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WATER AVAILABILITY INSIDE PROPOSED YUCCA MOUNTAIN REPOSITORY BREACHED WASTE PACKAGES

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We present a model to evaluate the water mass balance inside a breached waste package in Yucca Mountain (YM) repository environments. The amount of water as liquid or vapor that can accumulate inside or percolate through the package in the emplacement drift is modeled as a function of the temperature and relative humidity (RH) near the waste package, the dripping rate of water from seepage, the area of failure patches on the waste package, and the extent of waste degradation. The water activity inside the waste package is assumed to be determined by both matric and osmotic potentials in the porous waste degradation products that also includes hygroscopic salts. We implemented the model and conducted a set of Monte Carlo simulations to gain insight into the variability and uncertainty associated with model predictions. The model shows that water vapor diffusion can be as important as the advective seepage flow. In addition, chemical reactions during waste degradation can consume a significant fraction of water accumulated in the waste package.

I. INTRODUCTION

Waste packages are one of the key engineered barriers designed to reduce long-term radionuclide release from the proposed Yucca Mountain repository. It is anticipated that some fraction of waste packages will be breached as a result of degradation processes such as stress cracking and/or general corrosion. Water may enter a breached package through failure openings by either vapor diffusion or liquid advection. The amount of water available inside a waste package will ultimately determine the extent of waste degradation and the subsequent release and transport of radionuclides from the waste package.

In this paper, we present an in-package water balance model, in which the amount of water accumulating inside or percolating through a breached waste package is calculated as a function of the relative humidity and temperature in the surrounding drift environment, the rate of water dripping onto the waste package as seepage, the area of breaches on the waste package, and the extent of waste degradation. Our model shows that the combination of the unique Yucca Mountain in-drift environment and the physical configuration of the waste packages can effectively prevent large quantities of water from accumulating inside a waste package and therefore the resulting conditions are not favorable for radionuclide transport. It should be noted that this model has not been used or implemented to directly support any work related to the Department of Energy (DOE) license application (LA) to the Nuclear Regulatory Commission (NRC) for the YM repository. Instead, the results of this model provide additional confirmation that sub-models used in support of the Total System Performance Assessment (TSPA) model for the license application are adequate. This modeling effort is part of the Performance Margin Analysis (PMA) conducted as part of the evaluation of conservatisms of the TSPA model. The PMA is documented in Appendix C of the TSPA-LA report model report MDL-WIS-PA-000005 REV00 (2007)[1].

II. MATHEMATICAL MODEL

II.A. Relevant processes

The quantity of water that can accumulate inside or percolate through a waste package is controlled by complex hydrologic, thermal, and chemical processes. At early stages after repository closure, heat generation from radioactive decay is high, and a steep temperature gradient will be maintained between the inside and the outside of a waste package, which, in the event of waste package breach, will tend to prevent water vapor condensation onto waste components inside the package. With time, temperatures decrease as heat generation by radioactive decay decreases, allowing for an increase in moisture in the package. In addition, as wastes degrade, certain hydroscopic salts (e.g., CsI, NaCl) are released into water occupying the pore space of the degradation products[2], creating a lower water activity inside the waste package due to an osmotic effect. This osmotic potential, together with the matric potential of the porous degradation products, will cause water vapor to condense inside the waste package. However, chemical reactions during waste degradation may consume a significant amount of the water entering the waste package.

For simplification, our model treats the inside of a waste package as a uniformly mixed compartment in terms of chemical composition and water distribution (Figure 1). The model assumes that all seepage water entering the drift will fall on the upper surfaces of waste packages forming a liquid water sheet or rivulets. A fraction of this water will percolate through breaches on waste package surface, and the rest will run off the package surface. Water will enter the waste package by vapor diffusion or liquid advection through the breaches. Water vapor diffusion depends on the gradient in vapor density between the inside and the outside of the waste package. The amount of seepage water that can enter the package by advective flow is determined by how long the water droplets will stay on the package surface and how fast the liquid water can percolate through the breaches. Part of water accumulated inside the waste package will be consumed by degradation reaction of internal materials. The model also assumes that the degradation products will form a porous medium, that will tend to hold liquid water in pores through a capillarity effect, and salts released from degradation reaction will create an osmotic potential, which will enhance water vapor condensation.

