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CSER 03-011: Transfer from Tank 241-C-106 to Tank 241-AN-106 Using Oxalic Acid Dissolution

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Abstract: This document provides an evaluation of the criticality safety of oxalic acid dissolution to remove residual waste from tank 241-C-106 and transfer to tank 241-AN-106. Limits and Controls for the activity are provided.

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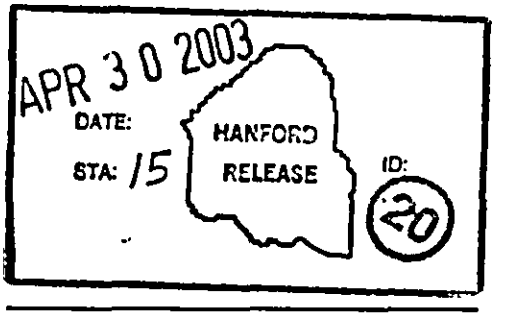

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CSER 03-011: Transfer From Tank 241-C-106 To Tank 241-AN-106 Using Oxalic Acid Dissolution

April 2003

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LIST OF TERMS

BBI	best basis inventory
CFR	code of federal regulations
CSER	criticality safety evaluation report
CWM	conservative waste model
DOE	U.S. Department of Energy
DST	double-shell tank
HLAN	Hanford site local area network
NCS	nuclear criticality safety
SST	single-shell tank
TWINS	Tank Waste Information Network System

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1.0 INTRODUCTION

This Criticality Safety Evaluation Report (CSER) analyzes the nuclear criticality safety (NCS) of transferring high-level waste from Single-Shell Tank (SST) 241-C-106 to Double-Shell Tank (DST) 241-AN-106. For this evaluation these tanks will be referred to as SST C-106 and DST AN-106. These tanks are located in the 200 East Area on the Hanford Site. This analysis provides justification that a nuclear criticality is incredible for the activities described with the controls described.

This CSER evaluates liquid transfers from SST C-106 to DST AN-106 using an oxalic acid dissolution method that includes the following activities:

- Oxalic acid insertion into SST C-106,
- Sludge material redistribution in SST C-106,
- Transfer of oxalic solution from SST C-106 to DST AN-106,
- Discharge of oxalic solution into SST AN-106, including redistribution of waste.

RPP-7475, *Criticality Safety Evaluation of Hanford Tank Farms Facility* (Weiss et al. 2002) covers the criticality safety for activities at Tank Farms. However, the planned activity is substantially different in that acid will be used to dissolve waste solids to permit them to be transferred from one tank to another. This process will violate the Technical Safety Requirement that waste liquids have a pH greater than 8 and introduces chemical processes that have not been previously analyzed. The present evaluation applies only to the transfer of waste from SST C-106 to DST AN-106. This CSER supercedes RPP-7475 (Weiss et al. 2002) only for the specific processes described. If a similar activity is proposed in the future, a new evaluation will be required that evaluates the specific conditions and material contents of the tanks involved.

2.0 DESCRIPTION

Most waste has been removed from SST C-106; however, a mass of hard solid waste remains. Because the remaining waste resists easy removal, an acid dissolution process is considered.

2.1 DESCRIPTION OF ACTIVITIES

Prior to transfer of SST C-106 waste, the entire contents of DST AN-101 will be transferred to DST AN-106. This waste will be caustic supernatant liquid with a pH greater than 10. This transfer will increase the liquid content in DST AN-106, but it will not change the volume of solids. The increase in plutonium inventory due to this transfer is estimated to be only 0.009 kg.

The majority of the contents of SST C-106 were previously removed, leaving three or four piles of hard waste. These piles are up to 4 ft high and are widely spaced. Because of the

difficulty in removing this waste, it is to be dissolved in oxalic acid. The use of acid is a departure from standard activities, which require waste to be caustic.

Between 38 kL (10 kgal) and 132 kL (35 kgal) of oxalic acid at a time will be added in a sluicing process that will break up the hard surface and knock down the piles to facilitate dissolution. The acid depth should not exceed 45.7 cm (18 in.); however, the actual depth is not important to this evaluation. The acid will remain in SST C-106 to allow maximum dissolution before being pumped to DST AN-106. This process will be repeated (up to 10 times) until all solids have been removed or no further significant dissolution is taking place. The total volume of acid will not exceed 795 kL (210 kgal).

A directly connected, dedicated, hose-in-hose assembly will be connected between SST C-106 and DST AN-106 to provide the route for transfer and to contain any leak that might develop. The inner hose with a nominal 5-cm (2-in.) diameter will be inside of a nominal 10-cm (4-in.) diameter hose. The route will pass through the SST C-106 heel pit and then overland to the DST AN-106 heel pit. This direct connection with an overland hose will preclude the possibility of incorrect routing to a different tank. There is an elevation gain of approximately 9.1-m (30-ft) between the SST C-106 heel pit and DST AN-106.

The incoming SST C-106 acid waste will be discharged into the dome space of DST AN-106 where it will fall into the supernatant liquid. Prior to receipt of the SST C-106 waste, the supernate will be charged with sufficient caustic to neutralize the acid stream to a pH greater than 10. The acid waste will have a pH between 1 and 3 as it is received into DST AN-106. Because of the large vessel size and the momentum of the entering waste stream, rapid mixing and neutralization of the acid waste stream is expected. The average pH in DST AN-106 will always be kept above 10.

2.2 SUBCRITICAL LIMIT ABSORBER-TO-PLUTONIUM MASS RATIOS

The ratio of the mass of a neutron absorber to the mass of plutonium, also called the X/Pu mass ratio, plays an important role in ensuring subcriticality. The subcritical limit mass ratio is the smallest absorber-to-plutonium (X/Pu) mass ratio that will ensure subcriticality in an unlimited volume mixed homogeneously with water in any proportion.

It is important that neutron absorbers remain associated with the plutonium. Water-soluble components (e.g., nitrates, nitrites, and sodium compounds) are capable of being separated from the plutonium and removed. Insoluble components provide a much higher assurance of remaining with the plutonium under credible accident conditions and are therefore more important to criticality safety. *High-Level Waste Tank Subcriticality Safety Assessment* (Braun et al. 1994) provides a discussion of the importance of having insoluble components. Nevertheless, the soluble components increase the margin of subcriticality and make a contribution to criticality safety.

Table 2-1 shows subcritical limit absorber-to-plutonium (X/Pu) mass ratios for components of interest reported by *An Analytical Model for Evaluating Subcritical Limits for*

Waste in Hanford Site Storage Tanks, (Rogers 1993). These subcritical limit mass ratios are based on an upper safety limit of k_{eff} of 0.90 and are conservative. Components are listed according to whether they are soluble or insoluble. The subcritical limit mass ratio is given for natural uranium in which the ^{235}U content is 0.72 wt%. This value is used for both natural and depleted uranium. A subcritical limit mass ratio of 770 is conservative when used for depleted uranium.

Table 2-1. Subcritical Limit Mass Ratios for Soluble and Insoluble Components

Insoluble[a] Component	Subcritical Limit Mass Ratio (X/Pu)[b]	Soluble[a] Component	Subcritical Limit Mass Ratio (X/Pu)[b]
Manganese	32	Water	150
Nickel	105	Nitrite (NO_3)	270
Chromium	135	Sodium	360
Iron	160	Calcium	770
Natural uranium	770	Aluminum	910

[a]Classification as soluble or insoluble is based on Braun et al. (1994).

[b]X/Pu mass ratios are taken from Rogers (1993).

Waste is subcritical when the neutron absorption cross-section per atom of plutonium is greater than required for criticality. The contribution of a waste component to subcriticality is proportional to its concentration divided by the plutonium concentration; this is the X/Pu. This contribution can be represented as a fraction by dividing by the subcritical limit mass ratio for that component. The contribution of different absorbers may be added together to obtain a total subcritical mass fraction.

The following rules can be used to evaluate a waste composition.

- Calculate the actual X/Pu for each waste component. Divide each actual mass ratio by its corresponding minimum subcritical mass ratio to obtain a fraction. This fraction will be referred to as the actual-to-minimum subcritical mass fraction.

- When the sum of the actual-to-minimum subcritical mass fractions for all individual components is greater than unity, the waste is subcritical.

Therefore,

$$\sum_{j=1}^{N} \frac{\left(\frac{X_j}{Pu}\right)_{actual}}{\left(\frac{X_j}{Pu}\right)_{subcritical}} \geq 1$$

where:

$\left(\frac{X_j}{Pu}\right)_{actual}$ = Actual absorber mass divided by plutonium mass.

$(\frac{X_j}{Pu})_{subcritical}$ = Minimum subcritical mass ratio.

When the sum of fractions is greater than 1.0, the total neutron absorption is sufficient to ensure subcriticality in an infinite system of homogeneous waste. The greater the sum of the fractions, the greater the margin of safety.

The absorbers of greatest importance to this evaluation are iron and manganese. These components have a tendency to form agglomerates with plutonium and to remain associated with each other (*Tank Farms Nuclear Criticality Review*, Bratzel et al. 1996).

2.3 SST C-106

The Best Basis Inventory (BBI), a database on tank inventories, is maintained on the Tank Waste Information System (TWINS) website (*Tank Waste Information network*, TWINS 2002). The BBI information used in this evaluation was downloaded on April 1, 2003. Table 2-2 shows absorber-to-plutonium subcritical mass fractions for soluble and insoluble components in SST C-106, based upon the BBI (see Appendix B for more details).

Table 2-2. Absorber-to-Plutonium Subcritical Mass Fractions for SST C-106

Component	Subcritical Mass Fractions*		
	Supernate	Sludge	Total
Sum of Soluble Fractions	4007.4	5.7	21.4
Sum of Insoluble Fractions	57.7	36.4	36.5
Sum of Fractions	4065.2	42.1	57.9
Insoluble Absorber-to-Plutonium Subcritical Mass Fractions			
Aluminum	31.4	1.6	1.7
Chromium	4.3	0.3	0.3
Iron	1.4	14.7	14.7
Manganese	1.3	18.3	18.2
Nickel	0.7	0.8	0.8
Silicon	2.3	0.0	0.0
Zirconium	0.0	0.0	0.0
Depleted uranium	14.7	0.04	0.1

*Subcritical mass fractions are derived from BBI. See Table B8 for sludge BBI.

The contents of SST C-106 were listed as 70 kL (19 kgal) of supernatant liquid and 34 kL (9 kgal) of sludge. Following the listing of this inventory data, 68 kL (18 kgal) of supernate was pumped to DST AN-102, leaving behind only 3 kL (0.80 kgal) of supernate. This remaining supernate was diluted with about 4.2 kL (1.1 kgal) of water. Before the oxalic acid is added, the solids will be washed with about 265 kL (70 kgal) of water.

The plutonium-equivalent inventory is 1.92 kg, virtually all of which is in sludge. The supernate contains only 7 g of plutonium. Based upon the BBI, the average plutonium

concentration in the sludge is 0.056 g/L. The sludge contains 60.8 kg of depleted uranium with a 0.64-wt% ^{235}U content, 4514.0 kg of iron, and 1121.7 kg of manganese. There are also other neutron absorbing waste components that increase the margin of subcriticality, but which are not included in this evaluation.

For SST C-106 the 99% upper confidence level on the total alpha is 4.450 μCi/g, on the sludge density is 1.630 g/cm^3, and on the sludge volume is 40.9 kL (10.8 kgal). Based on these upper bound values, the upper bound plutonium inventory is found to be 4.87 kg. This value is 2.5 times larger than the expected value provided by the official inventory.

Herting (2003a, b, c, d, e) performed chemical analyses on samples taken from SST C-106 and DST AY-102 to determine the concentration of various analytes dissolved in oxalic acid. DST AY-102 sludge was considered similar to SST C-106 sludge because much of it was previously transferred from SST C-106. The analyses for dissolved waste was designated as Phase I, and the later analyses to determine the quantity of suspended residue was designated as Phase II. Phase I data are summarized in Table 2-3 (see Appendix B for more details) for plutonium, uranium, iron, and manganese and there mass ratios are calculated, based on the measured concentrations.

There is a wide range of variation in the values in Table 2-3. The average absorber-to-plutonium mass ratios for DST AY-102 samples are more than twice as large as for the SST C-106 samples. Since the SST C-106 samples are taken directly from the waste to be transferred, their average values will be assumed to be the Best Estimate values.

Table 2-3. Analytical Results for Components Dissolved in Oxalic Acid (Phase I)

Sample Ident/Size	Pu (g/L)	U (g/L)	Fe (g/L)	Mn (g/L)	U/Pu	Fe/Pu	Mn/Pu
Dissolved SST C-106 Solids							
A4 (15 mL)	0.00127	<0.010	0.444	0.654	<7.9	350	515
A5 (40 mL)	<0.00121	<0.025	2.730	2.680	21	>2260	>2210
A6 (40 mL)	0.00101	----	----	----	----	----	----
Average*	0.00116	0.018	1.587	1.667	15	1370	1440
Dissolved DST AY-102 Solids							
B4 (15 mL)	0.00212	<0.250	2.970	4.270	<118	1400	2010
C4 (15 mL)	0.00088	0.068	3.150	3.330	77	3580	3780
B5 (40 mL)	0.00091	<0.130	7.240	2.010	<142	7930	2200
B6 (40 mL)	0.00067	0.068	8.040	2.470	101	12000	3690
C5 (40 mL)	0.00088	<0.050	4.460	1.690	<57	5070	1920
C6 (40 mL)	0.00052	0.051	4.550	1.900	98	8750	3650
Average	0.00100	0.103	5.068	2.612	103	5070	2610

Reference: Herting (2003d)

*Average values for SST C-106 are assumed to be Best Estimate for waste transferred to DST AN-106.

The samples were later analyzed in Phase II for their content of residual solids, both suspended and settled solids. Table 2-4 shows the concentrations of plutonium, iron, and manganese in residual solids, based upon data provided in Appendix C (Herting 2003e).

Table 2-4. Analytical Results for Residual Solids (Phase II)

Sample Ident/Size	Pu (g/L)	U (g/L)	Fe (g/L)	Mn (g/L)	U/Pu	Fe/Pu	Mn/Pu
Residual SST C-106 Solids							
A4 (15 mL)	0.00566	----	4.27	2.16	----	754	382
A4 (15 mL)	0.02610	----	12.73	13.80	----	488	529
A5 (40 mL)	0.00443	----	2.77	1.25	----	625	282
Average*	0.01210	----	6.59	5.74	----	545	474
Residual DST AY-102 Solids							
B4 (15 mL)	0.03110	----	26.20	8.60	----	842	276

Reference: Appendix C (Herting 2003e)
*Average values for SST C-106 are assumed to be Best Estimate for waste transferred to DST AN-106.

2.4 DST AN-101

Waste in DST AN-101 is to be transferred to DST AN-106 prior to the transfer of the contents of SST C-106. Table 2-5 shows the sum of the absorber-to-plutonium subcritical mass fractions for soluble and insoluble components.

Table 2-5. Subcritical Mass Fractions for DST AN-101

Component	Supernate Subcritical Mass Fraction*
Sum of Soluble Fractions	119429
Sum of Insoluble Fractions	2115.0
Sum of Component Fractions	121545
Insoluble Subcritical Mass Fractions	
Aluminum	1985.2
Chromium	108.1
Iron	7.0
Manganese	7.0
Nickel	4.3
Silicon	1.9
Zirconium	0.1
Uranium (< 0.72 wt% ^{235}U)	1.7

*Subcritical mass fractions are derived from BBI.

The BBI for April 1, 2003, lists the contents of DST AN-101 as 956 kL (253 kgal) of supernatant liquid with no sludge or saltcake. The plutonium-equivalent inventory is 0.009 kg. The plutonium concentration is less than 10^{-5} g/L. The BBI inventory contains 11.4 kg of uranium with a 0.70 wt% ^{235}U content.

2.5 DST AN-106 PRIOR TO RECEIPT OF DST AN-101 WASTE

The BBI for DST AN-106 for April 1, 2003, showed 526 kL of supernate and 65 kL of saltcake. The plutonium-equivalent inventory was 1.26 kg, with 0.20 kg in supernate and 1.06 kg in saltcake. The average plutonium concentration was found to be 0.016 g/L in the saltcake and 0.0004 g/L in the supernate. The saltcake contains 81.5 kg of natural uranium (0.71 wt% ^{235}U) and the supernatant liquid contains 1883 kg of depleted uranium (0.65 wt% ^{235}U).

