

Uranium diffusion through Opalinus Clay in the absence and presence of citric acid

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Argillaceous rocks are investigated as potential host rocks of deep geological repositories for high-level waste (HLW). Uranium is one of the major components of spent nuclear fuel. In the case of water-mediated release of uranium from HLW containers, the uranium migration through an argillaceous host rock is governed by molecular diffusion due to its low permeability. In contrast to low / intermediate level waste (L/ILW), HLW does not contain significant amounts of organic compounds. Nevertheless, interactions with compounds such as citric acid (CC) have to be taken into account for L/ILW and HLW co-disposal concepts. Moreover, low molecular weight organic acids can be released by the organic matter fraction of natural clay rocks. Citric acid forms aqueous complexes with U(VI) but also with U(IV) and, consequently, can mobilize uranium. Batch sorption studies with Opalinus Clay (OPA) from Mont Terri, Switzerland, proved a decrease of U(VI) sorption with increasing CC concentration¹. The uranium / CC system acts very light-sensitive, meaning CC is able to reduce U(VI) to U(IV).

The aim of this study was to examine the potential U(VI) mobilization by CC during diffusion through OPA. For this, ²³³U(VI) diffusion experiments were conducted in the absence and presence of CC under anaerobic conditions and under exclusion of light. The experiments were run for three months ($c_0[^{233}\text{U(VI)}] = 1 \times 10^{-6}$ mol/L; $c_0[\text{CC}] = 1 \times 10^{-3}$ mol/L; pH 8.5; $I = 0.36$ mol/L OPA pore water). Sorption and transport parameter values were calculated based on Fick's laws by inverse modeling of the concentration profiles using COMSOL Multiphysics.

Diffusion parameter values for U(VI) in the absence of CC were in agreement with literature values for diffusing $\text{Ca}_2\text{UO}_2(\text{CO}_3)_3(\text{aq})^2$. In the presence of CC, U(VI) diffusion was significantly retarded, which can be attributed to a probable reduction to U(IV). Up to now, there is no thermodynamic data available for the observed diffusing U(VI)-CC species. This complex still needs to be described.

1. Schmeide and Joseph, *Annual Report 2011, HZDR-013*, **2012**, Helmholtz-Zentrum Dresden-Rossendorf, Dresden, p. 41.
2. Joseph, et al., *Geochim. Cosmochim. Acta*, **2013**, 109, 74.

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