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Criticality Safety Evaluation of Disposing of K Basin Sludge in Double-shell Tank AW-105

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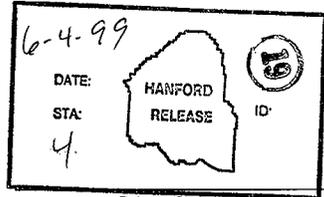
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Abstract: A criticality safety evaluation is made of the disposal of K Basin sludge in Double-shell Tank (DST) AW-105 located in the 200 East Area on the Hanford Site. The technical basis is provided for limits and controls to be used in the development of a Criticality Prevention Specification (CPS). A model of K Basin sludge is developed to account for fuel burnup. The iron/uranium mass ratio required to ensure an acceptable margin of subcriticality is determined.

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Criticality Safety Evaluation of Disposing of K Basin Sludge in Double-Shell Tank AW-105

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LIST OF TERMS

AST	AST Research, Inc.
CPS	Criticality Prevention Specification
CSER	Criticality Safety Evaluation Report
CWM	conservative waste model
DESH	DE&S Hanford, Inc.
DST	double-shell tank
ENDF	Evaluated Nuclear Data File
ERDF	Environmental Restoration Disposal Facility
Fe/(U+Pu)	mass ratio of iron to uranium plus plutonium
HLW	High-level waste
IRF	iron reduction factor
IWTS	Integrated Water Treatment System
k_{∞}	infinite neutron multiplication constant
KE	K East
KW	K West
LAW	Low-activity waste
MTU	Metric tons of uranium
NITAWL	code to process cross sections
ORIGEN	Oak Ridge Isotope Generation and decay code to calculate fuel composition changes during irradiation
PCF	plutonium concentration factor
PUREX	Plutonium Uranium Extraction Facility
SCALE	Standardized Computer Analysis for Licensing Evaluation
SCATS	Safeguards Control Accountability Transaction System
SPR	Single Pass Reactor
TEMPEST	Transient Energy Momentum and Pressure Equations in Three-dimensions
TWRS	Tank Waste Remediation Systems
USL	Upper Safety Limit
X/Pu	neutron absorber-to-plutonium mass ratio
XSDRNP	1-dimensional deterministic criticality code

1.0 INTRODUCTION

Fuel elements discharged from reactors on the Hanford Site have been stored under water in the 105-K East (KE) and 105-K West (KW) Basins, located in the 100 Area at the northern edge of the Hanford Site. These elements have been subjected to the stress of irradiation and reactor discharge operations. Subsequent cracking has allowed contact with water to oxidize uranium metal causing deterioration and, for some elements, partial disintegration. Oxidized uranium has sloughed off and broken pieces have fallen away from the elements. Uranium from deteriorated elements, mixed with debris on the basin floor, forms "K Basin Sludge." K Basin sludge will be removed from the basins, treated to meet Tank Farms acceptance criteria, and transferred to double-shell (DST) tank 241-AW-105.

This Criticality Safety Evaluation Report (CSER) reviews the criticality safety of storage of K Basin sludge at tank farms and provides the basis for proposed technical limits and controls, including a determination of the quantity of iron needed to ensure chemical compatibility and long-term subcriticality of the stored sludge. Removal of sludge from the basins, processing prior to receipt at tank farms, and transport to tank farms are not part of this evaluation.

K Basin sludge is characteristically different from the waste currently stored at tank farms. Plutonium generated when the uranium was irradiated in a reactor has not been removed, thereby resulting in a higher plutonium/uranium ratio than in existing tank waste. In its original form much of the K Basin sludge has a low ratio of neutron absorbing solids. Recommendations are made as to the quantity of neutron absorbers required. This report follows requirements outlined in HNF-PRO-539, *Criticality Safety Evaluations*, for the development of CSERs and guidance provided in *Guidelines for Preparing Criticality Safety Evaluations at Department of Energy Non-Reactor Nuclear Facilities* (DOE 1998).

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2.0 SUMMARY AND CONCLUSIONS

K Basin sludge is divided into three primary types - floor pit sludge, canister sludge, and fuel wash sludge. Spent N Reactor fuel stored in the KE and KW Basins has undergone deterioration leading to uranium corrosion products mixing with floor debris to form floor sludge. Canister sludge is composed of corrosion products from broken elements remaining inside of their storage canisters. The sludges are subdivided into five sludge streams according to storage location within the basin. Before receipt at tank farms, sludge streams will be processed to meet tank farms acceptance criteria, and iron will be added to provide neutron absorption.

This report evaluates criticality safety of discharge into and long-term storage in DST AW-105 located in the 200 East Area tank farms on the Hanford Site. Consideration is given to fuel burnup during irradiation (i.e., to the decrease in ^{235}U and the increase in ^{239}Pu and ^{240}Pu) to determine the quantity of iron required for an adequate margin of subcriticality. A model of K Basin sludge is developed to envelope the range of possible compositions, and this model is used to develop the bases for new criticality safety limits and controls to cover operations involving K Basin sludge at tank farms.

2.1 TANK FARM CRITICALITY PREVENTION SPECIFICATIONS

Limits and controls for criticality safety during the transfer and storage of sludge are governed by a Criticality Prevention Specification (CPS). The present CPS limit on the maximum permitted plutonium equivalent concentration is 1 g/L in settled solids (DESH 1997). This may be exceeded by K Basin sludge. This evaluation provides justification for revising the CPS limits and controls to permit storage of K Basin sludge at tank farms. Proposed limits are described in Section 3.0.

2.2 BASIS FOR CRITICALITY SAFETY EVALUATION

All green KE Basin uranium (i.e., before irradiation), and about half of green KW Basin uranium, was 0.947 wt% ^{235}U enriched. The remaining half of green KW Basin uranium was 1.25 wt% ^{235}U enriched. Because of having been irradiated, the ^{235}U enrichment in sludge uranium is less than these values, but, at the same time, sludge uranium contains plutonium generated during irradiation. Since plutonium is fissionable and present in quantities that exceed its minimum critical mass, it must be taken into account.

A model for K Basin sludge is developed too partially account for burnup (see Section 5). This model assumes all uranium is enriched to 0.84 wt% ^{235}U and contains plutonium derived from the irradiation process. The composition of K Basin uranium was obtained from the Safeguards Control Accountability Transaction System (SCATS) database (Schlosser 1990) which shows composition after irradiation. According to SCATS, the average ^{235}U enrichment is

0.74 wt% for KE Basin and 0.81 wt% for KW Basin. The highest burnup for any K Basin uranium corresponds to a plutonium/uranium mass ratio of 0.0030 (0.30 wt%) and a ^{240}Pu /plutonium mass ratio less than 0.167 (16.7 wt%).

In the K Basin sludge model the ^{235}U enrichment is held constant, and maximum k_{∞} occurs when the plutonium content in the uranium corresponds to highest burnup. When optimally moderated, fully reflected, and formed into a sphere, K Basin sludge at any level of burnup will exhibit a k_{eff} less than that of the model, which assumes that the plutonium content is at its highest possible value.

2.3 CHEMISTRY

This evaluation does not provide new information on chemistry, but rather it presents information from published documents. The primary sources of information on plutonium chemistry are Serne et al. (1996) and Whyatt et al. (1996), and the primary source for uranium chemistry is Daling et al. (1997). Uranium and plutonium have different chemical characteristics and are assumed capable of chemically separating from each other.

Tank waste is maintained alkaline with a minimum pH of 8.0 to ensure that uranium and plutonium remain combined with solids. Whyatt et al. (1996) concludes that plutonium primarily resides in the solid phase in the form of agglomerates (conglomerates). The tendency of iron to coprecipitate and to agglomerate with plutonium is a primary reason for using it as a neutron absorber. The solubility of plutonium in alkaline salt solution is low enough that saturation concentrations in waste liquids are at least 30 times lower than the minimum concentration needed to support criticality.

2.4 PARTICLE SIZE

Resonance neutron absorption in low enriched uranium particles in water is affected by particle size. For 0.84 wt% enriched uranium criticality is not possible for any particle size. For 0.95 wt% enriched uranium, criticality is precluded when particles are less than 0.13-cm (0.05-in.) diameter.

The settling velocities in a liquid are different for particles of different size and density. In the process of gravity separation, waste components may settle into layers according to particle size. Processing of K Basin sludge will reduce the predominant particle size to less than 10 μm . At this particle size the greatest increase of plutonium concentration possible by gravity separation will not exceed 3.0 times the initial concentration (Serne et al. 1996, Daling et al. 1997). Although a small weight fraction of particles might be larger than 10 μm , their proportion decreases rapidly with increasing size, and they were taken into account in establishing the maximum segregation factor. At this small size, variations in density are not important to settling velocity. When particles agglomerate, a much larger particle size may be produced, but it is the size of the primary particles that is important because the proportion of primary particle types in the agglomerate will reflect the overall sludge composition.

There are two processes being considered for achieving the 10 μm particle size in the sludge to be transferred. In one process high energy milling would be used to process the sludge in order to achieve the small particle size. In the other process the sludge would be dissolved in acidic solution and then reprecipitated.

2.5 NEUTRON ABSORBERS

The basic parameter to specify the quantity of neutron absorber material to be added is the absorber/heavy-metal mass ratio, where heavy metal refers to the sum of uranium plus plutonium. Since plutonium is less than 0.30 wt% of the heavy metal, the heavy metal and the uranium is very nearly the same in quantity. For this reason, the absorber/uranium mass ratio, which is easier to describe and implement, is used to specify the iron content. Because uranium and plutonium contents are related through the K Basin sludge model, the state of the sludge is determined by the iron/uranium mass ratio, and there is no need to specify the iron/²³⁵U and iron/plutonium mass ratios as separate parameters.

A limit of 0.84 wt% (maximum) is placed on ²³⁵U enrichment in K Basin sludge received at tank farms. If depleted uranium is added to lower the enrichment to less than 0.84 wt%, the depleted uranium must not contain more than 0.05 wt% plutonium. The depleted uranium shall be blended with the sludge uranium and have a similar particle size distribution. Once blended, the depleted uranium cannot be chemically or physically separated from sludge uranium by ordinary mechanisms.

Iron is added as a neutron absorber to ensure subcriticality for credible increases in plutonium concentration. The iron/uranium mass ratio for K Basin sludge shall be no less than 0.768 (minimum). This quantity, when homogeneously mixed, ensures that k_{∞} will not exceed the Upper Safety Limit (USL) of 0.881 under credible abnormal conditions, including a 3-fold increase in plutonium concentration or a 3-fold decrease in iron concentration. Neutron absorbing components in sludge, other than iron and ²³⁸U, that are not taken into account in this evaluation will provide added conservatism.

2.6 COMBINING K BASIN AND DST AW-105 SLUDGE

DST AW-105 is a 23-m (75-ft) diameter cylindrical tank with a capacity of 3.8 ML (1.0 Mgal). It contains 1,643,000 L (434,000 gal) of waste from the processing of fuel at the Plutonium Uranium Extraction (PUREX) facility (Hanlon 1998). The plutonium inventory is 22.95 kg, and the solids/plutonium mass ratio is estimated to be 55,400 (LMHC 1998). The average plutonium areal density is estimated to be 55.9 g/m² (5.2 g/ft²), or 2.2% of the minimum critical areal density. The largest measured plutonium concentration is 0.024 g/L (Braun et al. 1994, WHC 1995), less than 1% of the minimum required for criticality. This measurement was made before the PUREX transfers in 1995. Available information for DST AW-105 sludge composition indicates a high mass ratio of neutron absorbers to fissile material and a large margin of subcriticality. Studies of the waste generation processes have concluded that no significant volumes exist with a plutonium concentration much above the tank average.

Knowledge of the origin of K Basin sludge and the process for disposal provides high assurance of its composition. In DST AW-105 K Basin sludge will primarily determine k_{eff} . At any water content the combined sludge will not produce a k_{eff} that exceeds what is possible for K Basin sludge by itself. The calculational model assumes an unlimited volume of sludge at the most reactive allowed composition.

DST AW-105 sludge and K Basin sludge have large mass ratios of absorbers in relationship to fissile isotopes. Mixing of these sludge types will not change the overall mass ratio of neutron absorbing solids to fissile isotopes. Even if mixed, the absorber/fissile mass ratio will remain large throughout the sludge, and the margin of subcriticality will remain large. Before being received at tank farms, samples from each batch of K Basin sludge will be analyzed to verify composition.

2.7 CALCULATIONAL METHOD

XSDRNPM (two-dimensional deterministic criticality code), a code in the Standardized Computer Analysis for Licensing Evaluation (SCALE) code package (Version 4.3), is used to calculate k_{∞} . These calculations are supported by *Verification and Validation of the XSDRNPM Code for Tank Waste Calculations* (Rogers and Niemer 1999). This report justifies an USL on k_{∞} of 0.90 after code bias and calculational uncertainty at the 95% confidence level are taken into account. This defines the upper bound for acceptable calculations. To satisfy the USL, a XSDRNPM calculated k_{∞} must not exceed 0.881 for 27-group ENDF/B-IV cross sections, or 0.882 for 44-group ENDF/B-V cross sections, for any waste configuration over the entire range of credible compositions, including optimal moderation and full reflection. This evaluation uses 27-group cross sections.

3.0 SCOPE, CRITERIA, AND LIMITS

Basic assumptions used in this evaluation, criteria for acceptability, and limits and controls to be used in the discharge and storage of K Basin sludge at tank farms are provided.

3.1 OBJECTIVES

This evaluation has two objectives. The first is to establish acceptance criteria for the discharge and storage of K Basin sludge into DST-AW-105, and the second is to recommend activities that ensure safe, efficient, and cost effective compliance with acceptance criteria. Negative impacts to current and future waste management operations by the addition of neutron absorbing solids will be minimized to the extent possible.

3.2 SCOPE

This evaluation establishes criticality safety limits and controls for the discharge and storage of K Basin sludge (floor, pits, canister, and fuel wash) into DST AW-105. A technical basis is established for limits and controls that minimize the potential for segregation of fission isotopes from neutron absorbers during waste transfer and storage operations. Since the chemistry of waste tank contents is largely unobservable, verification of compliance with TWRS requirements must be completed either during the dissolution and precipitation process or by sampling the final product.

Criticality safety evaluation of the solids treatment process is not in the scope of this evaluation. However, the resulting chemistry of that process is important to the disposition at tank farms, and controls are identified to help ensure compliance with TWRS requirements.

3.3 BASIC ASSUMPTIONS

Some parameters basic to this evaluation have been established using assumptions that bound the true composition of the sludge. When received, sludge must fall within the description and requirements provided by these basic assumptions.

1. Basin sludge chemistry will be compatible with tank waste chemistry.
2. Heat generation and hydrogen gas production are not part of the criticality safety evaluation, since they do not increase the potential for a criticality.
3. Basin sludge will be stored in DST AW-105.
4. The ^{235}U enrichment of uranium received at tank farms will be no greater than 0.84 wt%.

5. Iron will be added and integrally mixed prior to receipt at tank farms to provide an acceptable margin of subcriticality.
6. The particle size will be small enough to preclude gravity segregation of plutonium by more than a factor of 3.0.

3.4 CRITERIA OF ACCEPTABILITY

Criteria are established to provide a margin of subcriticality such that waste management operations are not negatively impacted.

3.4.1 Double Contingency

The basic criteria for criticality safety is the Double-Contingency Principle stated in DOE Order 5480.24 (DOE 1992) as:

Process designs shall incorporate sufficient factors of safety to require two unlikely, independent, and concurrent changes in process conditions before a criticality is possible. Protection shall be provided by either:

1. The control of two independent process parameters (which is the preferred approach, if practical) or
2. A system of multiple (at least two) controls on a single parameter.

In all cases, no (credible) single failure shall result in the potential for a criticality accident.

A contingency, as defined by DOE-STD-3007-93 (DOE 1998), is a "possible but unlikely change in a condition/control important to the nuclear criticality safety of a fissionable material operation that would, if it occurred, reduce the number of barriers (either administrative or physical) that are intended to prevent an accidental nuclear criticality."

3.4.2 Criteria for Limits and Controls

Specific criteria for establishing limits and controls to ensure compliance to the Double-Contingency Principle are:

1. k_{∞} shall not exceed 0.90 at the 95% confidence level, after accounting for the range of possible compositions, optimal moderation, full reflection, calculational bias, and possible changes in waste composition resulting from segregation through gravity settling, chemical processes, and measurement inaccuracies.

(Note: This is specified in HNF-PRO-537 (HNF 1997).)

2. The criterion for k_{∞} shall be met after consideration of the following conditions:
 - a. Separation of uranium from other solids.
 - b. Separation of plutonium from iron and uranium.
 - c. Separation of iron from uranium and plutonium.
3. The plutonium/uranium mass ratio must not exceed 0.0030.
4. If depleted uranium is added:
 - a. The depleted uranium and sludge uranium shall be mixed to achieve uniformity of enrichment. Calculation of the final enrichment of the blended uranium shall include compensation for uncertainties and variations in the initial enrichment and the ^{235}U content of the depleted uranium added.
 - b. The plutonium content shall not exceed 0.05 wt% of the uranium.
5. The predominant particle size for uranium, plutonium and iron shall not exceed $10\ \mu\text{m}$ (0.0004 in). Size of other components is not limited.

(Note: This is intended to mean that particles larger than $10\ \mu\text{m}$ compose only a small mass fraction and are not capable of increasing the segregation factor above 3-fold.)
6. When combined with tank farms sludge, k_{eff} of the combined sludge shall not exceed 0.90, under the worst credible conditions, after accounting for optimal moderation, full reflection, and possible changes in waste composition.

3.4.3 Discussion

The criterion for k_{∞} ensures subcriticality under normal and credible accident conditions. k_{∞} is determined for an unlimited (infinite) volume of waste at optimal moderation. This is conservative for any finite volume of waste with full reflection.

The criterion on k_{∞} is taken from procedure HNF-PRO-537, Sections 1.4.1.2 and 1.4.1.3 (HNF 1997). This procedure provides for three categories of subcritical margin, depending on the accuracy of experimental data available to which the calculated configuration can be compared. The quality of available experimental data for uranium and plutonium is sufficient to justify a maximum k_{∞} of 0.95. Although benchmark experiments are available in which iron is present as fuel element cladding and as structural material, none of the available experimental

descriptions included near the quantity of iron in K Basin sludge and none could be considered as homogeneous. Rogers and Niemer (1999) concludes that the quality of available experimental data for iron is not sufficient to justify this criterion and stipulate that the acceptable USL on k_{eff} for XSDRNPM calculations of tank waste systems with a high iron content be 0.90. At the 95% confidence level, this provides an USL of 0.881 on the calculated k_{eff} .

The *Nuclear Criticality Safety Guide* (Pruvost and Paxton 1996) provides a subcritical limit on ^{235}U enrichment for uranium mixed homogeneously with water as 0.93 wt% for uranium metal and 0.96 wt% for UO_2 , UO_3 , U_3O_8 or UO_3F_2 . By restricting enrichment to a maximum of 0.84 wt% ^{235}U , k_{∞} for uranium is assured of being less than 0.95, and criticality involving only uranium is precluded, even after separation of uranium from neutron absorbing solids. Since it is impossible under tank waste storage conditions to separate ^{235}U from ^{238}U , no other neutron absorbing solids are needed to maintain subcriticality for the uranium portion of the sludge.

An important reason for restricting the enrichment of uranium to no more than 0.84 wt% is to remove the need to consider higher enrichments in the calculational model. This restriction reduces the quantity of iron that has to be added to meet the subcriticality criterion.

Plutonium contained in the uranium makes this evaluation more complex. Uranium enriched to 0.84 wt% ^{235}U and also containing 0.30 wt% plutonium can be made critical with optimal moderation. This plutonium requires that iron be added to ensure an adequate margin of subcriticality.

The expected maximum neutron multiplication for existing DST AW-105 sludge is low, and neutronic interaction with K Basin sludge under expected conditions will have a negligible impact towards increasing neutron multiplication. Available characterization information for AW-105 sludge supports the conclusion that no significant localized pockets of high plutonium concentration exist (Braun et al. 1994).

3.5 CPS LIMITS AND CONTROLS

All waste brought into tank farms is governed by limits and controls provided in an applicable CPS.

3.5.1 Current Tank Farms CPS Requirements

The current Criticality Prevention Specification for tank farms, CPS-T-149-00010, Rev. I-0, limits the plutonium equivalent concentration in incoming waste mixtures to no more than 0.033 g/L (DESH, 1997). In addition, the neutron absorber to plutonium mass ratio must be greater than at least one of the minimum subcritical ratios provided in an accompanying table. However, when the plutonium concentration in a batch is less than 0.001 g/L, transfer is permitted without a requirement on the absorber content. This limit is justified by the low importance of individual transfers at this concentration and is helpful because of the much greater difficulty of determining the absorber/plutonium mass ratio for low concentrations.

In addition a surveillance program is required to compare characterization data to an investigation level for the plutonium concentration in settled solids of 1.0 g/L (3.8 g/gal). An investigation level is defined as the measured value at which further investigation of the waste composition is required to verify compliance to criticality safety requirements. The investigation level provides an assessment of the overall state of the waste. In practice it is difficult to accurately characterize a batch of waste with a very low concentration of solids. The impact of individual batches on criticality safety is small, and there is no need to require a mass ratio of solids much above the minimum subcritical mass ratio. However, the mass ratio of total solids to total plutonium over many batches is more important. For this reason, the true limit for criticality safety is the surveillance limit of 1.0 g Pu/L in settled solids.

The plutonium inventory includes the total plutonium and the "plutonium equivalence" of the ^{235}U content in excess of 0.72 wt%. This definition excludes natural and depleted uranium. For 0.95 wt% and 1.25 wt% enriched uranium, the excess ^{235}U is equal to 0.23 wt% and 0.53 wt% of the uranium, respectively. At the highest uranium concentration possible in canister sludge, the ^{235}U inventory would be greater than permitted in the existing tank farms CPS. This CSER provides justification for revising the CPS to permit a higher concentration of fissile material in settled K Basin sludge solids. This higher concentration is permitted because of the accurate characterization of this sludge provided by a detailed knowledge of its history.

3.5.2 Limits and Controls for K Basin Sludge

The following limits and controls are established for K Basin sludge to be discharged into DST AW-105:

1. The ^{235}U enrichment shall not exceed 0.84 wt% (maximum).
2. The iron/uranium mass ratio shall be at least 0.768 (minimum).
3. The plutonium content shall not exceed 0.30 wt% (maximum) of the uranium.
4. Depleted uranium added to K Basin sludge shall not contain more than 0.05 wt% (maximum) plutonium. No other plutonium shall be added.
5. Added depleted uranium and iron shall be uniformly mixed with the sludge.
6. The predominant particle size for uranium, plutonium, and iron shall be less than 10 μm (0.004 in.).

Note: Dissolution and reprecipitation will guarantee a particle size less than 10 μm (0.0004 in.). Size of other components is not limited.
7. pH shall be at least 8.0 (minimum).

8. Plutonium, uranium, and ^{235}U enrichment shall be verified by sample analysis on a batch-by-batch basis.
9. An independent verification of sample analysis accuracy and CPS compliance shall be documented.

3.5.3 Discussion

The basis for the above limits and controls are provided in this evaluation report.

For K Basin sludge there are fixed relationships between ^{238}U and ^{235}U , between ^{239}Pu and ^{240}Pu , and between total uranium and plutonium, and the range of these relationships is known. This makes it possible to define the fissile component simply as heavy metal, which is the sum of the uranium and plutonium. The iron/heavy-metal mass ratio (same as $\text{Fe}/(\text{U}+\text{Pu})$) can then be used as a measure of subcriticality. Since uranium mass is smaller than heavy metal mass by less than 0.30 wt%, the iron/heavy-metal mass ratio is essentially identical to the iron/uranium mass ratio. Since the iron/uranium mass ratio is simpler to understand and to implement, it is used for CPS limits

Separation into layers according to differences in particle size and density occurs as a result of differences in settling velocity. This separation is referred to as gravity segregation. Serne et al. (1996) reports that an increase in fissile concentration of a factor of 2.5 is possible with a particle size of 10 μm as a result of gravity segregation of tank waste components that have been mixed and allowed to settle. However, a segregation factor of 3 is used for this evaluation, based on a recommendation by Daling et al. (1997) for K Basin sludge. The margin of subcriticality must be large enough to compensate for this degree of segregation.

Plutonium/uranium mass ratios are accurately determined from the irradiation history. Values used to define the model for K Basin sludge were selected to be conservative relative to the actual sludge. If plutonium content in excess of 0.30 wt% of the uranium is obtained from an analytical measurement, a review should be made to verify accuracy and to understand the reason for the high value.

4.0 FACILITIES, EQUIPMENT, AND OPERATIONS

This section describes equipment and operations pertinent to understanding the characteristics of K Basin sludge and the treatment processes that have a direct bearing on criticality safety at tank farms. Details of some operations prior to receipt at tank farms are included to help understand the overall process.

4.1 DESCRIPTION OF K BASINS

The KE and KW Basins are 125 ft long by 67 ft wide, water-filled pools located in the 105-KE and 105-KW spent fuel storage facilities. The basins are divided into a complex of seven interconnecting pool areas; each designated for a specific operational function.

Spent N Reactor fuel elements are stored in fuel canisters. Some fuel elements were damaged when discharged from the reactor to the extent that the uranium metal fuel is exposed to the water. Over time, the slow corrosion of exposed uranium metal has led to the formation of particulate material settled on the basin floor. In some cases, partial disintegration and breakage of fuel elements has occurred. Particulate and broken pieces of uranium fuel have mixed with debris on the KE Basin floor to form "KE Basin Sludge." In addition, fuel fragments in the bottom of storage canisters will also be disposed of as sludge. At the KW Basin the sludge is primarily found in the North loadout pit and in storage canisters.

4.1.1 Fuel Element and Assembly Inventories

A large storage array for N Reactor fuel elements and assemblies is maintained in each basin. Fuel assemblies are stored in canisters consisting of two cylindrical barrels welded together. Each barrel is designed to hold seven assemblies, and a full canister holds fourteen assemblies. An N Reactor fuel assembly consists of an "inner" and an "outer" fuel element. The outer element is an annular tube into which the inner cylindrical element is inserted. The primary types of N Reactor fuel assemblies stored in the basins are Mark IA and Mark IV. An unirradiated Mark IV assembly contains uranium enriched to 0.95 wt% ^{235}U in both the inner and the outer element. A fresh Mark IA assembly contains an inner element of uranium enriched to 0.95 wt% and an outer element enriched to 1.25 wt%.

A detailed description of fuel stored in the K Basins is provided in *105-K Basin Material Design Basis Feed Description for Spent Fuel Project Facilities, Volume 1, Fuel Praga* (1998). Praga describes the inventory of irradiated N Reactor fuel as follows:

The KE Basin stores 3,672 canisters containing 51,073 fuel assemblies, elements, or pieces. The KW Basin stores 3,841 canisters containing 53,964 assemblies, elements, or pieces. The total mass of fuel elements at the KE Basin is approximately 1,233 metric tons, and the total

mass in the KW Basin is approximately 1,038 metric tons. Of this total mass, there are approximately 1,143,600 kg of uranium and 2,155 kg of plutonium in the KE Basin and approximately 951,900 kg of uranium and 1,875 kg of plutonium in the KW Basin.

The fissile material in the K Basin facilities from N Reactor is comprised of 0.71, 0.95, and 0.95/1.25 wt% uranium fuel assemblies as shown in Table 4-1. Before 1998 all fuel in the KE Basin was comprised of uranium enriched to no more than 0.947 wt%. In October 1998 Schwinkendorf (1998) issued a criticality safety evaluation report for the storage of 39 additional N Reactor fuel elements in KE Basin, of which 17 were to be Mark IA elements containing 1.25 wt% uranium. KW Basin, however, contains 628 metric tons of uranium (MTU) which, before irradiation, was enriched to 1.25 wt%.

Table 4-1. Basin Inventories of N Reactor Fuel in Metric Tons.

Location	0.71 wt% ²³⁵ U	0.95 wt% ²³⁵ U	0.95/1.25 wt% ²³⁵ U
KE Basin	8.8	1141	----- ¹
KW Basin	1.12	323	628

Note:

¹Temporary storage of 17 Mark IA elements with several hundred kilograms of 1.25 wt% uranium.

In addition to N Reactor elements, there are fuel elements from eight single-pass reactors (SPR) which operated at Hanford to produce plutonium between 1944 and 1955. Schwinkendorf (1997) states that fuel from the Hanford single pass reactors comprises "3.4 metric tons of the total irradiated fuel inventory (i.e., 0.16 wt%)." The enrichment before irradiation was as high as 2.1 wt% ²³⁵U. The current inventory in SPR fuel in K Basins is given as 106 kg ²³⁵U of 1.25 wt% enriched uranium and 183 kg ²³⁵U of 0.95 wt% enriched uranium. The remainder of the SPR fuel in the basins is natural or depleted uranium metal. The majority of this fuel is assumed to be in good condition with minimal cladding damage. Praga (1998) provides a physical description of this fuel.

The vast majority of the fuel assemblies are zircalloy clad. However, in KW Basin, 830 elements are aluminum clad, and in KE Basin, 138 elements are aluminum clad.

4.1.2 Limits and Controls for Basin Operations

Chapter 6 of the *K Basins Safety Analysis Report* (Meichle 1996) discusses controls used for criticality safety. It states:

The storage of irradiated N Reactor fuel in the KE and KW Fuel Storage Facilities has been evaluated for potential nuclear criticality accidents, during normal K Basin fuel handling and storage, and found to be safe and within the nuclear criticality safety criteria

and limits established for the fuel storage basins operational activity (Toffer and Eaves 1981, Roblyer 1982, Toffer and Tollefson 1982, Tollefson 1983).

Additional criticality evaluations of the accumulation of fissile isotopes in ion exchange columns, sand filters, cartridge filters, and other components, were performed by Erickson (1994), Wittekind (1994a), and Schwinkendorf (1994).

Nearly all fuel in the basins has been in the form of fuel elements and assemblies. Generally, fuel in a form different from a complete assembly or element is called "scrap." Limits provided in the K Basin CPS for scrap were determined using the assumption that the pieces are of an optimal size and are separated at an optimal spacing in water. The mass of uranium required to achieve criticality for undamaged elements is greater than for broken pieces of fuel. The smaller mass limit for scrap is conservative when applied to uranium metal of any size, whether large or small. Criticality safety limits used for scrap batches in basin activities, based upon Schwinkendorf (1995) and Tollefson (1983), are shown in Table 4-2.

Table 4-2. K Basin Criticality Prevention Specification Mass Limits.

Maximum ²³⁵ U enrichment	0.95 wt%	1.25 wt%
Maximum mass of uranium and scrap with no geometry control	2,031 kg	933 kg

Sludge on basin floors was spread over a wide area with an areal density well below the minimum for which criticality is possible.

4.1.3 Sludge Characterization Data

Sludge characterization data in *105-K Basin Material Design Basis Feed Description for Spent Fuel Project Facilities, Volume 2, Sludge* (Pearce et al. 1998) "utilizes the most current characterization data available to define the various sludge inventories." Feed descriptions in this document apply to K Basin sludge that has not been treated. Waste as it will be received at tank farms will have undergone a treatment process. However, the uranium enrichment and plutonium content will not be changed by treatment.