Table 2-6 shows the absorber-to-plutonium subcritical mass fractions. This is the composition prior to receipt of waste from DST AN-101 or from SST C-106.

Table 2-6. Absorber-to-Plutonium Subcritical Mass Fractions for DST AN-106

Component	Subcritical Mass Fractions*		
	Supernate	Saltcake	Total
Sum of Soluble Fractions	672.7	106.2	194.6
Sum of Insoluble Fractions	18.1	6.5	8.3
Sum of Component Fractions	690.8	112.7	202.9
Insoluble Absorber-to-Plutonium Subcritical Mass Fraction			
Aluminum	1.7	2.6	2.4
Chromium	2.3	3.2	3.1
Iron	0.1	0.2	0.2
Manganese	0.0	0.2	0.1
Nickel	1.3	0.2	0.4
Silicon	0.1	0.0	0.0
Zirconium	0.3	0.0	0.0
Depleted uranium	12.4	0.1	2.0

*Subcritical mass fractions are derived from BBI. See Table B9 for sludge values.

2.6 DST AN-106 AFTER RECEIPT OF DST AN-101 WASTE

Table 2-7 shows the absorber-to-plutonium subcritical mass fractions for DST AN-106 after the receipt of waste from AN-101. Based on the BBI for April 1, 2003, the waste volume will be 1482 kL of supernate and 65 kL of saltcake. The plutonium-equivalent inventory will be 1.27 kg, with 0.205 kg in the supernate and 1.065 kg in the saltcake. The average plutonium concentration is found to be 0.016 g/L in the saltcake and 0.00001 g/L in the supernate.

2.7 HEEL PIT

The pump will be located in the heel pit (also known as a pump pit) associated with SST C-106. The dimensions of this pit are nominally 1.8 m (6 ft) wide by 2.7 m (9 ft) long by 0.65 m (25.5 in.) deep below the cover block. The cover block is 0.6 m (24 in.) thick. The pit drain is plugged with grout. The pump is inserted through the heel pit riser into the waste at the bottom of SST C-106. The hose will be run overland to DST AN-106.

A leak detector is in place that will detect the presence of liquid flowing back into the pump through the outer hose. Detection of liquid in the outer hose will cause the pump to stop.

Table 2-7. Absorber-to-Plutonium Subcritical Mass Fractions for DST AN-106 After Receipt of DST AN-101 Waste

Component	Subcritical Mass Fractions*		
	Supernate	Saltcake	Total
Sum of Soluble Fractions	5602.1	106.2	993.3
Sum of Insoluble Fractions	105.1	6.5	22.4
Sum of Component Fractions	5707.2	112.7	1015.7
Insoluble Absorber-to-Plutonium Subcritical Mass Fraction			
Aluminum	84.1	2.6	15.7
Chromium	6.7	3.2	3.8
Iron	0.3	0.2	0.2
Manganese	0.3	0.2	0.2
Nickel	1.4	0.2	0.4
Silicon	0.1	0.0	0.0
Zirconium	0.2	0.0	0.0
Depleted uranium	12.0	0.1	2.0

*Subcritical mass fractions are derived from BBI for April 1, 2003.

2.8 CONNECTION TO DST AN-106

Connection to DST AN-106 will be made using the overland hose coming from SST C-106. Special care must be taken to ensure that this hose is properly connected to DST AN-106 and is not connected to any other tank.

The connecting hose will use a hose-in-hose assembly with the inner hose having a nominal 10-cm (2-in.) diameter. The hose is reinforced with steel wire and is rated for a pressure of 400 PSIA. In tests a hose of this construction has withstood a pressure greater than 1,000 PSIA. Because of the steel reinforcement, there is little stretching of the hose. The length of this hose will be about 183 m (600 ft). The total hose volume will be about 1500 L (396 gal) and the central hose will have a volume of about 375 L (99 gal).

3.0 REQUIREMENTS DOCUMENTATION

The presence of fissionable material makes it necessary to exercise criticality safety over the transfer of waste. The goal of the Tank Farm criticality safety program is to assure that all waste remains in a form or distribution that is subcritical under normal and credible abnormal operating conditions. A summary is provided of the requirements of the Criticality Safety Program.

3.1 REGULATORY REQUIREMENTS

DOE Order 420.1A, *Facility Safety* (DOE 2002), establishes the facility safety requirements for the Department of Energy for criticality safety. Section 4.3 mandates that a program be established ensuring that "criticality safety is comprehensively addressed and receives an objective review, with all identifiable risks reduced to acceptably low levels and management authorization of the operations documented."

Code of Federal Regulations (CFR) 10 Part 830, *Nuclear Safety Management* (DOE 2001), "governs the conduct of DOE contractors, DOE personnel, and other persons conducting activities (including providing items and services) that affect, or may affect, the safety of DOE nuclear facilities."

TFC-ENG-CHEM-P-04, *Criticality Safety Evaluations* (CHG 2002), describes the actions to be taken when a new Criticality Safety Evaluation Report (CSER) is needed. The formatting and content of this CSER conforms to requirements provided in DOE-STD-3007-93, *Guidelines for Preparing Criticality Safety Evaluations at Department of Energy Non-Nuclear Facilities* (DOE 1998).

3.2 TECHNICAL SAFETY REQUIREMENTS

HNF-SD-WM-TSR-006, *Tank Farms Technical Safety Requirements* (CHG 2003a) contains Administrative Control (AC) 5.7 for the NCS program. This specifies the criticality safety requirements for waste transfers into tank farms from non-tank farm facilities. Implementation of AC 5.7 is provided by Chapter 5.7 of HNF-IP-1266, *Tank Farms Operation Administrative Controls* (CHG 2003b), Criticality Prevention Specifications (CPS), and RPP-14330, *Tank Farms Criticality Safety Manual* (CHG 2003c). The activities covered by this evaluation will involve the transfer of waste between tanks and compliance to the Technical Safety Requirements need not be demonstrated.

The only material to be introduced into tank farms facilities will be oxalic acid to SST C-106 and caustic to DST AN-106 and SST C-106. Neither of these liquids will contain fissionable material. However, the oxalic acid will violate the requirement that the waste have a pH greater than 8. It will be necessary to obtain a waiver for this requirement. This criticality safety evaluation report provides justification that criticality will remain incredible after the introduction of up to 795 kL (210 kgal) of oxalic acid into SST C-106. Upon transfer to DST AN-106, this acid will be neutralized with caustic, and the final pH will exceed a pH of 8.

3.3 TANK FARMS SPECIFIC CRITICALITY SAFETY CRITERION

The criticality safety criterion for tank waste is that the form or distribution of plutonium must ensure that criticality is not credible. For settled waste solids, a high proportion of neutron absorbers provide an assurance of subcriticality. A low plutonium mass and concentration in relationship to the corresponding critical values provide additional assurance of subcriticality. The low plutonium saturation concentration in caustic liquids and in oxalic acid provides

additional assurance that the plutonium concentration will always be well below the critical concentration.

4.0 METHODOLOGY

Criticality safety for the acid dissolution method of waste transfer is more complicated than criticality safety for a static, caustic waste configuration, as has been previously analyzed in RPP-7475 (Weiss et al. 2002) and predecessor evaluations. In the dissolution and reprecipitation process both the plutonium and the neutron absorber form and distribution will change. This requires that a detailed compilation of the chemical characterization of the waste be estimated for each stage of the transfer process. The methodology used to ensure incredibility is to first describe a bounding envelope of material composition that might arise from chemical transformations and waste combinations in the transfer activities. Then, all of these configurations must be shown to remain subcritical under all credible conditions

This evaluation will show that the plutonium mass required to achieve criticality in any given volume when mixed with the waste will exceed the plutonium mass that might credibly be concentrated into that volume. An important chemical property of plutonium dissolved in oxalic acid is that the saturation concentration is well below the minimum critical concentration. This evaluation will examine initial and final states of the waste using material compositions that are conservative with respect to the actual waste. The low concentration of the plutonium during transfer and the small diameter of the connecting hose preclude criticality outside of the storage tanks.

Basic criticality parameters were obtained from WHC-SD-SQA-CSA-507, *Criticality Parameters for Tank Waste Evaluation* (Rogers et al., 1996). Subcritical limit absorber-to-plutonium mass ratios for iron, manganese, uranium, and other waste components were used to obtain subcritical mass fractions. Subcriticality was demonstrated by showing that the sum of the subcritical mass fractions remains above 1.0 with an adequate safety factor at all times.

Herting (2003a, b, c, d, e) supplied the primary data used in this evaluation. The assumed transfer rates of waste components were based upon this data.

Tank waste characterization data was obtained from the Best Basis Inventory (BBI) on the Tank Waste Information Network System (TWINS) database maintained on the Hanford Site Local Area Network (HLAN). The BBI provides the most current estimate of chemical inventories for Hanford underground waste storage tanks.

The calculations performed for this evaluation involved simple arithmetic operations. The inventory for an isotope in grams was found by dividing the inventory in curies by the specific activity. Simple division was used to obtain mass ratios and mass fractions. An EXCEL spreadsheet calculated mass ratios and mass fractions based on BBI data.

5.0 DISCUSSION OF CONTINGENCIES

The transfer of the contents of DST AN-101 to AN-106 will not create any criticality safety concerns due to the very low plutonium-equivalent inventory of 9 g in DST AN-101. The sum of the absorber-to-plutonium subcritical mass fractions for insoluble components in DST AN-101 is estimated to be more than 2000. There are no solids in the transfer, and the solids volume in DST AN-106 will not change. The transferred waste will be highly caustic, and the final pH for DST AN-106 will remain relatively unchanged.

The solids in SST C-106 will be both dissolved and suspended in oxalic acid and transferred as a mixture to DST AN-106. The following conditions are assumed for this evaluation of the transfer process:

- The total SST C-106 plutonium content may be transferred to DST AN-106.
- The total SST C-106 plutonium content may remain in SST C-106 (this is a very low probability).
- The BBI provides an upper limit on the mass of plutonium, both in a waste layer and in the entire tank. The plutonium-equivalent inventory for SST C-106 is given as less than 1.9 kg, with an upper bound of 4.87 kg at the 99% confidence level.

  The plutonium-equivalent inventory for DST AN-106 is given as 1.3 kg before the transfer begins. The 99% upper bound is about 3.3 kg, assuming the same factor increase as for SST C-106. This plutonium will remain immobile and well subcritical throughout the transfer process.
- Before or during pumping of the acidic liquid to DST AN-106, sufficient caustic will be added to maintain the pH of the supernate above 10 in DST AN-106.

If one of the following criteria is met at every location within the waste, subcriticality will be assured:

- The subcritical mass fraction for insoluble absorbers in a liter of settled solids will exceed 3.0.
- The plutonium concentration is less than 10 g/L, the minimum critical concentration for 2.0 kg of plutonium in water (*Criticality Handbook,* Carter et al. 1969, III.A.6.100-3)).
- The total mass of plutonium in a 17-L sphere is less than 530 g, the minimum critical mass in water (Carter et al. 1969, III.A-2)).
- The plutonium areal density is less than 2.6 kg/m^2 (240 g/ft^2), the minimum critical areal density in a homogenous infinite plane (Carter et al. 1969, III.A.8.100-3).

It is not possible to know whether a criterion is met at every location within the waste. A criterion is assumed to be met for a waste layer when the average value is less than the criterion

and the waste layer is quasi-homogeneous.

5.1 SST C-106 BEFORE ACID ADDITION

Subcriticality in SST C-106 is ensured by the limits that applied when the waste was discharged from the processing facilities. These discharges were covered by criticality safety evaluations that required that the plutonium concentration in settled solids remain less than 1 g/L at all times. The discharged waste was required to be highly caustic to ensure that the concentration of dissolved plutonium was very low. The plutonium coprecipitated with neutron absorbers and the caustic nature of the waste prevented chemical processes capable of changing the form or distribution of waste solids. The mass of insoluble waste components exceeds the minimum required to ensure subcriticality by more than 30 times. In addition the plutonium concentration as determined by analysis of waste samples was found to be much less than the minimum concentration for which criticality is possible. The solids in SST C-106 are well subcritical, and there are no natural processes capable of significantly reducing the margin of subcriticality.

5.2 SST C-106 AFTER ACID ADDITION

Acid dissolution in SST C-106 is a complex chemical process that takes plutonium and other waste components into solution. This process creates criticality safety concerns by changing the relative proportions of plutonium to the various neutron absorbers. Nevertheless, there are several barriers that must be crossed before criticality is possible.

- First, the sum of the neutron absorber-to-plutonium mass ratios must fall below the minimum ratio that prevents criticality over a volume large enough for criticality to occur. The absorber-to-plutonium mass ratio in SST C-106 sludge is estimated to be 36 times greater than the value that prevents criticality in a homogeneous mixture and the initial waste configuration is quasi-homogeneous, based on BBI inventories. It would be extremely unlikely for the absorber-to-plutonium mass ratio in even a small volume of this sludge to approach 1.0.

- Second, the plutonium concentration must exceed the minimum for which criticality is possible and this must occur over a volume large enough for criticality.

  The plutonium saturation concentration in oxalic acid is about 0.06 g Pu/liter at 50° C. This is much less than the minimum concentration for which criticality is possible. In a plutonium-water configuration the plutonium minimum critical concentration is at least 7.2 g/L (Carter et al. 1969, III.A-2). For oxalic acid the plutonium minimum critical concentration is expected to be about the same, but perhaps slightly less, than for water. When the dissolved absorbers are taken into account, the plutonium minimum critical concentration will increase and the value for pure water will be conservative. The plutonium saturation concentration is 100 times less than the minimum concentration that can be made critical.

Based on the Conservative Waste Model (Rogers 1993) the plutonium concentration in the solids must exceed the subcritical limit of 2.6 g/L before criticality is possible. For criticality to occur at this low concentration requires that there be no water or oxalic acid combined with the solids. Otherwise, the minimum critical plutonium concentration will be larger.

At the minimum critical concentration, the plutonium critical mass is infinite. For a plutonium inventory of 2 kg (the mass actually available in DST C-106) criticality cannot occur until the plutonium concentration exceeds 10 g/L.

- Third, the plutonium mass in a compact volume must exceed the minimum for which criticality is possible. In a purely plutonium-water system the plutonium mass would have to exceed 530 g in a sphere in the range of 17 L (with a concentration about 30 g/L). As absorbers are added the minimum critical mass will increase.

- Fourth, the plutonium areal density must exceed 2.6 kg/m^2 (240 g/ft^2) (Carter et al. 1969, III.A.8.100-3). The areal density is defined as the plutonium mass above a unit area of floor. In practical terms, the mass above a 0.30-m (1-ft) square area must exceed 240 g. In addition, a sizeable area is needed for criticality.

5.3 THE ACID SOLUTION DURING TRANSFER

During transfer the waste will be confined to an approximately 5-cm (2-in.) diameter hose. Should a leak develop in the inner hose, the solution would flow into the annulus and drain back to the SST C-106 heel pit. Then it would flow through a drain in the pump plate back into SST C-106. Before criticality would be possible within a fully-reflected, 10-cm (4-in.) diameter hose, the plutonium concentration would have to exceed 4 kg/L (Carter et al. 1969, III.A.4-1) in a pure plutonium-water solution. It is not credible for the outer hose to stretch under pressure due to the strength of the hose, which is rated to 400 PSIA and which has withstood a pressure of 1,000 PSIA in tests. These pressures are much higher than expected during transfer activities. If it were hypothesized that the hose ruptured, the waste would flow onto the ground. Criticality in the transfer hose is not credible.

5.3.1 Heel Pit

The waste is pumped through a 5-cm (2-in.) diameter hose inside of a 10-cm (4-in.) diameter hose. Between the pump in the SST C-106 heel pit and DST AN-106 there is an elevation gain of about 9.1 m (30 ft). A leak in the inner hose would drain into the outer hose and flow back towards the heel pit. When the returning waste reached the pit, a leak detector would detect its presence and shut off the pump. The maximum volume of waste that could flow back into the heel pit would be no greater than the volume of the outer hose, or less than 1500 L (396 gal). The plutonium contained in this volume of waste would be much less than the minimum critical mass of 530 g in a purely water solution. In addition, the mass ratio of neutron absorbing solids to plutonium would be high. Criticality in the heel pit is not credible.

