Goldschmidt 2021 Abstract
https://doi.org/10.7185/gold2021.3156

Spatially resolved sorption of Cm(III) on crystalline rock: influence of surface roughness and mineralogy

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Many countries will use deep geological repositories to dispose of their highly active nuclear waste. Crystalline rock is a potential host rock because of its high stability, heat resistance and low solubility. However, it possesses a high inherent mineralogical heterogeneity. Using sophisticated techniques that allow spatial resolution we characterized the nanostructure of such crystalline rock surfaces and the speciation of the actinide Cm(III) thereon.

Namely, we combined vertical scanning interferometry, calibrated autoradiography, and Raman microscopy coupled to µTRLFS (*micro-focus time-resolved laser-induced spectroscopy*).[1] Thus we were able to correlate mineralogy, topography, and grain boundary effects with radionuclide speciation, allowing us to identify important radionuclide retention processes and parameters.

Investigations focussed on granite from Eibenstock (Germany) and migmatised gneiss from Bukov (Czech Republic). Cm(III) sorption on the rock's constituing minerals - primarily feldspar, mica and quartz - was analyzed quantitatively and qualitatively. We observed that Cm(III) sorption uptake and speciation depends not only on the mineral phase, but also the surface roughness. An increasing surface roughness leads to higher sorption uptake and a stronger coordination of the sorbed Cm(III). On the same mineral grains sorption differed significantly depending if an area exhibits a low or high surface roughness. In case that one mineral phase dominates the sorption process, sorption of Cm(III) on other mineral phases will only occur at strong binding sites, typically where surface roughness is high. Areas of feldspar and quartz with high surface roughness additionally showed the formation of sorption species with particularly high sorption strength that could either be interpreted as Cm(III) incorporation species or ternary complexes on the mineral surface.

We conclude that in addition to mineral composition, surface roughness needs to be considered adequately to describe interfacial speciation of contaminants and respective retention patterns for the safety assessments of nuclear waste repositories.

[1] Molodtsov, Schymura, Rothe, Dardenne & Schmidt (2019), *Scientific Reports* 9, 6287.