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A XAS study of baryte nucleation: implications on radium mobility

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Baryte is of interest to nuclear waste disposal as it is considered to be the main scavenger of ^{226}Ra, a long-lived nuclide playing a major role in the safety assessment of planned disposal sites [1]. It is characterized by a complex kinetic behavior, particularly metastability of supersaturated solutions. Therefore, it can be anticipated that the release and distribution of radium in repository systems will strongly depend on baryte precipitation kinetics.

In this study, we carried out X-ray absorption experiments at the Ba-L_{III} edge on $BaCl_2$-Na_2SO_4 solution droplets shortly after mixing of the two components. The degree of supersaturation was controlled by fixing the relative humidity in a dedicated cell sealed with a 200 nm thick Si_3N_4 window. XANES spectra were recorded to detect the onset of homogeneous precipitation as a function of saturation index (SI) and compared to those of well-defined Ba references. The results (Fig. 1) indicate that mixed $BaCl_2$-Na_2SO_4 solutions are stable up to SI ~ 3, in agreement with predictions of classical nucleation theory (CNT) [2].

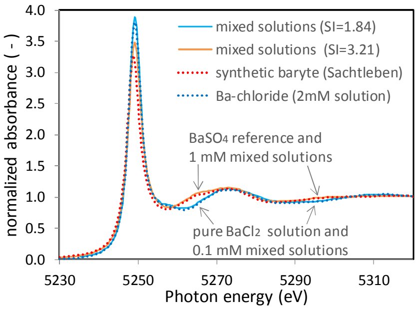

Figure 1: XANES spectra of supersaturated $BaCl_2$-Na_2SO_4 aqueous mixtures compared to reference spectra.

CNT, including both homogeneous and heterogeneous baryte precipitation calibrated to the experimental data, was implemented in a lattice Boltzmann model simulating radium precipitation in a reactive porous medium. The model allows exploring the influence of baryte kinetics on the partitioning of Ra between aqueous phase and solid at the pore scale.

[1] Brandt et al. (2015), *GCA* 155,1-15; [2] Prieto (2014) *Mineral. Mag.* 78,1437-1447.

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