Reactive Transport Model for Fracture and Matrix Geochemistry at Yucca Mountain, Nevada

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Abstract

Reactive transport models for the potential nuclear waste repository at Yucca Mountain (YM) provide information on evolving water chemistries and secondary mineralogies, which may affect engineered barrier system performance, radionuclide releases, and radionuclide transport. Although reactive transport models permit explicit analysis of coupled thermal-hydrological-chemical processes important to estimation of long-term repository performance, these estimates have uncertainty that is difficult to quantify. Confidence can be gained in reactive transport models by demonstrating their capability to represent natural conditions.

Site characterization studies at YM have revealed significant differences between the hydrogeochemical properties of fracture and matrix materials in the unsaturated zone (UZ) overlying the potential waste emplacement setting. A quantitative evaluation of the most significant hydrogeochemical processes that caused these differences is helpful to develop detailed estimates of the quantity and chemistry of water contacting engineered materials in a thermally-perturbed repository setting—a risk-significant component of performance for YM. Developing a reliable explanation for observed differences between UZ matrix and fracture materials at YM provides a good test for the reactive transport models that may be used to evaluate performance of a potential repository at YM.

We developed a 1D, dual continuum, reactive transport model of the ambient UZ matrix/fracture system at YM in order to evaluate the origin and evolution of groundwater compositions and secondary minerals in fracture and matrix materials overlying the location of the potential repository. Sensitivity tests were conducted to gauge the importance of data and model uncertainties.

Basic Model Properties

Groundwater flow: Rock matrix and fracture networks in the unsaturated zone (UZ) are depicted as interacting porous continua. Heat/water flow between continua are controlled by:

* Darcy's law coupled with constitutive relationships and equations

* van Genuchten function with Mualem assumption for moisture retention/relative permeability

* the active fracture model (Liu et al., 1998)

* parameter values adopted from CRWMS M&O (2001).

Code: MULTIFLO version 1.5.2 (Litchner and Seth, 1996; Painter et al., 2001)

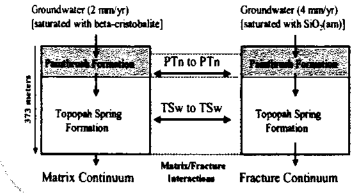


Figure 1. General structure of our dual permeability model, showing infiltration of SiO_2(am)-undersaturated waters into matrix and SiO_2(am)-(super)saturated waters into the fractures of the ptn26 unit.

Gridding: a 56 cell structured grid with a mixed upper boundary condition (i.e. specified gas pressure, temperature and liquid flux) and a gravity drainage lower boundary condition.

Geochemical Model: The model considers:

*dissolved species and gas: Cl^-, Ca^{2+}, H^+, HCO_3-, CO_2(aq), CO_3^{2-}, SiO_2(aq), $HSiO_3^-$, $Al(OH)_4^-$, Na^+, K^+, OH^-, and Al^{3+}, and CO_2(g)

* kinetically reactive phases: low albite, calcite, rhyolitic glass, amorphous silica, and endmember Na, Ca, and K-smectites

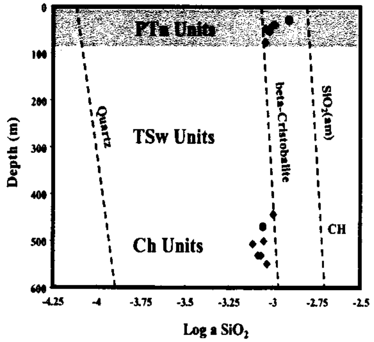
Hydrostratigraphy: Ten different hydrostratigraphic units bounded by the ptn26 and tsw38 layers (CRWMS, 2000). Each model unit is:

* horizontally-oriented with isotropic hydraulic properties

- defined by chemically and hydraulically distinct matrix and fracture continua

* occupied by different volumetric proportions of kinetically reactive phases.

What is the Origin of SiO_2(am) in the TSw Fractures?



Revised analytical pore water compositions (Browning et al., 2001) from SD-9 and other borehole cores indicate that matrix waters from Yucca Mt. are undersaturated in SiO_2(am).

There are no measured fracture water compositions from Yucca Mt., but the occurrence of SiO_2(am) in the TSw fractures indicates that fracture waters are (super)saturated in SiO_2(am).

