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REVIEW OF THE TECHNICAL ISSUES RELATED TO INTERIM STORAGE AND DISPOSAL OF ALUMINUM BASED SPENT NUCLEAR FUEL

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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 evaluation reported here.

EXECUTIVE SUMMARY

In accordance with the Nuclear Waste Policy Act of 1982 (NWPA), the U.S. Department of Energy (DOE) is responsible for the interim management and ultimate disposal of government owned spent nuclear fuel (SNF), which includes the foreign and domestic Al-based research reactor fuels. In 1995, the DOE decided to process and store the Al-based fuels at the Savannah River Site (SRS) in sealed containers until they can be disposed at a geologic repository. This storage configuration is termed "road ready" by the DOE. Al-based research reactor fuels and targets are a small part (less than 1 percent by volume) of the total inventory of SNF and high-level waste to be disposed in a geologic repository. A total of 266 m^3 [23.8 metric tons of heavy metal (MTHM)] of Al-based SNF is expected to be sent to SRS for processing and temporary storage. These fuels are slated to arrive from domestic and foreign research reactors as well as other DOE sites. Al-based fuels at various sites are currently stored underwater, but continued storage under these conditions is not desirable because of the susceptibility to corrosion of the cladding and fuel. While these fuels constitute only a small fraction of the total amount of SNF to be emplaced in the proposed geologic repository at Yucca Mountain, disposition of these fuels, including treatment, temporary storage, and permanent disposal, presents a significant challenge because of the diverse geometries, high enrichments (20 to greater than 90 percent U^{235}), and the complex metallurgical characteristics of the fuel.

The DOE Office of Spent Fuel Management chartered the Research Reactor SNF Task Team to assist in the development of a technical strategy for interim management and ultimate disposition of research reactor SNF. In 1996, the task team recommended that the DOE should proceed with the parallel development of two treatment technologies: direct disposal (or co-disposal) in a repository and melt and dilute followed by disposal. The direct/ co-disposal option entails placing the SNF in type 316L stainless steel canisters that will be emplaced along with high-level waste glass canisters in a disposal overpack. The melt and dilute option would involve melting an appropriate mixture of Al-based SNF with depleted uranium and aluminum to achieve the desired composition and enrichment, casting them in type 316L stainless steel pour canisters, followed by co-disposal with glass waste canisters. The task team also recommended that the DOE should utilize the existing SRS processing capabilities and should begin to work immediately with the Nuclear Regulatory Commission (NRC) and other regulatory agencies to reach agreement on SNF disposal requirements.

A memorandum of understanding and Interagency Agreement between the NRC and the DOE were established for the NRC to assist the DOE in identifying potential technical issues associated with the ultimate disposition of these fuels (Stablein, 1997). This report provides an evaluation of the technical and regulatory issues related to the final disposal of the Al-based SNF. The processing, transportation, and interim storage safety issues that are related to the requirements in 10 CFR Parts 70, 71, and 72 are outside the scope of this review. It is also not the purpose of this report to provide input to the selection of one treatment technology over the other. The evaluation of technical issues relevant to disposal of the Al-clad fuels is performed with reference to the regulatory requirements in 10 CFR Part 60:

10 CFR 60.113: Performance of particular barriers after permanent closure
10 CFR 60.131: General design criteria for the geologic repository operations area
10 CFR 60.135: Design criteria for the waste package and its components

However, it is anticipated that the NRC regulatory requirements, especially in terms of postclosure subsystem performance requirements, will change as a result of pending NWPA legislation and the National Academy of Science (NAS) recommendations that were made as directed in the Energy Policy Act. Hence, the

postclosure performance evaluation may change to focus on criticality and dose to a critical group at a selected location.

Interim Dry Storage and Its Effects on Preclosure Operations

According to 10 CFR 60.131(b), the structures, systems, and components important to safety in the geologic repository operations area shall be designed so that they will perform the necessary safety functions, assuming the occurrence of design basis events (DBE). In this context, the integrity of fuel canisters is important during waste transfer operations.

The Al-based fuels in the direct/co-disposal technology option may be stored at SRS for periods up to 50 years (a period of 100 years is mentioned in some documents) prior to disposal at a geologic repository, although the intended design life of the dry storage canister is 40 years (WSRC, 1997). The detailed design and method of loading the SNF into the storage canisters have not been decided at this time. It is expected that the fuels will be placed in a road ready canister made of type 316L stainless steel and sealed by welding so that preclosure operations involving waste transfer from transportation to disposal overpacks will not involve handling bare fuel. The fuel will be placed in a basket structure made of carbon steel or stainless steel containing appropriate neutron absorbers. Interim dry storage can affect waste transfer operations through (i) deterioration of the road ready canister due to either internal corrosion or thermal embrittlement of the welds or (ii) alteration of the fuel and cladding geometry or mechanical properties. Alteration of the fuel and cladding would be expected to affect waste transfer operations only in the event that the road ready canister is breached prior to emplacement of the waste in the repository.

The DOE has not considered the deterioration of the road ready canisters during interim storage. Such deterioration may occur inside the canister due to the presence of residual moisture in combination with deleterious fission products such as I^{129} which may form iodides in aqueous solution as well as oxidizing species (e.g., iodates) in the presence of radiolysis. Stainless steels and Ni-base alloys have been shown to suffer localized corrosion in iodide environments, although the localized corrosion severity of iodide is less than that of chloride (Koch, 1995). Long-term exposure of stainless steel welds that contain ferrite to temperatures around 300 °C may result in the formation of brittle alpha-prime phase. However, this effect is not considered to be significant for the road ready canister because of lower anticipated temperatures during storage and the discontinuous nature of the ferrite phase in the stainless welds (Sridhar et al., 1994). The DOE should include evaluation of the internal environment chemistry and corrosion of their instrumented test canister intended for the study of the drying behavior of road ready canister.

The effect of temporary storage on eventual performance in a repository has been considered by the DOE and specific acceptance criteria for the drying and storage conditions have been formulated. The DOE has performed considerable testing of the cladding corrosion and has analyzed the creep of the cladding and fuel. Based on these tests, a 200 °C temperature limit has been placed as an acceptance criterion for temporary storage. Center for Nuclear Waste Regulatory Analyses (CNWRA) staff have concluded that the 200 °C temperature limit for dry storage of 50 years may not be sufficiently conservative based on the data generated by SRS on cladding and simulated fuel materials (WSRC, 1997). For example, based on the measured weight gain and metal-loss data, the time to penetrate 3.0 mils of alloy Al 1100 cladding at 200 °C is greater than 50 years. However, for the alloy Al 5052 cladding material, the time to penetrate 3 mils is less than 50 years without breakaway corrosion. If breakaway corrosion occurs after 1,400 hours at 200 °C, the time to penetrate 3 mils is less than 1 year. Furthermore, under breakaway corrosion conditions (WSRC, 1997), the 0.38 mm (15 mil) thick cladding will be completely penetrated in 1 year. The acceptance criteria do not

address Al-based fuels that will be received from INEEL that are known to be corroded significantly (Guenther et al., 1995).

While considerable testing of the cladding has taken place, providing valuable insight into the behavior of these materials under vapor phase corrosion conditions, it does not appear to be sufficient to determine whether corrosion under steam or water vapor would deteriorate the cladding sufficiently to prevent safe fuel transfer, if necessary. This is because (i) some of the tests have not been carried out for a sufficiently long time (a few thousand hours at most), (ii) tests under radiation conditions may result in breakaway corrosion at longer time periods, and (iii) metallographic examination of the corroded specimens has not been reported to ascertain whether internal oxidation would arrest once weight gain stopped. Indeed, long-term autoclave tests at 250 °C (WSRC, 1997) showed that the boehmite (hydrated aluminum oxide) was transformed to corundum (anhydrous oxide) suggesting that water may be released to continue further oxidation internally. Based on the data reported to date, a bounding assessment of the oxidation or humid-air corrosion of cladding and fuel cannot be performed. The tests performed to date indicate the importance of water chemistry and radiolysis on vapor corrosion, but do not provide sufficient guidance on estimating the degree of oxidation or penetration of the cladding and fuel under dry storage conditions. For example, it is unclear what the chemistry of the environment would be in the dry storage cask. Because the loading of the road ready package with fuel is going to occur in the storage pool, the initial water chemistry is likely to be related to pool water chemistry rather than condensate water chemistry, which is related to atmospheric chemistry at SRS.

The creep analysis, while conservative in some respects, assumes that the only source of stress is the weight of the fuel elements, and ignores the thermal stresses as short-lived. The thermal stresses are indicated to be several orders of magnitude higher than that due to the fuel weight and hence may cause significant creep deformation in the short term. Additionally, the creep rate has a cubic dependence on grain size and there is some uncertainty regarding the assumed grain size. DOE analyses of structural integrity of co-disposal canister focus only on tip-over accident as a credible DBE. The bases for ignoring other credible DBEs such as vertical and horizontal drops are not provided. Finally, although embrittlement of the irradiated U-Al alloy can occur at temperatures between 150 to 200 °C, this is neglected because of the low stress from fuel weight. However, impact loading from a variety of credible DBE is ignored in determining whether this embrittlement can affect fuel assembly integrity. Hence, the 200 °C temperature limit for storage is not conservative from the point of view of creep and embrittlement. The thermal analysis needed to determine whether the peak temperatures would exceed the DOE limits should be performed with consistent input data and boundary conditions.

Finally, the preclosure performance of the road ready canister containing the melt-dilute waste form may be influenced by the thermal history of the canister during the waste form pouring and storage periods. However, corrosion due to residual moisture from pool water is not a likely factor in the melt and dilute option. Additionally, because of the larger cross sectional area of the melt-dilute waste form, creep and oxidation of the waste form are not expected to adversely affect the waste transfer operations.

Postclosure Performance

As mentioned previously, the postclosure performance requirements are anticipated to change as a result of pending legislation and the NAS recommendations. The evaluation of the DOE reports did not consider the subsystem performance requirements in 10 CFR 60.113, but considered the source term issues relevant to overall system performance. However, the design criteria for the waste package and its components in

10 CFR 60.131 and 60.135 may still be applicable. Specifically, design criteria in terms of criticality control, and explosive, pyrophoric, and chemically reactive materials were evaluated.

The criticality analyses for the direct/co-disposal option have many conservative assumptions. However, some aspects of the calculations need to be addressed to eliminate potential nonconservatisms:

- Justification is needed for the assumption that 1 percent of the uranium in the fuel is U-234. Since this isotope can act as an efficient neutron absorber and is not typically present in these quantities in fresh fuel, its inclusion may represent implicit burnup/decay credit.

- More compact neutronic systems may need to be considered where there is a segregation of the degraded fuel plates due to corrosion and creep and the resultant gravity driven compaction from the neutron absorber plates which are left intact.

- The segregation of the neutron absorber as a result of migration of the absorber in the case of degraded fuel should be addressed.

- The effect of reconfiguration of the fissile material outside the waste package into a critical configuration needs to be addressed. It was indicated that these calculations will be made in the future.

- While gadolinium has several advantages as a neutron absorber over boron, the higher burnup of the gadolinium needs to be considered in determining the initial concentration of gadolinium.

The criticality of the melt-dilute waste, although less of a concern than the direct/co-disposal fuel, needs to be addressed in order to determine whether neutron absorber materials need to be incorporated in the melt.

