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Recrystallization of Barite in the presence of Radium

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The uptake of Radium by Barite via solid solution formation is an important process controlling the solubility of Ra in aqueous systems. Recent studies have focused on ambient conditions (Bosbach et al., 2010; Curti et al., 2010). Here, we have focused on the understanding of kinetics and uptake mechanisms at room temperatures. Pure barite powder was put into contact with an aqueous solution with an initial Ra/Ba ratio of 0.3 ($5 \cdot 10^{-6}$ mol/L Ra) at neutral pH. Two barites of different morphology and specific surface area were used during the recrystallization experiments at close to equilibrium conditions and with variation of solid to liquid ratio.

The experimental results show a decrease of the Ra concentration to $3.5 \cdot 10^{-9}$ to $7 \cdot 10^{-9}$ mol/L within the first 70 days of the experiment at a solid/liquid ratio of 5 g/L. At a solid/liquid ratio of 0.5 g/L a slower decrease of the Ra concentration to $2 \cdot 10^{-8}$ mol/L is observed after 180 days. The decrease of the Ra concentration is apparently not related to the specific surface area of the barite crystals. The final radium concentrations are in the range as can be expected from thermodynamic calculations assuming full re-equilibration of the barite to a $Ra_xBa_{1-x}SO_4$ solid. Different thermodynamic models describing the mechanism of Ra incorporation into barite are discussed: (1) Ba – Ra exchange into the crystal volume, combining the Ra – Ba exchange with the Ba – Ba recrystallization rate at equilibrium conditions (Curti et al, 2010);

(2) the formation of a Ra-Ba-Phase on barite surfaces. The formation of a Ra-Ba phase on the barite surfaces could be possible because all experiments are already slightly supersaturated with regard to $Ra_xBa_{1-x}SO_4$ after about one day. Crystallization rates calculated according to this model are in a similar range for all experiments when normalized to the barite surface area.

The results of this study will provide the basis for further spectroscopic and microscopic investigations in order to obtain a molecular-level understanding of the Ra incorporation into barite.

TOF-SIMS measurements were performed in order to investigate the spatial distribution of Ra within the barite crystal. Sachtleben barite crystals of 0.5 g/L experiments at RT were investigated after different time intervals. At the end of the experiment (350 days) the results show a homogeneous distribution of Ra in the whole volume of the crystals.

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[2] Curti, E.; Fujiwara, K.; Iijima, K.; Tits, J.; Cuesta, C.; Kitamura, A.; Glaus, M. & Müller, W. (2010) Geochimica et Cosmochimica Acta, 74, 3553-3570