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# Controlling Features and Processes for Radionuclide Release from Unsaturated Zone Geological Repositories

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**CONTROLLING FEATURES AND PROCESSES FOR RADIONUCLIDE  
RELEASE FROM UNSATURATED ZONE GEOLOGICAL  
REPOSITORIES**

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Lubna Kamel Hamdan

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## **Dedication**

I dedicate this work to my dear family, especially  
my beloved parents. Their support,  
encouragement, and prayer helped me overcome  
the difficulties and eased the pressure that I faced  
during my PhD study

**وَقُلْ رَبِّ زِدْنِي عِلْمًا**

**And say: O my Lord increase me in  
knowledge.**



CONTROLLING FEATURES AND PROCESSES FOR RADIONUCLIDE  
RELEASE FROM UNSATURATED ZONE GEOLOGICAL REPOSITORIES

by

LUBNA KAMEL HAMDAN, B. S., M. S

DISSERTATION

Presented to the Faculty of the Graduate School of

The University of Texas at El Paso

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of the Requirements

for the Degree of

DOCTOR OF PHILOSOPHY

Environmental Science and Engineering Program

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## **Abstract**

In the long run, nuclear waste packages will fail gradually due to localized and general corrosion and radionuclides will be transported to the accessible environment by ground water. Two main failure scenarios are expected for the waste packages: Flow-through model and bathtub model; in the flow-through model water flows through the waste container, while in the bathtub model water pools inside the waste package. The Department of Energy, in their performance assessment of the proposed repository at Yucca Mountain, excluded the bathtub model from their analysis and assumed the diffusion and advection to be in the same direction in the flow-through model. In this research, a new conceptual model is introduced for radionuclide release from a flow-through category failed waste container. Due to the residual heat release of the nuclear waste, this model expects a bidirectional transport for radionuclides in the sheltered areas; advection toward the warmest region and diffusion in the opposite direction. As a result, a sequestration process is expected for the radionuclides in the sheltered areas, which would delay the radionuclide release and make it gradual. The analysis showed that the sequestration will be more effective at higher rates of heat generation and lower moisture conditions. As for the bathtub model, conditions inside the bathtub category failed waste container are investigated. The analysis showed that temperature stratification is expected in the lower half of the waste package, which would limit oxygen availability and slow the corrosion of the spent nuclear fuel and other waste package components. As a result, the estimated life for all waste package components will be longer and consequently radionuclide release will be delayed.

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# Chapter 1

## 1. General Introduction

After twenty years of little or no net growth in installed nuclear power capacity in most of the countries, there is today again great interest in nuclear power. Many energy analysts expect now a notable nuclear energy renaissance worldwide, motivated by common global concerns about: global warming, energy independence, and energy security. The expected expansion of nuclear energy use will result in hundreds of thousands of metric tons of spent nuclear fuel (SNF) (Feiveson, 2007), (Gentenaar, 2009).

To produce electricity, nuclear reactors use fuel made of solid ceramic pellets of enriched uranium ( $\text{UO}_2$  powder) sealed in strong metal tubes usually of zircalloy or stainless steel. The tubes are bundled together to form a nuclear fuel assembly. When the uranium pellets are no longer efficient for producing electricity the assembly is removed from the reactor. The removed assembly, now called SNF, is highly radioactive and gives off thermal energy. Therefore, the removed fuel assemblies are stored in special water pond located at the reactor site to allow the decrease of their heat release and radioactivity, or they can be dry stored in engineered facilities, cooled by air. However both kinds of storages are interim step intended only to make it easier to handle the SNF due to the radioactivity decay. After the temporary storage, the SNF is either reprocessed to recover the usable portion of it or sent to long-term storage and final disposal without reprocessing. (World Nuclear Association, 2006).

To solve the problem of SNF accumulation, permanent disposal sites are needed. Among technical experts, the accepted method for radioactive waste disposal is to contain the waste and isolate it from the accessible environment. Scientists and researchers worldwide have studied

various ways to dispose nuclear waste; they considered disposal in ocean floor, space, ice sheets, and remote islands. They agreed that the emplacement of radioactive waste at significant depths underground, i.e. the geological disposal, is the best option for isolating it from the environment accessible to humans. In a geological repository, waste containment and isolation are provided by: the containers into which the nuclear waste is put before being emplaced in the repository, various additional engineered barriers, and the natural barrier provided by the host rock, see Figure 1.1. In addition, geological disposal assures long term safety without surveillance and maintenance (IAEA, 2003).

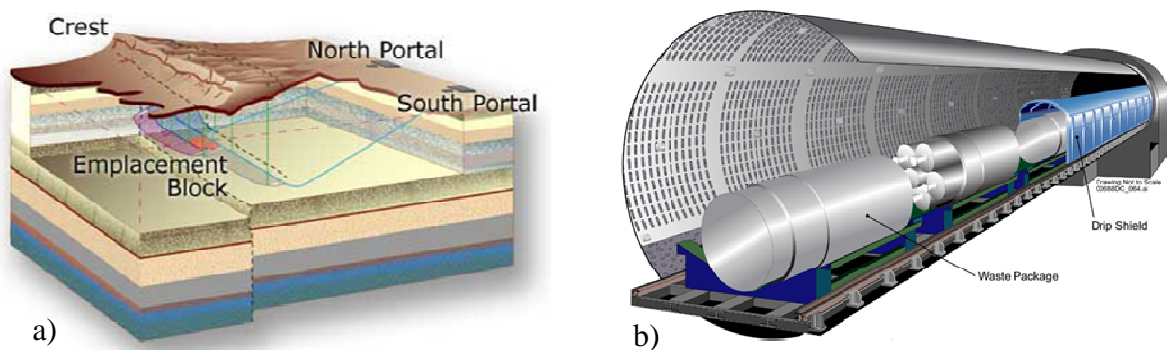


Figure 1.1 a) Natural barriers include the rock, the soil, and other natural features of Yucca Mountain. b) Engineered barriers include the waste container, the drip shield, and the repository construction (DOE, 2007 a).

Up to December 2005, the United States accumulated about 53,440 metric tons of spent nuclear fuel. In addition, there will be about 22,000 canisters of solid high-level radioactive waste (highly radioactive material associated with weapons production) for future disposal in a repository. By the year 2035 this amount will increase to an estimated 119,000 metric tons (DOE, 2007 b). The DOE planned to construct a geological repository at Yucca Mountain (YM) to dispose SNF and high-level radioactive waste. Yucca Mountain is located on federal land in a

remote area in Nye County, Nevada, about 100 miles northwest of Las Vegas, see Figure 1.2. YM was chosen as a site for the geological repository because its natural characteristics aid in repository performance. These Natural characteristics include: a semiarid climate, relatively stable site geology (the YM area has a low incidence of large magnitude seismic activity and volcanic activity in the YM region has declined through recent geologic time), a deep water table, and limited groundwater flow through the unsaturated zone (UZ) and the saturated zone (SZ) (SNL, 2008 ). The proposed nuclear repository inside YM is to be built between 660 and 1400 feet (201 - 427 m) below the mountain's surface and between 570 and 1,200 feet (174 - 366 m) above the water table. The DOE would emplace around 70,000 MTHM of SNF and high-level radioactive waste in the repository (OSTI, 2000).



Figure 1.2 Location of Yucca Mountain in Nevada state (Hamburger, 2006).

To protect people and the environment, the design of YM repository depends on natural and engineered barriers to isolate the nuclear waste and keep it dry as long as possible, Figure 1.1 shows the design of YM repository. However, in the long run nuclear waste packages will fail due to localized and general corrosion and water will have access to the nuclear waste. As a result, radionuclides will be transported to the accessible environment by groundwater.

The license application, to be submitted by DOE to obtain the Nuclear Regulatory Commission (NRC) license to construct a repository in YM, should present scientifically defensible analyses of how the repository will perform over 10,000 years. To assess the behavior of the repository over 10,000 years and beyond, DOE used a computer model called “Total System Performance Assessment (TSPA) Model/Analysis for the License Application”. This model uses data and results from individual process models to assess the performance of all parts of the repository when they work together to keep the waste isolated. The TSPA model also simulates the effect of damaging events, such as earthquakes and volcanoes, on the repository total performance and calculates the consequences to the people and environment (SNL, 2008).

Release of radionuclides from the Yucca Mountain repository is a key factor determining the repository performance. Release rate calculations are performed by the Engineered Barrier System Radionuclide Transport Abstraction (EBS RT). The purpose of the EBS RT Abstraction is to provide the conceptual model, used in the TSPA model, to determine the time-dependent flux of radionuclides from the EBS to the UZ. This model consists of two main components: a flow model and a transport model. The flow model defines the pathways for water flow in the EBS and computes the flow rate in each pathway; Figure 1.3 shows the potential flow pathways in the EBS. The transport model considers both advective and diffusive transport from a breached waste package. The advective transport occurs when the dissolved radionuclides (or

absorbed onto collides) are carried from the waste package by the seepage flux, which passes through waste package breaches. Diffusive transport, occurs as a result of a gradient in radionuclides concentration, is the sole way of transport when there is no flow through a waste package. This situation is assumed to predominate during the regulatory compliance period of the repository (SNL, 2007).

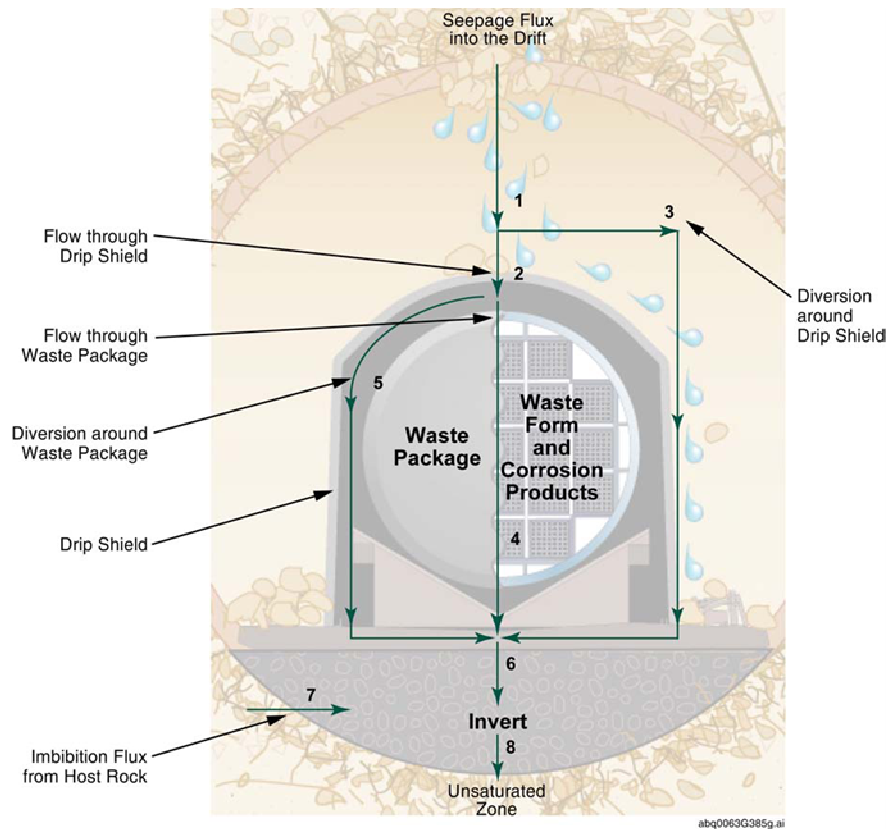


Figure1.3 The potential pathways for water flow in the EBS (SNL, 2007).

Source term (the transport mechanisms determining the amounts and types of radionuclides released) is an area with extremely high conceptual uncertainty at Yucca Mountain. Conceptual uncertainty is not fully captured in performance assessment calculations such as the DOE TSPA model. 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. The goal of this research is to develop conceptual models of radionuclide release from the EBS and explore some of the implications of the revised conceptual models. These models are based on natural processes anticipated to take place inside a partially failed waste container as a result of persistent heat release from the nuclear waste.

Two main scenarios are expected for waste packages failure: flow-through model (in which the waste package is penetrated from top and bottom and water flows through it) and bathtub model (in which the waste package is penetrated from the top only causing water to pool inside it); Figure 1.4 shows schematic diagrams for both kinds of failure. In the flow-through model, DOE assumed that advective transport and diffusive transport are in the same direction (Figure 1.4 a). This research considered the effect of partial evaporation for water entering a flow-through category failed waste container, which would lead to bidirectional transport (the diffusive transport is in opposite direction to the advective transport). The bathtub model was screened out by DOE as an alternative conceptual model to the flow-through model, in order to overestimate radionuclide transport (SNL, 2007). This research, made a detailed study for the bathtub model, considering two possibilities: temperature stratification of the pooled water versus mixing process and their effect on oxygen availability and radionuclide release.



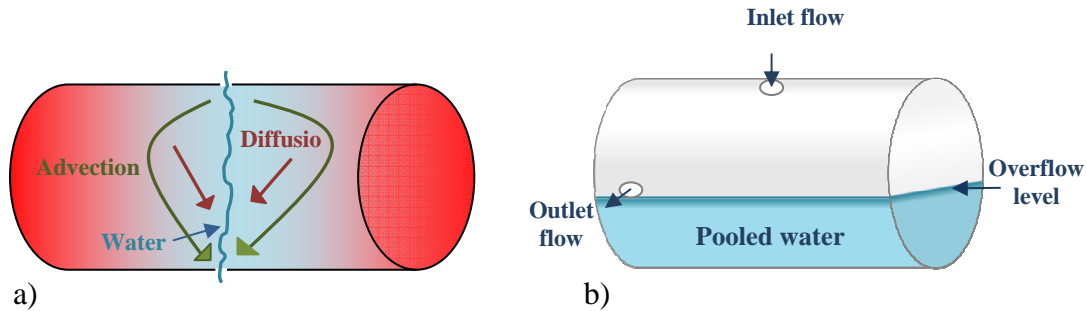


Figure 1.4 Schematic diagrams for the main scenarios of the waste packages failure: a) Flow-through model. b) Bathtub model.

**Finally, it is worthwhile to mention that although the calculations of the models introduced in this research are based on the EBS of the proposed repository at YM, these models apply, in general, to any unsaturated zone geological nuclear waste repository. These models are based on a key characteristic of the nuclear waste, heat production, and the geology, unsaturated zone location.**

## REFERENCES

Department of Energy: Office of Scientific and Technical Information (OSTI). (2000, October).

*Yucca Mountain facts at a glance*. Retrieved February 2010, from Information Bridge:

DOE Scientific and Technical Information:

<http://www.osti.gov/bridge/purl.cover.jsp;jsessionid=869DF1CD6BF3193F9353D81F9B92D2AA?purl=/860320-0j7JQv/>

Feiveson, H. A. (2007). Faux Renaissance: Global Warming, Radioactive Waste Disposal, and the Nuclear Future. *Arms Control Today* , 37, 13-17.

Gentenaar, H. (2009). The Nuclear Energy Renaissance. *BusinessWeek* , Electronic document, available at [http://www.businessweek.com/adsections/2009/pdf/05.04.09\\_Nuclear.pdf](http://www.businessweek.com/adsections/2009/pdf/05.04.09_Nuclear.pdf).

Hamburger, L. (2006). *Yucca Mountain: A Nuclear Waste Repository*. Retrieved 2010, from  
Volcanoes of the Eastern Sierra Nevada:

<http://www.indiana.edu/~sierra/papers/2006/hamburger.html>

International Atomic Energy Agency, IAEA. (2003). *THE LONG TERM STORAGE OF  
RADIOACTIVE WASTE: SAFETY AND SUSTAINABILITY*. Vienna, Austria: Electronic  
document, available at [www-pub.iaea.org/MTCD/publications/PDF/LTS-RW\\_web.pdf](http://www-pub.iaea.org/MTCD/publications/PDF/LTS-RW_web.pdf):  
IAEA.

Sandia National Laboratories, SNL. (2007). *EBS Radionuclide Transport Abstraction, ANL-WIS-  
PA-000001 REV 03*. Las Vegas, Nevada: Sandia National Laboratories. ACC:  
DOC.20071004.0001. section 6.3.4.

Sandia National Laboratories, SNL. (2008 ). *Total System Performance Assessment  
Model/Analysis for the License Application, MDL-WIS-PA-000005 REV00*. Las Vegas,  
Nevada: Sandia National Laboratories, DOC.20080204.0003, section 6.3.2.

U.S. Department of Energy, DOE. (2007 a). *OCRWM-Engineering and design*. Retrieved  
December 6, 2007, from  
[http://www.ocrwm.doe.gov/ym\\_repository/studies/engdesign/index.shtml](http://www.ocrwm.doe.gov/ym_repository/studies/engdesign/index.shtml)

U.S. Department of Energy, DOE. (2007 b). *OCRWM-How Much Nuclear Waste Is In United  
States*. Retrieved September 12, 2007, from  
[http://www.ocrwm.doe.gov/ym\\_repository/about\\_project/waste\\_explained/howmuch.shtml](http://www.ocrwm.doe.gov/ym_repository/about_project/waste_explained/howmuch.shtml)

World Nuclear Association. (2006). *The Nuclear Fuel cycle*. Electronic document, available at  
<http://www.world-nuclear.org/education/nfc.htm>.

## **Chapter 2**

### **Limitations and Patterns of Radionuclide Release from Partially Failed Containers**

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## **2. 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.*

## **2.1 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.

## **2.2 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 2.1 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.