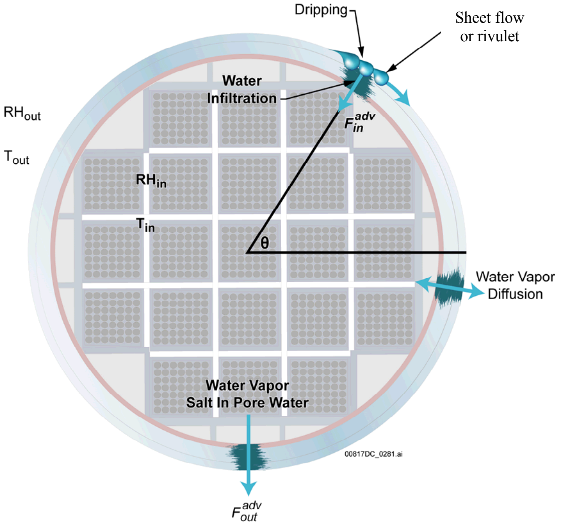

Figure 1. Schematic diagram of water balance inside a waste package.

II.B. Water Potential

We assume that the total water potential (ψ, bars) inside the pores of waste degradation or metal corrosion products is determined by both the matric potential (ψ_m, bars) and the osmotic potential (ψ_o, bars)[3]:

$$\psi = \psi_m + \psi_o \tag{1}$$

The matric potential, as a function of water saturation (s_w), can be described by a modified Campbell's function[4]:

$$\psi_m = \psi_e (s_w)^{-b} [1 - \alpha (T_{in} - T_0)](1 + \beta m) \tag{2}$$

where ψ_e (referred as an air entry potential), b, α and β are constants; T_{in} is the temperature (K) inside the waste package; and T_0 represents the reference temperature (298.15 K). The modified Campbell's equation accounts for the effects of the effects of elevated temperature and dissolved salt on water-air surface tension, based on the observations that the surface tension of water linearly decreases with the temperature and increases with dissolved salt concentration (m, molality) over the parameter ranges of interest[5,6].

The osmotic potential is created by salt dissolution in pore water. For a simple symmetrical electrolyte solution (e.g., NaCl), the osmotic potential of water can be calculated by using the van't Hoff relation[7]:

$$\psi_o = \omega \eta m \rho_{water} R T_{in} \tag{4}$$

where ω is the osmotic coefficient; η is the number of osmotically active species per molecule of solute (e.g., value of 2 for NaCl); ρ_{water} is the density of liquid water (kg/m^3); and R designates the gas constant (82.06 cm^3 atm/mole/K).

The total water potential in Equation (1) is related to the relative humidity inside a waste package (RH_{in}) by[8]:

$$RH_{in} = \frac{\rho_{in}(T_{in})}{\rho_s(T_{in})} = e^{-\frac{\psi v_w^0}{R T_{in}}} \tag{5}$$

where $\rho_{in}(T_{in})$ is the water vapor density inside the waste package (kg/m^3) at T_{in}; $\rho_s(T_{in})$ is the saturated water vapor density of pure water (kg/m^3) at T_{in}; and v_w^0 is the molar volume of liquid water (m^3/mole). It is assumed that v_w^0 doesn't change significantly within the temperature range of interest (i.e., T_{in} <100°C) and is thus treated as a constant.

