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A. Jenni, U. Mäder

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Coupling of chemical and hydromechanical properties in bentonite

A. Jenni^{*}, U. Mäder

RWI, Institute of Geological Sciences, University of Bern, Baltzerstrasse 3, CH-3012 Bern, Switzerland
(andreas.jenni@geo.unibe.ch, urs.maeder@geo.unibe.ch), +41 31 631 4531

^{*} corresponding author

Abstract

Bentonite plays a key role as buffer/backfill in many engineered barrier concepts for nuclear waste repositories and is required to perform over long periods of time. Swelling pressure and porosity distribution, two of the crucial properties for the performance of bentonite, vary with the chemical environment. This contribution reviews available data, explores a simple coupling model, and applies it to a fresh water / saline water scenario.

Systematic swelling pressure data for bentonite or montmorillonite under controlled chemical conditions are scarce. Evaluation of literature data shows that swelling pressure drops by 75% when increasing ionic strength of the equilibrated NaCl electrolyte from 0 to 3 M for an effective montmorillonite dry density corresponding to 1500 kg/m³ (MX-80 bentonite). In contrast, swelling pressure of CaCl₂-equilibrated montmorillonite drops only by 40% for the same ionic strength increase. Cation occupancy strongly affects swelling pressure at densities below 1500 kg/m³ (monovalent vs. divalent), and ionic strength still controls swelling pressure at higher density.

The chemical influence on the pore size/shape distribution in confined constant-volume systems is poorly investigated. The presence and quantity of a “free” porosity containing charge-balanced electrolyte (not affected by charged clay surfaces) in highly compacted clay systems is debated. Experimental data states different average montmorillonite interlayer distances at different ionic strengths at constant dry density. Assuming a constant volume for solids and electrolyte, any change in interlayer volume must be compensated by a corresponding change in some other type of porosity, most likely in a larger-size porosity domain. This coupling of ionic strength to porosity can explain differences in advective and diffusive transport commonly observed in clays tested with different background electrolyte concentrations, but at the same density.

The existence of any effect of cation occupancy on porosity distribution has not been confirmed so far. Observed differences in advective and diffusive solute transport between Na and Ca montmorillonite might be caused by different microstructures originating from sample preparation. However, our own experimental approach avoiding such artefacts also results in transport properties dependent on the exchanger cation.

Own and critically reviewed data from literature indicate a coupling of ionic strength or cation occupancy with porosity distribution. The influence of ionic strength is stronger and supported by experimental data. This HMC coupling (hydromechanical-chemical) was incorporated into a dual-porosity multicomponent reactive transport code. The code maps the complex porosity distribution into two domains: the *Donnan porosity* contains the porewater influenced by the negatively charged clay sheet surfaces, and has to compensate for this permanent charge with cations (representing the exchangeable cations of the clay). The Donnan porosity comprises clay interlayers and a region near clay particle outer surfaces (diffuse double layer). The Donnan porewater composition is at equilibrium with the charge-balanced “free porewater” not affected by surface charge. Both, the interlayer distance and the thickness of the diffuse double layer are assumed to depend linearly on the Debye length (dependent on ionic strength).

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