Hanford Tank 241-C-106: Residual Waste Contaminant Release Model and Supporting Data

May 2005

The cover image of tank C-106 residual material is from a scanning electron microscope. See page 3.40 for discussion.

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Hanford Tank 241-C-106: Residual Waste Contaminant Release Model and Supporting Data

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May 2005

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Summary

The CH2M HILL Hanford Group, Inc. (CH2M HILL) is producing risk/performance assessments to support the closure of single-shell tanks at the U.S. Department of Energy's Hanford Site. As part of this effort, staff at Pacific Northwest National Laboratory were asked to develop release models for contaminants of concern that are present in residual sludge remaining in tank 241-C-106 (C-106) after final retrieval of waste from the tank. The primary contaminants of concern in the sludge are ^{99}Tc, ^{238}U, ^{129}I, and Cr because of their potential mobility in the environment and the long half-lives of the radionuclides. A key result from this work is that high percentages (>90%) of these primary contaminants are not readily leachable from the residual waste. This minimizes their future release rates to the environment, and is similar to ^{99}Tc results found in related studies of sludges from tanks AY-102, C-203, and C-204.

Developing release models consists of laboratory testing to produce contaminant release data and a conceptual source release model. After development, the release model can be incorporated into a fate and transport model as part of a long-term risk assessment for the closed tank.

Initial laboratory tests (Tier 1) were conducted to characterize the sludge and identify water-leachable constituents. Based on the results of Tier 1 tests, additional analyses were performed to augment the characterization of the material and determine the controlling mechanism(s) for release of contaminants. Tier 2 tests consisted of X-ray diffraction (XRD) and scanning electron microscopy/energy dispersive spectrometry (SEM/EDS) analyses of the solids to identify reactive phases, and selective extractions to quantify the release of contaminants from particular solid phases.

The laboratory results of sludge and liquid testing were used to develop source term models that describe the release of contaminants as infiltrating water contacts the solids in the future. These models simulate the geochemical system in the tank sludge and take into account interactions between the solution phase and the contaminant-containing solids. The release models are simplifications of the complex geochemical interactions occurring; however, they adequately represent the release of the primary contaminants of concern from the sludge as measured in the laboratory tests.

Because of the highly complex chemical nature of tank C-106 residual sludge, clear and quantitative phase associations of the contaminants of concern with the phases known to exist in the sludge are difficult to specify. Although the various characterization methods employed in this study have revealed a number of important observations and have provided valuable data for constructing a scientifically defensible release model, many questions remain. Because a thorough understanding of all the important phase associations for the contaminants of concern cannot be developed at this time, an empirically based release model has been developed. Although less satisfying from a mechanistic point of view, this approach provides a release model that can be used now and is conservative in nature. Later work may provide a better understanding of the phase associations with the contaminants of concern and the release mechanisms from these phases. In this case, a less conservative, but more scientifically defensible release model could be developed.

The major conclusions from this work are summarized in the following items:

- The total concentrations of the primary contaminants of concern are relatively low in the residual C-106 sludge. The average ^{99}Tc concentration is 1.2 µg/g and the ^{238}U concentration is 247 µg/g. The ^{129}I concentration had an average value of 0.62 µg/g (110 pCi/g). The Cr concentration in the sludge was near the instrument quantification limit, and was estimated to be 897 µg/g.

- Only 2 to 3.5% of the ^{99}Tc in the residual sludge was water leachable and only 1.3 to 4% of the ^{238}U was leachable. The water leachability of ^{129}I from the sludge was found to be much less than 1%. The Cr concentrations in solutions from the water leach tests were below the estimated quantitation limit; however the semi-quantitative results show leachabilities from 1 to 7%.

- Mechanistic release models of contaminants from the sludge could not be developed from the available data because of the complexity of the geochemical system. Empirical release models based on measured total sludge concentrations and maximum solution concentrations in water leaching tests should be used for risk/performance assessment modeling. These models are expected to provide conservatively high release rates into fresh water percolating through the residual waste.

- The concentrations of major metals and anions in the sludge were:
  - Al – 131,483 µg/g
  - Mn – 117,767 µg/g
  - Fe – 43,777 µg/g
  - Ca – 38,221 µg/g
  - Na – 60,400 µg/g
  - oxalate – 63,900 µg/g
  - carbonate – 39,500 µg/g

- The oxalic acid treatment of sludge in the tank during retrieval removed much of the water-leachable constituents from the solid. Leachable amounts of Al and Fe were barely detectable, while about 6% of the Ca, 40% of the Mn and 50% of the Na were water leachable. The primary water leachable anions were oxalate and carbonate.

- Several of the metals and contaminants could not be leached from the sludge even under the condition of aggressive leaching with concentrated nitric acid. It was found that 56% of the ^{99}Tc could not be leached under these conditions and 40% of the Cr was recalcitrant. Fe and Al were also relatively immobile with 65% and 41% not leachable, respectively.

- The XRD results indicate that the unleached sludge samples contain detectable quantities of the following crystalline phases:
  - lindbergite [$MnC_2O_4 \cdot 2H_2O$]
  - gibbsite [$Al(OH)_3$]
  - dawsonite [$NaAlCO_3(OH)_2$]
  - hematite (Fe_2O_3)
  - böhmite [$AlO(OH)$]
  - rhodochrosite ($MnCO_3$)
  - whewellite (Ca oxalate monohydrate, $CaC_2O_4 \cdot H_2O$)

Testing of sludge from tank C-106 has shown the need for future development of analytical techniques and release models. These developments include the following:

- Improved selective extraction methods to identify the solid phases containing trace levels of contaminants

- Testing of contaminant release from sludge under environmental conditions other than fresh water infiltration (e.g., cement grout/sludge system)

Acknowledgments

Acronyms and Abbreviations

AEA	alpha energy analysis
ASTM	American Society for Testing and Materials
BSE	backscattered electron
CCV	continuing calibration verification
CH2M HILL	CH2M HILL Hanford Group, Inc.
DDI	double deionized (water)
DOE	U.S. Department of Energy
DUP	duplicate sample
EDS	energy-dispersive X-ray spectrometry
EPA	U.S. Environmental Protection Agency
EQL	estimated quantitation limit
GEA	gamma energy analysis
HASQARD	Hanford Analytical Services Quality Assurance Requirements Document
HF	hydrofluoric
ICP-MS	inductively coupled plasma-mass spectroscopy (spectrometer)
ICP-OES	inductively coupled plasma-optical emission spectroscopy (same as ICP-AES)
ICDD	International Center for Diffraction Data, Newtown Square, Pennsylvania
JCPDS	Joint Committee on Powder Diffraction Standards
LEPS	Low Energy Photon System
LSC	liquid scintillation counting
NIST	National Institute of Standards and Technology
PDF™	powder diffraction file
pe	redox potential ($-\log a_{e^-}$)
PNNL	Pacific Northwest National Laboratory
QA	quality assurance
RPL	Radiochemical Processing Laboratory
SBMS	Standards-Based Management System
SE	secondary electron
SEM	scanning electron microscopy (or microscope)
SI	Saturation Index
SRM	Standard Reference Material
TEM	transmission electron microscopy (or microscope)
TRU	transuranic (waste)
XRD	X-ray powder diffractometry analysis (commonly called X-ray diffraction)

Units of Measure

Å   angstrom

θ   angle of incidence (Bragg angle)

$\Delta_f G_{298}^{\circ}$ Gibbs energy of formation from the elements in their reference states at 298.15 K

ºC   temperature in degrees Celsius [T(ºC) = T(K) – 273.15)

Ci   Curie

eV   electron volt

ft   foot

g   gram

gal   gallon

in.   inch

K   temperature in degrees (without degree symbol) Kelvin [T(K) = T(ºC) + 273.15]

K_{298}° equilibrium constant at 298.15 K

kcal   kilocalorie, one calorie equals 4.1840 joules

keV   kilo-electron volt

kJ   kilojoule, one joule equals 4.1840 thermochemical calories

L   liter

μ   micro (prefix, 10^{-6})

μCi   microCurie

μeq   microequivalent

μg   microgram

μm   micrometer

M   molarity, mol/L

mA   milliAmpere

mg   milligram

mL   milliliter

mm   millimeter

mM   millimolar

mol   mole

nCi   nanoCurie

ng   nanogram

rpm   revolution per minute

I/I_o relative intensity of an XRD peak to the most intense peak

λ   wavelength

wt%   weight percent

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1.0 Introduction

This report describes the development of release models for contaminants of concern that may be present in residual sludge in Hanford tank 241-C-106 (C-106) after closure. These release models are necessary components of the risk assessments being conducted as part of the closure process. From the perspective of long-term risk to the environment, the primary contaminants of concern are ^{99}Tc, ^{238}U, ^{129}I, and Cr because of their mobility in the environment and long half-lives for the radionuclides. Sludge samples and a sample of the liquid from tank C-106 were collected after final sludge retrieval to characterize the geochemistry of the reactive phases and to quantify the release of primary contaminants into water that may contact residual sludge after tank closure.

The remainder of this section describes the scope of work for laboratory testing and release model development as well as background information on this tank. The samples and laboratory testing procedures for this project are described in Section 2 of this report, and the results are provided in Section 3. Release models are discussed in Section 4 and general conclusions in Section 5. Cited references are listed in Section 6, and supporting material is included in the appendices.

1.1 Scope

Initial (Tier 1) laboratory tests were conducted to characterize the sludge and identify water-leachable constituents. The Tier 1 tests consisted primarily of fusion and acid digestions (which measured element concentrations in the solid) and water leaching of contaminants from the sludge to evaluate their mobility in infiltrating water. Based on the results of Tier 1 tests, additional analyses were performed to augment the characterization of the material and determine the controlling mechanism(s) for release of contaminants. Tier 2 tests consisted of analyses of the solids using X-ray diffraction (XRD) and scanning electron microscopy/energy dispersive spectrometry (SEM/EDS) to identify reactive phases, and selective extractions in order to quantify the release of contaminants from particular solid phases.

The laboratory results of residual sludge and liquid testing were used to develop source term models that describe the release of contaminants as infiltrating water contacts the solids in the future. These models simulate the geochemical system in the tank sludge and take into account interactions between the solution phase and the contaminant-containing solids. The release models are simplifications of the complex geochemical interactions occurring between the phases; however, they adequately represent the release of the key contaminants ^{99}Tc, ^{238}U, ^{129}I, and Cr from the sludge as measured in laboratory tests.

1.2 C-106 Tank Description

Tank C-106 is a single-shell underground waste tank located in the C Tank Farm in the 200 East Area of the Hanford Site (Figure 1.1). It was constructed between 1943 and 1944 and put into service during September 1947. This tank is 22.8 m (75 ft) in diameter and has a capacity of 2,006,268 L (530,000 gal) when filled to a depth of 5.2 m (17 ft). Figure 1.2 is a diagram showing the configuration of tank C-106.

Figure 1.1. Hanford C Tank Farm

1.2

Table 1.1 shows the history of waste transfers into and removals from tank C-106 through March 1979 when it was removed from service. This table is from Schreiber et al. (1996, Table 2-3) who obtained the information from Anderson (1990) and Agnew et al. (1995). The transfer history shows that a large variety of waste has been stored in this tank throughout its operational period. The tank was placed on the High-Heat Load Watch List in January 1991, and active ventilation was used to cool the tank by evaporation. The heat was produced by decay of radionuclides in the waste, principally ^{90}Sr. Periodic water additions were made to replace evaporated moisture. In 1995, the *Environmental Assessment* (DOE 1995) stated that approximately 22,712 L (6,000 gal) of water were added to the tank each month for cooling purposes. As of May 31, 1996, the tank contained an estimated 866,859 L (229,000 gal) of waste (745,726 L [197,000 gal] of sludge plus 121,133 L [32,000 gal] of supernatant liquid) classified as non-complexed (Hanlon 1996; Schreiber et al. 1996). From November 1998 through September 1999, the contents of tank C-106 were sluiced to tank 241-AY-102 using supernatant from tank 241-AY-102 as the sluicing fluid. Approximately, 97% of the waste was removed, which left approximately 68,137 L (18,000 gal) in the tank.

In late 2003, sludge from tank C-106 was further removed using a 0.9 M oxalic acid solution to dissolve and suspend the solids and pump out as much as possible. The goal was to lower the sludge volume from about 68,137 to 10,599 L (18,000 to 2,800 gal [360 ft^3]). This goal was achieved by several additions and removals of the oxalic acid solution. After the final removal, the sludge was rinsed with water to remove as much of the acid solution as possible. Approximately 151,416 L (40,000 gal) or 35.5 cm (14 in.) of water was added to measure the sludge volume, and then as much liquid as possible was removed. A 0.5 M NaOH solution was added to neutralize the residual waste, and then removed. The residual liquid in the tank was sampled for analysis through Riser 14 (Figure 1.2), and multiple sludge samples were acquired using a clamshell device by CH2M HILL in January 2004. All samples were delivered to the Hanford 222-S Laboratory for processing and characterization. Figure 1.3 is a picture of the sludge at the 222-S Laboratory. Subsamples were sent to PNNL for testing and release model development.

Figure 1.2. Tank C-106 Configuration (Conner 1996)

1.4

Figure 1.3. Tank C-106 Sludge at 222-S Laboratory

Table 1.1. Tank C-106 Waste Transfer History through 1979 (from Schreiber et al. 1996)

Transfer Location	Waste Type	Time	Estimated Waste Volume[a]	
			In[b]	Out[b]
241-C-105	Metal waste supernate	1947	530,000 gal	NA
241-BY-102, 241-C-104, 241-C-105, 241-C-202, 241-C-203, 241-C-204	Metal waste	1953	1,387,000 gal	NA
Miscellaneous	Flush water	1953	1,828,000 gal	NA
U-Plant	Sluicing metal waste for uranium recovery	1953-1954	538,000 gal	3,680,000 gal
Miscellaneous	Flush water	1957	164,000 gal	NA
241-C-109, 241-C-112	Uranium recovery waste for ferrocyanide scavenging	1957	NA	550,000 gal
241-A-101, 241-A-102	Organic wash waste and high-level PUREX waste	1957	481,000 gal	NA
241-BY-103, 241-BY-110	Supernatant	1957-1958	NA	463,000 gal
PUREX	Cladding waste	1958, 1960	420,000 gal	NA
241-B-101, 241-B-107	Supernatant	1963	NA	353,000 gal
241-A-102	PUREX waste	1963-1964	427,000 gal	99,000 gal
CR Vault	Decontamination waste	1965	36,000 gal	NA
241-C-105	Supernatant	1968-1969	NA	637,000 gal
244-AR Vault, 241-A-106	Washed PUREX waste	1969-1970	1,019,000 gal	NA
241-A-102, 241-C-103, 241-C-105	PUREX and low-level B-Plant supernate	1970-1971	638,000 gal	1,446,000 gal
Miscellaneous	Flush water	1971	27,000 gal	NA
244-AR Vault, 241-A-106	Washed PUREX waste	1971-1972	151,000 gal	NA
241-AX-103	Supernatant	1974	NA	221,000 gal
Miscellaneous	Flush Water	1974-1975	26,000 gal	NA
241-C-103, 241-C-104	Supernatant	1974-1976	NA	2,392,000 gal
B-Plant	Low-level B-Plant waste	1974-1976	2,937,000 gal	NA
241-A-102	Strontium recovery waste from B-Plant	1976-1977	247,000 gal	362,000 gal
241-AZ-101	Supernatant	1978	NA	85,000 gal
241-A-102	Complexed and evaporator waste	1978-1979	365,000 gal	445,000 gal

(a) Waste volumes do not include unknown transfers or transfers out to the condenser.
(b) Volumes are given in English units; to convert to liters (metric), multiple gallons by 3.785.
NA = Not applicable.

2.0   Materials and Laboratory Test Methods

Sludge and liquid samples from tank C-106 were collected by CH2M HILL during post retrieval activities in January 2004. This section provides a description of the samples and the various tests used to characterize the material, measure contaminant release, and identify controlling solids.

2.1   Tank C-106 Samples

On January 26, 2004, after neutralization of tank waste with NaOH and during pumping of residual fluid from tank C-106, a sample of the liquid was collected and transported to the Hanford 222-S Laboratory for storage and analysis. On January 29, 2004, nine clamshell samples of residual sludge were collected from the tank and sent to the 222-S Laboratory. On January 30 the sludge samples were extruded in the 11A Hot Cell. Part of the extruded samples was combined to produce the Field Primary Solid Composite. The remaining sludge was combined to produce the Field Duplicate Solid Composite. On April 13, 2004, samples of the sludge and liquid were shipped to the PNNL Radiochemical Processing Laboratory (RPL). Table 2.1 lists the samples received by PNNL.

Testing of sludge samples to develop contaminant release models was conducted on Field Primary Solid Composite S04T000404 (primary 404, Figure 2.1) and Field Duplicate Solid Composite S04T000405 (duplicate 405, Figure 2.2). Liquid sample S04T000403 (403) was also analyzed to determine the porewater concentrations of contaminants in the sludge.

Table 2.1. Tank C-106 Samples Provided by 222-S Laboratory to PNNL

Sample	Jar Size (mL)	Labcore Number	Net Weight/Volume of Material (g)
Field Primary Solid Composite	60	S04T000109	20.4
Field Primary Solid Composite	60	S04T000404	20.6
Field Duplicate Solid Composite	60	S04T000405	20.1
6C-03-11 (Liquid)	60	S04T000403	61.6

2.2   Sludge Composition by Fusion and Acid Digestions

The bulk compositions of the sludge solids were determined using PNNL internal procedure AGG-ESL-001[a] and a modified version of U.S. Environmental Protection Agency (EPA) Method 3050B (EPA 1996a). These methods were used to measure the element composition of the sludge, but are not appropriate for the anion concentrations due to the acids used in the analyses. The anion compositions were measured separately in solutions obtained by water leaching of the solids (see Section 2.5.5).

(a) Lindberg MJ. 2003. "Solubilization of Metals from Solids Using a KOH-KNO$_3$ Fusion." AGG-ESL-001 (Rev. 0), unpublished PNNL Technical Procedure, Pacific Northwest National Laboratory, Richland, Washington.

Figure 2.1. Tank C-106 Field Primary Solid Composite Sludge Sample 404

Figure 2.2. Tank C-106 Field Duplicate Solid Composite Sludge Sample 405

The potassium hydroxide (KOH)-potassium nitrate (KNO_3) fusion-dissolution procedure is the most commonly used method for solubilization of Hanford tank sludge samples for chemical analysis by inductively coupled plasma-mass spectroscopy (ICP-MS) and other methods (De Lorenzo et al. 1994; Simpson 1994; Fiskum et al. 2000; Smith et al. 2001). Benefits of this procedure include effective metathesizing of insoluble salts such as $SrSO_4$, $PuPO_4$, PuF_3, and ThF_3 into acid soluble hydroxides; fusion completed at relatively low temperature (550°C) compared to other fluxing agents, such as 1100°C for the $LiBO_2$ (lithium metaborate) fluxing agent; and use of nickel or zirconium crucibles, as opposed to the more costly platinum crucibles, for the fusion.[a]

The KOH-KNO_3 fusion-dissolution procedure consists of chemical analyses of a solution resulting from water and acid dissolutions of a solid that has been fused at a high temperature with a caustic fluxing agent. In this procedure, 0.3 g of the tank waste sludge material was mixed with 10 mL of a 20% KOH and 2% KNO_3 solution as a fluxing agent in a zirconium crucible. The crucible was then placed on a hot plate and allowed to evaporate to dryness, after which it was covered and transferred to a muffle furnace preheated to 550ºC. Fusion was accomplished by heating the sample-flux mixture for 60 minutes at 550ºC. After 60 minutes, the crucible was removed from the furnace and allowed to cool to ambient room temperature. The fused solid was then dissolved in double deionized (DDI) water. The resulting solution was transferred to a 100-mL volumetric flask to which 1 mL of 1.0 M hydroxylamine HCl was added. Ten mL of an 8-M nitric acid (HNO_3) solution was added to the crucible to try to dissolve any remaining residual solid. The acid wash solution was also added to the volumetric flask. The crucible was then triple-rinsed with DDI water, and these washes were also added to the volumetric flask. The resulting solution was diluted up to a total volume of 100 mL with DDI water. Prior to chemical analysis, the final 100-mL solution was passed through a Whatman 41 filter with final filtration through a 0.45-µm pore-size syringe filter to remove any particles that did not dissolve in the HNO_3 solution. The insoluble fractions were not characterized because of the difficulty in removing them from the syringe filter media.

Chemical analyses of an acid digestion of the sludge solids were also completed for comparison with the KOH-KNO_3 fusion procedure. For the acid digestion, 0.3 g of tank sludge was digested following the basic procedure described in U.S. Environmental Protection Agency (EPA) Method 3050B (EPA 1996a) with the following exceptions: 1) all reagent volumes were reduced by 50% because safety considerations necessitated smaller sample masses due to the high radioactivity levels associated with these sludge solids, and 2) HCl was not added during the digestion because it interferes with analysis of the resulting solutions by ICP-MS. If HCl is used, an $ArCl^+$ species is formed during ICP-MS analysis, which creates a spectral interference that impedes analysis of certain analytes. Throughout the remainder of this report, this treatment of sludge solids will be referred to as "acid digestion."

For the acid digestion procedure, 300 mg of the sample was placed in a 50 mL Griffin beaker, 8 M HNO_3 was added to the sample, the beaker was covered with a watch glass, and the unit was heated to 95°C. Successive additions of concentrated HNO_3 and heating are performed until no reaction with the sample was noted. The sample was then allowed to digest for two hours at 95°C. The sample was removed and allowed to cool, 30% H_2O_2 was added to the sample, and the temperature was increased to 95°C. Successive addition of H_2O_2 and heating are performed until no reaction was noticed. The sample was allowed to digest for 2 hours at 95°C. After cooling, the solution was filtered through a Whatman 41

(a) Personal communication from WI Winters (CH2M HILL) to the authors, December 22, 2003.

filter to remove any insoluble particles and brought to a final volume of 50 mL with DDI water. Samples were filtered through a 0.45-µm pore-size syringe filter prior to analysis. The limited quantity of the insoluble fractions and the inability to remove this material from the filter media precluded their characterization.

Table 2.2 lists the digestion factors (wet solid-to-solution ratios) for the samples of C-106 sludge solids used for the KOH-KNO$_3$ fusion treatments and acid digestion. These factors were calculated from the wet weight of sludge material divided by the volume of extracting solution. The digestion factors were then multiplied by the percent solids, as determined from moisture content analysis, to convert to a dry weight basis. All EPA acid-digestion and fused-sample solutions were filtered using 0.45-µm pore-size syringe filters prior to analysis. The dissolved concentrations and the total beta and total alpha activities for the filtered solutions were then analyzed by a combination of methods, including ICP-MS, inductively coupled plasma-optical emission spectroscopy (ICP-OES), and several radiochemical analytical techniques. These analytical methods are described in Lindberg and Deutsch (2003).

Table 2.2. Digestion Factors for Samples of Tank C-106 Residual Sludge Solids Used for the EPA Acid Digestion and KOH-KNO$_3$ Fusion Treatments

Treatment	Sample Number	Dry Weight Corrected Digestion Factor (g/L)
KOH-KNO$_3$ fusion	404	2.0880
	404 DUP	1.8505
	405	2.5348
	405 DUP	2.1843
EPA acid digestion	404	4.0515
	404 DUP	3.4368
	405	4.6821
	405 DUP	3.8920
(a) DUP = Duplicate sample.		

2.3   XRD Analysis

Crystalline phases present in the unleached (as-received), 1-month and 82-day water-leached, and hydrofluoric (HF)-extracted sludge samples 404 and 405 from tank C-106 were characterized by standard powder XRD techniques. The water leach and selective extraction tests are described in Sections 2.5 and 2.6, respectively. Because the sludge materials were highly radioactive, dispersible powders, it was necessary to prepare the XRD mounts of these samples inside a fumehood regulated for handling radioactive materials. Sludge samples were prepared for XRD analysis by placing milligram quantities of each sample into a mixture of water and collodion solution. The collodion solution consists of 2% nitrocellulose dissolved in amyl acetate, and is an X-ray amorphous, viscous binder commonly used to make random powder mounts for XRD when only a limited amount of sample is available. Apparent shifts in *d*-spacings (i.e., measured 2θ angles) were observed in some of the XRD patterns for the water leached and HF-extracted sludge samples. To determine if these shifts were due to a minor misalignment of the mounted sample or variations in the composition of one or more of the crystalline solids detected by XRD, mounts of the 82-day water leached samples were also prepared with the addition of trace

quantities of reference-material corundum powder (α-Al_2O_3, alumina) [National Institute of Standards and Technology Standard Reference Material (NIST SRM) 676] to each sample slurry. The corundum particles in each sample provided an internal 2θ standard for each XRD pattern. The measured patterns for the sludge samples analyzed with corundum confirmed that the observed peak shifts were due to slight misalignments of the mounted samples. Based on these results, 2θ corrections were estimated using JADE® XRD processing software and applied to the XRD patterns for the water-leached and HF-extract sludge samples.

Using a pipette, each slurry was transferred onto a circular-shaped platform (1-cm diameter) and placed on top of the post located on the base inside a disposable XRD specimen holder (Figure 2.3). This specimen holder was designed specifically for safe handling of dispersible powders containing highly radioactive or hazardous materials (Strachan et al. 2003). After allowing samples to air dry overnight, the holder was assembled and a piece of Kapton® film was placed between the cap and the retainer. The holder was sealed with wicking glue and removed from the fumehood.
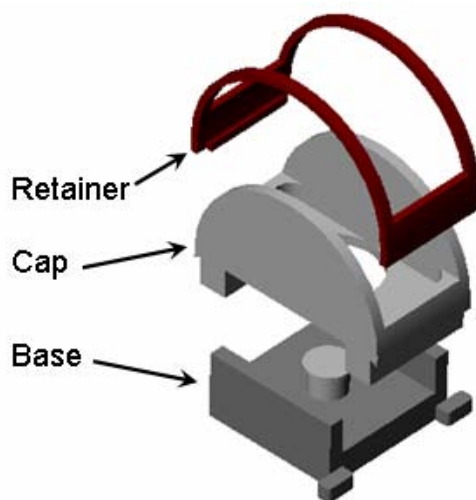
Figure 2.3. Exploded Schematic View of the XRD Sample Holder (Kapton® film not shown)

Each sample was analyzed using a Scintag XRD unit equipped with a Pelter thermoelectrically-cooled detector and a copper X-ray tube. The diffractometer was operated at 45 kV and 40 mA. Individual scans were obtained from 2 to 65° 2θ with a dwell time of 4 and 14 seconds. Scans were collected electronically and processed using the JADE® XRD pattern-processing software.

Krupka et al. (2004) prepared and analyzed by XRD a sample consisting of only a dry film of the collodion solution so that its contribution relative to the background signals of the XRD patterns for the sludge samples could be quantified. The resulting XRD pattern for the collodion solution film is shown in Figure 2.4. The most obvious feature of this diffraction pattern is the broad peak positioned between 10º and 30°2θ. The symmetry of this peak is characteristic of those resulting from the XRD of amorphous (noncrystalline) material. Although subtracting the collodion background from sludge XRD patterns allows for better phase matching, this process may eliminate minor reflections and inconspicuous features of a pattern. Therefore, each as-measured XRD pattern was examined before and after back-ground subtraction to ensure that the integrity of the pattern was maintained. For background subtraction,

the JADE® software provides the user with control over the selection of background-subtraction points. This process allows a better fit to 2θ regions under broad reflections, such as those resulting from amorphous materials. On average, 30 to 40 background points were selected from each XRD pattern, and a cubic-spline curve was then fit through each set of points. Adjustments to this curve were made by selecting additional background points in regions of a pattern that were difficult to fit. Once a well-matched curve was fitted to a pattern, the background was subtracted from each as-measured XRD pattern, resulting in a smooth tracing.
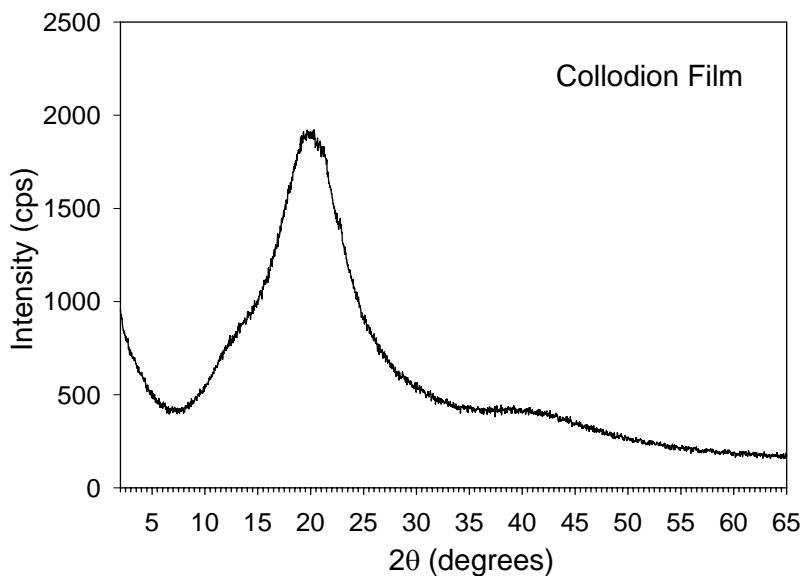
Figure 2.4. XRD Pattern for Collodion Film Measured in the Absence of any Sludge Material (from Krupka et al. 2004)

Identification of the mineral phases in the background-subtracted patterns was based on a comparison of the XRD patterns measured for the sludge samples with the mineral powder diffraction files (PDF™) published by the Joint Committee on Powder Diffraction Standards (JCPDS) International Center for Diffraction Data (ICDD). As a rule of thumb, a crystalline phase must be present at greater than 5 wt% of the total sample mass (greater than 1 wt% under optimum conditions) to be readily detected by XRD. In general, the measured peak intensities depend on several factors, including the combined mass of each crystalline phase in the sample. Due to the physical characteristics of these tank sludge samples such as high radioactivity, high dispersibility, and variable moisture content, the mass of tank sludge combined with the collodion solution for each XRD mount could not be controlled or easily determined. Dis-similarities in mineral segregation (settling) resulting from the different densities of minerals mixed with the collodion solution and associated effects on relative peak intensities also influence the overall pattern intensity. The combined effect of these factors could have some effect on the characteristic mineral peak intensities, which precluded quantitative comparisons of peak intensities for equivalent reflections in background-subtracted XRD patterns for different sludge samples.

2.4   SEM/EDS Analysis

The morphologies, sizes, surface textures, and compositions of phases present in the unleached (as-received), 1-month and 82-day water leached, and HF-extracted sludge samples 404 and 405 from tank C-106 were characterized by SEM/EDS. Two or three mounts were prepared of each sample to compensate for the possibility that one or more less-than-optimum mounts of a sample might occur, thus improving the likelihood of obtaining representative SEM images of each sample. The mounts used for SEM/EDS consisted of double-sided carbon tape attached to standard aluminum mounting stubs. For each mount, small aliquots of each sludge sample were placed on the exposed upper surface of the carbon tape using a micro spatula. Each mount was then coated with carbon using a vacuum sputter-coater to improve the conductivity of the samples and thus the quality of the SEM images and EDS signals.

A JEOL JSM-840 SEM was used for high-resolution imaging of micrometer/submicrometer-sized particles from the sludge samples. The SEM system is equipped with an Oxford Links ISIS 300 EDS that was used for qualitative element analysis. Operating conditions consisted of 10 to 20 keV for SEM imaging and 20 keV, 100 live seconds[a] for the EDS analyses. The EDS analyses of particles are limited to elements with atomic weights heavier than boron. Photomicrographs of high-resolution secondary electron (SE) images and backscattered electron (BSE) images were obtained as digital images and stored in electronic format. To help identify particles that contain elements with large atomic numbers, such as uranium, the SEM was typically operated in the BSE mode. Secondary electrons are low-energy electrons ejected from the probed specimen as a result of inelastic collisions with beam electrons, whereas backscattered electrons are primary electrons emitted as a result of elastic collisions. Backscattered electron emission intensity is a function of the element's atomic number ─ the larger the atomic number, the brighter the signal. Backscattered electron images are obtained in exactly the same way as secondary electron images.

The SEM micrographs included in the main body of this report (Section 3.6) were selected because they show typical morphologies, sizes, and surface textures of particles in the sludge subsample mounts. The entire area of each SEM mount was examined by SEM at low magnification (typically 50 to 100x) to identify those particles and surface features that were typical or unusual for the sample. During this examination, SEM micrographs were recorded at low magnification (e.g., 100x) for typically two to four areas of the mount to show a general perspective of the sizes, types, and distributions of particles that make up the SEM mount. Within these imaged regions, additional SEM micrographs were recorded of several particles at greater magnifications to provide a more detailed representation of the particles' characteristics, and selected points on these particles then analyzed by EDS. Depending on the perceived importance of such particles, regions on these particles were sometimes analyzed by SEM and EDS at even greater magnifications.

All of the SEM micrographs and EDS spectra determined for sludge samples 404 and 405 from tank C-106 are shown in Appendices B (unleached [as-received] sludge), C (1-month and 82-day water leached sludge), and D (HF Stage I extracted). The name of each digital image file, sample identification

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(a) Live time is when (real time less dead time) the EDS system is available to detect incoming X-ray photons. Dead time is the portion of the total analyzing time that is actually spent processing or measuring X-rays. While each X-ray pulse is being measured, the system cannot measure another X-ray that may enter the detector and is, therefore, said to be "dead."

number, and a size scale bar are given, respectively, at the bottom left, center, and right of each SEM micrograph in this report. Micrographs labeled BSE near the digital image file name indicate that the micrograph was collected with BSE. Areas outlined by a white or black dotted-line squares or particles indicated by arrows in a micrograph designate sample material that is imaged at higher magnification and typically shown in the next figure of the series for that subsample.

Areas labeled "eds" in SEM micrographs in this report indicate areas of particles for which EDS spectra were recorded. Compositions determined by EDS are qualitative and have large uncertainties resulting from alignment artifacts caused by the variable sample and detector configurations that exist when different particles are imaged by SEM.

After the studies and analyses of tank C-106 sludge had been completed and presented in draft form, the instrument hardware and software for the JEOL JSM-840 SEM used for the SEM/EDS analyses described above were upgraded to INCA Energy EDS System[a] to automate the collection of EDS spectra over multi-micrometer-sized areas of an SEM-imaged sample. This upgrade permits the mapping of the spatial distributions over user-selected areas and/or lines of the relative concentrations of any user-specified element detectable by EDS. The INCAEnergy EDS System was demonstrated by using it to generate element distribution maps for a limited number of imaged areas of existing SEM mounts of unleached, 82-day water leached, and HF-extracted sludge samples. A few sets of element distribution maps are shown and discussed in Section 3.6, and the remainder of the generated element maps are shown in Appendix E.

2.5   Tier 1 Tests

Sludge samples were analyzed in a tiered approach similar to the one developed to investigate contaminant fate and transport issues associated with past single-shell tank leaks in the vadose zone. Such an approach allows for initial (Tier 1) screening of samples using relatively inexpensive analytical techniques. This is followed by an analysis of the data to determine the need for further testing (Tier 2). The Tier 1 tests are described in this section, and the Tier 2 tests are described in Sections 2.6 through 2.8. In addition to the analysis of the sludge samples, the sample of the liquid collected near the termination of retrieval activities was analyzed using many of the Tier 1 methods described in this section.

All laboratory activities were conducted in accordance with the requirements of Title 10, Code of Federal Regulations, Part 830.120 "Quality Assurance" and the *Hanford Analytical Services Quality Assurance Requirements Document* (HASQARD, DOE 1998). These requirements were implemented using PNNL's internal quality assurance (QA) plan.[b] PNNL's QA Plan is based on the requirements of U.S. DOE Order 414.1A , the HASQARD, relevant elements of NQA-1, as well as recognized industry standards (e.g., EPA, ASTM, American National Standards Institute).

(a)   Oxford Instruments, Concord, Massachusetts.
(b)   PNNL. 2001. *Conducting Analytical Work in Support of Regulatory Pro*grams. Internal unpublished procedure, Pacific Northwest National Laboratory, Richland, Washington.

2.5.1   Moisture Content

The moisture contents of the tank waste samples were measured to calculate dry weight concentrations for constituents in the waste. Dry weight concentrations provide a consistent measurement unit for comparison purposes that eliminates the effect of variable water content on sample concentrations.

Gravimetric water content of the waste material was determined using ASTM procedure D2216-98, *Standard Test Method for Laboratory Determination of Water (Moisture) Content of Soil and Rock by Mass* (ASTM 1998) with the following minor exceptions: 1) the volume of sample recommended was decreased due to radiological concerns and 2) the sample was dried at a lower oven temperature, 105°C, for a longer period of time to prevent dehydration of the solids.

Sludge samples were placed in tarred containers, weighed, and dried in an oven until a constant weight was achieved, usually requiring 24 to 48 hours. The container was then removed from the oven, sealed, cooled, and weighed. All weighings were performed using a calibrated balance. The gravimetric water content is computed as the percentage change in soil weight before and after oven drying (i.e., [{wet weight - dry weight}/dry weight]).

2.5.2   Water Extracts

Water-soluble inorganic constituents were determined using a DDI water extraction method. The extract was prepared by adding 30 mL of DDI water to 0.2 to 0.4 g of the residual sludge sample contained in a 50 mL polypropylene centrifuge tube. The centrifuge tube was sealed and briefly shaken by hand, and then placed on a mechanical orbital shaker for three time periods: 1 day, 2 weeks, and 1 month. After shaking for the predetermined time, the tube was placed in a centrifuge and spun at 4,000 rpm for 20 minutes. The supernatant was carefully decanted, filtered through 0.45-μm pore size membrane, and analyzed. For this test, the first stage of the Periodic Replenishment test (Section 2.5.3) was used as the 1-day leach sample. The 2-week and 1-month leach tests used the same sludge sample. After 2 weeks, a 5 mL aliquot was removed and analyzed. The remaining 25 mL was allowed to shake for 2 more weeks and represents the 1-month leach sample. The 2-week and 1-month leach sample results were corrected for a 30 mL final volume. More details of the test method can be found in ASTM Procedure D3987-85, *Standard Test Method for Shake Extraction of Solid Waste with Water* (ASTM 1999).

2.5.3   Periodic Replenishment Tests

Periodic replenishment tests were conducted on samples of residual sludge from tank C-106. In this test, the water-leachate solution was periodically removed and replaced with an equal volume of fresh solution. This test was conducted to evaluate whether the solution concentration might be limited by the solubility of one or more solid phases. For these tests, the samples were contacted with DDI water a total of five times. The contact periods ranged from 2 to 3 days, the length of time between replenishment of water-leachate solutions. The sludge samples were prepared and handled in the same manner as the single-contact water extracts for each repetitive step. After the fifth stage, samples were replenished with fresh solutions and placed back on the shaker for evaluation of long-term solubilities. The contact time for one of these long-term samples was 43 days (Stage 6A). For the other long-term test the contact time

was 82 days (Stage 6B). After these long-term tests, the samples were centrifuged and the supernatant carefully decanted and filtered through 0.45-µm pore size membranes prior to analysis for the same constituents as the shorter-term tests.

2.5.4   pH

The pH values of the solutions were measured using a solid-state pH electrode and a pH meter calibrated with buffers bracketing the expected range. This measurement is similar to EPA Method 9040B (EPA 1995).

2.5.5   Anion Analysis

Anion analysis was performed using an ion chromatograph. Fluoride, acetate, formate, chloride, nitrite, bromide, nitrate, carbonate, sulfate, oxalate, and phosphate were separated on a Dionex AS17 column with a gradient elution technique from 1 mM to 35 mM NaOH and measured using a conductivity detector. This methodology is similar to EPA Method 9056A (EPA 1994b) with the exception of using gradient elution with NaOH.

2.5.6   Cations and Trace Metals

Major cation analysis (including Al, Ca, Fe, K, Mg, Mn, Na, and Si) was performed by ICP-OES EPA Method 6010B (EPA 1996b). Trace metals analysis (including Ag, As, Cd, Cr, Mo, Pb, Se, ^{99}Tc, ^{238}U, ^{239}Pu, ^{241}Am, and ^{237}Np) was performed by ICP-MS. This method is similar to EPA Method 6020 (EPA 1994a). For both ICP-OES and ICP-MS, high-purity calibration standards were used to generate calibration curves and to verify continuing calibration during the analysis. Multiple dilutions of selected samples (ranging from 3x to 100x) were made and analyzed to investigate and correct for matrix interferences.

ICP-MS data is reported as total element concentration in terms of the specific isotope measured. The instrument software converts the concentration of an isotope of an element to the total concentration of the element based on the distribution of isotopes in the natural environment. For example, the total Cr concentration is reported from the raw count rates for both ^{52}Cr and ^{53}Cr based on taking the raw counts and dividing by the fraction of ^{52}Cr and ^{53}Cr found in nature to yield estimates of total Cr in the sample.

Standard ICP-MS operating software does not allow the direct input of the various element isotope concentrations as percent relative abundance. Under most circumstances, such as when only naturally occurring isotopes are present, the lack of correction for percent abundances creates no analytical quantification issues. However, care must be taken to interpret data under circumstances where the products of nuclear fission could be present. In this case, the greatest impact to the quantification of element data using standard calibration methods would be an overestimation of the concentration of an element in the presence of a fission product. For instance, Mo has seven natural isotopes with relative abundances ranging from 9.25 to 24.13%. When the ICP-MS is calibrated, the counts per second measured at a specific mass are assigned a concentration by the operator. Unfortunately, the current manufacturer-supplied software does not account for the relative percent abundance of the seven Mo isotopes, nor will it allow the operator to assign different standard concentrations for the isotopes. In other words, for a 1 ng/mL calibration standard, 1 ng/mL must be assigned as the concentration for all

seven Mo isotopes rather than 1 ng/mL multiplied by the percent abundance of the natural isotope. As mentioned in the preceding paragraph, this is not an issue when dealing only with natural systems, which will always have the relative abundance breakdown of 9.25 to 24.13% for the seven Mo isotopes. However, if ^{95}Mo, ^{97}Mo, ^{98}Mo, or ^{100}Mo fission products are present in the samples being analyzed, the actual fission product contribution to the total isotope concentration will be overestimated by a factor of 100 divided by the percent relative abundance of the natural isotope.

For future studies of tank sludge, the new Perkin Elmer Elan DRC II ICP-MS's software will be available. The Elan software allows the operator to assign different concentration values for each isotope of a respective element. This feature will enable user calibration based on the percent relative abundance of the natural isotopes. Measurement in this manner will permit the determination of individual masses without corrections for natural abundance. As the new ICP-MS is brought online, this technique will be tested with the objective that all subsequent calibration will be the direct quantification of total isotope concentrations. This will aid in the interpretation and possible quantification of fission products present in tank sludge.

2.5.7   Alkalinity

The sample alkalinity was measured by standard titration. A volume of standardized sulfuric acid (H_2SO_4) was added to the sample to an endpoint of pH 8.3 and then an endpoint of pH 4.5. The volume of H_2SO_4 needed to achieve each endpoint is used to calculate the phenolphthalein ($OH^- + CO_3^{2-}$) and total ($OH^- + HCO_3^- + CO_3^{2-}$) alkalinity as calcium carbonate ($CaCO_3$). The alkalinity procedure is similar to Standard Method 2320 B (Clesceri et al. 1998).

2.5.8 ^{129}I Extraction and Analysis

From a long-term risk standpoint, ^{129}I is a key potential contaminant in residual Hanford tank waste. For this reason, its presence in the waste material and mobility in infiltrating water is of interest. Although ^{129}I is generally considered mobile as a dissolved constituent in water, small partition coefficients (0.2 to 1 mL/g) are typically calculated when its uptake is measured on Hanford sediments (Cantrell et al. 2003; Um et al. 2004). Therefore, it is imperative to identify an extraction method that will enable quantitative measurement of total iodine in solid samples such as tank waste. Previous research (Brown et al. 2004) has shown the potential applicability of water leaches and KOH:KNO$_3$ water fusions for the removal of iodide from solid samples spiked with ^{129}I. The results from Brown et al. (2004) have led to the modification of the accepted PNNL internal procedure, to determine the ^{129}I concentration in sludge solids. In the case of ^{129}I, the KOH-KNO$_3$ fusion-dissolution procedure (Section 2.2) was modified (steps 10 through 14 were omitted) to prevent volatilization of iodine. In this modified procedure, 0.3 g of the tank waste sludge material was mixed with 10 mL of a 20% KOH and 2% KNO$_3$ solution as a fluxing agent in a Zr crucible. Several process spike samples, which were used to determine extraction efficiencies, were prepared by adding 0.05 µg ^{129}I to a crucible containing KOH-KNO$_3$ solution (blank spike) or sample and KOH-KNO$_3$ solution (matrix spike). The crucible was then placed in a 95ºC oven and allowed to evaporate to dryness, after which it was covered and transferred to a muffle furnace preheated to 550ºC. Fusion was accomplished by heating the sample-flux mixture for 60 minutes at 550ºC. After 60 minutes, the crucible was removed from the furnace and allowed to cool to

ambient room temperature. The fused solid was then dissolved in DDI water. The resulting solution was transferred to a 50-mL centrifuge tube. The crucible was then triple-rinsed with DDI water, and these wash solutions were also added to the centrifuge tube. Up to 25 mL of 1:1 concentrated sulfuric acid:1 M sodium bisulfite was added to the centrifuge tube to dissolve any remaining solids that may have transferred from the crucible during solution transfer. The contents of the centrifuge were swirled to facilitate mixing/dissolution. Once dissolution was complete, the resulting solution was diluted to a total volume of 50 mL with DDI water (the final solution volume in the centrifuge tube was determined gravimetrically and corrected for solution density).

Table 2.3 lists the digestion factors (wet solid-to-solution ratios) for the samples of tank C-106 sludge solids used for the modified KOH-KNO$_3$ water fusion treatments to measure ^{129}I. These factors were calculated from the wet weight of sludge material divided by the volume of extracting solution. The digestion factors were then multiplied by the percent solids, as determined from moisture content analysis, to convert to a dry weight basis. The fused sludge samples, as well as the samples from the one-day, single-contact water leach tests (Section 2.5.2), were analyzed for dissolved ^{129}I concentrations using either a VG Elemental PQS ICP-MS or a Perkin Elmer Elan DRC II ICP-MS. Spectrasol CFA-C from Spectrasol, Inc. (Warwick, NY) was diluted in DDI water (18 MΩ-cm) to create a 1% working solution.

Table 2.3. Digestion Factors for Samples of C-106 Sludge Solids Used for the Modified KOH-KNO$_3$ Water Fusion Treatment to Measure ^{129}I

Treatment	Sample Number	Dry Weight Corrected Digestion Factor (g/L)
KOH-KNO$_3$ fusion	404 C-106 (Primary)	3.888
	404 C-106 DUP[a] (Primary)	3.414
	405 C-106 (Duplicate)	4.834
	405 C-106 DUP[a] (Duplicate)	5.406
Matrix spike	405 C-106 (Duplicate) MS	6.627
(a) DUP = Duplicate sample.		

Calibration standards were prepared by diluting a 1 mg/L ^{129}I certified stock standard (NIST, Gaithersburg, MD) into appropriate volumes of the 1% Spectrasol CFA-C solution containing 5 ng/mL ^{121}Sb as the internal standard. Internal standards are chosen based on their proximity (atomic mass) to an element of interest and are used to verify instrument performance and correct for instrument drift. An independent calibration check standard was prepared from a 1 mg/L ^{129}I certified stock standard (Amersham, Piscataway, NJ) in 1% Spectrasol CFA-C. One percent Spectrasol CFA-C was used to prepare instrument blanks and was used as the rinse solution throughout the run.

2.5.9 Radioanalysis

In addition to the radionuclides listed in Section 2.5.6 that were analyzed in solution by ICP-MS, short-lived radionuclides were analyzed by conventional counting methods as described in the following sections.

2.5.9.1 Gamma Energy Analysis

All samples for gamma energy analysis (GEA) were analyzed using 60% efficient intrinsic-germanium gamma detectors. All germanium counters were efficiency calibrated for distinct geometries

using mixed gamma standards traceable to the NIST. Direct solids, acid extracts, and water extracts were analyzed for gamma energy. Spectral analysis was conducted using libraries containing most mixed-fission products, activation products, and natural decay products. Control samples were run throughout the analysis to ensure correct operation of the detectors. The controls contained isotopes with photo peaks spanning the full detector range and were monitored for peak position, counting rate, and full-width half-maximum. Details are found in PNNL internal procedure RRL-001.[a.]

2.5.9.2 ^{90}Sr Analyses

Aliquots of filtered acid extracts, fusions, and water extracts were diluted in 8 M HNO_3 and submitted for strontium separation and analysis by internal PNNL procedure PNL-RRL-003.2.[b] A 0.1 to 5 mL aliquot of sample was spiked with ^{85}Sr tracer and passed through a SrSpec® column (Eichrom Technologies, Chicago) to capture Sr. The columns were washed with 10 column volumes (20 mL) of 8 M HNO_3. The strontium was eluted from the SrSpec column into glass liquid scintillation vials using 15 mL of deionized water. The vials were placed under a heat lamp overnight to evaporate the water to dryness. A 15 mL Optifluor® scintillation cocktail was added to each vial. Gamma spectroscopy was used to determine the chemical yield from the added ^{85}Sr tracer. The samples were then analyzed by liquid scintillation counting (LSC) to determine the amount of ^{90}Sr originally present in the sludge sample. A matrix spike, a blank spike, a duplicate, and blanks were run with each sample set to determine the efficiency of the separation procedure as well as the purity of reagents.

2.5.9.3 Gross Alpha and Beta Analysis

Gross alpha and beta measurement were made on both the water and acid extracts. For each extract, 0.100 mL sample volume was placed in a 20-mL liquid scintillation vial containing 15 mL of scintillation cocktail.[c] The samples were then mixed and counted on a Wallace model 1415 liquid scintillation counter as prescribed in internal PNNL procedure AGG-RRL-002.[d]

2.6 Selective Extraction Tests

2.6.1 Phase 1

In order to identify the relative amount of potentially mobile contaminants (^{99}Tc, ^{238}U, and Cr) that may be sequestered within the three metal oxyhydroxide solids (Mn, Al, and Fe), the following sequence of selective extractions was conducted. The standard 0.3 g of residual sludge per 30 mL of solution was

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(a) RRL-001, *Gamma Energy Analysis, Operation, and Instrument Verification using Genie2000 Support Software*, unpublished PNNL Technical Procedure, Pacific Northwest National Laboratory, Richland, Washington.

(b) PNL-RRL-003.2. 2000. *Tc99 and Sr90 Analysis using Eichrom TEVA-spec and Sr-spec resin.* Technical Procedure, Pacific Northwest National Laboratory, Richland, Washington.

(c) The scintillation cocktail used is Packard Optifluor, which is based on the high flash-point solvent LAB (Linear Alkylbenzene) (http://las.perkinelmer.com/catalog/Product.aspx?ProductId=6013199).

(d) AGG-RRL-002, *Liquid Scintillation Counting and Instrument Verification using the 1400 DSA^{TM} Support Software*, unpublished PNNL Technical Procedure, Pacific Northwest National Laboratory, Richland, Washington.

changed to 0.1 g per 30 mL of solution for this set of extractions. For the manganese oxyhydroxide extractions, a much higher solution-to-solid ratio was required (2000:1). This series of selective extractions was done in duplicate for both samples 404 and 405. The entire sequence of extractions is shown schematically in Figure 2.5.

Selective Extraction Stage 1. 30 mL of DDI water was added to between 0.1 to 0.14 g of sample 404 or 405 sludge material. The samples were placed on a shaker table for 24 hours, then removed and centrifuged. The supernatant was removed and stored for later analysis. This step was performed to remove readily soluble components (i.e., salts) from the sludge.

Selective Extraction Stage 2. 200 mL of 0.1 M NH_2OH-HCl (pH~3.6) was added to each sample and placed on the shaker table for 2 hours. The samples were removed and centrifuged. The supernatant was then removed and stored for later analysis. This step removes the Mn oxyhydroxides and associated contaminants from the sludge.

Selective Extraction Stage 3 and 4. Two successive extraction steps consisting of a 0.01 M HF acid/0.01 M sodium fluoride buffer (pH approximately 3.2) were performed on the solids. The contact time on the shaker table was 2 hours. The samples were centrifuged and the supernatant removed after each stage and stored for analysis. These steps were performed to remove Al oxyhydroxides and associated contaminants.

Selective Extraction Stage 5. The remaining solids were transferred from the centrifuge cones to 50 mL Griffin beakers and contacted with an 8N HNO_3 solution. The solids and solutions were heated to 95°C. Over the subsequent 2 hours, two 1-mL aliquots of concentrated HNO_3 were added to the slurries. The samples were allowed to cool and brought to a 30-mL final volume. The solution was filtered through a 0.45-µm syringe filter and the filtrate stored for later analysis.

The solutions from each stage were analyzed for ^{99}Tc, ^{238}U, Cr, Mn, Al, and Fe. The pH was also measured on the extract solutions prior to and following each extraction. Extra samples were run so that the solids after the hydroxyl amine hydrochloride and the second fluoride buffer stages could be saved for analysis by XRD and SEM/EDS.

2.6.2   Phase 2

A second series of extractions was conducted to compare the HF/NaF buffer solution extraction with a hot concentrated NaOH extraction method for selective removal of Al oxyhydroxides. This set of extractions was conducted on both samples 404 and 405 in duplicate. Phase 2a was the HF/NaF buffer solution extraction, and the Phase 2b was the NaOH extraction method.

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┌─────────────────────────┐
│   0.1 g C-106 Sludge    │
└─────────────────────────┘
            │
            ▼
┌─────────────────────────┐
│  30 mL Deionized Water  │ ────────▶  Remove readily soluble salts
└─────────────────────────┘
            │
            │                          ┌──────────────────────────────┐
            ├──────────────────────────▶│ Leachates analyzed for $^{99}$Tc, │
            │                          │ U, Cr, Fe, Al, Mn, and pH.   │
            ▼                          └──────────────────────────────┘
┌─────────────────────────┐
│      200 mL 0.1 M       │ ────────▶  Remove Mn
│   NH$_2$OH-HCl pH 3.6      │            oxyhydroxides.
└─────────────────────────┘
            │                          ┌──────────────────────────────┐
            ├──────────────────────────▶│ Leachates analyzed for $^{99}$Tc, │
            │                          │ $^{238}$U, Cr, Fe, Al, Mn, and pH. │
            ▼                          └──────────────────────────────┘
┌─────────────────────────┐
│         30 mL           │
│  0.01 M HF/0.01 M NaF   │ ────────▶  Remove Al
│   Buffer pH 3.2 (Twice) │            oxyhydroxides.
└─────────────────────────┘
            │
            ▼
┌─────────────────────────┐
│    Hot Concentrated     │ ────────▶  Remove recalcitrant Fe and Al
│         HNO$_3$           │            oxyhydroxides.
└─────────────────────────┘
            │                          ┌──────────────────────────────┐
            └──────────────────────────▶│ Leachates analyzed for $^{99}$Tc, │
                                       │ $^{238}$U, Cr, Fe, Al, Mn, and pH. │
                                       └──────────────────────────────┘
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Figure 2.5. Phase 1 Selective Extraction Sequence Used to Determine Phase Associations of ^{99}Tc, ^{238}U, and Cr in Tank C-106 Sludge

2.6.2.1 Phase 2a

Selective Extraction Stage 1. 30 mL DDI water was added to 0.3 to 0.33 g of sludge samples 404 and 405. The samples were placed on a shaker table and extracted for 24 hours. The samples were centrifuged, and the supernatant was discarded. This step removed the readily soluble constituents.

Selective Extraction Stage 2 and 3. Two successive extractions consisting of 0.01 M HF acid/0.01 M NaF buffer (pH approximately 3.2) were performed on the solids. The contact time on the shaker table was 2 hours for each extraction. The slurry was then centrifuged and the supernatant removed after each stage and stored for analysis. These steps were intended to selectively remove Al oxyhydroxide.

Each of the extract solutions was analyzed for ^{99}Tc, ^{238}U, Mn, Cr, Al, and Fe.

2.6.2.2 Phase 2b

Selective Extraction Stage 1. 30 mL DDI water was added to 0.1 to 0.12 g of sludge samples 404 or 405 to remove readily soluble constituents. The samples were placed on a shaker table and extracted for 24 hours. The samples were centrifuged, and the supernatant was discarded. This step removed the readily soluble constituents.

Selective Extraction Stage 2. The sludge materials were rinsed into 6 mL distillation vessels with DDI water and placed in a drying oven. The solids were allowed to come to dryness and 5 ml of 3 M NaOH was added to each vessel. Cooling columns were attached and the sludge samples were heated to 95°C for 5 hours. The resulting solution was removed and brought to a final volume of 10 mL and stored for later analysis.

Selective Extraction Stage 3. Five mL of fresh 3 M NaOH was added to each vessel and heated to 95°C for 11 days. The solution was removed and brought to a final volume of 10 mL and stored for later analysis.

The solutions from each stage were analyzed for ^{238}U, Cr, Mn, Al, and Fe.

3.0   Laboratory Results

This section provides the results of the tests conducted on sludge and liquid samples from tank C-106. The discussion begins with sludge composition by fusion and acid digestions in Section 3.1. Section 3.2 presents ^{129}I extraction and measurement results, and Section 3.3 provides the results of the analysis of the liquid sample collected during final retrieval operations. Section 3.4 contains the results of batch water-leaching tests, including single-contact and periodic replenishment methods. X-ray diffraction and SEM/EDS analyses are described in Sections 3.5 and 3.6, while Section 3.7 discusses selective extractions.

3.1   Sludge Composition

An important component of calculating contaminant release rates is an accurate measurement of the total concentration of contaminants in the source material. As described in Section 2.2, the total metal and radionuclide concentrations of the sludge samples were measured using two methods (fusion and acid digestions). The results of these analyses are described in this section. The anionic (nonmetal) composition of the sludge was estimated by water extraction as part of the Tier 1 analyses (Section 2.5.2). The results are discussed in Section 3.4.1.

An insoluble solid representing a very small fraction of the original mass of the sludge samples was observed at the end of the fusion preparations and acid digestions for both samples 404 and 405. The residuals of both preparations consisted of very fine rust-colored particles. Because the insoluble fractions are not a significant amount of the total masses of the sludge, their presence is not expected to have a large impact on the measured total element compositions of the sludge.

Concentrations listed in parentheses in the tables are defined as less than the estimated quantitation limit (EQL) but greater than a zero instrument signal. These values are reported for informational purposes only. They may reflect actual concentrations that are real but have larger associated uncertainties than values above the EQL or they may reflect values that were calculated from the instrument's background signal and are not representative of actual sludge composition. The EQL of an element is determined by analyzing a suite of continuing calibration verification (CCV) standards at the beginning and end of each analytical run. The lowest CCV standard that is within ±10% of its certified value is multiplied by the dilution factor for the sample to determine the EQL for the element for the particular analytical run. The EQL may vary with each analysis depending on sample matrix, dilution factors, and instrument performance.

Concentrations listed as less than (<) values in the tables refer to instrument measurements that are less than zero. In these instances, the reported analyte concentration is assigned a value of "<EQL" using the EQL value appropriate for that particular analyte and set of analytical conditions.

The following discussion of element concentrations of the sludge is organized in terms of the analytical method used to measure concentrations in the solution extract. These methods were ICP-OES, ICP-MS, GEA, wet chemical separations and ICP-MS (actinides), wet chemical separations and liquid scintillation (^{90}Sr), and gross alpha/beta analysis. For the tables, the solution concentrations have been converted from a per-liter basis to a dry sludge mass basis. Each table provides results from the fusion and acid digestion methods. Table 3.1 lists the moisture contents (relative to total sludge mass) of the C-106 sludge samples used for the fusion and acid digestions. These values are used with the digestion factors (Table 2.2) to convert the solution analyses of the extracts from the treatments to dry weight solid concentrations. The moisture contents of the sludge samples ranged from 42.2 to 58.9%, suggesting that the samples were completely water saturated.

Tables 3.2 through 3.4 contain the results of ICP-OES analyses of the sludges. Because K-containing compounds were used as the fluxing agent for the fusion technique, K concentrations are not reported in Table 3.3 for the fusion analysis. The metals detected above their respective EQLs were Al, Mn, Na, Fe, Ca, Pb, Mg, Ni, and Ba. Figure 3.1 shows the concentrations of the major metals in the sludge based on fusion analyses of the primary and duplicate samples. The elements present in highest concentrations in the samples with their maximum values in units of μg/g are Al (1.49 x 10^5), Mn (1.23 x 10^5), Na (6.17 x 10^4), Fe (4.7 x 10^4), and Ca (3.95 x 10^4). For the metals, there is some variability in concentrations between the fusion method and the EPA acid digestion technique. The fusion method gave higher concentrations of the major metals by 10 to 40% compared to the concentrations measured by acid digestion.

Table 3.1. Moisture Contents of Tank C-106 Sludge Samples 404 and 405 [(wet wt – dry wt)/dry wt]

Sample Number	Moisture Content
404	58.86%
404 DUP	56.11%
405	42.19%
405 DUP	44.88%
DUP = Duplicate sample.	

Table 3.2. Concentrations of Elements Measured by ICP-OES per Gram of Dry Sludge

Sample Number	Al	As	B	Ba	Be	Bi	Ca	Cd	Co	Cr
	-- µg/g --									
KOH-KNO$_3$ Fusions										
404	1.32E+05	(6.03E+02)	(7.15E+02)	1.49E+03	(6.47E+01)	(3.5E+02)	3.75E+04	(2.74E+02)	(1.0E+02)	(8.61E+02)
404 DUP	1.49E+05	<6.76E+03	(3.26E+02)	(6.47E+02)	(5.77E+01)	(1.9E+02)	3.95E+04	(2.33E+02)	<1.35E+03	(8.20E+02)
405	1.23E+05	<4.93E+03	(2.64E+02)	(5.87E+02)	(3.87E+01)	<4.93E+03	3.75E+04	(2.41E+02)	<9.86E+02	(9.77E+02)
405 DUP	1.23E+05	<5.72E+03	(2.22E+02)	1.39E+03	(4.14E+01)	<5.72E+03	3.84E+04	(2.41E+02)	<1.14E+03	(9.27E+02)
404 Avg	1.40E+05	<3.68E+03	(5.20E+02)	1.07E+03	(6.12E+01)	(2.7E+02)	3.85E+04	(2.54E+02)	<7.27E+02	(8.41E+02)
405 Avg	1.23E+05	<5.33E+03	(2.43E+02)	9.89E+02	(4.01E+01)	<5.33E+03	3.79E+04	(2.41E+02)	<1.07E+03	(9.52E+02)
EPA Acid Digestion										
404	8.21E+04	(1.03E+02)	(1.76E+02)	1.03E+03	(1.81E+01)	<3.09E+03	2.73E+04	(2.35E+02)	<6.17E+02	(7.55E+02)
404 DUP	7.80E+04	<3.64E+03	(9.50E+01)	9.22E+02	(2.28E+01)	<3.64E+03	1.07E+05	(2.01E+02)	<7.27E+02	(6.47E+02)
405	8.84E+04	<2.67E+03	(6.47E+01)	9.06E+02	(1.52E+01)	<2.67E+03	2.49E+04	(2.41E+02)	<5.34E+02	7.36E+02
405 DUP	7.83E+04	<3.21E+03	(1.66E+02)	7.97E+02	(1.64E+01)	<3.21E+03	2.69E+04	(2.41E+02)	<6.42E+02	(7.72E+02)
404 Avg	8.01E+04	<1.87E+03	(1.35E+02)	9.76E+02	(2.05E+01)	<3.36E+03	6.71E+04	(2.18E+02)	<6.72E+02	(7.01E+02)
405 Avg	8.33E+04	<2.94E+03	(1.15E+02)	8.52E+02	(1.58E+01)	<2.94E+03	2.59E+04	(2.41E+02)	<5.88E+02	(7.54E+02)

Avg = Average.
DUP = Duplicate.
EPA = U.S. Environmental Protection Agency.
ICP-OES = Inductively coupled plasma-optical emission spectroscopy.
KNO$_3$ = Potassium nitrate.
KOH = Potassium hydroxide.

Table 3.3. Concentrations of Elements Measured by ICP-OES per Gram of Dry Sludge

Sample Number	Cu	Fe	K	Li	Mg	Mn	Mo	Ni	P	Pb
	--- µg/g ---									
KOH-KNO$_3$ Fusions										
404	(8.51E+02)	4.7E+04	NR	<1.20E+05	3.09E+03	1.20E+05	(3.46E+01)	1.92E+03	(7.82E+03)	5.01E+03
404 DUP	(5.96E+02)	4.0E+04	NR	<1.35E+05	(2.25E+03)	1.07E+05	(2.14E+02)	2.17E+03	(7.22E+03)	4.61E+03
405	(6.13E+02)	4.4E+04	NR	<9.86E+04	2.21E+03	1.21E+05	<9.86E+02	1.44E+03	(7.85E+03)	4.65E+03
405 DUP	(6.92E+02)	4.5E+04	NR	<1.14E+05	2.38E+03	1.23E+05	(7.43E+01)	1.32E+03	(7.88E+03)	5.09E+03
404 Avg	(7.23E+02)	4.33E+04	NR	<1.27E+05	2.67E+03	1.14E+05	(1.24E+02)	2.04E+03	(7.52E+03)	4.81E+03
405 Avg	(6.53E+02)	4.43E+04	NR	<1.07E+05	2.30E+03	1.22E+05	<5.30E+02	1.38E+03	(7.86E+03)	4.87E+03
EPA Acid Digestion										
404	(5.39E+02)	3.80E+04	(7.98E+02)	<6.17E+04	1.84E+03	1.11E+05	<6.17E+02	5.57E+03	(6.78E+03)	5.0E+03
404 DUP	(5.25E+02)	3.41E+04	(3.90E+03)	<7.27E+04	7.25E+03	1.00E+05	(3.63E+02)	5.03E+03	1.43E+04	4.60E+03
405	(4.58E+02)	3.79E+04	<1.33E+04	<5.34E+04	1.75E+03	1.09E+05	<5.34E+02	5.51E+03	(6.11E+03)	4.79E+03
405 DUP	(4.3E+02)	3.67E+04	<1.61E+04	<6.42E+04	1.81E+03	1.12E+05	<6.42E+02	5.38E+03	(5.82E+03)	4.91E+03
404 Avg	(5.32E+02)	3.61E+04	2.35E+03	<6.72E+04	4.54E+03	1.06E+05	(4.90E+02)	5.30E+03	(1.05E+04)	4.78E+03
405 Avg	(4.44E+02)	3.73E+04	1.47E+04	<5.88E+04	1.78E+03	1.11E+05	<5.88E+02	5.45E+03	(5.96E+03)	4.85E+03

Avg = Average.
DUP = Duplicate sample.
EPA = U.S. Environmental Protection Agency.
ICP-OES = Inductively coupled plasma-optical emission spectroscopy.
KNO$_3$ = Potassium nitrate.
KOH = Potassium hydroxide.
NR = Not Reported – K is a major component of the fluxing agent using in the fusion analysis.

Table 3.4. Concentrations of Elements Measured by ICP-OES per Gram of Dry Sludge

Sample Number	Se	Sr	Tl	V	Zn	Na	Si	S	Ti	Zr
	— µg/g									
KOH-KNO$_3$ Fusions										
404	<1.20E+04	(2.65E+02)	<1.20E+07	(3.49E+02)	(2.38E+03)	6.13E+04	(1.93E+04)	<2.39E+05	(1.82E+02)	(7.39E+01)
404 DUP	<1.35E+04	(2.10E+02)	<1.35E+07	(4.22E+02)	(3.52E+03)	6.17E+04	(1.86E+04)	<2.70E+05	(1.17E+02)	(7.28E+01)
405	<9.86E+03	(2.80E+02)	<9.86E+06	(2.45E+02)	(2.20E+03)	5.77E+04	(1.89E+04)	<1.97E+05	(2.02E+02)	(4.24E+01)
405 DUP	<1.14E+04	(2.70E+02)	<1.14E+07	(2.32E+02)	(2.35E+03)	6.09E+04	(1.95E+04)	<2.29E+05	(1.42E+02)	(2.32E+01)
404 Avg	<1.27E+04	(2.38E+02)	<1.27E+07	(3.85E+02)	(2.95E+03)	6.15E+04	(1.90E+04)	<2.55E+05	(1.49E+02)	(7.33E+01)
405 Avg	<1.07E+04	(2.75E+02)	<1.07E+07	(2.39E+02)	(2.27E+03)	5.93E+04	(1.92E+04)	<2.13E+05	(1.72E+02)	(3.28E+01)
EPA Acid Digestion										
404	<6.17E+03	(4.11E+02)	<6.17E+06	(1.55E+02)	(1.52E+03)	4.39E+04	(2.04E+03)	<1.23E+05	(4.71E+01)	(1.13E+02)
404 DUP	<7.27E+03	7.75E+02	<7.27E+06	(1.81E+02)	5.08E+04	4.96E+04	(1.28E+04)	(4.44E+04)	(4.89E+01)	(5.27E+01)
405	<5.34E+03	(3.90E+02)	<5.34E+06	(1.18E+02)	(1.49E+03)	5.00E+04	(1.96E+03)	(8.6E+01)	(5.50E+01)	(5.66E+01)
405 DUP	<6.42E+03	(3.97E+02)	<6.42E+06	(2.10E+02)	(1.65E+03)	4.34E+04	(2.78E+03)	<1.28E+05	(4.33E+01)	<6.42E+02
404 Avg	<6.72E+03	(5.93E+02)	<6.72E+06	(1.68E+02)	(2.62E+04)	4.67E+04	(7.42E+03)	<8.39E+04	(4.80E+01)	(8.26E+01)
405 Avg	<5.88E+03	(3.94E+02)	<5.88E+06	(1.64E+02)	(1.57E+03)	4.67E+04	(2.37E+03)	<6.43E+04	(4.92E+01)	(3.49E+02)

Avg = Average.
DUP = Duplicate sample.
EPA = U.S. Environmental Protection Agency.
ICP-OES = Inductively coupled plasma-optical emission spectroscopy.
KNO$_3$ = Potassium nitrate.
KOH = Potassium hydroxide.
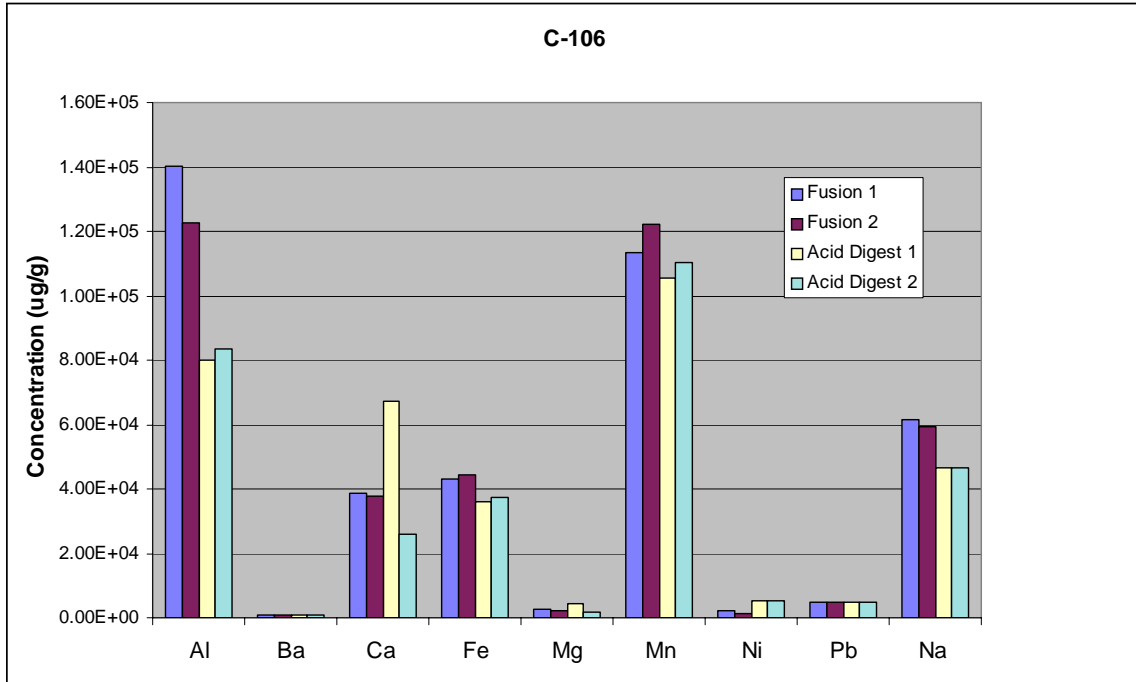
Figure 3.1. Metals Concentrations in Tank C-106 Sludge

The element concentrations in Tables 3.5 and 3.6 were derived from the ICP-MS analyses, which can obtain lower detection limits than ICP-OES for some metals and allow for measurement of trace metals. The elements listed in Tables 3.5 and 3.6 with concentrations greater than their EQLs are Pb, Cr, Cd, and Ru. The average Pb concentrations (based on ^{208}Pb and ^{208}Pb) in the sludge measured by the fusion method were 4,560 µg/g and by EPA acid digestion 4,610 µg/g, which are within 10% of the ICP-OES respective values of 4,840 µg/g and 4,820 µg/g. The average Cr concentrations (based on ^{52}Cr and ^{53}Cr) in the C-106 sludge measured by the fusion method were 699 µg/g and by EPA acid digestion 624 µg/g. The average Cd concentrations (based on ^{111}Cd and ^{114}Cd) measured by the fusion method were 290 µg/g and by EPA acid digestion 295 µg/g. The average Ru concentrations (based on ^{101}Ru, ^{102}Ru, and ^{104}Ru) measured by the fusion method were 270 µg/g and by EPA acid digestion 163 µg/g.

The sludge concentrations of ^{99}Tc and ^{238}U measured by ICP-MS are listed in Table 3.7. The ^{99}Tc concentration measured by the fusion method is in the range of 0.875 to 1.42 µg/g with an average value of 1.16 µg/g. These values are similar to the range of 1.05 to 1.22 µg/g measured by the EPA acid digestion method with an average value of 1.14 µg/g. The ^{238}U sludge concentration measured by the fusion method was in the range of 232 to 271 µg/g with an average value of 247 µg/g. The ^{238}U sludge concentration measured by the acid digestion method was in the range of 297 to 327 µg/g with an average value of 311 µg/g.

Table 3.5. Concentrations of Elements Determined from ICP-MS Analysis per Gram of Dry Sludge

Sample Number	Cr - total based on		As - total based on	Se - total based on	Mo - total based on		
	^{52}Cr	^{53}Cr	^{75}As	^{82}Se	^{95}Mo	^{97}Mo	^{98}Mo
	--- µg/g ---						
KOH-KNO$_3$ Fusions							
404	6.69E+02	7.91E+02	(4.15E+01)	<2.39E+03	(5.60E+01)	(3.39E+01)	(2.00E+01)
404 DUP	3.75E+02	4.24E+02	(3.71E+01)	<2.70E+03	(4.53E+01)	(2.83E+01)	(1.61E+01)
405	8.13E+02	8.65E+02	(2.69E+01)	(3.71E+00)	(4.86E+01)	(2.61E+01)	(1.19E+01)
405 DUP	8.01E+02	8.52E+02	(5.67E+01)	<2.29E+03	(5.00E+01)	(2.71E+01)	(1.51E+01)
404 Avg	5.22E+02	6.07E+02	(3.93E+01)	<2.55E+03	(5.07E+01)	(3.11E+01)	(1.81E+01)
405 Avg	8.07E+02	8.58E+02	(4.18E+01)	(1.15E+03)	(4.93E+01)	(2.66E+01)	(1.35E+01)
EPA Acid Digestion							
404	6.84E+02	7.12E+02	(6.90E+01)	<6.17E+03	(5.17E+01)	(4.87E+01)	(2.28E+01)
404 DUP	(5.94E+02)	6.33E+02	(1.11E+02)	(1.28E+01)	(4.62E+01)	(6.18E+01)	(2.47E+01)
405	6.17E+02	6.46E+02	(9.45E+01)	(3.11E+01)	(5.17E+01)	(5.41E+01)	(1.82E+01)
405 DUP	(5.45E+02)	5.58E+02	(8.79E+01)	<6.42E+03	(4.64E+01)	(4.96E+01)	(2.08E+01)
404 Avg	6.39E+02	6.73E+02	(9.01E+01)	<3.09E+03	(4.90E+01)	(5.53E+01)	(2.38E+01)
405 Avg	5.81E+02	6.02E+02	(9.12E+01)	(3.23E+03)	(4.90E+01)	(5.19E+01)	(1.95E+01)

Avg = Average.
DUP = Duplicate sample.
EPA = U.S. Environmental Protection Agency.
ICP-OES = Inductively coupled plasma-optical emission spectroscopy.
KNO$_3$ = Potassium nitrate.
KOH = Potassium hydroxide.
(a) The indicated isotope is the suggested isotope for use to quantify the total concentration of that element.

Table 3.6. Concentrations of Elements Determined from ICP-MS Analysis per Gram of Dry Sludge

Sample Number	Ru -total based on			Ag - total based on		Cd - total based on		Pb - total based on	
	^{101}Ru	^{102}Ru	^{104}Ru	^{107}Ag	^{109}Ag	^{111}Cd	^{114}Cd	^{206}Pb	^{208}Pb
	-- µg/g --								
KOH-KNO$_3$ Fusions									
404	4.39E+02	2.08E+02	2.05E+02	(2.24E+01)	(2.12E+01)	3.06E+02	2.85E+02	4.40E+03	4.93E+03
404 DUP	3.81E+02	1.92E+02	1.70E+02	(2.43E+01)	(2.37E+01)	2.67E+02	2.57E+02	3.98E+03	4.45E+03
405	4.23E+02	2.01E+02	1.96E+02	(1.90E+01)	(1.97E+01)	3.10E+02	2.92E+02	4.56E+03	5.10E+03
405 DUP	4.15E+02	2.03E+02	1.96E+02	(2.28E+01)	(2.28E+01)	2.94E+02	2.90E+02	4.26E+03	4.77E+03
404 Avg	4.10E+02	2.00E+02	1.87E+02	(2.34E+01)	(2.24E+01)	2.87E+02	2.71E+02	4.19E+03	4.69E+03
405 Avg	4.19E+02	2.02E+02	1.96E+02	(2.09E+01)	(2.12E+01)	3.02E+02	2.91E+02	4.41E+03	4.94E+03
EPA Acid Digestion									
404	(2.38E+02)	1.28E+02	1.29E+02	1.29E+03	1.33E+03	3.12E+02	2.86E+02	4.58E+03	5.14E+03
404 DUP	(2.35E+02)	(1.21E+02)	(1.23E+02)	1.09E+03	1.10E+03	2.85E+02	2.64E+02	4.12E+03	4.60E+03
405	(2.53E+02)	1.25E+02	1.32E+02	1.32E+03	1.33E+03	2.98E+02	2.83E+02	4.35E+03	4.86E+03
405 DUP	(2.34E+02)	(1.23E+02)	(1.21E+02)	1.07E+03	1.08E+03	3.45E+02	2.88E+02	4.40E+03	4.85E+03
404 Avg	(2.37E+02)	(1.24E+02)	(1.26E+02)	1.19E+03	1.22E+03	2.99E+02	2.75E+02	4.35E+03	4.87E+03
405 Avg	(2.43E+02)	1.24E+02	1.26E+02	1.19E+03	1.21E+03	3.22E+02	2.85E+02	4.37E+03	4.85E+03
Avg = Average. DUP = Duplicate sample. (a) The indicated isotope is the suggested isotope for use to quantify the total concentration of that element.									

Table 3.7. Concentrations of ^{99}Tc and ^{238}U, Measured by ICP-MS per Gram of Dry Sludge

Sample Number	^{99}Tc	^{238}U
	-------------- µg/g --------------	
KOH-KNO$_3$ Fusions		
404	1.15	239
404 DUP	0.875	232
405	1.42	245
405 DUP	1.21	271
404 Avg	1.01	236
405 Avg	1.31	258
EPA Acid Digestion		
404	1.21	302
404 DUP	1.06	297
405	1.22	327
405 DUP	1.05	315
404 Avg	1.14	300
405 Avg	1.14	321
Avg = Average. DUP = Duplicate. EPA = U.S. Environmental Protection Agency. ICP-MS = Inductively coupled plasma-mass spectroscopy. KNO$_3$ = Potassium nitrate. KOH = Potassium hydroxide.		

The ^{60}Co and ^{137}Cs concentrations in C-106 sludge measured by GEA are listed in Table 3.8. Data are presented for the untreated sludge and for the fusion and acid digestion extracts. For ^{60}Co, the values are similar for the three analyses and range from 0.3301 to 0.4521 µCi/g (2.9×10^{-4} to 3.99×10^{-4} µg/g). For ^{137}Cs, the values are also similar for the three analyses and range from 106 to 173 µCi/g (1.2 to 2.0 µg/g). The highest concentrations for both isotopes are associated with the direct readings and the lowest readings are generally found for the acid extracts.

Table 3.9 lists the concentrations of the transuranic (TRU) isotopes ^{239}Pu, ^{237}Np, and ^{241}Am in the sludge samples. The average ^{239}Pu concentration in 404 sludge measured by the fusion method is 29.5 µg/g (1.83 µCi/g) and the average value by the EPA acid digestion method is 27.3 µg/g (1.70 µCi/g). The ^{237}Np concentration in 404 sludge measured by the fusion method is 8.84 µg/g (6.28×10^{-3} µCi/g) and the average value by the EPA acid digestion method is 8.71 µg/g (6.18×10^{-3} µCi/g). The average ^{241}Am concentration in 404 sludge measured by the fusion method is 1.78 µg/g (6.05 µCi/g), and the average value by the EPA acid digestion method is 2.03 µg/g (6.89 µCi/g).

Table 3.8. ^{137}Cs and ^{60}Co Concentrations in Dry Sludge as Measured by GEA

Sample Number	^{60}Co ----μCi/g-----	^{60}Co ------μg/g---------	^{137}Cs ----μCi/g-----	^{137}Cs ------μg/g---------
Untreated (Raw) Solid				
404	4.521E-01	4.110E-04	1.583E+02	1.820E+00
404 DUP	4.312E-01	3.920E-04	1.545E+02	1.776E+00
405	4.287E-01	3.897E-04	1.697E+02	1.951E+00
405 DUP	4.128E-01	3.753E-04	1.733E+02	1.992E+00
404 Avg	4.417E-01	4.015E-04	1.564E+02	1.798E+00
405 Avg	4.208E-01	3.825E-04	1.715E+02	1.971E+00
KOH-KNO$_3$ Fusions				
404	3.413E-01	3.103E-04	1.411E+02	1.622E+00
404 DUP	4.483E-01	4.075E-04	1.566E+02	1.799E+00
405	4.186E-01	3.805E-04	1.599E+02	1.837E+00
405 DUP	4.108E-01	3.735E-04	1.610E+02	1.851E+00
404 Avg	3.948E-01	3.589E-04	1.488E+02	1.711E+00
405 Avg	4.147E-01	3.770E-04	1.604E+02	1.844E+00
EPA Acid Digestion				
404	3.899E-01	3.544E-04	1.06E+02	1.224E+00
404 DUP	3.301E-01	3.001E-04	1.18E+02	1.351E+00
405	3.726E-01	3.387E-04	1.32E+02	1.514E+00
405 DUP	4.069E-01	3.699E-04	1.283E+02	1.474E+00
404 Avg	3.600E-01	3.273E-04	1.120E+02	1.287E+00
405 Avg	3.897E-01	3.543E-04	1.300E+02	1.494E+00

Avg = Average.
DUP = Duplicate.
EPA = U.S. Environmental Protection Agency.
GEA = Gamma energy analysis.
KNO$_3$ = Potassium nitrate.
KOH = Potassium hydroxide.

Table 3.9. ICP-MS Analysis for Actinides per Gram of Dry Sludge

Sample Number	^{239}Pu μCi/g	^{239}Pu μg/g	^{237}Np μCi/g	^{237}Np μg/g	^{241}Am μCi/g	^{241}Am μg/g
KOH-KNO$_3$ Fusions						
404	1.82	29.36	6.64E-03	9.35	6.22	1.83
404 DUP	1.84	29.72	5.92E-03	8.34	5.88	1.73
405	1.85	29.82	7.11E-03	10.01	6.30	1.85
405 DUP	1.88	30.31	6.65E-03	9.36	6.43	1.89
404 Avg	1.83	29.54	6.28E-03	8.84	6.05	1.78
405 Avg	1.86	30.06	6.88E-03	9.69	6.36	1.87
EPA Acid Digestion						
404	1.85	29.80	6.62E-03	9.32	7.12	2.09
404 DUP	1.54	24.88	5.75E-03	8.09	6.65	1.96
405	1.78	28.78	6.44E-03	9.07	7.10	2.09
405 DUP	1.69	27.30	6.85E-03	9.64	6.90	2.03
404 Avg	1.70	27.34	6.18E-03	8.71	6.89	2.03
405 Avg	1.74	28.04	6.64E-03	9.36	7.00	2.06

Avg = Average.
DUP = Duplicate.
EPA = U.S. Environmental Protection Agency.
ICP-MS = Inductively coupled plasma-mass spectroscopy.
KNO$_3$ = Potassium nitrate.
KOH = Potassium hydroxide.

The average ^{239}Pu concentration in 405 sludge measured by the fusion method is 30.1 µg/g (1.86 µCi/g), and the average value by the EPA acid digestion method is 28.0 µg/g (1.74 µCi/g). The ^{237}Np concentration in 405 sludge measured by the fusion method is 9.69 µg/g (6.88 x 10^{-3} µCi/g), and the average value by the EPA acid digestion method is 9.36 µg/g (6.64 x 10^{-3} µCi/g). The ^{241}Am concentration in 405 sludge measured by the fusion method is 1.87 µg/g (6.36 µCi/g), and the average value by the EPA acid digestion method is 2.06 µg/g (7.00 µCi/g).

DOE defines TRU waste as radioactive waste that at the time of assay contains more than 100 nCi/g of alpha-emitting isotopes with atomic numbers greater than 92 and half-lives greater than 20 years (DOE Order 5820.2A). ^{239}Pu, ^{237}Np, and ^{241}Am are transuranic isotopes with half-lives greater than 20 years. The sum of the averages for sludge samples 404 and 405 for ^{239}Pu, ^{237}Np, and ^{241}Am isotopes measured by the fusion method is 8,057 nCi/g and by the EPA acid digestion method 8,670 nCi/g. Based on the definition, this would classify the sludge itself in C-106 as TRU waste; however, the final waste form will be a mixture of residual sludge and a tank filling grout, which will lower the concentrations of contaminants an unspecified amount.

Table 3.10 lists the gross beta, gross alpha, and ^{90}Sr concentrations in sludge samples from tank C-106. The average gross beta values for the primary (404) and duplicate (405) samples measured by fusion extraction were 5,426 and 6,729 µCi/g; by EPA acid digestion the average values were 8,064 and 8,312 µCi/g. The average gross alpha values for this sludge measured by fusion extraction were 959 and 1,364 µCi/g and by EPA acid digestion were 3,083 and 3,099 µCi/g. The average ^{90}Sr value measured by fusion extraction was 3,620 µCi/g, and the average by EPA acid digestion was 6,080 µCi/g.

Table 3.10. Total Beta and Total Alpha Activities and ^{90}Sr Concentrations per Gram of Dry Sludge

Sample Number	Total Beta	Total Alpha	^{90}Sr	
	------------------µCi/g -----------------		------µCi/g -------	------µg/g-------
KOH-KNO$_3$ Fusions				
404	5.949E+03	1.118E+03	3.51E+03	2.51E+01
404 DUP	4.903E+03	7.988E+02	2.75E+03	1.97E+01
405	7.011E+03	1.482E+03	4.16E+03	2.974E+01
405 DUP	6.447E+03	1.245E+03	4.02E+03	2.87E+01
404 Avg	5.426E+03	9.586E+02	3.13E+03	2.24E+01
405 Avg	6.729E+03	1.364E+03	4.09E+03	2.92E+01
EPA Acid Digestion				
404	8.823E+03	2.81E+03	6.15E+03	4.39E+01
404 DUP	7.304E+03	3.39E+03	5.71E+03	4.08E+01
405	8.680E+03	2.52E+03	6.22E+03	4.44E+01
405 DUP	7.944E+03	3.64E+03	6.24E+03	4.46E+01
404 Avg	8.064E+03	3.099E+03	5.93E+03	4.24E+01
405 Avg	8.312E+03	3.083E+03	6.23E+03	4.45E+01
Avg = Average. DUP = Duplicate. EPA = U.S. Environmental Protection Agency. KNO$_3$ = Potassium nitrate. KOH = Potassium hydroxide.				

Table 3.11 provides a summary of the major components of the sludge as measured for the metals by fusion and acid digestion analysis. For completeness, the anion analyses, as measured by the periodic replenishment water leaching tests (Section 3.4.2.5) have been included in this table. Figure 3.2 shows the components of the tank C-106 sludge that were measured at concentrations greater than 1,000 µg/g by the fusion method for the metals and represents the sum of the period replenishment water-leaching experiments for the anions (Section 3.4.1.7). The predominant metals are Al (13.1 wt%), Mn (11.8 wt%), Na (6.0 wt%), and Fe (4.4 wt%). The predominant anions are oxalate (6.4 wt%) and carbonate (4.0 wt%).

Table 3.11. Summary of Average Concentrations for Tank C-106 Residual Sludge

Analyte	C-106 (µg/g)	
	Fusion	EPA Acid Digestion
Al	131,483	81,699
Ba	1,028	914
Ca	38,221	46,490
Cr	(896)	(727)
Fe	43,777	36,663
K	N/A	8,526
Mg	2,485	3,162
Mn	117,767	108,069
Na	60,400	46,720
Ni	1,712	5,373
Pb	4,841	4,814
Si	(19,086)	(4,895)
Sr	(256)	(493)
^{99}Tc	1.16	1.14
^{238}U	247	310
Water Leach		
F$^-$	33	
Formate	<65	
Cl$^-$	87	
NO$_2^-$	<73	
NO$_3^-$	<70	
CO$_3^{2-}$	39,500	
SO$_4^{2-}$	<66	
Oxalate	63,900	
PO$_4^{3-}$	<91	
EPA = U.S. Environmental Protection Agency.		

The Hanford 222-S Laboratory also conducted extensive characterization of the residual sludge samples collected from tank C-106. Results of these analyses are reported in the *Analytical Results for Tank 241-C-106 Solid Clam Shell Samples Supporting Closure Action* (McKinney 2004). Results by the laboratories are similar for most analytical methods (ICP/MS, IC, alpha energy analysis, gamma energy analysis, ^{99}Tc, percent water). Differences were observed for some analyses/analytes (ICP/OES and ^{129}I). The differences in the measured concentrations for the two laboratories are due to a variety of factors.

Sample heterogeneity is a primary factor in measurement variability for this material. Given the wide variety of sludge solid phases with their variable particle sizes and densities, it is very difficult to mix the material uniformly and produce exact replicate samples. Also, very small amounts (tens of grams) of material were available producing a greater likelihood that duplicate samples collected separately from the tank could have significantly different compositions. Even if the samples were identical, analytical precision and accuracy in the laboratory affects the ability to reproduce the same measurement (most analytical instruments are calibrated within the specification of $\pm 10\%$). Therefore, there will be some variability when the same sample is measured several times on an instrument.

Another factor leading to variability in the tank C-106 results between the two laboratories is the use of different extraction/digestion techniques for the sludge. The 222-S Laboratory used an acid digestion technique to prepare liquid aliquots for the analysis of nonradioactive components of the sludge. The PNNL laboratory used both the fusion technique and an acid digestion method that differs somewhat from the method used by 222-S Laboratory. As shown in Table 3.11, the two methods used to dissolve the sludge resulted in different concentrations for the PNNL analyses, and, thus, it is not surprising that there are differences between the laboratories. For the major metals, these differences are most likely due to the ability of the fusion method to more completely dissolve the silicate minerals and more recalcitrant oxide and hydroxide phases. The digestion methods used by both laboratories produced a solid residue that could not be analyzed. The likely variability in the compositions of these residues adds to the variability in the reported sludge compositions. For these reasons, it is expected that some differences in reported concentrations of the sludges will occur and should be considered a natural outcome of the characterization process of these materials. Chemists from the two laboratories are comparing preparation and analytical techniques for potential improvements in the analysis of Hanford tank waste.

In the case of ^{129}I, discrepancies in the reported concentrations from the two laboratories exceeded a ratio that could easily be explained by sample heterogeneity. The PNNL results provided in the following section are approximately 3 times greater than the analytical detection limit reported by chemists from the 222-S Laboratory. An analytical detection limit was reported for ^{129}I by the 222-S Laboratory staff because it was not detected in the C-106 sludge samples analyzed at the laboratory. The discrepancy in the ^{129}I data from the two laboratories could be an artifact of two things: sample preparation and analytical techniques. Both laboratories performed a KOH:KNO$_3$ fusion/water extraction to remove ^{129}I from the sludge. However, due to the presence of oxidized metals and residual solids in the extracted samples, researchers at PNNL added a mixture of concentrated H_2SO_4 and 1 M NaHSO$_3$ to the sample. This additional step resulted in complete sample dissolution while preventing oxidation of the dissolved species.

The way in which researchers at the two laboratories analyzed the extracted samples differed considerably. Scientists at PNNL performed dilutions on the fully dissolved samples and analyzed them for ^{127}I and ^{129}I using ICP-MS. Chemists at the 222-S Laboratory took the KOH:KNO$_3$ fusion/water extracted samples through a multi-step liquid:liquid extraction/purification process, followed by precipitation as AgI. The AgI precipitate was eventually counted (X-rays) using a Low Energy Photon Spectrometry (LEPS) detector. Although the liquid:liquid procedure utilized by researchers at the 222-S Laboratory is a common technique, care must be taken throughout the various steps to ensure

volatilization of iodine does not occur. Given the thoroughness of the PNNL extraction procedure and the straightforward analytical technique, it would be prudent and conservative to use the ^{129}I values provided in this report (Section 3.2) for risk assessment calculations.

3.2 ^{129}I Extraction and Measurement

Table 3.12 contains results of the ^{129}I analysis of the modified KOH:KNO$_3$ water fusion of tank C-106 sludge material. The data are reported as pCi ^{129}I per gram of sludge (calculated on a dry weight basis). The italicized data reported for sample 405 C-106 matrix spike indicate that the reported value was corrected for the ^{129}I spike contribution. ICP-MS analysis of ^{129}I was better than ±10% of certified reference standards, with the linear operating range extending three orders of magnitude (0.01 to 10 ng/mL). The measured values are in the range of 73.4 to 143 pCi/g for sludge samples 404 and 405.

The ^{129}I concentrations in extracts from the standard KOH:KNO$_3$ fusion method were greater than the EQL (see Section 3.1 for a discussion of EQL calculation). The instrument EQL for this analysis was 0.88 pCi/L, which resulted in dilution-corrected sample EQLs ranging from 13.3 pCi/g to 22.6 pCi/g. The modified KOH:KNO$_3$ water fusion method was successful at dissolving the entire sludge sample tested. Additionally, the ^{129}I recovery of the blank spike sample analyzed as part of this dataset was excellent at 98.7%. Therefore, the fusion data reported in Table 3.12 represent the total ^{129}I present in tank C-106 sludge.
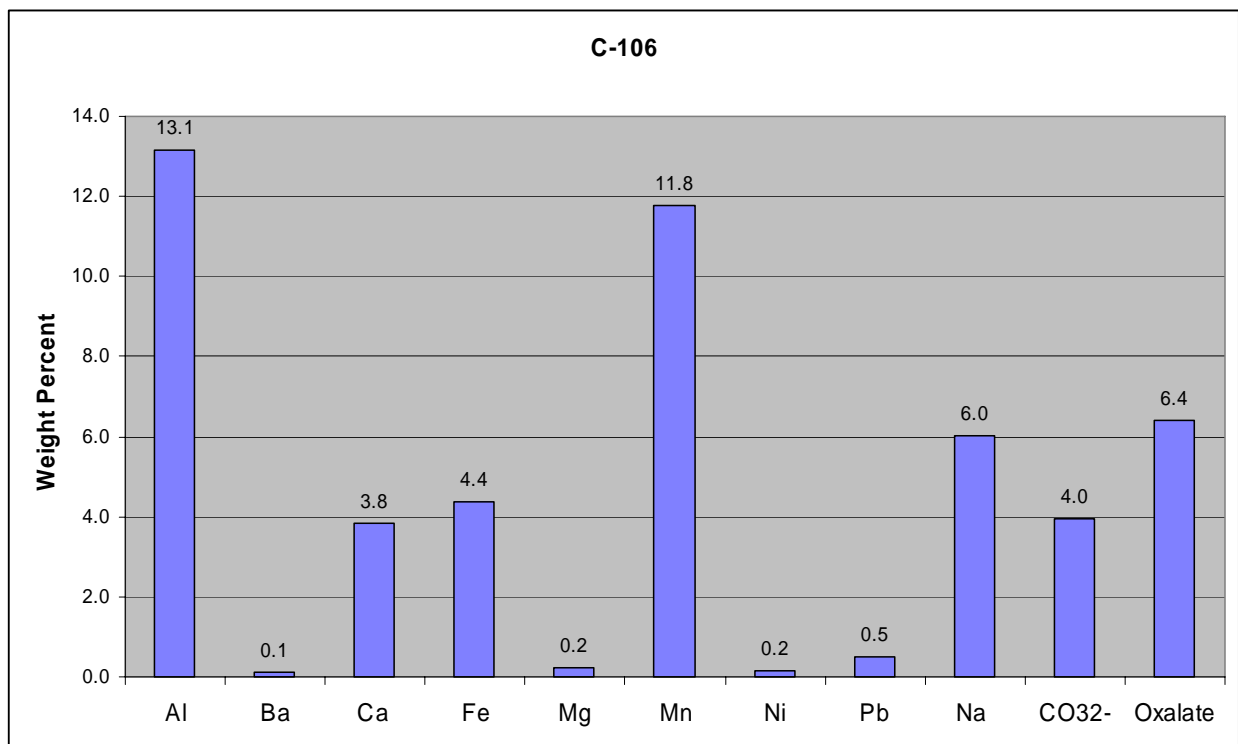
Figure 3.2. Tank C-106 Sludge Composition

3.14

Table 3.12. Summary of ^{129}I Concentrations for Modified KOH-KNO$_3$ Water Fusion Extracts for Tank C-106

Sample Number	^{129}I pCi/g
404 C-106	105
404 C-106 DUP	112
405 C-106	143
405 C-106 DUP	116
405 C-106 Matrix Spike	*73.4*
Italicized data means spike contribution has been removed. DUP = Duplicate sample.	

3.3 Composition of Tank Liquid

Sample 403 is a sample of the liquid in the tank collected near the cessation of final pumping during retrieval. It is representative of any supernatant and porewater in the residual sludge remaining in the tank. The composition of this liquid is provided in Table 3.13. The solution pH is 12.9 because of the addition of 0.5 M NaOH to neutralize remaining oxalic acid in the tank. The major cations in solution are Na (10,200 mg/L avg) and Al (121 mg/L avg); while the major anions are carbonate (9,930 mg/L avg) and oxalate (1,345 mg/L avg).

Table 3.13. Sample 403 – Tank C-106 Liquid Sample Composition

Analyte	Concentration (mg/L$^{(a)}$)	
	Primary	Duplicate
pH (standard units)	12.9	
Metals		
Al	126.0	116
Ba	3.5	(0.09)
Ca	21.9	(4.1)
Cr	(0.28)	(0.34)
Fe	(1.9)	(0.6)
K	(14)	(12)
Mg	(0.9)	(0.24)
Mn	(0.16)	(0.1)
Na	10,300	10,100
Ni	3.9	3.0
Pb	(0.16)	(0.13)
Si	(158)	(141)
Radionuclides		
^{99}Tc	0.0003	(0.00012)
^{238}U	1.24	1.03
^{137}Cs (μCi/L)	505	442
Total Alpha (μCi/L)	30.3	30.2
Total Beta (μCi/L)	470	469
Anions		
Oxalate	1,340	1,350
CO_3^{2-}	10,020	9,840
Cl^-	20.5	20.5
NO_3^-	9.43	9.41
F^-	1.38	<1.17
SO_4^{2-}	27.6	27.6
PO_4^{3-}	75.1	74.6
(a) mg/L unless otherwise noted.		

3.4 Water-Leaching Tests

The results of the water-leaching tests of sludge samples are discussed in this section. These include results from the single-contact and periodic replenishment tests. Concentration values in this section are given in terms of μg or μCi per gram of dry sludge. Dissolved concentrations in extracts from these tests are provided on a per liter of solution basis in Appendix F.

3.4.1 Single-Contact Test Results

The single-contact water-leach tests were run in duplicate with contact times of 1 day, 2 weeks, and 1 month. DDI water was used as the leaching solution. The results of the solution analyses of the water extracts are described in this section.

3.4.1.1 Digestion Factors and Moisture Contents – Single-Contact Water Extracts

The digestion factors for the 404 and 405 sludge samples used for the water extracts varied from 7 to 13.5 g/L. These digestion factors are the ratios of wet weight of sludge to 30 mL DDI water used to dissolve the soluble portion of the solid. The digestion factors were then multiplied by the percent solids, as determined from moisture content analysis, to convert to a dry weight basis. The variability is a function of the mass of sludge used, which ranged from approximately 0.2 to 0.4 g. The moisture contents averaged about 50.5% for the 404 and 405 samples (Table 3.1).

3.4.1.2 Water Extract pH and Alkalinity – Single-Contact Water Extracts

The average pH values of the duplicate water extracts of the sludge samples are listed in Table 3.14. The effluent pH values for sludge sample 404 were 7.03 and 7.17, and the values for 405 were 6.71 and 7.41. The pH values increase slightly between the 1-day and 1-month tests.

Table 3.14. Water Extract pH and Alkalinity Values Corrected to Grams of Dry Sludge

Sample Number	pH	Total Alkalinity as $CaCO_3$ mg/g
404 1 Day Avg	7.03	42.7
404 2 Week Avg	INS	INS
404 1 Month Avg	7.17	15.52
405 1 Day Avg	6.71	59.6
405 2 Week Avg	INS	INS
405 1 Month Avg	7.41	21.38
Avg = Average. INS = Instrument malfunction.		

The total alkalinities of the water extracts reported in units of $CaCO_3$ mg/g sludge are also shown in Table 3.14. The values for sludge sample 404 were 15.52 (for the month-long test) and 42.7 mg/g (for the day-long test), and the values for sample 405 after 1 month and 1 day were 21.38 and 59.6 mg/g, respectively. The alkalinities decrease by about a factor of three between the 1-day and 1-month tests. This suggests the precipitation of a carbonate mineral.

3.4.1.3 ^{99}Tc and ^{238}U – Single-Contact Water Extracts

^{99}Tc and ^{238}U are two of the more important potential long-term risk constituents in tank sludge because of their long half-lives and high mobility once dissolved in water. Table 3.15 lists the concentrations of these two radionuclides in units of µg leached/g-solid and µCi leached/g-solid for the single-contact water extracts. Table 3.16 lists the percentage of the total ^{99}Tc and ^{238}U leached from the sludge by each water-leach test versus the total available concentration given by the fusion analysis. The water-leachable amount of ^{99}Tc in sample 404 ranges from 0.0180 to 0.0339 µg/g, which is 1.78 to 3.35% of the total ^{99}Tc in the sludge. For sample 405, the water-leachable ^{99}Tc ranges from 0.0161 to 0.0349 µg/g, which is 1.22 to 2.66% of the available ^{99}Tc. The lowest amount of ^{99}Tc leached in the one-day contact test, and similar amounts leach at contact times of 2 weeks and 1 month. That is, a small amount of ^{99}Tc leaches within 1 day but the leaching rate decreases such that the total amount leached after 30 days is still small. The recalcitrant nature of some of the ^{99}Tc in other Hanford tank sludge has been documented in similar water-leach studies of sludges from tanks AY-102, C-203, and C-204 (Lindberg and Deutsch 2003; Krupka et al. 2004).

Table 3.15. ^{99}Tc and ^{238}U Concentrations in Tank C-106 Sludge from Single-Contact Water-Leach Tests

Sample Number	^{99}Tc	^{238}U	^{99}Tc	^{238}U
	---------- µg/g ----------		---------- µCi/g ----------	
404 1 Day Avg	0.018	6.41	3.07E-04	2.18E-06
404 2 Week Avg	0.0339	3.67	5.76E-04	1.25E-06
404 1 Month Avg	0.0313	3.10	5.32E-04	1.05E-06
405 1 Day Avg	0.0162	7.66	2.73E-04	2.61E-06
405 2 Week Avg	0.0349	3.99	5.94E-04	1.36E-06
405 1 Month Avg	0.0326	3.70	5.54E-04	1.26E-06
Avg = Average.				

Table 3.16.   Water-Leachable Percentages of ^{99}Tc and ^{238}U in C-106 Sludge Samples Compared with Fusion Results

Sample Number	^{99}Tc	^{238}U
	Percent Water Leachable	
404 1 Day Avg	1.78%	2.72%
404 2 Week Avg	3.35%	1.56%
404 1 Month Avg	3.09%	1.31%
405 1 Day Avg	1.22%	2.97%
405 2 Week Avg	2.66%	1.55%
405 1 Month Avg	2.48%	1.43%
Avg = Average.		

The water-leachable amount of ^{238}U in sample 404 ranges from 3.10 to 6.41 µg/g, which is 1.31 to 2.72% of the total ^{238}U in the sludge. For sample 405, the water-leachable ^{238}U ranges from 3.70 to 7.66 µg/g, which is 1.43 to 2.97% of the available ^{238}U. The water leachability of ^{238}U appears to decrease with contact time; however, this decrease is small and not considered to be statistically significant.

3.4.1.4 ^{129}I Concentrations – Single-Contact Water Extracts

Table 3.17 contains results of the ^{129}I analysis of the one-day water extracts of the C-106 sludge samples. The data are reported as pCi ^{129}I per gram of sludge (calculated on a dry weight basis). ICP-MS measurement uncertainties for ^{129}I were smaller than ±10% of certified reference standards, with the linear operating range extending 3 orders of magnitude (0.01 to 10 ng/mL). The water-leachable ^{129}I concentrations in extracts from these tests were less than the EQL (see Section 3.1 for a discussion of EQL calculation) for the analysis. The instrument EQL for this analysis was 1.8 pCi/L, which resulted in dilution-corrected sample EQLs ranging from 2.2 to 3.2 pCi/g for the water extracts. Although the water leachable ^{129}I results were less than the EQL of the analysis, it is apparent from a comparison with the total ^{129}I in the sludge (73.4 to 143 pCi/g, Section 3.2) that very little (less than 3%) of the ^{129}I in C-106 residual sludge samples was leachable in this one-day, water contact test.

3.4.1.5 Selected Metal Concentrations – Single-Contact Water Extracts

Metals detected at measurable concentrations in one or more samples in the water extracts or the fusion analyses are listed in Table 3.18. A discussion of the meaning of the less-than values (<) and the values in parentheses are provided in Section 3.1. The fusion analyses showed that Al, Mn, Na, Fe, and Ca are the dominant metals present in the tank C-106 sludge (Figure 3.1 and Figure 3.2). The water extracts show that Na and Mn are the primary water-soluble constituents with much smaller amounts of Ba, Ca, Mg, Ni, and (perhaps) Si also being leachable. Very little Al or Fe are water leachable from the sludge. (Note that the concentrations measured in the water leachates are the sums of the water-leachable amounts and the initial dissolved pore water concentrations.)

Table 3.17. Summary of ^{129}I Concentrations from the One Day Single-Contact Water-Leach Tests for Tank C-106 Sludge Samples

Sample Number	^{129}I pCi/g
404 1 Day	(1.28)
4041 Day DUP	(1.44)
405 1 Day	(1.28)
405 1 Day DUP	(1.59)
DUP = Duplicate.	

Table 3.18. Water-Leachable Average Metal Concentrations in Tank C-106 Single-Contact Water Extracts

Sample Number	Al	Ba	Ca	Cr	Fe	K	Mg	Mn	Na	Ni	Pb	Si	Sr
								--µg/g--					
404 1 Day Avg	83.2	35.1	329	(1.79)	(24.2)	(85.4)	152	8,870	10,700	183	(2.63)	(357)	(4.23)
404 2 Week Avg	(27.3)	9.81	69.5	(2.89)	(4.36)	(77.6)	193	10,400	11,900	121	(0.89)	(281)	(5.37)
404 1 Month Avg	(10.1)	15.3	70.4	2.51	(1.09)	(77.9)	176	8,680	10,400	96.4	(0.39)	314	5.04
405 1 Day Avg	126	91.1	739	(32.7)	(58.6)	(141)	247	15,500	9,890	298	(10)	(524)	(8.74)
405 2 Week Avg	(34.7)	18.6	115	(4.38)	(4.75)	(121)	235	15,000	10,900	154	(1.66)	(364)	(8.59)
405 1 Month Avg	(12.0)	23.7	112	3.74	(2.16	(89.2)	211	12,700	9,600	121	(1.20)	451	7.94
Avg = Average.													

Table 3.19 lists the leachable percentages of metals present in the single-contact water extracts. These values show that 16.2% to 19.3% of the total Na is water-leachable and 4.7% to 21.6% of the Ni is leachable. About 7% to 12% of the Mn and Mg are dissolved in water. Other elements that have leachabilities greater than 1% in one or more of the tests are Ba, Ca, Si, and Sr. Elements with lower water leachabilities are Al, Cr, Fe, and Pb.

3.4.1.6 Anion Concentrations – Single-Contact Water Extracts

The anion concentrations in water extracts from the tank C-106 sludge samples are listed in Table 3.20. The primary leachable anions are carbonate and oxalate, with much smaller quantities of chloride and fluoride leaching. The presence of high concentrations of oxalate (25,911 to 39,211 µg/g) in the water extracts shows that the sludge has the capacity to act as a reductant for more oxidized species. However, its presence in the sludge in contact with air suggests the oxalate is not readily oxidized in this environment.

3.4.1.7 Radioanalytical Results – Single-Contact Water Extracts

The results of the GEA analysis for ^{60}Co and ^{137}Cs in the water extractions are listed in Table 3.21. For the tank C-106 sludge samples, ^{60}Co was not detected in any of the leachates and ^{137}Cs was measured at values of 2.03 and 2.65 µg/g in the one-day water leach.

