

Title: THE APPLICATION OF SURFACE COMPLEXATION MODELING TO THE ADSORPTION OF URANIUM(VI) ONTO HEMATITE IN THE PRESENCE OF HUMIC AND FULVIC ACIDS (METAL SPECIATION)

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Abstract: Uranium is a common element in ground and surface waters in the western United States. It exists in oxidation states from III to VI, although U(VI) is the dominant oxidation state in surface and oxic groundwaters. Historically, negligent handling of spent tailings and leached ore has led to high levels of uranium in production water at many mining and milling operations. When including sites contaminated by nuclear weapons production and handling with these contaminated mining sites, the number of Superfund sites currently proposed or listed on the NPL that have uranium concentrations exceeding the proposed regulatory limit would be at least 25. Assessing the fate of uranium at these sites and developing innovative strategies for remediation hinge on a thorough understanding of the chemical form (speciation) of uranium. Fundamental to this understanding is expanding our knowledge regarding the role of mineral interfaces, natural organic matter (NOM) and colloids in regulating uranium speciation and fate in heterogeneous systems.

The fate of metals in aquatic systems is a function of: (1) metal speciation and (2) the processes that act to transport those metals. In most aquatic systems, species of dissolved organic carbon, like humic and fulvic acids, constitute an important pool of ligands for complexing metals. In spite of the fact that dissolved natural organic matter (NOM) is ubiquitous in aquatic environments, the details of the role of NOM in affecting metal speciation are still unquantified. The determination of the effects of NOM on the speciation and migration of trace metals, like uranium, is critical to the development of accurate models for metal solubility and transport.

In this study, the adsorption of uranium(VI) onto hematite in the presence of Suwannee river humic (HA) and fulvic acid (FA) is studied. The objectives of this study were to: (1) evaluate the speciation of uranium in systems with a mineral phase and different MW fractions of NOM and (2) develop a model to account for these interactions.

Experimental results of batch sorption, ion exchange, electrophoretic mobility and potentiometric titration experiments are discussed. HA was found to form stronger complexes with U(VI) than FA. The HA and FA complexation of U(VI) was interpreted using Schubert's method and FITEQL as a mixture of 1:1 and 1:2 complexes. For the systems studied, HA sorbed more readily than FA on a mass basis, but sorbed equally on a molar basis. In the ternary systems, both HA and FA enhanced the sorption of U(VI) by hematite.

These results allow for the systematic characterization of the "binary" systems (U/hematite, U/HA, U/FA, FA/hematite and HA/hematite), the "ternary" systems

(U/hematite/FA and U/hematite/HA) and the development of equilibrium models. To include the reactions of HA and FA with protons, uranium(VI) and mineral surfaces into equilibrium geochemical models, a discrete ligand, non-electrostatic model was used. This approach was integrated with surface complexation modeling theory in order to account for all chemical interactions in a consistent mathematical fashion. The end result of this study is a modeling approach that allows for the complete simulation of heterogeneous ternary systems with metals, mineral surfaces and natural macromolecules.