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Study of Th(IV) complexation with hydroxamic acids by affinity capillary electrophoresis

Emanuele ZANDA,^{a)} **Veronika ZINOVYEVA,**^{a)} **Vladimir SLADKOV,**^{a)}
Michel MEYER,^{b)} **Jean-Claude CHAMBRON**^{c)}

^{a)} *Laboratoire de Physique des 2 Infinis Irène Joliot Curie (IJCLab), UMR 9012, CNRS/IN2P3 Université Paris-Saclay, 15 rue Georges Clemenceau, 91405 Orsay, France*

^{b)} *Institut de Chimie Moléculaire de l'Université de Bourgogne (ICMUB), UMR 6302, CNRS, Université Bourgogne-Franche-Comté, 9 avenue Alain Savary, BP 47870, 21078 Dijon Cedex, France*

^{c)} *Institut de Chimie de Strasbourg, UMR 7177, CNRS, Université de Strasbourg, 1 rue Blaise Pascal, BP 296 R 8, 67008 Strasbourg Cedex, France*

emanuele.zanda@ijclab.in2p3.fr

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]. Therefore, the presence of actinide(IV) (Th, Pu) radionuclides after accidental discharges raises the question of their transfer in the environment and the resulting risk of food chain contamination. Siderophores, as natural iron-specific chelators, have to be considered in this context [2]. 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.

This work is devoted to the complexation study of Th(IV) with a number of commercially available simple hydroxamic acids (Figure 1a) and of (DFO(Me)PIPO)₄ (Figure 1b), a new tetrahydroxamic acid obtained by coupling the 6-membered 1-hydroxypiperidine-2-one moiety to the bacterial siderophore desferrioxamine B [3]. These compounds are effective ligands in capturing and transporting actinides [4].

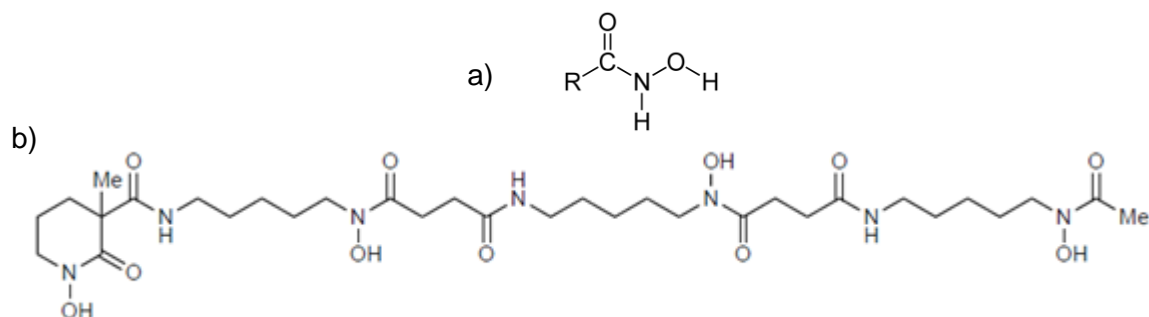


Figure 1. a) General formula of simple hydroxamic acids; b) structure of (DFO(Me)PIPO)₄.



Affinity capillary electrophoresis (ACE), which requires only a few μL of solutions, with UV detection was used to unravel the speciation of these systems. ACE is based on the change in the electrophoretic mobility of the detected species due to the interaction with other species present in the electrolyte. Two sets of runs were performed using (H,Na)ClO₄ mixtures at constant pH and ionic strength. On a first approach, the background electrolyte (BGE) contained varying amounts of Th(IV), while the second approach consisted in analysing solutions containing increasing amounts of ligand in the BGE. The electrophoretic mobility changes with the increasing concentrations of the reagent dissolved in the BGE solution was 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 [5].

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