Total beta and total alpha levels for the one-day, water-leach experiments are listed in Table 3.22. Comparing these results with the fusion analysis levels (Table 3.10) shows that about 2% of the total beta is water leachable from tank C-106 sludge. Approximately 0.02% of the total alpha is water leachable from the sludge. Table 3.22 also lists the water-leachable concentrations of ^{90}Sr. Comparing these levels to the fusion analysis values (Table 3.10) shows that 2.0% is water leachable from tank C-106 sludge.

Table 3.23 lists the water-leachable concentrations of ^{239}Pu, ^{237}Np, and ^{241}Am in terms of the original sludge compositions. The percent water leachabilities for these actinides are listed in Table 3.24. The water leachability of ^{239}Pu and ^{241}Am are much less than 1%. The leachability of ^{237}Np is in the range of 7.78 to 9.8% for the 1-day and 1-month contact tests. No actinide measurements were made on the 2-week water leach test solutions.

3.4.2 Periodic Replenishment Test Results

The periodic replenishment tests were conducted by contacting each sludge sub-sample with sequential 30-mL quantities of fresh DDI water. As shown in Table 3.25, the sludge was contacted six times with fresh water. The first five contacts were of short duration (1 or 4 days each), which was assumed sufficient for leaching contaminants of concern from the sludge. To test this assumption, an

Table 3.19. Water-Leachable Metals in Tank C-106 Single-Contact Water Extract Compared with Fusion Results (%)

Sample Number	Al	Ba	Ca	Cr	Fe	K	Mg	Mn	Na	Ni	Pb	Si	Sr
	--% Water Leachable--												
404 1 Day Avg	0.06	3.29	0.86	(0.21)	(0.06)	N/A	5.69	7.81	17.33	8.96	(0.05)	(1.88)	(1.78)
404 2 Week Avg E	(0.02)	0.92	0.18	(0.34)	(0.01)	N/A	7.22	9.18	19.31	5.91	(0.02)	(1.48)	(2.26)
404 1 Month Avg	(0.0)	1.44	0.18	(0.30)	(0.00)	N/A	6.57	7.64	16.85	4.71	(0.01)	1.66	2.12
405 1 Day Avg	0.10	9.20	1.95	(3.44)	(0.13)	N/A	10.73	12.73	16.69	21.64	(0.21)	(2.73)	(3.18)
405 2 Week Avg E	(0.03)	1.88	0.30	(0.46)	(0.01)	N/A	10.21	12.29	18.43	11.14	(0.03)	(1.89)	(3.12)
405 1 Month Avg	(0.01)	2.40	0.30	0.39	(0.00)	N/A	9.18	10.39	16.20	8.79	(0.02)	2.35	2.89
Avg = Average.													
N/A = Not applicable - potassium salts are used as fluxing agent.													

Table 3.20. Average Anion Concentrations in Tank C-106 Single-Contact Water Extract

Sample Number	Fluoride	Acetate	Formate	Chloride	Nitrite	Nitrate	Carbonate	Sulfate	Oxalate	Phosphate
	-- μg/g --									
404 1 Day Avg	35.3	<173	<51.1	82.6	<57	<55	20,829	<52	25,911	<84
404 2 Week Avg	INS	INS	INS	INS	INS	INS	INS	INS	INS	INS
404 1 Month Avg	<129	<1,508	<445	519	<496	<475	<55,026	<450	34,350	<0.72
405 1 Day Avg	34.4	<285	<84	99	<94	<90	12,633	<85	39,211	<105
405 2 Week Avg	INS	INS	INS	INS	INS	INS	INS	INS	INS	INS
405 1 Month Avg	191	<2,176	<642	410	<716	<686	<79,404	<650	37,946	<0.96
Avg = Average.										

Table 3.21. GEA for Tank C-106 Single-Contact Water Extractions and Direct Solids

Sample Number	^{60}Co ----μCi/g-----	^{60}Co ------μg/g---------	^{137}Cs ----μCi/g-----	^{137}Cs ------μg/g---------
404 1 Day Avg	<1.77E-02	<1.61E-05	2.65E+00	3.044E-02
405 1 Day Avg	<2.92E-02	<2.65E-05	2.03E+00	2.329E-02
Avg = Average.				

Table 3.22. Total Beta, Total Alpha, and ^{90}Sr – Single-Contact Water Extracts

Sample Number	Total Beta --------μCi/g-------	Total Alpha --------μCi/g-------	^{90}Sr --μCi/g--	^{90}Sr --μg/g--
404 1 Day Avg	9.97E+01	1.68E-01	4.71E+01	3.37E-01
405 1 Day Avg	1.75E+02	2.87E-01	8.53E+01	6.09E-01
Avg = Average.				

Table 3.23. Actinide Analysis for Tank C-106 Single-Contact Water Extracts

Sample Number	^{239}Pu --μCi/g--	^{239}Pu --μg/g--	^{237}Np --μCi/g--	^{237}Np --μg/g--	^{241}Am --μCi/g--	^{241}Am --μg/g--
404 1 Day	(3.81E-04)	(6.14E-03)	5.33E-04	7.51E-01	2.57E-03	7.55E-04
404 1 Month	9.10E-05	1.47E-03	4.88E-04	6.88E-01	4.88E-04	1.66E-03
405 1 Day	(1.18E-02)	6.74E-04	9.50E-01	<1.92E-03	<5.65E-04	(1.18E-02)
405 1 Month	2.33E-03	6.74E-04	9.49E-01	7.38E-04	2.51E-03	2.33E-03

Table 3.24. Water-Leachable Percentage for Actinides in C-106 Single-Contact Water Extracts Compared to Fusion Analysis

Sample Number	^{239}Pu	^{237}Np	^{241}Am
% Water Leachable			
404 1 Day	(0.02%)	8.49%	0.04%
404 1 Month	(0.005%)	7.78%	0.09%
405 1 Day	(0.04%)	9.80%	0.03%
405 1 Month	(0.008%)	9.79%	0.13%

extended period of leaching was conducted in which the sludge, after the first five stages of leaching, was contacted with water for 43 days (Stage 6A) or 82 days (Stage 6B). The objective was to evaluate the long-term leaching characteristics of contaminants from the sludge. Very little of the primary contaminants of concern, ^{99}Tc and ^{238}U, were leachable during these tests. Over 90% of the ^{99}Tc and ^{238}U remained in the sludge and was not leachable during the six stages of water leaching.

3.4.2.1   Digestion Factors and Moisture Contents – Periodic Replenishment Water Tests

The digestion factors for the 404 and 405 sludge samples used for the replenishment tests varied from 4.5 g/L to 8.1 g/L. These digestion factors are the ratios of wet weight of sludge to the amount of DDI water (30 mL) used to dissolve the soluble portion of the solid. The digestion factors were then multiplied by the percent solids, as determined from moisture content analysis, to convert concentrations to a dry weight basis. The variability in digestion factors is a function of the mass of sludge used, which ranged from approximately 0.2 to 0.38 g.

The average moisture contents [(wet wt – dry wt)/dry wt] were 57.5% for the 404 samples and 43.5% for the 405 samples.

3.4.2.2   Water Extract pH and Alkalinity – Periodic Replenishment Water Tests

Table 3.25 lists the pH values and alkalinities for the leachates from the replenishment tests. The leachate pH values for sample 404 during the first five stages are in the range 6.87 to 7.03 and then increase to an average of 7.5 in Stage 6. There is a similar trend in pH for sample 405, although the initial pH values are lower in the range of 6.68 to 6.71. The increase in pH at Stages 6A and 6B may reflect equilibration with a carbonate mineral. The alkalinity values decrease from values of 42.71 and 59.58 mg $CaCO_3$ per gram of sludge to about 13 and 16 mg $CaCO_3$ per gram of sludge in samples 404 and 405, respectively.

Table 3.25.   Contact Times, Average pH Values, and Alkalinities for Periodic Replenishment Tests on Tank C-106 Sludge Samples

Contact Stage	Contact Duration (days)	pH	Alkalinity as mg/g $CaCO_3$
Sample 404			
1	1	7.03	42.71
2	1	6.87	<31.62
3	4	6.93	<31.62
4	1	6.75	<31.62
5	1	6.73	<31.62
6A	43	7.43	12.43
6B	82	7.62	13.89
Sample 405			
1	1	6.71	59.58
2	1	6.70	53.59
3	4	6.79	<52.09
4	1	6.68	<52.09
5	1	6.70	<52.09
6A	43	7.50	15.04
6B	82	7.70	16.99

3.4.2.3 ^{99}Tc and ^{238}U – Periodic Replenishment Tests

Table 3.26 lists the amount of ^{99}Tc and ^{238}U leached from the two sludge samples during the replenishment tests, and Table 3.27 lists the percentages of the available radionuclides that were leached. As was found in the single-contact water leach tests (Table 3.16), very little of the ^{99}Tc or ^{238}U was water leachable. Only 2.3% of the ^{99}Tc is leachable in the first five contact stages for sample 404, while an additional 3.57% and 3.73% were leachable during Stages 6A and 6B, respectively. Similar amounts of ^{99}Tc were leachable throughout the six stages of leaching sample 405.

Approximately 4% of the ^{238}U was water leachable throughout the six contact stages for sample 404, with similar amounts leaching from sample 405. These results show the recalcitrant nature of the residual ^{99}Tc and ^{238}U in the sludge in tank C-106 after the oxalic acid sluicing campaign.

Table 3.26. ^{99}Tc and ^{238}U Concentrations in Tank C-106 Sludge Samples - Periodic Water Replenishment Tests

Contact Stage	Contact Duration (days)	^{99}Tc ---------- µg/g ----------	^{238}U	^{99}Tc ---------- µCi/g ----------	^{238}U
Sample 404					
1	1	1.98E-02	7.03E+00	3.36E-04	2.39E-06
2	1	(2.31E-03)	1.31E+00	(3.93E-05)	4.46E-07
3	4	3.56E-03	5.56E-01	6.06E-05	1.89E-07
4	1	(2.51E-03)	2.28E-01	(4.27E-05)	7.75E-08
5	1	<3.79E-03	9.25E-02	<6.45E-05	3.14E-08
6A	43	3.62E-02	2.04E-01	6.15E-04	6.92E-08
6B	82	3.77E-02	1.66E-01	6.41E-04	5.63E-08
Sample 405					
1	1	1.61E-02	7.66E+00	2.73E-04	2.61E-06
2	1	(3.60E-03)	1.68E+00	(6.12E-05)	5.71E-07
3	4	(5.29E-03)	6.65E-01	(9.00E-05)	2.26E-07
4	1	<4.20E-03	1.89E-01	<7.13E-05	6.44E-08
5	1	<6.25E-03	8.28E-02	<1.06E-04	2.81E-08
6A	43	3.62E-02	2.45E-01	6.16E-04	8.35E-08
6B	82	4.63E-02	2.74E-01	7.87E-04	9.31E-08

3.4.2.4 Selected Metal Concentrations – Periodic Replenishment Water Extracts

Table 3.28 lists the concentrations of metals leached during the periodic replenishment tests, and Table 3.29 lists the percentage of the total available metals that were leached during each stage of the test. The leachable metals from the two sludge samples are Ba, Ca, Mg, Mn, Na, and Ni. Non-leachable metals throughout the periodic replenishment tests were Al, Cr, Fe, Pb, Si, and Sr.

Table 3.27. Water-Leachable Percentages of ^{99}Tc and ^{238}U in Tank C-106 Sludge Samples - Periodic
Water Replenishment Tests Relative to Fusion Results

Contact Stage	Contact Duration (days)	^{99}Tc	^{238}U
		% Water Leachable	
Sample 404			
1	1	1.95	2.98
2	1	(0.23)	0.56
3	4	0.35	0.24
4	1	(0.25)	0.10
5	1	<0.37	0.04
6A	43	3.57	0.09
6B	82	3.73	0.07
Sample 405			
1	1	1.22	2.97
2	1	(0.27)	0.65
3	4	(0.40)	0.26
4	1	<0.32	0.07
5	1	<0.48	0.03
6A	43	2.76	0.10
6B	82	3.53	0.11
< EQ L = below the estimated quantitation limit.			

There are two trends observable in the metals that are leachable in this test. The amount leachable, as a percentage of the total metal in the sludge, during the first five stages of leaching for the metals Ba, Ca, and Ni, is approximately constant. The percent water leachable ranges for these metals in sample 404 are Ba (2.13 – 4.34%), Ca (0.92 – 1.69%), and Ni (9.30 – 10.83%). The percent water leachable ranges for these metals in sample 405 are Ba (5.24 – 14.38%), Ca (1.54 – 2.09%), and Ni (15.97 – 26.88%). For the metals Mg, Mn, and Na, there is a strong, uniform, decreasing trend of leachability for the first five stages of this test. For example, in sample 404 the Mn leachability decreases from 8.57 to 1.62% and the Na leachability decreases from 19.01 to 1.48% for stages 1 and 5. Constant levels of leachability, such as those observed for Ba, Ca, and Ni, suggest equilibrium control on solution concentration by a single mineral for each metal. The decreasing leachability trend observed for Mg, Mn, and Na suggests the dissolution and removal from the sludge of a soluble mineral containing the metal.

At Stages 6A (43-day contact) and 6B (82-day contact), the leachable percentages of Ba, Mg, Mn, and Ni decrease from values observed in the first five stages of leaching, whereas the value for Ca is fairly constant. The leachability of Na during stages 6A and 6B increases relative the values for Stages 3 through 5, but decreases compared to Stages 1 and 2. Sr leachability at measurable amounts above the EQL only occurs in stages 6A and 6B.

3.25

Table 3.28. Water-Leachable Average Metals in Tank C-106 Sludge Samples - Periodic Water Replenishment Tests

Contact Stage	Al	Ba	Ca	Cr	Fe	K	Mg	Mn	Na	Ni	Pb	Si	Sr
	--µg/g--												
Sample 404													
1	9.13E+01	3.85E+01	3.61E+02	(1.97E+00)	(2.66E+01)	(9.37E+01)	1.67E+02	9.73E+03	1.17E+04	2.01E+02	(2.88E+00)	(3.91E+02)	(4.64E+00)
2	8.16E+01	3.45E+01	3.97E+02	<1.97E+01	(2.17E+01)	<4.12E+02	1.59E+02	9.45E+03	7.41E+03	1.89E+02	(4.52E+00)	(3.52E+02)	(5.48E+00)
3	(6.68E+01)	3.42E+01	3.54E+02	(3.95E+01)	(2.06E+01)	(4.25E+02)	1.57E+02	9.02E+03	2.12E+03	1.90E+02	(4.19E+00)	(3.19E+02)	(5.86E+00)
4	6.41E+01	4.48E+01	3.55E+02	<2.10E+01	(2.50E+01)	<5.11E+01	1.50E+02	7.88E+03	1.90E+03	1.95E+02	(2.46E+00)	(3.34E+02)	(6.46E+00)
5	6.18E+01	(4.62E+01)	3.62E+02	<2.10E+01	(2.24E+01)	(2.51E+01)	1.28E+02	6.39E+03	1.63E+03	1.94E+02	<1.54E+00	(3.38E+02)	(7.03E+00)
6A	(4.47E+00)	1.52E+01	2.52E+02	(8.78E-01)	(4.57E-01)	(2.04E+01)	4.14E+01	1.54E+03	3.32E+03	7.96E+01	<1.63E+01	(2.47E+02)	1.11E+01
6B	(7.05E+01)	1.99E+01	(4.30E+02)	(1.19E+00)	1.13E+00	(8.65E+01)	8.38E+01	1.81E+03	4.02E+03	7.96E+01	<1.53E+01	(2.23E+02)	1.71E+01
Sample 405													
1	1.26E+02	9.11E+01	7.39E+02	(3.27E+01)	(5.86E+01)	(1.41E+02)	2.47E+02	1.55E+04	9.89E+03	2.98E+02	(1.00E+01)	(5.24E+02)	(8.74E+00)
2	1.10E+02	5.19E+01	7.00E+02	<3.52E+01	(4.91E+01)	(6.90E+02)	1.95E+02	1.17E+04	2.05E+03	3.71E+02	<6.13E+01	(3.85E+02)	(9.58E+00)
3	1.11E+02	6.50E+01	5.84E+02	<6.51E+01	(3.25E+01)	(8.10E+01)	1.40E+02	7.45E+03	1.98E+03	3.59E+02	(3.09E+00)	(4.62E+02)	(1.11E+01)
4	1.09E+02	8.54E+01	7.32E+02	<6.51E+01	(1.89E+01)	<1.30E+03	(5.16E+01)	2.71E+03	1.34E+03	3.22E+02	(3.35E+00)	(3.14E+02)	(1.19E+01)
5	1.13E+02	1.42E+02	7.94E+02	(2.91E+00)	(1.54E+01)	(1.40E+02)	(3.98E+01)	1.39E+03	1.26E+03	2.20E+02	(1.04E+01)	(2.69E+02)	(1.30E+01)
6A	(1.36E+01)	1.99E+01	2.92E+02	(1.11E+00)	(9.84E-01)	(3.47E+01)	3.35E+01	9.24E+02	4.80E+03	3.87E+01	<2.45E+01	(2.49E+02)	2.47E+01
6B	(1.32E+02)	3.45E+01	(7.83E+02)	(1.76E+00)	(1.50E+00)	(2.17E+01)	(9.40E+01)	1.37E+03	5.90E+03	3.68E+01	(1.20E+00)	(2.92E+02)	(3.71E+01)

3.27

Table 3.29. Percent Water-Leachable Average Metal Concentrations in Tank C-106 Sludge Samples - Periodic Water Replenishment Tests

Contact Stage	Al	Ba	Ca	Cr	Fe	K	Mg	Mn	Na	Ni	Pb	Si	Sr
					-----% Water Leachable-----								
Sample 404													
1	0.07	3.61	0.94	(0.23)	(0.06)	N/A	6.24	8.57	19.01	9.83	(0.06)	(2.06)	(1.95)
2	0.06	3.24	1.03	<2.34	(0.05)	N/A	5.94	8.32	12.04	9.22	(0.09)	(1.86)	(2.30)
3	(0.05)	3.21	0.92	(4.70)	(0.05)	N/A	5.88	7.94	3.45	9.30	(0.09)	(1.68)	(2.47)
4	0.05	4.20	0.92	<2.49	(0.06)	N/A	5.63	6.94	3.09	9.53	(0.05)	(1.76)	(2.72)
5	0.04	(4.34)	0.94	<2.49	(0.05)	N/A	4.78	5.63	2.66	9.47	<0.03	(1.78)	(2.96)
6A	(0.003)	1.43	0.65	(0.10)	(0.001)	N/A	1.55	1.36	5.40	3.89	<0.34	(1.30)	4.67
6B	(0.05)	1.87	(1.12)	(0.14)	0.003	N/A	3.14	1.59	6.54	3.89	<0.32	(1.18)	7.18
Sample 405													
1	0.10	9.20	1.95	(3.44)	(0.13)	N/A	10.73	12.73	16.69	21.64	(0.21)	(2.73)	(3.18)
2	0.09	5.24	1.84	<3.70	(0.11)	N/A	8.47	9.56	3.46	26.88	<1.26	(2.00)	(3.48)
3	0.09	6.57	1.54	<6.84	(0.07)	N/A	6.10	6.11	3.34	26.06	(0.06)	(2.40)	(4.04)
4	0.09	8.63	1.93	<6.84	(0.04)	N/A	(2.25)	2.22	2.26	23.35	(0.07)	(1.64)	(4.34)
5	0.09	14.38	2.09	(0.31)	(0.03)	N/A	(1.73)	1.14	2.13	15.97	(0.21)	(1.40)	(4.73)
6A	(0.01)	2.01	0.77	(0.12)	(0.002)	N/A	1.46	0.76	8.10	2.81	<0.50	(1.30)	8.97
6B	(0.11)	3.49	(2.06)	(0.18)	(0.003)	N/A	(4.09)	1.12	9.96	2.67	(0.02)	(1.52)	(13.49)

N/A = Not applicable; potassium salts are used as fluxing agent.

Table 3.30. Average Anion Concentrations in Tank C-106 Sludge Samples - Periodic Water Replenishment Tests

Contact Stage	Fluoride	Acetate	Formate	Chloride	Nitrite	Nitrate	Carbonate	Sulfate	Oxalate	Phosphate
	--µg/g--									
Sample 404										
1	3.53E+01	<1.73E+02	<5.11E+01	8.26E+01	<5.71E+01	<5.48E+01	2.08E+04	<5.18E+01	2.59E+04	<8.38E+01
2	<1.48E+01	<1.73E+02	<5.11E+01	4.04E+01	<5.71E+01	<5.48E+01	<6.32E+03	<5.18E+01	1.71E+04	<6.39E+01
3	<1.48E+01	<1.73E+02	<5.11E+01	3.20E+01	<5.71E+01	<5.48E+01	<6.32E+03	<5.18E+01	1.24E+04	<6.39E+01
4	<1.48E+01	<1.73E+02	<5.11E+01	<3.16E+01	<5.71E+01	<5.48E+01	<6.32E+03	<5.18E+01	6.80E+03	<6.39E+01
5	<1.48E+01	<1.73E+02	<5.11E+01	<2.99E+01	<5.71E+01	<5.48E+01	<6.32E+03	<5.18E+01	4.01E+03	<6.39E+01
6A	<1.53E+01	3.14E+02	1.06E+02	<3.07E+01	1.05E+02	1.43E+02	<6.53E+03	<5.34E+01	4.01E+03	2.06E+02
6B	<1.43E+01	5.68E+02	2.50E+02	<2.88E+01	1.33E+02	1.34E+02	<6.12E+03	<5.01E+01	4.37E+03	<6.18E+01
Sample 405										
1	3.44E+01	<2.85E+02	<8.42E+01	9.87E+01	<9.40E+01	<9.02E+01	1.26E+04	<8.53E+01	3.92E+04	<1.05E+02
2	<2.44E+01	<2.85E+02	<8.42E+01	6.18E+01	<9.40E+01	<9.02E+01	<1.04E+04	<8.53E+01	2.29E+04	<1.05E+02
3	<2.44E+01	<2.85E+02	<8.42E+01	<5.00E+01	<9.40E+01	<9.02E+01	<1.04E+04	<8.53E+01	1.49E+04	<1.05E+02
4	<2.44E+01	<2.85E+02	<8.42E+01	<4.92E+01	<9.40E+01	<9.02E+01	<1.04E+04	<8.53E+01	6.12E+03	<1.05E+02
5	<2.44E+01	<2.85E+02	<8.42E+01	<5.82E+01	<9.40E+01	<9.02E+01	<1.04E+04	<8.53E+01	2.65E+03	<1.05E+02
6A	<2.29E+01	3.43E+02	1.82E+02	<4.60E+01	1.44E+02	2.25E+02	<9.79E+03	1.04E+02	3.60E+03	1.90E+02
6B	<2.59E+01	7.73E+02	3.66E+02	<5.19E+01	1.79E+02	2.24E+02	<1.11E+04	<9.04E+01	3.56E+03	<1.12E+02

Cr water leachability from the sludge is potentially important because it can be a major contributor to noncarcinogenic risk and the Hazard Index calculation. Cr was not detected above its EQL in any of the periodic water replenishment tests or the single-contact water leach tests (Table 3.19), except for the sample 405 single contact 1-month test where 0.39% of the Cr was water leachable. Cr in the sludge (estimated at <900 µg/g, Table 3.11) has very low water leachability.

3.4.2.5   Anion Concentrations – Replenishment Water Extracts

The leachable anion concentrations at the six stages of the periodic replenishment tests are listed in Table 3.30. The primary leachable anions at Stage 1 were oxalate and carbonate, with much less leachable quantities of chloride and fluoride. Oxalate continues to leach from the sludge samples in measurable quantities at all stages of the test; however, the amount of carbonate leached is below the detection limit after stage 1. (The detection limit for carbonate is high at a level of about 6,300 µg/g in sample 404 and 10,000 µg/g in sample 405.) The amount of oxalate leaching from the sludge decreases fairly uniformly throughout the six stages of leaching. The amount leached from sample 404 in Stage 1 was 25,900 µg/g and in Stage 6B, it was 4,370 µg/g. The amount leached from sample 405 in Stage 1 was 39,200 µg/g and in Stage 6B, it was 3,560 µg/g.

3.4.2.6   Radioanalytical Results – Periodic Replenishment Water Tests

The results of the GEA analysis of the periodic replenishment water tests for ^{60}Co and ^{137}Cs are listed in Table 3.31. ^{60}Co was not detected in any of the test samples. The water leachable concentrations of ^{137}Cs for the first contacts were 2.65 and 2.03 µCi/g for sludge samples 404 and 405, respectively. This represents about 2% of the total ^{137}Cs in the sludge samples (Table 3.8). The subsequent concentrations in each leachate decrease by over a factor of ten compared to the initial level. This shows that a small amount of the ^{137}Cs is very leachable, but the remaining majority is relatively recalcitrant to water leaching.

Table 3.31. GEA for Tank C-106 Periodic Water Replenishment Tests

Contact Stage	Contact Duration (days)	^{60}Co ----µCi/g-----	^{60}Co --µg/g-	^{137}Cs ----µCi/g--	^{137}Cs ------µg/g---
		Sample 404			
1	1	<0.0177	<1.61E-05	2.65	3.044E-02
2	1	<0.0177	<1.61E-05	0.128	1.471E-03
3	4	<0.0177	<1.61E-05	0.0389	4.473E-04
4	1	<0.0177	<1.61E-05	0.0244	2.806E-04
5	1	<0.0177	<1.61E-05	0.0244	2.806E-04
6A	43	NA	NA	NA	NA
6B	82	NA	NA	NA	NA
		Sample 405			
1	1	<0.0292	<2.65E-05	2.03	2.329E-02
2	1	<0.0292	<2.65E-05	0.0402	4.623E-04
3	4	<0.0292	<2.65E-05	0.0402	4.623E-04
4	1	<0.0292	<2.65E-05	0.0402	4.623E-04
5	1	<0.0292	<2.65E-05	0.0402	4.623E-04
6A	43	NA	NA	NA	NA
6B	82	NA	NA	NA	NA
NA = Not analyzed.					

Total beta, total alpha and ^{90}Sr levels for the periodic replenishment tests are listed in Table 3.32. These results show that, for each of these constituents, the values start out relatively low in the first extraction and then increase by a factor of two of more by the fifth extraction. For example, the total beta in sample 404 starts at 99.7 µCi/g and increases to 200 µCi/g by Stage 5, and total alpha in sample 405 increases from 0.287 to 0.843 µCi/g over the five stages.

The ^{239}Pu, ^{237}Np, and ^{241}Am leachable concentrations for the replenishment tests are listed in Table 3.33, and the leachable percentages are shown in Table 3.34. ^{239}Pu was not measured above its EQL during any of the stages of leaching. The estimated extent of ^{239}Pu leaching was in the range of 0.01 to 0.04% of the total ^{239}Pu in the sludge. Measurable quantities of leachable ^{237}Np were measured at most stages of the test. The highest amounts (8.5% and 9.8%) were leached during Stage 1 with decreasing percentages to Stage 5 values of approximately 0.1%. The leachability of ^{237}Np increased to about 1% for the longer-term contact Stages 6A and 6B. ^{241}Am was only measured above its EQL during the first stage of leaching of sample 404. The percentage leached for this stage was 0.04%. Similar low percentages of leaching were estimated for ^{241}Am for the other stages.

Table 3.32.   Total Beta, Total Alpha, and ^{90}Sr Analysis for Tank C-106 Periodic Water Replenishment Tests

Contact Stage	Total Beta	Total Alpha	^{90}Sr	
	----------------µCi/g---------------		-------µCi/g-------	------µg/g------
Sample 404				
1	99.7	0.168	47.1	0.337
2	160	0.380	77.3	0.552
3	202	0.576	105	0.753
4	192	0.517	95.9	0.685
5	200	0.465	95.3	0.681
6a	NA	NA	NA	NA
6b	NA	NA	NA	NA
Sample 405				
1	175	0.287	85.3	0.609
2	262	0.627	141	1.01
3	317	0.869	155	1.11
4	312	0.719	151	1.08
5	341	0.843	180	1.29
6a	NA	NA	NA	NA
6b	NA	NA	NA	NA
NA = Not analyzed.				

Table 3.33. Actinide Analysis for Tank C-106 Periodic Water Replenishment Tests

Contact Stage	^{239}Pu --µCi/g--	^{239}Pu --µg/g--	^{237}Np --µCi/g--	^{237}Np --µg/g--	^{241}Am --µCi/g--	^{241}Am --µg/g--
			Sample 404			
1	(3.81E-04)	(6.14E-03)	5.93E-05	8.35E-02	2.57E-03	7.55E-04
2	(1.32E-04)	(2.13E-03)	2.53E-05	3.57E-02	(9.86E-04)	(2.90E-04)
3	(1.05E-04)	(1.69E-03)	1.75E-05	2.47E-02	<2.15E-03	<6.32E-04
4	(1.40E-04)	(2.25E-03)	1.01E-05	1.43E-02	<2.15E-03	<6.32E-04
5	(1.01E-04)	(1.63E-03)	1.03E-05	1.45E-02	<2.15E-03	<6.32E-04
6A	(1.30E-04)	(2.09E-03)	7.37E-05	1.04E-01	<2.22E-03	<6.53E-04
6B	(5.31E-05)	(8.57E-04)	6.93E-05	9.76E-02	<2.08E-03	<6.12E-04
			Sample 405			
1	(7.31E-04)	(1.18E-02)	8.07E-05	1.14E-01	(1.92E-03)	(5.65E-04)
2	(5.02E-04)	(8.09E-03)	3.14E-05	4.42E-02	(2.81E-03)	(8.27E-04)
3	(4.63E-04)	(7.47E-03)	1.88E-05	2.65E-02	<3.54E-03	<1.04E-03
4	(2.57E-04)	(4.15E-03)	9.49E-06	1.34E-02	<3.54E-03	<1.04E-03
5	(1.65E-04)	(2.66E-03)	(6.92E-06)	(9.75E-03)	<3.54E-03	<1.04E-03
6A	(2.60E-04)	(4.20E-03)	7.85E-05	1.11E-01	<3.76E-03	<1.11E-03
6B	(1.27E-04)	(2.06E-03)	8.24E-05	1.16E-01	<3.33E-03	<9.79E-04

Table 3.34. Water-Leachable Percentage for Actinides in Tank C-106 Periodic Water Replenishment Tests Compared with Fusion Analysis

Contact Stage	^{239}Pu	^{237}Np	^{241}Am
	-------------% Water Leachable-------------		
	Sample 404		
1	(0.02)	8.5	0.04
2	(0.01)	3.4	(0.02)
3	(0.01)	0.2	<0.04
4	(0.01)	0.1	<0.04
5	(0.01)	0.1	<0.04
6A	(0.01)	1.0	<0.04
6B	(0.003)	1.0	<0.03
	Sample 405		
1	(0.04)	9.8	(0.03)
2	(0.03)	3.9	(0.04)
3	(0.02)	0.3	<0.06
4	(0.01)	0.1	<0.06
5	(0.01)	(0.07)	<0.06
6A	(0.01)	1.1	<0.06
6B	(0.01)	1.0	<0.05

3.5   XRD Results

This section discusses the crystalline solids identified in the XRD patterns for the unleached, 1-month and 82-day water-leached, and HF Stage 1 sludge samples 404 and 405. The as-measured and background-subtracted XRD patterns for these samples are found in Appendix A. Phase identification was based on a comparison of the peak reflections and intensities observed in each pattern to the mineral powder diffraction files (PDF™) published by the Joint Committee on Powder Diffraction Standards (JCPDS) International Center for Diffraction Data (ICDD). Phase identification from the XRD patterns was refined in an iterative fashion by considering phases with particle compositions that were determined by SEM/EDS (see Section 3.6) as present in the sludge samples. The XRD patterns in Appendix A show greater detail than those plotted in this section because they do not include the schematic PDF-XRD database patterns (as plotted in this section) used for phase identification.

Each pattern in this section and Appendix A is shown as a function of degrees 2θ based on $Cu_{K\alpha}$ radiation (λ=1.5406 Å). The vertical axis in each pattern represents the intensity or relative intensity of the XRD peaks. The XRD patterns included in this section show, for comparison purposes, one or more schematic database (PDF) patterns considered for phase identification. The height of each line in the schematic PDF patterns represents the relative intensity of an XRD peak (i.e., the most intense [the highest] peak has a relative intensity [I/I_o] of 100%). Quantitative analyses of the relative masses of individual phases present in each solid sample were not estimated using these XRD patterns due to the factors discussed at the end of Section 2.3. Also, as noted previously in Section 2.3, a crystalline phase typically must be present at greater than 5 wt% of the total sample mass (greater than 1 wt% under optimum conditions) to be readily detected by XRD.

The background subtracted XRD patterns for the unleached, 1-month and 82-day water leached, and HF-extracted sludge samples 404 and 405 from tank C-106 are plotted with the PDF patterns used for phase identification in Figure 3.3 through Figure 3.6, respectively. The XRD results indicate that the unleached, 1-month and 82-day water-leached, and HF-extracted sludge samples contain detectable quantities of the following crystalline phases based on the indicated PDF database patterns:

- gibbsite ─ $Al(OH)_3$ (PDF 33-0018)
- dawsonite ─ $NaAlCO_3(OH)_2$ (PDF 45-1359)
- hematite ─ Fe_2O_3 (PDF 86-0550)
- böhmite ─ $AlO(OH)$ (PDF 83-1505)
- rhodochrosite ─ $MnCO_3$ (PDF 83-1763)
- whewellite ─ Ca oxalate monohydrate, $CaC_2O_4 \cdot H_2O$ (PDF 20-0231)

The XRD patterns were also consistent with the possible presence of an Ag-Hg phase (PDF 43-1465). Its identification, however, was based on a match to a single reflection at 37.153 °2θ, which is insufficient for a conclusive identification of any phase by XRD. However, the search of the XRD patterns for the possible presence of Ag-Hg phases was instigated by the discovery of Ag-Hg particles by SEM/EDS analysis of the sludge samples (see Section 3.6). It was not possible to further validate its presence in these samples by XRD.

The XRD patterns also show that lindbergite (Mn oxalate dihydrate, $MnC_2O_4 \cdot 2H_2O$) (PDF 25-0544) is also present in the unleached sludge samples 404 and 405 from tank C-106. The major reflections for lindbergite (PDF 25-0544), including the split peak at 18.4°θ that is due to the overlap of the 100% peaks for gibbsite and lindbergite (see red arrow in Figure 3.3), are absent in the XRD patterns for the 1-month and 82-day water-leached and HF-extracted sludge samples. The split peak at 18.4°θ is more apparent in the XRD patterns for the unleached samples in Appendix A.

There were no unassigned reflections in the background-subtracted XRD patterns for the samples of unleached, 1-month and 82-day water-leached, and HF-extracted sludge samples 404 and 405. This suggests that all crystalline phases present at greater than 5-10 wt% in these sludge samples were identified by XRD. Based qualitatively on the observed relative heights for peaks in the background subtracted XRD patterns (Figure 3.3 through Figure 3.6), gibbsite, dawsonite, and rhodochrosite appear to be the most dominant crystalline phases present in the unleached, 1-month and 82-day water-leached, and HF-extracted sludge samples 404 and 405.
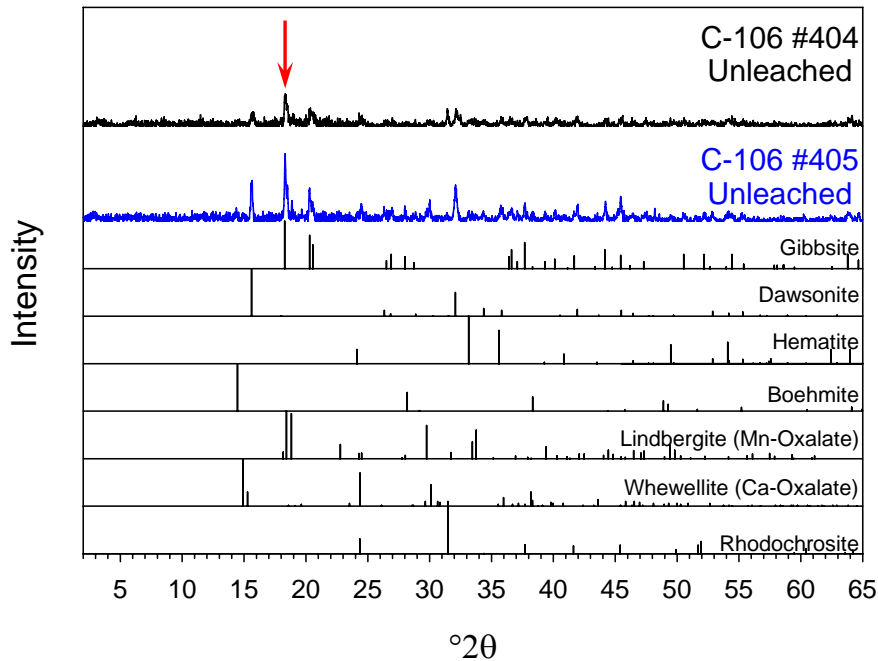
Figure 3.3. Background-Subtracted XRD Pattern for the Unleached Sludge Samples 404 and 405 from Tank C-106 Shown with Matching PDF Database Patterns
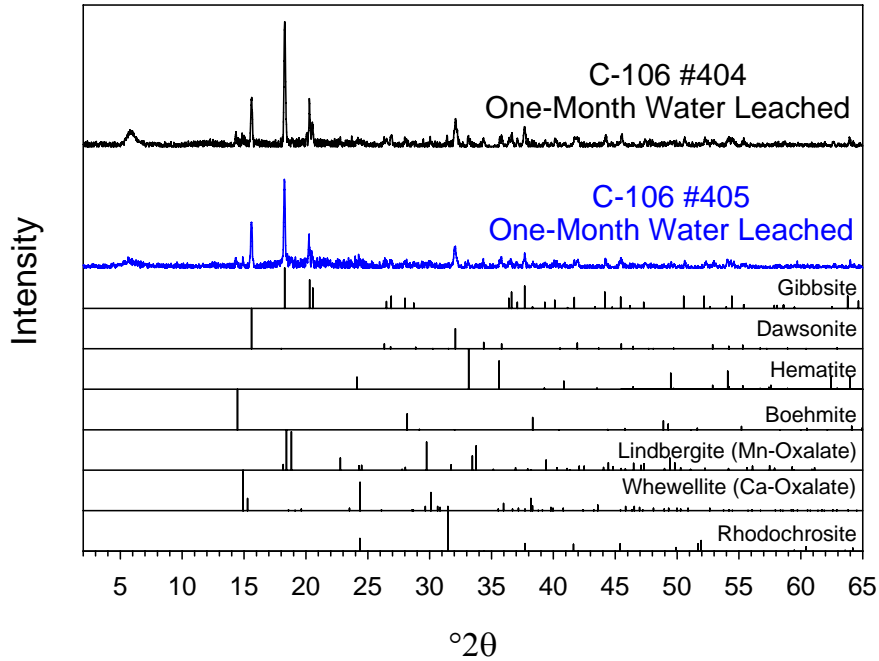
Figure 3.4. Background-Subtracted XRD Pattern for the 1-Month Water-Leached Sludge Samples 404 and 405 from Tank C-106 Shown with Matching PDF Database Patterns
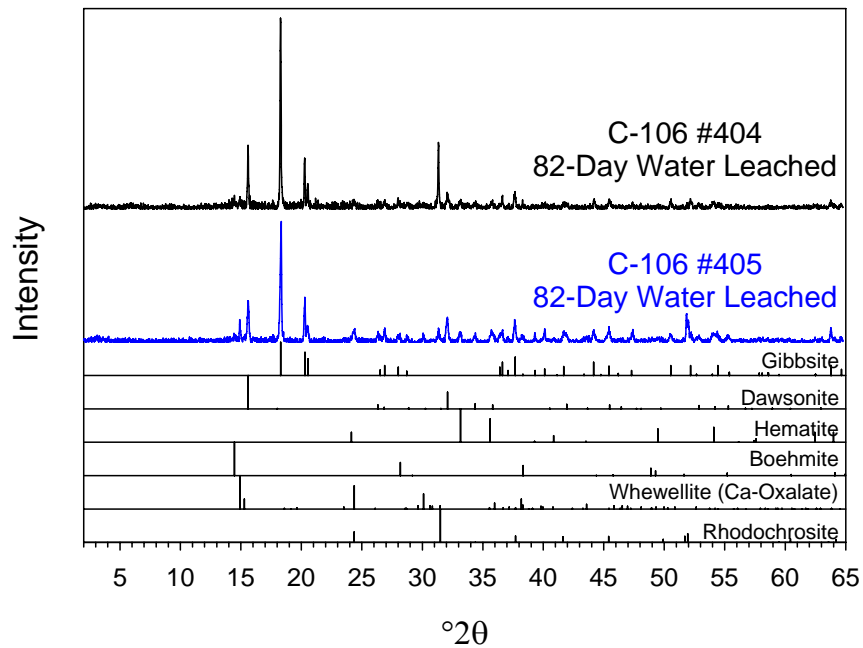
Figure 3.5. Background-Subtracted XRD Pattern for the 82-Day Water-Leached Sludge Samples 404 and 405 from Tank C-106 Shown with Matching PDF Database Patterns
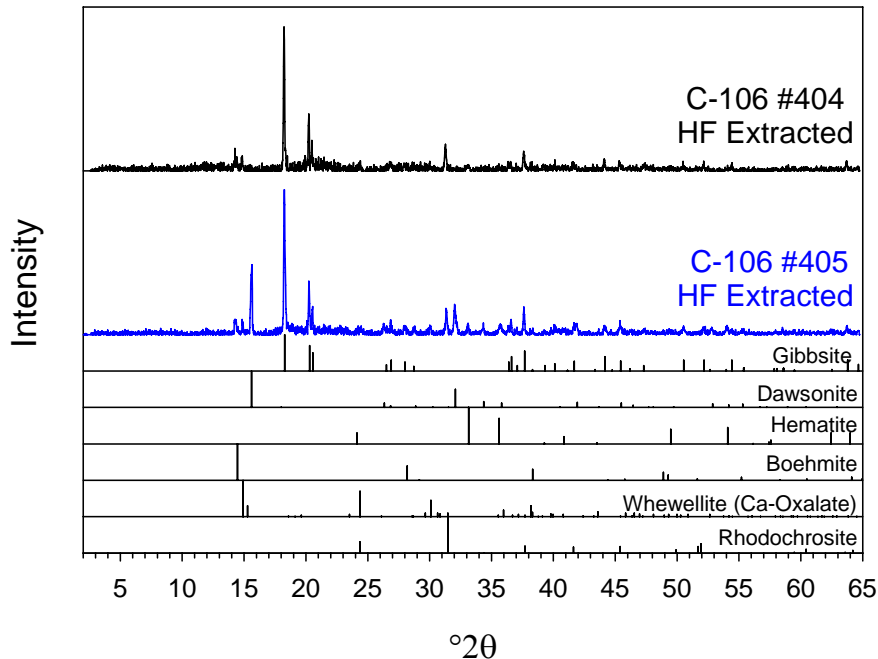
Figure 3.6. Background-Subtracted XRD Pattern for the HF-Extracted Sludge Samples 404 and 405 from Tank C-106 Shown with Matching PDF Database Patterns

The background-subtracted XRD patterns for the unleached, water leached, and HF-extracted sludge samples 404 and 405 from tank C-106 are generally very similar to each other. For example, Figure 3.7 shows a comparison of the XRD patterns for the unleached, 1-month and 82-day water-leached, and HF-extracted sample 405. Besides the presence of reflections for lindbergite (Mn oxalate dihydrate, $MnC_2O_4 \cdot 2H_2O$) in the unleached tank C-106 samples, only two other minor differences were observed between the XRD patterns for the four samples of tank C-106 sludge. One obvious difference is the low peak intensities (i.e., peak heights) observed in the background-subtracted XRD patterns for the unleached samples relative to those in the patterns for the water-leached and HF-extracted samples. If the same amount of solid sludge material was used for each of the four sample types, then it can be assumed from the low peak intensities that the amount of amorphous relative to crystalline material is greater in the unleached samples than in the water leached and HF-extracted sludge samples 404 and 405 from tank C-106. Because of the hazardous nature of these sludge samples, it is not possible to control the exact amount of material used for each XRD mount. However, when these XRD mounts are prepared, a concerted effort is made to follow the same procedure and use the same amount of sludge material as closely as possible.
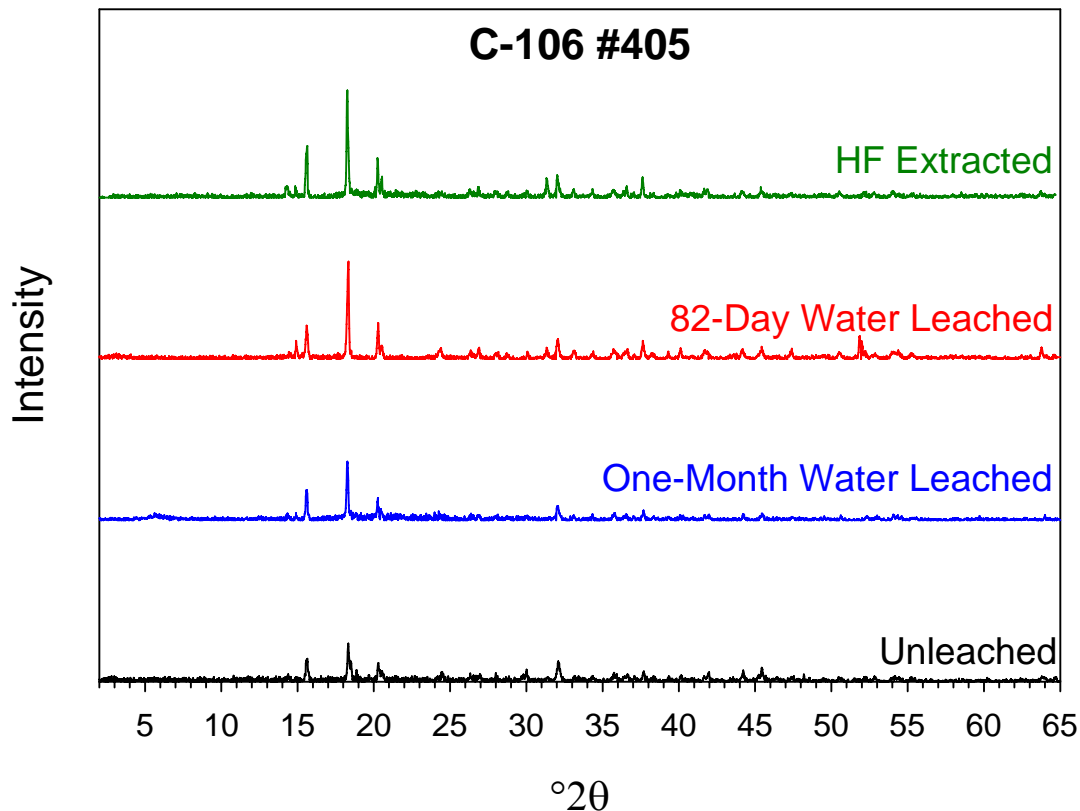
Figure 3.7. Comparison of Background-Subtracted XRD Pattern for Unleached, 1-Month and 82-Day Water-Leached, and HF-Extracted Sludge Sample 405 from Tank C-106 Shown with Matching PDF Database Patterns

The other difference is the presence of a small, broad peak at approximately 5.9°θ (15 Å) that only exists in the patterns for the 1-month water-leached sludge samples 404 and 405 from tank C-106 (Figure 3.4). This peak is likely due to reflections from the Kapton® film used in the specimen holder described in Section 2.3, and has occasionally been observed in other patterns where samples were mounted in this type of specimen holder.

Other minor differences include the absence of major reflections for rhodochrosite, such as the major peak at 31.36 °2θ, in the XRD pattern for the 1-month water-leached 405 sample, and for dawsonite in the XRD pattern for the HF-extracted 404 sample. Because these phases were identified in the other patterns for sludge samples 404 and 405, it is likely that rhodochrosite and dawsonite were also present in these two particular samples but at levels near or below the limits of detection by XRD.

3.6   SEM/EDS Results

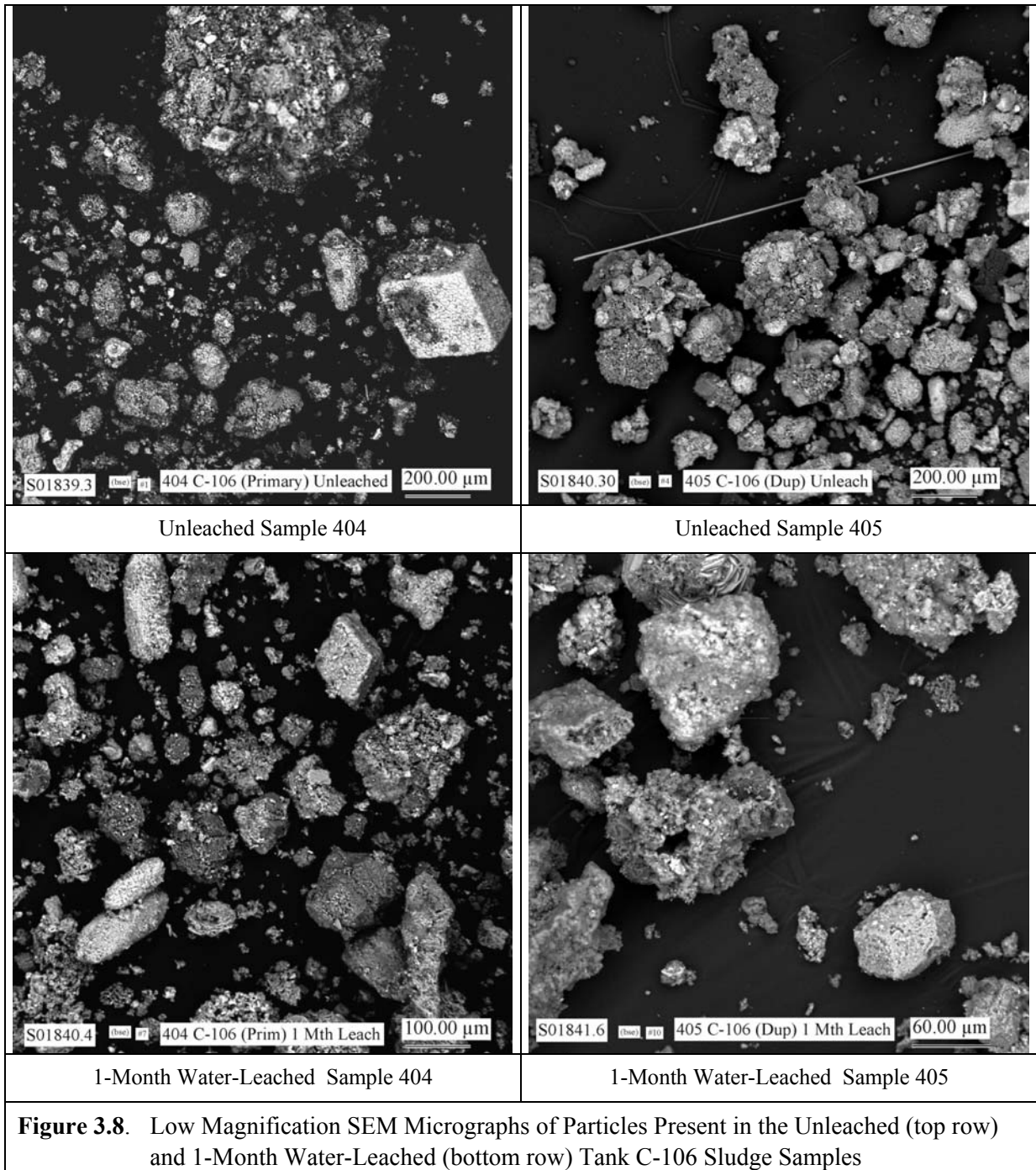This section discusses the results of the SEM/EDS analyses for the unleached (as-received), 1-month and 82-day water-leached, and HF-extracted sludge samples 404 and 405 from tank C-106. The SEM micrographs presented in this section show representative morphologies, sizes, and surface textures of
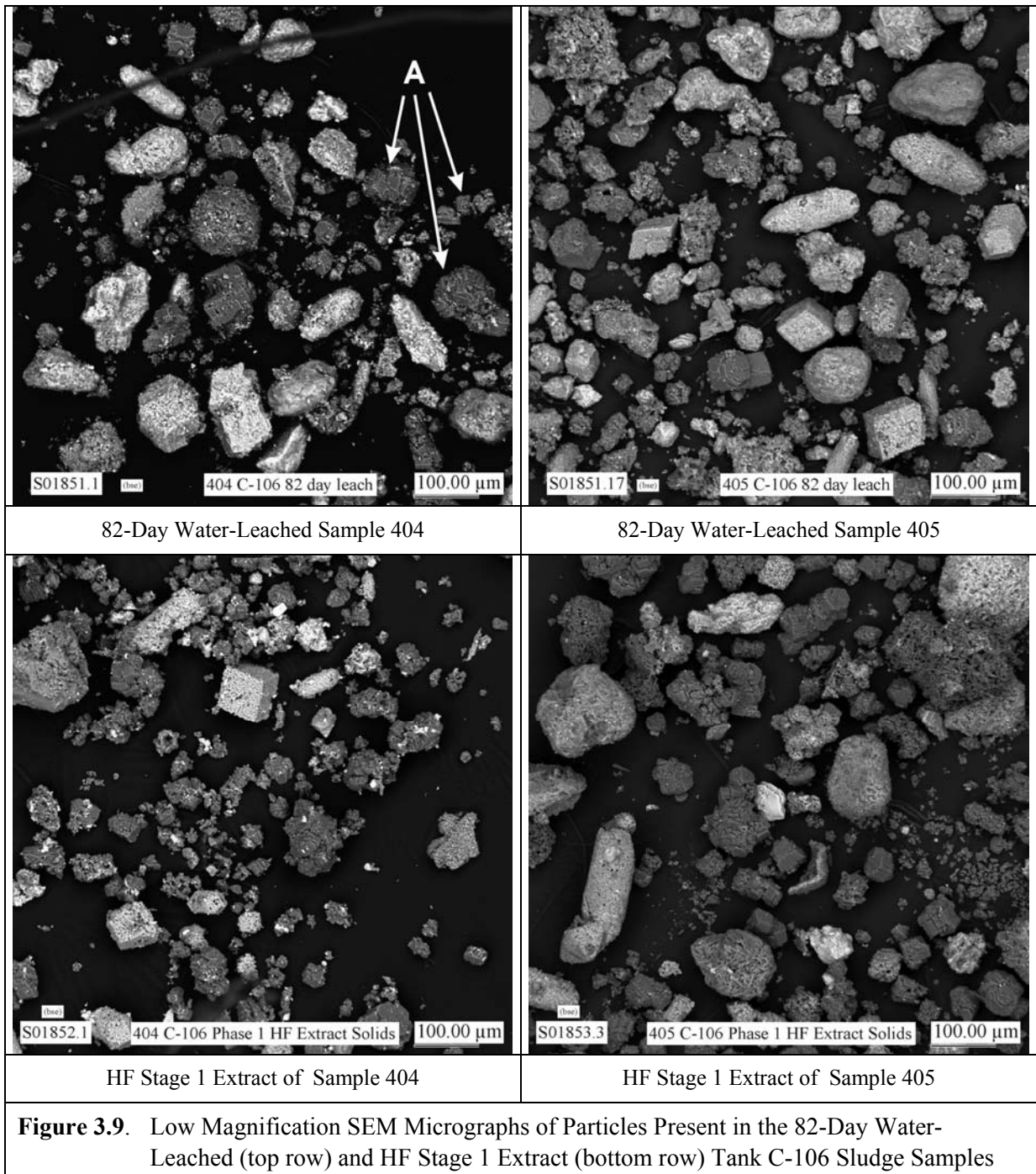
particles in each of these samples. After the studies and analyses of tank C-106 sludge had been completed and presented in draft form, the instrument hardware and software for the JEOL JSM-840 SEM were upgraded to the INCAEnergy EDS System to automate the collection of EDS spectra over multi-micrometer-sized areas of an SEM-imaged sample. This new capability was demonstrated using existing SEM mounts of the tank C-106 sludge samples. All of the SEM micrographs and EDS spectra determined for sludge samples 404 and 405 prior to the installation of the INCAEnergy EDS System are shown without discussion or interpretation in Appendices B (unleached [as received] sludge, C (1-month and 82-day water-leached sludge), and D (HF Stage 1 extracted). Three sets of SEM micrographs and element distribution maps collected with the INCAEnergy EDS System are presented at the end of this section. The remainder of the SEM micrographs and element distribution maps determined using this system upgrade is shown in Appendix E.

Each micrograph included in this section before Figure 3.18 shows the name of the SEM digital image file, sample identification number, and a size scale bar, respectively, at the bottom left, center, and right of each image. Micrographs labeled with "BSE" near the digital image file name indicate that the micrograph was collected with backscattered electrons. Areas labeled by "eds" identify locations of particles for which EDS spectra were recorded and presented in the appendices. The micrographs presented in this section are typically reproduced at reduced size to conserve page space. To get a more detailed view of these micrographs, the reader is referred to Appendices B through D, where the micrographs are shown at a larger size.

Figure 3.8 and Figure 3.9 show SEM micrographs obtained at relatively low magnification of material present in the unleached (top row in Figure 3.8), 1-month water-leached (bottom row in Figure 3.8), 82-day water-leached (top row in Figure 3.9), and HF-extracted (bottom row in Figure 3.9) sludge samples. The SEM micrographs for the sludge samples 404 and 405 are presented in the left and right columns, respectively, in both Figure 3.8 and Figure 3.9. The sludge particles range in size from a few hundred micrometers to submicrometer in size, and occur often as aggregates of one or more compositionally distinct phases. Generally, there are no obvious major differences visible in the morphologies and distributions of particles in the SEM images for the same treatment (or lack of) of 404 and 405 samples, or between the unleached, water-leached, or HF extract samples. Although very subjective, it appears that with extended water leaching, especially after HF extraction, the samples may possibly contain fewer particle aggregates and an increased relative mass of dark gray particles having multi-faceted, blocky shapes (see particles labeled A in the SEM micrograph in the top left of Figure 3.9). The surfaces of these blocky dark gray particles are also typically free of coatings or other sorbed particles, and do not show any obvious dissolution features regardless of their treatment. As discussed below, these multi-faceted, blocky dark gray particles primarily contain Al-O±H or Al-Na-O-C±H.

Figure 3.10 shows several large (tens of micrometers or larger on edge) particles that have the same "composition" as determined by EDS but at least four or five distinguishing morphologies. These particle shapes were common in all tank C-106 samples inspected by SEM, and can be seen in Figure 3.8 and Figure 3.9. The EDS analyses of these particles indicate the presence of Mn, Al, Fe, Na, P, Si, O, and possibly C and H (listed generally in decreasing order of peak heights of metal elements in the EDS spectra). The particles include morphologies that are oblong and rounded (micrograph A), orthorhombic (90° angles) (micrograph B), rhombohedral (oblique angles) (micrograph C), and pyramidal rhombic

3.37

S01839.3 (bse) #1 404 C-106 (Primary) Unleached 200.00 µm	S01840.30 (bse) #4 405 C-106 (Dup) Unleach 200.00 µm
Unleached Sample 404	Unleached Sample 405
S01840.4 (bse) #7 404 C-106 (Prim) 1 Mth Leach 100.00 µm	S01841.6 (bse) #10 405 C-106 (Dup) 1 Mth Leach 60.00 µm
1-Month Water-Leached Sample 404	1-Month Water-Leached Sample 405

Figure 3.8. Low Magnification SEM Micrographs of Particles Present in the Unleached (top row) and 1-Month Water-Leached (bottom row) Tank C-106 Sludge Samples

82-Day Water-Leached Sample 404	82-Day Water-Leached Sample 405
HF Stage 1 Extract of Sample 404	HF Stage 1 Extract of Sample 405

Figure 3.9. Low Magnification SEM Micrographs of Particles Present in the 82-Day Water-Leached (top row) and HF Stage 1 Extract (bottom row) Tank C-106 Sludge Samples

A	B	C
Sample 404 1-MonthWater Leach	Sample 405 Unleached	Sample 404 HF Stage 1 Extract

D	E
Sample 404 82-DayWater Leach	Sample 405 HF Stage 1 Extract

Figure 3.10.   Unique Morphologies of Large Particles Common in all Tank C-106 Samples Inspected by SEM/EDS (Note: all consist of Al, Ca, Fe, Mn, Na, O, P, Si, and possibly C and H)

(micrograph D) in Figure 3.10. Another particle shape having this composition and common in these samples is shown at the top of micrograph E in Figure 3.10. The particle shape in micrograph D is likely an elongated form of that in micrograph E, where the particle faces labeled A, B, and C in micrograph D are equivalent, respectively, to those in micrograph E.

It is not known if the differently shaped particles in Figure 3.10 are related crystallographically to each other, or if these particles consist entirely of a single phase (or phase assemblage) or are just coated with a phase(s) consisting of Mn, Al, Fe, Na, P, Si, O, and possibly C and H. Given the distinctive geometric shapes of these particles, some phase at the core of these particles is likely controlling the external morphologies of these grains. The SEM images in micrographs A and B in Figure 3.11 suggest that these particles may have a coating of this bulk composition. This conclusion is further supported in other SEM images that show surface cavities containing "darker" particles (usually Al-rich) that are different in texture and composition from the surface material, such as the areas labeled D in

micrograph D in Figure 3.10 and B in micrograph C in Figure 3.11. The surface textures of the particles in micrographs C and D in Figure 3.11 also suggest that the material at the surfaces of these particles consists of more than one phase. The EDS spectra (Figure 3.12) for the HF extract samples typically indicate the particle surfaces have become deficient in Al, Na, P, Si, and Ca relative to Mn and Fe, when compared to the compositions of the unleached samples. These particles exist primarily as large isolated grains, and are rarely observed to be intergrown, except for a few rare instances such as those shown in micrographs C (particles labeled A) and D (particles labeled C) in Figure 3.11. Unfortunately, the results of our studies did not provide any additional data that would allow further identification of these particles or explain the conditions of their existence.

Figure 3.13 through Figure 3.17 show examples of other particles typically present in the unleached, 1-month and 82-day water-leached, and HF-extracted Stage 1 sludge samples 404 and 405. The EDS analyses indicate the following compositions for particles in these samples ("±" symbol indicates the following element was possibly present):

- Mn-Al-Fe-Na-P-Si-Ca-O±C±H – see Figure 3.10, Figure 3.11, Figure 3.13(B), Figure 3.15(A) and (D), Figure 3.16(A)
- Al-O±H – see Figure 3.13(A) and (B), Figure 3.15(A)
- Ag-Hg±O±H – see Figure 3.13(A), Figure 3.15(A) and (B)
- Ca-O±C±H – see Figure 3.13(C) and Figure 3.16(B)
- Mn-O-C±H – possibly two different phases based on morphology, see Figure 3.14(A) and (B)
- Al-Na-O-C±H – see Figure 3.13(C) and (D), Figure 3.15 (B), (C), and (D)
- Mn-O-P±Al±C±H – see Figure 3.14(C) and Figure 3.15(D)
- Si-Al-Na-O±C±H – see Figure 3.15(A)
- Fe-Cr-O±C±H – see Figure 3.16(A)
- Fe-Mn-O±C±H - see Figure 3.16(D) (rare morphology) and Figure 3.17
- Rare earth element (REE)-rich – see Figure 3.16(C) (rare but present in most samples)
- Ca-Si-Al-O±C±H – see Figure 3.13(C) (rare)

The SEM/EDS analyses did not indicate the presence of I, Tc, or U in any of particles present in the unleached, 1-month and 82-day water-leached, and HF-extracted sludge samples 404 and 405 from tank C-106. This was due to their low concentrations in the sludge and the high detection limits for this analytical method.

Some unusual needle-like particles were observed in some micrographs (e.g., Figure 3.8 [upper right], Figure 3.11[D], and Figure 3.13[B]), especially for samples of the unleached tank C-106 sludge. These needle-like particles had clean shafts, and when not broken, terminated in a fine, sharp point at one end. They ranged up to approximately 1 mm in length. The EDS analyses indicated that these needle-like particles consisted primarily of O and Si and with lesser amounts of Ca, Na, Al, and possibly Mg (listed in estimated order of abundance and common components in manufactured glass products). These needle-like particles are probably from fibrous glass wool that was used in the laboratory to prevent dispersion of these radioactive powders during the drying of wet sludge samples. They are not present in the residual waste in the tank.
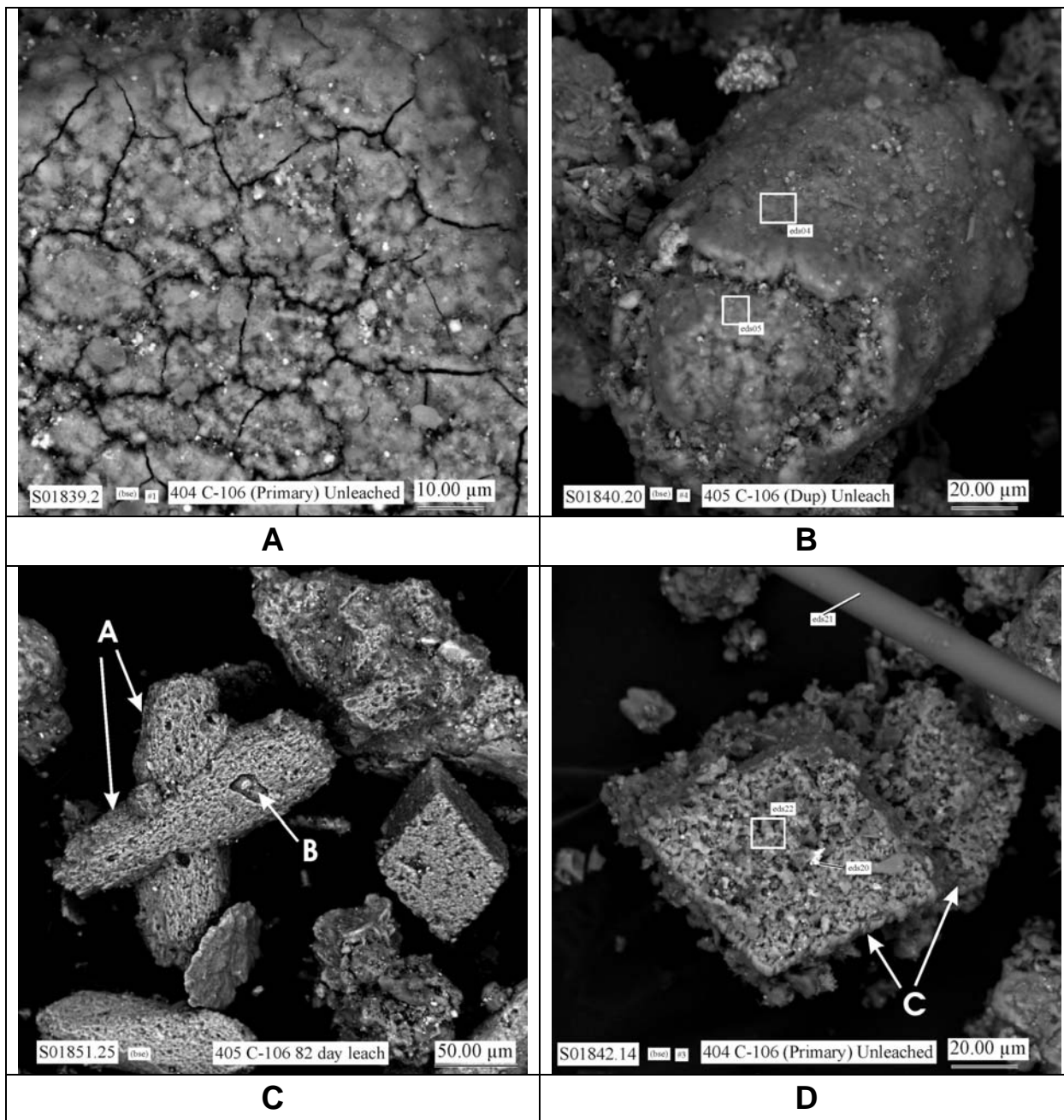
Figure 3.11.   Additional Examples of Large Particles with Unique Morphologies Shown in Figure 3.10 and Consisting of Mn, Al, Fe, Na, P, Si, Ca, O, and Possibly C and H
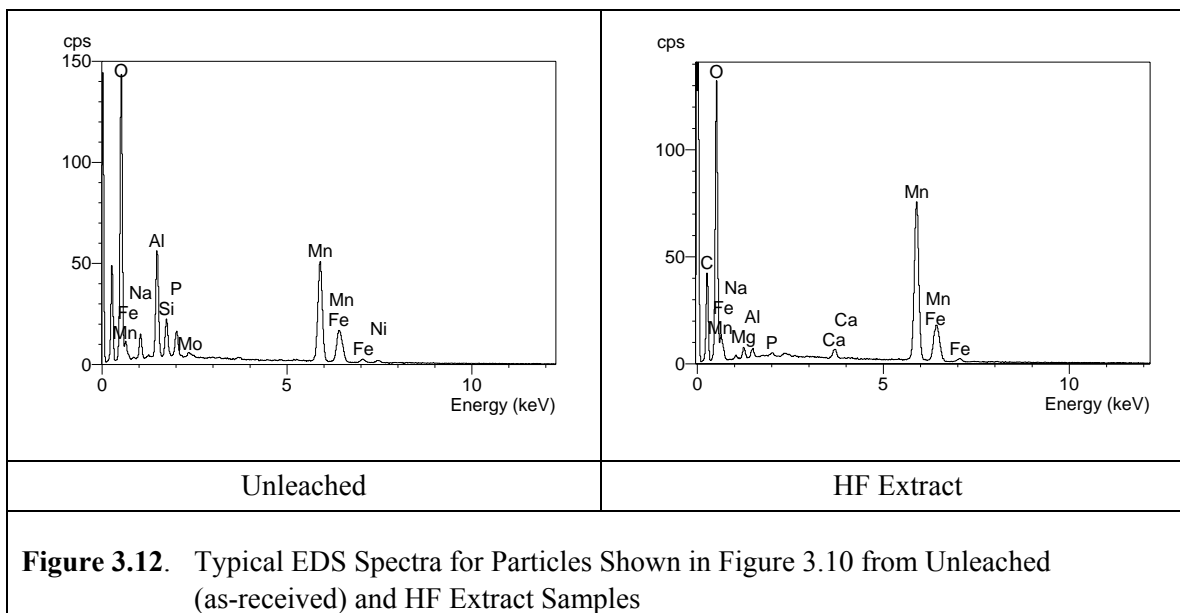
Figure 3.12.   Typical EDS Spectra for Particles Shown in Figure 3.10 from Unleached
(as-received) and HF Extract Samples

As noted in Section 3.1, Mn is a major component of sludge samples 404 and 405 from tank C-106. This is supported by the SEM/EDS analyses which show that four phases, two of which are common, in these samples contain Mn as the main element component. These include the phases consisting of Mn-Al-Fe-Na-P-Si-Ca-O±C±H, Mn-O-C±H, and Mn-O-P±Al±C±H. Based on morphology, the SEM analyses suggest that there may possibly be two different phases consisting of Mn-O-C±H. These phases include the blocky rhombohedral-like particles shown in Figure 3.14(A), and the cracked, blade-like particles in Figure 3.14(B). The latter form of particles was only observed in the unleached sludge samples 404 and 405.

The bright particle aggregates shown in Figure 3.13(A) and Figure 3.15(A) contained primarily Ag, Hg, O, and possibly C. These Ag-Hg particle aggregates were common in all of the unleached, 1-month and 82-day water-leached, and HF-extracted sludge samples 404 and 405. Although some of these aggregates were several tens of micrometers in size, many micrometer-to-submicrometer sized Ag-Hg particles also existed, such as the bright (white) "specks" observable in most of the micrographs shown in Figure 3.13 through Figure 3.16.

The SEM/EDS analyses also indicated the presence of numerous particles and particle aggregates containing Al oxide/hydroxide[a] in the unleached, 1-month and 82-day water-leached, and HF-extracted tank C-106 sludge samples. These multi-faceted, blocky particles appear dark gray in the SEM micrographs and were exceptionally common in the samples, especially the HF-extracted materials.

(a) Note that hydrogen (H) cannot be detected by EDS analysis and is, therefore, listed as possibly present in the compositions shown in the SEM micrographs.

3.43

Figure 3.13. Representative SEM Micrograph Images of Particles Identified in Unleached Sludge Samples 404 and 405 from Tank C-106
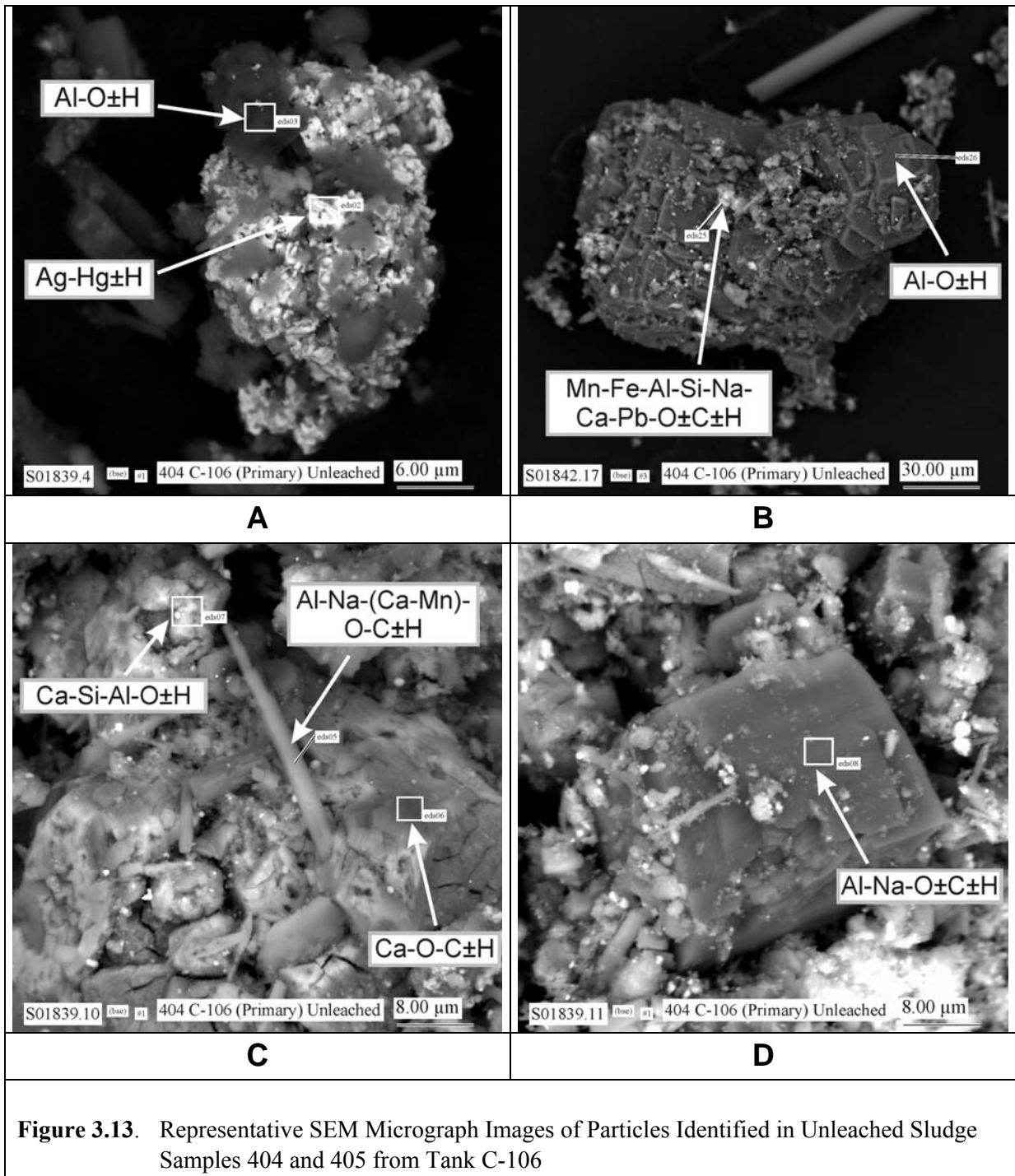
Figure 3.14. Representative SEM Micrograph Images of Particles Identified in Unleached Sludge Samples 404 and 405 from Tank C-106

Figure 3.15. Representative SEM Micrograph Images of Particles Identified in 1-Month Water-Leached Sludge Samples 404 and 405 from Tank C-106
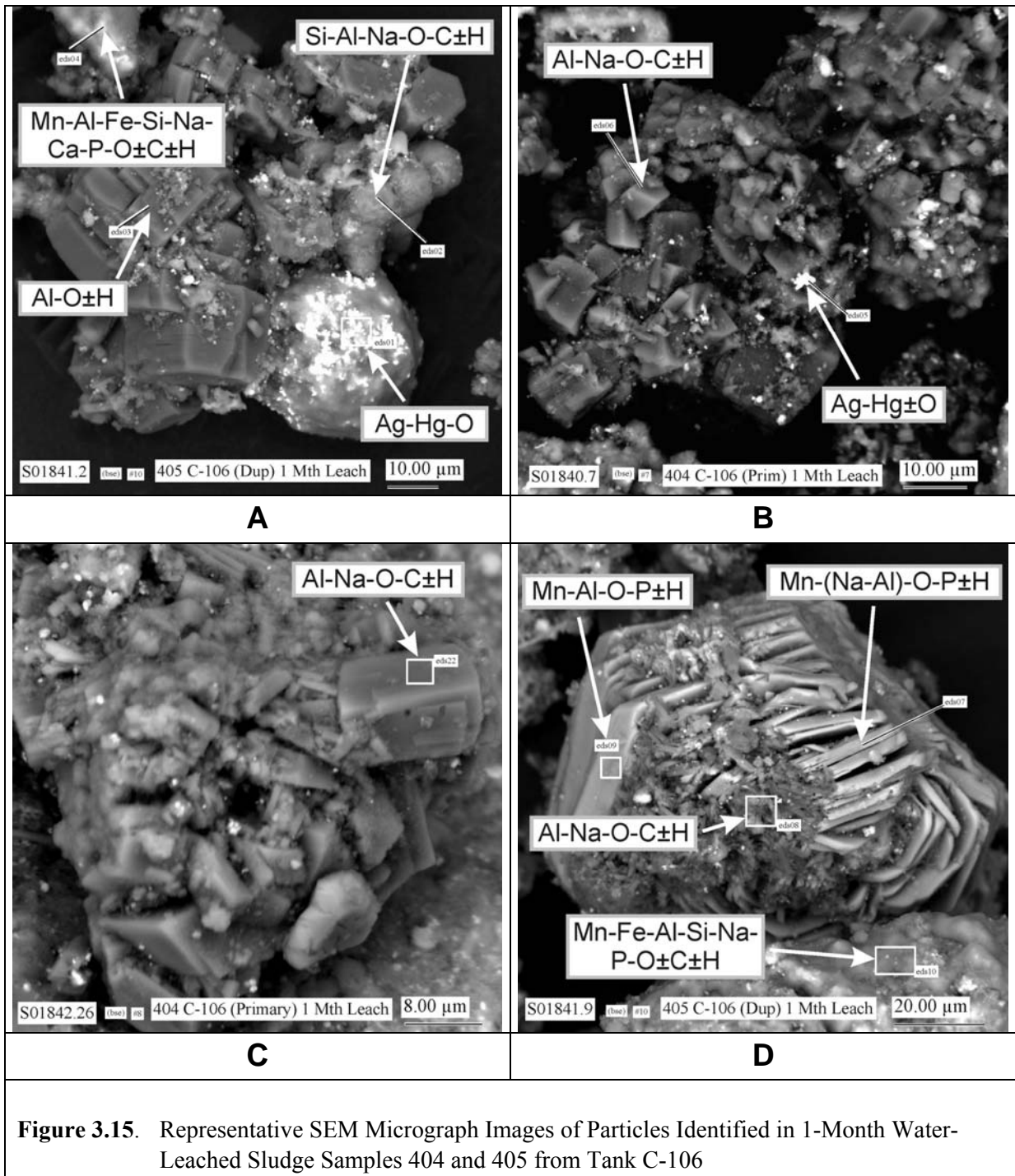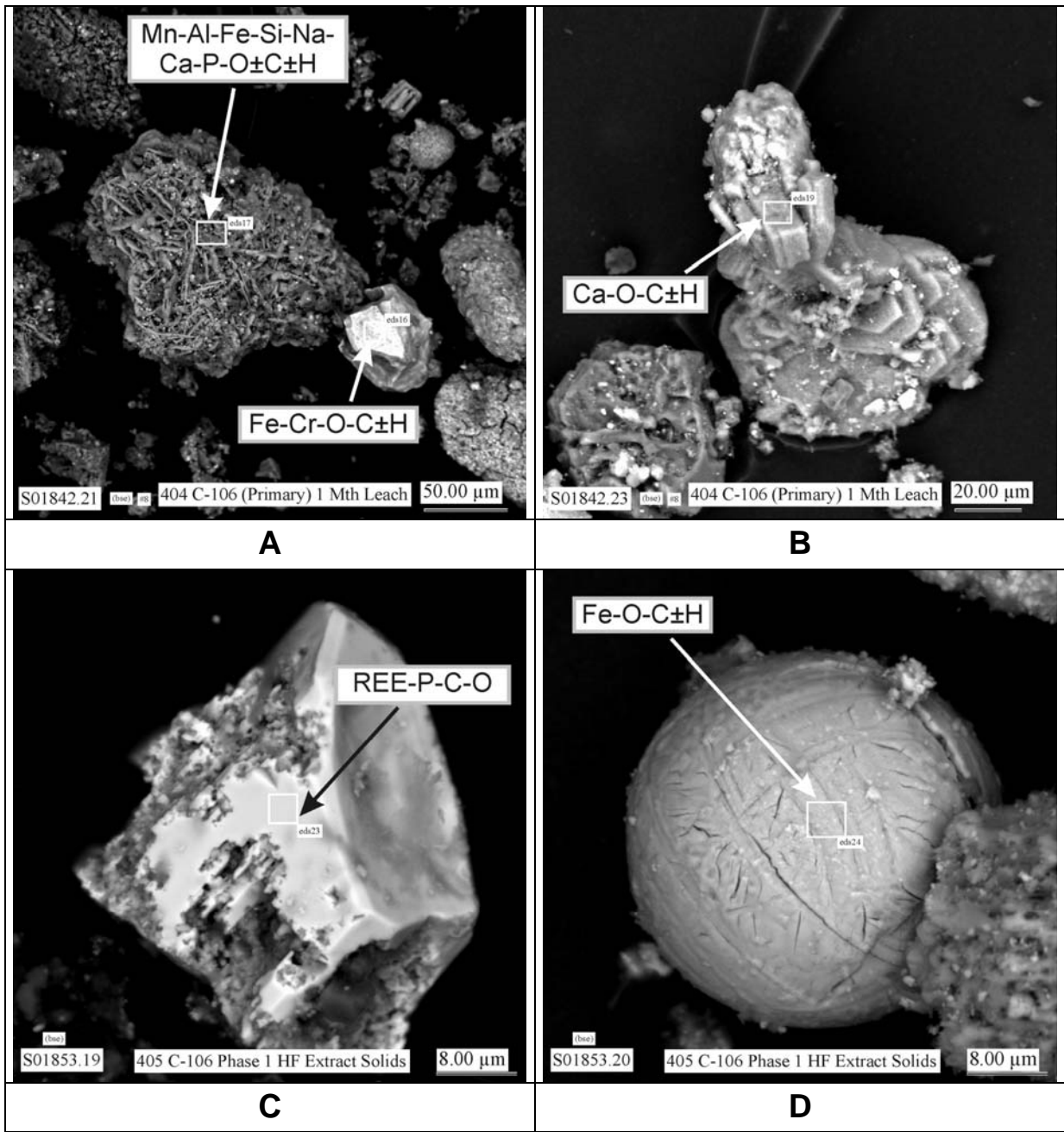
Figure 3.16. SEM Micrograph Images of Particles Identified in 1-Month Water-Leached and HF-Extracted Sludge Samples 404 and 405 from Tank C-106

Although less common than the particle compositions discussed in the preceding paragraphs, some relatively large (tens of micrometers) particles consisting primarily of Fe, O, possibly H, and often with Mn occur in the unleached, 1-month and 82-day water-leached, and HF-extracted tank C-106 sludge samples. When Cr and Ni were detected by EDS, these elements were always associated with these Fe oxides. Examples of these particles are shown in Figure 3.17. The surfaces of these particles do not exhibit 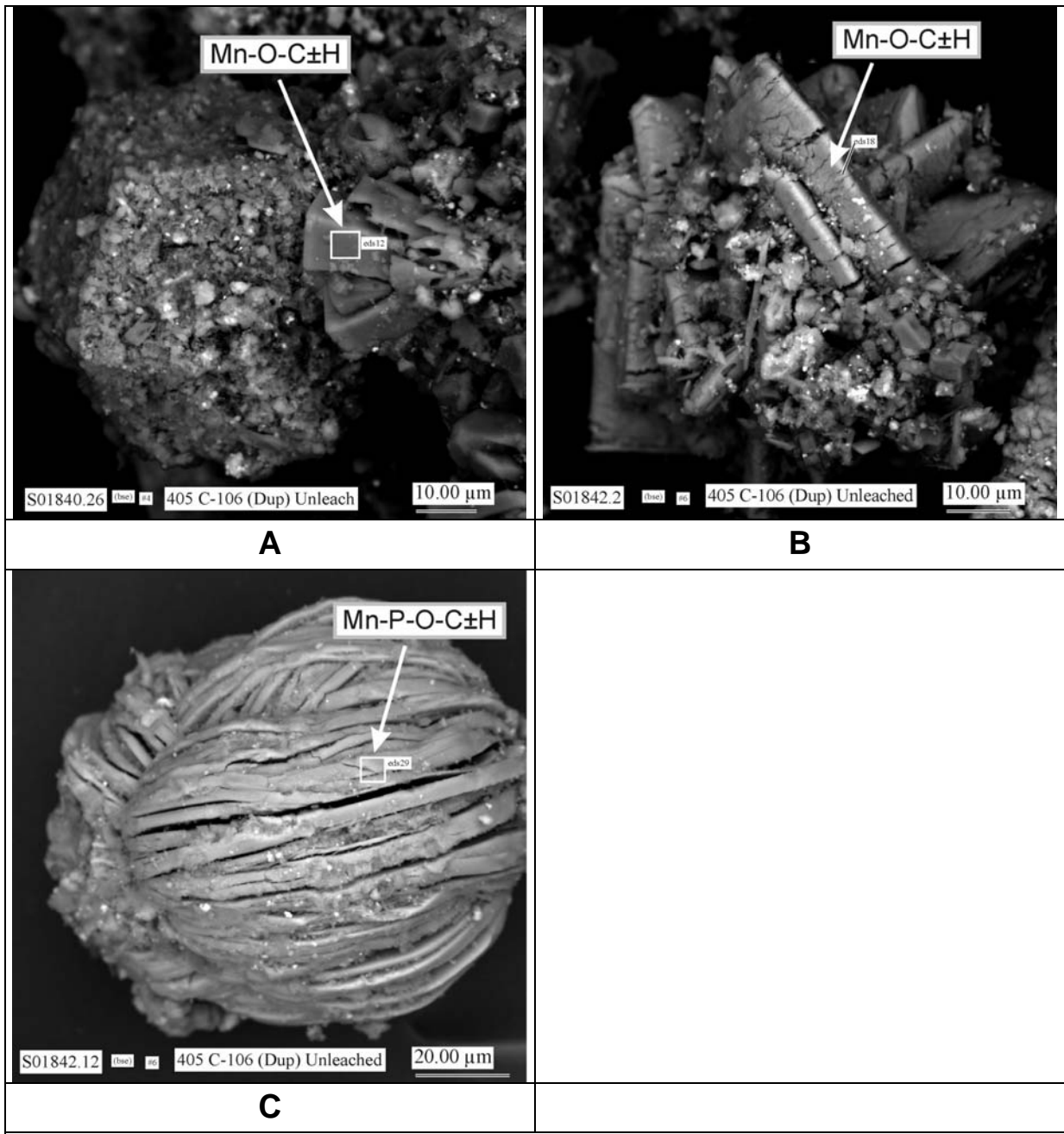any crystal faces, and except for being heavily pitted, were generally nondescript. Micrograph C at the bottom of Figure 3.17 is a higher magnification SEM image of the surface of an Fe oxide particle shown in Figure 3.17(B) from a sample of the HF extract of tank C-106 sludge. This micrograph shows the complex submicrometer layering and particle structure present in the Fe oxides that exist in these tank C-106 sludge samples. Detailed EDS analyses of the necklace-like structures in the lower left and right corners of micrograph C were not possible due their submicrometer size. However, based on the black-and-white contrast of these structures compared to the other material in this micrograph, these necklace-like structures possibly contained Fe and/or Mn as primary components. The conditions that produce these structures are not known. The cavities and complex submicrometer layering and particle structure present in the Fe oxides (see micrograph C in Figure 3.17) suggest that some solid phases, which may contain constituents of importance to sluicing and long-term stability of residual sludges and their risk assessment, can become encapsulated during precipitation of these Fe oxides and thus isolated from tank waste or sluicing solutions. Except when exposed at the surfaces of the Fe oxides, the dissolution of these sequestered phase(s) will likely be delayed and controlled ultimately by the low solubility of the Fe-oxide host phase.

Three sets of SEM micrographs and element distribution maps collected with the newly installed INCAEnergy EDS System are presented in Figures 3.18, 3.19, and 3.20. As noted previously, the capabilities of the INCAEnergy EDS System were demonstrated by completing a limited number of SEM/EDS analyses of SEM mounts of unleached, 82-day water-leached, and HF-extracted sludge samples. Figures 3.18 and 3.19 are low magnification BSE SEM micrographs and element distribution maps for particles in unleached and 82 day water leached sludge from Tank C-106, respectively. SEM micrographs and element distribution maps for the areas indicated by the white dotted rectangles in Figures 3.18 and 3.19 are shown at higher magnification in Appendix E. The concentration of each listed element is directly proportional to the regions of brightness (i.e., brighter the area, the higher the concentration of the selected element) in the corresponding distribution map. The element associations indicated by the distribution maps in Figures 3.18 and 3.19 and Appendix E are consistent with the EDS analyses discussed previously in this section. However, the element distribution maps provide a detailed visualization of the spatial distributions of the selected elements within the imaged particle assemblage, which are not apparent from numerous spot EDS analyses for different particles. Element distribution maps prepared using the INCAEnergy EDS System did not indicate any regions in any particles in the unleached, 82 day water-leached, and HF-extracted sludge samples with detectable concentrations for ^{129}I, ^{99}Tc, or ^{238}U. The element distribution maps however did indicate the presence of some particles, which typically also contained Ag and Hg, with detectable quantities of Cu and/or Pb. Although these two metals were sporadically detected in a few particles of tank C-106 sludge by spot EDS analyses, particles enriched in these metals had not been previously identified.

3.48
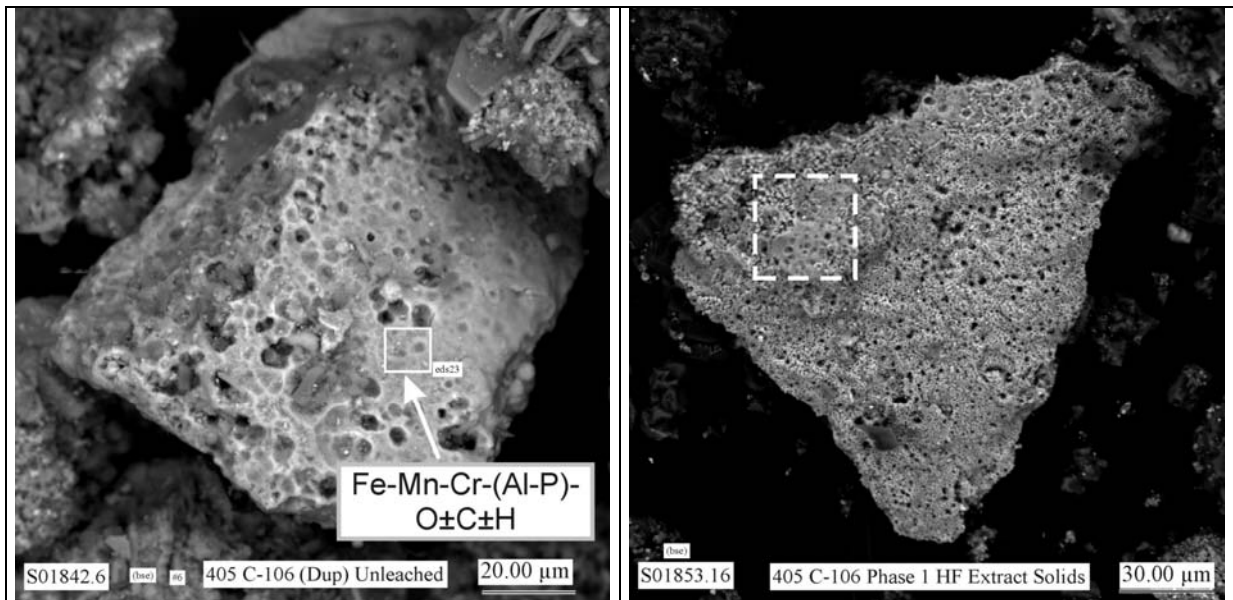
A – Unleached Sample 405

B – HF Stage I Extract Sample 405
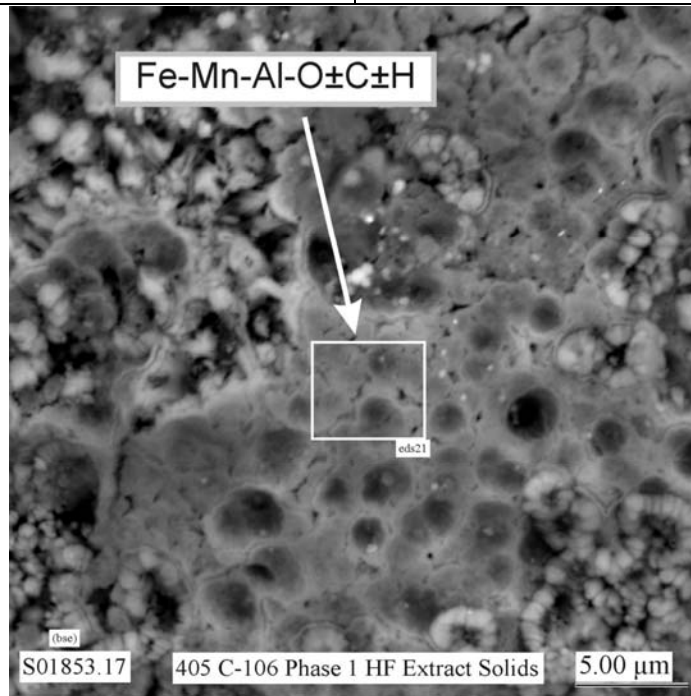
C – SEM micrograph showing at higher magnification the area in micrograph B (above)
indicated by the white, dashed-line square

Figure 3.17.   SEM Micrographs of Fe Oxide Particles in Unleached, 1-Month and 82-Day Water-
Leached, and HF-Extracted Sludge Samples 404 and 405 from Tank C-106
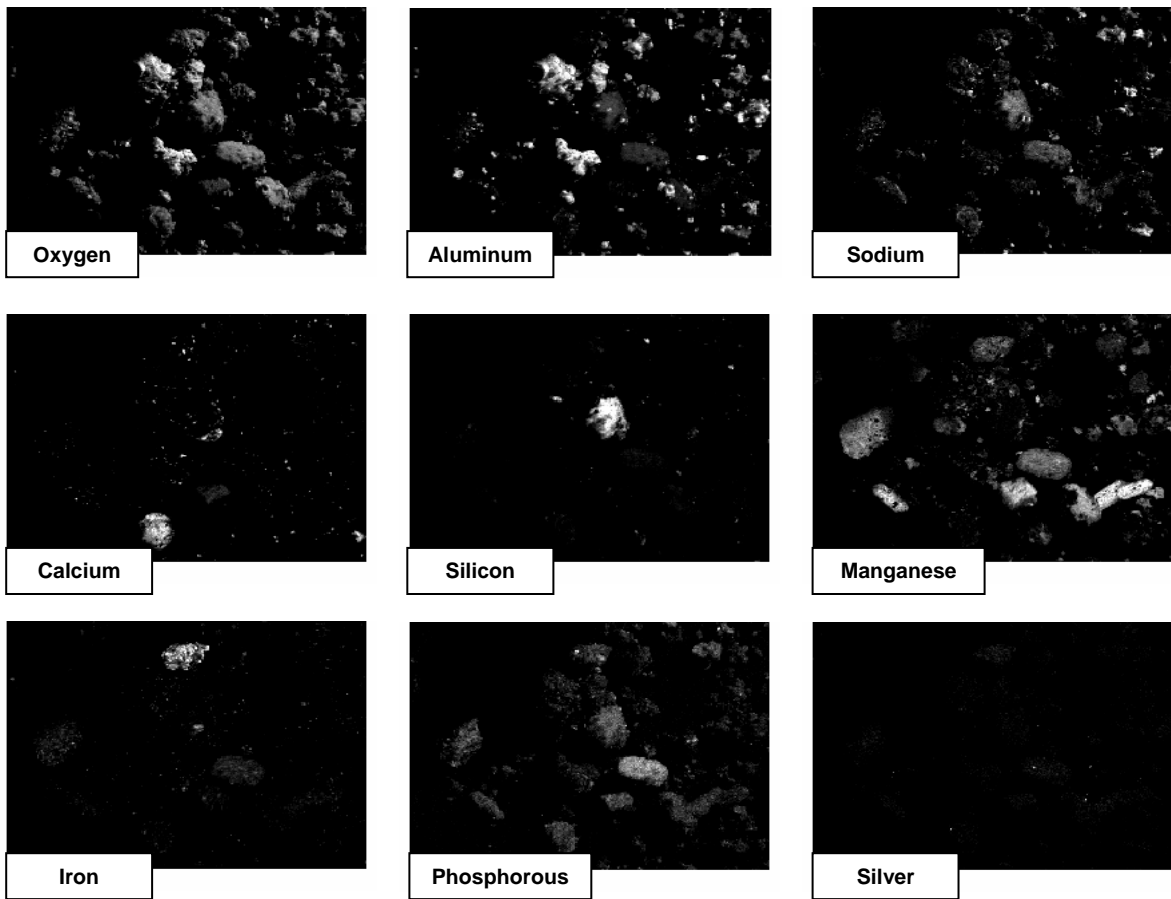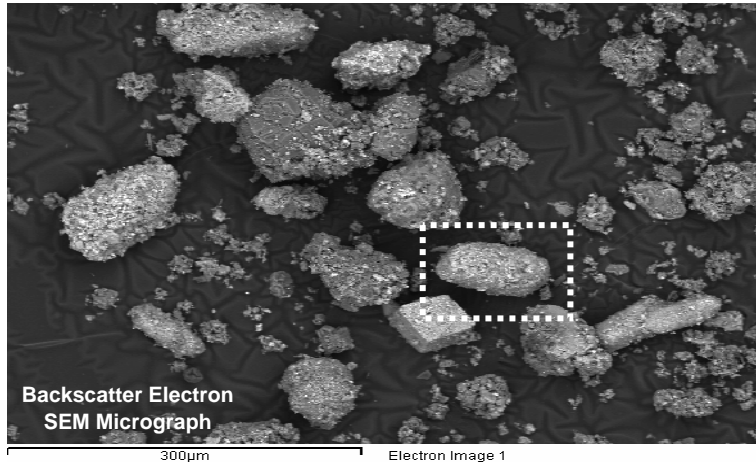
Figure 3.18. Low Magnification Backscatter-Electron SEM Micrograph and Element Distribution Maps for Particles in Unleached Sludge from Tank C-106
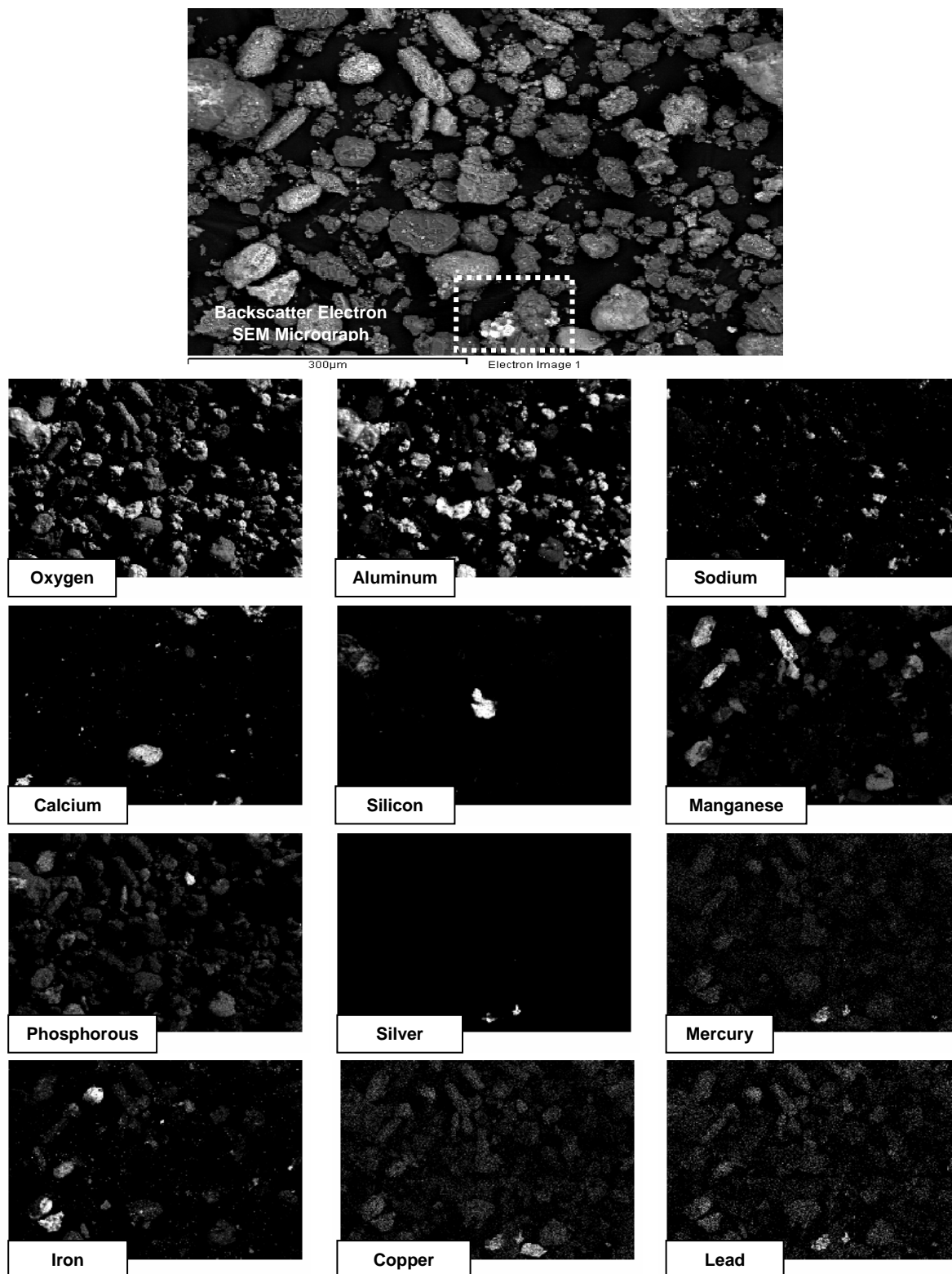
Figure 3.19. Low Magnification Backscatter-Electron SEM Micrograph and Element Distribution Maps for Particles in 82-Day Water-Leached Sludge from Tank C-106
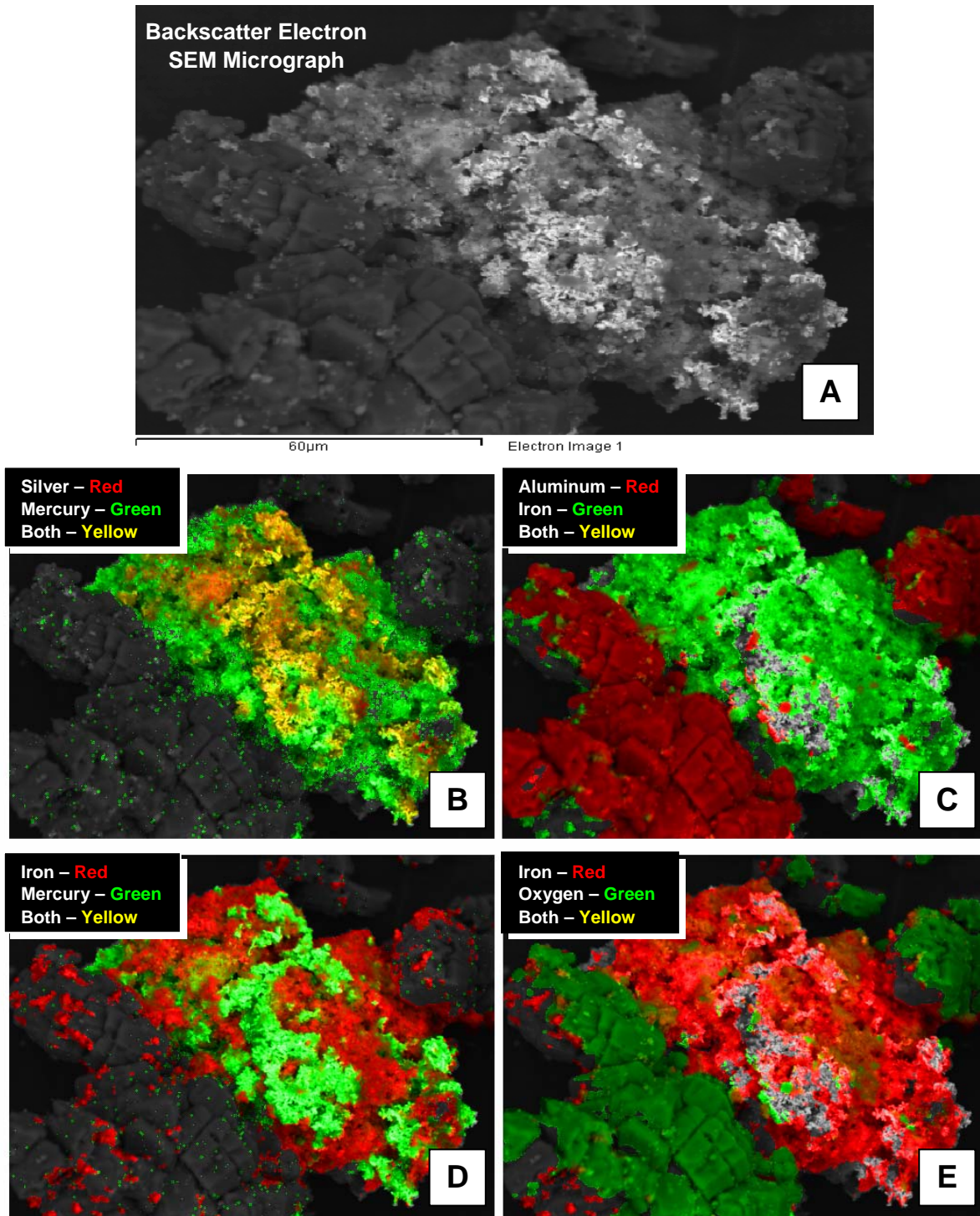
Figure 3.20. Backscatter-Electron SEM Image (A) and Colorized Element Maps (B Through E) for a Particle Aggregate from the HF Extract of Tank C-106 Sludge

Figure 3.20 shows examples of colorized element maps (Figure 3.20B to Figure 3.20E) for an SEM-imaged particle aggregate (Figure 3.20A) from an HF-extracted sample of C-106 residual sludge. The colors in the element maps denote the presence of the indicated, user-specified elements, where the "red areas" signifies the presence of one specified element, the "green areas" for the presence a second element, and the "yellow areas" for the presence both of the selected elements. The INCAEnergy software has the capability to create colorized maps for up to three elements selected by the user. The maps in Figure 3.20 show the relationship between the presence of Ag, Hg, and Fe (Figure 3.20B and Figure 3.20D) and of Fe to Al and O (Figure 3.20C and Figure 3.20E, respectively) for this particle aggregate. These elements correspond to the presence of phases {e.g., the Ag-Hg solid; hematite (Fe_2O_3); and gibbsite [$Al(OH)_3$], böhmite [$AlO(OH)$], and/or dawsonite [$NaAlCO_3(OH)_2$]} determined by bulk X-ray diffraction analysis (XRD) (see Section 3.5). These results also show for the first time that there might be two Hg-containing solids where one is enriched in Ag, and the possible coexistence of Fe with the Hg-rich/Ag-poor solid. Some of the very small colored areas, such as the small green and red points in the southwest areas of Figure 3.20B and Figure 3.20D, respectively, are false positives resulting from overlaps in the corresponding energies of the EDS peaks.

3.7   Comparison of XRD and SEM/EDS Results

The XRD and SEM/EDS results are consistent with each other and with the results reported by Lumetta et al. (1996) and Bechtold et al. (2003). Lumetta et al. (1996) used transmission electron microscopy (TEM) to identify the solids present in samples of an as-received sample of C-106 sludge (obtained several years ago when most of the contents of tank C-106 were sluiced into tank AY-102) and a sludge leached by NaOH. In the as-received tank C-106 sludge, they identified only the presence of amorphous $Al(OH)_3$ and amorphous aluminosilicates. In the caustic leached residue, Lumetta et al. (1996) found amorphous aluminosilicates, $FeO(OH)$, Ag_2O, and ZrO_2. Bechtold et al. (2003) used XRD and SEM/EDS to characterize a baseline sample of C-106 sludge and a baseline sample leached with 1 M oxalic acid. They identified dawsonite, hematite, cancrinite [$Na_6Ca_{1.5}Al_6Si_6O_{24}(CO_3)_{1.6}$], and possibly sidorenkite ($Na_2MnPO_4CO_3$) in the baseline C-106 sample. In residue from baseline C-106 sludge treated with 1 M oxalic acid, Bechtold et al. (2003) reported the presence of hematite, gibbsite, böhmite, possibly "Mn(II) oxalate" ($MnC_2O_4 \cdot 2H_2O$), and traces of Nd-rich and Pb-rich particulates.

Table 3.35 lists the characterization data available for tank C-106 sludge prior to treatment by oxalic acid from Bechtold et al. (2003) and for unleached AY-102 sludge from Krupka et al. (2004). As noted in Section 1.2, the contents of tank C-106 were sluiced to tank AY-102 using supernatant from tank AY-102 as the sluicing fluid, and therefore should be similar to tank C-106 sludge prior to its treatment by oxalic acid.

