible that they exist as a very small number of discrete large particles which are unlikely to be part of a small subsample for techniques such as XRD. One, and only one, discrete particle was conclusively seen to generate bubbles during these examinations. This particle was identified during wet sieving and is shown in Figure 4.1. It could not be recovered for individual analysis.

The canister sludge samples continued to generate gas at a reasonably steady rate for days after resuspension for settling studies. Although the formation of very large bubbles under the sludge gives the impression of increased gas generation after sparging (with helium or air), a dramatic increase in rate seems generally not to be the case. Formation of large trapped bubbles (100 ml of more) appears to be related to a decrease in bed permeability following long term sludge settling (days) including settling of gas generating species to the bottom of the sludge bed. This behavior may have resulted in capture of the gas thus making the gas more obvious than the case where bubbles form at the bottom and quickly migrate to the sludge surface.

Canister 5465 in K East Basin was observed to be producing bubbles during a visual survey prior to the acquisition of its sludge sample, 96-06, but not at the faster rate implied by the current study. Occasionally, disturbance of other floor and canister sludge has been observed to release bubbles to the basin water. Sludge collection, sparging, and transfer operations may affect particle surface areas (by breaking agglomerated large particles or surface layers) and thus affect the reaction rates observed in the laboratory. Also, hot cells are generally warmer than the water basins, which perhaps leads to an increased reaction rate. It has also been noted by Bredt 1996, that the formation of large pockets of hydrogen may affect both the dissipation of reaction heat and the access of corroding sludge to dissolved oxygen in water both of which may influence observed reaction rate.

4.3 DISCUSSION OF PHYSICAL PROPERTIES

Sieving of sludge has shown that for the fueled canisters a significant fraction of the sample mass (approximately half) is attributable to large particles (greater that 700 μm in diameter). In Figure 4.2 these data are presented on a volume basis after approximate conversion from a mass basis using measured densities. In contrast, the iron rich sludge from nonfueled canisters apparently consists of mostly small particles. The laser scattering technique gives the particle size distribution for much finer increments of particle diameter but is only applicable to the small particles (less than 700 μm in diameter). Overlays of various particle size distributions on a number and volume basis, (from laser scattering for three samples) are also presented in Figure 4.2. It is important to understand that a very small number of large particles can and do account for a significant fraction of the volume/mass of canister sludge even though their large size and infrequent occurrence make them uncountable by the automated laser scattering technique.

As-settled densities (from sludge associated with fueled canisters) show a good correlation with uranium content i.e., higher density corresponds to more uranium which is primarily in the form of oxide. When lower densities are encountered, they are due, in part, to the presence of uranium hydrates. Metallic uranium may play some part in the highest measured sludge densities, but as discussed in Section 4.2, the contribution is not large.

Table 4.1. K East Canister Sludge Characterization Data--Per Gram Centrifuged Sludge.

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
Ag.icp.w	μg/g	2.32E+00	1.71E+02	13	6.58E+01	6.25E+01	95.0	2.33E+02
Ag.icp.wo	μg/g	2.32E+00	1.71E+02	11	7.67E+01	6.20E+01	80.8	2.51E+02
Al.icp	μg/g	9.08E+03	5.61E+04	13	2.02E+04	1.28E+04	63.4	5.44E+04
Alpha Total	μCi/g	1.14E-01	1.96E+02	13	8.87E+01	6.66E+01	75.1	2.67E+02
^{241}Am.aea.w	μCi/g	9.43E-02	8.06E+01	13	3.83E+01	2.91E+01	75.9	1.16E+02
^{241}Am.aea.wo	μCi/g	9.43E-02	8.06E+01	12	4.00E+01	2.97E+01	74.3	1.21E+02
^{241}Am.gea.w	μCi/g	< 1.26E+00	3.37E+02	13	7.20E+01	8.77E+01	121.8	3.06E+02
^{241}Am.gea.wo	μCi/g	4.70E+00	3.37E+02	12	7.79E+01	8.88E+01	114.1	3.21E+02
B.icp	μg/g	8.30E+01	1.59E+02	13	1.24E+02	2.44E+01	19.8	1.89E+02
Ba.icp.w	μg/g	< 9.95E+00	1.54E+02	13	8.04E+01	4.39E+01	54.7	1.98E+02
Ba.icp.wo	μg/g	2.91E+01	1.54E+02	10	9.14E+01	3.78E+01	41.4	2.01E+02
Be.icp	μg/g	< 1.97E+00	2.61E+01	13	NA	NA	NA	NA
Beta Total	μCi/g	1.39E+01	4.05E+03	13	1.91E+03	1.61E+03	84.3	6.21E+03
Bi.icp.w	μg/g	< 1.99E+01	5.75E+02	13	2.41E+02	2.10E+02	87.4	8.02E+02
Bi.icp.wo	μg/g	3.95E+01	5.75E+02	10	2.99E+02	2.06E+02	68.9	8.98E+02
^{212}Bi.gea	μCi/g	< 1.39E-01	< 1.68E+01	13	NA	NA	NA	NA
Br^-.ic @	μg/g	< 2.04E-01	< 1.25E+00	10	NA	NA	NA	NA
Ca.icp.w	μg/g	1.12E+02	8.59E+02	13	4.34E+02	2.42E+02	55.7	1.08E+03
Ca.icp.wo	μg/g	1.12E+02	8.59E+02	10	5.04E+02	2.33E+02	46.2	1.18E+03
Cd.icp.w	μg/g	5.98E+00	6.38E+01	13	2.14E+01	1.45E+01	67.6	6.02E+01
Cd.icp.wo	μg/g	5.98E+00	6.38E+01	11	2.35E+01	1.49E+01	63.5	6.55E+01
^{144}Ce/Pr.gea	μCi/g	< 1.51E-01	< 2.82E+01	13	NA	NA	NA	NA
Cl^-.ic @	μg/g	1.29E-01	1.05E+00	10	4.52E-01	2.72E-01	60.1	1.24E+00
$^{243/244}$Cm	μCi/g	< 1.13E-02	< 1.81E+01	13	NA	NA	NA	NA
^{57}Co.gea	μCi/g	2.30E-02	< 1.21E+01	13	NA	NA	NA	NA
^{60}Co.gea.w	μCi/g	< 3.41E-02	< 1.45E+00	13	7.04E-01	4.49E-01	63.7	1.90E+00
^{60}Co.gea.wo	μCi/g	1.25E-01	8.52E-01	7	5.18E-01	2.70E-01	52.2	1.44E+00
Cr.icp.w	μg/g	< 1.86E+01	1.77E+03	13	1.98E+02	4.76E+02	240.7	1.47E+03
Cr.icp.wo	μg/g	3.93E+01	1.77E+03	7	3.51E+02	6.29E+02	179.3	2.49E+03
^{134}Cs.gea	μCi/g	< 3.37E-02	< 2.00E+00	13	NA	NA	NA	NA
^{137}Cs.gea	μCi/g	4.87E+00	9.94E+02	13	4.29E+02	2.96E+02	69.1	1.22E+03
Cu.icp	μg/g	7.31E+00	2.60E+02	13	1.05E+02	7.24E+01	69.3	2.98E+02
^{152}Eu.gea	μCi/g	< 3.92E-02	< 6.30E+00	13	NA	NA	NA	NA

Table 4.1. K East Canister Sludge Characterization Data--Per Gram Centrifuged Sludge. (Continued)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
^{154}Eu.gea.w	µCi/g	< 1.26E-01	1.62E+01	13	6.39E+00	5.05E+00	79.0	1.99E+01
^{154}Eu.gea.wo	µCi/g	6.28E-01	1.62E+01	12	6.92E+00	4.90E+00	70.8	2.03E+01
^{155}Eu.gea	µCi/g	2.67E-01	1.06E+01	13	NA	NA	NA	NA
F^-.ic @	µg/g	< 2.45E-02	1.29E-01	10	NA	NA	NA	NA
Fe.icp	µg/g	3.53E+02	8.92E+04	13	1.78E+04	2.50E+04	140.7	8.46E+04
K.icp	µg/g	< 9.95E+01	2.76E+03	13	NA	NA	NA	NA
Mg.icp.w	µg/g	< 1.99E+01	1.93E+03	13	7.04E+02	6.92E+02	98.3	2.55E+03
Mg.icp.wo	µg/g	1.03E+02	1.93E+03	9	9.66E+02	6.82E+02	70.6	3.03E+03
Mn.icp.w	µg/g	< 1.86E+01	1.94E+02	13	5.87E+01	5.54E+01	94.4	2.07E+02
Mn.icp.wo	µg/g	3.41E+01	1.94E+02	7	9.19E+01	5.78E+01	62.9	2.89E+02
NO_2^-.ic @	µg/g	< 1.76E-01	< 1.08E+00	10	NA	NA	NA	NA
NO_3^-.ic @	µg/g	< 2.26E-01	1.67E+00	10	NA	NA	NA	NA
Na.icp	µg/g	1.58E+02	1.92E+05	13	1.50E+04	5.32E+04	353.3	1.57E+05
^{94}Nb.gea	µCi/g	< 2.09E-02	< 1.57E+00	13	NA	NA	NA	NA
Ni.icp.w	µg/g	2.03E+01	1.17E+02	13	4.91E+01	2.71E+01	55.2	1.21E+02
Ni.icp.wo	µg/g	2.03E+01	1.17E+02	7	5.70E+01	3.61E+01	63.4	1.80E+02
^{237}Np.w	µCi/g	< 3.81E-03	1.51E-02	13	8.45E-03	4.11E-03	48.6	1.94E-02
^{237}Np.wo	µCi/g	4.07E-03	1.51E-02	8	1.04E-02	4.11E-03	39.7	2.35E-02
Oxalate.ic @	µg/g	< 1.72E-01	< 1.05E+00	10	NA	NA	NA	NA
P.icp	µg/g	< 7.90E+01	1.70E+03	13	NA	NA	NA	NA
PO_4^{3-}.ic @	µg/g	< 1.95E-01	< 1.20E+00	10	NA	NA	NA	NA
Pb.icp.w	µg/g	5.20E+01	< 2.12E+02	13	1.42E+02	6.54E+01	45.9	3.17E+02
Pb.icp.wo	µg/g	5.20E+01	1.79E+02	7	9.34E+01	4.94E+01	52.9	2.61E+02
^{238}Pu.w	µCi/g	4.14E-03	< 5.88E+01	13	1.17E+01	1.52E+01	130.0	5.24E+01
^{238}Pu.wo	µCi/g	4.14E-03	1.46E+01	12	7.80E+00	5.92E+00	75.8	2.40E+01
$^{239/240}$Pu	µCi/g	3.85E-02	1.33E+02	13	5.50E+01	4.33E+01	78.7	1.71E+02
^{226}Ra.gea	µCi/g	< 3.02E-01	< 5.28E+01	13	NA	NA	NA	NA
^{106}Ru/Rh.gea	µCi/g	< 2.37E-01	< 3.90E+01	13	NA	NA	NA	NA
SO_4^{2-}.ic.w @	µg/g	7.17E-01	3.57E+00	10	1.69E+00	8.63E-01	51.2	4.20E+00
SO_4^{2-}.ic.wo @	µg/g	7.17E-01	3.57E+00	8	1.80E+00	9.35E-01	51.9	4.78E+00
Se.icp	µg/g	< 1.99E+01	< 2.12E+02	13	NA	NA	NA	NA
Sm.icp	µg/g	< 1.99E+01	< 2.12E+02	13	NA	NA	NA	NA
$^{89/90}$Sr	µCi/g	3.62E+00	1.91E+03	13	8.07E+02	7.51E+02	93.1	2.81E+03

Table 4.1. K East Canister Sludge Characterization Data--
Per Gram Centrifuged Sludge. (Continued)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
TIC	µg/g	2.51E+02	1.10E+04	13	1.49E+03	2.91E+03	195.7	9.25E+03
TOC	µg/g	3.73E+02	1.05E+04	13	1.44E+03	2.75E+03	191.4	8.78E+03
^{99}Tc	µCi/g	2.26E-01	1.27E+01	6	5.56E+00	4.66E+00	83.7	2.28E+01
Tl.icp	µg/g	< 3.98E+01	< 4.25E+02	13	NA	NA	NA	NA
^{208}Tl.gea	µCi/g	< 2.00E-01	< 3.32E+01	13	NA	NA	NA	NA
Total Carbon	µg/g	5.52E+02	2.28E+04	13	2.96E+03	5.99E+03	202.5	1.89E+04
U.icp.w	µg/g	< 1.97E+02	6.52E+05	13	3.20E+05	2.91E+05	90.7	1.10E+06
U.icp.wo	µg/g	1.32E+04	6.52E+05	12	3.47E+05	2.86E+05	82.6	1.13E+06
U.las	µg/g	3.24E+04	7.68E+05	10	4.88E+05	2.83E+05	57.9	1.31E+06
U.phos	µg/g	7.12E+02	6.18E+05	13	2.65E+05	2.41E+05	90.8	9.08E+05
^{233}U.tims &	µg/g	< 1.59E-01	< 3.76E+00	10	NA	NA	NA	NA
^{234}U.tims &	µg/g	4.71E+00	7.08E+01	10	3.99E+01	2.49E+01	62.5	1.13E+02
^{235}U.tims &	µg/g	2.32E+02	5.96E+03	10	3.44E+03	2.12E+03	61.7	9.63E+03
^{236}U.tims &	µg/g	2.46E+01	6.05E+02	10	3.13E+02	1.83E+02	58.5	8.46E+02
^{238}U.tims &	µg/g	3.22E+04	7.63E+05	10	4.84E+05	2.81E+05	58.0	1.30E+06
%Water.grav	wt%	1.47E+01	9.01E+01	12	4.61E+01	2.96E+01	64.2	1.27E+02
Water.grav	µg/g	1.47E+05	9.01E+05	12	4.61E+05	2.96E+05	64.2	1.27E+06
Zn.icp.w	µg/g	1.85E+01	4.95E+02	13	1.19E+02	1.38E+02	116.3	4.88E+02
Zn.icp.wo	µg/g	1.85E+01	4.95E+02	11	1.37E+02	1.44E+02	105.0	5.41E+02
Zr.icp.w	µg/g	9.19E+00	2.18E+02	13	7.33E+01	6.53E+01	89.2	2.48E+02
Zr.icp.wo	µg/g	9.19E+00	2.18E+02	8	1.07E+02	6.34E+01	59.5	3.09E+02
pH @	pH units	7.07E+00	8.38E+00	7	7.74E+00	5.11E-01	6.6	9.47E+00

*: Per gram centrifuged sludge.
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: Tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.w: Less than values were included (i.e., 3 for <3) when calculating the summary statistics.
.wo: Less than values were deleted when calculating the summary statistics.
NA: Not applicable for these data.
@: Analyses performed using sludge supernate.
&: Calculated using the PNNL total uranium (laser fluoresence) data.

Table 4.2. K East Canister Sludge Characterization Data--
Per Gram As-Settled Sludge.

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
Ag.icp.w	µg/g	1.23E+00	1.52E+02	13	5.47E+01	5.64E+01	103.1	2.05E+02
Ag.icp.wo	µg/g	1.23E+00	1.52E+02	11	6.39E+01	5.67E+01	88.6	2.23E+02
Al.icp	µg/g	5.23E+03	3.60E+04	13	1.43E+04	7.98E+03	55.7	3.57E+04
Al.icp/ms	µg/g	7.79E+02	8.16E+03	2	4.47E+03	5.22E+03	116.8	NA
Alpha Total	µCi/g	1.01E-01	1.78E+02	13	6.97E+01	5.86E+01	84.0	2.26E+02
Alpha Total.PNNL	µCi/g	1.06E+02	2.14E+02	2	1.60E+02	7.63E+01	47.7	NA
^{241}Am.aea.w	µCi/g	8.39E-02	7.35E+01	13	2.93E+01	2.45E+01	83.5	9.47E+01
^{241}Am.aea.wo	µCi/g	8.39E-02	7.35E+01	12	3.04E+01	2.52E+01	83.0	9.94E+01
^{241}Am.gea.w	µCi/g	< 1.12E+00	2.23E+02	13	5.38E+01	6.04E+01	112.2	2.15E+02
^{241}Am.gea.wo	µCi/g	2.50E+00	2.23E+02	12	5.82E+01	6.09E+01	104.6	2.25E+02
^{241}Am.gea.PNNL	µCi/g	2.15E-01	5.27E+01	2	2.65E+01	3.71E+01	140.3	NA
B.icp	µg/g	5.33E+01	1.41E+02	13	9.41E+01	3.37E+01	35.8	1.84E+02
Ba.icp.w	µg/g	< 5.30E+00	1.37E+02	13	6.24E+01	4.03E+01	64.7	1.70E+02
Ba.icp.wo	µg/g	1.86E+01	1.37E+02	10	7.04E+01	3.82E+01	54.3	1.82E+02
Be.icp	µg/g	< 1.75E+00	1.81E+01	13	NA	NA	NA	NA
Beta Total	µCi/g	7.43E+00	3.70E+03	13	1.56E+03	1.45E+03	92.9	5.42E+03
Beta Total.PNNL	µCi/g	3.90E+03	8.67E+03	2	6.29E+03	3.38E+03	53.7	NA
Bi.icp.w	µg/g	< 1.06E+01	5.24E+02	13	1.90E+02	1.79E+02	94.0	6.68E+02
Bi.icp.wo	µg/g	2.53E+01	5.24E+02	10	2.37E+02	1.79E+02	75.2	7.57E+02
^{212}Bi.gea	µCi/g	< 8.08E-02	< 1.42E+01	13	NA	NA	NA	NA
Br^-.ic	µg/g	< 3.09E-01	< 1.31E+00	10	NA	NA	NA	NA
Ca.icp.w	µg/g	5.96E+01	6.19E+02	13	3.19E+02	1.83E+02	57.4	8.08E+02
Ca.icp.wo	µg/g	5.96E+01	6.19E+02	10	3.62E+02	1.89E+02	52.2	9.13E+02
Ca.icp/ms	µg/g	4.85E+01	8.62E+01	2	6.74E+01	2.66E+01	39.5	NA
Cd.icp.w	µg/g	5.32E+00	4.42E+01	13	1.55E+01	9.98E+00	64.6	4.21E+01
Cd.icp.wo	µg/g	5.32E+00	4.42E+01	11	1.69E+01	1.02E+01	60.1	4.56E+01
^{144}Ce/Pr.gea	µCi/g	< 8.69E-02	< 2.52E+01	13	NA	NA	NA	NA
Cl^-.ic	µg/g	1.90E-01	1.69E+00	10	6.04E-01	4.50E-01	74.6	1.91E+00
$^{243/244}$Cm	µCi/g	< 1.01E-02	< 1.63E+01	13	NA	NA	NA	NA
^{57}Co.gea	µCi/g	1.51E-02	< 6.41E+00	13	NA	NA	NA	NA
^{60}Co.gea.w	µCi/g	< 3.05E-02	< 1.30E+00	13	5.40E-01	3.88E-01	71.8	1.58E+00
^{60}Co.gea.wo	µCi/g	6.67E-02	7.09E-01	7	3.63E-01	2.26E-01	62.3	1.13E+00
^{60}Co.gea.PNNL	µCi/g	1.13E-02	3.18E-01	2	1.65E-01	2.17E-01	131.7	NA

Table 4.2. K East Canister Sludge Characterization Data--
Per Gram As-Settled Sludge. (Continued)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
Cr.icp.w	µg/g	< 1.17E+01	1.57E+03	13	1.63E+02	4.26E+02	261.3	1.30E+03
Cr.icp.wo	µg/g	2.28E+01	1.57E+03	7	2.88E+02	5.68E+02	196.9	2.22E+03
^{134}Cs.gea	µCi/g	< 2.20E-02	< 1.79E+00	13	NA	NA	NA	NA
^{134}Cs.gea.PNNL	µCi/g	< 2.05E-03	1.91E-01	2	9.66E-02	NA	NA	NA
^{137}Cs.gea	µCi/g	2.63E+00	9.10E+02	13	3.40E+02	2.66E+02	78.2	1.05E+03
^{137}Cs.gea.PNNL	µCi/g	7.99E-01	4.70E+02	2	2.36E+02	3.32E+02	140.9	NA
Cu.icp	µg/g	6.50E+00	1.80E+02	13	7.29E+01	4.65E+01	63.8	1.97E+02
^{152}Eu.gea	µCi/g	< 2.38E-02	< 5.14E+00	13	NA	NA	NA	NA
^{154}Eu.gea.w	µCi/g	< 1.12E-01	1.48E+01	13	4.98E+00	4.33E+00	86.9	1.65E+01
^{154}Eu.gea.wo	µCi/g	3.34E-01	1.48E+01	12	5.39E+00	4.25E+00	79.0	1.70E+01
^{154}Eu.gea.PNNL	µCi/g	2.99E-02	7.59E+00	2	3.81E+00	5.34E+00	140.3	NA
^{155}Eu.gea	µCi/g	1.43E-01	9.68E+00	13	NA	NA	NA	NA
^{155}Eu.gea.PNNL	µCi/g	1.24E-02	3.83E+00	2	1.92E+00	2.70E+00	140.5	NA
F^-.ic	µg/g	< 3.61E-02	2.07E-01	10	NA	NA	NA	NA
Fe.icp	µg/g	3.14E+02	6.17E+04	13	1.20E+04	1.70E+04	142.6	5.75E+04
Fe.icp/ms	µg/g	1.15E+03	4.95E+03	2	3.05E+03	2.69E+03	88.2	NA
K.icp	µg/g	< 5.30E+01	2.46E+03	13	NA	NA	NA	NA
Mg.icp.w	µg/g	< 1.06E+01	1.76E+03	13	5.55E+02	6.14E+02	110.7	2.19E+03
Mg.icp.wo	µg/g	7.14E+01	1.76E+03	9	7.57E+02	6.42E+02	84.8	2.70E+03
Mn.icp.w	µg/g	< 1.17E+01	1.34E+02	13	4.14E+01	3.87E+01	93.6	1.45E+02
Mn.icp.wo	µg/g	2.10E+01	1.34E+02	7	6.27E+01	4.30E+01	68.6	2.09E+02
NO_2^-.ic	µg/g	< 2.66E-01	< 1.13E+00	10	NA	NA	NA	NA
NO_3^-.ic	µg/g	< 3.43E-01	1.83E+00	10	NA	NA	NA	NA
Na.icp	µg/g	9.20E+01	1.71E+05	13	1.33E+04	4.73E+04	354.4	1.40E+05
^{94}Nb.gea	µCi/g	< 1.16E-02	< 1.43E+00	13	NA	NA	NA	NA
Ni.icp.w	µg/g	1.07E+01	1.04E+02	13	3.70E+01	2.33E+01	63.1	9.92E+01
Ni.icp.wo	µg/g	1.07E+01	1.04E+02	7	4.03E+01	3.22E+01	80.0	1.50E+02
^{237}Np.w	µCi/g	2.59E-03	1.37E-02	13	6.73E-03	3.69E-03	54.9	1.66E-02
^{237}Np.wo	µCi/g	2.59E-03	1.37E-02	8	8.41E-03	3.77E-03	44.9	2.04E-02
Oxalate.ic	µg/g	< 2.60E-01	< 1.11E+00	10	NA	NA	NA	NA
P.icp	µg/g	< 4.60E+01	1.51E+03	13	NA	NA	NA	NA
PO_4^{3-}.ic	µg/g	< 2.96E-01	< 1.26E+00	10	NA	NA	NA	NA

Table 4.2. K East Canister Sludge Characterization Data--
Per Gram As-Settled Sludge. (Continued)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
Pb.icp.w	µg/g	3.35E+01	< 1.82E+02	13	1.10E+02	6.10E+01	55.7	2.72E+02
Pb.icp.wo	µg/g	3.35E+01	1.24E+02	7	6.17E+01	3.41E+01	55.3	1.78E+02
^{238}Pu.w	µCi/g	3.70E-03	< 5.30E+01	13	9.51E+00	1.39E+01	145.9	4.66E+01
^{238}Pu.wo	µCi/g	3.70E-03	1.29E+01	12	5.89E+00	4.92E+00	83.5	1.93E+01
$^{239/240}$Pu	µCi/g	3.42E-02	1.21E+02	13	4.36E+01	3.85E+01	88.3	1.47E+02
^{239}Pu.icp/ms	µCi/g	1.07E+01	7.71E+01	2	4.39E+01	4.70E+01	107.0	NA
^{240}Pu.icp/ms	µCi/g	< 3.11E-02	3.09E+01	2	1.55E+01	NA	NA	NA
^{226}Ra.gea	µCi/g	< 1.73E-01	< 4.73E+01	13	NA	NA	NA	NA
^{106}Ru/Rh.gea	µCi/g	< 1.37E-01	< 3.53E+01	13	NA	NA	NA	NA
SO$_4^{2-}$.ic.w	µg/g	1.09E+00	4.18E+00	10	2.13E+00	9.30E-01	43.7	4.83E+00
SO$_4^{2-}$.ic.wo	µg/g	1.09E+00	4.18E+00	8	2.33E+00	9.38E-01	40.3	5.32E+00
^{125}Sb.gea.PNNL	µCi/g	< 1.37E-02	8.78E-01	2	4.46E-01	NA	NA	NA
Se.icp	µg/g	< 1.06E+01	< 1.82E+02	13	NA	NA	NA	NA
Sm.icp	µg/g	< 1.06E+01	< 1.82E+02	13	NA	NA	NA	NA
$^{89/90}$Sr	µCi/g	1.93E+00	1.74E+03	13	6.65E+02	6.68E+02	100.5	2.45E+03
TIC	µg/g	1.55E+02	9.79E+03	13	1.19E+03	2.60E+03	218.8	8.14E+03
TOC	µg/g	2.67E+02	9.34E+03	13	1.17E+03	2.47E+03	209.9	7.76E+03
^{99}Tc	µCi/g	2.01E-01	8.41E+00	6	4.29E+00	3.40E+00	79.4	1.69E+01
Tl.icp	µg/g	< 2.12E+01	< 3.65E+02	13	NA	NA	NA	NA
^{208}Tl.gea	µCi/g	< 1.16E-01	< 3.02E+01	13	NA	NA	NA	NA
Total Carbon	µg/g	4.36E+02	2.02E+04	13	2.41E+03	5.38E+03	223.4	1.68E+04
U.icp.w	µg/g	< 1.75E+02	5.94E+05	13	2.61E+05	2.55E+05	98.0	9.43E+05
U.icp.wo	µg/g	6.99E+03	5.94E+05	12	2.82E+05	2.54E+05	90.0	9.77E+05
U.las	µg/g	2.25E+04	7.00E+05	10	4.18E+05	2.71E+05	64.7	1.21E+06
U.phos	µg/g	6.45E+02	5.57E+05	13	2.16E+05	2.14E+05	98.9	7.87E+05
U.icp/ms	µg/g	4.44E+05	5.10E+05	2	4.77E+05	4.70E+04	9.9	NA
^{233}U.tims &	µg/g	< 1.10E-01	< 3.43E+00	10	NA	NA	NA	NA
^{233}U.icp/ms	µg/g	1.59E-01	2.97E-01	2	2.28E-01	9.77E-02	42.9	NA
^{234}U.tims &	µg/g	3.26E+00	6.45E+01	10	3.41E+01	2.33E+01	68.3	1.02E+02
^{234}U.icp/ms	µg/g	2.79E+01	3.38E+01	2	3.09E+01	4.16E+00	13.5	NA
^{235}U.tims &	µg/g	1.61E+02	5.30E+03	10	2.95E+03	2.00E+03	67.8	8.79E+03
^{235}U.icp/ms	µg/g	2.64E+03	4.31E+03	2	3.48E+03	1.18E+03	34.0	NA
^{236}U.tims &	µg/g	1.71E+01	5.51E+02	10	2.63E+02	1.71E+02	64.8	7.60E+02
^{236}U.icp/ms	µg/g	2.30E+02	2.53E+02	2	2.41E+02	1.63E+01	6.8	NA

Table 4.2. K East Canister Sludge Characterization Data--
Per Gram As-Settled Sludge. (Continued)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
^{238}U.tims &	µg/g	2.23E+04	6.96E+05	10	4.15E+05	2.68E+05	64.7	1.20E+06
^{238}U.icp/ms	µg/g	4.38E+05	5.04E+05	2	4.71E+05	4.69E+04	10.0	NA
^{238}U.gea.PNNL	µg/g	1.82E+05	< 1.18E+07	2	5.99E+06	NA	NA	NA
Water.grav	µg/g	2.23E+05	9.48E+05	12	5.55E+05	2.91E+05	52.5	1.35E+06
%Water.grav	wt%	2.23E+01	9.48E+01	12	5.55E+01	2.91E+01	52.5	1.35E+02
Zn.icp.w	µg/g	1.19E+01	3.43E+02	13	7.99E+01	9.28E+01	116.2	3.28E+02
Zn.icp.wo	µg/g	1.19E+01	3.43E+02	11	9.12E+01	9.70E+01	106.4	3.64E+02
Zr.icp.w	µg/g	8.17E+00	1.40E+02	13	4.88E+01	3.99E+01	81.7	1.55E+02
Zr.icp.wo	µg/g	8.17E+00	1.40E+02	8	6.92E+01	3.87E+01	56.0	1.93E+02
Zr.icp/ms	µg/g	1.04E+02	2.20E+03	2	1.15E+03	1.48E+03	128.6	NA
m/z241.icp/ms	µg/g	< 1.37E-01	4.00E+01	2	2.00E+01	NA	NA	NA

*: Per gram as-settled sludge
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: Tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.w: Less than values were included (i.e., 3 for <3) when calculating the summary statistics.
.wo: Less than values were deleted when calculating the summary statistics.
NA: Not applicable for these data.
&: Calculated using the PNNL total uranium (laser fluoresence) data.

Table 4.3. K East Canister Sludge Characterization Data--
Per ml As-Settled Sludge.

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
Ag.icp.W	µg/ml	1.31E+00	3.79E+02	13	1.19E+02	1.37E+02	115.3	4.84E+02
Ag.icp.WO	µg/ml	1.31E+00	3.79E+02	11	1.39E+02	1.39E+02	100.0	5.31E+02
Al.icp	µg/ml	7.40E+03	5.26E+04	13	2.34E+04	1.25E+04	53.6	5.67E+04
Al.icp/ms	µg/ml	1.62E+03	1.91E+04	2	1.03E+04	1.23E+04	119.2	NA
Alpha Total	µCi/ml	2.12E-01	4.17E+02	13	1.44E+02	1.52E+02	105.4	5.48E+02
Alpha Total.PNNL	µCi/ml	2.21E+02	5.00E+02	2	3.60E+02	1.97E+02	54.7	NA
^{241}Am.aea.W	µCi/ml	1.75E-01	1.80E+02	13	5.79E+01	6.15E+01	106.3	2.22E+02
^{241}Am.aea.WO	µCi/ml	1.75E-01	1.80E+02	12	5.94E+01	6.40E+01	107.8	2.35E+02
^{241}Am.gea.W	µCi/ml	< 2.34E+00	2.65E+02	13	9.72E+01	9.71E+01	99.9	3.56E+02
^{241}Am.gea.WO	µCi/ml	2.66E+00	2.65E+02	12	1.05E+02	9.69E+01	92.2	3.70E+02
^{241}Am.gea.PNNL	µCi/ml	4.49E-01	1.23E+02	2	6.18E+01	8.68E+01	140.4	NA
B.icp	µg/ml	5.68E+01	3.64E+02	13	1.76E+02	1.13E+02	63.9	4.77E+02
Ba.icp.W	µg/ml	< 5.64E+00	2.97E+02	13	1.22E+02	1.04E+02	85.3	3.99E+02
Ba.icp.WO	µg/ml	2.00E+01	2.97E+02	10	1.38E+02	1.08E+02	78.1	4.52E+02
Be.icp	µg/ml	< 3.66E+00	< 2.72E+01	13	NA	NA	NA	NA
Beta Total	µCi/ml	7.91E+00	1.08E+04	13	3.34E+03	3.75E+03	112.1	1.33E+04
Beta Total.PNNL	µCi/ml	9.11E+03	1.81E+04	2	1.36E+04	6.35E+03	46.7	NA
Bi.icp.W	µg/ml	< 1.13E+01	1.23E+03	13	3.92E+02	4.35E+02	111.1	1.55E+03
Bi.icp.WO	µg/ml	2.71E+01	1.23E+03	10	4.94E+02	4.49E+02	90.9	1.80E+03
^{212}Bi.gea	µCi/ml	< 8.61E-02	< 4.26E+01	13	NA	NA	NA	NA
Br$^-$.ic	µg/ml	< 6.97E-01	< 1.40E+00	10	NA	NA	NA	NA
Ca.icp.W	µg/ml	6.34E+01	1.45E+03	13	5.62E+02	4.11E+02	73.1	1.66E+03
Ca.icp.WO	µg/ml	6.34E+01	1.45E+03	10	6.11E+02	4.58E+02	74.9	1.94E+03
Ca.icp/ms	µg/ml	1.01E+02	2.02E+02	2	1.51E+02	7.09E+01	46.9	NA
Cd.icp.W	µg/ml	< 8.29E+00	5.69E+01	13	2.62E+01	1.72E+01	65.8	7.21E+01
Cd.icp.WO	µg/ml	9.78E+00	5.69E+01	11	2.87E+01	1.75E+01	61.0	7.80E+01
^{144}Ce/Pr.gea	µCi/ml	< 9.25E-02	< 7.07E+01	13	NA	NA	NA	NA
Cl$^-$.ic	µg/ml	3.47E-01	3.25E+00	10	1.08E+00	1.13E+00	104.9	4.39E+00
$^{243/244}$Cm	µCi/ml	< 2.10E-02	< 4.33E+01	13	NA	NA	NA	NA
^{57}Co.gea	µCi/ml	2.21E-02	< 1.46E+01	13	NA	NA	NA	NA
^{60}Co.gea.W	µCi/ml	< 6.36E-02	< 3.46E+00	13	1.05E+00	1.02E+00	96.7	3.77E+00
^{60}Co.gea.WO	µCi/ml	7.11E-02	1.66E+00	7	5.63E-01	5.42E-01	96.3	2.40E+00
^{60}Co.gea.PNNL	µCi/ml	2.36E-02	7.44E-01	2	3.84E-01	5.09E-01	132.7	NA
Cr.icp.W	µg/ml	< 1.66E+01	3.27E+03	13	3.10E+02	8.93E+02	287.5	2.69E+03
Cr.icp.WO	µg/ml	2.49E+01	3.27E+03	7	5.45E+02	1.21E+03	221.2	4.64E+03

Table 4.3. K East Canister Sludge Characterization Data--
Per ml As-Settled Sludge. (Continued)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
^{134}Cs.gea	µCi/ml	< 2.36E-02	< 5.05E+00	13	NA	NA	NA	NA
^{134}Cs.gea.PNNL	µCi/ml	< 4.27E-03	4.47E-01	2	2.25E-01	NA	NA	NA
^{137}Cs.gea	µCi/ml	2.80E+00	2.13E+03	13	6.90E+02	6.90E+02	99.9	2.53E+03
^{137}Cs.gea.PNNL	µCi/ml	1.67E+00	1.10E+03	2	5.51E+02	7.76E+02	141.0	NA
Cu.icp	µg/ml	1.36E+01	2.23E+02	13	1.14E+02	6.03E+01	52.8	2.75E+02
^{152}Eu.gea	µCi/ml	< 2.53E-02	< 1.54E+01	13	NA	NA	NA	NA
^{154}Eu.gea.W	µCi/ml	< 2.35E-01	3.45E+01	13	1.01E+01	1.08E+01	106.6	3.88E+01
^{154}Eu.gea.WO	µCi/ml	3.56E-01	3.45E+01	12	1.09E+01	1.08E+01	98.9	4.05E+01
^{154}Eu.gea.PNNL	µCi/ml	6.24E-02	1.77E+01	2	8.90E+00	1.25E+01	140.4	NA
^{155}Eu.gea	µCi/ml	1.52E-01	2.26E+01	13	NA	NA	NA	NA
^{155}Eu.gea.PNNL	µCi/ml	2.59E-02	8.96E+00	2	4.49E+00	6.32E+00	140.6	NA
F^-.ic	µg/ml	< 6.66E-02	3.97E-01	10	NA	NA	NA	NA
Fe.icp	µg/ml	6.54E+02	7.66E+04	13	1.57E+04	2.06E+04	131.4	7.07E+04
Fe.icp/ms	µg/ml	2.39E+03	1.16E+04	2	6.98E+03	6.48E+03	92.9	NA
K.icp	µg/ml	< 5.64E+01	5.12E+03	13	NA	NA	NA	NA
Mg.icp.W	µg/ml	< 1.13E+01	4.10E+03	13	1.09E+03	1.41E+03	129.9	4.86E+03
Mg.icp.WO	µg/ml	8.85E+01	4.10E+03	9	1.47E+03	1.57E+03	106.8	6.21E+03
Mn.icp.W	µg/ml	< 1.66E+01	1.99E+02	13	6.36E+01	5.66E+01	89.0	2.15E+02
Mn.icp.WO	µg/ml	2.29E+01	1.99E+02	7	8.67E+01	7.01E+01	80.8	3.25E+02
NO_2^-.ic	µg/ml	< 6.01E-01	< 1.21E+00	10	NA	NA	NA	NA
NO_3^-.ic	µg/ml	< 7.72E-01	1.99E+00	10	NA	NA	NA	NA
Na.icp	µg/ml	9.80E+01	3.56E+05	13	2.78E+04	9.87E+04	355.0	2.91E+05
^{94}Nb.gea	µCi/ml	< 1.23E-02	< 4.28E+00	13	NA	NA	NA	NA
Ni.icp.W	µg/ml	1.14E+01	2.16E+02	13	6.65E+01	5.30E+01	79.7	2.08E+02
Ni.icp.WO	µg/ml	1.14E+01	2.16E+02	7	6.09E+01	7.07E+01	116.0	3.01E+02
^{237}Np.W	µCi/ml	2.76E-03	3.36E-02	13	1.29E-02	1.03E-02	79.7	4.05E-02
^{237}Np.WO	µCi/ml	2.76E-03	3.36E-02	8	1.75E-02	1.07E-02	61.0	5.17E-02
Oxalate.ic	µg/ml	< 5.86E-01	< 1.18E+00	10	NA	NA	NA	NA
P.icp	µg/ml	< 5.01E+01	3.15E+03	13	NA	NA	NA	NA
PO_4^{3-}.ic	µg/ml	< 6.66E-01	< 1.34E+00	10	NA	NA	NA	NA
Pb.icp.W	µg/ml	3.57E+01	< 5.45E+02	13	2.12E+02	1.72E+02	81.4	6.71E+02
Pb.icp.WO	µg/ml	3.57E+01	1.54E+02	7	7.96E+01	4.20E+01	52.7	2.22E+02
^{238}Pu.W	µCi/ml	7.71E-03	< 1.30E+02	13	2.07E+01	3.51E+01	169.8	1.14E+02
^{238}Pu.WO	µCi/ml	7.71E-03	3.81E+01	12	1.15E+01	1.27E+01	109.7	4.61E+01
$^{239/240}$Pu	µCi/ml	7.14E-02	2.84E+02	13	9.01E+01	9.64E+01	107.0	3.47E+02

Table 4.3. K East Canister Sludge Characterization Data--
Per ml As-Settled Sludge. (Continued)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit \$
^{239}Pu.icp/ms	µCi/ml	2.23E+01	1.80E+02	2	1.01E+02	1.12E+02	110.2	NA
^{240}Pu.icp/ms	µCi/ml	< 6.47E-02	7.23E+01	2	3.62E+01	NA	NA	NA
^{226}Ra.gea	µCi/ml	< 1.85E-01	< 1.39E+02	13	NA	NA	NA	NA
^{106}Ru/Rh.gea	µCi/ml	< 1.46E-01	< 1.06E+02	13	NA	NA	NA	NA
SO_4^{2-}.ic.w	µg/ml	< 1.50E+00	5.20E+00	10	3.28E+00	1.39E+00	42.4	7.31E+00
SO_4^{2-}.ic.wo	µg/ml	2.66E+00	5.20E+00	8	3.71E+00	1.17E+00	31.6	7.45E+00
^{125}Sb.gea.PNNL	µCi/ml	< 2.85E-02	2.05E+00	2	1.04E+00	NA	NA	NA
Se.icp	µg/ml	< 1.13E+01	< 5.45E+02	13	NA	NA	NA	NA
Sm.icp	µg/ml	< 1.13E+01	< 5.45E+02	13	NA	NA	NA	NA
$^{89/90}$Sr	µCi/ml	2.05E+00	4.71E+03	13	1.45E+03	1.69E+03	117.1	5.97E+03
TIC	µg/ml	2.20E+02	2.04E+04	13	2.24E+03	5.48E+03	244.8	1.69E+04
TOC	µg/ml	3.09E+02	1.95E+04	13	2.23E+03	5.20E+03	233.2	1.61E+04
^{99}Tc	µCi/ml	4.19E-01	1.89E+01	6	7.78E+00	7.13E+00	91.7	3.42E+01
Tl.icp	µg/ml	< 2.26E+01	< 1.09E+03	13	NA	NA	NA	NA
^{208}Tl.gea	µCi/ml	< 1.23E-01	< 9.04E+01	13	NA	NA	NA	NA
Total Carbon	µg/ml	5.47E+02	4.22E+04	13	4.57E+03	1.13E+04	247.7	3.48E+04
U.icp.w	µg/ml	< 3.66E+02	1.76E+06	13	5.61E+05	6.41E+05	114.3	2.27E+06
U.icp.wo	µg/ml	7.45E+03	1.76E+06	12	6.07E+05	6.46E+05	106.3	2.37E+06
U.las	µg/ml	2.79E+04	2.00E+06	10	9.13E+05	7.17E+05	78.5	3.00E+06
U.phos	µg/ml	1.34E+03	1.44E+06	13	4.68E+05	5.44E+05	116.4	1.92E+06
U.icp/ms	µg/ml	9.26E+05	1.19E+06	2	1.06E+06	1.89E+05	17.8	NA
^{233}U.tims &	µg/ml	< 1.36E-01	< 9.80E+00	10	NA	NA	NA	NA
^{233}U.icp/ms	µg/ml	3.71E-01	6.20E-01	2	4.96E-01	1.76E-01	35.4	NA
^{234}U.tims &	µg/ml	4.04E+00	1.93E+02	10	7.51E+01	6.31E+01	84.1	2.59E+02
^{234}U.icp/ms	µg/ml	6.53E+01	7.05E+01	2	6.79E+01	3.70E+00	5.4	NA
^{235}U.tims &	µg/ml	2.00E+02	1.48E+04	10	6.46E+03	5.19E+03	80.3	2.16E+04
^{235}U.icp/ms	µg/ml	6.18E+03	9.00E+03	2	7.59E+03	1.99E+03	26.3	NA
^{236}U.tims &	µg/ml	2.11E+01	1.65E+03	10	5.76E+02	4.98E+02	86.5	2.03E+03
^{236}U.icp/ms	µg/ml	5.27E+02	5.37E+02	2	5.32E+02	6.85E+00	1.3	NA
^{238}U.tims &	µg/ml	2.76E+04	1.98E+06	10	9.06E+05	7.11E+05	78.5	2.98E+06
^{238}U.icp/ms	µg/ml	9.14E+05	1.18E+06	2	1.05E+06	1.88E+05	17.9	NA
^{238}U.gea.PNNL	µg/ml	3.80E+05	< 2.76E+07	2	1.40E+07	NA	NA	NA
Water.grav	µg/ml	5.02E+05	1.01E+06	12	8.01E+05	1.90E+05	23.7	1.32E+06
Zn.icp.w	µg/ml	1.69E+01	4.25E+02	13	1.08E+02	1.08E+02	100.1	3.97E+02
Zn.icp.wo	µg/ml	1.69E+01	4.25E+02	11	1.20E+02	1.14E+02	95.3	4.41E+02

Table 4.3. K East Canister Sludge Characterization Data-- Per ml As-Settled Sludge. (Continued)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
Zr.icp.w	µg/ml	< 1.66E+01	2.04E+02	13	6.96E+01	5.02E+01	72.1	2.04E+02
Zr.icp.wo	µg/ml	1.70E+01	2.04E+02	8	8.98E+01	5.48E+01	61.0	2.64E+02
Zr.icp/ms	µg/ml	2.44E+02	4.59E+03	2	2.42E+03	3.07E+03	127.1	NA
m/z241.icp/ms	µg/ml	< 2.85E-01	9.34E+01	2	4.69E+01	NA	NA	NA

*: Per ml as-settled sludge.
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: Tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.w: Less than values were included (i.e., 3 for <3) when calculating the summary statistics.
.wo: Less than values were deleted when calculating the summary statistics.
NA: Not applicable for these data.
&: Calculated using the PNNL total uranium (laser fluoresence) data.

Table 4.4. K East Canister Sludge Characterization Data--Per Gram Dried Sludge.

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
Ag.icp.W	µg/g	1.82E+01	1.95E+02	12	8.62E+01	6.68E+01	77.4	2.69E+02
Ag.icp.WO	µg/g	1.82E+01	1.95E+02	10	9.92E+01	6.58E+01	66.4	2.91E+02
Al.icp	µg/g	1.04E+04	1.39E+05	12	6.15E+04	5.22E+04	84.9	2.04E+05
Al.icp/ms	µg/g	1.14E+03	1.03E+04	2	5.71E+03	6.46E+03	113.2	NA
Alpha Total	µCi/g	1.49E-01	3.44E+02	12	1.43E+02	9.38E+01	65.5	4.00E+02
Alpha Total.PNNL	µCi/g	1.55E+02	2.69E+02	2	2.12E+02	8.06E+01	38.0	NA
^{241}Am.aea.W	µCi/g	1.23E-01	2.20E+02	12	7.00E+01	5.79E+01	82.7	2.28E+02
^{241}Am.aea.WO	µCi/g	1.23E-01	2.20E+02	11	7.44E+01	5.86E+01	78.7	2.39E+02
^{241}Am.gea.W	µCi/g	< 1.64E+00	9.08E+02	12	1.49E+02	2.45E+02	164.4	8.18E+02
^{241}Am.gea.WO	µCi/g	1.79E+01	9.08E+02	11	1.62E+02	2.52E+02	155.3	8.71E+02
^{241}Am.gea.PNNL	µCi/g	3.15E-01	6.64E+01	2	3.33E+01	4.67E+01	140.1	NA
B.icp	µg/g	1.53E+02	1.02E+03	12	3.21E+02	2.56E+02	79.7	1.02E+03
Ba.icp.W	µg/g	< 2.57E+01	3.34E+02	12	1.49E+02	7.05E+01	47.2	3.42E+02
Ba.icp.WO	µg/g	1.25E+02	3.34E+02	9	1.71E+02	6.36E+01	37.2	3.64E+02
Be.icp	µg/g	< 2.57E+00	1.87E+02	12	NA	NA	NA	NA
Beta Total	µCi/g	1.42E+02	4.66E+03	12	2.62E+03	1.82E+03	69.4	7.60E+03
Beta Total.PNNL	µCi/g	4.91E+03	1.27E+04	2	8.80E+03	5.51E+03	62.6	NA
Bi.icp.W	µg/g	< 5.13E+01	6.63E+02	12	3.38E+02	1.79E+02	53.0	8.28E+02
Bi.icp.WO	µg/g	2.06E+02	6.63E+02	9	3.97E+02	1.61E+02	40.6	8.86E+02
^{212}Bi.gea	µCi/g	< 7.54E-01	< 1.79E+01	12	NA	NA	NA	NA
Br$^-$.ic	µg/g	< 3.89E-01	< 2.51E+01	10	NA	NA	NA	NA
Ca.icp.W	µg/g	< 2.25E+02	2.47E+03	12	1.05E+03	7.53E+02	71.8	3.11E+03
Ca.icp.WO	µg/g	4.12E+02	2.47E+03	9	1.32E+03	6.71E+02	50.8	3.35E+03
Ca.icp/ms	µg/g	7.11E+01	1.09E+02	2	8.98E+01	2.65E+01	29.5	NA
Cd.icp.W	µg/g	7.79E+00	2.29E+02	12	7.02E+01	7.49E+01	106.6	2.75E+02
Cd.icp.WO	µg/g	7.79E+00	2.29E+02	11	7.56E+01	7.61E+01	100.6	2.90E+02
^{144}Ce/Pr.gea	µCi/g	< 1.66E+00	< 3.57E+01	12	NA	NA	NA	NA
Cl$^-$.ic	µg/g	2.43E-01	9.71E+00	10	2.74E+00	2.86E+00	104.7	1.11E+01
$^{243/244}$Cm	µCi/g	< 1.48E-02	< 2.17E+01	12	NA	NA	NA	NA
^{57}Co.gea	µCi/g	3.73E-02	< 1.22E+02	12	NA	NA	NA	NA
^{60}Co.gea.W	µCi/g	< 4.47E-02	2.51E+00	12	1.34E+00	7.29E-01	54.4	3.34E+00
^{60}Co.gea.WO	µCi/g	6.94E-01	2.51E+00	7	1.59E+00	7.29E-01	45.7	4.07E+00
^{60}Co.gea.PNNL	µCi/g	1.66E-02	4.01E-01	2	2.09E-01	2.71E-01	130.2	NA
Cr.icp.W	µg/g	< 2.23E+01	2.30E+03	12	4.26E+02	6.53E+02	153.4	2.21E+03
Cr.icp.WO	µg/g	1.20E+02	2.30E+03	7	7.13E+02	7.42E+02	104.0	3.24E+03

Table 4.4. K East Canister Sludge Characterization Data--
Per Gram Dried Sludge. (Continued)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
^{134}Cs.gea	µCi/g	< 1.77E-01	< 2.70E+00	12	NA	NA	NA	NA
^{134}Cs.gea.PNNL	µCi/g	< 3.00E-03	2.41E-01	2	1.22E-01	NA	NA	NA
^{137}Cs.gea	µCi/g	5.01E+01	1.95E+03	12	7.23E+02	5.38E+02	74.3	2.19E+03
^{137}Cs.gea.PNNL	µCi/g	1.17E+00	5.92E+02	2	2.97E+02	4.18E+02	140.9	NA
Cu.icp	µg/g	9.52E+00	1.28E+03	12	3.79E+02	4.50E+02	118.6	1.61E+03
^{152}Eu.gea	µCi/g	< 1.10E-01	< 6.46E+00	12	NA	NA	NA	NA
^{154}Eu.gea.w	µCi/g	< 1.65E-01	2.41E+01	12	1.03E+01	6.89E+00	66.9	2.92E+01
^{154}Eu.gea.wo	µCi/g	2.52E+00	2.41E+01	11	1.12E+01	6.41E+00	57.1	2.93E+01
^{154}Eu.gea.PNNL	µCi/g	4.38E-02	9.55E+00	2	4.80E+00	6.72E+00	140.1	NA
^{155}Eu.gea	µCi/g	< 6.43E-01	< 1.41E+01	12	NA	NA	NA	NA
^{155}Eu.gea.PNNL	µCi/g	1.82E-02	4.83E+00	2	2.42E+00	3.40E+00	140.4	NA
F$^-$.ic	µg/g	< 4.62E-02	< 2.40E+00	10	NA	NA	NA	NA
Fe.icp	µg/g	4.59E+02	2.56E+05	12	6.73E+04	8.75E+04	130.0	3.07E+05
Fe.icp/ms	µg/g	1.68E+03	6.23E+03	2	3.95E+03	3.21E+03	81.3	NA
K.icp	µg/g	< 6.60E+02	3.60E+03	12	NA	NA	NA	NA
Mg.icp.w	µg/g	< 5.13E+01	2.21E+03	12	1.11E+03	8.42E+02	75.5	3.42E+03
Mg.icp.wo	µg/g	2.96E+02	2.21E+03	8	1.58E+03	6.07E+02	38.4	3.52E+03
Mn.icp.w	µg/g	< 2.23E+01	8.10E+02	12	2.25E+02	2.90E+02	128.5	1.02E+03
Mn.icp.wo	µg/g	9.18E+01	8.10E+02	7	3.70E+02	3.09E+02	83.5	1.42E+03
NO$_2^-$.ic	µg/g	< 3.35E-01	< 2.16E+01	10	NA	NA	NA	NA
NO$_3^-$.ic	µg/g	< 4.31E-01	< 2.78E+01	10	NA	NA	NA	NA
Na.icp	µg/g	4.01E+02	2.50E+05	12	2.15E+04	7.20E+04	335.3	2.18E+05
^{94}Nb.gea	µCi/g	< 4.89E-02	< 1.80E+00	12	NA	NA	NA	NA
Ni.icp.w	µg/g	< 4.44E+01	4.98E+02	12	1.37E+02	1.39E+02	101.1	5.16E+02
Ni.icp.wo	µg/g	6.17E+01	4.98E+02	7	2.02E+02	1.53E+02	75.9	7.23E+02
^{237}Np.w	µCi/g	< 7.44E-03	4.94E-02	12	1.96E-02	1.32E-02	67.4	5.59E-02
^{237}Np.wo	µCi/g	7.94E-03	4.94E-02	7	1.87E-02	1.41E-02	75.1	6.65E-02
Oxalate.ic	µg/g	< 3.27E-01	< 2.11E+01	10	NA	NA	NA	NA
P.icp	µg/g	< 2.63E+02	2.21E+03	12	NA	NA	NA	NA
PO$_4^{3-}$.ic	µg/g	< 3.72E-01	< 2.40E+01	10	NA	NA	NA	NA
Pb.icp.w	µg/g	6.77E+01	7.66E+02	12	3.18E+02	2.13E+02	67.1	9.01E+02
Pb.icp.wo	µg/g	6.77E+01	7.66E+02	7	3.79E+02	2.70E+02	71.2	1.30E+03
^{238}Pu.w	µCi/g	5.41E-03	< 7.04E+01	12	1.89E+01	1.94E+01	102.5	7.19E+01
^{238}Pu.wo	µCi/g	5.41E-03	4.07E+01	11	1.42E+01	1.11E+01	78.3	4.56E+01
$^{239/240}$Pu	µCi/g	5.01E-02	1.90E+02	12	8.62E+01	5.47E+01	63.5	2.36E+02

Table 4.4. K East Canister Sludge Characterization Data--
Per Gram Dried Sludge. (Continued)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
^{239}Pu.icp/ms	µCi/g	1.57E+01	9.70E+01	2	5.64E+01	5.75E+01	102.0	NA
^{240}Pu.icp/ms	µCi/g	< 4.54E-02	3.89E+01	2	1.95E+01	NA	NA	NA
^{226}Ra.gea	µCi/g	< 3.30E+00	< 6.81E+01	12	NA	NA	NA	NA
^{106}Ru/Rh.gea	µCi/g	< 2.62E+00	< 5.37E+01	12	NA	NA	NA	NA
SO$_4^{2-}$.ic.w	µg/g	1.44E+00	< 2.76E+01	10	1.10E+01	1.04E+01	94.4	4.14E+01
SO$_4^{2-}$.ic.wo	µg/g	1.44E+00	2.43E+01	8	9.73E+00	9.67E+00	99.4	4.06E+01
^{125}Sb.gea.PNNL	µCi/g	< 2.00E-02	1.11E+00	2	5.63E-01	NA	NA	NA
Se.icp	µg/g	< 5.13E+01	< 2.56E+02	12	NA	NA	NA	NA
Sm.icp	µg/g	< 5.13E+01	< 2.56E+02	12	NA	NA	NA	NA
$^{89/90}$Sr	µCi/g	2.81E+01	2.19E+03	12	1.00E+03	8.39E+02	83.5	3.30E+03
TIC	µg/g	2.89E+02	1.43E+04	12	3.38E+03	4.07E+03	120.4	1.45E+04
TOC	µg/g	4.32E+02	1.37E+04	12	3.40E+03	4.12E+03	121.2	1.47E+04
^{99}Tc	µCi/g	2.94E-01	3.42E+01	6	1.33E+01	1.41E+01	106.7	6.57E+01
Tl.icp	µg/g	< 1.03E+02	< 5.10E+02	12	NA	NA	NA	NA
^{208}Tl.gea	µCi/g	< 2.20E+00	< 4.45E+01	12	NA	NA	NA	NA
Total Carbon	µg/g	6.38E+02	2.96E+04	12	6.51E+03	8.14E+03	125.0	2.88E+04
U.icp.w	µg/g	< 2.57E+02	7.49E+05	12	4.18E+05	2.97E+05	71.0	1.23E+06
U.icp.wo	µg/g	7.79E+04	7.49E+05	11	4.56E+05	2.79E+05	61.1	1.24E+06
U.las	µg/g	9.33E+04	8.81E+05	10	6.93E+05	2.63E+05	38.0	1.46E+06
U.phos	µg/g	9.44E+02	7.40E+05	12	3.54E+05	2.46E+05	69.6	1.03E+06
U.icp/ms	µg/g	6.43E+05	6.50E+05	2	6.46E+05	5.30E+03	0.8	NA
^{233}U.tims	mass%	< 5.00E-04	< 5.00E-04	10	NA	NA	NA	NA
^{233}U.tims &	µg/g	< 4.56E-01	< 4.31E+00	10	NA	NA	NA	NA
^{233}U.icp/ms	µg/g	2.00E-01	4.35E-01	2	3.18E-01	1.66E-01	52.3	NA
^{234}U.tims	mass%	5.10E-03	1.48E-02	10	8.84E-03	2.62E-03	29.6	1.65E-02
^{234}U.tims &	µg/g	1.35E+01	8.65E+01	10	5.64E+01	2.36E+01	41.9	1.25E+02
^{234}U.icp/ms	µg/g	3.52E+01	4.95E+01	2	4.23E+01	1.01E+01	24.0	NA
^{235}U.tims	mass%	4.97E-01	9.55E-01	10	7.09E-01	1.17E-01	16.4	1.05E+00
^{235}U.tims &	µg/g	6.68E+02	7.76E+03	10	4.88E+03	2.13E+03	43.6	1.11E+04
^{235}U.icp/ms	µg/g	3.33E+03	6.32E+03	2	4.82E+03	2.11E+03	43.9	NA
^{236}U.tims	mass%	3.90E-02	9.75E-02	10	6.98E-02	1.86E-02	26.6	1.24E-01
^{236}U.tims &	µg/g	7.07E+01	6.93E+02	10	4.61E+02	1.93E+02	41.8	1.02E+03
^{236}U.icp/ms	µg/g	2.89E+02	3.70E+02	2	3.30E+02	5.73E+01	17.4	NA
^{238}U.tims	mass%	98.983	99.459	10	99.212	1.19E-01	0.1	99.560
^{238}U.tims &	µg/g	9.25E+04	8.76E+05	10	6.88E+05	2.61E+05	38.0	1.45E+06

Table 4.4. K East Canister Sludge Characterization Data--
Per Gram Dried Sludge. (Continued)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
^{238}U.icp/ms	µg/g	6.35E+05	6.42E+05	2	6.38E+05	4.60E+03	0.7	NA
^{238}U.gea.PNNL	µg/g	2.67E+05	< 1.49E+07	2	7.58E+06	NA	NA	NA
Zn.icp.w	µg/g	2.23E+01	1.42E+03	12	4.75E+02	5.73E+02	120.6	2.04E+03
Zn.icp.wo	µg/g	2.23E+01	1.42E+03	10	5.65E+02	5.89E+02	104.3	2.28E+03
Zr.icp.w	µg/g	1.20E+01	8.90E+02	12	2.80E+02	3.15E+02	112.4	1.14E+03
Zr.icp.wo	µg/g	1.20E+01	8.90E+02	8	4.09E+02	3.16E+02	77.1	1.41E+03
Zr.icp/ms	µg/g	1.32E+02	3.22E+03	2	1.68E+03	2.18E+03	130.3	NA
m/z241.icp/ms	µg/g	< 2.00E-01	5.03E+01	2	2.53E+01	NA	NA	NA

*: Per gram dried sludge.
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: Tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.w: Less than values were included (i.e., 3 for <3) when calculating the summary statistics.
.wo: Less than values were deleted when calculating the summary statistics.
NA: Not applicable for these data.
&: Calculated using the PNNL total uranium (laser fluoresence) data.

Table 4.5. K East Sludge Characterization Data--K East Basin Floor/Weasel Pit Versus K East Canisters.

Analyte	Units As-Settled Sludge	K East Canister		K East Basin Floor/Weasel Pit	
		Minimum Concentration Observed	Maximum Concentration Observed	Minimum Concentration Observed	Maximum Concentration Observed
Ag.icp	µg/ml	1.31E+00	3.79E+02	< 1.02E+00	< 3.77E+01
Al.icp	µg/ml	7.40E+03	5.26E+04	8.54E+03	5.87E+04
Al.icp/ms	µg/ml	1.62E+03	1.91E+04	NA	NA
Alpha Total	µCi/ml	2.12E-01	4.17E+02	4.52E-01	1.11E+02
Alpha Total.PNNL	µCi/ml	2.21E+02	5.00E+02	NA	NA
^{241}Am.aea	µCi/ml	1.75E-01	1.80E+02	1.57E-01	5.60E+01
^{241}Am.gea	µCi/ml	< 2.34E+00	2.65E+02	1.82E-01	5.35E+01
^{241}Am.gea.PNNL	µCi/ml	4.49E-01	1.23E+02	NA	NA
B.icp	µg/ml	5.68E+01	3.64E+02	< 4.01E+01	7.66E+02
Ba.icp	µg/ml	< 5.64E+00	2.97E+02	2.32E+01	5.64E+02
Be.icp	µg/ml	< 3.66E+00	< 2.72E+01	1.76E+00	2.64E+01
Beta Total	µCi/ml	7.91E+00	1.08E+04	2.07E+01	3.09E+03
Beta Total.PNNL	µCi/ml	9.11E+03	1.81E+04	NA	NA
Bi.icp	µg/ml	< 1.13E+01	1.23E+03	NA	NA
^{212}Bi.gea	µCi/ml	< 8.61E-02	< 4.26E+01	< 7.59E-02	< 3.55E+00
Br$^-$.ic	µg/ml	< 6.97E-01	< 1.40E+00	NA	NA
CN$^-$	µg/ml	NA	NA	< 3.11E-01	< 1.34E+00
Ca.icp	µg/ml	6.34E+01	1.45E+03	2.68E+02	3.31E+04
Ca.icp/ms	µg/ml	1.01E+02	2.02E+02	NA	NA
Cd.icp	µg/ml	< 8.29E+00	5.69E+01	1.90E+01	7.64E+01
^{144}Ce/Pr.gea	µCi/ml	< 9.25E-02	< 7.07E+01	< 2.93E-01	< 1.15E+01
Cl$^-$.ic	µg/ml	3.47E-01	3.25E+00	1.70E-02	5.28E-01
$^{243/244}$Cm	µCi/ml	< 2.10E-02	< 4.33E+01	< 1.14E-01	< 1.28E+01
^{57}Co.gea	µCi/ml	2.21E-02	< 1.46E+01	NA	NA
^{60}Co.gea	µCi/ml	< 6.36E-02	< 3.46E+00	8.46E-02	2.34E+00
^{60}Co.gea.PNNL	µCi/ml	2.36E-02	7.44E-01	NA	NA
Cr.icp	µg/ml	< 1.66E+01	3.27E+03	4.15E+01	1.91E+03
^{134}Cs.gea	µCi/ml	< 2.36E-02	< 5.05E+00	< 3.83E-02	< 8.83E-01
^{134}Cs.gea.PNNL	µCi/ml	< 4.27E-03	4.47E-01	NA	NA
^{137}Cs.gea	µCi/ml	2.80E+00	2.13E+03	2.73E+01	1.48E+03
^{137}Cs.gea.PNNL	µCi/ml	1.67E+00	1.10E+03	NA	NA
Cu.icp	µg/ml	1.36E+01	2.23E+02	2.46E+01	9.47E+02

Table 4.5. K East Sludge Characterization Data--K East Basin Floor/Weasel Pit Versus K East Canisters. (Continued)

Analyte	Units As-Settled Sludge	K East Canister		K East Basin Floor/Weasel Pit	
		Minimum Concentration Observed	Maximum Concentration Observed	Minimum Concentration Observed	Maximum Concentration Observed
DSC.dry (Exotherms) *	Joules/g dry	0.00E+00	5.84E+01	0.00E+00	1.90E+02 222-S 1.12E+01 PNNL
DSC.wet (Exotherms) *	Joules/g wet	0.00E+00	4.49E+01	0.00E+00	6.14E+00 222-S 8.00E+00 PNNL
^{152}Eu.gea	µCi/ml	< 2.53E-02	< 1.54E+01	< 1.40E-02	< 7.02E-01
^{154}Eu.gea	µCi/ml	< 2.35E-01	3.45E+01	< 1.87E-02	8.90E+00
^{154}Eu.gea.PNNL	µCi/ml	6.24E-02	1.77E+01	NA	NA
^{155}Eu.gea	µCi/ml	1.52E-01	2.26E+01	< 5.41E-02	4.10E+00
^{155}Eu.gea.PNNL	µCi/ml	2.59E-02	8.96E+00	NA	NA
F$^-$.ic	µg/ml	< 6.66E-02	3.97E-01	< 1.08E-03	1.62E+00
Fe.icp	µg/ml	6.54E+02	7.66E+04	6.92E+03	5.22E+05
Fe.icp/ms	µg/ml	2.39E+03	1.16E+04	NA	NA
K.icp	µg/ml	< 5.64E+01	5.12E+03	< 6.25E+01	2.54E+03
Mg.icp	µg/ml	< 1.13E+01	4.10E+03	1.28E+02	4.82E+03
Mn.icp	µg/ml	< 1.66E+01	1.99E+02	6.14E+01	9.84E+02
NH3	µg/ml	NA	NA	< 7.52E-02	< 1.23E+01
NO$_2^-$.ic	µg/ml	< 6.01E-01	< 1.21E+00	< 1.41E-02	< 9.04E-01
NO$_3^-$.ic	µg/ml	< 7.72E-01	1.99E+00	3.46E-02	2.15E+00
Na.icp	µg/ml	9.80E+01	3.56E+05	9.03E+01	1.49E+04
^{94}Nb.gea	µCi/ml	< 1.23E-02	< 4.28E+00	< 1.23E-02	< 4.50E-01
Ni.icp	µg/ml	1.14E+01	2.16E+02	NA	NA
^{237}Np	µCi/ml	2.76E-03	3.36E-02	< 7.21E-04	< 8.54E-03
Oxalate.ic	µg/ml	< 5.86E-01	< 1.18E+00	NA	NA
P.icp	µg/ml	< 5.01E+01	3.15E+03	NA	NA
PO$_4^{3-}$.ic	µg/ml	< 6.66E-01	< 1.34E+00	< 2.45E-02	< 2.38E+00
Pb.icp	µg/ml	3.57E+01	< 5.45E+02	4.20E+01	1.08E+03
^{238}Pu	µCi/ml	7.71E-03	1.30E+02	2.83E-02	< 1.09E+01
$^{239/240}$Pu	µCi/ml	7.14E-02	2.84E+02	1.84E-01	4.48E+01
^{239}Pu.icp/ms	µg/ml	2.23E+01	1.80E+02	NA	NA
^{240}Pu.icp/ms	µg/ml	< 6.47E-02	7.23E+01	NA	NA
^{226}Ra.gea	µCi/ml	< 1.85E-01	< 1.39E+02	< 5.86E-01	< 2.20E+01
^{106}Ru/Rh.gea	µCi/ml	< 1.46E-01	< 1.06E+02	< 4.27E-01	< 1.68E+01
SO$_4^{2-}$.ic	µg/ml	< 1.50E+00	5.20E+00	2.45E-02	2.03E+01
^{125}Sb.gea.PNNL	µCi/ml	< 2.85E-02	2.05E+00	< 3.02E-02	< 3.75E+00

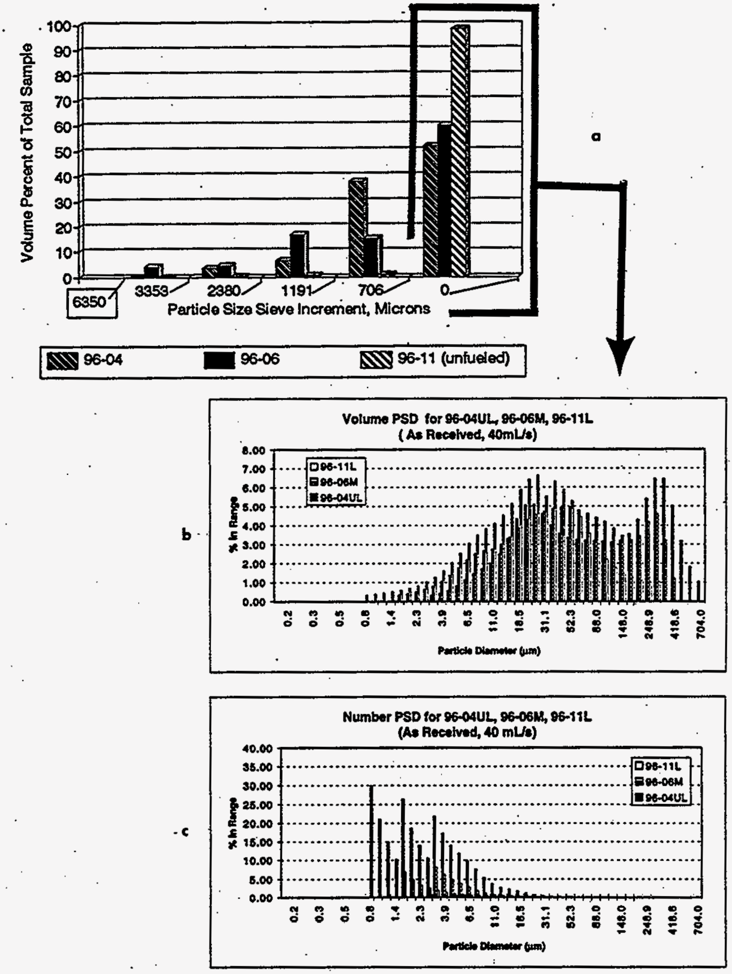
Table 4.5. K East Sludge Characterization Data--K East Basin Floor/Weasel Pit Versus K East Canisters. (Continued)

Analyte	Units As-Settled Sludge	K East Canister		K East Basin Floor/Weasel Pit	
		Minimum Concentration Observed	Maximum Concentration Observed	Minimum Concentration Observed	Maximum Concentration Observed
Se.icp	µg/ml	< 1.13E+01	< 5.45E+02	< 1.02E+01	< 3.77E+02
Sm.icp	µg/ml	< 1.13E+01	< 5.45E+02	< 1.02E+01	< 3.77E+02
$^{89/90}$Sr	µCi/ml	2.05E+00	4.71E+03	1.77E+00	1.38E+03
%Water (222S) #	wt%	2.23E+01	9.48E+01	4.54E+01	9.05E+01
%Water.tga (PNNL)	wt%	1.77E+01	8.13E+01	9.51E+00	8.53E+01
TIC	µg/ml	2.20E+02	2.04E+04	7.50E+00	3.26E+03
TOC	µg/ml	3.09E+02	1.95E+04	3.87E+02	4.03E+03
^{99}Tc	µCi/ml	4.19E-01	1.89E+01	NA	NA
Tl.icp	µg/ml	< 2.26E+01	< 1.09E+03	< 2.04E+01	< 7.55E+02
^{208}Tl.gea	µCi/ml	< 1.23E-01	< 9.04E+01	< 9.51E-02	< 1.19E+01
Total Carbon	µg/ml	5.47E+02	4.22E+04	6.68E+02	6.55E+03
U.icp	µg/ml	< 3.66E+02	1.76E+06	1.14E+03	9.18E+04
U.las	µg/ml	2.79E+04	2.00E+06	1.44E+03	4.27E+05
U.phos	µg/ml	1.34E+03	1.44E+06	1.31E+03	3.67E+04
U.icp/ms	µg/ml	9.26E+05	1.19E+06	NA	NA
^{233}U.tims	µg/ml	< 1.36E-01	< 9.80E+00	< 1.41E-02	< 4.18E+00
^{233}U.icp/ms	µg/ml	3.71E-01	6.20E-01	NA	NA
^{234}U.tims	µg/ml	4.04E+00	1.93E+02	1.10E-01	3.49E+01
^{234}U.icp/ms	µg/ml	6.53E+01	7.05E+01	NA	NA
^{235}U.tims	µg/ml	2.00E+02	1.48E+04	1.00E+01	3.10E+03
^{235}U.icp/ms	µg/ml	6.18E+03	9.00E+03	NA	NA
^{236}U.tims	µg/ml	2.11E+01	1.65E+03	1.10E+00	2.12E+02
^{236}U.icp/ms	µg/ml	5.27E+02	5.37E+02	NA	NA
^{238}U.tims	µg/ml	2.76E+04	1.98E+06	1.43E+03	4.24E+05
^{238}U.icp/ms	µg/ml	9.14E+05	1.18E+06	NA	NA
^{238}U.gea.PNNL	µg/ml	3.80E+05	< 2.76E+07	NA	NA
Zn.icp	µg/ml	1.69E+01	4.25E+02	5.44E+01	2.09E+03
Zr.icp	µg/ml	< 1.66E+01	2.04E+02	2.82E+01	1.06E+03
Zr.icp/ms	µg/ml	2.44E+02	4.59E+03	NA	NA
m/z241.icp/ms	µg/ml	< 2.85E-01	9.34E+01	NA	NA
Residue	µg/ml	NA	NA	5.80E+03	4.45E+05

#: 222-S %Water analyses by TGA for the Floor/Weasel Pit samples and by gravimetric for canister samples.

Figure 4.1. One Sludge Particle Observed Bubbling during Sieving for Particle Size Determinations

Figure 4.2. Particle Size Measurements.
(a) Summary of seiving results, (b and c) examples of data from laser scattering measurements which apply only to the smallest particles. Comparisons shown for seived volumes are, at best, estimates since measurements are taken in mass units and converted to volume using density data. Various assumptions were made when all the layers from a given sample were not examined.



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APPENDIX A

SAMPLING EQUIPMENT, CANISTER LOCATIONS, OPERATIONAL SEQUENCE, AND BACKGROUND

R. B. Baker

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APPENDIX A

SAMPLING EQUIPMENT, CANISTER LOCATIONS, OPERATIONAL SEQUENCE, AND BACKGROUND

A.1 SAMPLING EQUIPMENT BACKGROUND

The overall sequence of collection using the canister sludge sampling equipment is provided below. An additional process flow schematic for equipment is provided in Figure A.1. A detailed summary of the sample container support pole, control rod assembly and the sample container is provided in Figure A.2. Not shown is that, on the grating level, the required pump and video monitors were mounted on a special cart. Between the drawing of each sample the equipment components which come in contact with sample material were either replaced for each sample or were backflushed with deionized water to remove any residual sludge materials from the system. Note that the pump only handled air, not water or sludge; its function was to provide the vacuum to draw the sample into the sample container.

The general sequence for drawing a sample was:

1. The canister to be sampled was chosen (Makenas 1996b) from criteria described in Sampling and Analysis Plan (Welsh 1996). Parameters considered are discussed in Section 3.1.2.

2. The grating area over the canister to be sampled was prepared with installation of the sample container support pole assembly in the grating slot nearby. On the grating the special cart carrying the vacuum pumping system and supporting equipment (including video monitors for underwater cameras used to monitor sampling and the safety delay container) was also readied.

3. If needed, the sludge sample extraction tube and nozzle were backflushed to clean any residue from prior sampling. The extraction tube assembly was then moved to the sampling location. A uniquely numbered sample container was moved into the container support assembly brackets. This clean container was sealed (the valves on lid closed) prior to it being placed in the basin water (i.e., container contains only air at one atmosphere pressure at this point).

4. The sample control rod assembly with safety delay container was moved over the inlet and outlet ports on the lid of the sample container. The quick-disconnect fittings of these ports are engaged by pressing down the specially designed rod assembly. No water enters the sample container during this operation (i.e., the sample container remains filled only with air).

5. The identity of the fuel canister barrel to be sampled was verified and noted in the logbook. The support valves are verified to be the correct positions for sampling.

6. The video cameras were positioned. One monitored the safety delay container (a redundant safety precaution to assure no water reaches the pump system; if water would have been noted in this container, sampling would be halted). The other camera monitors the fuel storage canister barrel and extraction nozzle position.

7. The vacuum pump was started and the sampling container evacuated of air. The extraction nozzle was lowered into position in the canister barrel just above the surface of the sludge in the canister barrel. Promising candidate locations between fuel elements were previously determined by ultrasound.

8. The sludge sample was then drawn. The operator on the grating controlled the sample material being drawn into the 10.3 liter sample container with handle-controls linked to valves underwater. The nozzle was lowered extracting sludge in each channel between the fuel elements (or between the fuel elements and the canister barrel wall). To obtain a representative sample, each channel was cleaned out to the bottom of the barrel before sampling to the next channel. The sampling was continued until the sample container was filled. If container volume permitted and if there were any "outer" fuel elements which did not enclose an "inner" element, the extraction nozzle was used to pull sludge from the annuli of elements.

9. The completely filled sample container was then uncoupled from the control rod assembly. The container was then removed from the support pole assembly and moved to a storage area in the basin and monitored for gas generation. Once six containers were ready the cask was loaded underwater in the K East Basin South Loadout Pit and transported by truck to the Hanford 327 Building Facility. Here the cask was placed in the pool and the containers removed and stored.

10. The containers were then removed from the pool through a hot cell and transported in a smaller cask to the 325 Building Laboratory hot cells. Here the samples were recovered from the primary sample containers and the detailed analyses begun.

A.2 ADDITIONAL BACKGROUND ON CANISTERS SAMPLED

Tables 3.1 and 3.2 of the main text provide summary design and operational parameters for the canister barrels sampled for sludge at the K East Basin. Figures A.3 through A.7 show general overviews of the top of the canister barrels sampled. These views were taken from underwater video surveys made of the candidate canisters considered for sampling. These videos were taken prior to sampling and were part of the information used to decide which canisters would be sampled for sludge and fuel elements to receive destructive examinations in the hot cells.

The white corrosion nodules on the canister barrels are especially visible in Figure A.7. Note that the holes visible in the bottom of the canister barrel sampled for 96-09, Figure A.5. This barrel did not contain any fuel elements. It is expected that the sludge residing below these wire mesh openings in the canister barrel bottom was also drawn into the sample taken, as well as the material above. The canister barrels sit directly on the concrete floor in K East Basin.

Figure A1. Schematic Process Flow Diagram for Canister Sludge Sampler.

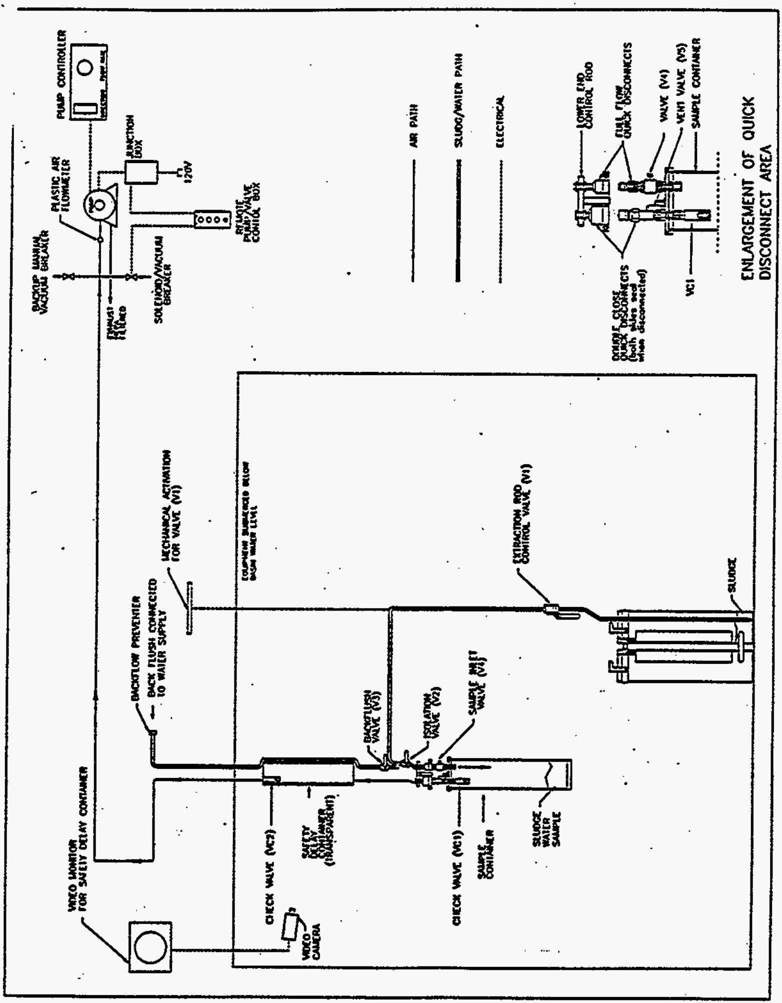


Figure A2. Sample Container Support Pole Assembly, Control Rod Assembly, and Sample Container.

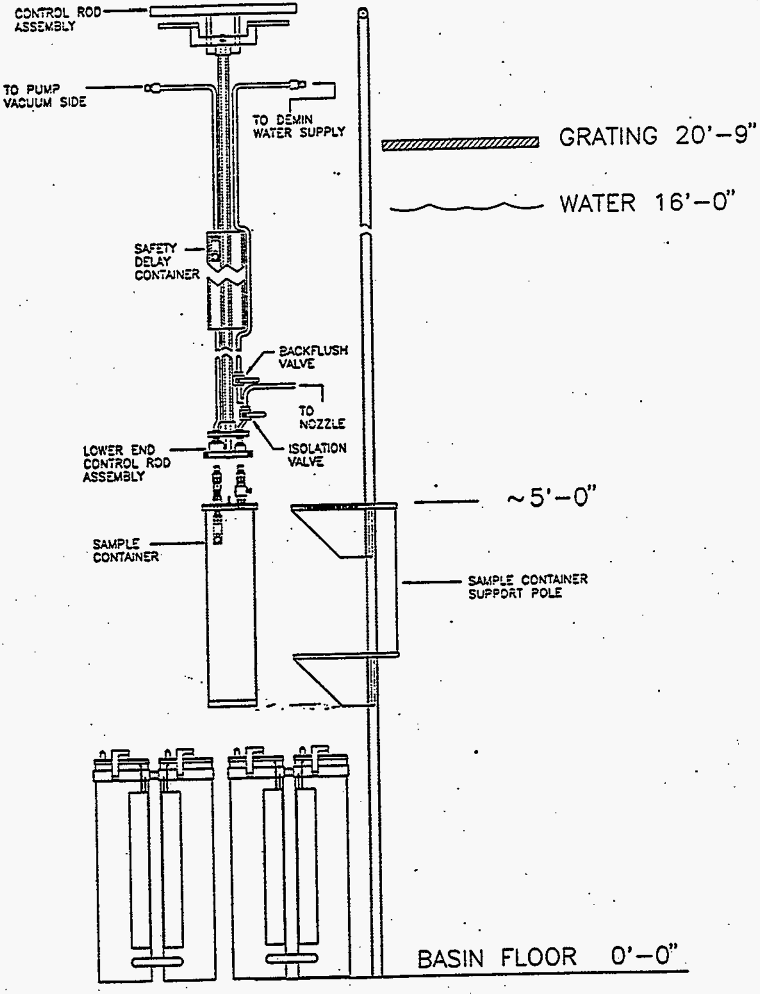


Figure A3. Overview of Canister Barrels Sampled for Sludge Samples 96-01 (Top) and 96-04 (Bottom).

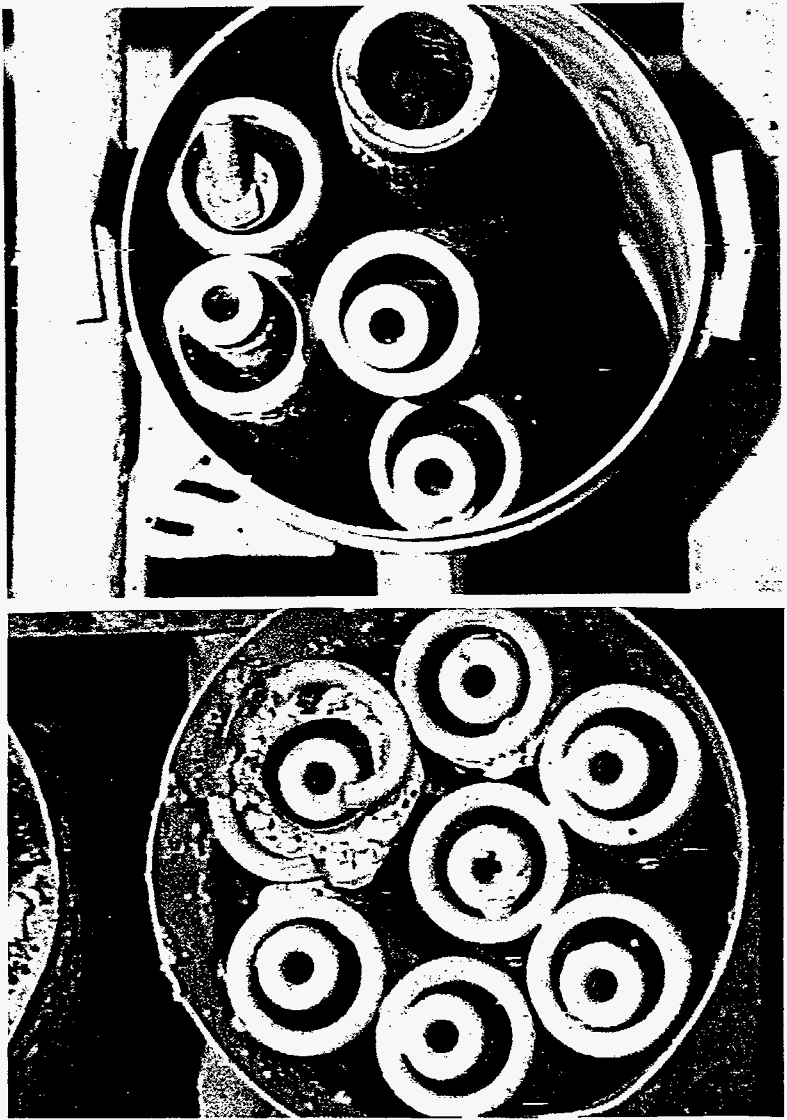


Figure A4. Overview of Canister Barrels Sampled for Sludge Samples 96-05 (Top) and 96-06 (Bottom).

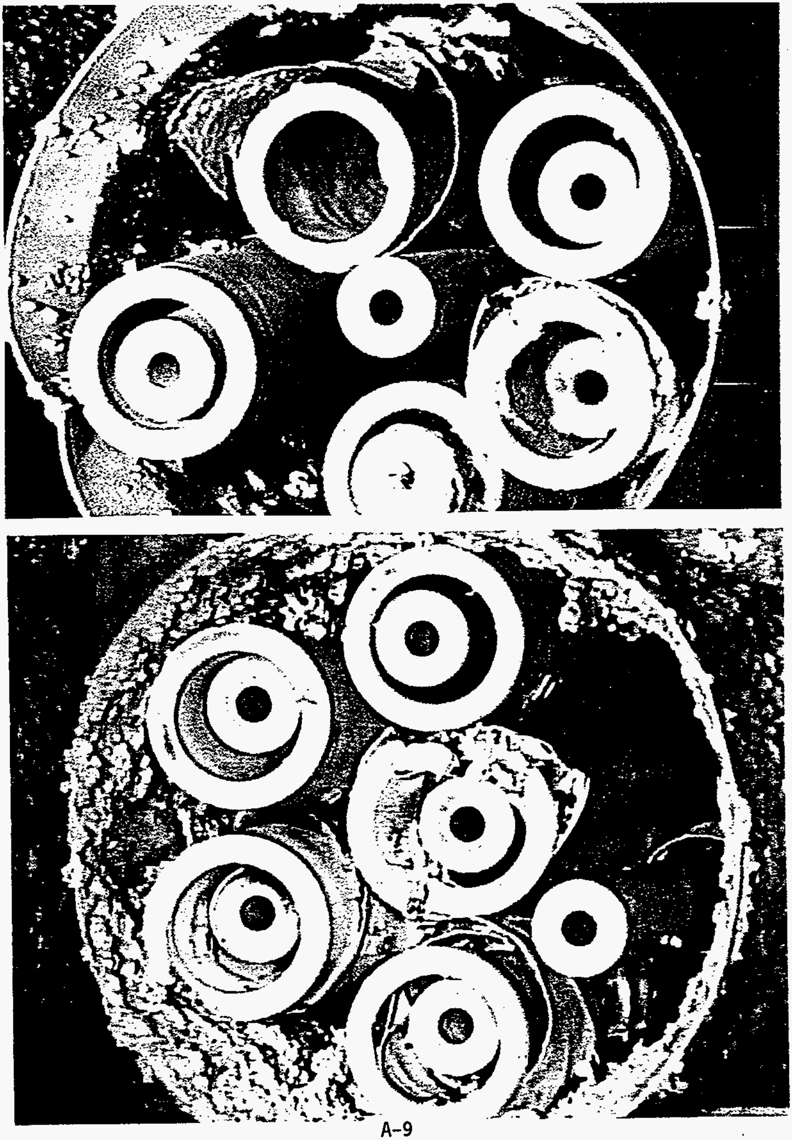


Figure A5. Overview of Canister Barrels Sampled for Sludge Samples 96-08 (Top) and 96-09 (Bottom photograph, top empty barrel).

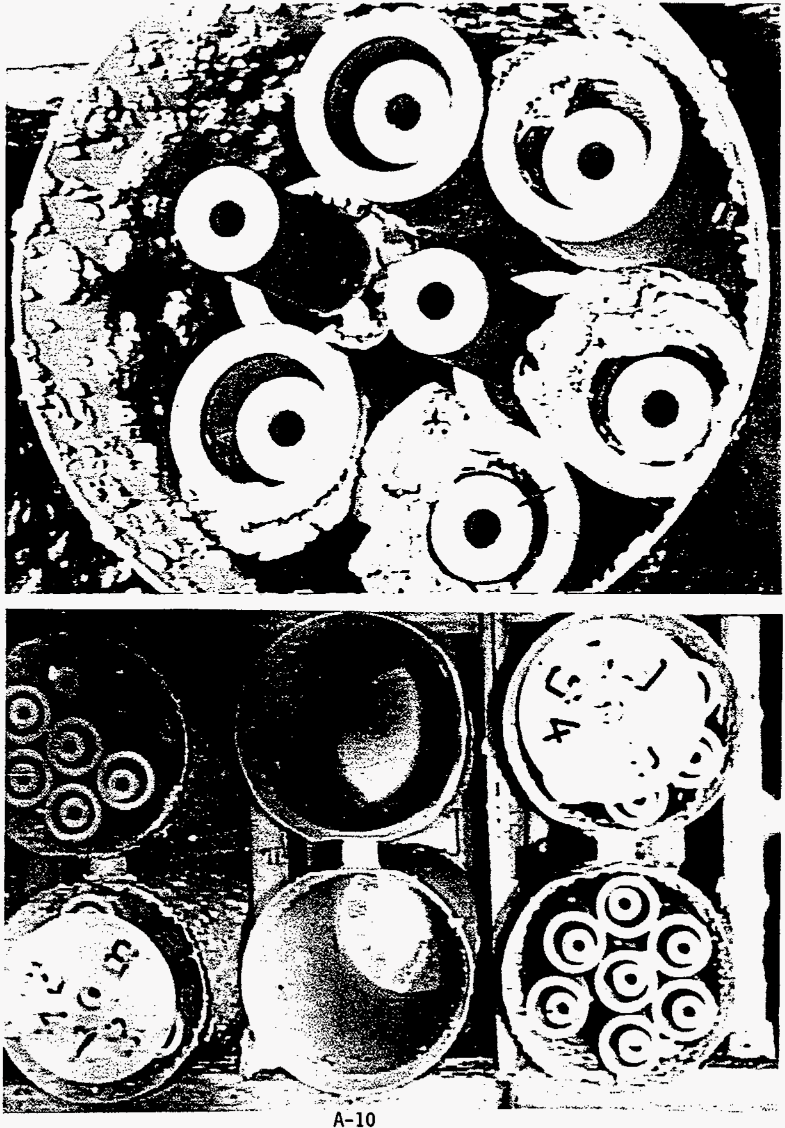


Figure A6. Overview of Canister Barrels Sampled for Sludge Samples 96-11 (Top photograph, bottom empty barrel) and 96-13 (Bottom).

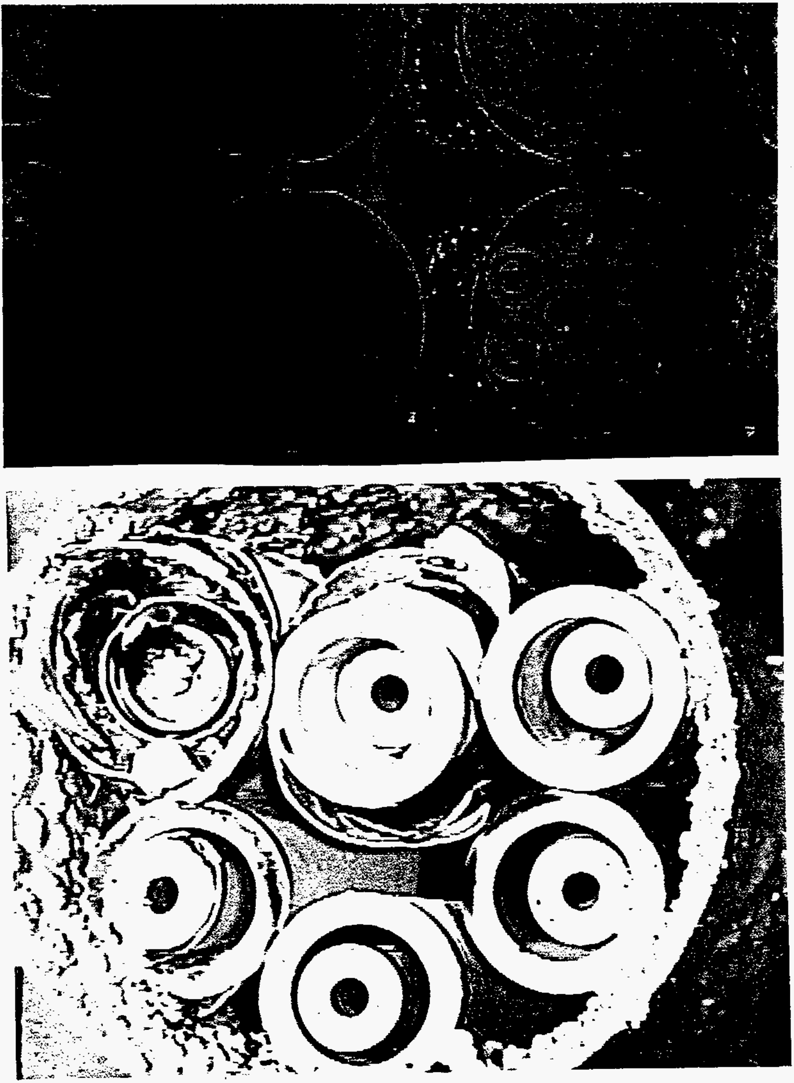
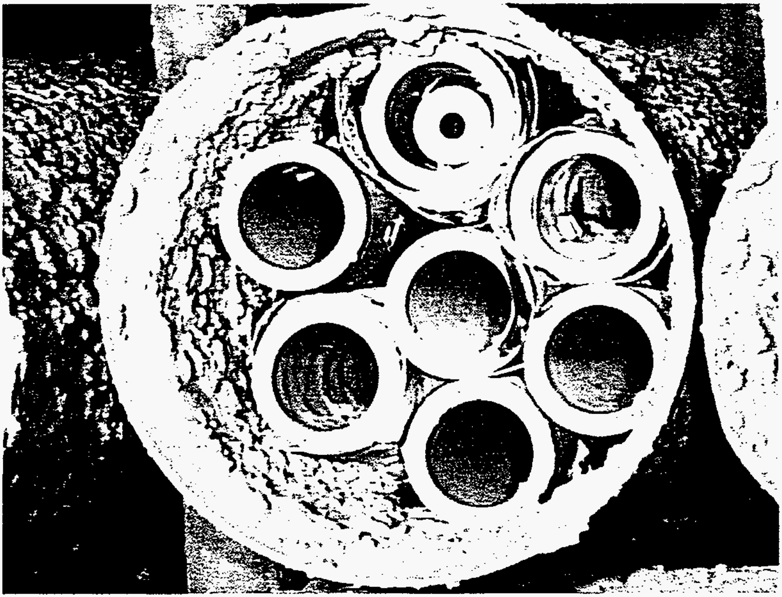


Figure A7. Overview of Canister Barrel Sampled for Sludge Sample 96-15.



APPENDIX B

SUMMARY OF K EAST BASIN CANISTER SLUDGE GAS SAMPLING RESULTS

P. R. Bredt and D. E. Rinehart

(Excerpted in part from Reference Silvers 1997a)

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APPENDIX B

SUMMARY OF K EAST BASIN CANISTER SLUDGE GAS SAMPLING RESULTS

B.1 INTRODUCTION

This appendix describes a scoping study which includes the collection of gas from the sludge samples and the conditions under which the gas was collected. With the exception of isotopic mass spectrometric results, the data presented are semi-quantitative.

Between June 11-20, 1996, nine K Basin canister sludge samples were delivered to the 325A High Level Radiochemistry Facility (325A HLRF). These samples were contained in sealed stainless steel shipping canisters with an approximate volume of 10.5 L each. Tables 3.1 and 3.2 list sampling information.

