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Laboratory-scale reactivity of a crystalline host rock for geological disposal of radioactive waste. Fracture vs. matrix.

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The Finnish geological repository for spent nuclear fuel is being constructed at the ONKALO® underground facility in Southwestern Finland. The bedrock is composed of migmatitic gneisses including brittle fault zones where groundwater flows. A relevant process to consider is the dissolution of chlorite, which releases Fe(II) that can lead to FeS precipitation and regulate HS^- concentrations in groundwater. Elevated HS^- is a concern for the potential corrosion of the copper canisters containing the spent fuel in the repository.

Two powder chlorite samples from fractures at this site were provided by Posiva Oy and characterized for their composition, solubilities and dissolution rates. XRD analysis showed the presence of both chlorite and muscovite. Compositions were measured by electron probe microanalysis. The chlorites in the two samples were slightly different; muscovite showed a single composition.

Chlorite 1: $(Mg_{1.444}Fe^{2+}{}_{1.958}Na_{0.018}K_{0.499})(Si_{3.246}Al_{2.565}O_{10})(OH)_8$

Chlorite 2: $(Mg_{1.973}Fe^{2+}{}_{2.515}Na_{0.007}K_{0.006})(Si_{2.733}Al_{2.693}O_{10})(OH)_8$

Muscovite: $(K_{0.797}Na_{0.020})(Al_{2.479}Mg_{0.240}Fe^{2+}{}_{0.179})Si_{3.227}O_{10}(OH)_2$

Apparent solubilities were consistent with those reported in existing thermodynamic databases. Far-from-equilibrium steady-state dissolution rates at near-neutral pH ranged between 10^{-13} and 10^{-12} mol/m^2/s, consistent also with previous results in the literature.

Two infiltration experiments using a gneiss core with a single fracture were performed to address the reactivity of this rock. The core was 5 cm in diameter and 6.2 cm in length. Groundwater from the site and Milli-Q® water were injected in the two experiments, respectively. The results (outlet solution chemistry), including the dissolution of chlorite, were interpreted by 1D and 2D reactive transport modeling (CrunchFlow). The 1D model considered flow, solute transport and reaction only along the fracture. Very large mineral surface areas, much larger than the exposed areas on the fracture surfaces, were needed to reproduce the experimental results. An alternative 2D model was developed, which also included diffusive transport and reactions in the rock matrix. The 2D model did not need the large surface areas in the fracture to match the experimental results. These results show the importance of the rock matrix in the overall reactivity of the fractured rock, despite the small porosities (ca. 1%) and diffusion coefficients. However, the 1D approach could still prove useful for large repository-scale calculations, given appropriate calibration.