Table 3.35. Comparison of Characterization Data for Tank C-106 Sludge Prior to Treatment by Oxalic Acid

Type of Sample	XRD and SEM/EDS Analyses by Krupka et al. (2004)		Characterization Results by Bechtold et al. (2003)
	Unleached AY-102 as a Surrogate for C-106 Sludge Prior to Treatment by Oxalic Acid		"Baseline Sample" of C-106 Sludge Prior to Treatment by Oxalic Acid
C-106 sludge prior to treatment by oxalic acid	Dawsonite [$NaAlCO_3(OH)_2$]	Lath-shaped, Na ±Al ±Fe particles	Dawsonite
	Hematite (Fe_2O_3)	Globular aggregates of Fe ±Na ±Mn particles	Hematite
	Gibbsite [$Al(OH)_3$]		Gibbsite
		Na,Al,Si-rich particles	Cancrinite [$Na_6Ca_{1.5}Al_6Si_6O_{24}(CO_3)_{1.6}$]
			Sidorenkite ($Na_2MnPO_4CO_3$)
		Na-rich particles	
		Ag-containing particles	
		U,Na-containing crusty particles	
		Large carbon-rich (graphite?) particles	

Table 3.36 compares the XRD (Section 0) and SEM/EDS (Section 3.6) results from this study to those reported by Bechtold et al. (2003) for tank C-106 sludge treated with 1 M oxalic acid. The XRD and SEM/EDS results from this study overall are consistent with each other and with phases identified by Bechtold et al. (2003). Several phases, however, observed by SEM/EDS were not detected by XRD in this study or do not correspond to any of phases identified by Bechtold et al. (2003). Some of the phases that were not identified by XRD may be amorphous or present at concentrations too low for detection by XRD.

As discussed in the Section 3.6, the phase that consists of Mn-Al-Fe-Na-P-Si-Ca-O±C±H was present on many of the particles in all of the unleached, water-leached, and HF-extracted samples 404 and 405 of sludge from tank C-106. The identity of this phase is not known and does not correspond to any of the solids identified by this XRD analyses or those of Bechtold et al. (2003). The texture of these particles, as characterized by SEM, does not provide any definitive information to evaluate if this material is amorphous or crystalline. Moreover, the results of HF extractions suggest that this phase may consist of more than one phase. Without additional characterization by techniques, such as TEM, it can be presumed at this time that the Mn-Al-Fe-Na-P-Si-Ca-O±C±H phase is possibly amorphous or consists of a mixture of two or more of the phases identified by XRD.

Table 3.36. Comparison of XRD and SEM/EDS Characterization Results from This Study to Those Reported by Bechtold et al. (2003) for Tank C-106 Sludge treated with Oxalic Acid

Type of Sample	XRD Analyses From This Study	SEM/EDS Analyses From This Study	Characterization Results by Bechtold et al. (2003)
C-106 sludge treated by oxalic acid (prior to water leaches and HF extraction)		Mn-Al-Fe-Na-P-Si-Ca-O±C±H	
	Gibbsite	Al-O±H	Gibbsite
	Böhmite		Böhmite
	Dawsonite	Al-Na-O-C±H	
	Hematite	Fe-Cr-O±C±H Fe-Mn-O±C±H	Hematite
	Rhodochrosite Lindbergite ($MnC_2O_4 \cdot 2H_2O$)	Mn-O-C±H (possibly two different phases based on morphology)	"Mn(II) oxalate"??
	Whewellite ($CaC_2O_4 \cdot H_2O$)	Ca-O±C±H	
	Possible Ag-Hg phase	Possibly 1 or 2 phases with Ag-Hg±Fe±Pb±Cu±O±H	
		Mn-O-P±Al±C±H	
		Si-Al-Na-O±C±H	
		REE-rich oxide	Nd-rich particules
		Ca-Si-Al-O±C±H	
		Pb-containing phase	Pb-rich particules
Oxalic acid-treated C-106 sludge after water leaches and HF extraction	Same as XRD results above except for the absence of lindbergite	Same as SEM/EDS results	No data available, since similar water leach and HF extraction studies were not part of the studies by Bechtold et al. (2003)

There are several similarities in the phases identified in sludge from tanks C-106 and AY-102 that has not been leached by oxalic acid with respect to those observed in the tank C-106 sludge tested in this study. For example, gibbsite, dawsonite, and hematite were common to both types of sludge. The phase determined by SEM/EDS to contain Si-Al-Na-O-C±H [see ball-shaped crystals in the upper right of Figure 3.15(A)] possibly corresponds to cancrinite [$Na_6Ca_{1.5}Al_6Si_6O_{24}(CO_3)_{1.6}$] that Bechtold et al. (2003) identified in the baseline C-106 sample prior to oxalic acid treatment. The ball-shaped crystals in the upper right of Figure 3.15(A) look like balls of twine, which is similar to the crystal habit identified by others as being the mineral cancrinite (Bickmore et al. 2001; Bredt et al. 2003; Buck and McNamara 2004). Although highly speculative, the Ag-Hg and Mn-O-P±Al±C±H phases determined by SEM/EDS in this study might be related, respectively, to the Ag-containing phase observed in unleached AY-102 sludge by Krupka et al. (2004) and to sidorenkite ($Na_2MnPO_4CO_3$) reported for baseline tank C-106 sludge by Bechtold et al. (2003) (see Table 3.35).

3.8   Selective Extractions

3.8.1   Phase 1

Phase 1 selective extraction results for samples 404 and 405 are shown in Table 3.37. Data for ^{99}Tc, ^{238}U, Cr, Fe, Al, and Mn are tabulated on the basis of mass of contaminant released per gram of dry sludge. The total contaminant concentrations used in the calculations were those determined by fusion analysis. The results displayed in parentheses are less than the lowest instrument EQL but greater than a zero instrument signal. These values may reflect real concentrations but have larger associated uncertainties. Values with a < sign are instrument values that are less than zero. The number indicated is the EQL. To facilitate easier evaluation of these data, the values are presented as a percentage of the total (fusion) concentrations in Table 3.38. Also included in this table are totals determined by summation of the results from each individual extraction step.

Table 3.37.   Phase 1 Selective Extraction Results for Sludge Samples 404 and 405 from Tank C-106 (in µg/g)

Sample Number	Extractant	^{99}Tc (µg/g)	^{238}U (µg/g)	Cr (µg/g)	Fe (µg/g)	Al (µg/g)	Mn (µg/g)
404	Deionized Water	1.86E-02	6.03E+00	(2.62E+00)	(2.64E+01)	7.46E+01	8.79E+03
404 DUP	Deionized Water	1.75E-02	6.78E+00	(9.66E-01)	(2.20E+01)	9.18E+01	8.94E+03
404	NH$_2$OH-HCl	2.25E-01	1.25E+02	8.63E+01	3.37E+03	1.06E+04	6.24E+04
404 DUP	NH$_2$OH-HCl	2.24E-01	1.58E+02	9.82E+01	4.10E+03	1.36E+04	8.76E+04
404	0.01HF/0.01 KF	4.92E-02	6.71E+01	6.03E+01	2.57E+03	1.91E+04	6.86E+03
404 DUP	0.01HF/0.01 KF	6.10E-02	7.76E+01	8.74E+01	3.03E+03	2.36E+04	5.92E+03
404	0.01HF/0.01 KF	5.36E-02	1.29E+01	5.85E+01	1.80E+03	7.96E+03	2.47E+03
404 DUP	0.01HF/0.01 KF	6.84E-02	1.78E+01	1.17E+02	3.58E+03	8.53E+03	7.04E+03
404	Hot Conc. HNO$_3$	3.65E-01	2.46E+01	4.70E+02	2.30E+04	7.34E+04	3.23E+04
404 DUP	Hot Conc. HNO$_3$	4.12E-01	1.95E+01	3.09E+02	1.68E+04	5.85E+04	2.23E+04
Total	Fusion	1.01E+00	2.36E+02	8.41E+02	4.33E+04	1.40E+05	1.14E+05
405	Deionized Water	1.92E-02	7.37E+00	(4.23E+00)	(8.22E+01)	1.38E+02	1.64E+04
405 DUP	Deionized Water	1.29E-02	7.95E+00	<6.12E+01	(3.50E+01)	1.15E+02	1.47E+04
405	NH$_2$OH-HCl	2.05E-01	1.25E+02	9.86E+01	3.83E+03	1.22E+04	8.05E+04
405 DUP	NH$_2$OH-HCl	1.44E-01	9.66E+01	6.96E+01	2.99E+03	9.63E+03	6.87E+04
405	0.01HF/0.01 KF	5.24E-02	7.56E+01	8.05E+01	2.81E+03	2.10E+04	7.43E+03
405 DUP	0.01HF/0.01 KF	4.73E-02	5.98E+01	5.25E+01	2.18E+03	1.76E+04	9.66E+03
405	0.01HF/0.01 KF	5.84E-02	1.56E+01	8.37E+01	2.59E+03	6.55E+03	2.57E+03
405 DUP	0.01HF/0.01 KF	4.40E-02	1.63E+01	6.48E+01	2.07E+03	8.15E+03	2.27E+03
405	Hot Conc. HNO$_3$	2.40E-01	1.42E+01	2.18E+02	1.05E+04	3.26E+04	7.72E+03
405 DUP	Hot Conc. HNO$_3$	5.67E-01	2.12E+01	7.19E+02	3.29E+04	9.63E+04	4.94E+04
Total	Fusion	1.31E+00	2.58E+02	8.07E+02	4.33E+04	1.23E+05	1.22E+05
Conc = Concentrated. DUP = Duplicate sample.							

Table 3.38. Phase 1 Selective Extraction Results for Sludge Samples 404 and 405 from Tank C-106 (in percentages)

Sample Number	Extractant	^{99}Tc	^{238}U	Cr	Fe	Al	Mn
		------------------------------------% Leached------------------------------------					
404	Deionized Water	1.8	2.6	0.3	0.1	0.1	7.7
404 DUP	Deionized Water	1.7	2.9	0.1	0.1	0.1	7.8
404	NH$_2$OH-HCl	22.3	53.0	10.3	7.8	7.6	54.7
404 DUP	NH$_2$OH-HCl	22.2	66.8	11.7	9.5	9.7	76.8
404	0.01 HF/0.01 KF	4.9	28.4	7.2	5.9	13.7	6.0
404 DUP	0.01 HF/0.01 KF	6.0	32.9	10.4	7.0	16.9	5.2
404	0.01 HF/0.01 KF	5.3	5.4	7.0	4.2	5.7	2.2
404 DUP	0.01 HF/0.01 KF	6.8	7.6	14.0	8.3	6.1	6.2
404	Hot Conc. HNO$_3$	36.1	10.4	55.9	53.2	52.4	28.3
404 DUP	Hot Conc. HNO$_3$	40.8	8.3	36.8	38.7	41.8	19.5
404	Total	70.4	99.8	80.6	71.1	79.5	99.0
404 DUP	Total	77.6	118.5	72.9	63.5	74.5	115.6
405	Deionized Water	1.5	2.9	0.5	0.2	0.1	13.4
405 DUP	Deionized Water	1.0	3.1	0.0	0.1	0.1	12.0
405	NH$_2$OH-HCl	15.6	48.3	12.2	8.6	9.9	65.9
405 DUP	NH$_2$OH-HCl	11.0	37.4	8.6	6.8	7.8	56.3
405	0.01 HF/0.01 NaF	4.0	29.3	10.0	6.4	17.1	6.1
405 DUP	0.01 HF/0.01 NaF	3.6	23.2	6.5	4.9	14.3	7.9
405	0.01 HF/0.01 NaF	4.5	6.0	10.4	5.8	5.3	2.1
405 DUP	0.01 HF/0.01 NaF	3.4	6.3	8.0	4.7	6.6	1.9
405	Hot Conc. HNO$_3$	18.3	5.5	27.0	23.8	26.5	6.3
405 DUP	Hot Conc. HNO$_3$	43.3	8.2	89.1	74.2	78.3	40.5
405	Total	43.9	92.0	60.1	44.8	58.9	93.9
405 DUP	Total	62.2	78.2	112.2	90.7	107.2	118.6
Conc = Concentrated.							
DUP = Duplicate sample.							

The most notable result for the deionized water leaches is the fact that a relatively large amount of Mn (8-13%) dissolved. Chemical equilibrium calculations conducted using the average concentration of the sample and duplicate for the sample 404 water leach solutions indicate that the water leachate solutions are near equilibrium (somewhat oversaturated) with respect to manganese oxalate (SI= 0.43, where SI = Log (Q/K$_{sp}$, Q is the ion activity product for the solid and K$_{sp}$ is the solubility constant for the solid, see Appendix G for details). Manganese oxalate has been identified by XRD in samples 404 and 405 (see Section 0). Chemical equilibrium modeling also indicates that during the 0.9 M oxalic acid treatment used in tank C-106, manganese (Mn(III) and Mn(IV)) oxyhydroxide phases would be reduced to Mn^{2+}(aq) ion, which would subsequently precipitate as manganese oxalate (see Appendix G). Batrakov et al. (1998) have demonstrated that reductive dissolution of manganese dioxide to Mn^{2+} by oxalic acid is rapid under acidic conditions. They found that the maximum dissolution rate occurred at

pH 1.6. The rate decreased to near zero above pH 4.5. Tank C-106 sludge also contained sidorenkite ($Na_3MnPO_4CO_3$) prior to oxalic acid treatment. It is expected that sidorenkite would readily dissolve under the acidic conditions of the oxalic acid treatment step. When sidorenkite dissolves in this process, the Mn^{2+} released to solution would also precipitate as manganese oxalate.

The hydroxylamine hydrochloride ($NH_2OH\cdot HCl$) extraction step was designed to selectively dissolve manganese oxyhydroxides in the sludge; however, it is clear from the previous discussion that it is likely that, as a result of the 0.9 M oxalic acid treatment step, a significant fraction of the Mn occurring as oxyhydroxides or sidorenkite in the tank sludge would have been transformed to Mn oxalate. The hydro-xylamine hydrochloride extraction solution has a pH of approximately 3.6. This low pH would tend to protonate oxalate and result in dissolution of Mn-oxalate. As a result, the hydroxylamine hydrochloride extraction would also be effective at dissolving Mn oxalate in addition to manganese oxyhydroxides. The amount of Mn solids dissolved during this selective extraction step ranged from 55 to 77%. Along with the Mn, a large fraction of the ^{238}U was released (37 to 67%). A fair amount of ^{99}Tc was also released (11 to 22%). Lower quantities of Cr (9 to 12%), Fe (7-10%), and Al (8-10%) were released.

The 0.01 HF/0.01 NaF extraction steps (pH 3.2) were intended to selectively release aluminum oxyhydroxide phases; however, during both extraction stages only between 19 and 23% of the aluminum was released. The 0.01 HF/0.01 NaF extraction steps were more effective at releasing uranium. Between 30 and 41% of the uranium was released during the two 0.01 HF/0.01 NaF extraction steps. Smaller amounts of ^{99}Tc (7-13%), Fe (10-15%), Mn (8-11%), and Cr (14-24%) were also released.

Results of the final hot concentrated HNO_3 extraction indicate that a significant amount of the Fe, Al, Mn, ^{99}Tc, ^{238}U, and Cr, remained immobile during the earlier mildly acidic extraction steps. The amounts of constituents released by the hot concentrated HNO_3 extractions ranged from 24-74% for Fe, 27-78% for Al, 6-40% for Mn, 18-43% for ^{99}Tc, 6-10% for ^{238}U, and 27-89% for Cr. The sum of all the selective extractions indicate that significant fractions of ^{99}Tc, Cr, Fe, and Al, were frequently not dissolved even after the hot concentrated HNO_3 extraction (up to 56% for ^{99}Tc, up to 40% for Cr, up to 65% for Fe and up to 41% for Al).

Some general conclusions and observations can be made regarding these selective extraction results. Cr was found to exhibit a fairly strong correlation with Fe. This was also observed in the SEM/EDS analyses (Section 3.6). The ^{238}U appears to be most highly correlated with the release of Mn. Some association of ^{99}Tc with the release of Mn may occur; however, the correlation is not as strong as the correlation with Fe. In general, the selective extraction methods were less selective than anticipated for the C-106 residual sludge samples. One possible reason for this result may be cation substitution among the various Fe, Al, Mn oxyhydroxides. The formation of the secondary Mn phases [Mn-oxalate and rhodochrosite ($MnCO_3$)] during the sludge removal process may have also contributed to the problem by co-precipitating some of the trace constituents that were initially released by sludge dissolution reactions.

3.8.2   Phase 2a

Phase 2a selective extraction results for samples 404 and 405 are shown in Table 3.39. Data for ^{99}Tc, ^{238}U, Cr, Fe, Al, and Mn are tabulated on the basis of mass of contaminant released per gram of dry

Table 3.39. Phase 2a Selective Extraction Results for C-106 Sludge Samples 404 and 405 (in µg/g)

Sample Number	Extractant	^{99}Tc (µg/g)	^{238}U (µg/g)	Cr (µg/g)	Fe (µg/g)	Al (µg/g)	Mn (µg/g)
404	0.01 HF/0.01 KF	3.58E-02	5.97E+01	1.54E+01	7.11E+02	6.47E+03	1.47E+04
404 DUP	0.01 HF/0.01 KF	3.51E-02	3.04E+01	1.11E+01	(4.34E+00)	7.12E+03	1.70E+04
404	0.01 HF/0.01 KF	3.90E-02	5.50E+01	1.98E+01	4.18E+01	7.66E+03	1.63E+04
404 DUP	0.01 HF/0.01 KF	3.74E-02	5.45E+01	1.92E+01	1.29E+01	7.33E+03	1.62E+04
Total	Fusion	1.01E+00	2.36E+02	8.41E+02	4.33E+04	1.40E+05	1.14E+05
405	0.01 HF/0.01 KF	3.33E-02	3.99E+01	8.36E+00	1.30E+01	6.58E+03	1.43E+04
405 DUP	0.01 HF/0.01 KF	3.39E-02	4.50E+01	1.04E+01	1.28E+01	7.35E+03	1.53E+04
405	0.01 HF/0.01 KF	3.42E-02	5.55E+01	1.58E+01	1.31E+01	6.82E+03	1.47E+04
405 DUP	0.01 HF/0.01 KF	2.82E-02	4.22E+01	2.39E+01	1.19E+03	5.65E+03	8.29E+03
Total	Fusion	1.31E+00	2.58E+02	8.07E+02	4.33E+04	1.23E+05	1.22E+05
DUP = Duplicate sample.							

sludge. The total contaminant concentrations used in the calculations were those determined by fusion analysis. To facilitate easier evaluation of these data, the results are presented as a percentage of the total (fusion) concentrations in Table 3.40. Also included in this table are totals determined by summation of the results from both extraction steps. These selective extractions were designed to selectively remove Al oxyhydroxides. The results in Table 3.40 indicate that this extraction did not work as intended on sludge from tank C-106. Only about 10 to 11% of the total Al was removed with this method. Much higher

Table 3.40. Phase 2a Selective Extraction Results for Primary (404) and Duplicate (405) Sludge Samples from Tank C-106 (in percentages)

Sample Number	Extractant	^{99}Tc (%)	^{238}U (%)	Cr (%)	Fe (%)	Al (%)	Mn (%)
404	0.01 HF/0.01 KF	3.5	25.3	1.8	1.6	4.6	12.9
404 DUP	0.01 HF/0.01 KF	3.5	12.9	1.3	0.0	5.1	14.9
404	0.01 HF/0.01 KF	3.9	23.3	2.4	0.1	5.5	14.3
404 DUP	0.01 HF/0.01 KF	3.7	23.1	2.3	0.0	5.2	14.2
404	Total	7.4	48.6	4.2	1.7	10.1	27.2
404 DUP	Total	7.2	36.0	3.6	0.0	10.3	29.1
405	0.01 HF/0.01 NaF	2.5	15.5	1.0	0.0	5.3	11.7
405 DUP	0.01 HF/0.01 NaF	2.6	17.4	1.3	0.0	6.0	12.5
405	0.01 HF/0.01 NaF	2.6	21.5	2.0	0.0	5.5	12.0
405 DUP	0.01 HF/0.01 NaF	2.2	16.4	3.0	2.7	4.6	6.8
405	Total	5.2	37.0	3.0	0.1	10.9	23.8
405 DUP	Total	4.7	33.8	4.3	2.8	10.6	19.3
DUP = Duplicate sample.							

percentages of Mn and [238]U were extracted with this method. The reason for the lack of intended specificity for this extraction is due primarily to an unexpected increase in pH that occurred during the extraction. The final pH values measured in the extracts ranged from 4.39 to 6.64. The initial pH of the 0.01 HF/0.01 NaF buffer was approximately 3.2. The reason for neutralization of the pH appears to be excessive proton consumption through dissolution of $Al(OH)_3$, Mn oxyhydroxides, and carbonate phases. In other words, the buffer concentration used was not high enough to effectively dissolve all the $Al(OH)_3$ and other proton-consuming phases in the sludge. The high fluoride concentrations may have been responsible for the relatively high concentrations of [238]U that were solubilized in these extractions. Fluoride is a fairly strong complexing agent for U(VI) (Grenthe et al. 1992).

3.8.3   Phase 2b

Phase 2b selective extraction results for tank C-106 samples 404 and 405 are shown in Table 3.41. Data for [238]U, Cr, Fe, Al, and Mn are tabulated on the basis of mass of contaminant released per gram of dry sludge. The total contaminant concentrations used in the calculations were those determined by fusion analysis. To facilitate easier evaluation of these data, the results are presented as a percentage of the total (fusion) concentrations in Table 3.42. Also included in Table 3.42 are totals determined by summation of the results from both extraction steps. The 3 M NaOH extraction was also designed to selectively extract Al oxyhydroxides. This method was not particularly effective at solubilizing Al from C-106 residual sludge. Only about 11 to 13% of the total Al was extracted. The method does appear to be more selective than the previous methods used with respect to Fe and Mn. Very little Fe and Mn were extracted; however 7.4 to 11.3% of the [238]U was extracted. Interestingly, 3 to 4% of the Cr was mobilized during the second extraction step. (Note: the first extraction (E1) was conducted for 24 hours, whereas the second extraction (E2) was allowed to continue for 11 days.)

Table 3.41.   Phase 2b Selective Extraction Results for Sludge Samples 404 and 405 from Tank C-106

Sample Number	Extractant	[238]U (µg/g)	Cr (µg/g)	Fe (µg/g)	Al (µg/g)	Mn (µg/g)
404	3 M NaOH (E1)	1.61E+01	(1.07E+00)	8.38E+00	1.27E+04	6.07E+00
404 DUP	3 M NaOH (E1)	1.94E+01	(7.03E-01)	8.98E+00	1.41E+04	5.93E+00
404	3 M NaOH (E2)	4.29E+00	2.51E+01	2.04E+01	2.49E+03	(4.73E-01)
404 DUP	3 M NaOH (E2)	7.26E+00	3.49E+01	2.73E+01	3.65E+03	2.35E+00
Total	Fusion	2.36E+02	8.41E+02	4.33E+04	1.40E+05	1.14E+05
405	3 M NaOH (E1)	NA	(1.19E+00)	7.35E+00	1.18E+04	(1.71E-01)
405 DUP	3 M NaOH (E1)	1.46E+01	(5.46E-01)	1.41E+01	1.26E+04	8.26E+00
405	3 M NaOH (E2)	4.23E+00	3.03E+01	2.11E+01	2.52E+03	(4.41E-01)
405 DUP	3 M NaOH (E2)	4.39E+00	2.54E+01	2.05E+01	2.97E+03	(4.77E-01)
Total	Fusion	2.58E+02	8.07E+02	4.33E+04	1.23E+05	1.22E+05
DUP = Duplicate sample.						

Table 3.42. Phase 2b Selective Extraction Results for Sludge Samples 404 and 405 from Tank C-106 (in percentages)

Sample Number	Extractant	^{238}U (%)	Cr (%)	Fe (%)	Al (%)	Mn (%)
404	3 M NaOH (E1)	6.8	(0.1)	0.0	9.1	0.0
404 DUP	3 M NaOH (E1)	8.2	(0.1)	0.0	10.1	0.0
404	3 M NaOH (E2)	1.8	3.0	0.0	1.8	0.0
404 DUP	3 M NaOH (E2)	3.1	4.2	0.1	2.6	0.0
404	Total	8.6	3.1	0.1	10.8	0.0
404 DUP	Total	11.3	4.2	0.1	12.7	0.0
405	3 M NaOH (E1)	NA	(0.1)	0.0	9.6	0.0
405 DUP	3 M NaOH (E1)	5.7	(0.1)	0.0	10.2	0.0
405	3 M NaOH (E2)	1.6	3.7	0.0	2.0	0.0
405 DUP	3 M NaOH (E2)	1.7	3.1	0.0	2.4	0.0
405	Total	NA	3.9	0.1	11.7	0.0
405 DUP	Total	7.4	3.2	0.1	12.6	0.0
DUP = Duplicate sample.						

4.0   Contaminant Release Model

The primary objective of this project is to develop source release models for contaminants of concern present in residual waste upon closure of Hanford single-shell tanks. As shown in Figure 4.1, developing these models consists of laboratory testing to produce contaminant release data and a conceptual source release model. After development, the release model can be incorporated into a fate and transport model as part of a long-term risk/performance assessment for the closed tank. This section describes the conceptual release models developed for ^{238}U, ^{99}Tc, Cr, and ^{129}I from the laboratory data collected during testing of residual sludge from tank C-106.

Data collected and analyses conducted as part of this study indicated that contaminant release from tank C-106 residual sludge will be geochemically complex. Contaminant release from the sludge is expected to involve mineral dissolution and precipitation, reduction-oxidation reactions, solution phase complexation, and surface adsorption. Some of these reactions can be modeled as equilibrium reactions whereas others will be rate controlled. Although significant insight was gained in this study regarding the contaminant release mechanisms that will be important for tank C-106 sludge, it was not possible to adequately characterize the phase associations of the contaminants of concern in sufficient detail to produce a mechanistically rigorous geochemical release model. This could change with additional work; however, in the meantime, a release model based on empirical solubilities has been developed. The approach used is inherently conservative and will over-estimate contaminant release. Further characterization work could allow the development of a more mechanistically rigorous geochemical release model, if warranted.
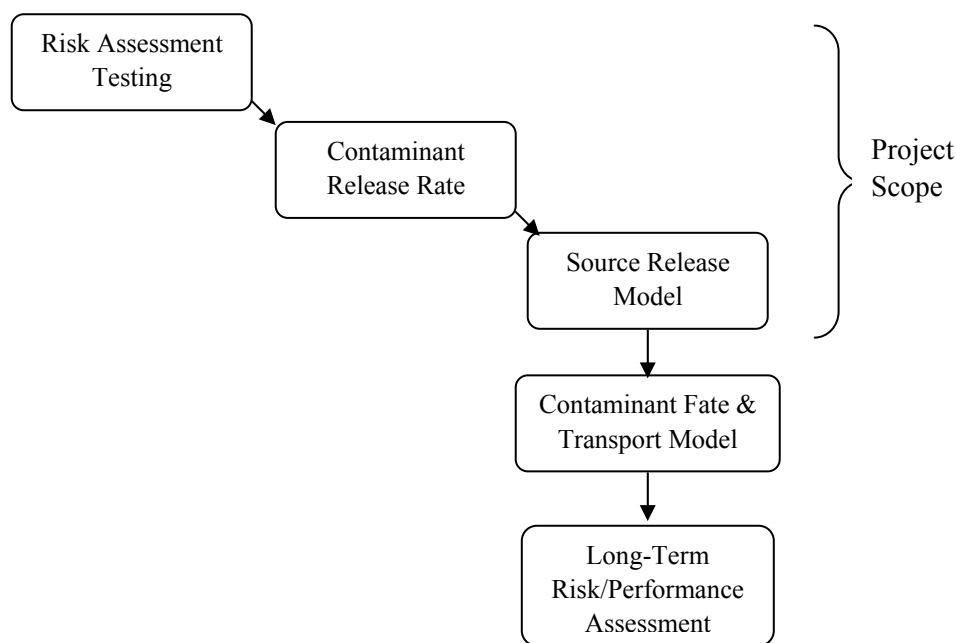

Figure 4.1. Source Release Model Development for Long-Term Risk/Performance Assessments

The contaminant release model that has been developed for residual sludge in tank C-106 is based on empirical solubilities of the contaminants of concern. Empirical solubilities can be influenced by many geochemical conditions (e.g., pH, Eh, ionic strength, presence/concentration of complexing species) that are potentially dynamic throughout the history of the evolving sludge environment. In addition, available data indicate that portions of the contaminants of concern are incorporated into other slightly soluble phases. The dissolution of these relatively insoluble phases likely control the long-term release of the contaminants of concern in the residual sludge. For these reasons, the release models described in this section are expected to be conservative only to tank conditions that are currently expected at closure and will need to be modified as additional information becomes available regarding anticipated tank conditions (such as the addition of grout).

Before discussing the release models for the four individual contaminants of concern (^{238}U, ^{99}Tc, Cr, and ^{129}I), a conceptual model of the chemical transformations that have occurred in the tank as a result of chemical treatments used in the sludge removal process will be described. This has important implications with regard to how contaminants would be released from tank C-106 residual sludge.

4.1 Conceptual Model of Chemical Transformations of Tank C-106 Sludge Resulting from the Retrieval Process

A chemical treatment was used as part of the sludge retrieval process for tank C-106. In this process, a solution of 0.9 M oxalic acid was added to the tank to dissolve and suspend sludge solids. Then, as much of the resulting suspension as possible was pumped out. This was repeated 4 to 6 times. After the final oxalic acid removal step, a single water rinse was conducted to remove as much of the acid solution as possible. The final step in the process was the addition of a 0.5 M NaOH solution to neutralize the remaining acidity in the sludge. As much of this neutralizing solution as possible was removed through pumping. It is estimated that approximately 75% of the sludge volume was removed throughout the entire retrieval process. Based on the results presented in this report (the average of the fusion results for samples 404 and 405) and previous analyses of untreated tank C-106 sludge (Bechtold et al. 2003, Table 7.1), it is apparent that the 0.9 M oxalic acid treatment process has significantly altered the composition of the residual sludge. For example, Table 4.1 shows the weight percent of three major sludge components (Al, Fe, and Mn) before and after treatment. Also included in this table are concentrations of three major contaminants of concern (^{99}Tc, ^{238}U, and Cr). These results indicate that as a

Table 4.1. Three Major Sludge Components (Weight Percent) and Major Contaminants of Concern in Tank C-106 Sludge Before and After 1 M Oxalic Acid Treatment

Sample	Al %	Fe %	Mn %	^{99}Tc (µg/g)	^{238}U (µg/g)	Cr (µg/g)
C-106 Before	3.7	6.5	6.5	1.3	81[a]	2,100
404/405 Avg	13.2	4.4	11.8	1.2	247	897
Avg = Average. (a) This result is assumed to be in µg/g, rather than mg/g as indicated in Bechtold et al. (2003).						

result of the oxalic acid treatment of tank C-106 sludge, Al, Mn, and ^{238}U have become enriched in the residual sludge. Fe and Cr have been depleted. Based on these results, and the fact that approximately 75% of the sludge was removed by the oxalic acid process, it would appear that very little, if any, of the Al was removed by this process.

A conceptual model has been developed to describe the chemical transformations that occurred in tank C-106 sludge during the sludge retrieval process and subsequent transformations that are expected to occur during future exposure to infiltrating natural recharge water. This model is illustrated schematically in Figure 4.2. Prior to the oxalic acid treatment, gibbsite [$Al(OH)_3$], hematite [Fe_2O_3], dawsonite [$NaAlCO_3(OH)_2$], cancrinite [$Na_6Ca_{1.5}Al_6Si_6O_{24}(CO_3)_{1.6}$], and sidorenkite ($NaMnPO_4CO_3$] were identified by XRD and SEM/EDS (Bechtold et al. 2003). It is possible that poorly crystalline/isomorphically substituted Mn and Fe oxyhydroxides also occurred in the sludge, but could not be identified by XRD. During the oxalic acid treatment of the sludge, significant quantities of the Fe and Mn oxyhydroxides were likely to have dissolved through reductive dissolution (e.g., Batrakov et al. 1998; Suter et al 1991; Zhang et al. 1985). As the dissolved Mn(II) concentration increased, Mn(II) oxalate would precipitate as a result of its low solubility and the high oxalate concentration in the solution phase. Based on the NH_2OH-HCl selective extraction results, it would appear that some of the ^{99}Tc, ^{238}U, and (possibly) Fe, released during the reductive dissolution of the Mn-oxyhydroxides, co-precipitated with the Mn-oxalate.

After the sludge was treated multiple times with the oxalic acid solution, the residual sludge was rinsed with water and then treated with a NaOH solution. The final pH of the solution in contact with the residual sludge was 12.9. After the base neutralization step, the sludge contained significant amounts of carbonate (see Table 3.12). The source of this carbonate is not clear; however, it is not likely that carbonates existing within the original sludge would have survived the oxalic acid treatments. After the base neutralization step, the sludge could have absorbed carbon dioxide from the head space in the tanks. Carbon dioxide could have also entered the tanks along with the NaOH solution used for the neutralization step. Carbon dioxide contamination of base can easily occur through exposure to air, both in solution or solid NaOH used to make up the NaOH solution. In any case, rhodochrosite ($MnCO_3$) was identified by XRD in the residual sludge (Figure 3.14). Rhodochrosite has a lower solubility constant than Mn-oxalate and would likely form at the expense of Mn-oxalate as the carbonate concentrations increased in the sludge.

Although both XRD and SEM/EDS data (Sections 3.6 and 3.7) support the existence of Mn-oxalate and/or rhodochrosite in the residual sludge, data in Table 3.12 suggest that a significant amount of the residual Mn in the sludge is still in the form of Mn oxyhydroxides. For example, if we assume that all the oxalate occurs in the residual sludge as Mn-oxalate and all the carbonate exists as rhodochrosite, 35% of the Mn would still remain unaccounted for. It is assumed that at least this much Mn occurs as Mn oxyhydroxides in the residual sludge. In addition to Mn-oxalate and rhodochrosite, gibbsite [$Al(OH)_3$], hematite [Fe_2O_3], dawsonite [$NaAlCO_3(OH)_2$], boehmite [$AlO(OH)$], and whewellite [Ca-oxalate] were identified by XRD and SEM/EDS in the residual sludge.
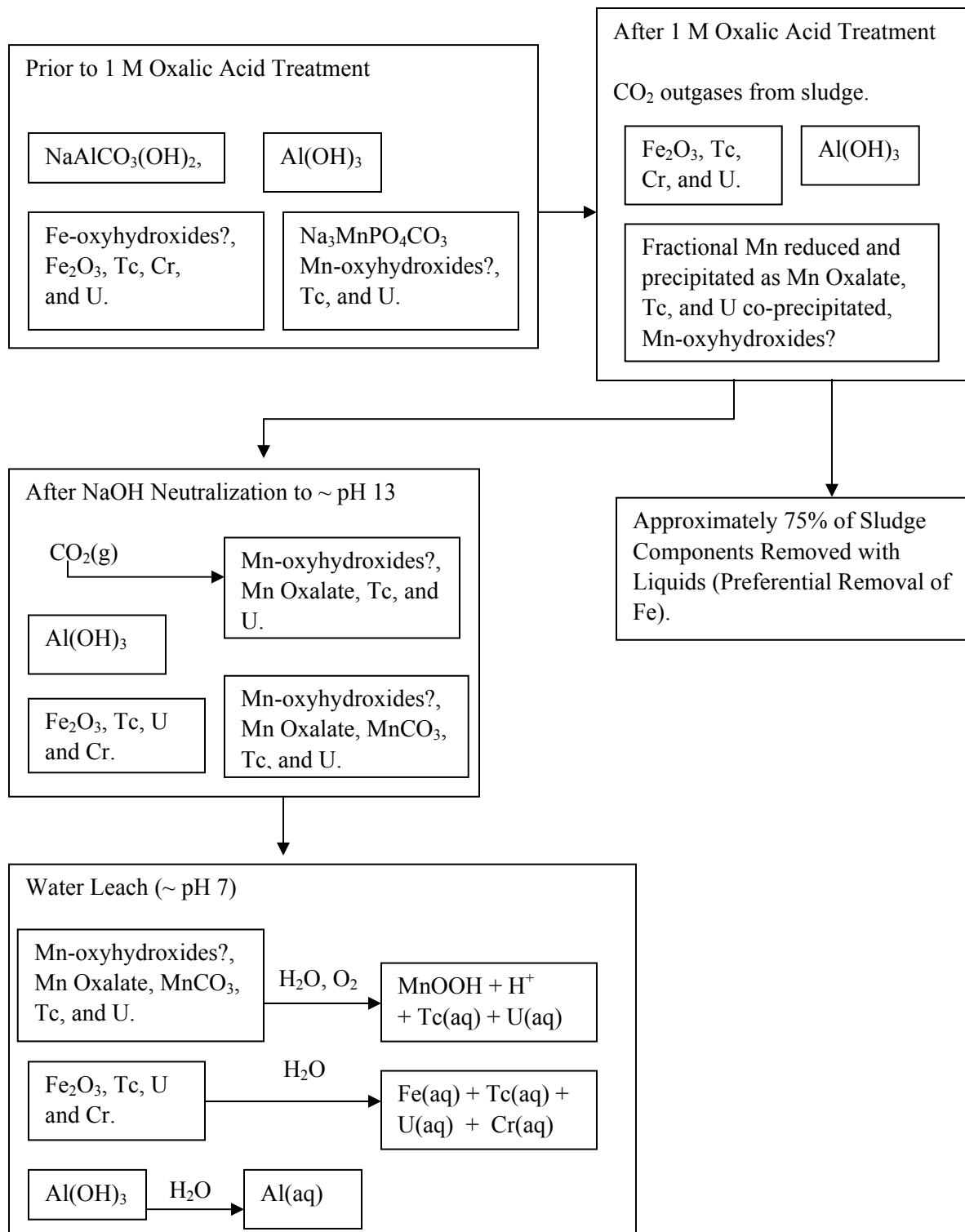
Figure 4.2. Conceptual Model of Chemical Transformations for Tank C-106 Sludge Resulting from Tank Retrieval Process and Water Leaching Relevant to Contaminant Release Modeling

As part of this study, sequential water leaches were conducted. Results of the water leach experiments indicated unexpectedly low pH values. For example, the tank C-106 liquid had a pH of 12.9 (Table 3.13). Assuming a simple 100:1 water to sludge ratio for the sequential leach experiments and a 50% moisture content for the sludge (Table 3.1), a pH of approximately 10.6 could be expected for the sequential leach final solutions. The pH values measured during these experiments along with Mn and oxalate concentrations, and oxalate/Mn ratios are listed in Table 4.2. As can be seen, the measured pH values in the extracts of these experiments are in the range of 6.7 to 7.7. After analysis of all the analytical data for these extracts, the only plausible source of acidity that could explain the depressed pH values in these experiments is oxidation of Mn(II) to Mn(III) and subsequent precipitation as a Mn oxyhydroxide. For example:

$$Mn^{2+} \; + \; 3/2H_2O \; + \; 1/2O_2 \; \rightarrow \; MnOOH \; + \; 2H^+ \tag{4.1}$$

Table 4.2. Contact Times and Average pH Values, Oxalate and Mn Concentrations, and Oxalate/Mn Ratios, for the Periodic Replenishment Test on Tank C-106 Sludge Samples

Contact Stage	Contact Duration (days)	pH	Ox (mol/L)	Mn (mol/L)	Ox/Mn
Sample 404					
1	1	7.03	2.33E-03	1.40E-03	1.66
2	1	6.87	1.54E-03	1.30E-03	1.18
3	4	6.93	1.11E-03	9.22E-04	1.21
4	1	6.75	6.13E-04	4.74E-04	1.30
5	1	6.73	3.61E-04	2.66E-04	1.37
6a	43	7.43	3.49E-04	2.15E-04	1.63
6b	82	7.62	4.06E-04	2.69E-04	1.51
Sample 405					
1	1	6.71	2.14E-03	1.36E-03	1.57
2	1	6.70	1.26E-03	1.02E-03	1.23
3	4	6.79	8.14E-04	6.51E-04	1.25
4	1	6.68	3.35E-04	2.37E-04	1.41
5	1	6.70	1.42E-04	1.21E-04	1.18
6a	43	7.50	2.09E-04	8.59E-05	2.43
6b	82	7.70	1.83E-04	1.12E-04	1.63
Ox = oxalate.					

This reaction could take place during the contact period subsequent to dissolution of Mn-oxalate from the residual sludge. After dissolution, the relatively low oxalate concentrations and high pH combine to create conditions that are favorable for the oxidation and subsequent precipitation of the Mn as MnOOH (or other Mn oxyhydroxide phase) and concomitant release of protons. Note that the Ox/Mn ratio is significantly greater than 1, suggesting that as Mn-oxalate dissolves and Mn is oxidized and precipitated, free oxalate in solution becomes elevated relative to Mn. The implication of this process for the release model is that, as Mn-oxalate dissolves and releases Mn(II), the Mn oxidizes and precipitates. This would tend to increase the overall dissolution rate of Mn-oxalate and precipitation of Mn oxyhydroxides. It is

likely, that this process will be limited by the amount of dissolved oxygen in the infiltrating water. For example, at 8 mg/L the dissolved oxygen concentration is 2.5×10^{-4} mol/L. From equation 4.1, it is apparent that this could potentially oxidize 5×10^{-4} mol/L of Mn(II).

Chemical equilibrium modeling indicates (Appendix G), that the first sequential contact (sample 404) is significantly oversaturated with respect to rhodochrosite (SI= 1.4). Two explanations are possible to account for this apparent oversaturation with respect to rhodochrosite. The first is that Mn oxyhydroxide colloids formed during the oxidation of Mn(II) and that these colloids escaped filtration and were analyzed as part of the total Mn. An alternative explanation is that Mn(III) was stabilized to some degree as oxalate complexes. The stability constants for Mn(III) oxalate complexes appear to be quite large. For example, an available published value for log K_1 for the formation of the first oxalate complex:

$$Mn^{3+} + C_2O_4^{2-} \leftrightarrow MnC_2O_4^+ \qquad (4.2)$$

is 10.0, determined in 2 mol/L perchloric acid by Taube (1948). The Mn(III) oxalate stability constants of Taube (1948) were determined by an indirect method and are apparently the only published values available. As a result, these constants must be considered as relatively uncertain. The formation of strong Mn(III) oxalate complexes may act to promote the oxidation of Mn(II) to Mn(III) in solution at high pH/pe values.

Although it is conceivable that an equilibrium geochemical model could be constructed to estimate the release of contaminants contained within the Mn phases (Mn-oxalate, rhodochrosite, and Mn oxyhydroxides) of the sludge, several factors arise that put significant constraints on the reliability of this approach. The most important factor is that the distribution of the contaminants of concern among the various phases is still very uncertain at this point. In addition, it is not clear what would happen to the contaminants of concern released during the dissolution of Mn oxalate. For example, available evidence suggests that, as Mn oxalate dissolves, Mn oxyhydroxides or rhodochrosite could precipitate. Contaminants released along with the Mn oxalate could be free to migrate or they might adsorb to the newly formed Mn oxyhydroxides or co-precipitate with rhodochrosite.

As a result of these uncertainties, a release model has been developed from empirical solubility data for the contaminants of concern determined from concentration measurements obtained from the water leach tests. This model is described in Section 4.2.

4.2   Technetium, Uranium, Iodine, and Chromium Release Models

Because of the highly complex chemical nature of tank C-106 residual sludge, clear and quantitative phase associations of the contaminants of concern with the phases known to exist in the residual sludge are difficult to specify. Although the various characterization methods employed in this study have revealed a number of important observations and have provided valuable data for constructing a scientifically defensible release model, many questions remain. Because a thorough understanding of all the important phase associations for the contaminants of concern cannot be developed at this time, an empirically based release model has been developed. Although less satisfying from a mechanistic point of view, this provides a release model that can be used now and is conservative in nature. Later work

may provide a better understanding of the phase associations with the contaminants of concern and the release mechanisms from these phases. In this case, a less conservative, but more scientifically defensible release model could be developed.

To estimate the maximum release rates for ^{99}Tc, ^{238}U, and Cr, their concentrations measured in the single-contact and sequential water extractions were evaluated. The highest concentrations in water correlate with the highest release rates from the sludge. In general, the single-contact water extracts contained the highest concentrations of the contaminants of concern. It was then determined which time period (1 day, 2 week, or 1 month) for the single-contact water-leach tests had the highest concentrations. Once the appropriate time period was selected, the maximum solution concentrations of the 404 and 405 samples were averaged to determine a release concentration. In the case of ^{129}I, data are only available for the 1-day single-contact water extract. These values are provided in Table 4.3 (column 3). The total concentrations were determined from either the fusion or EPA acid digestions of the sludge, whichever had the highest concentration. These totals were then averaged for the primary and duplicate samples for both samples 404 and 405. The results are provided in Table 4.3, column 2.

Table 4.3. Summary of Contaminant Release Model Data for C-106

Contaminant	Sludge Concentration	Release Concentration	Release Control
^{99}Tc	1.2 µg ^{99}Tc/g-sludge (0.2 pCi ^{99}Tc/g-sludge)	0.21 µg/L (3.6 pCi/L)	solubility
^{238}U	310 µg ^{238}U/g-sludge	46 µg/L	solubility
^{129}I	0.62 µg ^{129}I/g-sludge (110 pCi ^{129}I/g-sludge)	0.059 µg/L (10 pCi/L)	solubility
Cr	897 µg Cr/g-sludge	19 µg/L	solubility

These suggested empirical constant concentration (solubility) limits are based on contact with water that resembles natural infiltration (i.e., rainwater). Water modified by grout placed in the tank or any other amendments could significantly alter the solubilities (hence release concentrations) of these contaminants.

5.0   Conclusions

This report provides the results of laboratory tests on residual sludge and liquid samples from Hanford tank C-106 and describes the development of source term release models for the primary contaminants of concern. The major conclusions from this work are discussed in this section.

- The geochemistry of the residual sludge in tank C-106 is complex because of the variety of minerals present and the impact of chemical treatment of the waste with oxalic acid and NaOH during retrieval. The initial (pre-retrieved) waste consisted of the minerals gibbsite [$Al(OH)_3$], hematite [Fe_2O_3], dawsonite [$NaAlCO_3(OH)_2$], cancrinite [$Na_6Ca_{1.5}Al_6Si_6O_{24}(CO_3)_{1.6}$], and sidorenkite ($NaMnPO_4CO_3$) (Bechtold et al. 2003). It is possible that poorly crystalline Mn and Fe oxyhydroxides also occurred in the sludge. ^{99}Tc, ^{238}U, ^{129}I, and Cr occurred as trace constituents in these minerals. The oxalic acid treatment method for retrieval dissolved and removed approximately 75% of the sludge, but preferentially removed Fe (and Cr) from the sludge while concentrating Al and Mn in the residual solid. It appears that Al was concentrated in the sludge because gibbsite was not very soluble in the oxalic acid solution. Mn was concentrated in the residual sludge because most of the Mn that entered the solution from the dissolution of Mn minerals reprecipitated as Mn oxalate and, perhaps, Mn oxyhydroxide minerals. Some of the primary contaminants remained in the undissolved sludge probably associated with the Fe minerals. Some of the contaminants released to solution became associated, and immobilized, with the precipitating Mn minerals. Furthermore, the precipitating Mn minerals appear to have coated many of the original sludge particles, thereby affecting their future interaction with water that might come into contact with the residual sludge. The result is a complicated geochemical system in which the contaminants are present at trace levels associated with a variety of minerals with solubilities that are dependent on each other.

- Mechanistic release models of contaminants from the sludge could not be developed from the available data because of the complexity of the geochemical system. However, empirical release models based on measured total sludge concentrations and maximum solution concentrations in water-leaching tests can be used for performance/risk assessment modeling. These empirical release models are expected to provide conservatively high release rates into recharge water percolating through the residual waste. The values for the chosen empirical constant concentration release models are:

Contaminant	Sludge Concentration	Release Concentration	Release Control
^{99}Tc	1.2 µg ^{99}Tc/g-sludge (0.2 pCi ^{99}Tc/g-sludge)	0.21 µg/L (3.6 pCi/L)	solubility
^{238}U	310 µg ^{238}U/g-sludge	46 µg/L	solubility
^{129}I	0.62 µg ^{129}I/g-sludge (110 pCi ^{129}I/g-sludge)	0.059 µg/L (10 pCi/L)	solubility
Cr	897 µg Cr/g-sludge	19 µg/L	solubility

- The concentrations of major metals and anions in the residual sludge were:

    o Al – 131,483 µg/g
    o Mn – 117,767 µg/g
    o Fe – 43,777 µg/g
    o Ca – 38,221 µg/g
    o Na – 60,400 µg/g
    o oxalate – 63,900 µg/g
    o carbonate – 39,500 µg/g

- The sum of the TRU constituents (^{239}Pu, ^{237}Np, and ^{241}Am) measured in the tank C-106 residual sludge samples by the fusion method was 8,057 nCi/g; by the EPA acid digestion method it was 8,670 nCi/g. Based on the DOE definition of TRU waste (>100 nCi/g), this would classify the residual sludge itself in tank C-106 as TRU waste. However, the final waste form in the tank will be a mixture of residual sludge and a tank filling grout, which will lower the concentrations of contaminants an amount dependent on the mixing ratio of the sludge and grout.

- The oxalic acid treatment of the sludge in the tank during retrieval removed much of the water-leachable constituents from the solid. Less than 3% of the ^{129}I and less than 5% of the ^{99}Tc and ^{238}U in the sludge were water leachable during a variety of single and multiple-contact water leach tests. Leachable amounts of Al and Fe were barely detectable, while about 6% of the Ca, 40% of the Mn and 50% of the Na were water leachable. The primary water leachable anions were oxalate and carbonate.

- Several of the metals and contaminants could not be leached from the sludge even under the condition of aggressive leaching with concentrated nitric acid. It was found that 56% of the ^{99}Tc could not be leached under these conditions and 40% of the Cr was recalcitrant. Fe and Al were also relatively immobile with 65% and 41% not leachable, respectively.

- The XRD results indicate that the unleached sludge samples contain detectable quantities of the following crystalline phases:

    o lindbergite [$MnC_2O_4 \cdot 2H_2O$]
    o gibbsite [$Al(OH)_3$]
    o dawsonite [$NaAlCO_3(OH)_2$]
    o hematite (Fe_2O_3)
    o böhmite [$AlO(OH)$]
    o rhodochrosite ($MnCO_3$)
    o whewellite (Ca oxalate monohydrate, $CaC_2O_4 \cdot H_2O$)

Except for lindbergite, these minerals were also present in the 1-month and 82-day water-leached and HF-extracted sludge samples. A possible Ag-Hg phase was identified by a single reflection, and may be present in the sludge. Amorphous phases of Al and Fe that are not detectable by XRD are also likely to be present in the sludge.

SEM/EDS analysis of the sludge showed a wide variety of morphologies, sizes, surface textures and compositions. Particles range in size from submicrometer to a few hundred micrometers. An Al-rich, multi-faceted, blocky shaped particle is common to all of the samples and may represent gibbsite. Another common occurrence is particles containing Mn, Al, Fe, Na, P, Si, Ca, O, and possibly C and H that have distinctive shapes (oblong and rounded, orthorhombic, rhombohedral, and pyramidal rhombic). Coatings are present on many of these particles, and may account for some of the elements present in the analysis. The surfaces of the Fe oxide particles contain pits and solution cavities. Cr and other contaminants are likely associated with this phase. These contaminants will only be released as the host Fe oxide solid(s) dissolves. Other solids analyzed by EDS have the following compositions:

- Ag-Hg±O±H
- Ca-O±C±H
- Mn-O-C±H
- Al-Na-O-C±H
- Mn-O-P±Al±C±H
- Si-Al-Na-O±C±H
- Fe-Cr-O±C±H
- Fe-Mn-O±C±H
- Rare earth element (REE)-rich
- Ca-Si-Al-O±C±H (rare)

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Appendix A

**X-Ray Diffraction Patterns for Unleached,
Water-Leached, and HF-Extracted Sludge Samples
from Tank C-106**

Appendix A

X-Ray Diffraction Patterns for Unleached, Water-Leached, and HF-Extracted Sludge Samples from Tank C-106

This appendix presents the as-measured and background-subtracted X-ray powder diffraction (XRD) patterns for the unleached (as-received), 1-month and 82-day water-leached, and hydrofluoric (HF)-extracted sludge samples 404 and 405 from tank 241-C-106 (C-106). The instrumentation and procedures used for measuring, subtracting background, and interpreting the XRD patterns for these materials are described in the main report. The vertical axis in each of the following patterns represents the intensity in counts per second (cps) of the XRD peaks. The horizontal axis is in terms of degrees 2θ based on Cu$_{K\alpha}$ radiation (λ=1.5406 Å), and is related to *d* spacing according to the Bragg law (Cullity 1956).[a] For comparison to the background signal in the as-measured XRD patterns included in this appendix, Figure A.1 shows the XRD pattern for collodion film measured in the absence of any sludge material and reported by Krupka et al. (2004).[b]

Figure A.1. XRD Pattern for Collodion-Solution Film (from Krupka et al. 2004)
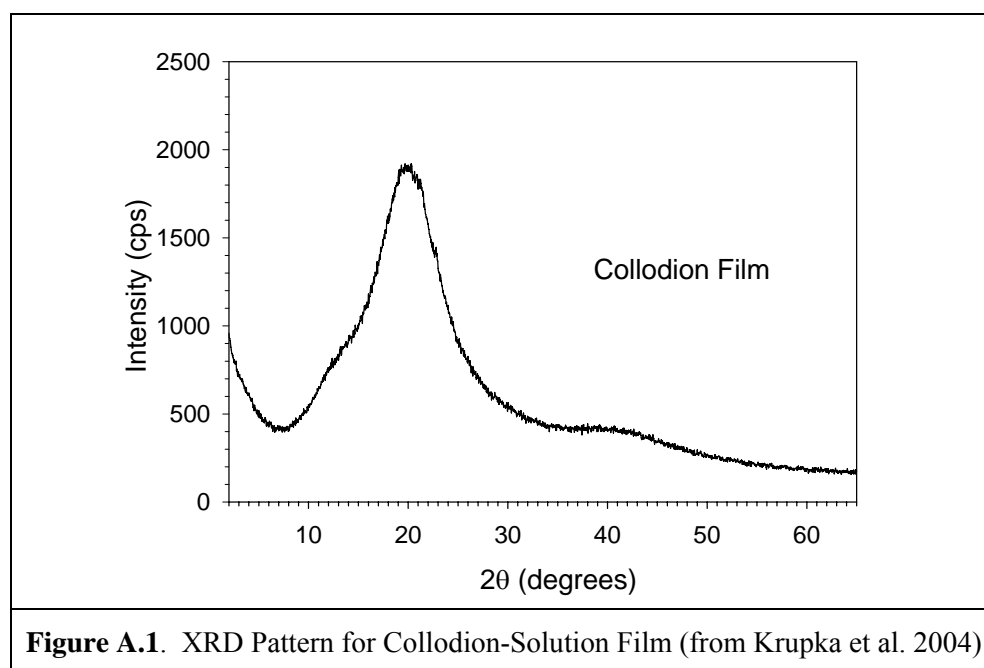
(a) Cullity BD. 1967. *Elements of X-Ray Diffraction*. Addison-Wesley Publishing Company, Inc., Reading, Massachusetts.

(b) Krupka KM, WJ Deutsch, MJ Lindberg, KJ Cantrell, NJ Hess, HT Schaef, and BW Arey. 2004. *Hanford Tanks 241-AY-102 and 241-BX-101: Sludge Composition and Contaminant Release Data*. PNNL-14614, Pacific Northwest National Laboratory, Richland, Washington.

Figure A.2.   As-Measured XRD Pattern (without background subtraction) for Unleached (as-received) Sludge Sample 404

Figure A.3.   Background-Subtracted XRD Pattern for Unleached (as-received) Sludge Sample 404

Figure A.4. As-Measured XRD Pattern (without background subtraction) for Unleached (as-received) Sludge Sample 405
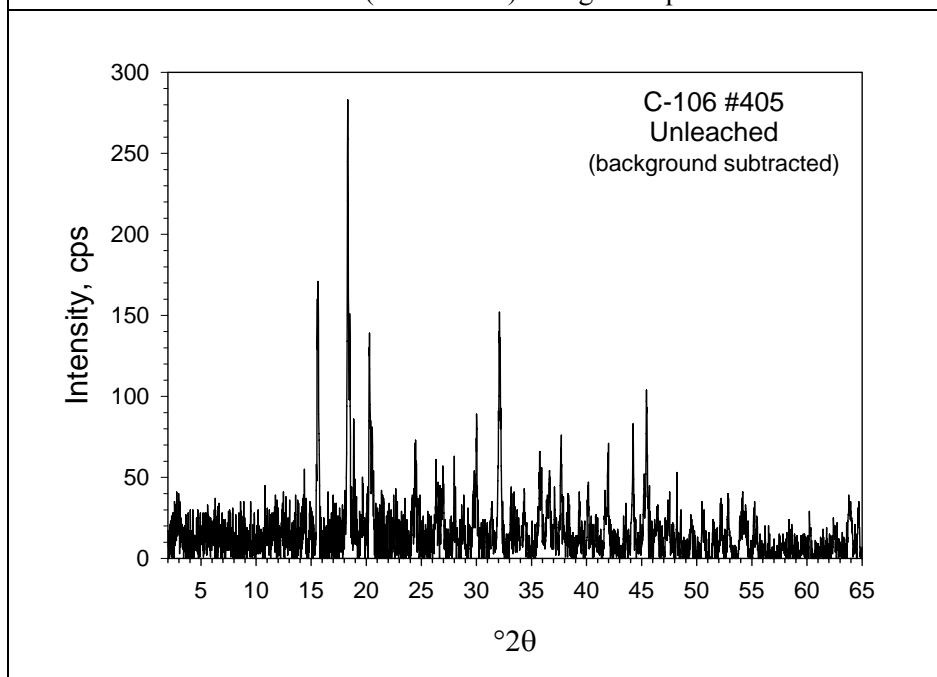
Figure A.5. Background-Subtracted XRD Pattern for Unleached (as-received) Sludge Sample 405
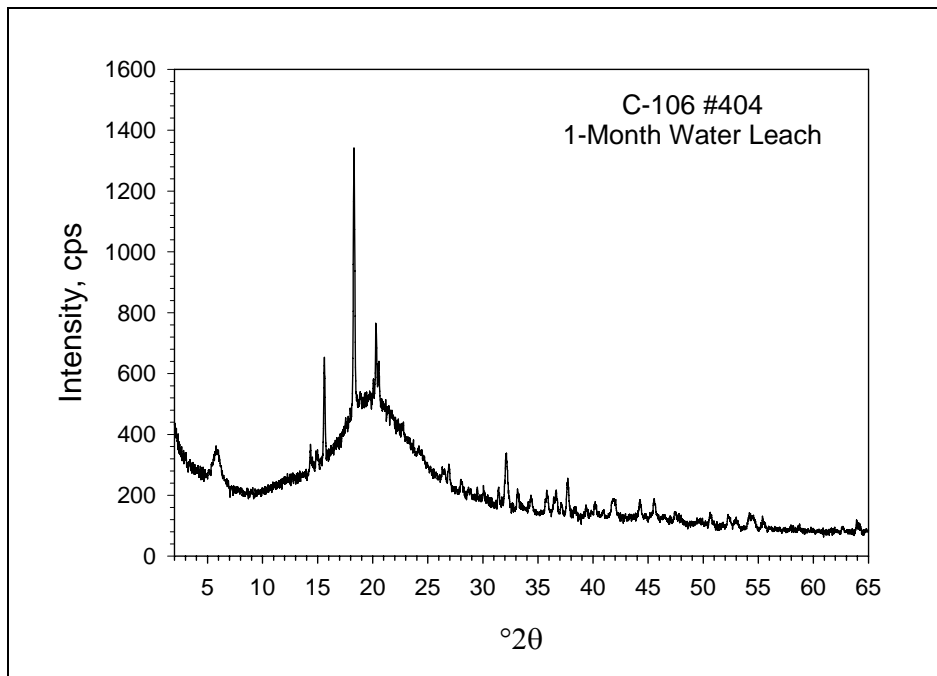
Figure A.6.   As-Measured XRD Pattern (without background subtraction) for 1-Month Water-Leached Sludge Sample 404
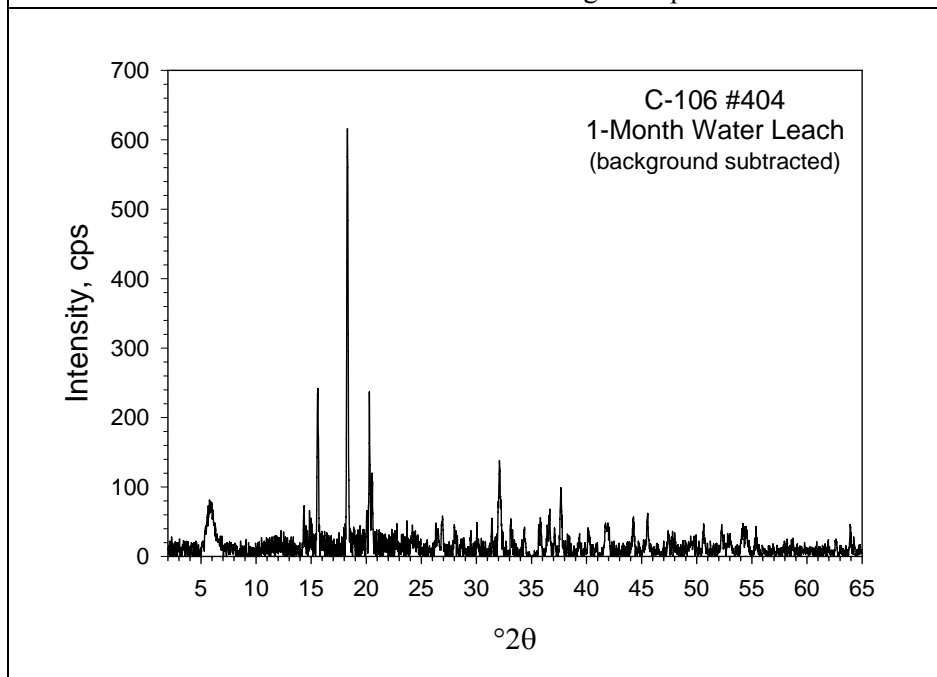
Figure A.7.   Background-Subtracted XRD Pattern for 1-Month Water-Leached Sludge Sample 404
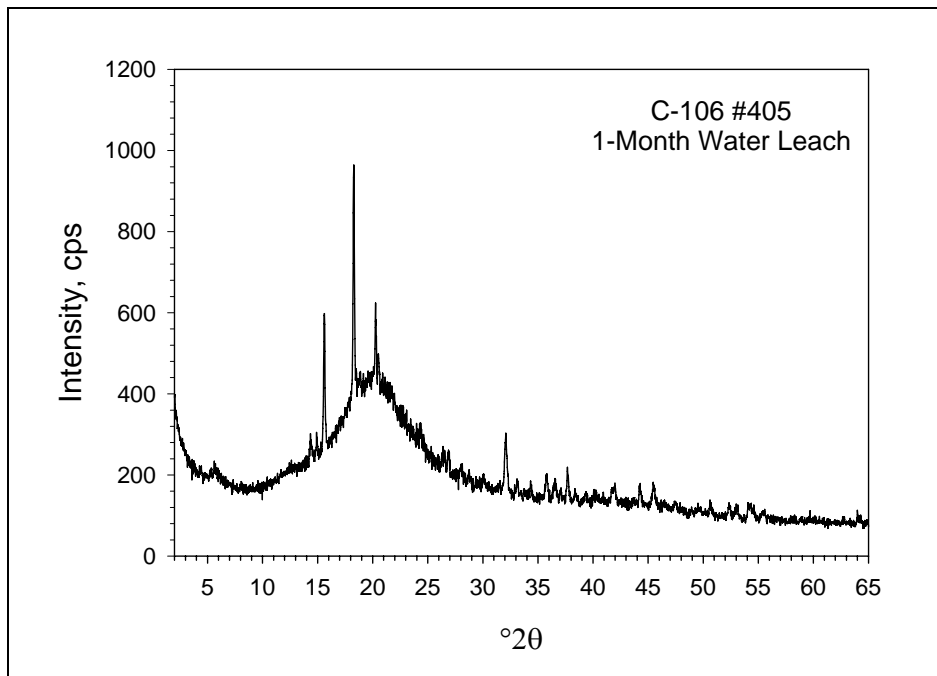
Figure A.8. As-Measured XRD Pattern (without background subtraction) for 1-Month Water-Leached Sludge Sample 405
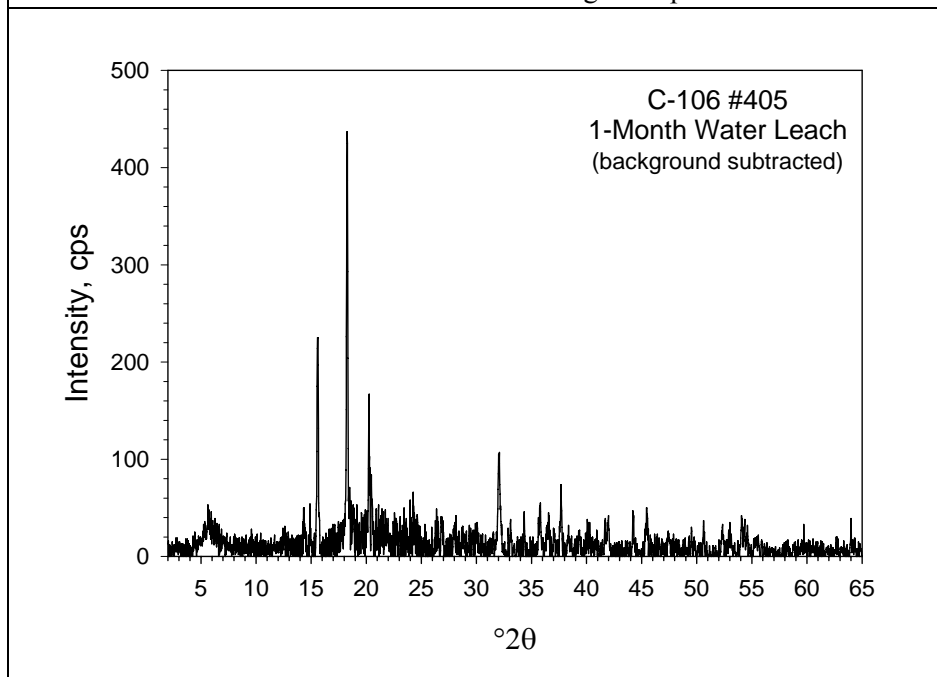
Figure A.9. Background-Subtracted XRD Pattern for 1-Month Water-Leached Sludge Sample 405

A.5
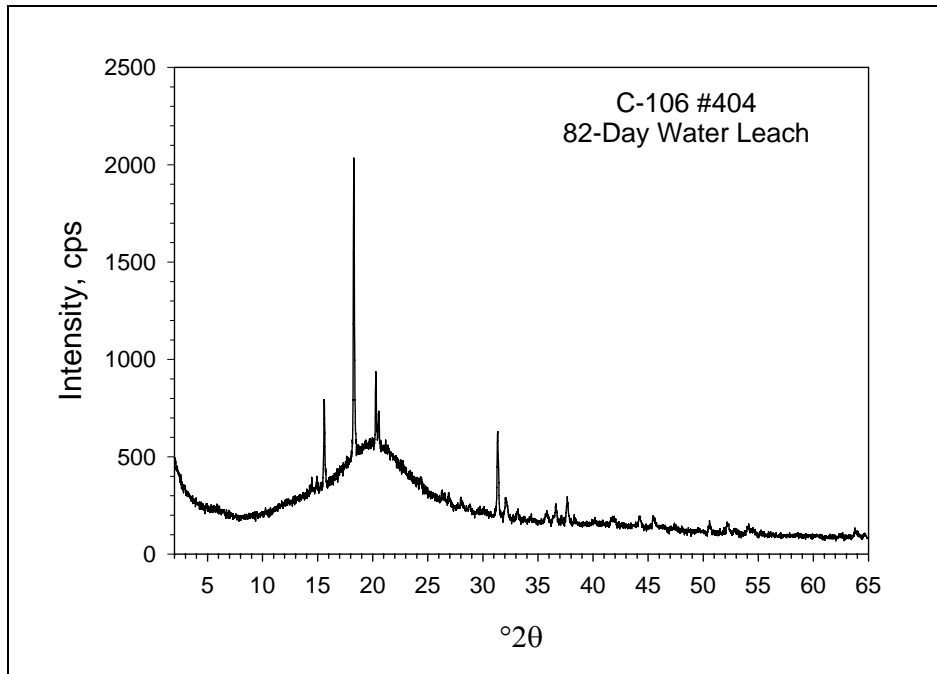
Figure A.10. As-Measured XRD Pattern (without background subtraction) for 82-day Water-Leached Sludge Sample 404
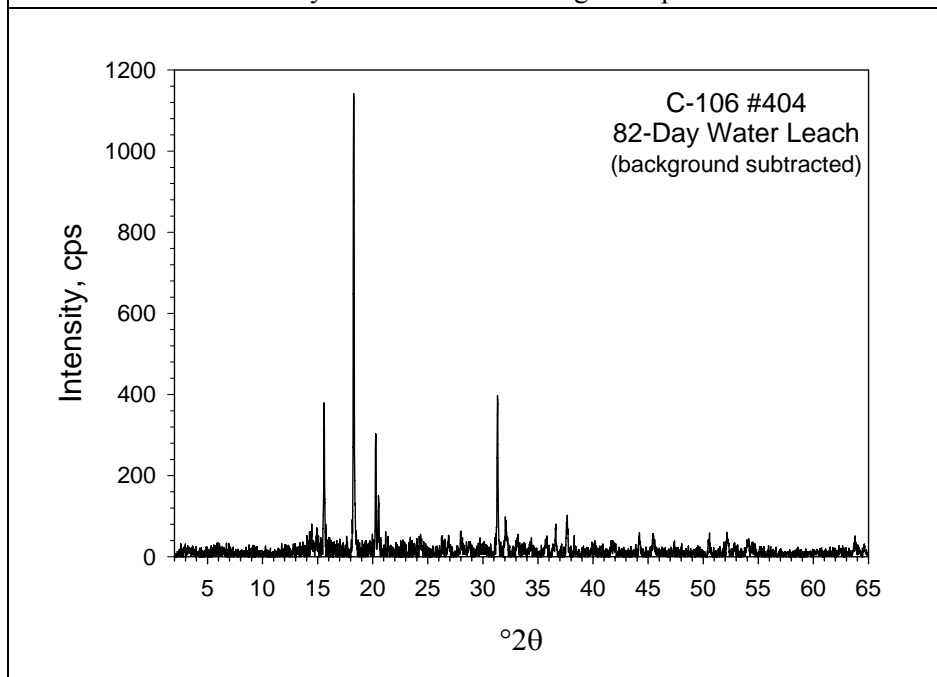
Figure A.11. Background-Subtracted XRD Pattern for 82-day Water-Leached Sludge Sample 404
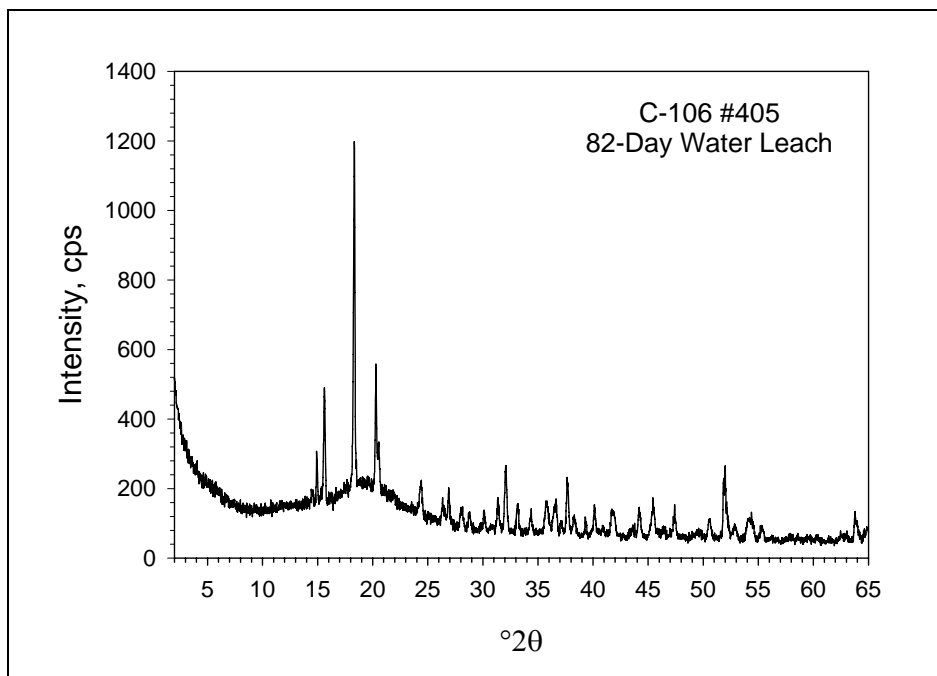
Figure A.12.   As-Measured XRD Pattern (without background subtraction) for 82-day Water-Leached Sludge Sample 405
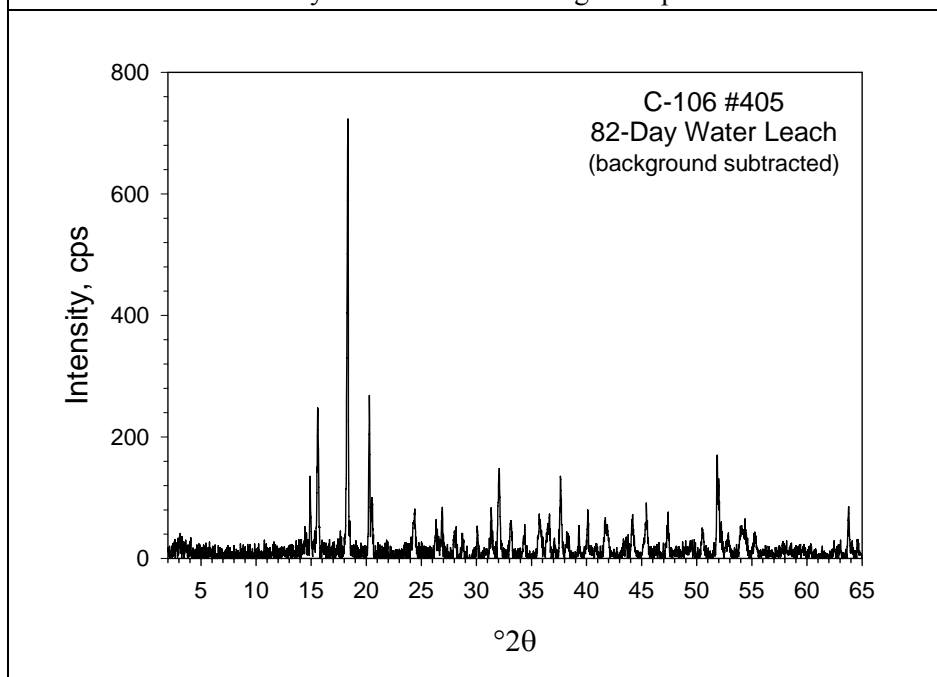
Figure A.13.   Background-Subtracted XRD Pattern for 82-day Water-Leached Sludge Sample 405
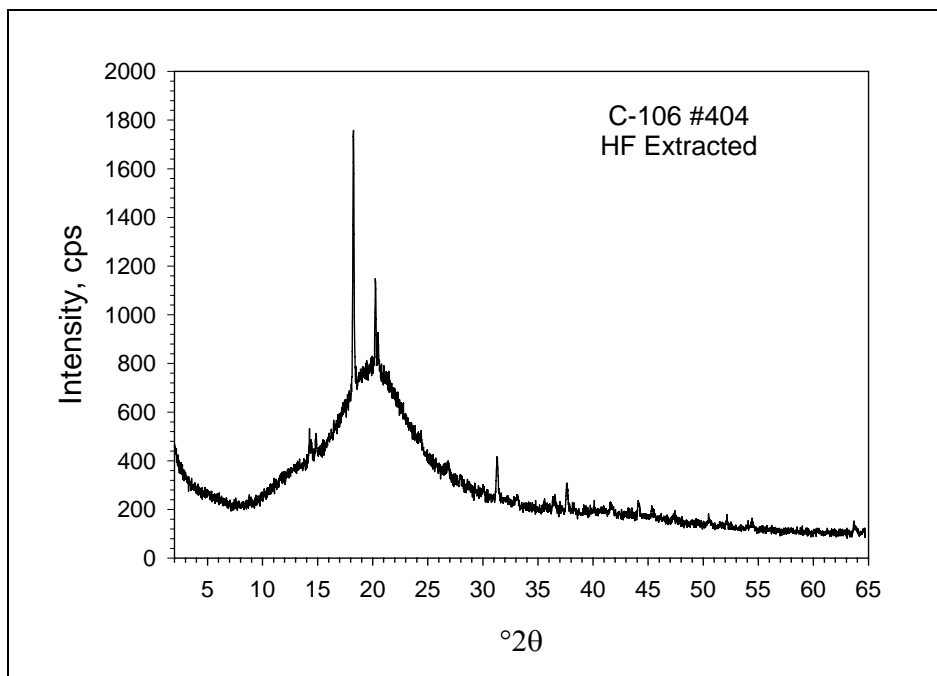
Figure A.14.　As-Measured XRD Pattern (without background subtraction) for HF-Extracted Sludge Sample 404

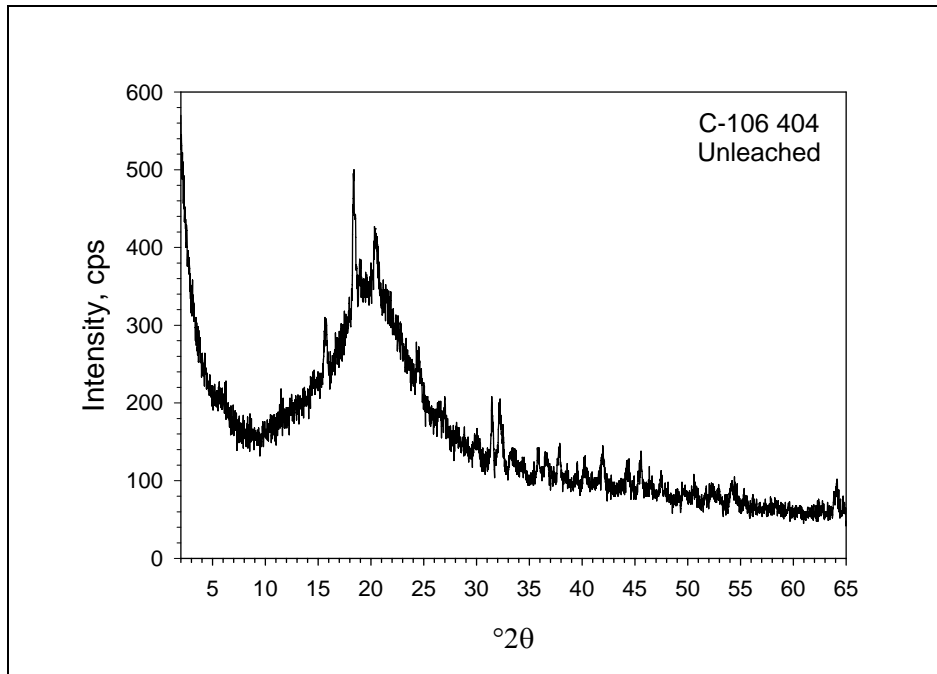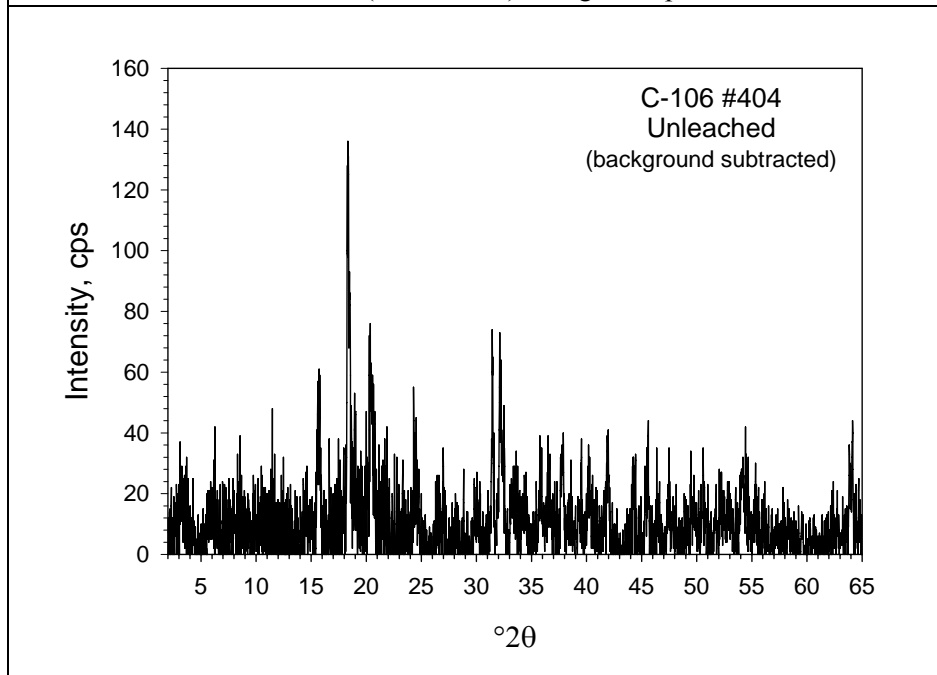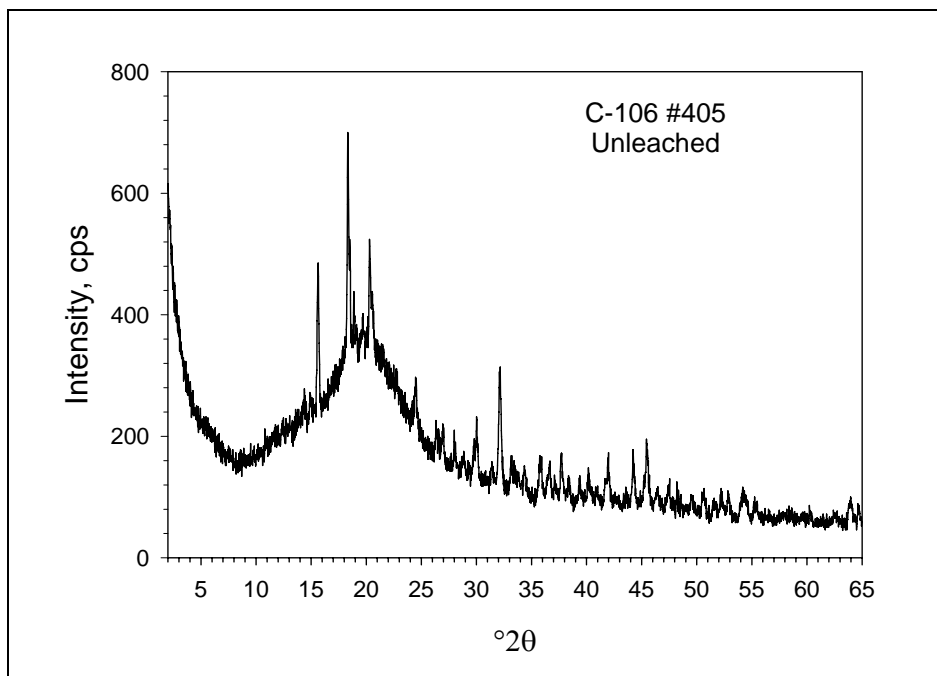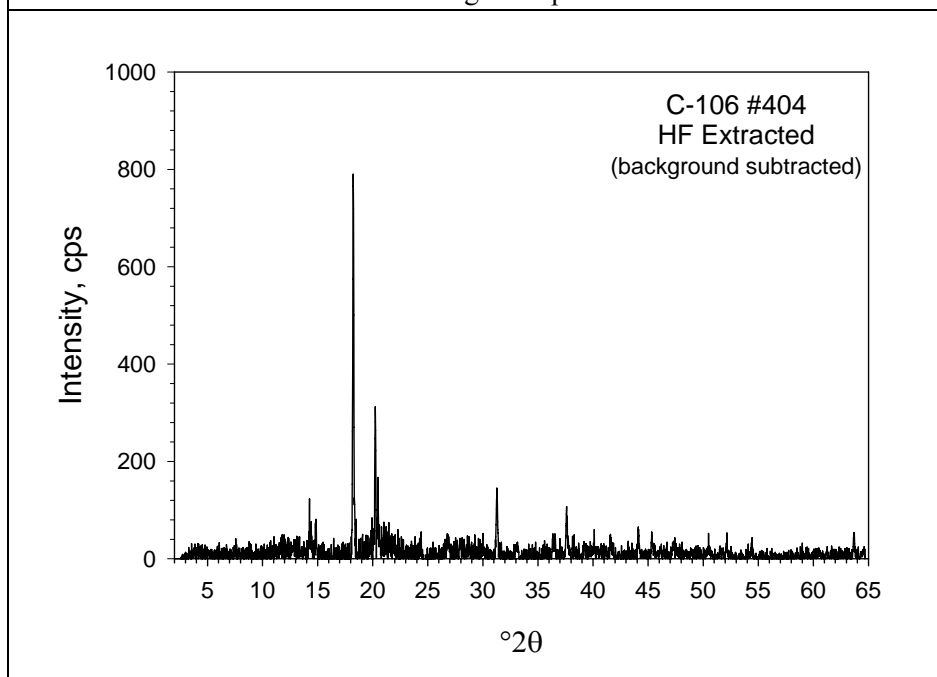**Figure A.15**.　Background-Subtracted XRD Pattern for HF-Extracted Sludge Sample 404

A.8
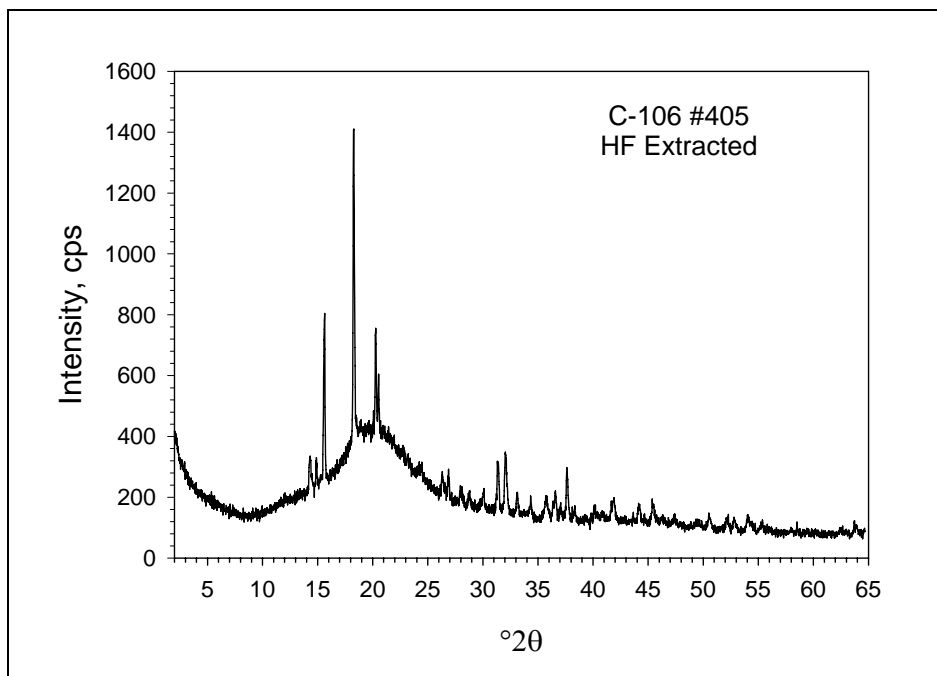
Figure A.16.   As-Measured XRD Pattern (without background subtraction) for HF-Extracted Sludge Sample 405
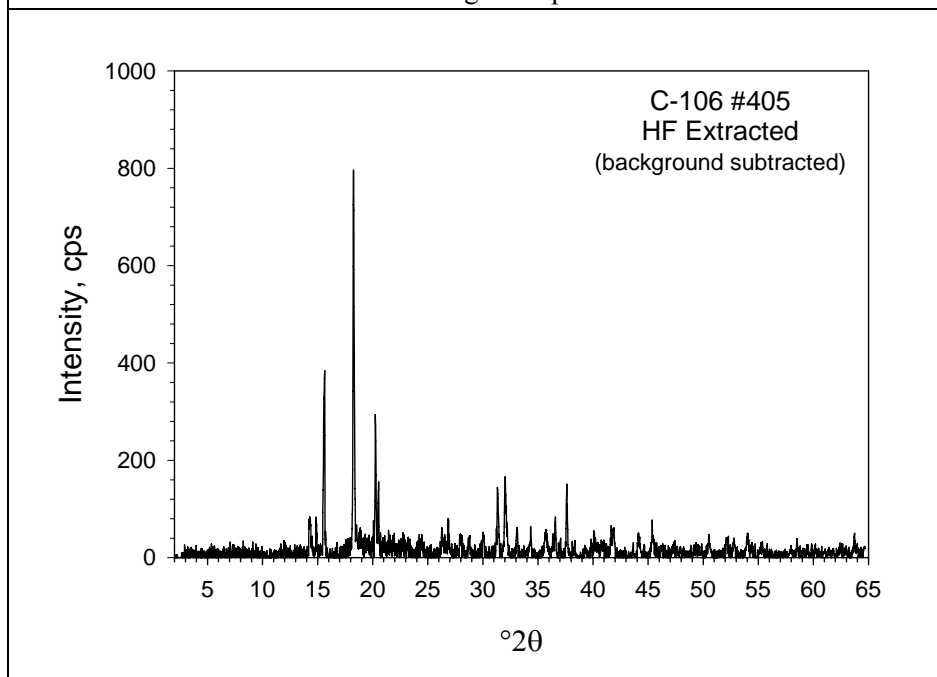
Figure A.17.   Background-Subtracted XRD Pattern for HF-Extracted Sludge Sample 405

A.9

Appendix B

SEM Micrographs and EDS Spectra
for Unleached Tank C-106 Sludge

Appendix B

SEM Micrographs and EDS Spectra for Unleached
Tank C-106 Sludge

This appendix includes the scanning electron microscope (SEM) micrographs and the energy-dispersive X-ray spectrometry (EDS) spectra for samples of unleached residual tank waste from tank 241-C-106 (C-106) (primary 404 and duplicate 405). The operating conditions for the SEM and procedures used for mounting the SEM samples are described in Section 2.4 of the main report.

The name of each SEM digital image file, sample identification number, and a size scale bar are given, respectively, at the bottom left, center, and right of each SEM micrograph in this appendix. Micrographs labeled by "BSE" to the immediate right of the digital image file name indicate that the micrograph was collected with backscattered electrons. Areas identified by a letter and/or outlined by a dashed-line square in a micrograph designate sample material that was imaged at higher magnification, which is typically shown in figure(s) that immediately follow in the series for that sample.

Areas labeled by "eds" in the following SEM micrographs in this appendix identify locations of particles for which EDS spectra were recorded. The "eds" label given with each EDS spectrum correspond to the same "EDS" label used in the SEM micrographs for this sample.

B.1 Unleached Sludge Sample 404

Two mounts of unleached (raw) residual sludge from tank C-106 (primary 404) were analyzed by SEM/EDS. The SEM micrographs for the first mount (1) of sample 404 sludge are shown in Figures B.1 through B.22. The EDS spectra for this mount are given in Figures B.23 through B.27. The SEM micrographs for the second mount (3) of sample 404 sludge are shown in Figures B.28 through B.35, and the EDS spectra for the second mount are given in Figures B.36 through B.38.
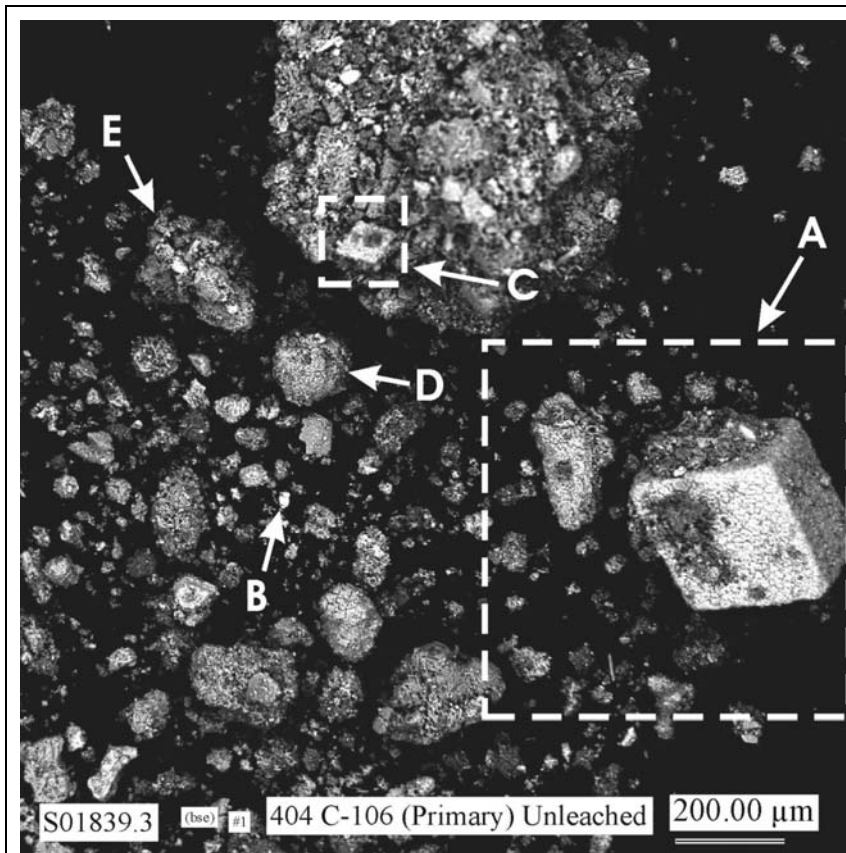
Figure B.1. Low Magnification SEM Micrograph Showing General Morphologies of Particles in SEM Mount 1 of Unleached Sludge Sample 404
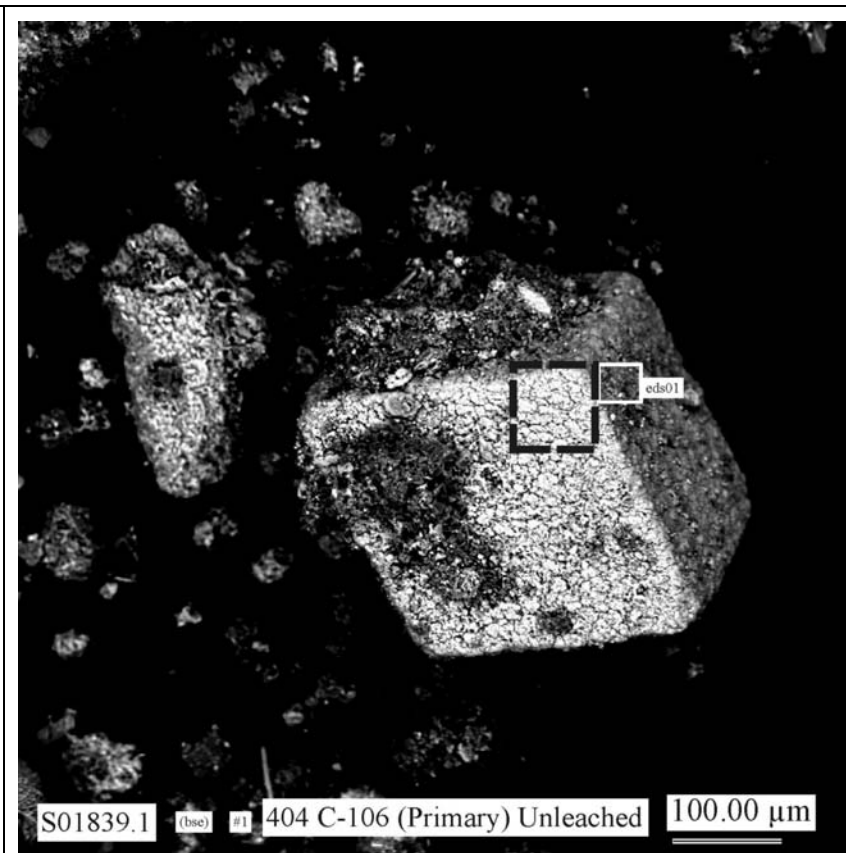
Figure B.2. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-line Square Labeled A in Figure B.1
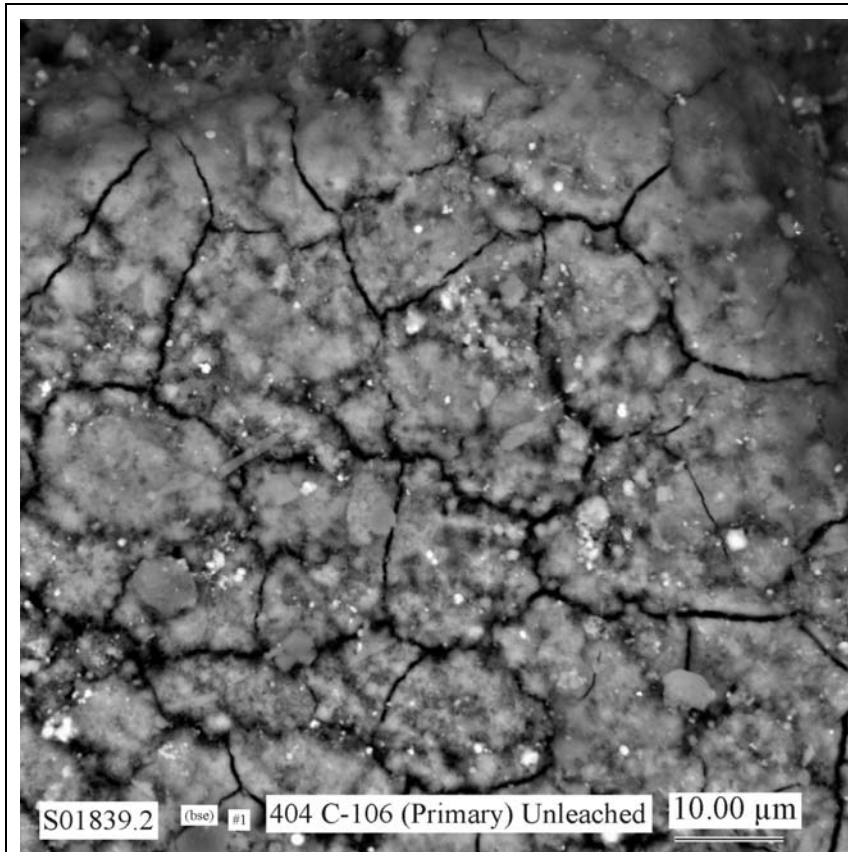
Figure B.3. Micrograph Showing at Higher Magnification the Area Indicated by the Black Dashed-Line Square in Figure B.2
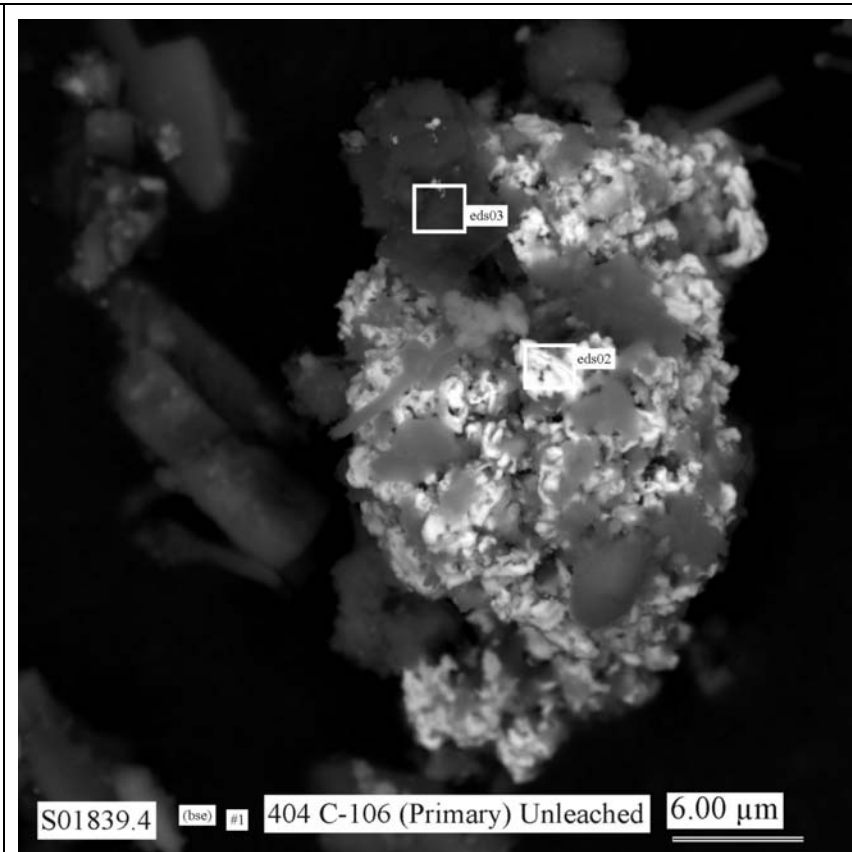
Figure B.4. Micrograph Showing at Higher Magnification the Particle Labeled B in Figure B.1
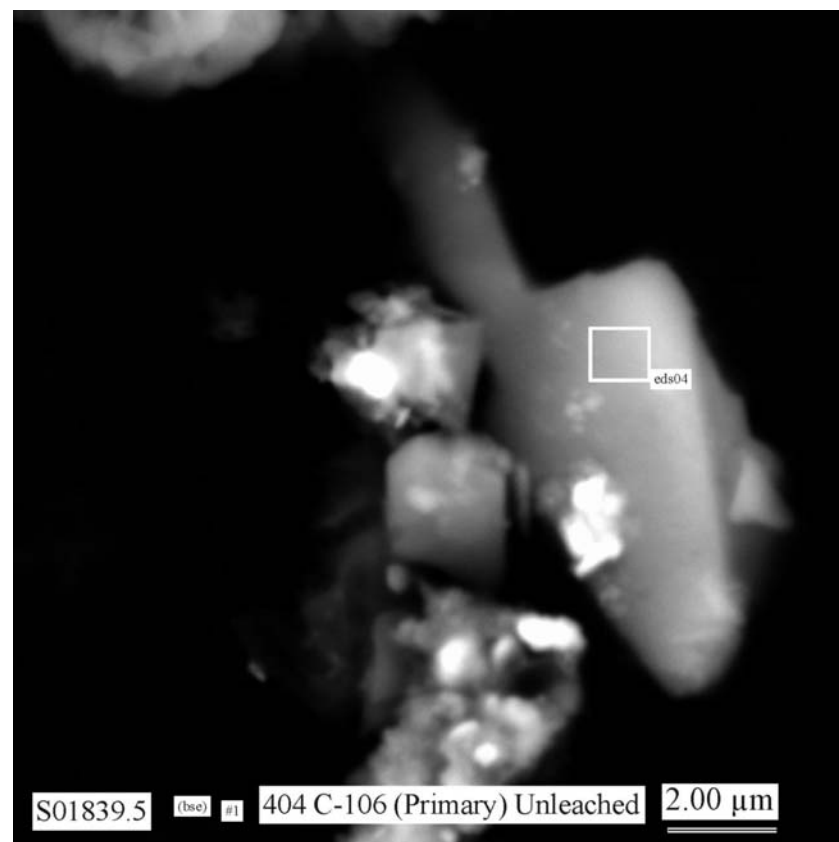
Figure B.5. Micrograph Showing at Higher Magnification a "Bright" Particle Present in the Field of View in Figure B.1

Figure B.6. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled C in Figure B.1

Figure B.7. Micrograph Showing at Higher Magnification the Particle Labeled D in Figure B.1

Figure B.8. Micrograph Showing at Higher Magnification the Particle Labeled E in Figure B.1

Figure B.9. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square in Figure B.8

Figure B.10. Micrograph Showing Morphologies of Typical Particles in SEM Mount 1 of Unleached Sludge Sample 404

Figure B.11. Micrograph Showing Morphologies of Typical Particles in SEM Mount 1 of Unleached Sludge Sample 404

Figure B.12. Micrograph Showing Morphologies of Typical Particles in SEM Mount 1 of Unleached Sludge Sample 404

S01839.14 (bse) #1 404 C-106 (Primary) Unleached 10.00 μm

eds13

Figure B.13. Micrograph Showing Morphologies of Typical Particles in SEM Mount 1 of Unleached Sludge Sample 404

S01839.15 (bse) #1 404 C-106 (Primary) Unleached 80.00 μm
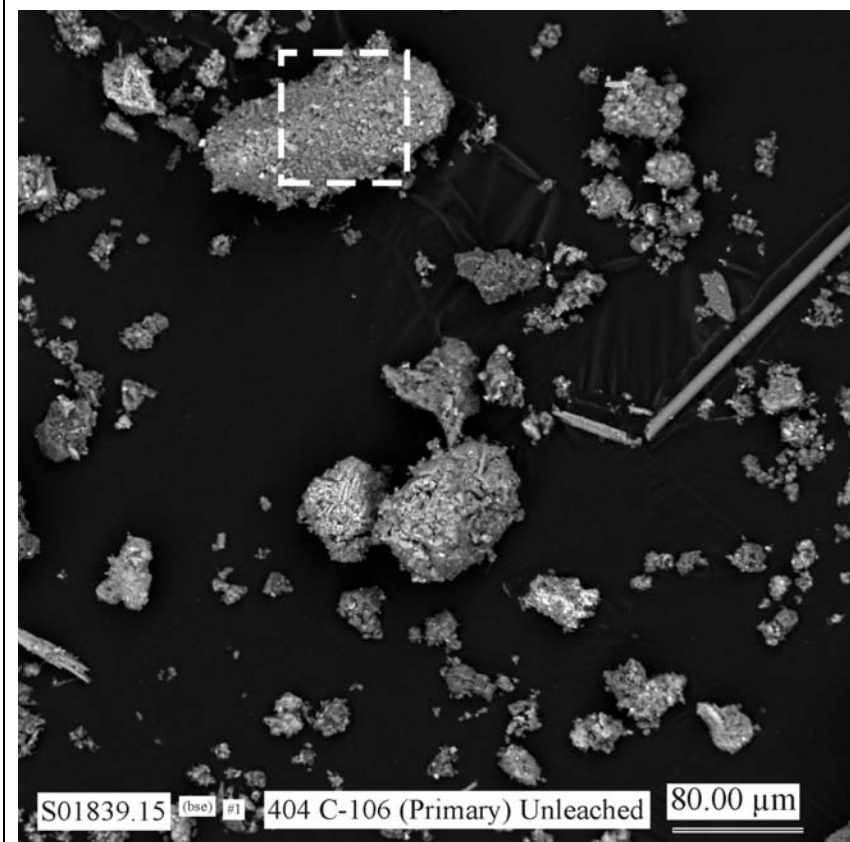
Figure B.14. Micrograph Showing Morphologies of Typical Particles in SEM Mount 1 of Unleached Sludge Sample 404
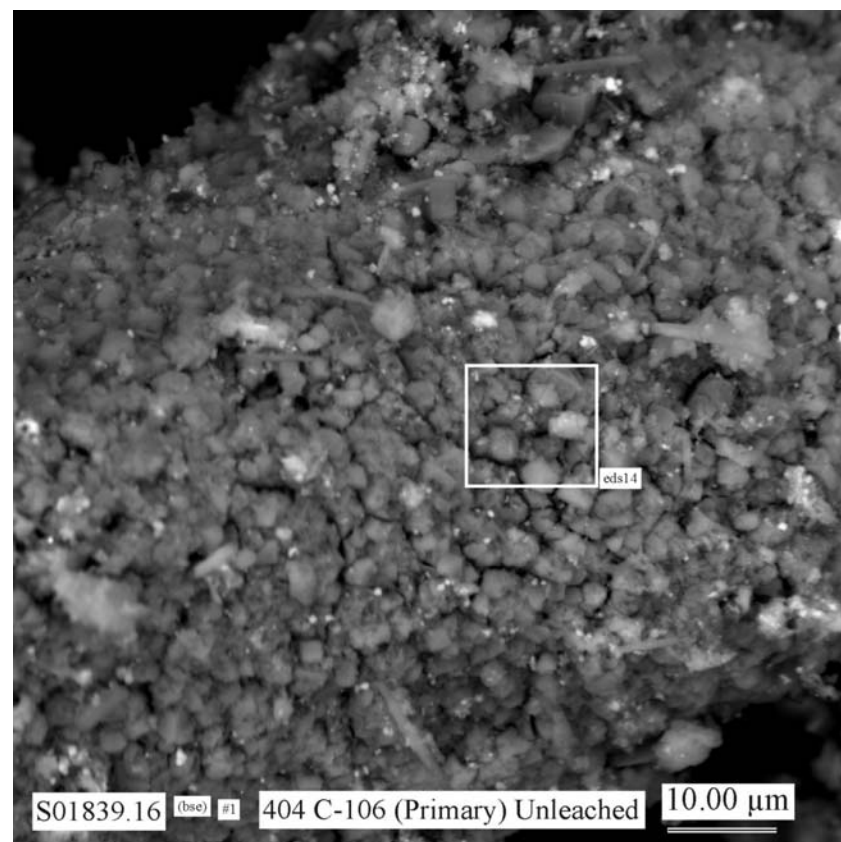
Figure B.15. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square in Figure B.14
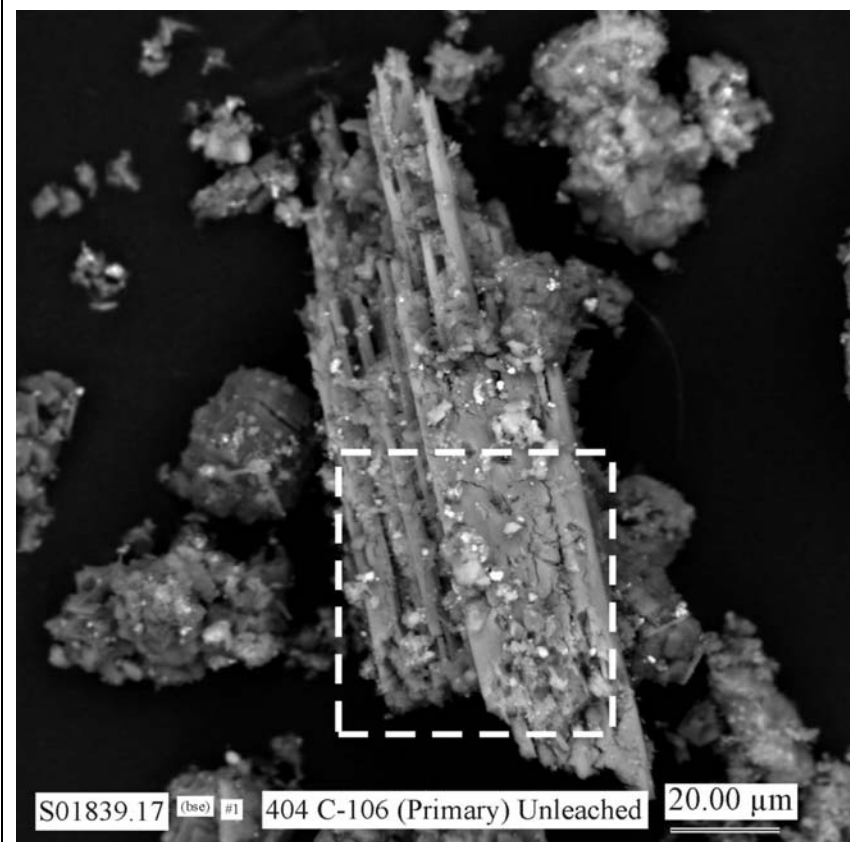
Figure B.16. Micrograph Showing Morphologies of Particles in SEM Mount 1 of Unleached Sludge Sample 404

