

# **SCIENTIFIC NOTEBOOK # 1098E**

## **Development of an Integration Model to Evaluate Affect of Residual Water on Fuel and Cladding Oxidation**

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# Table of Contents

|  |    |
|--|----|
| INITIAL ENTRIES .....                                  | 3  |
| Model Development.....                                 | 4  |
| Integration Model.....                                 | 4  |
| Inputs.....  | 4  |
| Calculation Sequences .....                            | 5  |
| Residual Water Amount.....                             | 12 |
| Radiolysis Kinetics .....                              | 12 |
| Fuel and Cladding Temperature.....                     | 13 |
| Decay Rate of Fuel and Cladding Temperature.....       | 13 |
| Cladding Failure Rate .....                            | 13 |
| Mechanisms for Oxygen Contacting the Exposed Fuel..... | 13 |
| Model Simulations .....                                | 14 |
| Cladding Oxidation.....                                | 18 |
| Fuel and Cladding Initial Temperature .....            | 18 |
| Cladding Failure Rate .....                            | 24 |
| Radiolysis Kinetics .....                              | 24 |
| Water Amount.....                                      | 25 |
| Thermal Decay Constant.....                            | 25 |
| Mode of Oxygen Diffusion.....                          | 26 |
| Fuel Oxidation .....                                   | 27 |
| Fuel and Cladding Initial Temperature .....            | 27 |
| Cladding Failure Rate .....                            | 29 |
| Radiolysis Kinetics .....                              | 30 |
| Water Amount.....                                      | 32 |
| Thermal Decay Constant.....                            | 33 |
| Mode of Oxygen Diffusion.....                          | 33 |
| Residual Oxygen.....                                   | 35 |
| Flammability Evaluation.....                           | 36 |
| Summary on Fuel and Cladding Oxidation .....           | 40 |
| Hydrogen Absorption by Cladding.....                   | 40 |
| Corrosion Calculations.....                            | 46 |
| Radiolysis Calculations.....                           | 51 |

INITIAL ENTRIES: Pavan K. Shukla  
Date: October 14, 2011

Title: Development of a Integrated Model to Analyze Water Consumption in a Dry Storage Cask System

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Project: This notebook documents development of a integrated model to analyze cladding and fuel oxidation due to water left in a dry storage cask system.

Initial Entry: This report presents analyses and supporting information to assess the potential impacts of residual water left inside a canister during fuel loading and subsequent drying. Typically, a dry storage cask system consists of a canister and overpack. However, some storage cask systems consist of a canister only. The canister is used to store spent nuclear fuel assemblies that have been cooled inside a spent fuel pool. The residual water could impact integrity of cladding, fuel, and other structural components of the canister's internals such as fuel basket, neutron absorbing materials, and the canister material. Analyses and supporting information have also been presented to assess the impact of residual water on criticality, canister pressurization, and flammability of the canister's environment.

The amount of residual water is assumed to be range from 1 to 55 moles. Even though there is an uncertainty regarding the amount of residual water, the analyses conducted using this range are expected to provide insight on potential impact of the residual water on the canister's internals. It is assumed that the canister is perfectly sealed and no ambient air ingresses. The residual water is expected to vaporize and undergo radiolytic decomposition because of heat load and radiation field of the spent nuclear fuel.

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## Model Development

The objective of quantitative analysis is to estimate the extent of cladding and fuel oxidation that could occur due to residual water in the canister. The amount of residual water will decrease with time primarily due to radiolysis. As outlined in Chapter 3, the rate of radiolysis would depend upon the fuel burn up and radiation field which is expected to have both temporal and spatial variations. The radiolysis product of water would include hydrogen, oxygen, hydrogen peroxide, and other species. For the sake of simplicity, it is assumed that the radiolysis of the water results in production of hydrogen and oxygen. It is recognized that hydrogen peroxide is also a dominant species that could be produced due to radiolysis of water. However, literature information indicates that hydrogen peroxide would rapidly oxidize  $\text{UO}_2$  even in excess of hydrogen (Lewis, et. al, 2002). Based upon this, it is assumed that hydrogen peroxide reacts with  $\text{UO}_2$  similar to reaction between oxygen and  $\text{UO}_2$ . Parallel information regarding reaction between cladding and hydrogen peroxide could not be found in literature. Nonetheless, it is also assumed that hydrogen peroxide reacts with zirconium-based cladding similar to reaction between oxygen and the cladding material.

The water could also directly react with cladding, exposed fuel, and other cask internal materials provided sufficient conditions exist for such reaction. For example, iron reacts with water in humid air when relative humidity is equal or greater than 60 percent (Vernon, 1935). Similarly, cladding, fuel, and cask internal materials could directly react with water when relative humidity is above a threshold value. A threshold value for water reaction with cladding is not available, for this reason, it is assumed that cladding directly reacts with water when relative humidity is greater than 20 percent.

The fuel and cladding oxidation models, presented in previous chapter, are implemented using a computer code developed in MATLAB<sup>®</sup>. This is referred as integration model. This model accounts for both temporal and spatial variation of temperature and relative humidity and their effects of cladding and fuel oxidation. The model is simulated for various cases to assess the uncertainties in water content, rate of radiolysis, and the thermal characteristics of cask system (i.e., temperature) on the fuel and cladding oxidation.

## Integration Model

The model consists of inputs, outputs, and calculations. A description of model inputs, calculation method, and model outputs are provided in the next three subsections. The model assumptions, wherever applicable, are also mentioned.

## Inputs

The model inputs include cask parameters, fuel temperature at the time of loading, residual water amount, cask internal volume, number of fuel assemblies, fuel rods per fuel assembly, dimensions of each fuel rod and fuel pellet, number of fragments per pellets, size of each grain in a fuel pellet, density of various  $\text{UO}_{2+x}$  phases, and void fraction in each fuel pellet. Since the fuel and cladding temperature is expected to vary spatially, the canister inside the cask volume is divided in the five zones. The inputs also include the radiolysis rate of water, and storage time of 300 years. In each zone, it is assumed that the fuel and cladding temperatures are uniform. Moreover, it is also assumed that the fuel temperature decays in each zone with same

rate. Initial fuel and cladding temperature and volume fraction of each zone is input in the model. A fraction of failed cladding percentage is also input. This fraction is used to calculate exposed fuel pellets available for oxidation. For example, a failed cladding fraction of 0.1 percent amount to failure of 1 fuel rod out of 1,000. The failed rod is assumed to have a crack of certain length. The crack length is specified in the model.

## Calculation Sequences

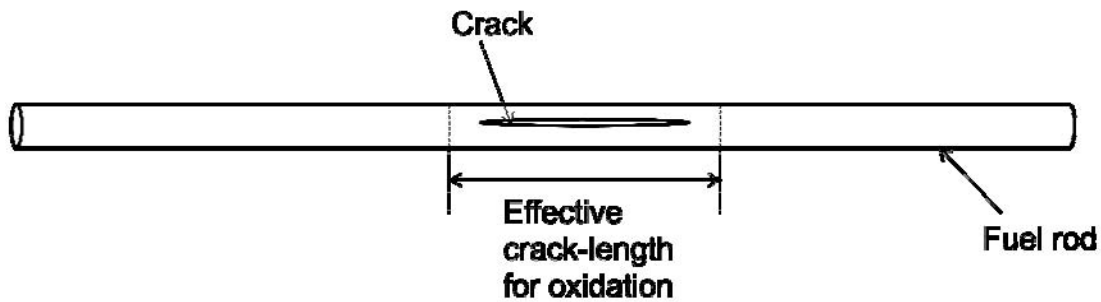
In the initial step, storage time is divided into several time steps. Before conducting sequential calculations as a function of time, the following calculations are conducted.

- Cladding surface areas in each zone are estimated based upon volume fraction in each zone. Cladding surface areas are estimated by multiplying the zone volume fraction with the total surface area of the cladding. The number of fuel rods in each zone is also estimated by multiplying the zone volume fraction with total rods in a canister.
- Number of failed fuel rods in each zone is estimated by multiplying number of fuel rods in each zone with the failure rate which is same as the failure rate of the cladding. The model inputs include cladding failure rate of 0.1 and 0.01 percent, and 4368 fuel rods. The cladding failure rate of 0.1 and 0.01 percent yield 4 and 1 failed rods, respectively. It is assumed that there is one failed rod in each of the hotter zone (i.e., zone 1-4) for the case of 0.1 percent failure rate. Similarly, for the 0.01 percent failure rate, the failed rod is assumed to be located in zone 1.
- The number of affected fuel pellets exposed to the canister environment due to failed cladding in a zone is estimated. Literature information (Einziger and Cook, 1985) indicates that the affected fuel pellets due to a crack of specified length in a fuel rod include pellets and 3 cm on either side along the axial length of the crack. Thus, effective crack-length for fuel oxidation is equal to dimension of the crack along the length of fuel rod plus 6 cm. A schematic diagram depicting a crack oriented axially along fuel rod in presented in Figure 1. As seen in the figure, the effective crack-length for fuel oxidation exceeds the crack length. The number of affected fuel pellets in a zone is calculated using the following equation

$$N_{\text{pellets\_zone}} = \text{floor} \left( \frac{\text{effective crack length}}{\text{pellet length}} \times \text{number of failed fuel rods in a zone} \right) \quad (1)$$

where  $N_{\text{pellets\_zone}}$  denotes the number of affected pellets. The *floor* function in Eq. (1) rounds the calculated number within parenthesis to lowest integer.

- Literature information (Einziger and Cook, 1985) also indicate fuel pellets directly underneath a crack tends to oxidize more compared to other fuel pellets not directly exposed but underneath the effective crack-length for oxidation. Considering this, it is assumed that the fuel pellets directly underneath the crack undergo oxidation before the other fuel pellets. Moreover, it is also assumed that the oxidation of the other pellets



**Figure 1. Schematic Representation of a Crack Oriented Along the Length of the Fuel Rod. The Effective Crack-Length for Fuel Oxidation is Equal to Crack Length Plus 6. cm.**

begin only after the directly exposed fuel pellets have completely oxidized. The number of directly-exposed fuel pellets in a zone are determined by the following equation

$$N_{\text{directly-exposed-pellets-zone}} = \text{floor} \left( \frac{\text{crack length}}{\text{pellet length}} \times \text{number of failed fuel rods in a zone} \right) \quad 2$$

where  $N_{\text{directly-exposed-pellets-zone}}$  denotes the number of directly-exposed fuel pellets in a zone. The number of other fuel pellets in a zone is calculated by subtracting number of directly-exposed fuel pellets from number of affected fuel pellets.

- The exposed area for fuel oxidation is also estimated. A fuel pellet is expected to fragment into 10-30 pieces during the reactor operation. There are two possible mechanisms for oxygen contacting the exposed fuel: (i) oxygen diffuses through grain boundaries and thus each fuel grain oxidizes simultaneously and (ii) through the surface of each fuel fragment. For the light water reactor fuel, the first case is referred as the base case whereas the second case is considered for uncertainties associated with the fuel oxidation model described in the following Table.

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| Table. Criteria for Temperature and Relative Humidity for Fuel Oxidation and Hydration in the Canister Environment  |           |   |  |
|---|-----------|---|--|
| Temperature and Relative Humidity   |           | Primary Phase Considered                      | Applicable Kinetic Equation and Comments   |
| T ≥ 230 °C [T ≥ 446 °F]<br>(independent of RH)  |           | U <sub>3</sub> O <sub>8</sub>                 | The equation for UO <sub>2.4</sub> , $w = (2kt)^{0.5}$ [Eq.(A3-4)] is used assuming all UO <sub>2.4</sub> is fully convert to U <sub>3</sub> O <sub>8</sub> .  |
| 150 ≤ T < 230 °C<br>[302 ≤ T < 446 °F]  | RH* < 40% | UO <sub>2.4</sub> †                           | $w = (2kt)^{0.5}$  |
|   | RH > 40%  | U <sub>3</sub> O <sub>8</sub>                 | U <sub>3</sub> O <sub>8</sub> is not normally observed below 230 °C [446 °F] in dry air with RH < 40%. U <sub>3</sub> O <sub>8</sub> can eventually form on a long-term at high relative humidity (no clear time at which formation starts) based on observations of U <sub>3</sub> O <sub>8</sub> formation for unirradiated UO <sub>2</sub> and used CANDU fuel tested at 150 °C ≤ T < 230 °C [302 °F ≤ T < 446 °F] with a high moisture level. The equation $w = (2kt)^{0.5}$ is applicable assuming conversion of UO <sub>2</sub> to U <sub>3</sub> O <sub>8</sub> . |
| T < 150 °C<br>[T < 302 °F]  | RH < 40%  | UO <sub>2.4</sub> ‡                           | $w = (2kt)^{0.5}$  |
|   | RH > 40%  | UO <sub>3</sub> •xH <sub>2</sub> O<br>(x < 2) | The kinetics to form schoepite or other hydrate forms can be used as the dissolution rate obtained from the aqueous condition.† The rate ranges from 0.01 to 6.85 mg/m <sup>2</sup> /day.  |
| <p>*relative humidity<br/>         †UO<sub>2.4</sub> could be a quasi-stable intermediate phase that can be eventually converted to U<sub>3</sub>O<sub>8</sub> only when the oxygen is available to react with UO<sub>2.4</sub> to form U<sub>3</sub>O<sub>8</sub> during a long-term conversion time {e.g., 1.16 × 10<sup>4</sup> years required at 150 °C [305 °F]} (see Table A3-2)<br/>         ‡NRC. NUREG-1914, "Dissolution Kinetics of Commercial Spent Nuclear Fuels in the Potential Yucca Mountain Repository Environment." ML083120074. Washington, DC: U.S. Nuclear Regulatory Commission. 2008. In NUREG-1914, the kinetics assume sufficient oxidants are present. Otherwise, the supply of oxidants can control the kinetics.</p> |           |   |  |

- For the first mechanism, i.e., base case, surface area for fuel oxidation per pellet is calculated by determining the number of grains per pellet and then multiplying it with the surface area of each grain. The number of grains per pellets is calculated by the following equation

$$N_{grains\_per\_pellet} = floor \left[ \frac{V_{pellet} \times (1 - f_{void})}{V_{grain}} \right] \quad (3)$$

where

|                           |   |                                  |
|---------------------------|---|----------------------------------|
| $N_{grains\_per\_pellet}$ | — | number of grains per pellet      |
| $V_{pellet}$              | — | volume of a pellet               |
| $V_{grain}$               | — | volume of a grain                |
| $f_{void}$                | — | void volume fraction in a pellet |

The corresponding surface area per pellet for fuel oxidation for the first mechanism is calculated by the following equation

$$A_{oxidation\_per\_pellet\_fm} = N_{grains\_per\_pellet} \times A_{grain} \quad (4)$$

where

- $A_{oxidation\_per\_pellet\_fm}$  — surface area for fuel oxidation per pellet for the first mechanism
- $A_{grain}$  — surface area of each grain

For the second mechanism of oxygen contacting each fuel fragment, surface area for fuel oxidation per pellet is determined by the following equation

$$A_{oxidation\_per\_pellet\_sm} = N_{fragments\_per\_pellet} \times A_{fragment} \quad (5)$$

where

- $A_{oxidation\_per\_pellet\_sm}$  — surface area for fuel oxidation per pellet for the second mechanism
- $A_{fragment}$  — surface area of a fragment

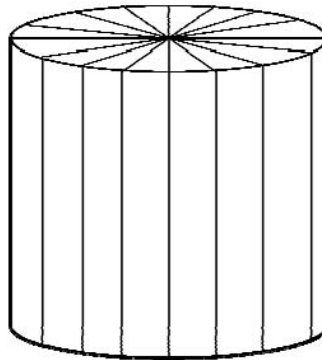
Each cylindrical fuel pellet is assumed to be fragmented into  $N_{fragments\_per\_pellet}$ . A schematic diagram depicting a fragmented fuel pellet is presented in Figure 2. As seen in the figure, the pellet fragments along the azimuthal direction.

- The temperature of each zone is calculated as a function of time. It is implicitly assumed that the fuel and cladding temperatures are not affected by the amount of residual water. The temperature of the fuel and cladding is assumed to vary with time according to the following equation.

$$T_{fuel\_cladding\_zone} = (T_{mean} - T_{ambient})exp(-at) + T_{ambient} \quad (6)$$

where

- $T_{fuel\_cladding\_zone}$  — temperature of fuel and cladding in a zone
- $T_{mean}$  — mean value of initial temperature in a zone at the time of fuel loading
- $T_{ambient}$  — ambient temperature, 309 K
- $a$  — thermal decay constant



**Figure 2. Schematic Representation of a Cylindrical Fuel Pellet Fragmented Into 16 Pieces**



- Before marching in time, the relative humidity in each zone is also calculated at the time of loading and first time step. It is assumed that the water is distributed in each zone according to the following equation

$$n_{water\_zone} = n_{water} \times f_{zone} \quad (7)$$

where

|                   |   |                                |
|-------------------|---|--------------------------------|
| $n_{water\_zone}$ | — | moles of water in a zone       |
| $n_{water}$       | — | moles of water in the canister |
| $f_{zone}$        | — | volume fraction of a zone      |

The partial pressure of water in each zone is calculated using the ideal gas law, and relative humidity is calculated by dividing the partial pressure with the saturated pressures. Additional details regarding the relative humidity calculations are provided in Chapter 2.

- The calculations are marched in time. The following calculations are conducted at each time step.
  - (1) The amount of oxygen produced due to radiolysis between the two time steps is calculated. The fuel and cladding temperature between the two time steps in a zone is assumed to be equal to average of the temperature at the two time steps. Similarly, relative humidity in a zone between the two time steps in a zone is assigned to be equal to the average of the relative humidity at the two time steps.
  - (2) The amount of oxygen consumed by the fuel pellets and cladding between the two time steps using the rate models in each zone is calculated. If the total oxygen produced by radiolysis is more than oxygen consumed by fuel and cladding oxidation in all the zones, the calculations are continued to the next time step. However, if the total oxygen consumed by fuel and cladding oxidation in all the zones is more than the oxygen generated by the radiolysis, the radiolysis-controlled fuel and cladding oxidation model is applied. This is explained next.

In the radiolysis-controlled oxidation model, amount of oxygen consumed by fuel and cladding oxidation in zone 5, the coldest zone, is estimated. If the moles of oxygen generated due to radiolysis in the zone 5 is less than amount needed for fuel and cladding oxidation, the amount of oxygen generated between the two time steps is divided between the fuel and cladding according to the following equations

$$n_{oxygen\_fuel\_oxidation\_zone\_5} = \left( \frac{A_{exposed\_fuel\_zone\_5}}{A_{exposed\_fuel\_zone\_5} + A_{clad\_zone\_5}} \right) \times n_{oxygen\_generated\_zone\_5} \quad (8)$$

$$n_{oxygen\_clad\_oxidation\_zone\_5} = \left( \frac{A_{clad\_zone\_5}}{A_{exposed\_fuel\_zone\_5} + A_{clad\_zone\_5}} \right) \times n_{oxygen\_generated\_zone\_5} \quad (9)$$

where

|  |   |  |
|--|---|--|
| $n_{oxygen\_fuel\_oxidation\_zone\_5}$ | — | moles of oxygen consumed by fuel oxidation in zone 5       |
| $n_{oxygen\_clad\_oxidation\_zone\_5}$ | — | moles of oxygen consumed by cladding oxidation in zone 5   |
| $n_{oxygen\_generated\_zone\_5}$       | — | moles of oxygen generated between two time steps in zone 5 |
| $A_{exposed\_fuel\_zone\_5}$           | — | surface area of the exposed fuel in zone 5                 |
| $A_{clad\_zone\_5}$                    | — | surface area of cladding in zone 5                         |

However, if the moles of oxygen generated due to radiolysis in the zone 5 is more than oxygen amount needed for fuel and cladding oxidation, the leftover oxygen moles in zone 5 is calculated according to the following equation

$$\begin{aligned}
n_{oxygen\_leftover\_zone\_5} &= n_{oxygen\_generated\_zone\_5} - n_{oxygen\_fuel\_oxidation\_zone\_5} \\
&\quad - n_{oxygen\_clad\_oxidation\_zone\_5}
\end{aligned} \tag{10}$$

where  $n_{oxygen\_leftover\_zone\_5}$  denotes moles of oxygen not consumed by cladding and fuel oxidation in zone 5. These moles of oxygen are added to the moles of oxygen generated in zone 4. Thus, the effective moles of oxygen present in zone four between the two time steps are equal to

$$n_{oxygen\_present\_zone\_4} = n_{oxygen\_generated\_zone\_4} + n_{oxygen\_leftover\_zone\_5} \tag{11}$$

where

|                                  |   |  |
|----------------------------------|---|--|
| $n_{oxygen\_present\_zone\_4}$   | — | moles of oxygen present in zone 4 for fuel and cladding oxidation        |
| $n_{oxygen\_generated\_zone\_4}$ | — | moles of oxygen generated by radiolysis in zone 4 between two time steps |

Again, the moles of oxygen consumed by fuel and cladding oxidation in zone 4 are calculated. If the moles of oxygen present in the zone 4 are less than amount needed for fuel and cladding oxidation, the moles of oxygen partitioned between the cladding and fuel according to the following equations

$$\begin{aligned}
n_{oxygen\_fuel\_oxidation\_zone\_4} &= \left( \frac{A_{exposed\_fuel\_zone\_4}}{A_{exposed\_fuel\_zone\_4} + A_{clad\_zone\_4}} \right) \times n_{oxygen\_present\_zone\_4}
\end{aligned} \tag{12}$$

$$\begin{aligned}
n_{oxygen\_clad\_oxidation\_zone\_4} &= \left( \frac{A_{clad\_zone\_4}}{A_{exposed\_fuel\_zone\_4} + A_{clad\_zone\_4}} \right) \times n_{oxygen\_present\_zone\_4}
\end{aligned} \tag{13}$$

where

|  |   |  |
|--|---|--|
| $n_{oxygen\_fuel\_oxidation\_zone\_4}$ | — | moles of oxygen consumed by fuel oxidation in zone 4     |
| $n_{oxygen\_clad\_oxidation\_zone\_4}$ | — | moles of oxygen consumed by cladding oxidation in zone 4 |
| $A_{exposed\_fuel\_zone\_4}$           | — | surface area of the exposed fuel in zone 4               |

$A_{clad\_zone\_4}$

— surface area of cladding in zone 4

Otherwise, the excess moles of oxygen are transferred to zone 3. This process is repeated for the remaining three zones.

- (3) A counter is used to keep track of all of the oxygen moles produced by radiolysis and consumed by cladding and fuel oxidation throughout the preceding time steps. If the moles of oxygen produced by radiolysis reach a plateau, and the difference between moles of oxygen produced by radiolysis and the total moles of oxygen consumed by fuel and cladding oxidation is less than a specified tolerance limit, calculations are stopped.
- (4) When the criterion to stop the calculations is not met, it is checked whether the directly-exposed fuel pellets in a zone have completely oxidized or not. If the directly-exposed fuel pellets have completely oxidized in a zone, the other fuel pellets undergo oxidation in the next time step. It is also checked whether the affected fuel pellets (i.e., the directly-exposed and other fuel pellets) have completely oxidized in a zone or not. If the affected fuel pellets have completely oxidized, only cladding oxidation is implemented in the next time step.
- (5) The calculation steps 1–4 are repeated till the criterion to stop the calculations is met.

## Outputs

The integrated model calculates extent of fuel and cladding oxidation, and moles of oxygen consumed by both fuel and cladding in each zone. The model also calculates fuel and cladding temperature, relative humidity, moles of oxygen, hydrogen, and water in the canister as a function of time.

## Model Parameters

The values of different parameters used in the model are provided. Some of parameters are fixed while others are varied in the model. The parameters whose values are fixed in the model are listed in Table 1.

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| <b>Table 1. Cladding, Fuel, and Canister Parameter Values Used in the Integration Model</b> |   |
|---|---|
| <b>Parameter</b>  | <b>Values</b>                                 |
| Cask volume   | 2,100 L                                       |
| Number of fuel assemblies   | 21  |
| Fuel rods per assembly  | 208 in 15 x 15 Babcock & Wilcox Fuel Assembly |
| Fuel rod length   | 3.9 m   |
| Fuel rod outer diameter   | 10.92 mm                                      |
| Pellet diameter   | 9.36 mm                                       |
| Pellet length   | 15.24 mm                                      |
| Axial length of a crack on a failed rod   | 3.5 cm  |
| Fuel pellet void volume   | 5%  |
| UO <sub>2</sub> density   | 10.96 g/cm <sup>3</sup>                       |
| UO <sub>2.4</sub> density   | 11.30 g/cm <sup>3</sup>                       |
| U <sub>3</sub> O <sub>8</sub> density   | 8.35 g/cm <sup>3</sup>                        |
| UO <sub>3</sub> ·xH <sub>2</sub> O* (x <2) density  | 4.89 g/cm <sup>3</sup>                        |
| UO <sub>2</sub> grain shape   | spherical                                     |
| UO <sub>2</sub> grain radius  | 10 μm   |
| Volume fraction zones 1, 2, 3, 4, and 5   | 19, 33, 33.7, 12.4, and 1.9%, respectively    |
| ZrO <sub>2</sub> density  | 5.6 g/cm <sup>3</sup>                         |
| *In the integration model simulation, x is assumed to be zero.                              |   |

The model results are presented for several potential scenarios which account for uncertainties in the amount of residual water, radiolysis kinetics, decay heat of the fuel loaded in the canister, rate of temperature decay of fuel, and mechanisms for oxygen contacting the exposed fuel. These uncertainties and the inputs used to model these uncertainties are described next.

## Residual Water Amount

The residual water amount could vary between 1 to 55 moles. Specific values of 5.5, 17.4, and 55 moles of the residual water are used in the model. The potential effects of the residual water amount less than 5.5 moles are also discussed in the next section.

## Radiolysis Kinetics

The residual water could completely decompose between 4.77 to 71.62 years. The rate of radiolysis would determine the rate of production of oxidizing species such as oxygen and hydrogen peroxide. Moreover, since the oxidizing species are consumed during fuel and cladding oxidation, the rate of radiolysis is affected by the fuel and cladding oxidation. Considering this, the rate of radiolysis as discussed in Section 3.2 is input using a linear and kinetic rate. For the linear rate, the water, oxygen, and hydrogen concentrations are calculated using the following equations

$$n_{water}(t) = n_{water}(1 - bt) \quad (14)$$

$$n_{oxygen}(t) = \frac{n_{water} - n_{water}(t)}{2} \quad (15)$$

$$n_{hydrogen}(t) = n_{water} - n_{water}(t) \quad (16)$$

where

|                   |   |                               |
|-------------------|---|-------------------------------|
| $n_{water}(t)$    | — | moles of water at time $t$    |
| $n_{water}$       | — | moles of the residual water   |
| $b$               | — | constant                      |
| $t$               | — | time in years                 |
| $n_{oxygen}(t)$   | — | moles of oxygen at time $t$   |
| $n_{hydrogen}(t)$ | — | moles of hydrogen at time $t$ |

The values of  $b$  are  $1/4.77$  and  $1/71.62$  for the decomposition periods of 4.77 to 71.62 years, respectively. The values of  $b$  is selected such that complete radiolytic decomposition of the residual water occurs either in 4.77 years or 70 years, respectively. For the exponential kinetic rate, the amount of water is calculated using the following equation

$$n_{water}(t) = n_{water} \exp(-ct) \quad (17)$$

where values of  $c$  is a constant whose value are input as -1.9290 and -0.12860 for complete radiolytic decomposition of the residual water in 4.77 years and 71.62 years, respectively. Equations (15) and (16) are used to calculate oxygen and hydrogen moles.

## Fuel and Cladding Temperature

The fuel temperature could significantly affect the fuel and cladding oxidation in the dry storage cask system. This uncertainty is considered in the model by inputting two sets of fuel and cladding initial temperatures in different zones. These values are assumed to be fuel and cladding temperatures at the time of canister loading, and referred as low and high-end fuel and cladding initial temperatures. These values are listed in Table 2 for the sake of convenience.

## Decay Rate of Fuel and Cladding Temperature

Equation (6) is used to model the evolution of fuel and cladding temperature. The thermal characteristics of a storage cask system would affect the evolution of fuel and cladding temperature. To account for this uncertainty, two values of parameter  $a$ , also referred as the thermal decay constant, is input in the model. The value of  $a$  in Eq. (6) is set equal to either 0.023 or 0.064. The fuel and cladding temperature decreases slower with time for  $a$  equal to 0.023 than 0.064.

## Cladding Failure Rate

The cladding failure rate of 0.1 and 0.01 percent is used in the model. These values are based upon the literature information.

