

Sorption of Tetravalent Plutonium onto Kaolinite in the Presence of Humic Substances

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Humic substances (HS), a mixture of organic macromolecules, are distributed ubiquitously in the ecosphere and play an important role in the interaction processes with actinide ions and clay minerals. The ternary system plutonium-kaolinite-humic substance is studied here in order to understand and predict the speciation and migration behavior of plutonium in a geogenic aquatic system.

The influence of HS on the sorption of Pu(IV) onto kaolinite was investigated as a function of pH, contact time, concentration of HS, and adding sequences of the reactants. The sorption studies were performed by batch experiments under aerobic conditions. A pH range of 1-9 was studied with Pu(IV) concentrations of 7.1×10^{-8} M, 0-100 mg/L of HS, and a solid phase concentration of 4 g/L kaolinite.

The ternary experiments were performed by using three adding sequences. In sequence 1, all reactants were mixed at the same time; in sequence 2, a pre-equilibrium of metal ion and kaolinite was achieved and then the HS solution was added; in sequence 3, a pre-equilibrium of kaolinite and HS was achieved; then the Pu(IV) solution was added. They all indicated that the systems reach a sorption equilibrium within a few hours after addition of Pu(IV) and fulvic acid (FA) to the pre-equilibrated kaolinite suspension. Comparing the different adding sequences of the reactants, no significant difference in the Pu(IV) sorption behavior was found.

The presence of FA influences significantly the sorption of Pu(IV) onto kaolinite. This was studied at the pH 1, 3, 6, and 9. The pH dependence of sorption onto kaolinite in varying adding sequences (1, 2, 3) was studied at a FA concentration range of 10-100 mg/L, see Figure 1. In general, Pu sorption onto kaolinite decreases when FA concentration increases regardless of the pH and adding sequence of the reactants [1]. This can be explained by the increased number of binding sites with increasing FA concentration in the solution. Particularly striking is the strong decrease of sorption at pH 1.

It is interesting that the adding sequence of the reactants in the ternary system has almost no influence on the sorption of Pu(IV) onto kaolinite whereas a significant influence on the sorption of Pu(III) onto kaolinite has been found [2].

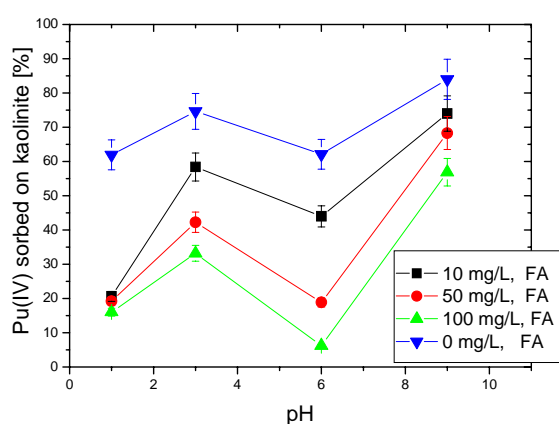


Figure 1: Influence of FA on the sorption of Pu(IV) onto kaolinite as a function of pH; adding sequence 3.

The minimum in the sorption of Pu(IV) onto kaolinite around pH 6 that had already been found in the binary system Pu(IV)-kaolinite and had been shown to be due to oxidation of Pu(IV) to Pu(V) is also present in the ternary system with FA. A similar behavior is also found in the case of humic acid.

References

- [1] Banik. N. L., Doctoral thesis, Institut für Kernchemie, Universität Mainz, (2006)
- [2] Buda. R. A., Doctoral thesis, Institut für Kernchemie, Universität Mainz, (2006)