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**EVALUATION OF THE U.S. DEPARTMENT
OF ENERGY ALUMINUM-BASED SPENT
FUEL CRITICALITY ANALYSIS**

Prepared for

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QUALITY OF DATA, ANALYSES, AND CODE DEVELOPMENT

No independent data or codes were used for evaluations performed in this report.

1 INTRODUCTION

Aluminum (Al)-based research reactor fuel is a small part (less than 1 percent by volume) of the total inventory of spent nuclear fuel (SNF) and high-level waste (HLW) to be disposed in a geologic repository. However, disposition of these fuels, including treatment, temporary storage, and permanent disposal, presents a significant challenge because of the diverse geometry, high enrichments (20 to greater than 90 percent), and the complex metallurgical characteristics of the fuel. A total of 255 m³ (62.4 metric tons of heavy metal) of Al-based SNF is anticipated to be sent to the Savannah River Site for processing by the year 2035. The U.S. Department of Energy (DOE) has conducted a parallel development of two treatment technologies: (i) direct disposal (or co-disposal) in a repository, and (ii) melt and dilution with depleted uranium followed by disposal. The development of these two technologies is termed the Alternate Technology Program (ATP).

In fiscal year (FY) 1998, Center for Nuclear Waste Regulatory Analyses (CNWRA) conducted a topical review of documents related to the permanent disposal of Al-based fuels treated according to direct co-disposal and melt and dilute options. Based on this review and subsequent discussions with the DOE during FY98, it was agreed to continue the review of additional documents supplied by the DOE. Based on a recent assessment of the relative merits of direct, co-disposal and melt and dilute technologies, the DOE has decided to focus the ATP on validation of melt and dilute treatment and waste form performance. Therefore, the CNWRA review in FY99 will focus primarily on repository performance issues related to melt and dilute waste form. However, as part of its activities related to criticality analysis of the direct co-disposal fuels, the DOE performed criticality calculations considering the degradation of canisters and other components of the co-disposal waste package (WP) (Civilian Radioactive Waste Management System, 1998a). This report provides an evaluation of the DOE criticality calculations and delineates further information needs. However, it is recommended that future evaluation of the DOE reports be combined with the review of melt and dilute waste form in a topical review report.

2 CRITICALITY CALCULATIONS

2.1 U.S. DEPARTMENT OF ENERGY CALCULATIONS

The evaluation of the potential for criticality of Al-based SNF that is disposed with HLW glass in a co-disposal canister is divided into three phases. The first phase evaluated criticality events within the canister containing only SNF. The second phase of the analysis assesses criticality for the degradation of both the HLW glass and the SNF within the co-disposal canister. Finally, phase three will determine whether criticality is possible outside of the WP. The documents reviewed here pertain to phase two of the project, criticality of degraded HLW glass and SNF within the co-disposal package.

The reactivity of the SNF canister was analyzed with the MCNP4A computer code using the ENDF-B/V cross section library. A series of benchmark runs of the code were conducted, and the results were compared to reviewed experiments. The worst experimental uncertainty was 0.015 k_{eff} . To be conservative, the bias value was set to 0.02 k_{eff} . For a given scenario to be considered subcritical, the sum of k_{eff} plus two standard deviations plus the bias value must be less than 0.95.

After performing some preliminary calculations to determine the likely performance of the container materials and possible separation mechanisms for the uranium and neutron absorber, the analysis divided the criticality calculations into three possible degradation scenarios. The first scenario was the degraded SNF contained within an intact DOE SNF canister. The second was the degraded SNF in a layer on top of the degraded HLW. The third scenario evaluated degraded SNF in a layer on the bottom of the co-disposal canister mixed with degraded HLW and covered with degraded HLW. These scenarios were evaluated for both the Massachusetts Institute of Technology (MIT) fuel enriched to 93.5 percent U-235 and the Operations Readiness Review (ORR) fuel enriched to 20 percent U-235. However, in all cases, the MIT fuel was more reactive than the ORR fuel, so this review will focus on the criticality analyses for the MIT fuel.

