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Context

The U.S. Nuclear Regulatory Commission (NRC) is preparing to review a license application from the U.S. Department of Energy (DOE) for the potential high-level radioactive waste repository at Yucca Mountain, Nevada (Figure 1). NRC has developed an independent performance assessment model, Total-system Performance Assessment (TPA), used by NRC staff to gain risk insights into repository performance.

TPA simulates the entire repository system (Figure 2), from water infiltration at the surface to radionuclide exposure. In the portions of the code representing radionuclide release and transport (colored boxes in Figure 2), we have abstracted colloidal effects.

This poster describes the colloid-associated release and transport abstractions, which involve simplified approaches to modeling complex processes. A challenge was the need to fit the abstraction into the existing TPA computational framework. The abstractions do not explicitly include all possible features or processes, but are designed to allow flexibility in formulating analyses that may computationally incorporate or bound other processes. The key risk motivation for simulating colloid effects is the potential groundwater dose significance of plutonium.

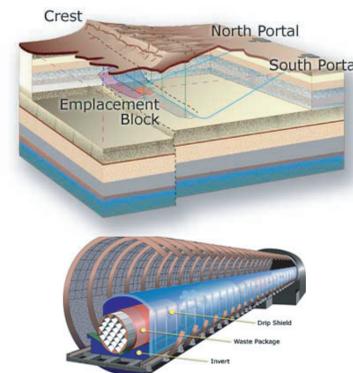


Figure 1. Features of the potential repository. http://www.ocrwm.doe.gov/ym_repository/about_project/concept/index.shtml

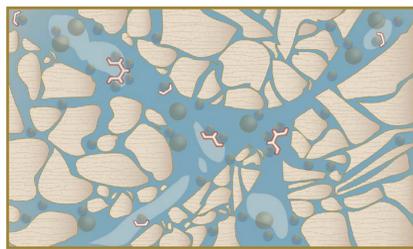


Figure 3. DOE illustration of conceptual model of colloid transport for use in performance assessment. Figure 3-60 from U.S. DOE (2001).

Key Features and Assumptions of the Colloid-Facilitated Release and Transport Abstractions

- Irreversible attachment to colloids is modeled during release, and irreversible and reversible attachment are modeled during transport (Figure 3).

- To capture uncertainty and variability, most parameters are sampled from probability distributions.

Release (EBSREL)

- As the waste form degrades, colloids resulting from corrosion of steel waste package components are available for irreversible attachment of Th-230, Pu-239, Pu-240, Am-241, Am-243, Cm-245, and Cm-246. Irreversible attachment to waste form colloids and intrinsic colloid association are not explicitly included, but may be studied within the framework.

- For radionuclides irreversibly attached to colloids, the total released concentration (including dissolved radionuclide) may exceed the solubility limit.

- Irreversible attachment is limited by the sorption capacity of the colloids and competition among actinides.

Transport (UZFT and SZFT)

- Radionuclides irreversibly attached to colloids are treated in UZFT and SZFT as separate species ("J-species") distinct from dissolved radionuclides.

- In the reference-case unsaturated zone irreversible model, colloidal J-species are retarded in volcanic tuff matrix, but unretarded in fractures. Some permanent filtration of J-species occurs at the interfaces of matrix units.

- In the saturated zone irreversible model, J-species are retarded during flow through fractured tuff and alluvium, but are not permanently removed by filtration.

- In the reversible model, equilibrium is assumed among radionuclides dissolved, sorbed to the host rock, and reversibly sorbed to colloids (Figure 4).

- The reversible model also accounts for reversible sorption of colloids to host rock (Figure 4). The combined effects of reversible attachment of radionuclides to colloids and colloid retardation in host rock are abstracted by modifying the retardation factor for the radionuclide. Highly sorptive elements (e.g., plutonium) are more strongly affected by reversible attachment than weakly sorbing elements.

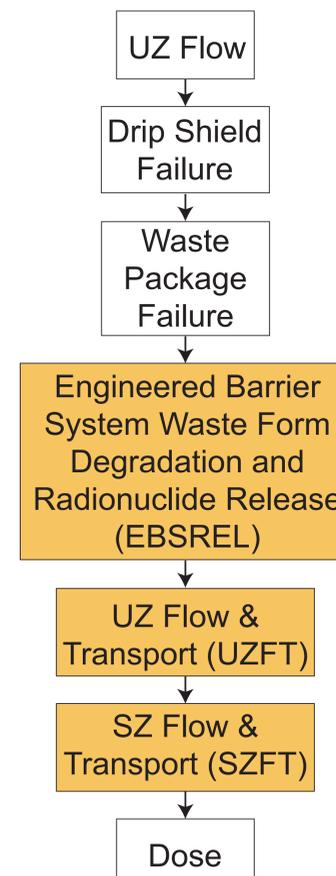


Figure 2. Schematic of information flow within TPA in the absence of disruptive events. Colloid abstractions are in the modules in the colored boxes.

