Conceptual Model and Controls on Radionuclide Release at Yucca Mountain and Multivariate Analyses of Water Chemistry: Surface and Ground Water Interactions

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I. INTRODUCTION

During the past year we have continued our work on evolution of groundwater chemistry and also began work on the processes controlling release of radionuclides from the Engineered Barrier System (EBS). The groundwater work during the past year is summarized in the paper published in Ground Water, the Journal of the National Groundwater Association: Woocay and Walton, *Multivariate Analyses of Water Chemistry: Surface and Ground Water Interactions*. This report summarizes our work to date on radionuclide release.

The release of radionuclides from the EBS is one of the most fundamental portions of the performance of the proposed repository at Yucca Mountain. Once radionuclides are released into the host rock most of them eventually make it out into the broader environment. Source term is an area with extremely high conceptual uncertainty at Yucca Mountain. Conceptual uncertainty (as opposed to parameter uncertainty) is not fully captured in performance assessment calculations such as the DOE Total System Performance Assessment (TSPA) process. For this reason areas identified by DOE as contributing to uncertainty may reflect mathematical projection of subjective judgments. In order to ensure protection of public health, improved understanding of the processes actually controlling radionuclide release at Yucca Mountain is important. In order to ascertain whether the solutions presented by DOE are conservative or realistic, one must have at least an approximate conceptual model – a picture in one's mind about what will happen. The goal of this work is to develop a more realistic conceptual model of radionuclide release from the EBS and to explore some of the implications of the revised conceptual model.

Release is currently described as a process of advection and diffusion from the waste containers (Baca and others, 2008). Advection is presumed to be driven by seepage into a flow-through package and diffusion is presumed to dominate in the absence of seepage. Whenever a liquid pathway of any type extends from the waste container, release begins. This release essentially never stops until depletion of the inventory and never reverses direction. This report explains how radionuclide release is more properly conceived in terms of counter fluxes of aqueous advective transport toward or away from the waste and diffusive transport away from the waste, as well as the balance of diffusive versus advective transport and advective transport direction control if and when radionuclides are released in the majority of situations.

The revised conceptual model differs substantially from the traditional approach of assuming that diffusive and advective liquid transport are in the same (outward) direction. It significantly changes the timing of radionuclide release from the repository.

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. Imagine that after initial penetration of the Alloy-22 the internal metallic components all begin to corrode at different rates. 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 zircalloy, may or may not proceed in an anticipated order. Much effort is spent by DOE and other organizations and analysts on maximum or worst case corrosion rates; less information is available on very long term passive corrosion rates of the type shown in Figure 1. Sometimes objects last much longer than anticipated. In particular, portions of the Alloy-22 may last indefinitely. 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 which pieces will last the longest. It is not guaranteed that the most corrosion-resistant materials will last longest.

Figure 1. Example of general corrosion that is non-uniform. North Percha Creek mine, New Mexico.

As parts lose structural integrity and corrosion products (which usually have a greater volume than the original metal) fill voids there will be 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 controlling processes on release rate it would seem likely that we could only be successful if the controlling processes were 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 2 gives the current design line load heat release for the repository. The line load is the average rate of heat release per unit length of drift in the axial direction. The graph indicates that the generation of thermal energy dies out after the first few thousand years and is near zero at 10,000 years.

