LSNReviews

From: Budhi Sagar [bsagar@cnwra.swri.edu]
Sent: Thursday, September 13, 2007 5:23 PM
To: 'kaxler@swri.org'; 'smohanty@swri.edu'
Cc: 'gwittmeyer@swri.org'; 'Dave Turner'; 'jwinterle@swri.edu'; 'achowdhury@swri.edu'; 'epearcy@cnwra.swri.edu'; 'pdubreuilh@cnwra.swri.edu'; 'Robert Lenhard'; 'wpatrick@swri.org'
Subject: FW: Slides for ACNW&M
Attachments: ACNW_0907_EBS_BEH_OGC.PPT; ACNW Corrosion 9 18 2007 Ahn Final.ppt; ACNW Juvenile WP Failure 9 18 2007 Dunn Final.ppt; ACNW Spent Fuel 9 18 2007 Ahn Final.ppt

Keith and Sitakanta,

We have conference room A 237 reserved for this. There would be a video connection. Please make sufficient copies for use during the presentations. I assume that Keith will be the coordinator of this meeting from our end.

Budhi

-----Original Message-----
From: Tae Ahn [mailto:TMA@nrc.gov]
Sent: Thursday, September 13, 2007 2:59 PM
To: Christopher Brown
Cc: Budhi Sagar; Yi-ming Pan
Subject: Slides for ACNW&M

Chris, please find four sets of slides to be presented to ACNW&M on September 18, 2007. Thank you, Tae

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Introduction to Engineered Barrier System Discussions

182nd Advisory Committee on Nuclear Waste and Materials Meeting
September 18, 2007

Dr. Brittain Hill

Sr. Technical Advisor for Repository Science
Office of Nuclear Materials Safety & Safeguards
Division of High-Level Waste Repository Safety
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Today's Goals

- Summarize Important Processes for
  - Waste package & drip shield corrosion
  - Juvenile failure of waste packages
  - Waste form dissolution
- Discuss Process-Level Uncertainties
- Provide Insights on Risk
- Background for October TPA Discussions

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Risk Insights

- High Significance to Performance
  - Passive film persistence
- Medium Significance to Performance
  - Localized & crevice corrosion
  - Drip shield integrity
  - Waste form degradation rate
- Low Significance to Performance
  - Juvenile failure of waste package

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Next Steps

- Approve and Release TPA 5.1
  - Users guide
  - Code
- October 2007 ACNW&M Meeting
  - Overview of changes from TPA 4.1j to TPA 5.1

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Evaluation of Waste Package and Drip Shield Juvenile Failure Rates

Presentation to the Advisory Committee on Nuclear Waste and Materials (ACNW&M)

Darrell Dunn

Contributors
T. Ahn (NRC), D. Daruwalla (SwRI), T. Mintz (CNWRA),
K. Chiang (CNWRA), Y.-M. Pan (CNWRA), O. Pensado (CNWRA),
K. Axler (CNWRA), Tina Ghosh (NRC)

September 18-20, 2007
Rockville, MD

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Outline

- Definition of Juvenile Failure
- Factors That Influence Juvenile Failure Rates
- Industrial Failure Rate Data
- Uncertainties For Waste Packages and Drip Shields
- Summary

Juvenile Failure

- DEFINITION: Penetration through the waste package or the drip shield during the preclosure period
- Defects that do not penetrate the waste package and drip shield may also exist
- In the TPA code, juvenile failures are conservatively estimated to occur at the start of the postclosure period (t = 0)
- The TPA code contains mechanistic models to evaluate waste package and drip shield performance during postclosure

Factors That Influence Juvenile Failure Rates

- Design codes and requirements
- Materials selection
- Fabrication processes
- Operating parameters and procedures
- Nondestructive examination and inspection
- Human reliability considerations

Industrial Failure Rate Data

- Failure rates from industrial experience are not directly applicable to waste packages or drip shields
  - Some similarity in materials, fabrication processes and design codes
  - Dissimilar operating conditions and inspection criteria
- Decreased industrial failure rates as a result of operating experience, improvements in nondestructive examination, and use of design codes
- Industrial data examples
  - Boiler and pressure vessels use similar fabrication processes and design codes
  - Fuel rods and storage casks selected as examples from the nuclear industry
- Dry Storage Casks for Spent Nuclear Fuel
  - No reported failures in service (licensed since 1986)
  - 4 reported cases of weld defects found during post-weld inspection of VSC-24 casks (19 in service through July 1998)

Uncertainties For Waste Packages and Drip Shields

- Weld defect densities and nondestructive examination
- Residual stress mitigation
  - Non uniform heating/cooling, process variability, and laser peening for waste packages
  - Processes, procedures, and inspection methods for the drip shield not fully defined
- Handling procedures and emplacement processes
- Task analysis and human error rates

