Melt-Dilute Spent Nuclear Fuel Form Criticality Summary Report

by

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Executive Summary

Criticality analysis of the proposed Melt-Dilute (MD) form of aluminum-based spent nuclear fuel (SNF), under geologic repository conditions, was performed following the methodology, documented in the *Disposal Criticality Analysis Methodology Topical Report*. This methodology evaluates the potential for nuclear criticality as determined by the composition of the waste and its geometry, namely waste form configuration, including presence of moderator, reflecting structural material, and neutron absorbers. The initial emplaced configuration of the SNF form is a dry package placed in a mined repository passageway. Criticality calculations show that even with waste package failure, followed by degradation of material within the waste package and potential loss of neutron absorber materials, sub-critical conditions can be maintained.

Detailed analyses and findings reported in the Bechtel-SAIC Report, TDR-EDC-NU-000006 Rev 00, "Evaluation of Codisposal Viability for Melt and Dilute DOE-Owned Fuel," July 2001 were used to construct this summary report and should be referred to for additional information as needed. The results show that the proposed melt-dilute form containing gadolinium and/or hafnium as neutron absorbers will maintain subcriticality and that the interim repository subcriticality criterion $k_{eff} + 2\sigma \leq 0.93$ can be met.[a]

The 5-DHLW/DOE SNF waste package for repository disposal is comprised of one 18-in.-outer diameter DOE standardized SNF canister containing the MD ingots, surrounded by five defense high-level radioactive waste (DHLW) glass canisters as shown in Figure ES-1. This intact waste package design is subjected to degradation scenarios comprised of a combination of features, events, and processes (FEPs) that can result in degraded configurations to be evaluated for criticality. The assessment of the criticality potential of the waste package involves (i) degradation scenarios analyses; (ii) geochemistry analysis; and (iii) criticality analysis. These are briefly summarized herein.

ES.1 Degradation Scenarios

Three master degradation scenarios, shown as IP-1, IP-2 and IP-3 in Figure ES-1, were used for evaluation of the physical and chemical interactions (e.g. ground water flow, corrosion, and precipitation) that can occur between the emplaced material and site surroundings and development of criticality analysis models.

The master scenarios (IP-1, IP-2, and IP-3) were based on liquid accumulating in the waste package coupled with scenarios assuming the MD ingots degrading before, concurrently and after, and degradation of other internal components (OICs). The development and selection of these waste package degraded configurations is discussed in Section 2.0.

ES.2 Geochemistry Analysis

A principal objective of the geochemistry analysis was to estimate the chemical composition of the degradation products remaining in a waste package containing MD ingots and high-level waste glass. Two compositions were considered in the analysis. The first composition was 13.2 ± 5 wt% uranium, enriched at less than 20% ^{235}U, and 0.5 wt% gadolinium metal, with the balance being aluminum. The

[a] The criterion of $k_{eff} + 2\sigma \leq 0.93$ has been used in calculations performed in preparation of the subject document. This value has been derived as unity (critical) less the sum of a five percent margin (10 CFR 60.131(h)) and estimates for calculational bias, and the uncertainty of the experiments used to validate the method of calculation. That is, k_{eff} + uncertainty + bias + margin = 1; where uncertainty = 2σ, bias = 0.02, and margin = 0.05. The estimates of bias and bias-uncertainty are taken as the worst-case values calculated from the MCNP simulations of the validation experiments. These estimates will be confirmed at a later time.

second composition is the same as the first except that 2.5 wt% of hafnium is included with the balance being aluminum. Although most of the geochemistry analysis cases showed that more than 80% of the initial Gd content will remain in the waste package under postulated conditions, there were several computational scenarios selected which degraded the MD ingots first and then the DHLW glass while suppressing (a calculational suppression) the formation of compounds such as $GdPO_4 \cdot 10H_2O$. Such calculations show significant loss of Gd, including total depletion of gadolinium while retaining U.

The formation of $GdPO_4 \cdot 10H_2O$ is expected and will be an asset in retaining Gd in sufficient quantity to avoid criticality as shown by these analyses. Experimental verification of the formation of $GdPO_4 \cdot 10H_2O$ in model systems is being pursued in laboratory tests at SRTC.

ES.3 Criticality Analysis

Three-dimensional Monte Carlo criticality calculations were performed for all anticipated intact- and degraded-mode configurations and the results are shown in Table ES-1. Calculations show that the requirement of $k_{eff}+2\sigma$ values less than or equal to the interim critical limit of 0.93 is satisfied for the MD codisposal package if at least 7.5% of the original Gd loading remains mixed with the fissile material. Hafnium, used in the alternate MD ingot composition, is predicted to remain in the DOE SNF canister or waste package in the limited number of conditions analyzed, thus preventing a critical condition even if all Gd is removed from the system.

ES.4 Shielding Calculations

Three-dimensional shielding calculations were performed using a Monte Carlo particle transport code, MCNP, Version 4B2LV to calculate average dose rates at the external surfaces of the waste package. The design criteria specify that a maximum dose rate on all external surfaces not exceed 1,450 rem/hr. A maximum surface dose rate of approximately 200 rem/hr was calculated. This level is well within the prescribed limit. The purpose of this 1,450 rem/hr limit is to limit the personnel exposure at the repository handling facility. In addition, it has been shown that the radiation generated by the MD-SNF form will not lead to irradiation enhanced waste package degradation under repository disposal conditions.

ES.5 Conclusions

A criticality and shielding analysis of the 5-DHLW/DOE waste package has been completed. The results summarized above show that subcritical conditions can be achieved and maintained and that the waste package surface radiation level is well within the specified limit.

This report summarizes criticality and shielding findings pertinent to the 5-DHLW/DOE Melt Dilute short waste package and therefore completes the work requirements contained in Subtask 1.1, Appendix A of SRT-MTS-2000-2035, Rev. 0, 2/1/2001.

Figure ES-1 Criticality Analysis Logic

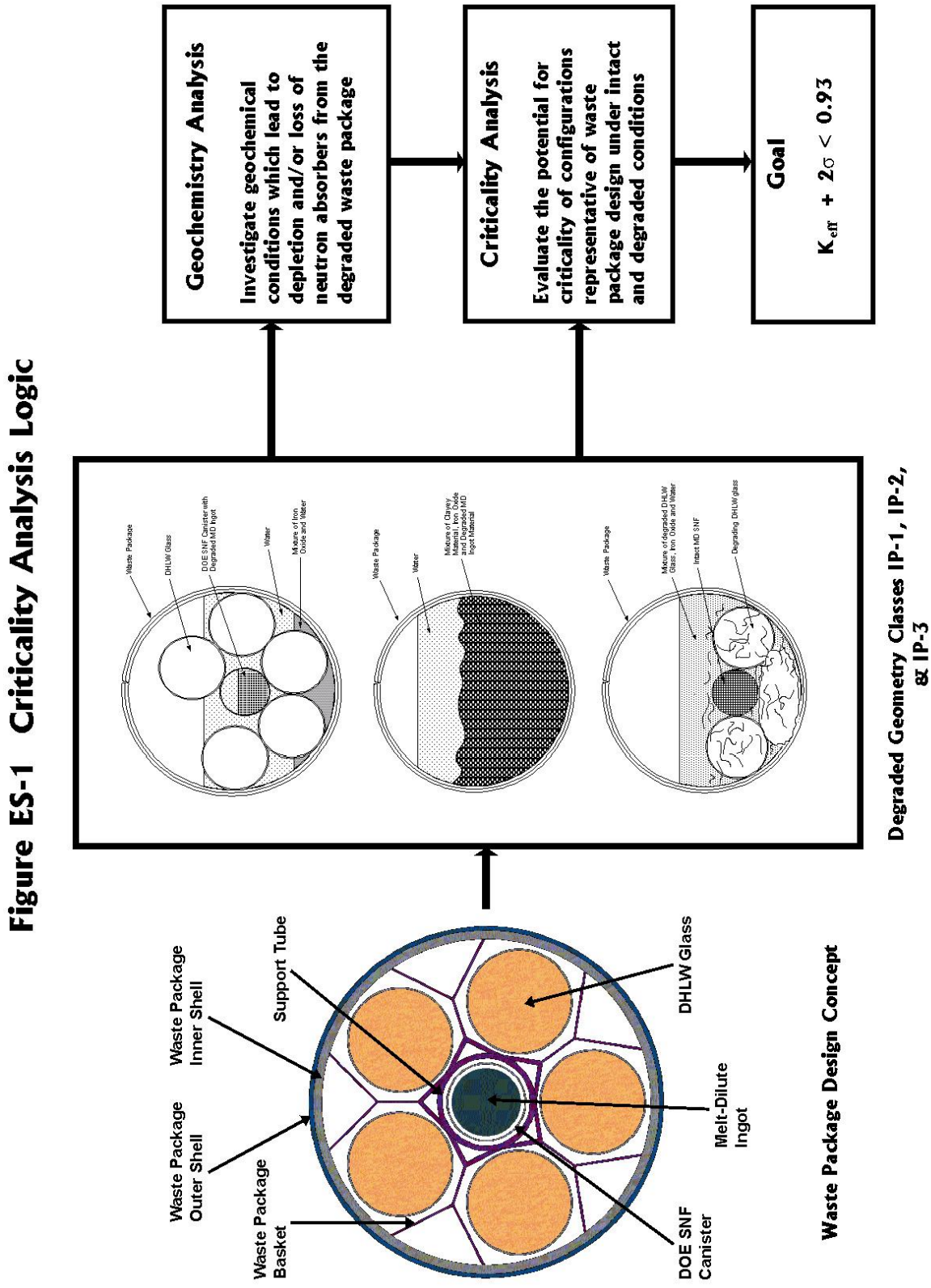

Table ES.1 Criticality Evaluation Summary for Degraded Configurations

Master Scenarios	Description		Configuration Classes	Geochemistry Results	Criticality Results			Comments
					Reference	% of initial Gd	$(k_{eff}+2\sigma)_{max}$	
IP-1	Liquid accumulates in WP	MD ingots degrade before OICs	IP-1-A/B: MD ingots degraded; DOE SNF canister shell intact	For most conditions and variations of parameters the geochemistry results indicated that the minimum content of Gd remaining at any time inside the DOE SNF canister or WP is at least 80% of the initial content.	Section 3.4 Figure 3.2 Tables 3.3 and 3.4	10	0.800	$(k_{eff}+2\sigma)_{max} < 0.93$
			IP-1-C: All WP components degraded		Section 3.5.1 Figures 3.3 and 3.4 Table 3.5	0	0.927	$(k_{eff}+2\sigma)_{max} < 0.93$
IP-2		MD ingots degrade concurrently with OICs	IP-2-A: All WP components degraded		Section 3.5.2 Figures 3.3 and 3.4 Table 3.6	0	1.019	At least 2.5% of initial Gd content must remain in the WP bottom layer mixed with U to prevent criticality.
IP-3		MD ingots degrade after OICs	IP-3-C: All WP components degraded			2.5	0.883	
			IP-3-A: MD ingots and DOE SNF canister shell intact; WP basket degraded		Section 3.6.1 Figure 3.4 Tables 3.7, 3.8 and 3.9	0	0.764	$(k_{eff}+2\sigma)_{max} < 0.93$
					Section 3.6.3[a] Figures 3.4 and 3.5 Table 3.11	1	1.268	At least 7.5% of initial Gd content must remain inside the DOE SNF canister mixed with U to prevent criticality.
						7.5	0.900	
			IP-3-B: MD ingots intact; WP basket, DHLW glass and DOE SNF canister degraded		Section 3.6.2 Figure 3.7 Table 3.10	10	0.816	$(k_{eff}+2\sigma)_{max} < 0.93$
IP-4	WP bottom is penetrated allowing liquid to flow through	MD ingots degrade before OICs	These classes were not discussed (see the explanation in the second cell at right)	The geochemistry calculation does not address this group of scenarios (see explanation in the next cell at right).	All cases are bounded by IP-1, IP-2 and IP-3 as the latter have better moderation (water is pooling inside WP), and more favorable conditions for neutron absorber loss (mainly Gd) concurrent with high fissile material (U) retention.			-
IP-5		MD ingots degrade concurrently with OICs						
IP-6		MD ingots degrade after OICs						

NOTE: [a] MD ingots are degraded.