The DOE has addressed the issues of pyrophoricity and reactivity in a reasonably conservative fashion. The Al-based fuels are not as pyrophoric as metallic uranium. The limitation placed on the volume of water as part of the dry storage acceptance criteria and the pressurization of the canister by helium ensure that the concentration of hydrogen generated through corrosion and radiolysis along with any oxidants that may be present will not reach flammability limits.

The Al-based fuels, due to their small volume compared to the commercial and other DOE fuels, are not expected to contribute significantly to the overall dose from the repository. The DOE preliminary total system performance assessment (PA) of the DOE fuels has indicated that the contribution to dose from a representative Al-based fuel is similar to that from an equivalent MTHM commercial spent fuel (Duguid et al., 1997). The dose at a time period less than 20,000 years is dictated by I^{129} and Tc^{99}, whereas the dose at longer time periods is dictated by ^{237}Np. The peak dose for the Al-based SNF occurs at about 10,000 years and is predicted to be about 2.5 times that for an equivalent MTHM of commercial spent fuel (Duguid et al., 1997). At longer times, the total dose is essentially equal for the two types of spent fuel. Independent calculation of the dose from Al-based fuels have not been made by the NRC or CNWRA and is recommended for the future. The calculation of radionuclide inventory using ORIGEN may have uncertainties due to the uncertain history of some of the fuels. Additionally, it is not clear whether ORIGEN has been validated for spent fuels which bound the range of composition and burnup characteristics represented by the Al-based SNF.

The corrosion tests of the co-disposal and the melt-dilute fuels have not progressed sufficiently to determine the relationship between dissolution rate of the fuel and the radionuclide release rate. This relationship is especially important since the fuel material in both cases is highly heterogeneous and the dissolution is selective. The test protocol developed by WSRC (1997) and currently in balloting in the American Society for Testing and Materials appears to be deficient in that an insufficient range of environmental conditions is specified. For example the effect of steel and basket dissolution products and radiolysis is not considered in the test protocols. In tests of Al cladding materials in hot brine (Brodda and Fachinger, 1995), it has been shown that the presence of dissolved iron corrosion products caused very high corrosion of Al, whereas galvanic coupling with stainless steel or alloy C-4 did not cause a significant increase in the corrosion rate. The presence of iron also increased the radionuclide release of both α and γ emitters by two orders of magnitude. Finally, the corrosion tests should include actual fuel materials in addition to depleted U-Al alloys as simulants because of the great dependence of the dissolution behavior on microstructure and radiolysis.

In the case of the melt-dilute option, the partitioning of radionuclides occurs between alloy, slag, and the offgas. The DOE reports do not provide detailed information on partitioning, which may be important if the slag and alloy are cast in the same canister and if filters used to collect the radionuclides in the offgas are also disposed in a like manner. The partitioning of the radionuclides is still being investigated by SRS. The Product Consistency Test (PCT) is mentioned as a PA test. PCT, which was conceived as a test to determine the consistency of vitrified waste form, is not a PA test. Furthermore, the test measures the release of Na, Li, and B which are absent in the melt-dilute waste forms. Since the melt-dilute waste form is metallic, tests appropriate to direct/co-disposal waste fuels can be used.

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1 INTRODUCTION

In accordance with the Nuclear Waste Policy Act of 1982, the U.S. Department of Energy (DOE) is responsible for the interim management and ultimate disposal of government owned spent nuclear fuel (SNF), which includes the foreign and domestic Al-based research reactor fuels. In 1995, the DOE decided to process and store the Al-based fuels in sealed canisters at the Savannah River Site (SRS) until they can be disposed at a geologic repository. This storage configuration is termed "road ready" by the DOE. Al-based research reactor fuels and targets are a small part (less than 1 percent by volume) of the total inventory of SNF and high-level waste (HLW) to be disposed in a geologic repository. Among the thirty-two categories of DOE fuels totaling about 2,430 metric tons of heavy metal (MTHM), SRS Al-clad fuels belong to seven groups for the purpose of PA (Duguid et al., 1997). A total of 266 m^3 (23.8 MTHM) of Al-based SNF is anticipated to be sent to SRS for processing by the year 2035 (WSRC, 1997b). These fuels are slated to arrive from domestic and foreign research reactors as well as from other DOE sites. Al-based fuels at various sites are currently stored underwater, but continued storage under these conditions is not desirable because of the susceptibility to corrosion of the cladding and fuel. While these fuels constitute only a small fraction of the total amount of SNF to be emplaced in the proposed geologic repository at Yucca Mountain (YM), disposition of these fuels, including treatment, temporary storage, and permanent disposal, presents a significant challenge because of the diverse geometries, high enrichments (20 to greater than 90 percent U^{235}), and the complex metallurgical characteristics of the fuel.

The DOE Office of Spent Fuel Management chartered the Research Reactor SNF Task Team to assist in the development of a technical strategy for interim management and ultimate disposition of research reactor SNF. In 1996, the task team produced a report (U.S. Department of Energy, 1996) that identified the various types of Al-based SNF, the broad issues involved in storage and disposal, and recommended that the DOE should proceed with the parallel development of two treatment technologies: direct disposal (or co-disposal) in a repository, and melt and dilute followed by disposal. The direct/co-disposal option entails placing the SNF in type 316L stainless steel canisters that will be emplaced along with HLW glass canisters in a disposal overpack. The melt and dilute option would involve melting an appropriate mixture of Al-based SNF with depleted uranium and aluminum to achieve the desired composition and enrichment, followed by co-disposal with glass waste canisters. The task team also recommended that the DOE should utilize the existing SRS processing capabilities and should begin to work immediately with the Nuclear Regulatory Commission (NRC) and other regulatory agencies to reach agreement on SNF disposal requirements.

A memorandum of understanding and Interagency Agreement (IA) between the NRC and the DOE were established for the NRC to assist the DOE in identifying potential technical issues associated with the ultimate disposition of these fuels (letter from N.K. Stablein to L. Watkins, dated August 28, 1997). This report provides an evaluation of the technical and regulatory issues related to the final disposal of the Al-based SNF. The processing, transportation, and interim storage safety issues that are related to the requirements in 10 CFR Parts 70, 71, and 72 are outside the scope of this review. It is also not the purpose of this report to provide input to the selection of one treatment technology over the other. The evaluation of technical issues relevant to disposal of the Al-clad fuels is performed with reference to the regulatory requirements in 10 CFR Part 60:

10 CFR 60.113: Performance of particular barriers after permanent closure
10 CFR 60.131: General design criteria for the geologic repository operations area
10 CFR 60.135: Design criteria for the waste package and its components

However, it is anticipated that the NRC regulatory requirements, especially in terms of postclosure subsystem performance requirements, will change as a result of pending legislation and the National Academy of Science (NAS) recommendations. Hence, the focus of postclosure performance evaluation will be criticality and dose to an individual at a selected location.

Of the Al-based SNF, the Materials and Test Reactor (MTR) fuel forms the major fraction, accounting for about 180 m^3 or 71 percent by volume (U.S. Department of Energy, 1996). This volume includes the 36 m^3 (3.31 MTHM) of Advanced Test Reactor (ATR) fuels that will be received from the Idaho National Engineering and Environment Lab (INEEL) (U.S. Department of Energy, 1997b). The design of the MTR/ATR fuels consists of flat or curved plates of U-Al, UO_2-Al, or U-Si alloy clad with aluminum.. The total thickness of the fuel element is 1.27 mm, the cladding being 0.38 mm and fuel (meat) being 0.51 mm thick. The plates are stacked 19 to 25 high with a gap between plates of about 1.9 mm. There are variations within this general design. For example, the Massachusetts Institute of Technology (MIT) fuel is thicker (2.54 mm including cladding and fins) and 14 fuel plates are stacked in a rhombohedral cross section (CRWMS M&O, 1997a). The gap between fuel plates is about 1.4 mm. The Oak Ridge SNF consists of 19 curved plates of 1.27 mm thickness. The other significant fuel types are the High Flux Isotope Reactor and the Reactor a-Haut Flux fuels which involve dual annulus, involute plate designs (U.S. Department of Energy, 1996). The involute plates are 1.27 mm thick with a 1.27-mm coolant gap. However, the detailed analyses presented to date have focused on the MTR/ATR fuel design. Hence, the review presented in this report will concentrate on the MTR/ATR fuels.

Since 1997, several reports have been produced by the SRS team pertaining to the two alternate technologies. The evaluation of co-disposal viability and the alternative SNF treatment technology reports (CRWMS M&O, 1997a; WSRC, 1997a,b) are the three main reports that provide an overview of the current status of the alternative technologies. These reports are supported by a number of technical reports and work/test plans. In the direct/co-disposal technology option, the activities have involved the development of storage acceptance criteria, an instrumented, shielded test canister to study the drying process, thermal and criticality analyses, corrosion testing of waste form simulants in vapor and aqueous environments, and analysis of the creep and mechanical properties of the fuel cladding (WSRC, 1997b). In the melt-dilute technology option, the range of U-Al compositions is being investigated in terms of bench-scale process development. Limited corrosion tests have been performed on the waste simulants. A test protocol is being developed for corrosion testing of the direct/co-disposal fuels and the melt-dilute waste forms.

The objective of this report is to assist the NRC in identifying potential technical issues relating to the disposal of Al-based SNF in a geologic repository in order to determine whether Al-based SNF can be disposed in the geologic repository with commercial SNF and vitrified waste. The review (i) identifies repository performance issues that may affect the choice of processing technology, (ii) evaluates DOE designs of interim storage facilities as they affect performance in the repository, and (iii) assesses DOE testing and modeling methodologies related to waste form performance. The processing, transportation, and interim storage safety issues that are related to the requirements in 10 CFR Parts 70, 71, and 72 are outside the scope of this review. It is also not the purpose of this report to provide input to the selection of one treatment technology option over the other.

The subsequent chapters are divided in terms of the two main technical issues pertaining to geologic disposal: preclosure operations, and postclosure performance. Preclosure operations include issues related to oxidation, corrosion, and mechanical behavior of fuel, cladding and the canister during temporary storage. Included in the postclosure performance are issues related to criticality, pyrophoricity, thermal and structural characteristics, and radionuclide release.

2 PRECLOSURE OPERATIONS

The general design criteria for safety in the geologic repository operations area (GROA) are described in 10 CFR 60.131 which species that the structures, systems, and components important to safety shall be designed so that they will perform the necessary safety functions assuming occurrence of design basis events (DBE). Since the purpose of the road ready canister is to avoid handling bare fuel during waste transfer from transportation to disposal overpacks and during any waste retrieval operations involving repackaging of wastes, the integrity of the road ready package is important. If bare fuel has to be handled due to actual or suspected deterioration of the road ready canister, the potential for damage to fuel and cladding becomes important. The criticality control issues are also addressed in 10 CFR 60.131(h) as part of the design criteria for GROA. The DOE reports address criticality during the postclosure period and hence the Center for Nuclear Waste Regulatory Analyses (CNWRA) evaluation of criticality is reported in the next chapter. This chapter evaluates the effects of interim dry storage of the Al-clad fuels at SRS on preclosure operations. It is subdivided into issues, each issue being discussed in terms of the statement of the issue, a summary of the DOE technical approach and results, and the CNWRA evaluation.