Figure B.17. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square in Figure B.16

Figure B.18. Micrograph Showing Morphologies of Typical Particles in SEM Mount 1 of Unleached Sludge Sample 404

S01839.20 (bse) #1 404 C-106 (Primary) Unleached 8.00 μm

eds17

S01839.21 (bse) #1 404 C-106 (Primary) Unleached 10.00 μm

eds18

| **Figure B.19.** | Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square in Figure B.18 | **Figure B.20.** | Micrograph Showing Morphologies of Typical Particles in SEM Mount 1 of Unleached Sludge Sample 404 |

eds19

S01839.22 (bse) #1 404 C-106 (Primary) Unleached 10.00 μm

Figure B.21. Micrograph Showing Morphologies of Typical Particles in SEM Mount 1 of Unleached Sludge Sample 404

S01839.23 (bse) #1 404 C-106 (Primary) Unleached 3.00 μm

Figure B.22. Micrograph Showing at Higher Magnification the "Bright" Particles in the Area Indicated by the White Dashed-Line Square in Figure B.18

| C-106 404 (#1) Primary Unleached eds01 [See Figure B.2] | C-106 404 (#1) Primary Unleached eds02 [See Figure B.4] |
| C-106 404 (#1) Primary Unleached eds03 [See Figure B.4] | C-106 404 (#1) Primary Unleached eds04 [See Figure B.5] |

Figure B.23.   EDS Spectra for Analyses eds01 Through eds04 for Particles in SEM Mount 1 of Unleached Sludge Sample 404

C-106 404 (#1) Primary Unleached eds05 [See Figure B.9]	C-106 404 (#1) Primary Unleached eds06 [See Figure B.9]
C-106 404 (#1) Primary Unleached eds07 [See Figure B.9]	C-106 404 (#1) Primary Unleached eds08 [See Figure B.10]

Figure B.24. EDS Spectra for Analyses eds05 Through eds08 for Particles in SEM Mount 1 of Unleached Sludge Sample 404

C-106 404 (#1) Primary Unleached eds09 [See Figure B.11]	C-106 404 (#1) Primary Unleached eds10 [See Figure B.12]
C-106 404 (#1) Primary Unleached eds11 [See Figure B.12]	C-106 404 (#1) Primary Unleached eds12 [See Figure B.12]

Figure B.25.   EDS Spectra for Analyses eds09 Through eds12 for Particles in SEM
Mount 1 of Unleached Sludge Sample 404

C-106 404 (#1) Primary Unleached
eds13 [See Figure B.13]

C-106 404 (#1) Primary Unleached
eds14 [See Figure B.15]

C-106 404 (#1) Primary Unleached
eds15 [See Figure B.17]

C-106 404 (#1) Primary Unleached
eds16 [See Figure B.17]

Figure B.26.   EDS Spectra for Analyses eds13 Through eds16 for Particles in SEM
Mount 1 of Unleached Sludge Sample 404

C-106 404 (#1) Primary Unleached
eds17 [See Figure B.19]

C-106 404 (#1) Primary Unleached
eds18 [See Figure B.20]

C-106 404 (#1) Primary Unleached
eds19 [See Figure B.21]

Figure B.27.   EDS Spectra for Analyses eds17 Through eds19 for Particles in SEM
Mount 1 of Unleached Sludge Sample 404

Figure B.28. Low Magnification SEM Micrograph Showing General Morphologies of Particles in SEM Mount 3 of Unleached Bulk Sludge Sample 404

Figure B.29. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled A in Figure B.28
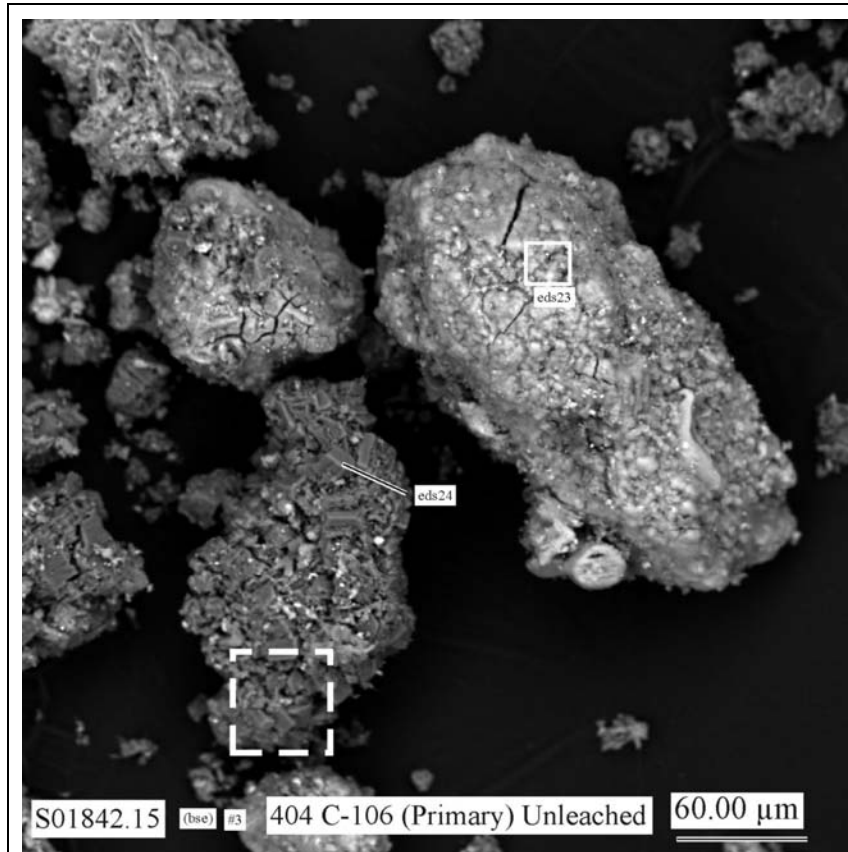
Figure B.30. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled B in Figure B.28
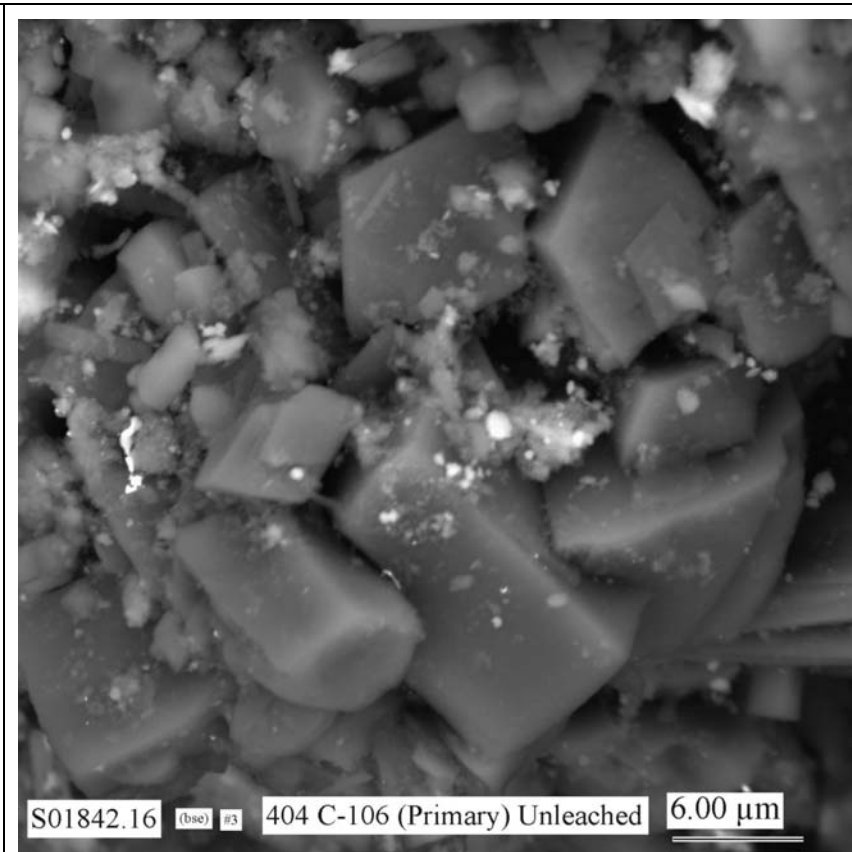
Figure B.31. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square in Figure B.30
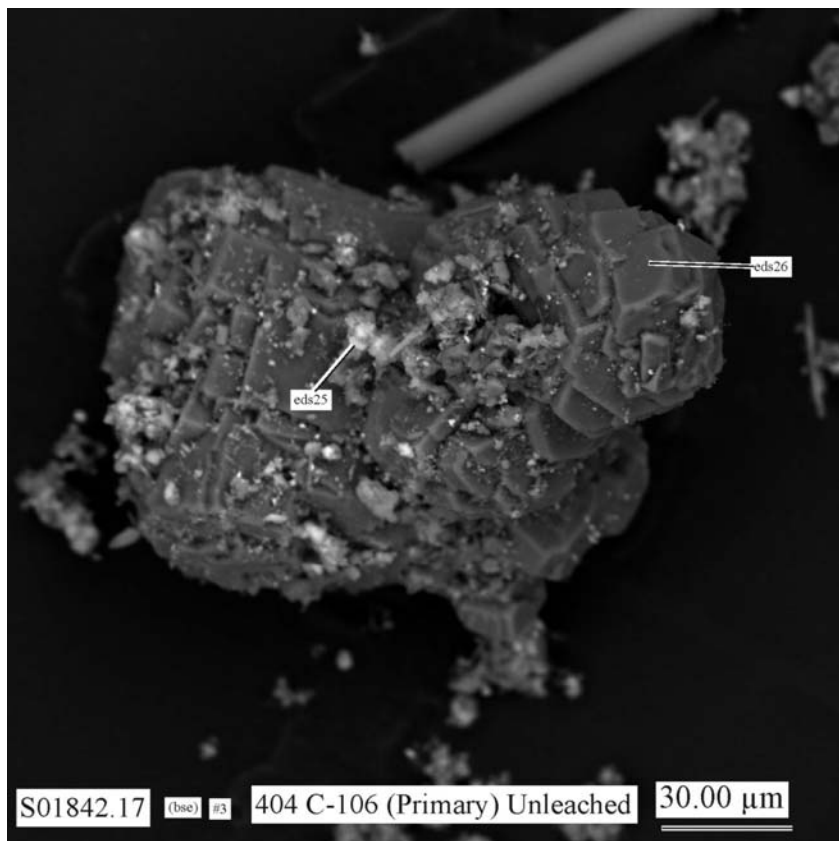
Figure B.32. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled C in Figure B.28
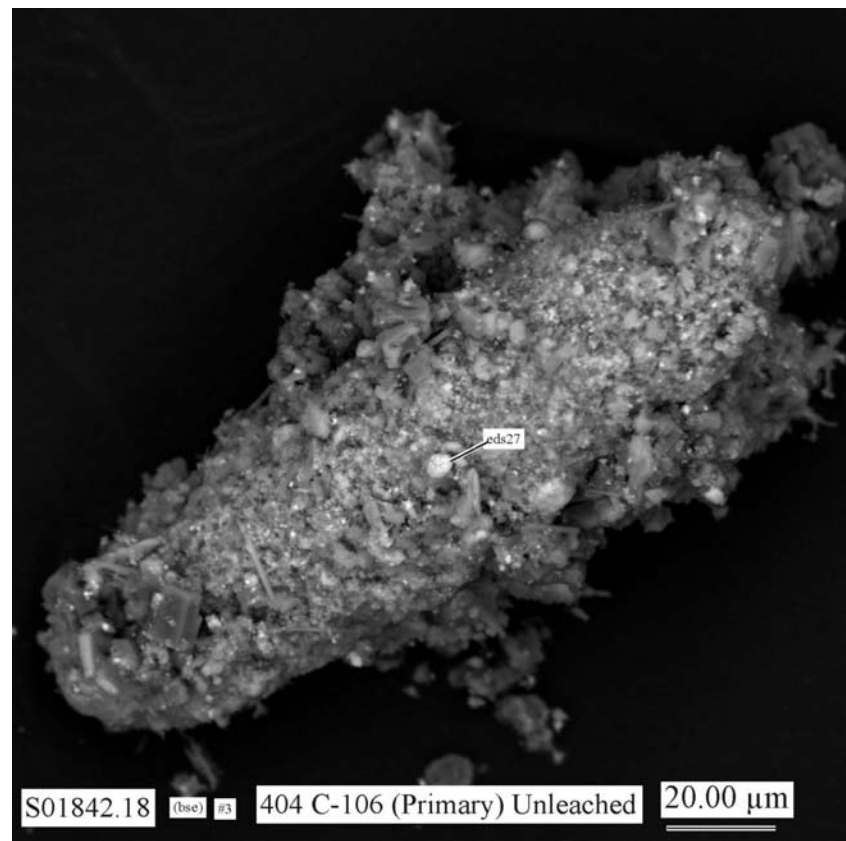
Figure B.33. Micrograph Showing Morphologies of Typical Particles in SEM Mount 3 of Unleached Sludge Sample 404
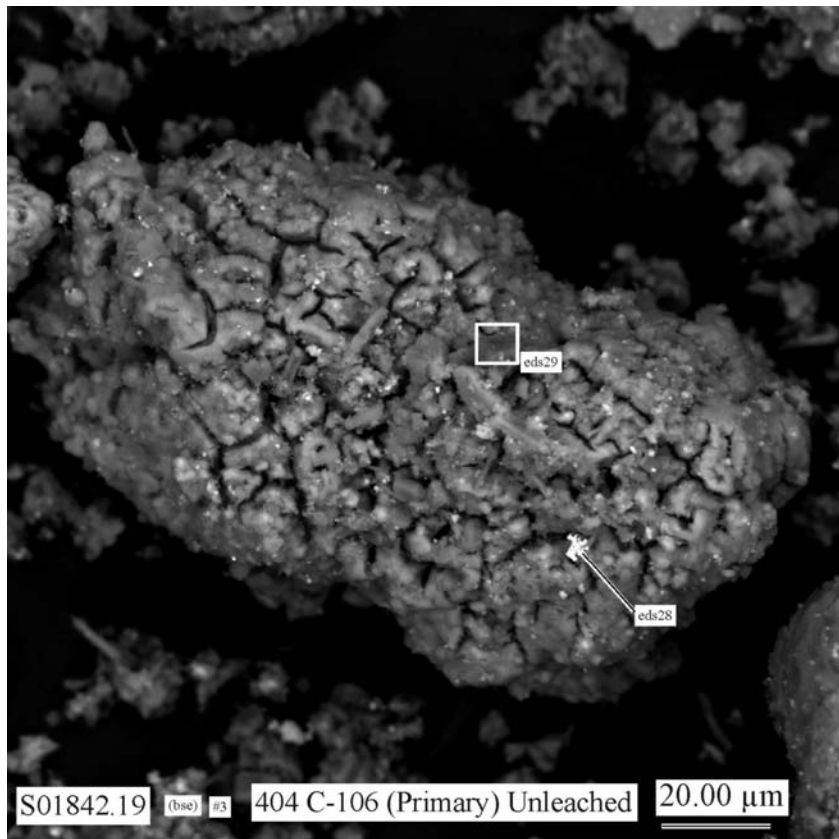
Figure B.34. Micrograph Showing Morphologies of Typical Particles in SEM Mount 3 of Unleached Sludge Sample 404

Figure B.35. Micrograph Showing Morphologies of Typical Particles in SEM Mount 3 of Unleached Ludge Sample 404

C-106 404 (#3) Primary Unleached eds20 [See Figure B.29]	C-106 404 (#3) Primary Unleached eds21 [See Figure B.29]
C-106 404 (#3) Primary Unleached eds22 [See Figure B.29]	C-106 404 (#3) Primary Unleached eds23 [See Figure B.30]

Figure B.36.   EDS Spectra for Analyses eds20 Through eds23 for Particles in SEM Mount 3 of Unleached Sludge Sample 404

C-106 404 (#3) Primary Unleached eds24 [See Figure B.30]	C-106 404 (#3) Primary Unleached eds25 [See Figure B.32]
C-106 404 (#3) Primary Unleached eds26 [See Figure B.32]	C-106 404 (#3) Primary Unleached eds27 [See Figure B.33]

Figure B.37. EDS Spectra for Analyses eds24 Through eds27 for Particles in SEM Mount 3 of Unleached Sludge Sample 404

C-106 404 (#3) Primary Unleached
eds28 [See Figure B.34]

C-106 404 (#3) Primary Unleached
eds29 [See Figure B.34]

C-106 404 (#3) Primary Unleached
eds30 [See Figure B.35]

Figure B.38.　　EDS Spectra for Analyses eds28 Through eds30 for Particles in SEM
Mount 3 of Unleached Sludge Sample 404

B.2 Unleached Sludge Sample 405

   Two mounts of unleached (raw) residual sludge from tank C-106 (duplicate 405) were analyzed by SEM/EDS. The SEM micrographs for the first mount (4) of sample 405 sludge are shown in Figures B.39 through B.52. The EDS spectra for this mount are given in Figures B.53 through B.57. The SEM micrographs for the second mount (6) of sample 405 sludge are shown in Figures B.58 through B.67, and the EDS spectra for the second mount are given in Figures B.68 through B.70.
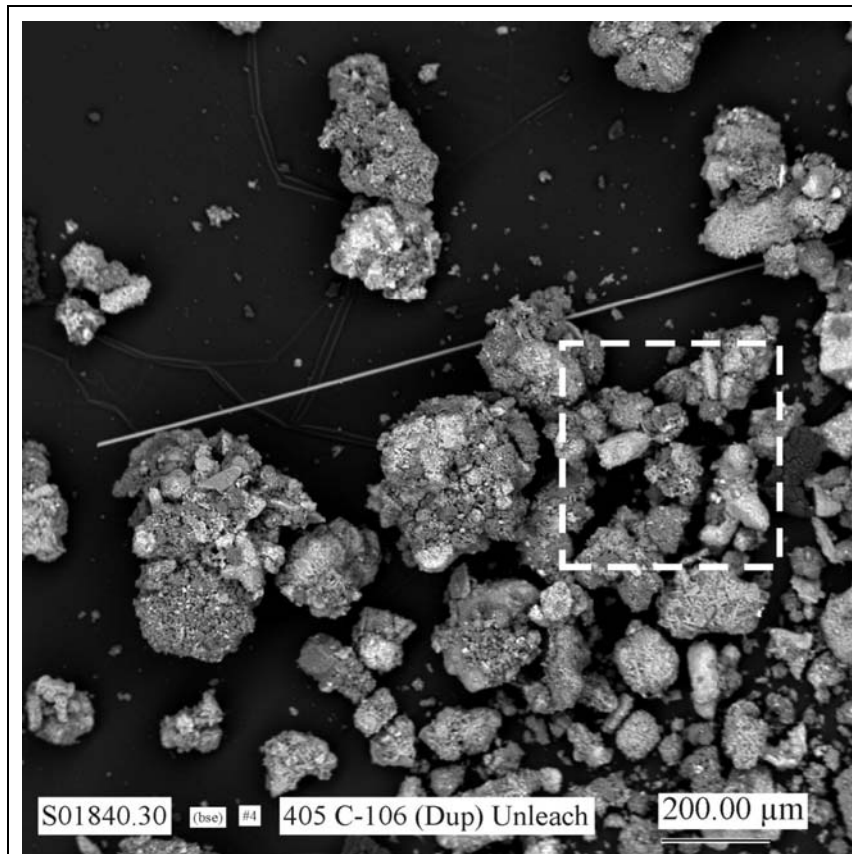
Figure B.39. Low Magnification SEM Micrograph Showing General Morphologies of Particles in SEM Mount 4 of Unleached Bulk Sludge Sample 405
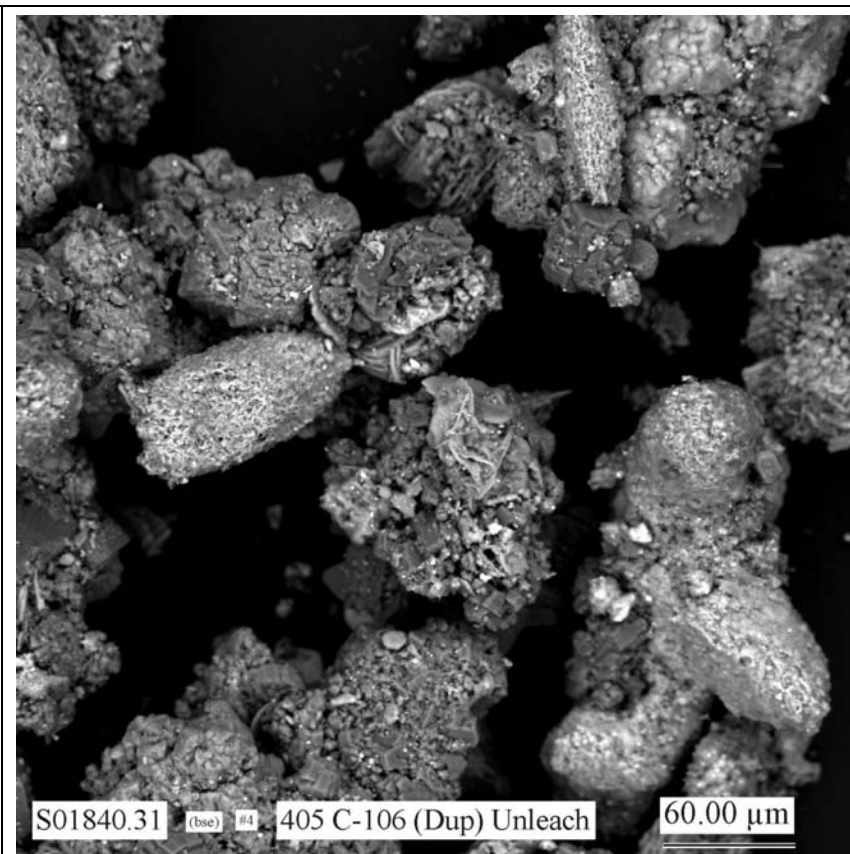
Figure B.40. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square in Figure B.39
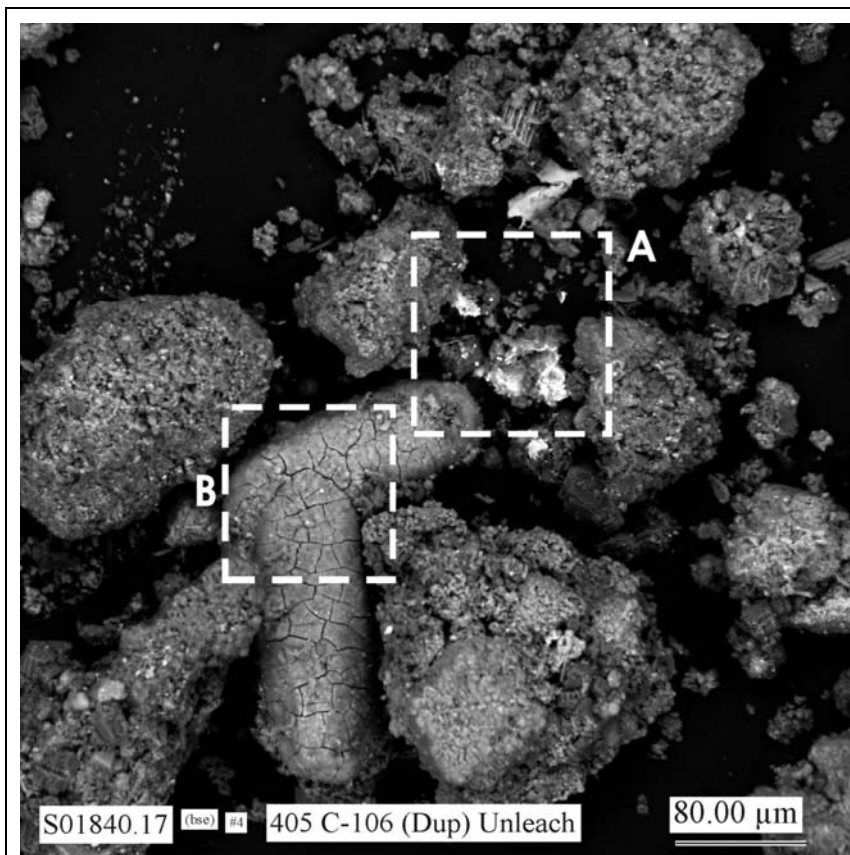
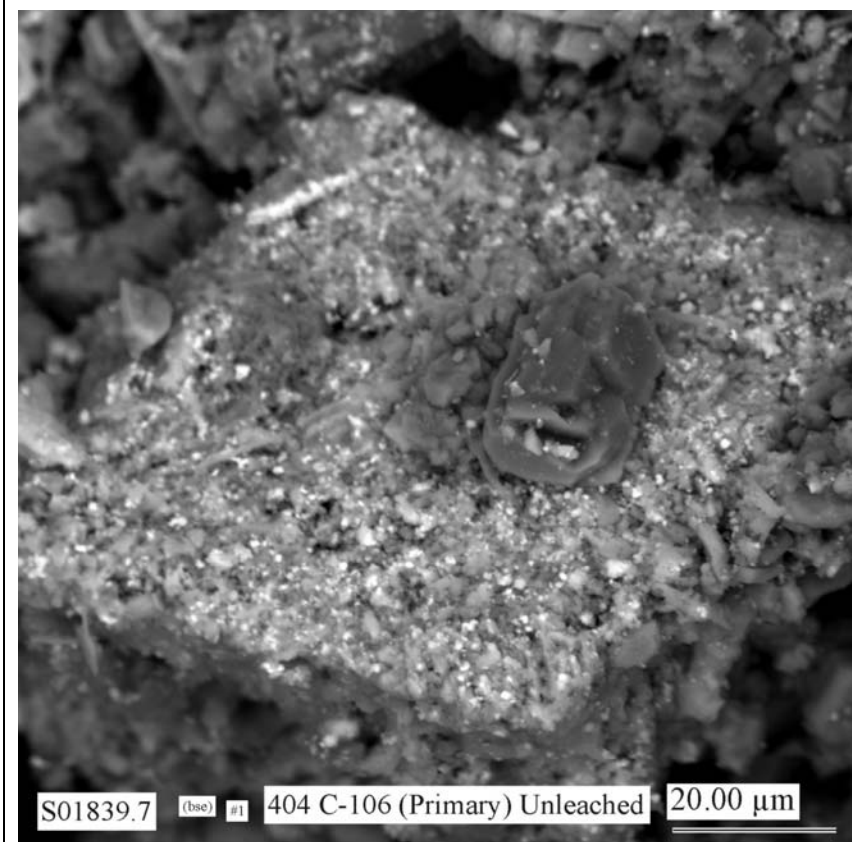**Figure B.41.** Micrograph Showing Morphologies of Typical Particles in SEM Mount 4 of Unleached Sludge Sample 405
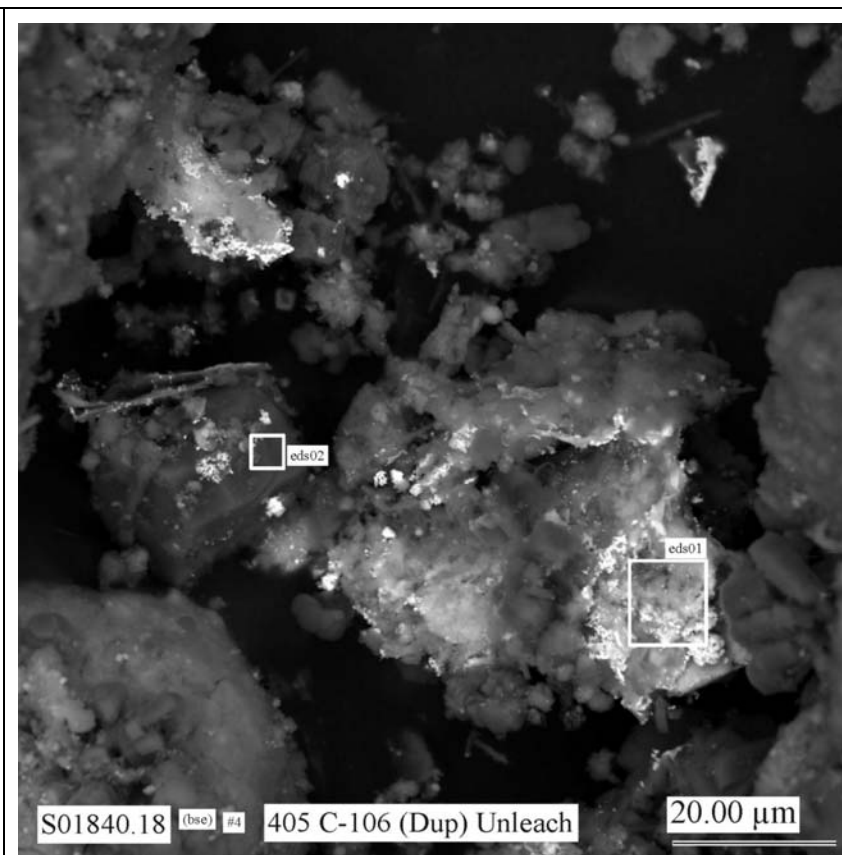
Figure B.42. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled A in Figure B.41
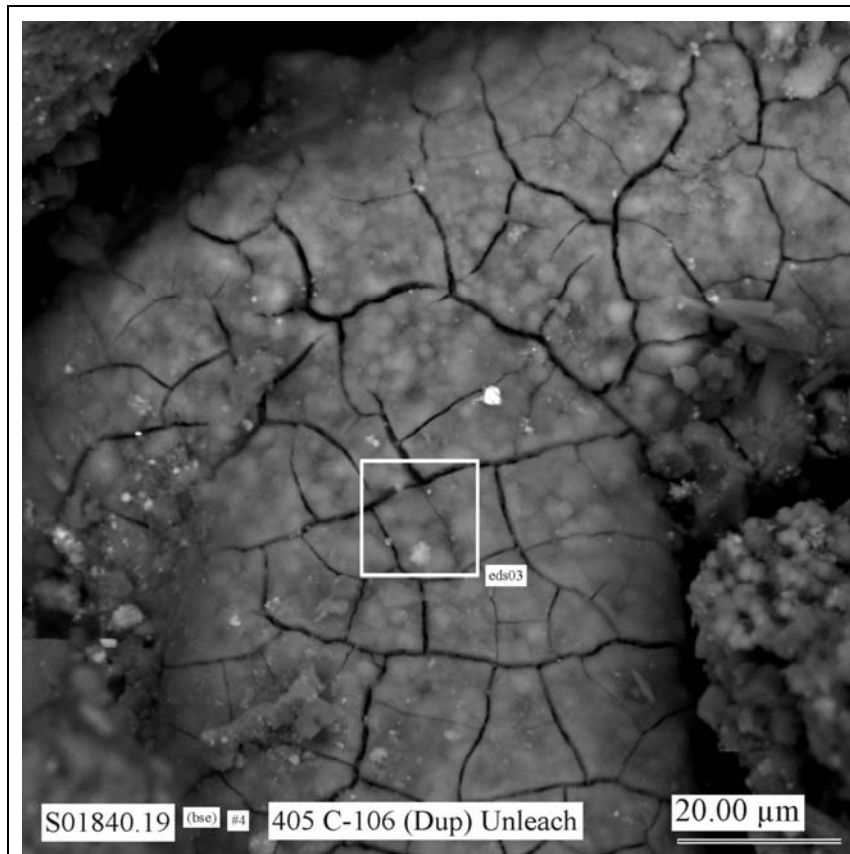
Figure B.43. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled B in Figure B.41
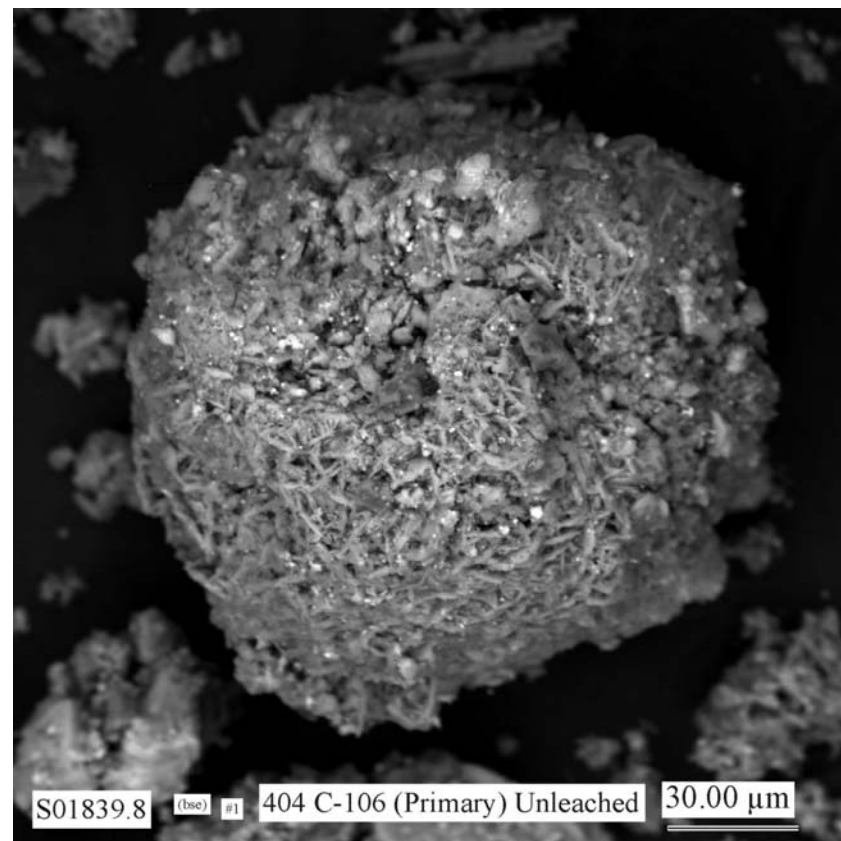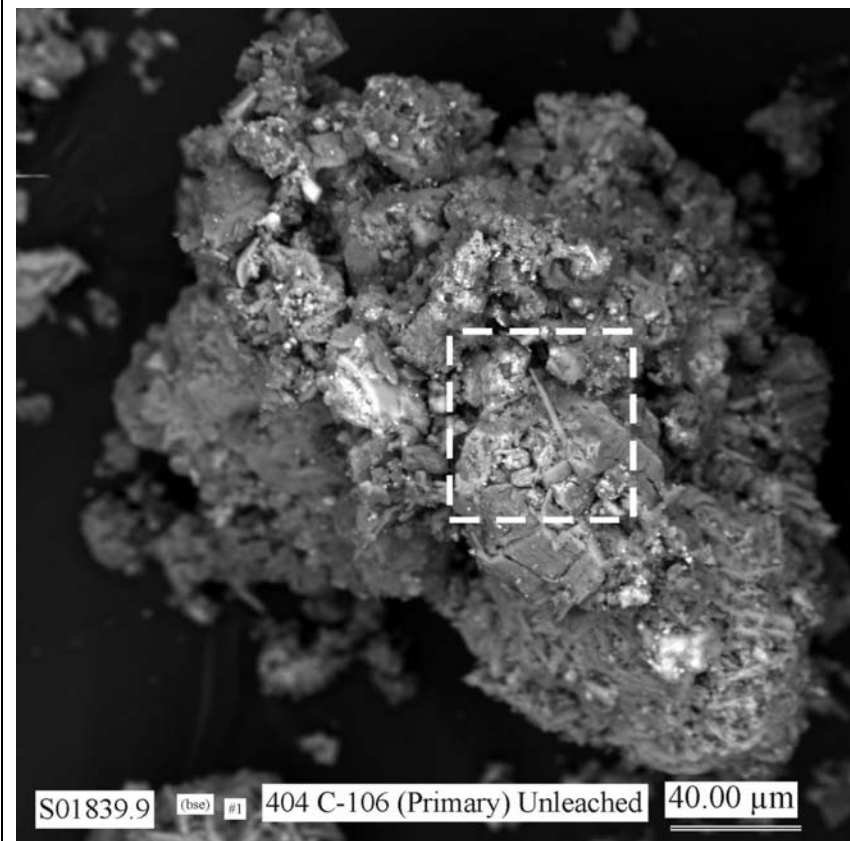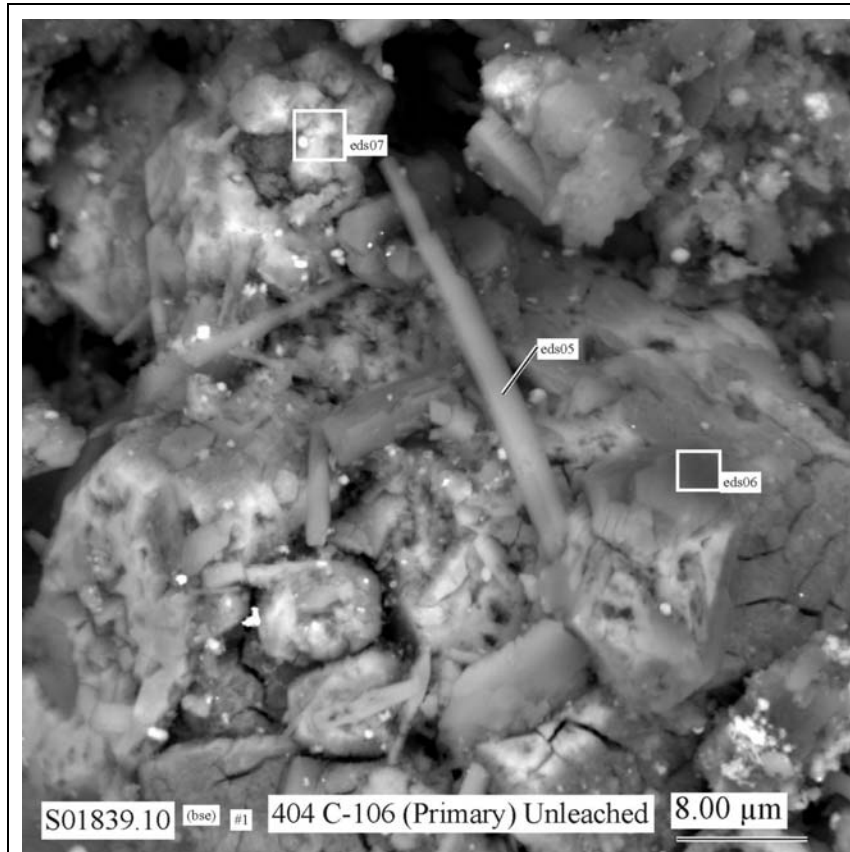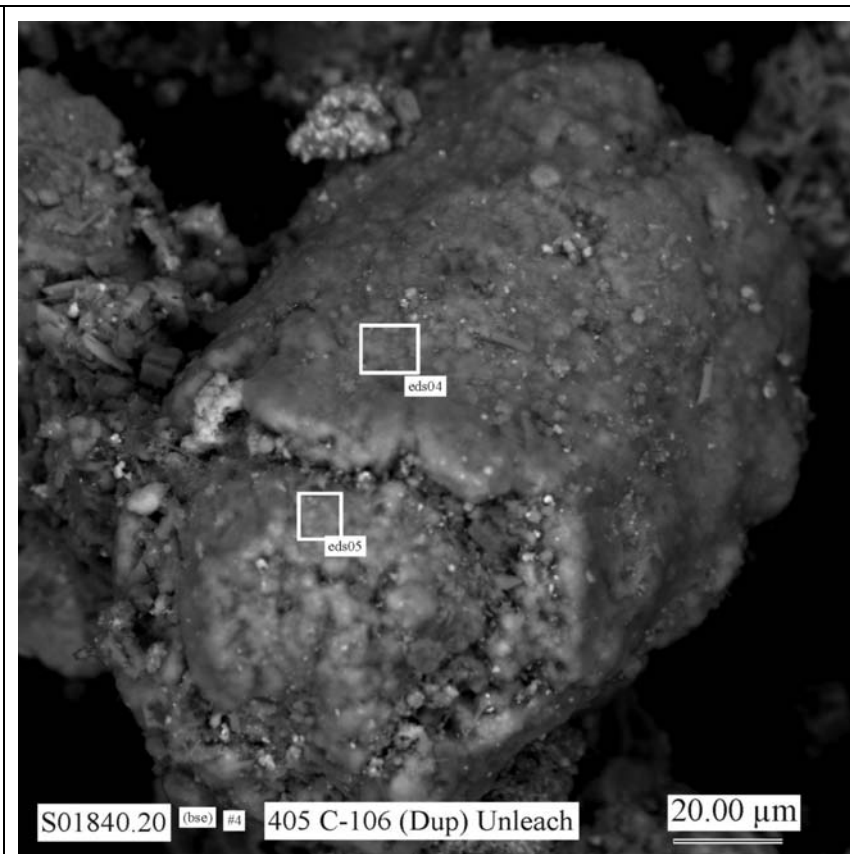
Figure B.44. Micrograph Showing Morphologies of Typical Particles in SEM Mount 4 of Unleached Sludge Sample 405
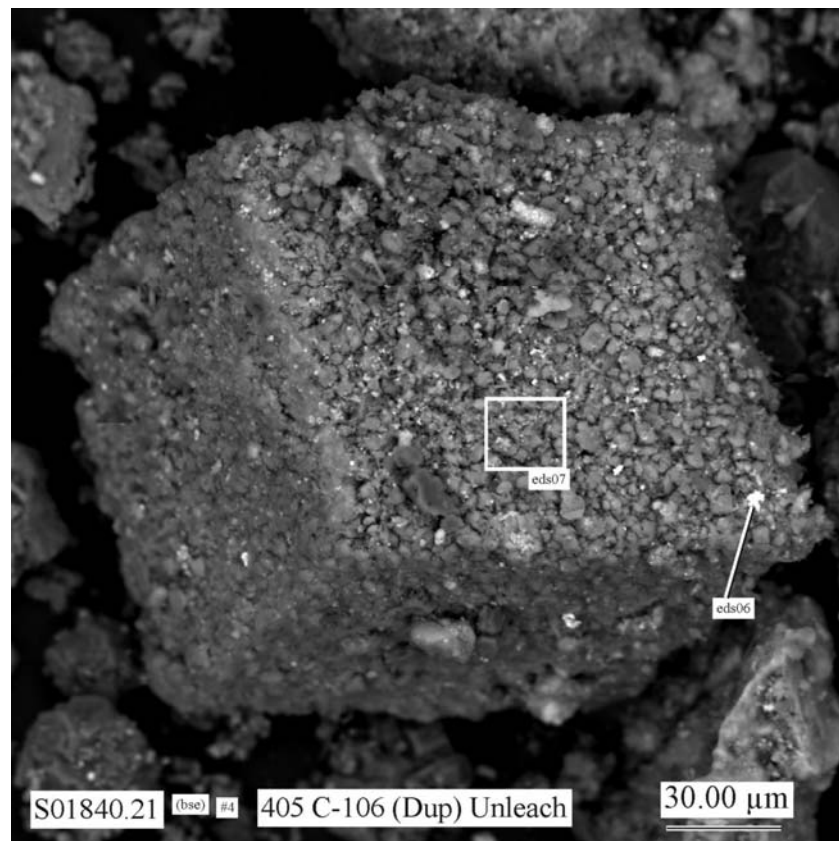
Figure B.45. Micrograph Showing Morphologies of Typical Particles in SEM Mount 4 of Unleached Sludge Sample 405
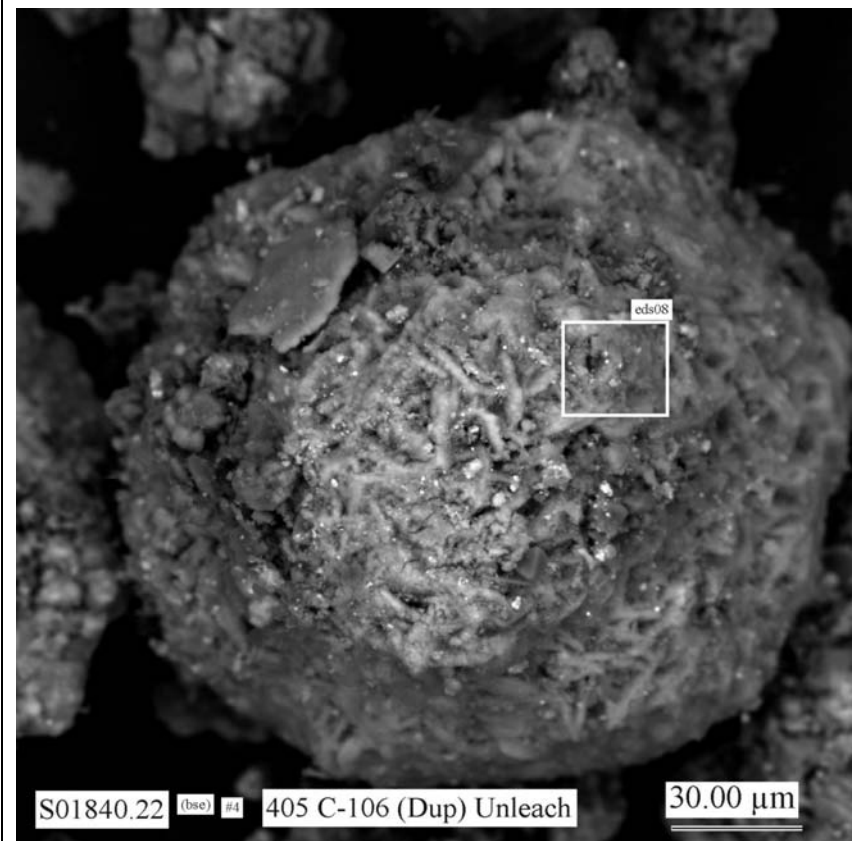
Figure B.46. Micrograph Showing Morphologies of Typical Particles in SEM Mount 4 of Unleached Sludge Sample 405
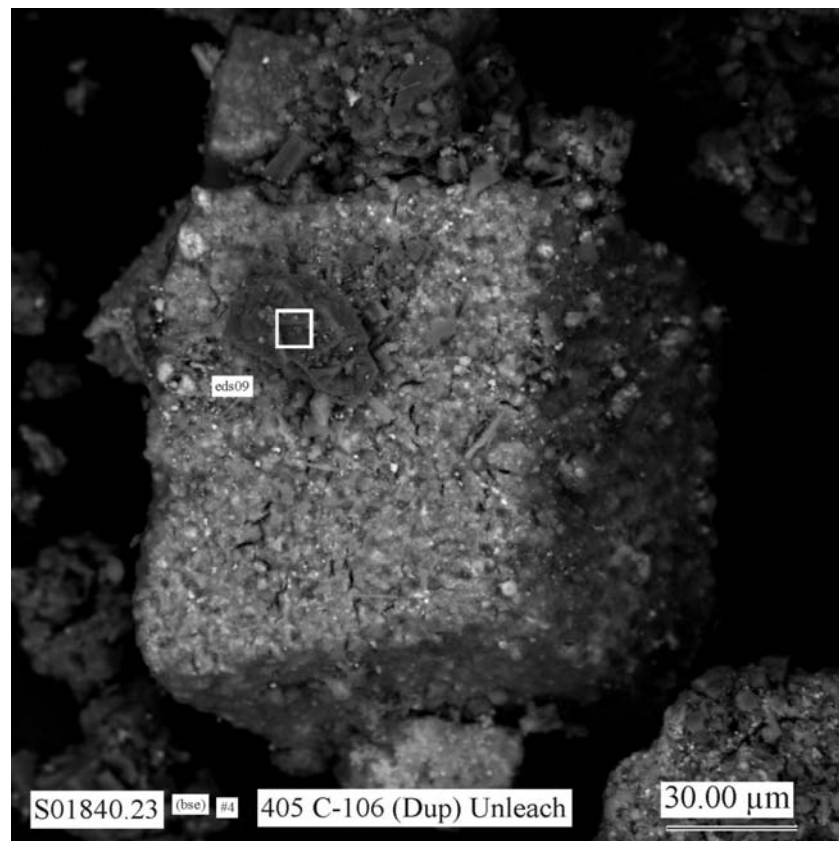
Figure B.47. Micrograph Showing Morphologies of Typical Particles in SEM Mount 4 of Unleached Sludge Sample 405
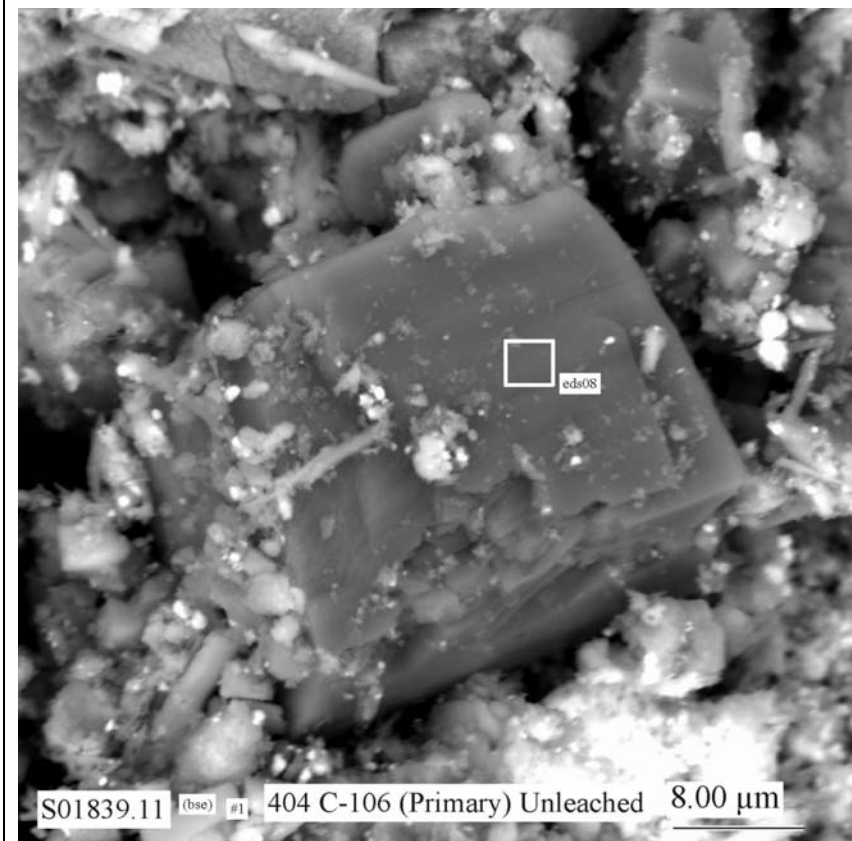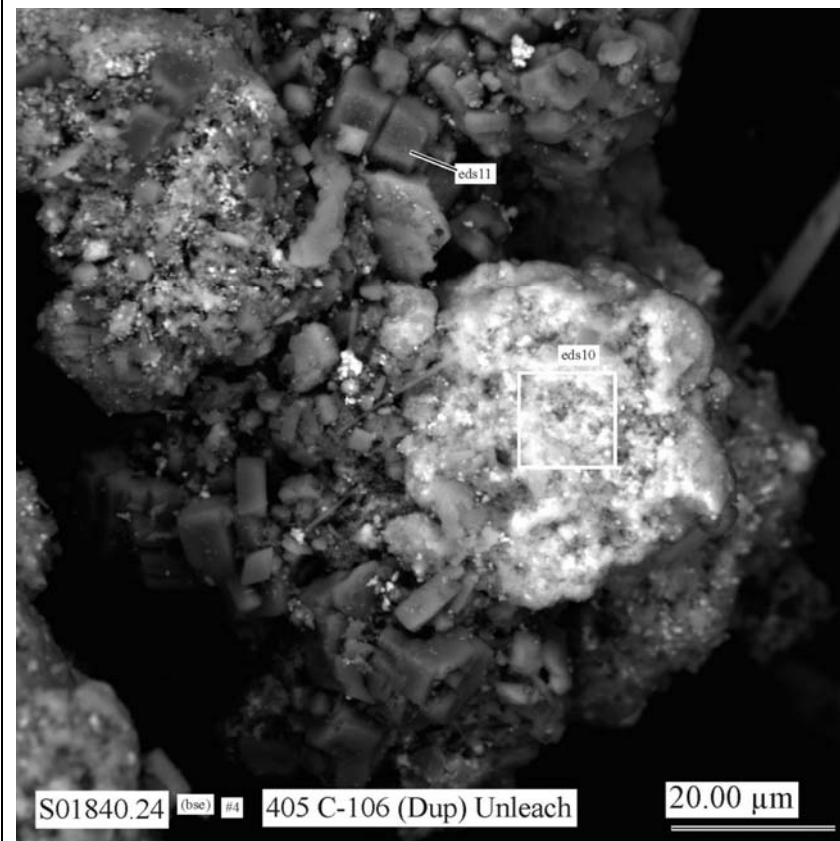
Figure B.48. Micrograph Showing Morphologies of Typical Particles in SEM Mount 4 of Unleached Sludge Sample 405
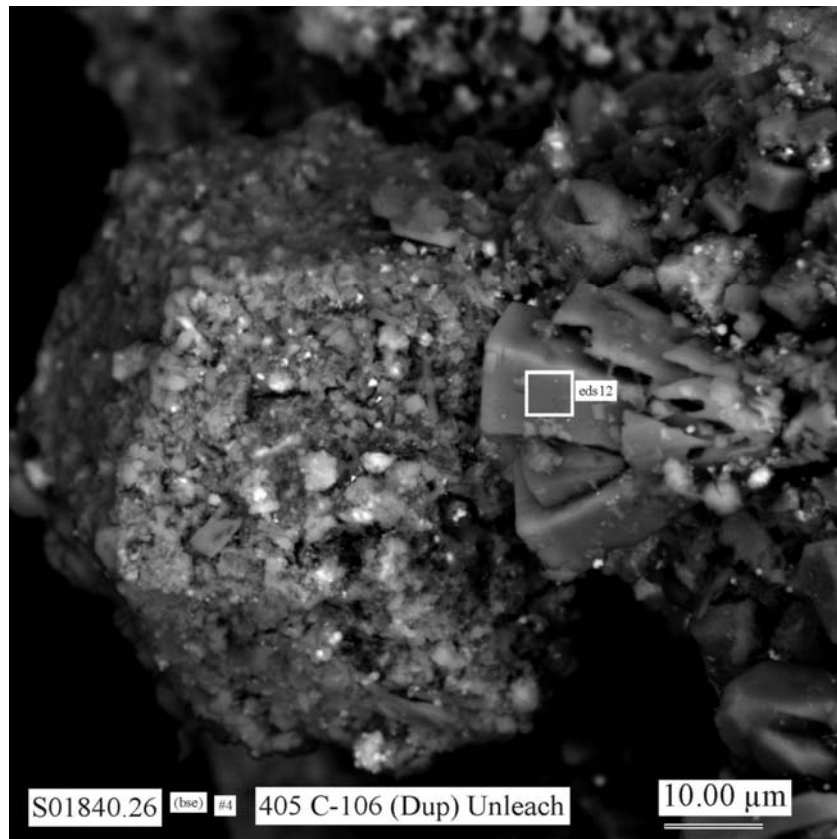
Figure B.49. Micrograph Showing Morphologies of Typical Particles in SEM Mount 4 of Unleached Sludge Sample 405
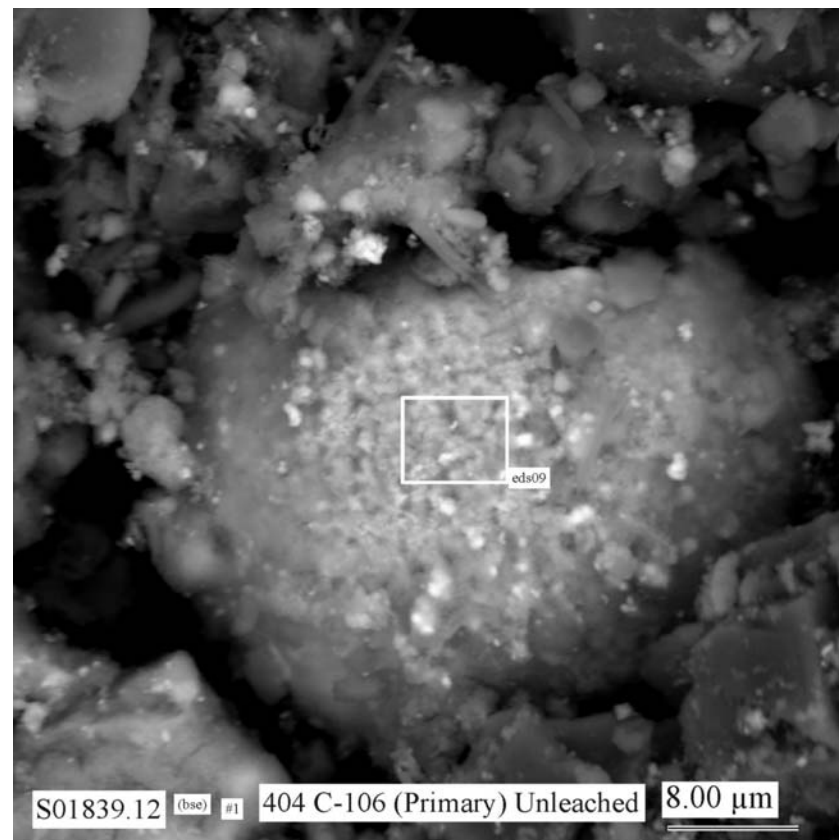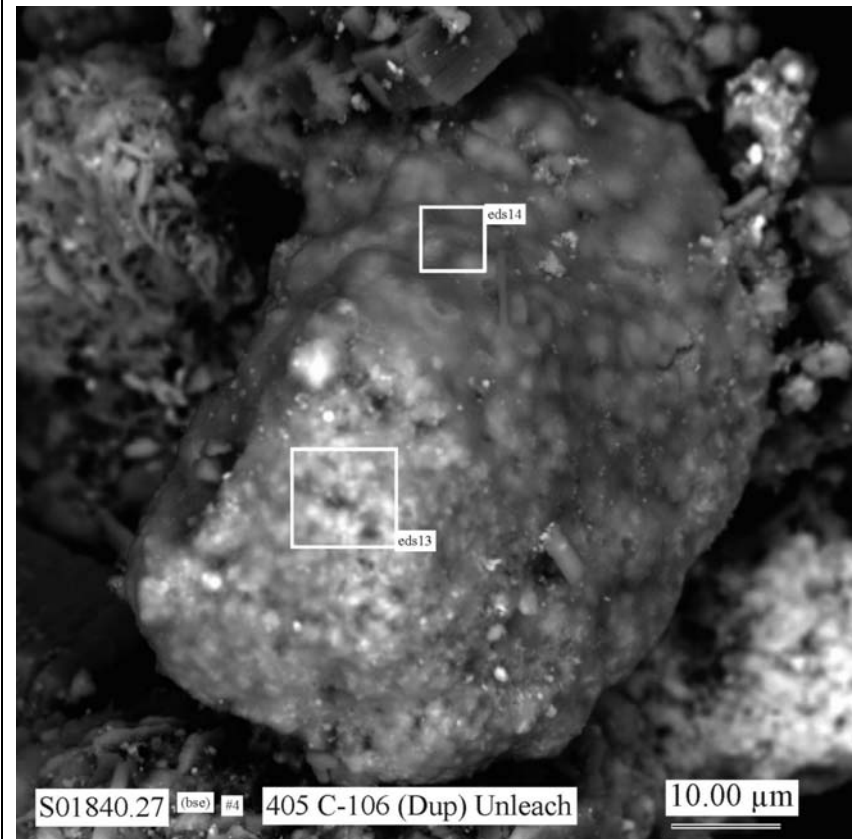
Figure B.50. Micrograph Showing Morphologies of Typical Particles in SEM Mount 4 of Unleached Sludge Sample 405
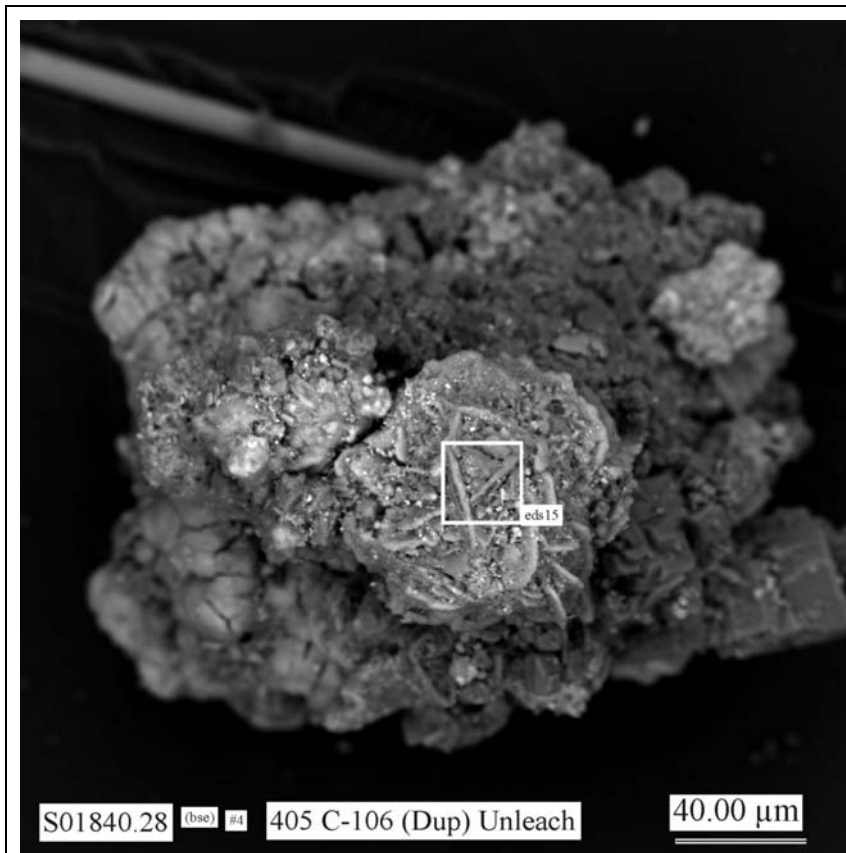
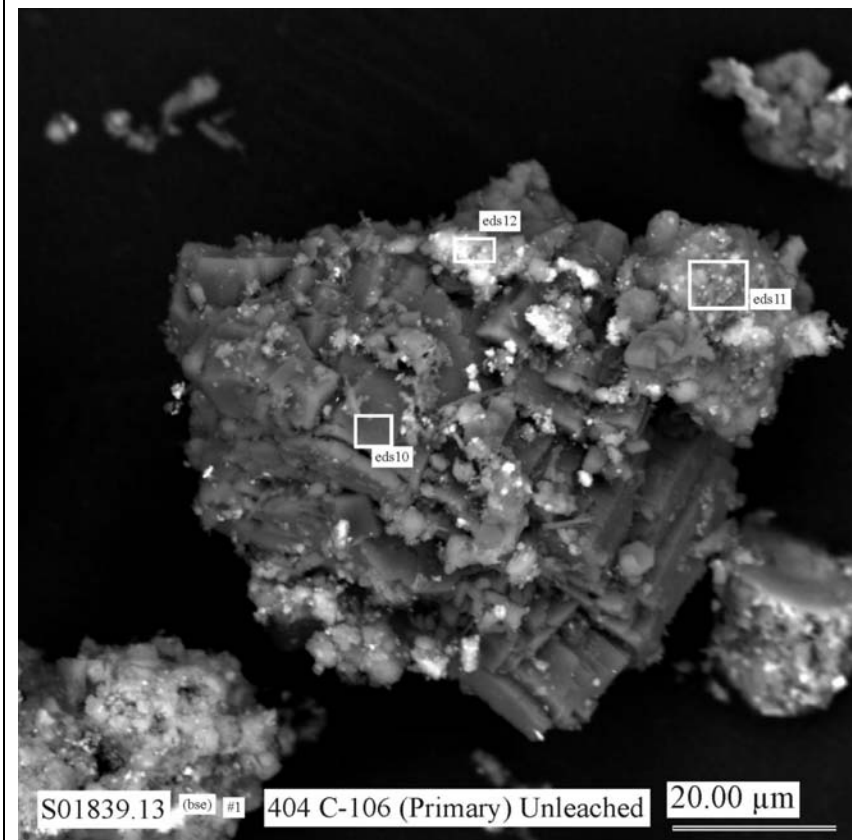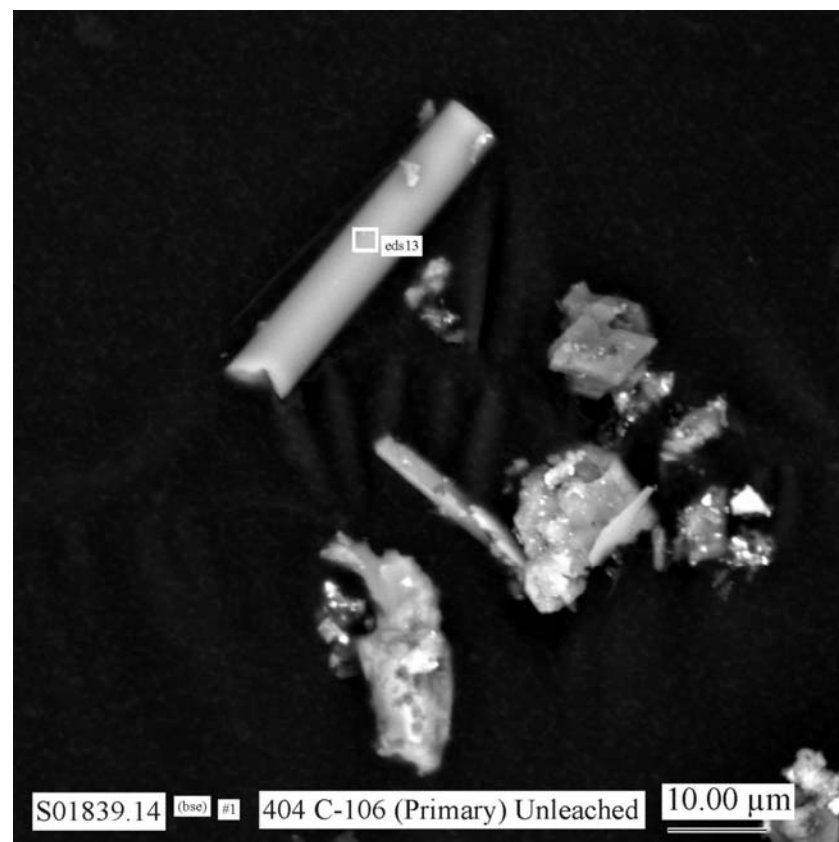| **Figure B.51.** | Micrograph Showing Morphologies of Typical Particles in SEM Mount 4 of Unleached Sludge Sample 405 | **Figure B.52.** | Micrograph Showing Morphologies of Typical Particles in SEM Mount 4 of Unleached Sludge Sample 405 |
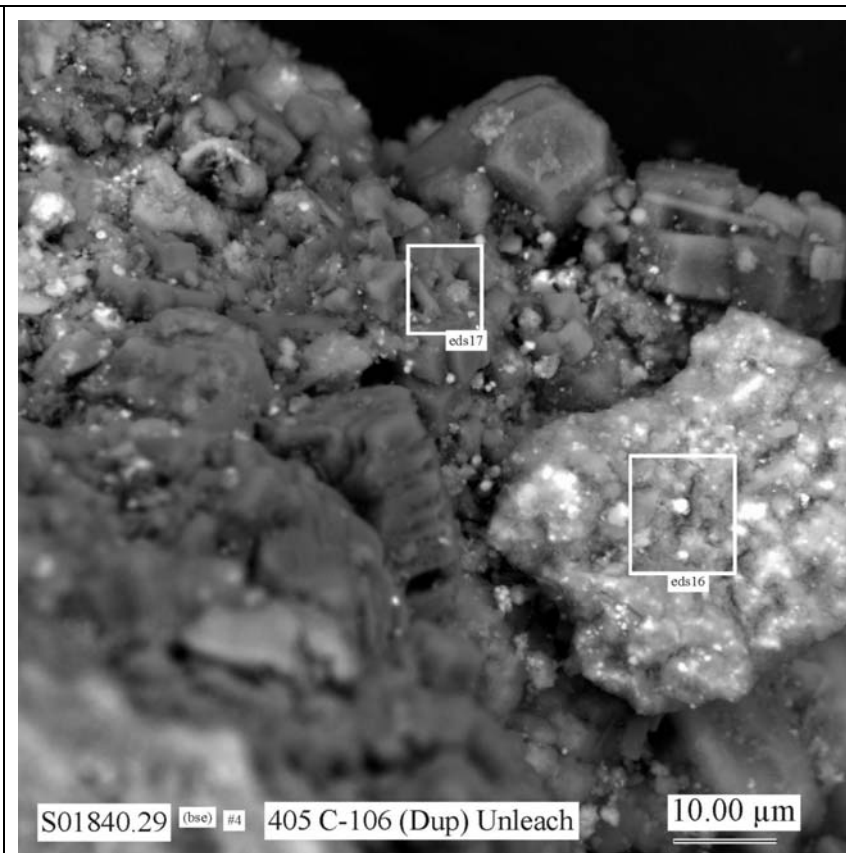
C-106 405 (#4) Duplicate Unleached
eds01 [See Figure B.42]

C-106 405 (#4) Duplicate Unleached
eds02 [See Figure B.42]

C-106 405 (#4) Duplicate Unleached
eds03 [See Figure B.43]

C-106 405 (#4) Duplicate Unleached
eds04 [See Figure B.44]

Figure B.53.   EDS Spectra for Analyses eds01 Through eds04 for Particles in SEM Mount 4 of
Unleached Sludge Sample 405

C-106 405 (#4) Duplicate Unleached
eds05 [See Figure B.44]

C-106 405 (#4) Duplicate Unleached
eds06 [See Figure B.45]

C-106 405 (#4) Duplicate Unleached
eds07 [See Figure B.45]

C-106 405 (#4) Duplicate Unleached
eds08 [See Figure B.46]

Figure B.54.   EDS Spectra for Analyses eds05 Through eds08 for Particles in SEM Mount 4 of
Unleached Sludge Sample 405

B.34

C-106 405 (#4) Duplicate Unleached eds09 [See Figure B.47]	C-106 405 (#4) Duplicate Unleached eds10 [See Figure B.48]
C-106 405 (#4) Duplicate Unleached eds11 [See Figure B.48]	C-106 405 (#4) Duplicate Unleached eds12 [See Figure B.49]

Figure B.55.   EDS Spectra for Analyses eds09 Through eds12 for Particles in SEM Mount 4 of Unleached Sludge Sample 405

C-106 405 (#4) Duplicate Unleached eds13 [See Figure B.50]	C-106 405 (#4) Duplicate Unleached eds14 [See Figure B.50]
C-106 405 (#4) Duplicate Unleached eds15 [See Figure B.51]	C-106 405 (#4) Duplicate Unleached eds16 [See Figure B.52]

Figure B.56. EDS Spectra for Analyses eds13 Through eds16 for Particles in SEM Mount 4 of Unleached Sludge Sample 405

C-106 405 (#4) Duplicate Unleached
eds17 [See Figure B.52]

Figure B.57.   EDS Spectra for Analysis
eds17 for a Particle in SEM
Mount 4 of Unleached
Sludge Sample 405
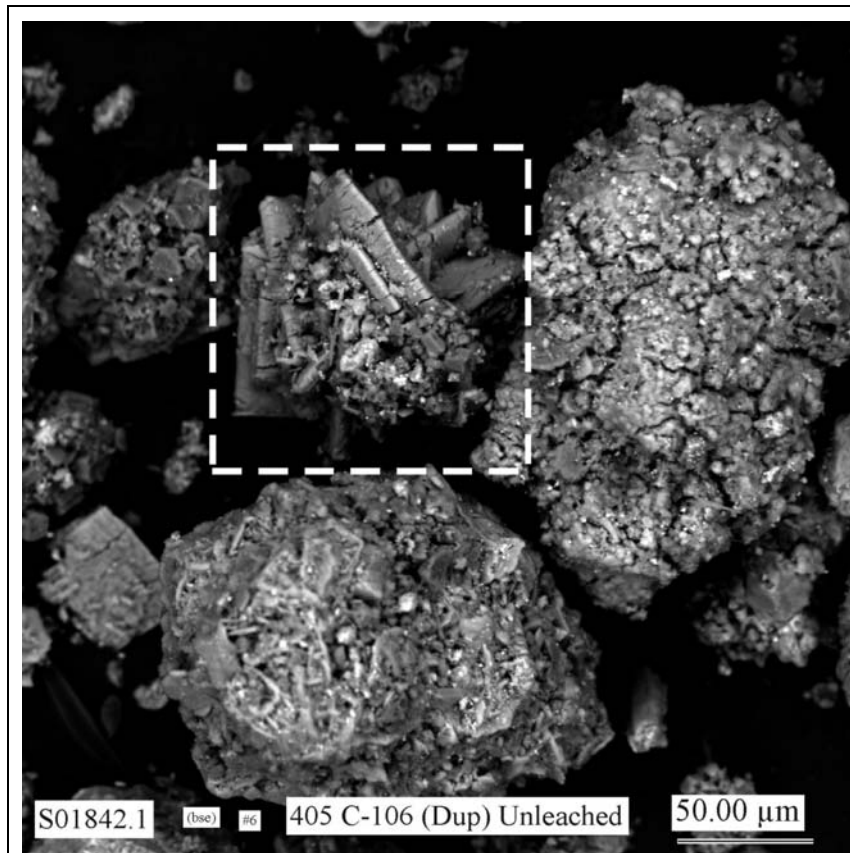
Figure B.58. Low Magnification SEM Micrograph Showing General Morphologies of Particles in SEM Mount 6 of Unleached Bulk Sludge Sample 405
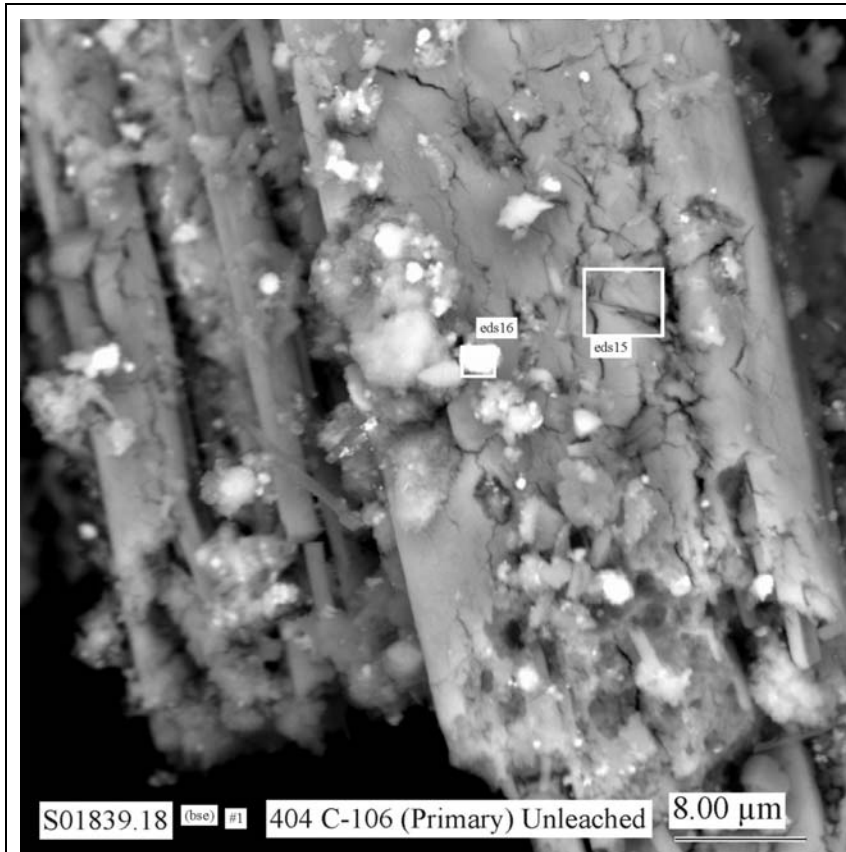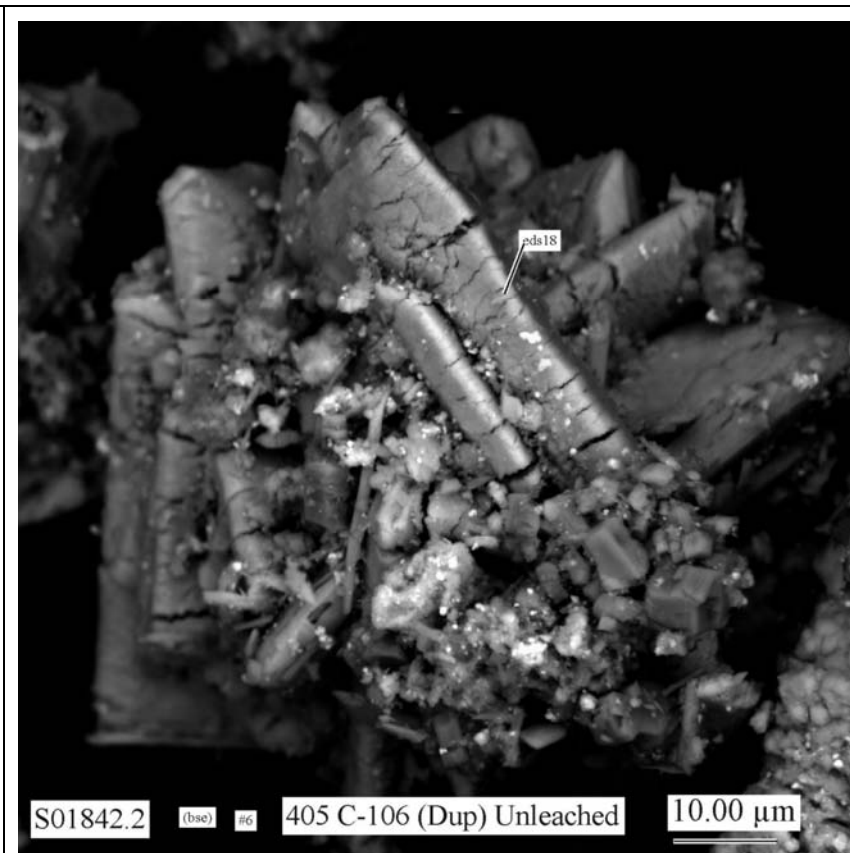
Figure B.59. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square in Figure B.58
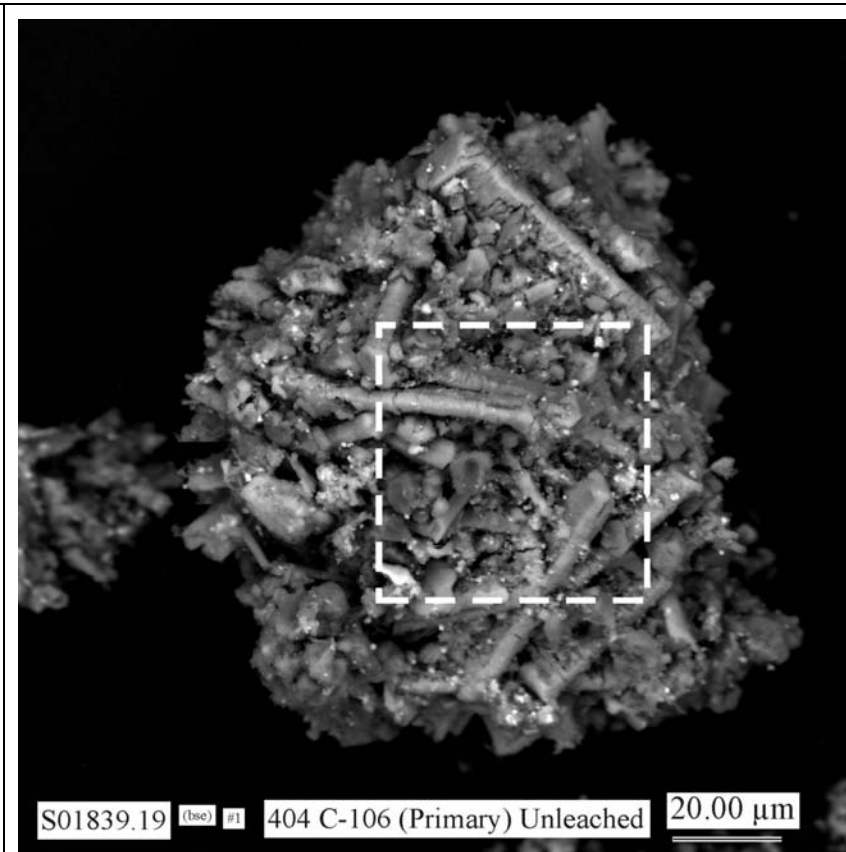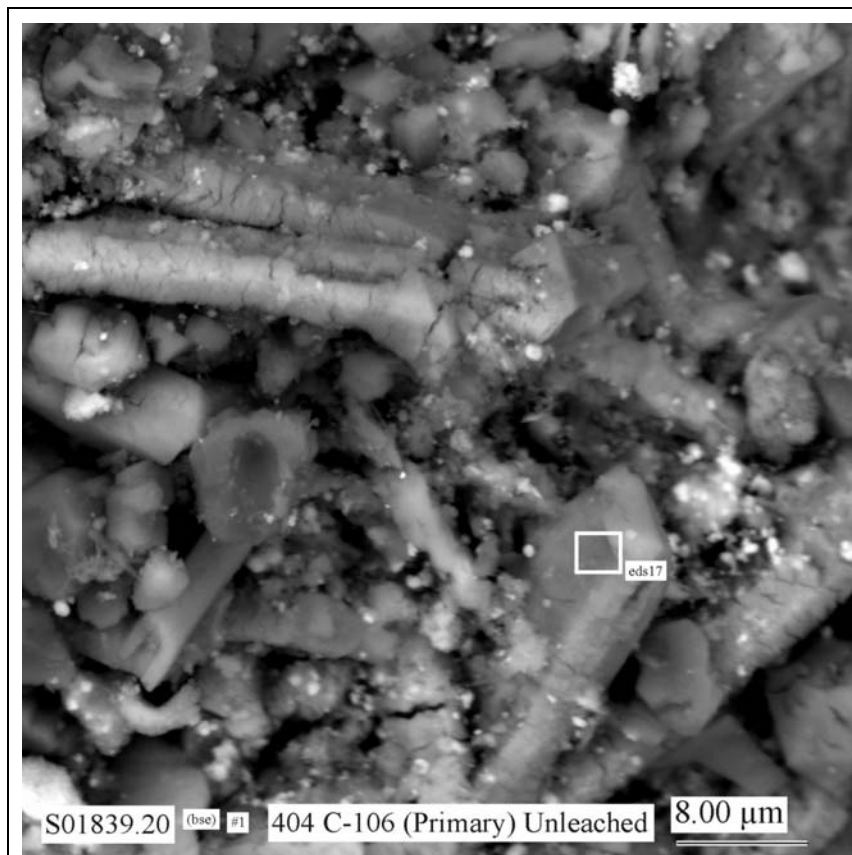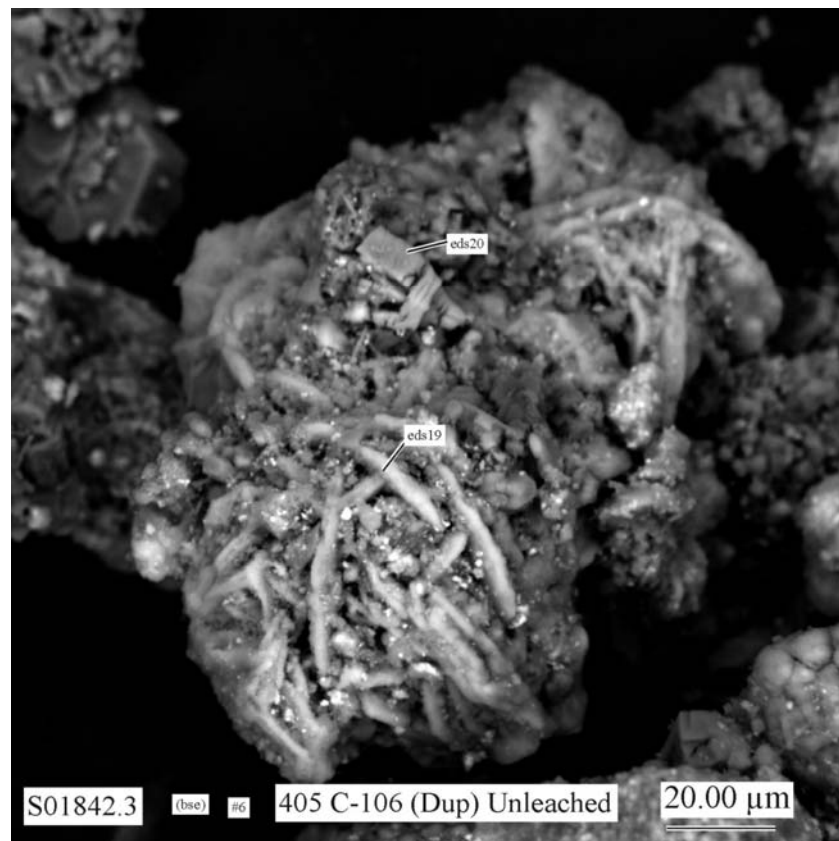
Figure B.60. Micrograph Showing Morphologies of Typical Particles in SEM Mount 6 of Unleached Sludge Sample 405
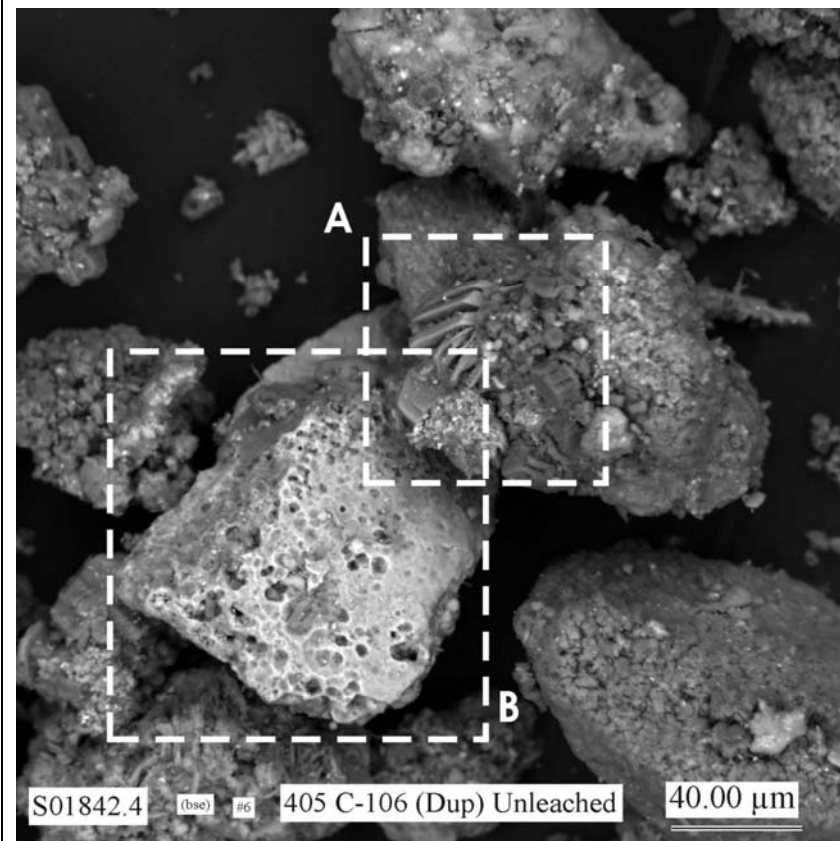
Figure B.61. Micrograph Showing Morphologies of Typical Particles in SEM Mount 6 of Unleached Sludge Sample 405

Figure B.62. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled A in Figure B.61

Figure B.63. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled b in Figure B.61
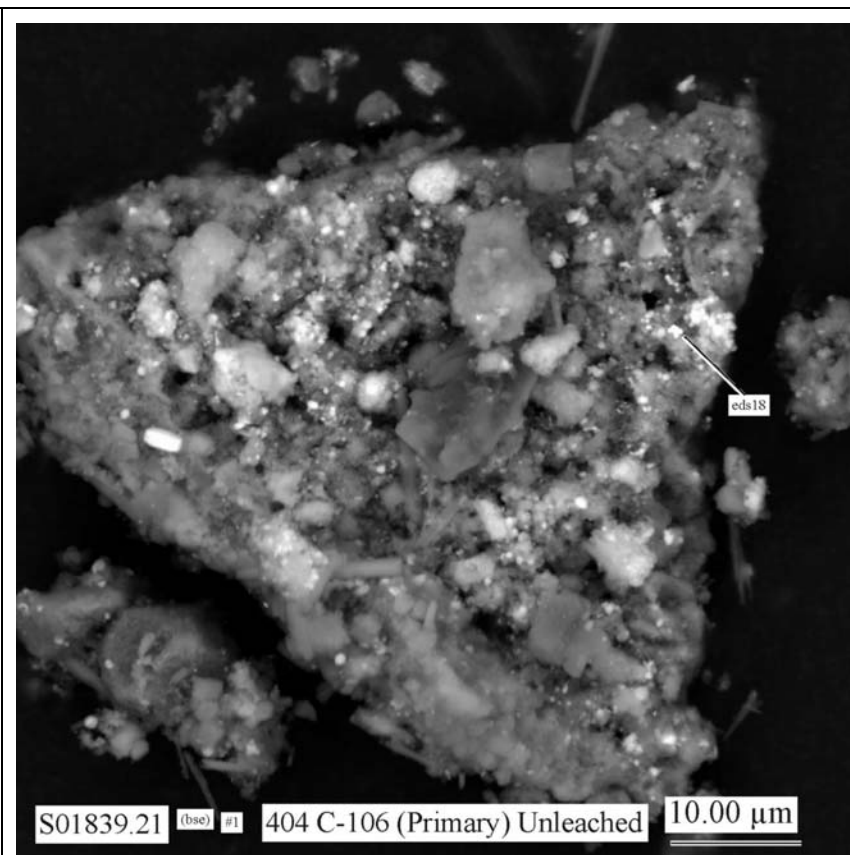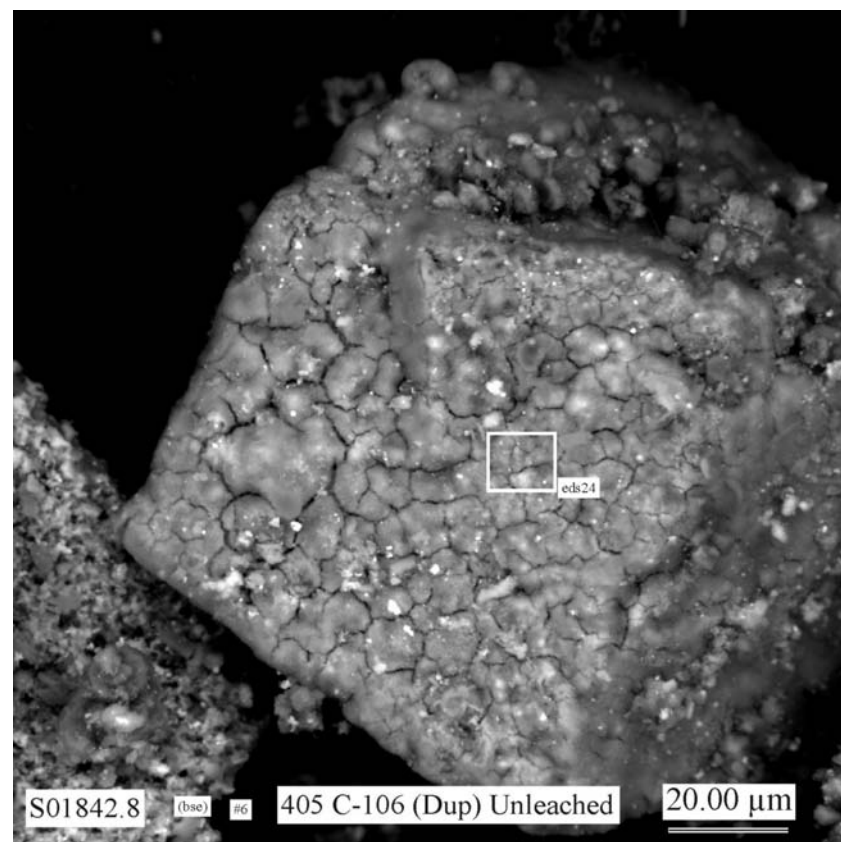
Figure B.64. Micrograph Showing Morphologies of Typical Particles in SEM Mount 6 of Unleached Sludge Sample 405
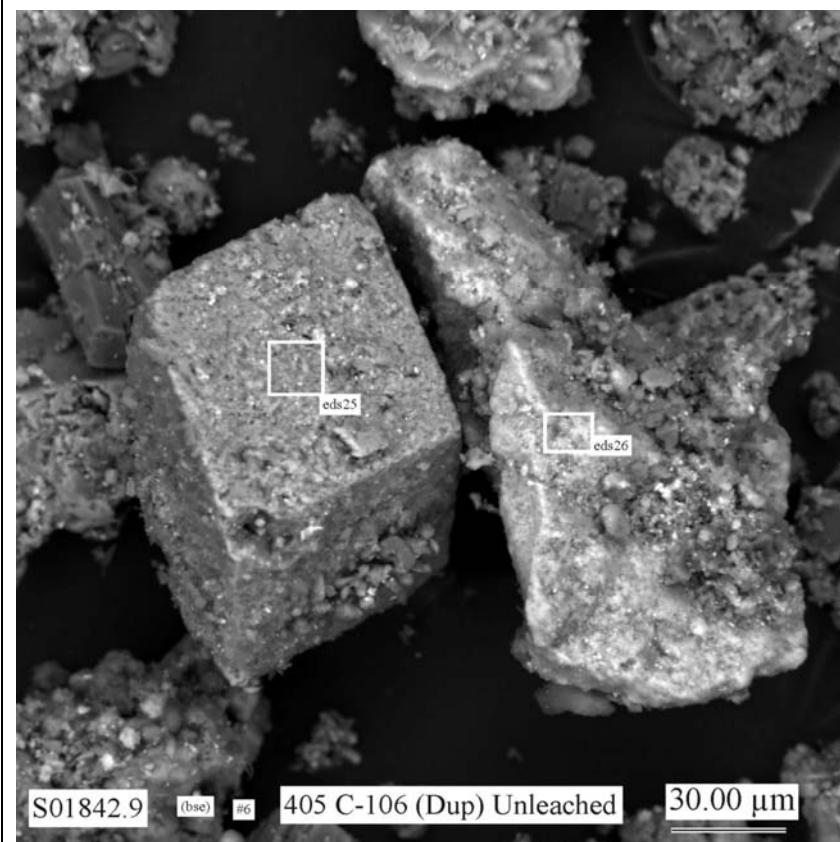
Figure B.65. Micrograph Showing Morphologies of Typical Particles in SEM Mount 6 of Unleached Sludge Sample 405
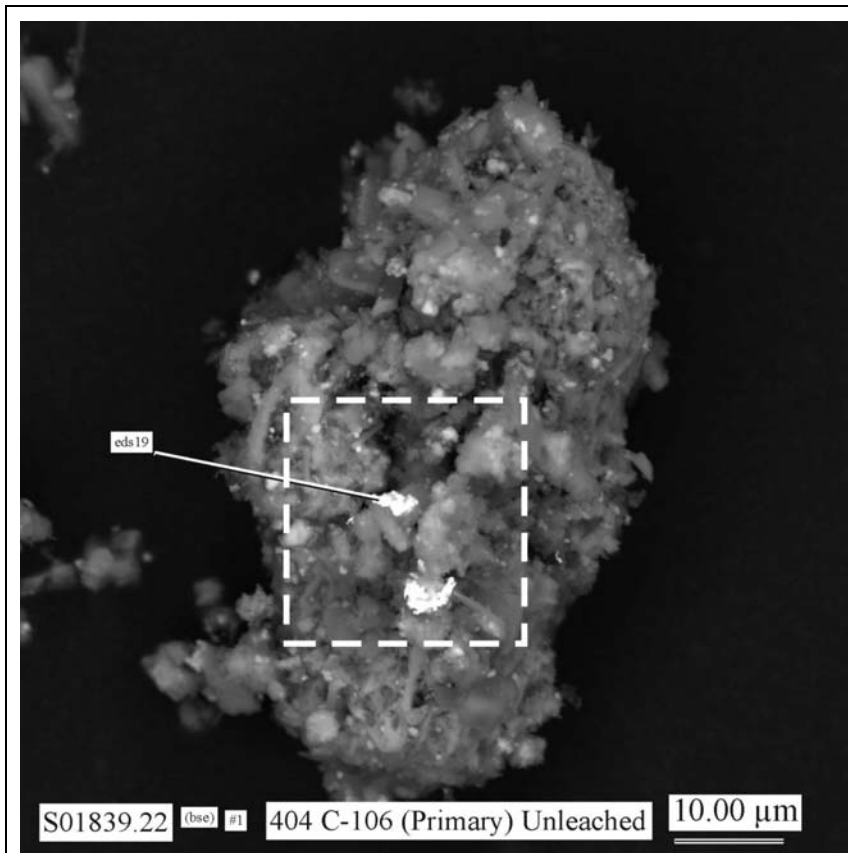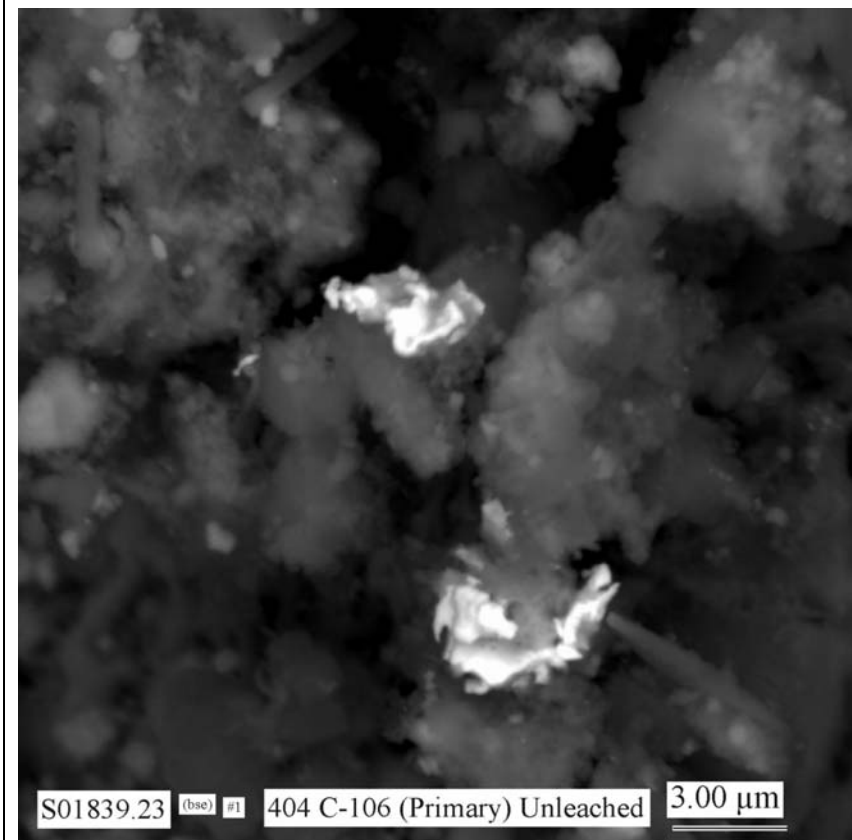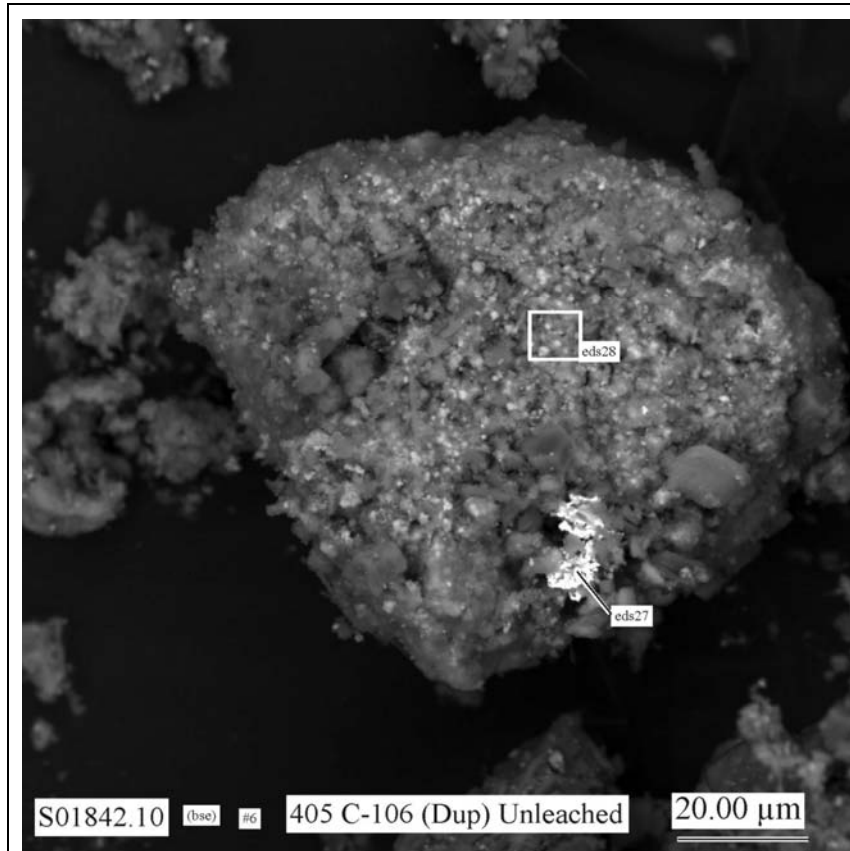
Figure B.66. Micrograph Showing Morphologies of Typical Particles in SEM Mount 6 of Unleached Sludge Sample 405
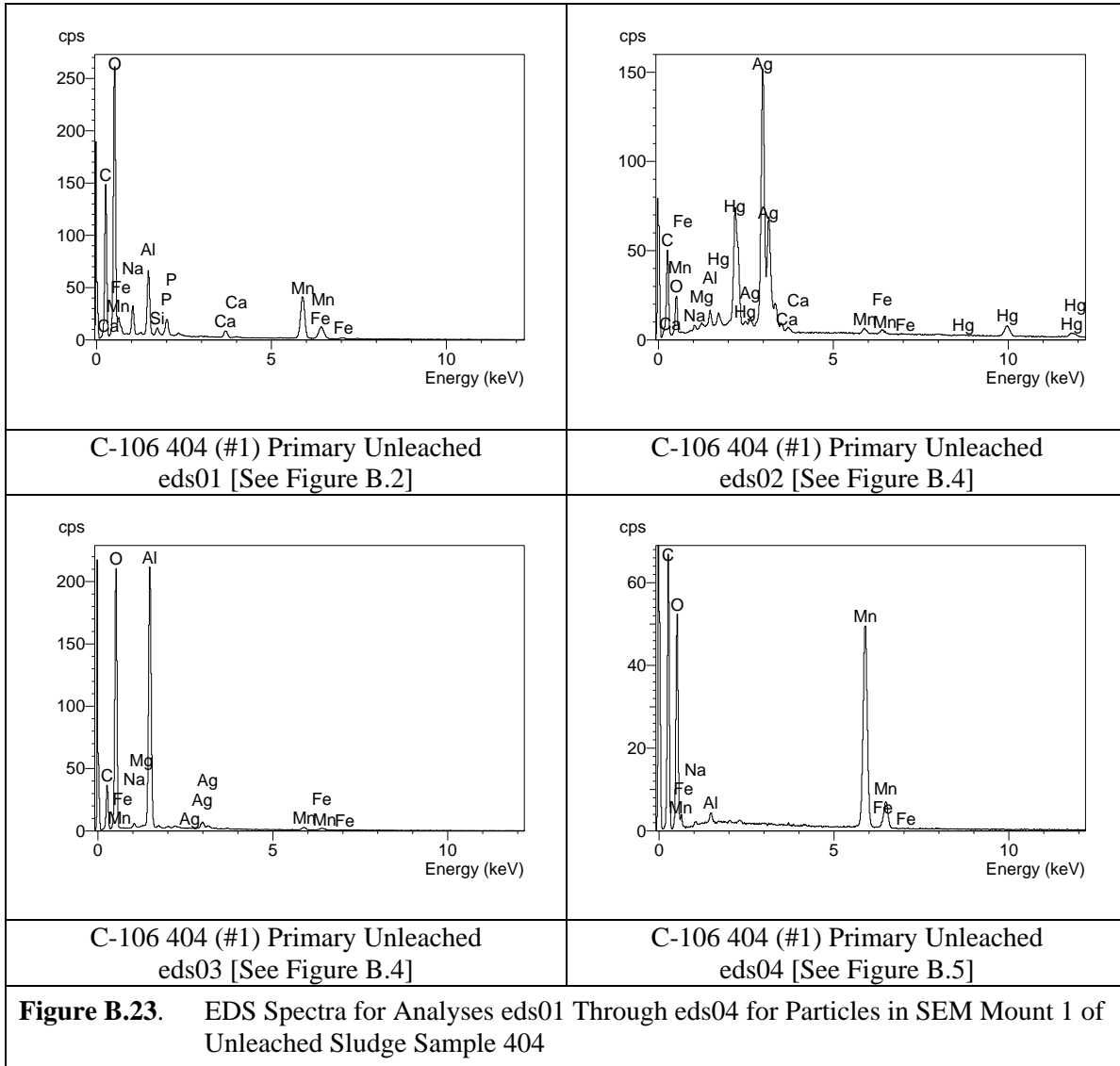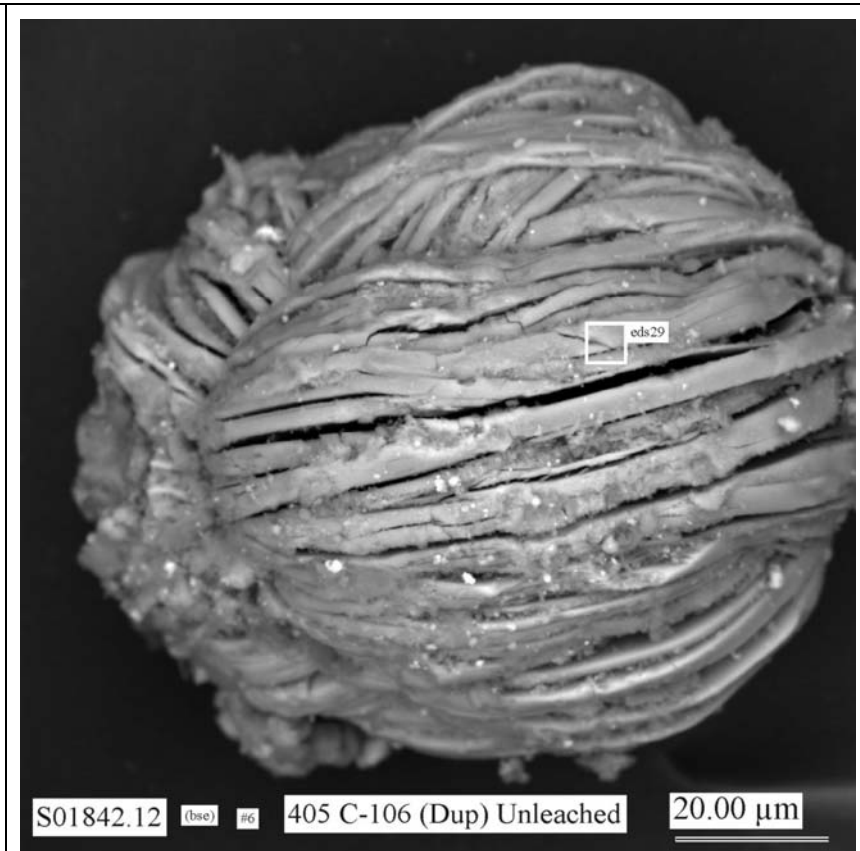
Figure B.67. Micrograph Showing Morphology of Unusual Particle (composed primarily of Mn, P, O, and possibly C) in SEM Mount 6 of Unleached Sludge Sample 405

C-106 405 (#6) Duplicate Unleached
eds18 [See Figure B.59]

C-106 405 (#6) Duplicate Unleached
eds19 [See Figure B.60]

C-106 405 (#6) Duplicate Unleached
eds20 [See Figure B.60]

C-106 405 (#6) Duplicate Unleached
eds21 [See Figure B.62]

Figure B.68.　　EDS Spectra for Analyses eds18 Through eds21 for Particles in SEM Mount 6 of Unleached Sludge Sample 405

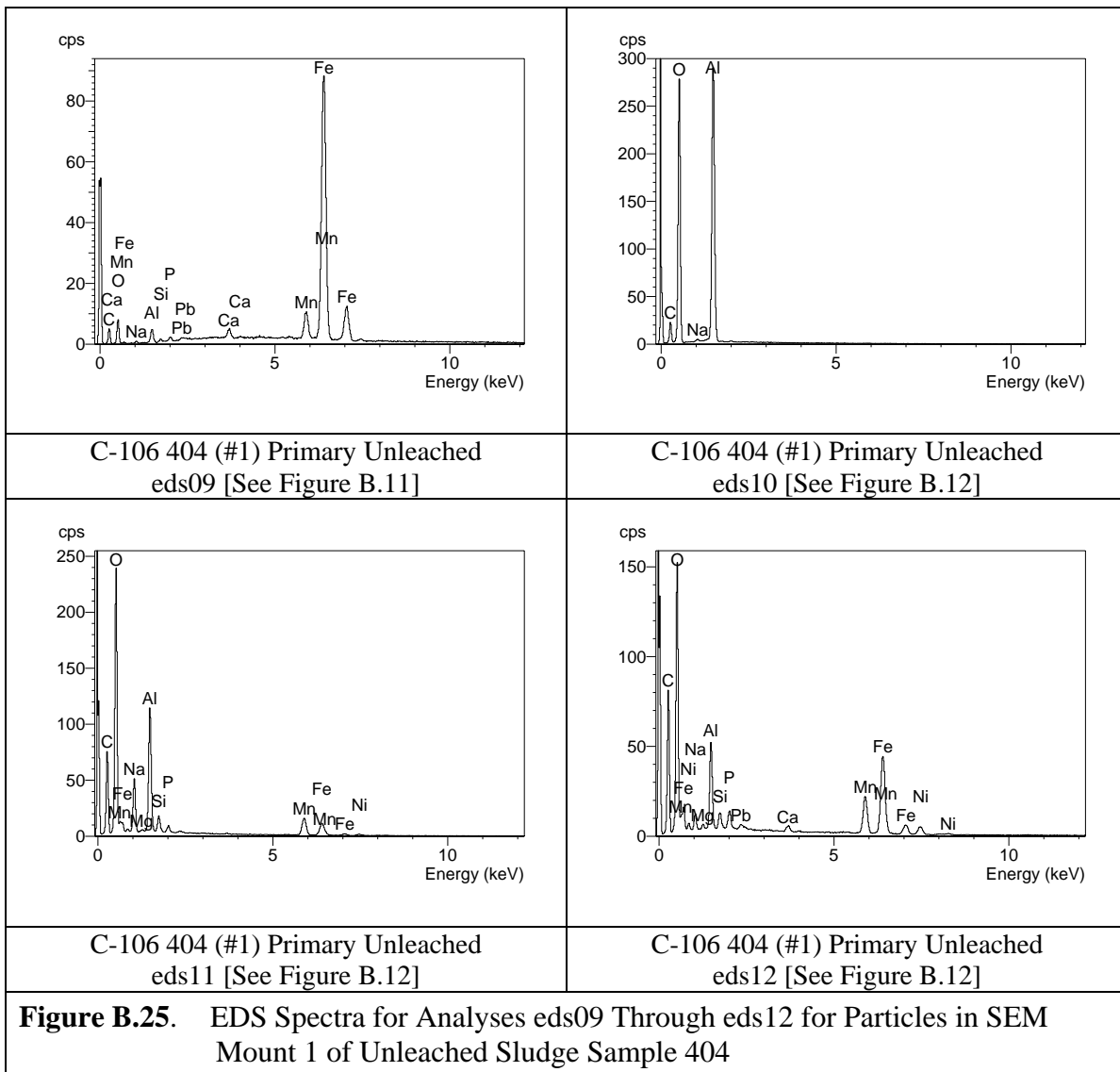| C-106 405 (#6) Duplicate Unleached eds22 [See Figure B.62] | C-106 405 (#6) Duplicate Unleached eds23 [See Figure B.63] |
| C-106 405 (#6) Duplicate Unleached eds24 [See Figure B.64] | C-106 405 (#6) Duplicate Unleached eds25 [See Figure B.65] |

Figure B.69. EDS Spectra for Analyses eds22 Through eds25 for Particles in SEM Mount 6 of Unleached Sludge Sample 405

C-106 405 (#6) Duplicate Unleached
eds26 [See Figure B.65]

C-106 405 (#6) Duplicate Unleached
eds27 [See FigureB.66]

C-106 405 (#6) Duplicate Unleached
eds28 [See Figure B.66]

C-106 405 (#6) Duplicate Unleached
eds29 [See Figure B.67]

Figure B.70.   EDS Spectra for Analyses eds26 Through eds29 for Particles in SEM Mount 6 of
Unleached Sludge Sample 405

Appendix C

SEM Micrographs and EDS Spectra
for Water-Leached Sludge from Tank C-106

Appendix C

SEM Micrographs and EDS Spectra for Water-Leached Sludge from Tank C-106

This appendix includes the scanning electron microscope (SEM) micrographs and the energy-dispersive X-ray spectrometry (EDS) spectra for samples of tank 241-C-106 (C-106) (primary 404 and duplicate 405) 1-month and 82-day water-leached residual tank waste. The operating conditions for the SEM and procedures used for mounting the SEM samples are described in Section 2.4 of the main report.

