NUCLEAR WASTE TECHNICAL REVIEW BOARD
FULL BOARD MEETING

SUBJECT: OVERVIEW OF SPENT FUEL MODELING CONCEPTS

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PLAZA SUITE HOTEL • LAS VEGAS, NEVADA
OCTOBER 14 - 16, 1992
Spent Fuel Characterization Activities

Objective:

- To provide data, testing, and models for the physical properties, the degradation responses, and the radioactive release responses of spent-fuel waste forms for waste package and system performance assessments in the Yucca Mountain Project

Product:

- Preliminary Waste Form Characteristics Report [MO3]

Contents are the available

- Physical property data for existing and projected SFWP & DHLW inventories
- Radionuclide data for existing and projected SFWP & DHLW inventories
- Test data and models for potential release rates from SFWP & DHLW
Outline

• Introduction: Spent fuel characteristics

• Release modes: gaseous and aqueous

• Models for test matrix design and for preliminary design: Performance Assessment (PA)
  - Gaseous
  - Cladding
  - Oxidation
  - Dissolution
  - Hardware

• Summary
Spent Fuel Model Development: Problem of Length Scale
Spent Fuel Features Important to Oxidation and Release Responses

Illustration of the cladding gap and the fragment surfaces. Fragment surface consists of grain boundary and grain areas.

- Fuel grain release ~ actinide species
- Gap release ~ soluble species
- Grain boundary release ~ soluble species
- Crack release
- Fuel cladding
- Pellet interfacial gap

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Source Term Release-Rate Diagram

Total Radionuclide Release Rate
Gaseous plus constrained solution

Radionuclide
Inventory in
Waste Packages
i. gaseous
ii. fuel & (glass)
iii. metal (hardware)

Container Failure Response
Rods and Hardware Exposed
Rod Failure Response
Fuel (+ glass) Exposed

Potential water contact for all temperatures < ~ 95°C
Air contact at all temperatures
Potential water contact for all temperatures < ~ 95°C
Air contact at all temperatures
Potential water contact for all temperatures < ~ 95°C

Waste Package Environmental (Air, Water, Temperature) History

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Spent Fuel Responses

Potential event sequence in time

- Gaseous release response from cladding
  - Carbon $^{14}$ release
  - $\dot{G}$ and $\Delta G$

- Rod failure time response
  - $\dot{R}_f$

- Cladding degradation response

- Oxidation response
  - $\dot{O}$

- UO$_2$ oxidation response

- Phase change kinetics
  - $P_{NM}$, eg $P_{49} \sim U_4O_9$
  - $P_{38} \sim U_3O_8$

- UO$_2$ dissolution response

- Dissolution response
  - $D$ (gm/m$^2$/day) and $\Delta D$

- Geochemistry solution response

- Colloidal response
  - Solubility limits and $S_L$ and $C_R$

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Spent Fuel: Knowledge & Data Base Activities

Main topics - Spent Fuel
1. Gaseous Release Response
2. Cladding Failure Response
3. Oxidation Response
4. Dissolution Response
5. Hardware Release

Pre-advanced Conceptual Design & Assessments: Preliminary Waste Form Characteristics Report v. 1.0

Advanced Conceptual Design & Assessments: Waste Form Characteristics Report v. n.0

License Application Design: Waste Form Characteristics Report
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Spent Fuel Inventory: History and Projection

Current Inventory: 20,000 tons
Projected Inventory: 80,000 tons

Burnup (GWd/MTU)

COURTESY: ORNL

BWR
PWR

1968-1987
1968-2037

Thousands of Tons of Waste

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Assembly Class Quantities and Typical Dimensions

GE BWR/4,5,6
- Assembly length ~ 175 in.
- Assembly area ~ 6 in. x 6 in.
- Assembly weight ~ 562 lbs.
- #Rods/assemble ~ 63
- #Pellets/Rod ~ 360

GE BWR/2,3
- Dresden 1
- Humboldt Bay
- Big Rock Point
- Lacrosse
- Elk River
- WE 15 x 15 array
- WE 17 x 17 array

WE PWR typical attributes
- Assembly length ~ 160 in.
- Assembly area ~ 8.5 in. x 8.5 in.
- Assembly weight ~ 1373 lbs.
- #Rods/Assembly ~ 264
- #Pellets/Rod ~ 288