## Mechanisms for Oxygen Contacting the Exposed Fuel

The model used one of the following mechanisms for oxygen contacting the exposed fuel (i) oxygen diffuses through grain boundaries (also referred as the base case) and thus each fuel grain oxidizes simultaneously and (ii) through the surface of each fuel fragment. This uncertainty is input in the model through surface area of the exposed fuel available for oxidation.

| <b>Zone</b> | <b>Mean Values of Low-End Fuel and Cladding Initial Temperature (K) [°F]</b> | <b>Mean Values of High-End Fuel and Cladding Initial Temperature (K) [°F]</b> |
|-------------|--|---|
| 1           | 575 [575.3]  | 673 [751.7]   |
| 2           | 525 [485.3]  | 623 [661.7]   |
| 3           | 475 [395.3]  | 573 [571.7]   |
| 4           | 425 [305.3]  | 523 [481.7]   |
| 5           | 375 [215.3]  | 481 [406.1]   |

### Model Simulations

The model is simulated for various combinations of parameters listed in Table 3. Several cases are constructed by combination of these parameters. The cases are listed in Table in Table 4. These cases correspond to various uncertainties discussed above.

The only difference between the first four cases is the selection of the radiolysis model. Case 1 to 4 can be seen as combination of several sub cases because several varying parameters are included in each case.

| <b>Parameter</b>                      | <b>Value</b>  |
|---------------------------------------|---|
| Fuel and cladding initial temperature | Low- or high-end fuel and cladding initial temperature (as listed in Table 2)   |
| Cladding failure rate                 | 0.1 or 0.01%  |
| Radiolysis kinetics                   | Exponential decomposition in 4.77 years, or linear decomposition in 4.77 years, or exponential decomposition in 71.62 years, or linear decomposition in 71.62 years |
| Residual water amount                 | 5.5 or 17.4 or 55 moles   |
| Thermal decay constant                | $\alpha$ equal to 0.023 or 0.064  |
| Mode of oxygen contacting the fuel    | Either through grains boundaries or through fragment surface  |

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| Table 4. Values of Varying Parameters and Corresponding Cases for Presenting the Simulation Data of the Integrated Model |   |
|--|---|
| Case No.   | Values of Varying Parameters Listed in Table  |
| 1  | The residual water amounts of 5.5, 17.4, and 55 moles. Cladding failure rate of 0.1 and 0.01. Low- and high-end initial fuel temperature. Oxygen diffusion through grain boundaries and contacting the each grain simultaneously (i.e., the base case). Thermal decay constant $\alpha$ equal to 0.023. Exponential decomposition of the residual water in 4.77 years due to radiolysis.                          |
| 2  | All parameters are same as case 1 except linear decomposition of the residual water in 4.77 years due to radiolysis.  |
| 3  | All parameters are same as case 1 except exponential decomposition of the residual water in 71.62 years due to radiolysis.  |
| 4  | All parameters are same as case 1 except linear decomposition of the residual water in 71.62 years due to radiolysis.   |
| 5  | 17.4 moles of residual water. Cladding failure rate of 0.1 percent. Low-end initial fuel temperature. Oxygen diffusion through grain boundaries (i.e., the base case). Linear decomposition of the residual water in 71.62 years due to radiolysis. Thermal decay constant $\alpha$ equal to 0.064. The case 4 data is used for comparison.   |
| 6  | Water amount of 17.4 moles. Cladding failure rate of 0.1 percent. Low-end initial fuel and cladding temperature. Exponential decomposition of the residual water in 4.77 years due to radiolysis. Thermal decay constant $\alpha$ equal to 0.023. Oxygen diffusion through fuel pellet fragments' surface. A fuel pellet is considered to have fragmented into 10 pieces. The case 1 data is used for comparison. |

Case 5 is constructed to highlight the effect of thermal decay constant on cladding and fuel oxidation. In this case, linear decay of water in 71.62 years is selected because the decay constant is expected to maximally influence fuel and cladding oxidation when water decomposes over longer period. This is expected because fuel and cladding temperature is expected to decay more rapidly when  $\alpha$  equal to 0.064, and thus, expected to influence fuel and cladding oxidation. The case 4 data is used for comparison, and highlight the affect of the thermal decay constant on cladding and fuel oxidation.

Case 6 is constructed to estimate the effect of mode of oxygen contacting the fuel. The case 1 data is used for comparison.

## Model Results

The cladding failure would cause a fuel rod to develop a crack in it. The cladding failure rate and length of the crack per failed rod parameter values are used to determine number of exposed fuel pellets in each zone. The calculated number of failed rods and corresponding exposed fuel pellets are summarized in Table 5. As long as cladding failure rates are specified in a case, the number of failed rods and corresponding number of exposed pellets remain unchanged.

Similarly, the surface areas of the exposed cladding in each zone also remain unchanged through a simulation. The surface area of cladding surface in each zone is directly proportional of volume fraction of each zone. The exposed cladding surface area was calculated by

multiplying the total surface area with the volume fraction of each zone. The exposed surface area of the cladding in each zone is provided in Table 6.

| Zone | Cladding Failure Rate of 0.1% |   |  | Cladding Failure Rate of 0.01% |   |  |
|------|-------------------------------|---|--|--------------------------------|---|--|
|      | Failed Rods                   | Number of Exposed Pellets Directly Underneath the 3.5 cm-Long Crack | Number of Pellets Underneath the Extended Length of the Crack* | Failed Rods                    | Number of Exposed Pellets Directly Underneath the 3.5 cm-Long Crack | Number of Pellets Underneath the Extended Length of the Crack* |
| 1    | 1                             | 2   | 4  | 1                              | 2   | 4  |
| 2    | 1                             | 2   | 4  | 0                              | 0   | 0  |
| 3    | 1                             | 2   | 4  | 0                              | 0   | 0  |
| 4    | 1                             | 2   | 4  | 0                              | 0   | 0  |
| 5    | 0                             | 0   | 0  | 0                              | 0   | 0  |

\*The fuel pellets underneath the extended length are not directly exposed.

| Zone                | 1<br>(m <sup>2</sup> ) [ft <sup>2</sup> ] | 2<br>(m <sup>2</sup> ) [ft <sup>2</sup> ] | 3<br>(m <sup>2</sup> ) [ft <sup>2</sup> ] | 4<br>(m <sup>2</sup> ) [ft <sup>2</sup> ] | 5<br>(m <sup>2</sup> ) [ft <sup>2</sup> ] |
|---------------------|---|---|---|---|---|
| <b>Surface Area</b> | 111<br>[1194.8]                           | 192.9<br>[2076.4]                         | 196.9<br>[2119.4]                         | 72.5<br>[780.4]                           | 11.1<br>[119.5]                           |

The simulation data for fuel and cladding oxidation for the six cases are presented in various tables. The cladding oxidation data are presented in terms of thickness of zirconium oxide layer. The fuel oxidation data are presented in terms of amount of UO<sub>2+x</sub> phase that would form due to exposure to oxygen produced by radiolysis. The fuel oxidation data also include the extent of oxidation which is equal to percentage of the exposed fuel pellets that have undergone oxidation to particular UO<sub>2+x</sub> phase. The extent of oxidation is separately calculated for the directly exposed fuel pellets, and fuel pellets in the extended crack length but not directly exposed.

The cladding oxidation data for the case 1 to 4 are listed in Tables 7 to 10, respectively. Note that the cladding oxidation model by Daalgard (1976) was used in the simulations. Analysis presented in Chapter 4 indicate that the cladding oxidation model by Daalgard (1976) are expected to provide highest cladding oxidation rates compared to the other models. The selection of the cladding oxidation model by Daalgard (1976) is based upon the following two considerations: i) this is expected to provide a bounding assessment of the cladding oxidation in the limited oxidizing environment in the canister, ii) this is also expected to provide a bounding assessment on fuel oxidation in the canister. Regarding the second consideration, when cladding consumes most of the available oxygen, the oxidation of the any exposed fuel is expected to be bounding. The cladding oxidation data for case 5 and 6 are provided in the next subsection where effects of various varying parameters on cladding oxidation are discussed.

The fuel oxidation data for the case 1 to 4 are listed in Tables 11 to 14, respectively. The data from these tables is selected to highlight the effects of various varying parameters. The case 5 and 6 data is presented when fuel oxidation data for case 1-4 are discussed.



| Table 7. Cladding Oxidation Data for Case 1<br>(Exponential Decomposition of the Residual Water in 4.77 Years) |   |                        |                        |   |                        |                        |                             |
|--|---|------------------------|------------------------|---|------------------------|------------------------|-----------------------------|
| Zone   | Low-End Fuel and<br>Cladding Initial Temperature                                  |                        |                        | High-End Fuel and<br>Cladding Initial Temperature                                 |                        |                        | Cladding<br>Failure<br>Rate |
|  | Cladding Oxides Layer Thickness ( $\mu\text{m}$ )<br>[mil] for Moles of the Water |                        |                        | Cladding Oxides Layer Thickness ( $\mu\text{m}$ )<br>[mil] for Moles of the Water |                        |                        |                             |
|  | 5.5   | 17.4                   | 55                     | 5.5   | 17.4                   | 55                     |                             |
| 1  | 1.24<br>[4.87E-02]  | 4.80<br>[1.89E-01]     | 7.32<br>[2.88E-01]     | 3.84E-01<br>[1.51E-02]  | 1.22<br>[4.78E-02]     | 7.03<br>[2.77E-01]     | 0.1                         |
| 2  | 3.09E-01<br>[1.22E-02]  | 7.58E-01<br>[2.98E-02] | 1.19<br>[4.67E-02]     | 3.85E-01<br>[1.52E-02]  | 1.53<br>[6.02E-02]     | 5.36<br>[2.11E-01]     | 0.1                         |
| 3  | 5.12E-02<br>[2.01E-03]  | 8.23E-02<br>[3.24E-03] | 1.36E-01<br>[3.53E-03] | 4.18E-01<br>[1.65E-02]  | 1.23<br>[4.86E-02]     | 1.95<br>[7.66E-02]     | 0.1                         |
| 4  | 4.77E-03<br>[1.88E-04]  | 5.44E-03<br>[2.14E-04] | 1.00E-02<br>[3.95E-04] | 2.54E-01<br>[1.00E-02]  | 3.52E-01<br>[1.39E-02] | 4.41E-01<br>[1.74E-02] | 0.1                         |
| 5  | 1.89E-04<br>[7.45E-06]  | 2.03E-04<br>[8.00E-06] | 4.63E-04<br>[1.82E-05] | 6.21E-02<br>[2.45E-03]  | 7.53E-02<br>[2.96E-03] | 8.69E-02<br>[3.42E-03] | 0.1                         |
| 1  | 1.33<br>[5.22E-02]  | 4.88<br>[1.92E-01]     | 7.32<br>[2.88E-01]     | 3.84E-01<br>[1.51E-02]  | 1.22<br>[4.78E-02]     | 7.21<br>[2.84E-01]     | 0.01                        |
| 2  | 3.15E-01<br>[1.24E-02]  | 7.71E-01<br>[3.03E-02] | 1.19<br>[4.67E-02]     | 3.86E-01<br>[1.52E-02]  | 1.61<br>[6.32E-02]     | 5.36<br>[2.11E-01]     | 0.01                        |
| 3  | 5.15E-02<br>[2.03E-03]  | 8.37E-02<br>[3.29E-03] | 1.36E-01<br>[3.53E-03] | 4.55E-01<br>[1.79E-02]  | 1.23<br>[4.86E-02]     | 1.94<br>[7.66E-02]     | 0.01                        |
| 4  | 4.57E-03<br>[1.80E-04]  | 5.53E-03<br>[2.18E-04] | 1.00E-02<br>[3.95E-04] | 2.48E-01<br>[9.77E-02]  | 3.51E-01<br>[1.38E-02] | 4.41E-01<br>[1.74E-02] | 0.01                        |
| 5  | 1.89E-04<br>[7.45E-06]  | 2.03E-04<br>[8.00E-06] | 4.63E-04<br>[1.82E-05] | 6.21E-02<br>[2.45E-03]  | 7.53E-02<br>[2.96E-03] | 8.69E-02<br>[3.42E-03] | 0.01                        |

| Table 8. Cladding Oxidation Data for Case 2<br>(Linear Decomposition of the Residual Water in 4.77 Years) |   |                        |                        |   |                        |                        |                             |
|---|---|------------------------|------------------------|---|------------------------|------------------------|-----------------------------|
| Zone  | Low-End Fuel and<br>Cladding Initial Temperature                              |                        |                        | High-End Fuel and<br>Cladding Initial Temperature                             |                        |                        | Cladding<br>Failure<br>Rate |
|   | Cladding Oxides Layer Thickness ( $\mu\text{m}$ )<br>[mil] for Moles of Water |                        |                        | Cladding Oxides Layer Thickness ( $\mu\text{m}$ )<br>[mil] for Moles of Water |                        |                        |                             |
|   | 5.5   | 17.4                   | 55                     | 5.5   | 17.4                   | 55                     |                             |
| 1   | 6.29E-01<br>[2.48E-02]  | 3.98<br>[1.57E-01]     | 7.32<br>[2.88E-01]     | 3.84E-01<br>[1.51E-02]  | 1.22<br>[4.78E-02]     | 3.84<br>[1.51E-01]     | 0.1                         |
| 2   | 6.66E-01<br>[2.62E-02]  | 7.23E-01<br>[2.85E-02] | 1.19<br>[4.67E-02]     | 3.85E-01<br>[1.52E-02]  | 1.22<br>[4.78E-02]     | 5.04<br>[2.00E-01]     | 0.1                         |
| 3   | 7.76E-02<br>[3.06E-03]  | 7.82E-02<br>[3.08E-03] | 1.36E-01<br>[5.35E-03] | 3.85E-01<br>[1.52E-02]  | 1.44<br>[5.65E-02]     | 4.02<br>[1.59E-01]     | 0.1                         |
| 4   | 5.11E-03<br>[2.01E-04]  | 5.10E-03<br>[2.05E-04] | 1.00E-02<br>[3.95E-04] | 4.26E-01<br>[1.62E-02]  | 7.08E-01<br>[2.79E-02] | 6.65E-01<br>[2.60E-02] | 0.1                         |
| 5   | 1.64E-04<br>[6.48E-06]  | 1.66E-04<br>[6.54E-06] | 4.63E-04<br>[1.82E-05] | 1.04E-01<br>[4.08E-02]  | 1.04E-01<br>[4.08E-03] | 1.04E-01<br>[4.08E-03] | 0.1                         |
| 1   | 7.11E-01<br>[2.80E-02]  | 4.16<br>[1.64E-01]     | 7.32<br>[2.88E-01]     | 3.84E-01<br>[1.51E-02]  | 1.22<br>[4.78E-02]     | 3.84<br>[1.51E-01]     | 0.01                        |
| 2   | 6.78E-01<br>[2.67E-02]  | 7.35E-01<br>[2.89E-02] | 1.19<br>[4.67E-02]     | 3.86E-01<br>[1.52E-02]  | 1.22<br>[4.78E-02]     | 5.09<br>[2.00E-01]     | 0.01                        |
| 3   | 7.74E-02<br>[3.05E-03]  | 7.96E-02<br>[3.14E-03] | 1.36E-01<br>[5.35E-03] | 3.87E-01<br>[1.52E-02]  | 1.49<br>[5.87E-02]     | 4.05<br>[1.59E-01]     | 0.01                        |
| 4   | 5.06E-03<br>[1.99E-04]  | 5.21E-03<br>[2.05E-04] | 1.00E-02<br>[3.95E-04] | 4.27E-01<br>[1.68E-02]  | 6.60E-01<br>[2.60E-02] | 6.60E-01<br>[2.60E-02] | 0.01                        |
| 5   | 1.64E-04<br>[6.48E-06]  | 1.70E-04<br>[6.70E-06] | 4.63E-04<br>[1.82E-05] | 1.04E-01<br>[4.08E-03]  | 1.04E-01<br>[4.08E-03] | 1.04E-01<br>[4.08E-03] | 0.01                        |

| <b>Table 9. Cladding Oxidation Data for Case 3<br/>(Exponential Decomposition of the Residual Water in 71.62 Years)</b> |  |             |           |  |             |           |                                      |
|---|--|-------------|-----------|--|-------------|-----------|--------------------------------------|
| <b>Zone</b>   | <b>Low-End Fuel and<br/>Cladding Initial Temperature</b>                                       |             |           | <b>High-End Fuel and<br/>Cladding Initial Temperature</b>                                      |             |           | <b>Cladding<br/>Failure<br/>Rate</b> |
|   | <b>Cladding Oxides Layer Thickness (<math>\mu\text{m}</math>)<br/>[mil] for Moles of Water</b> |             |           | <b>Cladding Oxides Layer Thickness (<math>\mu\text{m}</math>)<br/>[mil] for Moles of Water</b> |             |           |                                      |
|   | <b>5.5</b>   | <b>17.4</b> | <b>55</b> | <b>5.5</b>   | <b>17.4</b> | <b>55</b> |                                      |
| 1   | 4.65E-01   | 3.54E+00    | 7.37E+00  | 3.84E-01   | 1.22E+00    | 3.95E+00  | 0.1                                  |
| 2   | 7.17E-01   | 1.19E+00    | 1.19E+00  | 3.85E-01   | 1.22E+00    | 4.27E+00  | 0.1                                  |
| 3   | 1.36E-01   | 1.36E-01    | 1.36E-01  | 3.85E-01   | 1.31E+00    | 4.51E+00  | 0.1                                  |
| 4   | 1.01E-02   | 1.00E-02    | 1.00E-02  | 4.13E-01   | 1.06E+00    | 1.11E+00  | 0.1                                  |
| 5   | 4.34E-04   | 4.39E-04    | 4.63E-04  | 1.80E-01   | 1.80E-01    | 1.80E-01  | 0.1                                  |
| 1   | 4.87E-01   | 3.65E+00    | 7.36E+00  | 3.84E-01   | 1.22E+00    | 3.98E+00  | 0.01                                 |
| 2   | 7.44E-01   | 1.19E+00    | 1.19E+00  | 3.86E-01   | 1.23E+00    | 4.31E+00  | 0.01                                 |
| 3   | 1.36E-01   | 1.36E-01    | 1.36E-01  | 3.88E-01   | 1.33E+00    | 4.55E+00  | 0.01                                 |
| 4   | 9.99E-03   | 9.99E-03    | 1.00E-02  | 4.13E-01   | 1.05E+00    | 1.10E+00  | 0.01                                 |
| 5   | 4.38E-04   | 4.39E-04    | 4.63E-04  | 1.80E-01   | 1.80E-01    | 1.80E-01  | 0.01                                 |

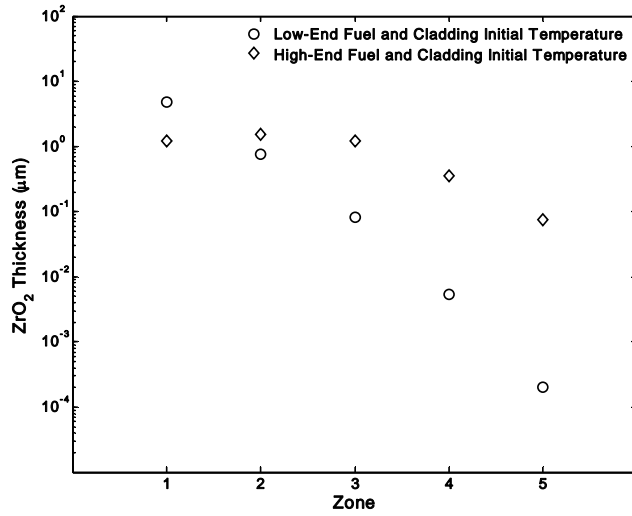
| <b>Table 10. Cladding Oxidation Data for Case 4<br/>(Linear Decomposition of the Residual Water in 71.62 Years)</b> |  |             |           |  |             |           |                                      |
|---|--|-------------|-----------|--|-------------|-----------|--------------------------------------|
| <b>Zone</b>   | <b>Low-End Fuel and<br/>Cladding Initial Temperature</b>                                       |             |           | <b>High-End Fuel and<br/>Cladding Initial Temperature</b>                                      |             |           | <b>Cladding<br/>Failure<br/>Rate</b> |
|   | <b>Cladding Oxides Layer Thickness (<math>\mu\text{m}</math>)<br/>for [mil] Moles of Water</b> |             |           | <b>Cladding Oxides Layer Thickness (<math>\mu\text{m}</math>)<br/>[mil] for Moles of Water</b> |             |           |                                      |
|   | <b>5.5</b>   | <b>17.4</b> | <b>55</b> | <b>5.5</b>   | <b>17.4</b> | <b>55</b> |                                      |
| 1   | 2.75E-01   | 7.70E-01    | 2.14E+00  | 3.07E-01   | 8.70E-01    | 2.43E+00  | 0.1                                  |
| 2   | 1.80E-01   | 4.57E-01    | 9.75E-01  | 2.46E-01   | 6.72E-01    | 1.79E+00  | 0.1                                  |
| 3   | 8.75E-02   | 1.36E-01    | 1.36E-01  | 1.93E-01   | 5.00E-01    | 1.24E+00  | 0.1                                  |
| 4   | 1.12E-02   | 1.03E-02    | 1.01E-02  | 1.40E-01   | 3.29E-01    | 6.98E-01  | 0.1                                  |
| 5   | 4.38E-04   | 4.38E-04    | 4.63E-04  | 8.51E-02   | 1.55E-01    | 1.80E-01  | 0.1                                  |
| 1   | 2.82E-01   | 7.90E-01    | 2.18E+00  | 3.11E-01   | 8.79E-01    | 2.46E+00  | 0.01                                 |
| 2   | 1.85E-01   | 4.77E-01    | 9.97E-01  | 2.49E-01   | 6.78E-01    | 1.81E+00  | 0.01                                 |
| 3   | 9.10E-02   | 1.36E-01    | 1.36E-01  | 1.95E-01   | 5.06E-01    | 1.26E+00  | 0.01                                 |
| 4   | 9.99E-03   | 9.99E-03    | 1.00E-02  | 1.40E-01   | 3.30E-01    | 6.99E-01  | 0.01                                 |
| 5   | 4.38E-04   | 4.38E-04    | 4.63E-04  | 8.51E-02   | 1.55E-01    | 1.80E-01  | 0.01                                 |

## Cladding Oxidation

This subsection provides analysis of cladding oxidation data to identify the effects of various varying parameters on the oxidation.

### Fuel and Cladding Initial Temperature

The cladding oxidation data from Case 1 is extracted to illustrate the effect of fuel and cladding initial temperature. The cladding oxidation data for 17.4 moles of the residual water, 0.1 percent cladding failure rate and low- and high-end fuel and cladding initial temperature is presented in the Figure 3.



**Figure 3. ZrO<sub>2</sub> Oxide Layer Thickness Formed During Cladding Oxidation for 17.4 Moles of Residual Water, 0.1 Percent Cladding Failure Rate, Exponential Decomposition of the Water in 4.77 Years, and Low- and High-End Fuel and Cladding Initial Temperature, and Thermal Decay Constant of 0.023**

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**Table 11. Fuel Oxidation Simulation Data for Case 1 (Exponential Decomposition of the Residual Water in 4.77 Years)**

| Zone | Low-End Fuel and Cladding Initial Temperature  |   |  |   |  |   | High-End Fuel and Cladding Initial Temperature |   |  |   |  |  | Cladding Failure Rate |
|------|--|---|--|---|--|---|--|---|--|---|--|--|-----------------------|
|      | Moles of Water                                 |   |  |   |  |   |  |   |  |   |  |  |                       |
|      | 5.5  |   | 17.4   |   | 55   |   | 5.5  |   | 17.4   |   | 55   |  |                       |
|      | UO <sub>2+x</sub> Mass [oz]                    | Extent of Oxidation*  | UO <sub>2+x</sub> Mass [oz]                    | Extent of Oxidation*  | UO <sub>2+x</sub> Mass [oz]                    | Extent of Oxidation*  | UO <sub>2+x</sub> Mass [oz]                    | Extent of Oxidation*  | UO <sub>2+x</sub> Mass [oz]                    | Extent of Oxidation*  | UO <sub>2+x</sub> Mass [oz]                    | Extent of Oxidation*   |                       |
| 1    | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 68.3 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 68.4 g [2.41] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 2.4 g [0.08] of U <sub>3</sub> O <sub>8</sub>  | 10.4% to U <sub>3</sub> O <sub>8</sub> , 0                                    | 7.5 g [0.26] of U <sub>3</sub> O <sub>8</sub>  | 32.8% to U <sub>3</sub> O <sub>8</sub> , 0                                    | 63.5 g [2.24] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 89.7% to U <sub>3</sub> O <sub>8</sub> | 0.1%                  |
| 2    | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 2.4 g [0.08] of U <sub>3</sub> O <sub>8</sub>  | 10.4% to U <sub>3</sub> O <sub>8</sub> , 0                                    | 9.4 g [0.33] of U <sub>3</sub> O <sub>8</sub>  | 41.4% to U <sub>3</sub> O <sub>8</sub> , 0                                    | 68.4 g [2.41] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub>  | 0.1%                  |
| 3    | 49.2 g [1.73] of UO <sub>2.4</sub>             | 100% to UO <sub>2.4</sub> , 60.0% to UO <sub>2.4</sub>                        | 67.1 g [2.36] of UO <sub>2.4</sub>             | 100% to UO <sub>2.4</sub> , 100% to UO <sub>2.4</sub>                         | 67.1 g [2.36] of UO <sub>2.4</sub>             | 100% to UO <sub>2.4</sub> , 75.9% to UO <sub>2.4</sub>                        | 2.6 g [0.09] of U <sub>3</sub> O <sub>8</sub>  | 11.4% to U <sub>3</sub> O <sub>8</sub> , 0                                    | 69.3 g [2.44] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub>  | 0.1%                  |
| 4    | 10.0 g [0.34] of UO <sub>2.4</sub>             | 44.7% to UO <sub>2.4</sub> , 0  | 12.1 g [0.43] of UO <sub>2.4</sub>             | 54.1% to UO <sub>2.4</sub> , 0  | 19.8 g [0.70] of UO <sub>2.4</sub>             | 47.5% to UO <sub>2.4</sub> , 0  | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub>  | 0.1%                  |
| 1    | 68.3 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 68.3 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 68.4 g [2.41] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 2.4 g [0.08] of U <sub>3</sub> O <sub>8</sub>  | 10.4% to U <sub>3</sub> O <sub>8</sub> , 0                                    | 7.5 g [0.26] of U <sub>3</sub> O <sub>8</sub>  | 32.8% to U <sub>3</sub> O <sub>8</sub> , 0                                    | 65.8 g [2.32] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 97.4% to U <sub>3</sub> O <sub>8</sub> | 0.01%                 |

\*Extent of Oxidation: This provides information on the percentage of exposed fuel pellets that have undergone oxidation to particular UO<sub>2+x</sub> phase. The extent of oxidation is separately calculated for the directly exposed fuel pellets and fuel pellets in the crack length, plus 6 cm area of a failed rod, but not directly exposed. The extent of oxidation values for the directly exposed fuel pellets and the adjacent fuel pellets, not directly exposed, are separated by a comma.