2.1 DETERIORATION OF ROAD READY CANISTER DURING STORAGE

2.1.1 Statement of Issue

The Al-based fuels in the direct/co-disposal technology option may be stored at SRS for periods up to 50 years (a period of 100 years is mentioned in some documents) prior to disposal at a geologic repository, although the intended design life of the dry storage canister is 40 years (WSRC, 1997b). The detailed design and method of loading of the SNF into the storage canisters have not been decided at this time. It is expected that the fuels will be placed in a road ready canister made of type 316L stainless steel and sealed by welding so that preclosure operations involving waste transfer from transportation to disposal overpacks will not involve handling bare fuel. The fuel will be placed in a basket structure made of carbon steel or stainless steel containing appropriate neutron absorbers. Interim dry storage can affect waste transfer operations through the deterioration of the road ready canister due to either internal corrosion or thermal embrittlement of the welds

2.1.2 DOE Technical Approach and Results

The DOE has mainly focused on the effect of interim storage conditions on the fuel and cladding performance. An instrumented canister, made of type 304 stainless steel and loaded with MTR fuel assembly, has been assembled to test the environment inside the canister during storage (temperature, pressure, and relative humidity) and fuel performance (WSRC, 1997b). After loading the fuel assembly and vacuum drying, a controlled amount of water (unspecified in the report) will be reinjected into the canister. However, the instrumented canister is not intended to monitor the response of the canister material to dry storage conditions. Additionally, no provision is made to sample the chemistry of water collecting inside the canister in order to evaluate potential corrosion processes.

2.1.3 CNWRA Evaluation

Significant internal corrosion of the road ready canister is not expected if the canister is vacuum dried and the temperature is maintained sufficiently high to prevent condensation. However, depending on the internal structure and the presence of corrosion products on the fuel assemblies, water can be retained even at relatively low relative humidity values. For example, small pores present in the corrosion products or hygroscopic corrosion products can enable retention of aqueous environment in the canister. Assuming that a maximum of 30 g of water is allowed per canister [based on hydrogen generation (WSRC, 1997b)] the concentration of fission products, essentially I^{129} can be estimated. Assuming an iodine inventory per assembly of 5 g (WSRC, 1997b), approximately 0.5 kg of U^{235} per assembly, quick release fraction of 0.01 of iodine, a loading of 14 kg of U^{235} per canister, and complete solubility, one can estimate the aqueous iodine concentration to be about 0.4 molar. The actual concentration may be less because not all the I^{129} released will go into solution and some of the iodine may combine with Cs. This concentration, in conjunction with radiolytically generated oxidizing species, can be sufficient to induce localized corrosion of stainless steel (Koch, 1995), especially in weldments which are not as corrosion resistant as the base metal. Internal corrosion of the welds may result in the breach of road ready canister and hence expose the bare fuel during waste handling. The tests conducted on instrumented canisters should include monitoring the internal corrosion of the canister and measuring condensed water chemistry.

Thermal embrittlement of type 316L weldments under storage conditions due to precipitation of alpha-prime phase in the weld ferrite is not considered to be kinetically significant (Sridhar et al., 1994). Since the weld ferrite in type 316L stainless steels is discontinuous, any embrittlement of the ferrite is unlikely to reduce the overall fracture toughness of the weld significantly.

2.2 OXIDATION AND CORROSION OF CLADDING AND FUEL IN VAPOR ENVIRONMENT

2.2.1 Statement of Issue

The storage system must be designed such that humid air corrosion or oxidation of the cladding and fuel during storage does not impair the ability to transfer the fuel, if it becomes necessary, at any time prior to permanent closure of the repository. Here, humid air corrosion encompasses corrosion in a water vapor environment where a thin film of condensate forms (i.e., at relative humidity values where the formation of several monolayers of water occurs). Oxidation refers to degradation under dry conditions where no condensed layer of water exists on the surface or, at most, a monolayer of water is stabilized. The oxidation may impair retrievability due to volume expansion inside the canister or brittleness and spallation of the oxidation products. Humid air corrosion (general or localized) may impair retrievability by generating mobile corrosion products or by the loss of strength and breakage of the fuel bundle during handling. Oxidation is not an important issue for the melt and dilute waste form because the waste form fills all the space inside the canister.

2.2.2 DOE Technical Approach and Results

The Al-based fuels that are received at SRS will be placed in Type 316L dry storage canisters under the direct co-disposal and melt and dilute options. The fuels are considered to be able to be handled

mechanically, even though about 10 percent have already been damaged by corrosion or mechanical forces (WSRC, 1997a,b). Hence in the direct co-disposal option, they will be placed in the storage canisters without further canning. The DOE has performed vapor phase corrosion studies of various Al alloys that simulate the Al-cladding and, to a lesser extent, on depleted U-Al alloys to simulate fuels (WSRC, 1997b; Lam et al., 1997). Vapor corrosion tests were performed under conditions of unlimited water (in a 1-gal. stainless steel autoclave where the condensate was replenished at various time intervals) and limited water (in a sealed 78 ml capsule) at a variety of calculated initial relative humidities. The water used for these tests was obtained by condensing the SRS atmosphere using a cooling coil. The initial analysis of the condensate (Lam et al., 1997) indicated a chloride concentration of 446 to 3,560 ppm by weight, sulfate of 1,360 to 7,670 ppm, calcium of 577 to 4,500 ppm, magnesium of 198 to 4,350 ppm, sodium of 1076 to 7891 ppm, and the pH ranged from 5.9 to 7.9. Post test analysis of the water indicated considerable concentration of the chloride (8,290 to 41,600 ppm), sulfate (12,800 to 55,000 ppm), calcium (2,140 to 22,174 ppm), magnesium (340 to 14,407 ppm), sodium (11,855 to 55,660 ppm), but no significant change in pH, suggesting significant evaporation of water or incorporation into hydrated corrosion products during the test. The lack of increase in pH due to evaporation may have been due to the hydrolysis of magnesium cations. Tests were also conducted using the condensate water modified by the addition of nitric acid to simulate radiolysis products of air-water mixture and condensate water under gamma irradiation conditions (Lam et al., 1997).

Breakaway corrosion, defined as the change in the oxidation kinetics from a parabolic to a linear rate, was seen for Al 5052 alloy (Al–0.1 wt%, Zn–2.5%, Mg) at 200 °C and 150 °C after 2,000 and 16,000 hours respectively, but not for alloys Al 1100 (commercially pure Al) and Al 6061 (Al–0.25%, Zn–1%, Mg–0.6%, Si). The breakaway corrosion was attributed to internal oxidation along grain boundaries due to the presence of Mg in alloy Al 5052. When nitric acid was added to the condensate water, the vapor phase corrosion was two orders of magnitude higher for all three alloys and pitting was noted for alloy Al 5052. In capsule tests with limited water availability, weight-gain was observed at relative humidities as low as 20 percent, indicating that the critical relative humidity was probably less than this value. Since tests were carried out for only 9,000 hours at temperatures up to 150 °C , it is difficult to ascertain whether breakaway corrosion can occur under these circumstances and lack of availability of sufficient corrodent would limit corrosion. Since only weight gain was reported and not depth of corrosion, it is not possible to determine whether continued penetration of the cladding by oxidation can occur even though the total weight gain may be arrested by limited water. The corrosion rate was higher when acid was added to the condensate water. The corrosion rate was also higher when J-13 water (i.e., water from a well near the proposed repository at YM) was used instead of condensate water. Since the initial chloride and sulfate concentration of J-13 water are about 6 and 20 ppm, respectively, compared to the 3,560 and 7,670 ppm, respectively of the condensate water, the high corrosion rate in J-13 water cannot be attributed to chloride or sulfate concentration. It is possible that the evolution of the pH during the evaporation of J-13 water to slightly alkaline values (9.5 to 10) may have increased the corrosion rate of Al, whereas the pH of the condensate water seems to have been buffered by the presence of Mg and Ca.

Tests at various temperatures were also performed in capsules under gamma radiation field (Lam et al., 1997). These tests were carried out for about 1,440 hours and indicated that gamma radiation enhanced corrosion significantly. Significant blistering of the surfaces of the three alloys was noted under gamma radiation field in contrast to the fairly smooth oxide (boehmite) layer on the unirradiated specimens exposed to a similar environment. At least in alloy Al 5052, the internal oxidation observed under breakaway corrosion in an unirradiated environment appears similar to the internal oxidation at early times under gamma radiation conditions. The blisters observed on the surface seem to be indications of internal oxidation in which boehmite crystals are formed under the external surface. Since, the depth of internal oxidation was shown only for 1 and 12 weeks, it is impossible to ascertain whether continued internal oxidation occurred

after 8 weeks despite lack of weight gain. The depth of internal oxidation after 12 weeks at 200 °C in this alloy was about 120 microns (the cladding thickness is about 380 microns). In alloys Al 1100 and Al 6061, internal oxidation was not observed under gamma radiation, but thick oxides that spalled may have led to high corrosion rates.

Much more limited study of the fuel material under vapor conditions has been conducted (WSRC, 1997b). Tests have been conducted on U-10 wt% Al alloy and tests are planned on U-18 wt% Al and U-33 wt% Al alloys that are more representative of the fuel materials awaiting disposal (WSRC, 1997b). All these tests are on alloys containing depleted U and, hence, do not involve radiation effects of the fuel materials. The tests conducted to date on U-10 percent Al alloy have involved hot-rolled sheet and extruded rod materials, and significant differences in the corrosion rates have been observed between these, indicating the effect of second phase distribution and morphology on corrosion (Lam et al., 1997). Tests have been conducted only in air that was 100 percent saturated with water vapor at 200 °C under conditions of unlimited corrodent availability. For the hot-rolled material, the corrosion rates are at least four times higher than for the extruded material. The corrosion rates are comparable to that of Al 1100. The test times of 1,700 hours are too short to indicate breakaway corrosion. Detailed microstructural investigation of the corrosion modes has not been reported.

Planned tests of cladding materials include tests in vapors in equilibrium with J-13 water, tests at 250 °C without radiation effects, and tests at 200 and 78 °C with gamma radiation. Planned tests for the U-19 wt% Al, chosen as the representative material for all the fuels, include tests in vapors in equilibrium with J-13 water up to 200 °C (with and without radiation).

For the melt and dilute option, vapor corrosion tests have not been reported. However, judging from the reported microstructures of the various compositions (WSRC, 1997b), the corrosion modes may be similar to the fuel materials in the co-disposal option. Since the cross-sectional size of the melt and dilute waste form is expected to be greater than that of the co-disposal fuel, the preclosure mechanical stability may not be adversely affected by corrosion.

