1/23

REVIEW OF THE U.S. DEPARTMENT OF ENERGY EVALUATION OF THE DISPOSABILITY OF ALUMINUM-BASED SPENT NUCLEAR FUEL FINAL REPORT

Prepared for

Nuclear Regulatory Commission
Contract NRC-02-97-009

Prepared by

C. Sean Brossia

Center for Nuclear Waste Regulatory Analyses
San Antonio, Texas

September 1999

2/23

CONTENTS

3/23

CONTENTS (cont'd)

4/23

ACKNOWLEDGMENTS

This report was prepared by the Center for Nuclear Waste Regulatory Analyses (CNWRA) for the Nuclear Regulatory Commission (NRC), Office of Nuclear Material Safety and Safeguards, Division of Waste Management, under Contract No. NRC-02-97-009. This report is an independent product of the CNWRA and does not necessarily reflect the views or regulatory position of the NRC.

The author acknowledges the helpful comments and suggestions provided by Dr. C. Greene (NRC), J. Weldy (CNWRA), and Dr. A. Chowdhury (CNWRA) as well as the technical review provided by Dr. N. Sridhar (CNWRA) editorial review by B. Long, and programmatic review by Dr. B. Sagar (CNWRA). The assistance of A. Ramos in preparing this report is also appreciated.

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 aluminum (Al)-based research reactor fuels from both domestic and foreign sources. Al-based SNF represents less than approximately 1 vol% of the total inventory of SNF and high-level waste (HLW) to be disposed in a geologic repository. 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. Based on several factors, the DOE decided to proceed with the melt-dilute option (Westinghouse Safety Management Solution, 1998). After the melt-dilution process, the fuel ingot was to be placed in a "road-ready" disposal canister for interim storage and eventual transport from the Savannah River Site to the repository, with immediate emplacement into waste packages (WPs) along with vitrified HLW.

The objective of this report is to assist the Nuclear Regulatory Commission in identifying potential technical concerns relating to the disposability of Al-based SNF in a geologic repository. The review identifies performance issues that could impact preclosure operations and those that could affect postclosure performance. From the preclosure integrated safety analysis (ISA) performance perspective, there are two primary issues. Potential deterioration of the road-ready canister during interim storage is the first. The DOE should further investigate the actual corrosion rate of the road-ready canister under interim storage conditions considering corrosion from within the canister together with external atmospheric corrosion through the use of monitored canisters. This is particularly critical in light of the interim storage times considered—up to 100 yr—which are in excess of the current design canisters' lifetime by nearly a factor of 2. The second issue concerns the possible detrimental effects of long-term thermal exposure on the integrity of the canister and the waste form. Though it is likely that the internal temperatures of the canister and WP will not exceed the DOE set limit of 350 °C, there is still some concern regarding the potential for accelerated degradation of the canister resulting from long-term thermal exposure during interim storage and preclosure. Further evaluation of these effects is suggested.

With regard to postclosure performance of Al-based SNF in a geologic repository, four key issues need to be resolved:

- Determination of adequate criticality control. Based on what has been presented by the DOE, it seems likely that criticality poisons will be added to the melt-dilute waste form. DOE should describe techniques to verify that the poisons are uniformly distributed throughout the melt-dilute ingot.

- Exclusion of pyrophoric and explosive materials from the WP is important to prevent premature breach of the WP and subsequent radionuclide release. The DOE calculated the maximum quantity of water that can be trapped in the melt-dilute canister that would lead to corrosion of Al and subsequent buildup of hydrogen gas in the canister. The approach and assumptions used seem reasonable, however, the maximum volume of water allowable represents approximately 0.4–1 vol% of the canister. It is unclear how achievable this low water level is in practice.

- There is a general lack of data on waste dissolution rates and morphology associated with the melt-dilute ingots. In particular, it is likely that the ingot will be inhomogeneous, which could lead to spatial variations in the dissolution mechanisms (localized versus uniform) and rates. It is suggested that further evaluation be performed of the dissolution behavior of melt-dilute ingot waste form compositions in the environment anticipated to contact the waste form.

- Determination of the effects of long-term thermal exposure. Neither the effects of long-term thermal exposure on material performance nor the potential detrimental effects of long-term thermal exposure on eventual radionuclide release through alterations of WP internal components have been examined fully.

REFERENCE

Westinghouse Safety Management Solution. 1998. *Criticality Evaluation of DOE SNF Codisposal Canister with Melt and Dilute MTR Fuel.* WSMS–CRT–98-0003. Revision 0. Aiken, SC: Westinghouse Savannah River Company.

7/23

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 aluminum (Al)-based research reactor fuels from both domestic and foreign sources. Al-based SNF represents less than approximately 1 vol% 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 Al-based fuels represent, the high enrichment levels (20 to > 90 percent), complex metallurgical structure, and varied fuel geometries complicate disposability issues.

In an effort to examine multiple disposal scenarios, the Alternate Technology Program was developed (Westinghouse Savannah River Company, 1997) to determine the suitability and advantages of directly disposing the fuel in the repository (direct disposal) and melting the fuel elements and reformulating their compositions through the addition of depleted U which would decrease the concentration of enriched U (melt-dilute). Based on several factors, the DOE decided to proceed with the melt-dilute option (Westinghouse Safety Management Solution, 1998). In both cases, the fuel was to be placed in a "road-ready" disposal canister for interim storage and eventual transport from SRS to the repository with immediate emplacement into waste packages (WPs) along with vitrified HLW.

In fiscal year 1998 (FY1998), the Center for Nuclear Waste Regulatory Analyses (CNWRA) completed 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 early FY1999, the CNWRA prepared a review 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). Because the DOE has decided to proceed with the melt-dilute option for disposal rather than direct disposal, this report will not discuss in detail analyses performed by the DOE examining the direct disposal case. Furthermore, as 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 FY1999 (Weldy et. al., 1999), criticality will not be addressed in extensive detail. If the DOE decides to pursue direct disposal further, additional analyses of the technical bases used will likely be needed.

