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Selective actinide(III) separation using 2,6-bis[1-(propan-1-ol)-1,2,3-triazol-4-yl]pyridine (PyTri-Diol) in the innovative-SANEX process: laboratory scale counter current centrifugal contactor demonstration

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Abstract: An innovative-SANEX process for the selective separation of the trivalent actinides americium and curium from a simulated PUREX raffinate solution was successfully demonstrated on the laboratory scale using a 16-stage 1 cm annular centrifugal contactor setup. The solvent was composed of 0.2 mol L⁻¹ *N,N,N',N'*-tetra-*n*-octyl-diglycolamide (TODGA) and 5% v/v 1-octanol in a kerosene diluent. Zr(IV) and Pd(II) co-extraction was prevented using *trans*-1,2-diaminocyclohexane-*N,N,N',N'*-tetraacetic acid (CDTA) as a masking agent in the feed. The actinide(III) selective

back-extraction was achieved using 2,6-bis[1-(propan-1-ol)-1,2,3-triazol-4-yl]pyridine (PyTri-Diol) in 0.45 mol L⁻¹ HNO₃ as a CHON alternative to the sulfur-containing stripping agent used in a previous version of the innovative-SANEX process. The new process described in this paper showed excellent performance for the recovery of An(III). An An(III) product with a quasi-quantitative recovery of americium and curium (>99.9%) and very good separation from fission and activation products was obtained (decontamination factors >4000). Only a slight contamination with Zr and Ru was observed. This test demonstrates the successful use of molecules containing only carbon, hydrogen, oxygen, and nitrogen atoms (so-called CHON molecules) for the selective separation of An(III) from a simulated PUREX raffinate solution. By avoiding sulfur- or phosphorous-containing molecules, the generation of secondary radioactive waste during process operation can be reduced drastically.

Keywords: actinide separation; CHON; innovative-SANEX; PyTri-Diol; TODGA.

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1 Introduction

Nuclear electricity production is currently applied in 32 countries worldwide with 439 operating nuclear power reactors and 51 reactors under construction, as of beginning 2022 [1]. Globally, used fuel in storage accumulates at an annual rate of ca. 7000 metric tons of heavy metal (t HM) and the stored inventory is approaching 300,000 t HM [2]. Additionally, ca. 3000 t HM used fuel can be treated annually in reprocessing facilities [3]. Most countries use a once-through fuel cycle, while only a few countries apply reprocessing on an industrial scale (e.g. France, Russia) for the recovery of uranium and plutonium [4]. For the future

application of nuclear electricity production and improvements of sustainability, further advancements of the nuclear fuel cycle and innovative reactor concepts are considered [5–8]. In Europe, several research projects were funded by the European Commission, addressing innovative hydrometallurgical processes for the separation of actinides from used nuclear fuel [9–17]. One of the objectives was the development of a process for the separation of the trivalent minor actinides (An(III)) americium and curium from a PUREX (Plutonium Uranium Reduction Extraction) process raffinate [18].

The extractant *N,N,N',N'*-tetraoctyl diglycolamide (TODGA, Figure 1) is the most prominent extractant of the class of diglycolamide extractants. It has a high affinity for the extraction of trivalent metal ions, especially An(III) and trivalent lanthanides (Ln(III)) [19–22]. Several separation processes have been studied using TODGA or a related diglycolamide extractant [23–34]. Commonly, An(III) and Ln(III) are effectively extracted, but the undesired co-extraction of Sr(II), Pd(II), Zr(IV) and Ru (numerous nitrosyl complexes) [35] is also observed [36]. While the co-extraction of Pd(II) and Zr(IV) can be overcome using *trans*-1,2-diaminocyclohexane-*N,N,N',N'*-tetraacetic acid (CDTA, Figure 1) as a masking agent [37], and Sr(II) can be scrubbed using less concentrated nitric acid, Ru co-extraction is still problematic and has to be addressed during solvent regeneration. Nevertheless, the co-extracted Ru is not recovered in the An(III) product [24, 26, 33, 34]. TODGA proved to be hydrolytically stable and the radiolytic stability was also found to be sufficient for long-term operation of an An(III) selective separation process [23, 38–45]. As TODGA alone extracts both An(III) and Ln(III) and a separation of the two groups is not possible, selective stripping of An(III) from the loaded solvent and separation from Ln(III) was accomplished using different approaches. On the one hand, An(III) selective stripping was