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Summary

- Industrial failure rates not directly applicable to waste packages and drip shields, however, a correlation can be made to general industrial fabrication technologies and inspection methods/criteria that will be utilized in waste package construction
- For industrial components considered in our study, failures early in service are due to fabrication defects, deficient nondestructive evaluation/inspection, and human errors
- Decrease in industrial failure rates attributed to increased experience, improvements in nondestructive examination and use of design codes
- Uncertainties identified for waste packages and drip shields are expected to be addressed

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This presentation describes work performed by the Center for Nuclear Waste Regulatory Analyses (CNWRA) for the U.S. Nuclear Regulatory Commission (NRC) under Contract No. NRC–02–02–012. The activities reported here were performed on behalf of the NRC Office of Nuclear Material Safety and Safeguards, Division of High-Level Waste Repository Safety. This presentation is an independent product of the CNWRA and does not necessarily reflect the view or regulatory position of the NRC. The NRC staff views expressed herein are preliminary and do not constitute a final judgment or determination for the matters addressed or of the acceptability of a license application for a geologic repository at Yucca Mountain.

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Introduction to Engineered Barrier System Discussions

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Corrosion of Waste Package and Drip Shield Materials in Potential Repository Conditions

Presented by T. Ahn, NRC

Team Leads: S. Whaley, NRC, and Y. Pan, Center for Nuclear Waste Regulatory Analyses (CNWRA)

Other Contributors: H. Jung, X. He, L. Yang, and O. Pensado, CNWRA

182nd Meeting of the Advisory Committee on Nuclear Waste and Materials (ACNW&M), September 18– 20, 2007

U.S. Nuclear Regulatory Commission, Rockville, MD

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Outline

- Purpose
- Engineered Barrier System
- Waste Package Environment and Corrosion Modes
- General Corrosion: Persistence of Passive Film
- Dust Deliquescence Corrosion
- Seepage Water Brines: Crevice Corrosion
- Additional Corrosion Processes
- Summary
- References

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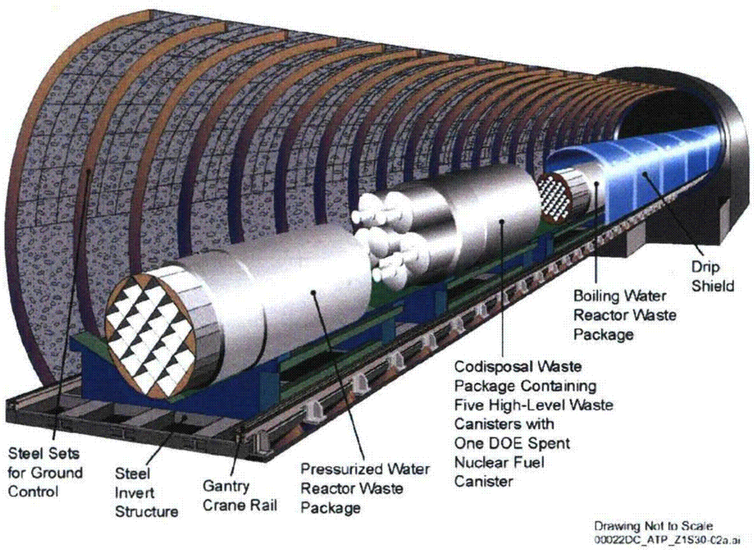
Purpose

- Summarize key processes affecting corrosion in the waste package (WP) and the drip shield (DS) at the potential Yucca Mountain (YM) repository

- Discuss current understanding of potentially significant uncertainties in corrosion processes for Alloy 22 and titanium alloys