At the start of the characterization effort four of the nine samples were vacuum transferred out of the stainless steel shipping canisters. Liquids were transferred into 10 L glass carboys while the settled sludge was transferred into 2 L glass graduated cylinders. In all four cases, supernatant was transferred back to the canister to aid in the recovery of the sludge remaining in the canisters. Supernatant was added to the graduated cylinders to bring the volume in each cylinder to approximately 1.7 L. Sample 96-05 was transferred on July 11, 1996, and Samples 96-01, 96-06, and 96-08 were transferred on July 12, 1996. Upon opening the ball valves on canisters containing Samples 96-05 and 96-06, supernatant was forced out of the valves to a height of approximately 2 ft indicating pressurization of the canisters had occurred. A fine particulate layer was noted on the bottom of the shipping canisters following the initial vacuum transfer. Repeated additions of supernatant followed by vacuum transfer were used to recover these fine particles. The fine particles were added to the slurry in the graduated cylinders. The cell temperature was 35 °C and the cells were maintained at a vacuum of approximately 0.26 in. of water relative to ambient air.

Within a few days after samples were loaded into the graduated cylinders, bubbles were observed releasing from the settled sludge layer in Samples 96-05 and 96-06. This gas release was not quantified nor was the gas collected.

The four samples in the 2 L glass graduated cylinders were sparged with air on July 29, 1996 for 5 min. to mobilize the solid layer and obtain a homogeneous slurry for the settling study. Air sparging was decided upon after attempts to mobilize the solid layer using magnetic stirrers as well as motorized blade mixers failed. The magnetic stir bar sat in the sludge and would not rotate. The motorized blade mixer stirred the solution, but did not lift the sludge off the bottom of the cylinders. Three video cameras were used to collect images both real time and time lapse as the slurry settled. Video images and visual observations showed gas generation in

Samples 96-05, 96-08, and 96-06, with gas retention in Sample 96-05. The gas appeared to be generated at the bottom of Sample 96-06 and released through preferred paths in the solids close to and along the walls of the graduated cylinder. In Sample 96-05, a gas pocket formed near the bottom of the sludge layer and expanded across the cylinder lifting the overlying solid layer. The gas pocket continued to grow until July 31, 1996 when the gas quickly released from the sludge. The presence of the gas pocket near the bottom of the sludge along with the lack of gas release in the overlying sludge indicates the gas was generated predominantly in the faster settling portion of the sludge. The top of the sludge layer for Sample 96-05 on July 30, 1996 at 4 p.m. was at the 290 ml mark on the graduated cylinder. Just before the gas pocket released, July 31, 1996 at 7:40 a.m., the top of the sludge was at the 320 ml mark. Given this volume and time data, an approximate generation rate of 1.9 ml/hr was calculated for Sample 96-05. This gas generation rate should be considered a minimum since some gas could have been released from the sludge without being observed. Since the gas generated in Sample 96-06 released through the sludge layer, the volume of gas generated by Sample 96-06 during this time is not known, and therefore a generation rate cannot be calculated for this sample during this period. The Fuel Characterization Project group representative observed bubbles releasing from the sludge in Sample 96-08 while it was in the graduated cylinder several weeks after the settling study was completed. This suggests Sample 96-08 was generating gas, but at a slower rate than Samples 96-05 and 96-06. No gas bubbles were observed releasing from Sample 96-01.

B.2 GAS COLLECTION AND ANALYSIS

A sampling system was designed and used to vent the five unopened shipping canisters and collect the vented gas. The system included a stainless steel tool with an O-ring seal to seat around the shipping canister vent tube with an internal slot for rotating the venting pin. Any pressurized gas was transferred to a 500 ml Tedlar bag through approximately 2 ft of food grade Tygon tubing. A 1/3 psi pressure relief valve was added to the line to prevent pressurization of the Tedlar bag. The results of this venting are summarized in Table B1. Pressurization was noted in canisters 96-11, 96-13, and 96-15. Sufficient pressure existed in 96-13 and 96-15 to collect gas in the Tedlar bag. Gas collected from shipping canisters 96-13 and 96-15 were speciated by isotopic mass spectrometry (IMS). Results of this speciation are presented in Table B2.

It was decided that the most expedient way to collect gas samples from the graduated cylinders in use for the settling studies was to seal the graduated cylinders with a rubber stopper. Off-gas was collected using a Tedlar bag attached to a piece of Tygon tubing run through a hole in the rubber stopper. On August 14, 1996 at approximately 6 p.m., stoppers fitted with Tedlar bags were sealed on the graduated cylinders containing the settled from Samples 96-05 and 96-06. The gas collection from these samples was discontinued at approximately 9 a.m. on August 19, 1996. The volumes of gas collected were estimated at approximately 40 to 75 ml from Sample 96-05 and 75 to 150 ml from Sample 96-06. Using these volumes, the gas generation rates were estimated at 0.4 to 0.7 ml/hr for Sample 96-05 and 0.7 to 1.4 ml/hr for Sample 96-06. Gas collected from each of the graduated cylinders was speciated by IMS. Results of this speciation are presented in Table B3.

On August 20, 1996 at approximately 4 p.m., the graduated cylinders containing the settled samples from 96-05 and 96-06 were sparged with helium gas for 5 min. During this sparging, the sludge layer was mobilized and the samples appeared well mixed. Following sparging, the cylinders were sealed with stoppers fitted with Tedlar bags. The gas collection from these samples was discontinued on the morning of August 22, 1996 at approximately 8 a.m. The volumes of gas collected were estimated at approximately 15 to 30 ml from Sample 96-05 and 40 to 75 ml from Sample 96-06. Using these volumes, the gas generation rates were estimated at between 0.4 to 0.8 ml/hr for Sample 96-05 and between 1.0 and 1.9 ml/hr for Sample 96-06. Given the error in the estimated volumes, approximately ±50%, the rates before and after sparging with helium are not significantly different. The gas collected from Sample 96-06 was speciated by IMS. Results of this speciation are presented in Table B3. Since the volume of gas collected from Sample 96-05 was small compared to the air contained in the head space of the graduated cylinder at the start of gas collection, it was estimated that greater than 80% of the gas collected from Sample 96-05 was air, and therefore, this sample was not submitted for IMS.

On August 30, 1996 a gas pocket was observed at the bottom of Sample 96-06. Time lapse video images were collected as the pocket grew pushing up the overlying sludge. At 10 a.m. the top of the gas pocket (bottom of the overlying sludge) was at the 130 ml level on the graduated cylinder. The pocket continued to grow until September 3, 1996 when the operator vented the pocket to prevent the overlying sludge from reaching the top of the cylinder. At 9 a.m. on September 3, 1996 the top of the gas pocket was at the 615 ml level. Given this volume and time data, an approximate generation rate of 5.1 ml/hr was calculated for Sample 96-06.

The solids from Samples 96-05 and 96-01 were transferred from the graduated cylinders to glass jars during the week of September 11, 1996. The solids from Sample 96-05 were still generating gas at the time of the transfer. Once samples were transferred to glass jars it was not possible to view gas generation. The solids from Sample 96-08 were transferred to a glass jar on October 24, 1996 and the solids from Sample 96-06 on November 6, 1996. The solids from Sample 96-06 were still generating gas at the time of transfer.

B.3 SECOND SETTLING GROUP

Solids from the remaining five samples (96-04, 96-09, 96-11, 96-13, and 96-15) were transferred out of the stainless steel shipping containers and into graduated cylinders between August 26, 1996 and October 4, 1996. Upon transfer to the graduated cylinders, gas bubbles were observed releasing from the solids in Samples 96-13 and 96-15. Gas generation in the solids from Sample 96-13 appeared greater than in Sample 96-15; however, generation rates were not measured.

The final five graduated cylinders were sparged with helium on October 9, 1996 for a minimum of 5 min. to mobilize the solid layer and obtain a homogeneous slurry. Three video cameras were used to collect images both real time and time lapse as the slurry settled. Large gas pocket formations were not observed in the solids from Sample 96-13 or 96-15, and no bubble releases were observed from Samples 96-04, 96-09, or 96-11 solids. Solids from Samples 96-04 and 96-11 were transferred from the 2 L graduated cylinders to glass jars on November 6, 1996. Solids from Samples 96-09, 96-13, and 96-15 were transferred from the graduated cylinders to glass jars between October 25, 1996 and October 29, 1996. Samples 96-13 and 96-15 were still generating gas when transferred to the glass jars. Table B3 presents a timeline of significant events covered in this report.

B.4 CALCULATIONS

Gas sample IMS results in Table B3 show the presence of argon, which is a component of ambient air and would not be generated by the K Basin samples. Using the "U.S. Standard Atmosphere, 1976" values for air concentrations (N_2 = 78.08%, O_2 = 20.95%, Ar = 0.934%, and CO_2 = 0.0314%) it is possible to subtract out the air contribution to the gas sample results. In this calculation, the argon concentration is used to determine the percent air in the gas sample. Once the air contribution is known, the contribution of N_2, O_2, and CO_2 from air can be calculated and subtracted. For example, the gas sample collected from Sample 96-05 following air sparging and settling contained 0.5% argon. The concentration of argon in air is 0.935%, therefore the fractional air content of the sample is 0.5/0.934 = 0.54. The fractional air content is then multiplied by the O_2, N_2, and CO_2 concentrations in standard air to determine the contribution of these gases in the sample from intruded air, O_2 = 20.95% x 0.54 = 11%, N_2 = 78.08% x 0.54 = 42%, and CO_2 = 0.0314% x 0.54 = 0.017%.

Subtraction of air from the results in Table B3 yielded negative concentrations for both O_2 and N_2, in all samples indicating consumption of these gases. To normalize the remaining gases to 100%, both O_2 and N_2 were set to zero. While O_2 and N_2 are consumed in the system, they are not a net product and therefore removing them from the calculation is appropriate. In addition, helium introduced to the cell air during sparging operations was set to zero prior to normalization. The results following the air and helium removal calculation are listed in Table B4.

B.5 DISCUSSION

The results of the off-gas analysis strongly suggest an oxidation process is occurring in four of the nine K East samples delivered to the 325A HLRF. These four samples include 96-05, 96-06, 96-13, and 96-15. Sufficient gas was collected from each of these four samples to perform speciation studies. Results of work conducted thus far support an oxidation of fine fuel particles. The reasons for this conclusion are listed below.

- Radiolysis of water, a mechanism initially proposed for gas generation in these samples, can be ruled out as a significant contributing mechanism. Chemical analysis has shown these samples are greater than 90% uranium oxide by weight. As a result, radiolysis and therefore gas generation should occur throughout the sample. However, gas generation was only observed at the very bottom of the solids. In addition, the predominant component of the off-gas in all samples is H_2 at approximately 98%. Radiolysis should generate H_2 and O_2 at a molar ratio of 2 to 1. While the possible presents of organics in the samples could reduce some of this oxygen, the data shows an overall O_2 consumption and not generation.

- Given the current knowledge of this system, two species could be oxidized to give H_2 as shown in Reactions 1 and 2.

$$U + 2\ H_2O \rightarrow UO_2 + 2\ H_2 \tag{1}$$

$$2\ UH_3 + 4\ H_2O \rightarrow 2\ UO_2 + 7\ H_2 \tag{2}$$

Since uranium hydride is not expected in sludge samples collected from K East Basin due to the low temperature and open canister storage (Swanson 1992), Reaction 2 should be considered unlikely. Uranium hydride formation is possible in occluded regions of cracked fuel which may be oxygen depleted favoring hydride formation. Reaction 2 is of more concern for samples taken from sealed canisters in K West Basin where fuel is stored in sealed canisters under a nitrogen cover gas, "if the moist air is a contaminant in an inert gas the amount of hydride ultimately formed may be appreciable" (Wilkinson 1962, p. 816).

- The presence of fission gases (i.e., isotopes of Kr and Xe listed in Tables B2 and B4) in the off-gas support the corrosion of spent fuel. Using an estimated Xe concentration in the fuel of 311 atomic ppm an anticipated Xe concentration of 0.016% was calculated for the off-gas.* Given the assumptions used to calculate the Xe concentration in the fuel, this value compares well with the measured values of between 0.03 and 0.08% in Table B4.

- Samples were vacuum transferred into the graduated cylinders using 0.305 in. ID stainless steel tubing; therefore, the fuel must be in the form of small particles, <0.3 in. It is likely that the fine particulate layer observed on the bottom of the shipping canisters was uranium metal. With a density of approximately 19 g/ml, fine uranium metal would be difficult to transfer by this vacuum technique. These particles could have been formed by preferential corrosion along grain boundaries or microcracks in the fuel.

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*Communication with R. B. Baker of Duke Engineering & Services Hanford, Inc. November 15, 1996.

Methane was detected in the off-gas at concentrations of up to 1.44%. Fuel used in N Reactor was a uranium alloy containing, among other components, 365 to 735 ppm carbon (Geier 1989). Methane may be formed during the corrosion; however, assuming a ratio of 2 moles of H_2 produced for every mole of U corroded, only 0.04% of the off-gas should be methane compared to the measured values of between 0.26 and 1.44% in Table B4. Determination of the additional methane source was not within the scope of this work.

The estimated gas generation rates for Samples 96-05 and 96-06 show greater gas generation for the samples once a gas pocket formed in the sludge. The generation rate in Sample 96-05 was estimated at a minimum of 1.9 ml/hr when a pocket formed and between 0.4 and 0.8 ml/hr during the gas collection in Tedlar bags. Sample 96-06 was between 1.0 and 1.9 ml/hr during collection in the Tedlar bags and increased to 5.1 ml/hr when a pocket formed. At least two explanations for this behavior include (1) the H_2 pocket forms a barrier to the transport of dissolved O_2. Oxygen retards uranium metal corrosion, and therefore, the H_2 pocket prevents dissolved O_2 from retarding the corrosion rate, (2) the corrosion generates heat which, without the gas pocket is transferred to the solution. The pocket forms an insulating barrier and allows the uranium metal to heat up increasing the rate of corrosion.

Given the many factors affecting the corrosion rate of uranium metal including (but not limited to) temperature, pH, and grain size, it is not possible to accurately estimate the H_2 generation rate from the K East canister sludge following recovery. In addition, all seven of the samples taken from canisters containing fuel elements were observed to generate gas either while in the basin or in the laboratory. This suggests that all canisters containing fuel elements contain reactive uranium metal fragments.

B.6 REFERENCES

Geier, R. G., 1989, *PUREX Technical Manual*, WHC-SP-0479, Westinghouse Hanford Company, Richland, Washington.

Swanson, J. L., 1992, "Project Plan for PNL Activities Regarding Characterization of Irradiated N Fuel," Letter Report, April 1992, Pacific Northwest Laboratory, Richland, Washington.

Wilkinson, W. D., 1962, "Uranium Metallurgy Volume II, Uranium Corrosion and Alloys," Interscience Publishers.

Table B1. Summary of Unopened Canister Venting and Sampling. (Volumes were visually estimated and are most likely accurate to within ±50%)

Sample	Vent Date	Gas Collected	Water Collected	Comment
96-04	August 12 1996	N/A*	None	Since no water was vented, this sample is assumed to have been unpressurized.
96-09	August 13, 1996	None	None	Unpressurized
96-11	August 12, 1996	None	<10 ml	Water entered tubing, but did not reach Tedlar bag indicating only minor pressurization of canister.*
96-11	August 22, 1996	None	<10 ml	Water entered tubing, but did not reach Tedlar bag, indicating only minor pressurization of canister.
96-13	August 13, 1996	200 ml	15 ml	Tedlar bag damaged before IMS analysis.
96-13	August 22, 1996	75 ml	15 ml	Unpressurized
96-15	August 13, 1996	200 ml	100 ml	Pressure relief opened during vent. Estimated gas volume 400 ml.

*Tubing detached from Tedlar bag prior to venting operation

Table B2. Results of Isotopic Mass Spectrometry on Gas Samples.
(Units are in mole percent)

Analyte	Sample 96-05 Air Sparge	Sample 96-06 Air Sparge	Sample 96-06 Helium Sparge	Sample 96-13 Shipping Canister	Sample 96-15 Shipping Canister
Argon	0.5	0.58	0.82	0.26	0.296
Carbon dioxide	0.26	0.17	0.14	0.044	0.234
Carbon monoxide	<0.01	<0.01	<0.01	<0.01	<0.01
Helium	<0.001	<0.001	0.83	0.014	<0.001
Hydrogen	56.6	53.4	12.6	72.1	75.6
Methane	0.15	0.17	0.04	0.43	1.11
Nitrogen	33.9	36.7	68.1	21.2	19.3
Oxygen	8.5	9	17.4	5.8	3.37
Ethane	0.012	0.013	<0.01	0.024	0.062
Other hydrocarbons	<0.01	<0.01	0.01	0.03	0.03
Krypton-84	0.002	0.002	0.0014	0.002	<0.0005
Krypton-85	<0.0005	0.003	<0.0005	<0.0005	<0.0005
Krypton-86	0.003	<0.0005	0.0017	0.003	<0.0005
Xenon-131	0.004	0.003	0.0017	0.004	0.0023
Xenon-132	0.006	0.005	0.002	0.005	0.0038
Xenon-134	0.009	0.007	0.003	0.008	0.0064
Xenon-136	0.013	0.01	0.004	0.012	0.0091

Table B3. Timeline for Gas Related Events during Characterization of K East Canister Sludge Samples. (All events occurred during 1996)

Date	Event
June 11-20, 1996	Nine K East Canister Sludge samples delivered to the 325A.
July 11-12, 1996	Samples 96-05, 96-01, 96-06, and 96-08 transferred out of shipping canisters. Canister Samples 96-05 and 96-06 were pressurized. Within a few days bubbles were observed releasing from Samples 96-05 and 96-06.
July 29, 1996	Samples 96-05, 96-01, 96-06, and 96-08 were air sparged.
July 29-31, 1996	Gas generation observed in Samples 96-05 and 96-06. The gas generated in Sample 96-05 was retained in the sludge while the gas generated in Sample 96-06 was released. A generation rate of 1.9 ml/hr was calculated for Sample 96-05.
August 12-22, 1996	Shipping canister Samples 96-04, 96-09, 96-11, 96-13, and 96-15 were vented. Significant pressurization was noted in canister Samples 96-13 and 96-15.
August 14-19, 1996	Gas collected from the top of the graduated cylinder Samples 96-05 and 96-06. Estimated generation rates were 0.4 and 0.7 ml/hr for Sample 96-05 and 0.7 and 1.4 ml/hr for Sample 96-06.
August 20-22, 1996	Graduated cylinder Samples 96-05 and 96-06 were sparged with helium and then gas collected. Gas generation rates were estimated at between 0.4 and 0.8 ml/hr for Sample 96-05 and between 1.0 and 1.9 ml/hr for Sample 96-06.
August 30 - September 3, 1996	Gas pocket formation in Sample 96-06. A generation rate of 5.1 ml/hr was calculated.
September 25 - October 4, 1996	Samples 96-04, 96-09, 96-11, 96-13, and 96-15 were transferred out of the stainless steel shipping containers. Gas generation observed in Samples 96-13 and 96-15.

Table B4. Gas Sample Isotopic Mass Spectrometry Results Following the Air Removal Calculation.

Analyte	Sample 96-05 Air Sparge	Sample 96-06 Air Sparge	Sample 96-06 Helium Sparge	Sample 96-13 Shipping Canister	Sample 96-15 Shipping Canister
Argon	0	0	0	0	0
Carbon dioxide	0.43	0.28	0.90	0.049	0.29
Carbon monoxide	0	0	0	0	0
Helium	0	0	0	0.	0
Hydrogen	99	99	99	99	98
Methane	0.26	0.32	0.31	0.59	1.44
Nitrogen	0	0	0	0	0
Oxygen	0	0	0	0	0
Ethane	0.02	0.024	ND	0.033	0.080
Other hydrocarbons	ND	ND	0.078	0.041	0.039
Krypton-84	0.004	0.004	0.0011	0.003	ND
Krypton-85	ND	0.006	ND	ND	ND
Krypton-86	0.005	ND	0.013	0.004	ND
Xenon-131	0.007	0.006	0.013	0.006	0.003
Xenon-132	0.011	0.009	0.016	0.007	0.005
Xenon-134	0.016	0.013	0.023	0.011	0.008
Xenon-136	0.023	0.019	0.031	0.017	0.012

ND = Not detected.

APPENDIX C

CHEMISTRY OF CENTRIFUGED K EAST CANISTER SLUDGE

T. L. Welsh

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APPENDIX C

CHEMISTRY OF CENTRIFUGED K EAST CANISTER SLUDGE

The 222-S analytical data presented in the following tables were generated using an electronic transfer from LABCORE, the 222-S Laboratory database. The PNNL analytical data presented in the following tables were obtained from Silvers 1997. The sample numbers, used in the following tables, are defined as follows;

Sludge Sample Number	K East Cubical Position	Canister Barrel	Number of Fuel Elements in Barrel
96-01	1845	East	5
96-05	3128	West	6
96-08	2350	East	7
96-09	4638	East	0
96-13	5055	West	6
96-15	6070	West	7
96-04 Upper layer	2711	East	7
96-04 Lower layer			
96-06 Upper layer	5465	West	6
96-06 Middle layer			
96-06 Lower layer			
96-11 Upper layer	6073	West	0
96-11 Lower layer			

The tables in this appendix do not include the duplicate analyses data which were performed according to the SAP (Welsh et. al. 1996). If both the analytical result and the duplicate result were "real" numbers, then the two values were averaged. If the analytical result was a "real" number and the duplicate result was a "less than" number, then the "real" number was reported. If the analytical result was a "less than" number and the duplicate result was a "real" number, then the "real" number was reported. If both the analytical result and the duplicate result were both "less than" numbers, then the maximum value of the "less thans" was reported.

222-S Laboratory

Thirteen "sludge" samples were shipped to 222-S Laboratory on a centrifuged sludge basis. Research Sample 96-06 Upper layer, consisted of a small quantity of material; therefore not all analyses specified in the SAP were performed. Inorganic analyses (icp, total uranium by phosphorescence, %water by gravimetric, caustic demand, and TIC), organic analyses (TOC and total carbon), and radiochemistry analyses (total alpha, total beta, gea, ^{99}Tc, $^{89/90}$Sr, ^{238}Pu, $^{239/240}$Pu, ^{241}Am, $^{243/244}$Cm, ^{237}Np) were performed on these samples.

Seven "supernate" samples, obtained from the settled solids graduate cylinders, were shipped to 222-S for IC analyses. The supernate represents the water digest (the usual sample preparation for IC analyses) of the settled sludge. For the IC analytical results, the conversion of $\mu g/ml_{liquid}$ where the liquid is the supernate from the as-settled sludge cylinder to $\mu g/g$ centrifuged sludge utilized the following formula.

$$\frac{\mu g_{analyte}}{g_{centrifuged\ sludge}} = \frac{\mu g_{analyte}}{mL_{liquid}} \times \frac{1}{\rho_{liquid}} \times \frac{g_{liquid}}{g_{water}} \times \frac{g_{water}}{g_{centrifuged\ sludge}} \qquad (1)$$

where:

ρ_{liquid} is the density of the liquid (mean density is 0.995 g_{liquid}/ml_{liquid} from six PNNL measurements; Table D-1, Appendix D),

g_{liquid} is the g_{water} plus the g_{TDS} (TDS is total dissolved solids; the ratio g_{liquid}/g_{water} using the Sample 96-05 cylinder analytical results is 1.0004, the ratio g_{liquid}/g_{water} using the Sample 96-01 cylinder analytical results is 1.0002),

$g_{water}/g_{centrifuged\ sludge}$ is the percent water divided by 100.

The 222-S percent water results (gravimetric) are listed in the following tables. The IC $\mu g/ml_{liquid}$ analytical results are listed in Appendix D.

PNNL

Two samples were selected for accelerated laboratory analyses; Sample 96-01 (non bubbler) and Sample 96-05 (bubbler). The laboratory analyses for these accelerated samples included icp/ms, gea, total alpha, total beta, and total uranium. The samples were analyzed on a dried sludge basis.

Ten additional samples (one subsample from each of the six non-research sludge samples and four subsamples from the different layers of the three research samples) were analyzed at PNNL for total uranium and uranium isotopic analyses on a dried sludge basis. There was not enough sample for PNNL to analyze subsamples from all the layers from each of the research sludge samples. Percent water (TGA) and DSC results were obtained for the six non-research samples on an as-settled sludge basis.

The conversion of dried sludge results to centrifuged sludge results utilized the formulas provided in Appendix D and Appendix E. The dried sludge results were first converted to as-settled sludge using Equation 2 of Appendix E. The as-settled sludge results were then converted to centrifuged sludge results using Equation 5 of Appendix D. The water sample results are provided in Appendix D. The water samples were not analyzed using the accelerated sample analytical procedures. Therefore, the accelerated sample results were not converted to a centrifuged sludge basis. The TGA and DSC results were not converted to a centrifuged sludge basis.

The PNNL uranium isotopic results were reported in atom %. The units for the uranium isotopics were changed to μg/g centrifuged sludge using the following formula.

For each sample, $\bar{A}_u$ was calculated using Equation 2.

$$\overline{A_u} = \begin{matrix} 233.04\, f_{^{233}U} + 234.04\, f_{^{234}U} + 235.04\, f_{^{235}U} \\ + 236.05\, f_{^{236}U} + 238.05\, f_{^{238}U} \end{matrix} \qquad (2)$$

where:

$$f_{u_i} = \frac{\text{atom\%}u_i}{100} \qquad (3)$$

Equation 4 was then used to convert the units, each isotope separately, to g/g centrifuged sludge.

$$C_{U_i} = C_U \frac{A_i}{\overline{A_u}} \frac{\%i}{100} \qquad (4)$$

where:

$U_1 = {}^{233}U$, $U_2 = {}^{234}U$, $U_3 = {}^{235}U$, $U_4 = {}^{236}U$, $U_5 = {}^{238}U$,

$A_1 = 233.04$, $A_2 = 234.04$, $A_3 = 235.04$, $A_4 = 236.05$, $A_5 = 238.05$,

$\%_i$ is the atom% for the uranium isotope of interest,

C_u is the total uranium concentration (C), as measured by PNNL (units of μg/g centrifuged sludge), for the sample of interest.

	Alpha Total	^{241}Am.aea	^{241}Am.gea	Beta Total	^{212}Bi.gea
Sample	µCi/g	µCi/g	µCi/g	µCi/g	µCi/g
96-01	1.14E-01	9.43E-02	< 1.26E+00	3.15E+02	< 5.77E-01
96-05	1.96E+02	8.06E+01	1.16E+02	4.05E+03	< 1.03E+01
96-08	5.96E+01	2.87E+01	3.37E+02	9.86E+02	< 9.60E-01
96-09	1.07E+01	4.96E+00	6.25E+00	6.08E+01	< 2.15E-01
96-13	1.35E+02	< 1.81E+01	6.01E+01	3.48E+03	< 9.60E+00
96-15	1.22E+02	5.20E+01	6.15E+01	3.49E+03	< 1.53E+01
96-04 U	6.23E+01	3.98E+01	3.38E+01	7.39E+02	< 2.30E+00
96-04 L	1.24E+02	6.70E+01	7.61E+01	1.73E+03	< 4.50E+00
96-06 U	1.52E+02	6.49E+01	7.06E+01	3.28E+03	< 1.68E+01
96-06 M	1.25E+02	6.41E+01	7.89E+01	2.63E+03	< 7.80E+00
96-06 L	1.52E+02	6.61E+01	8.36E+01	3.96E+03	< 1.56E+01
96-11 U	7.72E+00	6.38E+00	4.70E+00	1.39E+01	< 1.39E-01
96-11 L	1.00E+01	5.04E+00	6.22E+00	1.23E+02	< 9.94E-01

	^{144}Ce/Pr.gea	$^{243/244}$Cm.aea	^{57}Co.gea	^{60}Co.gea	^{134}Cs.gea
Sample	µCi/g	µCi/g	µCi/g	µCi/g	µCi/g
96-01	< 1.78E+00	< 1.13E-02	< 1.18E-01	< 3.41E-02	< 1.36E-01
96-05	< 2.15E+01	< 6.90E+00	< 6.86E+00	7.76E-01	< 1.56E+00
96-08	< 1.73E+00	< 2.73E+00	< 1.76E+00	2.57E-01	< 1.36E-01
96-09	< 4.12E-01	< 5.12E-01	< 3.52E-01	4.68E-01	< 3.37E-02
96-13	< 1.83E+01	< 1.81E+01	< 3.80E+00	< 7.56E-01	< 1.28E+00
96-15	< 2.82E+01	< 3.59E+00	< 3.58E+00	< 1.45E+00	< 2.00E+00
96-04 U	< 6.46E+00	< 3.75E+00	< 1.99E+00	4.54E-01	< 4.88E-01
96-04 L	< 1.22E+01	< 1.01E+01	2.30E-02	8.52E-01	< 9.38E-01
96-06 U	< 2.70E+01	< 4.41E+00	< 5.48E+00	< 1.34E+00	< 1.85E+00
96-06 M	< 1.22E+01	< 1.41E+01	< 4.06E+00	< 6.77E-01	< 8.66E-01
96-06 L	< 2.59E+01	< 1.59E+01	2.94E+00	< 1.27E+00	< 1.85E+00
96-11 U	< 1.51E-01	< 6.18E-01	< 1.21E+01	1.25E-01	< 1.34E-01
96-11 L	< 1.81E+00	< 8.15E-01	< 4.11E-01	6.94E-01	< 1.47E-01

µCi/g: µCi/g centrifuged sludge

	^{137}Cs.gea	^{152}Eu.gea	^{154}Eu.gea	^{155}Eu.gea	^{94}Nb.gea
Sample	µCi/g	µCi/g	µCi/g	µCi/g	µCi/g
96-01	2.53E+02	< 8.33E-02	< 1.26E-01	< 4.93E-01	< 3.74E-02
96-05	9.94E+02	< 2.85E+00	1.62E+01	1.06E+01	< 1.02E+00
96-08	4.38E+02	< 9.64E-02	3.92E+00	1.95E+00	< 8.45E-02
96-09	5.29E+01	< 4.68E-02	8.02E-01	3.05E-01	< 2.34E-02
96-13	5.38E+02	< 3.37E+00	7.62E+00	< 5.49E+00	< 8.07E-01
96-15	6.91E+02	< 5.12E+00	8.22E+00	< 8.53E+00	< 1.41E+00
96-04 U	3.52E+02	< 7.24E-01	4.36E+00	< 2.55E+00	< 2.51E-01
96-04 L	6.19E+02	< 1.18E+00	1.07E+01	5.70E+00	< 5.95E-01
96-06 U	6.26E+02	< 6.30E+00	1.17E+01	< 8.24E+00	< 1.41E+00
96-06 M	2.47E+02	< 2.47E+00	8.49E+00	< 5.41E+00	< 8.42E-01
96-06 L	6.81E+02	< 5.63E+00	9.50E+00	< 7.43E+00	< 1.57E+00
96-11 U	4.87E+00	< 3.92E-02	6.28E-01	2.67E-01	< 2.09E-02
96-11 L	7.84E+01	< 2.28E-01	8.75E-01	< 5.16E-01	< 9.73E-02

	^{237}Np	^{238}Pu	$^{239/240}$Pu	^{226}Ra.gea	^{106}Ru/Rh.gea
Sample	µCi/g	µCi/g	µCi/g	µCi/g	µCi/g
96-01	< 5.60E-03	4.14E-03	3.85E-02	< 3.68E+00	< 2.69E+00
96-05	6.83E-03	1.41E+01	1.33E+02	< 4.18E+01	< 3.16E+01
96-08	< 3.95E-03	6.46E+00	3.47E+01	< 3.50E+00	< 2.66E+00
96-09	< 3.81E-03	1.13E+00	6.52E+00	< 8.50E-01	< 6.43E-01
96-13	1.51E-02	< 5.88E+01	9.21E+01	< 3.43E+01	< 2.65E+01
96-15	1.09E-02	7.18E+00	8.35E+01	< 5.28E+01	< 3.90E+01
96-04 U	< 7.02E-03	7.36E+00	3.44E+01	< 1.23E+01	< 9.71E+00
96-04 L	6.94E-03	1.37E+01	6.30E+01	< 2.27E+01	< 1.77E+01
96-06 U	1.37E-02	1.31E+01	9.48E+01	< 5.11E+01	< 3.61E+01
96-06 M	1.50E-02	1.46E+01	8.22E+01	< 2.28E+01	< 1.68E+01
96-06 L	1.05E-02	1.40E+01	8.10E+01	< 5.08E+01	< 3.87E+01
96-11 U	4.07E-03	8.62E-01	4.52E+00	< 3.02E-01	< 2.37E-01
96-11 L	< 6.68E-03	1.22E+00	5.99E+00	< 3.73E+00	< 2.74E+00

µCi/g: µCi/g centrifuged sludge

	$^{89/90}$Sr	^{99}Tc	^{208}Tl.gea	Total Carbon	TOC
Sample	µCi/g	µCi/g	µCi/g	µg/g	µg/g
96-01	4.86E+01	2.26E-01	< 2.44E+00	2.28E+04	1.05E+04
96-05	1.91E+03	5.63E+00	< 2.66E+01	9.37E+02	5.77E+02
96-08	3.49E+02	1.27E+01	< 2.29E+00	1.77E+03	4.32E+02
96-09	5.39E+00	5.22E+00	< 5.59E-01	1.98E+03	1.60E+03
96-13	1.48E+03	8.54E+00	< 2.22E+01	8.17E+02	4.52E+02
96-15	1.38E+03	1.08E+00	< 3.32E+01	8.71E+02	5.66E+02
96-04 U	1.09E+02	NA	< 8.04E+00	1.41E+03	6.44E+02
96-04 L	6.31E+02	NA	< 1.49E+01	2.76E+03	5.98E+02
96-06 U	1.43E+03	NA	< 3.09E+01	7.58E+02	4.51E+02
96-06 M	1.41E+03	NA	< 1.43E+01	6.17E+02	5.11E+02
96-06 L	1.73E+03	NA	< 3.31E+01	5.52E+02	3.73E+02
96-11 U	3.62E+00	NA	< 2.00E-01	9.33E+02	5.11E+02
96-11 L	2.17E+01	NA	< 2.40E+00	2.29E+03	1.47E+03

	TIC	Ag.icp	Al.icp	B.icp	Ba.icp
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	1.10E+04	1.40E+01	2.30E+04	1.59E+02	< 1.97E+01
96-05	2.51E+02	1.11E+02	1.15E+04	1.35E+02	1.16E+02
96-08	8.81E+02	1.88E+01	2.92E+04	1.39E+02	5.85E+01
96-09	8.06E+02	< 3.95E+00	2.67E+04	9.08E+01	2.91E+01
96-13	3.41E+02	1.30E+02	1.21E+04	1.39E+02	1.09E+02
96-15	4.20E+02	1.71E+02	1.40E+04	1.58E+02	1.54E+02
96-04 U	7.47E+02	9.77E+00	2.33E+04	9.66E+01	6.04E+01
96-04 L	2.26E+03	3.27E+01	5.61E+04	1.18E+02	1.03E+02
96-06 U	2.78E+02	9.12E+01	9.47E+03	1.29E+02	1.17E+02
96-06 M	4.61E+02	1.25E+02	1.15E+04	1.28E+02	< 1.01E+02
96-06 L	2.64E+02	1.39E+02	9.08E+03	1.34E+02	1.09E+02
96-11 U	7.17E+02	2.32E+00	1.38E+04	1.00E+02	< 9.95E+00
96-11 L	8.89E+02	< 7.85E+00	2.28E+04	8.30E+01	5.91E+01

µCi/g: µCi/g centrifuged sludge
µg/g: µg/g centrifuged sludge
NA: not analyzed

	Be.icp	Bi.icp	Ca.icp	Cd.icp	Cr.icp
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	< 1.97E+00	< 3.94E+01	3.17E+02	5.98E+00	1.77E+03
96-05	< 9.89E+00	5.75E+02	6.79E+02	2.05E+01	< 1.98E+01
96-08	2.10E+01	1.27E+02	5.22E+02	1.52E+01	1.13E+02
96-09	1.94E+01	3.95E+01	3.09E+02	2.80E+01	1.19E+02
96-13	< 9.29E+00	5.54E+02	5.83E+02	1.69E+01	< 1.86E+01
96-15	< 9.85E+00	3.76E+02	< 1.97E+02	< 9.85E+00	< 1.97E+01
96-04 U	1.09E+01	6.62E+01	3.85E+02	1.55E+01	3.93E+01
96-04 L	1.12E+01	2.01E+02	7.91E+02	3.09E+01	7.59E+01
96-06 U	< 1.06E+01	5.60E+02	4.81E+02	< 1.06E+01	< 2.12E+01
96-06 M	< 1.01E+01	2.35E+02	< 2.03E+02	1.83E+01	< 2.03E+01
96-06 L	< 1.00E+01	2.57E+02	< 2.00E+02	2.09E+01	< 2.00E+01
96-11 U	1.86E+01	< 1.99E+01	1.12E+02	2.27E+01	6.26E+01
96-11 L	2.61E+01	< 7.85E+01	8.59E+02	6.38E+01	2.79E+02

	Cu.icp	Fe.icp	K.icp	Mg.icp	Mn.icp
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	7.31E+00	3.53E+02	2.76E+03	< 3.94E+01	1.08E+02
96-05	8.18E+01	6.09E+03	< 9.89E+02	1.93E+03	< 1.98E+01
96-08	9.49E+01	2.70E+04	< 3.07E+02	6.22E+02	3.41E+01
96-09	2.46E+02	4.04E+04	< 1.97E+02	3.80E+02	1.34E+02
96-13	6.74E+01	2.35E+03	< 9.29E+02	1.59E+03	< 1.86E+01
96-15	5.96E+01	5.74E+03	< 9.85E+02	1.69E+03	< 1.97E+01
96-04 U	8.35E+01	1.45E+04	< 1.98E+02	2.56E+02	3.61E+01
96-04 L	1.36E+02	2.99E+04	< 4.16E+02	7.81E+02	5.81E+01
96-06 U	7.54E+01	1.37E+03	< 1.06E+03	1.35E+03	< 2.12E+01
96-06 M	7.04E+01	1.43E+03	< 1.01E+03	< 2.03E+02	< 2.03E+01
96-06 L	6.10E+01	1.04E+03	< 1.00E+03	< 2.00E+02	< 2.00E+01
96-11 U	1.17E+02	1.20E+04	< 9.95E+01	< 1.99E+01	8.02E+01
96-11 L	2.60E+02	8.92E+04	< 3.93E+02	1.03E+02	1.94E+02

µg/g: µg/g centrifuged sludge

	Na.icp	Ni.icp	P.icp	Pb.icp	Se.icp
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	1.92E+05	1.17E+02	1.70E+03	5.20E+01	< 3.94E+01
96-05	3.63E+02	< 3.96E+01	< 3.96E+02	< 1.98E+02	< 1.98E+02
96-08	2.70E+02	2.29E+01	< 1.23E+02	6.18E+01	< 6.13E+01
96-09	1.58E+02	9.56E+01	1.81E+02	1.47E+02	< 3.95E+01
96-13	3.59E+02	< 3.71E+01	< 3.71E+02	< 1.86E+02	< 1.86E+02
96-15	4.12E+02	< 3.94E+01	< 3.94E+02	< 1.97E+02	< 1.97E+02
96-04 U	1.82E+02	5.35E+01	< 7.90E+01	6.58E+01	< 3.95E+01
96-04 L	3.28E+02	4.55E+01	< 1.66E+02	8.54E+01	< 8.32E+01
96-06 U	4.66E+02	< 4.25E+01	< 4.25E+02	< 2.12E+02	< 2.12E+02
96-06 M	3.50E+02	< 4.05E+01	< 4.05E+02	< 2.03E+02	< 2.03E+02
96-06 L	3.50E+02	< 4.01E+01	< 4.01E+02	< 2.00E+02	< 2.00E+02
96-11 U	1.73E+02	2.03E+01	1.12E+02	6.31E+01	< 1.99E+01
96-11 L	2.01E+02	4.49E+01	2.14E+02	1.79E+02	< 7.85E+01

	Sm.icp	Tl.icp	U.icp	U.phos	U.las
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	< 3.94E+01	< 7.88E+01	< 1.97E+02	7.12E+02	6.32E+05
96-05	< 1.98E+02	< 3.96E+02	6.52E+05	5.10E+05	7.68E+05
96-08	< 6.13E+01	< 1.23E+02	1.43E+05	1.15E+05	1.50E+05
96-09	< 3.95E+01	< 7.90E+01	2.51E+04	2.51E+04	1.69E+05
96-13	< 1.86E+02	< 3.71E+02	6.24E+05	6.18E+05	6.85E+05
96-15	< 1.97E+02	< 3.94E+02	5.85E+05	4.30E+05	7.11E+05
96-04 U	< 3.95E+01	< 7.90E+01	5.76E+04	5.37E+04	NA
96-04 L	< 8.32E+01	< 1.66E+02	2.01E+05	1.73E+05	3.45E+05
96-06 U	< 2.12E+02	< 4.25E+02	5.97E+05	4.71E+05	NA
96-06 M	< 2.03E+02	< 4.05E+02	5.95E+05	4.85E+05	6.56E+05
96-06 L	< 2.00E+02	< 4.01E+02	6.45E+05	5.29E+05	7.35E+05
96-11 U	< 1.99E+01	< 3.98E+01	1.32E+04	1.26E+04	NA
96-11 L	< 7.85E+01	< 1.57E+02	2.71E+04	2.55E+04	3.24E+04

µg/g: µg/g centrifuged sludge
NA: not analyzed

	Zn.icp	Zr.icp	%Water.grav	%Water.grav	Density centrifuged sludge
Sample	µg/g	µg/g	wt%	µg/g	g/ml
96-01	1.85E+01	9.19E+00	3.49E+01	3.49E+05	2.56
96-05	4.55E+01	< 1.98E+01	1.61E+01	1.61E+05	2.62
96-08	1.74E+02	1.22E+02	6.67E+01	6.67E+05	1.53
96-09	2.67E+02	1.07E+02	7.96E+01	7.96E+05	1.20
96-13	1.86E+01	< 1.86E+01	1.54E+01	1.54E+05	4.52
96-15	4.38E+01	5.87E+01	1.85E+01	1.85E+05	2.82
96-04 U	1.25E+02	1.61E+02	8.19E+01	8.19E+05	1.323 #
96-04 L	1.72E+02	2.18E+02	4.36E+01	4.36E+05	1.96
96-06 U	2.15E+01	< 2.12E+01	INS	INS	2.51
96-06 M	< 2.03E+01	< 2.03E+01	2.19E+01	2.19E+05	2.36
96-06 L	< 2.00E+01	< 2.00E+01	1.47E+01	1.47E+05	3.71
96-11 U	1.25E+02	7.80E+01	9.01E+01	9.01E+05	1.27
96-11 L	4.95E+02	9.93E+01	6.98E+01	6.98E+05	1.38

	^{233}U.tims	^{234}U.tims	^{235}U.tims	^{236}U.tims	^{238}U.tims
Sample	atom%	atom%	atom%	atom%	atom%
96-01	< 0.0005	0.00945	0.9545	0.0535	98.9825
96-05	< 0.0005	0.00510	0.4970	0.0390	99.4590
96-08	< 0.0005	0.00800	0.6570	0.0820	99.2535
96-09	< 0.0005	0.00730	0.7020	0.0750	99.2150
96-13	< 0.0005	0.00880	0.7315	0.0650	99.1945
96-15	< 0.0005	0.00700	0.7175	0.0455	99.2300
96-04 U	NA	NA	NA	NA	NA
96-04 L	< 0.0005	0.00755	0.6070	0.0975	99.2880
96-06 U	NA	NA	NA	NA	NA
96-06 M	< 0.0005	0.01065	0.7490	0.0805	99.1600
96-06 L	< 0.0005	0.00980	0.7480	0.0830	99.1590
96-11 U	NA	NA	NA	NA	NA
96-11 L	< 0.0005	0.01475	0.7255	0.0765	99.1825

µg/g: µg/g centrifuged sludge
INS: insufficient sample
NA: not analyzed
#: Upper/lower interface layer; not upper layer

	^{233}U.tims $	^{234}U.tims $	^{235}U.tims $	^{236}U.tims $	^{238}U.tims $
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	< 3.09E+00	5.87E+01	5.96E+03	3.35E+02	6.26E+05
96-05	< 3.76E+00	3.85E+01	3.77E+03	2.97E+02	7.63E+05
96-08	< 7.34E-01	1.18E+01	9.73E+02	1.22E+02	1.49E+05
96-09	< 8.28E-01	1.21E+01	1.17E+03	1.26E+02	1.68E+05
96-13	< 3.35E+00	5.93E+01	4.95E+03	4.41E+02	6.79E+05
96-15	< 3.48E+00	4.89E+01	5.04E+03	3.21E+02	7.05E+05
96-04 U	NA	NA	NA	NA	NA
96-04 L	< 1.69E+00	2.56E+01	2.07E+03	3.33E+02	3.42E+05
96-06 U	NA	NA	NA	NA	NA
96-06 M	< 3.21E+00	6.86E+01	4.85E+03	5.23E+02	6.50E+05
96-06 L	< 3.60E+00	7.08E+01	5.43E+03	6.05E+02	7.28E+05
96-11 U	NA	NA	NA	NA	NA
96-11 L	< 1.59E-01	4.71E+00	2.32E+02	2.46E+01	3.22E+04

$: based on the total uranium (U.las µg/g centrifuged sludge) by PNNL
µg/g: µg/g centrifuged sludge
NA: not analyzed

	Br^-.ic	Cl^-.ic	F^-.ic	NO_2^-.ic	NO_3^-.ic
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-04 U	< 1.14E+00	5.25E-01	< 1.09E-01	< 9.80E-01	1.67E+00
96-04 L	< 6.06E-01	2.80E-01	< 5.79E-02	< 5.22E-01	8.91E-01
96-06 M	< 3.04E-01	1.05E+00	1.29E-01	< 2.62E-01	< 3.37E-01
96-06 L	< 2.04E-01	7.07E-01	8.63E-02	< 1.76E-01	< 2.26E-01
96-08	< 9.25E-01	4.73E-01	< 8.85E-02	< 7.98E-01	< 1.03E+00
96-09	< 1.10E+00	2.95E-01	< 1.06E-01	< 9.52E-01	< 1.22E+00
96-11 U	< 1.25E+00	4.84E-01	< 1.20E-01	< 1.08E+00	< 1.39E+00
96-11 L	< 9.68E-01	3.75E-01	< 9.26E-02	< 8.35E-01	< 1.07E+00
96-13	< 2.23E-01	1.96E-01	9.52E-02	< 1.93E-01	< 2.48E-01
96-15	< 2.56E-01	1.29E-01	< 2.45E-02	< 2.21E-01	< 2.84E-01

	Oxalate.ic	PO_4^{3-}.ic	SO_4^{2-}.ic		
Sample	µg/g	µg/g	µg/g		
96-04 U	< 9.55E-01	< 1.09E+00	2.34E+00		
96-04 L	< 5.09E-01	< 5.79E-01	1.25E+00		
96-06 M	< 2.56E-01	< 2.91E-01	1.69E+00		
96-06 L	< 1.72E-01	< 1.95E-01	1.13E+00		
96-08	< 7.78E-01	< 8.85E-01	3.57E+00		
96-09	< 9.28E-01	< 1.06E+00	2.46E+00		
96-11 U	< 1.05E+00	< 1.20E+00	< 1.38E+00		
96-11 L	< 8.14E-01	< 9.26E-01	< 1.07E+00		
96-13	< 1.88E-01	< 2.14E-01	7.17E-01		
96-15	< 2.15E-01	< 2.45E-01	1.25E+00		

µg/g: µg/g centrifuged sludge
NA: not analyzed

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APPENDIX D

CHEMISTRY OF AS-SETTLED K EAST CANISTER SLUDGE

T. L. Welsh

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APPENDIX D

CHEMISTRY OF AS-SETTLED K EAST CANISTER SLUDGE

The conversion of centrifuged sludge results to as-settled sludge results utilized the following formulas.

The assumption for converting from a centrifuged basis to an as-settled basis is as follows.

> Centrifugation of settled sludge removes δg grams of excess water containing u_w concentrations of analytes by analysis. The remaining water and solids with their respective analytes stay with the centrifuged sludge.

The following table defines the symbols used in deriving the formula to convert centrifuged-state analyses and water analyses to a settled-state.

Symbol	Meaning	Units
c	gravimetric concentration	µg/g or µCi/g
g	mass	g
u	volumetric concentration	µg/ml or µCi/ml
δ	excess	
ρ	density	g/ml
subscript $_f$	centrifuged state	
subscript $_s$	as-settled state	
subscript $_w$	water state	

By mass balance:

$$g_s - \delta g_w = g_f \tag{1}$$

and

$$c_s g_s - \frac{u_w}{\rho_w} \delta g_w = c_f g_f \tag{2}$$

or

$$c_s = c_f \frac{g_f}{g_s} + \frac{u_w}{\rho_w} \frac{\delta g_w}{g_s} \qquad (3)$$

But, by (1) we have

$$\frac{g_f}{g_s} + \frac{\delta g_w}{g_s} = 1 \qquad (4)$$

Therefore, by substitution:

$$c_s = c_f \frac{g_f}{g_s} + \frac{u_w}{\rho_w} \left(1 - \frac{g_f}{g_s} \right) \qquad (5)$$

The average density (ρ_w), as measured by PNNL on the supernate (water) samples, is equal to 0.995 g/ml (see Table D-1 below). The values for g_f/g_s are listed Table D-2. The values for g_f and g_s are provided in Silvers 1997a. The values for c_f and the sample information are listed in Appendix C. The values for u_w, the water concentration, are listed in the following tables. The water concentrations (u_w) from Sample 96-05 Cylinder were used in Equation 5 for the "bubbled" sludge samples (96-05, 96-06, 96-13, and 96-15). The water concentrations (u_w) from Sample 96-01 Cylinder were used in Equation 5 for the "non bubbled" sludge samples (96-01, 96-04, 96-08, 96-09, and 96-11).

For the IC analytical results, the conversion of $\mu g/ml_{liquid}$ where the liquid is the supernate from the as-settled sludge cylinder to $\mu g/g$ as-settled sludge utilized Equation 6. This assumes that the analytes (sulfate, phosphate, nitrate, nitrite, chloride, fluoride, oxalate, bromide) are soluble and that the analyte does not stay with the sludge.

$$\frac{\mu g_{analyte}}{g_{as\text{-}settled\ sludge}} = \frac{\mu g_{analyte}}{mL_{liquid}} \times \frac{1}{\rho_{liquid}} \times \frac{g_{liquid}}{g_{water}} \times \frac{g_{water}}{g_{as\text{-}settled\ sludge}} \qquad (6)$$

where

ρ_{liquid} is the density of the liquid (mean density is 0.995 g/ml from six PNNL measurements,

g_{liquid} is the g_{water} plus the g_{TDS} (TDS is total dissolved solids; the ratio g_{liquid}/g_{water} using the 96-05 cylinder analytical results is 1.0004),

$g_{water}/g_{\text{as-settled sludge}}$ is the percent water divided by 100.

The 222-S percent water results (gravimetric), adjusted from a centrifuged basis to an as-settled basis using the data in Table D-2, are listed in the following tables. The $\mu g/ml_{liquid}$ analytical results are listed in the following tables.

The conversion of dried sludge results to as-settled sludge results utilized Equation 2 of Appendix E. The PNNL uranium isotopic results were reported in atom% (Appendix C). The units for the uranium isotopics were changed to µg/g as-settled sludge using Equation 3 of Appendix C, where C_u is the total uranium concentration (C), as measured by PNNL laser fluorimetry (units of µg/g as-settled sludge), for the sample of interest.

The icp/ms analytical results were reported with units of µg/g sludge (dried basis converted to as-settled basis). The icp/ms analytical results for ^{239}Pu and ^{240}Pu were converted to µCi/g sludge (dried or as-settled) using the specific activity for the isotope of interest; 0.062 Ci/g for ^{239}Pu and 0.227 Ci/g for ^{240}Pu. The PNNL gea analytical results for ^{238}U were reported with units of µCi/g (dried basis converted to as-settled basis) using the specific activity of 3.36E-07 Ci/g.

The TGA and DSC subsamples were reported with units of wt% ($g/g_{\text{as-settled sludge}}$) and Joules/$g_{\text{as-settled sludge}}$. Sample 96-13 was the only subsample which exhibited an exotherm. All samples exhibited endotherms. However, since the endotherm started below the baseline an accurate result is not possible (Silvers 1997a). Therefore, the endotherms provided in this appendix have been reported as greater than values. Most of the subsamples obtained for the TGA analyses exhibited a mass loss prior to the TGA analysis itself. The TGA results reported in this appendix have accounted for the initial mass loss (data provided in Silvers 1997a) and the TGA mass loss.

Table D-1. Supernate Density Results.

Sample	Supernate Density	Sample	Supernate Density
96-01 Supernate	1.001	96-09 Supernate	0.994
96-04 Supernate	1.000	96-11 Supernate	1.001
96-05 Supernate	0.991	96-13 Supernate	0.983
96-08 Supernate	0.992	96-15 Supernate	0.997
Mean Supernate Density = 0.995			

Table D-2. Conversion Factors--Centrifuged to As-Settled.

Sample	g_f/g_s	Sample	g_f/g_s
96-01	0.8895	96-04 Upper	0.5812
96-05	0.9118	96-04 Lower	0.6429
96-08	0.6623	96-06 Upper	0.5522
96-09	0.6389	96-06 Middle	0.8302
96-13	0.9011	96-06 Lower	0.9113
96-15	0.8927	96-11 Upper	0.5299
		96-11 Lower	0.6926

	Alpha Total	Alpha Total icp/ms PNNL	^{241}Am.aea	^{241}Am.gea	^{241}Am.gea (PNNL)
Sample	µCi/g	µCi/g	µCi/g	µCi/g	µCi/g
96-01	1.01E-01	1.06E+02	8.39E-02	< 1.12E+00	2.15E-01
96-05	1.78E+02	2.14E+02	7.35E+01	1.06E+02	5.27E+01
96-08	3.95E+01	NA	1.90E+01	2.23E+02	NA
96-09	6.84E+00	NA	3.17E+00	3.99E+00	NA
96-13	1.21E+02	NA	< 1.63E+01	5.42E+01	NA
96-15	1.08E+02	NA	4.64E+01	5.49E+01	NA
96-04 U	3.62E+01	NA	2.31E+01	1.96E+01	NA
96-04 L	7.94E+01	NA	4.30E+01	4.89E+01	NA
96-06 U	8.37E+01	NA	3.59E+01	3.92E+01	NA
96-06 M	1.04E+02	NA	5.32E+01	6.56E+01	NA
96-06 L	1.38E+02	NA	6.03E+01	7.62E+01	NA
96-11 U	4.09E+00	NA	3.45E+00	2.50E+00	NA
96-11 L	6.94E+00	NA	3.54E+00	4.31E+00	NA

	Beta Total	Beta Total PNNL	^{212}Bi.gea	^{144}Ce/Pr.gea	$^{243/244}$Cm.gea
Sample	µCi/g	µCi/g	µCi/g	µCi/g	µCi/g
96-01	2.80E+02	8.67E+03	< 5.15E-01	< 1.58E+00	< 1.01E-02
96-05	3.70E+03	3.90E+03	< 9.41E+00	< 1.97E+01	< 6.29E+00
96-08	6.53E+02	NA	< 6.41E-01	< 1.15E+00	< 1.81E+00
96-09	3.89E+01	NA	< 1.43E-01	< 2.68E-01	< 3.27E-01
96-13	3.14E+03	NA	< 8.67E+00	< 1.65E+01	< 1.63E+01
96-15	3.13E+03	NA	< 1.37E+01	< 2.52E+01	< 3.21E+00
96-04 U	4.29E+02	NA	< 1.34E+00	< 3.76E+00	< 2.18E+00
96-04 L	1.11E+03	NA	< 2.90E+00	< 7.85E+00	< 6.49E+00
96-06 U	1.87E+03	NA	< 9.38E+00	< 1.52E+01	< 2.44E+00
96-06 M	2.20E+03	NA	< 6.51E+00	< 1.02E+01	< 1.17E+01
96-06 L	3.62E+03	NA	< 1.42E+01	< 2.37E+01	< 1.45E+01
96-11 U	7.43E+00	NA	< 8.08E-02	< 8.69E-02	< 3.32E-01
96-11 L	8.49E+01	NA	< 6.93E-01	< 1.26E+00	< 5.68E-01

µCi/g: µCi/g as-settled sludge
NA: not analyzed

	^{57}Co.gea	^{60}Co.gea	^{60}Co.gea PNNL	^{134}Cs.gea	^{134}Cs.gea PNNL
Sample	µCi/g	µCi/g	µCi/g	µCi/g	µCi/g
96-01	< 1.05E-01	< 3.05E-02	1.13E-02	< 1.21E-01	< 2.05E-03
96-05	< 6.26E+00	7.09E-01	3.18E-01	< 1.43E+00	1.91E-01
96-08	< 1.17E+00	1.71E-01	NA	< 9.05E-02	NA
96-09	< 2.25E-01	3.00E-01	NA	< 2.20E-02	NA
96-13	< 3.43E+00	< 6.83E-01	NA	< 1.16E+00	NA
96-15	< 3.20E+00	< 1.30E+00	NA	< 1.79E+00	NA
96-04 U	< 1.16E+00	2.64E-01	NA	< 2.84E-01	NA
96-04 L	1.51E-02	5.48E-01	NA	< 6.03E-01	NA
96-06 U	< 3.04E+00	< 7.47E-01	NA	< 1.04E+00	NA
96-06 M	< 3.38E+00	< 5.65E-01	NA	< 7.26E-01	NA
96-06 L	2.68E+00	< 1.16E+00	NA	< 1.69E+00	NA
96-11 U	< 6.41E+00	6.67E-02	NA	< 7.16E-02	NA
96-11 L	< 2.85E-01	4.81E-01	NA	< 1.02E-01	NA

	^{137}Cs.gea	^{137}Cs.gea PNNL	^{152}Eu.gea	^{154}Eu.gea	^{154}Eu.gea PNNL
Sample	µCi/g	µCi/g	µCi/g	µCi/g	µCi/g
96-01	2.25E+02	7.99E-01	< 7.48E-02	< 1.12E-01	2.99E-02
96-05	9.10E+02	4.70E+02	< 2.60E+00	1.48E+01	7.59E+00
96-08	2.90E+02	NA	< 6.60E-02	2.60E+00	NA
96-09	3.38E+01	NA	< 3.22E-02	5.13E-01	NA
96-13	4.88E+02	NA	< 3.04E+00	6.87E+00	NA
96-15	6.21E+02	NA	< 4.58E+00	7.34E+00	NA
96-04 U	2.05E+02	NA	< 4.23E-01	2.54E+00	NA
96-04 L	3.98E+02	NA	< 7.61E-01	6.88E+00	NA
96-06 U	3.62E+02	NA	< 3.51E+00	6.48E+00	NA
96-06 M	2.11E+02	NA	< 2.06E+00	7.05E+00	NA
96-06 L	6.23E+02	NA	< 5.14E+00	8.66E+00	NA
96-11 U	2.63E+00	NA	< 2.38E-02	3.34E-01	NA
96-11 L	5.43E+01	NA	< 1.60E-01	6.07E-01	NA

µCi/g: µCi/g as-settled sludge
NA: not analyzed

	^{155}Eu.gea	^{155}Eu.gea PNNL	^{94}Nb.gea	^{237}Np	^{238}Pu
Sample	µCi/g	µCi/g	µCi/g	µCi/g	µCi/g
96-01	< 4.39E-01	1.24E-02	< 3.34E-02	< 5.08E-03	3.70E-03
96-05	9.68E+00	3.83E+00	< 9.31E-01	6.31E-03	1.29E+01
96-08	1.29E+00	NA	< 5.63E-02	< 2.93E-03	4.28E+00
96-09	1.96E-01	NA	< 1.53E-02	< 2.77E-03	7.22E-01
96-13	< 4.96E+00	NA	< 7.29E-01	1.37E-02	< 5.30E+01
96-15	< 7.63E+00	NA	< 1.26E+00	9.78E-03	6.40E+00
96-04 U	< 1.48E+00	NA	< 1.46E-01	< 4.47E-03	4.28E+00
96-04 L	3.67E+00	NA	< 3.83E-01	4.79E-03	8.77E+00
96-06 U	< 4.63E+00	NA	< 7.86E-01	7.98E-03	7.23E+00
96-06 M	< 4.52E+00	NA	< 7.02E-01	1.26E-02	1.21E+01
96-06 L	< 6.79E+00	NA	< 1.43E+00	9.61E-03	1.28E+01
96-11 U	1.43E-01	NA	< 1.16E-02	2.59E-03	4.57E-01
96-11 L	< 3.59E-01	NA	< 6.77E-02	< 4.91E-03	8.45E-01

	$^{239/240}$Pu	^{239}Pu.icp/ms PNNL	^{240}Pu.icp/ms PNNL	^{226}Ra.gea	^{106}Ru/Rh.gea
Sample	µCi/g	µCi/g	µCi/g	µCi/g	µCi/g
96-01	3.42E-02	1.07E+01	< 3.11E-02	< 3.28E+00	< 2.40E+00
96-05	1.21E+02	7.71E+01	3.09E+01	< 3.82E+01	< 2.89E+01
96-08	2.30E+01	NA	NA	< 2.33E+00	< 1.77E+00
96-09	4.17E+00	NA	NA	< 5.53E-01	< 4.20E-01
96-13	8.29E+01	NA	NA	< 3.10E+01	< 2.40E+01
96-15	7.45E+01	NA	NA	< 4.73E+01	< 3.49E+01
96-04 U	2.00E+01	NA	NA	< 7.16E+00	< 5.65E+00
96-04 L	4.05E+01	NA	NA	< 1.46E+01	< 1.14E+01
96-06 U	5.23E+01	NA	NA	< 2.88E+01	< 2.03E+01
96-06 M	6.82E+01	NA	NA	< 1.91E+01	< 1.41E+01
96-06 L	7.38E+01	NA	NA	< 4.64E+01	< 3.53E+01
96-11 U	2.39E+00	NA	NA	< 1.73E-01	< 1.37E-01
96-11 L	4.15E+00	NA	NA	< 2.59E+00	< 1.91E+00

µCi/g: µCi/g as-settled sludge
NA: not analyzed

	$^{89/90}$Sr	^{99}Tc	^{208}Tl.gea	Total Carbon	TOC
Sample	µCi/g	µCi/g	µCi/g	µg/g	µg/g
96-01	4.32E+01	2.01E-01	< 2.17E+00	2.02E+04	9.34E+03
96-05	1.74E+03	5.13E+00	< 2.43E+01	8.58E+02	5.29E+02
96-08	2.31E+02	8.41E+00	< 1.52E+00	1.18E+03	2.99E+02
96-09	3.45E+00	3.33E+00	< 3.64E-01	1.28E+03	1.04E+03
96-13	1.34E+03	7.69E+00	< 2.01E+01	7.40E+02	4.11E+02
96-15	1.24E+03	9.60E-01	< 2.97E+01	7.81E+02	5.09E+02
96-04 U	6.35E+01	NA	< 4.68E+00	8.36E+02	3.91E+02
96-04 L	4.05E+02	NA	< 9.59E+00	1.79E+03	3.99E+02
96-06 U	8.09E+02	NA	< 1.74E+01	4.36E+02	2.67E+02
96-06 M	1.17E+03	NA	< 1.20E+01	5.19E+02	4.31E+02
96-06 L	1.58E+03	NA	< 3.02E+01	5.07E+02	3.43E+02
96-11 U	1.93E+00	NA	< 1.16E-01	5.13E+02	2.90E+02
96-11 L	1.50E+01	NA	< 1.67E+00	1.60E+03	1.03E+03

	TIC	Ag.icp	Al.icp	Al.icp/ms PNNL	B.icp
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	9.79E+03	1.25E+01	2.04E+04	7.79E+02	1.41E+02
96-05	2.29E+02	1.01E+02	1.05E+04	8.16E+03	1.23E+02
96-08	5.85E+02	1.25E+01	1.93E+04	NA	9.23E+01
96-09	5.17E+02	< 2.53E+00	1.71E+04	NA	5.82E+01
96-13	3.08E+02	1.17E+02	1.09E+04	NA	1.25E+02
96-15	3.75E+02	1.52E+02	1.25E+04	NA	1.41E+02
96-04 U	4.36E+02	5.68E+00	1.35E+04	NA	5.64E+01
96-04 L	1.45E+03	2.10E+01	3.60E+04	NA	7.61E+01
96-06 U	1.55E+02	5.04E+01	5.23E+03	NA	7.16E+01
96-06 M	3.83E+02	1.03E+02	9.58E+03	NA	1.06E+02
96-06 L	2.41E+02	1.27E+02	8.27E+03	NA	1.22E+02
96-11 U	3.82E+02	1.23E+00	7.29E+03	NA	5.33E+01
96-11 L	6.17E+02	< 5.44E+00	1.58E+04	NA	5.77E+01

µCi/g: µCi/g as-settled sludge
µg/g: µg/g as-settled sludge
NA: not analyzed

	Ba.icp	Be.icp	Bi.icp	Ca.icp	Ca.icp/ms PNNL
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	< 1.75E+01	< 1.75E+00	< 3.51E+01	2.82E+02	4.85E+01
96-05	1.05E+02	< 9.02E+00	5.24E+02	6.19E+02	8.62E+01
96-08	3.88E+01	1.39E+01	8.41E+01	3.46E+02	NA
96-09	1.86E+01	1.24E+01	2.53E+01	1.97E+02	NA
96-13	9.78E+01	< 8.37E+00	4.99E+02	5.25E+02	NA
96-15	1.37E+02	< 8.79E+00	3.36E+02	< 1.76E+02	NA
96-04 U	3.51E+01	6.31E+00	3.85E+01	2.24E+02	NA
96-04 L	6.63E+01	7.20E+00	1.29E+02	5.09E+02	NA
96-06 U	6.46E+01	< 5.86E+00	3.09E+02	2.66E+02	NA
96-06 M	< 8.39E+01	< 8.39E+00	1.95E+02	< 1.69E+02	NA
96-06 L	9.93E+01	< 9.11E+00	2.34E+02	< 1.82E+02	NA
96-11 U	< 5.30E+00	9.83E+00	< 1.06E+01	5.96E+01	NA
96-11 L	4.09E+01	1.81E+01	< 5.44E+01	5.95E+02	NA

	Cd.icp	Cr.icp	Cu.icp	Fe.icp	Fe.icp/ms PNNL
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	5.32E+00	1.57E+03	6.50E+00	3.14E+02	1.15E+03
96-05	1.86E+01	< 1.81E+01	7.45E+01	5.55E+03	4.95E+03
96-08	1.01E+01	7.48E+01	6.29E+01	1.79E+04	NA
96-09	1.79E+01	7.60E+01	1.57E+02	2.58E+04	NA
96-13	1.52E+01	< 1.68E+01	6.07E+01	2.11E+03	NA
96-15	< 8.79E+00	< 1.76E+01	5.32E+01	5.12E+03	NA
96-04 U	8.98E+00	2.28E+01	4.85E+01	8.43E+03	NA
96-04 L	1.98E+01	4.88E+01	8.71E+01	1.92E+04	NA
96-06 U	< 5.86E+00	< 1.17E+01	4.16E+01	7.57E+02	NA
96-06 M	1.52E+01	< 1.69E+01	5.84E+01	1.19E+03	NA
96-06 L	1.90E+01	< 1.82E+01	5.55E+01	9.51E+02	NA
96-11 U	1.20E+01	3.31E+01	6.20E+01	6.36E+03	NA
96-11 L	4.42E+01	1.93E+02	1.80E+02	6.17E+04	NA

µg/g: µg/g as-settled sludge
NA: not analyzed

	K.icp	Mg.icp	Mn.icp	Na.icp	Ni.icp
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	2.46E+03	< 3.51E+01	9.56E+01	1.71E+05	1.04E+02
96-05	< 9.02E+02	1.76E+03	< 1.81E+01	3.31E+02	< 3.61E+01
96-08	< 2.03E+02	4.12E+02	2.26E+01	1.79E+02	1.52E+01
96-09	< 1.26E+02	2.43E+02	8.56E+01	1.01E+02	6.11E+01
96-13	< 8.37E+02	1.43E+03	< 1.68E+01	3.24E+02	< 3.34E+01
96-15	< 8.79E+02	1.50E+03	< 1.76E+01	3.67E+02	< 3.52E+01
96-04 U	< 1.15E+02	1.49E+02	2.10E+01	1.06E+02	3.11E+01
96-04 L	< 2.68E+02	5.02E+02	3.74E+01	2.11E+02	2.92E+01
96-06 U	< 5.86E+02	7.46E+02	< 1.17E+01	2.58E+02	< 2.35E+01
96-06 M	< 8.39E+02	< 1.69E+02	< 1.69E+01	2.91E+02	< 3.36E+01
96-06 L	< 9.11E+02	< 1.82E+02	< 1.82E+01	3.19E+02	< 3.65E+01
96-11 U	< 5.30E+01	< 1.06E+01	4.25E+01	9.20E+01	1.07E+01
96-11 L	< 2.72E+02	7.14E+01	1.34E+02	1.39E+02	3.11E+01

	P.icp	Pb.icp	Se.icp	Sm.icp	Tl.icp
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	1.51E+03	4.62E+01	< 3.51E+01	< 3.51E+01	< 7.01E+01
96-05	< 3.61E+02	< 1.81E+02	< 1.81E+02	< 1.81E+02	< 3.61E+02
96-08	< 8.15E+01	4.10E+01	< 4.06E+01	< 4.06E+01	< 8.15E+01
96-09	1.16E+02	9.39E+01	< 2.53E+01	< 2.53E+01	< 5.05E+01
96-13	< 3.34E+02	< 1.68E+02	< 1.68E+02	< 1.68E+02	< 3.34E+02
96-15	< 3.52E+02	< 1.76E+02	< 1.76E+02	< 1.76E+02	< 3.52E+02
96-04 U	< 4.60E+01	3.83E+01	< 2.30E+01	< 2.30E+01	< 4.60E+01
96-04 L	< 1.07E+02	5.49E+01	< 5.35E+01	< 5.35E+01	< 1.07E+02
96-06 U	< 2.35E+02	< 1.17E+02	< 1.17E+02	< 1.17E+02	< 2.35E+02
96-06 M	< 3.36E+02	< 1.69E+02	< 1.69E+02	< 1.69E+02	< 3.36E+02
96-06 L	< 3.65E+02	< 1.82E+02	< 1.82E+02	< 1.82E+02	< 3.65E+02
96-11 U	5.92E+01	3.35E+01	< 1.06E+01	< 1.06E+01	< 2.12E+01
96-11 L	1.48E+02	1.24E+02	< 5.44E+01	< 5.44E+01	< 1.09E+02

µg/g: µg/g as-settled sludge

	U.icp	U.phos	U.las PNNL	U.icp/ms PNNL	U.las PNNL accelerated sample
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	< 1.75E+02	6.45E+02	5.62E+05	4.44E+05	5.83E+05
96-05	5.94E+05	4.65E+05	7.00E+05	5.10E+05	5.27E+05
96-08	9.47E+04	7.62E+04	9.93E+04	NA	NA
96-09	1.60E+04	1.61E+04	1.08E+05	NA	NA
96-13	5.62E+05	5.57E+05	6.17E+05	NA	NA
96-15	5.22E+05	3.84E+05	6.35E+05	NA	NA
96-04 U	3.34E+04	3.12E+04	NA	NA	NA
96-04 L	1.29E+05	1.11E+05	2.22E+05	NA	NA
96-06 U	3.30E+05	2.60E+05	NA	NA	NA
96-06 M	4.94E+05	4.03E+05	5.44E+05	NA	NA
96-06 L	5.87E+05	4.82E+05	6.69E+05	NA	NA
96-11 U	6.99E+03	6.73E+03	NA	NA	NA
96-11 L	1.88E+04	1.77E+04	2.25E+04	NA	NA

	Zn.icp	Zr.icp	Zr.icp/ms PNNL	Water.grav	
Sample	µg/g	µg/g	µg/g	µg/g	
96-01	1.65E+01	8.17E+00	2.20E+03	4.21E+05	
96-05	4.15E+01	< 1.81E+01	1.04E+02	2.35E+05	
96-08	1.15E+02	8.08E+01	NA	7.79E+05	
96-09	1.71E+02	6.84E+01	NA	8.70E+05	
96-13	1.68E+01	< 1.68E+01	NA	2.38E+05	
96-15	3.91E+01	5.24E+01	NA	2.72E+05	
96-04 U	7.24E+01	9.36E+01	NA	8.95E+05	
96-04 L	1.11E+02	1.40E+02	NA	6.38E+05	
96-06 U	1.19E+01	< 1.17E+01	NA	NA	
96-06 M	< 1.69E+01	< 1.69E+01	NA	3.52E+05	
96-06 L	< 1.82E+01	< 1.82E+01	NA	2.23E+05	
96-11 U	6.60E+01	4.13E+01	NA	9.48E+05	
96-11 L	3.43E+02	6.88E+01	NA	7.91E+05	

µg/g: µg/g as-settled sludge
INC: laboratory analyses not finished
NA: not analyzed

	^{233}U.tims \$ PNNL	^{234}U.tims \$ PNNL	^{235}U.tims \$ PNNL	^{236}U.tims \$ PNNL	^{238}U.tims \$ PNNL
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	< 2.75E+00	5.23E+01	5.30E+03	2.98E+02	5.57E+05
96-05	< 3.43E+00	3.51E+01	3.43E+03	2.71E+02	6.96E+05
96-08	< 4.86E-01	7.81E+00	6.45E+02	8.08E+01	9.86E+04
96-09	< 5.29E-01	7.76E+00	7.49E+02	8.04E+01	1.07E+05
96-13	< 3.02E+00	5.34E+01	4.46E+03	3.98E+02	6.12E+05
96-15	< 3.11E+00	4.37E+01	4.50E+03	2.86E+02	6.30E+05
96-04 U	NA	NA	NA	NA	NA
96-04 L	< 1.08E+00	1.64E+01	1.33E+03	2.14E+02	2.20E+05
96-06 U	NA	NA	NA	NA	NA
96-06 M	< 2.66E+00	5.70E+01	4.03E+03	4.35E+02	5.40E+05
96-06 L	< 3.28E+00	6.45E+01	4.94E+03	5.51E+02	6.64E+05
96-11 U	NA	NA	NA	NA	NA
96-11 L	< 1.10E-01	3.26E+00	1.61E+02	1.71E+01	2.23E+04
	^{233}U.icp/ms PNNL	^{234}U.icp/ms PNNL	^{235}U.icp/ms PNNL	^{236}U.icp/ms PNNL	^{238}U.icp/ms PNNL
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	2.97E-01	3.38E+01	4.31E+03	2.53E+02	4.38E+05
96-05	1.59E-01	2.79E+01	2.64E+03	2.30E+02	5.04E+05
	^{125}Sb.gea PNNL	m/z241 icp/ms PNNL			^{238}U.gea
	µCi/g	µg/g			µg/g
96-01	< 1.37E-02	< 1.37E-01			1.82E+05
96-05	8.78E-01	4.00E+01			< 1.18E+07

\$: based on the total uranium (U.las µg/g as-settled sludge) by PNNL
µg/g: µg/g as-settled sludge
µCi/g: µCi/g as-settled sludge
NA: not analyzed

	Br^-.ic	Cl^-.ic	F^-.ic	NO_2^-.ic	NO_3^-.ic
Sample	µg/ml	µg/ml	µg/ml	µg/ml	µg/ml
96-04	< 1.38E+00	6.38E-01	< 1.32E-01	< 1.19E+00	2.03E+00
96-06	< 1.38E+00	4.78E+00	5.84E-01	< 1.19E+00	< 1.53E+00
96-08	< 1.38E+00	7.05E-01	< 1.32E-01	< 1.19E+00	< 1.53E+00
96-09	< 1.38E+00	3.69E-01	< 1.32E-01	< 1.19E+00	< 1.53E+00
96-11	< 1.38E+00	5.35E-01	< 1.32E-01	< 1.19E+00	< 1.53E+00
96-13	< 1.38E+00	1.21E+00	5.89E-01	< 1.19E+00	< 1.53E+00
96-15	< 1.38E+00	6.94E-01	< 1.32E-01	< 1.19E+00	< 1.53E+00

	Oxalate.ic	PO_4^{3-}.ic	SO_4^{2-}.ic	pH	
Sample	µg/ml	µg/ml	µg/ml	pH units	
96-04	< 1.16E+00	< 1.32E+00	2.85E+00	8.20E+00	
96-06	< 1.16E+00	< 1.32E+00	7.67E+00	7.31E+00	
96-08	< 1.16E+00	< 1.32E+00	5.33E+00	7.29E+00	
96-09	< 1.16E+00	< 1.32E+00	3.08E+00	7.94E+00	
96-11	< 1.16E+00	< 1.32E+00	< 1.52E+00	8.38E+00	
96-13	< 1.16E+00	< 1.32E+00	4.43E+00	7.98E+00	
96-15	< 1.16E+00	< 1.32E+00	6.74E+00	7.07E+00	

µg/ml: $\mu g_{analyte}/ml_{liquid}$

	Br^-.ic	Cl^-.ic	F^-.ic	NO_2^-.ic	NO_3^-.ic
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-04 U	< 1.24E+00	5.74E-01	< 1.19E-01	< 1.07E+00	1.83E+00
96-04 L	< 8.85E-01	4.09E-01	< 8.46E-02	< 7.63E-01	1.30E+00
96-06 M	< 4.88E-01	1.69E+00	2.07E-01	< 4.21E-01	< 5.41E-01
96-06 L	< 3.09E-01	1.07E+00	1.31E-01	< 2.66E-01	< 3.43E-01
96-08	< 1.08E+00	5.53E-01	< 1.03E-01	< 9.33E-01	< 1.20E+00
96-09	< 1.21E+00	3.23E-01	< 1.15E-01	< 1.04E+00	< 1.34E+00
96-11 U	< 1.31E+00	5.09E-01	< 1.26E-01	< 1.13E+00	< 1.46E+00
96-11 L	< 1.10E+00	4.25E-01	< 1.05E-01	< 9.46E-01	< 1.22E+00
96-13	< 3.38E-01	2.97E-01	1.44E-01	< 2.92E-01	< 3.75E-01
96-15	< 3.78E-01	1.90E-01	< 3.61E-02	< 3.26E-01	< 4.19E-01

	Oxalate.ic	PO_4^{3-}.ic	SO_4^{2-}.ic		
Sample	µg/g	µg/g	µg/g		
96-04 U	< 1.04E+00	< 1.19E+00	2.56E+00		
96-04 L	< 7.44E-01	< 8.46E-01	1.82E+00		
96-06 M	< 4.10E-01	< 4.67E-01	2.71E+00		
96-06 L	< 2.60E-01	< 2.96E-01	1.72E+00		
96-08	< 9.09E-01	< 1.03E+00	4.18E+00		
96-09	< 1.01E+00	< 1.15E+00	2.69E+00		
96-11 U	< 1.11E+00	< 1.26E+00	< 1.45E+00		
96-11 L	< 9.22E-01	< 1.05E+00	< 1.21E+00		
96-13	< 2.84E-01	< 3.24E-01	1.09E+00		
96-15	< 3.17E-01	< 3.61E-01	1.84E+00		

µg/g: µg/g as-settled sludge

	DSC Exotherm Result 1	DSC Exotherm Result 2	DSC Exotherm Mean	DSC Endotherm Result 1	DSC Endotherm Result 2
Sample	Joules/g	Joules/g	Joules/g	Joules/g wet	Joules/g wet
96-01	0	0	0	> 184	> 247
96-05	0	0	0	> 220	> 199
96-08	0	0	0	> 933	> 380
96-09	0	0	0	> 111	> 558
96-13	-44.9 wet basis -58.4 dry basis	-16.7 wet basis -23.6 dry basis	-30.8 wet basis -41.0 dry basis	> 108	> 62
96-15	0	0	0	> 123	> 49

	Initial Loss	TGA Loss	Total Mass Loss	Mean Mass Loss	
Sample	wt%	wt%	wt%	wt%	
96-01	---	23.1, 20.3	23.1, 20.3	21.7	
96-05	---	19.8, 15.5	19.8, 15.5	17.7	
96-08	33.1, 61.0	57.6, 35.7	71.6, 74.9	73.3	
96-09	85.5, 56.7	7.70, 44.6	86.7, 76.0	81.3	
96-13	9.48, 26.8	15.0, 3.40	23.1, 29.3	26.2	
96-15	14.7, 14.0	10.0, 15.0	23.3, 26.9	25.1	

Joules/g wet: Joules/g as-settled sludge (wet basis)
Joules/g dry : Joules/g as-settled sludge (dry basis)

The conversion of μCi/g as-settled sludge or μg/g as-settled sludge utilized the as-settled density (g as-settled sludge/ml as-settled sludge) measured for each of the samples. The as-settled density measurements are listed in Table D-3. These values are provided in Silvers 1997a.

The PNNL uranium isotopic results were reported in atom % (Appendix C). The units for the uranium isotopics were changed to μg/ml as-settled sludge using Equation 3 of Appendix C, where C_u is the total uranium concentration (C), as measured by PNNL (units of μg/ml as-settled sludge), for the sample of interest.

Table D-3. As-Settled Sludge Density.

Sample	Density g/ml As-settled sludge
96-01	2.09
96-05	2.34
96-08	1.19 &
96-09 $	1.07 &
96-13	2.458
96-15	1.845
96-04 Upper/Lower *	1.09
96-04 Lower #	1.46
96-06 Upper	1.42
96-06 Middle #	1.92
96-06 Lower #	2.99
96-11 Upper	1.06
96-11 Lower #	1.24

$ = Calculated from data gathered while obtaining the sample for rheology measurements.

& = The data were rounded after the duplicate density measurements were averaged versus rounding each density measurement, averaging, and then rounding again.

* = No data available for 96-04 Upper; the interface data is used to approximate the as-settled density of the upper layer since the visual appearances of the two layers were similar.

= The following equation was used to determine the as-settled density. The equation used 1g/ml as the supernate density.

$$Density_{as\text{-}settled} = \frac{g_{set\ sol}}{\dfrac{g_{cf\ sol}}{Density_{cf}} + \dfrac{(g_{set\ sol} - g_{cf\ sol})}{Density_{supernate}}}$$

	^{155}Eu.gea	^{155}Eu.gea PNNL	^{94}Nb.gea	^{237}Np	^{238}Pu
Sample	µCi/ml	µCi/ml	µCi/ml	µCi/ml	µCi/ml
96-01	< 9.16E-01	2.59E-02	< 6.96E-02	< 1.06E-02	7.71E-03
96-05	2.26E+01	8.96E+00	< 2.18E+00	1.48E-02	3.01E+01
96-08	1.54E+00	NA	< 6.69E-02	< 3.48E-03	5.08E+00
96-09	2.11E-01	NA	< 1.65E-02	< 2.97E-03	7.75E-01
96-13	< 1.22E+01	NA	< 1.79E+00	3.36E-02	< 1.30E+02
96-15	< 1.41E+01	NA	< 2.33E+00	1.81E-02	1.18E+01
96-04 U	< 1.62E+00	NA	< 1.59E-01	< 4.87E-03	4.66E+00
96-04 L	5.35E+00	NA	< 5.59E-01	7.00E-03	1.28E+01
96-06 U	< 6.55E+00	NA	< 1.11E+00	1.13E-02	1.02E+01
96-06 M	< 8.68E+00	NA	< 1.35E+00	2.41E-02	2.32E+02
96-06 L	< 2.03E+01	NA	< 4.28E+00	2.87E-02	3.81E+02
96-11 U	1.52E-01	NA	< 1.23E-02	2.76E-03	4.87E-01
96-11 L	< 4.45E-01	NA	< 8.40E-02	< 6.09E-03	1.05E+00

	$^{239/240}$Pu	^{239}Pu.icp/ms PNNL	^{240}Pu.icp/ms PNNL	^{226}Ra.gea	^{106}Ru/Rh.gea
Sample	µCi/ml	µCi/ml	µCi/ml	µCi/ml	µCi/ml
96-01	7.14E-02	2.23E+01	< 6.47E-02	< 6.83E+00	< 5.00E+00
96-05	2.84E+02	1.80E+02	7.23E+01	< 8.94E+01	< 6.75E+01
96-08	2.73E+01	NA	NA	< 2.76E+00	< 2.10E+00
96-09	4.47E+00	NA	NA	< 5.94E-01	< 4.51E-01
96-13	2.04E+02	NA	NA	< 7.63E+01	< 5.89E+01
96-15	1.37E+02	NA	NA	< 8.72E+01	< 6.44E+01
96-04 U	2.17E+01	NA	NA	< 7.80E+00	< 6.16E+00
96-04 L	5.91E+01	NA	NA	< 2.13E+01	< 1.66E+01
96-06 U	7.41E+01	NA	NA	< 4.07E+01	< 2.88E+01
96-06 M	1.31E+03	NA	NA	< 3.67E+01	< 2.71E+01
96-06 L	2.21E+03	NA	NA	< 1.39E+02	< 1.06E+02
96-11 U	2.55E+00	NA	NA	< 1.85E-01	< 1.46E-01
96-11 L	5.14E+00	NA	NA	< 3.21E+00	< 2.36E+00

µCi/ml: µCi/ml as-settled sludge
NA: not analyzed

	$^{89/90}$Sr	^{99}Tc	^{208}Tl.gea	Total Carbon	TOC
Sample	µCi/ml	µCi/ml	µCi/ml	µg/ml	µg/ml
96-01	9.01E+01	4.19E-01	< 4.53E+00	4.22E+04	1.95E+04
96-05	4.07E+03	1.20E+01	< 5.69E+01	2.01E+03	1.24E+03
96-08	2.75E+02	9.99E+00	< 1.81E+00	1.40E+03	3.56E+02
96-09	3.70E+00	3.58E+00	< 3.91E-01	1.37E+03	1.11E+03
96-13	3.29E+03	1.89E+01	< 4.94E+01	1.82E+03	1.01E+03
96-15	2.28E+03	1.77E+00	< 5.48E+01	1.44E+03	9.39E+02
96-04 U	6.92E+01	NA	< 5.10E+00	9.11E+02	4.26E+02
96-04 L	5.92E+02	NA	< 1.40E+01	2.61E+03	5.82E+02
96-06 U	1.15E+03	NA	< 2.47E+01	6.18E+02	3.78E+02
96-06 M	2.26E+03	NA	< 2.30E+01	9.96E+02	8.28E+02
96-06 L	4.71E+03	NA	< 9.04E+01	1.51E+03	1.03E+03
96-11 U	2.05E+00	NA	< 1.23E-01	5.47E+02	3.09E+02
96-11 L	1.86E+01	NA	< 2.07E+00	1.98E+03	1.27E+03

	TIC	Ag.icp	Al.icp	Al.icp/ms PNNL	B.icp
Sample	µg/ml	µg/ml	µg/ml	µg/ml	µg/ml
96-01	2.04E+04	2.60E+01	4.26E+04	1.62E+03	2.95E+02
96-05	5.36E+02	2.36E+02	2.45E+04	1.91E+04	2.87E+02
96-08	6.95E+02	1.48E+01	2.30E+04	NA	1.10E+02
96-09	5.55E+02	< 2.71E+00	1.83E+04	NA	6.25E+01
96-13	7.57E+02	2.88E+02	2.69E+04	NA	3.07E+02
96-15	6.93E+02	2.81E+02	2.31E+04	NA	2.60E+02
96-04 U	4.75E+02	6.19E+00	1.47E+04	NA	6.14E+01
96-04 L	2.12E+03	3.06E+01	5.26E+04	NA	1.11E+02
96-06 U	2.20E+02	7.13E+01	7.40E+03	NA	1.01E+02
96-06 M	7.36E+02	1.98E+02	1.84E+04	NA	2.03E+02
96-06 L	7.19E+02	3.79E+02	2.47E+04	NA	3.64E+02
96-11 U	4.07E+02	1.31E+00	7.76E+03	NA	5.68E+01
96-11 L	7.65E+02	< 6.75E+00	1.96E+04	NA	7.15E+01

µCi/ml: µCi/ml as-settled sludge
µg/ml: µg/ml as-settled sludge
NA: not analyzed

	Ba.icp	Be.icp	Bi.icp	Ca.icp	Ca.icp/ms PNNL
Sample	µg/ml	µg/ml	µg/ml	µg/ml	µg/ml
96-01	< 3.66E+01	< 3.66E+00	< 7.31E+01	5.87E+02	1.01E+02
96-05	2.46E+02	< 2.11E+01	1.22E+03	1.45E+03	2.02E+02
96-08	4.60E+01	1.65E+01	1.00E+02	4.11E+02	NA
96-09	2.00E+01	1.33E+01	2.71E+01	2.12E+02	NA
96-13	2.40E+02	< 2.06E+01	1.23E+03	1.29E+03	NA
96-15	2.54E+02	< 1.62E+01	6.19E+02	< 3.25E+02	NA
96-04 U	3.82E+01	6.87E+00	4.19E+01	2.43E+02	NA
96-04 L	9.68E+01	1.05E+01	1.88E+02	7.43E+02	NA
96-06 U	9.15E+01	< 8.29E+00	4.38E+02	3.77E+02	NA
96-06 M	< 1.61E+02	< 1.61E+01	3.75E+02	< 3.24E+02	NA
96-06 L	2.97E+02	< 2.72E+01	7.00E+02	< 5.45E+02	NA
96-11 U	< 5.64E+00	1.05E+01	< 1.13E+01	6.34E+01	NA
96-11 L	5.08E+01	2.24E+01	< 6.75E+01	7.38E+02	NA

	Cd.icp	Cr.icp	Cu.icp	Fe.icp	Fe.icp/ms PNNL
Sample	µg/ml	µg/ml	µg/ml	µg/ml	µg/ml
96-01	1.11E+01	3.27E+03	1.36E+01	6.54E+02	2.39E+03
96-05	4.36E+01	< 4.22E+01	1.74E+02	1.30E+04	1.16E+04
96-08	1.20E+01	8.89E+01	7.47E+01	2.12E+04	NA
96-09	1.92E+01	8.17E+01	1.69E+02	2.77E+04	NA
96-13	3.73E+01	< 4.12E+01	1.49E+02	5.19E+03	NA
96-15	< 1.62E+01	< 3.24E+01	9.81E+01	9.45E+03	NA
96-04 U	9.78E+00	2.49E+01	5.28E+01	9.18E+03	NA
96-04 L	2.90E+01	7.12E+01	1.27E+02	2.80E+04	NA
96-06 U	< 8.29E+00	< 1.66E+01	5.89E+01	1.07E+03	NA
96-06 M	2.91E+01	< 3.24E+01	1.12E+02	2.28E+03	NA
96-06 L	5.69E+01	< 5.45E+01	1.66E+02	2.84E+03	NA
96-11 U	1.28E+01	3.53E+01	6.60E+01	6.77E+03	NA
96-11 L	5.48E+01	2.40E+02	2.23E+02	7.66E+04	NA

µg/ml: µg/ml as-settled sludge
NA: not analyzed

	K.icp	Mg.icp	Mn.icp	Na.icp	Ni.icp
Sample	µg/ml	µg/ml	µg/ml	µg/ml	µg/ml
96-01	5.12E+03	< 7.31E+01	1.99E+02	3.56E+05	2.16E+02
96-05	< 2.11E+03	4.10E+03	< 4.22E+01	7.73E+02	< 8.44E+01
96-08	< 2.42E+02	4.89E+02	2.68E+01	2.13E+02	1.80E+01
96-09	< 1.35E+02	2.61E+02	9.19E+01	1.09E+02	6.56E+01
96-13	< 2.06E+03	3.52E+03	< 4.12E+01	7.95E+02	< 8.22E+01
96-15	< 1.62E+03	2.78E+03	< 3.24E+01	6.78E+02	< 6.49E+01
96-04 U	< 1.26E+02	1.62E+02	2.29E+01	1.16E+02	3.39E+01
96-04 L	< 3.91E+02	7.33E+02	5.45E+01	3.08E+02	4.27E+01
96-06 U	< 8.29E+02	1.06E+03	< 1.66E+01	3.65E+02	< 3.32E+01
96-06 M	< 1.61E+03	< 3.24E+02	< 3.24E+01	5.58E+02	< 6.46E+01
96-06 L	< 2.72E+03	< 5.45E+02	< 5.45E+01	9.53E+02	< 1.09E+02
96-11 U	< 5.64E+01	< 1.13E+01	4.52E+01	9.80E+01	1.14E+01
96-11 L	< 3.38E+02	8.85E+01	1.66E+02	1.73E+02	3.86E+01

	P.icp	Pb.icp	Se.icp	Sm.icp	Tl.icp
Sample	µg/ml	µg/ml	µg/ml	µg/ml	µg/ml
96-01	3.15E+03	9.64E+01	< 7.31E+01	< 7.31E+01	< 1.46E+02
96-05	< 8.44E+02	< 4.22E+02	< 4.22E+02	< 4.22E+02	< 8.44E+02
96-08	< 9.69E+01	4.87E+01	< 4.83E+01	< 4.83E+01	< 9.69E+01
96-09	1.24E+02	1.01E+02	< 2.71E+01	< 2.71E+01	< 5.43E+01
96-13	< 8.22E+02	< 4.12E+02	< 4.12E+02	< 4.12E+02	< 8.22E+02
96-15	< 6.49E+02	< 3.24E+02	< 3.24E+02	< 3.24E+02	< 6.49E+02
96-04 U	< 5.01E+01	4.17E+01	< 2.50E+01	< 2.50E+01	< 5.01E+01
96-04 L	< 1.56E+02	8.02E+01	< 7.81E+01	< 7.81E+01	< 1.56E+02
96-06 U	< 3.32E+02	< 1.66E+02	< 1.66E+02	< 1.66E+02	< 3.32E+02
96-06 M	< 6.46E+02	< 3.24E+02	< 3.24E+02	< 3.24E+02	< 6.46E+02
96-06 L	< 1.09E+03	< 5.45E+02	< 5.45E+02	< 5.45E+02	< 1.09E+03
96-11 U	6.30E+01	3.57E+01	< 1.13E+01	< 1.13E+01	< 2.26E+01
96-11 L	1.84E+02	1.54E+02	< 6.75E+01	< 6.75E+01	< 1.35E+02

µg/ml: µg/ml as-settled sludge

	U.icp	U.phos	U.las PNNL	U.icp/ms PNNL	U.las PNNL accelerated sample
Sample	µg/ml	µg/ml	µg/ml	µg/ml	µg/ml
96-01	< 3.66E+02	1.34E+03	1.17E+06	9.26E+05	1.22E+06
96-05	1.39E+06	1.09E+06	1.64E+06	1.19E+06	1.23E+06
96-08	1.13E+05	9.05E+04	1.18E+05	NA	NA
96-09	1.72E+04	1.73E+04	1.16E+05	NA	NA
96-13	1.38E+06	1.37E+06	1.52E+06	NA	NA
96-15	9.63E+05	7.08E+05	1.17E+06	NA	NA
96-04 U	3.64E+04	3.40E+04	NA	NA	NA
96-04 L	1.89E+05	1.62E+05	3.23E+05	NA	NA
96-06 U	4.67E+05	3.68E+05	NA	NA	NA
96-06 M	9.48E+05	7.73E+05	1.05E+06	NA	NA
96-06 L	1.76E+06	1.44E+06	2.00E+06	NA	NA
96-11 U	7.45E+03	7.16E+03	NA	NA	NA
96-11 L	2.33E+04	2.19E+04	2.79E+04	NA	NA

	Zn.icp	Zr.icp	Zr.icp/ms PNNL	Water.grav	Density as-settled sludge
Sample	µg/ml	µg/ml	µg/ml	µg/ml	g/ml
96-01	3.43E+01	1.70E+01	4.59E+03	8.78E+05	2.09
96-05	9.69E+01	< 4.22E+01	2.44E+02	5.49E+05	2.34
96-08	1.37E+02	9.60E+01	NA	9.26E+05	1.19
96-09	1.83E+02	7.34E+01	NA	9.34E+05	1.07
96-13	4.12E+01	< 4.12E+01	NA	5.85E+05	2.458
96-15	7.21E+01	9.67E+01	NA	5.02E+05	1.845
96-04 U	7.88E+01	1.02E+02	NA	9.74E+05	1.09
96-04 L	1.61E+02	2.04E+02	NA	9.31E+05	1.46
96-06 U	1.69E+01	< 1.66E+01	NA	NA	1.42
96-06 M	< 3.24E+01	< 3.24E+01	NA	6.75E+05	1.92
96-06 L	< 5.45E+01	< 5.45E+01	NA	6.66E+05	2.99
96-11 U	7.03E+01	4.40E+01	NA	1.01E+06	1.06
96-11 L	4.25E+02	8.53E+01	NA	9.81E+05	1.24

µg/ml: µg/ml as-settled sludge
NA: not analyzed

	^{233}U.tims $^{\$}$ PNNL	^{234}U.tims $^{\$}$ PNNL	^{235}U.tims $^{\$}$ PNNL	^{236}U.tims $^{\$}$ PNNL	^{238}U.tims $^{\$}$ PNNL
Sample	µg/ml	µg/ml	µg/ml	µg/ml	µg/ml
96-01	< 5.74E+00	1.09E+02	1.11E+04	6.22E+02	1.16E+06
96-05	< 8.01E+00	8.21E+01	8.03E+03	6.33E+02	1.63E+06
96-08	< 5.78E-01	9.28E+00	7.66E+02	9.60E+01	1.17E+05
96-09	< 5.68E-01	8.33E+00	8.05E+02	8.63E+01	1.15E+05
96-13	< 7.43E+00	1.31E+02	1.10E+04	9.78E+02	1.50E+06
96-15	< 5.73E+00	8.06E+01	8.29E+03	5.28E+02	1.16E+06
96-04 U	NA	NA	NA	NA	NA
96-04 L	< 1.58E+00	2.40E+01	1.94E+03	3.13E+02	3.21E+05
96-06 U	NA	NA	NA	NA	NA
96-06 M	< 5.12E+00	1.09E+02	7.73E+03	8.34E+02	1.04E+06
96-06 L	< 9.80E+00	1.93E+02	1.48E+04	1.65E+03	1.98E+06
96-11 U	NA	NA	NA	NA	NA
96-11 L	< 1.36E-01	4.04E+00	2.00E+02	2.11E+01	2.76E+04

	^{233}U.icp/ms PNNL	^{234}U.icp/ms PNNL	^{235}U.icp/ms PNNL	^{236}U.icp/ms PNNL	^{238}U.icp/ms PNNL
Sample	µg/ml	µg/ml	µg/ml	µg/ml	µg/ml
96-01	6.20E-01	7.05E+01	9.00E+03	5.27E+02	9.14E+05
96-05	3.71E-01	6.53E+01	6.18E+03	5.37E+02	1.18E+06

	^{125}Sb.gea PNNL	m/z241 icp/ms PNNL			^{238}U.gea
	µCi/ml	µg/ml			µg/ml
96-01	< 2.85E-02	< 2.85E-01			3.80E+05
96-05	2.05E+00	9.34E+01			< 2.76E+07

$: based on the total uranium (U.las µg/ml as-settled sludge) by PNNL
µg/ml: µg/ml as-settled sludge
µCi/ml: µCi/ml as-settled sludge
NA: not analyzed

	Br^-.ic	Cl^-.ic	F^-.ic	NO_2^-.ic	NO_3^-.ic	Oxalate.ic	PO_4^{3-}.ic	SO_4^{2-}.ic
Sample	µg/ml	µg/ml	µg/ml	µg/ml	µg/ml	µg/ml	µg/ml	µg/ml
96-04 U	< 1.35E+00	6.25E-01	< 1.29E-01	< 1.17E+00	1.99E+00	< 1.14E+00	< 1.29E+00	2.79E+00
96-04 L	< 1.29E+00	5.97E-01	< 1.24E-01	< 1.11E+00	1.90E+00	< 1.09E+00	< 1.24E+00	2.66E+00
96-06 M	< 9.37E-01	3.25E+00	3.97E-01	< 8.08E-01	< 1.04E+00	< 7.88E-01	< 8.96E-01	5.20E+00
96-06 L	< 9.24E-01	3.20E+00	3.91E-01	< 7.97E-01	< 1.02E+00	< 7.77E-01	< 8.84E-01	5.13E+00
96-08	< 1.28E+00	6.56E-01	< 1.23E-01	< 1.11E+00	< 1.42E+00	< 1.08E+00	< 1.23E+00	4.96E+00
96-09	< 1.30E+00	3.47E-01	< 1.24E-01	< 1.12E+00	< 1.44E+00	< 1.09E+00	< 1.24E+00	2.89E+00
96-11 U	< 1.40E+00	5.42E-01	< 1.34E-01	< 1.21E+00	< 1.55E+00	< 1.18E+00	< 1.34E+00	< 1.54E+00
96-11 L	< 1.36E+00	5.27E-01	< 1.30E-01	< 1.17E+00	< 1.51E+00	< 1.14E+00	< 1.30E+00	< 1.50E+00
96-13	< 8.32E-01	7.29E-01	3.55E-01	< 7.17E-01	< 9.22E-01	< 6.99E-01	< 7.96E-01	2.67E+00
96-15	< 6.97E-01	3.50E-01	< 6.66E-02	< 6.01E-01	< 7.72E-01	< 5.86E-01	< 6.66E-01	3.40E+00

µg/ml: µg/ml as-settled sludge

Six water samples were collected and analyzed by both laboratories. The six water samples were:

- Water collected from the 96-01 cylinder (Cyl)
- Water collected from the 96-05 carboy (Car)
- Water collected from the 96-05 cylinder
- An equipment blank (Eq Blk)
  - Water from 305 Building Pool
- An equipment blank
  - Water from 305 Building Pool--post pool sample
- A hot cell blank.

