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Stability investigations of actinide doped calcium silicate hydrate phases in highly saline electrolytes

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To evaluate the retention potential of the concrete barrier in a nuclear waste repository towards actinides in the presence of high saline water, leaching experiments with actinide-doped calcium silicate hydrate (CSH) phases were performed in highly saline electrolytes. Therefore, U(VI)- and Cm(III)-doped CSH phases with different C/S ratios (1.0-2.0) were synthesized directly in presence of either U(VI) or Cm(III) and characterized by time-resolved laser-induced fluorescence spectroscopy (TRLFS), infrared (IR) spectroscopy, powder X-ray diffraction (XRD), thermogravimetric analysis (TGA), differential scanning calorimetry (DSC), and scanning electron microscopy (SEM). The time-dependent release of Ca, Si, U, and Cm from CSH phases into brines containing either 2.5 M NaCl, 2.5 M NaCl/0.02 M Na_2SO_4 , 2.5 M NaCl/0.02 M NaHCO_3 or 0.02 M NaHCO_3 for U(VI)-doped CSH phases or 2.5 M NaCl/0.02 M NaHCO_3 or 0.02 M NaHCO_3 for Cm(III)-doped CSH phases was monitored in batch leaching experiments for 30 or 60 days, respectively. Subsequently, leaching induced changes of the CSH structure and of the U(VI) or Cm(III) coordination environment were investigated with TRLFS, IR spectroscopy and XRD.

Site-selective TRLFS studies of the Cm(III)/CSH binding at 8 K revealed a luminescence line-narrowing effect, which could be ascribed to the presence of a continuum of similar curium species on two different sorption sites in the CSH interlayer. The leached CSH phases showed pronounced differences in terms of decomposition behavior and actinide release depending on their C/S ratio, leaching electrolyte, and incorporated actinide. U(VI)-doped CSH phases leached in 2.5 M NaCl showed an increased release of Ca and a U(VI) precipitation as uranophane while the leaching in 0.02 M NaHCO_3 led to a mobilization of U(VI) as an aqueous $\text{Ca}_2\text{UO}_2(\text{CO}_3)_3$ species.

In contrast, Cm(III) was not mobilized by NaHCO_3 but incorporated into newly formed secondary phases like calcite and vaterite as detected by site-selective TRLFS. The comparison between leaching experiments performed in 0.02 M NaHCO_3 and 2.5 M NaCl/0.02 M NaHCO_3 revealed that the presence of 2.5 M NaCl increases the carbonate-induced U(VI) mobilization from CSH phases with higher C/S ratios while no influence on the Cm(III) retention was detectable.

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