

Radiolytic effects on actinide(III) complexation with a hydrophilic PyTri ligand

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Summary. — The selective Am and Cm recovery from the highly active raffinate could be achieved by innovative-Selective ActiNide EXtraction (i-SANEX) process downstream major actinides removal. The TetraOctyl-DiGlycolAmide (TODGA) extractant and a hydrophilic complexing agent, 2,6-bis[1-(propan-1-ol)-1,2,3-triazol-4-yl]pyridine (PTD) being the most promising one, could be synergically used to separate Am and Cm from chemically similar lanthanides. Besides promising selectivity and efficiency for Am and Cm extraction, such systems must prove radiolytic and hydrolytic stability, given the high radionuclide content of the feed. In this work, PTD selectivity for trivalent Am and Cm towards Eu, chosen as representative lanthanides, has been studied at relevant conditions by different spectroscopic techniques. In order to mimic the process conditions, solutions of PTD were γ -irradiated up to 200 kGy by a ⁶⁰Co source or aged up to 100 days in 0.44 mol/L HNO₃ before complexometric titration experiments. The obtained UV-Vis and Time-Resolved Laser Fluorescence Spectroscopy (TRLFS) results were compared with those of fresh PTD solutions, in order to evaluate the impact of radiolysis and hydrolysis on metal-ligand complex formation. In accordance with the stability outlined by previous studies, also the complexation capabilities resulted to be unaltered. These results further support PTD as a valid candidate for partitioning processes.

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1. – Introduction

The reduction of volume and hazards of the high-level waste as well as the optimization of natural resources exploitation and final repository environmental footprint must be pursued by a careful management of long-term radiotoxic U, Np, Pu and Minor Actinides (MA: Am and Cm) contained in the Spent Nuclear Fuel (SNF) [1,2]. The recovery of major actinides (U and Pu) from the SNF has been successfully accomplished by industrially mature processes, as Plutonium and Uranium Reduction Extraction (PUREX) like processes [3-6]. Then, the recovered U and Pu are currently recycled as Mixed OXide (MOX) fuel. Following such PUREX-like processes, a Highly Active Raffinate (HAR) solution is obtained. Since it contains radioactive MA, fission and corrosion products, its long-term safety management is challenging, due to high decay heat and radiotoxicity. Partitioning and transmutation of MA is a promising strategy to deal with such hazardous legacy. First, the approach entails the separation (*i.e.*, partitioning) of MA from the HAR solution. Afterwards, with a view to obtaining shorter-living or even stable nuclides (*i.e.*, transmutation), the recovered MA would be used to fabricate a more complex fuel to be burned in advanced nuclear reactors. In the last decades, researchers' efforts and joint research projects funded by the European Commission have been dedicated to developing hydrometallurgical processes capable of efficiently and selectively recovering MA for their multi-recycling in advanced nuclear system [7-11]. Among all, the promising innovative-Selective ActiNide EXtraction (*i*-SANEX) process would allow the selective separation of Am and Cm from the chemically similar lanthanide fission products, as they are neutronic poisons that would hinder the MA transmutation [12]. In the first step of the *i*-SANEX process, trivalent MA and Ln are extracted from the acidic HAR aqueous feed by a lipophilic extractant, such as N,N,N',N'-TetraOctyl DiGlycolAmide (TODGA) [13, 14]. Afterwards, the MA are selectively back-extracted by a hydrophilic complexing agent [15-17]. In order to reduce the secondary radioactive waste generation and ease the following conversion steps, that are some of the main issues of sulphur-containing ligands, another hydrophilic complexing agent, 2,6-bis[1-(propan-1-ol)-1,2,3-triazol-4-yl]pyridine (PTD, see fig. 1), totally constituted by merely C, H, O and N atoms, has been proposed [18]. Moreover, this ligand showed so promising extraction selectivity and efficiency for Am and Cm that it was selected as reference complexing agent for the *i*-SANEX process [19, 20]. The hydrolytic and radiolytic stability is another key requirement in view of the future implementation of a solvent extraction system in the SNF reprocessing, due to the highly radioactive environment generated by the dissolved radionuclides [21, 22]. Preliminary studies showed the outstanding stability of extraction performance of aged and irradiated PTD solutions. Furthermore, its main degradation products were hypothesised [23].

