Long-Term Materials Behavior at the Potential Yucca Mountain Repository

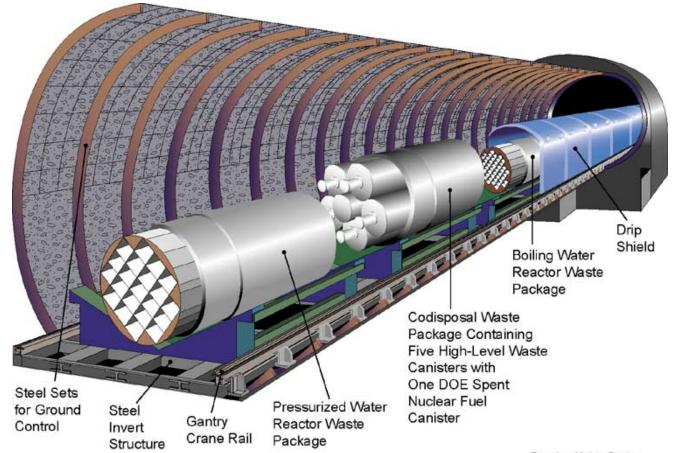
Tae M. Ahn U.S. Nuclear Regulatory Commission Washington, D.C. 20555-0001, U.S.A.

Presented to

Workshop on Design and Assessment of Radioactive Waste Packages in European Commission, Joint Research Center, Institute for Energy, Petten, The Netherlands October 5 – 7, 2005 **Disclaimer:**

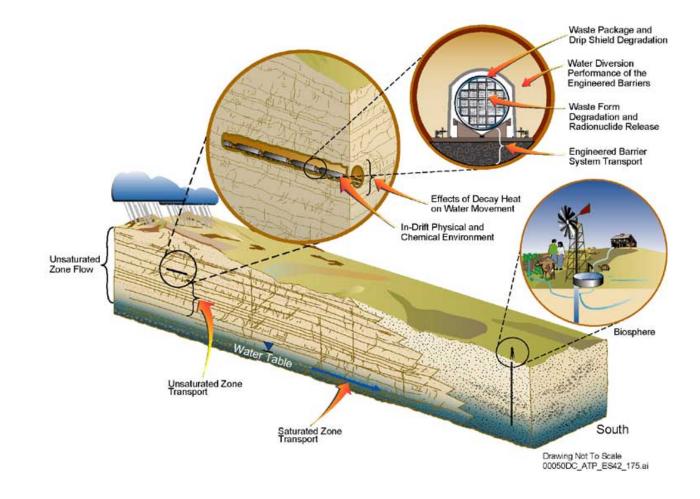
The NRC staff views expressed herein are preliminary and do not constitute a final judgment or determination of the matters addressed or of the acceptability of a license application for a geological repository at Yucca Mountain.

Schematic Illustration of the Emplacement Drift with Cutaway Views of Different Waste Packages (DOE, 2002)