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**Table 12. Fuel Oxidation Simulation Data for Case 2 (Linear Decomposition of the Residual water in 4.77 Years)**

| Zone | Low-End Fuel and Cladding Initial Temperature  |   |  |   |  |   | High-End Fuel and Cladding Initial Temperature |  |  |   |  |   | Cladding Failure Rate |
|------|--|---|--|---|--|---|--|--|--|---|--|---|-----------------------|
|      | Moles of Water                                 |   |  |   |  |   |  |  |  |   |  |   |                       |
|      | 5.5  |   | 17.4   |   | 55   |   | 5.5  |  | 17.4   |   | 55   |   |                       |
|      | UO <sub>2+x</sub> Mass [oz]                    | Extent of Oxidation*  | UO <sub>2+x</sub> Mass [oz]                    | Extent of Oxidation*  | UO <sub>2+x</sub> Mass [oz]                    | Extent of Oxidation*  | UO <sub>2+x</sub> Mass [oz]                    | Extent of Oxidation*                       | UO <sub>2+x</sub> Mass [oz]                    | Extent of Oxidation*  | UO <sub>2+x</sub> Mass [oz]                    | Extent of Oxidation*  |                       |
| 1    | 3.6 g [0.13] of U <sub>3</sub> O <sub>8</sub>  | 16% to U <sub>3</sub> O <sub>8</sub> , 0                                      | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 68.2 g [2.41] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 2.4 g [0.09] of U <sub>3</sub> O <sub>8</sub>  | 10.4% to U <sub>3</sub> O <sub>8</sub> , 0 | 7.5 g [0.26] of U <sub>3</sub> O <sub>8</sub>  | 32.8% to U <sub>3</sub> O <sub>8</sub> , 0                                    | 24.4 g [0.86] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 3.7% to U <sub>3</sub> O <sub>8</sub> | 0.1%                  |
| 2    | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 2.4 g [0.09] of U <sub>3</sub> O <sub>8</sub>  | 10.4% to U <sub>3</sub> O <sub>8</sub> , 0 | 7.5 g [0.26] of U <sub>3</sub> O <sub>8</sub>  | 41.4% to U <sub>3</sub> O <sub>8</sub> , 0                                    | 39.1 g [1.37] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 36% to U <sub>3</sub> O <sub>8</sub>  | 0.1%                  |
| 3    | 67.1 g [1.73] of UO <sub>2,4</sub>             | 100% to UO <sub>2,4</sub> , 100% to UO <sub>2,4</sub>                         | 67.1 g [2.36] of UO <sub>2,4</sub>             | 100% to UO <sub>2,4</sub> , 100% to UO <sub>2,4</sub>                         | 67.1 g [2.36] of UO <sub>2,4</sub>             | 100% to UO <sub>2,4</sub> , 75.9% to UO <sub>2,4</sub>                        | 2.4 g [0.09] of U <sub>3</sub> O <sub>8</sub>  | 10.4% to U <sub>3</sub> O <sub>8</sub> , 0 | 8.8 g [0.31] of U <sub>3</sub> O <sub>8</sub>  | 38.7% to U <sub>3</sub> O <sub>8</sub> , 0                                    | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 0.1%                  |
| 4    | 11.6 g [0.34] of UO <sub>2,4</sub>             | 44.7% to UO <sub>2,4</sub> , 0  | 11.8 g [0.42] of UO <sub>2,4</sub>             | 52.7% to UO <sub>2,4</sub> , 0  | 19.8 g [0.70] of UO <sub>2,4</sub>             | 47.5% to UO <sub>2,4</sub> , 0  | 2.3 g [0.08] of U <sub>3</sub> O <sub>8</sub>  | 10.1% to U <sub>3</sub> O <sub>8</sub> , 0 | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 0.1%                  |
| 1    | 4.4 g [0.16] of U <sub>3</sub> O <sub>8</sub>  | 19.4% to U <sub>3</sub> O <sub>8</sub> , 0                                    | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 68.4 g [2.41] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 2.4 g [0.09] of U <sub>3</sub> O <sub>8</sub>  | 10.4% to U <sub>3</sub> O <sub>8</sub> , 0 | 7.5 g [0.26] of U <sub>3</sub> O <sub>8</sub>  | 32.8% to U <sub>3</sub> O <sub>8</sub> , 0                                    | 24.4 g [0.86] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 3.7% to U <sub>3</sub> O <sub>8</sub> | 0.01%                 |

\*Extent of Oxidation: This provides information on the percentage of exposed fuel pellets that have undergone oxidation to particular UO<sub>2+x</sub> phase. The extent of oxidation is separately calculated for the directly exposed fuel pellets and fuel pellets in the crack length, plus 6 cm area of a failed rod, but not directly exposed. The extent of oxidation values for the directly exposed fuel pellets and the adjacent fuel pellets, not directly exposed, are separated by a comma.

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**Table 13. Fuel Oxidation Simulation Data for Case 3 (Exponential Decomposition of the Residual Water in 71.62 Years)**

| Zone | Low-End Fuel and Cladding Initial Temperature  |   |   |   |  |   | High-End Fuel and Cladding Initial Temperature                                       |  |   |   |   |  | Cladding Failure Rate |
|------|--|---|---|---|--|---|--|--|---|---|---|--|-----------------------|
|      | Moles of Water   |   |   |   |  |   |  |  |   |   |   |  |                       |
|      | 5.5  |   | 17.4  |   | 55   |   | 5.5  |  | 17.4  |   | 55  |  |                       |
|      | UO <sub>2+x</sub> Mass g [oz]  | Extent of Oxidation*  | UO <sub>2+x</sub> Mass g [oz]   | Extent of Oxidation*  | UO <sub>2+x</sub> Mass g [oz]                  | Extent of Oxidation*  | UO <sub>2+x</sub> Mass g [oz]  | Extent of Oxidation*   | UO <sub>2+x</sub> Mass g [oz]   | Extent of Oxidation*  | UO <sub>2+x</sub> Mass g [oz]   | Extent of Oxidation*   |                       |
| 1    | 2.0 g [0.07] of U <sub>3</sub> O <sub>8</sub> and 1.5 g [0.05] of UO <sub>2.4</sub>  | 8.8% to U <sub>3</sub> O <sub>8</sub> and 6.7% to UO <sub>2.4</sub> , 0                           | 31.8 g [1.12] of U <sub>3</sub> O <sub>8</sub> and 35.7 g [1.26] of UO <sub>2.4</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 20% to U <sub>3</sub> O <sub>8</sub> and 80% to UO <sub>2.4</sub> | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 2.3 g [0.08] of U <sub>3</sub> O <sub>8</sub> and 0.1 g [0.003] of UO <sub>2.4</sub> | 10.1% to U <sub>3</sub> O <sub>8</sub> and 0.5% to UO <sub>2.4</sub> , 0 | 7.2 g [0.25] of U <sub>3</sub> O <sub>8</sub> and 0.4 g [0.014] of UO <sub>2.4</sub>  | 31.7% to U <sub>3</sub> O <sub>8</sub> and 1.8% to UO <sub>2.4</sub> , 0                          | 23.0 g [0.81] of U <sub>3</sub> O <sub>8</sub> and 25.6 g [0.90] of UO <sub>2.4</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 0.6% to U <sub>3</sub> O <sub>8</sub> and 57.2% to UO <sub>2.4</sub> | 0.1%                  |
| 2    | 2.0 g [0.07] of U <sub>3</sub> O <sub>8</sub> and 44.3 g [1.56] of UO <sub>2.4</sub> | 8.8% to U <sub>3</sub> O <sub>8</sub> and 91.2% to UO <sub>2.4</sub> , 53.4% to UO <sub>2.4</sub> | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub>  | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub>                             | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 2.2 g [0.08] of U <sub>3</sub> O <sub>8</sub> and 0.3 g [0.01] of UO <sub>2.4</sub>  | 9.7% to U <sub>3</sub> O <sub>8</sub> and 1.3% to UO <sub>2.4</sub> , 0  | 7.0 g [0.25] of U <sub>3</sub> O <sub>8</sub> and 0.9 g [0.03] of UO <sub>2.4</sub>   | 30.8% to U <sub>3</sub> O <sub>8</sub> and 4.0% to UO <sub>2.4</sub> , 0                          | 24.6 g [0.87] of U <sub>3</sub> O <sub>8</sub> and 42.9 g [1.51] of UO <sub>2.4</sub> | 100% to U <sub>3</sub> O <sub>8</sub> and 4.1% to UO <sub>2.4</sub> , 95.9% to UO <sub>2.4</sub>             | 0.1%                  |
| 3    | 67.1 g [2.36] of UO <sub>2.4</sub>   | 100% to UO <sub>2.4</sub> , 100% to UO <sub>2.4</sub>   | 67.1 g [2.36] of UO <sub>2.4</sub>  | 100% to UO <sub>2.4</sub> , 100% to UO <sub>2.4</sub>   | 67.1 g [2.36] of UO <sub>2.4</sub>             | 100% to UO <sub>2.4</sub> , 100% to UO <sub>2.4</sub>                         | 1.9 g [0.07] of U <sub>3</sub> O <sub>8</sub> and 0.7 g [0.03] of UO <sub>2.4</sub>  | 8.4% to U <sub>3</sub> O <sub>8</sub> 3.1% to UO <sub>2.4</sub> , 0      | 6.4 g [0.22] of U <sub>3</sub> O <sub>8</sub> and 12.0 g [10.42] of UO <sub>2.4</sub> | 28.2% to U <sub>3</sub> O <sub>8</sub> and 53.7% to UO <sub>2.4</sub> , 0                         | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub>  | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub>                                | 0.1%                  |
| 4    | 19.1 g [0.67] of UO <sub>2.4</sub>   | 85.4% to UO <sub>2.4</sub> , 0  | 19.4 g [0.68] of UO <sub>2.4</sub>  | 86.7% to UO <sub>2.4</sub> , 0  | 19.8 g [0.69] of UO <sub>2.4</sub>             | 88.6% to UO <sub>2.4</sub> , 0  | 1.1 g [0.04] of U <sub>3</sub> O <sub>8</sub> and 3.4 g [0.12] of UO <sub>2.4</sub>  | 4.8% to U <sub>3</sub> O <sub>8</sub> and 15.2% to UO <sub>2.4</sub> , 0 | 3.5 g [0.12] of U <sub>3</sub> O <sub>8</sub> and 63.6 g [2.24] of UO <sub>2.4</sub>  | 15.4% to U <sub>3</sub> O <sub>8</sub> and 84.6% to UO <sub>2.4</sub> , 100% to UO <sub>2.4</sub> | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub>  | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub>                                | 0.1%                  |
| 1    | 2.0 g [0.07] of U <sub>3</sub> O <sub>8</sub> and 5.8 g [0.20] of UO <sub>2.4</sub>  | 8.8% to U <sub>3</sub> O <sub>8</sub> and 25.9% to UO <sub>2.4</sub> , 0                          | 32.3 g [1.14] of U <sub>3</sub> O <sub>8</sub> and 35.3 g [1.24] of UO <sub>2.4</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 21% to U <sub>3</sub> O <sub>8</sub> and 79% to UO <sub>2.4</sub> | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 2.3 g [0.08] of U <sub>3</sub> O <sub>8</sub> and 0.1 g [0.003] of UO <sub>2.4</sub> | 10.1% to U <sub>3</sub> O <sub>8</sub> and 0.5% to UO <sub>2.4</sub> , 0 | 7.2 g [0.25] of U <sub>3</sub> O <sub>8</sub> and 0.4 g [0.014] of UO <sub>2.4</sub>  | 31.7% to U <sub>3</sub> O <sub>8</sub> and 1.8% to UO <sub>2.4</sub> , 0                          | 23.1 g [0.81] of U <sub>3</sub> O <sub>8</sub> and 25.9 g [0.91] of UO <sub>2.4</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 0.8% to U <sub>3</sub> O <sub>8</sub> and 57.9% to UO <sub>2.4</sub> | 0.01%                 |

\*Extent of Oxidation: This provides information on the percentage of exposed fuel pellets that have undergone oxidation to particular UO<sub>2+x</sub> phase. The extent of oxidation is separately calculated for the directly exposed fuel pellets and fuel pellets in the crack length, plus 6 cm area of a failed rod, but not directly exposed. The extent of oxidation values for the directly exposed fuel pellets and the adjacent fuel pellets, not directly exposed, are separated by a comma.

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**Table 14. Fuel Oxidation Simulation Data for Case 4 (Linear Decomposition of the Residual Water in 71.62 Years)**

| Zone | Low-End Fuel and Cladding Initial Temperature   |  |  |   |   |   | High-End Fuel and Cladding Initial Temperature  |  |  |  |   |   | Cladding Failure Rate |
|------|---|--|--|---|---|---|---|--|--|--|---|---|-----------------------|
|      | Moles of Water  |  |  |   |   |   |   |  |  |  |   |   |                       |
|      | 5.5   |  | 17.4   |   | 55  |   | 5.5   |  | 17.4   |  | 55  |   |                       |
|      | UO <sub>2+x</sub> Mass [oz]   | Extent of Oxidation*   | UO <sub>2+x</sub> Mass [oz]  | Extent of Oxidation*  | UO <sub>2+x</sub> Mass [oz]   | Extent of Oxidation*  | UO <sub>2+x</sub> Mass [oz]   | Extent of Oxidation*   | UO <sub>2+x</sub> Mass [oz]  | Extent of Oxidation*   | UO <sub>2+x</sub> Mass [oz]   | Extent of Oxidation*  |                       |
| 1    | 0.5 g [0.02] of U <sub>3</sub> O <sub>8</sub> and 25.3 g [0.90] of UO <sub>2,4</sub>  | 2.2% to U <sub>3</sub> O <sub>8</sub> and 97.8% to UO <sub>2,4</sub> , 7.6% to UO <sub>2,4</sub> | 2.0 g [0.07] of U <sub>3</sub> O <sub>8</sub> and 65.1 g [2.30] of UO <sub>2,4</sub> | 8.8% to U <sub>3</sub> O <sub>8</sub> and 91.2% to UO <sub>2,4</sub> , 100% to UO <sub>2,4</sub>  | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub>  | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub>                     | 0.9 g [0.03] of U <sub>3</sub> O <sub>8</sub> and 17.0 g [0.60] of UO <sub>2,4</sub>  | 4% to U <sub>3</sub> O <sub>8</sub> and 76% to UO <sub>2,4</sub> , 0     | 2.9 g [0.10] of U <sub>3</sub> O <sub>8</sub> and 48.0 g [1.69] of UO <sub>2,4</sub> | 12.8% to U <sub>3</sub> O <sub>8</sub> and 87.2% to UO <sub>2,4</sub> , 63.6% to UO <sub>2,4</sub> | 36.9 g [0.13] of U <sub>3</sub> O <sub>8</sub> and 30.7 g [1.73] of UO <sub>2,4</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 31.2% to U <sub>3</sub> O <sub>8</sub> and 68.8% to UO <sub>2,4</sub> | 0.1%                  |
| 2    | 0.2 g [0.007] of U <sub>3</sub> O <sub>8</sub> and 20.4 g [0.72] of UO <sub>2,4</sub> | 0.9% to U <sub>3</sub> O <sub>8</sub> and 91.2% to UO <sub>2,4</sub> , 0                         | 0.6 g [0.02] of U <sub>3</sub> O <sub>8</sub> and 63.2 g [2.22] of UO <sub>2,4</sub> | 2.6% to U <sub>3</sub> O <sub>8</sub> and 97.4% to UO <sub>2,4</sub> , 97.5% to UO <sub>2,4</sub> | 14.9 g [0.53] of U <sub>3</sub> O <sub>8</sub> and 52.4 g [1.85] of UO <sub>2,4</sub> | 65.6% to U <sub>3</sub> O <sub>8</sub> and 34.4% to UO <sub>2,4</sub> , 100% to UO <sub>2,4</sub> | 0.7 g [0.03] of U <sub>3</sub> O <sub>8</sub> and 14.5 g [1.73] of UO <sub>2,4</sub>  | 3% to U <sub>3</sub> O <sub>8</sub> and 64.8% to UO <sub>2,4</sub> , 0   | 2.2 g [0.08] of U <sub>3</sub> O <sub>8</sub> and 36.9 g [1.30] of UO <sub>2,4</sub> | 9.7% to U <sub>3</sub> O <sub>8</sub> and 90.3% to UO <sub>2,4</sub> , 37.3% to UO <sub>2,4</sub>  | 11.1 g [0.13] of U <sub>3</sub> O <sub>8</sub> and 56.2 g [1.73] of UO <sub>2,4</sub> | 48.9% to U <sub>3</sub> O <sub>8</sub> and 51.1% to UO <sub>2,4</sub> , 100% to UO <sub>2,4</sub>             | 0.1%                  |
| 3    | 20.2 g [0.71] of UO <sub>2,4</sub>  | 90.3% to UO <sub>2,4</sub> , 0   | 67.1 g [2.36] of UO <sub>2,4</sub>   | 100% to UO <sub>2,4</sub> , 100% to UO <sub>2,4</sub>   | 67.1 g [2.36] of UO <sub>2,4</sub>  | 100% to UO <sub>2,4</sub> , 100% to UO <sub>2,4</sub>   | 0.4 g [0.13] of U <sub>3</sub> O <sub>8</sub> and 13.2 g [0.51] of UO <sub>2,4</sub>  | 1.8% to U <sub>3</sub> O <sub>8</sub> and 59% to UO <sub>2,4</sub> , 0   | 1.4 g [0.05] of U <sub>3</sub> O <sub>8</sub> and 31.4 g [1.11] of UO <sub>2,4</sub> | 6.2% to U <sub>3</sub> O <sub>8</sub> and 93.8% to UO <sub>2,4</sub> , 23.2% to UO <sub>2,4</sub>  | 4.8 g [0.17] of U <sub>3</sub> O <sub>8</sub> and 64.2 g [1.73] of UO <sub>2,4</sub>  | 21.1% to U <sub>3</sub> O <sub>8</sub> and 78.9% to UO <sub>2,4</sub> , 100% to UO <sub>2,4</sub>             | 0.1%                  |
| 4    | 16.4 g [0.58] of UO <sub>2,4</sub>  | 70.3% to UO <sub>2,4</sub> , 0   | 18.6 g [0.65] of UO <sub>2,4</sub>   | 83.2% to UO <sub>2,4</sub> , 0  | 17.0 g [0.60] of UO <sub>2,4</sub> and 51.9 g [1.83] of UO <sub>3</sub>               | 76% to UO <sub>2,4</sub> and 24% to UO <sub>2,4</sub> , 100% to UO <sub>3</sub>                   | 0.1 g [0.003] of U <sub>3</sub> O <sub>8</sub> and 13.3 g [0.47] of UO <sub>2,4</sub> | 0.4% to U <sub>3</sub> O <sub>8</sub> and 59.5% to UO <sub>2,4</sub> , 0 | 0.4 g [0.13] of U <sub>3</sub> O <sub>8</sub> and 32.9 g [1.73] of UO <sub>2,4</sub> | 1.9% to U <sub>3</sub> O <sub>8</sub> and 98.2% to UO <sub>2,4</sub> , 38.5% to UO <sub>2,4</sub>  | 1.5 g [0.05] of U <sub>3</sub> O <sub>8</sub> and 65.6 g [2.31] of UO <sub>2,4</sub>  | 6.6% to U <sub>3</sub> O <sub>8</sub> and 93.4% to UO <sub>2,4</sub> , 100% to UO <sub>2,4</sub>              | 0.1%                  |
| 1    | 0.5 g [0.02] of U <sub>3</sub> O <sub>8</sub> and 26.2 g [0.92] of UO <sub>2,4</sub>  | 2.2% to U <sub>3</sub> O <sub>8</sub> and 97.8% to UO <sub>2,4</sub> , 9.6% to UO <sub>2,4</sub> | 2.0 g [0.07] of U <sub>3</sub> O <sub>8</sub> and 65.1 g [2.30] of UO <sub>2,4</sub> | 8.8% to U <sub>3</sub> O <sub>8</sub> and 91.2% to UO <sub>2,4</sub> , 100% to UO <sub>2,4</sub>  | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub>  | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub>                     | 0.9 g [0.03] of U <sub>3</sub> O <sub>8</sub> and 17.4 g [0.61] of UO <sub>2,4</sub>  | 4% to U <sub>3</sub> O <sub>8</sub> and 77.8% to UO <sub>2,4</sub> , 0   | 2.9 g [0.10] of U <sub>3</sub> O <sub>8</sub> and 48.9 g [1.72] of UO <sub>2,4</sub> | 12.8% to U <sub>3</sub> O <sub>8</sub> and 87.2% to UO <sub>2,4</sub> , 65.6% to UO <sub>2,4</sub> | 38.8 g [1.37] of U <sub>3</sub> O <sub>8</sub> and 28.9 g [1.02] of UO <sub>2,4</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 35.4% to U <sub>3</sub> O <sub>8</sub> and 64.6% to UO <sub>2,4</sub> | 0.01%                 |

\*Extent of Oxidation: This provides information on the percentage of exposed fuel pellets that have undergone oxidation to particular UO<sub>2+x</sub> phase. The extent of oxidation is separately calculated for the directly exposed fuel pellets and fuel pellets in the crack length, plus 6 cm area of a failed rod, but not directly exposed. The extent of oxidation values for the directly exposed fuel pellets and the adjacent fuel pellets, not directly exposed, are separated by a comma.

As seen in the figure, the cladding oxidation thickness from zones 2 to 4 for low-end fuel and cladding initial temperature is less compared to high-end fuel and cladding initial temperature. However, this trend reversed for zone 1, i.e., more cladding oxidation occurs for low-end fuel and cladding initial temperature compared to high-end fuel and cladding initial temperature in zone 1. An explanation for this observation is following. The rate of cladding and fuel oxidation is dependent upon temperature and also on the rate of radiolysis of water. Moreover, any unused oxygen in a zone is transferred to next hotter zone in the integration model. Since, temperatures of zones 2 to 5 for low-end fuel and cladding initial temperature condition are less compared to the high-end fuel and cladding initial temperature condition, the rate of oxidation is also less, and thus more oxidation of cladding occurs for zones 2 to 4 for high-end fuel and cladding initial temperature condition. However, more oxidation occur for zone 1 cladding for low-end fuel and cladding initial temperature condition because more unused oxygen becomes available in zone 1 compared to the high-end fuel and cladding initial temperature condition. This trend is observed in case 1 to 3 cladding oxidation data.

However, the same is not observed in the case 4 data. This is attributed to slow the rate of radiolysis in case when the residual water linearly decomposed in 71.62 years. For the condition of low-end fuel and cladding initial temperature, the cladding cools and no appreciable amount of oxidation occurs beyond 20 years; however, the cladding oxidation continues for the long time period for the low-end fuel and cladding initial temperature condition.

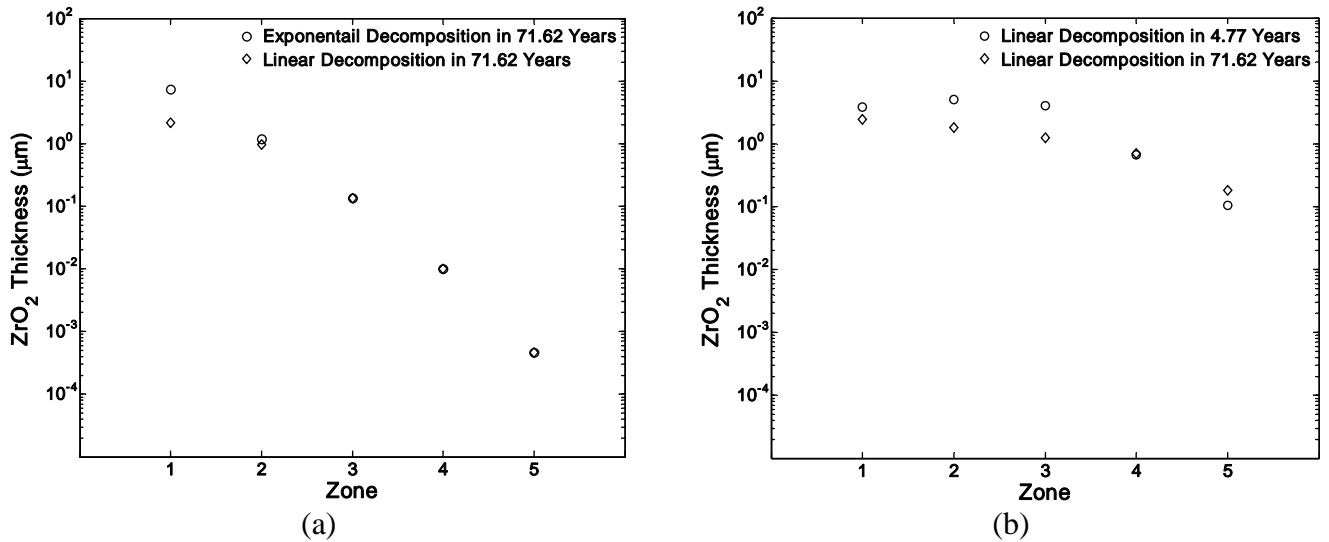
### **Cladding Failure Rate**

The cladding failure rate does not significantly affect the cladding oxidation. This trend is observed for cases 1 to 4. The cladding failure rate is used to determine the number of exposed fuel pellets to the canister. Thus, the higher cladding failure rate results in more number of exposed fuel pellets. However, this does not significantly affect the extent of cladding oxidation. This is because the amount of oxygen needed to oxidize fuel pellet is much smaller compared to the cladding. For example, only 0.013 moles of oxygen is needed to completely oxidize a fuel pellet from  $\text{UO}_2$  to  $\text{U}_3\text{O}_8$ .

### **Radiolysis Kinetics**

The data listed in Tables 7 and 8 for case 1 and 2, respectively, indicate that selection of either exponential or linear decomposition of the residual water in 4.77 years does not significantly affect the extent of cladding oxidation. However, the same is not observed when decomposition period is 71.62 years. The data from case 3 and 4 for 55 moles of water, 0.1 percent cladding failure, and low-end fuel and cladding initial temperature condition is used to show the effect of linear and exponential decomposition of the residual water in 71.62 years on the cladding oxidation. This data is presented in Figure 4(a). As seen in the figure, the cladding oxidizes more for zone 1 and 2 when the kinetic model for the decomposition of the residual water is exponential compared to linear. However, no different in the  $\text{ZrO}_2$  thickness is noted for zone 3, 4, and 5. This observation is attributed to the compound effect of the fuel temperature and oxygen generation rate. When water decomposes via exponential kinetics, more oxygen is available within first 10 years after storage compared to linear kinetics. The cladding oxidation in zone 3, 4, and 5 is dependent upon temperatures whereas in zone 1 and 2 is dependent upon rate of oxygen generation. Because more oxygen is generated with exponential kinetics, the more oxidation of the cladding occurs in zone 1 and 2 compared to linear kinetics.





**Figure 4. (a) Cases 3 and 4 Cladding Oxidation Data for 55 Moles of Water, 0.1 Percent Cladding Failure, and Low-End Fuel and Cladding Initial Temperature to Highlight The Effect of Radiolysis Model. (b) Cases 2 and 4 Cladding Oxidation Data for 55 Moles of Water, 0.1 Percent Cladding Failure, and High-End Fuel and Cladding Initial Temperature to Highlight the Effect of Decomposition Periods on Cladding Oxidation.**

The cladding oxidation data for case 1 and 3 indicated that there is little or no difference in cladding oxide thickness when the water decomposes via linear kinetics either in 4.77 or 71.62 years. More pronounced differences are observed when Case 2 and 4 cladding oxidation data for 55 moles of water, 0.1 percent Cladding Failure, and High-End Fuel and Cladding Initial Temperature is selected. This data is graphically presented in Figure 4(b). As seen in the figure, the ZrO<sub>2</sub> thickness in zone 1, 2, and 3 is more for the decomposition period of 4.77 compared to 71.62 years. This is a result of compound effect of temperature and availability of oxygen. When more oxygen is available at higher temperature and the rate of cladding oxidation is controlled by availability of the oxygen, more oxidation occurs for the shorter decomposition period.

### Water Amount

It is observed that more cladding oxidation occurs for higher amounts of residual water. This trend is observed for cases 1 to 4, and is independent of cladding failure rate. For example, in case 1 for the low-end fuel and cladding temperature, the cladding oxide thickness increased from 1.24 μm [4.87E-02 mils] to 7.32 μm [2.88E-01 mils] for the zone 1 exposed cladding when the residual water amount was increased from 5.5 to 55 moles. The same trend is observed in case 2, 3, and 4 also. When the water amount is less than 5.5 moles, it is expected that the extent of oxidation would be also be lesser compared the values obtained for the 5.5 moles of the residual water.

### Thermal Decay Constant

The cladding oxidation data for case 5 is listed in Table 15 to highlight the effect of thermal decay constant. The list data is for low-end fuel and cladding initial temperature, cladding failure rate of 0.1 percent, radiolysis kinetics of exponential decay of residual water in 71.62 years, water amount of 17.4 moles, and oxygen diffusion through grain boundaries. The value of thermal decay constant is varied.

The data indicates that more cladding oxidation occurs with the lower value decay constant in all five zones. This is expected because cladding temperature remains at higher values for longer periods with the lower value of thermal decay constant. Hence, more cladding oxidizes.

| <b>Table 15. Cladding Oxidation Simulation Data for Case 5<br/>(Linear Decomposition of the Residual Water in 71.62 Years, Low-End Fuel and Cladding Initial Temperature, 17.4 Moles of Residual Water, and Cladding Failure Rate of 0.1 Percent). The Values of Decay Constant is Varied.</b> |   |   |
|--|---|---|
| <b>Zone</b>  | <b>Cladding Oxides Layer Thickness (<math>\mu\text{m}</math>) [mil] for Thermal Decay Constant Value of 0.023</b> | <b>Cladding Oxides Layer Thickness (<math>\mu\text{m}</math>) [mil] for Thermal Decay Constant Value of 0.064</b> |
| 1  | 7.70E-01  | 2.74E-01  |
| 2  | 4.57E-01  | 1.63E-01  |
| 3  | 1.36E-01  | 4.90E-02  |
| 4  | 1.03E-02  | 3.95E-03  |
| 5  | 4.38E-04  | 1.81E-04  |

### Mode of Oxygen Diffusion

The cladding oxidation data for case 6 is listed in Table 16 to highlight the effect of mode of oxygen diffusion through exposed fuel pellets. The listed data is for low-end fuel and cladding initial temperature, cladding failure rate of 0.1 percent, radiolysis kinetics of exponential decomposition in 4.77 years, 17.4 moles of the residual water, and thermal decay constant equal to 0.023. The mode of oxygen diffusing through the exposed fuel pellet is varied.

