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Influence of the purity and irradiation stability of Cyanex 301 on the separation of trivalent actinides from lanthanides by solvent extraction

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The solvent extraction of trivalent actinides and lanthanides from aqueous 1M $NaNO_3$ by the sulfur containing acidic extractant Cyanex 301 was studied. This commercial extractant shows very interesting features for An(III)/Ln(III) separations depending on the purification of its main constituent bis(2,2,4-trimethylpentyl)dithiophosphinic acid. Efficient separations were observed with nonpurified Cyanex 301 for high Ln(III) concentrations, but not for low Ln(III) concentrations. Successful purification of Cyanex 301 was performed by precipitation of the ammonium dithiophosphinate. Very high An(III)/Ln(III) separation factors (higher than 10^3) were obtained for micro or macro concentrations of Ln(III) at pH 3–4.

Introduction

The selective extraction of transplutonides(III) from lanthanides(III) is still one of the most difficult separation procedures in chemistry. There are few differences in the chemical and physical properties of these elements, which makes it difficult to develop efficient processes for separating these metal ions. It is well known that extractants containing so-called soft donor atoms (N or S) have a certain selectivity for transplutonides(III). It is assumed that this effect can be explained by a greater complex stability (metal-ligand covalent bonding or steric effects) between the transplutonides and the soft donor ligands.[4]

Recently ZHU et al.[1] reported at the Global 95 conference in 1995 that they had obtained very high separation factors (>5000) between Am(III) and Eu(III) after purifying Cyanex 301, whose main constituent is bis(2,2,4-trimethylpentyl)-dithiophosphinic acid. A separation with unpurified Cyanex 301 only took place after prior saponification at pH values >5. In contrast, the monosubstituted dithiophosphinic acid (Cyanex 302) only showed minor selectivity. JARVINEN et al.[2] examined the synergistic effect of TBP for three different dithiophosphinic acids on the separation of trivalent actinides from lanthanides. Very high separation factors of 1000 were also achieved between Am(III) and Eu(III) in a single extraction step combining, e.g., dicyclohexyldithiophosphinic acid with TBP. Surprisingly, no separation was achieved with purified Cyanex 301 without TBP addition.

These contradictory results induced us to carry out comparable studies. The purpose of this work has been to investigate, in particular, the extraction behavior of unpurified and purified Cyanex 301. Furthermore, we also examined the radiation stability of purified Cyanex 301.

Experimental

Reagents

The extractants Cyanex 272, 302 and 301 were kindly provided by Cytec Ind., Canada, and used as received. Cyanex 301 was additionally purified using two different methods. n-Dodecane (from Merck) as well as all the other chemicals were of analytical grade. The radiotracers delivered by Blaseg Isotopendienst, ^{241}Am (3.7 MBq in 1 ml 1M HCl) and $^{152-154}$Eu (3.7 MBq in 5 ml 0.5M HCl) were diluted to 100 ml with dist. H_2O. A 1:1 mixture of both solutions (pH 2) was used as stock solution.

Procedure

In a glass vial with teflon stopper 2 ml aqueous phase and 2 ml organic phase were equilibrated after tracering with the aid of a MiniShaker, type IKA Vibro-Fix, for 10 minutes at 21 °C. The tracer concentrations of $^{152-154}$Eu and ^{241}Am were approximately 10^{-7} mol/l. Preliminary experiments showed that the extraction equilibrium was already attained within 5 minutes. After centrifuging, an aliquot of 1 ml was sampled from both phases for analysis. In order to avoid cross contamination in the aqueous phase, the organic phase was completely removed before.

The γ-active nuclides were analysed with the aid of a germanium detector (Ortec). The lines at energies of 59 and 122 keV were used for evaluating the ^{241}Am and $^{152-154}$Eu, respectively. Care was taken during measurements to ensure that the pulse rates were generally at least 10 000 in order to maintain the statistical error of individual measurements in the order of 1%. The measuring period varied between 5 minutes

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and 20 hours. The distribution ratios D_{Am} and D_{Eu} are calculated from the quotient of radioactivity per ml in the organic and aqueous phases. Distribution ratios between 0.01 and 1000 exhibit a maximum error of 5%. The error may be up to 20% for smaller and larger values.

