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Complexation of U(VI) with nitrogen and phosphorous containing ligands in aqueous solution

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The long-term safety assessment of nuclear waste disposals requires detailed knowledge of the transport and interaction behavior of actinides within the technical, geotechnical and geological barriers. In the case of accidental release of radionuclides into the surrounding environment, their migration behavior can be effectively influenced by the ambient conditions like pH, temperature, ionic strength and especially the presence of inorganic and organic complexing ligands, e.g. bioligands like humic acids, amino acids or phospholipids. Due to their high complexing capacity towards metal ions these ligands may influence the mobility of actinides.

We studied the U(VI) complexation with nitrogen and phosphorous containing model ligands in order to simulate the functionalities of bioligands. The aim of this work is to determine the influence of various nitrogen and phosphorous containing functional groups on the U(VI) complexation and to evaluate their contribution in comparison to oxygen containing functional groups. As nitrogen containing ligands we used the biologically important substances anthranilic acid (AA) and nicotinic acid (NA). Furthermore, phenylphosphonic acid (PPA) was used as aromatic phosphorous containing ligand.

The complexation studies in aqueous solution were performed as a function of pH using time-resolved laser-induced fluorescence spectroscopy (TRLFS). All ligands show a complex formation with U(VI) under the given experimental conditions ($[U(VI)] = 0.05 \text{ mM}$, $[ligand] = 0 - 0.5 \text{ mM}$, $pH \text{ 2-4}$, $I = 0.1 \text{ M}$, $T = 25^\circ\text{C}$). The binding of U(VI) by AA and NA takes place via the carboxylic group of the ligands. It can be concluded, that oxygen containing functional groups dominate the U(VI) complexation in the investigated pH range. The nitrogen functionalities of the ligands play only a subordinate role. AA forms a 1:1 complex, with a corresponding stability constant of $\log \beta_{11} = 3.14 \pm 0.17$ [1]. The formation of 1:1 and 1:2 complexes was detected for the U(VI)-nicotinate system ($\log \beta_{11} = 3.73 \pm 0.30$, $\log \beta_{12} = 7.46 \pm 0.17$ [1]) as well as for the U(VI)-phenylphosphonate system ($\log \beta_{11} = 3.58 \pm 0.17$, $\log \beta_{12} = 6.81 \pm 0.10$). At high U(VI) concentrations (1 mM), a precipitation of the formed complexes was observed for the U(VI) complexation by AA and PPA. The precipitates were analyzed with FT-IR spectroscopy and X-ray diffraction. To study the influence of different functional groups on the complexation of actinides in different oxidation states, comparable complexation studies are performed with Am(III).

References:

[1] B. Raditzky, K. Schmeide, S. Sachs, G. Geipel, G. Bernhard, *Polyhedron* (2009), doi:10.1016/j.poly.2009.08.010.

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