K Basin sludge is categorized as being from KE Basin or from KW Basin. Five process streams, designated KE1, KE2, KW1, KW2, and KW3, are defined by the locations where the sludge is collected and held in interim storage. Interim storage for process stream KE1 is in the KE Basin Weasel Pit and for the KE2 stream is in the Integrated Water Treatment System (IWTS) Knockout Pots. Process stream KW1 contains sludge retrieved from pit and floor areas. Interim storage for process stream KW2 is in the IWTS Knockout Pots and for process stream KW3 is in the settler tanks. See Appendix C for more information.

Particle size is an important determiner of the ability of different waste components to segregate from other components. In its original state, K Basin sludge contains larger particles

than will be permitted to be in sludge stored at tank farms. Process streams KE1, KE2, KW1, and KW2 contain particles up to 6350 μm (0.25 in.) in diameter. Streams KE2 and KW2 do not contain particles smaller than 250 μm and 500 μm , respectively. The maximum particle size for process stream KW3 is 500 μm .

4.2 SLUDGE TREATMENT PROCESS

Two processes are being considered for the treatment of K Basin sludge. The first is a chemical process in which the uranium and neutron absorbers are dissolved in nitric acid. The second is a high-energy milling process that grinds the sludge components into extremely small particles. In either process the sludge will be treated to meet TWRS acceptance criteria.

4.2.1 Chemical Baseline Process

Sludge treatment will be done by dissolving the fuel constituents in nitric acid, separating the insoluble material, adding neutron absorbers for criticality safety, and reacting the solution with caustic to coprecipitate the uranium and plutonium. A truck will transport the resulting slurry to DST AW-105. The undissolved solids will be treated to reduce the transuranic (TRU) and ^{137}Cs content, stabilized in grout, and transferred to the Environmental Restoration Disposal Facility (ERDF) for disposal. The preliminary design flowsheet for chemical treatment includes the following operations (Westra et al. 1998):

- Sieving the sludge on a screen to remove the organic resin beads and some inorganic ion exchange media followed by separation of the resin from larger sludge particles in an elutriation column.
- Dissolving the sludge in nitric acid.
- Physically separating residual solids (mostly zirconium, sand, dirt, and the remaining inorganic ion exchange media) from the solution.
- Adding iron and/or depleted uranium as a neutron absorber
- Precipitating the solution using caustic solution
- Chemically adjusting the solution using sodium nitrite
- Leaching the organic resin beads and insoluble solids to remove absorbed TRU constituents
- Combining and stabilizing the resin beads and insoluble solids in a grout matrix.

4.2.2 High-Energy Milling Process

The second sludge treatment process under consideration is a high-energy milling process using a grinder to reduce the sludge particle size through fracturing and mechanical abrasion. Upon arrival at the facility, sludge would be removed from the transport container and placed in the lag storage vessel to await transfer to the grinder. Grinding would reduce the sludge to fine particles, and the metallic uranium particles would oxidize in the water to a stable form.

From the grinder, the slurry would pass through a screen. Particles smaller than 10 μm diameter would go to the adjustment tank and larger particles would return to the grinder. Neutron absorber material (i.e., iron) would be added as needed to meet the criticality safety requirement. Sodium hydroxide and sodium nitrate would be added to ensure alkalinity.

Cladding solids greater than 1000 μm diameter would be removed and sent to be grouted. Oversize material remaining after completion of milling would be solidified and disposed of at the ERDF. Polychlorinated biphenyl (PCB) extraction would be accomplished primarily by adsorption of the PCBs onto the polyurethane liner of the grinder.

The treated slurry would be transferred to a DST for eventual processing with other Hanford tank waste at the planned vitrification facility. The low-activity waste (LAW) would be disposed in Land Disposal facilities, and the high-level waste (HLW) would be sent to the geologic repository.

4.3 SLUDGE RECEIVING SYSTEM

The Sludge Transportation System will move K Basin sludge to tank farms in batches limited to about 6000 L. Sludge will arrive at tank farms in the transport container on a flatbed trailer. The transporter will be positioned at the Sludge Receiving Station where the sludge will be transferred into DST AW-105.

The DST AW-105 Sludge Receiving Station will have four primary features: 1) a spill retention basin; 2) a male coupler for connecting to the Sludge Transportation System; 3) a sludge transfer line; and 4) a pump and associated equipment. Sludge will be transferred through a flexible 3.8 cm (1.5 in.) diameter hose at a maximum rate of 3 L/sec (50 gpm).

During sludge transfer, the trailer will be stationed on the spill retention basin with the wheels chocked. The volume of the spill retention basin is larger than the volume of the transport container. Features will be provided to automatically stop the pump upon detection of leakage.

4.4 DOUBLE-SHELL TANK AW-105 RECEIVER TANK

DST AW-105 is a 23-m (75-ft) diameter storage tank with an operational capacity of 3.78 million L (1.0 million gal). Prior to addition of K Basin sludge, AW-105 contains

1,643,000 L (434,000 gal) of waste from the processing of fuel at the Plutonium Uranium Reduction Extraction Plant (PUREX) (Hanlon 1998). At its operational storage limit the waste depth would be 30 ft, 2 in. If overfilled, the maximum volume of waste would be 4.4 million L at a liquid level of 35 ft, 2 in. This tank is located underground with about 8 ft of soil covering the dome. Information on the contents of this tank is provided in Appendix D.

The Double-Shell Tank Plutonium Inventory Database shows the October 1998 inventory for DST AW-105 to be 22.95 kg of plutonium, based on sample analysis and the total solids mass to be 55,400 times greater than this (LMHC 1998). The largest measured plutonium concentration reported by Braun et al. (1994) for this tank is 0.024 g/L, less than 1% of the minimum required for criticality under the most idealized conditions. The average plutonium areal density is estimated to be 55.9 g/m² (5.2 g/ft²), or only 2.2% of the minimum critical areal density. Wyatt et al. (1996) concludes that the plutonium in tank waste primarily resides in the solid phase and that the saturation concentrations of plutonium in supernatant liquid are at least 30 times lower than needed to support criticality.

A detailed discussion of the contents of this tank is provided in Appendix A of *Feasibility Report on Criticality Issues Associated With Storage of K Basin Sludge in Tank Farms* (Daling et al. 1997).

4.4.1 History Prior To 1995

Discharges to this tank have always been made according to limits provided in a CPS. Prior to discharge, waste was held at the processing facility in a holdup vessel until the accumulated volume became large enough for transfer. The volume of the holdup tank was typically 18,900 L (5,000 gal). At the time of discharge the average plutonium concentration in a batch was required to be less than 0.013 g/L (0.05 g/gal).

On January 1, 1994, the inventory is listed as 15,180 g Pu, and the tank averaged solids/plutonium mass ratio was 88,898. On January 1, 1995, the inventory is listed as 17,230 g Pu, and the tank averaged solids/plutonium mass ratio was 68,539. Until the 1995 transfers described below, no single batch ever contained more than 205 g of plutonium.

4.4.2 PUREX Transfers in 1995

The top layers of solids in DST AW-105 contain the highest plutonium concentration in this tank. These layers were formed from 18 transfers between January and April 1995 using discharge limits requiring the presence of cadmium to compensate for allowing an increase in the plutonium concentration (Carter 1984). The Plutonium Inventory Database shows that these transfers increased the plutonium inventory by 5,613 g from 17,230 g to 22,843 g. At least part of this increase in "plutonium inventory" was due to ²³⁵U in enriched uranium being treated as if it were plutonium. The average plutonium content per transfer was 311 g, while the largest transfer contained 446 g. The average plutonium equivalent concentration in these discharges was 0.018 g/L, and the highest value for a single transfer was 0.031 g/L.

The average uranium concentration per transfer varied between 12.2 g/L and 34.2 g/L. The lowest uranium/plutonium mass ratio for any transfer was 653, while the largest value was 3181. Out of the 18 transfers, only two had an uranium/plutonium mass ratio smaller than 770, the minimum subcritical mass ratio for natural uranium, and none was smaller than 650. The median mass ratio was about 1,050. The $^{238}\text{U}/(^{235}\text{U} + \text{Pu})$ ratio was greater than needed to ensure subcriticality for an unlimited volume of this waste. However, the ratio was small enough to require cadmium to meet the CPS requirements (Carter 1984).

The cadmium to fissile atom ratio in the solution varied between 2.6 and 11. This quantity of cadmium ensures subcriticality under all conditions, regardless of other components present in the solution. Upon entering DST AW-105 each batch of waste would have spread out into a layer and settled. The batches with the lower U/Pu mass ratios would be in layers sandwiched between layers formed by batches with higher U/Pu mass ratios. If the cadmium settled at a different speed than the plutonium bearing solids, this would lead to layers of enhanced cadmium concentration formed between layers of higher uranium and plutonium concentrations. Nevertheless, the final configuration would remain well subcritical.

If it is assumed that the 5,613 g of plutonium is spread uniformly over the entire area of the tank, the areal density would be 13.7 g/m^2 (1.27 g/ft^2). This areal density is 188 times smaller than the minimum critical plutonium areal density of $2,582 \text{ g/m}^2$ (240 g/ft^2). No scenarios can be postulated in which the ^{235}U and/or plutonium can be segregated into a volume compact enough and a concentration high enough for criticality to occur. Even if the cadmium is assumed not to have remained with the enriched uranium, the proportion of ^{238}U is sufficient to ensure subcriticality.

4.4.3 Mass Ratios And Subcritical Fractions

The mass ratio of neutron absorbing solids to plutonium is a measure of the margin of subcriticality. For a specific waste component the actual-to-minimum subcritical fraction is defined as the actual mass ratio divided by the corresponding minimum subcritical limit mass ratio. The actual-to-minimum subcritical fraction is often just called the subcritical fraction. When the sum of subcritical fractions exceeds 1.0, homogeneous waste is subcritical.

Using process records, Agnew (1995) determined the sum of actual-to-minimum subcritical fractions to be 18.6 for the insoluble components and 54.4 for the soluble components.

Whyatt et al. (1993) concludes that zirconium is the most accurately known constituent in the waste stream sent to DST AW-105. Process records show there to be 7.0 times as much zirconium as is required to maintain subcriticality. In addition, there is 1.4 times as much iron as required to maintain subcriticality. However, when a core sample was analyzed, the fractions obtained from the analysis were found to be significantly smaller. Based on a single core sample, the subcritical mass fractions are 1.28 for zirconium, 0.78 for iron, and 0.46 for lanthanum.

Braun et al. (1994) reports data from eight waste samples from DST AW-105. The largest measured plutonium concentration is 0.024 g/L. This value corresponds to an areal density of plutonium of 55.9 g/m², a value 46 times less than the minimum required for criticality. The solids-to-plutonium mass ratio is estimated to be at least 50,000. The fraction of the actual-to-minimum subcritical mass ratio for this waste is estimated to be 64. From eight samples the smallest sum of the fractions found is 15.5, and this is broken down to be 0.50 for insoluble components and 15 for soluble components. The plutonium concentration in the composite sample from which these low values were derived is listed as 0.019 g/L.

These values clearly indicate that the neutron absorber/plutonium mass ratio is far more than the minimum required for subcriticality. Nevertheless, the low mass ratio reported by Braun et al. (1994) for insoluble-components appears to run counter to Agnew (1995) and Whyatt et al. (1996). The iron fraction in this sample is only 0.08, far less than expected. The quality of the analytical measurement that gave this result is unknown. The total tank iron inventory based on process records should provide a much better indicator of the state of the waste, and Whyatt uses this to provide an average subcritical fraction for iron of 1.4.

4.4.4 Stratification

An important consideration to criticality safety is the formation of strata of different composition. Waste was discharged into DST AW-105 from PUREX as a large number of batches. Since the composition differed from one batch to another, the compositions of strata reflect the variation in batches. Transfer records provide a means of estimating the composition and thickness of strata and a measurement of the total depth of sludge provides a check on the total quantity discharged. This data can be used to build a model of plutonium concentration as a function of depth. During the period of days between discharges the solids in a batch had time to settle before the next batch was introduced. The mass of plutonium in a layer is assumed to be the same as in the discharge.

Differences in particle size and density will cause differences in the rate at which particles settle. This difference in settling velocities can result in particle segregation into layers. Whyatt et al. (1996) concluded that particles are flocculated under normal tank conditions and this flocculent waste contains agglomerates in which the plutonium is tightly bound with other solids. This agglomeration counteracts any tendency of plutonium to separate from other solids. However, Whyatt et al. (1996) used the Transient Energy Momentum and Pressure Equations in Three Dimensions (TEMPEST) code to model particle dynamics and to estimate the degree by which the plutonium concentration might increase upon settling. These simulations indicated that the concentration increase would not exceed a factor of 2.5 for particle size below 10 μm . An important assumption in this model is that the plutonium associates with a particular particle size. In practice this assumption is conservative because the plutonium would actually be composed of a range of particle sizes, and the distribution of these particle sizes would be similar to the distribution of particle sizes for other waste components. After gravity segregation the relative proportion of components will remain about the same within each layer of waste. There would be very little, if any, change in the plutonium concentration within these layers.

Since this model did not take agglomeration into account, this concentration increase overestimates the actual factor of increase. Also, plutonium was discharged into the tank as part of a batch of waste. Even if particles containing plutonium settled at a different velocity, all of the waste in a given batch would settle into a layer before discharge of the next batch. This would result in plutonium-rich layers sandwiched between layers of neutron absorbing solids. The total quantity of plutonium in a layer must equal the quantity discharged in a batch. A layer of high plutonium concentration would have to be thin to maintain a constant total mass of plutonium.

Daling et al. (1997) examined the variation in plutonium concentration in sludge formed from discrete discharges of waste from PUREX. Each discharge formed a layer on top of the existing waste surface. The final waste configuration is one of stratification. Daling et al. (1997) concluded for DST AW-105 that elevated plutonium concentrations as high as 0.31 g/L are possible, but only in thin layers (< 1 cm thick). Over a thicker slice of waste the average plutonium concentration will be less, with the average for the tank as a whole being 0.02 g/L. Measured from the bottom of the sludge, a region of high plutonium concentration is seen to exist between 88 and 94 in. This layer contains almost 7 kg of plutonium. This layer contains the PUREX transfers made in 1995. Above this region is a 10-in. thick layer with very little plutonium (< 0.01 g/L). The top of the sludge lies at 104 in.

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5.0 METHOD OF ANALYSIS

A model of K Basin Sludge is developed by which conservative critical parameters can be determined. This model takes into account irradiation history.

5.1 CALCULATIONAL METHOD

XSDRNPM (Greene and Petrie, 1995), a deterministic criticality code, was used to calculate criticality parameters. This code is included in the Standardized Computer Analysis for Licensing Evaluation (SCALE-4.3) nuclear criticality safety software package. Calculations were made on a 200 MHz AST Bravo MS 5200M personal computer using the SCALE 27-group ENDF/B-IV (Evaluated Nuclear Data File) cross-section library.

SCALE 4.3 is a collection of modules designed to perform criticality calculations. For this evaluation two functional modules were used which require user input: NITAWL-II (Greene et al., 1995) and XSDRNPM. NITAWL-II applies the Nordheim Integral Treatment to perform neutron cross-section processing in the resolved resonance range for ENDF/B resonance parameter data. The analyst specifies resonance parameters based on two options. The first option is to treat the resonance region as if the fissile material were in an infinite homogeneous medium. The second option is for the fissile material to be treated as a finite lump. XSDRNPM is a general purpose, discrete-ordinates code that solves the 1-D Boltzmann equation in slab, cylindrical, or spherical geometry.

Verification and Validation of XSDRNPM Code For Tank Waste Calculations by Rogers and Niemer (1999) provides a validation study that demonstrates that XSDRNPM accurately calculates k_{∞} for water-moderated, homogeneous systems of low enriched uranium, plutonium, and iron for simple geometries.

An USL is determined that defines the upper bounds on k_{eff} for acceptable calculations. The USL incorporates an administrative margin of subcriticality of 0.10, such that an acceptable calculation must not exceed a k_{eff} of 0.90. After taking into account code bias and after subtracting calculational uncertainty at the 95% confidence level, the USL is found to be 0.881 for the SCALE 27-group ENDF/B-IV cross section library and 0.882 for the 44-group ENDF/B-V library.

The range of applicability for XSDRNPM calculations is defined by: 1) uranium of any ^{235}U enrichment; 2) plutonium containing up to 20 wt% ^{240}Pu ; 3) iron in any proportion; 4) an average lethargy of neutrons causing fissioning (AEF) between 0.029 and 0.344 eV; and 5) geometric configurations for which neutron leakage is negligible. For these configurations the hydrogen-to-fissile atom (H/X) ratio falls between 125 and 3694, and the smallest dimension is large enough such that the neutron multiplication constant (k_{eff}) is indistinguishable from the neutron multiplication constant for an unlimited volume (k_{∞}).

5.2 IRRADIATION HISTORY

When heat is generated in a reactor, the quantity of ^{235}U decreases in a process called "burnup." This process of irradiation is likened to burning of wood, and unburned fuel is analogously called "green." During irradiation ^{238}U atoms absorb neutrons, and some of these decay and transform into ^{239}Pu . As irradiation continues, the quantity of ^{239}Pu increases, although part is lost through fissioning and part absorbs neutrons and becomes ^{240}Pu . ^{240}Pu does not fission with thermal neutrons and contributes very little to the fission process. In fact, absorption of neutrons in ^{240}Pu increases the quantity of plutonium needed for a self-sustaining chain reaction.

Fissioning of plutonium offsets part of the decrease in worth of the ^{235}U in maintaining a neutron chain reaction. If neutron absorption by fission products is ignored and, at the same time, conservative assumptions are made of the reduction in ^{235}U and increase in ^{239}Pu , the calculated reactivity might actually increase. However, Schwinkendorf (1997) made a careful assessment of the impact of irradiation on uranium enriched to 0.95 wt% and to 1.25 wt% ^{235}U and found that the maximum achievable k_{∞} will always be less than that for "green" uranium. This means that the quantity of neutron absorbers required to reduce the maximum achievable k_{∞} in K Basin sludge will be less for uranium that has been irradiated. The greater the degree of irradiation the smaller will be the quantity of absorbers required. The worth of plutonium generated relative to uranium burned is discussed in Appendix F.

When burnup is taken into account, a significant reduction can be made in the quantity of neutron absorbers required to maintain a margin of subcriticality. In this section a strategy is developed to account at least in part for burnup and to use this information to reduce need for adding neutron absorbers.

An inventory of fuel stored in the K Basins used to be maintained in the Safeguards Control Accountability Transaction System (SCATS) database spreadsheet (Schlosser 1990). In this spreadsheet fuel elements are grouped according to three types: Mark IA, Mark IV, or natural uranium. Each element is given a key number according to type and irradiation history. Based upon the original fuel element composition, its location in the reactor, and the length of time it was irradiated, the composition within each key category after irradiation was determined by the Oak Ridge Isotope Generation (ORIGEN) computer code. ORIGEN takes into account radioactive decay and outputs concentrations of important radioactive isotopes after a specified period of irradiation. The SCATS spreadsheet used for this evaluation was generated for January 31, 1992, (Wittekind 1998). Changes in fissile concentrations since 1992 are small, and it is conservative to ignore them. No ORIGEN calculations were made for this evaluation.

Table 5-1 show isotopes included in the SCATS database, and Table 5-2 shows the average composition of K Basin fuel. The quantity of ^{236}U is small relative to the quantity of ^{238}U , and its importance to criticality safety is negligible. For this evaluation ^{236}U is replaced by ^{238}U . The isotopes ^{238}Pu , ^{241}Pu and ^{242}Pu comprise a small fraction of the plutonium. Parametric studies in Appendix A were made with these isotopes replaced by an equal mass of ^{239}Pu .

However, the ^{241}Pu isotope can be worth as much as 2 times its weight as ^{239}Pu , and a simple one-to-one replacement may not be conservative. The impact of this replacement on k_{∞} is discussed in Section 5.4, and correction is made to the final results to ensure conservatism.

Table 5-1. Components Listed in SCATS Database.

Element	Isotopes
Uranium	^{235}U , ^{236}U , ^{238}U
Plutonium	^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu
Neptunium	^{237}Np
Krypton	^{85}Kr

Table 5-2. Average Composition of K Basin Fuel.

Element Ratio	KE Basin Average, ¹ wt%	KW Basin Average, ¹ wt%
$^{235}\text{U}/\text{U}$	73.6	81.3
$\text{U}(> 0.84) / \text{U}$ ²	33.0	50.0
$\text{Pu} / (\text{Pu} + \text{U})$	18.8	19.5
$^{239}\text{Pu} / \text{Pu}$	85.5	85.1
$^{240}\text{Pu} / \text{Pu}$	12.6	12.5
$^{238}\text{Pu} / \text{Pu}$	0.12	0.11
$^{241}\text{Pu} / \text{Pu}$	1.52	2.02
$^{242}\text{Pu} / \text{Pu}$	0.28	0.31

Notes:

¹Average for all fuel in basins, based on SCATS.

² $\text{U}(> 0.84) / \text{U}$ is fraction of all uranium enriched above 0.84 wt%.

Stress cracking of fuel elements is the precursor of sludge formation. Since the amount of stress cracking is proportional to irradiation time, most uranium in sludge will be found to originate from fuel with longer irradiation times. A longer irradiation time results in a greater reduction in ^{235}U enrichment, so the average ^{235}U enrichment in sludge is expected to be bounded by the average over all fuel in the basin.

Using the SCATS database, the average ^{235}U enrichment for all fuel in KE Basin is found to be 0.736 wt%. This agrees with characterization data provided by Welsh et al. (1996), which shows a high enrichment of 0.73 w% and an average enrichment of about 0.71 wt%. About one-third of the fuel in KE Basin is found to be enriched above 0.84 wt%.

The average ^{235}U enrichment for KW Basin fuel is found to be 0.813 wt%. Almost half of the uranium in KW Basin has an enrichment greater than 0.84 wt%.

KE Basin uranium contains an average plutonium content of about 0.188 wt%, and KW Basin uranium contains about 0.195 wt%. For plutonium in both basins the average ^{240}Pu /plutonium ratio content is found to be about 12.5 wt%.

A more current inventory of N Reactor fuel in K Basins is maintained in the Accountability Database described in Appendix A of Praga (1998). Three batch keys show a ^{240}Pu /plutonium ratio greater than 16 wt% (i.e., keys 11540, 12565, and 12852), with the highest value being 16.73 wt%. These 3 keys contain a total of 22.6 MTU, which represents about 1.1% of the approximately 2100 MTU stored in both basins. In the SCATS database only one of these keys (i.e., 11540) has a ^{240}Pu /plutonium ratio greater than 16 wt%, and that value is 16.33 wt%.

Representative examples of KE Basin and KW Basin fuel taken from the SCATS database for various levels of burnup are shown in Tables 5-3 and 5-4.

Table 5-3. Selected Fuel Keys from SCATS Database for KE Basin.

Key #	Pu kg	U kg	^{235}U kg	$^{240}\text{Pu}/\text{Pu}$ %	Pu/U %	$^{235}\text{U}/\text{U}$ %
10764	0.00	1972.00	18.67	0.00	0.000	0.947
15204	82.29	97003.40	831.21	5.89	0.085	0.857
14355	9.80	5883.98	45.20	9.41	0.166	0.768
10294	62.55	34667.16	260.43	10.19	0.180	0.751
10385	66.27	33857.17	247.86	11.03	0.196	0.732
10456	6.54	3079.31	21.94	12.14	0.212	0.712
11036	6.39	2595.92	17.42	13.76	0.246	0.671
10679	38.25	14240.30	91.12	15.02	0.269	0.640
11540	2.90	977.12	5.91	16.33	0.297	0.605
KE Basin	2154.87	1146166.	8436.48	12.57	0.188	0.736
	Total	Total	Total	Average	Average	Average

Table 5-4. Selected Fuel Keys From SCATS Database From KW Basin.

Key #	Pu kg	U kg	²³⁵ U kg	²⁴⁰ Pu/Pu %	Pu/U %	²³⁵ U/U %
10764	0.00	16.57	0.19	0.00	0.000	1.150
15445	1.52	1965.91	20.62	5.25	0.077	1.049
13648	0.04	33.11	0.33	7.17	0.117	0.989
10201	20.34	14095.26	133.38	9.02	0.144	0.946
9993	13.97	8448.59	77.14	10.12	0.165	0.913
13017	44.49	26850.17	204.00	11.01	0.166	0.760
13686	35.80	17856.03	152.85	12.03	0.200	0.856
13525	38.19	17551.51	144.96	13.02	0.218	0.826
12565	19.82	6833.33	42.03	15.93	0.290	0.615
KW Basin	1858.95	952196.	7744.65	12.47	0.195	0.813
	Total	Total	Total	Average	Average	Average

5.3 ACCOUNTING FOR BURNUP

If uranium is assumed to have the ²³⁵U content of green fuel and at the same time to contain the plutonium concentration found in highly burned fuel, K Basin sludge would be assured of being conservative. However, the quantity of absorbers required for criticality safety would be unnecessarily high. If compensation is made for burnup, a sludge model will include a smaller content of fissile material and this, in turn, will reduce the quantity of absorbers required to achieve an acceptable margin of subcriticality. This section discusses the method used to account for burnup. No attempt is made, however, to account for the contribution of fission products to neutron absorption.

5.3.1 Decrease in ²³⁵U Enrichment

Based upon characterization data (see Appendix C), the average value of ²³⁵U enrichment for sludge in KE Basin Stream 1 or Stream 2 is less than 0.68 wt%. For KW Basin sludge the average ²³⁵U enrichment for uranium in each of the three waste streams is less than 0.79 wt%. For all waste streams the average ²³⁵U enrichment is less than 0.84 wt%, the upper limit on enrichment to be permitted in DST AW-105.

Average values do not indicate the range of burnup present. The actual ^{235}U enrichment in a batch of sludge could be higher than average. When enrichment in a batch is greater than 0.84 wt%, this enrichment can be lowered by combining it with another batch known to be less enriched. In this way, batch enrichment can be adjusted without requiring addition of depleted uranium. Over one-third of K Basin uranium is enriched above 0.84 wt% ^{235}U . Consideration should be given to the need for mixing batches and, if necessary, to add depleted uranium to ensure an acceptable enrichment.

5.3.2 Plutonium Generation

Schwinkendorf (1997) calculates maximum k_{∞} values for N Reactor fuel elements in water as a function of exposure time in a reactor. For 1.25 wt% ^{235}U enrichment, k_{∞} decreases monotonically as the irradiation time increases. For 0.95 wt% ^{235}U enrichment, k_{∞} decreases for irradiation times less than 60 days, but then increases for longer times. After reaching a maximum value only slightly less than for green uranium, k_{∞} then decreases monotonically at a slow rate. The decrease in k_{∞} is greater for 1.25 wt% uranium than for 0.95 wt%. An important conclusion, nevertheless, is that maximum k_{∞} of both irradiated 0.95 wt% and 1.25 wt% enriched uranium fuel in water is always less than for the same fuel when green.

Although green KW uranium is more highly enriched, the difference between it and KE uranium decreases as the irradiation time increases. At maximum irradiation the sum of the ^{235}U and plutonium contents is 0.902 wt% for KE Basin fuel and 0.905 wt% for KW Basin fuel. For maximum irradiation the ^{235}U enrichments for KE and KW Basins are 0.605 wt% and 0.615 wt%, respectively. These enrichments are the total ^{235}U in the basin divided by total uranium (i.e., weighed average).

^{240}Pu is produced when ^{239}Pu absorbs a neutron. The production rate of ^{240}Pu is proportional to the quantity of plutonium, so the ^{240}Pu fraction increases with irradiation time. At the maximum irradiation received for any K Basin fuel the ^{240}Pu content reaches 16.33 wt% of the plutonium. The average ^{240}Pu /Plutonium ratio of 12.5 wt% shown in Tables 5-3 and 5-4 is the total ^{240}Pu in the basin divided by total plutonium (i.e., weighted average).

Batch key 11,540 in KE Basin is the only SCATS fuel with a ^{240}Pu /plutonium ratio greater than 16.0 wt%, and it comprises less than 0.1% of the total uranium. However, the Accountability Database shows 3 batch keys with 1.1% of the fuel to have a ^{240}Pu /plutonium ratio above 16.0 wt%. Nevertheless, the K Basin sludge model remains conservative for several reasons. First, 1.1% is a small proportion of the total plutonium, and in sludge it will be combined with other plutonium. Second, the sludge model maximum plutonium content of 0.30 wt% is larger than any value in SCATS, including batch key 11,540. The Accountability Database shows only a small percentage of fuel with a plutonium content greater than 0.30 wt% and the average value for all fuel is about 0.20 wt%. Third, the increase in k_{∞} , even if all fuel were assumed to have a higher plutonium content, is small (see Figure A-4). For these reasons, a 16 wt% upper limit on the ^{240}Pu /plutonium ratio for the K Basin sludge model ensures a calculated maximum k_{∞} larger than any actual k_{∞} over the range of credible compositions of real sludge.

Table 5-5 shows ^{235}U enrichment and ^{240}Pu content as a function of plutonium content, based on the SCATS database. The ^{240}Pu content in Table 5-5 is selected to be on the lower end of the range of corresponding values in SCATS. When the plutonium content is held constant, a smaller than actual ^{240}Pu content is conservative. The relationship between total plutonium and ^{240}Pu content is shown graphically in Figure 5-1. At highest irradiation when the plutonium content reaches 0.30 wt%, the ^{235}U enrichment of both KE and KW Basin fuel drops to about 0.61 wt%. This decrease in ^{235}U counteracts the increase in plutonium.

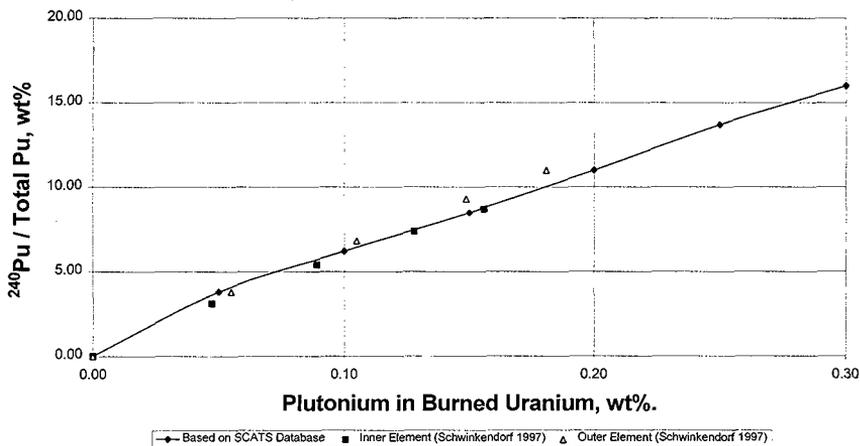
Table 5-5. ^{235}U and ^{240}Pu Contents as a Function of Plutonium Content Based on SCATS.

Pu/U wt%	Associated $^{240}\text{Pu}/\text{Pu}$, wt% ¹	Corresponding $^{235}\text{U}/\text{U}$, wt%	
		KE Basin Fuel	KW Basin Fuel
0.00	0.00	0.95	1.25
0.05	3.80	0.89	1.08
0.10	6.20	0.84	1.02
0.15	8.50	0.79	0.93
0.20	11.0	0.74	0.85
0.25	13.7	0.67	0.78
0.30	16.0	0.61	0.62

Note:

¹Both KE and KW Basin fuel inventories were examined to obtain this $^{240}\text{Pu}/\text{Pu}$ ratio.