1.0 INTRODUCTION AND BACKGROUND

There are more than 250 forms of U.S. Department of Energy (DOE)-owned spent nuclear fuels (SNF). Due to the variety of the spent nuclear fuel, the National Spent Nuclear Fuel Program has designated nine representative fuel groups for disposal criticality analyses based on fuel matrix, primary fissile isotope, and enrichment. The Melt and Dilute (MD) fuel form has been designated as the representative fuel for the high-enriched U-Al fuel group.[1] Demonstration that other fuels in this group are bounded by the MD fuel analysis remains to be done before acceptance of these fuel forms. The information for the representative fuel type is provided in the Reference 2. The results of the analyses performed by using the information from this reviewed data report will be used to develop waste acceptance criteria which must be met by all fuel forms within the high-enriched U-Al fuel group. The items that are important to criticality control are identified based on analysis needs and result sensitivities. Prior to acceptance of the fuel from the high-enriched U-Al fuel group for disposal, the important items of the fuel types that are being considered for disposal under the high-enriched U-Al fuel group must be demonstrated to satisfy the conditions set in Section 8.6 of Reference 3, "Items Important to Criticality".

The MD technology development program is focused on the development and implementation of a treatment technology for diluting high-enriched U-Al SNF to low-enriched U levels (<20 wt%) and qualifying this low-enriched U-Al SNF form, MD DOE SNF, for geologic repository disposal. The following assumptions for the MD form were used as design information to use in the criticality analyses.

The MD ingots are homogeneous and monolithic cylinders that will range in height from 15 to 30 in. (381 mm to 762 mm) and will likely be contained in a plain carbon steel crucible liner. The liner will have the maximum outer diameter of 16.5 in. (419.1 mm). The composition is 13.2 ± 5 wt% uranium, enriched at less than 20% ^{235}U and 0.5 wt% gadolinium metal, with the balance of the ingot being aluminum. A second composition is also considered, which is identical to the first for uranium and gadolinium, except that in this case 2.5 wt% of the ingot is hafnium, with the balance of the ingot being aluminum.

The 5-DHLW/DOE SNF waste package is comprised of one 18-in.-outer diameter DOE standardized SNF canister containing the MD ingots, surrounded by five defense high-level radioactive waste (DHLW) glass canisters as shown in Figure 3.1. These are emplaced within the waste package structure consisting of two concentric cylindrical shells. The outer shell is made of a corrosion resistant nickel-based alloy (Alloy 22) and the inner shell is composed of stainless steel 316 NG (nuclear grade). The waste package design has three lids at the one end of the waste package (one for the inner shell and two for the outer shell) and two lids at the other end of the waste package (one for each shell). The DOE SNF canister containing three to six MD ingots is placed in a carbon steel support tube that becomes the center of this waste package.

Criticality analyses have been performed by the DOE-Office of Civilian Radioactive Waste Management (RW) according to the *Disposal Criticality Analysis Methodology Topical Report*.[4] This report had been submitted to the U.S. Nuclear Regulatory Commission as part of the pre-license exchange of information. The methodology includes analyzing the geochemical and physical processes that can breach the waste package and degrade the waste forms as well as the intact and degraded component criticality analyses. Addenda to the topical report will be required to establish the critical limit for the DOE SNF types once sufficient critical benchmarks are identified and run.

Shielding calculations were performed according to Reference 5, Dose Rate Calculation for the Codisposal Waste Package of HLW and the Melt Dilute Al-SNF. The detailed analyses and findings are reported in Reference 3 and were used to construct the summaries that follow.

2.0 CODISPOSAL WASTE PACKAGE DEGRADATION ANALYSIS

2.1 Systematic Investigation of Degradation Scenarios and Configurations

Degradation scenarios comprise a combination of features, events, and processes that result in degraded configurations to be evaluated for criticality. A configuration is defined by a set of parameters characterizing the amount and physical arrangement, at a specific location, of the materials that can significantly affect criticality (e.g., fissile materials, neutron absorbing materials, reflecting materials, and moderators). The variety of possible configurations is best understood by grouping them into classes. A configuration class is a set of similar configurations whose composition and geometry is defined by specific parameters that distinguish one class from another. Within a configuration class, the values of configuration parameters may vary over a given range.

A master scenario list and set of configuration classes relating to internal criticality is given in the *Disposal Criticality Analysis Methodology Topical Report*.[4] This list was developed by a process that involved workshops and peer review. The comprehensive evaluation of disposal criticality for any waste form must include variations of the standard scenarios and configurations to ensure that no credible degradation scenario is neglected. All of the scenarios that can lead to criticality begin with the breaching of the waste package, followed by entry of water, which eventually leads to degradation of the spent nuclear fuel and/or other internal components of the waste package.

2.2 Application of Standard Scenarios to Melt and Dilute Ingots

The MD SNF has the following characteristics in terms of geometrical arrangement inside the codisposal WP and the distribution of the neutron absorber:

1. There is no internal structure inside the DOE SNF canister. The ingots fill most of the space inside the DOE SNF canister and thus do not need a support structure, but a carbon steel crucible liner may encase the MD ingot. This implies that configurations following from degradation of DOE SNF canister basket structure are not valid for the MD SNF disposal.

2. Neutron absorber and SNF are merged metallurgically in the ingot. Physical separation of neutron absorber is not possible, as is degradation of the neutron absorber while fuel stays intact. This means that separation mechanisms such as differential settling of solid particles of different densities (see section 6.4.2 of Reference 6) are not applicable for the MD SNF.

Based on these characteristics the application of scenarios is as follows:

IP-1: The configurations resulting from IP-1 scenario (see Figure 2.1) involve the MD ingots degrading before other internal components and depends on the degradation rates of the various materials that make up the OICs as compared to the degradation rate of the ingots. The degradation rates show that the ingot high rate is 4.8×10^{-12} mol·cm^{-2}·s^{-1} while the low rate of the SS components is 2.5×10^{-14} mol·cm^{-2}·s^{-1}. The carbon steel has a degradation rate of 1.8×10^{-11} mol·cm^{-2}·s^{-1}. Therefore, the degradation of the carbon steel basket and the ingot, with the stainless steel and DHLW glass components intact, is possible. Since there is no basket structure in the DOE SNF canister associated with the MD ingots, configuration variations within the DOE SNF canister are limited. Possible variations are configurations with partial or total degradations of the components outside the DOE SNF canister. The DOE SNF canister falls to the bottom of the WP. Near the end of this sequence, layers of degradation products in the WP might result surrounding a partially degraded DOE SNF canister shell.

IP-2: In the configurations resulting from IP-2 scenario (see Figure 2.2), the SNF may degrade simultaneously with the other components in the WP if the environmental conditions favor glass degradation rates that are comparable to ingot and steel degradation rates. Figure 5-4 is an example. In this scenario the gradual degradation of the various constituents could result in a configuration where higher density material collects at the bottom of the waste package while lower density material stays on top. The potential for criticality could be significant if the neutron absorber (Gd as $GdPO_4$ – the most likely mineral to form enters into solution and is flushed out of the WP while the fissile material is in a geometry favorable to criticality. Because the Gd is integral to the MD ingots, this would require complete degradation of the ingots. Gd loss also depends on the fraction of $GdPO_4$ formed as a result of the geochemistry analysis.

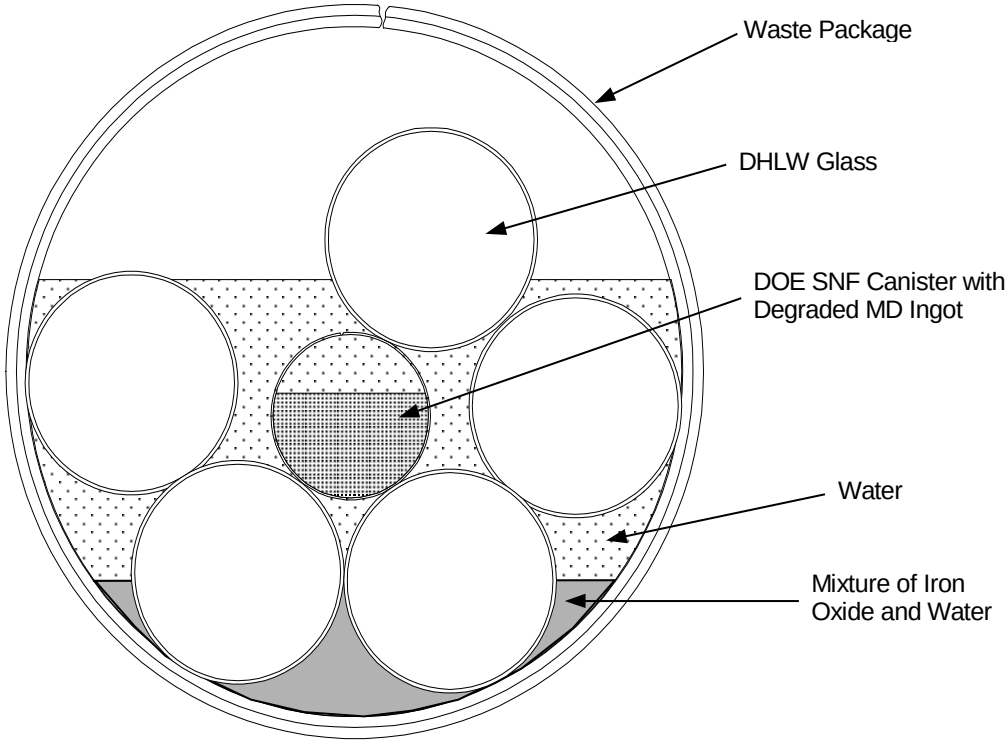

Figure 2.1 Conceptual Sketch of WP for Degradation Scenario IP-1

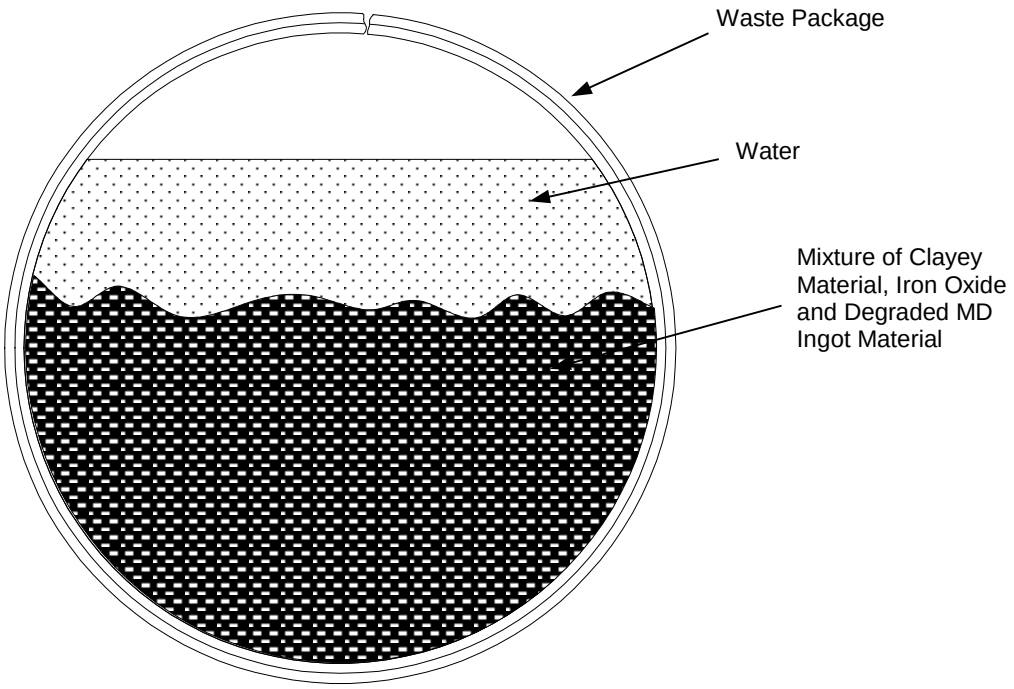

Figure 2.2 Conceptual Sketch of WP for Degradation Scenario IP-2

IP-3: The configurations resulting from IP-3 scenario (see Figure 2.3) for SNF degrading after OICs would require that the ingots have a low degradation rate and the 316L stainless steel of the DOE SNF canister have substantially lower rates than the 304L stainless steel of the DHLW canisters, along with high degradation rates for the DHLW glass. In this configuration the ingots collect at the bottom of the WP while surrounded by degradation products (e.g., clayey material). As long as the ingots are intact there is no possibility for criticality since the neutron absorber is maintained. Loss of the neutron absorber ($GdPO_4$) if it enters solution and is flushed out of the WP while the fissile material is in a geometry that is favorable to criticality should be considered. Possible variations are configurations with DOE SNF canister degraded and intact SNF accumulated at the WP bottom with partial or total degradation of WP components.