The distribution ratios of the inactive lanthanides La, Ce, Pr, Nd, Sm, Eu, Gd and Yb were determined by ICP-MS (inductively coupled plasma mass spectrometry). The aqueous phase was directly measured after adequate dilution. In order to determine the element concentration in the organic phase, stripping with 1M HNO_3 was performed before. The radioactivity of ^{244}Cm was determined by liquid scintillation counting (LSC).

The pH of the aqueous phase was adjusted by adding NaOH and HNO_3 and measured after extraction with the aid of a combined Ag/AgCl micro glass electrode (Metrohm) in combination with a digital pH meter, type 691 (Metrohm). The pH meter was previously calibrated using commercial pH 3.00 and 5.00 buffer solutions.

^{31}P-NMR spectra were recorded on a Bruker AMX 400 WB spectrometer. Chemical shifts were measured in δ scale with respect to an internal reference of H_3PO_4.

Purification of Cyanex 301

Purification over zinc dithiophosphinate: The purification procedure is described in detail by Jarvinen et al.[2] We also obtained transparent crystals after two weeks of crystallization, which were filtrated and dried in air for 2–3 hours. The yield was very low (<5%). The purity was higher than 99.5% according to ^{31}P-NMR.

Purification over NH_4 dithiophosphinate:[3] 52 g of Cyanex 301 (0.13 mol) was dissolved in 200 ml benzene and heated to 70 °C while being stirred. After adding 14 g of ammonium carbonate the solution was left to react at this temperature for another hour. The solution was filtrated and kept overnight in a refrigerator (10 °C). The crystallized NH_4 salt was filtrated and recrystallized two times from benzene. A white crystalline product (38.5 g) was obtained after drying, which was gathered with 300 ml petroleum benzine (40–60 °C) and shaken twice with 300 ml 4M HCl. The organic phase was then separated, washed twice with dist. H_2O and finally dried with 20 g Na_2SO_4. Solvent was then removed at 30 °C and 40 mbar using a rotary evaporator. The initially viscous pale yellowish product crystallized after the last solvent traces had been removed at 12 mbar. The purity was higher than 99.5% according to ^{31}P-NMR. We were able to also produce the NH_4 salt by introducing gaseous NH_3 into a benzolic solution of Cyanex 301. After a NH_3 surplus had been introduced at 10 °C, the salt precipitated after several hours in the cold. Processing was then carried out as described above.

Radiation stability

Gamma irradiation took place in the cooling pond of the DIDO research reactor at the Research Centre Jülich. The γ-dose rate was 4200 Gy/h. For the ^{31}P-NMR investigations approx. 100 mg of solid purified Cyanex 301 was irradiated with a total dose of $4\cdot10^3$, $1\cdot10^5$, $7\cdot10^5$ and $1\cdot10^6$ Gy. For the extraction studies 10 ml of 0.5M Cyanex 301 in n-dodecane was irradiated with a total dose of $1\cdot10^4$, $1\cdot10^5$ and $7\cdot10^5$ Gy.

Results and discussion

Extraction studies with unpurified Cyanex 272, 302 and 301

The results of the extraction of tracer amounts of Am(III) and Eu(III) from 1M $NaNO_3$ with 12.5 wt% Cyanex 272, 302 and 301 in n-dodecane are shown in Fig. 1. With the extractants Cyanex 301 and 302 Eu(III) is slightly better extracted than Am(III) in the pH range between 0.5 and 3. The Am(III)/Eu(III) separation factors are in the range of 0.5. Cyanex 272 shows a pronounced selectivity for Eu(III) with Am(III)/Eu(III) separation factors of 0.2 at pH 1–3.