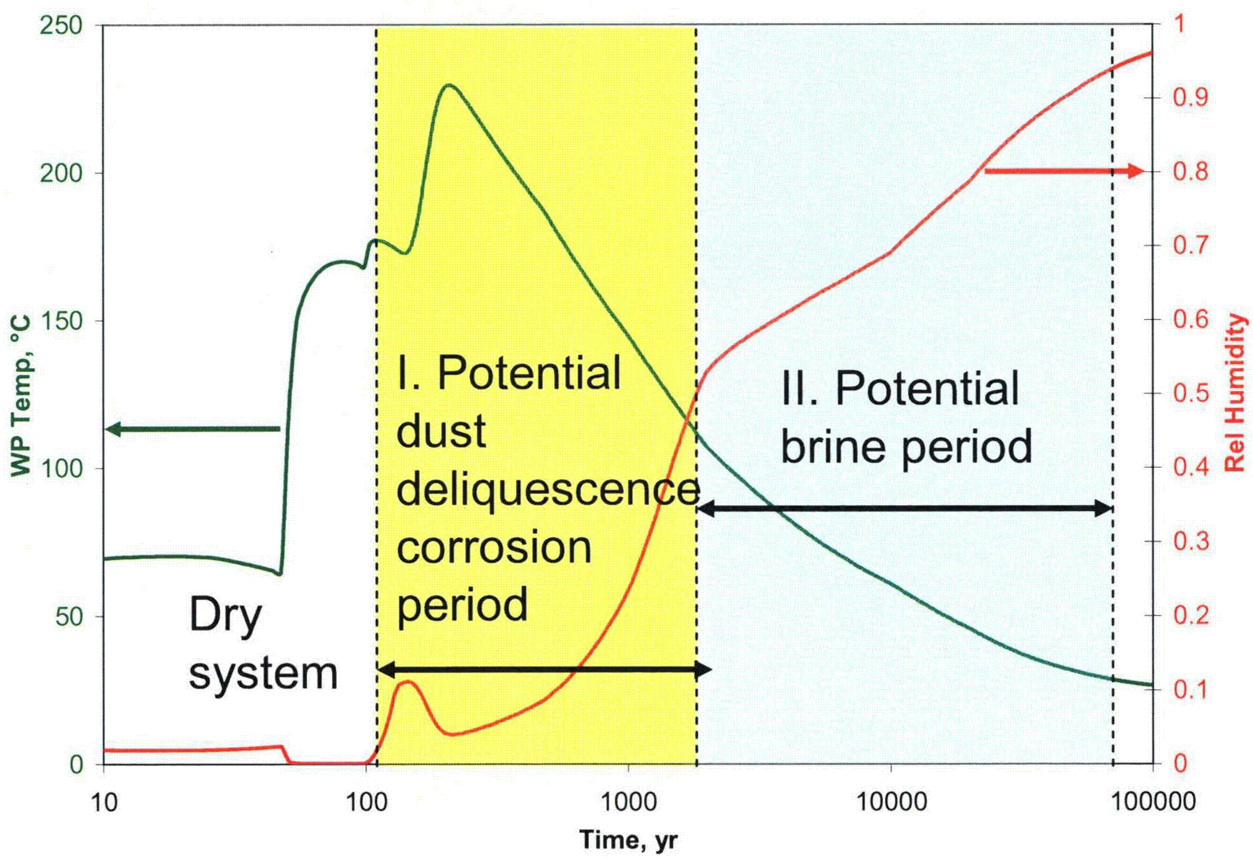
Engineered Barrier System

(DOE, 2002)



- Drip Shield – Titanium Alloys (Grades 7 and 29)
  (i) prevents contact of seepage water with WP
  (ii) prevents rockfall impact on WP
- Waste Package – Alloy 22 (Ni-22Cr-13Mo-3W-4Fe)
  (i) prevents water contact with waste form
  (ii) controls radionuclide release
- Good understanding of potential corrosion mechanisms, with residual uncertainties

WP Environment and Corrosion Modes



(After Pensado, 2006)

- Persistence of long-term passive film in general corrosion (I, II, and longer period)
- Dust deliquescence corrosion (I)
- Seepage water brines - crevice corrosion (II)
- Microbially influenced corrosion (II, and longer period)
- Hydrogen effects on Titanium (II, and longer period)

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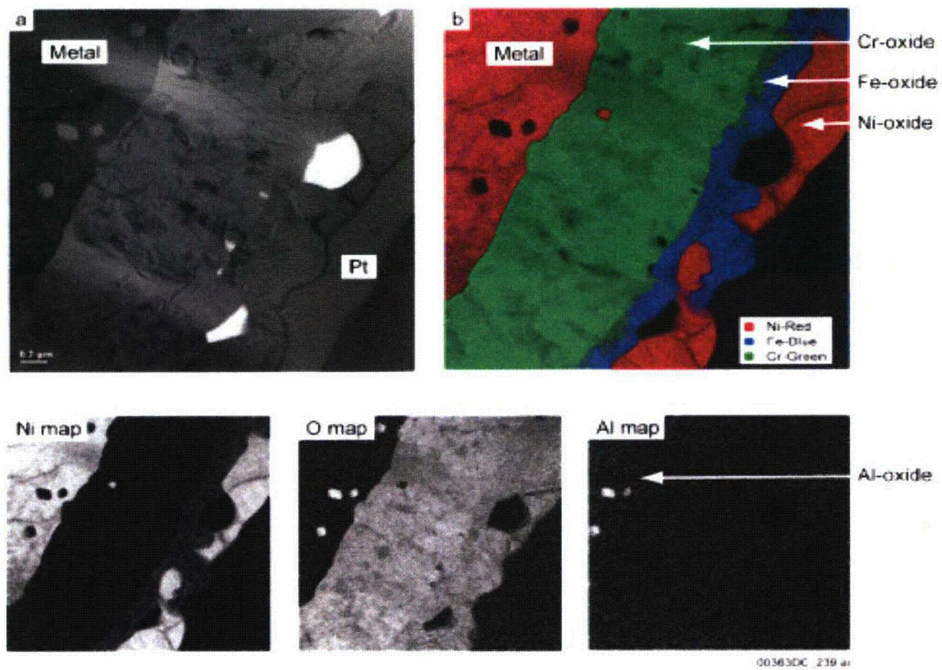
WP Environment and Corrosion Modes *(Contd.)*

Corrosion Modes Not Seen As Risk Significant

- Alloy 22:
  stress corrosion cracking, hydrogen embrittlement, galvanic corrosion, and dry oxidation

