U.S.NRC
United States Nuclear Regulatory Commission
Protecting People and the Environment

EXTENDED DRY STORAGE AND TRANSPORTATION: MODEL FOR EVALUATING VACUUM DRYING ADEQUACY

T. Ahn,[1] H. Jung,[2] P. K. Shukla,[2] E. L. Tipton[2] and K. Das,[2]

[1]U.S. Nuclear Regulatory Commission, Washington, DC 20555, USA

[2]Center for Nuclear Waste Regulatory Analyses (CNWRA®), Southwest Research Institute, San Antonio, Texas 78238, USA

Contact: Tae M. Ahn, tae.ahn@nrc.gov

2013 International High-Level Radioactive Waste Management Conference (IHLRWMC), April 28 – May 2, 2013, Albuquerque, New Mexico, U.S.A.

U.S.NRC
United States Nuclear Regulatory Commission
Protecting People and the Environment

DISCLAIMER

The NRC staff views expressed herein are preliminary and do not constitute a final judgment or determination of the matters addressed or of the acceptability of any licensing action that may be under consideration at the NRC.

ACKNOWLEDGMENTS

The authors thank Dr. C. Markley of the NRC for his review of this paper.

U.S.NRC
United States Nuclear Regulatory Commission
Protecting People and the Environment

Introduction

- **Potential excessive residual moisture following incomplete drying of spent nuclear fuel (SNF)**

- **A time-dependent integration model, to assess potential radiolysis, producing oxygen and hydrogen, and potential effects on the integrity of SNF, cladding, internals, and flammability**

- **Interim progress made for the process of model abstraction, model integration, and uncertainty assessments. Quantitative results will be presented in the future.**

U.S.NRC
United States Nuclear Regulatory Commission
Protecting People and the Environment

Outline

- **Quantity of Water Remaining after Drying: water reacts with SNF, cladding and internals**
- **Temperature: affects reactions**
- **Radiolysis: produces oxygen and hydrogen; affects reactions; poses flammability potential**
- **Relative Humidity (RH): affects reaction**
- **Oxidation and Hydration of SNF: affect SNF integrity**
- **Cladding Oxidation: affects cladding integrity**

U.S.NRC
United States Nuclear Regulatory Commission
Protecting People and the Environment

Outline (continued)

- **Cladding Splitting - Strain Estimate due to SNF Swelling: affects integrity of SNF and cladding**
- **Flammability**
- **Hydrogen-Absorption-Induced Damage**
- **Aqueous Corrosion: affects reactions**
- **Integration and Benchmarking**

U.S.NRC
United States Nuclear Regulatory Commission
Protecting People and the Environment

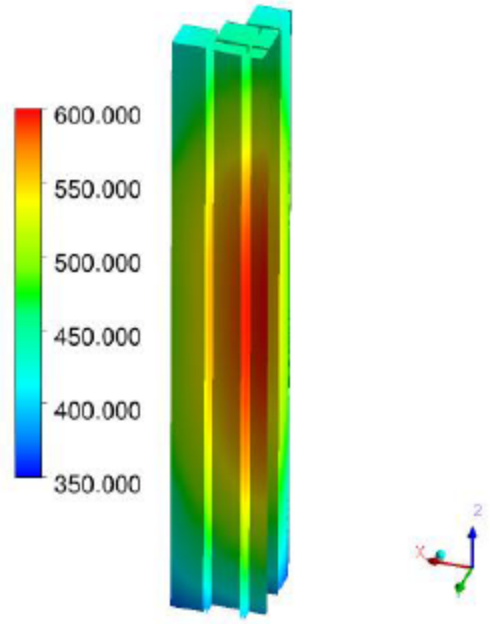
Quantity of Water Remaining after Drying and Temperature

