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Model for the Evolution of Thermohydrology and Its Effects on Chemistry and Corrosion of Engineered Barrier Materials

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ABSTRACT

The evolution of the environment within the emplacement drifts of a potential high-level nuclear waste repository at Yucca Mountain, Nevada, may be influenced by several factors, including quantity and chemistry of seepage water, temperature, and relative humidity. Understanding the evolution of these environmental conditions is important, as it determines the duration and initiation time of aggressive conditions that could cause early degradation of a waste package or drip shield. This task is challenging as a result of the heterogeneity of the natural system, interactions between the engineered and natural components, the unsaturated conditions at the repository horizon, and the potential for natural drift degradation in a thermally perturbed environment. This poster presents a phased study that involves experimental and numerical models to (a) simulate the in-drift conditions by controlling the heat load, infiltration flux and initial water compositions, and (b) measure the changes in temperature, relative humidity, and chemistry of the water contacting the surfaces of the engineered barrier materials. This study is a collaborative effort that integrates the unsaturated flow processes occurring during the thermal period with geochemical and corrosion processes. Relationships between evolution of the water chemistry as a function of temperature, relative humidity, infiltration flux, and corrosion of the waste package and drip shield materials are evaluated. Analytical and experimental studies to determine the composition of the salts forming due to evaporation from a wide range of initial pore water compositions were completed in the first phase. The second phase, now underway, considers the evolution of coupled thermohydrological-chemical conditions and its impact on corrosion processes using experimental and numerical models. Effects of drift degradation will be included in the third phase of this study.

INTRODUCTION

The evolution of environmental conditions within the emplacement drifts of a potential high-level nuclear waste repository at Yucca Mountain, Nevada, may be influenced by several factors, including the temperature and relative humidity within the emplacement drifts and the composition of seepage water. The performance of the waste package and the drip shield may be affected by the evolution of the environmental conditions within the emplacement drifts and in the surrounding host rock. The complex relationships among thermal, hydrological, and chemical processes lead to difficulty in estimating the in-drift environmental conditions that influence the quantity and chemistry of water potentially contacting the engineered barrier system (Figures 1 and 2). In-drift thermohydrological conditions significantly affect temperature and relative humidity gradients and lead to moisture redistribution (Fedors et al., 2004). These conditions, in turn, affect the chemistry of water contacting the drip shield and waste package, corrosion of the engineered barrier system, and transport of radionuclides through the invert to the unsaturated zone below the drifts. In addition, temperature estimates can be significantly modified when (i) drift degradation leading to formation of a rubble pile, (ii) natural convection, and (iii) the cold-trap process are considered.

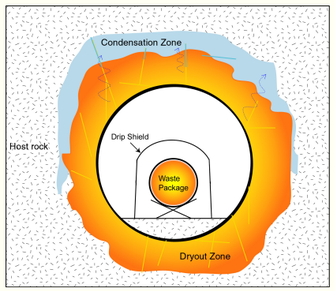

Figure 1- Schematic of the Thermohydrological Processes in the Near-field Environment

Waste packages for the potential repository will consist of an Alloy 22 (56Ni-22Cr-13.5Mo-3W-4Fe) outer container surrounding a Type 316 nuclear grade (NG) stainless steel (SS) inner container. Degradation of the Alloy 22 waste package outer containers may occur by uniform corrosion, pitting or crevice corrosion, and stress corrosion cracking. Work has been conducted by DOE and by NRC and the Center for Nuclear Waste Regulatory Analyses (CNWRA) to characterize the conditions under which the various degradation mechanisms may occur (Dunn et al., 2006).