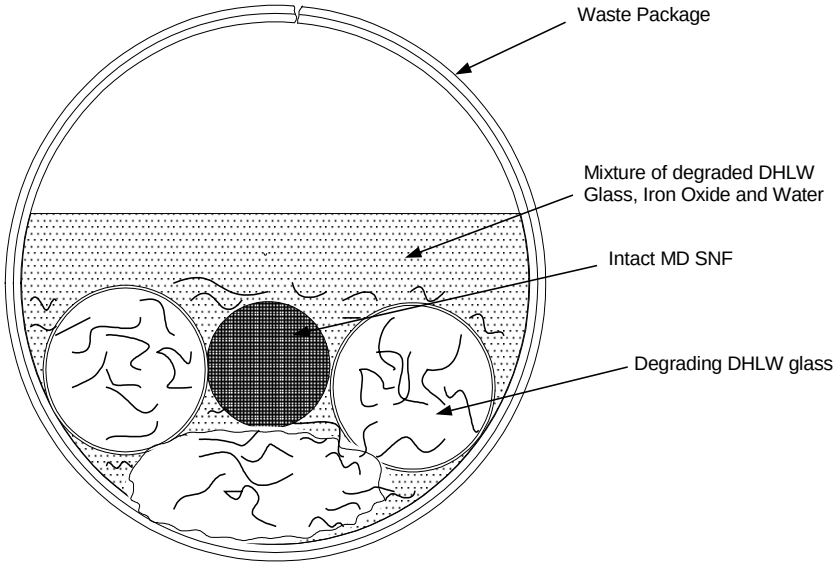

Figure 2.3 Conceptual Sketch of WP for Degradation Scenario IP-3

It should be noted that for the scenarios presented "flushing out of the neutron absorber" requires that water over-flows through the hole in the top of the WP.

Other degradation scenarios designated as IP-4, IP-5 and IP-6 that allow for water flow-through require a top and bottom breach in the waste package. However, for these scenarios to lead to potential critical configurations there must be some plugging of the hole(s) in the bottom, so that water can accumulate to provide neutron moderation. In addition, geochemistry calculations assume that a material does not get flushed out unless it is in solution. Therefore, the resulting configurations are the same as the configurations for the top breach only cases (IP-1, IP-2 and IP-3).

3.0 CRITICALITY ANALYSIS

3.1 Items Important to Criticality Control and Acceptance

As part of the criticality licensing strategy, items that are important to criticality control will be identified during evaluation of the representative fuel types designated by the National Spent Nuclear Fuel Program. As a result of the analyses performed for the evaluation of the codisposal viability of Al-based DOE-owned fuel, several items are identified as important to criticality control. The DOE SNF canister shell is naturally an item that is important to criticality control since it initially confines the fissile elements to a specific geometry and location within the waste package. The fissile mass limit in the canister, the linear density of the ^{235}U in the DOE SNF canister, and the fuel enrichment are also important to criticality control.

All calculations are based on a maximum of 38.3 kg ^{235}U per DOE SNF canister. The degraded configurations of the Melt and Dilute ingots bound the other types of Al-based DOE-owned spent nuclear fuel, as long as the limits on mass of uranium and its enrichment, and the linear density are not exceeded.

Hence, the total mass of fissile element (^{235}U) should not exceed the mass used in deriving the conclusions of this report, which is 38.3 kg of ^{235}U per DOE SNF canister. The maximum ^{235}U enrichment is 20 wt%. The linear density of the ^{235}U should not exceed 151 g/cm in the DOE SNF canister. This value is calculated by considering the maximum diameter and the maximum U content (18.2 wt%) for the MD ingots.

Table 3.1 indicates the relationships between the degradation scenarios described in Section 2.0 that could lead to removal of soluble neutron absorbers from the waste package and the geochemistry and criticality models (see Section 4.0). Criticality models to analyze the configuration classes shown are identified and discussed below.

3.2 Computer Software

The Monte Carlo particle transport code, MCNP, Version 4B2LV[7], is used to estimate the effective neutron-multiplication factor (k_{eff}) of the codisposal waste package. The information regarding the code and its use for the criticality analysis is documented in Reference 8.

The MCNP Version 4B2LV is used to estimate the k_{eff} values for various geometrical configurations of the MD SNF in the 5-DHLW/DOE SNF-short waste package. The k_{eff} results represent the average combined collision, absorption, and track-length estimator from the MCNP calculations. The standard deviation (σ) represents the standard deviation of k_{eff} related to the average combined collision, absorption, and track-length estimate due to the Monte Carlo calculation statistics. The calculations are performed using ENDF/B-V continuous energy cross-section libraries that are part of the qualified MCNP code system.

The MCNP calculated results are presented in the following sections to demonstrate that all foreseeable intact and degraded configurations inside the codisposal waste package have been investigated and the values of k_{eff} are below the interim critical limit of 0.93. Although each of the degradation configurations discussed in Reference 4 is not specifically modeled, the criticality configurations selected were bounding type analyses and therefore extensive criticality sensitivity analyses were not necessary.

3.3 Intact Geometry Criticality Analysis

This section presents the results of an intact-geometry mode criticality analysis representing a water intrusion situation. This criticality configuration (see Figure 3.1) represents a waste package, which has been breached, allowing inflow of water, and wherein the internal components of the waste package have maintained their as-loaded geometry. For all these calculations (unless otherwise specified), the waste package has reflected boundaries acting as a mirror (i.e., no neutron leakage) and represents a very conservative approach. Variations of postulated water intrusion were examined to identify the configuration that results in the highest calculated k_{eff} value for a range of possible water intrusion conditions. The results are shown in Table 3.1.

Intact cases were investigated first with a gadolinium loading of 0.5 wt% and the MD SNF form completely filling the DOE SNF canister. For these cases, approximately 212 kg of U and 5.8 kg of Gd are used. Cases were run with the MD ingot composition filling the interior of the DOE SNF canister and the 10% void in the MD ingots dry, half-filled, and filled with water. Ingot/gap height combinations from 10 to 60 cm high were also run to investigate the effects of ingot height. An additional case was evaluated to determine the effect of 2 wt% Si in the MD ingot composition. This case simulates the composition of the MD-SNF form that is expected to result from the treatment of U-Al-Si base, Al-clad SNF.

The results show that the configuration with the wet ingot (10% void in the MD ingots filled with water) has higher $k_{eff}+2\sigma$ than the case with dry ingots. The addition of 2 wt% Si in the MD ingot has negligible effect to criticality (0.3561 versus 0.3571 for case without Si). The highest $k_{eff}+2\sigma$ occurs for the case with the wet ingot filling the entire DOE SNF canister. For this case $k_{eff}+2\sigma = 0.3571$. Variation of ingot height $k_{eff}+2\sigma$ results in a lower $k_{eff}+2\sigma$. A highly moderated case without Gd in the waste package resulted in a $k_{eff}+2\sigma$ below 0.90, thereby confirming, for the water-filled but intact waste package, no Gd is required in the MD ingots to maintain subcriticality.

This finding is significant since it represents potential conditions prior to the onset of long-term degradation. The following sections discuss degraded conditions represented by the IP-1, IP-2, and IP-3 configurations described in Section 2.0.

Table 3.1 Criticality Evaluation Summary for Degraded Configurations

Master Scenarios	Description		Configuration Classes	Geochemistry Results	Criticality Results			Comments
					Reference	% of initial Gd	$(k_{eff}+2\sigma)_{max}$	
IP-1	Liquid accumulates in WP	MD ingots degrade before OICs	IP-1-A/B: MD ingots degraded; DOE SNF canister shell intact	For most conditions and variations of parameters the geochemistry results indicated that the minimum content of Gd remaining at any time inside the DOE SNF canister or WP is at least 80% of the initial content.	Section 3.4 Figure 3.2 Tables 3.3 and 3.4	10	0.800	$(k_{eff}+2\sigma)_{max} < 0.93$
			IP-1-C: All WP components degraded		Section 3.5.1 Figures 3.3 and 3.4 Table 3.5	0	0.927	$(k_{eff}+2\sigma)_{max} < 0.93$
IP-2		MD ingots degrade concurrently with OICs	IP-2-A: All WP components degraded		Section 3.5.2 Figures 3.3 and 3.4 Table 3.6	0	1.019	At least 2.5% of initial Gd content must remain in the WP bottom layer mixed with U to prevent criticality.
						2.5	0.883	
IP-3			IP-3-C: All WP components degraded		Section 3.6.1 Figure 3.4 Tables 3.7, 3.8 and 3.9	0	0.764	$(k_{eff}+2\sigma)_{max} < 0.93$
		MD ingots degrade after OICs	IP-3-A: MD ingots and DOE SNF canister shell intact; WP basket degraded		Section 3.6.3[a] Figures 3.4 and 3.5 Table 3.11	1	1.268	At least 7.5% of initial Gd content must remain inside the DOE SNF canister mixed with U to prevent criticality.
						7.5	0.900	
			IP-3-B: MD ingots intact; WP basket, DHLW glass and DOE SNF canister degraded		Section 3.6.2 Figure 3.7 Table 3.10	10	0.816	$(k_{eff}+2\sigma)_{max} < 0.93$
IP-4	WP bottom is penetrated allowing liquid to flow through	MD ingots degrade before OICs	These classes were not discussed (see the explanation in the second cell at right)	The geochemistry calculation does not address this group of scenarios (see explanation in the next cell at right).	All cases are bounded by IP-1, IP-2 and IP-3 as the latter have better moderation (water is pooling inside WP), and more favorable conditions for neutron absorber loss (mainly Gd) concurrent with high fissile material (U) retention.			-
IP-5		MD ingots degrade concurrently with OICs						
IP-6		MD ingots degrade after OICs						

NOTE: [a] MD ingots are degraded.

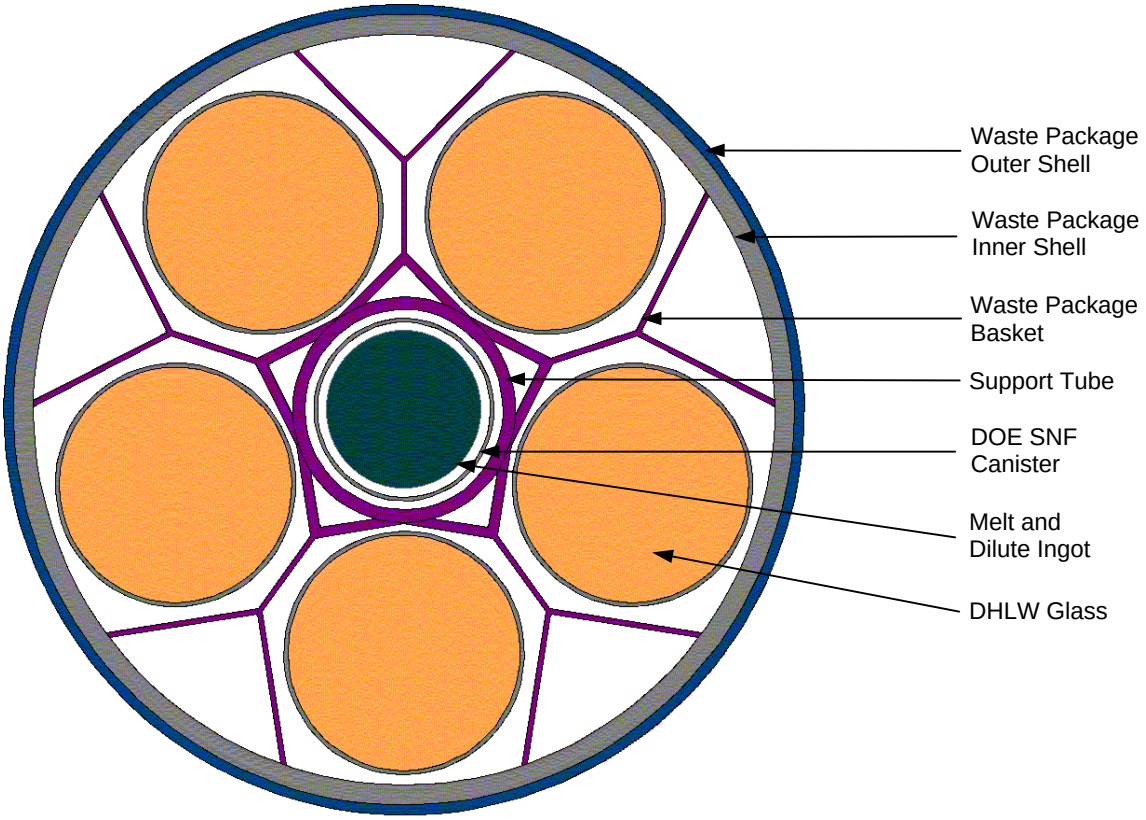

Figure 3.1 Cross-section View of the 5-DHLW/DOE Waste Package in an as-Loaded (intact) Configuration