The extraction behavior of Cyanex 301 in the presence of macro amounts of Eu will be examined in the following. The Eu concentrations in the aqueous solutions varied between 0.0001 and 0.3M Eu(III). For comparability of the measurement results shown in Table 1, the nitrate content of the aqueous solution was set to 1M by the addition of $NaNO_3$.

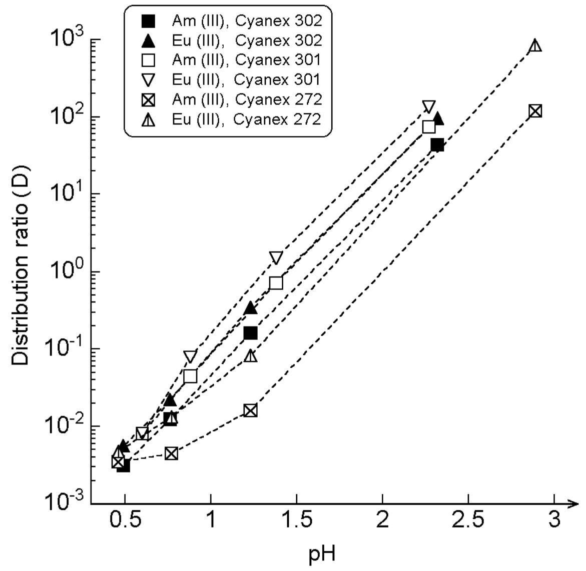

Fig. 1. Extraction of Am(III) and Eu(III) by Cyanex 272, 302 and 301. The organic solution contained 12.5 wt% extractant in n-dodecane. The initial aqueous phase was a solution of <10^{-7} M Am(III) and Eu(III) in 1M $NaNO_3$

Table 1. Extraction of Am(III) and Eu(III) by 12.5 wt% Cyanex 301 in n-dodecane. Effect of the Eu(III) concentration. The initial aqueous phase was a solution of $<10^{-7}$ M ^{241}Am and ^{152}Eu + variable $Eu(NO_3)_3$, 1M NO_3^-

[Eu], mol/l	pH	$D_{Am(III)}$	$D_{Eu(III)}$	$\alpha = D_{Am(III)}/D_{Eu(III)}$
$<10^{-7}$	0.60	0.0081	0.0081	1
	0.88	0.0443	0.0784	0.56
	1.38	0.719	1.499	0.48
	2.27	73.767	133.73	0.55
0.0001	1.59	1.368	2.866	0.48
	2.41	60.37	75.61	0.80
	2.74	97.4	88.99	1.09
	4.79	>1000	547.96	–
0.001	1.56	0.343	0.663	0.53
	2.26	12.18	6.33	1.92
	2.62	31.11	14.42	2.15
	4.95	>1000	414.67	–
0.01	2.09	0.0664	0.538	0.12
	2.39	0.136	0.632	0.22
	3.57	18.64	1.115	16.71
	4.26	772.35	3.476	222.13
0.1	2.07	0.0114	0.046	0.25
	2.36	0.0274	0.0534	0.51
	3.11	0.912	0.0699	13.04
	3.77	39.83	0.103	383.28
0.3	1.85	0.00388	0.0183	0.21
	2.22	0.0194	0.019	1.0
	3.56	16.34	0.0358	456.1
	4.37	160.41	0.070	2280.4

As can be seen from Table 1 and Fig. 2, a Eu concentration of 0.01M is already sufficient for Cyanex 301 to display a pronounced selectivity for Am(III) above pH 3. At higher Eu concentrations, a separation already takes place at lower pH values. As can be seen from Fig. 2, the Am(III)/Eu(III) separation factors significantly increase with rising pH values and reach values >1000 at pH>4. No significant separation was achieved with Cyanex 302, the monothio-phosphinic acid, even at Eu concentrations of 0.1M.

