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Effects of uncertainties in sorption modelling in crystalline rocks

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One of the most important radionuclides (RN) retardation process in the vicinity of nuclear waste repositories and radioactive contamination is sorption (described by distribution coefficients – K_d values). This term usually subsumes ion exchange, surface complexation, surface precipitation, and formation of secondary minerals including solid solutions. Its modelling is often based on mechanistic approaches (smart-K_d) which require a full parameterization for all relevant mineral surfaces – in real systems this results in rather large matrices, or to be more precisely, sparsely populated matrices.

Dealing with associated data gaps in an appropriate way is not trivial and application-specific. Options to consider are the use of chemical analogies (for both RN and mineral phases), established estimation methods (e.g. Linear Free Energy Relationship - LFER), or, at the extreme, ignoring some interactions.

Building on the discussions in a recent sorption workshop held at the Amphos21 site in Barcelona in September 2024, this contribution attempts to quantify the impact of the different approaches on the derivation of sensible, consistent and comprehensive parameter sets required for calculating the K_d values of various RN. The calculations focused on U, Np & Pu, and only considered surface complexation models (SCM) and ion exchange (IEx). The work originated from a geostatistical treatment of sorption in crystalline rocks, based on characterizations (mineral content and pore water composition) of real samples from the Lusatian region in Germany.

The mineral composition was dominated by quartz, feldspars, biotite, chlorite, muscovite and hematite. While the SCM / IEx parameterization is acceptable for many RN-mineral combinations, the case of biotite is a real challenge. The following approximations were tested: basic case (ignoring the mineral), approximation by a combination of illite and hematite, approximation by chlorite, parameterization with SCM derived from (low-quality) raw data.

It was found that ignoring biotite is not an acceptable option, nor is the surrogate by a combination of illite and hematite. In case of poor quality SCM data, at least the assigned error can be used in a sensitivity analyses to quantify the effects on K_d values, and consequently identify the need for additional experimental effort.