AUG 21 1996 21

ENGINEERING DATA TRANSMITTAL

Page 1 of 1

1. EDT 156471

2. To: (Receiving Organization) Tank Waste Remediation System	3. From: (Originating Organization) Criticality and Shielding	4. Related EDT No.: N/A
5. Proj./Prog./Dept./Div.: Tank Farm Transitions Projects	6. Cog. Engr.: C. A. Rogers	7. Purchase Order No.: N/A
8. Originator Remarks: This document supports the Hanford Tank Farms Criticality Team report on the criticality safety of tank waste.		9. Equip./Component No.: N/A
		10. System/Bldg./Facility: Tank Farms
11. Receiver Remarks:		12. Major Assm. Dwg. No.: N/A
		13. Permit/Permit Application No.: N/A
		14. Required Response Date: August 20, 1996

15. DATA TRANSMITTED					(F)	(G)	(H)	(I)
(A) Item No.	(B) Document/Drawing No.	(C) Sheet No.	(D) Rev. No.	(E) Title or Description of Data Transmitted	Approval Designator	Reason for Transmittal	Originator Disposition	Receiver Disposition
1	WHC-SD-SQA-CSA-507		0	CSER 96-010: Criticality Parameters for Tank Waste Evaluation	N/A S KN	1,2		

16. KEY									
Approval Designator (F)		Reason for Transmittal (G)				Disposition (H) & (I)			
E, S, Q, D or N/A (see WHC-CM-3-5, Sec.12.7)		1. Approval 2. Release 3. Information		4. Review 5. Post-Review 6. Dist. (Receipt Acknow. Required)		1. Approved 2. Approved w/comment 3. Disapproved w/comment		4. Reviewed no/comment 5. Reviewed w/comment 6. Receipt acknowledged	
17. SIGNATURE/DISTRIBUTION (See Approval Designator for required signatures)									
(G)	(H)	(J) Name	(K) Signature	(L) Date	(M) MSIN	(J) Name	(K) Signature	(L) Date	(M) MSIN
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21. DOE APPROVAL (if required) Ctrl. No. ☐ Approved ☐ Approved w/comments ☐ Disapproved w/comments									

Criticality Parameters for Tank Waste Evaluation

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U.S. Department of Energy Contract DE-AC06-87RL10930

EDT/ECN: 156471 UC: 510
Org Code: 8M730 Charge Code: N1FC3
B&R Code: EW3120071 Total Pages: 129

Key Words: criticality, tank waste, subcritical limit, critical
parameters, safety, plutonium, mass ratio, conservative
waste model

Abstract: A summary is provided of basic parameters used to evaluate criticality safety of high-level tank waste at the Hanford Site. Critical sizes and plutonium concentrations are based on a conservative waste model with reduced neutron absorption and optimized water. Figures are provided of sphere and slab minimum critical dimensions and plutonium critical masses. Minimum subcritical limit absorber/plutonium mass ratios are provided for selected waste components. Component contributions to subcriticality can be combined by adding the individual actual-to-minimum subcritical mass fractions. A discussion is provided of the margin of safety inherent in tank waste.

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Karen H. Noland 8/21/96
Release Approval Date

Approved for Public Release

CSER 96-010

CRITICALITY PARAMETERS FOR TANK WASTE EVALUATION

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LIST OF TERMS

CSER	criticality safety evaluation report
CWM	conservative waste model
DCRT	double-contained receiver tank
DST	double-shell tank
SST	single-shell tank
X/Pu	component/plutonium mass ratio
XS	cross section

ACKNOWLEDGEMENT

Santiago Parra helped define the composition of supernatant waste and helped with some of the figures. He read and provided comments on drafts of the report, and his help is greatly appreciated.

CRITICALITY PARAMETERS FOR TANK WASTE EVALUATION

1.0 INTRODUCTION AND SUMMARY

This study summarizes basic parameters used for the criticality safety evaluation of high-level waste at the Hanford Site.

For criticality to be possible it is necessary for fissionable material to be present. Low concentrations of fissionable material, primarily ^{235}U and ^{239}Pu , are present in Hanford Site tank waste. Criticality is influenced by many parameters, including the following:

- Type of fissionable material
- Concentration
- Size and shape (geometry)
- Composition of the waste
- Effectiveness of neutron absorbers
- Effectiveness of neutron reflectors
- Moderator content (especially water).

1.1 CONSERVATIVE WASTE MODEL (CWM)

The complexity of an unknown heterogeneous distribution of waste types can be overcome by replacing the entire configuration with a single composition known to have a smaller macroscopic neutron absorption cross section than any of the individual waste types. A Conservative Waste Model (CWM) with reduced neutron absorption and optimized water is defined to provide a waste composition for which critical parameters are conservative relative to any real waste. Because all discharged waste has a higher neutron absorption than the CWM, it is conservative to represent the entire volume of waste by the CWM.

1.2 WASTE CONFIGURATIONS

Under normal conditions, waste discharged to tank farms is well mixed. Upon entering the storage tank, the highly fluid waste forms into a relatively uniform layer over the entire area of the tank. In practice, however, the plutonium may not actually reside in a uniform layer, but may have a non-uniform distribution.

In assessing criticality safety, the potential for changing shape and composition must be taken into account. The shape that minimizes the volume and mass of plutonium required to achieve criticality is the sphere. A spherical shape minimizes leakage of neutrons and has the smallest possible surface-to-volume ratio. For this reason, sphere critical parameters can be used as a conservative estimate of critical parameters for other geometric shapes. Compact geometries, such as cubes and cylinders with a height/diameter ratio of about 1, will have critical parameters close to those for spheres of the same volume.

1.3 PRIMARY SAFETY PARAMETERS

A parameter of primary importance is the mass ratio of an absorber material to the plutonium present. The minimum subcritical mass ratio determines the relative quantity of an absorber material which, if present individually with the plutonium, would provide subcriticality for any degree of moderation. The minimum subcritical absorber/plutonium mass ratios were calculated for the more important waste components, selected for their relative abundance and neutron absorption capacity. A subcritical mass ratio can also be defined in terms of the total solids present without requiring a knowledge of the waste composition. When the settled solids/plutonium mass ratio exceeds 770, criticality is not possible in homogeneous waste.

Another parameter of primary importance is the plutonium concentration. Subcriticality is ensured whenever the plutonium concentration is less than 2.6 g/L. This is a general limit applicable to all waste, including settled waste solids, regardless of the water content.

The presence of a large quantity of inert material provides the surest guarantee of continued subcriticality. Traditionally, a limit of 1 g/L on the concentration of plutonium in settled solids has been used. In reality, the average plutonium concentration in settled solids is considerably lower.

1.4 COMBINING THE EFFECT OF SEVERAL ABSORBERS

The contributions of different components to the total absorption cross section can be combined by adding their individual contributions, which are proportional to their individual mass ratios divided by the corresponding subcritical limits. When the sum of the fractions is greater than unity, the resulting configuration will be subcritical, if homogeneous. Subcriticality is ensured in a non-homogeneous mixture if this condition holds for waste at any location within the waste.

1.5 SPECIFIC CONDITIONS AFFECTING CRITICAL MASS

Three specific conditions that affect the plutonium critical mass in waste are discussed:

- The enhanced production of plutonium at the outer surface of the fuel and the discharge of this outer surface as part of the cladding waste
- The formation of intermetallic particles which might be denser and physically distinct from other waste, and the possibility that they might separate from other waste as the result of mechanical processes to form localized accumulations
- The presence of fission products.

1.6 MARGIN OF SUBCRITICALITY

Several specific examples are provided to illustrate the inherent margin of subcriticality in tank waste.

The margin of subcriticality for tank waste was evaluated by determining actual-to-minimum subcritical fractions for actual waste samples. These samples were selected from a large number of available samples for their high plutonium concentration and low concentration of neutron-absorbing materials. For these samples, the plutonium concentration ranged between 0.214 and 0.060 g/L. The sum of the actual-to-minimum subcritical fractions for these five samples ranged between 4.6 and 35.

A value of k_{∞} was calculated for waste samples selected on the basis of their apparent low neutron absorber content. Of five samples evaluated, the largest value of k_{∞} was 0.0826, while three samples indicated a value of k_{∞} less than 0.02. Therefore, a k_{∞} of 0.083 should represent a reasonable upper limit for real waste under actual storage conditions.

For the tank farms as a whole, the plutonium inventory is conservatively estimated to be less than 981 kg, and the average plutonium concentration is estimated to be 0.0157 g/L of solids. A conservative estimate of the solids/plutonium mass ratio for the tank farms as a whole was 74,500, a value 100 times greater than required to ensure subcriticality in well-mixed waste.

For tank 241-SY-102, which receives Plutonium Finishing Plant waste, the June 1996 estimated plutonium inventory is 41.4 kg, and the estimated solids/plutonium mass ratio is at least 7,790. This is the lowest ratio for any tank. Tank 241-C-104, with an estimated inventory of 89 kg of plutonium, has one of the largest plutonium inventories of any tank. The solids/plutonium mass ratio of tank 241-C-104 is estimated to be 15,055.

The minimum tank inventory required to achieve criticality, assuming the plutonium is distributed uniformly over the area of the tank, is 1,060 kg of plutonium. This value is found by multiplying the minimum critical areal density by the cross-sectional area of a 22.9-m-diameter tank. This quantity exceeds the estimated total inventory for all tanks combined, and is 10 times greater than the largest inventory expected in any single tank.

Figure 1-1 illustrates the criticality safety envelope for 22.9-m- (75-ft-) diameter waste storage tanks. For criticality to be possible for a given plutonium inventory, the plutonium concentration must fall to the right of the curve of minimum critical mass. The curve shown is the subcritical mass limit based on a k_{eff} of 0.935 for CWM solids in spherical geometry with optimal water moderation. The operating zone refers to the range of plutonium concentration and tank inventory size considered acceptable. For the illustration, the operational limits are shown for a tank plutonium inventory of 125 kg and a concentration of 2 g/L. The operating zone may be different for different tanks, based upon specific evaluations. The region of "Maximum Tank Parameters" is defined by the largest actual tank inventory and the largest actually measured plutonium concentration. The region of "Average Tank Parameters" indicates the average over all 177 storage tanks, based upon the estimated total inventories at tank farms.

Figure 1-1. Criticality Safety Envelope for 22.9-m- (75-ft-) Diameter Waste Storage Tanks.

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Concentration and mass are controlled at the facility from which the waste is discharged. At tank farms the only control over incoming waste is verification (prior to transfer) that an incoming transfer complies to tank farm specifications. The ability to control waste composition within storage tanks is limited. Changes made by tank farms operations are of secondary importance to ensuring a proper composition for incoming waste. Multiple, major violations of limits and controls in a processing facility would be required before it would be possible to discharge a sufficient quantity of out-of-specification waste for criticality in a storage tank.

Section 6.5 shows the comparison between the plutonium critical mass as a function of plutonium concentration in CWM waste and in "real" waste. In simulated real waste the concentration of plutonium required for criticality increases dramatically. The minimum plutonium concentration for criticality in fully water-saturated waste becomes 11 g/L, more than four times as large as the value in dry CWM waste. Below 8.9 g Pu/L, more than 1,000 kg of plutonium is required for criticality, regardless of water content.

2.0 OBJECTIVE AND SCOPE

2.1 OBJECTIVE

The objective of this study is to define and quantify key parameters for criticality safety limits and controls for high-level waste in storage tanks on the Hanford Site. The goal of criticality safety is to prevent a nuclear criticality accident and associated releases of radiation and energy.

2.2 SCOPE

This study provides critical parameters for high-level waste in Hanford Site waste tanks. Properties of tank waste that influence critical parameters are defined, and their relevance to criticality safety is described.

Much of the information that follows was obtained from three reports:

- *High-Level Waste Tank Subcriticality Safety Assessment*, WHC-SD-WM-SARR-003 (Braun et al. 1994)
- *CSER 92-009: An Analytical Model for Evaluating Subcritical Limits for Waste in Hanford Site Storage Tanks*, WHC-SD-SQA-CSA-20356 (Rogers 1993)
- *CSER 94-004: Criticality Safety of Double-Shell Waste Storage Tanks*, WHC-SD-SQA-CSA-20368 (Rogers 1994).

Although the original intention was to summarize material from the above reports, major new information has been developed. Most critical parameters in this report have been recalculated using a newer version of the computer code and in some cases using different criteria of acceptability. For these reasons, values provided are sometimes more conservative than in the above reports.

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3.0 BACKGROUND

The properties of waste and its arrangement inside a waste storage tank influence the amount of fissionable material required to achieve a nuclear criticality. Characteristics of waste and its storage that have importance to criticality safety evaluations are briefly described.

3.1 DEFINITION OF NUCLEAR CRITICALITY IN WASTE

Nuclear criticality is when a self-sustaining neutron chain reaction occurs in fissionable material. A fissionable nucleus becomes unstable upon absorbing a neutron, splits into smaller nuclei, and releases additional neutrons. These neutrons cause even more atoms to fission. When the number of neutrons in the material remains constant (number produced equals number that escape or are absorbed), the system is said to be critical, and the neutron multiplication factor (k) is equal to 1.0. When k is greater than 1.0, the system is supercritical, meaning the number of neutrons will increase exponentially with time. When k is less than 1.0, the material is subcritical, meaning the number of neutrons will decrease with time.

The multiplication constant for an infinitely large volume of material is written as k_{∞} . For a finite volume, the effective multiplication constant is written as k_{eff} . Tank waste is expected to remain subcritical with a k_{eff} less than 1.

The k_{eff} value is influenced by many parameters including the following:

- The geometry of the container, which influences the neutron leakage (i.e., the rate at which neutrons escape)
- The composition of the waste, which determines the fraction of absorbed neutrons that cause fissioning and produce additional neutrons
- The water content, which determines how quickly neutrons are thermalized (water also absorbs neutrons).

3.2 FISSIONABLE NUCLIDES IN HANFORD SITE WASTE

The fissionable nuclides potentially present in Hanford Site waste are ^{233}U , ^{235}U , ^{238}Pu , ^{239}Pu , ^{240}Pu , and ^{241}Pu . The proportions of ^{238}Pu and ^{241}Pu in the plutonium are small, and these isotopes, along with the ^{240}Pu , are all assumed to be ^{239}Pu (i.e., added to the ^{239}Pu content). The ^{235}U can usually be ignored because ^{235}U is almost always combined with a large enough quantity of ^{238}U to ensure that the uranium is subcritical. An unlimited mass of the isotope ^{238}U cannot be made critical, and it therefore does not have a critical mass associated with it.

In practice, fissionable nuclides capable of contributing to a neutron chain reaction are converted to their "plutonium-equivalents." Both ^{235}U and ^{233}U are usually treated as if they were ^{239}Pu and added to the plutonium

inventory. Although at higher concentrations ^{233}U can have a critical mass that is smaller than that of ^{239}Pu , at the low concentrations in tank waste the replacement of ^{233}U with ^{239}Pu is conservative. Therefore, the fissionable material inventory of a tank is generally taken as its plutonium-equivalent inventory in which the quantities of ^{235}U (excluding that in natural and depleted uranium) and ^{233}U are replaced by an equivalent mass of plutonium.

Nuclides ^{238}Pu and ^{240}Pu are fissionable, but not fissile. The presence of even a small quantity of moderating material increases the critical mass of these isotopes very rapidly until criticality is not possible with them alone. In the tank waste environment, ^{240}Pu acts as a mild poison, and its presence increases the quantity of ^{239}Pu required to achieve criticality. Generally, the content of ^{238}Pu is very small and does not need to be taken into account. It is conservative to assume all plutonium is ^{239}Pu (i.e., to replace ^{240}Pu and ^{238}Pu with ^{239}Pu).

Plutonium may not be uniformly distributed and there is considerable uncertainty as to the degree of non-uniformity. Conservative assumptions must be made to compensate for the lack of information.

Although ^{237}Np and ^{241}Am are fissionable, they are not fissile, and criticality is possible only with fast neutrons. The water- and steel-reflected minimum critical masses for ^{237}Np are 83 kg and 55 kg, respectively (Clayton 1979). For criticality with ^{237}Np , the concentration must exceed 12.7 kg/L. An estimated 12.7 kg of ^{237}Np were sent to tank 241-AZ-102. This total mass of ^{237}Np would have to be placed within a single liter to achieve the minimum critical concentration, but even then the mass would be far less than required for criticality.

The water- and steel-reflected minimum critical masses for ^{241}Am are 51 kg and 34 kg, respectively (Clayton 1979). Just as for ^{237}Np , a high concentration of ^{241}Am is required for criticality. However, ^{241}Am is present in tank 241-SY-102 in low concentrations. The low concentration and the presence of moderation ensure that ^{237}Np and ^{241}Am cannot contribute to a criticality.

3.3 TYPES OF WASTE

Waste in a storage tank consists of liquid and settled solids in various proportions. Liquid is present as free-standing supernatant or as interstitial liquid in the void spaces of solid wastes. "Solids" refers to the non-liquid components of waste that settle when left undisturbed, and can be separated from the liquid using a centrifuge or by draining the waste through a fine mesh. Solids are primarily of two types: sludge and salt cake.

The waste makeup of most single-shell tanks (SST) is primarily solid. The majority of the supernatant liquid has been removed from the SSTs because of the interim stabilization efforts.

The waste in most double-shell tanks (DST) is primarily liquid, with some sludge and salt cake. Double-shell slurry is a mixture of inorganic salt crystals in high salt mother liquor, with small amounts of metal hydroxide solids.

Aging waste is new waste that is placed in a DST with heat removal and some mixing capability for removing the decay heat from the radionuclides.

3.3.1 Inert Material

Controls to ensure that the plutonium concentration remains low are maintained by the presence of a large fraction of inert material. The plutonium remains combined with this material after settling and mixing. The chemical makeup of the waste provides assurance against the dissolution and separation of solids from the plutonium. Traditionally, a limit of 1 g/L on the concentration of plutonium in settled solids has been used. In reality, the average plutonium concentration in settled solids is considerably lower.

3.3.2 Water Content

Water content in waste varies from very high in supernatant liquid to relatively low in dry waste. Ultimately, the drainable water (liquid) will be removed from all the waste in preparation for long-term undisturbed storage. Normally, the water content is not expected to be less than 25 wt%.

The water content at which the plutonium critical mass or the critical concentration are minimized is referred to as optimal. At the very low plutonium concentration in waste, optimal water content is generally a state of extreme dryness. However, in theoretical studies in which a much higher plutonium concentration is assumed, the minimum critical mass occurs at a higher water content. Unless otherwise stated, critical parameters in this report are provided for optimal water content.

3.3.3 Plutonium Distribution

Under normal conditions waste is discharged in a well-mixed, highly fluid suspension which tends to form into a relatively uniform layer over the entire area of the tank. In practice, however, the plutonium may not actually reside in a uniform layer. Postulated mechanisms capable of causing a non-uniform distribution of plutonium include differential settling, thermal transport effects in convection cells, the filtering effect of solid layers, preferential dissolution in organic layers, sorting by particle size during stirring, discharge of high concentrations, etc. Because of such possibilities, the actual distribution of plutonium may contain regions of significantly higher-than-average concentration.

3.4 TANK SIZE AND GEOMETRY

All DSTs are 22.9 m (75 ft) in diameter and have a nominal capacity of 3,800,000 L (1,000,000 gal). A DST is constructed of two steel shells with an

annular air space between them. The outer shell is surrounded by reinforced concrete. Older SSTs have the same diameter, but the height and capacity may be less. Double-contained receiver tanks (DCRT) used for intermediate storage are much smaller in diameter than DSTs and have a capacity less than 151,416 L. In addition, there are several tank vaults, each consisting of two 75,700-L tanks and two 18,925-L tanks. It is conservative to treat these vault tanks the same as the much larger tanks. For the purposes of criticality safety evaluation, all tanks are considered infinite in size.

Appendix B of the *High-Level Waste Tank Subcriticality Safety Assessment* (Braun et al. 1994) shows the current status of each of the Hanford Site waste tanks. It provides tables and figures that display the type and amount of waste, information about tank integrity, and watch list status as of August 1993. Detailed information is also available in the *Waste Tank Summary Report* (Hanlon 1996), which is issued monthly.

4.0 SUBCRITICAL LIMITS

Tank waste is a complex mixture of components, with uncertainties for every component. These uncertainties add complexity in determining critical parameters. Therefore, most subcritical limits for controlling waste are based on a CWM using values that, in spite of uncertainties, will ensure that the real case is always as safe or safer than the model. In a CWM, a value that is known to be subcritical after taking all the uncertainties into consideration is selected as a subcritical limit. The CWM developed in Rogers (1993) is used as a basis for setting the subcritical limits for this study.

As a rule, critical values are calculated assuming a homogeneous (uniform) mixture of plutonium, waste, and water. In practice, however, waste is not actually homogeneous, and the implications of complex waste geometries are examined. As a rule, heterogeneous configurations are examined by comparing them to more conservative homogeneous configurations.

In this report, the term "concentration" refers to the mass of one element in a mixture per unit volume of waste. The term "density" refers to the total mass of all the elements per unit volume of waste.

4.1 KEY PARAMETERS

Parameters (variables) that influence the concentration at which plutonium can be made critical include: (1) type of fissionable nuclide present, (2) fissionable nuclide concentration and distribution, (3) neutron absorbers present, (4) moderator content, (5) size of system, (6) shape of system, and (7) the effectiveness of neutron reflectors. The value of a parameter at which the system reaches critical is referred to as its critical value.

The k_{eff} (or k_{∞}) for subcritical limits calculated for this evaluation will not exceed 0.95 with a 95% confidence level. This criterion is met when the calculated k_{eff} (or k_{∞}) is 0.935, and the corresponding configuration is said to be critical. The difference between 0.95 and 0.935 accounts for calculational bias and code convergence uncertainty.

When a critical parameter is reported in an authoritative document, such as an American Nuclear Society Standard or the *Nuclear Safety Guide* (Thomas 1978), that parameter may be used as a subcritical limit. Two important examples are the experimentally determined plutonium minimum critical concentration of 7.2 g/L in water (Paxton and Pruvost 1987) and the minimum critical plutonium areal density provided by Carter et al. (1970) as 240 g Pu/ft (2,582 g/m²). These values are larger than the corresponding subcritical limit values calculated using the above criterion.

4.2 CONSERVATIVE WASTE MODEL

The CWM developed by Rogers (1993) was designed to address features of waste that are significant to criticality, and to always produce conservative operating limits. The CWM solids are intended to have a lower neutron absorption per unit volume than real waste. The CWM is used to calculate the critical plutonium concentrations and to determine minimum critical dimensions. Subcritical limits set by Rogers (1993) are the basis for this evaluation, and provide the basis for setting safe operating limits for the waste tanks.

4.2.1 Basis for the CWM

The CWM used component concentrations analyzed from 28 waste compositions from 16 SSTs. The primary goal in formulating this waste model was to provide a waste composition known to have a smaller macroscopic absorption cross section than that of any real waste. Critical parameters are calculated using the assumption of optimal water moderation. This ensures that subcritical limits calculated using the model are conservative relative to the actual critical values for any real waste.

As a rule, most of the plutonium is found in the solids portion of the waste. The minimum critical plutonium concentration is directly related to the minimum absorption cross section that can be achieved. Because the lowest possible degree of neutron absorption is determined by the minimum solids density and the minimum water content in the waste, the dry solids density is an important parameter for criticality safety. For the CWM, a dry solids density of 1,200 g/L is assumed.