The results described in this chapter clearly show that the purity level of the extractants used has a great influence on the extraction behavior of tracer amounts of Am and Eu. This influence seems to be less pronounced for the extractants Cyanex 302 and 272 than for Cyanex 301, the dithiophosphinic acid. It can be seen from the results that the impurities contained in Cyanex 301 are complexed by macro amounts of Eu, especially at high pH values. It was thus demonstrated that commercial Cyanex 301 only has a high selectivity for Am(III) at high Ln concentrations.

A similar behavior has already been observed by MUSIKAS.[4] In his studies with di(2-ethylhexyl)dithiophosphoric acid (HDEHDTP) as the extractant, an Am/Eu separation was also only achieved with macro amounts of Eu (0.05M) and with 0.25M TBP. MUSIKAS assumed that the di(2-ethylhexyl)thiophosphoric acid impurity formed by oxidation extracts Am and Eu better than HDEHDTP, but cannot separate them, and that a separation was only effected by complete complexation of the monothio acid with Eu(III).

Our ^{31}P-NMR investigations show that Cyanex 301 consists of 78.7% $R_2PS(SH)$, 8.8% R_3PS, 9.9% unknown compounds and 2.6% $R_2PS(OH)$ (R=2,4,4-trimethylpenthyl). The results thus show that the monothio derivative (Cyanex 302) is also contained as a byproduct in commercial Cyanex 301 and clearly influences the extraction of Am and Eu.

Extraction studies with purified Cyanex 301

In contrast to its viscous base product smelling obnoxiously of SO_2, H_2S, Cyanex 301 purified over NH_4 salt is solid and almost odourless at room temperature. A 0.5 molar solution was prepared in n-dodecane for the extraction experiments. The aqueous phase was a 1 molar $NaNO_3$ solution, as before. Initial extraction experiments yielded a very high separation factor of 5032 at a pH 3.3, confirming the results of ZHU et al.[1] The D_{Am} and D_{Eu} distribution ratios were 11.2 and 0.0022, respectively. After three days, extraction experiments were carried out with the same base solution at different pH values. It was not possible to reproduce the high separation factor achieved with a freshly prepared solution. After two weeks, the extraction results shown in Fig. 3 were obtained. It can be seen that the distribution ratios of europium, D_{Eu}, had increased, while D_{Am} remained almost constant, so that smaller separation factors were obtained.

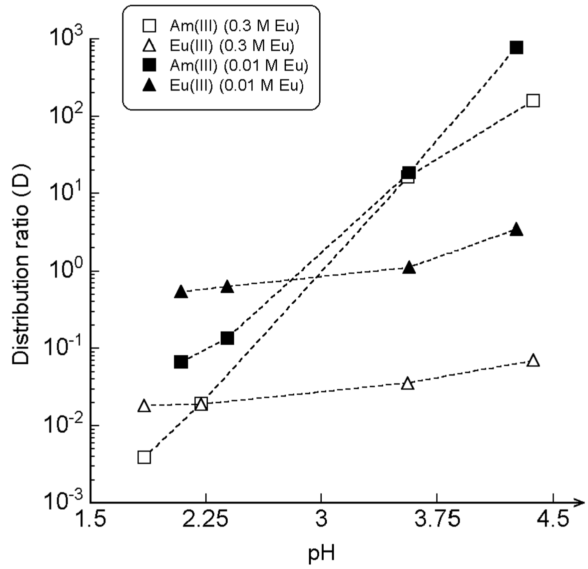

Fig. 2. Extraction of Am(III) and Eu(III) by 12.5 wt% Cyanex 301 in n-dodecane. Effect of the Eu(III) concentration. The initial aqueous phase was a solution of $<10^{-7}$ M ^{241}Am and ^{152}Eu + variable $Eu(NO_3)_3$, total 1M NO_3^-

