

LIMITATIONS AND PATTERNS OF RADIONUCLIDE RELEASE FROM PARTIALLY FAILED CONTAINERS

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Over time, the waste package is likely to evolve into a combination of failure locations mixed with relicts of intact Alloy-22. In this paper we address the potentially serious failure where multiple penetrations allow water to flow through the waste package. Residual heat release in the waste is anticipated to set up flow systems in the relict protected areas, where liquid water flows into the protected area toward the warmest region and vapor flows outward, effectively preventing release and sometimes sequestering radionuclides in the relict sheltered areas. We derive a dimensionless group that specifies the condition where residual heat from relict waste package materials should set up this internal flow pattern. The dimensionless group is used to estimate the minimum size of the covered areas required to sequester radionuclides and prevent release. Over time, the minimum area required for protection slowly increases, while general corrosion decreases the average size of relict areas. 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.

I. INTRODUCTION

Understanding the processes actually controlling radionuclide release at Yucca Mountain (YM) is essential to ensure the future protection of the public's health. A key factor determining the performance of the proposed repository is the possibility of radionuclide release from the Engineered Barrier System (EBS) at YM. Once radionuclides are released into the host rock and given ample time, most radionuclides will eventually make it out into the broader environment. Source term – the transport mechanisms determining the amounts and types of radionuclides released– is an area with high conceptual uncertainty, and the models used to estimate release may be largely based upon processes that may not in general occur.

In this paper we develop a conceptual model of radionuclide release from the EBS and explore some of

the implications of the conceptual model. This model addresses the potentially serious failure of a waste package, where multiple penetrations allow water to flow through the system, entering at a higher elevation and exiting at a lower elevation in a partially failed waste package container.

II. GEOMETRY AND ENVIRONMENT FOR RADIONUCLIDE RELEASE

Release rate analysis is complicated by the fact that the geometry and properties of the inside of a failed waste container are unknown, and fundamentally unknowable. Corrosion predictions are based upon short term data in perhaps non representative environments since the environment is also unknown. Thus the corrosion rate of Alloy-22, stainless steel, and Zircaloy, may or may not proceed in an anticipated order. Small cells of evaporation and condensation are likely to leave some portions of the waste package metals in more benign and others in more harsh corrosion environments, making it difficult to predict exactly which pieces will last the longest. It is not guaranteed that the most corrosion resistant materials will last longest. Over time, as parts lose structural integrity and corrosion products fill voids, the waste package is likely to evolve into a mixture of porous media and discrete objects. This heterogeneity may or may not form capillary breaks (i.e., discontinuous liquid transport pathways) within the failed waste package, further complicating mass transport. All of these properties will change slowly over time.

If it is possible to further understand the processes controlling release rate, it seems likely that we can only be successful if the controlling processes are robust and not tightly related to many of the evolving material properties. One potentially controlling process is the release of thermal energy (heat) from the radioactive waste over time. Figure 1 shows the rate of heat release based upon the line load plotted on a logarithmic scale. The figure indicates that heat release occurs, albeit at a very low rate, up-to one million years.

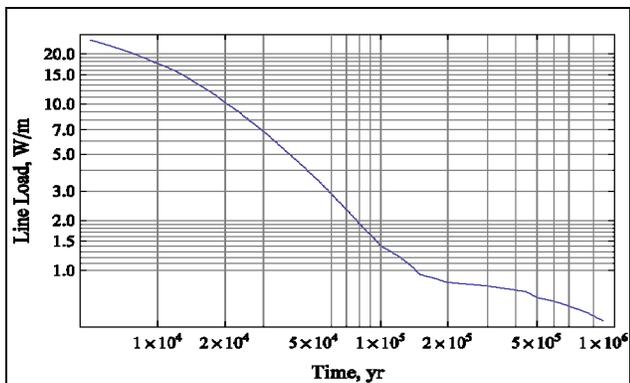


Figure 1. Line load on a logarithmic scale. Small, but significant, heat release occurs up-to one million years.