Table 4-1 shows the composition of the CWM solids. Only the waste solids are included. The model assumes the density of dry solids to be 1,200 g/L. The concentrations of plutonium and water are independent variables and not part of the waste model solids composition.

Table 4-1. Composition of Conservative Waste Model Solids.

Component	Wt%
Oxygen	40.7
Phosphorus	6.9
Silicon	3.8
Sodium	21.5
Aluminum	7.2
Nitrogen	0.0
Iron	19.9

The macroscopic absorption cross section for the CWM solids is 0.01096 cm^{-1} . Real waste is expected to have greater absorption. Although theoretical models of waste having less absorption than this are possible, it is expected that all real tank waste will satisfy this criterion.

4.2.2 Optimal Moderation

Although not actually water, the liquid portion of waste generally contains a large water fraction, and, from a criticality safety perspective, it is conservative to treat it as pure water. In this discussion it will be called water.

When saturated, water occupies a large fraction of the waste volume. After draining, the remaining water content may still be about 40 vol%. Removal of additional water to make the solids dry will result in part of the volume previously occupied by water becoming void. However, the void fraction will be less than the fraction previously occupied by water, because the solids will compact and partially fill the created void space. For this reason, the density of dry solids is expected to be greater than the concentration of solids when saturated with water. The degree of compaction depends on the characteristics of the solids.

Figure 4-1 shows the relationship between optimal water moderation and the plutonium minimum critical concentration. At low plutonium concentrations characteristic of tank waste, the water content which results in a minimum critical plutonium concentration is very low. In fact, at the subcritical limit concentration of 2.6 g Pu/L, optimal moderation means the solids are completely dry. In other words, at the lowest possible critical plutonium concentration, waste contains no water at all. The large fraction of solids provides moderation, while hydrogen in the water increases neutron absorption. With addition of water, the fraction of neutrons absorbed in elements other than plutonium is increased, and this loss of neutrons lowers the neutron multiplication constant.

At 2.6 g Pu/L, criticality is possible only with CWM solids that are completely dry. When the plutonium concentration is 3 g/L, the water content must be less than 7 wt% for criticality to be possible. As the plutonium content increases, so does the maximum water content at which an infinite volume of waste can go critical. At 4 g Pu/L, criticality is not possible when the water content exceeds 25 wt%. At plutonium concentrations above 6.5 g/L, much higher than found in tank waste, optimal water occurs when the waste is fully water-saturated.

To achieve criticality in the minimum volume requires a greater percentage of water than to achieve criticality at the plutonium minimum critical concentration. Figure 4-1 also shows the relationship between water content and the minimum critical volume. When the plutonium concentration exceeds 6.5 g/L (a conservative value verified experimentally to be at least 7.2 g/L), criticality is possible for any water content, provided a sufficient volume of waste is available. When the water content is much higher or much lower than the optimal value, the volume of waste required for criticality might be far greater than the minimum critical volume. Likewise, the mass of plutonium required might be far greater than the minimum value.

Figure 4-1. Optimal Water Content as a Function of Plutonium Concentration in Conservative Waste Model Solids.

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For waste not saturated with water, it is not possible for the solids to be suspended. For optimal water content at low plutonium concentrations, the waste is not water-saturated and solids cannot be suspended. The smeared (i.e., average) density of waste solids will be the same as for dry solids.

4.2.3 Density of Dry Solids

For CWM solids, a bulk dry solids density of 1,200 g/L is assumed after all water (liquid) has been removed. This value represents a lower density than likely to occur for real waste solids.

Lesser solids concentrations are possible for solids suspended in water. However, a configuration with suspended solids would not contain void space, and the water content would be much greater than that which results in criticality for plutonium concentrations near to 2.6 g/L. For solids suspended in water criticality would only be possible for a plutonium concentration much greater than this value.

When water is added to real waste solids, it first fills the void space. Only after the void space is filled does the increasing water fraction cause a decrease in the solids fraction. However, for this evaluation the conservative assumption was used in calculations that no void space occurs within dry CWM solids. Even a small volume fraction of water was assumed to displace an equal volume fraction of solids (i.e., the assumed density of water plus solids is less than actual). This means that the minimum critical concentration is determined for a solids concentration reduced by the volume fraction occupied by the water.

4.2.3.1 Waste with Lower Solids Density. If the dry density for CWM solids were to be lowered, the plutonium minimum critical concentration would decrease in almost direct proportion to the decrease in density. For this reason, it is necessary to evaluate the possibility that real waste might under some circumstances achieve a dry solids density less than 1,200 g/L.

Analytical results have been reported in which the solids concentration is less than 1,200 g/L. In these samples, the liquid was allowed to drain, but the sample remained fully water-saturated. After draining, interstitial water is estimated to occupy 40% of the sample volume. Upon reducing the sample density by 40% to account for the removal of water, the solids concentration is less than 1,200 g/L, and it has been suggested that a solids concentration as low as 800 g/L may be possible. However, this occurred for solids saturated with water (liquid); whereas, with removal of water, the solids would be expected to consolidate to partially fill voids created. This would cause the concentration of solids to increase. The possibility that the density of dry solids might under some conditions be less than 1,200 g/L cannot be ruled out.

In deriving the plutonium minimum critical concentration of 2.6 g/L, several conservative assumptions were made. First, the subcritical limit on k_0 was taken to be 0.935. In other words, a calculated k_0 of 0.935 is assumed to be critical, and the calculated minimum critical concentration will be smaller than the actual value. Second, the total macroscopic neutron absorption cross section of 0.01096 cm^{-1} for CWM solids is less than that of an equal mass of real waste. Third, real waste is expected to have a dry solids density greater than 1,200 g/L. Fourth, optimal moderation for low plutonium concentrations occurs when the waste is extremely dry. In practice, a water content less than 10 wt% would be extremely unlikely, whereas this water content would be sufficient, if added to CWM solids, to increase the plutonium minimum critical concentration to more than 3 g/L.

Per unit mass, neutron absorption in water is more than twice that of CWM solids. Because the plutonium minimum critical concentration of 2.6 g/L occurs for completely dry solids, any water in real waste can actually be thought of as being equivalent to an increase in the dry solids density. In fact, 100 g of water increases the minimum critical concentration as much as 200 g of dry solids.

These conservative assumptions ensure that the dry solids density would have to fall well below 1,200 g/L and the water content would have to be extremely low before the minimum critical plutonium concentration would actually fall below 2.6 g/L. The probability of any real waste achieving a

composition capable of lowering the plutonium minimum critical concentration below this value is extremely small.

4.3 CRITERIA FOR SUBCRITICALITY

Providing assurance that a large margin of subcriticality is maintained within a 3.8-million L storage tank is extremely difficult. Compilations have been made of critical configurations, and these may be used to evaluate the margin of safety. Nevertheless, it is helpful to have generalized criteria against which to evaluate criticality safety. Criteria are now presented that, if any one is met, will ensure subcriticality.

- Criterion 1. If no 1-L sphere (or cube) anywhere in the waste has a plutonium content that exceeds 2.6 g, subcriticality is ensured.

This subcritical limit is based on the minimum critical plutonium concentration in an infinite volume of homogenized waste, assuming the waste has a macroscopic absorption cross section that is at least as large as that for the CWM solids.

- Criterion 2. If the total mass of plutonium is less than 520 g, subcriticality is ensured.

The smallest mass of plutonium that can be made critical in a plutonium-waste-water system is 520 g. This minimum occurs at a plutonium concentration of 30 g/L in pure water when no waste solids are present (Carter et al. 1970). This is the largest mass of plutonium that is ensured to be subcritical under all possible conditions.

- Criterion 3. If there is no cubic volume 58 cm on a side that encloses at least 520 g of plutonium, subcriticality is ensured.

This criterion combines three concepts. First, less than 520 g of plutonium are subcritical for all degrees of moderation, full reflection, uniform (homogeneous) or non-uniform (heterogeneous) distribution of plutonium, and any waste composition. Therefore, no critical spheres are possible. Second, for 520 g of plutonium in a 58-cm cube, the average concentration is less than 2.6 g/L, the minimum which can be made critical. Third, the 58-cm spacing, center-to-center, between adjacent volumes is adequate to ensure neutronic isolation for subcritical optimized spheres. This is true because a high water content would be required to achieve a sphere approaching critical, and at the same time a high water content would ensure neutronic isolation between adjacent spheres.

- Criterion 4. If the plutonium areal density does not exceed $2,582 \text{ g/m}^2$ (240 g/ft^2), subcriticality is ensured.

Carter et al. (1970) provide 240 g/ft^2 as the smallest areal density for which criticality is possible in a plutonium-water system. This value is independent of the plutonium concentration and will increase if waste solids are taken into account. This criterion applies for all waste compositions, thicknesses of waste, degrees of moderation, and degrees of reflection.

Although the above criteria are simple to state, the waste tank environment makes it difficult to guarantee that at least one is met under all conditions.

4.4 CONFIGURATIONS WITH SMALL MASS AND VOLUME

Examples are provided of configurations that achieve criticality with a small mass of plutonium or in a small volume. This section illustrates that for these configurations, the plutonium concentration required to achieve criticality is high. See Section 5.2 for additional critical parameters for spheres.

4.4.1 Minimum Critical Mass

The minimum critical mass is defined as the smallest mass of plutonium (or other fissile nuclide) that can be made critical. In practice, the minimum critical mass is reduced to include a margin of safety to guarantee subcriticality over the range of uncertainties in the derivation. This is called the subcritical mass limit. When the total tank inventory is known to be less than the minimum critical mass, subcriticality is ensured without placing any restrictions on the actual waste composition or the plutonium distribution. Only a few tanks have a plutonium inventory smaller than the minimum critical mass.

The minimum critical mass of plutonium in pure water is 520 g, and this value is conservative when applied to tank waste. Criticality occurs at a plutonium concentration of 30 g/L. This plutonium concentration is more than 10 times larger than the minimum critical concentration in an unlimited volume (i.e., 2.6 g/L).

4.4.2 Small Critical Volume

Criticality is possible in a small spherical volume of 17 L, but the concentration must be 30 g Pu/L with a critical mass of 520 g of plutonium. In a volume of 300 L, criticality occurs at 10 g Pu/L with a critical mass of 3.0 kg of plutonium.

4.5 GENERAL LIMITS FOR LARGE VOLUMES

Examples are provided to illustrate conditions required to achieve criticality in a large volume of waste. For these configurations, criticality is achieved with a low plutonium concentration. Nevertheless, the plutonium concentration expected in tank waste is much lower than the minimum that can be made critical. See Section 5.2 for additional critical parameters for spheres and Section 5.6 for slabs.

4.5.1 Minimum Critical Concentration

The minimum critical concentration is the smallest concentration of plutonium (or other fissile nuclide) that can be made critical over the range of variation possible for all parameters. This means that the water content, an important variable, is optimized. In practice, a slightly smaller value called the minimum subcritical limit concentration is used to ensure subcriticality despite uncertainties in the derivation.

There may be little or no solids in supernatant liquid, and it is conservative to assume that supernatant liquid is all water. In a plutonium-water mixture, criticality is not possible unless the plutonium concentration exceeds 7.2 g/L.

When solids with small neutron absorption cross sections are present, criticality becomes possible at lower concentrations than in a purely plutonium-water solution. This condition occurs in settled solids with a low water content. Under ideal conditions, criticality is possible for a plutonium concentration as low as 2.6 g/L. This value is the subcritical limit for plutonium concentration, based on the CWM with optimal water content.

4.5.2 Large Critical Spherical Volume

To achieve criticality at a plutonium concentration of 4.0 g/L requires a 3.7-m-diameter, 27,000-L-sphere of waste containing 108 kg of plutonium (see Figures 5-1 and 5-2). To achieve criticality at 3.6 g Pu/L requires a 69,400-L sphere containing 250 kg of plutonium. These values are more conservative than reported in earlier studies (see Section 2.2). As the plutonium concentration is decreased below this value, the critical volume will increase rapidly with an asymptote at a concentration of 2.6 g Pu/L. Below this concentration, criticality is not possible. Section 5.2 provides more details.

4.5.3 Criticality in a Slab

The slab against which tank waste is commonly compared has infinite lateral dimensions and is critical at a plutonium concentration of 4 g/L. For this slab to go critical it must be at least 1.7 m thick. Below a plutonium concentration of 2.6 g/L, criticality is not possible no matter how thick the slab. Section 5.6 provides more details.

4.5.3.1 Minimum Critical Areal Density. The plutonium areal density is the total quantity of plutonium above a unit area of floor, including plutonium located all the way to the top surface. The minimum critical areal density for plutonium in water is conservative when applied to waste. The smallest areal density that can be made critical is 2,582 g Pu/m² (Carter et al. 1970). This value is often stated as 240 g Pu/ft². Section 5.6 provides more details.

4.6 MINIMUM SUBCRITICAL ABSORBER-TO-PLUTONIUM MASS RATIOS

Criticality prevention in the Hanford Site high-level waste tanks relies on the dilution of fissile material and the presence of neutron absorber materials mixed with the plutonium compounds in the waste matrix. Water is a major part of the liquid that provides dilution for most in-tank waste solids.

Because the wastes are maintained at a high pH value, most of the plutonium is in the solids (Rai and Serne 1978). Thus, the plutonium concentration in the supernate is so low as to present an insignificant criticality safety concern.

One way to maintain subcriticality in the solid waste is to ensure that there are sufficient neutron absorbers in the waste media such that neutron absorption exceeds the neutron production capability of the fissile material. The following sections discuss the basis for a set of minimum subcritical X/Pu mass ratios, where X is used to designate the absorber type.

It should be noted that the actual-to-minimum subcritical mass ratio is based on homogeneous waste configurations. Although subcriticality is ensured in a homogenous mixture when this ratio is greater than unity, the same cannot be said for a heterogeneous mixture where conditions in one location might be different from another location. Nevertheless, the actual-to-minimum mass ratio is an important parameter because it allows an assessment of the margin of subcriticality based on individual waste samples. The greater the number of waste samples shown to exhibit a high actual-to-minimum mass ratio, the higher the confidence that the waste as a whole exhibits a large margin of subcriticality.

4.6.1 Subcritical Ratios for Uranium, Iron, and Manganese

An important property of uranium, iron, and manganese is that they are insoluble under normal and credible accident conditions found in tank waste. Table 4-2 presents the minimum subcritical X/Pu mass ratios for these elements. The ratios determine the minimum relative concentrations for absorber materials which, if present, provide subcriticality for any degree of moderation. The derivations of the minimum subcritical X/Pu ratios are addressed in the following subsections.

Table 4-2. Minimum Subcritical Absorber-to-Plutonium Ratios to Ensure Subcriticality.

Element or compound	Atomic or molecular weight	Minimum subcritical X/Pu ratio (by weight)
Natural uranium	238.07	770
Iron	55.85	160
Manganese	54.94	32

4.6.2 Minimum Subcritical Uranium-to-Plutonium Mass Ratio

Natural uranium contains 0.72 wt% ^{235}U and 99.28 wt% ^{238}U . Because ^{238}U absorbs so many of the neutrons produced by the fission of ^{235}U , natural uranium cannot be made critical in a homogeneous water uranium system. Criticality remains impossible, even after a small quantity of ^{239}Pu is added, provided the plutonium/uranium mass ratio does not exceed the "always safe" value of 0.0013 (see Table 4-3). For a ratio below this always safe value, criticality cannot be achieved regardless of the type or proportion of other materials present or degree of moderation.

Table 4-3, taken from *Nuclear Safety Guide* (Thomas 1978), shows the limiting mass ratios for plutonium in natural uranium. For mass ratios smaller than those shown, criticality is not possible. For tank waste evaluation, the subcritical limit value of 0.0013 is used to ensure conservatism.

Table 4-3 illustrates that under actual conditions in which the moderation may not be optimized and the moderator may not be pure water, the plutonium concentration required to achieve criticality will increase. Therefore, the actual margin of safety should be greater than obtainable using conservative assumptions as to the uncertainties in the waste composition.

The mass ratio limits in Table 4-3 use the ratio $\text{Pu}/(\text{Pu} + \text{U})$. This evaluation uses a plutonium/uranium ratio instead, because it is more conservative and it simplifies calculations.

Table 4-3. Subcritical Concentration Limits for ^{239}Pu in Mixtures of Plutonium and Natural Uranium in Unlimited Mass.

Material	Mass Ratio $\text{Pu}/(\text{Pu} + \text{U})$
Dry oxide, $\text{H}:(\text{Pu}+\text{U}) = 0$	0.044
Damp oxides, $\text{H}:(\text{Pu}+\text{U}) \leq 0.4$	0.018
Oxides in water	0.0013
Nitrate solutions	0.0065

4.6.3 Iron-to-Plutonium and Manganese-to-Plutonium Mass Ratios

Clemmons and Goslen (1992a, 1992b) calculated the amount of iron and manganese required in a water solution to ensure subcriticality of either ^{235}U or ^{239}Pu . The iron and manganese content is specified as the mass ratio relative to the fissile nuclide. Criticality is not possible regardless of water moderation if the iron- or manganese-to-fissile isotope mass ratios exceed $\text{Fe}/^{239}\text{Pu} = 160$ or $\text{Mn}/^{239}\text{Pu} = 32$.

4.6.4 Other Minimum Subcritical Absorber-to-Plutonium Mass Ratios

In addition to those previously mentioned, other elements and compounds can be considered for minimum subcritical X/Pu mass ratios. This is because of their high neutron absorption cross sections, or because of their relative abundance in the waste. The minimum subcritical absorber-to-plutonium mass ratios for these are shown in Table 4-4. The minimum subcritical ratios are developed for the water content that minimizes the critical plutonium concentration.

A group of waste components selected for their low neutron absorption is shown in Table 4-5. This group is important to criticality safety because it raises the possibility of criticality at very low plutonium concentrations. These components are discussed in more detail in Section 4.6.5.

4.6.4.1 Basis for Mass Ratios. The fact that k_{∞} depends only on the macroscopic cross section for absorption provides a simple method for deriving the mass ratios of other elements, based on known mass ratios. The detailed analyses (Rogers 1993) of the minimum subcritical mass ratios for uranium, iron, and manganese provide the basis for the minimum subcritical mass ratios shown in Tables 4-4 and 4-5. For example, if a large volume of iron and plutonium is slightly subcritical, the mass will remain slightly subcritical when the mass of iron is replaced by another material having an equivalent amount of neutron absorption. This equivalent amount of neutron absorption can then be used to define a mass ratio for the substituted material.

Derivation of values in Tables 4-4 and 4-5 is based on maintaining the thermal neutron macroscopic absorption cross section unchanged. Because a conservative constant of proportionality was used, the subcritical limit ratios are expected to be larger than the true values. Calculations are provided in Appendix A for most of the elements in the tables to verify conservatism of the subcritical mass ratios. For verification, k_{∞} was calculated over the entire range of moderation from extremely dry to over-

Table 4-4. Minimum Subcritical Absorber-to-Plutonium Mass Ratios to Ensure Subcriticality.

Component	Minimum subcritical X/Pu mass ratio
Calcium	770
Chromium	135
Copper	130
Lanthanum	121
Nickel	105
Nitrogen	61
Nitrate	270
Sodium	360
Thorium	243

Table 4-5. Minimum Subcritical Absorber-to-Plutonium Mass Ratios for Low Neutron Absorbers.

Component	Minimum subcritical X/Pu mass ratio
Aluminum	910
Bismuth	48,000
Bismuth phosphate	10,100
Carbon (graphite)	28,000
Silicon dioxide	1,400
Zirconium	4,000

moderated. Because no experimental benchmarks were available to validate these calculations, an upper limit criterion on k_{∞} was set at 0.90 at a 95% confidence level. This criterion was met by all the component mass ratios, except for calcium.

For a calcium to plutonium mass ratio of 720 obtained using the Rogers' formula, the k_{∞} reached a maximum value of 0.903. Although this is well subcritical, it does not meet the conservative k_{∞} criterion stated above. To meet this criterion, the subcritical mass ratio limit for calcium was increased to 770.

These calculations provide confirmation that criticality is not possible at any degree of water moderation when the mass ratios exceed the subcritical limits.

4.6.5 Materials with Low Neutron Absorption Cross Sections

The CWM assumes that the macroscopic absorption cross section will be greater than 0.01096 cm^{-1} . However, some components in waste, if taken alone, exhibit a macroscopic neutron absorption cross section smaller than this value. This raises the possibility that the minimum plutonium concentration that can be made critical might, under some circumstances, be less than 2.6 g/L (the CWM value).

Components of primary interest because of their low neutron absorption cross sections are aluminum, bismuth phosphate, carbon (graphite), silicon dioxide, and zirconium. Table 4-6 shows microscopic and macroscopic cross sections and Table 4-5 shows the minimum subcritical absorber-to-plutonium mass ratios for these components.

Silicon dioxide is a common compound found in sandy soils. Bismuth phosphate comprises a significant portion of some waste streams. Zirconium was used in fuel element cladding and is found in cladding waste. Carbon is a common substance, although in waste it seldom appears in a pure form.

Table 4-6. Components with Small Absorption Cross Sections.

Component	Atomic or mol. weight	Density (g/cm ³)	Microscopic absorption XS (barns)	Macroscopic absorption XS (cm ⁻¹)
Aluminum	26.98	2.699	0.241	0.015
Bismuth	209	9.747	0.034	0.001
Bismuth phosphate	304.0	6.323	0.224	0.0029
Carbon (graphite)	12.0	1.60	0.0034	0.00027
Silicon dioxide	60.06	2.32	0.16	0.0037
Zirconium	91.22	6.4	0.185	0.008

4.6.5.1 Silicon Dioxide. The plutonium concentration necessary for criticality in silicon dioxide is very low. The macroscopic absorption cross section for silicon dioxide is only 0.0037 cm⁻¹. Although waste contains relatively little silicon dioxide, sandy soils surrounding waste tanks may contain a high proportion. Therefore, silicon dioxide is important when evaluating waste liquids that have leaked into the soil.

Sanchez et al. (1996) provide a detailed study of critical parameters for plutonium-silicon dioxide-water systems. This study is helpful in understanding the critical parameters of other compounds with low absorption cross sections.

The minimum critical plutonium concentration in silicon dioxide is provided by Sanchez et al. as 1 g/L, and this occurs when the silicon dioxide is completely dry. However, Altschuler and Doto (1991) provide a slightly lower minimum critical plutonium concentration of 0.8 g/L. In tank waste studies, a subcritical limit value of 0.7 g Pu/L has been used.

When the plutonium critical concentration is low, the critical mass and the critical volume will be large. In silicon dioxide, the plutonium minimum critical mass is 30 kg, and this occurs at a plutonium concentration of about 4 g/L in a completely dry state. Also, it is interesting that the critical mass remains near 30 kg while the plutonium concentration varies between 2 and 12 g/L. Over this range, the plutonium critical mass changes by less than 25%, while the volume varies from 4,000 to 15,000 L. Only for very high plutonium concentrations greater than 1 kg/L will the critical mass fall below the 30 kg minimum for low concentrations.

At a silicon/plutonium mass ratio of 705 (atom ratio of 6,000), the minimum critical plutonium concentration is about 1.5 g/L, and the minimum critical mass is 200 kg of plutonium. At this very low plutonium concentration, a volume of at least 130,000 L of silicon dioxide is required for criticality.