The reported units for these samples are either µg/ml liquid or µCi/ml liquid. The water results for all but the hot cell blank are also included here.

Water Sample Analytical Results				
	Alpha Total	^{241}Am.aea	^{241}Am.gea	Beta Total
Sample	µCi/ml	µCi/ml	µCi/ml	µCi/ml
305 Eq Blk	NA	NA	NA	NA
305 Post Pool	NA	NA	NA	NA
96-01 Cyl	< 2.03E-04	< 2.87E-04	< 1.06E-02	1.32E-01
96-05 Car	< 1.90E-04	< 2.78E-04	< 3.71E-02	3.41E+00
96-05 Cyl	1.56E-01	1.51E-01	< 4.39E-01	1.23E+02
	^{212}Bi.gea	^{144}Ce/Pr.gea	$^{243/244}$Cm.aea	^{57}Co.gea
Sample	µCi/ml	µCi/ml	µCi/ml	µCi/ml
305 Eq Blk	NA	NA	NA	NA
305 Post Pool	NA	NA	NA	NA
96-01 Cyl	< 1.52E-02	< 1.45E-02	< 2.87E-04	< 9.59E-04
96-05 Car	< 1.76E-02	< 5.07E-02	< 2.78E-04	< 3.30E-03
96-05 Cyl	< 2.30E-01	< 5.89E-01	< 1.03E-02	< 3.90E-02
	^{60}Co.gea	^{134}Cs.gea	^{137}Cs.gea	^{152}Eu.gea
Sample	µCi/ml	µCi/ml	µCi/ml	µCi/ml
305 Eq Blk	NA	NA	NA	NA
305 Post Pool	NA	NA	NA	NA
96-01 Cyl	< 1.58E-03	< 1.34E-03	1.05E-01	< 6.38E-03
96-05 Car	< 1.69E-03	< 4.21E-03	3.31E+00	< 4.55E-03
96-05 Cyl	< 1.57E-02	< 4.25E-02	3.63E+01	< 5.88E-02
	^{154}Eu.gea	^{155}Eu.gea	^{94}Nb.gea	^{237}Np
Sample	µCi/ml	µCi/ml	µCi/ml	µCi/ml
305 Eq Blk	NA	NA	NA	NA
305 Post Pool	NA	NA	NA	NA
96-01 Cyl	< 3.16E-03	< 4.15E-03	< 1.08E-03	< 9.21E-04
96-05 Car	< 2.88E-03	< 1.40E-02	< 1.18E-03	< 9.21E-04
96-05 Cyl	< 4.10E-02	< 1.67E-01	< 1.56E-02	< 9.21E-04

NA: not analyzed

Water Sample Analytical Results

	^{238}Pu	$^{239/240}$Pu	^{226}Ra.gea	^{106}Ru/Rh.gea
Sample	µCi/ml	µCi/ml	µCi/ml	µCi/ml
305 Eq Blk	NA	NA	NA	NA
305 Post Pool	NA	NA	NA	NA
96-01 Cyl	< 1.78E-04	< 1.78E-04	< 2.81E-02	< 2.52E-02
96-05 Car	< 1.99E-04	< 1.99E-04	< 1.06E-01	< 7.88E-02
96-05 Cyl	< 2.09E-03	< 2.09E-03	< 1.21E+00	< 8.69E-01

	$^{89/90}$Sr	^{208}Tl.gea	Total Carbon	TOC
Sample	µCi/ml	µCi/ml	µg/ml	µg/ml
305 Eq Blk	NA	NA	< 4.00E+01	< 4.00E+01
305 Post Pool	NA	NA	< 4.00E+01	< 4.00E+01
96-01 Cyl	1.61E-02	< 2.02E-02	NA	NA
96-05 Car	8.73E-02	< 6.83E-02	NA	NA
96-05 Cyl	4.97E+01	< 7.71E-01	NA	NA

	TIC	Ag.icp	Al.icp	B.icp
Sample	µg/ml	µg/ml	µg/ml	µg/ml
305 Eq Blk	< 5.00E+00	< 1.00E-02	1.11E-01	6.14E-01
305 Post Pool	< 5.00E+00	< 1.00E-02	1.07E-01	6.37E-01
96-01 Cyl	NA	< 1.00E-02	4.26E-01	6.17E-01
96-05 Car	NA	< 1.00E-02	1.19E-01	7.09E-01
96-05 Cyl	NA	< 1.00E-02	1.58E-01	7.18E-01

	Ba.icp	Be.icp	Bi.icp	Ca.icp
Sample	µg/ml	µg/ml	µg/ml	µg/ml
305 Eq Blk	< 5.00E-02	< 5.00E-03	< 1.00E-01	3.60E-01
305 Post Pool	< 5.00E-02	< 5.00E-03	< 1.00E-01	2.23E-01
96-01 Cyl	< 5.00E-02	< 5.00E-03	< 1.00E-01	2.03E-01
96-05 Car	< 5.00E-02	< 5.00E-03	< 1.00E-01	2.35E-01
96-05 Cyl	< 5.00E-02	< 5.00E-03	< 1.00E-01	6.88E-01

NA: not analyzed

Water Sample Analytical Results

	Cd.icp	Cr.icp	Cu.icp	Fe.icp
Sample	µg/ml	µg/ml	µg/ml	µg/ml
305 Eq Blk	< 5.00E-03	< 1.00E-02	1.93E-02	5.47E-02
305 Post Pool	< 5.00E-03	< 1.00E-02	< 1.00E-02	< 5.00E-02
96-01 Cyl	< 5.00E-03	< 1.00E-02	< 1.00E-02	< 5.00E-02
96-05 Car	< 5.00E-03	< 1.00E-02	< 1.00E-02	< 5.00E-02
96-05 Cyl	< 5.00E-03	< 1.00E-02	1.44E-02	< 5.00E-02

	K.icp	Mg.icp	Mn.icp	Na.icp
Sample	µg/ml	µg/ml	µg/ml	µg/ml
305 Eq Blk	< 5.00E-01	< 1.00E-01	< 1.00E-02	8.31E-01
305 Post Pool	< 5.00E-01	< 1.00E-01	< 1.00E-02	8.45E-01
96-01 Cyl	< 5.00E-01	< 1.00E-01	< 1.00E-02	7.97E-01
96-05 Car	< 5.00E-01	< 1.00E-01	< 1.00E-02	9.34E-01
96-05 Cyl	< 5.00E-01	< 1.00E-01	1.67E-02	1.41E+00

	Ni.icp	P.icp	Pb.icp	Se.icp
Sample	µg/ml	µg/ml	µg/ml	µg/ml
305 Eq Blk	< 2.00E-02	< 2.00E-01	< 1.00E-01	< 1.00E-01
305 Post Pool	< 2.00E-02	< 2.00E-01	< 1.00E-01	< 1.00E-01
96-01 Cyl	2.78E-02	< 2.00E-01	< 1.00E-01	< 1.00E-01
96-05 Car	< 2.00E-02	< 2.00E-01	< 1.00E-01	< 1.00E-01
96-05 Cyl	2.89E-02	< 2.00E-01	< 1.00E-01	< 1.00E-01

	Sm.icp	Tl.icp	U.icp	U.phos
Sample	µg/ml	µg/ml	µg/ml	µg/ml
305 Eq Blk	< 1.00E-01	< 2.00E-01	< 5.00E-01	NA
305 Post Pool	< 1.00E-01	< 2.00E-01	< 5.00E-01	NA
96-01 Cyl	< 1.00E-01	< 2.00E-01	< 5.00E-01	1.03E+02
96-05 Car	< 1.00E-01	< 2.00E-01	< 5.00E-01	1.85E+02
96-05 Cyl	< 1.00E-01	< 2.00E-01	1.46E+00	3.16E+02

NA: not analyzed

Water Sample Analytical Results

	U.las	Zn.icp	Zr.icp	^{233}U.tims
Sample	µg/ml	µg/ml	µg/ml	atom%
305 Eq Blk	NA	2.62E-02	< 1.00E-02	NA
305 Post Pool	NA	< 1.00E-02	< 1.00E-02	NA
96-01 Cyl	1.73E+01	1.22E-02	< 1.00E-02	< 5.00E-04
96-05 Car	2.12E+00	< 1.00E-02	< 1.00E-02	< 5.00E-04
96-05 Cyl	9.17E+01	1.39E-01	< 1.00E-02	< 5.00E-04

	^{234}U.tims	^{235}U.tims	^{236}U.tims	^{238}U.tims
Sample	atom%	atom%	atom%	atom%
305 Eq Blk	NA	NA	NA	NA
305 Post Pool	NA	NA	NA	NA
96-01 Cyl	8.30E-03	9.44E-01	5.50E-02	9.8992E+01
96-05 Car	5.70E-03	5.02E-01	4.15E-02	9.9451E+01
96-05 Cyl	7.40E-03	5.11E-01	4.80E-02	9.9434E+01

	Br^-.ic	Cl^-.ic	F^-.ic	NO_2^-.ic
Sample	µg/ml	µg/ml	µg/ml	µg/ml
305 Eq Blk	< 1.25E-01	1.35E-01	< 1.20E-02	< 1.08E-01
305 Post Pool	< 1.25E-01	7.60E-02	< 1.20E-02	< 1.08E-01
96-01 Cyl	< 1.26E+01	2.67E+00	4.43E+01	< 1.09E+01
96-05 Car	NA	NA	NA	NA
96-05 Cyl	< 1.26E+01	4.70E+00	< 1.21E+00	< 1.09E+01

	NO_3^-.ic	Oxalate.ic	PO_4^{3-}.ic	SO_4^{2-}.ic
Sample	µg/ml	µg/ml	µg/ml	µg/ml
305 Eq Blk	< 1.39E-01	< 1.05E-01	< 1.20E-01	3.79E-01
305 Post Pool	< 1.39E-01	< 1.05E-01	< 1.20E-01	3.13E-01
96-01 Cyl	1.74E+01	< 1.06E+01	< 1.21E+01	< 1.39E+01
96-05 Car	NA	NA	NA	NA
96-05 Cyl	1.58E+02	< 1.06E+01	< 1.21E+01	2.26E+01

NA: not analyzed

Water Sample Analytical Results	
	pH
Sample	pH units
305 Eq Blk	6.38
305 Post Pool	6.68
96-01 Cyl	6.06
96-05 Car	NA
96-05 Cyl	6.78
96-05 Cyl	4.41

NA: not analyzed

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APPENDIX E

CHEMISTRY OF DRIED K EAST CANISTER SLUDGE

T. L. Welsh

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APPENDIX E

CHEMISTRY OF DRIED K EAST CANISTER SLUDGE

The conversion of as-settled sludge results (which were converted from the 222-S Laboratory centrifuged sludge results) to as-dried sludge results utilized the following formulas. It is assumed that the analytes are non-volatile. The following table defines the symbols used in deriving the formula to convert the as settled-state analyses to a dried-state.

Symbol	Meaning	Units
c	gravimetric concentration	µg/g or µCi/g
g	mass	g
d	dried concentration	µg/g or µCi/g
subscript $_s$	as-settled state	
subscript $_d$	dried state	

By mass balance:

$$c_s g_s = c_d g_d \quad (1)$$

Solving for c_d gives the following conversion equation.

$$c_d = c_s \frac{g_s}{g_d} \quad (2)$$

The values for c_s are listed in Appendix D and the sample information are listed in Appendix C. The values for g_d/g_s are listed in Table E-1. The values for g_d and g_s are provided in Silvers 1997a.

For volatile analytes, the concentration on a dried sludge basis is zero.

The PNNL analyses were performed on dried samples (except for TGA and DSC) and reported with units of either µg/g dried sludge or µCi/g dried sludge. Two samples (called accelerated samples) were analyzed on a priority basis for total uranium (laser), uranium isotopics (tims), radionuclides (gea and icp/ms), total alpha, total beta, and some metals (icp/ms). All samples were analyzed for total uranium (laser) and uranium isotopics (tims).

The PNNL uranium isotopic results (tims) were reported in atom % (Appendix C). The units for the uranium isotopics were changed to µg/g dried sludge using Equation 3 of Appendix C, where C_u is the total uranium concentration (C), as measured by PNNL (units of µg/g dried sludge), for the sample of interest.

Table E-1. Conversion Factors Dried Sludge to As-Settled Sludge.

Sample	$g_{dry}/g_{as\text{-}settled}$
96-01	0.6829
96-05	0.7944
96-08	0.2459
96-09	0.1227
96-13	0.7527
96-15	0.7809
96-04 Upper	0.1052
96-04 Lower	0.4057
96-06 Upper	NA
96-06 Middle	0.6589
96-06 Lower	0.7945
96-11 Upper	0.0525
96-11 Lower	0.2410

NA: Not available; insufficient sample mass

	Alpha Total	Alpha Total icp/ms PNNL	^{241}Am.aea	^{241}Am.gea	^{241}Am.gea (PNNL)
Sample	µCi/g	µCi/g	µCi/g	µCi/g	µCi/g
96-01	1.49E-01	1.55E+02	1.23E-01	< 1.64E+00	3.15E-01
96-05	2.24E+02	2.69E+02	9.25E+01	1.33E+02	6.64E+01
96-08	1.61E+02	NA	7.73E+01	9.08E+02	NA
96-09	5.57E+01	NA	2.58E+01	3.26E+01	NA
96-13	1.61E+02	NA	< 2.17E+01	7.20E+01	NA
96-15	1.39E+02	NA	5.95E+01	7.04E+01	NA
96-04 U	3.44E+02	NA	2.20E+02	1.87E+02	NA
96-04 L	1.96E+02	NA	1.06E+02	1.21E+02	NA
96-06 U	NA	NA	NA	NA	NA
96-06 M	1.58E+02	NA	8.08E+01	9.95E+01	NA
96-06 L	1.74E+02	NA	7.58E+01	9.59E+01	NA
96-11 U	7.79E+01	NA	6.58E+01	4.76E+01	NA
96-11 L	2.88E+01	NA	1.47E+01	1.79E+01	NA

	Beta Total	Beta Total PNNL	^{212}Bi.gea	^{144}Ce/Pr.gea	$^{243/244}$Cm.gea
Sample	µCi/g	µCi/g	µCi/g	µCi/g	µCi/g
96-01	4.10E+02	1.27E+04	< 7.54E-01	< 2.32E+00	< 1.48E-02
96-05	4.66E+03	4.91E+03	< 1.18E+01	< 2.47E+01	< 7.92E+00
96-08	2.66E+03	NA	< 2.61E+00	< 4.68E+00	< 7.35E+00
96-09	3.17E+02	NA	< 1.16E+00	< 2.19E+00	< 2.67E+00
96-13	4.18E+03	NA	< 1.15E+01	< 2.20E+01	< 2.17E+01
96-15	4.01E+03	NA	< 1.75E+01	< 3.23E+01	< 4.10E+00
96-04 U	4.08E+03	NA	< 1.28E+01	< 3.57E+01	< 2.07E+01
96-04 L	2.73E+03	NA	< 7.14E+00	< 1.93E+01	< 1.60E+01
96-06 U	NA	NA	NA	NA	NA
96-06 M	3.35E+03	NA	< 9.89E+00	< 1.55E+01	< 1.78E+01
96-06 L	4.56E+03	NA	< 1.79E+01	< 2.98E+01	< 1.82E+01
96-11 U	1.42E+02	NA	< 1.54E+00	< 1.66E+00	< 6.34E+00
96-11 L	3.52E+02	NA	< 2.88E+00	< 5.22E+00	< 2.36E+00

µCi/g: µCi/g dried sludge
NA: not analyzed

	^{57}Co.gea	^{60}Co.gea	^{60}Co.gea PNNL	^{134}Cs.gea	^{134}Cs.gea PNNL
Sample	µCi/g	µCi/g	µCi/g	µCi/g	µCi/g
96-01	< 1.54E-01	< 4.47E-02	1.66E-02	< 1.77E-01	< 3.00E-03
96-05	< 7.88E+00	8.92E-01	4.01E-01	< 1.80E+00	2.41E-01
96-08	< 4.74E+00	6.94E-01	NA	< 3.68E-01	NA
96-09	< 1.84E+00	2.44E+00	NA	< 1.79E-01	NA
96-13	< 4.55E+00	< 9.07E-01	NA	< 1.54E+00	NA
96-15	< 4.10E+00	< 1.66E+00	NA	< 2.29E+00	NA
96-04 U	< 1.10E+01	2.51E+00	NA	< 2.70E+00	NA
96-04 L	3.73E-02	1.35E+00	NA	< 1.49E+00	NA
96-06 U	NA	NA	NA	NA	NA
96-06 M	< 5.13E+00	< 8.57E-01	NA	< 1.10E+00	NA
96-06 L	3.38E+00	< 1.46E+00	NA	< 2.13E+00	NA
96-11 U	< 1.22E+02	1.27E+00	NA	< 1.37E+00	NA
96-11 L	< 1.18E+00	1.99E+00	NA	< 4.24E-01	NA

	^{137}Cs.gea	^{137}Cs.gea PNNL	^{152}Eu.gea	^{154}Eu.gea	^{154}Eu.gea PNNL
Sample	µCi/g	µCi/g	µCi/g	µCi/g	µCi/g
96-01	3.29E+02	1.17E+00	< 1.10E-01	< 1.65E-01	4.38E-02
96-05	1.14E+03	5.92E+02	< 3.28E+00	1.86E+01	9.55E+00
96-08	1.18E+03	NA	< 2.68E-01	1.06E+01	NA
96-09	2.76E+02	NA	< 2.63E-01	4.18E+00	NA
96-13	6.48E+02	NA	< 4.04E+00	9.12E+00	NA
96-15	7.95E+02	NA	< 5.86E+00	9.40E+00	NA
96-04 U	1.95E+03	NA	< 4.03E+00	2.41E+01	NA
96-04 L	9.81E+02	NA	< 1.88E+00	1.70E+01	NA
96-06 U	NA	NA	NA	NA	NA
96-06 M	3.20E+02	NA	< 3.13E+00	1.07E+01	NA
96-06 L	7.85E+02	NA	< 6.46E+00	1.09E+01	NA
96-11 U	5.01E+01	NA	< 4.53E-01	6.37E+00	NA
96-11 L	2.25E+02	NA	< 6.63E-01	2.52E+00	NA

µCi/g: µCi/g dried sludge
NA: not analyzed

	^{155}Eu.gea	^{155}Eu.gea PNNL	^{94}Nb.gea	^{237}Np	^{238}Pu
Sample	µCi/g	µCi/g	µCi/g	µCi/g	µCi/g
96-01	< 6.43E-01	1.82E-02	< 4.89E-02	< 7.44E-03	5.41E-03
96-05	1.22E+01	4.83E+00	< 1.17E+00	7.94E-03	1.62E+01
96-08	5.26E+00	NA	< 2.29E-01	< 1.19E-02	1.74E+01
96-09	1.60E+00	NA	< 1.25E-01	< 2.26E-02	5.89E+00
96-13	< 6.59E+00	NA	< 9.68E-01	1.81E-02	< 7.04E+01
96-15	< 9.77E+00	NA	< 1.61E+00	1.25E-02	8.20E+00
96-04 U	< 1.41E+01	NA	< 1.39E+00	< 4.25E-02	4.07E+01
96-04 L	9.04E+00	NA	< 9.44E-01	1.18E-02	2.16E+01
96-06 U	NA	NA	NA	NA	NA
96-06 M	< 6.86E+00	NA	< 1.06E+00	1.91E-02	1.83E+01
96-06 L	< 8.54E+00	NA	< 1.80E+00	1.21E-02	1.61E+01
96-11 U	2.73E+00	NA	< 2.21E-01	4.94E-02	8.71E+00
96-11 L	< 1.49E+00	NA	< 2.81E-01	< 2.04E-02	3.51E+00

	$^{239/240}$Pu	^{239}Pu.icp/ms PNNL	^{240}Pu.icp/ms PNNL	^{226}Ra.gea	^{106}Ru/Rh.gea
Sample	µCi/g	µCi/g	µCi/g	µCi/g	µCi/g
96-01	5.01E-02	1.57E+01	< 4.54E-02	< 4.80E+00	< 3.51E+00
96-05	1.53E+02	9.70E+01	3.89E+01	< 4.81E+01	< 3.64E+01
96-08	9.35E+01	NA	NA	< 9.46E+00	< 7.20E+00
96-09	3.40E+01	NA	NA	< 4.51E+00	< 3.42E+00
96-13	1.10E+02	NA	NA	< 4.12E+01	< 3.18E+01
96-15	9.54E+01	NA	NA	< 6.05E+01	< 4.47E+01
96-04 U	1.90E+02	NA	NA	< 6.81E+01	< 5.37E+01
96-04 L	9.97E+01	NA	NA	< 3.60E+01	< 2.81E+01
96-06 U	NA	NA	NA	NA	NA
96-06 M	1.04E+02	NA	NA	< 2.90E+01	< 2.14E+01
96-06 L	9.29E+01	NA	NA	< 5.84E+01	< 4.45E+01
96-11 U	4.56E+01	NA	NA	< 3.30E+00	< 2.62E+00
96-11 L	1.72E+01	NA	NA	< 1.08E+01	< 7.91E+00

µCi/g: µCi/g dried sludge
NA: not analyzed

	$^{89/90}$Sr	^{99}Tc	^{208}Tl.gea	Total Carbon	TOC
Sample	µCi/g	µCi/g	µCi/g	µg/g	µg/g
96-01	6.32E+01	2.94E-01	< 3.18E+00	2.96E+04	1.37E+04
96-05	2.19E+03	6.46E+00	< 3.06E+01	1.08E+03	6.66E+02
96-08	9.40E+02	3.42E+01	< 6.20E+00	4.81E+03	1.22E+03
96-09	2.81E+01	2.72E+01	< 2.97E+00	1.04E+04	8.45E+03
96-13	1.78E+03	1.02E+01	< 2.67E+01	9.83E+02	5.46E+02
96-15	1.58E+03	1.23E+00	< 3.81E+01	1.00E+03	6.52E+02
96-04 U	6.04E+02	NA	< 4.45E+01	7.95E+03	3.72E+03
96-04 L	9.99E+02	NA	< 2.36E+01	4.40E+03	9.83E+02
96-06 U	NA	NA	NA	NA	NA
96-06 M	1.78E+03	NA	< 1.82E+01	7.87E+02	6.54E+02
96-06 L	1.98E+03	NA	< 3.81E+01	6.38E+02	4.32E+02
96-11 U	3.67E+01	NA	< 2.20E+00	9.78E+03	5.52E+03
96-11 L	6.22E+01	NA	< 6.92E+00	6.63E+03	4.26E+03

	TIC	Ag.icp	Al.icp	Al.icp/ms PNNL	B.icp
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	1.43E+04	1.82E+01	2.99E+04	1.14E+03	2.07E+02
96-05	2.89E+02	1.27E+02	1.32E+04	1.03E+04	1.54E+02
96-08	2.38E+03	5.06E+01	7.86E+04	NA	3.75E+02
96-09	4.21E+03	< 2.06E+01	1.39E+05	NA	4.75E+02
96-13	4.09E+02	1.56E+02	1.45E+04	NA	1.66E+02
96-15	4.81E+02	1.95E+02	1.60E+04	NA	1.80E+02
96-04 U	4.14E+03	5.40E+01	1.29E+05	NA	5.36E+02
96-04 L	3.59E+03	5.17E+01	8.88E+04	NA	1.88E+02
96-06 U	NA	NA	NA	NA	NA
96-06 M	5.81E+02	1.57E+02	1.45E+04	NA	1.61E+02
96-06 L	3.03E+02	1.59E+02	1.04E+04	NA	1.53E+02
96-11 U	7.29E+03	2.35E+01	1.39E+05	NA	1.02E+03
96-11 L	2.56E+03	< 2.26E+01	6.55E+04	NA	2.39E+02

µCi/g: µCi/g dried sludge
µg/g: µg/g dried sludge
NA: not analyzed

	Ba.icp	Be.icp	Bi.icp	Ca.icp	Ca.icp/ms PNNL
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	< 2.57E+01	< 2.57E+00	< 5.13E+01	4.12E+02	7.11E+01
96-05	1.33E+02	< 1.14E+01	6.59E+02	7.79E+02	1.09E+02
96-08	1.58E+02	5.66E+01	3.42E+02	1.41E+03	NA
96-09	1.52E+02	1.01E+02	2.06E+02	1.61E+03	NA
96-13	1.30E+02	< 1.11E+01	6.63E+02	6.98E+02	NA
96-15	1.76E+02	< 1.13E+01	4.30E+02	< 2.25E+02	NA
96-04 U	3.34E+02	6.00E+01	3.66E+02	2.13E+03	NA
96-04 L	1.63E+02	1.78E+01	3.18E+02	1.25E+03	NA
96-06 U	NA	NA	NA	NA	NA
96-06 M	< 1.27E+02	< 1.27E+01	2.96E+02	< 2.56E+02	NA
96-06 L	1.25E+02	< 1.15E+01	2.95E+02	< 2.29E+02	NA
96-11 U	< 1.01E+02	1.87E+02	< 2.02E+02	1.14E+03	NA
96-11 L	1.70E+02	7.50E+01	< 2.26E+02	2.47E+03	NA

	Cd.icp	Cr.icp	Cu.icp	Fe.icp	Fe.icp/ms PNNL
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	7.79E+00	2.30E+03	9.52E+00	4.59E+02	1.68E+03
96-05	2.35E+01	< 2.27E+01	9.38E+01	6.98E+03	6.23E+03
96-08	4.09E+01	3.04E+02	2.56E+02	7.27E+04	NA
96-09	1.46E+02	6.20E+02	1.28E+03	2.10E+05	NA
96-13	2.02E+01	< 2.23E+01	8.07E+01	2.81E+03	NA
96-15	< 1.13E+01	< 2.25E+01	6.81E+01	6.56E+03	NA
96-04 U	8.54E+01	2.17E+02	4.61E+02	8.01E+04	NA
96-04 L	4.89E+01	1.20E+02	2.15E+02	4.73E+04	NA
96-06 U	NA	NA	NA	NA	NA
96-06 M	2.30E+01	< 2.56E+01	8.86E+01	1.80E+03	NA
96-06 L	2.40E+01	< 2.29E+01	6.99E+01	1.20E+03	NA
96-11 U	2.29E+02	6.32E+02	1.18E+03	1.21E+05	NA
96-11 L	1.83E+02	8.02E+02	7.46E+02	2.56E+05	NA

µg/g: µg/g dried sludge
NA: not analyzed

	K.icp	Mg.icp	Mn.icp	Na.icp	Ni.icp
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	3.60E+03	< 5.13E+01	1.40E+02	2.50E+05	1.52E+02
96-05	< 1.14E+03	2.21E+03	< 2.27E+01	4.16E+02	< 4.55E+01
96-08	< 8.27E+02	1.68E+03	9.18E+01	7.28E+02	6.17E+01
96-09	< 1.03E+03	1.98E+03	6.98E+02	8.25E+02	4.98E+02
96-13	< 1.11E+03	1.90E+03	< 2.23E+01	4.30E+02	< 4.44E+01
96-15	< 1.13E+03	1.93E+03	< 2.25E+01	4.71E+02	< 4.50E+01
96-04 U	< 1.10E+03	1.41E+03	1.99E+02	1.01E+03	2.96E+02
96-04 L	< 6.60E+02	1.24E+03	9.21E+01	5.20E+02	7.20E+01
96-06 U	NA	NA	NA	NA	NA
96-06 M	< 1.27E+03	< 2.56E+02	< 2.56E+01	4.41E+02	< 5.10E+01
96-06 L	< 1.15E+03	< 2.29E+02	< 2.29E+01	4.01E+02	< 4.60E+01
96-11 U	< 1.01E+03	< 2.02E+02	8.10E+02	1.75E+03	2.05E+02
96-11 L	< 1.13E+03	2.96E+02	5.56E+02	5.79E+02	1.29E+02

	P.icp	Pb.icp	Se.icp	Sm.icp	Tl.icp
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	2.21E+03	6.77E+01	< 5.13E+01	< 5.13E+01	< 1.03E+02
96-05	< 4.55E+02	< 2.27E+02	< 2.27E+02	< 2.27E+02	< 4.55E+02
96-08	< 3.32E+02	1.67E+02	< 1.65E+02	< 1.65E+02	< 3.32E+02
96-09	9.43E+02	7.66E+02	< 2.06E+02	< 2.06E+02	< 4.12E+02
96-13	< 4.44E+02	< 2.23E+02	< 2.23E+02	< 2.23E+02	< 4.44E+02
96-15	< 4.50E+02	< 2.25E+02	< 2.25E+02	< 2.25E+02	< 4.50E+02
96-04 U	< 4.37E+02	3.64E+02	< 2.19E+02	< 2.19E+02	< 4.37E+02
96-04 L	< 2.63E+02	1.35E+02	< 1.32E+02	< 1.32E+02	< 2.63E+02
96-06 U	NA	NA	NA	NA	NA
96-06 M	< 5.10E+02	< 2.56E+02	< 2.56E+02	< 2.56E+02	< 5.10E+02
96-06 L	< 4.60E+02	< 2.29E+02	< 2.29E+02	< 2.29E+02	< 4.60E+02
96-11 U	1.13E+03	6.38E+02	< 2.02E+02	< 2.02E+02	< 4.04E+02
96-11 L	6.15E+02	5.14E+02	< 2.26E+02	< 2.26E+02	< 4.51E+02

µg/g: µg/g dried sludge

	U.icp	U.phos	U.las PNNL	U.icp/ms PNNL	U.las PNNL accelerated sample
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	< 2.57E+02	9.44E+02	8.24E+05	6.50E+05	8.54E+05
96-05	7.48E+05	5.85E+05	8.81E+05	6.43E+05	6.64E+05
96-08	3.85E+05	3.10E+05	4.04E+05	NA	NA
96-09	1.31E+05	1.31E+05	8.81E+05	NA	NA
96-13	7.47E+05	7.40E+05	8.20E+05	NA	NA
96-15	6.68E+05	4.92E+05	8.13E+05	NA	NA
96-04 U	3.18E+05	2.97E+05	NA	NA	NA
96-04 L	3.18E+05	2.73E+05	5.46E+05	NA	NA
96-06 U			NA	NA	NA
96-06 M	7.49E+05	6.11E+05	8.26E+05	NA	NA
96-06 L	7.39E+05	6.06E+05	8.43E+05	NA	NA
96-11 U	1.33E+05	1.28E+05	NA	NA	NA
96-11 L	7.79E+04	7.33E+04	9.33E+04	NA	NA

	Zn.icp	Zr.icp	Zr.icp/ms PNNL		
Sample	µg/g	µg/g	µg/g		
96-01	2.41E+01	1.20E+01	3.22E+03		
96-05	5.22E+01	< 2.27E+01	1.32E+02		
96-08	4.69E+02	3.29E+02	NA		
96-09	1.39E+03	5.57E+02	NA		
96-13	2.23E+01	< 2.23E+01	NA		
96-15	5.00E+01	6.71E+01	NA		
96-04 U	6.88E+02	8.90E+02	NA		
96-04 L	2.73E+02	3.45E+02	NA		
96-06 U			NA		
96-06 M	< 2.56E+01	< 2.56E+01	NA		
96-06 L	< 2.30E+01	< 2.29E+01	NA		
96-11 U	1.26E+03	7.88E+02	NA		
96-11 L	1.42E+03	2.85E+02	NA		

µg/g: µg/g dried sludge
NA: not analyzed

	^{233}U.tims \$ PNNL	^{234}U.tims \$ PNNL	^{235}U.tims \$ PNNL	^{236}U.tims \$ PNNL	^{238}U.tims \$ PNNL
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	< 4.03E+00	7.65E+01	7.76E+03	4.37E+02	8.15E+05
96-05	< 4.31E+00	4.42E+01	4.32E+03	3.41E+02	8.76E+05
96-08	< 1.98E+00	3.18E+01	2.62E+03	3.29E+02	4.01E+05
96-09	< 4.31E+00	6.32E+01	6.11E+03	6.55E+02	8.74E+05
96-13	< 4.01E+00	7.10E+01	5.92E+03	5.29E+02	8.13E+05
96-15	< 3.98E+00	5.59E+01	5.76E+03	3.67E+02	8.06E+05
96-04 U	NA	NA	NA	NA	NA
96-04 L	< 2.67E+00	4.05E+01	3.27E+03	5.28E+02	5.42E+05
96-06 U	NA	NA	NA	NA	NA
96-06 M	< 4.04E+00	8.65E+01	6.11E+03	6.59E+02	8.19E+05
96-06 L	< 4.12E+00	8.12E+01	6.22E+03	6.93E+02	8.36E+05
96-11 U	NA	NA	NA	NA	NA
96-11 L	< 4.56E-01	1.35E+01	6.68E+02	7.07E+01	9.25E+04

	^{233}U.icp/ms PNNL	^{234}U.icp/ms PNNL	^{235}U.icp/ms PNNL	^{236}U.icp/ms PNNL	^{238}U.icp/ms PNNL
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	4.35E-01	4.95E+01	6.32E+03	3.70E+02	6.42E+05
96-05	2.00E-01	3.52E+01	3.33E+03	2.89E+02	6.35E+05

	^{125}Sb.gea PNNL	m/z241 icp/ms PNNL			^{238}U.gea
	µCi/g	µg/g			µg/g
96-01	< 2.00E-02	< 2.00E-01			2.67E+05
96-05	1.11E+00	5.03E+01			< 1.49E+07

\$: based on the total uranium (U.las µg/g dried sludge) by PNNL
µg/g: µg/g dried sludge
µCi/g: µCi/g dried sludge
NA: not analyzed

	Br$^-$.ic	Cl$^-$.ic	F$^-$.ic	NO$_2^-$.ic	NO$_3^-$.ic
Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-04 U	< 1.18E+01	5.45E+00	< 1.13E+00	< 1.02E+01	1.74E+01
96-04 L	< 2.18E+00	1.01E+00	< 2.09E-01	< 1.88E+00	3.21E+00
96-06 M	< 7.41E-01	2.57E+00	3.13E-01	< 6.39E-01	< 8.21E-01
96-06 L	< 3.89E-01	1.35E+00	1.65E-01	< 3.35E-01	< 4.31E-01
96-08	< 4.40E+00	2.25E+00	< 4.21E-01	< 3.79E+00	< 4.88E+00
96-09	< 9.84E+00	2.63E+00	< 9.41E-01	< 8.48E+00	< 1.09E+01
96-11 U	< 2.51E+01	9.71E+00	< 2.40E+00	< 2.16E+01	< 2.78E+01
96-11 L	< 4.55E+00	1.76E+00	< 4.35E-01	< 3.93E+00	< 5.05E+00
96-13	< 4.50E-01	3.94E-01	1.92E-01	< 3.88E-01	< 4.98E-01
96-15	< 4.83E-01	2.43E-01	< 4.62E-02	< 4.17E-01	< 5.36E-01

	Oxalate.ic	PO$_4^{3-}$.ic	SO$_4^{2-}$.ic		
Sample	µg/g	µg/g	µg/g		
96-04 U	< 9.92E+00	< 1.13E+01	2.43E+01		
96-04 L	< 1.83E+00	< 2.09E+00	4.50E+00		
96-06 M	< 6.23E-01	< 7.08E-01	4.11E+00		
96-06 L	< 3.27E-01	< 3.72E-01	2.16E+00		
96-08	< 3.70E+00	< 4.21E+00	1.70E+01		
96-09	< 8.27E+00	< 9.41E+00	2.20E+01		
96-11 U	< 2.11E+01	< 2.40E+01	< 2.76E+01		
96-11 L	< 3.83E+00	< 4.35E+00	< 5.01E+00		
96-13	< 3.78E-01	< 4.30E-01	1.44E+00		
96-15	< 4.06E-01	< 4.62E-01	2.36E+00		

µg/g: µg/g dried sludge
NA: not analyzed

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APPENDIX F

STATISTICAL ANALYSIS OF K EAST CANISTER SLUDGE CHEMISTRY DATA

T. L. Welsh

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APPENDIX F

STATISTICAL ANALYSIS OF K EAST CANISTER SLUDGE CHEMISTRY DATA

Boxplots are an effective way to view a batch of data. A boxplot shows (1) where the middle of the data (also known as the median) lies, (2) how spread out the middle is, and (3) how the tails relate to it. The box encloses the middle 50 percent of the data. The median is the vertical line inside the box; the position of this line is an indication of the symmetry of the data. Horizontal lines (called whiskers) extend from each end of the box; the left whisker goes from the box (also known as the lower hinge) to the smallest data point within 1.5 interquartile ranges, while the right whisker goes from the box (also known as the upper hinge) to the largest data point within 1.5 interquartile ranges. The box and the whiskers provide a graphical view of the distribution of the data. Any data that are further than 3 times the interquartile ranges from the box are called "outliers" and are plotted as individual points.

The data illustrated in the following boxplots are listed in either Appendix D or Appendix E depending on the units provided with each figure. A boxplot does not exist if more than half the data were reported as "less than" values. For those analytes with less than half the data reported as "less than" values, the upper value of the "less than" (e.g., 3.5 for < 3.5) was used to represent the analytical result.

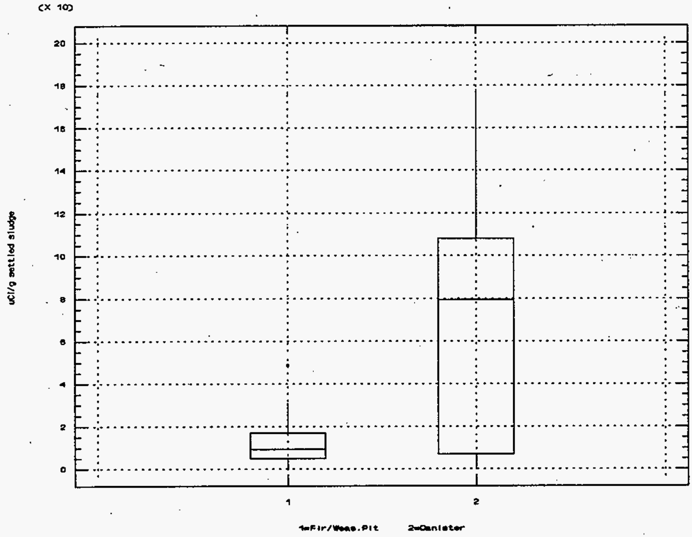
For comparison purposes the boxplot illustrating the KE Basin Floor and Weasel Pit data (Makenas 1996b) is provided.

Interpretations of two boxplots are as follows.

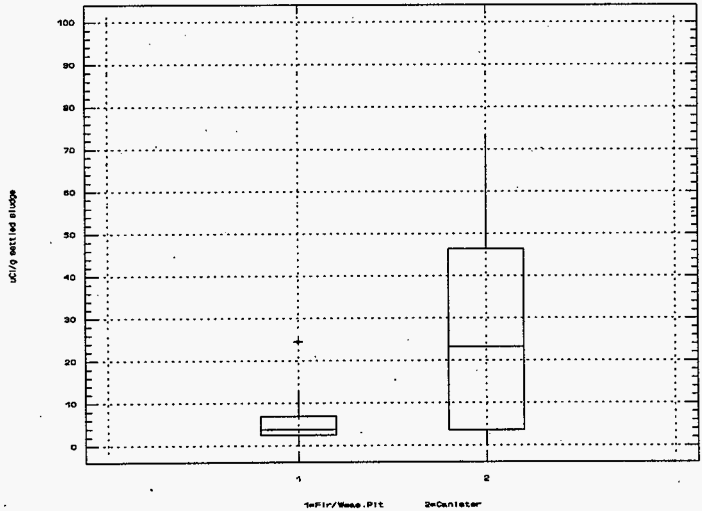
The boxplot for ^{137}Cs on page F-6 indicates that for the Basin floor/Weasel Pit data there is one "outlier" on the upper end of this data set. This "outlier" represents the data from location 0-09. The plot depicts a strong skewness toward the lower end (the right whisker is longer than the left whisker, plus the median is to the left of center). The ^{137}Cs canister boxplot indicates that there are no "outliers" in this data set. The distribution of the data appears to be symmetrical, i.e., there is no evidence of skewness. Comparison of the two boxplots indicate that (1) the range (maximum-minimum) from the canister data is close to the range from the Basin floor/Weasel Pit data and (2) the median concentration for the canister data is larger than the median concentration for the Basin floor/Weasel Pit data.

The boxplot for ^{238}U on page F-7 indicates that for the Basin floor/ Weasel Pit data there are no "outliers" in this data set. The distribution of the data appears to be symmetrical, i.e., there is no evidence of skewness. The ^{238}Pu canister boxplot indicates that there are three "outliers" in this data set. The distribution of the data appears to be symmetrical, i.e., there is no evidence of skewness. Comparison of the two boxplots indicate that (1) the range (maximum-minimum) from the canister data is greater than the range from the Basin floor/Weasel Pit data and (2) the median concentration for the canister data is larger than the median concentration for the Basin floor/Weasel Pit data.

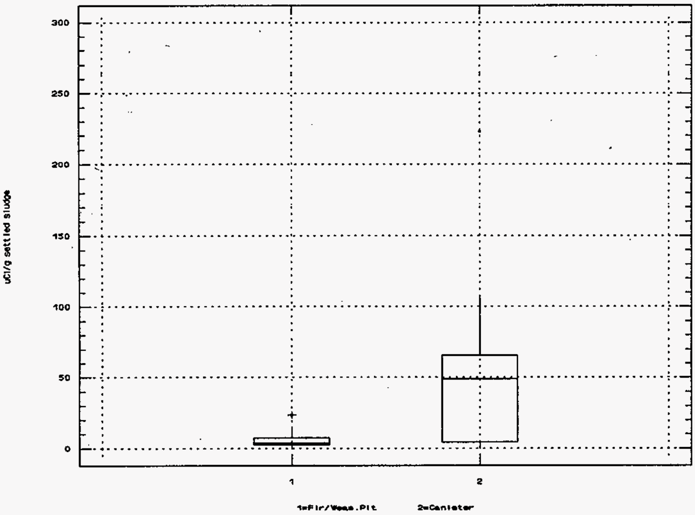
Total Alpha (uCi/$g_{settled\ sludge}$)



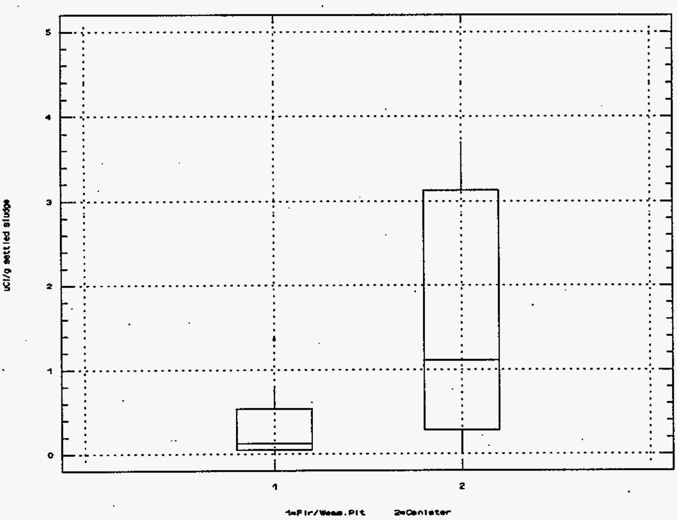
^{241}Am.aea (uCi/$g_{settled\ sludge}$)



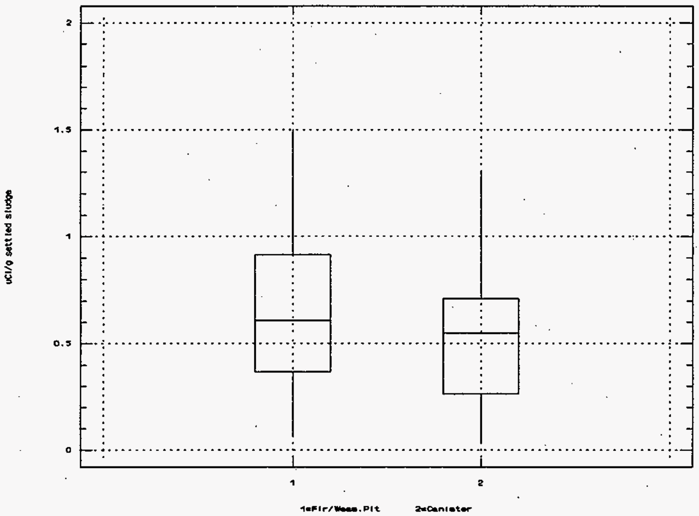
^{241}Am.gea (uCi/g$_{\text{settled sludge}}$)



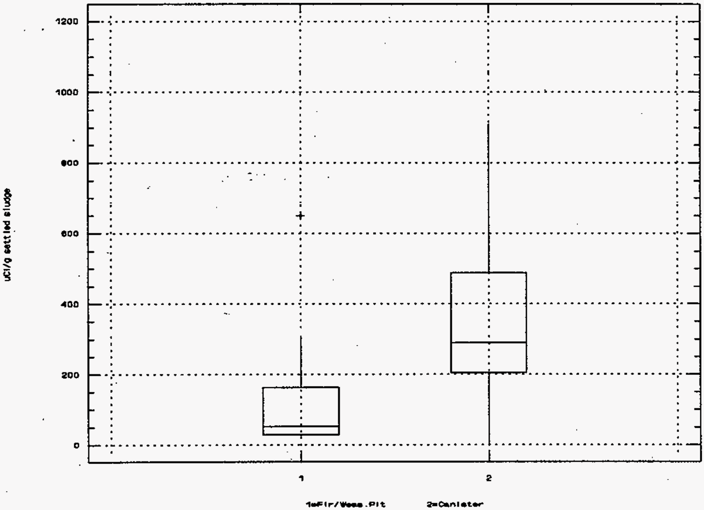
Total Beta (uCi/g$_{\text{settled sludge}}$)



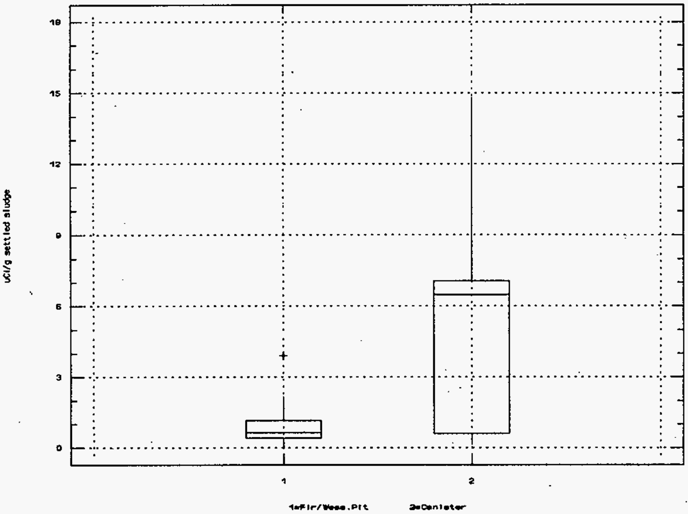
^{60}Co.gea (uCi/g$_{settled\ sludge}$)



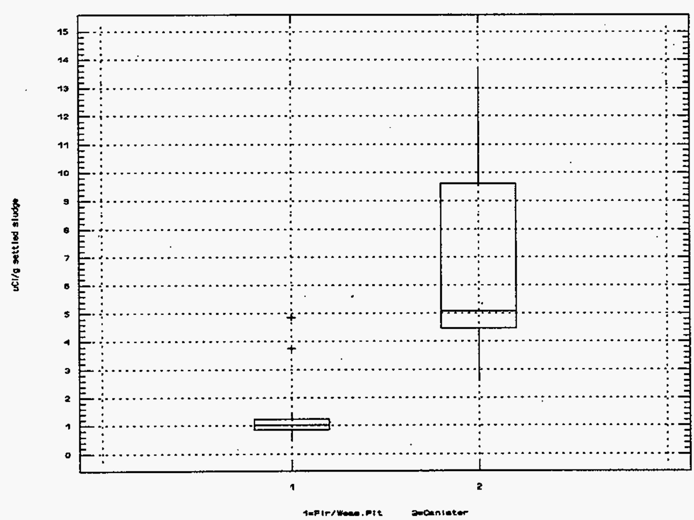
^{137}Cs.gea (uCi/g$_{settled\ sludge}$)



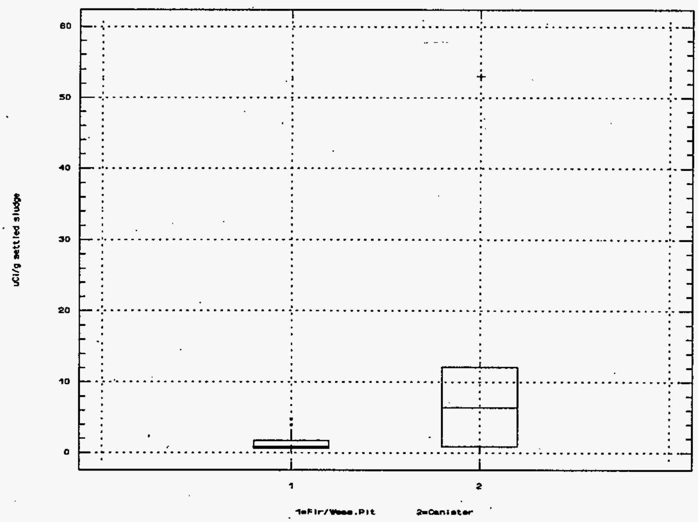
^{154}Eu.gea (uCi/g$_{settled\ sludge}$)



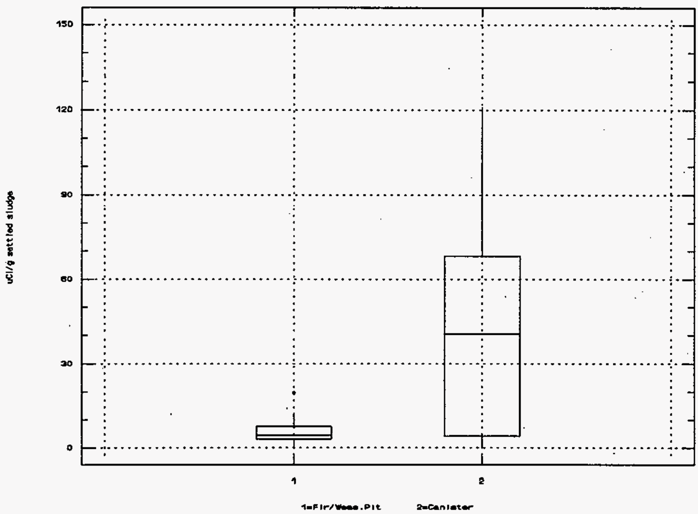
^{237}Np (uCi/g$_{settled\ sludge}$)



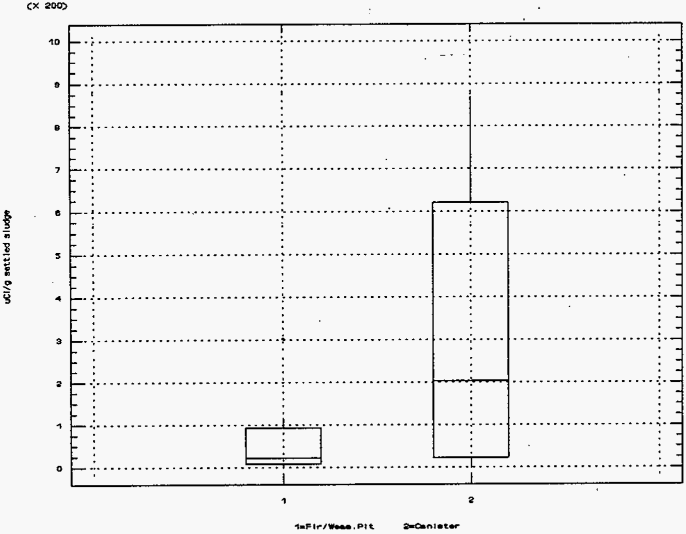
^{238}Pu (uCi/g$_{\text{settled sludge}}$)



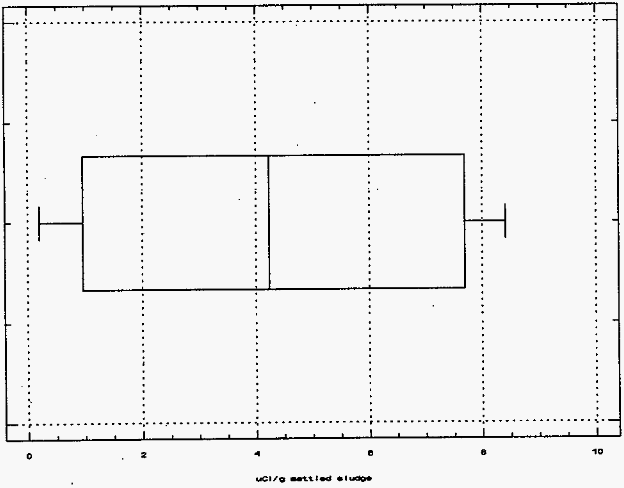
$^{239/240}$Pu (uCi/g$_{\text{settled sludge}}$)



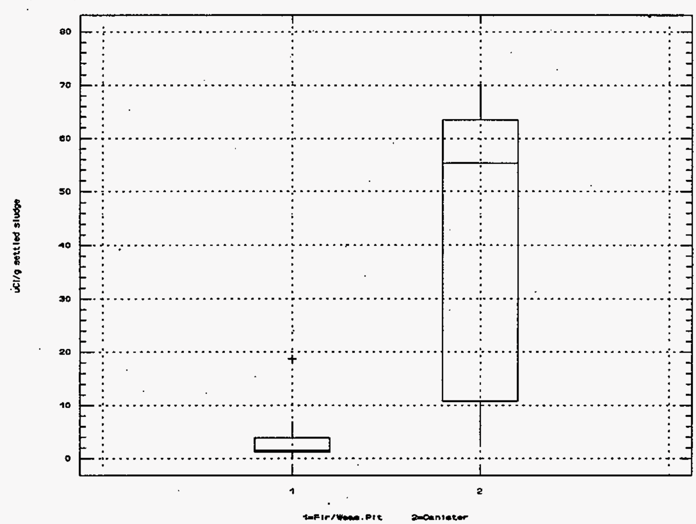
$^{89/90}Sr$ (uCi/g$_{settled\ sludge}$)



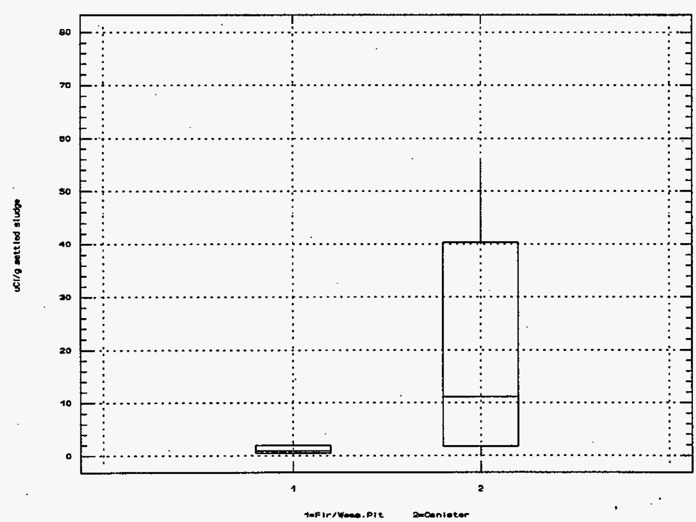
^{99}Tc (uCi/g$_{settled\ sludge}$)



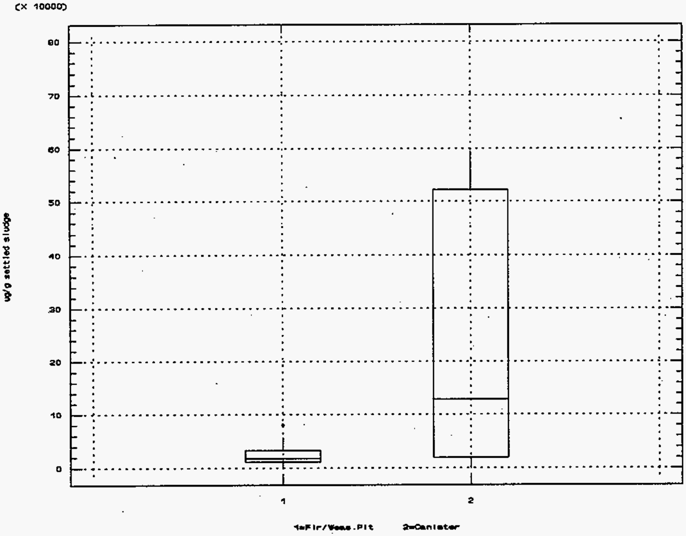
Uranium.las ($ug/g_{settled\ sludge}$)



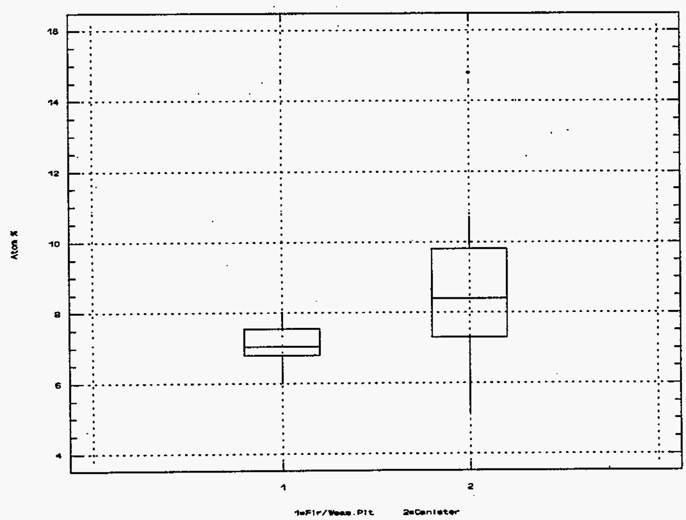
Uranium.phos ($ug/g_{settled\ sludge}$)



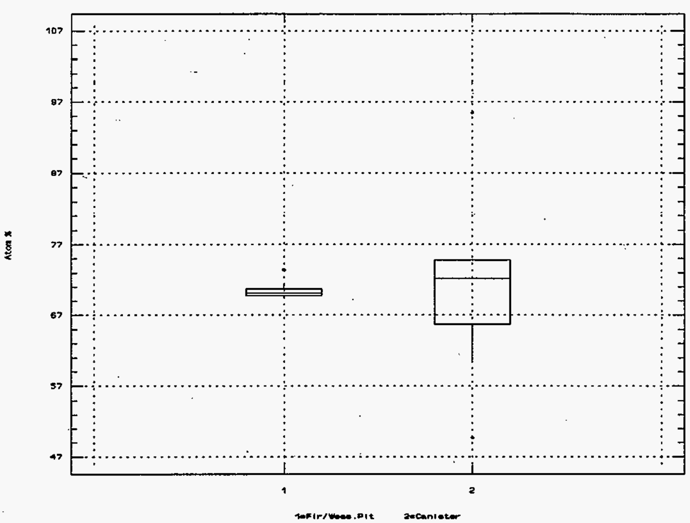
Uranium.icp (ug/$g_{\text{settled sludge}}$)



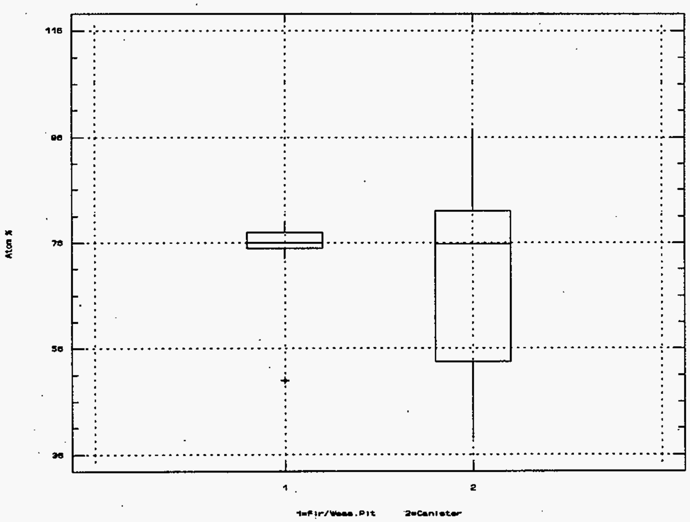
^{234}U.tims (atom%)



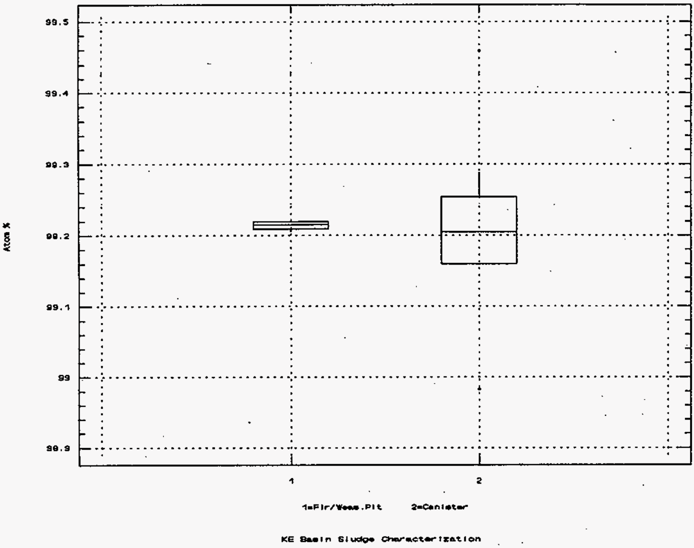
^{235}U.tims (atom %)

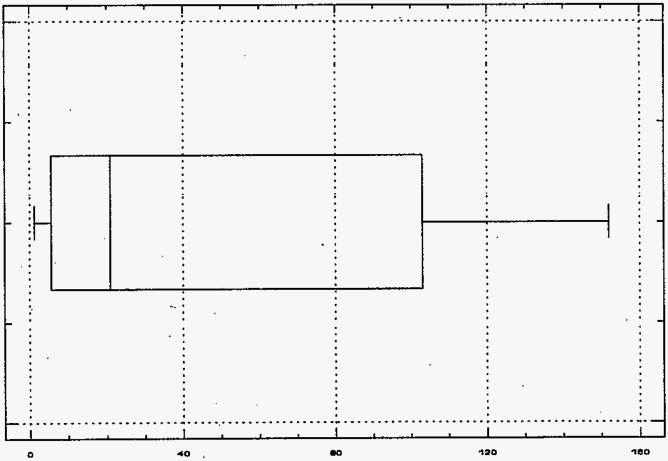


^{236}U.tims (atom%)



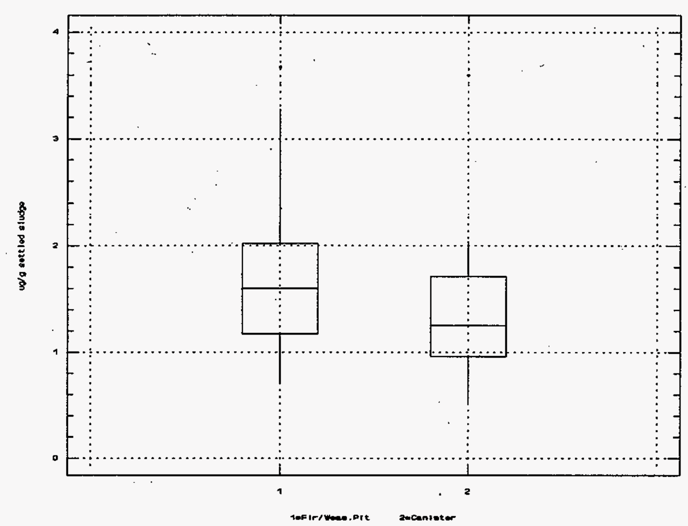
^{238}U.tims (atom %)



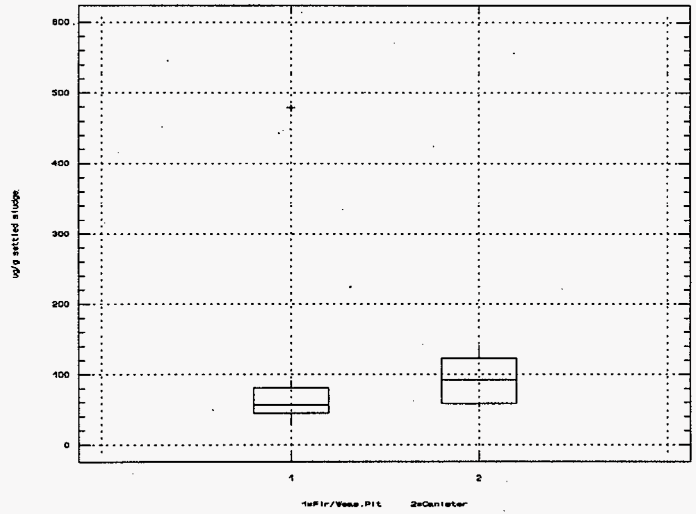


Ag.icp (ug/g$_{\text{settled sludge}}$)

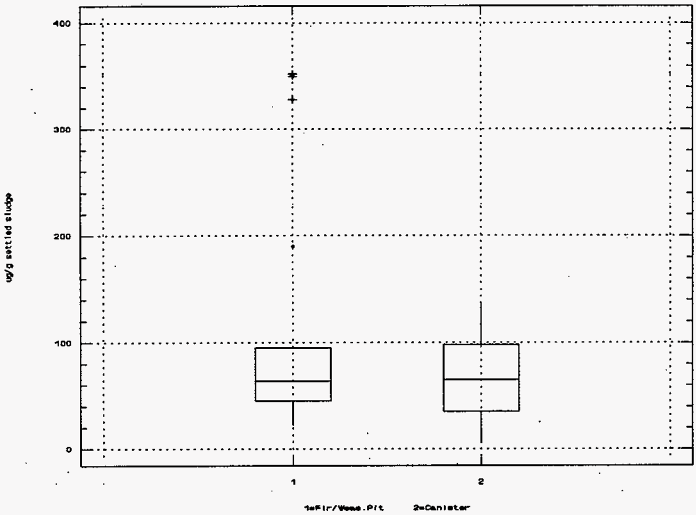
Al.icp (ug/g$_{\text{settled sludge}}$)



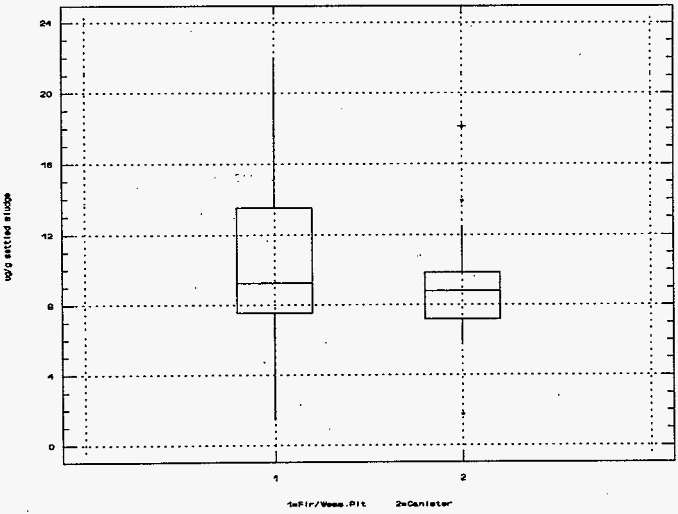
B.icp (ug/g$_{\text{settled sludge}}$)

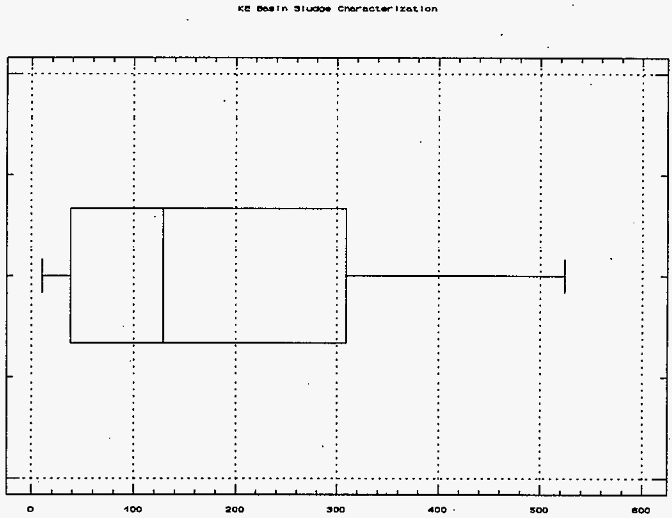


Ba.icp (ug/$g_{\text{settled sludge}}$)



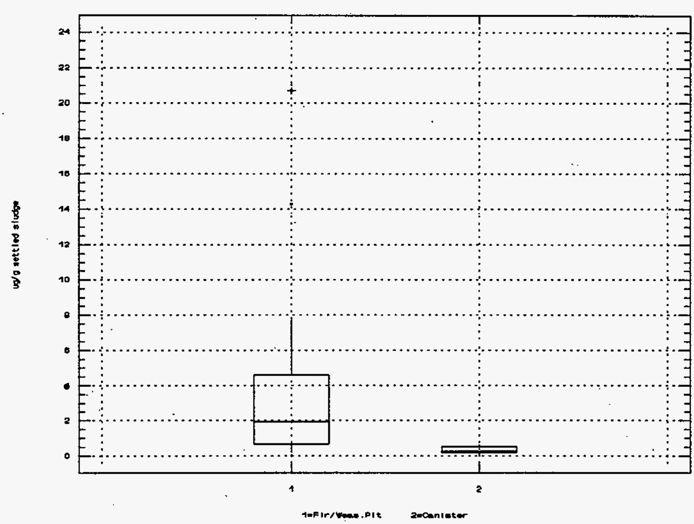
Be.icp (ug/$g_{\text{settled sludge}}$)



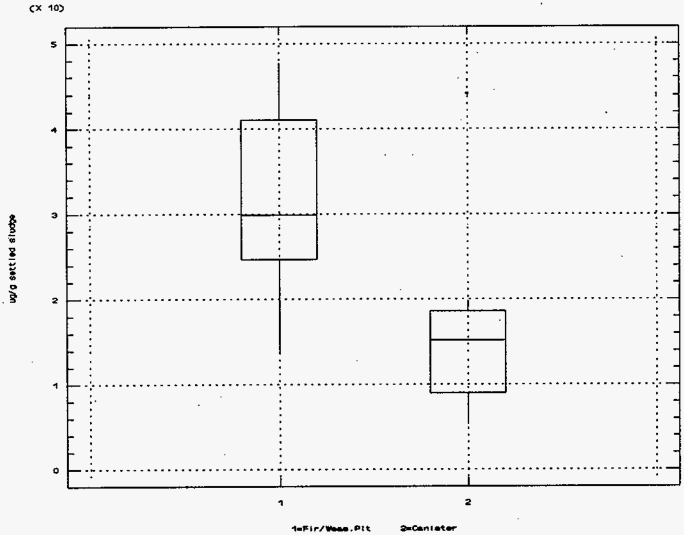


Bi.icp (ug/$g_{\text{settled sludge}}$)

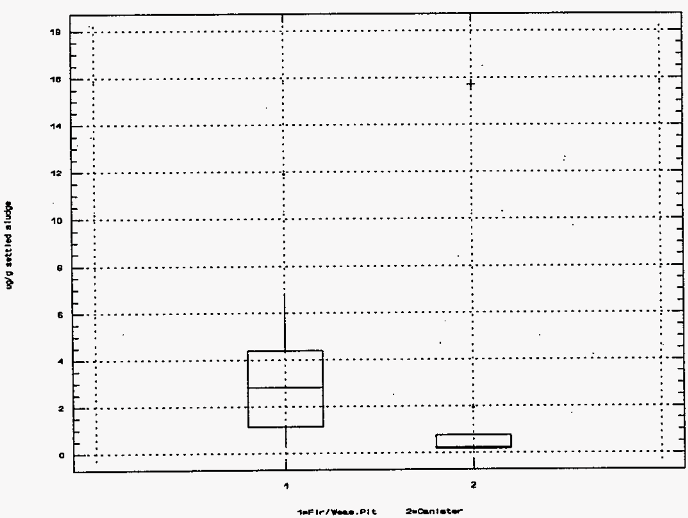
Ca.icp (ug/$g_{\text{settled sludge}}$)



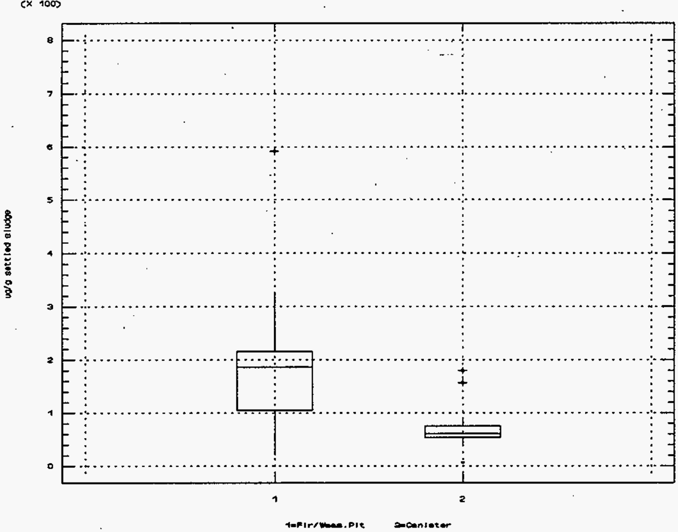
Cd.icp (ug/g$_{\text{settled sludge}}$)



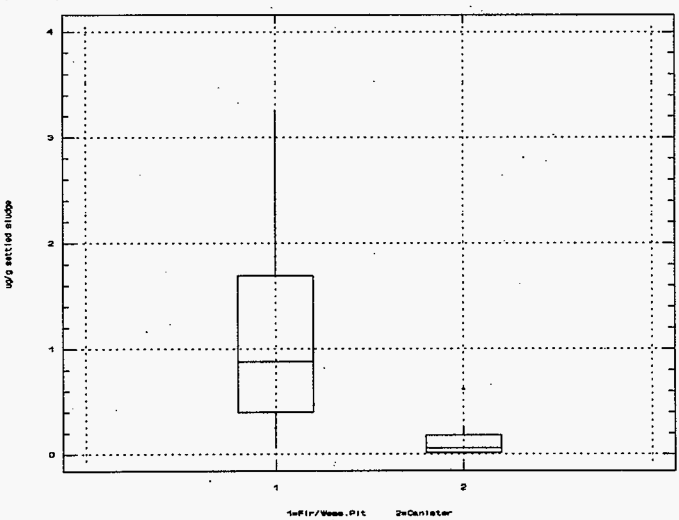
Cr.icp (ug/g$_{\text{settled sludge}}$)



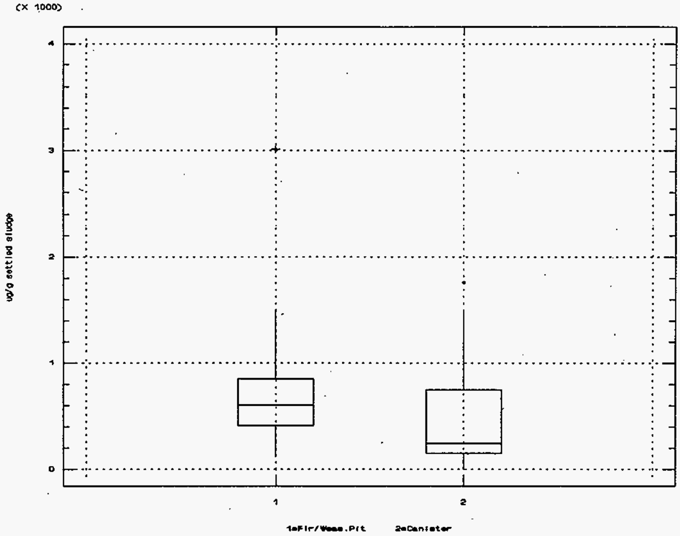
Cu.icp (ug/g$_{\text{settled sludge}}$)



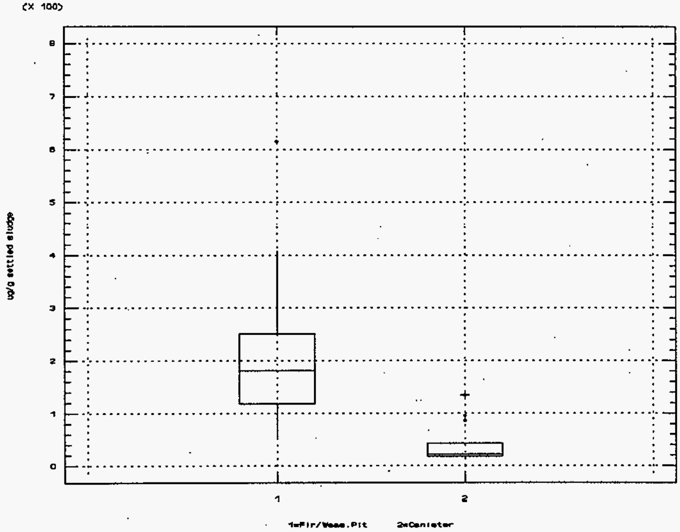
Fe.icp (ug/g$_{\text{settled sludge}}$)

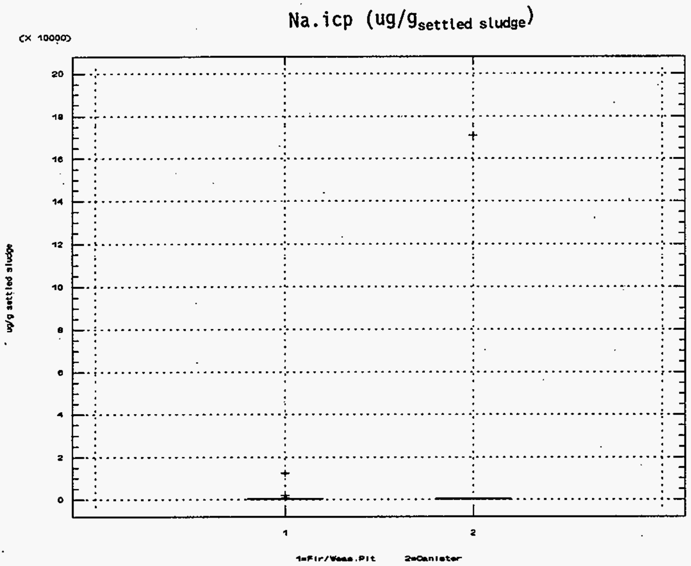


Mg.icp (ug/$g_{settled\ sludge}$)

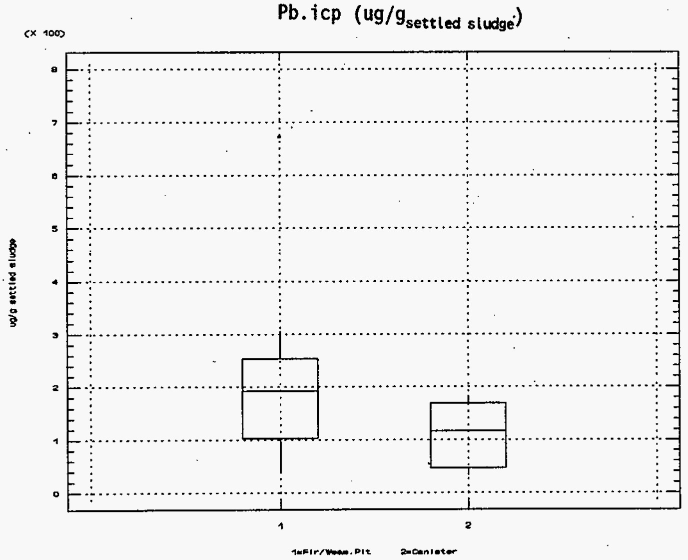


Mn.icp (ug/$g_{settled\ sludge}$)



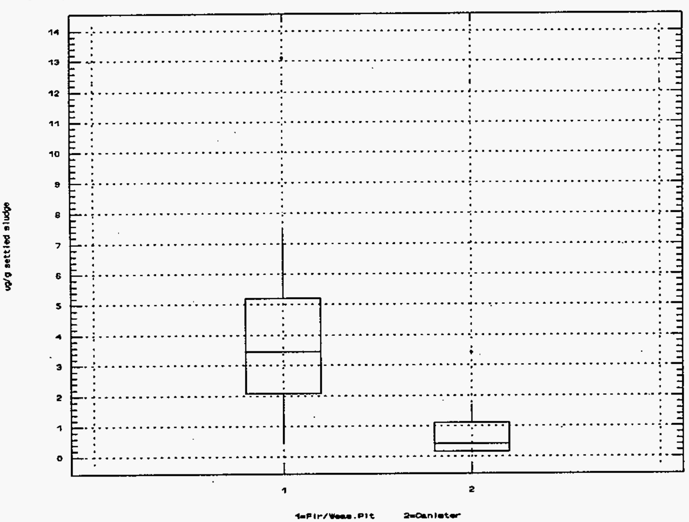


Na.icp (ug/g$_{\text{settled sludge}}$)

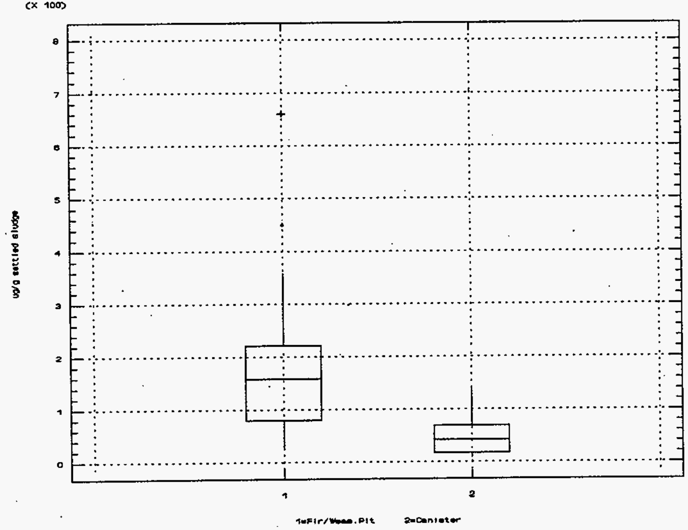


Pb.icp (ug/g$_{\text{settled sludge}}$)

Zn.icp (ug/g$_{\text{settled sludge}}$)

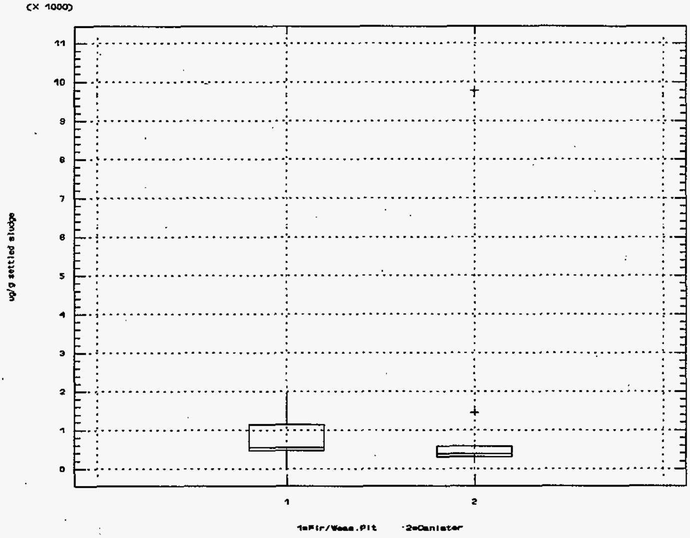


Zr.icp (ug/g$_{\text{settled sludge}}$)



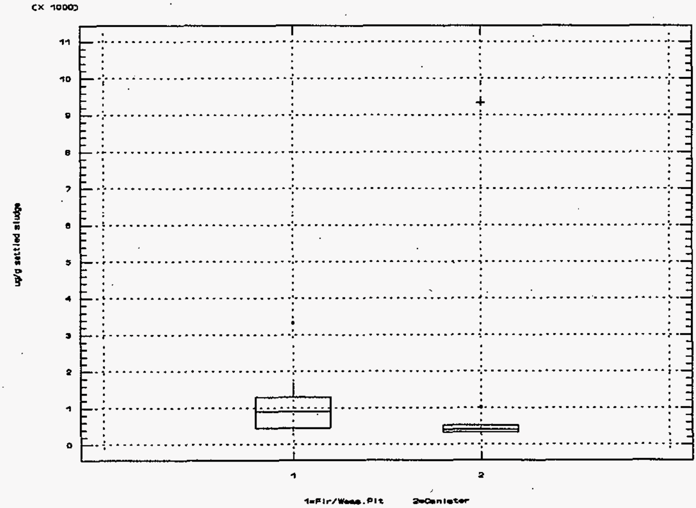
TIC ($ug/g_{settled\ sludge}$)

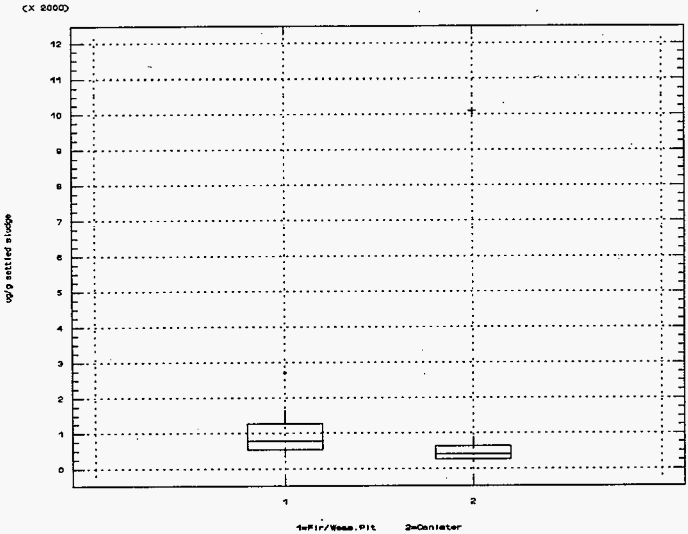
Total Inorganic Carbon



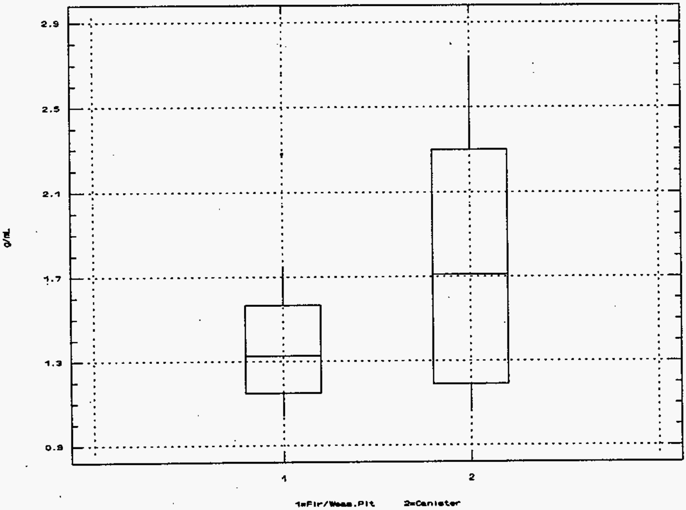
TOC ($ug/g_{settled\ sludge}$)

Total Organic Carbon



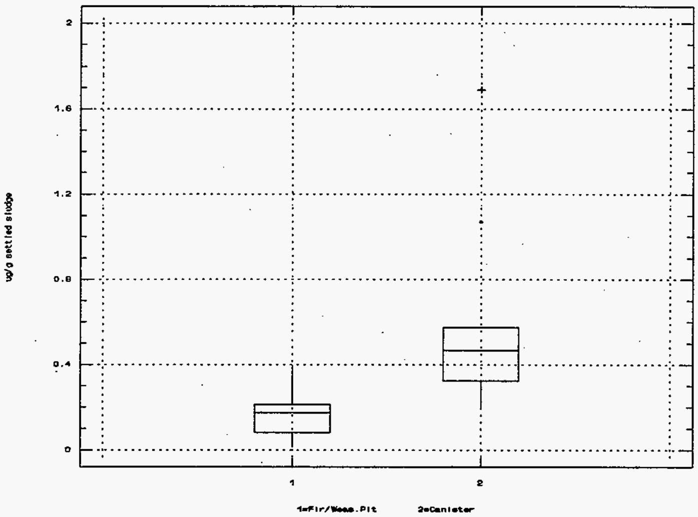


Total Carbon (ug/g$_{\text{settled sludge}}$)

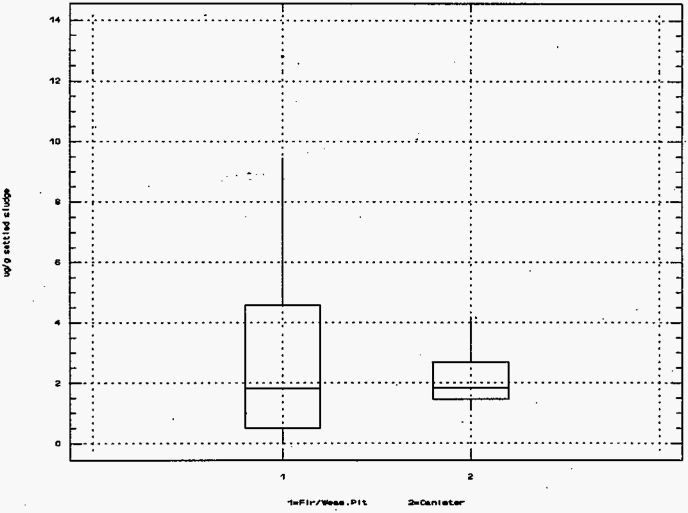


Density (as-settled)

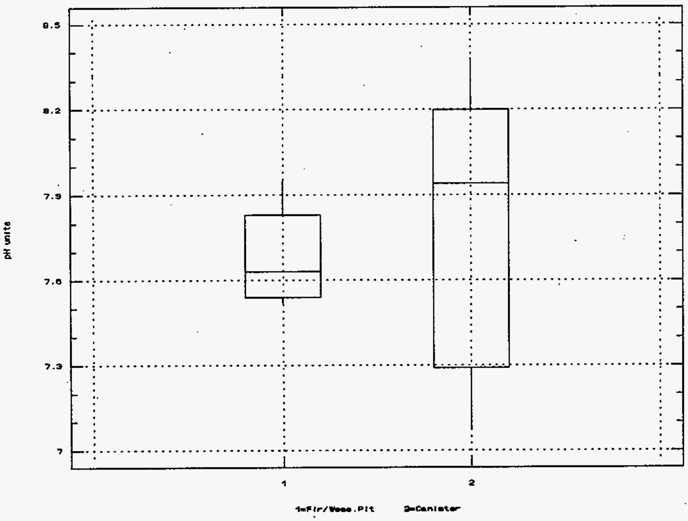
Cl^{-}.ic (ug/$g_{settled\ sludge}$)



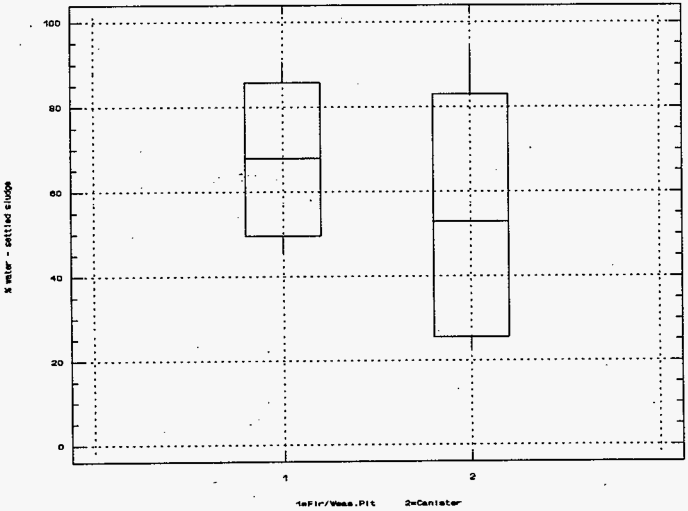
SO_4^{2-}.ic (ug/$g_{settled\ sludge}$)



pH



%Water.222S (wt%-settled sludge)



ANALYTICAL VARIABILITY

Random Analytical Variability

The sampling and analysis plan called for duplicate analyses in order to estimate the random analytical variability. The duplicate data are presented in Miller 1997a and Silvers 1997a. A one-way analysis of variance was computed for each analyte that had two or more duplicate analyses. The one-way analysis of variance used the following model;

$$X_{ij} = \mu + S_i + \epsilon_{ij}$$

where X_{ij} is the analyte of interest (e.g., ^{137}Cs or Al), μ represents the population mean concentration, S_i represents the different samples, and ϵ_{ij} represents the duplicate analyses performed by the laboratory. The analysis of variance computation provides the variance estimate ($\hat{\sigma}^2_\epsilon$) for each analytical method. The relative standard deviation (RSD) was then calculated using the following formula.

$$\text{RSD}(\%) = \frac{\sqrt{\hat{\sigma}^2_\epsilon}}{\overline{X}} \times 100 \text{ , where } \overline{X} = \text{overall mean}$$

The random analytical variability estimate for each analyte is provided in Table F-1. In addition to the RSDs, the number of samples with duplicate measurements used in computing the variability estimate (i.e., the degrees of freedom associated with the variability estimate) is listed in Table F-1.