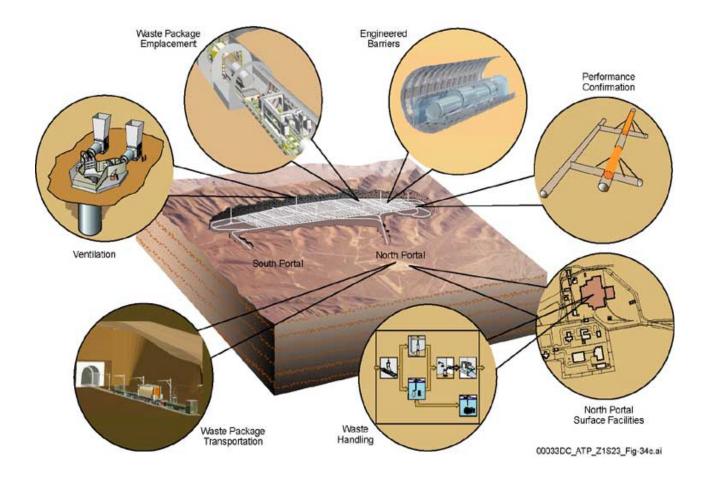


Drawing Not to Scale 00022DC_ATP_Z1S30-02a.ai

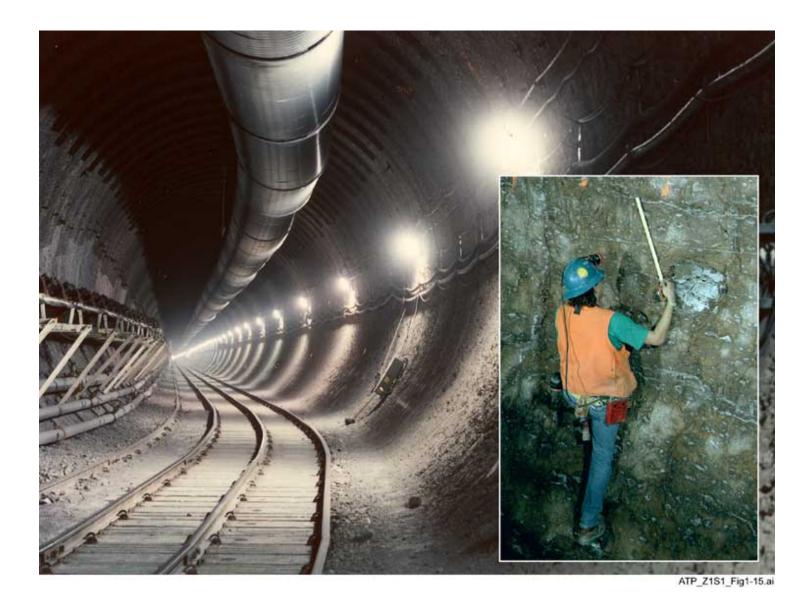
Schematic Illustration of the Ten General Processes Considered and Modeled for Total System Performance Assessment (DOE, 2002)



Proposed Monitored Geologic Repository Facilities at Yucca Mountain (DOE, 2002)



View Looking Down Exploratory Studies Facility (DOE, 2002)



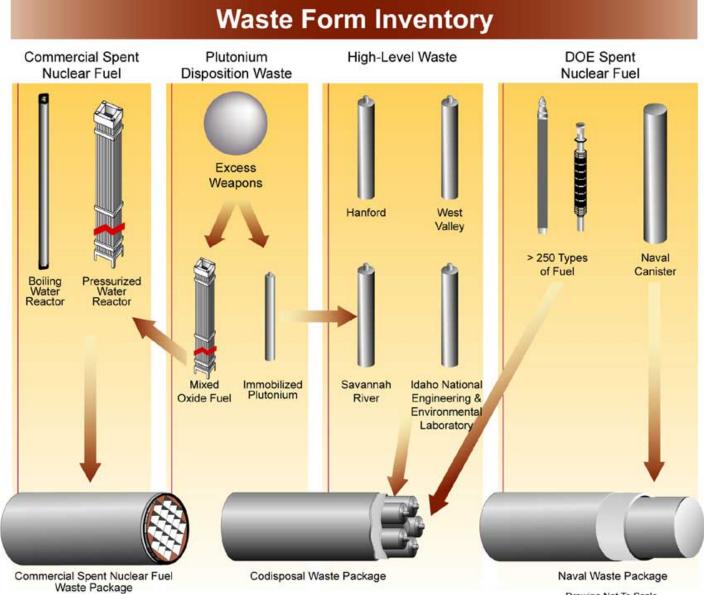
Risk-Informed Performance-Based Licensing

- Pre-Closure Period
- Post-Closure Period

• Performance Objectives: 10 CFR Part 63

Tools for Risk Assessments

- Pre-Closure Safety Analysis (PCSA)
- Total-system Performance Assessment (TPA)
- Screening of Event Sequences and Scenarios
- Models
- Bases for Model Supports