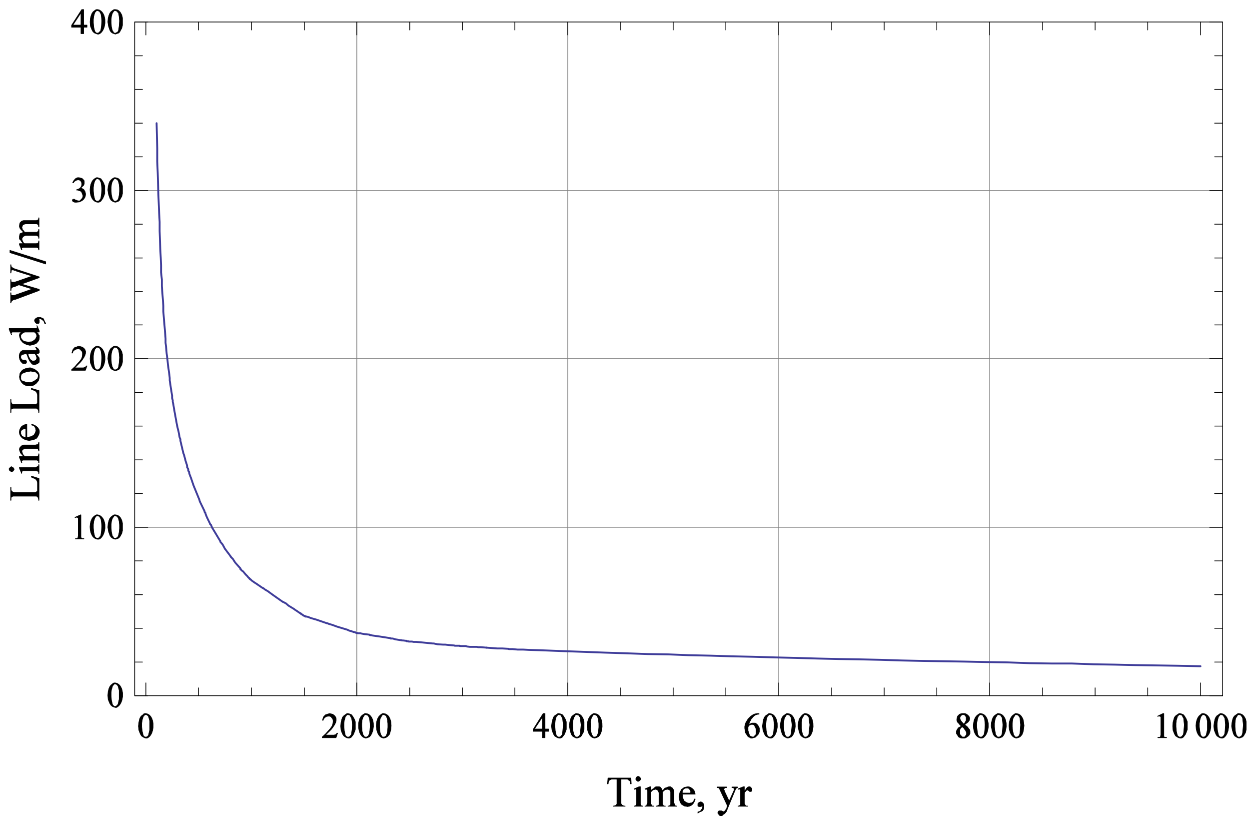

Figure 2. Heat release over time based upon the average line load. Heat release largely disappears after the first 10,000 years.

III. NATURAL ANALOGS

Given the complexity of the situation natural analogs seem appropriate. The following pictures were taken at the 1880's era silver mine on North Percha Creek, NM. At an unknown time presumably in the past 30 years, 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 upwards 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.

Figure 3. North Percha Creek mine berm with formation of efflorescent crusts.

Figure 4. Close-up picture of crusts. The evaporative concentration process appears to survive even in a wet system.

Figure 5. Crust formation in a different mine shaft on North Percha Creek, NM. Notice the proximity of standing water.

IV. FLOW AND TRANSPORT SYSTEM ANTICIPATED FOR FAILED WASTE CONTAINERS

In the mine, evaporation is driven by the low humidity air near the entrance. In Yucca Mountain the residual heat from the waste will drive localized evaporation and (perhaps) condensation cells. Evaporating water may condense within the volume of the initial waste package, or somewhere else in the drift.

In the current repository design, the waste package is underlain by a capillary barrier of crushed tuff called invert ballast. The capillary barrier from the crushed tuff means that, in the absence of direct seepage or condensation dripping at any specific location, water availability is limited as the water must either arrive in the vapor phase or wick through the capillary barrier of crushed tuff. Figure 6 is the rate of heat release based upon the line load plotted on a logarithmic scale. The figure indicates that heat release, albeit at a very low rate, occurs beyond one million years.