- Titanium alloys:
  localized corrosion (pitting and crevice corrosion), stress corrosion cracking, microbially influenced corrosion, galvanic corrosion, and dry oxidation

General Corrosion: Persistence of Passive Film



A Cross-Sectional View of a Solution Annealed Alloy 22 Substrate (Orme, 2005)

- **Uncertainties of passive film stability affect long-term general corrosion rates**
- **Passive film stability is primarily affected by changes in**
  - **chemical composition**
  - **microstructure**
  - **thickness**

General Corrosion: Persistence of Passive Film *(Condt.) Chemical Composition, Microstructure, and Thickness – Conformance of Chromium Oxide*

Models, analogue information, and limited laboratory data suggest that a chromium-rich oxide layer is responsible for the persistence of passive film (i.e., conformance).

- **Models: conformance by point defect model (PDM, Macdonald et al., 2004; Chao et al. and Lin et al., 1981; Urquidi-Macdonald and Macdonald, 1987); finite thickness of passive film (Urquidi-Macdonald and Macdonald, 2003), and minimal effects of potential void formation (Pensado et al., 2002)**

  - uncertainties: long-term changes in chemical compositions and microstructure

- **Long lifetime of natural analogue, nickel-iron meteorites (Sridhar and Cragnolino, 2002)**

  - uncertainties: applicability to Alloy 22 in the YM environment (e.g., role of chromium)

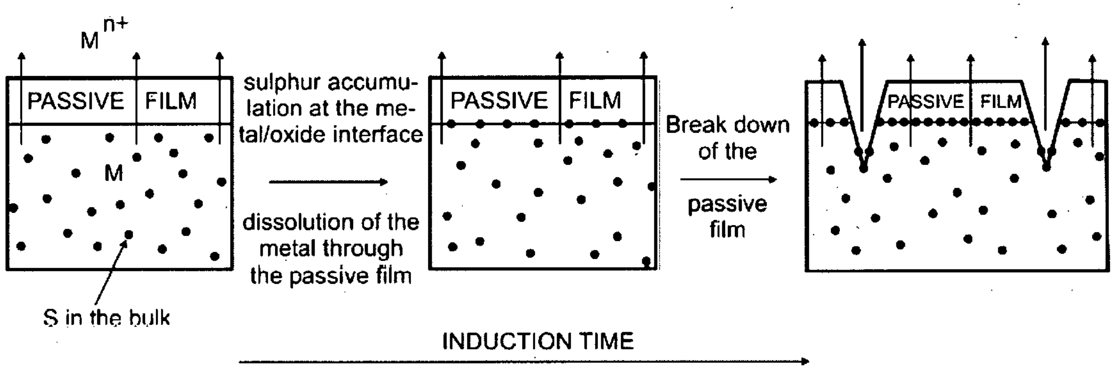
- **Limited laboratory data: decrease of general corrosion rates of Alloy 22 with time**

  - example uncertainties: properties of passive film under deliquescence conditions (Yang et al., 2007); effects of foreign deposits such as silica

General Corrosion: Persistence of Passive Film

(Contd.) Chemical Composition - Anodic Sulfur Segregation

Mechanism of the breakdown of the passive film induced by enrichment of sulfur at the metal-passive film interface (Marcus. 1995)



(Reprinted from "Sulfur-Assisted Corrosion Mechanisms and the Role of Alloyed elements" by P. Marcus in *Corrosion Mechanisms in Theory and Practice* edited by P. Marcus and J. Oudar, Marcel Dekker, Copyright (1995), with permission from Marcel Dekker)

Uncertainties

- Dissolution rate of segregated sulfur with molybdenum

- Repassivation rate in chloride solution with chromium and oxyanions

(Ahn, et al., 2007; Passarelli et al., 2005)

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Dust Deliquescence Corrosion

- Dust deliquescence corrosion is potentially important for approximately first 2000 years after closure.
- Dust on WP may form brines from deliquescent at elevated temperatures. Some deliquescent brines can induce general and crevice corrosion of Alloy 22.
- Brines formed from very corrosive salt mixtures (Na-K-Cl-NO_3) at elevated temperatures will more likely result in general corrosion rather than crevice corrosion:
  - CNWRA experiments indicate high general corrosion rates at elevated temperatures, on the order of 1µm/yr [0.04 mil/yr], only during an approximately 2000-year thermal period; and
  - Extent of corrosion depends on distribution of dust and duration of corrosive brine formation.
- Current uncertainties in deliquescence-induced corrosion rates result from extrapolating short-term experimental results to repository time scales.

