The uptake of ^{226}Ra during the recrystallization of Barite: Effect of ionic strength, Sr_{aq} and temperature

M. KLINKENBERG[1]*, F. BRANDT[1], J. POONOOSAMY[1], J. WEBER[2], D. BOSBACH[1]

[1]Institute of Energy and Climate Research (IEK-6), Forschungszentrum Jülich GmbH, 52425 Jülich, Germany, (*correspondence: m.klinkenberg@fz-juelich.de, f.brandt@fz-juelich.de, j.poonoosamy@fz-juelich.de, d.bosbach@fz-juelich.de)

[2]University of Arizona, Kuiper Imaging & Microscopy Facility, Tucson, Arizona 85721, USA (julianeweber@email.arizona.edu)

In recent safety cases for the deep geological disposal of spent nuclear fuel, ^{226}Ra is regarded as a relevant radionuclide. Therefore, retention mechanisms e.g. sorption or uptake in solid solutions are studied, aiming at a more realistic thermodynamic description of the fate of ^{226}Ra under repository relevant conditions. Due to the presence of Ba in spent nuclear fuel which may be released during corrosion and sulfate in many groundwaters, the uptake of ^{226}Ra and the thermodynamics of the $(Ra,Ba)SO_4$ solid are currently studied. Despite the presence of strontium in many groundwaters, so far little is known about the effect of aqueous Sr concentrations (Sr_{aq}) on the recrystallization and ^{226}Ra uptake process as well as about the effect of ionic strength.

Here, we present new experimental data on the recrystallization of barite to unravel the effect of Sr_{aq} and ionic strength on the ^{226}Ra uptake kinetics and solubility. Final Ra concentrations (Ra_{aq}) are interpreted based on a recent thermodynamic model for the $(Ba,Sr,Ra)SO_4$ solid solution. With higher ionic strength, the final ^{226}Ra concentrations are close to predicted values for low solid/liquid (S/L) ratios whereas at high S/L further kinetically driven Ra-uptake was observed. In contrast to ionic strength, the presence of Sr_{aq} can have a significant inhibiting effect on the ^{226}Ra-uptake into barite which depends on temperature and the S/L ratio. In the extreme case of low S/L ratio and ambient temperature conditions, a complete inhibition of Ra-uptake into barite was observed. At higher temperatures the uptake kinetics were affected by the presence of Sr, the final Ra_{aq} were close to the predicted thermodynamic equilibrium [1].

[1] Brandt *et al.* (2018) *Minerals* **8**(11), 520.