Alteration at bentonite-cement interfaces – An experimental approach

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The identification of alteration processes at the interface of clay and contacting cement is crucial for ensuring the long-term stability of underground nuclear waste repositories, i.e. the Aspö field test site (Sweden).

In order to investigate alteration features directly at the bentonite-cement interfaces, three flow-through laboratory experiments were realized. Differences in the water uptake behavior and related changes in the mineralogy and chemistry of Portland cement clinker and air-dried MX-80 bentonite were monitored using wet-cell X-ray diffraction (XRD) and transmission electron microscopy analyses, covering an experimental period of 1 year.

The water uptake rate was ~44-times higher in the clay-cement experiments than that of the pure MX-80 bentonite reference experiment, and the steady state was reached after ~22 and ~963 h, respectively. XRD data display progressive hydration of the Na-montmorillonite interlayer sites, as expressed by the stepwise increase of the water layers (WL) from 12.4 Å (1 WL) and 15.7 Å (2 WL) to 18.6-19.1 Å (3 WL). *CALCMIX* modeling of the montmorillonite revealed 65 ± 2% 3 WL and 35 ± 2% 4 WL at the steady state, suggesting complete hydration of the cement and bentonite was reached in the less altered zone. In contrast, cation exchange of Na^+ (0.36 to 0.08 a.p.f.u.) for Ca^{2+} (0.08 to 0.12 a.p.f.u.) was recognized in the montmorillonite interlayer sites close to the clay-cement contact, corresponding with a general depletion in CaO of the contacting Portland cement by a factor of 2.5. In addition, various of original cement phases were preserved, and only minor proportions of C-S-H phases were found.

Cation exchange within the clay and the preservation of original cement phases suggests that intense alteration processes occurred at the bentonite-cement contact. The Ca for Na substitutions strongly reduced the smectites swelling pressure and inhibited the formation of stable cement phases, by quantitative removal of CaO. These processes destabilized the clay-cement buffer and need further investigation for long-term nuclear waste disposal in sustainable underground repository sites that require clay-cement sealings.

Coupled spectromicroscopic investigations for improved conceptual models of soil carbon cycling

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Useful numeric models can only be constructed from accurate conceptual models. Biogeochemical interfaces or hotspots result from the spatial and temporal convergence of two or more different materials or processes. A comprehensive understanding of interfaces requires analytical techniques that complement each other in spatial, temporal, and elemental sensitivity. Our team has built expertise in the application of multiple (spectro)microscopic techniques to laboratory investigations of soil carbon cycling. We have sought to understand the interface between in inorganic mineral phases, organic compounds, and biological organisms occurring at the micron scale. We have made particular use of synchrotron based (spectro)microscopic techniques such as SR-FTIR, STXM/NEXAFS, hard X-ray microprobe, and X-ray microtomography as well as high resolution secondary ion mass spectrometry (NanoSIMS). This presentation will include technical considerations and challenges associated with sample preparation and handling required for successful application of multi-modal imaging analysis. In addition, the specific findings from several different investigations and the associated insights will be presented. The data shown will illustrate the challenges associated with obtaining statistically robust and quantitative measurements with this approach along with its' power for improving conceptual mechanistic models.

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