

HLWYM HEmails

From: David Pickett
Sent: Wednesday, October 29, 2003 6:42 PM
To: Christianne Ridge
Subject: RE: Colloids in TPA 5.0o
Attachments: reversible colloids.wpd; colloidsummary.wpd; irreversible colloid release factor; FW: UZ filtration of colloids with irreversible attachment; migration 03 paper for NRC revised.doc; Kd's and Rd's; RE: new table for tpa.inp

Follow Up Flag: Follow up
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Christianne,

First, I've attached a lot of info. The four attached emails should be fairly quick reads and give you a good idea of implementation. "reversible colloids.wpd" shows how the Kd and Rd adjustment is made for reversible attachment. "colloidsummary.wpd" is Ron's more sketchy account. As for your questions:

> Does the parameter ColloidReleaseFactor represent the fraction of
> released waste that is in colloidal form?

Yes.

> parameter is currently set to a value of 0.1 for each of the species
> representing radionuclides irreversibly sorbed to colloids (ie, Ja,
> Jc, Jp, and Jt). What is the basis for this value?

There really is no strong basis for this value; it's a placeholder. The attached "irreversible colloid release factor" email shows how I came up with 0.1. My intention has been to look at sensitivity. Perhaps we can get some clues on this from the new TBDoc; see, for example, page 3-16, sec 3.2.7.3.

> Does the parameter PermanentLossColloidFilterFactor represent the
> fraction of Ja, Jc, Jp, and Jt that are permanently removed through
> filtration? What is the basis for the values for this parameter for
> the invert and various subsurface layers?

Yes, it is the fraction removed. See attached "FW: UZ filtration of colloids with irreversible attachment" email for the bases for the UZ values. Invert value is a placeholder, largely because the invert design has not been finalized (as far as I know). Again, I feel these values have very large uncertainties; these are starting points.

> My understanding is that radionuclides that are reversibly bound to
> colloids are modeled by reducing the Kd and Rd values for the
> dissolved species off line based on the colloid concentration. What
> are the colloid concentrations used for this adjustment?

As seen in "reversible colloids.wpd," I used 0.001 kg/m³ - equivalent to 1 mg/L or 1 ppm. As you can see in the TBDoc, pp 3-19 to 3-20, DOE's range now goes up to 200 ppm. So we can play with this value quite a bit.

David

Hearing Identifier: HLW_YuccaMountain_Hold_EX
Email Number: 656

Mail Envelope Properties (dpickett@cnwra.swri.edu20031029184200)

Subject: RE: Colloids in TPA 5.0o
Sent Date: 10/29/2003 6:42:10 PM
Received Date: 10/29/2003 6:42:00 PM
From: David Pickett

Created By: dpickett@cnwra.swri.edu

Recipients:
"Christianne Ridge" <Christianne.Ridge@nrc.gov>
Tracking Status: None

Post Office:

Files	Size	Date & Time
MESSAGE	2070	10/29/2003 6:42:00 PM
reversible colloids.wpd	70491	
colloidssummary.wpd	6858	
irreversible colloid release factor		4917
FW: UZ filtration of colloids with irreversible attachment		7172
migration 03 paper for NRC revised.doc		116801
Kd's and Rd's	82574	
RE: new table for tpa.inp	6874	

Options

Priority: Standard
Return Notification: No
Reply Requested: No
Sensitivity: Normal
Expiration Date:
Recipients Received: Follow up

Proposed Equations and Parameters for Reversible Colloid Attachment in TPA Transport Abstractions

David Pickett
January 21, 2003

The TPA approach to modeling reversible attachment involves an (optional) adjustment to transport parameters that account for the increased mobility due to reversible attachment. Values for K_D and R_D are re-calculated. Retardation in fractures is not modeled, consistent with the non-colloid case. **Note: none of these adjustments should be made to the new irreversible colloid species.**

1. Unsaturated Zone

The affected parameters in the UZ are the matrix K_D 's for the various tuff units (because there is no retardation in the fractures). The adjustment will be made to all radioelements that have a non-zero K_D , but it should be noted that the effect is smaller the smaller the starting K_D . The affected elements are Am, Pu, U, Np, Th, Ra, Pb, Cs, Ni, and Se. Curium should be included in this list if it is given a non-zero K_D ; the current zero value is highly conservative.

Alternatively, the adjustment could be made to all radioelements, because those that are assigned a zero value will remain zero. This would simplify later efforts if any elements are no longer assumed to have no sorption. In any case, the adjustment should be made after the K_D is sampled. The adjustment should be made to every unit, e.g., MatrixKD_TSw_Am, MatrixKD_ChnvAm, etc.