It is observed that there is less than 10 percent difference in the two values of the oxide layer thickness in each zone. The  $\text{ZrO}_2$  thickness is slightly less for the oxygen diffusion through grain boundaries compared to the fragment surface. This is attributed to the fact that more oxygen is consumed by the fuel when oxygen diffuses through the fuel

| <b>Table 16. Fuel Oxidation Simulation Data for Case 6 (Low-End Fuel and Cladding Initial Temperature, Cladding Failure Rate of 0.1 Percent, Exponential Decay of Residual Water in 4.77 Years, Water Amount 17.4 Moles, and Decay Constant Value of 0.023). The Mode of Oxygen Diffusion is Varied.</b> |   |  |
|--|---|--|
| <b>Zone</b>  | <b>Cladding Oxides Layer Thickness (<math>\mu\text{m}</math>) [mil] for Oxygen Diffusion Through Grain Boundaries of Each Exposed Fuel Pellet</b> | <b>Cladding Oxides Layer Thickness (<math>\mu\text{m}</math>) [mil] for Oxygen Diffusion Through Exposed Surfaces of Fuel Pellet Fragments</b> |
| 1  | 4.80E+00  | 4.92E+00   |
| 2  | 7.58E-01  | 7.78E-01   |
| 3  | 8.23E-02  | 8.45E-02   |
| 4  | 5.44E-03  | 5.56E-03   |

|   |          |          |
|---|----------|----------|
| 5 | 2.03E-04 | 1.83E-04 |
|---|----------|----------|

## Fuel Oxidation

This section provides information on various varying parameters effects on fuel oxidation.

### Fuel and Cladding Initial Temperature

The fuel oxidation data form Case 1 is extracted to illustrate the fuel and cladding initial temperature effect. The fuel oxidation data for low- and high-end fuel and cladding initial temperature, 0.1 percent cladding failure rate, exponential decomposition of the residual water in 4.77 years, 5.5 moles of water, oxygen diffusing through grain boundaries, and decay constant equal to 0.023 is listed in Table 17.

The listed data indicate that exposed fuel oxidizes more for the low-end fuel and cladding initial temperature condition compared to the high-end fuel and cladding initial temperature. However,  $UO_{2+x}$  phase in zone 3 and 4 for the low-end condition is  $UO_{2.4}$  whereas the oxide phase is  $U_3O_8$  in the two zones for the high-end fuel and cladding initial temperature condition. An examination of the Case 2 data for 5.5 moles of water also indicates the same trend.

To further examine this trend, the data from the four cases for 0.1 percent cladding failure rate is listed in Table 18. The mass of various  $UO_{2+x}$  phase in the five zones is added. The listed data indicate that for 5.5 moles of residual water, more fuel oxidizes with the low-end condition in the four cases. However, for 17.4 moles of the residual water, more fuel oxidizes with the low-end condition in case 1, 2, and 3. In case 4, more fuel oxidation occurs for 17.4 moles with the high-end condition.

For the 55 moles of the residual water, more fuel oxidizes for the low-end condition in case 1 and 2. However, most of the oxidized fuel for the high-end condition in case 1 and 2 results in formation of  $U_3O_8$ . In case 3, more fuel oxidizes and forms  $U_3O_8$  for the high-end condition compared to the low-end condition. In case 4, the amount of oxidized fuel for the low- and high-end conditions are comparable. However, note that more  $U_3O_8$  forms for the low-end condition compared to the high-end condition.

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**Table 17. Mass of  $UO_{2+x}$  Phase and Extent of Oxidation for Exponential Decomposition of the Residual Water in 4.77 Years, 0.1 Percent Cladding Failure Rate, 5.5 Moles of Water, Oxygen Diffusing Through Grain Boundaries, and Decay Constant Equal to 0.023**

| Zone | Low-End Fuel and Cladding Initial Temperature |   | High-End Fuel and Cladding Initial Temperature |  |
|------|---|---|--|--|
|      | Mass of $UO_{2+x}$ Phase [oz]                 | Extent of Oxidation*                        | Mass of $UO_{2+x}$ Phase [oz]                  | Extent of Oxidation*                   |
| 1    | 68.2 g [2.40]<br>of $U_3O_8$                  | 100% to $U_3O_8$ ,<br>100% to $U_3O_8$      | 2.4 g [0.08] of<br>$U_3O_8$                    | 10.4% to $U_3O_8$ , 0                  |
| 2    | 68.2 g [2.40]<br>of $U_3O_8$                  | 100% to $U_3O_8$ ,<br>100% to $U_3O_8$      | 2.4 g [0.08] of<br>$U_3O_8$                    | 10.4% to $U_3O_8$ , 0                  |
| 3    | 49.2 g [1.73]<br>of $UO_{2.4}$                | 100% to $UO_{2.4}$ ,<br>60.0% to $UO_{2.4}$ | 2.6 g [0.09] of<br>$U_3O_8$                    | 11.4% to $U_3O_8$ , 0                  |
| 4    | 10.0 g [0.34]<br>of $UO_{2.4}$                | 44.7% to $UO_{2.4}$ , 0                     | 68.2 g [2.40]<br>of $U_3O_8$                   | 100% to $U_3O_8$ ,<br>100% to $U_3O_8$ |

\*Extent of Oxidation: This provides information on the percentage of exposed fuel pellets that have undergone oxidation to particular  $UO_{2+x}$  phase. The extent of oxidation is separately calculated for the directly exposed fuel pellets and fuel pellets in the crack length, plus 6 cm area of a failed rod, but not directly exposed. The extent of oxidation values for the directly exposed fuel pellets and the adjacent fuel pellets, not directly exposed, are separated by a comma.

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| <b>Table 18. Mass of UO<sub>2+x</sub> Phase for Different Water Amount Under Cases 1 to 4. The Data is for 0.1 Percent Cladding Failure Rate. The Fuel Oxidation Data for the Five Zones is Added.</b> |  |   |  |   |  |  |
|--|--|---|--|---|--|--|
| <b>Case Number</b>   | <b>Low-End Fuel and Cladding Initial Temperature</b>                                 |   |  | <b>High-End Fuel and Cladding Initial Temperature</b>                               |  |  |
|  | <b>Mass of UO<sub>2+x</sub> Phase [oz] for Water Amount (moles)</b>                  |   |  | <b>Mass of UO<sub>2+x</sub> Phase [oz] for Water Amount (moles)</b>                 |  |  |
|  | <b>5.5</b>   | <b>17.4</b>   | <b>55</b>  | <b>5.5</b>  | <b>17.4</b>  | <b>55</b>  |
| 1  | 136.4 g [4.8] of U <sub>3</sub> O <sub>8</sub> and 59.2 g [2.1] of UO <sub>2.4</sub> | 136.5 g [4.8] of U <sub>3</sub> O <sub>8</sub> and 79.2 g [2.8] of UO <sub>2.4</sub>  | 136.6 g [4.8] of U <sub>3</sub> O <sub>8</sub> and 86.9 g [3.1] of UO <sub>2.4</sub>                                     | 75.6 g [2.7] of U <sub>3</sub> O <sub>8</sub>                                       | 153.3 g [5.4] of U <sub>3</sub> O <sub>8</sub>                                       | 268.3 g [9.5] of U <sub>3</sub> O <sub>8</sub>                                       |
| 2  | 72.0 g [2.5] of U <sub>3</sub> O <sub>8</sub> and 78.8 g [2.8] of UO <sub>2.4</sub>  | 136.4 g [4.8] of U <sub>3</sub> O <sub>8</sub> and 79.0 g [2.8] of UO <sub>2.4</sub>  | 136.4 g [4.8] of U <sub>3</sub> O <sub>8</sub> and 86.9 g [3.1] of UO <sub>2.4</sub>                                     | 9.4 g [0.3] of U <sub>3</sub> O <sub>8</sub> and 0.5 g [0.02] of UO <sub>2.4</sub>  | 92.0 g [3.2] of U <sub>3</sub> O <sub>8</sub>  | 199.8 g [7.0] of U <sub>3</sub> O <sub>8</sub>                                       |
| 3  | 4.0 g [0.14] of U <sub>3</sub> O <sub>8</sub> and 131.9 g [4.6] of UO <sub>2.4</sub> | 100.1 g [3.5] of U <sub>3</sub> O <sub>8</sub> and 122.2 g [4.3] of UO <sub>2.4</sub> | 136.4 g [4.8] of U <sub>3</sub> O <sub>8</sub> and 86.9 g [3.1] of UO <sub>2.4</sub>                                     | 7.5 g [0.26] of U <sub>3</sub> O <sub>8</sub> and 4.5 g [0.16] of UO <sub>2.4</sub> | 24.1 g [0.84] of U <sub>3</sub> O <sub>8</sub> and 76.9 g [2.7] of UO <sub>2.4</sub> | 183.9 g [6.5] of U <sub>3</sub> O <sub>8</sub> and 68.5 g [2.4] of UO <sub>2.4</sub> |
| 4  | 0.6 g [0.02] of U <sub>3</sub> O <sub>8</sub> and 214.1 g [7.6] of UO <sub>2.4</sub> | 2.6 g [0.09] of U <sub>3</sub> O <sub>8</sub> and 86.5 g [7.5] of UO <sub>2.4</sub>   | 83.1 g [2.9] of U <sub>3</sub> O <sub>8</sub> and 136.6 g [4.8] of UO <sub>2.4</sub> and 51.9 g [1.8] of UO <sub>3</sub> | 2.2 g [0.08] of U <sub>3</sub> O <sub>8</sub> and 58.1 [2.1] g of UO <sub>2.4</sub> | 6.9 g [0.24] of U <sub>3</sub> O <sub>8</sub> and 149.3 g [5.3] of UO <sub>2.4</sub> | 51.4 g [1.9] of U <sub>3</sub> O <sub>8</sub> and 214.9 g [7.6] of UO <sub>2.4</sub> |

The fuel oxidation in a zone of the canister is affected by availability of the oxygen, and rate of cladding and fuel oxidation. The unused oxygen from colder zones is transferred to hotter zones in the integration model. For the low-end fuel and cladding initial temperature, more unused oxygen is transferred from colder zones to hotter zones compared to the high-end fuel and cladding initial temperature. This result is higher extent of oxidation of the exposed fuel in the hotter zones for the low-end temperature compared to the high-end condition. However, the exposed fuel for the high-end condition tends to oxidize to U<sub>3</sub>O<sub>8</sub>. This occurs when the residual water decomposes either through exponential or linear kinetics in 4.77 years and exponential kinetics in 71.62 years. When the residual water decomposes through linear kinetics in 71.62 years, a compound effect of cladding oxidation, fuel oxidation rate, and availability of the oxygen determine the extent of fuel oxidation. In the integration model, the available oxygen is divided between fuel and cladding according to surface area proportions (see Eqs. 12 and 13). When more oxygen is available in during time step in a zone, the more oxygen is allocated to the fuel.

### Cladding Failure Rate

Higher amount of fuel is exposed to the radiolysis-generated oxygen when the cladding failure rate is 0.1 percent compared to 0.01 percent. Thus, more fuel gets oxidized for higher value of cladding failure rate. Only fuel pellets in zone 1 is exposed to the oxygen for cladding failure rate of 0.01 percent whereas fuel pellets in zones 1 to 4 are exposed to the oxygen for cladding failure rate of 0.1 percent. To illustrate the affect of cladding failure rate on oxidation of zone 1 fuel pellet, the data is extracted from the first four cases, and is listed in Table 19. The data indicate that there is less than 20 percent change in the extent of fuel oxidation between the two cladding failure rates provided other conditions are same. This trend is observed in data for four cases. This data also indicate that the higher value of cladding failure rate does not affect the extent of fuel oxidation in different zones. This is due to fact that very small amount of oxygen is needed to oxidize a fuel pellet compared to total oxygen generated by radiolysis of either 5.5 or 17.4 or 55 moles of water.

### **Radiolysis Kinetics**

To discern the effect of radiolysis kinetics on fuel oxidation, the data form case 1, 2, and 4 is extracted and listed in Table 20. The compiled data is for low-end fuel and cladding initial temperature, 0.1 percent cladding failure rate, 17.4 moles of water, oxygen diffusion through grain boundaries, and the thermal decay constant equal to 0.023.

It is observed that the extent of oxidation is different for zone 4 exposed fuel pellets when radiolysis model is changed from exponential to linear for the decomposition period of 4.77 years (i.e., cases 1 and 2). The difference in the extent of oxidation is less than 10 percent. This indicate that when the residual water decomposes relatively soon compared to the storage period, the radiolysis kinetic model (i.e., either linear or exponential), does not significantly affect the extent of fuel oxidation. Between the four cases, it is observed that extent of oxidation for zones 3 and 4 exposes fuel pellets is nearly same. The cladding oxidation in zones 3 and 4 is negligible compared to zones 1 and 2 because of its temperature. Therefore, most of the available oxygen in zones 3 and 4 is used to oxidize fuel.

It is also observed that the extent of oxidation is significantly different for zone 1 and 2 exposed fuel pellets when radiolysis model is changed from exponential to linear for the decomposition

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**Table 19. Effect of Cladding Failure Rate on Fuel Oxidation for Zone 1 Exposed Fuel Pellets. The Fuel Oxidation Data is Extracted From the First Four Cases. The Extracted Data Is for Low-End Fuel and Cladding Initial Temperature.**

| Water Amount (Moles) | Mass of UO <sub>2+x</sub> [oz] Phase for Cladding Failure Rate of 0.1% |  |  |  | Mass of UO <sub>2+x</sub> [oz] Phase for Cladding Failure Rate of 0.01% |  |   |  |
|----------------------|--|--|--|--|---|--|---|--|
|                      | Case 1   | Case 2   | Case 3   | Case 4   | Case 1  | Case 2   | Case 3  | Case 4   |
| 5.5                  | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub>                         | 3.6 g [0.13] of U <sub>3</sub> O <sub>8</sub>  | 2.0 g [0.07] of U <sub>3</sub> O <sub>8</sub> and 1.5 g [0.05] of UO <sub>2.4</sub>  | 2.4 g [0.0847] of U <sub>3</sub> O <sub>8</sub>                                      | 68.3 g [2.40] of U <sub>3</sub> O <sub>8</sub>                          | 4.4 g [0.16] of U <sub>3</sub> O <sub>8</sub>  | 2.0 g [0.07] of U <sub>3</sub> O <sub>8</sub> and 5.8 g [0.20] of UO <sub>2.4</sub>   | 0.5 g [0.02] of U <sub>3</sub> O <sub>8</sub> and 26.2 g [0.92] of UO <sub>2.4</sub> |
| 17.4                 | 68.3 g [2.40] of U <sub>3</sub> O <sub>8</sub>                         | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 0.5 g [0.02] of U <sub>3</sub> O <sub>8</sub> and 25.3 g [0.90] of UO <sub>2.4</sub> | 2.0 g [0.07] of U <sub>3</sub> O <sub>8</sub> and 65.1 g [2.30] of UO <sub>2.4</sub> | 68.3 g [2.40] of U <sub>3</sub> O <sub>8</sub>                          | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 32.3 g [1.14] of U <sub>3</sub> O <sub>8</sub> and 35.3 g [1.24] of UO <sub>2.4</sub> | 2.0 g [0.07] of U <sub>3</sub> O <sub>8</sub> and 65.1 g [2.30] of UO <sub>2.4</sub> |
| 55                   | 68.4 g [2.41] of U <sub>3</sub> O <sub>8</sub>                         | 68.2 g [2.41] of U <sub>3</sub> O <sub>8</sub> | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub>                                       | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub>                                       | 68.4 g [2.41] of U <sub>3</sub> O <sub>8</sub>                          | 68.4 g [2.41] of U <sub>3</sub> O <sub>8</sub> | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub>  | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub>                                       |

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**Table 20. Compilation of Cases 1, 2, 3, and 4 Simulation Data for Identification of Radiolysis Kinetics Effect on Fuel Oxidation. The Selected Data Is for Low-End Fuel and Cladding Initial Temperature, 0.1 Percent Cladding Failure Rate, 17.4 Moles of the Residual Water, Oxygen Diffusion Through Grain Boundaries, and Thermal Decay Constant Equal to 0.023.**

| Zone | Radiolysis Model                               |   |  |   |   |   |  |   |
|------|--|---|--|---|---|---|--|---|
|      | Exponential in 4.77 Years                      |   | Linear in 4.77 Years                           |   | Exponential in 71.62 Years  |   | Linear in 71.62 Years  |   |
|      | Mass of UO <sub>2+x</sub> Phase [oz]           | Extent of Oxidation*  | Mass of UO <sub>2+x</sub> Phase [oz]           | Extent of Oxidation*  | Mass of UO <sub>2+x</sub> Phase [oz]  | Extent of Oxidation*  | Mass of UO <sub>2+x</sub> Phase [oz]   | Extent of Oxidation*  |
| 1    | 68.3 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 31.8 g [1.12] of U <sub>3</sub> O <sub>8</sub> and 35.7 g [1.26] of UO <sub>2.4</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 20% to U <sub>3</sub> O <sub>8</sub> and 80% to UO <sub>2.4</sub> | 2.0 g [0.07] of U <sub>3</sub> O <sub>8</sub> and 65.1 g [2.30] of UO <sub>2.4</sub> | 8.8% to U <sub>3</sub> O <sub>8</sub> and 91.2% to UO <sub>2.4</sub> , 100% to UO <sub>2.4</sub>  |
| 2    | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub>  | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub>                             | 0.6 g [0.02] of U <sub>3</sub> O <sub>8</sub> and 63.2 g [2.22] of UO <sub>2.4</sub> | 2.6% to U <sub>3</sub> O <sub>8</sub> and 97.4% to UO <sub>2.4</sub> , 97.5% to UO <sub>2.4</sub> |
| 3    | 67.1 g [2.36] of UO <sub>2.4</sub>             | 100% to UO <sub>2.4</sub> , 100% to UO <sub>2.4</sub>                         | 67.1 g [2.36] of UO <sub>2.4</sub>             | 100% to UO <sub>2.4</sub> , 100% to UO <sub>2.4</sub>                         | 67.1 g [2.36] of UO <sub>2.4</sub>  | 100% to UO <sub>2.4</sub> , 100% to UO <sub>2.4</sub>   | 67.1 g [2.36] of UO <sub>2.4</sub>   | 100% to UO <sub>2.4</sub> , 100% to UO <sub>2.4</sub>   |
| 4    | 12.1 g [0.43] of UO <sub>2.4</sub>             | 54.1% to UO <sub>2.4</sub> , 0  | 11.8 g [0.42] of UO <sub>2.4</sub>             | 52.7% to UO <sub>2.4</sub> , 0  | 19.4 g [0.68] of UO <sub>2.4</sub>  | 86.7% to UO <sub>2.4</sub> , 0  | 18.6 g [0.65] of UO <sub>2.4</sub>   | 83.2% to UO <sub>2.4</sub> , 0  |

\*Extent of Oxidation: This provides information on the percentage of exposed fuel pellets that have undergone oxidation to particular UO<sub>2+x</sub> phase. The extent of oxidation is separately calculated for the directly exposed fuel pellets and fuel pellets in the crack length, plus 6 cm area of a failed rod, but not directly exposed. The extent of oxidation values for the directly exposed fuel pellets and the adjacent fuel pellets, not directly exposed, are separated by a commas.

period of 71.62 years (i.e., cases 3 and 4). The residual water decomposes slowly with the decomposition period of 71.62 years compared to 4.77 years. With the exponential model, more oxygen is generated in the first 20 years compared to the linear model. This results in more oxygen being available to oxidize fuel.

The compiled data for linear radiolysis in 4.77 and 71.62 years indicate that the extent of oxidation increases for exposed pellets in zone 4 when the decomposition period is increased from 4.77 to 71.62 years. Moreover, the oxide phase in zone 1 and 2 exposed pellets shifts from U<sub>3</sub>O<sub>8</sub> to UO<sub>2.4</sub>. For example, 68.2 g of U<sub>3</sub>O<sub>8</sub> forms in zone 1 with linear radiolysis in 4.77 years whereas only 2.0 g of U<sub>3</sub>O<sub>8</sub> forms in zone 1 with linear radiolysis in 71.62 years. The same trend is observed for the exposed fuel pellets in zone 2. This trend is because of slower rate of oxygen generation during linear radiolysis in 71.62 years compared to linear radiolysis in 4.77 years. The slower rate of oxygen generation results in formation of UO<sub>2.4</sub> in the integration model.

### Water Amount

The data compiled in Table 19 also illustrates the effect of water amount on fuel oxidation. The fuel oxide amount increases with increasing water amount. This is expected because the amount of oxygen



produced due to radiolysis also increases with increasing water amount. This trend is observed for the first four cases. For example, the data in Table 18 for case 1 and the low-end fuel and cladding temperature specifies that the amount of fuel oxidized is 136.4 g [4.8] of  $U_3O_8$  and 59.2 g [2.1] of  $UO_{2.4}$  for 5.5 moles of the residual water. The oxidized fuel amount increases to 136.6 g [4.8] of  $U_3O_8$  and 86.9 [3.1] g of  $UO_{2.4}$  for the same set of conditions in case 1 but for 55 moles of the residual water. This trend is also observed for case 2, 3, and 4 data listed in Table 18. This clearly indicates that the increasing amount of residual water increases extent of fuel oxidation.

### **Thermal Decay Constant**

The simulation data for case 3 and 5 is used to discern the affect of thermal decay constant on fuel oxidation. The data is listed in Table 21. The key differences between the two cases are the extent of oxidation and  $UO_{2+x}$  phase. As noted in the table, the extent of oxidation for zone 4 fuel pellets is higher with higher value of the decay constant. However, no difference is observed in the extent of oxidation for zone 1, 2, and 3 exposed fuel pellets between the two cases.

The data also indicate that more  $U_3O_8$  forms with the lower value of the decay constant in zones 1 and 2, whereas more  $UO_{2.4}$  and  $UO_3$  forms with the higher values of the decay constant. This is expected because exposed fuel in zones 1 and 2 remain at higher temperature for longer time with lower value of the decay constant.

### **Mode of Oxygen Diffusion**

The simulation data for Case 6 is used to discern the affect of mode of oxygen diffusion. The case 6 data is presented in Table 22. As noted in the table, the extent of oxidation for each zone decreases significantly with when oxygen diffuses through fragment surface instead of

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| <b>Table 21. Fuel Oxidation Simulation Data for Case 5 (Low-End Fuel and Cladding Initial Temperature, 0.1 Percent Cladding Failure, Exponential Decomposition of the Residual Water in 71.62 Years, Water Amount 17.4 Moles, and Oxygen Diffusion Through Grain Boundaries). The Values of Decay Constant Is Varied.</b>   |   |   |   |  |
|---|---|---|---|--|
| <b>Zone</b>   | <b>Decay Constant Equal to 0.023</b>  |   | <b>Decay Constant Equal to 0.064</b>  |  |
|   | <b>UO<sub>2+x</sub> Mass [oz]</b>   | <b>Extent of Oxidation*</b>   | <b>UO<sub>2+x</sub> Mass [oz]</b>   | <b>Extent of Oxidation*</b>  |
| 1   | 31.8 g [1.12] of U <sub>3</sub> O <sub>8</sub> and 35.7 g [1.26] of UO <sub>2.4</sub> | 100% to U <sub>3</sub> O <sub>8</sub> , 20% to U <sub>3</sub> O <sub>8</sub> and 80% to UO <sub>2.4</sub> | 0.7 g [0.02] of U <sub>3</sub> O <sub>8</sub> and 34.0 g [1.2] of UO <sub>2.4</sub> and 33.6 g [1.2] of UO <sub>3</sub>   | 3.1% to U <sub>3</sub> O <sub>8</sub> and 96.7% to UO <sub>2.4</sub> , 26% to UO <sub>2.4</sub> and 74% to UO <sub>3</sub>     |
| 2   | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub>  | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub>                             | 0.2 g [0.008] of U <sub>3</sub> O <sub>8</sub> and 27.3 g [0.96] of UO <sub>2.4</sub> and 41.0 g [1.4] of UO <sub>3</sub> | 0.9% to U <sub>3</sub> O <sub>8</sub> and 99.1% to UO <sub>2.4</sub> , 11.5% to UO <sub>2.4</sub> and 88.5% to UO <sub>3</sub> |
| 3   | 67.1 g [2.36] of UO <sub>2.4</sub>  | 100% to UO <sub>2.4</sub> , 100% to UO <sub>2.4</sub>   | 32.2 g [1.1] of UO <sub>2.4</sub> and 36.2 g [1.3] of UO <sub>3</sub>   | 100% to UO <sub>2.4</sub> , 21.8% to UO <sub>2.4</sub> and 78.2% to UO <sub>3</sub>  |
| 4   | 19.4 g [0.68] of UO <sub>2.4</sub>  | 86.7% to UO <sub>2.4</sub> , 0  | 10.6 g [0.37] of UO <sub>2.4</sub> and 58.6 g [2.1] of UO <sub>3</sub>  | 47.4% to U <sub>3</sub> O <sub>8</sub> and 52.6% to UO <sub>2.4</sub> , 100% to UO <sub>3</sub>                                |
| *Extent of Oxidation: This provides information on the percentage of exposed fuel pellets that have undergone oxidation to particular UO <sub>2+x</sub> phase. The extent of oxidation is separately calculated for the directly exposed fuel pellets and fuel pellets in the crack length, plus 6 cm area of a failed rod, but not directly exposed. The extent of oxidation values for the directly exposed fuel pellets and the adjacent fuel pellets, not directly exposed, are separated by a comma. |   |   |   |  |

| <b>Table 22. Fuel Oxidation Simulation Data for Case 6 (Exponential Decomposition of the Residual Water in 4.77 Years, Low-End Fuel and Cladding Initial Temperature, 17.4 Moles of the Residual Water, 0.1 Percent Cladding Failure Rate, and Thermal Decay Constant of 0.023). The Mode of Oxygen Diffusion Is Varied.</b>  |  |   |   |   |
|---|--|---|---|---|
| <b>Zone</b>   | <b>Oxygen Diffusing Through Grain Boundaries</b> |   | <b>Oxygen Diffusing Through Each Fragment Surface</b>                                 |   |
|   | <b>UO<sub>2+x</sub> Mass [oz]</b>                | <b>Extent of Oxidation*</b>   | <b>UO<sub>2+x</sub> Mass [oz]</b>   | <b>Extent of Oxidation*</b>   |
| 1   | 68.3 g [2.40] of U <sub>3</sub> O <sub>8</sub>   | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 3.3 g [0.12] of U <sub>3</sub> O <sub>8</sub>   | 14.5% to U <sub>3</sub> O <sub>8</sub> , 0                              |
| 2   | 68.2 g [2.40] of U <sub>3</sub> O <sub>8</sub>   | 100% to U <sub>3</sub> O <sub>8</sub> , 100% to U <sub>3</sub> O <sub>8</sub> | 1.2 g [0.04] of U <sub>3</sub> O <sub>8</sub> and 0.07 g [0.002] of UO <sub>2.4</sub> | 5.3% to U <sub>3</sub> O <sub>8</sub> and 0.3% to UO <sub>2.4</sub> , 0 |
| 3   | 67.1 g [2.36] of UO <sub>2.4</sub>               | 100% to UO <sub>2.4</sub> , 100% to UO <sub>2.4</sub>                         | 0.4 g [0.013] of UO <sub>2.4</sub>  | 1.8% to UO <sub>2.4</sub> , 0   |
| 4   | 12.1 g [0.43] of UO <sub>2.4</sub>               | 54.1% to UO <sub>2.4</sub> , 0  | 0.08 g [0.003] of UO <sub>2.4</sub>   | 0.4% to UO <sub>2.4</sub> , 0   |
| *Extent of Oxidation: This provides information on the percentage of exposed fuel pellets that have undergone oxidation to a particular UO <sub>2+x</sub> phase. The extent of oxidation is separately calculated for the directly exposed fuel pellets and fuel pellets in the crack length, plus 6 cm area of a failed rod, but not directly exposed. The extent of oxidation values for the directly exposed fuel pellets and the adjacent fuel pellets, not directly exposed, are separated by a comma. |  |   |   |   |

grain boundaries. This is attributed to decrease in surface area available for oxidation per pellet. When oxygen diffuses through grain boundaries and each grain is being oxidized simultaneously, the calculated surface area for fuel oxidation is 0.299 m<sup>2</sup> [3.22 ft<sup>2</sup>] per pellet, whereas when oxygen diffuses through

surface area of each fragment, the effective surface area available for fuel oxidation is  $0.002 \text{ m}^2$  [ $0.022 \text{ ft}^2$ ] per pellet. This decrease in surface area directly affects the extent of oxidation.