Figure 2. Revised analytical pore water compositions from the SD-9 borehole (Browning et al., 2001); dashed lines show solubility curves for quartz, beta cristobalite, and SiO_2(am) with depth in the PTn, TSw and CH units.

High SiO_2 concentrations and precipitation of SiO_2(am) in Yucca Mt. Fractures may be explained by:

1) air flow and evaporation of water in fractures,
   ...but, DOE mountain scale thermal hydrological models indicate no significant air flow in TSw fractures (CRWMS, 2001).

2) SiO_2-rich waters infiltrating into fractures from the overlying alluvium or colluvium (Meijer, 2002),
   ...but, the geochemical consequences of high-silica infiltration waters have not been considered in the context of dual permeability flow and transport models.

Model Results

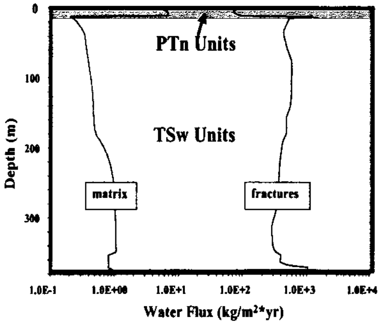


Figure 3. Simulated flux of water in and between the matrix and fracture continuum with depth in the PTn and TSw units of Yucca Mt., NV, where the depth at the top of the ptn 26 is defined as zero.

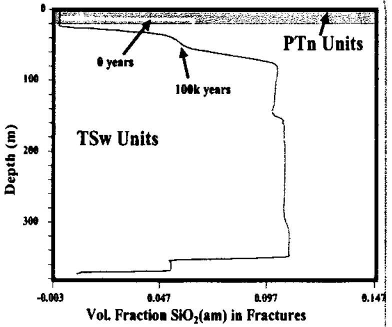


Figure 4. Comparison between initial volume fractions of SiO_2(am) in the fractures with depth and those predicted to occur after 100k years, where the depth at the top of the ptn 26 is defined as zero.

Water flows slowly from the fractures into the matrix, except in portions of the ptn26 and tsw37 units.

Simulated fracture-matrix interaction is most significant at the base of the PTn, where a permeability barrier may force large amounts of PTn matrix waters to pour into TSw fractures.

Simulations show SiO_2(am) dissolution in the TSw fractures, because fracture water compositions in the TSw are largely inherited from the overlying matrix PTn units, and the model neglects evaporation.

Conclusions/Discussion

Reactive transport simulations of fracture and matrix geochemistry at Yucca Mt. indicate that:

- The chemical composition of fracture waters percolating into TSw units may be very similar to the composition of measured pore water compositions in the overlying PTn units.

  Groundwater entering the TSw fractures is undersaturated with respect to SiO_2(am) and will dissolve, rather than precipitate, SiO_2(am).

  Observations of SiO_2(am) in TSw fractures cannot be explained by infiltration of SiO_2(aq)-rich waters into the PTn units from the overlying alluvium or colluvium.

- Flow and transport across matrix-fracture interfaces plays a significant role in controlling fracture water compositions.

  Further evaluation of assumptions inherent within the active fracture model (Liu et al., 1998) is needed to determine their effects on geochemical modeling results.

  Hydrologic processes that redistribute water in matrix and fracture materials (i.e. advection, vapor pressure lowering, and especially evaporation) may be required to explain SiO_2(am) and other secondary minerals observed in TSw fractures.

References: Browning et al. Scientific Basis for Nuclear Waste Management XXIII. R. Smith and D. Shoesmith, eds. Symposium Proceedings 663, 2000. Browning et al. Computers & Geosciences, vol. 23, number 3, 247-265, 2003; Browning and Murphy (2002) CNWRA Letter Report; TDR-NBS-HS-000002 REV 00 ICN 02. CRWMS M&O, 2000. TDR-NBS-HS-000002 REV 00 ICN 02. CRWMS M&O, 2001. ANL-EBS-MD-000049, Revision 00, ICN02, North Las Vegas, Nevada.; P.C. Lichtner and M. Seth, In Proc. Intl. Conf. on Deep Geo. Disposal of Radio. Waste, Can. Nuclear Soc., 1996 p 133-142. Liu et al. Water Resources Research 34(10), pp. 2633-2646, 1998; Meijer, Applied Geochem. 17. 793-805. Painter et al. CNWRA, 2001.

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