

Effect of Physical Processes on Radionuclide Release from Partially Failed Containers-9260

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ABSTRACT

In the long run, nuclear waste packages at the Yucca Mountain repository are likely to evolve into a combination of corroded materials mixed with relicts of intact Alloy-22 and other waste package materials. Different rates of corrosion, due to physical and chemical disturbances in the environment of the repository, will lead to different times of penetration between waste packages and at different locations on the same waste package. Radionuclides are released from waste packages by dissolution and transport in water. In this paper, we shed some light on the effect of residual heat release, and other physical processes that take place in the waste package during penetration times, on radionuclide release. We develop a flow-through conceptual model for a probable serious failure in which multiple penetrations allow water to flow through a partially failed waste container. This model demonstrates that evaporation at hotter protected areas creates a capillary pressure gradient that causes water to flow with its dissolved and suspended contents toward these relict protected areas, effectively preventing radionuclide release. We derive a dimensionless group to estimate the minimum size of the covered areas required to sequester radionuclides and prevent release, and explore the implication of the flow-through model on the Yucca Mountain repository performance.

INTRODUCTION

The proposed repository at Yucca Mountain, NV is intended to store highly radioactive spent nuclear fuel, and high-level radioactive waste associated with weapons production (The total amount of Defense Waste will be very small relative to the amount of spent fuel). The layers of rock and dirt will shield the waste radiation, but the nuclear waste can be transported to the accessible environment by water. To protect people and the environment, the U.S. Department of Energy designed the Yucca Mountain repository that depends on multiple natural and engineered barriers, to isolate waste and keep it dry as long as possible. However, the waste packages will be penetrated eventually, and the water will have access to the nuclear waste, where its fate depends on the geometry and environment inside the failed waste container.

Geometry and Environment inside a Waste Package

Nuclear waste packages at Yucca Mountain repository are expected to fail gradually and at different rates, due to localized and general corrosions that are affected by physical and chemical disturbances in the environment of the repository. As the waste package fails, small cells of evaporation and condensation, inside the failed container, are projected to leave some portions of the waste package metals in more harsh corrosion environments than other parts. Over time, under these different rates of corrosion the waste package is likely to evolve to into a combination of corroded materials (i.e., porous media) mixed with relicts of intact Alloy-22 and other waste package materials. In this heterogeneous geometry, capillary breaks (i.e., discontinuous liquid transport pathways) may or may not exist.

Under these conditions, heat release from the nuclear waste will continue, in general, apart from the waste evolving material properties. Figure 1 shows the average rate of heat release per unit volume over time for two classes of radioactive waste packages at Yucca Mountain; the class of the highest release: pressurized water reactor [21PWR], and the class of the lowest release: defense glass logs [DHLW-L] [1]. Figure 1 indicates that the heat release from radioactive waste persists, although at very low rates, beyond one million years.

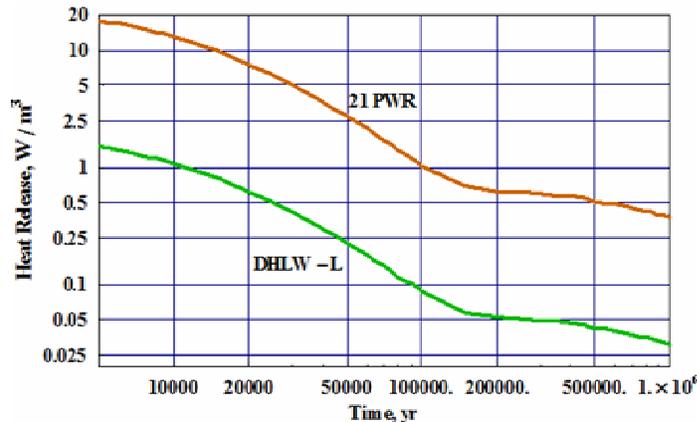


Fig. 1. The rate of heat release per volume from radioactive waste packages over time, plotted on a logarithmic scale. Small, but significant, heat release continues up to one million years.

Given all these conditions inside a partially failed waste container in the long run, what role can very little heat release play in radionuclide release process?

