Key Processes and Parameters in a Generic Clay Disposal System Model

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INTRODUCTION

Sensitivity analysis was performed with respect to various key processes and parameters affecting long-term post-closure performance of geologic repositories in clay media. Based on the detailed computational Clay Generic Disposal System Model (GDSM) developed by the Used Fuel Disposition (UFD) campaign [1], these results provide an overview of the relative importance of processes that affect the repository performance of a generic clay disposal concept model. Further analysis supports a basis for development of rapid abstracted models in the context of system level fuel cycle simulation. Processes and parameters found to influence repository performance include the rate of waste form degradation, timing of waste package failure, and various coupled geochemical and hydrologic characteristics of the natural system including diffusion, solubility, and advection.

DESCRIPTION OF THE ACTUAL WORK

Clay Generic Disposal System Model

These analyses were performed using the Clay GDSM developed by the UFD campaign [1].

The Clay GDSM is built on the GoldSim simulation framework and contaminant transport model. This radionuclide transport toolset simulates chemical and physical attenuation processes including radionuclide solubility, dispersion phenomena, and reversible sorption [2, 3]. Model input parameters supporting modeling of these tranport processes include geometry specifications (e.g. repository depth), geologic material properties (e.g. clay porosity), geochemical data (e.g. elemental solubility limits), and environmental parameters (e.g. natural system velocity). Probabilistic elements of the GoldSim modeling framework enable the models to incorporate simple probabilistic Features, Events, and Processes (FEPs) that affect repository performance.

The disposal concept modeled by the Clay GDSM is an array of spent nuclear fuel packages within a clay repository envrionment, 500 meters beneath the earth's surface. In this repository concept, the waste packages contain an inventory of spent nuclear fuel, are emplaced horizontally in excavated tunnels, and are backfilled by a reducing bentonite clay buffer material within the tunnel [4].

In the UFD GDSM tool that was used for this analysis, the clay repository concept is modeled using a single rectangular disposal cell. The disposal cell models a single cylindrical spent nuclear fuel package surrounded by bentonite buffer material and emplaced in the clay geologic medium. Using reflective boundary conditions in the horizontal plane, an infinite disposal cell array is modeled [**?**].

The model includes vertical advective fast pathway as well as an Engineered Barrier System (EBS) which can undergo rate based dissolution and barrier failure. Releases from the EBS enter near field and subsequently far field host rock regions in which diffusive and advective transport take place, attenuated by solubility limits as well as sorption and dispersion phenomena [1].

Sampling Scheme

This analysis has undertaken an analysis strategy to develop a many dimensional overview of the key factors in modeled repository performance. To achieve this, both individual and dual parametric cases were performed.

Individual parameter cases varied a single parameter of interest in detail over a broad range of values. Dual parameter cases were performed for pairs of parameters expected to exhibit some covariance. For each parameter or pair of parameters, forty simulation groups varied the parameter or parameters within the range considered. Each case and its parametric range are detailed in Table 1.

Simulation Cases

Case	Parameter	Units	Min. Value	Max. Value
I	D_{eff}	$[m^2 \cdot s^{-1}]$	10^{-8}	10^{-5}
	Inventory	[MTHM]	10^{-4}	10^{1}
II	$V_{adv,y}$	$[m \cdot yr^{-1}]$	6.31×10^{-8}	6.31×10^{-4}
	D_{eff}	$[m^2 \cdot s^{-1}]$	10^{-8}	10^{-5}
III	S_i	$[mol \cdot m^{-3}]$	$(1 \times 10^{-9})\langle S_i \rangle$	$(5 \times 10^{10})\langle S_i \rangle$
IV	$K_{d,i}$	$[m^3 \cdot kg^{-1}]$	$(1 \times 10^{-9})\langle K_{d,i} \rangle$	$(5 \times 10^{10})\langle K_{d,i} \rangle$
V	$R_{WFDeg.}$	$[yr^{-1}]$	10^{-9}	10^{-2}
	Inventory	[MTHM]	10^{-4}	10^{1}
VI	t_{WPFail}	$[yr]$	10^{3}	10^{7}
	D_{eff}	$[m^2 \cdot s^{-1}]$	10^{-8}	10^{-5}

TABLE 1: Each dual and single parameter simulation case had 40 simulation groups of 100 realizations each.

For each simulation group, a 100 realization simulation was completed. Each realization held the parameters being analyzed as constant and sampled stochastic values for uncertain parameters not being studied. A sampling scheme developed in previous generic disposal media modeling was implemented in this model in order to ensure that the each 100 realization simulation sampled identical values for uncertain parameters [1, 4].