The adjustment is as follows:

$$K_{D,eff} = \frac{K_D}{1 + C_U F_T K_D}$$

(Contardi et al., 2001, after Vilks et al., 1998) where K_D is the given matrix K_D (m^3/kg) sampled in TPA, C_U is the colloid concentration in unsaturated zone water (kg/m^3), and F_T is a unitless factor accounting for surface area differences between tuff matrix and colloids (Contardi et al., 2001). C_U and F_T should be easily adjustable by the user; default values are:

$C_U = 0.001 \text{ kg/m}^3$ (bounds 78% of the concentration data compiled by Contardi et al., 2001)

$F_T = 590$ (typical value for tuff in Contardi et al., 2001).

FractureRD_... values for the UZ can remain unaltered, as long as they continue to be unity.

2. Saturated Zone

A. SZ Tuff

For transport in the SZ tuff, relevant parameters are the “immobile” phase retardation factors and the diffusion rate. (Fracture_RD_... values can remain unaltered as long as they continue to be unity.) These two parameters affect how solute is transferred from the mobile fracture phase to the “immobile” matrix. They are affected by reversible colloid attachment according to these equations (note: these are not the equations that will be used in TPA):

$$R_{D,eff} = 1 + \frac{(1 - \phi)\rho K_D}{\phi(1 + C_T F_T K_D)}$$

(Contardi et al., 2001, after Vilks et al., 1998) and

$$D_{eff} = \frac{D}{(1 + C_T F_T K_D)^2}$$

(modified from CRWMS M&O, 2000, after CRWMS M&O, 1997). ϕ is the tuff matrix porosity, equivalent to the input parameter ImmobilePorosity_STFF (currently a constant value of 0.2). ρ is the grain density of the tuff matrix; a value of 2650 kg/m³ is appropriate (Contardi et al., 2001). D is the diffusion coefficient, K_D is the matrix K_D (m³/kg), C_T is the colloid concentration in water in saturated tuff matrix (kg/m³), and F_T is a unitless factor accounting for surface area differences between matrix and colloids (Contardi et al., 2001). C_T and F_T should be easily adjustable by the user; default values are:

$C_T = 0.001$ kg/m³ (bounds 78% of the concentration data compiled by Contardi et al., 2001)

$F_T = 590$ (typical value for tuff in Contardi et al., 2001).

The retardation factors (e.g., ImmobileRD_STFF_Am) are element specific, and so could be adjusted for colloids with the above equation. However, a single diffusion coefficient, DiffusionRate_STFF = 0.001 m²/yr, is applied to all elements in TPA; adjustment of this parameter would be inappropriate because elements are affected to very different degrees by reversible attachment (see K_D term in D_{eff} equation). According to equations 10-1 and 10-2 of CNWRA (2002; TPA 4.0 Module Description and User's Guide), the retardation factor is inversely proportional to the rate of change of concentration in the immobile phase, while the diffusion coefficient is directly proportional. Therefore, the ratio of the diffusion coefficient to the retardation factor, both adjusted for reversible attachment, implements the colloidal effect on matrix diffusion. Because the diffusion coefficient cannot be adjusted (i.e., we cannot calculate D_{eff} for each element), the retardation factor $R_{D,eff}$ can be further adjusted so that the ratio is element specific. That is:

$$\frac{0.001}{R'_{D,eff}} = \frac{D_{eff}}{R_{D,eff}}$$

Using the equations above for D_{eff} and $R_{D,eff}$, knowing that $D = 0.001 \text{ m}^2/\text{yr}$, and rearranging gives:

$$R'_{D,eff} = (1 + C_T F_T K_D) \left[(1 + C_T F_T K_D) + \left(\frac{1 - \phi}{\phi} \right) \rho K_D \right]$$

This calculation should be applied to each “immobile” R_D value in the TPA input (e.g., ImmobileRD_STFF_Am) using an element-specific K_D (see below). The colloid adjustment can either be applied only to those radioelements with a non-unity value (Am, Np, Cm, U, Pu, Th, Ra, Pb, Cs, Ni, Se, and Nb) or to all radioelements (values that are unity will remain at unity, because the K_D calculated below will be zero). This formulation accounts for the competing effects of the two parameters on matrix diffusion.

The K_D value in the equation is not in the TPA input file; it must be extracted from the starting R_D (e.g., ImmobileRD_STFF_Am) in the input file, using this equation:

$$K_D = \frac{\phi}{(1 - \phi)\rho} (R_D - 1)$$

(Freeze and Cherry, 1979), with ϕ and ρ as defined above. $R_{D,eff}$ can now be calculated and used in place of the input file value for the “immobile” R_D for each element.

