Interaction of corroding iron with bentonite at repository conditions

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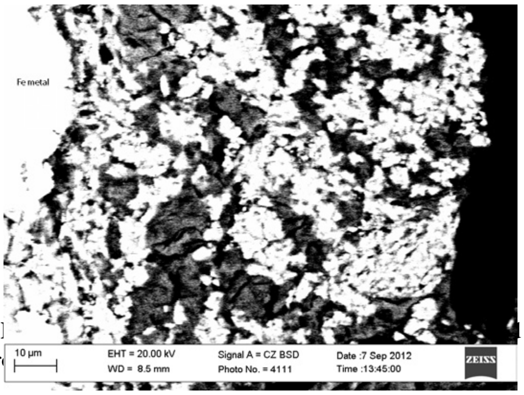
Iron canisters containing high-level nuclear waste surrounded by swelling bentonite backfill are foreseen to be emplaced in geologic formations. Corrosion will release Fe(II) species which will interact with the bentonite. This may induce cementation and clay mineral transformations. The details of this process are still poorly understood. In an in-situ experiment (ABM-1) at the Äspö Hard Rock Laboratory (Sweden), compacted bentonite materials in contact with steel were exposed to temperatures up to 140°C for 2 years. After overcoring, clay samples in contact with the Fe were sampled and analysed with microscopic techniques and μ-Raman spectroscopy. Both sides of an Fe metal structure, the Fe-water and the Fe-clay interfaces were studied.

The Fe-water side reveals a 100-200 μm thick corrosion layer conposed of magnetite, siderite, hematite, hydrous ferric oxide. The Fe-clay side exhibts a web-shaped ~100 μm thick layer of iron corrosion products (bright) and clay (dark). Siderite and Fe oxyhydroxides could be identified. The clay matrix is somewhat enriched in Fe and partly also in Mg, but no change in A/Si ratio was noted. At a distance of about 1 mm from the metal formation of $CaSO_4$ is observed. The front enriched in Fe exends 1-2 mm into the clay. Overall, the data suggest an intimate association of iron corrosion product and montmorillonite, but no formation of Fe silicates.



iron

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Insights into U-series weathering chronometers from size fraction distribution of U and Th nuclides in Himalayan soils and sediments

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As a result of varying mobility between different nuclides, disequilibria within the U-series decay chain potentially provides valuable constraints on the timescales of Earth surface processes. Models describing the evolution of both ($^{234}U/^{238}U$) and ($^{230}Th/^{238}U$) activity ratios during leaching are increasingly used to determine the duration of weathering and sediment transport. However, important assumptions in these models remain untested. While the basic mechanisms driving observed disequilibria during weathering are well-established (e.g. alpha recoil and preferential leaching), the picture remains somewhat fuzzy in terms of how bulk rocks, soils, and sediments, and the complementary dissolved phase, actually acquire their U and Th signature. Important puzzles, such as the reason for ^{234}U enrichment of some bulk solid phases despite the expected more rapid leaching of this nuclide, have yet to be completely resolved.

This study focuses on measurements of the U and Th nuclide abundance in a suite of samples (rocks, soils, sediments, and stream water) from the Middle Hills of Nepal, where solid products of weathering show distinct depletion in soluble elements (e.g. Ca, Na). Bulk soils and sediments have ($^{234}U/^{238}U$) above equilibrium ($\delta^{234}U$=+15-30‰ relative to equilibrium), increasing with weathering extent in the soil profiles. Variability in $^{234}U/^{238}U$ between size fractions of the Nepal soils demonstrates that the ^{234}U enrichment can be quantitatively explained by retention in the very fine (<0.2μm) fractions, which have measured $\delta^{234}U$>+100‰ and REE patterns characteristic of Fe-oxides. These results suggest that bulk solid phase ($^{234}U/^{238}U$)>1 can result from modern weathering processes because of retention in oxide phases.

Observed ($^{230}Th/^{238}U$) is highly variable and would imply long (tens to hundreds kyr) weathering duration based on simple models of leaching and decay. These durations require residence times that are inconsistent with the depth of weathering profiles and erosion rates inferred from ^{10}Be inventories in quartz from these locations. One possible explanation is that the measured sediment ($^{230}Th/^{238}U$) from these sites may be strongly influenced by grain size sorting.

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