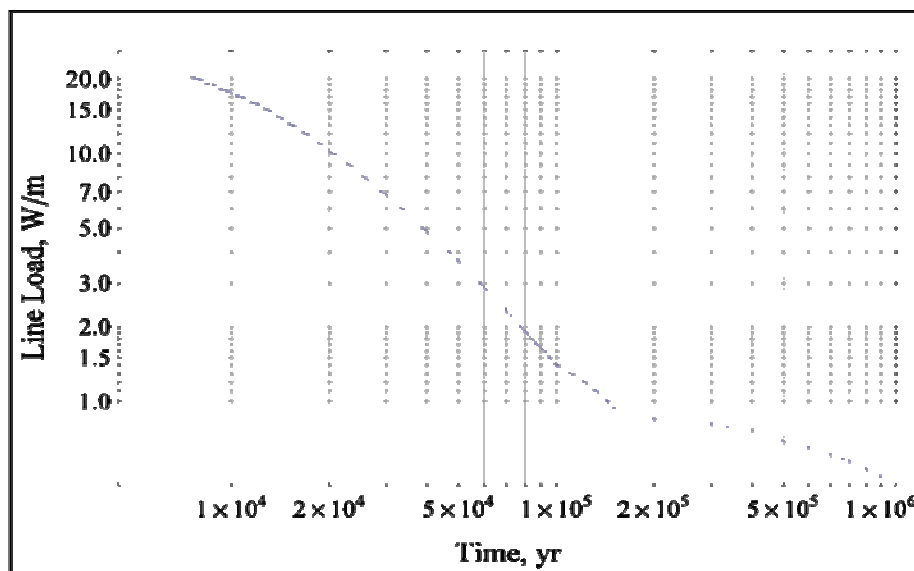


Figure 2.1 Line load on a logarithmic scale. Small, but significant, heat release occurs up-to one million years.

### 2.3 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 2.2). Diffusion-only release is included in this model.

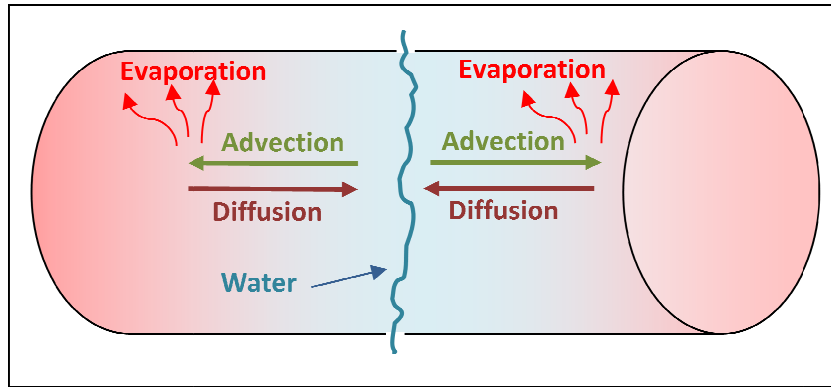


Figure 2.2 Schematic drawing showing physical processes in a flow through system.

Predicted temperature increase from the outside to the inside of a hypothetical failed waste container is shown in Figure 2.3, 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 ( $\text{W}/\text{m}^3$ )

$r_o, r$  = radii of the cylinder surface and the inner point (m)

$K$  = thermal conductivity (W/ (m°C))

Since the precise thermal properties of the rubble and how it changes with time are unknown, Figure 2.3 is intended only to illustrate, in a semi-quantitative fashion, the amount of heat energy available.

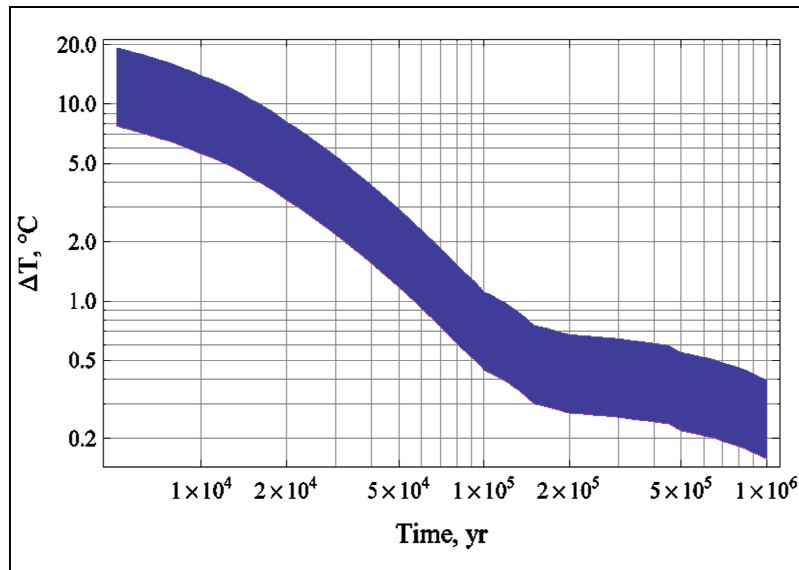


Figure 2.3 Plausible temperature rise from outside to inside of the rubble.

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 2.2).

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<sup>2</sup>/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 2.4. 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 2.4 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

## 2.4 NATURAL ANALOG

Another example illustrating the physical processes involved in this conceptual model comes from nature. Figure 2.5 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 2.6.



Figure 2.5 North Percha Creek mine berm with formation of efflorescent crusts.



Figure 2.6 A close-up picture of crusts. The evaporative concentration process appears to survive even in a wet system.

## 2.5 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 2.7 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.

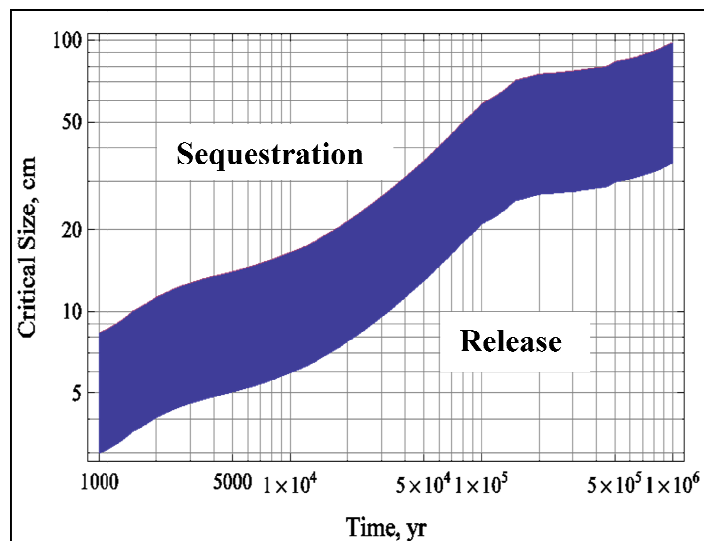


Figure 2.7 Minimum size of sheltered (no drip) area required to protect the waste from leaching. Sheltered areas larger than this size do not support radionuclide release.

## **2.6 CONCLUSIONS**

As Figure 2.7 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 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, 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.

## ***ACKNOWLEDGMENT***

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## **REFERENCES**

- [1] Jury, W.A., W.R. Gardner and W.H. Gardner. 1991. Soil Physics, pp. 181. Fifth Edition.  
New York: John Wiley & Sons, Inc.
- [2] Carslaw, H.S. and J.C. Jaeger. 1959. Conduction of Heat in Solids, pp. 191. Second Edition.  
Oxford, UK: Oxford University Press.
- [3] Conca, J.L. and J. Wright. 1992. "Diffusion and Flow in Gravel, Soil, and Rock." Applied  
Hydrogeology, 1, 5-24. Hanover, Germany: Verlag Heinz Heise GmbH. TIC: 224081.

## **Chapter 3**

### **Safety Implication for an Unsaturated Zone Nuclear Waste Repository**

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### **3. Safety Implication for an Unsaturated Zone Nuclear Waste Repository**

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## **ABSTRACT**

In this paper we shed some light on the safety of unsaturated zone nuclear geological repositories in the long run, by examining the effect of physical and chemical processes that take place inside a partially failed nuclear waste container. Our analysis addresses the safety of the proposed nuclear repository at Yucca Mountain, which is intended to store high-level nuclear waste. Our study is independent of the U. S. Department of Energy (DOE) analysis, which involves a number of complex computer codes and assumptions, and relies on the performance of an engineered barrier system. Our safety analysis could be applied in general to any geological repository designed to be in an unsaturated zone, since it is based on the geology, unsaturated zone location, and a key characteristic of the waste, heat production. This analysis shows that the radionuclide release from a partially failed waste container, stored in an unsaturated zone geological repository, is likely to be gradual and long delayed.

**Keywords:** Geological repository, Advective transport, Radionuclide sequestration.

## **3.1 INTRODUCTION**

In December 2006, an international workshop focused on the infrastructure issues that must be addressed before embarking on a nuclear power program, was held at the International Atomic Energy Agency's headquarters in Vienna. Twenty nonnuclear countries (potential new entrants to nuclear energy use) participated in this workshop which reflects the growing global interest in the nuclear energy (ANS, 2007). This renewed interest in nuclear energy is a result of an increasing concern for the global climate and the realization that greenhouse gas emissions,



which mainly result from the burning of fossil fuels, need to be reduced (Nuttall, 2005). Some scientists believe that nuclear power should not be favored since there are more effective ways to reduce carbon emissions, for instance: improvements in end-use efficiency (the efficiency of converting the energy delivered to the point of end use, such as electrical energy, into desired energy services such as hot showers and cold drinks (Lovins, 2005), improvements in the efficiency of the power sector itself, and the future availability of other low-carbon electricity supply options such as coal plants with carbon capture and storage, and wind farms (Feiveson, 2007). Nevertheless, nuclear power will play an increasing role, at least, as part of the global clean energy mix in the future, to meet the predicted global surge in electricity demand, and to increase energy security and independence (Squassoni, 2007).

Although nuclear power is considered clean energy, it has major environmental (radiation exposure) and security (nuclear proliferation) concerns regarding the treatment of spent nuclear fuel (SNF). Some countries reprocess the SNF for possible future use. Other countries use the strategy of interim storage of the SNF with the object of either ultimate disposal in a geological repository or making a later decision (reprocessing or emplacement in a geological repository) (Feiveson, 2007). Since there will be inevitable expansion of nuclear power use in the future, it is projected that hundreds of thousands of metric tons of SNF will accumulate. Reprocessing of SNF reduces the amount of nuclear waste and its decay heat, but it does not eliminate nuclear waste altogether, which eventually needs to be disposed in a safe and permanent place (Vujic, 2007). Geological repositories are reasonable solution for the problems of accumulated SNF. Inside a geological repository, such as the proposed repository at Yucca Mountain, large quantities of SNF can be disposed permanently in a secure and environmentally safe place. Disposal of nuclear waste in a geological repository will reduce the nuclear proliferation risks

(Feiveson, 2007) and protect people and the environment. In addition, recent work has shown that geological disposal can be developed in stages, such that the possibility of retrieval is retained for a long time (IAEA, 2003), which allows spent fuel retrieval for reprocessing purposes in the future, once efficient and suitable reprocessing technologies are available (Vujic, 2007)

### **3.2 CONDITIONS INSIDE THE GEOLOGICAL REPOSITORY AND WASTE PACKAGES IN THE LONG RUN**

The main purpose of the unsaturated zone geological repository is to shield nuclear radiation (by layers of soils and rock), and keep the nuclear waste dry as long as possible to prevent the release of radionuclides by corrosion of the storage canisters and transport by water to the accessible environment. The DOE, after extensive studies, chose to locate the geological repository at Yucca Mountain (an arid region) in the unsaturated zone, and to protect the nuclear waste with multiple engineered barriers to isolate them from water as long as possible (DOE, 2001). In the first period after the permanent closure of the repository, a zone of boiling to above-boiling conditions is expected to form through and around the emplacement drift (dry-out zone). As the heat output from the waste packages declines with time, due to radioactive decay, the dry-out zone shrinks and eventually the condensed water will be available to drip inside the emplacement drift (SNL, 2007a). Over time, due to physical and chemical disturbances in the repository environment, engineered barrier system and waste packages are likely to corrode at different rates and different locations (general and localized corrosions), and ultimately water will have access to the nuclear waste. As the waste package fails, small cells of evaporation and condensation are expected to build up inside the waste package, and leave some portions of the waste package materials in more harsh corrosion environments than others. Under these

different rates of corrosion, the waste package is anticipated to evolve into a mixture of corroded material and intact relicts of Alloy 22 and other waste package components.

Meanwhile, heat release from nuclear waste will continue, in general, apart from the evolving material properties of the waste package. The average heat release per unit volume over time for two types of waste packages intended to be stored in the repository are shown in Figure 3.1. The first type is pressurized water reactor [21PWR], with the highest average heat release, and the second type is defense glass logs [DHLW-L], which has the lowest average heat release (SNL, 2008 a). Figure 3.1 indicates that although heat release from nuclear waste packages declines over time, it persists (at very small rates) beyond one million years.

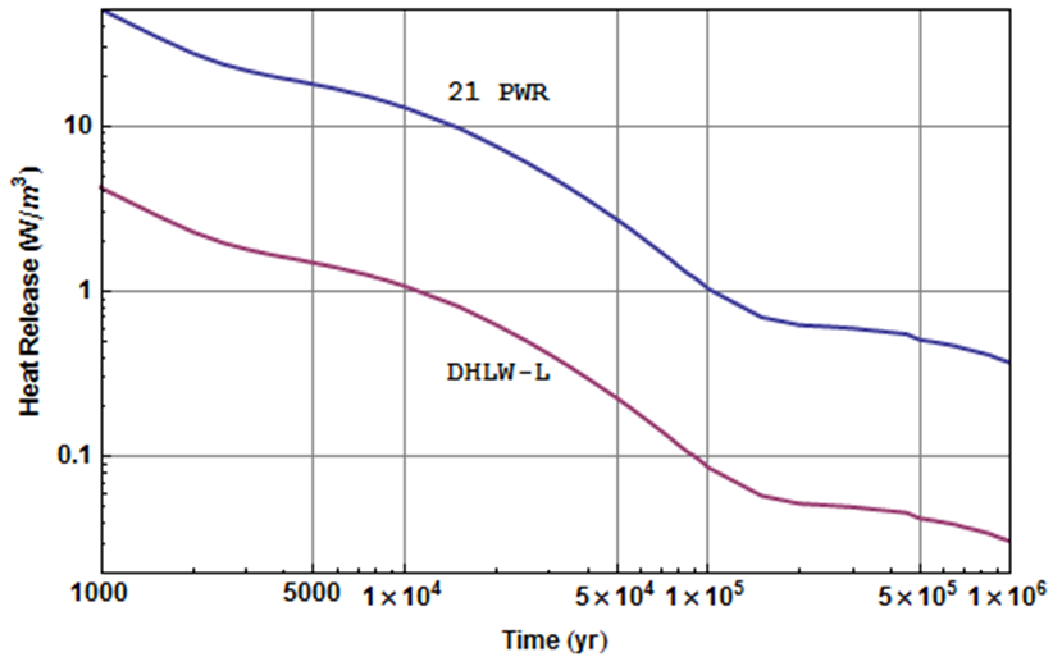


Figure3.1 The rate of heat release per volume from radioactive waste packages over time, plotted on a logarithmic scale. Small, but significant, heat release continues up to one million years.

In summary, in the long run, the interior of a waste package is likely to be a combination of porous material (the corroded material) and discrete pieces of Alloy 22 and other waste

package objects, present in a moist environment accompanied by residual little heat release. What effects might these factors have on radionuclide release from partially failed waste containers?

### **3.3 THEORY**

To examine the effect of the moisture inside a partially failed waste container on radionuclide release in the presence of a little heat release, we first need to shed some light on two phenomena: heat flow in soil or porous media (since the interior of the waste package is a mixture of porous material and discrete objects), and a natural sequestration of minerals, in North Percha Creek mine, NM, formed by a process similar to those anticipated to exist inside a partially failed waste container in the long run.

#### **3.3.1 Heat flow in soil (or porous media)**

The two main processes of heat transfer in soil are conduction, which refers to heat transport by molecular collisions, and convection of latent heat (energy per mass required for vaporization), which refers to latent heat energy transport in water vapor (in a porous media at low temperature and low temperature gradient radiation and convection could be neglected). Movement of water vapor from one location to another in the soil constitutes a transport of heat in a latent form because, during its movement, water vapor goes through internal condensation-evaporation phase changes which will result in heat energy liberation or consumption (Jury et al., 1991). Figure 3.2 shows a schematic diagram illustrating the condensation-evaporation phase change of vapor during its movement in soil. This condensation and evaporation mechanism of vapor transport is postulated to be partially responsible for the enhanced vapor diffusion in soil (compared to air diffusion rates) in the presence of its liquid (Philip & De Vries, 1957). Since

the presence of a temperature gradient in moist soil will cause the movement of heat as well as liquid water and water vapor, thermal conductivity of soil cannot be measured directly (De Vries, 1958). Instead, the effective thermal conductivity of porous media can be measured experimentally, which includes the effects of conduction and convection of latent heat (Jury et al., 1991). However, Hiraiwa and Kasubuchi (2000) estimated the fraction of thermal conductivity of soil attributed to the convection of latent heat by subtracting the effective thermal conductivity of soil near 0 °C from that at any higher temperature. This fraction depends on the temperature and volumetric water content of soil. Cass et al. (1984) measured effective thermal conductivity of soil as a function of temperature, water content, and pressure to separate the fraction due to the convection of latent heat. Wildenschild and Roberts (2001) used the same method for Topopah Spring Tuff.

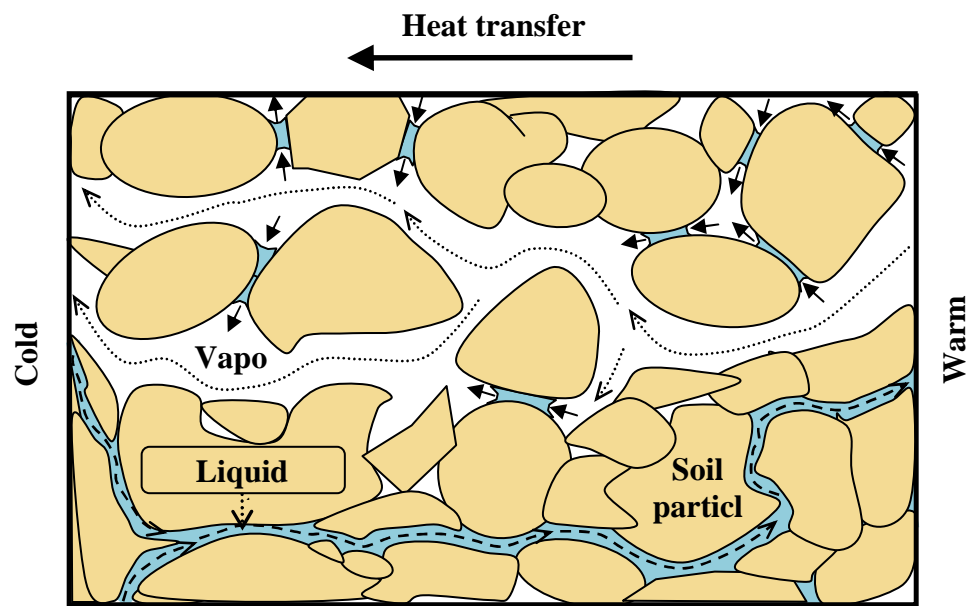


Figure 3.2 Schematic diagram illustrates the condensation-evaporation phase change of vapor during its transport in soil. The small arrows represent the direction of water vapor movement; as vapor enters the small pores it condensates and heat is released, and when it passes the small pores it evaporates and heat is consumed.