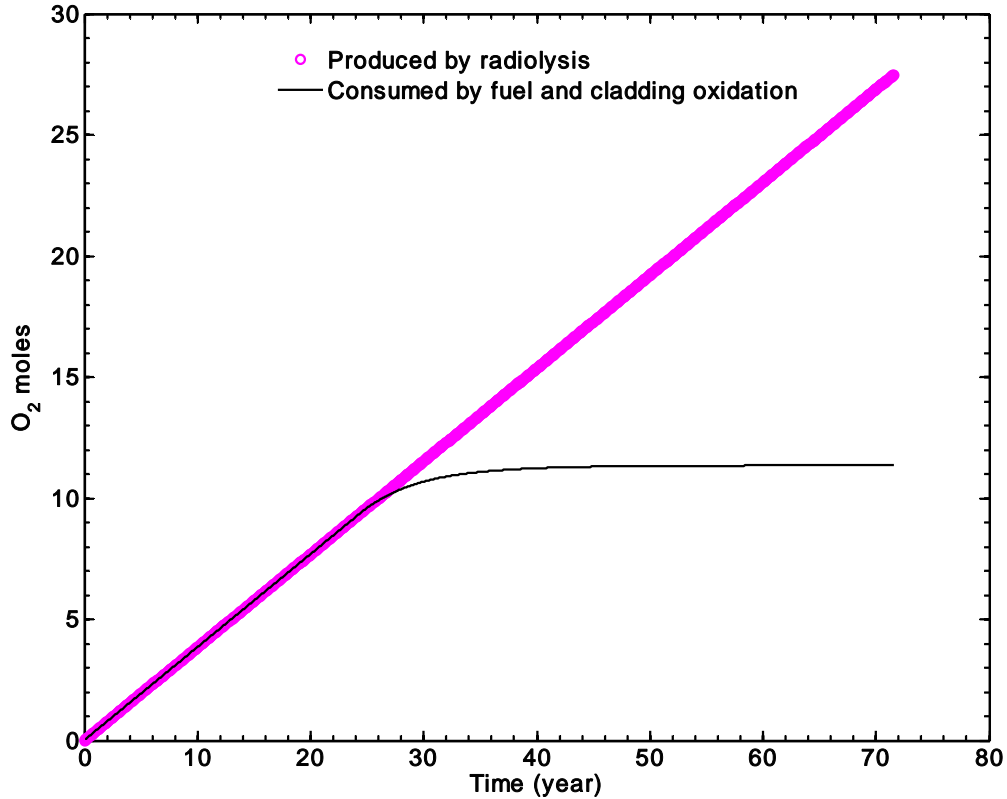
## **Residual Oxygen**

The residual oxygen is defined as radiolysis-generated oxygen but consumed in the fuel and cladding oxidation. The oxygen production and consumption data for a set of case 4 conditions is presented in Figure 5. As seen in the figure, the generated oxygen amount increases linearly with time. The generated oxygen is consumed by the cladding and fuel only for the first 45 years. The oxygen consumption then reaches a plateau. The difference between the generated and consumed oxygen after 71.62 years is the residual oxygen which remains in the canister.

The simulation data for cases 1 to 4 is used to estimate the amount of residual oxygen. The residual oxygen amounts for each case are listed in Table 23. An examination of the case 1 data indicate that the residual oxygen is left in the canister for 55 moles of the residual water under the low-end fuel and cladding initial temperature condition only. The residual oxygen amounts for the case 3 conditions include low-end fuel and cladding initial temperature and 17.4 and 55 moles of the residual water. In case 4, there is residual oxygen for every combination of the condition.

The residual oxygen is left in the cask when fuel and cladding is too cold to react with oxygen. The listed data in Table 22 indicate that the cladding failure rate does not significantly affect the residual moles of oxygen. In fact, the difference between the moles of residual oxygen for

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**Figure 5. Moles of Oxygen Produced and Consumed by Fuel and Cladding Oxidation As a Function of Time for Linear Decay in 71.62 Years, High-End Fuel and Cladding Initial Temperature, 55 Moles of Residual Water, 0.1 percent Cladding Failure Rate, Decay Constant Equal to 0.023, and Oxygen Diffusion Through Grain Boundaries. The Residual Moles of Oxygen Are Determined by Calculating the Difference Between the Produced and Consumed Moles After 71.62 Years.**

two values of cladding failure rate is less than 10 percent. The residual oxygen increases with increasing moles of residual water and generally with the decomposition period.

### Flammability Evaluation

The radiolysis-generated hydrogen is not expected to be absorbed by the cladding because the produced hydrogen would be in molecular form. The residual oxygen could help ignite the hydrogen. According to flammability criterion specified in the Standard Review Plan for Transportation Packages for Radioactive Material in NUREG-1609 (NRC, 1999), volume fraction of any flammable gas should be less than 5 percent.

Since hydrogen is flammable, its mole fraction is calculated as a function of backfill pressure and radiolysis kinetics. Mole fraction is representative of volume fraction in a closed canister of a dry storage cask system. To evaluate flammability of the canister environment, it is assumed that the canister was backfilled with helium at room temperature. Furthermore, it is assumed that the physical properties of helium, hydrogen, and oxygen can be described by the ideal gas law inside the canister environment. The data on mole fraction of hydrogen as a residual water amount and backfill pressure of helium is

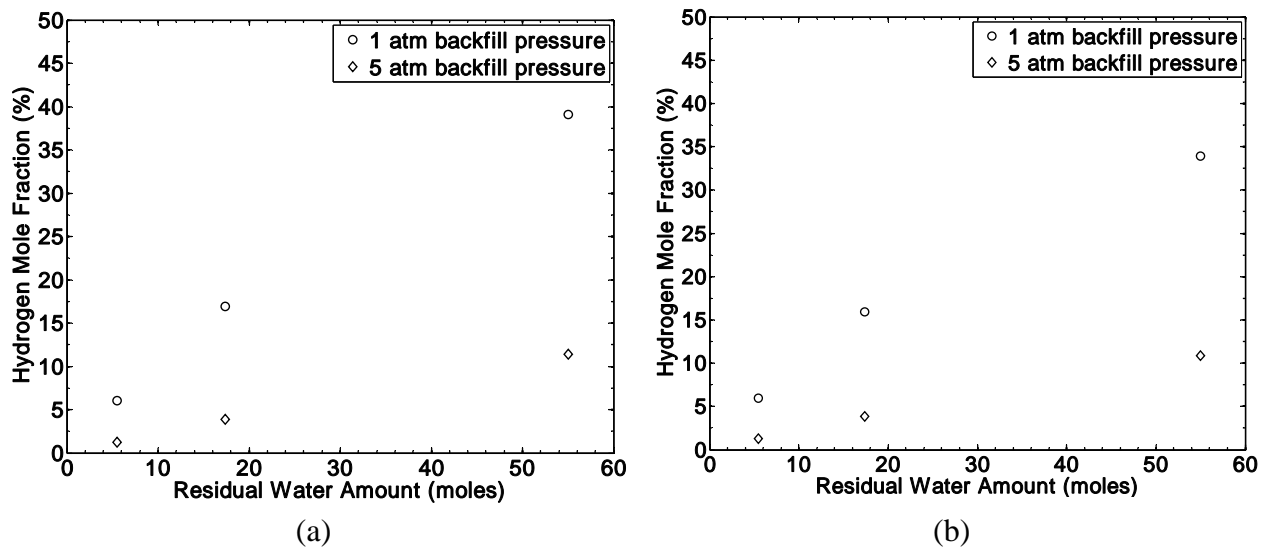
presented in Figure 6(a). The data on mole fraction of hydrogen considering the residual oxygen is presented in Figure 6(b).

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**Table 23. Residual Oxygen For Cases 1, 2, 3, and 4 Conditions**

| Residual Water Amount (moles) | Case 1 (Exponential Decomposition in 4.77 Years) |       |  |       | Case 2 (Linear Decomposition in 4.77 Years)   |       |  |       | Case 3 (Exponential Decomposition in 71.62 Years) |       |  |       | Case 4 (Linear Decomposition in 4.77 Years)   |       |  |       |
|-------------------------------|--|-------|--|-------|---|-------|--|-------|---|-------|--|-------|---|-------|--|-------|
|                               | Low-End Fuel and Cladding Initial Temperature    |       | High-End Fuel and Cladding Initial Temperature |       | Low-End Fuel and Cladding Initial Temperature |       | High-End Fuel and Cladding Initial Temperature |       | Low-End Fuel and Cladding Initial Temperature     |       | High-End Fuel and Cladding Initial Temperature |       | Low-End Fuel and Cladding Initial Temperature |       | High-End Fuel and Cladding Initial Temperature |       |
|                               | Cladding Failure Rate                            |       | Cladding Failure Rate                          |       | Cladding Failure Rate                         |       | Cladding Failure Rate                          |       | Cladding Failure Rate                             |       | Cladding Failure Rate                          |       | Cladding Failure Rate                         |       | Cladding Failure Rate                          |       |
|                               | 0.1%   | 0.01% | 0.1%   | 0.01% | 0.1%  | 0.01% | 0.1%   | 0.01% | 0.1%  | 0.01% | 0.1%   | 0.01% | 0.1%  | 0.01% | 0.1%   | 0.01% |
| <b>5.5</b>                    | 0  | 0     | 0  | 0     | 0   | 0     | 0  | 0     | 0   | 0     | 0  | 0     | 1.7   | 1.7   | 1.1  | 1.1   |
| <b>17.4</b>                   | 0  | 0     | 0  | 0     | 1.2   | 1.1   | 0  | 0     | 0.6   | 0.6   | 0  | 0     | 6.1   | 6.1   | 4.3  | 4.4   |
| <b>55</b>                     | 14.3   | 14.4  | 0  | 0     | 14.3  | 14.4  | 0  | 0     | 14.2  | 14.3  | 0  | 0     | 21.7  | 21.8  | 16.1   | 16.2  |

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**Figure 6. Hydrogen Mole Fraction vs. Moles of Residual Water for 1 and 5 ATM He Backfill Pressure. (a) No Residual Oxygen and (b) Residual Oxygen Data in Table 23 for Linear Decay of Residual Water in 71.62 Years and Low-End Fuel and Cladding Initial Temperature and 0.1 percent Cladding Failure Rate.**

The data presented in Figure 6(a) is calculated assuming that there is no residual oxygen in the cask whereas data in Figure 6(b) is calculated considering residual oxygen values listed in Table 23 for linear decay of residual water in 71.62 years and low-end fuel and cladding temperature. As seen in the figures, the hydrogen mole fraction is more than 5 percent for 1 atm backfill pressure and irrespective of residual oxygen amount. However, the hydrogen mole fraction is more than 5 percent for 5 atm backfill pressure only when residual water is 55 moles. The presence of residual oxygen slightly decreases the hydrogen mole fraction.

The presence of oxygen is necessary for hydrogen to become flammable, however, no criterion exists that specifies ratio of oxygen to hydrogen needed for the flammability of hydrogen with helium as inert gas. Zlochower and Green (2009) conducted experiments to determine limiting oxygen concentration for the flammability of hydrogen in presence of nitrogen as inert gas. The limiting oxygen concentration is defined as the minimum amount of oxygen that can support flame propagation and lead to explosion.

Zlochower and Green (2009) determined that limiting oxygen concentration is close to 5 mole percent for hydrogen mole fraction varying between 4 to 76 mole percent. If it is assumed that the limiting oxygen concentration value determined by Zlochower and Green (2009) applies hydrogen mixed with helium as inert gas, the oxygen concentration exceeds 5 mole percent only when residual oxygen is more than 7.4, 5.4, and 4.8 moles for 55, 17.4, and 5.5 moles of residual water, respectively, and backfill pressure of 1 atm. Similarly, for 5 atm backfill pressure, the oxygen concentration exceeds 5 mole percent when residual oxygen is more than 25.7, 24.4, and 22.8 moles for 55, 17.4, and 5.5 moles of residual water, respectively. Note that these values of oxygen amounts are for 2,100 L canister volume and backfill pressure measured at room temperature. Considering this and data in Table 23, the oxygen concentration could exceed 5 mole percent when 55 moles of the residual water undergo radiolysis irrespective of radiolysis

kinetics. The oxygen concentration could also exceed 5 mole percent when 17.4 moles of the residual water when the water undergoes radiolysis via linear decomposition in 71.62 years and fuel and cladding at the low end.

### **Summary on Fuel and Cladding Oxidation**

The simulation data presented in the section indicate that additional cladding oxidation is insignificant compared to original clad layer thickness from reactor. The maximum  $ZrO_2$  layer that could form is expected to be no more than 7  $\mu\text{m}$  whereas the original cladding layer thickness is expected to be in the range of 0.6 to 0.9 mm (NRC, 1983). Therefore, the formation of  $ZrO_2$  layer due to cladding oxidation is not expected to affect integrity of the cladding; hence, cladding oxidation is not a concern due to the residual water.

The simulation data also indicate that exposed fuel could be completely oxidized to  $U_3O_8$  provided the fuel rod is located in sufficiently hot parts of the canister, amount of residual water is sufficiently high, water undergoes complete radiolysis in a relatively short period, and diffusion of oxygen occurs through grain boundaries. If the amount of residual water is fixed, the key uncertainty in the integration model is the radiolysis kinetics. These uncertainty directly affect the extent of oxidation of the exposed fuel. State cladding rupture case – it would matter only extreme cases of water volume, radiolysis kinetics, and cladding initial failure rate

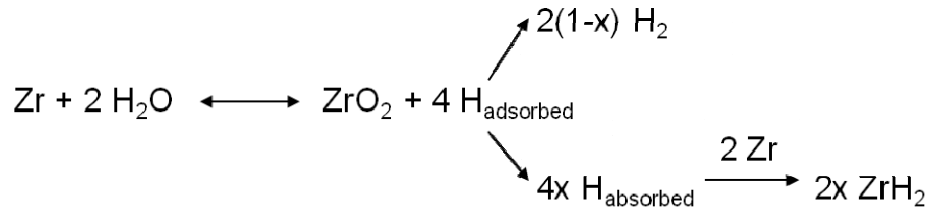
Significant amount of residual oxygen, generated by radiolysis, could be left inside a storage cask when sufficient quantity of the residual water undergoes radiolytic decomposition. The residual oxygen could pose a risk of flammability when both residual hydrogen and oxygen mole fractions exceed 5 percent.

### **Hydrogen Absorption by Cladding**

The cladding material exposed to coolant water during reaction operations would absorb hydrogen during the operating period. The concentration of absorbed hydrogen in the cladding material ranges between 100 to 600 ppm. This process is depicted in form of sequences of chemical reactions in Figure 7. As seen in the figure, water first reacts with the zirconium in the cladding material. This results in release of atomic hydrogen. A fraction of the released atomic hydrogen is absorbed by the cladding, and remaining is released as hydrogen gas. The fraction of atomic hydrogen absorbed is also known as hydrogen uptake fraction, and is denoted by symbol  $x$  in Figure 7. The absorbed hydrogen could react with cladding material and precipitate in form of zirconium hydride when fuel is placed inside a dry storage cask. This could occur because of cooling of the fuel and consequent cooling of the cladding material, and the fact that the solubility of dissolved hydrogen in cladding decreases with decreasing temperature.

The analyses presented in this report indicate that most of the hydrogen would be generated by the radiolysis of the residual water. This radiolysis-generated-hydrogen would be in the molecular form and is not expected to be absorbed by the cladding. The hydrogen could be absorbed by the cladding when water directly reacts with cladding. This would occur when





**Figure 7. Sequence of Chemical Reactions Depicting Zirconium Reaction With Water and Release of Hydrogen Gas, and Absorption of Hydrogen by Zirconium**

water contacting the cladding material is either in liquid phase or relative humidity is above a threshold value provide cladding temperature is sufficiently high. The threshold relative humidity value is assumed to be 20 percent. A justification for is provided in Chapter 4. The relative humidity inside the cask is a function of following three variables (i) fuel and cladding temperature, (ii) water amount, and (iii) rate of radiolysis. RH data as a function of the variables was generated in the integration model, and it is summarized in Table 24.

The information listed in Table 24 indicates that the relative humidity values would briefly exceed more than 20 percent in zone 5 only for 17.4 and 55 moles of water, low-end fuel and cladding initial temperature, and the following three radiolysis models (i) exponential decomposition in 4.77 years, (ii) linear decomposition in 4.77 years, and (iii) exponential decomposition in 71.62 years. For radiolysis kinetic model of linear decomposition in 71.62 years, the RH values in several zones are expected to be more than 20 percent in several zones.

Affect of RH values exceeding more than 20 percent in zones 5 for low-end fuel and cladding initial temperature and for the three kinetic models—(i) exponential decomposition in 4.77 years, (ii) linear decomposition in 4.77 years, and (iii) exponential decomposition in 71.62 years—is discussed first. The cladding surface area that would be exposed in zone 5 is approximately 11.1 m<sup>2</sup> [119.5 ft<sup>2</sup>]. The temperature of the fuel and cladding in zone 5 would vary between 375 and 370 K for the first five years after storage begins, 375 and 355 K for the first 16 years after storage begins (see Figure 2-5 in Chapter 2). Considering this, it can be assumed that the cladding temperature in zone 5 is 375 K for the first 16 years. The amount of water that would react with the exposed cladding in zone 5 at 375 K as a function of time is calculated, and data is presented in Figure 8. As seen in the figure, the maximum water amount that could react with exposed cladding in zone 5 in 16 years after storage begins is less than 6 × 10<sup>-4</sup> moles. Assuming cladding thickness of 0.8 mm and water amount of 6 × 10<sup>-4</sup> moles, the maximum amount of hydrogen that could be absorbed by the cladding would be 6 × 10<sup>-4</sup> moles. This would yield an increase in hydrogen concentration of approximately 24 ppb in the cladding material. This indicates that hydrogen absorption by cladding is not a concern for the three radiolysis models.

The effect of RH values exceeding more than 20 percent in different zones for both low and high-end fuel and cladding initial temperature and for the kinetic model of linear radiolysis in 70 years is discussed. The RH profile for low-end fuel and cladding initial temperature, and 17.4 and 55 moles of residual water is presented in Figures 9(a) and 9(b), respectively. As seen in Figure 9(a), the RH in zone 5 is higher than 20 percent until 62 years after storage begins, and RH in zone 4 exceeds 20 percent after 38 years of storage and remains more than 20 percent for

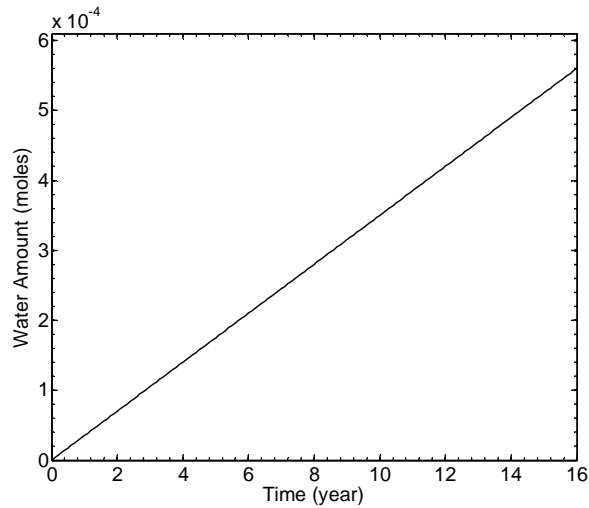
10 years. The extrapolation of information presented in Figure 8 indicates that cladding in zone 5 would at most react with  $2.4 \times 10^{-3}$  moles of water, and corresponding increases in hydrogen concentration of approximately 100 ppb in the cladding material. Moreover, the fuel and cladding temperature in zone 4 after 38 years of storage is below 375 K but above 350 K up to 50 years after storage. The cladding surface area in zone 4 is approximately  $72.5 \text{ m}^2$  [ $780.4 \text{ ft}^2$ ] (see Table 5-6 in Chapter 5). Using the information presented in Figure 8 and considering the cladding the surface are in zone 4, the maximum amount of water that could directly react with the cladding is  $2.5 \times 10^{-3}$  moles. This would result in increases of hydrogen concentration of approximately 15 ppb in the zone 4 cladding material.

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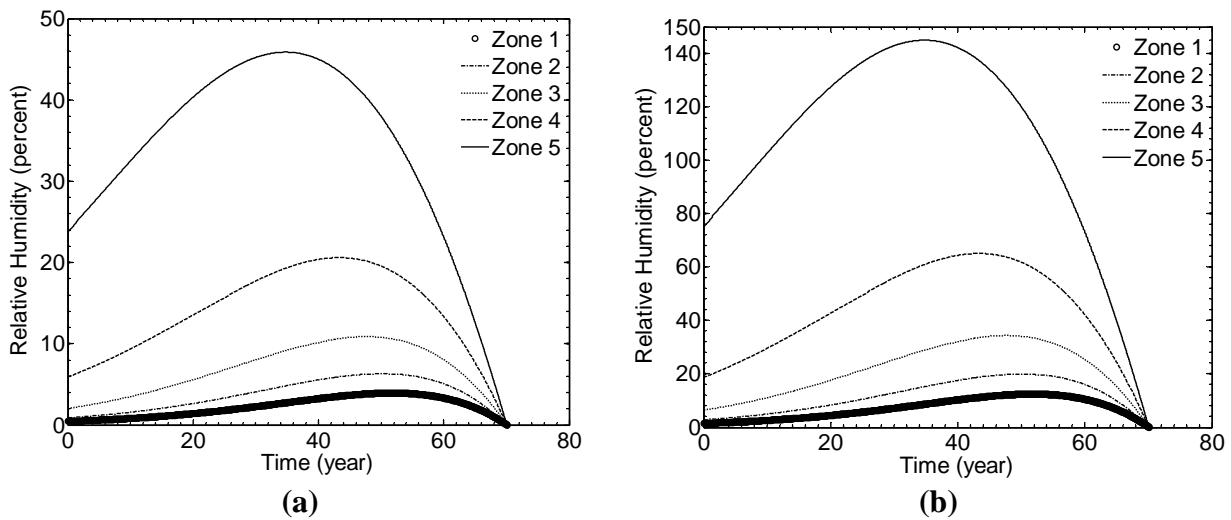
**Table 24. Summary of the Relative Humidity Data Generated in the Integration Model. The Data is Summarized As a Function of Radiolysis Kinetic Model, Low- and High-End Fuel and Cladding Initial Temperature, and Water Amount. The Thermal Decay Constant is 0.023**

| Radiolysis Kinetic Model                 | Low-End Fuel and Cladding Initial Temperature    |   |  | High-End Fuel and Cladding Initial Temperature   |  |  |
|--|--|---|--|--|--|--|
|  | Water Amount (Moles)                             |   |  | Water Amount (Moles)                             |  |  |
|  | 5.5  | 17.4  | 55   | 5.5  | 17.4   | 55   |
| Exponential Decomposition in 4.77 Years  | RH do not exceed more than 20% value in any zone | Maximum RH of 21.5% in zone 5. This values decreases below 20% in less than 1.5 years after storage begins. | Maximum RH of 75% in zone 5. This values decreases below 20% in less than 4 years after storage begins.  | RH do not exceed more than 20% value in any zone | RH do not exceed more than 20% value in any zone | RH do not exceed more than 20% value in any zone                                 |
| Linear Decomposition in 4.77 Years       | RH do not exceed more than 20% value in any zone | Maximum RH of 23.6% in zone 5. This values decreases below 20% in less than 1.5 years after storage begins. | Maximum RH of 75% in zone 5. This values decreases below 20% in less than 4 years after storage begins.  | RH do not exceed more than 20% value in any zone | RH do not exceed more than 20% value in any zone | RH do not exceed more than 20% value in any zone                                 |
| Exponential Decomposition in 71.62 Years | RH do not exceed more than 20% value in any zone | Maximum RH of 23.6% in zone 5. This values decreases below 20% in less than 2 years after storage begins.   | Maximum RH of 75% in zone 5. This values decreases below 20% in less than 16 years after storage begins. | RH do not exceed more than 20% value in any zone | RH do not exceed more than 20% value in any zone | RH do not exceed more than 20% value in any zone                                 |
| Linear Decomposition in 70 Years         | RH do not exceed more than 20% value in any zone | RH remains above 20% in zones 4 and 5. See Figure 9(a) for additional information.                          | RH exceeds above 20% in zones 3, 4, and 5. See Figure 9(b) for additional information.                   | RH do not exceed more than 20% value in any zone | RH do not exceed more than 20% value in any zone | RH exceeds above 20% in zones 4 and 5. See Figure 10 for additional information. |

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**Figure 8. Water Amount in Moles That Would React With Exposed Cladding in Zone 5 As a Function to Time**



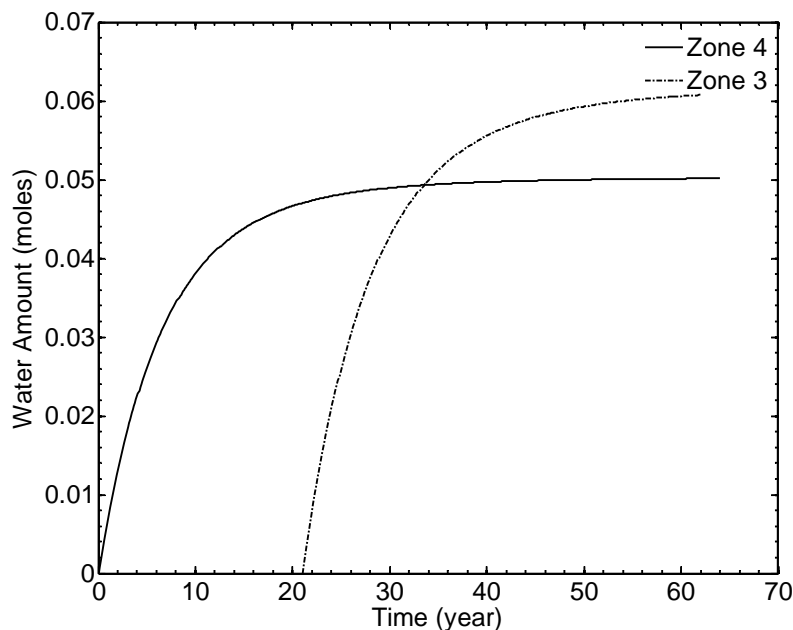
**Figure 9. Relative Humidity As a Function of Time for the Radiolysis Kinetic Model of Linear Decay in 70 Years and Low-End Fuel and Cladding Initial Temperature in Different Zones Inside Canister of the Dry Storage Cask System. (a) 17.4 and (b) 55 Moles of Residual Water.**

As seen in Figure 9(b), the RH in zones 3, 4, and 5 is higher than 20 percent. Specifically, RH in zone 5 is higher than 20 percent until 68 years after storage begins. Moreover, RH in zone 4 exceeds 20 percent for until 66 years after storage begins, and RH in zone 3 exceeds 20 percent after 21 years of storage and remains above 20 percent for 40 years. RH in zone 2 is close to 20 percent but does not exceed 20 percent. The analysis presented for zone 5 due to RH profile in Figure 9(a) can be applied for the zone due to RH profile in Figure 9(a). This indicates that

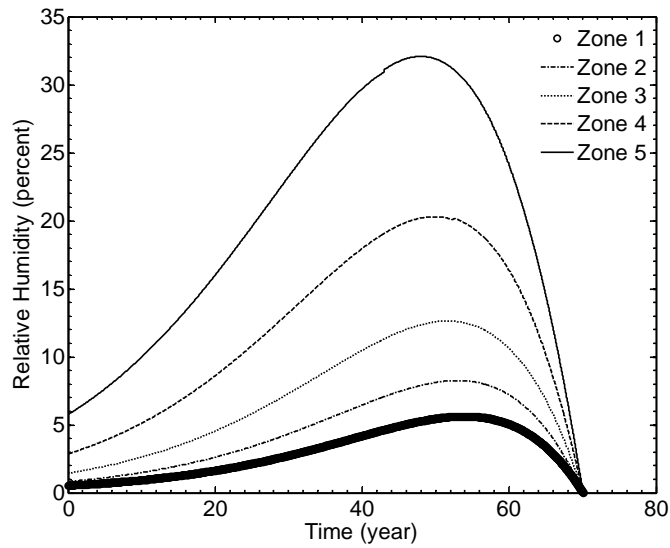
exposed cladding in zone 5 would not absorb more than  $2.4 \times 10^{-3}$  moles of hydrogen, and as a result, the hydrogen concentration would not exceed more than 100 ppb in the zone 5 cladding material. The amount of water that could react with cladding in zones 3 and 4 due to RH and temperature profile is presented in Figure 10. The amount of water is approximately equal to 0.06 and 0.05 moles for zone 3 and 4, respectively. This would result in an increase in hydrogen concentration by 0.13 and 0.3 ppm by zones 3 and 4 cladding, respectively.