In this work, the selectivity of PTD for trivalent Am and Cm towards Eu, chosen as representative of the lanthanides, has been assessed by complexometric titration

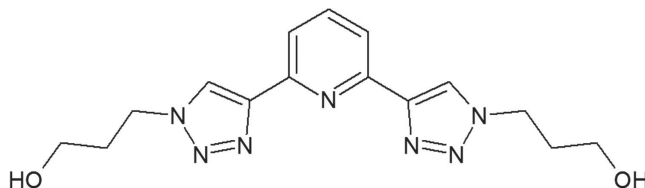


Fig. 1. – Molecular structure of PTD.

experiments in the aqueous phase followed by UV-Vis spectrophotometry and Time-Resolved Laser Fluorescence Spectroscopy (TRLFS) techniques, under relevant conditions. The complexometric titration investigates the formation of soluble coordination complexes between the cation and the ligand upon stepwise addition (*i.e.*, titration) of one of the above-mentioned reagents. The cations chemical species (*i.e.*, the metal-ligand complexes) can be studied by analysing the absorption and emission spectra acquired by UV-Vis and TRLFS techniques, respectively. The choice of the experimental conditions is of primary importance to properly mimic the radiolytic damage of molecules in the aqueous and organic phases in the reprocessing facility. The common procedure consists in irradiating synthetic samples prior to analysis. Essential parameters to be considered are the type of radiation, dose rate, and total absorbed dose, depending on SNF composition, burn-up, and cooling time after discharge. Even if the main contribution to radiolysis is due to alpha emitters, the irradiation is usually performed by gamma sources, since it is technically easier and the induced effects are conservative [22,24]. In order to possibly appreciate a radiation-induced modification, PTD solutions were γ -irradiated by 2.5 kGy/h ^{60}Co source up to 200 kGy, that is far above the absorbed dose received at process conditions. For proper comparison, some PTD solutions were aged up to 100 days before complexometric titration experiments. Metal-ligand speciation was studied in the conditions of process, that is 0.44 mol/L HNO_3 . The obtained results were compared with those already available for fresh PTD solutions, in order to evaluate the impact of radiolysis and hydrolysis on metal-ligand complex formation [18,19].

2. – Material and methods

2.1. Chemicals. – All chemicals employed in this work were acquired from Sigma Aldrich or Alfa Aesar and used without any purification. The Cm(III) stock solution has a concentration of $2.12 \cdot 10^{-5}$ mol/L in 0.017 mol/L HClO_4 (with isotopic mass distribution of 89.7% Cm-248, 9.4% Cm-246, $\leq 1\%$ Cm-243, Cm-244, Cm-245 and Cm-247). The ^{241}Am (III) stock solution has a concentration of $1.2 \cdot 10^{-2}$ mol/L in 6.1 mol/L HNO_3 . PTD was synthesized at the University of Parma according to an elsewhere described procedure [18]. The ligand stock solutions for complexometric titration experiments were prepared by dissolving weighed amounts of PTD in 0.44 mol/L HNO_3 (or in ultrapure water in the case of Eu titration) yielding a final PTD concentration of 80 mmol/L. Ultrapure water (Millipore, Billerica, USA; $18.2 \text{ M}\Omega \cdot \text{cm}$) was used for the preparation of all solutions.

2.2. Ageing and irradiation of solutions. – In this work, a ^{60}Co gamma source characterized by 2.5 kGy/h dose rate was used. The irradiations were performed at different absorbed doses up to 200 kGy. In order to standardize the irradiation protocol, all solutions were stored in glass vials closed with a plastic lid and kept in the dark at room temperature ($22^\circ\text{C} \pm 1^\circ\text{C}$). At the end of the irradiation, the solutions were stored at $4^\circ\text{C} \pm 1^\circ\text{C}$ until analysis. Unirradiated samples, with same ageing and thermal treatment of the irradiated samples, were taken as reference.