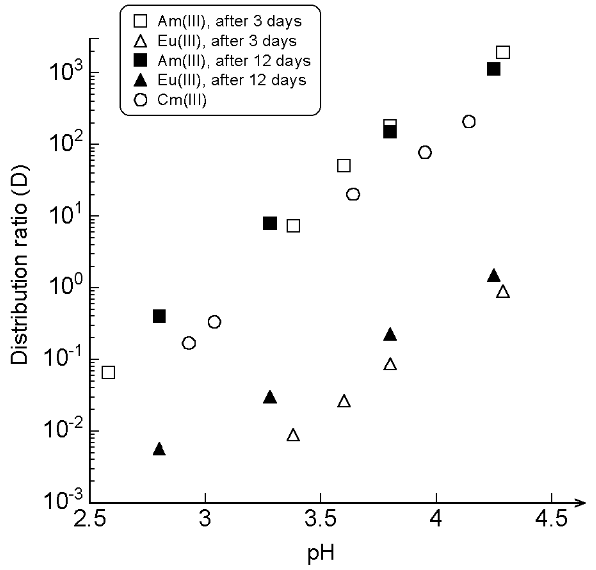

Fig. 3. Extraction of Am(III), Cm(III) and Eu(III) by purified 0.5M Cyanex 301 in n-dodecane. Effect of the ageing. The initial aqueous phase was a solution of $<10^{-7}$ M ^{241}Am, ^{244}Cm and ^{152}Eu and 1M $NaNO_3$

Obviously, the purified Cyanex 301 slowly decomposed in both the dissolved and solid state. The impurity formed by decomposition extracts the Eu better than the dithiophosphinic acid. It must be borne in mind, however, that the effects observed can only be caused by minimal impurities with the amounts of ^{241}Am and ^{152}Eu tracers being $<10^{-7}$ mol/l. The distribution ratios of trace amounts of Cm(III) were determined separately and also plotted in Fig. 3. The distributon ratios are similar to the values of Am(III).

In the following, the influence of the europium concentration on Am/Eu separation with purified Cyanex 301 will be examined. The aqueous solutions contained 0.0001–0.1 mol/l inactive $Eu(NO_3)_3$. The total nitrate content was again set to 1 mol/l with $NaNO_3$. The results are shown in Table 2. An extraction experiment with tracer amounts of Eu is shown for comparison (first line). It can be seen that the Eu content does not exert a great influence on the distribution ratios of Am. In the case of Eu concentrations $>10^{-5}$ mol/l, D_{Eu} drops clearly below 10^{-2} and the separation factors, D_{Am}/D_{Eu}, assume values greater than 1000. This is indicative of the fact that even minimal impurities contained in the purified Cyanex 301 are complexed by Eu. The same series of tests was repeated with six-month old extraction agent. The separation factors achieved with tracer amounts (cf. first line in Table 2) dropped from 326 to 29. The results obtained for higher Eu concentrations are again comparable.

Table 2. Extraction of Am(III) and Eu(III) by purified 0.5M Cyanex 301 in n-dodecane. Effect of the Eu(III) concentration. The initial aqueous phase was a solution of $<10^{-7}$ M ^{241}Am and ^{152}Eu + variable $Eu(NO_3)_3$, 1M NO_3^-, pH 3.3

[Eu], mol/l	$D_{Am(III)}$		$D_{Eu(III)}$		$\alpha=D_{Am(III)}/D_{Eu(III)}$	
	a)	b)	a)	b)	a)	b)
$<10^{-7}$	8.570	8.063	0.0262	0.300	326.2	28.9
10^{-4}	8.402	9.095	0.00805	0.0116	1043.7	785.6
10^{-3}	8.017	7.949	0.00259	0.00292	3092.6	2722.3
10^{-2}	6.067	5.389	0.00231	0.00223	2626.4	2414.5
10^{-1}	5.210	4.985	0.00220	0.00175	2064.6	2846.0

a) A few days after purification.
b) Six months after purification.