II.C. Mass Balance

The water balance inside a breached waste package can be described by the following mass conservation equation:

$$\frac{dQ}{dt} = \rho_{water}\left(F_{in}^{adv} - F_{out}^{adv}\right) + \frac{D\phi^{5/3}A}{L}(\rho_{out} - \rho_{in}) - M_w \sum_i n_i^w S_i s_w R_i \tag{6}$$

with

$$Q = \frac{V_s \phi}{1-\phi}\left[s_w \rho_{water} + (1-s_w)\rho_{in}\right] + V_{void}\rho_{in} \tag{7}$$

where Q is the mass of water in kg as either liquid or vapor inside the waste package; t designates the time (year); V_s stands for the total volume of degradation products (m^3) inside the waste package; ϕ refers to the porosity of degradation products; ρ_{water} delineates the density of liquid water (kg/m^3); V_{void} symbolizes the volume of void space outside the fuel rods and corrosion rinds (m^3); F_{in}^{adv} and F_{out}^{adv} are the advective fluxes of liquid water into and out of the waste package respectively (m^3/year); ρ_{out} signifies the water vapor density outside the waste package (kg/m^3); D corresponds to the molecular diffusivity of water vapor in air (m^2/year); A represents the cross-sectional area of breaches on the outer layer of the package for water vapor transport (m^2); L denotes the thickness of the outer layer of the waste package (m); M_w specifies the molecular weight of water (kg/mole); n_i^w equals the number of moles of water consumed per kilograms of material degraded; S_i defines the surface area of material component i (m^2); and R_i assigns the degradation rate of component i (kg/m^2/year). It is assumed that in an unsaturated porous medium, reactive surface areas are proportional to water saturation [9,10]. The left-side term of Equation (6) represents the accumulation rate of water mass inside a waste package. The three terms on the right side of the equation represent the water flux by advective flows, the water flux by vapor diffusion, and the water consumption rate, respectively. The model also treats the corrosion product filling in the failure openings as a porous medium for water transport. The factor $\phi^{5/3}$ accounts for the effect of tortuosity in the porous medium[3].

Similarly, the mass balance for dissolved salt can be formulated as follows:

$$\frac{dM_{salt}}{dt} = 1000(F_{in}^{adv} m_{seep} - F_{out}^{adv} m) + \sum_i n_i^{salt} S_i s_w R_i$$

where M_{salt} is the total salt dissolved in the pore water (moles); n_i^{salt} specifies the moles of salt released per kilograms of material degraded; and m_{seep}(6)defines the concentration of dissolved salt in the seepage water.

(3)

II.D. Advective Water Fluxes

The advective water influx term (F_{in}^{adv}) in Equation (9) is evaluated based on the assumption that a water drip falling onto the waste package surface will form a liquid sheet or a rivulet with a nominal thickness of h (m) and a surface area of A_w (m^2), which will move down across the package surface. When the liquid sheet flows over the breaches of waste package, a certain fraction of water will be able to percolate into the waste package through the corrosion products in the breaches, with the remainder running off the surface.

The mean velocity for sheet flow ($\bar{u}_{sheet}$, m/s) on an inclined surface is described by [11,12]:

$$\bar{u}_{sheet} = \frac{\rho_{water} g h^2 \sin(\frac{\pi}{2} - \theta)}{3\mu} \tag{9}$$

where g is the gravity acceleration (9.81 m/s^2); θ denotes the angle as shown in Figure 1; μ refers to the viscosity of water (Pa s), and h delineates the liquid film thickness (m). The evolution of the water sheet can be described by:

$$\frac{d(A_w h)}{dt} = -\phi A_w f K_c^s \sin\theta \tag{10}$$

$$\frac{d\theta}{dt} = -\frac{\bar{u}_{sheet}}{2\pi R_{wp}} \tag{11}$$

where f is the fraction of breaches on the total waste package surface area; K_c^s is the hydraulic conductivity of the corrosion products filling the breaches under water saturated conditions (m/s); and R_{wp} is the radius of the waste package.