The objective of this report is to assist the Nuclear Regulatory Commission (NRC) in identifying potential technical concerns relating to the disposability of Al-based SNF in a geologic repository. The review identifies repository performance issues from the prospective of those that could impact preclosure operations and those that could affect postclosure performance. As such, issues that influence processing such as the ultimate disposition of Cs resulting from melt-dilute processing and the potential for steam explosions during processing are not considered. Preclosure issues include those related to the integrity of the road-ready canister; the effects of thermal aging on the fuel, other WP components, and the canister; and any detrimental effects of interim dry storage on disposability. Issues related to postclosure performance include the effects of thermal aging on fuel, other WP components and the canister, degradation of the fuel and subsequent radionuclide release, assurance of criticality control, and pyrophoricity. Safety issues associated with interim dry storage facilities, processing, and transportation of the fuel are outside the scope of this report.

2 PRECLOSURE OPERATIONS

The ISA requirements for the geologic repository operations area are described in (draft) 10 CFR 63.112 (Nuclear Regulatory Commission, 1999), which specifies that structures, systems, and components important to safety be designed so they will still perform their necessary contribution to safety assuming the occurrence of design basis events. Because the primary purpose of the road-ready canister is to avoid bare fuel handling during waste transfer from transportation to disposal overpacks and during potential waste retrieval operations that could require repackaging the waste, the integrity of the road-ready canister is important. In line with this, thermal conditions of the WPs and the internal components (including waste canisters) and the waste forms themselves as influenced by radioactive decay should not significantly impair each component's safety function or result in the acceleration of waste form dissolution and, thus, radionuclide release. The establishment of acceptance criteria for Al-based SNF and the creep of fuel rods may also play a role in determining eventual release. In addition, there are several other objective measures that must be met to facilitate disposal, including dimensions of the road-ready canister and canister labeling. Because these issues are quantitatively met or are not met and do not rely on interpretation or assumptions, they will not be examined in further detail because no technical evaluation is necessary. Maintenance of criticality control is also an important consideration in draft 10 CFR 63.112(e)(6) for the preclosure ISA (Nuclear Regulatory Commission, 1999). The DOE reports address criticality during the postclosure period; this review will examine that issue in the next chapter. This chapter evaluates the effects listed on preclosure operations related to the disposability of Al-based SNF. It is divided into four sections, each defining the issue, providing a summary of the DOE technical approach and the results examining the issue, and presenting the CNWRA evaluation.

2.1 DETERIORATION AND CONFIGURATION OF ROAD-READY/ DISPOSABLE CANISTER DURING INTERIM STORAGE

2.1.1 Statement of Issue

After processing, the melt-dilute Al-based SNF waste form may be stored at SRS in the road-ready canisters for periods up to 100 yr prior to permanent disposal at a geologic repository, even though the design life of the canister is only 40 yr (Westinghouse Savannah River Company, 1997). The detailed design of the canister and the loading method have not been finalized completely, but it is expected that the canisters will be constructed of a low-carbon, stabilized austenitic stainless steel (SS) (e.g., Nitronic 50[1]) with a depleted U or other suitable end plug material that will be seal-welded to the canister shell after backfilling the canister with an inert gas (Westinghouse Savannah River Company, 1998a,b). Deterioration of the canister through corrosion or thermal embrittlement of the welds could also affect waste transfer operations and acceptability at the repository as a result of loss of mechanical strength to withstand handling operations or penetration and premature radionuclide release. In addition, there are a number of canister configuration issues including maximum canister weight that should be considered.

[1]UNS S20910 (ASTM XM–19) with a nominal composition (wt%) of 0.06 C max, 4.0–6.0 Mn, 0.04 P max, 0.03 S max, 1.0 Si max, 20.5–23.5 Cr, 11.5–13.5 Ni, 1.5–3.0 Mo, 0.20–0.40 N, 0.10–0.30 Nb, and 0.10–0.30 V.

2.1.2 U.S. Department of Energy Technical Approach and Results

The DOE has mainly focused on the material performance of Al-based fuel and fuel cladding (Westinghouse Savannah River Company, 1998a). Examination for internal corrosion of a canister constructed of Type 304 SS has been performed, however, the objective was not to examine the performance of the canister material under dry storage conditions (Westinghouse Savannah River Company, 1997, 1998a). DOE has plans to evaluate degradation under dry storage conditions through monitoring relative humidity (RH) and temperature, as well as through nondestructive evaluation inspections of canisters (Westinghouse Savannah River Company 1998b). This activity has not been initiated to date. DOE also examined the disposability issues with regard to road-ready canisters that may be damaged as a result of handling, corrosion, or both. It was determined that, as long as the canister has sufficient structural integrity that it can be handled safely, it would be considered acceptable for disposal (Westinghouse Savannah River Company, 1998a).

2.1.3 Center for Nuclear Waste Regulatory Analyses Evaluation

Significant internal corrosion of the road-ready canister is not anticipated if the canister is adequately filled with the melt-dilute waste form and vacuum dried and back-filled with an inert gas. It is unlikely there would be any trapped water in the melt-dilute waste form, so the only water of concern is water that could condense from the air space in the canister. Based on pyrophoricity, the maximum water content currently allowed to be present is approximately 0.4–1 vol% of the canister (Westinghouse Savannah River Company, 1998a). Previous calculations have estimated that a concentration of 0.4 M aqueous iodide could develop inside the canister, but would likely be lower as a result of interactions between iodide and Cs (Sridhar et al., 1998). In the case of the melt-dilute waste form, however, it is predicted that at least some fraction of the Cs present in the fuel elements will be released during the melt-dilute process (Westinghouse Savannah River Company, 1998c). Due to the potential formation of oxidizing radiolysis products (e.g., peroxide), the possibility for localized corrosion of the canister exists (Szklarska-Smialowska, 1986; Sedriks, 1996). Localized corrosion could also occur at weldments that are known to act as preferential sites for localized corrosion (Sedriks, 1996). As localized corrosion proceeds at a significantly faster local dissolution rate than the rest of the material surface (Kelly, 1994), breach of the canister at weldments would result in exposure of the fuel during handling. As a result, the tests conducted on instrumented canisters should include monitoring of internal corrosion and periodic sampling and measuring the condensed water chemistry inside a canister containing the melt-dilute waste form. More emphasis should also be placed on monitoring the conditions (e.g., RH and temperature) in the storage area and possible degradation of the canister in the storage environment.

