

Retention of trivalent actinides (Am, Cm) and lanthanides (Eu) by Ca feldspars

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Abstract. The transport of radionuclides in the environment is a major problem for the safety assessment of radioactive waste repositories. Storage in deep geological repositories is considered a safe disposal strategy because of their ability to isolate hazardous components from the biosphere for hundreds of thousands of years. Minor actinides (i.e. Am, Cm and Np) dominate the radiotoxicity of spent nuclear fuel over geological timescales. In underground repositories, reducing conditions are expected, and therefore the trivalent oxidation state is dominant for Am and Cm as well as possibly for Pu. For investigations of the mobility of the trivalent actinides Am^{3+} and Cm^{3+} , the less toxic trivalent rare earth elements, in particular Eu^{3+} , are commonly used.

Besides clay and salt, crystalline rock is considered a possible host rock for deep geological repositories. Crystalline rock (e.g. granite) consists mainly of quartz, mica and feldspar. The latter forms common aluminosilicates making up ~ 60 vol. % of the earth's crust, but their sorption behaviour is not well understood, especially for the Ca-bearing members of the group.

Here, we study the sorption of trivalent actinides and their rare earth element homologues on plagioclase which are Ca-bearing feldspars, quantitatively and mechanistically. Zeta potentials of various Ca feldspars show an unexpected increase at pH 4–7, which becomes more pronounced as the amount of Ca in the crystal lattice increases. This can be interpreted by assuming uptake of Al^{3+} and/or the precipitation of an Al phase, where Al originates from feldspar dissolution at different pH values.

Nevertheless, only minor differences were found in the retention and surface speciation of Cm^{3+} on Ca and K feldspars (Neumann et al., 2021). Ca feldspar has a slightly higher potential to retain trivalent metal ions compared to K feldspar. An inner-sphere (IS) complex and its two hydrolysis forms have been identified on both minerals, but the hydrolysis of the IS complex is stronger in the Ca-rich mineral.

A surface complexation model for Ca feldspar was developed by combining the batch sorption data and the spectroscopically identified surface complexes to describe the experimental data. These data will be the basis for the improvement of transport simulations for a reliable safety assessment of potential radioactive waste repositories in crystalline rock.

References

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