Based on the heat transfer phenomenon in a porous media, we assume that a fraction of the residual heat in the waste package is available for water evaporation and that it is equal to the fraction of thermal conductivity attributed to the convection of latent heat. Figure 3.3 shows the fraction of thermal conductivity attributed to the convection of latent heat ( $\beta$ ) versus volumetric water content ( $\theta$ ), for four kinds of soil estimated at a temperature of 32.5 °C from Hiraiwa and Kasubuchi (2000) and Cass et al. (1984).

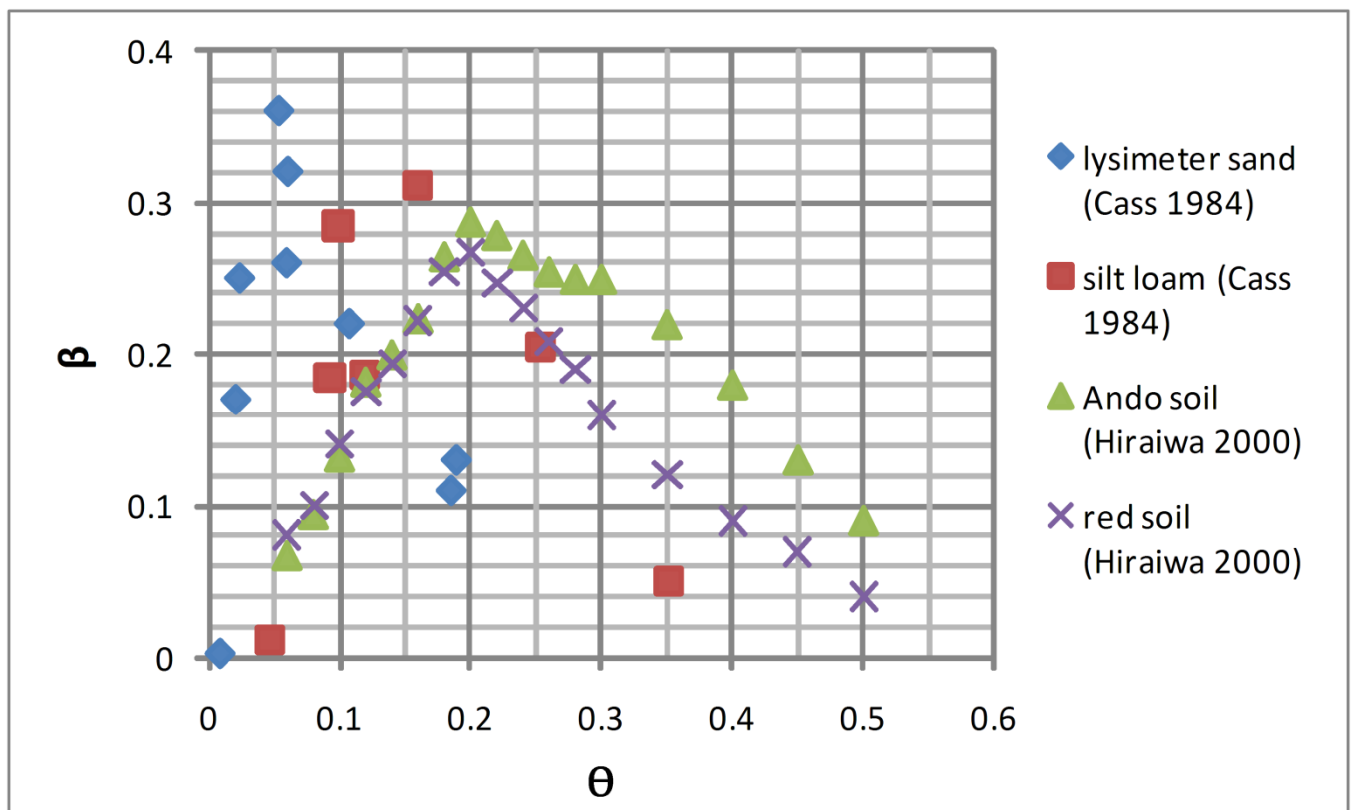


Figure 3.3 The fraction of thermal conductivity attributed to the convection of latent heat ( $\beta$ ) versus volumetric water content ( $\theta$ ) for different kinds of soil estimated at 32.5 °C from Hiraiwa and Kasubuchi (2000) and Cass et al. (1984).

### **3.3.2 North Percha Creek mine, NM**

Figure 3.4 shows a picture of the mouth of an old deserted silver mine on North Percha Creek, NM. The mouth of the mine is blocked with berms of mine tailings to reduce the seepage of contaminated water from the mine tunnels. Due to the significant hydraulic head gradient ( $\sim 20$  cm/100 cm) across the berms, we expect water to flow through the berms, carrying with it the dissolved minerals from the tailings forming the berms. Instead, as Figure 3.4 shows, the water flow through the berms resulted in a sequestration process for some of the minerals of the tailings on the top of the berms (the efflorescent crusts formed on the top of the tailings berms). Indeed, the low humidity of the air near the entrance of the drift causes continuous evaporation of the water at the upper surfaces of the wet tailings. This evaporation creates a capillary pressure gradient that pulls water, with its dissolved and suspended minerals, upward through the berms. As water evaporation continues, the concentration of the minerals increases and they start to precipitate forming the crusts on the top of berms. Interestingly, some tailings minerals are sequestered rather than released, due to the water evaporation driven by low air humidity.

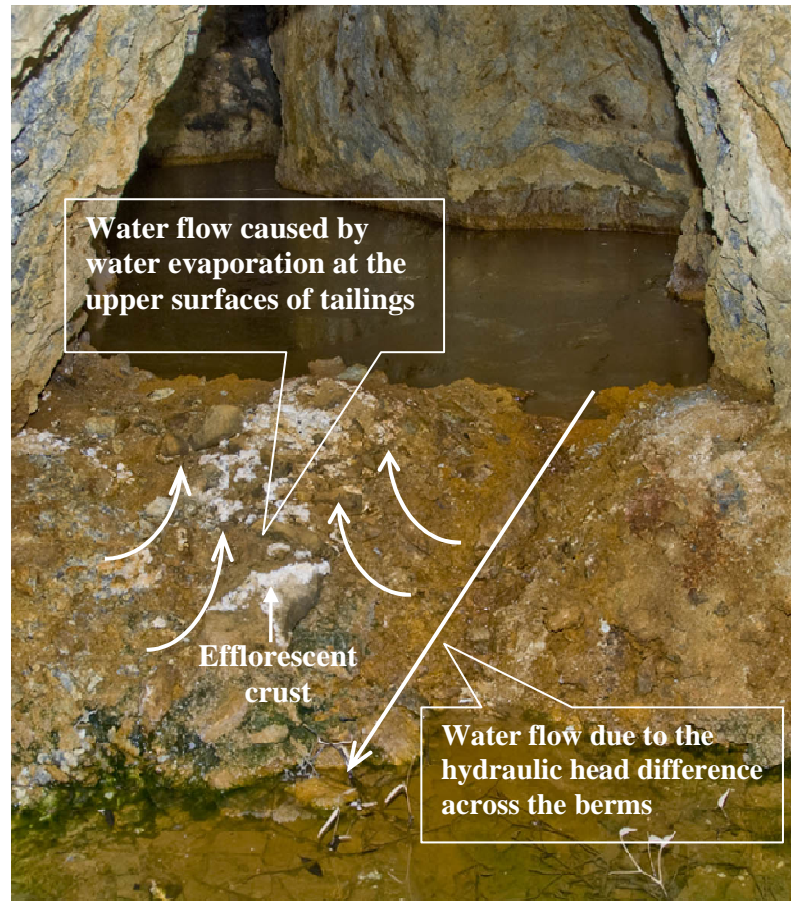


Figure 3.4 Mouth of North Percha Creek mine. Efflorescent crusts form on top of the wet tailings as water evaporates at the upper surfaces. Water evaporation creates a capillary pressure gradient that causes water to rise upward in the berms.

Based on the above phenomena, and given the fact that the temperature inside the waste package is not homogenous (usually center of the waste package due to heat loss, and areas where heavy metals concentrate, are hotter than others), we introduce a conceptual model to examine the effect of the nuclear waste residual heat release on radionuclide release from a partially failed waste container. The process is similar but not identical to the process at Percha Creek.



### **3.3.3 Radionuclide sequestration model**

This model addresses a potential serious failure case of the waste packages, in which several penetrations in the package allow water to flow through it from top to bottom (see Figure 3.5). In this case we anticipate water evaporation at sheltered (from direct seepage and condensate) hotter areas to create a capillary pressure gradient that causes at least part of the direct seepage to divert its direction toward the hotter regions, carrying with it dissolved and suspended radionuclides; in the same manner as water evaporation at the upper surfaces of the mine berms did to the contaminated water flowing through the berms. This means that there will be convective transport of the radionuclides toward the sheltered areas. As water evaporation continues, the concentration of radionuclides in the diverted seepage increases and radionuclides start to precipitate (as tailings minerals did); that is, radionuclides will be sequestered in the sheltered hot areas. We notice that this process, in the meantime, will create a gradient in the radionuclide concentration in the diverted seepage that initiates diffusive transport of radionuclides in the opposite direction (toward the direct seepage). Figure 3.5 shows a schematic diagram for the anticipated radionuclide transport processes inside a partially failed waste container.

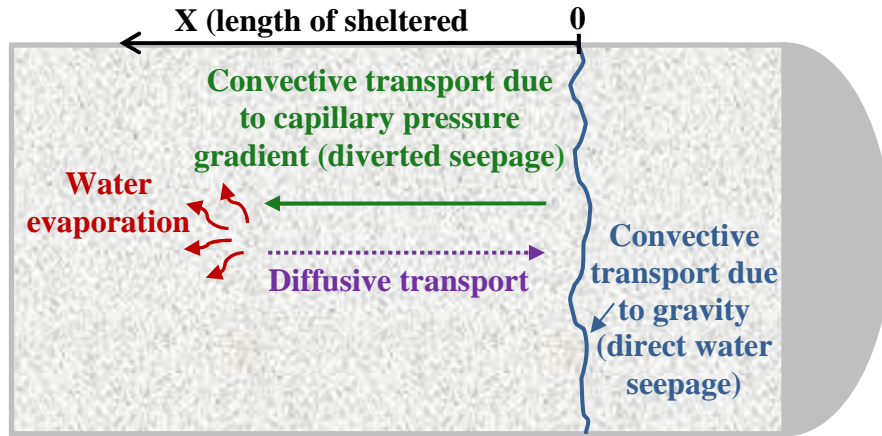


Figure 3.5 Schematic diagram illustrates the physical processes inside a partially failed container. Evaporation in the hotter regions drives water to flow toward sheltered regions (advective transport), while the increase in radionuclide concentration (due to evaporation) causes a diffusive transport in the opposite direction.

According to our model, radionuclides transported by convection (in the diverted seepage) will be sequestered in the sheltered areas, while radionuclides transported by diffusion could be released with the direct seepage. Under such conditions, radionuclides can only be released from the sheltered areas by diffusion when diffusive transport rate is greater than convective transport rate of the diverted seepage. On the contrary, if convection rate is greater than diffusion rate, radionuclides will be sequestered instead of released. To give a quantitative perception for radionuclide transport toward sheltered regions, simple one-dimensional calculations are made for this phenomenon, using the equations of water transport in soil, since the rubble inside the waste package is projected to resemble the physical properties of soil (porous media).

### 3.3.4 Mathematical treatment

In these calculations, the interior of the waste package is assumed to be homogenous and with no source or sink for radionuclides. At steady state, the first order advection-dispersion equation (in this equation, dispersion is treated mathematically like diffusion) for a chemical fully dissolved in water gives the critical case, which separates between the sequestration and release conditions in our model, convection rate equals diffusion rate (Walton, 2008):

$$VC = \theta\tau D \frac{dC}{dx} \quad (1)$$

where:

$V$  = specific discharge or Darcy velocity ( $\text{m/s} = \text{m}^3 / (\text{m}^2 \cdot \text{s})$ )

$C$  = concentration of chemical in water ( $\text{kg/m}^3$ ) or ( $\text{kmol/m}^3$ )

$\theta$  = volumetric water content

$\tau$  = tortuosity and constrictivity factor

$D$  = diffusion coefficient of the chemical in water ( $\text{m}^2/\text{s}$ ).

$(\theta\tau D)$  in Equation 1 represents the diffusivity of the chemical in the bulk porous medium. Conca and Wright (1992), and Conca et al (1993) measured the diffusion coefficients for several granular materials, over wide range of volumetric water content. Based on these measured data, the diffusion coefficient of a chemical in a bulk porous medium can be written, as a function of volumetric water content ( $\theta$ ), in this form of Archie's law (SNL, 2007 b):

$$D_{bulk} \equiv \theta\tau D = \theta^{1.863} D \quad (2)$$

To correct for the diffusion coefficient dependence on temperature, the following equation can be used (SNL, 2007 b):

$$D_T = D_{T_0} \frac{T}{T_0} 10^{\left[ \frac{1.3272(293.15-T_0)-0.001053(T_0-293.15)^2}{T_0-168.15} \right] - \left[ \frac{1.3272(293.15-T)-0.001053(T-293.15)^2}{T-168.15} \right]} \quad (3)$$

where:

$D_T$  is the diffusion coefficient ( $\text{m}^2/\text{s}$ ) at temperature  $T$  (K)

$D_{T_o}$  is the diffusion coefficient ( $\text{m}^2/\text{s}$ ) at temperature  $T_o$  (K)

In Equation 3,  $T_o = 298 \text{ K}$ .

The specific discharge or Darcy velocity can be estimated from evaporation flux, since it derives the convective transport:

$$V = \frac{E}{A} = \frac{Q' \beta v}{A \rho \gamma}$$
$$V = \frac{Q' \beta x}{\rho \gamma} \quad (4)$$

where:

$E$  = evaporation flux ( $\text{m}^3/\text{s}$ )

$A$  = area ( $\text{m}^2$ )

$Q'$  = rate of heat generation ( $\text{W}/\text{m}^3$ )

$\beta$  = fraction of heat goes to water evaporation

$v$  = volume ( $\text{m}^3$ )

$\rho$  = water density ( $\text{Kg}/\text{m}^3$ )

$\gamma$  = latent heat ( $\text{J}/\text{Kg}$ )

$x$  = distance from the direct seepage (m)

Substituting Equations 2 and 4 into Equation 1 and separating variables give:

$$\frac{dC}{C} = \frac{Q' \beta x dx}{\rho \gamma \theta^{1.863} D} = \alpha x dx \quad (5)$$

where  $\alpha = \frac{Q' \beta}{\rho \gamma \theta^{1.863} D}$

Integrating Equation 5, using the following boundary conditions:

$C = C_0$  at  $x = 0$  (the location of direct seepage, see Figure 3.5)

$C = C_x$  at a distance  $x$  from the direct seepage toward the sheltered areas

gives:

$$\frac{C_x}{C_0} = e^{\left(\frac{\alpha x^2}{2}\right)} = e^{\left(\frac{Q\beta x^2}{2\rho\gamma\theta^{1.863}D}\right)} \quad (6)$$

Equation 6 gives the concentration profile of an infinitely soluble radionuclide over distance in terms of the ratio of its concentration at any point ( $x$ ; down capillary convection stream) to the initial point ( $x = 0$ ).

Since these calculations are based on the critical condition: advection = diffusion, solving for ( $x$ ) in Equation 6 gives the minimum length of sheltered area (i.e., region with no dripping or other externally driven liquid advection) required to sequester radionuclides and prevent release:

$$x = \sqrt{\frac{2 \ln(C_x/C_0)}{\alpha}}$$

$$x = \sqrt{\frac{2\rho\gamma\theta^{1.863}D \ln(C_x/C_0)}{Q\beta}} \quad (7)$$

Equation 7 can also be obtained using Peclet number (Pe), which gives the ratio between the advective and diffusive components of solute transport (Mills et al., 1993):

$$Pe = \frac{vL}{D} \quad (8)$$

where:

$v$  = average linear velocity or pore velocity (m/s);  $v = \frac{V}{\theta}$  for partially saturated environment.

$L$  = characteristic length (m).

Small values of the Peclet number indicate that diffusion dominates, while large values indicate that advection dominates. A  $Pe = 1$  represents the critical condition in our model: advection equals diffusion. In this case, substituting for  $(v)$  by  $(V/\theta)$  and multiplying  $(D)$  by  $(\tau)$  to correct for tortuosity and constrictivity we have:

$$\tau D = \frac{V}{\theta} L \quad (9)$$

Substituting Equations 2 and 4 into Equation 9 and solving for  $(L \text{ or } x)$  we have:

$$L = \sqrt{\frac{\rho \gamma \theta^{1.863} D}{Q \beta}} \quad (10)$$

If we choose  $(C_x/C_o = 1.649)$ , Equation 7 turns out to be the same as Equation 10.

### 3.4 RESULTS AND DISCUSSION

The two main factors controlling radionuclide release in this model are the amount of heat released from the nuclear waste ( $Q'$ ) and the amount of moisture (volumetric water content,  $\theta$ ). The transport of a radionuclide toward sheltered areas can be represented by its concentration profile over distance, given by Equation 6. Figure 3.6 shows the concentration profile for plutonium (Pu), which has a free water diffusion coefficient equal to  $1.3 \times 10^{-9}$  (SNL, 2007 b), in a 21 PWR waste package. The concentration profile is calculated for three different times (different values of  $Q'$ ) and two different values of volumetric water content for each time, using the parameters shown in Table 3.1. The fraction of heat that goes to water evaporation ( $\beta$ ) is estimated using Hiraiwa and Kasubuchi (2000) because it covers a wide range of temperatures and volumetric water contents and soil is a reasonable analog for the interior of the waste package; approximate values are used for the time one million years. We have

performed sensitivity calculations with other values for  $\beta$  and find that although the specific predictions vary, the resultant conclusions do not.