B. SZ Alluvium

In TPA, transport in the alluvial aquifer is simulated with an element-specific retardation factor, e.g., AlluviumMatrixRD_SAV_Am. The equation for the R_D that accounts for reversible attachment is:

$$R_{D,eff} = 1 + \frac{(1 - \phi)\rho K_D}{\phi(1 + C_A F_A K_D)}$$

(Contardi et al., 2001, after Vilks et al., 1998), where K_D is the matrix K_D (m^3/kg), C_A is the colloid concentration in water in saturated alluvium (kg/m^3), and F_A is a unitless factor accounting for surface area differences between alluvium and colloids (Contardi et al., 2001). C_A and F_A should be easily adjustable by the user; default values are:

$$C_A = 0.001 \text{ kg}/\text{m}^3 \text{ (in middle of range of concentration data compiled by Contardi et al., 2001)}$$

$F_A = 630$ (calculated for alluvium in Contardi et al., 2001).

ϕ is the alluvium matrix porosity; a value of 0.125, in the middle of the range for TPA parameter AlluviumMatrixPorosity_SAV, should be used to be consistent with Contardi et al. (2001). ρ is the grain density of the alluvium matrix; a value of 2500 kg/m³ should be used (Contardi et al., 2001).

The K_D value in the equation is not in the TPA input file; it must be extracted from the starting R_D (e.g., AlluviumMatrixRD_SAV_Am) in the input file, using this equation:

$$K_D = \frac{\phi}{(1 - \phi)\rho} (R_D - 1)$$

(Freeze and Cherry, 1979), with ϕ and ρ as defined in the previous paragraph (note that ϕ and ρ differ for the tuff and alluvial aquifers). $R_{D,eff}$ can now be calculated. This calculation can either be applied to only those radioelements with non-unity alluvium retardation factors (Am, Np, Cm, U, Pu, Th, Ra, Pb, Cs, Ni, Se, and Nb) or to all radioelements. Those with R_D equal to one will be unaffected by the calculation because calculated K_D will be zero. **Note: this adjustment should be made after the starting R_D has been sampled and/or calculated with D. Turner's method.**

Here is a summary of how the colloids are handled for TPA5.0d

1. The colloid release fraction is used from tpa.inp to create a new release output file from releaset to divide the releases into aqueous and colloidal releases. This only applies to element pairs (Cm, Jc), (Am, Ja), (Pu, Jp), and (Th, Jt).
2. The invert Rd and the permanent loss colloid invert filter factor for the individual colloids are used by ebsfilt to prepare a release from invert output file. The loss factor is applied to the output of ebsfilt.
3. The matrix Kds and fracture Rds for the colloidal elements are retrieved from the tpa.ip file while Kds and Rds for the non-colloidal form of Am, Np, Pu, Th, and U are calculated with Turner's coefficients.
4. The permanent loss colloid filter factor for each of the UZ layers are retrieved from the tpa.inp file. One loss factor for each layer is used for all colloidal elements in that layer.
5. The infiltration for the current subarea is organized into 500 year time steps.
6. The maximum infiltration over all time is used to set the media type for each layer. If the infiltration exceeds the conductivity the layer is omitted from the NEFMKS run as fracture flow.
7. A total composite effective filter factor is calculated based on the remaining matrix flow layers.
8. The original infiltration values with TPA time steps are reinstated.
9. Loop over all time steps with the following:
 - 9.1 Calculate the matrix fraction, up to 1.0, for each layer as $\text{cond}(\text{layer}) / \text{infiltration}(\text{time})$.
 - 9.2 Select the minimum matrix fraction over all layers for this time step.
 - 9.3 Calculate the fracture fraction as $1.0 - \text{matrix fraction}$.
 - 9.4 Divide the release array at each time step into matrix and fracture values for their respective arrays.
 - 9.5 Remove colloids from the matrix release array using the total composite effective filter factor before calling NEFMKS.
10. Calculate the flow velocity for each layer and each time step based on the infiltration, porosity and SATM or SATF Van Genuchten parameters.
11. Limit the travel time in each layer to a minimum of 5 years.

12. Calculate the travel time for each layer at each time step.
13. Calculate two average travel times for each layer, one for the compliance period and one for all time. Select the higher value as the average ground water travel time for a given layer.
14. Calculate the total average GWTT for the UZ as a sum of the individual layers.
15. Set the repository leg velocity for each time step to the value used for the first matrix leg.
16. Further select layers for analysis based on the average GWTT being greater than 10 years and also being greater than the total average GWTT / 10.0.
17. Retrieve dispersion factors for each layer from tpa.inp.
18. Set dispersion limit for each layer based on leg length. If leg length is greater than 40 meters then dispersion limit is 0.1 meters. Otherwise the dispersion limit is 0.01 meters.
19. Calculate the dispersion for each layer as the minimum of its dispersion limit and its $\text{leg_length} * \text{dispersion_factor}$.
20. Run NEFMKS and pass the output on to SZFT.
21. In SZFT colloids are not filtered by layer filter factors, but the following is applied; restrict the SAV Rd for Ja and Jp to be no more than 5 times the STFF Rd values for Ja and Jp.
22. Run NEFMKS and move the colloid releases to their non-colloidal form.