Table 3.2 Results for Intact Mode Configurations

Ingot Height	Ingot Type	wt% Gd	$k_{eff}+2\sigma$
Full canister height	Dry ingot Dry waste package	0.5	0.1521
Full canister height	Wet ingot	0.5	0.3571
Full canister height	Dry ingot Filled waste package	0.5	0.2155
Full canister height	Half-wet ingot	0.5	0.2969
Full canister height	Wet ingot w/ 2 wt% Si	0.5	0.3561
19cm (1cm water gap)	Wet ingot	0.5	0.3475
29cm (1cm water gap)	Wet ingot	0.5	0.3464
59cm (1cm water gap)	Wet ingot	0.5	0.3450
9cm (1cm water gap)	Wet ingot	0.5	0.3482
8cm (2cm water gap)	Wet ingot	0.5	0.3014
8cm (2cm water gap)	Wet ingot	0.0	0.8949

3.4 Waste Form Degrades Before the Internal Components of the Waste Package

In this section, cases where the waste form degrades before any other internal components of the waste package are investigated and corresponds to configuration IP-1 (see Figure 2.1, Section 2.2). This configuration assumes a rapid degradation of the ingots in the canister while the rest of the waste package internals remain intact. For the purpose of the calculations presented in this section, the configuration shown in Figure 3.2 was used. This configuration is different from that of Figure 2.1 since most conservative features were added, i.e., the waste package internal components (but external to the DOE SNF canister) were considered intact and at the closest position relative to the DOE SNF canister and the waste package was filled with water (for best reflection and moderation). The uranium is conservatively represented in the form of UO_2 that is distributed in the canister. The incorporation of U degradation products as UO_2 maximizes the available volume for water, and thus, for H. This UO_2 is mixed with water, gibbsite [$Al(OH)_3$] and gadolinium. Above this MD bearing mixture is a mixture of water and gibbsite. In all the cases considered, various amounts of the ingot Al material were assumed to remain in the canister and 90% of the original gadolinium was assumed to be dissolved and removed. The amount of gibbsite that was removed from the canister was varied in order to observe the effects of different degrees of moderation. Cases were run with the height of the accumulation of degradation products in the canister varying from 10 to 40 cm (chord height in Table 3.3). For each height investigated the gibbsite fraction was varied from 100% to 0% of the original mass (water replaces gibbsite). The results in the geochemistry calculation indicate that less than 20% of the initial Gd content would be lost in this configuration[9], so these cases have a factor of 8 margin in Gd concentration. Degradation of most of the steel components would also be required to allow the loss of the Gd from the degraded MD ingots.

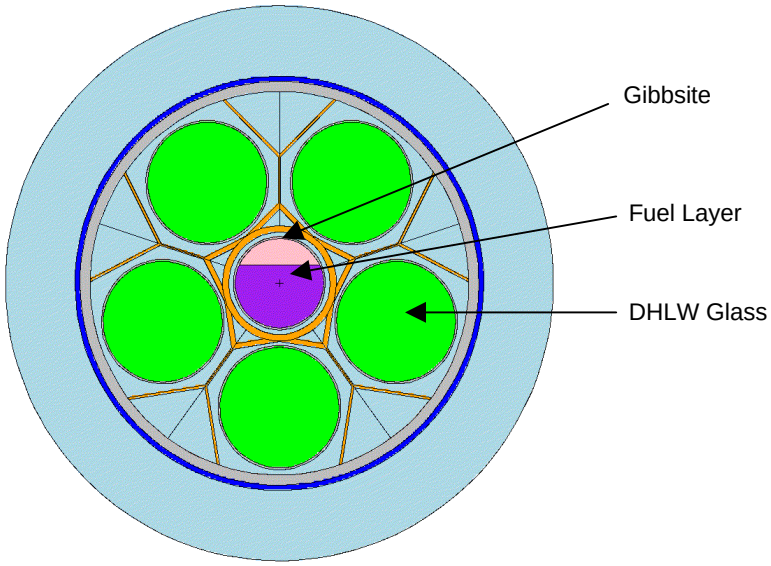

Figure 3.2 Cross-section View of Degraded Fuel in an Intact Waste Package

Table 3.3 shows results for criticality calculations where the void regions in the waste package are filled with water and the content of the gadolinium remaining in the waste package is chosen to be consistent with 90% of the original gadolinium leaving the system. Various mixtures of UO_2, gibbsite, and water are run to identify optimum compositions. Water replaces gibbsite as the water fraction goes up. Table 3.4 shows results for calculations where the void regions of the waste package are left void. These cases have approximately 191.3 kg of U and 0.5 kg of Gd in the degraded fuel layer (constrained by the original

dimensions of the ingots). The column titled water volume % in Table 3.3 and Table 3.4, refers to the available volume after UO_2 is considered.

Cases were run with the height of the accumulation of degradation products in the canister varying from 10 to 40 cm. For each height investigated the gibbsite fraction was varied from 100% to 0% of the original mass in the space not occupied by UO_2 (water replaces gibbsite) in the layer. The water fraction in the gibbsite/water layer above the UO_2-bearing layer matches the UO_2-bearing layer value. Under all circumstances $k_{eff}+2\sigma$ remained under 0.80 with a factor of 8 margin on Gd mass as indicated by the geochemistry results, and the maximum value corresponds to a case with an accumulation 30 cm deep with all the gibbsite present. The cases with void space filled with water have slightly lower $k_{eff}+2\sigma$ values than corresponding non-physical cases with empty void space.

Table 3.3 Results for Degraded Fuel in Intact DOE SNF Canister and Waste Package

Chord Height (cm)	Water Volume %	$k_{eff}+2\sigma$
10	0	0.6609
10	20	0.6791
10	40	0.6743
10	60	0.6736
10	80	0.6694
10	100	0.6699
20	0	0.7507
20	20	0.7671
20	40	0.7612
20	60	0.7610
20	80	0.7559
20	100	0.7488
30	0	0.7852
30	20	0.7822
30	40	0.7782
30	60	0.7804
30	80	0.7712
30	100	0.7682
40	0	0.7818
40	20	0.7816
40	40	0.7766
40	60	0.7800
40	80	0.7757
40	100	0.7740

NOTE: In all cases the void inside waste package is filled with water.

Table 3.4 Results for Degraded Fuel in Intact DOE SNF Canister and Waste Package

Chord Height (cm)	Water Volume %	$k_{eff}+2\sigma$
10	0	0.6922
10	20	0.6933
10	40	0.6874
10	60	0.6913
10	80	0.6886
10	100	0.6832
20	0	0.7831
20	20	0.7824
20	40	0.7765
20	60	0.7735
20	80	0.7759
20	100	0.7704
30	0	0.7994
30	20	0.7949
30	40	0.7949
30	60	0.7903
30	80	0.7854
30	100	0.7858
40	0	0.7947
40	20	0.7938
40	40	0.7981
40	60	0.7957
40	80	0.7902
40	100	0.7929

NOTE: In all cases the void inside waste package is empty.

3.5 All Components Internal to Waste Package Degraded

These configurations represent the final stage of degradation (IP-2) at which time all components in the waste package have formed the geometry illustrated by Figure 3.3, and water surrounds the degraded package shown. Reference 9 gives the composition of the clay resulting from the degradation of the internal components of the waste package. This clay is referred as post-breach clay. If all of the U is eventually removed while $GdPO_4$ remains there is no potential for criticality. However, if the Gd is removed before the U is all gone criticality can occur. Homogenizing the Gd in the clay will only increase its effectiveness in absorbing neutrons.

Table 6 of Reference 9 gives a post-breach degraded component composition for an alternative EQ3/6 case. In this case, the MD ingots degrade with the steel components of the waste package, but before the DHLW glass degrades and removes the U. However, since suppression of $GdPO_4$ formation in the EQ3/6 calculations led to the removal of Gd before the U is removed (see Section 4.4), a number of criticality runs were made to investigate such sensitivities.

MCNP cases were run where the amount of water in this mix is varied to determine the optimum moderation. The maximum water fraction in the fuel-bearing layer is very conservatively represented to be 50%. Cases were run with a layer of UO_2 mixed with water and for various mixtures of UO_2, water, and degraded components. Determination of the minimum mass of Gd or Hf is made to cause the $k_{eff}+2\sigma$ to fall below 0.93. The geochemistry calculation demonstrates that Hf remains in the DOE SNF canister or waste package in each of the limited number of conditions considered.[9]

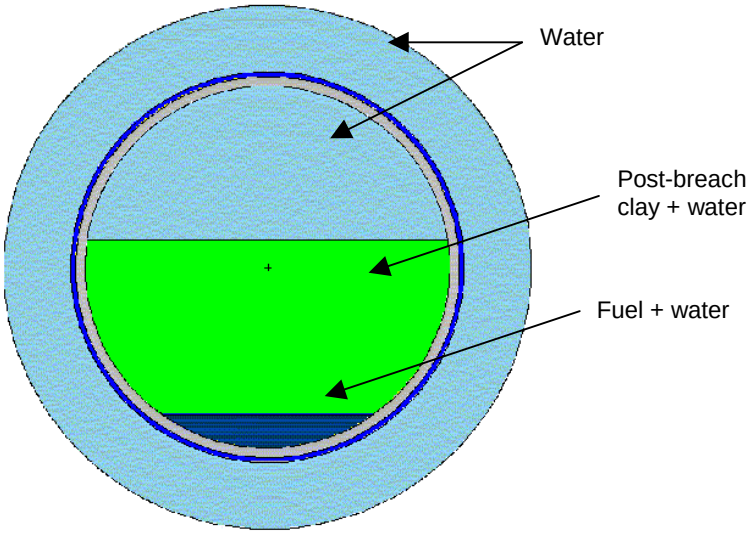

Figure 3.3 Cross-section View of WP with All Components Degraded

The effect of limited tilting of the waste package to the $k_{eff}+2\sigma$ was also investigated for a tilt angle of 13.72° as shown in Figure 3.4. The slope is chosen arbitrarily and the case included for completeness.

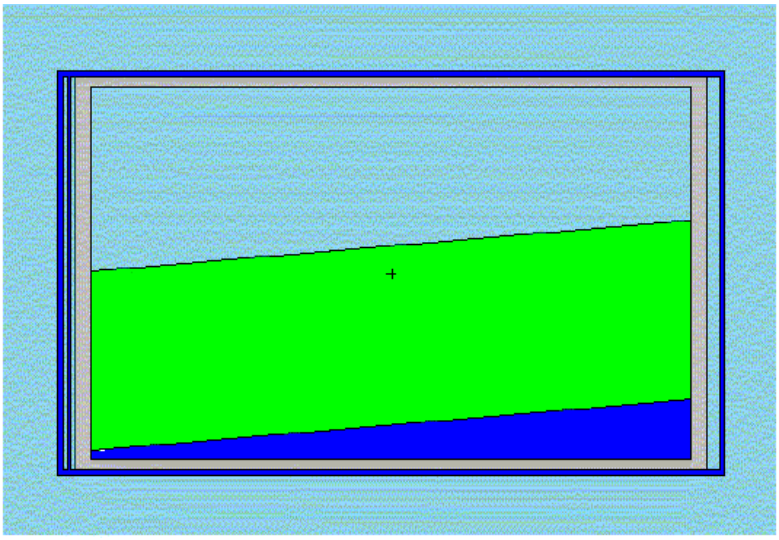

Figure 3.4 Cross-section View of Simulated Tilt of Waste Package

3.5.1 Stratified Layers of UO_2 and Post-breach Clay Without Neutron Absorber

Table 3.5 shows results for cases comprised of a fuel layer (UO_2 and water) on bottom and clay layer on top. The cases are for 50% water content and 75% water content in the fuel layer, respectively and there is no neutron absorber in the fuel layer and are represented by Figure 3.3. These cases have 191.3 kg of U in the fuel layer.

Table 3.5 Results for Stratified UO_2 and Clay Inside Waste Package

Water Content (%)	$k_{eff}+2\sigma$
50	0.6949
75	0.9270

The result shows that there is no criticality concern for this particular configuration when the water content in the fuel layer is 50%. The case with 75% water content in the fuel layer shows $k_{eff}+2\sigma$ of 0.9270, which is just below the critical limit. However, this configuration is not realistic due to lack of physical mechanism that could promote homogenization of 25% UO_2 with 75% water in stratified layers.

