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Nuclear Regulatory Commission*

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**SOME IMPORTANT CONSIDERATIONS  
IN WASTE FORM SOURCE-TERM MODELING**

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**For presentation at Workshop on Alternative Models  
and Interpretations for the Waste Form  
Degradation and Radionuclide Mobilization  
Expert Elicitation (WFEE) Project**

**December 15-16, 1997  
San Francisco, CA**

## OBSERVED DISSOLUTION RATE OF PRIMARY PHASE OF BARE SPENT FUEL

- at Room Temperature:

  - 9 mg/m<sup>2</sup>-day in J-13 well water

  - 72 mg/m<sup>2</sup>-day in carbonate water

- at 90 C:

  - 77 mg/m<sup>2</sup>-day in J-13 well water

All are based on geometric surface area and immersion/flow-through tests; data at 90 C is close to ANL results of drip tests [Gray, 1997] (ref.: ~ 7 mg/m<sup>2</sup>-day for UO<sub>2</sub>). The consensus (of the technical community) up to now is that the environment is oxidizing from oxygen inflow [Stockman, the first of this WFEE meeting, 1997] and from alpha/beta radiolysis.

- Life Time of 1 mm Fragment:

Extrapolating linearly, the above data correspond to lifetimes of ~ 3040, ~ 380, and ~ 360 years respectively. Lifetime assuming carbonate water at 90 C is ~ 45 years. These do not appear to be realistic values for use in source-term modeling

- Based on these rates, most soluble radionuclides will be released quickly from congruent releases.

CONSIDERATIONS RELATED TO WET FRACTION  
OF SPENT FUEL IN THE REPOSITORY

- Practically no experimental data are available.
- Codell: probabilistic analysis including geosphere, near field and waste packages (next presentation in this meeting)
- Example factors to be considered in calculating decreased dissolution rates:

The area fraction of pits on the container surface can vary from 1 to  $\sim 10^9$ , based on review of literature data on measured pit density and pit size [Ahn, 1994].

The volume of a container is  $\sim 10^7 \text{ cm}^3$ ; 21 PWR spent fuel assemblies will occupy 0.05  $\sim$  0.1 fraction.

Pits are likely to be clogged with corrosion products. Although the inside in growing pits may be severely acidic and aqueous [Szkarska-Smialowska, 1986], through-wall pits are likely to have porous solid corrosion products. Solid corrosion products were shown in pits and crevices during steel corrosion [Raman and Nasrazadani, 1990].

Assuming water can get to all fuel fragments, 1 liter of water per container will cover all 1 mm spent fuel particle with thickness  $\sim 4 \mu\text{m}$  for 21 PWR spent fuel assemblies. At relative humidity 80 %, 5  $\sim$  10 monolayers of water will be condensed at outdoor temperatures [Leygraf, 1995]. Therefore,  $\sim 4 \mu\text{m}$  water is likely to be aqueous.

100 % of bare spent-fuel surface was exposed to humid air in ANL tests. However, the release is  $\sim 1$  % of the equivalent release from high drip rate tests [Bates, the first of this WFEE meeting, 1997]

CLADDING PROTECTION MODES  
AND CONSIDERATIONS RELATED TO CLADDING FAILURE MECHANISMS

(1) Protection Modes

Intact

Limited water intrusion through pits and cracks

Limited radionuclide release through pits and cracks

(2) Localized Corrosion

Pitting<sub>3</sub> can take place in acidic environments (pH < 4.5) with Fe<sup>3+</sup> from container corrosion (~ ppm) [McGuire, 1997]. At neutral pH, high Cl<sup>-</sup> concentration can also cause pitting.