Table 3.1 Parametric values used to calculate the concentration profiles in Figure 3.6.

Time after closure (yr)	Heat release ( $\text{W/m}^3$ )	Anticipated (T) for waste package ( $^{\circ}\text{C}$ ) <sup>a</sup>	Temperature correction for $D_{\text{bulk}}$ Eq. (3)	Fraction of heat ( $\beta$ )	
				$\theta = 0.1$	$\theta = 0.2$
10,000 yr	13	55	1.94	0.25	0.44
100,000 yr	1.04	25	1	0.1	0.21
1,000,000 yr	0.369	20	0.87	0.08	0.15

<sup>a</sup> (SNL, 2008 b)

Figure 3.6 shows that the concentration of radionuclides increases as we move toward the heat source in the sheltered areas, indicating mass transport for the radionuclides toward the sheltered areas (a sequestration process). Figure 3.6 indicates that the concentration ratios are greater in earlier times, since the heat release that causes the advective transport toward sheltered areas is greater. Since we assume an infinite solubility for the radionuclides, the concentration ratios reach very large values, especially for the time 10,000 years, as we move deeper in the sheltered areas. This does not happen in reality since every material has a limited solubility and starts to precipitate after a critical concentration. In addition, concentration ratios are greater for the lower values of volumetric water contents. These results are reasonable, since radionuclides are released by water transport (diffusive transport).

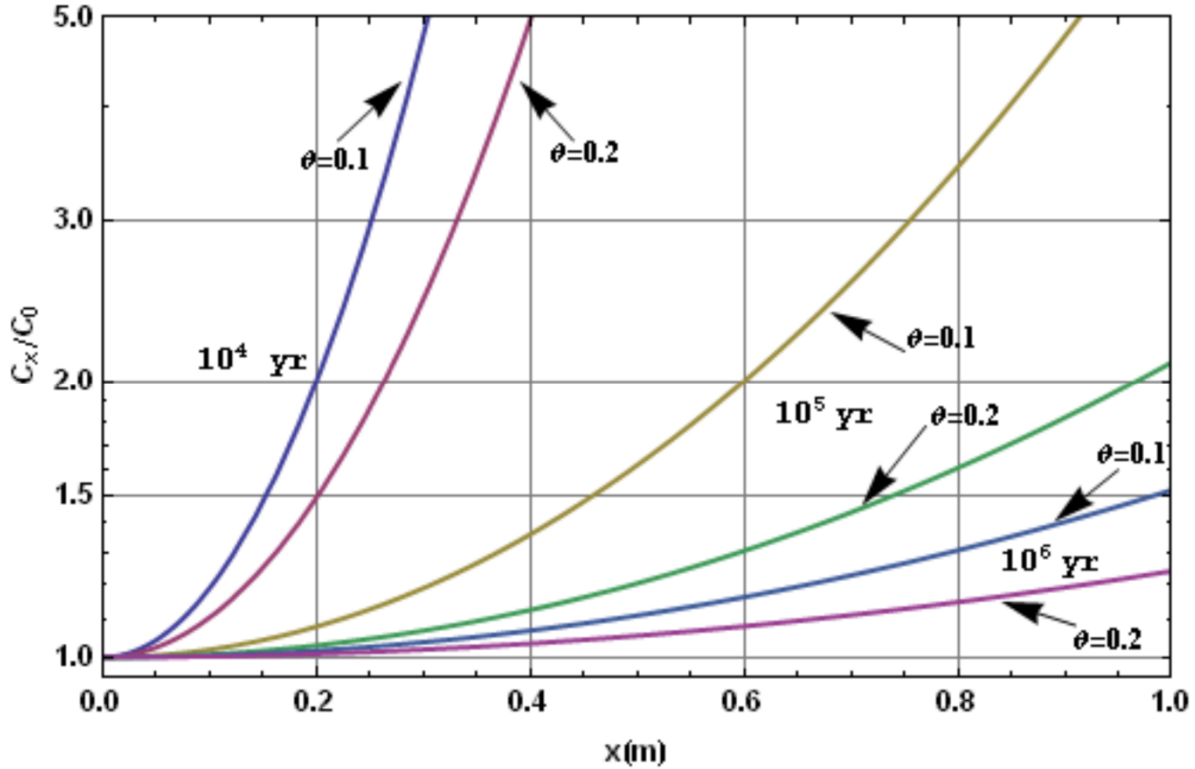


Figure 3.6 Ratio of radionuclide concentration ( $C_x / C_0$ ) vs. distance, calculated at three different times with two volumetric water content values.

Equation 10 (or Equation 7 with the value  $C_x / C_0 = 1.649$ ) is used to estimate the minimum length of sheltered areas in the waste package needed to prevent radionuclide release over time. Results are shown in Figure 3.7 for a 21 PWR waste package with two different volumetric water content values.

Figure 3.7 indicates that the minimum length of sheltered areas increases gradually over time (as the heat release that causes radionuclide sequestration decreases), but with significant protection extending out to one million years. In these calculations, the temperature corrections for the diffusion coefficient are not included, and an interpolation for the values of  $\beta$  over time is used. As it is expected, the minimum length of sheltered areas is smaller for the volumetric



water content of value 0.1 since the diffusion process, which leads to radionuclide release in our model, will be less.

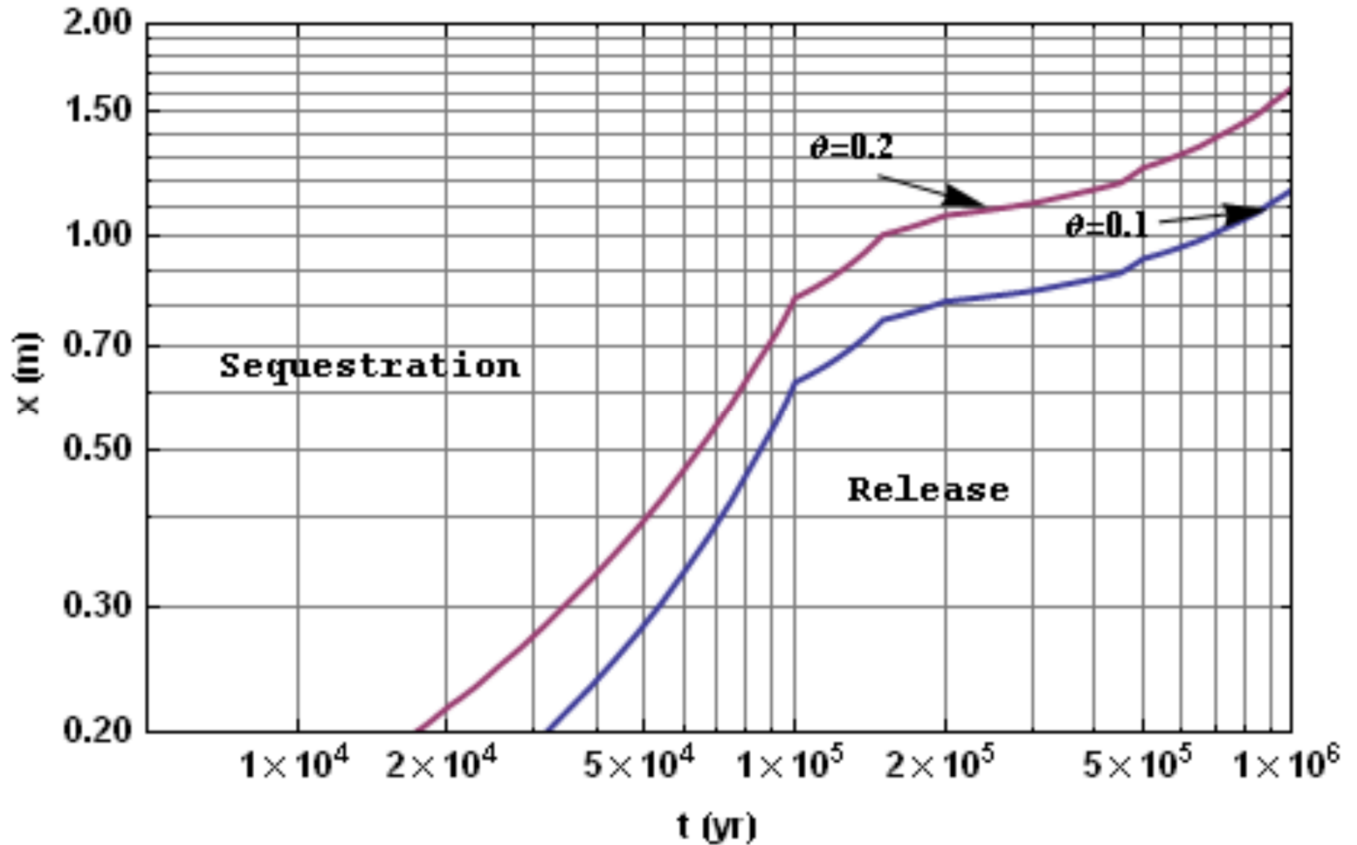


Figure 3.7 Minimum size of sheltered (no drip) area over time, required to protect the waste from leaching, for two values of volumetric water content. Sheltered areas larger than this size do not support radionuclide release.

### 3.5 CONCERNS

It is worthwhile to mention that accumulation of soluble salts may affect or defeat the flow system (advection-diffusion) generated in the sheltered areas. In addition, in our analysis we assumed that the interior of the waste package is homogenous, while it is expected to be heterogeneous and that will affect the transport properties of the system (capillary breaks may exist). However, in our calculations we used the average heat release, whereas the localized heat

release will be greater in the areas where heavy metals concentrate. This will enhance the flow system (since it is generated due to water evaporation), especially if the heavy metals are existed in the sheltered areas.

### **3.6 CONCLUSIONS**

Heat release characteristics of the nuclear waste contribute to the safety of geological nuclear waste repositories constructed in the unsaturated zone, since heat-generating waste stored in a partially saturated environment tends to sequester radionuclides in the sheltered areas and prevent their release. The radionuclide sequestration model showed that the sequestration process is more effective at higher rates of heat generation and lower moisture conditions. In addition, the minimum size of sheltered areas needed to prevent radionuclide release increases over time. On the other hand, the size of sheltered areas decreases over time due to general corrosion. A convolution of these two processes suggests that radionuclide release is likely to be gradual and long delayed.

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### **REFERENCES**

American Nuclear Society, ANS. More countries looking to produce nuclear power. Nuclear News . p. 41; 2007, February.

- Cass, A.; Campbell, G. S.; Jones, T. L. Enhancement of Thermal Water Vapor Diffusion in Soil. *Soil Sci. Soc. Am. J.* 48: 25-32; 1984.
- Conca, J. L.; Wright, J. Diffusion and Flow in Gravel, Soil, and Whole Rock. *Applied Hydrogeology Journal*. 1(1): 5-24; 1992.
- Conca, J. L.; Apted, M.; Arthur, R. Aqueous Diffusion in Repository and Backfill Environments. In: Interrante, C. G.; Pabalan, R. T. eds. *Scientific Basis for Nuclear Waste Management XVI* (pp. 395-402). Pittsburgh, Pennsylvania: Materials Research Society. TIC: 208880; 1993.
- De Vries, D. A. Simultaneous Transfer of Heat and Moisture in Porous Media. *Trans. Am. Geophys. Union*. 39: 909-916; 1958.
- DOE (U.S. Department of Energy): Office of Civilian Radioactive Waste Management. *Yucca Mountain Science and Engineering Report, DOE/RW-0539, chapter 1*. Las Vegas, Nevada; 2001, May.
- Feiveson, H. A. Faux Renaissance: Global Warming, Radioactive Waste Disposal, and the Nuclear Future. *Arms Control Today*. 37:13-17; 2007
- Hiraiwa, Y.; Kasubuchi, T. Temperature Dependence of Thermal Conductivity of Soil Over a Wide Range of Temperature (5-75°C). *European Journal of Soil Science*. 51: 211-218; 2000.
- International Atomic Energy Agency, IAEA. (2003). *THE LONG TERM STORAGE OF RADIOACTIVE WASTE: SAFETY AND SUSTAINABILITY*. Vienna: IAEA.
- Jury, W. A.; Gardner, W. R.; Gardner, W. H. *Soil Physics* (5th ed.). New York: John Wiley & Sons, Inc.; 1991

Lovins, A. B. Energy End-Use Efficiency. Inter Academy Council, Amsterdam, as part of  
“Transitions to Sustainable Energy Systems” study; 2005, September.

[www.rmi.org/images/PDFs/Energy/E05-16\\_EnergyEndUseEff.pdf](http://www.rmi.org/images/PDFs/Energy/E05-16_EnergyEndUseEff.pdf).

Mills, I.; Cvitas, T.; Homan, K.; Kuchitsu, K. Quantities, Units and Symbols in Physical  
Chemistry (2nd ed.). Oxford: Blackwell; 1993.

Nuttall, W. J. Nuclear Renaissance: Technologies and Policies for the Future of Nuclear Power.  
New York: Taylor & Francis Group; 2005.

Philip, J. R.; De Vries, D. A. Moisture Movement in Porous Materials under Temperature  
Gradients. Trans. Am. Geophys. Union. 38: 222-232; 1957.

SNL (Sandia National Laboratories). EBS Radionuclide Transport Abstraction, ANL-WIS-PA-  
000001 REV 03. Las Vegas, Nevada: Sandia National Laboratories. ACC:  
DOC.20071004.0001. section 6.3.4; 2007 b.

SNL (Sandia National Laboratories). Multiscale Thermohydrologic Model. ANL-EBS-MD-  
000049 REV 03 ADD 02. Las Vegas, Nevada: Sandia National Laboratories. ACC:  
DOC.20080201.0003, page 6-5; 2008 a.

SNL(Sandia National Laboratories). Multiscale Thermohydrologic Model. ANL-EBS-MD-  
000049 REV 03ADD 01. Las Vegas, Nevada: Sandia National Laboratories. ACC:  
DOC.20070831.0003 chapter 6; 2007a.

SNL (Sandia National Laboratories). Total System Performance Assessment Model/Analysis for  
the License Application, MDL-WIS-PA-000005 REV00. Las Vegas, Nevada: Sandia  
National Laboratories, DOC.20080204.0003, section 6.3.2; 2008 b.

Squassoni, S. Risks and Realities: The New Nuclear Energy Revival. *Arms Control Today* ,  
37(4): 6-12; 2007.

Vujic, J. Nuclear Energy Renaissance in the U.S. ,seminar presented at Power Systems  
Engineering Research Center Tele-Seminar; 2007, April.  
[www.pserc.org/ecow/get/generalinf/presentati/psercsemin1/pserc2007s/vujic\\_pserc\\_tele-](http://www.pserc.org/ecow/get/generalinf/presentati/psercsemin1/pserc2007s/vujic_pserc_tele-seminar_april_2007.pdf)  
[seminar\\_april\\_2007.pdf](http://www.pserc.org/ecow/get/generalinf/presentati/psercsemin1/pserc2007s/vujic_pserc_tele-seminar_april_2007.pdf). .

Walton, J. C. Fate and Transport of Contaminants in the Environment (1st ed.). Glen Allen,  
USA: College Publishing; 2008.

Wildenschild, D.; Roberts, J. Experimental tests of Enhancements of Vapor Diffusion in  
Topopah Spring tuff. *Journal of Porous Media.*, 4(1): 1-13; 2001.

## **Chapter 4**

### **Oxidation/Reduction Status of Water Pooled in a Penetrated Nuclear Waste Container**

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#### **4. Oxidation/Reduction Status of Water Pooled in a Penetrated Nuclear Waste Container -10265**

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##### **ABSTRACT**

Nuclear power use is expected to expand in the future and result in hundreds of thousands of metric tons of spent nuclear fuel (SNF). One of the main concerns of nuclear energy use is SNF disposal. Storage in geological repositories is a reasonable solution for the accumulation of SNF. One of the key factors that determine the performance of the proposed geological repository at Yucca Mountain (YM), NV is the release of radionuclides from the engineered barrier system (EBS) by water transport. Over time, EBS, including nuclear waste containers, is expected to fail gradually due to general and localized corrosion. Physical and chemical disturbances in the environment of the repository will lead to different corrosion rates at different locations of the waste packages. Considering the inherent uncertainty of the failure sequence of a waste package, two main failure scenarios are expected: flow through model (penetrations are on the top and bottom of the waste package causing water to flow through it) and bathtub model (penetrations are on the top with the waste package filling with water). In this paper, we consider a bathtub category failed waste container and shed some light on chemical and physical processes that take place in the pooled water and their effects on radionuclide release. We

consider two possibilities: temperature stratification of the pooled water versus mixing. Our calculations show that there will be temperature stratification of the pooled water in the lower half of the waste package, and mixing in the upper half. The effect of these situations on oxygen availability and consequently spent fuel alteration and waste container components corrosion is also considered.

#### **4.1 INTRODUCTION**

YM repository design depends on multiple natural and engineered barriers to isolate nuclear waste and keep it dry as long as possible. The U. S. Department of Energy (DOE) made comprehensive and detailed studies to assess the performance of the proposed repository at YM. In the long run, as the EBS is expected to fail, DOE (to be conservative) screened out the bathtub model as an alternative to the flow-through conceptual model. Their analysis showed that the flow-through model increases radionuclide releases relative to the bathtub model [1]. However, the bathtub model in which water pools inside the waste package is a possible scenario for the waste package failure. It is worthwhile to predict the conditions inside the pooled water and estimate their effects on corrosion of the waste form and other waste package components.

#### **4.2 CONDITIONS INSIDE THE POOLED WATER OF THE BATHTUB MODEL**

A main factor in the corrosion process is oxygen availability. The limited solubility of oxygen in water and the existence of temperature stratification may restrict oxygen accessibility inside the pooled water. Temperature variance inside the pooled water is expected since the nuclear waste will continue heat release beyond one million years [2]. Temperature variance of the pooled water inside a failed waste package was calculated by discretization (finite difference) method using Excel. In these calculations we assumed: the porosity of the corroded materials is



50%, the waste package is half filled with water, and heat transfer inside the waste package is by conduction only. The thermal conductivity of soil is used for the corroded materials [3]. Figure 4.1 shows the temperature variance inside a pressurized water reactor spent fuel (21 PWR) 20,000 years after permanent closure of the repository.