CONCEPTUAL MODEL

The conceptual model examines the effect of residual heat release, and related physical processes, on radionuclide release from partially failed waste containers. We introduce our model with an example from nature which resembles, to a plausible extent, the conditions and processes that take place inside a partially failed waste package. Figure 2 shows a picture was taken at the 1880's era silver mine on North Percha Creek, NM. The mouth of the mine is blocked with two berms of mine tailings, to slow the release of contaminated water from the mine tunnels. Despite the presence of a significant hydraulic head difference (~20 cm/100 cm) that drives water to flow through the berms, efflorescent crusts form on the top of the wet tailings (see Figure 2). The evaporation of water at the upper surfaces of the wet tailings, driven by the low humidity air near the drift entrance, creates a capillary pressure gradient that causes water to wick upward in the berms, that is, advective transport takes place for the dissolved minerals from the tailings forming the berms. As the water evaporation continues, the concentration of dissolved species increases near the tailings/air interface and minerals begin to precipitate, forming the efflorescent crusts. Interestingly, due to these processes, the minerals from tailings are sequestered on the top of the berms rather than being released into the water.

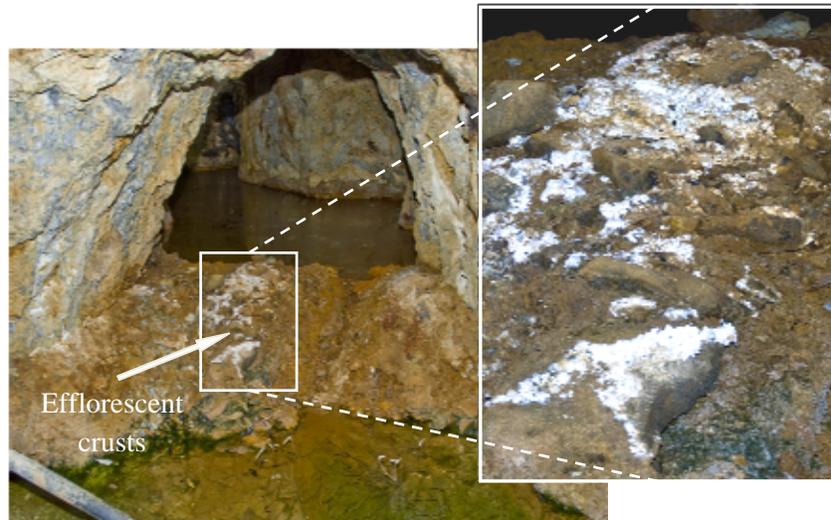


Fig. 2. North Percha Creek mine. Efflorescent crusts form on the top of the wet tailings, when water evaporates at the upper surfaces. Water evaporation leads to a gradient in capillary pressure that causes water to rise upward in the berms.

The environment in the tailings berms of North Percha Creek mine is a good analogy to the environment inside a partially failed waste package in the long run. The inside of a waste container will be a mixture of porous media and discrete objects; that is, it has physical properties similar to soil. The residual heat inside a waste package plays the same role as the low humidity air near the drift entrance, in driving the evaporation process.

The conceptual model addresses a serious failure of a waste package, where multiple penetrations allow water to flow through the waste package. Figure 3 shows a schematic drawing for this model. Residual heat is anticipated to set up flow systems in the areas protected from seepage. Evaporation at hotter areas, usually where the greatest concentration of heavy metals is present, creates a gradient in the capillary pressure that causes water (with its dissolved and suspended constituents), in the absence of direct seepage or condensate, to flow or wick toward the warmest region; that is, advective transport for radionuclides occurs toward the heat source in the protected areas. As water continues to evaporate, the concentration of radionuclides in these regions increases and radionuclides start to precipitate, in the same manner as the minerals in the tailings berms. On the other hand, the continuous evaporation of water creates gradient in radionuclide concentration, in these areas that drives diffusive transport for the radionuclides in the opposite direction (away from hotter regions; see Figure 3).

Since the advective transport of radionuclides is toward the sheltered hotter areas, radionuclide release from these areas can only occur by diffusive transport, when the rate of diffusive transport is greater than the rate of advective transport.