According to the method of JARVINEN et al.^{2} Cyanex 301 was purified by precipitating the Zn-dithiophosphinate and subsequent acidification with HCl. As also described by the authors, the dithiophosphinic acid only crystallized after a few days. The product was then filtered and dried for 2 to 3 hours in air. The residual mother liquor also solidified after a few days. In the extraction experiments, whose results are summarized in Table 3, no separation between Am(III) and Eu(III) was achieved even after repeated purification attempts. A comparison of the Am and Eu distribution ratios with those of unpurified Cyanex 301 (see Fig.1) shows that they are lower by about two orders of magnitude after purification. The results shown in the bottom line of Table 3 were achieved with the crystallized mother liquor as the extractant. These results are again comparable with those of unpurified Cyanex 301.

JARVINEN et al. did not obtain any separation under similar conditions (extractant dissolved in toluene, 0.1M $NaNO_3$ instead of 1M). In the presence of 0.3 to 0.5M TBP, on the other hand, the Am(III)/Eu(III) separation factors increased up to a maximum of 50. He interpreted his results that separation was only achieved by the synergistic effect of TBP.

The investigations described in this section were carried out prior to the experiments with Cyanex 301 purified according to the Chinese method and can now be explained as follows with the results obtained there. Similarly to JARVINEN et al., we obtained a pure product (>99.5%) after purification. However, the product purified according to the Zn method still contained minimal impurities (<0.5%), which especially extracted traces of Eu so that no separations were achieved. JARVINEN et al. and also we had first interpreted the results incorrectly. However, if the Cyanex 301 is purified according to the NH_4 method, these impurities (especially the monothiophosphinic acid) are separated and high Am(III)/Eu(III) separation rates are obtained.

Table 3. Extraction of Am(III) and Eu(III) by purified 0.5M Cyanex 301 in n-dodecane. The initial aqueous phase was a solution of $<10^{-7}$ M ^{241}Am and ^{152}Eu and 1M $NaNO_3$. Purification according to Reference 2

No.	pH	$D_{Am(III)}$	$D_{Eu(III)}$	$\alpha = D_{Am(III)}/D_{Eu(III)}$
1	2.83	1.29	1.12	1.15
2	2.60	1.82	1.76	1.03
3	2.86	1.78	1.22	1.45
3*	2.15	135.46	159.40	0.85

* Crystallized mother liquor.

Effect of lanthanides on Am(III)/Ln(III) separation

The extraction of trace amounts of ^{241}Am(III) and ^{152}Eu(III) in the presence of other lanthanides showed no influence on their extraction behavior. The aqueous solution was a lanthanide mixture containing 0.001 mol/l of La, Ce, Pr, Nd, Sm, Eu, Gd, and Yb each. The concentrations of lanthanides were chosen according to their concentrations in the spent fuel. The results are shown in Fig. 4. It can be seen that the Am(III)/Eu(III) separation factors increase significantly with rising pH. The measured values clearly exceed 1000 above pH 3.3 and reach an incredibly high value of 9000 at pH 4.2.

Inactive experiments (in absence of ^{241}Am and ^{152}Eu) were carried out to determine also the distribution ratios of the individual lanthanides, which are also summarized in Fig. 4. It can be seen that the distribution ratios are very low even at high pH values. With the exception of Ce(III) and Pr(III), the distribution ratios are clearly below 10^{-1} in the pH range examined. The distribution ratios of the lanthanides decrease in the order Ce > Pr > Nd > La > Sm > Gd > Eu >Yb.

Stability of purified Cyanex 301

It is known that sulphur-containing organophosphoric compounds tend to oxidation and decomposition, especially in the presence of acids and strong oxidants. In comparison to the dialkyldithiophosphoric acids, which have a weak hydrolytic and thermal stability, the dialkyldithiophosphinic acids are characterized by higher stability due to the lack of the ether bridge. Sole et al.[6] studied the long-term stability of unpurified Cyanex 301 in the presence of sulphuric acid and nitric acid. Gradual decomposition was only observed at high nitric acid concentrations (>2M).