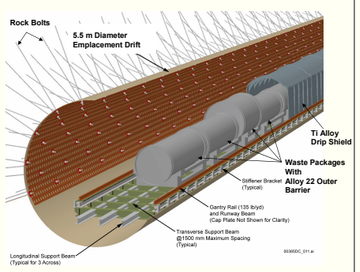

Figure 2. Schematic of the In-Drift Components at Yucca Mountain (Dunn et al., 2006) [1 m = 3.28 ft]

The study presented in this poster involves experimental and numerical models designed to (a) simulate in-drift conditions by controlling the heat load, infiltration flux and initial water compositions, and (b) measure the changes in temperature, relative humidity, and chemistry of the water contacting the surfaces of potential engineered barrier materials. The experimental and numerical models are being developed in a phased manner. The first phase, essentially complete, involved analytical and experimental studies to determine the composition of the salts forming due to evaporation from a wide range of initial pore water compositions. The second phase, now underway, considers the evolution of coupled thermohydrological-chemical conditions due to heat generated in an intact drift (i.e., no degradation) using experimental and numerical models. The third phase will consider the evolution of thermohydrological-chemical conditions in a degraded drift.

Phase 1 – Determination of the salt composition formed due to evaporation and its dependence on the starting water chemistry (Dunn et al., 2006)

Modeling Approach

- Initial water compositions represented Yucca Mountain unsaturated zone pore waters from three water types (calcium-chloride, neutral, and alkaline)
- Water chemistry evolution was assessed using thermodynamic simulations of evaporation, supplemented by chemical divide analysis

Testing Approach

- CNWRA conducted heater tests (Figure 3) using initially dilute calcium-chloride, neutral, and alkaline waters synthesized from reagent-grade chemicals at 110° C [[230 °F]
- The deposits were analyzed to determine final salt composition and evaluate the effects of corrosion on materials that are being considered for waste packages

Conclusions

- The testing program assessed a wide range of solution compositions typical of unsaturated zone pore waters from Yucca Mountain
- Final salt deposit composition depended on the initial composition
- The tests suggest that deliquescent salt deposits or brines are not likely to form for the given test conditions.

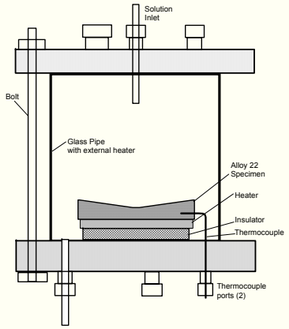

Figure 3. Schematic of Heated Specimen Test Cell

Phase 2 – Evolution of Thermohydrological and Chemical Environment and its Effect on Corrosion

Objectives

- Simulate the waste package and drip shield surfaces using a test system under thermohydrological conditions expected within the emplacement drifts in the potential repository at Yucca Mountain.
- Measure the evolution of near field chemistry in the host rock and on the surfaces of the engineered barrier materials.
- Determine the influence of temperature, infiltration, and chemistry variations on the corrosion of the waste package and drip shield materials.

Experimental Model

A schematic illustration and a photograph of the experimental setup under construction are shown in Figures 4 and 5 respectively. Quartz sand is used for the surrounding porous medium (i.e., host rock) as it is inert and will not modify the water chemistry by rock-water interactions. The effect of rock-water interactions on chemistry will be examined in the next phase, where rubble will replace quartz sand to simulate a degraded drift. The porous media is restrained using stainless steel mesh to form a simulated drift.

Multiple tests will be conducted with known starting water compositions and flux rates. Because the range of possible water compositions that may contact a waste package and drip shield is of interest, the starting water composition will be varied to include the most benign (low chloride and high nitrate), most aggressive (low nitrate and high chloride), and the highest boiling point (high chloride and high nitrate) combinations for the alkaline, neutral, and Ca-Cl brines.

