

Ra uptake by the Sr-rich solid solution of (Sr,Ba)SO₄

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Solid solution formation is an important mechanism of radionuclide uptake by mineral phases which is favored in natural systems due to a lowered configurational entropy. Recent studies have shown that the formation of a (Ba,Ra)SO₄ solid solution significantly reduces the solubility of ²²⁶Ra in aqueous systems. This result is very relevant for the direct disposal of spent nuclear fuel in a deep geological formation, where ²²⁶Ra would dominate the dose after 100,000 years [1]. In natural systems SrSO₄ often occurs along with BaSO₄ implying that Ra-uptake should be assessed within the system of (Sr,Ba,Ra)SO₄ + H₂O. A recent thermodynamic modelling study [2] predicted a significant uptake of Ra into the ternary (Sr,Ba,Ra)SO₄ solid-solution.

Here we present results of long-term batch-recrystallisation experiments on Ra-uptake in a Sr-rich part of the ternary system. A mechanical mixture of celestite with a small amount of barite and a (Sr,Ba)SO₄ solid solution of equivalent composition were put into a contact with ²²⁶Ra,aq. We observed a significant uptake of ²²⁶Ra in both cases proceeding via the formation of the minor ternary Sr-, Ba- and Ra-rich phase within the major Sr-rich phase. In longer experiments the ternary phase disappeared leading to the formation of a mixture of Sr- and Ba-rich phases, within which Ra could not be detected by scanning transmission electron microscopy-energy-dispersive X-ray spectroscopy (STEM-EDX). The final ²²⁶Ra concentrations in both experiments were similar and close to predicted levels. The formation of the intermediate phase is interpreted based on structural and kinetic considerations.

[1] Norrby, S. et al. (1997). "SKI SITE-94 Sakerhetsanalys foer Djupfoervar iett Kristallint berg". Stockholm, Sweden. [2] Vinograd et al. (2018). Applied Geochemistry, in press.