ASSESSMENT OF THE POTENTIAL EFFECTS OF COLLOIDAL RADIONUCLIDE TRANSPORT ON
NUCLEAR WASTE REPOSITORY PERFORMANCE

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Abstract

The U.S. Nuclear Regulatory Commission (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 potential high-level nuclear waste repository at Yucca Mountain, Nevada. For the evolving conditions of the potential 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 represented by the performance assessment being developed by the DOE. Preliminary simplified calculations providing a conservative estimate 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. Keywords: colloid, radionuclide, transport, nuclear, waste, plutonium.

1. Introduction

Yucca Mountain, Nevada, is being studied by the U.S. Department of Energy (DOE) as a potential site for disposal of high-level nuclear waste. One of the technical concerns being investigated by project scientists is whether radionuclide transport could be enhanced by colloids to a degree that an adverse dose consequence will result. Colloids have the potential to enhance the release and migration of highly sorptive and insoluble radioelements, such as plutonium (Pu), that might not otherwise be considered mobile. The U.S. Nuclear Regulatory Commission (NRC), with technical assistance from the Center for Nuclear Waste Regulatory Analyses (CNWRA), is reviewing site characterization studies and data at the potential repository site, in preparation for reviewing an anticipated license application from the DOE. In this review, the critical performance measure is the exposure dose via the groundwater pathway. In addition, the DOE will be required to describe the capabilities of engineered and natural barriers to delay transport of contaminants. Therefore, radionuclide

transport, in which colloids play a role, is a key technical issue requiring resolution. Our current focus involves developing independent capabilities to evaluate colloid-facilitated transport and review the DOE's implementation in its total system performance assessment (TSPA) model. Determining the potential risk (dose) 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.

Potential sources of colloids at Yucca Mountain include naturally-occurring groundwater colloids, products of corrosion of engineered barrier system components, vitrified waste forms and spent fuel, and materials introduced during construction. Colloids could play a significant role in transport of radionuclides due to their high specific surface area, which facilitates sorption or precipitation of dissolved radionuclides (Manaktala et. al., 1995). In addition, microbial biocolloids may need to be considered, depending on the evolving geochemical environments and migration pathways in the unsaturated and saturated zones at Yucca Mountain. Elements that can be significantly concentrated in colloidal form typically have low solubility and high particle reactivity; Pu and americium (Am) are two such elements (Wilson, 1988). It is notable, then, that in 1000 year-old spent nuclear fuel, about 45% of the activity inventory would be $^{239+240}\text{Pu}$ and 51% would be ^{241}Am (Wilson, 1988).

The NRC is independently evaluating lab, field, and modeling work of the DOE while developing alternative modeling capabilities to upgrade performance assessment simulations. This paper presents our approach to assessing the potential importance of colloidal radionuclide transport and identifying scientific aspects of colloidal transport for which a better understanding is needed.

2. DOE Model and Key Uncertainties

The DOE TSPA approach to colloidal release and transport is described in detail in CRWMS M&O (2000a) and references therein. The models consider three colloid types: groundwater, corrosion product, and waste form (glass only). Released waste form colloids containing irreversibly-attached radionuclides are transported and retarded in the unsaturated and saturated zones. Radionuclide release may be augmented by reversible attachment to each of the three colloid types; transport of reversibly-attached radionuclides is modeled by adjusting the radionuclide transport parameters (i.e., sorption and diffusion). The most recently published TSPA showed colloidal ^{239}Pu as one of the major contributors to dose after 50,000 years (CRWMS M&O, 2000a).

More recently, the DOE has considered changes to the model and parameters (Aguilar and Alcorn, 2003), but

the basic approach is unchanged. Key concepts and parameters that are uncertain include:

- concentrations of colloids of each type leaving the waste package,
- waste forms for which colloid release is implemented,
- the choice of radionuclides for which colloid effects should be modeled,
- degree and reversibility of radionuclide attachment to colloids,
- effects of the near-field environment,
- colloid concentrations along the groundwater transport path,
- model abstraction of physical and chemical filtration of colloids,
- applicability of parameters derived from synthetic microsphere tests, and
- dispersion and dilution effects on colloidal radionuclide concentrations.

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 represent 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.