It takes only a small fraction of water to greatly increase the critical mass when the plutonium concentration is low. When the silicon/plutonium mass ratio is greater than 705, an addition of 1 wt% water is sufficient to cause the critical mass to increase to more than 1,000 kg of plutonium.

At a silicon/plutonium mass ratio of 330 (atom ratio of 2,812), the plutonium minimum critical concentration is about 3 g/L, and the minimum

critical mass is 10 kg of plutonium. For this mass ratio the minimum critical mass occurs when the water content is 4 wt%. In other words, optimal moderation is 4 wt% water. When no water is present, the critical mass at this plutonium concentration is greater than 30 kg. At a mass ratio of 330, a water content of 11 wt%, or greater, is sufficient to increase the plutonium critical mass to more than 1,000 kg.

To achieve criticality with a plutonium concentration less than 2.6 g/L, the minimum for CWM solids, the silicon/plutonium mass ratio must be greater than 330. In other words, the addition of silicon dioxide to tank waste solids will not lower the minimum critical concentration until the waste is largely composed of silicon dioxide.

4.6.5.2 Bismuth Phosphate. Bismuth phosphate was an important part of the bismuth phosphate process used to separate plutonium from uranium, and it ranks very low as an absorber of neutrons. Its macroscopic absorption cross section is only 0.0029 cm^{-1} .

For waste composed entirely of bismuth phosphate, the minimum critical plutonium concentration should be between 0.6 and 0.8 g/L, based on a comparison with silicon dioxide. Because of the smaller neutron absorption cross section, the minimum critical mass when no water is present will be less than that of silicon dioxide, but its value has not been determined.

The presence of even low concentrations of water and other absorbers would greatly increase the plutonium concentration required for criticality. For plutonium in pure water, subcriticality is ensured when the hydrogen/plutonium atom ratio exceeds 3,600, and this value holds no matter the plutonium concentration. When water is added to bismuth phosphate, neutron absorption in the bismuth phosphate will reduce the hydrogen/plutonium atom ratio required to ensure subcriticality, and the degree of reduction increases as the bismuth phosphate fraction increases. When the plutonium concentration is 3 g/L in bismuth phosphate, the concentration of water required to ensure subcriticality will be about 0.3 g/L. At 2 g Pu/L, the water concentration to ensure subcriticality will be about 0.15 g/L.

Four major waste streams were sent to underground tanks from the bismuth phosphate process. Two of these, coating waste and metal waste, contained considerable uranium and sodium nitrate. The other two streams, first cycle and second cycle waste, contained considerable sodium nitrate and sodium phosphate. In addition, these streams contained lesser quantities of iron, chromium, and ammonium compounds. For these four streams Agnew (1995) reports the highest bismuth phosphate concentration to be only 0.014 molar (first cycle waste). These waste streams have sufficient absorbers to ensure that the minimum critical plutonium concentration does not fall below the 2.6 g/L found for the CWM.

4.6.5.3 Zirconium. Zirconium was used for fuel element cladding. The plutonium minimum critical concentration in zirconium should be about 1.9 g/L, based on a comparison to silicon dioxide. Because zirconium is only present with other components that have larger neutron cross sections, its presence would not be expected to cause the total macroscopic neutron absorption to fall below that of the CWM solids.

4.6.5.4 Carbon. Carbon has an extremely low neutron absorption cross section of 0.0034 barns, and for graphite the macroscopic absorption cross section is only 0.00027 cm⁻¹. Sanchez et al. (1996) provide a graph of the critical mass of ²³⁵U in graphite that shows a minimum critical concentration of between 0.2 and 0.3 g ²³⁵U/L. The plutonium minimum critical concentration in pure carbon is expected to be similar to that of ²³⁵U.

The minimum critical mass of ²³⁵U in graphite is about 700 g and occurs at a ²³⁵U concentration of about 3 g/L. The spherical critical mass remains below 1.0 kg over a range of ²³⁵U concentration between 0.8 and 12 g/L. This clearly shows that the critical concentration and the critical mass can be significantly reduced in a configuration composed almost entirely of carbon. For criticality at 0.8 g ²³⁵U/L, at least 1,250 L of very pure carbon would be required. The volume of pure carbon required for a critical configuration increases rapidly as the uranium critical concentration decreases below this value.

Because carbon is only present with a large volume fraction of other components that have larger neutron cross sections, its presence would not be expected to cause the total macroscopic neutron absorption to fall below that of the CWM solids. For example, an atom of hydrogen has 98 times the neutron absorption capacity of an atom of carbon, while an atom of iron has more than 700 times the absorption cross section. Only a small fraction of water or other waste material combined with the carbon is required to significantly increase the minimum critical concentration. Looked at another way, a large fraction of carbon must be combined with normal waste before the plutonium critical concentration can fall below 2.6 g/L.

4.7 GENERALIZED SOLIDS/PLUTONIUM MASS RATIO LIMIT

A limit on the solids/plutonium mass ratio is versatile because it applies over all plutonium concentrations and solids densities. It eliminates the need for separate limits on the plutonium concentration and the solids density.

A solids/plutonium mass ratio can be determined by dividing the solids density by the minimum critical plutonium concentration. However, CWM solids have a lower neutron absorption per unit volume, rather than per unit mass, than does real waste. For this reason, a solids/plutonium mass ratio calculated for CWM solids would not be expected to have a degree of conservatism as high as does the critical plutonium concentration.

The subcritical solids/plutonium mass ratio limit is 476 for CWM solids at a density of 1,200 g/L. Although this value is conservative for most waste, it is possible to postulate a few waste compositions for which it does not apply. For example, because the natural uranium/plutonium subcritical mass ratio is 770, a solids/plutonium of 770 should be used for the few tanks with a large fraction of natural or depleted uranium.

When taken alone, aluminum, bismuth, silicon, and zirconium exhibit a solids/plutonium subcritical limit larger than the 770 value for natural uranium. However, those components are always mixed with a large fraction of other components with a smaller component/plutonium mass ratio, such that the

overall mass ratio is less than 770. For this reason, a solids/plutonium mass ratio of 770 is conservative when applied to real waste. Therefore, this value is taken as the subcritical solids/plutonium mass limit for tank waste.

If the solids/plutonium mass ratio is less than the subcritical limit, criticality does not become possible until the region affected is large enough to contain more than a minimum critical mass of plutonium. In practice, waste sent to storage is well mixed, and its mass ratio is greater than the limit.

4.8 COMBINING THE EFFECT OF SEVERAL ABSORBERS

Waste is made up of many components. For this reason it is more realistic to base an evaluation of subcriticality on the combined effects of all the neutron absorbers rather than on a single component.

4.8.1 Relationship Between k_{∞} and Neutron Absorption

Two factors determine the degree of subcriticality in tank waste. These are the ability of the waste to either absorb neutrons (with or without a resulting fission) or to scatter neutrons. In a very large system from which very few neutrons escape, k_{∞} is almost independent of the neutron scattering, and the value of k_{∞} is determined almost entirely by the macroscopic absorption cross section. This means that k_{∞} for an infinitely large system will remain nearly unchanged when the composition of the waste is changed, as long as the macroscopic absorption cross section remains unchanged. It is this relationship that provides the basis for summing the actual-to-minimum subcritical ratios. For a given total sum, the total neutron absorption will remain unchanged regardless of the actual components of the waste, and consequently, k_{∞} will remain unchanged.

A simple definition of k_{∞} can be expressed as the neutron production divided by the neutron absorption. The formula is as follows.

$$k_{\infty} = \frac{\nu \Sigma_f}{\Sigma_a}$$

where:

$$\begin{aligned} \nu \Sigma_f &= \text{Neutron production term} \\ \Sigma_a &= \text{Neutron absorption term.} \end{aligned}$$

4.8.2 Sum of Actual-to-Minimum Subcritical Ratios

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

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

- Calculate the actual X/Pu for each waste component. Divide each actual mass ratio by its corresponding minimum subcritical mass ratio to obtain a fraction. This fraction will be referred to as the actual-to-minimum subcritical mass fraction.
- When the sum of the actual-to-minimum subcritical mass fractions for all individual components is greater than unity, the waste is subcritical.

Therefore

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

where:

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

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

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

Even if the sum of the fractions for a waste sample is determined to be less than 1.0, the waste configuration may still be subcritical. For example, uncertainty in the analyzed composition may be high, not all the waste components may have been included in the calculation, etc. However, additional evaluation is required to demonstrate subcriticality.

4.9 BORON AND CADMIUM

Boron and cadmium are effective absorbers of neutrons. Boron in waste is assumed to be natural boron composed of 20 wt% ^{10}B and 80 wt% ^{11}B .

If the ratio of hydrogen to fissile atoms exceeds 250, subcriticality is ensured when either the cadmium/fissile atom ratio exceeds 0.5 or the boron/fissile atom ratio exceeds 2.0. (Fissile refers to ^{235}U , ^{233}U , and ^{239}Pu .)

Cadmium and boron are not included in the CWM because their continued presence can not be guaranteed. Both of these elements are very strong neutron absorbers, and the presence of either element in any significant quantity would dominate the neutron absorption. As a practical matter, it may be impossible to guarantee the presence of these poisons mixed uniformly throughout the waste. Still, the large neutron absorption of cadmium and boron means they are potentially important to subcriticality studies. If their presence is independently verified, they provide additional assurance of subcriticality.

4.10 URANIUM

In a uranium-water system that is homogenized, criticality is not possible in tank waste unless the enrichment exceeds 1.03 wt%. Nearly all uranium sent to tank storage contains less than 0.72 wt% ^{235}U and falls under the description of natural or depleted uranium. There is no chemical or physical means by which the ^{235}U can be separated from the ^{238}U to inadvertently cause a criticality.

Although criticality is possible at enrichments as low as 0.72 wt% in precisely defined heterogeneous arrays with low neutron-absorbing moderators, such as graphite or heavy water, these configurations would be impossible to achieve in tank waste.

5.0 WASTE GEOMETRIES

Many parameters, some independent and some interrelated, work together to determine whether a critical configuration is possible. The complexity of waste makes it difficult to analyze the interplay of these parameters.

When the solids content, the plutonium content, and the liquid (water) content are the same at every location, they are said to be homogeneous. "Real" waste is non-homogeneous, and this makes it difficult to evaluate criticality safety. Nevertheless, it is possible to greatly simplify most configurations by replacing them by conservative, homogeneous configurations. Critical parameters based on simple homogeneous configurations are used to assess criticality safety.

Figures used in this report were selected to best illustrate the margin of safety for tank waste. Additional figures of critical parameters for a mixture of plutonium and waste solids in water are found in Braun et al. (1994).

To be conservative, a configuration having a k_{eff} of 0.935 was assumed to be critical. For finite volumes, a 25.0-cm-thick water reflector was assumed to surround the critical region. The CWM was used for the solids composition.

5.1 INFINITE HOMOGENEOUS GEOMETRY

The only acceptable condition for tank waste is for k_{∞} to be much less than 1.0. At a plutonium concentration of 1.0 g/L, k_{∞} will always be less than 0.6.

In a purely water solution, k_{∞} is equal to 1.0 at a plutonium concentration of 7.2 g/L. When the plutonium concentration is less than 2.6 g/L, k_{∞} will never exceed 1.0, even in an unlimited volume of CWM solids when completely dry.

For plutonium concentrations ranging from 1.0 to 8.0 g Pu/L in waste, k_{∞} increases as the proportion of solids increases and reaches a peak value for waste that is almost completely dry. At the very low plutonium concentrations that occur in tank waste under normal circumstances, optimal moderation (i.e., highest k_{∞}) occurs when the water content is very low.

5.2 SPHERES

A sphere is the geometry in which criticality can be achieved with the smallest mass of plutonium, the smallest volume, or the lowest plutonium concentration. Other compact geometries of the same volume (i.e., cubes and cylinders with a height/diameter ratio of about 1) will have similar critical parameters. Therefore, critical parameters for spheres can be used as a conservative estimate of critical parameters for all other shapes.

The subcritical limit on plutonium concentration in a water solution is 7.2 g Pu/L. However, in waste solids it is possible to create critical

configurations with a lower plutonium concentration than is possible in pure water. In CWM solids, criticality is not possible when the plutonium concentration is less than 2.6 g/L.

As the plutonium concentration increases, the mass required for criticality decreases. When the plutonium concentration is 3.6 g/L, the smallest mass required for criticality is 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.

As the plutonium concentration increases, the difference between the minimum critical mass in pure water and in CWM solids decreases. Above 10 g P/L, minimizing the critical mass for CWM solid requires that the solids content be reduced to zero, thereby resulting in a mixture of plutonium in pure water. This explains the merging of the CWM solids and pure water curves shown in Figures 5-1 through 5-4.

For critical spheres of CWM solids and water, the following general observations are made (Braun et al. 1994).

- When the plutonium concentration is lower, the smallest critical sphere diameter occurs at a water content that is lower, and the range of water content for which criticality is possible is narrower.
- When the plutonium concentration is less than 5 g/L, the critical diameter is large. Criticality is not possible when the water content is high.
- At a plutonium concentration of 8 g/L, the critical sphere diameter remains relatively unchanged over a wide range of water content from 100 vol% down to less than 30 vol%.
- As the plutonium concentration increases, the smallest critical diameter occurs at a higher water content. Above 8 g Pu/L, the smallest critical diameter occurs for plutonium in pure water.

Figure 5-1 shows the sphere minimum critical diameter as a function of plutonium content for three different postulated waste compositions: (1) CWM solids in water, (2) pure water, and (3) a saturated solution of sodium compounds (nitrate, nitrite, aluminate, and hydroxide) to simulate "real" waste. For CWM solids, the water content is optimized to provide the smallest diameter. A 25-cm-thick, close-fitting water reflector is assumed.

Calculations for Figure 5-1 and other figures that follow were made using the WIMS-E computer code (Gubbins et al. 1982). These calculations are discussed in Appendixes B and C. Because phosphorus was not available in the WIMS-E cross-section library, the phosphorus in CWM solids was replaced by an equivalent mass of silicon to maintain the macroscopic absorption cross section unchanged.

Figure 5-1. Sphere Minimum Critical Diameter versus Plutonium Concentration.

CWM = conservative waste model

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The asymptote values shown in Figure 5-1 are for the plutonium concentrations for which k_{sc} equals 0.935 in each of the waste media. The 6.5 g/L asymptote for water is less than the subcritical limit of 7.2 g/L used elsewhere in this report. This difference derives from basing the asymptote on a subcritical limit of 0.935 for k_{sc} , whereas the larger subcritical limit is based on experiments and is more precise. The smaller value shown in Figure 5-1 verifies that the curves are calculated with a conservative bias.

A saturated solution of sodium compounds was used to simulate "real" waste, based on a composition described by Reynolds and Herting (1984). The composition selected for the calculations provides the largest k_{sc} from among nine compositions evaluated. Table 5-1 summarizes the composition of this waste, which can be described as a saturated supernatant liquid (see Appendixes B and C for more details). Its density is 1.42 g/cm³, with 50.6 wt% in solutes and the remaining 49.4 wt% being water.

Table 5-1. Composition of Simulated Supernatant Liquid.

Constituent compounds	Wt%	Constituent elements	Wt%
NaNO_3	32.4	Hydrogen	5.7
NaNO_2	9.7	Nitrogen	7.3
NaAlO_2	2.9	Oxygen	70.0
NaOH	5.6	Sodium	16.0
H_2O	49.4	Aluminum	1.0

Figure 5-2 shows the sphere minimum critical mass as a function of plutonium content for three different postulated waste compositions: (1) CWM solids with water optimized, (2) pure water, and (3) a solution of sodium compounds intended to simulate "real" waste as a saturated supernatant liquid. A 25-cm-thick, close-fitting water reflector is assumed.

Figure 5-2. Sphere Minimum Critical Mass versus Plutonium Concentration.

—●— CWM solids —■— pure water —▼— supernate

CWM = conservative waste model

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5.3 HEMISPHERES

Under some circumstances, waste entering a storage tank might form a pile of solids under the inlet pipe in the shape of either a hemisphere or a cone. In practice, a hemisphere would be the most conservative shape for such a pile of solids. A hemisphere and a cone would require greater volumes of solids than a sphere to achieve criticality, given the same plutonium concentration. Therefore, it is conservative to compare the volume to that of a critical sphere. Only when waste is well characterized can the shape of a pile be taken into account when evaluating the criticality potential of the waste.

5.4 CYLINDERS

Plutonium-bearing waste might occupy a cylindrical volume. The maximum height of such a cylinder would be the depth of the settled waste solids. A cylinder for which the height to diameter ratio is near 1.0 will result in critical parameters very nearly the same as those for a sphere of the same volume.

If the waste region of interest occupies the total cross-sectional area of the tank, the cylinder becomes effectively a slab, and critical parameters for slabs apply. A cylinder for which the height to diameter ratio is small can be compared to a slab.

5.5 ANNULAR CYLINDERS AND TOROIDAL VOLUMES

Waste falling from a height and flowing radially outward from a central point might deposit solids in a ring. A slurry distributor is capable of creating waste distributions in the shape of an annular cylinder or toroid. A toroid is similar in shape to an annular cylinder. From a criticality safety perspective, qualitative arguments used for annular cylinders may often be used for toroidal volumes.

When the central diameter of the annular cylinder or toroid is large, interaction of neutrons across the central region will become small. When the central region is filled with waste solids or liquids, the opposite sides may be isolated from each other. The annular cylinder then resembles a slab bent into a circle and the toroid resembles a cylinder bent into a circle. The critical plutonium concentration would then be about the same as that for the geometry that these shapes resemble. An estimate of the critical mass can be made by multiplying the critical concentration by the volume.

It is conservative to replace an annular cylinder by a solid cylinder of the same height and volume. The critical mass of plutonium in the solid cylinder will be less.

The critical volume and the critical mass for an annular cylinder or in a toroid will be greater than for a sphere, and in many cases they will be much greater. The critical plutonium mass may be found to be very large for waste in an annular cylinder or toroidal geometry. However, caution must be used when evaluating criticality safety. Even though the critical mass of a toroid may be large, a relatively small increase in plutonium concentration or a

small change in shape over a portion of the toroid may dramatically change the conditions under which criticality is possible. In assessing criticality safety, the potential for changing shape and composition must be taken into account.

5.6 SLABS

The most probable final geometry for a waste stream is a slab occupying the entire cross-sectional area of the storage tank.

Figure 5-3 shows the minimum critical slab thickness as a function of the plutonium concentration for three waste compositions: (1) CWM solids with water optimized, (2) water, and (3) a saturated solution of sodium compounds (primarily nitrate) intended to simulate "real" waste. Calculations were performed using the WIMS-E computer code. For the CWM, the value shown is the minimum over the range of possible water content. A 25-cm-thick water reflector is assumed on top and bottom.

Figure 5-4 shows the slab minimum critical plutonium mass as a function of the plutonium concentration in a 22.9-m-diameter tank for three waste compositions: (1) CWM solids with water optimized, (2) water, and (3) a saturated solution of sodium compounds (primarily nitrate) intended to simulate "real" waste. The plutonium mass is calculated assuming the waste covers the entire area of the tank at a uniform depth, and there is a 25-cm-thick water reflector on top and bottom.

Figure 5-3. Slab Minimum Critical Thickness versus Plutonium Concentration.

CWM = conservative waste model

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Figure 5-4. Slab Minimum Critical Mass versus Plutonium Concentration in 22.9-m- (75-ft-) Diameter Tank.

CWM = conservative waste model

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Over the range of all plutonium concentrations, the minimum critical plutonium mass in a uniform slab is shown in Figure 5-4 as somewhat less than 1,000 kg, based on a subcritical limit on k_{eff} of 0.935. When a less conservative, but well established, minimum critical areal density of 2,582 g/m² is used, the minimum critical mass is 1,060 kg (see Section 5.6.1). This larger value is generally used in discussions of criticality safety. It is important to note that the minimum critical plutonium mass in a uniform slab occurs at a high plutonium concentration.

The following observations are made concerning the relationship between the waste critical slab thickness and solids concentration (Braun et al. 1994).

- For a plutonium concentration of 5 g/L, or less, criticality is not possible unless the water content of the waste is less than 50 wt%. The minimum critical slab thickness occurs when the water content is less than 25 wt% and the waste solids content is more than 75 wt%.
- The infinite slab critical thickness increases rapidly as the plutonium concentration decreases below 5 g/L plutonium (see Figure 5-3).

- At a plutonium concentration of 8 g/L, the critical slab thickness remains almost unchanged over a wide variation in the water content. Optimum moderation occurs when the solids content is 40 wt%.
- As the plutonium concentration increases, the minimum critical slab thickness occurs at a higher water content and a smaller solids content.

5.6.1 Plutonium Critical Areal Density in Slabs

The plutonium areal density is defined as the total mass of plutonium in a column of waste extending from a unit area of floor to the top of the waste. The minimum areal density of plutonium that can be made critical for an optimized plutonium-water mixture is 2,582 g/m². This is often expressed as 240 g Pu/ft². When the areal density is less than the minimum critical areal density, criticality is impossible for a uniform slab, regardless of the slab thickness or the degree of moderation.

Figure 5-5 shows the minimum critical areal density as a function of CWM solids content, based on criticality corresponding to a k_{eff} of 1.00 with a 25-cm-thick water reflector on top and bottom. Curves are shown for the addition of solids having a density of 1.2 g/cm³ and for solids having a density of 3.6 g/cm³. The minimum critical areal density, when plotted as a function of the water to solids weight ratio, is almost identical for the densities of the two solids.

The minimum critical areal density for plutonium in pure water was 2,582 g Pu/m², the same as established by Carter et al. (1970). The addition of solids to a plutonium-water solution increases the minimum critical plutonium areal density. Therefore, a subcritical limit of 2,582 g/m² on the plutonium areal density is conservative for all compositions of waste solids and water.

For a uniform layer of waste containing plutonium at the minimum critical areal density, the tank inventory required to achieve criticality in a 22.9-m-diameter tank is 1,060 kg of plutonium. This quantity exceeds the estimated total inventories for all the tanks combined, and is 10 times greater than the largest inventory expected in any single tank.

5.7 COMPLEX WASTE GEOMETRIES

When the distribution of waste components is not uniform, the conditions under which criticality is possible become more complex.

5.7.1 Vertical Stratification (Layering)

Waste was sent to tank storage in a series of waste streams. A waste stream is defined as the waste discharged from a single process. In tank storage, this entire volume of waste appears as a single layer of more or less

Figure 5-5. Minimum Critical Areal Density versus Conservative Waste Model Solids Content.

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uniform composition. Waste within a single storage tank is often composed of several waste streams that may form distinct layers. This process of forming layers is referred to as vertical stratification or layering.

Even though waste in a storage tank may consist of a number of distinct layers, critical parameters developed based on the CWM can be used to represent the entire contents of the tank. The CWM was developed to represent a wide variety of waste so that all waste types can be replaced by the CWM, and the result will be more conservative than the original combination of waste types.

5.7.2 Variation Within a Layer

Mixing of waste streams may create a waste composition that is not uniform even within a given layer. Nevertheless, if individual regions of waste can be replaced by the CWM, the entire layer of waste can be replaced by the CWM. Therefore, variations in composition within a layer will not lower the minimum critical concentration below that of the CWM.

