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Revision 1

Parameter Selection for Department of Energy Spent Nuclear Fuel to be Used in the Yucca Mountain License Application

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ABSTRACT

This report contains the chemical, physical, and radiological parameters that were chosen to represent the U.S. Department of Energy spent nuclear fuel in the Yucca Mountain license application. It also contains the selected packaging requirements for the various fuel types and the criticality controls that were used. The data are reported for representative fuels and bounding fuels in groups of fuels that were selected for the analysis. The justification for the selection of each parameter is given. The data reported were not generated under any quality assurance program.

CONTENTS

ACRONYMS

ATR	Advanced Test Reactor
CSNF	commercial spent nuclear fuel
DOE	U.S. Department of Energy
EOL	end-of-life
FFTF	Fast Flux Test Facility
FSV	Fort Saint Vrain
HEU	highly enriched uranium
HLW	high-level waste
HTGR	High Temperature Gas Reactor
HWCTR	Heavy Water Components Test Reactor
INEEL	Idaho National Engineering and Environmental Laboratory
LEU	low-enriched uranium
MOX	mixed oxide
MTHM	metric tons of heavy metal
PWR	pressurized water reactor
QA	quality assurance
RW	Office of Civilian Radioactive Spent Nuclear Fuel
SNF	spent nuclear fuel
TMI-2	Three Mile Island Unit 2
TSPA	total system performance assessment

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1. INTRODUCTION

To perform a Total System Performance Assessment (TSPA) on the U.S. Department of Energy (DOE) spent nuclear fuel (SNF), it is necessary to obtain data on the characteristics and performance of the DOE fuel in a repository setting. The Office of Civilian Radioactive Waste Management (RW) has a good database of characteristics of commercial SNF due to many years of research and characterization studies. No such database exists for the DOE SNF. There has been some research in a few performance parameters that could be applied to the DOE SNF inventory. In addition, some of the commercial SNF parameters can be adapted or modified as deemed appropriate to obtain the parameters for analysis of the DOE SNF. In some cases, best engineering judgment has to be used to estimate the parameter, because there is no existing data on the SNF. In these cases, groups of knowledgeable individuals met to make the estimates.

The data requested for the DOE SNF include:

- Chemical and physical properties
  - Air oxidation rate
  - Wet dissolution rate
  - Surface area of the matrix
  - Clad failure fraction
  - Free radionuclide inventory fraction
  - gap radionuclide inventory fraction
  - fuel area per package
  - heat generation
- Radiological inventory
- Packaging
- Criticality controls.

2. APPROACH

A fuel type was selected to represent a group of fuels that had similar characteristics. This approach has been used in prior DOE fuel analysis and prior TSPA calculations. The representative fuel is selected based on (1) its quantity as compared to the total quantity of fuel in the group, and (2) the availability of characterization data. The representative fuel is not a "worst case" or "bounding" fuel in its characteristics, nor is it meant to be a composite fuel for the total inventory. A bounding fuel for each fuel characteristic was also selected for each group based on the fuel characteristics. The bounding fuel in many cases comprises only a small fraction of the total inventory of the group. The bounding fuel characteristics bound all the other fuels' performance characteristics or physical parameters in the group. In some cases the representative fuel is also the bounding fuel for a group.

2.1 Chemical and Physical Properties

2.1.1 Air Oxidation Rate

The air oxidation rates for most materials were unknown. DOE-RW uses the value of 0 for commercial spent nuclear fuel (CSNF) in its TSPA calculations. There were a few cases where information about the DOE SNF could be extrapolated from existing data. However, the effect of the air oxidation rate on the DOE fuels is insignificant compared to the wet dissolution rate.

2.1.2 Wet Dissolution Rate

Wet dissolution is the primary method for making the radionuclides available for transport in the repository. There exists in the chemical literature a large compilation of dissolution data for unirradiated materials. There are very little data on the wet dissolution of irradiated materials. Where there were good models for wet dissolution, they were used. Where no models existed, the model recommended was based on an extrapolation from an existing model using expert judgment.

2.1.3 Surface Area of the Matrix

The surface area was based on the known geometry of the fuel. Where the existing geometry was unknown, estimates were made from similar known fuel types. The National SNF database contains the dimensions of many of the fuel assemblies but does not always contain the dimensions of the individual fuel rods or plates in the dimension section. In some cases, the dimensions of the rods or plates were contained in the comment section. If it was not in the National SNF database, then other references were obtained with the needed dimensions. The geometric dimensions were used to calculate the surface area. Because the thickness of the clad is thin, its surface area was neglected. The worst-case or bounding fuel surface area was determined by calculation for all the fuels in the group. Because metal and oxide surfaces have some roughness, a surface roughness factor was used to compensate for the roughness. The RW Analysis/Model report, *CSNF Waste Form Degradation: Summary Abstraction*, ANL-EBS-MD-000015, Revision 0 uses a surface roughness factor of 3 for CSNF, which has pellets of sintered uranium oxide.

The surface area of a plate assembly is given by the formulas:

$$SA_{plate} = 2lw + 2t(l + w) \quad . \tag{1}$$

For thin plates the thickness can be ignored.

$$SA_{assembly} = SA_{plate} \times \text{number of plates} \tag{2}$$

The surface area of a solid rod is given by the formula:

$$SA_{rod} = \pi \, l \, OD + 1/2 \, \pi \, OD^2 \quad . \tag{3}$$

For small diameter rods, the surface area on the top and bottom of the rod can be ignored.

$$SA_{assembly} = SA_{rod} \times \text{number of rods in an assembly} \quad . \tag{4}$$

The surface area of an annular fuel rod is given by the formula:

$$SA_{rod} = \pi \, l \, [OD + ID] + 1/2 \, \pi \, (OD^2 - ID^2) \tag{5}$$

The surface area of a double annulus fuel assembly is given by the formula:

$$SA_{assembly} = \pi \, l \, [OD_{out} + ID_{out} + OD_{in} + ID_{in}] + 1/2 \, \pi \, (OD_{out}^2 - ID_{out}^2) + \tfrac{1}{2} \, \pi \, (OD_{in}^2 - ID_{in}^2) \tag{6}$$

where

Width = w, Length = l, Thickness = t, Surface area = SA, Outside Diameter = OD, Inside diameter = ID

The surface area for the fuels was calculated using the fuel geometry and dimensions and appropriate equations in an Excel spread sheet. In all cases, the specific surface area of the fuel, cm^2 per gram of matrix, was also obtained by dividing the calculated surface area by the mass of the matrix material in grams. For the DOE SNF the geometric surface area was multiplied by a roughness factor of 5 to 10 depending on the fuel materials and condition for conservatism.

2.1.4 Volume of the Fuel Matrix

The volume of water contained in a waste package is important in the calculation of the release of radionuclides. The volume of water is best estimated by knowing the volume of the SNF in the waste package. The volume of the fuel can be calculated from the dimensions of the fuel rods, plates, and assemblies, or it can be calculated from the mass of the matrix divided by the density of the matrix material. By using the volume of the fuel derived from the density of the fuel matrix, the results are very conservative because they neglect the volume of the cladding and structural materials.

The volume of a plate assembly is given by the formulas:

$$V_{plate} = lwt \tag{7}$$

$$V_{assembly} = V_{plate} \times \text{number of plates} \quad . \tag{8}$$

The volume of a solid rod is given by the formulas:

$$V_{rod} = l\pi(OD/2)^2 \tag{9}$$

$$V_{assembly} = V_{rod} \times \text{number of rods in an assembly} \quad . \tag{10}$$

The volume of an annular fuel rod is given by the formula:

$$V_{rod} = l\pi \ [(OD/2)^2 - (ID/2)^2] \quad . \tag{11}$$

The volume of a double annulus fuel assembly is given by the formula:

$$V_{assembly} = l\,\pi\,[(OD_{out}/2)^2 - (ID_{out}/2)^2 + (OD_{in}/2)^2 - (ID_{in}/2)^2] \quad . \qquad (12)$$

The volume for the fuels was calculated using the fuel geometry and dimensions and appropriate equations in an Excel spread sheet and indepently using the mass and the density of the fuel matrix. The number of fuel assemblies in a standard canister was obtained from the National SNF database. The volume of SNF per standard canister was obtained from the product of the volume per assemblies and the number of assemblies per standard canister. The bounding case was determined by the smallest volume of SNF in the canister from the two methods.

2.1.5 Clad Failure Fraction

The cladding failure fraction was based on known characterization data or extrapolated from similar types of fuel.

2.1.6 Free Radionuclide Inventory Fraction

The free radionuclide inventory is the quantity of radionuclides that are available for immediate release when the waste package or the SNF container is breached without having to dissolve the fuel cladding or matrix. For a radionuclide to become part of the free inventory, it needs to be released out of the fuel matrix and the clad. If the fuel clad is intact, the radionuclides enter into a gap between the fuel matrix and the clad where appropriate. If the clad is not intact, the radionuclides are released through the clad breaches into the space outside the fuel or into the free space. Most of the driving force to move the radionuclides from the matrix is the temperature of the fuel, the radionuclide concentration gradient, the internal partial pressure, and the degree of grains in the fuel matrix. Gaseous radionuclides migrate best, and semivolatile radionuclides can migrate if the temperatures are high enough. The Idaho National Engineering and Environmental Laboratory (INEEL) experience with the monitoring of disrupted fuel in dry storage has shown that only a very small quantity, just above the detection limits, of gaseous fission products are released from the fuel. Only gaseous fission products have been detected.

The RW Analysis/ Model report, *CSNF Waste Form Degradation: Summary Abstraction*, ANL-EBS-MD-000015, Revision 0 gives the main isotopes in the gap and grain boundaries to be cesium, technetium, strontium, and iodine. It states that the gap inventory for CSNF is composed of two main isotopes, I-129 and Cs-137. The measured values are reported as a function of the fission gas release. For I-129 it is 1, and for Cs-137 it is 1/3. The report states that the quantity is driven primarily by the degree of friability of the fuel matrix and the temperature of the fuel. The previously used value of 2% was reported to be nonconservative for CSNF.

The free radionuclide inventory fraction was also based on the fuel construction methods, the characteristics of the fuel matrix, the as-stored condition of the fuel, and the treatment of the fuel that was projected prior to being transported to the repository. If the fuel is dried and placed in an inert gas atmosphere, the existing free radionuclide inventory will be removed by the process.

2.1.7 Gap Radionuclide Inventory Fraction

The gap radionuclide inventory is the quantity of radionuclides that are available for immediate release when the cladding is breached without having to dissolve the fuel matrix. For a radionuclide to become part of the gap inventory it needs to be released out of the fuel matrix and into the gap between the matrix and the clad where appropriate. Some SNF types are constructed without a gap between the fuel matrix and the cladding. Most of the driving force to move the radionuclides from the matrix is the

temperature of the fuel, the internal partial pressure of the radionuclide, the concentration gradient of the radionuclide, and the degree of grains in the fuel matrix. Gaseous radionuclides migrate best, and semivolatile radionuclides can migrate if the temperatures are high enough.

The RW Analysis/ Model report, *CSNF Waste Form Degradation: Summary Abstraction*, ANL-EBS-MD-000015, Revision 0 gives the main isotopes in the gap and grain boundaries to be cesium, technetium, strontium, and iodine. It states that the gap inventory for CSNF is composed of two main isotopes, I-129 and Cs-137. The measured values are reported as a function of the fission gas release. For I-129 it is 1, and for Cs-137 it is 1/3. The report states that the quantity is driven primarily by the degree of friability of the fuel matrix and the temperature of the fuel. The previously used value of 2% was reported to be nonconservative for CSNF.

The gap radionuclide inventory fraction was also based on the condition of the fuel clad, fuel construction methods, the characteristics of the fuel matrix, the as-stored condition of the fuel, and the treatment of the fuel that was projected prior to being transported to the repository.

2.1.8 Fuel Area Per Package

The fuel area per package was obtained from the surface area of fuel and the projected packaging of the fuel into the waste package. The projected number of packages was obtained from the National SNF Program database version 4.4.0. The number of packages may change slightly as the packaging in refined and new versions are released.

2.1.9 Heat Generation

The heat generation was obtained from ORIGEN-2 generated data. The worst-case fuel and package were used to give a worst-case heat generation number.

2.2 Radiological Inventory

Radionuclide calculations have been performed for a variety of DOE Environmental Management fuels. The methodologies established for conducting these calculations have been reviewed by the National Spent Nuclear Fuel Program Quality Assurance (QA) organization. This evaluation showed that the methodology essentially meets the QA requirements for determining radionuclide inventories of the fuels.

ORIGEN-2 was used to perform the calculations. Where deemed adequate and applicable, the established cross-section libraries packaged with the program were used for the fuels. However, most of the DOE fuels are not enveloped by the established ORIGEN-2 cross-section libraries and had to be generated from basic reactor physics parameters of the fuel, reactor, and operating environment.

For each cross-section library, the methodology, calculations, assumptions, and outputs were validated by a qualified individual familiar with ORIGEN-2 input parameter requirements. The QA review of the methods for calculating both the new libraries and the validation check was used to generate each cross-section library indicated so that they essentially met the QA requirements.

