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U.S. Nuclear Regulatory Commission
Attn: Dr. C.A. Greene
Office of Nuclear Material Safety and Safeguards
TWFN Mail Stop 7C-24
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Washington, DC 20555

Subject: Transmittal of Report "Review of the U.S. Department of Energy Evaluation of Thermal Performance Analysis and Dissolution Rates of Melt and Dilute Aluminum-Based Spent Nuclear Fuel" (IM 20.01407.001.940)

Dear Dr. Greene:

Enclosed please find the subject report "Review of the U.S. Department of Energy Evaluation of Thermal Performance Analysis and Dissolution Rates of Melt and Dilute Aluminum-Based Spent Nuclear Fuel" (IM 20.1407.001.940) detailing the Center for Nuclear Waste Regulatory Analyses review of the two recent U.S. Department of Energy reports on thermal analysis and dissolution of Al-based spent nuclear fuel.

If you need further clarification, please do not hesitate to call me at 210.522.5538 or Sean Brossia at 210.522.5797.

Sincerely yours,

N. Sridhar

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REVIEW OF THE U.S. DEPARTMENT OF ENERGY EVALUATION OF THERMAL PERFORMANCE ANALYSIS AND DISSOLUTION RATES OF MELT AND DILUTE ALUMINUM-BASED SPENT NUCLEAR FUEL

Prepared for

Nuclear Regulatory Commission
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Prepared by

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Center for Nuclear Waste Regulatory Analyses
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QUALITY OF DATA, ANALYSES, AND CODE DEVELOPMENT

DATA: The CNWRA did not generate any original data presented in this report. Sources for other data should be consulted for determining the level of quality for those data.

ANALYSES AND CODES: The CNWRA did not perform any independent calculations or develop a code in the evaluations reported herein.

EXECUTIVE SUMMARY

Based on the Nuclear Waste Policy Act of 1982, the U.S. Department of Energy (DOE) is responsible for the ultimate disposal of government owned spent nuclear fuel (SNF), which includes Al-based research reactor fuels from both domestic and foreign sources. Al-based SNF represents less than approximately 1 volume percent of the total inventory of SNF and high-level waste (HLW) to be disposed in a geologic repository. Despite the small volume fraction Al-based fuels represent, the high enrichment levels (20 to >90 percent), complex metallurgical structure, and varied fuel geometries complicate disposability issues. Based on several factors, DOE decided to proceed with a melt/dilute process (Westinghouse Safety Management Systems, 1998). After this process, the fuel ingot is to be placed in a road-ready disposal canister, transported from the Savannah River Site to the repository and emplaced into waste packages (WPs) along with vitrified HLW.

The objective of this report is to assist the U.S. Nuclear Regulatory Commission in identifying potential technical issues relating to the disposability of the melt/dilute option for Al-based SNF in a geologic repository. The issues related to disposability considered in this report include the effects of thermal aging on the fuel, other WP components and the canister, and degradation of the fuel and subsequent radionuclide release. Safety issues related to interim dry storage facilities and processing and transportation of the fuel are outside the scope of this report.

Thermal analysis (Westinghouse Savannah River Company, 1999) of the melt/dilute canisters is necessary to demonstrate that the DOE temperature limit goals (i.e., <350 °C) for codisposal WPs and their concomitant components (i.e., HLW glass and their canisters and melt/dilute SNF and their canisters) will not be exceeded during the postclosure period. Four two-dimensional numerical modeling methodologies have been developed and applied to the thermal analysis of codisposal WPs. Three of these models are used to assess the relative influence of conduction, convection, and radiation modes of heat transfer within the confines of the codisposal WP. The fourth model was used to assess the relative merits of the different boundary conditions applied to the exterior surface of the codisposal WP in the three WP models by considering the effects of the surrounding geologic media and the potential presence of a Richard's Barrier within the emplacement drift. Although no independent analyses were performed by the Center for Nuclear Waste Regulatory Analyses to verify the modeling methodologies and results presented, a review of the mathematical bases was accomplished. This review indicated that the analytical techniques used to approximate the codisposal WP temperatures after emplacement within the proposed repository drift appear acceptable. Some concerns pertaining to the boundary conditions employed in these models were identified. An overall lack of clarity regarding which boundary conditions were used to generate some of the results was identified as well.