Drawing Not To Scale 00022DC-ATP-Z1S30-03a.ai

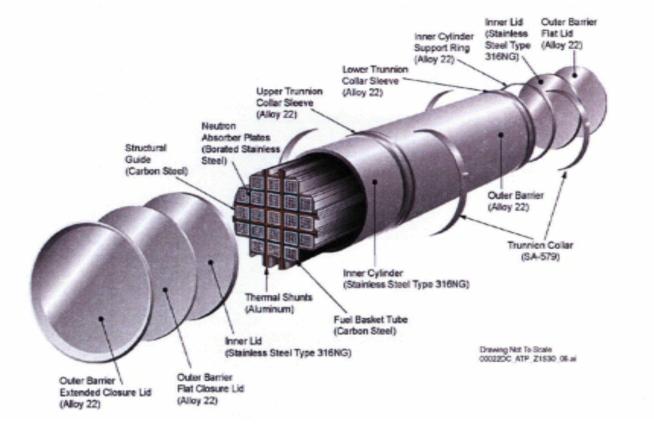
Waste Package (Alloy 22, Ni-22Cr-13.5Mo-3W-4Fe)

- Long-Term Passivity in Uniform Corrosion
- Localized Corrosion
- Stress Corrosion Cracking
- Long-Term Phase Stability
- Mechanical Failure
- Fabrication and Reliability

Drip Shields (Ti-7 and Ti-24, Pd addition)

- Long-Term Passivity in Uniform Corrosion
- Fluoride Uniform Corrosion
- Hydride Embrittlement
- Stress Corrosion Cracking
- Creep

Pressurized Water Reactor Absorber Plate Waste Package Design (DOE, 2002)



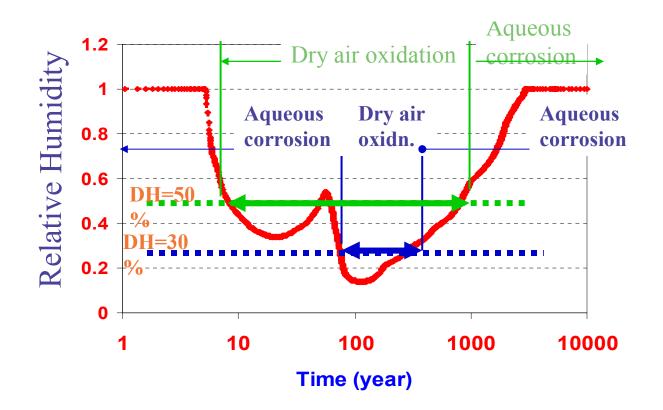
Spent Nuclear Fuel

- Matrix Dissolution- Tc and I
- Secondary Phase Formation and Solubility- Np
- Matrix Oxidation Pu and Am
- Colloid Formation Pu
- Hydride Reorientation

High-Level Waste Glass

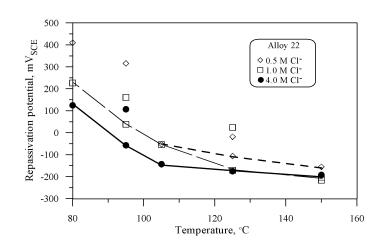
- Leaching
- Colloid Formation Pu

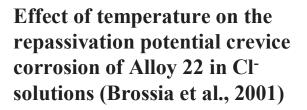
Environmental Conditions



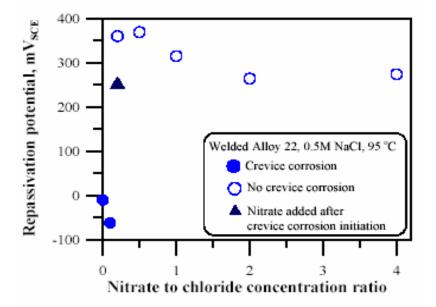
Deliquescence Humidity and Aqueous or Dry Air Corrosion (Yang, 2001)