If ORIGEN-2 values or similar calculations from other programs existed for a particular fuel type, the input parameters for the ORIGEN-2 code were evaluated. If the input parameters, assumptions, and calculations were satisfactory, compared to the existing QA reviewed methodology, the cross-section library was used for that particular fuel. When the input parameters were not satisfactory to the QA reviewed methodology, then they were recalculated.

The National Spent Nuclear Fuel Program established a process to estimate the radionuclide source term for all the DOE SNF. The process used the existing ORIGEN-2 data and known fuel parameters to identify a template to be used in the calculations. The result of the analysis was published as DOE/SNF/REP-078, *Source Term Estimates for DOE Spent Nuclear Fuels*.

2.3 Packaging

Packaging data were based on an analysis that included individually each fuel type in the group, rather than based on just the representative fuel.

2.4 Criticality Controls

A representative fuel type was chosen for each group, based on the quantity of that fuel and the total quantity of fuel in the group. In some cases, the representative fuel was not the largest contributor to the group, but a fuel on which there was existing data. The parameters for the group are those of the representative fuels. If there is variation in any parameter within the representative fuel, an average value was used. No attempt was made to use the worst-case values for the parameters in each group. Some fuels in a group could potentially have worse characteristics in a particular parameter than the representative fuel. But, because of the small quantity of the other fuel, it was decided that the parameter should not drive the projected performance of the whole group. As additional data become available, these parameters may be modified.

3. DATA SOURCES

National SNF database version 4.4.0.

Waste Form Degradation: Summary Abstraction, ANL-EBS-MD-000015 REV 00
Handbook of Chemistry and Physics
Sandia PA on DOE SNF, SAND-93-2330

Listed references

4. DOE SNF DATA PRESENTED BY TOTAL SYSTEM PERFORMANCE ASSESSMENT GROUPS

4.1 Uranium Metal

4.1.1 Group Description

There are 13 separate entries in the National SNF database for this group, containing 2,109 MTHM. The group is dominated by N-reactor fuel containing 2,100 MTHM. N-reactor fuel was chosen to represent the group because its mass was so large that the performance of the rest of the group's fuels, even if they might be worse than N-reactor fuel, would not change the overall group performance. The bounding fuel was chosen for each property based on the individual fuel characteristic.

4.1.2 Chemical and Physical Properties

4.1.2.1 Air Oxidation Rate. Uranium oxidation process has been studied for many years, but a good understanding of the mechanism has only been obtained fairly recently. Reference 1 reports the data are for unirradiated uranium metal. The irradiated uranium metal may oxidize at a faster rate than the unirradiated uranium metal. The linear reaction rate for long time periods for this reaction in the temperature range of 38 to 300°C is:

$$k = 1.09E+8 \exp[-71.3 \text{ kJ/mole/RT}] \text{ mg U/cm}^2 \text{ h} \quad . \tag{13}$$

The bounding rate for the reaction was determined in the temperature range of 40 to 300 C and is reported as:

$$k = 7.95E+8 \exp[-76.6 \text{ kJ/mole/RT}] \text{ mg U/cm}^2 \text{ h} \quad . \tag{14}$$

4.1.2.2 Wet Dissolution Rate. A review of the oxidation rates for metallic fuels was reported in Reference 1. Linear or paralinear kinetics was observed. Linear reaction rates were developed. In the case of paralinear kinetics, linear reaction rates were determined for times after initial parabolic oxide growth. Temperature dependent reaction rates were developed by standard regression methods. The recommended Arrhenius dependent reaction rates were reported. The reaction rate of the uranium metal-water system bounded the reaction rate of all other systems and was reported to be

$$K_l = 5.03E+9 \exp[-66.4 \text{ kJ/mole/RT}] \text{ mg U/cm}^2 \text{ h for temperatures from 20–300°C.} \tag{15}$$

A bounding or worst-case rate for the metallic uranium system is the instantaneous release model, where one assumes that all the radionuclides are ready for transport when the container is breached.

4.1.2.3 Surface Area of the Matrix. The surface area of the matrix is necessary because the release rate from corrosion of the matrix is dependent on the surface area. The uranium metal surface in an uncorroded condition would be relatively smooth, and the surface area could be calculated from the geometric dimensions of the fuel. Because the N-reactor fuel is irradiated and corroded, the surface area is significantly larger, but not as high as commercial fuel that has a relatively rough surface with a large number of cracks and porosity. By using Equation (6) and the mass of the element as 22.68 kg, the ideal surface area is 1.4E-5 m^2/g, assuming the largest size Mark IV element.

$$SA = 66.3\ \pi\ [6.15+4.32+3.25+1.22] + \pi/2(6.15^2 - 4.32^2) + \pi/2\ (3.25^2 - 1.22^2) = 3{,}156 \text{ cm}^2 \tag{16}$$

$SA/g = 0.3156\ m^2/22{,}680\ g = 1.39\ E\text{-}5\ m^2/g$ (17)

A roughness factor of 5 to 10 should be used to account for the actual condition. DOE has used a roughness factor of 3 for U.S. CSNF. A roughness factor of 5 was used making the nominal surface area 7.0E-5 m^2/g for the group.

The fuel with the largest surface area and the smallest thickness will have the largest surface area per gram of uranium. Several fuel types were evaluated and using the appropriate equation and the mass of 56.4 kg, the Experimental Breeder Water Reactor fuel has a calculated surface area of 2.78E-5 m^2/g.

$SA = 137 \times 9.53 \times 2 \times 6 = 1.57\ E4\ cm^2$ (18)

$SA/g = 1.57 m^2/56{,}400\ g = 2.78E\text{-}5\ m^2/g$ (19)

A roughness factor of 10 should be used to account for the worst-case condition, thereby making the bounding surface area equal to 2.78E-4 m^2/g.

4.1.2.4 Volume of the Fuel. The volume of the fuel can be calculated from the dimensions of the fuel rods, plates, and assemblies, or it can be calculated from the mass of the matrix divided by the density of the matrix material. By using the volume of the fuel derived from the density of the fuel matrix, the results are very conservative because they neglect the volume of the cladding and structural materials.

By using Equation (12) for N-reactor fuel, the volume is calculated to be 1,457 cm^3/assembly

$V = 66.3\ \pi\ [3.075^2 - 2.16^2 + 1.625^2 - 0.61^2] = 1{,}470\ cm^3$ (20)

By using the mass of an assembly and the density of uranium metal, the volume is calculated to be 1,243 cm^3/assembly.

$V = 22{,}680\ g/19.05\ g/cm^3 = 1{,}191\ cm^3$ (21)

For this use, the 1,191 cm^3/assembly is most conservative, because it allows the most water into the waste package. The remainder of the fuel types in this group had larger volumes per assembly. By using the smaller volume and 247 assemblies per MCO, the volume is 294,000 cm^3 per MCO.

The worst-case fuel in this group was determined to be the Experimental Breeder Water Reactor fuel. By using Equation (8), the volume per assembly was determined to be 4,018 cm^3/assembly. Based on the mass of an assembly and the density of uranium metal, the volume is calculated to be 2,951 cm^3. By using the smallest volume per canister and twelve assemblies per standard canister, the bounding volume of fuel in a canister is 35,410 cm^3/standardized canister.

4.1.2.5 Clad Failure Fraction. Much (50–80%) of the N-reactor fuel cladding has been reported to be disrupted. No additional documentable characterization data exist on any cladding conditions. A conservative approach is to assume that all the cladding is failed and not give any credit for the protection of the fuel meat by the cladding. The bounding clad failure fraction is 1 (100% failed).

4.1.2.6 Free Radionuclide Inventory Fraction. The free radionuclide inventory is the quantity of radionuclides that are available for immediate release when the waste package or the SNF container is breached without having to dissolve the fuel matrix or cladding

Typical burnup and hence heat generation rates for commercial fuel are 50,000 MWD/MTU. N-reactor fuel maximum burnup is reported to be 6,000 MWD/MTU, which is also the worst-case burnup for the group. Uranium metal fuel is significantly less friable than the uranium oxide fuel of CSNF. Therefore, because of the low burnup and fission product inventory, the low heat generation rate, and the physical structure of the fuel matrix, the quantity of fission products released from the fuel matrix will be significantly less than CSNF. In addition these fuels will be conditioned and repackaged, which will further reduce the free radionuclide fraction. N-reactor fuel is both the representative and bounding fuel in this group. The free fraction was arbitrarily set at 0.001 based on the discussion above. The bounding fraction is set at 10 times higher, or 0.01. There are no supporting analytical data for this value.

4.1.2.6 ***Gap Radionuclide Inventory Fraction***. Because of the fuel manufacturing methods, there is no gap between the fuel meat and the cladding. Because of the cladding condition, if there were a gap inventory it would have been previously released to the environment as the free inventory. For this reason, the gap inventory fraction is set to zero.

4.1.2.7 Fuel Area Per Package. A loading per package (247 assemblies) was determined from the National SNF database. The surface area for each type of fuel in the group and the loading was used to determine the surface area in the package. The value derived for this group is 78 m^2/package based on N-reactor fuel. The N-reactor fuel is also the bounding fuel for the group. Using a surface roughness factor of 5, the surface area is 400 m^2/package.

4.1.2.8 Heat Generation. The National SNF database reports the heat generation rate of an N-reactor assembly to be 3.5 W. Present packaging of the N-reactor fuel into MCOs places 247 assemblies into one MCO. This makes the heat generation rate of 1 MCO to be 864 W. The worst-case heat generation rate for the group is 270 N-reactor assemblies in a MCO, which has 945 W per container.

4.1.3 Radiological Inventory

The radiological inventory for this group is contained in DOE/SNF/REP-078, *Source Term Estimates for DOE Spent Nuclear Fuels*.

4.1.4 Packaging

The numbers of the various types of containers are:

- 11—18-in. × 10-ft standard canister
- 2—18-in x 15-ft standard canister
- 412—24-in. × 15-ft MCO.

4.1.5 Criticality Controls

Criticality analysis will be performed on each fuel type for transportation and storage of intact fuel assemblies and canisters. Criticality for the degraded modes will be performed on a worst-case fuel. All the fuels in this group are less than 2.5% enriched.

4.2 Uranium-Zirconium Alloy

4.2.1 Group Description

There are 6 separate entries in the National SNF database for this group, containing 0.6548 MTHM. The group is dominated by the Heavy Water Components Test Reactor (HWCTR) fuel containing 0.6518 MTHM. It was chosen as the representative fuel because it was the largest part of the inventory. Much of the HWCTR fuel has been sections, disrupted, and destructively examined. The bounding fuel was chosen for each property based on the individual fuel characteristics.

4.2.2 Chemical and Physical Properties

4.2.2.1 Air Oxidation Rate. No data exist for air oxidation of irradiated uranium-zirconium alloy fuel. Data for unirradiated uranium metal exist, but the irradiated alloy will not oxidize nearly as fast as the uranium metal. RW presently uses the value of zero for the air oxidation of CSNF. The repository will be wet when the waste package is breached. It is felt that the wet dissolution rate would be so much faster than the air oxidation that the air oxidation rate would be immaterial. For this reason, the value for air oxidation of uranium metal fuel was set to zero.

4.2.2.2 Wet Dissolution Rate. A review of the oxidation rates for metallic fuels was reported in Reference 1. Linear or paralinear kinetics was observed. Linear reaction rates were developed. In the case of paralinear kinetics, linear reaction rates were determined for times after initial parabolic oxide growth. Temperature dependent reaction rates were developed by standard regression methods. The recommended Arrhenius dependent reaction rates were reported. The reaction rate for the uranium-zirconium alloy water system for zirconium concentrations in the 20–97% range was reported to be

K_l = 1.13E+3 exp[-51.9 kJ/mole/RT] mg metal/cm^2 h for temperatures from 100–363°C. (22)

A bounding or worst-case rate for this group is the instantaneous release model, where one assumes that all the radionuclides are ready for transport when the container is breached.

4.2.2.3 Surface Area of the Matrix. The surface area of the matrix is necessary because the release rate from corrosion of the matrix is dependent on the surface area. The uranium metal alloy surface is an uncorroded condition would be relatively smooth, and the surface area could be calculated from the geometric dimensions of the fuel. Based on the HWCTR element dimensions of the fuel matrix

(5.8 × 290 cm) with the fuel being a tube with a hollow center, and using Equation (5), the geometric surface area is calculated to be 10,207 cm^2. The uranium loading end-of-life (EOL) averages 83,920 g matrix material. The calculated specific surface area is 1.2E-5 m^2/g. Adjusting this specific surface area with a roughness factor of 10 because of the disrupted nature of the fuel gives a surface area of 1.2E-4 m^2/g.

The other fuel types in the group were evaluated, using the appropriate equations. The thin plates of the Shippingport Pressurized Water Reactor (PWR) Core 1 assemblies have a combined surface area of 17,035 cm^2/assembly. Dividing the surface area by the matrix mass of 21.75 kg/assembly, the surface area is 7.8E-5 m/g. A roughness factor of 5 to account for the worst case gives a surface area of 3.1E-4 m/gram of matrix material. The Shippingport PWR fuel was determined to be the worst-case condition of this fuel for the group.

4.2.2.4 Volume of the Fuel. The volume of the fuel can be calculated from the dimensions of the fuel rods, plates, and assemblies, or it can be calculated from the mass of the matrix divided by the density of the matrix material. By using the volume of the fuel derived from the density of the fuel matrix, the results are very conservative because they neglect the volume of the cladding and structural materials.