Because radionuclide release rates can be governed by the dissolution rate and mode of the waste form, corrosion of the waste form is also a key component in determining disposability. DOE has conducted several tests to examine the effects of environmental variables and U-Al alloy composition in both the irradiated and nonirradiated states, however, no efforts regarding the examination of the melt/dilute waste form have been reported (Westinghouse Savannah River Company, 1998, 2000). Using single pass flow-through tests in nominal J–13 water, the release rate for both irradiated and unirradiated alloys was approximately 0.2–0.45 $mgU/m^2 \cdot d$. In both bicarbonate and nitric acid solutions, all fuel forms exhibited higher dissolution rates by a factor of at least 150 with the unirradiated U-Al alloys and the U-Al SNF showing even higher dissolution rates than the other fuel forms. The heterogeneous nature of these fuels was also indicated by the observation that the relative release rates of the various radionuclides present in the SNF showed some differences compared to the U release rate, and in some cases was claimed to indicate that different radionuclides dissolved from different phases within the SNF. Corrosion testing of Al-based SNF, however, has not progressed sufficiently to determine the relationship between the dissolution rate of the fuel and the subsequent radionuclide release rate. Furthermore, DOE has not fully addressed the possibility that prior processing history may alter the behavior of U particles present in the Al matrix. The release rates determined by DOE also depend heavily on the results of the single pass flow-through tests that may be nonconservative because the primary corrosion processes responsible for release occur at the interface between the Al matrix and the U particles. This release may be accelerated in stagnant solutions by the buildup of aggressive ionic species in the occluded region between the particle and the matrix.

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1 INTRODUCTION

Based on the Nuclear Waste Policy Act of 1982, the U.S. Department of Energy (DOE) is responsible for the ultimate disposal of government owned spent nuclear fuel (SNF), which includes Al-based research reactor fuels from both domestic and foreign sources. Al-based SNF represents less than approximately 1 volume percent of the total inventory of SNF and high-level waste (HLW) to be disposed in a geologic repository. It is anticipated that a total of 255 m^3 (62.4 metric tons of heavy metal) of Al-based SNF will be received by the Savannah River Site (SRS) for processing by the year 2035. Despite the small volume fraction that Al-based fuels represent, the high enrichment levels (20 to >90 percent), complex metallurgical structure, and varied fuel geometries complicate disposability issues.

To examine multiple disposal scenarios, the alternate technology program was developed to determine the suitability and advantages of (i) directly disposing of the fuel in the repository (direct disposal) and (ii) melting the fuel elements and reformulating their composition through the addition of depleted uranium, thereby decreasing the concentration of enriched uranium (melt/dilute). Based on several factors, the DOE decided to proceed with the melt/dilute option (Westinghouse Safety Management Systems, 1998). In both cases, the fuel was to be placed in a road-ready disposal canister, ready for transport from SRS to the repository and immediate emplacement into waste packages (WPs) along with the vitrified HLW.

In fiscal year (FY) 1998, the Center for Nuclear Waste Regulatory Analyses (CNWRA) performed a topical review of the documents related to permanent disposal of Al-based fuels examining both the direct and melt/dilute options (Sridhar et al., 1998). In FY 1999, the CNWRA completed two comprehensive reviews of the analyses performed by DOE concerning the criticality issues associated with Al-based fuels (including both the direct and melt/dilute options) (Weldy et al., 1999) and issues related to ultimate disposability of these fuels (Brossia, 1999). Since the issuance of the CNWRA reports in FY 1999, the DOE has submitted two reports for review and comment by the U.S. Nuclear Regulatory Commission (NRC). These reports detail analyses performed in the areas of thermal performance of the melt/dilute waste form (Westinghouse Savannah River Company, 1999) and dissolution of Al-based SNF (Westinghouse Savannah River Company, 2000). Because no new information has been brought forth by the DOE in the area of criticality of the melt/dilute option since the issuance of the CNWRA report in early FY 1999, criticality will not be addressed in this report.

The objective, then, of this report is to assist the NRC in identifying potential technical issues relating to the disposability of the melt/dilute option for Al-based SNF in a geologic repository. The issues related to disposability considered in this report includes the effects of thermal aging on the fuel, other WP components and the canister, and degradation of the fuel and subsequent radionuclide release. Issues related to the safety of interim dry storage facilities and processing and transportation of the fuel are outside the scope of this report unless they impact the disposability of Al-based SNF in the repository.