5.3.2 Routing to Incorrect Tank

Another potential accident would be to route the acid solution to the wrong tank. This would require connection of the hose to the inlet pipe to the wrong tank. Administrative controls will be used to ensure that the hose from SST C-106 is connected to the correct inlet pipe. An examination of the connection to DST AN-106 will be made independently by at least two individuals.

5.4 DST AN-106 BEFORE ACID ADDITION

The addition of AN-101 liquid does not lower the margin of subcriticality for DST AN-106. However, assurance must be provided that the routing of waste is correct and only the contents of DST AN-101 are pumped into DST AN-106.

The barriers described for SST C-106 before acid addition apply to DST AN-106 before receipt of waste from SST C-106. The sum of the absorber-to-plutonium subcritical mass fractions for DST AN-106 exceeds that for SST C-106. Therefore, the margin of subcriticality in DST AN-106 prior to receipt of SST C-106 waste is greater than that for SST C-106. There are no natural processes capable of reducing the margin of subcriticality for DST AN-106 prior to the transfer of waste from SST C-106. Criticality is not credible for this waste.

5.5 DST AN-106 AFTER ACID ADDITION

The barriers described for SST C-106 after acid addition apply to the receipt of this waste into DST AN-106. However, the precipitation of solids upon entry into the highly caustic waste in DST AN-106 adds a concentration mechanism and also increases the plutonium inventory. For these reasons, the possibility must be considered that the margin of subcriticality might be reduced. An evaluation of the receipt of waste into DST AN-106 bounds the entire transfer process.

6.0 EVALUATION AND RESULTS

The primary criticality safety concerns arise with the mixing of acidic waste from SST C-106 with liquid waste in DST AN-106. For this reason, the following evaluation is primarily directed to the process of combining the wastes.

6.1 BASE CASE CONDITIONS

Before transfer activities, SST C-106 contains 70 kL (19 kgal) of supernate liquid and 34 kL (9 kgal) of sludge. Between 38 kL (10 kgal) and 132 kL (35 kgal) of oxalic acid will be added at a time and allowed to remain in the tank over a period of time to dissolve as much sludge as possible. Chemical activity during dissolution will release gas and cause fine particulate matter to break away from the hard sludge and to become suspended in the acid. Although no data on particle size has been provided, a large proportion of the material that breaks away from the hard sludge will certainly consist of extremely small particles. These particles will remain suspended for a long period of time and will be carried along by currents within the liquid. Much of this particulate will be pumped out of SST C-106 during each transfer of waste.

After the acidic waste, which contains both dissolved and suspended solids, is transferred to DST AN-106, the process will be repeated. Up to 10 episodes of acid addition may be required to dissolve the majority of the sludge. The total volume of oxalic acid used in the dissolution process will not exceed 795 kL (210 kgal).

Before receipt of the acid waste into DST AN-106, a sufficient volume of caustic will be added to the supernate in DST AN-106 to neutralize the incoming acid. The final pH after neutralization will be no lower than 10. The volume of waste in DST AN-106 will increase by the volume of the transferred SST C-106 waste, the added oxalic acid, and the added caustic.

The expected composition of the waste pumped to DST AN-106 is a homogeneous mixture of 795 kL (210 kgal) of oxalic acid, 34 kL (9 kgal) of sludge, and 70 kL (18 kgal) of supernate. The composition of the sludge and supernate for SST C-106 is as described in Section 2.3 and in Appendix B. The saltcake in DST AN-106 prior to transfer is assumed to be homogeneous and to remain homogeneous throughout the transfer process. The compositions of the supernatant liquid and the saltcake in DST AN-106 are described in Section 2.6 and in Appendix B.

6.2 CONTINGENCY SUMMARY

Table 6-1 provides a summary of contingencies associated with discharging waste from SST C-106 into DST AN-106. Contingencies are discussed in the following sections.

6.3 DISTRIBUITON OF PRECIPITATED WASTE SOLIDS IN DST AN-106

Before the transfer of acid from SST C-106, the supernatant liquid in DST AN-106 will be at least 3.3 m (10.8 ft) deep. Prior to each acid waste addition, sufficient caustic will be added to ensure a pH of 10 upon completion of the transfer. Acid waste will enter DST AN-106 through the dome and fall into the supernatant. The momentum of the acid stream entering the supernatant will cause rapid mixing that will result in a rapid acid/base neutralization reaction. During neutralization the oxalic acid will be converted to sodium oxalate, which then

Table 6-1. Contingency Summary For DST AN-106 After Receipt of SST C-106 Waste

Contingency Description	Affected Parameters	Barriers that Make Contingency Unlikely
Absorber-to-plutonium mass ratio below limit	Absorption	Large proportion of neutron absorbers in relationship to the plutonium. Agglomeration of solids with plutonium during precipitation. Low plutonium concentration.
Exceed minimum critical concentration in solution	Concentration	Low plutonium saturation concentration in solution (about 0.06 g Pu/liter at 50° C (Harmon et al. 1961)). Tendency of waste components to diffuse. Lack of effective concentration mechanism.
Exceed minimum critical concentration in solids	Concentration	Lack of effective concentration mechanisms. High insoluble component-to-plutonium mass ratio. Tendency for solids to agglomerate with plutonium.
Exceed minimum critical mass in 20 L volume	Mass Volume Concentration	Low incoming plutonium concentration. Lack of effective concentration mechanisms. High insoluble component-to-plutonium mass ratio.
Exceed critical areal density over an area greater than 0.30-m (1-ft) on a side	Mass Concentration	Large area of tank bottom. Low incoming plutonium concentration. Tendency for flocculent precipitate to spread out while settling. No identified concentration mechanism.

co-precipitates with dissolved metals. The precipitate will be a flocculent, low density solid that

will be easily transported by currents in the supernatant liquid. Induced currents will easily disturb these suspended, flocculent solids and will carry them throughout the supernatant layer.

Between each transfer of acid, there will be a period of several days or more until the next transfer. This will provide time for the solids to slowly settle to the bottom of the tank. No determination has been made of the time required for the flocculent particulate to reach the bottom of the tank. However, the time will be more than long enough for the suspended particulate to spread over the entire area of the tank. The layer of solids formed should have a more-or-less uniform depth over the entire floor of the tank. After completion of transfer, the solids are expected to form a layer no more than about 0.61 m (24 in.) deep at any location.

If it is hypothesized that a mounding of solids were to form, the mound would have the following characteristics:

- The solids would have a low bulk density and would be easily disturbed by currents in the supernate.
- The high point of mounded solids should be almost directly below the point of entry.
- The mound would have a very low angle of repose.
- The plutonium areal density would be low over the area of the mound.

6.4 MAINTAINING AN ADEQUATE ABSORBER-TO-PLUTONIUM MASS RATIO

The primary parameter that provides assurance of subcriticality is the sum of the absorber-to-plutonium subcritical mass fractions. In both tanks the total subcritical mass fraction in the supernatant liquid exceeds 4000 due to the very low concentration of dissolved plutonium. Criticality in the supernatant prior to receipt of acid waste is not credible.

Table 6-2 lists values of parameters important to criticality safety for the sludge in SST C-106 prior to acid dissolution. Criticality is not credible in DST C-106 before the addition of acid due to the low plutonium concentration and high total absorber-to-plutonium subcritical mass fraction.

Table 6-2. BBI Parameters for SST C-106 Sludge

Component	Best Estimate
Starting Inventory	
Plutonium	1.9 kg
Iron	4514.0 kg
Manganese	1121.7 kg
Uranium	60.8 kg
Starting Average Concentration	
Plutonium	0.056 g/L
Starting Subcritical Mass Fractions	
Sum of Insoluble	36.5
Iron	14.7
Manganese	18.3
Uranium	0.04

6.4.1 Gravity Segregation

The acidic solution entering DST AN-106 mixes with the caustic supernatant liquid to form a low-density, flocculent precipitate that sinks very slowly to the bottom. During the settling process particulate will separate according to their settling rates. The particles that are denser will settle at a faster rate. Because of the different settling rates, the waste will be formed into layers according to the relative size and density of particles. This process of segregating particles according to size is called gravity segregation, and it provides a mechanism for concentrating plutonium. If the plutonium-bearing particles are formed with a relatively uniform size and density, then a plutonium-rich layer might form in the settled solids. *The Potential for Criticality in Hanford Tanks Resulting from Retrieval of Tank Waste,* (Whyatt et al. 1996) concluded that gravity segregation might increase the plutonium-to-solids mass ratio by a factor of 3 for tank waste solids settling through a liquid layer. In other words, a pile of homogeneous waste dropped into a layer of water might settle into a pile that is not homogeneous. The plutonium concentration within a layer inside this pile might be 3 times higher than the concentration in the original pile. However, this layer of highest plutonium concentrations is very likely to be thin.

Using BBI data for the SST C-106 sludge, the total water-insoluble absorber-to-plutonium subcritical mass fraction is estimated to be 36. If this sludge were broken up into very small particles and dropped into the DST AN-106 supernatant, it might be possible for the absorber-to-plutonium mass ratio to decrease by a factor of 3 within a layer of settled solids. The resulting absorber-to-plutonium subcritical mass fraction might drop to 12 within this layer. A total absorber-to-plutonium subcritical mass fraction of 12 nevertheless provides a large margin of subcriticality. The probability of reducing the absorber-to-plutonium mass fraction to less than 1 by gravity segregation alone is extremely unlikely. For such a high degree of concentration to occur over a slab thickness and also over an area large enough to result in criticality is not credible. Before criticality would be possible the dissolution process must provide an additional mechanism that is effective at concentrating the plutonium.

6.4.2 Dissolution of Waste Components

Acid dissolution provides another potential concentration mechanism. Differences both in dissolution rates of waste components and precipitation rates might result in a composition for the settled solids in DST AN-106 that is markedly different from the original composition. In a worst-case scenario the plutonium dissolution rate might be low, while the absorber dissolution rate might be high. Under these conditions, all of the neutron absorbers might be transferred, while most of the plutonium remains behind. In another worst-case scenario the opposite might be postulated in which all of the plutonium is transferred without any of the neutron absorbers. The ability of the oxalic acid to dissolve plutonium and to dissolve waste components is important.

Iron, manganese, and uranium are selected as important neutron absorbers that occur in relatively large quantities. If an adequate margin of subcriticality can be demonstrated for these selected components alone, the effect of the other components need not be evaluated. This results in a simplification of the evaluation. At the same time the justification for subcriticality is conservative, because other waste components also contribute significantly to neutron absorption and help ensure subcriticality.

Table 6-3 shows the minimum, average, and maximum concentrations for plutonium, uranium, iron, and manganese when SST C-106 and DST AY-102 sludge samples are dissolved in oxalic acid. These values were supplied by Herting (2003d). The mass ratios were then derived from the concentrations. This information is described in greater detail in Section 2.3 and in Appendix B.

Table 6-3. Concentrations and Mass Ratios in Oxalic Acid Solution

	Pu (g/L)	U (g/L)	Fe (g/L)	Mn (g/L)	U/Pu	Fe/Pu	Mn/Pu
SST C-106 Samples							
Minimum	0.00101	0.010	0.444	0.654	7.9	350	515
Average*	0.00116	0.018	1.587	1.667	15	1370	1440
Maximum	0.00127	0.025	2.730	2.680	21	2260	2210
DST AY-102 Samples							
Minimum	0.00052	0.050	2.970	1.690	57	1400	1920
Average	0.00100	0.103	5.068	2.612	103	5070	2610
Maximum	0.00212	0.250	8.040	4.270	142	12000	3780

Reference: Herting (2003d). See Appendix B2.1 for more detail.
*The average for SST C-106 samples is taken to be the Best Estimate.

This data show a relatively large spread in component concentrations. The question of uncertainty in the Best Estimate for a waste component will be discussed later.

The plutonium concentration is the most important parameter. If the plutonium concentration is very small, only small amounts of plutonium will be transferred with the other components. If this were to happen, sluicing might be required to suspend solids to ensure that

they are transferred. The last transfer might then contain a much lower absorber-to-plutonium mass ratio. This scenario would complicate criticality safety.

The most probable plutonium concentration in solution was estimated to be 0.00116 g/L. At this concentration the total plutonium transferred in 795 kL (210 kgal) of oxalic acid solution would be 922 g. The lower bound on the plutonium transferred would be several hundred grams smaller. In other words, the quantity of plutonium dissolved in the oxalic acid is not sufficient to ensure transfer all of the plutonium. Complete transfer of this waste requires the transfer of a relatively large proportion as suspended solids.

6.4.3 Transfer of Suspended Waste Solids

The process of dissolution will release gas and agitate the mixture. During this process fine particulate will separate from the hard sludge and become suspended in the liquid. Phase II of the chemical analysis measured the quantity of residual solids (i.e., residue). After allowing time for dissolution of the waste sample to be completed, the residue was separated from the oxalic acid by centrifuging. The results of the Phase II analyses are reported in Appendix C and summarized in Appendix B2.2. The fine particles generated in the acid dissolution process should remain suspended for a relatively long time. They would be easily carried along with the liquid pumped from SST C-106. This evaluation assumes that the residue content reported in Appendix C provides a reasonable estimate of the composition of suspended solids transferred with the acid solution.

Table 6-4 shows the minimum, average, and high values reported for the plutonium, iron, and manganese concentrations. The average values are taken to be the Best Estimate of the suspended solids. The uranium concentration was measured to be very low and was omitted from the data. The uncertainty associated with the concentrations is of the order of 50 %. The Best Estimate of the plutonium concentration in the residual solids is 10 times larger than for the dissolved plutonium. These residual solids consist of suspended solids and settled solids. The suspended solids clearly play an important role in the transfer. Although the fraction of the residual solids that are suspended is not known, the composition of the residual and suspended solids are the same. The smallest subcritical mass fraction in the residual solids was found to be 12.7 for iron and manganese alone. These two components therefore provide sufficient neutron absorption to ensure subcriticality in the suspended solids

Table 6-4. Analytical Results for Residual SST C-106 Solids (Phase II)

	Pu (g/L)	Fe (g/L)	Mn (g/L)	Fe/Pu	Mn/Pu	Subcritical Fraction
Minimum	0.00443	2.77	1.25	625	282	12.7
Average*	0.01210	6.59	5.74	545	474	18.2
High	0.02610	12.73	13.80	488	529	19.6

Reference: Appendix C (Herting 2003e)
*The average concentration of SST C-106 samples is taken to be the Best Estimate concentration.

The expected constancy of the uranium-to-plutonium mass ratio is contradicted by the chemical analysis results. The BBI data shows the U/Pu mass ratio in SST C-106 sludge to be

32, and this equates to a subcritical mass fraction of 0.04. Herting (2003e) indicates that the U/Pu mass ratio in transferred waste is small. Therefore uranium cannot be counted as an absorber in the transferred waste. In addition, the total quantity of uranium is too small to contribute much to subcriticality. Nevertheless, the uranium does increase the margin of subcriticality for the waste that does not get transferred.

The Best Estimate of the absorber-to-plutonium mass ratios in residual solids is higher than the minimum required to ensure subcriticality. The absorber-to-plutonium mass ratios in the suspended solids sent to DST AN-106 would be expected to be the same as for the residual solids as a whole. Therefore, the transferred waste has a subcritical mass fraction for iron of 3.4 and for manganese of 14.8, making a total subcritical mass fraction of 18.2. If one assumes gravity segregation occurs during settling through the supernate layer, then the final subcritical mass fraction in the settled solids might drop by a factor as high as 3. This might result in a layer of waste that has a subcritical mass fraction as low as 5.5. This remains sufficient to guarantee subcriticality. The final absorbers mass ratio is found to be adequate to ensure that criticality is incredible during the precipitation and settling process.

If the quantity of iron and manganese transferred were to be only half as great as assumed, then the subcritical mass fraction in the settled solids might be as low as 2.7. This remains sufficient to ensure subcriticality, based solely on the absorber-to-plutonium mass ratios in the transfers.

6.4.4 Waste Remaining in SST C-106

Table 6-5 shows four scenarios in which the quantities of components remaining after each transfer are calculated.

