

United States Nuclear Regulatory Commission- - *n-A*

## ANALYSIS OF WASTE PACKAGE PERFORMANCE

Presented by: T. Ahn

(301) 415-5812, TMA@NRC.GOV U.S. Nuclear Regulatory Commission Engineering and Geoscience Branch Division of Waste Management Office of Nuclear Material Safety and Safeguards

For Presentation at Appendix 7 Meeting on Waste Package Materials February 11-12, 1998 Lawrence Livermore National Laboratory Livermore, CA

# Acknowledgments

 $\mathbf{G}^{\dagger}$ 

 $\ddot{\phantom{a}}$ 

 $\ddot{\phantom{a}}$ 

- K. Chang (NRC)
- R. Codell (NRC)
- G. Cragnolino (CNWRA)
- J. Davis (NRC)
- D. Dunn (CNWRA)
- J. Firth (NRC)
- K. Gruss (NRC)
- S. Mohanty (CNWRA)
- N. Sridhar (CNWRA)

#### OUTLINE OF PRESENTATION

Present Recent NRC/CNWRA Analyses of Waste Package Performance with Total System Performance Assessment Code (TPA) Version 3.1

- \* Inputs of Temperature, Relative Humidity, Saturation, pH, and Chloride Concentration
- Dry Air Oxidation of Outer Containers
- Corrosien:

I

- Humid Air Corrosion of Outer Container
- General Aqueous Corrosion
- Localized Aqueous Corrosion
- Galvanic Corrosion
- **Mechanical Failure of Outer Container**
- **Results of Analyses** 
	- Galvanic Protection
	- Chloride Concentration
	- Oxygen Partial Pressure
	- Backfill Emplacement
	- Other Sensitive Parameters
- \* Cladding Protection
- **Summary**

# FLOW CHART SHOWING VARIOUS COMPONENTS CONSIDERED IN WASTE PACKAGE PERFORMANCE

 $\epsilon_{\rm{th}}$ 

 $\sim$  $\sim$   $\sim$ 



 $\sim$ 



# TEMPERATURE AND RELATIVE HUMIDITY CALCULATIONS FOR VARIOUS VALUES OF THE AREAL MASS LOADING AND DIFFERENT VENTILATION AND BACKFILLING CONDITIONS (Manteufel, 1996)



# COMPUTED MULTIFLO VERSION 1.0 CALCULATIONS AT DIFFERENT TIMES ASSUMING NO INFILTRATION: (a) AND (b) SATURATION PROFILES, (c) pH PROFILE, AND (d) CHLORIDE CONCENTRATION PROFILE<br>(Lichtner, 1997)  $\sim \frac{3}{\beta}$



6

#### DRY AIR OXIDATION OF OUTER CONTAINER

*---*

Extrapolation of Laboratory Test Results: Negligible

2 um after 10,000 yrs, at 200 C (Stahl, 1993)

- 127  $\mu$ m after exposure to temperatures decreasing from 280 C to 210 C over a 5000 yr. period; with spalling of oxide, 350 to 1600  $\mu$ m over 5000 yrs. (Henshall, 1996)
- at most 100  $\mu$ m at 200 C for 10,000 yrs. by localized penetration (Ahn, 1996)
- $(3 4)$  µm for 1000 yrs. at 250 C (Larose and Rapp, 1996)

Areas of Uncertainty:

Intergranular Penetration of Oxygen, especially with Alloying Elements

Effects of Prior Dry Oxidation on Aqueous Corrosion

Calculations: Matrix and Grain Boundary Diffusion of Oxygen

#### **CORROSION**

*---*

 $\mathbf{L}$ 

Relative Humidity (RH) Determines Humid Air Corrosion and Aqueous Corrosion of Carbon Steel.

Critical RH Values: Capillary Effects and Chemistry Effects

humid air corrosion - *0.55* aqueous corrosion - 0.80

Humid Air Corrosion of Carbon Steel: Constant Rate

Aqueous Corrosion of Carbon Steel:

at  $pH > 8$ : - passive general corrosion - localized corrosion

Passive General Corrosion:

carbon steel: rate  $\tilde{ }$  12  $\mu$ m/yr alloy 825: rate  $\tilde{\phantom{a}}$  1.0  $\mu$ m/yr

 $1.1610^{3}$  m/y

Localized Corrosion for Outer and Inner Containers

initiation:

 $\epsilon$  .