By using Equation (9) for HWCTR fuel, the volume is calculated to be 6,936.9 cm^3/assembly.

By using the mass of an assembly and the density of the uranium alloy, which is 18.34, the volume is calculated to be 3,064 cm^3/assembly.

For this use, the 3,064 cm^3/assembly is most conservative. It allows the most water into the waste package. The National SNF database states that there will be 22 assemblies in a standard canister. The total volume of fuel in the standard canister will be 6.74E+4.

The Shippingport PWR fuel has the smallest volumes per assembly and will be packaged only one assembly to a standard canister. The Shippingport PWR fuel is the bounding fuel in this group at 851.8 cm^3 per standard canister.

4.2.2.5 Clad Failure Fraction. The HWCTR elements are tubes with some known to be disrupted. A clad failure fraction is assumed to be 1. The bounding case is that all the clad has failed, and no credit for the clad can be taken.

4.2.2.6 Free Radionuclide Inventory Fraction. Typical burnup and hence heat generation rates for commercial fuel is 50,000 MWD/MTU. No maximum burnup is reported in the National SNF database for the HWCTR fuel, but it believed to be low. Because of the low burnup and the age of the fuel, it is assumed that there was low fission product inventory. Therefore, the quantity of fission products released from the fuel matrix will be significantly less than CSNF. Because of the anticipated stability of the uranium-zirconium matrix, it is not anticipated that there will be a large release of fission products into the waste container prior to container failure. In addition the HWCTR fuel will be conditioned and repackaged, further reducing the free radionuclide fraction. HWCTR is the representative fuel in this group. The free radionuclide inventory fraction is assumed to be .00001 of the total radionuclide inventory, because it is expected to be very low as compared to CSNF. The bounding fraction is set at 10 times higher, or 0.0001 The HWCTR is also the bounding fuel for the group.

4.2.2.7 Gap Radionuclide Inventory Fraction. The fuel is formed without a gap between the fuel meat and the cladding. The gap inventory is set at zero because there is no gap.

4.2.2.8 Fuel Area Per Package. A loading per package was determined from the National SNF database. The surface area for each type of fuel in the group and the loading was used to determine the surface area in the package. For the HWCTR fuel the surface area is 10,207 cm^2/assembly. It is proposed that 22 assemblies be placed into a standard canister giving the value of 22.46 m^2/package. The HWCTR is also the worst-case fuel in this group.

4.2.2.9 Heat Generation. No heat generation data exist on these fuels. Based on analysis of other fuel types of the same age, it is believed that l0 W/assembly is conservatively high. Nine assemblies are assumed to be placed into one canister. The heat generation is estimated to be 90 W/canister.

4.2.3 Radiological Inventory

The radiological inventory for this group is contained in DOE/SNF/REP-078, *Source Term Estimates for DOE Spent Nuclear Fuels*.

4.2.4 Packaging

The number of the various types of containers are:

- 8—18-in. × 15-ft standard canister.

4.2.5 Criticality Controls

Criticality analysis will be performed on each fuel type for transportation and storage of intact fuel assemblies and canisters. Criticality analysis for the degraded modes will be performed on a worst-case fuel.

4.3 Uranium-Molybdenum Alloy

4.3.1 Group Description

There are eight separate entries in the National SNF database for this group. Fermi driver Cores 1 and 2 are the major fuels in the group. There are seven containers of HWCTR 3 EMT-2 from two assemblies (.0081 MTHM) and a small quantity (.0024 MTHM) of SPEC (ORME) fuel also in the group. Fermi driver was chosen as the representative fuel type. The molybdenum content ranges from 2 to 10 wt%. The bounding fuel was chosen for each property based on the individual fuel characteristics.

4.3.2 Chemical and Physical Properties

4.3.2.1 Air Oxidation Rate. No data exist for this parameter. RW presently uses the value of zero for CSNF. The repository will be wet when the waste package is breached. It was concluded that the wet dissolution rate would be so much faster than the air oxidation that the air oxidation rate would be immaterial. For this reason the value for the air oxidation of uranium-molybdenum alloy fuel was set to zero.

4.3.2.2 Wet Dissolution Rate. A review of the oxidation rates for metallic fuels was reported in Reference 1. Linear or paralinear kinetics was observed, but they were different for alloys containing greater than or less than 8 wt% molybdenum. Linear reaction rates were developed. In the case of paralinear kinetics, linear reaction rates were determined for times after initial parabolic oxide growth. The initial reaction rate with water was lower than the corresponding reaction of pure uranium metal by one to three orders of magnitude. However, the reaction rates in tests with small samples often accelerated

after a long time. The apparent accelerated rate corresponded to discontinuous failure and concomitant with severe cracking. Discontinuous failure probably occurs by preferential attack along nonhomogeneous secondary phases that results in cracks and an increased surface area. This phenomenon is sometimes referred to as matrix breakup and occurs after a long time period.

Temperature dependent reaction rates were developed by standard regression methods. The recommended Arrhenius dependent reaction rates were reported. The reaction rate of the uranium metal water system bounded the reaction rate of all other systems and was reported to be

K_l = 1.15E+8 exp[-66.5 kJ/mole/RT] mg matrix/cm^2 h for temperatures from 100–178°C and molybdenum less than 8 wt%. (23)

Because of the breakup phenomenon that has been observed, the reaction rate for this group should be the same as the rate for uranium metal, which is one and one half magnitudes faster than that reported for the uranium-molybdenum alloy.

The bounding or worse-case rate for this group should be the instantaneous rate.

4.3.2.3 Surface Area of the Matrix. The surface area of the matrix is necessary because the release rate from corrosion of the matrix is dependent on the surface area. The uranium-molybdenum metal alloy surface in an uncorroded condition would be relatively smooth, and the surface area could be calculated from the geometric dimensions of the fuel. Based on the Fermi element dimensions of the fuel matrix (0.4 × 84 cm) with the fuel being a rod, and by using Equation (3), the geometric surface area is calculated to be 105.5 cm^2/rod. The uranium loading EOL averages 147 g uranium-molybdenum per rod. The calculated specific surface area is 7.18E-5 m^2/g. Adjusting this specific surface area with a roughness factor of 5 gives a surface area of 3.6E-4 m^2/g.

The fuel with the largest surface area and the smallest diameter will have the largest surface area per gram of uranium. All fuel types in this group were evaluated and used the appropriate equations to determine the bounding surface area. The HWCTR with a surface area of 3,468 cm^2/assembly and a mass of 1.29 kg uranium-molybdenum/assembly has a calculated surface area of 2.7E-4 m^2/g. A roughness factor of 10 should be used to account for the worst-case condition, thereby making the bounding surface area 2.7E-3 m^2/g.

4.3.2.4 Volume of the Fuel. The volume of the fuel can be calculated from the dimensions of the fuel rods, plates, and assemblies, or it can be calculated from the mass of the matrix divided by the density of the matrix material. By using the volume of the fuel derived from the density of the fuel matrix, the results are very conservative because they neglect the volume of the cladding and structural materials.

By using Equation (9) for Fermi fuel, the volume is calculated to be 10.55 cm^3/rod.

By using the mass of an assembly, 160 g, and the density of the uranium molybdenum alloy, which is 17.5, the volume is calculated to be 9.1 cm^3/rod.

For this use, the 9.1 cm^3/assembly is most conservative because it allows the most water into the waste package. The National SNF database states that there will be 3,840 rods in a standard canister. The total volume of fuel in the standard canister will be 4.1E +4 cm^3/ standardized canister.

The worst-case fuel in this group was determined to be the Fermi fuel.

4.3.2.5 Clad Failure Fraction. Most of the Fermi fuel pins in this group are reported to have intact cladding; 1,956 of 29,952 (6.5%) have had their cladding breached intentionally. We must assume 0.1 cladding failures fraction based on the existing data. The bounding case is that all the clad has failed, and no credit for the clad can be taken.

4.3.2.6 ***Free Radionuclide Inventory Fraction.*** Typical burnup and hence heat generation rates for commercial fuel are 50,000 MWD/MTU. An average burnup for Fermi fuel of 1,600 and maximum burnup of 2,765 is reported in the National SNF database. Because of the age and low burnup of the fuel, it is assumed that there was low fission product inventory. Therefore, the quantity of fission products released from the fuel matrix will be significantly less than CSNF. Because of the anticipated stability of the uranium-molybdenum matrix and the intact cladding, it is not anticipated that there will be a large release of fission products into the waste container prior to container failure. In addition the Fermi fuel will be conditioned and repackaged, further reducing the free radionuclide fraction. Fermi is the representative fuel in this group. The free radionuclide inventory fraction is assumed to be 0.00001 of the total radionuclide inventory because it is expected to be very low as compared to CSNF. The bounding case is assumed to be a factor of 10 worse, or 0.0001.

4.3.2.7 Gap Radionuclide Inventory Fraction. The fuel is formed without a gap between the fuel meat and the cladding. The gap inventory is set at zero because there is no gap.

4.3.2.8 Fuel Area Per Package. The loading per package was determined from the National SNF database. The surface area for each type of fuel in the group and the loading was used to determine the surface area in the package. For the Fermi fuel the surface area is 105.5 cm^2/rod. It is proposed that 3,840 rods be placed into a standard canister giving the value of 4.05E+3 m^2/package. The Fermi fuel is also the worst-case fuel in this group.

4.3.2.9 Heat Generation. Based on ORIGEN-2 data, the worst-case heat generation rate for the Fermi driver core is 1.1 W/assembly. With 27.4 equivalent assemblies per waste canister, the worst-case heat generation rate is 30 W/waste canister.

4.3.3 Radiological Inventory

The radiological inventory for this group is contained in DOE/SNF/REP-078, *Source Term Estimates for DOE Spent Nuclear Fuels.*

4.3.4 Packaging

The numbers of the various types of containers are:

- 8—18-in. × 10-ft standard canister.

4.3.5 Criticality Controls

Criticality analysis will be performed on each fuel type for transportation and storage of intact fuel assemblies and canisters. Criticality analysis for the degraded modes will be performed on a worst-case fuel.

4.4 Intact Uranium Oxide

4.4.1 Group Description

There are 73 separate entries in the National SNF database for this group, containing approximately 70 MTHM. The group is dominated by typical commercial fuel. A Westinghouse 15 × 15 commercial assembly was chosen to represent the group, because it is the single largest contributor to the group. The bounding fuel was chosen for each property based on the individual fuel characteristic.

4.4.2 Chemical and Physical Properties

4.4.2.1 Air Oxidation Rate. Limited data exist for this parameter. RW presently uses zero for CSNF. The repository will be wet when the waste package is breached. It was felt that the wet dissolution rate would be so much faster than the air oxidation rate that the air oxidation rate would be immaterial. For this reason the value was set to zero.

4.4.2.2 Wet Dissolution Rate. A dissolution rate is recommended that was developed by RW for CSNF. This is conservatively high because much of the uranium oxide DOE SNF has a ceramic matrix, like Shippingport PWR, and will have a much slower dissolution rate than the typical commercial fuel. The group is represented by CSNF, and the dissolution rate, as reported in Reference 2, is:

$$\text{Log}_{10}(\text{rate}) = a_0 + a_1/T_k + a_2 * pCO_3 + a_3 * pO_2 + a_4 * pH \quad (24)$$

where

a_0 ….. a_4 = constants

T_k = waste package temperature (K)

pCO_3 = $-\log_{10}$ (molar concentration of CO_3)

pH = $-\log_{10}$ of molar hydrogen ion concentration inside the waste package

pO_2 = $-\log_{10}$ of the O_2 concentration inside the waste package

rate = rate of dissolution in mg/m^2–day.

	Intrinsic Dissolution Rate Coefficients				
In-package pH	a_0	a_1	a_2	a_3	a_4
pH>7	4.69	-1085	-0.12	-0.32	0
pH<7	7.13	-1085	0	-0.32	-0.41

where the term $[CO_3]$ is the total carbonate concentration in units of mole/L and $[O_2]$ is the oxygen overpressure in atmospheres.

This expression for alkaline conditions transforms to the more usual kinetics format

$$k = 5 \times 10^4 [CO_3]^{0.12} [O_2]^{0.32} \exp[-20.8 \text{ kJ/mole/RT}] \text{ mg } UO_2/m^2 d \quad . \quad (25)$$

For acidic conditions the rate is

$$k = 1.35 \times 10^{7} [H^{+}]^{0.41} [O_2]^{0.32} \exp[-20.8 \text{ kJ/mole/RT}] \text{ mg } UO_2/m^2d \quad . \qquad (26)$$

The CSNF also represents the bounding or worst-case dissolution rate for the group because the other fuels all have slower dissolution rates.

4.4.2.3 Surface Area of the Matrix. The specific surface area for a Westinghouse 15 ×15 assembly that contains 206 fuel rods is calculated by using Equation (3) to be 2.04E-5 m^2/g. The surface area for a BWR assembly ranges from 4.3E-4 m^2/g to 7.9E-4 m^2/g. A value was derived from a number of references for CSNF and is given as 9.5E-4 m^2/g. This value should be used as the representative value. The fuel from the dry rod consolidation test contains fuel rods that have undergone a consolidation of rods so that twice as many rods occupy the same space. Therefore, the surface per assembly should be doubled. However, the specific surface area per gram of uranium should be the same because the mass also doubles.