2.3. TRLFS analysis. – TRLFS analyses were performed using a Nd:YAG laser (Continuum Surelite) combined with a pumped dye laser system (NARROWscan D-R Dye laser, 10 Hz repetition rate) set at 396.6 nm excitation wavelength for Cm(III). For the spectral decomposition, a spectrograph (Shamrock 303i) with 300, 900 and 1200 lines per mm grating turret was used. The fluorescence emission spectra were detected by

an ICCD camera (iStar Gen III, ANDOR). In order to avoid Rayleigh scattering and organic ligand fluorescence, 1 μ s delay time between excitation and analysis was applied. The Cm(III) stock solution was diluted by 0.44 mol/L HNO₃ to a concentration of 10⁻⁷ mol/L. The quartz cuvette was kept at controlled temperature of 25 °C during data acquisition. Afterwards, the titration experiment was performed by subsequently adding aliquots of solutions of PTD in 0.44 mol/L HNO₃, up to a final PTD concentration of approximately 30 mmol/L. The system was allowed to reach chemical equilibrium before recording each spectrum. Spectroscopic data were analysed by Origin software (OriginLab Corporation). The spectra of solvated cation (Cm) and of CmL_{*j*} species, where L is the ligand and *j* ranges from 0 to 3, obtained in a previous work, were used to deconvolute the acquired spectra and obtain the species distribution, considering a fluorescence intensity factor correction [19]. The conditional stability constants of CmL_{*j*} (*j* = 1–3) complexes were calculated accordingly with the stepwise complexation model confirmed by the slope analysis.

2.4. UV-Vis analysis. – UV-Vis analyses were carried out at controlled temperature (25 °C) in 1 cm optical path length quartz cuvette by a CARY 5000 spectrophotometer (Agilent) linked with a glove box for the handling of radioactive samples. The speciation between Am(III) and PTD was studied following a similar approach than Cm(III), *i.e.*, by stepwise additions of known amounts of ligand to the cation solution, in 0.44 mol/L HNO₃ medium. The Am concentration was kept at 1.8 · 10⁻⁴ mol/L, while the ligand concentration was increased up to 4 · 10⁻² mol/L. Since the UV-Vis signal of europium is too weak, the Eu(III)-PTD speciation was studied by a different approach, *i.e.*, by stepwise additions of known amounts of cation to the PTD solution. The ligand concentration was kept equal to 1 · 10⁻⁴ mol/L, while the cation concentration was stepwisely increased up to 5 · 10⁻³ mol/L. These analyses were performed in water medium, since nitrates and PTD absorb at similar wavelength. Few minutes between each titration step and UV-Vis spectra acquisition proved to be sufficient for the complexation reaction to reach equilibrium. The AmL_{*j*} and EuL₁, where L is the ligand and *j* ranges from 1 to 3, conditional stability constants were computed by HypSPEC software.

3. – Results and discussion

The PTD complexation capabilities have been studied by spectroscopic investigation under relevant process conditions. Therefore, the experiments were performed on aged and irradiated solutions of PTD dissolved in 0.44 mol/L HNO₃, unless otherwise specified. Within the An family, Am(III) and Cm(III) speciation was followed by UV-Vis and TRLFS techniques, respectively. Among the Ln series, Eu(III) was studied by UV-Vis spectrophotometry.

3.1. PTD-Cm complexation by TRLFS. – The speciation of Cm(III) with PTD was studied by complexometric titrations followed by TRLFS measurements. Aged and irradiated 0.08 mol/L PTD in 0.44 mol/L HNO₃ solutions were analysed in order to obtain the stoichiometry and the conditional stability constants of CmL_{*j*} complexes under hydrolysis and radiolysis conditions. In order to simulate the hydrolytic degradation, the PTD solution was aged for 29 days. The combination of hydrolytic and radiolytic effects was studied with the ligand solution irradiated at 100 kGy with 2.5 kGy/h dose rate ⁶⁰Co source. For proper comparison, the same 29 days ageing period was applied to the irradiated sample. The titration experiment was performed by stepwise

