Effect of ionizing radiation on actinide-ligand complexes

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x oral ☐ poster

In nuclear fuel reprocessing, the organic ligands in solution undergo multiple attacks: chemical, thermal, but especially radiolytic. This complexity is reinforced by the biphasic nature of the chemical system and the presence of numerous solutes in the aqueous and organic phase [1]. These phenomena lead to a modification of the composition of the solution and to an alteration of the extraction and complexation properties in terms of efficiency and selectivity. However, very few studies report the effect of the radiation on the molecular speciation of complexes in solution.

The aim of this work is the investigation of the effect of radiolysis on the speciation of the actinides – ligand complexes in solution. Ligands with amide functional groups are chosen for this study. A molecular characterization of the alpha emitter-actinide (such as Pu, Am) – ligand complexes in separation processes was performed and the effect of the in-situ radiolysis (irradiation coming directly from the actinide included in the solution) on the speciation was investigated (Figure 1). To determine if the linear energy transfer (LET) of the radiation impacts ligand degradation and if this in turn impacts the complexation, ligands were also irradiated by a gamma source. After irradiation, extraction experiments were then performed to determine if gamma radiolysis of the ligand would impact actinide complexation differently than alpha radiolysis. Several complementary spectroscopic techniques were used such as UV-visible spectrophotometry and electrospray ionization mass spectrometry, which can also be used to suggest possible changes in metal speciation [2].

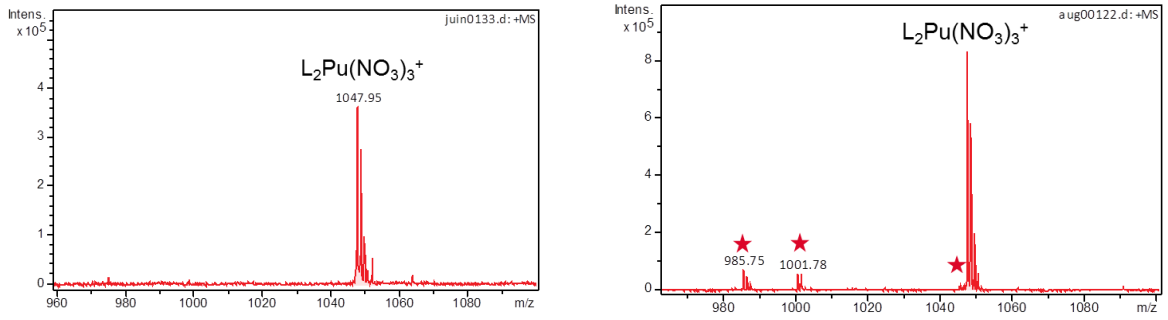

Fig. 1: ESI-MS spectra (positive ionization mode) of the Pu-N,N-dialkylamide organic phase solution before and after radiolysis. a. day 1, 0 kGy; b. day 69, 603 kGy. 970-1100 m/z mass range. Dilution 1/30th in acetonitrile. L being a N,N-dialkyl amide ligand. Stars indicate the appearance of new Pu species.

[1] Berthon, L. and Charbonnel, M.-C., in Ion Exch. Solvent Extr., ed. B. Moyer, CRC Press Taylor & Francis Group, Boca Raton, London, New York, (2010), vol. 19, pp. 429-513.

[2] Drader, J. A.; Boubals, N.; Cames, B.; Guillaumont, D.; Guilbaud, P.; Saint-Louis, G.; Berthon, L., Radiolytic stability of N, N-dialkyl amide: effect on Pu(IV) complexes in solution. Dalton Trans. (2018), 47 (1), 251-263