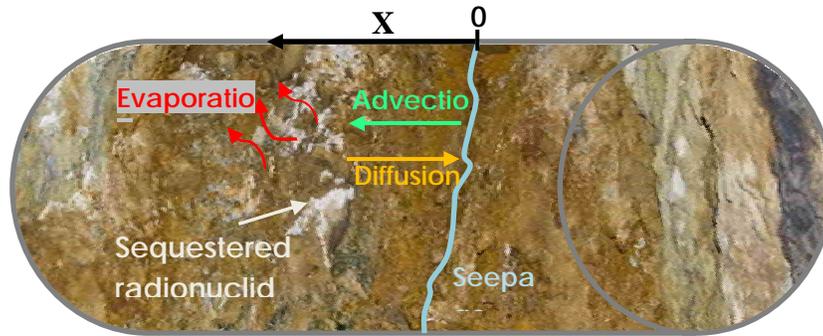


Fig. 3. A schematic drawing illustrates the physical processes in a flow-through system model. Evaporation in the hotter regions causes water to flow toward these regions (advective transport), while the increase in radionuclide concentration (due to evaporation) causes diffusive transport in the opposite direction.

The dimensionless Peclet number gives the ratio of advection to diffusion:

$$Pe = \frac{VL}{D} \quad (1)$$

Where V is inward advective velocity (m/s), L is length of non drip region (m), and D is the effective diffusion coefficient (m^2/s).

In the areas exposed to direct seepage or condensate, diffusion and advective transport are in the same direction; therefore $Pe > 0$ and there will be a rapid release for radionuclides from these areas. On the other hand, in the relict protected areas, where diffusive and advective transports are in opposite directions, if $0 > Pe > -1$, diffusive transport is greater than advective transport and a gradual release will occur, while if $Pe < -1$, the advective transport is greater and sequestration process for radionuclides will take place rather than release.

DISSCUSSION

According to the conceptual model, radionuclides will be sequestered in the relict sheltered (from direct seepage or condensate) areas, unless the diffusive transport is greater than advective transport. To estimate the minimum length of sheltered area (x , see Figure 3), required to sequester and prevent radionuclide release, over time, we used the dimensionless Peclet number with value equals (-1); that is, advection and diffusion are equal and in opposite directions. Hiraiwa and Kasubuchi estimated that the fraction of the measured thermal conductivity in moist soils with a water content of (0.2) that can be attributed to latent heat (vapor diffusion) is approximately equals (0.3), at temperature $30^{\circ}C$ [2]. Since at lower water content the contribution of latent heat declines, and we based our calculations on a water content of (0.1) inside the partially failed waste container, we assumed that (20%) of the heat generated by the

waste is available to evaporate water. Accordingly, the advective velocity is estimated from the heat generation of the waste (the calculations are made for a [21PWR] waste package), and the latent heat of evaporation of water. The effective diffusion coefficient is estimated using Archie's Law, which assumes that the porosity and tortuosity corrections are equal to volumetric water content squared [3]. The equations are solved for the minimum protected (no drip) areas required to prevent release over time; the results are shown in Figure 4.

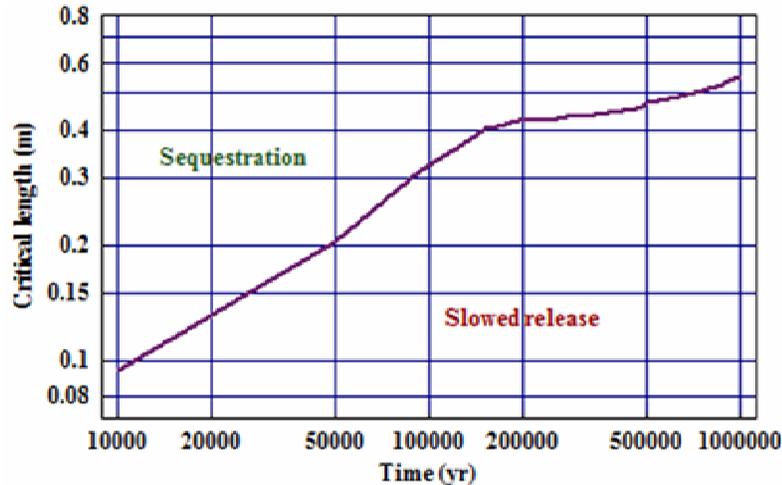


Fig. 4 Minimum size of sheltered (no drip) area required to protect the waste from leaching. Sheltered areas larger than this size do not support radionuclide release.

Figure 4 indicates that the minimum size required for radionuclide sequestration increases gradually over time, but with significant protection extending out to one million years. On the other hand the average size of sheltered areas decreases over time, due to general corrosion. Convolution of the two processes suggests that radionuclide release from the flow-through category of partially failed waste packages will be gradual and long delayed.