The name of each SEM digital image file, sample identification number, and a size scale bar are given, respectively, at the bottom left, center, and right of each SEM micrograph in this appendix. Micrographs labeled by "BSE" to the immediate right of the digital image file name indicate that the micrograph was collected with backscattered electrons. Areas identified by a letter and/or outlined by a dashed-line square in a micrograph designate sample material that was imaged at higher magnification, which is typically shown in figure(s) that immediately follow in the series for that sample.

Areas labeled by "eds" in the following SEM micrographs in this appendix identify locations of particles for which EDS spectra were recorded. The "eds" label given with each EDS spectrum correspond to the same "EDS" label used in the SEM micrographs for this sample.

C.1 Water-Leached (1-Month) Sample 404

Two mounts of 1-month water-leached residual sludge from tank C-106 (primary 404) were analyzed by SEM/EDS. The SEM micrographs for the first mount (7) of the 1-month water-leached sample 404 are shown in Figures C.1 through C.15. The EDS spectra for this mount are given in Figures C.16 through C.19. The SEM micrographs for the second mount (8) of the 1-month water-leached sample 404 are shown in Figures C.20 through C.27, and the EDS spectra for the second mount are given in Figures C.28 through C.30.

C.2
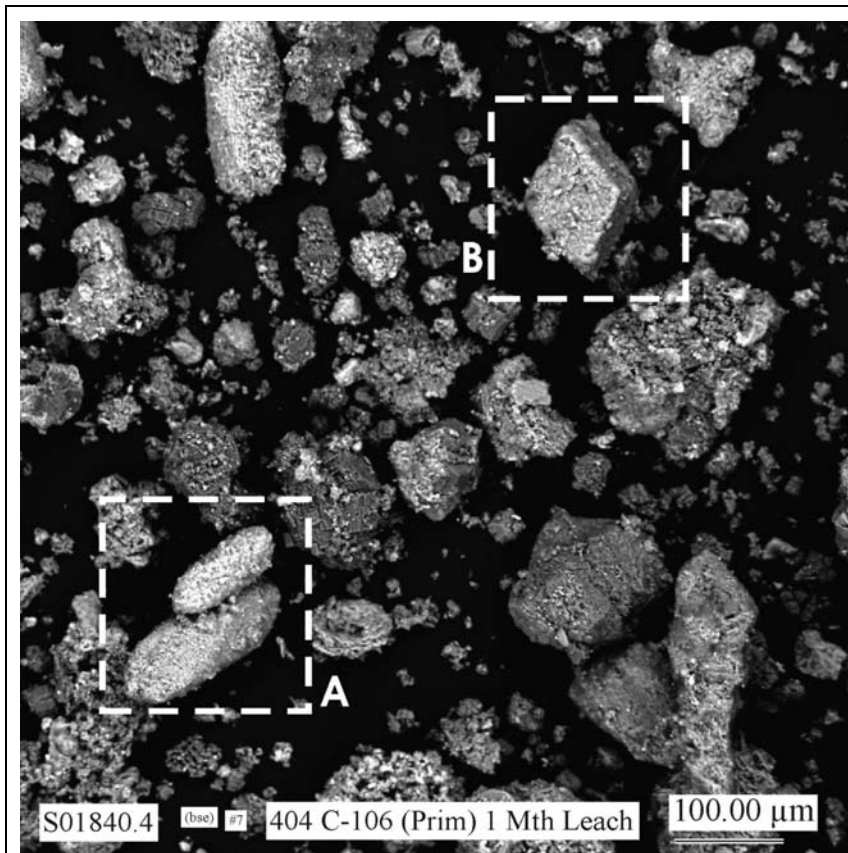
Figure C.1. Low Magnification SEM Micrograph Showing General Morphologies of Particles in SEM Mount 7 of 1-Month Water-Leached Sludge Sample 404

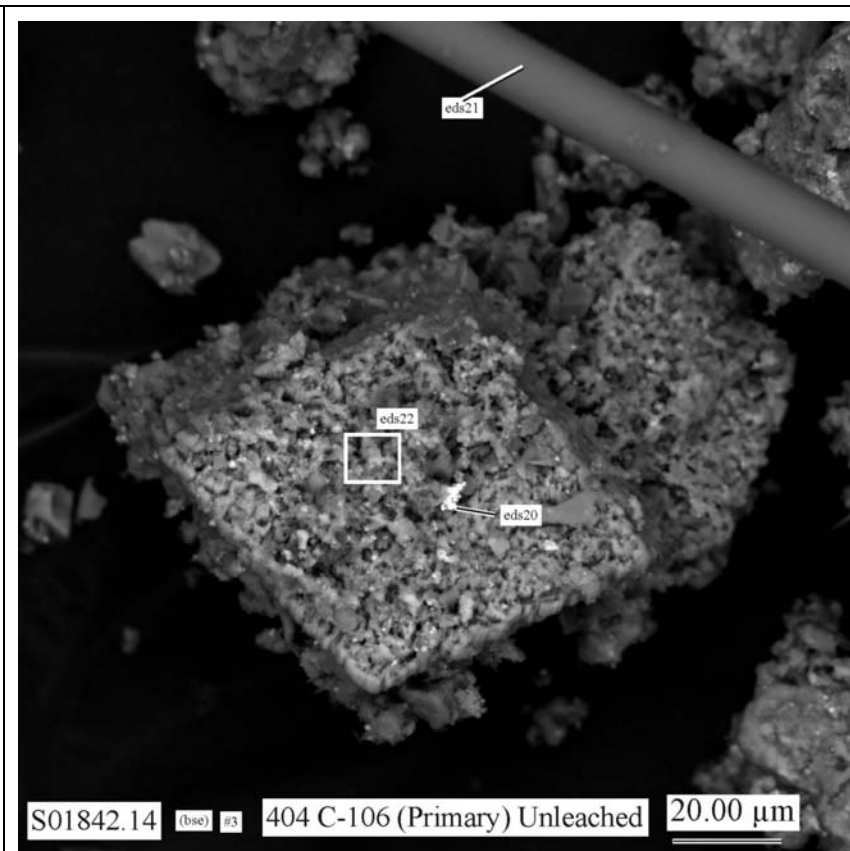**Figure C.2.** Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled A in Figure C.1

| S01840.5 | (bse) | #7 | 404 C-106 (Prim) 1 Mth Leach | 30.00 μm |

| S01840.7 | (bse) | #7 | 404 C-106 (Prim) 1 Mth Leach | 10.00 μm |

Figure C.3. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled B in Figure C.1
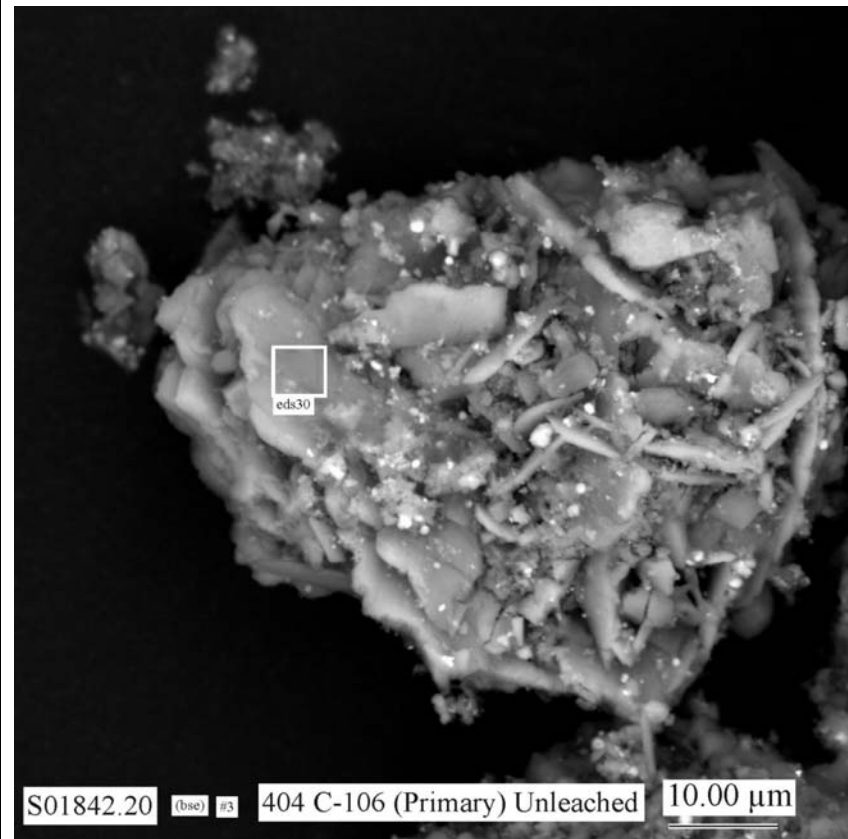
Figure C.4. Micrograph Showing Morphologies of Typical Particles in SEM Mount 7 of 1-Month Water-Leached Sludge Sample 404

S01840.1 (bse) #7 404 C-106 (Prim) 1 Mth Leach 50.00 µm
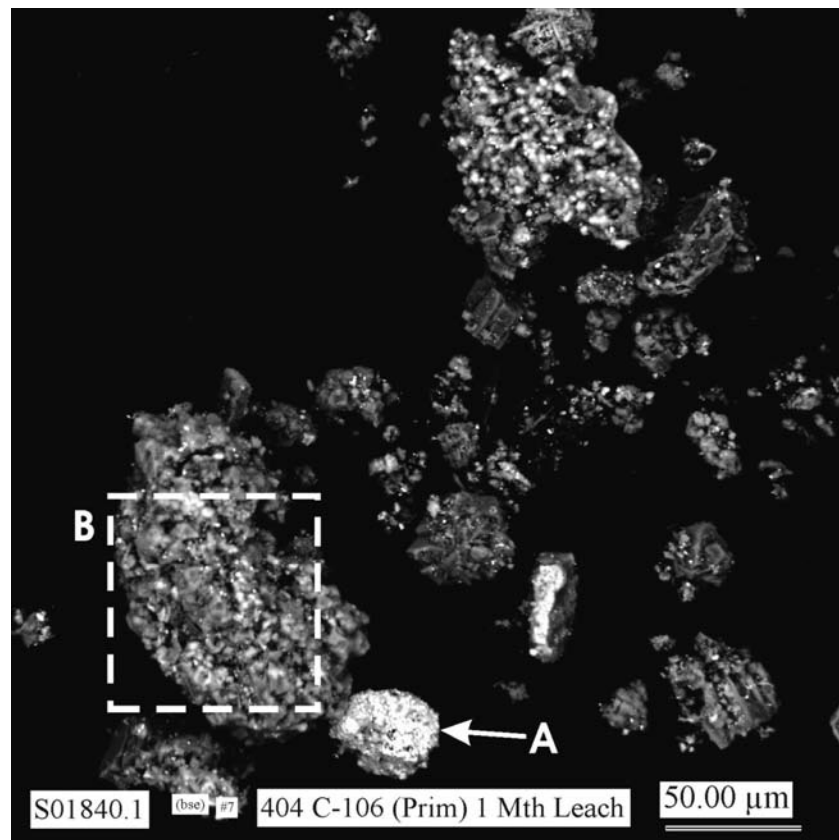
Figure C.5. Micrograph Showing Morphologies of Typical Particles in SEM Mount 7 of the 1-Month Water-Leached Sludge Sample 404

S01840.2 (bse) #7 404 C-106 (Prim) 1 Mth Leach 8.00 µm

Figure C.6. Micrograph Showing at Higher Magnification the Particle Labeled A in Figure C.5

Figure C.7. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled B in Figure C.5
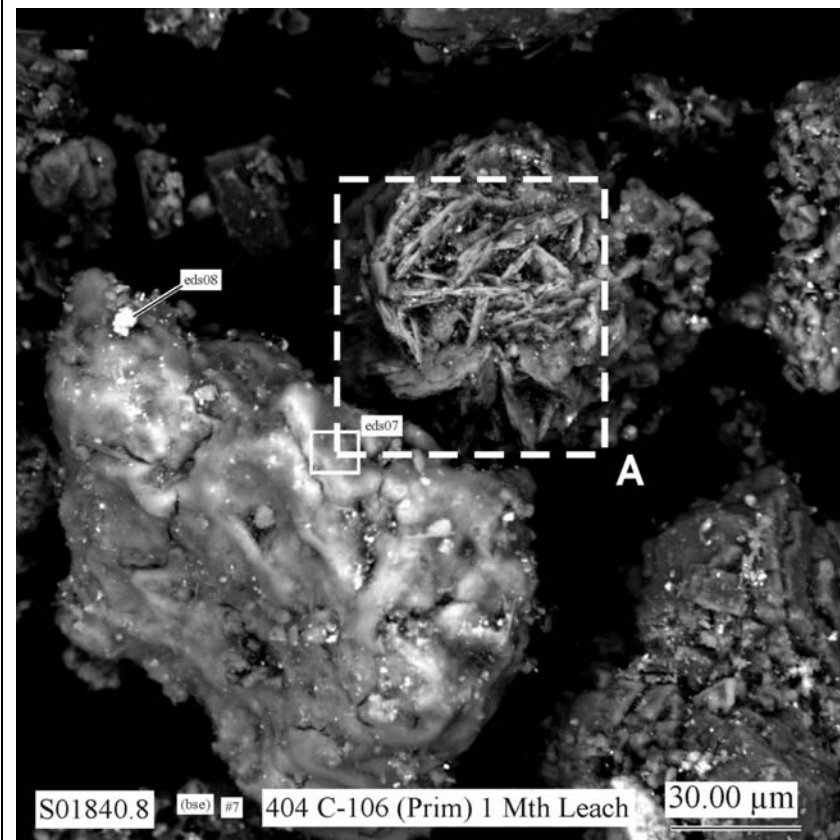
Figure C.8. Micrograph Showing Morphologies of Typical Particles in SEM Mount 7 of the 1-Month Water-Leached Sludge Sample 404

S01840.9 (bse) #7 404 C-106 (Prim) 1 Mth Leach 10.00 µm
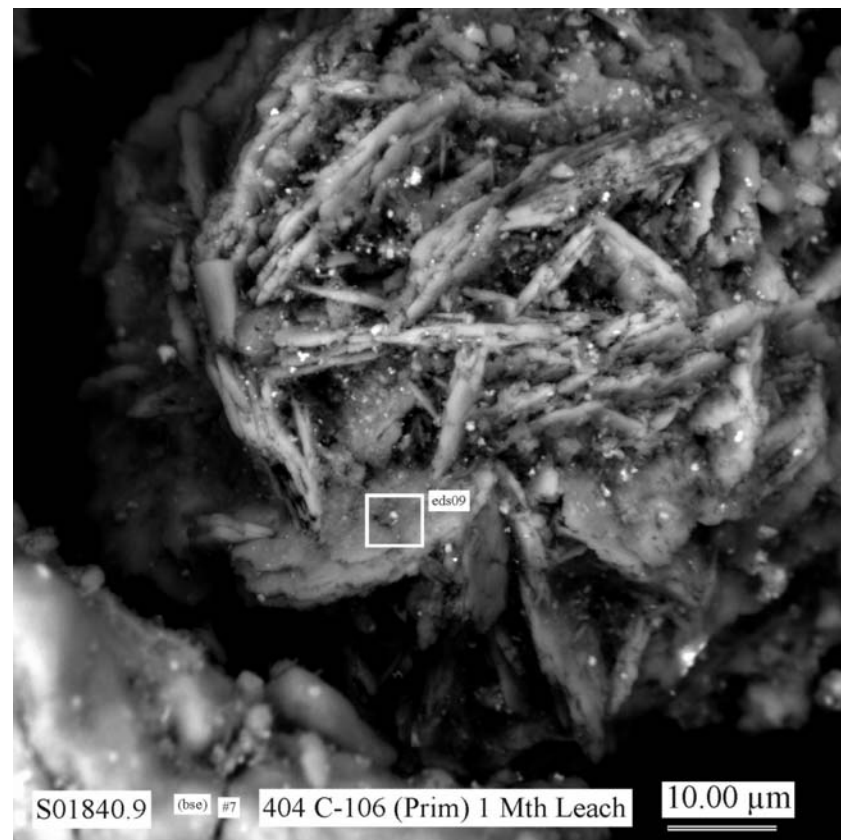
Figure C.9. Micrograph Showing at Higher Magnification the Particle Labeled A in Figure C.8

S01840.11 (bse) #7 404 C-106 (Prim) 1 Mth Leach 10.00 µm
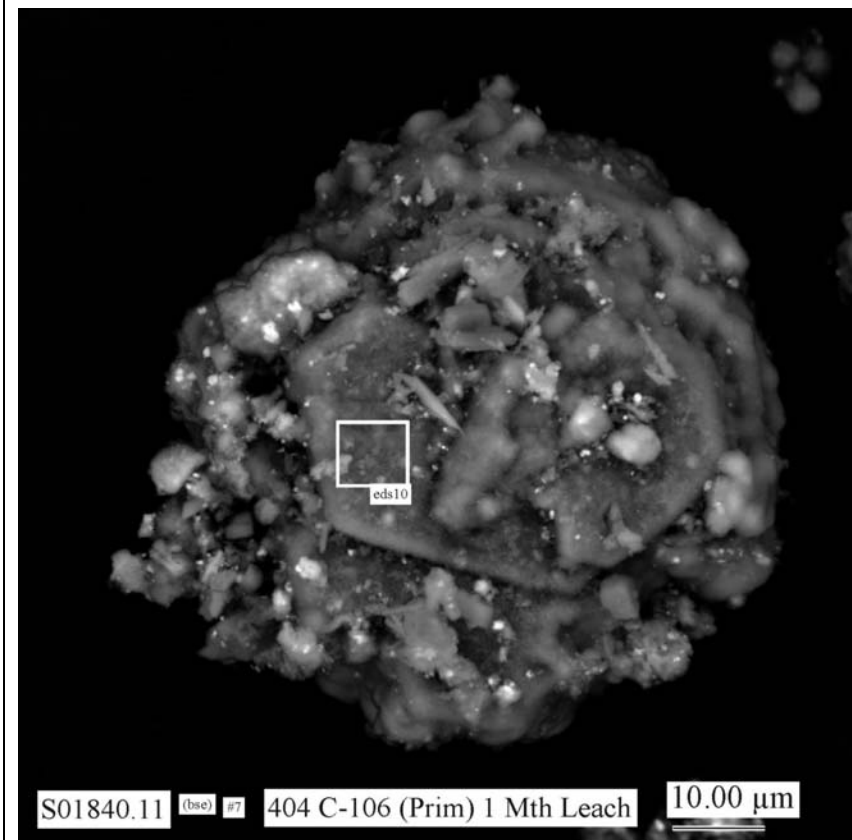
Figure C.10. Micrograph Showing Morphologies of Typical Particles in SEM Mount 7 of the 1-Month Water-Leached Sludge Sample 404
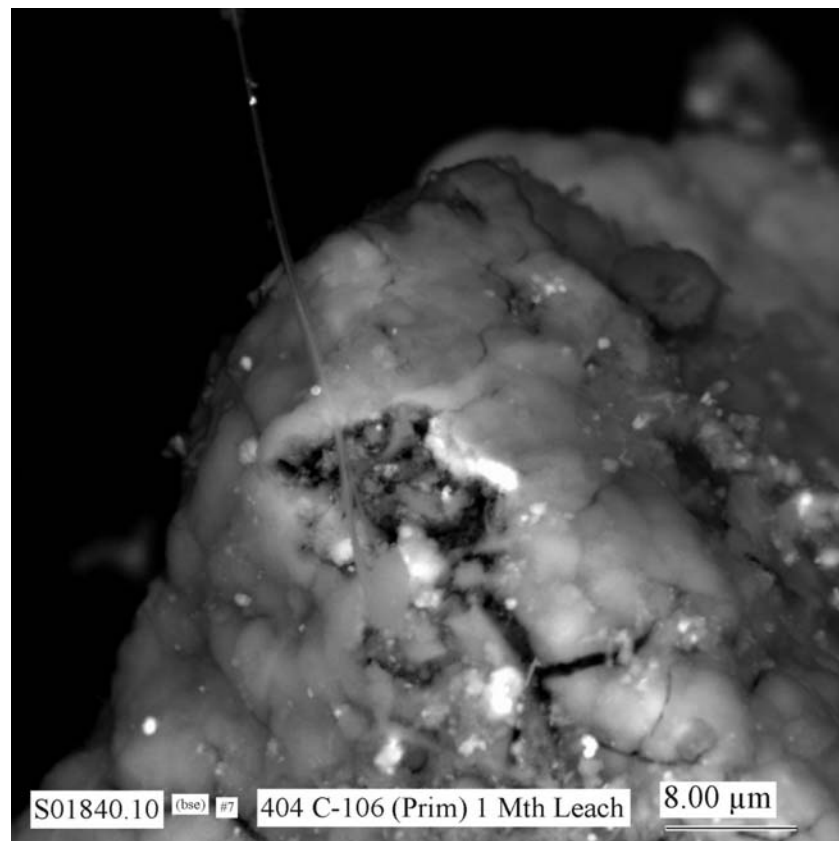
Figure C.11. Micrograph Showing Morphologies of Typical Particles in SEM Mount 7 of the 1-Month Water-Leached Sludge Sample 404
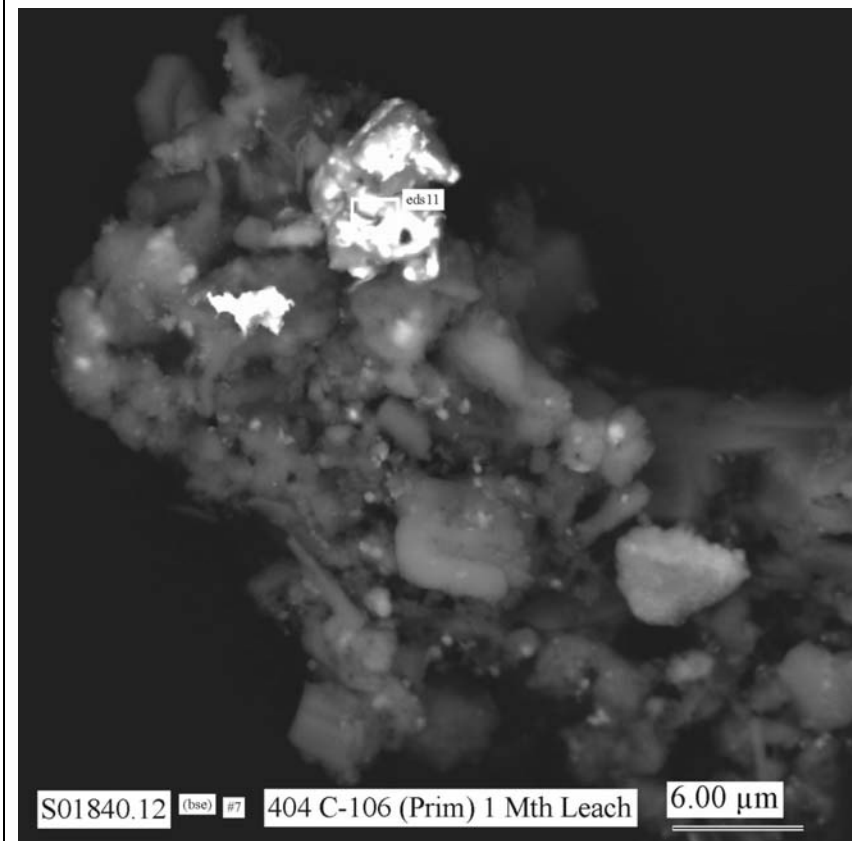
Figure C.12. Micrograph Showing Morphologies of Typical Particles in SEM Mount 7 of the 1-Month Water-Leached Sludge Sample 404
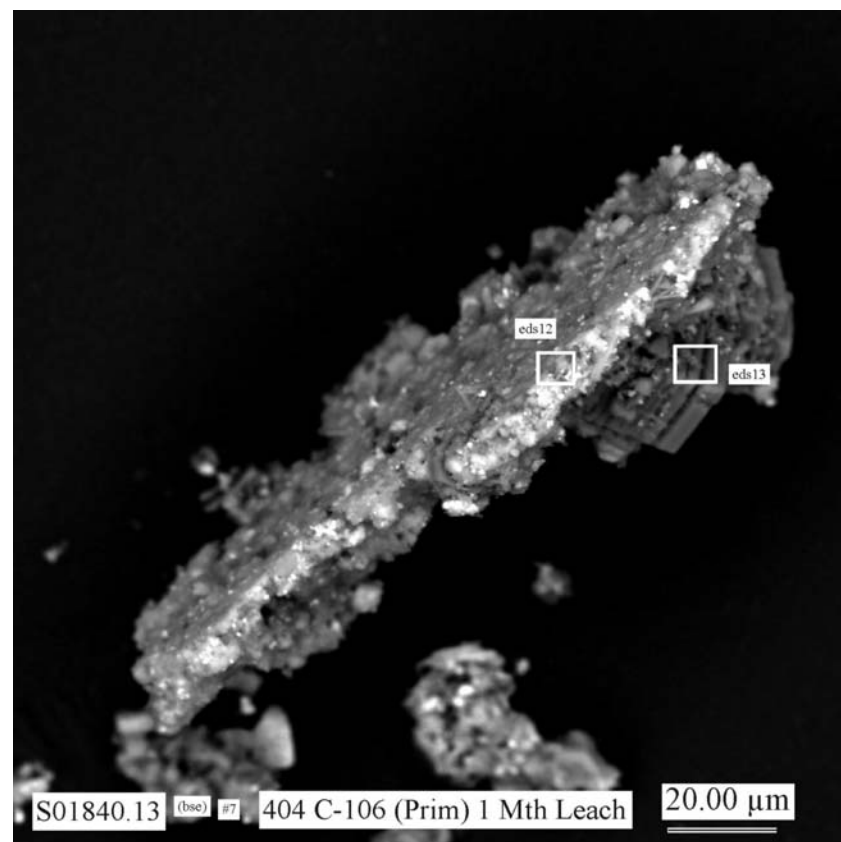
Figure C.13. Micrograph Showing Morphologies of Typical Particles in SEM Mount 7 of the 1-Month Water-Leached Sludge Sample 404
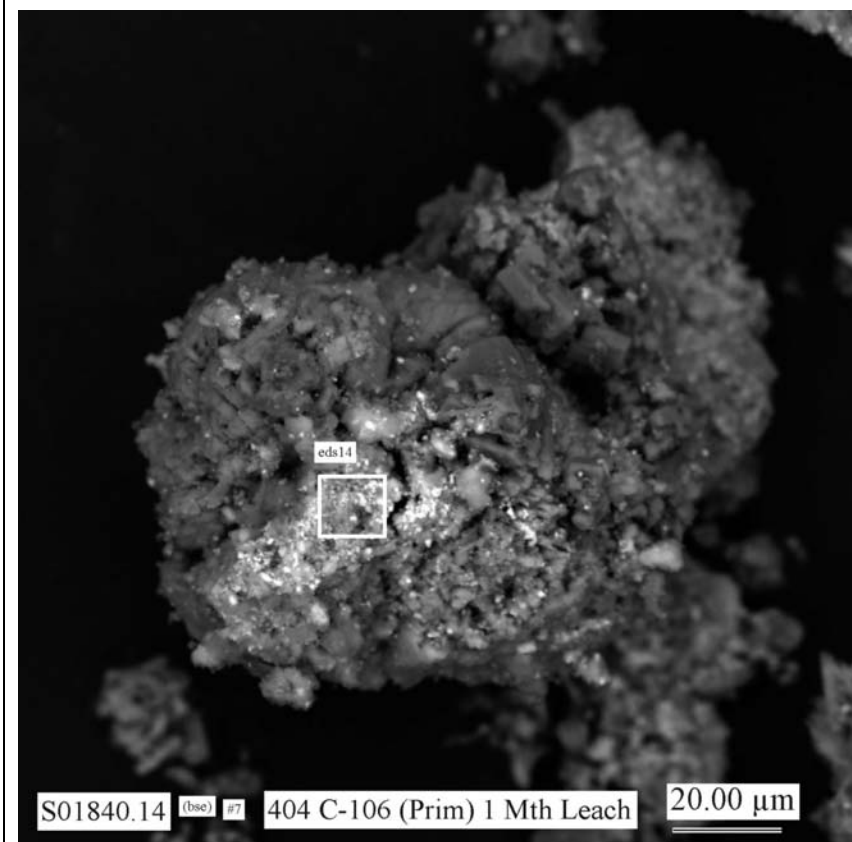
Figure C.14. Micrograph Showing Morphologies of Typical Particles in SEM Mount 7 of the 1-Month Water-Leached Sludge Sample 404
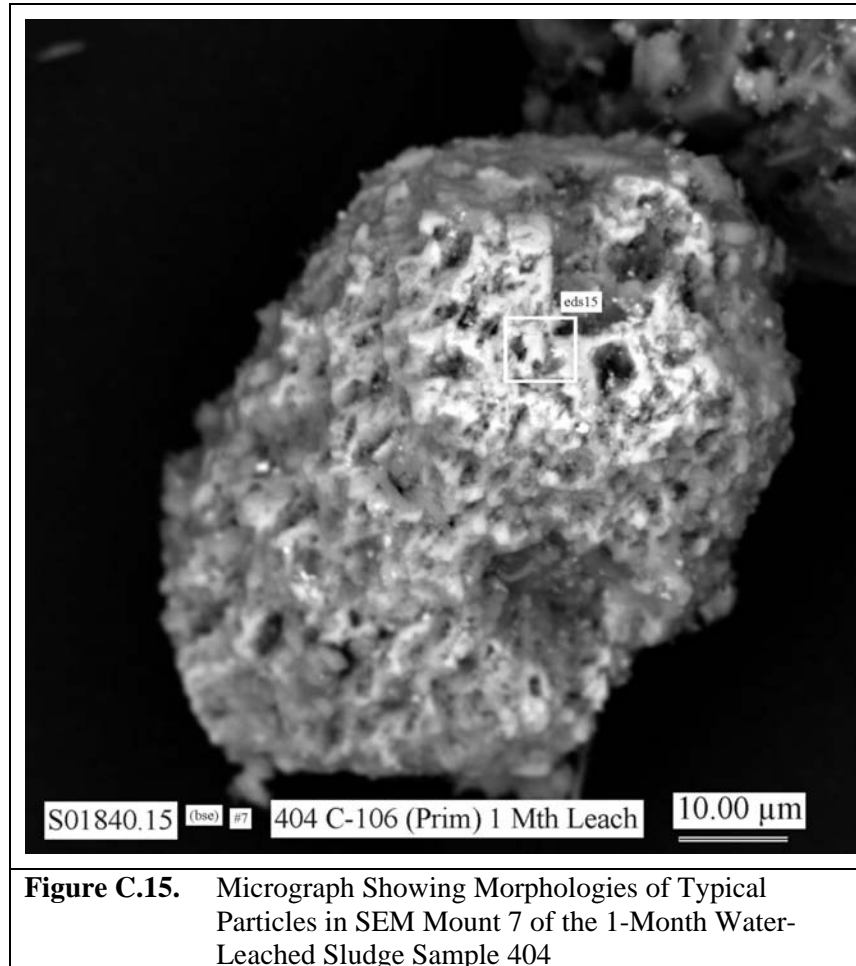
Figure C.15. Micrograph Showing Morphologies of Typical Particles in SEM Mount 7 of the 1-Month Water-Leached Sludge Sample 404

C-106 404 (#7) Primary 1-month Leached eds01 [See Figure C.6]	C-106 404 (#7) Primary 1-month Leached eds02 [See Figure C.7]
C-106 404 (#7) Primary 1-month Leached eds03 [See Figure C.3]	C-106 404 (#7) Primary 1-month Leached eds04 [See Figure C.2]

Figure C.16.   EDS Spectra for Analyses eds01 Through eds04 for Particles in SEM Mount 7 of the 1-Month Water-Leached Sludge Sample 404

| C-106 404 (#7) Primary 1-month Leached eds05 [See Figure C.4] | C-106 404 (#7) Primary 1-month Leached eds06 [See Figure C.4] |
| C-106 404 (#7) Primary 1-month Leached eds07 [See Figure C.8] | C-106 404 (#7) Primary 1-month Leached eds08 [See Figure C.8] |

Figure C.17.   EDS Spectra for Analyses eds05 Through eds08 for Particles in SEM Mount 7 of the 1-Month Water-Leached Sludge Sample 404

| C-106 404 (#7) Primary 1-month Leached eds09 [See Figure C.9] | C-106 404 (#7) Primary 1-month Leached eds10 [See Figure C.10] |
| C-106 404 (#7) Primary 1-month Leached eds11 [See Figure C.12] | C-106 404 (#7) Primary 1-month Leached eds12 [See Figure C.13] |

Figure C.18.   EDS Spectra for Analyses eds09 Through eds12 for Particles in SEM Mount 7 of the 1-Month Water-Leached Sludge Sample 404

C-106 404 (#7) Primary 1-month Leached
eds13 [See Figure C.13]

C-106 404 (#7) Primary 1-month Leached
eds14 [See Figure C.14]

C-106 404 (#7) Primary 1-month Leached
eds15 [See Figure C.15]

Figure C.19. EDS Spectra for Analyses eds13 Through eds15 for Particles in SEM Mount 7 of the 1-Month Water-Leached Sludge Sample 404

C.13

S01842.21 (bse) #8 404 C-106 (Primary) 1 Mth Leach 50.00 µm

| **Figure C.20.** | Micrograph Showing Morphologies of Typical Particles in SEM Mount 8 of the 1-Month Water-Leached Sludge Sample 404 |

S01842.22 (bse) #8 404 C-106 (Primary) 1 Mth Leach 20.00 µm
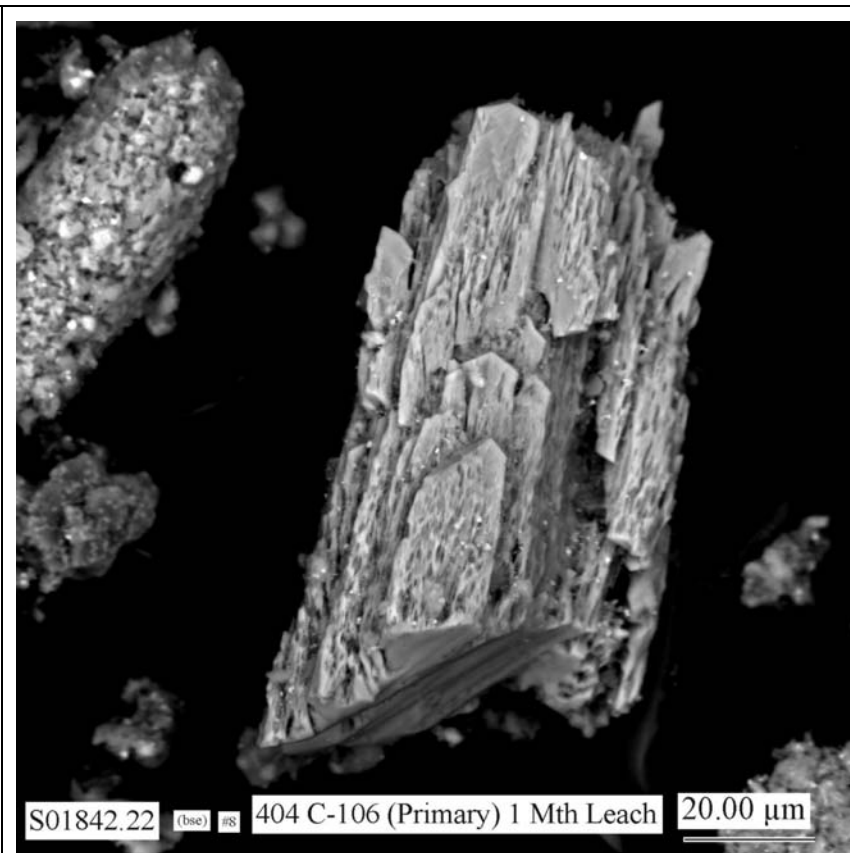
| **Figure C.21.** | Micrograph Showing Morphologies of Typical Particles in SEM Mount 8 of the 1-Month Water-Leached Sludge Sample 404 |
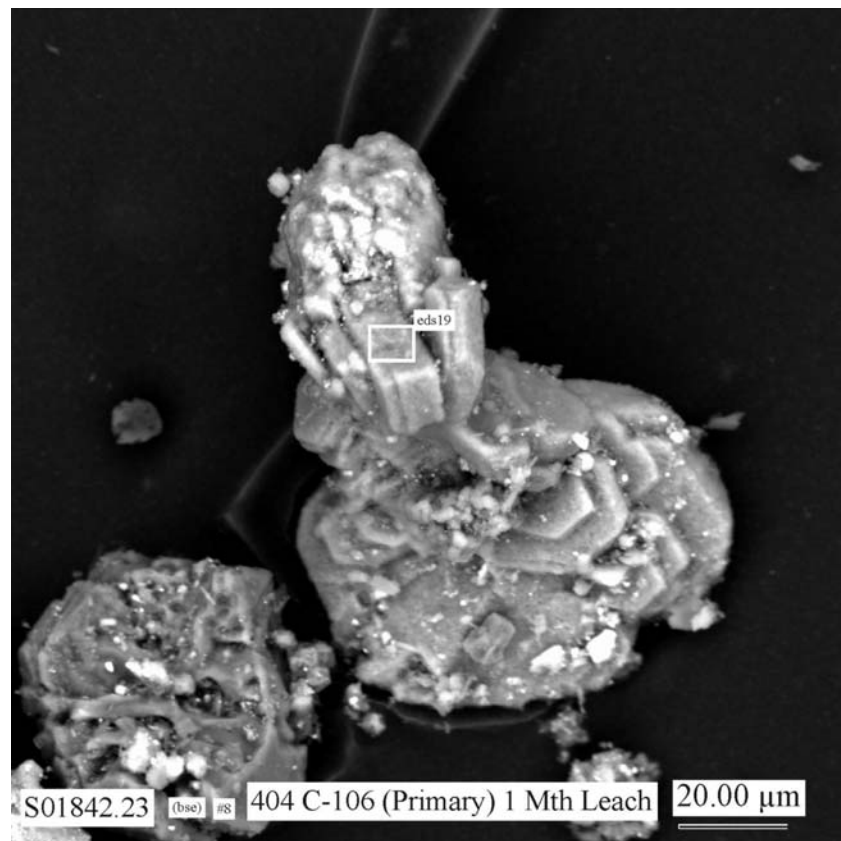
Figure C.22. Micrograph Showing Morphologies of Typical Particles in SEM Mount 8 of the 1-Month Water-Leached Sludge Sample 404
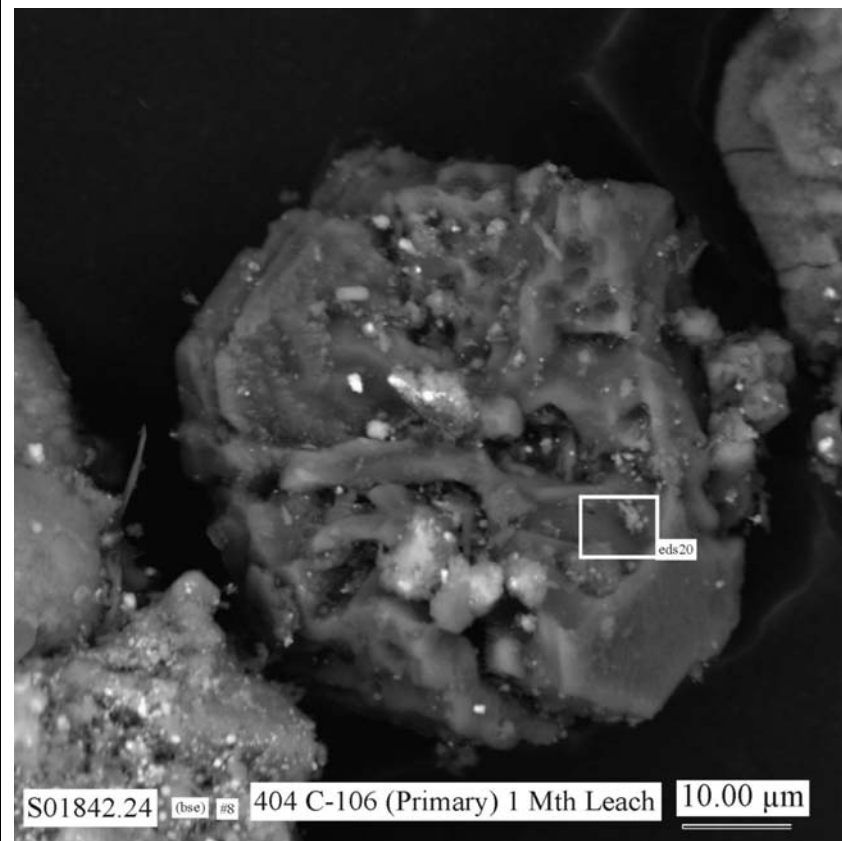
Figure C.23. Micrograph Showing Morphologies of Typical Particles in SEM Mount 8 of the 1-Month Water-Leached Sludge Sample 404

S01842.25 (bse) #8 404 C-106 (Primary) 1 Mth Leach 100.00 μm
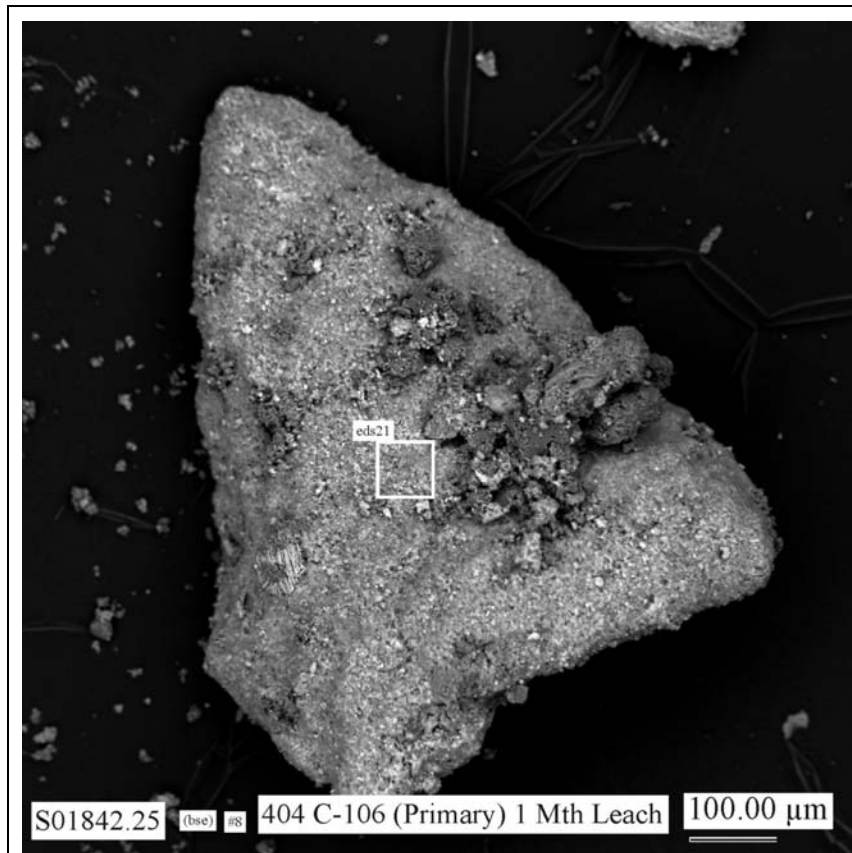
Figure C.24. Micrograph Showing Morphologies of Typical Particles in SEM Mount 8 of the 1-Month Water-Leached Sludge Sample 404

S01842.26 (bse) #8 404 C-106 (Primary) 1 Mth Leach 8.00 μm
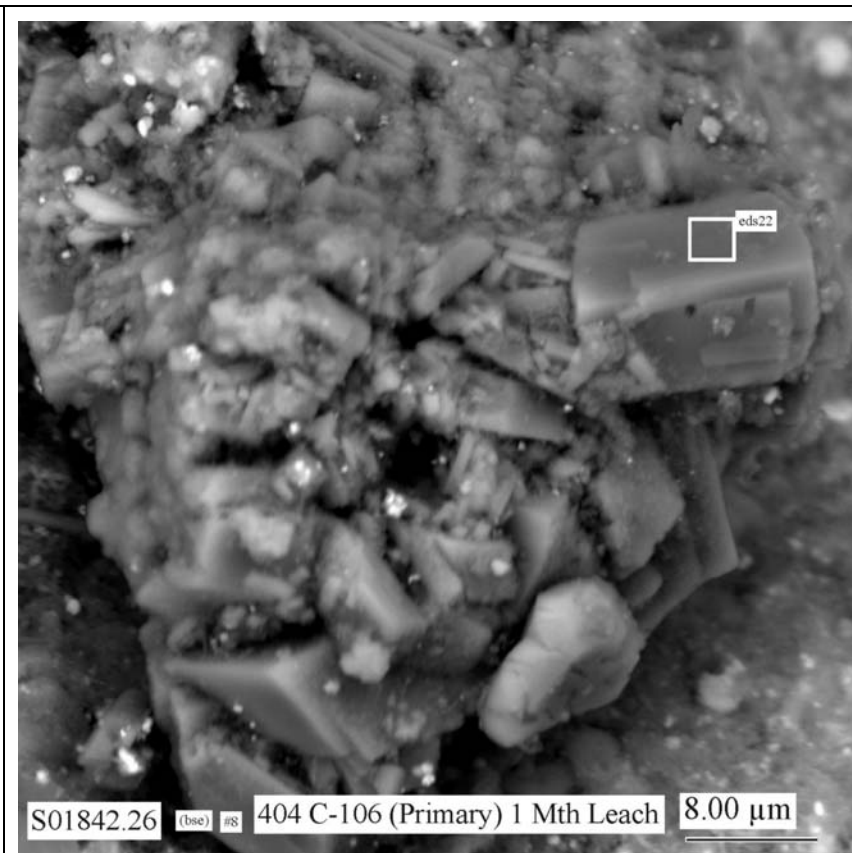
Figure C.25. Micrograph Showing Morphologies of Typical Particles in SEM Mount 8 of the 1-Month Water-Leached Sludge Sample 404

S01842.27 (bse) #8 404 C-106 (Primary) 1 Mth Leach 10.00 μm
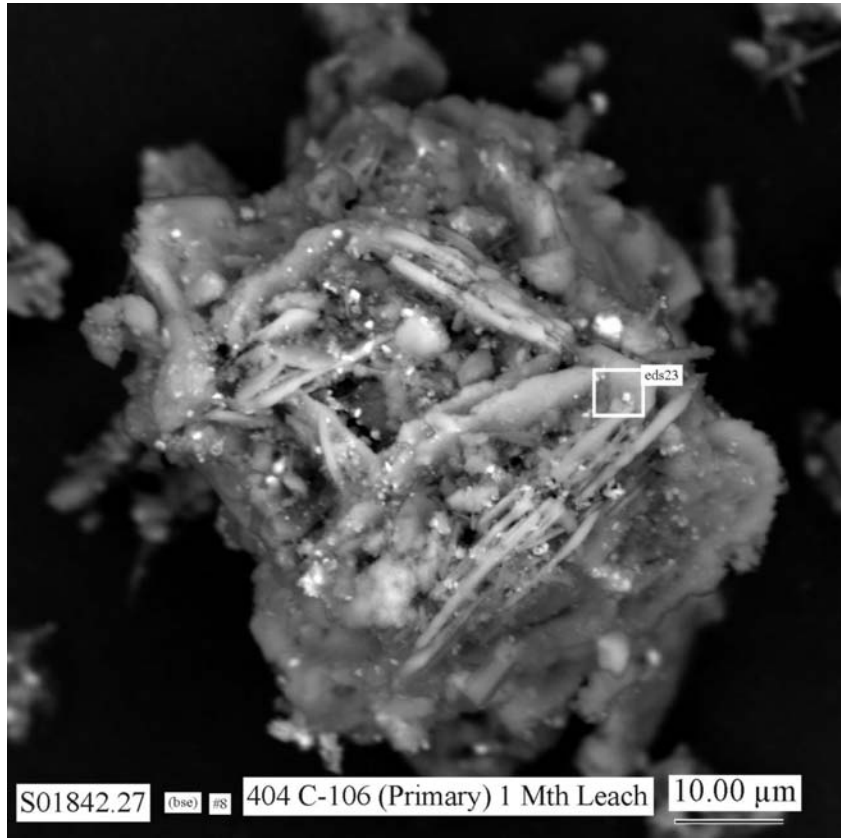
Figure C.26. Micrograph Showing Morphologies of Typical Particles in SEM Mount 8 of the 1-Month Water-Leached Sludge Sample 404

S01842.28 (bse) #8 404 C-106 (Primary) 1 Mth Leach 20.00 μm
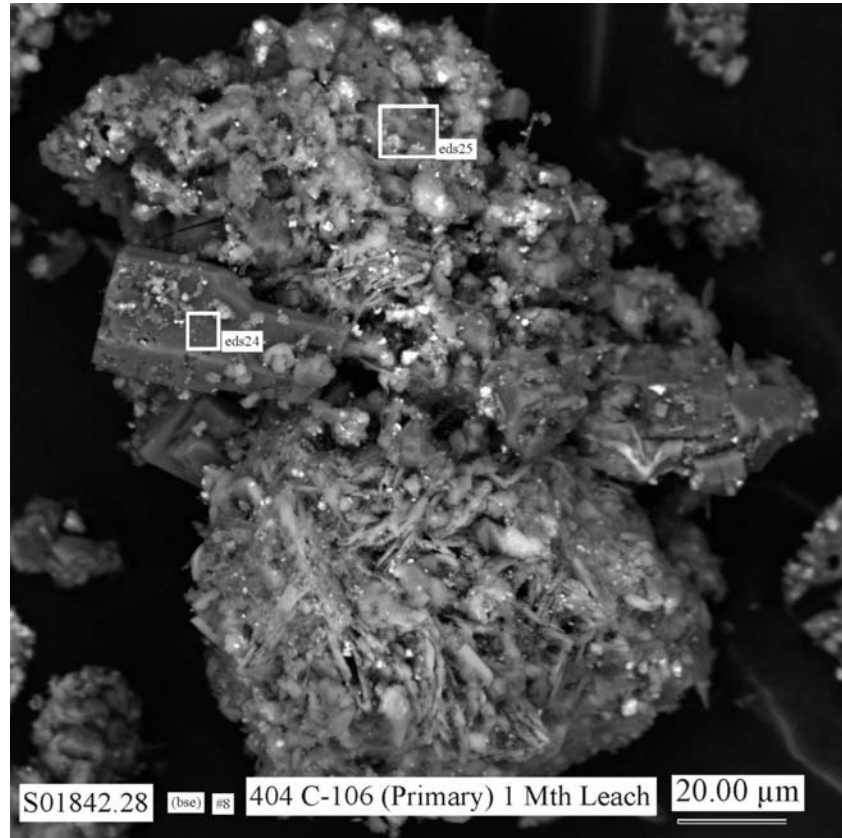
Figure C.27. Micrograph Showing Morphologies of Typical Particles in SEM Mount 8 of the 1-Month Water-Leached Sludge Sample 404

C-106 404 (#8) Primary 1-month Leached
eds16 [See Figure C.20]

C-106 404 (#8) Primary 1-month Leached
eds17 [See Figure C.20]

C-106 404 (#8) Primary 1-month Leached
eds19 [See Figure C.22]

C-106 404 (#8) Primary 1-month Leached
eds20 [See Figure C.23]

Figure C.28.   EDS Spectra for Analyses eds16, 17, 19, and 20 for Particles in SEM Mount 8 of
the 1-Month Water-Leached Sludge Sample 404

C-106 404 (#8) Primary 1-month Leached eds21 [See Figure C.24]	C-106 404 (#8) Primary 1-month Leached eds22 [See Figure C.25]
C-106 404 (#8) Primary 1-month Leached eds23 [See Figure C.26]	C-106 404 (#8) Primary 1-month Leached eds24 [See Figure C.27]

Figure C.29.   EDS Spectra for Analyses eds21 through eds24 for Particles in SEM Mount 8 of the 1-Month Water-Leached Sludge Sample 404

C-106 404 (#8) Primary 1-month Leached
eds25 [See Figure C.27]

Figure C.30.   EDS Spectra for Analyses
eds24 through eds25 for
Particles in SEM Mount 8 of
the 1-Month Water-Leached
Sludge Sample 404

C.2 Water-Leached (1-Month) Sample 405

Two mounts of the 1-month water-leached residual sludge from tank C-106 (duplicate 405) were analyzed by SEM/EDS. The SEM micrographs for the first mount (10) of the 1-month water-leached sample 405 sludge are shown in Figures C.31 through C.44 The EDS spectra for this mount are given in Figures C.45 through C.48. The SEM micrographs for the second mount (11) of sample 405 are shown in Figures C.49 through C.54, and the EDS spectra for the second mount are given in Figures C.55 through C.56.

S01841.1 (bse) #10 405 C-106 (Dup) 1 Mth Leach   60.00 µm

S01841.2 (bse) #10 405 C-106 (Dup) 1 Mth Leach   10.00 µm
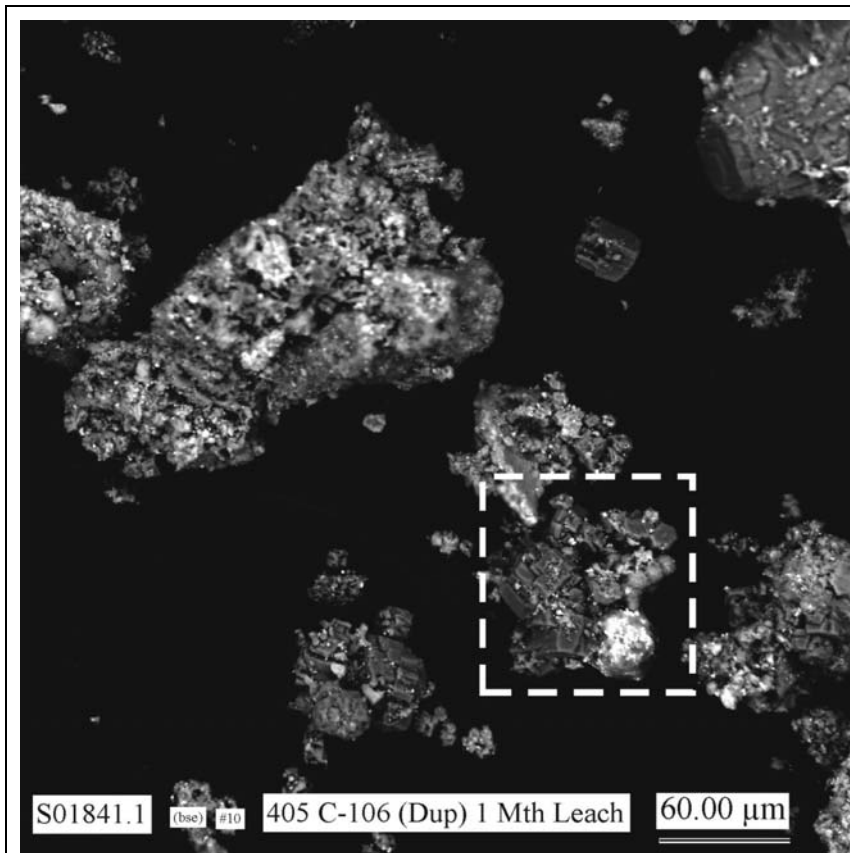
| **Figure C.31**. | Low Magnification SEM Micrograph Showing General Morphologies of Particles in SEM Mount 10 of the 1-Month Water-Leached Sludge Sample 405 | **Figure C.32.** | Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square in Figure C.31 |

S01841.3 (bse) #10 405 C-106 (Dup) 1 Mth Leach 3.00 μm

Figure C.33. Micrograph Showing at Higher Magnification the Spherical Particles Labeled A in Figure C.32

S01841.4 (bse) #10 405 C-106 (Dup) 1 Mth Leach 4.00 μm

Figure C.34. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled B in Figure C.32

Figure C.35. Low Magnification SEM Micrograph Showing General Morphologies of Particles in SEM Mount 10 of the 1-Month Water-Leached Sludge Sample 405

Figure C.36. Micrograph Showing at Higher Magnification the Large Particle in the Area Indicated by the White Dashed-Line Square Labeled A in Figure C.33
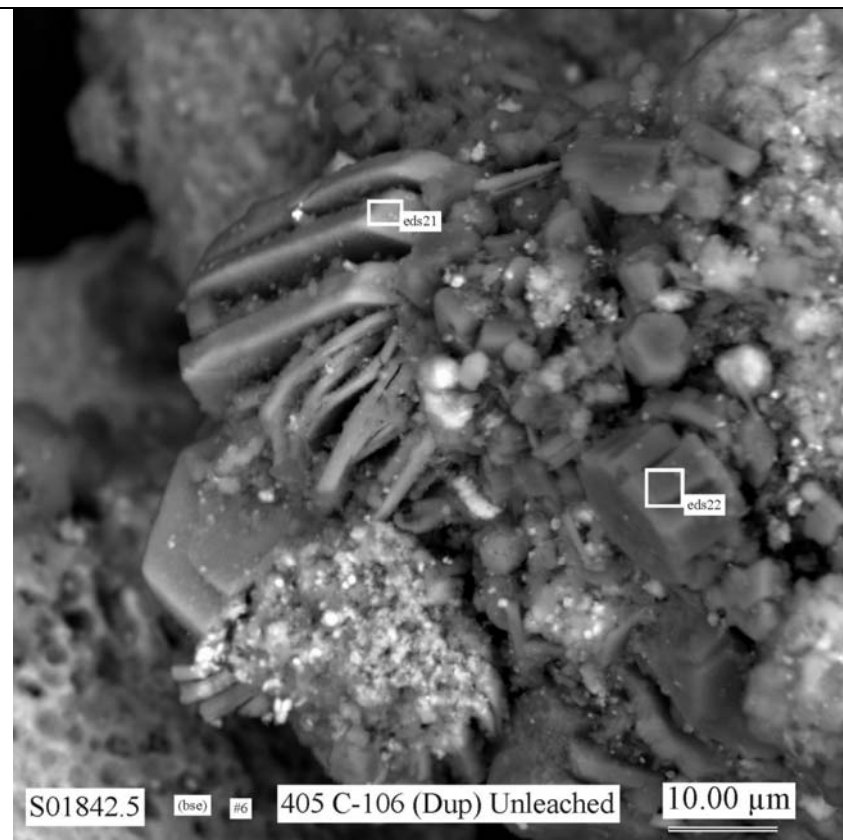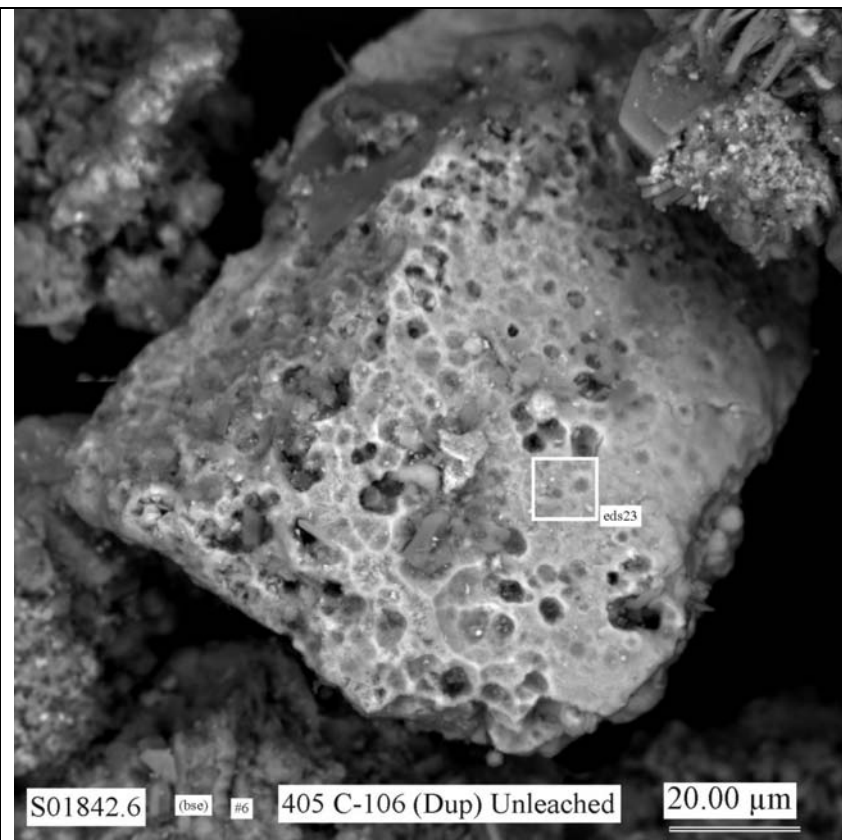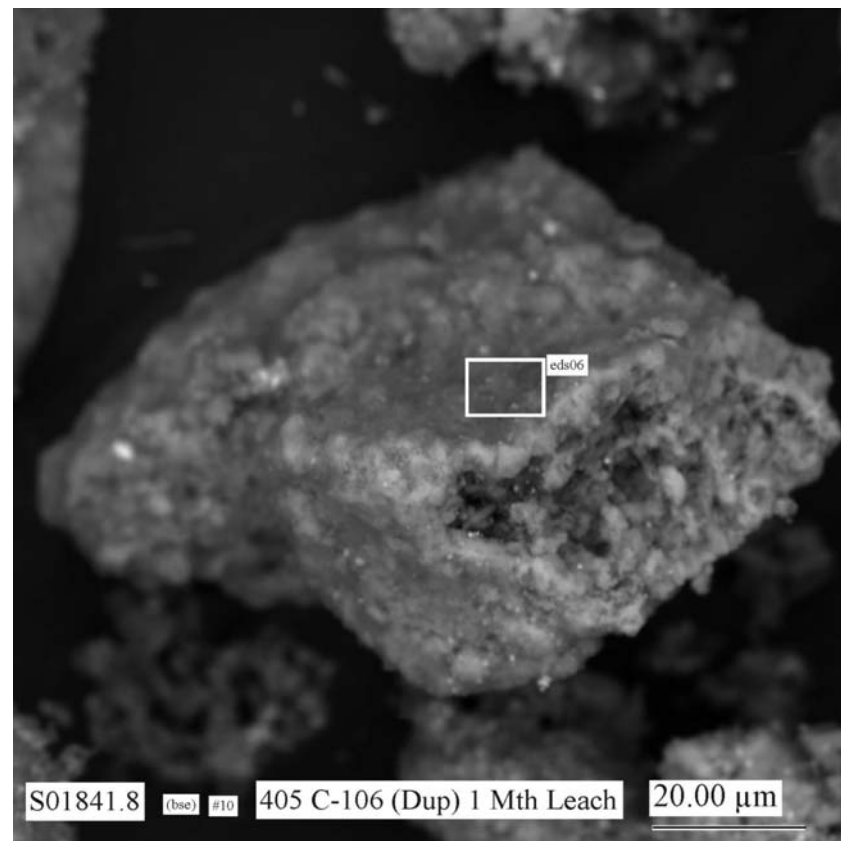
Figure C.37. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled B in Figure C.33

Figure C.38. Micrograph Showing at Higher Magnification the Large Particle Labeled C in Figure C.33

S01841.10 (bse) #10 | 405 C-106 (Dup) 1 Mth Leach | 20.00 µm
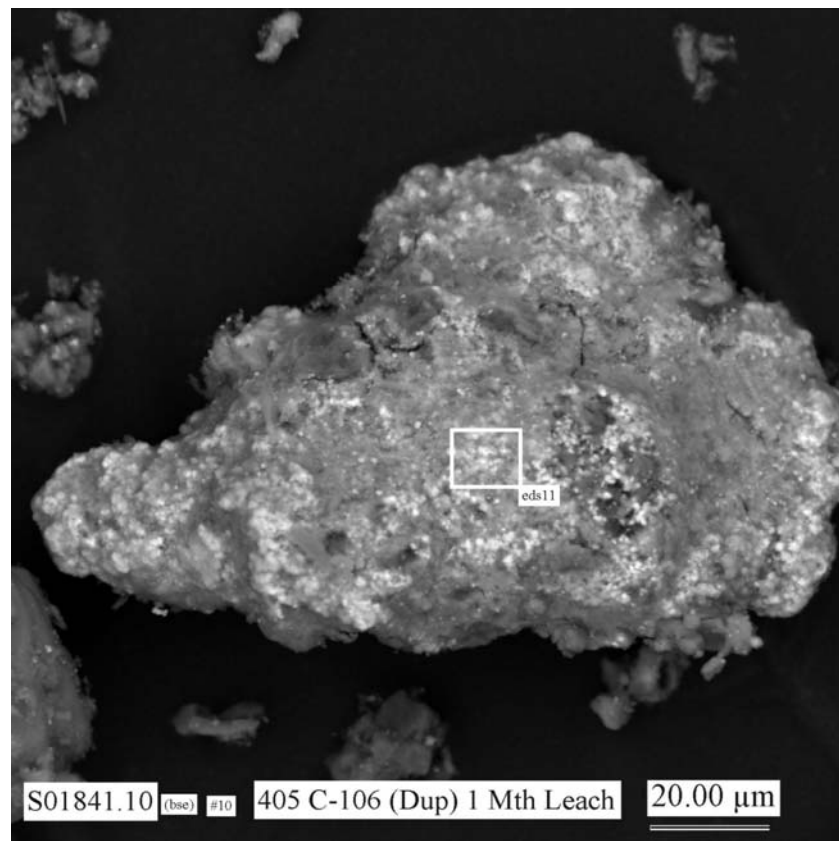
| **Figure C.39.** | Micrograph Showing Morphologies of Typical Particles in SEM Mount 10 of the 1-Month Water-Leached Sludge Sample 405 |

S01841.11 (bse) #10 | 405 C-106 (Dup) 1 Mth Leach | 30.00 µm
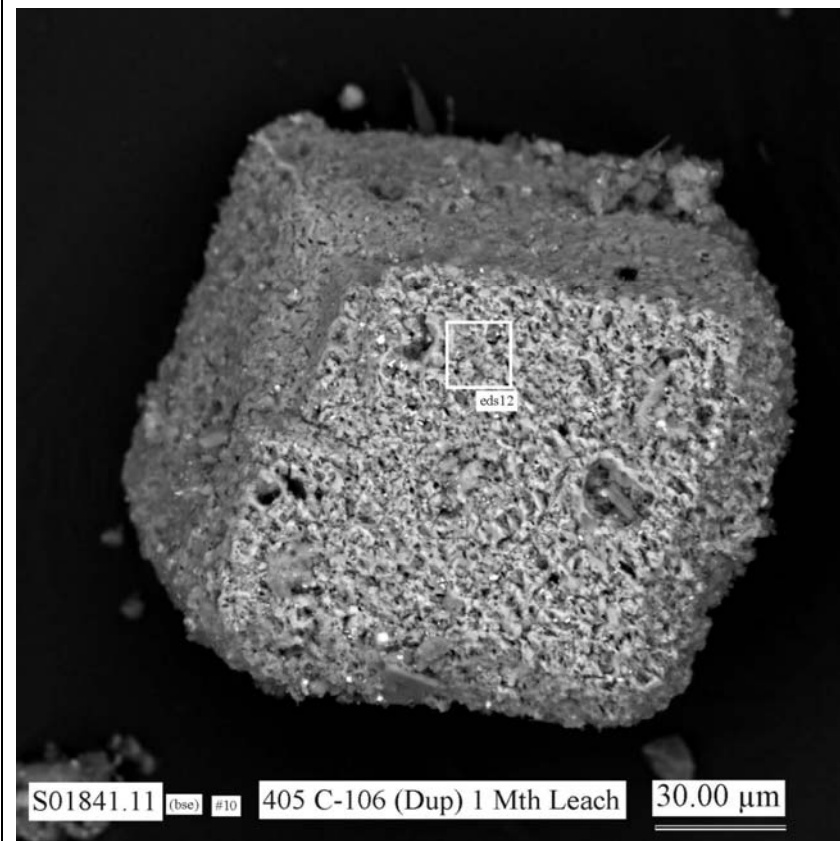
| **Figure C.40.** | Micrograph Showing Morphologies of Typical Particles in SEM Mount 10 of the 1-Month Water-Leached Sludge Sample 405 |
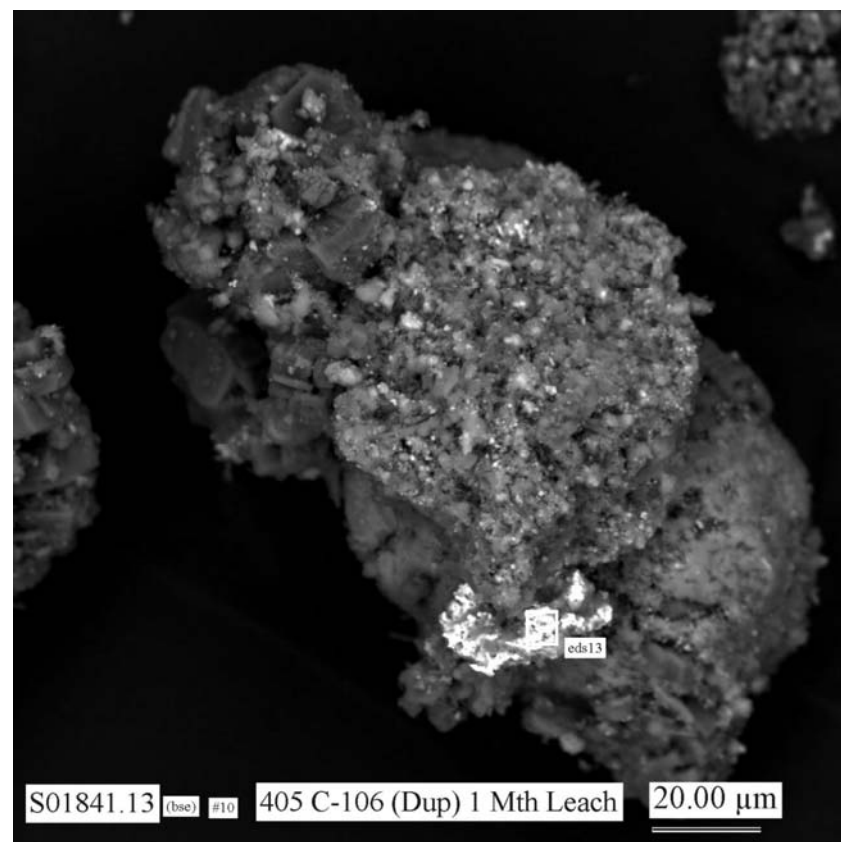
Figure C.41. Micrograph Showing Morphologies of Typical Particles in SEM Mount 10 of the 1-Month Water-Leached Sludge Sample 405
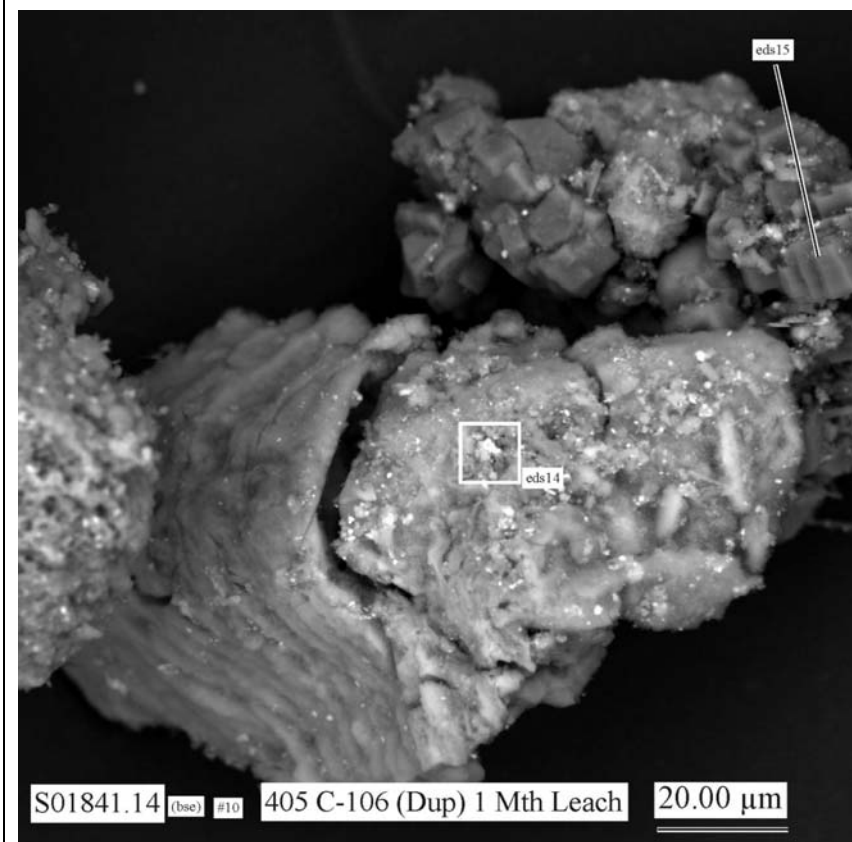
Figure C.42. Micrograph Showing Morphologies of Typical Particles in SEM Mount 10 of the 1-Month Water-Leached Sludge Sample 405

S01841.15 (bse) #10 405 C-106 (Dup) 1 Mth Leach 50.00 µm

S01841.16 (bse) #10 405 C-106 (Dup) 1 Mth Leach 50.00 µm
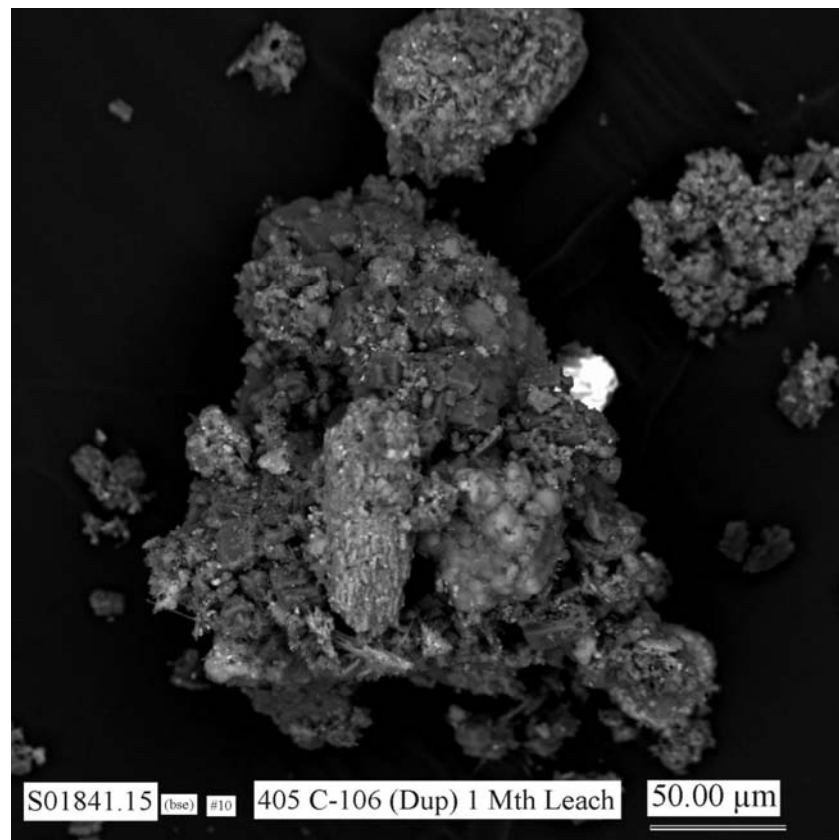
Figure C.43. Micrograph Showing Morphologies of Typical Particles in SEM Mount 10 of the 1-Month Water-Leached Sludge Sample 405
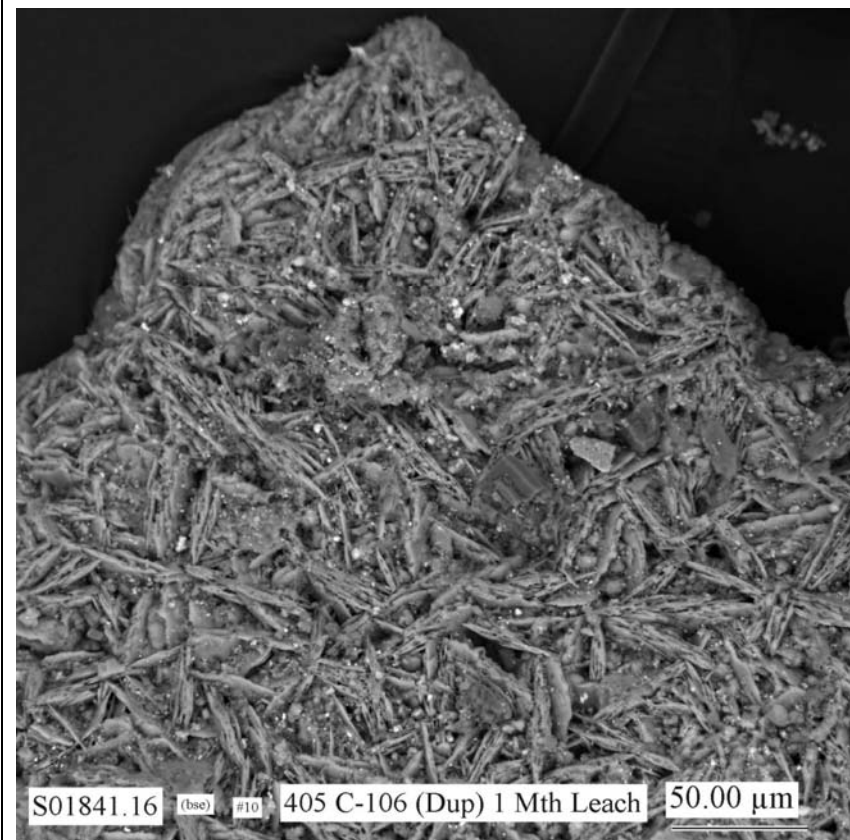
Figure C.44. Micrograph Showing Morphologies of Typical Particles in SEM Mount 10 of the 1-Month Water-Leached Sludge Sample 405

C-106 405 (#10) Duplicate 1-month Leached eds01 [See Figure C.32]	C-106 405 (#10) Duplicate 1-month Leached eds02 [See Figure C.32]
C-106 405 (#10) Duplicate 1-month Leached eds03 [See Figure C.32]	C-106 405 (#10) Duplicate 1-month Leached eds04 [See Figure C.32]

Figure C.45.   EDS Spectra for Analyses eds01 Through eds04 for Particles in SEM Mount 10 of the 1-Month Water-Leached Sludge Sample 405

C-106 405 (#10) Duplicate 1-month Leached eds05 [See Figure C.36]	C-106 405 (#10) Duplicate 1-month Leached eds06 [See Figure C.37]
C-106 405 (#10) Duplicate 1-month Leached eds07 [See Figure C.38]	C-106 405 (#10) Duplicate 1-month Leached eds08 [See Figure C.38]

Figure C.46.   EDS Spectra for Analyses eds05 Through eds08 for Particles in SEM Mount 10 of the 1-Month Water-Leached Sludge Sample 405

C-106 405 (#10) Duplicate 1-month Leached eds09 [See Figure C.38]	C-106 405 (#10) Duplicate 1-month Leached eds10 [See Figure C.38]
C-106 405 (#10) Duplicate 1-month Leached eds11 [See Figure C.39]	C-106 405 (#10) Duplicate 1-month Leached eds12 [See Figure C.40]

Figure C.47.   EDS Spectra for Analyses eds09 Through eds12 for Particles in SEM Mount 10 of the 1-Month Water-Leached Sludge Sample 405

C-106 405 (#10) Duplicate 1-month Leached eds13 [See Figure C.41]	C-106 405 (#10) Duplicate 1-month Leached eds14 [See Figure C.42]
C-106 405 (#10) Duplicate 1-month Leached eds15 [See Figure C.42]	

Figure C.48.   EDS Spectra for Analyses eds13 Through eds15 for Particles in SEM Mount 10 of the 1-Month Water-Leached Sludge Sample 405

Figure C.49. Micrograph Showing Morphologies of Typical Particles in SEM Mount 11 of the 1-Month Water-Leached Sludge Sample 405
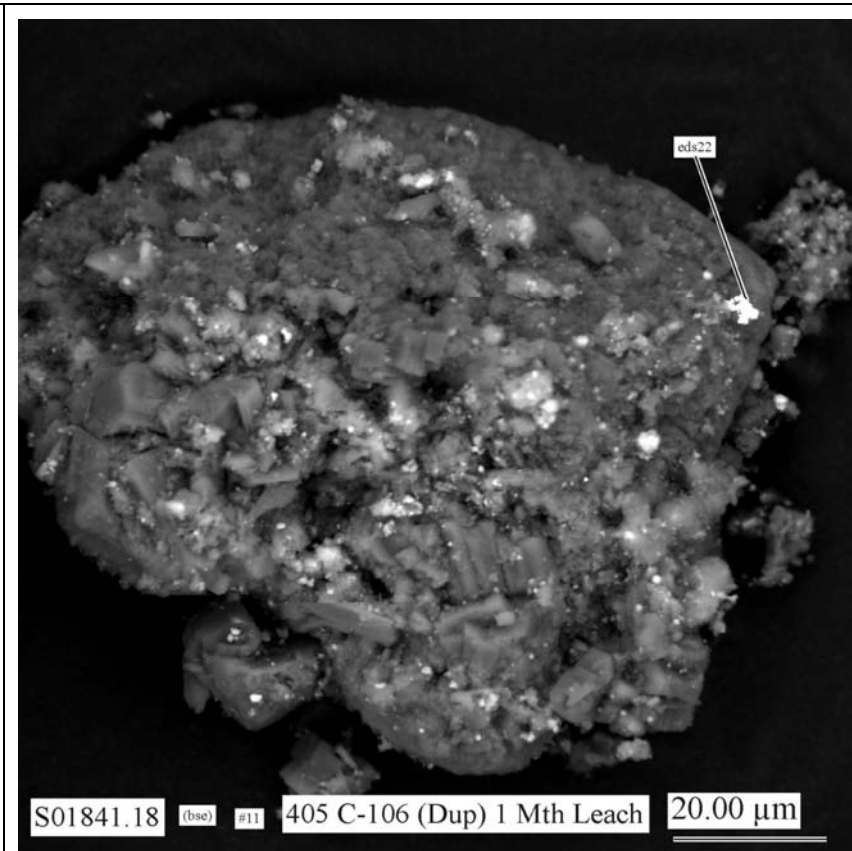
Figure C.50. Micrograph Showing Morphologies of Typical Particles in SEM Mount 11 of the 1-Month Water-Leached Sludge Sample 405
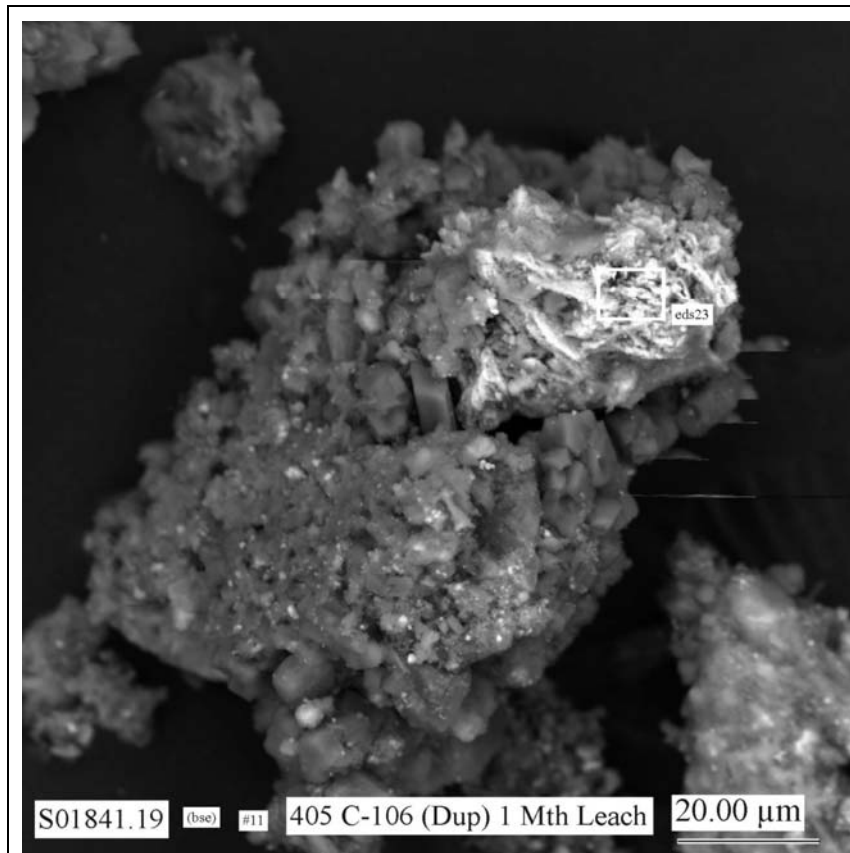
Figure C.51. Micrograph Showing Morphologies of Typical Particles in SEM Mount 11 of the 1-Month Water-Leached Sludge Sample 405
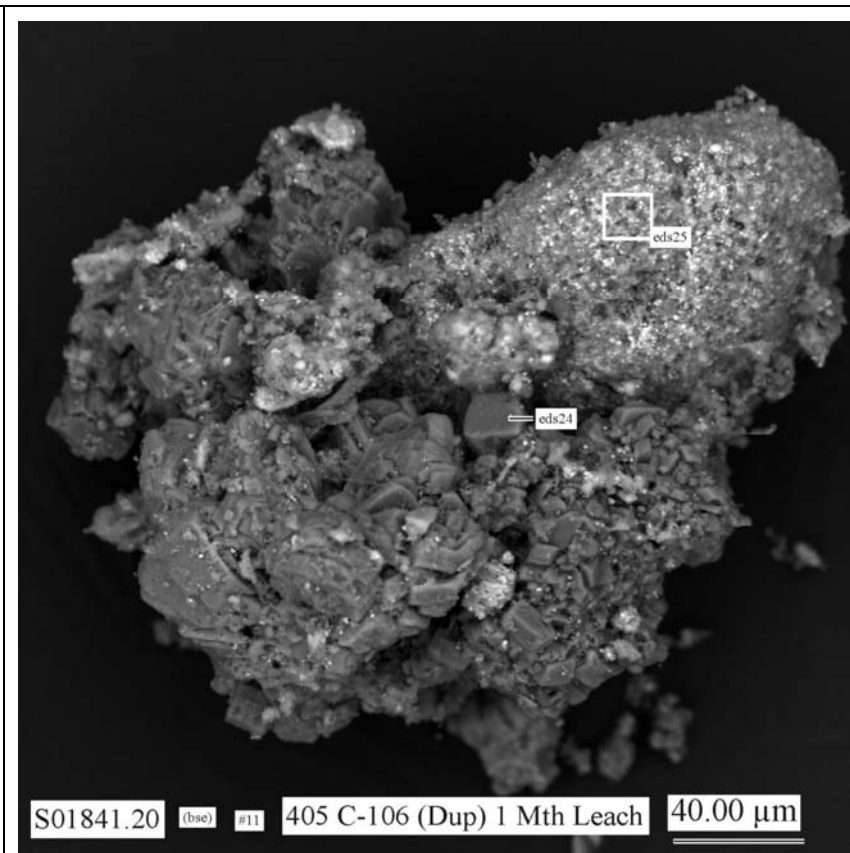
Figure C.52. Micrograph Showing Morphologies of Typical Particles in SEM Mount 11 of the 1-Month Water-Leached Sludge Sample 405

S01841.21 (bse) #11 405 C-106 (Dup) 1 Mth Leach 10.00 μm
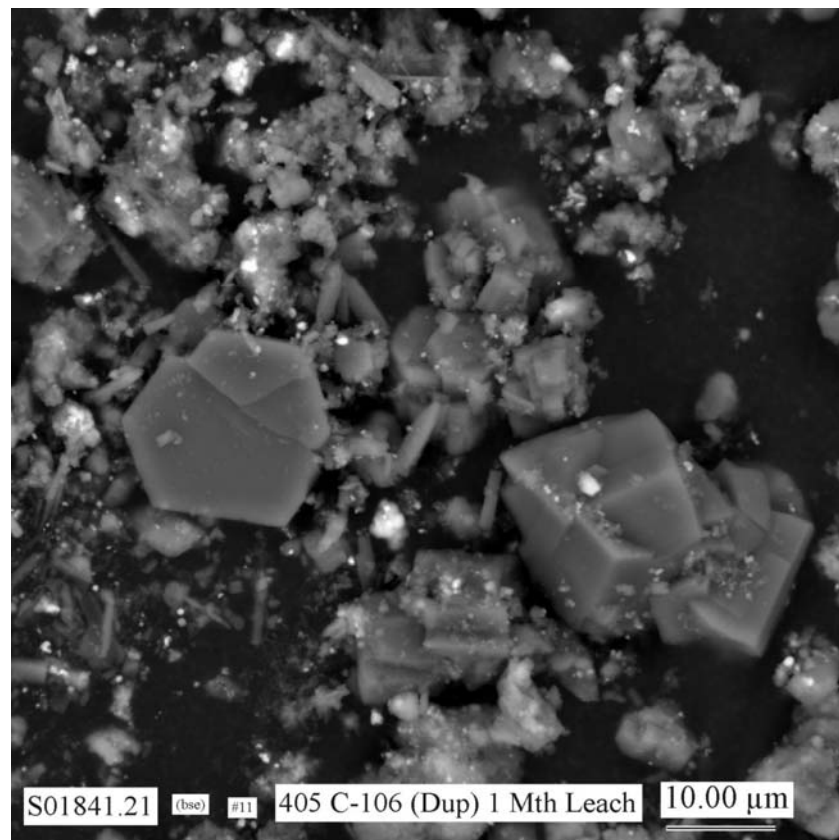
Figure C.53. Micrograph Showing Morphologies of Typical Particles in SEM Mount 11 of the 1-Month Water-Leached Sludge Sample 405

S01841.24 (bse) #11 405 C-106 (Dup) 1 Mth Leach 10.00 μm
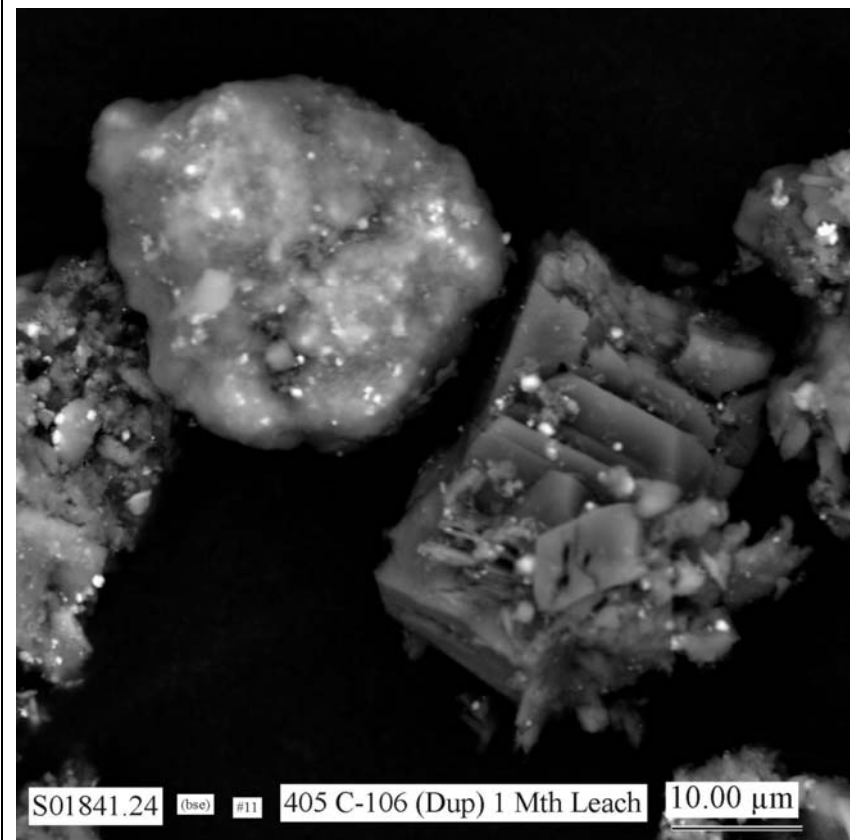
Figure C.54. Micrograph Showing Morphologies of Typical Particles in SEM Mount 11 of the 1-Month Water-Leached Sludge Sample 405

| C-106 405 (#11) Duplicate 1-month Leached eds21 [See Figure C.49] | C-106 405 (#11) Duplicate 1-month Leached eds22 [See Figure C.50] |
| C-106 405 (#11) Duplicate 1-month Leached eds23 [See Figure C.51] | C-106 405 (#11) Duplicate 1-month Leached eds24 [See Figure C.52] |

Figure C.55.   EDS Spectra for Analyses eds21 Through eds24 for Particles in SEM Mount 11 of the 1-Month Water-Leached Sludge Sample 405

cps
C-106 405 (#11) Duplicate 1-month Leached eds25 [See Figure C.52]
Figure C.56.   EDS Spectra for Analyses eds25 for a Particles in SEM Mount 11 of the 1-Month Water-Leached Sludge Sample 405