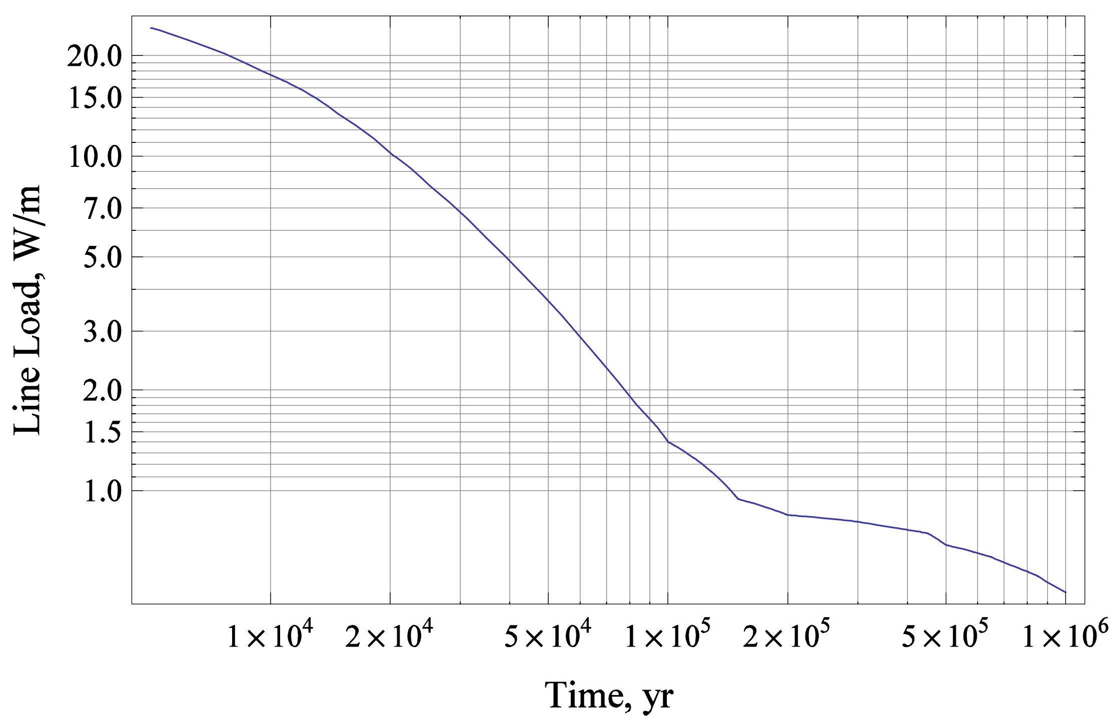

Figure 6. Line load on a logarithmic scale. Small but significant heat release occurs beyond one million years.

Predicted temperature increase from the outside to the inside of a hypothetical failed waste container is shown in Figure 7 over a range of potential thermal conductivities. Since the precise thermal properties of the rubble and how it changes with time are unknown, the figure is intended only to illustrate, in a semi-quantitative fashion, the amount of heat energy available.

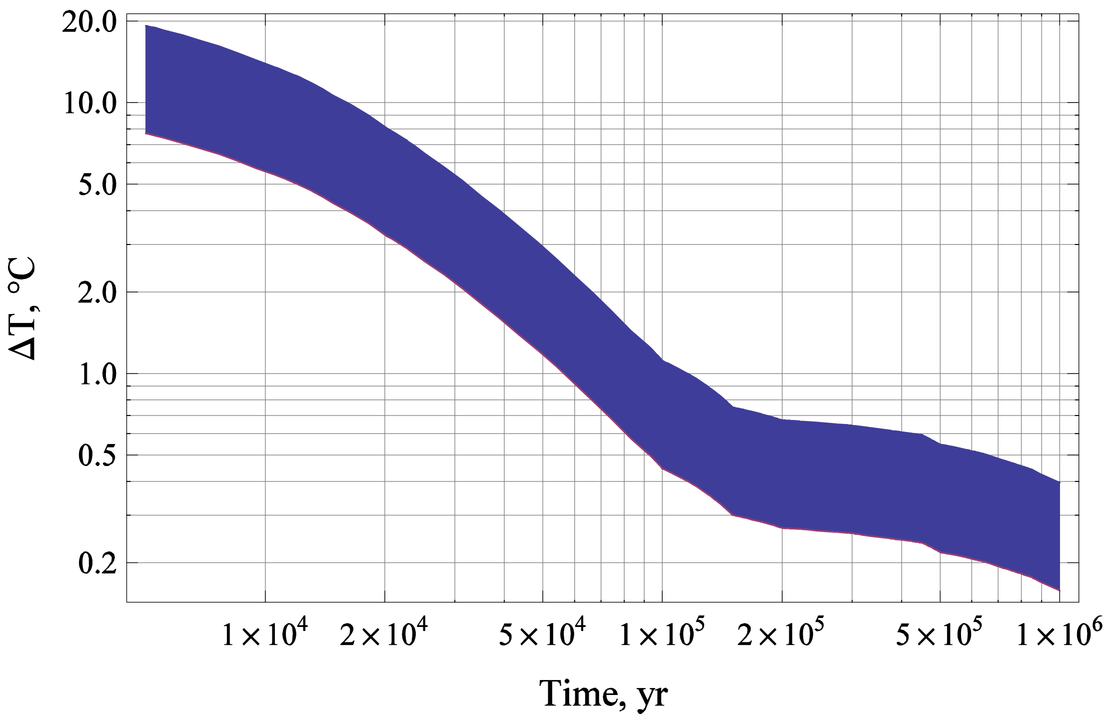

Figure 7. Plausible temperature rise from outside to inside of the rubble formed as waste containers fail. This simple graph assumes a range of thermal conductivity characteristic of soils.

The heat-driven flow system of interest is predominantly one where 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. In a quiescent system, such as the drifts tens or hundreds of thousands of years into the future, even very subtle forces can be important.