Radionuclide transport toward sheltered areas inside a failed waste package can be demonstrated by calculating the concentration profile of a radionuclide over distance. In the same manner, the calculations are based on the conceptual model. The first order continuity equation for advective and dispersive transport in groundwater, for the steady state, is used [4]. We assumed that the volumetric water content equals (0.2), and the fraction of released heat that goes to water evaporation is (30%) [2].

Figure 5 shows the concentration profile of the radionuclide over distance, in terms of the ratio of radionuclide concentration at any point (x; downstream), inside the waste container, to the initial point (x = 0; see Figure 3), where seepage occurs. The concentration profile is calculated for the time (25000) yr after closure.

Figure 5 indicates that the concentration of radionuclide increases as we move toward the heat source, in the sheltered areas of the waste package. The concentration ratio will not reach these

large values, shown in Figure 5, since any compound has limited solubility and start to precipitate after critical concentration. The order of precipitation for the radionuclides is illustrated in Figure 5. Normally the radionuclide with the lowest solubility will precipitate first. The light isotopes of the radionuclide are expected to precipitate closer to the initial point ($x=0$) since they have higher diffusion coefficients.

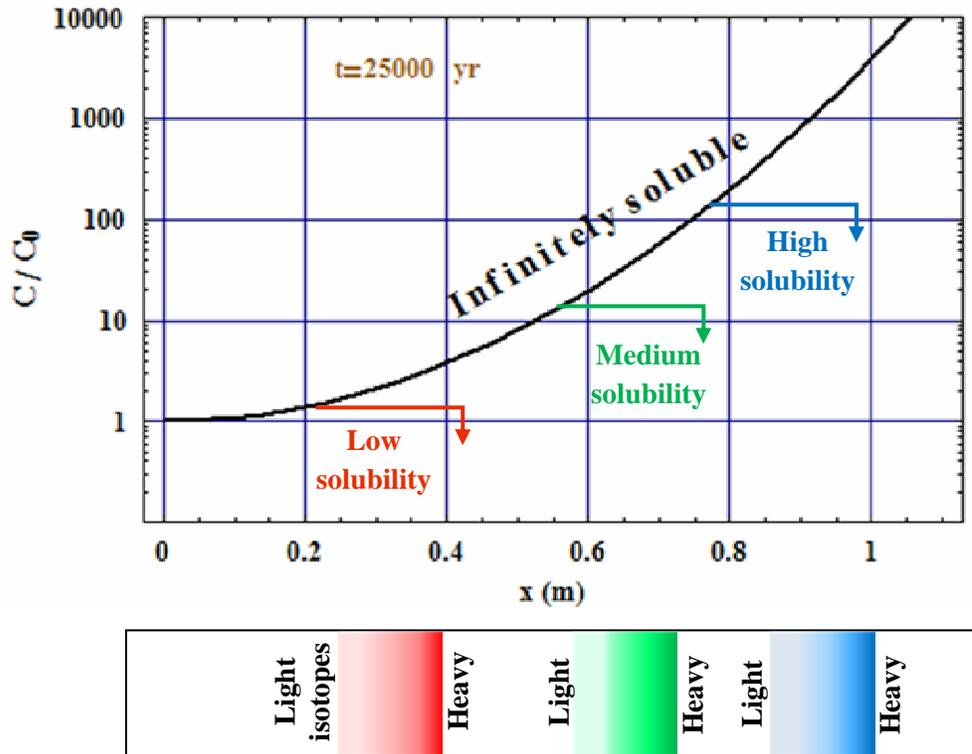


Fig. 5. The ratio of radionuclide concentration ($C(x) / C(x=0)$) vs. distance, calculated at time = 25000 yr after closure.

CONCLUSIONS

The conceptual model showed that the residual heat release from nuclear waste packages stored in a partially saturated environment, tends to accumulate radionuclides in the sheltered areas (where heat source exists) and prevents radionuclide release. Over time, the size of sheltered areas decreases by general corrosion and the size of sheltered areas required preventing release increases (as residual heat release decreases). As a result radionuclide release will be gradual and long delayed; that is heat releasing waste with an unsaturated environment make the Yucca Mountain repository inherently safe.

ACKNOWLEDGMENTS

WM2009 Conference, March 1-5, 2009, Phoenix, AZ

The Nye County Nuclear Waste Repository Project Office funded this work through funding provided by the U.S. Department of Energy Office of Civilian Radioactive Waste Management. We also thank the Center for Environmental Resource Management of The University of Texas at El Paso for their support.

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