2 ANALYSIS OF THERMAL CONDITIONS OF MELT/DILUTE ALUMINUM-BASED SPENT NUCLEAR FUEL IN THE PROPOSED REPOSITORY

2.1 STATEMENT OF ISSUE

Thermal analysis of the melt/dilute canisters is necessary to demonstrate that the DOE temperature limit goals (i.e., <350 °C based on cladding creep criteria) for codisposal WPs and their concomitant components (i.e., HLW glass and their canisters and melt/dilute SNF and their canisters) will not be exceeded during the postclosure period. The analysis should use reasonable assumptions for the boundary conditions and appropriate and consistent thermal input data. The results of the calculations should demonstrate that the predicted temperatures will not adversely affect the potential for thermal aging of waste forms and canisters such that acceptable waste form dissolution rates are not exceeded and premature failure of the canisters does not occur.

2.2 U.S. DEPARTMENT OF ENERGY TECHNICAL APPROACH AND RESULTS

Four two-dimensional (2D) numerical thermal simulations have been conducted of codisposal WPs (Westinghouse Savannah River Company, 1999). Three of these models are used to assess the relative influence of conduction, convection, and radiation modes of heat transfer within the confines of the codisposal WP. These three codisposal WP models are referred to as the Conduction, Baseline, and Detailed Models. The fourth model, called the Macro Model, was used to assess the relative merits of the different boundary conditions applied to the exterior surface of the codisposal WP in the three WP models by considering the effects of the surrounding geologic media and the potential presence of a Richard's Barrier within the emplacement drift.

All three codisposal WP models are limited in their scope in that they are only applicable to the WP and its contents. The Conduction Model, as its name implies, is only capable of capturing conduction heat transfer effects. The Baseline Model considers coupled conduction and radiation heat transport mechanisms in its formulation. In addition to conduction and radiation, the Detailed Model explicitly considers convection effects by using a computational fluid dynamics program. No solid-metal conduction heat transfer paths between the Al-SNF canister and HLW glass logs were accounted for in any of the three WP models. In other words, the codisposal WP basket has not been included in any of the WP models.

Several scenarios pertaining to variations in (i) the amount of Al-SNF contained within the codisposal WP, (ii) the boundary conditions applied to the exterior surface of the codisposal WP, and (iii) the type of fill gas (i.e., He versus air) used within the codisposal WP were investigated using the aforementioned WP models (i.e., Conduction, Baseline, and Detailed). Specifically, the volume percentages of the melt/dilute ingot contained within the centralized SNF canister of the codisposal WP considered were 50, 75, 90, and 100 percent. The influence of different boundary conditions applied to the exterior surface of the codisposal WP was also investigated. These boundary conditions included fixed temperatures intended to correspond to different time periods after emplacement of the WP within the drift (table 2-1). In addition, a natural convection heat flux was also applied to the exterior surface of the WP to assess its effects on the calculated

Table 2-1. Assumed codisposal waste package exterior surface temperatures for different time periods after emplacement within the drift (Westinghouse Savannah River Company, 1999).

Time Period After Emplacement (year)	Assumed Codisposal Waste Package Exterior Surface Temperature (°C)
0–1	150
1–5	175
5–50	190
>50	200

WP and waste form temperatures relative to the fixed temperature approach. The emplacement drift ambient air temperature was assumed to be 100 °C when applying the natural convection boundary condition.

The transient decay heat load for the Al-SNF contained in the centralized canister of the codisposal WP was estimated using the assumption that the Cs isotopes are not released during the conversion to the melt/dilute Al-SNF form. As a result, the melt/dilute form of the Al-SNF has the same transient decay heat load as the direct disposal option. Moreover, the transient decay heat loads used for the three WP only models (i.e., Conduction, Baseline, and Detailed) were based on the assumption that the Al-SNF and HLW glass logs are cooled for a period of 10 yr after fuel discharge from the reactor and production of the HLW glass logs, respectively, at the time of emplacement within the proposed repository.

As was mentioned previously, the Macro Model was used to assess the relative merits of the different boundary conditions applied to the exterior surface of the codisposal WP in the three WP models by considering the effects of the surrounding geologic media and the potential presence of a Richard's Barrier within the emplacement drift. To accomplish this task, the Macro Model considered (i) conduction, natural convection, and radiation modes of heat transfer between the emplacement drift wall and the exterior surface of the WP and (ii) conduction through the surrounding geologic medium. Unlike the three WP models, where a 10-yr Al-SNF and HLW glass logs cooling time was assumed, the Macro Model transient decay heat loads were based on a 16-yr cooling period at the time of emplacement within the proposed repository. Moreover, the decay heat loads of the Al-SNF and HLW within the WP was represented as a uniformly distributed heat flux applied to the inner surface of the codisposal WP outer barrier. The extent of the geologic medium considered in the study was defined by the radial distance from the center of the emplacement drift. The effect of changing the size of the geologic medium in the Macro Model was investigated. In particular, the outermost boundary of the geologic medium was modeled at 60, 80, 100, 120, and 160 ft from the center of the emplacement drift. The Macro Model used a fixed temperature of 30 °C at the outermost boundary of the surrounding geologic medium. In other words, a fixed ambient temperature of 30°C was assumed to exist within the geologic medium at 60, 80, 100, 120, and 160 ft away from the center of the emplacement drift.