Systematic Analytical Variability

The systematic analytical variability is estimated from the analysis of laboratory standards or from the analysis of spiked samples. The laboratory standard results and the spiked samples results are reported as percent recovery values using the following formulas.

$$\text{Percent Recovery (Laboratory standard)} = \frac{\text{Measured value}}{\text{known value}} \times 100$$

$$\text{Percent Recovery (Spiked sample)} = \frac{\text{(Spiked sample result - sample}}{\text{known spike amount}}$$

Table F-1. Random/Systematic Analytical Variability Estimates. (Page 1 of 3)

Analyte	Random Variability Estimated from the duplicate sample results		Systematic Variability Estimated from Spike or Standard Analyses	
	N *	RSD (%)	N $	RSD (%)
Ag.icp.w	11	8.07	1	48.5 rs
Ag.icp.wo	10	7.27	1	48.5 rs
Al.icp	11	10.51	1	11.6 rs
Total Alpha	11	5.62	1	15.9 sp
^{241}Am.aea.w	11	8.41	1	17.1 sp
^{241}Am.aea.wo	10	8.24	1	17.1 sp
^{241}Am.gea.w	11	8.39	NA	25.0 ct
^{241}Am.gea.wo	10	8.13	NA	25.0 ct
B.icp	11	12.76	1	8.2 sp
Ba.icp.w	11	8.53	1	8.0 sp
Ba.icp.wo	7	8.79	1	8.0 sp
Total Beta	11	8.76	1	2.0 sp
Bi.icp.w	11	10.34	1	2.2 sp
Bi.icp.wo	7	9.96	1	2.2 sp
Ca.icp.w	11	12.91	1	13.2 sp
Ca.icp.wo	8	13.00	1	13.2 sp
Caustic	3	9.68	1	2.8 rs

* = Number of samples with duplicate measurements
$ = Number of analytical results from which the systematic variability was estimated
.w = Less than values were used in the statistical analysis at the upper value (e.g., 3 for <3)
.wo = Less than values were deleted from the statistical analysis
sp = Systematic variability estimate calculated from spike analyses
rs = Systematic variability estimate calculated from reference standards analyses
ct = Systematic variability estimated calculated from counting statistics

Table F-1. Random/Systematic Analytical Variability Estimates. (Page 2 of 3)

Analyte	Random Variability Estimated from the duplicate sample results		Systematic Variability Estimated from Spike or Standard Analyses	
	N *	RSD (%)	N $	RSD (%)
Cd.icp.w	11	19.02	1	8.2 sp
Cd.icp.wo	9	19.15	1	8.2 sp
Cl^-.ic	5	3.13	1	3.4 sp
Cr.icp.w	11	9.82	1	15.8 sp
Cr.icp.wo	5	8.22	1	15.8 sp
^{137}Cs.gea	11	16.22	5	1.1 rs
Cu.icp	11	5.58	1	8.0 sp
^{154}Eu.gea.w	11	18.32	No Std	25.0 ct
^{154}Eu.gea.wo	9	8.99	No Std	25.0 ct
Fe.icp	11	36.08	1	12.8 rs
Mg.icp.w	11	9.51	1	1.0 sp
Mg.icp.wo	6	9.33	1	1.0 sp
Mn.icp.w	11	10.04	1	14.2 sp
Mn.icp.wo	5	9.46	1	14.2 sp
Na.icp	11	0.23	1	20.8 sp
Ni.icp.w	11	7.86	1	4.6 sp
Ni.icp.wo	5	9.81	1	4.6 sp
^{237}Np.w	11	13.99	1	22.3 sp
^{237}Np.wo	6	7.69	1	22.3 sp
^{238}Pu.w	11	15.64	No Std	5.0 ct
^{238}Pu.wo	10	9.67	No Std	5.0 ct

* = Number of samples with duplicate measurements
$ = Number of analytical results from which the systematic variability was estimated
.w = Less than values were used in the statistical analysis at the upper value (e.g., 3 for <3)
.wo = Less than values were deleted from the statistical analysis
sp = Systematic variability estimate calculated from spike analyses
rs = Systematic variability estimate calculated from reference standards analyses
ct = Systematic variability estimated calculated from counting statistics

Table F-1. Random/Systematic Analytical Variability Estimates. (Page 3 of 3)

	Random Variability		Systematic Variability	
	Estimated from the duplicate sample results		Estimated from Spike or Standard Analyses	
Analyte	N *	RSD (%)	N $	RSD (%)
$^{239/240}$Pu	11	6.04	1	1.3 sp
SO_4^{2-}.ic.w	5	5.75	1	2.1 sp
SO_4^{2-}.ic.wo	4	5.78	1	2.1 sp
$^{89/90}$Sr	11	4.55	1	14.8 sp
Total Carbon	12	2.62	5	5.8 sp
TIC	12	4.38	7	2.8 sp
TOC	12	14.68	7	11.6 sp
^{99}Tc	4	10.69	2	2.7 rs
U.icp.w	11	3.21	1	13.8 rs
U.icp.wo	10	3.11	1	13.8 rs
U.phos	11	7.87	1	12.6 sp
U.las	7	8.41	4	2.4 rs
^{234}U.tims	7	21.65	3	7.3 rs
^{235}U.tims	7	1.10	3	0.90 rs
^{236}U.tims	7	2.06	3	0.06 rs
^{238}U.tims	7	0.01	3	0.005 rs
%Water.grav	7	1.88	3	4.0 rs
Zn.icp.w	11	21.01	1	11.0 sp
Zn.icp.wo	8	21.34	1	11.0 sp
Zr.icp.w	11	16.74	1	4.8 sp
Zr.icp.wo	6	15.48	1	4.8 sp
pH	7	0.53	3	0.4 rs

* = Number of samples with duplicate measurements
$ = Number of analytical results from which the systematic variability was estimated
.w = Less than values were used in the statistical analysis at the upper value (e.g., 3 for <3)
.wo = Less than values were deleted from the statistical analysis
sp = Systematic variability estimate calculated from spike analyses
rs = Systematic variability estimate calculated from reference standards analyses
ct = Systematic variability estimated calculated from counting statistics

The systematic analytical variability (RSD) is estimated by the larger of either (1) the deviation of the mean percent recovery from 100 or (2) the standard deviation of the percent recovery values for the analyte of interest divided by the square root of n (the number of percent recovery values for the analyte of interest).

The estimates of the systematic analytical variability based on either the laboratory standards percent recovery or the spiked samples percent recovery are provided in Table F-1. The number of spike analyses performed and the number of laboratory standards analyzed is also listed in Table F-1. These systematic analytical variability estimates do NOT include any of the uncertainties associated with any of the steps prior to the actual laboratory (222-S or PNNL) analysis of the samples.

Comparison of the Uranium Analyses

The uranium analyses were performed using four different analytical measurement methods: phosphorescence by 222-S Laboratory, fluorescence by PNNL, inductively coupled plasma (icp) by 222-S Laboratory, and inductively coupled plasma/mass spectrometry (icp/ms) by PNNL. The data, by sludge sample, are listed in Table F-2 and illustrated in Figure F-1. The uranium results, as measured by 222-S Laboratory, for sludge Sample 96-01 are quite a bit smaller than all the other uranium results.

To evaluate the analytical results, the data for each sludge sample were pooled and the summary statistics computed. The summary statistics are listed in Table F-3. The pooled standard deviation incorporates variability due to the analytical procedure and the subsampling variability. Nine of the thirteen sludge samples have an RSD less than 25%. Only two sludge samples (96-01 and 96-09) had large (> 100%) RSDs.

Table F-2. Uranium Sludge Characterization Data.

	U (222-S) phosphorescence	U-PNNL fluorescence	U (222-S) icp	U-PNNL icp/ms Accelerated Sample	U-PNNL fluorescence Accelerated sample
Sludge Sample	µg/g	µg/g	µg/g	µg/g	µg/g
96-01	944	824000	< 257	650000	854000
96-05	585000	881000	748000	643000	664000
96-08	310000	404000	385000	NA	NA
96-09	131000	881000	131000	NA	NA
96-13	740000	820000	747000	NA	NA
96-15	492000	813000	668000	NA	NA
96-04 U	297000	NA	318000	NA	NA
96-04 L	273000	546000	318000	NA	NA
96-06 U	NA	NA	NA	NA	NA
96-06 M	611000	826000	749000	NA	NA
96-06 L	606000	843000	739000	NA	NA
96-11 U	128000	NA	133000	NA	NA
96-11 L	73300	93300	77900	NA	NA

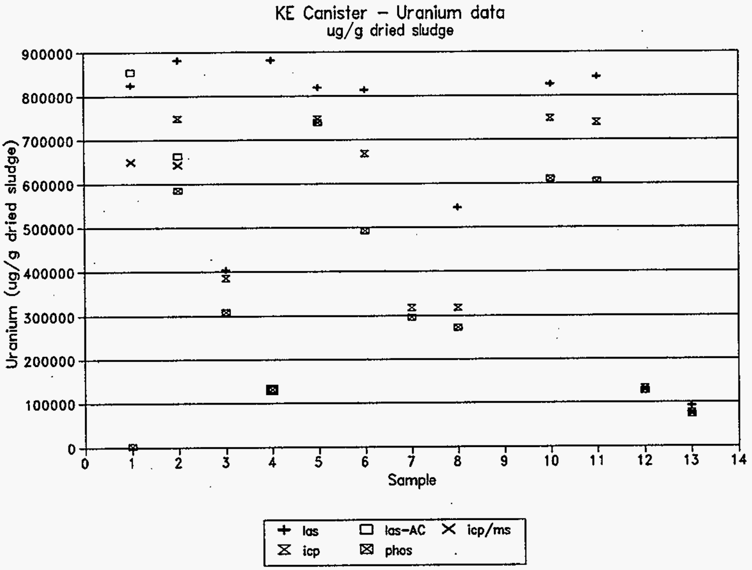
µg/g: µg/g dried sludge
NA: Not analyzed

Table F-3. Uranium Concentration by Sludge Sample.

	Mean Concentration	Standard Deviation	RSD (%)
Sludge Sample	µg/g	µg/g	µg/g
96-01 *	466000 (776000)	432000 (110000)	92.7 (14.2)
96-05	704000	115000	16.3
96-08	366000	49700	13.6
96-09	381000	433000	113.7
96-13	769000	44300	5.8
96-15	658000	161000	24.4
96-04 U	308000	14800	4.8
96-04 L	379000	146000	38.6
96-06 U	NA	NA	NA
96-06 M	729000	109000	15.0
96-06 L	729000	119000	16.3
96-11 U	131000	3540	2.7
96-11 L	81500	10500	12.9

*: The values listed in the () have the 222-S Laboratory data deleted.

Figure F-1. KE Canister Sludge Uranium Concentration Data.



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APPENDIX G

TEST PLAN FOR POLYCHLORINATED BIPHENYL ANALYSES

(A Comparison Between Environment Protection Agency
and Currently Utilized Methods)

E. W. Hoppe

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APPENDIX G

TEST PLAN FOR POLYCHLORINATED BIPHENYL ANALYSES

G.1 SUMMARY

The Pacific Northwest National Laboratory (PNNL) Organic Analysis Laboratories approach to analysis of PCBs in K Basin sludge and supernatant samples is based upon existing USEPA methodologies. The only modifications necessary are due to the amount of sample available; consequently, the procedure is scaled down. The modifications reduce the initial sample size and proportionately lower the amount of surrogate or spike compounds to be added. This reduction matches the concentrations in the sample to those in the USEPA procedures. To match the concentration of these compounds in the final extract residue, this volume is reduced as well. The PNNL approach uses the same advisory limits for surrogate and spike recoveries and precision as specified in the USEPA procedures.

G.2 EXTRACTION OF WATER SAMPLES

USEPA Contract Laboratory Program (CLP) or SW-846 Method 3510 procedures typically begin with nominally 1 liter of sample added to a separatory funnel. This step is followed by the addition of surrogate (tetrachloro-m-xylene, TCX, and decachlorobiphenyl, DCB) or spike compounds (e.g., Aroclor 1254 in this case). The surrogates are added to achieve a concentration in the sample of 1 to 5 μg/L. For the canister sludge supernate water samples, approximately 100-ml of supernatant sample will be available for each sample. Nominally, 100 ml of sample will be added to a separatory funnel, and one-tenth the quantity of surrogate or spike compound that would be added to a liter sample will be used. As in the USEPA methods, the concentration of surrogates in the 100-ml sample will be 1 μg/L. The spike will be added at a concentration of 1 μg/L of Aroclor 1254. This spike level is near the nominal detection limit for the gas chromatography/electron capture detector (GC/ECD) analysis, but less than the quantification level required for this testing (i.e., 3 μg/L). Aroclor 1254 is chosen based on results from the previous analysis of K East Basin sludge samples.

As in the USEPA methods, methylene chloride will be used as the partitioning solvent for three extractions. The methylene chloride extracts will be combined, concentrated, and exchanged into hexane. The final volume of the residue will be reduced from 10 ml specified in the USEPA procedures to 1 ml for the supernatant samples, which will result in the same concentration factor as the USEPA procedures. The samples will then be ready for analysis by GC/ECD.

G.3 EXTRACTION OF SLUDGE SAMPLES

In the previous K Basin sludge sample set, sample inhomogeneity was the greatest source of variability in analytical results. Extraction was performed on approximate 1 g samples. Normally, sample size can be increased to reduce these effects. However, because of limited sample availability and radiological concerns, sample size cannot be increased and inhomogeneity concerns cannot be reduced in this manner. To improve sample homogeneity where possible, the canister samples will be thoroughly mixed before obtaining discrete sample aliquots. Extractions will be performed in radiological hoods and glove boxes to the extent possible to reduce the complications associated with extractions performed in the remote handling facilities.

Ultrasonic extraction has been chosen as the extraction method for the K Basin sludge samples. It is known that very non-polar species, such as PCBs, may not extract well from organic materials using this method, however, it will be appropriate here since the matrix to be analyzed is not organic. Although the K Basin matrix is not organic material, the sample will be solvent contacted and sonicated three times rather than once as is done in the USEPA CLP or SW-846 3550 medium level methods to ensure adequate extraction. Once again, this set of samples will be extracted employing a slightly reduced sample size of 1 g rather than 2 g in the USEPA SW-846 3550 method. The USEPA CLP methods specify 1 g in the 1988 SOW, however, no provision is made for medium level extractions in the 1991 SOW. Sodium sulfate is added to the sample as a drying agent followed by the addition of 1 μg of each of the surrogate compounds, TCX and DCB.

Samples designated for spiking will have 1 μg of Aroclor 1254 added. The samples will be extracted using a methylene chloride/acetone (1:1) solvent mixture and subjected to high intensity sonication. The solvent will be removed and the extraction process repeated twice more. The resulting extracts will be combined and concentrated to 10 ml. From the 10-ml extract, 1 ml will be removed and exchanged into hexane. This will be used for GC/ECD analysis. The remaining 9 ml of solvent will be reduced to 0.9 ml for confirmation by gas chromatography/mass spectroscopy (GC/MS) if needed.

G.4 ANALYSIS

As with the USEPA methods, analysis will be performed using a dual capillary gas chromatograph and dual electron capture detector instrument. This allows for simultaneous analysis and confirmation of the sample residue. The method identifies and determines the concentrations of PCBs as Aroclor mixtures by direct comparison with the instrument calibration using authentic Aroclor mixtures. USEPA procedures demonstrate linearity using different pesticide compounds followed by a single-point calibration of Aroclors. For these analyses, a multipoint calibration for Aroclor 1254 will be performed rather than for the various pesticides on the GC/ECD. In addition to the GC/ECD analysis, GC/MS confirmation will be used. The GC/MS confirmation will only be employed if Aroclors are detected in sufficient concentration by the GC/ECD, in this case 10 ng/μL in the final extract. A multipoint calibration of the GC/MS will be performed for Aroclor 1254. Since GC/MS is less susceptible to interferences, it is anticipated to provide results that are more accurate than those produced from the GC/ECD instrument.

G.5 REFERENCES

Battelle, Analytical Chemistry Laboratory (ACL) Procedure Compendium, Methods PNL-ALO-120, 345, 346, 347. Analytical method for extraction and analysis of Pesticides/PCBs and Semivolatile compounds.

SW-846, Test Methods for Evaluating Solid Waste, Physical and Chemical Methods, Method 8082, Polychlorinated Biphenyls (PCBs) by Capillary Column Gas Chromatography, Rev. 0, January 1995.

USEPA Contract Laboratory Program, Statement of Work of Organic Analysis, Multi-Media, Multi-Concentration, 1988.

USEPA Contract Laboratory Program, Statement of Work of Organic Analysis, Multi-Media, Multi-Concentration, OLM01.8, August 1991.

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APPENDIX H

SUMMARY OF POLYCHLORINATED BIPHENYL RESULTS

E. W. Hoppe and A. J. Schmidt

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APPENDIX H

SUMMARY OF POLYCHLORINATED BIPHENYL RESULTS

H.1 OVERVIEW

All nine sludge samples collected from K East Basin fuel canisters and two supernatant water decant samples from the canisters were analyzed for polychlorinated biphenyls (PCBs) by the Pacific Northwest National Laboratory (PNNL). The PCB analyses were performed using a dual capillary gas chromatograph and dual electron capture detector (GC/ECD) instrument. Confirmation analyses were performed on select samples using gas chromatography/mass spectroscopy (GC/MS). The approaches used to extract and analyze for PCBs in sludge and supernatant samples were based on existing U.S. EPA methodologies.

The canister sludge samples were analyzed twice (approximately 30 days apart), and two data sets were generated. During the initial set of analyses (reported in Silvers 1997a), Aroclor 1254 was identified in all of the samples. Additionally, late eluting peaks were tentatively identified as Aroclor 1268 and, potentially, Aroclor 1262 in the majority of the sludge samples. Since the presence of Aroclor 1262 and 1268 was unexpected, calibration using acceptable reference standards was not possible. Only Aroclor 1254 was quantified in the first data set. Additionally, discrepancies in the individual PCB congener profiles between the samples and the best available (but out-of-date) Aroclor 1262 and 1268 standards resulted in their identification as being only tentative.

For the second analysis (Silvers 1997b), the sample extracts were further concentrated to verify the amount of Aroclor 1254 present, and current standards were obtained for more detailed examination of the tentatively identified higher Aroclors 1262 and 1268. While Aroclor 1254 was readily requantified, examination of the data generated from the reanalysis again revealed discrepancies of the late eluting peaks to the expected congener profiles. To resolve these issues, a select number of the sample extracts (those exhibiting higher concentrations of the late eluting peaks in question) were further concentrated and then analyzed using GC/MS. The late eluting peaks did not match the characteristic "fingerprint" mass spectra of PCBs, nor did the GC/MS data identify any chlorinated species within the late eluting peaks. Several of the late eluting peaks were identified as phthalates and adipates. [Within the EPA protocol, phthalates are specifically listed as compounds that can potentially interfere with PCB analysis by GC/ECD. Also, phthalates were identified as being present in the K East Basin Floor and Weasel Pit samples (Makenas 1996c).] Therefore, based on the discrepancies in the congener profiles obtained with GC/ECD analyses and the GC/MS results, it has been concluded that the late eluting peaks are not PCBs.

The results from the initial Aroclor 1254 analyses agreed fairly well with the results from the second, or reanalysis data set. The relative percent difference (RPD) between individual samples from the two data sets ranged from 0 to 76%. The results obtained from the reanalysis are lower than the data from the initial analyses. The reanalysis results are lower for several reasons. Some effect may be from the decreased signal to noise in the reanalysis because the extracts were further concentrated by a factor of four. However, because the surrogate recoveries were also lower in the reanalysis data, which would have been less effected by extract concentration changes. The greatest impact is likely due to additional storage and handling of the extracts prior to reanalysis.

The range of Aroclor 1254 concentration from both data sets was 11 to 1100 ppb (0.011 to 1.1 ppm). Based on these data sets, the canister sludge does not meet the definition of a regulated Toxic Substance Control Act (TSCA) waste (i.e., the canister sludge contains <50 ppm PCB). Furthermore, the level of PCB found in the canister sludge is less than the TSCA PCB treatment standard, which is 2 ppm per PCB congener.

No Aroclors were detected in either of the two canister water decant samples (i.e., canister decant water contained <0.10 μg/L PCB). The PCB content in the canister decant water is below the level at which the water could potentially be subject to regulation under TSCA (i.e., it is less than 3 ppb).

The results of the PCB analyses are summarized in Table H1 (canister sludge) and Table H2 (canister decant water). Table H1 also includes information on the number of fuel elements in the canisters from which the sludge samples were taken. The number of fuel elements present (or the lack of fuel elements altogether) does not appear to be correlated with the PCB concentration in the sludge. In both Tables H1 and H2 information is provided on the results of quality indicator sample analyses.

H.2 SAMPLE HANDLING AND PREPARATION

The extractions of PCBs from canister sludge and decant water were performed in accordance with PNL-ALO-347, Rev. 1, "Sample Preparations for Pesticides/PCBs Analysis in Water and Soil/Sediment" (with test plan modifications; see Appendix G), which is based on existing U.S. EPA methodologies. The only modifications necessary were to scale down the procedures proportionate to the amount of sample available due to sample availability and radiological considerations. Because sample sizes were reduced, the amount of surrogates or spike compounds were proportionately reduced to match those specified in the U.S. EPA procedures.

H.2.1 Canister Sludge Samples

Before subsample aliquots were collected for extraction and PCB analysis, the canister sludge was homogenized in accordance with Procedure PNL-ALO-135, "Procedure for Laboratory Homogenization of Solutions, Slurries, and Sludges (August 3, 1989)." From each canister, approximately 200 to 400 ml of settled sludge was subjected to homogenization.

Table H1. Polychlorinated Biphenyls in K East Basin Canister Sludge Samples Determined by Gas Chromatograph and Dual Electron Capture Detector (concentrations on settled sludge basis).

Sample ID	Comments	Initial Analysis (Silvers 1997a)				Reanalysis (Silvers 1997b)			RPD Between Initial and Reanalyses[3]
		ppb of Aroclor 1254 (µg/kg)	% Spike Recovery	RPD[1]	Aroclor 1268 Detected by GC/ECD ?[2]	ppb of Aroclor 1254 (µg/kg)	% Spike Recovery	RPD[1]	
Canister Sludge Samples									
96-01 96-01 Duplicate	Canister barrel contained 5 elements, sludge depth = 2.33 in.	140 130	NA	7.4	No No	120 120	NA	0	15 8.0
96-04* 96-04 Duplicate*	Canister contained 7 elements, sludge depth = 1.70 in.	180 110	NA	48	Yes Yes	99 110	NA	11	58 0
96-05 96-05 Duplicate	Canister contained 6 elements, sludge depth = 3.34 in.	120 160	NA	29	Yes Yes	92 72	NA	24	26 76
96-06	Canister contained 6 elements, sludge depth = 8.55 in.	<80	NA	NA	Yes	13	NA	NA	NA
96-08	Canister contained 7 elements, sludge depth = 3.44 in.	750	NA	NA	Yes	560	NA	NA	29
96-09	Canister contained 0 elements, sludge depth = 0.38 in.	1110	NA	NA	Yes	880	NA	NA	23
96-11 96-11 Duplicate	Canister contained 0 elements, sludge depth = 0.53 in.	170 330	NA	64	Yes Yes	100 170	NA	52	52 64
96-13 96-13 Duplicate	Canister contained 6 elements, sludge depth = 5.66 in.	<50 <70	NA	NA	Yes Yes	11 27	NA	84	NA
96-15*	Canister contained 7 elements, sludge depth > 12.13 in.	<60	NA	NA	Yes	24	NA	NA	NA
Canister Sludge Data Quality Indicator Samples									
96-04 MS 96-04 MSD	Aroclor 1254 matrix spike Aroclor 1254 matrix spike dup	880[4] 870[4]	92 95	3.2	Yes Yes	500[4] 770[4]	49 88	57	55 12
96-05 MS 96-05 MSD	Aroclor 1254 matrix spike Aroclor 1254 matrix spike dup	610[4] 970[4]	103 107	3.8	Yes Yes	480[4] 840[4]	87 98	12	24 14
Method Blank*		<100	NA	NA	No	<10	NA	NA	NA
Method Blank 2*		<100	NA	NA	No	<10	NA	NA	NA

[1] Relative percent difference between duplicates.

[2] Presence not confirmed by GC/MS.

[3] Example: For Sample 96-01, RPD between sets = (140 - 120)/0.5(140 + 120) = 15.

[4] Sum of PCB in sample + PCB from Aroclor 1254 spike.

*Denotes sample was subjected to GC/MS confirmation analysis. No PCB congeners were detected other than the DCB surrogate (GC/MS PCB congener detection limit was ~500 ppb).

Table H2. Polychlorinated Biphenyls in K East Basin Canister Water Decant Samples Determined by Gas Chromatograph and Dual Electron Capture Detector.

Sample ID	Comments	Aroclor 1254 (µg/L)[1]	Aroclor 1262 (µg/L)	Aroclor 1268 (µg/L)	Total PCB (µg/L)[2]
Canister Water Decant Samples					
96-01 Cyl	Canister contained 5 elements, sludge depth = 2.33 in.[3]	ND[4]	ND	ND	<0.1
96-05 Cyl 96-05 Cyl Duplicate	Canister contained 6 elements, sludge depth = 3.34 in.	ND ND	ND ND	ND ND	<0.1[5] <0.1[5]
Canister Water Data Quality Indicator Samples					
96-05 Cyl MS	Aroclor 1254 spike, 42% recovery	0.56[6]	ND	ND	0.56[6]
96-05 Cyl, MSD	Aroclor 1254 spike, 75% recovery	1.00[6]	ND	ND	1.00[6]
Method blank		ND	ND	ND	<0.1
Method blank matrix spike	124% recovery of Aroclor 1254 matrix spike	1.20	ND	ND	1.20
Filter blank		ND	ND	ND	<0.1
Filter blank matrix spike	5.6% recovery of Aroclor 1254 matrix spike	0.06	ND	ND	0.06
Laboratory Control Standard	1254, U.S. EPA WP036 #1, 48% of true value, acceptable range (0.627 -2.59 µg/L)	0.85	ND	ND	0.85

[1] µg/L = Microgram per liter, which is approximately equivalent to parts per billion.

[2] Total PCB is sum of Aroclor 1254 + Aroclor 1262 + Aroclor 1268.

[3] Number of fuel elements contained in canister barrel sampled, and depth of sludge measured in that canister barrel.

[4] ND = Not detected.

[5] Samples 96-05 Cyl and 96-05 Cyl Duplicate were filtered. Spike recovery of filter blank was only 5.6% - therefore, actual detection limit may be as high as 1.8 µg/L.

[6] Value is sum of PCB in sample + PCB from Aroclor 1254 spike.

Canister sludge sample aliquots ranged from 1 to 2.2 g. Consequently, some of the samples were extracted using a slightly reduced sample size rather than the 2 g recommended in the U.S. EPA SW-846 3550 Method for medium level PCB concentrations. Sodium sulfate was added to the samples as a drying agent, followed by the addition of 1 µg of each of the surrogate compounds, [tetrachloro-m-xylene (TCX) and decachlorobiphenyl (DCB)]. [Note: DCB is a PCB congener in which all ten available sites on the biphenyl molecule are occupied by chlorine atoms (i.e., DCB is a fully chlorinated PCB).] For samples designated for spiking, 1 µg of Aroclor 1254 was added. Ultrasonic extraction (PNL-ALO-347, Rev. 1, with test plan modifications) was used as the extraction method for the K East Basin canister sludge samples. The samples were solvent contacted [methylene chloride/acetone (1:1)] and sonicated three times.

The extracts resulting from the three sonication extractions were combined and concentrated to 10 ml. From each 10-ml extract, 2 ml were removed and exchanged into hexane and used for the initial set of canister sludge PCB analyses via GC/ECD. The remaining 8 ml of extract were later concentrated to 2 ml, exchanged into hexane, and used for the second set of canister sludge PCB analyses (GC/ECD), which was performed approximately 35 days after the initial extraction. The hold time recommendation from SW-846 for soil/sediments and sludges for Semivolatile Organochlorine Pesticides/PCBs and Herbicides is that the extracts be analyzed within 40 days of initial extraction.

Following the second set of GC/ECD analyses, 1.5 ml of the remaining extracts from select samples were concentrated to 50 µL (0.05 ml) and exchanged back to methylene chloride for confirmation analyses using GC/MS.

H.2.2 Canister Water Decant Samples

The K East Basin water samples were extracted in the PNNL shielded analytical (SAL) facility using solvent partition with methylene chloride in a separatory funnel. The solvent extracts were then concentrated in a radiological fume hood.

U.S. EPA Contract Laboratory Program or SW-846 Method 3510 procedures typically begin with 1 liter of sample being added to a separatory funnel. Next, surrogates (TCX and DCB) or spike compounds (Aroclor 1254 in this case) are added to the water sample. The surrogates are added to achieve a concentration in the sample of 1 to 5 µg/L. For K East Basin canister water decant, sample sizes were limited to 73 to 100 ml.

For these analyses, nominally, 100 ml of canister decant sample were added to a separatory funnel and one-tenth the quantity of surrogate or spike compounds that would be added to a liter sample was used. As in the U.S. EPA methods, the concentration of surrogates in the 100-ml sample was 1 µg/L. The spike was added at a concentration of 1 µg/L of Aroclor 1254. This spike level is near the nominal detection limit for the GC/ECD analysis, and Aroclor 1254 was chosen based on results from the analysis of K East Basin Floor and Weasel Pit sludge (Makenas 1996).

As with the U.S. EPA methods, methylene chloride was used as the partitioning solvent for three successive extractions. The methylene chloride extracts were combined, concentrated, and exchanged into hexane. The final volume of the residue was reduced from 10 ml specified in the U.S. EPA procedures to 1 ml for K East Basin samples. Thus, the final 1-ml extract was at the same concentration factor as recommended in the U.S. EPA procedures.

The canister water decant samples were sufficiently radioactive to limit their handling to the SAL. It was initially believed that the radioactivity was due to a small quantity of sediment present in the sample containers. An attempt was made to filter this material out of the sample so that the extraction could be performed in a radiological fume hood. A filter blank consisting of the same blank water used for the method blank was filtered through a glass-fiber Tuffryn™ filter. The eluate was collected for extraction and analysis. Blank water was also spiked with Aroclor 1254, a portion of which was reserved for extraction and analysis. Another portion of the spiked blank was filtered and the eluate collected for extraction and analysis. Samples 96-05 Cyl and 96-05 Cyl Duplicate were filtered, and the eluates collected. The results from the filtering tests are discussed later in this text. Because an insufficient reduction in radioactivity was observed, extraction of the water decant samples in the radiological fume hood was not possible. Consequently, the remaining sample, 96-01 Cyl, was not filtered.

H.3 SAMPLE ANALYSIS

Sludge and water samples were analyzed in accordance with PNL-ALO-346, Rev. 0, "Analysis for Pesticides/PCBs by Gas Chromatography with Electron Capture Detection," which is consistent with U.S. EPA methods. This method uses a dual capillary gas chromatograph and dual electron capture detector (GC/ECD) instrument (HP-5890II dual on-column injection, with DB-17 and DB-1701 columns). This setup allows for simultaneous analysis and confirmation of the sample residue. The method identifies and determines the concentrations of PCBs as Aroclor mixtures by direct comparison with the instrument calibration using Aroclor standards. The U.S. EPA procedures demonstrate linearity using several pesticide compounds, followed by a single-point calibration of Aroclors. The initial analysis of these samples incorporated a six-point calibration of the GC/ECD for Aroclor 1254 and the surrogates (TCX and DCB). Single point calibrations were performed with Aroclors 1016, 1221, 1238, 1242, and 1248. For the reanalyses, six-point calibrations for Aroclors 1254, 1260, 1262, 1268 and the surrogates (TCX and DCB) were performed on the GC/ECD.

Confirmation analyses on select sludge samples were performed in accordance with PNL-ALO-345, Rev. 1, using an HP-5890II/HP5972A GC/MS and splitless injection onto a DB-5 column. The GC/MS confirmation analyses on sludge extracts were performed after meeting tuning criteria for mass spectral abundances of decafluorotriphenylphosphine (DFTPP) specified in the procedure. No continuing calibration standard was analyzed. No spiking was performed other than the addition of internal standards at 40 μg/ml.

H.4 DISCUSSION

H.4.1 Canister Sludge Samples

Canister sludge samples were analyzed twice using GC/ECD, and two data sets were generated. In the initial data set, Aroclor 1254 was identified in all of the sludge samples, and late eluting peaks indicated the presence of higher molecular weight material. With the use of available out-of-date U.S. EPA Aroclor standards (obtained in 1990), reference chromatograms for Aroclors 1262 and 1268 were produced and used to tentatively identify them within the canister sludge. Discrepancies were seen in the individual PCB congener profiles between the samples and these Aroclors 1262 and 1268 standards. No attempt was made to perform quantifications using the out-of-date standards.

For the second set of canister sludge analyses (i.e., reanalyses), current Aroclors 1262 and 1268 standards were obtained, and used in the initial six-point calibration before reanalysis. Congeners for calibration were carefully chosen that were indicative of specific Aroclors or showed minimal cross contributions with other Aroclors. To further improve sensitivity (i.e., to lower detection and quantification limits), the remaining 8 ml of sludge extract residues were further concentrated and then analyzed by the GC/ECD. As in the initial analysis, Aroclor 1254 was easily identifiable. In addition, many of the chromatograms again exhibited late eluting material. The pattern for these materials resembled Aroclor 1268, or slightly degraded Aroclor 1268. However, unlike the initial analysis, due to the selection of non-cross contributing congeners, the ratio bias of Aroclor 1254 congeners was not observed. This resulted in better quantitation for Aroclor 1254, particularly for those samples where the amount of Aroclor 1254 present was similar to the quantity of late eluting material.

As expected, the reanalysis observed the same late eluting materials tentatively identified as higher Aroclors in the initial set of analyses. While the pattern and retention times for the late eluting material matched the Aroclor 1268 standard, the congener ratios were substantially different from the Aroclor 1268 standard. In addition, low levels of the late eluting material were observed in the method blanks, potentially indicative of interfering contaminants other than PCBs (i.e., phthalates).

Although the confirmation column results were consistent with the primary column results, additional analysis was deemed necessary to determine whether the sample peaks in question were generated from degraded Aroclors or were the result of interfering compounds. [Note: PNL-ALO-346, Rev. 0, "Analysis for Pesticides/PCBs by Gas Chromatography with Electron Capture Detection," analyzes each sample on two columns, one being a confirmation column.] To resolve this uncertainty, remaining sample extracts were prepared as described previously and analyzed using GC/MS.

Aliquots of the remaining sample extracts were further concentrated to obtain sufficient response from the GC/MS. Verification of response was provided by tracking the DCB surrogate during the GC/MS analysis. The DCB surrogate, which is a fully chlorinated PCB congener, was spiked (at a maximum concentration of 1 ppm) into the canister sludge samples prior to the initial extraction. The GC/MS analysis detected the DCB surrogate and generated full isotopic spectra; however, no other chlorinated species were identified late in the GC/MS chromatogram. Based on this information and that the GC/MS response factors for similar PCB congeners do not vary widely, it can be concluded that there are no late eluting individual PCB congeners present at a concentration greater than the maximum DCB surrogate concentration of 1 ppm.

The GC/MS was expected to detect the late eluting peaks since the GC/ECD response to the materials was as much as 50% of that of the DCB surrogate. As expected, several late eluting species were identified by GC/MS as phthalates and adipates which could give a response on the GC/ECD similar to that of Aroclors. The peak shape of these compounds on the GC/MS was sharp and similar to the GC/ECD chromatograms.

Because the late eluting material was not identified by GC/MS as PCB or any chlorinated species, and the GC/ECD indicated the congener ratios were different from the standard, the late eluting material cannot be identified as Aroclor 1268. If the late eluting material contained any higher PCB congeners below the GC/MS detection limit, then they would have to be present at levels much lower than the DCB surrogate (1 ppm). The conclusion drawn from the GC/MS analysis is that if higher Aroclors are present, they are at concentrations below regulatory significance (i.e., <2 ppm per PCB congener).

Comparison of the quantitative data (Table H1) for Aroclor 1254 from the two data sets shows some minor differences. The values for Aroclor 1254 from the reanalysis tend to be slightly lower than those obtained in the initial analyses. These differences may be attributed to two factors: (1) the additional extract storage period and handling from further concentration of the extract may have resulted in minor losses of Aroclor 1254 in the reanalysis (indicated by the lower spike and surrogate recoveries), (2) some late eluting mass was assigned as Aroclor 1254 in the first data set, potentially inflating Aroclor 1254 concentrations. This potential bias was minimized during quantitation of Aroclor 1254 in the reanalysis. Furthermore, for the reanalysis, lower detection and quantification limits were achieved since the sample extracts were subjected to additional concentration.

Both sets of analyses show the canister sludge does not meet the definition of TSCA waste (i.e., the canister sludge contains <50 ppm PCB). Furthermore, the level of PCB found in the canister sludge is less than the TSCA PCB treatment standard, which is 2 ppm per PCB congener.

H.4.2 Canister Water Decant Samples

No Aroclors were detected in either of the two canister water decant samples (i.e., canister decant water contained <0.10 µg/L PCB). The PCB content in the canister decant water is below the level at which the water could be potentially subject to regulation under TSCA (i.e., it is less than 3 ppb).

The water decant samples were analyzed following extraction in the SAL. Samples 96-05 Cyl and 96-05 Cyl Duplicate were filtered along with a filter blank and filter blank matrix spike. Analysis of the filtered blanks indicated no analytical interferences were leached into the water from the filter material. However, the filtrate showed substantial loss of Aroclor 1254 that was spiked into the blank water, which is exhibited by the blank spike water recovery of 124% for Aroclor 1254 and the filtered blank spike showing a recovery of only 5.6%. Due to the filter recovery issue, the detection limit for Samples 96-05 Cyl and 96-05 Cyl Duplicate is biased low by as much as 18 times. With this potential bias, the PCB detection limit for these samples may be as high as 1.8 µg/L, which is still below the TSCA limit of 3 ppb. Sample 96-01 Cyl, was not filtered and, therefore, not affected by filter recovery issues.

H.5 QUALITY CONTROL

Modifications were made to the extraction and analytical procedures to allow for a more pertinent calibration and analysis for PCBs as Aroclor mixtures. The extraction portion of the procedure was scaled down appropriately to account for radiological issues and limited sample volume availability. In addition, rather than using pesticides for determining instrument linearity, Aroclors were used for multipoint calibrations. The continuing calibration also consisted of using Aroclor 1254. Quantification was performed using a primary column, and the secondary column was used for confirming identification.

For quality assurance purposes, a number of standard measures were taken during the sample preparation and analyses. Method blank analyses were prepared and analyzed in parallel with the sludge and decant water samples. Two sets of matrix spikes and matrix spike duplicates were prepared and analyzed with the sludge samples. One matrix spike and matrix spike duplicate was prepared and analyzed with the canister water decant samples. Additionally, more than half the samples were prepared and analyzed in duplicate. Table H3 summarizes the quality control criteria and shows how the data sets performed against these criteria.

All samples, blanks, matrix spikes, and matrix spike duplicates were spiked with the surrogate spike compounds TCX and DCB. No control limits have been established for these compounds on these matrices; however, surrogate recoveries were judged acceptable. Acceptance of these surrogate recoveries is further supported by use of the U.S. EPA advisory limits set at 20% to 150%. During reanalysis of the sludge samples, one sludge sample, 94-04 MSD, exhibited a surrogate (DCB) recovery outside of the advisory limits. Table H4 presents a summary of surrogate recoveries for all samples.

H.5.1 Statistical Evaluation of Canister Sludge Polychlorinated Biphenyls Data

The summary statistics for the PCB results from the K East Canister sludge analyses sets are provided in Table H5. The differences observed between the analyses sets (initial analysis and reanalysis) are statistically

Table H3. Quality Assurance Criteria and Indicators Performance for Gas Chromatograph and Dual Electron Capture Detector.

QC Parameter	Comments
Target Compounds	Detection levels are based on sample size and the determination of instrument detection limits (IDLs) from multiple injections of a low standard. Met requirements.
Surrogate Recoveries Sludge - Data Set 1 TCX 62-120% DCB 49-101% Sludge - Data Set 2 TCX 37-90% DCB 9.5-85% Water TCX 40-83% DCB 26-77%	Surrogate spiking was performed on all samples using TCX and DCB. No control limits have been established for these compounds on these matrices. A summary of all surrogate recoveries is shown in Table H4. Surrogate recoveries were judged acceptable for all but one sample.
Matrix Spike Recovery Sludge Data Set 1[1] Aroclor 1254 92-107% Sludge Data Set 2[2] Aroclor 1254 49-98% Water Aroclor 1254 42-124%	Spiking was performed at an adequate frequency. Two sets of spikes and duplicates were performed for the sludge samples, and one set and an LCS was performed on the water samples. No control limits have been established for these compounds on these matrices. All spike recoveries and RPDs are listed in Tables H1, H2, and H3. Spike recoveries were judged acceptable.
Laboratory Control Standard (LCS) Check	U.S. EPA WP036 #1 analyzed as a blind water sample. 0.85 µg/L or 48% of true value obtained, acceptable range (0.627 - 2.59 µg/L).
Method Blank Summary	Method blanks were extracted with each batch of samples. Met requirements.
Initial Detection Limit Determination	Multiple analysis of standard <10 x IDL. Met requirements.
Initial Calibration	Met requirements. Calibration for Data Set 1 was performed with Aroclor 1254 and surrogates. Calibration for Data Set 2 was performed using Aroclors 1254, 1262, 1268, and surrogates. Met requirements
Carry-Over Evaluation	Met requirements.
Continuing Calibration Verification	Met requirements.

[1] Data Set 1, Initial Analysis (Silvers 1997a).

[2] Data Set 2, Reanalysis (Silvers 1997b).

Table H4. Summary of Surrogate Recoveries, Analysis of K East Basin Canister Sludge and Water Decant Samples for Polychlorinated Biphenyls.

Sample ID	TCX Recovery (%)			DCB Recovery (%)		
	Initial Analysis	Reanalysis	RPD	Initial Analysis	Reanalysis	RPD
Canister Sludge Samples						
Method Blank	94.8	39.8	82	90.3	31.0	98
Method Blank 2	105	89.0	16	99.9	84.3	17
96-01	120	86.8	32	72.4	66.8	8
96-01 Duplicate	111	87.5	24	72.3	68.3	6
96-04	69.7	41.3	51	67.6	19.6	110
96-04 Duplicate	62.3	60.0	4	48.6	30.0	47
96-04 MS	71.3	40.0	56	63.0	44.0	36
96-04 MSD	76.4	64.5	17	80.6	9.45	158
96-05	103	83.8	21	79.8	68.0	16
96-05 Duplicate	101	43.8	79	79.0	42.5	60
96-05 MS	106	81.0	27	77.8	68.3	13
96-05 MSD	90.6	76.8	16	64.1	78.5	20
96-06	92.6	51.5	57	65.9	44.3	39
96-08	92.7	76.0	20	90.7	76.0	18
96-09	104	89.8	15	95.9	85.0	12
96-11	105	62.0	52	101	55.8	58
96-11 Duplicate	75.1	37.3	67	82.0	39.5	70
96-13	101	82.0	21	66.1	76.3	14
96-13 Duplicate	74.2	55.8	28	73.9	48.5	42
96-15	94.2	78.8	18	71.8	57.5	22
Canister Water Decant Samples[1]						
Method blank	39.5	NA	NA	52.3	NA	NA
Method blank MS	83.4	NA	NA	72.6	NA	NA
Filter blank	48.4	NA	NA	26.0	NA	NA
Filter blank MS	57.6	NA	NA	32.6	NA	NA
LCS	49.8	NA	NA	55.8	NA	NA
96-01-Cyl	43.3	NA	NA	60.6	NA	NA
96-05-Cyl	53.1	NA	NA	44.9	NA	NA
96-05-Cyl dup	53.0	NA	NA	53.1	NA	NA
96-05-Cyl MS	43.0	NA	NA	42.9	NA	NA
96-05-Cyl MSD	75.6	NA	NA	76.5	NA	NA

[1] No reanalysis of water decant samples was required. NA = not applicable.

Table H5. Summary Statistics for Polychlorinated Biphenyls Results from K East Basin Canister Sludge.

K East Canister Sludge Samples Summary Statistics	Initial Analysis Aroclor 1254 As-Settled Sludge (μg/kg)	Reanalysis Aroclor 1254 As-Settled Sludge (μg/kg)
Minimum concentration observed	60[1] (135)	13
Maximum concentration observed	1110	880
Number of observations	9[1] (6)	9
Mean	300[1] (420)	220
Standard deviation	370[1] (410)	300
Relative standard deviation (%)	120[1] (100)	140
Upper tolerance limit (0.95, 0.95)[2]	1400[1] (2000)	1100
Upper tolerance limit (0.95, 0.99)[3]	1800[1] (2500)	1500
Analytical variability (1 σ) - Based on the sample duplicates	23%[1] (20%)	14%
Systematic variability (1 σ) - Based on the spike analyses	3.5%	20%

[1] The less than results were used in calculating the summary statistics (e.g., 3 for <3); the numbers in parentheses are the summary statistics when the less than values are deleted from the calculations.

[2] There is 95% confidence that 95% of the data lie below 1400 μg/kg as-settled sludge (assuming normality).

[3] There is 95% confidence that 99% of the data lie below 1800 μg/kg as-settled sludge (assuming normality).

significant at the 0.05 level (i.e., 95% confidence level). This difference indicates that the two data sets are distinct and should not be combined or averaged. Systematic variability, which is based upon spike recoveries, was much greater for the reanalysis data set. The higher systemic variability for the reanalysis data set can be mostly attributed to the low spike recovery for Sample 96-04 MS (49% recovery). Analytical variability, which is based on sample duplicates, ranged from 14 to 23%. Considering the very low quantitation limits achieved, the analytical variability is reasonable.

Upper tolerance levels were calculated to estimate the potential maximum concentration of PCB in the canister sludge. These results show that there is 95% confidence that 95% of the data is less than 1400 ppb (1.4 ppm) and a 95% confidence that 99% of the data is less than 1800 ppb (1.8 ppm) Aroclor 1254.

H.6 REFERENCES

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APPENDIX I

PHYSICAL AND RHEOLOGICAL PROPERTIES OF K EAST CANISTER SLUDGE

(Density, Viscosity, Settling Rates, Zeta Potential,
Particle Shape, and Particle Size)

G. R. Golcar, P. R. Bredt, and J. M. Tingey

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APPENDIX I

PHYSICAL PROPERTIES OF K EAST CANISTER SLUDGE

I.1 INTRODUCTION

The results from characterizing the physical properties of the K East (KE) Basin canister sludges are evaluated and summarized in this appendix. This work was based on the specific needs of the Spent Nuclear Fuel (SNF) sludge disposition project. The KE canister sludge measurements and the physical properties data are documented in detail in Silvers et al., 1997. In this appendix, the physical properties of sludge settling behavior, rheological measurements, sludge density, particle size distribution, sludge particle shape, and the zeta potential measurement are evaluated. These evaluations are based on the needs which influence the specification, design, and performance of the candidate canister sludge removal, washing, dewatering, and storage systems.

The two K Basins at the Hanford Site are water-filled concrete pools that contain over 2,000 tons of N Reactor metallic uranium elements stored in aluminum or stainless steel canisters. Visual inspections of the fuel elements in canisters have shown that the surfaces of the fuel elements are covered with a thin layer of corrosion products, and an accumulation of sludge in the bottom of the canisters covers the lower part of the fuel elements. The sludge in the canisters has accumulated by gravitational settling of corroded fuel, canister corrosion products, windblown debris such as sand and insects, and debris from the basin operations. Most of the residual corrosion products on the surface of the fuel elements are caused by the corrosion of metallic uranium fuel exposed to water when the zirconium alloy cladding is mechanically breached (Johnson 1995). In addition to the sludge found in the canisters, a settled sludge layer has accumulated on the main basin and remote pit floors, but this appendix does not address the floor sludge (see Makenas 1996 for related data on floor sludge).

During fiscal year FY 1997 and the fourth quarter of FY 1996, physical, rheological, radiochemical, and organic analyses of sludge samples obtained from nine KE canister sludge samples were performed (as specified in the sampling and analyses plan, Welsh 1996) by staff from the Pacific Northwest National Laboratory (PNNL) Process Chemistry Group and Analytical Chemistry Laboratory (ACL).

This appendix summarizes the proposed SNF project canister sludge cleaning and packaging systems and the sampling history of the KE Basin canister sludges. Measurements obtained for batch settling, particle size distribution (PSD), particle shape, sludge and solid mixture densities, and rheology of the canister sludge samples are reported and any potential impacts of these measurements to the proposed SNF project canister sludge cleaning and packaging systems are discussed.

I.2 CONCLUSIONS AND RECOMMENDATIONS

- Observation of the settled sludge layer suggests that the majority of solid particles at the top of the sediment layer are fine particles in the micron and sub-micron range, whereas most of the sludge that accumulated at the bottom of the sediment layer consisted of particles greater than several hundred microns in diameter.

- The sedimentation behavior of the canister sludge is characterized by an initial sedimentation rate and a final sedimentation volume as measured in 2 liter graduated cylinders. The majority of particles settled at a rate greater than 0.1 cm/min. For most of the KE canister sludge samples, the suspension settles at a rate of approximately 0.5 to 3 cm/min in the top 60% of the suspension volume. The final 40% of each settling volume will settle at a decreasing settling rate that falls from approximately 0.1 cm/min to less than 0.0001 cm/min.

- The PSD results from the wet sieving revealed that a major mass fraction (42 to 44 dry wt%) of the sludge, from the canisters with the fuel elements, was greater than 710 microns in diameter, but only a small mass fraction (approximately 2 dry wt%) of the nonfueled canister sludge sample was greater than 710 microns in diameter. These large particles will likely settle in the basin slurry transfer pipe line because of their large size and their high densities. These large particles will also likely separate in settling containers due to their high settling velocities.

- The wide spread of the PSD by a volume-weighted distribution of all the research samples shows that these samples were polydisperse, and as a result the mean size of the PSD by a volume-weighted distribution is much larger than the mean size of the PSD by a number-weighted distribution.

- The particle shape measurements suggest that canisters with corroded fuel contain nearly oblate to slightly elongated particles that are high in uranium content. More elongated to needle-like particles composed of quartz (SiO_2) are found in most of the canisters, but are in higher proportion to uranium particles in canisters that do not contain fuel elements.

- The canister sludge particle shape results are different than KE Basin floor and Weasel Pit sludge. The floor and Weasel Pit sludge samples contained needle-like particles not seen in the canister sludge samples. The needle-like particles seen in the KE Basin floor and Weasel Pit sludge, were sand particles that were composed of magnesium, aluminum, iron, and silicon. Uranium is the primary component of the canister sludge, not the components of iron and sand as were observed in the floor and Weasel Pit sludge.

- The results from the X-Ray Diffraction (XRD), the dry particle density, and the uranium concentration indicate that the major components of the canister sludge are uranium oxides with theoretical dry-solid densities between 8 and 11 g/mL. Other important components include silicates and iron oxides with dry densities between 2.5 and 6.5 g/mL, and possibly metallic uranium with a density of 19 g/mL. The density of a layer of these materials, when wet, are dependent on the packing efficiency. If a conservatively high packing factor of 70 vol% solids is assumed, then the range of wet densities for these materials, excluding uranium metal, would be between 8.0 and 2.0 g/mL. Most of the particles in the canister sludge are uranium oxides with wet densities between 8.0 and 5.9 g/mL.

- The zeta potential of the "nonfueled" 96-11 sample shows the same trend as the zeta potential of the floor sludge and the Weasel Pit sludge; however, the isoelectric point of Sample 96-11 occurs at about pH 6, which indicates that the surface of the particles is predominantly coated with a more basic oxide then was observed in the floor or Weasel Pit sludge. The magnitude of the zeta potential of the "fueled" canister samples (Samples 96-04 U/L, 96-04 L, 96-06 L, and 96-06 L) is higher than the floor sludge and the Weasel Pit sludge results. These results indicate that the concentration of the charge on the surface of the "fueled" canister sludge particles is higher and the "fueled" canister sludge particles tend to be more receptive to the adsorption of ions.

I.3 CANISTER SLUDGE CLEANING AND PACKAGING SYSTEM

An overview of the various proposed SNF project canister sludge cleaning and packaging systems is presented in this section. The role of various physical properties of the canister sludge and the necessary actual canister sludge data are discussed. This discussion is in the context of applying and operating these systems.

It is planned to retrieve K Basin SNF from the basins and repackage it into Multi-Canister Overpacks (MCOs) for eventual storage at the Canister Storage Building. In general sludge is removed from the canisters in the basin, water is separated from the sludges, and this concentrated sludge is disposed of through various processes. The processes for handling the sludge are currently being evaluated and include operations such as

- Washing of fuel elements and canisters to remove sludge
- Slurry transport pumping within the basin
- Solid-liquid separation processes at the integrated basin water treatment facility
- Storage and disposition of dewatered canister sludge.

A summary of these processes and the relevant key physical properties of these operations are given in the following paragraphs.

Baseline Primary Washing Machine and Cleaning Mechanism--In the process of retrieving fuel elements, a SNF washing process is used to remove sludge* from the surface of the fuel elements. A primary wash cycle is designed to remove the majority of the sludge from the canisters, clean SNF before it is removed from canisters, and minimize the impact to water quality downstream from the primary washing process.

The primary wash cycle system uses only physical processes with no chemical treatment to remove sludge from the spent fuel elements. Removal of sludge from the fuel elements depends on the sliding action of the fuel elements against one another and the erosion of the solid particles by the rinsing action of a water jet. The degree of adhesion between the solid particles and the surface of the fuel is the primary physical property affecting the solid particle erosion process.

In-Basin Slurry Pipeline Transfers--The basin slurry transfer pipe lines will be used to transfer the discharged slurries to the water treatment facility. Slurries will originate from the fuel retrieval process, fuel washing, the sludge retrieval process, the debris retrieval process, and the MCO/cask loadout process.

The physical properties important to slurry transport are the mean particle size, particle size distribution, particle shape, density of particles, concentration of solid particles, density of the water (the carrier liquid), and the viscosity of the slurry and the effect of temperature on the viscosity/rheology. A combination of these properties will affect the transport velocity and the pressure drop across the pipeline. For slurry transport robust turbulence is desired to keep all particle sizes suspended, or at a minimum, maintain a moving bed of sediment in the lower portion of the pipeline. Assuming fully suspended transport, the pressure drop across the transfer pipeline depends on the apparent viscosity of transported slurry. This slurry viscosity is dependent primarily on the carrier liquid viscosity and the slurry solids loading.

Sludge Separation Process--To decrease the filter loads hydrocyclones are included in the integrated water treatment system as a coarse solid-liquid separation step between the primary fuel-cleaning machine or the floor-sludge overflow stream and the backflushable filtration system which is intended to decrease the filter loads. The solid/liquid underflow mixture from this step will be routed to the Weasel Pit which acts as a "settling tank." The solid-liquid overflow is transferred to the water treatment process where it is filtered by a backflushable filtration system.

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*In this appendix, the canister sludge is defined by the SNF project as being made up of solid particles less than 0.25 in. (6350 microns) in diameter. Consistent with this the characterization sampling system precludes solid particles larger than 0.25 in. in diameter from being drawn into the canister sludge samples. In addition, the canister sludge is described as a nonhomogeneous mixture of corrosion products from metallic uranium fuel elements, canister corrosion products, and settled debris, such as windblown sand, that are found in the fuel element canisters.

The operating performance of hydrocyclone separation is measured by the amount of the solid fraction contained in the underflow. When suspended solid particles flow in a hydrocyclone, the particles are subjected to two forces: (1) the external and internal fields of acceleration caused by the force of gravity and centrifugation, and (2) the drag exerted on the particle by the flow. Typically, gravity may be neglected and only the centrifugal and drag forces are considered. The centrifugal force acts in the radial direction, which prevents the particle following the inward radial flow. If the centrifugal force acting on a particle exceeds the drag, the particle moves radially outward; and if the drag is greater than the centrifugal force, the particle is carried inward. When solids are thrown out to the wall, they flow down the inclined walls and exit at the bottom. The key physical properties of the feed slurry that affect the separation efficiency of hydrocyclones are the cumulative effect of density of solid particles verses the fluid density, apparent viscosity of slurry, and particle-size distribution of the solids.

I.4 K EAST CANISTER SLUDGE SAMPLING HISTORY

In June 1996, nine KE Basin canister sludge samples were transferred from the 327 Building to the 325 Building High Level Radiochemistry Facility (HLRF). These samples were contained in sealed stainless steel shipping containers with an approximate volume of 10.5 L each. Table I.4-1 lists the samples delivered along with the volume of settled sludge recovered from each of the shipping containers.

The samples were vacuum transferred out of the stainless steel shipping containers into 10-L glass carboys and 2-L glass graduated cylinders. The majority of the liquid was transferred into 10-L glass carboys while the settled sludges with some liquid were transferred into 2-L glass graduated cylinders. A fine particulate layer was noted on the bottom of all the shipping containers following the initial vacuum transfer. Repeated additions of supernatant from the same shipping container followed by vacuum transfer were used to recover these fine particles and transfer them to the graduated cylinders. Additional supernate was then added to the graduated cylinders to bring the volume in each cylinder to approximately 1.7 L. These cylinders were utilized for settling studies as described in Section I.5.

The residual sludge layer in the bottom of shipping container Sample 96-06 could not be removed by only supernatant addition and vacuum transfer; therefore, the stainless steel tip of the vacuum transfer line was used to scrape the sludge layer and to suspend this layer in the supernatant. After this layer was removed, the bottom of shipping container Sample 96-06 was visually inspected. The bottom of this shipping container appeared rough which increased the adhesional force and caused the solid particles to adhere more readily to the container walls.

Bubbles were observed rising from the settled sludge layer in several samples within a few days after the samples were loaded into the graduated cylinders. Gas samples were collected from the graduated cylinders and the remaining five sealed stainless steel shipping canisters. These samples were analyzed by isotopic mass spectroscopy, and the results suggested the presence of metallic uranium or uranium hydride oxidation in four of the nine samples (96-05, 96-06, 96-13, and 96-15). Details of this gas study, including estimated generation rates and gas-retention rates, are reported in Appendix B.

The carboys containing the sample supernate were reexamined several weeks after the initial transfer, and a layer of fine solid particles had settled in the carboys containing liquid from Samples 96-06 and 96-15. These solids were recovered but the masses of these layers were not measured, because the solids were not separated from the liquid.

I.5 VISUAL OBSERVATIONS, BATCH SETTLING MEASUREMENTS, AND LAYER SEPARATION

The graduated cylinders holding the transferred sludge were 41 cm in height and 8.0 cm in diameter. A few days after transferring the samples, the contents of the graduated cylinders were sparged to resuspend the solids, and the settling studies were initiated. The sludge and supernatant in the graduated cylinders were sparged with either air or helium gas for a minimum of 5 minutes to uniformly mobilize the solid layer and obtain a homogeneous slurry. Following gas sparging, the resulting slurries were left undisturbed for a minimum of 2 weeks while the settling behavior was observed. The settling measurements of all nine canister sludge samples were divided in two sets. The settling behavior of each graduated cylinder was monitored and recorded using several video cameras. In the first set of experiments, the settling behavior of the sludge from canister Samples 96-01, 96-05, 96-06, and 96-08 was monitored. In the second set, the settling behavior of the sludge from canister Samples 96-04, 96-09, 96-11, 96-13, and 96-15 was examined.

In general, the sedimentation of polydisperse and multi-density suspensions such as KE canister sludges can be measured by monitoring the formation of various interfaces within the length of the container. As a uniform suspension settles, the solid particle concentration becomes non-uniform, and approximately four regions of varying solid concentrations within the container length can be formed. A clear layer, which contains limited particles, is formed at the top; a sediment layer is formed at the bottom of the container; between these two layers, a cloudy settling suspension may exist. The upper region layer contains the slower settling particles, and the lower regions containing the faster settling particles.

When solid particles begin to settle in the graduated cylinder, they are subjected to four forces: (1) a downward gravitational force, (2) an upward buoyant force due to the displaced fluid, (3) a frictional drag force from the surrounding suspension, and (4) the interparticle interaction forces. In a column batch settling experiment, the overall settling behavior of solid particles can be categorized as either free falling, hindered settling, or a combination of both free falling and hindered settling.

In the free-falling settling, the sedimentation of a solid particle is independent on the particle concentration in the suspension, and the particles settle based on their terminal or free-falling settling velocity. In general, under free-falling conditions a distinct sediment layer forms at the bottom of the container. The volume of this sediment layer increases as a function of time, with the interface between the sediment and supernatant layer beginning at the bottom of the container and moving upward. As the particles settle the turbidity or cloudiness of the suspension decreases.

In hindered settling the settling rate decreases due to interaction of particles with surrounding particles. The closeness of the particle packing (crowding effect of surrounding particles) prevents the differential movement of any single particle; therefore, the settling rate of a suspension decreases with increasing solids concentration. Under hindered settling conditions, the solid layer appears to "condense" (decrease in volume) forming a clear supernatant layer that increases in volume with time. The supernatant and solid layers are separated by a sharp sediment-liquid interface with the sedimentation starting at the top of the sediment layer an moving downward with time.

Batch Settling Results--At the beginning of the settling experiment, the contents of each graduated cylinder were sparged with either air or helium to obtain a uniform suspension. The graduated cylinders containing sludge from canister Samples 96-01, 96-05, 96-06, and 96-08 were sparged with air, and the graduated cylinders containing sludge from canister Samples 96-04, 96-09, 96-11, 96-13, and 96-15 were sparged with helium. The resuspension of solid particles in the graduated cylinders by sparging may break existing weak aggregates in the canister sludge. However, such aggregate shearing is considered appropriate for these settling tests since in the basin, the canister sludge will experience the same or higher levels of shear when it is suspended in the basin.

In all of the batch settling studies, the only downward or settling force is gravitation. The initial sediment lay in all samples formed from the bottom of the graduated cylinder and the sediment-liquid interface moved in the upward direction. In Figure I.5-1, the settled volume percentage* as a function of settling time for the first set of settling experiments (canister Samples 96-01, 96-05, 96-06, and 96-08) is shown. Based on these results, Samples 96-01, 96-05, and 96-06 show similar settling trends. The sediment volumes of Samples 96-01, 96-05, and 96-06 drop steeply with the final sedimentation volume of these samples being reached in approximately 1 hour. During the first hour of settling the settling rate of Sample 96-08 was constant, followed by compaction of the solids in the next 3 hours. In Figure I.5-2, the settled volume percentage as a function of the settling time for the second set of settling experiments (canister Samples 96-04, 96-09, 96-11, 96-13, and 96-15) is presented. As in the case of the first set of settling experiments, within the first hour of settling, a sharp drop in the sedimentation volume of Samples 96-15, 96-13, and 96-04 is observed. A top and a bottom settling layer were observed for Samples 96-09 and 96-11. Well-defined settling layers were observed during the settling of Samples 96-09 and 96-11. The top layer of these samples settled at an

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*In this section, the settled volume percentage is defined as the percentage of settled layer volume to the initial volume of suspended slurry.

initial constant settling rate. With the exception of Samples 96-08 and 96-13, the solid layers in the graduated cylinders containing sludges from canister Samples 96-01, 96-05, 96-06, 96-04, the Sample 96-09 lower interface, Sample 96-11 lower interface, and Sample 96-15 dropped to about 15 to 25% of their initial uniform suspended volumes.

The sedimentation behavior can also be characterized by the initial sedimentation velocity and the final sedimentation volume. Figure I.5-3 shows the settling rate of the canister sludge samples as a function of the settled volume percentage. For most of the KE canister sludge samples, the suspension layer settles at a rate of approximately 0.5 to 3 cm/min in the top 60% of the suspension volume. The final 40% of each settling volume, settles at a decreasing settling rate that falls from approximately 0.1 cm/min to less than 0.0001 cm/min.

Photographs of the samples are provided in Figures I.5-4 through I.5-16. The settled solids from Samples 96-01, 96-05, 96-06, 96-08, 96-13, and 96-15 were dark gray-brown to black in color and the settled solids from Samples 96-04, 96-09, and 96-11 (while still dark) were more yellow-brown. Samples 96-09 and 96-11 appeared the lightest in color. Clear settled supernatant was observed in Samples 96-01, 96-05, 96-13, and 96-15. The settled supernatant from Samples 96-08, 96-09, and 96-11 was cloudy and yellow in color. The settled supernatant from Sample 96-04 was clear yellow-green. The settled supernatant for Sample 96-06 was cloudy and light gray-brown in color. Sample 96-06 could have been cloudy due to resuspension of solids resulting from continuous bubble release from the sludge layer. The hot cell environment may tend to influence perceived sample colors toward yellow because of the color of the hot cell lead glass windows.

Close-up images of Samples 96-04, 96-09, and 96-11 are included in Figures I.5-6, I.5-11, and I.5-13, respectively. These images show the granular nature of the material in the bottom layers of these samples. Several of the flakes at the bottom of Sample 96-04 were separated. A few of the flakes appeared glass like. Several of the non-glass flakes were analyzed for crystalline species by XRD. Results indicated these flakes were composed primarily of zirconium and zirconium hydride. The texture of the upper portion of these sludge samples was smoother and contained a higher proportion of finer sized particles.

The SNF Characterization Project group designated three samples as "research samples" (96-04, 96-06, and 96-11) and the remaining samples (96-01, 96-05, 96-08, 96-09, 96-13, and 96-15) as "normal samples." "Research samples" were intended for supplementary physical property determinations while "normal samples" received only chemistry related analyses after settling. Sample 96-04 was chosen as a research sample from a KE fuel storage canister made of stainless steel. Sample 96-06 was chosen as a research sample from a KE fuel storage canister made of aluminum, and Sample 96-11 was chosen as a research sample from a stainless steel canister with no fuel. The research samples were recovered in layers from the graduated cylinders after the settling experiment was completed. Each recovered layer was treated as a unique sample. Figures I.5-5, I.5-8, and I.5-12 identify the layers within the research samples. The layers were separated by vacuum transfer, if enough sample was available (>100 ml), or by decanting the layer with a glass pipet if the layer was too thin to perform adequate separation with vacuum transfer. The glass pipet was connected to a syringe with tygon tubing, and the syringe was used to pull the sample into the pipet. The mass of each sample layer and the method used to obtain these samples are reported in Table I.5-2.

Sample 96-04 was split at the interface between the smooth and granular layers. Sample 96-06 was split into three layers, an upper dark fluffy layer, a middle lighter layer, and a lower layer. Sample 96-11 was split at the interface between the thin light colored fluffy upper layer and the darker bulk of the sample. To help ensure that material from one layer was not mixed with that of the next, the interfaces between the identified layers were collected as separate samples. It was not originally intended to use these interface subsamples for physical and rheological characterization; however, since some layer samples were so small, some analyses were made on these separated interface samples.

The remaining six samples were designated as "normal" samples. The settled solids from the normal samples were recovered from the graduated cylinders and transferred to unique jars.

I.6 PARTICLE SIZE DISTRIBUTION MEASUREMENT

The particle size distribution measurements were performed on research Samples 96-04 U/L, 96-04 L, 96-06 carboy solids, 96-06 M, 96-06 L, and 96-11 L.* The photographs of the sludge samples after they had settled in the graduated cylinders (see Figures I.5-6, I.5-8, and I.5-13) showed that these sediments are composed of a wide range of particle sizes. The majority of solid particles at the top of the sediment layer appeared to be fine particles in the micron and sub-micron range, whereas most of the sludge that had accumulated at the bottom of the sediment layer was greater than several hundred microns in diameter.

Thus, to characterize such a broad PSD, two separate methods were used. A Microtrac X100 Particle Size Analyzer was used to measure particle sizes from 0.12 to 704 microns, and a wet sieving technique was used to fractionate particles as coarse, medium, and fine. To sort the particles in these categories, sieves with openings of 3.35, 2.36, 1.18, and 0.71 mm were used. Both particle size analyses were used to measure the PSD of research Samples 96-04 L, 96-06 M, 96-06 L, and 96-11 L. Only the Microtrac X100 Particle Size Analyzer was used to measure research Samples 96-04 U/L and 96-06 carboy solids. Because of insufficient sample the, wet sieving analyses could not be measured on research Samples 96-04 U/L and 96-06 carboy solids. The entire PSD of research Sample 96-06 carboy solids was smaller than 700 microns.

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*The information on the origin of these research samples and the criteria for their selection to perform detailed physical and rheological measurements are described in Section I.5.

In general, the objective of particle size analysis is to discriminate between particles of different sizes. However, the Microtrac X100 Particle Size Analyzer and wet sieving rely on different principles to discriminate and to fractionate particles of different sizes. The PSD data from the Microtrac X100 Particle Size Analyzer and from the wet sieving are not on the same basis. The Microtrac instrument uses light scattering to measure the dimensions of individual particles from which a particle volume-weighted distribution is generated. Wet sieving determines the mass of particles on each sieve, which results in a particle-mass distribution. Converting between the particle sizes on a mass-distribution basis and particle sizes on a volume-distribution basis requires knowledge of particle density. KE canister sludge is heterogeneous and it is possible that the average particle density varies with the particle sizes. Because of this, the PSDs cannot be quantitatively compared and a single rigorous PSD from 0.2 to 3350 microns cannot be generated. However, with an assumption of an "averaged uniform solid mixture" density for the sludge, a qualitative comparison can be made. These assumptions and comparisons are discussed in more detail in Section I.6.3. By making such an assumption, the PSD of the entire size range is obtained, which was the only appropriate route to characterize these radioactive samples.

The wet sieving results are summarized in Section I.6.1. In Section I.6.2 the result of particle size measurements using Microtrac X100 is presented. An overall PSD that results by joining information from both analyses is presented in Section I.6.3, and the necessary assumptions are discussed.

I.6.1 Particle Size Distribution Measurement from 700 to 3350 Microns

A wet-sieving technique was used to fractionate canister sludge samples as coarse, medium, or fine. To sort the canister sludge samples into these categories, sieve sizes of 3.35, 2.36, 1.18, and 0.71 mm were used. Detailed information about these sieves is summarized in Table I.6.1-1. The wet sieving method was used since the canister sludge samples are suspended in the basin water. In addition, the wet-sieving method was used instead of the dry sieving technique to prevent the formation of aggregates when the samples are dried, which will shift the PSD in favor of bigger particles.

In this technique, particles were classified into different sizes as they pass through a stack of sieves. The sieves were stacked in ascending order of aperture size. The largest sieve (Tyler sieve 6) was placed at the top of the sieve stack, and the smallest sieve (Tyler sieve 24) was nested at the bottom of the stack. A final closed pan, a receiver, was placed at the bottom of the stack to collect the particles smaller than 710 microns that passed through all the sieves. In any sieving operation, an important factor that influences the results of sieving operation is sieve loading. This is because errors in weighing and measurement precision results from insufficient weight of material. With the limited amount of canister sludge layers available, and the fact that only a maximum of four sieves could be stacked at each wet-screening operation, a total of four sieves were used to classify the canister sludge samples between a 710 to 3350 microns (but smaller than 6350 microns) size range. It should be mentioned that the sludge sampler device that was used to retrieve canister sludge samples from the KE Basin canisters is designed to sample particles smaller than 6350 microns (0.25 in.) in diameter and to leave behind particles greater than 6350 microns. Thus, the upper end of particle size is limited to particles of 6350 microns in diameter.

Experimental Procedure--In these experiments, a known amount of research sample was dispersed in the basin water and was poured through the stack of sieves. Then, the sludge content was washed through each sieve with the rinsing fluid. At the same time, the accumulated slurry in the receiver pan was flowed into a collection beaker. After the samples were washed through each sieve, the sieves and the beakers were dried in the oven until a stable mass was reached. The dried sample was weighed and the weight percent of solid particles (on dry solid basis) was calculated for each sieve and the beaker.

K East Canister Sludge Wet Sieving Results--The dry wt% of solids was calculated by dividing the dry mass of the material in the sieve or beaker by the sum of the masses of all the dry material in the sieves and the beaker. Table I.6.1-2 summarizes the wt% of solids on a dry basis for each particle size range.

The PSD from the wet sieving analyses shows that 42 to 44 wt% of solid particles (on a dry solid basis) of the samples from the canisters containing SNF (Samples 96-04 L, 96-06 M, and 96-06 L) are greater than 710 microns and smaller than 6350 microns. Also, 56 to 58 wt% of these solid particles are smaller than 710 microns. These results suggest that only 56 to 58 wt% of the PSD from Samples 96-04 L, 96-06 M, and 96-06 was analyzed by the Microtrac instrument. In addition, the sieving results show that 98 wt% of particles for the nonfueled sample (Sample 96-11 L) are smaller than 710 microns, and only a small mass percent of this sample (approximately 2 dry wt%) is greater than 710 microns. For the nonfueled Sample 96-11 L about 98 wt% of its particles were analyzed by the Microtrac instrument. The PSD results from the wet sieving revealed that a major mass fraction (42 to 44 dry wt%) of the sludge from the canisters with the fuel elements was greater than 710 microns, but only a small mass fraction (approximately 2 dry wt%) of the nonfueled canister sludge sample was greater than 710 microns. The layers in the sieving of sludge from fueled canisters showed differences in color and composition (Silvers et al., 1997a). Several pictures of the fueled and the nonfueled samples are attached (see Figures I.6.1-1 to I.6.1-4).

The XRD analysis of the sludge from canisters containing SNF, for instance the Sample 96-04 L, indicates that the primary crystalline materials in this canister were various forms of uranium oxide and uranium hydrates such as metaschoepite ($UO_3 \cdot 2H_2O$), schoepite ($UO_3 \cdot 2H_2O$), and uraninite (UO_2, U_4O_9, and U_3O_7). The theoretical density of these particles range from 10.96 g/cc for UO_2 to 4.87 g/cc for $UO_3 \cdot 2H_2O$. The wet sieving analyses indicate that 42 to 44 wt% of solid particles (on a dry solid basis) from the canisters containing SNF (Samples 96-04 L, 96-06 M, and 96-06 L) are greater than 710 microns and smaller than 6350 microns. This represents a significant fraction of the sludge mass. These larger particles will most likely settle in the basin slurry transferring pipelines due to their large size and their high densities. In addition, these large particles will most likely separate in the settling containers due to their very high settling velocities.

I.6.2 Particle Size Distribution Measurement from 0.12 to 704 Microns

A Leeds and Nortrup Microtrac X100 Particle Size Analyzer was used for these analyses. This device can measure particle diameters between 0.12 to 704 microns. The analyzer works by analyzing light scattered by the particles in a dilute suspension. The amount and direction of the light scattered by the particles is measured by an array of optical detectors and then analyzed to determine the size distribution of the particles.* To measure PSD, a sample is added manually to a sample reservoir. It mixes with the recirculating fluid so that a stream of well-dispersed particles passes through the sample cell for analysis.

Experimental Procedure--The sludge samples for particle size analyses were slurried before sub-sampling. Some of the samples had dried significantly before subsampling; therefore, approximately 0.25 g of solids were transferred with a spatula and diluted with 5 ml of supernatant from the same KE Basin canister sample. The state of the samples analyzed and the amount of solids added to the 5 ml of supernatant is reported in Table I.6.2-1.

Since solids were also observed in the supernatant from Sample 96-06 that was vacuum transferred from a shipping container into the 10 L carboy, a slurry of these solids was separated and the PSD measured using the Microtrac instrument. The radiation dose rate from these carboy solids was extremely high.

Each diluted sample was swirled or shaken to produce a slurry/suspension of the sample particles. A small amount (2 to 5 ml) of this slurry/suspension was pipetted into the reservoir of the particle-size analyzer to produce a suspension with sufficient particles to make an accurate determination of the PSD. The reservoir contained approximately 500 ml of demineralized and distilled water. The pH of each suspension is reported in Table I.6.2-2. The pH and mineral content of these suspensions compare well with the pH and salt content (as measured by the specific conductance of the water) in the KE Basin.

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*The instrument combines low-angle laser light scattering (LALLS) with 90° scattering at three different wavelengths and orthagonal polarities. This combination will extend the size range to a lower size than is usually used for forward light scattering alone. The forward light scattering and Fraunhofer theory are used to analyze particles coarser than 2 microns. The Mie theory and 90° scatter are used for smaller-sized particles.

A study of the effect of the analysis parameters on the PSD was performed on one of the K Basin research samples (96-04 U/L). The effect of flow rate, circulation time, sonication power and time, whether the particles are spherical or non-spherical, and particle transparency on the particle distribution was determined. As a result the particles were assumed to be non-spherical and transparent for these analyses. The particle size analyses were obtained after applying a variety of circulating flow rates and ultrasonication treatments. The treatments included (1) circulation at 60 mL/s with 25 W ultrasonication for 120 s, (2) circulation at 60 mL/s with 40 W ultrasonification for 300 s, (3) circulation at 40 mL/s with 40 W ultrasonification for 90 s, and (4) circulation at 70 mL/s with 40 W ultrasonification for 90 s. A detailed discussion of the equipment settings are presented in Silvers et al., 1997b.

The particle size analyses for the rest of samples were performed under a number of varying conditions. The flow rates for sample circulation included 40, 60, and 70 mL/s. The samples were measured before ultrasonication (as-received) and after ultrasonication at 25 W for 120 s at 60 mL/s, 40 W for 300 s at 60 mL/s, and 40 W for 90 s at 40 and 70 mL/s. The results under these conditions are reported in Table I.6.2-3.

Mictrotrac Particle Size Distribution Results--The plots for each of the research samples under all of the analysis parameters are presented in Figures I.6.2-1 to I.6.2-22. In these figures, the PSDs for each research measured sample are pictorially presented in a histogram and in a cumulative under-size-percentage plot. These are given on a particle number-weighted basis and on a particle volume-weighted basis. The number-weighted PSD is computed by counting each particle and by weighting all the particle diameters equally. The volume-weighted PSD, however, is weighted by the volume of each particle measured, which is proportional to the cube of the particle diameter. In this case, larger particles are treated as more important in determination of the distribution than the smaller particles. Therefore, the volume-weighted PSD gives information about how the volume and the mass (assuming uniform density) is distributed among particle sizes, and the number-weighted PSD provides information about the population of particles found in each size range. The mean diameters of the volume- and number-weighted distributions are presented in Table I.6.2-3.

The wide spread in the volume-weighted PSD of all the research samples shows that these samples were polydispersed, and as a result the mean size of the volume-weighted PSD is much larger than the mean size of the number-weighted PSD. In addition, the histogram and the cumulative plots of the particle volume distribution of the samples before and after ultrasonication present the bimodal or polymodal distribution nature of these canister sludge research samples. The polymodality of the "as-received" samples supports the contention that these samples are heterogeneous mixtures of various particles.

The histograms and cumulative volume-weighted and number-weighted distributions for research Samples 96-04 U/L and 96-04 L are presented in Figures I.6.2-1 to I.6.2-5 and Figures I.6.2-6 to I.6.2-10, respectively. The histograms and cumulative volume-weighted distributions and number-weighted distributions for research Sample 96-06 M are presented in

Figures I.6.2-11 to I.6.2-15, for research Sample 96-06 L are presented in Figure I.6.2-16, and for the research Sample 96-06 carboy solids are presented in Figure I.6.2-17. The same type of pictorial presentation of the PSDs for the research Sample 96-11 L are shown in Figures I.6.2-18 to I.6.1-22.

The plots of the volume-weighted PSD for the "as-received" research Samples 96-04 U/L, 96-04 L, and 96-06 M are presented in Figures I.6.2-1, I.6.2-6, and I.6.2-11 and they show that the particles in the 0.2 to 704 micron size range are a mixture of different particles and they represent a polymodal distribution. For instance, a subsample from the research Sample 96-04 U/L exhibits two distinct normal distributions of particles at 40 ml/s, which are widely spread out. Using the same subsample, when the instrument flow rate was changed to 70 ml/s and the circulation time (from 2 to 10 minutes) was increased, the wide spread of the bimodal distribution was narrowed significantly, but the distribution showed the same modality. In addition, the mean particle diameter of Sample 96-04 U/L (see Table I.6.2-3) decreased and the distribution shifted toward the smaller-sized particles. This behavior may suggest that the shift in PSD could be attributed to the removal of weakly bounded oxide flocs from the surface of the particles (or more compacted agglomerate centers) upon an increase of shearing forces induced by the change in the circulation time and the circulation flow rate. The measurements at 60 ml/s were performed on a different subsample of Sample 96-04 U/L that does not represent the same mixture of particles and does not capture the same distribution. The same type of conclusions can be made for the PSD of the "as-received" Samples 96-04 L and 96-06 M (see Figures I.6.2-6 and I.6.2-11).

The PSDs of the research samples were also measured after the samples were mechanically sonicated. Each sample was sonicated at various sonication powers and durations. These results are included in Figures I.6.2-1 to I.6.1-22. The ultrasonication of samples at (1) 60 ml/s with 25 W ultrasonication for 120 s, (2) 60 ml/s with 40 W ultrasonication for 300 s, (3) 40 ml/s with 40 W ultrasonication for 90 s, and (4) 70 ml/s with 40 W ultrasonication for 90 s are reported in this Appendix. The data on all the samples, either the canister sludge from fueled canisters (Samples 96-04 U/L, 96-04 L, 96-06 carboy solids, 96-06 M, and 96-06 L) and the canister sludge from the nonfueled sample (Sample 96-11 L) show that agglomerates were originally present in these samples as evidenced by an increase in the number of smaller particles after the ultrasonication.

I.6.3 A Qualitative Overall Particle Size Distribution Results for Combining Both Analysis

For each canister sludge layer, the weight percent of dry solid particles that is fractionated by a sieve size is presented in Table I.6.1-2. These results show that the mass fractions of particles analyzed by the Microtrac system for Samples 96-04 L, 96-06 M, 96-06 L, and 96-11 L were respectively 56, 58, 58, and 98 wt%. The analogy presented here is considered to be qualitative and the validity of such assumptions needs to be investigated. If one assumes that all the major components of the sludge are poly dispersed over the entire size range, then the volume-weighted PSD can also represents the mass distribution of the solids as a function of particle diameter. It is assumed that the average density of particles for each size interval is the same. For instance, the major components of the canister sludge such as uraninite with the density of 12.0 or quartz with the density of 2.7 must exist uniformly over the entire size range. In this case, the weight fraction of particles analyzed by the Microtrac system for Samples 96-04 L, 96-06 M, 96-06 L, and 96-11 L can be directly converted to volume percent. Then for Samples 96-04 L, 96-06 M, 96-06 L, and 96-11 L the volume fraction of particles analyzed by the Microtrac system are respectively 56, 58, 58, and 98 vol%. Further analyses need to made to investigate if the averaged mixture density of particles can be assumed to be constant over the entire size range. In general, it should be noted that the conversion between the PSD of the KE canister sludge by volume and by mass using the indicated techniques cannot be done rigorously and precisely.

I.7 PARTICLE SHAPE MEASUREMENTS

The shape of the particles was analyzed for research layer Samples 96-04 L, 96-06 M, 96-06 L, and 96-11 L. This analysis was conducted using a Brinkmann 2010 Image Analyzer. The Brinkmann 2010 Image Analyzer uses a video camera to capture still images of particles suspended in a glycerol/water solution.* The shape of the particles are obtained by transferring images of a measured area (taken with a video camera) to the computer for shape analysis. In this process, the "out of focus particles" that were caught by the camera are rejected from analysis. A typical example image is provided in Figure I.7-1. Several image from each sample were transferred to the data-analysis system, which identified the particles and measured several parameters, including, aspect ratio, shape factor, Ferret's diameter, particle size (by both number and volume density), particle area, particle volume, and average radius. The instrument is capable of analyzing particles between 0.1 and 60 microns.

The principle of operation for the Brinkmann 2010 analyzer is based on detecting, counting, and sizing individual particles by taking still images in a local measuring zone within the sample cell. A large number of samples needs to be scanned to obtain good statistics on PSD. The particle size and shape data can only be qualitatively compared because each process is run independently, many of the parameters are calculated from different sets of images, and each analysis selects acceptable particles based on different criteria.

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*The large or dense particles tend to settle quickly, which makes a uniform distribution of particles difficult to maintain when still images are taken. Glycerol is added to the water to increase the viscosity of the liquid phase, and thus decrease the settling rate of solid particles suspended in the liquid phase.

Microtrac X 100 particle size analysis system used to obtain the particle size distribution data measures the diameter of more particles in a shorter time interval and thus produces a more accurate statistical distribution than can be obtained by the Brinkmann Image Analyzer. In the Microtrac system the whole field of the particle sample (sample cell) is scanned by the laser light and the PSD is determined from the interaction of the light with the assembly of particles (see Section I.6 for the Microtrac instrument principle of operation). The Microtrac system does not measure the shape of the particles, and the principle of operation of the Brinkmann Image Analyzer is better suited for obtaining shape of individual particles; therefore, the Brinkmann Image Analyzer was used to determine the shape of the particles for these samples. The particle size distribution of the Brinkmann 2010 Image Analyzer which can cover diameters 0.1 to 60 microns, is not included in Section I.6, because it is not as statistically accurate as the data obtained by the Microtrac system.

<u>Experimental Procedure</u>--The research samples were suspended in a glycerol/water solution and were dispersed ultrasonically to break aggregates. A sub-sample of the resulting suspension was transferred to cuvette that contained a glycerol/water solution to achieve the desired dilution for taking pictures. A stirring propeller was used to maintain the content of the analytical cuvette uniform in between taking pictures. To take still images, the stirring propeller was turned off and images were taken.

<u>Particle Shape Results</u>--A complete set of the measured parameters is reported in Silvers 1997. The most useful parameters from the Brinkmann 2010 with respect to particle shape are aspect ratio and shape factor. The aspect ratio is defined as the ratio of the smallest to largest Ferret's diameters of a particle. The Ferret's diameter is the distance between two tangents on opposite sides of the particle, parallel to some fixed direction. The Ferret's diameter as measured by the Brinkmann 2010 are determined by rotating the particles at 0, 45, 90, and 135 degrees. The shape factor is defined as:

$$\text{Shape Factor} = \frac{\text{Surface area of a sphere having the same volume as the particles}}{\text{Surface area of particles}}$$

The shape factor is a measure of deviation of the shape of the particle from spherical. An aspect ratio or shape factor of one corresponds to a sphere and values approaching zero correspond to acicular (needle-shaped) particles.

Figures I.7-2 through I.7-5 present the aspect ratio and shape factor data for Samples 96-04 L, 96-06 M, 96-06 L, and 96-11 L. The mean values of shape factor and the aspect ratio of these samples are presented in Table I.7.1.

The aspect ratios of particles from Samples 96-04 L, 96-06 M, and 96-06 L show similarly shaped distributions. These three samples are from the KE fuel storage canisters, which contain fuel elements. The histograms of the aspect ratios for these three samples show two distinct distributions centered at 0.5 and 1.0 and a much less pronounced distribution around 0.7. As stated earlier, the aspect ratio is defined as the ratio of the smallest to largest

Ferret's diameters of a particle. The aspect ratio of 0.5 may indicate that particles of oblate or thick plate shape were present. In addition, in all three sludge samples, the percentage of the particles with the aspect ratio of 0.5 was approximately the same as the percentage of the particles with the aspect ratio of 1.0. The similar percentage of these aspect ratios may indicates that half of the time an edge view of the particles (the aspect ratio of 0.5) is measured, and the other half of the time, the face view of the particles (the aspect ratio of 1.0) is measured. The less pronounced distribution around 0.7 may suggest that the suspended particles are oblate to spherical with different aspect ratios. These results suggest that the particles in Samples 96-04 L, 96-06 M, and 96-06 L were asymmetrical and on the "average" oblate or thick plate shaped.

The histogram of shape factors for Samples 96-04 L, 96-06 M, and 96-06 L demonstrates that the shape factor of these canister sludge particles is smaller than 0.8 (95% of the time), and on the average the particle shape factors are about 0.65. The shape factors of 0.8 and smaller support the conclusions from the aspect ratio results, which show the deviation of these particles from spherical shape. The shape factor data further indicate that most of the particles in Samples 96-04 L, 96-06 M, and 96-06 L were oblate. Particles from Sample 96-11 L (from a nonfueled canister) were more oblate with an average aspect ratio and shape factor of 0.6 and 0.58, respectively. However, as with the other samples, Sample 96-11 L did not contain any needle-like particles. The smallest aspect ratio measured for any of the particles in Sample 96-11 L was 0.0625 with most particles above 0.4.

The data for the average radius of particles indicate that in all of the samples (Samples 96-04 L, 96-06 M, 96-06 L, and 96-11 L) 90 to 99% of the particles analyzed for shape had diameters of 2 microns or smaller. This indicates that the presence of a similar percentage of particles with the aspect ratios of either 0.5 and 1.0 is not attributable to particles of different sizes. These aspect ratios (0.5 and 1.0) suggest that these aspect ratios are independent of particle size, and particles of the same size have different shapes.

These particle shape results are different than those previously reported by J. Liu for KE Basin floor and Weasel Pit sludge (Silvers 1995). The floor and Weasel Pit sludge samples contained needle-like particles (not seen in the canister sludge samples) as well as some elongated particles. The needle-like particles were sand particles that were composed of magnesium, aluminum, iron, and silicon. Uranium is the primary component of the canister sludge. The TEM results for the floor and Weasel Pit sludge show the uranium-containing particles to be more elongated. This explains why the canister sludge particles, with the exception of Sample 96-11, were oblate to slightly elongated. Sample 96-11, which was only 10% uranium by weight, contained silica (SiO_2 determined by XRD), explaining the presence of a higher proportion of needle like particles relative to fueled canisters.

I.8 SLUDGE AND SOLID PARTICLES DENSITY MEASUREMENT

The supernate density and the density of the bulk settled sludge were measured on eight of the canister sludge samples. Five settled sludge samples (96-05, 96-08, 96-09, 96-13, and 96-15) were centrifuged, and their densities were measured. The density of the settled sludge layer was measured on two of the research layers (Samples 96-06 U and 96-11 U). The centrifuged sludge and dried sludge densities (density of dried solid mixture) were measured on most of the research samples.

Experimental Procedure--The density of the bulk settled sludge layer was measured by recording the volume and the mass of the settled sludge layer in the graduated cylinders after the settling experiments (see Section I.5) were completed, and the supernatant was removed. This procedure was used to measure the density of the bulk settled sludge for Samples 96-04, 96-08, 96-09, 96-11, 96-13, and 96-15. The mass and volume for Samples 96-01, 96-05, and the duplicate measurements of Samples 96-08 and 96-09 were made by determining the mass and volume of subsamples of the bulk settled solids in graduated centrifuge cones. The settled solids were centrifuged at approximately 1000 x g for 1 hour to remove any entrapped air bubbles which would affect the volume of the sample. Following centrifugation the volume and mass of the centrifuged supernatant, centrifuged sludge, and bulk settled solids (total sample in the centrifuge cone) were measured. The respective sums of these three mass and volume components were assumed to be the same as the settled sludge values without air bubbles.

The centrifuged solid densities for Samples 96-01, 96-05, 96-08, 96-09, 96-13, and 96-15, and the research Samples 96-04 U/L, 96-04 L, 96-06 U, 96-06 M, 96-06 L, 96-11 U, and 96-11 L were measured by transferring subsamples of the bulk settled sludge to graduated centrifuged cones, weighing the subsamples, and centrifuging the subsamples at approximately 1000 x g for 1 hour. Following centrifugation, the volume and mass of the centrifuged solids and supernatant were recorded. The centrifuged solids densities were then calculated by dividing the mass of the centrifuged sludge sample by its volume.

Dried solids densities for research Samples 96-04 U/L, 96-04 L, 96-06 M, 96-06 L, and 96-11 L were measured using a Micromeritics AccuPyc 1330 pycnometer modified for glovebox operation. This instrument measures the volume of small samples, 0.1 to 1.0 ml, by exposing the sample to a known pressure of ultra-high purity helium and then measuring a pressure drop as the gas is vented to a known volume. Samples were prepared by centrifuging at 1000 x g for approximately 1 hour. The centrifuged solids were transferred to a 1-dram (about 3.7 ml) vial and placed in an oven at 105 °C until a stable mass was reached (24 hours). These vials were placed back in the oven for an additional 24 hours and reweighed to verify that a stable mass had been reached. A weighed portion of the dried solids was placed in the pycnometer, and the volume was measured. The dried solid densities were then calculated by dividing the mass of dried solids by the measured volume. Due to the high dose rates associated with these dry materials, it was not possible to conduct duplicate analyses with different subsamples. Volume measurements were made twice with the same subsample.

Sludge and Solid Particles Density Measurement--The measured supernatant, settled solids, centrifuged solids, and dried solids densities are listed in Table I.8-1. In general, the density values reported for the sludges from canisters containing fuel are significantly higher than those previously reported for the KE floor and Weasel Pit sludge. Settled solids densities from canisters containing fuel are between 1.18 and 2.46 g/ml. Those samples whose uranium content was greater than 80% (five of the seven samples from fuel containing canisters) have measured densities between 1.84 and 2.46 g/ml. All but one of the 23 settled-solids samples from the floor and Weasel Pit (Silvers 1995) had measured densities below 2 g/ml with 12 having densities below 1.5 g/ml. Centrifuged floor and Weasel Pit sludge densities were measured between 1.13 and 3.81 g/ml with all but one sample below 2 g/ml. Centrifuged sludge densities from canisters containing fuel and uranium contents >80 wt% in the sludge were measured between 2.36 and 4.52 g/ml. The densities of the samples from canisters which did not contain fuel were similar to the KE floor and Weasel Pit sludges.

The high density values for the canister sludge compared to the floor and Weasel Pit sludge are explained by the uranium content. Uranium concentrations for the dried canister sludge are listed in Table I.8-2. Dried sludge from canisters without fuel (Samples 96-09 and 96-11) had uranium concentrations of 8.8 and 9.3 wt%, respectively. These two samples also had the lowest densities (approximately 1.07 g/ml for the settled sludge). The samples from canisters containing fuel have settled solids densities between 1.18 and 2.51 g/ml. Two of the samples from canisters containing fuel had uranium contents (on a dried weight basis) of 40 wt% (Sample 96-08) and 54 wt% (Sample 96-04 L) and had settled solids densities of 1.21 and 1.18 g/ml, respectively. The other samples from fuel containing canisters had much higher uranium contents (>80 wt%) and significantly higher settled solid densities (1.84 to 2.46 g/ml). By comparison, the uranium concentrations in the floor and Weasel Pit sludge samples were between 0.3 and 33 wt% (dry weight basis) with an average value of only 10 wt% and densities between 1.04 and 1.75 g/ml with the exception of one sample which had a density of 2.28 g/ml.

XRD data suggests that most of the uranium is probably in the form of UO_2 (or higher oxides) although both XRD and drying experiments, detailed in Appendix M, suggest the presence of hydrates.

Table I.8-3 summarizes the major crystalline components of the canister sludge as determined by XRD analysis. Also listed in this table are theoretical density values for these crystalline components. Sample 96-06 has the highest measured dried solids densities of the research layers (6.90 and 7.88 g/ml for the middle and lower layers, respectively). Sample 96-06 also has the highest uranium content (approximately 83 wt% on a dry weight basis). The only crystalline species identified in this sample were oxides of uranium. As Table I.8-3 indicates, these uranium oxides all have densities between 8 and 11 g/ml. The high densities of the lower and the middle layers of the Sample 96-06 may also be attributed to the presence of a small amount of uranium metal or hydride. While uranium metal was not identified in the XRD analysis for Sample 96-06, its presence is suggested by the gas analyses discussed in Appendix B.

Crystalline species identified in Sample 96-04 include zirconium, zirconium hydride, and uranium oxide hydrates, in addition to uranium oxides. The first three of these have densities below 6.5 g/ml. These lower densities explain why the dry particle densities measured for Sample 96-04, 4.68 and 4.76 g/ml for the U/L and L layers, respectively, are below those measured for Sample 96-06. Crystalline species identified in Sample 96-11 included only iron oxides and quartz. These species have densities below 5.25 g/ml with quartz having a density of only 2.65 g/ml. Therefore, Sample 96-11 has the lowest dry-particle density.

The results from the XRD, the dry particle density, and the uranium concentration indicate that the major components of the canister sludge are likely uranium oxides with theoretical dry-solid densities between 8 and 11 g/ml. Other important components include silicates and iron oxides with dry densities between 2.5 and 6.5 g/ml, and possibly metallic uranium with a density of 19 g/ml. The density of these materials, when packed as wet layers, is dependent on the packing efficiency. If a conservatively high packing factor of 70 vol% solids is assumed, then the range of wet densities for these materials, excluding uranium metal, would be between 8.0 and 2.0 g/ml. Most of the particles in the canister sludge are uranium oxides with wet densities between 8.0 and 5.9 g/ml.

I.9 RHEOLOGICAL MEASUREMENT

Various sludge removal, slurry transport, slurry settling, and solid/liquid separation systems are needed to clean the two Hanford K Basins and separate solid particles from the basin water. Solid/liquid suspensions ranging from dilute slurries to concentrated sludges will be processed throughout this cleaning process; therefore, the flow and deformation behaviors of a wide range of suspensions need to be examined. Rheological measurements are important for determining this behavior and for predicting the mobilization and transport of these suspensions in pipelines.

In newtonian fluids shear stress is linearly related to shear rate; however, the addition of solid particles to newtonian liquids generally results in non-newtonian behavior (shear stress is a non-linear function of shear rate); therefore, it is important to measure viscosity over a range of shear rates. Generally, the addition of solids to a liquid or to a slurry system results in higher viscosities and increased shear strength; therefore, rheological properties of a sample are generally measured at varying solid concentrations.

The rheological properties of each of the available research samples were determined by performing a stress sweep (steady state shear stress as a function of shear rate), an oscillatory stress sweep (shear stress as a function of shear rate at varying frequencies), and a shear strength measurement (shear stress as a function of time at a constant shear rate). For each research sample the stress sweep and oscillatory stress sweep were repeated at three solid concentrations (settled solids, 30% settled solids, and centrifuged solids). The samples were initially suspended in the KE Basin supernatant and allowed to settle for 3 days. The clear supernatant above the solids was removed and remaining material was analyzed as settled solids. The 30% settled solids sample was prepared by diluting the settled solids with supernatant at a supernatant to settled solids volume ratio of 0.7 to 0.3. The settled solids sample was centrifuged at 1000 x g for 1 hour to obtain centrifuged solids. It is anticipated these samples will encompass the expected range of solid concentrations that will be observed during canister sludge mobilization and slurry transport.

When the volume fraction of a suspension occupied by solids is larger than approximately 0.01, the particles increasingly enter the neighborhood of other particles and the interaction between particles plays a predominant role in determining the rheological properties of the system. In this range, increases in the particle volume fraction introduce deviations from Newtonian behavior (e.g., shear thinning, pseudoplasticity, and shear thickening) as well as increasing the viscosity of the system. Typically, the viscosity grows at an increasing rate as solids concentration is increased. With further increases in particle concentration, the interactive contact between the particles increases until a highly ordered structure is achieved. At this point, the system is referred to as a "solid" suspension and the viscosity of the system tends to infinity. At higher solid concentrations, the rheological properties of the suspension are characterized by the shear strength and viscoelastic moduli rather than viscosity.

