

Uranium(VI) sorption to bentonite.

Samer Bachmaf , Britta Planer-Friedrich , Broder J Merkel

TU Bergakademie Freiberg, Lehrstuhl für Hydrogeologie, Gustav-Zeuner-Str. 12, 09596 Freiberg, Germany

Uranium U(VI) sorption to bentonite was investigated as a function of pH, sulphate, carbonate, arsenate, and phosphate. Uranium sorption on bentonite was more effective at lower uranium concentrations with a maximum sorption at pH 5 (>90%) and a minimum sorption at pH 3 (70%) in the absence of complexing ligands. In the presence of sulphate, uranium sorption was observed to decrease significantly under acidic conditions (45%). While cation exchange predominates sorption mechanisms on bentonite under acidic conditions, modelling showed that the presence of sulfate lead to formation of a zero-charged uranyl-sulphate complex, thus explaining the decrease in sorption. With carbonate in solution, $\text{UO}_2(\text{CO}_3)_2^{2-}$ and $\text{UO}_2(\text{CO}_3)_3^{4-}$ complexes were modelled to form. In contrast to the $\text{UO}_2(\text{OH})_3^-$ complex that predominates in the absence of carbonate, these complexes showed almost no sorption to bentonite (10%). In the presence of phosphate, uranium sorption on bentonite was significantly enhanced to almost 100%, probably due to the formation of ternary $\text{UO}_2\text{HPO}_4^0$ or UO_2PO_4^- complexes. Kinetic batch experiments revealed that the sorption rate was generally rapid over the first 10 minutes of the experiments and then slowed down.

Similar to phosphate, increased uranium sorption was observed in the presence of arsenate. While uranyl-arsenate complexes were shown before to form under acidic conditions up to a pH of 5, evidence for their formation under neutral to alkaline conditions, analogous to uranyl-phosphates, e.g., as $\text{UO}_2\text{AsO}_4^-$ and $\text{UO}_2(\text{AsO}_4)_2^{2-}$ complexes, is new. Chromatographic separation of the uranium-arsenate mixed solutions by anion-exchange chromatography coupled to inductively-couple plasma mass-spectrometry confirmed that only 10% of the arsenate originally present still remained after addition of uranium, while 90% were converted to an unknown species not detectable with our current chromatographic separation methods. All our studies were carried out in sodium-containing solutions. In the absence of sodium, no chromatographic evidence for complexation was found, which might advocate the formation of sodium-uranyl-arsenate complexes instead of pure uranyl-arsenate complexes.