Mean of the Peak Annual Dose

In this analysis, repository performance is quantified by radiation dose to a hypothetical receptor. Specifically, this sensitivity analysis focuses on parameters that affect the mean of the peak annual dose. The mean of the peak annual dose,

$$D_{MoP,i} = \frac{\sum_{r=1}^{N} \max\left[D_{r,i}(t)|_{\forall t}\right]}{N} \quad (1)$$

where

$D_{MoP,i} =$ mean of peak annual dose due to isotope i $[mrem/yr]$
$D_{r,i}(t) =$ year t dose in realization r due to isotope i $[mrem/yr]$
$N =$ number of realizations,

is a conservative metric of repository performance and should not be confused with the peak of the mean annual dose.

RESULTS AND ANALYSIS

Case I : Diffusion Coefficient and Inventory

In clay media, diffusion dominates far field hydrologic transport due to characteristically low hydraulic head gradients and permeability.

The sensitivity of the peak dose to the effective diffusion coefficient, D_{eff}, in the host rock was analyzed in conjunction with the inventory total mass. The parametric range for these variables (see Table 1), were chosen to cover the full range of expected diffusion coefficients in clay and inventories in current wasteforms.

In order to isolate the effect of the far field behavior, the waste form degradation rate was set to be very high as were the solubility and advective flow rate through the EBS. This unhindered contaminant flowthrough in the near field and left far field transport as the sole remaining physical barrier to release.

Peak dose due to highly soluble, non-sorbing elements such as I and Cl was found to be proportional to the radionuclide inventory and to the relative diffusivity.

For sorbing and solubility limited elements, two diffusion coefficient regimes were visible in the results of this analysis. In the low diffusion coefficient regime, the diffusive pathway through the homogeneous permeable porous medium in the far field continues to be a dominant barrier to nuclide release for normal (non-intrusive) repository conditions. In the second regime, for very high diffusion coefficients, the effects of additional attenuation phenomena in the natural system can be seen.

The dependence of peak annual dose on mass factor was consistently directly proportional for all isotopic groups, though slight attenuation of this dependence was seen for higher mass factors.

Case II : Vertical Advective Velocity and Diffusion Coefficient

In this analysis, the threshold between primarily diffusive and primarily advective transport was investigated by varying the vertical advective velocity, $v_{adv,y}$, in conjunction with the diffusion coefficient, D_{eff}, as in Table 1. For the low diffusion coefficients and low advective velocities usually found in clay media, the model demonstrated behavior entirely in the diffusive regime. However, as the vertical advective velocity was increased, system behavior increasingly approached the advective regime.

Soluble and non-sorbing nuclides ^{129}I and ^{36}Cl are more sensitive to vertical advective velocity for lower vertical advective velocities. It was shown that for vertical advective velocities above a threshold ($6.31 \times 10^{-6}[m/yr]$), lower reference diffusivities are ineffective at attenuating the mean of the peak doses for soluble, non-sorbing elements.

Solubility limited and sorbing elements such as Tc and Np show a very weak influence on peak annual dose rate for low reference diffusivities, but show a direct proportionality between dose and reference diffusivity above a threshold. For ^{99}Tc, for example, that threshold occurs at $1 \times 10^{-11}[m^2/s]$. Dose contribution from these elements show a proportional relationship with vertical advective velocity above a regime threshold at $6.31 \times 10^{-5}[m/yr]$, above which the system exhibits sensitivity to advection.

Case III : Solubility Coefficients

The dissolution behavior of a solute in an aqueous solution is called its solubility. This behavior is limited by the solute's solubility limit, described by an equilibrium constant that depends upon temperature, water chemistry, and the properties of the element.

By varying the multiplication factor applied to the expected solubility limit, $\langle S_i \rangle$, this analysis varied elemental solubility limits, S_i, for each isotope i, as detailed in Table 1. This method preserves, in some sense, the relative solubility limits between isotopes, which vary over many magnitudes.

For solubility limits below a threshold, approximately $1 \times 10^{-10}[mol \cdot m^{-3}]$, dose releases were directly and strongly proportional to the solubility limit. This indicated that the radionuclide concentration saturated the groundwater up to the solubility limit near the waste form. For solubility limits above the threshold, however, further increase to the limit had no effect on the peak dose. This demonstrated the situation in which the solubility limit was so high that even complete dissolution of the waste inventory into the pore water was insufficient to reach the solubility limit.

Case IV : Partition Coefficient

This analysis investigated the sensitivity of repository performance to the elemental partition coefficient.

