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Melt-Dilute Form of Al-Based Spent Nuclear Fuel Disposal Criticality Summary Report

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MELT-DILUTE FORM OF Al-BASED SPENT NUCLEAR FUEL DISPOSAL CRITICALITY SUMMARY REPORT

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ABSTRACT

Criticality analysis of the proposed melt-dilute (MD) form of aluminum-based spent nuclear fuel (SNF), under geologic repository conditions, was performed [1] following the methodology documented in the *Disposal Criticality Analysis Methodology Topical Report* [2]. This methodology evaluates the potential for nuclear criticality for a waste form in a waste package. 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 readily demonstrated for the MD form of aluminum-based SNF.

INTRODUCTION

The 5-DHLW/DOE SNF waste package for repository disposal of the melt-dilute form of aluminum-based SNF 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. Criticality analysis of the proposed MD-SNF form, under geologic repository conditions, was performed [1] following the methodology documented in the Disposal Criticality Analysis Methodology Topical Report [2]. 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 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. There are three primary paths or degradation scenarios that encompass the range of potential events (see Fig. 1). Consideration is further given to the evolution of the geochemical environment within the waste package and the effect of the geochemical changes on the degradation characteristics of the waste package components including the spent fuel and the package internals. The integrated analysis results in configurations that are evaluated for criticality with the goal of maintaining k_{eff} less than 0.93. The goal for k_{eff} less than 0.93 has been set to allow for a 5% safety margin and uncertainty in the experiments used to validate the method to calculate k_{eff}. This paper provides a summary of the criticality analysis and supporting analyses in evaluating the MD-SNF form for criticality during the post-closure period of the Monitored Geologic Repository (MGR).

SOURCE TERM FOR THE CRITICALITY ANALYSIS

The DOE standardized canister that will be placed in the center of the codisposal WP will contain three to six MD ingots that are both homogenous and monolithic, depending on the dimensions of the individual ingots. These ingots will range in height from 15 to 30 inches and will likely be contained in a plain carbon steel crucible liner. The crucible liner will be standardized at approximately 20 to 30 inches. The maximum outer diameter of the crucible liner will be 15.5-16.5 inches and it will have a thickness of up to 0.5 inches. Due to uncertainty in the presence of a crucible liner, the criticality analysis considered a range of crucible liner thickness from 0.0 inches (i.e., no liner) to 0.5 inches. The outer diameter of the MD-SNF form will be dictated by the crucible dimensions.