C.3 Water-Leached (82-Day) Sample 404

One mount of sample from the 82-day water-leached residual sludge from tank C-106 (primary 404) was analyzed by SEM/EDS. The SEM micrographs for this sample are shown in Figures C.57 through C.72, and the EDS spectra for this mount are given in Figures C.73 through C.76.
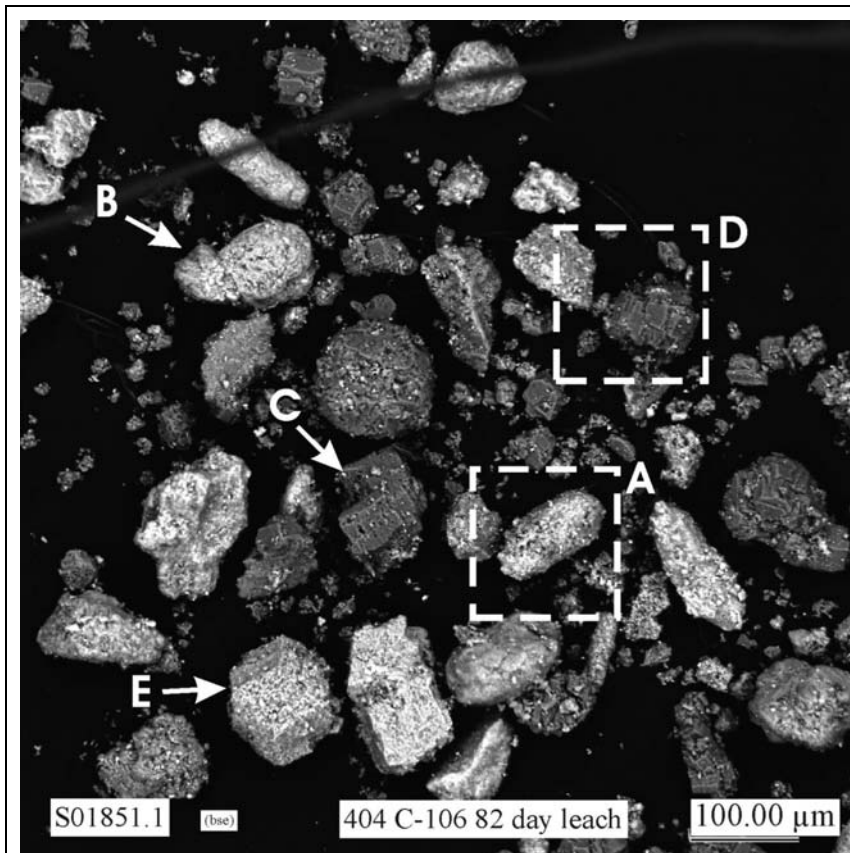
Figure C.57. Low Magnification SEM Micrograph Showing General Morphologies of Particles in Sample 404 of 82-Day Water-Leached Sludge
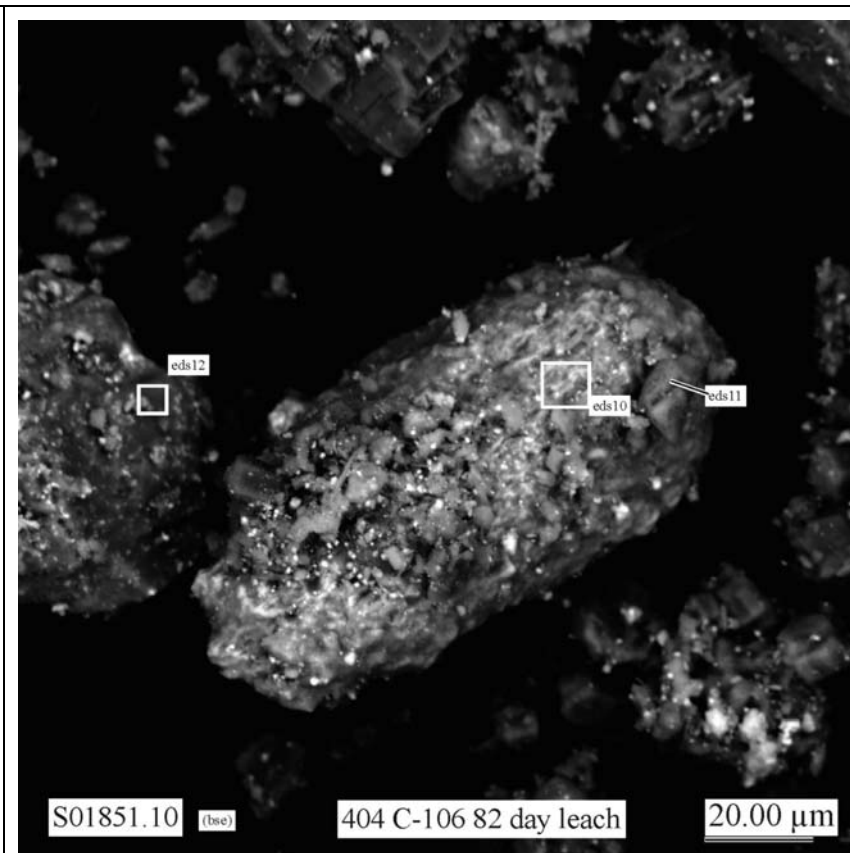
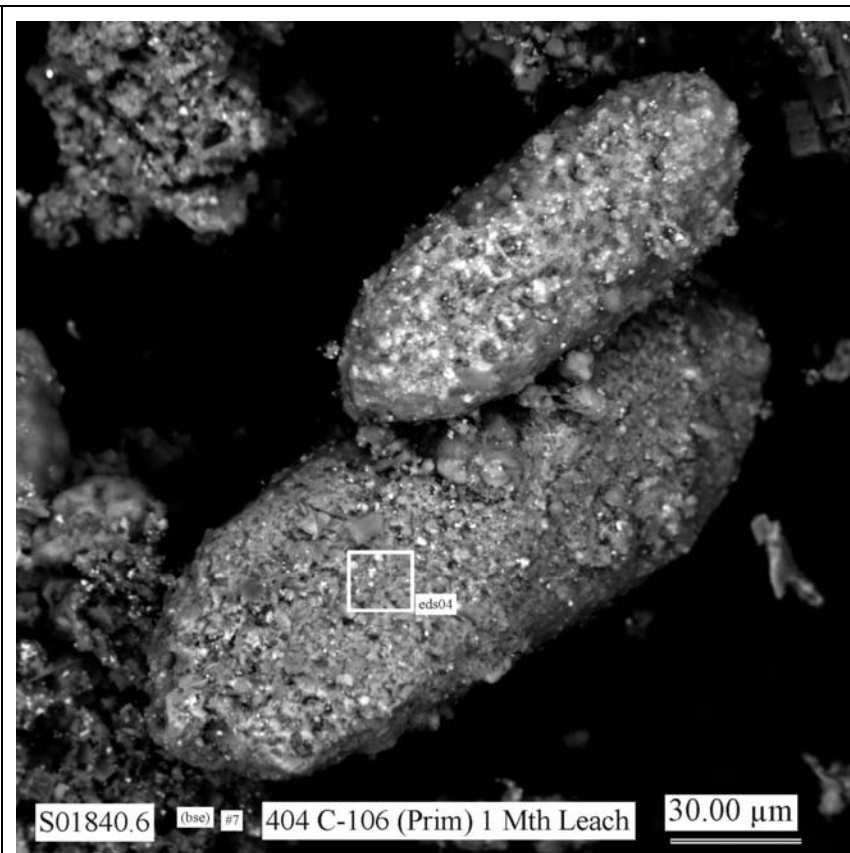**Figure C.58.** Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled A in Figure C.57
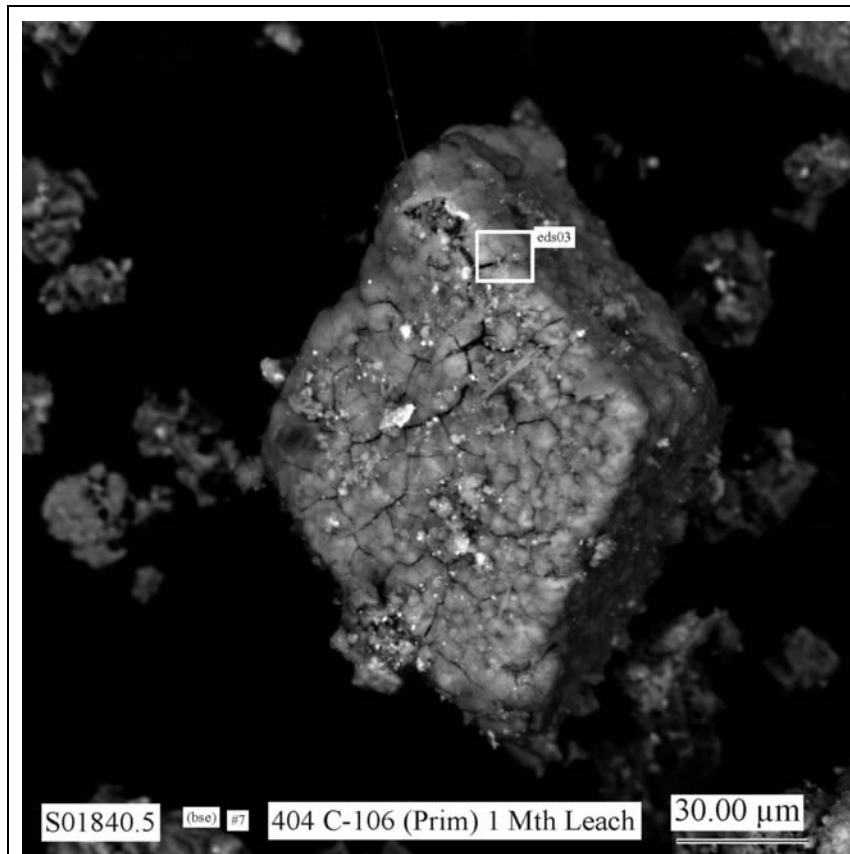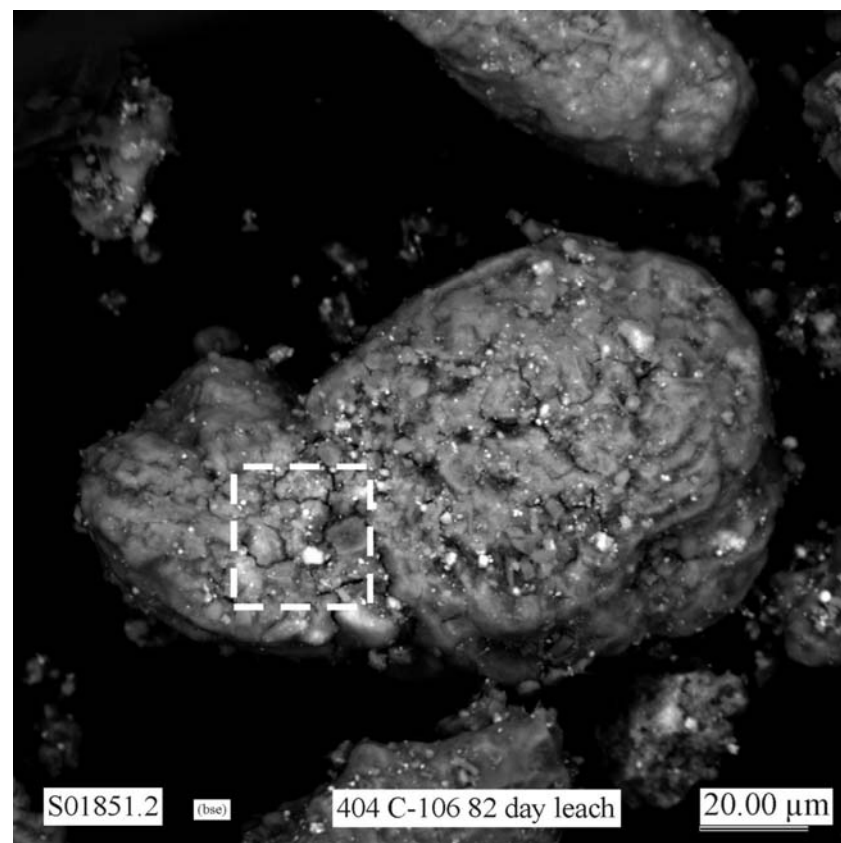
Figure C.59. Micrograph Showing at Higher Magnification the Particle Labeled B in Figure C.57
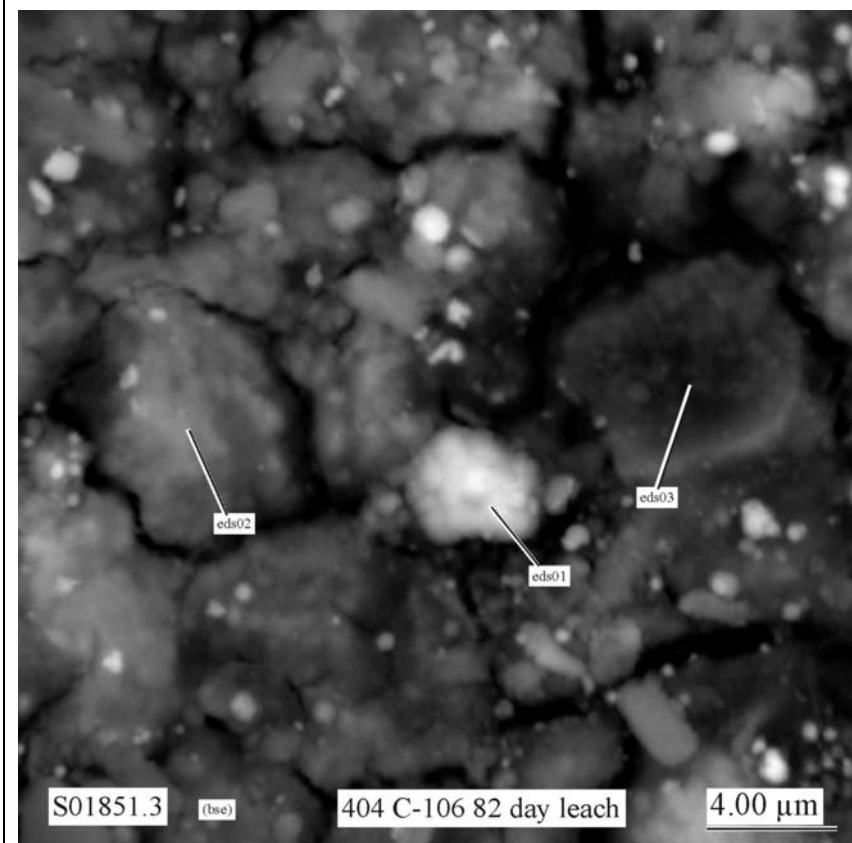
Figure C.60. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square in Figure C.59
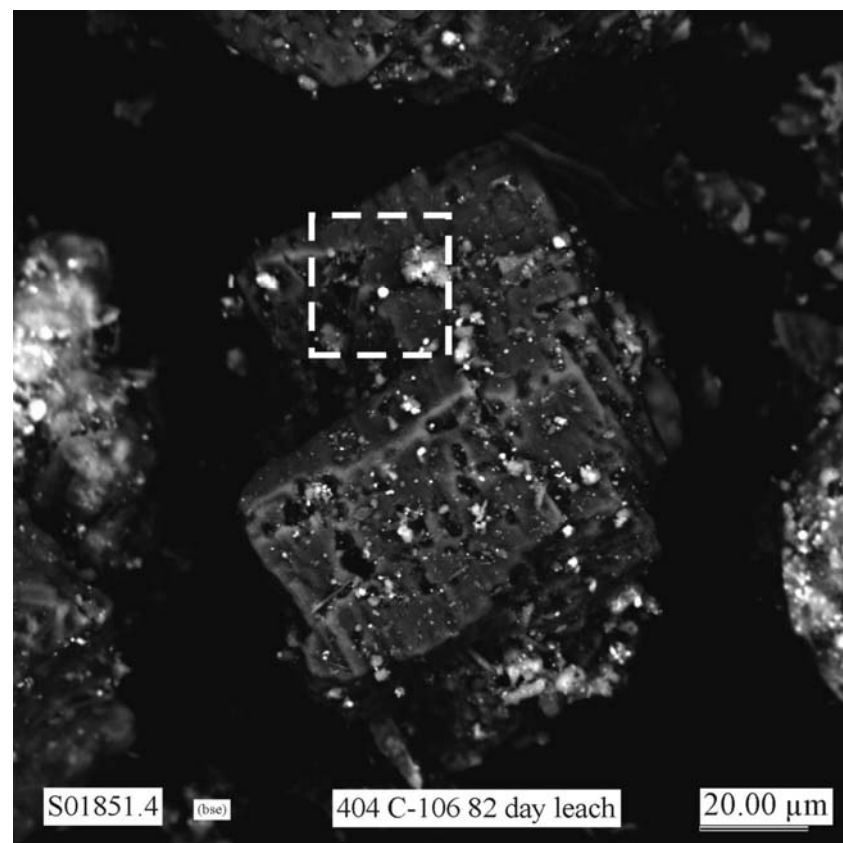
Figure C.61. Micrograph Showing at Higher Magnification the Particle Labeled C in Figure C.57
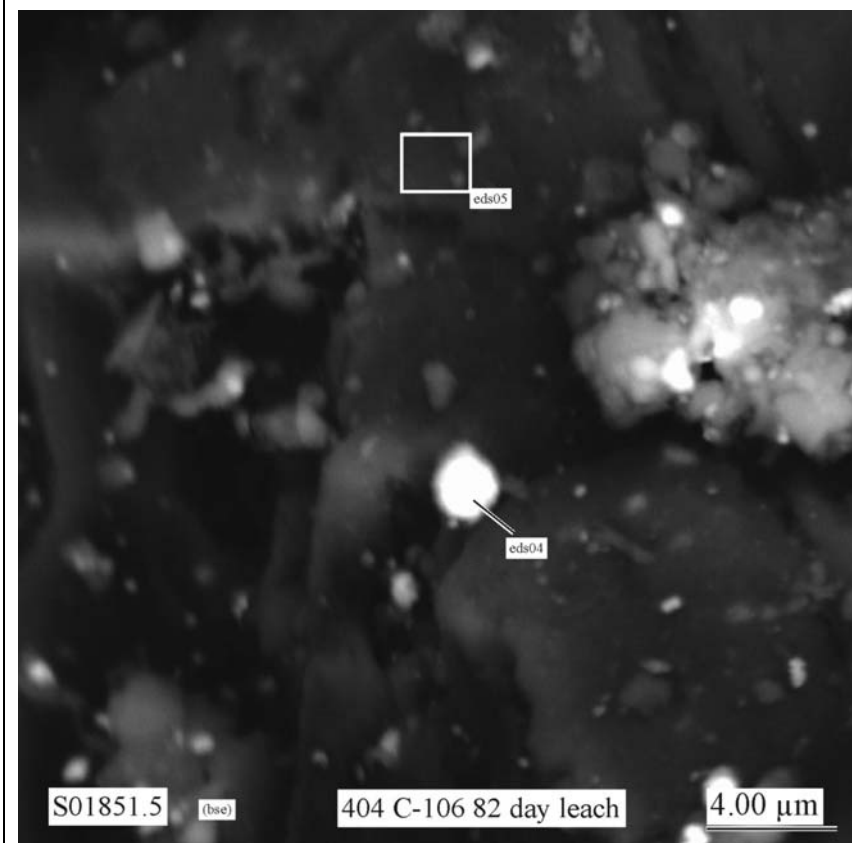
Figure C.62. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square in Figure C.61
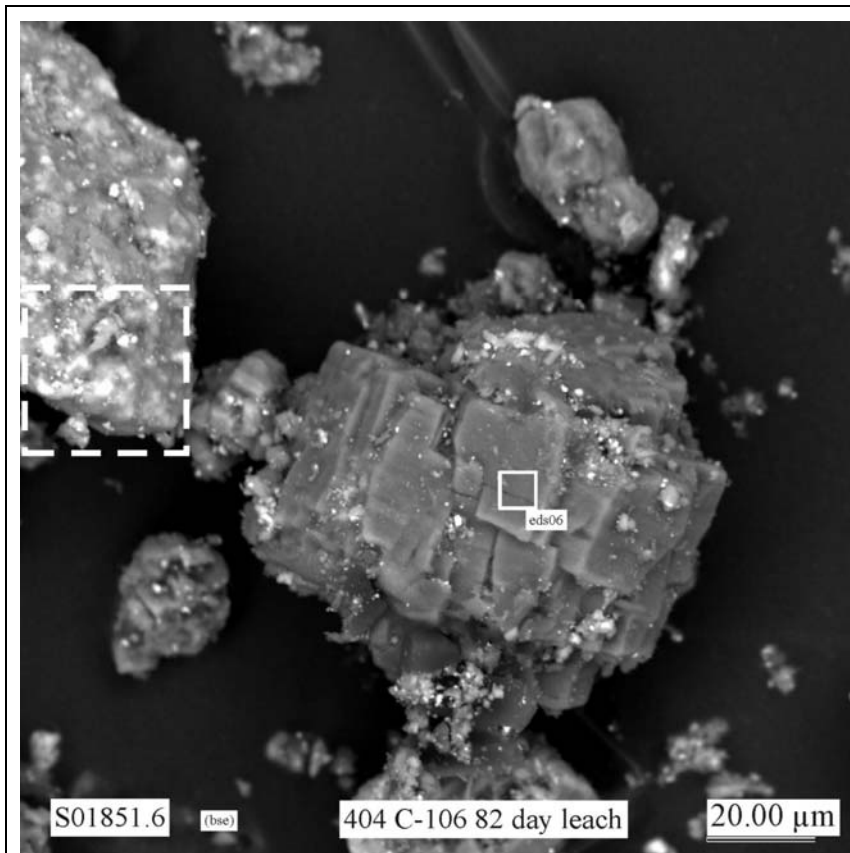
Figure C.63. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled D in Figure C.57
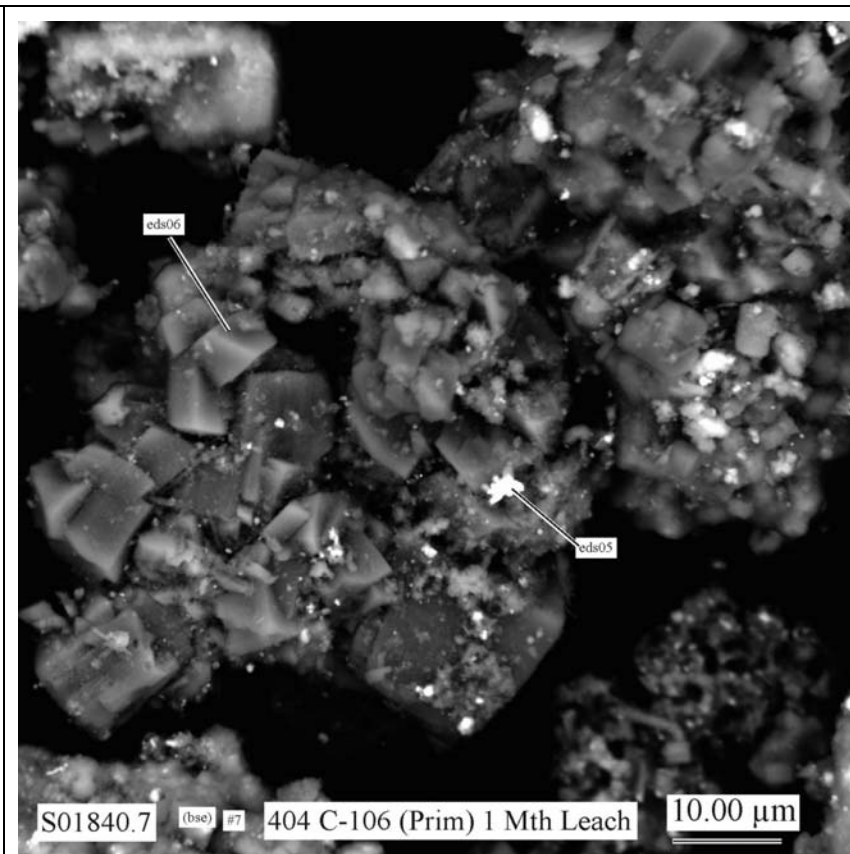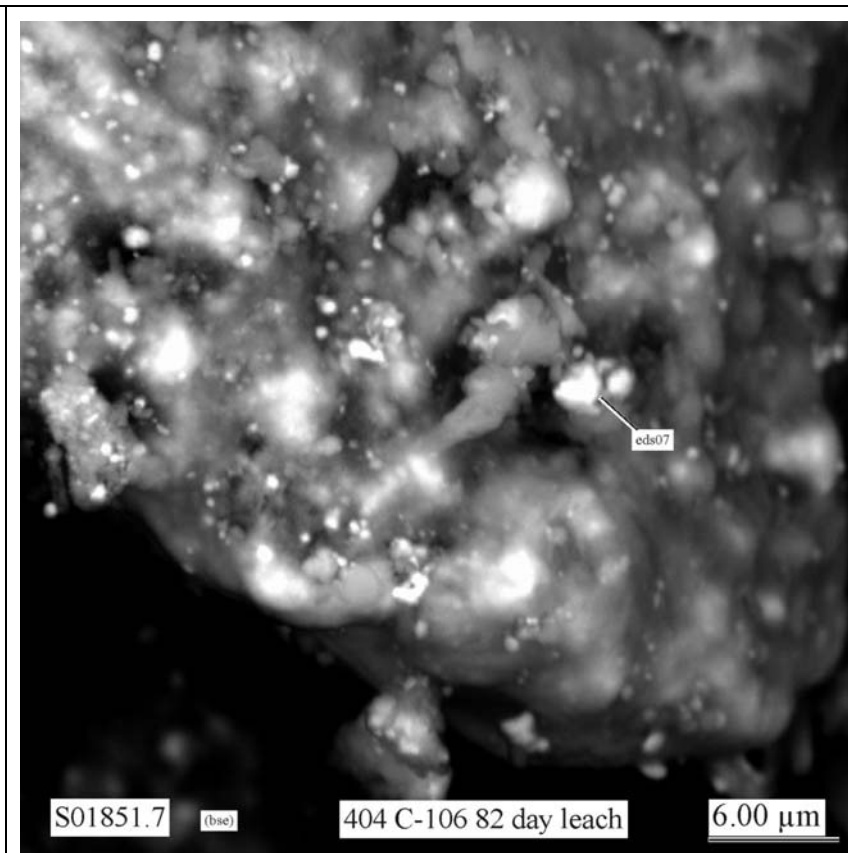

Figure C.64. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square in Figure C.63
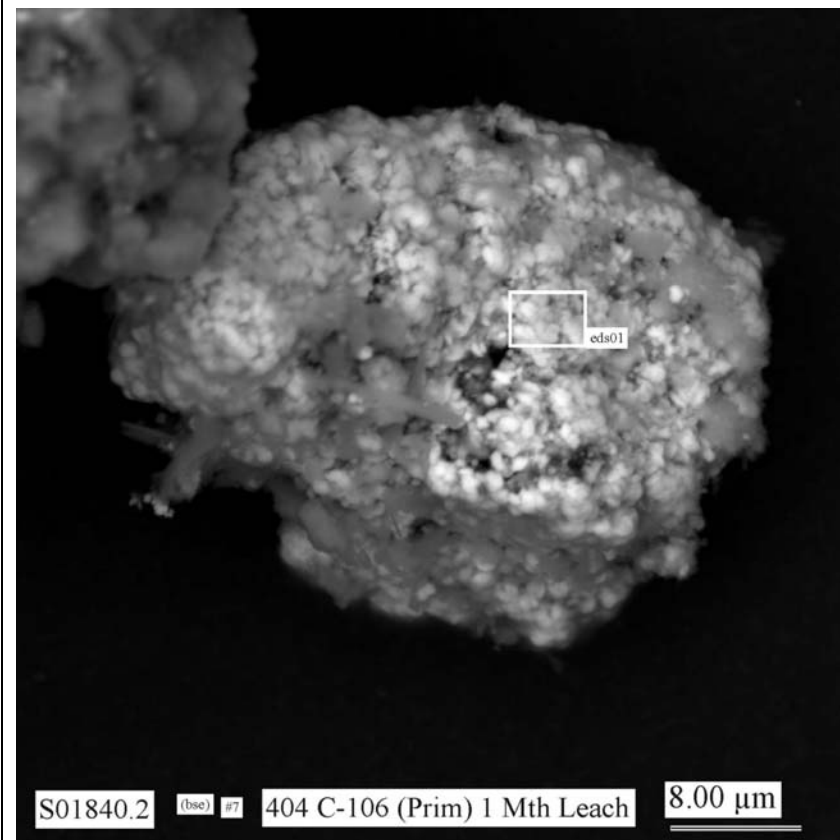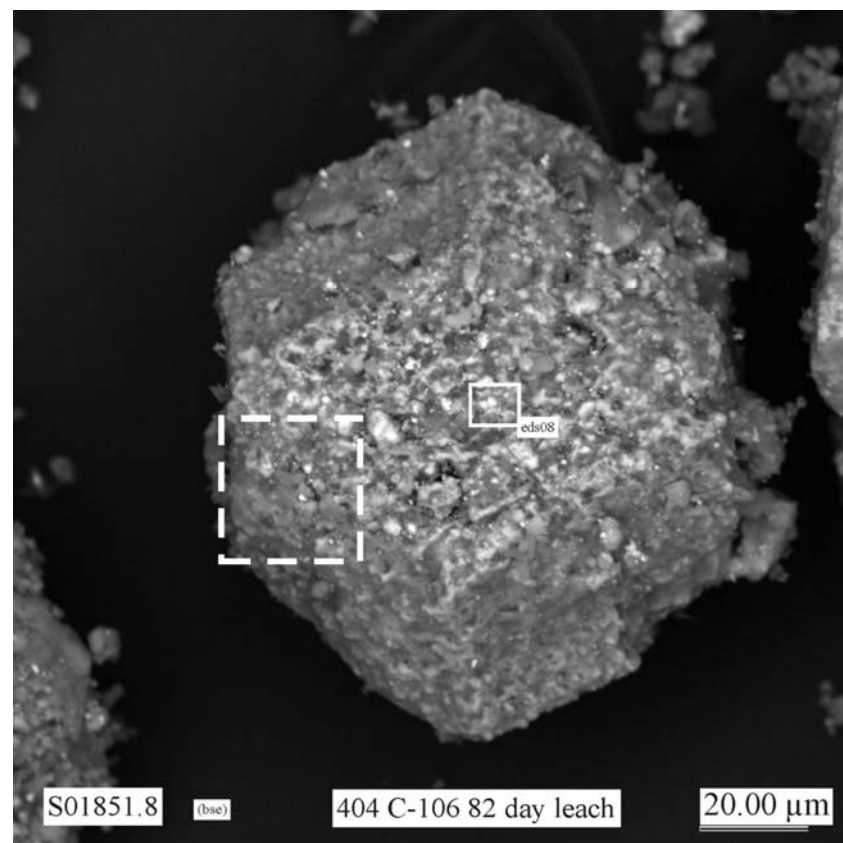
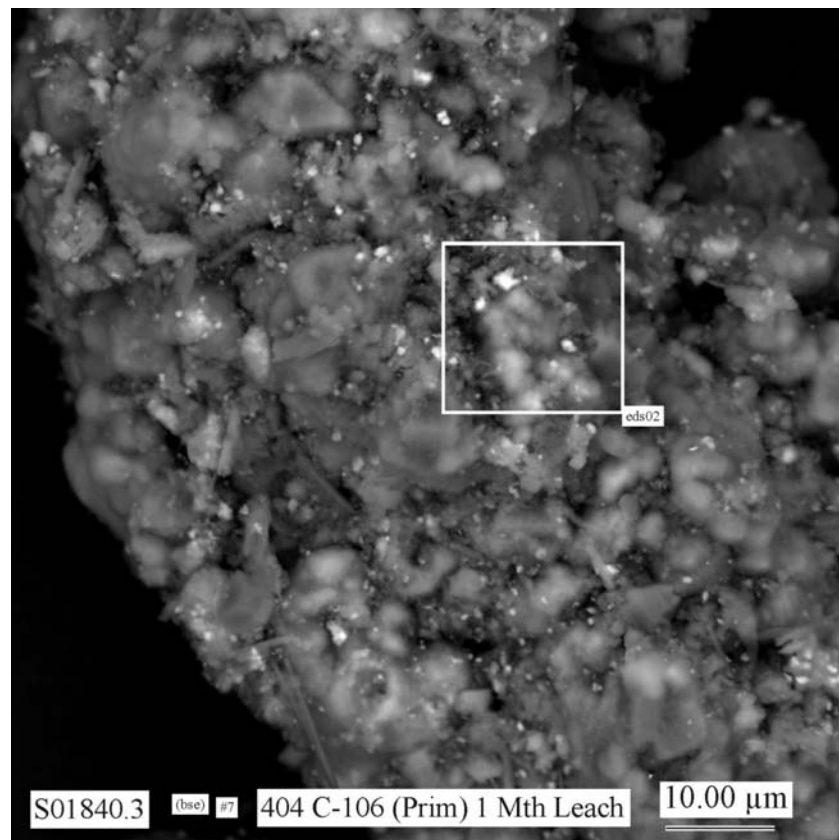**Figure C.65.** Micrograph Showing at Higher Magnification the Particle Labeled E in Figure C.57
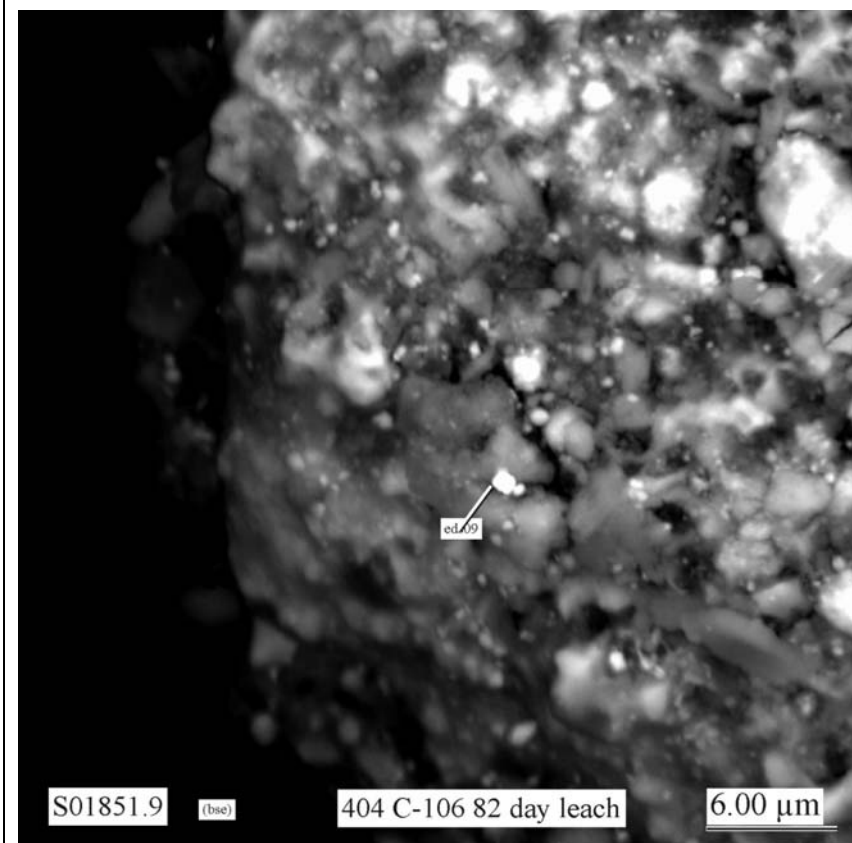
Figure C.66. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square in Figure C.65
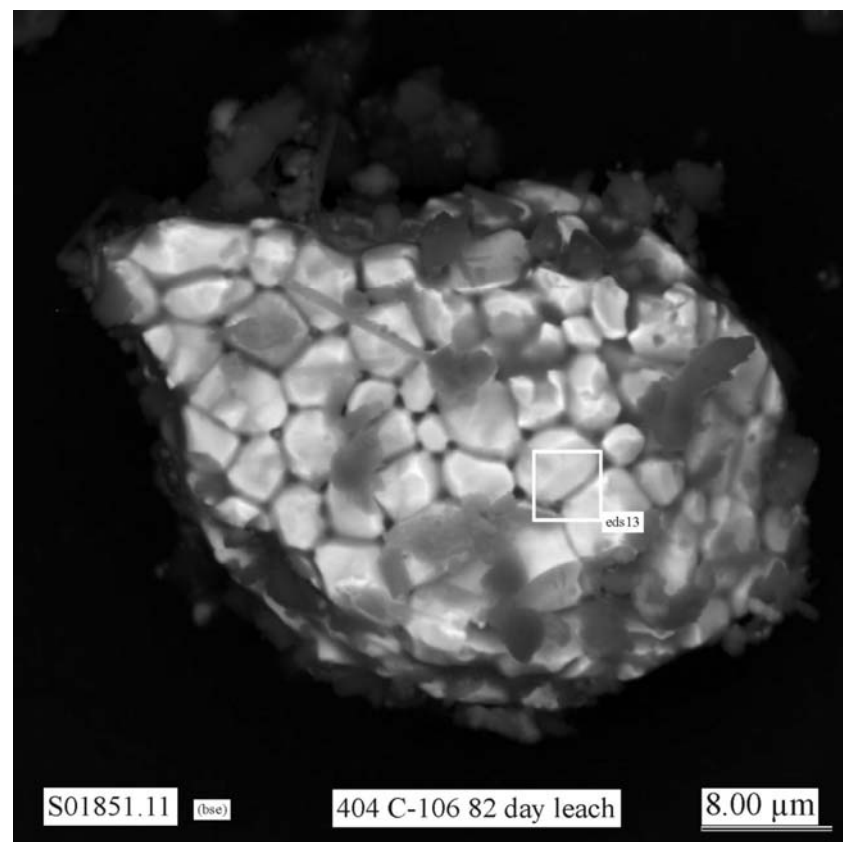
Figure C.67. Micrograph Showing Morphology of an Unusual Aggregate of Th-rich (possibly metallic) Particles in Sample 404 of 82-Day Water-Leached Sludge
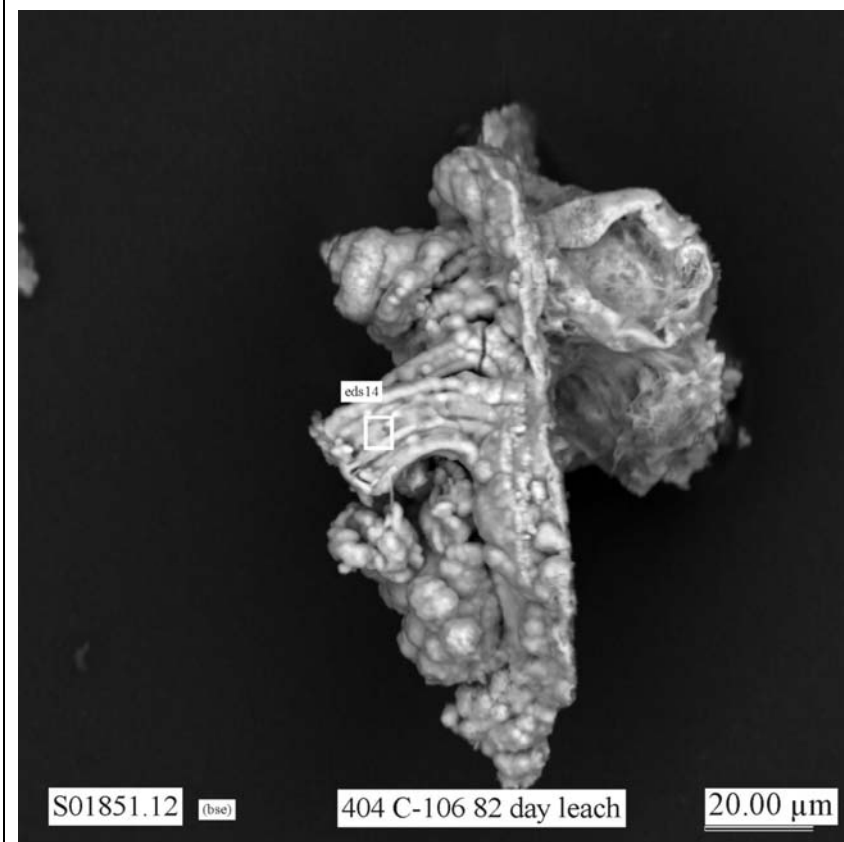
Figure C.68. Micrograph Showing Morphology of an Unusual Fe-Rich Particle in Sample 404 of 82-Day Water-Leached Sludge
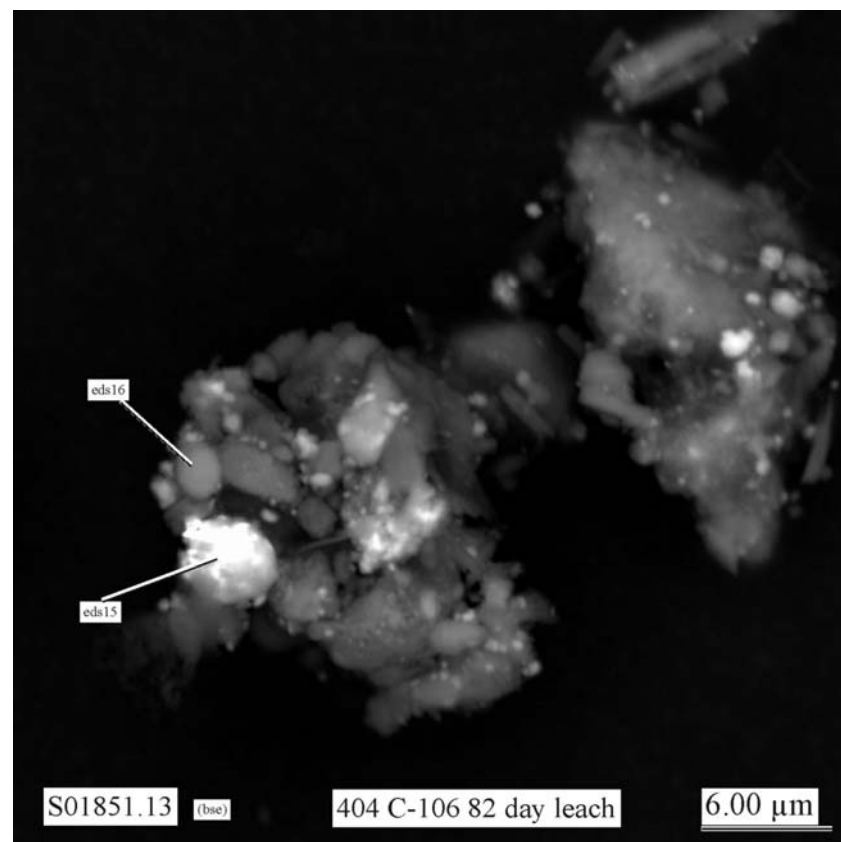
Figure C.69. Micrograph Showing Morphologies of Typical Particles in Sample 404 of 82-Day Water-Leached Sludge
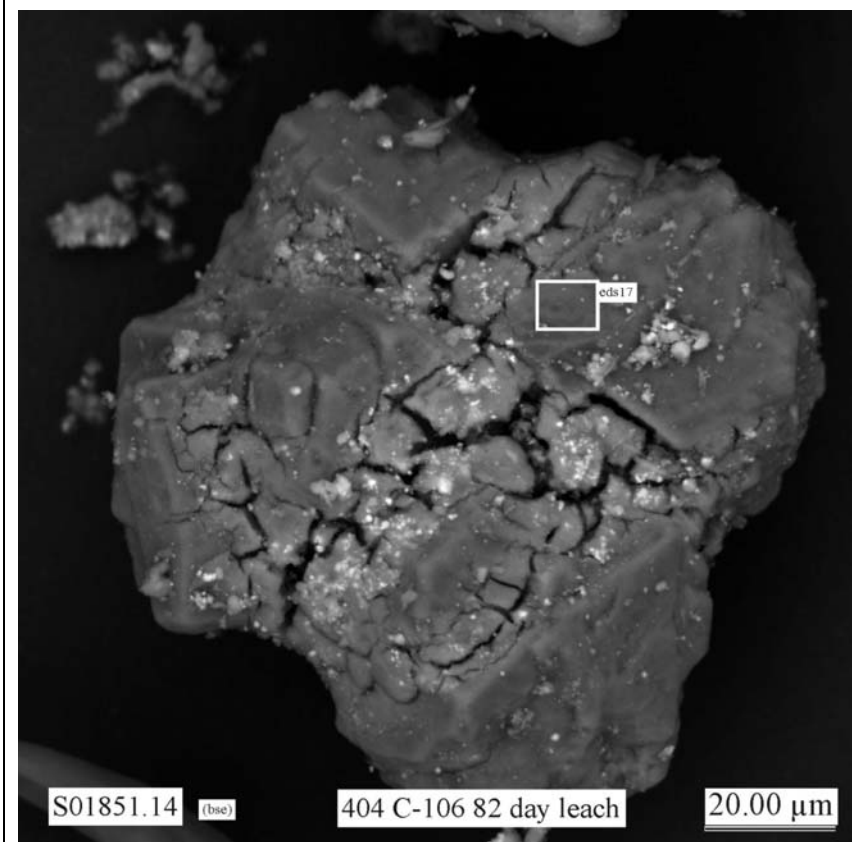
Figure C.70. Micrograph Showing Morphologies of Typical Particles in Sample 404 of 82-Day Water-Leached Sludge
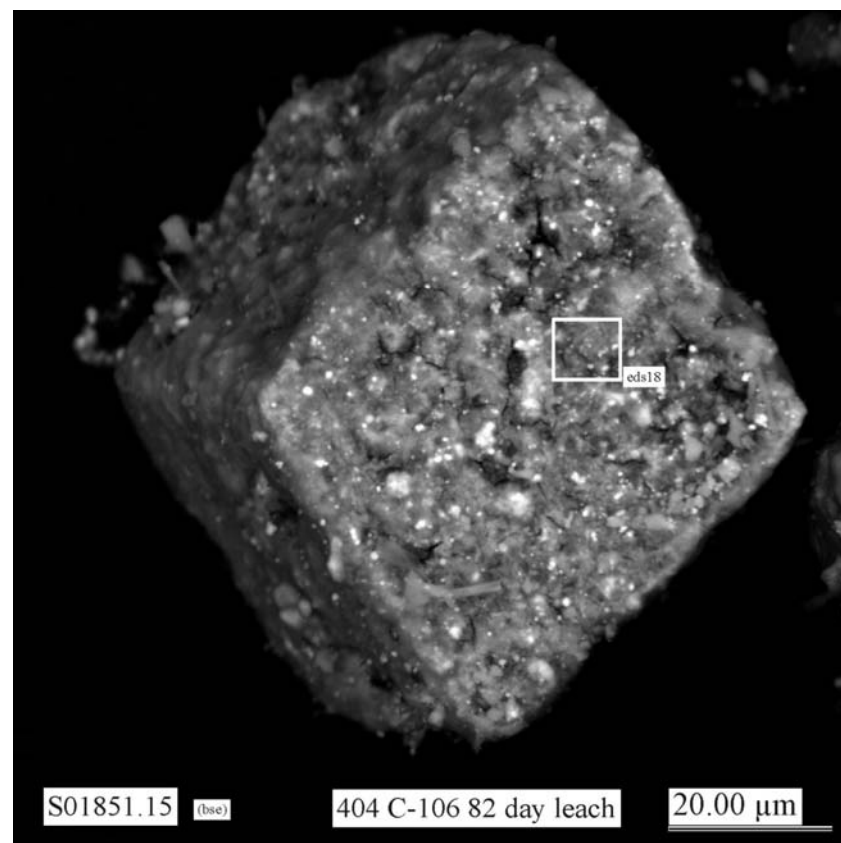
Figure C.71. Micrograph Showing Morphology a Common Particle in Sample 404 of 82-Day Water-Leached Sludge
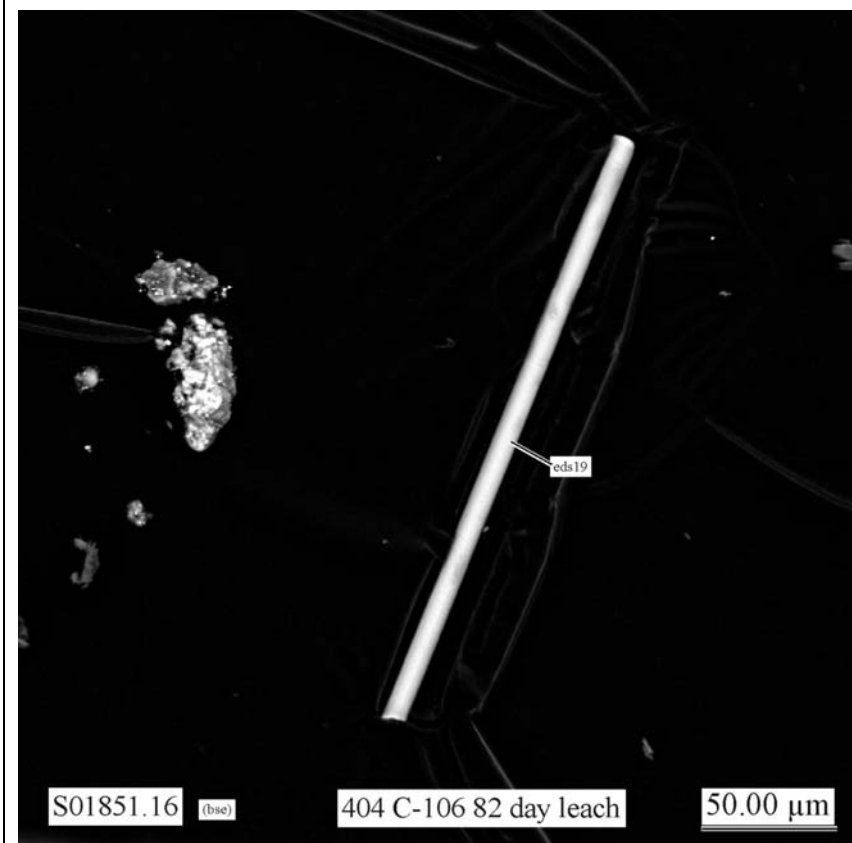
Figure C.72. Micrograph Showing a Rod-Like Particle in Sample 404 of 82-Day Water-Leached Sludge

C-106 404 Primary 82-day Leached
eds01 [See Figure C.60]

C-106 404 Primary 82-day Leached
eds02 [See Figure C.60]

C-106 404 Primary 82-day Leached
eds03 [See Figure C.60]

C-106 404 Primary 82-day Leached
eds04 [See Figure C.62]
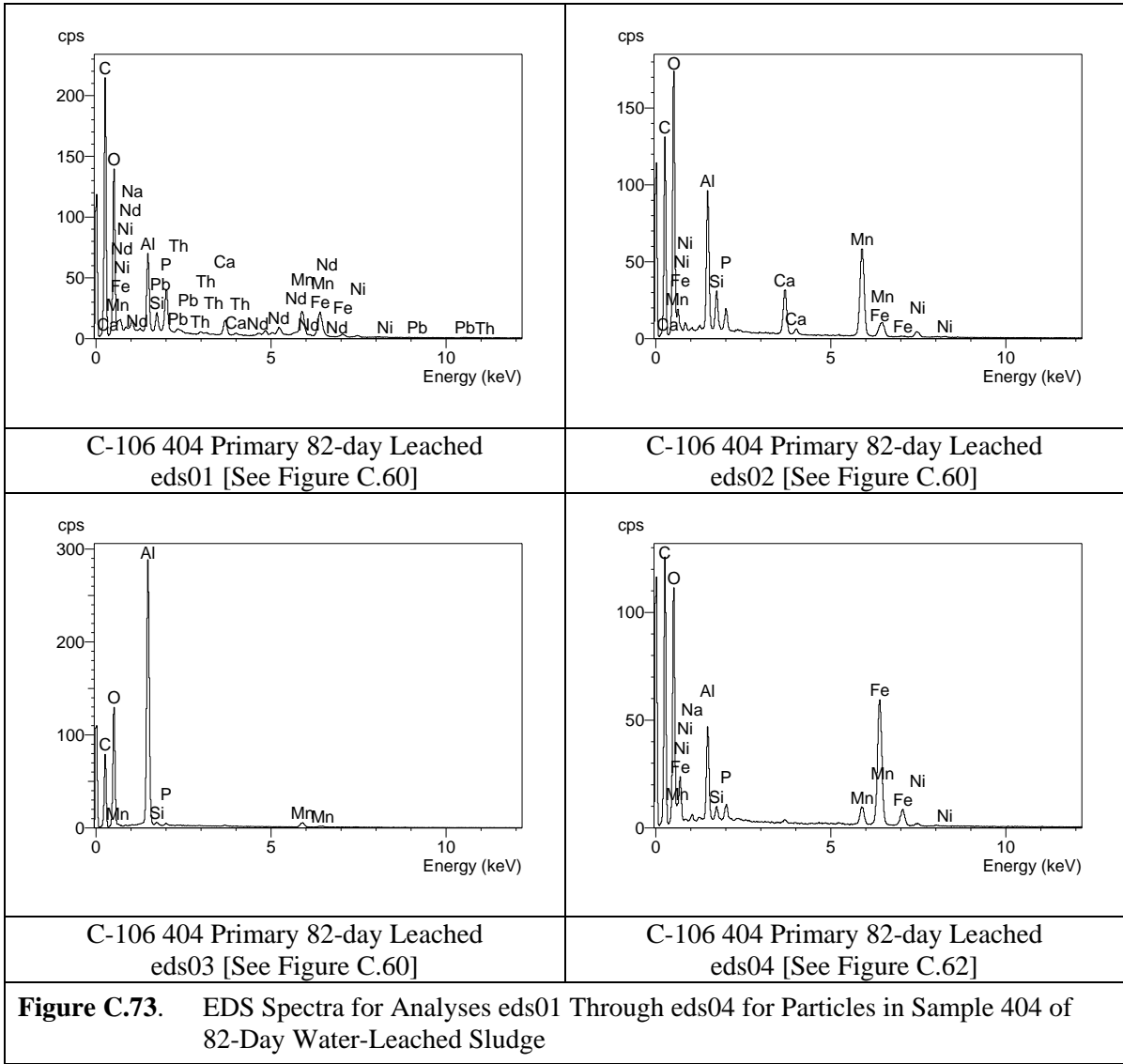
Figure C.73.   EDS Spectra for Analyses eds01 Through eds04 for Particles in Sample 404 of
82-Day Water-Leached Sludge

C-106 404 Primary 82-day Leached eds05 [See Figure C.62]	C-106 404 Primary 82-day Leached eds06 [See Figure C.63]
C-106 404Primary 82-day Leached eds07 [See Figure C.64]	C-106 404 Primary 82-day Leached eds08 [See Figure C.65]

Figure C.74.   EDS Spectra for Analyses eds05 Through eds08 for Particles in Sample 404 of 82-Day Water-Leached Sludge

C-106 404 Primary 82-day Leached
eds09 [See Figure C.66]

C-106 404 Primary 82-day Leached
eds10 [See Figure C.58]

C-106 404 Primary 82-day Leached
eds11 [See Figure C.58]

C-106 404 Primary 82-day Leached
eds12 [See Figure C.58]

Figure C.75.   EDS Spectra for Analyses eds09 Through eds12 for Particles in Sample 404 of 82-Day Water-Leached Sludge

| C-106 404 Primary 82-day Leached eds13 [See Figure C.67] | C-106 404 Primary 82-day Leached eds14 [See Figure C.68] |
| C-106 404 Primary 82-day Leached eds15 [See Figure C.69] | C-106 404 Primary 82-day Leached eds16 [See Figure C.69] |

Figure C.76.   EDS Spectra for Analyses eds13 Through eds16 for Particles in Sample 404 of 82-Day Water-Leached Sludge

C-106 404 Primary 82-day Leached eds17 [See Figure C.70]	C-106 404 Primary 82-day Leached eds18 [See Figure C.71]
C-106 404 Primary 82-day Leached eds19 [See Figure C.72]	

Figure C.77.   EDS Spectra for Analyses eds17 Through eds19 for Particles in Sample 404 of 82-Day Water-Leached Sludge

C.4 Water-Leached (82-Day) Sample 405

One mount of sample from the 82-day water-leached residual sludge from tank C-106 (primary 405) was analyzed by SEM/EDS. The SEM micrographs for this sample are shown in Figures C.78 through C.93, and the EDS spectra for this mount are given in Figures C.94 through C.98.

C.51
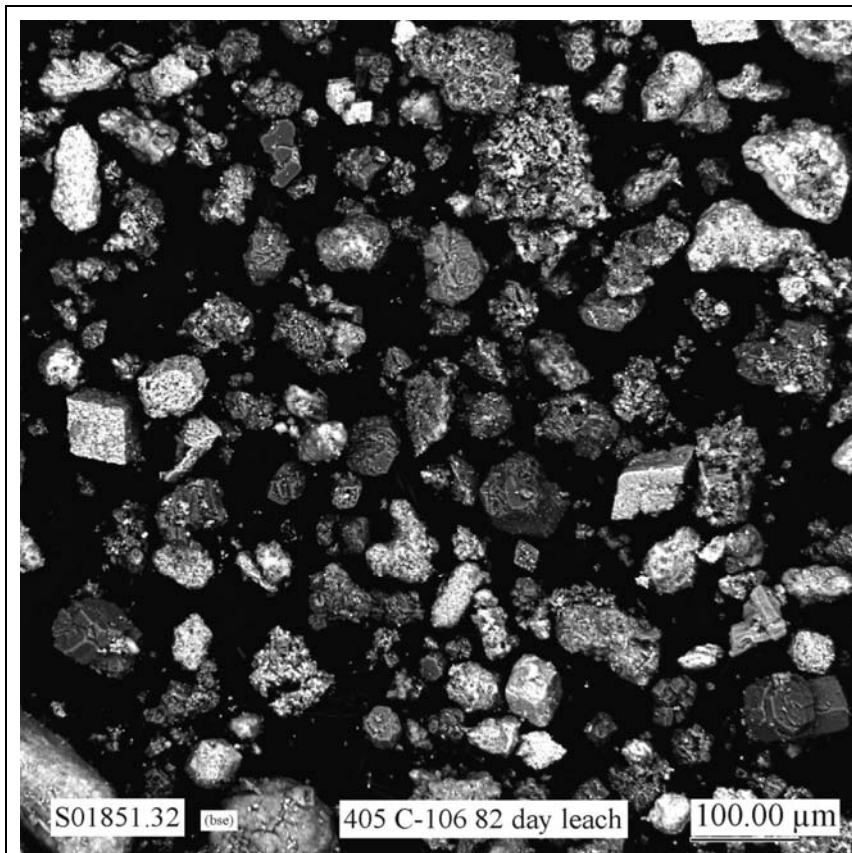
Figure C.78. Low Magnification SEM Micrograph Showing General Morphologies of Particles in Sample 405 of 82-Day Water-Leached Sludge
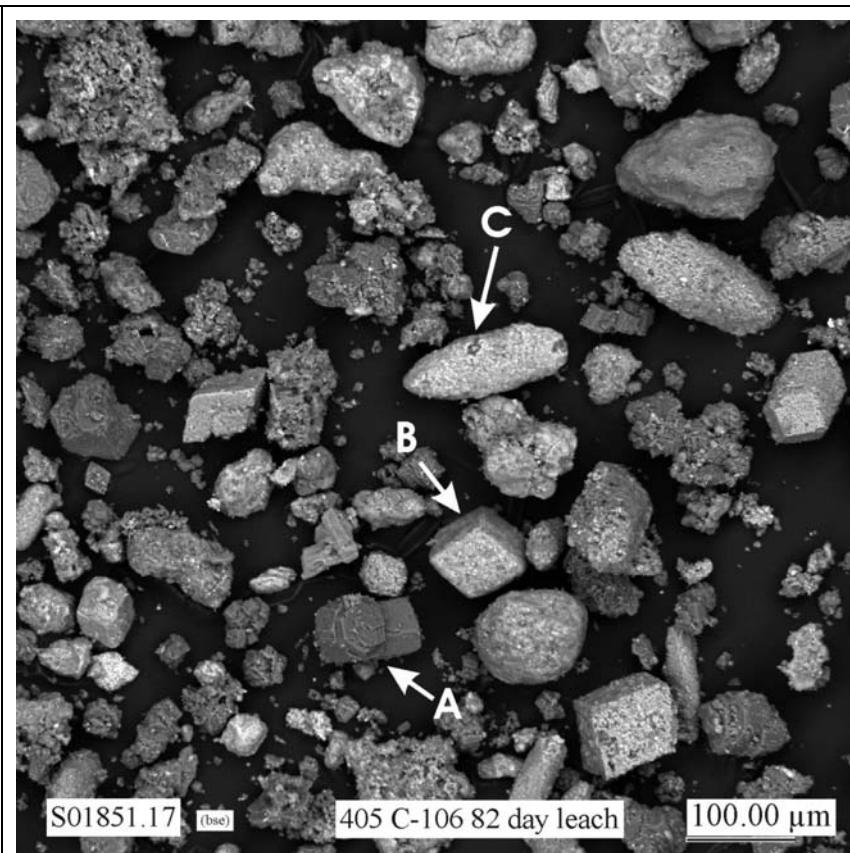
Figure C.79 Low Magnification SEM Micrograph Showing General Morphologies of Particles in Sample 405 of 82-Day Water-Leached Sludge
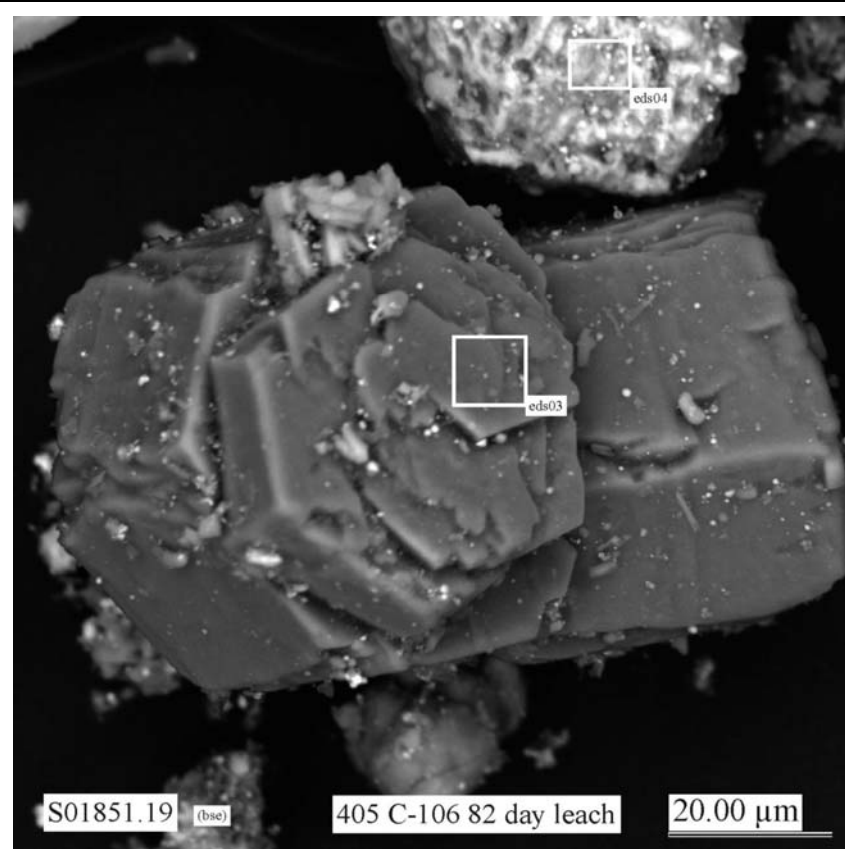
Figure C.80. Micrograph Showing at Higher Magnification the Particle Labeled A in Figure C.79
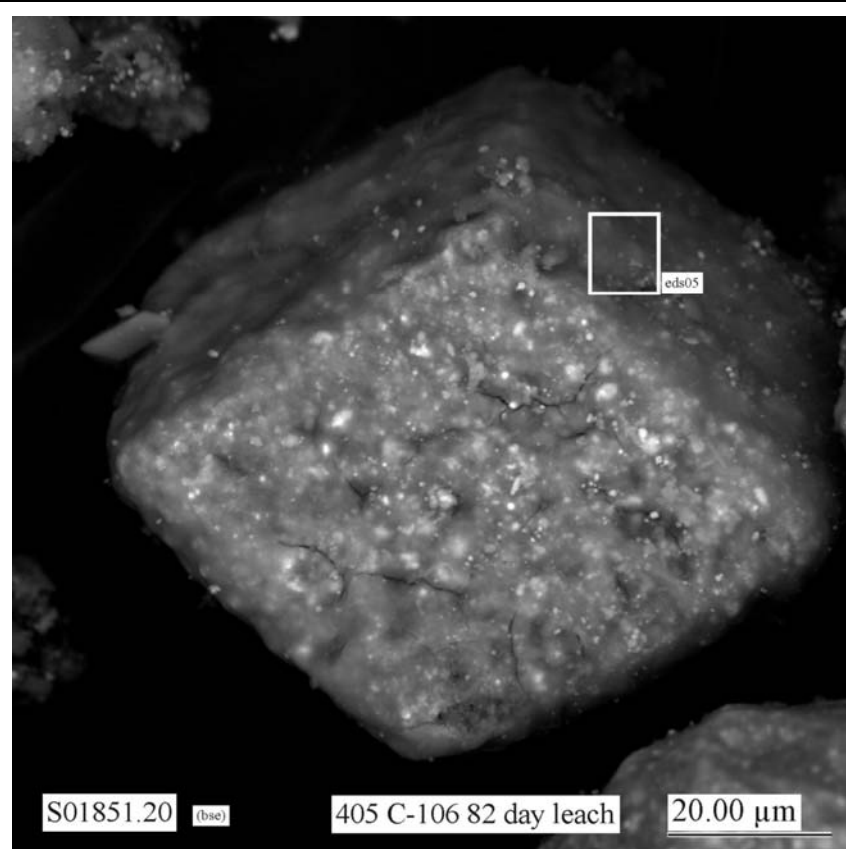
Figure C.81. Micrograph Showing at Higher Magnification the Particle Labeled B in Figure C.79
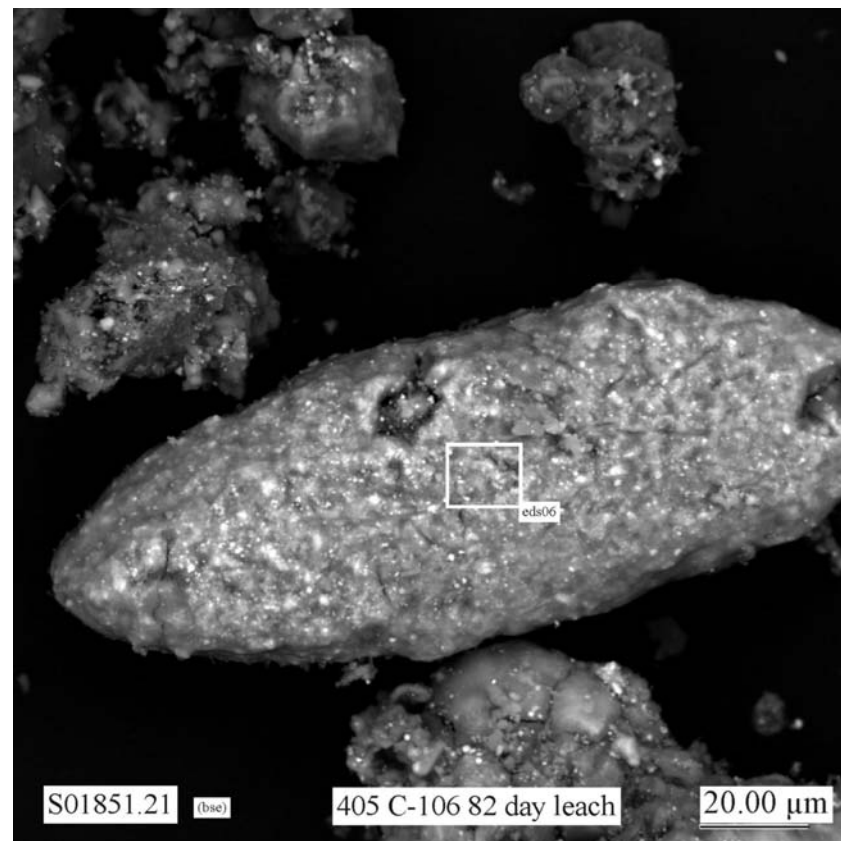
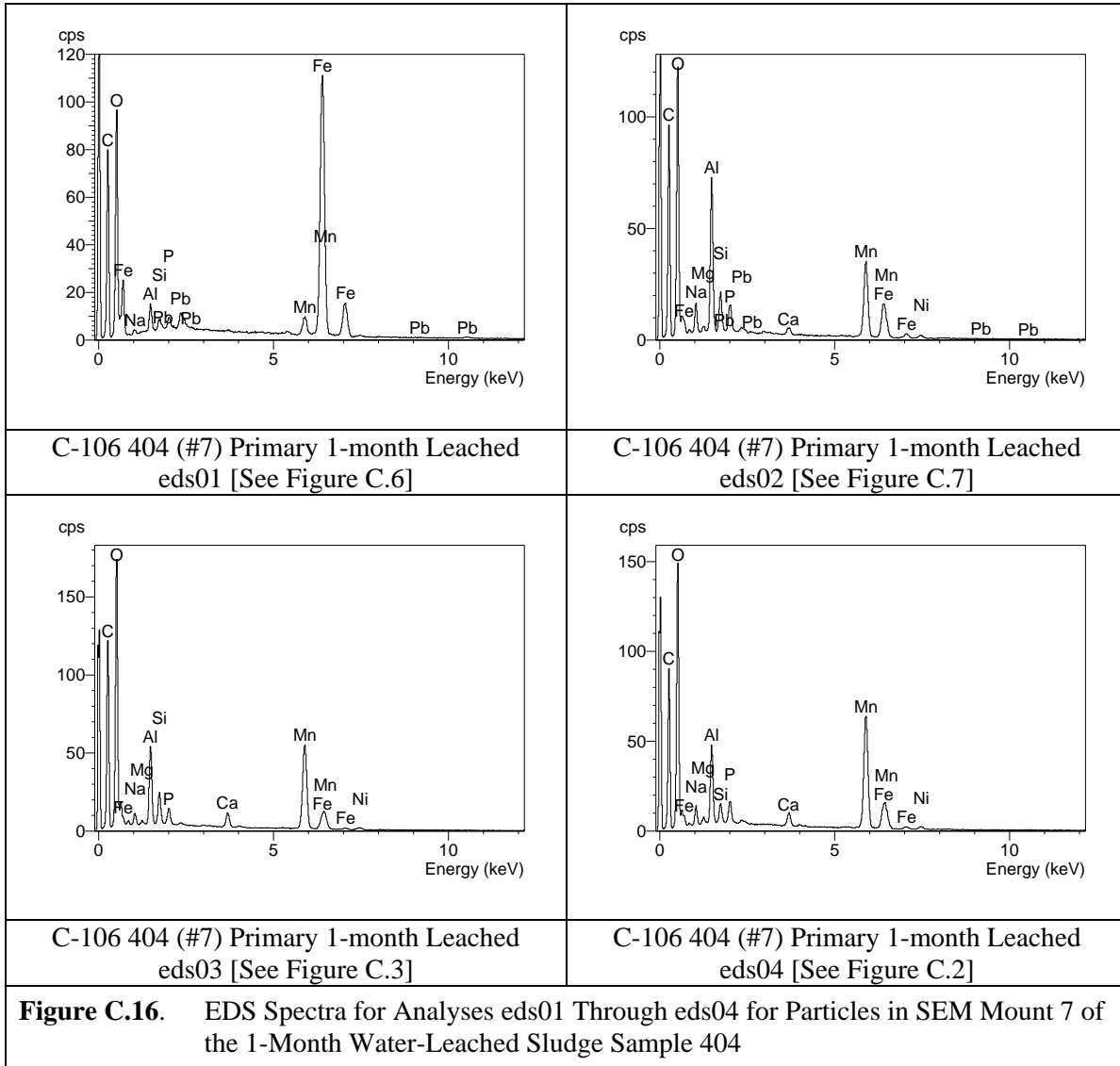**Figure C.82**. Micrograph Showing at Higher Magnification the Particle Labeled B in Figure C.79
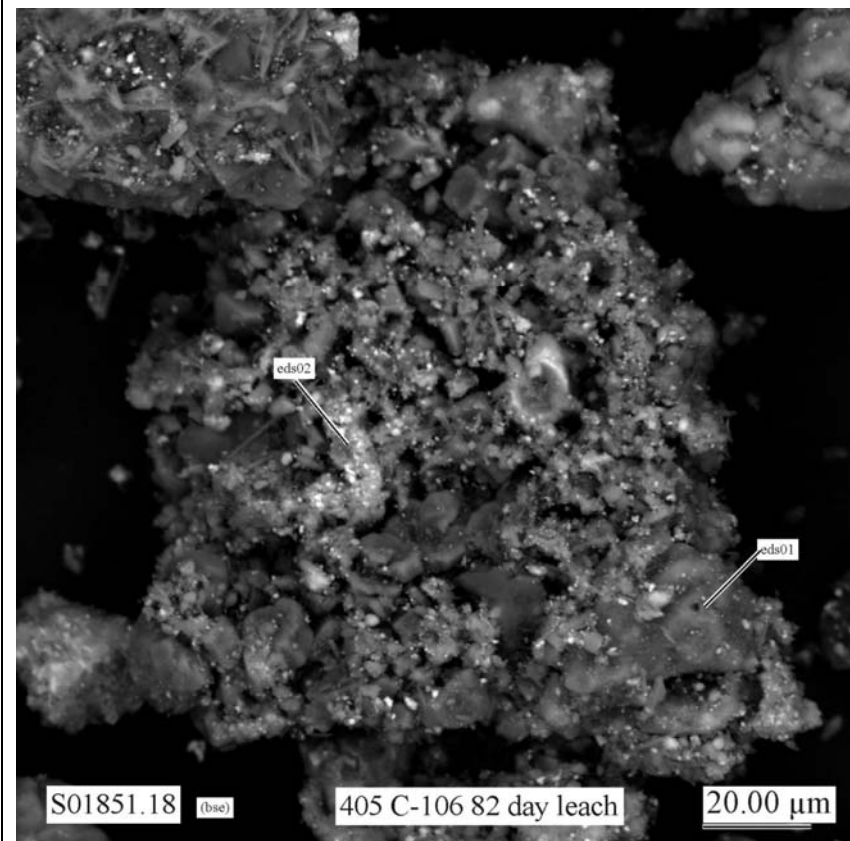
Figure C.82. Micrograph Showing Morphologies of Typical Particles in Sample 405 of 82-Day Water-Leached Sludge
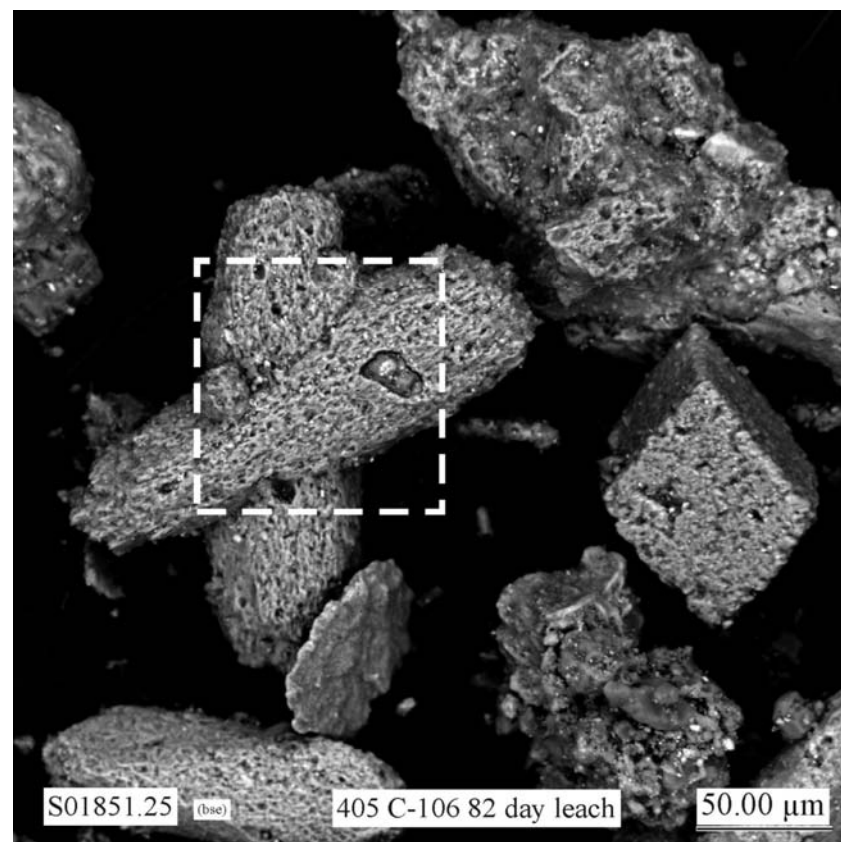
Figure C.84. Micrograph Showing Morphologies of Typical Particles in Sample 405 of 82-Day Water-Leached Sludge
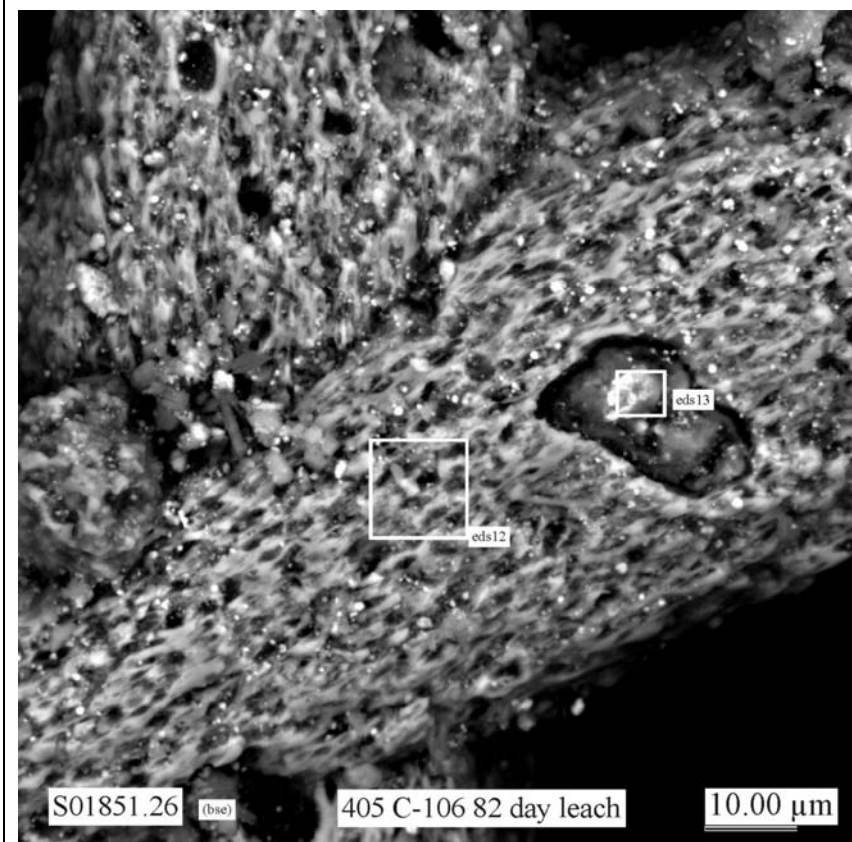
Figure C.85. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square in Figure C.84

S01851.23 (bse) 405 C-106 82 day leach 8.00 μm
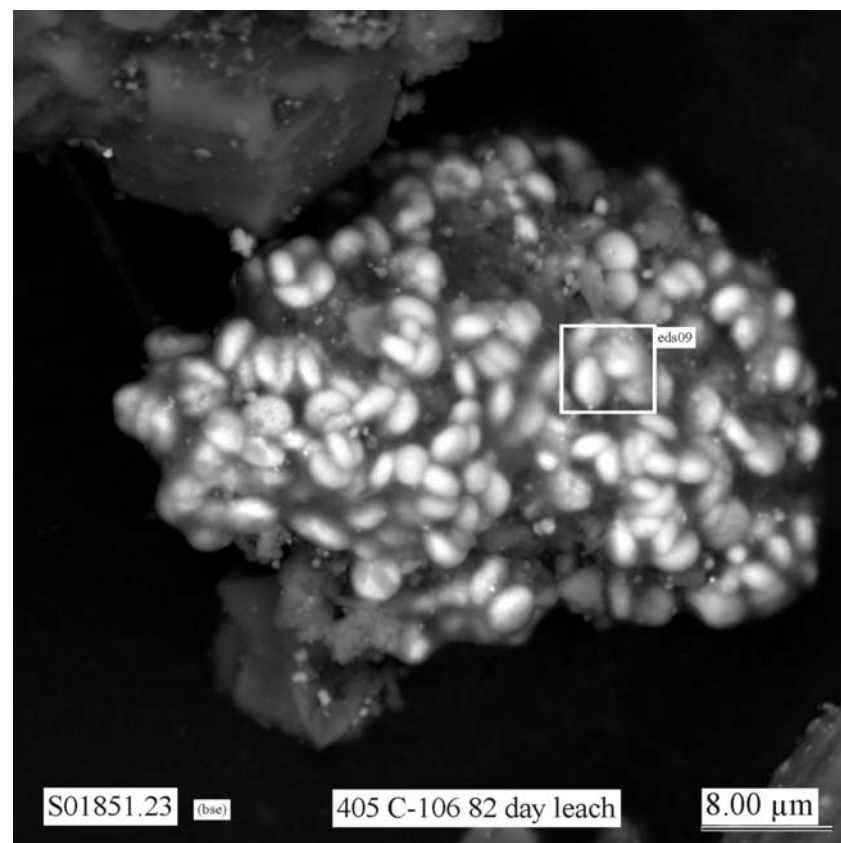
Figure C.86. Micrograph Showing Morphology of an Aggregate of Fe-Rich Particles in Sample 405 of 82-Day Water-Leached Sludge

S01851.24 (bse) 405 C-106 82 day leach 10.00 μm
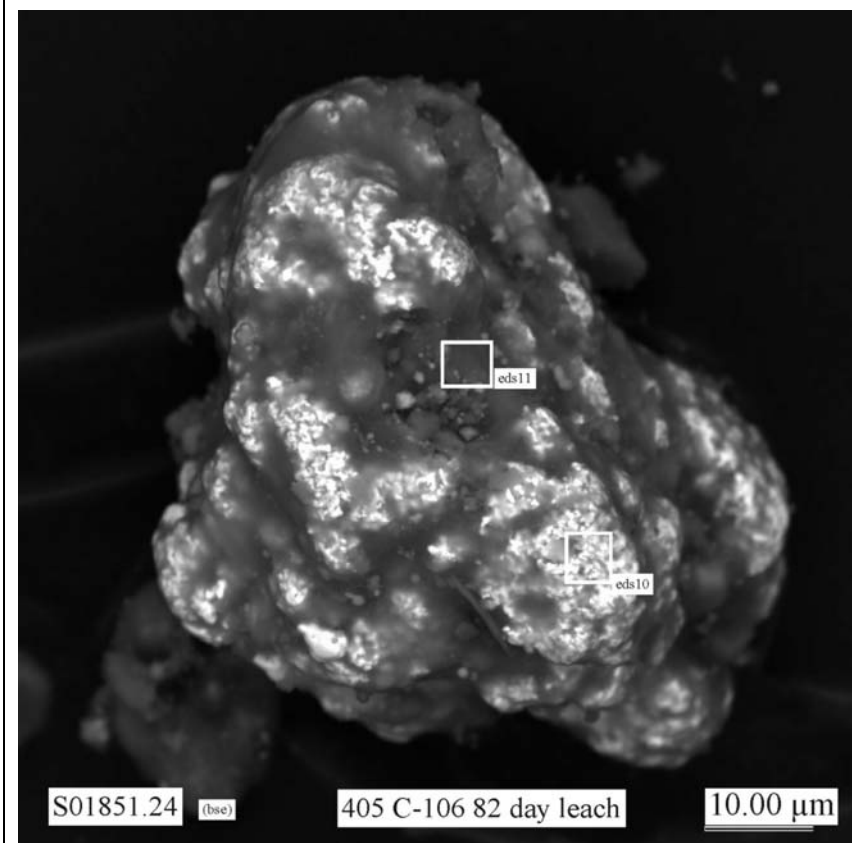
Figure C.87. Micrograph Showing Morphologies of a Typical Ag-Hg-Rich Particle in Sample 405 of 82-Day Water-Leached Sludge
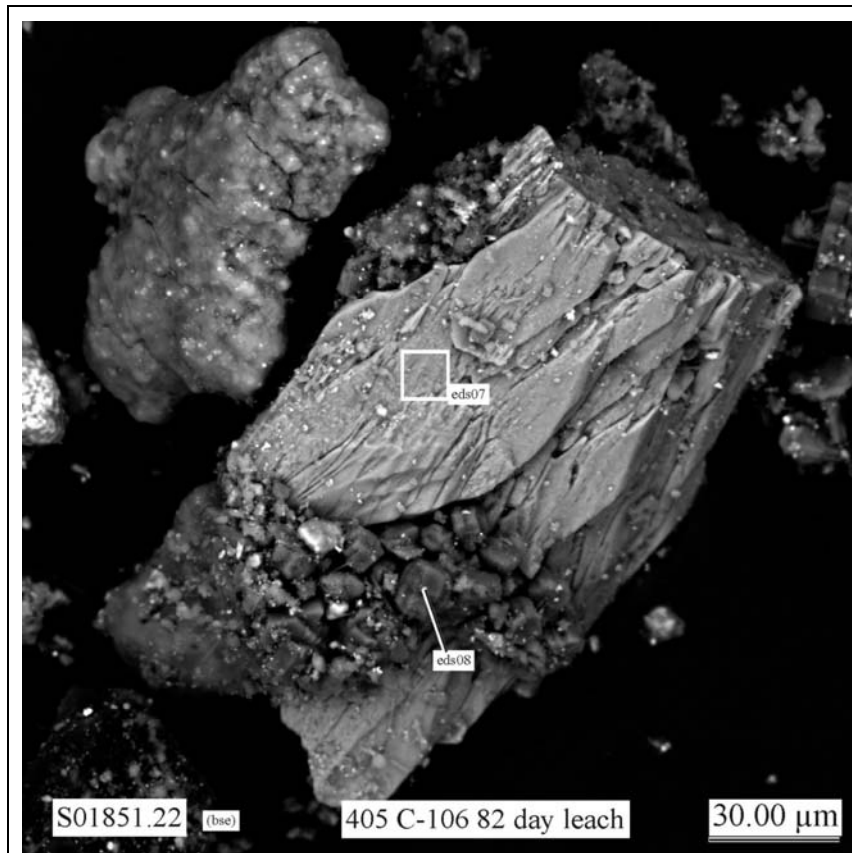
Figure C.88. Micrograph Showing Morphologies of Typical Particles in Sample 405 of 82-Day Water-Leached Sludge
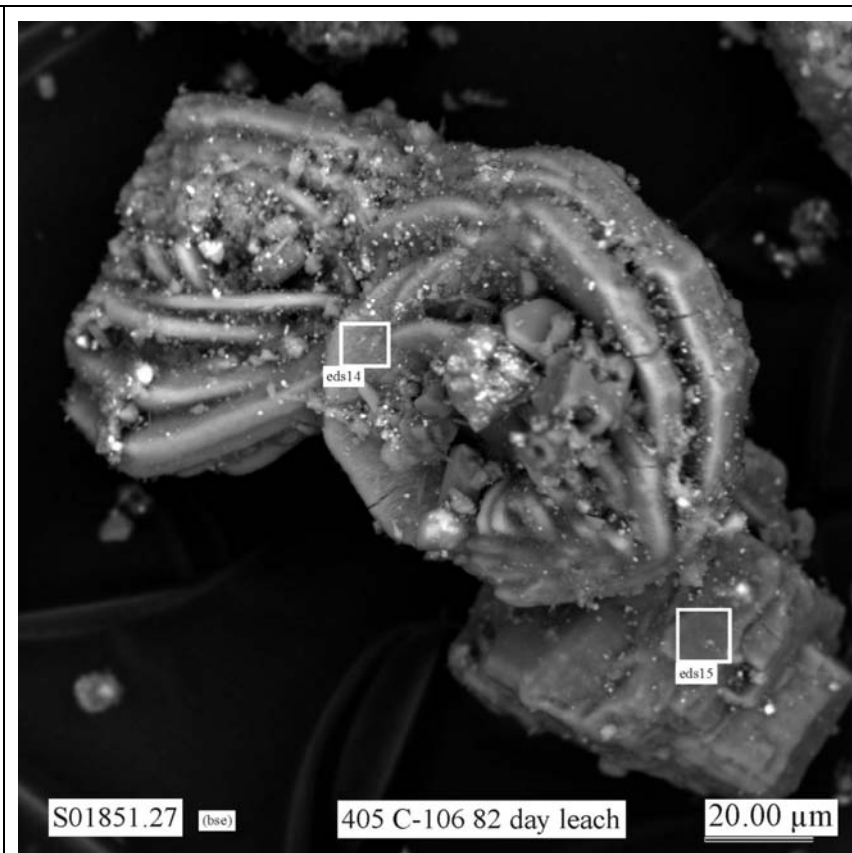
Figure C.89. Micrograph Showing Morphologies of Typical Particles in Sample 405 of 82-Day Water-Leached Sludge
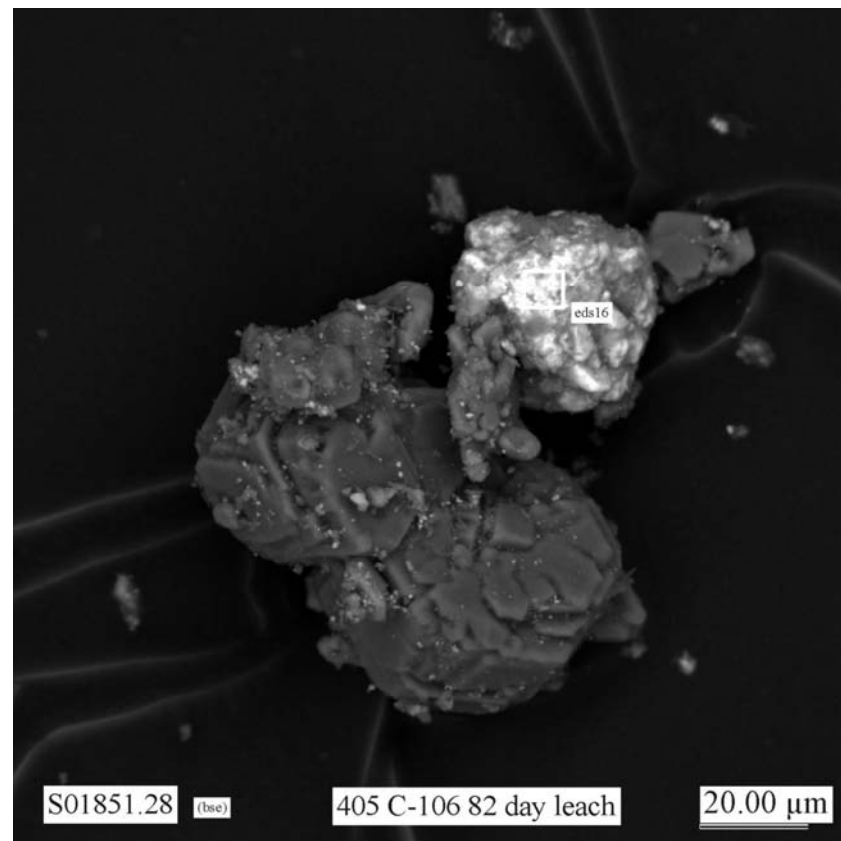
Figure C.90. Micrograph Showing Morphologies of Typical Particles in Sample 405 of 82-Day Water-Leached Sludge
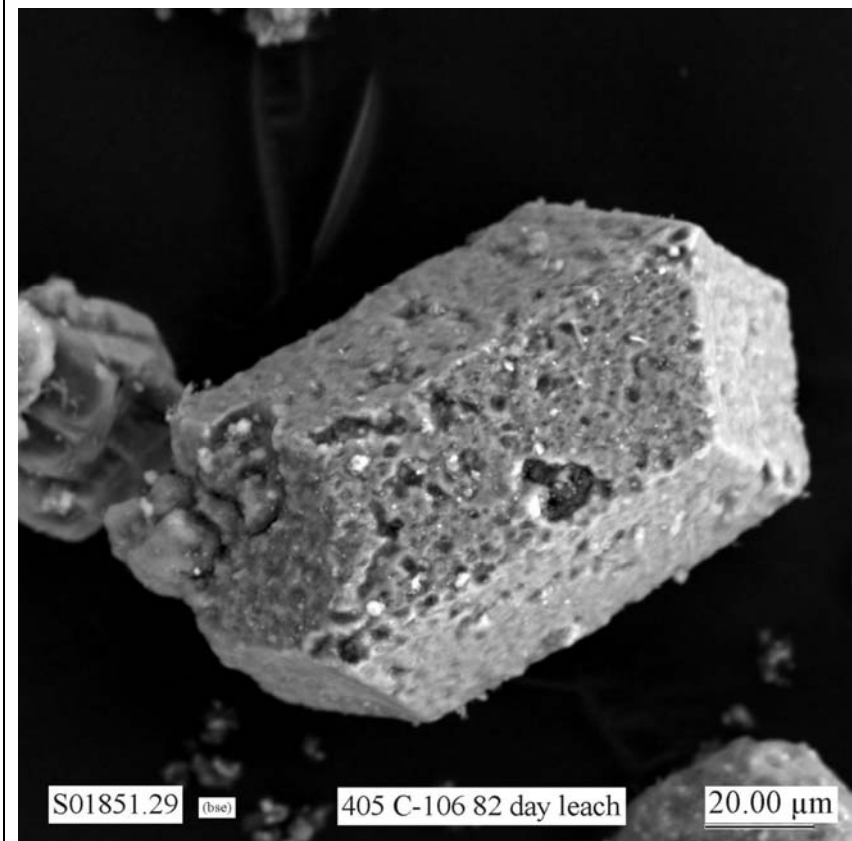
Figure C.91. Micrograph Showing Morphologies of Typical Particles in Sample 405 of 82-Day Water-Leached Sludge
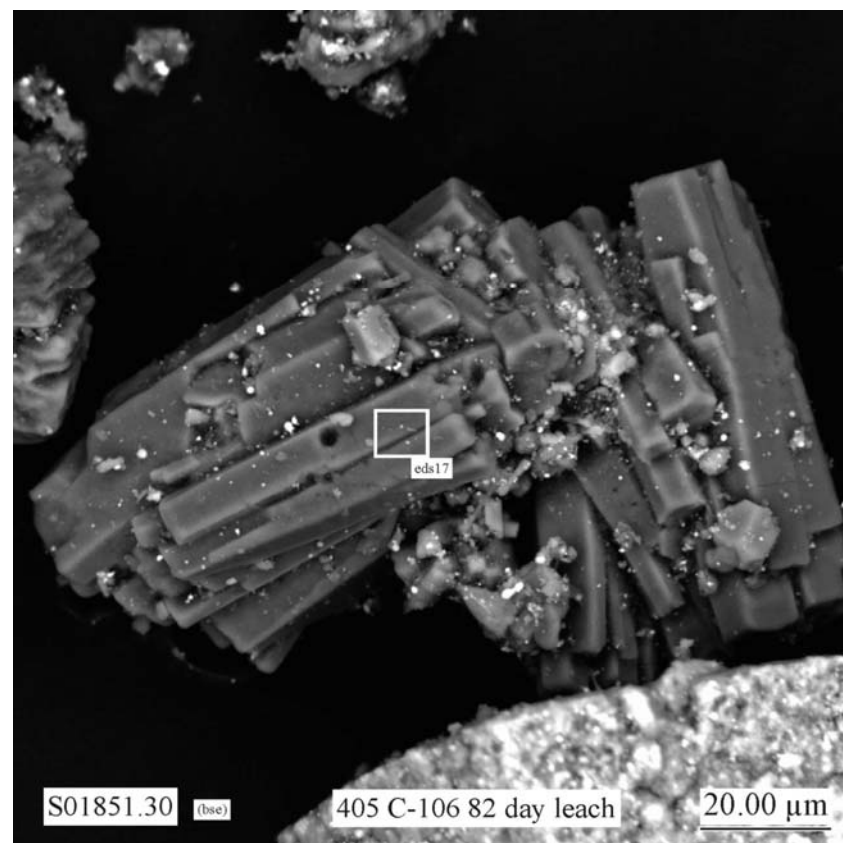
Figure C.92. Micrograph Showing Morphologies of Typical Particles in Sample 405 of 82-Day Water-Leached Sludge
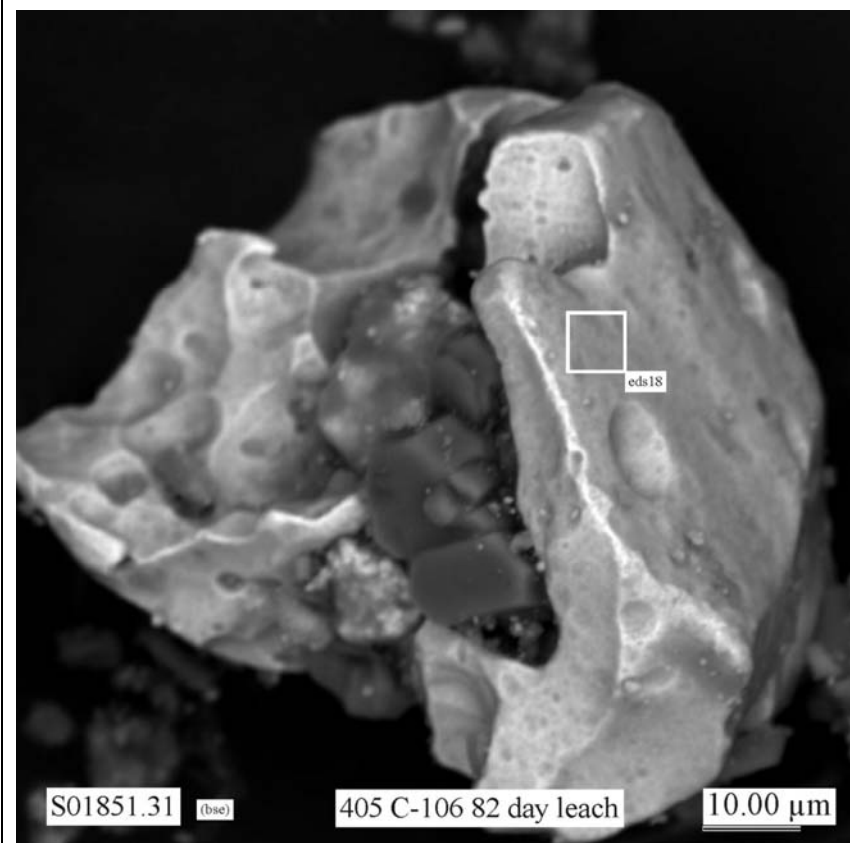
Figure C.93. Micrograph Showing Morphology of an Fe-Cr-Rich Particle in Sample 405 of 82-Day Water-Leached Sludge

C-106 405 Duplicate 82-day Leached
eds01 [See Figure C.82]

C-106 405 Duplicate 82-day Leached
eds02 [See Figure C.82]

C-106 405 Duplicate 82-day Leached
eds03 [See Figure C.80]

C-106 405 Duplicate 82-day Leached
eds04 [See Figure C.80]

Figure C.94. EDS Spectra for Analyses eds01 Through eds04 for Particles in Sample 405 of 82-Day Water-Leached Sludge

C-106 405 Duplicate 82-day Leached eds05 [See Figure C.81]

C-106 405 Duplicate 82-day Leached eds06 [See Figure C.82]

C-106 405 Duplicate 82-day Leached eds07 [See Figure C.88]

C-106 405 Duplicate 82-day Leached eds08 [See Figure C.88]

Figure C.95.   EDS Spectra for Analyses eds05 Through eds08 for Particles in Sample 405 of 82-Day Water-Leached Sludge

| C-106 405 Duplicate 82-day Leached eds09 [See Figure C.86] | C-106 405 Duplicate 82-day Leached eds10 [See Figure C.87] |
| C-106 405 Duplicate 82-day Leached eds11 [See Figure C.87] | C-106 405 Duplicate 82-day Leached eds12 [See Figure C.85] |

Figure C.96.   EDS Spectra for Analyses eds09 through eds12 for Particles in Sample 405 of 82-Day Water-Leached Sludge

| C-106 405 Duplicate 82-day Leached eds13 [See Figure C.85] | C-106 405 Duplicate 82-day Leached eds15 [See Figure C.89] |
| C-106 405 Duplicate 82-day Leached eds16 [See Figure C.90] | C-106 405 Duplicate 82-day Leached eds17 [See Figure C.92] |

Figure C.97.   EDS Spectra for Analyses eds13, eds15, eds16, and eds17 for Particles in Sample 405 of 82-Day Water-Leached Sludge

C-106 405 Duplicate 82-day Leached
eds18 [See Figure C.93]

Figure C.98.   EDS Spectra for Analyses eds18 for a
Particles in Sample 405 of 82-Day
Water-Leached Sludge

Appendix D

SEM Micrographs and EDS Spectra
for HF Sequential Extract of Tank C-106 Sludge

Appendix D

SEM Micrographs and EDS Spectra for HF Sequential Extract of Tank C-106 Sludge

This appendix includes the scanning electron microscope (SEM) micrographs and the energy-dispersive X-ray spectrometry (EDS) spectra for samples of residual sludge from tank 241-C-106 (C-106) (primary 404 and duplicate 405) remaining after the hydrofluoric (HF) (Stage 1) sequential extraction. The operating conditions for the SEM and procedures used for mounting SEM samples are described in Section 2.4 of the main report.

The name of each SEM digital image file, sample identification number, and a size scale bar are given, respectively, at the bottom left, center, and right of each SEM micrograph in this appendix. Micrographs labeled by "BSE" to the immediate right of the digital image file name indicate that the micrograph was collected with backscattered electrons. Areas identified by a letter and/or outlined by a dashed-line square in a micrograph designate sample material that was imaged at higher magnification, which is typically shown in figure(s) that immediately follow in the series for that sample.

Areas labeled by "eds" in the following SEM micrographs in this appendix identify locations of particles for which EDS spectra were recorded. The "eds" label given with each EDS spectrum correspond to the same "EDS" label used in the SEM micrographs for this sample.

One mount each of HF-extracted sludge from tank C-106 (primary 404 and duplicate 405) were analyzed by SEM/EDS. The SEM micrographs for the mount of HF-extracted sample 404 are shown in Figures D.1 through D.20. The EDS spectra for this mount are given in Figures D.21 through D.25. The SEM micrographs for the mount of HF-extracted sample 405 are shown in Figures D.26 through D.45, and the EDS spectra for this sample are given in Figures D.46 through D.51.
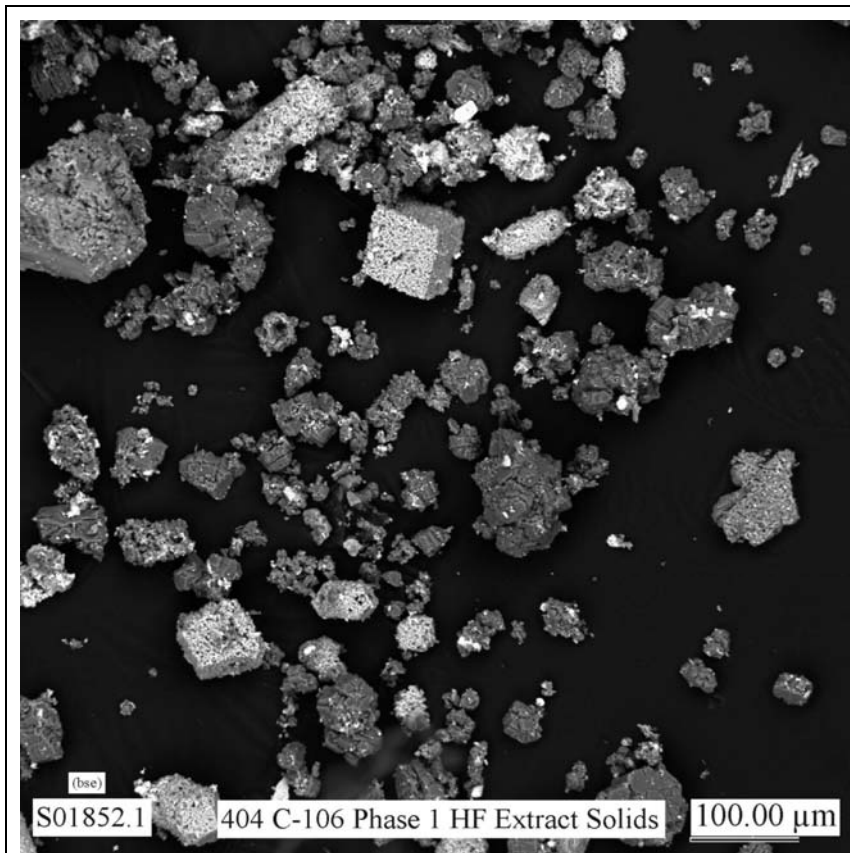
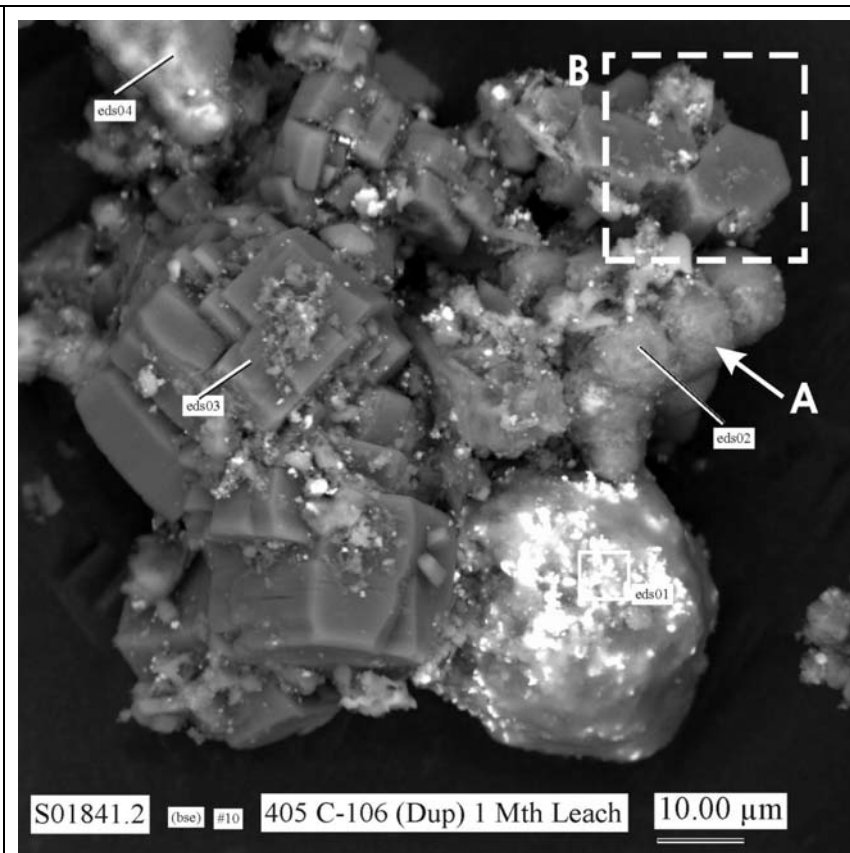| **Figure D.1**. | Low Magnification SEM Micrograph Showing General Morphologies of Particles in Sample 404 of HF-Extracted Sludge | **Figure D.2.** | Micrograph Showing Morphologies of Typical Particles in Sample 404 of HF-Extracted Sludge |
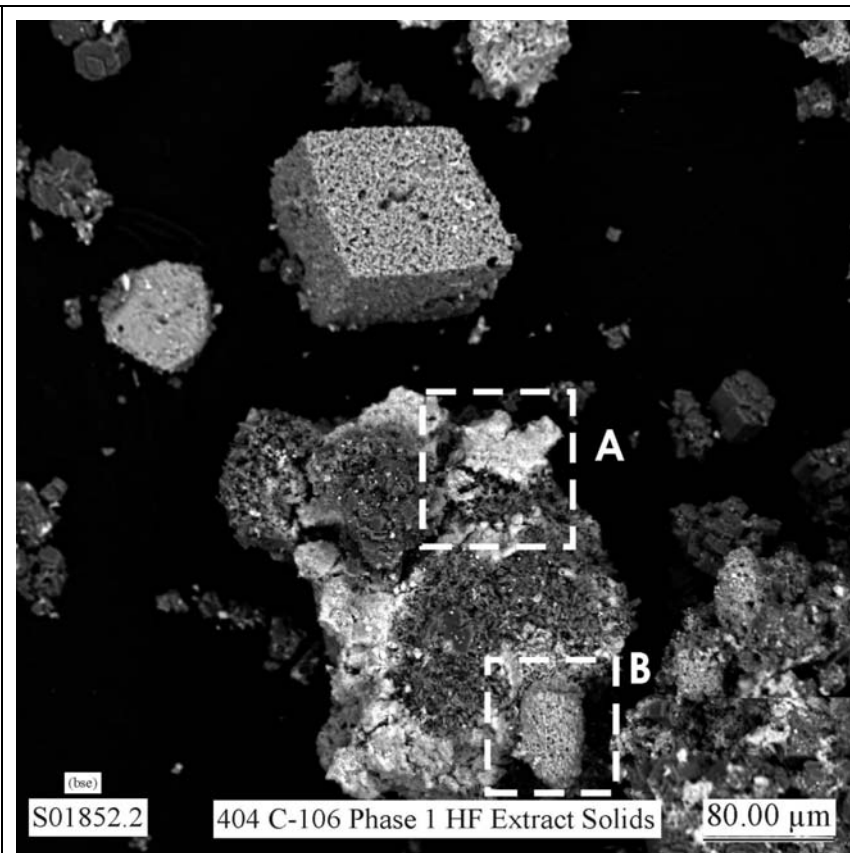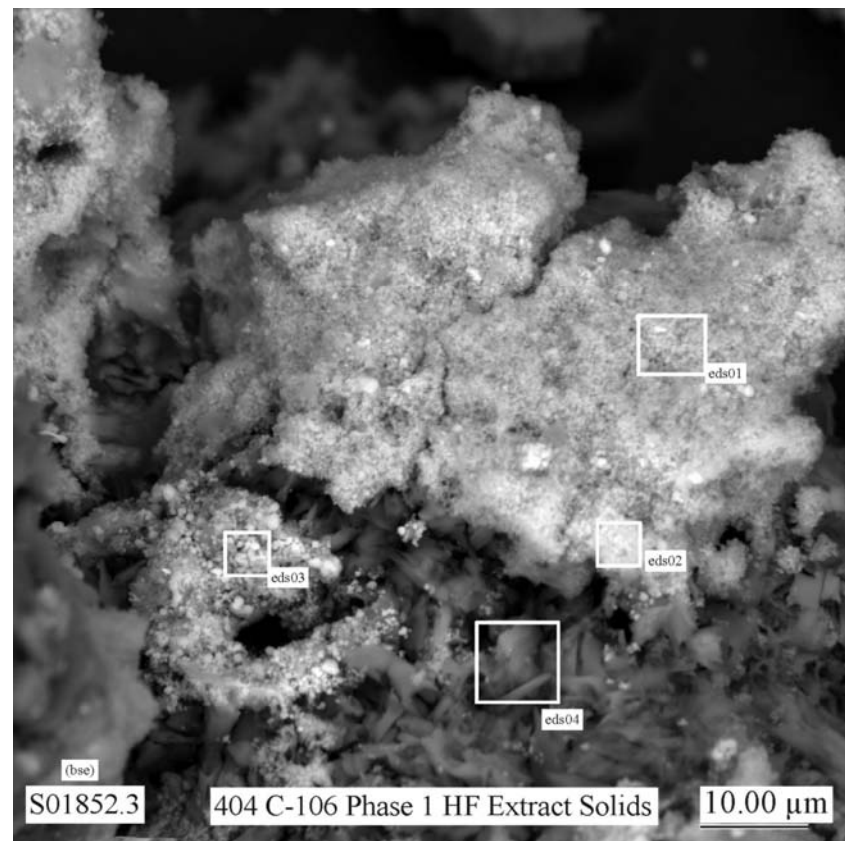
Figure D.3. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled A in Figure D.2
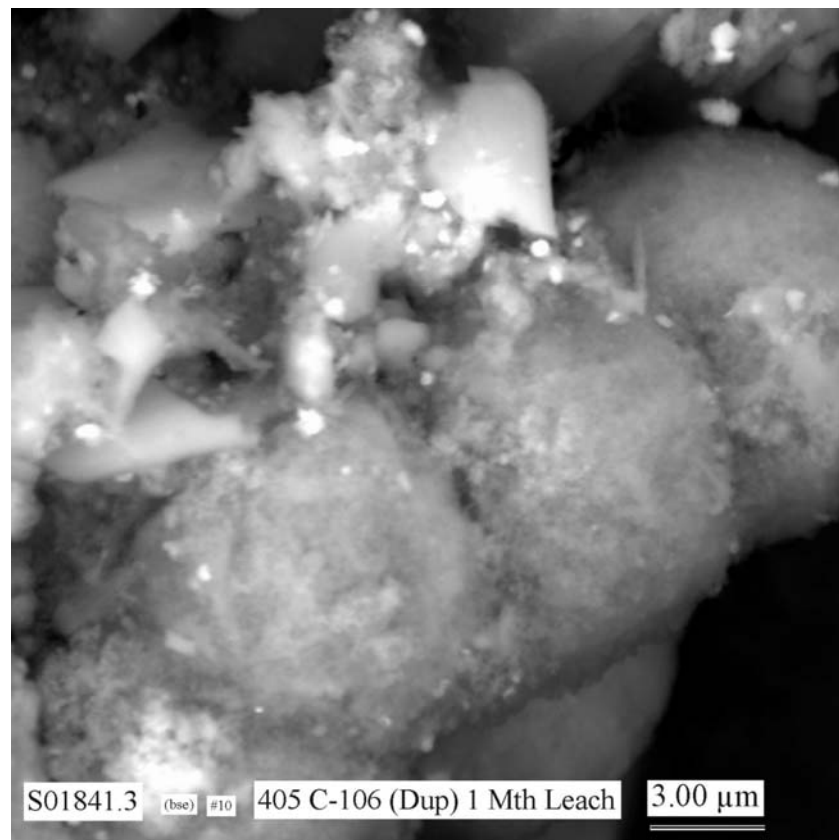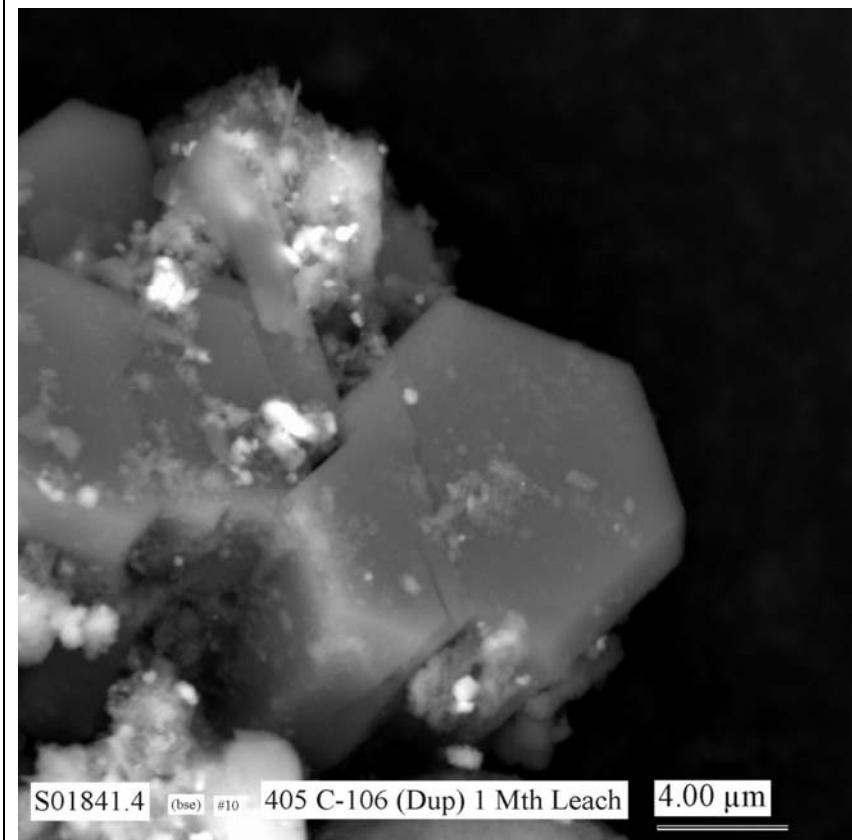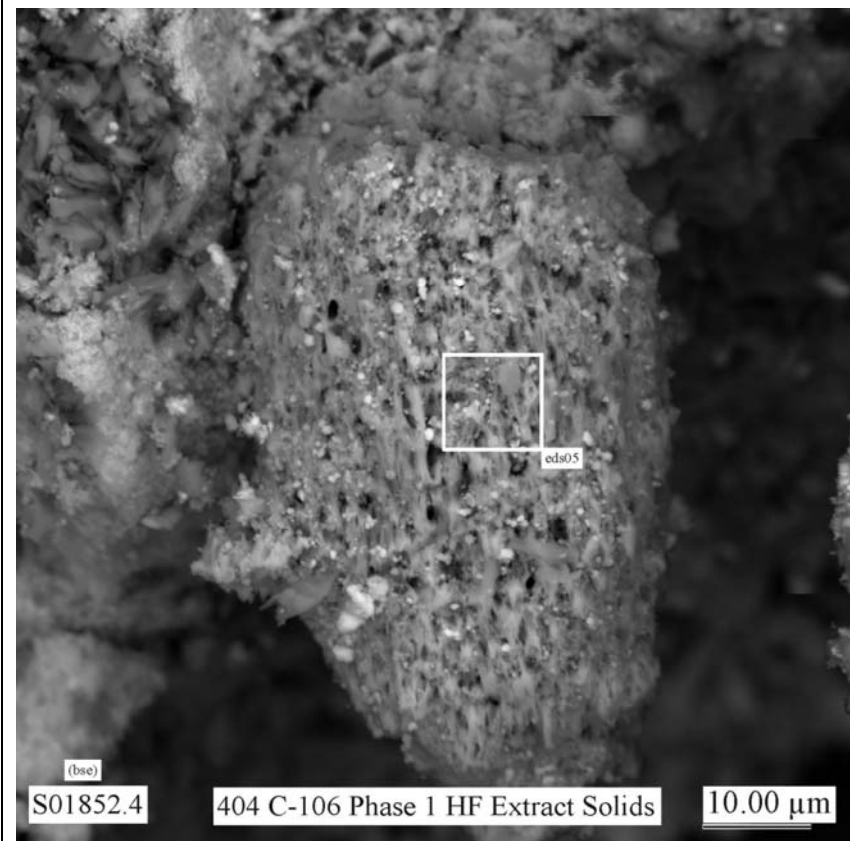

