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Colloid facilitated transport of lanthanides through discrete fractures in chalk

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Geological disposal of high-level radioactive waste is the internationally agreed-upon, long term solution for the disposal of long lived radionuclides and spent fuel. Eventually, corrosion of the waste canisters may lead to leakage of their hazardous contents, and the radionuclides can ultimately make their way into groundwater and pose a threat to the biosphere. Engineered bentonite barriers placed around nuclear waste repositories are generally considered sufficient to impede the transport of radionuclides from their storage location to the groundwater. However, colloidal-sized mobile bentonite particles eroding from these barriers have come under investigation as a potential transport vector for radionuclides sorbed to them. In addition, the presence of organic matter in groundwater has been shown to additionally facilitate the uptake of radionuclides by the clay colloids.

This study aims to evaluate the transport behaviors of radionuclides in colloid-facilitated transport through a fractured chalk matrix and under geochemical conditions representative of the Negev desert, Israel. Lanthanides are considered an acceptable substitute to actinides for research on radionuclide transportation due to their similar chemical behavior. In this study, the migration of Ce both with and without colloidal particles was explored and compared to the migration of a conservative tracer (bromide). Tracer solutions containing known concentrations of Ce, bentonite colloids, humic acid and bromide were prepared in a matrix solution containing salt concentrations representative of that of the average rain water found in the Negev. These solutions were then injected into a flow system constructed around a naturally fractured chalk core. Samples were analyzed for Ce and Br using ICP-MS, and colloid concentrations were determined using spectrophotographic analysis. Breakthrough curves comparing the rates of transportation of each tracer were obtained, allowing for comparison of transport rates and calculation of overall tracer recovery.

Preliminary results suggest that mobility of Ce as a solute is negligible, and in experiments conducted without bentonite colloids, the 2% of the Ce that was recovered during the experiments travelled as "intrinsic" colloids in the form of $Ce_2(CO_3)_3 \cdot 6H_2O$ precipitate. However, the total recovery of the Ce increased to 9% when it was injected into the core in the presence of bentonite colloids and 13% when both bentonite and the carbonate precipitate colloids were injected. In addition, the maximum relative concentration (C/C_0) of the Ce in the samples from the experiments conducted without bentonite colloids is about 0.002, whereas that of the experiments conducted in the presence of bentonite colloids reaches almost 0.2. This indicates that colloid presence does indeed markedly increase the mobility of radionuclides through fractured chalk matrices and should therefore be considered in models representing transport of radionuclide waste originating from nuclear repositories.