Several assumptions are made throughout the criticality calculations, which the DOE states are conservative. Assumption 4.3.1 states that the co-disposal canister is fully loaded with fuel, and assumption 4.3.2 indicates that no credit is taken for burnup. Assumption 4.3.4 states that all spaces within the canister that are not occupied by fuel or structural materials are assumed to be fully flooded with water. Assumption 4.3.12 states that the WP is assumed to be horizontal for estimating the fraction of neutronically significant material that could fall to the bottom of the SNF canister. This leads to a single collection of material at the bottom of the canister instead of several smaller collections in the corners between angled plates. Finally, although not stated explicitly in the assumptions section, the criticality analyses assume that the WP is surrounded by water.

2.2 NUCLEAR REGULATORY COMMISSION EVALUATION

The DOE criticality evaluation is reasonable for the scenarios being modeled. The computer code and cross section library used for the analyses are appropriate for the situations being modeled. However, it is not clear that all of the assumptions in the analysis are appropriate or conservative. Assumption 4.3.12 is confusing because it indicates that modeling the WP as horizontal is a conservative assumption, but it goes on to state that the collection of fissile material in several piles in the corners where plates meet (which would occur when the package is tilted) is a more reactive geometry than a single collection at the bottom (which would occur when the package was horizontal). If this statement is correct, the analysis should be

extended to determine if the collection of fissile material in smaller piles could lead to a criticality event. Also, the assumption that the canister is surrounded by water may not be the most conservative choice of reflectors for the system. Rockfall covering the package is a realistic scenario and may lead to a larger k_{eff} for the waste package than a water moderator because of its higher density. The DOE should evaluate the potential effects of different reflecting materials that may surround the waste package and should select the most conservative material for their analyses. More detailed review is provided below.

2.3 DEGRADED SPENT NUCLEAR FUEL CONTAINED WITHIN INTACT U.S. DEPARTMENT OF ENERGY SPENT NUCLEAR FUEL CANISTER

2.3.1 U.S. Department of Energy Calculations

A variety of possible SNF and HLW configurations were analyzed within this scenario to determine the most reactive condition. The DOE SNF canister was modeled as being in one of the following locations and states of degradation: (i) situated among five intact HLW canisters, (ii) lying on the bottom of the co-disposal container surrounded by the degraded remnants of the HLW glass, or (iii) situated among five degraded HLW canisters with the largest separation of the fuel and neutron absorber within the SNF canister. The degraded DOE SNF is homogenized within a basket position or within the canister depending on the degree of degradation of the basket. Variations of the conditions outside the DOE SNF canister are simulated to demonstrate conservatism. The basket is modeled at various stages of degradation and with stainless steel or carbon steel as the material of fabrication.

For the first set of conditions, it was assumed that all free space within the co-disposal container and all canisters was filled with water. The second set of conditions was modeled with a clay water content of 25 percent, which nearly fills the WP and a fully flooded SNF canister. The third set of conditions is modeled as a fully flooded canister. The calculations indicate that 1.00 kg of Gd_2PO_4 is required to keep the material sufficiently subcritical under these conditions when stainless steel is used as the basket material and 1.25 kg is required when carbon steel is used.

2.3.2 Nuclear Regulatory Commission Evaluation

These calculations appear to be reasonably conservative, but the DOE should address the following issues:

- (1) The analyses assume that the degraded spent fuel and clay material can absorb only enough water to fill the co-disposal canister. However, the document does not analyze the potential of holes in the canister allowing some of the degraded HLW clay and oxidated steel to leak out of the canister, and later being plugged up with clay to allow the package to fill with water. This scenario could allow the water content of the clay and other materials to increase above the amount currently assumed as maximum and possibly increase the reactivity of the package.
- (2) The analyses show that 1.00 kg of Gd_2PO_4 is required to keep a stainless steel package sufficiently subcritical and 1.25 kg of Gd_2PO_4 is required to keep a carbon steel package sufficiently subcritical. However, the analyses do not indicate how the DOE will ensure content and uniform coverage of the neutron absorbing material. The DOE should indicate

whether credit for the neutron absorbing material will be limited to 75 percent of the original material or that the uniform coverage of the material will be verified by measurement.