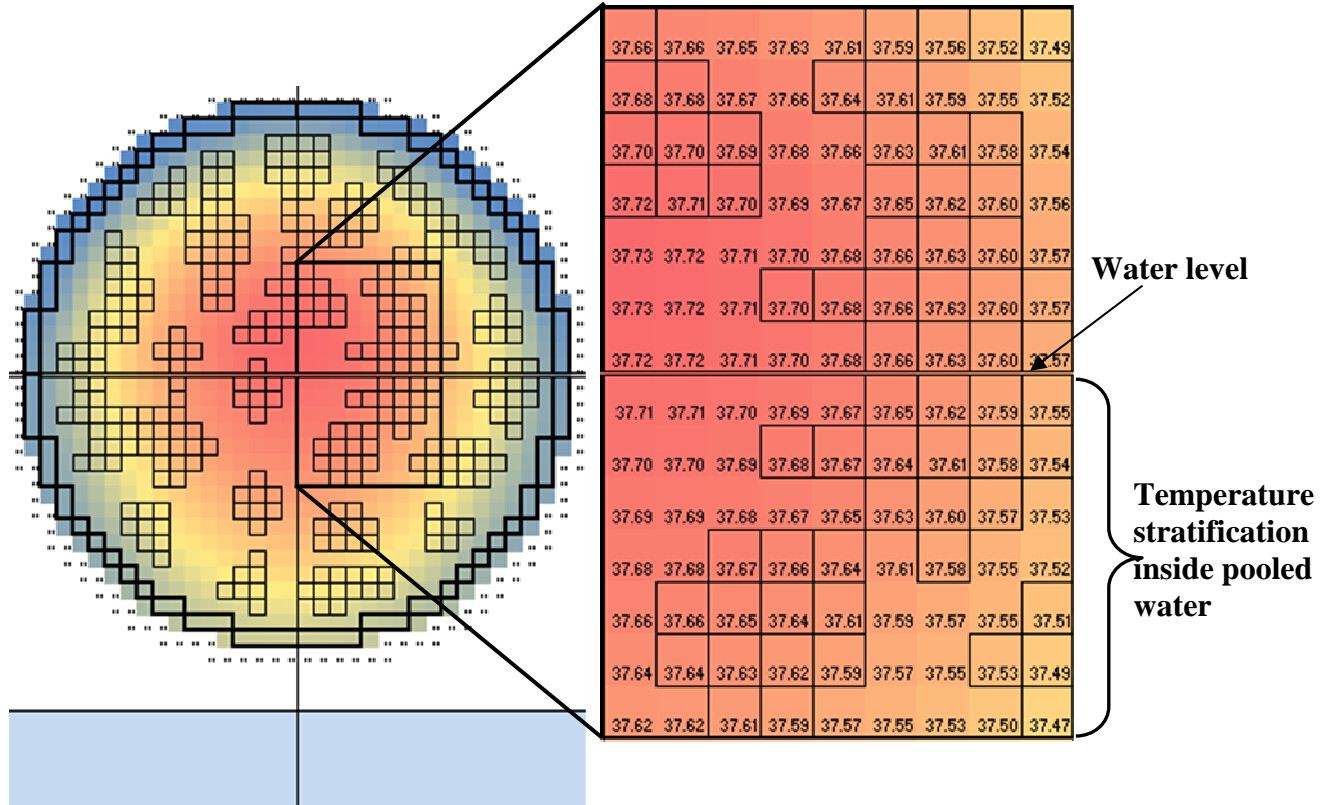


Figure 4.1 Excel calculated temperature variance inside a 21 PWR waste package, half filled with water, 20,000 years after permanent closure of the repository, calculated using the finite difference method. Red color represents the hottest region.

Figure 4.1 indicates that there is temperature stratification within the pooled water in the lower half of the waste package. This means that depleted oxygen (by corrosion) can be replenished only by diffusion. Similar calculations were made for a waste package fully filled with water; the calculations showed the same pattern for the lower part of the waste package (temperature stratification), while for the upper half of the waste package mixing process is

expected in the pooled water since the cold water is above the hot water. This process will make the oxygen concentration approximately homogeneous in this region.

### **4.3 CORROSION PROCESS WITHIN THE POOLED WATER**

Water as an additional chemical compound could affect the oxidation process and produce hydrated oxidation products. As a reaction medium, water allows oxidation to proceed by dissolution-precipitation and may enhance diffusion along grain boundaries. In addition, in the case of spent nuclear fuel, water is a potential source of radiolytic oxidation products, such as hydrogen peroxide. Radiolytic products may alter the kinetics and products of oxidation reaction [4]. However in this paper, we consider the effect of water as a reaction medium on oxygen availability. Oxygen plays a principal role in redox reactions, the main cause of materials corrosion. Consequently, oxygen has effects on radionuclide release. In addition, some radionuclides, such as uranium and plutonium, are less soluble under reducing conditions.

#### **4.3.1 Oxygen Availability in the Pooled Water**

Figure 4.2 is a schematic diagram illustrating the steady state balance of oxygen in the pooled water of a partially failed waste container (bathtub category) half filled with water.

Oxygen diffuses from air into water (input), such that the concentration of oxygen in water according to Henry's law equals:

$$C_w = k_H P \quad (\text{Eq. 1})$$

where  $C_w$  is the concentration of oxygen in water (mol/L),  $P$  is vapor pressure of oxygen in air (atm), and  $k_H$  is Henry's law constant (L. atm /mol) [5]. This makes the simplifying assumption that the air/water mass transfer coefficient is high relative to diffusion rates.

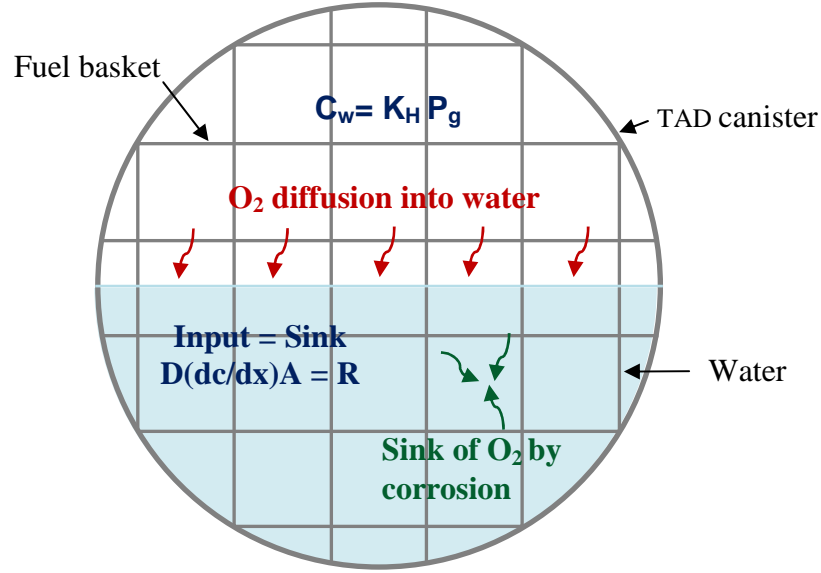


Figure 4.2 Schematic diagram illustrating steady state oxygen balance in water pooled inside a partially failed waste container half filled with water.

Accordingly, oxygen concentration in the water will be:

$$C_w = 1.07 \times 10^{-3} \times 0.2 = 2.14 \times 10^{-4} \text{ mol/L} = 6.848 \times 10^{-3} \frac{\text{kg}}{\text{m}^3}$$

where  $1.07 \times 10^{-3}$  mol/L-atm is the Henry's law constant of oxygen in water at a temperature of 37°C [6,7]. The transport of oxygen into the stagnant water is by diffusion:

$$\text{Input} \left( \frac{\text{kg}}{\text{s}} \right) = D \frac{dc}{dx} A \quad (\text{Eq. 2})$$

where  $D$  is diffusion coefficient of oxygen in water ( $\text{m}^2/\text{s}$ ),  $dc/dx$  is oxygen concentration gradient in water ( $\text{kg}/\text{m}^4$ ), and  $A$  is the surface area of water in contact with air ( $\text{m}^2$ ). Assuming that the concentration of oxygen 1 cm beneath the water surface equals zero, the input of oxygen into the water equals:

$$\text{Input} = 3.2 \times 10^{-9} \frac{6.848 \times 10^{-3}}{0.01} \times (8.7 \times 0.2) = 3.8 \times 10^{-9} \text{ kg/s} \quad (\text{Eq. 3})$$

Here  $8.7 \text{ m}^2$  is the surface area of water if the transportation, aging and disposal (TAD) canister contains only water and is half filled with it; the factor 0.2 is to account for the existence of spent

fuel rods and other waste package components, which occupy most of the surface. The diffusion coefficient of oxygen in water is at a temperature of 37°C [8].

#### 4.3.2 Oxygen Consumption Due to Stainless Steel Corrosion Only

Oxygen is consumed from water by corrosion of spent fuel and other waste package components. In this section, we consider the corrosion of Stainless Steel (SS) components only and their rate of oxygen consumption. SS is the most corrosion resistant material of the TAD canister components (Alloy 22 is the outer corrosion barrier of the waste package that contains the TAD canister). The composition of the TAD canister and its internal components is shown in Table 4.1 [9]; spent nuclear fuel is not included.

Table 4.1 Materials that Compose the TAD Canister and its Internal Components, along with the Surface Area of the Components Made from Each Material

Components of TAD canister		
Material	Surface area (m <sup>2</sup> )	
	Internal components	TAD canister shell
SS 316 L	269.8	31.5
SS 304B4 (borated SS)	137.5	-
Aluminum	57.9	-

Borated SS is treated as SS 316 L. To estimate the rate of oxygen consumption by SS 316 L corrosion, we assumed the alloy shown in Table 4.2. The alloy composition includes only the primary elements (weight %  $\geq 2$ ) as set by DOE. Borated SS does not include molybdenum in its alloy composition and 1.1 to 1.2 percent of its weight is boron, for our calculation purposes, this will not significantly affect the number of oxygen moles required for corrosion of one kg of SS [1].

Table 4.2 Primary elements that compose SS 316 L and number of O<sub>2</sub> moles required for corrosion of one mole of each element.

Element	Gram/Mole	Density (kg/m <sup>3</sup> )	Assumed percentage in SS 316 (7934 kg/m <sup>3</sup> )	No. of Moles in 1 kg of SS 316	No. of O <sub>2</sub> Moles required for corrosion of 1 Mole of element	
Fe	55.85	7874	68 %	12.2	$4\text{Fe} + 3\text{O}_2 \rightarrow 2\text{Fe}_2\text{O}_3$	<b>3/4</b>
Cr	52	7140	16 %	3.08	$2\text{Cr} + 3/2 \text{O}_2 \rightarrow \text{Cr}_2\text{O}_3$	<b>3/4</b>
Ni	58.69	8908	11 %	1.87	$\text{Ni} + 1/2 \text{O}_2 \rightarrow \text{NiO}$	<b>1/2</b>
Mo	95.94	10280	3 %	0.31	$\text{Mo} + \text{O}_2 \rightarrow \text{MoO}_2$	<b>1</b>
Mn	54.94	7470	2 %	0.36	$\text{Mn} + \text{O}_2 \rightarrow \text{MnO}_2$	<b>1</b>

Table 4.3 shows the parameters we used to calculate oxygen consumption due to the corrosion of SS 316 L. The rate of corrosion of SS 316 L (0.267  $\mu\text{m}/\text{yr}$ ) is converted to the units of (kg/m<sup>2</sup>/yr) [10].

Table 4.3 Parameters used to calculate rate of oxygen consumption by SS 316 L corrosion.

No. of O <sub>2</sub> Moles required for corrosion of 1 kg of SS 316	13.1
Degradation rate for SS 316 (kg/m <sup>2</sup> /yr)	$2.12 \times 10^{-3}$
Surface area of SS 316 inside the pooled water (m <sup>2</sup> )	219.4

Number of oxygen moles required for corrosion of one kg of SS is calculated from Table 4.2. The surface area inside the pooled water includes the borated SS surface area. The TAD canister shell surface area represents the internal surface area of a simple cylinder (the surface area of other shell components is not included). All areas are divided by two, since the canister is half filled with water.

Using the information in Table 4.3, the rate of oxygen consumption due to SS corrosion equals (R):

$$R = \frac{2.12 \times 10^{-3} \times 13.1 \times 32 \times 10^{-3} \times 219.4}{365 \times 24 \times 60 \times 60} = 6.2 \times 10^{-9} \text{ kg/s} \quad (\text{Eq. 4})$$

Here  $32 \times 10^{-3}$  is number of kilograms of oxygen per mole.  $R$  represents a sink to the oxygen inside water.

#### 4.4 DISCUSSION

Comparing Eq. 3 with Eq. 4 we find that oxygen input (by diffusion) into water is less than oxygen consumption by corrosion. It is worthwhile to note that our calculations did not include the corrosion of the other components inside the TAD canister including the spent nuclear fuel, which has higher rate of corrosion than SS. In addition, in these calculations the oxygen input into water is overestimated, since we assumed that oxygen concentration becomes zero after one cm beneath the water surface. However, SS corrosion may continue under anaerobic conditions.

At steady state, the continuity equation of oxygen in the pooled water (our control volume) is given by [10]:

Input = Sink

$$D \frac{dc}{dx} A = R \quad (\text{Eq. 5})$$

In this case the corrosion rate of materials inside the TAD canister may slow to reach the mass balance of oxygen in the pooled water.

#### 4.5 CONCLUSIONS

The rate of oxygen input into the pooled water is less than the rate of oxygen consumption caused by the corrosion of different components of the waste package in air. As a result, the rate of oxidation of spent fuel and other waste package components inside the pooled water is likely to slow until it reaches equilibrium with oxygen input rate. In this case, the

estimated life for all waste package components will become longer and radionuclide release will be delayed. We are currently evaluating the likely impact of pooled water upon spent fuel oxidation.

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#### **REFERENCES**

- [1] Sandia National Laboratories, “EBS Radionuclide Transport Abstraction”, ANL-WIS-PA-000001 REV 03. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20071004.0001, (2007)
- [2] L.K. Hamdan, J.C. Walton and A Woocay, “Effect of Physical Processes on Radionuclide Release from Partially Failed Containers”, Abstract 9260. Talk presented at Waste Management (WM) 2009 Symposia, Phoenix, AZ, March 4, (2009).
- [3] W. A. Jury, W. R. Gardner and Walter H. Gardner, “Soil Physics”, Fifth Edition, New York: John Wiley & Sons, Inc. (1991).
- [4] R. J. McEachern, P. Taylor, “A review of the oxidation of uranium dioxide at temperatures below 400 °C”, Journal of Nuclear Materials 254: 87-121, (1998).
- [5] R. Sander, “Compilation of Henry's Law Constants for Inorganic and Organic Species of Potential Importance in Environmental Chemistry”, Version 3, (1999).  
<http://www.henrys-law.org>

- [6] E. Wilhelm, R. Battino, and R. J. Wilcock, “Low-pressure solubility of gases in liquid water”. Chem. Rev., 77, 219–262, (1977).
- [7] R. Lide, and H. P. R. Frederikse, editors, “CRC Handbook of Chemistry and Physics”, 76th Edition: CRC Press, Inc., Boca Raton, FL, (1995).
- [8] Landolt-Bornstein: Group IV Physical Chemistry, “Gases in Gases, Liquids and their Mixtures”, volume 15A: Springer Berlin Heidelberg, (2007).
- [9] Sandia National Laboratories (SNL), “Total System Performance Assessment Data Input Package for Requirements Analysis for TAD Canister and Related Waste Package Overpack Physical Attributes Basis for Performance Assessment”, TDR-TDIP-ES-000006 REV 00. Las Vegas, Nevada: SNL. ACC: DOC.20070918.0005, (2007).
- [10] J. C. Walton, “Fate and Transport of Contaminants in the Environment”, First edition, College Publishing: Glen Allen, USA (2008).



## **Chapter 5**

### **Effect of Water Pooled in a Failed Nuclear Waste Package on Radionuclide Release**

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## **5. Effect of Water Pooled in a Failed Nuclear Waste Package on Radionuclide Release**

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### **ABSTRACT**

In the long term, nuclear waste packages stored in an unsaturated zone geological repository such as the proposed repository at Yucca Mountain, are expected to fail gradually due to general and localized corrosion and water will have access to the nuclear waste. In this case radionuclides will make their way to the accessible environment by groundwater transport. One possible scenario for the waste packages failure is the bathtub model, where the waste container is penetrated from the top only causing water to pool inside it. In this paper we consider a partially failed waste container from the bathtub category. We shed some light on chemical and physical processes that take place in the pooled water and their effects on radionuclide release. Finite difference method was used to estimate temperature variance inside a partially failed

waste package (the bathtub category). Our calculations showed that, temperature stratification inside the pooled water is expected in the lower half of the waste package, which would limit the oxygen availability required for corrosion. In addition, the rate of oxygen input into the pooled water by diffusion was estimated and found to be less than the rate of oxygen consumption by the corrosion of nuclear waste package components. The effect of oxygen deficiency inside the pooled water (reducing conditions) on the corrosion of spent nuclear fuel is investigated. Our analysis showed that the reducing conditions are likely to slow down the corrosion to very low rates causing a delay in radionuclide release.

**Keywords:** Geological repository; Bathtub model; Reducing conditions; Radionuclide release.

## 5.1 INTRODUCTION

As the nuclear energy use is expected to expand in the future, more concerns arise regarding the management of resultant nuclear waste. Safety (for people and the environment) and security (prevention of nuclear proliferation) are two key objectives of nuclear waste management. While nuclear waste material continues to accumulate in engineered storage facilities above ground or at very shallow depth, disposal in deep underground engineered facilities, i.e. geological disposal, is under investigation in most countries with significant amounts of nuclear waste (IAEA, 2003). Indeed, there is international consensus among the waste management experts that deep geological disposal is the best option currently available for high level and long lived radioactive waste (OECD, 1999). Worldwide, a number of rock types are considered for deep geological disposal, such as salt, clay, granite, and tuff. The Waste Isolation Pilot Plant (WIPP), constructed in bedded salt approximately 658 m below the ground surface near Carlsbad, New Mexico, in the United States, is currently the only repository in the world that receives nuclear waste (Hornberger, 2009). Other repositories are still under

discussion, with two countries have made formal Government decisions to go ahead with facilities for deep geological disposal (IAEA, 2003). WIPP is designed to store defense transuranic waste, while the proposed repository at Yucca Mountain (YM), Nevada is intended to store mainly highly radioactive spent nuclear fuel (SNF).