(3) Gamma Radiolysis on the Cladding Surface

fixation of nitrogen

$$N = 2 C_0 R [1 - \exp(-1.45 \times 10^{-5} \text{ GDt})] \text{ [Burns, et al., 1982]}$$

N: concentration of HNO<sub>3</sub> in water

C<sub>0</sub>: initial concentration of N<sub>2</sub> in air

R: ratio of air volume to water volume

G: yield

D: dose rate

t: time

For time intervals of water drip, (1 year and 3.5 days), 4 μm film thickness, ~ 50 rads/hour within 5000 years (from McCoy, 1997), and G = 1.9 [Burns et al., 1982], water film is likely to be acidic.

(4) Creep

Consensus (of the technical community) up to now: one % strain assumption is too conservative. (4 ~ 10) % (?)

Diffusion-controlled cavity growth (DCCG) has not been proven and is likely to be a conservative assumption.

(5) Hydrogen Effects

Delayed Hydride Cracking (DHC) is based on hydride precipitation at crack tip. There is a threshold toughness as a function of bulk hydrogen concentration (figure) [Shi and Puls, 1994].

Hydride reorientation requires ~ 290 C for hydride dissolution (~ 200 ppm solubility limit at 400 C) and ~ 95 MPa [Chan, 1996]. Hydrogen embrittlement with ~ 1000 ppm hydrogen decreases toughness by hydride growth or reorientation. An example is that the toughness decreases from 20 MPa m<sup>1/2</sup> to 2 MPa m<sup>1/2</sup> as cooling rates decrease (as in the repository) [Chan, 1996].

(6) Splitting by Matrix Volume Expansion

Volume expansion: 1.36 for U<sub>3</sub>O<sub>8</sub> from UO<sub>2</sub>; (2.67 ~ 5.55) for secondary minerals from uraninite [Lichtner, 1994]. Based on linear elasticity, the stress associated with volume expansion [Ahn, 1996] exceeds the cladding strength.

The altered phase is likely to be powdery and may not push adjacent rods at ~0.5 cm distance. It is considered that dissolved uranium may diffuse (or drip) out of cladding prior to secondary mineral formation. Even in the humid environment, ANL reports that water still condenses and drips.

(7) Stress Corrosion Cracking (SCC), Pellet Cladding Interaction (PCI), and Mechanical Properties

There is no evidence for iodine-induced SCC for PCI failure; the PCI failure is likely to be related to hydrogen or oxygen effects; and dynamic toughness for PCI is (5 ~ 10) MPa m<sup>1/2</sup> [Chung, 1997].

Fracture Toughness as a Function of Hydrogen in Solution [Shi and Puls, 1994]