3.5.2 Layers of Fuel Mixed with Clay

Table 3.6 shows criticality analysis cases for a layer of UO_2 mixed with the alternate post-breach clay composition corresponding to the extreme case where Gd is lost (see Sections 4.4 and 4.5) sitting on the bottom of the waste package as illustrated by Figure 3.3. This post-breach clay composition, without U, is on top. All these cases have 191.3 kg of U in the fuel layer. Following are cases to demonstrate the minimum mass of Hf and Gd required to prevent criticality.

Table 3.6 Layers of Fuel Mixed with Clay

Case #	Case Description	Clay Content (vol%)	Water Content (vol%)	UO_2 Content (vol%)	Neutron Absorber		$k_{eff}+2\sigma$
					Type	Content (g)	
1	Configuration shown in Figure 3.3	25	50	25	-	-	0.8074
2		30	50	20	-	-	0.8490
3		40	50	10	-	-	0.9654
4		45	50	5	-	-	1.0182
5		47.50	50	2.50	-	-	0.9510
6		45	50	5	Gd	525.59	0.6642
7		45	50	5	Gd	262.8	0.7862
8		45	50	5	Gd	131.4	0.8825
9		45	50	5	Hf	525.59	1.0124
10		45	50	5	Hf	5255.9	0.9347
11	Similar to case 6, but the WP is tilted 13.72°	45	50	5	Gd	525.59	0.7285
12	Similar to case 8, but 30-cm thick water is used as a reflector	45	50	5	Gd	131.4	0.8096
13	Similar to case 8, but 30-cm thick tuff is used as a reflector	45	50	5	Gd	131.4	0.8307

Table 3.6 shows that approximately 2.5% of the original Gd loading (131.4 g) must remain with this mixture to prevent criticality or approximately 25% of the Hf (approximately 5 kg) in the alternate MD ingot composition must remain. The geochemistry calculations (BSC 2001c) have demonstrated that Hf remains in the DOE SNF canister or waste package in each of the limited number of conditions considered. If confidence in the thermodynamic data for $GdPO_4$ formation is not sufficient to make the loss of Gd incredible, then the MD ingot composition with Gd and Hf will prevent a critical condition.

The effect of tilting the waste package is investigated in case 11 (see Table 3.6), for the maximum tilt angle possible. The $k_{eff}+2\sigma$ increased to 0.7285, which is significantly less than the critical limit.

Replacing the reflective boundary condition with a 30-cm thick water or tuff reflector decreases $k_{eff}+2\sigma$. This shows that use of the reflective boundary condition for this case is very conservative.

3.6 Internal Components of the Waste Package Degraded (outside intact DOE SNF canister) and Intact Ingots

This section describes configurations resulting from the scenario IP-3 (see Figure 2.3, Section 2.2) For the purpose of the calculations presented in this section, the configuration shown in Figure 3.4 was used. This configuration is different from that represented by IP-3 (Figure 2.3) in the sense that the features considered most conservative were added, i.e. the waste package internal components (external to the DOE SNF canister, which was considered intact) were considered completely degraded into a homogeneous slurry in which the DOE SNF canister is completely immersed (for best reflection). The composition of the slurry is given in Table 6 of Reference 9 and is referenced as the pre-breach clay. The amount of water mixed in this clay varies. There is ^{238}U present in the slurry from the degraded glass, but it is neglected in these calculations. The MD ingot-bearing canister is assumed to have dropped down to

the bottom of the waste package and is surrounded by a mixture of water and clay. The geometrical configuration is shown in Figure 3.5. Although the DOE SNF canister and MD ingots are assumed intact, they are also assumed fully flooded with water, which was determined to be the most reactive composition. Since the MD ingot is flooded, it is assumed 10% water by volume. This conservatively bounds the dry ingot case, since filling the porosity with water will increase (to the maximum limit) the moderation of the already under-moderated intact ingots. Both the gadolinium content of the ingots and the water volume fraction in the clay were varied. The density of dry pre-breach clay is 3.682 g/cm^3.[9]

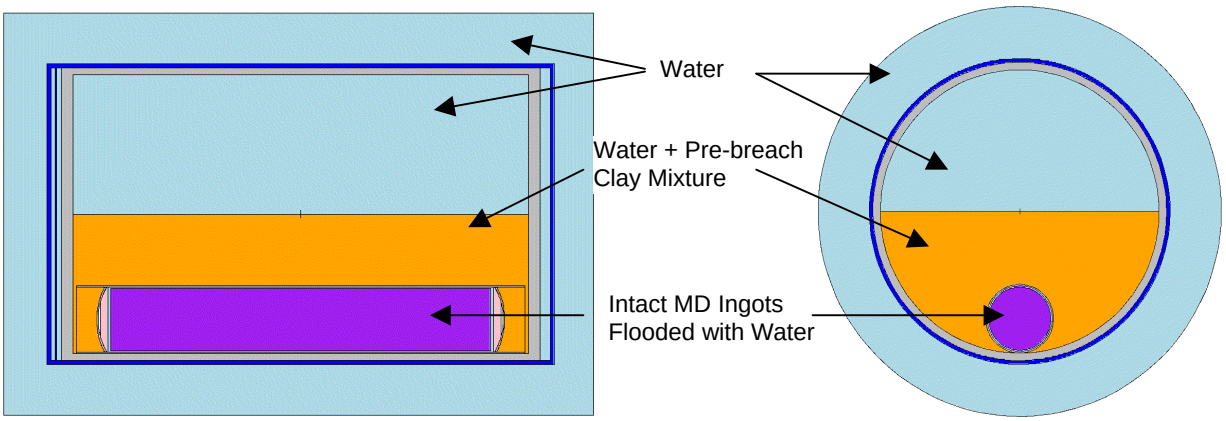

Figure 3.5 Cross-section Views of Intact DOE SNF Canister Degraded Internal Waste Package Components

The next stage of degradation involves the configuration described above with the degraded MD ingots within the DOE SNF canister as discussed in Section 3.4. The most conservative conditions identified in the previous calculations were used to characterize this combination. The minimum mass of Gd required to remain in the canister was identified and the effect of thinning the canister wall was investigated. Another case was run to simulate the effect of tilting of the DOE SNF canister inside the waste package as shown in Figure 3.6. The volume of the fuel region was conserved while the tilt angle was chosen as the maximum tilt that is conceivable for the DOE SNF canister inside the waste package, which is 13.72°.[8]

A bounding additional configuration was considered where ingots form an array inside the waste package and are surrounded by water as shown in Figure 3.7. The gadolinium linear density was chosen to be consistent with 90% of the original gadolinium leaving the system. Calculation was performed for short ingots (25.4 cm) forming an array with 9 units and for long ingots (76.2 cm) forming a array with 3 units inside the waste package.

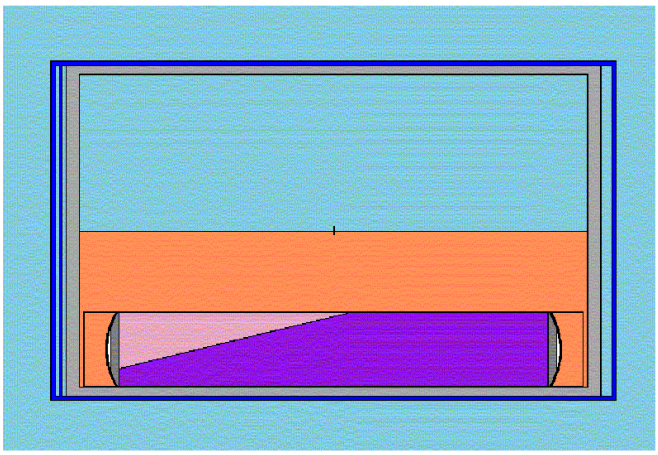

Figure 3.6 Cross-section View of Simulated Tilt of Intact DOE SNF Canister with Degraded Fuel and Degraded Internal Waste Package Components

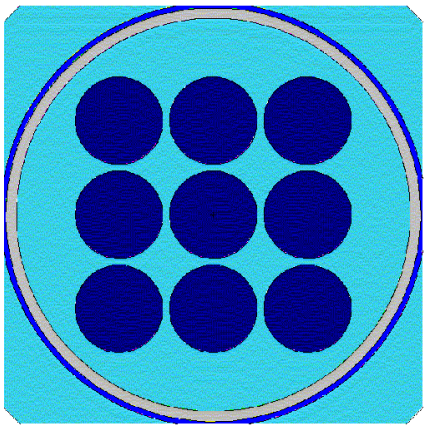

Figure 3.7 Array of 9 Ingots Inside the Waste Package Surrounded with Water

3.6.1 Wet Intact Ingots with Full or Partial Gd

Table 3.7 presents the results of the calculations for wet ingots with full Gd content of 0.5 wt% (5.8 kg/waste package) and a U content of 212 kg, corresponding to the MD fuel form completely filling the canister. The composition of the wet intact ingots is as described in Section 3.6 and corresponds to the highest k_{eff} values in Section 3.3. The basic case is illustrated in Figure 3.5, which shows the DHLW glass canisters and all the basket structure as degraded and forming a layer of pre-breach clay surrounding the DOE SNF canister.

Table 3.7 Wet Ingots with Full Gd Content

Water Content in Clay (volume %)	$k_{eff}+2\sigma$
0	0.4264
20	0.3934
40	0.3736
60	0.3589
80	0.3521
100	0.3428

k_{eff} +2σ is highest for the case when there is no water present in the pre-breach clay.

Table 3.8 presents the results of the calculations for wet ingots with partial Gd content of 0.05 wt% (0.58 kg/WP) and the MD fuel form completely filling the canister.

Table 3.8 Wet Ingots with Partial Gd Content

Water Content in Clay (volume %)	$k_{eff}+2\sigma$
0	0.6075
20	0.5601
40	0.5335
60	0.5146
80	0.5005
100	0.4949

Again, k_{eff} +2σ is highest for the case when there is no water in the pre-breach clay.

Table 3.9 presents the results of the calculations for wet ingots with partial Gd content of 0.025 wt% (0.29 kg/WP), Gd content of 0.0025 wt% (0.029 kg/WP) for the next before the last case and no Gd for the last case. The MD fuel form is filling the canister completely.

Table 3.9 Wet Ingots with Low Gd Content

Water Content in Clay (volume %)	Gd Content (g/WP)	$k_{eff}+2\sigma$
0	263	0.6586
20	263	0.6083
40	263	0.5840
60	263	0.5653
80	263	0.5530
100	263	0.5476
0	26.3	0.7503
0	0	0.7634

k_{eff} +2σ is highest for the case when there is no water in the pre-breach clay. Even without any Gd in the waste package k_{eff}+2σ is well below 0.93.

3.6.2 Intact Melt and Dilute Ingots in the Waste Package

Table 3.10 shows results for cases of ingots forming an array inside the waste package. The maximum value of k_{eff}+2σ is 0.8157 for case with 3 ingots. The first case is an array of 9 short ingots as shown in Figure 3.7 and the second case is for 3 long ingots. The ingots are sized such that they all fit within the DOE SNF canister. These cases have 0.05 wt% Gd.

Table 3.10 Results for Array of Ingots Inside Waste Package

Ingot Length (cm)	k_{eff}+2σ
25.4	0.7257
76.2	0.8157

3.6.3 Degraded Melt and Dilute Ingots

This configuration has degraded ingots within the intact DOE SNF canister as described in Section 3.4, but with degraded waste package internals. This configuration would be similar to the one shown in Figure 3.4, but with degraded MD ingots. The composition with which the highest k_{eff}+2σ values are associated from Section 3.4 is used (the case in Table 3.4 with a chord height of 30 cm and a water volume of 0%). The pre-breach clay with 0% water, with which the highest k_{eff} values are associated in Section 3.4 is used. Table 3.11 presents the results for this configuration. These cases have approximately 191.3 kg of U. The initial case has 10% of the original Gd loading.