5.7.3 Localized Pockets of Plutonium

The possibility exists that unknown circumstances might have caused pockets of high plutonium concentration to form. The great volume of waste, its chemical and physical complexity, and the lack of good characterization information make it difficult to ensure that there are no pockets of plutonium large enough to create a criticality safety concern.

Two things are required for criticality to occur. The plutonium concentration must be far greater than permitted by the discharge limits, and a large mass of plutonium must be concentrated into a localized volume. Either of these conditions alone is extremely unlikely, and it is far less likely that they would occur simultaneously and result in a criticality.

5.7.4 Lattice and Matrix Formation

A lattice of enriched uranium rods under idealized conditions may have a smaller critical mass of ^{235}U than does a homogeneous mixture. This phenomenon is made possible by absorption resonances in ^{238}U . However, to achieve the lower critical mass requires the uranium to be in the form of high-density rods in a water lattice. Such a configuration is not possible in tank waste because the uranium is in the form of small particles mixed more or less homogeneously with other waste.

Plutonium does not have absorption resonances like those in ^{238}U , and its critical mass will not decrease when formed into a lattice. Lumping plutonium into particles (or pellets) increases the neutron self-shielding. In other words, slow neutrons entering the lump tend to be absorbed near the surface, and the plutonium atoms near the center do not "see" as many slow neutrons as plutonium near the surface or outside of the lump. This effect increases the total quantity of plutonium required to achieve criticality. A lower fraction of neutrons will then be absorbed by the plutonium, and a higher fraction will be absorbed by the water and waste solids. For an infinite volume of waste containing a more or less uniform lattice of plutonium particles (or pellets), the mass of plutonium per liter required to achieve criticality will be greater than when the plutonium is homogeneously mixed with the waste. The minimum critical plutonium concentration for a homogenized configuration is conservative when applied to a non-uniform plutonium distribution.

5.7.5 Cratering

Waste entering from a point high above a sludge layer can acquire a significant velocity to splash and disperse as it impacts the waste layer or tank bottom. The splashing could cause a "crater" in the sludge layer, consisting of a central depression surrounded by a raised ring of material. The degree of subcriticality will increase when a crater is formed because fissile-bearing material is being removed from a region of higher neutron importance (inside the slab), and is being deposited on the top surface of the slab in a location of lower neutron importance.

6.0 MARGIN OF SUBCRITICALITY

A brief discussion is made of the margin of safety (subcriticality) for actual waste. This section is intended to provide the reader with a concise overview of the actual state of stored waste. The term "margin of safety" refers to the difficulty of moving from the actual state of the waste to the hypothetical critical state. The margin of safety is often difficult to quantify. Usually, barriers that prevent criticality from occurring are described and assurances provided that they will not be breached. The margin is said to be "low" when a criticality accident is conceivable and "high" when it is not. For tank waste, the only acceptable margin of safety is "high."

Although a compilation of critical parameters against which to evaluate tank waste is important, the parameters themselves do not provide an assessment of the actual margin of safety. A determination of the margin of safety requires an assessment of the actual conditions of the waste. In addition, it is necessary to assess the likelihood that changes in the waste might alter the condition of waste to make a criticality possible. The considerable complexity of tank waste, the uncertainties associated with its characterization, and the need to periodically change storage conditions require that a criticality safety program be maintained to monitor tank waste.

6.1 ACTUAL-TO-MINIMUM SUBCRITICAL FRACTIONS FOR SELECTED SAMPLES

The margin of subcriticality for tank waste is evaluated by determining actual-to-minimum subcritical fractions. Rogers (1993) lists the chemical composition of core samples taken from tank waste. Waste compositions derived from core samples represent the most complete and accurate compositions available at that time. For this reason, mass ratios derived from these samples should provide a good indication of what should be expected from actual waste. Table 6-1 shows actual-to-minimum subcritical fractions for selected components for the five samples having the largest plutonium concentrations.

Table 6-1. Actual-to-Minimum Subcritical Fractions for Selected Core Samples.

Component	Actual to minimum subcritical fractions				
	Tank 241-A-102 Sample 2	Tank 241-A-106 Sample 2	Tank 241-C-103 Sample 1	Tank 241-C-103 Sample 2	Tank 241-C-106 Sample 1
Aluminum	0.77	0.67	0.09	0.14	0.90
Iron	3.80	5.04	3.46	3.90	6.56
Manganese	2.76	1.45	0.42	1.87	11.58
Nitrate	13.83	6.47	0.03	0.13	0.07
Sodium	13.66	11.54	0.59	1.53	6.54
Uranium	0.07	0.005	0.01	0.05	0.01
Sum	34.89	25.18	4.60	7.62	25.66

The sum of the actual-to-minimum subcritical fractions for the five samples shown in Table 6-1 ranges between 4.6 and 35. For these samples, the plutonium concentration ranges between 0.214 and 0.06 g/L. The average plutonium concentration in 28 core samples is reported to be 0.032 g/L, and the mean plutonium concentration for a larger number of solid waste samples is 0.034 g Pu/L (Braun et al. 1994).

Because the plutonium concentrations for samples shown in Table 6-1 are from two to seven times higher than the average for all tank waste, the sum of the actual-to-minimum subcritical fractions for each of these samples is expected to be smaller than for most waste samples. This is true because the ratios are obtained by dividing by the plutonium concentration.

In practice, the chemical analyses of samples have often been limited in scope and may not have provided a complete representation of the waste. In those cases where the waste samples are incomplete, it may not be possible to calculate some of the minimum subcritical fractions. When a sample is incomplete, the sum of the actual-to-minimum subcritical fractions will be smaller than it would have been if sufficient data were available.

Because the exact nature of the chemical processes occurring within the waste is uncertain and because of the heterogeneity of the waste, there is a concern that the plutonium and neutron absorber concentrations might vary from one region to another within the waste. However, it is highly unlikely that the plutonium could be separated from neutron-absorbing components in the waste. Therefore, even with a heterogeneous waste, the sum of the actual-to-minimum subcritical fractions for insoluble components is a good measure of subcriticality.

6.2 CALCULATION OF k_{∞} FOR SELECTED SAMPLES

An assessment was made of the margin of criticality safety afforded by the composition of the tank waste (Braun et al. 1994). Waste samples were selected on the basis of their apparent low neutron absorber content.

The k_{∞} values were calculated for five analyzed waste compositions to represent waste configurations that would actually occur in a tank. These samples were selected from among several hundred available samples because they were expected to show the highest k_{∞} . Of the five samples, the largest k_{∞} value was 0.0826, the second largest was 0.0596, and the other three samples indicated a k_{∞} value less than 0.02. However, Braun et al. also report three additional samples were found to have unexpectedly high plutonium concentration and very low solids contents. One of these samples reports an extraordinarily high plutonium concentration of 0.10 wt% and a specific gravity of 1.5. This reported plutonium content of 1.5 g/L probably represents a value rounded upward to the nearest 0.1 wt%, and therefore represents only an upper limit value. Nevertheless, k_{∞} is only 0.3221, and the sample composition still represents waste that is well subcritical.

6.2.1 Decreasing the Water Content

The impact of removing water from the waste was examined using the above waste sample with the largest k_{∞} . For the actual waste compositions, k_{∞} remains very low for all degrees of waste moderation. Therefore, the plutonium concentration was arbitrarily increased to 4.0 g/L for a selected sample of waste. A high concentration of plutonium magnifies the effect of changing the water content. The actual water content in the sample is 559 g/L, and k_{∞} for this large plutonium concentration was 0.5079.

Next, k_{∞} was calculated as a function of reduced water content down to 35 g/L of water, a value only 6.3% of the original water content. As water is removed, k_{∞} increases. When the water content is 35 g/L, the value of k_{∞} is 0.6937. In practice it is impossible to remove this much water because some water will remain tightly bound as water of hydration. Nevertheless, the waste remains subcritical with these very conservative assumptions.

6.2.2 Increasing the Plutonium Content

Calculations were made assuming a water content of only 50 g/L to represent waste compositions after the drainable water has been removed. In addition, the plutonium concentration was assumed to be 3.0 g/L, a value more than 100 times larger than the average from among all available samples, and the density of dry solids was conservatively assumed to be only 850 g/L.

From the 10 waste samples described above, 5 were selected to represent waste likely to show a high k_{∞} . From these five samples, the largest value of k_{∞} for dry waste was 0.7468. The second largest value of k_{∞} was 0.4207. Therefore, even with a plutonium concentration much larger than expected and after drainable water has been removed, the waste compositions examined would still remain subcritical.

6.2.3 Removing Neutron Absorbers

Calculations were performed to assess the consequence of removing the following neutron absorbers: cadmium, boron, silicon, calcium, potassium, and mercury. These components were removed from the 10 analyzed waste compositions, and k_{∞} was recalculated. The change in k_{∞} was small. The largest value of k_{∞} for any of the revised samples was 0.1028, and the value of the original sample was 0.0596. These calculations show that a low concentration of plutonium, combined with a high water and solids content, ensures a large margin of subcriticality.

When the water content was reduced to 35 g/L, k_{∞} was 0.1958 without the above neutron absorbers.

6.3 TANK-AVERAGED PARAMETERS

When the total mass of plutonium and the total volume and mass of waste solids and liquids are known, the average plutonium concentration and the average solids/plutonium mass ratio can be found. This average value of a

parameter for tank farms will remain unchanged when waste is transferred between tanks, and it will change only slightly, if at all, when new waste is sent to tank farms.

6.3.1 Average Mass Ratios for Tank Farms

Roetman et al. (1994) estimate the total quantity of plutonium discharged to tank storage to have been 981 kg since the beginning of plutonium production. Volumes of various waste types are reported by Hanlon (1996). Based on Hanlon's information, the total volume of plutonium-bearing solids is conservatively estimated to be 62,297,000 L.

Using a volume of 62,297,000 L and a plutonium mass of 981 kg, the average plutonium concentration for tank farms is 0.0157 g/L of solids. Because a low estimate of volume and a high estimate of plutonium are used, this global average plutonium concentration should be larger than the actual value. Based on a conservative solids density of 1,200 g/L, the global average solids/plutonium mass ratio for tank farms is 74,500.

6.3.2 Average Mass Ratios for Selected Tanks

Presently, the Plutonium Finishing Plant is the primary facility sending waste to tank farms. This waste is sent to tank 241-SY-102, and the June 1996 estimate of its plutonium inventory is 41.4 kg of plutonium. For tank 241-SY-102, Hanlon (1996) reports the volume of solids and liquids to be 268,700 L and 1,170,000 L, respectively. Based only on the volume of solids, the average plutonium concentration is 0.154 g/L, and the solids/plutonium mass ratio is 7,790 (assuming a solids density of 1,200 g/L). This is the lowest ratio for any tank.

The largest fissile material inventories for any storage tank in tank farms are reported for tank 241-C-104. Assuming an 89-kg plutonium inventory and using a 1,117,000-L volume of sludge, the average plutonium concentration is 0.08 g/L, and the average solids/plutonium mass ratio is 15,055.

6.4 CONCENTRATING FACTORS

If a process (either chemical or physical) within the stored waste were to cause plutonium to concentrate, the likelihood of criticality would be related to the effectiveness of the process and to the total quantity of plutonium available. The concept of a "concentrating factor" might be used to describe the effectiveness of the process. The concentrating factor is defined as the highest plutonium concentration achieved by the process, divided by the initial (background) concentration. In the tank waste criticality safety evaluation, the primary utility of concentrating factors is to show how large of a plutonium inventory would be required to achieve criticality for a hypothetical concentrating process.

When a concentrating process is hypothesized, the total plutonium inventory that would be required in a tank to result in criticality depends on the concentrating factor assumed and on the volume of waste solids in the

tank. A smaller volume of waste requires a smaller plutonium inventory to achieve criticality. As an example, consider a concentrating process that causes criticality with 250 kg of plutonium at a concentration of 3.6 g/L. This requires a volume of 69,400 L, which is only 1.8% of the volume available in a 3.78 million-L tank. If one assumes a concentrating factor of 50, the plutonium concentration in the background waste would be 0.07 g/L, and the background inventory would be 272 kg. The total tank inventory would be 522 kg. For a concentrating factor of 100, the total inventory of the tank would be reduced to 386 kg. If the waste volume is reduced, the background inventory would be reduced proportionately.

The concept of a concentrating factor shows that the tank plutonium inventory for a hypothetical criticality would likely be significantly larger than the minimum required to achieve a critical configuration. In addition, it shows that a highly effective mechanism for concentrating plutonium is required when the initial plutonium concentration is very small. The primary utility of concentrating factors would be to show that the probability of a criticality would likely be far less than indicated by the requirements of the critical configuration taken by itself.

6.5 REAL WASTE

A comparison was made between the plutonium critical mass as a function of plutonium concentration in CWM waste and in "real" waste. The increase in plutonium concentration required for real waste is a measurement of the inherent conservatism for evaluations based on parameters obtained using CWM waste. In other words, this difference indicates a larger margin of safety for "real" waste, in comparison to that indicated by the evaluations.

6.5.1 Description of Average Waste Composition

To make this comparison requires that a "real waste" be defined, even though tank waste is highly inconsistent in its composition. In reality, no single waste composition can be said to represent all tank waste. Therefore, this comparison must be viewed as an indication of the most likely difference and cannot be applied to specific waste. For this comparison, "real waste" is separated into a liquid fraction and a solids fraction. The liquid fraction is assumed to have the composition of saturated supernatant liquid described in Section 5.2. This composition was selected from five compositions proposed by Reynolds and Herting (1984) as representing typical supernatant liquid. Of these five supernatant liquids, the selection was made on the basis of being the most conservative with respect to critical parameters.

The solids portion of the waste is defined on the basis of each component being of average concentration. The "Average Waste Solids Composition" was derived from 28 samples of tank waste documented in Table B-1 of *An Analytical Model for Evaluating Subcritical Limits for Waste in Hanford Site Storage Tanks* (Rogers 1993). Rogers lists the average concentrations in grams per liter of 39 elements and compounds commonly found in tank waste. However, the cross sections of only 19 of these constituents were available in the 1986 WIMS-E cross-section library, and for this reason only those 19 components are

used in this comparison. Table 6-2 lists these components and their average concentrations in tank waste. Although if all 39 components were used in the calculation, the critical plutonium concentrations would change, that change would not be expected to be large and the basic conclusions would remain the same. The comparison between CWM waste and "real" waste is qualitative and is intended only to provide a reasonable estimate of what might be expected.

To create an "average waste composition," the average solids composition described above was combined with the realistic supernate solution (Reynolds and Herting 1984) to produce a mixture with a density very nearly equal to the average density of the 28 samples. This mixture contained 10.35 wt% solids, 44.32 wt% water, and 45.33 wt% supernate solutes, and the mixture density was 1587 g/L. This waste contains a large fraction of water and is referred to as "wet waste."

Table 5-2 contains a description of the simulated supernatant liquid. This liquid composition is renormalized in Table 6-3 to show the composition after the waste solids have been added. Component concentrations are given as weight percents of the total waste, and the total weight percent is less than 100 wt% because solids content is not included.

A "dry" version of waste having an average waste composition was created by removing all water from the "wet" composition, while maintaining a constant volume. This results in a dry waste density of 883.8 g/L. This dry mixture contained 18.60 wt% solids and 81.40 wt% supernate solutes.

The component concentrations for the total waste, including the solids portion, are shown in Table 6-4. The composition of the dry waste differs from that for the wet waste only in that the water has been removed, and the components have been renormalized.

6.5.2 Comparison of Waste Types

Results of calculations are shown in Figure 6-1. The most conservative curve is that for CWM solids with optimized water. This curve represents a conservative lower limit on the concentration of plutonium which can be made critical in tank waste, and is used as a basis for setting limits on waste being discharged to tank farms. The minimum plutonium concentration for which criticality is possible is 2.6 g/L.

If water is allowed to drain and to evaporate from the waste over a period of time, there will still be water left in the waste. In practice, there is a lower limit on water content and water content would not likely fall below 25 wt%. The curve of critical mass for 25 wt% content requires a higher plutonium concentration for criticality. The minimum critical plutonium concentration increases to 4.2 g Pu/L.

In simulated real waste the concentration of plutonium required for criticality increases dramatically. The minimum plutonium concentration for criticality in fully water-saturated waste becomes 11 g/L, more than four times as large as the value in dry CWM waste.

Table 6-2. Average Concentrations of 19 Tank Waste Constituents.

Waste constituent	Concentration (g/L)	Waste constituent	Concentration (g/L)
Ag	0.163	Fe	37.91
Al	35.75	Mg	4.51
Am-241	0.0003	Mn	1.97
B	0.019	Ni	1.99
C (CO ₃)	13.4	Pb	1.26
C (TOC)	4.71	Si	47.01
Cd	0.172	⁹⁹ Tc	0.015
Cr	4.05	U	15.3
Cu	0.216	Zr	4.97
F	4.42	--	--

Table 6-3. Supernate Composition of the Average Waste Compositions.

Supernate constituent	Concentration wet waste (wt%)	Concentration dry waste (wt%)
NaNO ₃	29.01219	52.10239
NaNO ₂	8.69395	15.61350
NaOH	5.03971	9.05126
NaAlO ₂	2.58254	4.63742
H ₂ O	44.31717	0.00000
Total wt%	89.64556	81.40457

Table 6-4. Elemental Composition of "Average" Waste.

Waste component	Concentration (wt%)		Waste component	Concentration (wt%)	
	Wet	Dry		Wet	Dry
Ag	0.010	0.018	Mg	0.284	0.510
Al	2.252	4.045	Mn	0.124	0.223
²⁴¹ Am	0.00002	0.00003	N	6.546	11.756
B	0.0012	0.0022	Na	14.365	25.799
C	0.466	0.836	Ni	0.125	0.225
Cd	0.011	0.019	O	63.474	43.308
Cr	0.255	0.458	Pb	0.079	0.143
Cr	0.255	0.458	Si	2.962	5.319
F	0.228	0.500	⁹⁹ Tc	0.0095	0.0017
Fe	2.389	4.289	U	0.964	1.731
H	5.086	0.228	Zr	0.313	0.562

Figure 6-1. Comparison of Four Waste Types.

CWM = conservative waste model

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When water is removed from simulated real waste, the critical mass curve changes dramatically. As the waste becomes drier, the mass of plutonium required for criticality increases. For completely dry waste at a plutonium concentration of 14 g/L, more than 450 kg of plutonium is required for criticality. For dry waste, the critical mass decreases as the plutonium concentration increases, but at a slow rate. In real waste, the presence of a significant fraction of water is required before criticality is possible with less than 200 kg of plutonium.

In dry real waste, criticality is possible below 11 g Pu/L, but only for a plutonium mass exceeding 500 kg. Between 600 and 700 kg of plutonium, water content has relatively little effect on critical size. Over this narrow range of size, criticality is possible for plutonium concentrations between 10.5 and 11.5 g/L, regardless of water content. At less than 10.5 g Pu/L, criticality requires the waste to be very dry and the plutonium mass to exceed 500 kg. At 8.9 g Pu/L, more than 1,000 kg of plutonium is required for criticality.

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7.0 SPECIFIC CONDITIONS AFFECTING CRITICAL MASS

This section presents important topics that have specific applications for evaluating criticality safety of tank waste.

7.1 RADIAL NUCLIDIC PROFILE FOR ALUMINUM-CLAD FUEL ELEMENTS

Deccladding waste is assumed to consist of fuel cladding and the outer 5% of the active fuel volume. During fuel exposure, plutonium production is enhanced in the outer surface layer of the fuel element. This layer was potentially removed with the cladding waste. This possibility is examined to see if it raises any criticality safety concerns.

Schwinkendorf (Braun et. al. 1994) describes the radial distributions of the various uranium and transuranic nuclides in a typical Hanford Site single-pass reactor fuel element. The predicted radial variation in uranium and transuranic nuclides was based on the distribution of nuclides in "green" (unirradiated) fuel, depletion during irradiation, and the radial distribution of the nuclides generated during irradiation.

The minimum critical mass was calculated for a highly optimized configuration of material taken from the outer surface of fuel elements using the following conservative assumptions.

- Fission products have been removed from the heavy metal components.
- Uranium and plutonium (assumed to stay together) form chunks of pure metal of optimum size and are to be spaced at an optimum distance from each other.
- The interspersed water is pure and does not contain any neutron poisons.
- The regular lattice configuration formed into a sphere.
- The nuclides assumed in the calculations were characteristic of higher exposure than would be expected in the cladding waste.

Even for these ideal conditions, the mass required for criticality was in excess of 10 metric tons (10^4 kg) of heavy metal. This conservative minimum critical mass is considerably less than would be required for criticality with the actual waste, but is more than actually present in a waste tank. If the neutron absorption properties of the fission products were taken into account, this alone would greatly increase the critical mass (see Section 7.3).

7.2 PLUTONIUM INTERMETALLICS

Plutonium is capable of forming "intermetallic" alloys with other metals. An intermetallic alloy is an alloy of two metals having a progressive change in composition.

The formation of intermetallic particles that contain plutonium has been identified as a potential mechanism to create particles capable of being mechanically concentrated. Although this change can manifest itself by giving the compound different chemical properties, it is the physical properties that are of interest to criticality safety. Such particles might be denser and physically distinct from other waste and might therefore separate during differential settling, mixing, and shaking. These denser particles might accumulate in one place, resulting in a higher plutonium concentration than in the waste as a whole.

Intermetallics in spent nuclear fuel can be divided into two types. The first type is uranium or plutonium combined with aluminum, silicon, or zirconium at the interface between the fuel and the cladding. These intermetallics would be expected to be dissolved during fuel reprocessing, and would not enter the waste as small, high-density metal particles. The associated fissile materials would behave the same as the rest of the waste and would be of no special criticality concern.

The second type of intermetallic consists of particles of noble metals, ruthenium, rhodium, and palladium, combined with some plutonium or uranium. The lack of mobility of the noble metals at the operating temperatures of the Hanford Site reactors makes it unlikely that any such particles were formed. There is no evidence to indicate that this type of intermetallic particle was actually formed. Nevertheless, calculations were performed to estimate the impact of intermetallic particles on criticality safety, assuming such particles were present in the waste.

Schwinkendorf (Braun et al. 1994) performed a calculation of the relative abundance of plutonium atoms to atoms of noble metals. An estimate of the minimum subcritical mass ratio was calculated for each of the noble metal nuclides. The actual-to-minimum subcritical fraction was then calculated for each noble nuclide by dividing the nuclide-to-plutonium mass ratio by the minimum subcritical mass ratio.

To ensure subcriticality for an infinitely large mass of material, the actual-to-minimum subcritical fraction must be greater than 1.0. Because separation of noble metals from the intermetallic compound structure is extremely difficult, the important comparison fraction is the sum of the actual-to-minimum subcritical fractions obtained for the heavy metal nuclides taken as a whole. In this way a comparison fraction of 11 was obtained. Because the distribution of the various nuclides within the intermetallic particles would be homogeneous (or very nearly so), a comparison fraction of 11 is more than adequate to ensure that the particles are subcritical no matter how large the quantity of intermetallics.

7.3 FISSION PRODUCT POISONING

Fission product poisoning is usually ignored in criticality safety evaluations of tank waste. A great difficulty in evaluating the effect of this poisoning on k_0 is the inability to measure the concentration and distribution of specific radionuclides. Nevertheless, between 70 and 100 metric tons of fission products were produced on the Hanford Site, and a

large fraction of this material resides in tank storage. For every 1 kg of plutonium sent to tank farms, approximately 100 kg of fission products went along in the process.