- WE 15 x 15 array
- WE 17 x 17 array
- BW 15 x 15 array
- CE 14 x 14 array
- WE 14 x 14 array
- CE 16 x 16 array
- Haddam Neck
- Pallisades
- San Onofre 1
- Fort Calhoun
- Yankee Rowe
- Saint Lucie 2
- Indian Point 1
- BW 17 x 17 array
- South Texas 1&2

Dec. 31, 1987 2020 A.D.
Illustrative Rod Population Distribution of Gap and Grain Boundary Inventory

- Fission Gas Release
- Gap and Grain Boundary Distribution Function
- Average

Quantity of Spent Fuel # Rods (Unit Inventory)

Gap and Grain Boundary Inventory, %I, Cs, and Tc
Spent Fuel Inventory Attributes for Test Matrix (Illustrative)

Future SF testing inventory: low and high burnup with low and high FGR.
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Gaseous Release

\[ \dot{\text{Release}}_{(\text{gas})} = C \otimes R_o \otimes \dot{G}_{(\text{rod})} + C \otimes R \otimes \dot{G}_{(\text{fuel})} \\
+ \dot{C} \otimes R_o \otimes \Delta G_{(\text{rod})} + C \otimes \dot{R} \otimes \Delta G_{(\text{fuel})} \]

Waste Form Problems ... provide response functions (models) for R, \( \dot{R} \), — rod failure response

\( \dot{G}_{(\text{rod})} \) and \( \dot{G}_{(\text{fuel})} \) ... gaseous release rate

\( \Delta G_{(\text{rod})} \) and \( \Delta G_{(\text{fuel})} \) ... rapid release increment

**Primary Concern** — Carbon-14
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Zircaloy Cladding Degradation

Oxide film failure – stress corrosion cracking inhibitor

Hydride platelet precipitation

Zircaloy-fluoride corrosion response

Thin-walled tube
Radius/thickness ~8

Zirconium oxide film
Gas Pressure
Zircaloy

Zirconium oxide film
Zircaloy

Zirconium oxide film
Fluoride ion F⁻
Zircaloy

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Zircaloy Cladding Failure Modes Illustrated

Time to failure function will depend on stress-temperature-time histories and on initial hydrogen, oxide film, and stress conditions.
Pressurized Tube Testing System Developed

Developed apparatus for Zircaloy cladding failure testing:

1. High-temperature oxidation, time testing

2. High-temperature and high-stress deformation, time testing

3. Oxide film through-crack, strain-time testing

4. Hydride precipitation-reorientation, temperature-time testing

5. Oxide film crack propagation due to hydrides, stress-time testing
Zircaloy Cladding Degradation: Expected Total Modeling Response

Schematic of cumulative response curve for probable number of failed spent fuel rods at a time t

- Initial number rods failed
- High temperature high pressure failures
- Hydride stress reorientation failure
- Zircaloy-fluoride corrosion failures

Number rods failed

t time
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Oxidation Response

Fuel pellets, nominally 0.5cm to 0.6cm radius and ~2cm length, fracture into fragments due to thermal strains during first full power cycle.

Tests on spent fuel fragments clad have shown a grain boundary oxidation front moving into fragments, followed by a spatial zone where oxidation of individual grain volumes occur.

Oxidation kinetics depend statistically on fragment/grain sizes and shapes in a test sample; any fragment/grain can be subdivided into different sized pyramidal volume subsets to obtain a statistical distribution function.
Grain Volume Oxidation Front

Pyramidal volume in an oxidizing grain volume and its associated physical attributes.

Oxygen weight gain rate

\[ \dot{O}(t) = Ne_{ijk} a_j b_k c_i (1-C)^2 \dot{C} \]
Spent Fuel Oxidation Rate Response Surface

\[ \dot{O} \text{ (or } \dot{D}) \]
oxidation rate
\[ \text{gm/yr (or } \mu\text{m/yr)} \]
weight gain rate \( \sim \dot{O} \)
front propagation rate \( \sim \dot{h} \)

- Air moisture
- Temperature
- Oxidation phase
- FGR
- BWR/PWR/AECL Burnup
- Burnup

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Spent Fuel Oxidation Response - Conceptualization for Model

- Space-Time boundary between UO₂ and U₄O₉ phases
- Time to U₄O₉ surface
- Time to U₃O₈ surface
- Space-Time boundary between U₄O₉ and U₃O₈ phases
- Time to U₃O₈ grain

\[ \Delta \text{O/M} \]

UO₂₆₆

UO₂₄

Grain size

Time to U₄O₉ grain

\( t \) (time)
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Oxidation Front and Dissolution Front Analog

Grain boundary/volume oxidation front motion through a pellet fragment has geometrical and model development similarities to an idealized dissolution surface motion progressing into a fragment.