Assuming that the water advection is induced by the gravitational force only, the advective water flux out of the waste package (F_{out}^{adv}) in Equation (9) can be calculated by:

$$F_{out}^{adv} = A_{out} K_c(s_w) \qquad (8) \tag{12}$$

where A_{out} is the failure breach area on the bottom part of the waste package surface. The hydraulic conductivity K_c is a function of water saturation s_w [4]:

$$K_c = K_c^s (s_w)^{2b+3} \qquad (14)$$

where b is the Campbell constant in Equation (2).

III. MODEL SIMULATIONS

The mathematical model formulated above is implemented using a Monte-Carlo methodology. A total 30 model simulations have been performed, covering a time period of 10,000 years, for each of the two waste package types - the commercial spent nuclear fuel (CSNF) waste packages and the codisposal (CDSP) waste packages. In these simulations, waste package failure is assumed to occur 500 years after repository closure. The simulation results are presented in Figures 2 through 4.

The amounts of seepage water that can percolate through these breaches are shown in Figure 2. It can be seen that at maximum only a few liters of seepage water can enter a breached waste package per year over the first 10,000 years. This amount only accounts for less than one percent of total seepage flux, given seepage rates of 1 to 1,000 liters/package/year, and the rest of the water will run off the waste package surface. Thus, our modeling results indicate that a failed waste package can still maintain a large part of its barrier capability for reducing liquid water percolation through the package.

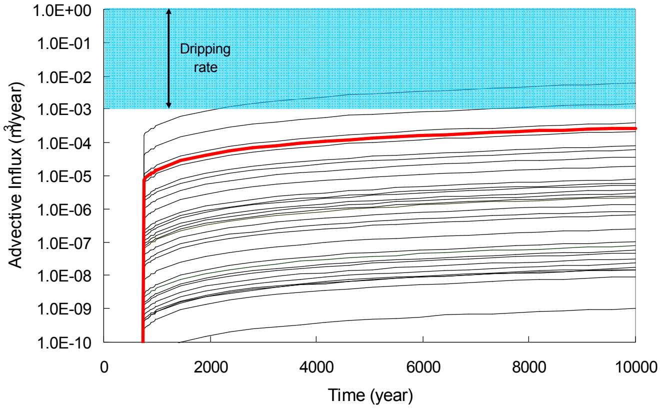

Figure 2. Rate of seepage water entering a breached waste package. Red curve represents mean values.

The evolution of water saturation inside a waste package is presented in Figure 9. Over the wide range of seepage rates used in the model simulations, the water saturation in both CSNF and CDSP waste packages is predicted to be relatively low. After the thermal event, the water saturation inside a CSNF waste package will range from 0.17 to 0.34 with the mean around 0.25. In contrast, the water saturation inside a CDSP waste package will generally be higher than that in the CSNF, ranging from 0.24 to 0.49 with the mean around 0.37. The higher water saturation in the CDSP package is partly attributed to the release of more salts from high-level waste glass.