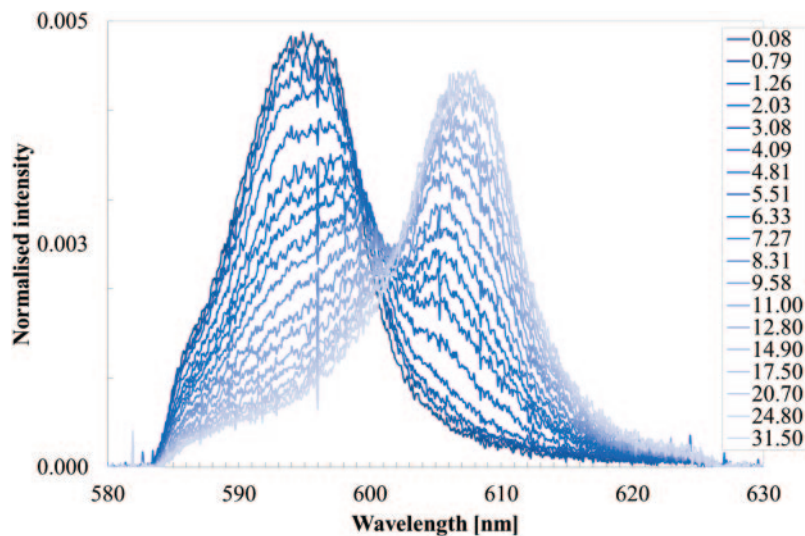


Fig. 2. – Normalised Cm(III) fluorescence spectra with increasing concentration of PTD in 0.44 mol/L HNO₃, [Cm(III)]_{in} = 10⁻⁸ mol/L. The ligand concentration ranges from 0 to 31.5 mmol/L, as reported in the graph. The ligand solution was aged for 29 days.

additions of ligand to the Cm(III) containing solution, being it at the initial concentration of about 10⁻⁷ mol/L. The final medium was 0.44 mol/L HNO₃. The normalised TRLFS spectra obtained with the aged and irradiated PTD solutions are reported in fig. 2 and fig. 3.

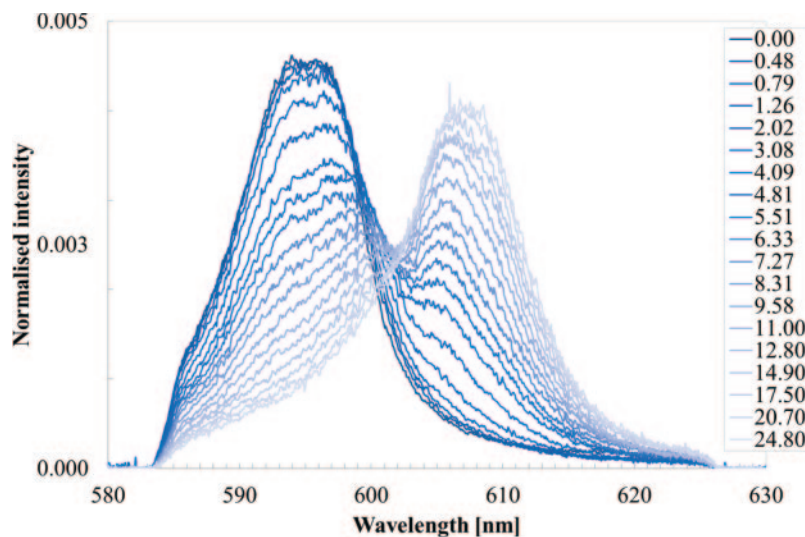


Fig. 3. – Normalised Cm(III) fluorescence spectra with increasing concentration of PTD in 0.44 mol/L HNO₃, [Cm(III)]_{in} = 10⁻⁸ mol/L. The ligand concentration ranges from 0 to 24.8 mmol/L, as reported in the graph. The irradiation was performed at 100 kGy absorbed dose with the 2.5 kGy/h dose rate ⁶⁰Co source.

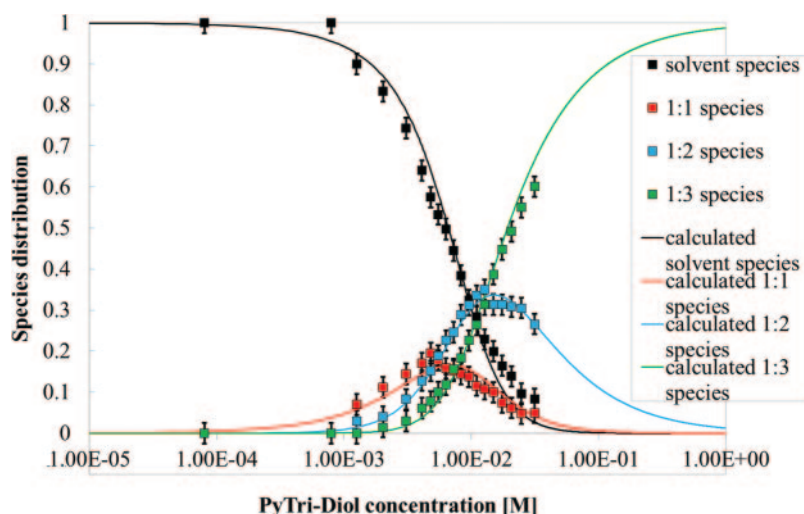


Fig. 4. – Cm(III) species distribution as a function of PTD concentration in 0.44 mol/L HNO_3 . The experiment was performed with PTD solution aged for 29 days.