4.4.2.4 Volume of the Fuel. The volume of the fuel can be calculated from the dimensions of the fuel rods, plates, and assemblies, or it can be calculated from the mass of the matrix divided by the density of the matrix material. By using the volume of the fuel derived from the density of the fuel matrix, the results are very conservative because they neglect the volume of the cladding and structural materials.

By using Equation (9) for a 15 × 15 commercial fuel assembly, the volume is calculated to be 9.470E+3 cm^3/assembly.

By using the mass of a 15 × 15 assembly, 460 kg, and the density of the uranium oxide, which is 10.96, the volume is calculated to be 4.197E+4 cm^3/assembly.

For this use, the 9.470E+3 cm^3/assembly is most conservative, because it allows the most water into the waste package. PWR fuel will be placed into packages with 21 assemblies per waste package. The total volume in a waste package will be 199,000 cm^3/waste package.

The worst-case fuel in this group was determined to be HWCTR fuel assembly. This fuel will be packaged 22 assemblies per standard canister for a total volume of 55,000 cm^3/standardized canister.

4.4.2.5 Clad Failure Fraction. The credit that can be taken for the containment of the radionuclide inventory in the fuel by the clad is dependent on the condition of the clad and the material properties of the clad material. Cladding composed of alloys of zirconium have demonstrated during testing sufficient corrosion resistance for credit to be taken. Other cladding materials have not demonstrated the same corrosion resistance. The fuel in this group is CSNF and is assumed to be intact and has only minor cracks and pin holes. For these conditions, the Nuclear Regulatory Commission and DOE-RW have taken credit for the clad integrity. For this group of fuels, the same clad failure fraction should be used as DOE-RW uses for intact zirconium-clad commercial fuel. The bounding case is the same as used by RW for commercial fuel.

4.4.2.6 Free Radionuclide Inventory Fraction. Because the clad is intact, there is no free radionuclide inventory for this fuel group. In addition, the conditions within the sealed repository disposal containers are benign and not likely to facilitate degradation of the fuel to produce a free inventory prior to the degradation of the waste package. The bounding free radionuclide inventory for this group is based on commercial fuel. Data from the monitoring of the gas inside the dry storage casks of commercial fuel at the INEEL have indicated that the free inventory of gaseous fission products is extremely low (the

same as commercial fuel) and the inventory of semivolatile radionuclides is below detection limits. The bounding case should be the same as used by RW for commercial fuel.

4.4.2.7 Gap Radionuclide Inventory Fraction. The RW Analysis/Model report, *CSNF Waste Form Degradation: Summary Abstraction*, ANL-EBS-MD-000015, Revision 0 gives the main isotopes in the gap and grain boundaries to be cesium, technetium, strontium, and iodine. It states that the gap inventory for CSNF is composed of two main isotopes, I-129 and Cs-137. The measured values are reported as a function of the fission gas release. For I-129 it is 1, and for Cs-137 it is 1/3. The report states that the quantity is driven primarily by the degree of friability of the fuel matrix and the temperature of the fuel. The previously used value of 2% was reported to be nonconservative for CSNF. The bounding case should be the same as used by RW for commercial fuel.

4.4.2.8 Fuel Area Per Package. If we assume that the package loaded with commercial fuel is a 21-assembly waste canister, the surface area per package is 446 m^2/package. An average loading of MTHM per package was determined from the MTHM and the package totals for all fuels in this category. This was used with the specific surface area determined above and appropriate conversion factors to calculate an average fuel meat surface area per package for this group. The value derived for this group is 500 m^2/package. This would be the bounding value.

4.4.2.9 Heat Generation. Based on repository requirements, the heat generation rate will be less than 1,500 W/assembly. If the CSNF is placed into a 21-assembly waste canister the total heat generation will be 31.5 kW/canister.

4.4.3 Radiological Inventory

The radiological inventory for this group is contained in DOE/SNF/REP-078, *Source Term Estimates for DOE Spent Nuclear Fuels*.

4.4.4 Packaging

The numbers of the various types of containers are:

- 60—18-in. × 10-ft standard canister
- 138—18-in. × 15-ft standard canister
- 18—MCO
- 14—21 commercial assembly canisters.

4.4.5 Criticality Controls

Criticality analysis will be performed on each fuel type for transportation and storage of intact fuel assemblies and canisters. Criticality analysis for the degraded modes will be performed on a worst-case fuel. The highly enriched uranium (HEU) fuel will be codisposed, and the low-enriched uranium (LEU) fuel will be disposed of in large diameter waste canisters.

4.5 Failed Uranium Oxide

4.5.1 Group Description

There are 33 separate entries in the National SNF database for this group, containing over 83 MTHM. The group is dominated by Three Mile Island Unit 2 (TMI-2) debris containing over 81 MTHM. TMI-2 was chosen as the representative fuel because it consists of more than 90% of the mass of the group. Other fuels in this group are completely disrupted down to the powder form. The bounding fuel was chosen for each property based on the individual fuel characteristics.

4.5.2 Chemical and Physical Properties

4.5.2.1 Air Oxidation Rate. Limited data exist for this parameter. RW presently uses zero for CSNF. The repository will be wet when the waste package is breached. It was felt that the wet dissolution rate would be so much faster than the air oxidation that the air oxidation rate would be immaterial. For this reason the value was set to zero.

4.5.2.2 Wet Dissolution Rate. A dissolution rate was used that was developed by RW for CSNF. This is conservative because some of the uranium oxide DOE SNF has a ceramic matrix and will have a much slower dissolution rate than the typical commercial fuel. The group is represented by CSNF, and the dissolution rate, as reported in Reference 2, is:

$$\text{Log}_{10}(\text{rate}) = a_0 + a_1/T_k + a_2 * pCO_3 + a_3 * pO_2 + a_4 * pH \quad (27)$$

where

$a_0 \ldots a_4$ = constants

T_k = waste package temperature (K)

pCO_3 = $-\log_{10}$ (molar concentration of CO_3)

pH = $-\log_{10}$ of molar hydrogen ion concentration inside the waste package

pO_2 = $-\log_{10}$ of the O_2 concentration inside the waste package

rate = rate of dissolution in mg/m^2–day.

	Intrinsic Dissolution Rate Coefficients				
In-package pH	a_0	a_1	a_2	a_3	a_4
pH>7	4.69	-1085	-0.12	-0.32	0
pH<7	7.13	-1085	0	-0.32	-0.41

For the TMI-2 fuel, the sections that were melted were converted into a ceramic type material that would have a lower dissolution rate than CSNF. The fuel was exposed to water for many years, so any easily dissolved species would have been removed prior to repository emplacement.

Some of the fuels in this group are uranium oxide powders that are not sintered and pressed into pellets. To accommodate those fuels in this group, the bounding wet dissolution should be 10 times the CSNF dissolution rate.

4.5.2.3 Surface Area of the Matrix. The specific surface area for a Westinghouse 15 × 15 assembly that contains 206 fuel rods is calculated to be 2.04E-5 m^2/g by using Equation (3). The specific surface area for a BWR assembly ranges from 4.3E-4 m^2/g to 7.9E-4 m^2/g. A value was derived from a number of references for CSNF and is given as 9.5E-4 m^2/g.

Because of the TMI-2 reactor accident, the potential surface area will be larger than that of normal CSNF. Some of the fuel in this group is in a powder form. The Fort Saint Vrain (FSV) fuel has a surface area per gram of 5.9E-3 m^2/g to 2.2E-2 m^2/g, depending on the particle size distribution. Based on a particle size of 200 μm in diameter, the surface area is 2.6E-3 m^2/g heavy metal. Based on these data points, the representative and the worst-case surface area for this group would be 2.2E-2 m^2/g.

4.5.2.4 Volume of the Fuel. The volume of intact fuel can be calculated from the dimensions of the fuel rods, plates, and assemblies, or it can be calculated from the mass of the matrix divided by the density of the matrix material. By using the volume of the fuel derived from the density of the fuel matrix, the results are very conservative because they neglect the volume of the cladding and structural materials.

By using Equation (9) for a 15 × 15 intact commercial fuel assembly, the volume is calculated to be 9.47E+3 cm^3/assembly.

By using the mass of a 15 × 15 assembly, 460 kg, and the density of the uranium oxide, which is 10.96, the volume is calculated to be 4.197E+4 cm^3/assembly. For this group the use of the 4.2E+4 cm^3/assembly is most conservative because it allows the most water into the waste package. PWR fuel will be placed into packages, 21 assemblies per waste package. The total volume in a waste package will be 1,194,000 cm^3.

For the failed uranium oxide fuel, there is no structure like an assembly. The density of the material is also uncertain, because it will not be close to the theoretical density. Therefore, the only way to address the volume of the fuel is to calculate the volume of the container that houses the fuel. The TMI-2 container is the largest container in the group. The usable volume for containing fuel is 22.8 × 22.8 × 381 cm or 1.98E+5 cm^3. This is also the bounding volume.

4.5.2.5 Clad Failure Fraction. Because of the disruptive nature of the TMI-2 accident, it should be assumed that 100% of the cladding has failed.

4.5.2.6 Free Radionuclide Inventory Fraction. All the fuel in this group has breached cladding in some degree. This breach in the clad allows the fission products that migrate through the fuel matrix into the gas spaces surrounding the fuel. All the fuels in this group are old and relatively cool, so the driving force for additional migration is low. Prior to sealing the fuel in the standard canister the fuel will be conditioned and dried, thus removing any existing free radionuclide inventory. Therefore, the free radionuclide inventory fraction is 0.

4.5.2.7 Gap Radionuclide Inventory Fraction. Not all the fuel was melted in the TMI-2 accident. There are many partial assemblies that still have their cladding intact, and potentially some gap inventory that has not been dissolved. A conservative estimate would be to treat the fuel like commercial fuel.

The RW Analysis/Model report, *CSNF Waste Form Degradation: Summary Abstraction*, ANL-EBS-MD-000015, Revision 0 gives the main isotopes in the gap and grain boundaries to be cesium, technetium, strontium, and iodine. It states that the gap inventory for CSNF is composed of two main isotopes, I-129 and Cs-137. The measured values are reported as a function of the fission gas release. For I-129 it is 1, and for Cs-137 it is 1/3. The report states that the quantity is driven primarily by the degree of friability of the fuel matrix and the temperature of the fuel. For the bounding case use the fraction of 1.

4.5.2.8 Fuel Area Per Package. If we assume an average uranium content for each of the waste packages and TMI-2 canisters is packaged four to a waste package, the surface area in the waste package will be 2.1E4 m^2/canister. By multiplying by a factor of 10 for conservatism, the worst-case would be 2E5 m^2/canister.

4.5.2.9 Heat Generation. Based on ORIGEN-2 data, the worst-case heat generation rate for the TMI-2 fuel is 3.7 W/assembly. The other fuel in the group is older, and therefore, the heat generation rate would be less than the TMI-2. An average of two assemblies is contained in each TMI-2 canister, and there are assumed to be four canisters in a waste package. The average heat generation rate is calculated to be 30 W per waste package. The worst case based on maximum possible loading would be 300 W.

4.5.3 Radiological Inventory

The radiological inventory for this group is contained in DOE/SNF/REP-078, *Source Term Estimates for DOE Spent Nuclear Fuels*.

4.5.4 Packaging

The numbers of the various types of containers are:

- 182—18-in. × 10-ft standard canister
- 347—18-in. × 15-ft standard canister
- 56—high integrity container.

4.5.5 Criticality Controls

Criticality analysis will be performed on each fuel type for transportation and storage of intact fuel assemblies and canisters. Criticality analysis for the degraded modes will be performed on a worst-case fuel. The HEU fuel will be codisposed with high-level waste (HLW) glass logs in the same waste canister, and the LEU fuel will be disposed in large diameter waste canisters.

4.6 Uranium-Aluminum Based

4.6.1 Group Description

There are 208 entries in the National SNF database for this type of fuel. The Advanced Test Reactor (ATR) fuel represents 4% of the mass of the group but is the single largest contributor. Because there are good data available on this fuel, it was chosen to represent the group. The bounding fuel was chosen for each property based on the individual fuel characteristics.

4.6.2 Chemical and Physical Properties

4.6.2.1 Air Oxidation Rate. No data exist for the dry oxidation of irradiated aluminum. There are data for the air oxidation of unirradiated aluminum. In normal air oxidation, the extent of oxidation is self-limiting at a given temperature. RW presently uses zero for CSNF. The repository will be wet when the waste package is breached. It was felt that the wet dissolution rate would be so much faster than the air oxidation that the air oxidation rate would be immaterial. For this reason the value was set to zero.

4.6.2.2 Wet Dissolution Rate. A review of the oxidation rates for metallic fuels was reported in Reference 1. Linear or paralinear kinetics was observed. Linear reaction rates were developed. In the case of paralinear kinetics, linear reaction rates were determined for times after initial parabolic oxide growth. Temperature-dependent reaction rates were developed by standard regression methods. The recommended Arrhenius dependent reaction rates were reported. The oxidation rates of aluminum-based dispersion fuels are controlled by the oxidation of the aluminum alloy matrix. The reaction rate of the aluminum metal-water system bounded the reaction rate of other systems and was reported to be

$K_l = 4.29 \exp[-32.8 \text{ kJ/mole/RT}]$ mg metal/cm^2 /hr for temperatures from 25–360°C . (28)

A bounding or worst-case rate for the aluminum-based system is the uranium metal release model, which is 10,000 times faster than the aluminum-based reaction.