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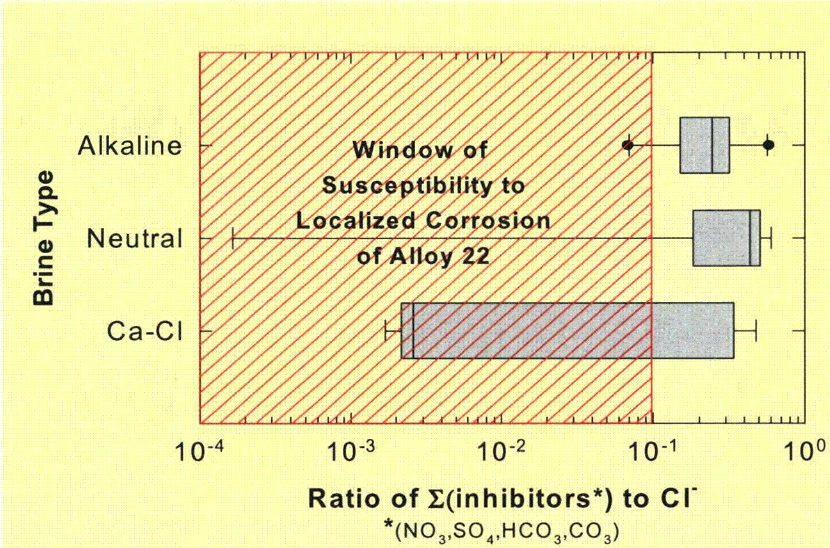
Seepage Water Brines: Crevice Corrosion

- Brines that form by evaporation of seepage waters are mostly benign to Alloy 22, but some compositions (less than approximately 10 percent) could initiate crevice corrosion for approximately 2,000 to several ten thousand year post closure.



Pabalan (2006); localize corrosion is crevice corrosion

  - Contact of seepage water may be prevented by DS.
  - The susceptibility of crevice corrosion decreases with decreasing temperature below approximately 110 °C.

- Uncertainties associated with likelihood of seepage-water brines to initiate crevice corrosion

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Seepage Water Brines: Crevice Corrosion *(Contd.)*

- In addition to temperature and water chemistry, tight contact environment is necessary to initiate crevice corrosion.

- Crevice area – tight area of buckled DS and WP under applied stress by rock falls and earthquakes
  - uncertainties in timing and extent of mechanical effects

- Weld area – more susceptible for crevice corrosion than base metal
  - uncertainties in weld characteristics from fabrication processes

- Crevice corrosion propagation was limited to some deep sites, after initial transient period, due to repassivation effects (He and Dunn, 2005)
  - uncertainties in long-term repassivation rate and under possible wide range of environment

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Additional Corrosion Processes

- **Microbially Influenced Corrosion (MIC)**

  - Models and limited laboratory data indicate low potential for MIC.

  - Uncertainties: Some MIC is difficult to detect (e.g., MIC-induced long-term pitting or crevice corrosion)

- **Hydrogen Effects in Titanium Alloys**

  - Preliminary analyses suggest some minor effects on long-term integrity of DS.

  - Uncertainties:
  (i) hydrogen sorption rates during titanium alloy corrosion; and
  (ii) hydrogen diffusion rates between dissimilar titanium metals (e.g., base metal and welds)

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Summary

- Long-term chemical or structural changes in passive film stability strongly affect uncertainties in Alloy 22 general corrosion rates.
- Current information indicates that crevice corrosion by dust deliquescence does not affect WP performance significantly.
- Crevice corrosion from seepage water (less than approximately 110 °C) requires tight crevices and aggressive brines (less than approximately 10 percent of expected seepage water compositions). Susceptibility decreases with decreasing temperature.
- Microbially influenced corrosion appears unlikely because of short induction time and no evidence of long-term pitting and crevice corrosion.
- Hydrogen effects on titanium alloy integrity appear to be low significance.
- Uncertainties in persistence of passive film appear more significant than uncertainties in other corrosion processes.
- Information from laboratory investigation, numerical models, and analog materials is available to support staffs review of corrosion processes.

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Dissolution Processes for Commercial Spent Nuclear Fuels in Repository Conditions

Presented by T. Ahn, NRC

Team Leads: T. Ahn, NRC, and D. Pickett, Center for Nuclear Waste Regulatory Analyses (CNWRA)

Other Contributor: S. Mohanty, CNWRA

182nd Meeting of the Advisory Committee on Nuclear Waste and Materials (ACNW&M), September 18 – 20, 2007

U.S. Nuclear Regulatory Commission, Rockville, MD

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Outline

- Purpose
- Commercial Spent Nuclear Fuel (CSNF) Dissolution
- Principal Factors for Matrix Dissolution
- In-Package Water Chemistry
- Condition of CSNF before Water Contact
- Groundwater Contact Modes for CSNF
- Summary
- References