Calculations were made of the effect of the buildup and decay of 36 specific fission products selected for their significance from a criticality viewpoint. When these 36 fission products are added into a ^{239}Pu -water solution, k_{∞} is suppressed dramatically. Across most of the range of plutonium concentration examined, a fission products/ ^{239}Pu mass ratio of 10 is predicted to reduce k_{∞} by a considerable degree (Braun et al. 1994).

The increase in the critical mass caused by fission products is substantial. The presence of fission products in waste and their intimate association with plutonium significantly increase the margin of subcriticality. It is clear that an evaluation of criticality safety that ignores the presence of fission products is conservative. However, the great difficulty in ensuring their presence with the plutonium makes it necessary to ignore their contribution toward making the waste even more subcritical.

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APPENDIX A
INDEPENDENT REVIEW

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APPENDIX A

INDEPENDENT REVIEW

Independent Reviewer

 J. Lee Geiger
 Senior Engineer
Date 26 July 25

J. L. Geiger of Criticality and Shielding performed an independent review of the contents of this report and provided the following comments and evaluation of conclusions. Additional minor comments were discussed with the author and are not included.

A1.0 MONTE CARLO CALCULATIONS

Benchmarks are lacking for k_{∞} measurements of the dry and nearly dry systems which provide optimal moderation for plutonium brought to a subcritical k_{∞} by these absorbers. Consequently, Section 5.1.3 of Chapter 2.0 of the *Nuclear Criticality Safety Manual* (WHC 1995) specifies $k_{\text{eff}} = 0.9$ as the appropriate subcritical limit before allowing for code bias.

Validation studies for the MONK6B¹ package (Macklin and Miller 1992; Miller 1994) supports a bias of 0.015, leading to a subcritical limit on the optimally moderated calculated k_{∞} of 0.885.

A validation study for MCNP² (Briesmeister 1993) has not been published by Westinghouse Hanford Company. Eight validation runs were performed using MCNP to support the calculational results presented in this appendix. The validation runs included unmoderated spheres and diluted spheres. Table A-1 presents the results of these runs, together with a brief description of each system. The average k_{eff} calculated for these systems is 1.00545, and the standard deviation of the eight benchmark calculations is 0.00670. The standard deviation of the average over eight benchmark calculations is 0.00249. The average of the standard deviations is 0.00219, with a standard deviation of 0.00022 and a standard deviation of the average (of the reported standard deviations) of 0.00008. The value of Student's t for seven degrees of freedom (the average of eight statistics) for a one-tailed 95% confidence interval is 1.895 (Perry and Green 1984, p. 1-24).

¹MONK6B and ANSWERS are trademarks of ANSWERS, the marketing service of the United Kingdom Atomic Energy Authority, Winfrith, England.

²A Monte Carlo computer code developed by Los Alamos National Laboratory.

Table A-1. Validation Results for MCNP.^a

Run ID	k _{eff}	Std ^b	Description
fast001	1.00387	0.00199	PU-MET-FAST-001, bare sphere of 4.5 Pu, 1.02 wt% Ga
fast002	1.00135	0.00212	PU-MET-FAST-002, bare sphere of 20.1 Pu, 1.01 wt% Ga
fast008	1.00218	0.00193	PU-MET-FAST-008, 5.1 Pu sphere refl. by Th cylinder
fast010	0.99463	0.00197	PU-MET-FAST-010, 4.8 Pu sphere refl. by nat. U
fast011	1.01375	0.00248	PU-MET-FAST-011, 5.2 Pu sphere refl. by water
pst0601	1.00665	0.00241	PU-SOL-THERM-006 Case 1: 15 in. sln. sphere, 3.12 Pu, 24.8 g/l
pst0602	1.01530	0.00237	PU-SOL-THERM-006 Case 2: 15 in. sln. sphere, 3.12 Pu, 25.56 g/l
pst0603	1.00587	0.00225	PU-SOL-THERM-006 Case 3: 15 in. sln. sphere, 3.12 Pu, 26.97 g/l

^aValidation cases were taken from NEA/NSC/DOC(95)03 (NEA 1995).^bStandard deviation for the calculated k_{eff} reported by the code.

ID = Identification

These statistics support a 95% confidence limit on the reported standard deviation of

$$1.895 \times \sqrt{.00008^2 + 0.00022^2} + 0.00219 \\ = 0.00263 ,$$

a 95% confidence limit on the bias of

$$1.895 \times \sqrt{.00249^2 + 0.0670^2} \\ = .01354 ,$$

and a 95% confidence that the actual k_{eff} is less than the calculated k_{eff} plus

$$\sqrt{.01354^2 + .00263^2} - .00545 \\ = 0.00834$$

95% of the time. A subcritical limit on the optimally moderated k_∞ as calculated by MCNP of 0.885 is therefore adequate to assure subcriticality.

A2.0 CALCULATED MINIMUM CRITICAL RATIOS

Section 4.6 of the main text and its subsections present the concept of the minimum subcritical mass ratio of an absorber to a fissile material. In addition to a citation of previous work, this part of the report extrapolates the data for iron and manganese to other absorbers via the thermal neutron absorption cross section. Although such a single-energy, one-reaction, one-interval hand calculation is valuable for scoping purposes, the potential exists for more complicated phenomena to cause some ratios so obtained to actually be supercritical.

The effective multiplication constant, k_{eff} , is a function of the system geometry and the products of the nuclide concentrations and microscopic cross sections (which have the dimension of area), making the k_{eff} of a system invariant for a given geometry and relative concentrations so long as the product of total concentration and characteristic length is invariant. Thus, for example, doubling the concentration of all nuclides and halving the diameter of a sphere will leave the k_{eff} unchanged. For an infinite system, the characteristic length is infinite and the k_{eff} is therefore a function of relative concentrations only. This fact was used to simplify the calculations by avoiding the need to provide a reasonable total concentration (material density): the ^{239}Pu concentration was arbitrarily set at 0.01 nuclei per barn-cm, and the other concentrations were adjusted to provide the stated ratios.

Table A-2 presents the results of k_{∞} calculations using the MONK6B code for several of the absorbers (ANSWERS 1992). The lack of cross sections for bismuth and lanthanum in the MONK6B package prevented calculations for these absorbers. Table A-3 presents the results of k_{∞} calculations using the MCNP code for absorber systems containing bismuth or neodymium. (One of the lanthanum-bearing streams into the tank waste contained a product called "rare earth nitrate," or REN, which contained primarily lanthanum and neodymium nitrates.) The tables present the standard deviation (Std) calculated by the Monte Carlo codes, which measures the uncertainty for the calculation and not the result. For clarity, these data are repeated in Figures A-1 through A-3.

A2.1 ALUMINUM

Calculations for aluminum were performed by MONK6B using an Al:Pu atomic ratio of 8145.7026. Table A-2 shows that as the H:Pu atomic ratio varies from 0 to 1000, the k_{∞} of 1 mass part plutonium mixed with 910 mass parts aluminum varies from a minimum of 0.73, at H:Pu = 1000, to a maximum of 0.87, at H:Pu = 140. Figure A-1 illustrates the data for aluminum. The Monte Carlo calculations confirm the scoping calculation for aluminum.

Table A-2. MONK6B Calculations of k_{∞} . (5 sheets)

Run ID	k_{∞}	Std*	H:Pu atomic ratio	Absorber species	Absorber to Pu mass ratio
ald	0.7650	0.0008	0	Al	910
alwc	0.7658	0.0007	0.01	Al	910
alwd	0.7655	0.0008	0.1	Al	910
alwu	0.7688	0.0008	1	Al	910
alwt	0.7876	0.0008	10	Al	910
alwh	0.8698	0.0008	100	Al	910
alwk	0.7531	0.0004	1000	Al	910
alwv	0.8227	0.0008	30	Al	910
alww	0.8570	0.0007	300	Al	910
alwx	0.8738	0.0008	180	Al	910
alwy	0.8742	0.0008	140	Al	910
cd	0.7389	<0.0001	0	C	28000
cwu	0.7388	<0.0001	1	C	28000
cwt	0.7380	<0.0001	10	C	28000
cwh	0.7308	<0.0001	100	C	28000
ckw	0.6650	<0.0001	1000	C	28000
cwv	0.7364	<0.0001	30	C	28000
cww	0.7149	<0.0001	300	C	28000
cad	0.8778	0.0008	0	Ca	720
cawc	0.8798	0.0008	0.01	Ca	720
cawd	0.8792	0.0008	0.1	Ca	720
cawu	0.8775	0.0008	1	Ca	720
cawt	0.8691	0.0009	10	Ca	720
cawh	0.9030	0.0009	100	Ca	720
cawk	0.7616	0.0004	1000	Ca	720
cawv	0.8756	0.0009	30	Ca	720
caww	0.8761	0.0009	300	Ca	720
cawx	0.8888	0.0009	55	Ca	720
cawy	0.8983	0.0008	180	Ca	720
cawz	0.9042	0.0009	140	Ca	720
ca2wh	0.8768	0.0009	100	Ca	770

Table A-2. MONK6B Calculations of k_{α} . (5 sheets)

Run ID	k_{α}	Std*	H:Pu atomic ratio	Absorber species	Absorber to Pu mass ratio
ca2wy	0.8747	0.0008	180	Ca	770
ca2wz	0.8765	0.0009	140	Ca	770
ca2wA	0.8609	0.0008	240	Ca	770
ca2wB	0.8725	0.0009	74	Ca	770
ca2wC	0.8678	0.0009	55	Ca	770
crd	0.6346	0.0006	0	Cr	135
crwc	0.6339	0.0006	0.01	Cr	135
crwd	0.6364	0.0006	0.1	Cr	135
crwu	0.6532	0.0007	1	Cr	135
crwt	0.6871	0.0010	10	Cr	135
crwh	0.8210	0.0009	100	Cr	135
crwk	0.7506	0.0004	1000	Cr	135
crwv	0.7137	0.0011	30	Cr	135
crww	0.8524	0.0007	300	Cr	135
crwx	0.8557	0.0008	180	Cr	135
crwy	0.8126	0.0005	550	Cr	135
crwz	0.8574	0.0007	240	Cr	135
cud	0.3904	0.0007	0	Cu	130
cuwc	0.3902	0.0007	0.01	Cu	130
cuid	0.3931	0.0007	0.1	Cu	130
cuwu	0.4230	0.0026	1	Cu	130
cuwt	0.5590	0.0011	10	Cu	130
cuwh	0.7876	0.0010	100	Cu	130
cuwk	0.7493	0.0004	1000	Cu	130
cuwv	0.6453	0.0012	30	Cu	130
cuiww	0.8359	0.0007	300	Cu	130
cuiwx	0.8311	0.0008	180	Cu	130
cuiwy	0.8049	0.0005	550	Cu	130
cuiwz	0.8354	0.0008	240	Cu	130
nd	0.6296	0.0011	0	N	61
nwc	0.6306	0.0011	0.01	N	61

Table A-2. MONK6B Calculations of k_{∞} . (5 sheets)

Run ID	k_{∞}	Std*	H:Pu atomic ratio	Absorber species	Absorber to Pu mass ratio
nwd	0.6266	0.0011	0.1	N	61
nwu	0.6304	0.0011	1	N	61
nwt	0.6459	0.0011	10	N	61
nwh	0.7119	0.0010	100	N	61
nwk	0.6935	0.0004	1000	N	61
nwv	0.6693	0.0010	30	N	61
nww	0.7381	0.0007	300	N	61
nwx	0.7345	0.0009	180	N	61
nwy	0.7246	0.0006	550	N	61
nad	0.7819	0.0007	0	Na	360
nawc	0.7822	0.0007	0.01	Na	360
nawd	0.7825	0.0007	0.1	Na	360
nawu	0.7862	0.0007	1	Na	360
nawt	0.8009	0.0008	10	Na	360
nawh	0.8543	0.0008	100	Na	360
nawk	0.7334	0.0004	1000	Na	360
nawv	0.8239	0.0008	30	Na	360
naww	0.8333	0.0007	300	Na	360
nawx	0.8424	0.0008	55	Na	360
nawy	0.8526	0.0007	180	Na	360
nid	0.7393	0.0007	0	Ni	105
niwc	0.7402	0.0007	0.01	Ni	105
niwd	0.7392	0.0007	0.1	Ni	105
niwu	0.7392	0.0007	1	Ni	105
niwt	0.7213	0.0010	10	Ni	105
niwh	0.8119	0.0009	100	Ni	105
niwk	0.7431	0.0004	1000	Ni	105
niwv	0.7332	0.0011	30	Ni	105
niww	0.8328	0.0007	300	Ni	105
niwx	0.8368	0.0008	180	Ni	105
niwy	0.7988	0.0005	550	Ni	105

Table A-2. MONK6B Calculations of k_{∞} . (5 sheets)

Run ID	k_{∞}	Std*	H:Pu atomic ratio	Absorber species	Absorber to Pu mass ratio
niwz	0.8309	0.0009	140	Ni	105
niwA	0.8359	0.0008	240	Ni	105
niwB	0.8182	0.0006	410	Ni	105
niwC	0.7733	0.0005	740	Ni	105
nitd	0.8236	0.0007	0	NO ₃	270
nitwc	0.8213	0.0007	0.01	NO ₃	270
nitwd	0.8222	0.0007	0.1	NO ₃	270
nitwu	0.8231	0.0007	1	NO ₃	270
nitwt	0.8216	0.0007	10	NO ₃	270
nitwh	0.8080	0.0007	100	NO ₃	270
nitwk	0.6958	0.0004	1000	NO ₃	270
nitwv	0.6376	0.0003	1800	NO ₃	270
nitww	0.5702	0.0002	3000	NO ₃	270
nitwx	0.4703	0.0001	5500	NO ₃	270
nitwy	0.3590	0.0001	10000	NO ₃	270
phosd	0.7848	0.0002	0	PO ₄	3700
phoswc	0.7853	0.0002	0.01	PO ₄	3700
phoswd	0.7848	0.0002	0.1	PO ₄	3700
phoswu	0.7853	0.0002	1	PO ₄	3700
phoswt	0.7848	0.0002	10	PO ₄	3700
phoswh	0.7743	0.0002	100	PO ₄	3700
phoswk	0.6964	0.0002	1000	PO ₄	3700
phoswv	0.7816	0.0002	30	PO ₄	3700
phosww	0.7549	0.0002	300	PO ₄	3700
phoswx	0.6412	0.0002	1800	PO ₄	3700
phoswy	0.5743	0.0002	3000	PO ₄	3700
phoswz	0.4734	0.0001	5500	PO ₄	3700
phoswA	0.3608	0.0001	10000	PO ₄	3700
sad	0.8406	0.0002	0	SiO ₂	3000
sid	0.9378	0.0006	0	Si	1400
siwc	0.9370	0.0006	0.01	Si	1400

Table A-2. MONK6B Calculations of k_{∞} . (5 sheets)

Run ID	k_{∞}	Std*	H:Pu atomic ratio	Absorber species	Absorber to Pu mass ratio
siwd	0.9381	0.0006	0.1	Si	1400
siwu	0.9377	0.0006	1	Si	1400
siwt	0.9374	0.0006	10	Si	1400
siwh	0.9159	0.0007	100	Si	1400
siwk	0.7440	0.0004	1000	Si	1400
siwv	0.6694	0.0003	1800	Si	1400
siww	0.5914	0.0002	3000	Si	1400
siwx	0.4820	0.0001	5500	Si	1400
siwy	0.3646	0.0001	10000	Si	1400
zrd	0.1356	0.0009	0	Zr	4000
zrwc	0.1349	0.0009	0.01	Zr	4000
zrwd	0.1354	0.0009	0.1	Zr	4000
zrwu	0.1400	0.0009	1	Zr	4000
zrwt	0.1703	0.0010	10	Zr	4000
zrwh	0.3832	0.0010	100	Zr	4000
zrwk	0.6271	0.0005	1000	Zr	4000
zrwv	0.5532	0.0008	300	Zr	4000
zrwl	0.6182	0.0004	1400	Zr	4000
zrwm	0.6039	0.0003	1800	Zr	4000
zrwn	0.5781	0.0003	2400	Zr	4000
zrwp	0.5536	0.0002	3000	Zr	4000
zrwx	0.6108	0.0006	550	Zr	4000
zrwy	0.6244	0.0006	740	Zr	4000

*Standard deviation reported by the code for the calculation.

ID = Identification

Table A-3. MCNP Calculations of k_{∞} .

Run ID	k_{∞}	Std*	H:Pu atomic ratio	Absorber species	Absorber to Pu mass ratio
bid	0.21617	0.00078	0	Bi	48000
biwu	0.21679	0.00072	1	Bi	48000
biwt	0.22604	0.00074	10	Bi	48000
biwh	0.30887	0.00090	100	Bi	48000
biwk	0.54548	0.00067	1000	Bi	48000
biws	0.53560	0.00049	3000	Bi	48000
biwv	0.42189	0.00066	300	Bi	48000
biww	0.24653	0.00082	30	Bi	48000
biwx	0.49140	0.00077	540	Bi	48000
biwy	0.52312	0.00072	740	Bi	48000
biwz	0.56202	0.00063	1800	Bi	48000
biwA	0.55976	0.00059	1400	Bi	48000
biwB	0.55152	0.00055	2400	Bi	48000
bipd	0.78508	0.00060	0	BiPO ₄	10100
bipwu	0.78644	0.00071	1	BiPO ₄	10100
bipwt	0.78579	0.00069	10	BiPO ₄	10100
bipwh	0.77593	0.00068	100	BiPO ₄	10100
bipwk	0.70319	0.00060	1000	BiPO ₄	10100
nddl13	0.88395	0.00097	0	Nd	68.2
ndwl13m	0.88394	0.00082	0.001	Nd	68.2
ndwl13c	0.88177	0.00088	0.01	Nd	68.2
ndwl13d	0.86226	0.00084	0.1	Nd	68.2
ndwl13u	0.72558	0.00073	1	Nd	68.2
ndwl13t	0.52135	0.00086	10	Nd	68.2
ndwl13h	0.49543	0.00105	100	Nd	68.2
ndwl13k	0.44379	0.00099	1000	Nd	68.2

*Standard deviation reported by the code for the calculation.

ID = Identification

Figure A-1. Calculated k_{∞} Values for Several Systems Containing Metal Absorbers. 7G960628.15
SRP-CSD

Figure A-2. Calculated k_{∞} Values for Systems with Various Absorbers Found in Tank Waste.

7G96060238.14
SAP-CSD

Figure A-3. Zirconium and Other Absorbers.

7G96060238.13
SRP-CSD

A2.2 CHROMIUM

Calculations for chromium were performed by MONK6B using a Cr:Pu atomic ratio of 620.6635. Table A-2 shows that as the H:Pu atomic ratio varies from 0 to 1000, the k_{∞} of 1 mass part plutonium mixed with 135 mass parts chromium varies from a minimum of 0.63, with no water present, to a maximum of 0.86, at H:Pu = 240. Figure A-1 illustrates the data for chromium. The Monte Carlo calculations confirm the scoping calculation for chromium.

A2.3 COPPER

Calculations for copper were performed by MONK6B using a Cu:Pu atomic ratio of 489.0435. Table A-2 shows that as the H:Pu atomic ratio varies from 0 to 1000, the k_{∞} of 1 mass part plutonium mixed with 130 mass parts copper varies from a minimum of 0.39, with no water present, to a maximum of 0.84, at H:Pu = 300. Figure A-1 illustrates the data for copper. The Monte Carlo calculations confirm the scoping calculation for copper.

A2.4 SODIUM

Calculations for sodium were performed by MONK6B using a Na:Pu atomic ratio of 3743.3436. Table A-2 shows that as the H:Pu atomic ratio varies from 0 to 1000, the k_{∞} of 1 mass part plutonium mixed with 360 mass parts sodium varies from a minimum of 0.73, at H:Pu = 1000, to a maximum of 0.85, at H:Pu = 100. Figure A-1 illustrates the data for sodium. The Monte Carlo calculations confirm the scoping calculation for sodium.

A2.5 NICKEL

Calculations for nickel were performed by MONK6B using a Ni:Pu atomic ratio of 427.53296. Table A-2 shows that as the H:Pu atomic ratio varies from 0 to 1000, the k_{∞} of 1 mass part plutonium mixed with 105 mass parts nickel varies from a minimum of 0.72, at H:Pu = 10, to a maximum of 0.84, at H:Pu = 180. Figure A-1 illustrates the data for nickel. The Monte Carlo calculations confirm the scoping calculation for nickel.

A2.6 CARBON

Calculations for carbon were performed by MONK6B using a C:Pu atomic ratio of 557270.353. Table A-2 shows that as the H:Pu atomic ratio increases from 0 to 1000, the k_{∞} of 1 mass part plutonium mixed with 28000 mass parts carbon decreases from a maximum of 0.74, for a completely dry system, to a minimum of 0.67, at H:Pu = 1000. Figure A-2 illustrates the data for carbon. The Monte Carlo calculations confirm the scoping calculation for carbon.

A2.7 NEODYMIUM

Although lanthanum cross sections were not available in either the MONK6B or the MCNP data sets, neodymium cross sections were available in the MCNP

data set. Because one of the lanthanum-bearing streams into the tank waste contained a product called "rare earth nitrate," or REN, which contained primarily lanthanum and neodymium nitrates, calculations for neodymium were performed by MCNP.

Not all the naturally occurring isotopes of neodymium were available in the cross-section libraries used by MCNP. Consequently, the available libraries were used, and the cross sections of the remaining isotopes were assumed to be zero. (That is, the available isotopes were used in the proportions that would be found if the Nd:Pu atomic ratio were 113, and the remaining isotopes were ignored.) Although the resulting calculations are not accurate, they are conservative.

Table A-3 shows that as the H:Pu ratio increases from 0 to 1000, the k_{∞} of 1 mass part plutonium mixed with 68 mass parts neodymium remains less than a maximum of 0.884, for a completely dry system, to less than 0.444, at H:Pu = 1000. Figure A-2 illustrates the data for neodymium.

A2.8 NITROGEN

Calculations for nitrogen were performed by MONK6B using a N:Pu atomic ratio of 1041.0855. Table A-2 shows that as the H:Pu atomic ratio varies from 0 to 1000, the k_{∞} of 1 mass part plutonium mixed with 61 mass parts nitrogen varies from a minimum of 0.63, for a completely dry system, to a maximum of 0.74, at H:Pu = 300. Figure A-2 illustrates the data for nitrogen. The Monte Carlo calculations confirm the scoping calculation for nitrogen.

A2.9 BISMUTH PHOSPHATE

Calculations for bismuth phosphate were performed by MCNP using a Bi:Pu atomic ratio of 7978.82642. Table A-3 shows that as the H:Pu atomic ratio increases from 0 to 1000, the k_{∞} of 1 mass part plutonium mixed with 10100 mass parts bismuth phosphate falls from a maximum of 0.79, for a completely dry system, to a minimum of 0.70, at H:Pu = 1000. Figure A-2 illustrates the data for bismuth phosphate. The Monte Carlo calculations confirm the scoping calculation for bismuth phosphate.