Figure 5-1. Estimated Relationship between Total Plutonium and ^{240}Pu Content During Burnup.



Schwinkendorf (1997) calculates the radial profile of ^{240}Pu and ^{239}Pu content in N Reactor fuel as a function of irradiation. The spreadsheet used by Schwinkendorf (1997) to develop his graphs was used to calculate the average plutonium/heavy-metal mass ratios shown in Table 5-6. In reality, the plutonium concentration is a function of the radial position within the element, with the highest concentration being at the surface. Table 5-6 compiles the average plutonium/heavy-metal mass ratio and the average ^{240}Pu /plutonium mass ratio as a function of irradiation time. For the same ^{240}Pu content inner elements have a slightly higher average plutonium content, when compared to outer elements. This small difference is due to shielding by the outer element causing the inner element to see a hardened neutron spectrum. The higher average energy of neutrons results in a slightly higher plutonium production. Data points for inner and outer fuel elements are plotted on Figure 5-1 to demonstrate the agreement between Schwinkendorf (1997) calculations and the SCATS data.

Criticality safety is based upon the largest attainable value of k_{∞} . For ^{240}Pu /plutonium mass ratios up to 16 wt%, the largest k_{∞} is associated with the highest ^{240}Pu content, and this corresponds to the largest plutonium content. The relationship between maximum k_{∞} and the ^{240}Pu /plutonium mass ratio is shown in Figure A-4.

Table 5-6. ^{235}U and ^{240}Pu Contents as a Function of Plutonium Content Based on Schwinkendorf (1997)

Irradiation Days	Mark IA & IV Fuel Inner Element		Mark IA & IV Fuel Outer Element	
	Pu/HM ¹ wt%	Associated ¹ $^{240}\text{Pu}/\text{Pu}$, wt%	Pu/HM ¹ wt%	Associated ¹ $^{240}\text{Pu}/\text{Pu}$, wt%
52	0.047	3.1	0.055	3.8
102	0.089	5.4	0.105	6.8
152	0.128	7.4	0.149	9.3
192	0.156	8.7	0.181	11.0

Note:

¹Estimated average over radial profile.

Wittekind (1994b) performed calculations of decay heat from all irradiated fuel in KE and KW Basins. The production of fission products, and therefore the production of decay heat, is proportional to the ^{240}Pu generated during irradiation. The decay heat production of fuel elements was categorized according to their ^{240}Pu content. Wittekind calculated the ^{240}Pu weight percent in the plutonium as a function of exposure in units of megawatt days per metric ton of uranium (MWd/MTU). The ^{240}Pu content reaches 6.0 wt% at an exposure of 1000 MWd/MTU. Above this exposure the rate of increase slows down and remains almost linear until the ^{240}Pu content reaches 16 wt% at 4000 MWd/MTU. This is the upper limit exposure.

5.4 CONSERVATIVE MODEL OF K BASIN SLUDGE

A conservative model of K Basin sludge is constructed. Sludge volume and geometry are assumed unrestricted. The model assumes that all uranium has a ^{235}U enrichment of 0.84 wt%, a value chosen to ensure that the uranium by itself will be subcritical under all conditions of storage. This value is a compromise between being small enough not to require an overly conservative quantity of iron and at the same time large enough to permit compliance with a CPS limit. In practice this may require uranium of higher enrichments be blended with uranium of lower enrichments to ensure a blended enrichment no greater than 0.84 wt%.

The SCATS average plutonium content is between 0.18 and 0.20 wt% of the uranium, and virtually all uranium contains less than 0.27 wt% plutonium. None of the uranium has a plutonium content greater than 0.30 wt%. The average ^{240}Pu content is 12.5 wt% of the plutonium.

The upper limit plutonium content in the sludge model is conservatively set at 0.30 wt%. The ^{240}Pu content is determined by Figure 5-1, which shows the relationship between total plutonium and ^{240}Pu .

Table C-2 shows selected sludge components, based upon Pearce et al. (1998). The highest ^{235}U enrichments reported as 0.68 wt% in KE sludge and 0.79 wt% in KW sludge are consistent with the sludge model described above. However, plutonium contents provided by Pearce et al. (1998) are conservative estimates based upon the assumption that all plutonium activity is from ^{239}Pu . This assumption results in an over-estimation of the total plutonium. A more accurate estimate of plutonium content is calculated by treating each of the plutonium isotopes separately. When this is done, the values of Pearce et al. (1998) are reduced to approximately 0.65 times the reported value, and the highest estimated plutonium content for any of the waste streams is 0.27 wt% of the uranium. The plutonium contents for all waste streams therefore fall within the envelope of the K Basin sludge model.

5.4.1 Plutonium Isotopic Composition

Plutonium is composed of five isotopes: ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu . The K Basin Sludge Model was simplified by replacing ^{238}Pu , ^{241}Pu , and ^{242}Pu by an equal mass of ^{239}Pu . Justification of this replacement was provided by the relatively small proportion of these isotopes and by the large minimum critical masses of ^{238}Pu and ^{242}Pu in moderated systems. However, ^{241}Pu has a minimum critical mass of only 260 g, as compared to 520 g for ^{239}Pu , and a one-for-one substitution is not conservative. When moderation is optimized, one gram of ^{241}Pu can be equivalent to as much as two grams of ^{239}Pu . The following discussion examines the substitution used in the K Basin Sludge Model to determine if the assumed substitutions result in a final composition that is conservative.

^{238}Pu and ^{242}Pu comprise a small proportion of the plutonium. Both ^{238}Pu and ^{242}Pu require fast neutrons to fission, and neither can be made critical with thermal neutrons. For highly moderated configurations these isotopes act as neutron absorbers. Wolfe (1970) describes a series of critical experiments performed to demonstrate the poisoning effect of ^{238}Pu on the criticality of ^{239}Pu -water solutions. He shows that any addition of ^{238}Pu results in an increase in critical mass. When the ^{238}Pu content is 5 wt%, the increase in critical mass is about 25%. For water-moderated systems the effect of ^{238}Pu on criticality is almost the same as ^{240}Pu . Therefore, it is acceptable to replace the ^{238}Pu by ^{240}Pu . The effect of this substitution on k_{∞} is minimal.

Although ^{242}Pu resembles ^{240}Pu , no data is available to provide direct comparison. Replacement by an equal quantity of ^{240}Pu would be expected to provide about the same value of k_{∞} . Since no data are available to confirm this, this discussion assumes that ^{242}Pu is replaced by a void. This substitution is conservative because ^{242}Pu in a moderated system causes k_{∞} to decrease, and its removal would increase k_{∞} .

The quantities of the ^{238}Pu , ^{241}Pu , and ^{242}Pu isotopes are all directly correlated with the quantity of ^{240}Pu . When the ^{240}Pu content is low, the contents of these other isotopes are correspondingly low. According to the SCATS database, the average ^{241}Pu /plutonium content is

2.02 wt% for the KW Basin and 1.52 wt% for the KE Basin. Less than 2% of the plutonium has a ^{241}Pu content greater than 2.5 wt%, and the highest ^{241}Pu content for either basin is 2.71 wt%. In sludge the small proportion of plutonium containing a high ^{241}Pu content is mixed with a much higher quantity with a lower content, and it would be very unlikely for a significant quantity of plutonium with ^{241}Pu greater than 2.5 wt% to accumulate in a small, compact volume. It is conservative to assume that the ^{241}Pu /plutonium content will not exceed 2.5 wt%. This ^{241}Pu content is equivalent to 5.0 wt% ^{239}Pu .

When the ^{240}Pu content is 16.0 wt%, the ^{238}Pu and ^{242}Pu contents are about 0.17 and 0.50 wt%, respectively, and this should be approximately equivalent to an equal quantity of ^{240}Pu , or 0.67 wt%. However, to ensure conservatism the ^{242}Pu is removed without replacement, and the resultant conservative equivalency is 0.17 wt% of ^{240}Pu .

A conservative isotopic composition for plutonium is 80.83 wt% ^{239}Pu , 16.00 wt% ^{240}Pu , 2.50 wt% ^{241}Pu , 0.50 wt% ^{242}Pu , and 0.17 wt% ^{238}Pu . A 100 g batch of this plutonium would contain 2.50 g of ^{241}Pu and 0.67 g total of ^{238}Pu and ^{242}Pu . Replacing this with 5.0 g of ^{239}Pu and 0.17 g of ^{240}Pu results in the equivalent composition of 85.83 g of ^{239}Pu and 16.17 g of ^{240}Pu . In other words, 100 g of high burnup plutonium can be replaced by 102.0 g of plutonium containing 84.15 wt% ^{239}Pu and 15.85 wt% ^{240}Pu . When the ^{240}Pu content is 16 wt%, a 1.0 % change in ^{240}Pu results in a 5.0 % change in critical mass (see Table F-1). If the ^{240}Pu content is reduced by 0.40 wt%, the critical mass will decrease by 2.00 wt%. This means that 100.0 g of plutonium comprised of 84.55 wt% ^{239}Pu and 15.45 wt% ^{240}Pu is equivalent to 100 g of maximum burnup plutonium as described above.

In conclusion, maximum burnup plutonium is conservatively equivalent to plutonium comprised of 84.55 wt% ^{239}Pu and 15.45 wt% ^{240}Pu . This composition compensates for the generation of ^{241}Pu in the irradiation process and does not account for the neutron absorption by ^{242}Pu . The K Basin Sludge Model assumed a composition of 84.0 wt% ^{239}Pu and 16.0 wt% ^{240}Pu for maximum burnup. The sludge model is therefore slightly non-conservative when compared to the composition model assumed in this discussion. The effect of this slight change in composition is examined in the next section.

5.4.2 k_{∞} for Limiting Sludge Composition

A parametric study of the K Basin sludge model is provided in Appendix A. For these calculations ^{238}Pu , ^{241}Pu , and ^{242}Pu were replaced by an equal mass of ^{239}Pu . Maximum k_{∞} for the sludge model occurs when the plutonium content is 0.30 wt% and the ^{240}Pu content is 16 wt%. Calculational results shown in Table 5-7 demonstrate that compensating for the plutonium isotopic composition by further reducing the ^{240}Pu /plutonium ratio is conservative. For these calculations the ^{240}Pu /plutonium ratio was set at 15.7 wt% and to 15.45 wt%. This small difference in ^{240}Pu content results in only a slight difference in k_{∞} . Only compositions of greatest interest were selected for these calculations. All configurations were selected with a water content that provides maximum k_{∞} . Uranium is assumed enriched to 0.84 wt% ^{235}U and to have a plutonium content of 0.30 wt%.

The reason for performing calculations in Table 5-7 is to determine how much iron is required to ensure that the USL is not exceeded after assuming that gravity segregation increases the concentration of plutonium by a factor of 3. In addition, calculations were performed with a constant plutonium concentration for varying contents of iron. Decreasing iron concentration by a factor of 3 results in a higher final reactivity than increasing the plutonium concentration by a factor of 3. When the ^{240}Pu /plutonium ratio is 15.45 wt%, an iron/heavy-metal mass ratio of 0.256 results in a maximum k_{∞} of 0.8708, a value slightly smaller than the USL of 0.881. When this iron is multiplied by 3 to give an iron/uranium mass ratio of 0.768, the USL is satisfied, after taking into account gravity segregating and the plutonium isotopic composition. Maximum k_{∞} is then 0.6658, and under normal conditions this is the highest value of k_{∞} that can be achieved.

Before the subcritical limit is reached, it is necessary to reduce the iron/uranium ratio by an additional factor of between 2.4 and 3. For criticality to be possible the iron/uranium mass ratio would have to drop below 0.100.

Table 5-7. Calculations Showing Safety Margin and Compliance to Upper Safety Limit

Case Identifier	$^{235}\text{U}/\text{U}$ wt%	Pu/U wt%	$^{240}\text{Pu}/\text{Pu}$ wt%	Fe/HM Mass Ratio	XSDRNPM Maximum k_{∞}
Changes in Iron Content					
F17E	0.84	0.30	15.45	0.768	0.6658
F15E	0.84	0.30	15.7	0.768	0.6652
F15A	0.84	0.30	15.7	0.710	0.6820
F15D	0.84	0.30	15.45	0.256	0.8716
F15D	0.84	0.30	15.7	0.256	0.8708
F15C	0.84	0.30	15.7	0.242	0.8780
F15B	0.84	0.30	15.7	0.238	0.8803
F15G	0.84	0.30	15.7	0.200	0.9017
F15K	0.84	0.30	15.7	0.175	0.9165
F15H	0.84	0.30	15.7	0.150	0.9321
F15J	0.84	0.30	15.7	0.100	0.9647
Change in Plutonium Content					
F15E	0.84	0.30	15.7	0.768	0.6652
F16A	0.84	0.90	15.7	0.768	0.8501
F16B	0.84	1.20	15.7	0.768	0.9125
F16F	0.84	1.50	15.45	0.768	0.9649
F26C	0.84	1.50	15.7	0.768	0.9633
F16D	0.84	1.80	15.7	0.768	1.0053

Calculations are made over a wide range of iron and plutonium content to show only the margin to ensure compliance to the USL and also the margin available to preclude criticality.

Calculations are made to determine how much iron is required to remain within the USL after gravity segregation increases the concentration of plutonium by a factor of 3. Between the USL and criticality there is an additional factor of 2 safety margin in the quantity of iron.

Calculations based upon the conservative sludge model described above form the bases for limits and controls to ensure subcriticality for operations in tank farms. The iron content is an independent variable with a limit value selected to ensure that maximum k_{∞} will not exceed the limit criterion. When the ^{240}Pu /plutonium ratio is 15.45 wt% and the iron/heavy-metal mass ratio is 0.256; maximum k_{∞} is slightly smaller than the USL of 0.881. An iron/uranium mass ratio of 0.0768 is found to be sufficient to satisfy the USL after taking into account the possibility of segregating by a factor of 3. This takes into account the isotopic composition of the plutonium.

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6.0 NEUTRON ABSORBERS

Criticality safety requires that no chemical processes capable of making significant changes to the relative concentrations of uranium, plutonium, and neutron absorbers are operating. In an acidic solution, the variety and complexity of active chemical processes would be much greater than when alkaline. Normally, to prevent corrosion of the steel tank walls and interconnecting piping, the pH is maintained near 12. Criticality safety requires that pH not fall below a minimum value of 8. A discussion of chemistry can be found in Appendix E.

Bratzel et al. (1996) studied physical and chemical phenomena and mechanisms that might concentration fissile material and concluded there are none capable of resulting in an accidental nuclear criticality for tank waste. Although the origin of K Basin sludge is different from waste already in tank storage, it will be processed into a form that more closely resembles tank waste.

Particle size is a primary determiner of the degree by which gravity segregation is possible. Complete assurance that component separation will not occur requires a particle size smaller than 10 μm or a detailed knowledge of waste chemistry. This evaluation assumes that particles are small enough that the increase in plutonium concentration as result of gravity segregation will not exceed a factor of 3.

Neutron absorber materials will be added to K Basin sludge to ensure an adequate margin of subcriticality. Subcriticality can be assured provided: 1) the solids are sufficient to provide adequate neutron absorption; 2) the solids and sludge are uniformly mixed; and 3) there are no processes capable of separating the solids from the uranium. Although there are many solid materials that might be used, depleted uranium and iron are considered best choices. This section provides a basis for determining the quantities of depleted uranium and iron required to meet the criterion for subcriticality.

6.1 DEPLETED URANIUM

Depleted uranium is the neutron absorber of choice to be blended with enriched uranium. The result is uranium of an intermediate enrichment. There is no chemical or physical processes in the waste storage environment capable of separating ^{238}U from ^{235}U , and the blended enrichment cannot be increased by processes in the sludge. It is important that depleted uranium be well blended with the enriched uranium to ensure a uniform enrichment.

The plutonium content in depleted uranium is a concern. This plutonium counteracts, at least in part, the reduction in k_{∞} from adding depleted uranium. Additional iron would be required to maintain the margin of subcriticality for the plutonium. So as not to increase the required iron, a limit of 0.05 wt% is placed on the plutonium content in depleted uranium to be added.

The *Nuclear Criticality Safety Guide* (Pruvost and Paxton 1996) provides a subcritical limit on ^{235}U enrichment for uranium mixed homogeneously with water as 0.93 wt% for uranium metal and 0.96 wt% for UO_2 , UO_3 , U_3O_8 or UO_3F_2 .

Clayton and Brown (1965) provide a minimum critical ^{235}U enrichment for UO_3 in a water solution of 1.030 ± 0.010 wt%. Extrapolation shows that k_{∞} will not exceed 0.95 when the ^{235}U enrichment is less than 0.88 wt%. Figure A-2 shows k_{∞} for homogeneous UO_2 as a function of water content. When the ^{235}U enrichment is 0.84 wt%, the maximum k_{∞} that can be achieved is 0.923. After taking into account calculational bias, this k_{∞} is less than 0.95 at a 95% confidence level.

For K Basin uranium the plutonium content might also be as high as 0.30 wt%. When this plutonium is taken into account, the maximum k_{∞} might be as high as 1.04, and neutron absorbers are also required to counteract the effect of the plutonium.

In practice the ^{235}U enrichment in K Basin uranium is likely to be less than 0.84 wt%. If this can be shown, then there is no need to add depleted uranium to lower the ^{235}U enrichment. In cases where the ^{235}U enrichment is greater than 0.84 wt%, it may be possible to combine this sludge with sludge from another location for which the uranium is known to have a lower enrichment. By mixing the more highly enriched uranium with the lower enriched uranium the need to add depleted uranium can be avoided.

The quantity of depleted uranium to be added to a quantity of enriched uranium to lower the combined enrichment to 0.84 wt% is determined from the following formula, where E(enriched) and E(depleted) are the ^{235}U enrichments of enriched and depleted uranium.

$$\text{U(depleted)/U(enriched)} = [\text{E(enriched)} - 0.84] / [0.84 - \text{E(depleted)}]$$

Table 6-1 shows the mass ratio of depleted uranium to be blended with 0.95 wt% and 1.25 wt% enriched uranium to reduce the enrichment to 0.84 wt%. The U(depleted)/U(enriched) mass ratio for any combination of depleted and enriched uranium can be determined using the formula.

Table 6-1. Depleted Uranium Required to Reduce Uranium Enrichment to 0.84 wt%.

^{235}U Content in Depleted Uranium (wt% ^{235}U)	Mass Ratio to Reduce 0.95 wt% to 0.84 wt%	Mass Ratio to Reduce 1.25 wt% to 0.84 wt%
0.20	0.145	0.540
0.30	0.203	0.756
0.40	0.250	0.932
0.50	0.323	1.20
0.60	0.458	1.71
0.72 (natural)	0.916	3.41

If a sizeable quantity of depleted uranium with no contained plutonium is added, the ^{235}U enrichment can be decreased below 0.84 wt% and the plutonium/uranium ratio would be reduced. This would also decrease the quantity of iron required to maintain an acceptable margin of safety.

6.2 IRON

Plutonium and uranium are different elements with different chemical properties and can be chemically separated from each other. If separation is precluded by avoiding environments (i.e., acids and complexing agents) which dissolve the absorber or leach the plutonium, either depleted uranium or iron can be used as a neutron absorber. However, a higher assurance of a continued margin of subcriticality is provided by addition of a neutron absorber that preferentially remains associated with plutonium in the tank environment. Iron will coprecipitate with both uranium and plutonium and is such an absorber.

The K Basin sludge model (see Section 5.0) is the basis for calculation of k_{∞} as a function of iron content. Figure A-7 shows the quantity of iron required to ensure a specified maximum k_{∞} when the plutonium content is 0.20 wt% (average burnup) and 0.30 wt% (maximum burnup). The quantity of iron required for 0.30 wt% plutonium content is the larger value, but the difference is not large.

The USL is the highest value of the calculated k_{∞} permitted under all normal and credible abnormal conditions of tank waste operations. Rogers and Niemer (1999) support an USL on k_{∞} of 0.881. The iron reduction factor (IRF) is defined as the minimum permitted iron concentration divided by the iron concentration that results in a maximum k_{∞} equal to the USL. The plutonium concentration factor (PCF) is defined as the plutonium concentration which results in a maximum k_{∞} equal to the USL divided by the maximum permitted plutonium concentration. The PCF is equal to the plutonium/uranium mass ratio at the USL divided by the maximum permitted plutonium/uranium mass ratio.

When the iron/uranium mass ratio is 0.256, maximum k_{∞} for the K Basin sludge model is found to be 0.8708, a value slightly less than the USL. Assuming an IRF of 3.0, the acceptable iron/uranium mass ratio becomes 0.768. Holding this iron content constant and increasing the plutonium/uranium mass ratio by a factor of 3 to 0.90 wt%, maximum k_{∞} increases to 0.8501, a value below the USL. For this iron content the PCF is 3.3. Details on these calculations are provided in Section 5.4.

In conclusion, an iron/uranium mass ratio of 0.768 ensures conformance to the USL after assuming that a gravity segregation factor of 3 is possible. When uniformly mixed, maximum k_{∞} will not exceed 0.67 for all degrees of burnup.

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7.0 COMBINING K BASIN AND TANK AW-105 SLUDGE

Scenarios associated with combining K Basin sludge with tank waste within DST AW-105 are evaluated as to whether interaction between sludge types and intermixing are capable of increasing k_{∞} above the USL.

7.1 INTERACTION WITH CONTENTS OF DST AW-105

Whyatt et al. (1996) concludes that plutonium in tank waste primarily resides in the solid phase. The solubility of plutonium in supernatant liquid is low enough that saturation concentrations are at least 30 times lower than concentrations needed to support criticality. For this reason, criticality in the supernatant liquid is not credible.

Processing of K Basin sludge provides high assurance that the composition will fall inside the composition envelope of the sludge model. However, the possibility is considered that pockets of high fissile concentration might exist in DST AW-105 sludge. Available evidence strongly indicates that regions of higher fissile concentration, should they exist, are confined to thin slab layers. Knowledge of waste generation processes supports the conclusion that there are no significant volumes having a plutonium concentration much above the tank average.

CPS limits are based on the assumption that an unlimited volume of waste is possible. Since optimal water moderation is assumed, maximum k_{eff} for any finite volume, even with full reflection, will be less than the maximum k_{∞} for an infinite volume.

There is a high degree of uncertainty associated with the composition of DST AW-105 sludge, and this makes it difficult to determine an accurate upper limit on k_{∞} . However, available information supports the conclusion that it is lower than that for K Basin sludge. k_{eff} for the combined sludge is determined primarily by the sludge type with the larger k_{∞} , and for sludges under consideration this is K Basin sludge. Any combination and geometric arrangement of these sludges should not be capable of a k_{eff} that exceeds the largest value possible for K Basin sludge alone.

Chemical compatibility of sludges is addressed only in so far as it concerns criticality safety. Chemistry documented by Serne et al. (1996), by Whyatt et al. (1996), and by Daling et al. (1997) applies before and after the transfer of K Basin sludge.

An estimated 68.8 vol% (729,550 L) of DST AW-105 sludge is from the PUREX zircalloy decladding waste stream. Subcritical fractions reported by Whyatt et al., based on process records, are shown in Appendix D4.2. The sum of actual-to-minimum subcritical fractions for iron and zirconium is found to be 8.4 from process records, as compared to 2.1 from an analysis of core samples.

Agnew (1995) calculated the sum of the absorber actual-to-minimum subcritical fractions to be 18.6 for insoluble components and 54.4 for soluble components. Braun et al. (1994)

estimates the solids/plutonium mass ratio to be 50,000, and the sum of the actual-to-minimum subcritical fractions to be at least 64. This value, which includes both soluble and insoluble components, is in good agreement with Agnew's value. The smallest sum of subcritical fractions reported by Braun et al. for soluble components was 15. For this same sample the sum of insoluble subcritical fractions was reported as 0.50. This low content of insoluble solids is thought to reflect an incomplete analysis of the waste sample. Nevertheless, the total quantity of neutron absorbers reported for soluble and insoluble components for this sample is considerably higher than required to ensure subcriticality.

The largest plutonium concentration reported by Braun et al. was 0.024 g/L, a value 20 times higher than indicated by process records. For this evaluation, the plutonium concentration is assumed to be 0.024 g/L for all sludge in DST AW-105. Based on this conservative plutonium concentration, the plutonium areal density is 55.9 g/m², a value 46 times smaller than the minimum required for criticality.

Both DST AW-105 and K Basin sludge have large ratios of absorbers, both soluble and insoluble, in relationship to fissile isotopes. When the sludges are mixed, the combined sludge will also contain a large ratio of absorbers to fissile isotopes. The characteristics of the combined sludge and the associated chemistry should be very much the same as that of each of the individual sludge types. Mixing the sludge will not reduce the margin of subcriticality.

7.1.1 Stratification and Interaction

According to Daling et al. (1997), the top 10-in. layer of sludge in DST AW-105 contains an estimated 110 g of plutonium at an average concentration less than 0.002 g/L. However, the 6-in. layer directly below this contains almost 30% of the "plutonium equivalent" inventory of the tank, or about 7 kg. Much of this "plutonium equivalent" inventory is probably comprised of ²³⁵U in enriched uranium. Only the portion of ²³⁵U greater than the content in natural uranium, or 0.72 wt%, is included. The highest estimated plutonium concentration is 0.31 g/L in a thin layer.

If spread uniformly over the tank, the areal density in this layer would be 17.0 g/m² (1.58 g/ft²), a value 152 times smaller than the minimum critical plutonium areal density of 2,582 g/m² (240 g/ft²). The mass ratio of natural uranium to plutonium in this layer exceeds 1,000, and the atom ratio of cadmium to plutonium exceeds 2. Either the natural uranium or the cadmium alone is sufficient to ensure subcriticality, if homogeneously combined with the plutonium.

7.2 MECHANISMS WHICH INCREASE CONCENTRATION

K Basin sludge is well characterized when received into DST AW-105. However, the distribution of components already present is less well defined. Under conditions defined by

historic discharge limits and by characterization data the neutronic interaction between K Basin sludge and DST AW-105 sludge is negligible. The possibility of a sizeable pocket of high plutonium concentration is extremely unlikely (Braun et al. 1994).

If neutron absorption is assumed to be at least as great as in the conservative waste model (CWM), the fissile concentration must exceed 2.6 g/L before criticality is possible (Rogers 1996). However, this limit does not apply to K Basin sludge, which falls outside of the description of the CWM. Nevertheless, it provides an estimate of the minimum fissile concentration for the hypothetical condition in which plutonium is separated from K Basin sludge and mixed with DST AW-105 sludge. For K Basin sludge to achieve criticality it would be necessary for the plutonium to be separated from the iron, to be deposited in a limited volume, and to be optimally moderated. Mechanisms capable of separation are discussed in the following subsections.

7.2.1 Settling of Suspended Fissile Material

The most obvious, and perhaps most effective, mechanism for concentrating uranium and/or plutonium is settling. If particles of fissile material settle faster than other waste particles, this would lead to formation of layers containing elevated uranium concentrations. The volume fraction occupied by neutron absorbing solids and the size distribution of waste particles determine the degree by which the concentration of fissile material can be increased upon settling through a liquid layer.

The fissile areal density is the total mass of fissile material above a unit of floor area. Criticality is not possible unless the plutonium equivalent areal density exceeds 240 g/ft^2 ($2,582 \text{ g/m}^2$). This areal density over the area of a storage tank would require more than 1,000 kg of plutonium. In addition, for criticality to actually occur the concentration must be greater than the minimum required for criticality. Based on the conservative waste model, the minimum critical concentration for plutonium in water is 2.6 g/L.

Based on the K Basin sludge model, the iron/uranium mass ratio must fall below 0.256 before it is possible for k_{∞} to exceed the USL. Before the USL can be actually exceeded, this low ratio would have to apply to a large mass of sludge.

7.2.2 Removing Liquid

Liquid is removed from waste through evaporation and pumping. As liquid is removed solids consolidate, and their density increases. However, the solids/plutonium and solids/uranium mass ratios remain unchanged. When the sum of mass ratios exceeds the minimum subcritical value, subcriticality is assured, no matter how dry the waste. Because of the low fissile concentration, sludge has sufficient neutron absorbers to remain well subcritical for all degrees of moderation. Criticality safety for K Basin sludge does not require a limit on water content.

7.2.3 Chemical Separation of Components

Chemical processes capable of increasing the uranium and plutonium concentrations can be postulated. These processes must dissolve the uranium (or plutonium) and then precipitate, or be otherwise deposited, preferentially at a different location. For dissolution to occur the chemical environment of the tank must change from its normal condition of high alkalinity to high acidity. This would require introduction of a large volume of acid in violation of operating requirements. This would then have to be followed by the reverse condition of alkalinity. These changes can only be possible when wastes of considerably different compositions are mixed. This would require a major deviation from normal operating procedures and multiple violations of applicable controls.

If chemicals were to dissolve neutron-absorbing components, natural controls prevent criticality. First, even if all absorbers were removed, the low plutonium areal density would preclude criticality. Second, the variety of chemicals in waste mitigates removal of a specific absorber. In addition there would be a tendency for components removed to remix with the waste.

7.2.4 Mixing

When mixed, a component concentration tends to become closer to an average value. As mixing continues, the overall mixture becomes increasingly uniform. When particles of varying size and density are mixed and allowed to settle, however, they may form layers of similar particles. The concentration of a component might be increased in one layer and decreased in another. Serne et al. (1996) describes particle segregation for natural geologic processes and ore beneficiation processes, and they discuss tank waste operations that might cause particle segregation, such as sluicing, mixing, and salt well pumping. None of these can be shown to cause criticality.

7.2.5 Conditions that Affect Plutonium Accumulation

Several conditions tend to prevent accumulation of plutonium in a small, compact volume. First, alkaline conditions limit the mass of plutonium and uranium in solution (dissolution). Second, the bulk of the plutonium and uranium would be present as precipitates amongst a large mass of metal hydrous oxide particulate neutron absorbers. During any mechanical disturbance these materials are capable of mixing with the fissile components and would not likely separate from them. Third, following a brief periodic transfer of new waste there are no physical forces disturbing the sludge until the next transfer. This provides a long period of quiescence that allows all solids from one transfer to settle before the next waste is received. Transfer of small volumes of waste slurry into the tank does not cause major mixing of existing sludge, especially sludge lying near the bottom under the weight of overlying material. Fourth, waste components agglomerate into tightly bound masses that are not easily broken apart or dissolved.

8.0 CONTINGENCIES

Contingencies that apply to disposing of K Basin sludge are listed in Table 8-1.

Table 8-1. Contingencies and Barriers.

Contingency		Barriers
No.	Description	
1	^{235}U enrichment	No uranium enriched above 1.25 wt%. Primarily high burnup uranium. Detailed inventory records. Chemical and isotopic analysis of samples.
2	Plutonium content	No other source of plutonium. Highest burnup assumed in evaluation. Chemical analysis of sludge samples.
3	^{240}Pu content	Burnup history
4	Iron content	Controls on iron addition. Chemical analysis of sludge samples.
5	Homogeneity	Individual batches mixed. Chemical analysis of sludge samples. Highly fluid sludge spreads into thin layer.
6	Particle size	Procedures for limiting particle size (either by chemical means or by grinding).
7	Alkalinity (pH)	Requirements for corrosion control. Chemical analysis of sludge samples.
8	Neutronic interaction with DST AW-105 waste	High solids/plutonium mass ratio. Tank characterization data. Waste generation history.