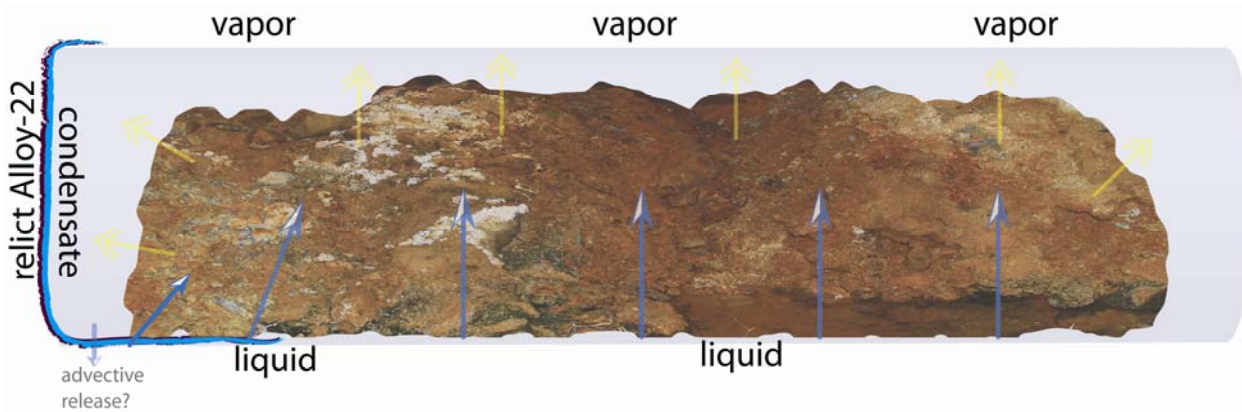

Figure 8. Flow system of liquid and vapor in rubble. This figure assumes a source of liquid water at the contact with the invert ballast (crushed rock) at the bottom of the figure. Liquid moves toward the hottest portions of the waste in response to capillary pressure gradients caused by evaporation at the thermally hotter portions of the waste. Water vapor moves in response to vapor pressure gradients sometimes into the drift and sometimes to cooler portions of relict Alloy-22 where it condenses. The flow of liquid water toward the waste will cause concentration of radionuclides at warmer regions and prevent most releases.

Figure 8 illustrates the conceptual flow and transport system. Since, in the absence of seepage or condensate formation at any particular point, the liquid flow is toward the heat source, liquid releases from these areas not only must be from diffusion but the diffusion is against the direction of advective transport; essentially the ions must "swim upstream" to escape. This is not the situation in precise locations with active dripping, which are anticipated to be relatively rare on a spatial basis.

Given that flow within this system is toward the majority of the radioactivity, 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:

$$Pe = VL/D$$

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 that exact geometries are not needed to make general conclusions. The interpretation of the Peclet number in this situation is 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.

If it is assumed that heat generated by the waste is available to evaporate water (if present), then 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 (Figure 9). Notice that we do not assume all water is evaporated. Diffusion coefficients for the graphs shown are estimated from the data of Conca and Wright, 1992. Other formulations such as Archie's Law give similar results. Since diffusive and advective transport are in opposite directions the Peclet number is negative. The equations predicting the Peclet number are solved for three different values of volumetric water content over time, assuming that the waste package degrades into a 1-meter high pile of rubble. The lowest value of volumetric water content (θ) shown is the one anticipated for the crushed tuff below the waste. For the case of crushed tuff (the edge of the capillary barrier) the anticipated volumetric water content is consistent with $Pe < (-1)$ for over a million years, suggesting that diffusional releases will, for the most part, not occur in the repository for the first one million years. Releases will be small and driven by advection at precise locations of seepage and condensation. Higher than anticipated volumetric water content will support diffusional releases after 300,000 years in the case modeled (volumetric water content = 0.2). Not surprisingly, the calculations indicate that very dry portions of rubble will not support liquid release.

