

Migration behaviour of carboxylic acids in compacted illite, kaolinite and Opalinus Clay

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Owing to its long half-life and high mobility in the geosphere, ¹⁴C is an important contributor to the annual radioactive dose predicted for a low- and intermediate-level radioactive waste repository.

To address possible weak retardation processes, we investigated the migration behaviour of selected carboxylic acids (CAs) in compacted illite, kaolinite and Opalinus Clay (OPA, potential host rock for Swiss nuclear waste repositories) using an infiltration technique with a pulse injection of the CAs [1].

The pure clay samples (infiltration with NaCl; uncontrolled pH) exhibited symmetrical breakthrough curves with no tailing. Test solutes eluted from illite in the following order: ³⁶Cl, aliphatic CAs, HTO, β-OH-CAs, α-OH-CAs [1]. The early breakthrough of ³⁶Cl suggests an anion exclusion effect caused by the negatively charged illite surface. In kaolinite, the elution order was: HTO, aliphatic CAs, ³⁶Cl, β-OH-CAs. Breakthrough curves of all test solutes from OPA (infiltration with artificial pore water; Br⁻ as the reference tracer) were asymmetrical with tailing, which might be caused by the increased predominance of diffusion processes. Compared with Br⁻, test aliphatic CAs were not retarded in OPA. α-OH-CAs could not be recovered in the effluents from kaolinite and OPA within the time of observation.

Variations of salinity and pH were conducted in the experiments with pure clays to (i) compare to the OPA system, and (ii) to get further insight into the retardation mechanisms. First results on illite indicated an increasing retardation for all test solutes with increasing salinity. Therefore, the retardation in illite can not be explained by electrostatic interaction or ligand exchange. Different retention behaviours of D-Lactate and L-Lactate enantiomers in illite were observed. It could be concluded that the hydroxy group in the alpha position is involved in the retardation. The sorption of hydroxylated acids is hypothetically caused by chelation of the α-/β-hydroxy group with the metal cations on the clay surface.

[1] Chen Y, Glaus M.A., Van Loon L, Mäder U (2015) *Goldschmidt Abstracts*, **2015** 527.