A2.10 BISMUTH

Calculations for bismuth were performed by MCNP using a Bi:Pu atomic ratio of 54907.172. Table A-3 shows that as the H:Pu atomic ratio varies from 0 to slightly over 1000, the k_{∞} of 1 mass part plutonium mixed with 48000 mass parts bismuth varies from a minimum of 0.22 for a completely dry system to a maximum of 0.56 at H:Pu = 1800. Figure A-3 illustrates the data for bismuth. The Monte Carlo calculations confirm the scoping calculation for bismuth.

A2.11 PHOSPHATE

Calculations for phosphate were performed by MONK6B using a P:Pu atomic ratio of 9313.25. (This concentration was derived by making two rather

dubious assumptions: that the ratios produced by the scoping calculation for bismuth and bismuth phosphate were accurate for the acceptance limit and that the effects of bismuth and phosphate were strictly additive.) Table A-2 shows that as the H:Pu atomic ratio varies from 0 to 10000, the k_{∞} of 1 mass part plutonium mixed with 3700 mass parts phosphate falls from a maximum of 0.785 for a completely dry system to a minimum of 0.361 at H:Pu = 10000. Figure A-3 illustrates the data for phosphate. The Monte Carlo calculations confirm that a PO_4 :Pu mass ratio of 3700 is sufficient to guarantee subcriticality.

A2.12 NITRATE

Calculations for nitrate were performed by MONK6B using a N:Pu atomic ratio of 1040.9506. Table A-2 shows that as the H:Pu atomic ratio increases from 0 to 10000, the k_{∞} of 1 mass part plutonium mixed with 270 mass parts nitrate decreases from a maximum of 0.82 for a completely dry system to a minimum of 0.36 at H:Pu = 10000. Figure A-3 illustrates the data for nitrate. The Monte Carlo calculations confirm the scoping calculation for nitrate.

A2.13 ZIRCONIUM

Calculations for zirconium were performed by MONK6B using a Zr:Pu atomic ratio of 10482.438. Table A-2 shows that as the H:Pu atomic ratio varies between 0 and 3000, the k_{∞} of 1 mass part plutonium mixed with 4000 mass parts zirconium varies from a minimum of 0.14, at H:Pu = 0.1, to a maximum of 0.63, at H:Pu = 1000. Figure A-3 illustrates the data for zirconium. The Monte Carlo calculations confirm the scoping calculation for zirconium.

A2.14 SILICON

Calculations for silicon were performed by MONK6B using a Si:Pu atomic ratio of 11916.001. Table A-2 shows that as the H:Pu atomic ratio increases from 0 to 10000, the k_{∞} of 1 mass part plutonium mixed with 1400 mass parts silicon falls from a maximum of 0.94, for a completely dry system, to a minimum of 0.36, at H:Pu = 10000. Figure A-3 illustrates the data for silicon. The Monte Carlo calculations do not confirm the scoping calculation for silicon.

Further inquiry shows that the scoping calculation for silicon consisted of ignoring the oxygen in a previously published result for silica (Sanchez et al. 1996). Re-adding the oxygen into the case yielded the completely dry case "sad," with $k_{\infty} = 0.8406$. Thus, the Monte Carlo calculation confirms the previously published minimum subcritical mass ratio of silica to plutonium of 3000.

A2.15 CALCIUM

Calculations for calcium were performed by MONK6B. The initial check on the scoping calculation used a Ca:Pu atomic ratio of 4294.347, in accordance

with the scoping calculation prediction that a calcium to plutonium mass ratio of 720 would be reliably subcritical. Table A-2 shows that, for H:Pu = 100, the k_{∞} = 0.90, which is above the acceptance criterion of 0.885.

A second scoping calculation was performed using the action counts listed in the MONK6B output for run cawz. The new estimate suggested that a ratio of 764 would yield a safe k_{∞} of 0.88. This ratio was rounded up to 770. Additional monk runs, listed in Table A-2, demonstrate that this ratio leads to a reliably subcritical infinite system. (Because the safety of this ratio is demonstrated by the MONK6B calculations, and because the action count output of MONK6B has not been validated, the calculations used in estimating this value are not shown.) Figure A-4 illustrates the results for calcium.

The Monte Carlo calculations demonstrate that a calcium to plutonium mass ratio of at least 770 is reliably subcritical.

A2.16 SUMMARY OF NEW CALCULATIONS

In summary, the Monte Carlo calculations support the analyzed subcritical minimum X:Pu mass ratios for aluminum, bismuth, bismuth phosphate, chromium, copper, nickel, sodium, nitrogen, nitrate, phosphate, and zirconium. Although the Monte Carlo calculations support the silicon dioxide ratio reported by Sanchez et al. 1996, they do not show the proposed silicon ratio to be sufficient. The Monte Carlo calculations furthermore show that a calcium to plutonium mass ratio of 770 is reliably subcritical, and that a neodymium to plutonium mass ratio of 68 is reliably subcritical. Further multigroup calculations will be necessary before the proposed lanthanum limits can be accepted as verified.

A2.17 CHECK ON IRON AND MANGANESE

As a check on the subcritical mass ratios reported by Clemmons and Goslen (1992) for iron and manganese, MONK6B calculations were performed at various H:Pu ratios using the cited concentrations of iron and manganese. Table A-4 presents the results, which are graphed in Figure A-5. The calculations suffice to independently establish the minimum subcritical Mn:Pu mass limit of 32.

Although the new calculations do not independently confirm the minimum subcritical Fe:Pu mass limit of 160, they do not challenge it either. The limits for iron and manganese are not proposed on the basis of new work, but on the basis of the detailed evaluation by Clemmons and Goslen (1992), which documents and establishes the minimum subcritical limits for iron and manganese.

Table A-4. MONK6B Calculations of k_{∞} .

Run ID	k_{∞}	Std*	H:Pu atomic ratio	Absorber species	Absorber to Pu mass ratio
fed	0.8225	0.0007	0	Fe	160
fewd	0.8227	0.0007	0.1	Fe	160
fewu	0.8203	0.0008	1	Fe	160
fewt	0.8016	0.0010	10	Fe	160
fewv	0.8143	0.0010	30	Fe	160
fewh	0.8892	0.0009	100	Fe	160
fewz	0.9007	0.0009	140	Fe	160
fewx	0.9038	0.0008	180	Fe	160
fewA	0.8987	0.0008	240	Fe	160
feww	0.8915	0.0007	300	Fe	160
fewy	0.8466	0.0005	550	Fe	160
fewk	0.7822	0.0004	1000	Fe	160
mnd	0.6760	0.0004	0	Mn	32
mnwd	0.6765	0.0005	0.1	Mn	32
mnwu	0.6631	0.0006	1	Mn	32
mnwt	0.6023	0.0010	10	Mn	32
mnwv	0.6169	0.0011	30	Mn	32
mnwh	0.7531	0.0009	100	Mn	32
mnwx	0.8048	0.0008	180	Mn	32
mnwz	0.8153	0.0007	240	Mn	32
mnww	0.8165	0.0007	300	Mn	32
mnwA	0.8073	0.0006	410	Mn	32
mnwy	0.7910	0.0005	550	Mn	32
mnwk	0.7407	0.0004	1000	Mn	32

*Standard Deviation reported by the code for the calculation.

ID = Identification

Figure A-4. Variation of k_{∞} with H:Pu for Systems of Plutonium and Calcium.

7G96060238.12
SRP-CSD

Figure A-5. Calculated k_{∞} for Various Moderated Plutonium with Iron or Manganese as an Absorber.

76860238.21
SRP-CSD

A3.0 CALCULATIONS DESCRIBED IN APPENDIX C

The technical review of Appendix C included independent derivation of the formulae used to produce compositions with waste solids and independent Monte-Carlo calculations to verify the results of the sample calculation. As detailed in Appendix C, the cross-section library for WIMS-E did not contain phosphorus and the WIMS-E/GOLF calculations substituted silicon for phosphorous. Consequently, two MONK6B calculations were performed for each WIMS-E/GOLF calculation: one substituting silicon for phosphorous and one using the phosphorous cross section included in the MONK6B package. A third calculation was made using MCNP as an independent check on the cross-section set.

Table A-5 presents the results of the MONK6B calculations. Each calculation used the composition specified as input to the WIMS-E/GOLF system, together with the radius found by GOLF for $k_{eff}=0.935$. Because MONK6B was expected to produce a k_{eff} of 0.935 for these cases, Table A-5 lists the difference $0.935-k_{calc}$, together with the calculation standard deviation from MONK6B. As can be seen, the Monte Carlo codes calculated lower values for k_{eff} than the WIMS-E/GOLF system. The student's t test shows the difference to be statistically significant. The reviewer believes the difference to be due to slightly different assumptions about the values of physical constants. Because the WIMS-E/GOLF results are more conservative than the Monte Carlo results, the difference is unimportant.

A comparison of the results for the modified composition used in the WIMS-E/GOLF calculation and the results for the original composition shows negligible effect of replacing phosphorus with silicon. A comparison of the results using MONK6B (which uses cross sections derived from the same database as the WIMS-E cross sections) and the results using MCNP (which uses cross sections from a different source) shows the two cross-section sets to be in agreement.

Table A-5. Differences in k_{eff} Between WIMS-GOLF and Monte Carlo Codes.

Percent solids	MONK6B, modified composition	MONK6B, original composition	MCNP, original composition
38	0.0064 ± 0.0021	0.0044 ± 0.0021	0.00781 ± 0.00163
39	0.0073 ± 0.0022	0.0068 ± 0.0022	0.00386 ± 0.00177
40	0.0053 ± 0.0022	0.0039 ± 0.0021	0.00699 ± 0.00163
41	0.0046 ± 0.0022	0.0067 ± 0.0022	0.00746 ± 0.00153
42	0.0064 ± 0.0021	0.0087 ± 0.0022	0.00849 ± 0.00165
Average	0.00600 ± 0.00108	0.00610 ± 0.00130	0.00692 ± 0.00109
t of the average*	5.56	4.69	6.39

*Student's t for a statistic is the statistic divided by its unbiased standard deviation.

A set of sphere radii calculated by WIMS-E/GOLF for a narrow range of compositions was provided for review. However, graphing these radii (Figure A-6) does not provide a clear assurance that the minimum radius had been found. Consequently, some further independent MCNP calculations were performed. In this set, the minimum radius found by the WIMS-E/GOLF system was used with a broader set of compositions to find the maximum calculated k_{eff} . (The composition with the maximum k_{eff} at a fixed radius would correspond to the composition with the minimum radius for the specified k_{eff} .) Because the flux did not appear sufficiently symmetrical in the first set of calculations, a second set was performed with more neutrons and more symmetry built into the model. Table A-6 presents the results of these calculations. Figure A-7 presents these results with a quadratic best fit. Figure A-7 demonstrates that the minimum-radius composition is indeed close to that found by the WIMS-E/GOLF calculation and that the minimum-radius calculation is reliable.

Table A-6. Summary of k_{eff} for Constant-Radius MCNP Calculations.

Mass percent solids	First calculation	Second calculation	Weighted average
30	0.93085 ± 0.00070	0.93216 ± 0.00050	0.93172 ± 0.00054
35	0.93257 ± 0.00073	0.93326 ± 0.00054	0.93303 ± 0.00047
38	0.93182 ± 0.00074	0.93238 ± 0.00050	0.93219 ± 0.00044
39	0.93181 ± 0.00078	0.93259 ± 0.00053	0.93233 ± 0.00049
40	0.93250 ± 0.00071	0.93214 ± 0.00052	0.93226 ± 0.00043
41	0.93175 ± 0.00071	0.93093 ± 0.00053	0.93120 ± 0.00048
42	0.93280 ± 0.00072	0.93125 ± 0.00049	0.93177 ± 0.00060
45	0.92899 ± 0.00078	0.92996 ± 0.00054	0.92964 ± 0.00052
50	0.92648 ± 0.00081	0.92665 ± 0.00056	0.92653 ± 0.00046

The first calculation used 400,000 neutron histories and 1/8 sphere.
 The second calculation used 800,000 neutron histories and 1/48 sphere.

Figure A-6. Radii from Sample Calculation in Appendix C.

Figure A-7. Search for Most Reactive Composition using MCNP.

A4.0 SAMPLE INPUT FILES

Sample MONK6B and MCNP input files (including all eight MCNP validation files) are included below. Each output includes the input file.

A4.1 SAMPLE MONK6B INPUT

```
* Wet Calcium and Pu239, mass ratio = 770:1  H:Pu = 100
FMODE 0 0 1 4
NUCNames
CONC PU239 0.01 CA 45.925678 HINH2O 1 0 0.5
CM
NEST 1
SPHERE 1 1.0
SUPERHIST 1 1.0
1 100 1000
600 0
END
END
```

A4.2 SAMPLE MCNP INPUT

```
Wet Neodymium and Plutonium.  Nd:Pu atom ratio = 113 H:Pu=1
1      4      0.320229746      -3      imp:n=1
2      0      +3      imp:n=0

3*      so      1000

m4      94239.55c  0.01      60143.50c  0.138770193
        60145.50c  0.093334063  60148.50c  0.063125489
        8016.50c  0.005      1001.50c  0.01
kcode   1000 1 1 101
ksrc    0 0 0
```

A4.3 VALIDATION INPUT "fast001"

MCNP Pu239 JEZEBEL 17.020 kg Pu(4.5) 10.2 w/o Ga.
 c NEA/NSC/DOC(95)03 PU-MET-FAST-001
 1 1 4.09014-2 -1
 2 0 +1

 1 so 6.38493

 mode n
 imp:n 1 0
 kcode 01000 1.0 15 115
 ksrc 0 0 0
 m1 94239.55c 3.7047-2 94240.50c 1.7512-3
 94241.50c 1.1674-4 31000.50c 1.3752-3
 dbcn 7j
 print

A4.4 VALIDATION INPUT "fast002"

K-EFF CALCULATION FOR Pu-240 JEZEBEL
 c NEA/NSC/DOC(95)03, Case PU-MET-FAST-002
 c Cell Cards
 1 1 4.055292e-2 -1 imp:n=1
 2 0 #1 imp:n=0

 c Surface Cards
 1 so 6.6595

 c Data Cards
 kcode 1000 1.0 15 115
 ksrc 0 0 0
 m1 94239.55c 0.029934
 94240.50c 0.0078754
 94241.50c 0.00015672
 31000.50c 0.0013722
 print

A4.5 VALIDATION INPUT "fast008"

PU(5.1) SPHERE, REFLECTED BY CYLINDER OF TH-232

c NEA/NSC/DOC(95)03, PU-MET-FAST-008

```

1 1 0.03945359 -1 imp:n=1
2 2 0.030054 1 -2 -3 4 imp:n=1
3 0 2:3:-4 imp:n=0

```

c Surface Cards

```

1 so 5.31
2 cx 26.67
3 px 26.67
4 px -26.67

```

c Data Cards

```

m1      94239.55c  3.6049-2      94240.50c  1.9562-3
        94241.50c  1.1459-4      31000.50c  1.3338-3
m2      90232.50c  0.030054
kcode 1000 1.0 15 115
ksrc 0 0 0
print

```

A4.6 VALIDATION INPUT "fast010"

NATURAL U REFLECTED PU(4.8) SPHERE

c NEA/NSC/DOC(95)03 PU-MET-FAST-010

```

1 1 0.04070346 -1 imp:n=1
2 2 0.04786702 1 -2 imp:n=1
3 0 2 imp:n=0

```

```

1 so 5.0419
2 so 9.1694

```

```

m1      94239.55c  0.037291      94240.50c  0.0019277
        94241.50c  0.00012196     31000.50c  0.0013628
m2      92238.50c  0.047518      92235.50c  0.00034902
kcode 1000 1.0 15 115
ksrc 0 0 0
print

```

A4.7 VALIDATION INPUT "fast011"

WATER REFLECTED PU(5.2) SPHERE
 c NEA/NSC/DOC(95)03 PU-MET-FAST-011

1 1 0.04971635 -1 imp:n=1
 2 2 0.100149 1 -2 imp:n=1
 3 0 2 imp:n=0

1 so 4.1217
 2 so 29.5217

m1	94239.55c	0.046982	94240.50c	0.0025852
	94241.50c	0.00014915	94242.50c	0.0000099432
m2	1001.50c	0.066766	8016.50c	0.033383

kcode 1000 1.0 15 115
 ksrc 0 0 0
 print

A4.8 VALIDATION INPUT "pst0601"

Case Pu006-1, 3.12 wt% Pu240, 24.80 g Pu/L, Water SS347
 Reflected Sphere, 15 in dia
 c Volume adjusted for solution in support pipe and contactors
 c Copied from NEA/NSC/DOC(95)03/I

1	0	3	imp:n=0	\$Outside everything
2	1	1.0045-01	-1 imp:n=1	\$Inner sphere, Pu(NO3)4

sln.
 3 2 8.6914-02 1 -2 imp:n=1 \$SS347 Spherical
 Container
 4 3 9.9933-02 2 -3 imp:n=1 \$Water reflector

1	so 19.0416	\$Steel sphere inner
radius		
2	so 19.1686	\$Steel sphere outer
radius		
3	so 49.1686	\$Water reflector outer
radius		

mode n				
kcode	1000 1.0 10 110			
ksrc	0 0 0			\$Point source at origin
m1	94239.55c 6.0526-05	94240.50c 1.9411-06		\$Solution
	7014.50c 1.1266-03	1001.50c 6.4266-02		
	8016.50c 3.5054-02	26000.55c 9.4892-07		
m2	26000.55c 6.0386-02	24000.50c 1.6678-02		\$SS347
	28000.50c 9.8504-03			
m3	1001.50c 6.6622-02	8016.50c 3.3311-02		\$Water
mt1	lwtr.01t			
mt3	lwtr.01t			
totnu				
ctme	2100.0			
print				

A4.9 VALIDATION INPUT "pst0602"

Case Pu006-2, 3.12 wt% Pu240, 25.56 g Pu/L, Water SS347
 Reflected Sphere, 15 in dia
 c Volume adjusted for solution in support pipe and contactors
 c Copied from NEA/NSC/DOC(95)03/I PU-SOL-THERM-006

1	0	3	imp:n=0	\$Outside everything
2	1	1.0042-01 -1	imp:n=1	\$Inner sphere, Pu(NO3)4

sln.

3	2	8.6914-02 1 -2	imp:n=1	\$\$\$347 Spherical
---	---	----------------	---------	---------------------

Container

4	3	9.9933-02 2 -3	imp:n=1	\$Water reflector
---	---	----------------	---------	-------------------

1	so	19.0416		\$Steel sphere inner
radius				
2	so	19.1686		\$Steel sphere outer
radius				
3	so	49.1686		\$Water reflector outer
radius				

mode n

kcode	1000 1.0 10 110			
ksrc	0 0 0			\$Point source at origin

m1	94239.55c 6.2381-05	94240.50c 2.0006-06		\$Solution
	7014.50c 1.4277-03	1001.50c 6.3492-02		
	8016.50c 3.5444-02	26000.55c 1.0136-06		
m2	26000.55c 6.0386-02	24000.50c 1.6678-02		\$\$\$347
	28000.50c 9.8504-03			
m3	1001.50c 6.6622-02	8016.50c 3.3311-02		\$Water
mt1	lwtr.01t			
mt3	lwtr.01t			
totnu				
ctme	2100.0			
print				

A4.10 VALIDATION INPUT "pst0603"

Case Pu006-3, 3.12 wt% Pu240, 26.97 g Pu/L, Water SS347
 Reflected Sphere, 15 in dia
 c Volume adjusted for solution in support pipe and contactors
 c Copied from NEA/NSC/DOC(95)03/I PU-SOL-THERM-006

1	0	3	imp:n=0	\$Outside everything
2	1	1.0022-01 -1	imp:n=1	\$Inner sphere, Pu(NO3) ₄ sln.
3	2	8.6914-02 1 -2	imp:n=1	\$SS347 Spherical
Container				
4	3	9.9933-02 2 -3	imp:n=1	\$Water reflector
1	so	19.0416		\$Steel sphere inner radius
2	so	19.1686		\$Steel sphere outer radius
3	so	49.1686		\$Water reflector outer radius
mode n				
kcode	1000	1.0	10	110
ksrc	0	0	0	
m1	94239.55c	6.5822-05	94240.50c	2.1109-06 \$Point source at origin
	7014.50c	2.0590-03	1001.50c	6.1880-02 \$Solution
	8016.50c	3.6223-02	26000.55c	1.0352-06
m2	26000.55c	6.0386-02	24000.50c	1.6678-02 \$SS347
	28000.50c	9.8504-03		
m3	1001.50c	6.6622-02	8016.50c	3.3311-02 \$Water
mt1	lwtr.01t			
mt3	lwtr.01t			
totnu				
ctme	2100.0			
print				

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APPENDIX B

COMPUTER CODES

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APPENDIX B

COMPUTER CODES

Calculations for the figures were made using the GOLF code with cross sections generated by the WIMS-E code. As part of the technical review, calculations were made with the MONK6B¹ Monte Carlo code. A description of these codes is provided. Calculations by GOLF and WIMS-E are described in Appendix C.

B1.0 WIMS-E CODE VALIDATION AND APPLICATION

The WIMS-E code (Gubbins et al. 1982) has been extensively validated against critical experimental data. Documented validation efforts have covered low-enriched uranium metal billets (Erickson 1992, Schwinkendorf 1985), MKIA fuel assemblies and uranium metal rods (Schwinkendorf 1992a), and low-enriched uranium solutions (Schwinkendorf 1992b). In addition, validations exist for WIMS-D, the earlier version of the code.

The WIMS-E code has been shown to be less conservative than the WIMS-D code, although this is believed to be more a function of the newer cross-section library than changes in the numerical methods employed in the code. WIMS-E follows the critical experimental data more accurately than WIMS-D but is still conservative. Because of its conservative nature, it has been the practice to neglect the imposition of a bias when using WIMS-E to calculate k_{∞} , buckling, or cross sections that are input to a diffusion theory code (to calculate idealized, finite dimensions).

The validation of the uranium metal rods included critical experimental data from two sources. The first (Hellens and Honeck 1962) was for 1.0 wt% ²³⁵U-enriched rods and included measured boron poison effects. The published results were in the form of bucklings, not critical masses or k_{eff} . To compare WIMS-E results to the published results, WIMS-E results were output as two-group lattice-averaged cross sections for each experiment, and an analytical formula was used to calculate buckling for each case. WIMS-E results compared very well with experimental results, both as a function of water-to-uranium volume ratio and as a function of amount of poison added. The second source (Kupinski and Toffer 1970) contained data over a range of rod outer diameters (OD) and ²³⁵U enrichments (0.444 cm [0.175 in.] OD to 7.62 cm [3.0 in.] OD and 3.0 wt% ²³⁵U to 4.89 wt% ²³⁵U). These results were in the form of critical masses in spherical and cylindrical geometry.

The newest version of WIMS-E, Version 6a, comes with a new cross-section library (1994) that was developed from the Joint European File (JEF 2.2) and

¹MONK6B is a trademark of ANSWERS, the marketing service of the United Kingdom Atomic Energy Authority, Winfrith, England.

ENDF/B-VI. The burnup chains now include transuranics up to and including ^{244}Cm . Burnup validation of the code was recently performed and excellent agreement was shown (Schwinkendorf et al. 1996, Wittekind 1984).

B2.0 GOLF DIFFUSION THEORY CODE

The GOLF computer code was used to perform diffusion theory calculations using cross sections generated by WIMS-E. Information on this code can be found in WHC-SD-NR-SWD-024, *Software Certification Package for the GOLF Code* (Schwinkendorf 1994). The following is taken from that document.