Figure D.4. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled B in Figure D.2
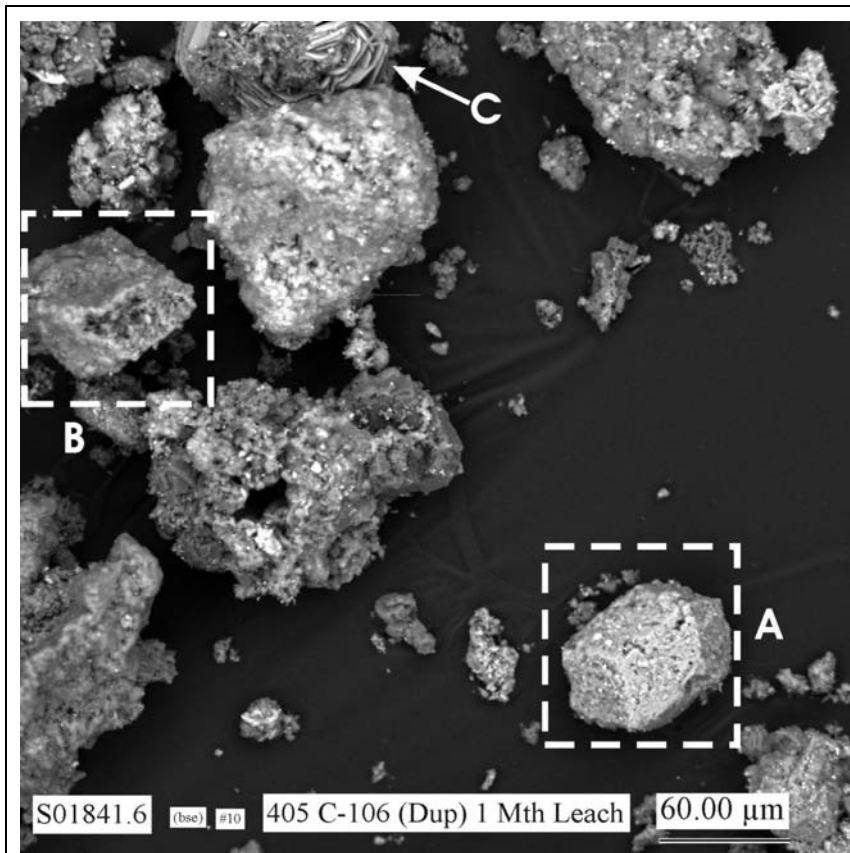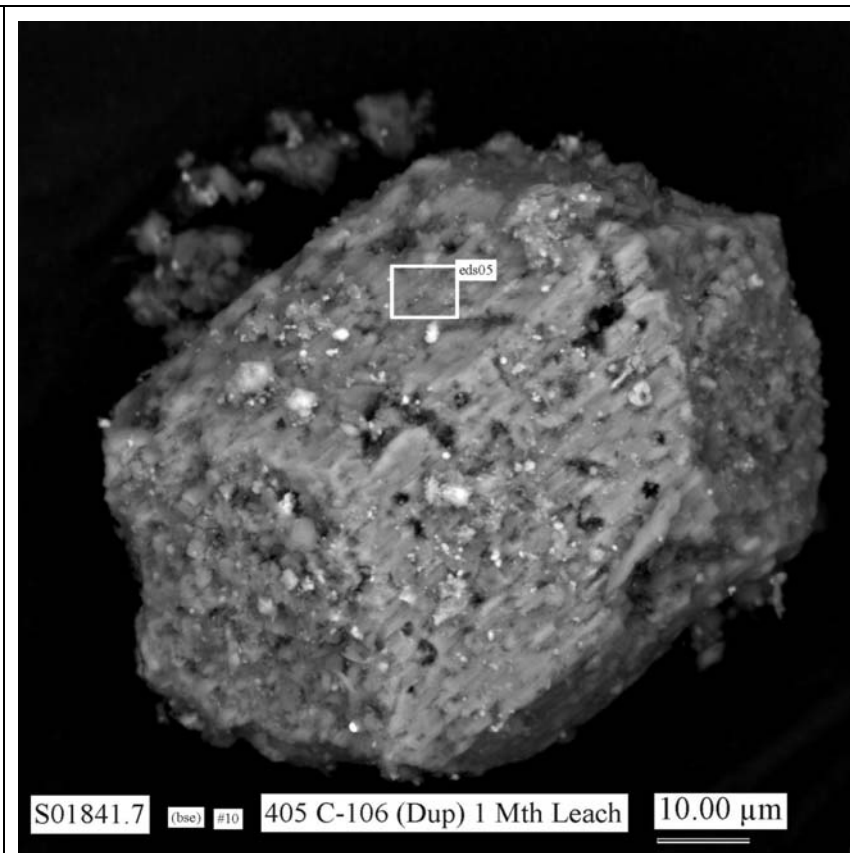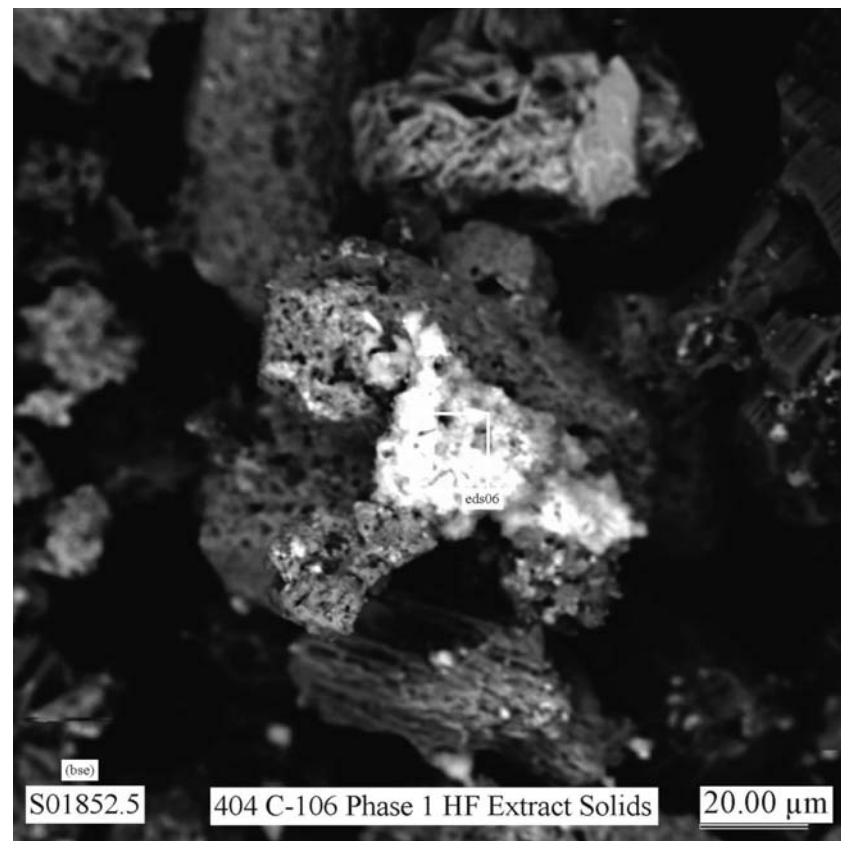
Figure D.5. Micrograph Showing Morphologies of Typical Particles in Sample 404 of HF-Extracted Sludge
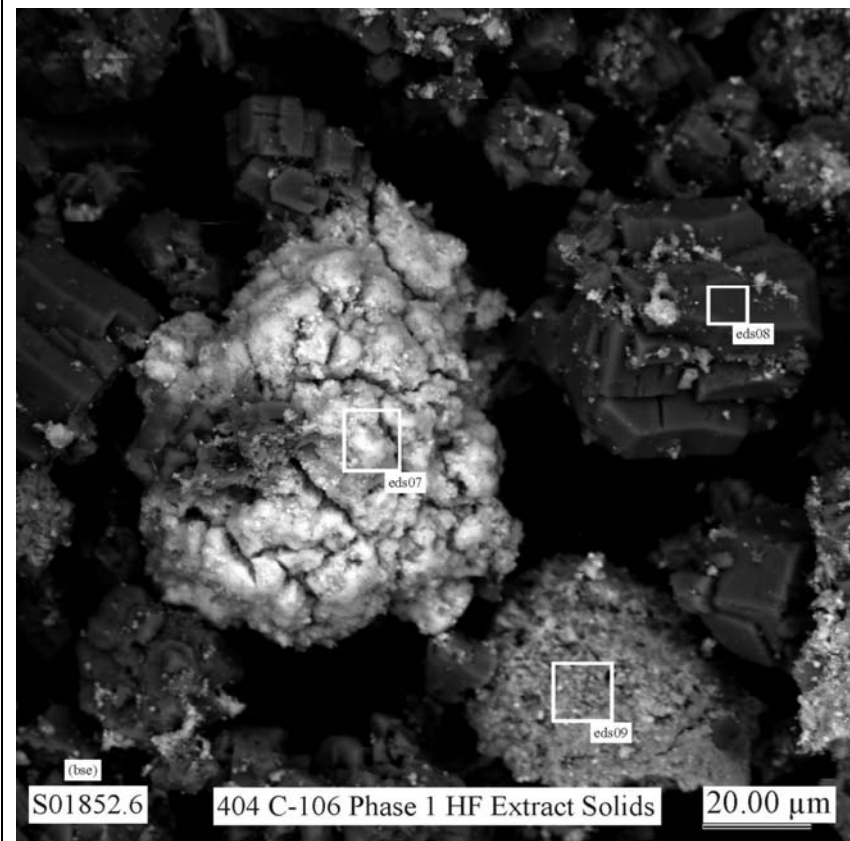
Figure D.6. Micrograph Showing Morphologies of Typical Particles in Sample 404 of HF-Extracted Sludge
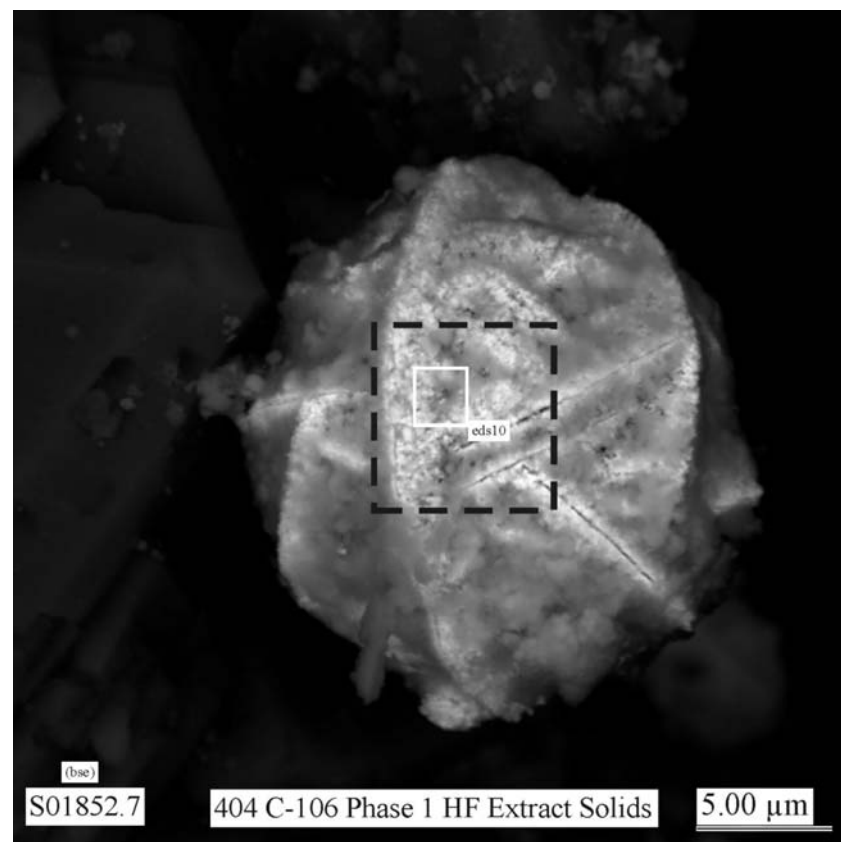
**Figure D.7.** Micrograph Showing Morphology of Unusual Hg-Pd-Ag Rich Particle in Sample 404 of HF-Extracted Sludge
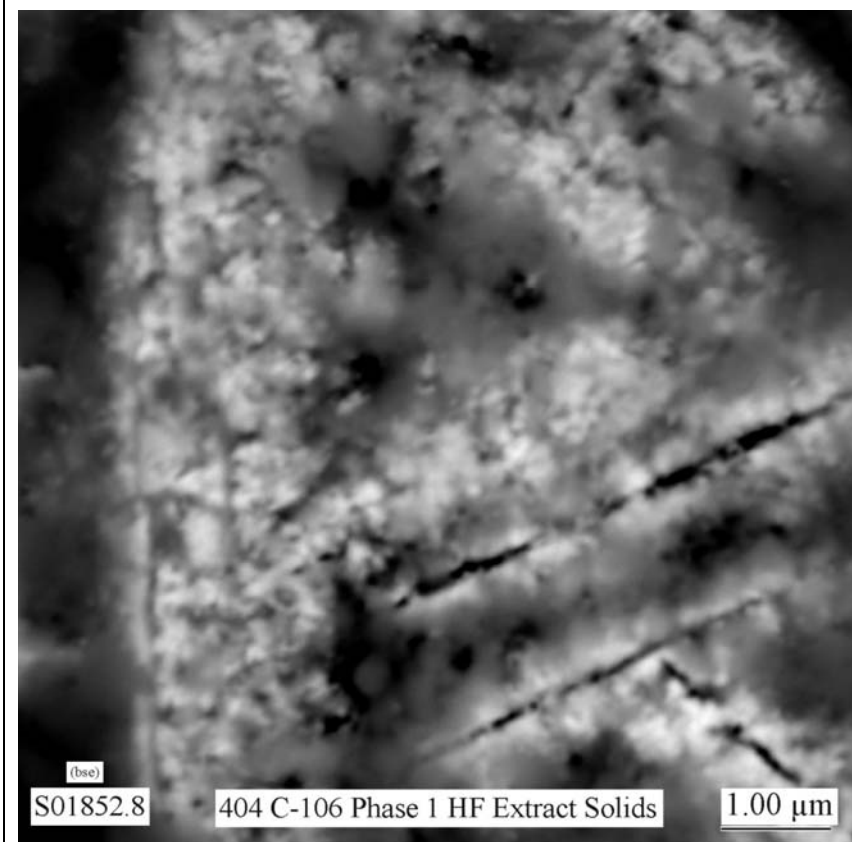
Figure D.8. Micrograph Showing at Higher Magnification the Area Indicated by the Black Dashed-Line Square in Figure D.7.
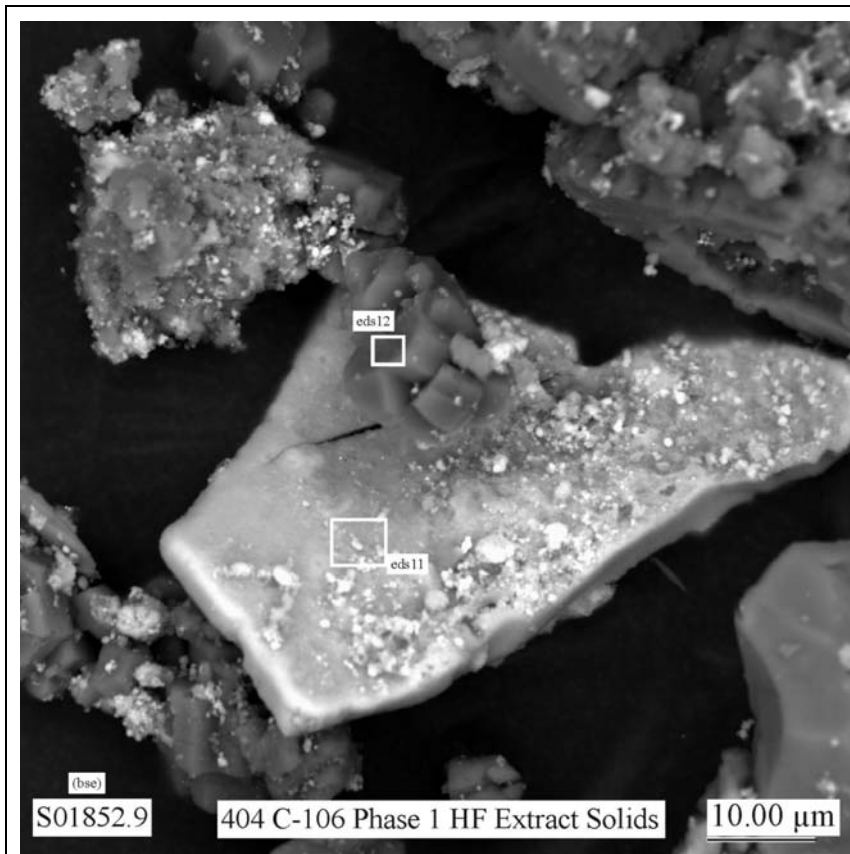
Figure D.9. Micrograph Showing Morphologies of Typical Particles in Sample 404 of HF-Extracted Sludge
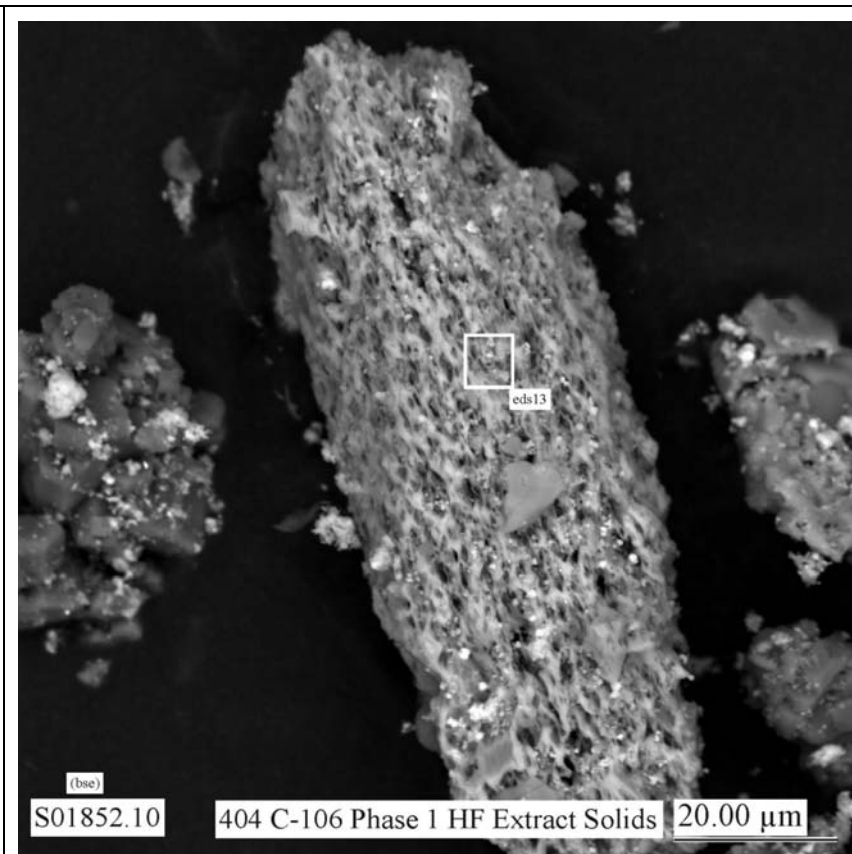
Figure D.10. Micrograph Showing Morphologies of Typical Particles in Sample 404 of HF-Extracted Sludge
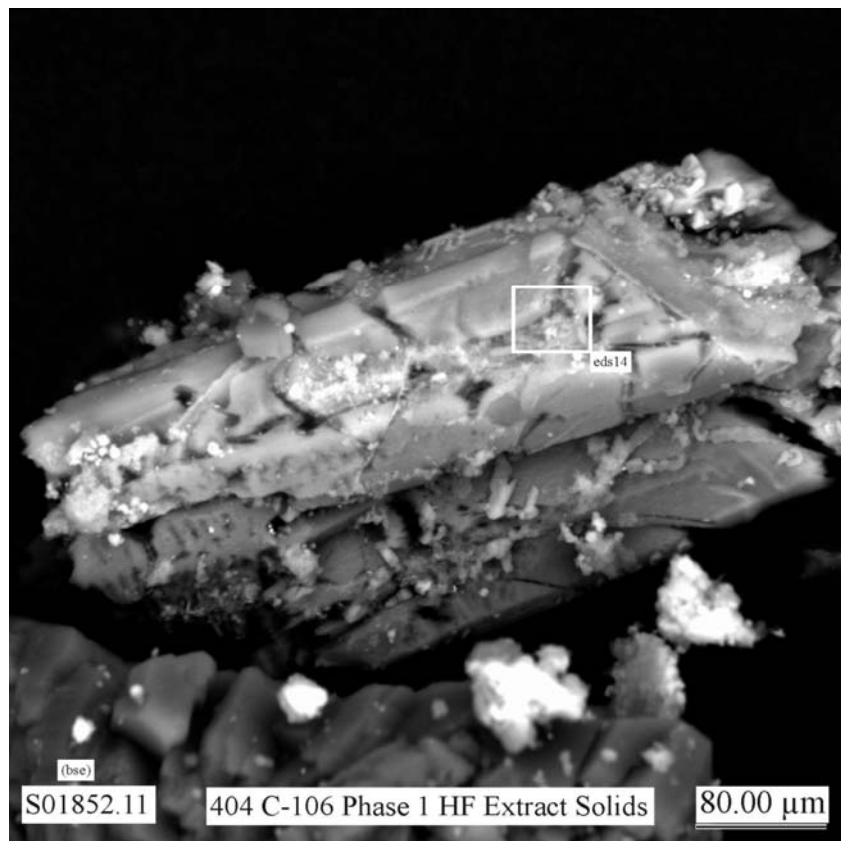
Figure D.11. Micrograph Showing Morphologies of Typical Particles in Sample 404 of HF-Extracted Sludge
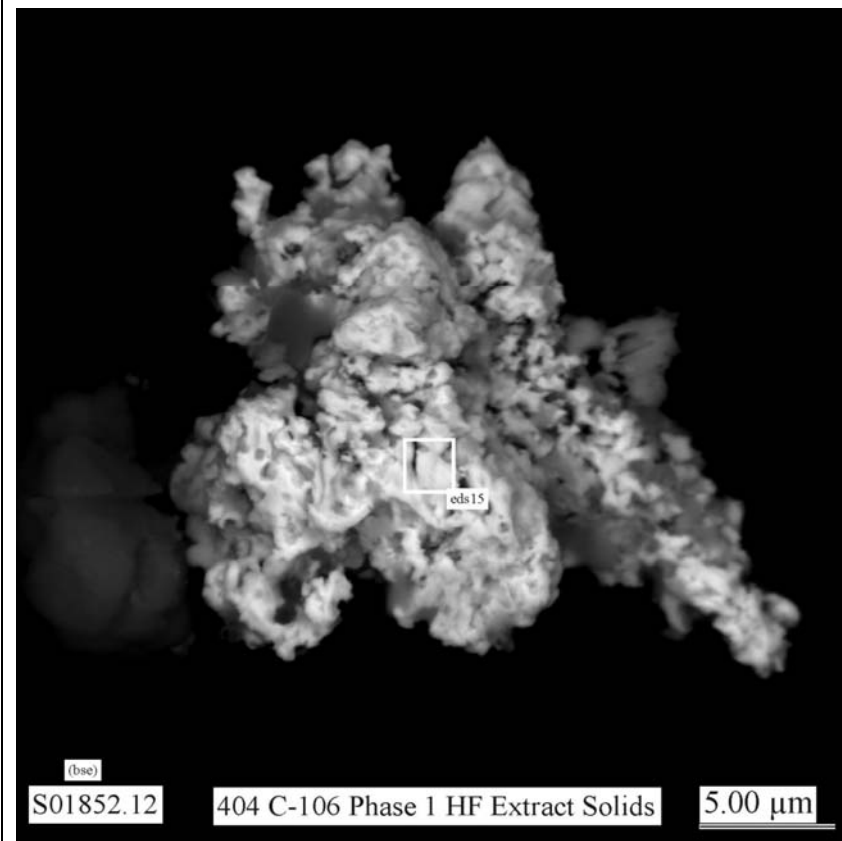
Figure D.12. Micrograph Showing Morphologies of Typical Particles in Sample 404 of HF-Extracted Sludge
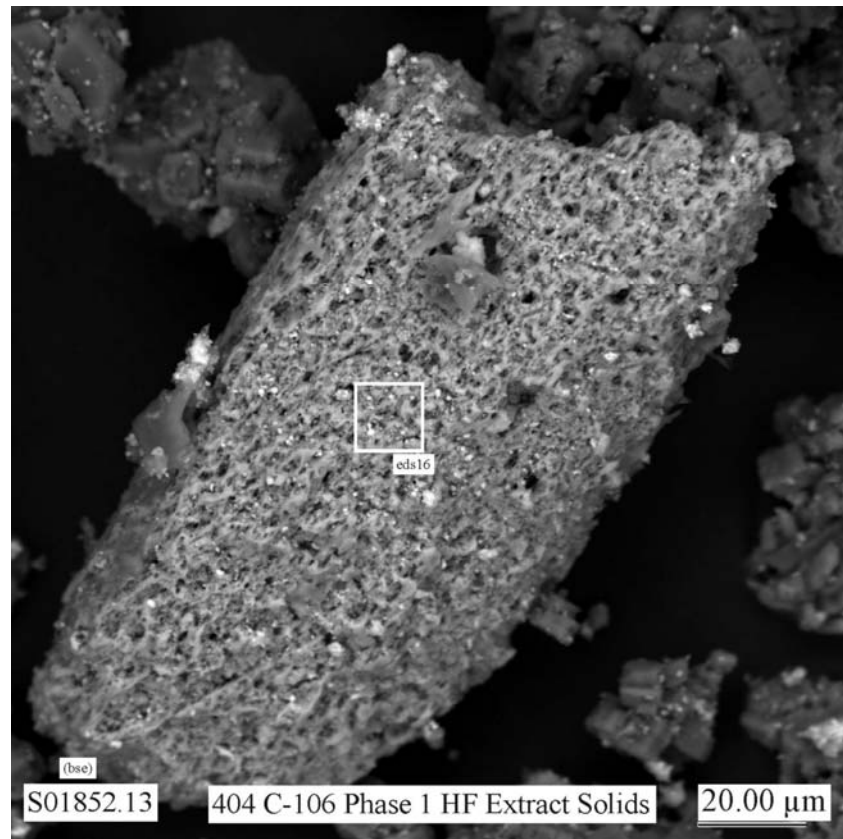
Figure D.13. Micrograph Showing Morphologies of Typical Particles in Sample 404 of HF-Extracted Sludge
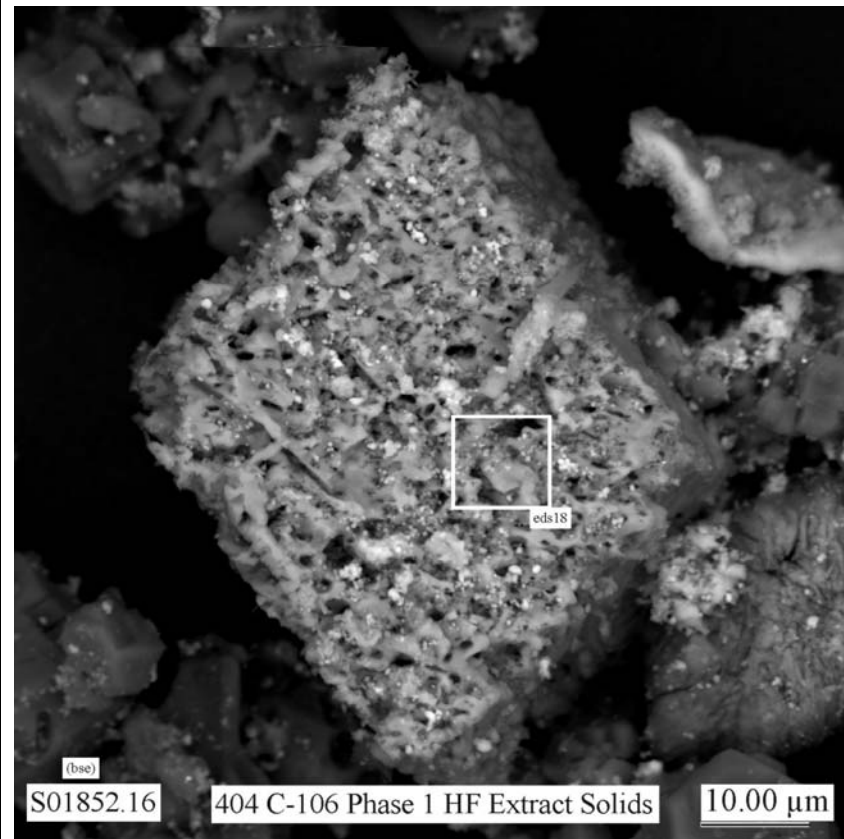
Figure D.14. Micrograph Showing Morphologies of Typical Particles in Sample 404 of HF-Extracted Sludge

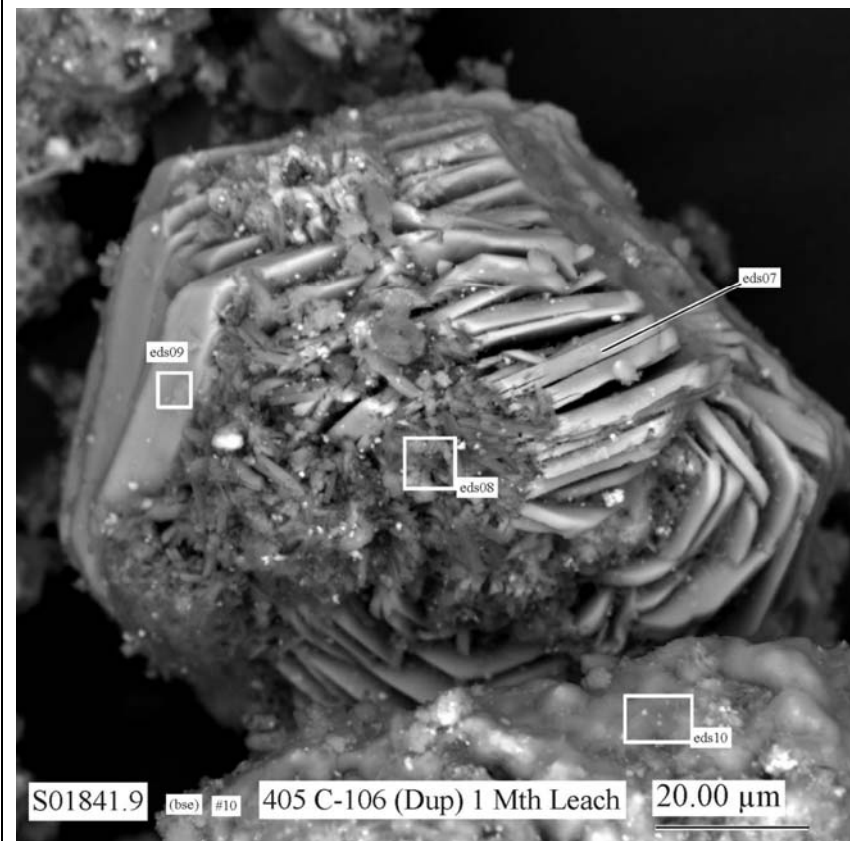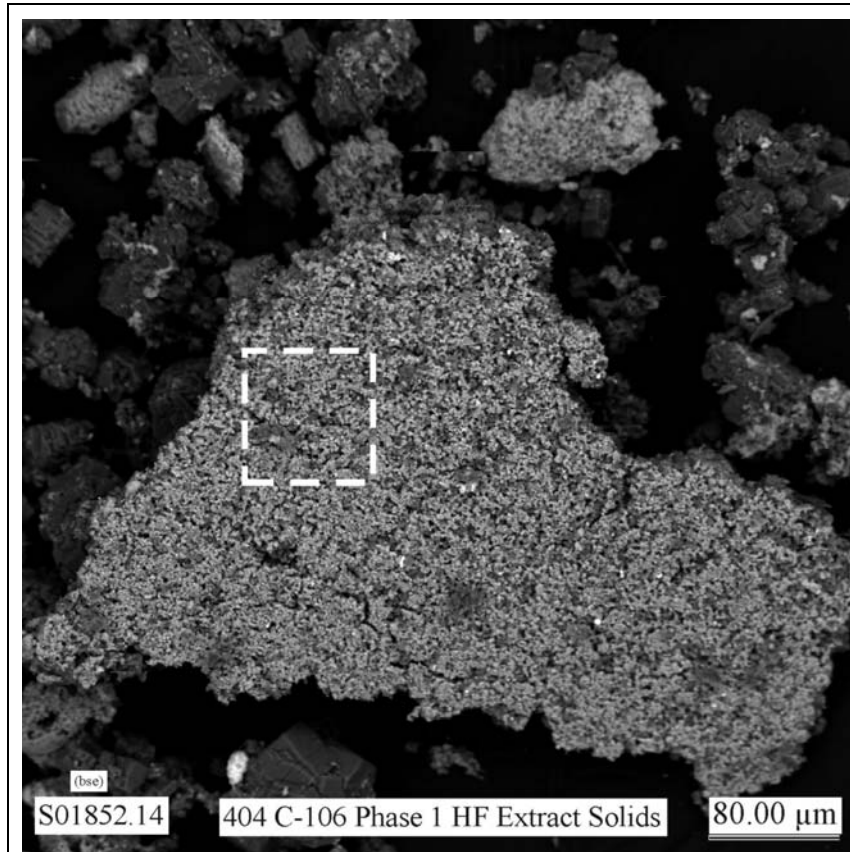| **Figure D.15.** | Micrograph Showing Morphologies of Typical Particles in Sample 404 of HF-Extracted Sludge | **Figure D.16.** | Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square in Figure D.15. |
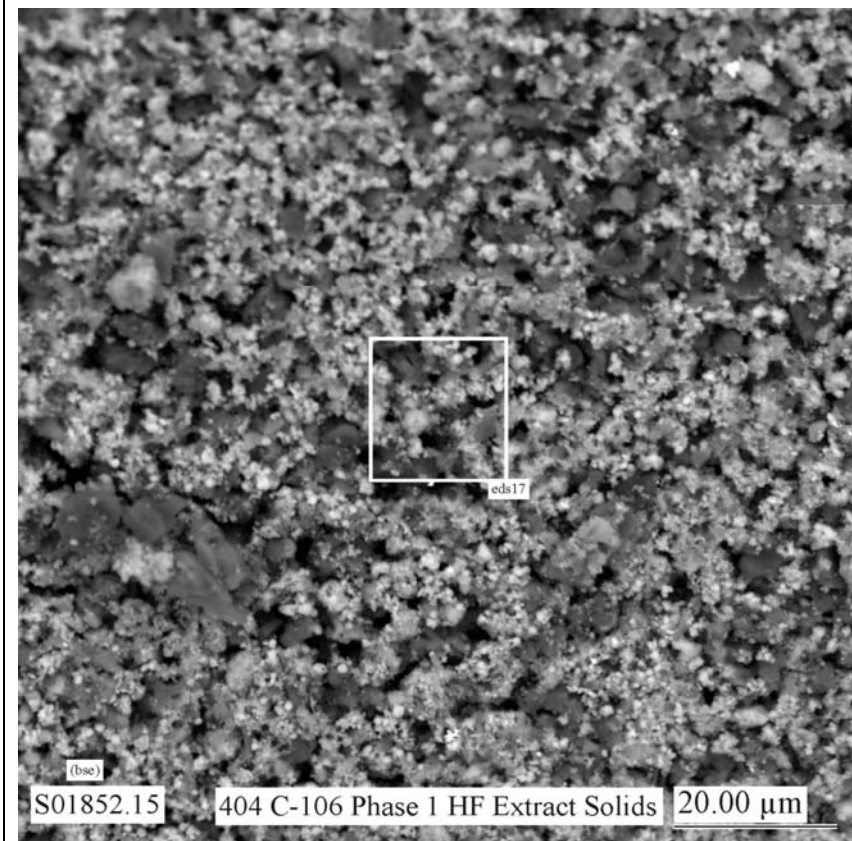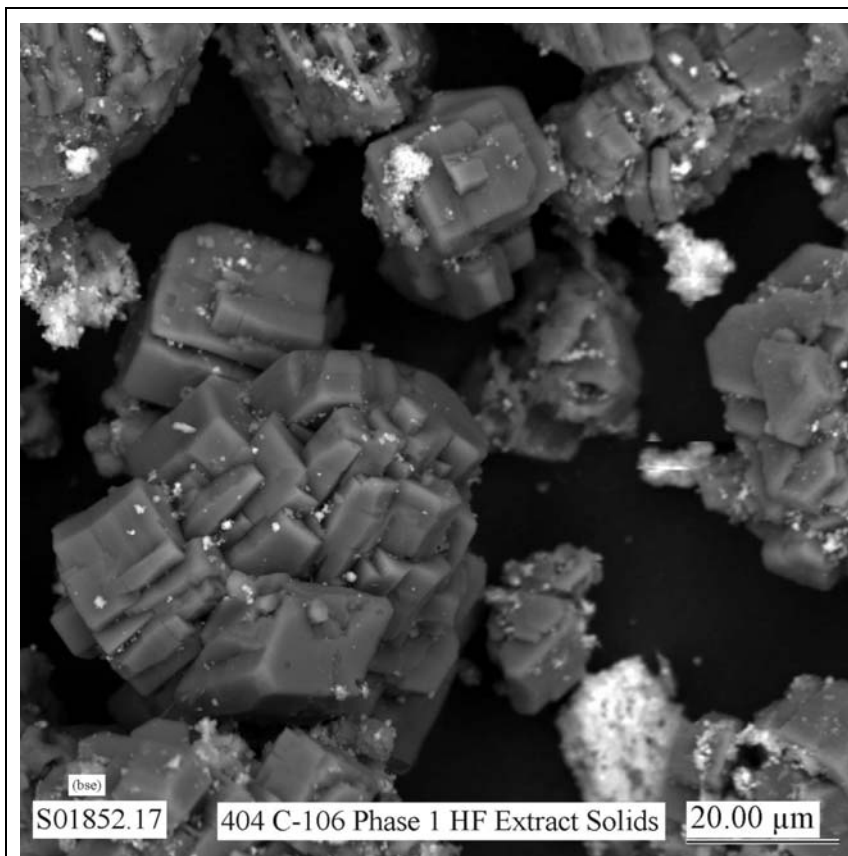
Figure D.17. Micrograph Showing Morphologies of Typical Particles in Sample 404 of HF-Extracted Sludge
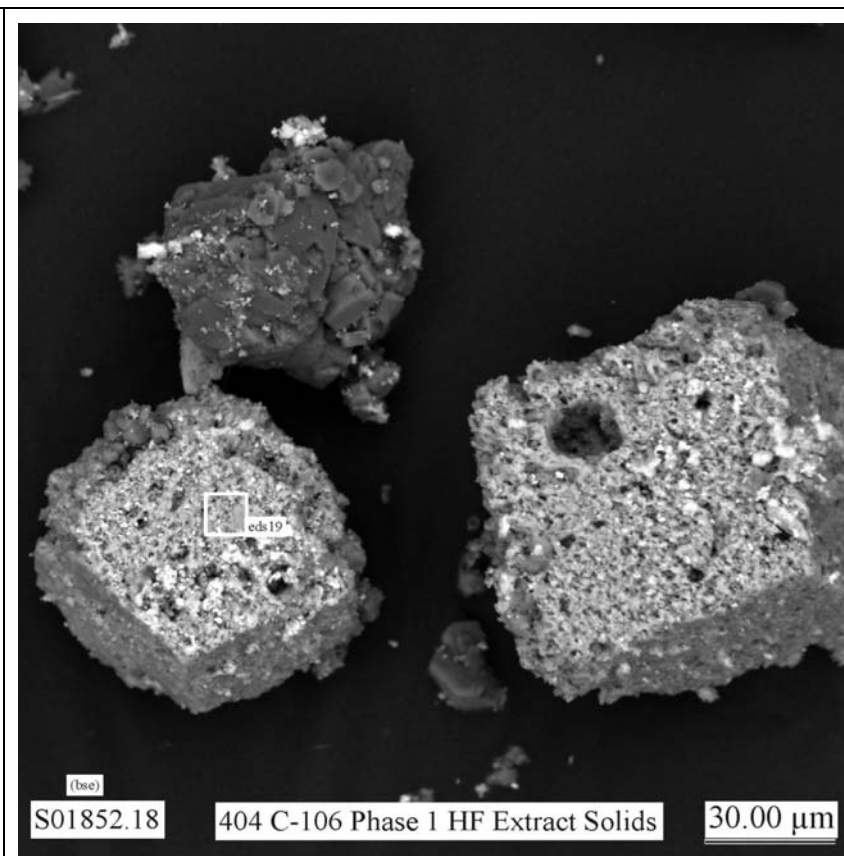
Figure D.18. Micrograph Showing Morphologies of Typical Particles in Sample 404 of HF-Extracted Sludge
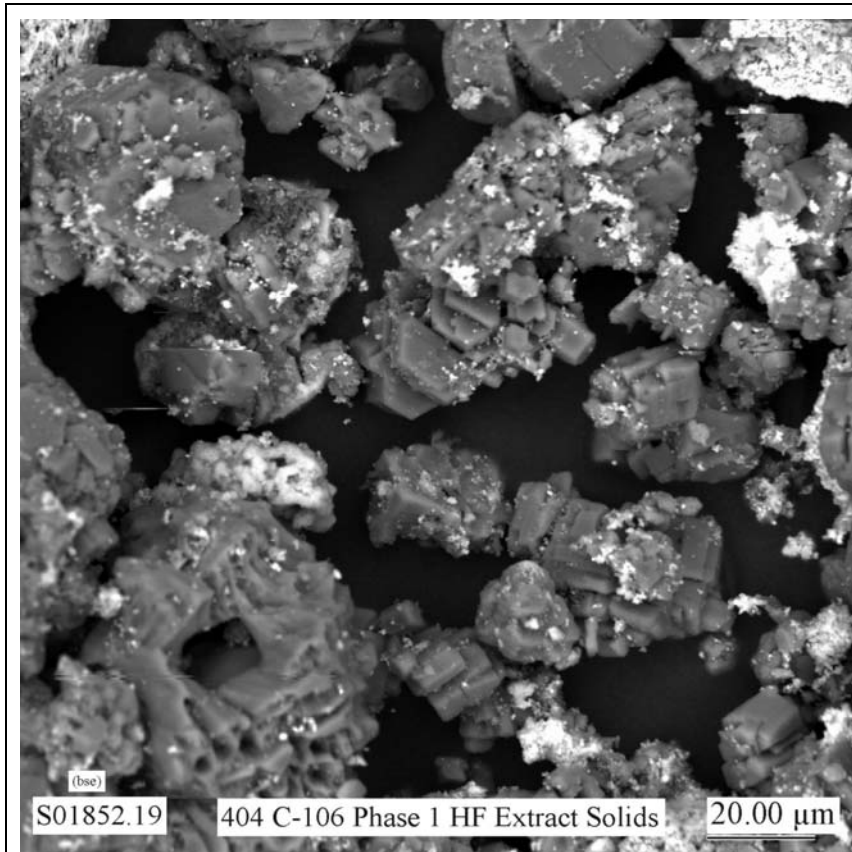
Figure D.19. Micrograph Showing Morphologies of Typical Particles in Sample 404 of HF-Extracted Sludge
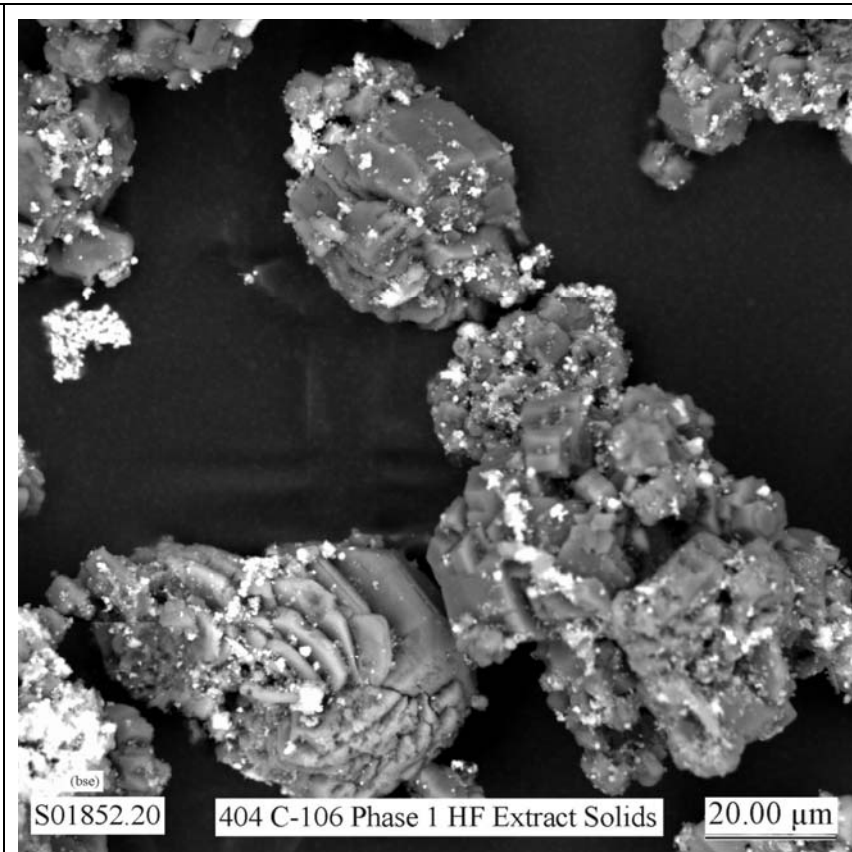
Figure D.20. Micrograph Showing Morphologies of Typical Particles in Sample 404 of HF-Extracted Sludge

C-106 404 Primary HF Extract
eds01 [See Figure D.3]

C-106 404 Primary HF Extract
eds02 [See Figure D.3]

C-106 404 Primary HF Extract
eds03 [See Figure D.3]

C-106 404 Primary HF Extract
eds04 [See Figure D.3]
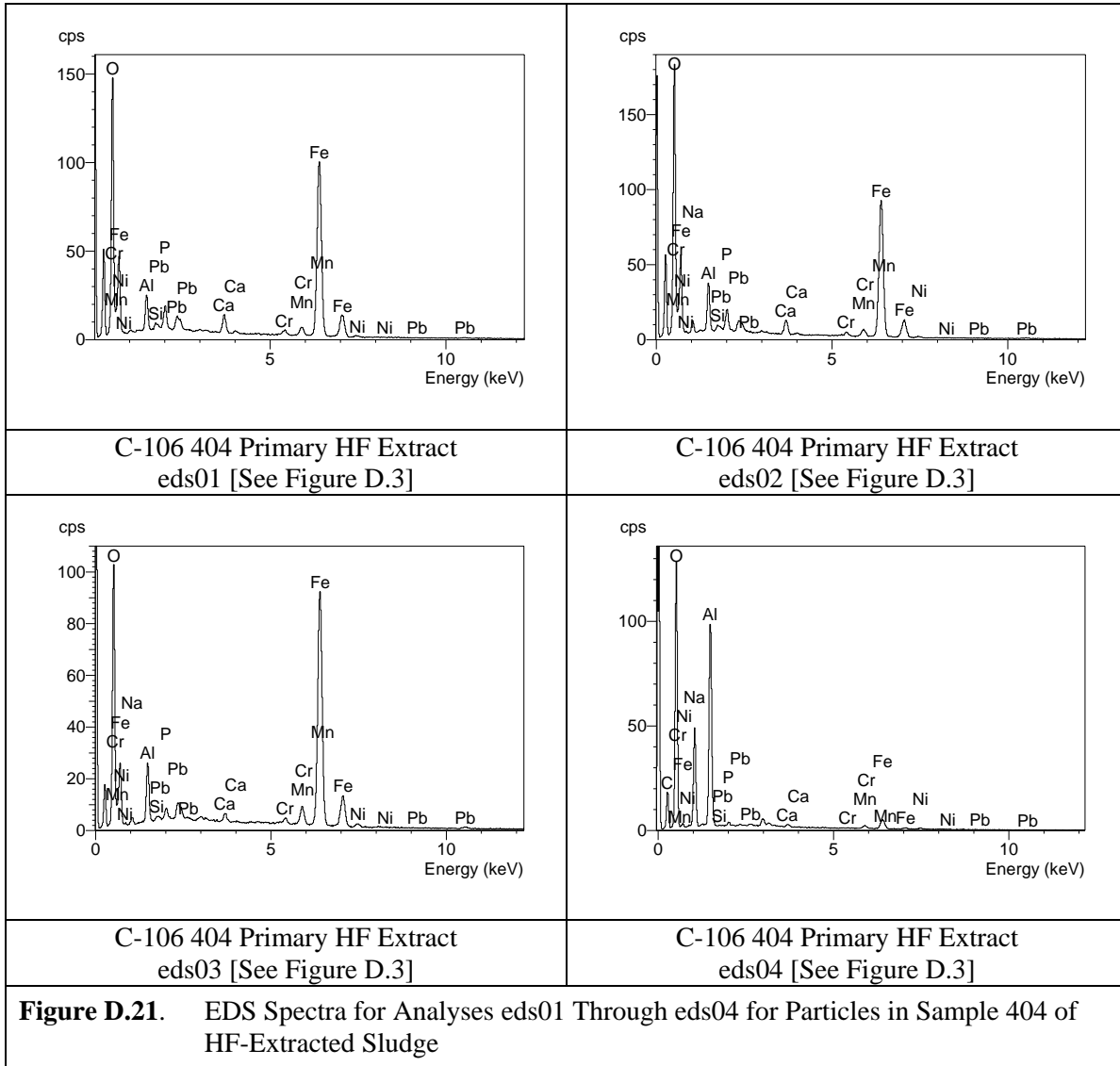
Figure D.21.   EDS Spectra for Analyses eds01 Through eds04 for Particles in Sample 404 of HF-Extracted Sludge

Figure D.22. EDS Spectra for Analyses eds05 Through eds08 for Particles in Sample 404 of HF-Extracted Sludge

C-106 404 Primary HF Extract
eds09 [See Figure D.6]

C-106 404 Primary HF Extract
eds10 [See Figure D.7]

C-106 404 Primary HF Extract
eds11 [See Figure D.9]

C-106 404 Primary HF Extract
eds12 [See Figure D.9]

Figure D.23.   EDS Spectra for Analyses eds09 Through eds12 for Particles in Sample 404 of HF-Extracted Sludge

| C-106 404 Primary HF Extract eds13 [See Figure D.10] | C-106 404 Primary HF Extract eds14 [See Figure D.11] |
| C-106 404 Primary HF Extract eds15 [See Figure D.12] | C-106 404 Primary HF Extract eds16 [See Figure D.13] |

Figure D.24. EDS Spectra for Analyses eds13 Through eds15 for Particles in Sample 404 of HF-Extracted Sludge

D.15

| C-106 404 Primary HF Extract eds17 [See Figure D.16] | C-106 404 Primary HF Extract eds18 [See Figure D.14] |

| C-106 404 Primary HF Extract eds19 [See Figure D.18] | |

Figure D.25.   EDS Spectra for Analyses eds13 Through eds15 for Particles in Sample 404 of HF-Extracted Sludge

Figure D.26. Low Magnification SEM Micrograph Showing General Morphologies of Particles in Sample 405 of HF-Extracted Sludge
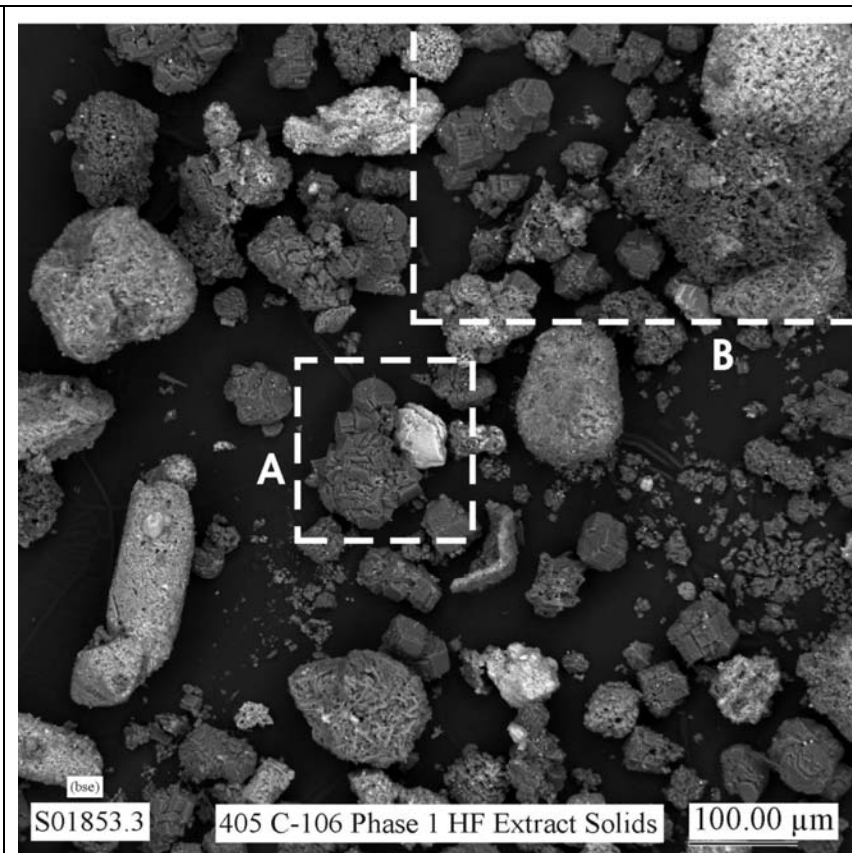
Figure D.27. Low Magnification SEM Micrograph Showing General Morphologies of Particles in Sample 405 of HF-Extracted Sludge
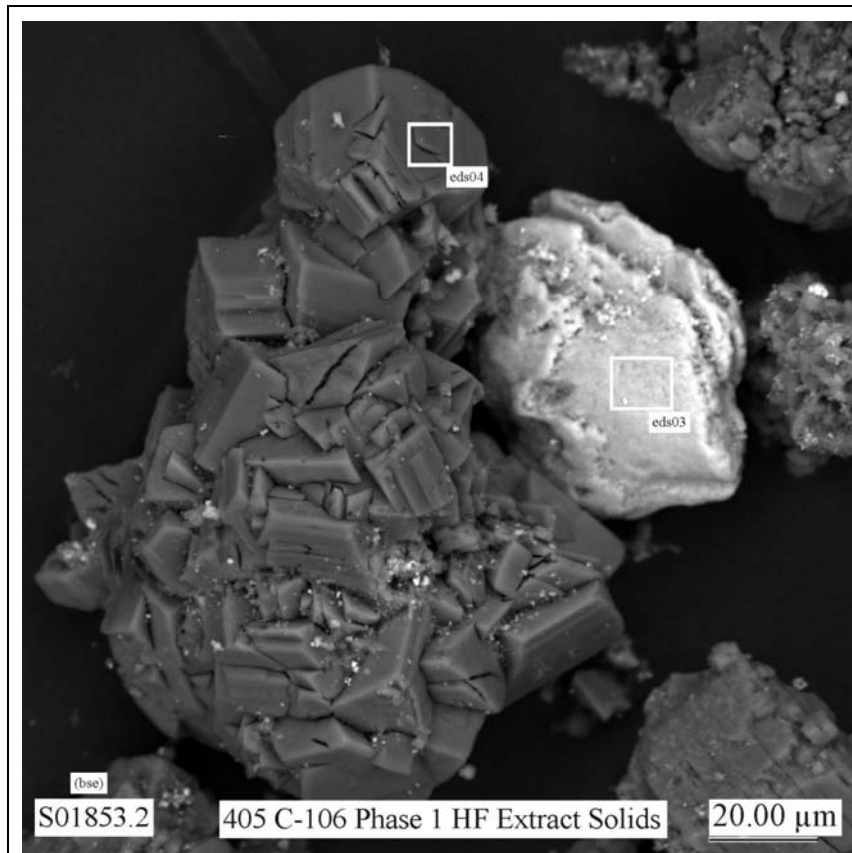
Figure D.28. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled A in Figure D.27
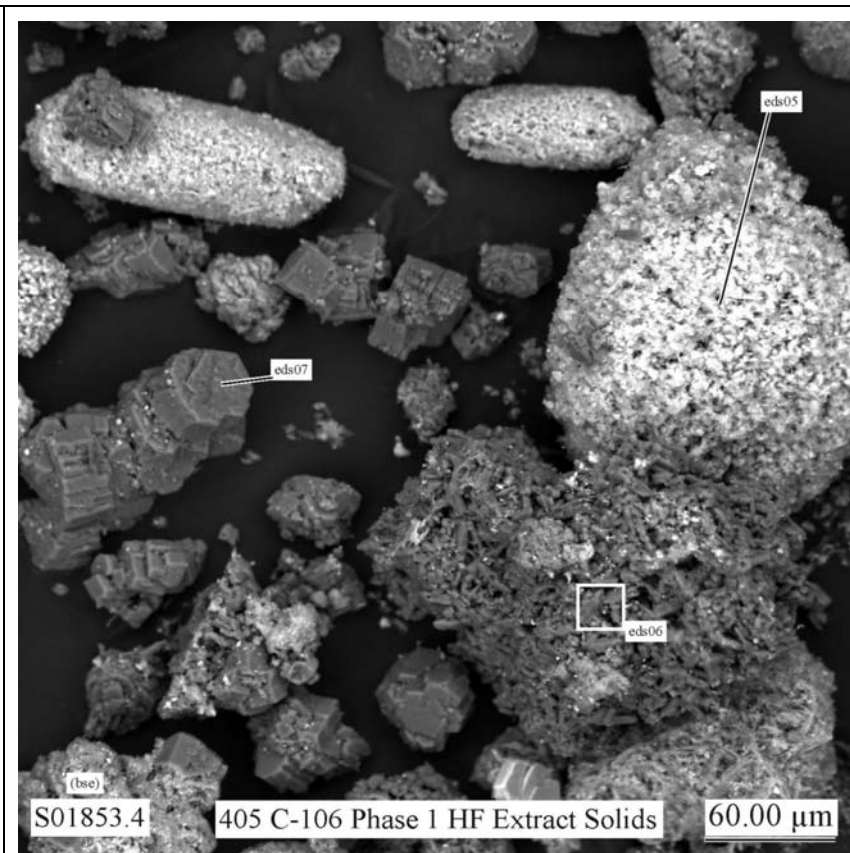
Figure D.29. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square Labeled B in Figure D.27
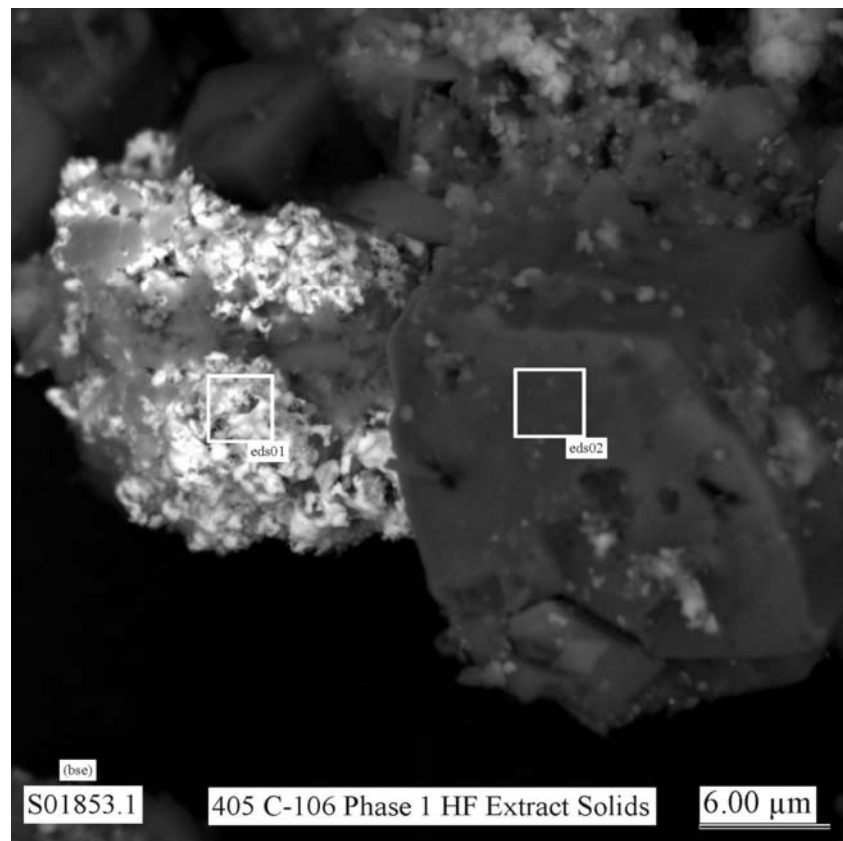
Figure D.30. Micrograph Showing Morphologies of Typical Particles in Sample 405 of HF-Extracted Sludge
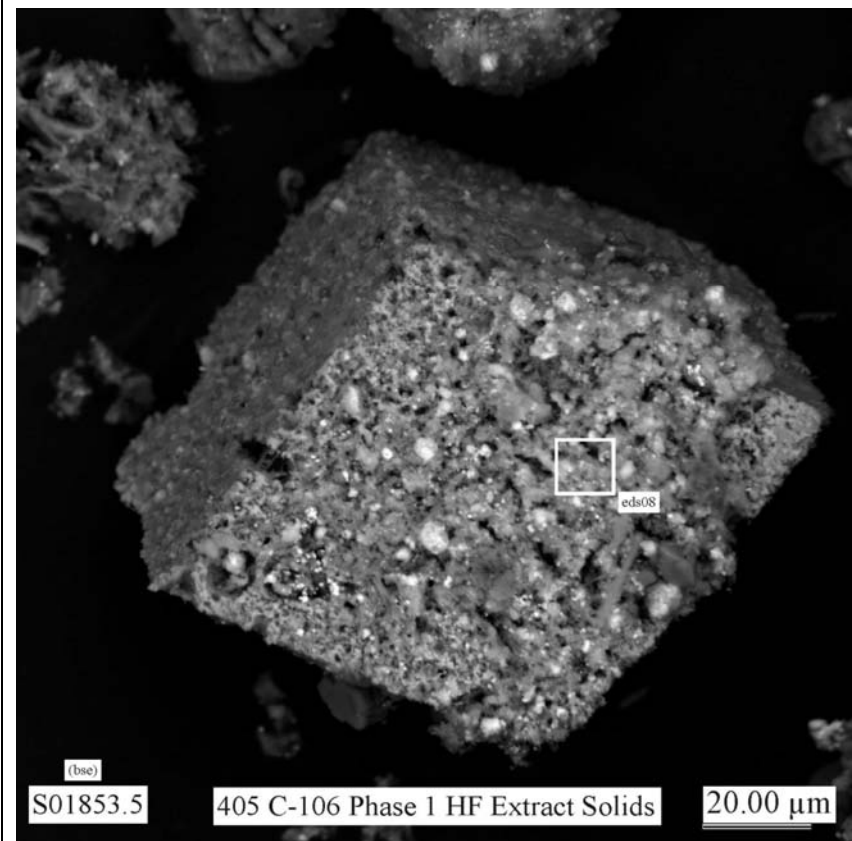
Figure D.31. Micrograph Showing Morphologies of Typical Particles in Sample 405 of HF-Extracted Sludge
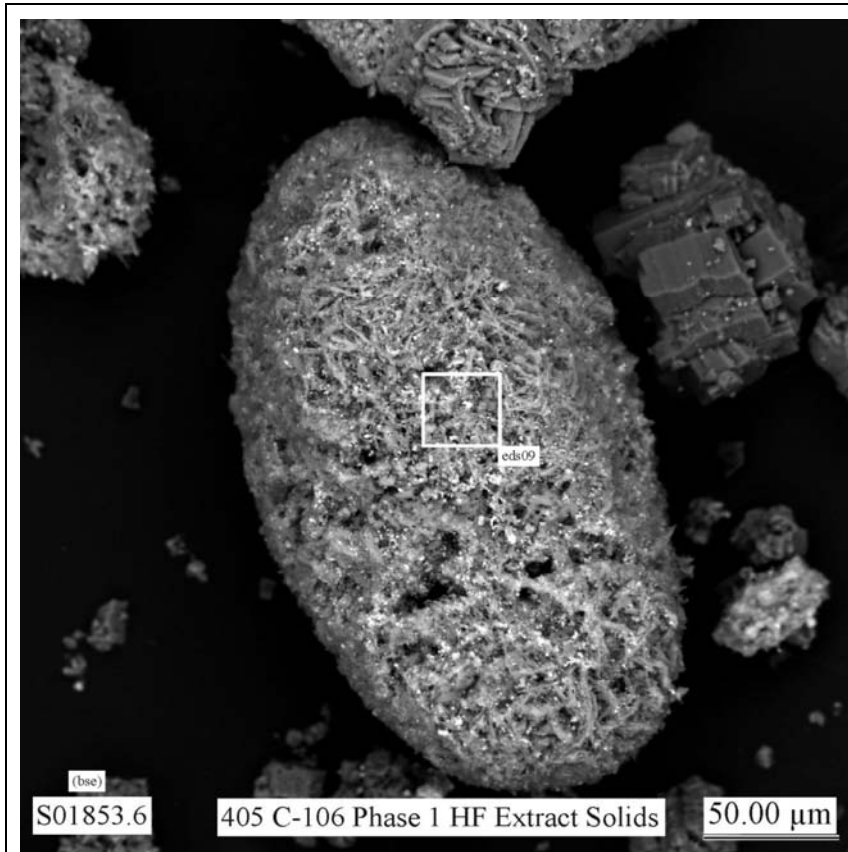
Figure D.32. Micrograph Showing Morphologies of Typical Particles in Sample 405 of HF-Extracted Sludge
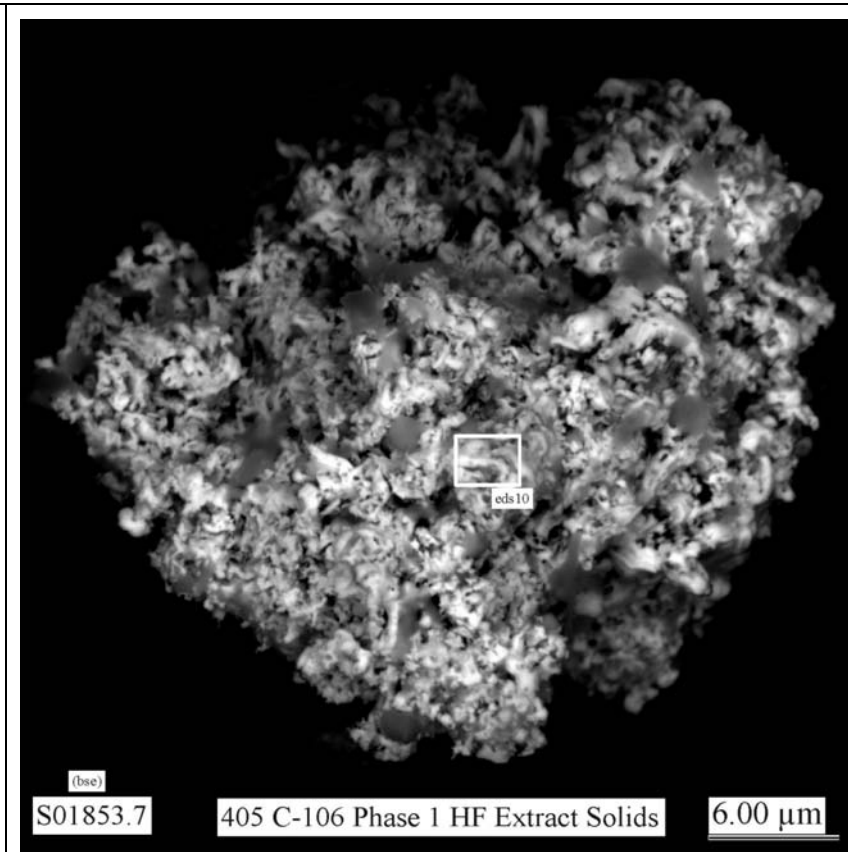
Figure D.33. Micrograph Showing Morphologies of Typical Particles in Sample 405 of HF-Extracted Sludge
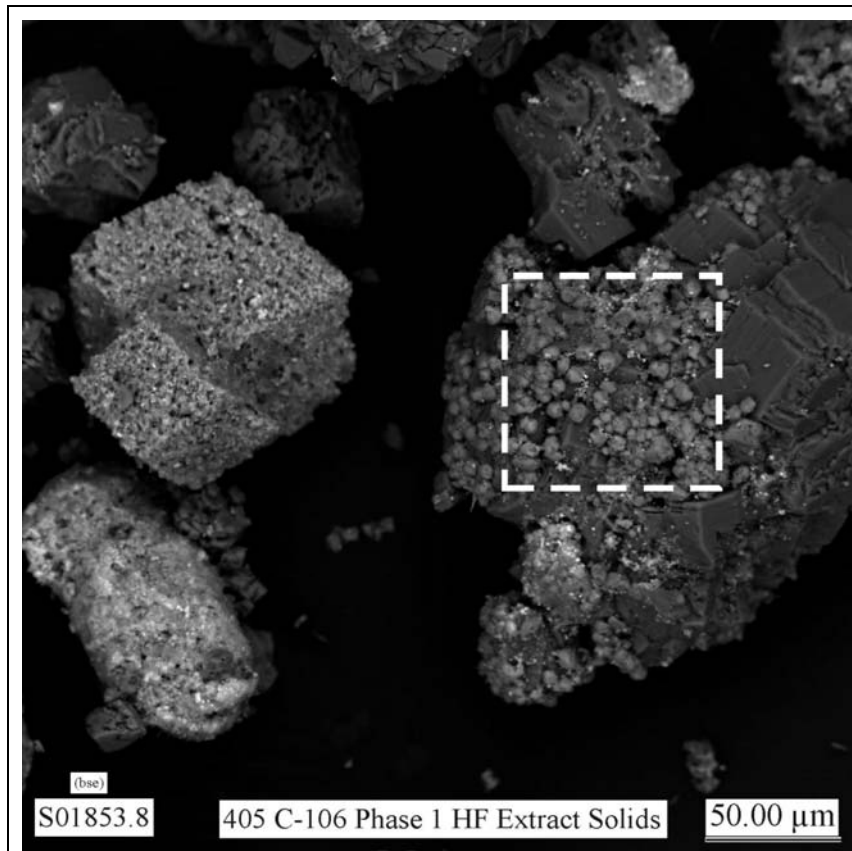
Figure D.34. Micrograph Showing Morphologies of Typical Particles in Sample 405 of HF-Extracted Sludge
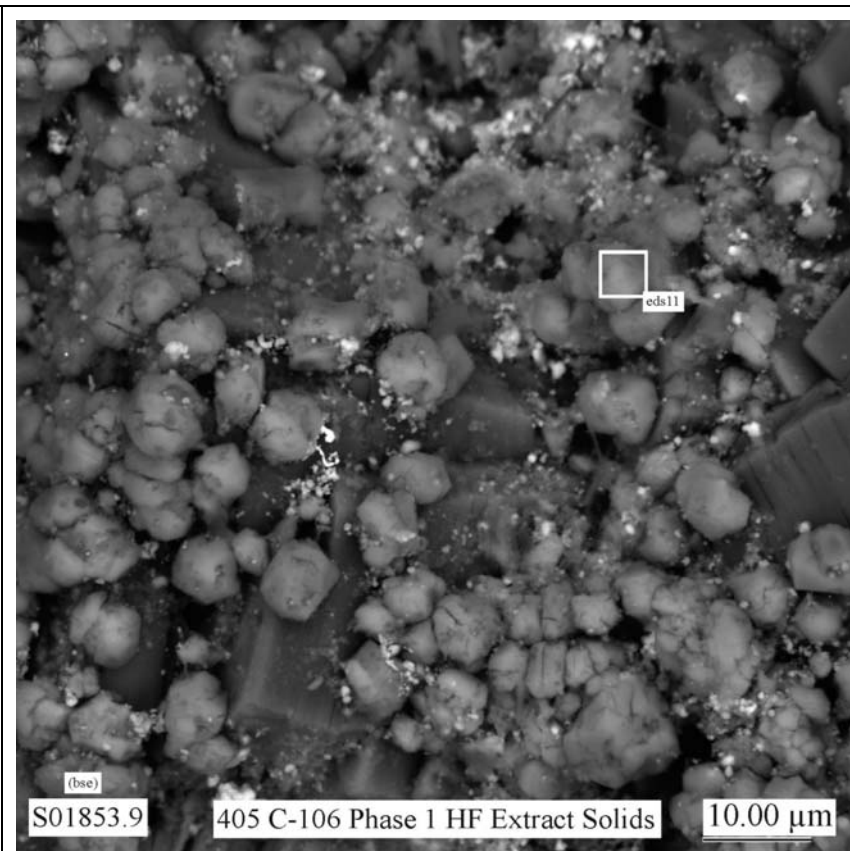
Figure D.35. Micrograph Showing at Higher Magnification the Crystals Coating the Area Indicated by the White Dashed-Line Square in Figure D.34
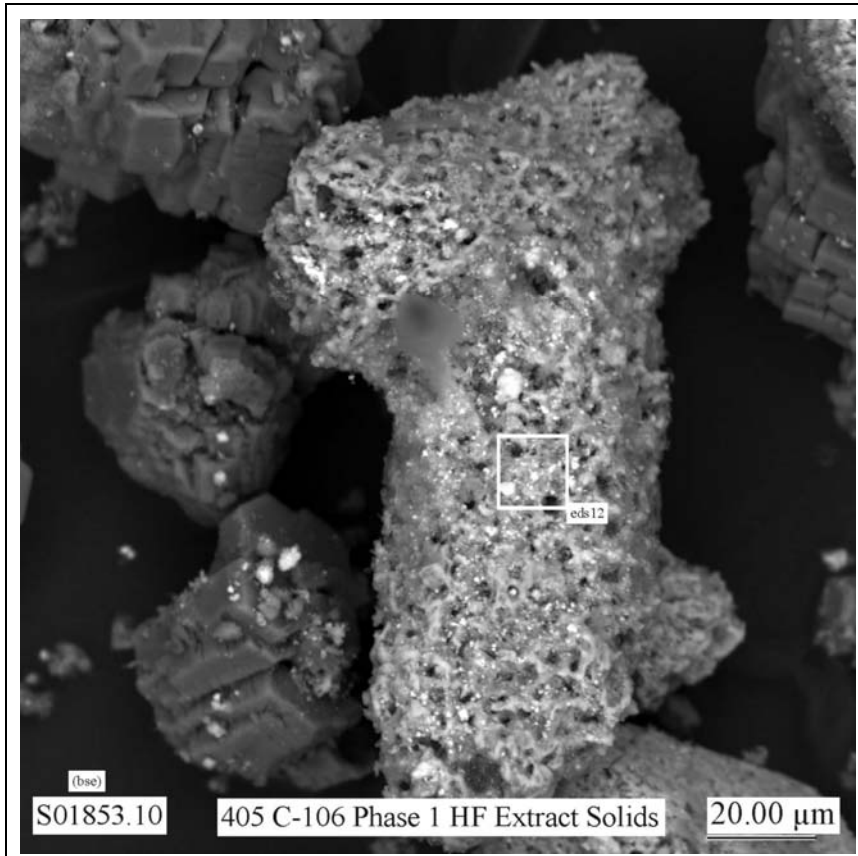
| **Figure D.36.** Micrograph Showing Morphologies of Typical Particles in Sample 405 of HF-Extracted Sludge | **Figure D.37.** Micrograph Showing Morphologies of Typical Particles in Sample 405 of HF-Extracted Sludge |
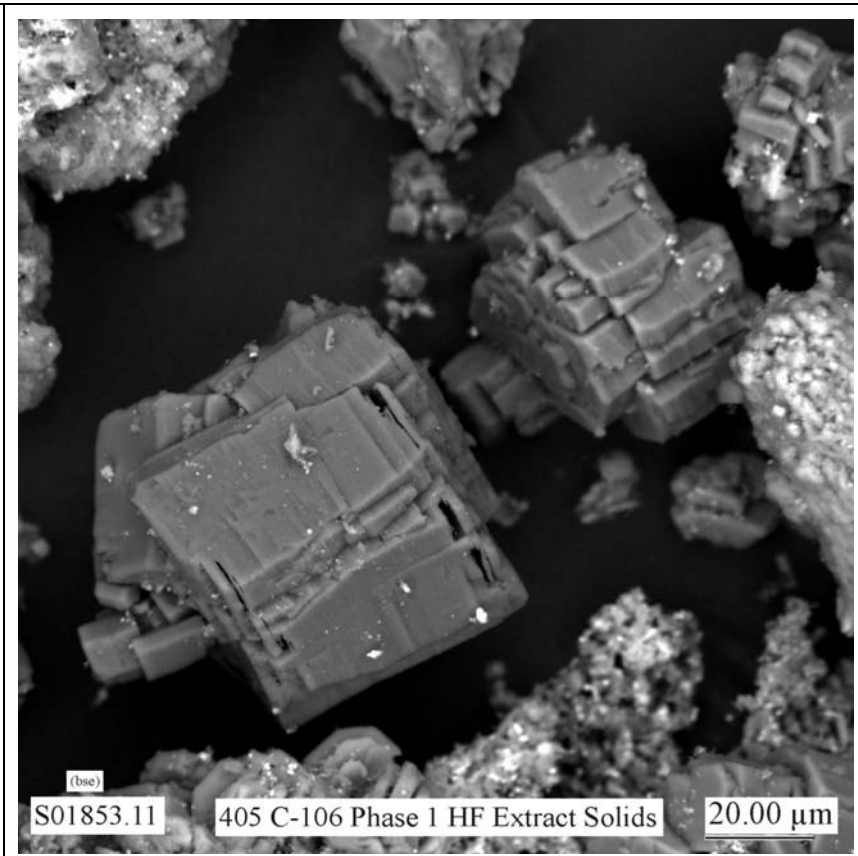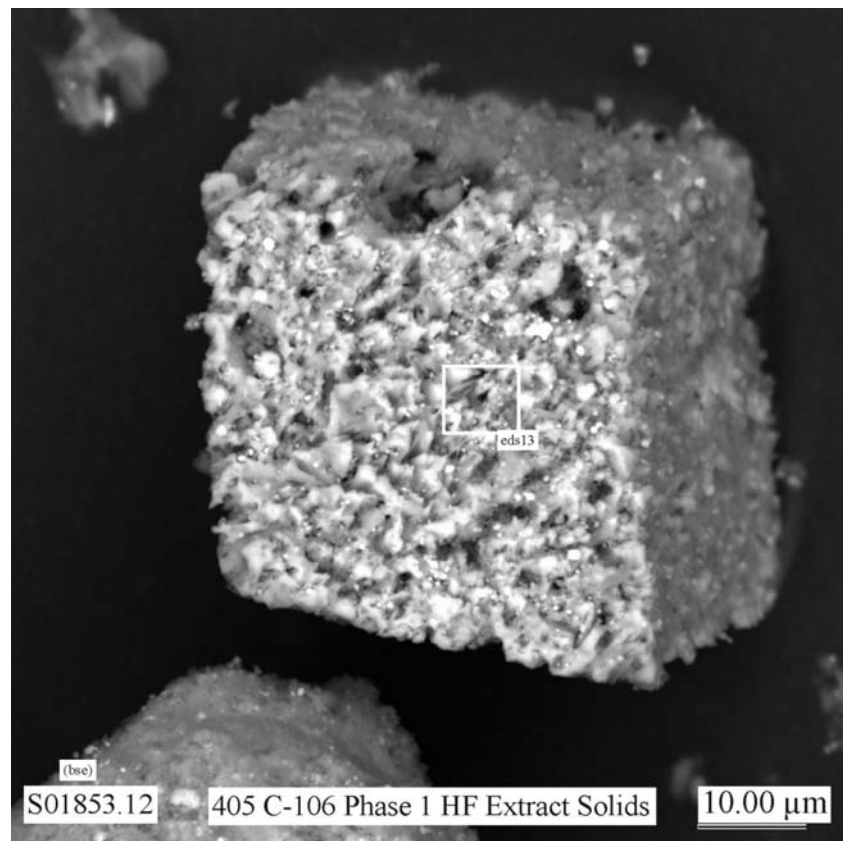
Figure D.38. Micrograph Showing Morphologies of Typical Particles in Sample 405 of HF-Extracted Sludge
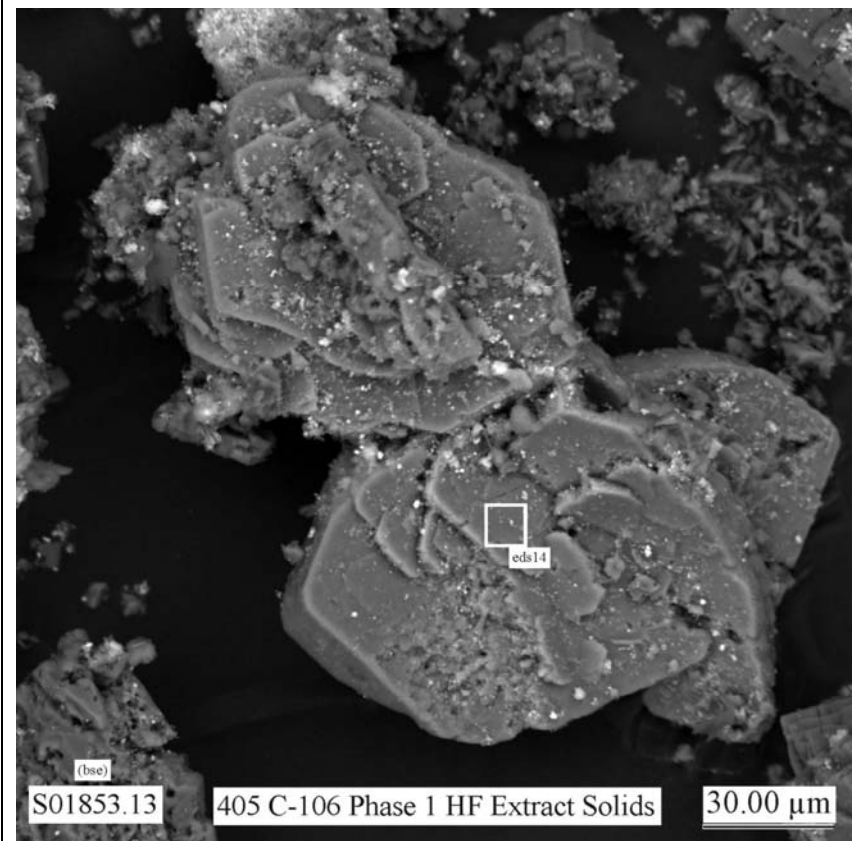
Figure D.39. Micrograph Showing Morphologies of Typical Particles in Sample 405 of HF-Extracted Sludge
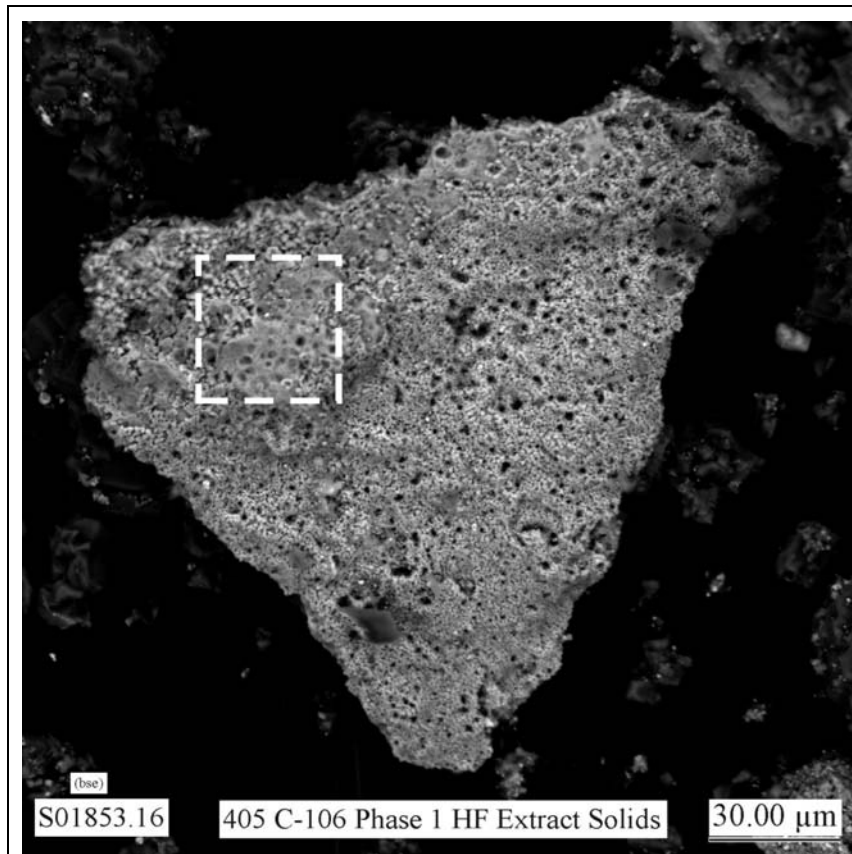
Figure D.40. Micrograph Showing Surface Morphology of a Fe-Rich Particle in Sample 405 of HF-Extracted Sludge
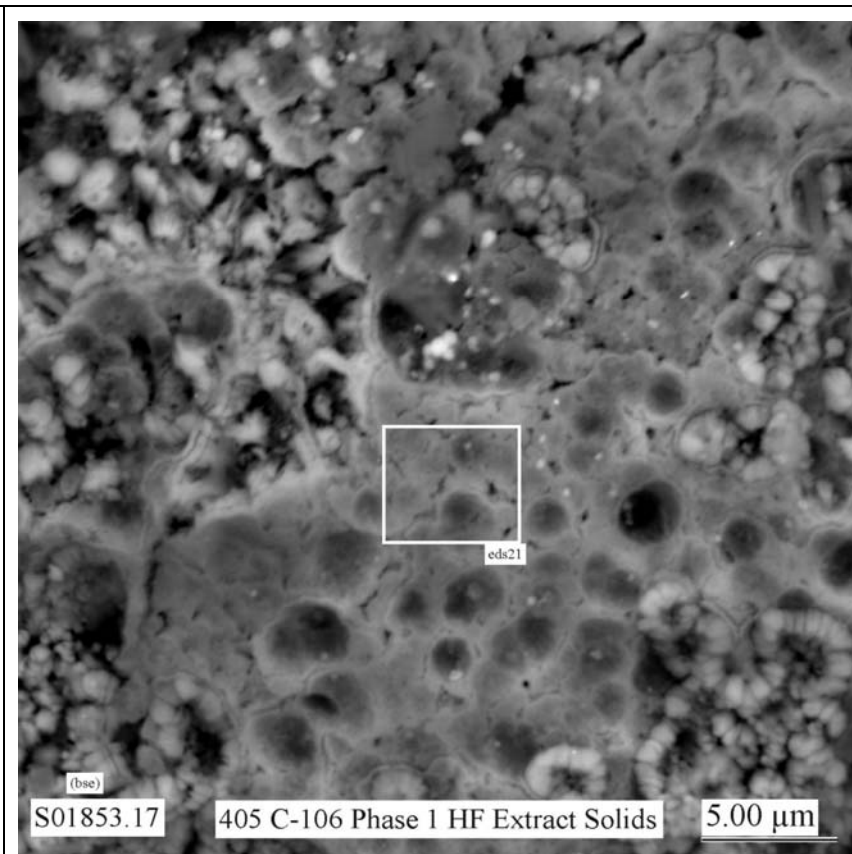
Figure D.41. Micrograph Showing at Higher Magnification the Area Indicated by the White Dashed-Line Square in Figure D.40
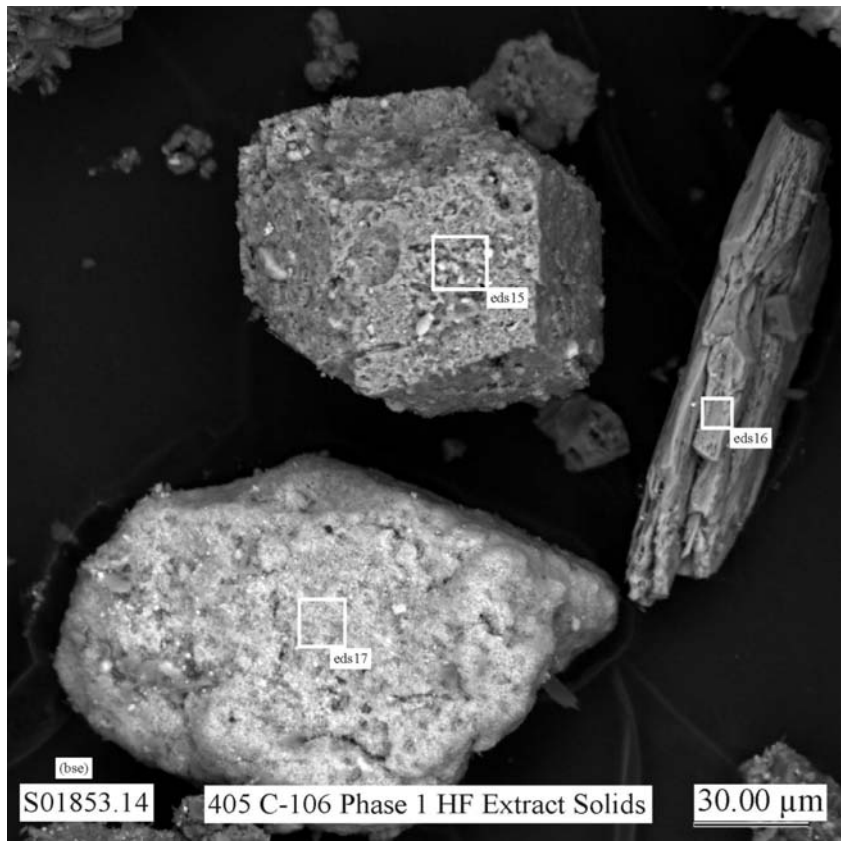
Figure D.42. Micrograph Showing Morphologies of Typical Particles in Sample 405 of HF-Extracted Sludge
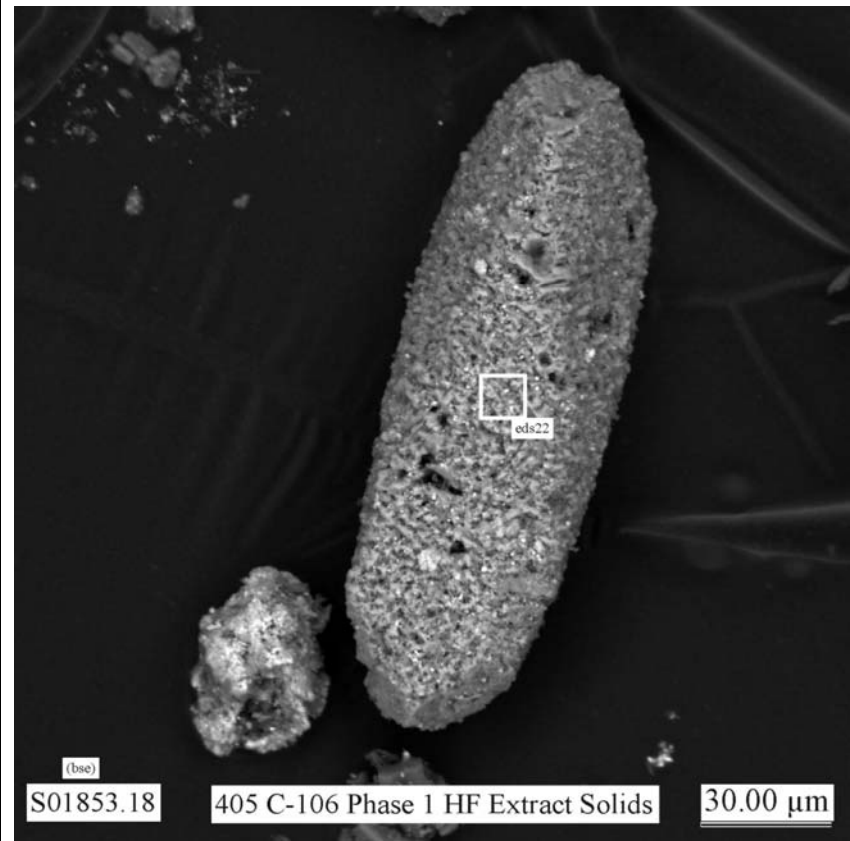
Figure D.43. Micrograph Showing Morphologies of Typical Particles in Sample 405 of HF-Extracted Sludge

| **Figure D.44.** | Micrograph Showing Morphology of an Unusual Rare Earth-Phosphorous Rich Particle in Sample 405 of HF-Extracted Sludge | **Figure D.45.** | Micrograph Showing Morphology of an Unusual Fe-Rich Particle in Sample 405 of HF-Extracted Sludge |

| C-106 405 Duplicate HF Extract eds01 [See Figure D.30] | C-106 405 Duplicate HF Extract eds02 [See Figure D.30] |
| C-106 405 Duplicate HF Extract eds03 [See Figure D.28] | C-106 405 Duplicate HF Extract eds04 [See Figure D.28] |

Figure D.46.   EDS Spectra for Analyses eds01 Through eds04 for Particles in Sample 405 HF-Extracted Sludge

C-106 405 Duplicate HF Extract eds05 [See Figure D.29]	C-106 405 Duplicate HF Extract eds06 [See Figure D.29]
C-106 405 Duplicate HF Extract eds07 [See Figure D.29]	C-106 405 Duplicate HF Extract eds08 [See Figure D.31]

Figure D.47.   EDS Spectra for Analyses eds05 Through eds08 for Particles in Sample 405 HF-Extracted Sludge

C-106 405 Duplicate HF Extract eds09 [See Figure D.32]	C-106 405 Duplicate HF Extract eds10 [See Figure D.33]
C-106 405 Duplicate HF Extract eds11 [See Figure D.35]	C-106 405 Duplicate HF Extract eds12 [See Figure D.36]

Figure D.48.   EDS Spectra for Analyses eds09 Through eds12 for Particles in Sample 405 HF-
Extracted Sludge

| C-106 405 Duplicate HF Extract eds13 [See Figure D.38] | C-106 405 Duplicate HF Extract eds14 [See Figure D.39] |
| C-106 405 Duplicate HF Extract eds15 [See Figure D.42] | C-106 405 Duplicate HF Extract eds16 [See Figure D.42] |

Figure D.49.   EDS Spectra for Analyses eds13 Through eds16 for Particles in Sample 405 HF-Extracted Sludge

C-106 405 Duplicate HF Extract eds17 [See Figure D.42]	C-106 405 Duplicate HF Extract eds18 [See Figure D.26]
C-106 405 Duplicate HF Extract eds19 [See Figure D.26]	C-106 405 Duplicate HF Extract eds20 [See Figure D.26]

Figure D.50.   EDS Spectra for Analyses eds17 Through eds20 for Particles in Sample 405 HF-Extracted Sludge

| C-106 405 Duplicate HF Extract
eds21 [See Figure D.41] | C-106 405 Duplicate HF Extract
eds22 [See Figure D.43] |
| C-106 405 Duplicate HF Extract
eds23 [See Figure D.44] | C-106 405 Duplicate HF Extract
eds24 [See Figure D.45] |

Figure D.51   EDS Spectra Analyses of eds21 through eds24 Particles in Sample 405 of HF-Extracted Sludge

Appendix E

SEM/EDS Element Distribution Maps
for Samples of Tank C-106 Sludge

Appendix E

SEM/EDS Element Distribution Maps for Samples of Tank C-106 Sludge

This appendix includes the scanning electron microscope (SEM) micrographs and corresponding energy-dispersive X-ray spectrometry (EDS) element distribution maps 9Figures E1 through E.10) for a limited number of imaged areas of SEM mounts of unleached, 82-day water-leached, and hydrofluoric (HF)-extracted sludge samples from tank 241-C-106 (C-106). The operating conditions for the SEM and procedures used for mounting SEM samples are described in Section 2.4 of the main report. After the studies and analyses of tank C-106 sludge had been completed and presented in draft form, the instrument hardware and software for the JEOL JSM-840 SEM used for the SEM/EDS analyses (see micrographs and EDS spectra in Appendices B, C, and D) were upgraded to the INCAEnergy EDS System to automate the collection of EDS spectra over multi-micrometer-sized areas of an SEM-imaged sample. This upgrade permits the mapping of the spatial distributions over user-selected areas of the relative concentrations of any user-specified element detectable by EDS. The installed INCAEnergy EDS System was tested and demonstrated using the sludge samples from tank C-106.

The top of each figure in this appendix shows an SEM micrograph of the area of the sample mount that was scanned by EDS using the INCAEnergy EDS System. Each of the included SEM micrographs was imaged using backscattered electrons (BSE). A series of element distribution maps that show the spatial distributions of the relative concentrations for the indicated elements is included below each micrograph. The concentration of each listed element is directly proportional to the regions of brightness (i.e., brighter the area, the higher the concentration of the selected element) in the corresponding distribution map.
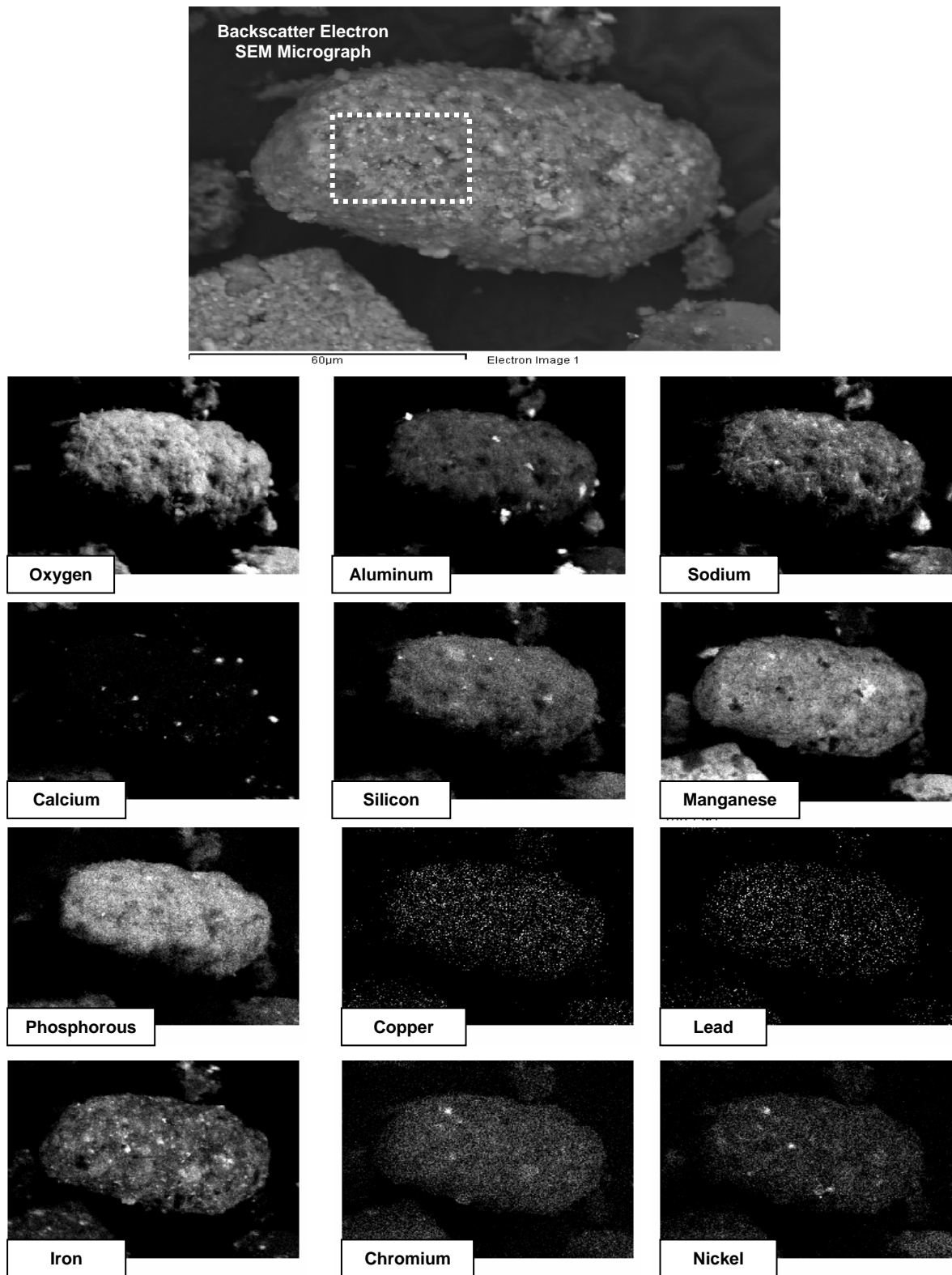
Figure E.1. SEM Micrograph and Element Distribution Maps for Particles in Unleached Sludge from Tank C-106
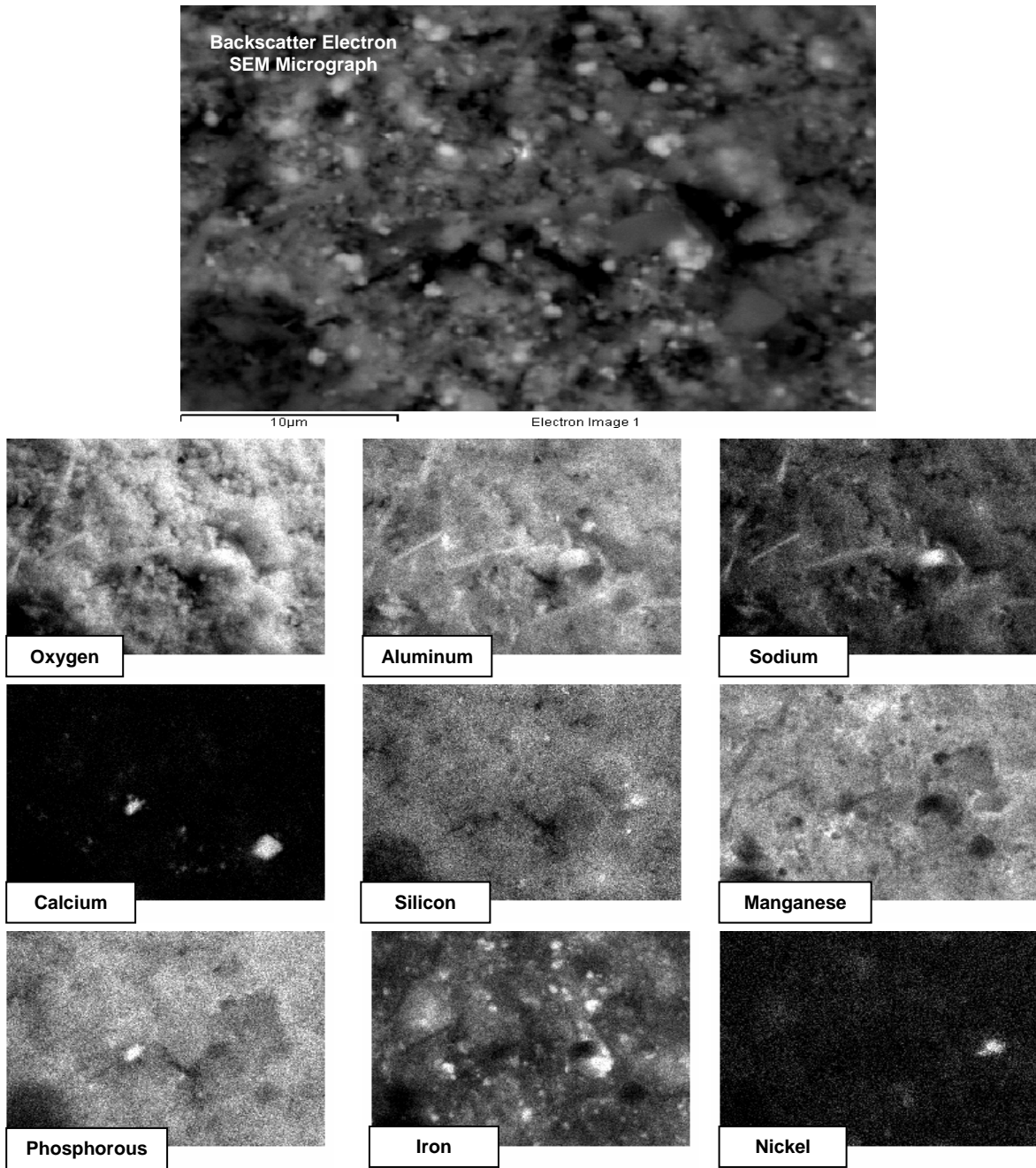
Figure E.2. SEM Micrograph and Element Distribution Maps for the Area of the Particle Indicated by the White Dashed-Line Rectangle in Figure E.1 of Unleached Sludge from Tank C-106
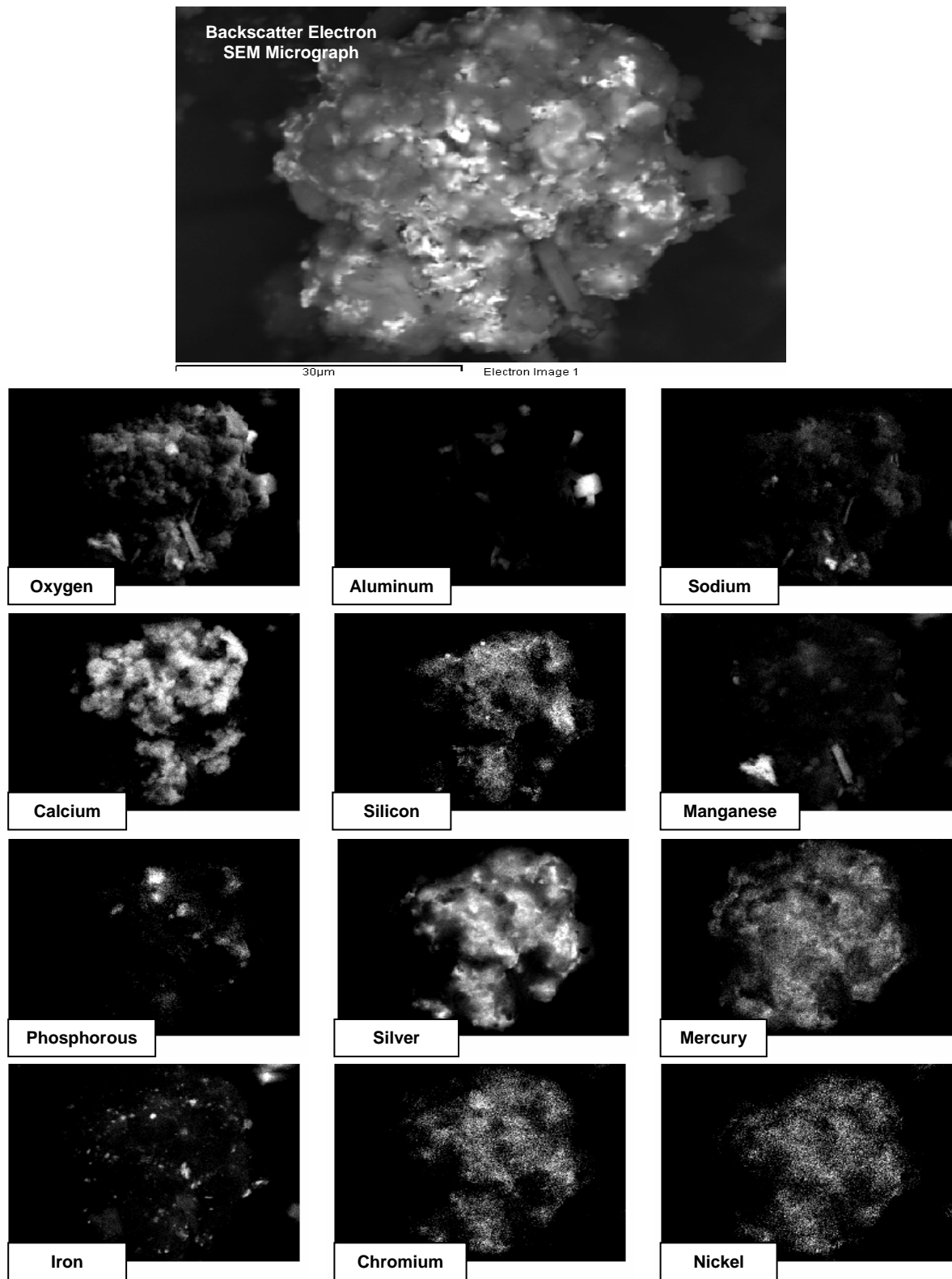
Figure E.3. SEM Micrograph and Element Distribution Maps for a Particle Aggregate in Unleached Sludge from Tank C-106
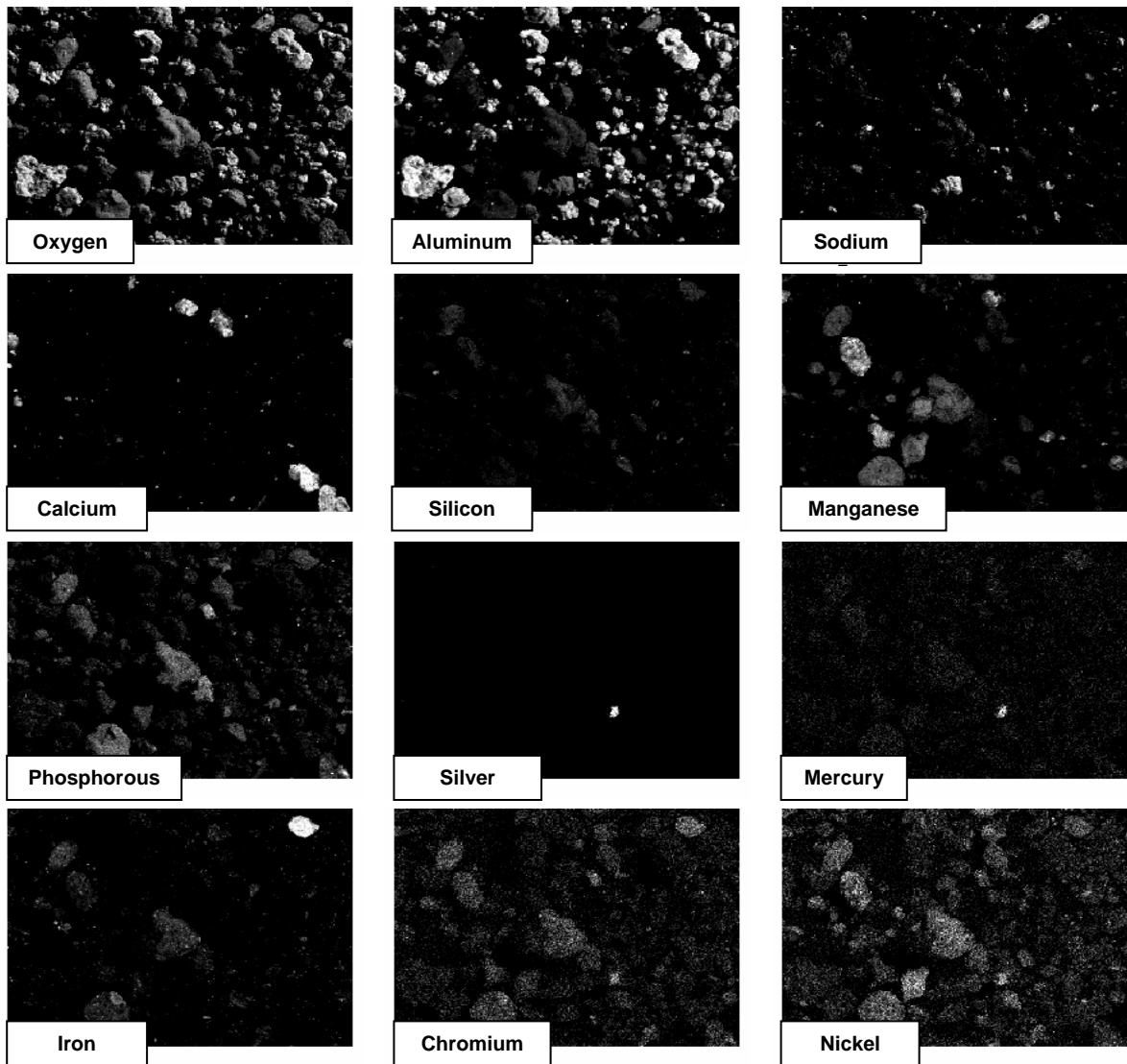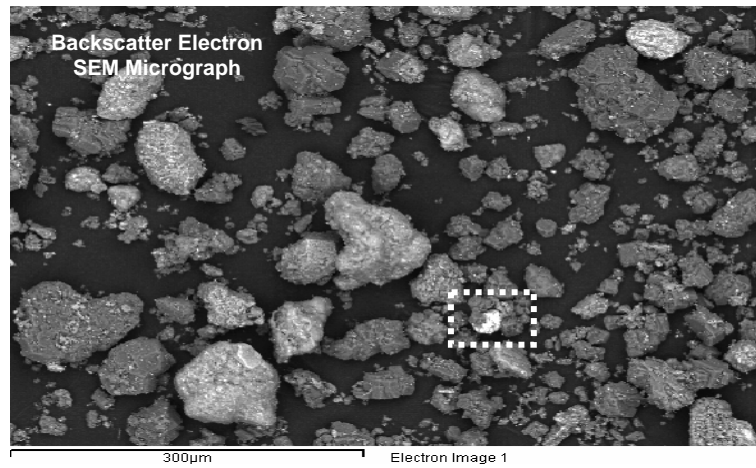
Figure E.4. Low Magnification SEM Micrograph and Element Distribution Maps for Particles in 82-Day Water-Leached Sludge from Tank C-106

Backscatter Electron SEM Micrograph

30μm   Electron Image 1

Oxygen

Aluminum

Sodium

Calcium

Silicon

Manganese

Phosphorous

Silver

Mercury

Iron

Chromium

Nickel
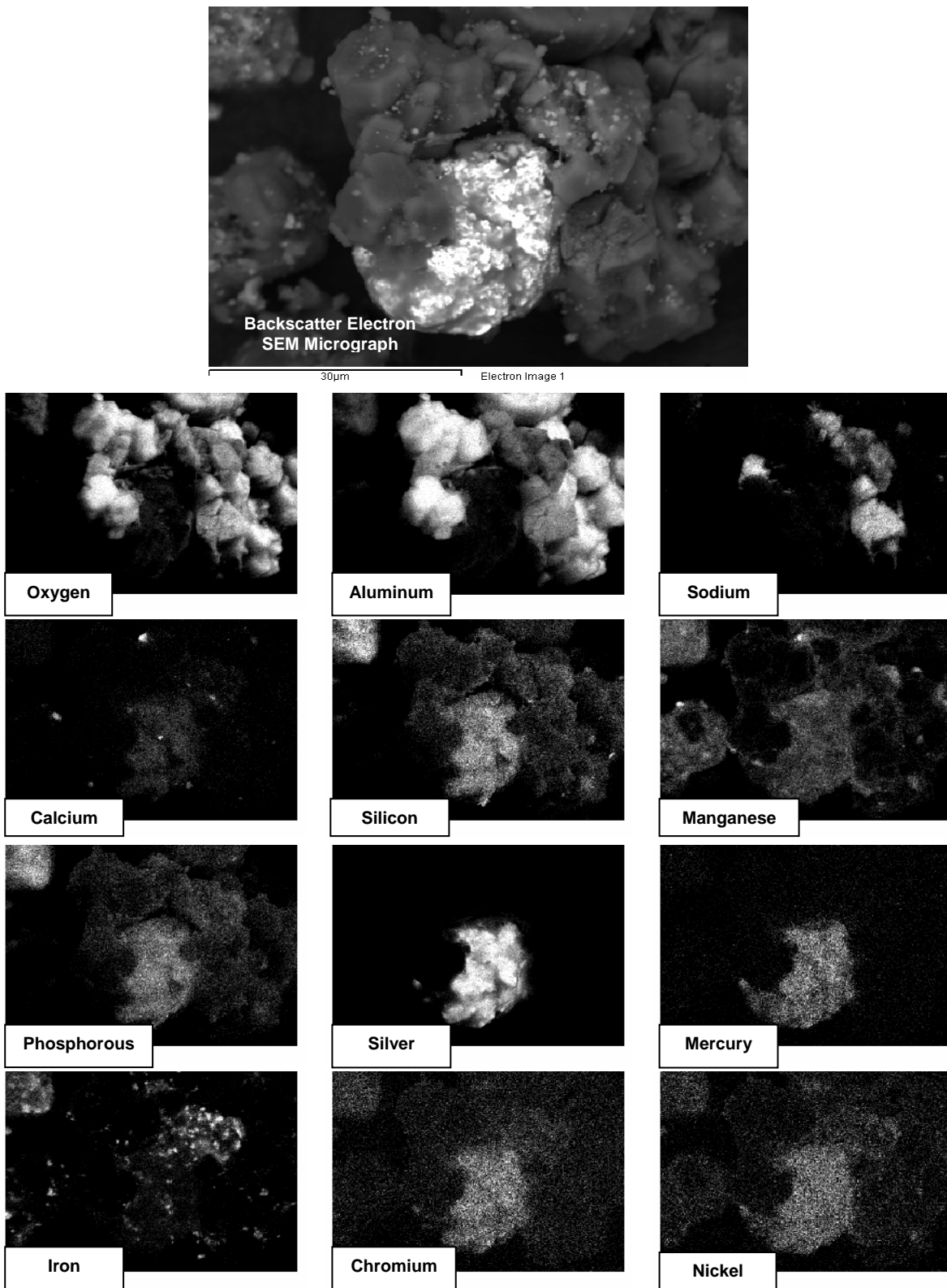
Figure E.5. SEM Micrograph and Element Distribution Maps for Particles in the Area Indicated by the White Dashed-Line Rectangle in Figure E.4 for 82-Day Water-Leached Sludge from Tank C-106

E.6
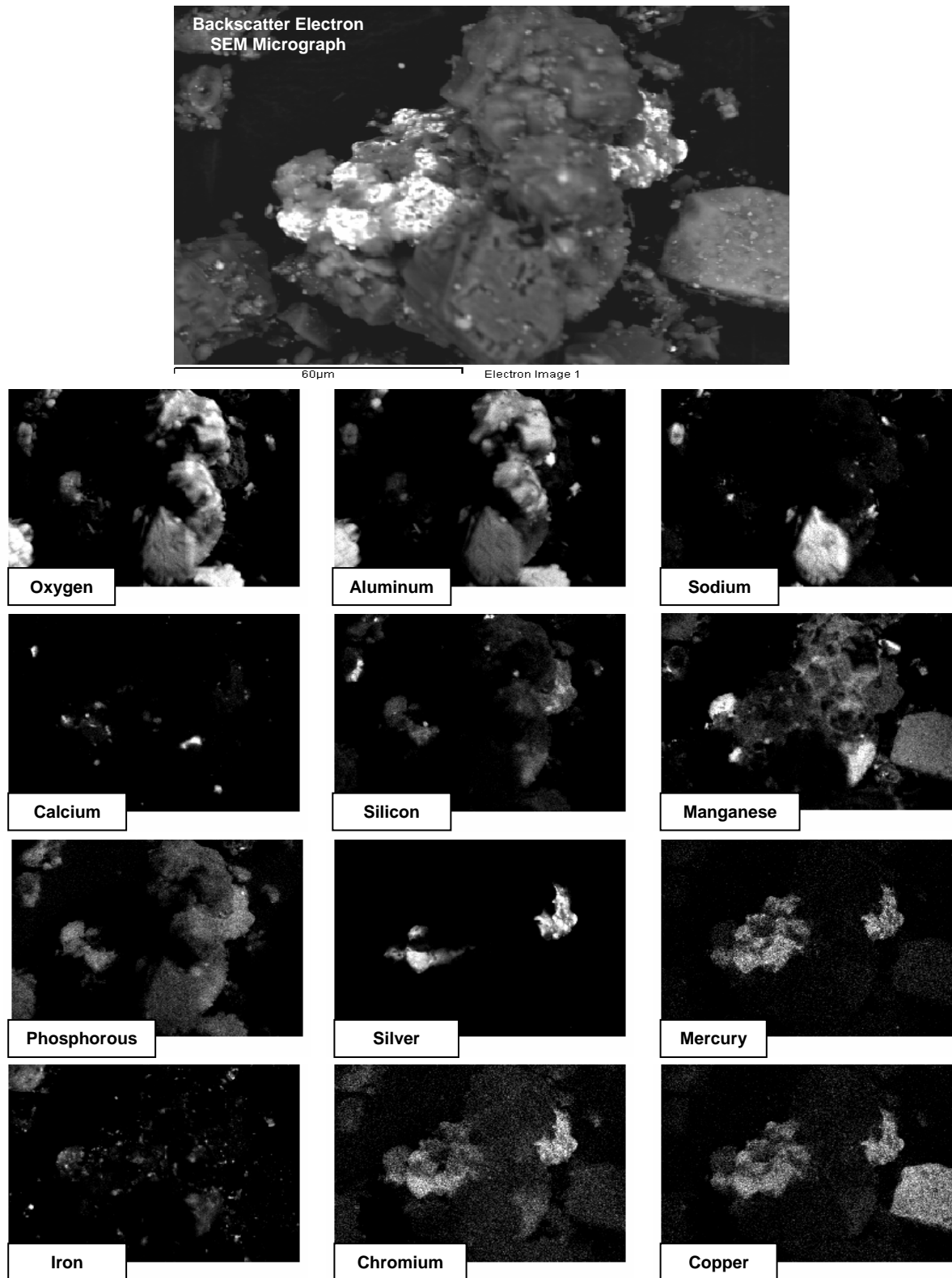
Figure E.6. SEM Micrograph and Element Distribution Maps for a Particle Aggregate in 82-Day Water-Leached Sludge from Tank C-106
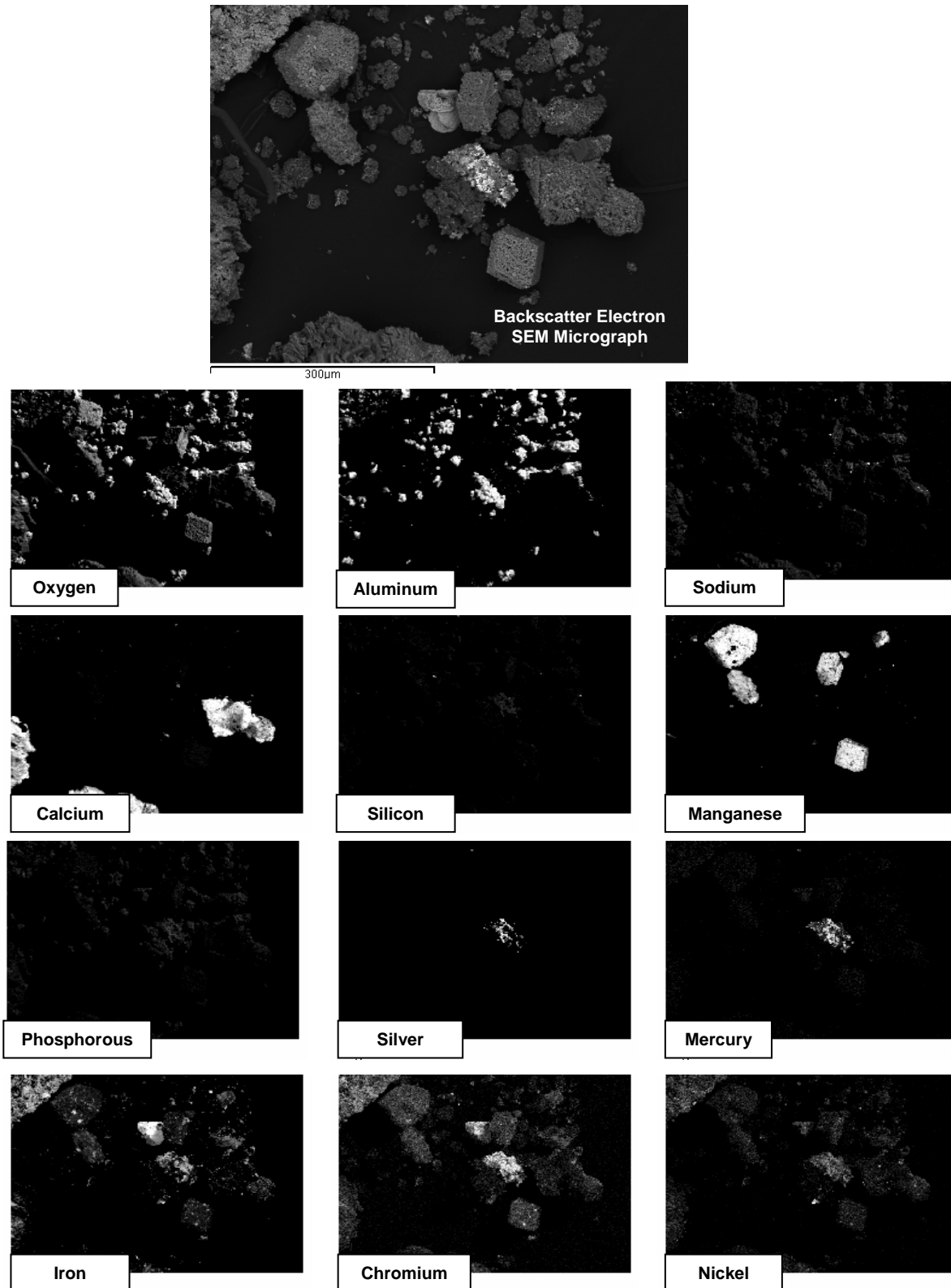
Figure E.7. Low Magnification SEM Micrograph and Element Distribution Maps for Particles in HF-Extracted Sludge from Tank C-106
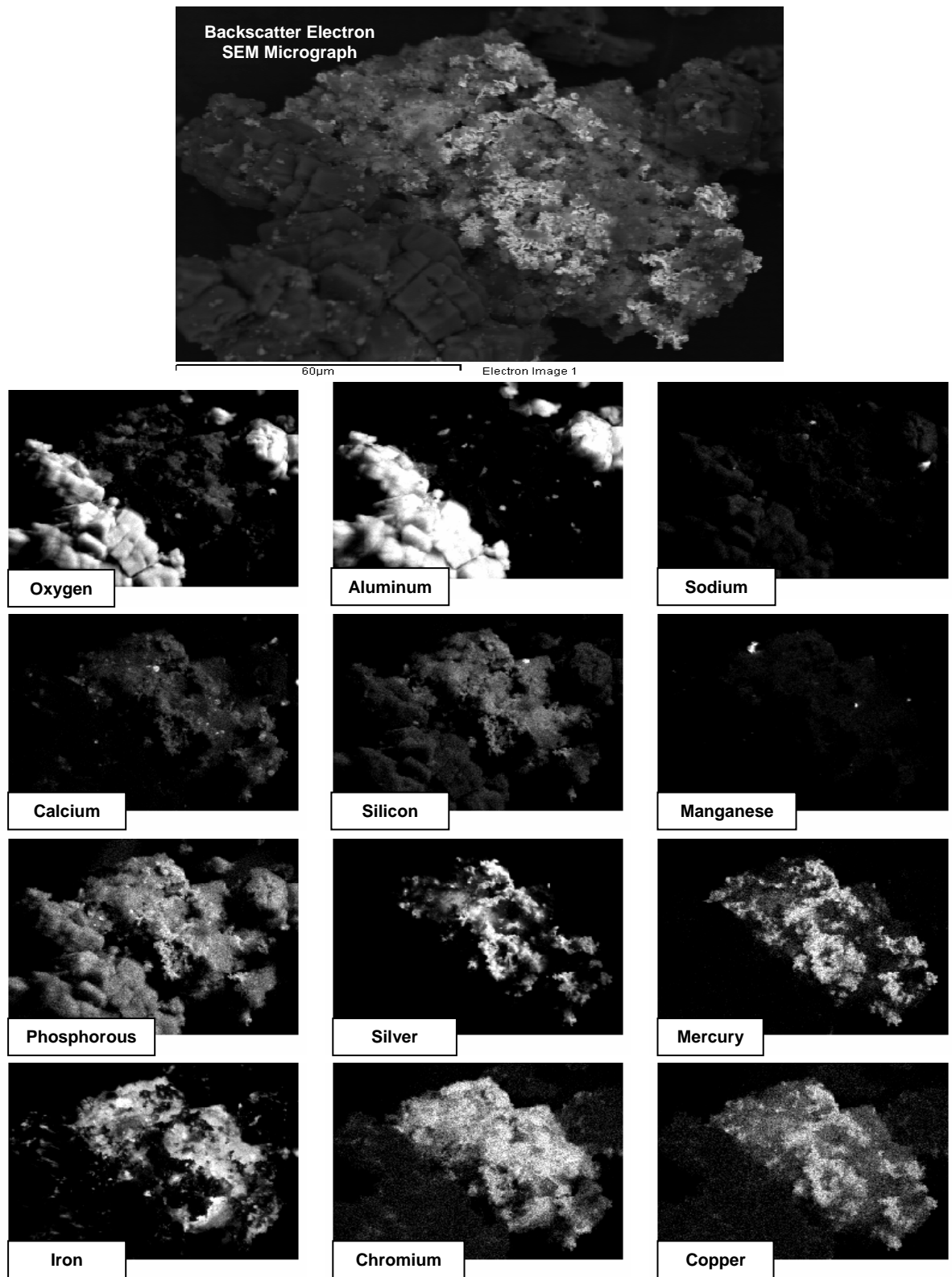
Figure E.8. SEM Micrograph and Element Distribution Maps for a Particle Aggregate in HF-Extracted Sludge from Tank C-106

Appendix F

Solution Concentrations of Tank C-106 Residual Liquid and Water Contact Tests with Residual Sludge

Appendix F

Solution Concentrations of Tank C-106 Residual Liquid and Water Contact Tests with Residual Sludge

This appendix provides the data (Tables F.1 through F.5) from water-leaching tests on sludge samples 404 and 405 from single-shell tank 241-C-106 (C-106) at the Hanford Site. These include results from the single-contact and periodic replenishment tests. Dissolved concentrations in extracts from these tests are provided on a per liter of solution basis.