The solvated Cm(III) species shows an emission band centred at 593.8 nm, belonging to the ${}^6\text{D}'_{7/2} \rightarrow {}^8\text{S}'_{7/2}$ transition. As shown in a previous work, a bathochromic shift is observed at increasing ligand concentration, due to three further emission bands centred at 600.1 nm, 605.5 nm and 608.6 nm [19]. This is coherent with the stepwise formation of CmL_j complex species, where j ranges from 1 to 3. The normalized spectra were deconvoluted using the pure components spectra calculated in the previous work, thus obtaining the species distribution. In order to compensate for the increasing fluorescence caused by complexation, the fluorescence intensity factors estimated in the previous work have been employed [19]. The fractions of the formed species are reported in fig. 4 and in fig. 5

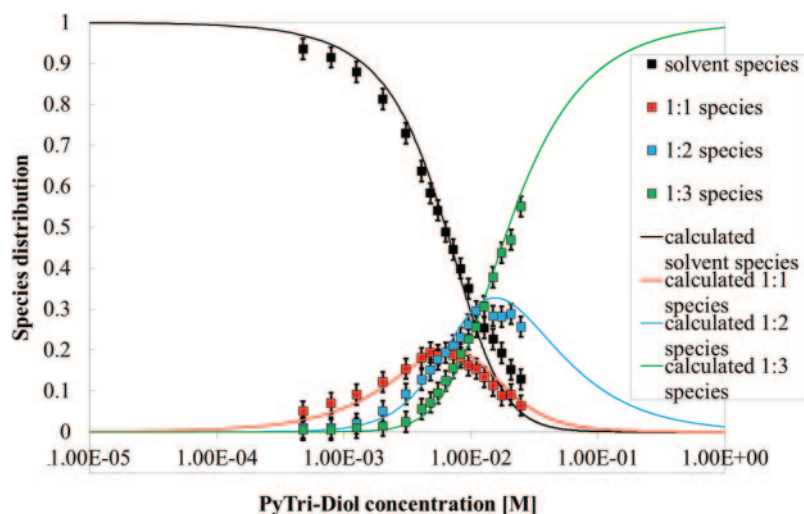


Fig. 5. – Cm(III) species distribution as a function of PTD concentration in 0.44 mol/L HNO_3 . The experiment was performed with PTD solution irradiated at 100 kGy.

TABLE I. – Conditional stability constants ($\log\beta_{1:j}$) of the CmL_j complexes obtained by TRLFS technique with fresh, aged and irradiated PTD in 0.44 mol/L HNO_3 , where j denotes the number of ligands in the complex and it ranges from 1 to 3. The aged solution was stored in the dark for 29 days before analysis. The irradiation was performed at 100 kGy by the 2.5 kGy/h dose rate ^{60}Co source.

	Fresh ¹⁴	Aging (29 days)	Radiolysis (100 kGy)
$\log\beta_{1:1}$	1.7 ± 0.2	1.7 ± 0.2	1.8 ± 0.2
$\log\beta_{1:2}$	4.0 ± 0.2	4.0 ± 0.2	4.0 ± 0.2
$\log\beta_{1:3}$	5.7 ± 0.2	5.9 ± 0.3	5.9 ± 0.3

as a function of the ligand concentration. Following the same computational approach described in the previous work, slope analyses have been performed to demonstrate the stepwise complexation model and the conditional stability constants have been calculated [19]. The conditional stability constants of CmL_j complexes under hydrolysis and radiolysis conditions are reported in table I and compared with the data obtained with fresh PTD solutions [19]. As evidenced, the results obtained with irradiated PTD are unaltered with respect to both aged and fresh PTD, within the uncertainty. These outcomes are further confirmation of the unprecedented ligand stability towards hydrolytic and radiolytic degradation [18, 23].