Also unknown are the effects of long-term (up to 40+ yr) thermal exposure at temperatures approaching 200 °C (Westinghouse Savannah River Company, 1998b) on material performance. Because thermal embrittlement of Type 316L SS weldments under these conditions is not considered kinetically significant (Sridhar et al., 1994), it is likely that thermal embrittlement of the Nitronic 50 proposed construction material will not be significant also. Given the differences in the chemistry and properties of these materials, however, the effect of long-term thermal exposure should be examined.

Also it should be noted that the 2,750 lb maximum canister weight defined by DOE may be exceeded when the canister is filled with the melt-dilute ingot (Westinghouse Savannah River Company, 1998b). Further clarification of the potential detrimental effects of this on handling and disposability is recommended.

10/23

2.2 PRECLOSURE THERMAL CONDITIONS OF MELT-DILUTE ALUMINUM-BASED SPENT NUCLEAR FUEL IN THE REPOSITORY

2.2.1 Statement of Issue

Thermal analysis of the melt-dilute canisters using reasonable assumptions on boundary conditions and appropriate and consistent thermal input data is necessary to demonstrate that the DOE temperature limit goals (<350 °C) for co-disposal WP and WP potential components (i.e., HLW glass and its canister, and melt-dilute SNF and its canister) have not been exceeded. The calculations performed should show that the predicted temperatures also will not adversely affect properties, including the potential for thermal aging of waste forms and canisters which could lead to enhanced waste form dissolution rates and premature failure of the canisters.

2.2.2 U.S. Department of Energy Technical Approach and Results

A two-dimensional (2D) finite element model of the melt-dilute canisters filled to either 75 or 90 vol% capacity (which represents 101 and 121 material test reactor fuel assemblies) was used to calculate the temperature profiles of the WP and its respective internal components to times approaching 2000 yr. All canisters were assumed to be vacuum dried and backfilled with He and all fuel assemblies were assumed to have been discharged from the reactor for at least 5 yr (Westinghouse Savannah River Company, 1998a) or 10 yr (Westinghouse Savannah River Company, 1998c) using an exponential SNF thermal decay profile as the thermal source term. Furthermore, the package geometry was assumed intact with a repository temperature of either 100 or 150 °C and WP emplacement at the centerline of the drift. The thermal load of the melt-dilute waste form was also assumed lower than the comparable direct disposal configuration as a result of the release of all ^{85}Kr and some Cs isotopes (^{134}Cs, ^{137}Cs, and ^{137}Ba daughter) during the melt-dilute process.

Given these assumptions, the temperature as a function of time for the two filling conditions (75 and 90 vol% filled) were calculated. The peak temperature at emplacement in the repository was calculated to be 284 °C for the 75 vol% filled cases and 264 °C for the 90 vol % filled case. At 90 yr after emplacement, the temperatures were calculated to have decayed to 135 and 133 °C (Westinghouse Savannah River Company, 1998c). Calculations were also performed assuming the canisters were backfilled with air rather than He. In this case, peak temperatures of 347 and 286 °C were calculated for the two cases at emplacement. Under all conditions examined, the calculated temperatures were well below the minimum melting temperatures of the metallic phases present in the melt-dilute form. For example, the lowest liquidus on the Al-U phase diagram is at the eutectic composition (86.8Al-13.2U) at 641 °C (ASM International, 1992). All other possible Al-U compositions have higher liquidus temperatures. Uranium silicides as well as U, Al, and Si oxides that also may be present all have liquidus temperatures even higher than this (ASM International, 1992). Because calculated temperatures were well below the melting temperatures and peak temperatures for He backfilled canisters are well below the DOE maximum allowable peak temperature of 350 °C, the DOE concluded that the melt-dilute waste form would be acceptable. The maximum peak temperature criteria of 350 °C originated from the Mined Geologic Disposal System Draft Disposability Interface Specifications (MGDS-DIS) document (U.S. Department of Energy, 1998a) to minimize the risk of cladding creep. The requirements set forth in the MGDS-DIS are derived from public and environmental safety standards established by the Environmental Protection Agency and the NRC.

2.2.3 Center for Nuclear Waste Regulatory Analyses Evaluation

Most assumptions are reasonable, including the idealizations regarding heat transfer modes, effective thermal conductivity, and convective cooling by He. Additionally, the canister surface temperatures reported as acceptance criteria are predicted to be met at times less than approximately 50 yr. Canister surface temperatures of less than 190 °C for the first 50 yr are specified in the DOE acceptance criteria (Westinghouse Savannah River Company, 1998c), yet the canister surface temperature does not drop below this until around 50 yr after emplacement. Further explanation and clarification on the apparent disconnect between the stated acceptance criteria and the predicted results is needed.