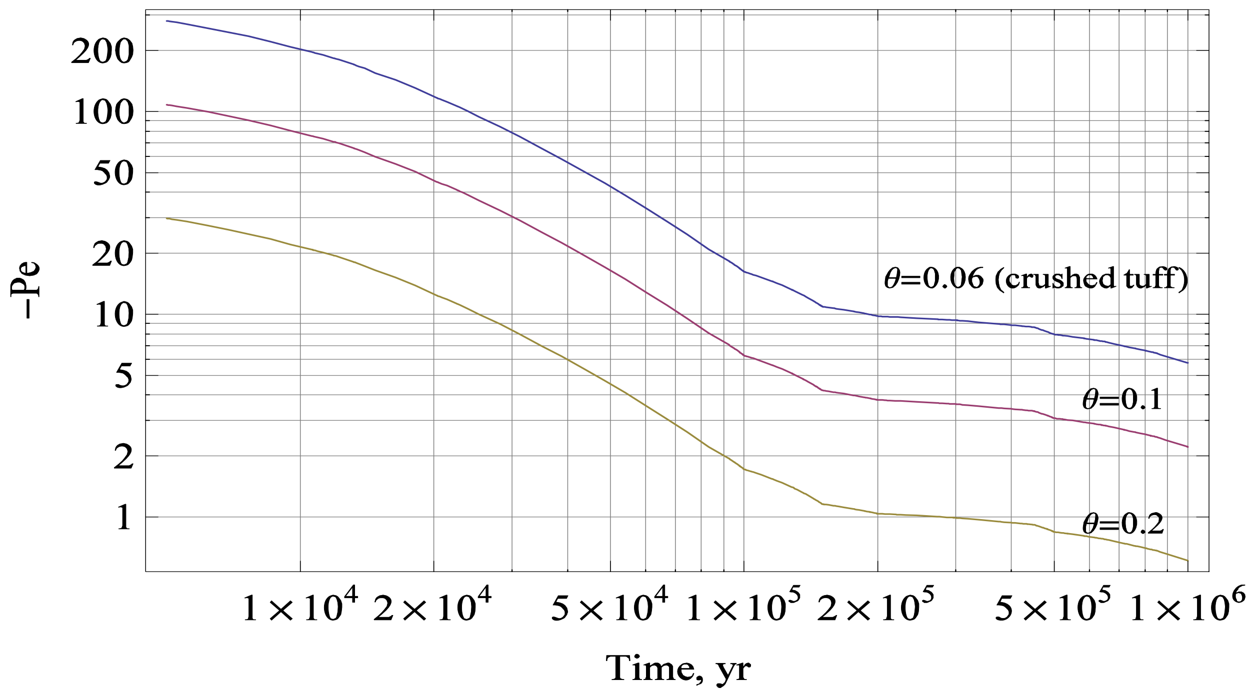

Figure 9. Calculation of the Peclet number over time. Calculation is based upon average line load heat release per unit volume of waste package. 100% of the heat release is assumed to go into evaporative cooling.

This simple calculation leads to several important conclusions:

- Low volumetric water content and capillary breaks (capillary barriers) are important factors in controlling release rate;
- Release occurs when the process described becomes overwhelmed by other factors such as direct dripping of seepage or condensate;
- Entry and exit of seepage to/from a waste package does not cause the entire waste package to be subject to release; and
- The process has the potential to last beyond regulatory time periods.

The process is shown schematically in Figure 10. In general, the Peclet number is negative with liquid flow toward the waste. Over time the Peclet Number increases toward zero (no heat-driven liquid flow). As the liquid flow slows, diffusional release will gradually begin to occur and increase with time. In locations of direct seepage, the heat-driven flow system is overwhelmed, leading to more rapid release of small portions of the inventory.

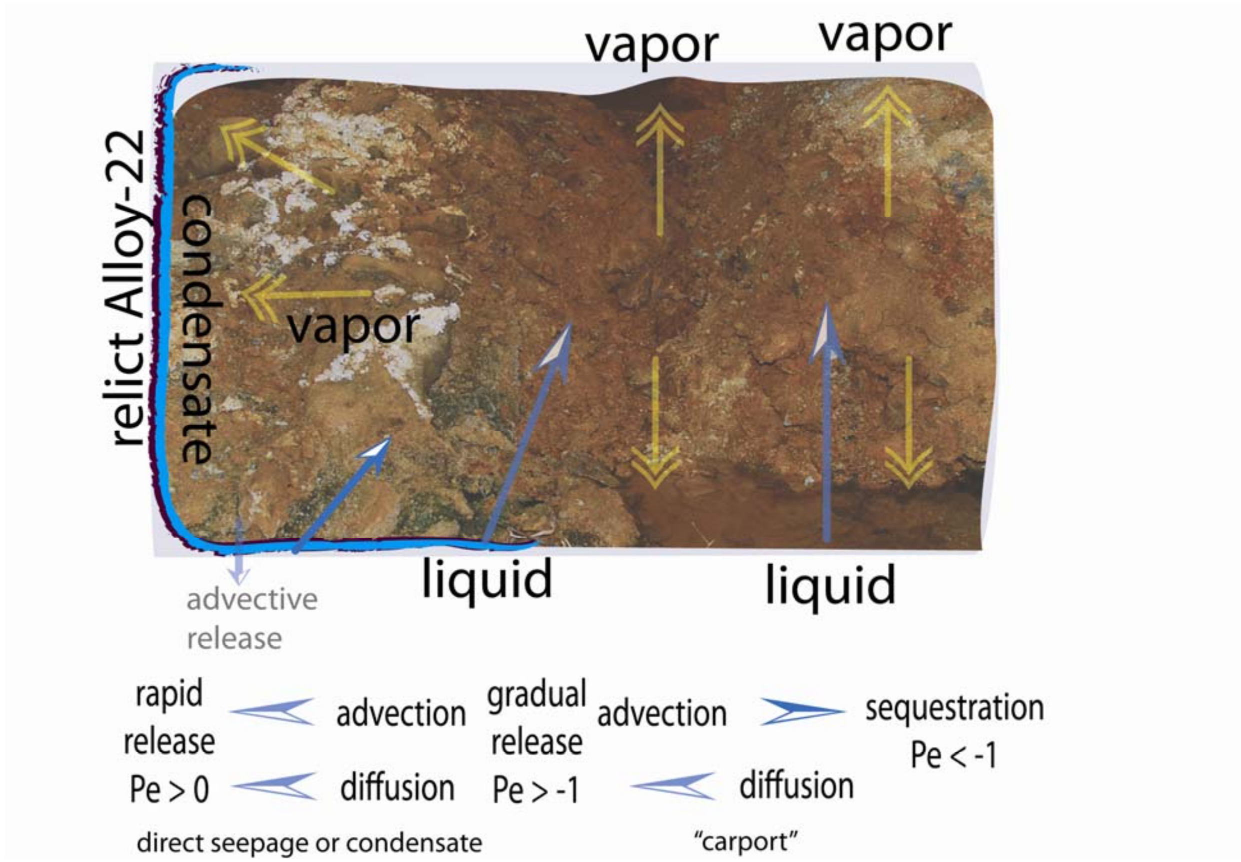

Figure 10. Schematic of flow and transport system inside a failed waste container. Release occurs only from the portion of the waste exposed to condensate flow. In areas sheltered from direct dripping or condensation ("carports") the waste initially concentrates in hotter regions followed by gradual release at very long (generally > one million years) time periods. Areas with direct seepage or condensation cause the natural inward flow direction to be overwhelmed, leading to more rapid release of small portions of the inventory.