3.2. PTD-Am complexation by UV-Vis. – The speciation between Am(III) and PTD was studied by complexometric titrations followed by UV-Vis spectrophotometry. Fresh

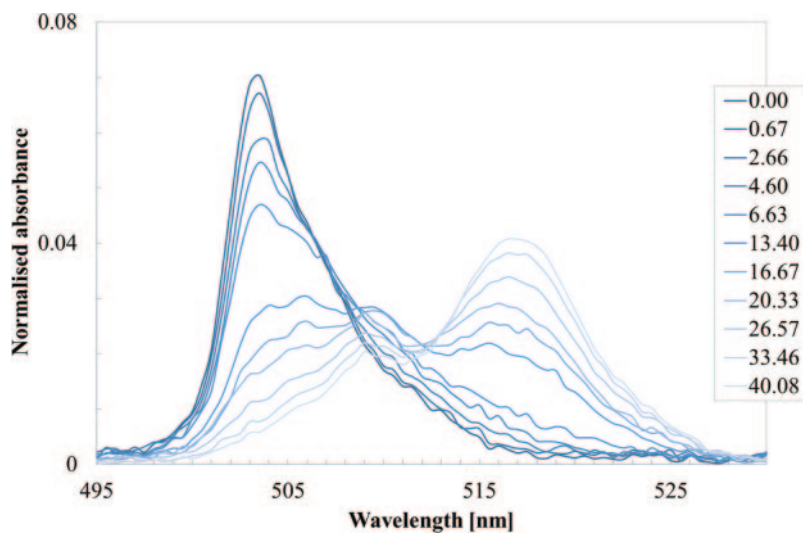


Fig. 6. – Normalised Am(III) absorbance spectra with increasing concentration of fresh PTD in 0.44 mol/L HNO_3 , $[\text{Am(III)}] = 1.8 \cdot 10^{-4}$ mol/L. The ligand concentration ranges from 0 to 40.08 mmol/L, as reported in the graph.

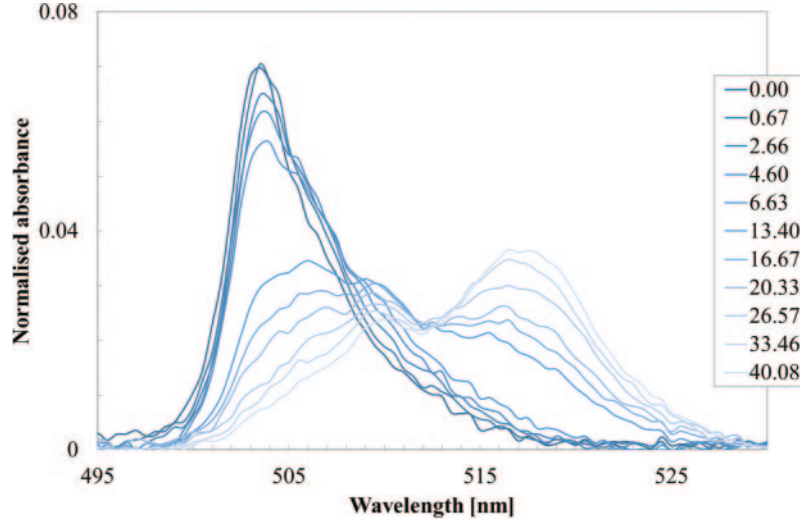


Fig. 7. – Normalised Am(III) absorbance spectra with increasing concentration of irradiated PTD in 0.44 mol/L HNO₃, [Am(III)] = $1.8 \cdot 10^{-4}$ mol/L. The ligand concentration ranges from 0 to 40.08 mmol/L, as reported in the graph. The irradiation was performed at 200 kGy absorbed dose with the 2.5 kGy/h dose rate ⁶⁰Co source.

and irradiated 0.08 mol/L PTD in 0.44 mol/L HNO₃ solutions were analysed in order to obtain the stoichiometry and the conditional stability constants of AmL_j complexes under hydrolysis and radiolysis conditions. The irradiation was performed with the 2.5 kGy/h dose rate ⁶⁰Co source at the absorbed dose of 200 kGy. The titration experiment was performed by stepwise addition of ligand to the cation solution, at fixed ²⁴¹AmCl₃ concentration of $1.8 \cdot 10^{-4}$ mol/L. The final medium was 0.44 mol/L HNO₃. The normalised absorption spectra obtained with the fresh and irradiated PTD solutions are reported in fig. 6 and fig. 7.