3. Simple Calculation

One way to approach initially an uncertainty issue with respect to waste disposal is to ask the question, "How much could this process or feature negatively influence dose?" We present here a calculation, modified after Dam et al. (2001), that addresses this question in a simplified, preliminary manner. Because the potential effect of colloids is to counteract retardation, we compare the relative hypothetical dose contribution from colloidal Pu in one commercial spent fuel waste package to contributions from radioactive technetium (Tc) and iodine (I). Because they have low sorption coefficients, these latter two elements are often assumed in groundwater systems to be unretarded (e.g., Mohanty et al., 2002). This calculation is not an attempt to predict accurately the dose effect; rather, it provides a means for comparison. If the potential dose from colloidal Pu is comparable to that from mobile radionuclides ^{99}Tc and ^{129}I , then the importance of characterizing colloidal influence on radionuclide transport is clear. We calculate the magnitude of mobilized colloidal Pu from spent fuel in one waste package by

applying laboratory release rates to projected flow rates at Yucca Mountain. A key assumption is that colloidal Pu concentration in groundwater is not reduced until diluted in the representative volume; in addition, decay is neglected. The equation used is:

$$dose_{rate} = \frac{[Pu] \times FR}{PR} \times DCF \quad (1)$$

with the variables as defined in the following paragraphs describing parameter development.

Colloidal Pu concentration [Pu]. Stagnant water leaching of spent nuclear fuel in a laboratory study resulted in up to 1.1×10^7 Bq/m³ (300 pCi/ml) ²³⁹⁺²⁴⁰Pu in the leachate (Wilson, 1988). This Pu was not demonstrated to be due to waste form colloids (and, in fact, the leachate was filtered at 0.4 μm), but we assume it was for the purposes of the calculation. With the Pu activity being 70% ²⁴⁰Pu and 30% ²³⁹Pu (Wilson, 1988), this activity concentration converts to 1.0×10^{-8} M, which is lower than the maximum colloidal Pu concentration of 8×10^{-8} M in high-level waste glass tests (CRWMS M&O, 2000b). Because spent nuclear fuel dominates the projected inventory of the potential repository (about 90% by mass; CRWMS M&O, 2000a), neglecting glass waste is not significant for the present calculation.

Colloidal Pu may also be released by sorption to groundwater and corrosion-product colloids. We assume conservatively no desorption from colloids during transport. We calculate this colloidal Pu component using the corresponding DOE model and parameters (CRWMS M&O, 2000c), that is, as the product of the dissolved Pu concentration, the K_d for Pu onto colloids, and the colloid mass concentration. Because data on the specific nature of spent fuel waste form colloids are not available, we will calculate sorbed colloidal Pu only for groundwater and iron-(hydr)oxide corrosion product colloids (Table 1). The resulting concentrations are 7.2×10^{-8} kg/m³ (3.0×10^{-10} M) for groundwater colloids and 2.4×10^{-5} kg/m³ (1.0×10^{-7} M) for iron-(hydr)oxide colloids. Adding these to the spent fuel “colloidal” Pu from Wilson (1988) yields a total of 2.6×10^{-5} kg/m³ (1.1×10^{-7} M) colloidal Pu, dominated by the component sorbed to iron-(hydr)oxide colloids. Given the activity distribution of ²³⁹Pu versus ²⁴⁰Pu (Wilson, 1988), this concentration corresponds to 1.2×10^8 Bq/m³ (3300 pCi/ml). Note that this concentration is an order of magnitude lower than the maximum Pu solubility limit employed in TPA (Mohanty et al., 2002).

Water flow through waste package (FR). Recent results from TPA Version 4.1 yield a flow rate through breached waste packages of 6.7×10^{-3} m³/y (6.7 L/y), averaged over the time period during which flow takes place

(S. Mohanty, personal communication, 2003).

Pumping rate (PR). The “representative volume” of groundwater by which radionuclides are delivered to the potential dose recipients is fixed by regulation (10 CFR Part 63) at $3.7 \times 10^6 \text{ m}^3/\text{y}$ (3000 acre-feet/y). The annually released radioactivity is diluted into this volume.

Dose conversion factor (DCF). Dose conversion factors for ^{239}Pu and ^{240}Pu are both $1.02 \times 10^{-6} \text{ (Sv/y)/(Bq/m}^3\text{)}$ [$3.79 \times 10^6 \text{ (rem/y)/(Ci/m}^3\text{)}$] for the nonpluvial environment reflecting drinking and irrigating under current conditions (Mohanty et al., 2002).

Using Equation (1), the calculated dose rate from colloidal $^{239+240}\text{Pu}$ in one waste package is $2.2 \times 10^{-7} \text{ Sv/y}$ (0.022 mrem/y). Key simplifying assumptions in this calculation include: no colloid retardation or attenuation (due to physical filtration) in either natural or engineered barriers, the use of maximum or high cumulative probability values for concentrations and K_d to colloids, no desorption from colloids, and neglect of radionuclides other than ^{239}Pu and ^{240}Pu .

