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Barite Recrystallization to Witherite in the Presence of Carbonate, and the Impact on Radium Retention

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For geological disposal of nuclear waste the (geo-)technical barrier system supports protection against groundwater contact. Since intrusion of solutions into disposal rooms has to be considered on a long term, chemical reactions between the waste forms and intruding solutions must be taken into account within the safety case of a repository. As sulfate containing groundwater contacts the waste, barite ($BaSO_4$) formation is expected. Radium (Ra) uptake by barite occurs when dissolved Ra reacts with barite, leading to Ra retention in a $(Ba,Ra)SO_4$ solid-solution [1]. On the other side, barite reaction with aqueous carbonate at high pH, e.g. due to the alteration of cementitious material inside a repository, may cause barite to convert into witherite ($BaCO_3$) [2]. Carbonate presence likely alters the chemical behaviour of barite surfaces, via surface mixing or by witherite layer formation through dissolution-precipitation [3]. Aim of this study is to investigate the effect of carbonate on Ra retention by barite / witherite. The presented results highlight barite transformation into witherite in the absence of Ra.

Batches of barite were contacted with carbonate solutions (1-100 mM, pH 7-11). X-ray diffraction (XRD) showed shifts of reflection peaks in batches with pH values of 9-11 and carbonate concentrations of 10-100 mM, indicating a witherite-barite solid-solution formation [3]. Reaction rates depend on the type of barite used. In general, reactions slow down after about 7 days, likely due to surface passivation. Cubes cut from natural barite, were reacted with 100 mM carbonate solutions at pH 11. As a result, a porous witherite layer with (271±32) µm formed giving insight into recrystallization process progress and mechanism.

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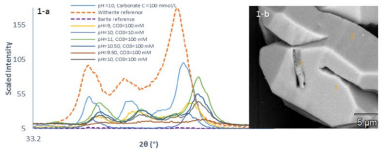

Fig. 1. (a-b) XRD reveals peaks shifts and a SEM image of barite powder replaced by witherite, respectively.

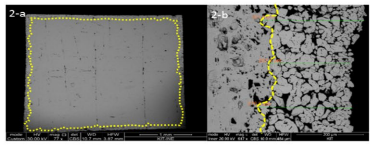

Fig. 2. (a-b) Witherite layer and its thickness for 15 day reaction period cube, respectively.