4.6.2.3 Surface Area of the Matrix. The ATR fuel was used as the representative fuel in this group. Based on the geometry of an ATR fuel assembly and the weight of the fuel meat (including the uranium and the aluminum), the specific surface area is 1.3E-3 m^2/g. Using a roughness factor of 5 the specific surface area recommended is 6.5E-3 m^2/g.

The group was examined for all the different fuel types, geometries, materials, and configurations using the appropriate equations to determine the bounding fuel. Twelve different fuels were chosen and evaluated. The bounding fuel for this group is the OMEGA West fuel that has a surface area of 1.6E-2 m^2/g of meat. Using a surface-roughness factor of 5, the bounding specific surface area is 8.0E-2 m^2/g of fuel meat.

4.6.2.4 Volume of the Fuel. The volume of the fuel can be calculated from the dimensions of the fuel rods, plates, and assemblies, or it can be calculated from the mass of the matrix divided by the density of the matrix material. By using the volume of the fuel derived from the density of the fuel matrix, the results are very conservative because they neglect the volume of the cladding and structural materials.

By using the appropriate equation for each fuel assembly, the volumes were calculated. The ATR fuel is the representative fuel for this group. Based on the geometry of the assembly, the volume is 1,923 cm^3. And by using the uranium fuel meat density and the mass of the fuel assembly, the volume is calculated to be 764 cm^3.

For this use, the 7.64E+2 cm^3/assembly is most conservative because it allows the most water into the waste package.

The worst-case fuel in this group was determined to be the ASTRA fuel because it has the smallest volume per assembly. The ASTRA fuel has a calculate volume of 1.04E+3 cm^3. Using the density of the fuel meat compound and the mass of the fuel the volume was calculated to be 8.6E+2 cm^3. The 7.6E+2 cm^3 should be used for the bounding value.

The volume per waste package was determined to be 3.8E4 cm^3/waste package for the ATR fuel. The bounding volume for this group was calculated to be for the Advanced Reactor Measurement Facility fuel with 1.9E4 cm^3/waste package

4.6.2.5 Clad Failure Fraction. In the repository environment, the aluminum cladding has a relatively short life expectancy. It is conservative not to take credit for the aluminum cladding, and it should be assumed that it is 100% failed.

4.6.2.6 Free Radionuclide Inventory Fraction. These fuels will be conditioned and repackaged, reducing the free radionuclide fraction. ATR fuel is both the representative and bounding fuel in this group. The free fraction was arbitrarily set at 0.001, based on the discussion above. The bounding fraction is set at 10 times higher, or 0.01. There are no supporting analytical data for this value.

4.6.2.7 Gap Radionuclide Inventory Fraction. The aluminum-based fuel is formed with the cladding thermally bonded to the sintered matrix, which essentially has no gap. The gap inventory will be less than that of CSNF, so a value of 0 is appropriate.

4.6.2.8 Fuel Area Per Package. Based on the projected number of assemblies per standard canister and the calculated surface area per assembly, the surface area per canister for the representative ATR fuel for this group is 77 m^2/canister. When the surface area including the roughness factor is included the value is 385 m^2/canister.

The bounding fuel for this parameter is the MURR (Missouri University Research Reactor) fuel, which has a surface area of 430 m^2/canister including the roughness factor.

4.6.2.9 Heat Generation. Based on ORIGEN-2 data, the worst-case ATR fuel has a heat generation rate of 117 W/assembly. Based on a waste canister loading of 20 ATR assemblies, the worst case is 2,340 W/waste canister. This is also the bounding value.

4.6.3 Radiological Inventory

The radiological inventory for this group is contained in DOE/SNF/REP-078, *Source Term Estimates for DOE Spent Nuclear Fuels*.

4.6.4 Packaging

- 1,029—18-in. × 10-ft standard canister
- 235—18-in. × 15-ft standard canister.

4.6.5 Criticality Controls

Criticality analysis will be performed on each fuel type for transportation and storage of intact fuel assemblies and canisters. Criticality analysis for the degraded modes will be performed on a worst-case fuel. The uranium-235 enrichment in these fuels ranges from 10% to 95%. The fuels will be codisposed with HWL glass logs in the same waste container to ensure that criticality safety requirements are met.

4.7 High Integrity Uranium-Thorium Carbide

4.7.1 Group Description

There are four entries in the National SNF database for this group. FSV fuel is selected to represent the group, as it is 95% of the mass of the group. There is also some Peach Bottom Core 2 fuel and a very small quantity of General Atomic high temperature gas reactor (HTGR) fuel that was made identical to the FSV fuel. The bounding fuel for the group was chosen for each property based on the individual fuel characteristics.

4.7.2 Chemical and Physical Properties

4.7.2.1 Air Oxidation Rate. Uranium dicarbide, UC_2, oxidizes readily to UO_2 and CO_2:

$$UC_2 + 3\ O_2 \rightarrow UO_2 + 2\ CO_2\ . \quad (29)$$

The oxidation of UC_2 in dry oxygen follows a parabolic rate law, with rate data for temperatures in the range of 150–250°C given by the following Arrhenius equation:

$$K = 1.05\ 10^{22} \exp[-91.5\ kJ/mole/RT]\ mg^2\ UC_2/m^4\ d\ . \quad (30)$$

Above 250°C the reaction rate is instantaneous.

While the uranium-thorium carbide rapidly reacts in water, it is protected by an encapsulation layer of silicon carbide. Silicon carbide (SiC) is a robust nonoxide ceramic that is highly resistant to oxidation. For this reason, SiC is being used in numerous applications that require high-temperature strength and resistance to oxidation. It has been used in nuclear fuels as the robust, impermeable barrier coating on $(U,Th)C_2$ fuel kernels in FSV graphite fuel.

SiC oxidation in the temperature range of 800–1,200°C is bounded by:

$$k = 3.12 \times 10^9 \exp[-94.3\ kJ/mol/RT]\ mg^2\ SiC/m^4 d\ . \quad (31)$$

Extrapolation of this relationship to temperatures significantly lower than 800°C may be problematic, because a different mechanism may operate at lower temperatures, such as logarithmic growth kinetics. At some temperature, the reaction will become so slow that the oxide thickness for transition from linear to parabolic may not be reached in a reasonable time limit, or the microstructure of the oxide may not result in an effective barrier layer. In this case, the reaction follows linear kinetics with the rate given by:

$$K = 4.03E9 \exp [-159\ kJ/mole/RT]\ mg\ SiC/m^2\ d\ . \quad (32)$$

4.7.2.2 Wet Dissolution Rate. The hydrolysis of UC_2 proceeds extremely rapidly in water. Hydrolysis of UC_2 produces UO_2, hydrogen, and a mixture of aliphatic hydrocarbons, in which ethane and methane are the primary constituents.

Reaction of porous samples is an apparent function only of the bulk surface area, not the specific surface area due to porosity and particle size. Rate constants based on bulk surface area define the observed hydrolysis rate better than when normalized to the BET specific surface area. This indicated that the reaction is so rapid at the surface that diffusion of oxygen or moisture into pore structure does not play

a large role in the reaction. Reaction of UC_2 at 50°C is 0.25 μg/cm^{2}sec, or 216 g/m^{2}d. The weight gain data correspond to the following hydrolysis rate for UC_2 in water vapor:

$$K = 5.37 \times 10^{11} \exp[-35.05 \text{ kJ/mole/RT}] \text{ mg } UC_2/m^2 \text{ d} \quad . \tag{33}$$

The hydrolysis of ThC_2 produces primarily hydrogen (20–60%) plus a mixture of methane and higher alkenes and alkanes. Face-centered cubic ThO_2 was the only identified inorganic hydrolysis product. Thorium carbide and thorium-containing UC_2 hydrolyze in moist air about 10 times faster than UC_2 and at ambient temperatures, <50°C, form the hydrous oxide $ThO_2 \bullet xH_2O$. The bounding rate for the reaction rate with water is instantaneous reaction.

The oxidation of silicon carbide by water or water vapor proceeds similar to the oxidation in air or oxygen. As in oxidation by air or oxygen, the hydrolysis reaction yields SiO_2 as the solid product, but the gaseous products are CO, CO_2, CH_4, and hydrogen.

The hydrolysis mechanism is comparable to the oxidation mechanism and generally follows similar kinetics. As in oxidation by air or oxygen, the initial phase of hydrolysis proceeds by linear kinetics as the water reacts with the pristine silicon carbide surface. As the protective SiO_2 layer builds and impedes water diffusion to the reaction interface, the kinetics transitions from linear to parabolic kinetics. However, because of the much greater solubility of water in SiO_2, the effective diffusion rate of water (i.e., permeation rate) is much faster than the diffusion rate of oxygen. Therefore, a much thicker SiO_2 film, on the order of 500–1,000 nm, is required to force the transition to parabolic kinetics. In addition, water or water vapor can effect the silicon carbide reaction by hydrolyzing the SiO_2 reaction product to form silicon hydroxide species. The bounding rate of parabolic hydrolysis of silicon carbide by water is:

$$K = 2.75E14 \exp[-2.02 \text{ kJ/mole/RT}] \text{ mg}^2 \text{ SiC/m}^4 \text{ d} \quad . \tag{34}$$

The effects of irradiation on silicon carbide oxidation have been investigated. They measured the weight change due to oxidation in dry oxygen of unirradiated silicon carbide and the weight change of silicon carbide samples that were neutron irradiated to fluences of $1.2–4.5 \times 10^{24}$ n/m^2 (E>1 MeV). The irradiated silicon carbide data from Yano were essentially the same as their unirradiated data, which suggest that there is no significant effect of irradiation to these fluences on the oxidation of silicon carbide.

4.7.2.3 Specific Surface Area of the Matrix. The specific surface area for the FSV fuel was calculated from the fuel design parameters by the INEEL. Based on the particle diameter and heavy metal content, the surface area ranges from 5.9 E-3 m^2/g heavy metal to 2.2E-2 m^2/g heavy metal. The largest surface area is selected to represent the group with no additional surface-roughening factor used.

The Peach Bottom Core 2 fuel was manufactured with a similar process, but without the final silicon carbide coating. To conservatively bound the entire group the highest surface area is multiplied by a factor of 5. This equates to a bounding surface area for the group of 1.1E-1 m^2/g heavy metal.

4.7.2.4 Volume of the Fuel. The volume of the fuel can be calculated from the dimensions of the fuel rods, plates, and assemblies, or it can be calculated from the mass of the matrix divided by the density of the matrix material. By using the volume of the fuel derived from the density of the fuel matrix, the results are very conservative because they neglect the volume of the cladding and structural materials.

By using the appropriate equation for each fuel assembly, the volumes were calculated. The FSV fuel is the representative fuel for this group. Based on the geometry of the assembly, the volume is

87,710 cm^3/assembly. This is also the bounding fuel for the group. The other fuel in the group is Peach Bottom fuel, which has a volume of 22,690 cm^3/assembly.

The volume per waste package is calculated from the volume per assembly and the number of assemblies in a waste package. The representative value for the FSV fuel is 4.4E5 cm^3/waste package. The bounding fuel is the Peach Bottom fuel that has 3.0E5 cm^3/waste package.

4.7.2.5 Clad Failure Fraction. Analysis of the FSV fuel after irradiation indicated less than a 1% failure or a fraction of 0.01 of the particles. There were no data on the clad failure fraction for the Peach Bottom Core 2 fuel. The Peach Bottom Core 2 fuel did not have the silicon carbide coating on the fuel particle, and it is possible that more of the particles could have ruptures. Therefore, the bounding cladding failure fraction should be 0.05.

4.7.2.6 Free Radionuclide Inventory Fraction. The silicon carbide coating is very impervious to fission product transport. The pyrolitic carbon coating is also impervious except for cesium. Based on this, the fission product distribution among all the elements, the less than 1% coating failure of the FSV fuel pellets, and the long-term storage in a vented facility, the free inventory fraction for the FSV fuel is less than 0.00001. The contribution of the Peach Bottom Core 2 fuel is small because of the small percentage to the MTHM of the group. However, the Peach Bottom fuel does not have a silicon carbide coating, and the long-term performance of the pyrolitic carbon coating is not known. Therefore, to bound this group, the commercial fuel model is recommended.

Typical burnup and hence heat generation rates for commercial fuel is 50,000 MWD/MTU. The Peach Bottom Core 2 burnup is 30,000–70,000 MWD/MTU. However the fuel will have cooled for over 35 years. This fuel will be conditioned and repackaged, reducing the free radionuclide fraction. The free fraction was arbitrarily set at 0.001, based on the discussion above. The bounding fraction is set at 10 times higher, or 0.01. There are no supporting technical analytical data for this value.

4.7.2.7 Gap Radionuclide Inventory Fraction. The pyrolytic carbon coating was designed to absorb fission products, so the gap inventory should be zero.

4.7.2.8 Fuel Area Per Package. An average loading of MTHM per package was determined from the projected loading of the standard canister for all fuels in this category. This was used with the specific surface area determined above and appropriate conversion factors to calculate the bounding surface area per package for this group. The bounding and representative fuel for this parameter is the FSV fuel with a value of 1,276 m^2/package.

