

Mineral-solution interfacial phenomena influences on porewater over-pressures in clay-rocks

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Long-term measurements (up to ~ 6 years) of pressures in sealed-off borehole intervals located at depths between roughly 420 and 530 m in the 130 m thick Callovo-Oxfordien (C-O) formation at the Andra URL (Bure, eastern France) show trends towards apparently stable values exceeding expected groundwater heads by several tens of meters. These observations need to be explained in order to properly model water and solute transport for feasibility studies of a potential underground storage facility for radioactive waste.

Anomalous over- (and under) pressures in a variety of geological contexts are reported in the literature, and osmotic phenomena-based interpretations have been proposed by a significant number of authors. Virtually all of the theoretical models proposed consider that the clay-rock acts as a semi-permeable membrane separating the solution compartment having the anomalous pressure from a solution having a reference composition and pressure. In all cases, corresponding water activities are calculated based on dissolved species concentrations, even when the model considers the clay-rock pore-space and pore solution as one of the 'compartments'. Interpretation of the observed C-O over-pressure data using such type models leads to the conclusion that the clay-rock must be a highly efficient semi-permeable membrane, blocking most solute movement while letting water molecules pass. This result is in contradiction with known solute diffusion characteristics for this rock measured under realistic porewater compositions.

The conceptual model presented in this communication resolves this inconsistency, while retaining osmosis as the underlying phenomena for over-pressure generation. The basic difference is consideration of other processes affecting, and fixing, water activity in a clay-rock volume, in particular interfacial phenomena (effects of exchangeable cations, water structuring...) and mineral solubility controls. Taking these additional phenomena into account results in a local water activity which is principally determined in the C-O clay-rock by the physical-chemical characteristics of the wetted mineral surfaces. The predicted over-pressure therefore varies mostly as a function of formation mineralogy and is relatively insensitive to variations in uncontrolled solute concentrations. The analogy can be extended to swelling pressures in bentonite-based engineered barrier materials.

Reactive transport models for deep radioactive waste disposal

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The conceptualisation of deep radioactive waste disposal envisages a multi-barrier design, with an engineered barrier system surrounding the nuclear waste (near field) in the geologic medium (far field). Most repository concepts take into consideration crystalline rocks or claystones as geologic medium due to their very low permeability, enhanced by the multi-barrier system (waste – steel and/or copper canister - compacted bentonite) surrounding the nuclear waste.

Although it seems these systems will maintain waste containment for long time periods, this is something that must be demonstrated. However, experiments can not reproduce the time span expected for the operational life of a repository. Evenmore, most experiments can not reproduce real repository conditions due to the low permeability of materials to be used (i.e. bentonite), slowing down the expected water-rock reactions. Therefore, the use of reactive-transport models became a very valuable tool to assess the long-term behaviour of deep nuclear waste repositories, as well as for individual parts of these systems.

In the present study we developed a numerical model to assess the geochemical evolution of the near field (bentonite barrier) of a repository based on the Swedish design performed by SKB. We assume the presence of a hypothetical fracture that is intersected by the deposition hole, where the canister containing the nuclear waste is emplaced. Thus groundwater flowing through the fracture interact with the bentonite barrier. The geochemical processes considered in the model are: dissolution/precipitation of bentonite accessory minerals, and smectite surface related reactions as cation exchange and protonation/deprotonation.

The results of the numerical simulations indicate that minor changes in pH occur in the bentonite pore water, increasing from 6.9 to 7.3. Accessory minerals play a decisive role in controlling the pH conditions, although the influence of cation exchange reactions and surface acidity cannot be underestimated. Cation exchange onto smectite surfaces results in a progressive depletion of calcium in the pore water, which is compensated by dissolution of Ca-bearing minerals (calcite and gypsum). The availability of these minerals is responsible of pH changes through time. Pore water is undersaturated in gypsum so that this mineral dissolves rapidly providing calcium for the exchange. Calcite dissolution takes place when gypsum is depleted in the system, and then, pH increases progressively.