Potential doses from ^{99}Tc and ^{129}I assume a uniform waste form release rate of 10^{-4} y^{-1} ; that is, the entire inventory is depleted over a 10,000-year period. Model spent fuel waste package inventories are $4.2 \times 10^{12} \text{ Bq}$ (114 Ci) for ^{99}Tc and $1.0 \times 10^{10} \text{ Bq}$ (0.28 Ci) for ^{129}I (Mohanty et al., 2002); thus, release rates are $4.2 \times 10^8 \text{ Bq/y}$ (0.011 Ci/y) for ^{99}Tc and $1.0 \times 10^6 \text{ Bq/y}$ ($2.8 \times 10^{-5} \text{ Ci/y}$) for ^{129}I . In Equation (1), these release rates replace the term “[Pu] $\times FR$,” the same value for the representative volume (*PR*) is used, and dose conversion factors (*DCF*) are non-pluvial environment values from Mohanty et al. (2002). The resulting doses are $5.9 \times 10^{-8} \text{ Sv/y}$ (0.0059 mrem/y) for ^{99}Tc and $2.2 \times 10^{-8} \text{ Sv/y}$ (0.0022 mrem/y) for ^{129}I . Thus, the model dose effect of colloidal Pu is more than 2.5 times higher than combined ^{99}Tc and ^{129}I . These results are not meant to be predictive, and the absolute dose values are not significant; rather, the calculation illustrates the potential risk significance of colloidal transport of Pu and, by extension, other apparently immobile radioelements. We conclude that further evaluation is warranted to understand the processes affecting colloid release and transport. More realistic model analyses are underway at the process level and in performance assessment calculations.

4. Process-Level Models

In assessing processes potentially affecting waste isolation, process-level models are aiding the interpretation of laboratory and field data and in, the absence of sufficient data, better defining which parameters and processes

are likely to most strongly affect model results. One aspect of colloid-facilitated transport that has not been extensively investigated for Yucca Mountain is kinetic control of solute sorption to colloids. NRC-sponsored work by Cvetkovic et al. (2002) and Painter et al. (2002) developed an analytical colloid sorption model that implemented 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) used this sorption model as part of a more comprehensive simulation of colloid-enhanced radionuclide transport. Figure 1 shows colloid-solute-host interactions that were considered in the model, including solute sorption to colloids and host rock, reversible sorption of colloids to host rock, and irreversible filtration of colloids by host rock. The model simulated continuous Pu release into a heterogeneous aquifer, with tracking of steady-state tracer discharge downstream. Emphasis was on kinetic rates of the exchanges depicted in Figure 1, with simplifying assumptions in some cases (e.g., neglect of solute sorption to immobilized colloids). 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 need to be employed 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. Cvetkovic et al. (2003) found that 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.

5. Total-system Performance Assessment implementation

The NRC/CNWRA TPA model—which has been developed for the purpose of better understanding the DOE performance assessment and how the various features, events, and processes in the potential repository system affect its ability to prevent or minimize dose to the public—has not in the past included explicit implementation of colloid release and transport. Previously, potential colloid total-system effects were simulated by assuming no

retardation for selected radioelements (Contardi et al., 2001; Mohanty et al., 2002). 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, 2000c; Aguilar and Alcorn, 2003), but is implemented in a way that the user can vary the inputs to test parameters developed offline. Radionuclide release is enhanced by colloids, and both reversible and irreversible attachment to colloids are included during transport. The approach to the implementation is not to precisely reproduce or mimic process-level models, but to allow flexibility in adjusting parameters so that different aspects of the colloid conceptualization can be evaluated.

Irreversible attachment is accommodated by introducing separate colloidal species for Th, Pu, Am, and curium (Cm) isotopes. Release of these species from the waste form is user-specified. These “irreversible colloid” species are partially permanently filtered by the invert and are then transported to the unsaturated zone, where they are (i) partially permanently filtered when flow takes place through a tuff matrix unit or, more commonly, (ii) transported unretarded and unattenuated through fractures. Filtration factors were calculated offline by comparing estimated colloid size distributions with matrix pore size distributions. In the saturated zone, the irreversible colloid species are subject to retardation, with the same retardation factor applied to all four species. The saturated zone flow path at Yucca Mountain passes first through fractured tuff, then an alluvial aquifer. We have initially adopted DOE-developed probability distributions for the retardation factors in matrix tuff (unsaturated and saturated) and alluvium (CRWMS M&O, 2000d), but will subject them to sensitivity analyses.

For reversible attachment, we have adopted the approach outlined in 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 offline of TPA 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. For transport in unsaturated zone matrix tuff, the following adjustment is applied to all radioelements with non-zero K_d :

$$K_{d,eff} = \frac{K_d}{1 + CFK_d}, \quad (2)$$

where K_d is the matrix value, C is the colloid concentration in water, and F is a unitless factor accounting for surface area differences between matrix and colloids (typically equal to 590; Contardi et al., 2001). For saturated

zone matrix tuff, the adjustment of R_d for radioelements with non-unity values follows:

$$R_{d,eff} = 1 + \frac{(1 - \phi)\rho K_d}{\phi(1 + CFK_d)}, \quad (3)$$

where ϕ is the tuff matrix porosity, ρ is the grain density of the tuff matrix, K_d is the matrix value, C is the colloid concentration in water in saturated tuff matrix, and F is as defined for Equation (2). Because the TPA input file does not have values for K_d for the saturated zone, Equation (3) is combined with

$$K_d = \frac{\phi}{(1 - \phi)\rho} (R_d - 1) \quad (4)$$

(Freeze and Cherry, 1979) to yield

$$R_{d,eff} = 1 + \frac{R_d - 1}{1 + CF \left[\frac{\phi}{(1 - \phi)\rho} \right] (R_d - 1)}. \quad (5)$$