- (3) Concerning the maximum separation of fuel and absorber, there are several issues that are unclear. First, it is not clear how the maximum percentage of uranium at the bottom of the canister, as shown in table 6.4-2, column 4, was calculated. Second, the footnote to that table indicates that analyses were performed to show that, for the minimum Gd at the bottom, the remaining distributed Gd will be sufficient to prevent criticality with all the uranium distributed. However, it does not address whether the package will be subcritical with the maximum percentage of Gd at the bottom of the package (assume 100 percent without any additional information) and all of the uranium distributed throughout the canister. These issues should be addressed to ensure that separation of fuel and absorber will not lead to a criticality event.

2.4 DEGRADED MASSACHUSETTS INSTITUTE OF TECHNOLOGY SPENT NUCLEAR FUEL ON TOP OF DEGRADED HIGH-LEVEL WASTE

2.4.1 U.S. Department of Energy Calculations

This scenario consists of a layer of hydrated oxides representing the degraded remnants of the DOE SNF canister and contents above a volume of clayey material from the degradation of HLW glass. This configuration is based on the degradation of the DOE SNF canister while resting on the surface of the clayey material. The optimum water content of the layer of oxides for neutron moderation was determined for this scenario to be 0.83 (i.e., a solids density of 17 percent). The results of these analyses showed that the quantity of Gd_2PO_4 required in the first scenario would be sufficient to maintain the system subcritical in this scenario.

2.4.2 Nuclear Regulatory Commission Evaluation

This analysis appears to be complete and reasonably conservative. There were no issues identified for this part of the evaluation.

2.5 DEGRADED MASSACHUSETTS INSTITUTE OF TECHNOLOGY SPENT NUCLEAR FUEL MIXED WITH DEGRADED HIGH-LEVEL WASTE CLAY

2.5.1 U.S. Department of Energy Calculations

This scenario consists of various fractions of the HLW clayey material mixed with the degraded DOE SNF accumulated starting in a canister-sized volume below the unmixed fraction of HLW and proceeding until the DOE SNF is mixed with the HLW clayey material in the layer just covering the canister. This configuration is based on the degradation of the DOE SNF canister surrounded by the HLW clayey material. The water fraction is kept consistent in the bottom fuel/clay mixture and the HLW clayey material on the top because it is not credible to have a less dense mixture at the bottom. The fraction of clay into which degraded fuel was mixed was varied along with the water fraction in the clay to determine the most

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reactive composition. The clay fraction was varied from 0 percent to 100 percent, while the water fraction was varied up to 0.272, which caused the clay to fill the container. The results of these analyses indicated that the system would remain subcritical with the quantity of Gd_2PO_4 that was determined to be necessary in the first scenario.

2.5.2 Nuclear Regulatory Commission Evaluation

Similar to the first scenario, this scenario does not evaluate the potential for loss of clay material out of the package, which could allow the water content of the clay and SNF layer to increase beyond the maximum evaluated in the analysis. This scenario has the potential to increase the reactivity of the system and should be addressed.

2.6 GEOCHEMICAL MODELING

2.6.1 U.S. Department of Energy Calculations

Studies documented in the subject report use geochemical modeling to determine compositions of materials remaining in the co-disposal WP and identify particular configurations affecting criticality control. More specifically, modeling is aimed at predicting the fate of fissile uranium and neutron absorbers as a result of aqueous degradation of materials as a function of time. The geochemical models are also important to criticality considerations in how they affect the solid phase assemblage because the physical arrangement of waste components and degradation products affects criticality calculations. For example, the report states in section 6.1 that the hydrated clay resulting from HLW glass corrosion "serves as the primary moderator."

The approach is to use the EQ6 geochemical reaction path code (Wolery, 1992) to simulate water reacting with and degrading engineered materials in the WP. Amounts in solution and in solids of U, B, and Gd are emphasized in analyzing the results as they apply to criticality potential. A number of assumptions are made regarding the water chemistry, corrosion rates, and design parameters. It is assumed that the J-13 well water composition is representative of the water reacting with the fuel and other materials. The corrosion rates assumed (table 4.1.6-1 of Civilian Radioactive Waste Management System, 1998a) for the corrosion-resistant materials correspond generally to the passive behavior of these alloys (of the order of 0.01 to 0.8 $\mu\text{m}/\text{yr}$). The corrosion rate for Alloy C-22 was assumed to be $8.12 \times 10^{-6} \mu\text{m}/\text{yr}$. It is also assumed that the dissolution rate of the fuel matrix is much faster than that of other WP components.