2.3 CENTER FOR NUCLEAR WASTE REGULATORY ANALYSES EVALUATION

Although no independent analyses were performed by the CNWRA to verify the modeling results presented in the report (Westinghouse Savannah River Company, 1999), a review of the mathematical bases and a qualitative comparison of the codisposal WP Conduction, Baseline, and Detailed Model results that were provided was performed. This review indicated that the analytical techniques used to approximate the codisposal WP temperatures after emplacement within the proposed repository drift appear acceptable. Some concerns pertaining to the boundary conditions employed in the three codisposal WP models and the Macro Model were identified, however. Moreover, a lack of clarity as to which boundary conditions were used to generate some of the results presented was identified as well.

It was stated in the report (Westinghouse Savannah River Company, 1999) that the peak temperature within the codisposal WP remained below 350 °C even when the exterior surface temperature of the WP was set to 200 °C for the entire 2000-yr analysis period (including the first 50 yr). It is not clear, however, if the maximum temperatures provided in figure 23 (Westinghouse Savannah River Company, 1999) were calculated using the Baseline Model with the temperature boundary conditions conveyed in table 2-1 of this review or a fixed 200 °C for the entire 2000-yr analysis period. It was also observed that figures 36, 39, 42, 43, and 45 of the report indicate that the assumed fixed temperature boundary conditions for the three codisposal WP models (see table 2-1) are significantly below the WP surface temperatures predicted by the Macro Model.

The ambient temperature assumed when the natural convection boundary condition was applied to the exterior surface of the codisposal WP Baseline Model was 100 °C. Results from the Macro Model that were presented in the report clearly indicate that the ambient temperature of the air within the emplacement drift is somewhere between 200 and 265 °C [see figures 36, 39, 42, 43, and 45 of the report (Westinghouse Savannah River Company, 1999)]. As a result, the codisposal WP temperatures predicted by the three codisposal WP models, when using a natural convection boundary condition for the exterior surface of the WP, may be significantly underestimated.

From the perspective of the Macro Model itself, three particular concerns need to be addressed: (i) the ambient soil temperature assumption and its concomitant implementation, (ii) the representation of the engineered barrier within the Macro Model, and (iii) the effect of active ventilation during the preclosure period not being considered.

With regard to the ambient soil temperature assumption and its implementation, it has been shown (Ofoegbu, 2000) that the soil temperature around a given emplacement drift of the proposed repository is dependent on the influence of the heat generated by WPs emplaced in neighboring drifts and the temperatures assumed for the aquifer and Yucca Mountain (YM) surface boundaries. As shown in figure 40 of the report (Westinghouse Savannah River Company, 1999), the allowable peak codisposal WP temperature may be exceeded when the ambient soil temperature is greater than approximately 70 °C at a distance of 80 ft (24.4 m) from the WP center, which may very well be the case when the appropriate soil boundary conditions are considered.

The method used to represent the potential presence of a Richard's Barrier within the Macro Model has also been identified as an aspect of the model that requires some revision. In particular, the thermal conductivity assumed for the Richard's Barrier was the same as the surrounding geologic medium. Tests performed by the CNWRA staff (Green et al., 1997) indicate that crushed tuff with significant matrix saturation has a measured thermal conductivity of 0.49 W/m-K up to thermal gradients of about 240 K/m. For unsaturated crushed tuff, the measured thermal conductivity was 0.26 W/m-K for thermal gradients up to about 600 K/m. In either case, the thermal conductivity is much lower than the 1.59 W/m-K assumed for the Richard's Barrier in the report (Westinghouse Savannah River Company, 1999). Using a more realistic value for the thermal conductivity of the Richard's Barrier may significantly increase the predicted peak codisposal WP temperatures. In addition, the geometry of the Richard's Barrier needs to be changed to better reflect its actual configuration within the emplacement drift. The issues pertaining to the Richard's Barrier may be moot, however, because the use of a Richard's Barrier or backfill within the proposed repository is no longer being considered. It is expected, however, that a drip shield will be deployed within the emplacement drifts and the effects this may have on WP temperatures should be considered in future analyses.