Natural and engineered barriers of the proposed repository at YM are anticipated to isolate radioactive waste from the human environment for very long periods of time. However, water will have access to the nuclear waste, due to localized and general corrosion in the long run. As a result, radionuclides will be transported by groundwater to the accessible environment. The physical and chemical disturbances in the environment of the waste packages will expose the waste packages to different rates of corrosion, leading to different times of penetration at different locations. Accordingly, two main scenarios are expected for the failure of waste packages: the flow-through model, in which penetrations on the top and bottom of the waste package will allow water to flow through it, and the bathtub model, where the penetrations are in the upper part of the waste package and water will pool inside the waste container. To be conservative, the U.S. Department of Energy (DOE) in their assessment to the performance of YM repository excluded the bathtub model. Their analysis showed that the bathtub model introduces a delay in radionuclide release to the engineered barrier system (EBS) but does not change the dose rate, therefore the flow-through model is bounding relative to the bathtub model (SNL, 2007 a). However, since the bathtub model is a possible scenario for the failure of waste packages, it is worthwhile to shed some light on the effect of pooled water on the corrosion process of waste package components. Given that many radionuclides of the waste inventory are redox sensitive (their solubility and mobility depend on their oxidation state), we will check oxygen availability inside the pooled water and its effect on radionuclide release, and whether a

failure in a bathtub model container from the bottom will cause a peak in the dose rate. To do that we need to check the physical and chemical conditions inside the pooled water.

## **5.2 CONDITIONS INSIDE THE POOLED WATER OF BATHTUB MODEL**

A main difference between the bathtub model and flow-through model is that in the bathtub model part of the nuclear waste will be submerged in the pooled water. As a chemical compound, water could affect the oxidation process and produce hydrated oxidation products, such as  $\text{UO}_3\text{-H}_2\text{O}$  compounds. As a reaction medium, water allows oxidation to proceed by dissolution-precipitation mechanisms and may enhance oxygen diffusion along grain boundaries in sintered  $\text{UO}_2$  pellets. In the case of SNF water is a potential source of radiolytic oxidation products, which may affect the kinetics and the products of oxidation (McEachern and Taylor, 1998). In addition, the chemistry of water, such as the pH and carbonate concentration, affects the corrosion rate of nuclear waste ( $\text{UO}_2$  oxidative – dissolution rate) (BSC, 2004 a). Finally, water as a reaction medium limits oxygen availability. This effect is significant, since oxygen is a principal factor in redox reactions, the main cause of materials corrosion. As well, some radionuclides, such as uranium and plutonium are less soluble under reducing conditions (Szecsody et al., 1998), (Runde et al., 2002). Oxygen availability is influenced by oxygen solubility in water, as well as by physical conditions inside the pooled water, whether there is temperature stratification or mixing.

### **5.2.1 Temperature stratification versus mixing inside pooled water**

Our calculations considered pressurized water reactor (21 PWR) type, since it has the greatest number of waste packages to be stored in YM repository (SNL, 2008) and has the highest rate of energy release (Danko and Bahrami, 2004). To examine the physical status of the

pooled water in the bathtub model, we calculated temperature variance inside a failed waste package by discretization (finite difference method) using Excel. In our calculations we used a simple model, in which the interior of the waste package is divided into square cells. The calculations are two dimensional because the waste packages will be placed in the repository in a line-load configuration with a waste package-to- waste package spacing of approximately 10 cm (SNL, 2008), and the length of the waste package is 5.69 m while its diameter is 1.88 m (SNL, 2007 b); consequently we assumed temperature symmetry in the direction of the line-load. The corroded materials are modeled as soil with 50 percent porosity and given thermal conductivity ( $k$ ) that depends on volumetric water content ( $\theta$ ) (Jury et al., 1991), while the nuclear fuel is given the thermal conductivity of the internal cylinder of the waste package (SNL, 2007 c). The intact SNF is distributed randomly inside the waste package. Heat transfer inside the waste package is by conduction only. Heat loss from the waste package is by convection, with the background temperature for bottom cells set 0.1 °C lower than adjacent cells to account for the conduction loss through the emplacement pallet (made of Alloy22 and Stainless Steel (SS) 316); the radiation loss is neglected since our calculations are in a late period (20,000 years after closure) and the differences in temperature between waste package, drip shield and drift wall are small (less than 1 °C) (SNL, 2007 c). Table 5.1 shows some of the parametric values used in the temperature variance model.

Table 5.1 Parametric values used in the temperature variance model calculations.

Heat generation rate of the waste package ( $E$ )	Convection heat-transfer coefficient for air ( $h_c$ )	$\Delta X$ for square cells	$k$ for lumped shell
$7.57 \text{ W/m}^3$ <sub>a</sub>	$8.7 \text{ W/m}^2\text{-K}$ <sub>b</sub>	0.05 m	$1.1 \text{ W/m-K}$ <sub>c</sub>

<sub>a</sub> For 21 PWR package at time 20,000 years after closure.

<sub>b</sub> The value of convection heat coefficient of air is  $10 \text{ W/m}^2\text{-K}$  and is multiplied by 0.87 (5.9, waste package perimeter, divided by 6.8, perimeter of convection surface in the model) to correct for the larger length of perimeter due to sheet configuration.

<sub>c</sub> The lumped shell contains: Alloy 22 (2.5 cm thickness,  $k = 8.5 \text{ W/m-K}$  (Special Metals Corporation, 2006)), SS 316 (7.5 cm thickness,  $k = 13.7 \text{ W/m-K}$  (Karditsas & Baptiste, 1995)), the gap to accommodate corrosion products (2 cm thickness,  $k = 0.4 \text{ W/m-K}$  (SNL, 2007 b)). The values of  $k$  are estimated at  $37^\circ\text{C}$ .  $k$  effective of the lumped shell is 2.1 and is multiplied by 0.42 to correct for the real thickness (5cm represents 12cm) and by 1.24 (6.6, the length in the model, divided by 5.34, the perimeter of model circle) to correct for the larger length of perimeter due to sheet configuration

Figure 5.1 shows a schematic diagram for the model used to calculate temperature variance along with the thermal conductivities used for the materials inside the waste package.

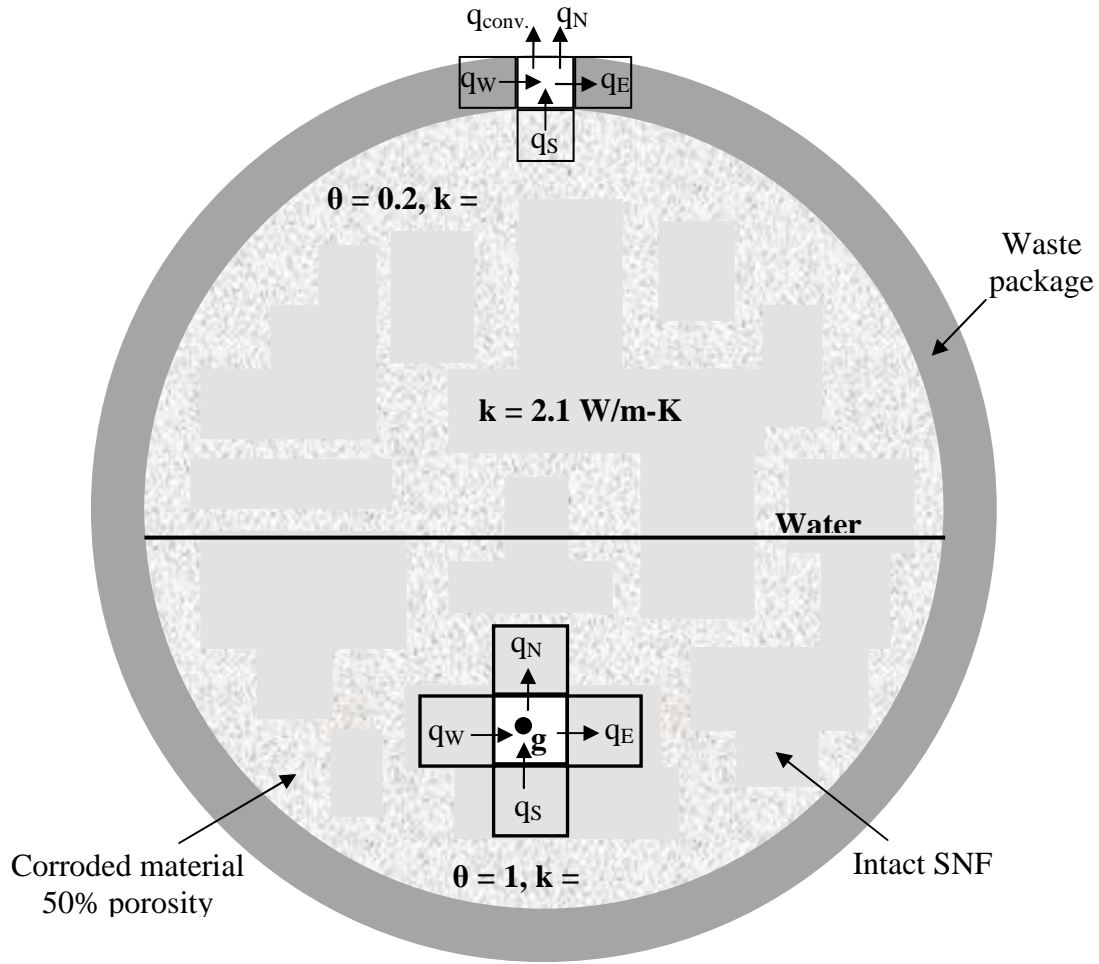


Figure 5.1 Schematic diagram illustrates the model used to calculate temperature variance inside a partially failed waste package.

Heat balance for the boundary cells is:

$$q_S + q_W = q_E + q_N + q_{conv.} \quad (1)$$

where  $q_S$  is rate of heat transfer by conduction from the southern cell (J/s), W stands for west, N for north, and E for east;  $q_{conv.}$  is rate of heat transfer by convection (J/s).

The rate of heat transfer by conduction is given by:

$$q = -kA \frac{\Delta T}{\Delta x} \quad (2)$$



where  $k$  is thermal conductivity of the medium (W/m-K),  $A$  is surface area through which the heat flows ( $m^2$ ),  $\Delta x$  is the distance of heat flow (m), and  $\Delta T$  is temperature difference between distance ends (K).

The rate of heat transfer by convection is given by:

$$q_{conv} = h_c A (T_{surface} - T_{\infty}) \quad (3)$$

where  $h_c$  is convection heat-transfer coefficient of the fluid (W/m<sup>2</sup>-K).

Substituting Equation 2 and Equation 3 into Equation 1 to solve for the temperature, we have the following equations:

For cells with two convection boundaries:

$$T = \frac{k_w T_w + k_N T_N + k_E T_E + k_S T_S + 2 * T_{\infty} h_c \Delta x}{k_w + k_N + k_E + k_S + 2 h_c \Delta x} \quad (4)$$

For cells with one convection boundary:

$$T = \frac{k_w T_w + k_N T_N + k_E T_E + k_S T_S + T_{\infty} h_c \Delta x}{k_w + k_N + k_E + k_S + h_c \Delta x} \quad (5)$$

For cells with no convection boundaries:

$$T = \frac{k_w T_w + k_N T_N + k_E T_E + k_S T_S}{k_w + k_N + k_E + k_S} \quad (6)$$

Here  $k_w$  is the effective thermal conductivity of the cell where temperature is being calculated and the neighbor cell to the west. The same analog applies for  $k_N$ ,  $k_E$ , and  $k_S$ .

Heat balance for the interior cells is:

$$q_w + q_s + E_g = q_E + q_N \quad (7)$$

$$E_g = EV \quad (8)$$

where  $E_g$  is rate of heat generation by nuclear waste (J/s),  $E$  heat generation per unit volume (J/s- $m^3$ ), and  $V$  is volume ( $m^3$ ).

Substituting Equation 2, Equation 3, and Equation 8 into Equation 7 to solve for the temperature, we have:

$$T = \frac{T_w k_w + T_s k_s + T_E k_E + T_N k_N + E \Delta x^2}{k_w + k_s + k_N + k_E} \quad (9)$$

Equation 4, Equation 5, Equation 6, and Equation 9 are per unit length in the direction of the line-load.

A finite difference method, with successive relaxation, was applied to different levels of water inside the failed waste package: half filled, three quarters filled and fully filled with water. The results showed the same pattern of temperature variance for all levels of water. Figure 5.2 shows the temperature variance for a waste package fully filled with water.

Figure 5.2 indicates that a mixing process is expected in the upper half of the waste package since cold water is above hot water. Consequently oxygen concentration in this region will be approximately homogenous and controlled by the rate of oxygen input at the water-air interface and its rate of consumption by corrosion within pooled water. On the other hand, there will be temperature stratification in the lower half of the pooled water, which means that oxygen depleted by corrosion will be replenished only by diffusion from the upper layers. Figure 5.2 shows also horizontal temperature variance between the cells, which leads to horizontal mixing. To account for horizontal mixing and examine its effect on temperature stratification, temperature variance was recalculated after the thermal conductivities in the horizontal direction were multiplied by the factor 10 and then by the factor 100. The results continued to show temperature stratification in the lower half of the waste package.

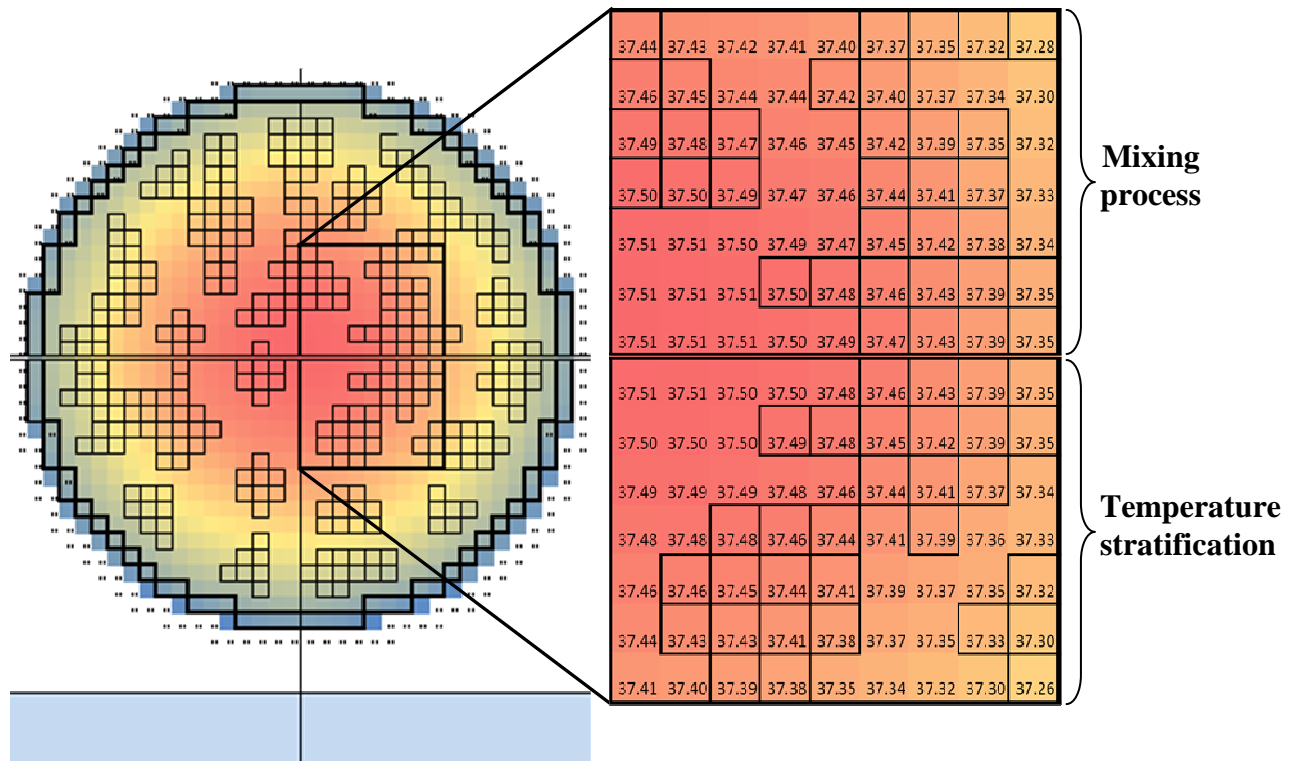


Figure 5.2 Temperature variance inside a 21 PWR waste package fully filled with water, after 20,000 years of permanent closure of the repository. The calculations are made by Excel using finite difference method. Red color represents the hottest region.

It is worthwhile to mention that the greater the heat loss from the lower parts compared to the upper parts, the more the stratification region expands, and the opposite is true, where the mixing region will expand downward if the heat loss is greater in the upper parts. However, the stratified region is likely to be larger due to the conduction loss through the emplacement pallet beneath the waste package. Given that oxygen solubility in water is very low, its diffusion is very slow, and it is consumed continuously by corrosion, it is expected that depleted oxygen cannot be replenished by diffusion, and conditions inside the pooled water turn into anaerobic, with more severe situation in the stratified regions.

### 5.2.2 Oxygen availability inside the pooled water

Anaerobic conditions inside the pooled water will slow down the corrosion of nuclear waste and hence radionuclide release. To evaluate the role of water in limiting oxygen availability, we need to compare the rate of oxygen input into the pooled water by diffusion with the rate of oxygen consumption by corrosion of the waste package components. To increase the area of water-air interface (through which oxygen diffuses) and at the same time include only the stratified region of pooled water, our calculations consider a failed waste package half filled with water. Figure 5.3 shows a schematic diagram for the steady state balance of oxygen in the pooled water.