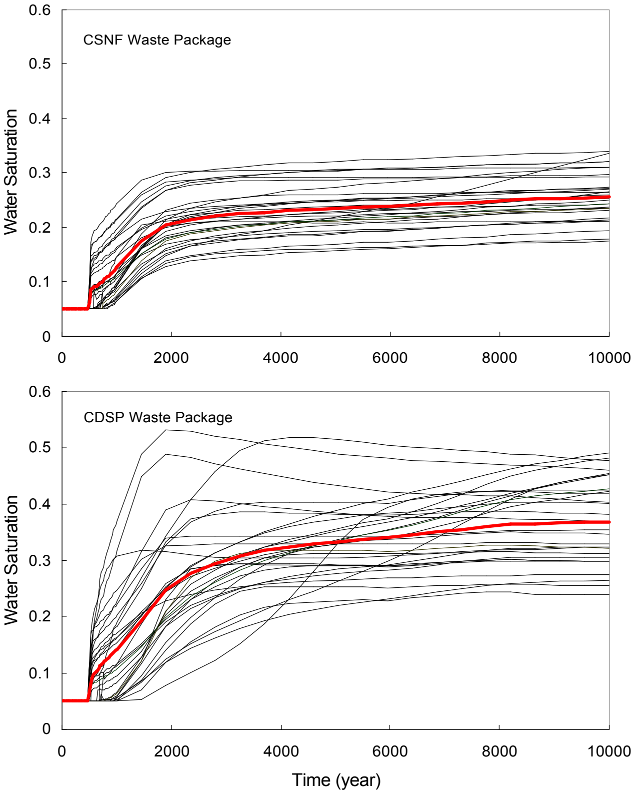

Figure 3. Water saturation in degradation products calculated as a function of time. The red curves represent mean values.

The model simulations show that a significant fraction of water that enters a waste package will be consumed by chemical reactions during material degradation. Because of water consumption and in-package water evaporation and also because of the remaining barrier capability of waste package, the advective water flow out of a waste package is predicted to be very small, as shown in Figure 4, in spite of a wide range of seepage rate (1 – 1,000 liter/year) used in the simulations. The flux for a CSNF waste package will be less than 1.0 liter/year, with the mean value of 0.05 liter/year, while the flux for the CDSP will be less than 5.3 liters/year with the mean value of 0.4 liter/year.

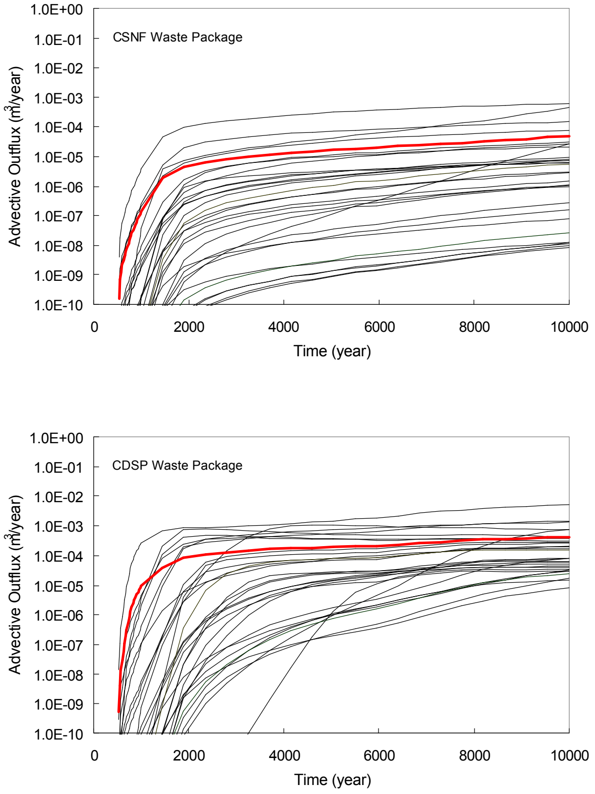

Figure 4. Advective water out-fluxes calculated as a function of time. The red curves represent mean values.

IV. CONCLUSIONS

The model simulations indicate that a breached waste package can maintain a large part of its barrier capability and only < 1% of the total seepage flux impinging on the waste package surface can enter the package. Vapor diffusion of water through the breaches can be as important as liquid water flow into the waste package. Waste degradation reactions can consume a significant fraction of water entering the waste package. Model simulations have demonstrated that the water saturation inside waste packages will be low (< 0.5) and the advective water flux out of a waste package will be small (with the mean value < 0.5 liter/package/year) over a wide range of seepage rates considered (1 – 1,000 liters/year). It is concluded that the combination of the unique Yucca Mountain in-drift environment and the physical configuration of the waste packages can effectively prevent large quantities of water from accumulating inside or percolating through a breached waste package and therefore the resulting conditions are not conducive of radionuclide transport.

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