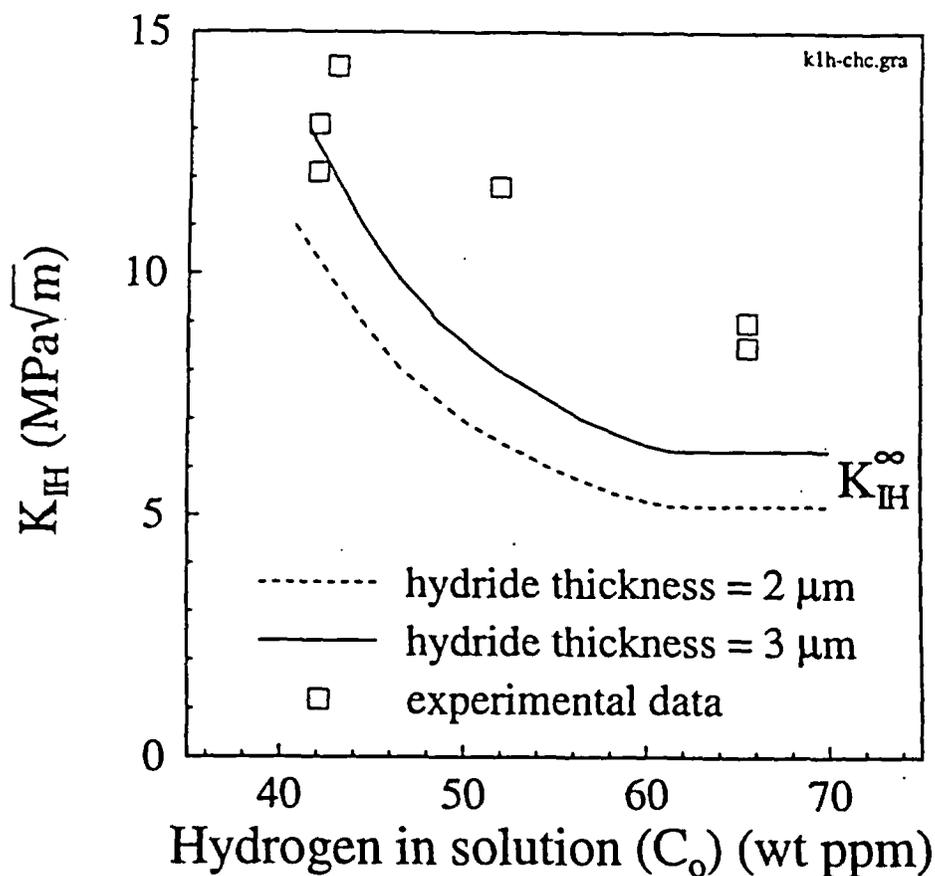


Table A-1 Engineered Barriers Performance Factors

Engineered Barrier Subsystem	Absolute Performance Factor <sup>1</sup>		Environment at which Calculations Performed <sup>2</sup>	Environmental Parameters which Influence Barrier <sup>3</sup>	Operational Considerations
	10k yrs	1M yrs			
Cladding	20	50	0.3 mm/yr	T; stress	T<350°C
Waste Package	-1	-1	0.3 mm/yr	q(flux), RH, T	---
Galvanic Protection	30	20	0.3 mm/yr	q(flux), RH, T	none
Drip Shield	>1	<1.5 <sup>4</sup>	0.3, 1.25 mm/yr	RH; T	need long life
Backfill	>1	15	0.3, 1.25 mm/yr	q(flux), RH, T	emplacement, high T
Backfill +galvanic	>1	20	0.3, 1.25 mm/yr	q(flux), RH, T	emplacement, high T
Richards Barrier Backfill	>1	<1.5 <sup>4</sup>	0.3, 1.25 mm/yr	q(flux), RH, T	not emplaceable
Repository Configuration	>1	-3	0.3 mm/yr	q(flux), $\phi$ , T	high T; logistics

<sup>1</sup> Absolute performance is based on dividing the estimated peak dose at 30 km for the ACD base case by the predicted doses with the respective barrier included.

<sup>2</sup> Current measurements indicate a percolation flux in TSw2 of 1 to 10 mm/yr with 5 to 7 mm/yr most likely.

<sup>3</sup> Environment includes such issues as temperature, RH, water chemistry, and percolation flux.

<sup>4</sup> This value could be larger if the barrier is very long lived (>7x10<sup>5</sup> years).

Limiting Factors to Determine Radionuclide Releases from Pinhole (~0.02 cm Diameter) or from Slit- (~0.015 Diameter x ~2.54 cm Length) Defected Cladding in J-13 Well Water. Ratios were calculated with respect to radionuclide releases from bare spent-fuel matrices. Data from Wilson [1990]

Radionuclide	Limiting Factor
Np-237	~70
Tc-99	~460
I-129	~7x10 <sup>5</sup>
Sr-90	~65
Pu-(239+240)	~7x10 <sup>4</sup>
Am-241	~3x10 <sup>5</sup>

\*: Data were selected for Cycle 2 and Cycle 3. The Cycle 1 is the first semi-static immersion test including the release of fuel grains and of gap inventory. Cycle 2 tests were retests of first-cycled samples that had been placed in new leachant, after removing the altered layer. In general, tests after Cycle 2 showed little gap inventory if it is not of a large amount initially. Detection limits of radionuclides were used in the calculation when hole-defected or slit-defected samples did not release detectable radionuclides.