An analogous effective retardation factor equation is used for saturated zone alluvium. For saturated zone matrix tuff, the diffusion coefficient D is also adjusted, primarily so that diffusion of colloids from tuff fractures (where there is no retardation) to matrix is slowed. The equation is based on the DOE approach for reversible attachment (modified from CRWMS M&O, 2000e; Robinson et al., 1997):

$$D_{eff} = \frac{D}{(1 + CFK_d)^2}. \quad (6)$$

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 Figure 2. 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. Different choices of parameters will, of course, affect the results; for example, Contardi et al. (2001, Figure 1) show the effect of increasing colloid concentration in lowering the maximum retardation factor.

The TPA abstraction does not directly implement all aspects of the detailed process-level models discussed in Section 4. 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 in a more sophisticated attempt to answer the question posed in Section 2: “How much could this process or feature negatively influence dose?”

6. 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 suggest that colloids have the potential to affect how natural barriers delay 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 implement colloid effects, and future analyses will focus on the sensitivity of dose results to varying inputs reflecting different conceptual models and transport parameters.

Acknowledgments

We thank S. Mohanty, R. Janetzke, T. McCartin, and S. Painter for helpful discussions, and D. Turner, B. Sagar, and J. Bradbury for constructive reviews. This paper, documenting work performed by the CNWRA for 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. 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.

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Table 1. Parameters for calculating Pu sorbed to colloids

Colloid type	Dissolved Pu*	K _d for Pu on colloid†	Colloid concentration‡
Groundwater	$2.4 \times 10^{-4} \text{ kg/m}^3$	$10 \text{ m}^3/\text{kg}$	$3 \times 10^{-5} \text{ kg/m}^3$
Iron-(hydr)oxide	$2.4 \times 10^{-4} \text{ kg/m}^3$	$100 \text{ m}^3/\text{kg}$	$1 \times 10^{-3} \text{ kg/m}^3$

*Maximum from Mohanty et al. (2002) solubility limit distribution.

†Mean plus one standard deviation for lognormal distributions in CRWMS M&O (2000c). The groundwater value is similar to a surface complexation model result for colloids by Cvetkovic et al. (2003).

‡Maxima from CRWMS M&O (2000c).