The WP and WP component temperatures predicted from analyzing Al-based SNF containing WPs are inconsistent with the temperatures reported in the total system performance assessment viability assessment (TSPA-VA) (U.S. Department of Energy, 1998). Based on the thermal analyses performed for Al-based SNF (Westinghouse Savannah River Company, 1998c), decay in temperature with increasing time is predicted for the WP. In the case of the TSPA-VA (U.S. Department of Energy, 1998b), however, the temperature is predicted to increase or remain nearly constant for the first 20 yr followed by decay. In addition, the peak temperature predicted by the analysis of Al-based SNF containing WPs [at t = 0 yr (i.e., emplacement)] was between 264 and 284 °C, depending on canister filling. In TSPA-VA, the average WP peak temperature occurs at around 20 yr after emplacement and is generally less than 170 °C. Thus, further clarification of these differences is recommended.

Based on DOE analyses, it does not seem likely that the WP or its components will exceed the peak temperature limit of 350 °C chosen by the DOE. Furthermore, it also does not seem likely there will be melting of any metallic or oxide compounds within the melt-dilute waste form. Analysis of the potential detrimental effects of long-term thermal exposure to the temperatures predicted on waste form stability and eventual radionuclide release (from both the Al-based SNF melt-dilute form as well as the HLW glass logs present) as well as the integrity and properties of both Al-based SNF and HLW canisters has not been addressed and should be examined further.

2.3 PYROPHORICITY AND EXPLOSION RESISTANCE

2.3.1 Statement of Issue

Though not explicitly required in the draft 10 CFR Part 63 (Nuclear Regulatory Commission, 1999), the possible detrimental effects of including pyrophoric or explosive materials in the WP should be examined to determine if such inclusions would significantly change the magnitude and time of the resulting expected annual dose.

2.3.2 U.S. Department of Energy Technical Approach and Results

With the processing steps involved in forming the melt-dilute ingot and filling the canisters (e.g., vacuum drying and backfilling with He), DOE assumed that the water content within the canister will be low. For explosion resistance, DOE limited the water content so that the maximum amount of hydrogen that can be produced from either Eqs. (2-1) or (2-2) (at temperatures less than and greater than 80 °C) will be less than 4 vol%. With this maximum hydrogen concentration, the maximum quantity of water that can be present in the canister was calculated as a fraction of the canister volume. Given the likely canister

12/23

dimensions, the maximum volume of water that can be present in the canister is approximately 1 vol% of the canister or about 2 L.

$$2Al + 6H_2O \rightarrow Al_2O_3 \cdot 3H_2O + 3H_2 \quad (2\text{-}1)$$

$$2Al + 4H_2O \rightarrow Al_2O_3 \cdot H_2O + 3H_2 \quad (2\text{-}2)$$

DOE has taken the approach of assuming that no pyrophoric materials in concentrations that could compromise safety over the possible internal WP conditions (25–400 °C and 1–5 atm) be allowed inside the WP (Westinghouse Savannah River Company, 1998a). The main pyrophoric material of concern in the analyses performed by DOE is UH_3. The formation of UH_3 is theoretically possible as a result of Al corrosion, which then produces hydrogen, as shown in Eq. (2-1) and (2-2), which could combine with U to form UH_3. Given the composition of the melt-dilute ingot, the U present is thought to be predominantly in the form of UAl_4, with some $U_xAl_ySi_z$ possibly present. As a result, it was not considered likely that UAl_4, U_3Si_2 or $U_xAl_ySi_z$ compounds would be reduced by the expected hydrogen pressures present (maximum of 4 vol% hydrogen) to produce any significant UH_3 (Westinghouse Savannah River Company, 1998a).

2.3.3 Center for Nuclear Waste Regulatory Analyses Evaluation

Regarding flamability and explosion resistance, the assumptions used to determine the maximum water content in the canister to prevent explosion, combustion, are both reasonable and conservative. There is some concern, though, about the likelihood of achieving the low water levels that the DOE calculates as necessary. Even though 2 L is a significant volume of water, it only represents around 1 vol% of the canister (75 vol% filled case). Though the DOE approach appears conservative, the technical basis for the claim that reduction of U-Al and U compounds to UH_3 is not likely should be provided.

2.4 ACCEPTANCE CRITERIA FOR INTERIM DRY STORAGE OF ALUMINUM-BASED SPENT NUCLEAR FUEL—EFFECTS ON DISPOSABILITY

2.4.1 Statement of Issue

The condition of the fuel elements and cladding at the time of receipt could influence handling and safety issues related to acceptance (10 CFR Part 72) and subsequent disposal [draft 10 CFR Part 63 (Nuclear Regulatory Commission, 1999)]. In particular, prior corrosion or damage to the fuel elements together with the presence of debris and corrosion products and cut fuel elements, which could contain entrapped water, must all be disposed safely similar to the safe disposal of intact fuel. The focus of this section though, is to examine the possible effects of the acceptance criteria for interim storage on disposability and not on the acceptance criteria themselves.

2.4.2 U.S. Department of Energy Technical Approach and Results

Because melt-dilute process removes any water present in either the fuel elements or corrosion products or debris, the possibility of entrapment of water in the canister is not significant (Westinghouse Savannah River Company, 1998a). Furthermore, as all fuel elements will be melted and recast as ingots, prior

corrosion or mechanical damage to the fuel is unimportant. Debris and any corrosion products present also will be incorporated into the cast ingot, which may or may not result in the formation of slag (Westinghouse Savannah River Company, 1998a). If slag forms, it too will be emplaced in the canister along with the ingot and, thus, is not considered to affect disposability (Westinghouse Savannah River Company, 1998a).

2.4.3 Center for Nuclear Waste Regulatory Analyses Evaluation

The DOE approach is acceptable assuming that the integrity of the canister is maintained throughout the interim storage period.