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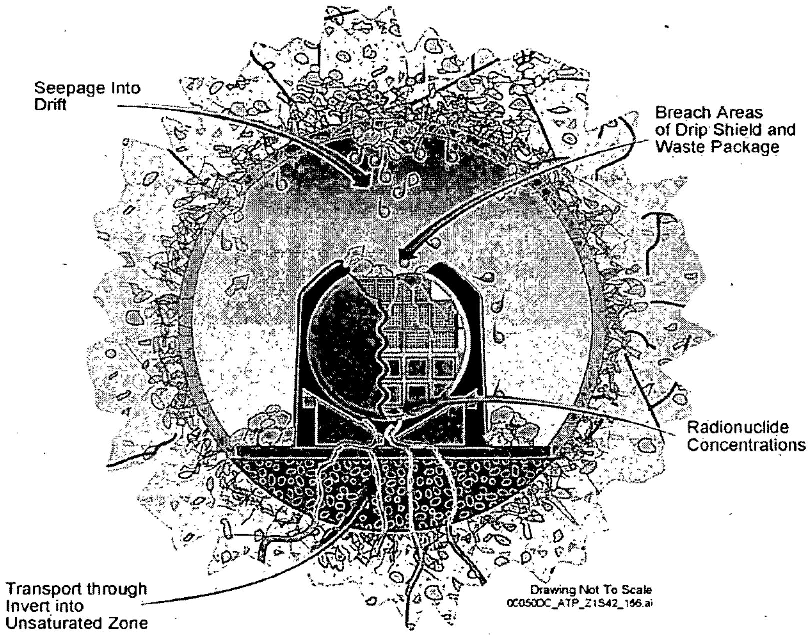
Purpose

- Present an overview of key processes for the dissolution of commercial spent nuclear fuels at conditions representative of the potential Yucca Mountain (YM) repository

- Discuss the significance of uncertainties in important processes that affect CSNF dissolution models
  - In-package water chemistry
  - CSNF characteristics
  - Groundwater contact modes

CSNF Dissolution

Matrix (i.e., irradiated UO_2) Dissolution – conqruent release of Tc-99 and I-129



(DOE, 2002)

Principal Factors for Matrix Dissolution

Reaction products of UO_2 with H_2O depend on electrochemical conditions of UO_2 for dissolution and hydrolysis of dissolved species.

(1) In-Package Water Chemistry

- Concentrations of carbonate/bicarbonate ions, oxygen, iron and other cations
- pH
- Temperature

(2) Condition of CSNF before Water Contact

- Extent of pre-oxidation/hydration
- Grain-boundary characteristics

(3) Groundwater Contact Modes for CSNF

- Seepage rate
- Immersion versus drip
- Extent of cladding protection

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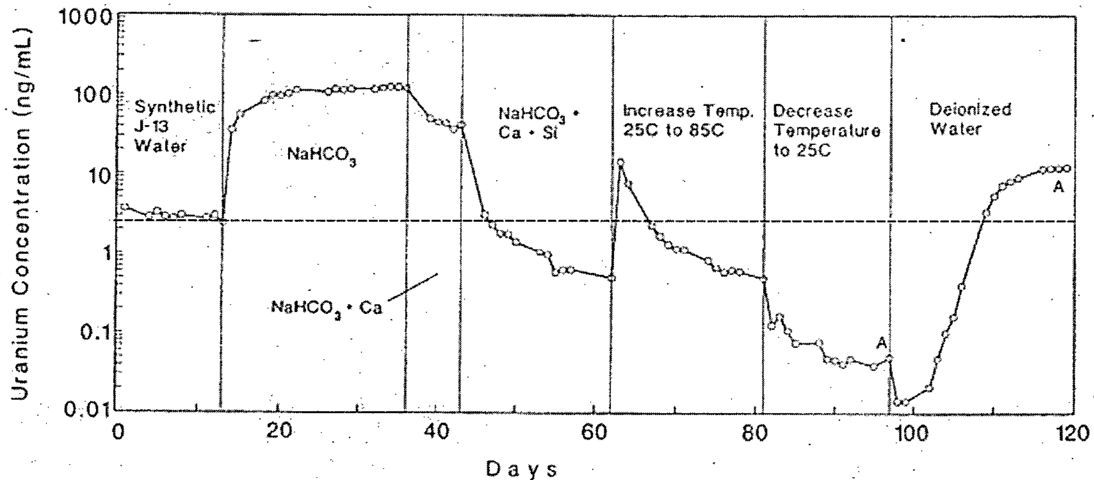
In-Package Water Chemistry
Cation Effects



(after Wilson and Gray, 1990)

- Ca and Si (Ca^{2+} and SiO_4^{4-}) tend to decrease the matrix dissolution rates by two orders of magnitude or more at 25 °C (77 °F) compared to those in carbonate solutions of $2x10^{-4} – 2x10^{-2}$ M.
- Primary uncertainties
  - Competing effects of cations and low pH
  - Rates of cation depletion
  - Formation of Schoepite inhibiting dissolution