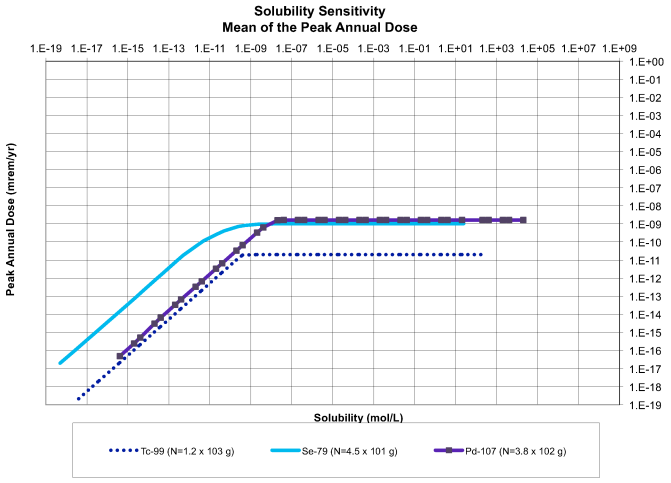

Fig. 1: Solubility limit sensitivity. The peak annual dose due to an inventory, N, of each isotope.

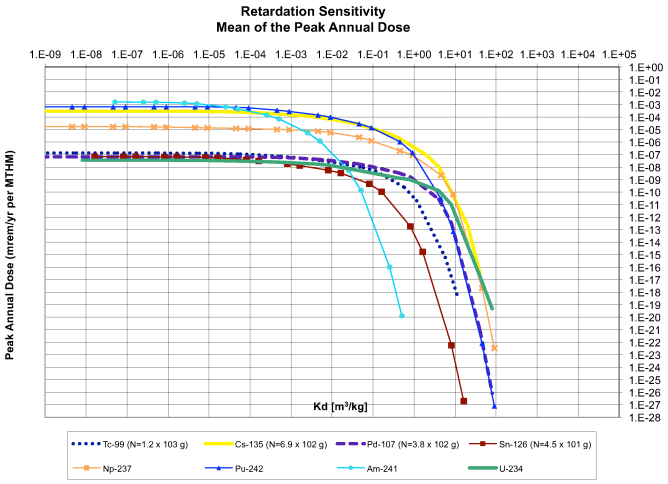

Fig. 2: K_d sensitivity. The peak annual dose due to an inventory, N, of each isotope.

The partition or distribution coefficient, K_d, relates the amount of contaminant adsorbed into the solid phase of the host medium to the amount of contaminant adsorbed into the aqueous phase of the host medium. It is a common empirical coefficient used to capture the effects of a number of retardation mechanisms. The coefficient $K_{d,i}$, in units of $[m^3 \cdot kg^{-1}]$, is the ratio of the mass of contaminant i in the solid to the mass of contaminant i in the solution.

As indicated in Table 1, the parameters in this model were all set to the default values except a multiplier applied to the expect partitioning coefficients, $\langle K_{d,i} \rangle$, for each isotope i. This multiplier preserved, in some sense, the widely varying relative sorption behavior among elements.

The expected inverse relationship between the partition coefficient resulting peak annual dose was found for all elements that were not assumed to be effectively infinitely soluble. It is clear from Figure 2 that for partition coefficients greater than a threshold, approximately $1 \pm 10[m^3 \cdot kg^{-1}]$ the relationship between peak annual dose and partition coefficient is a strong inverse one.

Case V : Waste Form Degradation Rate and Inventory

The sensitivity of peak dose rate to the waste form degradation rate was determined with respect to varying inventories of waste. These parameters were varied as indicated in Table 1.

For realizations in which the dominant dose contributing radionuclides have half-lives much shorter than the expected waste form lifetime, the waste form degradation rate did not have an effect. So too, for cases in which the primary barrier to release, the slow diffusive pathway, dominates overall repository performance, the waste form engineered barrier had a negligible effect on repository performance in comparison.

These results show two regimes. In the first regime, the mean of the peak annual dose rates is directly proportional to both the mass factor and the fractional waste form degradation rate. For some radionuclides, attenuation occurs for high values of both parameters as the release of radionuclides is limited by dispersion parameters. This phenomenon can be seen in the figures below in which transition between regimes for higher degradation rates happens at lower mass factors than transition between regimes for lower degradation rates.

The peaks for highly soluble, non sorbing elements such as I and Cl are directly proportional to mass factor for most values of waste form degradation rates.

In Figure 3, highly soluble and non-sorbing ^{129}I demonstrated a direct proportionality between dose rate and fractional degradation rate until a turnover where other natural system parameters dampened transport. It also domonstrated a direct proportionality to the inventory multiplier, as seen in Figure 4.

The peaks for solubility limited, sorbing elements such as Tc and Np, on the other hand, have a more dramatic turnover. For very high degradation rates, the dependence on mass factor starts to round off due to attenuation by solubility limits, as can be seen in Figures 5, and 6.