Scenario 1 is the "Best Estimate" scenario in which all parameters are assumed to be Best Estimates. The BBI provided the starting quantities. For the Best Estimate the quantity of plutonium, iron, and manganese in the residual solids is 10.4, 4.1, and 3.4 times larger, respectively, than the quantity in solution. These same ratios will apply to suspended solids that are transferred and to settled solids that are not. The suspended solids are expected to dominate in the transfers. When all residual solids are assumed to be suspended, only two transfers of 76 kL (20 kgal) each are required to remove the entire plutonium inventory. After the first transfer, the proportion of absorbers increases. At no time does the total subcritical mass fraction for the waste remaining in SST C-106 fall below its original value of 33. This large proportion of absorbers ensures that criticality remains incredible in SST C-106.

Scenario 2 increases the plutonium inventory by 253% to 4.87 kg. This is the 99% upper bound on the plutonium inventory for SST C-106. Five transfers are required to remove all plutonium. In this scenario the initial total subcritical mass fraction for iron and manganese is 13.0. The total subcritical mass fraction then decreases to 7.3 after two flushes. Each flush after

Table 6-5. Material Remaining in SST C-106 after 76 kL (20 kgal) Transfers

Scenario 1		Pu	U	Fe	Mn	Mass Fraction	
C-106 Inventory (kg)		1.92	60.8	4514.0	1121.7	Fe	Mn
Solution Conc.(g/L)		0.00116	0.018	1.587	1.667	8.6	45.0
Suspended Conc.(g/L)		0.01210	0.000	6.59	5.74	3.4	14.8
Transfer	Vol. (L)	Pu (kg)	U (kg)	Fe (kg)	Mn (kg)		
0	0	1.92	60.8	4514.0	1121.7	14.7	18.3
1	76	0.92	59.4	3894.9	560.9	26.6	19.1
2	152	0.00	58.1	3275.9	0.2	large	large
99% Upper Bound Plutonium Inventory							
Scenario 2		Pu	U	Fe	Mn	Mass Fraction	
C-106 Inventory (kg)		4.87	60.8	4514.0	1121.7	Fe	Mn
Solution Conc.(g/L)		0.00116	0.018	1.587	1.667	8.6	45.0
Suspended Conc.(g/L)		0.01210	0.000	6.59	5.74	3.4	14.8
Transfer	Vol. (L)	Pu (kg)	U (kg)	Fe (kg)	Mn (kg)		
0	0	4.87	60.8	4514.0	1121.7	5.8	7.2
1	76	3.87	59.4	3894.9	560.9	6.3	4.5
2	152	2.86	58.1	3275.9	0.2	7.2	0.1
3	227	1.86	56.7	2656.8	0.0	8.9	0.0
4	303	0.85	55.3	2037.7	0.0	14.9	0.0
Half the Plutonium Transfer Rate in Suspended							
Scenario 3		Pu	U	Fe	Mn	Mass Fraction	
C-106 Inventory (kg)		1.92	60.8	4514.0	1121.7	Fe	Mn
Solution Conc.(g/L)		0.00116	0.018	1.587	1.667	8.6	45.0
Suspended Conc.(g/L)		0.00605	0.000	6.59	5.74	3.4	14.8
Transfer	Vol. (L)	Pu (kg)	U (kg)	Fe (kg)	Mn (kg)		
0	0	1.92	60.8	4514.0	1121.7	14.7	18.3
1	75.7	1.37	59.4	3894.9	560.9	17.7	12.8
2	151.4	0.83	58.1	3275.9	0.2	24.7	0.0
3	227.1	0.28	56.7	2656.8	0.0	58.8	0.0
Half Plutonium Transfer Rate in Suspended and 56% Increase in Plutonium Inventory							
Scenario 4		Pu	U	Fe	Mn	Mass Fraction	
C-106 Inventory (kg)		3.00	60.8	4514.0	1121.7	Fe	Mn
Solution Conc.(g/L)		0.00116	0.018	1.587	1.667	8.6	45.0
Suspended Conc.(g/L)		0.00605	0.000	6.59	5.74	3.4	14.8
Transfer	Vol. (L)	Pu (kg)	U (kg)	Fe (kg)	Mn (kg)		
0	0	3.00	60.8	4514.0	1121.7	9.4	11.7
1	75.7	2.45	59.4	3894.9	560.9	9.9	7.1
2	151.4	1.91	58.1	3275.9	0.2	10.7	0.0
3	227.1	1.36	56.7	2656.8	0.0	12.2	0.0
4	302.8	0.82	55.3	2037.7	0.0	15.6	0.0
5	378.5	0.27	54.0	1418.7	0.0	32.8	0.0

the second increases the subcritical mass fraction. The iron content by itself remains sufficient to ensure subcriticality until all plutonium has been transferred.

Scenario 3 assumes that the plutonium concentration in suspended solids is half of the Best Estimate. All 1.92 kg of plutonium is removed in 3 transfers. After the first transfer, the proportion of absorbers increases. At no time does the total subcritical mass fraction for iron and manganese fall below a value of 24.7.

Scenario 4 increases the plutonium inventory to 3.0 kg and decreases the plutonium concentration in suspended solids by half. The initial total subcritical mass fraction for iron and manganese is 21. After the second transfer, all of the manganese is gone. However, iron-to-plutonium mass ratio increases. The total subcritical mass fraction drops to 10.7 after the second transfer, but it increases after each subsequent transfer. In all four scenarios there is always enough absorbers remaining in SST C-106 to ensure that criticality is incredible.

If the SST C-106 manganese inventory were less than expected and/or the transfer rates were higher, the iron alone would be enough to ensure subcriticality for 1.92 kg of plutonium (i.e., the normal inventory). If the iron were half the BBI inventory, there would still be enough to ensure subcriticality for the normal plutonium inventory at the expected transfer rates.

Based on Best Estimates, it is concluded that the proportion of iron and manganese transferred is sufficient to ensure subcriticality during the precipitation and settling process in DST AN-106. At the same time, a failure to transfer the solids that sloughed off of the hard sludge would not create a potential for criticality in SST C-106. This is true because they should contain the same quantities of iron and manganese in relationship to the plutonium as was determined by Herting (2003e) for the residual solids.

6.5 PLUTONIUM CONCENTRATION IN SOLUTION

Plutonium Reconversions, (Harmon et al. 1961) states that the plutonium (IV) saturation concentration in oxalic acid is about 0.06 g/L at 50° C and it can be as high as 1.16 g/L at 75° C (see Appendix B). Even the higher of these values is well below the minimum concentration that can be made critical in a hydrogenous solution. For oxalic acid the minimum critical concentration appears to be slightly less than for water. In an infinite volume the minimum plutonium concentration is approximately 7 g/L. At the same time, criticality at this concentration requires an infinite mass of plutonium.

For 2.0 kg of plutonium, the minimum plutonium concentration for which criticality is possible is about 10 g/L in pure water (Carter et al. 1969, III.A.6.100-3). For oxalic acid the minimum critical concentration would be very nearly the same. This means that the plutonium concentration would have to increase by a factor of at least 10, and possibly more than 100, above the saturation concentration before criticality is possible. To reach such a high concentration most of the plutonium would have to be present as suspended particulate. The highest plutonium concentration that Herting (2003e) found in suspended particulate was

0.0308 g/L. The plutonium minimum critical concentration is more than 300 times larger than this largest value.

Before criticality becomes possible, the entire plutonium inventory of SST C-106 or DST AN-106 would have to be concentrated into a compact volume. To achieve a the required high plutonium concentration would require that the following mechanisms occur:

(1) Oxalic acid in SST C-106 would have to dissolve or suspend a large fraction of the available plutonium within a single transfer of acid.

Discussion: If it were assumed that the entire plutonium inventory of SST C-106 (i.e., 1.92 kg) was suspended in a single transfer of 76 kL (20 kgal) of acid solution, the average plutonium concentration would be 0.025 g/L. It would be extremely unlikely for such a large plutonium mass to be contained in a single transfer.

After reaching DST AN-106, this concentration would have to be increased 400 times to reach the minimum concentration of 10 g/L for which criticality is possible.

(2) Dissolved plutonium in the incoming waste precipitates upon entering the supernate.

Discussion: This is assumed to happen. The precipitate would form very fine particulate that would remain suspended for a long period of time. This would allow settling over a large area.

(3) Most particulate settles onto a relatively small area.

Discussion: The momentum of the incoming acid stream would generate currents within the supernatant liquid that would carry the suspended and newly precipitated particulate along with it. This particulate would be very small and the settling rate would be slow. At the same time it would spread horizontally over a large area. It is likely that the waste solids would settle more or less uniformly over the entire bottom of the tank.

(4) Settling rate must be rapid enough to prevent dispersal, but slow enough to remain suspended until enough plutonium has been discharged.

Discussion: This would require particulate arriving first to settle at a slower rate than the particulate arriving later. It would be extremely unlikely for this to occur.

Although the DST AN-106 supernatant liquid already contains plutonium, its concentration is very low and would not contribute significantly towards reaching the minimum critical concentration.

It is not credible for the plutonium concentration in the supernatant liquid to even approach 10 g/L, even in a small volume. Based upon the maximum plutonium concentration that might be achieved, criticality in the supernatant liquid is not credible.

6.6 MAXIMUM PLUTONIUM CONCENTRATION IN SETTLED SOLIDS

Even if it is assumed that the plutonium is separated from the neutron absorbers, it is necessary for the plutonium concentration to exceed the minimum critical concentration before criticality is possible. The Conservative Waste Model (CWM) defines a waste composition that results in a smaller plutonium critical concentration than will occur in any real waste (Rogers 1993). The subcritical limit plutonium concentration for CWM solids is 2.6 g/L. The average plutonium concentration in SST C-106 solids is 0.056 g/L, based on the BBI inventory of 1.9 kg in 34 kL (9 kgal) of sludge. To reach the minimum concentration that might be made critical in dry CWM solids the plutonium concentration in the settled solids in DST AN-106 would have to be 46 times greater than the average concentration. Because of the tendency of the acid waste entering DST AN-106 to spread, it would be extremely unlikely for the settled solids to achieve a plutonium concentration as high as 2.6 g/L even over a small area. Even if this were to occur, the small plutonium mass of 1.9 kg would not be large enough for criticality.

In reality, the presence of moderators, especially water and oxalic acid, would increase the plutonium concentration required before criticality is possible. To achieve criticality with 2 kg of plutonium, it would necessary to have a high degree of water moderation and for the plutonium concentration to be at least 10 g/L in a compact volume. It is not credible for the settled solids to achieve a plutonium concentration high enough for criticality. In addition, the layer of high concentration would be relatively thin and the mass of plutonium required for criticality would be greatly increased. It would not be credible to form a layer of settled solids for which criticality would be possible.

6.7 EXCEEDING MINIMUM CRITICAL MASS

For criticality to occur, it would be necessary for more than a critical mass of plutonium to be concentrated into a small compact volume. As shown above, it would be extremely unlikely to achieve the minimum critical concentration. Before criticality would be possible, the mass of plutonium above the minimum critical concentration would also have to exceed the critical mass. This requirement further reduces the probability of criticality below that which is incredible.

The entire 1.92 kg of plutonium must be concentrated into a compact volume of only 192 L. If this volume formed a cube, it would be only 58 cm (1.9 ft) on a side. The volume of the supernatant layer in DST AN-106 is about 1,482,000 L. The average plutonium concentration in the supernatant liquid would be only 0.0013 g/L. For criticality to be possible, the plutonium would have to concentrate into only 0.013% of the available volume before settling into a layer. It is not credible for this to occur.

6.8 FINAL STATE OF SST C-106

The goal of SST C-106 retrieval is to leave less than 10.2 m^3 (360 ft^3) of waste in SST C-106, including both the liquid and the solids portion. Laboratory testing has shown that not all of the waste will dissolve. Once the remaining solids volume falls below 10.2 m^3 (360 ft^3), no

further addition of acid will be made. It is therefore assumed that the final volume of solids will be at least 7.1 m^3 (250 ft^3), but less than 10.2 m^3 (360 ft^3). Due to chemical action the solids are expected to be broken up into particles that resemble sand and to form a layer less than 15.2 cm (6 in.) deep. The final volume of solids should be less than 25% of the initial volume.

The plutonium inventory in SST C-106 after completion of the acid dissolution and transfer process is expected to be less than the minimum critical mass of 530 g of plutonium. This plutonium will be combined with at least 7.1 m^3 (250 ft^3) of solids at an average plutonium concentration no greater than 0.075 g/L. If spread uniformly over the floor of the tank the areal density would be about 0.0013 kg/m^2 (0.12 g/ft^2). The areal density of the plutonium will be far less than the minimum critical areal density of 2.6 kg/m^2 (240 g/ft^2) (Carter et al. 1969, III.A.8.100-3). The average areal density will be about 2,000 times smaller than the minimum critical areal density. The suspended solids that remain in SST C-106 will have a subcritical mass fraction as large as those that are transferred, or at least 16.4.

In the worst-case scenario for SST C-106 in which none of the plutonium is assumed transferred, the plutonium remaining would be 4.87 kg and the average plutonium concentration in the solids would be no greater than 0.68 g/L, assuming at least 7.1 m^3 (250 ft^3) of solids. The average areal density will be about 217 times smaller than the minimum critical areal density. Even under these conditions, subcriticality would be assured.

The high proportion of absorbers, the low plutonium areal density, and the small slab thickness will ensure that criticality is not credible for the waste that remains in SST C-106.

6.9 SUMMARY AND CONCLUSIONS

The acid dissolution process used is similar to the process used to generate the original waste at the processing facility. In addition, the process of precipitation in DST AN-106 is similar to the process used to precipitate the original solids in SST C-106. For these reasons, the composition of the settled waste solids in DST AN-106 is expected to be close to the composition of the sludge in SST C-106.

The dissolved components were found to remove a high proportion of neutron absorbers, while leaving plutonium behind. The suspended solids were found to have a significantly higher concentration of plutonium, iron, and manganese. In particular, the plutonium in suspended particulate was estimated about 15 times higher than in solution. Therefore, the data for suspended solids is of special importance for the justification of incredibility. For this evaluation the Best Estimate of suspended components was taken to be the average of the SST C-106 residue concentrations from Herting (2003e). The concentration of residue was assumed to be a reasonable estimate of the concentration of suspended solids in the transferred waste. These values have a relatively large uncertainty. Nevertheless, the large absorber-to-plutonium mass ratios for iron and manganese in the SST C-106 sludge, based on the BBI inventory, provides a large enough margin to balance uncertainties and to make criticality incredible.

Currents generated by the incoming acid waste falling into in the DST AN-106

supernatant liquid provide an effective mechanism to disperse precipitated and suspended particulate over a wide area. The small size of the particulate causes it to settle slowly and allows it to disperse. This particulate is expected to settle over the entire tank. The 1.9 kg of plutonium in SST C-106 corresponds to an average areal density of 0.0046 g/m^2 (0.43 g/ft^2). This is 500 times smaller than the minimum critical areal density of 2.6 kg/m^2 (240 g/ft^2). The greatest areal density at any location should not be more than an order of magnitude higher than the average. This would still be far lower than the minimum critical value.

At the 99% confidence level, the upper bound on the plutonium inventory in SST C-106 is 4.87 kg. The high proportion of neutron absorbing solids is judged sufficient to ensure subcriticality, even if the upper bound inventories were assumed. Before receiving waste from SST C-106, the plutonium inventory for DST AN-106 is 1.27 kg, with almost all plutonium locked up in the saltcake layer. The average concentration in the saltcake is very low. Interaction between the saltcake layer and the incoming plutonium is negligible.

Consideration was given to the high proportion of neutron absorbing components, the transfer rates, the small particulate size, the low plutonium concentrations in oxalic acid and in water, and the tendency to disperse in the supernatant. Based upon these considerations, it was concluded that criticality in DST AN-106 caused by the SST C-106 waste transfers is not credible.

In addition to the large absorber-to-plutonium mass ratio, there are two barriers that demonstrate that criticality is incredible. The first is the need to achieve a minimum critical concentration. After reaching DST AN-106, the plutonium concentration would have to increase by more than 400 before criticality becomes possible. In addition, nearly the entire plutonium content from SST C-106 would have to be brought together in a volume of about 192 L, which is only 0.0013% of the volume of the supernate. It is not credible to achieve a configuration in DST AN-106 for which criticality is credible, even if the proportion of neutron absorbers were far less than expected.