4.7.2.9 Heat Generation. The heat generation rate for the FSV fuel is estimated to be 50 W/assembly. With 5 assemblies in a canister, the heat generation rate is 250 W/canister.

4.7.3 Radiological Inventory

The radiological inventory for this group is contained in DOE/SNF/REP-078, *Source Term Estimates for DOE Spent Nuclear Fuels*.

4.7.4 Packaging

- 503—18 in. × 15 ft standard canister

4.7.5 Criticality Controls

Criticality analysis will be performed on each fuel type for transportation and storage of intact fuel assemblies and canisters. Criticality analysis for the degraded modes will be performed on a worst-case fuel. The fuel is all HEU and will be codisposed with HLW glass logs in the same waste container.

4.8 Low Integrity Uranium-Thorium Carbide

4.8.1 Group Description

Peach Bottom Core 1 fuel and scrap is the major fuel in the group. There is a small quantity of HTGR fuel.

4.8.2 Chemical and Physical Properties

4.8.2.1 Air Oxidation Rate. Uranium dicarbide, UC_2, oxidizes readily to UO_2 and CO_2:

$$UC_2 + 3\ O_2 \rightarrow UO_2 + 2\ CO_2 \quad . \tag{35}$$

The oxidation of UC_2 in dry oxygen follows a parabolic rate law with rate data for temperatures in the range of 150–250°C given by the following Arrhenius equation:

$$K = 1.05 \times 10^{22} \exp[-91.5\ \text{kJ/mole/RT}]\ \text{mg}^2\ UC_2/\text{m}^4\ \text{d} \quad . \tag{36}$$

The uranium carbide reacts with the air when the particle coating is breached. Presently 60 to 80% of the particles are breached. The oxidation of the uranium carbide will potentially release some fission products from the fuel. There are indications that the oxidation could proceed rapidly under the proper conditions so the instantaneous model is chosen to bound the group.

4.8.2.2 Wet Dissolution Rate. The hydrolysis of UC_2 proceeds much faster than uranium carbide. Hydrolysis of UC_2 produces UO_2, hydrogen, and a mixture of aliphatic hydrocarbons, in which ethane and methane are the primary constituents.

Reaction of porous samples is an apparent function only of the bulk surface area, not the specific surface area due to porosity and particle size. Rate constants based on bulk surface area define the observed hydrolysis rate better than when normalized to BET specific surface area. This indicated that the reaction is so rapid at the surface that diffusion of oxygen or moisture into pore structure does not play a large role in the reaction. Reaction of UC_2 at 50°C is 0.25 μg/cm^{2}sec or 216 g/m^{2}d. The weight gain data correspond to the following hydrolysis rate for UC_2 in water vapor:

$$K = 5.37 \times 10^{11} \exp[-35.05\ \text{kJ/mol/RT}]\ \text{mg}\ UC_2/\text{m}^2\ \text{d} \quad . \tag{37}$$

The hydrolysis of ThC_2 produces primarily hydrogen (20–60%) plus a mixture of methane and higher alkenes and alkanes. Face-centered cubic ThO_2 was the only identified inorganic hydrolysis product.

Thorium carbide and thorium-containing UC_2 hydrolyze in moist air about 10 times faster than UC_2; hydrolysis of ThC_2 in water vapor forms the hydrous oxide $ThO_2 \cdot xH_2O$ for reactions below 50°C. To bound this group, the instantaneous model was chosen.

4.8.2.3 Specific Surface Area of the Matrix. Based on the similar design parameters to the FSV fuel, the surface area ranges from 5.9E-3 m^2/g heavy metal to 2.2E-2 m^2/g heavy metal. To conservatively bound the entire group, the highest surface area is multiplied by a factor of 5. This equates to a bounding surface area for the group of 1.1E-1 m^2/g heavy metal.

4.8.2.4 Volume of the Fuel. The volume of the fuel can be calculated from the dimensions of the fuel rods, plates, and assemblies, or it can be calculated from the mass of the matrix divided by the density of the matrix material. By using the volume of the fuel derived from the density of the fuel matrix, the results are very conservative because they neglect the volume of the cladding and structural materials. By using the appropriate equation for each fuel assembly, the volumes were calculated. The fuel in the group is Peach Bottom Core 1 fuel, which has a volume of 22,690 cm^3/assembly. The volume per waste package is calculated from the volume per package and the number of assemblies in a waste package. Both the representative and bounding values are for the Peach Bottom fuel with 3E5 cm^3/waste package.

4.8.2.5 Clad Failure Fraction. Based on fuel examination, 60% to 80% of the fuel particles of the Peach Bottom Core 1 are failed. A cladding failure fraction is assumed to be 0.6 to 0.8. The bounding clad failure fraction is 1.

4.8.2.6 ***Free Radionuclide Inventory Fraction.*** The fuel will be vacuum dried, and the free inventory will be swept away. After drying, some of the inventory may migrate into the free space from the fuel matrix. Typical burnup and hence heat generation rates for commercial fuel is 50,000 MWD/MTU. Peach Bottom Core 1 fuel maximum burnup is reported to be 31,000 MWD/MTU. Therefore, because of the lower burnup and fission product inventory, the longer cooling time, the low heat generation rate, and the physical structure of the fuel matrix, the quantity of fission products released from the fuel matrix will be significantly less than commercial fuel.

A conservative fraction value of 0.5 was selected because characterization data have shown that many fission products (cesium, barium, strontium, hydrogen, carbon) were released from the fuel particles and were removed by the purge gas and deposited in the fission product trap inside the assembly. The fuel will be bounded by commercial fuel.

4.8.2.7 Gap Radionuclide Inventory Fraction. The gap in this fuel is assumed to be the pyrolytic carbon coating that is between the fuel meat and the outside of the particle. The coating was designed to absorb fission products. For those fuel particles that are breached, some fission products may accumulate after they are released by the air oxidation of the uranium carbide. The gap inventory will be assumed to be 0.5 of the inventory. The bounding fraction will be 1.

4.8.2.8 Fuel Area Per Package. An average loading of MTHM per package was determined from the National SNF database and the package totals for all fuels in this category. This was used with the specific surface area determined above and appropriate conversion factors to calculate an average fuel meat surface area per package for this group. The value derived for this group is 470 m^2/package.

4.8.2.9 Heat Generation. Based on ORIGEN-2 data, the maximum projected heat rate for the Peach Bottom fuel is 3.7 W/assembly. Based on a loading of 13 assemblies per canister, the heat generation rate is 48 W/canister.

4.8.3 Radiological Inventory

The radiological inventory for this group is contained in DOE/SNF/REP-078, *Source Term Estimates for DOE Spent Nuclear Fuels*.

4.8.4 Packaging

- 64—18 in. × 15 ft standard canister

4.8.5 Criticality Controls

Criticality analysis will be performed on each fuel type for transportation and storage of intact fuel assemblies and canisters. Criticality analysis for the degraded modes will be performed on a worst-case fuel. The fuel is all HEU and will be codisposed with HLW glass logs in the same waste container.

4.9 Nongraphite Uranium Carbide

4.9.1 Group Description

There are five entries in the National SNF database for this group; four of which are Fast Flux Test Facility (FFTF) carbide fuel. FFTF carbide assemblies were chosen to represent the group because of the number of entries. The FFTF carbide fuel was constructed from uncoated uranium and plutonium monocarbide spheres that were loaded directly into the fuel pins or were pressed into pellets that were loaded into the pins. The bounding fuel for the group was chosen for each property based on the individual fuel characteristics.

4.9.2 Chemical and Physical Properties

4.9.2.1 Air Oxidation Rate. Uranium monocarbide has poor resistance to oxidation and corrosion, but reacts more slowly in air than UC_2. The oxidation rate is dependent on the sample history; prior ageing in humid air increases the rate of oxidation. While freshly cast uranium carbide oxidizes very slowly even at 500°C in air, ageing in moist air can make aged uranium carbide pyrophoric in air at 350°C. The oxidation of uranium carbide is strongly exothermic, with a ΔG of –302 kcal/mole at 700 K (Matzke86) and will oxidize to CO and CO_2 as stated in reference 2.

Above 55°C, the oxidation rate is very fast and is limited by the oxygen diffusion to the reaction surface. Below 550°C, which is the expected repository condition, the data follow linear kinetics with a reaction rate given by:

$$K = 1.74\ 10^{15} \exp[-104\ \text{kJ/mole/RT}]\ \text{mg UC/m}^2\ \text{d} \quad . \tag{38}$$

which is the bounding rate.

Oxidation tests at 60–160°C indicated parabolic kinetics with a temperature dependence of the oxidation rate given by

$$K = 2.6\ \text{E+18} \exp[-105\ \text{kJ/mole/RT}]\ \text{mg}^2\ \text{UC/m}^4\text{d} \quad , \tag{39}$$

which is the representative rate.

The oxidation of the carbides will potentially release some fission products from the fuel matrix that could accumulate in the gap for early release. These will be accounted for in an increased gap inventory.

4.9.2.2 Wet Dissolution Rate. The hydrolysis of uranium carbide in moist air or water proceeds substantially faster than the oxidation in air and forms primarily methane (≈80%), hydrogen, and

hydrocarbon gases. The entire hydrolysis reaction can be adequately described by a linear rate law of the simple form

$$M = kt \quad . \tag{40}$$

Reference 2 reported a change in mechanism in reaction with liquid water at approximately 60 to 80°C, with a greater temperature dependence of the rate below 60°C than above. At 20 to 60°C, the rate constant for hydrolysis in liquid water is

$$k = 7.0 \times 10^{17} \exp[-61 \text{ kJ/mole/RT}] \text{ mg UC/m}^2\text{d} \quad , \tag{41}$$

whereas the rate constant for 60°C to 90°C is

$$k = 8.9 \times 10^{10} \exp[-17 \text{ kJ/mole/RT}] \text{ mg UC/m}^2\text{d} \quad . \tag{42}$$

In general, the hydrolysis rate of uranium carbide decreases with increasing irradiation. To bound this group the instantaneous model was chosen.

4.9.2.3 Surface Area of the Matrix. Based on a particle size of 200-μm diameter, the surface area is 2.6E-3 m^2/g heavy metal. This was chosen as both the representative and the bounding value for the group.

4.9.2.4 Volume of the Fuel. The volume of the fuel can be calculated from the dimensions of the fuel rods, plates, and assemblies, or it can be calculated from the mass of the matrix divided by the density of the matrix material. By using the volume of the fuel derived from the density of the fuel matrix, the results are very conservative because they neglect the volume of the cladding and structural materials.

By using the appropriate equation for each fuel assembly, the volumes were calculated. The FFTF fuel is the representative fuel for this group. Based on the geometry of the assembly, the volume is 5,215 cm^3/assembly. This is also the bounding fuel for the group. The other fuel in the group is a small quantity of scrap. The volume of the fuel per waste package is given by the volume of the assembly and multiplied by the number of assemblies per waste package. The representative and bounding number is that of the FFTF fuel and is 8.3E4 cm^3/waste package.

4.9.2.5 Clad Failure Fraction. The cladding is stainless steel, and there are a few reports of damage of leaking rods. Because the cladding is stainless steel, and stainless steel has a limited lifetime in the repository environment, a conservative assumption is to assume that 100% of the cladding has failed.

4.9.2.6 Free Radionuclide Inventory Fraction. The fuel will be vacuum dried, and the free inventory will be swept away. After drying, some of the inventory may migrate into the free space from the fuel matrix. There are no data available on how the fuel is expected to perform in the repository. Based on the low thermal output of the fuel, which is 2 W/assembly, it is conservative to model the fuel like commercial fuel. This is both representative and bounding for the group.

4.9.2.7 Gap Radionuclide Inventory Fraction. Based on the similarity between the construction of the FFTF and CSNF i.e., pellets of fuel matrix contained in a metal cladding tube, this fuel is treated as CSNF.

The RW Analysis/Model report, *CSNF Waste Form Degradation: Summary Abstraction*, ANL-EBS-MD-000015, Revision 0 gives the main isotopes in the gap and grain boundaries to be cesium, technetium, strontium, and iodine. It states that the gap inventory for CSNF is composed of two main

isotopes, I-129 and Cs-137. The measured values are reported as a function of the fission gas release. For I-129 it is 1, and for Cs-137 it is 1/3. The report states that the quantity is driven primarily by the degree of friability of the fuel matrix and the temperature of the fuel.

4.9.2.8 Fuel Area Per Package. An average loading of MTHM per package was determined from the MTHM and the package totals for all fuels in this category . This was used with the specific surface area determined above and appropriate conversion factors to calculate an average fuel meat surface area per package for this group. The value derived for this group is 76 m^2/package.

4.9.2.9 Heat Generation. The heat generation rate for these FFTF assemblies is estimated to be 2 W/assembly. The fuel will be loaded an average of 46 assemblies per canister, resulting in an average heat generation rate of 92 W/canister. The maximum loading will be 66 assemblies per canister so the maximum heat generation rate would be 132 W/canister.

4.9.3 Radiological Inventory

The radiological inventory for this group is contained in DOE/SNF/REP-078, *Source Term Estimates for DOE Spent Nuclear Fuels*.