The DOE assumes that water dripping on the WP penetrates the overpacks and the HLW glass canisters, which are codisposed with the Al-clad SNF canister. Degradation of glass is assumed to generate clay forming minerals, which fill up the HLW canisters and generate a neutral pH. At this point, several degradation sequences are considered leading to two final configurations: (i) U compounds collect at the bottom surrounded by a clay layer and (ii) U compounds concentrate on top of a clay layer. The first configuration with a hydrated clay surrounding the U layer was considered to be the most conservative by the DOE.

A key conclusion of the geochemical modeling is that Gd_2PO_4 phosphate should be used as a neutron absorber because it is a low-solubility solid phase over the range of chemical conditions predicted (i.e., it will not allow significant transport of Gd out of the WP). This review, therefore, focuses on aspects of modeling affecting Gd fate during and after waste degradation.

2.6.2 Nuclear Regulatory Commission Evaluation

In general, the approach is valid and the modeling is well-posed. Uncertainties in scenarios as a result of input and conceptual uncertainties are considered in determining which factors are most important to making a conservative judgment of criticality potential. For example, the investigators show that the high pH conditions resulting from HLW glass dissolution would be favorable in that fissile U would tend to be mobilized and escape the WP, but they reasonably conclude that this scenario is too uncertain to be relied upon for criticality control. Likewise, the potential for B to be solubilized and removed from the WP is judged to be probable enough that B is not considered viable as a neutron absorber in the WP. Therefore, this review is not specifically concerned with the modeling as it affects U and B because the report takes no credit for their favorable disposition with respect to criticality. However, the stability of Gd_2PO_4 is important for maintaining subcriticality. Hence model assumptions in the calculation of Gd_2PO_4 stability are examined in detail.

There is no physical basis for the assumption that Alloy C-22 corrodes at a rate of almost three orders of magnitude lower than that of Alloy 625. While Alloy C-22 is expected to be more corrosion resistant than Alloy 625 under repository environmental conditions, under the assumption used in criticality calculations that Alloy 625 is a passive material, the corrosion rates of both these alloys should be similar. Both these alloys are protected by a passive film that limits the corrosion rate to about 10^{-2} $\mu\text{m}/\text{y}$ (current density of 1×10^{-8} A/cm^2). This value is similar to those observed experimentally for passive corrosion rates for these alloys. While the DOE has not considered Alloy C-22 in their criticality calculations, the low corrosion rates of 8.12×10^{-6} $\mu\text{m}/\text{y}$ assumed for Alloy C-22 may be nonconservative in future calculations.

The chemistry of the water is assumed to be that of J-13 water. However, information regarding the starting pH is not presented explicitly with other water chemistry data in table 4.1.5-1. The carbon molality reported in table 4.1.5-1 should be justified. Other sources (Ogard and Kerrisk, 1984) report considerably higher J-13 bicarbonate molalities (which should be less than or equal to carbon molality): around 2×10^{-3} versus the 1.45×10^{-4} value used here. Carbonate species are critical to water chemistry and speciation of many constituents (e.g., U and Gd). It is suggested in section 4.1.5 that a range of water compositions need not be considered because of the wide range encountered at different stages of modeling. However, it is possible that initial parameters such as pH could determine reaction paths in ways that are not addressed merely by recognizing that a wide range of chemistries result from the modeling. Some assessment of the sensitivity of model results to initial conditions is warranted. For example, radiolysis of water may create locally acidic conditions. Also, acidic conditions may be created locally by the hydrolysis of dissolved cationic species such as Fe^{2+} , Al^{3+} , and Cr^{3+} . Such acidic pH is well recognized in localized corrosion studies (Smialowska, 1986).

Recent DOE modeling of the near-field (Hardin, 1998) predicts that temperatures of close to 100°C may persist at the repository horizon 5,000 yr after closure. This may have profound effects on degradation processes and rates. Furthermore, high temperatures would affect water chemistry [e.g., see composition of J-13 equilibrated with tuff at 90°C in Wronkiewicz et al. (1992)]. Hence the effect of higher than ambient temperatures on the chemistry of the internal environment should be considered.