The RH profile for high-end fuel and cladding initial temperature and 55 moles of residual water is presented in Figures 11. As seen in the figure, RH in zone 5 exceeds 20 percent after 24 years of storage and remains above 20 percent for approximately 28 years. Moreover, RH in zone 4 exceeds 20 percent after 46 years of storage and remains above 20 percent for approximately 8 years. The cladding temperature in zone 5 when RH exceeds 20 percent is expected to be in the range of 475 to 325 K. Similarly, cladding temperature in zone 4 when RH exceeds 20 percent is expected to be in the range of 390 to 380 K. Considering this, the cladding material in zones 5 and 4 would at most react with  $3.0 \times 10^{-3}$  and  $2.0 \times 10^{-3}$  moles of water, respectively. This would result in an increase in hydrogen concentration by 120 and 12 ppb in zone 5 and 4, respectively.

The analysis presented in this section indicate that amount of hydrogen absorbed by the cladding and resulting increase in dissolved hydrogen concentration in the cladding during extended storage is less compared to preexisting dissolved hydrogen in the cladding during reactor operations. Therefore, absorption of hydrogen by cladding due residual water is not a concern. However, there is concern regarding the zirconium hydride precipitation, and require further evaluation.



**Figure 10. Temperature vs. Water Amount in Moles That Would React With Exposed Cladding in Zones 3 and 4 Due to Relative Humidity Profiles Presented in Figure 9(b)**



**Figure 11. Relative Humidity As a Function of Time for the Radiolysis Kinetic Model of Linear Decomposition in 71.62 Years, High-End Fuel and Cladding Initial Temperature, and 55 Moles of Residual Water in Different Zones Inside Canister of the Dry Storage Cask System**

**Corrosion Calculations**

These calculations were conducted by OLI software. Dr. Roberto Pabalan conducted these calculations.

. Carbon steel; 1 wt% H<sub>2</sub>O<sub>2</sub>; O<sub>2</sub> saturated

| <b>Calculated Rates</b>                         |   |   |   |
|---|---|---|---|
|   | <b>25 °C<br/>(1.21E-03<br/>molal DO2)</b> | <b>75 °C<br/>(4.98E-04<br/>molal DO2)</b> | <b>125 °C<br/>(3 atm;<br/>5.96E-04<br/>molal DO2)</b> |
| <b>Corrosion Rate<br/>(mm/yr)</b>               | 0.602838                                  | 0.624629                                  | 1.30081   |
| <b>Corrosion<br/>Potential (V,<br/>SHE)</b>     | -0.47623                                  | -0.49659                                  | -0.49974  |
| <b>Repassivation<br/>Potential (V,<br/>SHE)</b> | >2.0                                      | >2.0                                      | >2.0  |
| <b>Current<br/>Density (µA/sq-<br/>cm)</b>      | 51.9536                                   | 53.8316                                   | 112.106   |
| <b>Pit Current</b>                              | 4.81E-05                                  | 0.018109                                  | 0.33964   |

|  |  |  |  |
|--|--|--|--|
| Density ( $\mu\text{A}/\text{sq-cm}$ ) |  |  |  |
|--|--|--|--|

2. Carbon steel; 5 wt%  $\text{H}_2\text{O}_2$ ;  $\text{O}_2$  saturated

| Calculated Rates                                      |   |   |  |
|---|---|---|--|
|   | 25 °C<br>(1.17E-03<br>molal $\text{DO}_2$ ) | 75 °C<br>(4.84E-04<br>molal $\text{DO}_2$ ) | 125 °C<br>(3 atm;<br>6.08E-04<br>molal $\text{DO}_2$ ) |
| Corrosion Rate<br>(mm/yr)                             | 0.533749                                    | 0.564001                                    | 1.23685  |
| Corrosion<br>Potential (V,<br>SHE)                    | -0.47756                                    | -0.49789                                    | -0.50043   |
| Repassivation<br>Potential (V,<br>SHE)                | >2.0  | >2.0  | >2.0   |
| Current<br>Density ( $\mu\text{A}/\text{sq-cm}$ )     | 45.9994                                     | 48.6066                                     | 106.594  |
| Pit Current<br>Density ( $\mu\text{A}/\text{sq-cm}$ ) | 3.14E-05                                    | 0.012184                                    | 0.317549   |

3. 304 Stainless steel ; 1 wt%  $\text{H}_2\text{O}_2$ ;  $\text{O}_2$  saturated

| Calculated Rates                                      |   |   |  |
|---|---|---|--|
|   | 25 °C<br>(1.21E-03<br>molal $\text{DO}_2$ ) | 75 °C<br>(4.98E-04<br>molal $\text{DO}_2$ ) | 125 °C<br>(3 atm;<br>5.96E-04<br>molal $\text{DO}_2$ ) |
| Corrosion Rate<br>(mm/yr)                             | 2.54E-03                                    | 6.13E-03                                    | 0.012189   |
| Corrosion<br>Potential (V,<br>SHE)                    | 0.302134                                    | 0.264955                                    | 0.261403   |
| Repassivation<br>Potential (V,<br>SHE)                | >2.0  | >2.0  | >2.0   |
| Current<br>Density ( $\mu\text{A}/\text{sq-cm}$ )     | 0.234567                                    | 0.564822                                    | 1.12387  |
| Pit Current<br>Density ( $\mu\text{A}/\text{sq-cm}$ ) | 0.23174                                     | 0.391842                                    | 0.393305   |

|     |  |  |  |
|-----|--|--|--|
| cm) |  |  |  |
|-----|--|--|--|

4. 304 Stainless steel; 5 wt% H<sub>2</sub>O<sub>2</sub>; O<sub>2</sub> saturated

| <b>Calculated Rates</b>                         |   |   |   |
|---|---|---|---|
|   | <b>25 °C<br/>(1.17E-03<br/>molal DO2)</b> | <b>75 °C<br/>(4.84E-04<br/>molal DO2)</b> | <b>125 °C<br/>(3 atm;<br/>6.08E-04<br/>molal DO2)</b> |
| <b>Corrosion Rate<br/>(mm/yr)</b>               | 2.55E-03                                  | 6.19E-03                                  | 0.012403  |
| <b>Corrosion<br/>Potential (V,<br/>SHE)</b>     | 0.302392                                  | 0.265075                                  | 0.263232  |
| <b>Repassivation<br/>Potential (V,<br/>SHE)</b> | >2.0                                      | >2.0                                      | >2.0  |
| <b>Current<br/>Density (μA/sq-<br/>cm)</b>      | 0.234735                                  | 0.570656                                  | 1.1436  |
| <b>Pit Current<br/>Density (μA/sq-<br/>cm)</b>  | 0.231929                                  | 0.391868                                  | 0.39358   |

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5. Aluminum 1199 (pure); 1 wt% H<sub>2</sub>O<sub>2</sub>; O<sub>2</sub> saturated

| <b>Calculated Rates</b>                         |  |  |  |
|---|--|--|--|
|   | <b>25 °C<br/>(1.21E-03<br/>molal DO<sub>2</sub>)</b> | <b>75 °C<br/>(4.98E-04<br/>molal DO<sub>2</sub>)</b> | <b>125 °C<br/>(3 atm;<br/>5.96E-04<br/>molal DO<sub>2</sub>)</b> |
| <b>Corrosion Rate<br/>(mm/yr)</b>               | 6.06E-04   | 1.50E-03   | 4.87E-03   |
| <b>Corrosion<br/>Potential (V,<br/>SHE)</b>     | -0.18469   | -0.40593   | -0.62806   |
| <b>Repassivation<br/>Potential (V,<br/>SHE)</b> | >2.0   | >2.0   | >2.0   |
| <b>Current<br/>Density (μA/sq-<br/>cm)</b>      | 0.055604   | 0.137239   | 0.446549   |
| <b>Pit Current<br/>Density (μA/sq-<br/>cm)</b>  | 0  | 0  | 0  |

6. Aluminum 1199 (pure); 5 wt% H<sub>2</sub>O<sub>2</sub>; O<sub>2</sub> saturated

| <b>Calculated Rates</b>                         |  |  |  |
|---|--|--|--|
|   | <b>25 °C<br/>(1.17E-03<br/>molal DO<sub>2</sub>)</b> | <b>75 °C<br/>(4.84E-04<br/>molal DO<sub>2</sub>)</b> | <b>125 °C<br/>(3 atm;<br/>6.08E-04<br/>molal DO<sub>2</sub>)</b> |
| <b>Corrosion Rate<br/>(mm/yr)</b>               | 6.95E-04   | 2.13E-03   | 7.27E-03   |
| <b>Corrosion<br/>Potential (V,<br/>SHE)</b>     | -0.17007   | -0.40192   | -0.6257  |
| <b>Repassivation<br/>Potential (V,<br/>SHE)</b> | >2.0   | >2.0   | >2.0   |
| <b>Current<br/>Density (μA/sq-<br/>cm)</b>      | 0.063812   | 0.195218   | 0.666773   |
| <b>Pit Current<br/>Density (μA/sq-<br/>cm)</b>  | 0  | 0  | 0  |

7. Alloy 600; 1 wt% H<sub>2</sub>O<sub>2</sub>; O<sub>2</sub> saturated

| <b>Calculated Rates</b>                         |   |   |   |
|---|---|---|---|
|   | <b>25 °C<br/>(1.21E-03<br/>molal DO2)</b> | <b>75 °C<br/>(4.98E-04<br/>molal DO2)</b> | <b>125 °C<br/>(3 atm;<br/>5.96E-04<br/>molal DO2)</b> |
| <b>Corrosion Rate<br/>(mm/yr)</b>               | 1.34E-04                                  | 4.80E-04                                  | 1.25E-03  |
| <b>Corrosion<br/>Potential (V,<br/>SHE)</b>     | 0.338247                                  | 0.316179                                  | 0.322177  |
| <b>Repassivation<br/>Potential (V,<br/>SHE)</b> | >2.0                                      | >2.0                                      | >2.0  |
| <b>Current<br/>Density (µA/sq-<br/>cm)</b>      | 0.012819                                  | 0.045878                                  | 0.119369  |
| <b>Pit Current<br/>Density (µA/sq-<br/>cm)</b>  | 6.93E-03                                  | 1.59E-03                                  | 6.94E-04  |

8. Alloy 600; 5 wt% H<sub>2</sub>O<sub>2</sub>; O<sub>2</sub> saturated

| <b>Calculated Rates</b>                         |   |   |   |
|---|---|---|---|
|   | <b>25 °C<br/>(1.17E-03<br/>molal DO2)</b> | <b>75 °C<br/>(4.84E-04<br/>molal DO2)</b> | <b>125 °C<br/>(3 atm;<br/>6.08E-04<br/>molal DO2)</b> |
| <b>Corrosion Rate<br/>(mm/yr)</b>               | 1.34E-04                                  | 9.62E-04                                  | 2.56E-03  |
| <b>Corrosion<br/>Potential (V,<br/>SHE)</b>     | 0.356845                                  | 0.297778                                  | 0.300758  |
| <b>Repassivation<br/>Potential (V,<br/>SHE)</b> | >2.0                                      | >2.0                                      | >2.0  |
| <b>Current<br/>Density (µA/sq-<br/>cm)</b>      | 0.012837                                  | 0.091882                                  | 0.244619  |
| <b>Pit Current<br/>Density (µA/sq-<br/>cm)</b>  | 8.90E-03                                  | 9.33E-04                                  | 3.96E-04  |

## Radiolysis Calculations

### Selecting G-value(s)

The selected G(H<sub>2</sub>O)-value is -7.4 molecules /100 eV, based upon the general values reported in Arkhipov, et al. (2007). Given this is a rough calculation, the value selected and will not greatly affect the value which is intended to be order of magnitude (the value could be scaled to reflect other assumed G-values).

Note, G-value is only considering value for Gamma. Note, for neutrons, the flux would be significantly lower than in core. Using only the G-value for Gamma is bounding since reported G-values for contribution from the fast neutrons are lower than the gamma. Therefore, here we assume all deposited radiation decomposes water based upon the g-value for gamma. Subsequent recombinations of decomposition products to water are being neglected.

### Radiation

Radulescu (2011) includes a map of the dose rate values for a cross-section through the center of the The source term used by Radulescu (2011) for this model includes neutron and gamma radiation and the model calculations were made using MAVRIC (Peplow, 2011) for dose rate values. Based upon this figure, the values of the dose rates depicted by the response value bin, which covers the majority of the canister volume, range from  $1.18 \times 10^2$  to  $1.69 \times 10^3$  Gy/hr [ $1.18 \times 10^4$  to  $1.69 \times 10^5$  rad/hr]. In order to convert from dose rate in rad/hr to units of eV/g/s for moist air, the relationship  $1 \text{ rad/h} = 1.7338 \times 10^{10} \text{ eV/g/s}$  is used. This is assumed reasonable in the present work; as stated in Radulescu (2011), the conversion to energy deposition rate from the dose rates are assumed applicable to moist air of varying density because the ANSI/ANS-6.1.1-1977 flux-to-dose were used in the MAVRIC dose rate calculations, which have been shown in CAL-MGR-NU-000006, Revision 00 (BSC, 2001) to lead to slightly higher energy absorption for moist air. Note that the values selected from Radulescu (2011) are at 20 years decay. Based upon this conversion, the values in units of eV/g/s are  $2.04 \times 10^{14}$  to  $2.93 \times 10^{15}$  in units of eV/g/s. For the calculation of decomposition rate, the values are rounded to  $2 \times 10^{14}$  to  $3 \times 10^{15}$ .

### Time for Water Decomposition

The time for complete decomposition of residual water is estimated using the selected G-value, amount of water, and the energy deposition. The amount of water present as water vapor was assumed to be 55 moles. The time to decompose 55 moles of water in the cask internal volume was estimated. This is done using several additional assumptions. First, the rate of energy deposition is assumed constant for all water vapor (in reality this would vary depending on location in the cask) and for all times considered (this value would be expected to decrease in time as the total radiation level drops). Further, subsequent recombination of decomposition products to water are not accounted for. Together these assumptions result in estimating a time needed for complete decomposition of the water that would be less than would be expected. Using the assumptions, the decomposition rate for water ( $R_D$ ) can be calculated via the relation

$$R_D = R_{ED} m_{water} G_{water}$$

where  $R_{ED}$  is the rate of energy deposition in units of eV/g/second,  $m_{water}$  is the mass of water in grams,  $G_{water}$  is the G-value in units of molecules of H<sub>2</sub>O decomposed per 100 eV, and the decomposition rate of water,  $R_D$ , is in units of molecules decomposed per second. Assuming 55 moles is approximately 1000 grams of water. The two values calculated for the water decomposition rate are:

$$R_D = R_{ED} \left( \frac{\text{eV}}{\text{g} \times \text{s}} \right) \times 1000 \text{ (g)} \times \frac{7.4 \text{ (Molecules)}}{100 \text{ (eV)}}$$

Therefore, for both energy deposition rates considered:

$$R_D = 3 \times 10^{15} \left( \frac{\text{eV}}{\text{g} \times \text{s}} \right) \times 1000 \text{ (g)} \times \frac{7.4 \text{ (Molecules)}}{100 \text{ (eV)}} = 2.22 \times 10^{17} \left( \frac{\text{Molecules}}{\text{s}} \right)$$

$$R_D = 2 \times 10^{14} \left( \frac{\text{eV}}{\text{g} \times \text{s}} \right) \times 1000 \text{ (g)} \times \frac{7.4 \text{ (Molecules)}}{100 \text{ (eV)}} = 1.48 \times 10^{16} \left( \frac{\text{Molecules}}{\text{s}} \right)$$

Based upon this the time to consume the mass of water is:

$$\text{Time (year)} = 1000 \text{ (gram)} \times 1/18.016 \text{ (mole/gram)} \times 6.02214 \times 10^{23} \text{ (molecules/mole)} \times \left( \frac{1}{R_D \left( \frac{\text{Molecules}}{\text{s}} \right)} \right) \times \left( \frac{1 \text{ hr}}{3600 \text{ s}} \right) \times \left( \frac{1 \text{ day}}{24 \text{ hr}} \right) \times \left( \frac{1 \text{ year}}{365 \text{ day}} \right).$$

The calculated times are 4.77 years and 71.62 years, respectively, for  $3 \times 10^{15} \left( \frac{\text{eV}}{\text{g} \times \text{s}} \right)$  and  $2 \times 10^{14} \left( \frac{\text{eV}}{\text{g} \times \text{s}} \right)$ .

## Model Expressions for Chemical Kinetics

Detailed chemical kinetics of the water decomposition reactions due to radiolysis in the canister is not modeled. Instead, bounding approaches were used to estimate the amount of chemical species of interest. Expressions are developed in the following to approximate the chemical kinetics as radiolysis products accumulate and react with cask system internals (e.g., cladding and fuel oxidation). The expressions represent the quantity of water in the cask internal volume as a function of time based upon two cases for the chemical kinetics. For the first case, the rate of decomposition of water is assumed independent of chemical species concentrations and subsequent consumption of those species by any chemical reaction. A linear rate of decomposition was assumed based upon the two periods estimated for complete radiolytic decomposition of 55 moles of residual water. In the second case, an exponential rate is assumed for the decomposition of the residual water to approximate contributions from chemical reactions that would consume radiolysis products of water. The underlying assumptions and technical bases for the two cases are explained next.

For the first case, the reduction in the quantity of water in the canister is assumed to follow the rate of decomposition from radiolysis, which is assumed to be constant. In other words, the rate of energy deposition was assumed constant and recombination of reaction products to water is not accounted for. Further, the decomposition rate of water is assumed independent of chemical species concentrations and consumption of those species by any chemical reaction that may occur in the canister environment. For this case, the expressions can be readily developed. For the two periods, 4.77 and 71.62 years, the expressions are given by the following equations:

$$n_{water}(t) = n_{water}(1 - t / 4.77)$$

and

$$n_{water}(t) = n_{water}(1 - t / 71.62),$$

respectively, where  $n_{water}(t)$  is the number of moles of water in the canister time  $t$ ,  $n_{water}$  is the initial number of moles of residual water in the canister, and  $t$  is the time in years.

For the second case, instead of a linear function, a time-dependent exponential function is used to calculate the residual water amount in the canister. This is done in order to reflect consumption of radiolysis-produced chemical species. The exponential function qualitatively accounts for consumption of radiolysis-generated species. Two expressions are developed for the two time frames estimated for decomposition of 55 moles of water from radiolysis. The expressions for the exponential term are found by assuming that at the two time periods, 4.77 and 71.62 years, only 0.01 % of the water is present (near zero). At time zero, the quantity of water is 55 moles and for the two time periods, the quantity is 0.55 moles. A trend of the form

$$n_{water}(t) = 55 \times \exp(a t)$$

is fit to the data for each time period. Using the FindFit function in Mathematica®, the constants were determined to be  $-1.92904$  and  $-0.128609$ .

The expressions developed to represent the amount of residual water as a function of time for the two time frames, 4.77 and 71.62 years, are given by the following equations

$$n_{water}(t) = n_{water} \exp(-1.92904 t)$$

$$n_{water}(t) = n_{water} \exp(-0.128609 t),$$

These expressions are also used when  $n_{water}$  is less than 55 moles.

### Additional Details of the Radiolysis Model

**Date: May 1, 2012**

**Entry by: Pavan Shukla**

#### Radiolysis Model

**Radiolysis model was updated. Details follow.**

It is assumed that the water decomposes to produce oxygen and water according to the following chemical equation



The decomposition rate for water  $R_D$  in unit of molecules per second was calculated using the following equation

$$R_D = R_{ED} m_{water} G_{water} \quad (2)$$

where  $R_{ED}$  is the rate of energy deposition in units of eV/g/second,  $m_{water}$  is the mass of water in grams,  $G_{water}$  is the G-value in units of molecules of  $\text{H}_2\text{O}$  decomposed per 100 eV. Following the law of mass action,  $m_{water}$  can be expressed as

$$m_{water}(t) = m_0 \exp\left(-\frac{R_{ED} G_w MW}{N_A} t\right) = m_0 \exp\left(-\frac{t}{\tau}\right) \quad (3)$$

where  $t$  is time,  $m_0$  is the initial amount of the residual water,  $MW$  is the molar mass of water, and  $N_A$  is Avogadro's number, and  $\tau$  is a time constant defined as

$$\tau = \frac{N_A}{R_{ED} G_W MW} \quad (4)$$

Eq. (3) can be rewritten as

$$n_{water}(t) = n_0 \exp\left(-\frac{t}{\tau}\right) \quad (5)$$

where  $n_{water}$  is the moles of water, and  $n_0$  is the initial moles of the residual water. Using the higher and lower values of radiation energy deposition rates, the values of the time constants are equal to 4.77 and 71.62 years for the radiation energy deposition rates equal to  $2.0 \times 10^{14}$  and  $3 \times 10^{15}$  eV/g/s, respectively. Additional analysis presented in the following subsection indicates that recombination could change the rate water decomposition. For the sake of the scoping analyses, it is assumed that 99.99 percent of the water decomposes in one time constant, with the remaining water computed as

$$n_{water}(t) = n_0 \exp(-1.929 t) \quad (6)$$

for time constant equal to 4.77 years, and

$$n_{water}(t) = n_0 \exp(-0.129 t) \quad (7)$$

for the time constant equal to 71.62 years. The following two equations were also used where water is assumed to decompose completely within a time constant following a linear function of time

$$n_{water}(t) = n_0(1 - t / 4.77) \quad (8)$$

and

$$n_{water}(t) = n_0(1 - t / 71.62) \quad (9)$$

These equations are reasonable given the uncertainty in the parameters, the wide range of radiation dose considered, and the scoping nature of this assessment. Eqs. (6) to (9) are used to analyze the effects of residual water on various canister components.

### Effect of Recombination Reaction

In the paper by Arkhipov, et al. (2007), more than 20 chemical equations are listed to model radiolytical decomposition of the vapor-phase water. Several of those reactions involve reaction between hydrogen and oxidizing species to produce water. This indicates that recombination reaction between hydrogen and oxygen could occur via a complex reaction pathways leading to the production of water in the canister. Recombination could change water depletion rate

embedded in Eq. (A2–5). The discussion that follows is a simplified analysis of potential changes in the depletion rate arising from recombination.

Considering this, Eq. (2), and stoichiometry of the Eq. (1), the following equation represents mass-balance for water in the canister

$$\frac{d[H_2O]}{dt} = -\frac{[H_2O]}{\tau} + k[H_2]^2[O_2] \quad (10)$$

where

|          |   |   |
|----------|---|---|
| $[H_2O]$ | — | water concentration in the canister = $n_{water}(t)/V$                    |
| $[H_2]$  | — | hydrogen concentration in the canister = $n_{hydrogen}(t)/V$              |
| $[O_2]$  | — | oxygen concentration in the canister = $n_{oxygen}(t)/V$                  |
| $k$      | — | rate constant for the recombination reaction in units of $M^{-2}sec^{-1}$ |
| $V$      | — | canister volume   |

and the symbols  $n_{hydrogen}(t)$  and  $n_{oxygen}(t)$  denote molar amounts of hydrogen and oxygen as functions of time. Based on stoichiometry of Eq. (A2–1), the following equations are valid in the canister environment

$$[O_2] = \frac{[H_2]}{2} \quad (11)$$

and

$$[H_2] = [H_2O]_0 - [H_2O] \quad (12)$$

where  $[H_2O]_0$  is initial water concentration. Equations (A2–11) and (A2–12) mean that the only source for  $H_2$  and  $O_2$  is water decomposition, and that reactions with uptake  $O_2$  and  $H_2$  are ignored, such as oxidation of fuel and cladding. Equation (A2-10) can be rewritten as

$$\frac{d[H_2O]}{dt} = -\frac{[H_2O]}{\tau} + \frac{k}{2} ([H_2O]_0^3 - 3[H_2O]_0^2[H_2O] + 3[H_2O]_0[H_2O]^2 - [H_2O]^3) \quad (13)$$

In the canister environment,  $[H_2O]_0$  is equal to 0.026 M for 55 moles of residual water and 2,100 L void volume; therefore, in general the concentration  $[H_2O]$  is small. A limit analysis Eq. (13) is provided to compare the rate of water depletion with respect to Eq. (5). First, a case of slow chemical kinetics is considered.

### Slow Recombination Reaction (i.e., small $k$ )

Because  $[H_2O]$  is small, the first order term with respect to  $[H_2O]$  would dominate in Eq. (13) for the canister environment for small  $k$  values. Therefore, neglecting second and third order terms with respect to  $[H_2O]$  the following approximation is valid

$$\frac{d[H_2O]}{dt} = -[H_2O] \left( \frac{1}{\tau} + \frac{3k[H_2O]_0^2}{2} \right) + \frac{k}{2} [H_2O]_0^3 = -\frac{[H_2O]}{\tau_{eff}} \quad (14)$$

where effective time constant  $\tau_{eff}$  considering the recombination reaction, in the limit of small  $k$ , is

$$\tau_{eff} = \frac{2\tau}{2 + 3k[H_2O]_0^2\tau} \quad (15)$$

Equation (15) indicates that  $\tau_{eff} < \tau$ , or that recombination would expedite water depletion in the limit of small  $k$ , with respect to the depletion rate implied in Eq. (5). Therefore, assumption of 99.99 percent of the residual water within one time constant is reasonable.

### Fast Recombination Reaction (i.e., large $k$ )

For large  $k$ , Eq. (A2–13) can be approximated as

$$\frac{d[H_2O]}{dt} = \frac{k}{2} ([H_2O]_0^3 - 3[H_2O]_0^2[H_2O] + 3[H_2O]_0[H_2O]^2 - [H_2O]^3) \quad (16)$$

At steady state, this equation has the trivial solution

$$[H_2O](t) = [H_2O]_0 \quad (17)$$

This solution is intuitive: in the limit of very fast chemical kinetics, any water decomposed by radiolysis is immediately reconstituted by the recombination reaction, leaving the amount of water unchanged. Thus, based on the limiting Eq. (17), as  $k$  increases, the rate of water depletion is slowed down compared to the depletion rate embedded in Eq. (5).

Based on the limiting analysis for small and large  $k$ , recombination can accelerate or slow down water depletion compared to the depletion rate in Eq. (5). To account, in a simplistic manner, for radiolysis and chemical kinetics, perturbations to Eq. (5) were examined, in the form of Eqs. (6) to (9).

### Effect of Oxygen Consumption

Radiolysis-generated oxygen can react with cladding and exposed fuel. Interaction of radiolysis-driven chemical reactions, such as  $2H_2O \rightarrow 2H_2 + O_2$ , with metal oxidation reactions can affect the rate of recombination reactions [e.g., the balance assumed in Eq. (A2–11) would be altered if metal oxidation is considered]. Thus, the overall rate of water decomposition (in which radiolysis and chemical kinetics play a role) can be a function of oxidation rates of metallic system components.

### Radilysis References

Arkhipov, O.P., A.O. Verkhovskaya, S.A. Kabakchi, and A.N. Ermakov. “Development and Verification of a Mathematical Model of the Radiolysis of Water Vapor.” *Atomic Energy*. Vol. 103, No. 5. pp. 870–874. 2007.



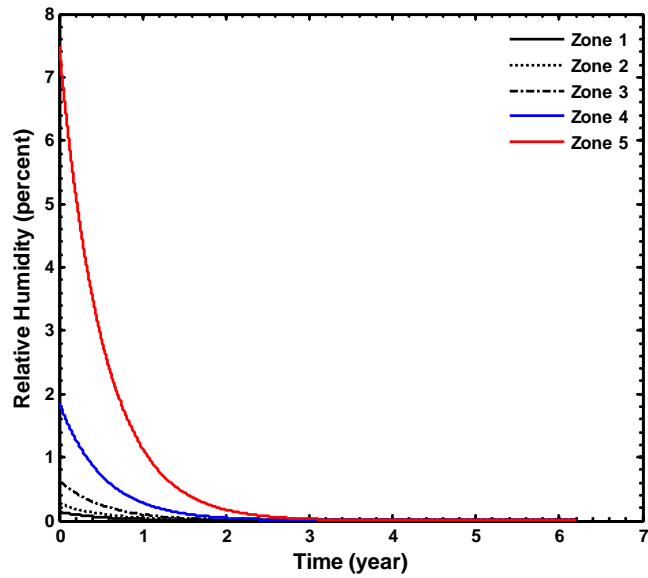
BSC. "Gamma and Neutron Radiolysis in the 21-PWR Waste Package." CAL-MGR-NU 000006, Rev. 00. Las Vegas, Nevada: Bechtel SAIC LLC. 2001b. Peplow, D.E. "Monte Carlo Shielding Analysis Capability With MAVRIC." *Nuclear Technology*. Vol. 174. p. 289. 2011.

Radulescu G. "Radiation Transport Evaluations for Repository Science." ORNL/LTR 2011/294. Oak Ridge, Tennessee: Oak Ridge National Laboratory. 2011.