The assumption of natural convection between the codisposal WP and emplacement drift wall during the preclosure period is conservative in that active ventilation of the drifts during this time frame is expected. As was indicated earlier, revision of the Macro Model boundary conditions needed to better reflect the influence of the heat being generated in neighboring drifts and the effect of the temperatures at the aquifer and YM surface boundaries may cause the predicted peak WP temperatures to exceed the allowable limit. Consequently, modeling of preclosure ventilation may be necessary to show that WP temperatures can be maintained within acceptable limits.

The report (Westinghouse Savannah River Company, 1999) indicated that the peak codisposal WP temperatures predicted by the three WP models (i.e., Conduction, Baseline, and Detailed) were within allowable limits. It needs to be emphasized, however, that these temperatures were obtained by employing boundary conditions on the exterior surface of the WP that the Macro Model clearly demonstrated as being unrealistic. This observation takes on additional significance when the assumed cooling time for the Al-SNF and HLW glass logs was 16 yr for the Macro Model as opposed to a 10-yr cooling time assumed for the Conduction, Baseline, and Detailed Models of the codisposal WP.

Figures 1 and 2 of the report (Westinghouse Savannah River Company, 1999) indicate that a concrete liner is to be used within the emplacement drifts. These illustrations should be updated to reflect the fact that a concrete drift liner is no longer a part of the repository design. Similarly, figure 9 of that report also should be updated to reflect the EDA-II design. And, lastly, an explanation for why the volumetric heat generation by the 75-percent volume Al-SNF case is greater than the 90-percent volume case (see tables 5 and 6 of the report) should be provided.

In summary, based on a review of the mathematical bases and a qualitative comparison of the codisposal WP Conduction, Baseline, and Detailed Model results, it was determined that the analytical techniques used to approximate the codisposal WP temperatures after emplacement within the proposed repository drift appear acceptable. Specific concerns pertaining to the boundary conditions employed in the three codisposal WP models and the Macro Model were identified. These boundary condition concerns must be addressed before a final evaluation finding can be rendered.

3 DISSOLUTION OF ALUMINUM-BASED SPENT NUCLEAR FUEL

3.1 STATEMENT OF ISSUE

Radionuclide release rates can be governed by the dissolution rate of the waste form. The release rate is also dependent on the dissolution mode of the waste form. Because the melt/dilute waste form is composed of U-rich second-phase particles in an Al solid solution matrix, dissolution occurs by selective release rather than by general overall dissolution. Incongruent dissolution may also cause a change in the makeup and orientation of criticality poisons leading to changes in the effectiveness of criticality control.

3.2 U.S. DEPARTMENT OF ENERGY TECHNICAL APPROACH AND RESULTS

Tests conducted thus far have examined the effects of environmental variables and U-Al alloy composition in both the irradiated and unirradiated states (Westinghouse Savannah River Company, 1998a, 2000), however, no efforts regarding the examination of the melt/dilute waste form have been reported. Initial test environments (Westinghouse Savannah River Company, 1998a) relied on simulated variants of the J–13 Well water chemistry at temperatures of 25 and 90 °C representing nominal, high chloride (60 ppm chloride total), low pH (~3 through additions of nitric acid), and high pH (~11 through additions of sodium hydroxide) cases with additional tests performed in nitric acid (pH ~3) and bicarbonate solutions (pH ~8). A new set of experiments (Westinghouse Savannah River Company, 2000) has been performed again in nitric acid, bicarbonate, and simulated J–13 Well water at ambient conditions under flowing solution conditions. This latest effort was conducted in a similar fashion as the Light Water Reactor SNF dissolution studies. In all cases, four fuel types were examined in the irradiated condition (U-Al, U-Al_x, U_3O_8, and U_3Si_2) as well as a limited set of unirradiated U-Al alloys of varied compositions.

