A microspectroscopic study on the influence of the inherent heterogeneity of waste repository materials on contaminant uptake

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Large-scale problems such as toxic metal environmental disturbance and nuclear waste remediation and storage are now recognized to be interconnected to small scale-mechanisms. In this study we have combined micro X-ray fluorescence (XRF), micro X-ray absorption spectroscopy (XAS) with ‘bulk’ XAS to gain spatially resolved information on the speciation and structural coordination environment of contaminants solidified in cement. Cement-based materials play an important role in multi-barrier concepts developed worldwide for the safe disposal of radioactive wastes in underground repositories. From a chemical standpoint, cement is a complex heterogeneous mixture (calcium (aluminium) silicate hydrates, Ca(OH)2, calcium aluminates, and some highly reactive minor phases) with discrete particles typically in the size range of a few to a few hundred micrometers.

An example addressing the heterogeneity of sorbent cement concerns the microbial influenced degradation of a cobalt-containing cement matrix. Since microorganisms can affect the chemical speciation of contaminants in a cementitious environment, the cobalt-containing cement material was analyzed by a set of analytical techniques before and after extended microbial degradation by chemotrophic material was analyzed by a set of analytical techniques before cementitious environment, the cobalt-containing cement affects the chemical speciation of contaminants in a cobalt-containing cement matrix. Since microorganisms can cement concerns the microbial influenced degradation of a.

Spatial and temporal variation of Zn species in the rhizosphere of a contaminated soil

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Contamination of soils with Zn from anthropogenic sources is an increasing problem in industrialized countries, potentially impeding the production of food and fiber. The effective, short-term effect of Zn depends on the amount and the chemical nature of the most mobile species, while the long-term ecotoxicity is determined by the resupply of the mobile pool from more stable phases. Thus, quantitative speciation as well as its variation with time is a prerequisite for long-term risk assessments.

Due to the heterogeneous distribution of water, minerals organic matter and biota, diverse chemical microenvironments may coexist on micrometer to millimeter scales, leading to gradients in Zn speciation. Especially in the rhizosphere where roots alter, e.g., pH, redox, and the concentration of organic chelators, Zn speciation may significantly deviate from that in bulk soil. Therefore, we investigated the Zn speciation after mixing a soil with dust particles emitted by a metal foundry, using bulk X-ray spectroscopy and selective sequential extractions (SSE) to follow the temporal variation, and using micro X-ray fluorescence (_XRF) and _XAFS spectroscopies (5 µm spatial resolution) to follow the spatial variation in the rhizosphere soil.

The dust particles consisted predominately of zincite (ZnO), which dissolved in the soil within 7 d (water saturated) or 6 months (unsaturated). After dissolution, the predominant part of Zn was bound as inner-sphere sorption complex. The figure below shows the Zn distribution within a 1.5 x 1.5 mm2 large area around a root (delineated by hatched lines). A Zn plume extends from a dissolving zincite particle towards the root and Mn- and Fe-rich zones. Micro-XAFS suggested that the dominant Zn species after 3 months is tetrahedrally coordinated Zn sorbed to root tissue and to Mn- and Fe-oxides.