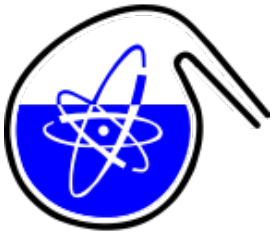

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Complexation of americium(III) with citric acid : role of the hydroxo function ?

Tuesday, 15 May 2018 09:00 (15 minutes)

Alpha-hydroxycarboxylic acids are important molecules in nuclear field. They can be used as complexing agent or buffer in aqueous phase in actinide selective separation processes. These molecules are also present in the environment as a degradation product of cellulose. Therefore the alpha hydroxyl-carboxylate ligands are likely to influence the speciation in the fuel cycle processes and in the migration mechanisms. In this double context, the behavior and complexation of americium with this type of ligand was studied, taking citric acid as a representative of this family.

Some studies have already been published on actinide(III)-citric acid complexation, but most of them are incomplete because the citric acid is also one of the most complicated compounds of the family due to its three carboxylic acids able to be deprotonated and to coordinate the metallic cation. On one side, the thermodynamic studies provide only the stoichiometry of the different complexes without any information on the coordination spheres (the number of complexing carboxylates, the presence or not of the hydroxo function as a coordinating group and the ligand protonation state). On the other side, the rare structural studies relate to polynuclear or polymeric species unlike the complexes identified in solution in the previous thermodynamic studies.

In this study, thermodynamics and structural data were assessed in solution for the same chemical conditions to finally describe the speciation of the Am(III)-citrate system. A focus was done on the role played by the hydroxyl group: does it coordinate the actinide? Is it protonated? To better understand its role, the Am-citrate system was compared to the Am-tricarballylate system (analogue to citric acid without the hydroxyl group). The two systems were studied at pH = 1 and 3. The complexation constants have been determined and, different complexes were characterized using X-ray absorption, NMR, TRLFS and capillary electrophoresis.

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