8.1 URANIUM-235 ENRICHMENT

Section A7.1 describes the sensitivity of k_{∞} to changes in ^{235}U enrichment when the iron/uranium (virtually the same as iron/heavy-metal) mass ratio is held constant (see Figures A-10 and A-11). Consider a configuration having an iron/uranium mass ratio of 0.53. When the ^{235}U enrichment is 0.84 wt% and the plutonium content is 0.30 wt%, maximum k_{∞} is 0.745. When plutonium content remains unchanged, an increase in ^{235}U enrichment to 0.95 wt% increases k_{∞} to 0.776, and an increase to 1.00 wt% increases k_{∞} to 0.789. This means that a change of 0.10 wt% in ^{235}U enrichment results in a change in k_{∞} of 0.028, when the iron/uranium mass ratio is 0.53. Extrapolating to 1.25 wt% ^{235}U , the highest enrichment available, k_{∞} increases to 0.86.

If the iron/uranium mass ratio is increased to 0.70, maximum k_{∞} for 0.84 wt% ^{235}U and 0.30 wt% plutonium content decreases to 0.684. For 1.25 wt% ^{235}U maximum k_{∞} would be 0.80. Therefore, if a large quantity of green (unirradiated) uranium were hypothetically combined with the sludge, the high iron content would maintain the sludge well subcritical.

In reality, green uranium contains no plutonium, and maximum k_{∞} after addition of green uranium would be less than described above. As shown in Figure A-4, 0.84 wt% enriched uranium with 0.30 wt% plutonium has a maximum k_{∞} of 1.034, while removal of the plutonium would reduce maximum k_{∞} to 0.923. Assuming removal of plutonium would change k_{∞} by the same ratio when the iron/uranium mass ratio is 0.70, maximum k_{∞} for green 1.25 wt% ^{235}U (without plutonium) would be 0.80 multiplied by 0.923/1.034, or about 0.71.

When the iron/uranium mass ratio is 0.70, replacement of irradiated uranium with green 1.25 wt% ^{235}U enriched uranium would cause the maximum k_{∞} to increase from 0.68 to 0.71. In other words, an inadvertent substitution of green uranium will not increase maximum k_{∞} by more than 0.03.

It is highly unlikely that the ^{235}U enrichment will exceed 0.84 wt%. Chemical analysis will be made of sludge samples from each batch received at tank farms, and this information will be used to verify compliance with the CPS limits.

In conclusion, the probability of violating the limit on ^{235}U enrichment is low. At the same time, a failure to meet the limit of 0.84 wt% (maximum) ^{235}U enrichment cannot increase maximum k_{∞} by more than 0.03, assuming the iron/uranium mass ratio is larger than 0.70. k_{∞} would remain well below the USL for any possible error in uranium characterization.

8.2 PLUTONIUM CONTENT

Figure A-15 shows maximum k_{∞} as a function of plutonium content when the ^{235}U enrichment is 0.84 wt% and the ^{240}Pu /plutonium ratio is 16 wt%. When the iron/uranium mass ratio is 0.768, a plutonium content of 0.90 wt%, 3 times the upper limit, results in a

maximum k of 0.850. This means that the PCF must exceed 3.0 to reach the USL. Violation of the USL requires that separated plutonium from an outside source be added, but no outside sources of plutonium are available.

To increase plutonium relative to uranium requires that plutonium be chemically separated and preferentially moved to a new location where it is concentrated. The presence of a large ratio of iron makes plutonium transport unlikely, even if separation is postulated.

A limit of 0.05 wt% is placed on the plutonium content in depleted uranium added to the sludge from an outside source. This prevents addition of significant quantities of plutonium. It seems likely that no depleted uranium will be added to this sludge. If depleted uranium is added, the total quantity is expected to be small.

A chemical analysis will be made of samples taken from each batch of sludge received at tank farms, and this information will be used to verify that the plutonium/uranium mass ratio does not exceed 0.30 wt%.

In conclusion, violation of the limit on initial plutonium content is not possible under credible conditions. It is extremely unlikely, if not impossible, that chemical or physical processes within the tank would be capable of increasing the plutonium concentration by the factor of at least 3.0 required to exceed the USL.

8.3 PLUTONIUM-240 CONTENT

The effect of a change in ^{240}Pu content on maximum k_{∞} depends on whether the ^{240}Pu content is independent or dependent of the plutonium content. Variation in ^{240}Pu content when total plutonium is held constant at 0.30 wt% is examined in Section A.7.2. In this case, an increase in ^{240}Pu is accompanied by a decrease in ^{239}Pu . With a ^{235}U enrichment of 0.84 wt% and an iron/uranium mass ratio of 0.53, an increase in ^{240}Pu content from 0 to 16 wt% results in a decrease in maximum k_{∞} from 0.795 to 0.745 (see Figure A-13). In other words, when the iron/uranium mass ratio is 0.53, a 1.0-wt% overestimation of ^{240}Pu results in an underestimation in k_{∞} of 0.003. For higher iron contents the change in k_{∞} would be smaller.

When ^{239}Pu and ^{240}Pu are related according to burnup, the concentrations of these isotopes increase together (see Figure 5-1). When the ^{240}Pu /plutonium ratio is 0 wt%, the plutonium/uranium ratio is also 0 wt%. When the ^{240}Pu /plutonium ratio is 16 wt%, the plutonium content is 0.30 wt%. For a constant 0.84 wt% ^{235}U enrichment, k_{∞} increases with the ^{240}Pu content because the plutonium content is also increasing. This relationship is shown in Figure A-4 for a ^{240}Pu /plutonium ratio between 0 and 16 wt% when no iron is present. When the total plutonium increases from 0 to 0.30 wt%, k_{∞} increases from 0.923 to 1.034, and the ^{240}Pu /plutonium ratio increases from 0 to 16 wt%.

A more realistic configuration for K Basin sludge would have an iron/uranium mass ratio of 0.70 and a ^{240}Pu content between 11 and 16 wt%. With this iron content maximum k_{∞} increases from 0.650 to 0.684 when the ^{240}Pu /plutonium ratio increases from 11 to 16 wt%.

CPS limits are based on the ^{240}Pu and total plutonium contents being related through burnup and the ^{240}Pu /plutonium ratio being 16 wt%. This relationship is not subject to change by processing or by waste handling operations at tank farms.

In conclusion, no changes can be made to K Basin sludge that would be capable of resulting in a ^{240}Pu content outside of the K Basin sludge model. Uncertainties in analytical measurement of ^{240}Pu content will not call into question conformance to the USL, provided the iron/uranium mass ratio is larger than its limit.

8.4 IRON CONTENT

The parameter of greatest importance to ensuring an adequate margin of subcriticality is the iron/uranium mass ratio. Iron absorbs neutrons and also has an affinity for plutonium.

Sludge components are expected to form agglomerates, and iron is a major component of this process. When components agglomerate, they form into a tightly-bound mass in which uranium, plutonium, iron, and other waste components are held in a fixed relationship. Agglomeration is a mechanism that inhibits an increase in fissile concentration. No credit is taken in this evaluation for the ability of agglomeration to inhibit separation of components.

Studies of gravity separation (see Appendix F4.0) show that a small particle size greatly reduces, or even prevents, separation of components according to size and density. If sludge is assumed composed of small individual spheres of pure components that exhibit no mutual attraction or cohesion, gravity separation into layers of varying component concentrations is possible. To preclude significant gravity separation of waste components into layers following agitation and settling, a limit of 10 μm is placed on particle size. Serne et al. (1996), in a study of tank waste chemistry, concludes that this small size ensures an upper limit of a factor of 2.5 on concentration increase. Daling et al. (1997) discusses gravity separation for K Basin sludge and concludes that a concentration factor of 3 applies.

When the iron/uranium mass ratio is less than 1.0 (i. e., k_{∞} greater than about 0.60), maximum k_{∞} decreases in almost direct proportion to increases in the quantity of iron (see Figure A-7). Above an iron/uranium mass ratio of 0.60, the effectiveness of additional iron towards further reducing k_{∞} diminishes.

To exceed the USL the iron/uranium mass ratio must drop below 0.256. Three times this quantity of iron, or an iron/uranium mass ratio of 0.768, will compensate for credible gravity segregation and ensure compliance to the USL. Since the K Basin sludge model envelopes credible sludge compositions, no additional iron is required to compensate for uncertainties in the model.

The USL was deliberately set low to conservatively compensate for uncertainties in the validation of iron cross sections. The USL of 0.881 is based on a maximum k_{∞} of 0.90 at a 95% confidence level. If justification were to be provided to permit an USL of 0.95 at a 95% confidence level, the requirement for iron could be reduced further.

In conclusion, an iron/uranium mass ratio of 0.768 provides an adequate margin of subcriticality to protect against gravity separation, uncertainties in characterization, and uncertainties in the calculational method. This quantity of iron ensures a k_{∞} no greater than 0.67 when mixed homogeneously with the uranium and plutonium.

8.5 HOMOGENEITY

A requirement that sludge be mixed is not the same as a requirement that it be uniform. Complete mixing of sludge would be very difficult to achieve and also very difficult to verify. The sludge need not be uniform throughout, but it is necessary that the iron/uranium mass ratio within any small spherical subvolume be no less than the required limit. For tank waste a volume of 100 L is considered small. Regions with more iron than required are acceptable. In other words, sludge in DST AW-105 can be formed into layers of different compositions so long as the iron/uranium mass ratio in each layer is greater than the CPS limit value.

Before processing the iron/uranium mass ratio in K Basin sludge is less than required for criticality safety in DST AW-105. After iron is added, the sludge will be mixed to reduce spatial variation in composition, thereby ensuring that the iron/uranium mass ratio in any subvolume will exceed the minimum required ratio. Since it is not practical to mix the entire volume of sludge as a single unit, individual transport container loads must be mixed before discharge into DST AW-105.

Before K Basin sludge is received at tank farms, analytic verification is to be made of its composition. Analyses of samples sent to an analytical laboratory must be completed before sludge arrives at tank farms. If the iron/uranium mass ratios in analyzed samples exceed the CPS limit, mixing is judged acceptable.

The highly fluid sludge will spread into a thin layer inside of DST AW-105. Variations in composition will translate into variations within thin layers. The process of layer formation will reduce the significance of differences in batches. For this evaluation the volume of sludge is assumed to be great enough to fill the entire tank. The finite volume of the sludge and the slab geometry into which it forms will create a less reactive configuration than that assumed in this evaluation. For these reasons the real margin of safety will be greater than stated in this evaluation.

8.6 PARTICLE SIZE

The ability of sludge to segregate into layers of different composition is dependent upon the relative size, density, and composition of particles. When sludge particle sizes are smaller than 10 μm , the segregation factor will be no greater than 3.0. At this small size differences in density are no longer important, and the settling rate is entirely a function of particle size. When particle size exceeds 50 μm , the segregation factor increases. This evaluation assumes that a particle size criterion of 10 μm is met for primary particles. The size of secondary particles made up of agglomerated primary particles is not important to criticality safety. The composition of an agglomerate will be similar to the average composition of the sludge.

The impact of not meeting the particle size criterion is difficult to assess. Even at sizes much larger than 10 μm , there are physical and chemical forces at work that act against segregation of sludge components. In reality the particles for each sludge component conform to a size distribution. If the distribution of sizes for different components are the same, gravity segregation will not occur, even if the average particle size is much larger than 10 μm . However, no studies have demonstrated that gravity segregation will not occur for particles larger than 10 μm , and the sludge will be processed to ensure a particle size of less than 10 μm .

Adherence to the particle size criterion will be demonstrated for the process used to achieve size reduction. Analytical verification will be made before shipment of sludge to tank farms.

8.7 ALKALINITY

A pH between 0 and 7 indicates acidity and between 7 and 14 indicates alkalinity. Tank waste is required to be alkaline with a pH of at least 8.0 (minimum).

pH is an indicator of the degree to which material contained in the waste is insoluble. A high pH guarantees a low solubility of most waste components, including uranium, plutonium, and iron. Dissolution of sludge by itself would not cause criticality, but it increases the complexity of chemical processes and creates the possibility of removal of the plutonium from uranium. A low pH makes waste components more mobile.

The parameter of primary importance to criticality safety is the mass ratio of solids (especially iron) to plutonium. To criticality safety, the importance of maintaining a high pH is to prevent dissolution of solids, including plutonium, thereby ensuring that most plutonium will be combined with solids settled at the bottom of the tank.

Even postulating a very high inventory of plutonium, such as 200 kg, the areal density will be well below that required for criticality. This remains true even if all neutron-absorbing solids are assumed removed. It is extremely unlikely that a plutonium concentration high enough over a large enough volume would be achieved to result in a k_{∞} even approaching the USL achieved, regardless of the pH.

Procedures are in place to ensure that waste in tank storage is alkaline. Measurement of the pH of K Basin sludge will be made prior to discharge into DST AW-105.

8.8 NEUTRONIC INTERACTION WITH DST AW-105 WASTE

When discharged into DST AW-105, K Basin sludge will form a layer on top of sludge already there. Criticality safety requires assurance that interaction between these waste types cannot result in a k_{∞} which might exceed the USL. A detailed description of DST AW-105 waste is provided in Appendix D.

Knowledge of waste generation processes supports the view that there are no significant volumes in DST AW-105 having a plutonium concentration much above the tank average. Regions of higher fissile concentration are confined to thin slab layers. The Double-Shell Tank Plutonium Inventory Database (LMHC 1998) shows the April 1999 inventory to be 22.95 kg. The total volume of waste is 430.5 kgal, of which 280 kgal is solids. The plutonium concentration in supernatant liquid is 3.22E-06 g/L and in the solids is 0.0217 g/L. The estimated solids/plutonium mass ratio is 55,409.

The largest measured plutonium concentration for this tank is 0.024 g/L (Braun et al. 1994, WHC 1995). This concentration is more than 100 times smaller than the minimum critical plutonium concentration. When the entire volume of DST AW-105 waste is assumed to contain plutonium at this conservative concentration, the plutonium areal density is estimated to be 55.9 g/m² (5.2 g/ft²), or about 46 times smaller than the minimum critical areal density.

DST AW-105 waste contains a high solids/plutonium mass ratio. Whyatt et al. (1996) reported the sum of actual-to-minimum subcritical fractions for iron and zirconium to be 8.4 from process records, as compared to 2.1 from an analysis of core samples. Agnew (1995) calculated the sum of the absorber actual-to-minimum subcritical fractions to be 18.6 for insoluble components and 54.4 for soluble components. Braun et al. (1994) estimates the solids/plutonium mass ratio to be 50,000, and the sum of the actual-to-minimum subcritical fractions to be at least 64.

According to Daling et al. (1997), the top 10-in. layer in DST AW-105 contains almost no plutonium, while the next 6-in. layer contains 7 kg, or about 30% of the tank inventory. No other 6-in.-thick layer contains near this much plutonium. If spread uniformly over the tank, the areal density in this layer would be 17.0 g/m² (1.58 g/ft²), a value 152 times smaller than the minimum critical plutonium areal density. The mass ratio of natural uranium to plutonium in this layer exceeds 1,000, and is sufficient to ensure subcriticality. This sludge was discharged with a cadmium/plutonium atom ratio greater than 2.

In conclusion, the solids/plutonium mass ratios for both DST AW-105 and K Basin sludge are much more than the minimum required to maintain the USL for an unlimited volume of sludge. When combined, the maximum achievable k_{∞} will be no greater than the higher value of the two sludge types alone.

8.9 CUMULATIVE EFFECT OF VARIATIONS

This evaluation is based upon the K Basin sludge model that assumes uranium to be 0.84 wt% ^{235}U enriched and the plutonium content to be consistent with the highest level of burnup. The K Basin sludge model is designed to provide a larger k_{∞} than is possible for any actual composition of uranium and plutonium resulting from the irradiation N Reactor fuel types stored in the basins.

The greatest possible deviation from the K Basin sludge model would be for the sludge to contain uranium enriched to 1.25 wt% ^{235}U . However, this enrichment is only possible for green fuel and would contain no plutonium. When homogeneously mixed at an iron/uranium mass ratio of 0.768, sludge containing 1.25 wt% enriched uranium would have a maximum k_{∞} no more than 0.03 higher than found for this evaluation. This is the largest possible increase in k_{∞} and would be possible only if a large proportion of the uranium were green. Care will be taken to ensure that sludge uranium is not enriched above 0.84 wt%. It is very unlikely that any increase in maximum k_{∞} above the evaluation level will occur as result of improper characterization of uranium.

Replacing ^{238}Pu , ^{241}Pu , and ^{242}Pu with ^{239}Pu and ^{240}Pu in the calculational model is done in a manner that is conservative. The ^{241}Pu content is assumed to be 2.5 wt%, a value larger than present in 99% of the plutonium. The iron/uranium mass ratio fully compensates for these isotopes.

Interaction between K Basin sludge and DST AW-105 sludge is difficult to quantify. The margin of subcriticality for DST AW-105 sludge is greater than that of K Basin sludge. When combined, K Basin sludge will dominate the overall multiplication constant. Maximum k_{∞} for any combination of these sludges will not be greater than that for K Basin sludge alone. When mixed, the resultant k_{∞} should fall between that of the two sludge types alone. The margin of subcriticality for K Basin sludge is considered large enough to compensate for uncertainties associated with DST AW-105 sludge characterization.

Uncertainties associated with the calculational method and material cross sections were incorporated into the USL. Failure to find benchmark experiments using iron contents that closely resemble K Basin sludge configurations is compensated for in a conservative USL based upon a maximum k_{∞} of 0.90 at the 95% confidence level.

Two mechanisms that might potentially increase k_{∞} are failure to add enough iron or failure to properly mix the sludge. Protection against gravity segregation is maintained by requiring an iron/uranium mass ratio of at least 0.768. This also provides protection against improper mixing. The iron/uranium mass ratio would have to fall below 0.256 over a sizeable volume before it is possible to exceed the USL.

Assurance of an iron/uranium mass ratio of 0.768 will be provided by written procedures for iron addition and by chemical analysis of samples drawn from the sludge before discharge.

Assurance that the required iron/uranium mass ratio will be maintained after discharge into the storage tank will be provided by requiring a small particle size and by mixing sludge in each batch before discharge.

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APPENDIX A

PARAMETRIC STUDY

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APPENDIX A

PARAMETRIC STUDY

Curves are provided to permit an assessment of variations in sludge composition on the margin of subcriticality. The relationship between iron content and k_{∞} is investigated as a basis for determining the minimum quantity of iron required to meet criticality safety criteria to assess sensitivity to changes in iron content.

A1.0 CALCULATIONAL METHOD

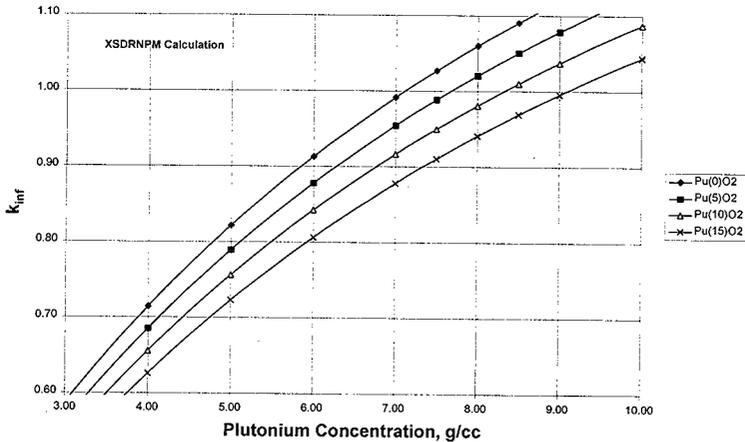
The XSDRNPM computer code was used to calculate criticality parameters using a 200 MHz AST Bravo MS 5200M personal computer. Calculations were made using Version 4.3 of the Standardized Computer Analyses for Licensing Evaluation (SCALE) code system (ORNL 1995) with cross-sections from the 27-group ENDF/B-IV cross-section library. Validation of XSDRNPM is documented in *Verification and Validation of XSDRNPM Code For Tank Waste Calculations* by Rogers and Niemer (1999).

A2.0 PLUTONIUM OXIDE IN WATER

Figure A-1 shows curves of k_{∞} for plutonium oxide in water as a function of plutonium concentration. Curves are shown for plutonium containing 0, 5, 10, and 15 wt% ^{240}Pu . When the ^{240}Pu content is indicated, such as in the legend on figures, it is placed in parentheses. For example, plutonium containing 10 wt% ^{240}Pu is written as Pu(10).

The smallest plutonium concentration that can be made critical in water is 7.1 g/L, and this occurs when the plutonium contains no ^{240}Pu . The minimum critical plutonium concentration increases as the ^{240}Pu content increases. For Pu(15)O₂ the minimum critical concentration becomes 9.0 g/L.

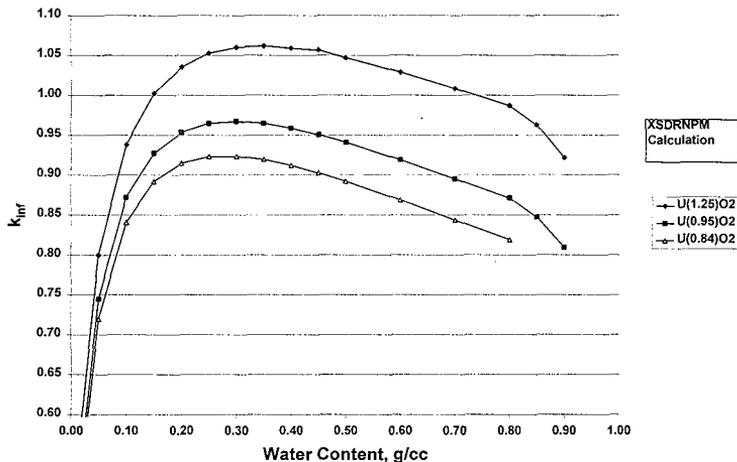
When confined to a finite volume, the smallest critical concentration for plutonium in water is larger than for an unlimited volume. Rogers et al. (1996) provides a graph showing the relationship between sphere minimum critical diameter and plutonium concentration. As the plutonium concentration increases above 7.1 g/L, the sphere critical diameter decreases rapidly. In like fashion, the quantity of plutonium required to achieve criticality also decreases rapidly.

Figure A-1. k_{∞} for Homogeneous PuO_2 in Water.

A3.0 URANIUM OXIDE IN WATER

Figure A-2 shows k_{∞} for homogeneous mixtures of UO_2 and water. Curves are shown for ^{235}U enrichments of 1.25, 0.95, and 0.84 wt%. The 1.25 wt% ^{235}U enrichment represents uranium stored in KW Basin, and the maximum k_{∞} of 1.062 is the highest possible k_{∞} for K Basin sludge. The 0.95 wt% ^{235}U enrichment represents uranium stored in KE Basin, and the maximum possible k_{∞} for this uranium is 0.967.

$\text{U}(0.84)\text{O}_2$ is to be the most reactive form of uranium permitted in sludge received at tank farms. $\text{U}(0.84)\text{O}_2$ has a maximum k_{∞} of 0.923, a value less than 0.95 at the 95% confidence level.

Figure A-2. k_{∞} for Homogeneous UO_2 in Water.

Rods of uranium of optimal diameter optimally arranged in water will have a larger k_{∞} than homogenized uranium-water mixtures. Uranium in K Basin sludge as it arrives at tank farms will be composed of very fine particles and will be governed by critical parameters for homogeneous systems. For this reason, no calculations are provided for arrays of uranium rods.

Figure A-2 is for pure uranium in water. In reality, sludge uranium has been irradiated and contains plutonium. For this reason the true maximum k_{∞} for sludge uranium enriched to 0.84 wt% is greater than shown. Uranium that has plutonium contained within it is discussed in Section A4.0.

A4.0 PLUTONIUM-URANIUM OXIDE MIXTURES IN WATER

Figure A-3 shows k_{∞} for homogeneous UO_2 - PuO_2 as a function of water content and ^{240}Pu content when the ^{235}U enrichment is held constant at 0.84 wt%. For these curves the relationship between ^{240}Pu content and total plutonium is determined by burnup, and a higher ^{240}Pu content correlates to a higher plutonium content. This relationship is shown in Figure 5-1. At 0.84 wt% enrichment and with no plutonium content the maximum k_{∞} is found to be 0.923 at 0.25 g/cc water content (bottom curve in Figure A-3). As plutonium content is increased, maximum k_{∞} increases, and the water content at which it occurs also increases. At highest irradiation K Basin uranium has a plutonium content of 0.30 wt%, and this plutonium contains 16.0 wt% ^{240}Pu .

When the uranium contains 0.30 wt% of highest burnup plutonium, a maximum k_{∞} of 1.034 occurs when water content is 0.35 g/cc. This is the maximum k_{∞} possible when ^{235}U enrichment is 0.84 wt% and the plutonium content is the result of irradiation of N Reactor fuel elements.

In Figure A-3, and in other figures that follow, at a water content above 0.80 g/cm³, k_{∞} decreases at a faster rate with increasing water. The abrupt change in the curve is due to saturation of the solids. Below 0.80 g water/cm³ the solids concentration is constant, while water fills in void space. Solids occupy about 20% of the volume and saturation occurs when water fills the remaining volume. Above 0.80 g water/cm³, there is no longer any void space and solids must be removed to add more water. Since the fissile concentration is reduced as water increases, k_{∞} decreases at a faster rate than before saturation.

Figure A-4 shows maximum k_{∞} from Figure A-3 as a function of ^{240}Pu content. For ^{240}Pu content between 0 and 16 wt% k_{∞} increases monotonically because the total plutonium content is also increasing. However, the rate of increase is slower at higher ^{240}Pu content because ^{240}Pu absorbs slow neutrons without fissioning. In fact, in moderated configurations ^{240}Pu is an effective neutron absorber. At some ^{240}Pu content greater than 16 wt%, k_{∞} will reach an absolute maximum beyond which any further increase in ^{240}Pu will cause k_{∞} to decrease. Over the range of burnup present in K Basin fuel the most reactive composition, based on a constant ^{235}U enrichment of 0.84 wt%, is the composition for greatest burnup (i.e. Pu(16)). In the absence of neutron absorbers (i.e., iron) the maximum k_{∞} for the K Basin sludge model is 1.034, and this occurs with highest burnup plutonium.

Average burnup uranium contains about 0.20 wt% plutonium with a ^{240}Pu content of 11.0 wt%. This composition better represents the uranium expected in well-mixed K Basin sludge. At average burnup maximum k_{∞} is 1.018, a value only 1.5% smaller than for highest burnup. Highest burnup composition is therefore only a little more conservative than average burnup composition. When deriving limits, highest burnup is assumed for all sludge uranium. When highest burnup is assumed, limits for absorber addition are conservative when applied to all K Basin sludge uranium.

Figure A-3. k_{inf} for Homogeneous $U(0.84)O_2$ - PuO_2 in Water.

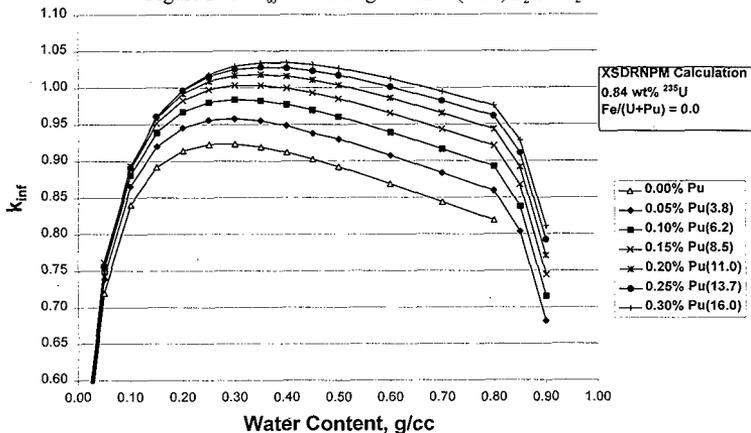
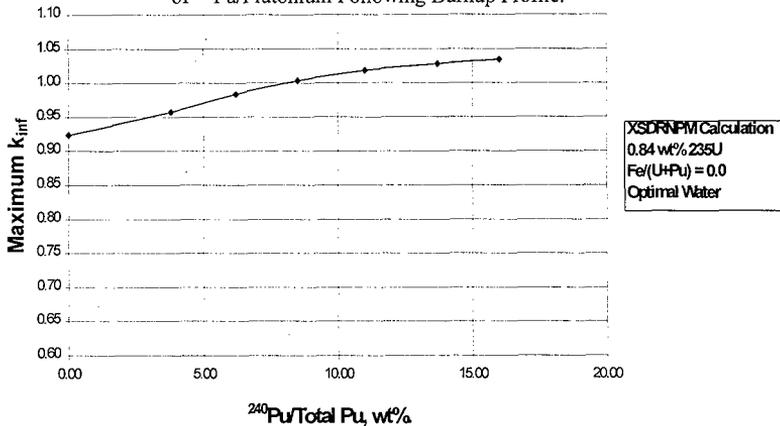


Figure A-4. Maximum k_{inf} for $U(0.84)O_2$ - PuO_2 as a Function of ²⁴⁰Pu/Plutonium Following Burnup Profile.



A5.0 IRON ADDITION FOR AVERAGE AND HIGH BURNUP URANIUM

Figure A-5 shows k_{∞} for average burnup $U(0.84)O_2$ - PuO_2 homogeneously mixed with FeO and water. Average burnup uranium contains 0.20 wt% plutonium with a ^{240}Pu content of 11 wt%. Figure A-6 shows k_{∞} for highest burnup $U(0.84)O_2$ - PuO_2 homogeneously mixed with FeO and water. Highest burnup uranium contains 0.30 wt% plutonium with a ^{240}Pu content of 16 wt%. The iron/heavy-metal mass ratio is varied from 0.0 to 1.76, while ^{235}U enrichment is held constant at 0.84 wt%.

Figure A-7 shows maximum k_{∞} in a homogeneous mixture of $U(0.84)O_2$ - PuO_2 , iron, and water as a function of iron content. Curves are derived from maximum values from Figures A-5 and A-6. High burnup uranium requires a larger quantity of iron to maintain a specified maximum k_{∞} . Figure A-7 can be used to determine sensitivity of maximum k_{∞} to changes in iron content.

Figure A-5. k_{∞} for Average Burnup $U(0.84)O_2$ - PuO_2 and FeO in Homogeneous Water Mixture.

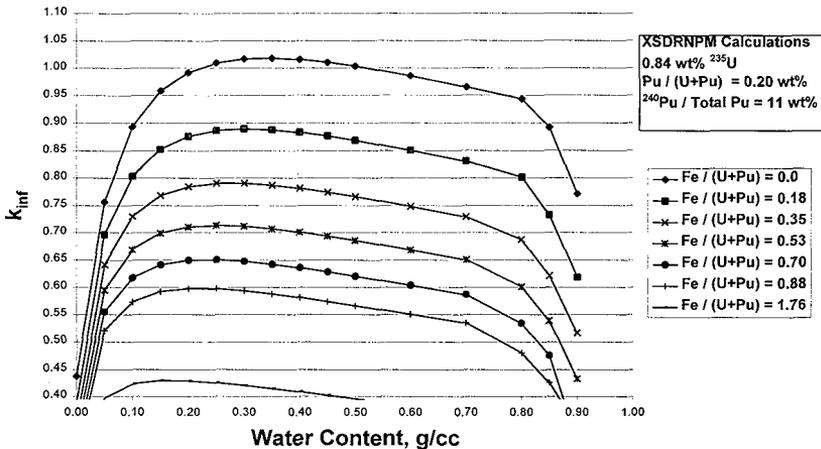


Figure A-6. k_{∞} for High Burnup $U(0.84)O_2$ - PuO_2 and FeO in Homogeneous Water Mixture.