4.9.4 Packaging

- 2—18 in. × 10 ft standard canister
- 2—18 in. × 15 ft standard canister.

4.9.5 Criticality Controls

Criticality analysis will be performed on each fuel type for transportation and storage of intact fuel assemblies and canisters. Criticality analysis for the degraded modes will be performed on a worst-case fuel. The effective enrichment of this fuel is greater than 15%. The fuel will be codisposed with HLW glass logs in the same waste canister to control criticality.

4.10 Mixed Oxide Fuel (MOX)

4.10.1 Group Description

There are 42 entries in the National SNF database for MOX fuel. Twelve of those entries are FFTF MOX test assemblies that are located at Hanford. These test assemblies contain 10.42 MTHM or over 80% of the MTHM of the group. The FFTF MOX test assemblies were chosen as the representative fuel for the group. All the fuel meat in the group was made by sintering and pressing mixtures of UO_2 and PuO_2. The bounding fuel for the group was chosen for each property based on the individual fuel characteristics.

4.10.2 Chemical and Physical Properties

4.10.2.1 Air Oxidation Rate. PuO_2 oxidizes very slowly at about 0.65% of the UO_2 oxidation rate. This relative rate of reaction is consistent with observations of the relative release rates of plutonium in the oxidation of MOX fuel samples. In the oxidation of MOX fuel samples, the PuO_2 component reacted more than 10 times more slowly than the matrix UO_2. The air oxidation rates for intact commercial fuel should be used for this group.

RW presently uses zero for CSNF. The repository will be wet when the waste package is breached. It was felt that the wet dissolution rate would be so much faster than the air oxidation that the air oxidation rate would be immaterial. For this reason the value was set to zero.

4.10.2.2 Wet Dissolution Rate. PuO_2 oxidizes very slowly at about 0.65% of the UO_2 oxidation rate. This relative rate of reaction is consistent with observations of the relative release rates of plutonium in the oxidation of MOX fuel samples. In the oxidation of MOX fuel samples, the PuO_2 component reacted more than 10 times more slowly than the matrix UO_2.

The oxidation rate of PuO_2 is

$$k = 0.165 \exp[-39 \text{ kJ/mole/RT}] \text{ mole } PuO_2/m^2d \ , \quad (43)$$

or

$$k = 44.6 \exp[-39 \text{ kJ/mole /RT}] \text{ g } PuO_2/m^2d \ . \quad (44)$$

The wet dissolution rate for intact commercial fuel should be used as bounding for this group.

This expression for alkaline conditions is

$$k = 5 \times 10^4 [CO_3]^{0.12} [O_2]^{0.32} \exp[-20.8 \text{ kJ/mole/RT}] \text{ mg } UO_2/m^2d \ . \quad (45)$$

For acidic conditions the rate is

$$k = 1.35 \times 10^7 [H^+]^{0.41} [O_2]^{0.32} \exp[-20.8 \text{ kJ/mole/RT}] \text{ mg } UO_2/m^2d \ . \quad (46)$$

4.10.2.3 Specific Surface Area of the Matrix. All the fuel is constructed of sintered pellets similar to commercial fuel. By using the appropriate equations, the calculated surface area for the FFTF assembly is 1.1E-4 m^2/g heavy metal. With a surface roughness factor of 5, the surface area is 5.5E-4 m^2/g heavy metal. The surface area for commercial fuel is 9.5E-4 m^2/g and should be used for the worst-case or bounding surface area because some of the fuel in this group is just like CSNF.

4.10.2.4 Volume of the Fuel. The volume of the fuel can be calculated from the dimensions of the fuel rods, plates, and assemblies, or it can be calculated from the mass of the matrix divided by the density of the matrix material. By using the volume of the fuel derived from the density of the fuel matrix, the results are very conservative because they neglect the volume of the cladding and structural materials.

By using Equation (9) for the FFTF fuel assembly the volume is calculated to be

$$5.215 \text{ E+3 } cm^3/\text{assembly} \ . \quad (47)$$

By using the number of assemblies per waste package, the representative volume for the FFTF fuel is 8.3E4 cm^3/waste package. And by using the appropriate equations, the worst-case fuel for the group is some EBR-II MOX pins that will have 2.0E3 cm^3/waste package.

4.10.2.5 Clad Failure Fraction. The FFTF fuel is stainless steel clad and an assumption of 0.1 fraction failures is made based on other stainless steel clad SNF. The bounding fuel in this group is the Pacific Northwest Laboratory fuel located at Sandia National Laboratory that has no cladding, so the bounding clad failure fraction is 1.

4.10.2.6 Free Radionuclide Inventory Fraction. The free radionuclide inventory for this group is based on commercial fuel. Prior to placement of the fuel into the standard canister it will be conditioned. There will be no free inventory, and the free inventory that develops over the storage life will depend on the driving force on the radionuclides to pass through the clad defects, such as internal pressure and heat. These fuels are old and cold so there is very little driving force. For this group, the free radionuclide inventory is assumed to be 0. The bounding value should be the same as commercial fuel.

4.10.2.7 Gap Radionuclide Inventory Fraction. The gap radionuclide inventory for this group is based on commercial fuel. The RW Analysis/Model report, *CSNF Waste Form Degradation: Summary Abstraction*, ANL-EBS-MD-000015, Revision 0 gives the main isotopes in the gap and grain boundaries to be cesium, technetium, strontium, and iodine. It states that the gap inventory for CSNF is composed of two main isotopes, I-129 and Cs-137. The measured values are reported as a function of the fission gas release. For I-129 it is 1, and for Cs-137 it is 1/3. The report states that the quantity is driven primarily by the degree of friability of the fuel matrix and the temperature of the fuel.

4.10.2.8 Fuel Area Per Package. There will be five FFTF assemblies in a standard canister so the fuel area per package will be five times the surface area of an FFTF assembly or 89 m^2/package.

An average loading of MTHM per package was determined from the MTHM and the package totals for all fuels in this category. This was used with the specific surface area determined above and appropriate conversion factors to calculate an average fuel meat surface area per package for this group. The value derived for this group is 89 m^2/package. The 89 m^2/package is the most conservative value for the FFTF fuel.

The surface area for commercial fuel is 9.5E-4 m^2/g. This value should be used for the worst-case or bounding surface area because some of the fuel in this group is just like CSNF. The Saxton MOX will be packaged, 32 assemblies per standard canister, so it will have the bounding surface area per package of 170 m^2.

4.10.2.9 Heat Generation. It is estimated that the heat generation rate for an FFTF assembly is 1.8 kW. It will be loaded up to five assemblies per canister, so the heat generation rate is 9 kW/package.

4.10.3 Radiological Inventory

The radiological inventory for this group is contained in DOE/SNF/REP-078, *Source Term Estimates for DOE Spent Nuclear Fuels*.

4.10.4 Packaging

- 22—18 in. × 10 ft standard canister
- 131—18 in. × 15 ft standard canister
- 3—BWR commercial assembly casks.

4.10.5 Criticality Controls

Criticality analysis will be performed on each fuel type for transportation and storage of intact fuel assemblies and canisters. Criticality analysis for the degraded modes will be performed on a worst-case fuel. The effective enrichment of the MOX fuel is greater than 15%. The fuel will be codisposed with HLW glass logs in the same waste container to control criticality.

4.11 Uranium-Thorium Oxide

4.11.1 Group Description

Shippingport LWBR was chosen as the representative fuel because it comprises 8 of the 12 records and represented approximately 85% of the MTHM inventory in this group. The remainder of the group is composed of 1,000 Dresden I rods, 190 Elk River Reactor assemblies, and some scrap material. The bounding fuel for the group was chosen for each property based on the individual fuel characteristic.

4.11.2 Chemical and Physical Properties

4.11.2.1 Air Oxidation Rate. The most stable oxidation state for thorium is Th(IV). In fact, no higher oxidation states are available to thorium, so that the chemistry of thorium is dominated by the Th(IV) oxidation state. The only stable oxide phase is thorium dioxide, ThO_2. Consequently, ThO_2 cannot oxidize further and is stable in air and water. Any reactions involving ThO_2 are limited to redox reactions that involve reduction of Th(IV) to a lower oxidation state, acid-base reactions that transform the thorium oxide to salts or compounds of the same or lower oxidation state, and solvation or chelation reactions.

4.11.2.2 Wet Dissolution Rate. ThO_2 cannot oxidize further and is stable in air and water. Any reactions involving ThO_2 are limited to redox reactions that involve reduction of Th(IV) to a lower oxidation state, acid-base reactions that transform the thorium oxide to salts or compounds of the same or lower oxidation state, and solvation or chelation reactions. It is conservative to assume the same dissolution rate as CSNF.

This expression for alkaline conditions transforms to the more usual kinetics format

$$k = 5 \times 10^4 [CO_3]^{0.12} [O_2]^{0.32} \exp[-20.8 \text{ kJ/mole/RT}] \text{ mg } UO_2/m^2d \quad . \tag{48}$$

For acidic conditions the rate is

$$k = 1.35 \times 10^7 [H^+]^{0.41} [O_2]^{0.32} \exp[-20.8 \text{ kJ/mole/RT}] \text{ mg } UO_2/m^2d \quad . \tag{49}$$

4.11.2.3 Surface Area of the Matrix. Based on the geometry and the heavy metal loading of a LWBR fuel pellet that includes a roughness factor of 5, the surface area is 3.6E-4 m^2/g heavy metal. Based on the rod dimensions and the heavy metal loading per rod, the bounding surface area is 1.1E-4 m^2/g heavy metal. Because some of the material in the group is scrap and leftover material from destructive examinations, the bounding surface is assumed to be 10 times the LWBR surface area, or 3.6E-3 m^2/g.

4.11.2.4 Volume of the Fuel. The volume of the fuel can be calculated from the dimensions of the fuel rods, plates, and assemblies, or it can be calculated from the mass of the matrix divided by the density of the matrix material. By using the volume of the fuel derived from the density of the fuel matrix, the results are very conservative because they neglect the volume of the cladding and structural materials.

By using Equation (9) for LWBR seed fuel assembly, the volume is calculated to be

$$9.1 \text{ E+4 } cm^3/\text{assembly} \quad . \tag{50}$$

The volume for the representative fuel per waste package is calculated to be 9.1E4 cm^3/waste package.

The worst-case fuel in this group is the Elk River Reactor fuel that has 3.7E4 cm^3/waste package.

4.11.2.5 Clad Failure Fraction. Based on the known 7% of the LWBR inventory that has been destructively examined, it is estimated that 0.1 fraction of the LWBR cladding has failed. The bounding case is to assume that all the cladding has failed and the clad failure fraction is 1.

4.11.2.6 Free Radionuclide Inventory Fraction. The free radionuclide inventory for this group is based on commercial fuel. Prior to placement of the fuel into the standard canister it will be conditioned. There will be no free inventory, and the free inventory that develops over the storage life will depend on the driving force on the radionuclides to pass through the clad defects, such as internal pressure and heat. These fuels are old and cold so there is very little driving force. For this group, the free radionuclide inventory is assumed to be part of the gap inventory. For the bounding inventory, it is assumed to be the same as commercial SNF.

4.11.2.7 Gap Radionuclide Inventory Fraction. The gap radionuclide inventory for this group is based on commercial fuel because of the similarities in construction. The RW Analysis/Model report, *CSNF Waste Form Degradation: Summary Abstraction*, ANL-EBS-MD-000015, Revision 0 gives the main isotopes in the gap and grain boundaries to be cesium, technetium, strontium, and iodine. It states that the gap inventory for CSNF is composed of two main isotopes, I-129 and Cs-137. The measured values are reported as a function of the fission gas release. For I-129 it is 1, and for Cs-137 it is 1/3. The report states that the quantity is driven primarily by the degree of friability of the fuel matrix and the temperature of the fuel.

4.11.2.8 Fuel Area Per Package. The surface area per package was based on the surface area of the LWBR assembly rods, and it is assumed that the fuel would be loaded one assembly per package. This calculation gives an average surface area of 61 m^2/package for the blanket assembly, which is the worst of the LWBR assemblies. The bounding fuel is KEMA that has an area of 634 m^2/package.

4.11.2.9 Heat Generation. Based on ORIGEN-2 data for Shippingport LWBR fuel, the maximum heat generation rate is 120 W/assembly. The fuel will be packaged 1 assembly per canister. This bounds the other fuels in the group.

4.11.3 Radiological Inventory

The radiological inventory for this group is contained in DOE/SNF/REP-078, *Source Term Estimates for DOE Spent Nuclear Fuels*.

4.11.4 Packaging

- 11—18 in. × 10 ft standard canister
- 17—18 in. × 15 ft standard canister
- 27—24 in. × 15 ft standard canister.

4.11.5 Criticality Controls

Criticality analysis will be performed on each fuel type for transportation and storage of intact fuel assemblies and canisters. Criticality analysis for the degraded modes will be performed on a worst-case fuel. This HEU fuel will be codisposed with HLW glass logs in the same waste container to ensure criticality safety.

4.12 Uranium-Zirconium-Hydride

4.12.1 Group Description

TRIGA comprises 98 % of this group, 143 of 146 entries in the National SNF database, and will be used as the representative fuel. There are several different types of TRIGA based on uranium-235 enrichment, uranium loading, and cladding material. However, the fuel matrix, the construction, and the size are the same. The bounding fuel for the group was chosen for each property based on the individual fuel characteristics.