A sensitive absorption band corresponding to the electronic transition from ⁷F₀ ground state to ⁵L₆ excited state was found at 503.5 nm, in agreement with the literature. Since first coordination sphere modifications strongly alter this band resulting in a bathochromic shift, several speciation information can be retrieved from it [25]. At increasing PTD concentration, three further absorption bands centred at $\lambda = 506$ nm,

TABLE II. – Conditional stability constants ($\log \beta_{1;j}$) of the AmL_j complexes obtained by UV-Vis technique with fresh and irradiated PTD in 0.44 mol/L HNO₃, where *j* denotes the number of ligands in the complex and it ranges from 1 to 3. The irradiation was performed at 200 kGy by the 2.5 kGy/h dose rate ⁶⁰Co source.

	Fresh	Irradiated (200 kGy)
$\log \beta_{1;1}$	1.85 ± 0.04	1.96 ± 0.12
$\log \beta_{1;2}$	3.69 ± 0.07	3.61 ± 0.13
$\log \beta_{1;3}$	5.59 ± 0.07	5.34 ± 0.13

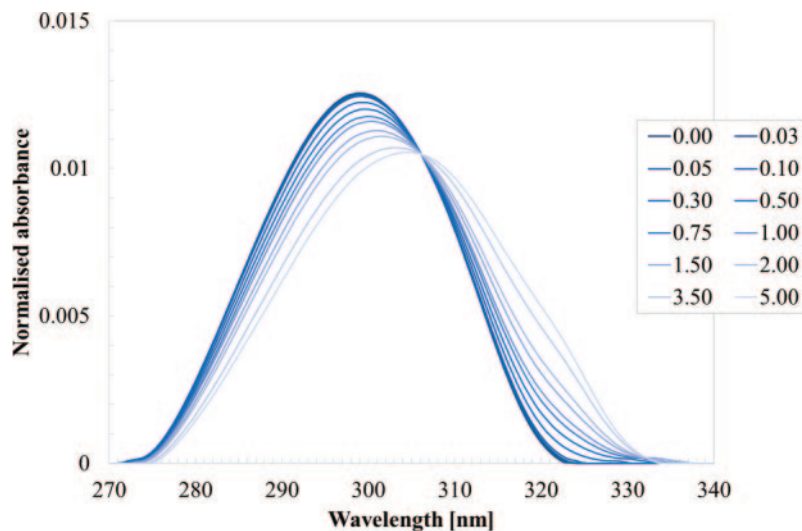


Fig. 8. – Normalised Eu(III) absorbance spectra with increasing concentration of Eu(III) in water, $[PTD] = 1 \cdot 10^{-4}$ mol/L. The cation concentration ranges from 0 to 5 mmol/L, as reported in the graph.

510 and 518 nm appear, consistently with the stepwise complexation between Am(III) and PTD. This bathochromic shift is similarly visible in the titrations of both fresh and irradiated PTD solutions. It results in the formation of AmL_j complex species, where j ranges from 1 to 3. The spectral data were analysed by the HypSPEC software and the computed conditional stability constants of AmL_j complexes are reported in table II. It

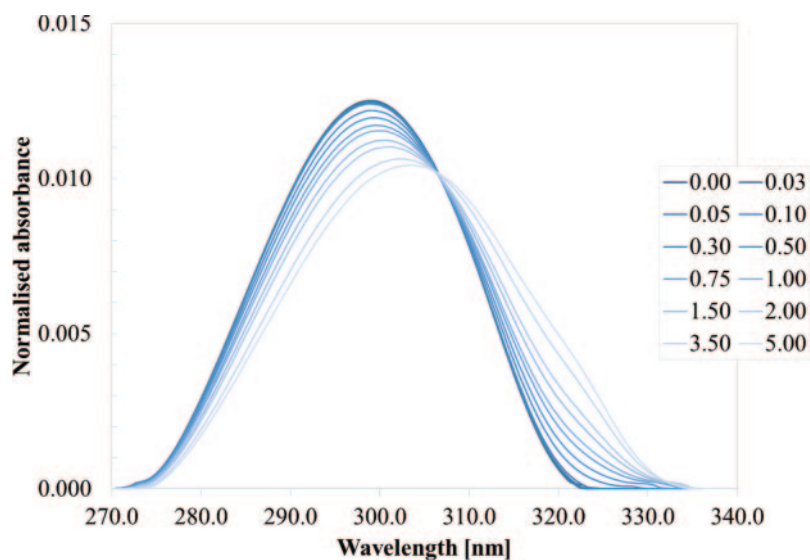


Fig. 9. – Normalised Eu(III) absorbance spectra with increasing concentration of Eu(III) in water, $[PTD] = 1 \cdot 10^{-4}$ mol/L. The cation concentration ranges from 0 to 5 mmol/L, as reported in the graph. The irradiation was performed at 200 kGy absorbed dose with the 2.5 kGy/h dose rate ^{60}Co source.