<u>Experimental Procedure</u>--Steady state shear stress (τ) as a function of shear rate (γ) was measured on three solid concentrations for three KE canister research samples (96-06 M, 96-11 L, and 96-04 U/L). These analyses were performed on the Bohlin Controlled Stress (CS) rheometer (Silvers 1997b). A water bath was used to hold the samples at 25 °C during the analyses. A concentric cylinder geometry consisting of an inner cylinder 25 mm in diameter and outer cylinder 26.5 mm in diameter(0.75mm gap,C25 geometry,small cell) was used to perform these analyses on the settled solids and 30% settled solid samples. This narrow gap (0.75 mm) provides for a uniform shear field across the sample therefore, any radial shear-induced migration of particles due to the gradient in the shear rate is negligible. The centrifuged solid samples could not be poured; therefore, a cone and plate geometry consisting of a plate with a 40 mm diameter cone with a 4° pitch (CP4/40 geometry) was used to perform these analyses of the centrifuged solids.

<u>Steady State Shear Stress (τ) - Shear Rate (γ) Experimental Results</u>--From the τ and γ measurements, the experimental values of yield stress, critical shear rate, and the plastic viscosity of canister sludge samples at three solid particle concentrations were determined. The yield stress (τ_y) arises from the residual effect of the interaction potential and frictional interlocking between particles. Also, the yield stress corresponds to the transition from an elastic solid to a viscous fluid. The numerical value of the yield stress is determined by extrapolation of the linear portion of the τ - γ curve to $\gamma = 0$. The critical shear rate (γ_{crit}) is the shear rate above which the τ - γ behavior of the sample shows a linear relationship. Also, the critical shear rate (γ_{crit}) is the shear rate above which viscous stresses will rupture all the flocs in a flocculated suspension system. The plastic viscosity of a sample can be determined from the gradient of the linear τ - γ curve above the γ_{crit}. Examining the variation of shear stress with shear rate can reveal some

information about the extent of flocculation and floc strength. As flocs are destroyed by shear forces, the viscosity of the slurry is reduced. The plastic viscosity gives a measure of the hydrodynamic units that remain undisturbed above the γ_{crit}.

Sample 96-06 M--Figure I.9-1 shows the viscosity data as a function of shear rate (from approximately 0.10 to 400.0 s^{-1}) for Sample 96-06 M. The viscosity as a function of shear rate is presented in a log-log plot to illustrate the numerical magnitude of viscosity change over the range of shear rate and the solids concentration. The results indicate similar decreases in the viscosity with increasing shear rate for the "as-settled solid" and 30 vol% solid samples. Sample 96-06 M contained large particles that interfered with the gap spacing in the cone and plate geometry which was used for the centrifuged samples. As a result, the sample could not be properly loaded and the data was not collected for Sample 96-06 M after centrifuging this sample. The viscosity of the "as-settled solid" decreased from approximately 10 Pa.s at a shear rate of 0.1 s^{-1} to about 0.01 Pa.s at a shear rate of 400 s^{-1}. The viscosity of the "30 vol%" sample drops from approximately 50 Pa.s at shear rate of 0.1 s^{-1} to about 0.1 Pa.s at shear rate of 400 s^{-1}. In the case of the as settled solid concentration, initially the sample's viscosity is at about 10,0000 Pa.s that rapidly decreases to 0.1 Pa.s at shear rate of 400 s^{-1}. These results indicate that the viscosity of Sample 96-06 M is influenced by a change in the solids concentration as the solid concentration increases from 30% solids by volume to the as-settled solid concentration.

The plot of shear rate verses shear stress (see Figure I.9-2) indicates that the yield stress of this sample increases from approximately 1 Pa (at 30% by volume) to about 10 Pa for the as settled case. Thus, the as settled solids sample begins to flow when a shear stress approaching 10 Pa is applied. The critical shear rate (γ_{crit}) is about 60 s^{-1}. In general, at shear rates above the γ_{crit}, no further floc destruction occurs with increasing shear stress.

Sample 96-04 U/L--Figure I.9-3 shows the viscosity plotted as a function of shear rate (from approximately 0.10 to 400.0 s^{-1}) for the Sample 96-06 U/L. The plot of the viscosity as a function of shear rate indicates that the decrease in the viscosity at the solid's concentration of the "as-settled solid" and the centrifuged sample are comparable and the initial viscosity of the centrifuged sample at a shear rate of 0.1 s^{-1} is approximately the same as the viscosity of the as-settled samples at the same shear rate. The viscosity of the "as-settled solid" sample decreased from approximately 20 Pa.s at a shear rate of 0.1 s^{-1} to about 0.05 Pa.s at shear rate of 400 s^{-1}. The viscosity of the centrifuged layer drops from approximately 40 Pa.s at a shear rate of 0.1 s^{-1} to about 0.05 Pa.s at a shear rate of 400 s^{-1}.

The plot of shear rate verses shear stress (see Figure I.9-4) indicates that the yield stress of this sample increases from approximately 8 Pa (for the as-settled sample) to about 10 Pa for the centrifuged case. Thus, the sample begins to flow when a shearing force greater that 10 Pa is applied at the centrifuged solids concentration. The γ_{crit} is about 100 s^{-1}.

Sample 96-11 L--Figure I.9-5 shows the viscosity data as a function shear rate (from approximately 0.10 to 400.0 s^{-1}) for the Sample 96-11 L. The results indicate that the relative change in the viscosity of this sample at three solid particle concentration is similar. The viscosity decreases from approximately 100 Pa.s at a shear rate of 0.1 s^{-1} (centrifuged solids) to about 0.001 Pa.s (30 vol% settled solids) (viscosity of water at 25 °C) at a shear rate of 400 s^{-1}. These results indicate that the viscosity of the "as-settled solid" sample and centrifuged sample are 10 to 100 times more viscous than the 30% by volume sample.

The plot of shear rate verses shear stress (see Figure I.9-6) indicates that the yield stress for Sample 96-11 L increases from approximately 0.2 Pa (at 30% by volume) to about 10 Pa for the centrifuged case. The sample begins to flow when a shearing force greater that 10 Pa is applied at the centrifuged solid concentration. The γ_{crit} is about 140 s^{-1}.

Shear Strength Measurements--The shear strength of Samples 96-04 U/L, 96-06 U/M, 96-06 M, 96-06 M/L, 96-06 L, and 96-11 U/L were measured using a shear vane and a Haake M5 measuring head (Silvers 1997b). In these experiments the sludge layers from the graduated cylinders were transferred to containers and left undisturbed for 2 weeks. For this discussion the shear strength is defined as the maximum stress required to cause failure (deformation) of the sludge in shear mode. The measurements were made by monitoring the shear stress of the sample as a function of time at a constant shear rate. The shear rate approaches the theoretical shear stress value measured at a shear rate of zero. The measured shear strength of these samples are reported in Table I.9.1.

The shear strength of a material is strongly dependent on the sample history. The canister sludge samples were subjected to various sample handling disturbances during the course of characterization. These disturbances include the initial mobilization of the canister sludge samples by the sampling device, removal of sample from the shipping container to the graduated cylinder, resuspension of the sludge layer by gas sparging and transferring the settled sludge layer to the measuring container. The interaction between solid particles in the sludge samples were disturbed in each of these processes. Since these samples were disturbed prior to the shear strength measurements it is hard to predict the shear strength of actual KE canister sludge upon mobilization. In fact the actual shear strength of the KE canister sludges in the basin may be considerably higher than these reported values. The shear strength results reported in this section provide an estimate of the lower bound of the shear strength for the KE canister sludges in the basin and provide data to estimate the shear strength of the material in the processes to clean the Hanford K Basins.

I.10 ZETA POTENTIAL MEASUREMENT

The quantity known as zeta potential can have a major effect on the behavior of small particles suspended in a low-salt-concentration liquid. In a suspension system of fine solid particles of roughly less than 10 microns the magnitude of the surface forces acting on the particles dominates the body/bulk forces. These surface forces can be quantified by measuring the zeta potential that represents the electrostatic charge on the surface of solid particles.* In a suspension, the surface of solid particles can be charged by adsorption of the dissolved ions existing in the aqueous phase. The ions of the opposite charge to the particle's charge will adsorb to the particle surface and create a layer of charge near the surface.

The zeta potential provides a measure of the strength of the repulsive forces between the particles. A strongly positive or negative zeta potential indicates that the repulsive forces between particles are strong. When the repulsive forces are strong enough to overcome the interparticle attractive forces (van der Waals and London dispersion forces), the particles will remain segregated from each other. Otherwise, the attractive forces between the particles dominate the particles' behavior, and the solid particles will tend to aggregate and form larger particles. Depending on the conditions of the suspension (i.e., the salt concentration in the liquid phase, the type of salt, the PSD and particle shapes, etc.), there can be a particle-to-particle distance at which the attractive forces and electrostatic repulsive forces balance such that the particles have a tendency to form loosely aggregated clumps known as floc. This process is called flocculation.

In dealing with the complex suspension that will be formed during the KE canister sludge washing, hydrocyclone solid-liquid separation and fine solid-liquid filtration, the separation of fine solid particles from the slurry can potentially be improved by the "floc" formation process. Therefore, it would be desirable to maximize the flocculation process. In this context the concept of zeta potential arises as a valuable measurement. The larger the zeta potential (with + or -), the more stable will be the suspension. As zeta potential approaches zero, the suspension is less stable and will tend to flocculate. At the isoelectric point, the zeta potential is zero, and the potential energy of repulsion between the particles is minimum. By measuring the zeta potential as a function of pH, the relative stability of the suspension with respect to flocculation can be evaluated.

<u>Experimental Procedure</u>--The Brookhaven Zetaplus instrument was used to measure zeta potential. In these experiments, the zeta potential measurement of selected KE canister sludge samples was determined as a function of pH using the acid-base titration. Initially, a small amount of sludge sample was suspended in deionized water. Each of these samples was divided in half, and one half was titrated with sodium hydroxide to a basic condition while measuring zeta potential. The other half was titrated with nitric acid to acidic conditions while measuring zeta potential (Silvers 1997a).

<u>K East Canister Sludge Zeta Potential Results</u>--The zeta potential, the pH values, and the conductivity of the as-received untitrated samples are presented in Table I.10-1.

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*The zeta potential is the effective electrostatic charge at the shearing or slipping plane between the bulk liquid and the envelope of liquid (i.e., water) that moves with the particle. The actual charge on the surface is different from the zeta potential, but, to some extent, the zeta potential is comparable to the surface potential.

The measured conductivity of all the as-received untitrated samples is low (6 to 26 μS), except the measured conductivity of Sample 96-06 M, which shows a higher sample conductivity. The conductivity is a measure of the electrolyte concentration in the solution. Higher conductivity decreases the double layer and particle velocities.

The measured zeta potential for all the research canister sludge samples as a function of pH is shown in Figure I.10-1. The results for the carboy solids exhibit a large scatter in the zeta potential measurement. Besides the carboy sample, the rest of the fueled canister sludge samples (Samples 96-04 U/L, 96-04 L, 96-06 M, and 96-06 L) show a positive zeta potential at the acidic conditions (approximately pH 3.0) and a negative zeta potential value at the basic condition (approximately pH 11.0). The zeta potential measurements of the fueled canister sludge samples (Samples 96-04 U/L, 96-04 L, 96-06 M, and 96-06 L) reveal that over the pH range of interest, from pH 6 to pH 11, the surface of the particles are negatively charged or not charged (zero zeta potential). Over the pH range of 6 to 11, the zeta potential of the fueled samples is approximately between 0.0 to -100 mV. The zeta potential of the Sample 96-04 U/L approaches its isoelectric point (zero zeta potential) at about a pH of 6.0. At this condition, the repulsive forces between the particles are zero and a suspension of the 96-04 U/L particles that allows the particles to aggregate is unstable.

At the high-acid concentration (approximately pH 3.0) and the high basic condition (approximately pH 11.0), the surface of the fueled samples continues to adsorb ions, and the particles show a high mobility. In addition, the zeta potential of the fueled canister samples (Samples 96-04 U/L, 96-04 L, 96-06 M, and 96-06 L) varies approximately from zero to -40 mV at pH 6 to about -80 mV at pH 11. From pH 6 to pH 11, the zeta potential of the nonfueled sample (Sample 96-11) is a change over a small range. The zeta potential of the Sample 96-11 varies from approximately 0 mV at pH 6 to -25 mV at pH 11. The uranium concentration of the sludge for Sample 96-11 was only about 9.3 wt%. This demonstrates that the particles are less receptive to adsorption of ions and show a smaller zeta potential variation compared to the fueled sludge samples. About 50 wt% of Samples 96-04 L and 96-04 U/L is likely made of uranium oxides or uranium hydroxide, and the uranium content of Samples 96-06 M and 96-06 L is about 80 wt%.

The zeta potential measurements for the research canister sludge samples and the floor and Weasel Pit sludge samples as a function of pH are shown in Figure I.10-2. The zeta potential of the "nonfueled" Sample 96-11 shows the same trend as the zeta potential of the floor sludge and the Weasel Pit sludge. However, the isoelectric point of Sample 96-11 occurs at about pH 6, which indicates that the surface the particles are predominantly coated with a more basic type oxide. The magnitude of the zeta potential (positive or negative deviation from zero) of the "fueled" canister samples (Samples 96-04 U/L, 96-04 L, 96-06 M, and 96-06 L) is generally higher than the floor sludge and the Weasel Pit sludge results. These results indicate that the concentration of the charge on the surface of the "fueled" canister sludge particles is higher and the "fueled" canister sludge particles tend to be more receptive to the adsorption of ions.

Table I.4-1. K East Sample Information.

Container Number	K East Cubical Position	Canister Barrel	Number of Fuel Elements	Volume of Settled Sludge in Samples (ml)
96-01	1845	East	5	265
96-04	2711	East	7	260
96-05	3128	West	6	285
96-06	5465	West	6	315
96-08	2350	East	7	422
96-09	4638	East	0	120
96-11	6073	West	0	220
96-13	5055	West	6	465
96-15	6070	West	7	160

Table I.5-2. Layers from the Research Samples.

As Observed in Settled Sludge:

Sample	Layer	Estimated Volume Percent
96-04	Upper	70
	Lower	30
96-06	Upper	5
	Middle	53
	Lower	42
96-11	Upper	7
	Lower	93

As Split for Analyses:

Sample	Layer	Sampling Method	Label	Mass (g)
96-04	Upper	Syringe with glass pipet	96-04 U	64.93
	Interface	Syringe with glass pipet	96-04 U/L	79.87
	Lower	Vacuum transfer	96-04 L	177.82
96-06	Upper	Syringe with glass pipet	96-06 U	9.62
	U/M interface	Syringe with glass pipet	96-06 U/M	81.89
	Middle	Syringe with glass pipet	96-06 M	145.31
	M/L interface	Syringe with glass pipet	96-06 M/L	117.22
	Lower	Vacuum transfer	96-06 L	379.08
96-11	Upper	Syringe with glass pipet	96-11 U	31.72
	Interface	Syringe with glass pipet	96-11 U/L	44.64
	Lower	Vacuum transfer	96-11 L	143.07

Table I.6.1-1. Aperture Sieve Summary.

Tyler Sieve Size	U.S.A. Equivalent	Aperture Opening (mm)
6	6	3350
8	8	2360
14	16	1180
24	25	710

Table I.6.1-2. Sieving Analysis Particle Size Distribution Reported as Weight Percent Dry Solid Basis.

Sample	Larger than 3350 but less than 6350 μm	2360 to 3350 μm	1180 to 2360 μm	710 to 1180 μm	Smaller than 710 μm
96-04 L	0	12	22	9	56
96-06 M	5	6	20	11	58
96-06 L	3	3	15	22	58
96-11 L	0	0.05	0.7	1	98

Table I.6.2-1. Particle Size Analysis Sampling for Microtrac Instrument.

Sample	Layer	Sample Condition	Mass (g)
96-04	Upper (U)	Not analyzed (sample was used for other analyses)	N/A
	Interface (U/L)	Wet sample (replaced Sample 96-04 U)	0.33
	Lower (L)	Very dry	0.24
96-06	Upper (U)	Not analyzed (sample was used for other analyses)	N/A
	Interface (U/M)	Interface layers not analyzed	N/A
	Middle (M)	Sticky sludge, slightly moist	0.23
	Interface (M/L)	Interface layers not analyzed	N/A
	Lower (L)	Very dry	0.26
96-11	Upper (U)	Not analyzed (sample was used for other analyses)	N/A
	Interface (U/L)	Interface layers not analyzed	N/A
	Lower (L)	Wet	0.4

Table I.6.2-2. pH of the Particle Size Analysis Suspensions.

Sample	Layer	pH
96-04	Interface (U/L)	5.74
	Lower (L)	5.51
96-06	Middle (M)	3.25 and 5.5*
	Lower (L)	5.53
	Carboy solids	5.59
96-11	Lower (L)	5.35

*The middle layer from sample 96-06 had a pH of 3.25. This low pH was most likely due to a small amount of nitric acid remaining in the reservoir. The lens of the particle size analyzer was washed with 0.5 M nitric acid and flushed with 3 volumes of distilled and demineralized water before beginning the particle size analyses. A small amount of acid may have remained in the system following this washing procedure. The duplicate run of this sample was done at a pH of 5.5.

Table I.6.2-3. Mean Particle Size of the K Basin Research Samples.

Sample	Flow Rate (ml/s)	Circulation Time (min.)	Sonication	Mean Diameter (μm)	
				Number Dist.	Volume Dist.
96-04 U/L	60	2	None	1.49	16.3
		8	25 W, 120 s	0.91	11.7
		16	40 W, 300 s	0.37	9.22
		2	None	3.79	27.6
		25	None	2.74	20.6
		35	25 W, 90 s	1.48	17.2
	40	2	None	6.02	142
		6	40 W, 90 s	2.01	35.5
	70	10	None	2.01	36
		14	40 W, 90 s	1.64	31.4
96-04 L	60	2	None	6.26	194
		7	25 W for 120 s	1.93	48.6
		14	40 W for 300 s	1.1	39.2
	40	2	None	1.43	212
		7	40 W, 90 s	0.76	76.9
	70	10	No additional	0.69	59
		16	40 W, 90 s	0.54	227
96-06 M	60	2	None	0.84	21.2
	40	2	None	2.86	82.9
		6	40 W, 90 s	1.12	36.9
	70	10	No additional	1.15	37.2
		16	40 W, 90 s	0.54	32.4
96-06 L	60	2	None	1.17	321
		8	25 W for 120 s	0.44	37.7
		15	40 W for 300 s	0.36	31.5
96-06 Carboy Solids	60	2	None	0.63	21
		9	25 W for 120 s	0.32	20.2
		16	40 W for 300 s	0.32	21.5
96-11 L	60	2	None	0.99	25.8
		6	25 W for 120 s	0.26	15.1
		14	40 W for 300 s	0.27	11.5
	40	2	None	1.36	38.4
		7	40 W, 90 s	0.49	27
	70	10	No additional	0.63	28.7
		16	40 W, 90 s	0.32	22.5

Table I.7-1. Mean Aspect Ratio and Shape Factor of K East Canister Sludge Research Samples.

Sample	Mean Aspect Ratio	Mean Shape Factor
96-04 L	0.72	0.65
96-06 M	0.71	0.65
96-06 L	0.71	0.66
96-11 L	0.60	0.58

Table I.8-1. Density Data for K East Canister Sludge and Supernate.

Sample	Density (g/ml)			
	Supernatant	Settled Solids	Centrifuged Solids	Dried Solids
96-01	1.000	2.09	2.56	NM
96-04	1.000	1.21	NM	NM
96-04 U/L	1.000	NM	1.32	4.68*
96-04 L	1.000	NM	1.95	4.76*
96-05	0.990	2.34	2.62	NM
96-06 U	NM	1.42	2.51	NM
96-06 M	NM	NM	2.36	6.90*
96-06 L	NM	NM	3.71	7.88*
96-08	0.992	1.18*	1.53	NM
96-09	0.994	1.08*	1.20	NM
96-11	1.001	0.99	NM	NM
96-11 U	1.001	1.06*	1.27*	NM
96-11 L	1.001	NM	1.38	3.49*
96-13	0.983	2.46	4.52	NM
96-15	0.997	1.84	2.82	NM

*Average of two measurements.
NM = Density was not measured for this sample.

Table I.8-2. Calculated Uranium Concentration in Canister Sludge.

Sample	Uranium* Content (wt% of Dried Sludge)
96-01	82
96-04 L	54
96-05	88
96-06 M	83
96-06 L	84
96-08	40
96-09	8.8
96-11 L	9.3
96-13	82
96-15	81

*Four different techniques were used to measure uranium content. Here the value from laser fluorescence was used. A more complete listing of composition is given in Appendices C, D, and E.

Table I.8-3. Theoretical Reference Densities for Components in K East Canister Sludge as Determined by X-Ray Diffraction and Gas Generation.

Name	Chemical Formula	Theoretical Density (g/ml)
Uraninite	UO_2	10.95[1]
Uraninite	U_4O_9	8 to 11[1]
Uranite	U_3O_7	11.32[2]
Metaschoepite	$UO_3 \cdot 2H_2O$	4.87
Schoepite	$UO_3 \cdot 2H_2O$	4.87
Uranium metal[3]	U	19.05[1]
Zirconium metal	Zr	6.51[1]
Zirconium hydride	$ZrH_{1.66}$, ZrH_2	5.63[4,5]
Calcium aluminum oxide formate hydrate	$C_2H_2CaO_4 \cdot 3CaO \cdot 11H_2O$	6.49[1]
Quartz	SiO_2	2.66[1]
Maghemite	Fe_2O_3	4.9 (hematite)
Magnetite	$FeFe_2O_4$	5.18[1]
Chromite	$FeCr_2O_4$	4.97[1]
Geothite	FeO(OH)	4.28[1]
Lepidocrocite	FeO(OH)	3.96

[1] David R. Linde, Handbook of Chemistry and Physics, CRC Press Ann Arbor, 1993.

[2] Joseph J. Katz and Eugene Rabinowitch, The Chemistry of Uranium, McGraw-Hill Book Company, Inc., New York, New York, 1951.

[3] Included in table based on gas generation results (Appendix B).

[4] Reactor Handbook Vol. I "Material" Interscience Publishers New York, p. 1069, 1960.

[5] R. D. Scheele and L. L. Burger, "Zirconium Hydride as a Storage Medium for Tritium," BNNL-2083, 1976.

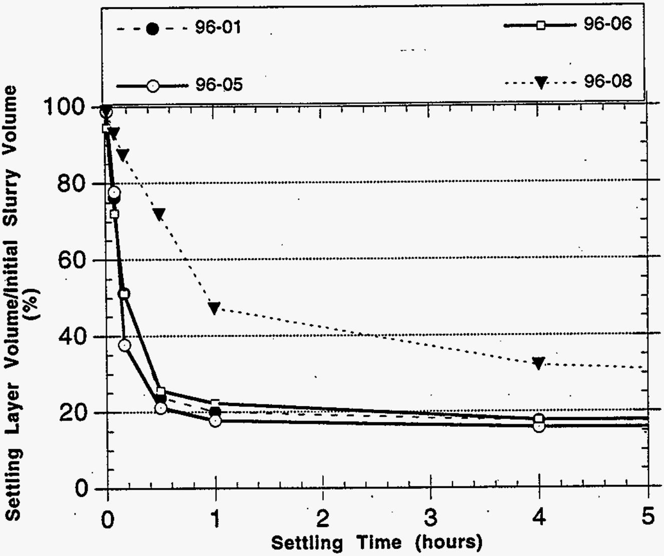
Table I.9-1. Measured Shear Strength in Units of Pascal.

Sample	Run 1 (Pa)	Run 2 (Pa)	Averaged (Pa)
96-04 U/L	<100	<100	<100
96-06 U/M	230	170	200
96-06 M	170	130	150
96-06 M/L	430	500	460
96-06 L	470	--	470
96-11 U/L	130	130	130

Table I.10-1. Zeta Potential, pH, and the Conductivity of As-Received Untitrated K East Canister Sludge Samples.

Sample	pH	Zeta Potential (mV)	Conductivity (μS)
96-04 U/L	2.1	-25.9	23.1
96-04 L	5.51	6.7	19.9
96-06 M	3.25	7.42	519.5
96-06 L	5.53	4.1	25.8
96-11 L	8.29	-13.2	21.4
Carboy solids	5.59	-9.6	6.08

Figure I.5-1. Settling Behavior of K East Canister Sludge Samples 96-01, 96-05, 96-06, and 96-08.



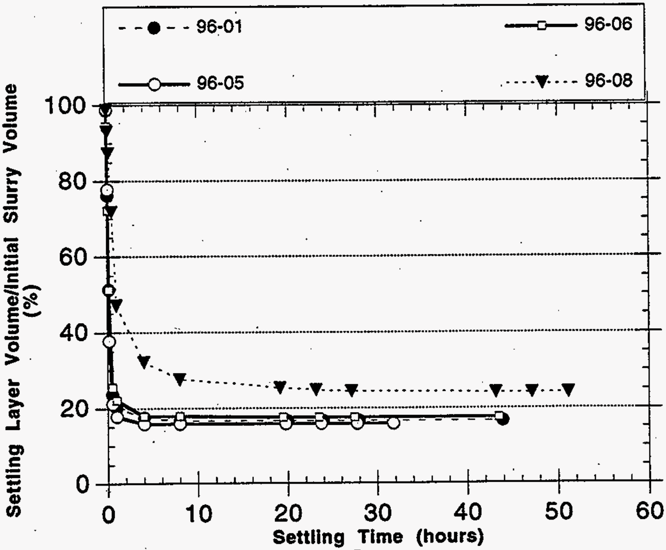
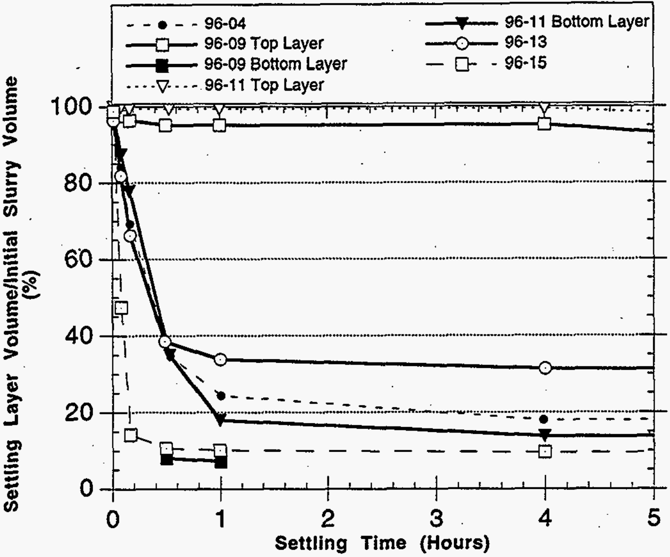


Figure I.5-2. Settling Behavior of K East Canister Sludge Samples 96-04, 96-09, 96-11, 96-13, and 96-15.



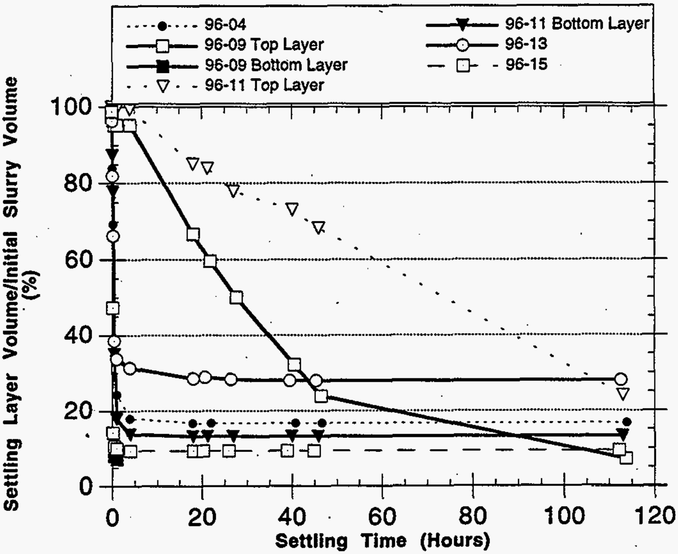


Figure I.5-3. Plot of Settling Rate as a Function of Suspension Interface Volume.

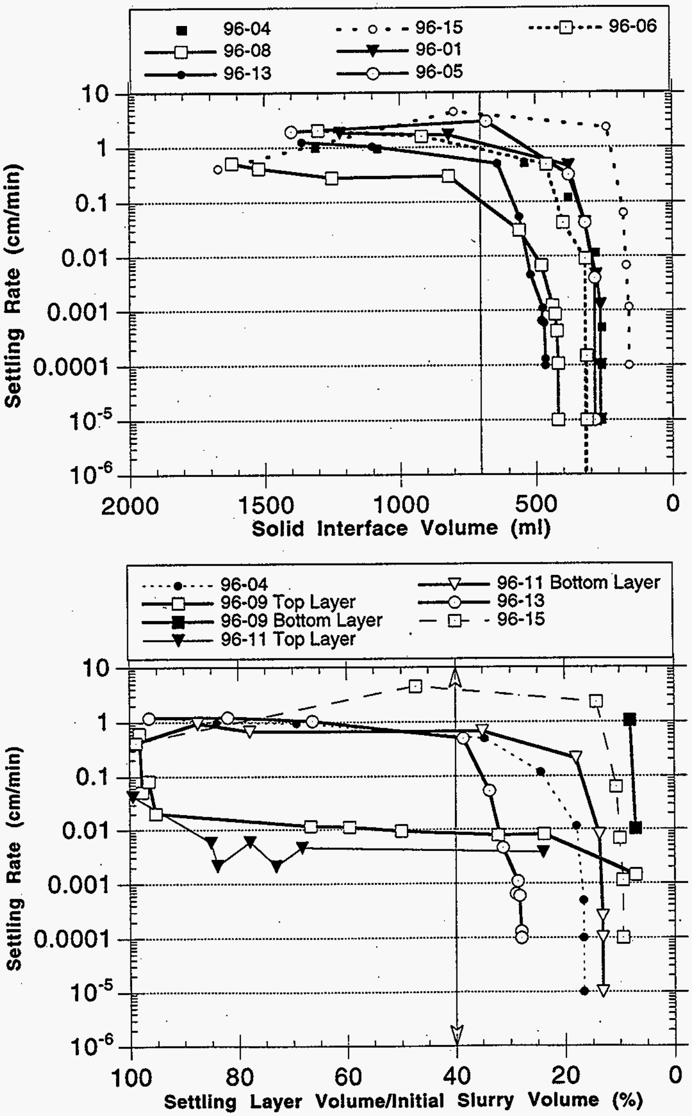


Figure I.5-4. Photograph of Settled Sludge Layer for Canister Sample 96-01.

96-01

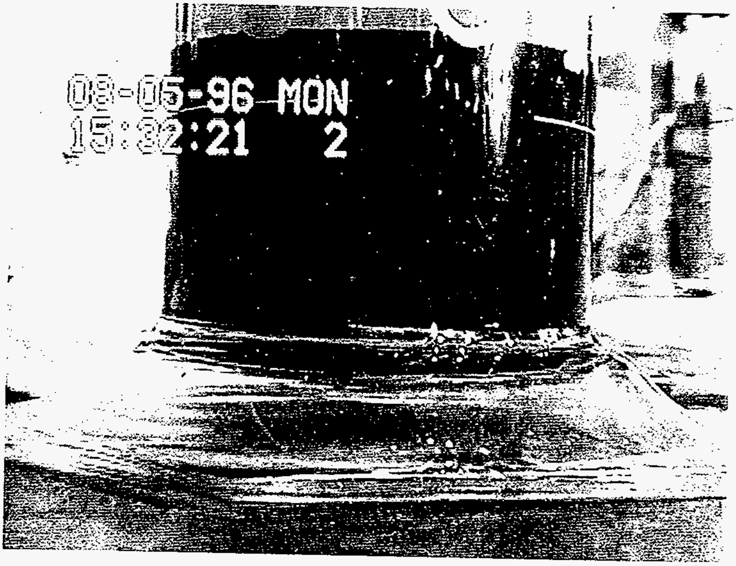


Figure I.5-5. Photograph of Settled Sludge Layer for Canister Sample 96-04.

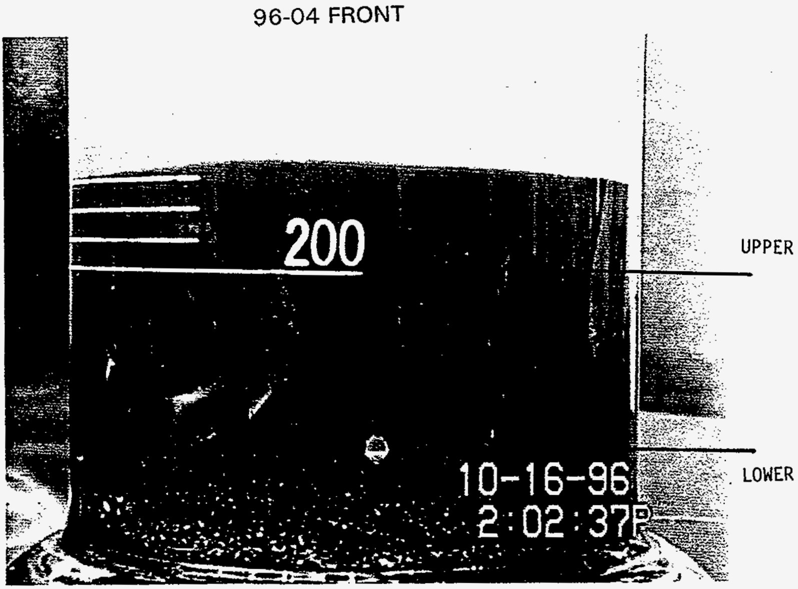
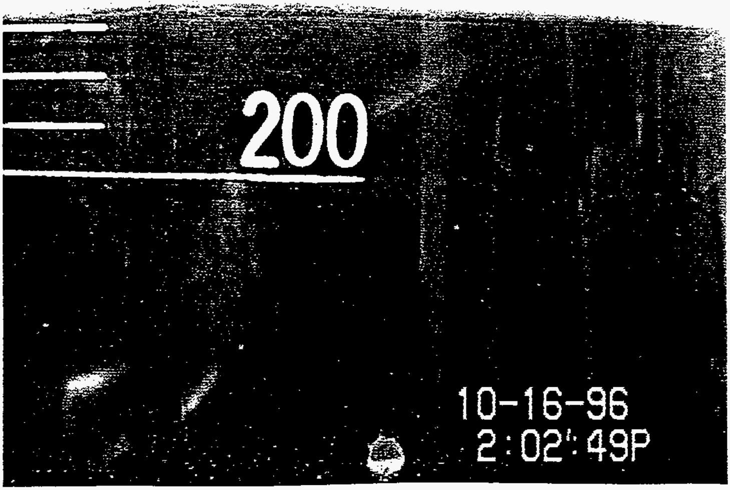


Figure I.5-6. Photograph of Settled Sludge Layer for Canister Sample 96-04.

96-04 FRONT - UPPER REGION



96-04 FRONT - GRANULAR REGION AT BOTTOM

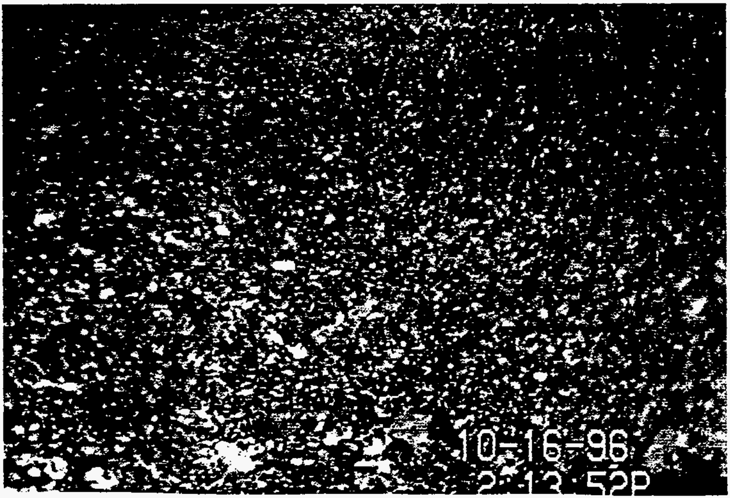


Figure I.5-7. Photograph of Settled Sludge Layer for Canister Sample 96-05.

96-05

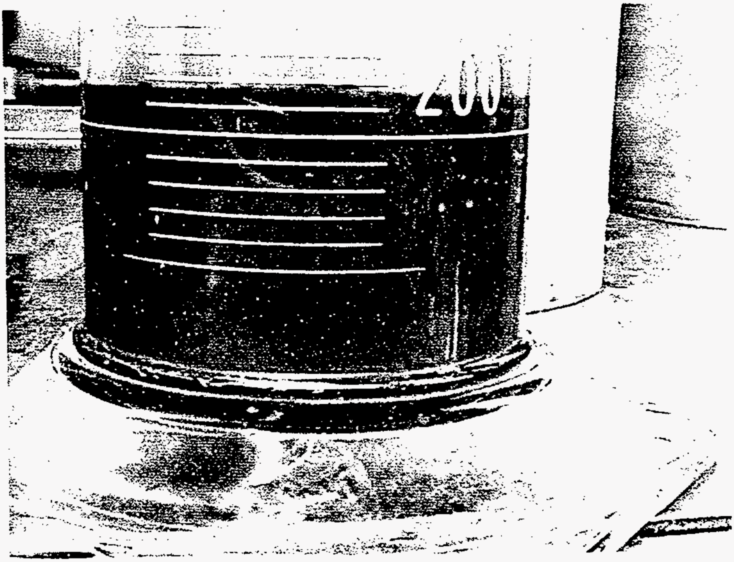
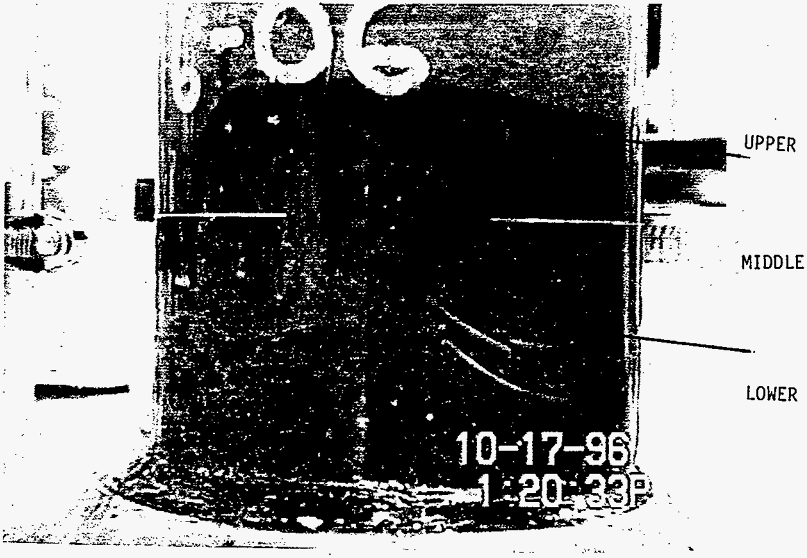


Figure I.5-8. Photograph of Settled Sludge Layer for Canister Sample 96-06.

96-06 FRONT



96-06 FRONT - PARTICLES AT 200 cc MARK

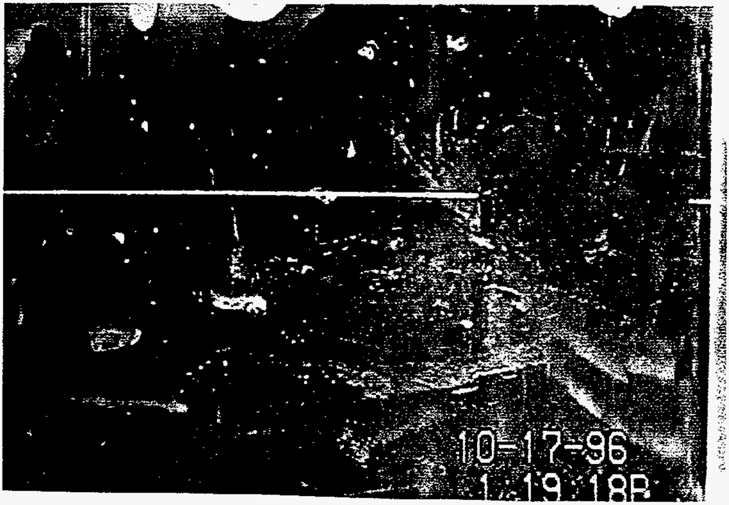
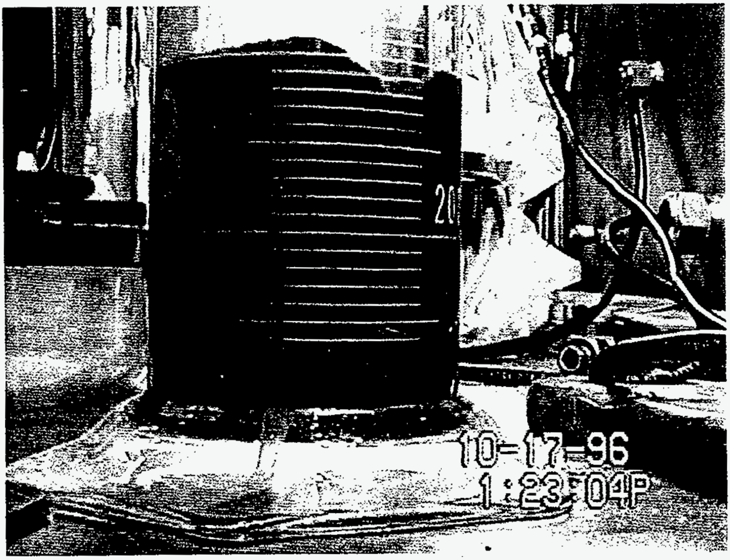


Figure I.5-9. Photograph of Settled Sludge Layer for Canister Sample 96-08.

96-08 FRONT



96-08 FRONT - LIGHT TOP SLUDGE LAYER SLOUGHING INTO CAVITY

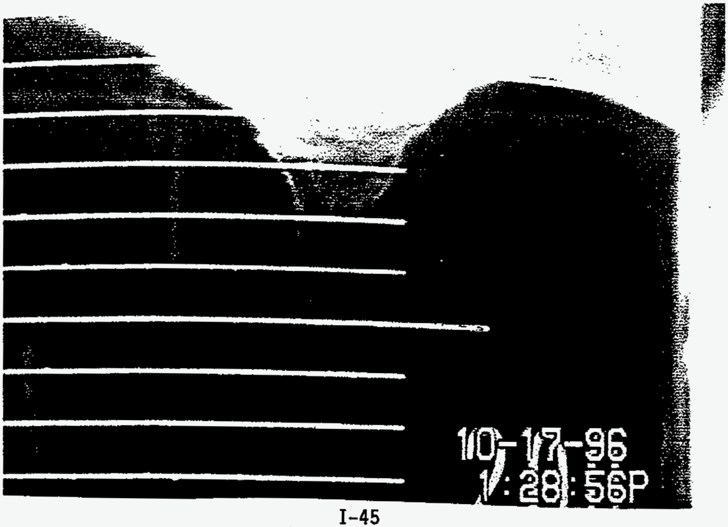
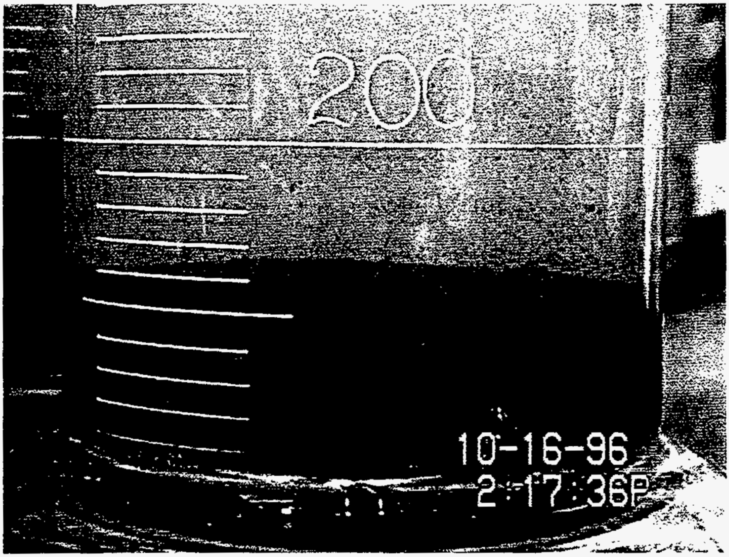


Figure I.5-10. Photograph of Settled Sludge Layer for Canister Sample 96-09.

96-09 FRONT



96-09 BACK

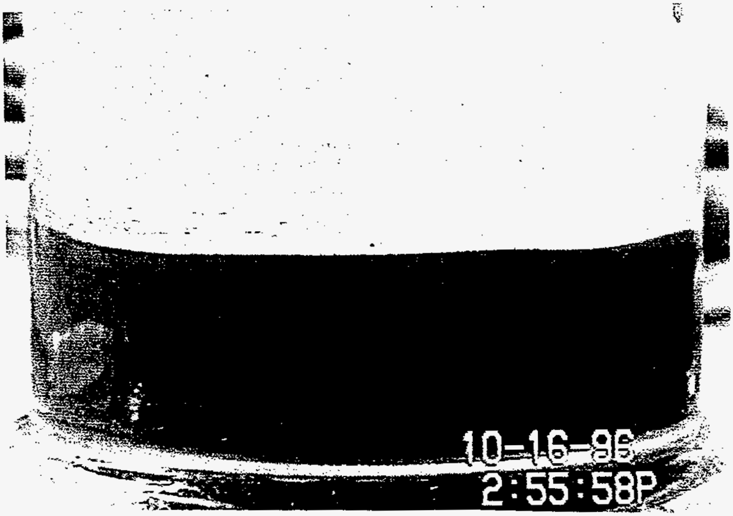
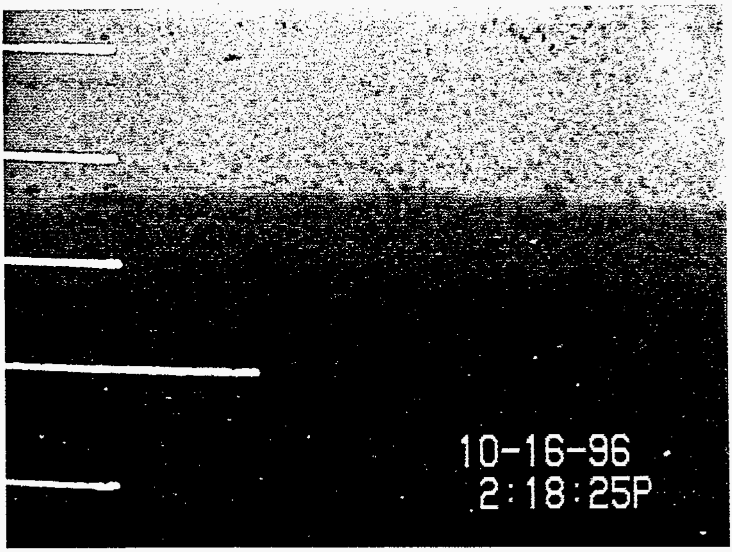


Figure I.5-11. Photograph of Settled Sludge Layer for Canister Sample 96-09.

96-09 FRONT - UPPER REGION



96-09 FRONT - BOTTOM REGION

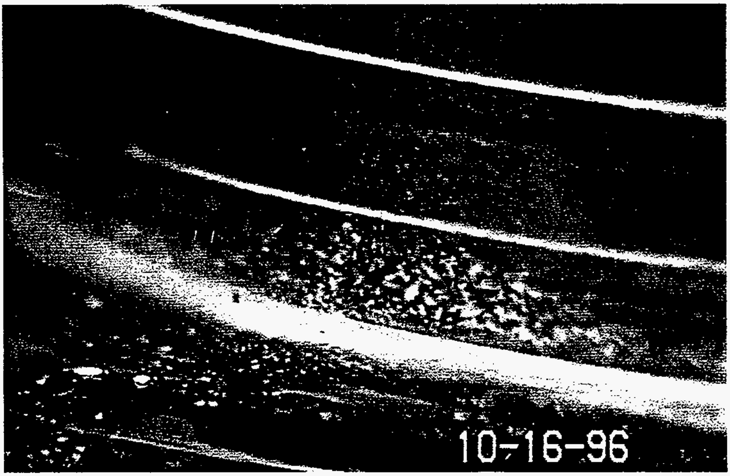


Figure I.5-12. Photograph of Settled Sludge Layer for Canister Sample 96-11.

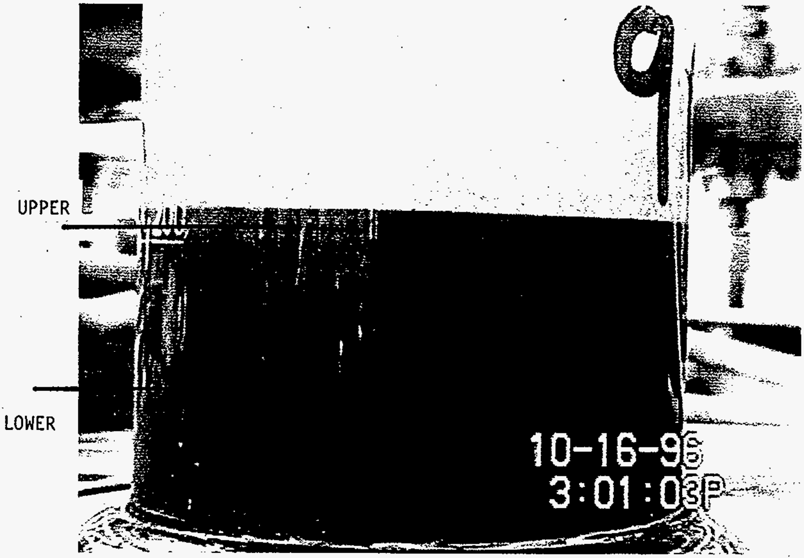
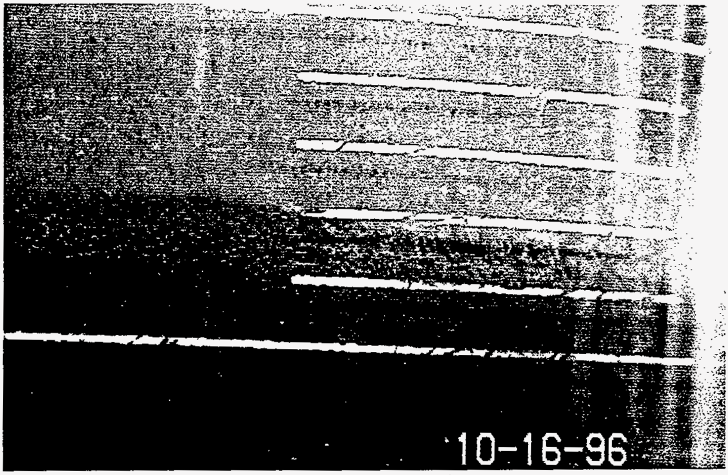


Figure I.5-13. Photograph of Settled Sludge Layer for Canister Sample 96-11.

96-11 FRONT - UPPER REGION



96-11 FRONT - BOTTOM REGION

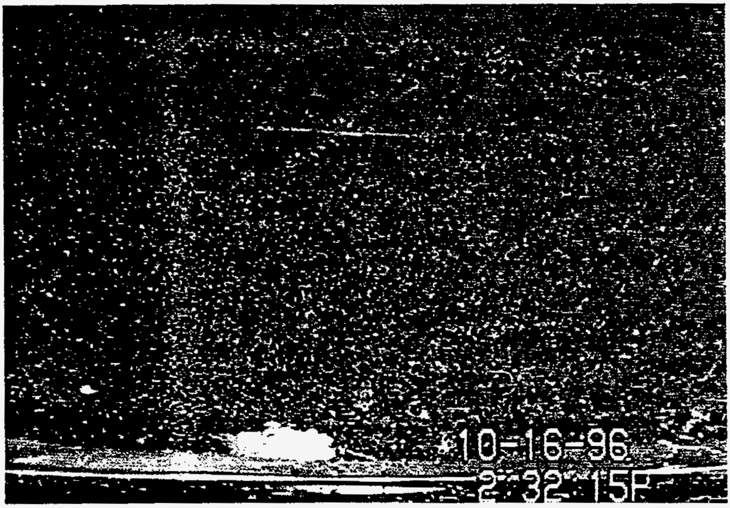


Figure I.5-14. Photograph of Settled Sludge Layer for Canister Sample 96-13.

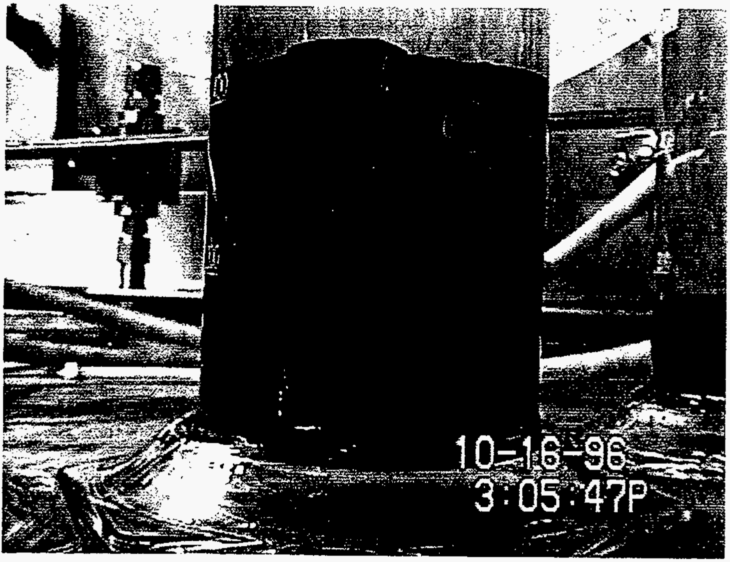
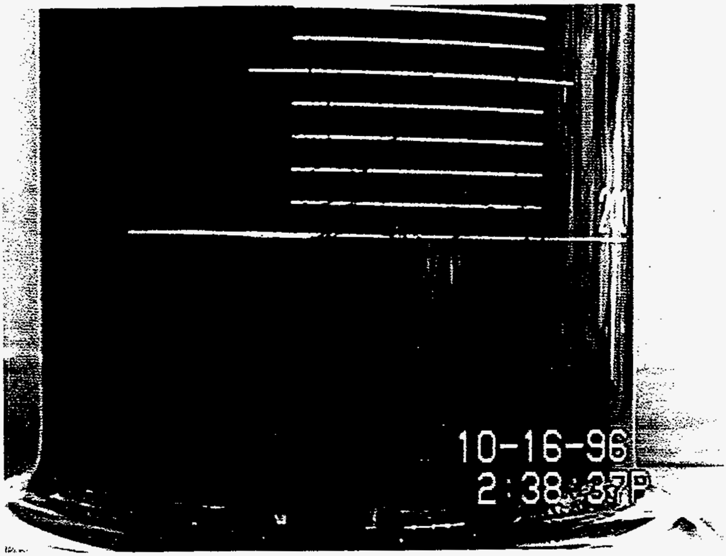
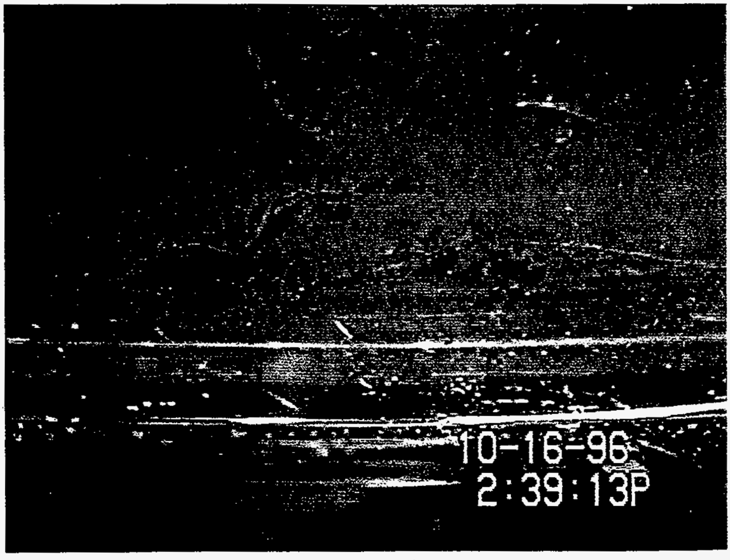


Figure I.5-15. Photograph of Settled Sludge Layer for Canister Sample 96-13.



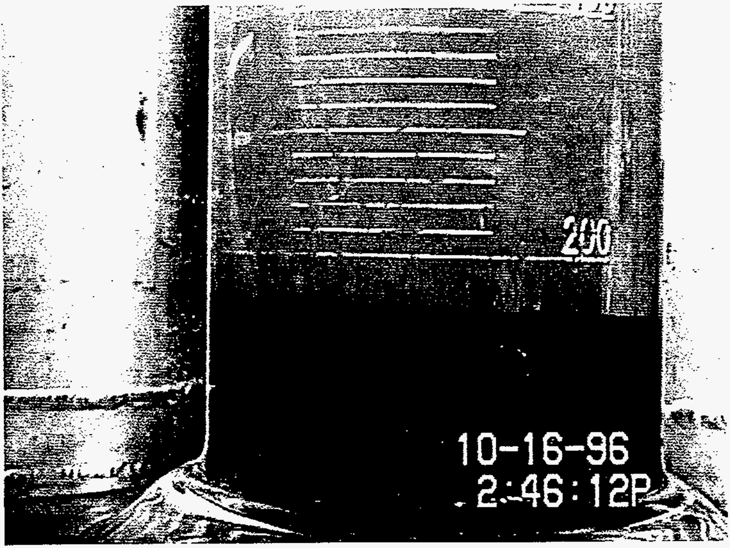
96-13 FRONT - LOWER 300 cc



96-13 FRONT - BOTTOM REGION

Figure I.5-16. Photograph of Settled Sludge Layer for Canister Sample 96-15.

96-15 FRONT



96-15 BACK

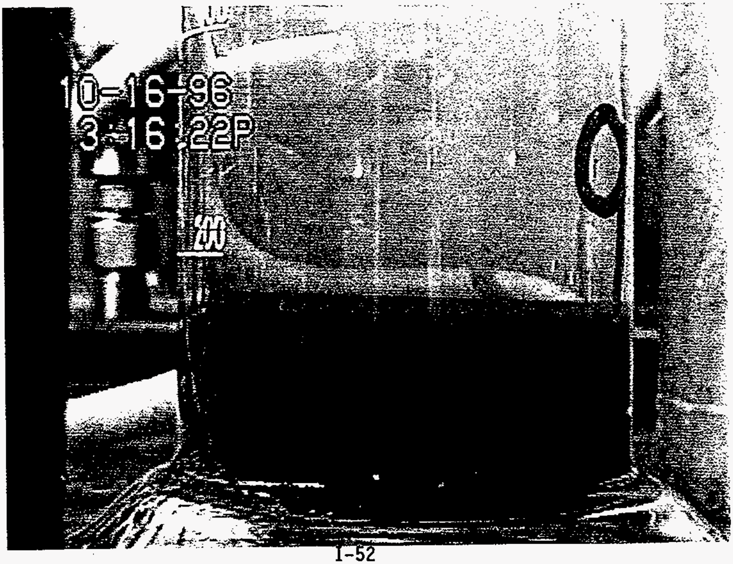


Figure I.6.1-1. Sample 96-06 M on the Tyler 6 Sieve After Rinsing. (Sieve openings are 3.3 mm)

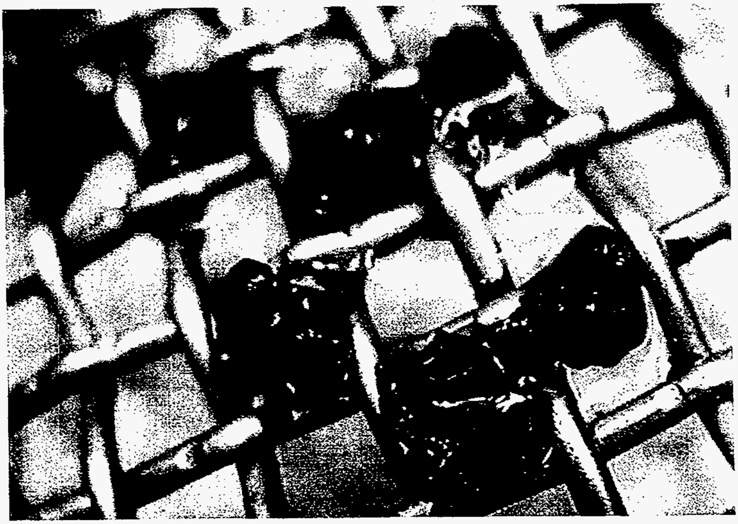


Figure I.6.1-2. Sample 96-06 M on the Tyler 8 Sieve After Rinsing. (Sieve openings are 2.36 mm)

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Figure I.6.1-3. Sample 96-04 L on the Tyler 14 Sieve After Rinsing. (Openings are 1.18 mm)

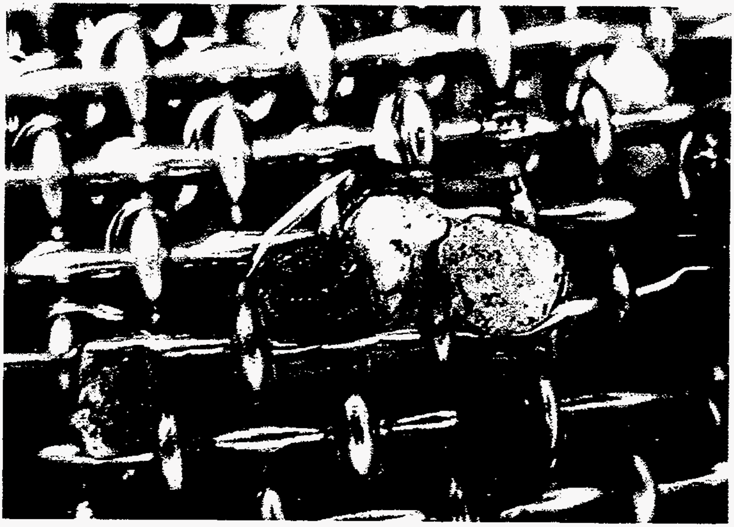

Figure I.6.1-4. Sample 96-04 L on the Tyler 24 Sieve After Rinsing. (Openings are 0.710 mm)

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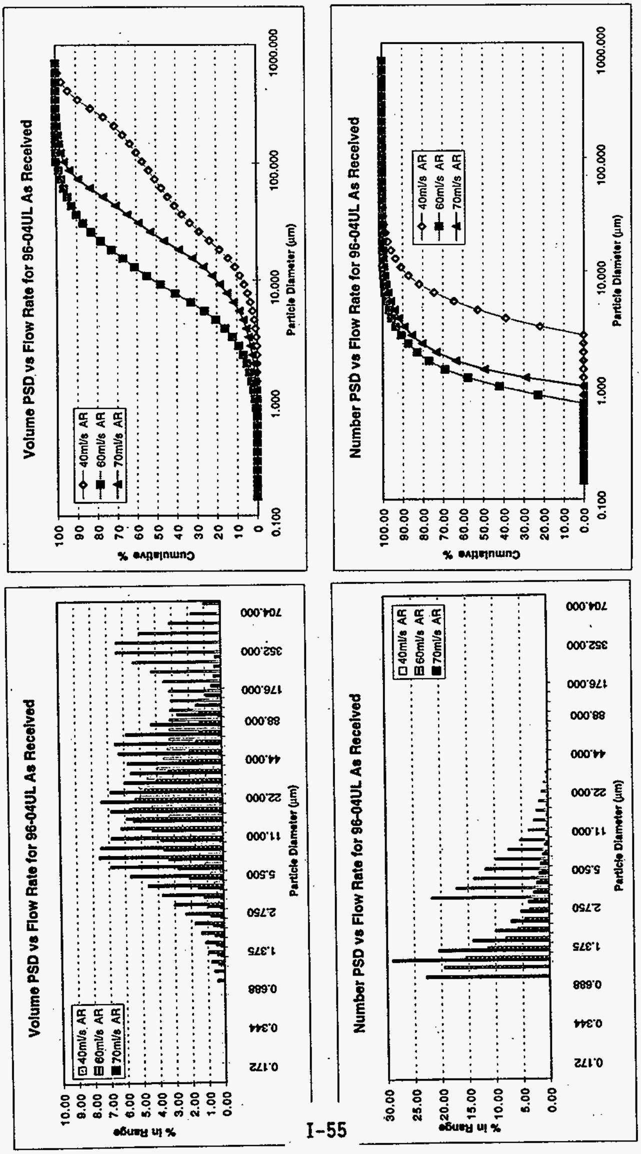
Figure I.6.2-1. Particle Size Versus Flow Rate for Sample 96-04 U/L. ("AR" means as-received)



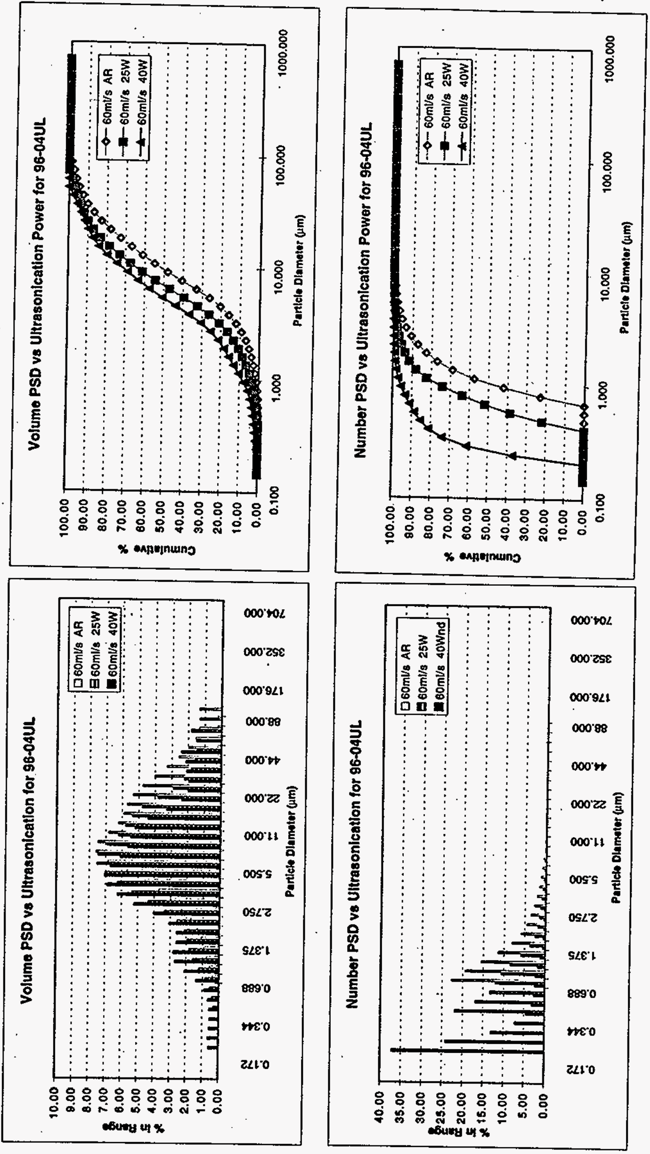
Volume PSD vs Flow Rate for 96-04UL As Received

Volume PSD vs Flow Rate for 96-04UL As Received

Number PSD vs Flow Rate for 96-04UL As Received

Number PSD vs Flow Rate for 96-04UL As Received

Figure I.6.2-2. Particle Size Versus Sonification Power for Sample 96-04 U/L.



40ml/s for 90 s; 70ml/s for 90 s; 60ml/s for 300 s

40ml/s for 90 s; 70ml/s for 90 s; 60ml/s for 300 s

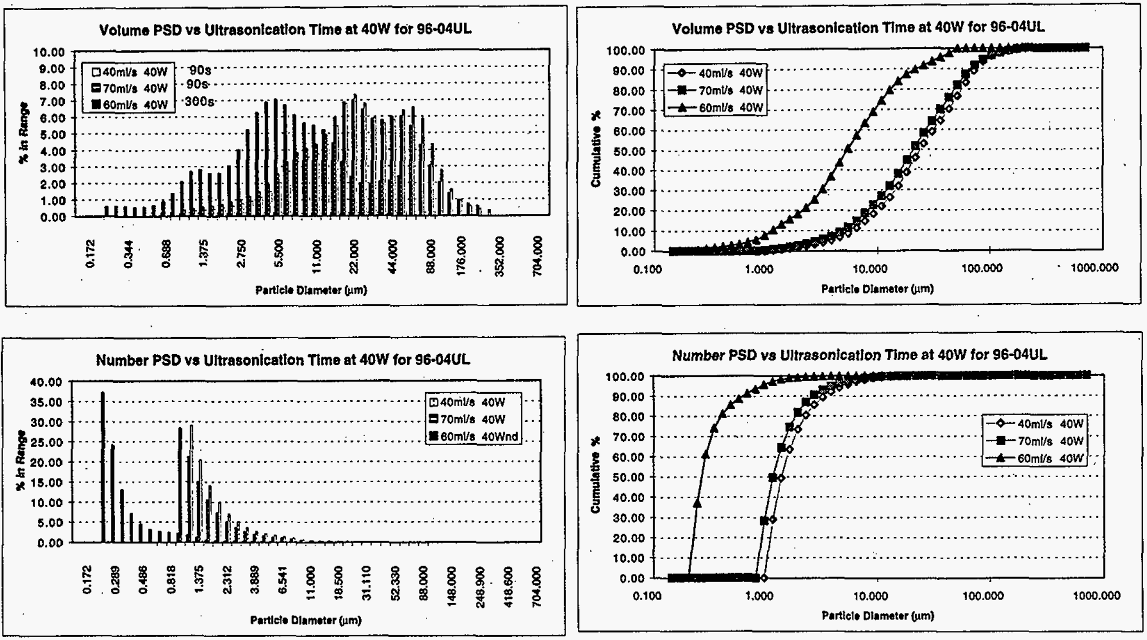


Figure I.6.2-3. Particle Size Versus Sonification Time for Sample 96-04 U/L.

Figure I.6.2-4. Particle Size Distribution Versus Sonification for Sample 96-04 U/L.

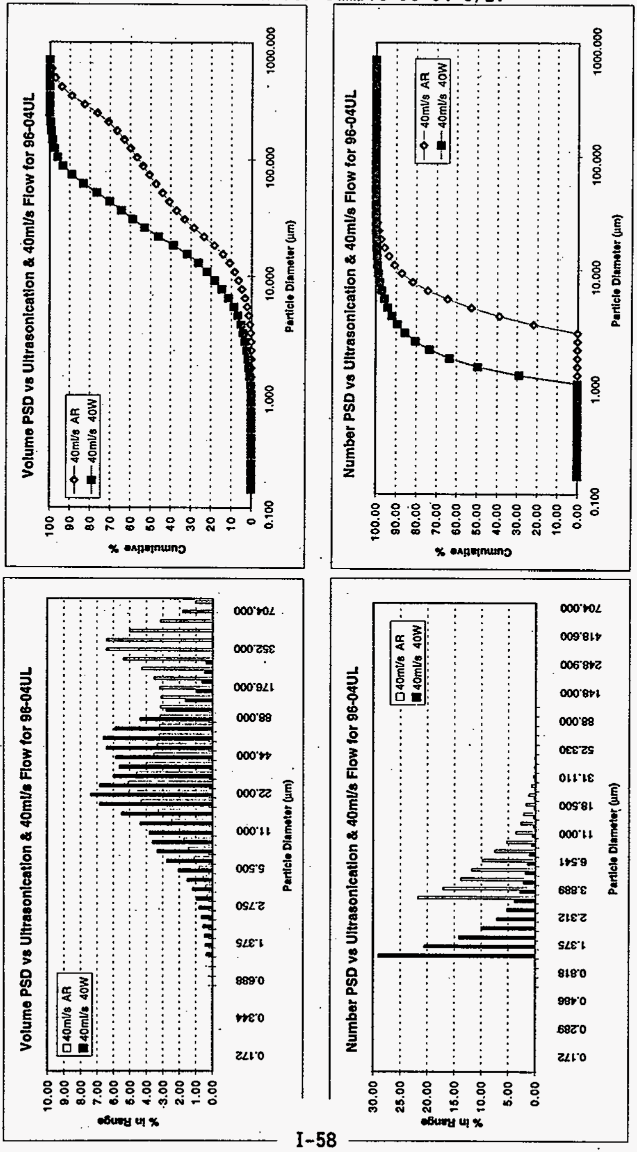


Figure I.6.2-5. Particle Size Distribution Versus Sonification for Sample 96-04 U/L.

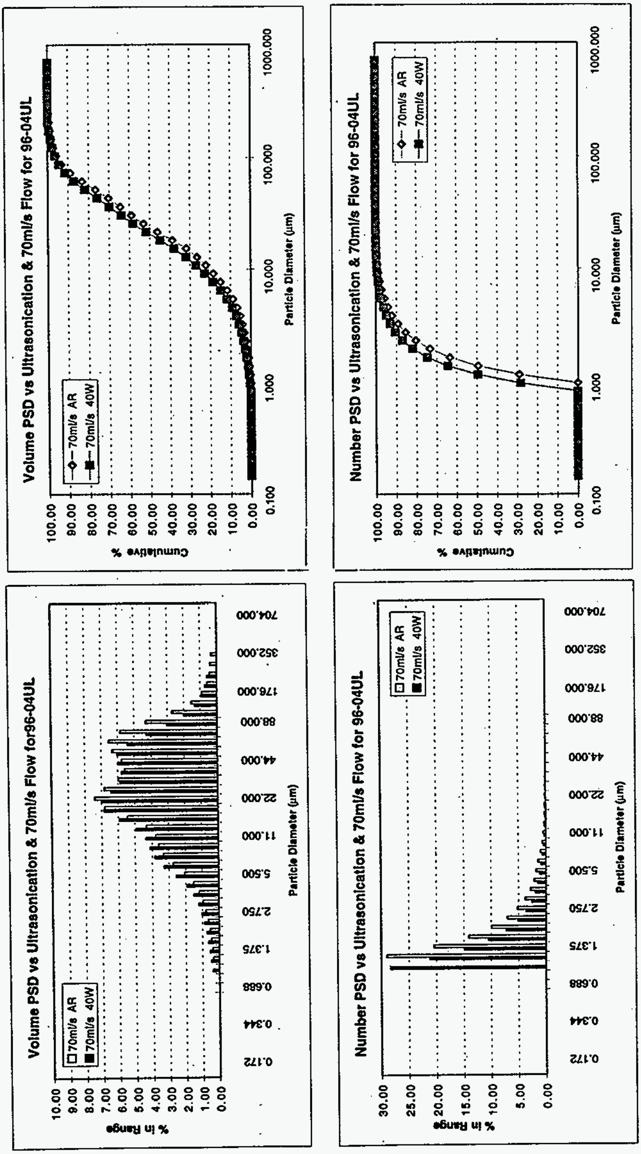


Figure I.6.2-6. Particle Size Distribution Versus Flowrate for Sample 96-04 L.

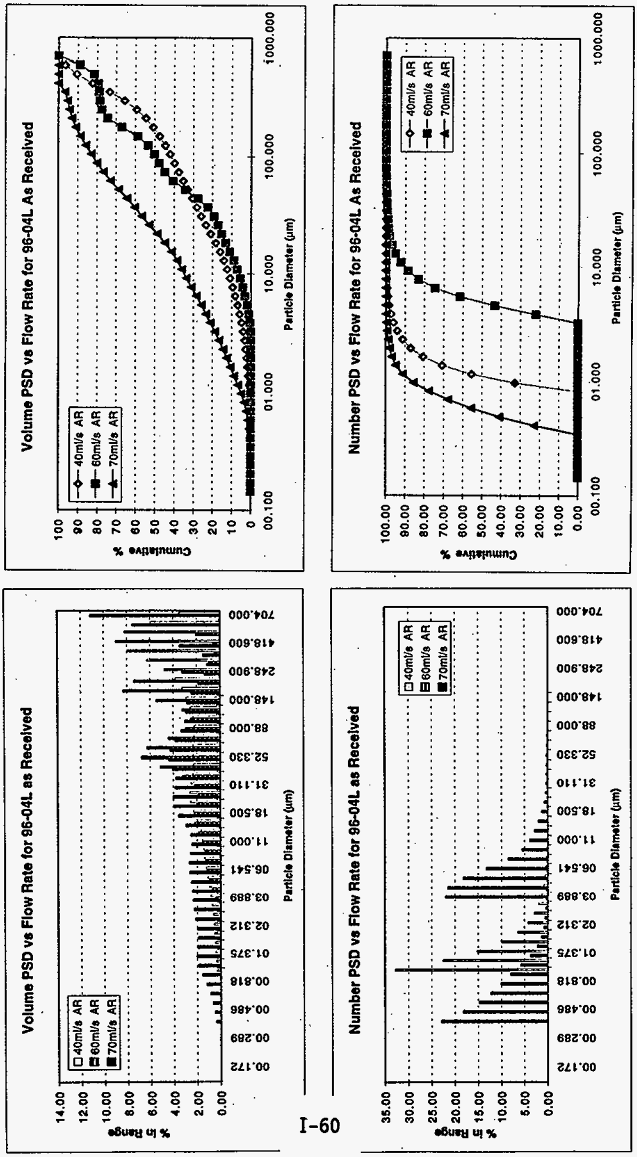


Figure I.6.2-7. Particle Size Distribution Versus Sonification for Sample 96-04 L.

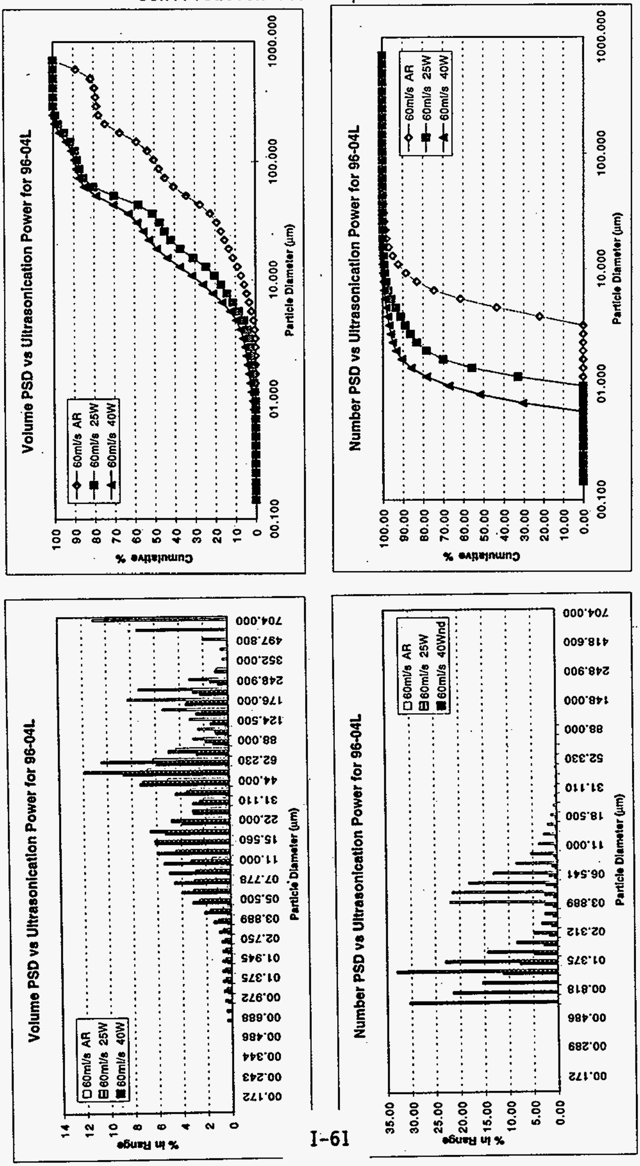
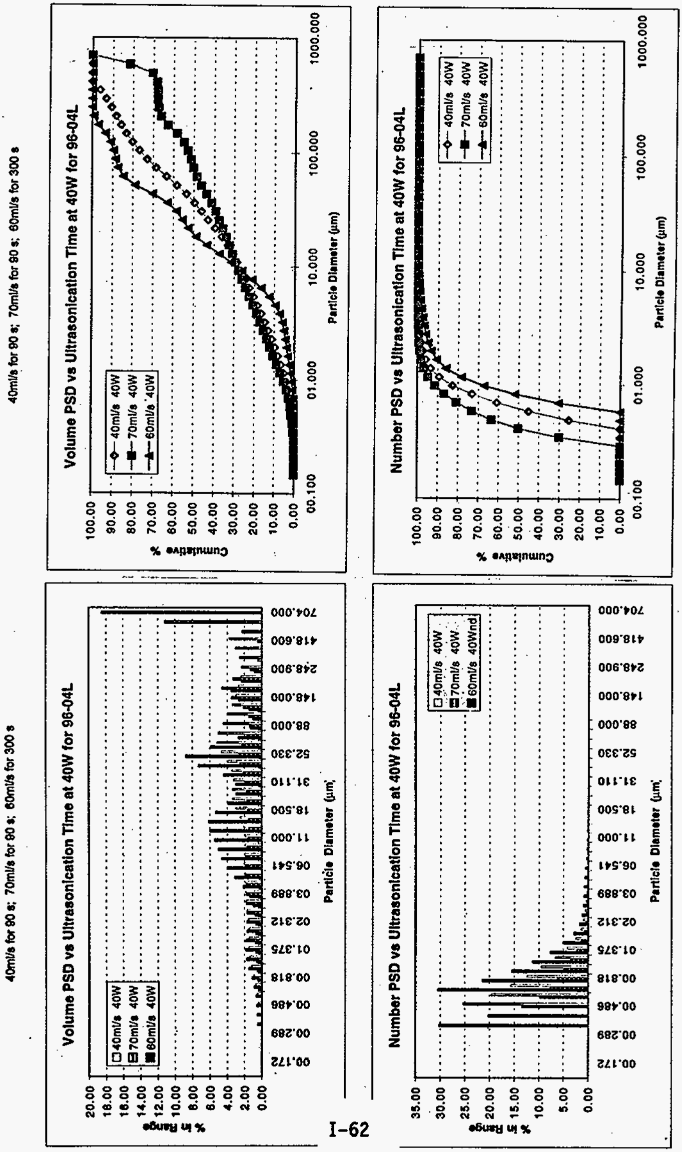


Figure I.6.2-8. Particle Size Distribution Versus Sonification Time for Sample 96-04 L.



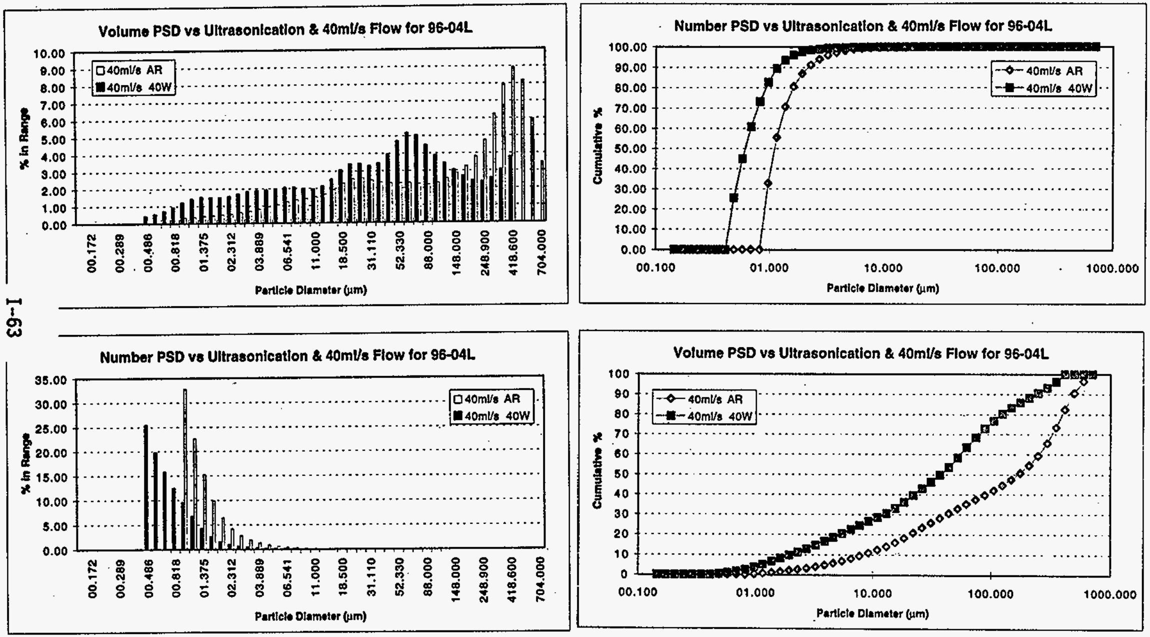
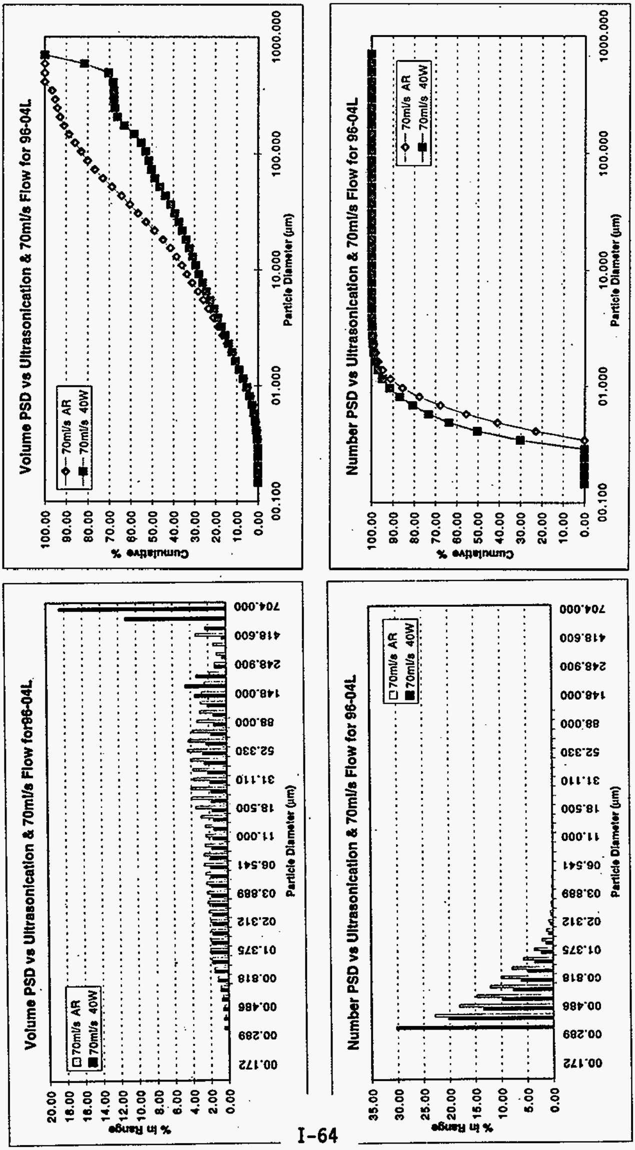


Figure I.6.2-9. Particle Size Distribution Versus Sonification for Sample 96-04 L.

Figure I.6.2-10. Particle Size Distribution Versus Sonification for Sample 96-04 L.



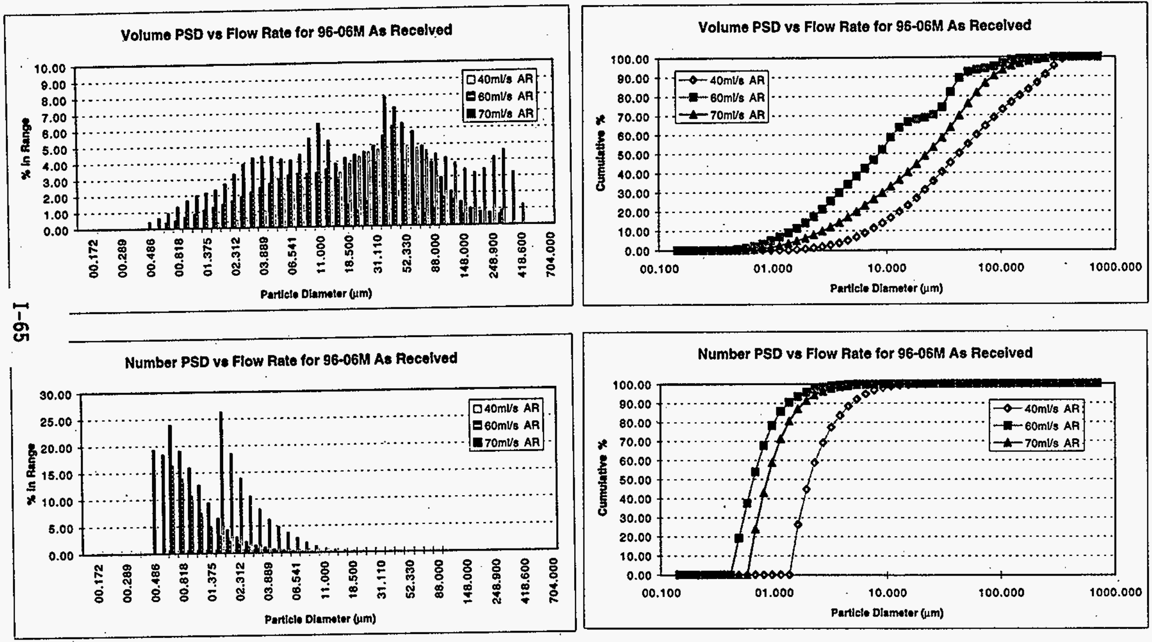


Figure I.6.2-11. Particle Size Distribution Versus Flowrate for Sample 96-06 M.

Figure I.6.2-12. Particle Size Distribution Versus Sonification Power for Sample 96-06 M.

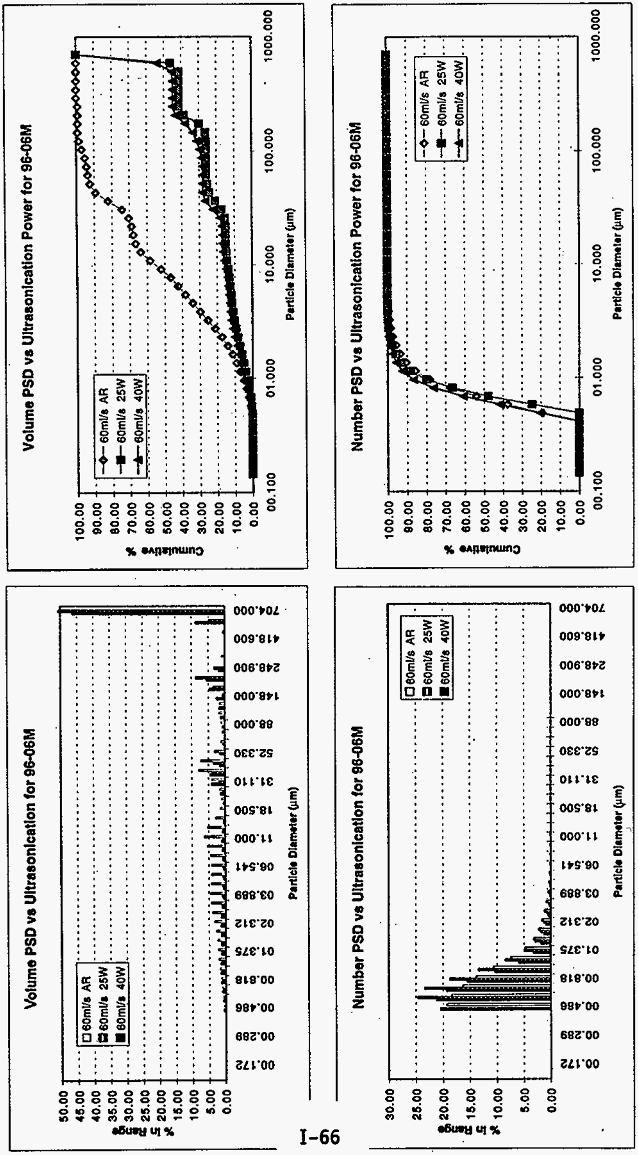
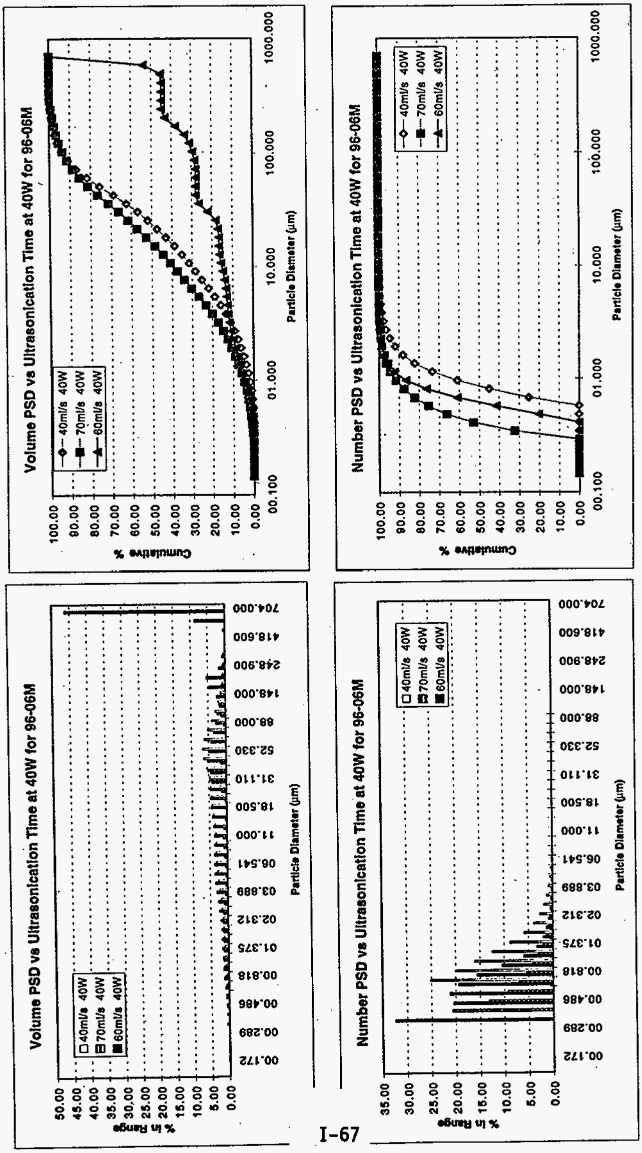


Figure I.6.2-13. Particle Size Distribution Versus Sonification Time for Sample 96-06 M.



40ml/s for 90 s; 70ml/s for 90 s; 60ml/s for 300 s

40ml/s for 90 s; 70ml/s for 90 s; 60ml/s for 300 s

Figure I.6.2-14. Particle Size Distribution Versus Sonification for Sample 96-06 M.

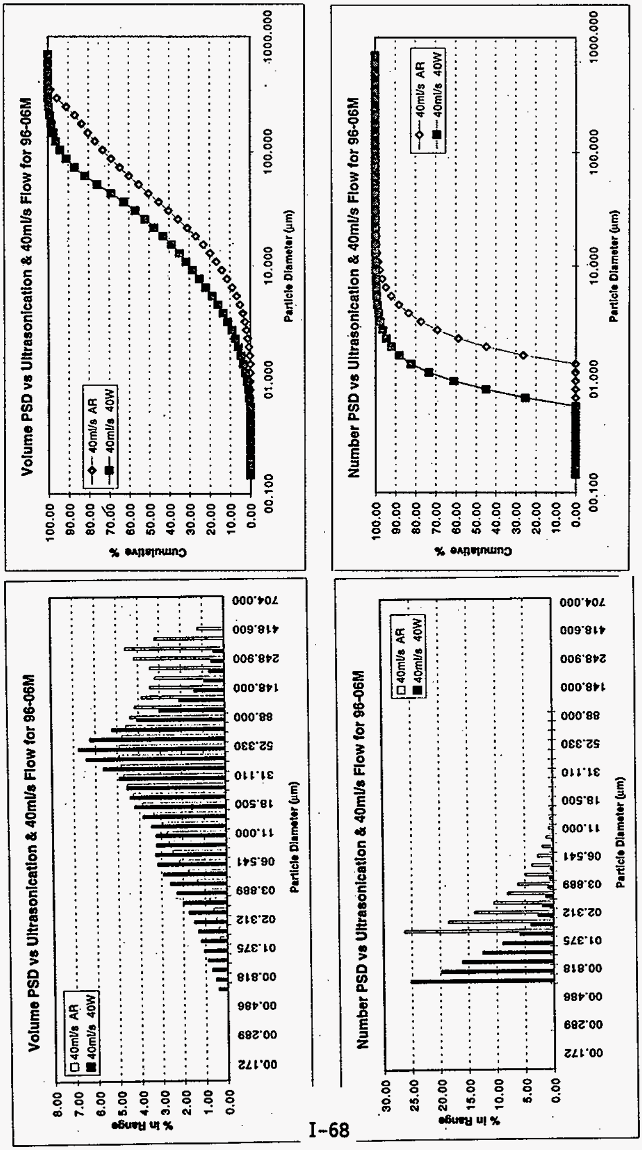


Figure I.6.2-15. Particle Size Distribution Versus Sonification for Sample 96-06 M.