2.2.3 CNWRA Evaluation

Preclosure performance of fuel and cladding is important only if handling bare fuel is necessary during waste transfer operations at the GROA. Bare fuel handling may be necessary if the road ready canister is breached during waste transfer operations. While considerable amount of testing of the cladding has taken place, providing valuable insight into the behavior of these materials under vapor phase corrosion conditions, they do not appear to be sufficient to determine whether corrosion under steam or water vapor would deteriorate the cladding sufficiently to prevent safe waste transfer, if necessary. For example, under limited water availability, it was shown that the weight gain would stabilize after some time period. However, these data are not sufficient to conclude whether penetration of cladding by oxidation would be arrested because (i) tests have not been carried out for a sufficiently long time (a few thousand hours at most), (ii) tests under radiation conditions may result in breakaway corrosion at longer time periods, and (iii) metallographic examinations of the corroded specimens have not been reported to ascertain whether internal oxidation would arrest once weight gain stopped. Indeed, long-term autoclave tests at 250 °C (WSRC, 1997b) showed that the boehmite (hydrated aluminum oxide) was transformed to corundum (anhydrous oxide) suggesting that water may be released to continue further oxidation internally. Based on the data reported to date, a bounding assessment of the oxidation or humid-air corrosion of cladding and fuel cannot be performed. The tests performed to date indicate the importance of water chemistry and radiolysis on vapor corrosion, but do not

provide sufficient guidance on estimating the degree of oxidation or penetration of the cladding and fuel under dry storage conditions. For example, it is unclear what the chemistry of the environment would be in the dry storage cask. Because the fuels will be transferred to the road ready packages in the spent fuel pool, the initial water chemistry is likely to be related to pool water chemistry rather than condensate water chemistry. The internal water chemistry may also be influenced by fission products such as iodine. The J-13 water chemistry is irrelevant to preclosure and retrieval performance because this is the chemistry of water in the saturated zone in the vicinity of the YM repository. The relevance of vapor corrosion for postclosure performance will be evaluated in another section.

2.3 CREEP OF THE CLADDING AND FUEL

2.3.1 Statement of Issue

Creep of cladding and fuel may lead to sloughing of the fuel plates or rupture. This may affect transfer of the fuel from the co-disposal canister, if deemed necessary. Since a certain spacing between fuel plates is assumed to avoid criticality during storage, slumping of the plates may affect criticality adversely. It may also affect postclosure performance of the waste form, which will be addressed in a later section.

2.3.2 DOE Technical Approach and Results

The DOE has performed a finite element analysis using ABAQUS, Version 5.4-1 and assuming Coble creep as the operating mechanism under the anticipated low stress (due to the weight of the fuel plates) and temperature (Miller and Sindelar, 1995; WSRC, 1997b). The stress on the cladding was calculated to be 89.6 kPa (13 psi) due to gravity loading alone and 10s of Mpa (several thousand psi) due to differential thermal expansion of the fuel and cladding during the heat-up phase (Miller and Sindelar, 1995). The authors note that these high-differential thermal stresses decrease to the gravitational stresses within approximately 1 month. Hence, Coble creep with a normalized stress (actual stress divided by the shear modulus of the material) of about 10^{-6} and a homologous temperature (actual temperature divided by melting point in absolute scale) of about 0.5 was assumed from the deformation map for pure aluminum. The deformation behavior of the fuel element was ignored based on the assumption that the creep rate of the fuel will be constrained by the creep of the cladding. Based on these assumptions, the authors calculated that the minimum temperature for exceeding a deflection of 2.54 mm (0.1 in.) was 150 °C for a grain size of 25 microns, 200 °C for a grain size of 50 microns, and 250 °C for a grain size of 100 microns. It was then argued that the microstructure of some of the cladding material showed a minimum grain size of 50 microns and, hence, a limit of 200 °C was deemed to be safe for dry storage. The authors also compared their analytical results with an independent analysis of the creep of ATR fuels, as well as literature data on creep of pure Al at 250 °C, and found their calculations to be conservative.

2.3.3 CNWRA Evaluation

The creep analysis performed by SRS (Miller and Sindelar, 1995) is conservative in some respects. It assumes a relatively high gravitational stress, assumes the behavior for pure Al rather than for a Al-U solid

solution which may be expected to creep less, and assumes a constant temperature of 200 °C. However, many uncertainties remain that may render this analysis nonconservative:

- As the authors acknowledged, the differential thermal expansion stresses between fuel and cladding may be several orders of magnitude higher than the gravitational stresses, at least initially, and may lead to accelerated creep. Additionally, Ashby et al. (1979) have indicated that, for a normalized stress of 2×10^{-4} and homologous temperature of 0.52, commercially pure Al can suffer intergranular creep fracture in 1.5 months. The data on which they based their analyses are rather old and further experimental study is needed to ascertain this conclusion.

- The assumption that a grain size of 50 microns is the minimum grain size for all cladding materials may be questionable. For example, an inspection of the internal oxidation structure of alloy Al 5052 published in WSRC (1997b, Figure 27b) indicates a wide range of grain sizes from less than 10 micron to greater than 50 microns.

Hence, if a 200 °C criterion is used for dry storage, further analysis of the creep and stress rupture behavior of cladding and the fuel package may be necessary. Examination of a lower temperature criterion (e.g., 150 °C) is also recommended.

2.4 STRUCTURAL INTEGRITY OF INTERIM STORAGE WASTE PACKAGES

2.4.1 Statement of Issue

Structural analyses of the co-disposal waste package (WP) with all relevant fuel assembly configurations must be conducted for all credible preclosure DBEs to demonstrate that the structural integrity of WP, canisters, and waste forms will be maintained during handling, emplacement operations, preclosure period storage, disposal, and retrieval operations. These analyses must show that the structural strength of the co-disposal WP and canisters is adequate under all credible preclosure scenarios to (i) prevent the fuel from being damaged, (ii) conduct emplacement operations, and (iii) conduct waste transfer operations from interim casks, transportation casks, and disposal casks. The credible preclosure DBEs to be considered must be consistent with the techniques to be used for handling, emplacement operations, preclosure storage, and retrieval operations, and the site-specific environment that may exist during the preclosure period. In the structural analyses, the effect of prolonged thermal exposure on the mechanical properties of the cladding and fuel material needs to be considered.

2.4.2 DOE Technical Approach and Results

The DOE has selected the MIT reactor and Oak Ridge Research (ORR) reactor SNF co-disposal canisters as representative canisters for inclusion in the co-disposal WPs (CRWMS M&O, 1997a). The DOE has conducted structural analyses involving only MIT SNF co-disposal canisters (CRWMS M&O, 1997b). These analyses were based on both actual and assumed input data and a variety of reasonable simplifying assumptions.

The DOE has developed a list of preclosure DBEs for Viability Assessment design of WPs (CRWMS M&O, 1997c). This list has been developed based on a four-step systematic approach. These

DBEs include, among others, various amounts of rock falling on WPs from various heights, vertical and horizontal drops of WPs, and WP tip-over from a vertical position. Using the finite element code ANSYS 5.1, the MIT SNF co-disposal canister was analyzed for dynamic impact loads due to only a tip-over event after being placed into a 5-canister defense high-level waste (DHLW) WP. The WP tip-over was from a position of 35 degrees from the vertical. It was assumed that the external load on the MIT SNF co-disposal canister was the dynamic impact load from only one DHLW canister acting at an angle of 45 degrees with the axis of the MIT SNF co-disposal canister. This impact load was simulated by applying nodal forces at the points of contact between the DHLW canister and the MIT SNF co-disposal canister. The other load considered to act on the MIT SNF co-disposal canister was the effect of gravitational acceleration due to the WP tip-over.

The results of these analyses were used to determine the locations where the displacements were large enough to cause the fuel assemblies to deform. The results of the analyses were also evaluated in terms of the maximum stress contours to determine if the co-disposal canister stresses exceeded the material yield or ultimate tensile strength. It was found that the structural strength of the MIT SNF co-disposal canister basket is adequate to prevent the fuel from being damaged in a WP tip-over accident, although some localized plastic deformations might occur within the basket structure at a high stress area.

While the structural analyses assumed the mechanical properties of annealed cladding and fuel materials, there is a potential for the mechanical properties to be degraded by thermal exposure. The effect of thermal exposure and neutron irradiation on the ductility of alloys Al 1100 and Al 6061 were reviewed by Caskey (1993). While the ductility of irradiated material can decrease to very low values upon thermal exposure to temperatures in the 150 to 200 °C regime, it was argued that the annealing of the previously irradiated cladding material during dry storage would restore the unirradiated material properties (Caskey, 1993). It was also argued that the anticipated stresses were low and, hence, a 200 °C limit was reasonably conservative.

2.4.3 CNWRA Evaluation

The DOE analyses predict only the performance of a MIT SNF co-disposal canister under a co-disposal WP tip-over accident from a position of 35 degrees from the vertical. For these analyses, the DOE did not consider other credible preclosure DBEs such as rockfall on WPs and vertical and horizontal drops of WPs as identified by CRWMS M&O (1997c), and did not provide any bases for not considering those credible preclosure DBEs. Thus the selection by the DOE of WP tip-over as the bounding preclosure DBE without considering other credible preclosure DBEs is inadequate. In these analyses, the assumptions such as (i) only one DHLW canister impacts the co-disposal canister, (ii) the tip-over dynamic load generated on a 6-canister co-disposal WP is the same as that generated on a 4-canister DHLW WP, and (iii) the Savannah River HLW canister is representative of the DHLW canister, may provide sources of nonconservatism. Furthermore, these analyses do not include co-disposal canisters for ORR SNF or other fuel types.

The effect on mechanical properties of prolonged exposure to 200 °C of previously irradiated aluminum alloys need further examination. While the stresses expected from gravitational loading may be quite small, impact loading from canister tip-over or other DBEs may result in the breakage of the fuel elements, thus impairing the ability to transfer wastes safely during the preclosure period. Alternatively, a lower storage temperature limit may need to be considered.

2.5 ACCEPTANCE CRITERIA FOR INTERIM DRY STORAGE

2.5.1 Statement of Issue

The SRS has established acceptance criteria for interim dry storage of three classes of Al-based SNF. The acceptance criteria are defined as the combined set of environmental limits for drying and storage to avoid excessive degradation of the fuels during a nominal 50-year storage period (Sindelar et al., 1996). For all types of fuels, the temperature limit specified is 200 °C. The maximum expected degradation under these conditions is stated to be less than 0.3 mil (7.6 microns) deep corrosion of cladding, negligible sloughing-off of oxide products, negligible fission product release, and less than 0.1 in. (2.54 mm) local distortion due to creep. In a subsequent document (WSRC, 1997b), the acceptance criteria are stated slightly differently. In addition to the temperature limit of 200 °C, a limit of the free water to prevent excessive hydrogen generation, a limit to general corrosion rate of 3 mils (76 microns), and a limit of creep deformation not to exceed 75 percent of space between fuel assembly and storage grid are specified (WSRC, 1997b).

2.5.2 DOE Technical Approach and Results

As mentioned before, the SRS is conducting an extensive study of the vacuum drying, and vapor corrosion of cladding and simulated fuel alloys under a variety of water chemistry and radiation conditions. Additionally, the creep and stress rupture behavior of Al has been considered.

2.5.3 CNWRA Evaluation

The 200 °C temperature limit for dry storage of 50 years may not be sufficiently conservative based on the data generated by the SRS on cladding and simulated fuel materials (WSRC, 1997b). For example, based on the measured weight gain and metal-loss data, the time to penetrate 3.0 mils of alloy Al 1100 cladding at 200 °C is greater than 50 years. However, for the alloy Al 5052 cladding material, the time to penetrate 3 mils is less than 50 years without breakaway corrosion, whereas if breakaway corrosion occurs after 1,400 hours at 200 °C, the time to penetrate 3 mils is less than 1 year. Furthermore, under breakaway corrosion conditions (WSRC, 1997b), the cladding will be completely penetrated in 1 year. Corrosion in a water vapor environment is mitigated by the pressurization of the storage canister with helium (intended for diluting the hydrogen to below the flammability limit) but, as pointed out previously, insufficient data exist on penetration of the cladding under limited water and radiolysis conditions.