During separation of the trivalent actinides from the An(III)/Ln(III) fraction, the extractant is exposed to a high radiation dose which can be up to 10^6 Gy. As a consequence of the radiolytic decomposition of the extractant, undesirable effects such as a decrease in selectivity and poorer phase separation occur due to the formation of decomposition products.

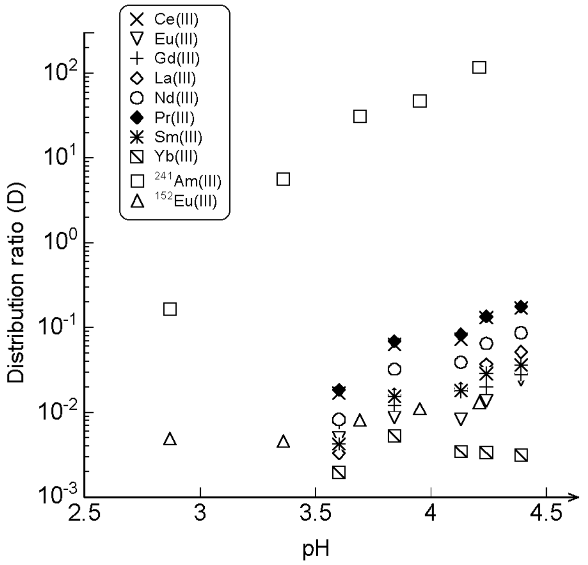

Fig. 4. Extraction of trace amounts of Am(III), Eu(III) and lanthanides (III) by purified 0.5M Cyanex 301 in n-dodecane. The initial aqueous phase was a solution of $<10^{-7}$ M ^{241}Am, ^{152}Eu and 0.001M La, Ce, Pr, Nd, Sm, Eu, Gd, and 1M NO_3^-

The extent of radiolytic degradation of the pure Cyanex 301 was first determined by estimating the residual Cyanex 301 content using quantitative ^{31}P-NMR. Table 4 shows the chemical shifts of the products formed and their amount in % as a function of the adsorbed dose. Apart from the starting compound with a chemical shift of $\delta = 68.4$ ppm, the Cyanex 302 (monothiophosphinic acid, R_2PSOH, $\delta = 55.2$ ppm) and the sulphur-free compound Cyanex 272 (R_2POOH, $\delta = 92–94$ ppm) were clearly identifiable. It can be seen from Table 4 that a decomposition of the Cyanex 301 is observed with increasing gamma dose. At a total dose of $1 \cdot 10^6$ Gy more than 80% of the Cyanex 301 has decomposed, forming approx. 6.3% Cyanex 302, 5% Cyanex 272 and a balance of undefinable compounds. In particular, the formation of these acid compounds, which are a much stronger extractant for An(III) and Ln(III) with lower selectivity, can lead to extraction problems.

For this purpose, the extraction behaviour of Am(III) and Eu(III) from 1M $NaNO_3$ was examined at pH 2.2–4.5 with γ-irradiated Cyanex 301/n-dodecane as a function of the total dose. The distribution data are shown in Fig. 5. The results reveal that the *D* values increase for Eu(III) with rising adsorbed dose and those of Am(III) have hardly changed in comparison to unirradiated Cyanex 301, so that the Am/Eu separation factors decrease. Up to a total dose of $1 \cdot 10^5$ Gy, Cyanex 301 seems to maintain its very good extraction properties. Whereas separation factors of 1000 were still achieved with unirradiated Cyanex 301 at pH 3.3, they decrease by two orders of magnitude to about 10 after γ-irradiation with a total dose of $7 \cdot 10^5$ Gy. As can be seen from Fig. 5, however, the Am/Eu separation factors are greatly dependent on the pH and rise from about 1 at pH 2.3 to 50 at pH 4.3 ($7 \cdot 10^5$ Gy). The decrease in selectivity can be explained by the fact that the extractant is decomposed at high γ-doses (cf. ^{31}P-NMR study) and the acid compounds Cyanex 302 and 272 are formed.

