

Trace element incorporation in barite under natural environmental conditions – implications for the long term safety of radioactive waste repositories

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In deep geological repository systems, containers for highly radioactive waste may encounter groundwater, which ultimately leads to the release of radionuclides like Ra, U, and Pu. In contact with contaminated fluids and under the relevant physico-chemical conditions, barite (BaSO_4) and calcite (CaCO_3) may (re)crystallize and incorporate significant quantities of radiotoxic elements. The solubility of these host phases limits the fixation and mineral-fluid partitioning coefficients are important to long-term analytical model calculations, which are the only way to demonstrate safety over the required time spans (e.g. 1 million years in Germany).

Usually these parameters are determined by laboratory-based experiments, the results of which, however, cannot be easily transferred to conditions of an actual repository site. This is evident from the discrepancy between experimental and natural calcite-fluid partition coefficients.

Hence, we investigated the trace element distribution and partition coefficients of natural barites from a borehole in the äspö Hard Rock Laboratory, Sweden, and several mines in SW-Germany by high-resolution LA-ICP-MS.

Preliminary results show, that La is particularly enriched in Sr-poor zones, indicating that the La uptake is linked to the amount of celestine in barite. However, the incorporation of U and La does not necessarily coincide, since while La is largely incorporated into the bulk crystal, U seems to be incompatible with the barite structure and is only found along cracks or in pores. The assumed analogy of La for U and other actinides is therefore questionable for barite.

SIMS-derived $\delta^{34}\text{S}$ values vary between 11-22 ‰ and seem to reflect fluid mixing events.

Barite-fluid partition coefficients could be obtained from the outermost crystal rims and fluids sampled from the borehole. These are $P_{\text{La}} = 2.29 \times 10^{-2}$ and $P_{\text{Sr}} = 1.74 \times 10^{-4}$ [with $P_{\text{Me}} = (\text{Me}/\text{Ba})_{\text{barite}}/(\text{Me}/\text{Ba})_{\text{solution}}$ expressed as ratios by weight]. For La this result is in line with published continental barite data. Barite partition coefficients for La and Sr are 3-4 orders of magnitude below calcite values of the same sampling location, indicating a much lower immobilization potential of barite compared to calcite.

By further examination of the large sample suite, a

