

## Characterization of (Ba,Ra)SO<sub>4</sub> solid solutions with true nm-scale resolution

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The (Ba,Ra)SO<sub>4</sub> solid solution is potentially relevant for the direct disposal of spent nuclear fuel because it could retain Ra in certain cases addressed *e.g.* in the swedish safety assessment for the planned final repository in Forsmark [1] [2]. Previous studies on Ra uptake into barite showed the replacement of barite by a (Ba,Ra)SO<sub>4</sub> solid solution under repository relevant conditions [1] [3]. Nevertheless, the spatial resolution of the characterizations allowed not to determine the details of the Ra distribution within the solid or the uptake mechanism [3].

Here, we present the characterization of (Ba,Ra)SO<sub>4</sub> solid solutions from Ra uptake experiments [1] [3] in comparison with Ra-free reference samples by applying a combined nanoanalytical approach. High-resolution transmission electron microscopy (HR-TEM) and atom probe tomography (APT) were used because these techniques provide chemical composition and structural information with true nm-scale resolution [4]. Both cross-section and plan-view lamellae were prepared by Focused Ion Beam (FIB) from 10 – 20 μm barite particles.

A characteristic layered internal structure was observed in the barite reference material and identified by APT & HR-TEM to be fluid inclusions (< 20 nm) with variations in the H<sub>2</sub>O, H<sub>3</sub>O, Na and Cl content. During the later stages of Ra uptake these fluid inclusions coalesce and become macropores (>1 μm). In contrast to this, the microstructure from the Ra-free barite reference sample remains unchanged. Hence, these fluid inclusions provide a possible path for the Ra into the crystal volume during a dissolution-reprecipitation process.

[1] Brandt et al. *Geochim. et Cosmochim. Acta*, 2015, **155**,1-15. [2] Hedin *et al.*, 2011. Technical Report, TR-11-01. [3] Klinkenberg et al., *Environ. Sci. Technol.*, 2014, **48**, 6620-6627. [4] Kelly & Larson, *Annu.Rev.Mater.Res.*, 2012, **42**, 1-31.