As can be seen from Fig. 1, especially the sulphur-free phosphinic acid (Cyanex 272) is responsible for this behavior because Cyanex 272 preferentially extracts the Eu with Eu/Am separation factors of 7 (pH 1–3).

However, since the alpha dose rate[5] is about $4 \cdot 10^2$ Gy/h in a solution with a ^{241}Am content of 1 g/l, it may be assumed that the extractant is stable enough to treat an An/Ln fraction originating from the high-level liquid waste (e.g. from TRUEX, TRPO or DIAMEX processes).

Table 4. Radiolytic degradation of purified Cyanex 301 as the function of adsorbed dose

Adsorbed dose	Peak area in %				
× 10^4 Gy	$\delta_{55.2}$, ppm[2]	$\delta_{67.2}$, ppm*	$\delta_{68.4}$, ppm	$\delta_{80\text{-}84}$, ppm*	$\delta_{92\text{-}94}$, ppm[1]
0.0			100.00		
0.4		22.26	71.44	5.46	0.87
10.0	1.32	35.21	48.19	15.54	1.95
70.0	3.18	25.03	35.94	39.54	3.64
100.0	4.92	5.87	17.89	64.33	6.34

The identified radiolysis products of Cyanex 301($\delta_{68.4}$ ppm) are:
[1] Cyanex 302 ($\delta_{92\text{-}94}$ppm) and [2] Cyanex 272 ($\delta_{55.2}$ ppm).
* Unknown degradation product of Cyanex 301.

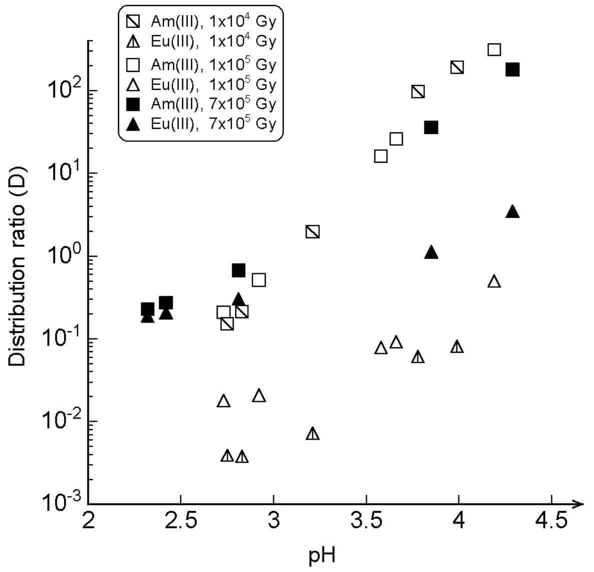

Fig. 5. Extraction of trace amounts of Am(III) and Eu(III) with γ-irradiated purified 0.5M Cyanex 301 in n-dodecane. The dose rate was $4.2 \cdot 10^3$ Gy/h and the total adsorbed doses were $1 \cdot 10^4$, $1 \cdot 10^5$ and $7 \cdot 10^5$ Gy. The initial aqueous phase was a solution of $<10^{-7}$ M ^{241}Am, ^{152}Eu and 0.001M Eu and 1M NO_3^-

Conclusions

Summarizing the results obtained with unpurified and purified Cyanex 301, the effects observed can be concluded as follows. In the case of unpurified Cyanex 301, an Am/Eu separation is only achieved by complexing the impurities which extract both Am and Eu clearly better and to the same extent by macro amounts of Eu. Purified Cyanex 301, on the other hand, shows a very high selectivity for Am(III), both with tracer amounts and also with macro amounts of Ln(III). This corresponds to the first confirmation of the results of ZHU et al. published in 1995. Since the Eu concentrations and those of the other lanthanides are >0.001 mol/l in real HLLW, it is to be assumed that the separation factors at pH values greater than 3 are very high and enable almost complete separation in a few extraction steps.

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