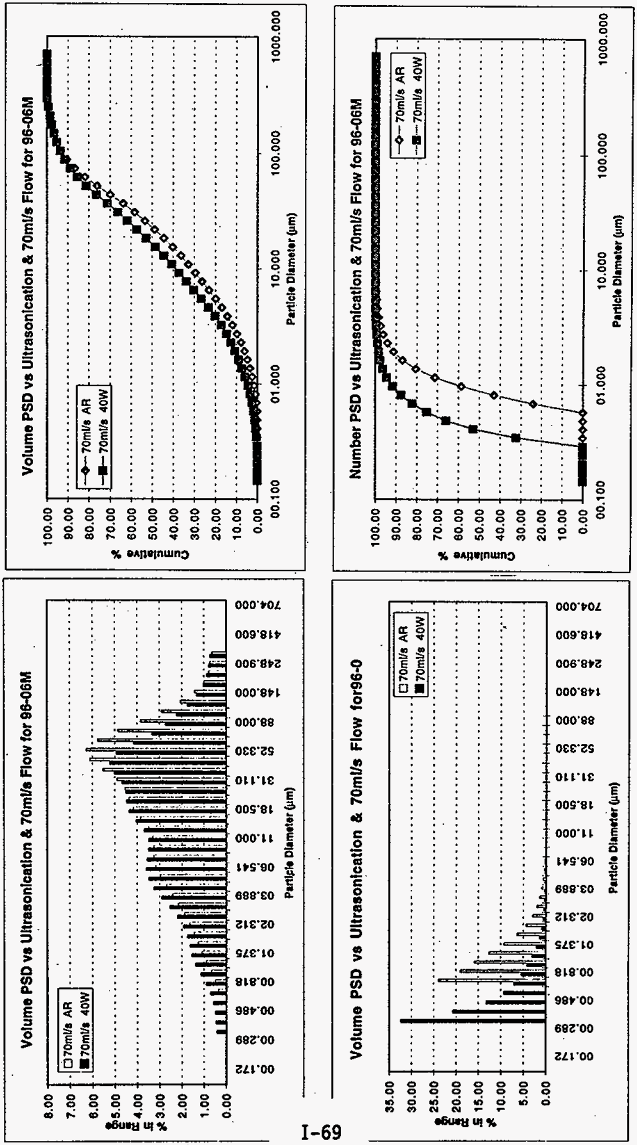
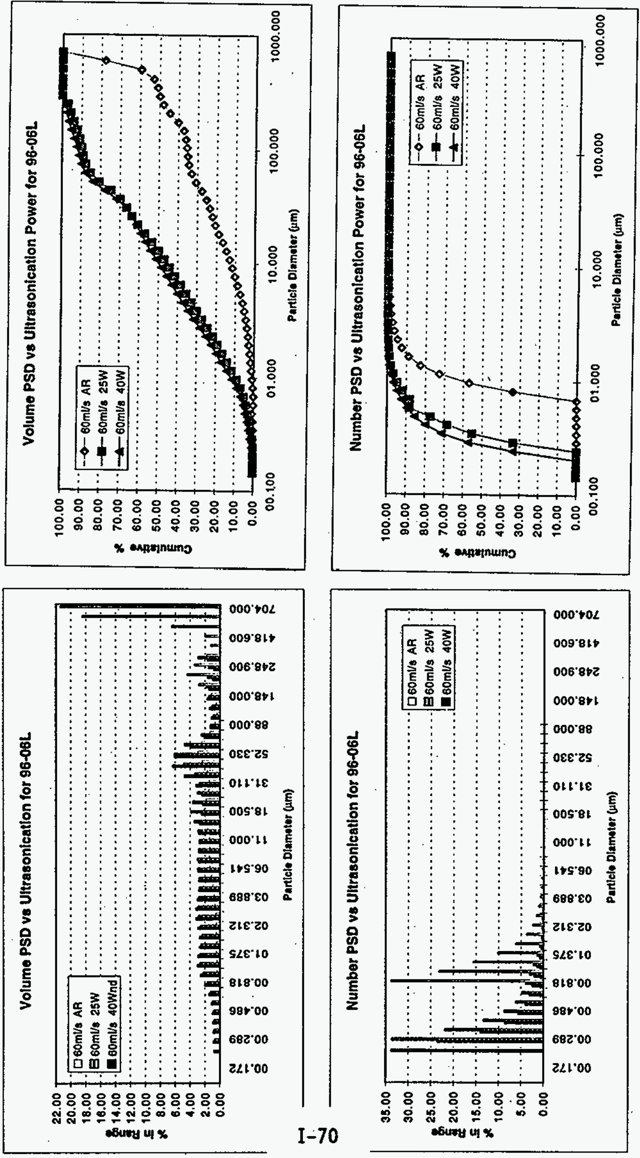


Figure I.6.2-16. Particle Size Distribution Versus Sonification Power for Sample 96-06 L.



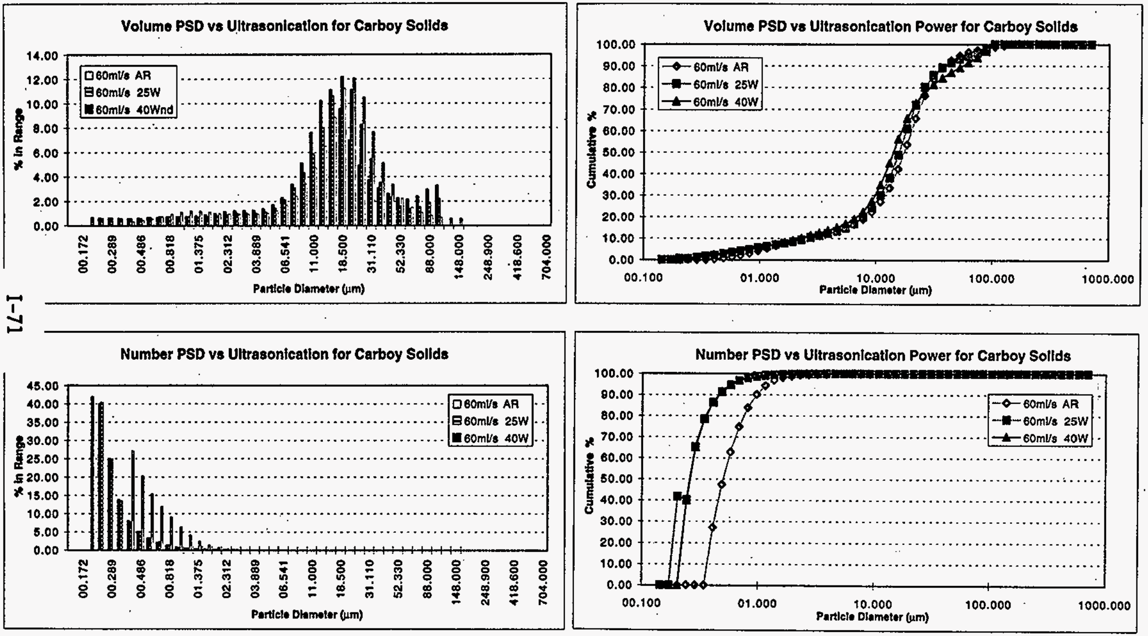


Figure 1.6.2-17. Particle Size Distribution Versus Sonification Power for Sample 96-06 Carboy Solids.

Figure I.6.2-18. Particle Size Distribution Versus Flowrate for Sample 96-11 L.

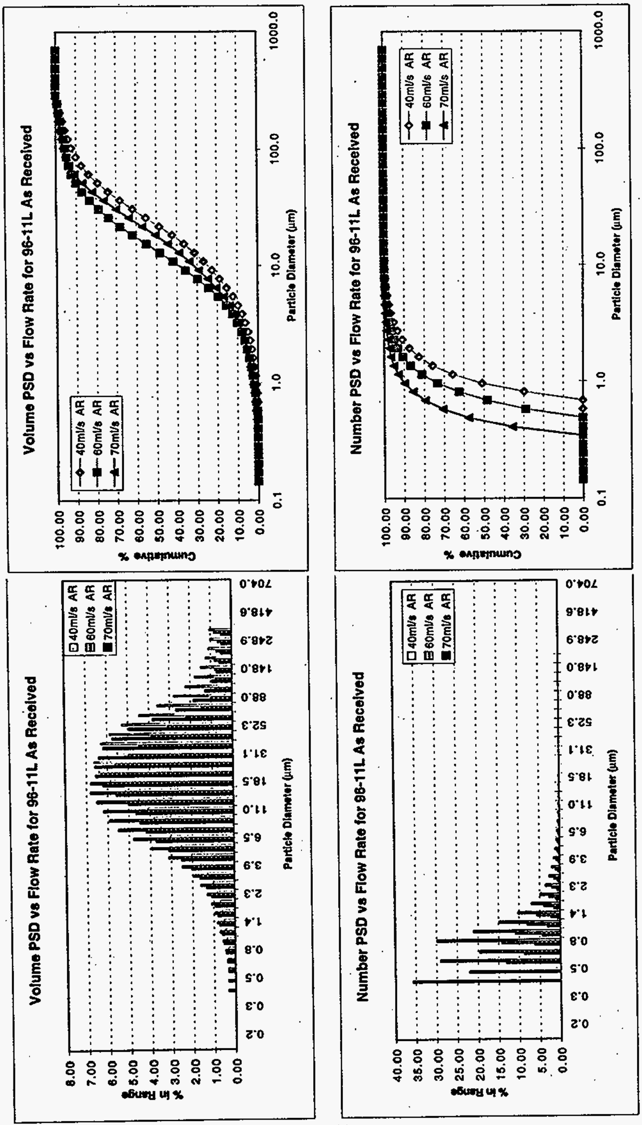


Figure I.6.2-19. Particle Size Distribution Versus Sonification Power for Sample 96-11 L.

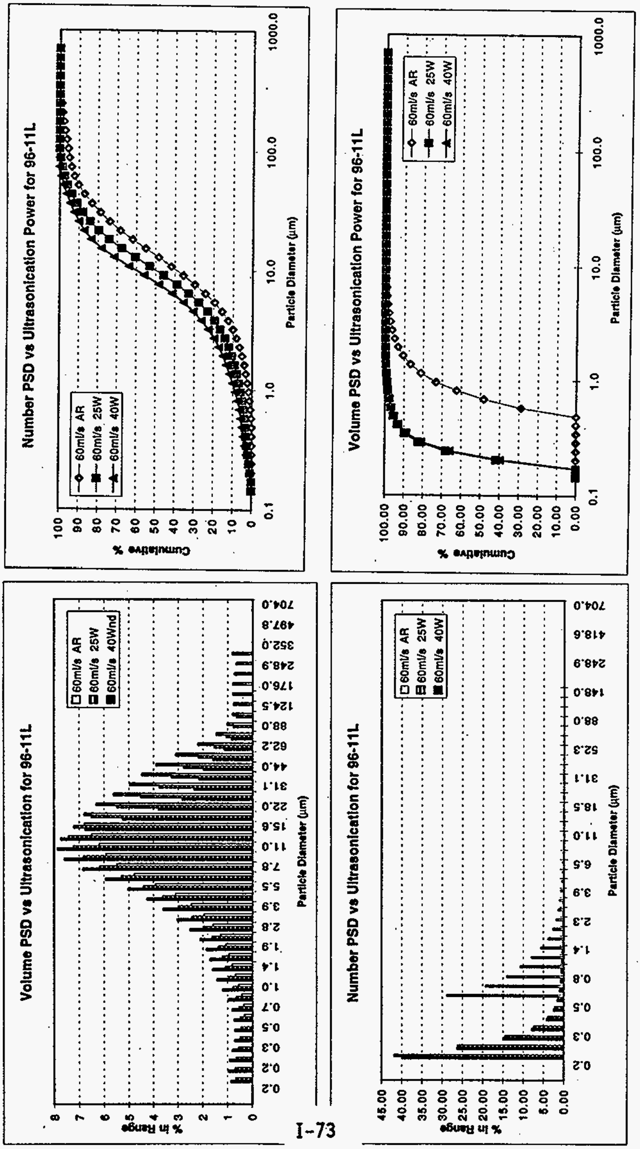
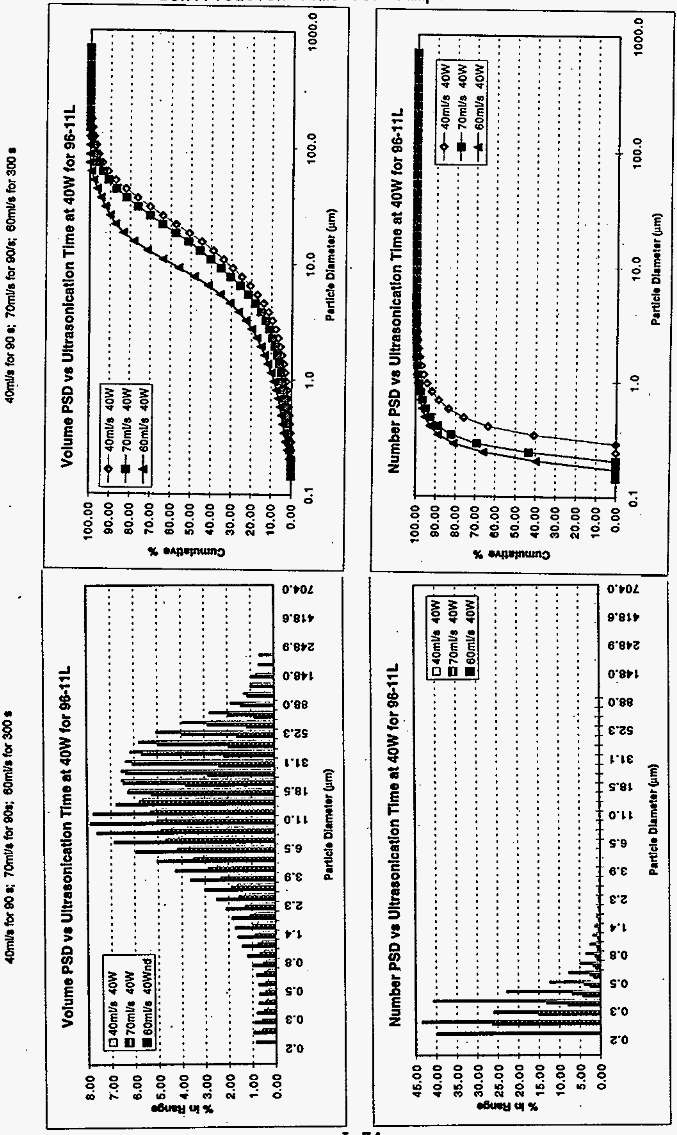


Figure I.6.2-20. Particle Size Distribution Versus Sonification Time for Sample 96-11 L.



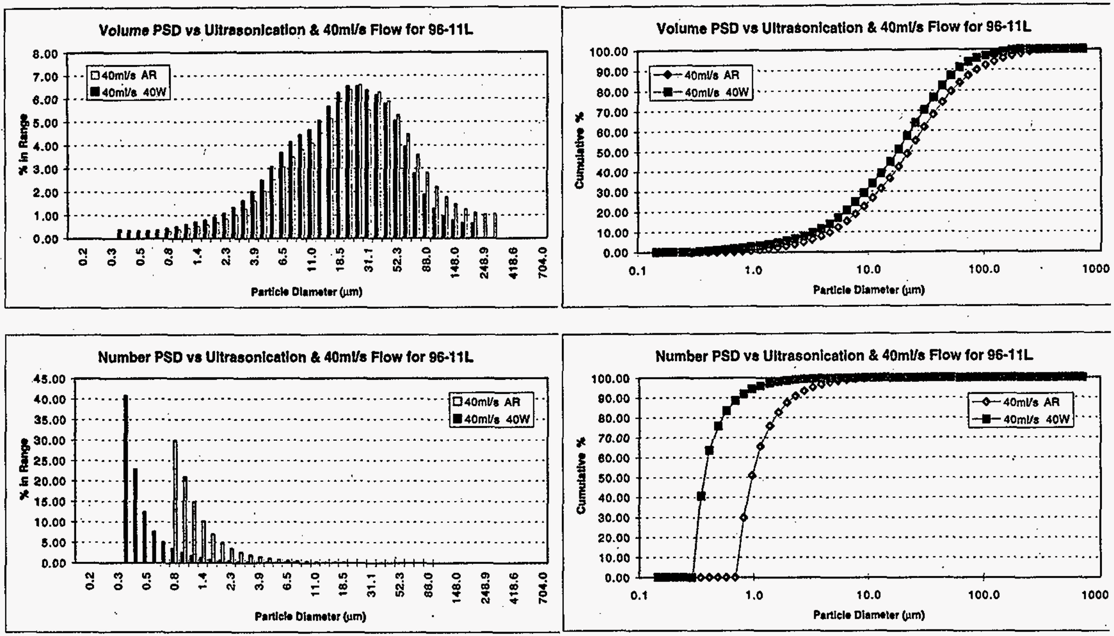


Figure I.6.2-21. Particle Size Distribution Versus Sonification for Sample 96-11 L.

Figure I.6.2-22. Particle Size Distribution Versus Sonification for Sample 96-11 L.

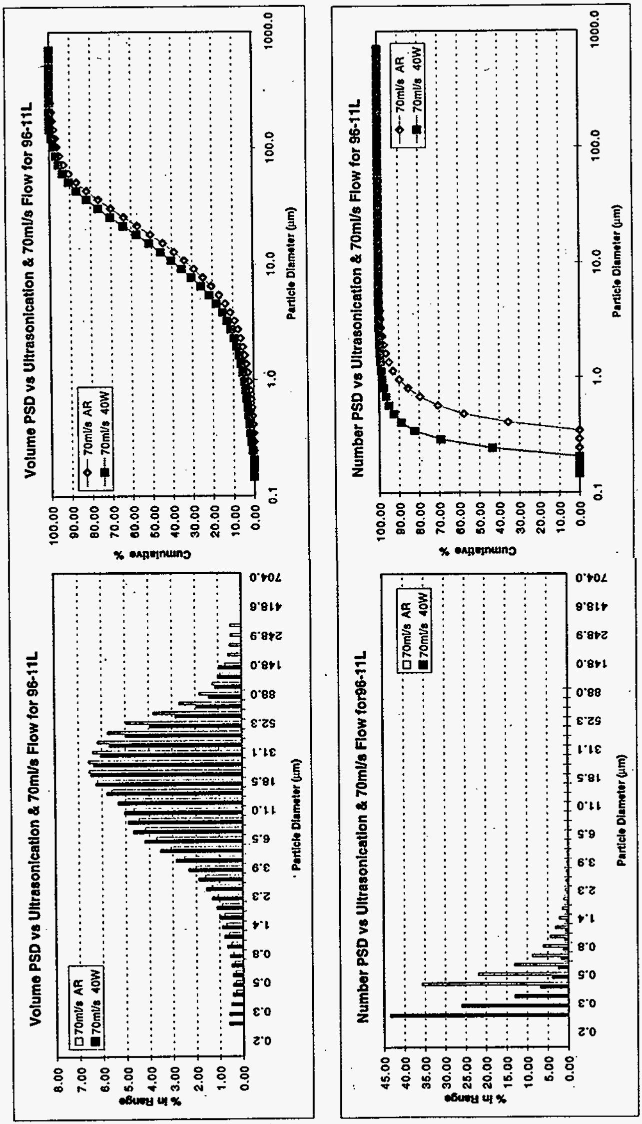


Figure I.7-1. Example Particle Shape Image for Canister Sludge (Sample 96-06 M).

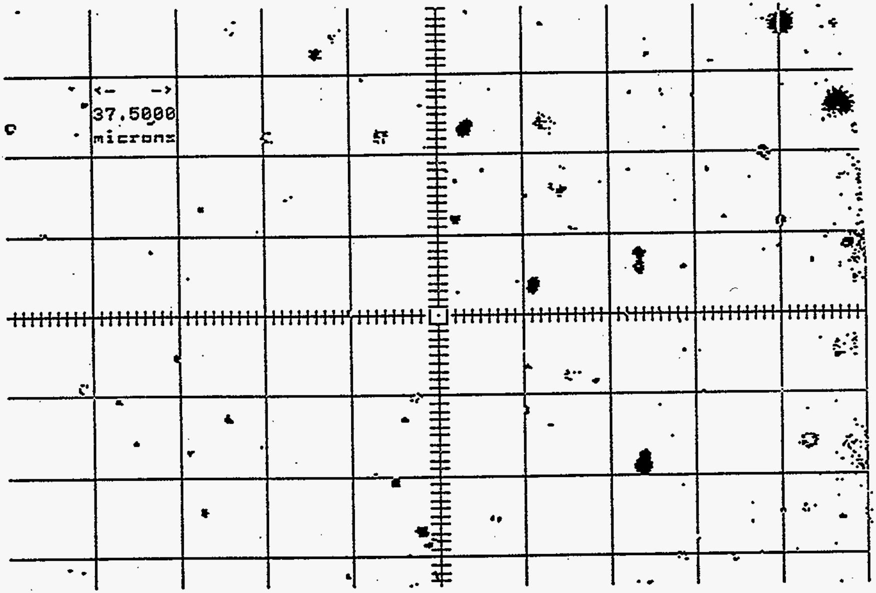


Figure I.7-2. Aspect Ratio and Shape Factor for Sample 96-04 L.

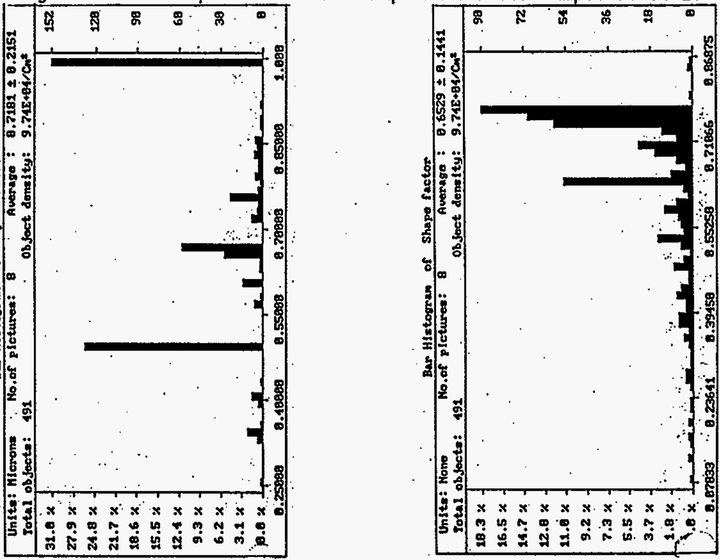


Figure I.7-3. Aspect Ratio and Shape Factor for Sample 96-06 M.

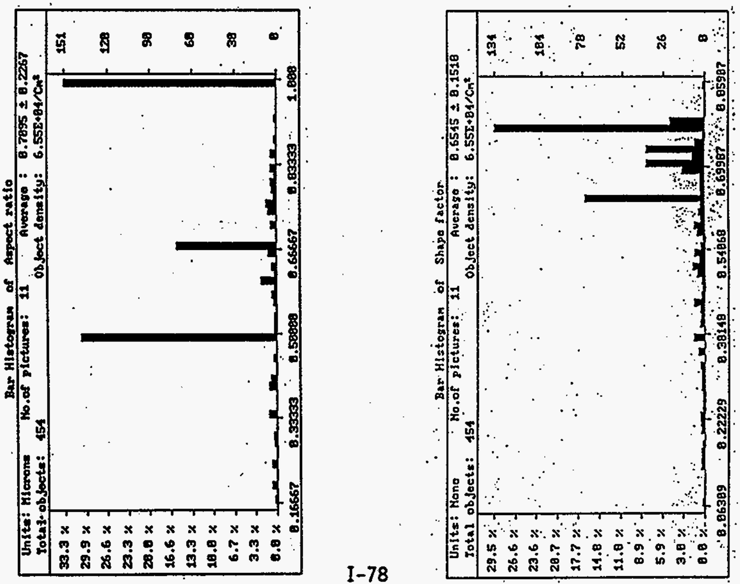


Figure I.7-4. Aspect Ratio and Shape Factor for Sample 96-06 L.

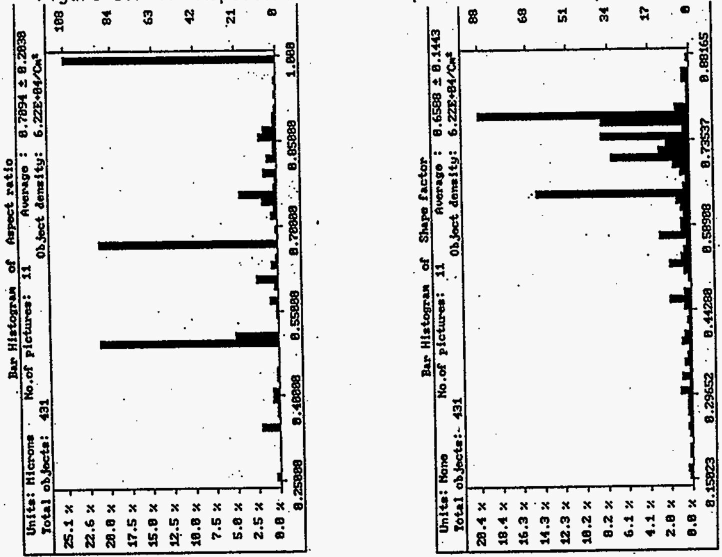


Figure I.7-5. Aspect Ratio and Shape Factor for Sample 96-11 L.

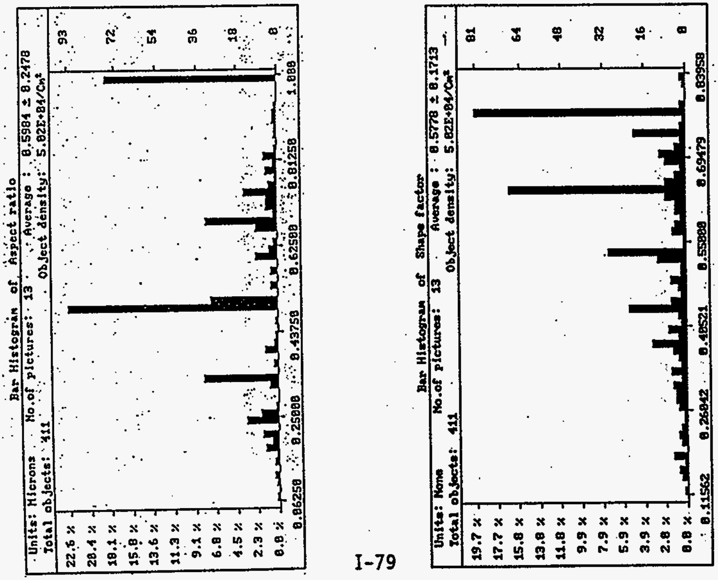


Figure I.9-1. Plot of Viscosity as a Function of Shear Rate for Sample 96-06 M.

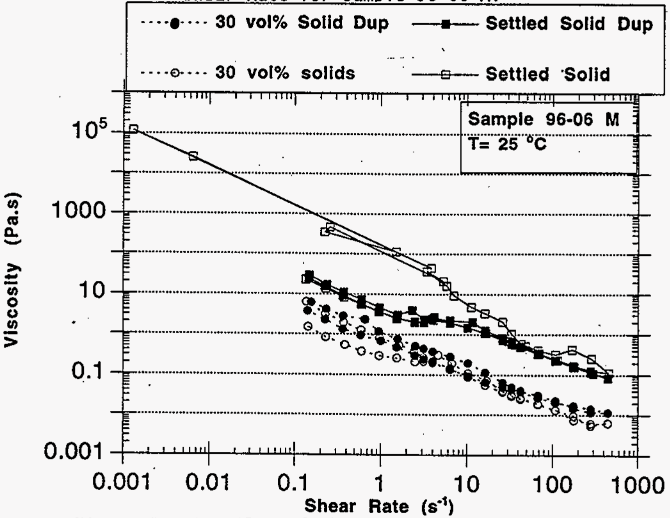


Figure I.9-2. Plot of Shear Rate as a Function of Shear Stress for Sample 96-06 M.

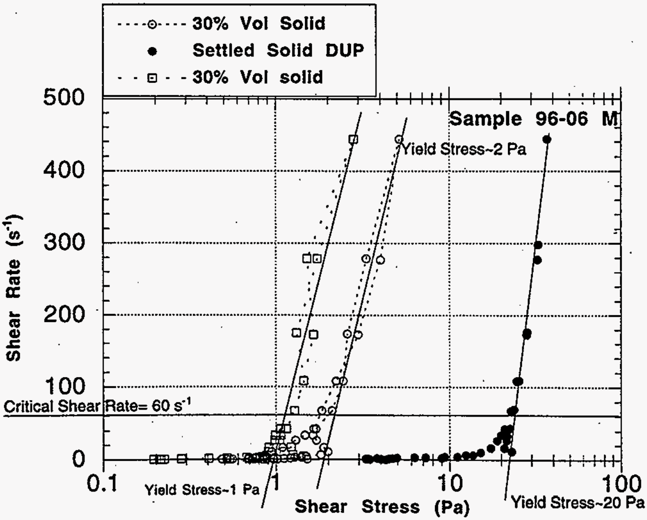


Figure I.9-3. Plot of Viscosity as a Function of Shear Rate for Sample 96-04 U/L.

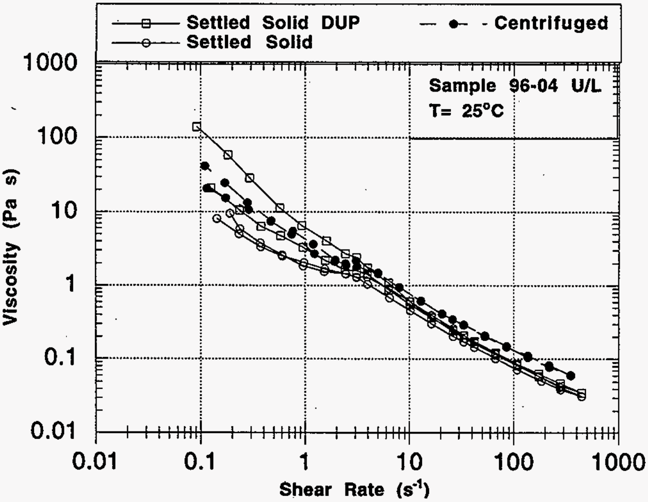


Figure I.9-4. Plot of Shear Rate as a Function of Shear Stress for Sample 96-04 U/L.

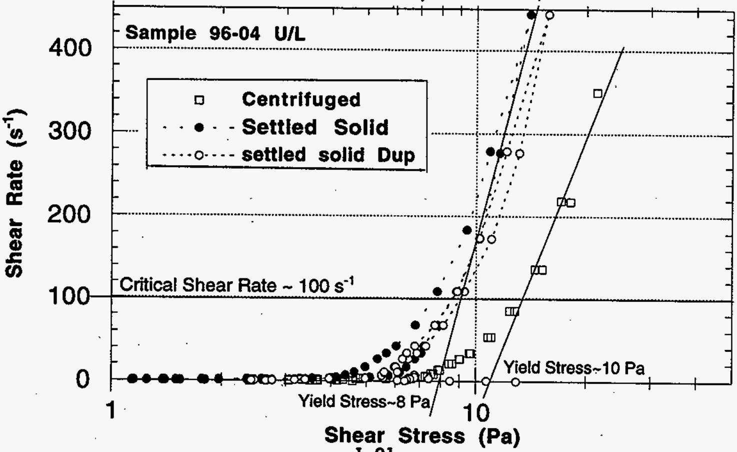


Figure I.9-5. Plot of Viscosity as a Function of Shear Rate for Sample 96-11 L.

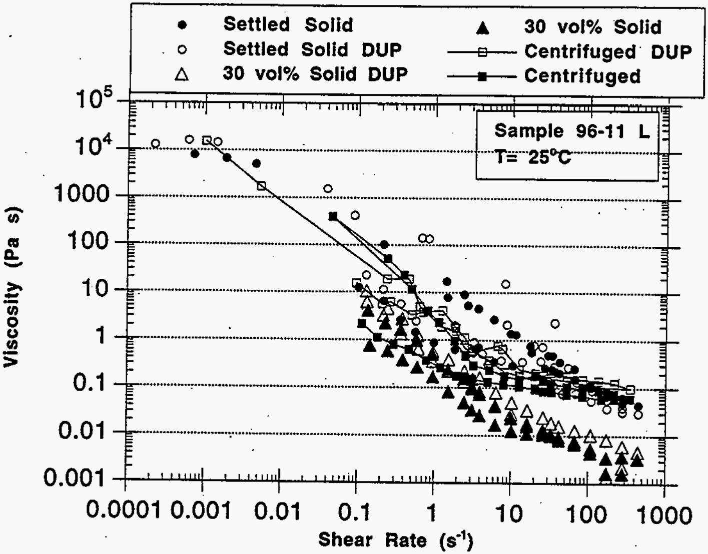
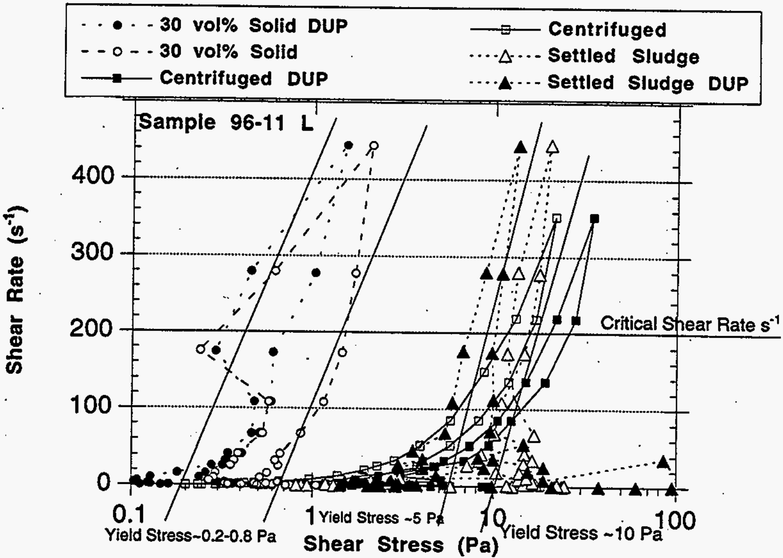


Figure I.9-6. Plot of Shear Rate as a Function of Shear Stress for Sample 96-11 L.



APPENDIX J

X-RAY DIFFRACTION RESULTS

(Excerpted from Reference K. L. Silvers 1997)

E. D. Jenson

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Table J1. Summary of Phases Found in
K East Basin Canister Sludge Samples. (Continued)

Sample ID	Crystalline Phase
96-01	UO_2 U_4O_9 U_3O_7
96-01 magnetic stirrer	$FeFe_2O_4$ Fe_2O_3 $FeCr_2O_4$ $FeO(OH)$ $(Ca,Mn)Mn_4O_9 \cdot 3H_2O$
96-04-L	UO_2 U_4O_9 U_3O_7 $UO_3 \cdot 2H_2O$
96-04-Flakes	$ZrH_{1.66}$ ZrH Zr $UO_3 \cdot 2H_2O$
96-04-U	$UO_3 \cdot 2H_2O$ (Two Forms) UO_2 U_4O_9 U_3O_7
96-05	UO_2 U_4O_9 U_3O_7 $UO_3 \cdot 2H_2O$
96-06-L	UO_2 U_4O_9 U_3O_7
96-06-M	UO_2 U_4O_9 U_3O_7 Al_2O_3
96-06-Carboy Solids	$UO_3 \cdot 2H_2O$ (Two Forms) $C_2H_2CaO_4 \cdot 3CaO \cdot 11H_2O$
96-08	UO_2 U_4O_9 U_3O_7

Table J1. Summary of Phases Found in K East Basin Canister Sludge Samples. (Continued)

Sample ID	Crystalline Phase
96-08-R	SiO_2 Unknown Number 1
96-09	FeO(OH) Fe_2O_3 FeO(OH) $FeFe_2O_4$
96-09-R	SiO_2
96-09-Flakes	$FeFe_2O_4$ Unknown Number 2*
96-11-L	FeO(OH) SiO_2 Fe_2O_3
96-11-R	Unknown Number 3
96-13-1	UO_2 U_4O_9 U_3O_7
96-13-2	UO_2 U_4O_9 U_3O_7
96-15	UO_2 U_4O_9 U_3O_7

*Energy dispersive X-ray analysis performed during Scanning Electron Microscopy indicates the presence of compound(s) containing Al, U, Fe, Ca, Si, and S.

Figure J1. X-Ray Pattern for Sample 96-01.

Note: Only uranium oxides were identified. It is probably not possible to identify minor constituents (less than 1 or 2%) nor to truly say that all three forms of oxide stoichiometry are present given overlapping peaks. Note Al_2O_3 was added as a standard.

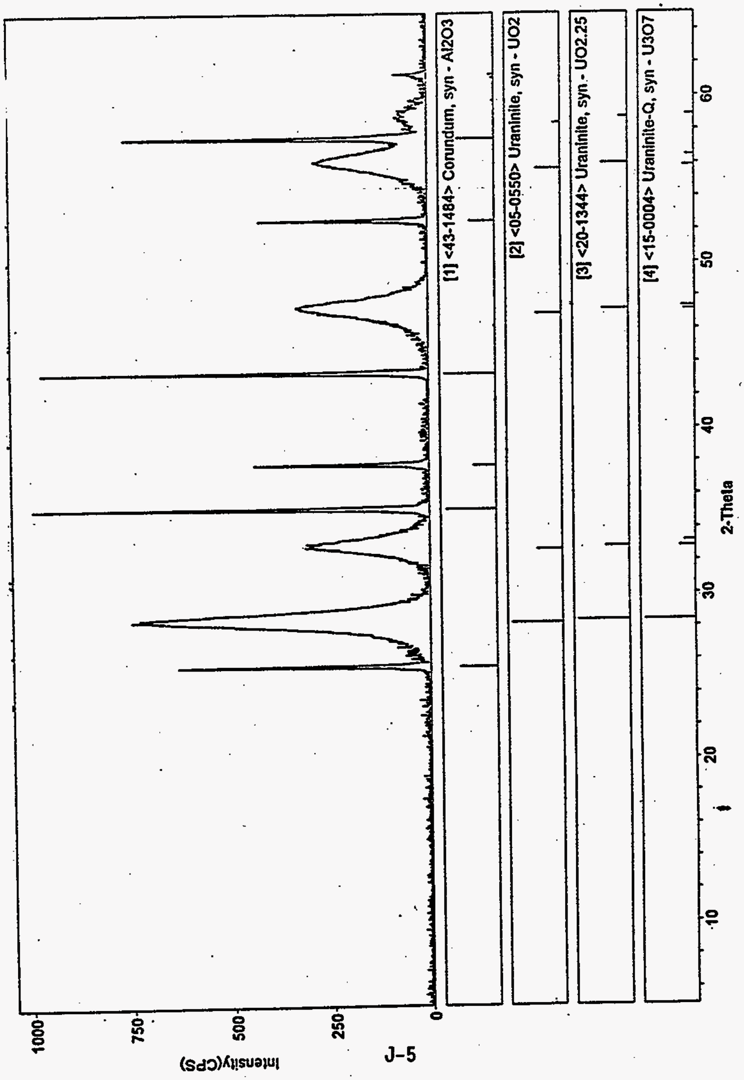


Figure J2. X-Ray Pattern from the Sample Showing the Greatest Indication of Hydrate, Sample 96-04.

This pattern is from the lower layer of the sample but the upper layer also demonstrated a strong indication.

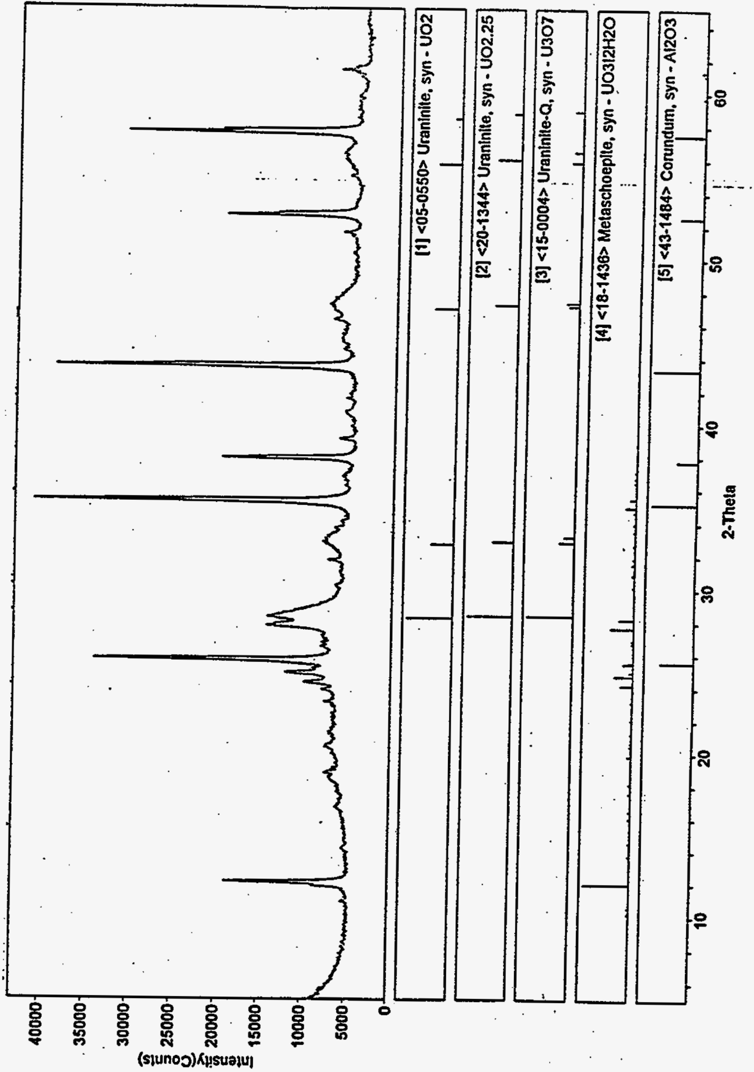


Figure J3. X-Ray Pattern from Sample 96-05
Showing a Weak Indication of Hydrate.

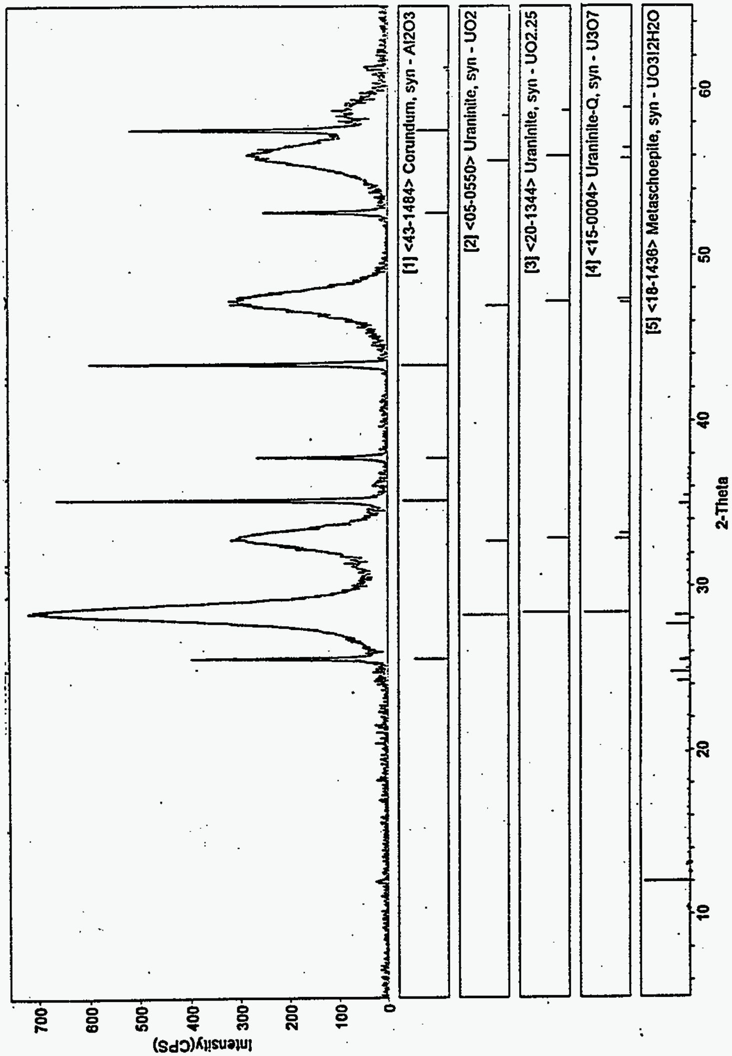
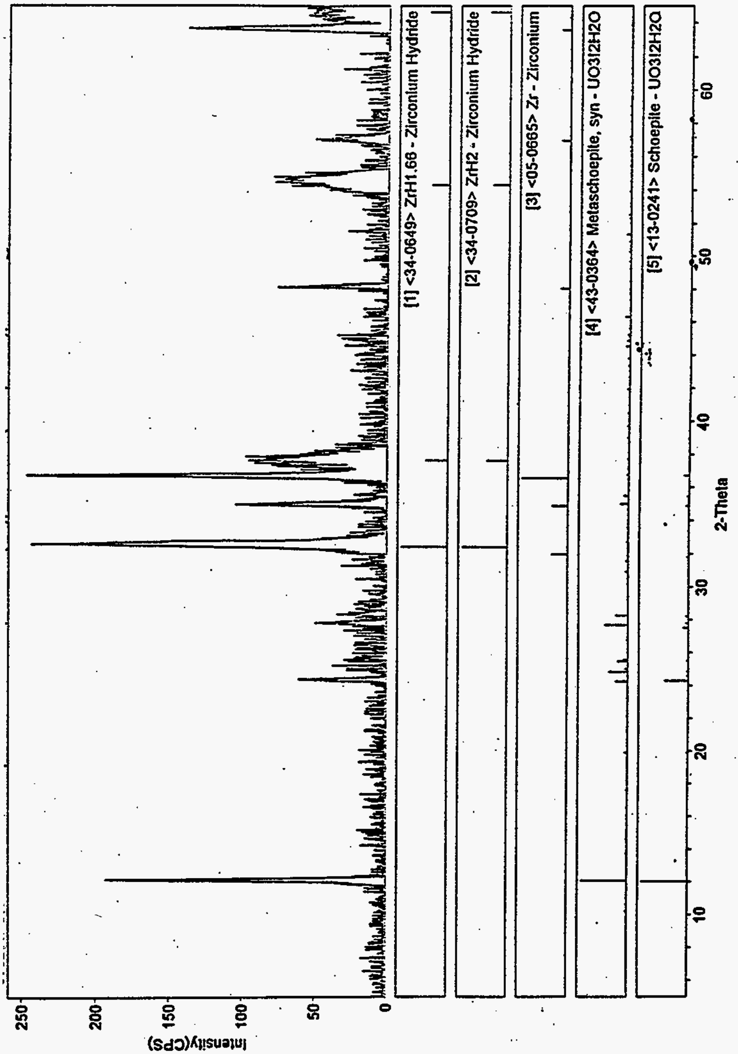


Figure J4. X-Ray Pattern from Shiny Flakes Recovered from Sample 96-04. This is the only indication of zirconium or zirconium hydride found.



APPENDIX K

THERMO-GRAVIMETRIC ANALYSES AND DIGITAL SCANNING CALORIMETRY IN A NITROGEN ATMOSPHERE

(Excerpted from the Work of J. M. Tingey
Reported in Reference Silvers 1997a)

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APPENDIX K

THERMO-GRAVIMETRIC ANALYSES AND DIGITAL SCANNING CALORIMETRY IN A NITROGEN ATMOSPHERE

K.1 THERMAL ANALYSIS

K.2 K EAST BASIN CANISTER SLUDGE

Duplicate analysis of the settled solids from canister Samples 96-01, 96-05, 96-08, 96-09, 96-13, and 96-15 were completed by Differential Scanning Calorimetry (DSC) and Thermo-Gravimetric Analysis/Differential Thermal Analysis (TG-DTA). The analyses were performed on a Seiko Series 5200 Thermal Analysis System according to Pacific Northwest National Laboratory (PNNL) technical procedure PNL-ALO-508 "Laboratory Procedure for Operation of the Differential Scanning Calorimeter (DSC), Thermo-Gravimetric Analyzer (TG), and High Temperature Differential Thermal Analyzer (DTA) and DSC." Approximately 20 mg of sample were placed in platinum pans, and the temperature of the sample was increased from ambient temperature to 773 K at a constant rate (5 K/min). The energy required to increase the temperature of the sample (DSC), the mass of the sample (TG), and the changes in the temperature of the sample (DTA) in relation to a reference sample (an empty platinum pan) were monitored as a function of temperature. These analyses were performed in a flowing nitrogen atmosphere.

The calibration of the thermal analysis system was checked with indium and lead standards and a calibrated weight. The literature values for the enthalpy and onset temperature of indium and lead were 28.59 J/g and 429.75 K (indium), and 23.22 J/g and 600.65 K (lead). The measured values for the onset temperature of indium and lead were 430.0 K and 600.7 K for the DSC and 429.8 K and 600.9 K for the DTA. The enthalpy of the indium and lead as measured by DSC are 28.9 and 23.6 J/g. The deviation of the measured values from the reported values is less than 2% for the enthalpy and within 0.3 K for the onset temperatures.

The results of these analyses are reported in Tables K1 and K2. Two major transitions are observed in all of the samples. The first transition occurs between ambient temperature and 398 K. This transition is due to the evaporation of water from the sample, and a significant sample mass loss is associated with this transition. The enthalpy for this transition is dependent upon the amount of water in the sample (wt% solids) and varies between samples. This water evaporation endotherm consists of multiple endotherms which cannot be resolved from Samples 96-08, 96-13, and 96-15.

In Samples 96-01, 96-05, and 96-09 the evaporation of water appears to be a single endothermic transition. The multiple endothermic transitions are indicative of evaporation of water from different environments (inner and outer coordination spheres and waters of hydration) and were expected.

In these K East Basin canister sludges, significant water evaporates at ambient temperatures; therefore, many of the samples had lost substantial water before the analyses were begun. In the DSC scans, this loss of water is indicated by curves beginning below the baseline; due to the premature loss of water, an accurate enthalpy for the evaporation of water cannot be determined for all samples. Similarly, loss of water in the TG curves is indicated by masses beginning at less than 100% of the original material mass. Therefore, small differences in TG scan results and values listed in Tables K1 and K2 will be observed; mass loss associated with reported transitions are based on mass of the original sample (100% mass).

The second transition observed in all the samples had an onset temperature of about 470 K. The enthalpy of this transition also varies between samples but is much smaller than the first transition. A small mass loss with the same onset temperature is observed for this transition. The first and second endothermic transitions were observed in both the DSC and DTA accompanied with mass losses in the TG analysis.

The second transition was not observed in the DSC for the Sample 96-01 duplicate. The mass loss for this transition was also not clearly observed. There was a mass loss in the sample following the evaporation of the water, but this loss continued throughout the temperature range of the analysis. In the DSC analysis it also appears that an endothermic transition may have occurred after the evaporation of water, but this transition cannot be clearly resolved from the baseline.

An additional transition is observed in the sludge from canister Sample 96-13. This transition is exothermic with an enthalpy between 17 and 44 J/g. The onset temperature for this exothermic behavior is about 630 K. this exothermic behavior was observed in both the sample and duplicate DSC analyses of Sample 96-13 but was not observed in the DSC analyses of any other sample. A mass loss of approximately 1.3% may be associated with this exotherm, but it is difficult to resolve the mass loss from the second and third transitions.

All of the data is based on wet weights. For more consistency between samples, the data can be calculated on a dry weight basis for the second and third transitions.

The thermal analysis results for these samples under these conditions are similar to the results observed in more detailed thermal analysis over a wider temperature range. The first two transitions were also observed in these more detailed analyses.

The weight percent water in the samples varied significantly between samples and even between the sample and duplicate. In these K East Basin canister sludges significant water evaporates at ambient temperatures; therefore, many of the samples lost substantial water while they are stored in vials. The mass loss can vary between vials based on how effective of a seal is placed on the vial. Since the caps of these samples are placed on the vials in the hot cells, the effectiveness of the seal will vary between vials.

Table K1. Differential Scanning Calorimetry Results.

Sample	Temperature Range (K)	Onset Temperature (K)	Enthalpy (J/g)
96-01 Duplicate	294-408	*	247
96-01 Sample	294-399 453-528	* 465.4	173 11.5
96-05 Sample	294-370 383-413 450-532	* ** 477.1	204 0.6 15.8
96-05 Duplicate	296-400 454-518	* 467.5	184 15.5
96-08 Sample	294-387 439-520	*	*** 27.1
96-08 Duplicate	293-394 425-528	* 467	*** 43.5
96-09 Sample	294-415 415-537	* 463	*** 111.3
96-09 Duplicate	296-393 426-576	* 466	*** 101.7
96-13 Sample	295-415 453-517 610-805	* 466 631	*** 7.7 -44.9
96-13 Duplicate	300-460 460-527 689-801	* 469 696	*** 6.7 -16.7
96-15 Sample	296-441 441-677	* 471	*** 59.2
96-15 Duplicate	294-429 434-611	* 467	*** 27.5

*This transition began at the start of the run; therefore, it was not possible to determine the onset temperature.

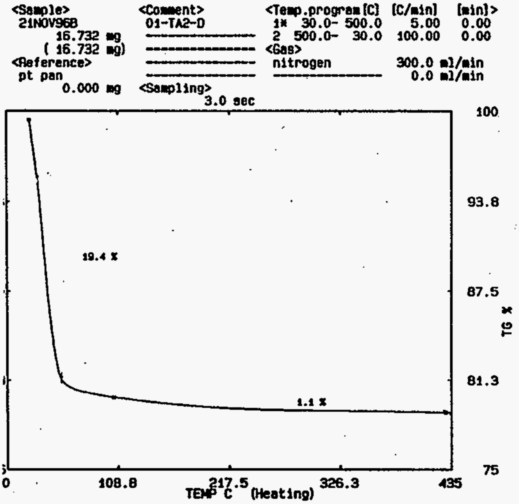
**This transition was so small that it was difficult to obtain an accurate onset temperature.

***The enthalpy of this water evaporation was not accurately measured because the endotherm began below the baseline due to premature evaporation of water from the sample.

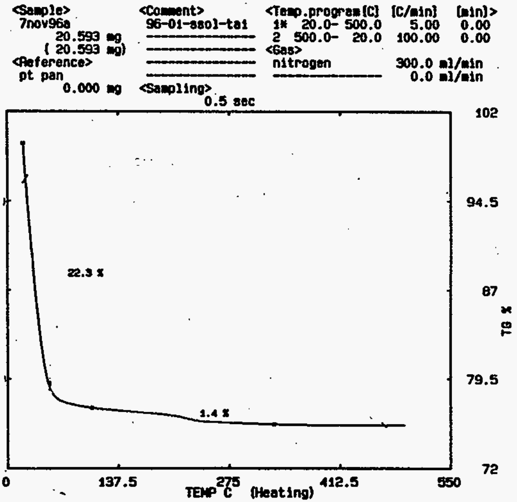
Table K2. Thermo-Gravimetric Analysis Results.

Sample	Temperature Range (K)	Mass Loss (%)
96-01 Duplicate	295-413 413-700	20.3 0.7
96-01 Sample	293-413 413-600	23.1 1.2
96-05 Sample	295-413 413-600 600-773	19.8 1.4 0.1
96-05 Duplicate	295-400 400-605	15.5 1.2
96-08 Sample	295-402 402-558 558-820	57.6 3.3 1.5
96-08 Duplicate	296-425 425-527 527-817	35.7 3.4 2.9
96-09 Sample	296-414 414-817	7.7 14.0
96-09 Duplicate	296-389 489-567 567-815	44.6 6.7 2.4
96-13 Sample	295-418 418-713	15.0 2.3
96-13 Duplicate	300-446 446-531 531-704	3.4 1.0 1.3
96-15 Sample	296-450 441-540 540-717	10.0 2.3 2.1
96-15 Duplicate	296-411 411-704	15.0 3.4

Figure K1. Thermo-Gravimetric Analyses Data from Canister Sludge Sample 96-01.

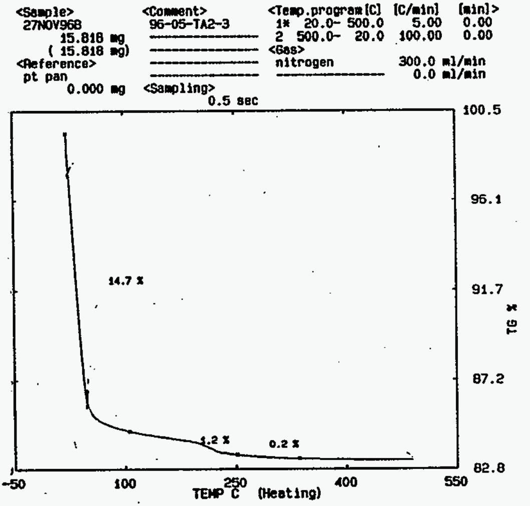


```
<Sample>         <Comment>          <Temp.program[C]  [C/min]   [min]>
 21NOV96B         01-TA2-D           1*  30.0- 500.0    5.00    0.00
      16.732 mg  ---------------     2  500.0-  30.0  100.00    0.00
    ( 16.732 mg) ---------------   <Gas>
<Reference>      ---------------    nitrogen          300.0 ml/min
 pt pan          ---------------    ---------------     0.0 ml/min
       0.000 mg  <Sampling>
                          3.0 sec
```

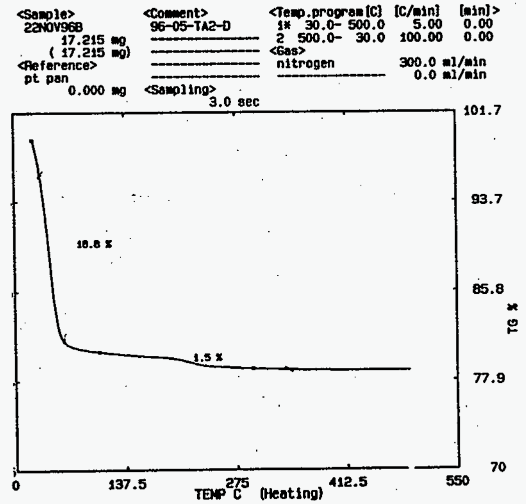


```
<Sample>         <Comment>          <Temp.program[C]  [C/min]   [min]>
 7nov96a          96-01-ssol-ta1     1*  20.0- 500.0    5.00    0.00
      20.593 mg  ---------------     2  500.0-  20.0  100.00    0.00
    ( 20.593 mg) ---------------   <Gas>
<Reference>      ---------------    nitrogen          300.0 ml/min
 pt pan          ---------------    ---------------     0.0 ml/min
       0.000 mg  <Sampling>
                          0.5 sec
```

Figure K2. Thermo-Gravimetric Analyses Data from Canister Sludge Sample 96-05.



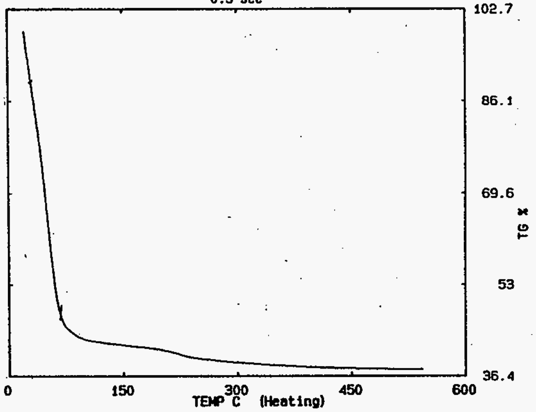
```
<Sample>             <Comment>            <Temp.program[C]  [C/min]   [min]>
 27NOV96B             96-05-TA2-3           1*  20.0- 500.0    5.00    0.00
       15.818 mg     ------------------     2  500.0-  20.0  100.00    0.00
     ( 15.818 mg)    ------------------   <Gas>
<Reference>          ------------------    nitrogen          300.0 ml/min
 pt pan              ------------------    ----------------    0.0 ml/min
        0.000 mg    <Sampling>
                           0.5 sec
```



```
<Sample>             <Comment>            <Temp.program[C]  [C/min]   [min]>
 22NOV96B             96-05-TA2-D           1*  30.0- 500.0    5.00    0.00
       17.215 mg     ------------------     2  500.0-  30.0  100.00    0.00
     ( 17.215 mg)    ------------------   <Gas>
<Reference>          ------------------    nitrogen          300.0 ml/min
 pt pan              ------------------    ----------------    0.0 ml/min
        0.000 mg    <Sampling>
                           3.0 sec
```

Figure K3. Thermo-Gravimetric Analyses Data from Canister Sludge Sample 96-08.

```
<Sample>            <Comment>            <Temp.program[C]  [C/min]   [min]>
 22JAN97B           96-08TGA1             1*  20.0- 550.0    5.00    0.00
         15.090 mg  ----------------      2  550.0-  20.0  100.00    0.00
     ( 15.090 mg)   ----------------     <Gas>
<Reference>         ----------------      nitrogen          300.0 ml/min
 pt pan             ----------------      ----------------    0.0 ml/min
        0.000 mg    <Sampling>
                              0.5 sec
```



```
<Sample>            <Comment>            <Temp.program[C]  [C/min]   [min]>
 22JAN97F           96-08TGA2             1*  20.0- 550.0    5.00    0.00
         21.704 mg  ----------------      2  550.0-  20.0  100.00    0.00
     ( 21.704 mg)   ----------------     <Gas>
<Reference>         ----------------      nitrogen          300.0 ml/min
 pt pan             ----------------      ----------------    0.0 ml/min
        0.000 mg    <Sampling>
                              0.5 sec
```

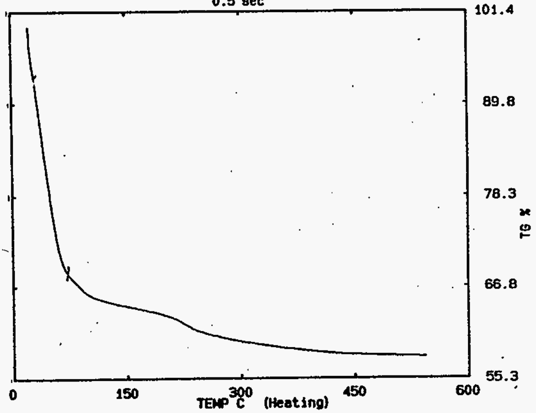
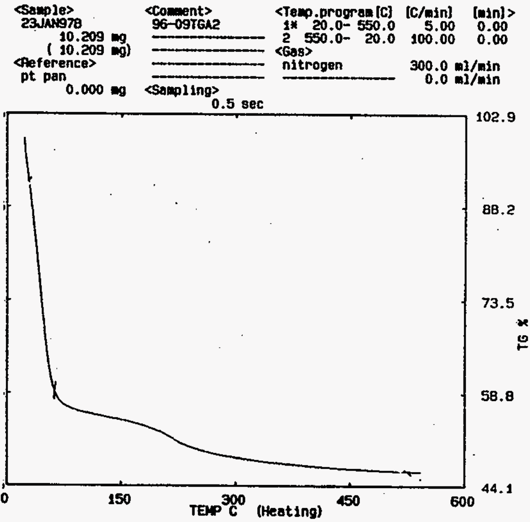
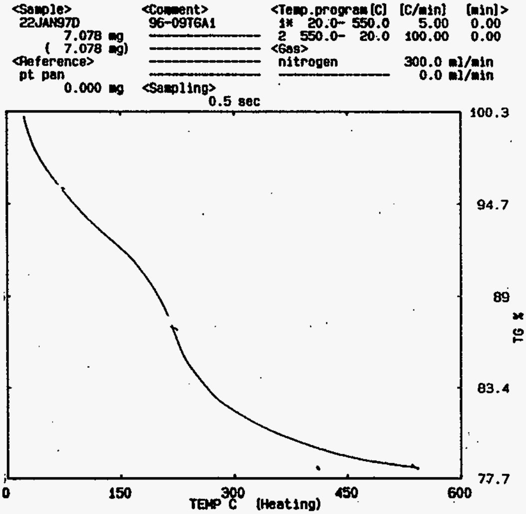


Figure K4. Thermo-Gravimetric Analyses Data from Canister Sludge Sample 96-09.

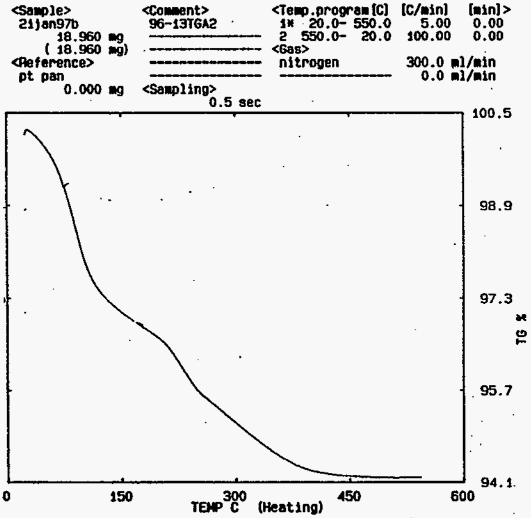


```
<Sample>          <Comment>        <Temp.program[C]  [C/min]   [min]>
 23JAN97B          96-09TGA2        1*  20.0- 550.0     5.00    0.00
      10.209 mg   ---------------   2  550.0-  20.0   100.00    0.00
    ( 10.209 mg)  ---------------  <Gas>
<Reference>       ---------------   nitrogen          300.0 ml/min
 pt pan           ---------------  ---------------      0.0 ml/min
       0.000 mg   <Sampling>
                            0.5 sec
```

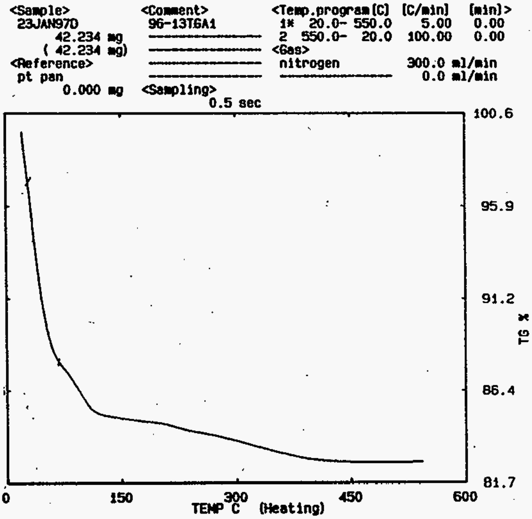


```
<Sample>          <Comment>        <Temp.program[C]  [C/min]   [min]>
 22JAN97D          96-09TGA1        1*  20.0- 550.0     5.00    0.00
       7.078 mg   ---------------   2  550.0-  20.0   100.00    0.00
     ( 7.078 mg)  ---------------  <Gas>
<Reference>       ---------------   nitrogen          300.0 ml/min
 pt pan           ---------------  ---------------      0.0 ml/min
       0.000 mg   <Sampling>
                            0.5 sec
```

Figure K5. Thermo-Gravimetric Analyses Data from Canister Sludge Sample 96-13.



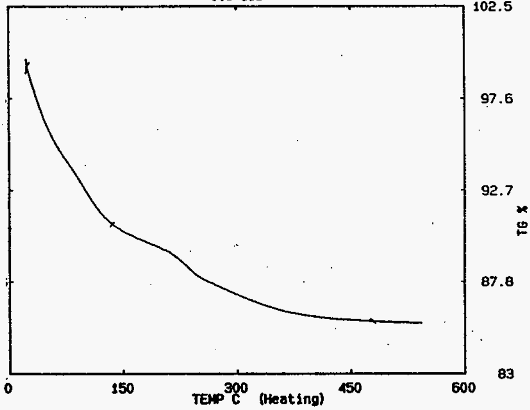
```
<Sample>            <Comment>           <Temp.program[C] [C/min]  [min]>
 21jan97b            96-13TGA2           1*  20.0- 550.0    5.00    0.00
       18.960 mg    ---------------     2  550.0-  20.0  100.00    0.00
     ( 18.960 mg)   ---------------    <Gas>
<Reference>         ---------------     nitrogen           300.0 ml/min
 pt pan             ---------------     ---------------      0.0 ml/min
        0.000 mg    <Sampling>
                              0.5 sec
```



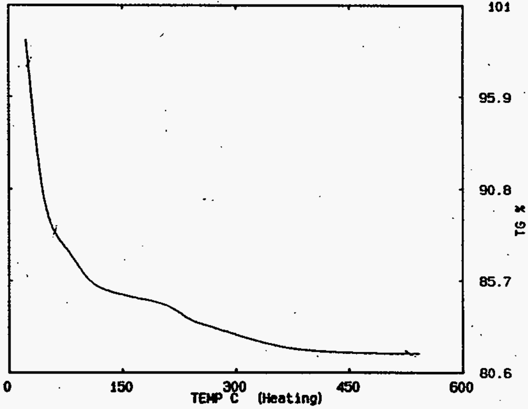
```
<Sample>            <Comment>           <Temp.program[C] [C/min]  [min]>
 23JAN97D            96-13TGA1           1*  20.0- 550.0    5.00    0.00
       42.234 mg    ---------------     2  550.0-  20.0  100.00    0.00
     ( 42.234 mg)   ---------------    <Gas>
<Reference>         ---------------     nitrogen           300.0 ml/min
 pt pan             ---------------     ---------------      0.0 ml/min
        0.000 mg    <Sampling>
                              0.5 sec
```

Figure K6. Thermo-Gravimetric Analyses Data
from Canister Sludge Sample 96-15.

```
<Sample>             <Comment>            <Temp.program[C]  [C/min]   [min]>
 14JAN97C             96-15-TGA1           1*  20.0- 550.0     5.00    0.00
      30.739 mg      ------------------    2  550.0-  20.0   100.00    0.00
    ( 30.739 mg)     ------------------   <Gas>
<Reference>          ------------------    nitrogen          300.0 ml/min
 PT PAN              ------------------    -----------------   0.0 ml/min
       0.000 mg      <Sampling>
                                0.5 sec
```



```
<Sample>             <Comment>            <Temp.program[C]  [C/min]   [min]>
 15jan97b             96-15tga2            1*  20.0- 550.0     5.00    0.00
      19.613 mg      ------------------    2  550.0-  20.0   100.00    0.00
    ( 19.613 mg)     ------------------   <Gas>
<Reference>          ------------------    nitrogen          300.0 ml/min
 pt pan              ------------------    -----------------   0.0 ml/min
       0.000 mg      <Sampling>
                                0.5 sec
```



APPENDIX L

THERMO-GRAVIMETRIC ANALYSES OF CANISTER SLUDGE IN A HELIUM ATMOSPHERE

B. J. Makenas, R. B. Baker, and J. M. Tingey

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APPENDIX L

THERMO-GRAVIMETRIC ANALYSES OF CANISTER SLUDGE IN A HELIUM ATMOSPHERE

Thermo-Gravimetric Analyses (TGA) monitors the weight of a sample as temperature is increased. When volatile species are evolved, the change is manifest as a loss in sample weight. TGA runs were targeted to determine the amount of residual water present in sludge [which might accompany fuel in a Multi-Canister Overpack (MCO)] after exposure to Hot Vacuum Drying (HVD) temperatures. Previous TGA data from floor sludge and from particulates found in fuel shipping containers (References L2 and L3) demonstrated no residual water above 300 °C. The most recent TGA runs discussed herein were made under a helium atmosphere with no provision for compositional analysis of off-gas. The temperature was raised from room temperature to 1000 °C with hold points at temperatures of particular interest. Four subsamples were run, two from primary Sample 96-05 and two from 96-01. Of these two only 96-05 had previously been observed to be generating hydrogen bubbles while 96-01 was quiescent. The four TGA runs to date have produced quite variable data.

Figure L1 illustrates the basic features of the TGA curves acquired to date:

I. Most of the weight loss occurs early at 50 °C indicating the loss of free water.

II. A small weight loss occurs at 100 °C.

III. A more significant weight loss at 300 °C.

IV. Gradual weight increase due to oxidation of the sample even under a He cover gas.

V. A false indication of a weight gain as 1000 °C is approached (due to buoyancy or instrument effects).

VI. A weight loss perhaps due to strongly bound water. This may overlap the weight gains in V above.

VII. Continued oxidation of the sample.

VIII. Recovery of the false weight gain seen in V above.

Figure L2 shows a TGA run which is quite featureless from a weight loss point of view with a high temperature weight reduction only at 300 °C. Figure L3 is similar to Figure L1 discussed above. Figure L4 however shows, for sample 96-05, a significant weight loss at high temperature. The implication here is a 6% residual water content (by weight) for canister sludge above 300 °C. It should be noted that, for this latter run, an air ingress to the TGA apparatus occurred inadvertently during the 100 °C portion of the curve which may interject some caution in the use of these data even though no weight increase due to reaction with air was seen.

X-RAY DIFFRACTION

X-Ray Diffraction (see Appendix J) has been run for one subsample from each of the two primary samples used in these TGA runs, 96-01 and 96-05. This technique gives a determination of the crystalline phases present but does not yield quantitative concentrations. Both samples were found to consist primarily of uranium oxides (UO_2 and U_4O_9) with no indications of hydride, or uranium metal phase. A subsequent run using a subsample of 96-05, spiked with an Al_2O_3 standard, demonstrated the accuracy of the angles ascribed to each X-ray peak. A very small indication of some $UO_3 \cdot 2H_2O$ was noted for Sample 96-05 during this run.

SLUDGE COMPOSITION

Uranium concentrations ascertained by several methods are given in Appendices C through E. The two samples discussed here were found to consist primarily of uranium (>60% of dry sludge). This is far in excess of the uranium content of K East Basin floor sludge (Reference L2). If the sludge is indeed primarily oxide, then a significant portion of the non-uranium constituents could be oxygen bound to uranium.

DISCUSSION

The lack of identified hydride or metal in the sludge contradicts a previous finding (see Appendix B) of fission gases xenon and krypton (along with hydrogen gas) during the observed bubbling of sludge. The liberation of fission gas would suggest corrosion of metal fuel or of hydride to oxide. TGA runs to date have not been consistent in weight loss history. Subsampling of sludge is always subject to issues related to inhomogeneity. If gas bubbles and high temperature water losses are due to a few discrete reactive sludge particles the various phenomena may not be manifest in or expected in all samples. It is also possible that ion exchange beads (previously found in floor sludge) could be in the sludge samples. Decomposition of such beads could explain weight losses at temperatures of 350 °C to 400 °C although no such beads were visually observed in the samples. The reader will find some additional discussion of TGA for helium atmospheres in the Appendix M.

REFERENCES

L1. B. J. Makenas and R. B. Baker, "Highlight Report on Observations of Bubbles in K East Basin Canister Sludge During Analysis," September 1996.

L2. B. J. Makenas, et al., *Analysis of Sludge from Hanford K East Basin Floor and Weasel Pit*, WHC-SP-1182, April 1996.

L3. J. Abrefah and S. C. Marschman, "Highlight Report: Analysis of Filter Residues from K West Fuel Sample SFEC10.4366," April 1996.

L4. E. D. Jenson, Letter to K. L. Silvers, "XRD Examination of K Basin Samples," October 28, 1996.

Figure L1. Thermo-Gravimetric Analysis from Sample 96-05 R. Original Sample Mass was 40.1 mg.

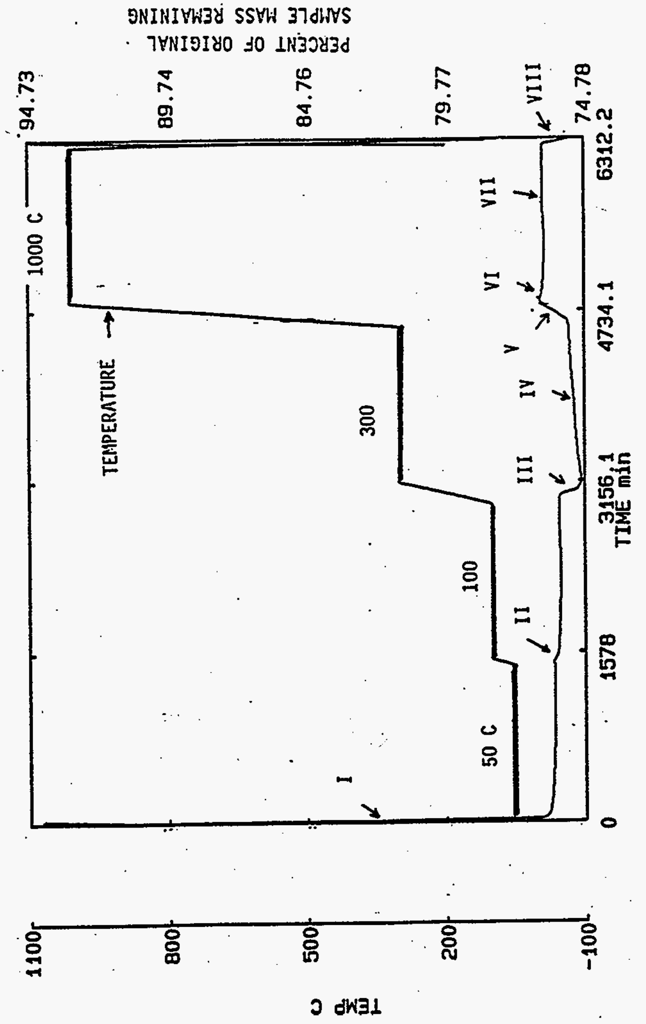


Figure L2. Thermo-Gravimetric Analysis from Sample 96-01 R. Original Sample Mass was 60.3 mg.

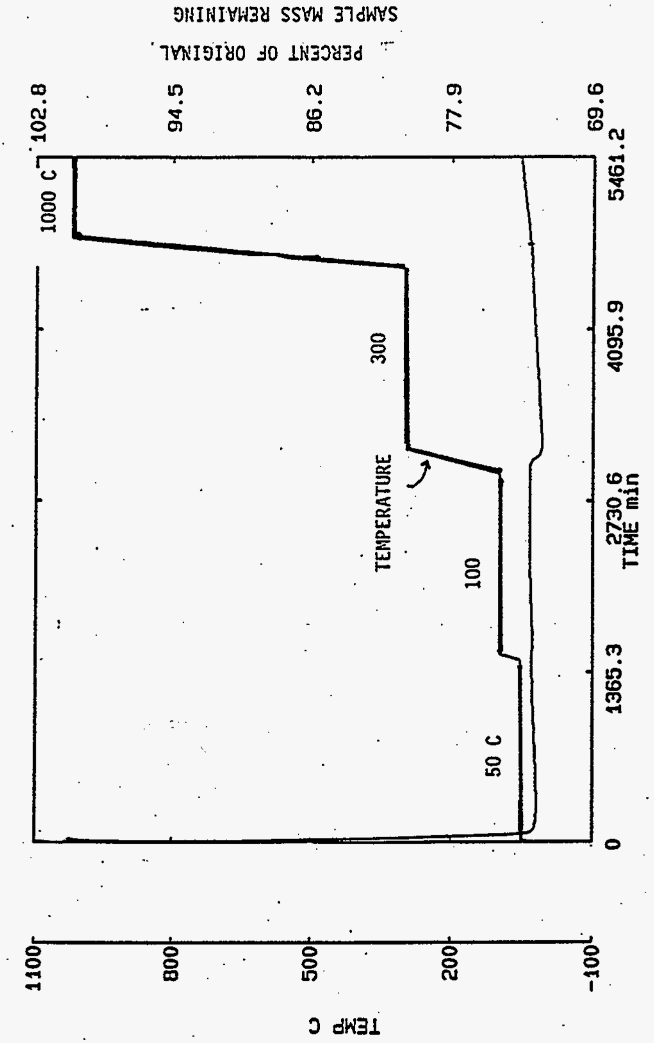


Figure L3. Thermo-Gravimetric Analysis from Sample 96-01.
Original Sample Mass was 19.4 mg.

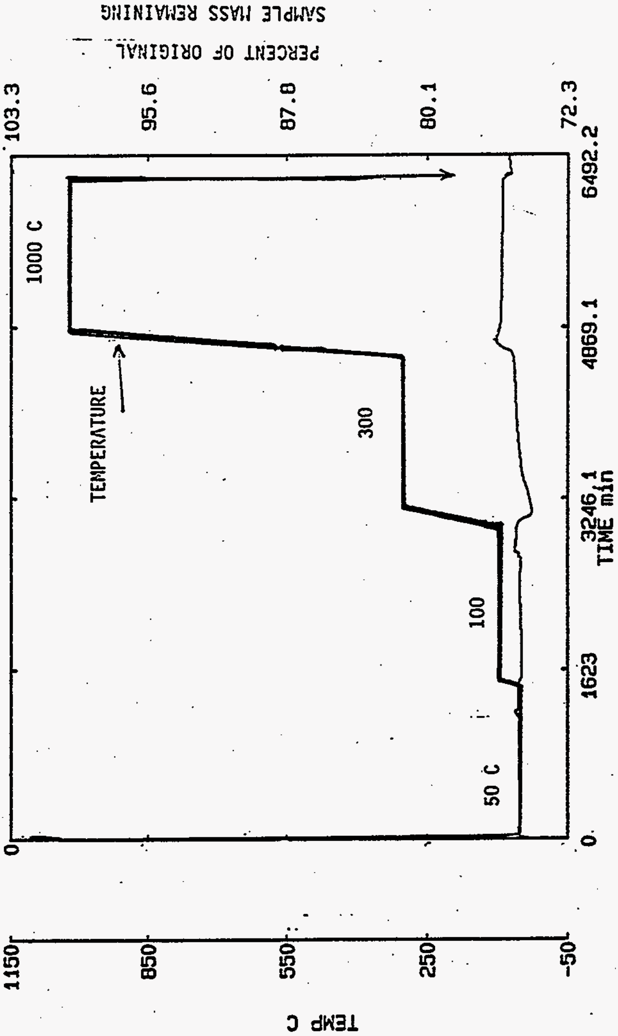
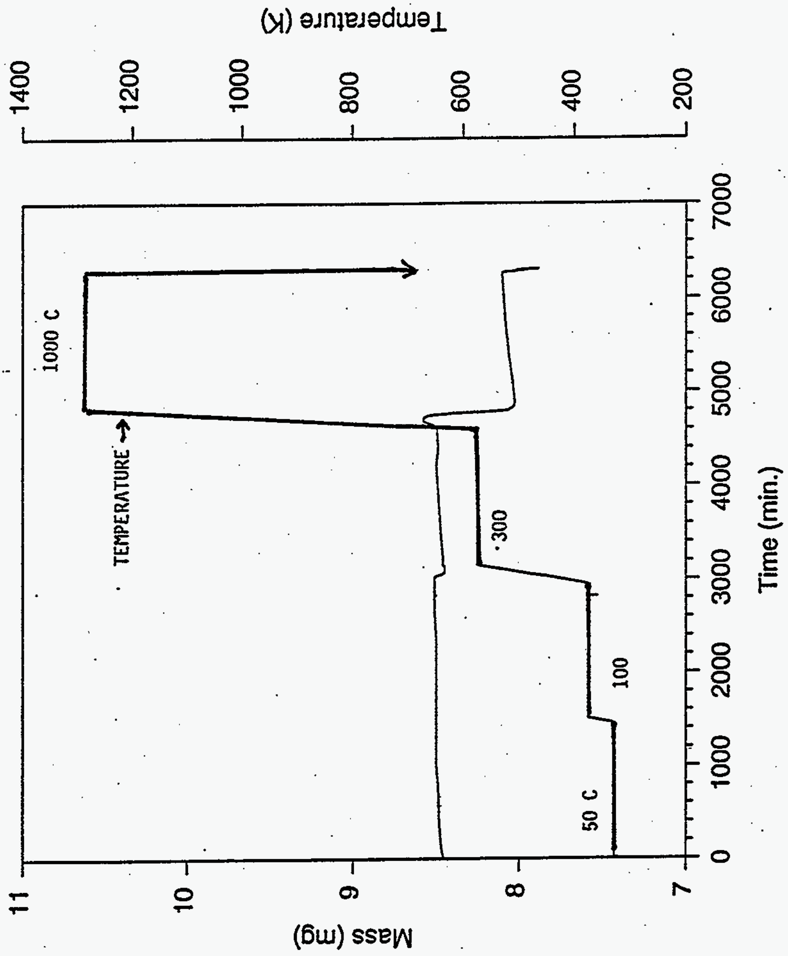


Figure L4. Thermo-Gravimetric Analysis from Sample 96-05.
Original Sample Mass was 10.5 mg.



APPENDIX M

THERMO-GRAVIMETRIC ANALYSIS IN VACUUM

(Three highlight reports summarizing Thermo-Gravimetric Analysis
for 14 samples of K East canister sludge)

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Highlight Report: Drying Behavior of K-East Canister Sludge

J Abrefah
HC Buchanan
SC Marschman
BJ Makenas (Duke Engineering & Services of Hanford)

March 1997

Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest National Laboratory
Operated for the U.S. Department of Energy
by Battelle

SNFCT97:025:R01

SUMMARY

Measurements have shown the water content (physically plus chemically bound) of five K-East canister sludge samples to range between 6.1 wt% and 18.9 wt%. The minimum and maximum water contents corresponded to samples removed from the same canister sample. The bound water released from the samples at temperatures above 50°C ranged from less than 1 wt % to 5 wt%. Results of the measurements are summarized in the following table. Water content based on the thermogravimetric measurements at higher temperature (greater than 300°C) represents a lower bound due to the ongoing sample oxidation in the test environment. Correspondingly, the results based on the mass spectrometer data for water released at high temperatures represent an upper bound due to release of trapped water in the system.

At present the data are insufficient to develop a composite drying curve for the sludge or even a meaningful average because of sample-to-sample variability. These results will be updated upon completion of similar measurements of the second group of five samples from the total of fourteen.

K-East Canister Sludge Drying Results Sample Weight Change (% of Initial Weight)

Run #	RM to 50°C	50°C to 75°C	75°C to 300°C	300°C to 430°C	430°C to 630°C
(A) Based On Thermogravimetric Data					
1	0.38	0.23	0.71	NE	NE
2	0.06	0.34	0.28	NE	0.04
3	0.62	NE	0.29	0.05	0.02
4	0.19	0.13	3.03	1.20	0.16
5	0.18	0.12	1.62	0.31	0.25
(B) Based On Mass Spectrometer Data					
1	NE	NE	1.66	0.93	NE
2	NE	NE	0.60	0.63	0.73
3	0.27	0.16	1.58	1.94	1.38
4	NE	NE	2.06	9.12	2.12
5	NE	NE	NE	NE	NE

NE = Not Evaluated
RM = Ambient Temperature

SNFCT97:025

INTRODUCTION

Storage of Spent Nuclear Fuel (SNF) in the K-Basins has generated corrosion products (i.e., oxides of uranium), some of which may accompany the SNF elements when they are loaded into the Multi-Canister Overpacks (MCOs). The oxides of uranium will provide a large surface area for adsorption of water. Additionally, the oxides can chemically react with water to generate hydrates. These two forms of water (i.e., physically bound and chemically bound) will be subjected to drying steps during the Integrated Process Strategy (IPS) treatment of the SNF for the interim dry storage. Data on the drying behavior of these forms of moisture are therefore, needed in support of the IPS.

This highlight report discusses results of drying samples of K-East canister sludge using a thermogravimetric/mass spectrometer (TGA/MS) system. This system monitors the weight change of the samples due to volatilization of species such as water and in some instances oxidation of the sample. The volatilized species were monitored by the quadrupole mass spectrometer. The tests were conducted to determine the relative amount of water in the samples and the fractional release of the moisture at certain desired temperatures. These TGA runs are complementary to those performed previously for smaller samples on an instrument without a mass spectrometer capability (Makenas 1996).

EXPERIMENTAL

The schematic of the TGA/MS testing system that was used in the drying studies is shown in Figure 1. A detailed description of the system is in the report of Marschman and Abrefah (1996).

The K-East canister subsamples used for the drying tests came from a batch of nine K-East canister sludge samples shipped to 325 Building for characterization. For each TGA run, about one gram of these samples was loaded into an alumina crucible, weighed and mounted on the thermogravimetric stick shown in Figure 1. The alumina crucible was covered with a perforated lid to prevent spread of the sample after drying. All the samples were dried in a vacuum atmosphere with ultra pure helium environment except run #33. Run #33 was performed in ultra pure helium at one atmosphere pressure. The drying was conducted within a temperature range of ambient to 1000°C with a typical temperature profiles such as the following:

(a) Heated at constant rate of 1°C/min. to a temperature of about 50°C and held it at this temperature for 8 hours.

(b) Heated at constant rate of 1°C/min. to a temperature of about 75°C and held it at this temperature for 10 hours.

(c) Heated at constant rate of 1°C/min. to a temperature of about 300°C and held it at this temperature for 12 hours.

(d) Heated at constant rate of 1°C/min. to a temperature of about 431°C and held it at this temperature for 12 hours.

(e) Heated at constant rate of 2°C/min. to a temperature of about 630°C and held it at this temperature for 6 hours.

The sample weight change was continuously monitored by the electrobalance throughout the test and the off-gas stream was analyzed by the attached quadrupole mass spectrometer. The sample was weighed after the test to estimate the total weight loss.

RESULTS AND DISCUSSION

Five sludge samples have been tested, three from the canister sample 96-05, and two from canister sample 96-01. A summary of the test samples and results are listed in Tables 1 and 2. The NE (not evaluated) in the tables indicate that the weight loss could not be determined from the electrobalance or the mass spectrometer (MS) data. In the case of the electrobalance, the reason for not evaluating the weight loss was that either no measurable weight change was detected or the weight increase due to oxidation of the sample affected the weight loss measurement. The MS data, on the other hand, occasionally could not be analyzed due to high background signal of moisture which obscured the moisture from the sample. A large fraction of the physically adsorbed water (free water) on the sample vaporized during the pump down to vacuum condition. This happened before the sample was heated and the weight loss could not be monitored by the data acquisition system.

Before and after weight measurements in Tables 1 and 2 indicate that samples for TGA runs 31, 36 and 37 lost about 19, 17 and 18 wt%, respectively, of their initial weight. Samples for TGA runs 32 and 33, however, lost about 6.1 and 6.4 wt%, respectively. The difference in these two groups of samples is the handling and storage environment. Samples for TGA runs 32 and 33 were extracted from the primary container and stored in a hot cell for several days before the TGA test samples were extracted. On the other hand, samples for TGA runs 31, 36 and 37 were taken directly from the primary samples. Thus storage time does affect the water content of small samples.

The percent weight loss for the samples with the same handling and storage environment are very reproducible within the experimental spread of data. This indicates that fractions of the physically adsorbed water (i.e., free water) and chemically bound water (hydrates and hydroxides) may not be significantly different from sample-to-sample. It can also be inferred that a significant fraction of water was physically adsorbed water. Comparing the results of the two groups, the fraction of physically adsorbed water can be at least 15 wt%.

Figures 2 through 6 show plots, for each run, of weight change data and the MS data for mass/charge ratio of 18, which is water (H_2O). In Figure 2 (TGA run 31), the weight change data show a small weight loss between ambient temperature and 50°C followed by another small loss between 50°C and 75°C. A more significant weight loss occurred during the ramp to 300°C. The sample started gaining weight when the temperature reached 300°C. The weight gain due to oxidation of the sample at this temperature and above was significant enough to offset any weight loss due to release of moisture from the sample. The total weight gain of the sample from Figure 2 was about 22 mg. The sludge sample, therefore, was not in its fully oxidized state at the beginning of the run, but eventually became fully oxidized as evidenced by the leveling off of the weight gain. The water loss from the sample within the temperature range of 75°C to 300°C was well above the background water signal. The MS signal shows the maximum water release rate occurred at about 250°C and continuous water release from the sample when it was still gaining weight. Another significant amount of water-release from the sample was measured by the MS within the temperature range of 300°C and 430°C. During the ramp to 1000°C, the MS signal indicated a large release of moisture, but most of that signal was probably due to increased background moisture. The small shoulder of that portion of curve, however, may be a release of water from the sample.

In Figure 3 (TGA run 32), the weight change curve shows a small weight loss between ambient temperature and 50°C and comparable weight losses within the temperature ranges of 50°C to 75°C, and 75°C to 250°C. The sample for this run showed small weight gain at temperatures above 300°C (due to oxidation) and the net weight change of the sample was almost zero. The MS signal curve indicates water released within the temperature ranges of 75°C to 250°C, 250°C to 430°C, and 430°C to 630°C.

Figures 4a and 4b show the drying results of TGA run 33. This run was plotted in two figures because the run was shut off accidentally at the hold point of 75°C (Figure 4a), and was restarted for the remaining segments (Figure 4b) of the test. This test was performed in ultra pure helium at atmospheric pressure and the helium flow rate was 200 cc/min. The sample lost significant weight between ambient temperature and 50°C and no measurable weight loss between 50°C and 75°C (Figure 4a). Figure 4b shows a significant weight loss between 75°C to 250°C followed by gradual weight changes at higher temperatures. The MS signal shows moisture release at each segment of temperature range (Figure 4b).

Figures 5 and 6 (for runs 36 and 37) show weight loss curves which are very similar to one another. A small weight loss occurred between ambient temperature and 75°C, a significant weight loss between 75°C and 300°C, a slight increase in weight at 300°C, and gradual weight loss above 300°C. The MS signal for these two runs was very noisy and the computationally smoothed curves may not accurately represent actual water released. Calculations of the weight loss from the MS data for these runs have large errors.

Calculations of the weight loss from the thermogravimetric (TG) measurements are listed in Table 1 and calculations from the MS data are listed in Table 2. The MS calculations were from the mass/charge ratio of 18 (H_2O) only. The small (but not negligible) fraction due to cracking of water molecules to OH radicals was not accounted in the preliminary calculation. The weight loss data in the two tables indicate that the chemisorbed water fraction in the sludge samples tested ranges between 3 to 5 weight percent and a large fraction of about 15 weight percent is physically absorbed water. It appears from the data that four different hydrate species are present in the sludge that can be released within the temperature ranges studied. Significant weight fractions of hydrates decomposed within the temperature ranges of 75°C to 300°C and 300°C to 430°C.

Additional runs using samples from the nine primary K-East canisters are being performed and the results of those tests will be reported. These next tests will increase the sludge drying characteristics data which may improve our understanding of the forms of hydrates that formed on the K-East canister sludge.

REFERENCES

Makenas BJ and RB Baker. 1996. *Accelerated Examinations of K East Basin Canister Sludge.* DESH-9655840, Duke Engineering & Services of Hanford, Richland, Washington.

Marschman SC and J Abrefah. 1996. *System description for the TGA/DSC/MS system.* SNFCT96:025:R00, Pacific Northwest National Laboratory, Richland, Washington.

Table 1. TG Drying Results of KE Canister Sludge

Run #		Status (Test Start Date)	System Press. (mTorr)	Sample				Sample Weight Loss (mg) for Different Temperature Ranges (°C)				
TGA	KE Sludge			ID	Weight Before Test (mg)	Weight After Test (mg)	Weight Loss (mg)	RM[a] to 50	50 to 75	75 to 300*	300* to 430	430 to 630**
31	1	12/17/96	181	96-05	1384	1123	261	3.915	3.163	9.759	NE[b]	NE
32	2	01/06/97	93	96-05	1017	955	62	0.569	3.470	2.846	NE	0.434
33	3	01/10/97	760x10^3	96-05	1000	836	64	6.216	NE	2.892	0.542	0.229
36	4	02/03/97	71	96-01	643	534	109	1.215	0.843	19.503	7.700	1.054
37	5	02/11/97	121	96-01	1054	860	194	1.898	1.265	17.078	3.268	2.635

Table 2. MS Drying Results of KE Canister Sludge

Run #		Status (Test Start Date)	System Press. (mTorr)	Sample				Sample Weight Loss (mg) for Different Temperature Ranges (°C)				
TGA	KE Sludge			ID	Weight Before Test (mg)	Weight After Test (mg)	Weight Loss (mg)	RM[a] to 50	50 to 75	75 to 300*	300* to 430	430 to 630**
31	1	12/17/96	181	96-05	1384	1123	261	NE	NE	23	12.84	NE
32	2	01/06/97	93	96-05	1017	955	62	NE	NE	6.11	6.44	7.47
33	3	01/10/97	760x10^3	96-05	1000	936	64	2.71	1.56	15.77	19.38	13.81
36	4	02/03/97	71	96-01	643	534	109	NE	NE	13.22	58.61	12.63
37	5	02/11/97	121	96-01	1054	860	194	NE	NE	NE	NE	NE

* 250°C for Runs 32 and 33; ** 1000°C for Run 31; (a) RM = ambient temperature; NE = not evaluated

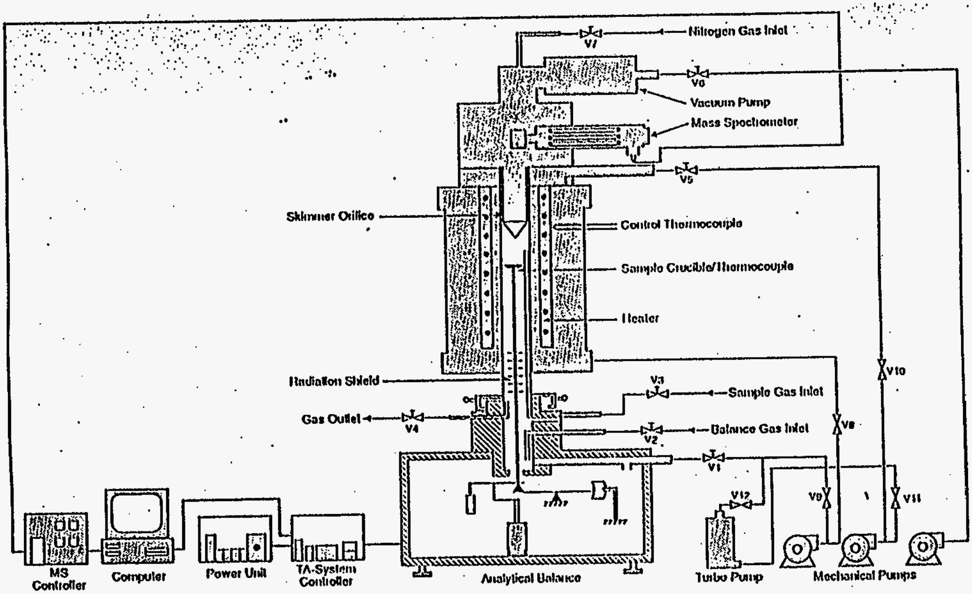

Figure 1. Schematic of the TGA/MS System

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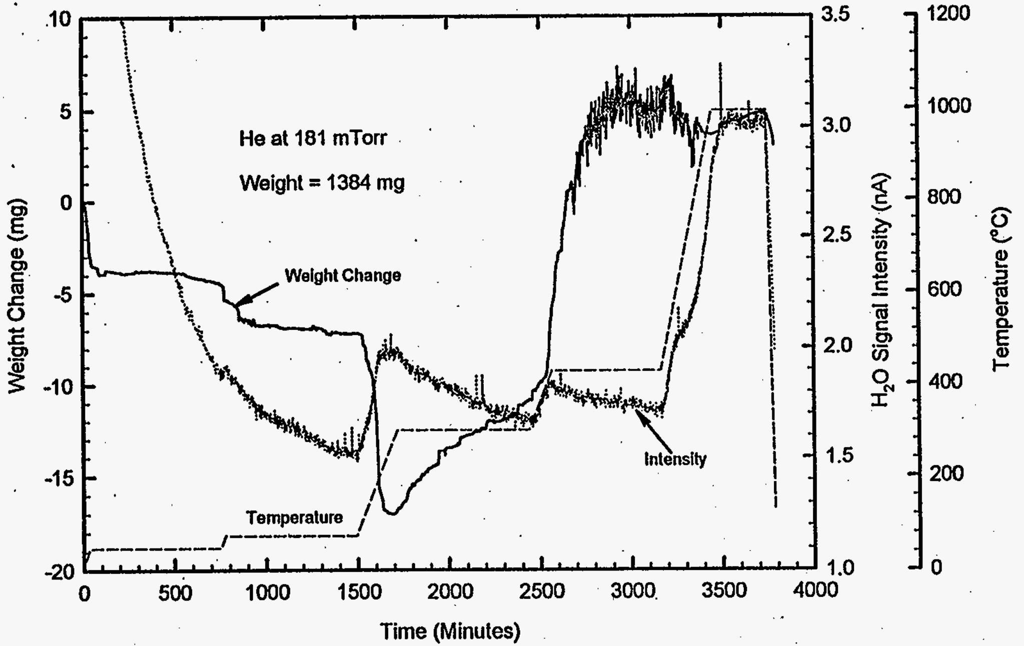

Figure 2. TGA Run 31 Plot Showing Weight Change, MS Signal for H_2O and Temperature versus Time. KE Canister Sludge 96-05.