3 POSTCLOSURE OPERATIONS

The postclosure performance objectives for the geologic repository are described in the draft regulations in 10 CFR 63.113 (Nuclear Regulatory Commission, 1999), which calls for the inclusion of multiple barrier systems composed of both natural and engineered systems such that the peak annual expected dose to a member of the critical group does not exceed 25 mrem during the first 10,000 yr after permanent closure. As a result, processes leading to the degradation of the engineered barrier system are important. Though not specifically stated in draft 10 CFR Part 63 (Nuclear Regulatory Commission, 1999), based on the potential effects of disposing Al-based SNF, such processes would include criticality control, pyrophoricity and explosion hazards, waste form dissolution and radionuclide release (including types of potentially released radionuclides), and the effects of long-term thermal aging on the WP and its components, including waste forms and internal canisters, to ensure that omission of such features, events, and processes (FEPs) would not significantly alter the time or magnitude of the expected annual dose. The CNWRA evaluation of DOE reports provided in this chapter addresses these particular aspects. Criticality control will only be summarized briefly, as this subject has been extensively reviewed and discussed in a separate report (Weldy et al., 1999).

3.1 CRITICALITY CONTROL

3.1.1 Statement of Issue

The postclosure requirements in the proposed NRC regulations in 10 CFR 63 (Nuclear Regulatory Commission, 1999) do not specifically require criticality control. According to the draft 10 CFR Part 63.114(c) and (f), determination of the effects of omitting FEPs (such as criticality) on the time and magnitude of the expected annual dose need to be examined. Calculation of the neutron multiplication factor (k_{eff}) and consequences of criticality are aspects of this requirement.

3.1.2 U.S. Department of Energy Technical Approach and Results

Determination of k_{eff} was performed using the SCALE computer code (developed by Oak Ridge National Laboratory) using the ENDF/B-IV cross-sectional library. It was assumed that degradation of the melt-dilute waste form had occurred within an intact canister with the canister free volume filled with water. Based on a $k_{eff} < 0.95$ criteria, the maximum canister free volume was calculated for the various ^{235}U enrichment levels being considered (20, 15, and 10 percent). For these ^{235}U enrichment levels in the fuel ingot, it was predicted that the maximum free volume percentage was 10 and 40 for the 20 and 15 percent enrichment cases. For an enrichment level of 10 percent, it was predicted that criticality could not be achieved (Westinghouse Savannah River Company, 1998a). As a result of the two principal ingot sizes considered (with canister fill volumes of 75 and 90 percent), addition of criticality poison materials was suggested, with further analysis recommended.

3.1.3 Center for Nuclear Waste Regulatory Analyses Evaluation

Evaluation of the approach taken by DOE to determine criticality control issues has been reviewed elsewhere (Weldy et al., 1999). Briefly it was suggested that future analyses address concerns regarding the conceptual model of assuming that the WP is surrounded by water as opposed to the possibility of rock, backfill, or both; the assumed density of the fuel ingot possibly being unrealistically low; and the possible effects of canister thinning by corrosion—all of which could lead to higher values of k_{eff} than predicted.

Additionally, DOE did not specify criticality poisons. Unlike the case for the direct disposal option where criticality poisons can be incorporated in the canister internal structure, criticality poisons in the melt-dilute case must be incorporated into the ingot. The DOE should examine the possibility of preferential dissolution and release of the poisons from the ingot during ingot dissolution together with segregation of poisons during casting and processing operations. The DOE should also ensure that the poisons chosen are added in sufficient concentration to maintain criticality control in the cases where the canisters are filled only to 75 percent and poisons should be added in sufficient concentration to account for variability in ingot size. The consequences of criticality on performance were not discussed by DOE.

3.2 PYROPHORICITY AND EXPLOSION RESISTANCE

3.2.1 Statement of Issue

Though not explicitly required in the draft 10 CFR Part 63 (Nuclear Regulatory Commission, 1999), the possible detrimental effects of including pyrophoric or explosive materials in the WP should be examined to determine if such inclusions would significantly change the magnitude and time of the resulting expected annual dose.

3.2.2 U.S. Department of Energy Technical Approach and Results

As discussed in section 2.3, with the processing steps involved with forming the melt-dilute ingot and filling of the canisters (e.g., vacuum drying and backfilling with He), DOE assumed that the water content within the canister will be low. The water content has been limited such that the maximum amount of hydrogen that can be produced will be less than 4 vol% of the canister. It was also noted that in the case of a breached canister, the quantity of water present would likely exceed the maximum allowable for explosion resistance. No analysis on the possibility of hydrogen generation and build up as a result of a canister breach was performed, however (Westinghouse Savannah River Company, 1998a).

3.2.3 Center for Nuclear Waste Regulatory Analyses Evaluation

Regarding flamability and explosion resistance, the assumptions used to determine the maximum water content in the canister to prevent explosion, combustion, or both are reasonable and conservative. There is some concern, though, if a canister breach occurs, significantly more water could be present that could lead to increased hydrogen production. DOE did not examine this scenario in any detail. Additional analysis examining the case where a breached canister leads to significant quantities of water should be provided.

3.3 WASTE FORM DISSOLUTION AND RADIONUCLIDE RELEASE

The dissolution rate of the waste form subject to solubility limitations and surface segregation can determine the rate of radionuclide release. Thus, the performance of the waste form is a critical aspect to achieving the system performance requirements in the draft 10 CFR Part 63 (Nuclear Regulatory Commission, 1999). The composition and radionuclide distribution in the waste form can also impact the release rate. This can arise if selective dissolution of the radionuclides preferentially occurs with respect to the Al matrix. Thus, there are two primary subsections in this section dealing with waste form dissolution and distribution of radionuclides in the waste form.

3.3.1 Waste Form Dissolution

3.3.1.1 Statement of Issue

As mentioned previously, the release rates of highly soluble radionuclides 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 most likely occurs by selective release rather than by uniform overall dissolution of both the Al matrix and U-Al particles.