The acceptance criteria do not address Al-based fuels that will be received from INEEL that are known to be corroded significantly (Guenther et al., 1995).

2.6 PRECLOSURE THERMAL CONDITIONS OF CO-DISPOSAL WASTE PACKAGE IN THE REPOSITORY

2.6.1 Statement of Issue

Thermal analyses of the co-disposal WP with all relevant fuel assembly configurations must be conducted with appropriate and consistent thermal input data to demonstrate that the DOE temperature limit goals on different components (HLW glass and its canister, MIT SNF and its canister, and co-disposal WP

shell) of the co-disposal WP have not been exceeded. These analyses must show that the calculated temperatures are such that they will not adversely affect the mechanical properties of WP and canister materials and behavior of Al-based SNF.

2.6.2 DOE Technical Approach and Results

Two dimensional (2-D) steady-state finite element thermal analyses were conducted on the co-disposal WP involving the MIT SNF co-disposal canister [CRWMS M&O (1997a)]. The analyses used the previously determined surface temperatures of the DHLW WP with four HLW canisters (Bahney, 1996) as the boundary conditions for the co-disposal WP, and applied the heat loads in the DHLW canisters and MIT SNF co-disposal canister to determine the internal temperature distribution at various times from emplacement out to 100 years. The analyses were conducted for a thermal load of 83 MTU/acre. The effects of drift backfilling were not considered for this analysis. The DOE has stated that there were inconsistencies in the thermal input data and has recommended another round of thermal analyses when the consistent thermal input data become available (CRWMS M&O, 1997a).

The calculations of the surface temperatures of DHLW WP with four HLW canisters considered WPs in the drift with different WP heat generation rates (Bahney, 1996). In this arrangement, the DHLW WPs alternate in position along the drift axis with the commercial SNF WPs. The spacings between the WPs varied to account for each WP MTU loading to provide a thermal load of 83 MTU/acre (CRWMS M&O, 1997a). Three dimensional (3-D) transient finite element analyses of the DHLW WP with four HLW canisters were conducted to calculate the surface temperatures of the DHLW WP that were used as the boundary conditions for the 2-D co-disposal WP.

On the basis of the preliminary analysis results, the DOE concluded (CRWMS M&O, 1997a) that the peak temperatures of the glass matrix of DHLW canister and the SNF of MIT co-disposal canister are below the DOE temperature limit goals and that the calculated temperatures of the materials used in the co-disposal WP are such that melting or rapid mechanical failure will not occur. Therefore, the proposed co-disposal canister can be loaded in the co-disposal WP.

2.6.3 CNWRA Evaluation

The analyses are based on a number of reasonable assumptions and idealizations regarding mode of heat transfer, effective thermal conductivity, convective cooling by helium, and modeling of fuel assembly internals. Since the peak temperatures at different components (HLW glass and its canister, MIT SNF and its canister, and co-disposal WP shell) of the co-disposal WP occur during the preclosure period, the preliminary analyses based on the DOE stated inconsistent thermal input data are not adequate to make the conclusion that the proposed co-disposal canister can be loaded in the co-disposal WP. The DOE needs to identify the inconsistencies in the thermal input data and conduct the thermal analyses with consistent thermal input data. In addition, the use of surface temperatures of DHLW WP with four HLW canisters as the boundary conditions for the co-disposal WP with five HLW canisters underestimated the temperatures of HLW glass and its canister, and MIT SNF and its canister. Thus the thermal analyses need to be done using consistent input data and correct boundary conditions to confirm that, during the preclosure period, the peak temperatures will not exceed the DOE temperature limit goals, that there will be no changes of mechanical properties of co-disposal WP that will adversely affect waste transfer operations, and that the behavior of Al-based SNF will not be affected adversely.

3 POSTCLOSURE PERFORMANCE

The current postclosure performance requirements are described in 10 CFR 60.112 for overall system performance requirement consistent with the U.S. Environmental Protection Agency (EPA) standard and 10 CFR 60.113, which provides for sub-system requirements (such as radionuclide release rates after container breach). In addition, according to 10 CFR 60.135, the WP and its components shall be designed considering factors such as solubility, corrosion reactions, hydriding, gas generation, thermal effects, mechanical strength, pyrophoric behavior, and explosion hazards. The criticality control issues are addressed in 10 CFR 60.131(h) as part of the design criteria for GROA. It is anticipated that the postclosure performance requirements will change in response to the pending legislation and the NAS recommendations and associated revisions to the EPA standard. The focus of the new NRC regulations may be dose to the critical group at a specified location considering the overall system performance, although performance of WPs will continue to be important in terms of a defense-in-depth approach. The CNWRA evaluation of DOE reports addressed the overall system performance in terms of source term issues. Waste form design issues were also evaluated. The DOE reports address criticality during the postclosure period and, hence, the CNWRA evaluation of criticality is also reported in this chapter.

3.1 CRITICALITY

The criticality requirement is described in the general design criteria for the GROA in 10 CFR 60.131. This section assesses the criticality calculations performed by the DOE. Criticality is more of a concern for direct co-disposal than for melt and dilute, but criticality calculations for both technologies are reviewed where available.

3.1.1 Criticality Safety for the Direct Disposal Option

3.1.1.1 Statement of Issue

Criticality calculations must be performed for the direct disposal of Al-clad fuels in co-disposal containers. These calculations must show that nuclear criticality is not possible unless two independent, unlikely, and concurrent or sequential events occur. The canister must be designed for criticality safety assuming occurrence of DBEs. The calculated multiplication factor must be sufficiently below unity to show a 5 percent margin of safety after accounting for calculational code bias and uncertainty.

3.1.1.2 DOE Technical Approach and Results

The DOE has run a series of criticality calculations for the co-disposal canister varying the fuel type, state of degradation of the fuel and canister, and the quantity of water in the canister. Two types of fuels were evaluated as bounding case scenarios: MIT reactor fuel that is enriched to 93.5 percent U^{235} and ORR fuel that is enriched to 20.56 percent U^{235}. Calculations were performed for three states of degradation of the fuel and canister: (i) intact fuel within the co-disposal canister; (ii) degraded highly enriched uranium (HEU) fuel within an intact co-disposal canister; and (iii) degraded HEU fuel, HLW glass, and canister material within the WP. The potential for a criticality event after reconfiguration of fuel outside of the WP is currently not being considered. Future calculations for the potential for a criticality event outside of the WP should consider at least the following two scenarios: (i) reconfiguration of the fuel outside of the WP after leaching out of the package; and (ii) the potential for disruptive events, such as fault slips or volcanism, being able

to breach canisters and separate fuel from the canister (and the neutron poisons) in a selectively intact state. The degradation of the fuel and basket within the co-disposal canister was modeled in three steps to determine the most reactive configuration. First, the fuel plates were homogenized with the interplate moderator volume. Second, the entire assembly, which consists of the fuel plates, structural combs, and water, was homogenized. Third, this homogenized material was dispersed throughout the free space within the basket.

Several assumptions were made in the evaluation of the potential for criticality of the fuel after emplacement in the repository. The co-disposal canister is assumed to be filled with as many assemblies as is physically possible. No credit is taken in the calculations for burnup of the fuel, so the fuel is assumed to be in its most reactive state. However, it was assumed that the fuel consisted of 1 percent U-234. The quantity of water in the cask is evaluated from no water to fully flooded to determine the most reactive condition. To account for the spatial variation of the quantity of poison materials, only 75 percent of the design quantity of poison materials are taken credit for, as no measurements will be made to ensure content and uniform coverage.

The fuel canister was modeled using the Monte Carlo N Particle Transport Code (MCNP) 4A code (Briesmeier, 1993). The MCNP model consisted of an exact model of the most reactive fuel within the co-disposal canister. The MCNP model of the fuel utilized the maximum widths and lengths of the fuel plates and the minimum thicknesses of the aluminum cladding components to maximize the fuel and moderator volume. The steel walls of the HLW canisters were not included in the modeling to allow for maximum neutronic interaction between the HLW and the co-disposal canister fuel. For the MIT fuel, neutron poisons will be incorporated into the plates between rows of fuel assemblies, the plates between individual fuel assemblies within a single row, and the plates that lie between axial layers of assemblies. For the ORR fuel, neutron poisons will be incorporated only into the plates that lie between axial layers of assemblies. The modeling to date has used borated steel as the neutron absorbing material, but the DOE has indicated that gadolinium is under consideration to become the primary neutron poison.

Results from these analyses show that neutron absorbers are necessary to ensure criticality safety. Intact MIT fuel has been explicitly modeled to show that it will remain adequately subcritical with neutron poisons only in the plates that lie between axial layers of assemblies and the plates between rows of fuel assemblies. The maximum calculated neutron multiplication factor was 0.90295 for fully flooded fuel after accounting for code bias and uncertainty. Intact ORR fuel has also been shown to remain subcritical with neutron absorbing material only in the plates between axial layers of fuel. The maximum calculated neutron multiplication factor was 0.89724 for fully flooded fuel after accounting for code bias and uncertainty.

Within the co-disposal canister, degraded MIT fuel has been shown to remain adequately subcritical with neutron absorbing material in all the plates mentioned above. The maximum calculated neutron multiplication factor was 0.94933 after accounting for code uncertainty and bias for the scenario where the entire cell is homogenized to fill the axial space between separator plates. However, if the neutron absorbers are removed from the plates between assemblies within a single row, this value has been shown to be 1.01618, which means that the fuel would be in a supercritical state. For the ORR fuel, it has been shown that only the plates that lie between axial layers of fuel assemblies are necessary to keep either intact or degraded fuel adequately subcritical. The maximum calculated neutron multiplication factor after accounting for code bias and uncertainty for degraded fuel was 0.93713. However, if the neutron absorbing material is eliminated from the axial plates, this value has been shown to be 0.967, which does not allow for a 5 percent margin of criticality safety.

The degradation of both the HEU fuel and the HLW glass within the WP has been evaluated by the DOE. Three possible degradation scenarios were evaluated based on the amount of mixing the HEU fuel with the HLW glass. The first scenario assumed that the degraded co-disposal canister did not mix with the HLW glass form and occupied the bottom of the fuel cask, with the degraded HLW glass above it. The second scenario assumed that the degraded co-disposal canister did not mix with the HLW glass, but the degraded HLW glass occupied the bottom of the fuel cask, with the degraded co-disposal canister above it. The final scenario assumed complete mixing and homogenization of the degraded HEU fuel and the degraded HLW glass within the fuel cask. The DOE has not provided these calculations to the NRC, but has stated that the results of the calculations show that the ORR fuel remains subcritical in all configurations and the MIT fuel requires the presence of 0.20 kg of Gd homogeneously distributed with the fuel to remain adequately subcritical in the most reactive configuration, which is a stratified layer of degraded MIT fuel on top of the HLW glass.

