

## Lanthanide and actinide inorganic complexes in natural waters. TRLIFS and ESI-MS studies

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Actinide chemistry in natural environments is of great interest to predict radionuclides migration from possible radioactive waste repositories. To determine stoichiometries and thermodynamic stabilities we have studied actinide interactions with inorganic ligands of groundwater such as carbonate, hydroxide and sulfate. Many data have been validated in the NEA-TDB critical reviews (Thermodynamic Data Base project of the Nuclear Energy Agency OECD) including data from our laboratory. Beside the lack of available data at temperatures above 25°C, the NEA-TDB have pointed out discrepancies in the literature, so that there is a need of confirmations. Typically supplementary experimental work is still needed to find out whether other stoichiometries of actinide species could be stable under environmental conditions, particularly hydroxocarbonate and hydroxosulfate mixed complexes. For this purpose, our laboratory has developed time-resolved laser-induced fluorescence spectroscopy (TRLIFS) as a speciation technique with lower detection limits than classical ones. Moreover, whereas TRLIFS is restricted to a few fluorescent elements, electrospray-ionisation mass spectrometry (ESI-MS) enables detection of any element and has to be validated as a new tool for analyses of inorganic trivalent and tetravalent metal ion complexes. Here we present our experimental data on Eu(III) and Cm(III) using TRLIFS. They provide quantitative results on carbonate complexation in good agreement with data on Am(III). An interesting point is to decide whether the limiting complex is  $M(\text{CO}_3)_3^{3-}$  or  $M(\text{CO}_3)_4^{5-}$  ( $M=\text{Eu}$  or  $\text{Cm}$ ). Both interpretations are tested on Eu(III) and discussed. This study has been extended to different temperatures from 10 to 50°C and other inorganic ligands. An investigation of La(III) hydroxosulfate complexes has been performed by ESI-MS.

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