

Thermodynamic and kinetic properties of U(VI) sorption on chalk substrates

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The mobility of uranium (U), a radioactive element, is one of the major concerns in safety risk assessments of sites exposed to mining and reprocessing. Leaching of U from the contaminated tailings may result in aquifer pollution in absence of bio-reductive immobilization. Therefore, effective remediation practices require mechanistic understanding of the long-term behaviour of U in porous aqueous media.

Here, we present experimental and modeling results of thermodynamic and kinetic properties of U(VI) sorption on two natural carbonated substrates: chalky soil and pure chalk. Batch experiments were performed using a system of reactors at controlled temperatures of 5°C, 12°C, 21°C, and 30°C. The equilibrium constants of calcium-carbonate-uranium complexes [2] were used to update the database adapted from Guillaumont *et al.* [1] and calculate U speciation using PHREEQC. The model indicates the key role of carbonate ions in U complexation and mobility in studied suspensions.

Calibration of the experimental data on U(VI) sorption as a function of temperature and time was performed according to the modified multi-reaction model of Zhang and Selim [3]. Results were used to compute thermodynamic properties, such as, distribution coefficients, activation energy, enthalpy, and entropy of reactions. The output of this experimental and modeling work will be used in a reactive transport model HYTEC for better estimation of U migration through soil and carbonate aquifer.

[1] Guillaumont, R.; Fanghanel, T.; Fuger, J.; Grenthe, I.; Neck, V.; Palmer, D.; Rand, M. H. *Update on the Chemical Thermodynamics of Uranium, Neptunium, Plutonium, Americium and Technetium*. (2003) Elsevier: Amsterdam.

[2] Dong, W.; Brooks, S.C. (2006) *Environ. Sci. Technol.*, **40**, 4689-4695.

[3] Hua Zhang and H. M. Selim. *Environ. Sci. Technol.*, **39**, 6101-6108.