- (1) corrosion potential greater than critical potential (i.e., repassivation potential)  $E_{\text{crit}} = E^{\circ}$  (T) + B(T) log [Cl<sup>-</sup>]
- (2) critical chloride concentration
- (3) no induction time

#### propagation:

- (1) outer container: a power law of time (Marsh and Taylor, 1988)  $P = A t<sup>n</sup>$
- (2) inner container: constant rate

Galvanic Corrosion

 $E^{\text{wp}} = (1 - \eta) E_{\text{corr}} + \eta E_{\text{conv}}$ 

- $\eta$  : efficiency of galvanic coupling  $0 < \eta < 1$
- **EwP:** corrosion potential of galvanic couple
- **Ecorr:** corrosion potential of inner layer
- **E**<sub>couple</sub>: experimentally measured values of potential of bimetallic couple assuming perfect galvanic coupling

#### MECHANICAL FAILURE OF OUTER CONTAINER

*---*

### Decrease Toughness by Thermal Embrittlement At Low Temperature for Long Times

Cause of Thermal Embrittlement: Sb, P, Sn and As Segregation along Prior Austenite Grain Boundaries (P is the Predominant Impurity in Commercial Steels.)

Source Stress: Residual Stress in Welds (Rockfalls Are Considered in Other Part of TPA Code.)

Thermal Embrittlement and Mechanical Failure May Not Occur in Carbon Steels Due to Prolonged Exposure to Temperatures above 250 C (Will Revisit Alloy Steel Susceptibility in Future Work)

## ANALYSES OF WASTE PACKAGE LIFETIME

(Preliminary Results)

Galvanic Protection

# Chloride Multiplication Factor: Concentration Factor from Chloride Concentration as a Function of Time (Calculated by MULTIFLO Code)

Oxygen Partial Pressure (No Time Dependence)

Backfill Emplacement

Other Sensitive Parameters:

- critical RH
- thickness of water film
- passive current density for corrosion of inner container
- coefficients for localized corrosion rate of outer container

**Effects of Galvanic Protection on Fraction of Failed Waste Packages** 



(The throwing power is on the order of the waste package dimensions because the resolution scale of modeling is the waste package.)



Scatter plots for total EBS release as a function of chloride multiplication factor (uniform; 1, 30) for 20,000 yr and 100 vectors. Case 20-oxygen partial pressure (triangular; 2.1e-5, 2.1e-1, 2.1e-1), Fow (constant; 1.0), FMult (constant; 0.05) and SAWetFr (constant; 0.5). All other parameters were constant at mean values, except UZFLOW flow parameters that were sampled.



**Chloride Multiplication Factor** Scatt :r plots for total EBS release as a function of chloride multiplication factor (uniform; 1, 30) for 20,000 yr and 100 vectors. Case 14--oxygen partial pressure (triangular; 2.1e-5, 2.1e-1, 2.1e-1). All other parameters were constant at mean values, except UZFLOW and EBSREL flow parameters that were sampled.

Edwar Plan paramile



U low the

Scatter plots for total EBS release as a function of oxygen partial pressure (triangular; 2.1e-5, 2.1e-1, 2.le-1) for 20,000 yr and 100 vectors. Case 20-chloride multiplication factor (uniform; 1, 30), Fow (constant; 1.0), FMult (constant; 0.05) and SAWetFr (constant; 0.5). All other parameters were constant at mean values, except UZFLOW flow parameters that were sampled.



Scatter plots for total EBS release as a function of oxygen partial Pressure (triangular; 2.1e-5, 2.1e-1,  $\angle$ .1e-1) for 20,000 yr and 100 vect rs. Case 14-chloride multiplication factor (uniform; 1, 30). All other parameters were constant at mean values, except UZFLOW and EBSREL flow parameters that were sampled.

# **Effects of Backfill Emplacement on Fraction of Failed Waste Packages**



15

## CLADDING PROTECTION

*-- -- --- - ---- - ----- -- - -- - -- - - -- - -- - -- - -- - -- - - \_- - - - -\_ - \_- - -*

Failure Modes:

 $\sim$   $\sim$ 



- creep

- mechanical failure

Protection:

- intact

- partially failed

Detailed Failure Models were not in the Code. Representative Parameters were Used in Calculations.

#### **SUMMARY**

Current NRC model abstraction and analyses of waste packages were presented. Thermal and environmental conditions were given as inputs in the analysis.

*---*

- Dry air oxidation was determined to be insignificant based on the extrapolation of laboratory test data.
- In corrosion, humid air corrosion and aqueous corrosion were considered, depending on RH. Corrosion was contrealed electrochemically. General passive corrosion for pH greater than 8.0 were considered for carbon steel.
- In localized corrosion, critical potential (i.e., repassivation potential) determined the initiation influenced by temperature and chloride concentration. Corrosion potential above critical potential and minimum chloride con
- Galvanic protection was considered using galvanic efficiency coefficient.
- The most sensitive parameters in corrosion of waste package materials include: galvanic protection, chloride multiplication factor, oxygen partial pressure, and backfill emplacement. Other sensitive parameters include critical RH, thickness of water film, passive current density for inner layer, and coefficient for localized corrosion rate of outer layer.
- Cladding appears to be an important barrier.