The Proposed Yucca Mountain Repository from a Corrosion Perspective

Joe H. Payer[1], Uziel Landau[2], C.C. Liu[2], Xi Shan[1], Meijun Shao[2], Pallavi Pharkya[1], Arun S. Agarwal[2], Hung Ha[1] and Hongyi Yuan[1]

CASE WESTERN RESERVE UNIVERSITY

[1]Department of Materials Science and Engineering, [2]Department of Chemical Engineering
Case Western Reserve University

RESEARCH SHOWCASE
CASE WESTERN RESERVE UNIVERSITY

ABSTRACT

The proposed Repository presents a familiar materials performance application that is regularly encountered in energy, transportation and other industries. The widely accepted approach to dealing with materials performance is to identify the performance requirements, to determine the operating conditions to which materials will be exposed and to select materials of construction that perform well in those conditions. A special feature of the proposed Yucca Mountain Repository is the extremely long time frame of interest, i.e. 10,000's of years and longer. Thus, the time evolution of the environment in contact with waste package surfaces and the time evolution of corrosion damage that may result are of primary interest in the determination of expected performance.

Researchers at Case are part of a Department of Energy Corrosion and Materials Performance Cooperative. This team of leading scientists/engineers from major universities and national laboratories is working together to further enhance the understanding of the role of engineered barriers in waste isolation. The team is organized to address important topics:

- Long-term behavior of protective, passive films
- Composition and properties of moisture in contact with metal surfaces
- Rate of penetration and extent of corrosion damage over extremely long times.

The work will also explore technical enhancements and seek to offer improvements in materials costs and reliability.

INTRODUCTION

- Corrosion is a primary determinant of waste package performance at the proposed Yucca Mountain Repository
  - The most likely degradation process
  - Controls the delay time for radionuclide transport from the waste package
  - Determines when packages will be penetrated and the shape size and distribution of those penetrations
- In this presentation a framework for the analysis of localized corrosion is presented and demonstrated for a scenario
  - Water chemistry of mixed salt solutions (sodium chloride-potassium nitrate)
  - Time-temperature-relative humidity profiles for a hot, mid and cool temperature waste package

Methodology For Determination Of Materials Performance

- Materials performance at the proposed Yucca Mountain Repository is amenable to a familiar and effective analytical methodology
  - Widely accepted in the energy, transportation and other industries
- Three components comprise the analysis
  - Definition of the performance requirements
  - Determination of the operating conditions to which materials will be exposed
  - Selection of materials of construction that perform well in those conditions
- A special feature of the proposed Repository is the extremely long time frame of interest, i.e. 10,000's of years and longer
  - Time evolution of the environment in contact with waste package surfaces
  - Time evolution of corrosion damage that may result

Locations of Spent Nuclear Fuel and High-Level Radioactive Waste

- Proposed Repository is about 300 m below the surface and 300 m above the water table
- Unsaturated zone, i.e. fractures and pores in rock are partially filled with water
- Desert area with about 18 cm of rain per year atmospheric pressure
- Ambient waters are dilute and near neutral pH
- Concentrated waters can form by condensation, deliquescence and evaporation

Design of The Proposed Yucca Mountain Repository, Emplacement Drift & Waste Canisters

BACKGROUND

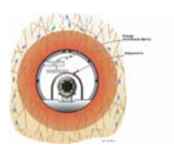
- Alloy 22 belongs to a family of Ni-Cr-Mo alloys
- Earlier alloys include C-276 and C-4 and later alloys include Inconel 686, Alloy 59, Hastelloy C-2000 and MAT-21
  - Alloy 22 (N06022) is a solid solution of Ni, Cr, Mo and W as the main alloying elements
  - Cr-Mo-W in Alloy 22 act synergistically to provide resistance to localized corrosion such as crevice corrosion
- Large industrial equipment in service for many years in harsh environments without corrosion
  - Alloy 22 has great toughness and over 50% elongation before failure
  - Can be hot or cold formed and is weldable by many methods
  - Can be fabricated into large structures and components

Corrosion Resistance is Crucial to Waste Package Performance

- Radionuclides are fully isolated if there are no penetrations
  - Even penetrated package can limit radionuclide movement
- Corrosion rates of passive metals are extremely low
  - Realistic rates are less than 1 µm/yr (a millionth of a meter per year) and much less
  - Alloy 22 layer is 2-cm thick (a stack of 12 U.S. quarters)
- Corrosion rates of approximately 0.01 µm/year are measured in exposures of over 5-years at the Long Term Test Facility at Lawrence Livermore National Laboratory

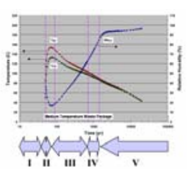
16,000 to 160,000 years to penetrate the thickness of one U.S. quarter for a corrosion rate of 0.1 to 0.01 µm/yr

Attributes of the Proposed Yucca Mountain Repository



- One long, slow heating/cooling cycle
- Packages cool to ambient over several thousands of years
- Waste packages on support pallets
- No immersion in waters
- No moving parts
- Low heat fluxes, slow heating and cooling, and modest thermal gradients
- Radiation effects at waste package surface negligible after a few hundred years
- Limited amount of water moving through the rock
- Limited salts and minerals carried into drifts by incoming water and dust

Relevant Time Periods Regards Corrosion



I - Emplacement of waste packages and preclosure
- Start to Year 50

II - Heat Up after closure
- Year 50 to ~65

III - Cool down/Thermal Barrier (drift wall above boiling temperature)
- Year ~65 to 750