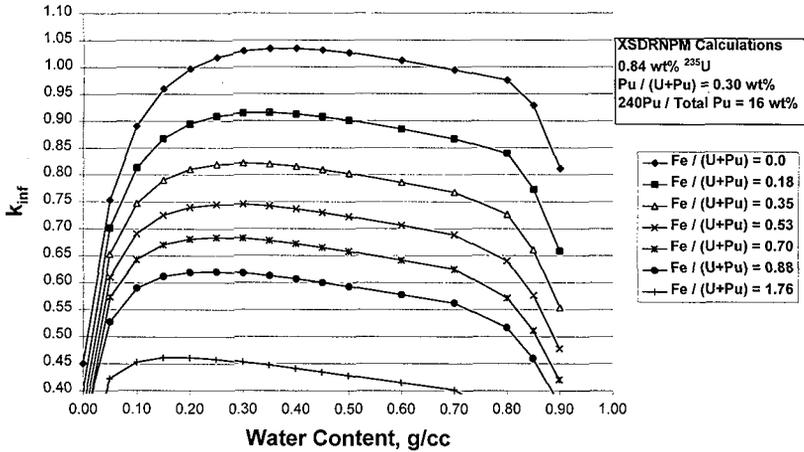
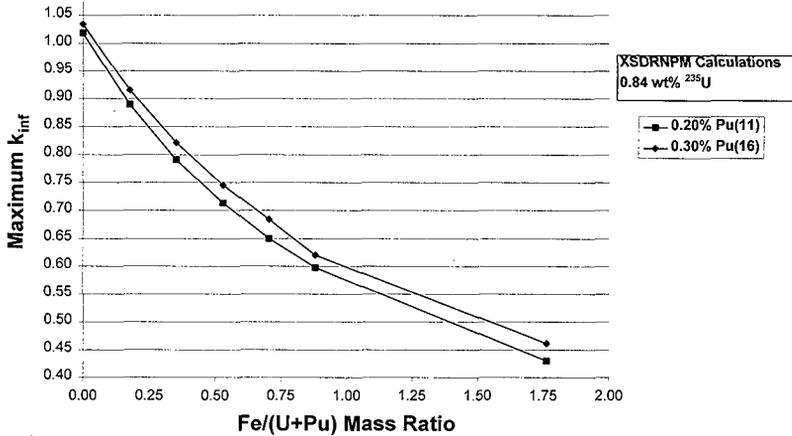


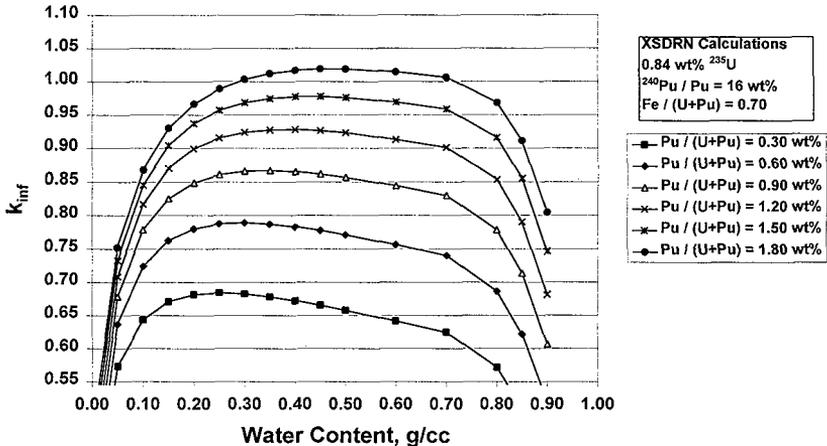
Figure A-7. Maximum k_{∞} for Homogeneous Sludge as a Function of Iron Content.



A6.0 IRON/HEAVY-METAL MASS RATIO OF 0.70

Figure A-8 shows k_{∞} for mixtures of high burnup $U(0.84)O_2$ - PuO_2 , iron, and water as a function of plutonium and water content. These graphs show the sensitivity of changing plutonium content when the iron/heavy-metal mass ratio is held constant at 0.70. The bottom curve shown is the most reactive possible for irradiated uranium under normal conditions. The curves for plutonium content above 0.30 wt% are purely hypothetical and are intended to show sensitivity to increases in plutonium concentration. The top curves shows the degree to which a concentrating mechanism would have to increase the plutonium concentration before criticality becomes possible. See Section A9.0 for a curve of maximum k_{∞} as a function of plutonium content.

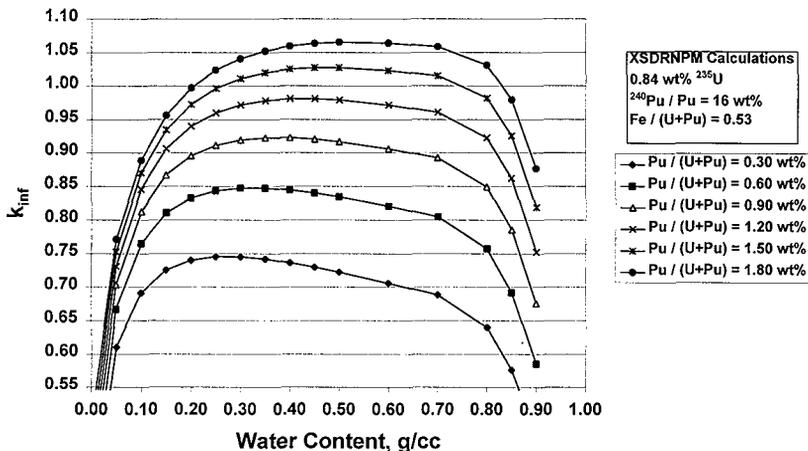
Figure A-8. k_{∞} for High Burnup $U(0.84)O_2$ - PuO_2
When the $Fe/(U+Pu)$ Mass Ratio is 0.70.



A7.0 IRON/HEAVY-METAL MASS RATIO OF 0.53

Figure A-9 shows k_{∞} for mixtures of high burnup $U(0.84)O_2$ - PuO_2 as a function of plutonium and water content when the iron ratio is held constant at 0.53. Figure A-9 repeats Figure A-8, but with a lower iron content in the sludge. Maximum k_{∞} as a function of plutonium content (based on these curves) is shown in Figure A-15 (Section A9.0).

Figure A-9. k_{∞} for High Burnup $U(0.84)O_2$ - PuO_2
When the $Fe/(U+Pu)$ Mass Ratio is 0.53.



A7.1 EFFECT OF URANIUM-235 ENRICHMENT ON k_{∞}

Figure A-10 shows k_{∞} for homogeneous mixtures of UO_2 , PuO_2 , iron, and water as a function of water content and ^{235}U enrichment between 0.75 wt% and 1.00 wt%. Plutonium content is constant at 0.30 wt% with a ^{240}Pu content of 16 wt% (high burnup). The iron/heavy-metal mass ratio is held constant at 0.53. This figure shows sensitivity of k_{∞} to changes in ^{235}U enrichment when the plutonium and absorber (iron) contents are near to that assumed for sludge discharged into DST AW-105.

Figure A-11 shows maximum k_{∞} as a function of ^{235}U enrichment for compositions in Figure A-10. Maximum k_{∞} increases almost linearly with enrichment. The iron content in these configurations causes the change in maximum k_{∞} to be relatively small over the range of ^{235}U enrichment possible. The maximum k_{∞} is 0.720 at 0.75 wt% ^{235}U enrichment and is 0.789 at 1.00 wt% ^{235}U enrichment.

Maximum k_{∞} for uranium in water when no iron is present is shown in Figure A-2.

Figure A-10. k_{inf} versus Water Content for Various ^{235}U Enrichments for Sludge Having an Fe/(U+Pu) Mass Ratio of 0.53.

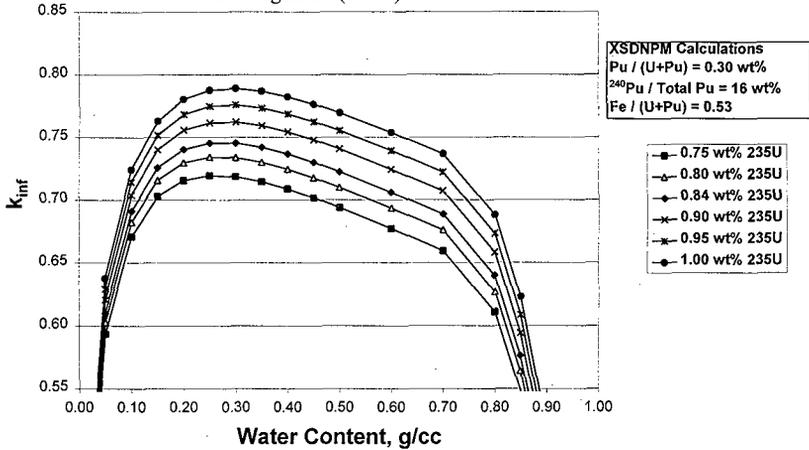
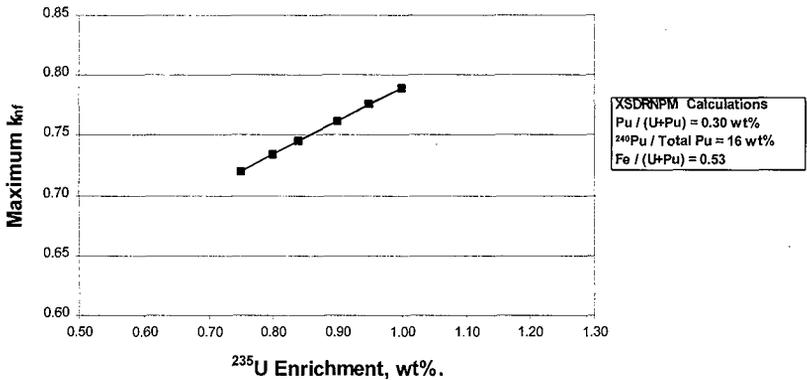


Figure A-11. Maximum k_{inf} as a Function of ^{235}U Enrichment for Sludge Having an Fe/(U+Pu) Mass Ratio of 0.53.



A7.2 CHANGE IN PLUTONIUM-240 WITH TOTAL PLUTONIUM CONSTANT

Figure A-12 shows k_{∞} as a function of ^{240}Pu and water content for an iron/heavy-metal mass ratio of 0.53. The total plutonium content in the uranium is held constant at 0.30 wt%, while ^{240}Pu content varies between 0 and 16 wt% of the plutonium. These curves show sensitivity of k_{∞} to errors in the ^{240}Pu /plutonium mass ratio

Figure A-12. k_{∞} versus Water Content for Various ^{240}Pu Contents for Sludge Having an Fe/(U+Pu) Mass Ratio of 0.53.

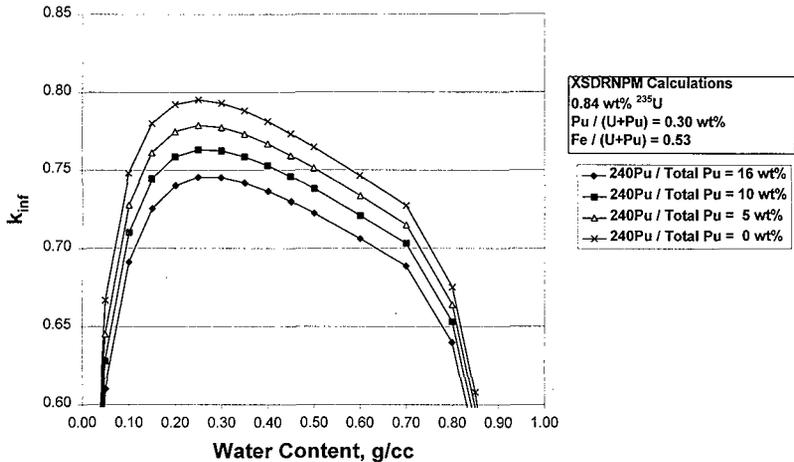


Figure A-13 plots maximum k_{∞} for curves in Figure A-12 as a function of ^{240}Pu content. Maximum k_{∞} decreases as ^{240}Pu content increases for two reasons. First, ^{240}Pu cannot be made critical with moderated neutrons and is therefore a mild neutron poison. Second, when ^{240}Pu content increases ^{239}Pu content decreases to maintain the total plutonium constant.

Figure A-13. Maximum k_{∞} as a Function of ^{240}Pu Content for Constant Total Plutonium at Iron/heavy-metal Mass Ratio of 0.53.

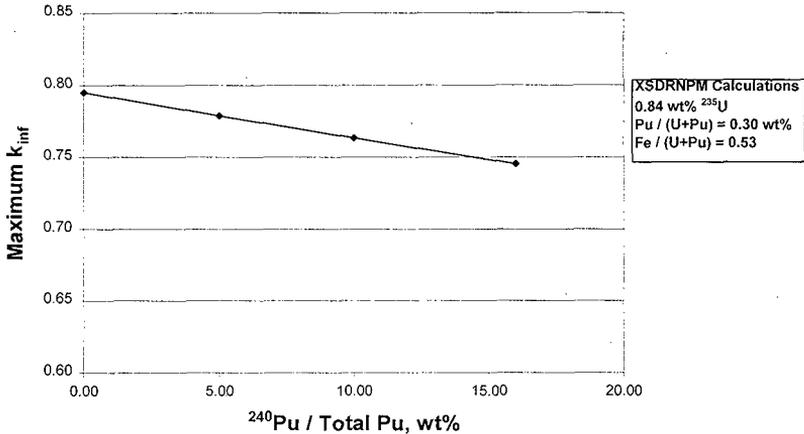
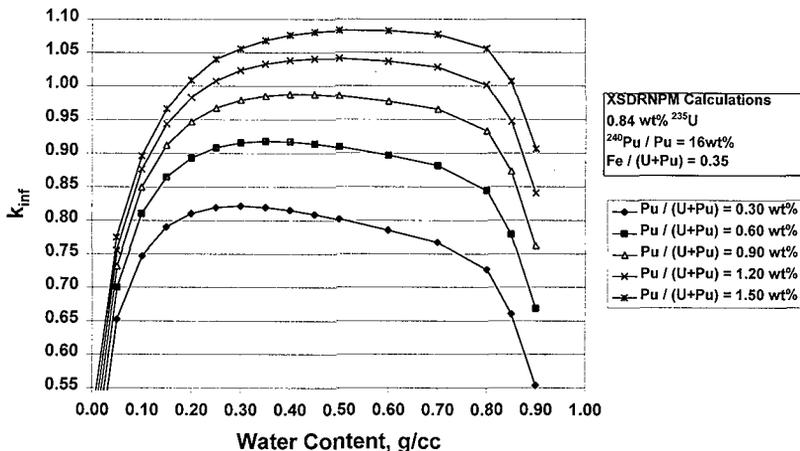


Figure A-13 differs from Figure A-4 because of the relationship between ^{240}Pu and the plutonium/uranium mass ratio. For Figure A-13 the plutonium/uranium mass ratio is held constant, and for Figure A-4 the ^{240}Pu content and the plutonium/uranium mass ratio are related through the relationship shown in Figure 5-1. For Figure A-4 a larger ^{240}Pu /plutonium mass ratio correlates to a larger plutonium content, and the corresponding maximum k_{∞} increases.

A8.0 IRON/HEAVY-METAL MASS RATIO OF 0.35

Figure A-14 shows k_{∞} for mixtures of high burnup $\text{U}(0.84)\text{O}_2\text{-PuO}_2$, iron, and water as a function of plutonium and water content when the iron/heavy-metal mass ratio is held constant at 0.35. These curves are the same as for Figures A-8 and A-9, except that the iron content has been reduced.

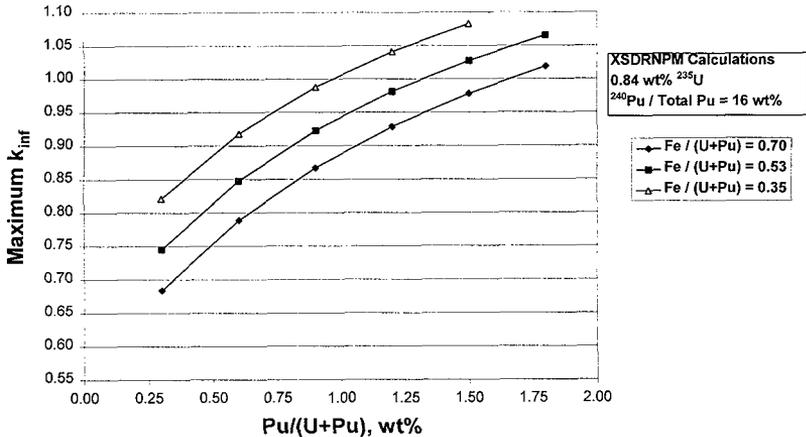
Figure A-14. k_{inf} versus Water Content for Various ^{235}U Enrichments for Sludge Having an $\text{Fe}/(\text{U}+\text{Pu})$ Mass Ratio of 0.35.



A9.0 COMPARISON OF IRON/HEAVY-METAL MASS RATIOS

Figure A-15 shows maximum k_{inf} as a function of plutonium content for high burnup $\text{U}(0.84)\text{O}_2\text{-PuO}_2$ for $\text{Fe}/(\text{U}+\text{Pu})$ mass ratios of 0.35, 0.53, and 0.70. Values shown are taken from Figures A-6, A-9, and A-14. This figure shows sensitivity of maximum k_{inf} to changes in plutonium and iron content. The lower end of these curves corresponds to the highest plutonium content actually present in K Basin sludge (i.e., 0.30 wt%).

Figure A-15. Maximum k_{∞} for $U(0.84)O_2$ - PuO_2 as a Function of the $Pu/(U+Pu)$ Mass Ratio.



A10.0 REFERENCES

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Rogers, C. A., and K. A. Niemer, 1999, *Verification and Validation of the XSDRNPM Codes for Tank Waste Calculations*, HNF-3950, Duke Engineering Services Hanford, Richland, Washington.

APPENDIX B
CRITICALITY PARAMETERS

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APPENDIX B

CRITICALITY PARAMETERS

A compilation of criticality parameters is provided to support this criticality safety evaluation.

B1.0 URANIUM CRITICAL PARAMETERS

For low enriched uranium the minimum critical mass for a heterogeneous lattice of optimal diameter rods in water is smaller than that for a homogenized solution. For optimized pieces criticality is possible for enrichments as low as the 0.72 wt% content in natural uranium (Paxton and Pruvost 1986). For a homogeneous mixture of uranium in water criticality can not occur unless the ^{235}U enrichment is at least 1.03 wt%.

Table B-1 shows critical masses provided by Schwinkendorf (1995) for low enriched uranium. Critical masses for uranium pieces (scrap) are based on optimally sized pieces at an optimal spacing in water. Critical masses for uranium pieces (scrap) are the smallest for which criticality is possible under any condition. The parameter shown for cylinders is the minimum critical mass per unit length and for slabs is the minimum critical mass per unit area.

A subcritical limit value based on a k_{eff} of 0.98 is provided in Table B-1. This defines the limit which is assured of being subcritical and is often used in setting operating limits for low enriched uranium, although sometimes a more conservative value based on a k_{eff} of 0.95 is used.

For 0.95 wt% enriched uranium pieces the minimum critical mass is 2886 kg uranium (27 kg ^{235}U). When k_{eff} is limited to 0.98, the minimum mass is 1951 kg. The large change in mass for a small change in k_{eff} clearly shows that the mass is a sensitive function of k_{eff} for 0.95 wt% enriched uranium. The minimum critical areal density is 2124 kg/m², and the minimum areal density for which k_{eff} can be 0.98 is 1763 kg of uranium. Criticality is precluded for 0.95 wt% uranium in the form of particles with dimension (diameter) less than 0.13 cm (0.05 in.).

For 1.25 wt% enriched uranium pieces the minimum critical mass is 672 kg uranium (8.4 kg ^{235}U). When k_{eff} is limited to 0.98, the minimum mass is 532 kg of uranium. The minimum critical areal density is 967 kg/m², and the minimum areal density for which k_{eff} can be 0.98 is 864 kg/m² of uranium. For 1.25 wt% enriched uranium criticality is possible no matter how small the particle size. However, 1364 kg of uranium (17.0 kg ^{235}U) is required to reach a k_{eff} of 0.98 when particles are smaller than 0.13 cm.

Table B-1. Critical Masses for Low Enriched Uranium ¹

	Mass (kg) For $k_{\text{eff}} = 0.98$		Mass (kg) For $k_{\text{eff}} = 1.00$	
	Heterogeneous Pieces	Homogeneous Solution	Heterogeneous Pieces	Homogeneous Solution
0.95 wt% ²³⁵ U Enrichment				
Sphere	1951	66460	2886	infinite
Hemisphere	3269	104600	4774	infinite
Cylinder, kg/m	1688	19090	2247	infinite
Slab, kg/m ²	1763	6870	2124	infinite
1.25 wt% ²³⁵ U Enrichment				
Sphere	532	1364	672	2070
Hemisphere	941	2303	1172	3434
Cylinder, kg/m	621	1204	740	1610
Slab, kg/m ²	864	1269	967	1533

Note:

¹ Schwinkendorf (1995)

Table B-2 shows critical dimensions provided by Schwinkendorf (1995) for low enriched uranium. For 0.95 wt% enriched uranium pieces the minimum critical sphere diameter is 101.3 cm (39.8 in.). The minimum sphere diameter for which k_{eff} is 0.98 is 91.4 cm (36.0 in.). For a slab the minimum critical thickness is 48.0 cm (18.9 in.) and the minimum thickness for which k_{eff} can be 0.98 is 46.5 cm (18.3 in.). A homogeneous mixture of 0.95 wt% enriched uranium can not be made critical.

For 1.25 wt% enriched uranium pieces the minimum critical sphere diameter is 69.6 cm (27.4 in.). The minimum sphere diameter for which k_{eff} can be 0.98 is 64.5 cm (25.4 in.). For a slab the minimum critical thickness is 33.5 cm (13.2 in.) and the minimum thickness for which k_{eff} can be 0.98 is 30.0 cm (11.8 in.).

As a homogeneous mixture, the minimum critical sphere diameter for 1.25 wt% enriched uranium is 103.4 cm (40.7 in.). The minimum sphere diameter for which k_{eff} can be 0.98 is 89.9 cm (35.4 in.). For a slab the minimum critical thickness is 48.8 cm (19.2 in.) and the minimum thickness for which k_{eff} can be 0.98 is 45.2 cm (17.8 in.).

Table B-2. Critical Dimensions for Low Enriched Uranium ¹.

	Dimension (cm) For $k_{\text{eff}} = 0.98$		Dimension (cm) For $k_{\text{eff}} = 1.00$	
	Heterogeneous Pieces	Homogeneous Solution	Heterogeneous Pieces	Homogeneous Solution
0.95 wt% ²³⁵ U Enrichment				
Sphere Diameter	91.4	295.9	101.3	infinite
Hemisphere Dia.	136.9	433.8	151.1	infinite
Cylinder Dia.	69.9	222.8	77.0	infinite
Slab Thickness	46.5	140.5	48.0	infinite
1.25 wt% ²³⁵ U Enrichment				
Sphere Diameter	64.5	89.9	69.6	103.4
Hemisphere Dia.	98.3	134.9	105.7	154.2
Cylinder Dia.	48.3	65.5	52.8	75.7
Slab Thickness	30.0	45.2	33.5	48.8

Note:

¹Schwinkendorf (1995)

B2.0 PLUTONIUM CRITICAL PARAMETERS

In most evaluations of tank waste, uranium is assumed to be natural or depleted, and critical parameters are defined in terms of plutonium content. In the past, the ²³⁵U in uranium enriched to 1.0 wt% has not been included in the fissile (plutonium) inventory. When enriched uranium is present, the ²³⁵U is usually replaced by its "plutonium equivalence." The fissile component of K Basin sludge is primarily enriched uranium, and the ²³⁵U must be taken into account. In this evaluation the ²³⁵U in uranium above an enrichment of 0.72 wt% (i.e., that of natural uranium) is treated as if it were plutonium and is added to the "plutonium equivalent" inventory. Usually this is referred to as the plutonium inventory.

For the evaluation of tank waste, plutonium concentration is a key parameter. When the plutonium concentration is everywhere less than the minimum critical concentration for the waste composition, subcriticality is assured. A Conservative Waste Model (CWM) with reduced neutron absorption and optimized water was developed by Rogers (1993) to provide a waste composition for which critical concentrations and critical dimensions are conservative relative to any real waste. The complexity of waste and the lack of knowledge of the distribution of waste

types can be overcome by replacing an entire configuration of waste layers and mixtures with the CWM. Based on the CWM, the subcritical limit for plutonium concentration in waste solids is 2.6 g/L. When applied to K Basin sludge, the subcritical limit on the sum of the ^{235}U and plutonium concentrations is 2.6 g/L.

When the plutonium concentration is 3.6 g/L, criticality requires at least 250 kg of plutonium in a spherical volume of 69,400 L. When the plutonium concentration is 10 g/L, criticality requires 3.0 kg of plutonium in a 300-L sphere. If no restriction is placed on the plutonium concentration, the smallest mass of plutonium that can be made critical is 520 g at a plutonium concentration of about 30 g/L in pure water. At optimal moderation the difference in critical plutonium content between plutonium as metal and as oxide is negligible.

The experimentally determined plutonium minimum critical concentration is 7.2 g/L in water (Paxton and Pruvost 1986) and the minimum critical plutonium areal density provided by Carter et al. (1970) is 240 g Pu/ft^2 (2,582 g/m^2). These parameters are supported by authoritative documents and are used as subcritical limits.

The most probable geometry for a waste stream is a slab. The minimum critical plutonium mass in a uniform slab covering the entire area of a 22.9-m-diameter tank at a uniform depth was calculated to be slightly less than 1,000 kg. When Carter's less conservative minimum critical areal density of 2,582 g/m^2 is used, the minimum critical mass is 1,060 kg. This quantity exceeds the total inventory for tank farms and is considerably larger than the projected inventory of DST AW-105 after K Basin sludge has been added.

The addition of solids to a plutonium-water solution increases the minimum critical plutonium areal density (Rogers et al. 1996). Therefore, a subcritical limit on the areal density of 2,582 g/m^2 derived from plutonium in pure water is conservative for all compositions of waste solids and water.

B3.0 ABSORBER-TO-FISSILE MINIMUM SUBCRITICAL MASS RATIOS

Subcriticality is ensured by a plutonium concentration below the minimum for which criticality is possible or by a high mass ratio of absorbers to plutonium. A set of minimum subcritical absorber-to-plutonium (X/Pu) mass ratios is defined, where X is used to designate the absorber type. The actual X/Pu mass ratio for a waste component divided by its corresponding minimum subcritical mass ratio is referred to as the actual-to-minimum subcritical mass fraction (usually shortened to subcritical fraction). When the sum of the subcritical fractions for components is greater than unity, the waste is subcritical.

Uranium, iron, and manganese have a very low solubility under conditions found in tank waste. These are important waste components for which detailed studies have been made. Table B-3 presents the X/Pu and X/ ^{235}U minimum subcritical mass ratios for these elements.

Table B-3. Absorber-to-Fissile Mass Ratios to Ensure Subcriticality.

Element or compound	Minimum subcritical X/Pu mass ratio	Minimum subcritical X/ ²³⁵ U mass ratio
²³⁸ U	---	Heterogeneous 139 Homogeneous 100
Natural uranium	770	---
Iron	160	77
Manganese	32	30

The minimum subcritical X/Pu mass ratios were calculated for waste components shown in Table B-4 (Rogers et al. 1996). These elements were chosen because of their high neutron absorption cross sections, or because of their relative abundance in the waste.

Table B-4. Absorber-to-Plutonium Mass Ratios to Ensure Subcriticality.

Component	Subcritical limit X/Pu mass ratio
Aluminum	910
Calcium	770
Chromium	135
Copper	130
Lanthanum	121
Nickel	105
Nitrogen	61
Nitrate	270
Sodium	360
Thorium	243
Zirconium	4,000

A $^{238}\text{U}/^{235}\text{U}$ minimum subcritical mass ratio of 139 is based upon the 0.72 wt% ^{235}U content in natural uranium being the lowest enrichment which can be made critical in a lattice of uranium rods. For a homogenous mixture of uranium and water criticality is not possible for an ^{235}U enrichment less than 1.0 wt% (Carter et al. 1969). The $^{238}\text{U}/^{235}\text{U}$ minimum subcritical mass ratio for a homogeneous mixture is therefore 100.

Whyatt et al (1996) conclude that plutonium primarily resides in the solid phase of the tank waste in the form of agglomerates. The solubility of plutonium in alkaline salt solution is low enough that saturation concentrations in waste liquids are at least 30 times lower than the minimum concentration needed to support a criticality. Tank waste is maintained alkaline to ensure that the uranium and plutonium remains combined with the solids. K Basin sludge is to be made alkaline with a pH of at least 8 before transfer into DST AW-105.

B4.0 COMBINING THE EFFECT OF ABSORBERS

The contributions of different components to the total absorption cross section can be combined by adding their individual contributions. The following rules can be used to evaluate a waste composition (Rogers et al. 1996).

- Calculate the actual absorber-to-plutonium mass ratio (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 or simply as the subcritical fraction.
- When the sum of the actual-to-minimum subcritical mass fractions for individual components is greater than unity, the waste is subcritical.

When the following expression is satisfied, the waste is subcritical:

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

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.

Even if a large fraction of the soluble components were to be removed, it would be unlikely for the plutonium concentration to increase sufficiently for criticality to occur. The areal density of plutonium is 55.9 g/m², a value 46 times smaller than the minimum required for criticality.

D5.0 k_{∞} OF DST AW-105 SLUDGE

Braun et al. discusses the neutron multiplication constant (k_{∞}) for waste samples from DST AW-105 selected because of "their relatively low neutron absorber content." Specifically, the samples selected did not meet the uranium/plutonium or the iron/plutonium minimum subcritical ratios, and the total mass ratio fractions were less than that for most other samples. The compositions of these samples are shown in Table D-4.

Values of k_{∞} were calculated for three conditions:

1. "As Analyzed" Composition: The plutonium concentration, solids composition and the water content are as analyzed. This composition provides a water content of between 70 and 80 wt%. The plutonium concentration varies from 0.013 to 0.024 g/L.
2. "As Analyzed" Composition with Selected Absorbers Removed: The composition is "as analyzed", except that boron, silicon, calcium, and potassium are removed and the solids renormalized to maintain the density unchanged. Because the water content is high and the boron content is small, the removal of these components reduces k_{∞} by only a small amount (i.e., it is not significant).
3. Dry Conditions with Greatly Elevated Plutonium Content: The water content is reduced to 50 g/L (5.5 wt%), and the solids density is increased to 850 g/L to replace the water. The solids density is less than expected for "real" dry sludge. The plutonium concentration is arbitrarily set at 3.0 g/L, a value 100 times greater than the measured value.