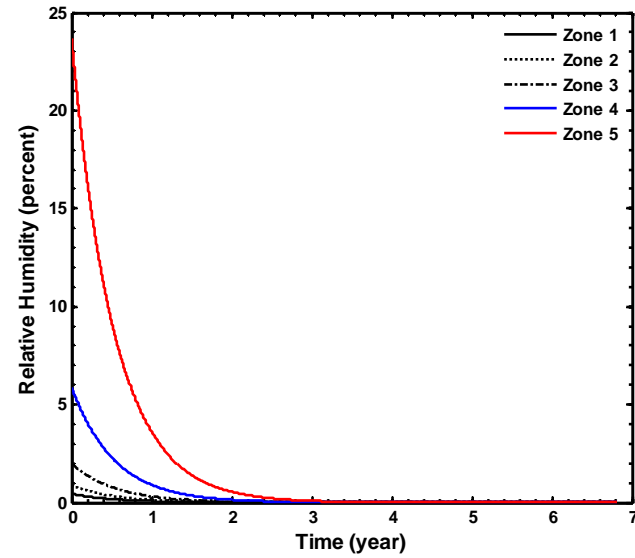
### **Relative Humidity Data for Table 23**

Relative humidity data generated using the integration models is for cases 1 to 4 is presented in Figures A5-7 to A5-14.

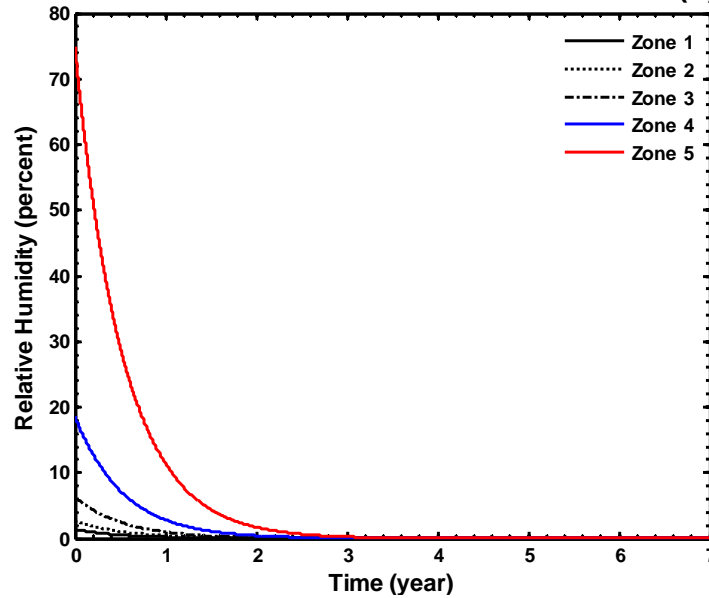
**(The remainder of this page is left blank)**



(a) 5.5 moles

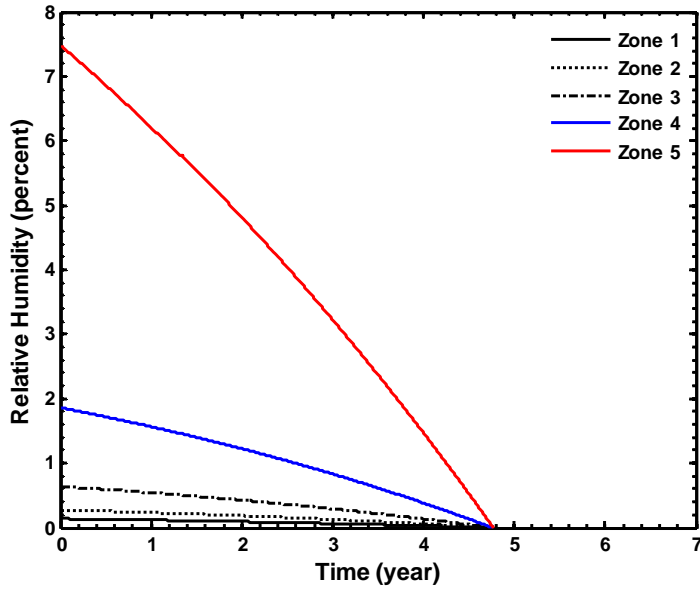


(b) 17.4 moles

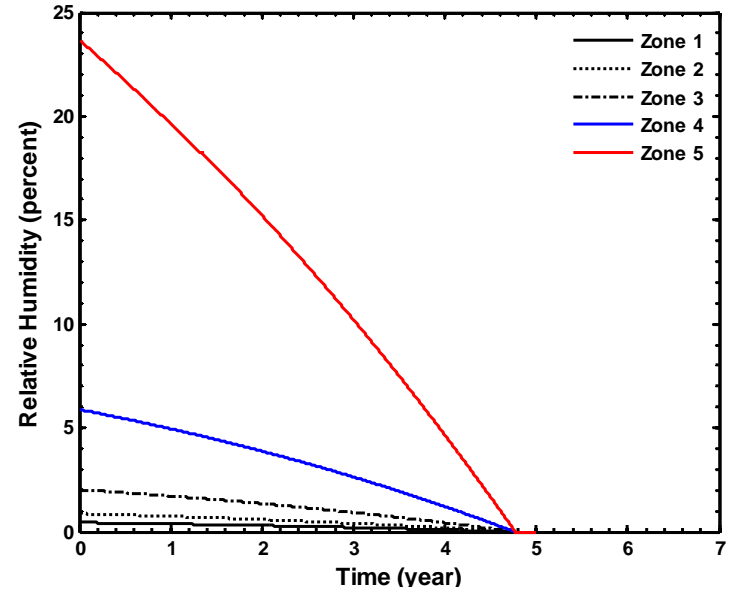


(c) 55 moles

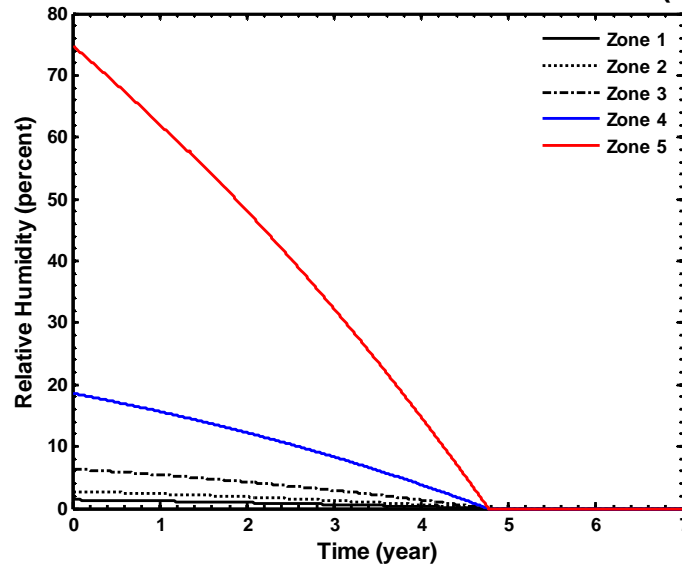
Figure A5-7. Relative Humidity Profiles for Exponential Decomposition in 4.77 years, Low-End Temperature Condition, and (a) 5.5, (b) 17.4, and (c) 55 moles of Residual Water



(a) 5.5 moles

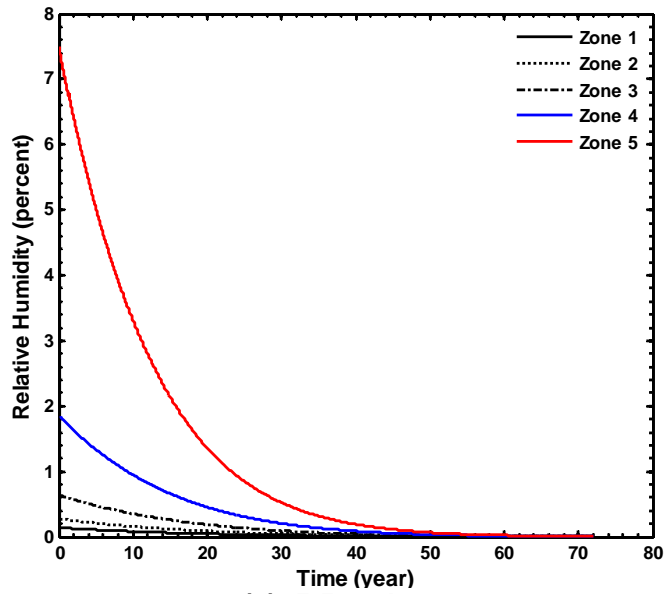


(b) 17.4 moles

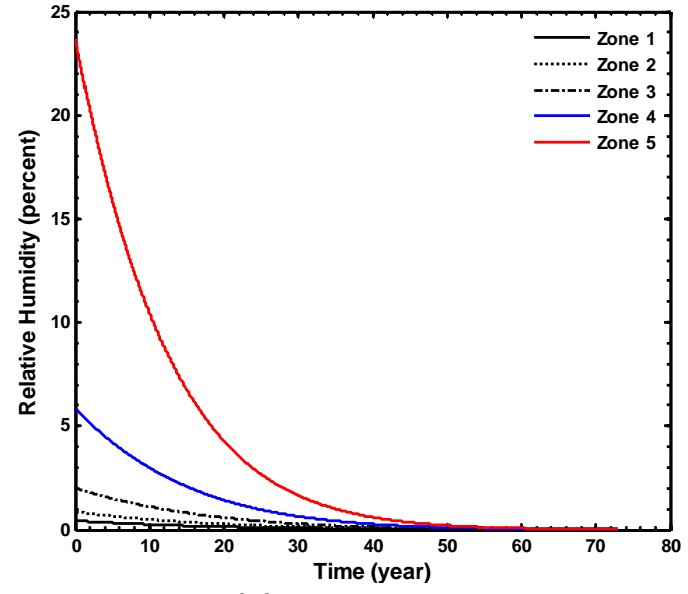


(c) 55 moles

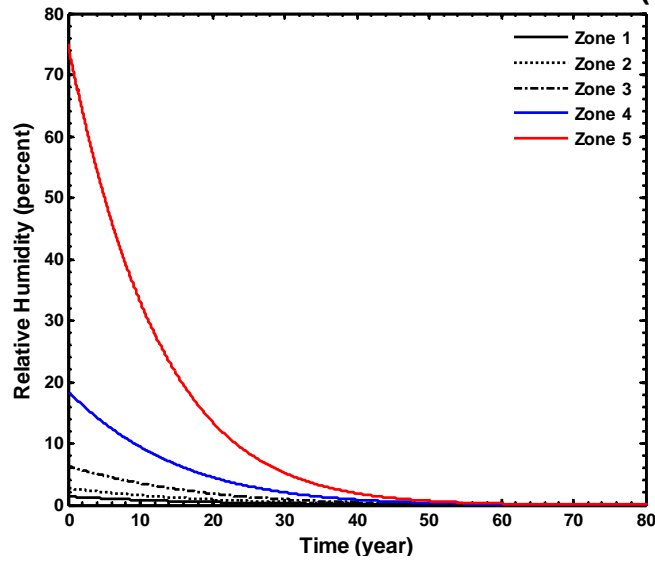
Figure A5-8. Relative Humidity Profiles for Linear Decomposition in 4.77 years, Low-End Temperature Condition, and (a) 5.5, (b) 17.4, and (c) 55 moles of Residual Water



(a) 5.5 moles

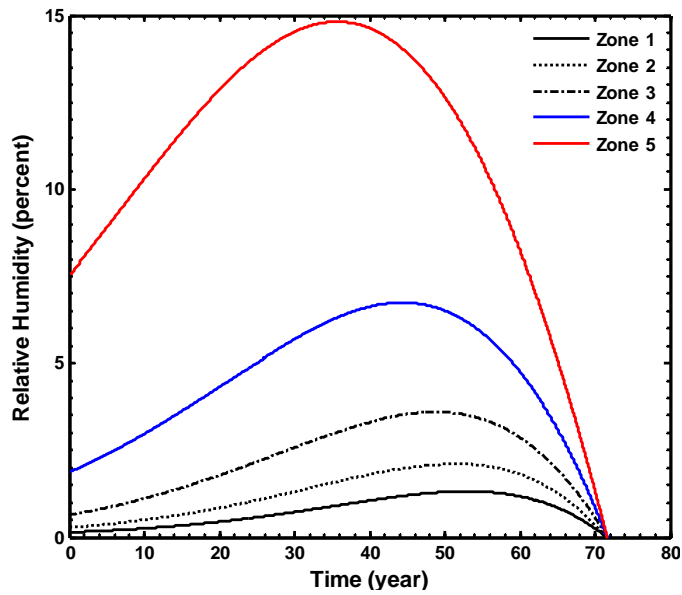


(b) 17.4 moles

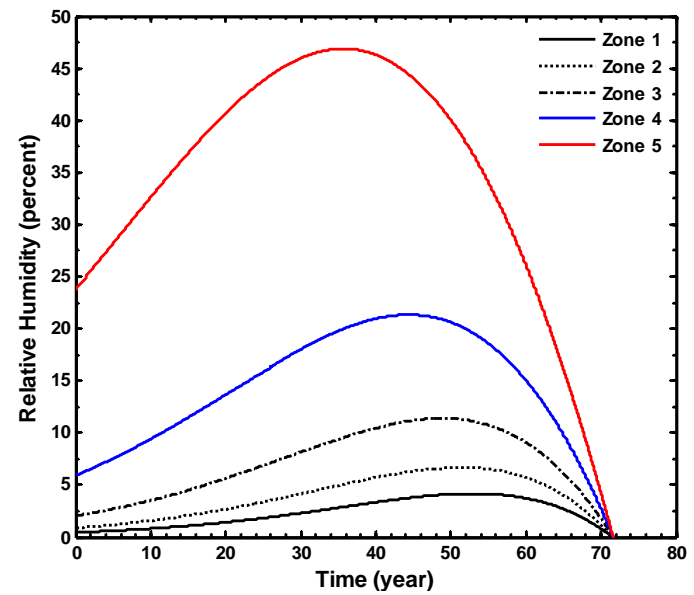


(c) 55 moles

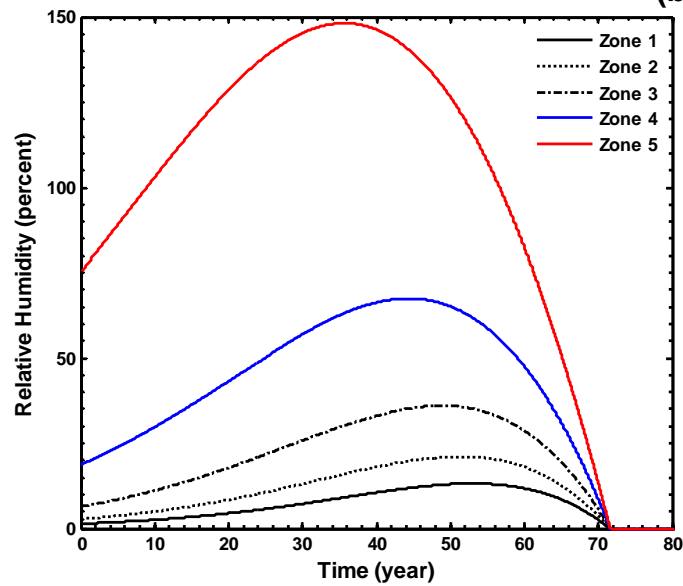
Figure A5-9. Relative Humidity Profiles for Exponential Decomposition in 71.62 years, Low-End Temperature Condition, and (a) 5.5, (b) 17.4, and (c) 55 moles of Residual Water



(a) 5.5 moles

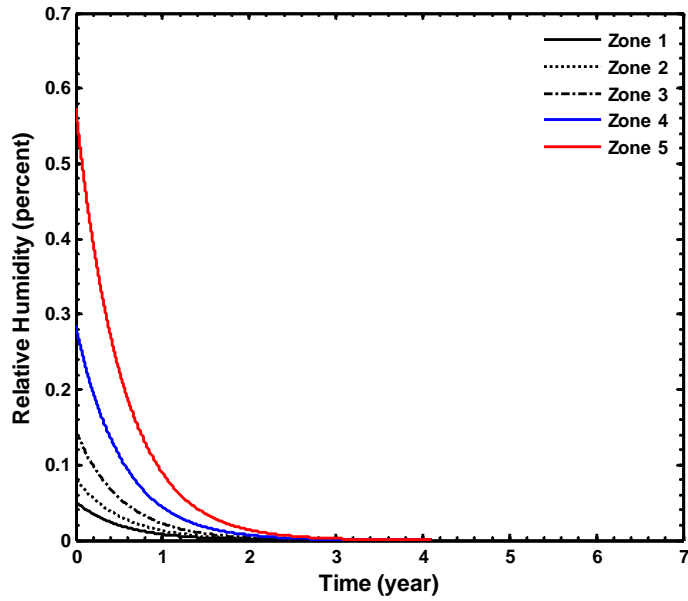


(b) 17.4 moles

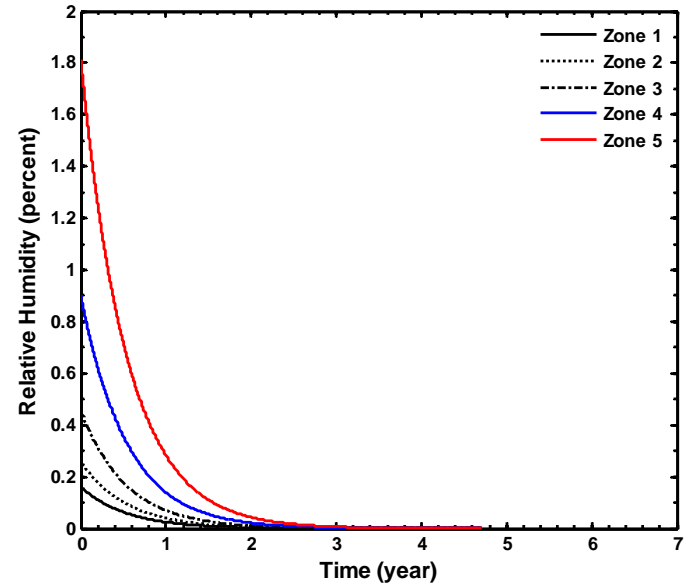


(c) 55 moles

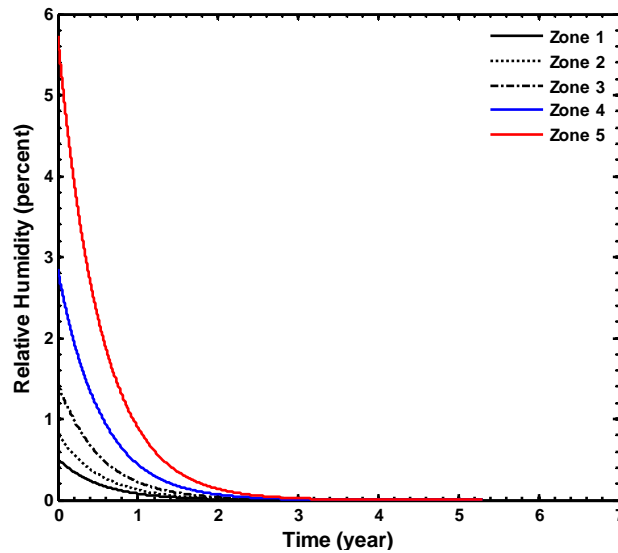
Figure A5-10. Relative Humidity Profiles for Linear Decomposition in 71.62 years, Low-End Temperature Condition, and (a) 5.5, (b) 17.4, and (c) 55 moles of Residual Water



(a) 5.5 moles

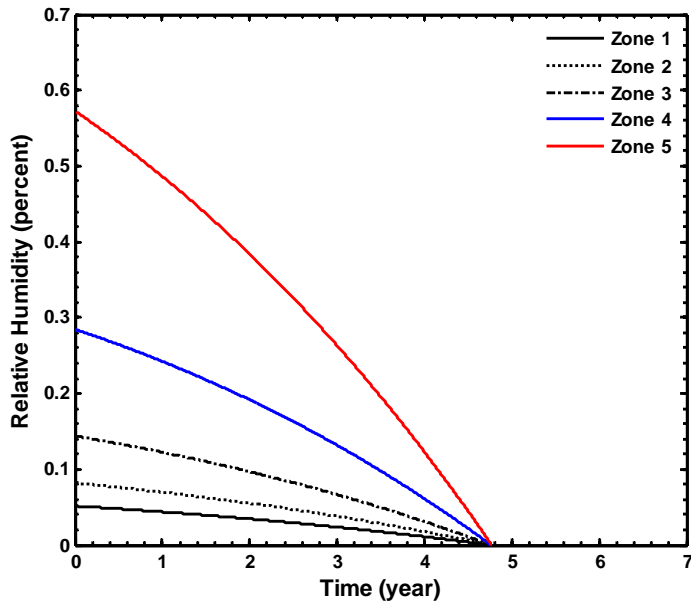


(b) 17.4 moles

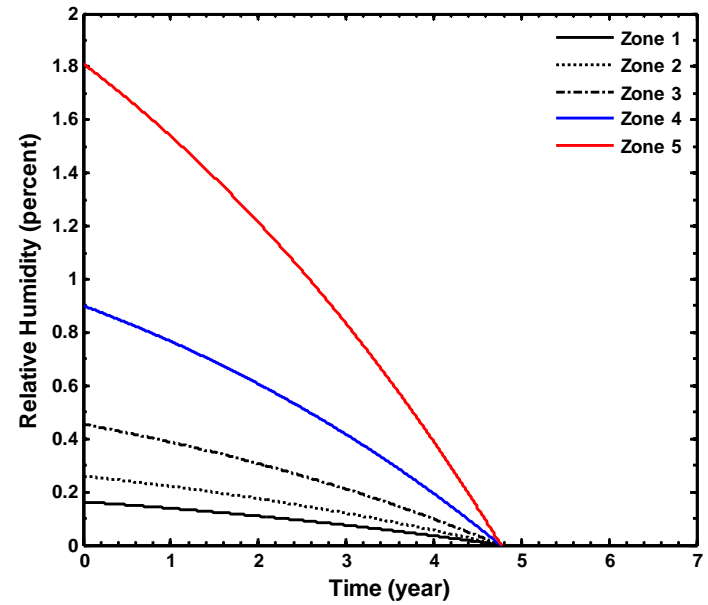


(c) 55 moles

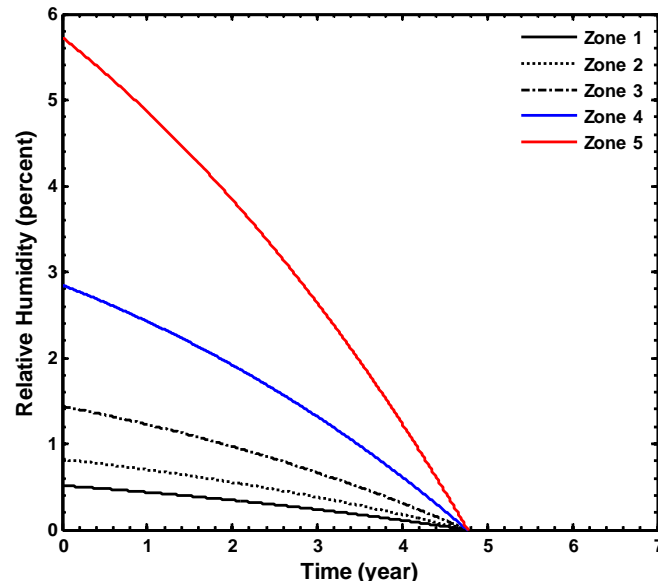
Figure A5-11. Relative Humidity Profiles for Exponential Decomposition in 4.77 years, High-End Temperature Condition, and (a) 5.5, (b) 17.4, and (c) 55 moles of Residual Water



(a) 5.5 moles

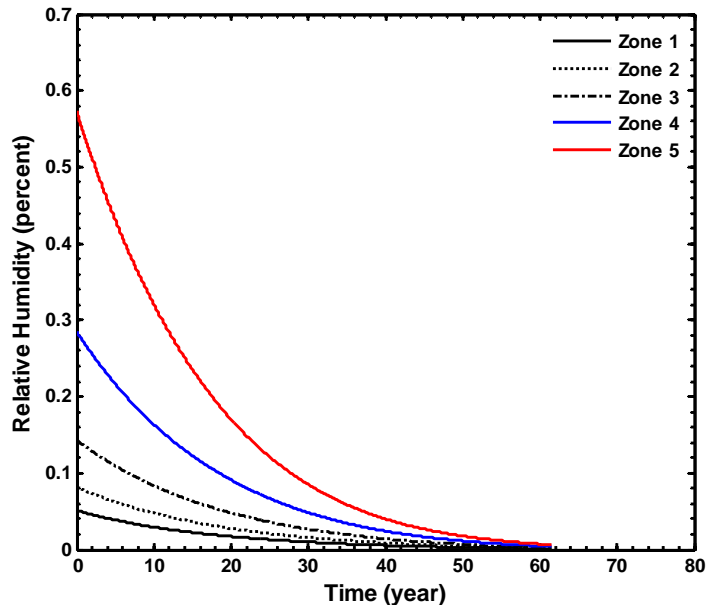


(b) 17.4 moles

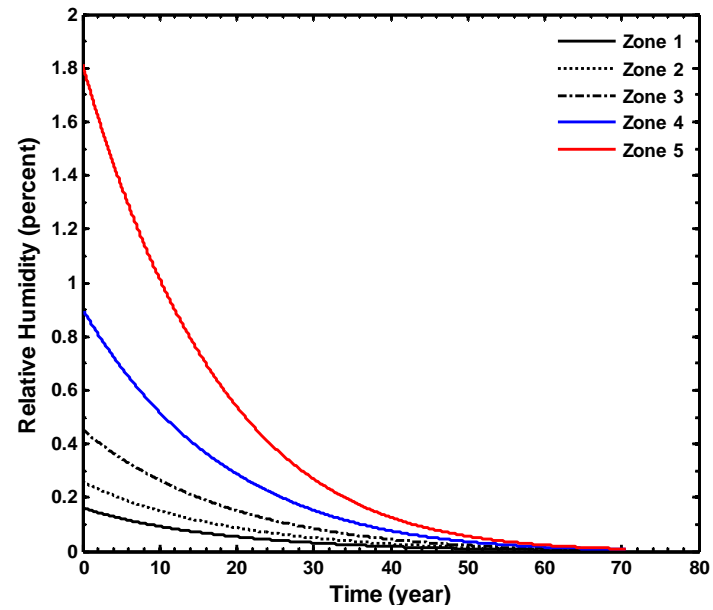


(c) 55 moles

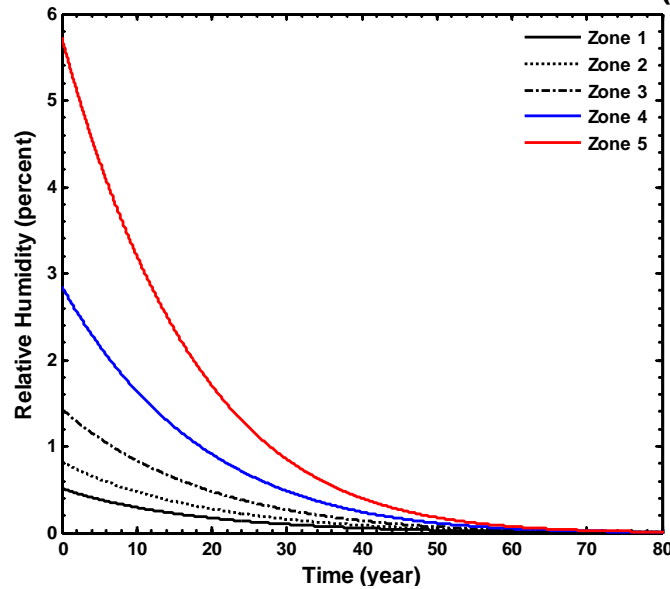
Figure A5-12. Relative Humidity Profiles for Linear Decomposition in 4.77 years, High-End Temperature Condition, and (a) 5.5, (b) 17.4, and (c) 55 moles of Residual Water



(a) 5.5 moles



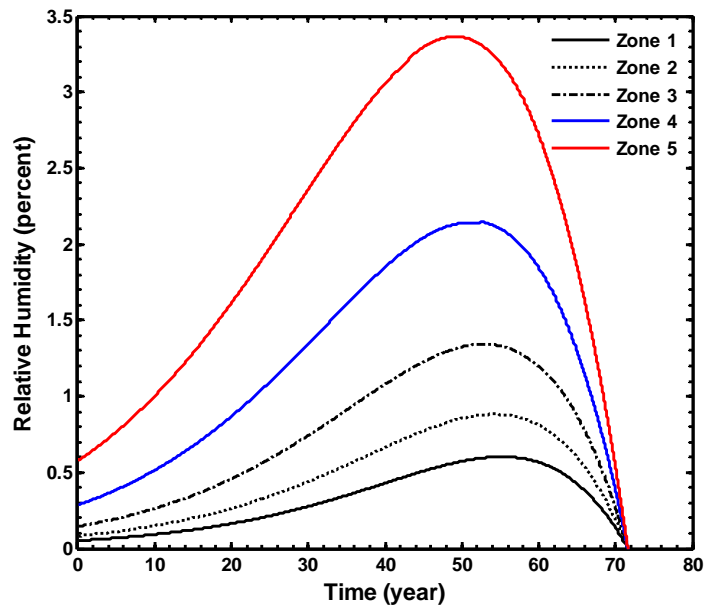
(b) 17.4 moles



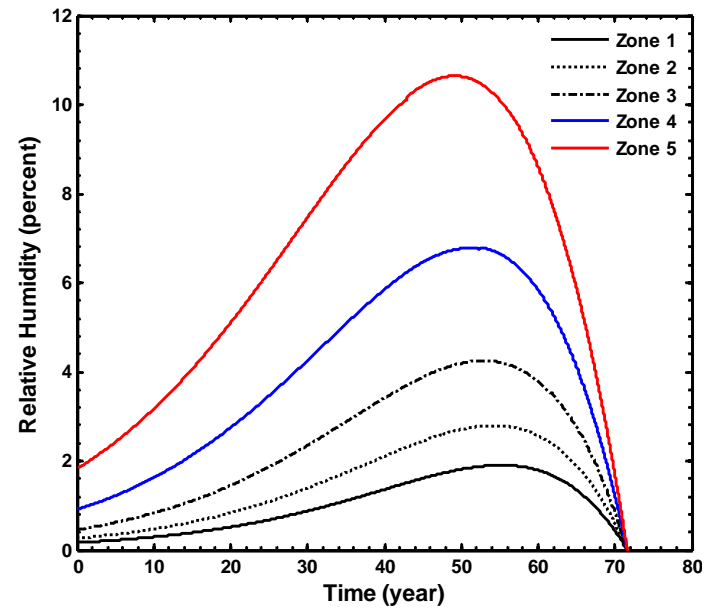
(c) 55 moles

Figure A5-13. Relative Humidity Profiles for Exponential Decomposition in 71.62 years, High-End Temperature Condition, and (a) 5.5, (b) 17.4, and (c) 55 moles of Residual Water

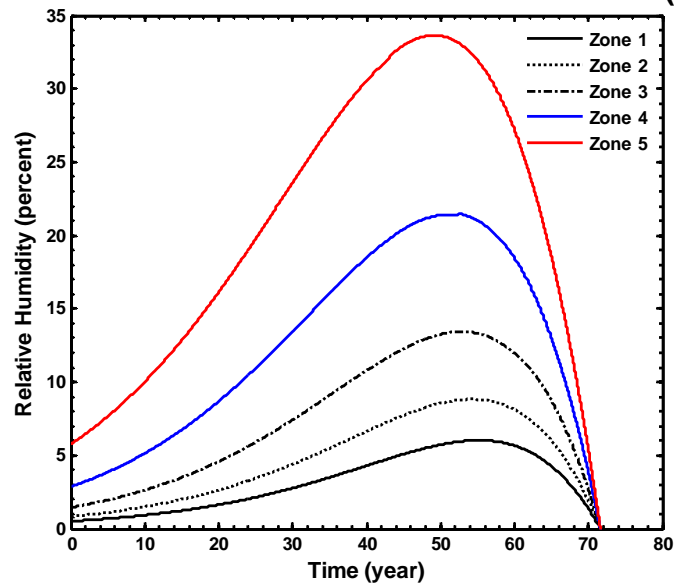




(a) 5.5 moles



(b) 17.4 moles



(c) 55 moles

Figure A5-14. Relative Humidity Profiles for Linear Decomposition in 71.62 years, High-End Temperature Condition, and (a) 5.5, (b) 17.4, and (c) 55 moles of Residual Water

**Entry: MATLAB Code****Date: May 14, 2012****By: Pavan Shukla**

The "run.m" is the main file. All of the files are associated and connected with this file. The MATLAB code and corresponding results are provided in the disk associated with this notebook.