In both the initial tests and the latest set of experiments, the fuel composition and irradiation state did not significantly influence release rates in solutions nominally equivalent to J–13 water. In the other solutions tested, however, there was a difference in dissolution rate observed between the fuels. Using single pass flow-through tests in nominal J–13 water, the release rates for both irradiated and unirradiated alloys was approximately 0.2 mgU/m^2·day (Westinghouse Savannah River Company, 1998a). Subsequent testing has shown similar results with an average dissolution rate for these materials of 0.45 mgU/m^2·day (Westinghouse Savannah River Company, 2000). In both bicarbonate and nitric acid solutions, the unirradiated U-Al alloys and U-Al SNF exhibited significantly higher dissolutions rates than the other fuel forms. For example, in bicarbonate and nitric acid solutions, U-Al_x, U_3O_8, and U_3Si_2 all had dissolution rates between 33 and 65 mgU/m^2·d, whereas U-Al SNF and unirradiated U-Al (19 wt% U) exhibited dissolution rates 107–320 mgU/m^2·d and 103–960 mgU/m^2·d. Similar, though smaller, differences were reported previously but were explained as resulting from differences in the exposed area of fuel particles for each fuel type (Westinghouse Savannah River Company, 1998a) and not from an inherent difference in the dissolution rates of the fuels. It should be noted that high dissolution rates were also observed in the low-pH J–13 variant and in static immersion nitric acid tests conducted previously (Westinghouse Savannah River Company, 1998a). The latest work (Westinghouse Savannah River Company, 2000) did not discuss the corrosion modes observed, however, it was previously reported (Westinghouse Savannah River Company, 1998a) that two distinct corrosion modes were observed of preferential dissolution of the Al matrix surrounding U particles followed by either general or localized corrosion of the Al matrix. The general or localized nature of the second stage attack was found to be dependent on the pH (at low pH, general corrosion; at neutral pH, localized).

The relative release rates of the various radionuclides present in the SNF also showed some differences compared to the U release rate. For example, the Pu release rate from all fuels except U-Al was less than 50 percent of the release rate for U, and this difference was not thought to be a result of Pu solubility limitations. Cs and Sr were both found to dissolve at rates greater than twice that of U in the U_3Si_2 fuel, which was claimed to indicate that both these radionuclides dissolved from a different phase than did U. The other fuels did not show any consistent trends with respect to Cs and Sr release rates nor was any trend apparent in the Tc release rate data.

3.3 CENTER FOR NUCLEAR WASTE REGULATORY ANALYSES EVALUATION

Corrosion testing of Al-based SNF has not progressed sufficiently to determine the relationship between the dissolution rate of the fuel and the subsequent radionuclide release rate. This relationship is particularly important considering the melt/dilute waste form has not been evaluated and tends to be heterogeneous in structure and dissolution of the fuels tends to be selective. Furthermore, DOE has not fully addressed the possibility that prior processing history may alter the behavior of U particles present in the Al matrix. Hence, testing of as-cast U-Al alloys and the various SNF forms may not be an accurate simulation of the melt/dilute ingot, and further testing of actual as processed melt/dilute waste forms is recommended. This testing will aid in determining the radionuclide release rates from actual fuel that could serve to verify the approach taken by the DOE thus far.

The release rates determined by DOE seem to depend heavily on the results of the single pass flow-through tests. Though these results are useful in providing a quantitative measure of the release rate, they may be nonconservative. In heterogeneous materials, such as the melt/dilute waste form, the primary corrosion processes responsible for release occur at the interface between the Al matrix and the U particles. This release may be accelerated in stagnant solutions by the buildup of aggressive ionic species in the occluded region between the particle and the matrix. The increase in the aggressiveness of the chemistry in this occluded region could then promote and further accelerate corrosion and radionuclide release. In a flowing solution, development of this aggressive chemistry in the occluded region is somewhat minimized as a result of constant dilution with fresh bulk solution. Thus, the release rates determined from the flow-through tests may not be conservative and should be compared to the results obtained from other test methods.

Though most of the SNFs exhibited similar dissolution rates when exposed to the same environment, the U-Al SNF and the unirradiated U-Al ingot experienced much higher dissolution rates. This difference may indicate a possible nonconservatism if the release rates from the other fuels are used to bound the performance of the melt/dilute ingot because U-Al more closely represents the melt/dilute waste form. Furthermore, the possible galvanic effect between U particles and the Al matrix has not been fully examined. Based on corrosion potential and corrosion rate measurements as a function of U content in U-Al alloys, Sridhar et al., (1998) highlighted that (i) U is more anodic compared to Al but U-Al particles are more cathodic than either U or Al, and (ii) the dissolution behavior of U-Al alloys is dependent on the volume fraction of U-Al particles present. This issue has not been adequately addressed and could become highly important if the rate of overall dissolution and subsequent release of U particles change with the U concentration in the melt/dilute ingot. The importance is further highlighted by the observation that the relative release rates of the radionuclides were not always the same, possibly indicating that the release rates are controlled by the dissolution rate of different phases present in the alloy. If this interpretation of the results is correct, the issue of galvanic interaction between the different phases and particles becomes even more important.

The possibility of significant segregation also exists and, thus, the dissolution and release rates may spatially vary within the melt/dilute ingot. The noncongruent nature of the dissolution of these fuels also may be important when considering the maintenance of criticality control. It has been reported that criticality poisons will likely be added to the melt/dilute waste form (Westinghouse Savannah River Company, 1998b). The DOE should examine the possibility that preferential dissolution and release of the poisons from the ingot during dissolution could result in a loss of criticality control.