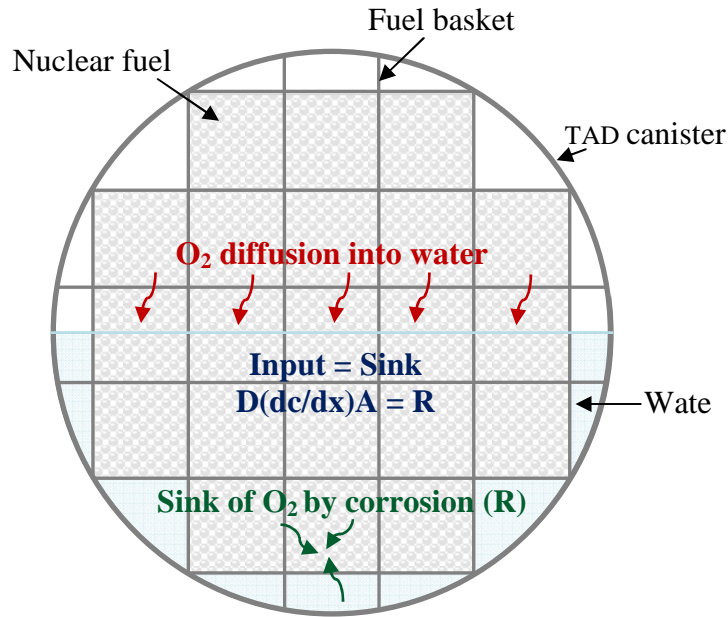


Figure 5.3 Schematic diagram illustrating steady state balance of oxygen in water pooled inside a partially failed waste container half filled with water.

Diffusion of oxygen from air into water is controlled by Henry's law:

$$C_w = k_H P \quad (10)$$

where  $C_w$  is oxygen concentration in water (mol/L),  $P$  is oxygen vapor pressure in air (atm), and  $k_H$  is Henry's law constant (L. atm /mol).

Given that oxygen vapor pressure in air is 0.2 atm, and Henry's law constant for oxygen water/air partitioning at 37 °C is  $1.07 \times 10^{-3}$  mol/L-atm (Sander, 1999), the concentration of oxygen at the surface of pooled water will be:

$$C_w = 1.07 \times 10^{-3} \times 0.2 = 2.14 \times 10^{-4} \text{ mol/L} = 6.85 \times 10^{-3} \frac{\text{kg}}{\text{m}^3}$$

With the pooled water inside the failed waste package being the control volume, oxygen mass balance at steady state is given by (Walton, 2008):

Input = Sink

$$D \frac{dC}{dx} A = S \quad (11)$$

where  $D$  is diffusion coefficient of oxygen in water ( $\text{m}^2/\text{s}$ ),  $\frac{dC}{dx}$  is oxygen concentration gradient in water ( $\text{kg}/\text{m}^4$ ),  $A$  is the surface area of water in contact with air ( $\text{m}^2$ ), and  $S$  is rate of oxygen consumption by corrosion ( $\text{kg}/\text{s}$ ).

The diffusion coefficient of oxygen in water at 37 °C is  $3.2 \times 10^{-9}$   $\text{m}^2/\text{s}$  (Landolt-Bornstein, 2007). Assuming that 1 cm beneath the water surface oxygen concentration is zero (this will over estimate oxygen flux), and oxygen flux occurs through 20 percent of the area (with the rest filled with solids) that equals to the diameter of transportation, aging and disposal (TAD) canister multiplied by its length ( $A = 8.7 \text{ m}^2$ ) (SNL, 2007 b), the rate of oxygen input will be:

$$\text{Input} = 3.2 \times 10^{-9} \frac{6.85 \times 10^{-3}}{0.01} \times (8.7 \times 0.2) = 3.8 \times 10^{-9} \text{ kg/s}$$

Oxygen is consumed by corrosion of TAD canister and its components including SNF. TAD canister and part of its components are made of SS 316; other parts are made of SS 304B4

(borated SS) and aluminum (SNL, 2007 b). In a previous paper we estimated the rate of oxygen consumption due to the corrosion of only main SS components ( $S_{SS}$ ), our calculations showed that  $S_{SS} = 6.2 \times 10^{-9}$  kg/s compared to oxygen input =  $3.8 \times 10^{-9}$  kg/s (Hamdan et al. 2010). With this rate of oxygen consumption, not to mention oxygen consumption by SNF and aluminum corrosion, conditions inside the pooled water will likely turn anaerobic (reducing conditions). Under reducing conditions steel continues to corrode, producing hydrogen gas and partially oxidized corrosion products (e.g.,  $Fe(OH)_2$ ,  $Cr(OH)_2$ ). Subsequent to complete corrosion of the metal, oxidation of the corrosion products will consume oxygen dissolved entering the pooled water. When this occurs the anaerobic period will be the longer of a) the corrosion time or b) the time for diffusion of oxygen in the solution to oxidize remaining corrosion products. As a result of the reducing conditions the corrosion rate of SNF and other components is likely to slow down until the pooled water returns to oxidizing.

To estimate for how long the reducing conditions inside the pooled water will prevail, we calculated the lifetime range and median lifetime for different components of the waste package using their rate of corrosion and their thickness. Table 5.2 shows the lifetime of main components of the waste package excluding the SNF. The corrosion rates presented in Table 5.2 are based on short-term experiments (few weeks to sixteen years) (BSC, 2004 b). Since general corrosion rates of metal and alloys tend to decrease over time as corrosion products build up on the surface and inhibit water and oxygen diffusion to the metal surface, the corrosion rates in the long term will be lower (SNL, 2007 d). Lower corrosion rates lead to lower rate of oxygen consumption. However our calculations for the rate of oxygen consumption did not include the corrosion of all waste package components, at the same time they overestimated oxygen input.

Table 5.2 Estimated lifetimes of the waste package components calculated from their thickness.

Component	Material	Nominal thickness (cm) (SNL, 2007 b)	Range and median of corrosion rate ( $\mu\text{m}/\text{yr}$ ) (BSC, 2004 b)	Lifetime range (kilo-year)	Median lifetime (kilo-year)
TAD shell	SS 316	2.54	0.001-0.048 <sub>a</sub>  0.003 <sub>a</sub>	25,400-530	8,467
TAD shell bottom		8.89		88,900-1,852	29,633
Shield plug		38.1		381,000-7,938	127,000
Basket guide assembly		0.95		9,500-198	3,167
Basket tubes		0.79		7,900-165	2,633
Inner vessel		5.08		50,800-1,058	16,933
Borated SS assembly	SS 304B4 <sub>b</sub>	1.11	161.54-252.98 <sub>c</sub>  211.84 <sub>c</sub>	0.069-0.044	0.052
Aluminum basket assembly	Al alloy	0.79	0.40-36.93 <sub>d</sub>  9.50 <sub>d</sub>	19.75-0.22	0.83

<sub>a</sub> Freshwater (29.5°C)

<sub>b</sub> Boron content of 1.1-1.2 wt percent

<sub>c</sub> 1.5 percent B, (ambient–boiling freshwater)

<sub>d</sub> Freshwater

### 5.3 EFFECT OF REDUCING CONDITIONS ON SNF CORROSION

Generally, SNF consists of  $\text{UO}_2$  matrix that contains actinides in their low oxidation state. Many important radionuclides in the SNF are redox sensitive. For example, uranium in its oxidation state IV is insoluble and immobile, but it can be remobilized by oxidation to soluble U (VI) (Szecsody et al., 1998), (Bots & Behrends, 2008). Plutonium, ranked second in terms of weight in the radionuclide inventories per waste package, has higher solubility and mobility in its higher oxidation states V and VI (Runde et al., 2002). As well, the long-lived radionuclides technetium and neptunium are abundant in nuclear waste and very soluble and mobile in their high oxidation states: Tc (VII) and Np (V) (Zachara et al, 2007), (Smellie & Karlsson, 1999), (Silin & Kareta, 1998). Consequently, under oxidizing conditions these radionuclides are oxidized to their soluble high oxidation states and released by oxidative-dissolution process. On the other hand, reducing conditions will cause reduction/precipitation mechanism and keep the radionuclides in their low oxidation states with low solubilities and mobilities, leading to delay in radionuclide release.

As we showed before, oxygen availability will be very limited in the pooled water since the rate of oxygen consumption by corrosion is greater than the rate of oxygen input by diffusion. As a result, conditions inside the pooled water are likely to be reducing and limit the oxidative-dissolution process, by which radionuclides are released. To give a quantitative perception for the effect of reducing conditions inside the pooled water on radionuclide release, we first need to estimate the rate of oxygen consumption by the corrosion of SNF under oxidizing conditions. The dissolution rate of commercial SNF (CSNF) depends on the physical and chemical prosperities of the pooled water such as: temperature, dissolved oxygen fugacity,



aqueous pH, and total carbonate concentration. Equation 12 gives the rate of CSNF dissolution as a function of these variables (BSC, 2004 a):

$$\log_{10}(F) = \log_{10}(A) + a_0 + a_1 \times IT + a_2 \times pCO_3 + a_3 \times pO_2 + a_4 \times pH \quad (12)$$

where:

$F$  = fractional dissolution rate of the CSNF ( $\text{day}^{-1}$ )

$IT$  = inverse temperature of the waste package ( $\text{K}^{-1}$ )

$pCO_3$  = negative base 10 log of total carbonate concentration (molar)

$pO_2$  = negative base 10 log of the oxygen partial pressure (atm)

$pH$  = in-package pH

$A$  = the fuel specific surface area ( $\text{m}^2/\text{mg}$ )

$a_0, a_1, a_2, a_3$ , and  $a_4$  are model parameters determined from experimental data

Oxygen fugacity is used in this equation instead of dissolved oxygen fugacity because dissolved oxygen concentration varies with temperature at constant oxygen fugacity. Under alkaline conditions ( $\text{pH} \geq 6.8$ ) Equation 12 becomes (BSC, 2004 a):

$$\log_{10}(F) = -6.7 + 4.705 - 1093.826 \times IT - 0.102 \times pCO_3 - 0.338 \times pO_2 \quad (13)$$

For temperature = 310 K (37 °C),  $pCO_3 = 2.7$  and oxygen partial pressure = 0.2 atm, the fractional dissolution rate ( $F$ ) for CSNF given by Equation 13 equals to  $9.2 \times 10^{-7}$  /d. The rate of radionuclide release ( $R_i$ ) can be found using the following equation (BSC, 2004 a):

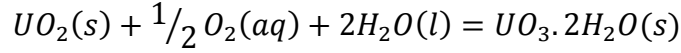
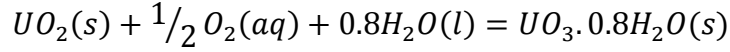
$$R_i = I_i \times F \quad (14)$$

Here  $I_i$  is the matrix inventory of the radionuclide ( $i$ ) in the waste package.

Substituting for the uranium inventory per waste package divided by two (to account only for the inventory in the pooled water) we have (SNL, 2008):

$$R_U = 4001775 \times 9.2 \times 10^{-7} = 3.68 \text{ g/d} = 4.26 \times 10^{-5} \text{ g/s} = 1.79 \times 10^{-7} \text{ mol/s}$$

The CSNF ( $UO_2$ ) is expected to undergo the following reactions upon contact with air saturated water (BSC, 2004 a):



Consequently, the rate of oxygen consumption due to CSNF corrosion is:

$$S_{CSNF} = \frac{1.79 \times 10^{-7}}{2} = 8.95 \times 10^{-8} \frac{mol}{s} = 2.86 \times 10^{-6} g/s = 2.9 \times 10^{-9} kg/s$$

The rate of oxygen consumption due to CSNF corrosion is of the same order as the rate of oxygen input into the pooled water ( $3.8 \times 10^{-9}$  kg/s). Given that there is also oxygen consumption due to the corrosion of SS ( $6.2 \times 10^{-9}$  kg/s) and Aluminum, the corrosion rate of the TAD components inside the pooled water is likely to slow down until it comes in equilibrium with oxygen rate input (a steady state).

Equation 13 applies for a range of oxygen partial pressure from 0.002 to 0.2 atm, which means a range of dissolved oxygen concentration from  $6.85 \times 10^{-3}$  to  $6.85 \times 10^{-5} \frac{kg}{m^3}$  (using Equation 10). Although oxygen concentration in the pooled water is expected to be below this range, Equation 13 can be used to examine the effect of oxygen deficiency on the corrosion rate of CSNF. Figure 5.4 shows the corrosion rate of uranium (i.e.  $UO_2$  matrix) as a function of oxygen partial pressure, using half of the uranium inventory per waste package. Figure 5.4 indicates that the corrosion rate of uranium increases as oxygen partial pressure and consequently dissolved oxygen concentration increases. This means that the oxygen deficiency inside the pooled water, which is enhanced by temperature stratification and competition on oxygen molecules, will lead to very low rates of corrosion for the materials submerged in the pooled water.

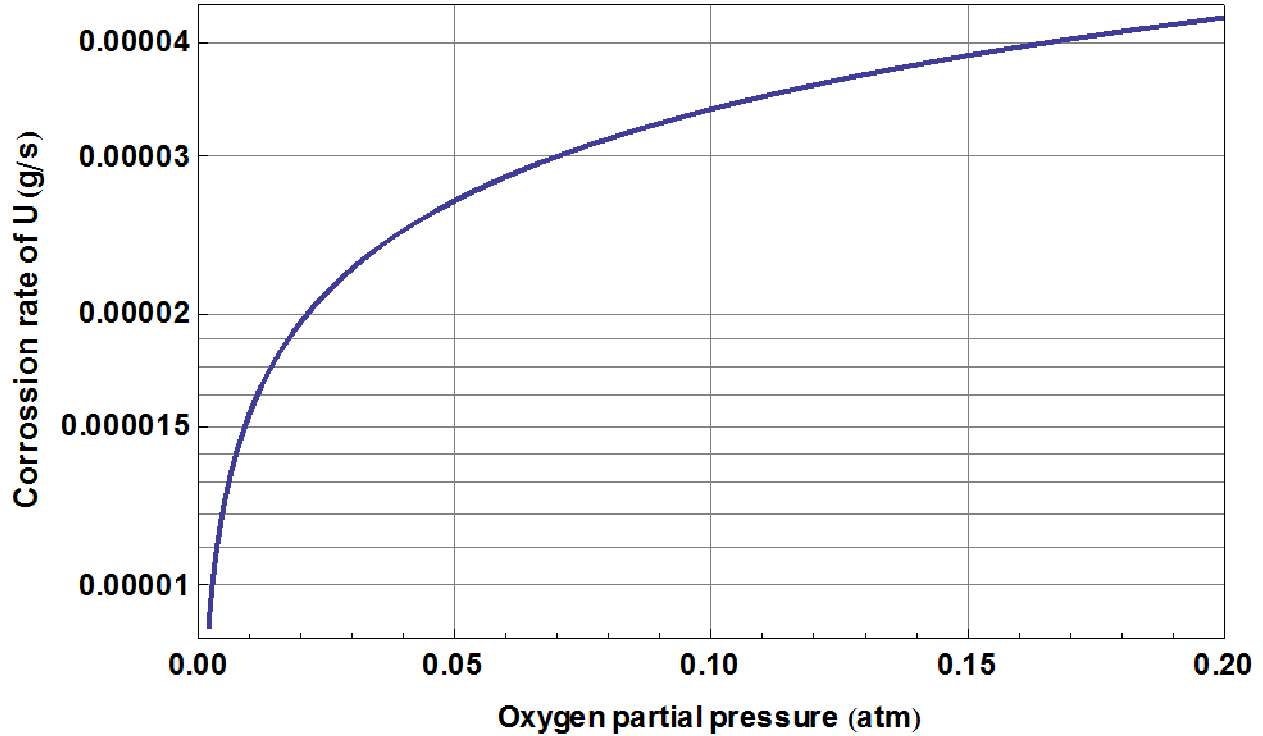


Figure 5.4 The corrosion rate of uranium as a function of oxygen partial pressure, calculated from Equation 13 at a temperature of  $310\text{ K}$  and  $pCO_3 = 2.7$

Equation 13 is also used to estimate the time required for a complete corrosion of CSNF as a function of oxygen partial pressure. The results are shown in Figure 5.5, using a temperature of  $310\text{ K}$  ( $37\text{ }^\circ\text{C}$ ), and  $pCO_3 = 2.7$  (molar). Figure 5.5 indicates that the time required for a complete corrosion of CSNF increases as the oxygen partial pressure and consequently dissolved oxygen concentration decreases. For example, the required time for a complete corrosion of CSNF increases approximately five times as the oxygen partial pressure decreases from  $0.2$  to  $0.002\text{ atm}$ . It is expected from the graph trend in Figure 5.5 that the life time of CSNF to be greater for oxygen concentrations below the range of Equation 13 (

—) and increases more rapidly as oxygen concentration decreases. Consequently, the risk

of a peak in the dose rate once the pooled water started to leak as the waste package fails from the bottom is attenuated by the very low rate of corrosion caused by oxygen deficiency.