In the figures on pages 25–33, clay is often shown filling large volumes of the WP. This physical configuration appears to be important to criticality calculations because of the role of hydrated clay as a moderator. These large volumes of degradation products should be justified by modeling results (or on the basis of conservativeness), as should the physical process for their dispersal through WP voids. In this regard,

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it may be helpful also to provide more specific information on the degradation product minerals involved. The standard database typically utilized in EQ3/6 version 7.2b (data0.com.R2) does not include materials such as waste glass and steel alloys. The investigators should explain how they incorporated degradation of such materials into EQ6 reaction path models. Other details of modeling that need to be provided include

- External imposition of gases, such as O₂ and CO₂, in EQ6. For example, without external imposition of oxygen fugacity, it is possible that oxidation of U and other metals during dissolution will consume all O₂ and dissolution will cease.
- Water replenishment in successive EQ6 runs. In section 5.2, brief mention is made of routines employed for replenishing water between successive EQ6 runs. To interpret the EQ6 results, it is important to fully understand how these routines were applied. For example, how often were they applied (i.e., how much time would pass in an individual EQ6 run before it was terminated)?
- Gd removal rate does not appear to scale directly with the drip rate. In section 6.3.4.2, it is calculated that, with an aqueous Gd concentration controlled by Gd₂PO₄ and a drip rate of 10 mm/yr, it would take 786,000 yr to remove only one gram of Gd from the WP. This would seem to scale to 78,600,000 yr for a drip rate of 0.1 mm/yr, corresponding to removal of 9×10^{-4} g in 70,000 yr. However, EQ6 results for the 0.1 mm/yr rate show "much less than 0.1 gram Gd loss in 70,000 yr." While these statements are strictly consistent, the EQ6 results seem to allow for much greater removal of Gd at higher drip rates than do the simple calculation results.
- Perhaps most important to the key design implication of the geochemical modeling—that Gd₂PO₄ should be used as the neutron absorber—is the reliability of the Gd results. The standard database typically used in EQ3/6 modeling (data0.com.R2) contains only one Gd solid (metal) and aqueous species include only Gd⁺⁺⁺ and a series of Gd acetates. Clearly, data for other species were incorporated into an EQ3/6 database. It is important to know the source and quality of the data supporting the conclusion that Gd₂PO₄ is sparsely soluble over the entire expected pH range. In addition, experimental validation of these results would strengthen the conclusion. The argument based on stability of rare earth element (REE) phosphates in nature (section 6.3.4.2), while supportive, is insufficient for confidence building. This argument does not address the fact that modeled WP conditions may be quite different from those encountered by natural minerals, which in any case are not particularly enriched in the "middle" REE such as Gd.

Reference was made to another detailed report (Civilian Radioactive Waste Management System, 1998b) which was not available to the reviewers.

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2.7 CRITICALITY EVALUATION OF U.S. DEPARTMENT OF ENERGY SPENT NUCLEAR FUEL CO-DISPOSAL CANISTER WITH MELT AND DILUTE MATERIALS AND TEST REACTOR FUEL

2.7.1 U.S. Department of Energy Calculations

This document evaluates the criticality potential of a co-disposal canister containing U-Al ingots composed of highly enriched U that has been melted and diluted with U-238 to reduce the U-235 enrichment to 10 to 20 percent. The U-Al ingot will initially fill a given percent of the canister volume. As the canister fills with water, the ingot will erode and the material will go into solution. The analyses vary the initial fill volume and the fraction of volume that is filled with solution to determine the most reactive set of conditions.

The code that was used to calculate the reactivity of the co-disposal canister for this document was the SCALE system (Oak Ridge National Laboratory) using the ENDF/B-IV cross section library. The results from this code were compared to an applicable set of experiments, and the bias was determined to be $0.018 k_{\text{eff}}$. This value will be added to all calculated values of k_{eff} plus two standard deviations to determine a final estimate of k_{eff} .

Several assumptions about the fuel have been made to perform the calculations. The calculations assume that all fuel is fresh, and no credit is taken for burnup or fission product poisoning. Although some fuel assemblies contain U-234, which is a neutron poison, no credit is taken for the presence of U-234 in the calculations. The WP is assumed to be fully flooded for the criticality calculations, which is the most conservative moderation condition possible for these scenarios. The density of the U-Al ingot is assumed to be 3.0 g/cc. Also, the WP is assumed to be surrounded on all sides with at least 30 cm of water.