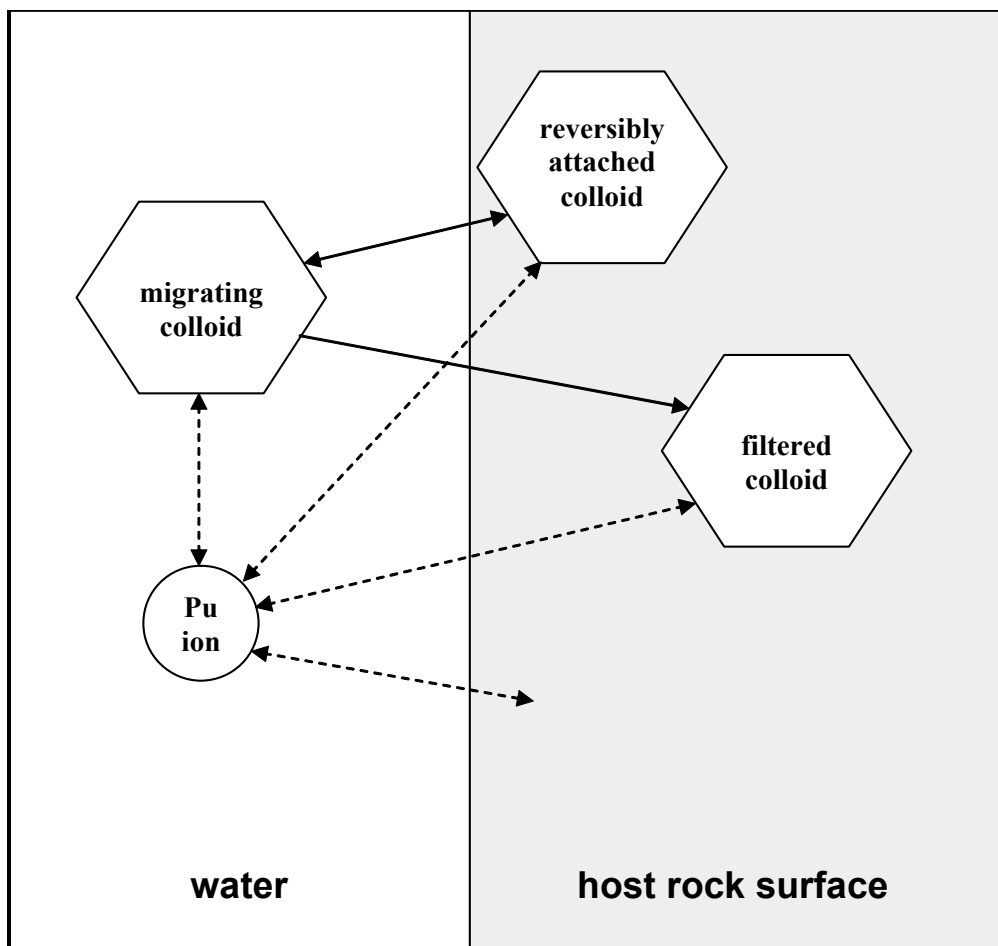


Figure 1. Schematic, after Cvetkovic et al. (2003), depicting solute-colloid interactions considered in process-level colloid-enhanced transport model. Solid lines indicate either exchange of colloids between water and reversible sites on host rock, or permanent removal by filtration. Dashed lines indicate reversible solute sorption onto colloids and host rock surfaces.

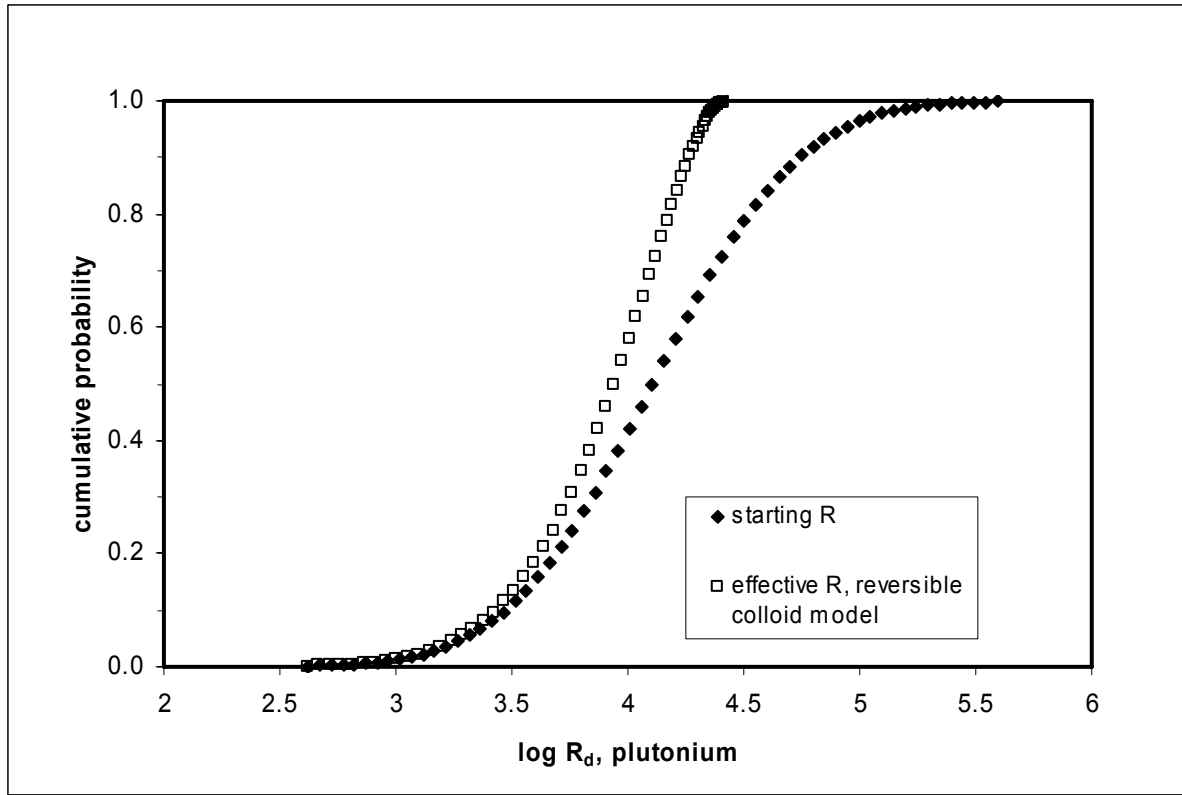


Figure 2. Cumulative probability distribution for the log of the plutonium retardation factor for alluvium in TPA 5.0, demonstrating the effect of the reversible colloid model adjustment. Equation (5) parameters are: $\phi = 0.125$, $\rho = 2500 \text{ kg/m}^3$, $C = 0.001 \text{ kg/m}^3$, and $F = 630$.