The settled solids that remain in SST C-106 will have the same high proportion of neutron absorbing components as the suspended solids that are transferred to DST AN-106. The associated total subcritical mass fraction is estimate to be at least 16.4, a quantity far larger than required to ensure subcriticality. In addition, the plutonium areal density in the remaining waste is estimated to be 2,000 times less than the minimum critical areal density. Based upon these considerations, it was concluded that criticality in SST C-106 is not credible.

After reviewing the range of credible configurations that might occur in SST C-106, in the transfer lines, and in DST AN-106, it is concluded that criticality is not credible for the acid dissolution and transfer process.

7.0 DESIGN FEATURES AND ADMINSTRATIVELY CONTROLLED LIMITS

7.1 LIMITS AND CONTROLS

The following limits and controls shall be used for the transfer of waste from SST C-106 to DST AN-106:

(1) No fissionable material bearing waste shall be added to SST C-106.

Basis: The primary parameter that determines criticality safety is the tank plutonium inventory. A smaller plutonium inventory provides a high margin of subcriticality.

Implementation: The only materials added to the tanks would be water, oxalic acid, and caustic. There is no credible way by which fissionable-bearing waste might be added except for acid waste flowing back from the transfer line. Since this waste originated in SST C-106, its return would not be considered a violation of this limit.

(2) The pH in DST AN-106 liquid waste shall be maintained above 8.

Basis: Maintaining a high pH in the DST AN-106 supernatant liquid will prevent fissionable material in the saltcake from dissolving and it will ensure that most of the plutonium dissolved in incoming waste will precipitate along with neutron absorbing components.

Implementation: This limit is implemented by procedural controls that will ensure that sufficient caustic has been placed in DST AN-106 before the transfer to neutralize the total volume of acidic waste received. The addition of caustic will be independently verified prior to receipt of acidic waste.

(3) The pH of SST C-106 liquid waste is not limited during transfer. After completion of transfers, caustic shall be added to ensure a pH greater than 8.

Basis: The addition of oxalic acid to SST C-106 is required to breakup and dissolve the waste components so they can be transferred to DST AN-106. The proportion of neutron absorbing solids in the waste is known to be far higher than required to ensure subcriticality. In addition, he average plutonium concentration in the liquid has been determined to be far less than the minimum that can be made critical. After completion of the transfers, caustic is added to return the waste to a state in compliance with the criticality safety requirements for tank waste.

Implementation: This limit is implemented by procedural controls that will ensure that sufficient caustic has been placed in SST C-106 after completion of the transfers to neutralize the acid and increase the pH to more than 8. The addition of caustic will be independently verified.

(4) The acid added to SST C-106 shall be oxalic acid.

Basis: This evaluation has been based upon the assumption that the acid to be used would be oxalic. The transfer rates assumed are for oxalic acid. The use of a different acid

would change the transfer rates and invalidate the conclusions of this evaluation.

Implementation: This limit is implemented by procedural controls that will ensure that the acid is oxalic. The acid type used will be independently verified prior to its use.

(5) The total volume of oxalic acid added to SST C-106 shall not exceed 795 kL (210 kgal).

Basis: The volume of acid used directly determines the quantity of caustic that must be added to DST AN-106 to neutralize the incoming acid. The need to use a larger volume of acid would only occur if the waste solids were not being broken up and dissolved as expected. This would create uncertainty concerning the accuracy of the reported dissolution and transfer rates and call into question the accuracy of the evaluation.

Implementation: This limit is implemented by procedural controls. The volume of acid used in each transfer will be independently verified. When the total volume of acid reaches 795 kL (210 kgal), the activities will stop and a review made of the status.

(6) Acid waste from SST C-106 shall not be transferred, either accidentally or on purpose, to any tank other than DST AN-106.

Basis: The discharge of acid into any waste storage tank without prior approval is forbidden. The chemistry studies of tank waste have not evaluated the consequence of acid dissolution of waste, and this has been identified as a means by which a criticality unsafe condition might be created.

Implementation: This limit is implemented by procedural controls. Routes by which the acidic waste might end up in a tank other than DST AN-106 are to be determined prior to the first transfer of waste. The possibility of connection to the wrong tank and the possibility acid waste draining back through lines different than originally taken are to be examined. Before the first transfer of waste, assurances must be provided that the proper connections have been made and that no accident condition might arise in which the acidic liquid would enter a tank other than SST C-106 or DST AN-106.

(7) The transfer of tank waste between SST C-106 and DST AN-106 must be made through a line that does not connect to any other tanks and that cannot drain back to any tank farm equipment, except that which is connected to these two tanks. Independent verification of the transfer line connections and routing must be made by at least two people.

Basis: The discharge of acid into any waste storage tank without prior approval is forbidden. The chemistry studies of tank waste have not evaluated the consequence of acid dissolution of waste, and this have been identified as a means by which a criticality unsafe condition might be created.

Implementation: This limit is implemented by procedural controls. Verification of a proper connection to DST AN-106 must be made independently by at least two people prior to the first transfer of acidic waste.

(8) No waste, other than waste from DST AN-101, shall be transferred to DST AN-106 prior to receipt of SST C-106 acid waste.

Basis: This evaluation is based upon the known contents of DST AN-106. If waste were to be transferred to DST AN-106 from any tank other than DST AN-101, the conditions under which this evaluation was made would no longer be valid.

Implementation: This limit is implemented by procedural controls.

7.2 PASSIVE DESIGN FEATURES

No passive design features were used on this evaluation.

7.3 ACTIVE DESIGN FEATURES

No active design features were used on this evaluation. No criticality safety alarm system is required.

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APPENDIX A

INDEPENDENT REVIEW

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Desk Instruction 2.0, Rev. 1

CHECKLIST FOR TECHNICAL PEER REVIEW

Document Reviewed - HNF-15682, Revision 0

Title: CSER 03-011: Transfer From Tank 241-C-106 To Tank 241-AN-106 Using Oxalic Acid Dissolution

Author: C. A. Rogers

Date: April 2003

Yes	No*	NA		
[X]	[ ]	[X]		Referenced analyses appropriate.
[X]	[ ]	[ ]		Problem completely defined and all potential configurations considered.
[X]	[ ]	[ ]		Accident scenarios developed in a clear and logical manner.
[X]	[ ]	[ ]		Necessary assumptions explicitly stated and supported.
[ ]	[ ]	[X]		Computer codes and data files documented.
[X]	[ ]	[ ]		Data used in calculations explicitly stated in document.
[X]	[ ]	[ ]		Data checked for consistency with original source information as applicable.
[X]	[ ]	[ ]		Mathematical derivations checked including dimensional consistency of results
[X]	[ ]	[ ]		Models appropriate and used within range of validity, or use outside range of established validity justified.
[X]	[ ]	[ ]		Hand calculations checked for errors. Spreadsheet results should be treated exactly the same as hand calculations.
[ ]	[ ]	[X]		Software input correct and consistent with document reviewed.
[ ]	[ ]	[X]		Software output consistent with input and with results reported in document reviewed.
[X]	[ ]	[ ]		Limits/criteria/guidelines applied to analysis results are appropriate and referenced. Limits/criteria/guidelines checked against references.
[X]	[ ]	[ ]		Safety margins consistent with good engineering practices.
[X]	[ ]	[ ]		Conclusions consistent with analytical results and applicable limits.
[X]	[ ]	[ ]		Results and conclusions address all points required in the problem statement.
[X]	[ ]	[ ]		Format consistent with applicable guides or other standards.
[X]	[ ]	[ ]	**	Review calculations, comments, and/or notes are attached.
[X]	[ ]	[ ]		Document approved (for example, the reviewer affirms the technical accuracy of the document).

D. G. Erickson D. G. Erickson 04/29/03
Technical Peer Reviewer (printed name and signature) Date

* All "no" responses must be explained below or on an additional sheet.

** Any calculations, comments, or notes generated as part of this review should be signed, dated and attached to this checklist. The material should be labeled and recorded in such a manner as to be understandable to a technically qualified third party.

Peer Review Comments

D. G. Erickson, a qualified Criticality Safety Specialist, of the Criticality and Shielding group in FFS Safety Analysis and Nuclear Engineering carried out an independent, technical review of Revision 0 of HNF-15682, *CSER 03-011: Transfer From Tank 241-C-106 To Tank 241-AN-106 Using Oxalic Acid Dissolution*. This was a complete review of the documentation including the appendices and calculations.

The technical arguments, and the basis for those arguments given in the report were found to be sound for qualifying the criticality safety of the transfer of the tank 241-C-106 contents to tank 241-AN-106, utilizing appropriate controls presented in Section 7.

The documentation provides a logical progression of the transfer, and the criticality incredibility argument for each step of the transfer, including the starting and ending state in both tanks. The arguments provided are only applicable to this operation on the specified tanks. If any other tank contents are involved, the justification of criticality incredibility is not valid.

The document went through several very significant evolutions to reach the final state. With a better understanding of the processes involved, and more appropriate data for the tanks and oxalic acid dissolution/suspension process, the justification for criticality incredibility was strengthened and simplified.

The analysis performed was conservative, and showed the transfer operation remains significantly subcritical at all times. There were no credible contingencies identified that did not maintain the criticality incredibility of the system.

The report was reviewed for technical accuracy, consistency, coverage of all credible contingencies, and adequacy of limits. Discrepancies, inconsistencies, editorial presentation issues and technical issues, etc., were raised and have been adequately resolved.

This reviewer affirms that based on the analysis contained in CSER 03-011, HNF-15682, Rev. 0, the transfer of the contents of tank 241-C-106 to tank 241-AN-106, with the appropriate controls, is safe from a nuclear criticality standpoint, and a criticality is incredible.

APPENDIX B

COMPILATION OF CHEMISTRY DATA

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COMPILATION OF CHEMISTRY DATA

Chemical analyses were made of sludge grab samples taken from SST C-106 in February 2003 and from other tanks to determine the concentrations of key components during the various phases of the waste transfer. A compilation is made of those components that are of greater importance to this criticality safety evaluation.

B1.0 DATA FROM REACTOR HANDBOOK

Harmon et al. (1961) describe the chemistry of plutonium oxalate and provides a typical flowsheet for the precipitation of plutonium (IV) oxalate. Precipitation is described as follows:

> The compound has been precipitated satisfactorily from solutions containing 1-300 g Pu/liter and enough nitric acid to make the final slurry 1.5-4.5 M. At acid concentrations below 1.5 M the coprecipitation of impurities is favored, and the precipitate is too finely divided for rapid settling or filtration. At slurry acid concentrations above 4.5 M, plutonium (IV) oxalate solubility is high and the precipitate is thixotropic.

The concentration of plutonium in the precipitated slurry is given as 0.5 g/L.

Concerning the solubility of plutonium (IV) oxalate, Harmon et al. (1961) provide the following information:

> Equilibrium solubilities of plutonium (IV) oxalate are much lower than those obtained in the usual quick precipitation process, and vary both with the acidity and with free oxalic acid concentrations. The optimum range of the free oxalic acid concentration is 0.05-0.15 M, depending on the purity of the solution. (The presence of oxalate-complexing cations in appreciable concentration requires a large excess of oxalic acid.) Slurry temperatures also affect the solubility of plutonium (IV) oxalate; in one case measured values indicated about 0.05 g Pu/liter at 25° C, about 0.06 g Pu/liter at 50° C, and 0.4-1.16 g Pu/liter at 75° C.

Harmon et al. (1961) state that plutonium (III) oxalate, $Pu_2(C_2O_4)_3 \bullet 9H_2O$, has a low solubility of 3.24 $(H^+)^3(H_2C_2O_4)^{-3/2}$ mg Pu/liter.

B2.0 CHEMICAL ANALYSES

Herting (2003a, b, c, d, e) conducted tests to determine the ability of oxalic acid to dissolve sludge samples from SST C-106 and DST AY-102. The sludge in DST AY-102 was analyzed because SST C-106 contents were previously transferred to DST AY-102. The sludge in DST AY-102 should be similar to sludge in SST C-106.

Tests were performed in two Phases. Phase I measured only dissolved components. This

analysis was then extended in Phase II to suspended residues that were obtained by centrifuging the samples.

B2.1 PHASE I DISSOLVED COMPONENTS ANALYSES

Table B1 and B2 shows analytical results for SST C-106 solids dissolved in water, in oxalic acid, and in a mixture of oxalic acid and nitric acid (Herting 2003d).

The total alpha is assumed to be entirely generated by ^{239}Pu. The plutonium concentration is obtained by dividing by the specific activity of 0.06133 Ci/g for ^{239}Pu (Clow et al. 1994) and converting to g/L. The plutonium concentration dissolved in oxalic acid was found to be about 0.0013 g/L, a value much smaller than the saturation concentration of about 0.06 g Pu/liter at 50° C reported by Harmon et al. (1961).

Table B1. SST C-106 Component Concentrations in Solution (Phase I)

	A1	A2	A3	A4	A5	A6	A7	A8
Reagent	**15 mL H_2O**	**40 mL H_2O**	**40 mL H_2O**	**15 mL Oxalic**	**40 mL Oxalic**	**40 mL Oxalic**	**15 mL Ox/Nit**	**40 mL Ox/Nit**
S03T000	**373**	**374**	**375**	**376**	**377**	**378**	**379**	**380**
pH	9.9	10.0	9.9	----	1.4	----	----	0.8
Components in Units of mg/L								
Al	2.5	16	18	7700	6600	----	----	2790
Cr	1.3	0.6	0.2	53	84	----	----	136
Fe	<1	9	3.5	444	2730	----	----	2990
Mn	2.0	19	6	654	2680	----	----	3990
Na	6400	1780	1760	16200	10500	----	----	9280
U	16	5.5	<5	<10	<25	----	----	<50
Plutonium (SpA for ^{239}Pu = 0.06133 Ci/g)								
α (mCi/L)	0.000082	0.00016	0.000076	0.078	<0.074	0.062	0.13	<0.058
Pu (mg/L)	0.00134	0.00261	0.00124	1.27	<1.21	1.011	2.12	<0.946
Absorber-to-Plutonium Mass Ratio (X/Pu)								
Fe/Pu	> 746	3450	2820	350	>2260	----	----	>3160
Mn/Pu	1490	7280	4840	515	>2210	----	----	4220
U/Pu	11,900	2110	>4030	>7.9	21	----	----	53

Reference: Herting (2003d)

Table B2. DST AY-102 Component Concentrations in Solution (Phase I)

	B4	B5	B6	B7	B8	C4	C5	C6
Reagent	15 mL Oxalic	40 mL Oxalic	40 mL Oxalic	15 mL Ox/Nit	40 mL Ox/Nit	15 mL Oxalic	40 mL Oxalic	40 mL Oxalic
S03T000	400	401	402	403	404	424	425	426
pH	----	1.1	1.1	3.2	0.7	2.8	1.0	1.2
Components in Units of mg/L								
Al	8060	3840	4410	8490	4580	7470	3970	4190
Cr	148	104	216	150	169	155	144	150
Fe	2970	7240	8040	1400	5530	3150	4460	4550
Mn	4270	2010	2470	4180	5210	3330	1690	1900
Na	10900	4480	5000	12100	5480	10700	4420	4630
U	<250	<130	68	<530	<100	68	<50	51
Plutonium (SpA for ^{239}Pu = 0.06133 Ci/g)								
α (mCi/L)	0.13	0.056	<0.041	0.16	0.10	0.054	0.054	0.032
Pu (mg/L)	2.12	0.913	<0.670	2.61	1.63	0.88	0.88	0.52
Absorber-to-Plutonium Mass Ratio (X/Pu)								
Fe/Pu	1400	7930	12000	536	3390	3580	5070	8750
Mn/Pu	2010	2200	3690	1600	3200	3780	1920	3650
U/Pu	<118	<142	101	<203	<61	77	<57	98

Reference: Herting (2003d)

B2.2 PHASE II RESIDUE ANALYSES

Herting (2003e) analyzed Phase I samples A1, A2, A4, A5 from SST C-106 and samples B1, B3, B4, B7, and B8 from DST AY-102 for sludge dissolution residues. Appendix C provides an overview of the results to be published for Phase II in May 2003. Information in Appendix C is preliminary, and has not been reviewed or released. Nevertheless, it is the only available data and is used for this evaluation. A summary of the Phase II results is provided in Table B3.