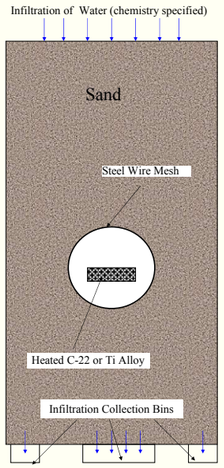

Figure 4 - Schematic of the Experimental Setup

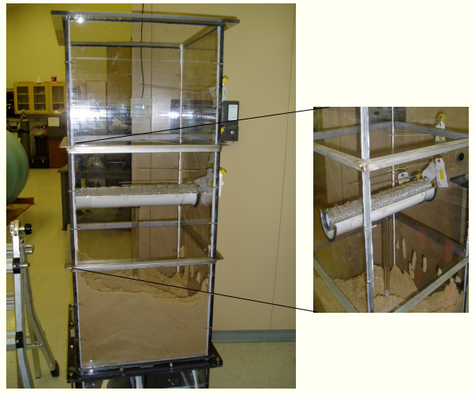

Figure 5 –Experimental Setup under Construction

Monitoring Quantity and Chemistry of Seepage: Collection and analyses of water at the bottom of the setup can be performed for each test case (Figure 4). Collection zones inside the drift and at the bottom of the apparatus are used to assess spatial variation of the flow paths. The relative amount of water that penetrates the drift will be determined as a function of flow rate and temperature. Sensors will be installed within the emplacement drifts and on the outside of the drifts to measure the presence of aqueous phase and the characteristics of the water such as pH, chloride concentration, temperature, and redox potential.

Monitoring Corrosion: Sensors will be used to determine the presence of moisture and measure corrosion rates. The experimental setup is designed to allow periodic removal and replacement of the test coupons to allow independent assessment of the corrosion of the waste package and drip shield materials and a comparison with the sensor measurements. Sensor locations will be adjusted based on the results of the water chemistry tests for each case.

Phase 2 – Evolution of Thermohydrological and Chemical Environment and its Effect on Corrosion (Contd.)

Numerical Model

A two-dimensional porous media model, using MULTIFLO 2.0.1 (Lichtner and Seth, 1996; Painter and Seth, 2003), was developed to represent thermohydrological processes in the experimental setup. MULTIFLO is a general code for simulating two-phase, two-component, non-isothermal flow and multicomponent reactive transport processes in porous/fractured systems. The thermal and hydrological properties of the porous medium used to design the experimental setup were based on typical values for sand (single continuum). Parameters will be calibrated as the experimental results become available.

Preliminary simulations assuming quasi-steady state conditions were used to guide the details of the experimental setup protocols, including (i) the height of the porous medium necessary to allow water flow to be influenced by the heat source within the drift; (ii) the lateral dimension of the experimental setup sufficient to accommodate the dryout zone; (iii) the heat load required to generate specified test temperature; and (iv) the location of wing heaters to maintain specified temperature gradients at the boundary.

Future simulations will be used to evaluate the (i) validity of assumptions related to unsaturated flow processes (e.g., flow focusing) in porous media models; (ii) use of equivalent thermal properties to represent heat transfer near the waste package; and (iii) ability of reactive transport models to represent coupled thermohydrological-chemical processes.

SUMMARY

The evolution of the environment within the emplacement drifts of a potential high-level waste repository at Yucca Mountain, Nevada, may be influenced by several factors, including quantity and chemistry of seepage water, temperature, and relative humidity. The complex relationships between thermal, hydrological, and chemical processes lead to difficulty in estimating the in-drift environmental conditions that influence the quantity and chemistry of water potentially contacting the engineered barrier system. A phased program of experimental and numerical studies are underway to (i) simulate the in-drift conditions by controlling the heat load, infiltration flux and initial water compositions, and (ii) measure the changes in temperature, relative humidity, and chemistry of the water contacting the surfaces of the engineered barrier materials.

The first phase involved analytical and experimental studies to determine the composition of the salts that form due to evaporation. Tests conducted in this phase used a wide range of solution compositions representative of calcium-chloride, neutral, and alkaline brines that may be produced through the evaporation and concentration of initially dilute seepage water at the potential repository at Yucca Mountain, Nevada. The composition of the salts that formed was dependent on the composition of the initially dilute water chemistry. The second phase will consider the evolution of thermohydrological conditions in the surrounding porous medium (i.e., host rock) using experimental and numerical models. The surrounding porous medium is represented using quartz sand to avoid modifying water chemistry by rock-water interactions. In the third phase, rubble (degraded drift scenario) will replace sand to represent the surrounding porous medium.

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