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Assessment of the Potential Effects of Colloidal Radionuclide Transport on Nuclear Waste Repository Performance

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Abstract

The NRC is independently evaluating technical issues such as colloidal transport in preparation for reviewing an anticipated license application from the U.S. Department of Energy (DOE) for a proposed high level nuclear waste repository at Yucca Mountain, Nevada. For the evolving conditions of the proposed repository over 10,000 years into the future, the influence of colloids in enhancing radionuclide transport is difficult to estimate and highly uncertain. NRC staff are conducting a multi pronged approach to assessing whether or not these uncertainties are sufficiently bounded by the performance assessment being developed by the DOE. Preliminary bounding calculations providing an estimate of the upper limit of calculated dose from colloidal Pu suggest that an effect on dose is plausible. A more sophisticated effort involves analytical modeling of colloidal Pu transport that uses laboratory and field data to represent more accurately processes such as kinetic controls on sorption and desorption of Pu to colloids. This modeling effort shows that slow desorption from colloids is a factor that could enhance radionuclide migration. Finally, an abstraction of colloidal transport is being implemented in the NRC total system performance assessment model in order to integrate potential colloidal effects at the system level. This implementation is flexible enough that a variety of sensitivity studies can be conducted that will aid identification of the model parameters most significant to transport.

Introduction

Yucca Mountain, Nevada, is a potential site for disposal of high level nuclear waste. The NRC, with technical assistance from the CNWRA, is reviewing DOE studies and models for the potential repository, and developing independent models, in anticipation of reviewing a DOE license application. One technical issue is whether radionuclide transport could be enhanced by colloids to a degree that an adverse dose consequence would result. Estimating the potential risk due to colloid transport may be highly uncertain; nevertheless, evaluating that risk may help focus our review on the individual processes and parameters that may be most significant for colloid transport.

Understanding the degree to which uncertainties can affect confidence in estimates of dose and barrier performance is central to evaluating performance assessments in support of a repository license application. This paper discusses our three-pronged approach to addressing whether DOE colloid release and transport models and parameters sufficiently bound uncertainties. The three components of this approach are: (i) simple, preliminary calculations of the potential dose effect of colloidal effects, (ii) more sophisticated process-level modeling, and (iii) abstraction of colloidal effects into the NRC's total-system performance assessment (TPA) model.

DOE Model Approach to Colloidal Release and Transport

The DOE Total System Performance Assessment - Site Recommendation (TSPA-SR; see diagram at right) considered three colloid types:

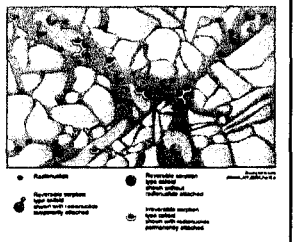
- groundwater
- corrosion product
- waste form (glass only).

Radionuclide release and transport are facilitated by colloids in two ways:

- reversible attachment to all three types
- irreversible attachment to waste form type.

Waste form colloids with irreversibly attached radionuclides are retarded in the unsaturated and saturated zones. Transport of reversibly-attached radionuclides is modeled by adjusting radionuclide parameters. DOE is considering some model changes (Aguilar and Alcorn, 2003), but the basic approach is unchanged. Key concepts and parameters that are uncertain include, but are not limited to:

- concentrations of colloids along the transport pathway,
- degree and reversibility of radionuclide attachment to colloids,
- model abstraction of physical and chemical filtration of colloids, and
- dispersion and dilution effects on colloidal radionuclide concentrations.



DOE illustration of conceptual model of colloid transport for use in performance assessment. Figure 3-60 from U.S. DOE (2001).

1 "Back-of-the-Envelope" Calculation

To what degree could colloidal transport influence dose? We calculate the magnitude of mobilized colloidal Pu from spent fuel by applying laboratory release rates to projected flow rates at Yucca Mountain. Key conservative assumptions are:

- all waste packages fail
- colloidal Pu concentration in groundwater is not reduced by retardation or filtration until diluted when pumped
- decay is neglected
- no desorption of Pu once sorbed to colloids.

Non-conservative assumptions include:

- only Pu is considered
- colloids may not be diluted uniformly in pumped groundwater
- supporting lab data may not have recovered all colloids
- some parameters may not be maxima.

On balance, the calculation is likely to be conservative. Therefore, this calculation is not intended to be realistic, but rather to demonstrate an upper bound effect. The equation used is:

$$doserate = \frac{[Pu] \times FR \times N}{PR} DCF$$

Parameter	Value	Source
$[Pu]$ = colloidal Pu concentration	1.2×10^{8} Bq/m^3	Leach test data[1] on ^{239}Pu and ^{240}Pu and calculation of Pu sorbed from solution–assuming no desorption–using high values for dissolved Pu, colloid concentration, and K_d onto colloids[2,3]
FR = water flow through waste package	6.7×10^{-3} m^3/y	TPA 4.1 average over time period during which flow takes place (S. Mohanty, personal communication, 2003).
N = number of affected waste packages	4,439	Conservatively assume all packages fail. TPA 4.1 average for fraction of failed packages that "see" water is one-half[3].
PR = well pumping rate at point of exposure	3.7×10^{6} m^3/y (3000 acre-feet/y)	The "representative volume" of groundwater by which radionuclides are delivered to the potential dose recipients is fixed by regulation[4].
DCF = dose conversion factor	1.02×10^{-4} (Sv/y)/(Bq/m^3)	TPA 4.1 values for for ^{239}Pu and ^{240}Pu for drinking and irrigating under current conditions[3].