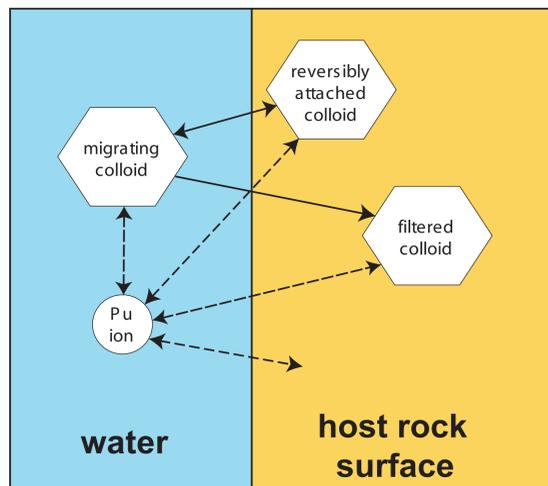


Figure 4. Schematic, after Cvetkovic, et al. (2004), depicting solute-colloid interactions considered in TPA colloid-associated transport abstractions. Solid lines indicate either exchange of colloids between water and reversible sites on host rock or permanent removal of colloids by filtration. Dashed lines indicate reversible solute sorption onto colloids and host rock surfaces.

Release Abstraction

The J-species (irreversibly attached radionuclides) are allocated from the radionuclide mass released during waste form degradation. Prior to this allocation, the sampled solubility limits for colloidal elements are increased so that the dissolved fraction (remaining after mass is transferred to the J-species) may be released at the solubility limit. In this way, if release is solubility limited, irreversible colloid attachment does not limit the dissolved radionuclide concentration to less than the solubility limit.

The allocation to J-species is computed assuming competition for colloid surface sites among plutonium, thorium, americium, curium, and uranium (although uranium is not then tracked as a J-species). A sorption capacity S_x is defined per unit water volume, which assumes a monolayer of sites on spherical, iron oxyhydroxide colloids and is the product of colloid concentration, colloid specific surface area, and the number of sorption sites per unit area.

A term Y is defined for each element to represent relative affinity for the sorption sites among the modeled actinides. The concentration of irreversible J-species follows this equation, using plutonium for illustration:

$$C_{JPu} = S_x \frac{Y_{Pu} \text{Min}(C_{Pu}, C_{S,Pu})}{\sum Y_i \text{Min}(C_i, C_{S,i})}$$

where

- C_{JPu} = total plutonium J species concentration (kg/m³)
- S_x = colloid sorption capacity (kg/m³)
- Y_{Pu} = plutonium affinity factor (dimensionless)
- C_{Pu} = total plutonium released concentration (kg/m³)
- $C_{S,Pu}$ = plutonium solubility limit (kg/m³)
- i = index for actinides in competition model

and the denominator is summed over plutonium, americium, thorium, curium, and uranium. Competition from other elements is neglected and would result in lower J-species concentrations.

Checks are performed to ensure mass balance is maintained (i.e., the maximum release is limited to the inventory in the dissolved waste form) and element mass is distributed to isotopes. The mass not allocated to J-species by the above equation remains in the dissolved state. Colloidal and dissolved species are passed from EBSREL to UZFT (Figure 2).

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Transport Abstraction

In the unsaturated zone (UZ) and saturated zone (SZ) radionuclide transport abstractions, the J-species (radionuclides irreversibly attached to colloids) are subject to retardation in the same way as dissolved species. The retardation factor probability distributions are based on theory and Yucca Mountain area test data; the same retardation factors are applied to all J-species radionuclides. In addition, permanent filtration at interfaces of tuff matrix units in the UZ is implemented by applying a reduction factor that is based on colloid size distributions and matrix pore size distributions.

Adjustment of retardation factors to account for reversible attachment to colloids is applied to all elements with the following equation:

$$R_D^{eff} = \frac{R_D + (K_D R_c)}{(1 + K_D)} = R_D \frac{1 + K_D(R_c/R_D)}{(1 + K_D)}$$

where

- R_D^{eff} = effective retardation factor accounting for colloids
- R_D = retardation factor for the element in the absence of colloids
- K_D = dimensionless distribution coefficient for sorption onto colloids
- R_c = retardation factor for colloids

It can be seen that, if an element sorbs more strongly to the host rock than do colloids (i.e., $R_D > R_c$), then $R_D^{eff} < R_D$, and element transport is enhanced by colloid association.

K_D is calculated by

$$K_D = C_c F K_o$$

where

- C_c = colloid concentration in water (kg/m³)
- F = ratio of the colloid specific surface area to the rock matrix specific surface area, about 600 (Contardi, et al., 2001)
- K_o = element distribution coefficient (m³/kg), generated by TPA and consistent with R_D

While equilibrium sorption is assumed in the abstractions, kinetic models were developed (e.g., Painter, et al., 2002; Cvetkovic, et al., 2004) to help guide parameter selection and aid in understanding how parameters and assumptions affect the results.

The SZFT model includes terms to account for matrix diffusion in fractured tuffs. A key parameter in this calculation is the mass transfer coefficient β (1/yr), which governs the rate of transfer of dissolved species between tuff matrix and fractures (Olague, et al., 1991). Because colloids will not be transported from fractures to matrix as readily as dissolved species, elements that sorb to them will also tend to remain in the fractures, enhancing their transport. This effect is accounted for by lowering the mass transfer coefficient:

$$\beta^{eff} = \beta / (1 + K_D)$$

Summary

- Simplified abstractions of colloid-facilitated radionuclide release and transport have been incorporated into the TPA performance assessment computational framework.
- The abstractions implement release and transport, as colloids, of selected actinides (most notably, plutonium) irreversibly attached to iron corrosion product colloids.
- Transport of all radionuclides is affected, to a lesser or greater degree, by reversible attachment to ambient groundwater colloids.
- The abstractions do not include all potential colloidal features and processes (e.g., waste form colloids, sorption kinetics), but are flexible enough to allow analyses that will improve NRC staff's understanding of colloidal effects in the potential repository system.

References

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