A simple experiment illustrating the flow system is shown in Figure 11. An ordinary bathroom towel was immersed in water spiked with dye. Over time, the dye travels up the towel and becomes concentrated at the point where evaporation becomes complete. The system is one of sequestration, not release.

Figure 11. Dye moves upward and concentrates in a towel dipped in water spiked with 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.

V. COMPLICATING FACTORS

A number of factors will influence how well radionuclides are sequestered by the flow system described above. In this section the factors are organized as a set of issues.

a. *Issue 1*: Soluble salts may accumulate and defeat the flow system described.

If the capillary barrier at any location is poor, or an excess of soluble salts have accumulated near the drift – the case with an above-boiling repository in portions of the drift – then accumulation of soluble salts will eventually slow the evaporation process through vapor pressure lowering. In later time periods the thermal gradients are very subtle (tenths or even hundredths of a degree; Figure 7) and even low concentrations of soluble salts with associated vapor pressure lowering could counteract the subtle increases in water vapor pressure caused by the small temperature differences along the flow path. At steady state in such a system the Peclet number would be set to -1.0, as the inward movement of soluble salts by advection is exactly counterbalanced by their diffusional outward movement. The evaporation and inward flow of water do not cease, but rather slow down to the Pe = -1 point. This means that the release of radionuclides will be greatly slowed below the rate expected by diffusion, but may not be zero in all cases. “Salting out” of the flow system is more likely in the absence of a capillary barrier below the waste and in an above-boiling repository.

In an above-boiling repository very large accumulations of soluble salts will occur near the rock/drift interface (Danko et al., in press). An adequate experimental database with corresponding theoretical development does not currently exist to predict how this system will behave and evolve over time given that the salt-related vapor pressure lowering and hygroscopic nature will significantly influence how it behaves in a system with partially saturated fracture/matrix flow, heat transfer, and very high concentration gradients (saturated brine versus distilled water). This excess salt may migrate into the invert or waste, thereby leading to the protective flow system prematurely salting out.

Figure 12. Dissolved constituents concentrate in filter paper. Over time the filter paper becomes armored with the precipitate.

b. *Issue 2*: Heat transfer is mostly by conduction, not the evaporation assumed in the calculations.

Calculations used to estimate the Peclet Number simplistically assume that evaporation is the major source of heat loss from the exposed waste. In the general case of dry waste this assumption is clearly false as conduction will dominate the heat transfer. The Peclet number calculations (Figure 10) also indicate that isolation is more complete and all the heat energy is not required in dry systems. Dry systems by definition have low volumetric water content, meaning that only a fraction of the heat transfer needs to be in the form of evaporation in order to prevent most release.

As the moisture content increases the answer is more difficult. Consider that the geometry, transport properties, and boundary conditions for partially failed waste containers are infinitely variable and specifically unknown. Precise answers require arbitrary assumptions about fundamentally unknown properties of the system, and are of little utility. We can say that the heat transfer tends to be more by evaporation in wet systems and more by conduction in dry systems. Wet areas require more evaporation to provide protection and are likely to have more evaporation than dry areas. These are favorable characteristics for waste isolation. Preliminary calculations indicate that, for moist systems, about half of the local heat transfer is likely to be in the form of evaporation/condensation and this fraction declines only slightly with time after the first few thousand years. In addition, a portion of the measured thermal conductivity for moist porous materials (e.g., soils) represents pore-scale latent heat transfer that is lumped into the value of the thermal conductivity. This is why thermal conductivity of moist soils varies with temperature. Hiraiwa and Kasubuchi, (2000) found about 1/3 of the thermal conductivity in two soils at 20% volumetric water content could be attributed to pore-scale latent heat transfer at a temperature of 30°C. Since the pore-scale evaporation/condensation cells also lead to inward water flow, they count toward waste isolation, although they are not generally modeled.

We plan to perform heat transfer calculations to further explore this question.

c. *Issue 3*: Some waste packages will see seepage and/or condensate.