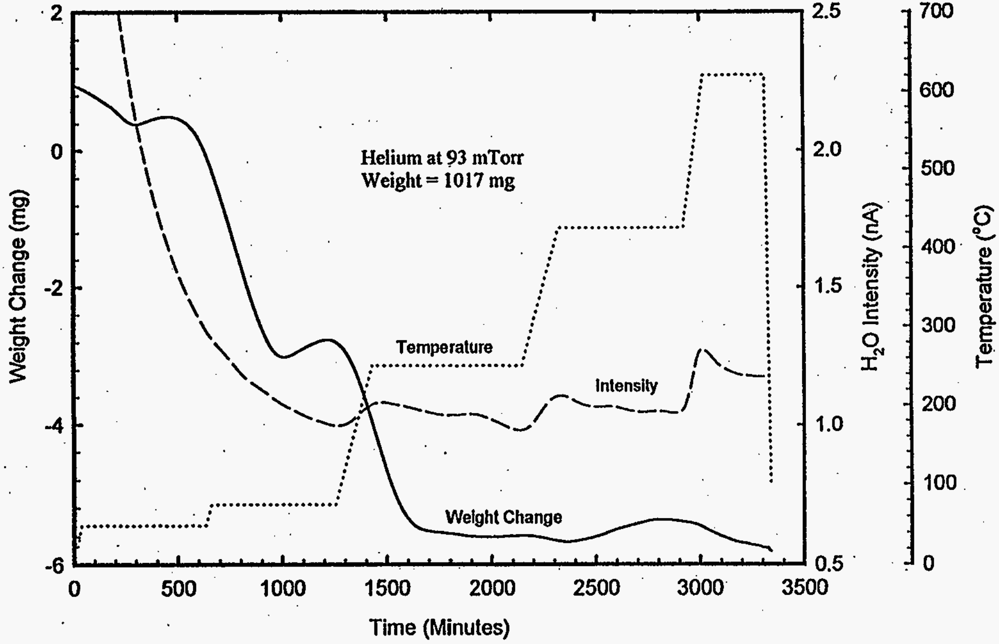

Figure 3. TGA Run 32 Plot Showing Weight Change, MS Signal for H_2O and Temperature versus Time. KE Canister Sludge 96-05.

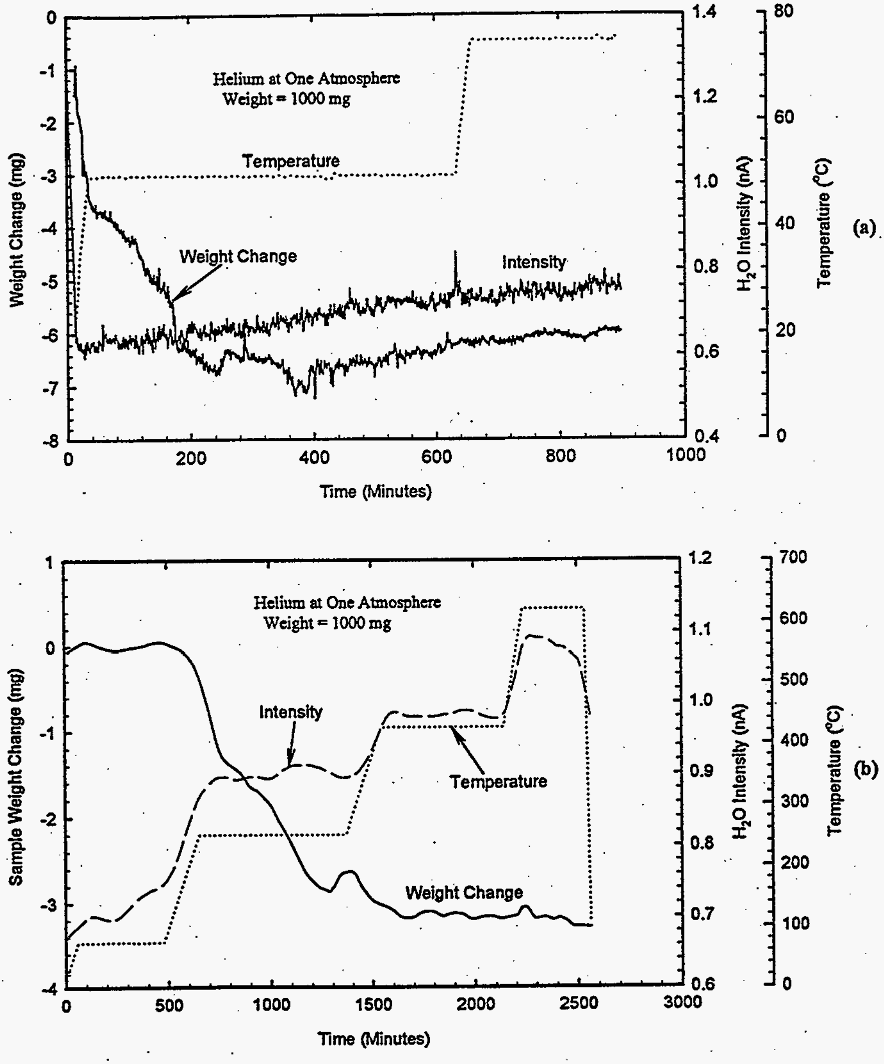

Figure 4. TGA Run 33 Plots Showing Weight Change, MS Signal for H_2O and Temperature versus Time; (a) First two Segments and (b) Remaining Segments. KE Canister Sludge 96-05

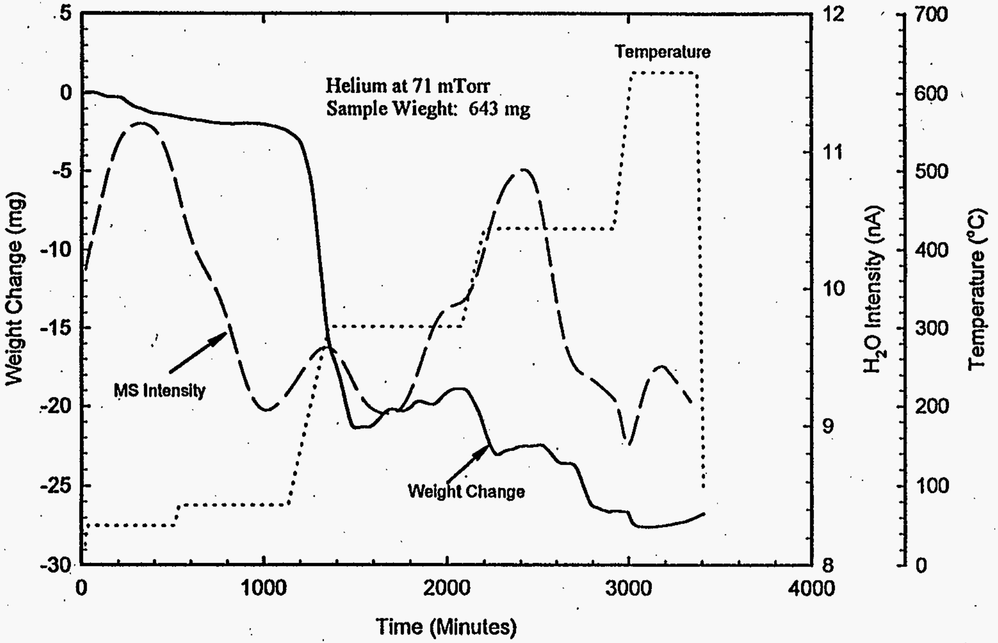

Figure 5. TGA Run 36 Plot Showing Weight Change, MS Signal for H_2O and Temperature versus Time. KE Canister Sludge 96-01.

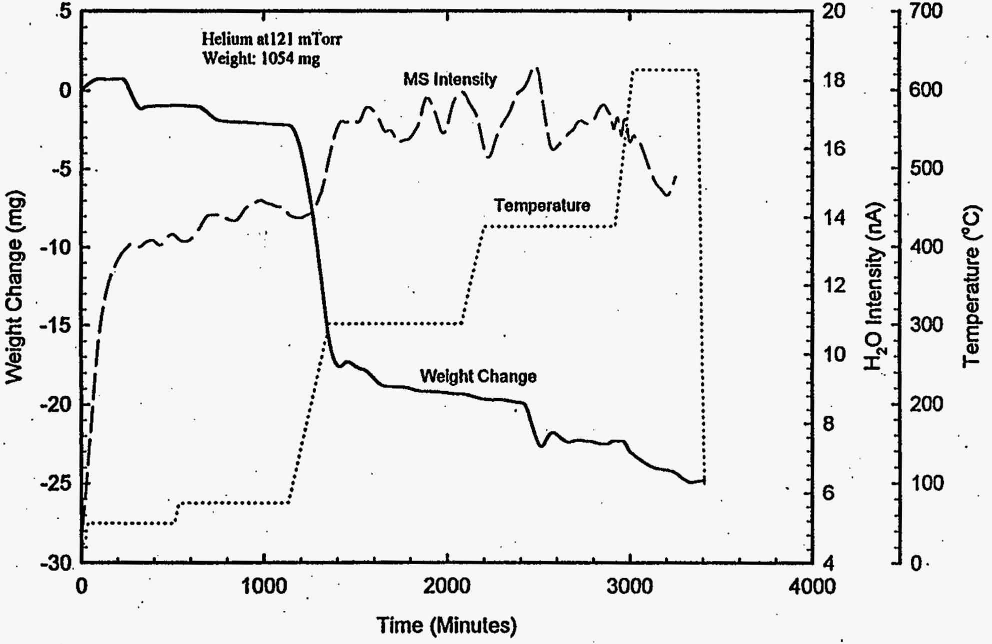

Figure 6. TGA Run 37 Plot Showing Weight Change, MS Signal for H_2O and Temperature versus Time. KE Canister Sludge 96-01.

Highlight Report: Second Report On Drying Behavior of K-East Canister Sludge

J Abrefah
HC Buchanan
SC Marschman
BJ Makenas (Duke Engineering & Services of Hanford)

April 1997

Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest National Laboratory
Operated for the U.S. Department of Energy
by Battelle

SNFCT97:042:R00

SUMMARY

Measurements have shown the water content (physically[1] plus chemically[2] bound) of the second group of five K-East canister sludge samples to range between 14.8 wt% and 76.9 wt%. The minimum water content corresponded to sample 96-13, and sample 96-08 showed the maximum water content. The hydrates constitutes about 2.1 wt% to 5.8 wt% of the total water content of the sludge samples. Most of the physically bound water was assumed to be released from the samples at ambient temperature during the time the system was pumped down to vacuum conditions of about 40 mTorr. The release of water resulting in sample weight loss above ambient temperature was ascribed to thermal decomposition of hydrates and a summary of these measurements is listed in the table below. The largest fraction of the hydrated species thermally decomposed within the temperature range of 75°C to 300°C. Quantitative analysis of the mass spectrometer (MS) signal for water could not be done to determine corresponding weight losses due to high background water interference.

These results will be updated upon completion of similar measurements of the last group of samples from the total of fourteen. After the fourteenth run there may be sufficient data to develop a composite drying curve for the sludge and to extract kinetic information from the data.

K-East Canister Sludge Drying Results Sample Weight Change (% of Initial Weight)

Run #	RM to 50°C	50°C to 75°C	75°C to 300°C	300°C to 425°C	425°C to 625°C
6	NE	0.30	0.79	0.21	0.82
7	NE	NE	0.92	NE	NE
8	0.20	0.29	2.42	0.21	0.18
9	0.16	0.31	1.89	0.15	0.14
10	0.55	0.93	3.70	0.37	0.22

NE = Not Evaluated
RM = Ambient Temperature

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[1] Physically bound water is the "free water" which is physically absorbed and adsorbed by the pores and surface of the sludge.

[2] Chemically bound water is the hydrate(s) and hydroxides formed by the reaction of water with the sludge and will henceforth be referred to as "hydrates".

INTRODUCTION

Storage of Spent Nuclear Fuel (SNF) in the K-Basins has generated corrosion products (i.e., oxides of uranium), some of which may accompany the SNF elements when they are loaded into the Multi-Canister Overpacks (MCOs). The oxides of uranium will provide a large surface area for adsorption of water. Additionally, the oxides can chemically react with water to generate hydrates. These two forms of water (i.e., physically bound and the hydrates) will be subjected to drying steps during proposed Integrated Process Strategy (IPS) treatment of the SNF for the interim dry storage. Data on the drying behavior of these forms of moisture are therefore, needed in support of the IPS.

This highlight report discusses drying results of five additional samples of K-East canister sludge using a thermogravimetric/mass spectrometer (TGA/MS) system. These TGA runs are complementary to those performed previously for smaller samples on an instrument without a mass spectrometer capability (Makenas 1996) and the first five results reported by Abrefah et al. (1997). The system monitors the weight change of the samples due to volatilization of species such as water and in some instances oxidation of the sample. The volatilized species were monitored by the quadrupole mass spectrometer. The tests were conducted to determine the relative amount of water in the samples and the fractional release of the moisture at certain desired temperatures.

EXPERIMENTAL

The testing system that was used in the drying studies is shown in the highlight report by Abrefah et al. (1997) and a detailed description of the system is in the report of Marschman and Abrefah (1996).

The K-East canister subsamples used for the drying tests came from a batch of nine K-East canister sludge samples shipped to 325 Building for characterization. For each TGA run, about one gram of a sample was loaded into an alumina crucible, weighed and mounted on the thermogravimetric stick. The alumina crucible was covered with a perforated lid to prevent spread of the sample after drying. All the samples were dried in a vacuum atmosphere with ultra pure helium environment. The drying was conducted within a temperature range of ambient to 625°C with a typical temperature profiles such as the following:

(a) Heated at constant rate of 1°C/min. to a temperature of about 50°C and held it at this temperature for 8 hours.

(b) Heated at constant rate of 1°C/min. to a temperature of about 75°C and held it at this temperature for 10 hours.

(c) Heated at constant rate of 1°C/min. to a temperature of about 300°C and held it at this temperature for 12 hours.

(d) Heated at constant rate of 1°C/min. to a temperature of about 425°C and held it at this temperature for 12 hours.

(e) Heated at constant rate of 2°C/min. to a temperature of about 625°C and held it at this temperature for 6 hours.

The test pressure was established by flowing ultra high purity helium gas through the sample environment and throttling a valve to reduce the pumping speed of the sample environment. This step was performed after achieving the lowest vacuum conditions (about 40 mTorr) of the system.

The sample weight change was continuously monitored by the electrobalance throughout the test and the off-gas stream was analyzed by a quadrupole mass spectrometer. The sample lost weight during the pump down to vacuum conditions which was not recorded by the data acquisition system. That weight loss was calculated from the before and after test weight measurements.

RESULTS AND DISCUSSION

Five additional sludge samples have been tested, two from the canister sample 96-08, two from canister sample 96-13 and one from canister sample 96-15. A summary of the test samples and results are listed in Table 1. The NE (not evaluated) in the tables indicate that the weight loss could not be determined from the electrobalance. The reason for not evaluating a specific weight loss was that either no measurable weight change was detected or the weight increase due to oxidation of the sample affected the weight loss measurement. The MS data, in general, could not be analyzed for these five runs due to high background moisture which obscured the moisture from the sample. A large fraction of the physically adsorbed water on the sample vaporized during the pump down to vacuum condition. This happened before the sample was heated and the weight loss could not be monitored by the data acquisition system.

Before and after weight measurements in Table 1 indicate that samples for TGA runs 38, and 39 (taken from canister sample 96-08) lost about 76.9 and 28.6 wt%, respectively, of their initial weights. These samples had enough water to decrease the sample temperature from ambient conditions to about 4°C during the pump down period. Samples for TGA runs 40 and 41 (taken from canister 96-13), however, lost about 15 wt%. The last sample (TGA 42) lost 16.6 wt% of its initial weight.

The hydrated water for the samples that could be analyzed ranges between 2.1 to 5.8 wt% (Table 1) and the characteristics of the thermal decomposition and release kinetics are shown in Figures 1 through 5. For each run, the weight change and the MS data for mass/charge ratio of 18, which is water (H_2O) are plotted. The MS data for all the plots were comparable to the background noise and analysis of the MS measurement will be very inaccurate. Hence the MS data calculations to give corresponding sample weight loss for the temperatures ranges listed in Table 1 will not be included in this highlight report. Runs 40 through 42 show considerably less interference from oxidation or absorption of water in the system than all the previous runs to date. This is due to addition of a chemical filter to the system background gas supply (helium) that reduced water and oxygen impurities in the gas.

In Figure 1 (TGA run 38), the weight change data show an increase during the ramp to 50°C. This increase in weight at such low temperatures may be due to pick up water by the sample. The source of the water might have been accumulated water in the system that originally was released from the sample during the ambient temperature drying. The oxidation of the sample could also effect sample weight gain but at these low temperatures, that contribution should be negligible unless there was a fraction of the sludge sample which was highly reactive (such reactions have not been observed). The sample showed a small weight loss in the temperature ranges 50°C to 75°C and 300°C to 425°C. A more significant weight loss occurred within the temperature ranges 75°C to 300°C, and 425°C to 625°C. The relatively significant weight loss at the last temperature segment is an exception to the rest of the test results from these five runs. This observation may be due to the low temperature weight gain of the sample. The sample may have reacted with the moisture released from the system to form hydrates and/or hydroxides. These newly formed hydrates and/or hydroxides might have contributed to the last temperature segment (425°C to 625°C) water release. The MS signal in the figure shows a high background noise which makes it very difficult to differentiate the water signal resulting from the sample.

Figure 2 (TGA run 39) shows a weight change curve that indicates weight loss between 75°C to 300°C. At the other temperature ranges, the sample showed no weight change, a small weight gain and a significant weight gain at temperatures above 300°C. The weight gain due to oxidation of the sample was significant enough to offset any weight loss due to release of moisture from the sample. The total weight gain of the sample above 300°C was about 23 mg. The overall data of this run was too noisy to be able to extract any accurate information. The technical source of the noise (a failing cooling pump on the system) was fixed prior to performing the next series of tests.

In Figure 3 (TGA run 40), the weight change curve shows a small weight loss between ambient temperature and 50°C followed by a weight losses within the temperature range of 50°C to 75°C. At the end of each of these temperature segments the weight change curve still showed a decreasing behavior (i.e., a non-zero slope). This can be attributed to slow decomposition of the higher temperature hydrates at these low temperatures. The weight loss measured within the next temperature range (75°C to 300°C) is the largest and there is an indication of a slope decrease for the weight change curve at about 219°C. The decrease in slope of the weight change indicates an increase in release rate of the moisture by the sample. Thus, assuming a constant source, the changes in slope can be used to determine the optimum temperature for thermal decomposition of this particular hydrated phase in the sludge sample. The sample for this run showed small weight losses at temperatures ranges 300°C to 425°C and 425°C to 625°C. The total weight loss measured at temperature above ambient was 3.3 wt% of the initial sample weight. Overall this sample lost a total of about 15 wt% of its initial weight, which calculates to be about 11.7 wt% of physically absorbed water (water removed at ambient temperature during system pump down) and 3.3 wt% chemically bound (water released during sample heat up to various temperature segments) water. The MS signal curve indicates water released within the temperature ranges considered, but the noise level was high enough to make any quantitative estimate invalid.

Figure 4 shows the drying results of TGA run 41. During this run the MS was not functional due to a filament failure. This figure only shows the weight change data. The sample lost significant weight between 75°C to 300°C, and measurable weight losses within the temperature ranges, ambient to 50°C, 50°C and 75°C, 300°C to 425°C and 425°C to 625°C. The behavior of a continuous decrease in weight at the two lower temperature segments was again observed in this run. The weight loss between 75°C to 300°C also show a change in slope at about 211°C, indicating an optimum decomposition temperature for the hydrate is around 211°C. The total weight loss of this run was also about 15 wt% and the fractional weight loss at temperatures above ambient was 2.7 wt%. Assuming the latter to be due to thermal decomposition of hydrated species, the physically absorbed and adsorbed water works out to be about 12.3 wt%. The TGA run 41 shows a good reproducible results compared to run 40 except the small changes in the fraction of hydrates measured. The samples for runs 40 and 41 came from the same canister and the difference in the hydrated fraction in both runs is an indication that not all the sludge is in a hydrated form and a testing study that uses small portions of the sample can be biased.

Figures 5 (TGA run 42) shows a weight loss curve which is very similar to runs 40 and 41. Small weight losses occurred within temperature ranges of ambient temperature to 50°C and 50°C to 75°C. A significant weight loss was measured between 75°C and 300°C. The temperature corresponding to the point where the weight change curve for this range changes slope occurred at about 218°C. The sample showed slight weight decreases within the temperature ranges of 300°C to 425°C and 425°C to 625°C. The MS signal for this run suffered from a high background signal which prevented quantitative analysis of the water signal corresponding to the weight losses. Qualitatively, the MS signal indicates release of water from the sample during measured weight losses.

The weight change data for reheating the sludge sample used in run 42, up to 1000°C is plotted in Figure 6. The figure indicates a decrease in the sample weight starting at about 645°C and continued to the end of the test. A change of slope for the decrease in weight occurred when the sample temperature reached about 1000°C. The total weight loss was 3.05 mg (about 0.42 wt% of initial sample weight). The decrease in sample weight upon reheating indicates that the release of all volatile species (including thermal decomposition of water) was not complete during run 42. The expanded plot of the last segment of Figure 5 (425°C to 625°C) shows a decreasing weight at the end of run 42 which is an indication that the process resulting in the weight loss was still happening when the run ended. If the weight loss at 625°C is identified to be a hydrate decomposition process then the same process may have contributed to the weight loss during the reheating test. On the other hand, the weight loss observed during the reheating can be due to release of a higher temperature hydrate and/or hydroxide. A decomposition of UO_3 to U_3O_8 at higher temperatures can also result in the weight loss observed during the reheating run.

Calculations of the weight loss for various temperature ranges are listed in Table 1. The weight loss data in the Table 1 (excluding run 39) indicate that the hydrated species in the sludge samples have a water content whose weight fraction ranges between 2.1 to 5.8 wt%. The largest fraction of hydrates decomposed within the temperature ranges of 75°C to 300°C. A detailed look at the weight change curve at the two lower temperature segments, ambient to 50°C and 50°C to 75°C indicates that the corresponding hydrates decomposed, may be the same (in small amounts) as the hydrate precisely decomposed within the temperature range of 75°C to 300°C. If this inference is true then we may be observing only three hydrated species in the runs. Thus, one hydrated species released between ambient and 300°C, a second one between 300°C to 425°C and a third hydrate above 425°C. The planned additional tests of the sludge samples from other K-East samples will increase the database and will improve our understanding of all the hydrated species observed in these runs and the runs reported earlier (Abrefah 1997).

The physically bound water content of the sludge samples was 11.7 wt% of the initial weight or greater. This calculation is based on the assumption that all the physically bound water was released and pumped out during the system pump down at ambient temperature. However, a small fraction of the physically bound water can remain in the sample to be released during the lower temperature segments of the tests. This high content of water in the sample indicates a very porous medium with a very high surface area for adsorption and absorption of water molecules.

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Table 1. TG Drying Results of KE Canister Sludge

Run #		Status (Test Start Date)	System Press. (mTorr)[*]	Sample				Sample Weight Loss (mg) for Different Temperature Ranges (°C)				
TGA	KE Sludge			ID	Weight Before Test (mg)	Weight After Test (mg)	Weight Loss (mg)	RM[*] to 50	50 to 75	75 to 300	300 to 425	425 to 625
38	6	02/19/97	70	96-08	917	212	705	NE	2.754	7.283	1.948	7.519
39	7	02/24/97	104	96-08	734	524	210	NE	NE	6.755	NE	NE
40	8	03/12/97	106	96-13	1159	986	173	2.369	3.313	28.080	2.475	2.041
41	9	03/19/97	125	96-13	768	654	114	1.265	2.349	14.548	1.175	1.084
42	10	03/24/97	96	96-15	723	603	120	3.976	6.690	26.744	2.711	1.626

RM = Ambient temperature
NE= Not evaluated

[*] These are the starting vacuum conditions of the runs and during the test the system pressure increases slightly due to increasing temperature and a balance between pumping speed and gas flow.

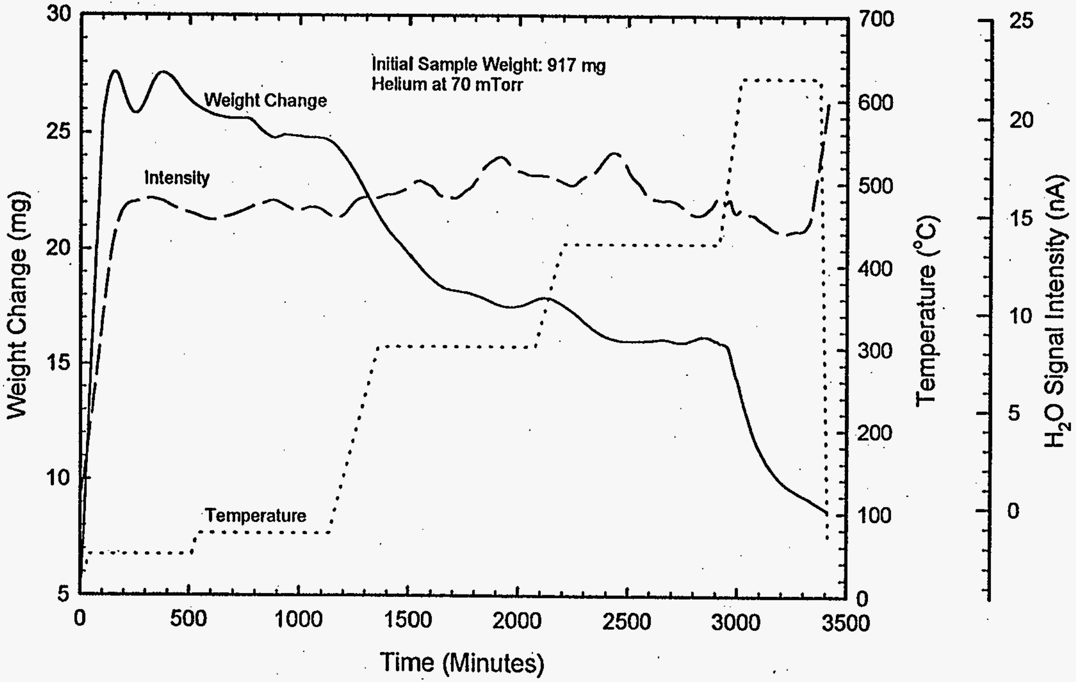

Figure 1. TGA Run 38 Plot Showing Weight Change, MS Signal for H_2O and Temperature versus Time. KE Canister Sludge 96-08.

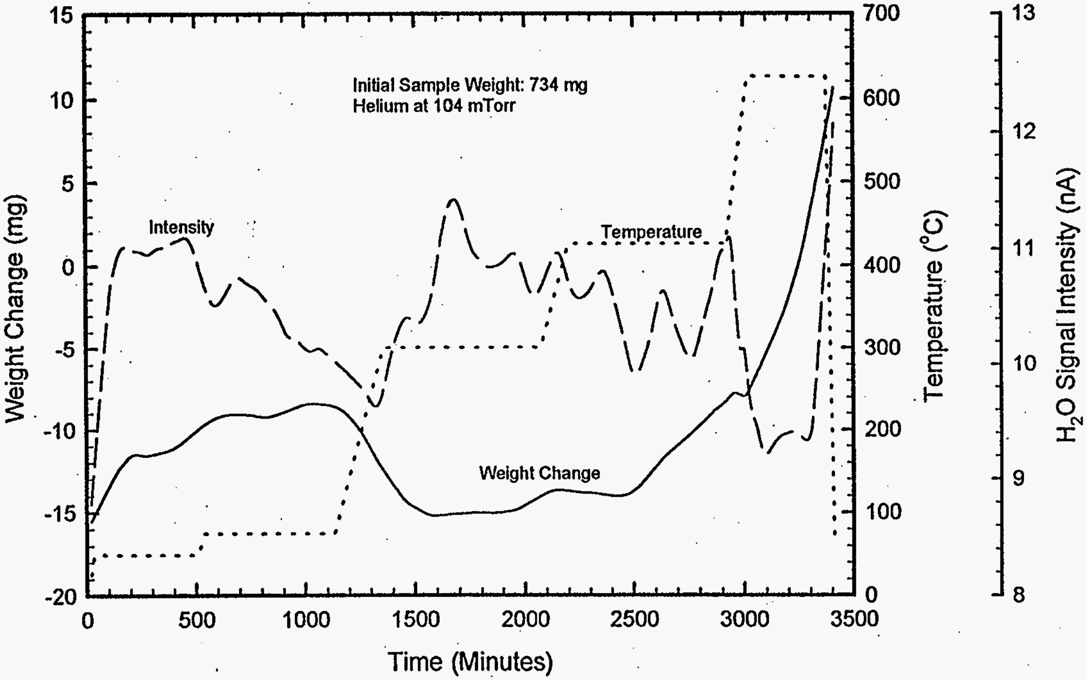

Figure 2. TGA Run 39 Plot Showing Weight Change, MS Signal for H_2O and Temperature versus Time. KE Canister Sludge 96-08.

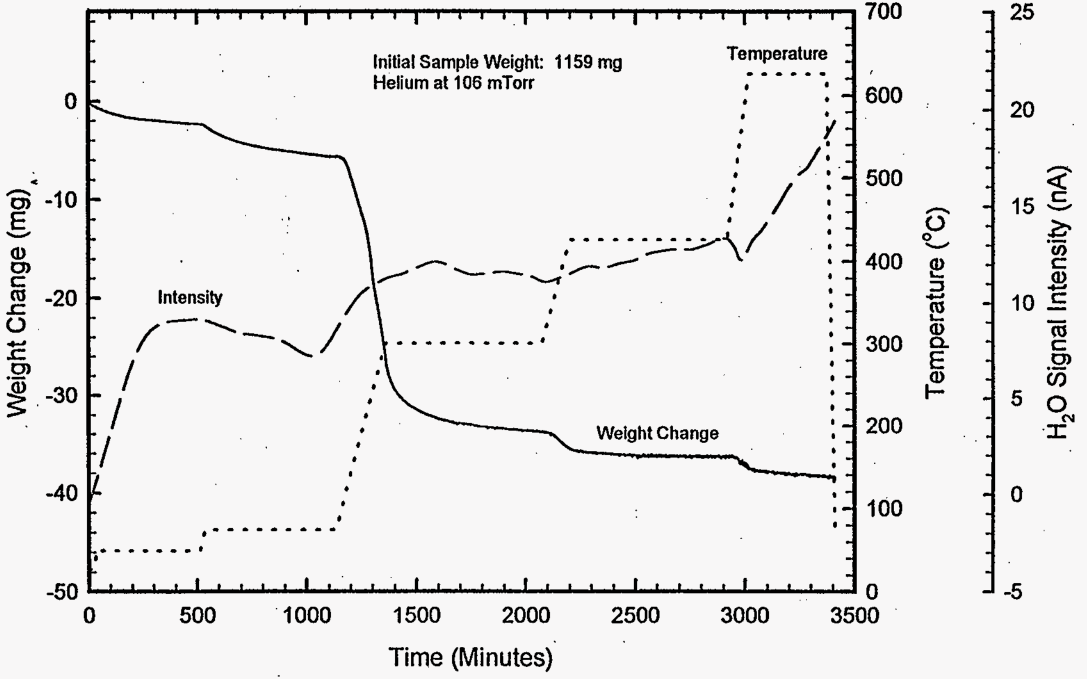

Figure 3. TGA Run 40 Plot Showing Weight Change, MS Signal for H_2O and Temperature versus Time. KE Canister Sludge 96-13.

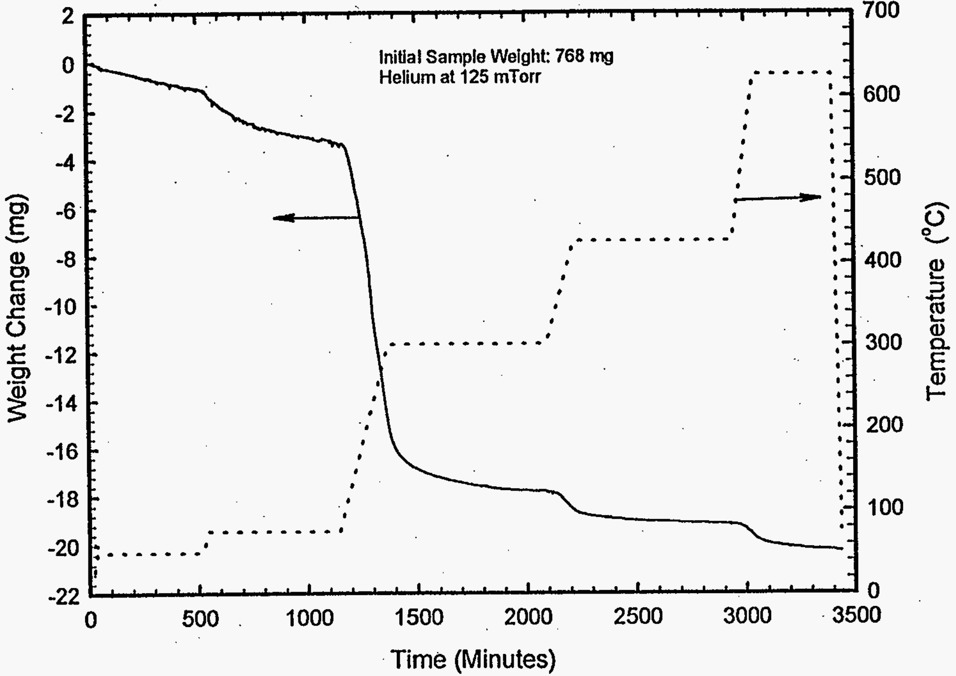

Figure 4. TGA Run 41 Plot Showing Weight Change, MS Signal for H_2O and Temperature versus Time. KE Canister Sludge 96-13.

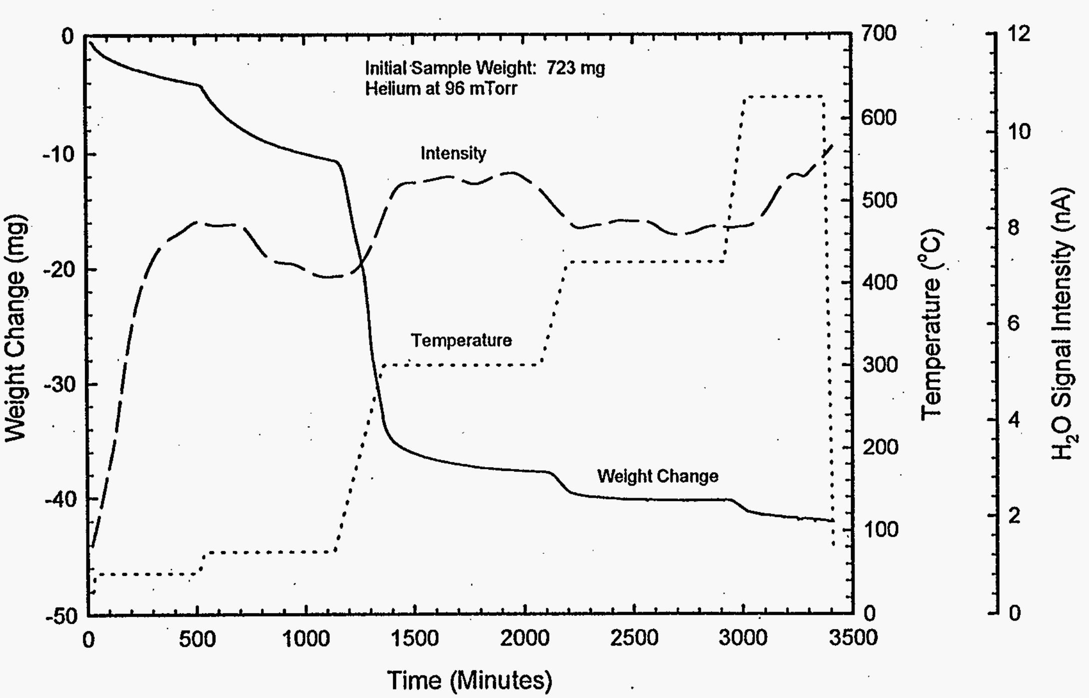

Figure 5. TGA Run 42 Plot Showing Weight Change, MS Signal for H_2O and Temperature versus Time. KE Canister Sludge 96-15.

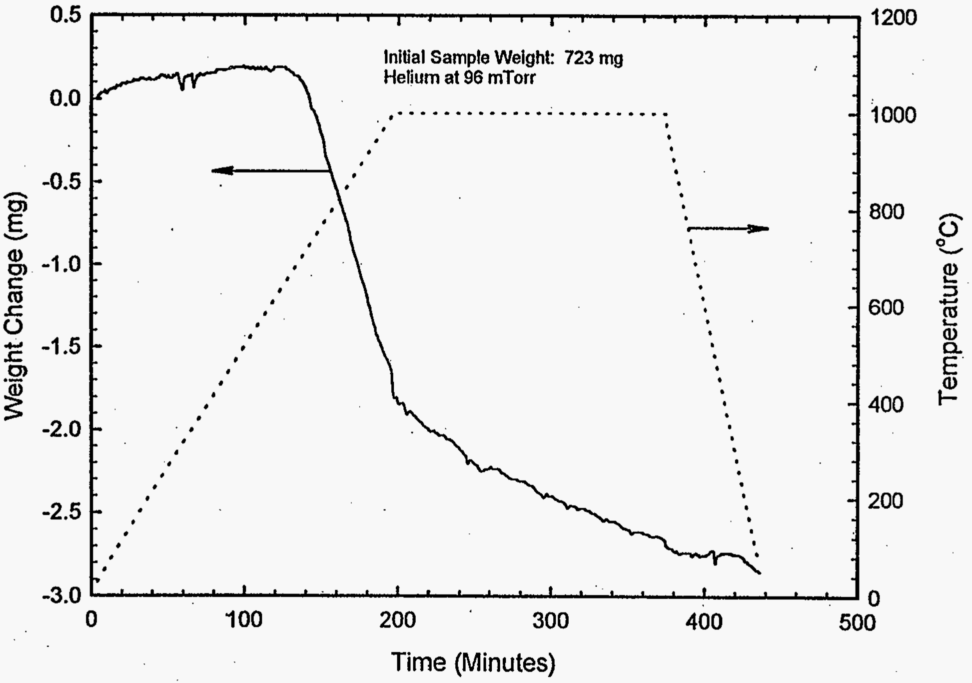

Figure 6. Plots of Weight Change and Temperature versus Time for Reheat of Sludge Sample 96-15 to 1000°C after TGA42.

Highlight Report: Third Report On Drying Behavior of K-East Canister Sludge

J Abrefah
HC Buchanan
SC Marschman
BJ Makenas (Duke Engineering & Services of Hanford)

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SUMMARY

Measurements have shown the water content (physically[1] plus chemically[2] bound) of the last group of four K-East canister sludge samples to range between 11 wt% and 68 wt%. The minimum water content corresponded to sample 96-15, and sample 96-08 showed the maximum water content. The hydrates constitute about 4 wt% to 15 wt% of the total water content of the sludge samples' initial weight. Most of the physically bound water was assumed to be released from the samples at ambient temperature for the runs in which the system was pumped down to vacuum conditions of about 40 mTorr. For the last run in which the test was conducted in helium at atmospheric pressure, a partial loss of the physically bound water was observed prior to the start of the test. Thus, the release of water resulting in sample weight loss above ambient temperature for the vacuum runs (11 to 13) was ascribed to thermal decomposition of hydrates. The weight loss observed for the first two segments of the last run was mostly due to release of physically bound water with possibly a small contribution from thermal decomposition of the hydrate(s). A summary of the measured weight losses within the corresponding test temperature segments are listed in the table below.

Drying Results of K-East Canister Sludge Sample Weight Loss (% of Initial Weight)

Run #	RM to 50°C	50°C to 75°C	75°C to 300°C	300°C to 425°C	425°C to 625°C
11	NE	0.20	3.36	0.23	0.18
12	0.22	0.22	3.04	0.38	0.29
13	0.63	2.30	10.4	1.50	0.69
14	7.74	1.44	4.08	0.62	0.31

NE = Not Evaluated; RM = Ambient Temperature

The largest fraction of the hydrated species thermally decomposed within the temperature range of 75°C to 300°C. At temperatures above 425°C the weight losses observed may be due to reduction of higher oxides of uranium by reactions such as:

$$3UO_3 = U_3O_8 + \frac{1}{2}O_2$$

rather than decomposition of hydrates. Quantitative analysis of the mass spectrometer (MS) signal for water was not done to determine corresponding weight losses because of a high background water signal.

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[1] Physically bound water is the "free water" which is physically absorbed and adsorbed by the pores and surface of the sludge.

[2] Chemically bound water is the hydrate(s) and hydroxides formed by the reaction of water with the sludge and will henceforth be referred to as "hydrates".

INTRODUCTION

Storage of Spent Nuclear Fuel (SNF) in the K-Basins has generated corrosion products (i.e., oxides of uranium), some of which may accompany the SNF elements when they are loaded into the Multi-Canister Overpacks (MCOs). The oxides of uranium will provide a large surface area for adsorption of water. Additionally, the oxides can chemically react with water to generate hydrates. These two forms of water (i.e., physically bound and the hydrates) will be subjected to drying steps during proposed Integrated Process Strategy (IPS) treatment of the SNF for the interim dry storage. Data on the drying behavior of these forms of moisture are therefore, needed in support of the IPS.

This highlight report is the third series of three covering the drying results of four additional samples of K-East canister sludge using a thermogravimetric/mass spectrometer (TGA/MS) system. These TGA runs are complementary to those performed previously for smaller samples on an instrument without a mass spectrometer capability (Makenas 1996) and the other results reported by Abrefah et al. (1997a, 1997b). The system monitors the weight change of the samples due to volatilization of species such as water. The volatilized species were monitored by the quadrupole mass spectrometer. The tests were conducted to determine the relative amount of water in the samples and the fractional release of the moisture at certain desired temperatures.

EXPERIMENTAL

The testing system that was used in the drying studies is shown in the highlight report by Abrefah et al. (1997) and a detailed description of the system is in the report of Marschman and Abrefah (1996).

The K-East canister sludge subsamples used for the drying tests came from a batch of nine K-East canister sludge samples shipped to the 325 Building for characterization. For each TGA run, about one gram of a sludge was loaded into an alumina crucible, weighed and mounted on the thermogravimetric stick. The alumina crucible was covered with a perforated lid to prevent loss of the sample after drying. The samples for TGA runs 43 to 45 were dried in a vacuum atmosphere with a ultra pure helium background. TGA run 46 was, however, performed in helium at about one atmosphere pressure. The drying was conducted within a temperature range of ambient to 625°C with typical temperature profiles such as the following:

(a) Heated at constant rate of 1°C/min. to a temperature of about 50°C and held at this temperature for 8 hours.

(b) Heated at constant rate of 1°C/min. to a temperature of about 75°C and held at this temperature for 10 hours.

(c) Heated at constant rate of 0.2°C/min. to a temperature of about 300°C and held at this temperature for 12 hours.

(d) Heated at constant rate of 1°C/min. to a temperature of about 425°C and held at this temperature for 12 hours.

(e) Heated at constant rate of 2°C/min. to a temperature of about 625°C and held at this temperature for 6 hours.

The system was evacuated to the lowest vacuum conditions (about 40 mTorr) after which the test vacuum pressure was established by flowing ultra high purity helium gas through the sample environment and throttling a valve to reduce the pumping speed of the sample environment. After runs 45 and 46, the samples were reheated to about 1000°C at a rate of 5°C/min to ascertain if there is any additional release of volatile species (e.g. water and oxygen) from the samples.

The sample weight change was continuously monitored by the electrobalance throughout the test and the off-gas stream was analyzed by the quadrupole mass spectrometer. The sample lost weight during the pump down to vacuum conditions was not recorded by the data acquisition system. That weight loss was calculated from the before and after test weight measurements.

One significant difference between these last four runs with the runs conducted previously (Abrefah 1997a, 1997b) was the slow heating rate from 75°C up to 300°C. During the first ten runs the ramp rate within this temperature range was 1°C/min compared to 0.2°C/min for the runs in this report.

RESULTS AND DISCUSSION

Four additional sludge samples have been tested, two from the canister sample 96-08, one from canister sample 96-04/Lower[3] and one from canister sample 96-15. A summary of the test samples and results are listed in Table 1. The NE (not evaluated) in the table indicates that a weight loss was not detectable within the noise level of the electrobalance. The MS data, in general, could not be analyzed for these four runs due to high background moisture which obscured the measurement of moisture from the sample. A large fraction of the physically adsorbed water on the sample vaporized during the pump down to vacuum condition. This happened before the sample was heated and the weight loss could not be monitored by the data acquisition system.

Before and after weight measurements in Table 1 indicate that samples for runs 44, and 46 (taken from canister sample 96-08) lost about 68 and 43 wt%, respectively, of their initial weights.

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[3] *The designation "lower" indicates that sample 96-04 was split into layers for analysis and the subsample for the TGA came from the lower layer.*

Samples for TGA runs 43 (taken from canister sample 96-15) and 45 (taken from canister sample 96-04/Lower), however, lost about 11 wt% and 24 wt%, respectively. The results of sludge samples taken from canister sample 96-08 including those reported in previous highlight reports (Abrefah 1997b) have indicated a high initial content of water which ranges between a low of about 29 wt% in run 39 (Abrefah 1997b) to as high as 77 wt%. In all cases a large fraction of the weight loss by each sample was due to removal of free water except the sample for run 45 (taken from canister sample 96-04/Lower), which indicated that more than 50% of the weight loss by the sample was due to release of chemically bonded volatile species (e.g. water and oxygen). The hydrated water for the samples ranges between 4 to 16 wt% (Table 1) with sample 96-04/Lower having the highest hydrate content. The 96-04/Lower is the sample that was analyzed by X-ray diffraction and found to contain a high content of schoepite ($UO_3 \cdot 2H_2O$) phase.

Comparison of the results of run 44 (sludge sample 96-08 in vacuum) and run 46 (sludge sample 96-08 in flowing helium at atmospheric pressure) suggest that most of the water released in run 46 within the temperature range of ambient to 50°C, and a large fraction of water released between 50°C and 75°C can be ascribed to free water. Thus, all the free water was not removed during drying in flowing gas at 50°C for about 10 hours. The inference from such an observation is that flowing gas drying for the KE canister sludge was not as efficient as the room temperature vacuum drying.

The characteristics of the thermal decomposition of the hydrate(s) and their release kinetics are shown in Figures 1 through 5. For each run, the weight change and the MS data for mass/charge ratio of 18, which is water (H_2O) are plotted. The MS data for all the plots were comparable to the background signal and analysis of the MS measurement for quantity of water released are therefore inaccurate. Hence, the MS data calculations for corresponding sample weight loss for the temperatures ranges listed in Table 1 will not be included in this highlight report. All the runs show considerably less, if any, interference from oxidation or absorption of water in the system as observed in the first 7 runs of the 14 run series. This is due to addition of a chemical filter to the system background gas supply (helium) that reduced water and oxygen impurities in the gas and system noise level improvement.

In Figure 1 (TGA run 43), the sample showed a very small weight loss at temperatures below 50°C which was difficult to evaluate giving the system noise level during the measurement. The weight loss in the temperature range 50°C to 75°C was again small but was measurable and could be evaluated. Most significant weight loss occurred within the temperature range of 75°C to 300°C. The majority of the weight loss for this temperature range occurred during the ramp from 75°C up to the 300°C (a total time of about 19 hours). The sample lost about 5 mg out of the total weight loss of 37.2 mg for that temperature range at the isothermal period of 10 hours at 300°C,. The weight losses within the temperature ranges 300°C to 425°C and 425°C to 625°C were also small but measurable. The MS signal in the figure shows a high background signal level which makes it very difficult to differentiate it from the water signal resulting from the sample drying.

In Figure 2 (TGA run 44), the weight change curve shows a small weight loss between ambient temperature and 50°C followed by yet another small weight losses within the temperature range of 50°C to 75°C. The last portion of the weight change curve for each of these temperature segments (ambient to 50°C, and 50°C to 75°C) still showed a decreasing behavior (i.e., a non-zero slope) at the end of the prescribed hold period. This incomplete release of moisture (i.e., the sample weight loss) at these two temperature segments may be due to slow decomposition of the higher temperature hydrate(s) at these low temperatures and vacuum condition of the test. The weight loss measured within the next temperature range (75°C to 300°C) is the largest and there is an indication of a slope decrease for the weight change curve. The decreasing slope indicates an increasing water release rate by the sample. Thus, assuming a constant source, the changes in slope can be used to determine the optimum temperature for thermal decomposition of this particular hydrated phase in the sludge sample. The minimum slope (Figure 3) occurred at a temperature of about 209°C. The sample for this run showed small weight losses at temperatures ranges 300°C to 425°C and 425°C to 625°C. The total weight loss measured at temperature above ambient was 4.2 wt% of the initial sample weight. Overall this sample lost a total of about 66 wt% of its initial weight, which calculates to be about 61.8 wt% of physically absorbed water (water removed at ambient temperature during system pump down) and 4.2 wt% chemically bound (water released during sample heat up to various temperature segments) water. The MS signal curve (curve smoothed using Lorentz method to remove the signal noise) indicates water was the volatile product released within the temperature ranges considered, but the noise and background signal levels were high enough to invalidate any quantitative estimates.

Figure 4 shows the drying results of TGA run 45. The sample lost significant weight between 75°C to 300°C, and small weight losses within the temperature ranges, ambient to 50°C, 50°C to 75°C, 300°C to 425°C and 425°C to 625°C. However, the weight loss for the temperature range 50°C to 75°C was higher than both the weight loss in the preceding temperature segment (ambient to 50°C for run 45) and the weight loss within the same temperature range (50°C to 75°C) for run 44 (Table 1). This increase in the weight loss suggests a probable increase in the hydrate content of this sample relative to the previous sample, and also an increase in the thermal decomposition of the hydrate at temperatures above 50°C. The weight change curve also indicates a continuous decrease in weight close to the end portion of the two lower temperature segments. This observation supports the assertion that this may be due to thermal decomposition of the same hydrated species. The weight loss between 75°C to 300°C shows a change in slope with a minimum slope at about 207°C, indicating an optimum decomposition temperature for the hydrate is around 207°C. The total weight loss of this run was about 24 wt% and the fractional weight loss at temperatures above ambient was 15.5 wt%. Assuming the latter to be due to thermal decomposition of hydrated species, the physically absorbed and adsorbed water correspond to approximately 8.5 wt%.

Figures 5 (TGA run 46) shows a weight loss curve for drying a K-East canister sludge in helium environment at one atmosphere pressure. A significant weight loss was measured at temperatures below 50°C because the preparation of this sample prior to the start of the test did not remove all the free water. The weight loss for the following range, 50°C to 75°C, was small compared to what was measured below 50°C but compared to the measured weight loss for similar sample (96-08 in run 44) the weight loss showed an increase of about a factor of 5. A significant weight loss was measured between 75°C and 300°C. The temperature corresponding to the point where the weight change curve for this range changes slope occurred at about 218°C. The sample showed slight weight decreases within the temperature ranges of 300°C to 425°C and 425°C to 625°C. The MS signal for this run suffered from a high background signal which prevented quantitative analysis of the water signal corresponding to the weight losses. Qualitatively, the MS signal indicates release of water from the sample during measured weight losses.

The weight change data for reheating the sludge sample used in run 45, up to 1000°C is plotted in Figure 6. The figure indicates a decrease in the sample weight starting at about 675°C and continued to the end of the test. A change of slope for the decrease in weight occurred when the sample temperature reached about 1000°C. The total weight loss was 6.26 mg (about 1.33 wt% of initial sample weight). The decrease in sample weight upon reheating indicates that the release of all volatile species (including thermal decomposition of water) was not complete during run 45. The weight loss observed during the reheating can be due to release of a higher temperature hydrate and/or hydroxide. A decomposition of UO_3 to U_3O_8 at higher temperatures can also result in the weight loss observed during the reheating run.

Calculated weight loss for various temperature ranges are listed in Table 1. If every weight loss by the sample is ascribed to thermal decomposition of the hydrated species in the sludge samples then the bound water content in the sludge ranges between 4.0 to 15.5 wt%. X-ray diffraction (XRD) analyses of the canister sludge have identified the trioxide hydrate, $UO_3 \cdot 2H_2O$, phase and the characteristics of the thermal decomposition data obtained might support only one form of hydrate. Assuming that the canister sludge contains $UO_3 \cdot 2H_2O$ phase, the theoretical weight fraction of the water content in the hydrate portion of the sludge sample would be 11.2 wt%. The maximum weight fraction observed in run 45 is greater than this theoretical estimate and there are two possible reasons for the additional weight loss. The first is a contribution by hydrates of the uranium peroxide (i.e., $UO_4 \cdot xH_2O$; $2<x<4$) and the second is the weight loss associated with reduction of the higher oxides of uranium (e.g. $3UO_3 = U_3O_8 + \frac{1}{2}O_2$). Since the $UO_4 \cdot xH_2O$ phase has not been identified by the XRD analysis, the latter reason may be the main factor in the increased weight loss above the expected theoretical estimate for run 45. Excluding the free water component in run 46, the largest fraction of the hydrate(s) thermally decomposed within the temperature range of 75°C to 300°C for all the tests. The slow temperature ramp rate (0.2°C/min) for this temperature range for the four runs caused most of the hydrated species present to decompose during the approximately 19 hours to reach 300°C. The temperature response of the first ten runs reported in Abrefah et al. (1997a and 1997b) for this range with faster ramp rate (1°C/min) is a faster release of the moisture from the sample.

In run 46, a fraction of the physically bound water was available after the first temperature drying segment of ambient to 50°C. The evidence of this was in comparing the weight loss during the second temperature segment for runs 44 and 46. The weight loss in run 46 which started with a large fraction of free water left in the sample due to changes in the experimental steps and conditions (one hour pump down for run 46 versus 24 hours pump down for run 44), is about 5 times greater than the weight loss in run 44. Thus, a flowing gas environment at 50°C for 10 hours was not adequate to dry out all the free water in the sludge. This observation supports the inference that in all the runs, a small fraction of the physically bound water can remain in the sample after ambient temperature pump down to be released during the initial temperature segments of the tests. The left over physically bound water may contribute to the increased weight loss above the theoretical expectation for the sludge sample in run 45.

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Makenas BJ and RB Baker. 1996. *Accelerated Examinations of K East Basin Canister Sludge*. DESH-9655840, Duke Engineering & Services of Hanford, Richland, Washington.

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Table 1. TG Drying Results of KE Canister Sludge

Run #		Status (Test Start Date)	System Press. (mTorr)	Sample				Sample Weight Loss (mg) for Different Temperature Ranges (°C)				
TGA	KE Sludge			ID	Weight Before Test (mg)	Weight After Test (mg)	Weight Loss (mg)	RM to 50	50 to 75	75 to 300	300 to 425	425 to 625
43	11	04/08/97	132*	96-15	1108	984	124	NE	2.169	37.23	2.530	1.987
44	12	04/15/97	90*	96-08	1004	322	682	2.169	2.169	30.54	3.795	2.892
45	13	04/18/97	140*	96-04/L	471	358	113	2.982	10.84	48.80	7.048	3.253
46	14	04/22/97	~760×10^3**	96-08	771	440	331	59.71	11.11	31.49	4.800	2.390

RM = Ambient temperature
NE= Not evaluated

* These are the starting pressures of the runs and during the test the system pressure increases slightly due to increasing temperature and a balance between pumping speed and gas flow.

** The system was at ambient pressure (i.e., about one atmosphere) and the actual pressure was not accurately measured.

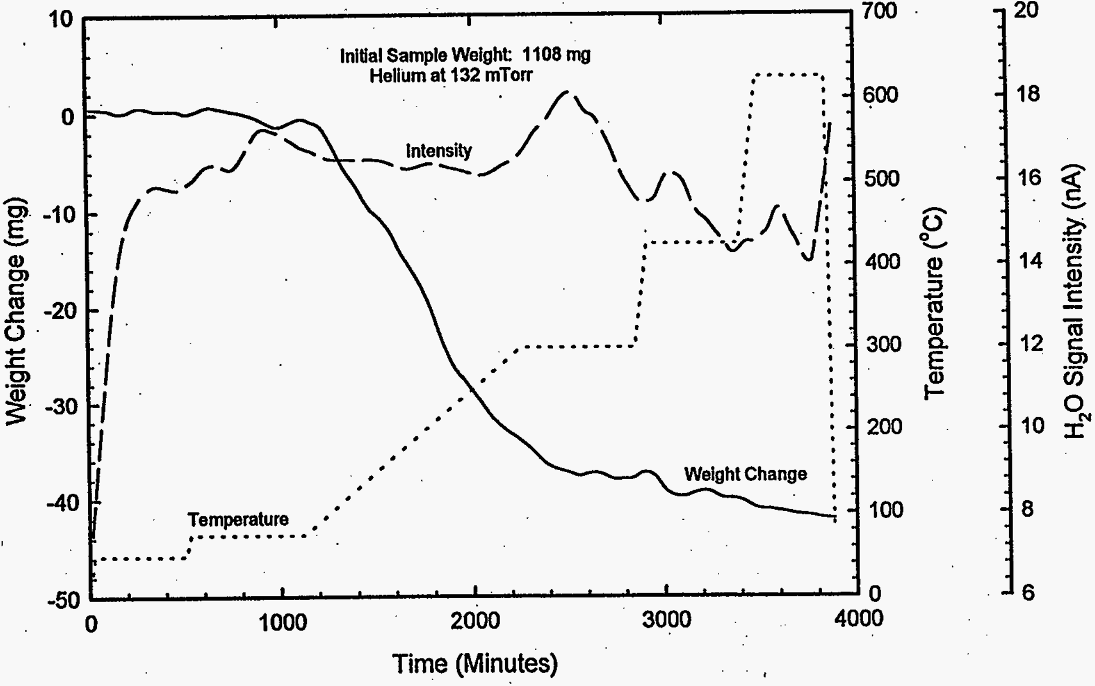

Figure 1. TGA Run 43 Plot Showing Weight Change, MS Signal for H_2O and Temperature versus Time for KE Canister Sludge 96-15.

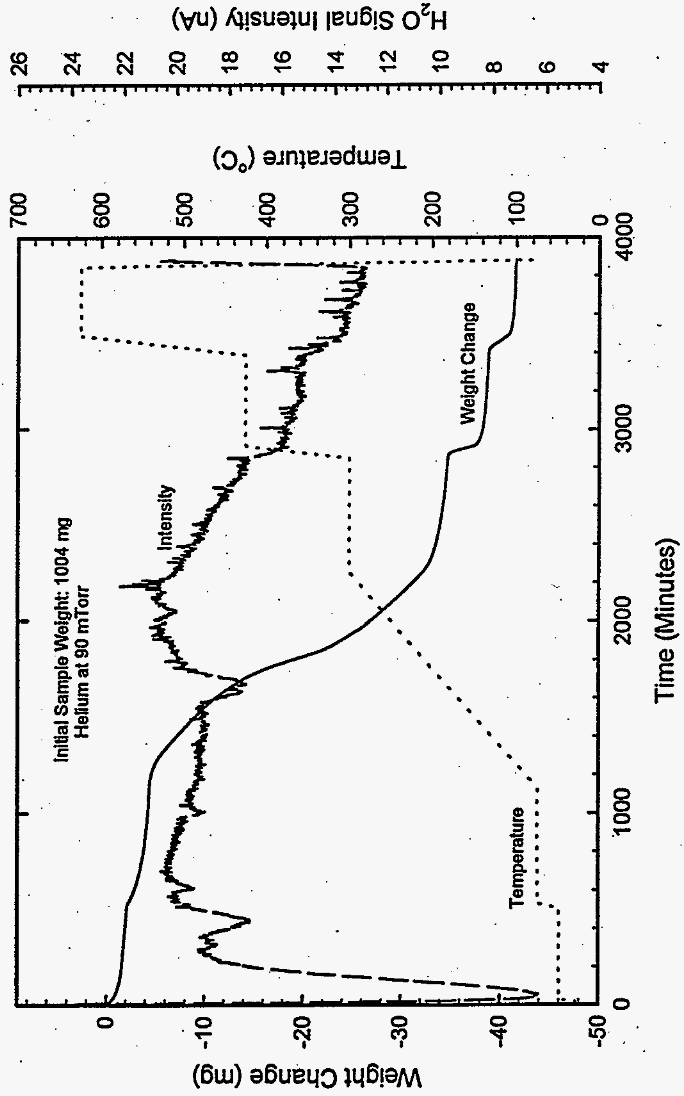

Figure 2. TGA Run 44 Plot Showing Weight Change, MS Signal for H_2O and Temperature versus Time for KE Canister Sludge 96-08.

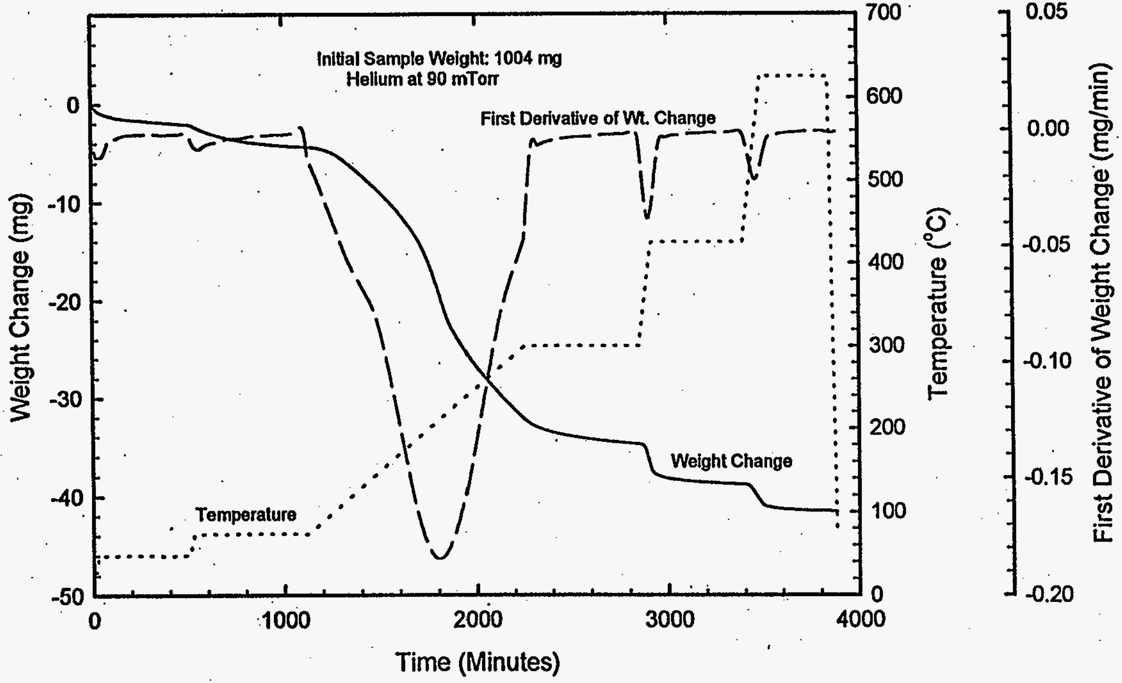

Figure 3. Plots of Weight Change, First Decrivative of the Weight Change and Temperature versus Time for TGA 44.

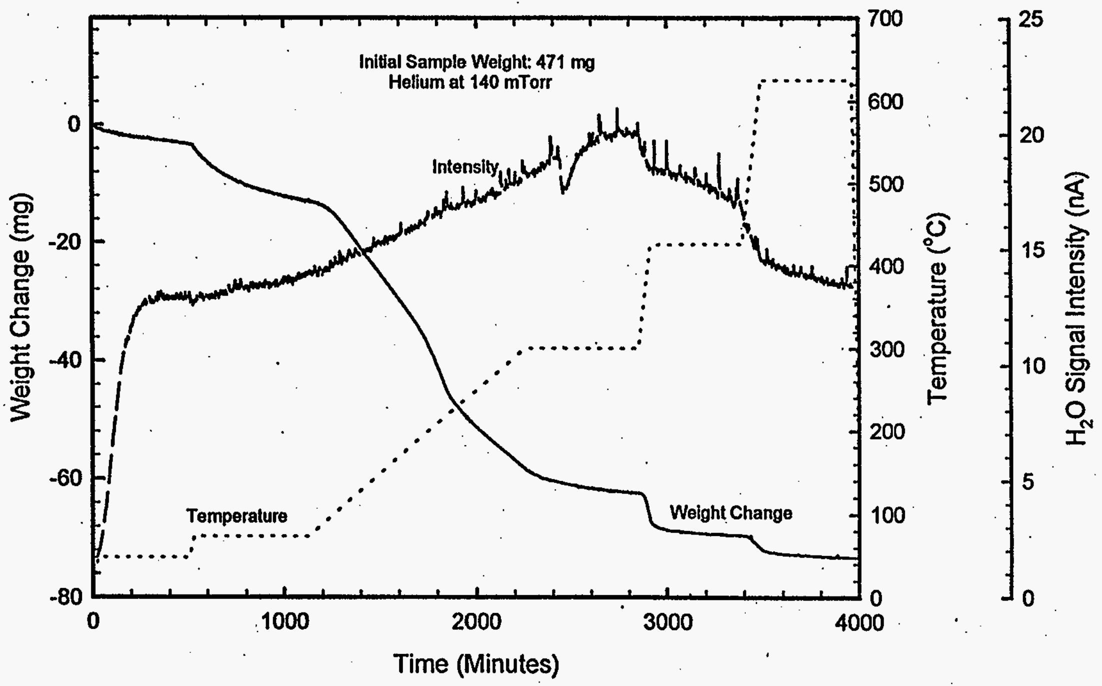

Figure 4. TGA Run 45 Plot Showing Weight Change, MS Signal for H_2O and Temperature versus Time for KE Canister Sludge 96-04/Lower.

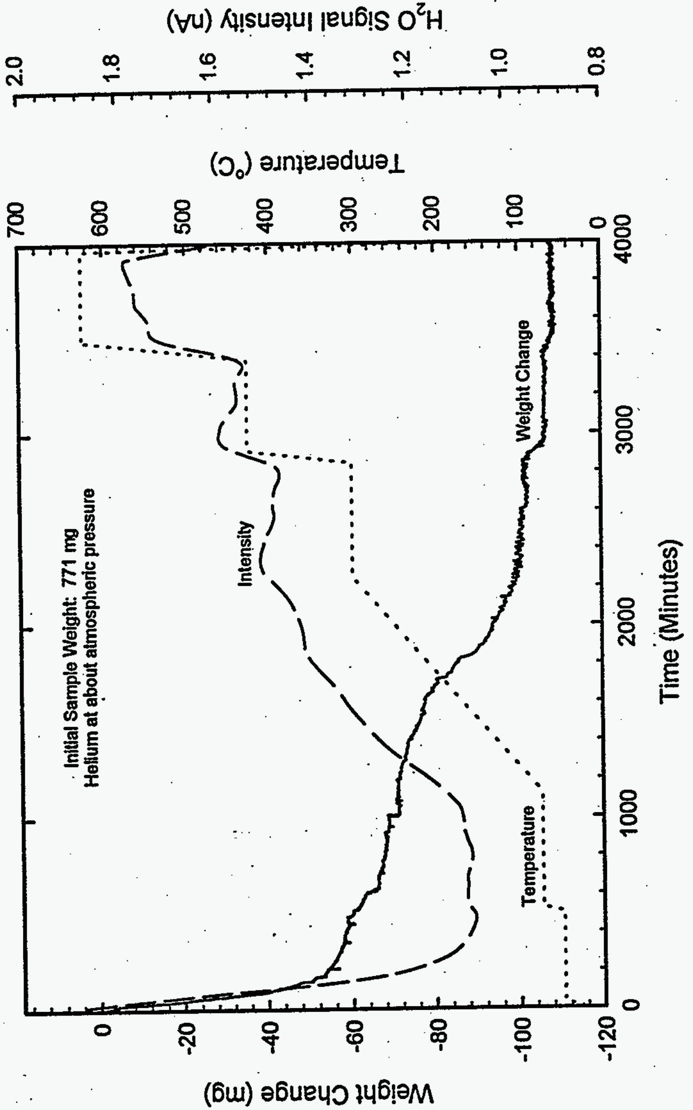

Figure 5. TGA Run 46 Plot Showing Weight Change, MS Signal for H_2O and Temperature versus Time for KE Canister Sludge 96-08.

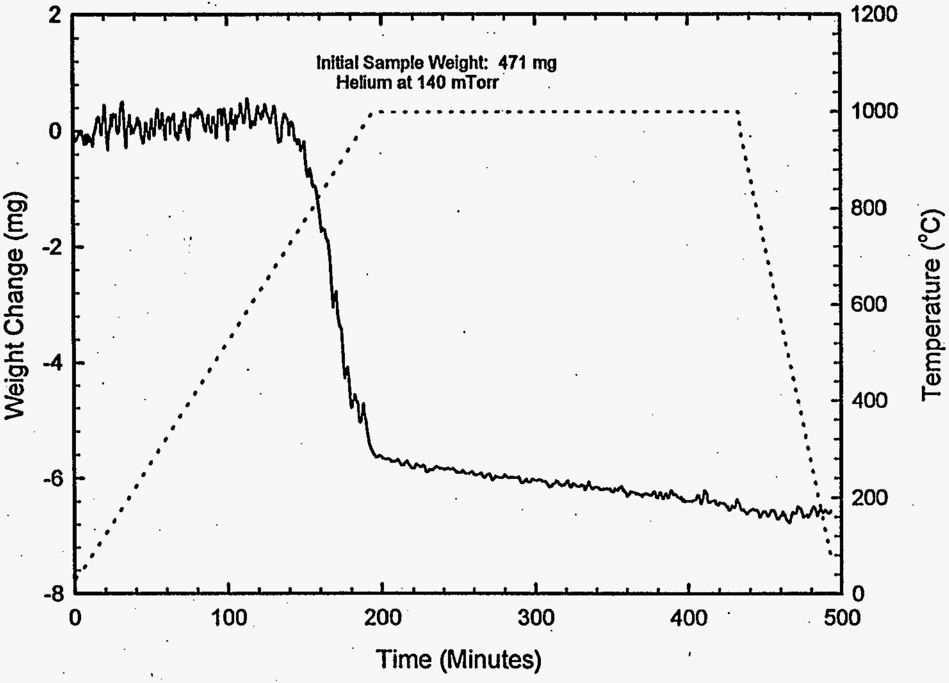

Figure 6. A Plot of Weight Change and Temperature versus Time for Reheating of Sludge sample 96-04/Lower used in TGA run 45.

APPENDIX N

COMPARISON OF EUROPIUM ISOTOPE AND PLUTONIUM CONCENTRATIONS

T. L. Welsh

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APPENDIX N

COMPARISON OF EUROPIUM ISOTOPE AND PLUTONIUM CONCENTRATIONS

Several correlations between the sludge characterization sample results are of interest; in particular, the correlations between (1) $^{239/240}$Pu and ^{154}Eu and (2) $^{239/240}$Pu and ^{155}Eu. If a relationship between fission product radioisotopes (measured by gamma energy analyses translatable to field applications) and plutonium concentration (measured by an analytical laboratory method) can be identified, then the field gamma measurements may be used to predict the plutonium concentration of the sludge leaving the basin.

Linear regression techniques were used to determine the correlations. Linear regression techniques provide equations of the form

$$Y = a + bX \qquad (1)$$

where Y is the dependent variable, X is the independent variable, b is the slope, and a is the y-intercept.

$^{239/240}$Pu and ^{154}Eu

The regression analysis used the ^{154}Eu data for the independent variable (X) and the $^{239/240}$Pu data as the dependent variable (Y). Regression analyses were performed for the four sets of sludge characterization data; µCi/g centrifuged sludge, µCi/g as-settled sludge, µCi/ml as-settled sludge, and µCi/g dried sludge.

Centrifuged Sludge Data (per gram basis)

Equation 2 was calculated from the regression analysis of the µCi/g centrifuged sludge data.

$$^{239/240}\mathrm{Pu} = -1.38324 + 8.438665 \times {}^{154}\mathrm{Eu} \qquad (2)$$

The R^2 value for Equation 2 is 93.4%. The R^2 value indicates the proportion of the variability in the dependent variable ($^{239/240}$Pu) that is explained by the independent variable (^{154}Eu). The data, the regression equation, the confidence interval for a mean Y value associated with a given X value, and the prediction interval for an individual Y value associated with a given X value are illustrated in Figure N-1. The regression equation (predicted values) is represented by the middle solid line. The 95% confidence interval (CI) is represented by the first set of lines, lower limit (LL) and upper limit (UL), which enclose the regression line. The 95% CI is used to make statements such as "We conclude, with 95% confidence, that the mean $^{239/240}$Pu concentration (μCi/g centrifuged sludge) is between 2.845 and 11.604 when the ^{154}Eu concentration (μCi/g centrifuged sludge)is 1.02." The 95% prediction interval (PI) is represented by the outside lines, LL and UL, which enclose the regression line. The prediction interval illustrates the variability associated with predicting (using the regression equation) a new individual value for Y given an X value. The data, the predicted value for the observed X values, the standard deviation used in calculating the 95% confidence interval, and the standard deviation used in calculating the 95% prediction interval are provided in Table N1.

As-Settled Sludge Data (per gram basis)

Equation 3 was calculated from the regression analysis of the μCi/g as-settled sludge data.

$$^{239/240}\mathrm{Pu} = -1.47829 + 8.715132 \times {}^{154}\mathrm{Eu} \qquad \textbf{(3)}$$

The R^2 value for Equation 3 is 94.6%. The data, the regression equation, the confidence interval for a mean Y value associated with a given X value, and the prediction interval for an individual Y value associated with a given X value are illustrated in Figure N-2. The data, the predicted value for the observed X values, the standard deviation used in calculating the 95% confidence interval, and the standard deviation used in calculating the 95% prediction interval are provided in Table N2.

As-Settled Sludge Data (per ml basis)

Equation 4 was calculated from the regression analysis of the μCi/ml as-settled sludge data.

$$^{239/240}\mathrm{Pu} = -2.13498 + 8.831050 \times {}^{154}\mathrm{Eu} \qquad \textbf{(4)}$$

The R^2 value for Equation 4 is 96.1%. The data, the regression equation, the confidence interval for a mean Y value associated with a given X value, and the prediction interval for an individual Y value associated with a given X value are illustrated in Figure N-3. The data, the predicted value for the observed X values, the standard deviation used in calculating the 95% confidence interval, and the standard deviation used in calculating the 95% prediction interval are provided in Table N3.

Dried Sludge Data (per gram basis)

Equation 5 was calculated from the regression analysis of the µCi/g dried sludge data.

$$^{239/240}\text{Pu} = -0.21809 + 7.607438 \times {}^{154}\text{Eu} \tag{5}$$

The R^2 value for Equation 5 is 87.0%. The data, the regression equation, the confidence interval for a mean Y value associated with a given X value, and the prediction interval for an individual Y value associated with a given X value are illustrated in Figure N-4. The data, the predicted value for the observed X values, the standard deviation used in calculating the 95% confidence interval, and the standard deviation used in calculating the 95% prediction interval are provided in Table N4.

The Sandfilter Backwash Pit (SFBWP) data (Warner 1994) were combined with the KE canister sludge characterization data (µCi/ml as-settled sludge). Equation 6 was calculated from the regression analysis of the combined µCi/ml as-settled sludge data.

$$^{239/240}\text{Pu} = -1.71466 + 8.807991 \times {}^{154}\text{Eu} \tag{6}$$

The R^2 value for Equation 6 is 96.4%. The data, the regression equation, the confidence interval for a mean Y value associated with a given X value, and the prediction interval for an individual Y value associated with a given X value are illustrated in Figure N-5. The data, the predicted value for the observed X values, the standard deviation used in calculating the 95% confidence interval, and the standard deviation used in calculating the 95% prediction interval are provided in Table N5.

The R^2 values, calculated for each of the five ^{154}Eu and $^{239/240}$Pu regression analyses (µCi/g centrifuged sludge, µCi/g as-settled sludge, µCi/ml as-settled sludge, µCi/g dried sludge, and the combined µCi/ml as-settled sludge), range in value from 87.0% (per g dried sludge basis) to 95.9% (per ml as-settled sludge basis). The variability associated with predicting a $^{239/240}$Pu concentration from knowing the ^{154}Eu concentration ranged from approximately 7.6 (per g as-settled sludge basis) to approximately 18.2 (per g dried sludge basis). Based on the R^2 values and the prediction variability the "best" prediction equation would be Equation 3 (per g as-settled sludge basis).

$^{239/240}$Pu and ^{155}Eu

The regression analysis used the ^{155}Eu data for the independent variable (X) and the $^{239/240}$Pu data as the dependent variable (Y). Regression analyses were performed for the four sets of sludge characterization data; µCi/g centrifuged sludge, µCi/g as-settled sludge, µCi/g dried sludge, and µCi/ml as-settled sludge.

Centrifuged Sludge Data (per gram basis)

Equation 7 was calculated from the regression analysis of the µCi/g centrifuged sludge data.

$$^{239/240}\mathrm{Pu} = 1.328622 + 12.052579 \times {}^{155}\mathrm{Eu} \tag{7}$$

The R^2 value for Equation 7 is 97.7%. The data, the regression equation, the confidence interval for a mean Y value associated with a given X value, and the prediction interval for an individual Y value associated with a given X value are illustrated in Figure N-6. The prediction interval illustrates the variability associated with predicting (using the regression equation) a new individual value for Y given an X value. The data, the predicted value for the observed X values, the standard deviation used in calculating the 95% confidence interval, and the standard deviation used in calculating the 95% prediction interval are provided in Table N6.

As-Settled Sludge Data (per gram basis)

Equation 8 was calculated from the regression analysis of the µCi/g as-settled sludge data.

$$^{239/240}\mathrm{Pu} = 0.773777 + 12.20963 \times {}^{155}\mathrm{Eu} \tag{8}$$

The R^2 value for Equation 8 is 98.7%. The data, the regression equation, the confidence interval for a mean Y value associated with a given X value, and the prediction interval for an individual Y value associated with a given X value are illustrated in Figure N-7. The data, the predicted value for the observed X values, the standard deviation used in calculating the 95% confidence interval, and the standard deviation used in calculating the 95% prediction interval are provided in Table N7.

As-Settled Sludge Data (per ml basis)

Equation 9 was calculated from the regression analysis of the μCi/ml as-settled sludge data.

$$^{239/240}\mathrm{Pu} = 0.708004 + 12.420384 \times {}^{155}\mathrm{Eu} \qquad (9)$$

The R^2 value for Equation 8 is 99.6%. The data, the regression equation, the confidence interval for a mean Y value associated with a given X value, and the prediction interval for an individual Y value associated with a given X value are illustrated in Figure N-8. The data, the predicted value for the observed X values, the standard deviation used in calculating the 95% confidence interval, and the standard deviation used in calculating the 95% prediction interval are provided in Table N8.

Dried Sludge Data (per gram basis)

Equation 10 was calculated from the regression analysis of the μCi/g dried sludge data.

$$^{239/240}\mathrm{Pu} = 6.400789 + 11.50259 \times {}^{155}\mathrm{Eu} \qquad (10)$$

The R^2 value for Equation 10 is 86.3%. The data, the regression equation, the confidence interval for a mean Y value associated with a given X value, and the prediction interval for an individual Y value associated with a given X value are illustrated in Figure N-9. The data, the predicted value for the observed X values, the standard deviation used in calculating the 95% confidence interval, and the standard deviation used in calculating the 95% prediction interval are provided in Table N9.

The R^2 values, calculated for each of the four ^{155}Eu and $^{239/240}$Pu regression analyses (μCi/g centrifuged sludge, μCi/g as-settled sludge, μCi/ml as-settled sludge, and μC/g dried sludge), range in value from 86.3% (per g dried sludge basis) to 99.5% (per ml as-settled sludge basis). The variability associated with predicting a $^{239/240}$Pu concentration from knowing the ^{155}Eu concentration ranged from approximately 3.1 (per g as-settled sludge basis) to approximately 15.0 (per g dried sludge basis). Based on the R^2 values and the prediction variability the "best" prediction equation would be Equation 8 (per g as-settled sludge basis). However, a word of caution is needed. The large R^2 values are influenced by the spacing of the X (^{155}Eu) observations. The ^{155}Eu data can be considered as two clusters; the first subset of data is clustered at the low concentration values of ^{155}Eu while the second subset (a single data point) is at the high concentration value of ^{155}Eu. Figures N-6, N-7, and N-8 illustrate the clustering of the data.

Table N1. $^{239/240}$Pu Versus ^{154}Eu--μCi/g Centrifuged Sludge.

Location	^{154}Eu μCi/g	$^{239/240}$Pu μCi/g	$^{239/240}$Pu μCi/g Predicted	Variability associated with a mean response $s(\hat{Y}_h)$ - μCi/g	Variability associated with the prediction of a new Y response $s(\hat{Y}_{h(new)})$ - μCi/g
1	3.54	8.98	28.5	1.80	9.84
2	0.934	5.90	6.50	2.16	9.91
3	1.09	7.73	7.82	2.12	9.90
4	2.34	14.8	18.4	1.89	9.85
5	1.02	6.61	7.22	2.13	9.90
6	0.706	5.75	4.57	2.21	9.92
9	4.37	22.0	35.5	1.82	9.84
10	0.321	2.24	1.33	2.31	9.94
11	1.14	7.90	8.24	2.11	9.90
12	0.256	1.83	0.777	2.33	9.95
16	1.29	11.4	9.50	2.07	9.89
17	1.11	8.19	7.98	2.11	9.90
18	0.241	1.78	0.650	2.33	9.95
19	0.636	5.17	3.98	2.23	9.92
20	1.85	14.7	14.2	1.96	9.87
21	2.31	16.2	18.1	1.89	9.85
23	0.705	4.85	4.57	2.21	9.92
2	16.2	133	135	5.71	11.2
3	3.92	34.7	31.7	1.80	9.84
4	0.802	6.52	5.39	2.19	9.91
5	7.62	92.1	62.9	2.48	9.98
6	8.22	83.5	68.0	2.66	10.0
7	4.36	34.4	35.4	1.82	9.84
8	10.7	63.0	88.9	3.53	10.3
9	11.7	94.8	97.3	3.91	10.4
10	8.49	82.2	70.3	2.75	10.1
11	9.50	81.0	78.8	3.09	10.2
12	0.628	4.52	3.92	2.23	9.92
13	0.875	5.99	6.00	2.17	9.91

Table N2. $^{239/240}$Pu Versus. ^{154}Eu--µCi/g As-Settled Sludge.

Location	^{154}Eu µCi/g	$^{239/240}$Pu µCi/g	$^{239/240}$Pu µCi/g Predicted	Variability associated with a mean response $s(\hat{Y}_h)$ - µCi/g	Variability associated with the prediction of a new Y response $s(\hat{Y}_{h(new)})$ - µCi/g
1	2.16	5.47	17.3	1.37	7.50
2	0.634	4.01	4.05	1.59	7.54
3	0.650	4.61	4.19	1.58	7.54
4	1.62	10.2	12.6	1.42	7.51
5	0.577	3.74	3.55	1.60	7.54
6	0.486	3.96	2.76	1.62	7.55
9	3.90	19.6	32.5	1.42	7.51
10	0.250	1.74	0.700	1.67	7.56
11	0.756	5.26	5.11	1.56	7.53
12	0.123	0.883	-0.406	1.70	7.57
16	1.10	9.66	8.11	1.50	7.52
17	0.892	6.58	6.30	1.54	7.53
18	0.197	1.45	0.239	1.69	7.56
19	0.545	4.42	3.27	1.61	7.54
20	0.964	7.65	6.92	1.52	7.53
21	1.76	12.3	13.9	1.40	7.50
22	1.23	7.54	9.24	1.48	7.52
23	0.641	4.41	4.11	1.59	7.54
2	14.8	121	127	4.92	8.86
3	2.60	23.0	21.2	1.35	7.49
4	0.513	4.17	2.99	1.61	7.55
5	6.87	82.9	58.4	2.10	7.66
6	7.34	74.5	62.5	2.25	7.71
7	2.54	20.0	20.7	1.35	7.49
8	6.88	40.5	58.8	2.10	7.67
9	6.48	52.3	55.0	1.99	7.63
10	7.05	68.2	60.0	2.16	7.68
11	8.66	73.8	74.0	2.68	7.84
12	0.334	2.39	1.43	1.65	7.55
13	0.607	4.15	3.81	1.59	7.54

Table N3. $^{239/240}$Pu Versus ^{154}Eu--μCi/ml As-Settled Sludge.

Location	^{154}Eu μCi/ml	$^{239/240}$Pu μCi/ml	$^{239/240}$Pu μCi/ml Predicted	Variability associated with a mean response $s(\hat{Y}_h)$ - μCi/ml	Variability associated with the prediction of a new Y response $s(\hat{Y}_{h(new)})$ - μCi/ml
1	2.48	6.29	19.8	2.88	15.2
2	0.761	4.81	4.59	3.12	15.2
3	0.676	4.79	3.84	3.13	15.2
4	1.86	11.8	14.3	2.95	15.2
5	0.641	4.16	3.53	3.14	15.2
6	0.607	4.95	3.23	3.14	15.2
9	8.90	44.8	76.5	2.97	15.2
10	0.337	2.35	0.841	3.19	15.2
11	0.862	5.99	5.48	3.10	15.2
12	0.173	1.24	-0.607	3.22	15.2
16	1.69	14.9	12.8	2.98	15.2
17	1.35	9.94	9.79	3.03	15.2
18	0.295	2.17	0.470	3.20	15.2
19	0.867	7.04	5.52	3.10	15.2
20	1.07	8.49	7.31	3.07	15.2
21	3.07	21.4	25.0	2.82	15.1
22	1.98	12.1	15.4	2.94	15.2
23	1.05	7.23	7.14	3.07	15.2
2	34.5	284	303	10.1	18.0
3	3.09	27.3	25.2	2.82	15.1
4	0.551	4.47	2.73	3.15	15.2
5	16.9	204	147	4.73	15.6
6	13.5	137	117	3.86	15.4
7	2.76	21.7	22.2	2.85	15.2
8	10.0	59.1	86.2	3.14	15.2
9	9.17	74.1	78.8	3.01	15.2
10	13.5	131	117	3.86	15.4
11	25.9	221	227	7.41	16.6
12	0.356	2.55	1.009	3.19	15.2
13	0.752	5.14	4.51	3.12	15.2

Table N4. $^{239/240}$Pu Versus ^{154}Eu--µCi/g Dried Sludge.

Location	^{154}Eu µCi/g	$^{239/240}$Pu µCi/g	$^{239/240}$Pu µCi/g Predicted	Variability associated with a mean response $s(\hat{Y}_h)$ - µCi/ml	Variability associated with the prediction of a new Y response $s(\hat{Y}_{h(new)})$ - µCi/ml
1	11.8	30.0	89.6	4.37	18.2
2	2.39	15.1	18.0	4.25	18.2
3	8.57	60.7	65.0	3.48	18.0
4	8.18	51.8	62.0	3.42	18.0
5	4.82	31.3	36.5	3.56	18.0
6	1.81	14.7	13.6	4.46	18.2
9	6.68	33.6	50.6	3.35	18.0
10	0.588	4.10	4.26	4.95	18.4
11	5.23	36.4	39.6	3.48	18.0
12	0.640	4.58	4.65	4.93	18.4
16	2.16	19.1	16.2	4.33	18.2
17	1.69	12.5	12.6	4.51	18.3
18	0.443	3.26	3.15	5.01	18.4
19	1.02	8.30	7.54	4.77	18.3
20	8.49	67.3	64.4	3.46	18.0
21	4.85	33.8	36.7	3.55	18.0
22	1.16	7.98	8.61	4.72	18.3
2	18.6	153	141	7.51	19.2
3	10.6	93.5	80.4	3.96	18.1
4	4.18	34.0	31.6	3.70	18.1
5	9.12	110	69.2	3.57	18.1
6	9.40	95.4	71.3	3.63	18.1
7	24.1	190	183	10.4	20.5
8	17.0	99.7	129	6.70	18.9
9	10.7	104	81.2	3.99	18.1
10	10.9	92.9	82.7	4.05	18.2
11	6.37	45.6	48.2	3.36	18.0
12	2.52	17.2	19.0	4.20	18.2

Table N5. Combined $^{239/240}$Pu Versus ^{154}Eu--µCi/ml As-Settled Sludge.

Location	^{154}Eu µCi/ml	$^{239/240}$Pu µCi/ml	$^{239/240}$Pu µCi/ml Predicted	Variability associated with a mean response $s(\hat{Y}_h)$ - µCi/ml	Variability associated with the prediction of a new Y response $s(\hat{Y}_{h(new)})$ - µCi/ml
1	2.48	6.29	20.1	2.19	13.3
2	0.761	4.81	4.99	2.35	13.4
3	0.676	4.79	4.24	2.36	13.4
4	1.86	11.8	14.7	2.24	13.3
5	0.641	4.16	3.93	2.37	13.4
6	0.607	4.95	3.63	2.37	13.4
9	8.90	44.8	76.7	2.51	13.4
10	0.337	2.35	1.25	2.40	13.4
11	0.862	5.99	5.88	2.34	13.3
12	0.173	1.24	-0.191	2.43	13.4
16	1.69	14.9	13.2	2.25	13.3
17	1.35	9.94	10.2	2.29	13.3
18	0.295	2.17	0.884	2.41	13.4
19	0.867	7.04	5.92	2.34	13.3
20	1.07	8.49	7.71	2.32	13.3
21	3.07	21.4	25.3	2.16	13.3
22	1.98	12.1	15.7	2.23	13.3
23	1.05	7.23	7.53	2.32	13.3
SFBWP	0.163	1.02	-0.279	2.43	13.4
SFBWP	0.164	0.928	-0.270	2.43	13.4
SFBWP	0.179	1.05	-0.138	2.43	13.4
SFBWP	0.281	1.68	0.760	2.41	13.4
SFBWP	0.161	1.31	-0.297	2.43	13.4
SFBWP	0.322	2.24	1.12	2.41	13.4
SFBWP	0.354	1.89	1.40	2.40	13.4
SFBWP	0.241	1.52	0.408	2.42	13.4
2	34.5	284	302	8.85	15.8
3	3.09	27.3	25.5	2.16	13.3
4	0.551	4.47	3.14	2.38	13.4
5	16.9	204	147	4.18	13.8
6	13.5	137	117	3.38	13.6
7	2.76	21.7	22.6	2.17	13.3
8	10.0	59.1	86.4	2.69	13.4
9	9.17	74.1	79.1	2.55	13.4
10	13.5	131	117	3.38	13.6
11	25.9	221	226	6.51	14.7
12	0.356	2.55	1.42	2.40	13.4
13	0.752	5.14	4.91	2.35	13.4

Table N6. $^{239/240}$Pu Versus ^{155}Eu--µCi/g Centrifuged Sludge.

Location	^{155}Eu µCi/g	$^{239/240}$Pu µCi/g	$^{239/240}$Pu µCi/g Predicted	Variability associated with a mean response $s(\hat{Y}^{h})$ - µCi/g	Variability associated with the prediction of a new Y response $s(\hat{Y}_{h(new)})$ - µCi/g
1	1.73	8.98	22.2	1.03	4.77
2	0.422	5.90	6.42	1.10	4.78
3	0.534	7.73	7.77	1.08	4.78
4	1.04	14.8	13.9	1.03	4.77
5	0.447	6.61	6.72	1.09	4.78
6	0.34	5.75	5.43	1.11	4.79
9	2.01	22.0	25.55	1.05	4.77
10	0.163	2.24	3.29	1.14	4.80
11	0.563	7.90	8.11	1.08	4.78
12	0.170	1.83	3.38	1.14	4.80
16	0.159	2.46	3.25	1.14	4.80
17	0.730	11.4	10.1	1.06	4.78
18	0.432	8.19	6.54	1.10	4.78
19	0.138	1.78	2.99	1.15	4.80
20	0.630	14.7	8.92	1.07	4.78
21	0.901	16.2	12.2	1.04	4.77
1	10.6	133	129	4.05	6.17
2	1.95	34.7	24.8	1.04	4.77
3	0.305	6.52	5.01	1.12	4.79
4	5.70	63	70.0	2.10	5.11
5	0.267	4.52	4.55	1.12	4.79

Table N7. $^{239/240}$Pu Versus ^{155}Eu--µCi/g As-Settled Sludge.

Location	^{155}Eu µCi/g	$^{239/240}$Pu µCi/g	$^{239/240}$Pu µCi/g Predicted	Variability associated with a mean response $s(\hat{Y}_h)$ - µCi/g	Variability associated with the prediction of a new Y response $s(\hat{Y}_{h(new)})$ - µCi/g
1	1.05	5.47	13.6	0.660	3.10
2	0.287	4.01	4.28	0.705	3.11
3	0.318	4.61	4.66	0.702	3.10
4	0.716	10.2	9.52	0.669	3.10
5	0.253	3.74	3.86	0.709	3.11
6	0.234	3.96	3.63	0.711	3.11
9	1.80	19.6	22.8	0.699	3.10
10	0.127	1.74	2.32	0.724	3.11
11	0.375	5.26	5.35	0.696	3.10
12	0.082	0.883	1.78	0.730	3.11
15	0.154	2.39	2.65	0.721	3.11
16	0.622	9.66	8.37	0.675	3.10
17	0.347	6.58	5.01	0.698	3.10
18	0.112	1.45	2.14	0.726	3.11
20	0.327	7.65	4.77	0.701	3.10
21	0.684	12.3	9.13	0.671	3.10
1	9.68	121	119	2.80	4.12
2	1.29	23	16.5	0.664	3.10
3	0.196	4.17	3.17	0.716	3.11
4	3.67	40.5	45.6	1.06	3.20
5	0.143	2.39	2.52	0.722	3.11

Table N8. $^{239/240}$Pu Versus ^{155}Eu--µCi/ml As-Settled Sludge.

Location	^{155}Eu µCi/ml	$^{239/240}$Pu µCi/ml	$^{239/240}$Pu µCi/ml Predicted	Variability associated with a mean response $s(\hat{Y}_h)$ - µCi/ml	Variability associated with the prediction of a new Y response $s(\hat{Y}_{h(new)})$ - µCi/ml
1	1.21	6.29	15.7	0.901	4.23
2	0.344	4.81	4.98	0.911	4.23
3	0.331	4.79	4.82	0.911	4.23
4	0.824	11.8	10.9	0.902	4.23
5	0.281	4.16	4.20	0.913	4.23
6	0.293	4.95	4.35	0.912	4.23
9	4.10	44.8	51.6	1.061	4.26
10	0.171	2.35	2.83	0.916	4.23
11	0.427	5.99	6.01	0.909	4.23
12	0.115	1.24	2.14	0.918	4.23
15	0.200	3.10	3.19	0.915	4.23
16	0.957	14.9	12.6	0.901	4.23
17	0.524	9.94	7.22	0.907	4.23
18	0.168	2.17	2.80	0.916	4.23
20	0.364	8.49	5.23	0.911	4.23
21	1.20	21.4	15.6	0.901	4.23
1	22.6	284	281	4.078	5.80
2	1.54	27.3	19.8	0.905	4.23
3	0.211	4.47	3.33	0.915	4.23
4	5.35	59.1	67.2	1.199	4.30
5	0.152	2.55	2.60	0.917	4.23

Table N9. $^{239/240}$Pu Versus ^{155}Eu--µCi/g Dried Sludge.