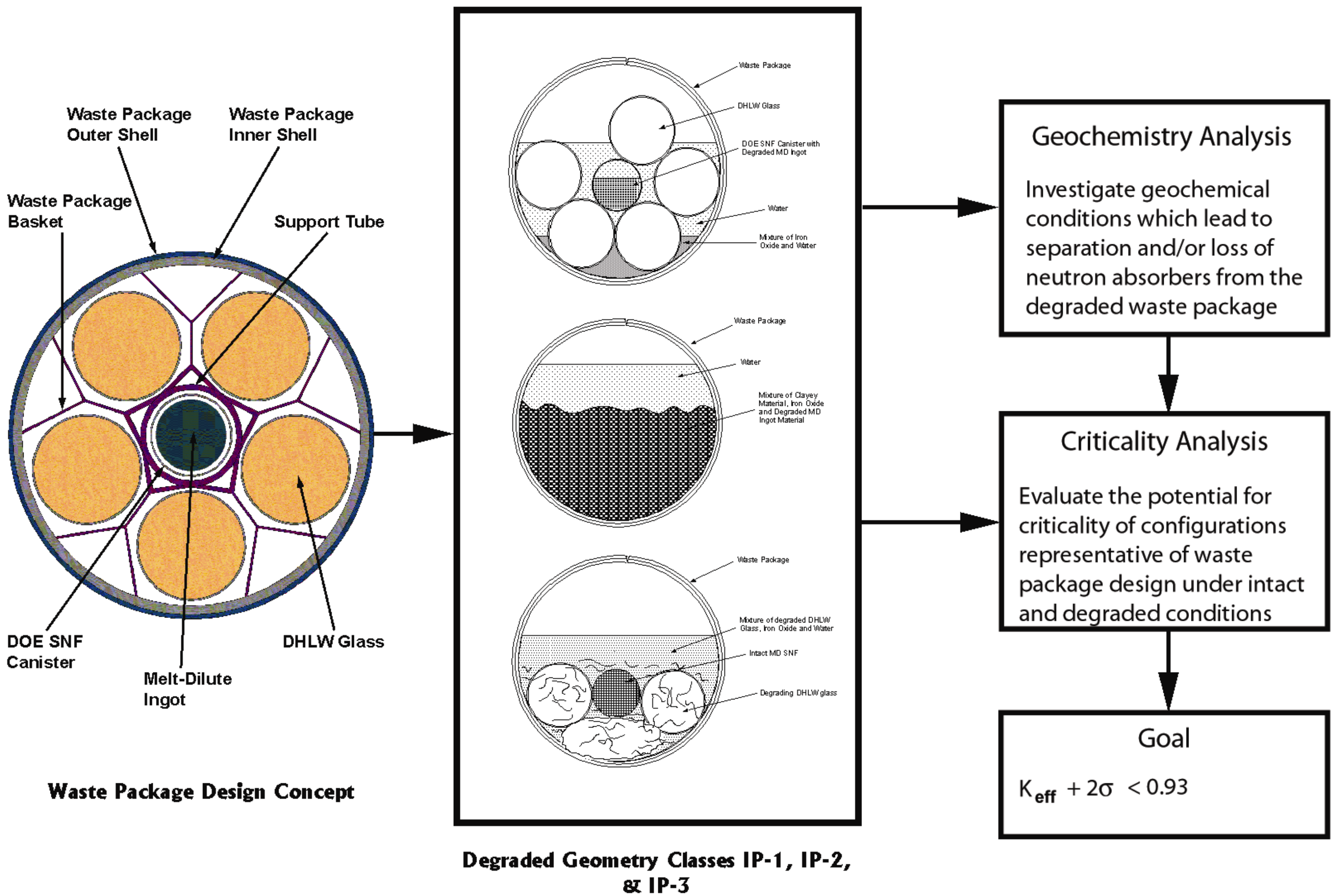

Figure 1 Criticality Analysis Logic

The melt-dilute ingot will have a density of approximately 3 g/cm^3 and a porosity of 5 to 10 percent. Two general ingot compositions have been considered in the criticality and geochemistry analyses. The first composition is 13.2 ± 5 percent by weight uranium, at 20% ^{235}U and 0.5 percent gadolinium metal by weight, with the balance of the ingot being aluminum. The second composition is 13.2 ± 5 percent by weight uranium, at 20% ^{235}U, 0.5 percent gadolinium metal by weight, and 2.5 percent hafnium metal by weight, with the balance of the ingot being aluminum.

CODISPOSAL WASTE PACKAGE DEGRADATION ANALYSIS

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* [2]. 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.

Application of Standard Scenarios to Melt-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 Ref. [3]) 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 Fig. 2) involve the MD ingots degrading before other internal components and depends on the degradation rates of the various materials that make up the other internal components (OICs) as compared to the degradation rate of the ingots. The degradation rates show that the ingot high rate is 4.8×10^{-12} $mol \cdot cm^{-2} \cdot s^{-1}$ while the low rate of the SS components is 2.5×10^{-14} $mol \cdot cm^{-2} \cdot s^{-1}$. Carbon steel has a degradation rate of 1.8×10^{-11} $mol \cdot cm^{-2}$ 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.

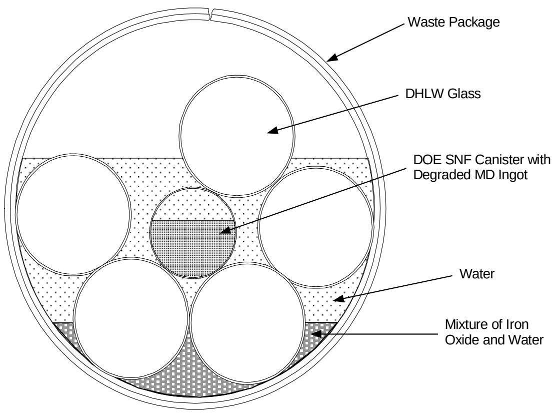

Figure 2 Conceptual Sketch of WP for Degradation Scenario IP-1

IP-2: In the configurations resulting from IP-2 scenario (see Fig. 3), 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. 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 for 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.

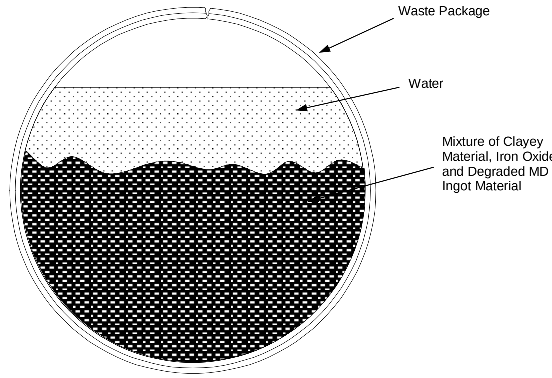

Figure 3 Conceptual Sketch of WP for Degradation Scenario IP-2

IP-3: The configurations resulting from IP-3 scenario (see Fig. 4) 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 into solution and is flushed out of the WP while the fissile material is in a geometry that is favorable for 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.

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

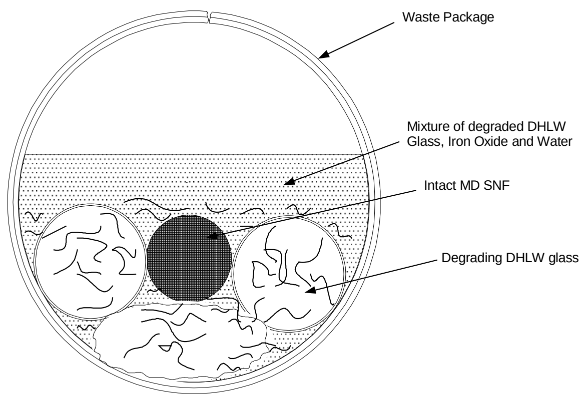

Figure 4 Conceptual Sketch of WP for Degradation Scenario IP-3

MOST PROBABLE DEGRADATION PATH

Based on the corrosion rates and the material thickness used in these analyses, 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.

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.

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. 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.

   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.

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.

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.

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.

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.

The degradation rates of each of the individual components of the melt-dilute codisposal waste package are varied within the analyses in consideration of the range of environmental conditions expected within the repository. Review of the corrosion rates of the components it is evident 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 lowest corrosion rate for the melt-dilute ingot is assumed equal to that given for U-Al fuel types.

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 high-level waste glass pour canisters; 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.

RESULTS AND CONCLUSION

The criticality analyses considered all aspects of intact and degraded configurations of the codisposal waste package containing MD 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.

Acknowledgments

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