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Does a stability constant decide on a repository permit?

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Safety of a nuclear waste repository is based to a large extent on the isolation of the radioactive waste within a suitable host rock. Clay rocks provide an option due to their very low hydraulic conductivity only allowing diffusive transport. Diffusion processes in clay formations are complex due to the diffuse double layers (DDL) enveloping the clay minerals to compensate their net surface charge and the associated different migration behaviour for cationic, anionic and neutral species. Therefore, determination of the speciation of an element in the porewater is essential to quantify migration lengths precisely. Safety assessments are based on numerical simulations to cover time periods of up to one million years and thus the predominant species of a radionuclide, dependent on the stability constants within the law of mass action, might be signififcant.

In the present study, we use uranium, one of the main components in spent fuel, as an example for the diffusion in the Swiss Opalinus Clay, a potential host rock for the storage of nuclear waste. In the geochemical system, uranium is mainly present as U(VI) in ternary uranyl complexes with calcium and carbonate, whereby speciation depends on the selected thermodynamic data (Hennig et al., 2020). For instance, the stability constants for the neutral uranyl complex $Ca_2UO_2(CO_3)_3$ differ slightly in literature. Depending on the selected one, either the neutral or the anionic complex $CaUO_2(CO_3)_3^{2-}$ is the predominant species in the system with an associated varying interaction with the DDL of the clay minerals. With our one-dimensional, multi-component diffusion models we quantified the effect of the selected stability constant on the diffusion length for the host rock scale.

The chemistry in the porewater of the three facies of the Opalinus Clay, shaly, sandy and carbonate-rich, plays a key role for the sorption processes (Hennig et al., 2020) as well as for the composition and thickness of the DDL and therewith the diffusive transport. Based on our results, we show, that the influence of the predominant uranium species on the migration lengths varies between the individual facies, but is overall negligible for the host rock scale. Consequently, a stability constant is not decisive for the required thickness of the host rock as geological barrier.

Hennig, T.; Stockmann, M.; Kühn, M. Simulation of diffusive uranium transport and sorption processes in the Opalinus Clay. Applied Geochemistry 2020, 123. doi:10.1016/j.apgeochem.2020.104777.