3.1.1.3 CNWRA Evaluation

Because the DOE calculations have not been provided for the complete degradation of the interior of the WP or for the reconfiguration of materials outside of the storage cask, this analysis is limited to cases in which the co-disposal canister inside the WP remained intact. The following assumptions for the criticality analysis are clearly conservative:

(i) The fuel is fresh with no burnup. Fresh fuel would provide the most reactive state by providing the greatest inventory of fissile material.

(ii) Infinite water reflectors are assumed to be present at each end of the system. Water is an excellent neutron reflector so the use of an infinite water reflector will provide the maximum neutron reflection back into the system.

(iii) Credit is taken for only 75 percent of the neutron absorber that will be initially present. This is conservative for the initial system because additional absorber material will remove more neutrons from the system and therefore lower the reactivity of the system. However, burnup of the absorber may need to be considered further, as discussed in section 3.1.3.3.

More justification is needed for the assumption that 1 percent of the uranium in the fuel is U-234. This isotope of uranium has a large thermal neutron cross section (100b) and does not undergo fission, so it acts as a fairly strong neutron poison in the fuel. Therefore, an assumption of its presence in the fuels is actually a criticality control measure and its use should be justified. There is no indication that this isotope is present in the fresh fuel in these quantities, so its presence in the calculations may indicate that credit is being taken for burnup or decay.

The calculations that were provided by the DOE limited the analysis to four cases. The first case is perhaps the simplest. This assumes that the fuel is located as placed in a basket structure and is completely flooded with pure water. This case makes no further assumptions in modeling the configuration than the global assumptions described above, and the canister and fuel are modeled explicitly. The case of the homogenized fuel assemblies also contains a conservative analysis of possible changes in geometry. Since the stainless steel remains intact, the degraded fuel would be expected to slump toward a low point in each section of the basket. This condition would create large voids which may provide for neutronic isolation. By dispersing the material evenly in the basket, the most reactive state is attained for this set of materials and

degradation state assumptions. Adequate protection is provided by the assumptions of fresh fuel, allowance for only 75 percent of the neutron absorbing material content, and the geometry assumptions. There are some questions regarding the third and fourth cases which deserve consideration. In the third case, the entire assembly—one vertical array of fuel assemblies—is homogenized. Arguably, this condition is not physically credible. However, if conditions were created in which these spaces could mix while the main separator plates remained, the analyzed condition may not be conservative. One would expect material to move with gravity into a more compact shape. Since the system appears to be larger than necessary for the fuel inventory to assure a critical shape, a more compact system could have a higher k_{eff}. Furthermore, as the material moved down, the neutron absorbing material in the separator plates would become less effective. The neutron absorbing material in the top of the plate would no longer be integral to the system. It would be helpful to discuss the effect of more compact neutronic systems.

For the final case, all structural parts of the system are assumed to have failed and mixed with the degraded fuel. At this point there are two primary concerns:

(i) Again, the system is assumed to maintain the size and shape of the storage basket. This would be reasonable only if the structural parts of the basket were assumed to perform this basic function. The effect of size on the neutronic system should be addressed.

(ii) Although it is acknowledged that segregation of neutron absorbing material might occur as a result of degradation, the impact of this migration is not addressed. Maintenance of a satisfactory reactivity margin is highly dependent on the effectiveness of the neutron absorbing material. More discussion on the potential for loss of neutron poison effectiveness should be included.

One problem found in the criticality calculations appears on page II-2 of the design analysis for criticality (CRWMS M&O, 1997e) in which a calculational error is discovered but not corrected. Rationale is provided for why an error which overstates the aluminum atom density is not likely to impact the overall result, but it would be very helpful if a modified MCNP run was performed on the worst case scenario to demonstrate this.

More discussion is needed about the fact that the Al-clad fuels have a wide variety of initial enrichments. The calculations reviewed were for bounding case type fuels, with the highly enriched MIT fuel and the lower enriched ORR fuel. The results showed that the MIT fuel required neutron absorbing material in plates between assemblies within the same row as well as axial plates, whereas the ORR fuel required only the axial neutron absorbing plates. However, the DOE does not indicate if all fuel types will contain within-row neutron absorbers (whether they are necessary or not), or if separate criticality evaluations will be performed for all fuels that have an enrichment between the two bounding cases to determine if within-row neutron absorbers are necessary. The DOE should indicate how other fuel types will be handled to ensure criticality safety after disposal.

Finally, evaluations should be done to determine if there is a potential for materials to reconfigure into a critical configuration outside of the WP, either in the repository or in the rocks beneath the repository. Documents reviewed thus far have simply indicated that these calculations will be performed elsewhere, but no specific references have been made. Future calculations of criticality outside of the WP should consider two scenarios. First, the fuel could leach out of the package and reconcentrate outside of the WP in a critical geometry. Second, disruptive scenarios, such as fault slip or volcanism, may have the ability to breach canisters and separate the fuel from the WP in a relatively intact state, creating a criticality event.

3.1.2 Criticality Safety for the Melt and Dilute Option

3.1.2.1 Statement of Issue

Criticality calculations must be performed for the disposal of Al-clad fuels after melting and diluting the fuel to 20 percent or less enrichment. These calculations must show that nuclear criticality is not possible unless two independent, unlikely, and concurrent or sequential events occur. The canister must be designed for criticality safety assuming occurrence of DBEs. The calculated multiplication factor must be sufficiently below unity to show a 5 percent margin of safety after accounting for calculational code bias and uncertainty.

3.1.2.2 DOE Technical Approach and Results

No evaluation of the potential for criticality has been performed for the disposal of the diluted fuel. It is expected that the diluted fuel will have a lower potential for criticality than the HEU fuels. If later evaluations determine that neutron poison materials are needed to ensure the subcriticality of the diluted fuel, the material will be incorporated into the alloy during melting.

3.1.2.3 CNWRA Evaluation

No evaluation can be made of the DOE approach to this issue because the DOE criticality calculations are not available. The potential for criticality of the diluted fuel, although less of a concern than for the very highly enriched fuel, should be addressed to determine if neutron absorbing material will be necessary for this option.

3.1.3 Selection of Neutron Absorbing Material

3.1.3.1 Statement of Issue

Because neutron absorbing materials have been shown to be necessary to ensure the subcriticality of the HEU fuels, the selection of the material to be used as a neutron poison is important to the safety of the system. The material selected must be able to keep the system subcritical throughout the time where significant fissile materials exists to enable the system to achieve super criticality under all states of degradation of the fuel.

3.1.3.2 DOE Technical Approach and Results

The DOE has used borated steel as their primary neutron absorbing material throughout the criticality analyses described above. However, in recent discussions, the DOE has indicated that they are considering the use of gadolinium, in the form of Gd_2O_3 or $GdPO_4$, as the primary neutron poison. Calculations by the DOE have shown that the gadolinium will remain in the WP for a much longer time than boron because of the high solubility of boron.

3.1.3.3 CNWRA Evaluation

The use of gadolinium as the primary neutron absorbing material has several advantages and disadvantages. Advantages include a much larger thermal neutron cross section and a longer lifetime inside the canister than boron. Another advantage is that when the isotopes of gadolinium with the largest thermal neutron cross sections (Gd^{155} and Gd^{157}) absorb a neutron, they simply become another isotope of gadolinium, instead of decaying to another element as boron does to lithium and helium. The thermal neutron cross sections of these newly formed isotopes of gadolinium (Gd^{156} and Gd^{158}) are much smaller than those of Gd^{155} and Gd^{157}, but they will continue to provide a small amount of neutron absorption. Also, because the reaction products of the neutron absorption are isotopes of the same element, damage to material properties of the steel support structure is less likely to occur. However, there are disadvantages to the use of gadolinium. Because of the larger cross section, burnup of the gadolinium may be a problem. The DOE should perform an evaluation of the fraction of gadolinium that will absorb neutrons over the time period of interest, effectively removing it as a neutron absorber. If a significant fraction of the gadolinium will be removed in this manner, the initial concentration of gadolinium in the steel may have to be increased to compensate for this loss. Another problem with gadolinium is that it may become soluble in acidic solutions. However, this becomes much less of a problem if $GdPO_4$ is used as the neutron absorber instead of Gd_2O_3.

3.2 PYROPHORICITY AND EXPLOSION RESISTANCE

3.2.1 Statement of Issue

As required by 10 CFR 60.135(b), the waste forms and WPs shall not contain explosive or pyrophonic materials or chemically reactive materials in an amount that could compromise the ability of the underground facility to contribute to waste isolation or the ability of the repository to satisfy performance objectives.

3.2.2 DOE Technical Approach and Results

The DOE has concluded that the potential for pyrophoricity of the Al-based SNF is much less than that for metallic uranium fuel (WSRC, 1997b). This is because of the lesser tendency to form uranium hydride and the possible dilution of the hydride by aluminum corrosion products. Further analyses are being performed. A recommended guide for pyrophoricity testing is being prepared under American Society for Testing and Materials (ASTM). The generation of explosive hydrogen as through the corrosion of aluminum or the fuel is minimized through reduction in free water content in the canister. The flammability limit for hydrogen in helium is assumed to be 4 volume percent. The limit on free water is calculated by assuming that for every mole of hydrogen produced, 1.33 moles of water is required. The DOE has proposed to dry the canister through the application of heat and vacuum. A test canister has been designed to study the drying behavior.

3.2.3 CNWRA Evaluation

The conclusion that Al-based SNF is not as pyrophoric as metallic U is reasonable. Further data using ASTM developed guidance documents may confirm this assumption. The calculation of the limit on water content in the canister based on hydrogen generation is reasonably conservative. Additional hydrogen will be generated as a result of the radiolysis of water. However, this is not expected to change the ratio of

water to hydrogen substantially. The vacuum drying and helium pressurization are expected to provide added safety factors.

3.3 POSTCLOSURE THERMAL EFFECTS

3.3.1 Statement of Issue

Postclosure thermal analyses must be conducted on the co-disposal WP with different fuel assembly configurations to demonstrate that the postclosure temperature of Al-based SNF will not exceed the critical temperature and that the preclosure thermal exposure during interim storage will not create a situation that will adversely affect the behavior of Al-based SNF.

3.3.2 DOE Technical Approach and Results

The DOE conducted 2-D steady state thermal analyses of the co-disposal WP using the finite element code ANSYS 5.1 for a heat load of 83 MTU/acre (CRWMS M&O, 1997d). These analyses used the previously determined surface temperatures of DHLW WP with four HLW canisters (Bahney, 1996) as the boundary conditions for the co-disposal WP, and applied the heat loads of DHLW and MIT SNF co-disposal canisters. This means the co-disposal WP thermal analyses involve a two-model approach. A 3-D transient finite element model of the DHLW WP with four HLW canisters provided the DHLW WP surface temperature history for use as boundary conditions for the 2-D co-disposal WP model. Since the repository rock temperatures change slowly with time and the HLW glass and MIT fuel heat generation rates decrease with time, the co-disposal WP was analyzed at several different times from emplacement out to 100 years as a steady state problem.

The results of preliminary analyses show that the temperature of Al-based SNF peaks 20 years after the time of emplacement of co-disposal WP, then subsequently decreases with time. Since the peak temperatures of the HLW glass and its canister, MIT SNF and its canister, and co-disposal WP shell are below the DOE temperature limit goals, the DOE concluded that the proposed co-disposal canister can be loaded in the co-disposal WP.