The dissolution rate was also found to be a function of the environment. Though it would be expected that the dissolution rate in low pH solutions would be higher than that observed in near-neutral environments based on changes in the stability of the passive film on Al (Pourbaix, 1974), the observation that the bicarbonate and the nitric acid environments exhibited similar dissolution rates is puzzling. Comparing the pH and likely aggressiveness of the bicarbonate solution to the simulated J–13 solution also used, similar results would be expected from these two environments. The dissolution rates in the bicarbonate solutions were consistently at or more than two orders of magnitude larger than the comparable rates observed in J–13. One possible explanation is the precipitation of corrosion products resulting from depressed solubility limits with the various species present in J–13 water not present in the bicarbonate solution. It should also be recognized that though the initial solution temperature was ~25 °C, the temperature of the solution likely increased as a result of radioactive decay heat from the irradiated SNF. Such an increase in temperature may have exacerbated the precipitation of corrosion products and released radionuclides further. Because no photographs were taken of the SNF samples after exposure, and no weight change measurements were performed, the possibility of precipitation and, as a result, low corrosion rates cannot be confirmed. It is suggested that this possibility be examined in subsequent testing. In addition, the technical basis for continued testing in J–13 water when it is clear this testing will not likely represent the water chemistry inside the WP, given evaporation, radiolysis, and interactions with other WP components, is also lacking.

To evaluate the impact of Al-based SNF on the eventual overall performance assessment (PA) case for the proposed repository at YM, the testing plan by the DOE does not provide any mechanistic information or data that can serve as input parameters for predicting performance. Additionally, the reports concerned with dissolution and radionuclide release did not make any recommendation nor did they provide a conclusive statement for the disposability of this fuel type based on the results obtained thus far. Furthermore, the true impact of radionuclide release from Al-based SNF on overall repository performance cannot be easily ascertained based on the work performed by DOE to date because there is no clear relationship between the environments chosen for investigation and the expected WP internal water chemistry.

4 SUMMARY

Potential technical issues pertaining to recent DOE reports on thermal analysis and dissolution of Al-based SNF were reviewed. The main focus of the review was on the methodologies, assumptions, and results used by the DOE and how these may influence the disposability of this waste form. The main points identified from these reports in the areas of thermal analysis and waste form dissolution are outline below.

Thermal Analysis

Thermal analysis of the melt/dilute option and dissolution of the various Al-based SNF waste forms were examined based on the new reports received from DOE. Thermal analysis of the melt/dilute canisters is necessary to demonstrate that the DOE temperature limit goals (i.e., <350 °C) for codisposal WPs and their concomitant components (i.e., HLW glass and their canisters and melt/dilute SNF and their canisters) will not be exceeded during the postclosure period. The analysis should use reasonable assumptions and the results of the calculations performed should demonstrate that the predicted temperatures will not adversely affect the potential for thermal aging of waste forms and canisters such that acceptable waste form dissolution rates are not exceeded and premature failure of the canisters does not occur.

Four sets of 2D numerical thermal simulations have been conducted of codisposal WPs. Three of these models are used to assess the relative influence of conduction, convection, and radiation modes of heat transfer within the confines of the codisposal WP. The fourth model (Macro Model) was used to assess the relative merits of the different boundary conditions applied to the exterior surface of the codisposal WP in the three WP models by considering the effects of the surrounding geologic media and the potential presence of a Richard's Barrier within the emplacement drift. The scope of these models was limited in that they are only applicable to the WP and its contents. Several scenarios pertaining to variations in (i) the amount of Al-SNF contained within the codisposal WP (50, 75, 90, and 100 percent filled), (ii) the boundary conditions applied to the exterior surface of the codisposal WP, and (iii) the type of fill gas (i.e., He versus air) used within the codisposal WP were investigated using the WP models.