The importance of the Phase II data is that it includes residual solids. The residual solids include suspended solids that are transferred and settled solids that are not transferred. Without accounting for the suspended solids, much of the plutonium would appear to be left behind in SST C-106. The solution concentrations derived from Appendix C data are the same as reported for Phase I. That is to say, the amount (mg or μCi) in the supernate is the Phase I value (μg/mL or μCi/mL) times the volume of liquid for that test (i.e., either 15 mL or 40 mL). The residue mass is the analytical value for the residue (in mg/g or μCi/g) times the final weight of centrifuged solids reported in the Phase I final report. Table B6 contains a summary of the concentrations and the derived mass ratios and subcritical fractions of selected components in the residue.

Table B3. Residual Solids Component Concentrations (Phase II)

Reagent	A4 (Orig) 15 mL Oxalic	A4 (Dup.) 15 mL Oxalic	A5 40 mL Oxalic	B4 15 mL Oxalic	Average
Components in Units of mg/sample*					
Fe	64.1	191	111	393	172
Mn	32.4	207	50	129	97
U	---	----	----	----	----
Components in Units of mg/L					
Fe	4270	12733	2770	26200	11470
Mn	2160	13800	1250	8600	6470
Plutonium (SpA for ^{239}Pu = 0.06133 Ci/g)					
α (mCi/sample)	5.2	24.0	10.9	28.7	----
α (mCi/L)	0.347	1.60	0.272	1.91	----
Pu (mg/L)	5.66	26.1	4.43	31.1	16.8
Absorber-to-Plutonium Mass Ratio (X/Pu)					
Fe/Pu	754	488	625	842	677
Mn/Pu	382	529	282	276	367
Subcritical Mass Fraction					
Fe	4.7	3.1	3.9	5.3	4.2
Mn	11.9	16.5	8.8	8.6	11.5
Total	16.6	19.6	12.7	13.9	15.7

*Reference: Herting (2003e)

B3.0 BEST BASIS INVENTORY DATA

An EXCEL® spreadsheet was used to calculate macroscopic absorption cross sections, actual-to-minimum subcritical mass ratios, and subcritical mass fractions using the Best Basis Inventory (BBI) downloaded from the Tank Waste Information System (TWINS) website. Spreadsheets were calculated for the supernatant layers in SST C-106, DST AN-106, and DST AN-101, the sludge layer in SST C-106, and the saltcake layer in DST AN-102.

The spreadsheets for SST C-106 are provided in Tables B4 (supernate) and B5 (sludge); the spreadsheets for DST AN-106 are provided in Tables B6 (supernate) and B7 (sludge); and the spreadsheet for DST AN-101 is provided in Table B8 (supernate). Because of the low plutonium concentrations in the supernatant liquid, the total plutonium content was very small and the absorber-to-plutonium mass ratios were very large.

Beginning with Column A on the left, the spreadsheet columns are labeled A through L. The columns contain the following information:

Column A identifies the tank and Column B gives the analyte name.

Columns C and D are labeled "Inventory." The inventory values were downloaded on April 1, 2003 from the Best Basis Inventory (BBI) in the TWINS database. In the spreadsheet the values is in Column C and the units are in Column D.

Column E is labeled "Ci/g (LA-12846)." This provides the conversion factor (i.e., the specific activity) used to convert from curies to grams. The specific activities are obtained from LA-12846, *Specific Activities and DOE-STD-1027-92 Hazard Category 2 Thresholds - LANL Fact Sheet* (Clow et al. 1994).

Column F lists the atomic weight of the analyte.

Column G shows microscopic neutron absorption cross sections taken from Parrington et al. (1996), *Nuclides and Isotopes*.

Column H is labeled "Mass Ratio X/Pu (Limit)." This provides subcritical limit absorber-to-plutonium (X/Pu) mass ratios (Rogers et al. 1996).

Column I shows the analyte mass in kg, based on the value in Column C.

Column J is labeled "Macroscopic Absorb XS." This is the macroscopic absorption cross section calculated from values in earlier columns. For waste component i:

$$\Sigma_i = (10^{-24} N)(m_i/V)(\sigma_{ai}/A_i)$$

$$= 0.6023(m_i/V)(\sigma_{ai}/A_i)$$

where Σ_i = macroscopic absorption cross section (Column J)

σ_{ai} = microscopic absorption cross section of component i in barns (Column G)
m_i = total mass of component i (Cell G38)
N = Avagadro's number = 6.023×10^{23} atoms/mole
A_i = atomic weight of component i (Column F)
V = volume of waste layer in liters (Cell F41)

The formula in the spreadsheet for aluminum (J12) is:

(I12/1000*F41)*(G12/F12)*(0.6023)

where I12 = mass of aluminum
F41 = volume of waste layer
G12 = microscopic absorption cross section for aluminum
F12 = atomic weight of aluminum

Column K is labeled "Mass Ratio X/Pu (Actual)."

For aluminum: X/Pu = I12/C38
where C38 = total fissile mass in kg

Column L is labeled "Actual/Min Mass Fraction."

For aluminum: Actual/Min Mass Fraction =K12/H12
where K12 = actual Al/Pu mass ratio
H12 = subcritical limit Al/Pu mass ratio

Table B4. Parameters for Supernatant Layer in SST C-106

Download Date: April 1, 2003

Supernatant Tank Name	Analyte	Inventory	Supernatant Ci/g (LA-12646)	Atom Wt.	Micro XS Absorption	Mass Ratio X/Pu (Limit)	Mass(kg)	Macroscopic Absorb XS	Mass Ratio X/Pu (Actual)	Actual/Min Mass Fraction
241-C-106	233U	0.00 Ci	9.842E-03	233.0	—	—	0.0			
241-C-106	235U	0.00 Ci	2.163E-06	235.0	—	—	0.5			
241-C-106	238U	0.03 Ci	3.364E-07	238.0	—	—	84.7			
241-C-106	238Pu	0.02 Ci	1.713E+01	238.0	—	—	0.0			
241-C-106	239Pu	0.43 Ci	6.133E-02	239.0	—	—	0.0			
241-C-106	240Pu	0.09 Ci	2.266E-01	240.0	—	—	0.0			
241-C-106	241Pu	1.06 Ci	1.031E+02	241.0	—	—	0.0			
241-C-106	242Pu	0.00 Ci	3.931E-03	242.0	—	—	0.0			
241-C-106	Al	216.02 kg		27.0	0.2330	910	216.0	0.00002	28564.05	31.39
241-C-106	Bi	0.83 kg		209.0	0.0340	—	0.8	0.00000	83.26	
241-C-106	Ca	3.56 kg		40.1	0.4300	770	3.6	0.00000	470.50	0.61
241-C-106	Cl	26.70 kg		35.5	33.6000	—	26.7	0.00022	3529.67	
241-C-106	Cr	4.36 kg		52.0	3.1000	135	4.4	0.00000	575.93	4.27
241-C-106	F	13.49 kg		19.0	0.0096	—	13.5	0.00000	1783.51	
241-C-106	Fe	1.68 kg		55.8	2.5600	160	1.7	0.00000	222.24	1.39
241-C-106	Hg	0.06 kg		200.6	374.0000	5	0.1	0.00000	8.16	1.63
241-C-106	K	81.84 kg		39.1	2.1000	—	81.8	0.00004	10821.83	—
241-C-106	La	0.00 kg		138.9	9.0000	—	0.0	0.00000	0.00	—
241-C-106	Mn	0.32 kg		54.9	13.3000	32	0.3	0.00000	42.50	1.33
241-C-106	Na	8957.13 kg		23.0	0.5300	360	8957.1	0.00178	1184363.06	3289.90
241-C-106	Ni	0.59 kg		58.7	4.5000	105	0.6	0.00000	77.56	0.74
241-C-106	NO2	1361.13 kg		46.0	1.9100	270	1361.1	0.00049	179978.74	666.58
241-C-106	NO3	61.29 kg		62.0	1.9100	364	61.3	0.00002	8104.76	22.27
241-C-106	OH	25.84 kg		17.0	0.3330	—	25.8	—	—	—
241-C-106	Pb	2.93 kg		207.2	0.1710	—	2.9	0.00000	387.82	—
241-C-106	PO4	530.15 kg		95.0	0.1810	3960	530.1	0.00001	70098.16	17.61
241-C-106	Si	24.76 kg		28.1	0.1710	1400	24.8	0.00000	3274.42	2.34
241-C-106	SO4	396.76 kg		96.1	0.5200	—	396.7	0.00002	52480.32	—
241-C-106	Sr	0.01 kg		87.6	1.2000	—	0.0	0.00000	1.04	—
241-C-106	TIC as CO3	9396.21 kg		60.0	0.0040	140000	9396.2	0.00001	1242420.06	8.87
241-C-106	TOC	337.21 kg		12.0	0.0035	26000	337.2	0.00000	44587.79	1.69
241-C-106	UTOTAL	85.36 kg		238.0	7.5700	770	85.4	0.00002	11286.28	14.66
241-C-106	Zr	0.00 kg		91.2	0.1840	4000	0.0	0.00000	0.14	0.00

241-C-106	Fissile Mass, kg	0.008	All Component Mass Sum (kg) =	21613.1	Sum X/Pu =	2843141.01	Total Fraction =	4065.17	
241-C-106	Pu Mass, kg =	0.007	CWM Components Mass (kg) =	11127.4	CWM X/Pu =	1474511.54	Insoluble Fract=	57.74	
241-C-106	235U Mass, kg =	0.550	Density =	1.13 g/mL	Macro Absorption XS =	0.00261	Soluble Fract =	4007.43	
241-C-106	233U Mass, kg=	0.000	Volume =	70.0 kL	Water Macro Absorp. XS=	0.01825			
241-C-106	235U Enrich. =	0.644	Component Mass/Volume (g/cc) =	0.309	Solids+ Water Macro XS =	0.02086			
241-C-106	Excess 235U, kg =	0.000	Water (g/cc) = (Density - M/V) =	0.821					
241-C-106	240Pu, kg =	0.000							

Table B5. Parameters for Sludge Layer in SST C-106

Download Date: April 1, 2003

Sludge Tank Name	Analyte	Inventory	Sludge Ci/g (LA-12846)	Atom Wt.	Micro XS Absorption	Mass Ratio X/Pu (Limit)	Mass(kg)	Macroscopic Absorb XS	Mass Ratio X/Pu (Actual)	Actual/Min Mass Fraction
241-C-106	233U	0.00 Ci	9.642E-03	233.0	—	—	0.0			
241-C-106	235U	0.00 Ci	2.163E-06	235.0	—	—	0.4			
241-C-106	238U	0.02 Ci	3.364E-07	238.0	—	—	60.3			
241-C-106	238Pu	6.16 Ci	1.713E+01	238.0	—	—	0.0			
241-C-106	239Pu	111.13 Ci	6.133E-02	239.0	—	—	1.8			
241-C-106	240Pu	22.70 Ci	2.268E-01	240.0	—	—	0.1			
241-C-106	241Pu	273.01 Ci	1.031E+02	241.0	—	—	0.0			
241-C-106	242Pu	0.00 Ci	3.931E-03	242.0	—	—	0.0			
241-C-106	Al	2706.39 kg		27.0	0.2330	910	2706.4	0.00041	1412.66	1.66
241-C-106	Bi	0.16 kg		209.0	0.0340	—	0.2	0.00000	0.09	
241-C-106	Ca	116.02 kg		40.1	0.4300	770	116.0	0.00002	60.56	0.08
241-C-106	Cl	5.58 kg		35.5	33.5000	—	5.6	0.00009	2.91	
241-C-106	Cr	84.45 kg		52.0	3.1000	136	84.4	0.00009	44.06	0.33
241-C-106	F	3.52 kg		19.0	0.0096	—	3.5	0.00000	1.84	
241-C-106	Fe	4514.01 kg		55.8	2.5600	160	4514.0	0.00367	2356.16	14.73
241-C-106	Hg	6.50 kg		200.6	374.0000	5	6.5	0.00021	3.39	0.68
241-C-106	K	31.16 kg		39.1	2.1000	—	31.2	0.00003	16.26	—
241-C-106	La	18.21 kg		138.9	9.0000	—	18.2	0.00002	9.51	—
241-C-106	Mn	1121.74 kg		54.9	13.3000	32	1121.7	0.00481	585.51	18.30
241-C-106	Na	3628.64 kg		23.0	0.5300	360	3628.6	0.00148	1894.06	5.26
241-C-106	Ni	156.36 kg		58.7	4.5000	105	156.4	0.00021	81.61	0.78
241-C-106	NO2	119.46 kg		46.0	1.9100	270	119.5	0.00009	62.35	0.23
241-C-106	NO3	17.38 kg		62.0	1.9100	364	17.4	0.00001	9.07	0.02
241-C-106	OH	kg		17.0	0.3330	—	—	—	—	—
241-C-106	Pb	232.58 kg		207.2	0.1710	—	232.6	0.00000	121.40	—
241-C-106	PO4	553.88 kg		95.0	0.1810	3960	553.9	0.00002	289.11	0.07
241-C-106	Si	61.35 kg		28.1	0.1710	1400	61.3	0.00001	32.02	0.02
241-C-106	SO4	89.64 kg		96.1	0.5200	—	89.6	0.00001	46.79	—
241-C-106	Sr	9.59 kg		87.6	1.2000	—	9.6	0.00000	5.01	—
241-C-106	TIC as CO3	6304.96 kg		60.0	0.0040	140000	6306.0	0.00001	3291.01	0.02
241-C-106	TOC	100.26 kg		12.0	0.0035	28000	100.3	0.00000	52.33	0.00
241-C-106	UTOTAL	60.76 kg		238.0	7.5700	770	60.8	0.00003	31.71	0.04
241-C-106	Zr	3.92 kg		91.2	0.1840	4000	3.9	0.00000	2.05	0.00

241-C-106	Fissile Mass, kg	1.916	All Component Mass Sum (kg) =	20009.1	Sum X/Pu =	10411.51	Total Fraction =		42.12
241-C-106	Pu Mass, kg =	1.916	CWM Components Mass (kg) =	11539.8	CWM X/Pu =	6046.06	Insoluble Fract=		36.42
241-C-106	235U Mass, kg =	0.391	Density =	1.52 g/mL	Macro Absorption XS =	0.01124	Soluble Fract =		5.69
241-C-106	233U Mass, kg=	0.000	Volume =	34.0 kL	Water Macro Absorp. XS=	0.02070			
241-C-106	235U Enrich. =	0.644	Component Mass/Volume (g/cc) =	0.589	Solids+ Water Macro XS =	0.03193			
241-C-106	Excess 235U, kg =	0.000	Water (g/cc) = (Density - M/V) =	0.931					
241-C-106	240Pu, kg =	0.100							