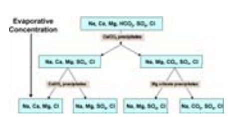
IV - Cool Down/Dripping and Seepage Possible
- Year 750 to 1375

V - Waste Packages below Critical Temp for Corrosion
- Year 1375 and beyond

This scenario is for Temp-RH shown above
- Waste package at 101°C when drift wall cooled to 96°C
- Critical corrosion temperature 90°C

Periods are determined by Temp-RH conditions
- Time when drift wall reaches 96°C
- Critical corrosion temperature for Alloy 22

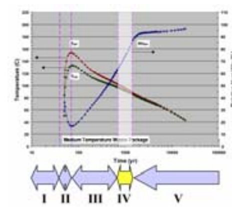
Chemical Divide Processes Determine the Categories of Waters

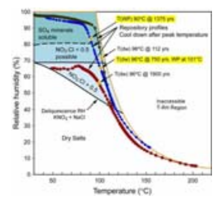


Ambient Waters
Dilute solutions
Na-Ca-Mg-HCO3-CO3-Cl-NO3-SO4
Near neutral pH

Waters can be concentrated
Modified during movement
Thermal-chemical processes
Modifications on waste package surface
Chemical and electrochemical processes

Period IV Analysis of T-RH-Solution Composition





Drift wall 96°C at 750 years;
Waste Package at 101°C;
Relative Humidity 65%

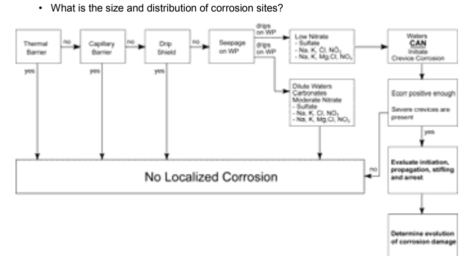
Critical Corrosion Temp 90°C
at year 1375; Relative Humidity 85%

The Temp-RH at any time fixes the possible waters. Can follow the trajectory with time

Number of non-corrosive solutions; Sodium chloride with low nitrate solutions can be corrosive

Decision-Tree Analysis

- ***A decision-tree for localized corrosion***
  - Are environments and crevices present to induce localized corrosion?
    - Consider conditions in moist layers of particulate and deposits
  - If localized corrosion initiates, will it persist?
    - Consider stifling and arrest processes as the corrosion proceeds
  - What amount of metal penetration occurs?
  - What is the size and distribution of corrosion sites?



Cyclic Polarization Provides a Widely Used Criteria for the Susceptibility to Localized Corrosion

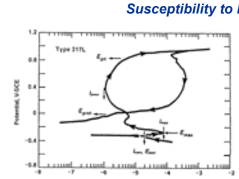


Figure 18. Anodic polarization behavior of type 317L stainless steel in an acid chloride solution.

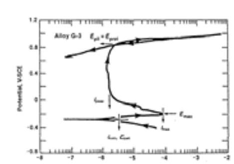
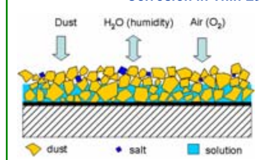
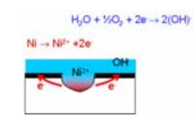


Figure 19. Anodic polarization behavior of alloy G-3 in an acid chloride solution.

Corrosion in Thin Layers of Particulate





- Dust deposited
- Soluble salts
- Particulate layer properties, such as conductivity, temperature, pH, degree of wetness etc.
- Localized environment on the surface
- Degree of wetness
- Gas composition and property, T, RH

Anode: Ni→Ni2 + 2e-   Cathode: H2O + ½O2 + 2e-→ 2(OH)-

Localized Corrosion can be Stifled by Limits on the Cathodic Processes

- In the proposed repository waste packages will never be fully immersed in solution
- Moisture and particulates may be present on surfaces
- Corrosion behavior in moist particulate can differ from full immersion
  - Limited size of corrosion site
  - Limited cathodic area to support localized corrosion
  - Limited cathodic kinetics could stifle corrosion

DOE/OST&I Multi-University Corrosion Cooperative (CorCoOp)

comprises of some 14 Principal Investigators and approximately 20 grad students and post docs

DOE CorCoOp is based at Case Western Reserve University

Collaborators

Arizona State University
Case Western Reserve University
The Ohio State University
Pennsylvania State University
University of California at Berkley
University of Minnesota
University of Toronto
University of Western Ontario
University of Virginia

SUMMARY

- Corrosion is a primary determinant of waste package performance
  - Controls the delay time for radionuclide transport from the waste package
- Two major aspects
  - Evolution of corrosion damage by localized corrosion
  - Durability of passive films
- Analysis of the potential for damage by corrosion is crucial and a major effort has been undertaken to enhance the technical basis for long-term behavior
  - Can corrosive environments form and persist?
  - Will localized corrosion start and persist?
  - What damage would result?

ACKNOWLEDGMENTS

The support of this work from the Office of Science and Technology and International (OST&I), Office of Civilian Radioactive Waste Management (OCRWM), U.S. Department of Energy (DOE) is gratefully acknowledged. The work is carried out as part of the DOE Multi-University Corrosion Cooperative (CorCoOp) under cooperative agreement DE-FC28-04RW12252. CorCoOp is directed by Professor Joe Payer at Case.