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Influence of the competition of Al on the retention of trivalent actinides and their homologues in feldspar

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Crystalline rock is one possible host rock for the final disposal of nuclear waste in a deep geological formation. For a proper safety assessment, it is of utmost importance to understand retention mechanisms of radionuclides at the water-mineral interface. The retention of trivalent actinides by K-feldspars, a main component of crystalline rock, was recently investigated by our group [1]. However, no sorption data of minor actinides is available for the series of Ca-feldspars (plagioclases) with different Al:Si ratios in the crystal lattice, which most likely causes differences in surface charge, dissolution, and sorption behavior.

Here, we study the sorption of trivalent actinides and their rare earth element homologues on Ca-feldspars quantitatively and mechanistically. We find that plagioclases show a stronger sorption affinity to actinides than K-feldspars (alkali feldspars). A possible explanation might be an increased concentration of Al^{3+} in the aqueous phase due to enhanced dissolution of the Ca-feldspars. The dissolved Al^{3+} may affect surface charge and sorption processes onto mineral surfaces, and in such case the underlying molecular mechanisms are expected to be complex and are clearly not well understood.

Therefore, we study the impact of Al^{3+} on the surface charge of K-feldspar and the concomitant effects on the retention of trivalent actinides and lanthanides. Zeta potential investigations reveal a strong impact of Al^{3+} on the surface charge of K-feldspar. The zeta potential increases for pH 4.5–7.5, and this increase is enhanced by higher added Al^{3+} concentration. Results from batch sorption experiments of Eu onto K-feldspar show a steeper sorption edge upon addition of Al^{3+}, which can be interpreted as a slightly stronger sorption affinity. Indeed, in the presence of Al^{3+}, K-feldspar behaves much like Ca-feldspar. To gain information about the formed surface complexes time resolved laser-induced fluorescence spectroscopy is applied. Potentially formed secondary phases of Al are evaluated by a combination of microscopy (SEM, TEM, Raman, AFM) and diffractometry (CTR/RAXR, high resolution PXRD). Ultimately, this will provide a better understanding of the fundamental mechanisms of sorption process of the minor actinides on naturally occurring mineral phases.

[1] Neumann, J. et al. (2021) J. Colloid Interface Sci. 591, 490-499.