4.12.2 Chemical and Physical Properties

4.12.2.1 Air Oxidation Rate. Zirconium hydride, ZrH_2, is one of the few metal hydrides with a high resistance to oxidation even at high temperatures. Its thermal and chemical stability have been important factors in the successful use of zirconium hydride as the fuel matrix in TRIGA reactors. However, information on the oxidation kinetics of ZrH_2 is extremely scarce in the open literature. The published data for the oxidation kinetics of ZrH_2 are limited almost entirely to qualitative or anecdotal observations. Qualitative reports in the literature indicate that ZrH_2 is more resistant to oxidation by air or water and to spontaneous ignition in air than the parent metal zirconium. Furthermore, the higher the hydrogen content of the zirconium hydride, the higher is the oxidation resistance. The relative chemical stability of ZrH_2 results from a thin tenacious oxide layer on the hydride surface. While the intrinsic reactivity of fresh ZrH_2 may be high, oxidation of the hydride surface creates a highly protective oxide layer that restricts oxygen diffusion. The excellent barrier characteristics of the ZrO_2 layer on ZrH_2 is demonstrated by the effective retention of hydrogen by ZrH_2 at temperatures as high as 950°C. For this reason the value was set to zero. The bounding rate is:

$$K = 4.58E5 \exp [-12.3 \text{ kJ/mole/RT}] \text{ mg}^2 \text{ ZrH}_2/\text{m}^4 \text{ d} \quad . \tag{51}$$

4.12.2.2 Wet Dissolution Rate. While the information on the kinetics of air oxidation of ZrH_2 is sparse and consists largely of qualitative observations, the available information on the reaction with water is even more limited. Quantitative kinetics studies have not been found for the oxidation of ZrH_2 in water or water vapor; the literature reports are composed only of qualitative observations. Nevertheless, the qualitative observations consistently have indicated that ZrH_2 is relatively inert to steam or water. Because of its native protective oxide film, ZrH_2 is relatively stable in water and steam up to at least 100°C, and reacts very slowly even at higher temperatures in steam. The reaction of a sample of 8 wt% U-$ZrH_{1.0}$ in boiling water resulted in the slow oxidation of the hydride and spallation of the oxide, with a cumulative weight loss of 2×10^{-4} g/cm^2 after exposure for 90 days. It is assumed that the spallation of the oxide signifies linear kinetics, and this translates to a weight loss rate of 2.2×10^{-6} g ZrO_2/cm^2 • d or 0.017 g ZrH_2/m^2 • d. In the only other report of quantitative corrosion rates, bare fuel specimens were heated in steam at 570°F (300°C) and 1,230 psi pressure for 400 hours. An observed weight gain rather than a weight loss as observed in boiling water and the formation of an adherent oxide film. It was reported that the maximum extent of corrosion penetration after 400 hr was less than 2 mils.

By assuming parabolic reaction kinetics, the formation of 'less than 2 mils' in 400 hours indicates a parabolic reaction rate that is less than 2×10^{-6} g^2 ZrH_2/cm^{4}hr or 5×10^3 g^2 ZrH_2/m^{4}hr.

For the bounding rate it is conservative to assume the same dissolution rate as CSNF.

This expression for alkaline conditions is

$$k = 5 \times 10^4 [CO_3]^{0.12} [O_2]^{0.32} \exp[-20.8 \text{ kJ/mole/RT}] \text{ mg } UO_2/\text{m}^2\text{d} \quad . \tag{52}$$

For acidic conditions the rate is

$$k = 1.35 \times 10^{7} [H^{+}]^{0.41} [O_2]^{0.32} \exp[-20.8 \text{ kJ/mole/RT}] \text{ mg } UO_2/m^2d \quad . \qquad (53)$$

4.12.2.3 Specific Surface Area of the Matrix. Based on geometry and the average TRIGA uranium zirconium hydride loading (8.5% uranium) per rod with a surface roughness factor of 5, the specific surface area is 1 E-4 m^2/g of the fuel matrix. The size of the rod is standard; however, the weight of the matrix varies. Based on the lightest loading to bounding surface area would be 1.1 E-4 m^2/g matrix.

4.12.2.4 Volume of the Fuel. The volume of the fuel can be calculated from the dimensions of the fuel rods, plates, and assemblies, or it can be calculated from the mass of the matrix divided by the density of the matrix material. By using the volume of the fuel derived from the density of the fuel matrix, the results are very conservative because they neglect the volume of the cladding and structural materials.

By using Equation (9) for TRIGA fuel rod, the volume is calculated to be 400 cm^3/assembly. This is also the worst-case volume for the group. The representative volume per waste package is 1.1E5 cm^3/waste package. The bounding volume per waste package, based on various waste package loadings, is 8.5E3 cm^3/waste package.

4.12.2.5 Clad Failure Fraction. Based on the fraction of the fuel that is either declad or that is aluminum clad for which no cladding credit is taken, the fraction of failed cladding is 0.10. The bounding case for this group is the declad material, which would have a clad failure fraction of 1.

4.12.2.6 Free Radionuclide Inventory Fraction. Because of the anticipated stability of the uranium-zirconium-hydride matrix, the stability of the ZrO_2 layer on the outside of the fuel matrix, it is not anticipated that there will be a large release of fission products into the waste container prior to container failure. The free radionuclide inventory fraction is assumed to be .00001 of the total radionuclide inventory. The bounding value for the group was conservatively estimated to be 10 times the representative value or 0.0001. There is no technical basis for the number.

4.12.2.7 Gap Radionuclide Inventory Fraction. The fission products are contained well in the matrix, but there is a design gap between the matrix and the cladding. The recoil release of the TRIGA fuel matrix is 1.5E-5 and is independent of fuel temperature. Below 400°C, the fractional release due to thermal diffusion through the matrix is less than 1.5E-5. Therefore, the gap fraction for the TRIGA fuel should be 1.5E-5.

The bounding case for this group was conservatively estimated to be 10 times the representative value or 0.00015.

4.12.2.8 Fuel Area Per Package. Based on the fuel rod surface area and a projected loading of 111 rods in a package, the area per package is calculated to be 4.8 m^2/package.

An average loading of MTHM per package was determined from the MTHM and the package totals for all fuels in this category. This was used with the specific surface area determined above and appropriate conversion factors to calculate an average fuel meat surface area per package for this group. The value derived for this group by this method is 22 m^2/package.

4.12.2.9 Heat Generation. Based on ORIGEN-2 data for TRIGA fuel, the average heat generation rate is 1 W/assembly. The fuel will be packaged 111 assemblies per canister making the heat generation rate 111 W/package . Based on the worst-case fuel, the bounding heat generation rate would be 5.5 kW/package.

4.12.3 Radiological Inventory

The radiological inventory for this group is contained in DOE/SNF/REP-078, *Source Term Estimates for DOE Spent Nuclear Fuels*.

4.12.4 Packaging

- 81—18 in. × 10 ft standard canister.

4.12.5 Criticality Controls

Criticality analysis will be performed on each fuel type for transportation and storage of intact fuel assemblies and canisters. Criticality analysis for the degraded modes will be performed on a worst-case fuel. The fuel is HEU and will be codisposed.

4.13 Miscellaneous

4.13.1 Group Description

This group consists of material that has minimal characterization data. There are 10 entries in the National SNF database for this group. The largest portion, as measured in MTHM, is a miscellaneous fuel at Argonne National Laboratory-West that is most likely uranium metal or alloy. This makes up more than 88% of the MTHM in the group. The bounding fuel for the group was chosen for each property based on the individual fuel characteristics.

4.13.2 Chemical and Physical Properties

4.13.2.1 Air Oxidation Rate. Uranium oxidation process has been studied for many years, but a good understanding of the mechanism has only been obtained fairly recently. These data are for unirradiated uranium metal. The irradiated uranium metal may oxidize at a faster rate than the unirradiated uranium metal. The linear reaction rate for long time periods for this reaction is:

$$k = 1.09\ E{+}8\ \exp[-71.3\ kJ/mole/RT]\ mg\ U/cm^2\ h\ . \quad (54)$$

The bounding value should be the same as commercial fuel.

4.13.2.2 Wet Dissolution Rate. There is a large variety of fuel in this group, from metal to oxide to nitride to salt. Many of these fuels are not well characterized. The largest fuel, as measured in MTHM, is a miscellaneous fuel at Argonne National Laboratory-West that is most likely uranium metal or alloy. A review of the oxidation rates for metallic fuels was reported in Reference 1. Linear or paralinear kinetics was observed. Linear reaction rates were developed. In the case of paralinear kinetics, linear reaction rates were determined for times after initial parabolic oxide growth. Temperature dependent reaction rates were developed by standard regression methods. The recommended Arrhenius dependent reaction rates were reported. The reaction rate of the uranium metal water system bounded the reaction rate of all other systems and was reported to be

$$K_l = 5.03\ E{+}9\ \exp[-66.4\ kJ/mole/RT]\ mg\ U/cm^2\ h \text{ for temperatures from 20–300°C.} \quad (55)$$

A bounding or worst-case rate for the metallic uranium system is the instantaneous release model, where one assumes that all the radionuclides are ready for transport when the container is breached.

4.13.2.3 Surface Area of the Matrix. The actual condition of the material is unknown. It is conservative to treat the surface area as if it were a particle such as the graphite fuels. The surface area for the FSV fuel was calculated from the fuel design parameters by the INEEL. The surface area ranges from 5.9E-3 m^2/g heavy metal to 2.2E-2 m^2/g heavy metal. The largest surface area is selected to represent the group with no additional surface roughness factor used.

The Peach Bottom Core 2 fuel was manufactured with a similar process, but without the final silicon carbide coating. To conservatively bound the entire group the highest surface area is multiplied by a factor of 5. This equates to a bounding surface area for the group of 1.1E-1 m^2/g heavy metal.

4.13.2.4 Volume of the Fuel. If one assumes that some of the material is a uranium metal or alloy, and that the lowest density for that type of material is 6.88, the volume of the fuel can be calculated from the uranium mass. The volume for all the mass in this group is 6.2E5 cm^3. The configuration in which the material will be packaged has not yet been determined. The maximum volume of a 10-ft-long, 18-in.-diameter standard canister is 6.5E4 cm^3. This is smaller and more limiting than assuming a maximum fissile material content of only 5 kg per canister. Bt using this assumption, 10 canisters will be required to contain all of this group. This equates to 6.2E4 cm^3/per package.

4.13.2.5 Clad Failure Fraction. Much of the fuel in this group is scrap from fuel development programs and from analysis of fuel remains. No documentable characterization data exist on any of the cladding conditions. A conservative approach is to assume that all the cladding is failed and not give any credit for the protection of the fuel meat by the cladding. The clad failure fraction is assumed to be 1 for both the representative and the bounding case.

4.13.2.6 Free Radionuclide Inventory Fraction. This group of SNF will need to be dried and conditioned prior to packaging for the repository. Both of these treatments may involve heating the fuel. The heating may release some of the fission products from the matrix and into the canister where they will be removed prior to sealing the canister. Because of this process, the free radionuclide inventory fraction is set to zero.

4.13.2.7 Gap Radionuclide Inventory Fraction. This fuel was probably manufactured without a gap between the clad and the fuel matrix. However, because there is insufficient information about the fuel, the worst-case is assumed to be commercial fuel. The RW Analysis/Model report, *CSNF Waste Form Degradation: Summary Abstraction*, ANL-EBS-MD-000015, Revision 0 gives the main isotopes in the gap and grain boundaries to be cesium, technetium, strontium, and iodine. It states that the gap inventory for CSNF is composed of two main isotopes, I-129 and Cs-137. The measured values are reported as a function of the fission gas release. For I-129 it is 1, and for Cs-137 it is 1/3. The report states that the quantity is driven primarily by the degree of friability of the fuel matrix and the temperature of the fuel.

4.13.2.8 Fuel Area Per Package. An average loading of MTHM per package was determined from the MTHM and the package totals for all fuels in this category. This was used with the specific surface area determined above and appropriate conversion factors to calculate an average fuel meat surface area per package for this group. The value derived for this group is 4.7E4 m^2/package.

4.13.2.9 Heat Generation. Most of this fuel is old and has very small heat generation rates. Because there are no data available and worst-case heat generation rate data are wanted, it will be assumed that the heat generation rate will be less than CSNF or 31.5 kW/canister.

4.13.3 Radiological Inventory

The radiological inventory for this group is contained in DOE/SNF/REP-078, *Source Term Estimates for DOE Spent Nuclear Fuels*.

4.13.4 Packaging

- 10—18 in. × 10 ft standard canister.

4.13.5 Criticality Controls

Criticality analysis will be performed on each fuel type for transportation and storage of intact fuel assemblies and canisters. Criticality analysis for the degraded modes will be performed on a worst-case fuel. Much or the fuel is HEU and will be disposed with mass limits for criticality control.

5. REFERENCES

1. Review of Oxidation Rates of DOE Spent Nuclear Fuel Part 1, Metallic Fuel, DOE/SNF/REP-054, Revision 0

2. *Review of Oxidation Rates of DOE Spent Nuclear Fuel Part 2. Nonmetallic Fuel*, DOE/SNF/REP-068, Revision 0