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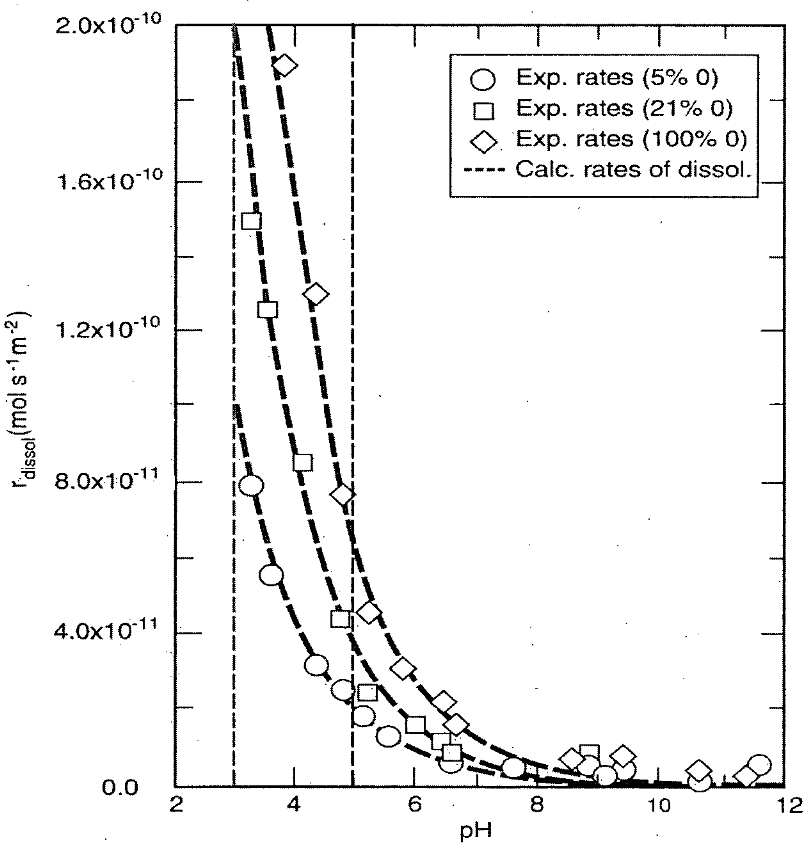
In-Package Water Chemistry *(Contd.)*

pH Effects



Rates of UO_2 dissolution as a function of pH and oxygen concentration [mol/s-m ^{2} : 6.5x10^{-4} mol/s-in^2] (after Torrero et al., 1997)

- **Data under oxidizing conditions (Torrero, et al., 1997)**
  - **Matrix dissolution rates increase by a factor of 4 – >10 at pH ~ 3, compared to pH below ~ 5.**
- **Metallic cations (e.g., Cr) from corrosion of waste package internals can lower pH in seepage water.**
- **Primary uncertainty is magnitude of pH variability.**

In-Package Water Chemistry *(Contd.)*

Effects of Oxygen and Iron Concentration

- Corroding steel components may decrease local oxygen concentration.
  - Decrease in the matrix dissolution rates by more than 10x in SIMFUEL tests in NaCl solution at ambient temperature (Quiñones, et al., 2001)

- Oxygen in an air-buffered repository environment is sufficiently abundant to offset the production of oxidants by radiolysis.

Effects of Temperature

- CSNF dissolution rate is most sensitive to variations in temperature.
- The activation energy for dissolution varies depending on test conditions (Shoesmith, 2000): 0 – 47 kJ/mol (11.2 kcal/mol).
- Activation energy of 24 – 33 kJ/mol (5.7 – 7.9 kcal/mol) for Tc-99 and I-129 from CSNF immersion tests (Wilson, 1990)

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Condition of CSNF before Water Contact

Pre-oxidation/Hydration

Uncertainties in the timing and extent of pre-oxidation and hydration processes:

- **$UO_{2.4}$ or U_3O_8 dissolves similarly with unoxidized UO_2. However, UO_3 xH_2O (x = 0.8, 2) dissolves 10 – 20 times faster than unhydrated oxides.**

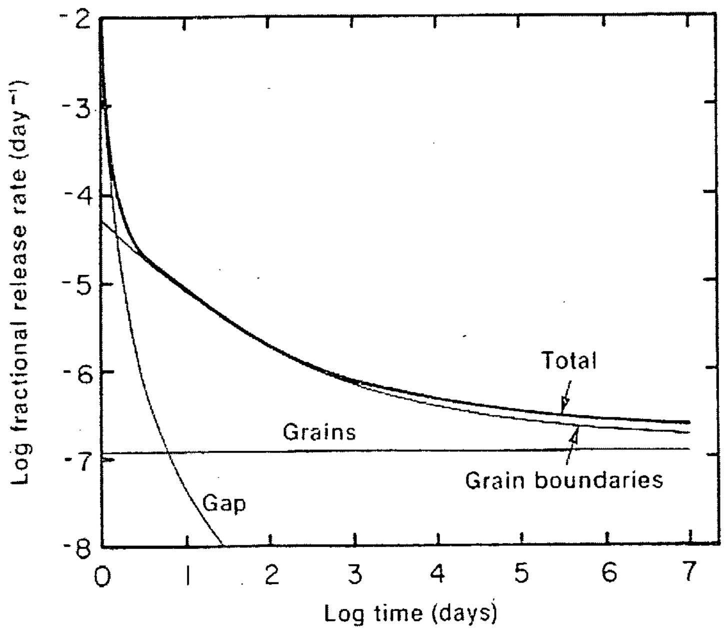
- **Oxidation to $UO_{2.4}$ tends to open up more grain boundaries, potentially increasing exposed surface area.**
  (i) Dissolution rates may increase for CSNF in initially failed WP if relative humidity is less than ~50 percent (Gray, 1997) for the oxidation during the early period in the initially failed WP.
  (ii) Grain boundary openings would be, however, masked with alteration (i.e., precipitated) products of dissolved uranium.