Table B6. Parameters for Supernatant Layer in DST AN-106

Download Date: April 1, 2003

Supernate Tank Name	Analyte	Inventory	Supernate Ci/g (LA-12846)	Atom Wt.	Micro XS Absorption	Mass Ratio X/Pu (Limit)	Mass(kg)	Macroscopic Absorb XS	Mass Ratio X/Pu (Actual)	Actual/Min Mass Fraction
241-AN-106	233U	0.216712 Ci	9.642E-03	233.0	—	—	0.0			
241-AN-106	235U	0.0262474 Ci	2.163E-06	235.0	—	—	12.1			
241-AN-106	238Pu	0.62594 Ci	1.713E+01	238.0	—	—	0.0			
241-AN-106	236U	0.162008 Ci	3.364E-07	236.0	—	—	481.6			
241-AN-106	239Pu	10.257 Ci	6.133E-02	239.0	—	—	0.2			
241-AN-106	240Pu	1.578 Ci	2.268E-01	240.0	—	—	0.0			
241-AN-106	241Pu	8.8368 Ci	1.031E+02	241.0	—	—	0.0			
241-AN-106	242Pu	5.0075E-05 Ci	3.931E-03	242.0	—	—	0.0			
241-AN-106	Al	312.444 kg		27.0	0.2330	910	312.4	0.00000	1587.53	1.74
241-AN-106	Bi	1.32026 kg		209.0	0.0340	—	1.3	0.00000	6.71	
241-AN-106	Ca	2.08718 kg		40.1	0.4300	770	2.1	0.00000	10.60	0.01
241-AN-106	Cl	319.282 kg		35.5	33.5000	—	319.3	0.00034	1622.27	
241-AN-106	Cr	60.49 kg		52.0	3.1000	135	60.5	0.00000	307.35	2.28
241-AN-106	F	1099.34 kg		19.0	0.0096	—	1099.3	0.00000	5586.76	
241-AN-106	Fe	1.86204 kg		55.8	2.5600	160	1.9	0.00000	9.46	0.06
241-AN-106	Hg	0 kg		200.6	374.0000	6	0.0	0.00000	0.00	0.00
241-AN-106	K	1236.1 kg		39.1	2.1000	—	1236.1	0.00005	6280.84	—
241-AN-106	La	0.0467086 kg		138.9	9.0000	—	0.0	0.00000	0.24	—
241-AN-106	Mn	0.157274 kg		54.9	13.3000	32	0.2	0.00000	0.80	0.02
241-AN-106	Na	28614.4 kg		23.0	0.5300	360	28614.4	0.00076	145390.04	403.86
241-AN-106	Ni	26.1422 kg		58.7	4.6000	105	26.1	0.00000	132.83	1.27
241-AN-106	NO2	9468 kg		46.0	1.9100	270	9468.0	0.00045	48107.00	178.17
241-AN-106	NO3	6206.8 kg		62.0	1.9100	364	6206.8	0.00022	31536.81	86.64
241-AN-106	OH	3597.84 kg		17.0	0.3330	—	3597.8	—	—	—
241-AN-106	Pb	1.57274 kg		207.2	0.1710	—	1.6	0.00000	7.99	—
241-AN-106	PO4	2224.96 kg		95.0	0.1810	3960	2225.0	0.00000	11305.14	2.84
241-AN-106	Si	12.4662 kg		28.1	0.1710	1400	12.5	0.00000	63.34	0.05
241-AN-106	SO4	1846.26 kg		96.1	0.5200	—	1846.3	0.00001	9380.66	—
241-AN-106	Sr	0.0417118 kg		87.6	1.2000	—	0.0	0.00000	0.21	—
241-AN-106	TIC as CO3	18357.4 kg		60.0	0.0040	140000	18357.4	0.00000	93274.12	0.67
241-AN-106	TOC	2896.26 kg		12.0	0.0035	26000	2896.3	0.00000	14726.09	0.53
241-AN-106	UTOTAL	1863.06 kg		238.0	7.5700	770	1863.1	0.00007	9567.96	12.43
241-AN-106	Zr	196.302 kg		91.2	0.1840	4000	196.3	0.00000	1007.57	0.25

241-AN-106	Fissile Mass, kg	0.197	All Component Mass Sum (kg) =	78862.6	Sum X/Pu =	379911.22	Total Fraction =	690.81
241-AN-106	Pu Mass, kg =	0.174	CWM Components Mass (kg) =	46828.5	CWM X/Pu =	215967.12	Insoluble Fract=	18.09
241-AN-106	236U Mass, kg =	12.136	Density = 1.12 g/mL	Macro Absorption XS =	0.00194	Soluble Fract =	672.72		
241-AN-106	233U Mass, kg=	0.022	Volume = 526.0 kL	Water Macro Absorp. XS=	0.02155				
241-AN-106	235U Enrich. =	0.644	Component Mass/Volume (g/cc) =	0.150	Solids+ Water Macro XS =	0.02350			
241-AN-106	Excess 235U, kg =	0.000	Water (g/cc) = (Density - MV) =	0.970					
241-AN-106	240Pu, kg =	0.007							

Table B7. Parameters for Saltcake Layer in DST AN-106

Download Date: April 1, 2003

Saltcake Tank Name	Analyte	Inventory	Saltcake Ci/g (LA-12846)	Atom Wt.	Micro XS Absorption	Mass Ratio X/Pu (Limit)	Mass(kg)	Macroscopic Absorb XS	Mass Ratio X/Pu (Actual)	Actual/Min Mass Fraction
241-AN-106	233U	0.08673006 Ci	9.842E-03	233.0	—	—	0.0			
241-AN-106	235U	0.00124267 Ci	2.163E-06	235.0	—	—	0.6			
241-AN-106	238U	0.0272155 Ci	3.364E-07	238.0	—	—	80.9			
241-AN-106	238Pu	2.09825386 Ci	1.713E+01	238.0	—	—	0.0			
241-AN-106	239Pu	61.8129341 Ci	6.133E-02	239.0	—	—	1.0			
241-AN-106	240Pu	10.4676272 Ci	2.268E-01	240.0	—	—	0.0			
241-AN-106	241Pu	107.777961 Ci	1.031E+02	241.0	—	—	0.0			
241-AN-106	242Pu	0.00094586 Ci	3.931E-03	242.0	—	—	0.0			
241-AN-106	Al	2476.07 kg		27.0	0.2330	910	2476.1	0.00020	2324.80	2.56
241-AN-106	Bi	17.6644 kg		209.0	0.0340	—	17.7	0.00000	16.59	
241-AN-106	Ca	44.5718 kg		40.1	0.4300	770	44.6	0.00000	41.87	0.06
241-AN-106	Cl	595.66 kg		35.5	33.5000	—	595.7	0.00021	559.60	
241-AN-106	Cr	462.15 kg		52.0	3.1000	135	462.2	0.00026	434.09	3.22
241-AN-106	F	71.1711 kg		19.0	0.0096	—	71.2	0.00000	66.85	
241-AN-106	Fe	32.9667 kg		55.8	2.5600	160	33.0	0.00001	30.97	0.19
241-AN-106	Hg	0 kg		200.6	374.0000	5	0.0	0.00000	0.00	0.00
241-AN-106	K	349.18 kg		39.1	2.1000	—	349.2	0.00017	327.96	—
241-AN-106	La	6.56253 kg		138.9	9.0000	—	6.6	0.00000	6.16	—
241-AN-106	Mn	5.48418 kg		54.9	13.3000	32	5.5	0.00001	5.15	0.16
241-AN-106	Na	17972.5 kg		23.0	0.5300	360	17972.5	0.00384	16881.32	46.89
241-AN-106	Ni	26.2912 kg		58.7	4.5000	105	26.3	0.00002	24.69	0.24
241-AN-106	NO2	8760.31 kg		46.0	1.9100	270	8760.3	0.00337	8228.44	30.48
241-AN-106	NO3	10966.9 kg		62.0	1.9100	364	10966.9	0.00314	10321.72	28.36
241-AN-106	OH	kg		17.0	0.3330	—	—	—	—	—
241-AN-106	Pb	9.16136 kg		207.2	0.1710	—	9.2	0.00000	8.62	—
241-AN-106	PO4	1671.31 kg		95.0	0.1610	3960	1671.3	0.00003	1475.91	0.37
241-AN-106	Si	43.9566 kg		28.1	0.1710	1400	44.0	0.00000	41.29	0.03
241-AN-106	SO4	1530.23 kg		96.1	0.5200	—	1530.2	0.00008	1437.32	—
241-AN-106	Sr	2.58804 kg		87.6	1.2000	—	2.6	0.00000	2.43	—
241-AN-106	TIC as CO3	8483.02 kg		60.0	0.0040	140000	8483.0	0.00001	7967.98	0.06
241-AN-106	TOC	847.275 kg		12.0	0.0035	28000	847.3	0.00000	795.83	0.03
241-AN-106	UTOTAL	81.5436 kg		238.0	7.5700	770	81.5	0.00002	76.59	0.10
241-AN-106	Zr	8.50356 kg		91.2	0.1840	4000	8.5	0.00000	7.99	0.00

241-AN-106	Fissile Mass, kg	1.065	All Component Mass Sum (kg) =	54468.6	Sum X/Pu =	51064.10	Total Fraction =		112.73
241-AN-106	Pu Mass, kg =	1.065	CWM Components Mass (kg) =	41801.1	CWM X/Pu =	29018.02	Insoluble Fract=		6.49
241-AN-106	235U Mass, kg =	0.575	Density = 1.58 g/mL		Macro Absorption XS =	0.01637	Soluble Fract =		106.24
241-AN-106	233U Mass, kg=	0.009	Volume = 65.0 kL		Water Macro Absorp. XS=	0.01649			
241-AN-106	235U Enrich. =	0.706	Component Mass/Volume (g/cc) =	0.838	Solids+ Water Macro XS =	0.03286			
241-AN-106	Excess 235U, kg =	0.000	Water (g/cc) = (Density - MV) =	0.742					
241-AN-106	240Pu, kg =	0.046							

Table B8. Parameters for Supernatant Layer in DST AN-101

Download Date: April 1, 2003
Supernatant Liquid

Tank Name	Analyte	Inventory	Ci/g (LA-12846)	Atom Wt.	Micro XS Absorption	Mass Ratio X/Pu (Limit)	Mass(kg)	Macroscopic Absorb XS	Mass Ratio X/Pu (Actual)	Actual/Min Mass Fraction
241-AN-101	233U	0.0114 Ci	9.642E-03	233.0	—	—	0.0			
241-AN-101	235U	0.0002 Ci	2.163E-06	235.0	—	—	0.1			
241-AN-101	238U	0.0038 Ci	3.364E-07	238.0	—	—	11.3			
241-AN-101	238Pu	0.0117 Ci	1.713E+01	238.0	—	—	0.0			
241-AN-101	239Pu	0.4301 Ci	6.133E-02	239.0	—	—	0.0			
241-AN-101	240Pu	0.0731 Ci	2.266E-01	240.0	—	—	0.0			
241-AN-101	241Pu	0.6142 Ci	1.031E+02	241.0	—	—	0.0			
241-AN-101	242Pu	0.0000 Ci	3.931E-03	242.0	—	—	0.0			
241-AN-101	Al	15396.86 kg		27.0	0.2330	910	15396.9	0.00008	1806514.11	1986.18
241-AN-101	Bi	19.12 kg		209.0	0.0340	—	19.1	0.00000	2243.36	
241-AN-101	Ca	19.12 kg		40.1	0.4300	770	19.1	0.00000	2243.36	2.91
241-AN-101	Cl	4794.37 kg		36.5	33.8000	—	4794.4	0.00285	562624.08	
241-AN-101	Cr	124.43 kg		52.0	3.1000	135	124.4	0.00000	14598.73	108.15
241-AN-101	F	200.78 kg		19.0	0.0096	—	200.8	0.00000	23557.96	
241-AN-101	Fe	9.56 kg		55.8	2.5500	160	9.6	0.00000	1121.68	7.01
241-AN-101	Hg	0.00 kg		200.6	374.0000	5	0.0	0.00000	0.00	0.00
241-AN-101	K	3313.26 kg		39.1	2.1000	—	3313.3	0.00011	388744.61	—
241-AN-101	La	9.56 kg		138.9	9.0000	—	9.6	0.00000	1121.68	—
241-AN-101	Mn	1.91 kg		54.9	13.3000	32	1.9	0.00000	224.34	7.01
241-AN-101	Na	132888.78 kg		23.0	0.5300	360	132888.8	0.00193	15591847.15	43310.69
241-AN-101	Ni	3.82 kg		58.7	4.6000	106	3.8	0.00000	448.67	4.27
241-AN-101	NO2	92261.16 kg		46.0	1.9100	270	92261.2	0.00241	10825005.97	40062.61
241-AN-101	NO3	111500.89 kg		62.0	1.9100	364	111500.9	0.00218	13082404.38	35940.67
241-AN-101	OH	25907.60 kg		17.0	0.3330	—	—	—	—	—
241-AN-101	Pb	42.56 kg		207.2	0.1710	—	42.5	0.00000	4992.02	—
241-AN-101	PO4	2236.82 kg		95.0	0.1810	3980	2236.8	0.00000	262448.80	65.94
241-AN-101	Si	23.24 kg		28.1	0.1710	1400	23.2	0.00000	2726.46	1.95
241-AN-101	SO4	1700.95 kg		96.1	0.8200	—	1700.9	0.00001	199661.17	—
241-AN-101	Sr	1.91 kg		87.6	1.2000	—	1.9	0.00000	224.34	—
241-AN-101	TIC as CO3	13001.60 kg		60.0	0.0040	140000	13001.6	0.00000	1526478.37	10.90
241-AN-101	TOC	1367.06 kg		12.0	0.0035	28000	1367.1	0.00000	160599.86	5.73
241-AN-101	UTOTAL	11.38 kg		238.0	7.5700	770	11.4	0.00000	1334.79	1.73
241-AN-101	Zr	1.91 kg		91.2	0.1640	4000	1.9	0.00000	224.34	0.06

241-AN-101	Fissile Mass, kg	0.009	All Component Mass Sum (kg) =	378942.3	Sum X/Pu =	44459968.69	Total Fraction =	121644.81	
241-AN-101	Pu Mass, kg =	0.007	CWM Components Mass (kg) =	354294.1	CWM X/Pu =	28488270.29	Insoluble Fract=	2116.36	
241-AN-101	235U Mass, kg =	0.079	Density =	1.29 g/mL	Macro Absorption XS =	0.00957	Soluble Fract =	119429.46	
241-AN-101	233U Mass, kg=	0.001	Volume =	966.0 kL	Water Macro Absorp. XS=	0.01985			
241-AN-101	235U Enrich. =	0.696	Component Mass/Volume (g/cc) =	0.396	Solids+ Water Macro XS =	0.02942			
241-AN-101	Excess 235U, kg =	0.000	Water (g/cc) = (Density - M/V) =	0.894					
241-AN-101	240Pu, kg =	0.000							

Table B9. Summary of BBI Waste Layers in Tanks C-106, AN-106, and AN-101

Tank	C-106			AN-101	AN-106			AN-106 + AN-101		
Waste Type	Supernate	Sludge	Total	Supernate	Supernate	Saltcake	Total	Supernate	Saltcake	Total
Volume (kL)	70	34	104	956	526	65	591	1482	65	1547
Fissile Mass (kg)	0.007	1.916	1.923	0.009	0.197	1.065	1.262	0.205	1.065	1.270
Pu Mass (kg)	0.007	1.916	1.923	0.007	0.174	1.055	1.229	0.182	1.055	1.237
^{235}U Mass (kg)	0.550	0.391	0.941	0.079	12.135	0.575	12.71	12.214	0.575	12.789
^{233}U Mass (kg)	0.000	0.000	0.000	0.001	0.022	0.009	0.031	0.024	0.009	0.033
^{235}U Enrich. (wt%)	0.644	0.644	0.644	0.695	0.644	0.705	0.651	0.645	0.705	0.648
Excess 235U Mass (kg)	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
^{240}Pu Mass (kg)	0.000	0.100	0.100	0.000	0.007	0.046	0.053	0.007	0.046	0.053
Components Mass Sum (kg)	21613	20009	41622	378942	78863	54469	133332	483736	54469	538205
Mass/Volume (g/cc)	0.309	0.589	0.400	0.396	0.150	0.838	0.226	0.326	0.838	0.348
Mass Unaccounted For (g/cc)	0.821	0.931	0.860	0.894	0.970	0.742	0.944	0.904	0.742	0.892
Density (g/cc)	1.13	1.52	1.26	1.29	1.12	1.58	1.17	1.23	1.58	1.24
Total Absorber-to-Plutonium Subcritical Mass Fractions										
Sum X/Pu	----	10411	21550	----	379911	51084	102414.	2209582	51084	399503.
Total Subcritical Fraction	4065.2	42.1	57.9	121545	690.8	112.7	202.9	5707.2	112.7	1015.7
Insoluble Subcritical Fraction	57.7	36.4	36.5	2115	18.1	6.5	8.3	105.1	6.5	22.4
Soluble Subcritical Fraction	4007.4	5.7	21.4	119429	672.7	106.2	194.6	5602.1	106.2	993.3
Insoluble Component Absorber-to-Plutonium Subcritical Mass Fractions										
Aluminum	31.4	1.6	1.7	1985.2	1.7	2.6	2.4	84.1	2.6	15.7
Chromium	4.3	0.3	0.3	108.1	2.3	3.2	3.1	6.7	3.2	3.8
Iron	1.4	14.7	14.7	7.0	0.1	0.2	0.2	0.3	0.2	0.2
Manganese	1.3	18.3	18.2	7.0	0.0	0.2	0.1	0.3	0.2	0.2
Nickel	0.7	0.8	0.8	4.3	1.3	0.2	0.4	1.4	0.2	0.4
Silicon	2.3	0.0	0.0	1.9	0.1	0.0	0.0	0.1	0.0	0.0
Zirconium	0.0	0.0	0.0	0.1	0.3	0.0	0.0	0.2	0.0	0.0
Uranium-238	14.7	0.04	0.1	1.7	12.4	0.1	2.0	12.0	0.1	2.0

B5.0 REFERENCES

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APPENDIX C

ANALYSIS OF PHASE II SLUDGE DISSOLUTION RESIDUES

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ANALYSIS OF PHASE II SLUDGE DISSOLUTION RESIDUES

The material presented in this appendix was received from D.L. Herting on April 11, 2003, in an email titled *Preliminary Residue Results*, as follows:

> Here (attached) are the preliminary results of the C-106/AY-102 Phase I residue analyses. I think they're quite interesting, and generally support what we concluded in the Phase I reports.
>
> The file is not in memo format, and has not been reviewed, so I have severely limited the distribution. The information will be included in the Phase II report, which we will issue in early May. If you need to have this info is a form (i.e., an official memo), which you can distribute before that time, please let me know.
>
> Supposedly, the ICP-MS analyses for Pu etc., have been completed, but I haven't seen those results yet. I suspect your criticality people (including Hans Toffer, who called yesterday) will want to see them along with the stuff I have here.
>
> Finally, please let me know if you think we should be analyzing any of the residues from the Phase II tests. Time is not on our side. The Test Plan says: "Sludge and/or residue samples from Phase II may be selected for analysis if any of the Phase II results are significantly different than the Phase I results. At a minimum the residue from test F1 will be analyzed."