III. FLOW-THROUGH CONCEPTUAL MODEL

This model addresses the potentially serious failure of the waste package, where multiple penetrations allow water to flow through the system (see Figure 2). Diffusion-only release is included in this model.

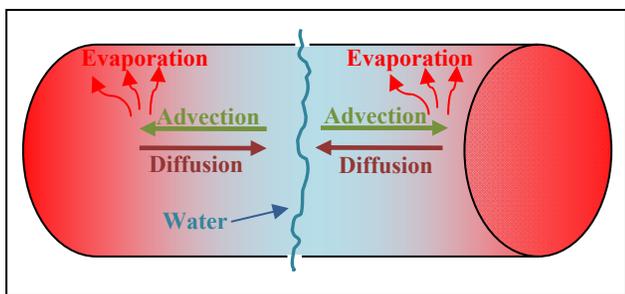


Figure 2. Schematic drawing showing physical processes in a flow through system.

Predicted temperature increase from the outside to the inside of a hypothetical failed waste container is shown in Figure 3, over a range of potential thermal conductivities characteristic of soils [1]. The calculations were performed using the equation for heat flow in a solid, infinite, circular cylinder with internal heat generation after [2]:

$$T = \frac{A_o (r_o^2 - r^2)}{4K} \quad (1)$$

where:

- T = temperature difference ($^{\circ}\text{C}$)
- A_o = rate of heat generation (W/m^3)
- r_o, r = radii of the cylinder surface and the inner point (m)
- K = thermal conductivity ($\text{W}/(\text{m}^{\circ}\text{C})$)

Since the precise thermal properties of the rubble and how it changes with time are unknown, Figure 3 is intended only to illustrate, in a semi-quantitative fashion, the amount of heat energy available.

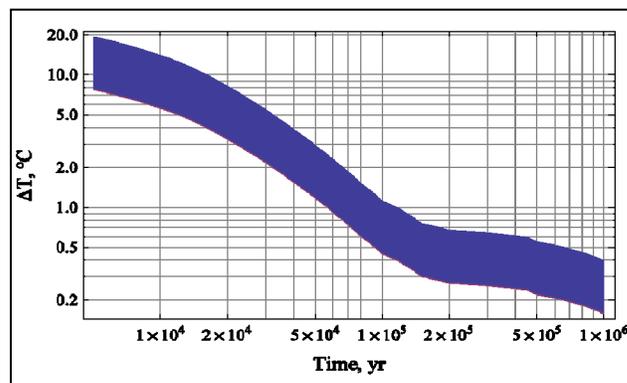


Figure 3. Plausible temperature rise from outside to inside of the rubble.

Residual heat release in the waste package is anticipated to set up flow systems in the relict protected areas, where liquid water flows into the center of the protected area, toward the warmest region, and vapor flows outward away from the warmest region. In the flow systems, created by residual heat release, water evaporates from the warmest regions, typically where the greatest concentration of heavy metal is present, leading to a capillary pressure gradient and subsequent wicking of water and dissolved or suspended constituents toward the heat source. Since the liquid flow is toward the heat source, liquid releases from these areas not only must be from diffusion, as a result of a gradient in radionuclide concentration, but the diffusion is against the advective transport direction: essentially the ions must “swim upstream” to escape (see Figure 2).

Given the flow system toward the majority of the radioactivity (heat source), radionuclides in these locations can only escape if the rate of diffusion is greater than the rate of advection. The ratio of advection to diffusion is given by the dimensionless Peclet number:

$$Pe = \frac{VL}{D} \quad (2)$$

where:

- V = inward advective velocity, m/s
- L = characteristic length (diameter of non drip region), m
- D = effective diffusion coefficient, m^2/s

Dimensionless numbers have the advantage of not needing exact geometries to make general conclusions. In this situation, the Peclet number is interpreted such that if $Pe < -1$, radionuclides entering the liquid water phase from the spent fuel will tend to accumulate at the hottest locations within the rubble and not be released into the crushed rock below. The Peclet number is negative because transport is in the opposite direction.