Calculations of k_{∞} were made for water content between 600 and 35 g/L. Over this entire range of water content, k_{∞} shows a monotonic increase as the water content is decreased. A water content of 50 g/L represents waste that is drier than considered possible in practice. Therefore, a water content of 50 g/L is conservative when compared to "real" waste.

Results of calculations are summarized in Table D-5. Using the measured water content (normal conditions), k_{∞} is calculated to be less than 0.03. Under hypothetical conditions using a water content reduced to 50 g/L and using a plutonium concentration arbitrarily increased to 3 g/L, k_{∞} is found to increase to 0.38. This is an extreme condition with less water content than should actually ever occur and with a plutonium concentration more than 100 times greater than measured. Even under these hypothetical conditions, the waste remains subcritical.

B6.0 REFERENCES

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APPENDIX C

K BASIN SLUDGE ORIGIN AND CHARACTERIZATION

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APPENDIX C**K BASIN SLUDGE ORIGIN AND CHARACTERIZATION**

Praga (1998) provides a description of design basis feed compositions for materials stored or processed by SNF facilities and activities in *105-K Basin Material Design Basis Feed Description for Spent Fuel Project Facilities, Volume 1, Fuel*, HNF-SD-SNF-TI-009. Chemical and radionuclide inventories are estimated for fuel stored in the KE and KW Basins. Praga (1998) includes the nominal (or overall average) inventory and a description of fuel element types.

Sludge characterization data in *105-K-Basin Material Design Basis Feed Description for Spent Fuel Project Facilities, Volume 2, Sludge* (Pearce et al. 1998) "utilizes the most current characterization data available to define the various sludge inventories." Feed descriptions apply to K Basin sludge that has not been treated. Sludge compositions and physical characteristics are compiled in Appendices A through F of Pearce et al. (1998). This data came from characterization campaigns conducted between 1994 and 1998 by the Hanford Site SNF Project. When waste is received at tank farms, it will have undergone a treatment process and will have changed from the description provided. However, the uranium enrichment and plutonium content will not change.

C1.0 PROCESS STREAMS

K Basin sludge is categorized as being from KE Basin or from KW Basin. Five process streams, designated KE1, KE2, KW1, KW2, and KW3, are defined by the locations where the sludge is collected and held in interim storage. Interim storage for process stream KE1 is in the KE Basin Weasel Pit and for the KE2 stream is in the Integrated Water Treatment System (IWTS) Knockout Pots. Process stream KW1 contains sludge retrieved from pit and floor areas. Interim storage for process stream KW2 is in the IWTS Knockout Pots and for process stream KW3 is in the settler tanks. Table C-1 lists the locations where sludge originates and locations where it is stored, based on Pearce et al. (1998).

Table C-1. Process Feed Streams.

Process Stream	Storage Location	Location of Origin	Volume m ³
KE1	Weasel Pit	Tech View	0.4
		Weasel Pit	10.1
		North Loadout Pit	6.3
		Dummy Elevator Pit	1.4
		Main Basin Floor	21.5
KE2	Knockout Pot <1/4 in. dia. >250 µm	Empty Canisters ¹	0.02
		Full Canisters	2.0
		Internal Sludge	0.26
		Fuel Pieces	0.15
KW1	Basin Storage	Tech View	0.07
		Weasel Pit	0.03
		North Loadout Pit	3.65
		Dummy Elevator Pit	0.04
		Main Basin Floor	0.82
KW2	Knockout Pot <1/4 in dia. >500 µm	Fuel Pieces	0.20
KW3	Settler Tanks < 500 µm dia.	Full Canisters	1.88
		Coating	
		Internal Sludge	

Note:

¹"Empty" refers to canisters that do not contain any fuel elements or pieces.

Sludge components of interest to criticality safety are shown in Table C-2, based on Pearce et al. The highest values for ^{235}U enrichment in KE and KW sludge are 0.68 wt% and 0.79 wt%, respectively. Since Pearce et al. (1998) was published, 17 Mark IA elements containing 1.25 wt% enriched uranium have been temporarily stored in the KE Basin, but these elements do not make any contribution to K Basin sludge.

Table C-2. Values for Selected Sludge Components.

Fissionable Component ¹	KE Basin Sludge Streams ^a		KW Basin Sludge Streams ^a		
	KE1	KE2	KW1	KW2	KW3
Total Mass, kg					
Plutonium, Conservative ²	8.08	10.23	0.318	5.19	9.26
Best Estimate ³	5.25	6.65	0.207	3.37	6.02
Uranium	2615.0	2819.2	87.24	1464.4	2207.8
FeO(OH)	7960.6	216.2	323.96	---	336.4
Uranium Isotopic Composition, wt%					
^{233}U	0.0000	0.0004	0.0067	---	---
^{235}U	0.6361	0.6769	0.6794	0.7800	0.7883
^{238}U	99.29	99.24	99.24	99.11	99.14
Plutonium Content, wt%					
Pu/(U+Pu), Conservative ²	0.308	0.362	0.363	0.353	0.418
Best Estimate ³	0.200	0.235	0.236	0.229	0.272
As-Settled Density, g/cm ³					
Dried solids content	0.564	1.655	0.382	8.242	1.915
Wet sludge density	1.403	2.359	1.292	8.549	2.582
Sludge Volume, m ³					
Sludge Volume	41.459	2.369	4.67	0.199	1.878

Notes:

¹ Based on Pearce et al. (1998), Tables 3-1 and 3-2.

² All plutonium activity assumed to be from ^{239}Pu .

³ ^{240}Pu , ^{238}Pu , and ^{239}Pu activities are used separately to find individual isotopic contents.

Pearce et al. (1998) derived plutonium content from activity measurements by assuming all activity of ^{238}Pu , ^{239}Pu , and ^{240}Pu is produced by ^{239}Pu . This approach over-estimates plutonium content. A "best estimate" plutonium content is made by treating the activities of ^{238}Pu and ^{240}Pu as separate from that of ^{239}Pu . Table C-3 shows the specific activities of these plutonium isotopes and their approximate contribution to total activity. When correction for the individual isotopic contributions is made, the total plutonium is reduced to less than 0.65 times the value reported by Pearce et al. Corrected values (shown in Table C-2 as "best estimates") are consistent with the K Basin sludge model.

Table C-3. Activity Fractions of Plutonium Isotopes in Irradiated N Reactor Fuel.

Isotope	Specific Activity Ci/g	Fraction of Total Activity
^{238}Pu	17.12	0.13 - 0.36
^{239}Pu	0.06217	0.40 - 0.55
^{240}Pu	0.2279	0.26 - 0.33

C2.0 FUEL BURNUP SUMMARY

Praga (1998) provides fuel burnup summaries for N Reactor Mark IA and Mark IV fuel elements, based on accountability records generated November 17, 1994. Appendix A of Praga lists the accountability database used. Tables C-4 and C-5 are copied from Praga.

Table C-4. N Reactor Mark IA Fuel Burnup Summary.

% ^{240}Pu Range	Mass, MTU	% of total mass
≤ 5	166.94	11.35
5 - 7	125.25	8.81
7 - 9	0.0598	0.00
9 - 11	62.988	4.28
11 - 13	270.56	18.39
13 - 15	714.01	48.54
> 15	131.29	8.92
Total	1471.1	100.

Table C-5. N Reactor Mark IV Fuel Burnup Summary.

% ²⁴⁰ Pu Range	Mass, MTU	% of total mass
≤ 5	36.124	5.75
5 - 7	3.373	0.54
7 - 9	0	0.00
9 - 11	68.008	10.83
11 - 13	118.59	18.88
13 - 15	401.88	64.00
> 15	0	0.00
Total	627.98	100

Data in Appendix C of Pearce et al. (1998) for KE Wash Sludge is used to construct Table C-6. The plutonium content falls between 0.10 and 0.25 wt%, and the ²⁴⁰Pu content is found to fall between 13.0 and 14.4 wt% of the plutonium. The K Basin sludge model is conservative when compared to these compositions.

Table C-6. Isotopic Breakdown of KE Wash Sludge.

Isotope	Sample CS1	Sample SSL1	Sample SSL2	Sample SSL2dup	Sample SSL3	Sample SSLmean	Fuel Pieces
Activity, $\mu\text{Ci/g(waste)}$							
²³⁸ Pu	16.5	27.7	26.0	25.3	36.6	29.98	49.34
²³⁹ Pu	61.7	128.0	102.0	101.0	150.0	126.50	94.61
²⁴⁰ Pu	36.3	76.6	59.0	58.7	93.1	76.18	51.95
g(Pu)/g(waste)							
²³⁸ Pu	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000
²³⁹ Pu	0.00100	0.00208	0.00165	0.00164	0.00243	0.00205	0.00153
²⁴⁰ Pu	0.00016	0.00034	0.00026	0.00026	0.00041	0.00034	0.00023
²⁴⁰ Pu/Plutonium, wt%							
²⁴⁰ Pu /Pu	13.8	14.0	13.6	13.7	14.4	14.2	13.0
Uranium Isotopic Content, wt%							
²³⁵ U	0.682	0.677	0.685	0.683	0.93	0.76	0.719
²³⁸ U	99.23	99.23	99.22	99.22	98.98	99.14	99.19

C3.0 REFERENCES

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- Praga, A. N., 1998, *105-K Basin Material Design Basis Feed Description for Spent Fuel Project Facilities, Volume 1, Fuel*, HNF-SD-SNF-TI-009, Volume 1, Rev. 2, Duke Engineering and Services Hanford, Richland, Washington.

APPENDIX D

DST AW-105 SLUDGE ORIGIN AND CHARACTERIZATION

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APPENDIX D

DST AW-105 SLUDGE ORIGIN AND CHARACTERIZATION

DST AW-105 is 23 m (75 ft) in diameter and contains 1,643,000 L (434,000 gal) of waste from the processing of fuel at the Plutonium Uranium Reduction Extraction Plant (PUREX) (Hanlon 1998). The Double Shell Tank Plutonium Inventory Database (LMHC 1998) shows the April 1999 inventory to be 22.95 kg. The total volume of waste is shown as 430.5 kgal, of which 280 kgal is solids. The plutonium concentration in the supernatant liquid is $3.22E-06$ g/L, and the plutonium concentration in the solids is 0.0217 g/L. The estimated solids/plutonium mass ratio for the tank is 55,409. The largest measured plutonium concentration for this tank is 0.024 g/L (Braun et al. 1994, WHC 1995), less than 1% of the minimum required for criticality under the most idealized conditions. The average plutonium areal density is estimated to be 55.9 g/m² (5.2 g/ft²), or 2.2% of the minimum critical areal density.

D1.0 CONTROLS ON INCOMING TRANSFERS

Waste presently in DST AW-105 was received under two basic CPS limits. First, the plutonium concentration in the holdup vessel at the facility from which the waste originated was required to be less than 0.013 g/L (0.05 g/gal) at the time of discharge. This is an average over the entire waste batch volume after being stirred just prior to discharge. Second, the plutonium concentration in the solids must not exceed 1.0 g/L after settling.

The PUREX tank from which most plutonium bearing waste was discharged has a volume of 18,900 L (5,000 gal). To meet the discharge limit of 0.013 g/L (maximum), the total mass of plutonium discharged at one time could therefore not exceed 250 g. Since there was a period of at least several days between discharges, the solids in each discharge had time to settle before the next discharge.

In 1995 waste was sent to DST AW-105 from PUREX under discharge limits requiring the presence of cadmium to compensate for allowing an increase in the plutonium concentration in settled solids. The cadmium concentration in these transfers was enough to maintain subcriticality at a plutonium concentration greater than 30 g/L, although the actual plutonium concentration was less than 0.05 g/L. The mass of plutonium in a transfer did not exceed 500 g, and the uranium/plutonium mass ratio was high enough to ensure subcriticality. This waste lies at the top of the sludge layer. These were the only transfers ever where cadmium was added to allow a higher concentration of plutonium to be transferred.

D2.0 TANK CONTENTS ORIGIN

Appendix A of Whyatt et al. (1996) and Chapter 9 of Serne et al. (1996) both contain the same review of tanks waste titled *Properties of Generated Waste Relevant to Criticality Hazard*. This is a detailed review of historic records of discharges to determine the contents of waste storage tank.

The official tank inventory is based upon Baseline Report WHC-SD-WM-TI-640 (January 1, 1995, with updates to April 1996) which reports that DST AW-105 contains 1,124,000 L of sludge, 288,000 L of liquid, and 22,947 g of plutonium (Whyatt et al. 1996). A second estimate based upon the Hanford Defined Waste (HDW) Model provides a plutonium inventory of 12,600 g (Agnew 1996).

The HDW model estimates a significantly smaller plutonium inventory than the official inventory. This difference reflects the fact that the official inventory conservatively assumes that the largest measured plutonium concentration applies to the entire volume of the tank.

Serne et al. (1996, Appendix D) characterizes waste streams to DST AW-105 as follows:

- (1) the primary stream was low-level waste from PUREX;
- (2) the secondary stream was aluminum and zircalloy cladding waste from the REDOX process;
- (3) the tertiary stream was slurry product from the evaporator; and
- (4) other waste was primarily decontamination waste from the T Plant containing mainly 0.24M NaNO₂ solution.

D3.0 ZIRCALLOY DECLADDING WASTE PROCESS RECORDS

Serne et al. (1996, Section 9.3.2) provides a description of waste transfers from PUREX. Zircalloy decladding waste was discharged between 1983 and 1990 to DSTs AW-103 and AW-105. During this period records show that 8.64 kg of plutonium and 11,697 kg of uranium were transferred to these two tanks. The transfers are described as follows:

Decladding waste from the PUREX dissolvers was sampled in tanks D2 and D3 before centrifuging or in tank E5 after centrifuging, nitrate addition, and neutralization. These tanks were agitated and duplicate samples were obtained for plutonium, uranium and pH analysis. For samples taken from TK-D2 and -E3, solids were allowed to settle and the liquid analyzed for plutonium and uranium. For samples taken from TK-E5 after the waste had been discharged and neutralized, only sample solution and acid soluble fines were analyzed with the solution. Some plutonium and/or uranium as fine particulate and

insoluble particulates may not have been accounted for by these sampling and analytical procedures. On verification that all batches contained less than 500 g plutonium, that the plutonium content was less than 0.013 g/L, and that the pH was greater than 12, the waste was transferred to the tank farm receiver tanks AW-103 and AW-105. Tank transfers were affected by steam jet, which resulted in about a 5 vol% water increase for each transfer.

The total volume of decladding waste transferred to DST AW-105 was 8,590,000 L, of which 729,550 L was sludge. Much of this liquid was later removed, but all of the sludge remains in the tank and comprises 68.8 vol% of the sludge in this tank. Averaged over the entire volume of all waste leaving PUREX, the uranium concentration was determined to be 0.525 g/L and the plutonium concentration was 0.00039 g/L. The contents in sludge were projected to be 6.48 g/L of uranium and 0.0048 g/L of plutonium.

According to Whyatt et al. (1996), zircalloy decladding waste contains 1.4 times as much iron and 7.1 times as much zirconium than required to ensure subcriticality for the quantity of plutonium present when homogenized. The quantity of uranium is reported to be 1,346 times larger than the quantity of plutonium. This information is shown in Table D-1.

Table D-1. Absorber/Plutonium Ratios and Subcritical Fractions
for Zircalloy Decladding Waste. ¹

Absorber	Absorber/Plutonium mass ratio	Minimum subcritical mass ratio	Actual-to-minimum subcritical fraction
Uranium	1,346	---	---
Iron	223-300	160	1.4
Zirconium	28,200	4,000	7.1

Note:

¹Whyatt et al. (1996)

D4.0 MASS RATIOS AND SUBCRITICAL FRACTIONS

D4.1 HANFORD DEFINED WASTE SUBCRITICAL FRACTION

Using process records, Agnew (1995) compiled compositions of Hanford wastes that define the HDW model. Agnew (1995) determined the sum of the insoluble absorber actual-to-minimum subcritical fractions to be 18.6 and the sum of subcritical fractions for the soluble components to be 54.4. A fraction greater than 1.0 ensures subcriticality in a homogeneous solution.

D4.2 TANK WASTE RETRIEVAL STUDY SUBCRITICAL FRACTIONS

Table D-2 shows component-to-minimum subcritical fractions provided by Whyatt et al. (1996) from analysis of a core. For iron and zirconium, process records provides values which are 2 and 5 times larger than the corresponding value based upon analysis of core samples. For process records, the sum of subcritical fractions for these two components is 8.4, as compared to 2.1, based on analysis of core samples.

Based upon process records, Whyatt et al. (1996) concludes that zirconium is the most accurately known constituent in the waste stream sent to DST AW-105. These records show there to be 7.1 times as much zirconium as is required to maintain subcriticality.

No actual-to-minimum subcritical fraction was found for uranium because the ^{235}U enrichment was not reported. Without including uranium, the sum of the subcritical fractions is 2.72. The sum of fractions obtained from the analysis of a core sample is significantly smaller than the sum reported by Agnew (1995). The largest mass fractions for individual elements are 1.28 for zirconium, 0.78 for iron, and 0.46 for lanthanum.

Table D-2. Component-to-Minimum Subcritical Fractions.¹

Component	Fraction Based on Process Records	Fraction Based on Core Sample Analysis
Zirconium	7.1	1.28
Iron	1.4	0.78
Lanthanum	---	0.46
Chromium	---	0.14
Aluminum	---	0.06
Sum of fractions	8.4	2.72

Note:

¹From Whyatt et al. (1996), Table A.32)

Whyatt et al. (1996) compared process record sludge concentration projections with actual tank sample analysis and found some significant differences:

As determined by AW-103 and AW-105 sludge analysis, the plutonium content of the accumulated sludge is significantly higher than shown by NMC (Nuclear Management Control) records of zirconium decladding waste batches transferred to these tanks during the 1980s. The zirconium content, as determined by sludge analysis and volume, is about a factor of two higher than was expected based on the total zirconium determined to be

added to the two tanks from fuel elements processed in PUREX. The actual masses of fuel processed through the PUREX Plant are known, so actual quantities are known for the zirconium in decladding waste. This makes the zirconium the most accurately known constituent in the waste stream.

Differences in plutonium content between the plant sending and the tank farm receiving the waste may be explained by sampling uncertainties in the plant batch-sampling procedures. The fines might not always have been representatively collected and analyzed. When the NMC decladding waste samples were obtained from Tanks TK-D2 and TK-E3, collected solids were not analyzed. The plant centrifuge was assumed to have removed the solids before the waste was transferred to the TK-E5 neutralization tank. Starting in March 1994, when NMC samples were obtained from TK-E5, any undissolved fines from the samples were not analyzed. Some difficulties were observed in plutonium sampling by operating personnel and the procedures were changed in 1987 to include analysis of fines.

Because fines were observed to be a concern in this stream, it could also be implied that fines may settle out of the supernatant unevenly in the tank farm receiving tank. The observation that the zirconium content of the sludge is greater than expected by a factor of about two could support a concern that uneven settling may have occurred. The waste was introduced into the tanks from near the top center of the tank through a pipe with an internal diameter of 0.957 cm angled about 20° from the horizontal and extending less than 0.1 m. The angled pipe was rotated after each batch transfer to distribute the waste in different directions. The discharge rate of about 2.8 L/s would put the slurry at about 8-m radius for a 7-m drop in elevation from the nozzle discharge point.

D4.3 SAFETY ASSESSMENT SUBCRITICAL FRACTIONS

Braun et al. (1994) reports data from eight waste samples taken from DST AW-105. The highest measured plutonium concentration is 0.024 g/L. This is also the highest plutonium concentration reported in *Tank Waste Source Term Inventory Validation* (WHC 1995). Based upon a plutonium concentration of 0.024 g/L, the solids-to-plutonium mass ratio is estimated to be at least 50,000. The fraction of the actual-to-minimum subcritical mass ratio for this waste is estimated to be 64. In other words, the mass of solids is estimated to be 64 times larger than required to ensure subcriticality when homogenized.

Braun et al. provides a listing of the actual-to-minimum subcritical mass fractions for waste components, based upon analyzed concentrations from samples taken from waste storage tanks. These samples are shown in Table D-3. Waste is separated into soluble and insoluble components. One sample shows the sum of the fractions for the insoluble components to only be 0.50. This is too small a fraction to assure subcriticality by the insoluble components alone. However, the soluble fraction is 15, a value far greater than required to assure subcriticality. In addition, the plutonium concentration in all samples is less than 0.025 g/L, a value less than 1% of the minimum required for criticality under the most idealized conditions.

Table D-3 Sample Data From DST AW-105.¹

Identification	RAT-AW-105-1	RAT-AW-105-2B	T-7937	T-7939
Sample Type	Sludge/Composite	Sludge	Composite	Composite
Pu Equiv., g/L	0.013	0.019	0.014	0.019
Insoluble Components				
U/Pu Fraction ²	0.94	0.54	0.71	0.34
Fe/Pu Fraction	0.92	0.13	0.10	0.08
Mn/Pu Fraction	2.4	0.02	---	0.01
Cr/Pu Fraction	0.47	0.08	0.06	0.04
Ni/Pu Fraction	0.14	0.04	0.02	0.03
Insoluble Sum	4.9	0.80	0.88	0.50
Soluble Components				
Al/Pu Fraction	0.26	0.07	0.06	0.05
Na/Pu Fraction	30.	20.	18.	14.
NO ₃ /Pu Fraction	12.	---	5.2	---
Soluble Sum	42.	20.	23.	15.
Identification	T-7940	T-7941	T-7942	T-7946
Sample Type	Composite	Composite	Composite	Composite
Pu Equiv., g/L	0.015	0.024	0.020	0.015
Insoluble Components				
U/Pu Fraction ²	0.56	0.44	0.59	3.5
Fe/Pu Fraction	0.15	0.15	0.11	10.
Mn/Pu Fraction	0.01	0.02	0.60	31.
Cr/Pu Fraction	0.08	0.06	0.18	4.9
Ni/Pu Fraction	0.04	0.06	0.14	0.85
Insoluble Sum	0.85	0.73	1.6	50.
Soluble Components				
Al/Pu Fraction	0.11	0.14	0.38	0.98
Na/Pu Fraction	28.	20.	27.	23.
NO ₃ /Pu Fraction	6.2	7.7	16.	22.
Soluble Sum	34.	28.	43.	46.

Notes:

¹ Braun et al. (1994)² Fraction refers to the component/plutonium mass ratio divided by the minimum subcritical mass ratio. A fraction of 1.0 means an unlimited volume of that component and plutonium will be just subcritical with optimal moderation.

Even if a large fraction of the soluble components were to be removed, it would be unlikely for the plutonium concentration to increase sufficiently for criticality to occur. The areal density of plutonium is 55.9 g/m², a value 46 times smaller than the minimum required for criticality.

D5.0 k_{∞} OF DST AW-105 SLUDGE

Braun et al. discusses the neutron multiplication constant (k_{∞}) for waste samples from DST AW-105 selected because of "their relatively low neutron absorber content." Specifically, the samples selected did not meet the uranium/plutonium or the iron/plutonium minimum subcritical ratios, and the total mass ratio fractions were less than that for most other samples. The compositions of these samples are shown in Table D-4.

Values of k_{∞} were calculated for three conditions:

1. "As Analyzed" Composition: The plutonium concentration, solids composition and the water content are as analyzed. This composition provides a water content of between 70 and 80 wt%. The plutonium concentration varies from 0.013 to 0.024 g/L.
2. "As Analyzed" Composition with Selected Absorbers Removed: The composition is "as analyzed", except that boron, silicon, calcium, and potassium are removed and the solids renormalized to maintain the density unchanged. Because the water content is high and the boron content is small, the removal of these components reduces k_{∞} by only a small amount (i.e., it is not significant).
3. Dry Conditions with Greatly Elevated Plutonium Content: The water content is reduced to 50 g/L (5.5 wt%), and the solids density is increased to 850 g/L to replace the water. The solids density is less than expected for "real" dry sludge. The plutonium concentration is arbitrarily set at 3.0 g/L, a value 100 times greater than the measured value.

Calculations of k_{∞} were made for water content between 600 and 35 g/L. Over this entire range of water content, k_{∞} shows a monotonic increase as the water content is decreased. A water content of 50 g/L represents waste that is drier than considered possible in practice. Therefore, a water content of 50 g/L is conservative when compared to "real" waste.

Results of calculations are summarized in Table D-5. Using the measured water content (normal conditions), k_{∞} is calculated to be less than 0.03. Under hypothetical conditions using a water content reduced to 50 g/L and using a plutonium concentration arbitrarily increased to 3 g/L, k_{∞} is found to increase to 0.38. This is an extreme condition with less water content than should actually ever occur and with a plutonium concentration more than 100 times greater than measured. Even under these hypothetical conditions, the waste remains subcritical.

Table D-4. Compositions of Selected Samples from DST AW-105.¹

Sample ID	RAT-AW105-1	T-7941	T-7942
Primary Parameters (g/L)			
Plutonium	0.0128	0.0238	0.0198
Water	827.0	764.0	747.0
Solids Density	211.8	259.2	314.2
Waste Solids Components (g/L)			
Uranium	9.3	8.04	9.02
Iron	1.88	0.564	0.352
Manganese	1.00	0.013	0.381
Silicon	4.97	6.97	3.17
Sodium	137.	174.	189.
Aluminum	2.99	2.94	6.75
Boron	0.499	0.697	0.352
Chromium	0.673	0.201	0.818
Nickel	0.188	0.161	0.196
Potassium	10.5	8.72	3.95
Nitrate	36.5	44.2	76.5
Nitrite	5.48	12.5	23.4

Note:

¹Braun et al. (1994)Table D-5. k_w Calculated for Analyzed Waste Sample Compositions.¹

Sample ID	RAT-AW105-1	T7941	T7942
"As Analyzed" Composition			
Plutonium, g/L	0.0128	0.0238	0.0198
Water, g/L	827	764	747
Solids, g/L	212	259	314
k_w	<0.02	<0.02	0.014
Water content reduced to minimum (50 g/L) Plutonium concentration arbitrarily increased to 3 g/L			
Plutonium, g/L	3	3	3
Water, g/L	50	50	50
Solids, g/L	850	850	850
k_w	0.25	0.23	0.38

Note:

¹Braun et al. (1994)

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APPENDIX E

CHEMISTRY

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APPENDIX E

CHEMISTRY

Chemical processes capable of changing uranium and/or plutonium concentrations are of concern to criticality safety. Studies of chemical processes capable of increasing or inhibiting the segregation of uranium and plutonium have been documented. This criticality safety evaluation report summarizes information from chemistry reports that have direct bearing on criticality safety, but does not develop new chemistry information.

In alkaline waste, plutonium solubility is low, and plutonium is bound within solids. Waste sent to tank storage is maintained alkaline to ensure that the plutonium remains combined with the solids. The upper limit of the concentration of dissolved plutonium in liquid waste should not exceed 0.017 g/L (Hobbs et al. 1993).

The origin of K Basin sludge is different from waste already in tank storage. K Basin sludge in its original form is not the product of operations in a separations plant in which the uranium and plutonium has been precipitated, after having first been dissolved. As it resides in the basins, K Basin sludge contains particles (and pieces) of enriched uranium metal with contained plutonium. However, before being sent to tank farms, K Basin sludge will be processed into a form that more closely resembles tank waste.

When waste is in solution, a number of chemical processes are possible for separating the plutonium from other components. For this reason, criticality safety requires that tank waste be maintained alkaline with a pH of at least 8. Normally, the pH is maintained at 12, or greater, to prevent corrosion. For this evaluation, separation of plutonium from uranium is assumed to occur.

E1.0 PLUTONIUM CHEMISTRY

Waste sent to tank farms in the past was the product of processes in which the uranium and plutonium was first dissolved and then precipitated. Evaluations of plutonium chemistry related to tank waste is documented by:

- *Tank Farm Nuclear Criticality Review* (Bratzel et al. 1996);
- *Fluid Dynamics, Particulate Segregation, Chemical Processes, Natural Ore Analog and Tank Inventory Discussions that Relate to the Potential for Criticality in Hanford Tanks* (Serne et al. 1996);
- *The Potential for Criticality in Hanford Tanks Resulting from Retrieval of Tank Waste* (Whyatt et al. 1996).

These documents provide a comprehensive review of the chemistry of waste from plants that extracted plutonium from reactor fuel.

Three Russian chemistry studies related to Hanford Site tank waste are documented by:

- *Plutonium(IV) Precipitates Formed in Alkaline Media in the Presence of Various Anions* (Krot et al. 1998);
- *Interaction of Pu(IV,VI) Hydroxides/Oxides with Metal Hydroxides/Oxides in Alkaline Media* (Fedoseev et al. 1998);
- *Purification of Alkaline Solutions and Wastes from Actinides and Technetium by Coprecipitation with Some Carriers Using the Method of Appearing Reagents* (Peretrukin et al. 1998).

E1.1 TANK FARM NUCLEAR CRITICALITY REVIEW

Bratzel et al. (1996) concludes that "no physical or chemical phenomena or mechanism has been identified that could concentrate fissile material at sufficient quantities or concentrations to result in an accidental nuclear criticality." This report identifies three potential chemical mechanisms that cause plutonium to enter the sludge:

- Sorption onto precipitated hydrated metal oxide sludge particles
- Precipitation as pure plutonium oxide crystals
- Formation of solid-solid solutions with non-neutron absorbers such as Zr(IV).

The importance of these mechanisms is described as follows:

If sorption were the dominant chemical mechanism, bonding of plutonium to strong neutron-absorbing metals such as iron and manganese ensures subcritical conditions. If pure hydrous plutonium oxide formed as a crystalline precipitate when acidic wastes were made alkaline, the plutonium oxide particles would be expected to agglomerate or mix with neutron absorbers. For nuclear criticality to occur for either of the latter cases, large (tens of micrometers in size) plutonium particles free of neutron absorbers must be created and then concentrated by some mechanism. All literature data, as well as calculations performed for modeling operational mechanisms such as salt well pumping, air lift circulators, and transfer pumps, indicate that creation and separation of such plutonium-bearing particles are not plausible.

Bratzel et al. (1996) states that "because other insoluble oxyhydroxides precipitated in great excess with plutonium oxyhydroxide, coprecipitation of plutonium with the sludge-forming solids occurred." Of several likely coprecipitation mechanisms, the most

important is "believed to be sorption of plutonium oxyhydroxide on the solid metal oxyhydroxides. In particular, the oxyhydroxides of iron and aluminum develop large, amorphous surfaces as they are formed that attract transition metal ions (Laitinen 1960)."

Bratzel et al. (1996) describes chemical mechanisms for concentrating the plutonium in a waste tank as follows:

Any chemical mechanism for concentrating the plutonium in a waste tank requires the transport of plutonium from various solid phases to an aqueous liquid phase and then to a compact geometry of a solid phase. Sorbed plutonium must be desorbed into the aqueous phase of the mixture of solid and liquid phases in the waste tanks. However, plutonium sorption on oxyhydroxides may be irreversible under certain chemical environments (DOE 1988, Alberts and Orlandini 1981) unless the solution composition in contact with the plutonium is dramatically altered. Such alteration requires the addition of organic or inorganic plutonium complexants, large changes in the REDOX potential of the solution by addition of oxidants or reductants, or by acidification of the solution.