[1]Wilson (1988). [2]Mohanty et al. (2002) [3]CRWMS M&O (2000b). [4]U.S. Code of Federal Regulations, Title 10, Part 63.

The calculated dose rate from colloidal $^{239+240}$Pu is 9.8×10^{-4} Sv/y (98 mrem/y) and the alpha activity concentration in the representative volume is 960 Bq/m^3 (26 pCi/L). There is no time component to this calculation; waste package lifetimes and transport times, therefore, are not considered. These results do not demonstrate that colloidal Pu will lead to unacceptable dose or groundwater radioactivity; rather, they merely show that colloidal effects are potentially significant and that the abilities of natural and engineered barriers to delay or attenuate colloidal radionuclides need to be demonstrated by DOE.

2 Process-Level Modeling

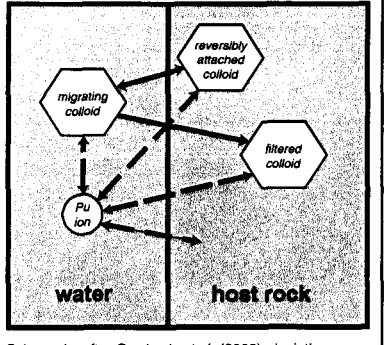
Process-level models aid the interpretation of laboratory and field data and in, the absence of sufficient data, help better define which parameters and processes are likely to most strongly affect model results. Cvetkovic et al. (2002) and Painter et al. (2002) developed an analytical colloid sorption model with first-order, two-site, forward and reverse sorption rates and used the model to interpret laboratory data on colloidal Pu sorption (Lu et al., 1998, 2000). This model pointed to the importance of solute-colloid sorption rates and the relative sluggishness of desorption.

Cvetkovic et al. (2003) expanded on this model. The figure to the right shows colloid-solute-host rock interactions that were considered, including:

- solute sorption to colloids and host rock,
- reversible sorption of colloids to host rock, and
- irreversible filtration of colloids by host rock.

Emphasis was on kinetic rates of the exchanges depicted in the figure. Three parameters that are particularly uncertain at Yucca Mountain are the solute-colloid desorption rate, the colloid irreversible removal (or filtration) rate, and the colloid retardation factor (i.e., for sorption of colloids to host rock). Generic and Yucca Mountain-specific test cases showed strong sensitivity to the first two parameters and lesser sensitivity to the last. These results suggest that, in the absence of new data, bounding values for these parameters would be appropriate in performance assessment models.

In the model, colloid retardation (except at very high values) is relatively ineffective at reducing Pu transport due to exchange between solution and reversibly attached colloids. Pu discharge was typically highest at colloid retardation factors of 10-100; therefore, using an apparently conservative value of one may actually underestimate the effect. Also, the model showed that the apparently conservative assumption of irreversible solute-colloid sorption is bounding only if colloid filtration is neglected.

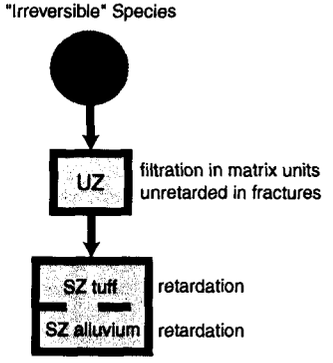


Schematic, after Cvetkovic et al. (2003), depicting solute-colloid interactions considered in process-level colloid-enhanced transport model. Solid lines indicate either exchange of colloids between water and reversible sites on host rock, or permanent removal by filtration. Dashed lines indicate reversible solute sorption onto colloids and host rock surfaces.

3 Total-system Performance Assessment Implementation

Previously, NRC/CNWRA simulated potential colloid total-system effects by assuming no retardation for selected radioelements. Contardi et al. (2001), using TPA version 3.2 and assuming no sorption or diffusion for Pu and Am isotopes, obtained a peak dose rate of 7.4×10^{-4} Sv/y (74 mrem/y) at 45,000 years–past the 10,000-year compliance period. However, the peak dose delay was more a function of waste package lifetime, rather than a direct reflection of colloidal processes or groundwater travel time (estimated average of 600 to 1,600 years; Mohanty et al., 2002). Similarly, Mohanty et al. (2002), using TPA version 4.1, assumed no retardation of Pu, Am, and thorium (Th) isotopes. Compared to the base case, this case gave dose rates one to three orders of magnitude higher, with a peak dose rate of 2×10^{-3} Sv/y (200 mrem/y) at 77,000 years. These results, though not indicative of a realistic scenario due to highly conservative assumptions, nevertheless demonstrate that colloid-enhanced radionuclide mobility can affect performance of the natural barrier below Yucca Mountain.