Localized Corrosion





$$(100 \ ^{\circ}C = 212 \ ^{\circ}F)$$



(Dunn et al., 2003)

Uniform Corrosion

- Data from DOE, CNWRA, industries and international community (e.g., long-term German tests in rocksalts) point out similarities of uniform corrosion rates. A container lifetime of greater than 10,000 years can be estimated.
- A long-term integrity of passive film is suggested by various models for point defects, chemistry segregation, and passive film growth.
- Analogue studies suggests that modern electrochemical theories for corrosion may explain the analogue observation: void formation, stoichiometric dissolution of meteorites and josephinite, possible passivity of Indian Pillar, and long-term passivity of carbon and stainless steel over half a century (Sridhar and Cragnolino, 2002).

Stress Corrosion Cracking (SCC)

Test conditions and results for the testing of Alloy 22 DCB specimens

Specimen ID	Test Solution and	Potential	Duration	Results
(Orientation)	Temperature	(mV _{SCE})	(hr)	
22-1(T-L)	0.9 molal Cl ⁻	-330 to	9,264	No SCC
	(5% NaCl), pH 2.7	-310 (OC)	(386 days)	
	90 °C, N ₂ deaerated			
22-2(T-L)	14.0 molal Cl ⁻	-280 to	9,264	No SCC
	(40% MgCl ₂), 110°C	-260 (OC)	(386 days)	Grain Boundary Attack
22-7(S-L)	14.0 molal Cl ⁻	-270 to	9,264	No SCC
	(40% MgCl ₂), 110°C	-250 (OC)	(386 days)	Secondary Cracking

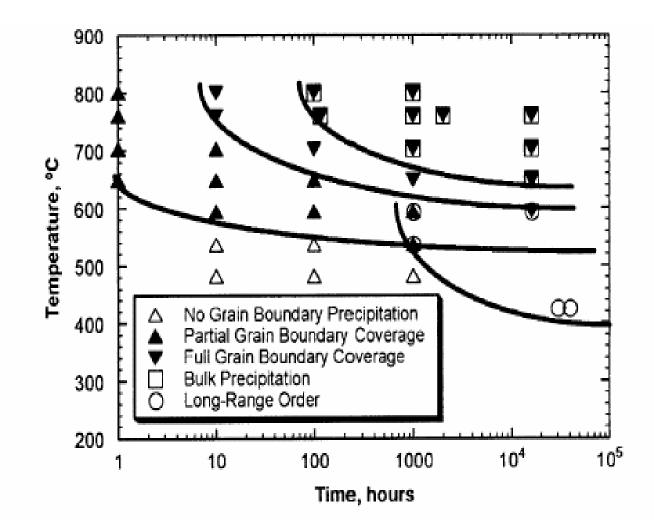
T-L – Transverse-Longitudinal; S-L – Short transverse-Longitudinal;

OC – Open-Circuit

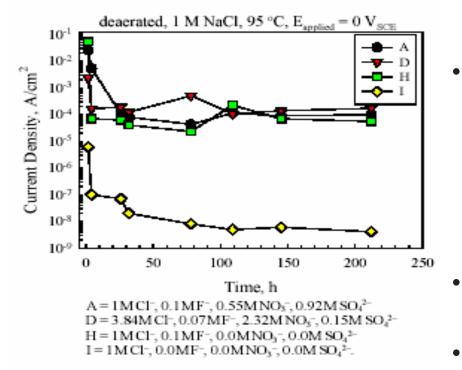
•No crack growth was observed at $K_I = 32.7 \text{ MPa} \cdot \text{m}^{1/2}$ after 386 days, implying a detection limit of 3 x10⁻¹³ m/s