"The GOLF computer code numerically solves the one-dimensional neutron diffusion equations for either slab, cylinder, or spherical geometries. The code employs a matrix factorization method for solving the tri-diagonal matrix that results from the finite differencing of the diffusion equations. The GOLF code iteratively searches for either k_{eff} or a system dimension that will satisfy input requirements. Perpendicular buckling (leakage) corrections may be supplied by the user for either cylinder or slab geometries. An internal, automatic buckling conversion is provided to transform the spherical geometry option into a hemisphere option. The GOLF sourcecode is written in standard FORTRAN77, and is therefore portable to a variety of computer platforms.

The equations solved are the multi-group neutron diffusion equations. For boundary conditions, either zero flux or zero incoming neutron current at the outer reflector extrapolated boundary may be selected. Flux symmetry is required at the core centerline. GOLF uses an upper/lower triangular matrix factorization method for calculating the fluxes for each source iteration. The fission source is extrapolated using a Chebychev polynomial predictor-corrector."

The GOLF Software Certification Package contains in Appendix B the verification and validation calculations for the GOLF computer code. Because cross sections were generated using the WIMS-E lattice transport code, correct prediction of critical experiments also validates the WIMS-E cross-section library.

Validation calculations were made using data from critical experiments with N Reactor MKIA fuel assemblies. These assemblies were arranged into a hexagonal lattice until criticality was approached (Brown et. al. 1965). Three different lattice spacings were used. To validate the GOLF code, WIMS-E was used to generate two-group cell cross sections for the core region. The W-PRES and W-RES modules were used to evaluate spatial self-shielding effects. After cross sections were generated, they were input to the GOLF code, executed in cylinder geometry. Axial neutron leakage was modeled using a perpendicular buckling appropriate for MKIA assemblies stacked two high.

In comparison to benchmark experiments, GOLF calculations using WIMS-E cross sections have proved to be conservative. No statistical uncertainties arising from Monte Carlo methods are associated with deterministic calculations, such as those by WIMS-E and GOLF. When calculations are made to a k_{eff} of 0.935, as is done for MONK6B, the calculated configuration is

expected to have a true k_{eff} less than 0.95 with a confidence level of at least 95%.

B3.0 MONK6B MONTE CARLO CODE

The MONK6B Monte Carlo code is an updated version of the MONK6A Monte Carlo code (UKAEA 1988a and 1988b), which was developed in the United Kingdom. Documentation for MONK6A applies to MONK6B, and the codes are basically the same. MONK6A has been used to calculate critical parameters for many years, and is derived from the GEM code introduced more than 25 years ago.

MONK6B can calculate critical parameters for complex geometries and material compositions. MONK6B results include the following:

- The effective neutron multiplication factor (k_{eff})
- The average neutron lifetime and generation time
- Energy-dependent fluxes
- Region-dependent fission densities.

Of these, k_{eff} is the most important parameter for assessing criticality safety. (By definition, k_{eff} less than 1.0 means subcriticality.) MONK6A "follows" neutrons through the system being modeled and compiles statistics on interactions. This statistical information is then used to compute k_{eff} for every region of the model.

MONK6B uses a library of cross sections that are specified at a large number of specific energy points. Thus, instead of averaging all the neutrons in a group together as is often done, the code allows calculations at a fine resolution along the energy spectrum. This eliminates the need to select cross-section sets averaged over broadly defined energy bands.

In MONK6B, cross sections for composite materials and mixtures are obtained by adding macroscopic cross sections of the individual elements of the material. Unless otherwise noted, code convergence uncertainties are specified for one-standard deviation.

B3.1 VALIDATION OF MONK6A CALCULATIONS

The MONK6A User's Guide describes 44 validation calculations (UKAEA 1988b) carried out to determine code bias using the MONK6A Point Energy Nuclear Library. These 44 experiments were selected to represent uranium, plutonium, and mixed systems over a wide range of moderation and reflection. These validation calculations provide considerable assurance that MONK6A calculations using point energy nuclear data accurately determine k_{eff} for a wide range of systems. Verification that MONK6B gives the same results for these configurations is provided by Miller and Friar (1995).

Macklin and Miller (1992) report 70 calculations in a report titled *MONK6A PU Validation*. Verification that the same values are provided by MONK6B is provided by Miller (1994). The critical experiments examined cover a wide range of moderation for spheres, cylinders, slabs, and simple arrays.

The calculations were made for critical configurations of plutonium and other materials, including hydrogen, oxygen, nitrogen, and iron, materials used in the conservative waste model. In addition, containers are modeled using stainless steel and aluminum.

B3.2 CALCULATIONAL BIAS DEFINED

Calculational bias is defined as the difference between the calculated value of k_{eff} and an experimentally determined value. Determining code bias requires careful calculations be performed using well-defined critical systems. The bias is associated with the particular code and cross-section sets used and may vary with moderation.

Bias is a factor that the analyst must consider when using a code. To the extent that bias is present, it makes the code results less certain. Further, the bias itself has an uncertainty that must be considered when applying a bias correction to a calculated critical parameter. For example, at the 95% confidence level, k_{eff} is obtained using the following formula:

$$k_{\text{eff}} = k_{\text{calc}} - \text{bias} + (U_b^2 + U_c^2)^{1/2}$$

where:

$$\begin{aligned} k_{\text{calc}} &= k_{\text{eff}} \text{ as given by calculation} \\ U_b &= 95\% \text{ confidence level uncertainty in bias} \\ U_c &= 95\% \text{ confidence level uncertainty in calculation.} \end{aligned}$$

From the above formula it can be seen that the calculated k_{eff} must be limited according to the following formula to ensure a 95% confidence level that the actual neutron multiplication factor is less than 0.95:

$$k_{\text{calc}} \leq 0.95 + \text{bias} - (U_b^2 + U_c^2)^{1/2}$$

B3.3 ACCEPTABLE SUBCRITICAL LIMIT ON k_{eff}

Macklin and Miller (1992) assume that calculations are run with a sufficient number of neutron histories to ensure a one-standard deviation convergence uncertainty of 0.010 or less. (This is the uncertainty shown on the MONK6A printout.) They determine a bias at the 95% confidence level which includes the uncertainty. No separate uncertainty is then required.

Macklin and Miller (1992, p. 21) reach the following conclusion:

"This validation study has determined that the code bias and statistical uncertainty is 0.015 for Pu only systems across the entire range $0 \leq H/\text{Fissile} \leq 2790$. A subcriticality margin of 0.05 should be used in conjunction with this value. Taking both of these into account results in a limiting value for k_{eff} of 0.935 for any

future calculation. The statistical analysis has compensated for standard deviations up to 0.010 associated with future calculations. Special analysis may be required for calculations of future k_{eff} 's with standard deviations >0.010 . No obvious dependencies were found between k_{eff} and the other parameters observed, so these results may be applied to all analyses that fall within the scope of this validation."

From this validation study, it can be seen that k_{calc} , the calculated k_{eff} , is acceptably subcritical if:

$$\begin{aligned} & k_{calc} \leq 0.935 \quad (<0.95 \text{ with } 95\% \text{ confidence}) \\ \text{and} \quad & U_c \leq 0.010 \quad (\text{one-sigma code convergence uncertainty}). \end{aligned}$$

B4.0 REFERENCES

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APPENDIX C

DESCRIPTION OF WIMS-E AND GOLF CALCULATIONS

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APPENDIX C

DESCRIPTION OF WIMS-E AND GOLF CALCULATIONS

C1.0 INTRODUCTION

This appendix documents calculations performed to create Figures 1-1, 5-1, 5-2, 5-3, 5-4, and 5-5 in the main text which show the minimum critical sizes and masses of spheres and slabs. Section C2.0 explains the process used to determine chemical compositions of conservative waste model (CWM) solids and of supernatant liquid, and Section C3.0 describes the computational tools used to calculate elemental compositions, cross sections, k_{∞} values, and minimum critical dimensions.

C2.0 MIXTURE DENSITIES AND COMPOSITIONS

C2.1 DENSITY AND COMPOSITIONS FOR CWM SOLIDS

The WIMS-E computer code, Version 6a (Gubbins et al. 1982) was used to generate cross sections. Because the 1986 cross-section library does not contain phosphorus, a modified waste composition (shown in Table C-1) was used in which the 6.9 wt% for phosphorus was added to silicon, giving a total silicon content of 10.7 wt%. The resulting macroscopic absorption cross section for CWM solids was then 0.010996 cm^{-1} , only 0.3% higher than the value for the original waste composition.

The elemental composition of mixtures of CWM solids, plutonium, and water was determined. First, the mixture density, which depends only on plutonium concentration and the weight percent of CWM solids, was calculated. The

Table C-1. Composition of Conservative Waste Model Solids.

Component	CWM Solids (wt%)	Modified CWM Solids (wt%)
Oxygen	40.7	40.7
Phosphorus	6.9	0.0
Silicon	3.8	10.7
Sodium	21.5	21.5
Aluminum	7.2	7.2
Nitrogen	0.0	0.0
Iron	19.9	19.9

CWM = Conservative waste model

remaining weight percent was water. Finally, the individual component concentrations were calculated using the weight percents given in Table C-1 for the modified CWM solids. The formulae used to calculate mixture densities and elemental compositions for mixtures of CWM solids, plutonium, and water are shown below.

$$\rho_{\text{mixture}} = \frac{1 - C_{\text{Pu}} \left[\frac{1}{\rho_{\text{Pu}}} - \frac{1}{\rho_{\text{water}}} \right]}{\frac{W_{\text{solids in mixture}}}{100 \rho_{\text{solids}}} + \frac{100 - W_{\text{solids in mixture}}}{100 \rho_{\text{water}}}}$$

where:

$$\rho_{\text{mixture}} = \text{Density of mixture in } \frac{\text{g}}{\text{cc}}$$

$$C_{\text{Pu}} = \text{Desired plutonium concentration in } \frac{\text{g}}{\text{cc}}$$

$$\rho_{\text{Pu}} = \text{Density of pure plutonium} \left(19.8 \frac{\text{g}}{\text{cc}} \right)$$

$$\rho_{\text{water}} = \text{Density of water} \left(1.0 \frac{\text{g}}{\text{cc}} \right)$$

$$W_{\text{solids in mixture}} = \text{Desired weight percent of solids in mixture}$$

$$\rho_{\text{solids}} = \text{Density of dry solids} \left(1.2 \frac{\text{g}}{\text{cc}} \right).$$

$$W_{Pu \text{ in mixture}} = 100 \frac{C_{Pu}}{\rho_{\text{mixture}}}$$

$$W_{\text{water in mixture}} = 100 - W_{\text{solids in mixture}} - W_{Pu \text{ in mixture}}$$

$$W_H \text{ in mixture} = W_{\text{water in mixture}} \left(\frac{11.19}{100} \right)$$

$$W_O \text{ in mixture} = W_{\text{water in mixture}} \left(\frac{88.81}{100} \right) + W_{\text{solids in mixture}} \left(\frac{40.7}{100} \right)$$

$$W_{Si} \text{ in mixture} = W_{\text{solids in mixture}} \left(\frac{10.7}{100} \right)$$

$$W_{Na} \text{ in mixture} = W_{\text{solids in mixture}} \left(\frac{21.5}{100} \right)$$

$$W_{Al} \text{ in mixture} = W_{\text{solids in mixture}} \left(\frac{7.2}{100} \right)$$

$$W_{Fe} \text{ in mixture} = W_{\text{solids in mixture}} \left(\frac{19.9}{100} \right)$$

C2.2 DENSITIES AND COMPOSITION FOR SUPERNATE

A saturated sodium nitrate solution is used to simulate the supernatant liquid commonly found in the waste tanks. Component concentrations were determined using formulae provided in Reynolds and Herting (1984). The solution composition selected for this evaluation was the most reactive in a group of nine different solutions examined. Table C-2 summarizes the composition of this liquid which contains 5.42 moles/L of NaNO_3 , 2.0 moles/L of NaNO_2 , 2.0 moles/L of NaOH , and 0.5 moles/L of NaAlO_2 at a temperature of 60 °C.

Table C-2. Composition of Supernatant Liquid.

Constituent compounds	Wt%	Constituent elements	Wt%
NaNO_3	32.4	Hydrogen	5.673
NaNO_2	9.7	Nitrogen	7.302
NaOH	5.6	Oxygen	70.052
NaAlO_2	2.9	Sodium	16.025
H_2O	49.4	Aluminum	0.948

The following formula relates the density of the supernatant liquid to the molarity of its components and to the temperature.

$$\begin{aligned}\rho_{\text{supernate}} = & 1.017 + 0.0587[\text{NaAlO}_2] - 0.01943[\text{OH}] - 0.000505(\text{TEMP}) \\ & + 0.0183([\text{NaAlO}_2] + [\text{NaOH}] + [\text{NaNO}_3] + [\text{NaNO}_2]) \\ & - 0.000883[\text{NaNO}_3][\text{NaNO}_2]\end{aligned}$$

$$\begin{aligned}[\text{NaNO}_3] = & 5.552 - 0.205[\text{NaNO}_2] - 0.499[\text{NaAlO}_2] - 0.362[\text{NaOH}] \\ & + 0.000347(\text{TEMP})^2\end{aligned}$$

where:

$$\rho_{\text{supernate}} = \text{Density of supernate in } \frac{\text{g}}{\text{cc}}$$

$$[\text{NaNO}_3] = \text{Concentration of NaNO}_3 \text{ in } \frac{\text{moles}}{\text{L}}$$

$$[\text{NaNO}_2] = \text{Concentration of NaNO}_2 \text{ in } \frac{\text{moles}}{\text{L}}$$

$$[\text{NaOH}] = \text{Concentration of NaOH in } \frac{\text{moles}}{\text{L}}$$

$$[\text{NaAlO}_2] = \text{Concentration of NaAlO}_2 \text{ in } \frac{\text{moles}}{\text{L}}$$

$$\text{TEMP} = \text{Temperature of saturated solution in } ^\circ\text{C}.$$

Formulae to calculate mixture densities and elemental compositions for mixtures are shown below. The density of the supernate without plutonium was 1.42284 g/cm³, and the density of the dry solutes was 2.42564 g/cm³. The solutes contribute 50.56387 wt% and water the remaining 49.43613 wt% to the supernatant liquid. Component weight percents used below are from Table C-2.

$$\rho_{\text{solution}} = \frac{1}{\frac{50.56387}{100 \rho_{\text{solut es}}} + \frac{49.43613}{100 \rho_{\text{water}}}}$$

$$\rho_{\text{mixture}} = C_{\text{Pu}} + \rho_{\text{solution}} \left(1 - \frac{C_{\text{Pu}}}{\rho_{\text{Pu}}}\right)$$

where:

$$\rho_{\text{solution}} = \text{Density of supernate solution in } \frac{\text{g}}{\text{cc}}$$

$$C_{\text{Pu}} = \text{Desired plutonium concentration in } \frac{\text{g}}{\text{cc}}$$

$$\rho_{\text{Pu}} = \text{Density of pure plutonium } (19.8 \frac{\text{g}}{\text{cc}})$$

$$\rho_{\text{water}} = \text{Density of pure water } (1.0 \frac{\text{g}}{\text{cc}})$$

$$\rho_{\text{solut es}} = \text{Density of dry solutes } (2.42564 \frac{\text{g}}{\text{cc}}).$$

$$W_{\text{Pu in mixture}} = 100 \frac{C_{\text{Pu}}}{\rho_{\text{mixture}}}$$

$$W_{\text{solution in mixture}} = 100 - W_{\text{Pu in mixture}}$$

$$W_{\text{O in mixture}} = W_{\text{solution in mixture}} \left(\frac{70.052}{100} \right)$$

$$W_{\text{H in mixture}} = W_{\text{solution in mixture}} \left(\frac{5.673}{100} \right)$$

$$W_{\text{N in mixture}} = W_{\text{solution in mixture}} \left(\frac{7.302}{100} \right)$$

$$W_{\text{Na in mixture}} = W_{\text{solution in mixture}} \left(\frac{16.025}{100} \right)$$

$$W_{\text{Al in mixture}} = W_{\text{solution in mixture}} \left(\frac{0.948}{100} \right)$$

C3.0 COMPUTATIONAL PROCEDURE

Critical dimensions were calculated using the GOLF computer code, a one-dimensional, multigroup neutron diffusion theory code (Schwinkendorf, 1994). Cross-sections input to GOLF were generated using the WIMS-E computer code (Gubbins et al. 1982).

Three FORTRAN programs were used to generate WIMS-E input files. WIMWST generated WIMS-E input files for waste mixtures containing CWM solids, plutonium, and water. WIMNIT generated input files for saturated sodium nitrate solution (supernate) and plutonium mixtures. ITERATOR generated pairs of weight percents and plutonium concentrations for input to WIMWST and WIMNIT.

Two Quattro Pro¹ for Windows spreadsheets, MIXWST86 and MIXNIT86, were used to check the correctness of the mixture densities and elemental compositions calculated by the two input file generator codes.

In addition to the above codes, several UNIX script files were used to develop WIMS-E cross-section sets for input into GOLF.

All calculations for this report were for simple geometries, either spheres or infinite slabs with close-fitting, 25-cm thick, water reflectors. The spheres are described by a diameter and a material composition, and the slabs by a thickness and a material composition. The material composition is entered into the GOLF code through the cross sections generated by WIMS-E. The sphere diameter, slab thickness, and reflector thickness are specified in the GOLF input.

C4.0 WIMS-E

MONK6B² and WIMS-E access section libraries of a "common ancestry." The MONK6B point-wise library and the WIMS 69-group library were created from the same British Joint European File (JEF) library. The British use the GENEX code to process "raw" cross section evaluations into libraries for different codes. Because WIMS-E and MONK use cross sections from the same point of origin (GENEX processing of JEF data), the bias in WIMS-E calculations is expected to be about the same as for the same calculations performed using MONK6B.

C4.1 PROCEDURE USED TO GENERATE WIMS-E INPUT

The procedure used to calculate minimum critical dimensions using WIMS-E and GOLF is now described.

1. Choose a waste type from the following list. The WIMS-E input generator code to be used for each type is shown in parentheses.
 - a. CWM solids in water (use WIMWST)
 - b. Pure water (use WIMWST; set CWM solids to zero)
 - c. Supernate (use WIMNIT)

¹Quattro Pro is a trademark of Borland International, Inc.

²MONK6B is a trademark of ANSWERS, the marketing service of the United Kingdom Atomic Energy Authority, Winfrith, England.

2. Choose the range of waste weight percents and plutonium concentrations to be analyzed. Use ITERATOR to generate input file ITER_OUT to be read by either WIMWST or WIMNIT.
3. Copy ITER_OUT file to an archive file called WTXXXXYY, where WT stands for weight percent, XXX is a three-character code specifying the waste type ("WST" for CWM solids, "WAT" for pure water, and "NIT" for supernate), and YY is a three-character code to identify either the plutonium concentration or waste weight percent used.
4. Copy ITER_OUT file to SOLN_IN and run either WIMWST or WIMNIT to generate WIMS-E input file.
5. Open WIMS-E input file SOLN_11 and check mixture densities and elemental weight percents against values generated in either the MIXWST86 or MIXNIT86 spreadsheet.
6. If spot check reveals no errors, copy SOLN_11 file to an archive file whose name is the six-letter mnemonic XXXYYY used in step 3.
7. Use UNIX "script" command to place cross sections generated by WIMS-E into file TYPESCRIPT.
8. Run script GO_6A with file XXXYYY as input. This script passes the XXXYYY file name to WIMS-E, which generates two output files, KINF.XXXYYY (contains k_{∞} values) and OXXXXYYY (contains complete output listing). Script GO_6A issues UNIX "ed" command to pipe in commands from shell script F_CMD_6A. This removes unwanted text from KINF.XXXYYY file, leaving only numerical k_{∞} values.
9. Deactivate UNIX "script" command using "exit" command.
10. Use UNIX command "dos2unix TYPESCRIPT TAPE7 XXXYYY" to replace unwanted ctrl-M characters in TYPESCRIPT file with standard UNIX return characters and to store modified text in TAPE7_XXXXYY file.
11. Edit TAPE7_XXXXYY file to remove unwanted lines, leaving only cross sections generated by WIMS-E. Archive this file.

C4.2 EXAMPLE

An example is provided to find critical dimensions for CWM solids with weight percents ranging from 38% to 42% in water. The goal is to identify the minimum critical sphere size for a plutonium concentration of 7.5 g Pu/L.

First use ITERATOR to generate weight percent/plutonium concentration data pairs. The ITERATOR output file ITER_OUT (Listing 1) contains this information.

Listing 1. Sample ITER OUT File

```

0.0
5
38.00000 7.50000
39.00000 7.50000
40.00000 7.50000
41.00000 7.50000
42.00000 7.50000

```

Copy ITER OUT to an archive file WTST7P5. Then copy ITER OUT to file SOLN_IN and execute WIMWST. This produces output file SOLN_11 which is copied to archive file WST7P5 (Listing 2). WST7P5 can now be used as input to the WIMS-E code.

Listing 2. Sample WIMS-E Input File (WIMWST Output File WST7P5)

```

*****
* pu solutions: chuck rogers model waste, plus water
*****
ncycle 1 5
whead 1
rmesh 7
nregion 1
rmaterial 1
endp
print 0
cent 7
mesh
qual 4 1
* model waste wt% = 38.00000 Pu g/L = 7.50
material 1 1.07522 296.0 1 6239 0.69753 1240 0.00000
6016 69.90907 3001 6.85940 29 4.06600
23 8.17000 27 2.73600 2056 7.56200
qual 4 2
* model waste wt% = 39.00000 Pu g/L = 7.50
material 1 1.07713 296.0 1 6239 0.69629 1240 0.00000
6016 69.42906 3001 6.74765 29 4.17300
23 8.38500 27 2.80800 2056 7.76100
qual 4 3
* model waste wt% = 40.00000 Pu g/L = 7.50
material 1 1.07906 296.0 1 6239 0.69505 1240 0.00000
6016 68.94906 3001 6.63589 29 4.28000
23 8.60000 27 2.88000 2056 7.96000
qual 4 4
* model waste wt% = 41.00000 Pu g/L = 7.50
material 1 1.08099 296.0 1 6239 0.69381 1240 0.00000
6016 68.46906 3001 6.52414 29 4.38700
23 8.81500 27 2.95200 2056 8.15900
qual 4 5
* model waste wt% = 42.00000 Pu g/L = 7.50
material 1 1.08293 296.0 1 6239 0.69257 1240 0.00000
6016 67.98905 3001 6.41238 29 4.49400
23 9.03000 27 3.02400 2056 8.35800
annulus 1 4.0000 1 * infinite region of solution
begin
*
* perseus calculations
*
wthes 1
print 0
begin
*
wpip 1
print 0
niter 1000
toler 1.0e-5
omega 1.25
begin
*
* smear all meshes into one material
*
wsmeas 1 2

```

```

mate 1
mesh 1
endp
newmat 1 1 2 3 4 5 6 7
mcode 1
begin
*
* collapse to 2 groups
*
wcond 2 3
group 2
endp
part 27 69
begin
*
* print 2-group cross sections save it on unit 7
*
winter 3
print
file 3
matrix
snap
begin
finish
stop

```

The lines beginning with the keyword "material" specify the composition of the material whose cross sections are being evaluated. As an example, consider the first material line in Listing 2:

```
material 1 1.07522 296.0 1 6239 0.69753 1240 0.00000
```

The "1" specifies the material number (only one homogeneous material was used), "1.07522" is the mixture density in g/cm³, "296.0" is the material temperature in K, and "1" indicates that this material contains fuel. Following this, eight pairs of values specify the isotopic components of the material and their corresponding weight percents. The isotope identifiers are as follows:

```

6239 for 239Pu,
1240 for 240Pu,
6016 for 16O,
3001 for 1H,
29 for 28Si,
23 for 23Na,
27 for 27Al,
2056 for 56Fe, and
14 for 14N.