3.3.1.2 U.S. Department of Energy Technical Approach and Results

Tests conducted by DOE thus far, examined the effects of environmental variables and U-Al alloy composition in both the irradiated and nonirradiated states (Westinghouse Savannah River Company, 1998d). All test environments relied on variants of simulated 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. Additional tests were performed in nitric acid solutions (pH ~3) and bicarbonate solutions (pH ~8). Four fuel types were examined in the irradiated condition (UAl, UAl_x, U_3O_8, and U_3Si_2) and unirradiated U-Al alloys ranging from 10 to 25 wt% U were also tested.

The fuel composition and irradiation states did not significantly influence release rates. The environmental composition, however, had a significant influence. For example, in single pass flowthrough tests in nominal J-13 water, the release rates for both irradiated and nonirradiated alloys were approximately 0.2 $mgU/m^2 \cdot d$. In nitric acid and the low pH J-13 variant (irradiated and nonirradiated alloys) flowthrough tests, the release rates were 30–100 and 200–400 $mgU/m^2 \cdot d$. Similar results were observed in the static immersion tests. Of note, the high pH and low pH J-13 solutions were found to be the most aggressive. At low and high pH values, Al does not form a stable protective oxide film and thus can undergo rapid dissolution (Pourbaix, 1974). In general, two distinct corrosion modes were observed. The first stage consisted of preferential dissolution of the aluminum matrix surrounding U-Al particles, leaving the particles in relief for subsequent release and fall out. In the second stage, either general or pitting corrosion of the Al matrix occurred depending on the pH of the environment (low and high pH resulted in general corrosion and neutral pH resulted in pitting).

A limited number of experiments were also performed in which the U-Al alloys were coupled to SS (a potential canister construction material) and Al. In these tests, the SS and Al plates acted as cathodes to the U-Al alloys. While coupling to the Al plate increased the corrosion rate slightly, coupling to the SS plate significantly increased the dissolution rate of the U-Al alloys in all environments examined. Thus, the possibility exists for enhanced fuel degradation and radionuclide release if coupling between the fuel and the canister occurs. The grade of SS used was not specified.

3.3.1.3 Center for Nuclear Waste Regulatory Analyses Evaluation

The corrosion tests of the melt-dilute fuel ingots have not progressed sufficiently to determine the relationship between the dissolution rate of the fuel and the subsequent radionuclide release rate; particularly considering the melt-dilute ingots are heterogeneous in structure and composition and dissolution of the fuels

may be selective and undergo different corrosion modes (e.g., uniform, pitting) in different regions. Testing of as-cast U-Al alloys may not be an accurate simulation of the melt-dilute ingot. Further testing of actual as-processed melt-dilute waste forms is recommended.

The release rates determined by DOE seem to depend solely on the results of the single pass flowthrough test results. Though these results are useful in providing a quantitative measure of the intrinsic dissolution rate without transport limitation, they may be nonconservative. This is because 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-Al particles that 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 would then promote and accelerate further corrosion and 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 flowthrough tests may not be conservative and should be compared to the results obtained from other test methods.

The galvanic effect between the U-Al particles and the Al matrix is still unclear. As noted in a previous report (Sridhar et al., 1998), DOE should explain the contradictions in the relationship between composition and corrosion rates. Sridhar et al. (1998) argued that, based on corrosion potential and corrosion rate measurements as a function of the U content in U-Al alloys, (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. The possible galvanic interaction between U-Al particles and the Al matrix as yet, has not been addressed adequately. This could become a critical issue if the rate of overall dissolution and subsequent release of U-Al particles changes with the U concentration in the melt-dilute ingot. Then, not all the results obtained thus far are clearly applicable. Furthermore, the possibility of significant segregation exists and thus the dissolution and release rates may spatially vary within the ingot.

The potential detrimental influence of microbial colonies developing on fuels has been reported by Guenther et al. (1995) and Wolfram et al. (1998), but has not been examined by the DOE. Because of the possibility that microbes may lay dormant but alive during the dry period and then cause accelerated corrosion during the wet period, a limited examination of the effect that these organisms could have on the dissolution of the fuel form and radionuclide release rate is recommended.

When evaluating the impact of Al-based SNF on the eventual overall performance assessment case for the proposed repository at Yucca Mountain, the testing plan thus far followed by the DOE does not provide any mechanistic information or data that can serve as input parameters for predicting performance. The draft ASTM standard (American Society for Testing and Materials, 1999), currently being evaluated, provides a number of recommended test methods for examining the corrosion behavior of spent nuclear fuel. However, these test methods are similar to those already employed by the DOE and also do not provide any mechanistic understanding of the dissolution process. Though only a small fraction of the total radionuclide inventory, the true impact of radionuclide release from Al-based SNF on overall repository performance cannot be easily ascertained based on work conducted by DOE to date. Furthermore, there is no clear relationship between the environments chosen for investigation and those expected in WP water chemistry.

3.3.2 Radionuclide Characterization and Inventory

3.3.2.1 Statement of Issue

The radionuclide inventory of Al-based SNF can affect factors important in determining disposability, including radiogenic heat production, criticality potential, and performance in a geologic repository. As a result of the heterogeneous structure of the melt-dilute waste form, there may be significant segregation of various actinides and fission products within the ingot. Because preferential selective dissolution of the ingot occurs (as was discussed in the previous section), the distribution of radionuclides within the ingot is an important factor in evaluating eventual release.

3.3.2.2 U.S. Department of Energy Technical Approach and Results

There are a number of differences between various Al-based SNFs that will eventually be received by SRS for treatment. These differences include burn-up rates, U enrichment, time since reactor discharge, and uncertainty and validity of available data and information on these characteristics, especially for fuel elements arriving from foreign research reactors (Westinghouse Savannah River Company, 1998a). Based on the likely fuel types to be received, the ORIGEN 2 computer code was used to calculate the likely isotopic composition and total fissile material concentration of the fuel elements.

