

Water Migration in Engineered Barrier Materials for Radioactive Waste Disposal

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INTRODUCTION

The US Department of Energy is evaluating various strategies for deep geological repositories for the permanent disposition of spent nuclear fuel. The development and evaluation of engineered barrier system design concepts and their potential interactions with natural barriers or with other engineered interfaces are inherently important to the long-term (i.e., tens of thousands of years) safety and performance of geological repositories.^[1]

Current understanding of barrier behavior and water migration is dependent on simulation modeling. Codes such as TOUGH2^[2,3] are used to characterize behavior of natural and engineered barriers. These codes have been used to simulate the observed progress of hydration phenomena through various media in the laboratory.^[4] There are, however, fundamental limitations to these simulations.

Both natural and engineered barriers are needed to limit the flow and movement of water or adsorb contaminants by chemical means. Media selection for barriers includes the use of materials that have very low permeability. However, the complete elimination of water movement through even low permeability material is not viewed as practical. Rather, what is needed for predictive modeling is a full understanding of the mineralogical behavior and pore structures of the material under saturation conditions and the transport characteristics of water through potential engineered barrier materials.

Neutron imaging and neutron scattering have been used to understand the transport of fluids through dense solids. For instance, the spontaneous imbibition of water into fine-grained granite matrices was studied using dynamic neutron radiography,^[5] showing that significant transport can occur in a material with a porosity as low as ~0.6%. In the work reported here, neutron imaging was coupled with ultra (U) small angle neutron scattering (SANS) measurements to qualitatively and quantitatively evaluate the multiscale structure of the media and the movement of water through it.^[6] Vibrational spectra of bonded hydroxyl groups and water molecules were also measured using inelastic neutron scattering.^[2] These techniques were used to answer fundamental questions associated with ground water transport necessary to evaluate the effectiveness of natural

and engineered barriers to safely contain spent fuel/waste over geologic time scales.

RESULTS

The experiments simulated wetting–drying–rewetting cycles to ascertain if changes in mineral hydration are temporary or permanent. Bentonite used in these experiments has undergone extensive characterization, and similar material has been used in hydrothermal experiments to provide the geochemical/mineralogical data used in transport models performed with the TOUGH2 computer code. Hydrothermal alterations included contact with deionized water, as well as saturated NaCl and saturated KCl solutions.

Initial work at Oak Ridge National Laboratory provided images of water saturation/swelling impact on small cores of bentonite with different sized aggregates packed to various bulk densities (Fig. 1). Transient water uptake data were also recorded for samples that had undergone different hydrothermal treatments (Fig. 1[A]).

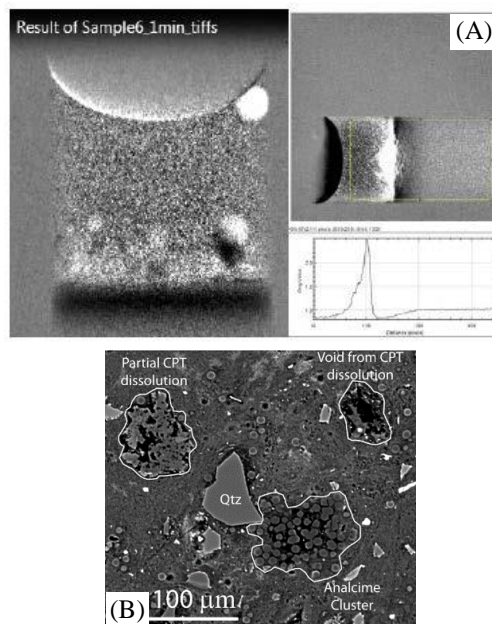


Fig. 1. (A) Neutron image of water saturation, swelling, and air expulsion in bentonite (12.7-cm-dia.). (B) Electron microprobe image showing the zeolite alteration/dissolution

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from the hydrothermal experiment developing new porosities (Image reprinted with permission from the *Mineralogical Society of America*).^[1]

Additional studies of the interaction of clays with water have also been carried out with focus on the (i) changes in mineralogy;^[1] (ii) cation absorption and interlayer interactions;^[7] and (iii) hydrogen bonding between the clay and adsorbed water.^[8] For inelastic neutron scattering studies, the bentonite samples were first equilibrated under various relative humidities at room temperature. Vibrational bands of H₂O in contact with the montmorillonite were observed at frequencies from 5 to 600 meV (up to 800 cm⁻¹). The intensities of the bands were proportional to the amount of water absorbed into the sample, determined by thermogravimetric analysis. Figure 2 shows vibrational spectra of H₂O collected on a bentonite sample over a temperature range of 5–300 K. A clear transition can be observed between 250 and 300 K, indicating a change in structure of the water in the sample. Differences were also observed in the spectra based on the hydrothermal treatment of the samples. The spectra will be analyzed using density functional theory to determine the binding energies of the water to the mineral surfaces. These findings will allow modeling of the hydration and dehydration of minerals and the effect of ion exchange on water transport through the bentonite matrix.

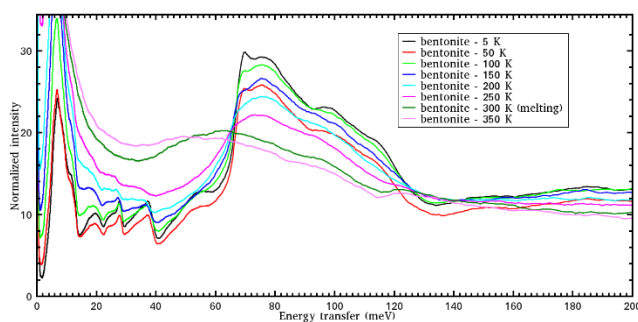


Fig. 2. Inelastic neutron scattering from bentonite samples from 0 to 100 meV, showing vibrational spectra for water absorbed in the sample matrix. These spectra, taken at temperatures from 5 to 300 K, show evidence of a phase transition between 250 and 300 K.

CONCLUSIONS

Results from SANS, imaging, and inelastic scattering experiments can be used to develop models for the interaction of water with mineral surfaces in engineered barriers. Future work will probe the effect of density and grain size, as well as changes in fluid composition. In addition, the experiments conducted to date were performed under ambient and static conditions, but water uptake needs to be evaluated under conditions conducive to fracture formation and elevated temperature and pressure. This will allow the swelling behavior and self-sealing characteristics

of the clays to be analyzed under more realistic environmental conditions that can be used to refine modeling parameters for evaluating the effectiveness of natural and engineered barriers. Results from these experiments will contribute to the development of well-calibrated models that can be extended beyond laboratory scales, which is essential for the proper selection and evaluation of barriers to radionuclide transport.

ENDNOTES

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