

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

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Deep geological repositories for nuclear waste are protected with (geo-)technical barriers against groundwater intrusion. However, over geological periods of time, water intrusion and subsequent corrosion are expected. When sulfate (SO_4) containing groundwater reaches the waste it may react with barium (Ba, fission product present in waste forms), to form barite (BaSO_4). On the long run, radium (Ra, uranium progeny product) will be released from the waste. Ra may be incorporated into available barite to form a $(\text{Ba,Ra})\text{SO}_4$ solid solution [1]. When carbonated groundwater with elevated pH due cementitious materials (a disposal barrier) reacts with barite, conversion into witherite (BaCO_3) is possible [2]. The conversion proceeds via a dissolution-precipitation mechanism [3]. The present study aims to investigate carbonate effect on Ra retention by barite or witherite and the presented results shed light on barite conversion into witherite.

Commercial and natural barite powders were equilibrated with 0.1M NaCl for several days and exposed to carbonate solutions (1-100 mM TIC, pH 9.5-11.0). Witherite formation was more pronounced at higher carbonate activity. The commercial powder showed a higher reaction rate due to higher surface reactivity. However, a commercial powder that was equilibrated for several years before carbonate exposure showed much less witherite growth. Natural powder formed less witherite and rate slowed down after several days, possibly due to surface passivation. The newly formed particles were either witherite crystals or witherite overgrown / intergrown barite crystals. The possibility to form witherite from barite indicates the relevance of the investigated process. However, the ability of witherite to incorporate Ra, i.e. to form $(\text{Ba,Ra})\text{CO}_3$ [4] indicates that even upon barite-witherite transformation only minor amounts of Ra may be released from the solid into the liquid phase.

[1] Heberling et al. (2018) *Geochim. Cosmochim. Acta* 232, 124-139.

[2] Rendón-Angeles et al. (2008) *J. Materials Science* 43, 2189-2197.

[3] Putnis and Putnis (2007) *J. Solid State Chemistry* 180, 1783-1786.

[4] P.L. Brwon et al. / *Geochimica et Cosmochimica Acta* 255 (2019) 88-104.

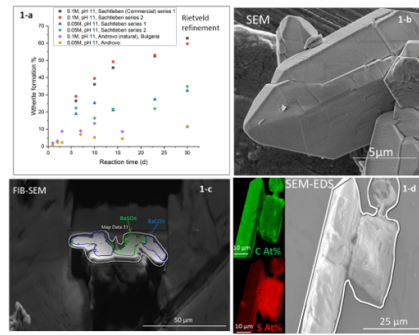


Fig. 1. (a) Witherite formation as function of time starting from two types of barite at various conditions, as indicated in the legend (b) SEM-SE image of witherite crystals grown from barite at elevated TIC and pH, (c) SEM-FIB cut for a witherite crystal overgrowing barite and (d) shows witherite growing adjacent to barite