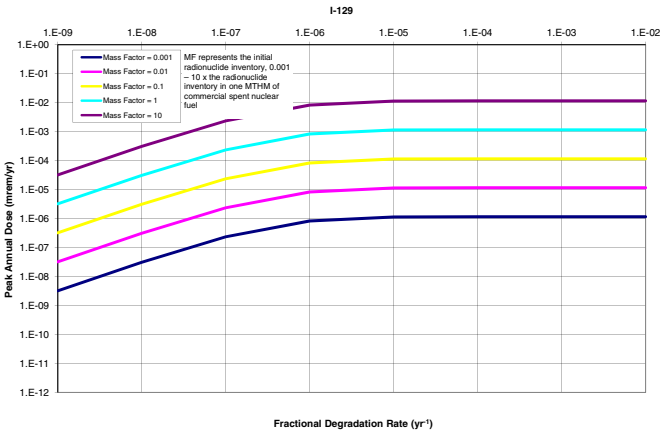

Fig. 3: ^{129}I waste form degradation rate sensitivity.

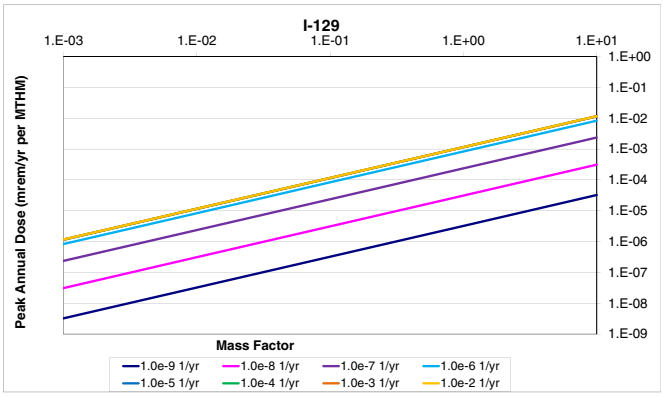

Fig. 4: ^{129}I inventory multiplier sensitivity.

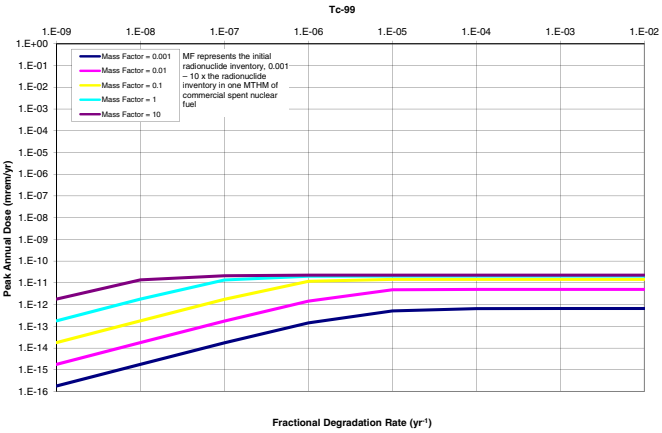

Fig. 5: ^{99}Tc waste form degradation rate sensitivity.

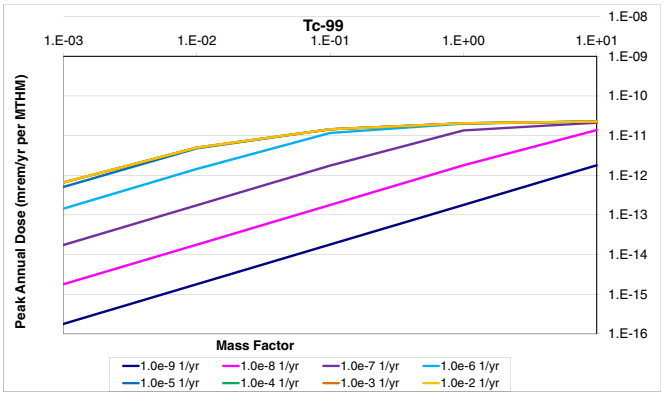

Fig. 6: ^{99}Tc inventory multiplier sensitivity.

Solubility limited and sorbing isotopes such as ^{99}Tc demonstrated a direct proportionality to fractional degradation rate until attenuation by their solubility limits and other natural system parameters.

Case VI : Waste Package Failure Time and Diffusion Coefficient

To investigate the effect of the waste package failure time, it was varied over five magnitudes from one thousand to one million years. Simultaneously, the reference diffusivity was varied over the eight magnitudes between 1×10^{-8} and 1×10^{-15} in order to determine the correlation between increased radionuclide mobility and the waste package lifetime.

For the clay repository, the waste package failure time was shown to be entirely irrelevant until waste package failure times reach the million year time scale, at which point it has an effect primarily on the dose due to long lived isotopes.

ACKNOWLEDGEMENTS

This work is supported by the U.S. Department of Energy, Basic Energy Sciences, Office of Nuclear Energy, under contract # DE-AC02-06CH11357.

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