Goldschmidt 2026 Abstract
https://doi.org/10.46427/gold2026.33460

Iron Corrosion Related Alteration Reactions of Bentonite and Their Impact on the Sorption and Diffusion of Selected Radionuclides.

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Concepts for nuclear waste disposal in deep geological formations often foresee the use of a bentonite barrier for the isolation of the waste packages from the geosphere. Properties, which qualify bentonite for this task are its swelling capacity, the low hydraulic conductivity, and the high sorption capacity towards radionuclides. Scenarios for the long-term evolution of waste repositories, usually consider a thermal phase with elevated temperatures and the saturation of the bentonite buffer with groundwater. When groundwater reaches iron-based waste canisters, oxygen will be rapidly consumed and canister corrosion will release Fe(II) and hydrogen. In the presence of aqueous Fe(II) smectite minerals in bentonite may undergo mineral alterations and properties of bentonite, which are essential for long-term repository safety, may be impeded.

In this contribution we present two types of experiments testing the possible impact of bentonite alteration on its long-term performance in a nuclear waste repository. In a first set of experiments mixed bentonite / iron powder suspensions are stored at 90°C for several months. Some experiments are subsequently stored at room temperature for further alteration. In all experiments we observe a rapid delamination of smectite minerals and an uptake of about 30 atom-% Fe(II). Diffraction and spectroscopic (IR, XAS) analyses indicate the formation of Fe-rich serpentine like alteration products.

In a second set of experiments, compacted bentonite is altered at 90°C inside through-diffusion cells next to a layer of compressed iron powder. After alteration the diffusion cells are subjected to through diffusion experiments using HTO and $^{36}Cl^-$ as radiotracers. Compared to bentonite exposed to 90°C in the absence of Fe, the Fe-altered samples show a decreased diffusivity for both tracers. Modelling indicates that diffusivity changes are mostly due to changes in tortuosity, decreases in pore volume play only a minor role. Post mortem analyses reveal magnetite and Fe-rich smectite as secondary alteration products. Only close to the iron layer smectite minerals delaminate like in the suspension experiments, and only locally SEM-EDX revealed the formation of Fe-serpentine minerals. Changes in altered bentonite's sorption capacity towards ^{60}Co(II) and ^{137}Cs(I) are minor, $\log_{10}K_D$(Co(L/kg)): 4.5±0.3 (unaltered), 4.8±0.5 (altered); $\log_{10}K_D$(Cs(L/kg)): 3.7±0.3, (unaltered), 3.9±0.2 (altered).