Location	^{155}Eu µCi/ml	$^{239/240}$Pu µCi/ml	$^{239/240}$Pu µCi/ml Predicted	Variability associated with a mean response $s(\hat{Y}_h)$ - µCi/ml	Variability associated with the prediction of a new Y response $s(\hat{Y}_{h(new)})$ - µCi/ml
1	5.76	30.0	72.7	4.31	15.0
2	1.08	15.1	18.8	3.70	14.8
3	4.19	60.7	54.6	3.39	14.7
4	3.63	51.8	48.2	3.21	14.7
5	2.12	31.3	30.8	3.25	14.7
6	0.872	14.7	16.4	3.82	14.8
9	3.08	33.6	41.8	3.13	14.7
10	0.299	4.10	9.84	4.20	15.0
11	2.59	36.4	36.2	3.16	14.7
12	0.425	4.58	11.3	4.11	14.9
15	0.275	4.25	9.56	4.22	15.0
16	1.23	19.1	20.5	3.62	14.8
17	0.658	12.5	14.0	3.96	14.9
18	0.253	3.26	9.31	4.23	15.0
20	2.88	67.3	39.5	3.13	14.7
21	1.89	33.8	28.1	3.33	14.7
1	12.2	153	147	10.23	17.6
2	5.26	93.5	66.9	3.96	14.9
3	1.60	34	24.8	3.44	14.8
4	9.04	99.7	110	7.13	16.0
5	2.73	45.6	37.8	3.14	14.7

Figure N-1

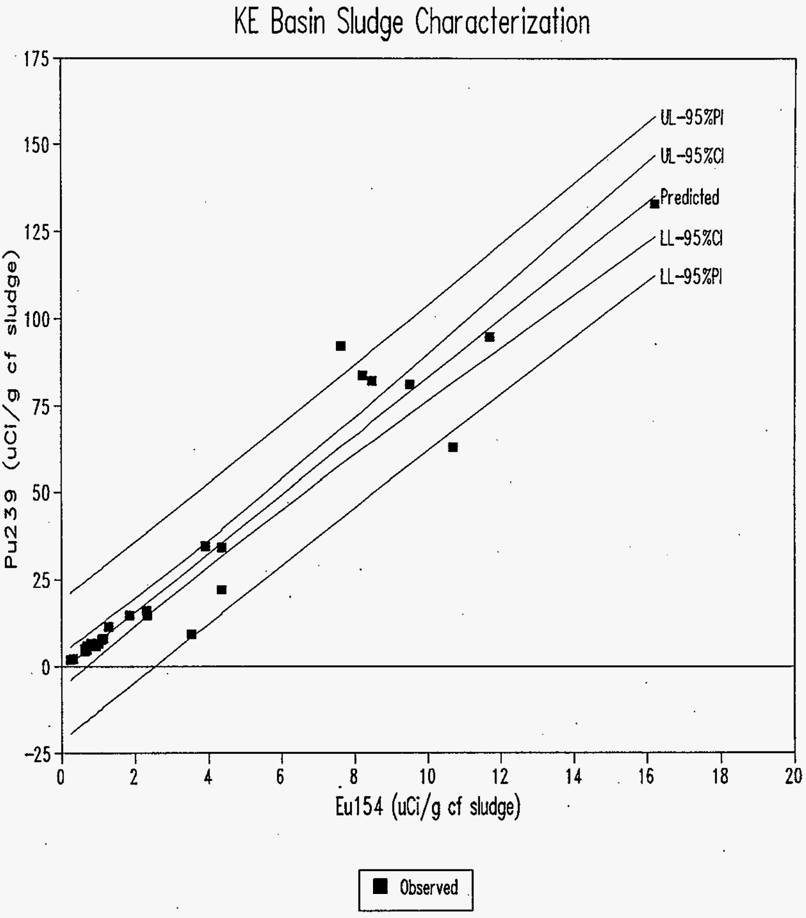

KE Basin Sludge Characterization
Pu239 (uCi/g of sludge)
175
150
125
100
75
50
25
0
-25
Eu154 (uCi/g of sludge)
0
2
4
6
8
10
12
14
16
18
20
UL-95%PI
UL-95%CI
Predicted
LL-95%CI
LL-95%PI
■ Observed

Figure N-2

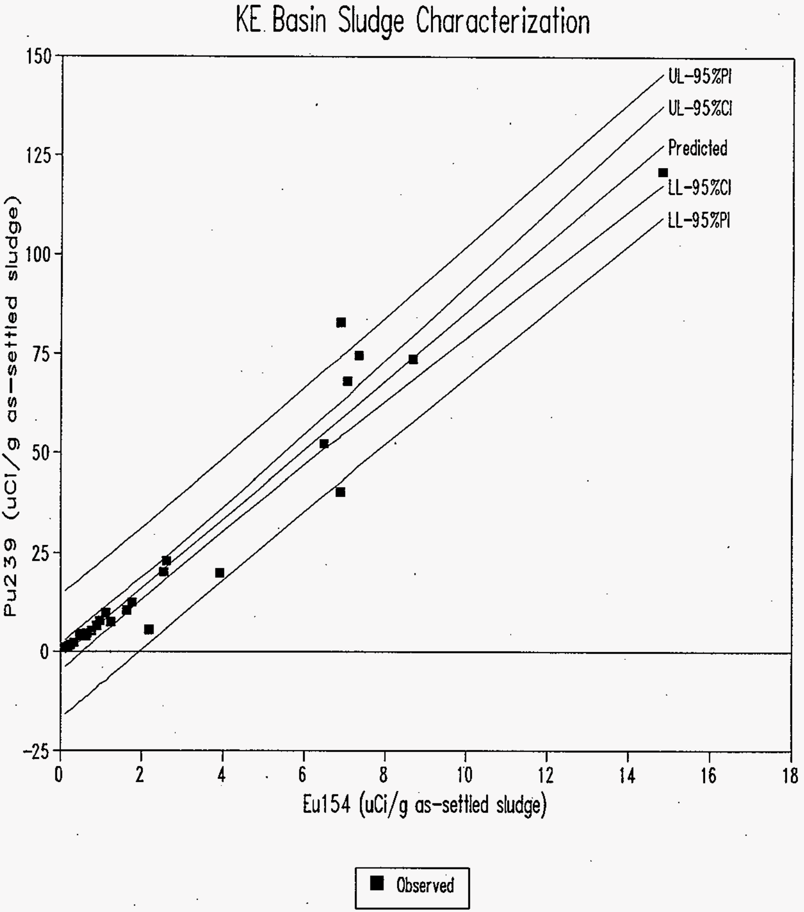

KE Basin Sludge Characterization
Pu239 (uCi/g as-settled sludge)
150
125
100
75
50
25
0
-25
0
2
4
6
8
10
12
14
16
18
Eu154 (uCi/g as-settled sludge)
UL-95%PI
UL-95%CI
Predicted
LL-95%CI
LL-95%PI
■ Observed

Figure N-3

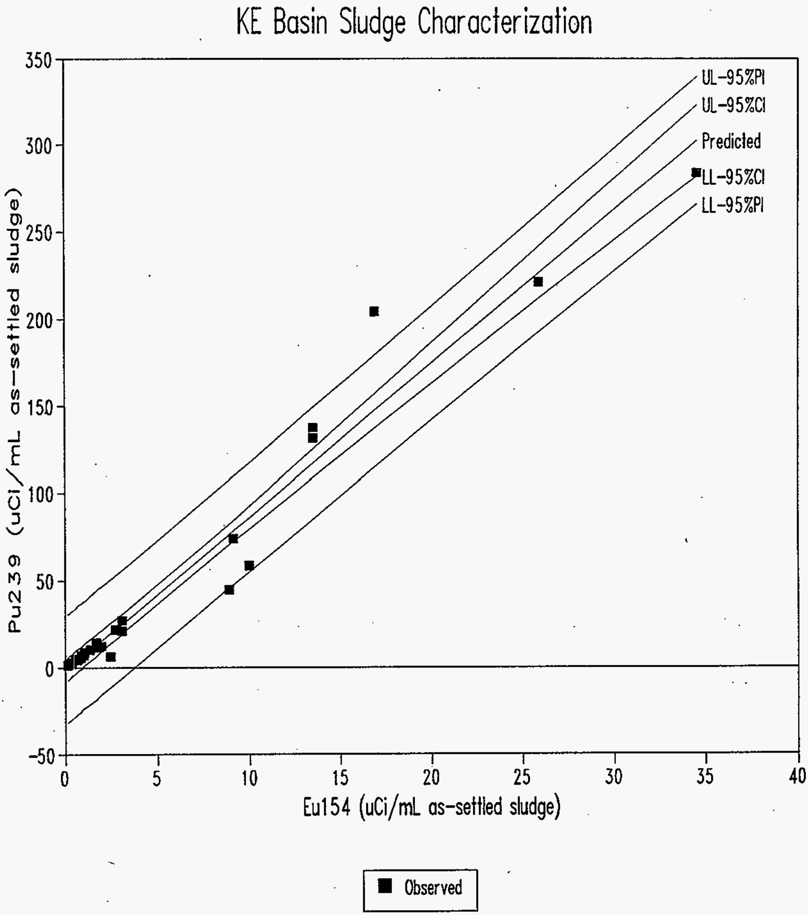

KE Basin Sludge Characterization
Pu239 (uCi/mL as-settled sludge)
350
300
250
200
150
100
50
0
-50
Eu154 (uCi/mL as-settled sludge)
0
5
10
15
20
25
30
35
40
UL-95%PI
UL-95%CI
Predicted
LL-95%CI
LL-95%PI
■ Observed

Figure N-4

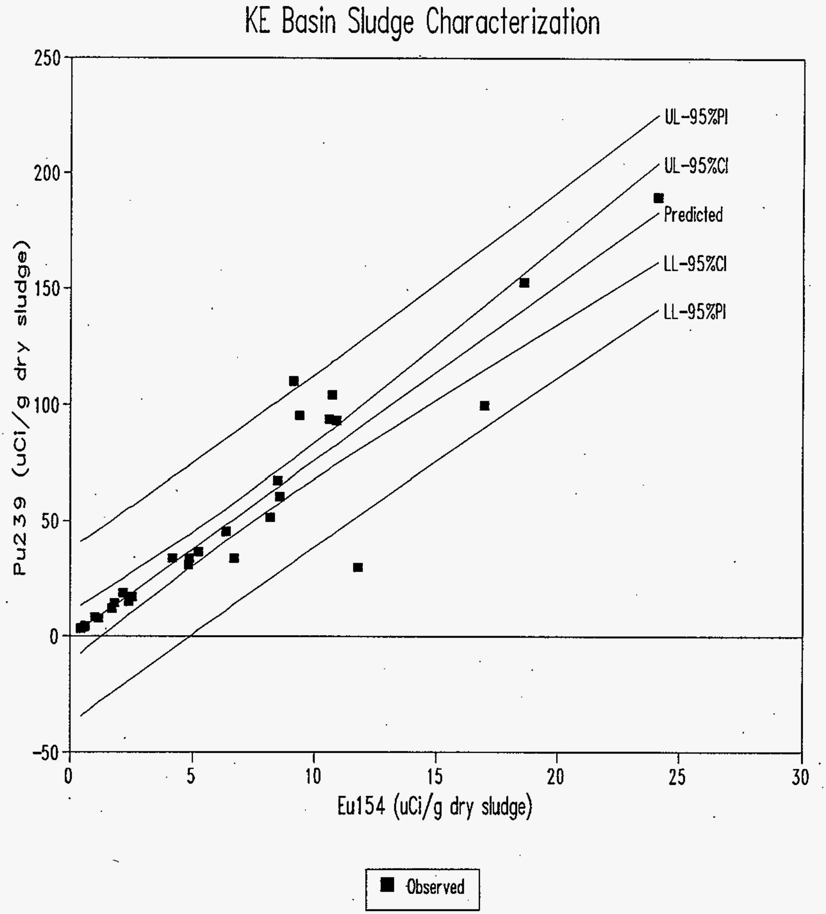

KE Basin Sludge Characterization
Pu239 (uCi/g dry sludge)
250
200
150
100
50
0
-50
Eu154 (uCi/g dry sludge)
0
5
10
15
20
25
30
UL-95%PI
UL-95%CI
Predicted
LL-95%CI
LL-95%PI
■ Observed

Figure N-5

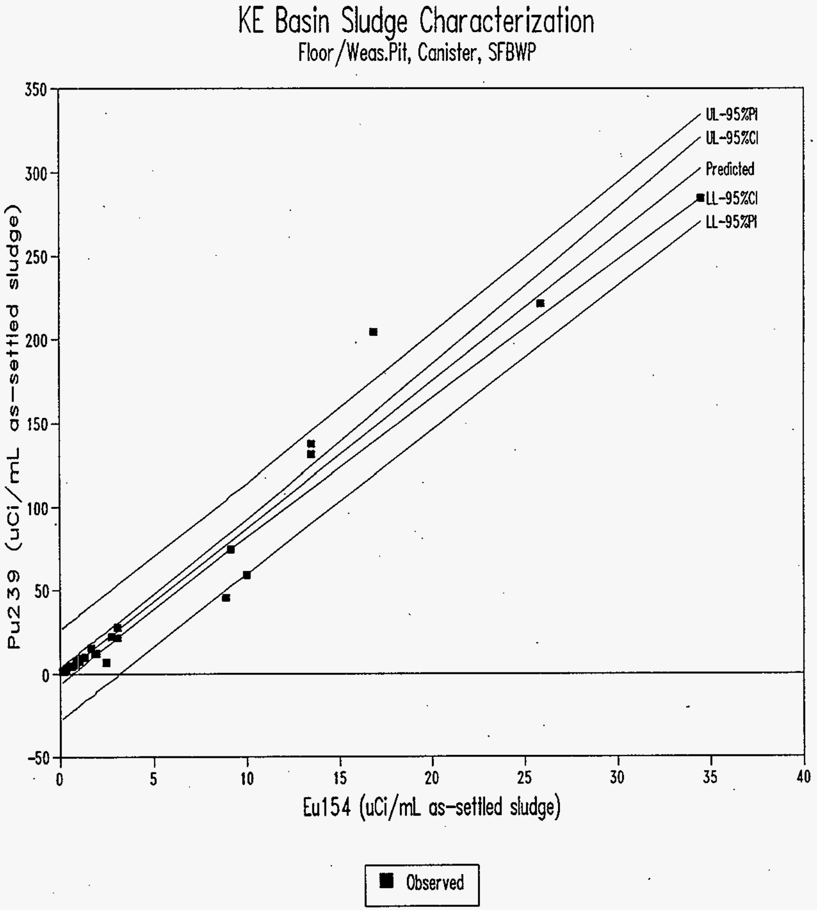

KE Basin Sludge Characterization
Floor/Weas.Pit, Canister, SFBWP
Pu239 (uCi/mL as-settled sludge)
350
300
250
200
150
100
50
0
-50
Eu154 (uCi/mL as-settled sludge)
0
5
10
15
20
25
30
35
40
UL-95%PI
UL-95%CI
Predicted
LL-95%CI
LL-95%PI
■ Observed

Figure N-6

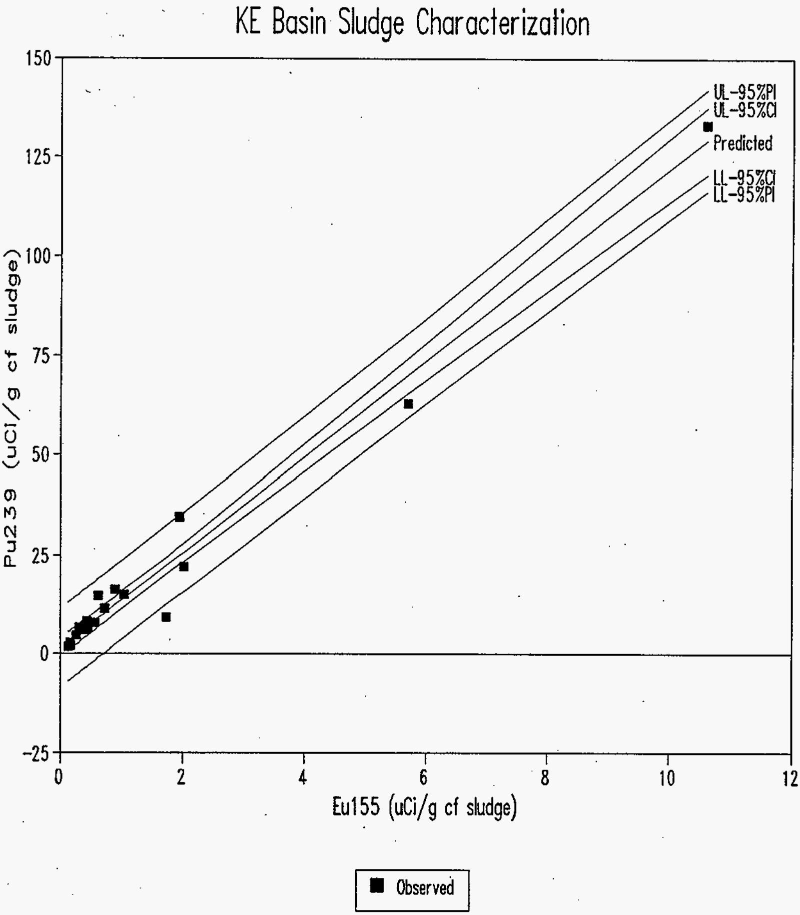

KE Basin Sludge Characterization
Pu239 (uCi/g cf sludge)
150
125
100
75
50
25
0
-25
Eu155 (uCi/g cf sludge)
0
2
4
6
8
10
12
UL-95%PI
UL-95%CI
Predicted
LL-95%CI
LL-95%PI
■ Observed

Figure N-7

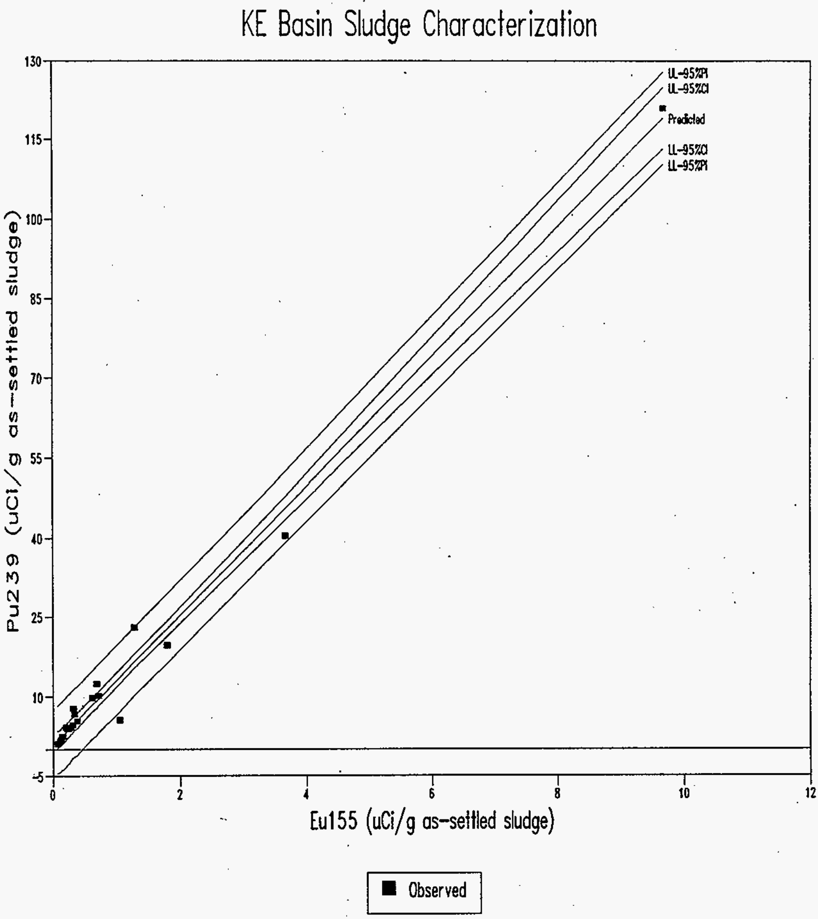

KE Basin Sludge Characterization
Pu239 (uCi/g as-settled sludge)
130
115
100
85
70
55
40
25
10
-5
Eu155 (uCi/g as-settled sludge)
0
2
4
6
8
10
12
UL-95%PI
UL-95%CI
Predicted
LL-95%CI
LL-95%PI
■ Observed

Figure N-8

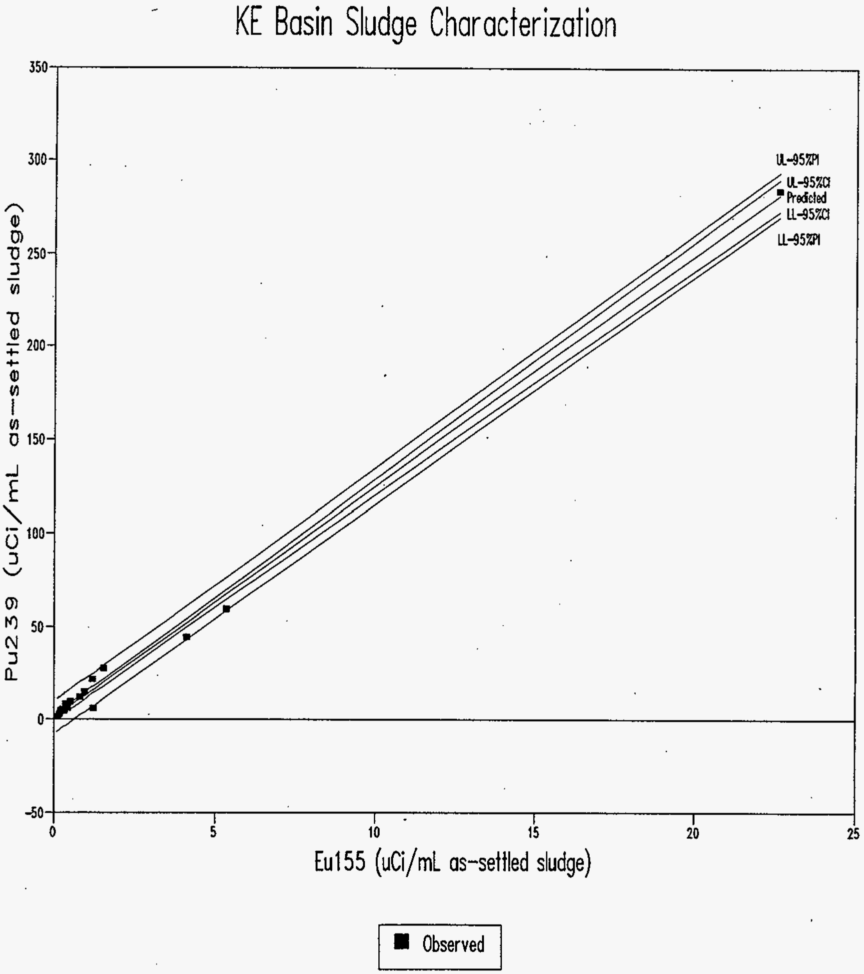
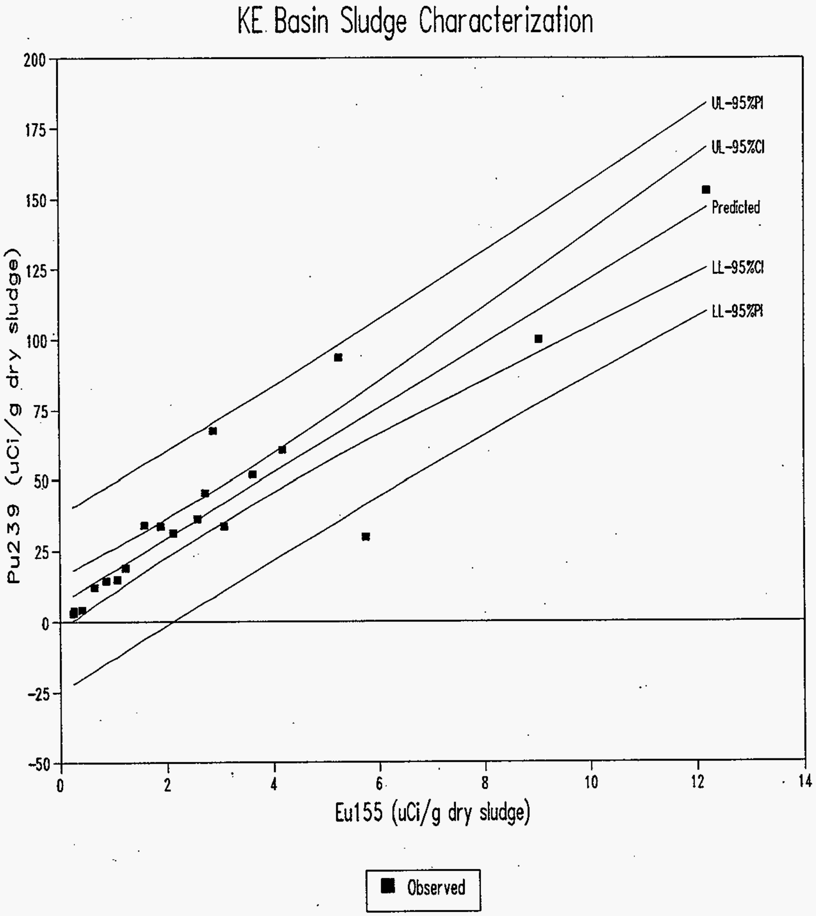

KE Basin Sludge Characterization
Pu239 (uCi/mL as-settled sludge)
350
300
250
200
150
100
50
0
-50
0
5
10
15
20
25
Eu155 (uCi/mL as-settled sludge)
UL-95%PI
UL-95%CI
Predicted
LL-95%CI
LL-95%PI
■ Observed

Figure N-9


KE Basin Sludge Characterization
200
175
150
125
100
75
50
25
0
-25
-50
Pu239 (uCi/g dry sludge)
0
2
4
6
8
10
12
14
Eu155 (uCi/g dry sludge)
UL-95%PI
UL-95%CI
Predicted
LL-95%CI
LL-95%PI
■ Observed

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APPENDIX O

K EAST CANISTER SLUDGE SUMMARY STATISTICS, EDITED DATA SET, WITHOUT SAMPLE 96-01

T. L. Welsh

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APPENDIX O

K EAST CANISTER SLUDGE SUMMARY STATISTICS, EDITED DATA SET, WITHOUT SAMPLE 96-01

From the data tables presented in Appendix C it can be observed that the analytical results for sample 96-01 are

- Over one order of magnitude lower than the remainder of the data (e.g., Alpha Total, ^{241}Am.aea, $^{239/240}$Pu) or
- Over two orders of magnitude higher than the remainder of the data (e.g., Total Carbon, TOC, TIC, Na.icp).

The summary statistics were recomputed without this sample's result for all analytes except (1) those that were measured by PNNL, (2) those that were measured by 222-S using the supernate samples (pH, IC), and (3) %water.grav by 222-S. These data are provided for information only. Outlier tests were performed and only the tests for Total Carbon, TIC, TOC, Cr.icp. and Na.icp indicated that Sample 96-01 analytical results were outliers.

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Table 0.1. K East Canister Sludge Characterization Data--
per Gram Centrifuged Sludge. (Page 1 of 3)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
Ag.icp.w	µg/g	2.32E+00	1.71E+02	12	7.01E+01	6.32E+01	90.2	2.43E+02
Ag.icp.wo	µg/g	2.32E+00	1.71E+02	10	8.29E+01	6.15E+01	74.2	2.62E+02
Al.icp	µg/g	9.08E+03	5.61E+04	12	2.00E+04	1.33E+04	66.9	5.65E+04
Alpha Total	µCi/g	7.72E+00	1.96E+02	12	9.61E+01	6.38E+01	66.3	2.71E+02
^{241}Am.aea.w	µCi/g	4.96E+00	8.06E+01	12	4.15E+01	2.79E+01	67.3	1.18E+02
^{241}Am.aea.wo	µCi/g	4.96E+00	8.06E+01	11	4.36E+01	2.82E+01	64.7	1.23E+02
^{241}Am.gea	µCi/g	4.70E+00	3.37E+02	12	7.79E+01	8.88E+01	114.1	3.21E+02
B.icp	µg/g	8.30E+01	1.58E+02	12	1.21E+02	2.30E+01	19.0	1.84E+02
Ba.icp.w	µg/g	< 9.95E+00	1.54E+02	12	8.54E+01	4.17E+01	48.9	2.00E+02
Ba.icp.wo	µg/g	2.91E+01	1.54E+02	10	9.14E+01	3.78E+01	41.4	2.01E+02
Be.icp	µg/g	< 9.29E+00	2.61E+01	12	NA	NA	NA	NA
Beta Total	µCi/g	1.39E+01	4.05E+03	12	2.04E+03	1.61E+03	78.6	6.44E+03
Bi.icp.w	µg/g	< 1.99E+01	5.75E+02	12	2.57E+02	2.10E+02	81.8	8.33E+02
Bi.icp.wo	µg/g	3.95E+01	5.75E+02	10	2.99E+02	2.06E+02	68.9	8.98E+02
^{212}Bi.gea	µCi/g	< 1.39E-01	< 1.68E+01	12	NA	NA	NA	NA
Br^{-}.ic @	µg/g	< 2.04E-01	< 1.25E+00	10	NA	NA	NA	NA
Ca.icp.w	µg/g	1.12E+02	8.59E+02	12	4.43E+02	2.50E+02	56.3	1.13E+03
Ca.icp.wo	µg/g	1.12E+02	8.59E+02	9	5.25E+02	2.37E+02	45.1	1.24E+03
Cd.icp.w	µg/g	< 9.85E+00	6.38E+01	12	2.27E+01	1.44E+01	63.1	6.20E+01
Cd.icp.wo	µg/g	1.52E+01	6.38E+01	10	2.52E+01	1.45E+01	57.4	6.74E+01
^{144}Ce/Pr.gea	µCi/g	< 1.51E-01	< 2.82E+01	12	NA	NA	NA	NA
Cl^{-}.ic.w @	µg/g	1.29E-01	1.05E+00	10	4.52E-01	2.72E-01	60.1	1.24E+00
$^{243/244}$Cm	µCi/g	< 5.12E-01	< 1.81E+01	12	NA	NA	NA	NA
^{57}Co.gea	µCi/g	2.30E-02	< 1.21E+01	12	NA	NA	NA	NA
^{60}Co.gea.w	µCi/g	1.25E-01	< 1.45E+00	12	7.60E-01	4.19E-01	55.1	1.91E+00
^{60}Co.gea.wo	µCi/g	1.25E-01	8.52E-01	7	5.18E-01	2.70E-01	52.2	1.44E+00
Cr.icp.w	µg/g	< 1.86E+01	2.79E+02	12	6.74E+01	7.62E+01	113.2	2.76E+02
Cr.icp.wo	µg/g	3.93E+01	2.79E+02	6	1.15E+02	8.59E+01	74.9	4.33E+02
^{134}Cs.gea	µCi/g	< 3.37E-02	< 2.00E+00	12	NA	NA	NA	NA
^{137}Cs.gea	µCi/g	4.87E+00	9.94E+02	12	4.43E+02	3.04E+02	68.6	1.28E+03

*: per gram centrifuged sludge
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.w: less than values were included (i.e. 3 for <3) when calculating the summary statistics.
.wo: less than values were deleted when calculating the summary statistics.
NA: not applicable for these data.
@: Analyses performed using sludge supernate.
&: Calculated using the PNNL total uranium (laser fluorescence) data.

Table O.1. K East Canister Sludge Characterization Data--
per Gram Centrifuged Sludge. (Page 2 of 3)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
Cu.icp	µg/g	5.96E+01	2.60E+02	12	1.13E+02	6.92E+01	61.5	3.02E+02
^{152}Eu.gea	µCi/g	< 3.92E-02	< 6.30E+00	12	NA	NA	NA	NA
^{154}Eu.gea	µCi/g	6.28E-01	1.62E+01	12	6.92E+00	4.90E+00	70.8	2.03E+01
^{155}Eu.gea	µCi/g	2.67E-01	1.06E+01	12	NA	NA	NA	NA
F^-.ic @	µg/g	< 2.45E-02	1.29E-01	10	NA	NA	NA	NA
Fe.icp	µg/g	1.04E+03	8.92E+04	12	1.92E+04	2.56E+04	132.9	8.92E+04
K.icp	µg/g	< 9.95E+01	< 1.06E+03	12	NA	NA	NA	NA
Mg.icp.w	µg/g	< 1.99E+01	1.93E+03	12	7.60E+02	6.92E+02	91.1	2.65E+03
Mg.icp.wo	µg/g	1.03E+02	1.93E+03	9	9.66E+02	6.82E+02	70.6	3.03E+03
Mn.icp.w	µg/g	< 1.86E+01	1.94E+02	12	5.46E+01	5.58E+01	102.1	2.07E+02
Mn.icp.wo	µg/g	3.41E+01	1.94E+02	6	8.93E+01	6.29E+01	70.4	3.23E+02
NO_2^-.ic @	µg/g	< 1.76E-01	< 1.08E+00	10	NA	NA	NA	NA
NO_3^-.ic @	µg/g	< 2.26E-01	1.67E+00	10	NA	NA	NA	NA
Na.icp	µg/g	1.58E+02	4.66E+02	12	3.01E+02	1.02E+02	33.8	5.79E+02
^{94}Nb.gea	µCi/g	< 2.09E-02	< 1.57E+00	12	NA	NA	NA	NA
Ni.icp.w	µg/g	2.03E+01	9.56E+01	12	4.35E+01	1.88E+01	43.2	9.49E+01
Ni.icp.wo	µg/g	2.03E+01	9.56E+01	6	4.71E+01	2.72E+01	57.8	1.48E+02
^{237}Np.w	µCi/g	< 3.81E-03	1.51E-02	12	8.69E-03	4.19E-03	48.3	2.02E-02
^{237}Np.wo	µCi/g	4.07E-03	1.51E-02	8	1.04E-02	4.11E-03	39.7	2.35E-02
Oxalate.ic @	µg/g	< 1.72E-01	< 1.05E+00	10	NA	NA	NA	NA
P.icp	µg/g	< 7.90E+01	< 4.25E+02	12	NA	NA	NA	NA
PO_4^{3-}.ic @	µg/g	< 1.95E-01	< 1.20E+00	10	NA	NA	NA	NA
Pb.icp.w	µg/g	6.18E+01	< 2.12E+02	12	1.50E+02	6.21E+01	41.4	3.20E+02
Pb.icp.wo	µg/g	6.18E+01	1.79E+02	6	1.00E+02	5.03E+01	50.1	2.87E+02
^{238}Pu.w	µCi/g	8.62E-01	< 5.88E+01	12	1.27E+01	1.55E+01	121.9	5.51E+01
^{238}Pu.wo	µCi/g	8.62E-01	1.46E+01	11	8.51E+00	5.65E+00	66.3	2.44E+01
$^{239/240}$Pu	µCi/g	4.52E+00	1.33E+02	12	5.96E+01	4.18E+01	70.1	1.74E+02
^{226}Ra.gea	µCi/g	< 3.02E-01	< 5.28E+01	12	NA	NA	NA	NA
^{106}Ru/Rh.gea	µCi/g	< 2.37E-01	< 3.90E+01	12	NA	NA	NA	NA

*: per gram centrifuged sludge
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.w: less than values were included (i.e. 3 for <3) when calculating the summary statistics.
.wo: less than values were deleted when calculating the summary statistics.
NA: not applicable for these data.
@: Analyses performed using sludge supernate.
&: Calculated using the PNNL total uranium (laser fluorescence) data.

Table 0.1. K East Canister Sludge Characterization Data--per Gram Centrifuged Sludge. (Page 3 of 3)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
SO_4^{2-}.ic.w @	µg/g	7.17E-01	3.57E+00	10	1.69E+00	8.63E-01	51.2	4.20E+00
SO_4^{2-}.ic.wo @	µg/g	7.17E-01	3.57E+00	8	1.80E+00	9.35E-01	51.9	4.78E+00
Se.icp	µg/g	< 1.99E+01	< 2.12E+02	12	NA	NA	NA	NA
Sm.icp	µg/g	< 1.99E+01	< 2.12E+02	12	NA	NA	NA	NA
$^{89/90}$Sr	µCi/g	3.62E+00	1.91E+03	12	8.70E+02	7.48E+02	86.0	2.92E+03
TIC	µg/g	2.51E+02	2.26E+03	12	6.93E+02	5.52E+02	79.7	2.20E+03
TOC	µg/g	3.73E+02	1.60E+03	12	6.81E+02	4.06E+02	59.6	1.79E+03
^{99}Tc	µCi/g	1.08E+00	1.27E+01	5	6.63E+00	4.31E+00	65.0	2.47E+01
Tl.icp	µg/g	< 3.98E+01	< 4.25E+02	12	NA	NA	NA	NA
^{208}Tl.gea	µCi/g	< 2.00E-01	< 3.32E+01	12	NA	NA	NA	NA
Total Carbon	µg/g	5.52E+02	2.76E+03	12	1.31E+03	7.26E+02	55.5	3.29E+03
U.icp	µg/g	1.32E+04	6.52E+05	12	3.47E+05	2.86E+05	82.6	1.13E+06
U.las	µg/g	3.24E+04	7.68E+05	10	4.88E+05	2.83E+05	57.9	1.31E+06
U.phos	µg/g	1.26E+04	6.18E+05	12	2.87E+05	2.37E+05	82.7	9.37E+05
^{233}U.tims &	µg/g	< 1.59E-01	< 3.76E+00	10	NA	NA	NA	NA
^{234}U.tims &	µg/g	4.71E+00	7.08E+01	10	3.99E+01	2.49E+01	62.5	1.13E+02
^{235}U.tims &	µg/g	2.32E+02	5.96E+03	10	3.44E+03	2.12E+03	61.7	9.63E+03
^{236}U.tims &	µg/g	2.46E+01	6.05E+02	10	3.13E+02	1.83E+02	58.5	8.46E+02
^{238}U.tims &	µg/g	3.22E+04	7.63E+05	10	4.84E+05	2.81E+05	58.0	1.30E+06
%Water.grav	µg/g	1.47E+05	9.01E+05	12	4.61E+05	2.96E+05	64.2	1.27E+06
Water.grav	Wt%	1.47E+01	9.01E+01	12	4.61E+01	2.96E+01	64.2	1.27E+02
Zn.icp.w	µg/g	1.86E+01	4.95E+02	12	1.27E+02	1.41E+02	110.8	5.13E+02
Zn.icp.wo	µg/g	1.86E+01	4.95E+02	10	1.49E+02	1.46E+02	98.0	5.73E+02
Zr.icp.w	µg/g	< 1.86E+01	2.18E+02	12	7.86E+01	6.52E+01	83.0	2.57E+02
Zr.icp.wo	µg/g	5.87E+01	2.18E+02	7	1.21E+02	5.37E+01	44.6	3.03E+02
pH @	pH units	7.07E+00	8.38E+00	7	7.74E+00	5.11E-01	6.6	9.47E+00

*: per gram centrifuged sludge
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.w: less than values were included (i.e. 3 for <3) when calculating the summary statistics.
.wo: less than values were deleted when calculating the summary statistics.
NA: not applicable for these data.
@: Analyses performed using sludge supernate.
&: Calculated using the PNNL total uranium (laser fluorescence) data.

Table 0.2. K East Canister Sludge Characterization Data--per Gram As-Settled Sludge. (Page 1 of 4)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
Ag.icp.w	µg/g	1.23E+00	1.52E+02	12	5.82E+01	5.74E+01	98.6	2.15E+02
Ag.icp.wo	µg/g	1.23E+00	1.52E+02	10	6.91E+01	5.70E+01	82.5	2.35E+02
Al.icp	µg/g	5.23E+03	3.60E+04	12	1.38E+04	8.12E+03	58.7	3.60E+04
Al.icp/ms	µg/g	7.79E+02	8.16E+03	2	4.47E+03	5.22E+03	116.8	NA
Alpha Total	µCi/g	4.09E+00	1.78E+02	12	7.55E+01	5.71E+01	75.6	2.32E+02
Alpha Total.PNNL	µCi/g	1.06E+02	2.14E+02	2	1.60E+02	7.63E+01	47.7	NA
^{241}Am.aea.w	µCi/g	3.17E+00	7.35E+01	12	3.17E+01	2.39E+01	75.2	9.70E+01
^{241}Am.aea.wo	µCi/g	3.17E+00	7.35E+01	11	3.31E+01	2.45E+01	73.9	1.02E+02
^{241}Am.gea	µCi/g	2.50E+00	2.23E+02	12	5.82E+01	6.09E+01	104.6	2.25E+02
^{241}Am.gea.PNNL	µCi/g	2.15E-01	5.27E+01	2	2.65E+01	3.71E+01	140.3	NA
B.icp	µg/g	5.33E+01	1.41E+02	12	9.01E+01	3.19E+01	35.4	1.77E+02
Ba.icp.w	µg/g	< 5.30E+00	1.37E+02	12	6.61E+01	3.97E+01	60.1	1.75E+02
Ba.icp.wo	µg/g	1.86E+01	1.37E+02	10	7.04E+01	3.82E+01	54.3	1.82E+02
Be.icp	µg/g	< 5.86E+00	1.81E+01	12	NA	NA	NA	NA
Beta Total	µCi/g	7.43E+00	3.70E+03	12	1.67E+03	1.46E+03	87.5	5.65E+03
Beta Total.PNNL	µCi/g	3.90E+03	8.67E+03	2	6.29E+03	3.38E+03	53.7	NA
Bi.icp.w	µg/g	< 1.06E+01	5.24E+02	12	2.03E+02	1.80E+02	88.7	6.97E+02
Bi.icp.wo	µg/g	2.53E+01	5.24E+02	10	2.37E+02	1.79E+02	75.2	7.57E+02
^{212}Bi.gea	µCi/g	< 8.08E-02	< 1.42E+01	12	NA	NA	NA	NA
Br^-.ic	µg/g	< 3.09E-01	< 1.31E+00	10	NA	NA	NA	NA
Ca.icp.w	µg/g	5.96E+01	6.19E+02	12	3.22E+02	1.91E+02	59.3	8.45E+02
Ca.icp.wo	µg/g	5.96E+01	6.19E+02	9	3.71E+02	1.98E+02	53.5	9.73E+02
Ca.icp/ms	µg/g	4.85E+01	8.62E+01	2	6.74E+01	2.66E+01	39.5	NA
Cd.icp.w	µg/g	< 5.86E+00	4.42E+01	12	1.63E+01	9.93E+00	60.9	4.35E+01
Cd.icp.wo	µg/g	8.98E+00	4.42E+01	10	1.81E+01	9.93E+00	54.8	4.70E+01
^{144}Ce/Pr.gea	µCi/g	< 8.69E-02	< 2.52E+01	12	NA	NA	NA	NA
Cl^-.ic	µg/g	1.90E-01	1.69E+00	10	6.04E-01	4.50E-01	74.6	1.91E+00
$^{243/244}$Cm	µCi/g	< 3.27E-01	< 1.63E+01	12	NA	NA	NA	NA
^{57}Co.gea	µCi/g	1.51E-02	< 6.41E+00	12	NA	NA	NA	NA

*: per gram as-settled sludge
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.w: less than values were included (i.e. 3 for <3) when calculating the summary statistics.
.wo: less than values were deleted when calculating the summary statistics.
NA: not applicable for these data.
&: Calculated using the PNNL total uranium (laser fluorescence) data.

Table 0.2. K East Canister Sludge Characterization Data--per Gram As-Settled Sludge. (Page 2 of 4)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
^{60}Co.gea.W	µCi/g	6.67E-02	< 1.30E+00	12	5.82E-01	3.72E-01	63.9	1.60E+00
^{60}Co.gea.WO	µCi/g	6.67E-02	7.09E-01	7	3.63E-01	2.26E-01	62.3	1.13E+00
^{60}Co.gea.PNNL	µCi/g	1.13E-02	3.18E-01	2	1.65E-01	2.17E-01	131.7	NA
Cr.icp.W	µg/g	< 1.17E+01	1.93E+02	12	4.57E+01	5.17E+01	113.1	1.87E+02
Cr.icp.WO	µg/g	2.28E+01	1.93E+02	6	7.48E+01	6.19E+01	82.7	3.04E+02
^{134}Cs.gea	µCi/g	< 2.20E-02	< 1.79E+00	12	NA	NA	NA	NA
^{134}Cs.gea.PNNL	µCi/g	< 2.05E-03	1.91E-01	2	9.66E-02	NA	NA	NA
^{137}Cs.gea	µCi/g	2.63E+00	9.10E+02	12	3.50E+02	2.75E+02	78.7	1.10E+03
^{137}Cs.gea.PNNL	µCi/g	7.99E-01	4.70E+02	2	2.36E+02	3.32E+02	140.9	NA
Cu.icp	µg/g	4.16E+01	1.80E+02	12	7.84E+01	4.39E+01	56.0	1.99E+02
^{152}Eu.gea	µCi/g	< 2.38E-02	< 5.14E+00	12	NA	NA	NA	NA
^{154}Eu.gea	µCi/g	3.34E-01	1.48E+01	12	5.39E+00	4.25E+00	79.0	1.70E+01
^{154}Eu.gea.PNNL	µCi/g	2.99E-02	7.59E+00	2	3.81E+00	5.34E+00	140.3	NA
^{155}Eu.gea	µCi/g	1.43E-01	9.68E+00	12	NA	NA	NA	NA
^{155}Eu.gea.PNNL	µCi/g	1.24E-02	3.83E+00	2	1.92E+00	2.70E+00	140.5	NA
F^-.ic	µg/g	< 3.61E-02	2.07E-01	10	NA	NA	NA	NA
Fe.icp	µg/g	7.57E+02	6.17E+04	12	1.29E+04	1.74E+04	134.8	6.06E+04
Fe.icp/ms	µg/g	1.15E+03	4.95E+03	2	3.05E+03	2.69E+03	88.2	NA
K.icp	µg/g	< 5.30E+01	< 9.11E+02	12	NA	NA	NA	NA
Mg.icp.W	µg/g	< 1.06E+01	1.76E+03	12	5.98E+02	6.20E+02	103.7	2.29E+03
Mg.icp.WO	µg/g	7.14E+01	1.76E+03	9	7.57E+02	6.42E+02	84.8	2.70E+03
Mn.icp.W	µg/g	< 1.17E+01	1.34E+02	12	3.69E+01	3.67E+01	99.6	1.37E+02
Mn.icp.WO	µg/g	2.10E+01	1.34E+02	6	5.72E+01	4.43E+01	77.6	2.22E+02
NO_2^-.ic	µg/g	< 2.66E-01	< 1.13E+00	10	NA	NA	NA	NA
NO_3^-.ic	µg/g	< 3.43E-01	1.83E+00	10	NA	NA	NA	NA
Na.icp	µg/g	9.20E+01	3.67E+02	12	2.26E+02	1.01E+02	44.6	5.03E+02
^{94}Nb.gea	µCi/g	< 1.16E-02	< 1.43E+00	12	NA	NA	NA	NA
Ni.icp.W	µg/g	1.07E+01	6.11E+01	12	3.14E+01	1.25E+01	39.7	6.55E+01
Ni.icp.WO	µg/g	1.07E+01	6.11E+01	6	2.97E+01	1.77E+01	59.4	9.52E+01

*: per gram as-settled sludge
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.W: less than values were included (i.e. 3 for <3) when calculating the summary statistics.
.WO: less than values were deleted when calculating the summary statistics.
NA: not applicable for these data.
&: Calculated using the PNNL total uranium (laser fluorescence) data.

Table 0.2. K East Canister Sludge Characterization Data-- per Gram As-Settled Sludge. (Page 3 of 4)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
^{237}Np.w	µCi/g	2.59E-03	1.37E-02	12	6.86E-03	3.82E-03	55.7	1.73E-02
^{237}Np.wo	µCi/g	2.59E-03	1.37E-02	8	8.41E-03	3.77E-03	44.9	2.04E-02
Oxalate.ic	µg/g	< 2.60E-01	< 1.11E+00	10	NA	NA	NA	NA
P.icp	µg/g	< 4.60E+01	< 3.65E+02	12	NA	NA	NA	NA
PO_4^{3-}.ic	µg/g	< 2.96E-01	< 1.26E+00	10	NA	NA	NA	NA
Pb.icp.w	µg/g	3.35E+01	< 1.82E+02	12	1.15E+02	6.05E+01	52.7	2.80E+02
Pb.icp.wo	µg/g	3.35E+01	1.24E+02	6	6.43E+01	3.66E+01	57.0	2.00E+02
^{238}Pu.w	µCi/g	4.57E-01	< 5.30E+01	12	1.03E+01	1.42E+01	137.7	4.91E+01
^{238}Pu.wo	µCi/g	4.57E-01	1.29E+01	11	6.43E+00	4.78E+00	74.3	1.99E+01
$^{239/240}$Pu	µCi/g	2.39E+00	1.21E+02	12	4.73E+01	3.79E+01	80.1	1.51E+02
^{239}Pu.icp/ms	µCi/g	1.07E+01	7.71E+01	2	4.39E+01	4.70E+01	107.0	NA
^{240}Pu.icp/ms	µCi/g	< 3.11E-02	3.09E+01	2	1.55E+01	NA	NA	NA
^{226}Ra.gea	µCi/g	< 1.73E-01	< 4.73E+01	12	NA	NA	NA	NA
^{106}Ru/Rh.gea	µCi/g	< 1.37E-01	< 3.53E+01	12	NA	NA	NA	NA
SO_4^{2-}.ic.w	µg/g	1.09E+00	4.18E+00	10	2.13E+00	9.30E-01	43.7	4.83E+00
SO_4^{2-}.ic.wo	µg/g	1.09E+00	4.18E+00	8	2.33E+00	9.38E-01	40.3	5.32E+00
^{125}Sb.gea.PNNL	µCi/g	< 1.37E-02	8.78E-01	2	4.46E-01	NA	NA	NA
Se.icp	µg/g	< 1.06E+01	< 1.82E+02	12	NA	NA	NA	NA
Sm.icp	µg/g	< 1.06E+01	< 1.82E+02	12	NA	NA	NA	NA
$^{89/90}$Sr	µCi/g	1.93E+00	1.74E+03	12	7.17E+02	6.70E+02	93.5	2.55E+03
TIC	µg/g	1.55E+02	1.45E+03	12	4.74E+02	3.39E+02	71.7	1.40E+03
TOC	µg/g	2.67E+02	1.04E+03	12	4.94E+02	2.64E+02	53.3	1.22E+03
^{99}Tc	µCi/g	9.60E-01	8.41E+00	5	5.11E+00	3.08E+00	60.3	1.80E+01
Tl.icp	µg/g	< 2.12E+01	< 3.65E+02	12	NA	NA	NA	NA
^{208}Tl.gea	µCi/g	< 1.16E-01	< 3.02E+01	12	NA	NA	NA	NA
Total Carbon	µg/g	4.36E+02	1.79E+03	12	9.20E+02	4.47E+02	48.6	2.14E+03

*: per gram as-settled sludge
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.w: less than values were included (i.e. 3 for <3) when calculating the summary statistics.
.wo: less than values were deleted when calculating the summary statistics.
NA: not applicable for these data.
&: Calculated using the PNNL total uranium (laser fluorescence) data.

Table 0.2. K East Canister Sludge Characterization Data--per Gram As-Settled Sludge. (Page 4 of 4)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
U.icp	µg/g	6.99E+03	5.94E+05	12	2.82E+05	2.54E+05	90.0	9.77E+05
U.las	µg/g	2.25E+04	7.00E+05	10	4.18E+05	2.71E+05	64.7	1.21E+06
U.phos	µg/g	6.73E+03	5.57E+05	12	2.34E+05	2.13E+05	90.9	8.16E+05
U.icp/ms	µg/g	4.44E+05	5.10E+05	2	4.77E+05	4.70E+04	9.9	NA
^{233}U.tims &	µg/g	< 1.10E-01	< 3.43E+00	10	NA	NA	NA	NA
^{233}U.icp/ms	µg/g	1.59E-01	2.97E-01	2	2.28E-01	9.77E-02	42.9	NA
^{234}U.tims &	µg/g	3.26E+00	6.45E+01	10	3.41E+01	2.33E+01	68.3	1.02E+02
^{234}U.icp/ms	µg/g	2.79E+01	3.38E+01	2	3.09E+01	4.16E+00	13.5	NA
^{235}U.tims &	µg/g	1.61E+02	5.30E+03	10	2.95E+03	2.00E+03	67.8	8.79E+03
^{235}U.icp/ms	µg/g	2.64E+03	4.31E+03	2	3.48E+03	1.18E+03	34.0	NA
^{236}U.tims &	µg/g	1.71E+01	5.51E+02	10	2.63E+02	1.71E+02	64.8	7.60E+02
^{236}U.icp/ms	µg/g	2.30E+02	2.53E+02	2	2.41E+02	1.63E+01	6.8	NA
^{238}U.tims &	µg/g	2.23E+04	6.96E+05	10	4.15E+05	2.68E+05	64.7	1.20E+06
^{238}U.icp/ms	µg/g	4.38E+05	5.04E+05	2	4.71E+05	4.69E+04	10.0	NA
^{238}U.gea.PNNL	µg/g	1.82E+05	< 1.18E+07	2	5.99E+06	NA	NA	NA
Water.grav	µg/g	2.23E+05	9.48E+05	12	5.55E+05	2.91E+05	52.5	1.35E+06
%Water.grav	Wt%	2.23E+01	9.48E+01	12	5.55E+01	2.91E+01	52.5	1.35E+02
Zn.icp.w	µg/g	1.19E+01	3.43E+02	12	8.52E+01	9.49E+01	111.4	3.45E+02
Zn.icp.wo	µg/g	1.19E+01	3.43E+02	10	9.87E+01	9.89E+01	100.2	3.87E+02
Zr.icp.w	µg/g	< 1.17E+01	1.40E+02	12	5.22E+01	3.97E+01	76.0	1.61E+02
Zr.icp.wo	µg/g	4.13E+01	1.40E+02	7	7.79E+01	3.23E+01	41.4	1.88E+02
Zr.icp/ms	µg/g	1.04E+02	2.20E+03	2	1.15E+03	1.48E+03	128.6	NA
m/z241.icp/ms	µg/g	< 1.37E-01	4.00E+01	2	2.00E+01	NA	NA	NA

*: per gram as-settled sludge
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.w: less than values were included (i.e. 3 for <3) when calculating the summary statistics.
.wo: less than values were deleted when calculating the summary statistics.
NA: not applicable for these data.
&: Calculated using the PNNL total uranium (laser fluorescence) data.

Table O.3. K East Canister Sludge Characterization Data--per ml As-Settled Sludge. (Page 1 of 4)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
Ag.icp.w	µg/mL	1.31E+00	3.79E+02	12	1.26E+02	1.40E+02	110.7	5.09E+02
Ag.icp.wo	µg/mL	1.31E+00	3.79E+02	10	1.51E+02	1.41E+02	93.8	5.62E+02
Al.icp	µg/mL	7.40E+03	5.26E+04	12	2.17E+04	1.16E+04	53.3	5.34E+04
Al.icp/ms	µg/mL	1.62E+03	1.91E+04	2	1.03E+04	1.23E+04	119.2	NA
Alpha Total	µCi/mL	4.35E+00	4.17E+02	12	1.56E+02	1.52E+02	97.5	5.71E+02
Alpha Total.PNNL	µCi/mL	2.21E+02	5.00E+02	2	3.60E+02	1.97E+02	54.7	NA
^{241}Am.aea.w	µCi/mL	3.40E+00	1.80E+02	12	6.27E+01	6.17E+01	98.3	2.31E+02
^{241}Am.aea.wo	µCi/mL	3.40E+00	1.80E+02	11	6.48E+01	6.43E+01	99.2	2.46E+02
^{241}Am.gea	µCi/mL	2.66E+00	2.65E+02	12	1.05E+02	9.69E+01	92.2	3.70E+02
^{241}Am.gea.PNNL	µCi/mL	4.49E-01	1.23E+02	2	6.18E+01	8.68E+01	140.4	NA
B.icp	µg/mL	5.68E+01	3.64E+02	12	1.66E+02	1.11E+02	67.1	4.71E+02
Ba.icp.w	µg/mL	< 5.64E+00	2.97E+02	12	1.29E+02	1.05E+02	81.6	4.17E+02
Ba.icp.wo	µg/mL	2.00E+01	2.97E+02	10	1.38E+02	1.08E+02	78.1	4.52E+02
Be.icp	µg/mL	6.87E+00	< 2.72E+01	12	NA	NA	NA	NA
Beta Total	µCi/mL	7.91E+00	1.08E+04	12	3.57E+03	3.82E+03	106.8	1.40E+04
Beta Total.PNNL	µCi/mL	9.11E+03	1.81E+04	2	1.36E+04	6.35E+03	46.7	NA
Bi.icp.w	µg/mL	< 1.13E+01	1.23E+03	12	4.18E+02	4.43E+02	106.0	1.63E+03
Bi.icp.wo	µg/mL	2.71E+01	1.23E+03	10	4.94E+02	4.49E+02	90.9	1.80E+03
^{212}Bi.gea	µCi/mL	< 8.61E-02	< 4.26E+01	12	NA	NA	NA	NA
Br^-.ic	µg/mL	< 6.97E-01	< 1.40E+00	10	NA	NA	NA	NA
Ca.icp.w	µg/mL	6.34E+01	1.45E+03	12	5.60E+02	4.29E+02	76.6	1.73E+03
Ca.icp.wo	µg/mL	6.34E+01	1.45E+03	9	6.14E+02	4.85E+02	79.1	2.09E+03
Ca.icp/ms	µg/mL	1.01E+02	2.02E+02	2	1.51E+02	7.09E+01	46.9	NA
Cd.icp.w	µg/mL	< 8.29E+00	5.69E+01	12	2.74E+01	1.73E+01	63.3	7.49E+01
Cd.icp.wo	µg/mL	9.78E+00	5.69E+01	10	3.04E+01	1.74E+01	57.2	8.11E+01
^{144}Ce/Pr.gea	µCi/mL	< 9.25E-02	< 7.07E+01	12	NA	NA	NA	NA
Cl^-.ic	µg/mL	3.47E-01	3.25E+00	10	1.08E+00	1.13E+00	104.9	4.39E+00
$^{243/244}$Cm	µCi/mL	< 3.51E-01	< 4.33E+01	12	NA	NA	NA	NA
^{57}Co.gea	µCi/mL	2.21E-02	< 1.46E+01	12	NA	NA	NA	NA

*: per mL as-settled sludge
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.w: less than values were included (i.e. 3 for <3) when calculating the summary statistics.
.wo: less than values were deleted when calculating the summary statistics.
NA: not applicable for these data.
&: Calculated using the PNNL total uranium (laser fluorescence) data.

Table 0.3. K East Canister Sludge Characterization Data--
per ml As-Settled Sludge. (Page 2 of 4)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
^{60}Co.gea.W	µCi/mL	7.11E-02	< 3.46E+00	12	1.13E+00	1.02E+00	89.6	3.92E+00
^{60}Co.gea.WO	µCi/mL	7.11E-02	1.66E+00	7	5.63E-01	5.42E-01	96.3	2.40E+00
^{60}Co.gea.PNNL	µCi/mL	2.36E-02	7.44E-01	2	3.84E-01	5.09E-01	132.7	NA
Cr.icp.W	µg/mL	< 1.66E+01	2.40E+02	12	6.34E+01	5.99E+01	94.5	2.27E+02
Cr.icp.WO	µg/mL	2.49E+01	2.40E+02	6	9.03E+01	7.75E+01	85.9	3.78E+02
^{134}Cs.gea	µCi/mL	< 2.36E-02	< 5.05E+00	12	NA	NA	NA	NA
^{134}Cs.gea.PNNL	µCi/mL	< 4.27E-03	4.47E-01	2	2.25E-01	NA	NA	NA
^{137}Cs.gea	µCi/mL	2.80E+00	2.13E+03	12	7.09E+02	7.17E+02	101.1	2.67E+03
^{137}Cs.gea.PNNL	µCi/mL	1.67E+00	1.10E+03	2	5.51E+02	7.76E+02	141.0	NA
Cu.icp	µg/mL	5.28E+01	2.23E+02	12	1.23E+02	5.45E+01	44.4	2.72E+02
^{152}Eu.gea	µCi/mL	< 2.53E-02	< 1.54E+01	12	NA	NA	NA	NA
^{154}Eu.gea	µCi/mL	3.56E-01	3.45E+01	12	1.09E+01	1.08E+01	98.9	4.05E+01
^{154}Eu.gea.PNNL	µCi/mL	6.24E-02	1.77E+01	2	8.90E+00	1.25E+01	140.4	NA
^{155}Eu.gea	µCi/mL	1.52E-01	2.26E+01	12	NA	NA	NA	NA
^{155}Eu.gea.PNNL	µCi/mL	2.59E-02	8.96E+00	2	4.49E+00	6.32E+00	140.6	NA
F^-.ic	µg/mL	< 6.66E-02	3.97E-01	10	NA	NA	NA	NA
Fe.icp	µg/mL	1.07E+03	7.66E+04	12	1.69E+04	2.10E+04	124.0	7.44E+04
Fe.icp/ms	µg/mL	2.39E+03	1.16E+04	2	6.98E+03	6.48E+03	92.9	NA
K.icp	µg/mL	< 5.64E+01	< 2.72E+03	12	NA	NA	NA	NA
Mg.icp.W	µg/mL	< 1.13E+01	4.10E+03	12	1.17E+03	1.44E+03	122.9	5.12E+03
Mg.icp.WO	µg/mL	8.85E+01	4.10E+03	9	1.47E+03	1.57E+03	106.8	6.21E+03
Mn.icp.W	µg/mL	< 1.66E+01	1.66E+02	12	5.22E+01	4.09E+01	78.3	1.64E+02
Mn.icp.WO	µg/mL	2.29E+01	1.66E+02	6	6.79E+01	5.41E+01	79.7	2.69E+02
NO_2^-.ic	µg/mL	< 6.01E-01	< 1.21E+00	10	NA	NA	NA	NA
NO_3^-.ic	µg/mL	< 7.72E-01	1.99E+00	10	NA	NA	NA	NA
Na.icp	µg/mL	9.80E+01	9.53E+02	12	4.28E+02	3.09E+02	72.0	1.27E+03
^{94}Nb.gea	µCi/mL	< 1.23E-02	< 4.28E+00	12	NA	NA	NA	NA
Ni.icp.W	µg/mL	1.14E+01	< 1.09E+02	12	5.41E+01	2.93E+01	54.2	1.34E+02
Ni.icp.WO	µg/mL	1.14E+01	6.56E+01	6	3.50E+01	1.92E+01	54.9	1.06E+02

*: per mL as-settled sludge
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.W: less than values were included (i.e. 3 for <3) when calculating the summary statistics.
.WO: less than values were deleted when calculating the summary statistics.
NA: not applicable for these data.
&: Calculated using the PNNL total uranium (laser fluorescence) data.

Table 0.3. K East Canister Sludge Characterization Data--per ml As-Settled Sludge. (Page 3 of 4)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
^{237}Np.w	µCi/mL	2.76E-03	3.36E-02	12	1.31E-02	1.07E-02	81.8	4.25E-02
^{237}Np.wo	µCi/mL	2.76E-03	3.36E-02	8	1.75E-02	1.07E-02	61.0	5.17E-02
Oxalate.ic	µg/mL	< 5.86E-01	< 1.18E+00	10	NA	NA	NA	NA
P.icp	µg/mL	< 5.01E+01	< 1.09E+03	12	NA	NA	NA	NA
PO$_4^{3-}$.ic	µg/mL	< 6.66E-01	< 1.34E+00	10	NA	NA	NA	NA
Pb.icp.w	µg/mL	3.57E+01	< 5.45E+02	12	2.21E+02	1.76E+02	79.6	7.03E+02
Pb.icp.wo	µg/mL	3.57E+01	1.54E+02	6	7.68E+01	4.52E+01	58.9	2.45E+02
^{238}Pu.w	µCi/mL	4.87E-01	< 1.30E+02	12	2.24E+01	3.61E+01	161.1	1.21E+02
^{238}Pu.wo	µCi/mL	4.87E-01	3.81E+01	11	1.26E+01	1.27E+01	101.1	4.84E+01
$^{239/240}$Pu	µCi/mL	2.55E+00	2.84E+02	12	9.76E+01	9.66E+01	99.0	3.62E+02
^{239}Pu.icp/ms	µCi/mL	2.23E+01	1.80E+02	2	1.01E+02	1.12E+02	110.2	NA
^{240}Pu.icp/ms	µCi/mL	< 6.47E-02	7.23E+01	2	3.62E+01	NA	NA	NA
^{226}Ra.gea	µCi/mL	< 1.85E-01	< 1.39E+02	12	NA	NA	NA	NA
^{106}Ru/Rh.gea	µCi/mL	< 1.46E-01	< 1.06E+02	12	NA	NA	NA	NA
SO$_4^{2-}$.ic.w	µg/mL	< 1.50E+00	5.20E+00	10	3.28E+00	1.39E+00	42.4	7.31E+00
SO$_4^{2-}$.ic.wo	µg/mL	2.66E+00	5.20E+00	8	3.71E+00	1.17E+00	31.6	7.45E+00
^{125}Sb.gea.PNNL	µCi/mL	< 2.85E-02	2.05E+00	2	1.04E+00	NA	NA	NA
Se.icp	µg/mL	< 1.13E+01	< 5.45E+02	12	NA	NA	NA	NA
Sm.icp	µg/mL	< 1.13E+01	< 5.45E+02	12	NA	NA	NA	NA
$^{89/90}$Sr	µCi/mL	2.05E+00	4.71E+03	12	1.56E+03	1.72E+03	110.1	6.26E+03
TIC	µg/mL	2.20E+02	2.12E+03	12	7.23E+02	4.71E+02	65.2	2.01E+03
TOC	µg/mL	3.09E+02	1.27E+03	12	7.90E+02	3.61E+02	45.7	1.78E+03
^{99}Tc	µCi/mL	1.77E+00	1.89E+01	5	9.25E+00	6.88E+00	74.4	3.82E+01
Tl.icp	µg/mL	< 2.26E+01	< 1.09E+03	12	NA	NA	NA	NA
^{208}Tl.gea	µCi/mL	< 1.23E-01	< 9.04E+01	12	NA	NA	NA	NA
Total Carbon	µg/mL	5.47E+02	2.61E+03	12	1.44E+03	6.09E+02	42.4	3.10E+03

*: per mL as-settled sludge
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.w: less than values were included (i.e. 3 for <3) when calculating the summary statistics.
.wo: less than values were deleted when calculating the summary statistics.
NA: not applicable for these data.
&: Calculated using the PNNL total uranium (laser fluorescence) data.

Table 0.3. K East Canister Sludge Characterization Data-- per ml As-Settled Sludge. (Page 4 of 4)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
U.icp	µg/mL	7.45E+03	1.76E+06	12	6.07E+05	6.46E+05	106.3	2.37E+06
U.las	µg/mL	2.79E+04	2.00E+06	10	9.13E+05	7.17E+05	78.5	3.00E+06
U.phos	µg/mL	7.16E+03	1.44E+06	12	5.06E+05	5.49E+05	108.5	2.01E+06
U.icp/ms	µg/mL	9.26E+05	1.19E+06	2	1.06E+06	1.89E+05	17.8	NA
^{233}U.tims &	µg/mL	< 1.36E-01	< 9.80E+00	10	NA	NA	NA	NA
^{233}U.icp/ms	µg/mL	3.71E-01	6.20E-01	2	4.96E-01	1.76E-01	35.4	NA
^{234}U.tims &	µg/mL	4.04E+00	1.93E+02	10	7.51E+01	6.31E+01	84.1	2.59E+02
^{234}U.icp/ms	µg/mL	6.53E+01	7.05E+01	2	6.79E+01	3.70E+00	5.4	NA
^{235}U.tims &	µg/mL	2.00E+02	1.48E+04	10	6.46E+03	5.19E+03	80.3	2.16E+04
^{235}U.icp/ms	µg/mL	6.18E+03	9.00E+03	2	7.59E+03	1.99E+03	26.3	NA
^{236}U.tims &	µg/mL	2.11E+01	1.65E+03	10	5.76E+02	4.98E+02	86.5	2.03E+03
^{236}U.icp/ms	µg/mL	5.27E+02	5.37E+02	2	5.32E+02	6.85E+00	1.3	NA
^{238}U.tims &	µg/mL	2.76E+04	1.98E+06	10	9.06E+05	7.11E+05	78.5	2.98E+06
^{238}U.icp/ms	µg/mL	9.14E+05	1.18E+06	2	1.05E+06	1.88E+05	17.9	NA
^{238}U.gea.PNNL	µg/mL	3.80E+05	< 2.76E+07	2	1.40E+07	NA	NA	NA
Water.grav	µg/mL	5.02E+05	1.01E+06	12	8.01E+05	1.90E+05	23.7	1.32E+06
Zn.icp.w	µg/mL	1.69E+01	4.25E+02	12	1.14E+02	1.11E+02	96.9	4.17E+02
Zn.icp.wo	µg/mL	1.69E+01	4.25E+02	10	1.28E+02	1.17E+02	90.8	4.68E+02
Zr.icp.w	µg/mL	< 1.66E+01	2.04E+02	12	7.40E+01	4.98E+01	67.2	2.10E+02
Zr.icp.wo	µg/mL	4.40E+01	2.04E+02	7	1.00E+02	4.99E+01	49.8	2.70E+02
Zr.icp/ms	µg/mL	2.44E+02	4.59E+03	2	2.42E+03	3.07E+03	127.1	NA
m/z241.icp/ms	µg/mL	< 2.85E-01	9.34E+01	2	4.69E+01	NA	NA	NA

*: per mL as-settled sludge
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.w: less than values were included (i.e. 3 for <3) when calculating the summary statistics.
.wo: less than values were deleted when calculating the summary statistics.
NA: not applicable for these data.
&: Calculated using the PNNL total uranium (laser fluorescence) data.

Table 0.4. K East Canister Sludge Characterization Data--per Gram Dried Sludge. (Page 1 of 4)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
Ag.icp.w	µg/g	< 2.06E+01	1.95E+02	11	9.24E+01	6.63E+01	71.8	2.79E+02
Ag.icp.wo	µg/g	2.35E+01	1.95E+02	9	1.08E+02	6.30E+01	58.2	2.99E+02
Al.icp	µg/g	1.04E+04	1.39E+05	11	6.44E+04	5.38E+04	83.5	2.16E+05
Al.icp/ms	µg/g	1.14E+03	1.03E+04	2	5.71E+03	6.46E+03	113.2	NA
Alpha Total	µCi/g	2.88E+01	3.44E+02	11	1.56E+02	8.63E+01	55.3	3.99E+02
Alpha Total.PNNL	µCi/g	1.55E+02	2.69E+02	2	2.12E+02	8.06E+01	38.0	NA
^{241}Am.aea.w	µCi/g	1.47E+01	2.20E+02	11	7.63E+01	5.61E+01	73.5	2.34E+02
^{241}Am.aea.wo	µCi/g	1.47E+01	2.20E+02	10	8.18E+01	5.60E+01	68.4	2.45E+02
^{241}Am.gea	µCi/g	1.79E+01	9.08E+02	11	1.62E+02	2.52E+02	155.3	8.71E+02
^{241}Am.gea.PNNL	µCi/g	3.15E-01	6.64E+01	2	3.33E+01	4.67E+01	140.1	NA
B.icp	µg/g	1.53E+02	1.02E+03	11	3.31E+02	2.66E+02	80.2	1.08E+03
Ba.icp.w	µg/g	< 1.01E+02	3.34E+02	11	1.61E+02	6.17E+01	38.4	3.34E+02
Ba.icp.wo	µg/g	1.25E+02	3.34E+02	9	1.71E+02	6.36E+01	37.2	3.64E+02
Be.icp	µg/g	< 1.11E+01	1.87E+02	11	NA	NA	NA	NA
Beta Total	µCi/g	1.42E+02	4.66E+03	11	2.82E+03	1.76E+03	62.5	7.78E+03
Beta Total.PNNL	µCi/g	4.91E+03	1.27E+04	2	8.80E+03	5.51E+03	62.6	NA
Bi.icp.w	µg/g	< 2.02E+02	6.63E+02	11	3.64E+02	1.62E+02	44.6	8.21E+02
Bi.icp.wo	µg/g	2.06E+02	6.63E+02	9	3.97E+02	1.61E+02	40.6	8.86E+02
^{212}Bi.gea	µCi/g	< 1.16E+00	< 1.79E+01	11	NA	NA	NA	NA
Br$^-$.ic	µg/g	< 3.89E-01	< 2.51E+01	10	NA	NA	NA	NA
Ca.icp.w	µg/g	< 2.25E+02	2.47E+03	11	1.11E+03	7.62E+02	68.8	3.25E+03
Ca.icp.wo	µg/g	6.98E+02	2.47E+03	8	1.43E+03	6.18E+02	43.1	3.40E+03
Ca.icp/ms	µg/g	7.11E+01	1.09E+02	2	8.98E+01	2.65E+01	29.5	NA
Cd.icp.w	µg/g	< 1.13E+01	2.29E+02	11	7.59E+01	7.58E+01	99.8	2.89E+02
Cd.icp.wo	µg/g	2.02E+01	2.29E+02	10	8.24E+01	7.66E+01	93.0	3.05E+02
^{144}Ce/Pr.gea	µCi/g	< 1.66E+00	< 3.57E+01	11	NA	NA	NA	NA
Cl$^-$.ic	µg/g	2.43E-01	9.71E+00	10	2.74E+00	2.86E+00	104.7	1.11E+01
$^{243/244}$Cm	µCi/g	< 2.36E+00	< 2.17E+01	11	NA	NA	NA	NA
^{57}Co.gea	µCi/g	3.73E-02	< 1.22E+02	11	NA	NA	NA	NA

*: per gram dried sludge
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.w: less than values were included (i.e. 3 for <3) when calculating the summary statistics.
.wo: less than values were deleted when calculating the summary statistics.
NA: not applicable for these data.
&: Calculated using the PNNL total uranium (laser fluorescence) data.

Table 0.4. K East Canister Sludge Characterization Data--per Gram Dried Sludge. (Page 2 of 4)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
^{60}Co.gea.w	µCi/g	6.94E-01	2.51E+00	11	1.46E+00	6.34E-01	43.5	3.24E+00
^{60}Co.gea.wo	µCi/g	6.94E-01	2.51E+00	7	1.59E+00	7.29E-01	45.7	4.07E+00
^{60}Co.gea.PNNL	µCi/g	1.66E-02	4.01E-01	2	2.09E-01	2.71E-01	130.2	NA
Cr.icp.w	µg/g	< 2.23E+01	8.02E+02	11	2.56E+02	2.94E+02	115.1	1.08E+03
Cr.icp.wo	µg/g	1.20E+02	8.02E+02	6	4.49E+02	2.72E+02	60.5	1.46E+03
^{134}Cs.gea	µCi/g	< 1.79E-01	< 2.70E+00	11	NA	NA	NA	NA
^{134}Cs.gea.PNNL	µCi/g	< 3.00E-03	2.41E-01	2	1.22E-01	NA	NA	NA
^{137}Cs.gea	µCi/g	5.01E+01	1.95E+03	11	7.59E+02	5.49E+02	72.3	2.30E+03
^{137}Cs.gea.PNNL	µCi/g	1.17E+00	5.92E+02	2	2.97E+02	4.18E+02	140.9	NA
Cu.icp	µg/g	6.81E+01	1.28E+03	11	4.13E+02	4.56E+02	110.4	1.70E+03
^{152}Eu.gea	µCi/g	< 2.63E-01	< 6.46E+00	11	NA	NA	NA	NA
^{154}Eu.gea	µCi/g	2.52E+00	2.41E+01	11	1.12E+01	6.41E+00	57.1	2.93E+01
^{154}Eu.gea.PNNL	µCi/g	4.38E-02	9.55E+00	2	4.80E+00	6.72E+00	140.1	NA
^{155}Eu.gea	µCi/g	< 1.49E+00	< 1.41E+01	11	NA	NA	NA	NA
^{155}Eu.gea.PNNL	µCi/g	1.82E-02	4.83E+00	2	2.42E+00	3.40E+00	140.4	NA
F^-.ic	µg/g	< 4.62E-02	< 2.40E+00	10	NA	NA	NA	NA
Fe.icp	µg/g	1.20E+03	2.56E+05	11	7.34E+04	8.91E+04	121.4	3.24E+05
Fe.icp/ms	µg/g	1.68E+03	6.23E+03	2	3.95E+03	3.21E+03	81.3	NA
K.icp	µg/g	< 6.60E+02	< 1.27E+03	11	NA	NA	NA	NA
Mg.icp.w	µg/g	< 2.02E+02	2.21E+03	11	1.21E+03	8.10E+02	66.9	3.49E+03
Mg.icp.wo	µg/g	2.96E+02	2.21E+03	8	1.58E+03	6.07E+02	38.4	3.52E+03
Mn.icp.w	µg/g	< 2.23E+01	8.10E+02	11	2.33E+02	3.02E+02	129.8	1.08E+03
Mn.icp.wo	µg/g	9.18E+01	8.10E+02	6	4.08E+02	3.20E+02	78.4	1.59E+03
NO_2^-.ic	µg/g	< 3.35E-01	< 2.16E+01	10	NA	NA	NA	NA
NO_3^-.ic	µg/g	< 4.31E-01	< 2.78E+01	10	NA	NA	NA	NA
Na.icp	µg/g	4.01E+02	1.75E+03	11	6.89E+02	4.04E+02	58.7	1.83E+03
^{94}Nb.gea	µCi/g	< 1.25E-01	< 1.80E+00	11	NA	NA	NA	NA
Ni.icp.w	µg/g	< 4.44E+01	4.98E+02	11	1.36E+02	1.45E+02	107.0	5.45E+02
Ni.icp.wo	µg/g	6.17E+01	4.98E+02	6	2.10E+02	1.66E+02	79.0	8.26E+02

*: per gram dried sludge
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.w: less than values were included (i.e. 3 for <3) when calculating the summary statistics.
.wo: less than values were deleted when calculating the summary statistics.
NA: not applicable for these data.
&: Calculated using the PNNL total uranium (laser fluorescence) data.

Table 0.4. K East Canister Sludge Characterization Data--per Gram Dried Sludge. (Page 3 of 4)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
^{237}Np.w	µCi/g	7.94E-03	4.94E-02	11	2.08E-02	1.33E-02	64.1	5.82E-02
^{237}Np.wo	µCi/g	7.94E-03	4.94E-02	7	1.87E-02	1.41E-02	75.1	6.65E-02
Oxalate.ic	µg/g	< 3.27E-01	< 2.11E+01	10	NA	NA	NA	NA
P.icp	µg/g	< 2.63E+02	1.13E+03	11	NA	NA	NA	NA
PO$_4^{3-}$.ic	µg/g	< 3.72E-01	< 2.40E+01	10	NA	NA	NA	NA
Pb.icp.w	µg/g	1.35E+02	7.66E+02	11	3.40E+02	2.08E+02	61.1	9.26E+02
Pb.icp.wo	µg/g	1.35E+02	7.66E+02	6	4.31E+02	2.55E+02	59.1	1.37E+03
^{238}Pu.w	µCi/g	3.51E+00	< 7.04E+01	11	2.06E+01	1.93E+01	93.8	7.51E+01
^{238}Pu.wo	µCi/g	3.51E+00	4.07E+01	10	1.57E+01	1.06E+01	67.9	4.66E+01
$^{239/240}$Pu	µCi/g	1.72E+01	1.90E+02	11	9.40E+01	4.98E+01	53.0	2.34E+02
^{239}Pu.icp/ms	µCi/g	1.57E+01	9.70E+01	2	5.64E+01	5.75E+01	102.0	NA
^{240}Pu.icp/ms	µCi/g	< 4.54E-02	3.89E+02	2	1.95E+02	NA	NA	NA
^{226}Ra.gea	µCi/g	< 3.30E+00	< 6.81E+01	11	NA	NA	NA	NA
^{106}Ru/Rh.gea	µCi/g	< 2.62E+00	< 5.37E+01	11	NA	NA	NA	NA
SO$_4^{2-}$.ic.w	µg/g	1.44E+00	< 2.76E+01	10	1.10E+01	1.04E+01	94.4	4.14E+01
SO$_4^{2-}$.ic.wo	µg/g	1.44E+00	2.14E+01	8	9.73E+00	9.67E+00	99.4	4.06E+01
^{125}Sb.gea.PNNL	µCi/g	< 2.00E-02	1.11E+00	2	5.63E-01	NA	NA	NA
Se.icp	µg/g	< 1.32E+02	< 2.56E+02	11	NA	NA	NA	NA
Sm.icp	µg/g	< 1.32E+02	< 2.56E+02	11	NA	NA	NA	NA
$^{89/90}$Sr	µCi/g	2.81E+01	2.19E+03	11	1.09E+03	8.23E+02	75.5	3.41E+03
TIC	µg/g	2.89E+02	7.29E+03	11	2.38E+03	2.27E+03	95.1	8.77E+03
TOC	µg/g	4.32E+02	8.45E+03	11	2.46E+03	2.67E+03	108.4	9.98E+03
^{99}Tc	µCi/g	1.23E+00	3.42E+01	5	1.59E+01	1.41E+01	89.1	7.52E+01
Tl.icp	µg/g	< 2.63E+02	< 5.10E+02	11	NA	NA	NA	NA
^{208}Tl.gea	µCi/g	< 2.20E+00	< 4.45E+01	11	NA	NA	NA	NA
Total Carbon	µg/g	6.38E+02	1.04E+04	11	4.41E+03	3.80E+03	86.2	1.51E+04

*: per gram dried sludge
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.w: less than values were included (i.e. 3 for <3) when calculating the summary statistics.
.wo: less than values were deleted when calculating the summary statistics.
NA: not applicable for these data.
&: Calculated using the PNNL total uranium (laser fluorescence) data.

Table 0.4. K East Canister Sludge Characterization Data--per Gram Dried Sludge. (Page 4 of 4)

Analyte	Units *	Minimum Concentration Observed	Maximum Concentration Observed	N	Mean Concentration	Standard Deviation	RSD # (%)	Upper Limit $
U.icp	µg/g	7.79E+04	7.49E+05	11	4.56E+05	2.79E+05	61.1	1.24E+06
U.las	µg/g	9.33E+04	8.81E+05	10	6.93E+05	2.63E+05	38.0	1.46E+06
U.phos	µg/g	7.33E+04	7.40E+05	11	3.86E+05	2.30E+05	59.7	1.03E+06
U.icp/ms	µg/g	6.43E+05	6.50E+05	2	6.46E+05	5.30E+03	0.8	NA
^{233}U.tims	mass%	< 5.00E-04	< 5.00E-04	10	NA	NA	NA	NA
^{233}U.tims &	µg/g	< 4.56E-01	< 4.31E+00	10	NA	NA	NA	NA
^{233}U.icp/ms	µg/g	2.00E-01	4.35E-01	2	3.18E-01	1.66E-01	52.3	NA
^{234}U.tims	mass%	5.10E-03	1.48E-02	10	8.84E-03	2.62E-03	29.6	1.65E-02
^{234}U.tims &	µg/g	1.35E+01	8.65E+01	10	5.64E+01	2.36E+01	41.9	1.25E+02
^{234}U.icp/ms	µg/g	3.52E+01	4.95E+01	2	4.23E+01	1.01E+01	24.0	NA
^{235}U.tims	mass%	4.97E-01	9.55E-01	10	7.09E-01	1.17E-01	16.4	1.05E+00
^{235}U.tims &	µg/g	6.68E+02	7.76E+03	10	4.88E+03	2.13E+03	43.6	1.11E+04
^{235}U.icp/ms	µg/g	3.33E+03	6.32E+03	2	4.82E+03	2.11E+03	43.9	NA
^{236}U.tims	mass%	3.90E-02	9.75E-02	10	6.98E-02	1.86E-02	26.6	1.24E-01
^{236}U.tims &	µg/g	7.07E+01	6.93E+02	10	4.61E+02	1.93E+02	41.8	1.02E+03
^{236}U.icp/ms	µg/g	2.89E+02	3.70E+02	2	3.30E+02	5.73E+01	17.4	NA
^{238}U.tims	mass%	98.983	99.459	10	99.212	1.19E-01	0.1	99.560
^{238}U.tims &	µg/g	9.25E+04	8.76E+05	10	6.88E+05	2.61E+05	38.0	1.45E+06
^{238}U.icp/ms	µg/g	6.35E+05	6.42E+05	2	6.38E+05	4.60E+03	0.7	NA
^{238}U.gea.PNNL	µg/g	2.67E+05	< 1.49E+07	2	7.58E+06	NA	NA	NA
Zn.icp.w	µg/g	2.23E+01	1.42E+03	11	5.16E+02	5.82E+02	112.9	2.15E+03
Zn.icp.wo	µg/g	2.23E+01	1.42E+03	9	6.25E+02	5.91E+02	94.6	2.42E+03
Zr.icp.w	µg/g	< 2.23E+01	8.90E+02	11	3.05E+02	3.19E+02	104.5	1.20E+03
Zr.icp.wo	µg/g	6.71E+01	8.90E+02	7	4.66E+02	2.93E+02	63.0	1.46E+03
Zr.icp/ms	µg/g	1.32E+02	3.22E+03	2	1.68E+03	2.18E+03	130.3	NA
m/z241.icp/ms	µg/g	< 2.00E-01	5.03E+01	2	2.53E+01	NA	NA	NA

*: per gram dried sludge
#: RSD (relative standard deviation); standard deviation divided by the mean.
$: tolerance interval; 95% confidence that 95% of the data lies below the stated value.
.w: less than values were included (i.e. 3 for <3) when calculating the summary statistics.
.wo: less than values were deleted when calculating the summary statistics.
NA: not applicable for these data.
&: Calculated using the PNNL total uranium (laser fluorescence) data.

Table 0.5. K East Sludge Characterization Data--K East Basin Floor/Weasel Pit Versus K East Canisters. (Page 1 of 4)

Analyte	Units as-settled sludge	KE Canister		KE Basin Floor/Weasel Pit	
		Minimum Concentration Observed	Maximum Concentration Observed	Minimum Concentration Observed	Maximum Concentration Observed
Ag.icp	µg/mL	1.31E+00	3.79E+02	< 1.02E+00	< 3.77E+01
Al.icp	µg/mL	7.40E+03	5.26E+04	8.54E+03	5.87E+04
Al.icp/ms	µg/mL	1.62E+03	1.91E+04	NA	NA
Alpha Total	µCi/mL	4.35E+00	4.17E+02	4.52E-01	1.11E+02
Alpha Total.PNNL	µCi/mL	2.21E+02	5.00E+02	NA	NA
^{241}Am.aea	µCi/mL	3.40E+00	1.80E+02	1.57E-01	5.60E+01
^{241}Am.gea	µCi/mL	2.66E+00	2.65E+02	1.82E-01	5.35E+01
^{241}Am.gea.PNNL	µCi/mL	4.49E-01	1.23E+02	NA	NA
B.icp	µg/mL	5.68E+01	3.64E+02	< 4.01E+01	7.66E+02
Ba.icp	µg/mL	< 5.64E+00	2.97E+02	2.32E+01	5.64E+02
Be.icp	µg/mL	6.87E+00	< 2.72E+01	1.76E+00	2.64E+01
Beta Total	µCi/mL	7.91E+00	1.08E+04	2.07E+01	3.09E+03
Beta Total.PNNL	µCi/mL	9.11E+03	1.81E+04	NA	NA
Bi.icp	µg/mL	< 1.13E+01	1.23E+03	NA	NA
^{212}Bi.gea	µCi/mL	< 8.61E-02	< 4.26E+01	< 7.59E-02	< 3.55E+00
Br^-.ic	µg/mL	< 6.97E-01	< 1.40E+00	NA	NA
CN^-	µg/mL	NA	NA	< 3.11E-01	< 1.34E+00
Ca.icp	µg/mL	6.34E+01	1.45E+03	2.68E+02	3.31E+04
Ca.icp/ms	µg/mL	1.01E+02	2.02E+02	NA	NA
Cd.icp	µg/mL	< 8.29E+00	5.69E+01	1.90E+01	7.64E+01
^{144}Ce/Pr.gea	µCi/mL	< 9.25E-02	< 7.07E+01	< 2.93E-01	< 1.15E+01
Cl^-.ic	µg/mL	3.47E-01	3.25E+00	1.70E-02	5.28E-01
$^{243/244}$Cm	µCi/mL	< 3.51E-01	< 4.33E+01	< 1.14E-01	< 1.28E+01
^{57}Co.gea	µCi/mL	2.21E-02	< 1.46E+01	NA	NA
^{60}Co.gea	µCi/mL	7.11E-02	< 3.46E+00	8.46E-02	2.34E+00
^{60}Co.gea.PNNL	µCi/mL	2.36E-02	7.44E-01	NA	NA
Cr.icp	µg/mL	< 1.66E+01	2.40E+02	4.15E+01	1.91E+03

#: 222-S %Water analyses by TGA for the Floor/Weasel Pit samples and by gravimetric for canister samples

Table 0.5. K East Sludge Characterization Data--K East Basin Floor/Weasel Pit Versus K East Canisters. (Page 2 of 4)

Analyte	Units as-settled sludge	KE Canister		KE Basin Floor/Weasel Pit	
		Minimum Concentration Observed	Maximum Concentration Observed	Minimum Concentration Observed	Maximum Concentration Observed
Cr.icp	µg/mL	< 1.66E+01	2.40E+02	4.15E+01	1.91E+03
^{134}Cs.gea	µCi/mL	< 2.36E-02	< 5.05E+00	< 3.83E-02	< 8.83E-01
^{134}Cs.gea.PNNL	µCi/mL	< 4.27E-03	4.47E-01	NA	NA
^{137}Cs.gea	µCi/mL	2.80E+00	2.13E+03	2.73E+01	1.48E+03
^{137}Cs.gea.PNNL	µCi/mL	1.67E+00	1.10E+03	NA	NA
Cu.icp	µg/mL	5.28E+01	2.23E+02	2.46E+01	9.47E+02
DSC.dry (Exotherms) *	Joules/g dry	0.00E+00	5.84E+01	0.00E+00	1.90E+02 222-S 1.12E+01 PNNL
DSC.wet (Exotherms) *	Joules/g wet	0.00E+00	4.49E+01	0.00E+00	6.14E+00 222-S 8.00E+00 PNNL
^{152}Eu.gea	µCi/mL	< 2.53E-02	< 1.54E+01	< 1.40E-02	< 7.02E-01
^{154}Eu.gea	µCi/mL	3.56E-01	3.45E+01	< 1.87E-02	8.90E+00
^{154}Eu.gea.PNNL	µCi/mL	6.24E-02	1.77E+01	NA	NA
^{155}Eu.gea	µCi/mL	1.52E-01	2.26E+01	< 5.41E-02	4.10E+00
^{155}Eu.gea.PNNL	µCi/mL	2.59E-02	8.96E+00	NA	NA
F$^-$.ic	µg/mL	< 6.66E-02	3.97E-01	< 1.08E-03	1.62E+00
Fe.icp	µg/mL	1.07E+03	7.66E+04	6.92E+03	5.22E+05
Fe.icp/ms	µg/mL	2.39E+03	1.16E+04	NA	NA
K.icp	µg/mL	< 5.64E+01	< 2.72E+03	< 6.25E+01	2.54E+03
Mg.icp	µg/mL	< 1.13E+01	4.10E+03	1.28E+02	4.82E+03
Mn.icp	µg/mL	< 1.66E+01	1.66E+02	6.14E+01	9.84E+02
NH3	µg/mL	NA	NA	< 7.52E-02	< 1.23E+01
NO$_2^-$.ic	µg/mL	< 6.01E-01	< 1.21E+00	< 1.41E-02	< 9.04E-01
NO$_3^-$.ic	µg/mL	< 7.72E-01	1.99E+00	3.46E-02	2.15E+00
Na.icp	µg/mL	9.80E+01	9.53E+02	9.03E+01	1.49E+04
^{94}Nb.gea	µCi/mL	< 1.23E-02	< 4.28E+00	< 1.23E-02	< 4.50E-01
Ni.icp	µg/mL	1.14E+01	< 1.09E+02	NA	NA

#: 222-S %Water analyses by TGA for the Floor/Weasel Pit samples and by gravimetric for canister samples

Table 0.5. K East Sludge Characterization Data--K East Basin Floor/Weasel Pit Versus K East Canisters. (Page 3 of 4)

Analyte	Units as-settled sludge	KE Canister		KE Basin Floor/Weasel Pit	
		Minimum Concentration Observed	Maximum Concentration Observed	Minimum Concentration Observed	Maximum Concentration Observed
^{237}Np	µCi/mL	2.76E-03	3.36E-02	< 7.21E-04	< 8.54E-03
Oxalate.ic	µg/mL	< 5.86E-01	< 1.18E+00	NA	NA
P.icp	µg/mL	< 5.01E+01	< 1.09E+03	NA	NA
PO_4^{3-}.ic	µg/mL	< 6.66E-01	< 1.34E+00	< 2.45E-02	< 2.38E+00
Pb.icp	µg/mL	3.57E+01	< 5.45E+02	4.20E+01	1.08E+03
^{238}Pu	µCi/mL	4.87E-01	< 1.30E+02	2.83E-02	< 1.09E+01
$^{239/240}$Pu	µCi/mL	2.55E+00	2.84E+03	1.84E-01	4.48E+01
^{239}Pu.icp/ms	µg/mL	3.60E+02	2.91E+03	NA	NA
^{240}Pu.icp/ms	µg/mL	< 2.85E-01	3.19E+02	NA	NA
^{226}Ra.gea	µCi/mL	< 1.85E-01	< 1.39E+02	< 5.86E-01	< 2.20E+01
^{106}Ru/Rh.gea	µCi/mL	< 1.46E-01	< 1.06E+02	< 4.27E-01	< 1.68E+01
SO_4^{2-}.ic	µg/mL	< 1.50E+00	5.20E+00	2.45E-02	2.03E+01
^{125}Sb.gea.PNNL	µCi/mL	< 2.85E-02	2.05E+00	< 3.02E-02	< 3.75E+00
Se.icp	µg/mL	< 1.13E+01	< 5.45E+02	< 1.02E+01	< 3.77E+02
Sm.icp	µg/mL	< 1.13E+01	< 5.45E+02	< 1.02E+01	< 3.77E+02
$^{89/90}$Sr	µCi/mL	2.05E+00	4.71E+03	1.77E+00	1.38E+03
%Water (222S) #	Wt%	2.23E+01	9.48E+01	4.54E+01	9.05E+01
%Water.tga (PNNL)	Wt%	1.77E+01	8.13E+01	9.51E+00	8.53E+01
TIC	µg/mL	2.20E+02	2.12E+03	7.50E+00	3.26E+03
TOC	µg/mL	3.09E+02	1.27E+03	3.87E+02	4.03E+03
^{99}Tc	µCi/mL	1.77E+00	1.89E+01	NA	NA
Tl.icp	µg/mL	< 2.26E+01	< 1.09E+03	< 2.04E+01	< 7.55E+02
^{208}Tl.gea	µCi/mL	< 1.23E-01	< 9.04E+01	< 9.51E-02	< 1.19E+01
Total Carbon	µg/mL	5.47E+02	2.61E+03	6.68E+02	6.55E+03
U.icp	µg/mL	7.45E+03	1.76E+06	1.14E+03	9.18E+04
U.las	µg/mL	2.79E+04	2.00E+06	1.44E+03	4.27E+05
U.phos	µg/mL	7.16E+03	1.44E+06	1.31E+03	3.67E+04
U.icp/ms	µg/mL	9.26E+05	1.19E+06	NA	NA

#: 222-S %Water analyses by TGA for the Floor/Weasel Pit samples and by gravimetric for canister samples

Table 0.5. K East Sludge Characterization Data--K East Basin Floor/Weasel Pit Versus K East Canisters. (Page 4 of 4)

Analyte	Units as-settled sludge	KE Canister		KE Basin Floor/Weasel Pit	
		Minimum Concentration Observed	Maximum Concentration Observed	Minimum Concentration Observed	Maximum Concentration Observed
^{233}U.tims	µg/mL	< 1.36E-01	< 9.80E+00	< 1.41E-02	< 4.18E+00
^{233}U.icp/ms	µg/mL	3.71E-01	6.20E-01	NA	NA
^{234}U.tims	µg/mL	4.04E+00	1.93E+02	1.10E-01	3.49E+01
^{234}U.icp/ms	µg/mL	6.53E+01	7.05E+01	NA	NA
^{235}U.tims	µg/mL	2.00E+02	1.48E+04	1.00E+01	3.10E+03
^{235}U.icp/ms	µg/mL	6.18E+03	9.00E+03	NA	NA
^{236}U.tims	µg/mL	2.11E+01	1.65E+03	1.10E+00	2.12E+02
^{236}U.icp/ms	µg/mL	5.27E+02	5.37E+02	NA	NA
^{238}U.tims	µg/mL	2.76E+04	1.98E+06	1.43E+03	4.24E+05
^{238}U.icp/ms	µg/mL	9.14E+05	1.18E+06	NA	NA
^{238}U.gea.PNNL	µCi/mL	1.28E-01	< 9.29E+00	NA	NA
Zn.icp	µg/mL	1.69E+01	4.25E+02	5.44E+01	2.09E+03
Zr.icp	µg/mL	< 1.66E+01	2.04E+02	2.82E+01	1.06E+03
Zr.icp/ms	µg/mL	2.44E+02	4.59E+03	NA	NA
m/z241.icp/ms	µg/mL	< 2.85E-01	9.34E+01	NA	NA
Residue	µg/mL	NA	NA	5.80E+03	4.45E+05

#: 222-S %Water analyses by TGA for the Floor/Weasel Pit samples and by gravimetric for canister samples

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