•It appears that $E_{corr} \le E_{SCC}$ and/or $K_I \le K_{Iscc}$

Conversion: 32.7 MPa m^{1/2} (29.7 ksi in^{1/2}), 3x10⁻¹³m/s (9.84x10⁻¹³ ft/s) (Cragnolino, 2003) Time-Temperature-Transformation Diagram for Alloy 22 Base Metal (DOE, 2000)



Ti-7 Corrosion



Effects of groundwater anions on fluorideinduced corrosion of T-7 (Brossia et al., 2001)

- DOE uses weight loss measurements of titanium in fluoride concentrations
 - No fluoride, $[F^-] = 7.37 \times 10^{-4} M$, and $[F^-] = 7.37 \times 10^{-2} M$
- General corrosion rate determined to be 3.25 x 10⁻⁴ mm/yr
- No enhanced corrosion by fluoride observed by DOE tests.

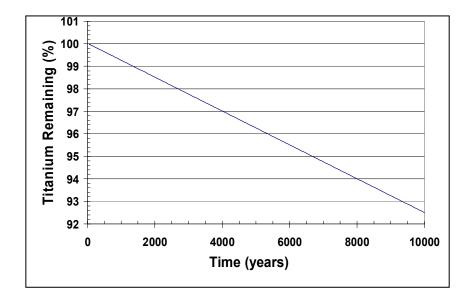
 $(1 \text{ A/cm}^2 = 1.55 \times 10^{-1} \text{ A/in}^2)$

 $1 \text{ mm/yr} = 3.94 \times 10^{-2} \text{ in/yr}$

Ti-7 Corrosion

 Most common products of titanium-fluoride reaction are TiF₆²⁻ and TiF₄

 Is there sufficient water for complete corrosion of drip shield?

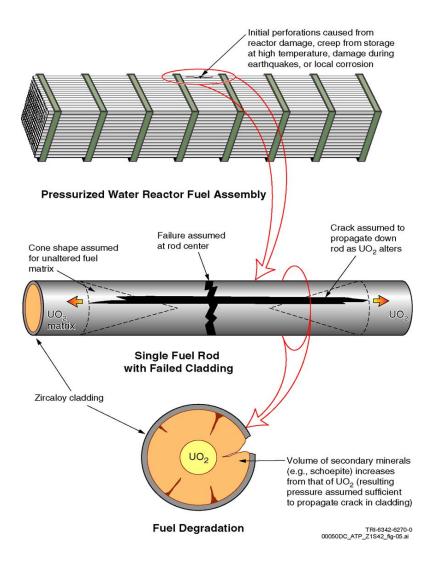


Cumulative reduction of drip shield titanium from corrosion by fluoride at average influx (Lin et al., 2003)

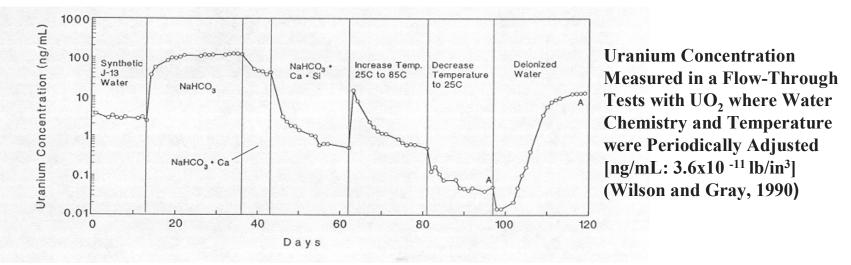
Low-Temperature Drip Shield Creep (Neuberger et al., 2002)

 Expected drip shield creep after rockfall loadings near material yield stress while drift operating temperatures are approximately 100 – 150° C

