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Actinides behavior in solution with polyaminocarboxylates cyclic ligands

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NMR spectroscopy is one of the most relevant and widespread techniques that provides structural information for solid or liquid samples. It can be applied in many fields from small molecule analysis in organic chemistry to large molecule protein analysis in biology. Recently, with the advent of “nuclearized” spectrometers, it has proven useful in nuclear chemistry as well. The presence of an actinide cation, with 5f electrons, generates a modification of the NMR spectrum i.e. a broadening and/or a shift of the chemical shift, which is characteristic of the cation paramagnetism. The structure of the complex in solution can then be deduced by the ^1H NMR spectrum. The total experimental shift (Δ_{tot}) depends on the paramagnetic shift (δ_{para}) whose equation can be solved by magnetic susceptibility measurements (equations a and b).

Quantification of the paramagnetic contribution involves measurement of magnetic susceptibilities, accessible by NMR spectroscopy, and determination of two parameters that depends on the electronic configuration of the metallic cation. Bleaney and Golding have determined and calculated these constants for lanthanide cations. For actinide cations (U, Np, Pu), they are unknown but first molar magnetic susceptibilities have been determined (equation b) for the free ion and the An – dipicolinate (DPC) complex.

For a better and more complete understanding of the magnetic behavior of 5f electrons, the present study is focused on the comparison between experiment and theory. Two areas of study are undertaken. First, a magnetic susceptibilities study of actinide (IV and VI) complexes in concentrated hydrochloric and nitric acid solutions (up to 9 M) were performed to complete literature data. Second, a study of the influence of the change in ligand symmetry, specifically from DPC to the DOTA ligand, for actinides (III and IV) are ongoing.