Although no independent analyses were performed by the CNWRA to verify the modeling methodologies and results presented in the report, a review of the mathematical bases was performed indicating that the analytical techniques used to approximate the codisposal WP temperatures after emplacement within the proposed repository drift appear acceptable. Some concerns pertaining to the boundary conditions in the three codisposal WP models and the Macro Model were identified, however, an overall lack of clarity which boundary conditions were used to generate some of the results presented was identified as well. For example, it was stated that the peak temperature within the codisposal WP remained below 350 °C even when the exterior surface temperature of the WP was set to 200 °C for the entire 2000-yr analysis period (including the first 50 yr). It is not clear, however, if the maximum temperatures provided were calculated using the Baseline Model with a range of temperature boundary conditions or a fixed 200 °C for the entire 2000-yr analysis period. Additionally, the ambient temperature assumed when the natural convection boundary condition was applied to the exterior surface of the codisposal WP Baseline Model was 100°C. Results from the Macro Model clearly indicate that the ambient temperature of the air within the emplacement drift is somewhere between 200 and 265 °C. As a result, the codisposal WP temperatures predicted by the three codisposal WP models, when using a natural convection boundary condition for the exterior surface of the WP, may be significantly underestimated.

From the perspective of the Macro Model itself, three particular concerns need to be addressed: (i) ambient soil temperature assumptions and implementation, (ii) the representation of the engineered barrier within the Macro Model, and (iii) the effect of active ventilation during the preclosure period. With regard to the ambient soil temperature assumptions and implementation, it has been shown (Ofoegbu, 2000) that the soil temperature around a given emplacement drift of the proposed repository is dependent on the heat generated by WPs emplaced in neighboring drifts and the temperatures assumed for the aquifer and YM surface boundaries. The method used to represent the potential presence of a Richard's Barrier within the Macro Model has also been identified as an aspect of the model that requires some revision. The report indicated that the peak codisposal WP temperatures predicted by the three WP models were within allowable limits. These temperatures were obtained, however, by employing boundary conditions on the exterior surface of the WP that the Macro Model clearly demonstrated were unrealistic. This observation takes on additional significance when the assumed cooling time for the Al-SNF and HLW glass log was 16 yr for the Macro Model as opposed to a 10-yr cooling time assumed for the Conduction, Baseline, and Detailed Models of the codisposal WP.

Dissolution and Radionuclide Release

Because radionuclide release rates can be governed by the dissolution rate and mode of the waste form, corrosion of the waste form is a key component to determine disposability. DOE has conducted several tests to examine the effects of environmental variables and U-Al alloy composition in both the irradiated and nonirradiated states, however, no efforts regarding the examination of the melt/dilute waste form have been reported. Using single pass flow-through tests in nominal J–13 water, the release rate for both irradiated and unirradiated alloys was approximately 0.2–0.45 $mgU/m^2 \cdot d$. In both bicarbonate and nitric acid solutions, all fuel forms exhibited higher dissolution rates by a factor of at least 150 with the unirradiated U-Al alloys and U-Al SNF showing even higher dissolution rates than the other fuel forms. The heterogeneous nature of these fuels was also indicated by the observation that the relative release rates of the various radionuclides present in the SNF showed some differences compared to the U release rate, and in some caseswas claimed to indicate that different radionuclides dissolved from a different phases within the SNF.

Corrosion testing of Al-based SNF, however, has not progressed sufficiently to determine the relationship between the dissolution rate of the fuel and the subsequent radionuclide release rate. This relationship is particularly important considering the melt/dilute waste form has not been evaluated. Furthermore, DOE has not fully addressed the possibility that prior processing history may alter the behavior of U particles present in the Al matrix. Hence, testing of as-cast U-Al alloys and the various SNF forms may not be an accurate simulation of the melt/dilute ingot, and further testing of actual as-processed melt/dilute waste forms is recommended. This testing will aid in determining the radionuclide release rates from actual fuel that could serve to verify the approach and methodology taken by the DOE thus far.

Based on the information presented to date, a number of clarifications are suggested. The release rates determined by DOE also depend heavily on the results of the single pass flow-through tests that may be nonconservative because the primary corrosion processes responsible for release occur at the interface between the Al matrix and the U particles. This release may be accelerated in stagnant solutions by the buildup of aggressive ionic species in the occluded region between the particle and the matrix. The possible galvanic effect between U particles and the Al matrix has not been fully examined. The possibility of inconsistencies in the results observed in simulated J–13 and bicarbonate solutions should also be addressed. More importantly, evaluating the impact of Al-based SNF on the eventual overall PA case for the proposed repository at YM cannot easily be supported by the testing plan used by the DOE because the plan does not provide any mechanistic information or data that can serve as input parameters for predicting performance. Additionally, the reports concerned with dissolution and radionuclide release did not make any recommendation nor did they provide a conclusive statement for the disposability of this fuel type based on the results obtained thus far. Furthermore, the true impact of radionuclide release from Al-based SNF on overall repository performance cannot be easily ascertained based on the work performed by DOE to date because there is no clear relationship between the environments chosen for investigation and the expected WP internal water chemistry.

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