A simple experiment illustrating the flow system is shown in Figure 4. In this case an ordinary bathroom towel was immersed in water spiked with dye. Over time,

the dye travels up the towel (by capillary effect) and becomes concentrated at the point where evaporation becomes complete. The system is one of sequestration, not release.



Figure 4. Dye moves upward and concentrates in a towel dipped in water and dye. This is an example of the sequestration method presented, although in this case the evaporation is driven by the low-humidity room air rather than heat generation.

IV. NATURAL ANALOG

Another example illustrating the physical processes involved in this conceptual model comes from nature. Figure 5 shows a picture taken at the 1880's era silver mine on North Percha Creek, NM. At an unknown time, the mouth of mine was blocked with two bars or berms of mine tailings to slow the release of contaminated water from the mine tunnels. Despite the presence of water on both sides of the berms and a significant hydraulic head difference (~20 cm/100 cm) driving water flow through the berms, efflorescent crusts form on the top of the wet tailings. The crusts form when water at the upper surfaces evaporates, leading to a gradient in capillary pressure that causes water to rise upward in the berms.

As the water continues to evaporate, the concentration of dissolved species increases near the tailings/air interface and minerals begin to precipitate. The interesting observation is that, rather than being released into the water, the minerals from the tailings forming the berms accumulate at the top. This process is driven by evaporation of water into the lower humidity air near the drift entrance. This is similar to the situation in a failed waste package where liquid water will flow by capillary forces toward thermally hot waste where

evaporation occurs. A close-up picture to the crusts atop the berms is shown in Figure 6.



Figure 5. North Percha Creek mine berm with formation of efflorescent crusts.



Figure 6. Close-up picture of crusts. The evaporative concentration process appears to survive even in a wet system.

V. CALCULATIONS AND RESULTS

Using Peclet number values, the minimum size of the covered areas required to sequester radionuclides and prevent release is estimated. In this derivation, it is assumed that heat generated by the waste is available to evaporate water. Accordingly, the advective velocity in a moist system can be estimated from the heat generation of the waste and the latent heat of evaporation of water. Diffusion coefficients are estimated from the data of Conca and Wright [3]. The equations are solved for the minimum protected (no drip) region (L) required to prevent release, using two different values of volumetric water content. Figure 7 shows that the minimum size for sequestration increases gradually over time, especially after 105 years, but with significant protection extending out to one million years. Unlike many conceptual models, this calculation is not highly sensitive to infiltration increases related to long-term climatic changes; thus, the potential for spikes in release related to changes in infiltration is small.

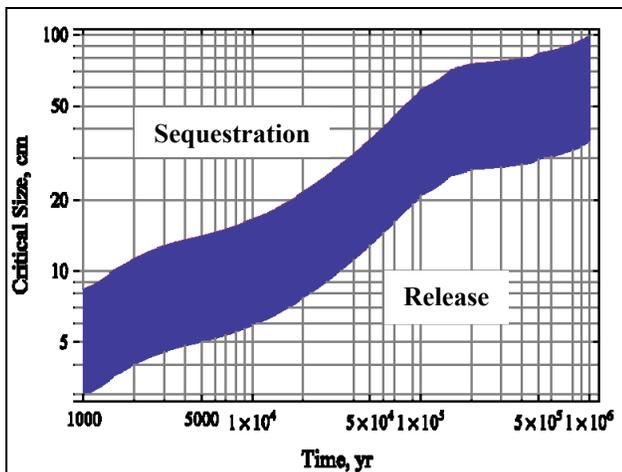


Figure 7. Minimum size of sheltered (no drip) area required to protect the waste from leaching. Sheltered areas larger than this size do not support radionuclide release.

VI. CONCLUSIONS

As Figure 7 demonstrates, the minimum area required for protection (sequestering the radionuclides and preventing their release) slowly increases over time, as the amount of heat generated from the waste package decreases. On the other hand, general corrosion decreases the average size of relic areas. 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, even in the case of early penetration by localized corrosion. In addition, colloids transported by advection into the warmest regions have very small diffusion coefficients. As a result, the release of radionuclides, attached to these colloids, by diffusion will be delayed more.

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