Potential complexing agents for plutonium are present in all the tanks. Organic complexing agents such as EDTA, HEDTA, nitrilotriacetate, citrate, oxalate, and others are in the various waste mixtures. Even though these compounds are powerful complexants for plutonium in weakly acidic solutions, they are not very effective at the high hydroxide ion concentrations of actual waste supernatant liquids (Delegard et al. 1984). Under alkaline conditions, the organic reagents cannot compete strongly with either hydroxide or carbonate ion bonding to plutonium ions. Anionic complexes of Pu(IV) with hydroxide and carbonate ion have been shown to form under conditions expected in waste tank supernatant liquors. With excess nitrite ions the proposed aqueous species are $\text{Pu}(\text{OH})_2(\text{CO}_3)_2^{(2-)}$ and $\text{Pu}(\text{OH})_4(\text{CO}_3)_2^{(4-)}$. These species have only very low solubility in waste tank supernatant liquids so they are not likely to cause significant plutonium desorption.

The supernatant liquids in the underground tanks contain high concentrations of dissolved nitrite ions, relative to plutonium concentrations, that act as a mild reducing agent, maintaining soluble plutonium almost exclusively as Pu(IV). Because Pu(IV) is sorbed more strongly or is more insoluble than either Pu(V) or Pu(VI), the nitrite ions help drive the plutonium to the solid phase. To change the reduction-oxidation potential of the supernatant liquid requires the addition of huge quantities of oxidizing or reducing agents to the tanks. Such additions are not reasonable in view of the tight control maintained over additions of materials to the tanks.

Dissolution of plutonium oxide to yield solutions containing soluble plutonium concentrations above 0.2 g Pu/L, in carbonate solutions at a pH in the range of 9 to 10, would require oxidation of Pu(IV) to either Pu(V) or Pu(VI). A very strong oxidant such as Ag(II), boiling nitric acid, chlorine, permanganate ion, etc., is required to effect such oxidation. Even under carefully controlled laboratory conditions, such oxidation of

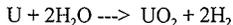
Pu(IV) is difficult to achieve. Thus, no dissolution mechanism is known to produce a sufficiently high plutonium concentration to be of criticality concern.

Acidification of wastes in the underground tanks could cause some dissolution of oxyhydroxides or oxides of plutonium and other metals on which the plutonium may be sorbed and thus result in dissolution of some of the plutonium. Of greater consequence would be the large volume of gases generated by reactions with nitrite ions to produce NO_x gases, with carbonate ions to produce CO_2 , and with the carbon steel tanks to produce hydrogen gas. Acidification would eventually cause failure of the carbon steel pipelines and steel liner, allowing liquid waste to escape into the environment. Because of these disastrous consequences, accidental acidification of the wastes must be avoided.

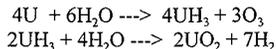
E2.0 URANIUM CHEMISTRY

Uranium in fuel elements is in the form of metal. When uranium metal comes into contact with water, it readily oxidizes into uranium oxide. The oxidation process occurs on the surface of the metal where the oxidized metal sloughs off and falls into the basin water. It is this process which causes deterioration of fuel elements and results in particles and broken pieces of uranium falling away from the elements and becoming part of the sludge on the basin floor or in the bottom of storage canisters. This process reduces the size of uranium metal particles and increases the fraction of particles of micron size. An important chemistry question is to what degree is this process completed over time. Uranium chemistry of K Basin sludge is discussed by Daling et al. (1997).

There are two chemical reactions by which the uranium is changed into uranium oxide. The most direct and also the more common reaction formed is:



The other reaction is:



E2.1 ISOTOPIC SEPARATION OF URANIUM-235 FROM URANIUM-238

^{235}U and ^{238}U are isotopes of the same element and chemically identical. Separation by chemical processes is not possible. Diffusion and other physical separation processes cannot occur during bulk storage. For this evaluation the $^{235}\text{U}/^{238}\text{U}$ ratio is assumed constant, except when batches of uranium having different ratios are combined.

E3.0 IRON CHEMISTRY

Iron content is a primary control for criticality safety. Iron is added to provide sufficient neutron absorption to keep neutron multiplication acceptably low and to prevent plutonium from segregating into regions of high concentration.

E3.1 COPRECIPITATION WITH PLUTONIUM

Uranium and its contained plutonium could be dissolved and coprecipitated with iron before being transferred to tank farms. Coprecipitation of plutonium with iron is a mechanism that ensures that the plutonium will remain associated with iron in a fixed proportion.

Fedoseev et al. (1998) addresses the question of uniformity of Pu(IV) coprecipitated with metals as follows:

Results in the Pu(IV)-Fe(III), Pu(IV)-Co(III), Pu(IV)-Cr(III), Pu(IV)-La(III), and Pu(IV)-U(VI) systems differed from those observed for the Pu(IV)-Ni(II) system. The dissolution rates of the precipitates obtained from NaOH addition to separate equimolar mixtures of Pu(IV) with Fe(III), Co(III), Cr(III), La(III), and U(VI) were considerably higher than those observed for pure $\text{PuO}_2 \cdot x\text{H}_2\text{O}$ treated under the same conditions. Based on these results, coprecipitation of Pu(IV) with the various metal ions apparently produces the corresponding hydroxides in which plutonium is distributed uniformly (on a molecular level) within the solids phases.

E3.2 FORMATION OF AGGLOMERATES WITH PLUTONIUM

Agglomeration is a mechanism in tank waste that ensures that plutonium will remain associated with other waste components. Agglomeration between plutonium and iron occurs when particle size is small. When particles agglomerate, they form larger particles composed of a random assortment of primary particles. If primary particles of differing types are uniformly mixed, the secondary agglomerated particles will reflect the original uniform distribution. A restriction on particle size therefore applies only to primary particles.

Formation of large masses of agglomerated particles greatly increases the difficulty of separating any waste component into a more highly concentrated form. When plutonium is agglomerated with iron, its relative proportion to iron will remain unchanged no matter how large the secondary particles and no matter how fine the agglomerates may be ground later.

E4.0 SEGREGATION BY PARTICLE SIZE

Particle size is of interest to criticality safety for two reasons. First, for uranium a larger particle size produces a smaller minimum critical mass. In addition larger particles of any waste component can lead to stratification within settled layers of solids.

^{238}U is a strong resonance absorber for epithermal neutrons. When neutrons are moderated outside of the uranium, they escape resonance capture, and the fraction of neutrons reaching thermalization can be increased. When uranium is in the form of pieces, neutrons may leave the uranium and be moderated in the intervening water. These neutrons reenter uranium pieces as thermal neutrons with a higher probability of fissioning ^{235}U atoms. The critical mass for low enriched uranium in optimally sized rods can be made smaller than is possible for powder or solution.

A definition of "small" for uranium particles to ensure that critical parameters for a homogeneous system applies is a diameter of less than 0.13 cm (1,300 μm). A maximum particle dimension of 0.13 cm (0.05 in.) is adequately small to ensure that mixtures and solutions containing 0.947% enriched uranium can not be made critical, even with optimal moderation. This small particle size, however, may not completely ensure subcriticality for 1.25 wt% enriched uranium.

The ability to concentrate particles during agitation and/or mixing is related to particle size and density. After dense particles are suspended in a liquid, as might occur during mixing or pumping, and then allowed to settle undisturbed, larger particles will more rapidly settle. The resulting settled configuration might therefore consist of layers having a higher concentration of some waste components. The possibility is considered that this reconfiguration might result in a high concentration of fissile components, thereby achieving an unacceptably high neutron multiplication constant.

In a study of tank waste chemistry Whyatt et al. reports that an increase in fissile concentration by a factor of 2.5 is possible as a result of segregation of waste components according to differences in particle size and density. This mechanism is based on the conservative assumption that plutonium might preferentially associate with one size particle. For their study they divided waste into nine size ranges, and using the TEMPEST code simulated mixing and allowing the waste to settle. With full 3-dimensional modeling they concluded that "solids being pumped out of C-106, of those same solids settling in AY-102, and of mixer pump operation in SY-102 show no indication of unusual segregation in any area of the tank. The greatest degree of solids enrichment in all simulations was about a factor of 2.5 for the largest solids in the distribution." This conclusion is based upon the maximum particle size being less than 50 μm . Above 50 μm the range of possible segregation increases considerably and arguments against segregation are difficult to make. Based upon Whyatt et al., a primary particle size less than 10 μm provides a high assurance that very little increase in plutonium concentration will occur as a result of gravity segregation.

Whyatt et al. states that "most gravity separation devices used in the mining industry are effective at recovering minerals with particle sizes down to approximately 100 μm ." Devices are available which can separate 10 μm particles, but the effectiveness of separation decreases rapidly below this size.

Daling et al. (1997) discusses particle size as it relates to criticality safety of K Basin sludge and the conditions necessary to provide assurance that separation will not occur. The following conclusions are stated for the mixing of uranium and iron:

There is significant potential for segregation of precipitated iron from untreated sludge. Elimination of metallic uranium will reduce the extent of segregation but is not sufficient to avoid segregation. The specific size to which reduction must be achieved will depend on how much dilution with iron is considered acceptable. However, the size is clearly less than 177 μm and is estimated to be 50 μm or less. Reduction to some size less than 10 μm would allow agglomeration arguments to be made which could assure that a safe ratio is maintained with the initial 3 times the safe ratio iron addition.

Based on these observations, it is concluded that a 3-fold increase in plutonium concentration bounds the spectrum of possible degrees of segregation in storage. If one assumes negligible interaction with DST AW-105 sludge, the quantity of neutron absorbing solids must be 3.0 times that required to ensure compliance to the USL.

Small particles are more likely to form agglomerates. In agglomerated material the uranium will remain in a fixed relationship to other solid material and is less likely to increase in concentration during waste transfers or processing. Solids combined with the uranium establish an upper limit on the uranium concentration and, at the same time, provide neutron absorption. The formation of agglomerations, therefore, ensures a larger margin of safety. It is the size of the primary particles from which the agglomerate is formed that determines the range of segregation possible. The size of agglomerated particles has no importance to component segregation in mixing and settling processes.

Small particles of uranium metal will oxidize to uranium oxide. This leads to a slight increase in the minimum critical mass. Oxidation occurs only on the surface of uranium. The greatest surface area occurs with finely divided particles. Upon oxidizing, uranium sloughs off fuel as finely divided particles. The process of oxidation transforms solid pieces of uranium metal fuel into finely divided particles for which critical parameters for homogenous uranium solutions apply. With oxidized uranium it is virtually impossible to achieve high-density lumps. The transformation of uranium metal to uranium oxide increases the minimum critical mass and the margin of safety. Oxidation is therefore helpful in ensuring subcriticality under all credible conditions.

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APPENDIX F

IRRADIATION AND CRITICAL MASS

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APPENDIX F

IRRADIATION AND CRITICAL MASS

This appendix describes the relationship between degree of irradiation in a reactor and critical parameters.

F1.0 IRRADIATION EFFECTS ON CRITICAL MASS

Toffer (1976) calculated the critical mass for 0.95 wt% and 1.25 wt% N Reactor fuel as a function of irradiation history. According to Toffer (1976), the critical mass of uranium in KE and KW Basin sludge would be expected to be at least 30% larger than the critical mass for unirradiated (green) fuel.

On the surface of fuel elements composed of low enriched uranium, plutonium is produced faster than ^{235}U is burned. However, as the $^{235}\text{U}/^{238}\text{U}$ ratio increases, the ratio of plutonium production to ^{235}U burnup decreases. Schwinkendorf (1997, Appendix G) looks at the radial isotopic evolution during burnup of 0.95 wt% and 1.25 wt% enriched uranium and reaches the following conclusion:

In all burnup calculations reported in this appendix, the production of plutonium is enhanced at the outer surfaces of the fuel; there is increased exposure near the surfaces because of self-shielding. However, this effect is at least partially mitigated by the fact that enhanced fissile uranium depletion also occurs near the fuel surfaces. In addition, the increased plutonium production near the surface also has a higher ^{240}Pu content, again, because of the increased exposure at the surface. The number of neutrons produced per fission is higher for ^{239}Pu than ^{235}U , and so the plutonium is worth more than the fissile uranium, but the effect is not large. Scrap material composed of the outer skin of the fuel is more reactive than the average fuel, but not to a significant degree.

Based only upon consideration of the concentrations of ^{235}U and ^{239}Pu , the outer 0.05-cm thick layer of each 0.95 wt% element is found to increase in reactivity worth during irradiation. However, below the surface of the element the fissile concentration decreases with fuel exposure. For the fuel element as a whole, the loss of ^{235}U is 1.6 to 1.8 times greater than the production of ^{239}Pu . The cladding of the fuel mitigates the preferential loss of the surface layer. Since considerable interior uranium would have to be removed to permit removal of the surface layer, sludge does not contain an elevated fraction of surface uranium. For 1.25 wt% enriched uranium, the sum of the ^{235}U and ^{239}Pu concentrations does not exceed that of green fuel at any radial position, regardless of exposure time.

Relative worth of fuel after irradiation compared to before irradiation depends on: (1) ^{235}U depletion, (2) plutonium production, (3) ^{240}Pu content, and (4) production of neutron absorbing fission products. Because of difficulty in determining concentrations and ensuring their

continued presence, fission products are usually not taken into account. However, fission products have a significant neutron absorption cross section, and it is conservative to ignore their effect.

Schwinkendorf (1997) calculates the maximum k_{∞} as a function of reactor exposure time for a lattice of uranium rods in water. Reactivity of 1.25 wt% enriched uranium decreases monotonically during irradiation. His calculations show that k_{∞} for 0.95 wt% enriched uranium remains nearly unchanged for the first 100 days of irradiation, but is always less reactive than green uranium.

F2.0 EFFECT OF PLUTONIUM-240 ON CRITICAL MASS

Plutonium consists primarily of the isotopes ^{239}Pu and ^{240}Pu . The low concentrations of other plutonium isotopes ensure that they have only a small impact on criticality safety. For evaluation the quantities of ^{241}Pu and ^{238}Pu are usually added to the ^{239}Pu inventory.

^{239}Pu and ^{240}Pu are very different in their ability to support a self-sustaining neutron chain reaction. ^{240}Pu cannot be made critical with thermal neutrons, and its minimum critical mass is much larger than that of ^{239}Pu . Because criticality with ^{240}Pu is only possible with fast neutrons all moderators would have to be removed for it to become critical. For thermal neutrons ^{240}Pu has a large absorption cross section and will not fission. Conditions which create a higher fraction of fast neutrons make ^{240}Pu more reactive, but these are the conditions which make ^{239}Pu less reactive. For these reasons, ^{240}Pu acts as a neutron absorber, rather than a contributor to criticality.

Table F-1 shows the relationship between ^{240}Pu content and critical mass. For every 1% increase in ^{240}Pu the plutonium critical mass increases by at least 4.0% (Hansen and Clayton 1969). When the ^{240}Pu content exceeds 15%, every 1% increase in ^{240}Pu causes the plutonium critical mass to increase by 5.0%. The plutonium minimum critical mass increases from 520 g for 0.0 wt% ^{240}Pu to 740 g for 10 wt% ^{240}Pu and to 1090 g for 20 wt% ^{240}Pu (Carter et al. 1969).

Table F-1. Effect of ²⁴⁰Pu on Plutonium Critical Mass.

Parameter	²³⁹ Pu: 100%	²³⁹ Pu: 95% ²⁴⁰ Pu: 5%	²³⁹ Pu: 90% ²⁴⁰ Pu: 10%	²³⁹ Pu: 85% ²⁴⁰ Pu: 15%	²³⁹ Pu: 80% ²⁴⁰ Pu: 20%
% change in critical mass per % change in ²⁴⁰ Pu ¹	---	4.0 (average 0 to 5%)	4.2 (average 0 to 10%)	5.0 (average 0 to 15%)	5.5 (average 0 to 20%)
Minimum Critical Mass ²	520 g	620 g	740 g	910 g	1090 g
²³⁵ U Equivalence ³	1.57	1.32	1.11	0.90	0.75

Notes:

¹Hansen and Clayton (1969)²Critical masses obtained from *Criticality Handbook* (Carter et al. 1969).³Minimum critical mass of ²³⁵U divided by plutonium minimum critical mass.

An ²³⁵U equivalence for ²³⁹Pu is found by dividing the minimum critical mass of 820 g for ²³⁵U by the minimum critical mass of plutonium. When plutonium contains no ²⁴⁰Pu, this ratio is 820/520, and the ²³⁵U equivalence is 1.57. In other words, if a gram of plutonium is replaced by 1.57 g of ²³⁵U, k_{∞} should remain unchanged.

F3.0 REFERENCES

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APPENDIX G
COMPUTER INPUT FILES

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APPENDIX G

COMPUTER INPUT FILES

Information is provided to document calculations and to permit reconstructing input listings for selected cases. For each waste composition calculations were made for 16 water contents from dry to saturation. Only the calculations for the water content that produced the maximum k_{∞} is shown. Maximum k_{∞} values are shown in this appendix with a higher precision than can be read from graphs.

Homogeneous configurations of infinite extent are calculated. A complete listing is provided for Case F1A-8, but for subsequent cases an abbreviated listing is provided showing only lines that are different. Lines that change are those that provide material densities or are derived from material densities. Lines shown after the case identification and before the "=nitawl" line are descriptive and not actually part of the input listing. An apostrophe (') in column 1 denotes a comment line. The XSDRNPM input format was developed for punch cards (30 years ago), and the number of characters on a line is limited to 72. A description of NITAWL and XSDRNPM input parameters is found in NUREG/CR-0200 (ORNL 1995) which documents the SCALE code package.

k_{∞} AS FUNCTION OF ^{235}U ENRICHMENT (NO PLUTONIUM OR IRON)

CASE F1A-8 Calculated $k_{\infty} = 1.06145$ (maximum value)
 1.25 wt% ^{235}U enriched uranium oxide in water. Infinite homogeneous system.
 2000 g UO_2/L . No plutonium. No iron. 350 g water/L.
 Case F1A-8 input is complete. Subsequent cases are abbreviated.

```
=nitawl
0$$ 82 e
1$$ a2 7 a8 4 e 1t
2$$ 1001 8016 26000 92235 92238 94240 94239
3**
 92235 293. 0 0.0 0.0 0.0 5.651e-05 1 1.008 8.455e+03 1
 16.068 1.534e+03 1 1.
 92238 293. 0 0.0 0.0 0.0 4.408e-03 1 1.008 1.082e+02 1
 16.068 1.967e+01 1 1.
 94240 293. 0 0.0 0.0 0.0 1.000e-08 1 1.008 1.000e+08 1
 16.068 1.000e+08 1 1.
 94239 293. 0 0.0 0.0 0.0 1.000e-08 1 1.008 1.000e+08 1
 16.068 1.000e+08 1 1.

2t
end
=xsdrrn
Case F1A-8, U(1.25) Oxide, 2000 g/L, FeO/UO2=0.0, H2O=350 g/L
1$$ 3 1 32 1 1 1 7 16 1 1 20 10 0 0 0
2$$ a7 -1 e
3$$ 1 a9 3 1 e
4$$ 0 4 20 -1 7 e
5** 2r1.-5 e 1t
13$$ 1 1 1 1 1 1 1
 1
  hydrogen oxygen iron U235 U238 Pu240 Pu239
14$$ 1001 8016 26000 92235 92238 94240 94239
```

```

15** 2.342-2 2.062-2 1.000-8 5.651-5 4.408-3 1.000-8 1.000-8
16$$ 11001 18016 126000 192235 192238 194240 194239
18## 6HH-1 6HO-16 6HFe-26 6HU-235 6HU-238 6HPu-240 6HPu-239 2T
33## f1 4t
35** 31i0.0 16.0
36$$ f1
49$$ 92235 92235 92238
50$$ 18 27 27
51$$ 5r1 5r2 5r3 12r4 5t
end
    
```

CASE F2A-7 Calculated k_{∞} = 0.96708 (maximum value)
 0.95 wt% ²³⁵U enriched uranium oxide. 2000 g UO₂/L. 300 g water/L.

```

=nitawl
3**
92235 293. 0 0.0 0.0 0.0 4.295e-05 1 1.008 9.536e+03 1 16.068 1.854e+03 1 1.
92238 293. 0 0.0 0.0 0.0 4.422e-03 1 1.008 9.263e+01 1 16.068 1.802e+01 1 1.
=xsdrn
hydrogen oxygen iron U235 U238 Pu240 Pu239
15** 2.008-2 1.897-2 1.000-8 4.295-5 4.422-3 1.000-8 1.000-8
    
```

CASE F3A-7 Calculated k_{∞} = 0.92332 (maximum value)
 0.84 wt% ²³⁵U enriched uranium oxide. 2000 g UO₂/L. 300 g water/L.

```

=nitawl
3**
92235 293. 0 0.0 0.0 0.0 3.798e-05 1 1.008 1.078e+03 1 16.068 2.098e+03 1 1.
92238 293. 0 0.0 0.0 0.0 4.427e-03 1 1.008 9.252e+01 1 16.068 1.799e+01 1 1.
=xsdrn
hydrogen oxygen iron U235 U238 Pu240 Pu239
15** 2.008-2 1.897-2 1.000-8 3.798-5 4.427-3 1.000-8 1.000-8
    
```

**k_{∞} AS FUNCTION OF IRON CONTENT
 (0.20 wt% PLUTONIUM)**

CASE F7A-8 Calculated k_{∞} = 1.01820 (maximum value)
 0.84 wt% ²³⁵U enriched uranium oxide in water. Infinite homogeneous system.
 0.20 wt% Pu. ²⁴⁰Pu/Pu = 11.0 wt% 2000 g UO₂/L. 350 g water/L.
 Mass Ratios: FeO/(U+Pu)O₂ = 0.00 Fe/(U+Pu) = 0.00

```

=nitawl
3**
92235 293. 0 0.0 0.0 0.0 3.790e-05 1 1.008 1.261e+04 1 16.068 2.287e+03 1 1.
92238 293. 0 0.0 0.0 0.0 4.418e-03 1 1.008 1.082e+02 1 16.068 1.962e+01 1 1.
94240 293. 0 0.0 0.0 0.0 9.739e-07 1 1.008 4.906e+05 1 16.068 8.701e+04 1 1.
94239 293. 0 0.0 0.0 0.0 7.913e-06 1 1.008 6.039e+04 1 16.068 1.096e+04 1 1.
=xsdrn
hydrogen oxygen iron U235 U238 Pu240 Pu239
15** 2.342-2 2.062-2 1.000-8 3.790-5 4.418-3 9.739-7 7.913-6
    
```

CASE F7D-6 Calculated k_{∞} = 0.71340 (maximum value)
 0.84 wt% ²³⁵UO₂; 0.20 wt% Pu; ²⁴⁰Pu/Pu = 11.0 wt%; 2000 g UO₂/L; 250 g water/L.
 Mass Ratios: FeO/(U+Pu)O₂ = 0.600 Fe/(U+Pu) = 0.529

```

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3**
92235 293. 0 0. 0. 0. 2.369E-05 1 1.008 1.441E+04 1 16.00 3.587E+03 1 1.
92238 293. 0 0. 0. 0. 2.761E-03 1 1.008 1.236E+02 1 16.00 3.078E+01 1 1.
94240 293. 0 0. 0. 0. 6.087E-07 1 1.008 5.607E+05 1 16.00 1.396E+05 1 1.
94239 293. 0 0. 0. 0. 4.946E-06 1 1.008 6.901E+04 1 16.00 1.718E+04 1 1.
=xsdrn
hydrogen oxygen iron U235 U238 Pu240 Pu239
    
```

15** 1.673E-02 2.023E-02 6.287E-03 2.369E-05 2.761E-03 6.087E-07 4.946E-06

CASE F7E-6 Calculated k_{∞} = 0.65018 (maximum value)
 0.84 wt% $^{235}\text{UO}_2$; 0.20 wt% Pu; $^{240}\text{Pu}/\text{Pu}$ = 11.0 wt%; 2000 g UO_2/L ; 250 g water/L.
 Mass Ratios: $\text{FeO}/(\text{U}+\text{Pu})\text{O}_2$ = 0.800 $\text{Fe}/(\text{U}+\text{Pu})$ = 0.705

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3**

92235	293.	0	0.	0.	0.	2.106E-05	1	1.008	1.621E+04	1	16.00	4.144E+03	1	1.
92238	293.	0	0.	0.	0.	2.454E-03	1	1.008	1.391E+02	1	16.00	3.556E+01	1	1.
94240	293.	0	0.	0.	0.	5.411E-07	1	1.008	6.308E+05	1	16.00	1.613E+05	1	1.
94239	293.	0	0.	0.	0.	4.396E-06	1	1.008	7.764E+04	1	16.00	1.985E+04	1	1.

=xsdrn

	hydrogen	oxygen	iron	U235	U238	Pu240	Pu239
15**	1.673E-02	2.078E-02	7.452E-03	2.106E-05	2.454E-03	5.411E-07	4.396E-06

**k_{∞} AS FUNCTION OF IRON CONTENT
 (0.30 wt% PLUTONIUM)**

CASE F9A-9 Calculated k_{∞} = 1.03419 (maximum value)
 0.84 wt% ^{235}U enriched uranium oxide in water. Infinite homogeneous system.
 0.30 wt% Pu. $^{240}\text{Pu}/\text{Pu}$ = 16.0 wt% 2000 g UO_2/L . 400 g water/L.
 Mass Ratios: $\text{FeO}/(\text{U}+\text{Pu})\text{O}_2$ = 0.000 $\text{Fe}/(\text{U}+\text{Pu})$ = 0.000

=nitawl

3**

92235	293.	0	0.0	0.0	0.0	3.786e-05	1	1.008	1.442e+04	1	16.068	2.475e+03	1	1.
92238	293.	0	0.0	0.0	0.0	4.413e-03	1	1.008	1.237e+02	1	16.068	2.123e+01	1	1.
94240	293.	0	0.0	0.0	0.0	2.215e-06	1	1.008	2.570e+05	1	16.068	4.410e+04	1	1.
94239	293.	0	0.0	0.0	0.0	1.120e-05	1	1.008	4.875e+04	1	16.068	8.366e+03	1	1.

=xsdrn

	hydrogen	oxygen	iron	U235	U238	Pu240	Pu239
15**	2.677-2	2.231-2	1.000-8	3.786-5	4.413-3	2.215-6	1.120-5

CASE F9D-6 Calculated k_{∞} = 0.74536 (maximum value)
 0.84 wt% $^{235}\text{UO}_2$; 0.30 wt% Pu; $^{240}\text{Pu}/\text{Pu}$ = 16.0 wt%; 2000 g UO_2/L ; 250 g water/L.
 Mass Ratios: $\text{FeO}/(\text{U}+\text{Pu})\text{O}_2$ = 0.600 $\text{Fe}/(\text{U}+\text{Pu})$ = 0.529

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3**

92235	293.	0	0.0	0.	0.	2.366E-05	1	1.008	1.442E+04	1	16.00	3.591E+03	1	1.
92238	293.	0	0.0	0.	0.	2.758E-03	1	1.008	1.237E+02	1	16.00	3.081E+01	1	1.
94240	293.	0	0.0	0.	0.	1.328E-06	1	1.008	2.570E+05	1	16.00	6.399E+04	1	1.
94239	293.	0	0.0	0.	0.	7.002E-06	1	1.008	4.875E+04	1	16.00	1.214E+04	1	1.

=xsdrn

	hydrogen	oxygen	iron	U235	U238	Pu240	Pu239
15**	1.673E-02	2.023E-02	6.287E-03	2.366E-05	2.758E-03	1.328E-06	7.002E-06

CASE F9E-6 Calculated k_{∞} = 0.68395 (maximum value)
 0.84 wt% $^{235}\text{UO}_2$; 0.30 wt% Pu; $^{240}\text{Pu}/\text{Pu}$ = 16.0 wt%; 2000 g UO_2/L ; 250 g water/L.
 Mass Ratios: $\text{FeO}/(\text{U}+\text{Pu})\text{O}_2$ = 0.800 $\text{Fe}/(\text{U}+\text{Pu})$ = 0.705

=nitawl

3**

92235	293.	0	0.0	0.	0.	2.104E-05	1	1.008	1.623E+04	1	16.00	4.149E+03	1	1.
92238	293.	0	0.0	0.	0.	2.452E-03	1	1.008	1.392E+02	1	16.00	3.559E+01	1	1.
94240	293.	0	0.0	0.	0.	1.181E-06	1	1.008	2.891E+05	1	16.00	7.392E+04	1	1.
94239	293.	0	0.0	0.	0.	6.224E-06	1	1.008	5.484E+04	1	16.00	1.402E+04	1	1.

=xsdrn

	hydrogen	oxygen	iron	U235	U238	Pu240	Pu239
15**	1.673E-02	2.078E-02	7.452E-03	2.104E-05	2.452E-03	1.181E-06	6.224E-06

**k_{∞} AS FUNCTION OF PLUTONIUM CONTENT
(IRON/HEAVY-METAL=0.529)**

CASE F12E-7 Calculated k_{∞} = 0.84754 (maximum value)
0.84 wt% ^{235}U enriched uranium oxide in water. Infinite homogeneous system.
0.60 wt% Pu. $^{240}\text{Pu}/\text{Pu}$ = 16.0 wt% 2000 g UO_2/L . 300 g water/L.
Mass Ratios: $\text{FeO}/(\text{U}+\text{Pu})\text{O}_2$ = 0.600 $\text{Fe}/(\text{U}+\text{Pu})$ = 0.529

=nitawl

3**

92235	293.	0	0.	0.	0.	2.359E-05	1	1.008	1.736E+04	1	16.00	3.900E+03	1	1.
92238	293.	0	0.	0.	0.	2.750E-03	1	1.008	1.489E+02	1	16.00	3.346E+01	1	1.
94240	293.	0	0.	0.	0.	2.656E-06	1	1.008	1.542E+05	1	16.00	3.464E+04	1	1.
94239	293.	0	0.	0.	0.	1.400E-05	1	1.008	2.925E+04	1	16.00	6.570E+03	1	1.

=xsdrn

	hydrogen	oxygen	iron	U235	U238	Pu240	Pu239
15**	2.008E-02	2.191E-02	6.287E-03	2.359E-05	2.750E-03	2.656E-06	1.400E-05

CASE F12C-9Calculated k_{∞} = 0.92266 (maximum value)0.90 wt% Pu; 0.84 wt% $^{235}\text{UO}_2$; $^{240}\text{Pu}/\text{Pu}$ = 16.0 wt%; 2000 g UO_2/L ; 400 g water/L.

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92235	293.	0	0.	0.	0.	2.352E-05	1	1.008	2.322E+04	1	16.00	4.509E+03	1	1.
92238	293.	0	0.	0.	0.	2.742E-03	1	1.008	1.992E+02	1	16.00	3.868E+01	1	1.
94240	293.	0	0.	0.	0.	3.984E-06	1	1.008	1.371E+05	1	16.00	2.662E+04	1	1.
94239	293.	0	0.	0.	0.	2.100E-05	1	1.008	2.600E+04	1	16.00	5.049E+03	1	1.

=xsdrn

	hydrogen	oxygen	iron	U235	U238	Pu240	Pu239
15**	2.677E-02	2.525E-02	6.287E-03	2.352E-05	2.742E-03	3.984E-06	2.100E-05

CASE F12D-10Calculated k_{∞} = 0.98088 (maximum value)1.20 wt% Pu; 0.84 wt% $^{235}\text{UO}_2$; $^{240}\text{Pu}/\text{Pu}$ = 16.0 wt%; 2000 g UO_2/L ; 450 g water/L.

