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Complexation of Th(IV) and U(VI) with a new tetrahydroxamic acid by affinity capillary electrophoresis

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The transfer of metal cations in soils and sediments is not only controlled by geological and physico-chemical parameters (clay and organic matter contents, pH, Eh, etc.) but also by microorganisms. The latter take part in the mobilization/immobilization of trace metals [1]. Siderophores, as natural iron-specific chelators, have to be considered in this context. These low molecular weight, water-soluble compounds are excreted by bacteria and fungi to overcome the limited bioavailability of iron under aerobic conditions (ca. 10^{-18} M at neutral pH) by dissolving iron oxohydroxides present in the soils. Because of their high affinity for hard Lewis acid cations, like Fe(III), hydroxamic siderophores are also able to effectively bind actinides (U, Th, Pu) [2-4]. The development of high affinity chelators for f-elements and the understanding of their coordination chemistry is of fundamental interest for the management and remediation of contaminated fields or the disposal of nuclear wastes [4].

This work is devoted to the complexation study of Th(IV) and U(VI) with (DFO(Me)PIPO)₄ (Figure 1), a new hydroxamic acid obtained by coupling the 6-membered 1-hydroxypiperidine-2-one moiety [5] to the bacterial siderophore desferrioxamine B [3,4]. This compound, which contains four hydroxamic acid groups, is an effective ligand of Zr⁴⁺ [6], as well as actinides, such as Th⁴⁺ and UO₂²⁺ even at pH values of about 2.

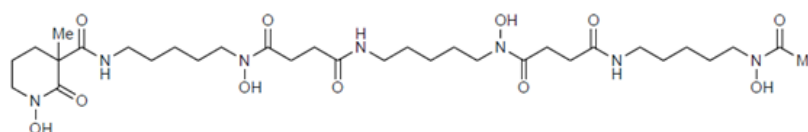


Figure 1. Structure of (DFO(Me)PIPO)₄.

Protonation constants of (DFO(Me)PIPO)⁴⁻ were obtained by potentiometry and UV absorption spectrophotometry, before unravelling the speciation of the ligand in the presence of Th⁴⁺ and UO₂²⁺ by affinity capillary electrophoresis (ACE). This electromigration technique, which requires only a few μ L of solutions, is based on the change in the electrophoretic mobility of the UV-detected analyte due to the interaction with the other species (metal or ligand) present in the electrolyte. These sets of runs were performed using (H,Na)ClO₄ mixtures to keep both the pH and the ionic strength ($I = 0.1$ M) constant. The electrophoretic mobility changes with the increasing concentrations of the reagent dissolved in the background electrolyte solution were used to assess the speciation and equilibrium constants. This method is fruitful for studying metal complex formation equilibria and gives valuable information for further modelling of their behaviour under environmental conditions [7]. Formation of 1:1 metal-to-ligand complexes was further ascertained by high-resolution ESI-MS spectrometry and circular dichroism (CD) measurements using the optically pure ligand.

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