Table F.1 Analysis of the Residual Liquid (Supernatant) Sample

	403 Liquid Avg	Sample Units
Al	1.21E+03	mg/L
Ba	(1.80)	mg/L
Ca	(1.30E+01)	mg/L
Fe	(1.27)	mg/L
Mg	(5.75E-01)	mg/L
Mn	(1.3E-01)	mg/L
Ni	3.48	mg/L
Pb	(1.44)	mg/L
Na	1.02E+04	mg/L
Fluoride	<1.28	mg/L
Chloride	2.05E+01	mg/L
N as Nitrate	9.42	mg/L
Carbonate	6.55E+03	mg/L
Sulfate	2.76E+01	mg/L
Oxalate	1.34E+03	mg/L
P as phosphate	7.49E+01	mg/L
^{90}Sr	1.44E-01	µCi/L
^{239}Pu	(4.7E-02)	µCi/L
^{237}Np	<5.00E-02	µCi/L
^{241}Am	<5.00E-02	µCi/L
^{129}I	1.64E-04	µCi/L
^{60}CO	<1.40E-01	µCi/L
^{137}CS	4.74E+02	µCi/L
TIC	2.80E+02	mg C/L
TOC	3.35E+02	mg C/L
Avg = Average.		
TIC = Total inorganic carbon.		
TOC = Total organic carbon.		

Table F.2. Solution Concentrations for Water-Leach Tests of Sample 404

Tank C-106 (404) Water Leach Results

Parameter	Units	Single Contact						Stage 1	Stage 1 (dup)	Stage 2	Stage 2 (dup)	Periodic Replenishment Tests		Stage 4	Stage 4 (dup)	Stage 5	Stage 5 (dup)	Stage 6 A	Stage 6 B
		1 day	1 day (dup)	2 week	2 week (dup)	1 month	1 month (dup)	Stage 1	Stage 1 (dup)	Stage 2	Stage 2 (dup)	Stage 3	Stage 3 (dup)	Stage 4	Stage 4 (dup)	Stage 5	Stage 5 (dup)	Stage 6 A	Stage 6 B
pH	std units	7.06	7.00			7.00	7.34	7.06	7.00	6.80	6.94	6.84	7.02	6.77	6.72	6.74	6.71	7.43	7.62
EC	mS/cm	0.464	0.517			0.819	0.455	0.464	0.517	0.149	0.149	0.123	0.128	0.082	0.096	0.063	0.062	0.161	0.197
Alkalinity	mg/L as CaCO3	307.44	366.00			127.37	109.80	307.44	366.00	<250.00	<250.00	<250.00	<250.00	<250.00	<250.00	<250.00	<250.00	95.16	113.46
TIC	mg C/L	19.98	13.74	15.21	17.10	9.12	8.10	19.98	13.74	4.18	3.55	7.07	10.78	16.71	3.91	4.29	9.30		
TOC	mg C/L	62.52	73.20	94.68	80.85	86.46	75.42	62.52	73.20	38.29	37.88	26.64	20.24	3.62	14.35	10.97	4.80		
TC	mg C/L	82.50	86.94	109.89	97.95	95.58	83.52	82.50	86.94	42.47	41.43	33.71	31.01	20.33	18.26	15.26	14.10		
Radionuclides																			
Sr^{90}	µCi/L	3.55E+02	3.89E+02					3.55E+02	3.89E+02	6.80E+02	5.46E+02	8.93E+02	7.78E+02	7.18E+02	7.96E+02	8.23E+02	6.88E+02		
^{99}Tc	mg/L	1.67E-04	1.47E-04	2.90E-04	2.30E-04	2.61E-04	2.18E-04	1.67E-04	1.47E-04	9.00E-06	2.70E-05	1.80E-05	(9.00E-06)	(3.00E-05)	(3.00E-05)	(3.00E-05)	(3.00E-05)	2.77E-04	3.08E-04
^{238}U	mg/L	5.41E-02	5.70E-02	3.29E-02	2.37E-02	2.70E-02	2.06E-02	5.41E-02	5.70E-02	1.06E-02	1.02E-02	4.50E-03	4.30E-03	2.19E-03	1.44E-03	8.20E-04	6.48E-04	1.56E-03	1.35E-03
^{129}I	mg/L	(3.50E-05)	(5.00E-06)	<2.50E-04	(7.00E-06)	(5.00E-06)	(5.20E-05)	(3.50E-05)	(5.00E-06)									(3.00E-05)	
^{239}Pu	µCi/L	(3.47E-03)	(2.58E-03)			(6.46E-04)	(7.23E-04)	(3.47E-03)	(2.58E-03)	(1.30E-03)	(8.06E-04)	(1.09E-03)	(5.89E-04)	(1.02E-03)	(1.18E-03)	(9.61E-04)	(6.51E-04)	(9.92E-04)	(4.34E-04)
^{237}Np	µCi/L	4.13E-03	4.30E-03			3.97E-03	3.49E-03	4.13E-03	4.30E-03	1.70E-03	1.71E-03	1.11E-04	1.08E-04	6.72E-05	5.98E-05	4.39E-05	3.80E-05	4.82E-04	5.25E-04
^{241}Am	µCi/L	1.82E-02	2.22E-02			4.16E-03	(3.32E-03)	1.82E-02	2.22E-02	(8.29E-03)	(7.34E-03)	<1.70E-02	<1.70E-02	<1.70E-02	<1.70E-02	<1.70E-02	<1.70E-02	<1.70E-02	<1.70E-02
Metals																			
Ag 107	mg/L	5.93E-03	1.15E-02			4.78E-03	2.83E-03	5.93E-03	1.15E-02	(5.90E-04)	(7.10E-04)	(3.60E-04)	(4.70E-04)	(3.00E-04)	(4.40E-04)	(4.30E-04)	(4.70E-04)	5.71E-04	
Ag 109	mg/L	5.58E-03	1.05E-02			4.41E-03	2.18E-03	5.58E-03	1.05E-02	(6.00E-04)	(5.00E-04)	(4.60E-04)	(4.70E-04)	(2.20E-04)	(3.30E-04)	(3.60E-04)	(3.20E-04)	(4.67E-04)	
Al	mg/L	6.69E-01	7.71E-01	(2.37E-01)	(1.82E-01)	(9.86E-02)	(5.87E-02)	6.69E-01	7.71E-01	5.11E-01	5.45E-01	(4.66E-01)	5.10E-01	5.15E-01	(4.87E-01)	5.43E-01	5.11E-01	(3.42E-02)	(5.76E-01)
As	mg/L	(7.71E-02)	(5.95E-02)	<5.00E-01	<5.00E-01	<2.50E-01	<2.50E-01	(7.71E-02)	(5.95E-02)	<1.25E+00	(1.51E-01)	<1.25E+00	(3.35E-02)	<1.25E+00	(1.25E-01)	<1.25E+00	<1.25E+00	(2.57E-03)	(1.49E-02)
As 75	mg/L	(1.08E-02)	(3.96E-03)			(1.88E-03)	(2.01E-03)	(1.08E-02)	(3.96E-03)	(2.92E-03)	<5.00E-02	(9.70E-04)	(7.70E-04)	(2.95E-03)	(6.45E-03)	<5.00E-02	(2.89E-03)	(1.23E-03)	
B	mg/L	(3.62E-01)	(3.20E-01)	(1.31E-01)	(1.16E-01)	(6.56E-02)	(5.26E-02)	(3.62E-01)	(3.20E-01)	(2.87E-01)	(1.96E-01)	(2.37E-01)	(2.19E-01)	(2.04E-01)	(1.68E-01)	(2.03E-01)	(1.63E-01)	(4.81E-02)	(9.94E-02)
Ba	mg/L	3.13E-01	2.97E-01	8.15E-02	6.86E-02	1.08E-01	1.22E-01	3.13E-01	2.97E-01	2.47E-01	2.93E-01	4.20E-01	3.15E-01	3.15E-01	3.12E-01	(1.67E-01)	(1.92E-01)	1.17E-01	1.63E-01
Be	mg/L	(1.61E-02)	(1.04E-02)	(2.09E-03)	(1.88E-03)	(1.32E-03)	(9.78E-04)	(1.61E-02)	(1.04E-02)	(8.06E-03)	(7.19E-03)	(6.98E-03)	(5.80E-03)	(6.71E-03)	(6.30E-03)	(5.56E-03)	(6.47E-03)	(8.39E-04)	(1.05E-03)
Bi	mg/L	<1.25E+00	<1.25E+00	<1.25E+00	<1.25E+00	<2.50E-01	<2.50E-01	<1.25E+00	<1.25E+00	<1.25E+00	<1.25E+00	<1.25E+00	<1.25E+00	<1.25E+00	<1.25E+00	<1.25E+00	<2.50E-01	<2.50E-01	(1.70E-02)
Ca	mg/L	2.37E+00	3.31E+00	4.97E-01	5.50E-01	5.13E-01	5.49E-01	2.37E+00	3.31E+00	2.96E+00	2.65E+00	2.97E+00	2.75E+00	2.78E+00	2.99E+00	7.12E+00	3.30E+00	1.93E+00	(3.51E+00)
Cd	mg/L	(1.41E-02)	(1.01E-02)	(1.72E-02)	(1.73E-02)	1.49E-02	1.52E-02	(1.41E-02)	(1.01E-02)	(1.23E-02)	<2.50E-01	(6.20E-03)	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	(3.64E-03)	(4.73E-03)
Cd 111	mg/L	1.90E-02	1.97E-02			1.40E-02	1.47E-02	1.90E-02	1.97E-02	1.97E-02	1.77E-02	1.60E-02	1.41E-02	1.08E-02	(9.03E-03)	(7.84E-03)	(5.78E-03)	4.41E-03	
Cd 114	mg/L	1.78E-02	1.80E-02			1.40E-02	1.45E-02	1.78E-02	1.80E-02	1.79E-02	1.65E-02	1.52E-02	1.31E-02	(9.86E-03)	(7.87E-03)	(5.97E-03)	(5.37E-03)	3.94E-03	
Co	mg/L	<2.50E-01	<2.50E-01	(1.76E-02)	(1.54E-02)	(9.01E-03)	(8.18E-03)	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	(1.17E-03)	<1.25E-02
Cr	mg/L	(2.34E-02)	(8.12E-03)	(2.33E-02)	(2.08E-02)	1.95E-02	1.85E-02	(2.34E-02)	(8.12E-03)	<3.13E-01	<3.13E-01	(9.07E-03)	<3.13E-01	<3.13E-01	<3.13E-01	(6.95E-03)	(6.72E-03)	(9.69E-03)	
Cr 52	mg/L	(4.00E-03)	(3.22E-03)			<2.17E-03	<8.33E-03	(4.00E-03)	(3.22E-03)	(3.30E-04)	<5.00E-02	(1.80E-04)	<5.00E-02	(3.20E-04)	<5.00E-02	(7.00E-04)	(1.47E-03)	(1.87E-03)	
Cr 53	mg/L	(1.15E-02)	(9.54E-03)			5.87E-03	3.81E-03	(1.15E-02)	(9.54E-03)	(5.37E-03)	(4.00E-03)	(5.10E-03)	(3.85E-03)	(2.13E-03)	(3.39E-03)	(3.79E-03)	(2.89E-03)	3.07E-03	
Cu	mg/L	(4.18E-01)	(3.82E-01)	(2.29E-01)	(1.90E-01)	(1.88E-01)	(1.59E-01)	(4.18E-01)	(3.82E-01)	(1.81E-01)	(1.89E-01)	(1.46E-01)	(1.34E-01)	(1.26E-01)	(9.51E-02)	(9.95E-02)	(9.02E-02)	(1.42E-02)	(2.86E-02)
Fe	mg/L	(2.37E-01)	(1.85E-01)	(4.79E-02)	(2.13E-02)	(9.68E-03)	(7.14E-03)	(2.37E-01)	(1.85E-01)	(1.67E-01)	(2.30E-01)	(1.28E-01)	(1.07E-01)	(1.30E-01)	(3.52E-01)	(7.76E-02)	(3.50E-02)	9.24E-03	
K	mg/L	(1.05E+00)	(4.49E-01)	(1.19E+00)	(1.14E-01)	(6.72E-01)	(5.25E-01)	(1.05E+00)	(4.49E-01)	<6.25E+00	(6.53E-01)	(1.38E-01)	(2.55E-01)	<6.25E+00	(6.03E-01)	(5.18E-01)	(2.70E-01)	(1.57E-01)	(7.07E-01)
Li	mg/L	<2.50E+01	<2.50E+01	<1.00E+00	<1.00E+00	<2.50E-01	<5.00E-01	<2.50E+01	<2.50E+01	<2.50E+01	<2.50E+01	<2.50E+01	<2.50E+01	<2.50E+01	<2.50E+01	<2.50E+01	<2.50E+01	<5.00E-01	<2.50E+00
Mg	mg/L	1.32E+00	1.32E+00	1.53E+00	1.41E+00	1.39E+00	1.28E+00	1.32E+00	1.32E+00	1.18E+00	1.30E+00	1.07E+00	9.51E-01	5.18E-01	(4.09E-01)	5.55E-01	(2.30E-01)	3.17E-01	6.85E-01
Mn	mg/L	7.88E+01	7.51E+01	7.91E+01	7.87E+01	6.57E+01	6.58E+01	7.88E+01	7.51E+01	7.42E+01	6.86E+01	5.55E+01	4.59E+01	3.00E+01	2.21E+01	1.65E+01	1.27E+01	1.18E+01	1.48E+01
Mo	mg/L	(6.43E-02)	(5.44E-02)	<1.00E-01	<1.00E-01	<5.00E-02	<5.00E-02	(6.43E-02)	(5.44E-02)	(1.79E-02)	<2.50E-01	(2.12E-02)	(3.71E-02)	(2.58E-02)	(1.31E-02)	(3.19E-02)	<2.50E-01	(3.60E-03)	<5.00E-02
Mo 100	mg/L	(1.07E-02)	(1.17E-02)			7.81E-03	6.73E-03	(1.07E-02)	(1.17E-02)	(6.48E-03)	(5.46E-03)	(4.76E-03)	(4.57E-03)	(3.66E-03)	(3.03E-03)	(2.87E-03)	(2.79E-03)	(2.01E-03)	
Mo 95	mg/L	(6.02E-03)	(5.83E-03)			(1.71E-03)	(1.42E-03)	(6.02E-03)	(5.83E-03)	(3.03E-03)	(4.87E-03)	(3.54E-03)	(3.46E-03)	(3.63E-03)	(3.51E-03)	(3.29E-03)	(3.93E-03)	(7.90E-04)	
Mo 98	mg/L	(3.30E-03)	(3.01E-03)			(9.87E-04)	(6.91E-04)	(3.30E-03)	(3.01E-03)	(1.40E-03)	(1.55E-03)	(2.09E-03)	(1.93E-03)	(1.72E-03)	(1.82E-03)	(2.14E-03)	(4.95E-04)		
Na	mg/L	8.46E+01	9.97E+01	1.03E+02	7.93E+01	9.10E+01	6.85E+01	8.46E+01	9.97E+01	1.46E+01	1.88E+01	1.10E+01	1.47E+01	7.44E+00	7.65E+00	7.22E+00	7.13E+00	2.54E+01	3.29E+01
Ni	mg/L	1.56E+00	1.61E+00	9.73E-01	8.70E-01	7.63E-01	7.03E-01	1.56E+00	1.61E+00	1.36E+00	1.64E+00	1.43E+00	1.62E+00	1.77E+00	1.73E+00	1.59E+00	1.42E+00	6.10E-01	6.50E-01
P	mg/L	(1.46E+00)	(1.65E+00)	(2.58E-01)	(1.81E-01)	(2.07E-01)	(1.53E-01)	(1.46E+00)	(1.65E+00)	(1.01E+00)	(1.18E+00)	(1.07E+00)	(9.66E-01)	(8.22E-01)	(8.15E-01)	(8.74E-01)	(8.59E-01)	(1.27E-01)	(6.10E-02)
Pb	mg/L	(1.72E-02)	(2.80E-02)	(5.52E-03)	(7.81E-03)	(4.10E-03)	(2.07E-03)	(1.72E-02)	(2.80E-02)	(4.40E-02)	(2.30E-02)	(1.57E-02)	(8.83E-03)	(1.24E-02)	(7.36E-02)	<5.00E-01	<5.00E-01	<1.25E-01	<1.25E-01
Pb 206	mg/L	7.58E-03	7.08E-03			(1.30E-03)	4.96E-04	7.58E-03	7.08E-03	2.21E-03	2.31E-03	2.18E-03	3.72E-03	3.60E-03	3.17E-03	2.30E-03	2.82E-03	<5.00E-04	
Pb 208	mg/L	8.20E-03	7.81E-03			(1.31E-03)	(5.86E-04)	8.20E-03	7.81E-03	(2.32E-03)	(2.42E-03)	(2.29E-03)	3.93E-03	3.65E-03	3.45E-03	2.88E-03	2.71E-03	(2.00E-06)	
Ru 101	mg/L	(6.48E-03)	(6.93E-03)			5.71E-03	5.13E-03	(6.48E-03)	(6.93E-03)	(3.68E-03)	(3.33E-03)	(3.65E-03)	(2.87E-03)	(2.38E-03)	(2.02E-03)	(1.62E-03)	(1.40E-03)	2.93E-03	
Ru 102	mg/L	(3.62E-03)	(4.22E-03)			2.81E-03	2.19E-03	(3.62E-03)	(4.22E-03)	(2.44E-03)	(1.98E-03)	(2.10E-03)	(1.73E-03)	(1.27E-03)	(1.19E-03)	(1.11E-03)	(1.18E-03)	1.36E-03	
Ru 104	mg/L	(3.36E-03)	(4.49E-03)			3.16E-03	2.41E-03	(3.36E-03)	(4.49E-03)	(2.30E-03)	(2.03E-03)	(2.17E-03)	(2.19E-03)	(1.27E-03)	(1.50E-03)	(1.64E-03)	(1.06E-03)	1.41E-03	
S	mg/L	(4.33E-01)	<5.00E+01	(2.98E-01)	(2.86E-01)	(2.94E-01)	(2.91E-01)	(4.33E-01)	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	(3.68E+00)	<5.00E+01	(1.18E-01)	(5.03E-02)
Se	mg/L	<2.50E+00	<2.50E+00	(5.41E-02)	(3.46E-02)	(3.55E-02)	(4.08E-02)	<2.50E+00	<2.50E+00	<2.50E+00	<2.50E+00	<2.50E+00	<2.50E+00	(3.5E-02)	<2.50E+00	<2.50E+00	<2.50E+00	<2.50E-01	(9.68E-02)
Se 82	mg/L	5.51E-03	<5.00E-01			4.17E-02	<1.60E-04	5.51E-03	<5.00E-01	<5.00E-01	<5.00E-01	9.00E-04	<5.00E-01	(7.28E-03)	<5.00E-01	(1.85E-02)	<5.00E-01	(1.67E-02)	(8.59E-02)
Si	mg/L	(3.05E+00)	(3.13E+00)	(2.21E+00)	(2.06E+00)	2.45E+00	2.33E+00	(3.05E+00)	(3.13E+00)	(2.41E+00)	(2.63E+00)	(2.64E+00)	(2.70E+00)	(1.88E+00)	(1.92E+00)	(1.81E+00)	(1.63E+00)	(1.89E+00)	(1.83E+00)
Sr	mg/L	(3.67E-02)	(3.66E-02)	(3.99E-02)	(4.12E-02)	3.67E-02	3.93E-02	(3.67E-02)	(3.66E-02)	(5.04E-02)	(4.26E-02)	(6.01E-02)	(5.13E-02)	(5.41E-02)	(5.09E-02)	(7.41E-02)	(5.07E-02)	8.51E-02	1.40E-01
Ti	mg/L	(8.81E-03)	<2.50E-01	<5.00E-02	<5.00E-02	(8.34E-04)	(5.51E-04)	(8.81E-03)	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-02	(1.05E-03)
Tl	mg/L							<2.50E+03	<2.50E+03	<2.50E+03	<2.50E+03	<2.50E+03	<2.50E+03	<2.50E+03	<2.50E+03	<2.50E+03	<2.50E+03	#VALUE!	0
V	mg/L	(6.17E-02)	(6.55E-02)	<1.25E-01	<1.25E-01	<6.25E-02	<6.25E-02	(6.17E-02)	(6.55E-02)	(7.84E-02)	(7.64E-02)	(6.46E-02)	(8.34E-02)	(5.45E-02)	(4.51E-02)	(5.80E-02)	(5.47E-02)	<6.25E-02	(4.23E-03)
Zn	mg/L	(5.60E-01)	(6.44E-01)	(1.79E-01)	(1.25E-01)	1.51E-01	1.76E-01	(5.60E-01)	(6.44E-01)	(6.43E-01)	(5.78E-01)	(5.15E-01)	(6.01E-01)	(4.87E-01)	(5.62E-01)	(3.93E-01)	(4.39E-01)	1.73E-01	2.54E-01
Zr	mg/L	(2.62E-02)	(1.23E-02)	(8.91E-04)	(1.12E-03)	(1.47E-03)	<6.25E-02	(2.62E-02)	(1.23E-02)	(7.89E-03)	(1.47E-02)	(7.97E-03)	<2.50E-01	<2.50E-01	(9.80E-03)	(8.56E-03)	(6.57E-03)	<6.25E-02	(2.10E-03)
Anions																			
NO_2^- as $NO2^-$	mg/L	<4.51E-01	<4.51E-01			<3.76E+00	<3.76E+00	<4.51E-01	<4.51E-01	<4.51E-01	<4.51E-01	<4.51E-01	<4.51E-01	<4.51E-01	<4.51E-01	<4.51E-01	<4.51E-01	8.03E-01	1.09E+00
NO_3^- as $NO3^-$	mg/L	<4.33E-01	<4.33E-01			<3.60E+00	<3.60E+00	<4.33E-01	<4.33E-01	<4.33E-01	<4.33E-01	<4.33E-01	<4.33E-01	<4.33E-01	<4.33E-01	<4.33E-01	<4.33E-01	1.09E+00	1.09E+00
CO_3^{2-}	mg/L	1.38E+02	1.90E+02			<4.17E+02	<4.17E+02	1.38E+02	1.90E+02	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01
SO_4^{2-}	mg/L	<4.09E-01	<4.09E-01			<3.41E+00	<3.41E+00	<4.09E-01	<4.09E-01	<4.09E-01	<4.09E-01	<4.09E-01	<4.09E-01	<4.09E-01	<4.09E-01	<4.09E-01	<4.09E-01	<4.09E-01	<4.09E-01
PO_4^{3-} as PO_4^{3-}	mg/L	<5.05E-01	8.09E-01			<4.62E+00	<4.50E+00	<5.05E-01	8.09E-01	<5.05E-01	<5.05E-01	<5.05E-01	<5.05E-01	<5.05E-01	<5.05E-01	<5.05E-01	<5.05E-01	1.58E+00	<5.05E-01
Cl^-	mg/L	6.50E-01	6.56E-01			2.23E+00	5.27E+00	6.50E-01	6.56E-01	3.21E-01	3.17E-01	2.45E-01	2.61E-01	<2.36E-01	2.63E-01	<2.36E-01	<2.36E-01	<2.35E-01	<2.35E-01
F^-	mg/L	2.50E-01	3.06E-01			<9.75E-01	<9.75E-01	2.50E-01	3.06E-01	<1.17E-01	<1.17E-01	<1.17E-01	<1.17E-01	<1.17E-01	<1.17E-01	<1.17E-01	<1.17E-01	<1.17E-01	<1.17E-01
Oxalate	mg/L	2.19E+02	1.92E+02			2.98E+02	2.30E+02	2.19E+02	1.92E+02	1.45E+02	1.25E+02	1.06E+02	9.01E+01	6.07E+01	4.72E+01	3.50E+01	2.86E+01	3.07E+01	3.57E+01
Acetate	mg/L	<1.37E+00	<1.37E+00			<1.14E+01	<1.14E+01	<1.37E+00	<1.37E+00	<1.37E+00	<1.37E+00	<1.37E+00	<1.37E+00	<1.37E+00	<1.37E+00	<1.37E+00	<1.37E+00	2.40E+00	4.64E+00
Formate	mg/L	<4.04E-01	<4.04E-01			<3.37E+00	<3.37E+00	<4.04E-01	<4.04E-01	<4.04E-01	<4.04E-01	<4.04E-01	<4.04E-01	<4.04E-01	<4.04E-01	<4.04E-01	<4.04E-01	8.13E-01	2.04E+00

Table F.3. Molar Concentrations for Water-Leach Tests of Sample 404

Tank C-106 (404) Water Leach Results

Parameter	Units	Single Contact						Periodic Replenishment Tests											
		1 day	1 day (dup)	2 week	2 week (dup)	1 month	1 month (dup)	Stage 1	Stage 1 (dup)	Stage 2	Stage 2 (dup)	Stage 3	Stage 3 (dup)	Stage 4	Stage 4 (dup)	Stage 5	Stage 5 (dup)	Stage 6 A	Stage 6 B
pH	std units																		
EC	mS/cm																		
Alkalinity	mM as CaCO3	3.07	3.66	0.00	0.00	1.27	1.10	3.07	3.66	<2.50	<2.50	<2.50	<2.50	<2.50	<2.50	<2.50	<2.50	0.95	1.13
TIC	mM C/g	1.67	1.15	1.27	1.43	0.76	0.67	1.67	1.15	0.35	0.30	0.59	0.90	1.39	0.33	0.36	0.77		
TOC	mM C/g	5.21	6.10	7.89	6.74	7.21	6.29	5.21	6.10	3.19	3.16	2.22	1.69	0.30	1.20	0.91	0.40		
TC	mM C/g	6.88	7.25	9.16	8.16	7.97	6.96	6.88	7.25	3.54	3.45	2.81	2.58	1.69	1.52	1.27	1.18		
Radionuclides																			
^{90}Sr	mM	2.82E-05	3.09E-05					2.82E-05	3.09E-05	5.39E-05	4.33E-05	7.09E-05	6.18E-05	5.70E-05	6.32E-05	6.53E-05	5.46E-05		
^{99}Tc	mM	1.68E-06	1.48E-06	2.93E-06	2.33E-06	2.64E-06	2.20E-06	1.68E-06	1.48E-06	9.09E-08	2.73E-07	(3.94E-07)	1.82E-07	(9.09E-08)	(3.03E-07)	(3.03E-07)	(3.03E-07)	2.80E-06	3.11E-06
^{238}U	mM	2.27E-04	2.39E-04	1.38E-04	9.97E-05	1.14E-04	8.67E-05	2.27E-04	2.39E-04	4.45E-05	4.28E-05	1.89E-05	1.81E-05	9.20E-06	6.05E-06	3.45E-06	2.72E-06	6.55E-06	5.68E-06
^{129}I	mM	(2.71E-07)	(3.88E-08)	<1.94E-06	(5.43E-08)	(3.88E-08)	(4.03E-07)	(2.71E-07)	(3.88E-08)									(2.33E-07)	
^{239}Pu	mM	(2.34E-06)	(1.74E-06)			(4.36E-07)	(4.88E-07)	(2.34E-06)	(1.74E-06)	(8.79E-07)	(5.44E-07)	(7.32E-07)	(3.97E-07)	(6.90E-07)	(7.95E-07)	(6.49E-07)	(4.39E-07)	(6.69E-07)	(2.93E-07)
^{237}Np	mM	2.46E-05	2.55E-05			2.36E-05	2.07E-05	2.46E-05	2.55E-05	1.01E-05	1.02E-05	6.61E-07	6.43E-07	3.99E-07	3.56E-07	2.61E-07	2.26E-07	2.87E-06	3.12E-06
^{241}Am	mM	2.22E-08	2.71E-08			5.08E-09	(4.06E-09)	2.22E-08	2.71E-08	(1.01E-08)	(8.96E-09)	<2.07E-08	<2.07E-08	<2.07E-08	<2.07E-08	<2.07E-08	<2.07E-08	<2.07E-08	<2.07E-08
Metals																			
Ag 107	mM	5.54E-05	1.07E-04			4.47E-05	2.65E-05	5.54E-05	1.07E-04	(5.51E-06)	(6.64E-06)	(3.36E-06)	(4.39E-06)	(2.80E-06)	(4.11E-06)	(4.02E-06)	(4.39E-06)	5.33E-06	
Ag 109	mM	5.12E-05	9.65E-05			4.05E-05	2.00E-05	5.12E-05	9.65E-05	(5.50E-06)	(4.59E-06)	(4.22E-06)	(4.31E-06)	(2.02E-06)	(3.03E-06)	(3.30E-06)	(2.94E-06)	(4.28E-06)	
Al	mM	2.48E-02	2.86E-02	(8.80E-03)	(6.75E-03)	(3.66E-03)	(2.17E-03)	2.48E-02	2.86E-02	1.89E-02	2.02E-02	(1.73E-02)	1.89E-02	1.91E-02	(1.80E-02)	2.01E-02	1.89E-02	(1.27E-03)	(2.13E-02)
As	mM	(1.03E-03)	(7.94E-04)	(<6.67E-03)	<6.67E-03	<3.34E-03	<3.34E-03	(1.03E-03)	(7.94E-04)	<1.67E-02	(2.01E-03)	<1.67E-02	(4.47E-04)	<1.67E-02	(1.67E-03)	<1.67E-02	(1.67E-02)	(3.44E-05)	(1.99E-04)
As 75	mM	(1.44E-04)	(5.28E-05)			(2.51E-05)	(2.69E-05)	(1.44E-04)	(5.28E-05)	(3.89E-05)	<6.67E-04	(1.29E-05)	(1.03E-05)	(3.93E-05)	(8.60E-05)	<6.67E-04	(3.85E-05)	(1.64E-05)	
B	mM	(3.35E-02)	(2.96E-02)	(1.21E-02)	(1.07E-02)	(6.07E-03)	(4.87E-03)	(3.35E-02)	(2.96E-02)	(2.65E-02)	(1.82E-02)	(2.20E-02)	(2.02E-02)	(1.89E-02)	(1.55E-02)	(1.87E-02)	(1.50E-02)	(4.45E-03)	(9.19E-03)
Ba	mM	2.28E-03	2.16E-03	5.93E-04	4.99E-04	7.90E-04	8.90E-04	2.28E-03	2.16E-03	1.80E-03	2.13E-03	3.06E-03	2.29E-03	2.29E-03	2.27E-03	(1.21E-03)	(1.39E-03)	8.49E-04	1.19E-03
Be	mM	(1.78E-03)	(1.15E-03)	(2.32E-04)	(2.09E-04)	(1.46E-04)	(1.09E-04)	(1.78E-03)	(1.15E-03)	(8.95E-04)	(7.98E-04)	(7.75E-04)	(6.44E-04)	(7.45E-04)	(7.00E-04)	(6.17E-04)	(7.18E-04)	(9.31E-05)	(1.16E-04)
Bi	mM	<5.98E-03	<5.98E-03	(2.32E-03)	<2.39E-03	<1.20E-03	<1.20E-03	<5.98E-03	<5.98E-03	<5.98E-03	<5.98E-03	<5.98E-03	<5.98E-03	<5.98E-03	<5.98E-03	<5.98E-03	<5.98E-03	<1.20E-03	(8.12E-05)
Ca	mM	5.92E-02	8.25E-02	1.24E-02	1.37E-02	1.28E-02	1.37E-02	5.92E-02	8.25E-02	7.37E-02	6.61E-02	7.41E-02	6.87E-02	6.92E-02	7.46E-02	1.78E-01	8.22E-02	4.81E-02	(8.76E-02)
Cd	mM	(1.25E-04)	(8.99E-05)	(1.53E-04)	(1.54E-04)	1.33E-04	1.35E-04	(1.25E-04)	(8.99E-05)	(1.10E-04)	<2.22E-03	(5.51E-05)	<2.22E-03	<2.22E-03	<2.22E-03	<2.22E-03	<2.22E-03	(3.24E-05)	(4.20E-05)
Cd 111	mM	1.71E-04	1.77E-04			1.26E-04	1.33E-04	1.71E-04	1.77E-04	1.77E-04	1.59E-04	1.44E-04	1.27E-04	9.76E-05	(8.14E-05)	(7.06E-05)	(5.21E-05)	3.97E-05	
Cd 114	mM	1.56E-04	1.58E-04			1.23E-04	1.27E-04	1.56E-04	1.58E-04	1.57E-04	1.45E-04	1.33E-04	1.15E-04	(8.65E-05)	(6.90E-05)	(5.24E-05)	(4.71E-05)	3.46E-05	
Co	mM	<4.24E-03	<4.24E-03	(2.98E-04)	2.61E-04	(1.53E-04)	(1.39E-04)	<4.24E-03	<4.24E-03	<4.24E-03	<4.24E-03	<4.24E-03	<4.24E-03	<4.24E-03	<4.24E-03	<4.24E-03	<4.24E-03	(1.98E-05)	<2.12E-04
Cr	mM	(4.51E-04)	(1.56E-04)	(4.47E-04)	(4.00E-04)	3.76E-04	3.56E-04	(4.51E-04)	(1.56E-04)	<6.01E-03	<6.01E-03	(1.74E-04)	<6.01E-03	<6.01E-03	<6.01E-03	<6.01E-03	(1.34E-04)	(1.29E-04)	(1.86E-04)
Cr 52	mM	(7.68E-05)	(6.19E-05)			<4.17E-05	<1.60E-04	(7.68E-05)	(6.19E-05)	(6.35E-06)	<9.62E-04	(3.46E-06)	<9.62E-04	(6.15E-06)	<9.62E-04	(1.35E-05)	(2.83E-05)	(3.60E-05)	
Cr 53	mM	(2.16E-04)	(1.80E-04)			1.11E-04	7.20E-05	(2.16E-04)	(1.80E-04)	(1.01E-04)	(7.55E-05)	(9.62E-05)	(7.26E-05)	(4.02E-05)	(6.40E-05)	(7.15E-05)	(6.55E-05)	5.79E-05	
Cu	mM	(6.57E-03)	(6.01E-03)	(3.60E-03)	(2.99E-03)	(2.96E-03)	(2.50E-03)	(6.57E-03)	(6.01E-03)	(2.85E-03)	(2.98E-03)	(2.30E-03)	(2.12E-03)	(1.98E-03)	(1.50E-03)	(1.57E-03)	(1.42E-03)	(2.23E-04)	(4.50E-04)
Fe	mM	(4.24E-03)	(3.31E-03)	(8.57E-04)	(3.81E-04)	(1.73E-04)	(1.28E-04)	(4.24E-03)	(3.31E-03)	(3.00E-03)	(4.12E-03)	(2.29E-03)	(1.92E-03)	(2.34E-03)	(6.31E-03)	(1.39E-03)	(6.27E-03)	1.65E-04	
K	mM	(2.69E-02)	(1.15E-02)	(3.04E-02)	(2.91E-03)	(1.72E-02)	(1.34E-02)	(2.69E-02)	(1.15E-02)	<1.60E-01	(1.67E-02)	(3.53E-03)	(6.51E-03)	<1.60E-01	(1.54E-02)	(1.33E-02)	(6.92E-03)	(4.00E-03)	(1.81E-02)
Li	mM	<3.60E+00	<3.60E+00	(1.44E-01)	<1.44E-01	<7.20E-02	<7.20E-02	<3.60E+00	<3.60E+00	<3.60E+00	<3.60E+00	<3.60E+00	<3.60E+00	<3.60E+00	<3.60E+00	<3.60E+00	<3.60E+00	<7.20E-02	<3.60E-01
Mg	mM	5.42E-02	5.44E-02	6.28E-02	5.80E-02	<7.20E-02	<7.20E-02	5.42E-02	5.44E-02	4.87E-02	5.35E-02	4.41E-02	3.91E-02	2.13E-02	(1.68E-02)	2.28E-02	(9.47E-03)	1.30E-02	2.82E-02
Mn	mM	1.43E+00	1.37E+00	1.44E+00	1.43E+00	1.20E+00	1.20E+00	1.43E+00	1.37E+00	1.35E+00	1.25E+00	1.01E+00	8.35E-01	5.46E-01	4.02E-01	3.01E-01	2.31E-01	2.15E-01	2.69E-01
Mo	mM	(6.71E-04)	(5.67E-04)	<1.04E-03	<1.04E-03	(5.21E-04)	<5.21E-04	(6.71E-04)	(5.67E-04)	(1.87E-04)	<2.61E-03	(2.21E-04)	(3.86E-04)	(2.69E-04)	(1.37E-04)	(3.33E-04)	<2.61E-03	(3.75E-05)	<5.21E-04
Mo 100	mM	(1.07E-04)	(1.17E-04)			7.81E-05	6.73E-05	(1.07E-04)	(1.17E-04)	(6.48E-05)	(5.46E-05)	(4.76E-05)	(4.57E-05)	(3.66E-05)	(3.03E-05)	(2.87E-05)	(2.79E-05)	(2.01E-05)	
Mo 95	mM	(6.34E-05)	(6.14E-05)			(1.80E-05)	(1.49E-05)	(6.34E-05)	(6.14E-05)	(3.19E-05)	(5.13E-05)	(3.73E-05)	(3.64E-05)	(3.82E-05)	(3.69E-05)	(3.46E-05)	(4.14E-05)	(8.32E-06)	
Mo 98	mM	(3.37E-05)	(3.07E-05)			(1.01E-05)	7.05E-06	(3.37E-05)	(3.07E-05)	(1.43E-05)	(1.58E-05)	(2.13E-05)	(1.97E-05)	(1.89E-05)	(1.76E-05)	(1.71E-05)	(2.18E-05)	(5.05E-06)	
Na	mM	3.68E+00	4.34E+00	4.49E+00	3.45E+00	3.96E+00	2.98E+00	3.68E+00	4.34E+00	6.35E-01	8.16E-01	4.79E-01	6.39E-01	3.24E-01	3.33E-01	3.14E-01	3.10E-01	1.11E+00	1.43E+00
Ni	mM	2.66E-02	2.75E-02	1.66E-02	1.48E-02	1.30E-02	1.20E-02	2.66E-02	2.75E-02	2.31E-02	2.80E-02	2.44E-02	2.77E-02	3.02E-02	2.95E-02	2.70E-02	2.42E-02	1.04E-02	1.11E-02
P	mM	(4.73E-02)	(5.33E-02)	(8.33E-03)	(5.83E-03)	(6.69E-03)	(4.93E-03)	(4.73E-02)	(5.33E-02)	(3.27E-02)	(3.83E-02)	(3.46E-02)	(3.12E-02)	(2.65E-02)	(2.63E-02)	(2.82E-02)	(2.77E-02)	(4.10E-03)	(1.97E-03)
Pb	mM	(8.30E-05)	(1.35E-04)	(2.66E-05)	(3.77E-05)	(1.98E-05)	(9.97E-06)	(8.30E-05)	(1.35E-04)	(2.12E-04)	(1.11E-04)	(7.60E-05)	(4.26E-05)	(6.01E-05)	(3.55E-04)	<2.41E-03	<2.41E-03	<6.03E-04	<6.03E-04
Pb 206	mM	3.68E-05	3.44E-05			(6.31E-06)	2.41E-06	3.68E-05	3.44E-05	1.07E-05	1.12E-05	1.06E-05	1.81E-05	1.75E-05	1.54E-05	1.12E-05	1.37E-05	<2.43E-06	
Pb 208	mM	3.94E-05	3.75E-05			(6.27E-06)	(2.82E-06)	3.94E-05	3.75E-05	(1.12E-05)	(1.16E-05)	(1.10E-05)	1.89E-05	1.75E-05	1.66E-05	1.38E-05	1.30E-05	(9.62E-09)	
Ru 101	mM	(6.42E-05)	(6.86E-05)			5.66E-05	5.08E-05	(6.42E-05)	(6.86E-05)	(3.64E-05)	(3.30E-05)	(3.61E-05)	(2.27E-05)	(2.36E-05)	(2.00E-05)	(1.60E-05)	(1.39E-05)	2.90E-05	
Ru 102	mM	(3.55E-05)	(4.14E-05)			2.76E-05	2.15E-05	(3.55E-05)	(4.14E-05)	(2.39E-05)	(1.94E-05)	(2.06E-05)	(1.70E-05)	(1.25E-05)	(1.17E-05)	(1.09E-05)	(1.16E-05)	1.33E-05	
Ru 104	mM	(3.23E-05)	(4.32E-05)			3.03E-05	2.31E-05	(3.23E-05)	(4.32E-05)	(2.21E-05)	(1.95E-05)	(2.09E-05)	(2.11E-05)	(1.44E-05)	(1.22E-05)	(1.02E-05)	1.36E-05		
S	mM	(1.35E-02)	<1.56E+00	(9.29E-03)	(8.93E-03)	(9.18E-03)	(9.08E-03)	(1.35E-02)	<1.56E+00	<1.56E+00	<1.56E+00	<1.56E+00	<1.56E+00	<1.56E+00	<1.56E+00	(1.15E-01)	<1.56E+00	(3.67E-03)	(1.57E-03)
Se	mM	<3.17E-02	<3.17E-02	(6.85E-04)	(4.38E-04)	(4.50E-04)	(5.16E-04)	<3.17E-02	<3.17E-02	<3.17E-02	<3.17E-02	<3.17E-02	<3.17E-02	(4.5E-04)	<3.17E-02	<3.17E-02	<3.17E-02	<3.17E-02	(1.23E-03)
Se 82	mM	(6.72E-05)	<6.10E-03			(5.08E-04)	<1.95E-06	(6.72E-05)	<6.10E-03	<6.10E-03	<6.10E-03	(1.10E-05)	<6.10E-03	(8.88E-05)	(2.26E-04)	<6.10E-03	(2.04E-04)	(1.05E-05)	
Si	mM	(1.09E-01)	(1.11E-01)	(7.88E-02)	(7.35E-02)	8.71E-02	8.30E-02	(1.09E-01)	(1.11E-01)	(8.58E-02)	(9.38E-02)	(9.41E-02)	(9.63E-02)	(6.71E-02)	(8.43E-02)	(6.73E-02)	(5.80E-02)	(6.73E-02)	(6.50E-02)
Sr	mM	(4.19E-04)	(4.18E-04)	(4.56E-04)	(4.70E-04)	4.19E-04	4.49E-04	(4.19E-04)	(4.18E-04)	(5.75E-04)	(4.86E-04)	(6.86E-04)	(5.86E-04)	(6.17E-04)	(5.81E-04)	(8.45E-04)	(5.79E-04)	9.71E-04	1.59E-03
Ti	mM	(1.84E-04)	<5.22E-03	<1.04E-03	<1.04E-03	(1.74E-05)	(1.15E-05)	(1.84E-04)	<5.22E-03	<5.22E-03	<5.22E-03	<5.22E-03	<5.22E-03	<5.22E-03	<5.22E-03	<5.22E-03	<5.22E-03	<5.22E-04	(2.20E-05)
Tl	mM	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	<1.22E+01	<1.22E+01	<1.22E+01	<1.22E+01	<1.22E+01	<1.22E+01	<1.22E+01	<1.22E+01	<1.22E+01	<1.22E+01	<1.22E+01	
V	mM	(1.21E-03)	(1.29E-03)	<2.45E-03	<2.45E-03	<1.23E-03	<1.23E-03	(1.21E-03)	(1.29E-03)	(1.54E-03)	(1.50E-03)	(1.27E-03)	(1.64E-03)	(1.07E-03)	(8.85E-04)	(1.14E-03)	(1.07E-03)	<1.23E-03	(8.31E-05)
Zn	mM	(8.56E-03)	(9.85E-03)	(2.74E-03)	(1.92E-03)	2.31E-03	2.69E-03	(8.56E-03)	(9.85E-03)	(9.83E-03)	(8.84E-03)	(7.88E-03)	(9.19E-03)	(7.45E-03)	(8.60E-03)	(6.72E-03)		2.64E-03	3.88E-03
Zr	mM	(2.88E-04)	(1.35E-04)	(9.77E-06)	(1.23E-05)	(1.61E-05)	<6.85E-04	(2.88E-04)	(1.35E-04)	(8.65E-05)	(1.62E-04)	(8.74E-05)	<2.74E-03	<2.74E-03	(1.07E-04)	(9.38E-05)	(7.20E-05)	<6.85E-04	(2.30E-05)
Anions																			
NO_2^- as NO2-	mM	<9.80E-03	<9.80E-03			<8.17E-02	<8.17E-02	<9.80E-03	<9.80E-03	<9.80E-03	<9.80E-03	<9.80E-03	<9.80E-03	<9.80E-03	<9.80E-03	<9.80E-03	<9.80E-03	1.74E-02	2.36E-02
NO_3^- as NO3-	mM	<6.98E-03	<6.98E-03			<5.81E-02	<5.81E-02	<6.98E-03	<6.98E-03	<6.98E-03	<6.98E-03	<6.98E-03	<6.98E-03	<6.98E-03	<6.98E-03	<6.98E-03	<6.98E-03	1.76E-02	1.76E-02
CO_3^{2-}	mM	2.29E+00	3.17E+00			<6.94E+00	<6.94E+00	2.29E+00	3.17E+00	<8.33E-01	<8.33E-01	<8.33E-01	<8.33E-01	<8.33E-01	<8.33E-01	<8.33E-01	<8.33E-01	<8.33E-01	<8.33E-01
SO_4^{2-}	mM	<4.26E-03	<4.26E-03			<3.55E-02	<3.55E-02	<4.26E-03	<4.26E-03	<4.26E-03	<4.26E-03	<4.26E-03	<4.26E-03	<4.26E-03	<4.26E-03	<4.26E-03	<4.26E-03	<4.26E-03	<4.26E-03
PO_4^{3-} as PO4^3-	mM	<5.32E-03	8.52E-03			<4.87E-02	<4.73E-02	<5.32E-03	8.52E-03	<5.32E-03	<5.32E-03	<5.32E-03	<5.32E-03	<5.32E-03	<5.32E-03	<5.32E-03	<5.32E-03	1.66E-02	<5.32E-03
Cl^-	mM	1.83E-02	1.85E-02			6.29E-02	1.49E-01	1.83E-02	1.85E-02	9.07E-03	8.94E-03	6.91E-03	7.36E-03	<6.66E-03	7.41E-03	<6.66E-03	<6.66E-03	<6.63E-03	<6.63E-03
F^-	mM	1.31E-02	1.61E-02			<5.13E-02	<5.13E-02	1.31E-02	1.61E-02	<6.16E-03	<6.16E-03	<6.16E-03	<6.16E-03	<6.16E-03	<6.16E-03	<6.16E-03	<6.16E-03	<6.16E-03	<6.16E-03
Oxalate	mM	2.49E+00	2.18E+00			3.39E+00	2.61E+00	2.49E+00	2.18E+00	1.65E+00	1.43E+00	1.20E+00	1.02E+00	6.90E-01	5.36E-01	3.97E-01	3.25E-01	3.49E-01	4.06E-01
Acetate	mM	<2.32E-02	<2.32E-02			<1.93E-01	<1.93E-01	<2.32E-02	<2.32E-02	<2.32E-02	<2.32E-02	<2.32E-02	<2.32E-02	<2.32E-02	<2.32E-02	<2.32E-02	<2.32E-02	4.07E-02	7.86E-02
Formate	mM	<8.98E-03	<8.98E-03			<7.48E-02	<7.48E-02	<8.98E-03	<8.98E-03	<8.98E-03	<8.98E-03	<8.98E-03	<8.98E-03	<8.98E-03	<8.98E-03	<8.98E-03	<8.98E-03	1.81E-02	4.53E-02

Table F.4. Solution Concentrations for Water-Leach Tests of Sample 405

Tank C-106 (405) Water Leach Results

Parameter	Units	Single Contact						Periodic Replenishment Tests											
		1 day	1 day (dup)	2 week	2 week (dup)	1 month	1 month (dup)	Stage 1	Stage 1 (dup)	Stage 2	Stage 2 (dup)	Stage 3	Stage 3 (dup)	Stage 4	Stage 4 (dup)	Stage 5	Stage 5 (dup)	Stage 6 A	Stage 6 B
pH	std units	7.06	7.00			7.00	7.34	6.70	6.72	6.70	6.69	6.77	6.81	6.68	6.67	6.72	6.68	7.50	7.70
EC	mS/cm	0.464	0.517			0.819	0.455	0.269	0.300	0.115	0.118	0.095	0.096	0.065	0.059	0.055	0.055	0.147	0.153
Alkalinity	mg/L as CaCO3	307.44	366.00			127.37	109.80	292.80	278.16	263.52	<250.00	<250.00	<250.00	<250.00	<250.00	<250.00	<250.00	76.86	76.86
TIC	mg C/L	8.93	9.17	15.03	16.89	7.08	5.88	8.93	9.17	4.36	9.11	19.06	14.15	8.08	8.35	7.16	7.09		
TOC	mg C/L	50.77	53.35	69.30	70.32	63.42	67.50	50.77	53.35	27.82	25.19	4.39	10.14	5.14	5.84	3.91	4.00		
TC	mg C/L	59.70	62.52	84.33	87.21	70.50	73.38	59.70	62.52	32.18	34.31	23.45	24.29	13.22	14.18	11.07	11.09		
Radionuclides																			
Sr^{90}	µCi/L	4.42E+02	3.73E+02					4.42E+02	3.73E+02	6.72E+02	6.80E+02	6.98E+02	7.99E+02	7.90E+02	6.54E+02	8.91E+02	8.34E+02	NA	NA
^{99}Tc	mg/L	8.70E-05	6.60E-05	1.73E-04	1.95E-04	1.63E-04	1.80E-04	8.70E-05	6.60E-05	6.00E-06	3.00E-05	(2.40E-05)	2.70E-05	(3.00E-05)	9.00E-06	(3.00E-05)	3.00E-05	1.85E-04	2.10E-04
^{238}U	mg/L	3.34E-02	4.06E-02	1.87E-02	2.38E-02	1.77E-02	2.15E-02	3.34E-02	4.06E-02	7.54E-03	8.66E-03	3.10E-03	3.29E-03	8.08E-04	1.02E-03	3.55E-04	4.45E-04	1.25E-03	1.24E-03
		(1.20E-05)	(2.50E-04)	(2.50E-04)	(5.80E-05)	<2.50E-04	(4.20E-05)	(1.20E-05)	<2.50E-04									(6.50E-05)	
^{239}Pu	µCi/L	(3.60E-03)	(3.41E-03)			(8.53E-04)	(6.46E-04)	(3.60E-03)	(3.41E-03)	(2.26E-03)	(2.57E-03)	(2.54E-03)	(1.86E-03)	(1.15E-03)	(1.33E-03)	(8.06E-04)	(7.75E-04)	(1.18E-03)	(6.51E-04)
^{237}Np	µCi/L	3.19E-03	3.29E-03			3.34E-03	3.77E-03	3.19E-03	3.29E-03	1.23E-03	1.32E-03	8.39E-05	8.13E-05	3.51E-05	3.91E-05	2.33E-05	2.45E-05	3.56E-04	3.55E-04
^{241}Am	µCi/L	(2.34E-03)	<1.70E-02			4.12E-03	(3.58E-03)	(2.34E-03)	<1.70E-02	(1.04E-02)	<1.70E-02	<1.70E-02	<1.70E-02	<1.70E-02	<1.70E-02	<1.70E-02	<1.70E-02	<1.70E-02	<1.70E-02
Metals																			
Ag 107	mg/L	(1.08E-03)	(8.30E-04)			1.30E-03	2.07E-03	(1.08E-03)	(8.30E-04)	(4.10E-04)	(4.60E-04)	(3.60E-04)	(5.00E-04)	(2.90E-04)	(3.60E-04)	(5.60E-04)	(4.00E-04)	(1.90E-03)	
Ag 109	mg/L	(8.40E-04)	(8.40E-04)			9.83E-04	1.76E-03	(8.40E-04)	(8.40E-04)	(5.00E-04)	(3.20E-04)	(3.80E-04)	(4.20E-04)	(3.80E-04)	(3.40E-04)	(4.90E-04)	(4.10E-04)	3.94E-03	
Al	mg/L	6.23E-01	5.88E-01	(1.71E-01)	(1.96E-01)	(4.63E-02)	(8.38E-02)	6.23E-01	5.88E-01	5.51E-01	5.04E-01	5.57E-01	5.09E-01	5.27E-01	5.14E-01	5.87E-01	(4.92E-01)	(6.94E-02)	(5.97E-01)
As	mg/L	(1.82E-01)	<1.25E+00	(1.14E-02)	<5.00E-01	<2.50E-01	<2.50E-01	(1.82E-01)	<1.25E+00	(1.03E-01)	(7.92E-02)	<1.25E+00	<1.25E+00	<1.25E+00	(1.03E-01)	(3.08E-02)	(9.63E-04)	(5.83E-03)	
As 75	mg/L	(1.39E-02)	(7.33E-03)			(2.23E-03)	(2.05E-03)	(1.39E-02)	(7.33E-03)	(6.01E-03)	<5.00E-02	(9.88E-03)	(3.50E-03)	(4.06E-03)	(8.10E-03)	(5.22E-03)	(5.09E-03)	(6.94E-02)	
B	mg/L	(3.27E-01)	(2.37E-01)	(7.18E-02)	(5.91E-02)	(8.08E-02)	(5.03E-02)	(3.27E-01)	(2.37E-01)	(1.37E-01)	(1.49E-01)	(1.53E-01)	(1.35E-01)	(9.84E-02)	(1.11E-01)	(1.13E-01)	(1.00E-01)	(2.87E-02)	(7.07E-02)
Ba	mg/L	3.61E-01	5.23E-01	7.88E-02	1.20E-01	1.26E-01	1.22E-01	3.61E-01	5.23E-01	2.48E-01	2.50E-01	2.84E-01	3.43E-01	3.40E-01	4.89E-01	1.05E+00	2.71E-01	1.02E-01	1.56E-01
Be	mg/L	(2.05E-02)	(1.27E-02)	(2.30E-03)	(1.87E-03)	(2.28E-03)	(1.59E-03)	(2.05E-02)	(1.27E-02)	(9.81E-03)	(8.17E-03)	(7.66E-03)	(6.95E-03)	(7.23E-03)	(6.22E-03)	(6.78E-03)	(6.27E-03)	(1.19E-03)	(2.47E-04)
Bi	mg/L	<1.25E+00	<1.25E+00	<5.00E-01	<5.00E-01	<2.50E-01	<2.50E-01	<1.25E+00	<1.25E+00	<1.25E+00	<1.25E+00	<1.25E+00	<1.25E+00	<1.25E+00	<1.25E+00	<1.25E+00	<1.25E+00	<2.50E-01	(2.83E-02)
Ca	mg/L	4.40E+00	2.58E+00	6.02E-01	6.06E-01	6.04E-01	5.72E-01	4.40E+00	2.58E+00	3.14E+00	3.61E+00	2.80E+00	2.80E+00	3.76E+00	3.23E+00	3.81E+00	3.82E+00	1.49E+00	(3.54E+00)
Cd	mg/L	(1.22E-02)	(1.26E-02)	(1.88E-02)	(1.96E-02)	1.73E-02	1.69E-02	(1.22E-02)	(1.26E-02)	(9.17E-03)	(7.69E-03)	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	(1.90E-03)	(3.09E-03)
Cd 111	mg/L	2.11E-02	2.20E-02			1.62E-02	1.59E-02	2.11E-02	2.20E-02	1.87E-02	2.01E-02	1.39E-02	1.24E-02	(7.21E-03)	(7.01E-03)	(4.98E-03)	(4.46E-03)	(1.17E-03)	
Cd 114	mg/L	2.01E-02	2.00E-02			1.62E-02	1.60E-02	2.01E-02	2.00E-02	1.79E-02	1.88E-02	1.23E-02	1.27E-02	(5.72E-03)	(6.64E-03)	(4.51E-03)	(3.86E-03)	(1.03E-03)	
Co	mg/L	<2.50E-01	<2.50E-01	(1.21E-02)	(1.47E-02)	(9.09E-03)	(9.25E-03)	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	(1.03E-03)	<1.25E-02
Cr	mg/L	(1.91E-02)	<3.13E-01	(2.30E-02)	(2.30E-02)	1.86E-02	2.09E-02	(1.91E-02)	<3.13E-01	<3.13E-01	(7.26E-03)	<3.13E-01	<3.13E-01	<3.13E-01	<3.13E-01	(1.40E-02)	(1.39E-02)	(5.67E-03)	(7.95E-03)
Cr 52	mg/L	(2.83E-03)	(2.56E-03)			<8.33E-03	<8.33E-03	(2.83E-03)	(2.56E-03)	<5.00E-02	<5.00E-02	<5.00E-02	<5.00E-02	(1.82E-03)	(1.35E-03)	(5.20E-03)	<5.00E-02	(3.64E-03)	
Cr 53	mg/L	(1.03E-02)	(1.30E-02)			3.88E-03	4.26E-03	(1.03E-02)	(1.30E-02)	(5.51E-03)	(5.37E-03)	(3.03E-03)	(2.86E-03)	(2.91E-03)	(4.37E-03)	(3.80E-03)	(6.80E-04)	4.41E-03	
Cu	mg/L	(3.34E-01)	(2.23E-01)	(1.60E-01)	(1.74E-01)	(1.37E-01)	(1.50E-01)	(3.34E-01)	(2.23E-01)	(1.57E-01)	(1.47E-01)	(9.18E-02)	(8.93E-02)	(3.56E-02)	(2.98E-02)	(2.32E-02)	(5.47E-02)	(1.12E-02)	(1.01E-02)
Fe	mg/L	(3.72E-01)	(1.79E-01)	(1.99E-02)	(3.11E-02)	(1.12E-02)	(1.15E-02)	(3.72E-01)	(1.79E-01)	(2.97E-01)	(1.67E-01)	(1.15E-01)	(2.03E-01)	(8.85E-02)	(9.28E-02)	(7.24E-02)	(7.57E-02)	(5.03E-03)	(6.80E-03)
K	mg/L	(6.50E-01)	(7.03E-01)	(3.48E-01)	(9.80E-01)	(4.88E-01)	(4.44E-01)	(6.50E-01)	(7.03E-01)	(7.09E-01)	<6.25E+00	(5.16E-01)	(2.45E-01)	<6.25E+00	<6.25E+00	(4.96E-01)	(8.68E-01)	(1.77E-01)	(9.83E-02)
Li	mg/L	<2.50E+01	<2.50E+01	<1.00E+00	<1.00E+00	<5.00E-01	<5.00E-01	<2.50E+01	<2.50E+01	<2.50E+01	<2.50E+01	<2.50E+01	<2.50E+01	<2.50E+01	<2.50E+01	<2.50E+01	<2.50E+01	<5.00E-01	<2.50E+00
Mg	mg/L	1.32E+00	1.03E+00	1.23E+00	1.23E+00	1.11E+00	1.10E+00	1.32E+00	1.03E+00	9.42E-01	9.24E-01	6.84E-01	6.60E-01	(2.43E-01)	(2.52E-01)	(1.54E-01)	(2.33E-01)	1.71E-01	(4.25E-01)
Mn	mg/L	7.42E+01	7.50E+01	7.84E+01	7.91E+01	6.63E+01	6.68E+01	7.42E+01	7.50E+01	5.41E+01	5.81E+01	3.57E+01	3.58E+01	1.24E+01	1.37E+01	6.58E+00	6.73E+00	4.72E+00	6.18E+00
Mo	mg/L	(6.13E-02)	<2.50E-01	<1.00E-01	<1.00E-01	<5.00E-02	<5.00E-02	(6.13E-02)	<2.50E-01	(1.76E-02)	(3.62E-02)	(1.98E-02)	(2.01E-02)	(3.21E-02)	(2.21E-02)	<2.50E-01	(6.51E-02)	<5.00E-02	
Mo 100	mg/L	(7.75E-03)	(8.16E-03)			5.35E-03	5.33E-03	(7.75E-03)	(8.16E-03)	(4.81E-03)	(5.24E-03)	(3.52E-03)	(5.13E-03)	(2.79E-03)	(3.11E-03)	(2.63E-03)	(1.88E-03)	(3.42E-02)	
Mo 95	mg/L	(3.65E-03)	(5.21E-03)			6.80E-03	(7.36E-04)	(3.65E-03)	(5.21E-03)	(3.85E-03)	(4.49E-03)	(3.86E-03)	(3.39E-03)	(3.87E-03)	(4.30E-03)	(3.43E-03)	(3.76E-03)	(2.87E-02)	
Mo 98	mg/L	(2.04E-03)	(1.88E-03)			(3.98E-04)	5.02E-04	(2.04E-03)	(1.88E-03)	(4.10E-03)	(7.78E-03)	(1.57E-03)	(2.07E-03)	<1.53E-03	(1.81E-03)	(1.55E-03)	(1.78E-03)	1.02E-01	
Na	mg/L	4.65E+01	4.85E+01	5.44E+01	6.09E+01	4.77E+01	5.36E+01	4.65E+01	4.85E+01	9.43E+00	1.03E+01	8.98E+00	1.01E+01	6.57E+00	6.27E+00	5.89E+00	6.22E+00	2.45E+01	2.67E+01
Ni	mg/L	1.49E+00	1.36E+00	7.98E-01	8.16E-01	6.34E-01	6.37E-01	1.49E+00	1.36E+00	1.83E+00	1.72E+00	1.72E+00	1.73E+00	1.47E+00	1.62E+00	9.55E-01	1.17E+00	1.98E-01	1.66E-01
P	mg/L	(5.93E-01)	(5.17E-01)	(1.15E-01)	(1.62E-01)	(1.16E-01)	(1.53E-01)	(5.93E-01)	(5.17E-01)	(4.85E-01)	(5.76E-01)	(4.95E-01)	(5.06E-01)	(3.86E-01)	(3.53E-01)	(5.13E-01)	(2.89E-01)	(7.01E-02)	(3.22E-02)
Pb	mg/L	(6.43E-02)	(2.97E-02)	(1.37E-02)	(2.58E-03)	(4.05E-03)	(9.07E-03)	(6.43E-02)	(2.97E-02)	<5.00E+00	(6.13E-02)	(1.10E-02)	(1.92E-02)	(6.32E-03)	(2.71E-02)	(4.46E-02)	(5.63E-02)	<1.25E-01	(5.44E-03)
Pb 206	mg/L	6.20E-03	3.52E-03			(3.61E-04)	6.31E-04	6.20E-03	3.52E-03	3.36E-03	3.20E-03	2.68E-03	8.59E-03	2.61E-03	2.43E-03	2.27E-03	2.68E-03	(5.67E-03)	
Pb 208	mg/L	5.71E-03	3.77E-03			(4.13E-04)	(7.32E-04)	5.71E-03	3.77E-03	3.63E-03	3.29E-03	2.55E-03	8.96E-03	2.65E-03	2.74E-03	2.49E-03	2.76E-03	5.71E-04	
Ru 101	mg/L	(4.05E-03)	(5.61E-03)			3.52E-03	3.86E-03	(4.05E-03)	(5.61E-03)	(2.50E-03)	(2.76E-03)	(2.19E-03)	(2.58E-03)	(1.77E-03)	(1.83E-03)	(1.63E-03)	(1.54E-03)	(1.19E-03)	
Ru 102	mg/L	(2.31E-03)	(2.85E-03)			1.73E-03	1.89E-03	(2.31E-03)	(2.85E-03)	(1.80E-03)	(2.05E-03)	(1.63E-03)	(1.42E-03)	(9.30E-04)	(1.19E-03)	(1.08E-03)	(1.07E-03)	<2.50E-01	
Ru 104	mg/L	(3.06E-03)	(2.77E-03)			1.82E-03	2.03E-03	(3.06E-03)	(2.77E-03)	(1.76E-03)	(2.00E-03)	(1.86E-03)	(1.90E-03)	(1.39E-03)	(9.70E-04)	(1.24E-03)	(1.34E-03)	1.49E+00	
S	mg/L	(1.55E+00)	<5.00E+01	(3.14E-01)	(2.99E-01)	(2.61E-01)	(2.36E-01)	(1.55E+00)	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	(2.14E-01)	(8.58E-02)
Se	mg/L	<2.50E+00	<2.50E+00	(4.96E-02)	(3.07E-02)	(4.47E-02)	(4.84E-02)	<2.50E+00	<2.50E+00	<2.50E+00	(8.31E-02)	<2.50E+00	<2.50E+00	(8.7E-02)	(9.14E-02)	<2.50E+00	(6.9E-02)	(9.67E-03)	(1.14E-01)
Se 82	mg/L	(3.49E-02)	<5.00E-01			4.33E-05	<4.17E-02	(3.49E-02)	<5.00E-01	<5.00E-01	<5.00E-01	(6.83E-03)	(2.37E-02)	(3.58E-03)	<5.00E-01	(9.63E-04)			
Si	mg/L	(2.52E+00)	(2.51E+00)	(1.61E+00)	(2.28E+00)	2.19E+00	2.58E+00	(2.52E+00)	(2.51E+00)	(1.81E+00)	(1.89E+00)	(2.14E+00)	(2.30E+00)	(1.46E+00)	(1.56E+00)	(1.24E+00)	(1.35E+00)	(1.27E+00)	(1.32E+00)
Sr	mg/L	(4.65E-02)	(3.68E-02)	(4.63E-02)	(4.36E-02)	4.36E-02	3.93E-02	(4.65E-02)	(3.68E-02)	(4.65E-02)	(4.53E-02)	(5.35E-02)	(5.30E-02)	(5.98E-02)	(5.46E-02)	(6.93E-02)	(5.47E-02)	1.26E-01	(1.68E-01)
Ti	mg/L	<2.50E-01	<2.50E-01	<5.00E-02	<5.00E-02	(1.94E-03)	(1.24E-03)	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	(5.30E-04)	(7.10E-05)
Tl	mg/L							<2.50E+03	<2.50E+03	<2.50E+03	<2.50E+03	<2.50E+03	<2.50E+03	<2.50E+03	<2.50E+03	<2.50E+03	<2.50E+03		
V	mg/L	(6.26E-02)	(6.72E-02)	<1.25E-01	<1.25E-01	<6.25E-02	<6.25E-02	(6.26E-02)	(6.72E-02)	(6.47E-02)	(5.71E-02)	(5.25E-02)	(6.30E-02)	(5.88E-02)	(8.67E-02)	(6.65E-02)	(7.18E-02)	<6.25E-02	(6.48E-03)
Zn	mg/L	(6.91E-01)	(5.82E-01)	(1.28E-01)	(1.27E-01)	1.65E-01	1.77E-01	(6.91E-01)	(5.82E-01)	(5.75E-01)	(5.48E-01)	(6.05E-01)	(6.92E-01)	(5.85E-01)	(5.80E-01)	(5.66E-01)	(5.24E-01)	1.54E-01	2.08E-01
Zr	mg/L	(2.48E-02)	(1.53E-02)	<1.25E-01	<1.25E-01	(2.10E-03)	(7.25E-04)	(2.48E-02)	(1.53E-02)	(1.09E-02)	(8.94E-03)	(5.09E-02)	(1.97E-02)	<2.50E-01	<2.50E-01	<2.50E-01	<2.50E-01	<6.25E-02	(2.87E-04)
Anions																			
NO_2^- as NO_2^-	mg/L	<4.51E-01	<4.51E-01			<4.51E+00	<4.51E+00	<4.51E-01	<4.51E-01	<4.51E-01	<4.51E-01	<4.51E-01	<4.51E-01	<4.51E-01	<4.51E-01	<4.51E-01	<4.51E-01	7.35E-01	8.08E-01
NO_3^- as NO_3^-	mg/L	<4.33E-01	<4.33E-01			<4.32E+00	<4.32E+00	<4.33E-01	<4.33E-01	<4.33E-01	<4.33E-01	<4.33E-01	<4.33E-01	<4.33E-01	<4.33E-01	<4.33E-01	<4.33E-01	1.15E+00	1.02E+00
CO_3^{2-}	mg/L	5.24E+01	7.00E+01			<5.00E+02	<5.00E+02	5.24E+01	7.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01	<5.00E+01
SO_4^{2-}	mg/L	<4.09E-01	<4.09E-01			<4.09E+00	<4.09E+00	<4.09E-01	<4.09E-01	<4.09E-01	<4.09E-01	<4.09E-01	<4.09E-01	<4.09E-01	<4.09E-01	<4.09E-01	<4.09E-01	5.32E-01	<4.09E-01
PO_4^{3-} as PO_4^{3-}	mg/L	<5.05E-01	<5.05E-01			<5.05E+00	<5.05E+00	<5.05E-01	<5.05E-01	<5.05E-01	<5.05E-01	<5.05E-01	<5.05E-01	<5.05E-01	<5.05E-01	<5.05E-01	<5.05E-01	9.73E-01	<5.05E-01
Cl^-	mg/L	4.05E-01	5.51E-01			2.78E+00	<2.35E+00	4.05E-01	5.51E-01	3.25E-01	2.64E-01	<2.36E-01	2.44E-01	<2.36E-01	<2.36E-01	<2.36E-01	3.28E-01	<2.35E-01	<2.35E-01
F^-	mg/L	1.53E-01	1.79E-01			1.23E+00	<1.17E+00	1.53E-01	1.79E-01	<1.17E-01	<1.17E-01	<1.17E-01	<1.17E-01	<1.17E-01	<1.17E-01	<1.17E-01	<1.17E-01	<1.17E-01	<1.17E-01
Oxalate	mg/L	1.93E+02	1.83E+02			2.39E+02	2.39E+02	1.93E+02	1.83E+02	1.03E+02	1.18E+02	7.26E+01	7.07E+01	2.74E+01	3.15E+01	1.61E+01	8.84E+00	1.84E+01	1.61E+01
Acetate	mg/L	<1.37E+00	<1.37E+00			<1.37E+01	<1.37E+01	<1.37E+00	<1.37E+00	<1.37E+00	<1.37E+00	<1.37E+00	<1.37E+00	<1.37E+00	<1.37E+00	<1.37E+00	<1.37E+00	1.75E+00	3.50E+00
Formate	mg/L	<4.04E-01	<4.04E-01			<4.04E+00	<4.04E+00	<4.04E-01	<4.04E-01	<4.04E-01	<4.04E-01	<4.04E-01	<4.04E-01	<4.04E-01	<4.04E-01	<4.04E-01	<4.04E-01	9.30E-01	1.66E+00

Table F.5. Molar Concentrations for Water-Leach Tests of Sample 405

Tank C-106 (405) Water Leach Results

Parameter	Units	Single Contact						Periodic Replenishment Tests											
		1 day	1 day (dup)	2 week	2 week (dup)	1 month	1 month (dup)	Stage 1	Stage 1 (dup)	Stage 2	Stage 2 (dup)	Stage 3	Stage 3 (dup)	Stage 4	Stage 4 (dup)	Stage 5	Stage 5 (dup)	Stage 6 A	Stage 6 B
pH	std units																		
EC	mS/cm																		
Alkalinity	mM as CaCO3	3.07	3.66			1.27	1.10	2.93	2.78	2.63	<2.50	<2.50	<2.50	<2.50	<2.50	<2.50	<2.50	0.77	0.77
TIC	mM C/g	0.74	0.76	1.25	1.41	0.59	0.49	0.74	0.76	0.36	0.76	1.59	1.18	0.67	0.70	0.60	0.59		
TOC	mM C/g	4.23	4.45	5.78	5.86	5.29	5.63	4.23	4.45	2.32	2.10	0.37	0.85	0.43	0.49	0.33	0.33		
TC	mM C/g	4.98	5.21	7.03	7.27	5.88	6.12	4.98	5.21	2.68	2.86	1.95	2.02	1.10	1.18	0.92	0.92		
Radionuclides																			
^{90}Sr	mM	3.51E-05	2.96E-05					3.51E-05	2.96E-05	5.34E-05	5.39E-05	5.54E-05	6.34E-05	6.27E-05	5.19E-05	7.07E-05	6.62E-05		
^{99}Tc	mM	8.79E-07	6.67E-07	1.75E-06	1.97E-06	1.65E-06	1.82E-06	8.79E-07	6.67E-07	6.06E-08	3.03E-07	(2.42E-07)	2.73E-07	(3.03E-07)	(9.09E-08)	(3.03E-07)	3.03E-07	1.87E-06	2.12E-06
^{238}U	mM	1.40E-04	1.71E-04	7.84E-05	9.98E-05	7.43E-05	9.03E-05	1.40E-04	1.71E-04	3.17E-05	3.64E-05	1.30E-05	1.38E-05	3.39E-06	4.30E-06	1.49E-06	1.87E-06	5.27E-06	5.21E-06
^{241}Am	mM	(2.43E-06)	(2.30E-06)			(5.75E-07)	(4.36E-07)	(2.43E-06)	(2.30E-06)	(1.53E-06)	(1.74E-06)	(1.72E-06)	(1.26E-06)	(7.74E-07)	(9.00E-07)	(5.44E-07)	(5.23E-07)	(7.95E-07)	(4.39E-07)
^{237}Np	mM	1.90E-05	1.95E-05			1.99E-05	2.24E-05	1.90E-05	1.95E-05	7.33E-06	7.82E-06	4.99E-07	4.83E-07	2.09E-07	2.32E-07	1.38E-07	1.46E-07	2.12E-06	2.11E-06
^{239}Pu	mM	(2.86E-09)	<2.07E-08			5.03E-09	(4.37E-09)	(2.86E-09)	<2.07E-08	(1.27E-08)	<2.07E-08	<2.07E-08	<2.07E-08	<2.07E-08	<2.07E-08	<2.07E-08	<2.07E-08	<2.07E-08	<2.07E-08
Metals																			
Ag 107	mM	(1.01E-05)	(7.76E-06)			1.22E-05	1.93E-05	(1.01E-05)	(7.76E-06)	(3.83E-06)	(4.30E-06)	(3.36E-06)	(4.67E-06)	(2.71E-06)	(3.36E-06)	(5.23E-06)	(3.74E-06)	(1.77E-05)	
Ag 109	mM	(7.71E-06)	(7.71E-06)			9.01E-06	1.61E-05	(7.71E-06)	(7.71E-06)	(4.59E-06)	(2.94E-06)	(3.49E-06)	(3.85E-06)	(3.49E-06)	(3.12E-06)	(4.50E-06)	(3.76E-06)	3.62E-05	
Al	mM	2.31E-02	2.18E-02	(6.32E-03)	(7.26E-03)	(1.72E-03)	(3.10E-03)	2.31E-02	2.18E-02	2.04E-02	1.87E-02	2.06E-02	1.89E-02	1.95E-02	1.90E-02	2.17E-02	(1.82E-02)	(2.57E-03)	(2.21E-02)
As	mM	(2.43E-03)	<1.67E-02	(1.52E-04)	<6.67E-03	<3.34E-03	<3.34E-03	(2.43E-03)	<1.67E-02	(1.38E-03)	(1.06E-03)	<1.67E-02	<1.67E-02	<1.67E-02	<1.67E-02	(1.38E-03)	(8.12E-04)	(1.29E-05)	(7.78E-05)
As 75	mM	(1.85E-04)	(9.77E-05)			(2.97E-05)	(2.73E-05)	(1.85E-04)	(9.77E-05)	(8.01E-05)	<6.67E-04	(1.32E-04)	(4.67E-05)	(5.41E-05)	(6.96E-05)	(9.25E-04)	(0.00E+00)		
B	mM	(3.03E-02)	(2.19E-02)	(6.64E-03)	(5.47E-03)	(7.48E-03)	(4.66E-03)	(3.03E-02)	(2.19E-02)	(1.26E-02)	(1.38E-02)	(1.42E-02)	(1.25E-02)	(9.10E-03)	(1.02E-02)	(1.04E-02)	(9.26E-03)	(2.66E-03)	(6.54E-03)
Ba	mM	2.63E-03	3.81E-03	5.74E-04	8.77E-04	9.20E-04	8.88E-04	2.63E-03	3.81E-03	1.81E-03	1.82E-03	2.07E-03	2.50E-03	2.48E-03	3.56E-03	7.63E-03	1.98E-03	7.42E-04	1.14E-03
Be	mM	(2.27E-03)	(1.41E-03)	(2.55E-04)	(2.07E-04)	(2.53E-04)	(1.76E-04)	(2.27E-03)	(1.41E-03)	(1.09E-03)	(9.07E-04)	(8.50E-04)	(7.71E-04)	(8.03E-04)	(6.90E-04)	(7.52E-04)	(6.95E-04)	(1.32E-04)	(2.74E-05)
Bi	mM	<5.98E-03	<5.98E-03	<2.39E-03	<2.39E-03	<1.20E-03	<1.20E-03	<5.98E-03	<5.98E-03	<5.98E-03	<5.98E-03	<5.98E-03	<5.98E-03	<5.98E-03	<5.98E-03	<5.98E-03	<5.98E-03	<1.20E-03	(1.35E-04)
Ca	mM	1.10E-01	6.44E-02	1.50E-02	1.51E-02	1.51E-02	1.43E-02	1.10E-01	6.44E-02	7.82E-02	9.00E-02	6.98E-02	7.00E-02	9.38E-02	8.07E-02	9.49E-02	9.52E-02	3.72E-02	(8.84E-02)
Cd	mM	(1.09E-04)	(1.12E-04)	(1.68E-04)	(1.74E-04)	1.54E-04	1.51E-04	(1.09E-04)	(1.12E-04)	(8.16E-05)	(6.84E-05)	<2.22E-03	<2.22E-03	<2.22E-03	<2.22E-03	<2.22E-03	<2.22E-03	(1.69E-05)	(2.75E-05)
Cd 111	mM	1.90E-04	1.98E-04			1.46E-04	1.43E-04	1.90E-04	1.98E-04	1.69E-04	1.81E-04	1.25E-04	1.12E-04	(6.50E-05)	(6.32E-05)	(4.49E-05)	(4.02E-05)	(1.05E-05)	
Cd 114	mM	1.76E-04	1.76E-04			1.42E-04	1.40E-04	1.76E-04	1.76E-04	1.57E-04	1.65E-04	1.08E-04	1.11E-04	(5.02E-05)	(5.82E-05)	(3.96E-05)	(3.39E-05)	(9.00E-06)	
Co	mM	<4.24E-03	<4.24E-03	(2.05E-04)	(2.50E-04)	(1.54E-04)	(1.57E-04)	<4.24E-03	<4.24E-03	<4.24E-03	<4.24E-03	<4.24E-03	<4.24E-03	<4.24E-03	<4.24E-03	<4.24E-03	<4.24E-03	(1.74E-05)	<2.12E-04
Cr	mM	(3.68E-04)	<6.01E-03	(4.42E-04)	(4.43E-04)	3.58E-04	4.01E-04	(3.68E-04)	<6.01E-03	(1.40E-04)	<6.01E-03	<6.01E-03	<6.01E-03	<6.01E-03	(2.69E-04)	(2.67E-04)	(1.09E-04)	(1.53E-04)	
Cr 52	mM	(5.44E-05)	(4.92E-05)			<1.60E-04	<1.60E-04	(5.44E-05)	(4.92E-05)	<9.62E-04	<9.62E-04	<9.62E-04	<9.62E-04	(3.50E-05)	(2.60E-05)	(1.00E-04)	<9.62E-04	(7.01E-05)	
Cr 53	mM	(1.94E-04)	(2.46E-04)			7.32E-05	8.05E-05	(1.94E-04)	(2.46E-04)	(1.01E-04)	(5.72E-05)	(5.40E-05)	(5.49E-05)	(8.25E-05)	(7.17E-05)	(1.28E-05)	8.32E-05		
Cu	mM	(5.26E-03)	(3.51E-03)	(2.52E-03)	(2.74E-03)	(2.16E-03)	(2.36E-03)	(5.26E-03)	(3.51E-03)	(2.47E-03)	(2.31E-03)	(1.44E-03)	(1.41E-03)	(5.61E-04)	(4.69E-04)	(3.66E-04)	(8.61E-04)	(1.76E-04)	(1.60E-04)
Fe	mM	(6.66E-03)	(3.20E-03)	(3.55E-04)	(5.56E-04)	(2.00E-04)	(2.07E-04)	(6.66E-03)	(3.20E-03)	(5.32E-03)	(2.99E-03)	(2.05E-03)	(3.64E-03)	(1.59E-03)	(1.30E-03)	(1.36E-03)	(9.00E-05)	(1.22E-04)	
K	mM	(1.66E-02)	(1.80E-02)	(8.89E-03)	(2.51E-02)	(1.25E-02)	(1.14E-02)	(1.66E-02)	(1.80E-02)	(1.81E-02)	<1.60E-01	(1.32E-02)	(6.27E-03)	<1.60E-01	<1.60E-01	(1.27E-02)	(2.22E-02)	(4.53E-03)	(2.51E-03)
Li	mM	<3.60E+00	<3.60E+00	<1.44E-01	<1.44E-01	<7.20E-02	<7.20E-02	<3.60E+00	<3.60E+00	<3.60E+00	<3.60E+00	<3.60E+00	<3.60E+00	<3.60E+00	<3.60E+00	<3.60E+00	<3.60E+00	<7.20E-02	<3.60E-01
Mg	mM	5.41E-02	4.25E-02	5.08E-02	5.04E-02	4.57E-02	4.52E-02	5.41E-02	4.25E-02	3.88E-02	3.80E-02	2.82E-02	2.71E-02	(1.00E-02)	(1.04E-02)	(6.34E-03)	(9.58E-03)	7.04E-03	(1.75E-02)
Mn	mM	1.35E+00	1.36E+00	1.43E+00	1.44E+00	1.21E+00	1.22E+00	1.35E+00	1.36E+00	9.84E-01	1.06E+00	6.50E-01	6.52E-01	2.25E-01	2.49E-01	1.20E-01	1.23E-01	8.59E-02	1.12E-01
Mo	mM	(6.39E-04)	<2.61E-03	<1.04E-03	<1.04E-03	(5.21E-04)	<5.21E-04	(6.39E-04)	<2.61E-03	(3.88E-04)	(1.83E-04)	(3.77E-04)	(1.64E-04)	(2.09E-04)	(3.34E-04)	(2.30E-04)	<2.61E-03	(6.78E-06)	<5.21E-04
Mo 100	mM	(7.75E-05)	(8.16E-05)			5.35E-05	5.33E-05	(7.75E-05)	(8.16E-05)	(4.81E-05)	(5.24E-05)	(3.52E-05)	(5.13E-05)	(2.79E-05)	(3.11E-05)	(2.63E-05)	(1.88E-05)	(3.42E-04)	
Mo 95	mM	(3.84E-05)	(5.48E-05)			7.16E-05	7.75E-06	(3.84E-05)	(5.48E-05)	(4.05E-05)	(4.73E-05)	(4.06E-05)	(3.78E-05)	(4.07E-05)	(4.53E-05)	(3.61E-05)	(3.96E-05)	(3.02E-04)	
Mo 98	mM	(2.08E-05)	(1.92E-05)			(4.06E-06)	5.12E-06	(2.08E-05)	(1.92E-05)	(1.43E-05)	(1.81E-05)	(1.60E-05)	(2.11E-05)	(1.56E-05)	(1.85E-05)	(1.58E-05)	(1.74E-05)	1.04E-03	
Na	mM	2.02E+00	2.11E+00	2.36E+00	2.65E+00	2.07E+00	2.33E+00	2.02E+00	2.11E+00	4.10E-01	4.49E-01	3.91E-01	4.38E-01	2.86E-01	2.73E-01	2.56E-01	2.71E-01	1.07E+00	1.16E+00
Ni	mM	2.54E-02	2.33E-02	1.36E-02	1.39E-02	1.08E-02	1.09E-02	2.54E-02	2.33E-02	3.12E-02	2.93E-02	2.93E-02	2.94E-02	2.51E-02	2.77E-02	1.63E-02	1.99E-02	3.37E-03	2.83E-03
P	mM	(1.92E-02)	(1.67E-02)	(3.72E-03)	(5.22E-03)	(3.76E-03)	(4.94E-03)	(1.92E-02)	(1.67E-02)	(1.57E-02)	(1.86E-02)	(1.60E-02)	(1.63E-02)	(1.25E-02)	(1.14E-02)	(1.66E-02)	(9.34E-03)	(2.26E-03)	(1.04E-03)
Pb	mM	(3.11E-04)	(1.43E-04)	(6.62E-05)	(1.25E-05)	(1.95E-05)	(4.38E-05)	(3.11E-04)	(1.43E-04)	<2.41E-03	(2.96E-04)	(5.32E-05)	(9.25E-05)	(3.05E-05)	(1.31E-04)	(2.15E-04)	(2.72E-04)	<6.03E-04	(2.63E-05)
Pb 206	mM	3.01E-05	1.71E-05			1.75E-06	3.06E-06	3.01E-05	1.71E-05	1.63E-05	1.55E-05	4.17E-05	1.27E-05	1.18E-05	1.10E-05	1.30E-05	(2.75E-05)		
Pb 208	mM	2.75E-05	1.81E-05			(1.99E-06)	(3.52E-06)	2.75E-05	1.81E-05	1.75E-05	1.58E-05	1.23E-05	4.31E-05	1.27E-05	1.32E-05	(1.20E-05)	1.33E-05	2.74E-06	
Ru 101	mM	(4.01E-05)	(5.55E-05)			3.48E-05	3.82E-05	(4.01E-05)	(5.55E-05)	(2.73E-05)	(2.17E-05)	(2.55E-05)	(1.75E-05)	(1.81E-05)	(1.61E-05)	(1.52E-05)	(1.18E-05)		
Ru 102	mM	(2.26E-05)	(2.79E-05)			1.70E-05	1.85E-05	(2.26E-05)	(2.79E-05)	(1.76E-05)	(2.01E-05)	(1.60E-05)	(1.39E-05)	(9.12E-06)	(1.17E-05)	(1.06E-05)	(1.05E-05)	<2.45E-03	
Ru 104	mM	(2.94E-05)	(2.66E-05)			1.75E-05	1.95E-05	(2.94E-05)	(2.66E-05)	(1.69E-05)	(1.92E-05)	(1.79E-05)	(1.83E-05)	(1.34E-05)	(9.33E-06)	(1.29E-05)	1.43E-02		
S	mM	(4.84E-02)	<1.56E+00	(9.79E-03)	(9.33E-03)	(8.14E-03)	(7.36E-03)	(4.84E-02)	<1.56E+00	<1.56E+00	<1.56E+00	<1.56E+00	<1.56E+00	<1.56E+00	<1.56E+00	<1.56E+00	<1.56E+00	(6.68E-03)	(2.67E-03)
Se	mM	<3.17E-02	<3.17E-02	(6.29E-04)	(3.88E-04)	(5.66E-04)	(6.13E-04)	<3.17E-02	<3.17E-02	<3.17E-02	(1.05E-03)	<3.17E-02	<3.17E-02	(1.1E-04)	(1.16E-03)	<3.17E-02	(8.8E-04)	(1.22E-04)	(1.45E-03)
Se 82	mM	(4.26E-04)	<6.10E-03			(5.28E-07)	<5.08E-04	(4.26E-04)	<6.10E-03	<6.10E-03	<6.10E-03	(8.33E-05)	(1.82E-04)	<6.10E-03	(2.89E-04)	(4.37E-05)	<6.10E-03	(1.17E-05)	
Si	mM	(8.97E-02)	(8.93E-02)	(5.72E-02)	(8.12E-02)	7.80E-02	9.20E-02	(8.97E-02)	(8.93E-02)	(6.44E-02)	(6.74E-02)	(7.61E-02)	(8.20E-02)	(5.21E-02)	(5.55E-02)	(4.41E-02)	(4.81E-02)	(4.53E-02)	(4.71E-02)
Sr	mM	(5.31E-04)	(4.20E-04)	(5.29E-04)	(4.97E-04)	4.98E-04	4.49E-04	(5.31E-04)	(4.20E-04)	(5.31E-04)	(5.17E-04)	(6.11E-04)	(6.05E-04)	(6.82E-04)	(6.23E-04)	(7.91E-04)	(6.25E-04)	1.44E-03	(1.91E-03)
Ti	mM	<5.22E-03	<5.22E-03	<1.04E-03	<1.04E-03	(4.05E-05)	(2.59E-05)	<5.22E-03	<5.22E-03	<5.22E-03	<5.22E-03	<5.22E-03	<5.22E-03	<5.22E-03	<5.22E-03	<5.22E-03	<5.22E-03	(1.11E-05)	(1.48E-06)
Tl	mM							0.00E+00	<1.22E+01	<1.22E+01	<1.22E+01	<1.22E+01	<1.22E+01	<1.22E+01	<1.22E+01	<1.22E+01	<1.22E+01	<1.22E+01	
V	mM	(1.23E-03)	(1.32E-03)	<2.45E-03	<2.45E-03	<1.23E-03	<1.23E-03	(1.23E-03)	(1.32E-03)	(1.27E-03)	(1.12E-03)	(1.03E-03)	(1.24E-03)	(1.15E-03)	(1.70E-03)	(1.31E-03)	(1.41E-03)	<1.23E-03	(1.27E-04)
Zn	mM	(1.06E-02)	(8.90E-03)	(1.97E-03)	(1.94E-03)	2.53E-03	2.70E-03	(1.06E-02)	(8.90E-03)	(8.39E-03)	(9.25E-03)	(1.06E-02)	(8.94E-03)	(8.88E-03)	(8.65E-03)	(8.02E-03)	2.35E-03	3.18E-03	
Zr	mM	(2.72E-04)	(1.67E-04)	<1.37E-03	<1.37E-03	(2.30E-05)	(7.95E-06)	(2.72E-04)	(1.67E-04)	(1.20E-04)	(9.80E-05)	(5.58E-04)	(2.15E-04)	<2.74E-03	<2.74E-03	<2.74E-03	<6.85E-04	(3.14E-06)	
Anions																			
NO_2^- as $NO2-$	mM	<9.80E-03	<9.80E-03			<9.80E-02	<9.80E-02	<9.80E-03	<9.80E-03	<9.80E-03	<9.80E-03	<9.80E-03	<9.80E-03	<9.80E-03	<9.80E-03	<9.80E-03	<9.80E-03	1.60E-02	1.76E-02
NO_3^- as $NO3-$	mM	<6.98E-03	<6.98E-03			<6.97E-02	<6.97E-02	<6.98E-03	<6.98E-03	<6.98E-03	<6.98E-03	<6.98E-03	<6.98E-03	<6.98E-03	<6.98E-03	<6.98E-03	<6.98E-03	1.85E-02	1.64E-02
CO_3^{2-}	mM	8.73E-01	1.17E+00			<8.33E+00	<8.33E+00	8.73E-01	1.17E+00	<8.33E-01	<8.33E-01	<8.33E-01	<8.33E-01	<8.33E-01	<8.33E-01	<8.33E-01	<8.33E-01	<8.33E-01	<8.33E-01
SO_4^{2-}	mM	<4.26E-03	<4.26E-03			<4.26E-02	<4.26E-02	<4.26E-03	<4.26E-03	<4.26E-03	<4.26E-03	<4.26E-03	<4.26E-03	<4.26E-03	<4.26E-03	<4.26E-03	<4.26E-03	5.54E-03	<4.26E-03
PO_4^{3-} as PO_4^{3-}	mM	<5.32E-03	<5.32E-03			<5.32E-02	<5.32E-02	<5.32E-03	<5.32E-03	<5.32E-03	<5.32E-03	<5.32E-03	<5.32E-03	<5.32E-03	<5.32E-03	<5.32E-03	<5.32E-03	1.02E-02	<5.32E-03
Cl-	mM	1.14E-02	1.55E-02			7.84E-02	<6.63E-02	1.14E-02	1.55E-02	9.18E-03	7.44E-03	<6.66E-03	6.90E-03	<6.66E-03	<6.66E-03	<6.66E-03	<6.66E-03	9.26E-03	<6.63E-03
F-	mM	8.07E-03	9.40E-03			6.45E-02	<6.16E-02	8.07E-03	9.40E-03	<6.16E-03	<6.16E-03	<6.16E-03	<6.16E-03	<6.16E-03	<6.16E-03	<6.16E-03	<6.16E-03	<6.16E-03	<6.16E-03
Oxalate	mM	2.19E+00	2.08E+00			2.71E+00	2.72E+00	2.19E+00	2.08E+00	1.18E+00	1.34E+00	8.25E-01	8.04E-01	3.11E-01	3.58E-01	1.83E-01	1.00E-01	2.09E-01	1.83E-01
Acetate	mM	<2.32E-02	<2.32E-02			<2.32E-01	<2.32E-01	<2.32E-02	<2.32E-02	<2.32E-02	<2.32E-02	<2.32E-02	<2.32E-02	<2.32E-02	<2.32E-02	<2.32E-02	<2.32E-02	2.97E-02	5.93E-02
Formate	mM	<8.98E-03	<8.98E-03			<8.98E-02	<8.98E-02	<8.98E-03	<8.98E-03	<8.98E-03	<8.98E-03	<8.98E-03	<8.98E-03	<8.98E-03	<8.98E-03	<8.98E-03	<8.98E-03	2.07E-02	3.68E-02

Appendix G

Chemical Equilibrium Modeling Calculations

Appendix G

Chemical Equilibrium Modeling Calculations

Chemical equilibrium calculations were made to estimate the saturation condition of the 1-day water-leach solution (sample 404) with respect to phases expected to be present in the sludge. MINEQL+ Version 4.0 for Windows (Schecher and McAvoy 1998) was used to make these calculations. The thermodynamic database used in MINEQL+ derives much of its data from the U.S. Environmental Protection Agency's MINTEQA2 database, which originally came from the U.S. Geological Survey WATEQ4F program. Additional thermodynamic data for components not included in MINTEQA2 database were taken from the original MINEQL program. Formation constants for manganese oxalate ($MnC_2O_4 \cdot 2H_2O$) and dawsonite are not in the MINEQL+ database and were added manually. The formation constant (the inverse of the solubility constant) for $MnC_2O_4 \cdot 2H_2O$ at 298.2 K (25°C) was calculated from thermodynamic data available in the National Bureau of Standards (NBS) Tables of Chemical Thermodynamic Properties (Wagman et al. 1982). The formation constant used for dawsonite at 298.2 K (25°C) was reported by Ferrante et al. (1976) and accepted in the compilation of thermodynamic values published by Robie and Hemingway (1995). The input concentrations and conditions used in most of the calculations discussed in the text are shown in Table G.1.

Table G.1. Initial Concentrations and Conditions Used for Most Equilibrium Modeling Calculations

Component or Parameter	Concentration (mol/L) or Condition
pH	7.03
pe	6.76
Ionic Strength	0.02 (fixed)
Precipitation	Not allowed
Alkalinity (total CO_2)	6.73×10^{-3} (eq/L) (8.13×10^{-3})
Na^+	4.0×10^{-3}
Oxalate ($C_2O_4^{2-}$)	2.3×10^{-3}
Mn^{2+}	1.4×10^{-3}
Ca^{2+}	7.1×10^{-5}
Mg^{2+}	5.4×10^{-5}
Al^{3+}	2.7×10^{-5}

The calculated saturation index (SI), for relevant minerals are shown in Table G.2. SI = Log (Q/K_{sp}), Q is the ion product for the solid, and K_{sp} is the solubility constant for the mineral.

Table G.2. Calculated Saturation Indices of Relevant Minerals for Water-Leached Sample 404

Mineral	SI
Dawsonite [AlNaCO$_3$(OH)$_2$]	-0.17
Mn oxalate [MnC$_2$O$_4$·2H$_2$O]	0.43
whewellite [CaC$_2$O$_4$·H$_2$O]	0.12
Gibbsite [Al(OH)$_3$]	0.09
Rhodochrosite [MnCO$_3$]	1.41
Calcite [CaCO$_3$]	-2.17
Manganite [MnOOH]	-0.83
Hausmannite [Mn$_3$O$_4$]	-2.07
Pyrolusite [MnO$_2$]	-3.08
SI = standard	

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