Radionuclide sorption in heterogeneous systems: From model mineral oxides to complex rocks

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The fate of radionuclides in natural rocks is governed by their sorption reactions onto heterogeneous systems. Fundamental process understanding of the retardation mechanisms is crucial in the long-term safety assessment of nuclear waste repositories.

The “Component Additivity” (CA) approach is widely used to model radionuclide sorption onto rocks or soils in a realistic manner. This bottom-up approach is based on the principle that the sorption in a complex material is determined by competitive sorption effects from the individual minerals. In the context of repository safety assessment the CA approach is used in the smart K_d-concept, which is developed for complex geochemical transport models to describe the radionuclide migration in the far-field of a repository more realistically [1].

In this work, batch sorption experiments of radionuclides, e.g. Np(V) and U(VI) onto mixtures of different mineral oxides, such as iron oxides, silicium dioxide, manganese oxides were performed varying the ratio of mineral oxides, solid-liquid-ratios and geochemical conditions. Vibrational (IR) and luminescence spectroscopy (TRLFS) were performed to identify sorbed species and to gain mechanistic understanding of the radionuclide sorption processes. Surface complexation parameters (such as surface protolysis and complex formation constants) of single minerals and mixtures thereof were derived, namely from titration and batch sorption experiments.

Finally, the experimental results were compared with results obtained from sorption predictions to verify the robustness and applicability of the CA approach. Based on the results obtained, a first estimation on the applicability of the CA approach for radionuclide sorption processes is presented.

[1] Stockmann *et al.* (2017), Chemosphere **187**, 277-285.