Table 3.11 Degraded Ingots in Intact DOE SNF Canister with Degraded Waste Package Internals

Case #	Case Description	Gd Content in WP		k_{eff}+2σ
		% of Initial Gd	(g)	
1	Degraded Ingots in the DOE SNF Canister and Intact Waste Package (chord height is 30 cm and water volume 0%)	10.0	525.59	0.8161
2		7.5	394.19	0.8914
3		5.0	262.80	1.0031
4		2.5	131.40	1.1425
5		1.0	52.56	1.2678
6	Similar to case 2, but the DOE SNF canister wall thinned to 0.3175 cm	7.5	394.19	0.8940
7	Similar to case 1, but the DOE SNF canister is tilted 13.72°	10.0	525.59	0.8443
8	Similar to case 2, but 30-cm thick water is used as a reflector	7.5	394.19	0.8810
9	Similar to case 2, but 30-cm thick tuff is used as a reflector	7.5	394.19	0.8810

The results show that retention of at least 7.5% of the original Gd inventory assures k_{eff}+2σ below 0.93, but reducing the Gd inventory below this value could result in values significantly over 1.0. Comparing the results of the first case in Table 3.10 with those for the case in Table 3.4 with a chord height of 30 cm and a water volume of 0%, leads to the observation that the current configuration is bounding.

The thinning of the canister wall down from 0.9525 cm to 0.3175 cm increases k_{eff}+2σ by less than 1% (k_{eff}+2σ of 0.8915 versus 0.8850). The effect of a 13.72° tilt (for the first case in Table 3.10) to k_{eff}+2σ is an increase of approximately 1.5%.

Replacing the reflective boundary condition with a 30-cm water or tuff reflector (cases 8 and 9 in Table 3.11, respectively) decreases $k_{eff}+2\sigma$. This shows that use of the reflective boundary condition is slightly conservative.

3.7 Most Probable Degradation Path

Based on the corrosion rates and the material thickness given in Table 3.12 and Table 3.13, respectively, the most probable degradation path for the waste package, the DOE SNF canister, and the MD ingots follows the sequence below:

1. Waste package is penetrated and flooded internally. Water has not yet penetrated the DOE SNF canister (see Table 3.1) for the results of criticality calculations for this configuration).

2. The waste package separation plates and DOE SNF canister support cylinder degrade first because of the high corrosion rate of A516 carbon steel. Degraded steel product (iron oxide) accumulates at the bottom of the waste package (criticality calculations in Section 3.3 are bounding for this configuration).

3. DHLW glass canister shell degrades and exposes the DHLW glass. The DHLW glass degrades at a much lower rate than the stainless steel components and only a small percentage degrades while the stainless steel degrades as demonstrated in the geochemistry calculations.[9] There are two possible degradation paths:

    3.a. DOE SNF canister stays intact. Intact DOE SNF canister with intact MD ingots fall and are surrounded by the iron-rich degradation products near the bottom of the waste package (see Section 3.6.2 for criticality calculations results and Figures 3.5 and 3.6 for configurations).

    3.b. DOE SNF canister starts to degrade.

Following 3b above, DOE SNF canister shell is penetrated but remains intact and DOE SNF canister interior is flooded (Section 3.6, Figure 3.5, and Figure 3.6 cover this case).

4. MD ingots in the DOE SNF canister are in contact with water. MD ingots start to degrade due to their high corrosion rate. The MD ingots degrade into hydrated Al and U oxides and Gd phosphate[9] (see Section 3.6.3 for criticality calculations results and Figure 3.5 and Figure 3.6 for configurations).

5. DOE SNF canister shell completely degrades. The degraded iron oxide mixes with the small percentage of degraded glass clay at the bottom of the waste package. The degraded MD ingot material falls out and scatters on top of or mixes with the clay/iron oxide mixture (see Section 3.4 for criticality calculations results and Figure 3.3 and Figure 3.4 for configurations).

6. Degraded glass clay product accumulates at the bottom of the waste package over or mixed with the iron-rich degradation products from the other OICs and the MD ingots (covered in Section 3.4).

A variation of the above sequence would retain the DOE SNF canister and subsequent degradation products trapped in the center of the DHLW glass logs, but the result is essentially the same.

Given a very long period of time, it is postulated that everything will degrade. All the internal components of the waste package will then be represented as sludge. This corresponds to degradation scenario group IP-2. The degraded MD ingots and other degradation products could mix and pile up near

the bottom of waste package. However, there is no mechanism to cause complete and uniform mixing of all the degradation products inside the waste package.

Table 3.12 Properties of Materials in Melt and Dilute Codisposal Waste Package

Reactant	*Rk* Reaction Rate (mol/cm^2·s)			*sk* Surface Area (cm^2)
	1	2	3	
DHLW Glass	rk1=8.858E-19 cdac1= -0.4 rk2=7.976E-13 cdac2= 0.6	rk1=1.076E-17 cdac1= -0.4 rk2=4.874E-12 cdac2= 0.6	Not Applicable (N/A)	1228.64
Ingots	1.399E-14	2.536E-13	4.830E-12	11.66
A516 Ingot Liner	1.798E-11	N/A	N/A	24.60
304L Glass Pour Canister	2.520E-14	2.520E-13	8.656E-12	141.5
A516 Outer Web	1.798E-11	N/A	N/A	120.0
A516 Impact Plates	1.798E-11	N/A	N/A	1.615
316L DOE SNF Canister	2.530E-14	2.530E-13	5.056E-13	22.67
316NG Inner Shell	2.530E-14	2.530E-13	5.056E-13	57.3

The degradation rates in Table 3.12 range from low to high (indicated by "1" to "3"). The true reaction rate is obtained by multiplying the reaction rate (*rk*) by the surface area (*sk*) to get mol/s. Inspection of the rates show that for a comparable surface area, the A516 carbon steel is expected to degrade much more rapidly than the stainless steels (316L, 316NG, and 304L). The ingot rate 1, from Reference 10, is the degradation rate given for U-Al fuel types. The ingot rate 2 is the degradation rate of pure aluminum 1100.

The outer web is composed of A-516 carbon steel, and serves two purposes: it centers, holds in place, and separates the DOE Canister and the GPCs; and prevents them from transmitting undue stress to each other in the event of a fall (tip-over) of the entire WP. In a breach scenario, the A516 WP components will be exposed to water and corrode before the rest of the WP, and are expected to degrade within a few hundred to a few thousand years.

Table 3.13 Materials and Thickness

Components	Material	Thickness (mm)
Waste package divider plate	A516 Carbon Steel	12.7
Waste package support tube	A516 Carbon Steel	31.75
High-level waste glass canister shell	304L Stainless Steel	9.525
High-level waste glass	Glass	N/A
DOE SNF canister shell	316L Stainless Steel	9.525
MD ingots	U-Al alloy	381 - 419

3.8 Intact and Degraded Component Criticality Conclusions

The criticality analyses considered all aspects of intact and degraded configurations of the codisposal waste package containing Melt and Dilute ingots, including optimum moderation condition, optimum reflection, geometry and composition. The results of three-dimensional Monte Carlo criticality calculations for the intact configuration show that the requirement of $k_{eff}+2\sigma$ values less than or equal to the interim critical limit of 0.93 is satisfied for the MD codisposal package. The criticality calculations results for all anticipated degraded-mode configurations developed through the degradation analysis show that the requirement of $k_{eff}+2\sigma$ values less than or equal to the interim critical limit of 0.93 is satisfied for the MD codisposal package if at least 7.5% of the original Gd loading (394.2 g) in the ingots without Hf remains mixed with the fissile material. In the alternate MD ingot composition (containing 2.5 wt% Hf), Hf remains in the DOE SNF canister or waste package in each of the limited number of conditions considered, therefore preventing a critical condition even if all Gd is removed from the system.

4.0 GEOCHEMISTRY ANALYSIS CONSIDERATIONS

4.1 Computer Software

Geochemistry analyses were performed using the EQ3/6 Version 7.2bLV[11-12] geochemistry software package in the solid-centered flow-through mode. The information regarding the code and its use for the degradation and geochemistry analysis is documented in Reference 9.

A principal objective of the geochemistry calculations was to assess the chemical circumstances that could lead to removal of neutron absorbers (mainly Gd) from a waste package containing MD ingots (Al-SNF) and DHLW glass, while fissile materials (U) remain behind. Such circumstances could increase the probability of a nuclear criticality occurrence within the waste package. EQ6 reaction path calculations were carried out to span the range of possible system behavior and to assess the specific and coupled effects of MD ingots degradation, steel corrosion, DHLW glass degradation, and fluid influx rate on U mobilization. Corrosion product accumulation was examined as well. The results presented in Reference 9, and summarized in this section, have been used as inputs to the criticality calculations described in Section 2.0 of this document.

4.2 Basic Design Approach for Geochemistry Analysis

The method used for this analysis involves eight steps as described below:

- Use the basic EQ3/6 capability to trace the progress of reactions as the chemistry evolves, including estimating the concentrations of material remaining in solution as well as the composition of precipitated solids. EQ3 is used to determine a starting fluid composition for EQ6 calculations; it does not simulate reaction progress.
- Evaluate available data on the range of dissolution rates for the materials involved to be used as material/species input for each time step.
- Use the "solid-centered flow-through" mode in EQ6. In this mode, an increment of aqueous "feed" solution is added continuously to the waste-package system, and a like volume of the existing solution is removed. This mode simulates a continuously stirred tank reactor.
- Determine the concentrations of fissile material in solution as a function of time (from the output of EQ6 simulated reaction times up to ~ 6×10^5 years).
- Calculate the amount of fissile material released from the waste package as a function of time (which, thereby, reduces the chance of criticality within the waste package).
- Determine the concentrations of neutron absorbers (most importantly Gd) in solution as a function of time (from the output of EQ6 over times up to ~ 6×10^5 years).
- Calculate the amount of neutron absorbers retained within the waste package as a function of time.
- Calculate the composition and amounts of solids (precipitated minerals or corrosion products and unreacted waste package materials).

4.3 Geochemistry Degradation Calculations and Results

The calculations begin using selected representative values from known ranges for composition, amounts, surface areas, and reaction rates of the various components of the MD waste packages. The input to EQ6

includes the composition of J-13 well water, a rate of influx to the waste package that corresponds to suitably chosen percolation rates into a drift, and a drip rate into the waste package, which is also the flow rate out of the waste package. In some cases, the degradation of the waste package is divided into stages (e.g., degradation of the DHLW glass before breach of the DOE SNF canister and exposure of the ingot material to the water). The EQ6 outputs include the compositions and amounts of solid products and the solution composition. The calculation process is presented in more detail in Reference 9.

Table 4.1 illustrates representative analysis cases that explore the different sequences of degradation: degrading the glass first and then the ingots, the ingots first and then the glass, or degrading the glass and ingots together.

Table 4.1 Cases Varying the Sequence of Degradation

md02_01	Maximum volume of ingots that fit in a DOE SNF canister with a minimum ingot liner thickness (1 mm). 2 stage run: Degrade glass and then expose ingots. High glass and drip rates, low ingot and SS rates.			
Reactant Fully Degraded	**Time (years)**	**pH**	**% Gd Remaining**	**% U Remaining**
Glass	14,839	8.81	100.00%	55.85%
Ingots	535,140	7.82	99.48%	22.91%
WP Liner	601,360	7.83	99.41%	19.53%
End	633,780	8.05	99.26%	17.94%
md02_02	Maximum volume of ingots that fit in a DOE can with a minimum ingot liner thickness (1 mm). 2 stage run: Degrade glass and then expose ingots. Stage 1: high glass and drip rates, low Ingot and SS rates. Stage 2: change to high stainless rate and low drip rate (causes a low pH in the second stage).			
Reactant Fully Degraded	**Time (years)**	**pH**	**% Gd Remaining**	**% U Remaining**
Glass	14,839	8.81	100.00%	55.85%
Ingots	16,721	7.02	100.00%	55.79%
WP Liner	44,545	5.32	99.59%	55.68%
End	634,170	7.90	99.54%	54.03%
md02_03	Maximum volume of ingots that fit in a DOE can with a minimum ingot liner thickness (1 mm). 2 stage run: Degrade ingots and then expose glass. Low glass rate, mean drip rate, high ingot and SS rates.			
Reactant Fully Degraded	**Time (years)**	**pH**	**% Gd Remaining**	**% U Remaining**
Ingots	1,506	5.44	77.35%	96.69%
WP Liner	30,091	5.75	77.21%	96.42%
Glass	229,650	8.68	77.21%	0.00%
End	633,820	8.07	77.06%	0.00%
md02_06	Maximum volume of ingots that fit in a DOE can with a minimum ingot liner thickness (1 mm). Ingots and glass degrade together, low glass rate, mean SS and drip rates, low ingot rate.			
Reactant Fully Degraded	**Time (years)**	**pH**	**% Gd Remaining**	**% U Remaining**
WP Liner	60,134	5.85	100.00%	99.97%
Glass	248,180	8.67	100.00%	29.17%
Ingots	519,930	8.07	99.62%	19.09%
End	633,800	8.07	99.37%	18.44%

The results of this illustrative table as well as most EQ6 runs show that in most of the cases more than 80% of the Gd will remain in the waste package, although the two-stage scenario exposing the MD ingots first and then the glass potentially loses more Gd.