3.3.3 CNWRA Evaluation

The DOE has conducted thermal analyses for only 100 years after the emplacement of the co-disposal WP. The DOE should conduct thermal analyses with consistent thermal input data and appropriate boundary conditions for the postclosure period that is beyond 100 years after the emplacement of co-disposal WP to provide a thermal history for the HLW glass and its canister, MIT SNF and its canister, and co-disposal WP shell. This postclosure thermal history may be necessary to evaluate that a long duration thermal exposure even at less than peak temperature will not adversely affect the postclosure behavior of Al-based SNF. For example, the dissolution rate of the fuel is dependent on the temperature and hence, time variation of the temperature will be needed to estimate the dissolution rate and flux of radionuclide released.

3.4 WASTE FORM DISSOLUTION AND RADIONUCLIDE RELEASE

3.4.1 Waste Form Dissolution

3.4.1.1 Statement of Issue

The dissolution rate of the waste form determines, in part, the rate of release of radionuclides, provided solubility limits and surface segregation do not determine the release rate. The relationship between dissolution rate and radionuclide release is also determined by the mode of dissolution. Since the Al-clad fuels, both for the co-disposal and melt-dilute options, have uranium rich second phase particles dispersed in an Al solid-solution matrix, dissolution is selective and radionuclide release is not by congruent dissolution.

3.4.1.2 DOE Technical Approach and Results

A test protocol has been proposed by the DOE (WSRC, 1997b) and is being balloted through the ASTM. The proposed test methods include static immersion tests, flowthrough tests, and electrochemical tests. The depleted U-Al alloys containing U ranging from 10 to 67 wt% will be tested. Tests on as-cast alloys simulating various melt-dilute compositions are in progress. However, tests on co-disposal fuel materials are yet to be completed. Although several environmental compositions will be tested, it appears that the focus of the test program is on simulated J-13 well water.

In the static tests on an Al-25 percent U alloy exposed to J-13 water, the Al matrix around the UAl_4 particles and in the eutectic corroded preferentially, indicating that it was anodic. The filtrate from the solution contained particles of UAl_3 and indicated that these have particles spalled off after corrosion of the matrix. Interestingly, the electrochemical measurements of a series of U-Al alloys indicated that the corrosion potential decreased with an increase in U content. Additionally, the corrosion potential of U was significantly more negative than the corrosion potential of pure Al (WSRC, 1997b). These electrochemical measurements are consistent with those reported by Chandler et al.(1997). Despite the more negative corrosion potential of higher U-containing alloys, in the electrochemical tests preferential corrosion of Al matrix was observed.

The flowthrough tests have not progressed sufficiently to enable a comparison of dissolution rate and radionuclide release rate.

3.4.1.3 CNWRA Evaluation

The corrosion tests of the co-disposal and the melt-dilute fuels have not progressed sufficiently to determine the relationship between dissolution rate of the fuel and the radionuclide release rate. This relationship is especially important since the fuel material in both cases is highly heterogeneous and the dissolution is selective. It must be noted that the behavior of the Al matrix and UAl_x ($x \approx 4$) phases may be highly dependent on the prior processing history, since segregation of various radionuclides to one or the other phase may be dependent on the processing history. Testing of as-cast U-Al simulants may not be reflective of the behavior of co-disposal or melt and dilute fuels. At present, there are few measurements of the radionuclide release from actual fuels. The corrosion measurements performed on fuels in the pools have focused on corrosion rate and morphology rather than radionuclide release rate.

The galvanic effect between the UAl_x particles and the Al matrix and cladding is not clear. The corrosion tests have indicated preferential dissolution of the Al matrix whereas corrosion potential measurements have indicated that U and alloys containing a higher proportion of U have more negative potentials. The corrosion rates also seem to increase with U content. These apparent contradictions suggest that (i) the U is more anodic than Al, but the UAl_x is more cathodic than either the Al or U, and (ii) the dissolution behavior is dependent on the volume fraction and distribution of the UAl_x. It must be noted that the potential measurements alone are insufficient to deduce the galvanic effects between disparate metals. The polarization behavior of the metals in contact is also important. Thus, two metals whose corrosion potentials are within 10s of millivolts apart may show significant galvanic effects if they do not polarize. Conversely, even if two metals show significant corrosion potential differences, the galvanic effect may not be significant if both metals passivate rapidly.

The flowthrough tests provide the advantages that radionuclide release rates can be measured easily and the "intrinsic" reaction rates can be measured without the limitation of diffusion controlled processes. Such a test has been used successfully in Light Water Reactor (LWR) fuel to estimate the forward reaction rates. However, for a heterogeneous material such as the Al-clad fuels, measurement of radionuclide release and dissolution rate using flowthrough tests may not be conservative, because in stagnant solution, corrosion of the interface between the Al matrix and uranium enriched particles may be accelerated by the accumulation of aggressive ionic species that will then result in the spalling of these particles, contributing to high release rates. In a flowing solution, the accumulation of deleterious ionic species is minimized due to the mixing with fresh solution. Hence it is important to compare the dissolution rates and the radionuclide release in stagnant and flowthrough tests.

The presence of microbiological colonies on the fuels has been reported (Guenther, 1995; Wolfram et al., 1998). The microbial colonies may survive the dry period (in an inactive state) and cause increased corrosion during the wet period. While it may not be necessary to conduct a detailed study of the effect of microbiological organisms on dissolution, the effect of these organisms in altering the near-field chemistry can be simulated by appropriate abiotic environments.

The test protocol in WSRC (1997b) and currently in ballot in the ASTM appears to be deficient in that an insufficient range of environmental conditions is specified. For example the effect of steel and basket dissolution products and radiolysis is not considered in the test protocols. In tests of Al cladding materials in hot brine (Brodda and Fachinger, 1995), it has been shown that the presence of dissolved iron corrosion products caused very high corrosion of Al, whereas galvanic coupling with stainless steel or alloy C-4 did not cause a significant increase in corrosion rate. Presence of iron also increased the radionuclide release of both α and γ emitters. Additionally, the near-field chemistry will be significantly different from the saturated zone well water (e.g., J-13 water) due to evaporation, radiolysis, and interactions with WP components. Hence, a predictive model for dissolution should consider the effect of a range of water chemistry.

3.4.2 Radionuclide Characterization and Inventory

3.4.2.1 Statement of Issue

The radionuclide inventory of an Al-based SNF form will significantly affect many factors important to selection of treatment option, such as radiogenic heat production, criticality potential, radiological safety during treatment, and performance in a geologic repository. Because of the heterogeneous microstructure, there may be a segregation of various actinides and fission products in the different phases being determined

by solid-state reactions and solubility in one or the other phases. Since preferential dissolution of the fuel occurs as discussed in section 3.4.1, the distribution of radionuclides is an important factor in evaluating radionuclide release.

3.4.2.2 DOE Technical Approach and Results

There is great variety among the Al-based SNFs with regard to important characteristics such as U^{235} content and burnup, and uncertainty in the validity of the available data on such characteristics. From these data, the computer program ORIGEN has been used to calculate inventories of activation and fission products (and their daughters) produced during fuel burnup (e.g., WSRC, 1997b, section 5.3.2.1). The reports acknowledge the uncertainties in ORIGEN calculations owing to incomplete knowledge of Al-SNF histories. More generally, some of the uncertainties of applying ORIGEN to the wide variety of DOE SNF forms were discussed by Duguid et al. (1997; section 3.2) in their PA of such wastes.

3.4.2.3 CNWRA Evaluation.

The distribution of radionuclides in different metallurgical phases has not been reported. This distribution may be important in calculating the release rate of radionuclides from the dissolution rate of fuel.

As mentioned in the reports, the accuracy of the Appendix A-type data packages is an open question which has not been resolved (e.g., Skidmore, 1997, p. 6). Skidmore (1997, figure 4.4) presents a table comparing pretreatment characterization needs for the two options that appears to favor the melt-dilute option. However, this comparison assumes that melt-dilute will not require extensive pretreatment characterization because the final waste form will differ significantly from the initial form. This assumption may be deficient, because lack of complete knowledge of pretreatment chemistry and/or radionuclide inventory could hinder the ability to assess (i) radiological safety and criticality potential during the treatment process and (ii) performance in a geologic repository of the final product of treatment.

3.5 MELT-DILUTE TECHNOLOGY

3.5.1 Statement of Issue

Waste form stability is one of the key criteria for the disposal of the WP and its components. As required by 10 CFR 60.135c, the waste form should be in a solid form and placed in a sealed container. While the Part 60 does not preclude the presence of several phases, performance of immiscible phases (slag/alloy) in the repository environment is a significant concern. The waste form should be designed such that the chemical composition and microstructure of the waste form are within the proposed target range and components are homogeneously distributed. Assessment of long term radionuclide leaching rates and transport characteristics for the melt-dilute waste form is important for PA.

3.5.2 DOE Technical Approach and Results

The HEU Al-SNF will be melted in a crucible and mixed with depleted uranium (and Al if needed) to dilute the HEU to low enriched uranium levels (<20%). The DOE will use induction melting to ensure rapid and complete mixing and melting of the uranium metal with aluminum. After mixing, a sample will be obtained to determine chemical composition of the melt. The melt will be cast either by pouring in a mold,

or solidified in the crucible. In addition, the DOE is investigating the range of microstructures resulting from various dilution and target comparisons (WSRC, 1997b).

3.5.3 CNWRA Evaluation

The following issues related to disposition of Al-SNF melt-dilute packages in the repository are not addressed in the report:

- Partitioning of Radionuclides—During melt-dilution, radionuclides present in the Al-SNF will be partitioned into three major macroscopic phases, U-Al intermetallic alloy phase, slag consisting of Mg, Si, Fe, and Ni impurities, and offgas consisting of water vapor and volatile gases such as Cs_2O. While U is anticipated to be contained as U-Al intermetallic phase, the actinides and fission products could be partitioned between offgas, slag, or U-Al phase. In addition, these radionuclides will be partitioned in two different host matrices (slag and alloy) which may have different waste form dissolution behavior. The DOE does not address the issues relating to radionuclide dissolution from slag and alloy.

- Formation and Settling of Crystals—As the melt-diluted waste form is cooled, crystals will form and may settle at the bottom of the crucible. Depending upon the host crystalline phase for radionuclides, the melt-dilute waste form may have inhomogeneous distribution of radionuclides that may impact waste form performance.

- Cracking of Waste Form—Depending upon the cooling rates, thermal stresses may be generated in the solidified waste form due to differential contraction of the slag and alloy. These stresses may be enough to fracture the alloy-slag interface and generate fines. The formation of fines may impact the radionuclide release rates.

- Chemical Durability—The product consistency test-A (PCT-A) for glass waste form is not a PA test. It is typically used to assess the quality of HLW glass to ensure the waste form has the characteristics within the prescribed range for release rate. The DOE has tested the release rate data for melt-dilute samples and compared them to the results of the glass leach tests. This kind of comparison is not valid because the PCT test for glass measures release rate of Na, Li, and B, which are absent in melt-dilute waste forms. The report is lacking specificity in regard to the methods that will be used to determine long-term performance of the melt-dilute waste form. In addition, test plan for determining the influence of dissolution of melt-dilute waste form on the glass waste form and vice-versa should be developed. Specifically the effects of the colloids, formed during waste form dissolution process, on radionuclide release and transport of melt-dilute waste form should be examined.