achieved using hydrophilic polyaminocarboxylic acids (e.g., diethylenetriamine-*N,N,N',N'',N'''*-pentaacetic acid (DTPA)). However, the pH of the An(III) stripping section needed to be controlled precisely in a very narrow range [25, 30, 33, 34]. The use of hydrophilic complexants containing only N-donor atoms, on the other hand, yielded much higher selectivity for An(III) and enabled stripping at higher HNO₃ concentrations compared to the polyaminocarboxylic acids [46]. The use of 2,6-bis(5,6-di(sulfophenyl)-1,2,4-triazin-3-yl)pyridine (SO₃-Ph-BTP, Figure 1) was successfully demonstrated on the laboratory scale in the innovative-SANEX process using centrifugal contactors [31]. It showed excellent performance for the recovery of An(III). The use of SO₃-Ph-BTP was an improvement over the formerly used buffered polyaminocarboxylic acid solutions, as precise pH control and salting out agents were no longer needed. SO₃-Ph-BTP was also used in the TRU-SANEX process, a combination of the EURO-GANEX [47, 48] and innovative-SANEX processes for heterogeneous recycling of the actinides from Np to Cm [49]. However, the SO₃-Ph-BTP molecule contains sulfur and is thus incompatible with the CHON principle, which describes that all chemical reagents used in a process should be totally destructible into gases, therefore only containing atoms of carbon (C), hydrogen (H), oxygen (O), and nitrogen (N) [50].

To overcome that issue, the applicability of 2,6-bis[1-(propan-1-ol)-1,2,3-triazol-4-yl]pyridine (PyTri-Diol, Figure 1) as a highly selective water-soluble stripping agent for trivalent actinides in the innovative-SANEX process was studied. It was shown that PyTri-Diol provides good separation of An(III) from Ln(III) under process-relevant conditions and good radiolytic and hydrolytic stability [51–55]. The stripping kinetics were shown to be sufficient to be used in a centrifugal contactor setup [56]. A flow sheet has been calculated using batch extraction data and kinetics data

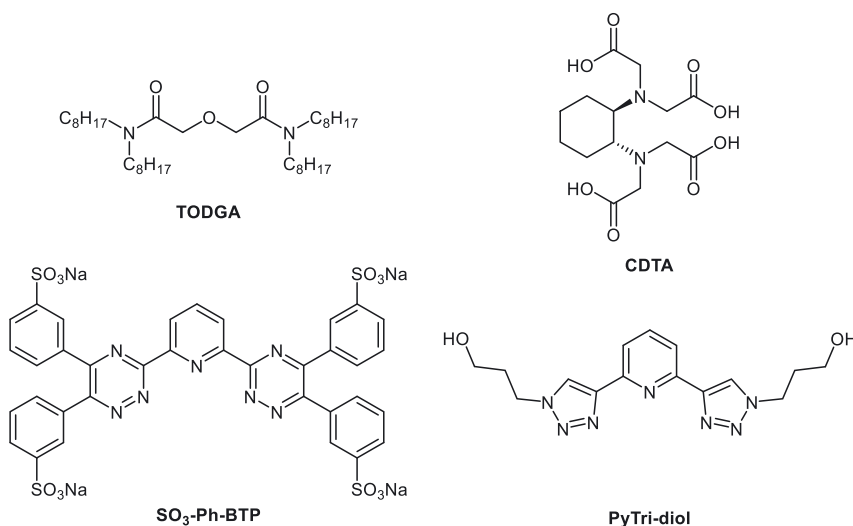


Figure 1: Chemical structures of TODGA, CDTA, SO₃-Ph-BTP, and PyTri-Diol.

Table 1: Composition of the synthetic PUREX raffinate solution used as the feed.

Component	Concentration [mg L ⁻¹ or as shown]	Component	Concentration [mg L ⁻¹]	Component	Concentration [mg L ⁻¹]
Na	2000	Al	2	Cr	106
Fe	1900	Ni	84	Cu	21
Se	6	Rb	73	Sr	184
Y	99	Zr	720	Mo	684
Ru	385	Rh	80	Pd	221
Ag	9	Cd	21	Sn	0.2
Sb	3	Te	109	Cs	532
Ba	298	La	261	Ce	594
Pr	240	Nd	757	Sm	156
Eu	35	Gd	27		
HNO ₃	4.4 mol L ⁻¹				
CDTA	0.05 mol L ⁻¹				

from the single centrifugal contactor tests with the SX Process code [56–58].

