

Experimental studies in the system (Ba,Sr,Ra)SO₄ + H₂O

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The relevance of solid-solution formation between pre-existing barite, BaSO₄, and Ra released from spent fuel is already well established and addressed in assessments regarding the long-term safety of high-level nuclear waste disposal in deep geological formations [1, 2]. Recent studies have demonstrated a significant incorporation of Ra(aq) into pure barite e.g in the review of Prieto et al. (2016) [3]. However, in natural systems, barite is often accompanied by celestite, SrSO₄, or is represented by intermediate compositions of the (Ba,Sr)SO₄ solid solution. Earlier studies are contradictory with regard to the effect of Sr on the uptake of Ra [4] [5] [6].

Here, we present new experimental results of batch-recrystallisation experiments on the uptake of Ra (1) by celestite, (2) by a (Ba, Sr)SO₄ solid solution and (3) by a mechanical mixture of celestite with small amounts of barite.

In the presence of celestite, already small amounts of barite result in a significant Ra decrease by the formation of a ternary solid solution. Even high purity celestite which is stable in reference experiments without Ra, is destabilized in the presence of radium leading to a release of Ba. Secondary radiobarite forms; in the case of primary barite, this is transformed to a secondary ternary solid solution. The microscopic features of the secondary phase next to the primary barite point to a classical dissolution-reprecipitation mechanism [7].

[1] Norrby, S. et al. (1997). "SKI SITE-94 Saekerhetsanalys foer Djupfoervar iett Kristallint berg". Stockholm, Sweden. [2] Nagra, 2002, Nagra Technical Report NTB 02-05 [3] Prieto, M., et al. (2016), Progress in Crystal Growth and Characterization of Materials. [4] Ceccarello S. et al. (2004). Miner. Eng. 17. [5] Jucker, H., Treadwell, W.D. (1954), Helvetica Chimica Acta, v. 37, No. 7. [6] Zhang, et al. (2014), ES&T, v. 48, no. 8. [7] Ruiz-Agudo, E., et al. (2014), Chem. Geol., 383.