This means that model development concepts for oxidation kinetics can be also applied to dissolution response for a distribution of fragments.

Initial outer surface at $t = 0$

Current surface at time $t$

Dissolved spatial domain

Zone of dissolution

Spent fuel undissolved

Fragment cross-section
Grain Boundary Attack on Bare Fuel
Turkey Point Fuel in Deionized Water for 1 Year

Photo

Photo

30 μm
A Schematic View of Spent Fuel Dissolution*

Release Rate: Volume of Fragments - Aqueous (Flow-thru Case Only)

Approximate Size (cm) \((2X_0)\) | Weight (Volume) Fraction
---|---
0.15 | 0.02
0.25 | 0.14
0.35 | 0.29
0.50 | 0.38
0.70 | 0.17

Temperature \(\circ C\) | Dissolution Time (years)
---|---
25 | \(8.0 \times 10^3\) | \(5.5 \times 10^4\)
85 | \(2.2 \times 10^3\) | \(1.5 \times 10^4\)

Fuel Fragments Distribution

monodisperse
Dissolution Release — Spent Fuel Pellets — Aqueous (Flow Thru)

\[
\dot{\text{Release}}_{(aq)} = C \otimes R \otimes A_e \otimes A_w \otimes V_w \otimes \dot{D} \\
+ \dot{C} \otimes R \otimes A_e \otimes A_w \otimes V_w \otimes \Delta D \\
+ C \otimes \dot{R} \otimes A_e \otimes A_w \otimes V_w \otimes \Delta D \\
+ C \otimes R \otimes A_e \otimes \dot{A}_w \otimes V_w \otimes \Delta D
\]

- \( C \) ... Container failure
- \( R \) ... Rod failure
- \( A_e \) ... Area exposed/rod(R)
- \( A_w \) ... Area wetted/V_w/A_e
- \( V_w \) ... Volume water rate
- \( \dot{D} \) ... gm/m^2/yr, dissolution rate
- \( \Delta D \) ... instantaneous dissolution

\( \dot{\text{Release}}_{(aq)} \) — no precipitation (solubility limits) constraints

\[
\dot{\text{Release}}_{(solution)} = \{ \dot{\text{Release}}_{(aq)} \text{ subject to solubility limits and colloidal response restraints} \}
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# Release Rate Response: Hardware

<table>
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<tr>
<th>Parameter</th>
<th>Description</th>
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<tbody>
<tr>
<td><strong>Release Rate</strong></td>
<td>- [Inventory] • [Dissolution Rate] • [Area]</td>
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<tr>
<td><strong>Hardware</strong></td>
<td></td>
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<tr>
<td><strong>Inventory</strong></td>
<td>- Activated species - used ORIGEN2 computer code</td>
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<tr>
<td><strong>Dissolution Rate</strong></td>
<td>- Metal corrosion rate (gm / area / time)</td>
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<td><strong>Area</strong></td>
<td>- Estimates have been made</td>
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<td>SS ~ 652,000 M²; Inconel ~ 1,480,000 M²; Zircaloy ~ 400,000 M²</td>
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Summary: Spent Fuel Response (Rate Processes)

Spent fuel attributes (SFA)
- Initial condition data have large spread (variance)
- Much remains to be acquired (significances addressed)

Repository environment (REV)
- Space-time-dependent boundary conditions that have uncertainty (Stochastic)
- Information remains to be characterized (uncertainty)

Mechanistic understanding of rate processes
- Knowledge base & models are being acquired with a multi-laboratory established testing program (concrete)
- Tests matrix for domain of SFA and REV (closure)
- Testing and modeling viewed as an iterative interface (checks)

Preliminary knowledge base and models
- Incomplete, but are being integrated in a “Preliminary Waste Form Characteristic Report” (document)
Organization of the Waste Form Characterization Report

Chapter 2
Technical Bases for Design

Chapter 3
Scientific Bases for mechanistic Model Development

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