=xsdrn

	hydrogen	oxygen	iron	U235	U238	Pu240	Pu239
15**	3.012E-02	2.693E-02	6.287E-03	2.345E-05	2.733E-03	5.312E-06	2.801E-05

CASE F12E-11Calculated k_{∞} = 1.02728 (maximum value)1.50 wt% Pu; 0.84 wt% $^{235}\text{UO}_2$; $^{240}\text{Pu}/\text{Pu}$ = 16.0 wt%; 2000 g UO_2/L ; 500 g water/L.

=xsdrn

	hydrogen	oxygen	iron	U235	U238	Pu240	Pu239
15**	3.346E-02	2.860E-02	6.287E-03	2.338E-05	2.725E-03	6.640E-06	3.501E-05

CASE F12F-11Calculated k_{∞} = 1.06530 (maximum value)1.80 wt% Pu; 0.84 wt% $^{235}\text{UO}_2$; $^{240}\text{Pu}/\text{Pu}$ = 16.0 wt%; 2000 g UO_2/L ; 500 g water/L.

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92235	293.	0	0.	0.	0.	2.331E-05	1	1.008	2.929E+04	1	16.00	5.153E+03	1	1.
92238	293.	0	0.	0.	0.	2.717E-03	1	1.008	2.513E+02	1	16.00	4.421E+01	1	1.
94240	293.	0	0.	0.	0.	7.968E-06	1	1.008	8.566E+04	1	16.00	1.507E+04	1	1.
94239	293.	0	0.	0.	0.	4.201E-05	1	1.008	1.625E+04	1	16.00	2.859E+03	1	1.

=xsdrn

	hydrogen	oxygen	iron	U235	U238	Pu240	Pu239
15**	3.346E-02	2.860E-02	6.287E-03	2.331E-05	2.717E-03	7.968E-06	4.201E-05

**k_{∞} AS FUNCTION OF PLUTONIUM CONTENT
(IRON/HEAVY-METAL=0.353)**

CASE F13B-8 Calculated $k_{\infty} = 0.91789$ (maximum value)
 0.84 wt% ^{235}U enriched uranium oxide in water. Infinite homogeneous system.
 0.60 wt% Pu. $^{240}\text{Pu}/\text{Pu} = 16.0$ wt% 2000 g UO_2/L . 350 g water/L.
 Mass Ratios: $\text{FeO}/(\text{U+Pu})\text{O}_2 = 0.400$ $\text{Fe}/(\text{U+Pu}) = 0.353$

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 3**

92235	293.	0	0.	0.	0.	2.696E-05	1	1.008	1.772E+04	1	16.00	3.564E+03	1	1.1.
92238	293.	0	0.	0.	0.	3.143E-03	1	1.008	1.520E+02	1	16.00	3.058E+01	1	1.1.
94240	293.	0	0.	0.	0.	3.036E-06	1	1.008	1.574E+05	1	16.00	3.166E+04	1	1.1.
94239	293.	0	0.	0.	0.	1.600E-05	1	1.008	2.986E+04	1	16.00	6.005E+03	1	1.1.

=xsdrn

	hydrogen	oxygen	iron	U235	U238	Pu240	Pu239
15**	2.342E-02	2.288E-02	4.790E-03	2.696E-05	3.143E-03	3.036E-06	1.600E-05

CASE F13C-9 Calculated $k_{\infty} = 0.98771$ (maximum value)
 0.90 wt% Pu; 0.84 wt% $^{235}\text{UO}_2$; $^{240}\text{Pu}/\text{Pu} = 16.0$ wt%; 2000 g UO_2/L ; 400 g water/L.

=xsdrn

	hydrogen	oxygen	iron	U235	U238	Pu240	Pu239
15**	2.677E-02	2.455E-02	4.790E-03	2.688E-05	3.133E-03	4.553E-06	2.401E-05

CASE F13D-11 Calculated $k_{\infty} = 1.04085$ (maximum value)
 1.20 wt% Pu; 0.84 wt% $^{235}\text{UO}_2$; $^{240}\text{Pu}/\text{Pu} = 16.0$ wt%; 2000 g UO_2/L ; 500 g water/L.

=xsdrn

	hydrogen	oxygen	iron	U235	U238	Pu240	Pu239
15**	3.346E-02	2.790E-02	4.790E-03	2.680E-05	3.124E-03	6.071E-06	3.201E-05

CASE F13E-11 Calculated $k_{\infty} = 1.08269$ (maximum value)
 1.50 wt% Pu; 0.84 wt% $^{235}\text{UO}_2$; $^{240}\text{Pu}/\text{Pu} = 16.0$ wt%; 2000 g UO_2/L ; 500 g water/L.

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3**

92235	293.	0	0.	0.	0.	2.672E-05	1	1.008	2.555E+04	1	16.00	4.385E+03	1	1.1.
92238	293.	0	0.	0.	0.	3.114E-03	1	1.008	2.192E+02	1	16.00	3.762E+01	1	1.1.
94240	293.	0	0.	0.	0.	7.589E-06	1	1.008	8.995E+04	1	16.00	1.544E+04	1	1.1.
94239	293.	0	0.	0.	0.	4.001E-05	1	1.008	1.706E+04	1	16.00	2.929E+03	1	1.1.

=xsdrn

	hydrogen	oxygen	iron	U235	U238	Pu240	Pu239
15**	3.346E-02	2.790E-02	4.790E-03	2.672E-05	3.114E-03	7.589E-06	4.001E-05

**k_{∞} AS FUNCTION OF ^{240}Pu CONTENT
(0.30 wt% PLUTONIUM)**

CASE F14A-6 Calculated k_{∞} = 0.76316 (maximum value)
 0.84 wt% ^{235}U enriched uranium oxide in water. Infinite homogeneous system.
 0.30 wt% Pu. $^{240}\text{Pu}/\text{Pu}$ = 10.0 wt% 2000 g UO_2/L . 250 g water/L.
 Mass Ratios: $\text{FeO}/(\text{U}+\text{Pu})\text{O}_2$ = 0.600 $\text{Fe}/(\text{U}+\text{Pu})$ = 0.529

=nitawl

3**

92235	293.	0	0.	0.	0.	2.366E-05	1	1.008	1.442E+04	1	16.00	3.591E+03	1	1.
92238	293.	0	0.	0.	0.	2.758E-03	1	1.008	1.237E+02	1	16.00	3.081E+01	1	1.
94240	293.	0	0.	0.	0.	8.300E-07	1	1.008	4.112E+05	1	16.00	1.024E+05	1	1.
94239	293.	0	0.	0.	0.	7.502E-06	1	1.008	4.550E+04	1	16.00	1.133E+04	1	1.

=xsdrn

	hydrogen	oxygen	iron	U235	U238	Pu240	Pu239
15**	1.673E-02	2.023E-02	6.287E-03	2.366E-05	2.758E-03	8.300E-07	7.502E-06

CASE F14B-6 Calculated k_{∞} = 0.77865 (maximum value)
 0.84 wt% $^{235}\text{UO}_2$; 0.30 wt% Pu; $^{240}\text{Pu}/\text{Pu}$ = 5.0 wt%; 2000 g UO_2/L ; 250 g water/L.

=xsdrn

	hydrogen	oxygen	iron	U235	U238	Pu240	Pu239
15**	1.673E-02	2.023E-02	6.287E-03	2.366E-05	2.758E-03	4.150E-07	7.918E-06

CASE F14C-6 Calculated k_{∞} = 0.79493 (maximum value)
 0.84 wt% $^{235}\text{UO}_2$; 0.30 wt% Pu; $^{240}\text{Pu}/\text{Pu}$ = 0.0 wt%; 2000 g UO_2/L ; 250 g water/L.

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94240	293.	0	0.	0.	0.	8.300E-10	1	1.008	4.112E+08	1	16.00	1.024E+08	1	1.
94239	293.	0	0.	0.	0.	8.334E-06	1	1.008	4.095E+04	1	16.00	1.020E+04	1	1.

=xsdrn

	hydrogen	oxygen	iron	U235	U238	Pu240	Pu239
15**	1.673E-02	2.023E-02	6.287E-03	2.366E-05	2.758E-03	8.300E-10	8.334E-06

**k_{∞} AS FUNCTION OF ^{235}U ENRICHMENT
(IRON/HEAVY-METAL = 0.539)**

CASE F14D-6 Calculated k_{∞} = 0.71951 (maximum value)
 0.75 wt% ^{235}U enriched uranium oxide in water. Infinite homogeneous system.
 0.30 wt% Pu. $^{240}\text{Pu}/\text{Pu}$ = 16.0 wt% 2000 g UO_2/L . 250 g water/L.
 Mass Ratios: $\text{FeO}/(\text{U}+\text{Pu})\text{O}_2$ = 0.600 $\text{Fe}/(\text{U}+\text{Pu})$ = 0.529

=nitawl

3**

92235	293.	0	0.	0.	0.	2.113E-05	1	1.008	1.615E+04	1	16.00	4.022E+03	1	1.
92238	293.	0	0.	0.	0.	2.761E-03	1	1.008	1.236E+02	1	16.00	3.078E+01	1	1.
94240	293.	0	0.	0.	0.	1.328E-06	1	1.008	2.570E+05	1	16.00	6.399E+04	1	1.
94239	293.	0	0.	0.	0.	7.002E-06	1	1.008	4.875E+04	1	16.00	1.214E+04	1	1.

=xsdrn

	hydrogen	oxygen	iron	U235	U238	Pu240	Pu239
15**	1.673E-02	2.023E-02	6.287E-03	2.113E-05	2.761E-03	1.328E-06	7.002E-06

CASE F14E-6 Calculated k_{∞} = 0.73412 (maximum value)
 0.80 wt% $^{235}\text{UO}_2$; 0.30 wt% Pu; $^{240}\text{Pu}/\text{Pu}$ = 16.0 wt%; 2000 g UO_2/L ; 250 g Water/L.

=xsdrn

15**	1.673E-02	2.023E-02	6.287E-03	2.254E-05	2.759E-03	1.328E-06	7.002E-06
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CASE F14F-7 Calculated k_{∞} = 0.76207 (maximum value)

0.90 wt% ²³⁵UO₂; 0.30 wt% Pu; ²⁴⁰Pu/Pu = 16.0 wt%; 2000 g UO₂/L; 300 g Water/L.

=xsdrn

15** 2.008E-02 2.191E-02 6.287E-03 2.535E-05 2.757E-03 1.328E-06 7.002E-06

CASE F14G-7

Calculated k_∞ = 0.77577 (maximum value)

0.95 wt% ²³⁵UO₂; 0.30 wt% Pu; ²⁴⁰Pu/Pu = 16.0 wt%; 2000 g UO₂/L; 300 g Water/L.

=xsdrn

15** 2.008E-02 2.191E-02 6.287E-03 2.676E-05 2.755E-03 1.328E-06 7.002E-06

CASE F14H-7

Calculated k_∞ = 0.78890 (maximum value)

1.00 wt% ²³⁵UO₂; 0.30 wt% Pu; ²⁴⁰Pu/Pu = 16.0 wt%; 2000 g UO₂/L; 300 g Water/L.

=nitawl

3**

92235	293.	0	0.	0.	0.	2.817E-05	1	1.008	1.454E+04	1	16.00	3.266E+03	1	1.
92238	293.	0	0.	0.	0.	2.754E-03	1	1.008	1.487E+02	1	16.00	3.341E+01	1	1.
94240	293.	0	0.	0.	0.	1.328E-06	1	1.008	3.084E+05	1	16.00	6.928E+04	1	1.
94239	293.	0	0.	0.	0.	7.002E-06	1	1.008	5.850E+04	1	16.00	1.314E+04	1	1.

=xsdrn

	hydrogen	oxygen	iron	U235	U238	Pu240	Pu239
15**	2.008E-02	2.191E-02	6.287E-03	2.817E-05	2.754E-03	1.328E-06	7.002E-06

G1.0 REFERENCES

ORNL, 1995, *SCALE, a Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation*, NUREG/CR-0200, Rev. 4, Vols. I-III (April 1995), Oak Ridge National Laboratory, Oak Ridge, Tennessee.

APPENDIX H
INDEPENDENT REVIEW

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APPENDIX H

INDEPENDENT REVIEW

Mr. Kevin N. Schwinkendorf of Fluor Daniel Northwest, Criticality and Shielding, performed an independent peer review. Mr. Warren D. Wittenkind of the same organization reviewed calculations made using the XSDRNPM code and reviewed the validation study for these calculations. The following is his description of this review.

H1.0 REVIEW

During Spent Nuclear Fuel Project (SNFP) basin retrieval operations, N Reactor fuel assemblies and broken fuel pieces (as well as a small quantity of Single-Pass Reactor (SPR) fuel) will be placed into Multicanister Overpack (MCO) containers for storage in the Canister Storage Building. Fuel assemblies will be placed into fuel baskets, while broken pieces (scrap) will be placed into MCO scrap baskets. Broken pieces of uranium metal which are smaller than the size criterion for acceptance into scrap baskets will be sent to waste tank AW-105 for disposal. The particle size required for low-enriched uranium metal to be considered as neutronically homogeneous is much larger than the particle size required for the mechanical segregation criterion to ensure a concentration factor of no greater than 3. Therefore, if treatment of the basin retrieval sludge results in particle sizes of less than 10 μm , this sludge easily qualifies as a neutronically homogeneous mixture. This CSER has this factor of 3 built into the limits to account for this degree of mechanical segregation, as predicted by previously documented fluid dynamics simulations using the TEMPEST code. After treatment, basin retrieval sludge will be more like existing tank waste (with regard to particle size), and the same arguments made previously for plutonium (regarding sorption and agglomeration) should also apply to retrieval sludge; there are no identified concentration mechanisms that could separate the fissile components to the degree where criticality could result.

Several editorial comments were also made, and these have been incorporated into the document.

H1.1 REVIEW COMMENT RECORD

Comments that required a response were put onto a Review Comment Record (RCR) form, and a copy is provided. The disposition of these comments is described on the RCR. In addition, a Checklist for Technical Peer Review is provided to show the scope of this review.

FLUOR DANIEL NORTHWEST

TECHNICAL PEER REVIEWS

CHECKLIST FOR TECHNICAL PEER REVIEW

Document Reviewed:

Title: Criticality Safety Evaluation of Disposing of K Basin Sludge in Double-shell Tank AW-105

Author: Charles Rogers

Date:

Scope of Review: Full document review

Yes	No*	NA	
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	** Previous reviews complete and cover analysis, up to scope of this review, with no gaps.
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Problem completely defined.
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Accident scenarios developed in a clear and logical manner.
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Necessary assumptions explicitly stated and supported.
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Computer codes and data files documented.
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Data used in calculations explicitly stated in document.
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Data checked for consistency with original source information as applicable.
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Mathematical derivations checked including dimensional consistency of results.
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Models appropriate and used within range of validity or use outside range of established validity justified.
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Hand calculations checked for errors. Spreadsheet results should be treated exactly the same as hand calculations.
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Software input correct and consistent with document reviewed.
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Software output consistent with input and with results reported in document reviewed.
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Limits/criteria/guidelines applied to analysis results are appropriate and referenced.
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Limits/criteria/guidelines checked against references.
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Safety margins consistent with good engineering practices.
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Conclusions consistent with analytical results and applicable limits.
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Results and conclusions address all points required in the problem statement.
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	** Review calculations, comments, and/or notes are attached.
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Traceability
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Document approved (i.e., the reviewer affirms the technical accuracy of the document).

Kevin N. Schwinkendorf *Kevin N. Schwinkendorf*

Reviewer: (Printed and Signed)

Date 5/17/99

* All "NO" responses must be explained below or on an additional page.

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

NUCLEAR ENGINEERING

FLUOR DANIEL NORTHWEST

TECHNICAL PEER REVIEWS

CHECKLIST FOR TECHNICAL PEER REVIEW

Document Reviewed: Title: Criticality Safety Evaluation of Disposing of K Basin Sludge in Double-shell Tank AW-105
 Author: Charles Rogers
 Date:
 Scope of Review: Review of appendix on computer input files for the XSDRNPM and NITAWL codes.

Yes	No*	NA	
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	** Previous reviews complete and cover analysis, up to scope of this review, with no gaps.
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Problem completely defined.
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Accident scenarios developed in a clear and logical manner.
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Necessary assumptions explicitly stated and supported.
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Computer codes and data files documented.
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Data used in calculations explicitly stated in document.
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Data checked for consistency with original source information as applicable.
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Mathematical derivations checked including dimensional consistency of results.
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Models appropriate and used within range of validity or use outside range of established validity justified.
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Hand calculations checked for errors. Spreadsheet results should be treated exactly the same as hand calculations.
<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Software input correct and consistent with document reviewed.
<input type="checkbox"/>	<input type="checkbox"/>	<input checked="" type="checkbox"/>	Software output consistent with input and with results reported in document reviewed.
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Limits/criteria/guidelines applied to analysis results are appropriate and referenced.
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Limits/criteria/guidelines checked against references.
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Safety margins consistent with good engineering practices.
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Conclusions consistent with analytical results and applicable limits.
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Results and conclusions address all points required in the problem statement.
<input type="checkbox"/>	<input type="checkbox"/>	<input checked="" type="checkbox"/>	** Review calculations, comments, and/or notes are attached.
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Traceability
<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	Document approved (i.e., the reviewer affirms the technical accuracy of the document).

Computer Input Files Appendix are consistent with stated case, output files were not in appendix and were not checked.

Warren D. Wittekind Warren D. Wittekind

17 May 1999
 Date

* All "NO" responses must be explained below or on an additional page.

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

NUCLEAR ENGINEERING

REVIEW COMMENT RECORD (RCR)			1. Date 03/29/99	2. Review No.
5. Document Number(s)/Title(s) HNF-3500, Rev. 0, <i>Criticality Safety Evaluation of Disposing of K Basin Sludge in Double-shell Tank 4H-105.</i>		6. Program/Project/ Building Number	3. Project No.	4. Page 1 of 6
7. Reviewer Kevin N. Schwinkendorf	8. Organization/Group Fluor Daniel Northwest/ Criticality and Shielding	9. Location/Phone M.S. 84-44/ 376-0960		
10. Agreement with indicated comment disposition(s) 11. CLOSED				
Organization Manager (Optional) _____ <i>Kevin N. Schwinkendorf</i> Reviewer/Point of Contact <i>Charles Rogers</i> Author/Originator Date: <u>5/20/99</u> Date: <u>5/20/99</u>				
12. Item	13. Comment(s)/Discrepancy(s) (Provide technical comment and dated recommendation of the action required to correct/resolve the discrepancy/problem indicated.)	14. Hold Point	15. Disposition (Provide justification if not accepted.)	
1.	This CSER does not follow the site-standard format required for CSERs, as given in HNF-PRO-539. Actually, the scope of this document is greater in extent than what could be easily fit into the 539 format. Still, supporting material (e.g., related to chemistry) might be moved to an appendix.		The format has been modified to be closer to the HNF-PRO format, and the chemistry section has been moved to an appendix.	
2.	There are noted in the marked-up copy, many editorial comments. Note that in several places, comments were made that later turned out to be unwarranted, because questions were answered, or points were clarified later in the document, and red ink does not erase.		Okay.	
17. Comment Submittal Approval:				

REVIEW COMMENT RECORD (RCR)		1. Date	2. Review No.
		3. Project No.	4. Page
		03/29/99	2 of 6
12. Item	13. Comment(s)/Discrepancy(ies) (Provide technical justification for the comment and detailed recommendation of the action required to correct/resolve the discrepancy/Discrepancy indicated).	14. Hold Point	15. Disposition (Provide justification if NOT accepted.)
3.	In the last paragraph of Section 2.5 <i>Neutron Absorbers</i> , reference is made to the 3-fold increase in plutonium density or a 3-fold decrease in iron density. I understand that the factor of 3 increase in Pu density came from TEMPEST simulations for high-density particles of plutonium with size less than 10 μm ; for particles greater than 50 μm in size, concentration factors increase rapidly (this is also explained nicely in Section 9.8, <i>Particle Size</i>). Do these assumptions/results also hold for the factor of 3 decrease in iron density? Are these ideas interchangeable? It seems to me that iron particles, being less than half as dense as plutonium, would have different values associated with concentration criteria (i.e., what concentration factor applies to iron particles as a function of particle size?)	X	The "factor of 3" assumption applies to a system containing plutonium particles and neutron absorbers. The formation of stable agglomerates containing plutonium and absorber particles is discussed in WMC-SD-WM-TI-725. For small particles, including those less than 10 microns, the density effect is minimal. A statement was added to the CSER to explain that density is of no importance for particles less than 10 microns in diameter. No further changes are required. However, it should also be noted that the direction provided to K Basin is to follow the technical guidance of WMC-SD-WM-TI-725.
4a.	This CSER applies an USL based on a limiting $k_{\text{eff}} = 0.90$, because of the lack of experimental data with systems with such large amounts of iron absorber. Reference is also made to earlier results for plutonium in tank waste, where the minimum critical concentration in CHM solids is 2.6 g Pu/L. Unfortunately, this result was based on a limiting $k_{\text{eff}} = 0.95$. Large quantities of iron and other absorbers were also present in these earlier analyses. How can you retain use of 0.95 (in practice, 0.935) for plutonium in the CHM, but apply 0.90 (in practice, 0.873) for the new CSER? Does this mean that the earlier work should have also been based on 0.90?	X	The determination of 2.6 g/L as the minimum critical concentration (MCC) of plutonium in tank waste was accomplished in a different document using the MDMNGA computer code and a different validation report. No changes are required.

REVIEW COMMENT RECORD (RCR)		1. Date	2. Review No.	16. Status
		03/29/99		
		3. Project No.	4. Page	3 of 6
12. Item	13. Comment(s)/discrepancy(s) (Provide technical justification for the comment and detailed recommendation of the action required to correct/resolve the discrepancy/problem indicated.)	14. Hold Point	15. Disposition (Provide justification if NOT accepted.)	
4b.	This reviewer was once told that the use of "two limit criteria in the same CSER is confusing." In my case, it was the "mixing" of 0.98 for K-Basin operations and 0.95 for multicamister overpack (MCO) requirements. This same criticism would appear to apply here as well, all you are "mixing" 0.95 and 0.90. (Just an observation.)		No response required.	
5.	There are several places where reference is made to (what used to be) the fact that the maximum U enrichment for any fuel present in KE Basin is 0.95 wt%. A short time ago, a relatively small quantity of Mark IA fuel (and 1.25 wt% scrap) was shipped to the KE Basin from the 327 Building, where it had been undergoing characterization. The quantity is on the order of several hundred kg; this is very much less than the 0.95 wt% material already there, but it is nevertheless no longer true that the highest enrichment present in KE Basin is 0.95 wt%. Does this actually affect your CSER substantively?	X	The additional fact has been noted in the CSER description. Since this material was only added recently (Spring of 1999), its contribution to KE Basin sludge is negligible.	
6.	There is also reference made to CSER-010 Rev. 1 for the 300 Area (for ideal geometry masses, dimensions, etc.). This document is currently being revised with the hemisphere results being replaced with MCNP results. It was found that for the higher enrichments, MCNP gave a smaller hemispherical radius for some desired keff than did the buckling leakage correction method as employed in GOLP. The discrepancy was highest for the 1.25 wt% scrap result, but essentially no difference was observed for 0.95 wt% materials. Also, all other geometries (sphere, cylinder, and slab) are still in excellent agreement (MCNP with WIMS-E/GOLP).		Reference to CSER-010 is not directly used in the calculations for K Basin Sludge. No action required.	

REVIEW COMMENT RECORD (RCR)		1. Date	2. Review No.	
		03/29/99		
		3. Project No.	4. Page	
			4 of 6	
12. Item	13. Comment(s)/Discrepancy(ies) (Provide technical justification for the review. Indicate the discrepancy/problem indicated.)	14. Find Point	15. Disposition (Provide justification if NOT accepted.)	16. Status
7.	How did ORIGEN results actually enter into this CSER? It appears as though any burnup information you used came from the SCATS database? The problem with ORIGEN2 (assuming you used the "straight-out-of-the-box" ORIGEN2 and associated cross section libraries from SCALE 4.3) is that the most recent N Reactor libraries were not used. The ORIGEN2 code requires burnup-dependent microscopic cross sections that appropriately reflect the self-shielding effects of exposing N Reactor tube-in-tube fuel assemblies in a thermal spectrum that is proper for a graphite-moderated reactor. The version of ORIGEN2 used for N Reactor fuel here at Hanford (maintained by Frank Schmittroth and then later Warren Wittkeind, when Frank retired) used WIMS-D-generated Libraries. Later (1996), WIMS-E was used to improve the ORIGEN2 Libraries, and this was documented by Randy Schwarz (1997). The current ORIGEN2 verification & validation will be released in the near future. The "closest match" libraries you could have used with ORIGEN2 (directly from SCALE 4.3) only has cross section libraries for "PMR".	X	The isotopic database was taken from SCATS as stated in Table 5-5. No new ORIGEN calculations were used. The point of Table 5-6 is to demonstrate that the data in Table 5-5 (and corresponding Figure 5-1) are conservative. Figure 5-1 will be further modified to incorporate the data from Table 5-6 to illustrate the bounding curve and to demonstrate the conservatism more clearly.	
8.	In Table 5-6, you visually pick data off graphs which were created by this reviewer. If you prefer (and have a charge code for doing so), I could get you these numbers directly from the spreadsheets these graphs were created in.		Table 5-6 provides adequate verification of conservatism. No further analysis is needed. However, Figure 5-1 will be modified to clarify the conclusion.	

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REVIEW COMMENT RECORD (RCR)		1. Date 03/29/99	2. Review No.
		3. Project No.	4. Page 5 of 6
12. Item	13. Comment(s)/Discrepancy(ies) for the comment and the description of the condition and the action required to correct/resolve the discrepancy/condition(s).	14. Hold Point	15. Disposition (Provide justification if NOT accepted.)
9a.	In the last paragraph in Section 5.2.2 <i>Plutonium Generation</i> , you make the statement that ^{240}Pu is the largest source of decay heat. How can this be? According to Radnuc-2A (which interpolates between ORIGEN2 data tables, sums keys, and decays all keys in its input file to a common date), certain fission products are much higher. In fact, the ratio of decay heat generation for ^{137}Ba to ^{240}Pu is 11, and even for ^{239}Pu , this ratio is 1.7. This uses the Radnuc-2A input for the KE/KH Combined Database from Mike Packer.	X	Text has been changed to clarify that the decay heat of the fuel elements is proportional to the amount of Pu-240 present.
9b.	Also, you assume that 16 wt% ^{240}Pu is conservative and bounding for all K Basin fuel. This is almost true, but, according to the database mentioned above (from Packer), there are three keys, 11540, 12565, and 12852, that have exposure greater than 16 wt% ^{240}Pu . These are: key 11540 is 16.5 wt%, key 12565 is 16.7 wt%, and key 12852 is 16.6 wt%.		The text will be modified to clarify that less than 2% of the K Basin Fuel might contain over 16 wt% Plutonium-240. Also note that the CSER for K Basin Sludge references Wittkind (SCATS), not Packer. The Packer values differ slightly from the SCATS values obtained from Wittkind. The text will acknowledge that 16% is bounding for virtually all K-Basin fuel.
10.	In Appendix A, you talk about XSDRNPM. Are you really still using ENDF/B-IV? Also, you've done the V&V (and established a bias), but I am concerned about what flux spectrum you used for the thermal region. Minimum critical mass results are found in a thermal spectrum, and so cross sections have to be collapsed using the proper weighting functions. We used to (in the late 1980's timeframe) use NUOY to generate new cross sections for WIMS-E when we needed them, and this was considered important.		Yes, the CSER really uses 27 group library ENDF/B-IV. The bias was determined for this cross section set and is being applied appropriately.

REVIEW COMMENT RECORD (RCR)		1. Date 03/29/99	2. Review No.
12. Item	13. Comment(s)/Discrepancy(s) (Provide technical justification for the review action required to correct/resolve the discrepancy/problem indicated.)	14. Hold Point	15. Disposition (Provide justification if NOT accepted.)
11.	In Appendix B, you state that if you have optimally-sized and spaced rods, natural uranium can go critical (and I'm reading between the lines here, but this implies that this is possible in light water). Did you mean that you can go critical in heavy water (this being the CANDU reactor design)? I reviewed some of my old WIMS-E results, and I found that if you extrapolate the maximum k_{eff} for optimal rod lattices at each of the enrichments (0.95, 1.15, and 1.25 wt%) down to 0.72 wt% $k_{eff} = 1.004$. So, did you get this result from me? It just stuck out because everybody knows you cannot make a practical reactor design out of natural uranium and light water.		Yes, we agree. The 0.72% represents the extrapolation of the handbook limits (ARCO 600) for an ideal lattice in light water. No changes are required.

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16. Status

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DISTRIBUTION SHEET

To Distribution	From NS&L/Criticality Support	Page 1 of 2
Project Title/Work Order Criticality Safety Evaluation of Disposing of K Basin Sludge in Double-Shell Tank AW-105		Date June 4, 1999
		EDT No. 625118
		ECN No. NA

Name	MSIN	Text With All Attach.	Text Only	Attach/Appendix Only	EDT/ECN Only
FDH					
S. T. Almodovar	A3-02	X			
J. E. Fialkovich	A3-02	X			
E. W. Gerber	R3-11	X			
L. B. McDaniel	R3-11	X			
LMHC					
D. R. Bratzel	S7-73	X			
R. J. Cash	S7-73	X			
C. E. Leach	R1-49	X			
E. J. Lipke	R1-49	X			
J. E. Meacham	R1-49	X			
C. A. Rogers (6)	R1-43	X			
L. E. Thomas	R3-01	X			
DESH					
K. H. Bergsman	X3-85	X			
M. C. Brady Raap	R1-43	X			
D. E. Bullock	R3-86	X			
J. R. Frederickson	R3-86	X			
M. A. Jensen	X3-79	X			
F. J. Muller	X3-85	X			
D. R. Precechtel	X3-85	X			
FDNW					
J. P. Estrellado	B4-44	X			
K. N. Schwinkendorf	B4-44	X			
W. D. Wittekind	B4-44	X			
WMH					
T. S. Vail	T4-56	X			

DISTRIBUTION SHEET

To Distribution	From NS&L/Criticality Support	Page 2 of 2			
Project Title/Work Order Criticality Safety Evaluation of Disposing of K Basin Sludge in Double-Shell Tank AW-105		Date June 4. 1999			
		EDT No. 625118			
		ECN No. NA			
Name	MSIN	Text With All Attach.	Text Only	Attach./Appendix Only	EDT/ECN Only
NHC					
T. A. Flament	H0-34	X			
F. W. Moore	H0-34	X			
K. L. Pearce	H0-34	X			
C. A. Petersen	H0-34	X			
W. W. Rutherford	H0-34	X			
J. P. Sloughter	H0-34	X			
DOE					
D. H. Alexander (3)	S7-54	X			
S. J. Altschuler	R3-79	X			
L. T. Nirider	A5-55	X			
Offsite					
Adolf S. Garcia,		X			
US DOE-ID					
850 Energy Drive					
Idaho Falls, ID 83401					
Dr. Jerry McKamy		X			
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19901 Germantown Rd.					
Germantown, MD 20874-129					
Dr. Kevin A. Niemer		X			
Duke Engineering, MSIN WC-26B					
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SNF Project Files A-13B	H6-08	X			
Central Files	A3-88	X			