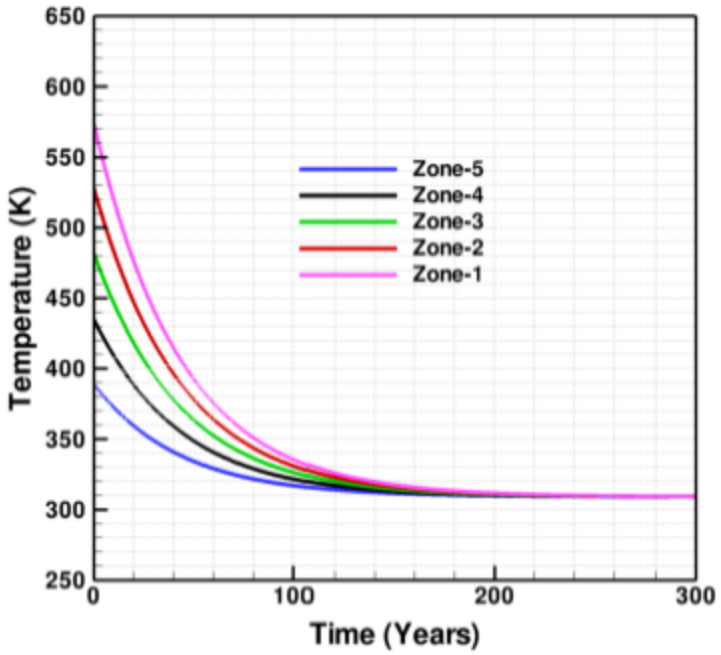
- **Quantity of Water Remaining after Drying**
  - **Unbound residual liquid water, unbound water vapor, and water chemically bound to hydroxide and hydrate species**
  - **Properly executed vacuum drying procedure result in: 1 to 5 moles (0.02 to 0.1 L [0.7 to 3.5 oz]) water inventory**
  - **up to 55 moles (~ 1 L [35 oz]) are conservatively assumed**
- **Temperature**
  - **Spatially distributed and decreases over time**

U.S.NRC
United States Nuclear Regulatory Commission
Protecting People and the Environment

Temperature (continued)

- **Temperature distribution in the SNF basket assembly (left). The scales for temperature are in K (F = 1.8 K – 459.4); SNF and cladding in the five zones calculated using mean values of low-end SNF and cladding initial temperatures (right)**





U.S.NRC
United States Nuclear Regulatory Commission
Protecting People and the Environment

Radiolysis and Relative Humidity (RH)

- **Radiolysis: Global Approximation for Various Intermediate Species**

$$2H_2O \leftrightarrow 2H_2 + O_2$$

  - **Decomposition rate of water by radiolysis: exponential time function**
  - **Recombination – additional steps for molecular collision needed, compared with decomposition**

- **Relative Humidity (RH)**
  - **The radiolysis removes water molecules by dissociating them into oxygen and hydrogen**
  - **RH is modified by radiolysis**

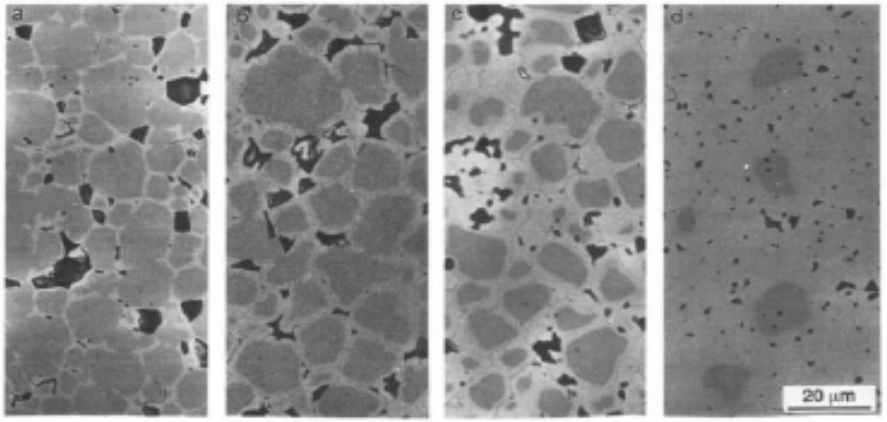
U.S.NRC
United States Nuclear Regulatory Commission
Protecting People and the Environment