We are now modifying the radionuclide release and transport abstractions in TPA version 5.0 to explicitly include colloid effects. The basic approach to the abstraction is similar to that adopted by the DOE (CRWMS M&O, 2000a, 2000b; Aguilar and Alcorn, 2003): radionuclide release is enhanced by colloids, and both reversible and irreversible attachment to



Irreversible attachment is accommodated by introducing separate colloidal species for Th, Pu, Am, and Cm isotopes. In the unsaturated zone, these "irreversible colloid" species are partially permanently filtered in matrix units, but unretarded and unattenuated through fractures (figure at left). Filtration factors were calculated offline by comparing estimated colloid size distributions with matrix pore size distributions. In the saturated zone flow path, which at Yucca Mountain passes first through fractured tuff, then an alluvial aquifer, the irreversible colloid species are subject to retardation (figure at left). We have initially adopted DOE-developed probability distributions for the retardation factors in matrix tuff (unsaturated and saturated) and alluvium (CRWMS M&O, 2000c), but will

For reversible attachment, we have adopted the approach of Contardi et al. (2001), after Vilks et al. (1998), in which the sorption coefficient (K_d) or retardation factor (R_d) for a dissolved radionuclide is reduced to account for equilibrium sorption to mobile colloids. During transport, it is assumed that solute sorbs to host rock and colloids according to the same K_d, though scaled for surface area. The K_d is thus adjusted by:

$$K_{d,eff} = \frac{K_d}{1+CFK_d}$$

where C is colloid concentration in water and F is a dimensionless factor accounting for surface area differences between matrix rock and colloids. In the saturated zone, TPA uses R_d rather than K_d:

$$R_{d,eff} = 1+\frac{(1-\phi)\rho K_d}{\phi\,(1+CFK_d)}$$

where ϕ is matrix porosity and ρ is bulk density. (K_d must be calculated from the starting R_d.)

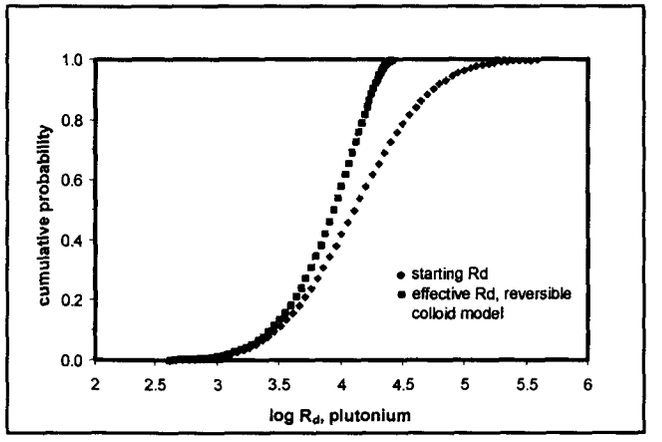
Many values for K_d and R_d are represented in the TPA input file as probability distributions. In order to adjust these values for reversible colloid attachment, it was necessary to discretize the starting distributions, apply the adjustment to each value, and then supply the tabulated colloid-adjusted distribution to the input file. An example of how a parameter distribution can be altered is shown in the plot to the right. It can be seen that, for this particular set of parameters, the adjustment is only significant at retardation factors greater than around $10^{3.5}$. At the high end of the distribution, R_d is lowered by a factor of 15.

The TPA abstraction does not directly implement all aspects of the detailed process-level models discussed in this poster. Rather, TPA implementation is designed to be flexible so that the results of process-level models can be tested within TPA by judicious choice of parameters. We will also perform TPA sensitivity analyses using bounding parameters in a more sophisticated attempt to answer the question: "How much could colloid-enhanced transport negatively influence dose?"

Summary

To prepare for reviewing colloid release and transport aspects of the license application for the potential Yucca Mountain repository, the NRC is performing simplified dose effect calculations, process-level modeling, and total-system performance assessment simulations. Results to date show that colloids have the potential to affect the performance of natural barriers in delaying radionuclide transport to the dose receptors. Models point to the importance of uncertain parameters controlling the rates of attachment of solutes to colloids and colloids to host rock. The TPA code has been modified to explicitly implement colloid effects, and future analyses will focus on the sensitivity of dose results to varying inputs reflecting different conceptual models and transport parameters.

This poster, documenting work performed by the CNWRA on behalf of the NRC Office of Nuclear Material Safety and Safeguards, Division of Waste Management, under Contract No. NRC-02-02-012, is an independent product of the CNWRA and does not necessarily reflect the views or regulatory position of the NRC. The NRC staff views expressed herein are preliminary and do not constitute a final judgment or determination of the matters addressed or of the acceptability of a license application for a geologic repository at Yucca Mountain.



Cumulative probability distribution for the log of the plutonium retardation factor for alluvium in TPA 5.0, demonstrating the effect of the reversible colloid model adjustment. Equation parameters are: ϕ = 0.125, ρ = 2500 kg/m^3, C = 0.001 kg/m^3, and F = 630.

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