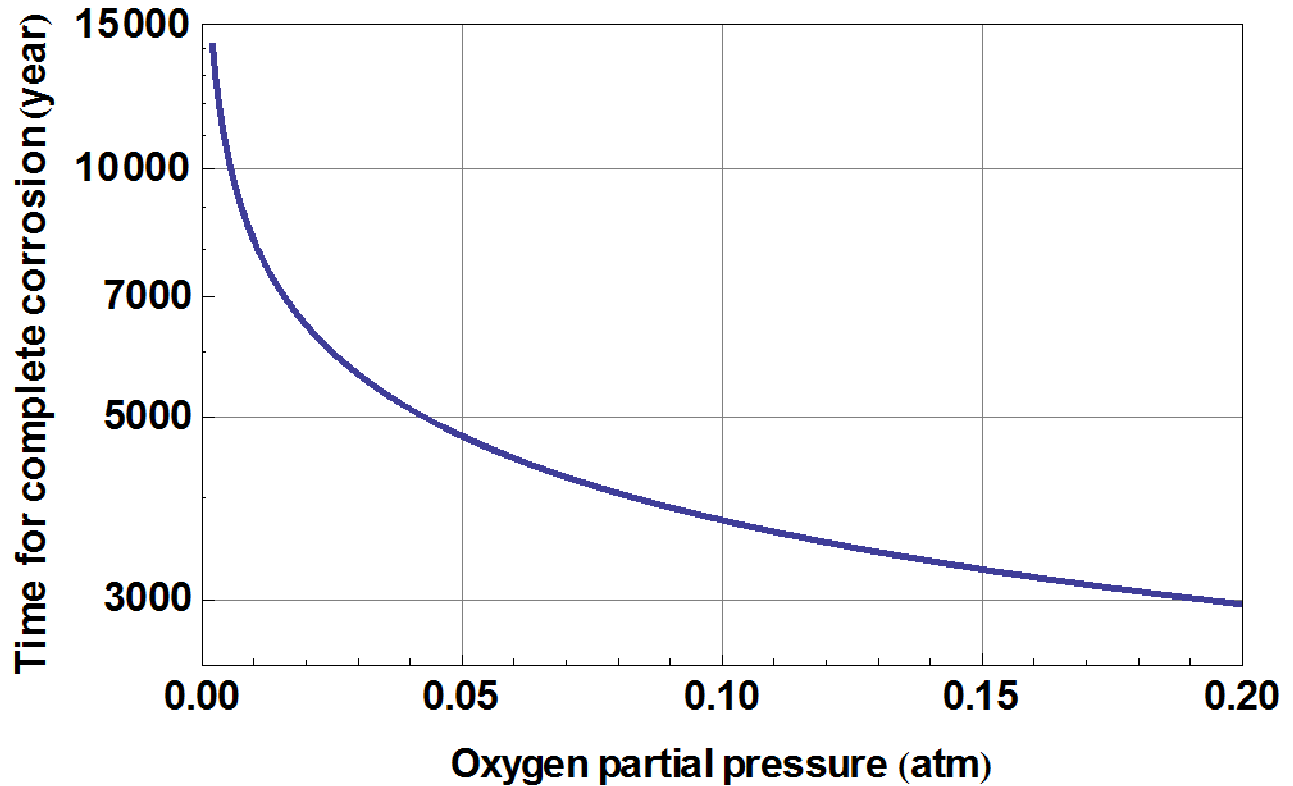


Figure 5.5 Time required for a complete corrosion of uranium as a function of oxygen partial pressure, calculated from Equation 13 at a temperature of  $310\text{ K}$  and  $pCO_3 = 2.7$

Finally, it is worthwhile to mention another possible effect of the expected reducing conditions inside the pooled water, namely, the corrosion of steel under reducing conditions. The anaerobic corrosion of iron in the waste canister will produce  $H_2$ , which is thought to inhibit the dissolution of  $UO_2$  according to some studies. Although efficient inhibition is observed in some cases, the mechanism for the inhibition of  $UO_2$  dissolution by  $H_2$  is still unclear, (Nilsson

& Jonsson, 2008) . However, as a result of this effect the radionuclide release from the waste package is expected to be more delayed.

#### **5.4 CONCLUSIONS**

Chemical and physical conditions inside the water pooled in a partially failed nuclear waste container are likely to limit the oxygen availability inside the pooled water. As a result, the rate of oxidative/dissolution process of the SNF and the corrosion rate of other TAD canister are likely to slow until the oxygen consumption by corrosion reaches equilibrium with oxygen input by diffusion. Under these conditions, the estimated life for all waste package components will be longer and consequently radionuclide release will be delayed.

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#### **REFERENCES**

- Bechtel SAIC Company ,BSC. (2004 b). Aqueous Corrosion Rates for Waste Package Materials 2.14 Concurrence Copy”, ANL-DSD-MD-000001E REV01E. Las Vegas, NV: BSC, ALA.20050323.1763.
- Bechtel SAIC Company ,BSC. (2004 a). CSNF Waste Form Degradation: Summary Abstraction, ANL-EBS-MD000015 REV 02. Las Vegas, NV: BSC. ACC: DOC.20040908.0001; DOC.20050620.0004.

- Bots, P., & Behrends, T. (2008). Uranium mobility in subsurface aqueous systems: the influence of redox conditions. *Mineralogical Magazine* , Vol. 72(1), pp. 381–384.
- Danko, G., & D Bahrami. (2004). Coupled, Multi-Scale Thermohydrologic-Ventilation Modeling with MULTIFLUX. Preprint 04-144, SME Annual Meeting and Exhibit, (pp. pp. 1-15). Denver, CO.
- Hamdan, L., Walton, J., & Woocay, a. A. (2010). Oxidation/Reduction Status of Water Pooled in a Penetrated Nuclear Waste Container. Linda Lehman(Editor), DOE Yucca Mountain Update: HLW and SNF Disposal and Perspectives from Around the Mountain. Phoenix, AZ: WM 2010 Symposium (under press).
- Hornberger, G. M. (2009). Geologic Repositories. Nuclear Fuel Cycle Course II, Vanderbilt University: Electronic document available at [www.cresp.org/NuclearFuelCycleCourseII/Presentations/20\\_Hornberger\\_Geological%20repositories\\_gmh\\_Jul09.pdf](http://www.cresp.org/NuclearFuelCycleCourseII/Presentations/20_Hornberger_Geological%20repositories_gmh_Jul09.pdf).
- International Atomic Energy Agency, IAEA. (2003). THE LONG TERM STORAGE OF RADIOACTIVE WASTE: SAFETY AND SUSTAINABILITY. Vienna, Austria: Electronic document, available at [www-pub.iaea.org/MTCD/publications/PDF/LTS-RW\\_web.pdf](http://www-pub.iaea.org/MTCD/publications/PDF/LTS-RW_web.pdf).
- Jury, W. A., Gardner, W. R., & Gardner, W. H. (1991). *Soil Physics* (5th ed.). New York: John Wiley & Sons, Inc.
- Karditsas, P. J., & Baptiste, M.-J. (1995). Thermal and Structural Properties of Fusion Related Materials: AUSTENITIC STAINLESS STEEL (316). Electronic document, available at <http://aries.ucsd.edu/LIB/PROPS/PANOS/ss.html>.

- Landolt-Bornstein. (2007). Group IV Physical Chemistry: Gases in Gases, Liquids and their Mixtures, volume 15A. Berlin/ Heidelberg, Germany: Springer.
- McEachern, R. J., & Taylor, P. (1998). A review of the oxidation of uranium dioxide at temperatures below 400 °C. *Journal of Nuclear Materials* , 254: 87-121.
- Nilsson, S., & Jonsson, M. (2008). On the catalytic effects of UO<sub>2</sub>(s) and Pd(s) on the reaction between H<sub>2</sub>O<sub>2</sub> and H<sub>2</sub> in aqueous solution . *Journal of Nuclear Materials* , Volume 372, Issues 2-3, Pages 160–163.
- OECD, RADIOACTIVE WASTE MANAGEMENT COMMITTEE OF THE OECD  
NUCLEAR ENERGY AGENCY. (1999). Geological Disposal of Radioactive Waste: Review of Developments in the Last Decade. Paris, France.
- Runde, W.; et al. (2002). Solubility and sorption of redox-sensitive radionuclides (Np, Pu) in J-13 water from the Yucca Mountain site: comparison between experiment and theory. *Applied Geochemistry* , 17:837–853.
- Sander, R. (1999). Compilation of Henry's Law Constants for Inorganic and Organic Species of Potential Importance in Environmental Chemistry. Version 3 , Electronic document, available at <http://www.henrys-law.org>.
- Sandia National Laboratories, SNL. (2007 a). EBS Radionuclide Transport Abstraction, ANL-WIS-PA-000001 REV 03. Las Vegas, Nevada: Sandia National Laboratories. ACC: DOC.20071004.0001. section 6.3.4.
- Sandia National Laboratories, SNL. (2007 d). General Corrosion and Localized Corrosion of the Drip Shield, ANLEBS-MD-000004 REV 02 ADD 01. Las Vegas, NV: SNL. ACC: DOC.20060427.0002; DOC.20070807.0004.

- Sandia National Laboratories, SNL. (2007 c). Multiscale Thermohydrologic Model. ANL-EBS-MD-000049 REV 03ADD 01. Las Vegas, NV: Sandia National Laboratories. ACC: DOC.20070831.0003 chapter 6.
- Sandia National Laboratories, SNL. (2007 b). Total System Performance Assessment Data Input Package for Requirements Analysis for TAD Canister and Related Waste Package Overpack Physical Attributes Basis for Performance Assessment, TDR-TDIP-ES-000006 REV 00. Las Vegas, NV: SNL. ACC: DOC.20070918.0005.
- Sandia National Laboratories, SNL. (2008 ). Total System Performance Assessment Model/Analysis for the License Application, MDL-WIS-PA-000005 REV00. Las Vegas, Nevada: Sandia National Laboratories, DOC.20080204.0003, section 6.3.2.
- Silin, V., & Kareta, A. (1998). The interaction of Np(V), Pu(VI) and Tc(VII) with metal in alkaline solutions. *Journal of Alloys and Compounds* , Volumes 271-273, Pages 803-808.
- Smellie, J. A., & Karlsson, F. (1999). The use of natural analogues to assess radionuclide transport. *Engineering Geology* , Volume 52, Issues 3-4, Pages 193-220.
- Special Metals Corporation. (2006). INCONEL® alloy 22. Publication Number SMC-049. Electronic document, available at:  
<http://www.specialmetals.com/documents/Inconel%20alloy%2022.pdf>.
- Szecsody, J. E.; et al. (1998). Uranium Mobility During In Situ Redox Manipulation of the 100 Areas of the Hanford Site. Pacific Northwest National Laboratory Richland, Washington 99352, PNNL-12048UC-2000,.
- Walton, J. C. (2008). Fate and Transport of Contaminants in the Environment (1st ed.). Glen Allen, USA: College Publishing.



Zachara, J. M.; et al. (2007). Reduction of pertechnetate [Tc(VII)] by aqueous Fe(II) and the nature of solid phase redox products. *Geochimica et Cosmochimica Acta* , 71: 2137–2157.

## Chapter 6

### 6. General Conclusions

**Heat release characteristics of the nuclear waste stored in an unsaturated zone geological repository contribute to the safety of the repository.** The effect of the residual heat release of the nuclear waste on radionuclide release was tested for the two main scenarios of the nuclear waste packages failure: the flow-through model and the bathtub model. In the flow-through model, heat release from the nuclear waste leads to bidirectional transport in the sheltered areas: advective transport toward the warmest regions and diffusive transport in the opposite direction. As a result, heat-generating waste stored in a partially saturated environment tends to sequester radionuclides in the sheltered areas and prevent their release. The radionuclide sequestration model showed that the sequestration process is more effective at higher rates of heat generation and lower moisture conditions. In addition, the minimum size of sheltered areas needed to prevent radionuclide release increases over time as the amount of heat generated from the waste package decreases. On the other hand, the size of sheltered areas decreases over time due to general corrosion. A convolution of these two processes suggests that radionuclide release is likely to be gradual and long delayed, even in the case of early penetration by localized corrosion.

In the bathtub model, heat release from the nuclear waste creates temperature stratification inside the water pooled in the lower half of the waste package. This will limit oxygen availability inside the pooled water, since the diffusion coefficient of oxygen in water is very small. As a result, the rate of oxygen input into the pooled water will be less than the rate of oxygen consumption caused by the corrosion of different components of the waste package and

the conditions will turn anaerobic. In this case, the rate of oxidative/dissolution process of the spent nuclear fuel (SNF) and the corrosion rate of other waste package components are likely to slow until the oxygen consumption by corrosion reaches equilibrium with oxygen input by diffusion. Under these conditions, the estimated life for all waste package components including SNF will be longer and consequently radionuclide release will be delayed.

## Vita

Lubna Kamel Hamdan was born in Kuwait. The second daughter of Kamel Hamdan and Nazeera Rashed, she graduated from the Department of Physics at Yarmouk University-Jordan in 1986. After her Bachelor's degree, she worked in Kuwait as a teacher of Science. She gained her Master's degree in physics from the University of Jordan in 1992. Her thesis was "*Ion Beam Mixing Theory & Experimentation*", where she used Argon ions ( $\text{Ar}^+$ ) to induce mixing in the bi-layer thin films system Selenium (Se) / Cadmium (Cd) evaporated on a silicon substrate. After graduation, she worked as a teacher of Physics and Math in community college and high school in Jordan.

Lubna came to the United States in August, 2005 to join the Environmental Science and Engineering PhD program at the University of Texas at El Paso. During her study at UTEP she participated as a speaker in several national and international conferences. While at UTEP, she authored and coauthored scientific publications in some of the most prestigious international journals in the fields of Energy, Environment, and Geological Engineering and recognized as a Miner Hero in the field of research in March 2010. Below are enlisted her recent publications and presentations.

### **Publications and Conference Presentations**

#### ***Peer-reviewed Journal***

**Lubna K. Hamdan**, Maryam Zarei, Russell R. Chianelli, and Elizabeth Gardner, 2008,

"Sustainable Water and Energy in Gaza Strip", Renewable Energy magazine, 33  
(2208): 1137 – 1146.

John C. Walton, **Lubna K. Hamdan**, and Arturo Woocay, 2010, “Conceptual Model for Radionuclide Release, Yucca Mt., Nevada, USA”, Applied Geochemistry, Accepted 11 Feb, 2010. doi:10.1016/j.apgeochem.2010.02.006.

**Lubna K. Hamdan**, John C. Walton, and Arturo Woocay, 2010, “SAFETY IMPLICATION FOR AN UNSATURATED ZONE NUCLEAR WASTE REPOSITORY”, Energy Policy: in Press

**Lubna K. Hamdan**, John C. Walton, and Arturo Woocay, 2010, “Effect of Water Pooled in a Failed Nuclear Waste Package on Radionuclide Release”, Environmental & Engineering Geoscience: Under review.

#### *Conference Papers*

**Hamdan, L.K.**, J.C. Walton, and A. Woocay, (2008), Limitations and patters of radionuclide release from partially failed containers, International High-Level Radioactive Waste Management Conference (IHLRWMC), Las Vegas, Nevada. La Grange Park, Illinois: American Nuclear Society. September 7-11, 2008, p. 348-351. ISBN: 978-0-89448-062-1

**Hamdan, L. K.** and J.C. Walton, (2008), “Limitations on Radionuclide Release from Partially Failed Containers”, Materials Research Society (MRS): Fall Meeting, Symposium Q (Volume 1124), Scientific Basis for Nuclear Waste Management XXXII. Boston, MA., December 1-5, 2008. Paper #: 1124-Q08-07 DOI: 10.1557/PROC-1124-Q08-07 [http://www.mrs.org/s\\_mrs/sec\\_subscribe.asp?CID=16763&DID=230332&action=detail](http://www.mrs.org/s_mrs/sec_subscribe.asp?CID=16763&DID=230332&action=detail)

**Hamdan, L. K.**, J. Walton and, A. Woocay, (2009), “Effect of Physical Processes on Radionuclide Release from Partially Failed Containers”, Waste Management Symposium 2009: Waste Management for the Nuclear Renaissance, Mar 1-5, 2009. Session 57, Publisher: Curran Associates, Inc. (Oct 2009) Pages: 5,143 (8 Vols) ISBN13: 9781615672523.

**Hamdan, L. K.**, J. Walton, A. Woocay, and Omar Al-Qudah, (2010), “Oxidation/Reduction Status of Water Pooled in a Penetrated Nuclear Waste Container.”, abstract 10265. Talk presented at Waste Management (WM) 2010 Symposia, Phoenix, AZ, March 10, 2010; in press in 2010 WM Symposium proceedings

### ***Abstracts***

**Hamdan,L.K.**, M. Zarei, R. Chianelli, and E. Gardner,(2007). “Sustainable Water and Energy in Gaza Strip”, Eos Trans. AGU, 88(52), Fall Meet. Suppl., Abstract H33G-1707, 2007 Presented at American Geophysics Union (AGU) 2007 Fall Meeting, San Francisco, CA, December 12, 2007.

Walton, J. C., **L. K. Hamdan**, and A. Woocay, (2008), “Controlling Features and Processes for Radionuclide Release from Yucca Mountain”, Presented at: Devil’s Hole Workshop, Furnace Creek, CA, May, 2008.

**Hamdan, L.K.**, J.C. Walton, and A. Woocay, (2008), “Conceptual Model for Radionuclide Release from the Engineered Barrier Systems (EBS) at Yucca Mountain.” Eos Trans. AGU, 89(53), Fall Meet. Suppl., Abstract H53E-1130, 2008, Presented at: American Geophysical Union Fall Meeting, San Francisco, CA, December 19, 2008.

- Hamdan, L. K.,** J.C. Walton, and A. Woocay, (2009), “Unsaturated Zone Nuclear Waste Repository - A Safety Case” Presented at: Devil’s Hole Workshop, Furnace Creek, CA, May, 2009.
- Hamdan, L. K.,** J.C. Walton, and A. Woocay, (2009), “A Case Study for the Performance of a Nuclear Geological Repository in the Unsaturated Zone”, Geological Society of America Abstracts with Programs, Vol. 41, No. 7, p. 89. Poster in Session 27 Paper 15, Portland, OR, October 18-21, 2009. ISSN: 0016-7592
- Hamdan, L. K.,** J.C. Walton, and A. Woocay, (2009), “Chemical and Physical Status of Water Pooled in a Failed Nuclear Waste Container”, Talk 426 presented at: 65rd Southwestern Regional Meeting of the American Chemical Society (ASC), El Paso, TX, November 4-7, 2009. <http://acs.confex.com/acs/swrm09/webprogram/Paper76634.html>.
- Hamdan, L.K.,** J.C. Walton and A. Woocay (2009), “Conditions inside Water Pooled in a Failed Nuclear Waste Container and its Effect on Radionuclide Release”, Eos Trans. AGU, 90(52), Fall Meet. Suppl., Abstract H31E-0835, 2009, Presented at: American Geophysical Union Fall Meeting, San Francisco, CA, December 16, 2009.
- Maryam Zarei, **Lubna Hamdan,** Russell R. Chianelli, and Elizabeth Gardner, (2010), “A Sustainable Water and Energy In Gaza Strip” , presented at Building Partnerships and Pathways to Address Engineering Grand Challenges Conference, February 8-10, 2010, **First Place Poster Presentation Award Winner.**

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