Modelling diffusion of sorbing cations in Opalinus Clay

P. KREJCI[1]*, TH. GIMMI[2] AND L. VAN LOON[3]

[1]Paul Scherrer Institute, 5232 Villigen PSI, Switzerland (*correspondence: philipp.krejci@psi.ch)
[2]University of Bern and Paul Scherrer Institute, Switzerland
[3] Paul Scherrer Institute, 5232 Villigen PSI, Switzerland

Opalinus Clay is a claystone formation which is considered as potential host rock for disposal of radioactive waste. Transport of radionuclides through clay formations or clay barriers is determined by their diffusion and sorption properties. Experimental studies of cation diffusion in clays led partly to diffusion coefficients D_e much larger than those of water tracers, especially when sorption is large. This behavior, and especially the dependence of D_e on the pore water chemistry, cannot be described consistently by simple Fick's law. In tracer experiments, diffusion coefficients of Na and Sr are typically derived from the steady state phase of breakthrough fluxes, and the sorption distribution coefficient from the transient phase, applying a single species model with linear sorption. Here, we use a generalized multisite surface diffusion model[1] implemented in the reactive transport code FLOTRAN[2] for modelling the enhanced diffusion of Na and Sr in Opalinus Clay. This model was already successfully applied to Cs diffusion in Opalinus Clay[3]. It accounts for a mobility of sorbed cations, which can move along the negatively charged clay surfaces and contribute to the overall mass flux. The model combines pore and surface diffusion, with their relative importance depending on the chemical conditions, in one single diffusion coefficient. Sorption is modeled as ion exchange with ion- and sorption site-specific surface mobilities. Surface mobilities for Na and Sr were estimated by fitting the model to experimental data. The mobilities fall in the range found in [1], and are about independent of the pore water chemistry. Thus, it is possible to predict Na and Sr diffusion coefficients for different chemical conditions. Finally, a sensitivity analysis of the sorption parameters was carried out in order to evaluate their effect on tracer breakthrough.

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