TABLE III. – *Conditional stability constants ($\log\beta_{1:j}$) of the EuL_1 complexes with fresh, aged and irradiated PTD. The titrations were carried out in water by UV-Vis spectrophotometry. The aged solution was stored in the dark for 100 days before analysis. The irradiation was performed at 200 kGy by the 2.5 kGy/h dose rate ^{60}Co source.*

	Fresh [18]	Ageing (100 days)	Radiolysis (200 kGy)
$\log\beta_{1:1}$	2.4 ± 0.1	2.4 ± 0.1	2.3 ± 0.1

could be observed that the results obtained with irradiated PTD are just slightly altered with respect to fresh PTD, if the uncertainty is considered. This result further confirms that hydrolytic and radiolytic induced modifications have an almost negligible impact on complexation, in agreement with the performance outlined in previous batch solvent extraction experiments, where fresh, aged and irradiated PTD solutions were tested to back-extract MA from the loaded organic phase containing both MA and Ln [18].

3.3. PTD-Eu complexation by UV-Vis. – The speciation between Eu(III) and PTD was studied by complexometric titrations followed by UV-Vis spectrophotometry. Since the Eu absorption band is too weak, the titration experiment was performed at fixed PTD concentration ($1 \cdot 10^{-4}$ mol/L) by stepwisely adding EuCl_3 up to 5 mmol/L. In order to discriminate the interfering absorption band of nitrate from PTD signal at around 300 nm, the titration was performed in water and not in 0.44 mol/L nitric acid. Consequently, the Eu(III) complexation results cannot be directly compared with those of Am(III) and Cm(III), since the latter are affected by ligand protonation as they were determined in acidic media [18]. Aged and irradiated solutions of 0.08 mol/L PTD in water were analysed in order to obtain the conditional stability constants of EuL_j complexes under hydrolysis and radiolysis conditions. In order to estimate both hydrolytic and radiolytic effects, a PTD solution was aged for 100 days and irradiated at 200 kGy with 2.5 kGy/h dose rate ^{60}Co source. For proper comparison, another solution was just aged for the same ageing period of 100 days.

The normalised absorption spectra of the titration experiment with aged and irradiated PTD solutions are reported in fig. 8 and fig. 9, respectively. Just a slight evolution is observable with increasing Eu(III) concentration, proving the cation complexation by PTD. The spectral data analysis was performed by the HypSPEC software. In contrast to the Am(III) and Cm(III) results, only the EuL_1 complex was identified. The corresponding conditional stability constants obtained with aged and irradiated PTD solutions are reported in table III and compared with the data obtained with fresh PTD solutions [11]. As evidenced, the results obtained with aged and irradiated PTD are unaltered with respect to both aged and fresh PTD, within the uncertainty. These results are in agreements with the unprecedented ligand stability towards both hydrolytic and radiolytic degradation [18, 23].

4. – Conclusions

In this work, the complexation of Am(III), Cm(III) and Eu(III) with PTD has been studied in aqueous solutions by UV-Vis and TRLFS experiments under experimental conditions relevant to an industrial application, *i.e.*, in 0.44 mol/L HNO_3 , after ageing

and gamma irradiation up to 200 kGy. This study has allowed to shed light on the effects of hydrolysis and radiolysis on PTD complexation capabilities. Beforehand, to the best of the author's knowledge, no analogous speciation research was performed on aged and irradiated solutions. The formed complexes M^{3+} -PTD have been identified and characterized. The conditional stability constants of Am(III), Cm(III) and Eu(III) have been determined with aged and irradiated PTD solutions and compared with those obtained with fresh PTD solutions. The absence of evolution of speciation and conditional stability constants, within the uncertainty, is in complete agreement with the results of previous solvent extraction experiments, further confirming the extraordinary stability of PTD-based stripping solvent.

Further efforts are underway to fully understand the impact of hydrolysis and radiolysis on solvent extraction performance and PTD degradation mechanism. To date, the high selectivity and stability exhibited by PTD look promising for its forthcoming application in the *i*-SANEX process for the An(III)/Ln(III) separation.

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