```

Parameters in the listing have the following meanings:

nitre 1000	Uses 1000 iterations (maximum) to reach convergence.
toler 1.0e-5	Uses convergence criterion of 1.0×10^{-5} .
omega 1.25	Uses an over-relaxation parameter of 1.25. This is the value recommended for use with water-moderated systems.
part 27 69	Specifies how the original 69 groups are to be partitioned into the final two groups. The cross sections for groups 1 through 27 are collapsed to form the final group 1 cross sections, and the cross sections for groups 28 through 69 are collapsed to form the final group 2 cross sections. The corresponding thermal cutoff value is 4.0 eV.

Next, spot-check mixture densities and compositions against values generated by the MIXWST86 spreadsheet.

Next, execute WIMS-E using WST7P5 as an input file. k_0 values are output to the file KINF.WST7P5 (Listing 3). As WIMS-E runs, it outputs cross-section sets for each of the five cases to the monitor, and these outputs are captured in the TYPESCRIPT file. After being edited and copied to file TAPE7 WST7P5 for archiving (Listing 4), the cross sections are copied to TAPE7, then converted to a GOLF-compatible format by running SNAP_G.X, which creates the file GOLFLIB.

Listing 3. Sample k_0 File KINF.WST7P5

```
1.105129
1.108204
1.111314
1.114457
1.117609
```

Listing 4. Sample Cross-Section File TAPE7 WST7P5

```
i      1 d
1.7228E+00 2.6200E-01
3.5492E-02 3.1831E-02
5.4531E-04 3.5296E-02
.0000E+00 3.4875E-02
3.3229E-06 .0000E+00
w      1
1.8832E-04 1.2296E-02
sp
1.0000E+00 .0000E+00
i      1 d
1.7357E+00 2.6579E-01
3.5011E-02 3.1722E-02
5.4553E-04 3.5274E-02
.0000E+00 3.4394E-02
3.3424E-06 .0000E+00
w      1
1.8840E-04 1.2288E-02
sp
1.0000E+00 .0000E+00
i      1 d
1.7489E+00 2.6970E-01
3.4529E-02 3.1610E-02
5.4577E-04 3.5252E-02
.0000E+00 3.3913E-02
3.3627E-06 .0000E+00
w      1
1.8848E-04 1.2281E-02
sp
1.0000E+00 .0000E+00
i      1 d
1.7623E+00 2.7374E-01
3.4045E-02 3.1497E-02
5.4602E-04 3.5229E-02
.0000E+00 3.3430E-02
3.3837E-06 .0000E+00
w      1
1.8857E-04 1.2273E-02
sp
1.0000E+00 .0000E+00
i      1 d
1.7760E+00 2.7792E-01
3.3559E-02 3.1385E-02
5.4626E-04 3.5205E-02
.0000E+00 3.2944E-02
3.4054E-06 .0000E+00
w      1
1.8866E-04 1.2264E-02
sp
```

1.0000E+00 .0000E+00

C5.0 GOLF

After WIMS-E generates file TAPE7 XXXYYY, copy TAPE7 XXXYYY file to TAPE7, and then run SNAP_G.X to create the GOLFLIB cross-section file to be used by GOLF.

Copy either TAPE3OCR file (for critical radii) or TAPE3OCT file (for critical half-thicknesses) to TAPE30, the command file used by GOLF. Listings 5 and 6 provide sample GOLF input files for sphere and slabs.

Listing 5. TAPE3OCR

GOLF Command File for Critical Spheres.

```
0.935 5.0e-5 1 1 1 0 1 1
2 2 2 1 0.000e-03 1.0e-05 1.0 0.0001 2.0e-03 1.0e-5 1
25.0 25.0
350 150
1.0 0.0
1.0 0
```

Listing 6. TAPE3OCT

GOLF Command File for Critical Slabs.

```
0.935 5.0e-5 1 1 1 0 1 1
2 2 0 1 0.000e-03 1.0e-05 1.0 0.0001 2.0e-03 1.0e-5 1
25.0 25.0
350 150
1.0 0.0
1.0 0
```

To run GOLF, use one of following commands:

```
"golf.x > CRXXXXYY" (for critical radii)
"golf.x > CTXXXXYY" (for critical half-thicknesses).
```

Calculated dimensions are piped into the named archive file.

C5.1 INSTRUCTIONS FOR GOLF

Three input files are used for GOLF. The primary file, TAPE30, consists of 6 or 7 lines of input data. Within each line, values are written in free format with individual values separated by one or more spaces. However, the number of parameters on each line must correspond to the input instructions.

Sample GOLF Input File: TAPE30

```
1.00 5.0e-5 1 1 0 0 1 1
4 2 2 1 0.00 1.0e-05 1.0 0.0001 2.0e-03 1.0e-5 1
25.0 25.0
350 100
```

7.63000E-01 2.36900E-01 1.00000E-04 0.00000E+00
0.0 1

The meaning of the parameters for each line is now described:

Line 1: desk, epsrt, ifopt, isopt, itopt, ihm, irl, ib

- desk k_{eff} for which dimension is calculated.
- epsrt Convergence criterion. If a dimension is required for an input k_{eff} , GOLF will iterate until the dimension converges to within the criterion, epsrt.
- ifopt Integer flag to tell GOLF whether or not to calculate k_{eff} of the core cross sections (0/1 for calculate/do not calculate k_{eff}).
- isopt Integer flag to tell GOLF whether to calculate a k_{eff} for an input set of dimensions (isopt=0) or to perform the dimension search for an input k_{eff} (isopt=1).
- itopt Integer flag. If itopt=0, GOLF will calculate Σ_f values, given the $v\Sigma_f$ data and a hard-wired v -spectrum. If itopt=1, GOLF will use user-supplied Σ_f data.
- ihm Tells GOLF (if spherical geometry is requested) whether or not to perform an internal buckling conversion so that the geometry to be treated is hemispherical.

If ihm=0, spherical geometry is used.
If ihm=1, conversion to hemispherical geometry is performed.
- irl Tells GOLF which reflector cross-section set to use. The library of reflector cross sections is read from TAPE9.

Use irl=1 for light water
irl=2 for heavy water
irl=3 for graphite.

For other reflector materials such as concrete, the applicable cross sections may be appended to those already present in the TAPE9 file.
- ib Tells GOLF whether to calculate its own group-dependent extrapolation distances (based on the cross sections of the outermost material region - ib=1), or to read in user-supplied values (ib=0).

Line 2: ng, nr, gf, rb, buck, eps, alp, epspw, b2, epsbt, iadj

- ng Number of energy groups.
- nr Number of material regions.
- gf Geometry factor. The geometry factor gf is equal to 0 for slab geometry, gf=1 for cylinder, and gf=2 for spherical geometry.

rb Boundary condition option that must currently be set to 1.0.

buck Perpendicular buckling correction (cm^{-2}).

eps Flux convergence criterion.

alp Initial over-relaxation parameter, and must be 1.0.

epspwr Region-wise power convergence criterion.

b2 Initial buckling (cm^{-2}) used in buckling search calculation.

epsbt Convergence criterion for the buckling search.

iadj Tells GOLF which fueled region width is to be adjusted to attain the required input k_{eff} . The regions are numbered starting from "1" at the core centerline, and incremented radially outward.

Line 3: w(i), i = 1,...,nr

w(i) Width, or thickness, of each of the nr material regions.

Line 4: ni(i), i = 1,...,nr

ni(i) The number of mesh intervals to be allocated for each of the nr material regions. The user should input at least 2 or 3 mesh intervals per neutron mean-free-path. For an infinite water reflector region (i.e., 25.0 cm), 100 mesh intervals should work.

Line 5: extrap(i), i = 1,...,ng (only if ib=0; delete line if ib=1)

extrap(i) The extrapolation distances to be added onto the width of the outermost material region. If ib=1, this line is deleted, and GOLF will calculate its own set of extrapolation distances based on that region's cross sections.

Line 6: x(i), i = 1,...,ng

x(i) The fission spectrum, one number for each group.

Line 7: sigbor, nf

sigbor An additional absorption cross section that may be added in to each of the nf fueled regions.

All macroscopic nuclear cross sections (except reflector) are read in from the GOLFLIB file.

Blocks of cross sections are read in for each material region, in turn. These blocks may be stacked for repeated cases. This is useful when a series of calculations are required such as to determine the minimum critical mass over a range of lattice spacing. A lattice code, such as WIMS-E, may be used to produce multigroup cross sections for each of several spacings. These

cross sections, once in the correct format (above), may then be combined into one file (GOLFLIB), for input into GOLF.

C5.2 GOLF OUTPUT

GOLF outputs file CRXXXXYY or CTXXXXYY. Edit the output file using UNIX command "ed CRXXXXYY < F2_CMD" to remove unwanted text leaving only numerical values.

Import desired values (k_{∞} values, sphere radii, or slab half-thicknesses) into a Quattro Pro for Windows spreadsheet by:

1. Issuing the TOOLS/IMPORT command
2. Selecting the "Comma and Delimited File" option
3. Highlighting name of desired file.

Search for minimum sphere radius (or slab half-thickness) over plutonium concentrations. After the appropriate values are obtained, the following manipulations can be made:

- Convert to diameter (or thickness)
- Convert to desired units
- Calculate sphere mass (or slab mass)
- Create graphs.

The GOLF output is piped into file CRWST7P5, which, after being edited, appears as shown in Listing 7. This file contains the radii at which each mixture goes critical (using $k_{eff} = 0.935$ as the criterion for criticality). As can be seen from Listing 7, the smallest radius at which criticality occurs is 47.27953 cm, corresponding to a CWM solids weight percent of 40%. After doubling the radius to obtain the diameter and converting the units from centimeters to meters, a value is obtained for the diameter of 0.94559 m at a plutonium concentration of 7.5 g Pu/L.

Listing 7. Sample Critical Radii File CRWST7P5

```
4.728828E+01
4.729015E+01
4.727953E+01
4.728079E+01
4.730901E+01
```


C6.0 SEARCH PARAMETERS USED IN CREATING THE FIGURES

Figures used in this report show graphs of minimum critical parameters (sphere diameter, sphere mass, slab thickness, slab mass, and areal density) as a function of plutonium concentration. Each point plotted represents the minimum value taken from another curve. For instance, the minimum critical sphere diameter is the smallest diameter over all possible water content in waste solids. Therefore, the total number of calculations made was an estimated 10 times greater than the number of points plotted. In all, more than 1,500 calculations were made to produce the 6 figures in this report.

Table C-3 summarizes weight percent/plutonium concentration parameters examined for all three waste media, and describes how the information was used.

Table C-3. Weight Percent/Plutonium Concentration Parameters Used.
(3 sheets)

Filename	Plutonium concentrations (g Pu/L)	Wt% (solids or solutes)	How the output was used
WST100	1 to 20	0 to fully dry in steps of 5	To estimate CWM solids weight percents at which maximum k_w values occur To estimate CWM solids weight percents at which minimum critical dimensions occur To find k_w values and minimum critical dimensions for water
WST999	1 to 10	90.0 to dry in steps of 0.1	To estimate CWM solids weight percents at which maximum k_w values occur
NIT1_8	1 to 8	50.56387 (fully saturated)	To find k_w values and minimum critical dimensions for supernate
NIT820	8 to 10 in steps of 0.1 11 to 20 in steps of 1	50.56387 (fully saturated)	To find k_w values and minimum critical dimensions for supernate
NIT805	8.05	50.56387 (fully saturated)	To find minimum critical dimensions for supernate
WAT6_7	6.0 to 7.0 in steps of 0.1	0 (pure water)	To find minimum critical dimensions and k_w values for pure water

Table C-3. Weight Percent/Plutonium Concentration Parameters Used.
(3 sheets)

Filename	Plutonium concentrations (g Pu/L)	Wt% (solids or solutes)	How the output was used
WAT6P5	6.53 to 6.60 in steps of 0.01	0 (pure water)	To find minimum critical dimensions and k_w values for pure water
WAT668	6.68 to 6.72 in steps of 0.01	0 (pure water)	To find minimum critical dimensions and k_w values for pure water
WAT7P5	7.5	0 (pure water)	To find minimum critical dimensions and k_w values for pure water
WAT8P5	8.05 8.1 to 8.5 in steps of 0.1	0 (pure water)	To find minimum critical dimensions and k_w values for pure water
SLA312	3.0 to 3.9 in steps of 0.1 4 5 6 7 8 9 10 11 12	85 to 99 83 to 97 73 to 87 58 to 72 48 to 62 28 to 42 18 to 32 0 to 14 0 to 14 0 to 14 in steps of 1%	To find minimum critical dimensions for CWM solids
SPH312	3.0 to 3.9 in steps of 0.1 4 5 6 7 8 9 10 11 12	85 to 99 83 to 97 73 to 87 58 to 72 38 to 52 23 to 37 3 to 17 0 to 14 0 to 14 0 to 14 in steps of 1%	To find minimum critical dimensions for CWM solids
WST4P5	4.5	79 to 93 in steps of 1%	To find minimum critical dimensions for CWM solids

Table C-3. Weight Percent/Plutonium Concentration Parameters Used.
(3 sheets)

Filename	Plutonium concentrations (g Pu/L)	Wt% (solids or solutes)	How the output was used
WST6P5	6.53 to 6.60 in steps of 0.01 6.7 to 7.0 in steps of 0.1	46 to 60 46 to 60 in steps of 1%	To find minimum critical dimensions for CWM solids
WST7P5	7.5	32 to 46 in steps of 1%	To find minimum critical dimensions for CWM solids
WST805	8.05	16 to 40 in steps of 1%	To find minimum critical dimensions for CWM solids
WST8P5	8.1 to 8.5 in steps of 0.1	16 to 40 in steps of 1%	To find minimum critical dimensions for CWM solids

CWM = Conservative waste model

C6.1 LISTING OF KEY CALCULATIONS

As documentation of calculations, a listing is provided (Tables C-4, C-5, and C-6) of the input to calculations selected to be representative. The large number of calculations performed make it impractical to list all in this manner.

Table C-4. Summary of Selected Sphere Calculations. (2 sheets)

Figure(s)	1-1 5-1 5-2	1-1 5-1 5-2	1-1 5-1 5-2	1-1 5-1 5-2	1-1 5-1 5-2
Curve type	CWM	CWM	CWM	CWM	CWM
k_{eff}	0.935	0.935	0.935	0.935	0.935
Pu conc. (g/L)	3.0	4.0	5.0	6.7	10.0
Diameter (cm)	923.1	372.5	211.7	115.5	60.23
Pu mass (kg)	1235.5	108.22	24.852	5.4097	1.1441
Reflector	Water	Water	Water	Water	Water
R. thick. (cm)	25.0	25.0	25.0	25.0	25.0
Density (g/L)	1198.6	1183.3	1154.9	1101.9	1009.5
Water (wt%)	1.7497 optimal	8.6620 optimal	21.5671 optimal	47.3919 optimal	99.0094 optimal
Solids (wt%)	98	91	78	52	0
Aluminum (wt%)	7.0560	6.5520	5.6160	3.7440	0.0
Iron (wt%)	19.5020	18.1090	15.5220	10.3480	0.0
Nitrogen (wt%)	0.0	0.0	0.0	0.0	0.0
Hydrogen (wt%)	0.1958	0.9692	2.4132	5.3029	11.0786
Oxygen (wt%)	41.4399	44.7297	50.8998	63.2530	87.9308
Plutonium (wt%)	0.2503	0.3381	0.4329	0.6081	0.9906
Silicon (wt%)	10.4860	9.7370	8.3460	5.5640	0.0
Sodium (wt%)	21.0700	19.5650	16.7700	11.1800	0.0

Table C-4. Summary of Selected Sphere Calculations. (2 sheets)

Figure(s)	5-1 5-2	5-1 5-2	5-1 5-2	5-1 5-2	5-1 5-2
Curve type	Supernate	Supernate	Supernate	Water	Water
k_{eff}	0.935	0.935	0.935	0.935	0.935
Pu conc. (g/L)	8.2	8.5	9.0	6.7	7.0
Diameter (cm)	338.5	207.1	143.8	284.0	167.5
Pu mass (kg)	166.55	39.532	14.015	80.355	17.224
Reflector	Water	Water	Water	Water	Water
R. thick. (cm)	25.0	25.0	25.0	25.0	25.0
Density (g/L)	1.4305	1.4307	1.4312	1006.4	1006.7
Water (wt%)	49.1527	49.1424	49.1253	99.3342	99.3046
Solutes (wt%)	50.2740	50.2635	50.2459	0.0	0.0
Aluminum (wt%)	0.9427	0.9425	0.9422	0.0	0.0
Iron (wt%)	0.0	0.0	0.0	0.0	0.0
Nitrogen (wt%)	7.2602	7.2587	7.2562	0.0	0.0
Hydrogen (wt%)	5.6408	5.6396	5.6376	11.1149	11.1116
Oxygen (wt%)	69.6503	69.6356	69.6113	88.2193	88.1930
Plutonium (wt%)	0.5732	0.5941	0.6288	0.6658	0.6954
Silicon (wt%)	0.0	0.0	0.0	0.0	0.0
Sodium (wt%)	15.9328	15.9294	15.9239	0.0	0.0

CWM = Conservative waste model

Table C-5. Summary of Selected Infinite Slab Calculations.

Figure(s)	5-3 5-4	5-3 5-4	5-3 5-4	5-3 5-4	5-3 5-4
Curve type	CWM	CWM	CWM	CWM	CWM
k_{eff}	0.935	0.935	0.935	0.935	0.935
Pu conc. (g/L)	3.0	4.0	5.0	6.7	10.0
Thickness (cm)	438.0	168.8	92.38	48.44	23.84
Pu mass (kg)	5393.2	2770.4	1895.9	1332.1	978.34
Reflector	Water	Water	Water	Water	Water
R. thick. (cm)	25.0	25.0	25.0	25.0	25.0
Density (g/L)	1198.6	1183.3	1159.3	1110.0	1028.4
Water (wt%)	1.7497 optimal	8.6620 optimal	19.5687 optimal	43.3964 optimal	88.0276 optimal
Solids (wt%)	98	91	80	56	11
Aluminum (wt%)	7.0560	6.5520	5.7600	4.0320	0.7920
Iron (wt%)	19.5020	18.1090	15.9200	11.1440	2.1890
Nitrogen (wt%)	0.0	0.0	0.0	0.0	0.0
Hydrogen (wt%)	0.1958	0.9692	2.1896	4.8558	9.8498
Oxygen (wt%)	41.4399	44.7297	49.9391	61.3326	82.6548
Plutonium (wt%)	0.2503	0.3381	0.4313	0.6036	0.9724
Silicon (wt%)	10.4860	9.7370	8.5600	5.9920	1.1770
Sodium (wt%)	21.0700	19.5650	17.2000	12.0400	2.3560

CWM = Conservative waste model

Table C-6. Summary of Selected Infinite Slab Calculations.

Figure(s)	5-3 5-4	5-3 5-4	5-3 5-4	5-3 5-4	5-3 5-4
Curve type	Supernate	Supernate	Supernate	Water	Water
k_{eff}	0.935	0.935	0.935	0.935	0.935
Pu conc. (g/L)	8.2	8.5	9.0	6.7	7.0
Thickness (cm)	162.1	96.60	65.15	140.6	77.57
Pu mass (kg)	5456.6	3370.0	2406.7	3866.7	2228.7
Reflector	Water	Water	Water	Water	Water
R. thick. (cm)	25.0	25.0	25.0	25.0	25.0
Density (g/L)	1430.5	1430.7	1431.2	1006.4	1006.7
Water (wt%)	49.1527	49.1424	49.1253	99.3342	99.3046
Solutes (wt%)	50.2740	50.2635	50.2459	0.0	0.0
Aluminum (wt%)	0.9427	0.9425	0.9422	0.0	0.0
Iron (wt%)	0.0	0.0	0.0	0.0	0.0
Nitrogen (wt%)	7.2602	7.2587	7.2562	0.0	0.0
Hydrogen (wt%)	5.6408	5.6396	5.6376	11.1149	11.1116
Oxygen (wt%)	69.6503	69.6356	69.6113	88.2193	88.1930
Plutonium (wt%)	0.5732	0.5941	0.6288	0.6658	0.6954
Silicon (wt%)	0.0	0.0	0.0	0.0	0.0
Sodium (wt%)	15.9328	15.9294	15.9239	0.0	0.0

CWM = Conservative waste model

C7.0 COMPARISON WITH EXPERIMENTAL VALUE

Figure C-1 shows the relationship between k_{eff} and plutonium concentration calculated using WIMS-E. From the curve for ^{239}Pu in pure water it can be seen that k_{eff} is equal to 1.00 when the plutonium concentration is 7.0 g/L. The following quote by Paxton and Pruvost (1986) shows that the WIMS-E value compares well with experimental data:

"The limiting critical concentration of 7.6 g ^{239}Pu /L, was deduced at Hanford from specifications of a critical plutonium solution in a 4-ft-diam sphere. This compares with 8.0 g ^{239}Pu /L as originally obtained from a Hanford Physical Constants Test Reactor (PCTR) measurement and was reduced to 7.2 g ^{239}Pu /L in a reexamination of the data."

Figure C-1. k_{∞} vs Plutonium Concentration in Water Solutions.

7G96030035.11
SRP-CSD

Carter et al. (1970) in the *Criticality Handbook* show curves of k_{∞} as a function of plutonium density calculated using the GAMTEC-II computer code. A comparison of Carter's curve for ^{239}Pu in water and the WIMS-E curve is made in Table C-7. The curves are found to be very close to each other.

Table C-7. Comparison of k_{∞} Calculations.

Pu Concentration g/L	k_{∞} Carter (1969)	k_{∞} WIMS-E
10	1.16	1.13
50	1.71	1.72
100	1.78	1.80
1000	1.80	1.84

C8.0 REFERENCES

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Criticality Parameters for Tank Waste Evaluation

Prepared for the U.S. Department of Energy
Assistant Secretary for Environmental Management

Westinghouse
Hanford Company Richland, Washington

Management and Operations Contractor for the
U.S. Department of Energy under Contract DE-AC06-87RL10930

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E. J. Lipke	S7-12	X
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E. M. Miller	H0-35	X
W. C. Miller	R1-56	X
N. J. Milliken	A3-37	X
S. R. Nelson	B4-55	X
L. T. Nirider	T5-53	X
L. L. Pedersen (4)	H0-35	X
J. G. Propson	R2-36	X
A. L. Ramble	A3-38	X
D. P. Reber	T4-07	X
S. H. Rifaey	R1-56	X
L. H. Rodgers	T5-54	X
C. A. Rogers (35)	R1-56	X
W. E. Ross	S5-07	X
R. L. Schlosser	R1-56	X
K. N. Schwinkendorf	H0-32	X
J. P. Stoughter	R2-54	X