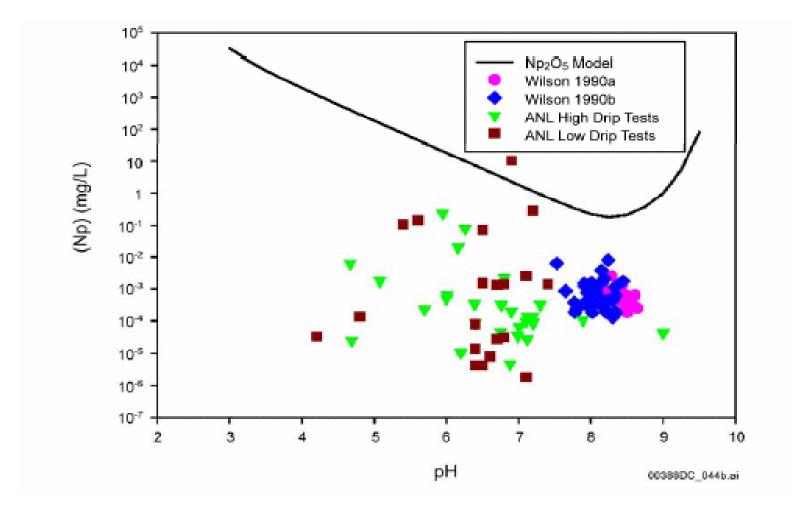
Conceptual Model of Commercial Spent Nuclear Fuel Cladding Degradation (DOE, 2002)



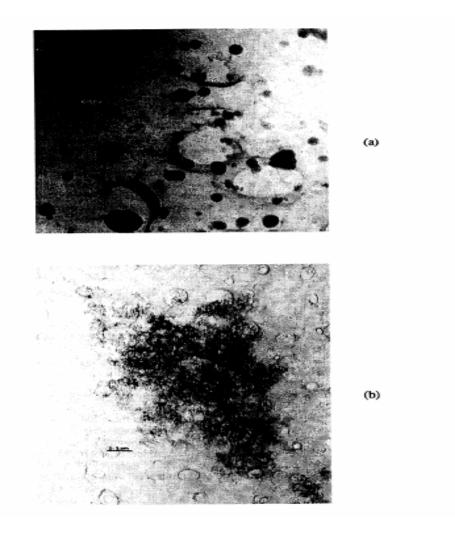
Environmental In-Package Chemistry Cation Effects



- Ca and Si (Ca²⁺ and SiO₄⁴⁻) tend to decrease SNF dissolution rates by as much as or more than two orders of magnitude at 25 °C (77 °F) compared with those in carbonate solutions of (2x10⁻⁴ - 2x10⁻² M).
- Carbonates and low pH ease the inhibition effects of these cations. However, repository relevant solutions did not show the effects (Table).
- These ions may be depleted in drip tests. However, Schoepite formed instead could impede the oxygen transport to the bare SNF resulting in the decrease of the dissolution rates.



Transmission Electron Microscopy (TEM) Micrographs of Particulate Material Isolated on a Holey Carbon TEM Grid: (a) Colloids Formed from Solution and (b) Material in Liquid Spalled from the Glass Surface (Bates et al., 1992)

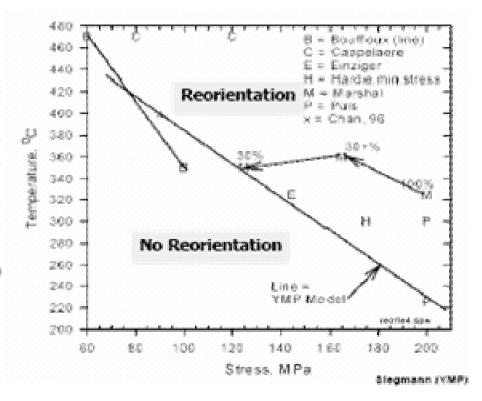


Dry Oxidation of Spent Fuel Matrix

- Weight Changes of Oxidized Bare Fragments of Turkey Point Fuel at Temperatures from 250° to 360° C (Einziger et al., 1992)
- At temperatures above 280° C, the plateau at O/M = 2.4 is very short and the oxidation proceeds rapidly to U_3O_8 .

