

Retention of U(VI) onto silica in presence of model organic molecules

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Abstract

It is well-known that the organic matter influences the retention of ions onto mineral surfaces. However, the major part of concerned studies implies humic substances and complex solids. Another approach for identifying the sorption mechanisms is possible by studying simpler solids than those present in natural medium. So, silica is chosen as mineral surface because of its abundance in soils and of the presence of Si-O groups in clayey minerals. Uranium (VI) is selected as cation. Simple organic molecules like acetic (one carboxylic group) and oxalic (two carboxylic functions) acids are considered as models of the natural organic matter for understanding their role in the retention of U(VI) onto powders and slides of silica.

Binary (organics/silica, U(VI)/silica) and ternary systems (organics/silica/U(VI)) are studied by complementary approaches. Sorption edges as function of pH are obtained by liquid scintillation methods and capillary electrophoresis. Different spectroscopic techniques are used to deduce the interactions between the organic matter and U(VI) sorbed onto the silica whose: Time-Resolved Laser-induced Fluorescence Spectroscopy (TRLFS), X-ray Photoelectron Spectroscopy (XPS), Nuclear Microprobe Analysis (NMA).

The results of the effect of these model organic molecules onto the U(VI) retention showed a good agreement between the different techniques. Concerning the acetic acid, there are not differences in the sorption percentages of uranyl (see the figure). All these results indicate that the uranyl-acetate complexes stay in the aqueous solution rather than sorbing onto the silica.

On the contrary, oxalic acid influences the sorption of U(VI) onto the silica surface. The sorption percentage of U(VI) in the ternary system (oxalic acid/silica/U(VI)) is lower than the binary system (U(VI)/silica) (see the figure). So, the presence of oxalic acid decreases the sorption of U(VI) onto the silica surface.

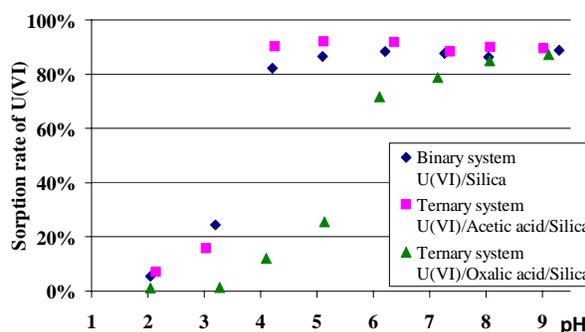


Figure 1: Sorption percentage of U(VI) onto silica with the model organic molecules

Moreover, the spectroscopic results show that there are not the same U(VI) chemical environment for the two systems (silica/U(VI) and silica/U(VI)/oxalate). The TRLFS results evidence an additional short lifetime for U(VI) observed in the ternary system silica/U(VI)/oxalate. The trace of sorbed uranyl-oxalate complexes is also observed by XPS spectroscopy, from the shift of 0.6 eV in the binding energy of the U4f signal. Besides, the results of NMA confirm the observations of the previous techniques, so that the presence of uranium onto silica is always associated to that of carbon.

Other techniques, as Attenuated Total Reflexion Fourier Transform Infra-Red spectrophotometry and Zeta-potential measurements will be used for determining of the structure of the organic and U(VI) sorbed complexes onto the silica surface (outer or inner sphere complexes).

Further work is under investigation with the same approach to study the effect of other model compounds (containing organic functions present in natural organic matter, as alcohols and carboxylic acids), as well as to apply this methodology to humic acids and to natural organic matter present in the Callovo-Oxfordian sediment in the northeast of the Paris Basin (Bure).