It is anticipated by the DOE that the radionuclide composition of the melt-dilute waste form will be performed through the analysis of dip samples taken from the molten ingot during processing and through postsolidification characterization (Westinghouse Savannah River Company, 1998a). The analytical techniques proposed include gamma-ray spectroscopy, thermal ionization mass spectroscopy, and x-ray fluorescence spectrometry. It is thought that through the use of these and possibly other methods, that the radionuclide inventory and the total fissile material concentration will be determined.

3.3.2.3 Center for Nuclear Waste Regulatory Analyses Evaluation

Because of the heterogeneous structure of the melt-dilute ingot, the possibility exists for segregation of radionuclides and fissile material throughout the ingot. Furthermore, the possibility also exists for segregation of these products into the oxide slag that would form during melting and casting operations. The distribution of these species may be important in calculating the ultimate release rate of radionuclides from the fuel and possible differences arising from variations in the dissolution rates of the fuel and slag. Furthermore, any samples taken for characterization during mixing will contain both molten fuel and slag. Thus, the radionuclide inventory and the fissile material concentration will represent an average of the fuel plus the slag. The DOE should examine further the possibility of radionuclide and fissile material segregation and the potential impact segregation between the fuel and slag could have on release. Methodologies to ensure that the composition of both the melt and the slag are accurately analyzed also should be established and validated.

3.4 POSTCLOSURE THERMAL CONDITIONS OF MELT-DILUTE ALUMINUM-BASED SPENT NUCLEAR FUEL IN THE REPOSITORY

3.4.1 Statement of Issue

Thermal analysis of the melt-dilute canisters using reasonable assumptions on boundary conditions and appropriate and consistent thermal input data is necessary to demonstrate that the DOE temperature limit goals (<350 °C) for codisposal WP and WP components (i.e., HLW glass and its canister, and melt-dilute SNF and its canister) will not be exceeded during the postclosure period. The calculations performed should show that the predicted temperatures will not adversely affect properties including the potential for thermal aging of waste forms and canisters that could lead to enhanced waste form dissolution rates and premature failure of the canisters.

3.4.2 U.S. Department of Energy Technical Approach and Results

A 2D finite element model of the melt-dilute canisters filled to either 75 or 90 vol% capacity (which represents 101 and 121 fuel assemblies) was used to calculate the temperature profiles of the WP and its respective internal components to times approaching 2,000 yr. All canisters were assumed to be vacuum dried and backfilled with He and all fuel assemblies were assumed to have been discharged from the reactor for at least 5 yr (Westinghouse Savannah River Company, 1998a) or 10 yr (Westinghouse Savannah River Company, 1998c) using an exponential SNF thermal decay profile as the thermal source term. Furthermore, the package geometry was assumed intact with a repository temperature of either 100 or 150 °C with WP emplacement at the centerline of the drift. The thermal load of the melt-dilute waste form was also assumed lower than the comparable direct disposal configuration as a result of the release of all ^{85}Kr and some Cs isotopes (^{134}Cs, ^{137}Cs, and ^{137}Ba daughter) during the melt-dilute process.

Given these assumptions, the temperature as a function of time for the two filling conditions (75 and 90 vol% filled) were calculated to 2,000 yr. The peak temperature at emplacement was calculated to be 284 °C for the 75 vol% filled case and 264 °C for the 90 vol% filled case. After 1,000–2,000 yr, the nominal temperature of the WP internal components approaches that of the ambient predicted repository temperature (Westinghouse Savannah River Company, 1998c). Under all conditions and times examined, the calculated temperatures were well below the minimum melting temperatures necessary for melting of the metallic phases present in the melt-dilute form. Because of this and that the peak temperatures for He backfilled canisters are well below the DOE maximum allowable peak temperature, it was concluded that the melt-dilute waste form would be acceptable.

3.4.3 Center for Nuclear Waste Regulatory Analyses Evaluation

Most assumptions are reasonable, including the idealizations regarding heat transfer modes, effective thermal conductivity, and convective cooling by He. One additional concern beyond those discussed for preclosure (section 2.2) is the imposed boundary condition that the repository temperature was assumed constant during the first 1,000–2,000 yrs at either 100 or 150 °C. The technical bases for these temperatures and the assumption that the repository temperature remains constant is unclear and needs further clarification once the melt-dilute waste form composition and canister dimensions are finalized.

4 SUMMARY

DOE reports pertaining to the disposition of Al-based SNF in a geologic repository were reviewed by focusing on the melt-dilute disposal option. The review examined the issue of disposability from the preclosure and postclosure time frames. An independent analysis of the effects of these fuels on the eventual repository performance has not been performed and is recommended.

4.1 PRECLOSURE PERFORMANCE

With the choice of the melt-dilute methodology for processing Al-based SNF, there are only two main issues important to preclosure performance in the proposed repository: deterioration of the road-ready canister during interim storage, and possible detrimental effects of long-term thermal exposure on the integrity of the canister and waste form. Regarding degradation of the road-ready canister during interim storage, the DOE should determine the actual corrosion rate under interim storage conditions considering both corrosion from within the canister and external atmospheric corrosion through the use of monitored canisters. This is particularly critical in light of the interim storage times considered—up to 100 yr which are in excess of the predicted design lifetime of the canisters by nearly a factor of 2. Though it is likely the temperature of the WP canister will not exceed the DOE set limit of 350 °C, there is still some concern regarding the potential for accelerated degradation of the canisters resulting from long-term thermal exposure during interim storage. Creep of cladding and fuel, though of importance in the direct disposal case, is not considered a likely event in the case of the melt-dilute waste form.

4.2 POSTCLOSURE PERFORMANCE

There are four main technical concerns with regard to postclosure performance of Al-based SNF in a geologic repository.

- Maintenance of adequate criticality control. Based on what has been presented by the DOE, it seems that the addition of criticality poisons to the melt-dilute waste form is necessary. DOE should describe techniques required to ensure that the poisons are uniformly distributed throughout the melt-dilute ingot are required.