- Waste form Sampling—The report is not specific regarding how samples will be obtained for characterization. Since there are at least two macroscopic immiscible phases, slag and alloy, during stirring, these two phases will be intermixed. Any sample taken during mixing will provide an average composition of the entire crucible contents. Such an approach is acceptable for determining average melt composition provided the slag and alloy phases on cooling remains homogeneously distributed. If on cooling, slag separates from the alloy and floats on the surface due to the lack of stirring, the waste form cannot be considered as

homogeneous because the two phases not only will have different composition but will have different radionuclide partitions and radionuclide release behaviors The DOE should provide the methodology for sampling that will ensure that the sampled melt-dilute waste form is similar to the final waste form.

- Role of Fission and Activation Products—The report does not discuss the influence of activation and fission products on the chemical stability of the waste form. Effect of activation and fission products on the chemical stability should be determined and evaluated. Similarly, the effect of impurities such as Mg, Si, Fe, and Ni; addition of poisons such as gadolinium; and incorporation of the dissolution products of the crucible material on the chemical stability of the waste form should be determined and evaluated.

- Density Measurements for Measuring Composition—The DOE plans to use density measurements to confirm U enrichment. The test data provided in the report focuses only on the pure sample mixtures. The influence of ternary components such as fission products, activation products, impurities, poisons, porosity, and intermixing of slags could significantly bias density measurement.

3.6 SYSTEM LEVEL PERFORMANCE ASSESSMENT

3.6.1 Statement of Issue

In addition to the overall system performance requirement in 10 CFR Part 60.112, there are subsystem performance requirements in terms of containment and controlled release of radionuclides in 10 CFR 60.113. However, following NAS recommendations, the subsystem requirements may be removed and, in their place, a total system performance requirement in terms of dose may be substituted. The effects of disposing of Al-based fuels in the repository should be evaluated by total system PA in terms of the dose to an individual at a defined location.

3.6.2 DOE Technical Approach and Results

A preliminary total system PA was conducted by the DOE (Duguid et al., 1997) using an approach similar to that used for commercial fuels. The dose analysis was performed in terms of dose to an individual drinking 2L of water per day from a well 5 km downgradient from the repository. The conceptual models for the container performance and radionuclide transport were essentially the same as for TSPA-95 conducted for commercial fuels (Andrews et al., 1995). For the metallic spent fuel, the dissolution rate used was derived from an earlier PA of the DOE fuels

$$M = A e^{-\frac{B}{T}} \left(t_2^{\,C} - t_1^{\,C} \right) D \cdot E \cdot S \tag{3-1}$$

where M is the mass of the layer corroded in a given time step, A is a pre-exponential term, B is the activation energy, T is the absolute temperature, t_2 and t_1 are the time at the end and beginning of the time step, C is an exponent for the time dependence, D is a saturation dependent term, E is an oxygen

concentration dependent term, and S is the surface area of the fuel. The radionuclide release (prior to exceeding the solubility limits) is then estimated assuming congruent release.

The details of the modeling are described in Duguid et al. (1997). The dose associated with each type of fuel was calculated by considering a canister filled with HLW and Al-clad fuel, only by glass HLW, and only by commercial spent fuel (of a quantity equal to the Al-based fuels). The resulting dose is governed by I^{129} and Tc^{99} at time period less than 20,000 years and by ^{237}Np at longer time periods. The peak dose occurs at 10,000 years and, for the Al-based fuels, is roughly 2.5 times that of the commercial fuels if compared on the basis of equivalent MTHM. At longer time periods, the dose from both fuels are equal due to solubility limitations of ^{237}Np. It must be noted that the solubility of ^{237}Np was lowered by two orders of magnitude based on a recent survey of available data (Duguid et al., 1997).

3.6.3 CNWRA Evaluation

While the PA calculations have many conservative assumptions regarding the distribution of the WPs and the container failure processes, there may be some nonconservatisms in the models. These include

- The dissolution rate equation is an empirical relationship without a sound mechanistic basis. For example, Eq. 3-1 assumes that the value of E is 0.2 based on atmospheric oxygen concentration and, hence, implies a linear dependence on the oxygen fraction in air. In reality, dissolved oxygen controls the corrosion potential and the dissolution current has an exponential dependence on the corrosion potential.

- Use of Eq. 3-1 for calculating release rate of soluble radionuclides (such as I^{129}) implies that the dissolution and radionuclide distribution in the fuel is uniform. In contrast, all the experimental work reported to date indicates that highly selective dissolution followed by spalling of UAl_x particles may result in pulsed release. As mentioned previously, the distribution of radionuclides in various microstructural features in unknown at present.

- The surface area of the fuel is an extremely important parameter and a technical basis for the value assumed for the calculation was not provided by Duguid et al. (1997).

- The use of two orders of magnitude lower Np solubility was based on unsaturated test results conducted Argon National Laboratory on commercial spent fuel where significant retention of Np was seen in secondary minerals. The use of the same solubility values for Al-based fuels where corrosion behavior is different from commercial SNF needs to be better justified.

4 SUMMARY

DOE reports pertaining to the two treatment technologies for the disposition of Al-clad fuels were reviewed in terms of the technical issues involved in permanent disposal. Independent analyses of the effect of these fuels in the repository on system performance was not performed and is recommended for the future. The DOE total system PA indicates that the disposal of the Al-clad fuels in the repository would not cause a disproportionate dose at the 5-km boundary. However, the uncertainties in this calculation include the validity of the model used to calculate the dissolution rate of the fuel, which is then used to calculate the flux of highly soluble radionuclides such as I^{129} and Tc^{99}, the use of appropriate surface area for the fuel, and the solubility of ^{237}Np. Further examination of these uncertainties and their effect on system level performance should be pursued.

Preclosure Performance

In terms of preclosure operations, the effect of interim storage on the integrity of road ready canister needs to be addressed by the DOE. The canister integrity may be adversely affected by internal corrosion from residual moisture containing fission products such as I^{129}.

The main concern in terms of the preclosure handling of the co-disposal fuel assembly is the effect of the DOE storage temperature maximum of 200 °C on the humid air oxidation, creep rupture, and embrittlement of the cladding. The vapor phase corrosion tests done to date indicate that the critical relative humidity for the onset of humid-air corrosion of Al cladding materials is quite low (less than 20 percent) and that breakaway corrosion leading to internal oxidation occurs at temperatures as low as 150 °C, especially in the presence of radiation. Because internal corrosion can occur at a rapid rate and a fresh supply of external water may not be needed for continued penetration of internal oxidation (water released from hydrated aluminum oxide can contribute to continued oxidation), the criterion that penetration should be less than 3 mils may not be met after prolonged exposure at 200 °C. The data generated thus far do not lead to a bounding determination of cladding and fuel penetration. Similarly, while the calculations of creep of fuel and cladding have some measure of conservatism, the assumption that the stress arises only from self-weight of the fuel is nonconservative. The residual stresses may be many orders of magnitude larger than the gravitational stresses and can cause intergranular failure within months. The effect of 200 °C exposure on thermal embrittlement of the cladding may also need to be considered in evaluating the behavior of the fuel assembly under DBEs. The thermal analyses used for determining whether the fuel temperature would meet the requirements must be performed using consistent input data and boundary conditions.

Postclosure Performance

The criticality analyses for the direct/co-disposal option have many conservative assumptions. However, some aspects of the calculations need to be addressed to eliminate potential nonconservatisms:

- Justification is needed for the assumption that 1 percent of the uranium in the fuel is U-234. Since this isotope can act as an efficient neutron absorber and is not typically present in these quantities in fresh fuel, its inclusion may represent implicit burnup/decay credit.

- More compact neutronic systems may need to be considered where there is a segregation of the fuel plates due to gravity driven compaction and the neutron absorber plates that are left intact.

- In the case of degraded fuel, the segregation of the neutron absorber as a result of migration should be addressed.

- The effect of reconfiguration of the fissile material outside the WP into a critical configuration needs to be addressed. It is indicated that these calculations will be made in the future.

- While gadolinium has several advantages over boron as a neutron absorber, the higher burnup of the gadolinium needs to be considered in determining the initial concentration of gadolinium.

The criticality of the melt-dilute waste, although less of a concern than the direct/co-disposal fuel, needs to be addressed in order to determine if neutron absorber materials need to be incorporated in the melt.

The DOE has addressed the issues of pyrophoricity and reactivity in a reasonably conservative fashion. The Al-based fuels are not as pyrophoric as metallic uranium. The limitation placed on the volume of water as part of the dry storage acceptance criteria and the pressurization of the canister by helium ensure that the concentration of hydrogen generated through corrosion and radiolysis will not reach flammability limits.

The Al-based fuels, due to their small volume compared commercial and other DOE fuels, are not expected to contribute significantly to the overall dose from the repository. The DOE preliminary total system PA of the DOE fuels has indicated that the contribution to dose from a representative Al-based fuel is equivalent to that from an equivalent MTHM commercial spent fuel (Duguid et al., 1997). The dose at a time period less than 20,000 years is dictated by I^{129} and Tc^{99}, whereas the dose at longer time periods is dictated by ^{237}Np. The peak dose for the Al-based SNF occurs at about 10,000 years and is predicted to be about 2.5 times that for equivalent MTHM of commercial spent fuel (Duguid et al., 1997). At longer times, the total dose is essentially equal for the two types of spent fuel. Independent calculation of the dose from Al-based fuels have not been made by the NRC or the CNWRA, and this is recommended for the future.

The corrosion tests of the co-disposal and the melt-dilute fuels have not progressed sufficiently to determine the relationship between dissolution rate of the fuel and the radionuclide release rate. This relationship is especially important since the fuel material in both cases is highly heterogeneous and the dissolution is selective. The test protocol in WSRC (1997b) and currently in ballot in the ASTM is too general for evaluation of its adequacy. An insufficient range of environmental conditions is specified. For example, the effect of steel and basket dissolution products and radiolysis is not considered in the test protocols. In tests of Al cladding materials in hot brine (Brodda and Fachinger, 1995), it has been shown that the presence of dissolved iron corrosion products caused very high corrosion of Al, whereas galvanic coupling with stainless steel or alloy C-4 did not cause a significant increase in the corrosion rate. The presence of iron also increased the radionuclide release of both α and γ emitters by two orders of magnitude. Finally, the corrosion tests should include actual fuel materials in addition to depleted U-Al alloys as simulants because of the great dependence of the dissolution behavior on the microstructure and radiolysis.

In the case of the melt-dilute option, the partitioning of radionuclides occurs between alloy, slag, and the offgas. The DOE reports do not provide detailed information on the partitioning that may be important if the slag and alloy are cast in the same canister and if filters used to collect the radionuclides in the offgas are also disposed in a like manner. The partitioning of the radionuclides is still being investigated by SRS. The PCT is mentioned as a PA test. PCT, which was conceived as a test to determine the consistency of vitrified waste

form and, consequently, is not a PA test. Furthermore, the test measures the release of Na, Li, and B, which are absent in the melt-dilute waste forms. Since the melt-dilute waste form is a metallic, tests appropriate to direct/co-disposal waste fuels should be used. If slag and melt are going to be poured into the same canister, there may be a risk of pulsed release upon canister breach because of the layered structure of the solidified waste form.

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