Various computational assumptions were tested, in order to investigate how various sequences of degradation might proceed and affect the retention of gadolinium. Such geochemical sensitivity analyses investigating the formation $GdPO_4 \cdot H_2O$ and/or iron minerals identified a potential for severe, or complete, depletion of Gd. These calculations are discussed next.

4.4 Impact of Suppression of $GdPO_4 \cdot 10H_2O$ Formation

$GdPO_4 \cdot 10H_2O$, is the most likely Gd mineral to form, and was artificially suppressed in two cases, to study the sensitivity of results to the formation of $GdPO_4 \cdot 10H_2O$. The suppression of formation of a mineral is an option available in EQ6 software code. Although completely suppressing the formation of minerals most likely to form is extreme and unrealistic, this is useful method in investigating the sensitivity of Gd and U. Table 4.2 illustrates findings from sensitivity calculations performed.

Table 4.2 Cases Suppressing $GdPO_4 \cdot 10H_2O$ Formation

md02_03 Maximum volume of ingots that fit in a DOE can with a minimum ingot liner thickness (1 mm).
(base case) 2 stage run: Degrade ingots and then expose glass. Low glass rate, mean drip rate, high ingot and SS rates.

Reactant Fully Degraded	Time (years)	pH	% Gd Remaining	% U Remaining
Ingots	1,506	5.44	77.35%	96.69%
WP Liner	30,091	5.75	77.21%	96.42%
Glass	229,650	8.68	77.21%	0.00%
End	633,820	8.07	77.06%	0.00%

md02_03 Maximum volume of ingots that fit in a DOE can with a minimum ingot liner thickness (1 mm).
Suppress $GdPO_4 \cdot 10H_2O$ 2 stage run: Degrade ingots and then expose glass. Low glass rate, mean drip rate, high ingot and SS rates.

Reactant Fully Degraded	Time (years)	pH	% Gd Remaining	% U Remaining
Ingots	1,506	5.68	18.09%	96.71%
WP Liner	30,091	5.75	0.00%	96.44%
Glass	229,090	8.84	0.00%	0.07%
End	633,820	8.07	0.00%	0.00%

md02_06 Maximum volume of ingots that fit in a DOE can with a minimum ingot liner thickness (1 mm).
(base case) Ingots and glass degrade together, low glass rate, mean SS and drip rates, low ingot rate.

Reactant Fully Degraded	Time (years)	pH	% Gd Remaining	% U Remaining
WP Liner	60,134	5.85	100.00%	99.97%
Glass	248,180	8.67	100.00%	29.17%
Ingots	519,930	8.07	99.62%	19.09%
End	633,800	8.07	99.37%	18.44%

md02_06 Maximum volume of ingots that fit in a DOE can with a minimum ingot liner thickness (1 mm).
Suppress $GdPO_4 \cdot 10H_2O$ Ingots and glass degrade together, low glass rate, mean SS and drip rates, low ingot rate.

Reactant Fully Degraded	Time (years)	pH	% Gd Remaining	% U Remaining
WP Liner	60,134	5.85	88.49%	99.98%
Glass	248,220	8.83	84.24%	29.22%
Ingots	519,930	8.07	83.37%	18.98%
End	633,820	8.07	83.12%	18.33%

Because the formation of $GdPO_4 \cdot 10H_2O$ was suppressed, the mineral $GdOHCO_3$ forms instead of $GdPO_4 \cdot 10H_2O$. The conservative two-stage case md02_03 retains 77% of the initial Gd content with the formation of $GdPO_4 \cdot 10H_2O$. However, with $GdPO_4 \cdot 10H_2O$ formation suppressed, all Gd is predicted to be lost (md02-03). This case demonstrates a strong sensitivity of Gd retention to $GdPO_4 \cdot 10H_2O$ formation, but requires the unlikely event of the ingots degrading in the absence of the glass in addition to the complete suppression of $GdPO_4 \cdot 10H_2O$ formation. Thermodynamic data indicate that $GdPO_4 \cdot 10H_2O$ will form, and since it's formation is key to retaining Gd, retention of Gd should be further investigated. This can be accomplished by a detailed analysis of the experiments on which the data is based to show applicability to the current situation, by further analysis and/or experiments.

In another case (md02_06), even with the $GdPO_4 \cdot 10H_2O$ formation suppressed, 83% of the Gd remains, as compared to the 99% that remained in the case where $GdPO_4 \cdot 10H_2O$ was allowed to form, since $GdOHCO_3$ is less likely to form than $GdPO_4 \cdot 10H_2O$. The suppression of $GdPO_4 \cdot 10H_2O$ formation does not affect the percentage of U remaining in either case.[9]

4.5 Suppressing the Formation of Iron Minerals to Control Ionic Strength

Hematite and goethite are predicted to form in the waste package under normal running of EQ6. At early times in the EQ3/6 runs when the stainless steels are degrading, the pH is low and the ionic strength is high due to the presence of Ni^{++}, $Cr_2O_7^{--}$, and $HCrO_4^-$ ions in solution. If the most stable iron oxides (hematite and goethite) are suppressed in the EQ3/6 runs, then the more soluble $Fe(OH)_3$ forms. Allowing $Fe(OH)_3$ to form causes the pH to increase closer to neutral and the ionic strength to decrease to less than 1.0. For the purpose of investigating the sensitivity, the formation of these minerals was suppressed to determine if there is any effect on the results. Table 4.3 illustrates the results of suppressing the formation of various iron minerals.

The case md02_03 is one of the few cases with an ionic strength greater than 1.0, therefore this case was used to see the effect of suppressing the formation of iron minerals. The first case, fe02_03, suppresses the formation of goethite, hematite, and andradite for the first thousand years, which is the time period where the ionic strength was high in case md02_03. The results of case fe02_03 show that the time span with an ionic strength of more than 1.0 is reduced from 470 years to 300 years, although the actual ionic strength peak for this case is higher. The second case, fs02_03, suppresses the formation of goethite, hematite, and andradite, like case fe02_03, and suppresses the formation of $GdPO_4 \cdot 10H_2O$. The ionic strength did not increase above 1.0 for this case. The third case, he02_03, suppresses the formation only for hematite. This case has a higher ionic strength peak than the original case. Case al02_03 suppressed the aluminum minerals diaspore and gibbsite to determine the sensitivity of the model on aluminum mineral production. Again, the effect of suppressing $GdPO_4 \cdot 10H_2O$ becomes evident.

The percentage of Gd remaining for these cases varies considerably. When the formation of the three iron minerals is suppressed in case fe02_03, the pH is lower from 1,000 to 3,000 years, therefore more of the Gd is washed out of the waste package during that time. The case that also suppresses $GdPO_4 \cdot 10H_2O$ formation (case fs02_03) loses all the Gd, but it is unlikely that $GdPO_4 \cdot 10H_2O$ will not form.[9] Case he02_03 retains more Gd because the pH is slightly higher from 1,000 to 1,200 years, as the ingots finish degrading, and that higher pH allows more $GdPO_4 \cdot 10H_2O$ to form. Results from the cases where hematite (case he02_03) or the aluminum minerals, diaspore and gibbsite, (case al02_03) are suppressed indicate only a slight change in Gd retention from that of the base case (case md02_03). The amount of U that remains in the waste package is unaffected by the suppression of any of the minerals considered in Table 4.3.

Table 4.3 Cases Suppressing the Formation of Various Minerals

md02_03 (base case) Maximum volume of ingots that fit in a DOE can with a minimum ingot liner thickness (1 mm). 2 stage run: Degrade ingots and then expose glass. Low glass rate, mean drip rate, high ingot and SS rates.

Reactant Fully Degraded	Time (years)	pH	% Gd Remaining	% U Remaining	% Al Remaining
Ingots	1,506	5.44	77.35%	96.69%	99.75%
WP Liner	30,091	5.75	77.21%	96.42%	99.74%
Glass	229,650	8.68	77.21%	0.00%	99.74%
End	633,820	8.07	77.06%	0.00%	99.74%

fe02_03 Maximum volume of ingots that fit in a DOE can with a minimum ingot liner thickness (1 mm). 2 stage run: Degrade ingots and then expose glass. Low glass rate, mean drip rate, high ingot and SS rates. Suppressed the formation of Fe minerals for 1st 1,000 years (andradite, goethite, and hematite).

Reactant Fully Degraded	Time (years)	pH	% Gd Remaining	% U Remaining
Ingots	1,506	5.03	60.79%	98.14%
WP Liner	30,091	5.75	60.23%	97.81%
Glass	229,220	8.84	60.23%	0.07%
End	633,820	8.07	60.09%	0.00%

fs02_03 Maximum volume of ingots that fit in a DOE can with a minimum ingot liner thickness (1 mm). 2 stage run: Degrade ingots and then expose glass. Low glass rate, mean drip rate, high ingot and SS rates. Suppressed the formation of Fe minerals for 1st 150 years and suppressed the formation of $GdPO_4 \cdot 10H_2O$.

Reactant Fully Degraded	Time (years)	pH	% Gd Remaining	% U Remaining
Ingots	1,506	5.68	18.09	96.49
WP Liner	30,091	5.75	0.00	96.22
Glass	229,030	8.84	0.00	0.07
End	443,540	8.26	0.00	0.00

he02_03 Maximum volume of ingots that fit in a DOE can with a minimum ingot liner thickness (1 mm). 2 stage run: Degrade ingots and then expose glass. Low glass rate, mean drip rate, high ingot and SS rates. Suppressed the formation of Hematite for the whole run.

Reactant Fully Degraded	Time (years)	pH	% Gd Remaining	% U Remaining
Ingots	1,506	5.21	78.12	96.76
WP Liner	30,090	5.27	75.07	95.87
Glass	276,660	8.27	74.92	0.00
End	633,820	8.12	74.54	0.00

al02_03 Maximum volume of ingots that fit in a DOE can with a minimum ingot liner thickness (1 mm). 2 stage run: Degrade ingots and then expose glass. Low glass rate, mean drip rate, high ingot and SS rates. Suppressed the formation of Al minerals Diaspore and Gibbsite

Reactant Fully Degraded	Time (years)	pH	% Gd Remaining	% U Remaining	% Al Remaining
Ingots	1,506	5.5857	76.84%	97.12%	99.67%
WP Liner	30,092	5.7452	76.72%	96.89%	99.64%
Glass	236,080	8.9339	76.69%	0.07%	99.62%
End	633,820	8.0771	76.52%	0.00%	99.62%

4.6 Geochemistry Analysis Conclusions

Most of the cases show that more than 80% of the initial Gd content will remain in the waste package, although the two-stage scenario exposing the MD ingots first and then the glass potentially loses more Gd. This case uses unrealistic input conditions, i.e. requires the unlikely event of the ingots degrading while isolated from the DHLW glass, in addition to the complete suppression of $GdPO_4 \cdot 10H_2O$ formation. Since thermodynamic data indicate that $GdPO_4 \cdot 10H_2O$ will form, and because its formation is key to retaining Gd, retention of Gd should be further demonstrated. This can be accomplished by further analysis of the experiments on which the data is based to show applicability to the current situation and/or experiments.

5.0 SHIELDING ANALYSIS

5.1 Computer Software

The Monte Carlo particle transport code, MCNP, Version 4B2LV (CRWMS M&O 1998f), is used to calculate average dose rates at the external surfaces of the waste package. The information regarding the code and its use for the shielding analysis is documented in BSC (2001b).

The Monte Carlo method for solving the integral radiation transport equation, which is implemented in the MCNP computer program, is used to calculate radiation dose rates for the waste packages. MCNP uses continuous-energy cross sections processed from the evaluated nuclear data files ENDF/B-V.[13] These cross-section libraries are part of the qualified MCNP code. The flux averaged over a surface tally is specified in calculations and the neutron and gamma flux-to-dose rate conversion factors[14] are applied to obtain surface dose rates.

5.2 Calculations & Results

Reference 5 specifies the design criteria for the maximum rate of 1,450 rem/hr on all external surface of the WP. The geometric representation of the waste package used in MCNP calculations is shown in Figure 5.1. The waste package contains two different radiation sources, which are volumetric sources uniformly distributed inside the cavity of the DOE SNF canister and the glass volume, respectively. A conservative approach is used, in which lower material densities for the SRS DHLW glass and the MD ingots are employed.