The results of the calculations show that if the U is initially enriched to 20 percent, the maximum achievable k_{eff} after accounting for bias and uncertainty was 1.020. U initially enriched to 15 percent will have a maximum possible k_{eff} of 0.952 after accounting for bias and uncertainty. A k_{eff} of 0.823 was the maximum possible for U initially enriched to 10 percent after accounting for bias and uncertainty.

2.7.2 Nuclear Regulatory Commission Evaluation

The U.S. Nuclear Regulatory Commission (NRC) reviewers find this analysis to be an excellent initial step for evaluating the potential for criticality in a melt and dilute canister. The computer code and cross section library used for the analyses are appropriate for the situations being modeled. The calculations show that without credit for any neutron poisons in the canister, U enriched to 15 percent U-235 will have a maximum possible k_{eff} of just over 0.95 after accounting for bias and uncertainty. The bias and uncertainty values may be able to be reduced with further code validation and iterations of the neutron multiplication calculations.

Three issues were identified that may increase the calculated k_{eff} and should be addressed for NRC review. The first issue is the assumption that the WP is surrounded by water on all sides. Although water is an excellent neutron reflecting material, it is not necessarily the most conservative choice for reflecting material for the WP. It is realistic to assume that the package may be buried under rockfall during disposal in the repository. This rockfall may be a better reflecting material than water because of its higher density and may lead to an increase in the k_{eff} calculated in this paper. Second, the assumption of the density of the

U-Al ingot material being 3.0 g/cm³ needs to be justified. Considering that the density of Al is 2.7 g/cm³ and the density of U is 18.68 g/cm³, this value seems unrealistically low. A larger value for the density of this material may increase the k_{eff} of the system. Third, the DOE should address the potential for the canister walls to become thinner over time because of corrosion. This would have the impact of replacing iron, which is a neutron poison and poor moderating material, with water, which has a very small neutron cross section and is an excellent moderating material. This replacement would cause the k_{eff} of the system to increase slightly and should be addressed in future criticality evaluations of the melt and dilute canister.

3 CONCLUSIONS

The DOE reports pertaining to the disposition of Al-clad fuels via both the direct co-disposal and the melt and dilute disposal options were reviewed in terms of the technical issues involved in permanent disposal. The reports reviewed include Phase II of the direct co-disposal option and the initial analysis of the melt and dilute option. In general, the criticality analyses use conservative assumptions to ensure that actual disposal conditions will be bounded by the analyses. However, there are several aspects of the analyses that do not appear to be conservative and should be addressed:

- A clearer explanation should be provided as to whether modeling the WP as horizontal is conservative
- The impact on the criticality calculations of replacing the water assumed to be surrounding the canister with rockfall should be evaluated
- The credibility and impacts on the criticality analyses of clay and oxidized steel leaking out of holes near the bottom of the package which are later plugged should be evaluated. This could increase the total quantity of moderator in the package and possibly increase the reactivity of the system. The physical models showing WP voids filled with clay degradation products should be explained and reconciled with the geochemical modeling results
- Analyses to evaluate the criticality potential of a package with the maximum percentage of Gd at the bottom and all of the U distributed throughout the canister should be conducted
- The potential for the thinning of the canister walls due to corrosion increasing the reactivity of the system should be evaluated

These issues need to be addressed to ensure that the criticality evaluations will be sufficient to show that a criticality event is unlikely to occur after the fuel is disposed of in the repository. In addition, the modeling of the criticality of direct co-disposal fuels in degraded WPs included a number of assumptions regarding the geochemistry of the environment surrounding and coming into contact with the fuels. The following items should be addressed to allow full evaluation of model interpretations:

- The pH used in geochemical modeling should be specified, and the low dissolved carbon content needs to be justified
- Geochemical model sensitivity to pH and temperature should be considered
- More detail should be provided on how degradation of glasses and alloys (including thermodynamic data) was incorporated into EQ6 calculations
- Other modeling details are needed in the areas of oxygen and carbon dioxide fugacities and water replenishment rates, and clarification is needed with regard to scaling of drip rates
- Thermodynamic data on gadolinium species need to be presented and justified

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