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Sulfide-bentonite interaction: experimental results for dispersed and compacted systems.

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Many geological repositories for nuclear waste disposal employ bentonite as a buffer confining and protecting waste canisters. Anaerobic conditions will prevail and sulfate reducing bacteria expected at the bentonite/host rock interface, eventually produce sulfide, which presents a potential threat for metallic canisters. In this study, sulfide solutions were reacted with different types of iron-bearing bentonites, pure montmorillonites and iron (oxyhydr)oxides in dispersed batch and diffusion experiments (compacted clay at various densities). Solutions were analyzed by colorimetry to determine final sulfide and polysulfide concentrations, and solids were analyzed by ^{57}Mössbauer spectrometry to determine iron speciation. A common CEC determination method (Copper-Triethylenetetramine) was also modified in order to determine both CEC and sulfide content.

Notable sulfide consumption was observed in dispersed batch experiments, ranging between 150 and 700 $mmol_S \cdot kg_{bentonite}^{-1}$. The main identified product of sulfide oxidation was elemental sulfur, which further reacted with excess sulfide to form polysulfides. The main pool of reactive iron was found to be smectite structural Fe^{3+}. The reaction between sulfide and iron (oxyhydr)oxides was inhibited because of large sulfide excess. Another sulfide immobilization mechanism was identified as structural exchange with clay structural hydroxyl groups (Hadi *et al.*, 2023). Reduction of structural Fe typically induces an increase of the negative layer charge; however, no notable CEC changes were observed. This indicated that dehydroxylation was the main charge compensating mechanism.

The reactivity of structural Fe in compacted clay cores was very similar. However, the accessory iron (oxyhydr)oxide was more reactive toward sulfide and formed mainly greigite. The reduction of structural Fe induced a small (5%) decrease in swelling pressure.

This study highlights that bentonites are efficient scavengers of sulfide and also that structural Fe reduction only marginally affects the main confining properties of the bentonite (swelling pressure, diffusivity of other ions and neutral species).

Reference

Hadi, J., Greneche, J.-M., Wersin, P., Koho, P. & Pastina, B. (2023) Determination of sulfide consumption by Fe-bearing components of bentonites. *Clays and Clay Minerals*, **71**, 577-599.