NRC-8, EuCheMS International Conference on Nuclear and Radiochemistry



Contribution ID: 116

Type: Poster

Trivalent Actinide/Lanthanide Sorption under Saline Conditions

Wednesday, 19 September 2012 18:00 (1h 50m)

Sorption processes on mineral surfaces play an important role in the retardation behaviour of radionuclides and have to be considered in performance assessment calculations. However, almost all available experimental data are restricted to low ionic strength conditions. Saline conditions are not restricted to solutions relevant to rocksalt formations. Elevated salt concentrations up to 6.5 M are also found in sedimentary rocks which are under investigation within nuclear waste disposal programs.

In the present work, experiments have been performed with quartz and illite. In all cases sorption of Eu(III) radiolabelled with Eu-152 was studied in batch experiments, where ionic strength was varied up to 5 M NaCl. Experiments with Cm(III) and the time resolved laser fluorescence spectroscopy (TRLFS) are directed to obtain detailed information on the metal ion speciation at the mineral surfaces as a function of ionic strength.

In all systems we observed a decreasing sorption with increasing ionic strength. While for quartz only innersphere surface complexation is expected for Eu(III), outer sphere sorption at low pH must be considered for illite.

Eu(III) sorption to illite was calculated by a slightly modified well established non-electrostatic sorption model, developed and tested so far only for lower ionic strength conditions (2SPNE/CE). The Pitzer formalism was used to calculate activity coefficients for aqueous species. Model calculations for different ionic strengths yield fair agreement with the experimental data. This finding suggests that the shift in sorption edges with increasing ionic strength is basically due to a variation of ion activities and a concomitant change in aqueous metal ion speciation.

TRLFS experiments with Cm(III) indicate no significant differences for sorbed inner-sphere Cm(III) complexes at low and high ionic strength conditions. Our preliminary study indicates that the modified non-electrostatic 2SPNE/CE model can be applied to predict sorption of trivalent actinide ions also for elevated ionic strength conditions at least in NaCl dominated solutions.

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Session Classification: Poster Session

Track Classification: Radioactive elements in the environment, radiation archeometry and Health Physics