Advection related to seepage water and/or condensate will occur on some waste packages. Two extremes of advection-affected waste packages are possible: a) flow-through system with penetrations on the top and the bottom, and b) bathtub system with penetrations only on the top. To date we have not addressed the bathtub situation except to observe that the inward flow system and sequestration do not occur under saturated conditions (Figure 10). Flow-through systems and partial bathtubs are amenable to the treatment shown above. Consider that even in a flow-through situation, not all portions of the partially failed waste package will necessarily be exposed to significant advection. Imagine a carport or "umbrella" situation where relict pieces of Alloy-22 still shelter portions of the waste package from seepage.

Rather than assuming the dimensions of the initial waste package, one can solve for the dimension or size of waste in a stagnant area carport that is necessary to keep the Peclet number < (-1). This is shown in Figure 13 for water content of (0.05 [bottom] to 0.20 [top]). The degree of protection decreases with time but significant protection of portions of the waste package continues beyond one million years. The gradual increase in size required for protection over time convolved with the gradual decrease in average size of the carports with continued corrosion suggests a gradual release of waste over a very long time period with a peak likely beyond one million years in the future.

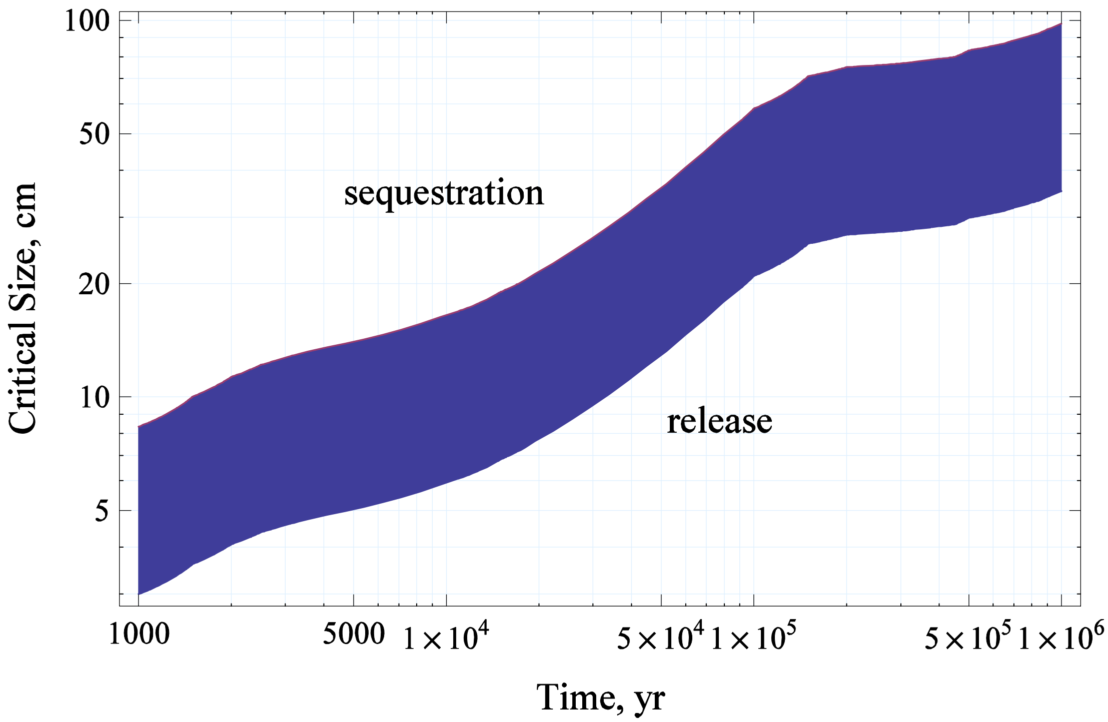

Figure 13. Minimum size of sheltered (no drip) area required to protect the waste from leaching. Sheltered areas larger than this size do not support radionuclide release.

d. *Issue 4*: The calculations are based on the average line load whereas some waste containers are hotter (and some are colder).

In general, containers with the most heavy metal (radioactivity) are thermally hotter and those with lower waste content tend to be colder (less hot). In the rubble of a partially failed waste package this will be true on finer scales. Some rubble will have high concentrations of waste; some rubble will be mostly metal corrosion products or rock. The key concept is that the heat and the waste are conjoint. This is one of the most important properties of the system; heat generating waste tends to accumulate, not disperse. The waste packages with the most waste will be better protected than those with less waste, and better protected than shown in the calculations included in this report. The waste packages with spent fuel have the most waste and are thermally hotter than the line load. Waste isolation in the hotter waste packages will work significantly better than shown in Figures 10 and 13.

e. *Issue 5:* The material properties will not be homogeneous and/or purely porous, as assumed in the calculations.

The rubble may eventually degrade to the point where the corrosion products create a heterogeneous porous media. During early stages of degradation a mixture of intact metals (zircalloy, stainless steel, Alloy-22) and spent fuel will be mixed with corrosion and spent fuel alteration products giving a mixture of porous and discrete (metal parts) pathway. The liquid transport pathway may become mostly discontinuous with transport occurring in hypothetical films of adsorbed water on metals along portions of the pathway between waste and invert ballast (crushed rock). This pathway may be constricted and tortuous and volumetric water content will vary spatially. This is a favorable property of the system. Waste will be isolated as long as $Pe < (-1)$ at any point along the transport pathway. The invert ballast is an example of one location along the pathway with estimated volumetric water content of 5%. Any point with sufficiently low liquid content in the liquid pathway will serve as a choke point and locally stop release. In the case of waste isolation, the strongest link in the chain dominates, not the weakest.

f. Issue 6: What will happen when the Peclet number begins to exceed -1?