In the calculation, the external surfaces of the waste package are divided in segments and the dose rate is averaged over each segment to evaluate the spatial distribution of the dose rate. Figure 5.2 and Figure 5.3 show the segments of the radial and axial segments used in the dose-rate calculations. The radial surface, between the bottom and top planes of DHLW glass, is equally divided into five segments, each of which is 47.886-cm high. The first radial segment (Segment 1), 64.57-cm high, corresponds to the empty portion of the DHLW canister, which is between the top of the waste package cavity and the top of the DHLW glass. The waste package top and bottom axial surfaces are divided into two radial segments of 0-30 cm (Segment 7) and 30-101.5 cm (Segment 8). For this waste package, the DOE canister is positioned in the center of the waste package and gamma source intensity of the MD ingots is twenty times the gamma source intensity of each individual SRS DHLW glass canister. Because the DHLW glass canisters are positioned near the disposal container, they attenuate the radiation emitted by the MD SNF and mostly determine the dose rates on the angular segments adjacent to them (Segments B). However, due to their higher source intensity, the MD ingots contribute to the dose rates averaged over Segments A. Therefore, an angular dependence of the waste package radial dose is expected and the radial surface is divided into ten equal angular segments, as shown in Figure 5.3.

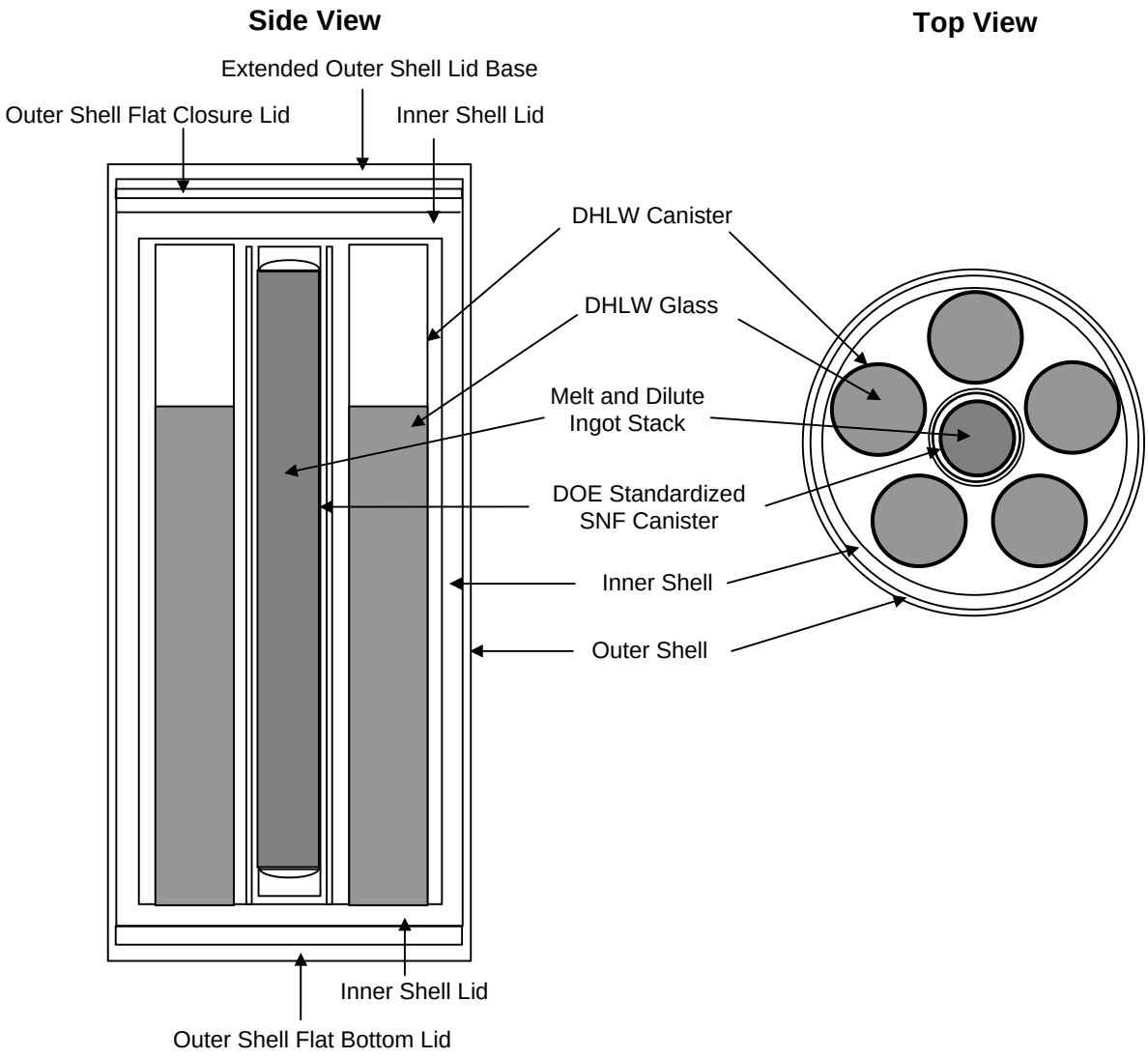

Figure 5.1 Vertical and Horizontal Cross Sections of MCNP Geometry Representation

Table 5.1 and Table 5.2 provide radial and axial dose rates on the outer surface of the waste package containing the five SRS DHLW glass canisters and the DOE SNF canister. The neutron source has an insignificant contribution to the total dose and the gamma dose dominates the total dose.

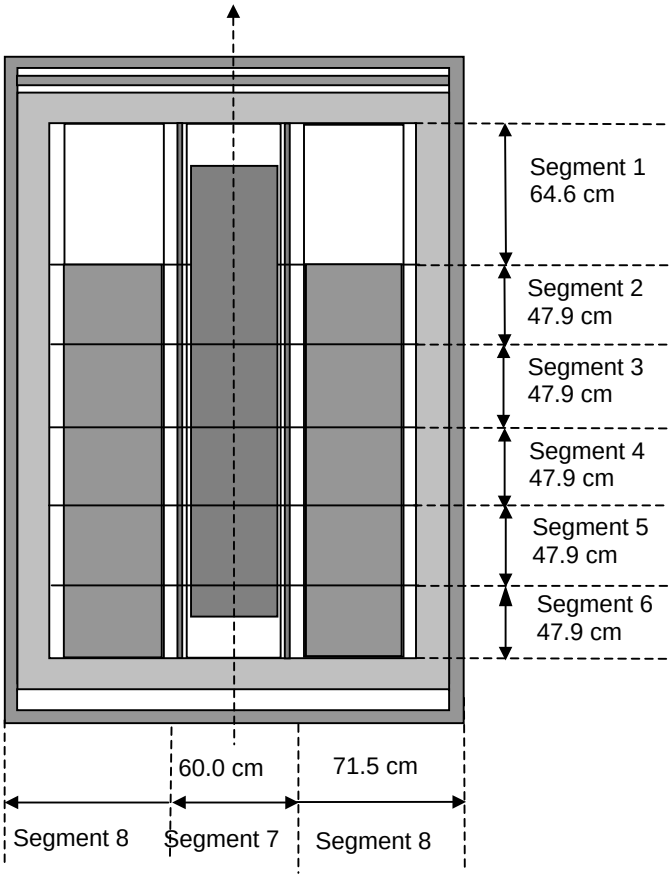

Figure 5.2 Surfaces and Segments (axial and radial) Used for Dose Rate Calculations

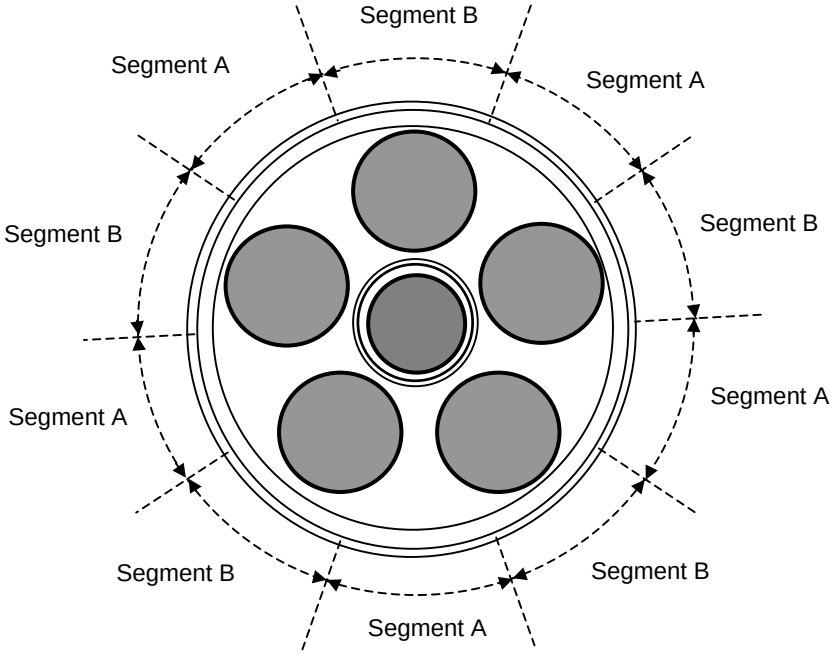

Figure 5.3 Angular Segments of the WP Outer Radial Surface Used in Dose Rate Calculations

Table 5.1 Dose Rates Averaged over Axial and Radial Segments of the WP Outer-Radial and Axial Surfaces[5]

Location	Gamma Dose Rate (rem/h)	Neutron Dose Rate (rem/h)	Total Dose Rate (rem/h)
Radial surface: Segment 1	85.47	0.14	85.61
Radial surface: Segment 2	133.53	0.14	133.67
Radial surface: Segment 3	144.49	0.15	144.64
Radial surface: Segment 4	143.34	0.15	143.49
Radial surface: Segment 5	136.42	0.14	136.57
Radial surface: Segment 6	105.20	0.11	105.31
Bottom surface: Segment 7	47.50	0.22	47.71
Bottom surface: Segment 8	13.76	0.08	13.84
Top surface: Segment 7	27.30	0.15	27.45
Top surface: Segment 8	4.82	0.08	4.89

NOTE: The dose rates listed in this table are the upper limits of the 95 percent confidence intervals of the Monte Carlo dose rate calculations.

The radial surface dose rates have an angular dependence, as shown in Table 5.2. The dose rate averaged over Segment A is approximately twice as much as the dose rate averaged over Segment B.

Table 5.2 Dose Rates Averaged Over Angular Segments of the WP Outer-Radial Surface

	Angular Segment A			Angular Segment B		
Axial Location	Gamma Dose Rate (rem/h)	Neutron Dose Rate (rem/h)	Total Dose Rate (rem/h)	Gamma Dose Rate (rem/h)	Neutron Dose Rate (rem/h)	Total Dose Rate (rem/h)
Segment 1	104.33	0.16	104.49	74.42	0.14	74.56
Segment 2	182.66	0.19	182.85	98.68	0.11	98.79
Segment 3	199.69	0.20	199.89	103.21	0.11	103.32
Segment 4	199.13	0.20	199.33	101.38	0.10	101.48
Segment 5	185.52	0.20	185.72	100.46	0.11	100.57
Segment 6	132.09	0.14	132.23	92.80	0.08	92.88

NOTE: The dose rates listed in this table are the upper limits of the 95 percent confidence intervals of the Monte Carlo dose rate calculations.

5.3 Shielding Analysis Summary

The maximum dose rate at the external surfaces of the waste package occurs on the radial surface and is 199.89 rem/h. The radial dose rate shows an angular distribution, with dose rates on Segments A being approximately twice as much as those on Segments B. The dose rates on the bottom and top surfaces of the waste package are about one-third and about one-fifth, respectively, of the maximum dose rate on the outer radial surface. The design criterion specifies that the maximum dose rate at all external surfaces of the waste package is 1,450 rem/h.[5] The dose rates in rem/h and rad/h are practically the same due to the insignificant contribution of the neutron dose rate to the total dose rate.

Radiation produces radiolytic species (e.g., hydrogen peroxide and nitric acid) that may enhance the corrosion of the waste package components. A study that has evaluated the effect of radiation on the corrosion of the material used for the fabrication of waste packages in the environments expected at Yucca Mountain[15] showed that a dose rate of 10^4 rad/h is required before any influence of radiation is observed on copper/nickel alloys. Since the calculated dose rate at the external surface of the MD waste package is approximately 200 rem/h, it is expected that no observable effect on the corrosion of waste package materials will be present.

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