**Entry: Key MATLAB Code Files****Date: May 7, 2013****By: Pavan Shukla**

The "run.m" is the main file. All of the files are associated and connected with this file. The MATLAB code and corresponding results are provided in the disk associated with this notebook.

Script of the key program files is provided.

Script for run.m

\*\*\*\*\*

```
%this is script file for the main run program
```

```
MolesOfWater=[1 5.5 17.4 55] ;
CladdingFailureRate=[0.01 0.1] ;
RadiolysisType='Exponential' ;
RadiationField=[2e14 1.5e16]
Tau_R=[5 70] ;
ThermalDecayConstant=0.023 ;
FuelOxidationModel=3;
NA=6.02214129e23 ;
MW=18.01528 ;
G=7.4/100 ;
sectyr=1/3600/24/365 ;

exponential
for p=1:2,
    if (p==1)
        FuelBurnType='High' ;
    else
        FuelBurnType='Low' ;
    end
for l=1:1,
    if (l==1)
        RadiolysisType='Exponential' ;
    else
        RadiolysisType='Linear' ;
    end
    for k=1:2,
        for i=1:4,
            for j=1:2,
                nwater=MolesOfWater(i);
                CladFailureRate=CladdingFailureRate(j);
                Tau_R(k)=(NA/MW/RadiationField(k)/G)*sectyr ;
                timeforradiolysis=Tau_R(k) ;

Success(i,j)=MainConsideringRadiolysis(nwater,CladFailureRate,RadiolysisType,
timeforradiolysis,...

FuelBurnType,ThermalDecayConstant,FuelOxidationModel) ;
end
```

```

        end
    end
end
end

%linear
for p=1:2,
    if (p==1)
        FuelBurnType='High' ;
    else
        FuelBurnType='Low' ;
    end
for l=2:2,
    if (l==1)
        RadiolysisType='Exponential' ;
    else
        RadiolysisType='Linear' ;
    end
    for k=1:2,
        for i=1:4,
            for j=1:2,
                nwater=MolesOfWater(i);
                CladFailureRate=CladdingFailureRate(j);
                Tau_R(k)=(NA/MW/RadiationField(k)/G)*sectyr
                timeforradiolysis=Tau_R(k)*5

Success(i,j)=MainConsideringRadiolysis(nwater,CladFailureRate,RadiolysisType,
timeforradiolysis,...

FuelBurnType,ThermalDecayConstant,FuelOxidationModel) ;
            end
        end
    end
end
end

%simulation for case5
% mwater=17.4;
% CladFailureRate=0.1;
% FuelBurnType='Low' ;
% RadiolysisType='Linear' ;
% timeforradiolysis=71.72 ;
% ThermalDecayConstant=0.064 ;
% Success=MainConsideringRadiolysis(mwater,CladFailureRate,...
%
RadiolysisType,timeforradiolysis,FuelBurnType,ThermalDecayConstant,FuelOxidat
ionModel);

%simulation for case6
% mwater=17.4;
% CladFailureRate=0.1;
% FuelBurnType='Low' ;
% RadiolysisType='Exponential' ;
% timeforradiolysis=5 ;
% ThermalDecayConstant=0.023 ;

```

```
% FuelOxidationModel=2 ;
% Success=MainConsideringRadiolysis(mwater,CladFailureRate,...
%
RadiolysisType,timeforradiolysis,FuelBurnType,ThermalDecayConstant,...
%
FuelOxidationModel);
```

script for MainConsideringRadiolysis.m file

```
*****
```

```
integration program for the dry storage cask system
```

```
%first calculating the temperature profile
```

```
function
```

```
Success=MainConsideringRadiolysis(mwater,CladFailureRate,RadiolysisType,timeforradiolysis,...
```

```
FuelBurnType,ThermalDecayConstant,FuelOxidationModel)
```

```
%clear
```

```
disp('in the main code')
```

```
%define temperature profile in the caks
```

```
t=0:0.0003:300 ; %time horizon to be 300 years
```

```
%t=0:0.1:300 ;
```

```
t=t' ;
```

```
%temperature and time data is provided by Kaushik and Debashis
```

```
n=max(size(t)) ; % number of time internals
```

```
%consider rate of radiolysis
```

```
%(1) exponential decomposition of water assuming all of the water decomposes in 4.77 years (SD case)
```

```
%(2) linear decomposition of water assuming all of the water decomposes in 4.77 years
```

```
%(3) exponential decomposition of water assuming all of the water decomposes in 71.62 years (LD case)
```

```
%(4) linear decomposition of water assuming all of the water decomposes in 71.62 years (LD case)
```

```
%Note SD stands for short duration and LD stands for long duration
```

```
% form of exponential decay is assumed to be a exp(bt)
```

```
% form of linear decay is assumed to be a exp(bt)
```

```
%defining all the parameters
```

```
%m_water=55; n_zones=5; Vcask=2100; n_assemblies=21 ; fuelrodsperassembly=208 ;
```

```
m_water=mwater; n_zones=5; Vcask=2100; n_assemblies=21 ;
```

```
fuelrodsperassembly=208 ;
```

```
rod_length=3.9 ; rod_od=10.92/1000 ;
```

```
CladArea=acos(-1)*rod_od*rod_length*fuelrodsperassembly*n_assemblies; % vcask is for VSC 17 cask system
```

```
nf=10; %number of fragments of each fuel pellet
```

```
Length_of_crack_per_rod= 3.5 ;
```

```
FailureRate=CladFailureRate/100 ; FuelBurnup=FuelBurnType ;
```

```
Expo='Yes'; Duration=timeforradiolysis ;
```

```
if (strcmp(RadiolysisType,'Exponential')==1)
```

```
Expo='Yes' ;
```

```
else
```

```
Expo='No' ;
```

```
end
```

```

    %parameter definition is finished
fnamesave=CreateFileName(mwater,CladFailureRate,RadiolysisType,FuelBurnType,t
imeforradiolysis)

[Conc_H2O_Selected,Conc_O2_Selected,Conc_H2_Selected]=WaterOxygenHydrogenConc
(m_water,t,Expo,Duration);

% figure(1)
% plot(t(1:700),Conc_O2_Selected(1:700,1))
%break

%defining parameters for cladding oxidation
[f_zones,V_zones,CladArea_zones]=DefineZones(n_zones,Vcask,CladArea) ;
%defining parameters for cladding oxidation
%length of a crack per rod is assumed to be 3.5 cm

[failed_rods_zones,Nexposed_pellets_zones_first,Nexposed_pellets_zones_second
,...

exposed_FuelArea_zones_first,exposed_FuelArea_zones_second,PelletDimentions_f
irst, ...

PelletDimentions_second,PelletDimentions_initial,FuelOxidationUnderneathCrack
,FuelOxidationAwayFromCrack] ...

=CalculateFailedRodsExposedArea(t,n_zones,nf,FuelOxidationModel,Length_of_cra
ck_per_rod,n_assemblies,...
                                fuelrodsperassembly,FailureRate);
FuelOxidationUnderneathCrack
FuelOxidationAwayFromCrack
%pause
% Nexposed_pellets_zones_first
% Nexposed_pellets_zones_second
% exposed_FuelArea_zones_first
% exposed_FuelArea_zones_second
Nexposed_pellets_zones=zeros(n_zones,1) ;
exposed_FuelArea_zones=zeros(n_zones,1) ;
Nexposed_pellets_zones=Nexposed_pellets_zones_first ;
exposed_FuelArea_zones=exposed_FuelArea_zones_first ;
PelletDimentions=PelletDimentions_first ;
PelletDimentions_first_rc=PelletDimentions_first ;
PelletDimentions_second_rc=PelletDimentions_second ;
PelletDimentions_first_kc=PelletDimentions_first ;
PelletDimentions_second_kc=PelletDimentions_second ;
%pause
%break
%calculating temperature and RH profile
[Temp_zones,RH_zones]=CalculateTempRHZones(t,Conc_H2O_Selected,Vcask,f_zones,
V_zones,FuelBurnup,ThermalDecayConstant) ;
% FuelOxideType =1 means U3O8
% FuelOxideType = 2 means U4O9
% FuelOxideType = 3 means UO3
% FuelOxidationModel =1 means base case fuel oxidation model which was
presented on October 26, 2011,
% FuelOxidationModel = 2 means that the U3O8 forms as soon as oxygen react
with UO2 above 230 degree Celsius, and area

```

## Scientific Notebook 1098E

---

```
% for oxygen diffusion is the surface area of each fragment, and
% FuelOxidationModel = 3 means that assuming the U3O8 forms as soon as oxygen
react with UO2 above 230 degree Celsius, and area
% for oxygen diffusion is the surface area of each grain of radius 10
micrometer.
```

```
moles_cladding_zones=zeros(n,n_zones) ;
moles_fueloxide_zones=zeros(n,n_zones) ;
moles_cladding_zones_kc=zeros(n,n_zones) ; %kc stands for kinetically
controlled
moles_fueloxide_zones_kc=zeros(n,n_zones) ;
moles_cladding_zones_rc=zeros(n,n_zones) ; %rc stands for radiolysis
controlled
moles_fueloxide_zones_rc=zeros(n,n_zones) ;
FT=zeros(n,n_zones) ; %FT stands for fuel thickness
```

```
moles_O2_consumed_AllZones_CladOnly=zeros(n,1);
moles_O2_consumed_AllZones_CladPlusFuel=zeros(n,1);
moles_O2_consumed_AllZones_CladOnly_kc=zeros(n,1);
moles_O2_consumed_AllZones_CladPlusFuel_kc=zeros(n,1);
moles_O2_consumed_AllZones_CladOnly_rc=zeros(n,1);
moles_O2_consumed_AllZones_CladPlusFuel_rc=zeros(n,1);
moles_O2_remaining=zeros(n,1) ;
moles_O2_remaining(1:n,1)=m_water/2 ;
O2signal=zeros(n,1) ;
O2_consumedByOxidation=zeros(n,1) ;
O2_producedbyRadiolysis=zeros(n,1) ;
T1_zones=zeros(1,n_zones) ;
T2_zones=zeros(1,n_zones) ;
RH1_zones=zeros(1,n_zones) ;
RH2_zones=zeros(1,n_zones) ;
%moles_O2_remaining(1:n)=m_water/2 ;
%define variables for UO2 oxides
UO2OxidesMass_KineticCase=zeros(n_zones,3) ;
UO2OxidesMass_KineticCase_Delta=zeros(n_zones,3) ;
UO2OxidesMass_RadiolysisCase=zeros(n_zones,3) ;
UO2OxidesMass_RadiolysisCase_Delta=zeros(n_zones,3) ;
UO2OxidesMass_Overall=zeros(n_zones,3) ;
deltamoles_O2_remaining=m_water/2 ;
nexit=n-1 ;
ControllingMechanism='KC' ;
% considering for fuel oxidation underneath the crack
```

```
for i=2:n,
    t1=t(i-1) ;
    t2=t(i) ;
    T1_zones(1,:)=Temp_zones(i-1,:); T2_zones(1,:)=Temp_zones(i,:);
    RH1_zones(1,:)=RH_zones(i-1,:); RH2_zones(1,:)=RH_zones(i,:);
    timestep=i ;
    %cladding and fuel part using kinetically controlled
    %disp('going inside Calculate MolesCladPlusFuelOxide_KC Case')
```

```
[molesOxygen_for_fueloxide_kc,moles_cladding_oxide_kc,PelletDimentions_first_
kc,PelletDimentions_second_kc,...

FT,UO2OxidesMass_KineticCase,UO2OxidesMass_KineticCase_Delta]=CalculateMolesC
ladPlusFuelOxide_KineticLimitingCase(T1_zones,T2_zones,...

RH1_zones,RH2_zones,t1,t2,Conc_O2_Selected,CladArea_zones,exposed_FuelArea_zo
nes_first,exposed_FuelArea_zones_second,...

f_zones,n_zones,timestep,PelletDimentions_first_kc,PelletDimentions_second_kc
,Nexposed_pellets_zones_first,Nexposed_pellets_zones_second,...

FT,FuelOxidationModel,UO2OxidesMass_KineticCase,FuelOxidationUnderneathCrack,
FuelOxidationAwayFromCrack) ;
    %disp('just got out of that function')
    %pause
    for j=1:n_zones,
        moles_cladding_zones_kc(i,j)=moles_cladding_zones_kc(i-
1,j)+moles_cladding_oxide_kc(j,1);
        moles_fueloxide_zones_kc(i,j)=moles_fueloxide_zones_kc(i-
1,j)+molesOxygen_for_fueloxide_kc(j,1);
    end

moles_O2_consumed_AllZones_CladOnly_kc(i,1)=sum(moles_cladding_zones_kc(i,:))
;

moles_O2_consumed_AllZones_CladPlusFuel_kc(i,1)=moles_O2_consumed_AllZones_Cl
adOnly_kc(i,1)+...
                sum(moles_fueloxide_zones_kc(i,:));

    %cladding and fuel part using radiolysis controlled
    %disp('going inside Calculate MolesCladPlusFuelOxide_OxygenLimiting
Case')

[molesOxygen_for_fueloxide_rc,moles_cladding_oxide_rc,PelletDimentions_first_
rc,PelletDimentions_second_rc,...

FT,UO2OxidesMass_RadiolysisCase,UO2OxidesMass_RadiolysisCase_Delta]=Calculate
MolesCladPlusFuelOxide_OxygenLimitingCase(T1_zones,T2_zones,...

RH1_zones,RH2_zones,t1,t2,Conc_O2_Selected,CladArea_zones,exposed_FuelArea_zo
nes_first,exposed_FuelArea_zones_second,...

f_zones,n_zones,timestep,PelletDimentions_first_rc,PelletDimentions_second_rc
,Nexposed_pellets_zones_first,Nexposed_pellets_zones_second,...

FT,FuelOxidationModel,UO2OxidesMass_RadiolysisCase,FuelOxidationUnderneathCra
ck,FuelOxidationAwayFromCrack) ;
    %disp('just got out of that function RC case')

    for j=1:n_zones,
        moles_cladding_zones_rc(i,j)=moles_cladding_zones_rc(i-
1,j)+moles_cladding_oxide_rc(j,1);
        moles_fueloxide_zones_rc(i,j)=moles_fueloxide_zones_rc(i-
1,j)+molesOxygen_for_fueloxide_rc(j,1) ;
```

```

end

moles_O2_consumed_AllZones_CladOnly_rc(i,1)=sum(moles_cladding_zones_rc(i,:))
;

moles_O2_consumed_AllZones_CladPlusFuel_rc(i,1)=moles_O2_consumed_AllZones_CladOnly_rc(i,1)+...
sum(moles_fueloxide_zones_rc(i,:));

if (moles_O2_consumed_AllZones_CladPlusFuel_kc(i,1) <=
Conc_O2_Selected(i,1))
    for j=1:n_zones,
        % moles_cladding_zones(i,j)=moles_cladding_zones_kc(i,j);
        % moles_fueloxide_zones(i,j)=moles_fueloxide_zones_kc(i,j) ;
        moles_cladding_zones(i,j)=moles_cladding_zones(i-
1,j)+moles_cladding_oxide_kc(j,1);
        moles_fueloxide_zones(i,j)=moles_fueloxide_zones(i-
1,j)+molesOxygen_for_fueloxide_kc(j,1) ;
    end

moles_O2_consumed_AllZones_CladOnly(i,1)=sum(moles_cladding_zones(i,:));

moles_O2_consumed_AllZones_CladPlusFuel(i,1)=moles_O2_consumed_AllZones_CladOnly(i,1)+...

sum(moles_fueloxide_zones(i,:));

UO2OxidesMass_Overall=UO2OxidesMass_Overall+UO2OxidesMass_KineticCase_Delta
;
    % UO2OxidesMass_KineticCase
    % moles_fueloxide_zones(i,:)
    moles_O2_remaining(i,1)=m_water/2-
moles_O2_consumed_AllZones_CladPlusFuel(i,1) ;
    % moles_fueloxide_zones(i,1)
    % moles_fueloxide_zones_kc(i,1)
    % moles_fueloxide_zones_rc(i,1)
    % molesOxygen_for_fueloxide_rc(1,1)
    % molesOxygen_for_fueloxide_kc(1,1)
    ControllingMechanism='KC' ;
    % disp(i)
    % disp('reach in the kinetic controlled zone')
    % pause
elseif ((moles_O2_consumed_AllZones_CladPlusFuel_kc(i,1) >
Conc_O2_Selected(i,1))&&(moles_O2_remaining(i-1,1)>0.001)...
&& (deltamoles_O2_remaining > 1.0e-8))
    for j=1:n_zones,
        % moles_cladding_zones(i,j)=moles_cladding_zones_rc(i,j);
        % moles_fueloxide_zones(i,j)=moles_fueloxide_zones_rc(i,j) ;
        moles_cladding_zones(i,j)=moles_cladding_zones(i-
1,j)+moles_cladding_oxide_rc(j,1);
        moles_fueloxide_zones(i,j)=moles_fueloxide_zones(i-
1,j)+molesOxygen_for_fueloxide_rc(j,1) ;
    end

moles_O2_consumed_AllZones_CladOnly(i,1)=sum(moles_cladding_zones(i,:));

```



```

moles_O2_consumed_AllZones_CladPlusFuel(i,1)=moles_O2_consumed_AllZones_CladOnly(i,1)+...

sum(moles_fueloxide_zones(i,:));

UO2OxidesMass_Overall=UO2OxidesMass_Overall+UO2OxidesMass_RadiolysisCase_Delta ;
% UO2OxidesMass_RadiolysisCase
% moles_fueloxide_zones(i,:)
moles_O2_remaining(i,1)=m_water/2-
moles_O2_consumed_AllZones_CladPlusFuel(i,1) ;
% moles_fueloxide_zones(i,1)
% moles_fueloxide_zones_kc(i,1)
% moles_fueloxide_zones_rc(i,1)
% molesOxygen_for_fueloxide_rc(1,1)
% molesOxygen_for_fueloxide_kc(1,1)
ControllingMechanism='RC' ;
% disp(i)
% disp('reach in the radiolysis controlled zone')
% pause
elseif (abs(deltamoles_O2_remaining) < 1.0e-8)
    nextit=i ;
    %pause
    break
else
    nextit=i ;
    %pause
    break
end
deltamoles_O2_remaining=moles_O2_remaining(i-1,1)-moles_O2_remaining(i,1)
;
%pause
% timestep
%disp('going insideCheckZonePelletOxidationMass')
[FuelOxidationUnderneathCrack,FuelOxidationAwayFromCrack]= ...

CheckZonePelletOxidationMass(ControllingMechanism,UO2OxidesMass_KineticCase, ..
..

UO2OxidesMass_RadiolysisCase,UO2OxidesMass_Overall,Nexposed_pellets_zones_first,Nexposed_pellets_zones_second,...

PelletDimentions_initial,n_zones,FuelOxidationUnderneathCrack,FuelOxidationAwayFromCrack,timestep);
% pause
end

for i=1:n_zones,
    moles_cladding_zones(nextit:n,i)=moles_cladding_zones(nextit-1,i);
    moles_fueloxide_zones(nextit:n,i)= moles_fueloxide_zones(nextit-1,i) ;
end
moles_O2_consumed_AllZones_CladOnly(nextit:n,1)=moles_O2_consumed_AllZones_CladOnly(nextit-1,1);
moles_O2_consumed_AllZones_CladPlusFuel(nextit:n,1)=moles_O2_consumed_AllZones_CladPlusFuel(nextit-1,1);
% end

```

```
disp('outside the main for loop')
ControllingMechanism
%pause

[FuelOxideMass_Check,cladthickness_zone]=CladThicknessFuelOxide(CladArea_zone
S, ...

moles_cladding_zones,moles_fueloxide_zones,nexit,n_zones,failed_rods_zones,...
.

Nexposed_pellets_zones,exposed_FuelArea_zones,ControllingMechanism, ...

UO2OxidesMass_KineticCase,UO2OxidesMass_RadiolysisCase,UO2OxidesMass_Overall)
;

% figure(1)
% plot(t,Temp_zones(:,1),'k-',...
%      t,Temp_zones(:,2),'k:',...
%      t,Temp_zones(:,3),'k-.',...
%      t,Temp_zones(:,4),'k--',...
%      t,Temp_zones(:,5),'r-')
%
% figure(2)
% plot(t(1:nexit),RH_zones(1:nexit,1),'k-',...
%      t(1:nexit),RH_zones(1:nexit,2),'k:',...
%      t(1:nexit),RH_zones(1:nexit,3),'k-.',...
%      t(1:nexit),RH_zones(1:nexit,4),'k--',...
%      t(1:nexit),RH_zones(1:nexit,5),'r-')
%
% figure(3)
% plot(t(1:nexit),Conc_O2_Selected(1:nexit),'rd',...
%      t(1:nexit),moles_cladding_zones(1:nexit,1),'k-',...
%      t(1:nexit),moles_cladding_zones(1:nexit,2),'k:',...
%      t(1:nexit),moles_cladding_zones(1:nexit,3),'k-.',...
%      t(1:nexit),moles_cladding_zones(1:nexit,4),'k--',...
%      t(1:nexit),moles_cladding_zones(1:nexit,5),'r-')
%
% figure(4)
% plot(t(1:nexit),moles_fueloxide_zones(1:nexit,1),'k-',...
%      t(1:nexit),moles_fueloxide_zones(1:nexit,2),'k:',...
%      t(1:nexit),moles_fueloxide_zones(1:nexit,3),'k-.',...
%      t(1:nexit),moles_fueloxide_zones(1:nexit,4),'k--',...
%      t(1:nexit),moles_fueloxide_zones(1:nexit,5),'r-')
%
% figure(5)
% plot(t(1:nexit),Conc_O2_Selected(1:nexit),'ro',...
%      t(1:nexit),moles_O2_consumed_AllZones_CladPlusFuel(1:nexit),'k-')
%
% figure(6)
% plot(t(1:nexit),Conc_O2_Selected(1:nexit),'ro',...
%      t(1:nexit),moles_O2_consumed_AllZones_CladPlusFuel(1:nexit),'k-')
% %
% % UO2OxidesMass_KineticCase
% % UO2OxidesMass_RadiolysisCase
% % UO2OxidesMass_Overall
```

```

% % ControllingMechanism
% %
% %
% figure(10)
% plot(t(1:200),moles_fueloxide_zones(1:200,1),'rd',...
%      t(1:200),moles_fueloxide_zones_rc(1:200,1),'k-',...
%      t(1:200),moles_fueloxide_zones_kc(1:200,1),'k-.')
% %
% % figure(11)
% % plot(t(1:200),moles_fueloxide_zones(1:200,1),'k-',...
% %      t(1:200),moles_fueloxide_zones_kc(1:200,1),'ro')
% %
% % figure(12)
% % plot(t(1:200),moles_fueloxide_zones(1:200,1),'k-',...
% %      t(1:200),moles_fueloxide_zones_rc(1:200,1),'rd')

[stringClad,CladResultSummary]=SummarizeCladThicknessResults(CladArea_zones,c
ladthickness_zone,n_zones);
disp(stringClad)
disp(CladResultSummary)

[stringFuel,FuelResultSummary]=SummarizeFuelThicknessResults(n_zones,failed_r
ods_zones,...

Nexposed_pellets_zones_first,Nexposed_pellets_zones_second,exposed_FuelArea_z
ones_first,...

exposed_FuelArea_zones_second,FuelOxidationUnderneathCrack,FuelOxidationAwayF
romCrack,UO2OxidesMass_Overall);
disp(stringFuel)
format short g;
disp(FuelResultSummary)
format short
save(fnamesave) ;
Success=1 ;
end
%[moles_fueloxide_zones(1:200,1) moles_fueloxide_zones_rc(1:200,1)
moles_fueloxide_zones_kc(1:200,1)]

*****

```

Other program files are available in the disk attached with this notebook.





# GEOSCIENCES AND ENGINEERING DIVISION

## SCIENTIFIC NOTEBOOK REVIEW CHECKLIST RECORD

Scientific Notebook No. 1098E Project Numbers: 20.14010.01.002 Title: Development of an Integration Model to Evaluate Affect of Residual Water on Fuel and Cladding Oxidation

### Accomplished

1. Initial entries per QAP-001
2. Dating of entries
3. Corrections (crossed out, one line through w/initials/date)
4. No white out used
5. Page number visible on copy or original notebook
6. In process entries per QAP-001
7. Figure information present
8. Text readable
9. Blank pages and blank portions of pages have a diagonal line to prevent unauthorized future entries.
10. Permanent ink or type only
11. Signing of entries (not required on each page)
12. Electronic media in the scientific notebook properly labeled
- N/A 13. Copyrighted or proprietary material fully referenced

Any discrepancies must be resolved before notebook closeout.

=====

I have reviewed this scientific notebook and find it in agreement with QAP-001.

\_\_\_\_\_  
Manager Signature

5/7/2013  
Date