Hydride Reorientation – Creep Tests

- Radial hydrides, as little as 40 wppm, can significantly degrade cladding's mechanical properties. (Marshall)
- Stress, temperature, cool-down rate, microstructure, H content, etc., all play important roles. (Einziger)
 - Threshold hoop stress for 400°C is ~ 100 MPa.





Metal Fabrication Reliability (Jain et al., 2003)

	Table 4-1. Ob:	served Weld F	law Frequenci	es*†	
Vessel	Location	Size of Cracks	Weld Volume	Number of Flaws	Flaws/m ³ [flaws/ft ⁸]
Pressure Vessel Research User Facility	Near Surface Zone {25 mm [0.98 in]}	< 3 mm [< 0.12 in]	0.014 m³ [0.49 ft³]	191	13,571 [384.3]
	Near Surface Zone {25 mm [0.98 in]}	> 3 mm [> 0.12 in]	0.014 m ³ [0.49 ft ³]	13	929 [26.3]
	Remaining thickness	< 5 mm [< 0.20 in]	0.20 m ³ [0.7 ft ³]	653	3,625 [102.6]
	Remaining thickness	> 5 mm [> 0.20 in]	0.20 m ³ [0.7 ft ³]	27	135 [3.8]
Shoreham Reactor Pressure Vessel	Inner 25 mm [0.98 in] surface	< 4 mm [< 0.16 in]	0.0226 m ³ [0.8 ft ³]	459	20,309 [574.5]
	Inner 25 mm [0.98 in] surface	> 4 mm [> 0.16 in]	0.0226 m ³ [0.8 ft ³]	9	398 [11.3]
	Outer 25 mm [0.98 in] surface	< 4 mm [< 0.16 in]	0.0241 m ³ [0.85 ft ³]	639	26,515 [750.8]
	Outer 25 mm [0.98 in] surface	> 4 mm [> 0.16 in]	0.0241 m ³ [0.85 ft ³]	19	788 [22.3]

*Doctor, S.R., G.J. Schuster, and F.A. Simonen. NUREG/CP-0166, Vol. 1, "Fabrication Flaws in Reactor Pressure Vessels." Proceedings of the Twenty-Sixth Water Reactor Safety Information Meeting, Bethesda, Maryland, October 26–28, 1998. Washington, DC: NRC. pp. 85–103. June 1999.

*Schuster, G.J., S.R. Doctor, S.L. Crawford, and A.F. Pardini. NUREG/CR-6471, Vol. 3, "Characterization of Flaws in U.S. Reactor Vessels—Density and Distribution of Flaw Indications in Shoreham Vessel." Washington, DC: NRC. November 1999.

Metal Fabrication Reliability (Jain et al., 2003)

Table 4-2. Causes of Fuel Failures in Pressurized Water Reactors*								
	Number of Assemblies							
Failure Cause	1989	1990	1991	1992	1993	1994	1995	1996 (Partial)
Handling Damage		6	2		-	1	1	1
Debris	146	11	67	20	13	6	10	1
Baffle jetting	_	-			-	_		_
Grid fretting	14	18	9	33	36	9	33	19
Primary hydriding		1	_	4	_		_	_
Crudding/corrosion	_	_	_	_		_	4	1
Cladding creep collapse	_			_	_	_	1	
Other fabrication	1	15	1	5	3	1	15	3
Other hydraulic	_	-	-	-	1	_		_
Inspected/unknown		_	_	_	36	36	13	2
Uninspected	43	58	35	61	14	3	12	1
Totals	204	109	114	123	103	56	89	27
Total discharged	2,196	3,461	2,937	3,302	3,612	2,636	3,666	
"Yang, R.L. "Meeting the Challenge of Managing Nuclear Fuel in a Competitive Environment." Proceedings of the 1997 International Topical Meeting on LWR Fuel Performance, Portland, Oregon, March 2–6, 1997. LaGrange Park, Illinois: American Nuclear Society. pp. 3–10. 1997.								