- **Oxidation to U_3O_8, or UO_3 xH_2O creates inter- or trans-granular fractures that increase exposed surface area by as much as 2 – 3 orders of magnitude - temperatures above 250 °C (482 °F) are necessary to form U_3O_8.**

- **Vapor hydration or aqueous dissolution may further weaken deeper grain boundaries. Mechanical impacts (e.g., seismicity) can result in additional grain breakage.**

Condition of CSNF before Water Contact

(Contd.) Grain Boundary Inventory

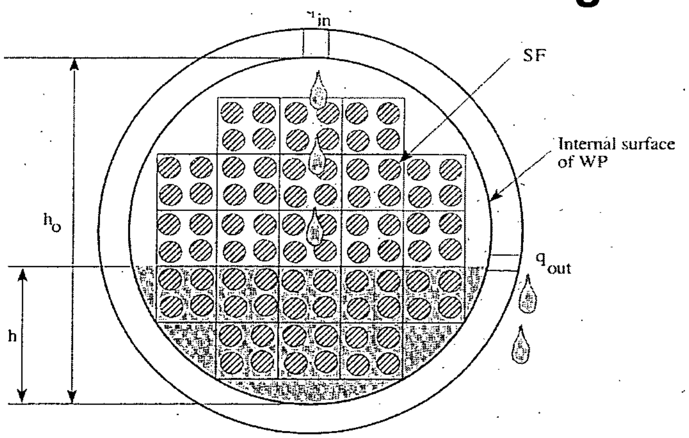


Schematic of three stages in fission product release from used CSNF (Johnson et al., 1985)

- **Radionuclide (RN) releases from grain boundaries and matrix dissolution.**
- **Tests with fragments (i.e., particles) under dripping, flow-through or immersion that show substantial amounts of grain-boundary RN in the results were used to determine matrix dissolution rates (Tc-99, I-129, Cs-137, or Sr-90) (OCRWM, 2003).**
- **Uncertainties in the magnitude and extent of long-term RN release from grain boundaries, relative to matrix**

Groundwater Contact Modes for CSNF

Perforations/Cracks on Waste Package
- uniform corrosion
- localized corrosion
- stress corrosion cracking



Schematic of bathtub model with incoming and outgoing water conduits (Mohanty et al., 2002)

- Increasing drip rates by 10x resulted in an increase in CSNF dissolution rate by at least 10x (Cunnane, 1999).
- Drip rates may be more than 10x lower than modeled repository seepage rate.
- Uncertainties in extrapolating tests to repository conditions (e.g., drip rate)

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Groundwater Contact Modes for CSNF *(Contd.)*

Failed Cladding Protection

- Immersion tests for slit (~0.015 cm [0.006 in] diameter x ~2.54 cm [1 in] length)- or hole (~ 0.02 cm [0.008 in] diameter)- CSNF cladding defects on release rate in J-13 Well Water at 85 ºC (185 ºF) (Wilson, 1990)

  - Relative to unclad CSNF, release decreased by a factor of ~ 140 for Tc-99, ~700,000 for I-129, and ~65 for Sr-90

- Drip tests of partial clad rod segments (OCRWM, 2003)

  - Tc-99 release, 2.6 x 10^{-8} fraction per day

- No decrease observed in release with 50 percent CSNF surface area exposed in immersion tests (Forsyth, 1997; Forsyth and Wermer, 1985; Stroes-Gascoyne, 1985)

- Significant uncertainties in the times and extent of cladding defects

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Summary

- Dissolution rates are most sensitive to variations in temperatures.
- Ca and Si ions could decrease CSNF dissolution rates more than an order of magnitude.
- Release from the grain boundary/gap inventory is substantial component of effective release rates, by a factor of 2 – 10 of the long-term matrix dissolution rates.
- Other factors that potentially decrease dissolution or release rates are low drip-rates of seepage water, small opening in cladding, and presence of iron compounds.
- Other factors that potentially increase dissolution rates are low pH, and pre-oxidation and hydration of CSNF before water contact.
- A range of information in analog, primarily from laboratory experiments, to support staff review of CNSF dissolution models.

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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 a license application for a geological repository at Yucca Mountain. This presentation describes work performed by the Center for Nuclear Waste Regulatory Analyses (CNWRA) for the NRC under contract number NRC-02-02-012. The activities reported here were performed on behalf of the NRC office of Nuclear Material Safety and Safeguards, Division of High Level Waste Repository Safety. This presentation is an independent product of the CNWRA and does not necessarily reflect the view or regulatory position of the NRC.

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