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Cesium uptake by Ca/Mg bentonite: evaluation of sorption experiments by a multicomponent two-site ion-exchange model

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Understanding the migration processes in the near-field of the geological disposal of nuclear wastes at the basic level can help to develop models, which may be used in transport codes that can predict the migration of radioactive contaminants in the field scale. Cesium is well adsorbed by clay minerals and many studies concerning adsorption of cesium on different types of clay minerals have been published so far. As in the Czech project of deep underground repository the application of local bentonite is proposed, in this work, cesium adsorption on Czech Ca/Mg bentonite (Rokle deposit, NW Bohemia) has been studied. In our previous study two main differences between Rokle bentonite and worldwide studied Na-bentonites were observed: (1) the great influence of phase ratio m/V on the shape of Cs sorption isotherm in the range of higher Cs concentration, and (2) a substantial content of micas and mica-type clay minerals (about 19 wt. %) that are believed to cause the specific Cs sorption in the lower concentration range due to the presence of the Frayed Edge Sites (FES).

In this work, cesium sorption on Ca/Mg-bentonite of the Czech origin was studied using batch technique for wide ranges of both bentonite-to-water ratio (m/V) and initial concentration of CsCl (10^{-7} – 0.1 mol/l), isotope Cs-137 was used as a tracer. The total cation-exchange capacity of the bentonite was 0.50 equiv/kg. The experiments revealed non-linear character of cesium sorption, substantially influenced by the m/V . The numerical evaluation of batch experiments performed with the use of PHREEQC, in which exchange reactions between added Cs⁺ and Mg²⁺, Ca²⁺, Na⁺ and K⁺ ions sorbed initially on the bentonite were taken into account, enabled to determine selectivity coefficients of all mentioned cations, according to the Gaines-Thomas convention. The model describing exchange reactions mentioned was successfully verified on a broader set of experimental data that was previously interpreted by a simple ion-exchange model taking into account the exchange of cesium with an unspecified divalent cation. The shape of equilibrium isotherms indicated for initial concentrations of cesium smaller than 0.001 mol/l the possibility to describe the cesium uptake on Ca/Mg-bentonite by a two site model. Using this assumption, capacity of the second type of sites with higher selectivity to cesium, was calculated as about 0.01 equiv/kg. This specific cesium sorption capacity, which is generally believed to be associated with the frayed edge sites (FES) of illitic materials, was also measured using AgTU method. The values resulting from both the methods were of the same order, differences were discussed. The first results of 1D transport modelling in the PHREEQC environment respecting the developed multicomponent ion-exchange model could help by the interpretation of results of diffusion transport of some cations in the layer of compacted bentonite.

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