Oxidation and Hydration of SNF

- **Upon cladding breach, the UO_2 could be oxidized to form:**

- U_4O_9, U_3O_7, and U_3O_8 in dry air (less than 40 percent RH)

- Hydrated uranium oxides, such as schoepite ($UO_3 \cdot xH_2O$, x = 0.5 to 2), in humid air (greater than 40 percent RH) or in an aqueous environment



Grain boundary oxidation of AM-105 spent nuclear SNF to U_4O_9. Optical ceramographs, as polished: (a) 95 hrs, Bulk O/M = 2.05; (b) 420 hrs, bulk O/M=2.17; (c) 775.5 hrs, bulk O/M = 2.24; and (d) 1,677 hrs, bulk O/M = 2.31 (Einziger, et al., 1992)

- **Chemical Arrhenius-type Kinetics**
- **Actual Rates are Controlled by Radiolysis Kinetics or Chemical Kinetics**

U.S.NRC
United States Nuclear Regulatory Commission
Protecting People and the Environment

Cladding Oxidation, and Cladding Splitting: Strain Estimate due to SNF Swelling

- **Cladding Oxidation**

  - **$Zr + O_2 = ZrO_2$ for dry air and**

  - **$Zr + 2H_2O = ZrO_2 + 2H_2$ for water or humid air (e.g., steam)**

  - **Model by Hillner, et al (1994) and 8 other similar kinetics**

  - **Large surface area of cladding**

  - **Actual Rates Controlled by Radiolysis Kinetics or Chemical Kinetics**

- **Cladding Splitting: Strain Estimate due to SNF Swelling**

  - **Oxidation of UO_2 to U_3O_8 can generate stress on cladding as U_3O_8 swells (36 percent when there is 100 percent conversion to U_3O_8).**

  - **Various threshold values of strain are assessed to determine the cladding splitting condition.**

  - **Observed threshold values:**
    6.5% strain (100% conversion);
    5.1% volume expansion correlation (25 % conversion);
    2% strain (50% conversion)

U.S.NRC
United States Nuclear Regulatory Commission
Protecting People and the Environment

Flammability and Hydrogen-Absorption-Induced Damage

- **Flammability**

  - **The cladding will not absorb radiolysis-generated hydrogen.**

  - **The flammability criterion requires the volume fraction of any flammable gas to be more than 5 percent, with oxygen and ignition.**

- **Hydrogen-Absorption-Induced Damage**

  - **Cladding: Zirconium oxide is expected to limit the absorption of molecular hydrogen:**

    Total possible amount of hydrogen from 55 mole water is assessed.

  - **Canister Internals: the mechanical properties may or may not be degraded with hydrogen concentration exceeding a threshold value.**

U.S.NRC
United States Nuclear Regulatory Commission
Protecting People and the Environment

Aqueous Corrosion, and Integration and Benchmarking

- **Aqueous Corrosion**

  - Aqueous corrosion could occur in vapor when RH is greater than a threshold value: At above the threshold RH

  (i) SCC of the canister's internal structural components could occur in carbon and stainless steels; no relevant data available

  (ii) Shadow corrosion (a form of galvanic corrosion) between cladding and spacer-grid material could occur.

- **Integration and Benchmarking**

  - In each time step, all reactions and gas generations are assessed

  - Bench marking with long-term demonstration data (e.g., hydrogen amount)

U.S.NRC
United States Nuclear Regulatory Commission
Protecting People and the Environment

Summary

Program is under development that evaluates:

- **Time-dependent integrated models for temperature, relative humidity, and radiolysis kinetics to decompose water into oxygen and hydrogen.**
- **Various abstracted models for temperature, radiolysis, relative humidity, and chemical reactions of SNF, cladding and internals.**
- **Time integrated model: in each time step, all reactions and gas generations are assessed.**
- **Assess degradation of SNF, cladding and internals.**
- **Possibility of flammability conditions.**