- Exclusion of pyrophoric and explosive materials from inclusion in the WP is the second concern because it is important to prevent premature breach of the WP and subsequent radionuclide release. The DOE has calculated the maximum quantity of water that can be trapped in the melt-dilute canister that would lead to Al corrosion and subsequent buildup of hydrogen gas in the canister. The approach and assumptions used seem reasonable. The only concern is that the maximum volume of water allowable represents approximately 0.4 to 1 vol% of the canister. It is unclear how reasonable the assumption is that this can be readily achieved during processing.

- There is a general lack of data on dissolution rates and morphology associated with the melt-dilute ingots. In particular, there exists the possibility that the ingot will be inhomogeneous, which could lead to spatial variations in the dissolution mechanisms (localized versus uniform) and rates. It is also unclear how the data generated to date reflect the likely anticipated conditions inside a breached WP. It is recommended that the DOE

further evaluate the dissolution behavior of the likely melt-dilute ingot waste forms under technically justifiable conditions.

- As was mentioned for preclosure, the effects of long-term thermal exposure on material performance should be examined further.

5 REFERENCES

American Society for Testing and Materials. 1999. *Draft Standard Guide for Corrosion Testing of Aluminum-Based Spent Nuclear Fuel in Support of Repository Disposal.* West Conshohocken, PA: American Society for Testing and Materials.

ASM International. 1992. Alloy phase diagrams. *Metals Handbook.* Volume 3. Materials Park, OH: ASM International.

Guenther, R.J., A.B. Johnson, Jr., A.L. Lund, E.R. Gilbert, S.P. Pednekar, F.M. Berting, L.L. Burger, S.A. Bryan, and T.M. Orlando. 1995. *Initial Evaluation of Dry Storage Issues of Spent Nuclear Fuels in Wet Storage at the Idaho Chemical Processing Plant.* INEL–96/0140. Richland, WA: Pacific Northwest Laboratory.

Kelly, R.G. 1994. Pitting. *Corrosion Tests and Standards: Application and Interpretation.* Philadelphia, PA: American Society for Testing and Materials.

Nuclear Regulatory Commission. 1999. *Disposal of High-Level Radioactive Wastes in a Proposed Geologic Repository at Yucca Mountain, Nevada.* Federal Register 64(34): 8663–8679. Washington, DC: U.S. Government Printing Office.

Pourbaix, M. 1974. *Atlas of Electrochemical Equilibria in Aqueous Solutions.* Houston, TX: NACE International.

Sedriks, A.J. 1996. *Corrosion of Stainless Steels.* New York: Wiley-Interscience Publications.

Sridhar, N., G.A. Cragnolino, D.S. Dunn, and H.K. Manaktala. 1994. *Review of Degradation Modes on Alternate Container Designs and Materials.* CNWRA 94-010. San Antonio, TX: Center for Nuclear Waste Regulatory Analyses.

Sridhar, N., A. Chowdhury, D. Deere, V. Jain, D. Pickett, and J. Weldy. 1998. *Review of the Technical Issues Related to Interim Storage and Disposal of Aluminum-Based Spent Nuclear Fuel.* San Antonio, TX: Center for Nuclear Waste Regulatory Analyses.

Szklarska-Smialowska, Z. 1986. *Pitting Corrosion of Metals.* Houston, TX: NACE International.

U.S. Department of Energy. 1998a. *Mined Geologic Disposal System Draft Disposability Interface Specification.* B00000000–01717–4600–00108. Revision 00. Las Vegas, NV: Office of Civilian Radioactive Waste Management.

U.S. Department of Energy. 1998b. *Viability Assessment of a Repository at Yucca Mountain Volume 3: Total System Performance Assessment.* DOE/RW–0508. Las Vegas, NV: Office of Civilian Radioactive Waste Management.

Weldy, J., D. Pickett, N. Sridhar, and S. Brossia. 1999. *Evaluation of the U.S. Department of Energy Aluminum-Based Spent Fuel Criticality Analysis*. San Antonio, TX: Center for Nuclear Waste Regulatory Analyses.

Westinghouse Safety Management Solution. 1998. *Criticality Evaluation of DOE SNF Codisposal Canister with Melt and Dilute MTR Fuel*. WSMS–CRT–98–0003. Revision 0. Aiken, SC: Westinghouse Savannah River Company.

Westinghouse Savannah River Company. 1997. *Alternative Aluminum Spent Nuclear Fuel Treatment Technology Development Status Report*. WSRC–TR–97–00345. Aiken, SC: Westinghouse Savannah River Company.

Westinghouse Savannah River Company. 1998a. *Disposability Assessment: Aluminum-Based Spent Nuclear Fuel Forms*. WSRC–TR–98–00227. Aiken, SC: Westinghouse Savannah River Company.

Westinghouse Savannah River Company. 1998b. *Bases for Functional Performance Requirements for a Spent Nuclear Fuel Treatment and Storage Facility*. WSRC–TR–98–00228. Aiken, SC: Westinghouse Savannah River Company.

Westinghouse Savannah River Company. 1998c. *Thermal Analysis of Repository Codisposal Waste Packages Containing Aluminum Spent Nuclear Fuel*. WSRC–TR–98–00158. Aiken, SC: Westinghouse Savannah River Company.

Westinghouse Savannah River Company. 1998d. *Preliminary Report on the Dissolution Rate and Degradation of Aluminum Spent Nuclear Fuels in Repository Environments*. WSRC–TR–98–00290. Aiken, SC: Westinghouse Savannah River Company.

Wolfram, J.H., R.E. Mizia, and W.J. Dirk. 1998. Microbial sampling of aluminum-clad spent nuclear fuel. *Proceeding of the Corrosion '98 Conference*. Paper No. 98162. Houston, TX: NACE International.