First, consider that the Peclet number is indicative of the direction of mass transport, but dimensionless numbers do not precisely predict any system in detail. As the system cools, especially beyond one million years, the Peclet number at many locations will gradually rise above -1, leading to radionuclide release, beginning in a few locations at one hundred thousand years and accelerating beyond one million years. As the Peclet numbers rise, residual, protective portions of the engineered barriers are also failing. Convolution of the two processes, both of them slowly evolving, will lead to a gradual increase in radionuclide release that is likely to peak beyond one million years. At this point most of the initial radioactivity will now be gone through natural attenuation. Even when the Peclet number is between -1 and zero, advective flow is still toward the waste and will slow release below the rate expected from diffusion. Interestingly, the mobility of the waste is approximately inverse to its toxicity (activity).

g. Issue 7: The inward flow system will cause precipitation of calcium carbonate, silica, and radionuclides on the waste.

Depending upon the connectivity of the liquid pathway between the waste and rock, evaporative concentration will cause precipitation of low solubility compounds such as calcium carbonate and silica on the waste. It is likely that this will tend to armor the waste as illustrated in the simple laboratory experiment shown in Figure 12. Armoring will be greatest for waste packages that experience seepage during the first 10,000 years when potential evaporation rates are greatest. Radionuclides released from the cooler portions of the waste package will concentrate in the hottest sections. Low- and moderate-solubility radionuclides will tend to move toward the heat and precipitate. Radionuclides such as radium will likely co-precipitate with calcium carbonate. High solubility radionuclides such as Technetium may obtain steady state concentration profiles with greater concentration in warmer regions. The effect of the armoring of the waste is unknown but likely to be favorable to waste isolation. Differences in solubility are likely to lead to some degree of separation of the radionuclides initially mixed together in the spent fuel. This separation is analogous to the separation outside the waste package discussed by Hall and Walton, 2006.

h. Issue 8: Much of the radionuclide release is in colloidal form.

The Peclet number approximates the rate of advection to diffusion. Colloids diffuse by Brownian Motion at a rate on the order of 1,000 times slower than dissolved constituents (Blaaderen et al, 1992). For colloids the Peclet numbers shown can all be multiplied by 1,000 meaning that diffusional release of colloids will not occur within the first million years.

VI. DISCUSSION

A new conceptual model is developed for release of radionuclides from the Engineered Barrier System at Yucca Mountain. A series of internal flow cells at different scales and sometimes in different directions are postulated to form. The internal flow cells tend to concentrate (sequester) radionuclides, other dissolved constituents, and mobile colloids. The flow cells consist of flow of liquid water toward localized heat sources in response to evaporation and water vapor transport away from the heat source. The new conceptual model does not postulate that all water entering the waste package will evaporate, only that evaporation/condensation cells will cause liquid movement predominantly toward the waste. Initially (< 100,000 years) these cells are very strong and robust. If a large supply of water is available they may lead to formation of deposits (“nodules”) of radionuclides, silica, and calcium carbonate. Over periods of hundreds of thousands of years the cells decline in strength, but remain active and important at times greater than one million years. Sequestration is based upon inward liquid phase mass transport being rapid relative to outward diffusional mass transport. Radionuclide release from this system will initially be spotty – only in precise areas of strong seepage and condensation. Over time the internal flow systems decrease in strength, allowing for a gradual release of radionuclides with a peak perhaps beyond one million years, even in the absence of protection from intact metallic barriers. In essence, heat release, one of the fundamental properties of the waste, makes it inherently safe for disposal in partially saturated environments.

A number of implications flow from the work shown above. These are:

- Heat generating waste placed in a partially saturated environment is inherently safe in the sense that it naturally tends to accumulate rather than disperse;
- The effect is stronger at higher rates of heat generation but continues to be significant at time scales of one million years or more;

- The natural process of accumulation can be enhanced by placing the waste inside a capillary barrier in a below-boiling repository;
- Bathtub situations within partially failed waste containers appear to be problematic but could be reduced in probability by enlightened design;
- Salting out of the protective flow system is more likely and less predictable in an above-boiling repository;
- The natural tendency for concentration rather than dispersal is amenable to treatment by a simple dimensionless group analysis, greatly reducing the need for over reliance on the "black box" of Total Systems Performance Assessment models;
- This is an example of the type of mechanism and simple quantitative treatment that can be used to develop a safety case; and
- Several aspects of the mechanism described are uncertain and have not been fully investigated.

This work is in its initial stages. All limitations and implications of the work have not been fully explored. Calculations have not yet been fully checked.

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