In Tables C1 through C9 the entry AT refers to "total alpha." For conservatism all of the alpha count is assumed to be coming from ^{239}Pu. Clow et al. (1994) gives the specific activity for ^{239}Pu as 0.06133 Ci/g.

For Tables C1 and C2 the units for ^{137}Cs, ^{90}Sr, ^{99}Tc, and AT entries (last four rows) are μCi/g. All other entries are in mg/g. For Table C3 through C8 the units for ^{137}Cs, ^{90}Sr, ^{99}Tc, and AT entries (last four rows) are μCi. All other entries are in mg.

Reference:

Clow, J., R. DeVore, J. Elder, G. Heindel, W. Inkret, and G. Miller, 1994, *Specific Activities and DOE-STD-1027-92 Hazard Category 2 Thresholds - LANL Fact Sheet*, LA-12846-MS, Los Alamos National Laboratory, Los Alamos, New Mexico.

C-106 and AY-102 Sludge Dissolution Residues were analyzed for Phase I samples A1, 2, 4, 5, and B1, 3, 4, 7, 8. Here is an overview of the results, which will be published in the Phase II report next month. This information is preliminary, and has not been reviewed or released.

Table C1. Residues after Contact with Water Only (mg/g or µCi/g)

Tank	C-106		AY-102	
Sample	A1	A2	B1	B3
TIC	19.5	18.0	14.0	12.9
TOC	< 1.0	< 1.0	1.3	2.2
Al	35.2	39.0	69.8	58.1
Ca	2.2	3.0	3.5	3.2
Cr	1.8	2.4	2.6	2.3
Fe	62.6	66.5	120.0	109.0
Mg	< 1	3.7	0.9	< 0.9
Mn	47.7	83.0	33.3	29.5
Na	70.4	108.0	50.5	43.3
P	19.6	37.8	3.9	2.5
Pb	4.5	4.2	6.6	6.2
Si	6.9	7.1	20.1	17.5
Sr	0.17	0.14	0.28	0.26
Zr	< 0.10	0.12	1.30	0.91
^{137}Cs	151	179	337	291
^{90}Sr	543	436	8670	7800
^{99}Tc	0.032	0.012	0.035	0.030
AT	4.7	8.1	7.7	7.2

Notes:

In general, the agreement between samples from the same tank is fairly good, with the exception of a few of the C-106 analytes, especially Mg, Mn, P, and AT.

AY-102 is roughly a factor of two higher than C-106 in Al, Fe, Si, Sr (ICP), and ^{137}Cs.

C-106 is ~1.5 to 2 times higher in Mn and Na, and ~10 times higher in P.

AY-102 is more than an order of magnitude higher in ^{90}Sr, and ~10 times higher in Zr.

From the beginning, sample homogeneity has been a major concern. The C-106 sample, in particular, was clearly a mixture of black and white materials, with some of the white chunks and streaks being relatively large. (The AY-102 sample was more evenly black.) One of the residue samples (A4) was analyzed in duplicate, fulfilling the standard QA requirement for one duplicate per batch. The results are not in good agreement, suggesting poor homogeneity.

Table C2. Agreement between Duplicate Analyses for Sample A4 (mg/g or µCi/g) Sample A4 held C-106 sludge after treatment with 15 mL of 1M oxalic acid

Sample	A4 Original	A4 Duplicate	Orig/Dup Ratio
TIC	20.2	na	--
TOC	33.7	na	--
Al	146	52.6	2.8
Ca	1.1	3.7	0.3
Cr	0.6	2.0	0.3
Fe	21.2	63.2	0.3
Mg	<0.8	2.1	~0.3
Mn	10.7	68.4	0.2
Na	135	112	1.2
P	3.3	44.2	0.07
Pb	1.4	4.3	0.3
Si	1.3	6.8	0.6
Sr	0.12	0.18	0.6
Zr	0.59	0.15	3.9
^{137}Cs	29	175	0.2
^{90}Sr	1220	5930	0.2
^{99}Tc	0.014	0.007	2.0
AT	1.7	7.9	0.2

Notes:

In the mass balance calculations that follow, the A4 Duplicate results are much more consistent with the starting material (average of A1 and A2) than the A4 Original results. Results for A4 Original appear to have been compromised by perhaps selecting a chunk of the white material – much higher in aluminum than the starting material, and much lower in Fe and Mn. Results for A4 Duplicate match the starting material fairly well (except for ^{90}Sr), suggesting little dissolution, in general agreement with the 21-24% dissolution by weight reported in Phase I final report (FH-0301381, March 27, 2003).

The following mass balance calculations show the total mg (or μCi) of each analyte recovered in each of the acid dissolution tests for which both residue analysis and supernate analysis are available.

The amount (mg or μCi) in the supernate is the analytical value reported in Phase I final report times the volume of liquid for that test (assumed to be 15 mL or 40 mL, despite the leakage).

The amount in the residue is the analytical value for the residue (in mg/g or μCi/g) times the final weight of centrifuged solids reported in the Phase I final report.

The "Total Found" is the sum of the amounts in the supernate and residue.

The "Initial Sample" is the average of A1 and A2 (or B1 and B3 for the B-series) from Table C1 above times the pre-acid-contact weight of washed centrifuged solids given in the Phase I final report.

"Total Found" and "Initial Sample" would be the same, if the samples were perfectly homogeneous and the analyses were error-free.

The "% Dissolved" is 100 times the amount in the supernate divided by the Total Found.

Table C3. Mass Balance for Sample A4 (Original Sample); Weights in mg (or µCi)

Sample A4 held C-106 sludge after treatment with 15 mL of 1M oxalic acid

	Supernate	Residue	Total Found	Initial Sample	% Dissolved
TIC	na	61.1	--	74.2	--
TOC	na	101.9	--	<4	--
Al	115	442	557	147	21
Ca	0.8	3.2	4.0	10.4	20
Cr	0.8	1.8	2.6	8.4	30
Fe	6.7	64.1	70.8	255	9
Mg	1.9	<2.4	--	7.9	--
Mn	9.8	32.4	42.2	259	23
Na	243	408	651	353	37
P	11.9	10.0	21.9	114	54
Pb	0.2	4.4	4.6	17.2	5
Si	1.6	3.8	5.4	27.8	29
Sr	0.10	0.35	0.45	0.60	22
Zr	0.7	1.8	2.5	0.5	27
^{137}Cs	104	88	192	653	54
^{90}Sr	2360	3690	6050	1940	39
^{99}Tc	0.014	0.044	0.058	0.087	24
AT	1.2	5.2	6.4	25.4	18

Notes:

Comparison of Found vs. Initial reflects the homogeneity problems shown previously (Table C2); The Found/Initial ratio is grossly high in Al, Na, Zr, and ^{90}Sr, and grossly low in Fe, Mn, P, Pb, Si, ^{137}Cs, and AT.

Table C4. Mass Balance for Sample A4 (Duplicate Sample); Weights in mg (or µCi)

Sample A4 held C-106 sludge after treatment with 15 mL of 1M oxalic acid

	Supernate	Residue	Total Found	Initial Sample	% Dissolved
TIC	na	61.1	--	74.2	--
TOC	na	102	--	<4	--
Al	115	159	274	147	42
Ca	0.8	11.2	12.0	10.4	7
Cr	0.8	6.2	7.0	8.4	11
Fe	6.7	191	198	255	3
Mg	1.9	6.3	8.2	7.9	23
Mn	9.8	207	217	259	5
Na	243	339	582	353	42
P	11.9	134	146	114	8
Pb	0.2	13.0	13.2	17.2	2
Si	1.6	20.7	22.3	27.8	7
Sr	0.10	0.54	0.64	0.60	15
Zr	0.7	0.5	1.2	0.5	59
^{137}Cs	104	529	633	653	16
^{90}Sr	2360	17930	20290	1940	12
^{99}Tc	0.014	0.022	0.036	0.087	39
AT	1.2	24.0	25.2	25.4	5

Notes:

Still high but much closer than A4 Original in Al, Na, and Zr. Much closer in Fe, Mn, P, Pb, Si, ^{137}Cs, and AT. Even higher than A4 Original in ^{90}Sr; now found more than 10 times higher than Initial. Suggests that ^{90}Sr is contained in highly-concentrated but dispersed particles, producing high variation from sample to sample.

Table C5. Mass Balance for Sample A5; Weights in mg (or µCi)

Sample A5 held C-106 sludge after treatment with 40 mL of 1M oxalic acid

	Supernate	Residue	Total Found	Initial Sample	% Dissolved
TIC	na	0.45	--	75.1	--
TOC	na	48.1	--	<4	--
Al	266	26	292	149	91
Ca	5.0	<2.1	<7.1	10.5	>70
Cr	3.4	1.1	4.5	8.5	74
Fe	109	111	220	258	50
Mg	3.0	<2.1	<5.1	8.0	>59
Mn	107	50	157	262	68
Na	420	14	434	357	97
P	74.8	<4.1	<79	115	>95
Pb	4.0	11.9	15.9	17.4	25
Si	20.1	<1.0	<21.1	28.1	>95
Sr	0.47	<0.21	<0.68	0.61	>69
Zr	6.5	0.7	7.2	0.5	91
^{137}Cs	396	<26	<422	661	>94
^{90}Sr	16040	1650	17690	1960	91
^{99}Tc	0.040	0.018	0.058	0.088	69
AT	<3.0	10.9	<13.9	25.7	<22

Notes:

58-74% dissolution by weight reported in Phase I final report, compares well with the component %Dissolved numbers here. Note that TOC (i.e., probably oxalate/binoxalate) is one of the major contributors to the residue. Also note that Na and Al appear closely tied in both samples A4 and A5, suggesting the presence of dawsonite, $NaAlCO_3(OH)_2$, which has been confirmed by XRD patterns of several of the residue samples.

Table C6. Mass Balance for Sample B4; Weights in mg (or μCi)

Sample B4 held AY-102 sludge after treatment with 15 mL of 1M oxalic acid

	Supernate	Residue	Total Found	Initial Sample	% Dissolved
TIC	na	23.8	--	51.9	--
TOC	na	72.9	--	6.9	--
Al	121	156	277	247	44
Ca	7.1	4.8	11.9	12.9	59
Cr	2.2	7.4	9.6	9.4	23
Fe	45	393	438	442	10
Mg	1.6	< 4.7	--	3.5	--
Mn	64	65	129	121	50
Na	164	86	250	180	65
P	7.7	9.5	17.2	12.3	45
Pb	< 1.5	24.1	< 25.6	24.6	< 6
Si	1.2	51.1	52.3	72.5	2
Sr	0.64	0.48	1.12	1.04	57
Zr	0.6	5.4	6.0	4.3	10
^{137}Cs	615	632	1247	1211	49
^{90}Sr	20100	14030	34130	31750	59
^{99}Tc	0.030	0.098	0.128	0.125	23
AT	2.0	28.7	30.6	28.7	6

Notes:

Much better homogeneity (agreement between Found and Initial) is displayed for all of the AY-102 samples.

Table C7. Mass Balance for Sample B7; Weights in mg (or µCi)

Sample B7 held AY-102 sludge after treatment with 15 mL of 0.5M oxalic/1.0M nitric acid

	Supernate	Residue	Total Found	Initial Sample	% Dissolved
TIC	na	18.5	--	54.4	--
TOC	na	35.7	--	7.3	--
Al	127	140	267	258	48
Ca	10.0	4.4	14.4	13.5	69
Cr	2.2	7.8	10.0	9.9	22
Fe	21	433	454	463	5
Mg	< 3.2	< 4.3	--	3.6	--
Mn	63	68	131	127	48
Na	181	74	255	190	71
P	< 6.3	12.0	< 18.3	12.9	< 35
Pb	< 3.2	22.5	< 25.7	25.8	< 13
Si	< 1.7	74.1	< 75.8	76.0	< 2
Sr	0.80	< 0.43	< 1.23	1.08	> 65
Zr	< 0.3	6.5	< 6.8	4.5	< 5
^{137}Cs	810	630	1440	1270	56
^{90}Sr	24450	12020	36470	33290	67
^{99}Tc	0.021	0.103	0.124	0.131	17
AT	2.4	29.6	32.0	30.1	7

Notes:

Table C8. Mass Balance for Sample B8; Weights in mg (or µCi)

Sample B8 held AY-102 sludge after treatment with 40 mL of 0.5M oxalic/1.0M nitric acid

	Supernate	Residue	Total Found	Initial Sample	% Dissolved
TIC	na	na	--	52.0	--
TOC	na	na	--	6.9	--
Al	183	77	260	247	70
Ca	11.6	< 5.7	< 17.3	13.0	> 67
Cr	6.8	3.6	10.4	9.4	65
Fe	221	301	522	443	42
Mg	2.5	< 5.7	--	3.5	--
Mn	100	27	127	121	79
Na	219	15	234	181	94
P	15.1	< 11.2	--	12.3	--
Pb	13.5	15.2	28.7	24.7	47
Si	72.0	7.9	79.9	72.7	90
Sr	0.96	< 0.56	< 1.52	1.04	> 63
Zr	5.2	2.6	7.8	4.3	67
^{137}Cs	1240	88	1328	1215	93
^{90}Sr	28000	3310	31310	31850	89
^{99}Tc	0.080	0.070	0.150	0.126	53
AT	4.0	36.5	40.5	28.8	10

Notes:

Table C9. Summary of Percent Dissolved Results

Sample	A4 (Orig)	A4 (Dup)	A5	B4	B7	B8
Volume (mL)	15	15	40	15	15	40
Al	21	42	91	44	48	70
Ca	20	7	> 70	59	69	> 67
Cr	30	11	74	23	22	65
Fe	9	3	50	10	5	42
Mg	--	23	> 59	--	--	--
Mn	23	5	68	50	48	79
Na	37	42	97	65	71	94
P	54	8	> 95	45	< 35	--
Pb	5	2	25	< 6	< 13	47
Si	29	7	> 95	2	< 2	90
Sr	22	15	> 69	57	> 65	> 63
Zr	27	59	91	10	< 5	67
^{137}Cs	54	16	> 94	49	56	93
^{90}Sr	39	12	91	59	67	89
^{99}Tc	24	39	69	23	17	53
AT	18	5	< 22	6	7	10
%DissSum[1]	21	21	58	--	22	53
%DissCSol[2]	24	24	74	--	-18	67

[1] [from Phase I preliminary report] %DissSum = percent sludge dissolved based on sum of components assumed to have dissolved to produce the concentrations found in the liquid phase.

[2] [from Phase I preliminary report] %DissCSol = percent sludge dissolved based on the final weight of centrifuged solids in comparison to the starting weight of wet centrifuged solids.

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