The present paper describes the results of a laboratory scale counter current demonstration of the innovative-SANEX process using PyTri-Diol as the An(III)-selective stripping agent.

2 Materials and methods

2.1 Chemicals and reagents

TODGA was purchased from Technocomm Ltd. (Wellbrae, Falkland, Scotland). PyTri-Diol was synthesized at Università di Parma, Parma, Italy, according to the procedure described elsewhere [51]. TPH (hydrogenated tetrapropene, a kerosene-like diluent commonly used in the French reprocessing facility) was obtained from CEA, Marcoule, France. CDTA (purity ≥ 99.0%) was purchased from Sigma-Aldrich, Germany. Nitric acid solutions (Merck AG) were prepared by dilution from a 65% nitric acid solution EMSURE® for analysis using ultra-pure water (18.2 MΩ cm). All commercially available chemicals were purchased in high purity and used without further purification. The radiotracers ²⁴¹Am, ²⁴⁴Cm, and ¹⁵²Eu were purchased from Isotopendienst M. Blaseg GmbH, Waldburg, Germany, Oak Ridge National Laboratory, Oak Ridge, USA, and Eckert & Ziegler Nuclitec GmbH, Braunschweig, Germany.

A synthetic PUREX raffinate was used as the feed. The composition is shown in Table 1. It was prepared by a specific protocol for the dissolution of the reagents, which is mainly based on the use of metal nitrate salts [59]. It corresponds to a PUREX raffinate with a volume of 5000 L t⁻¹ (t = metric ton) UO₂ fuel with an initial ²³⁵U enrichment of 3.5% and thermal burn-up of 33,000 MW d t_{HM}⁻¹ after 3 years of cooling [60–62]. The relatively high concentrations of Na and Fe in the raffinate are due to purification steps for the U/Pu product produced by the PUREX process and solvent clean-up steps. The initial HNO₃ concentration was 4.4 mol L⁻¹ HNO₃. 0.05 mol L⁻¹ CDTA was added as a masking agent for Zr and Pd to the innovative SANEX feed [37].

2.2 Centrifugal contactor setup

The demonstration of the process was carried out using 1 cm annular miniature centrifugal contactors produced by the Institute of Nuclear Energy Technology, Tsinghua University, Beijing, China, with the rotors made of titanium and the stator housings made of stainless steel [63, 64]. The process was run in counter current mode with a rotator speed of 4500 rpm. The speed was checked regularly during the experiment with a stroboscope tachometer. The contactor battery setup consists of four batteries with four stages each, resulting in a total available number of 16 stages. As the extraction/scrubbing sections of the innovative-SANEX process have been demonstrated in the same setup before [31], and no changes were made to the solvent composition, only the An-stripping and Ln re-extraction sections were tested in the centrifugal contactor setup. Calibrated syringe pumps (Kent Scientific Corp., Torrington, CT, USA) were used to deliver the organic and aqueous flows. The flow rates were optimized in single-stage centrifugal contactor experiments and 30 mL h⁻¹ were found to be optimal for each the An stripping, loaded solvent, and fresh solvent flows [56]. The flow rates are also given in the flow sheet shown in Figure 2.

2.3 Procedures and analytics

Batch solvent extraction experiments for the extraction and three consecutive scrubbing steps were carried out at room temperature (ca. 22°C) using equal volumes of 300 mL of each phase in glass bottles with Teflon screw caps. The bottles were shaken by hand for 30 min at room temperature for the extraction step, and 15 min at room temperature for each of the three scrubbing steps. After shaking, the bottles were left for phase separation. After phase separation was complete, the phases were sampled for analysis and the organic phase was transferred to another bottle for the next step according to the flow sheet given in Figure 2. ¹⁵²Eu, ²⁴¹Am, and ²⁴⁴Cm tracer were added in the last scrubbing step (Scrub 3) to reduce radiation dose to personnel, as the metal distribution ratios at 0.5 mol L⁻¹ HNO₃ are very high. Adding the tracers in the first extraction step would only have caused unnecessary dose with only tiny changing the composition of the loaded

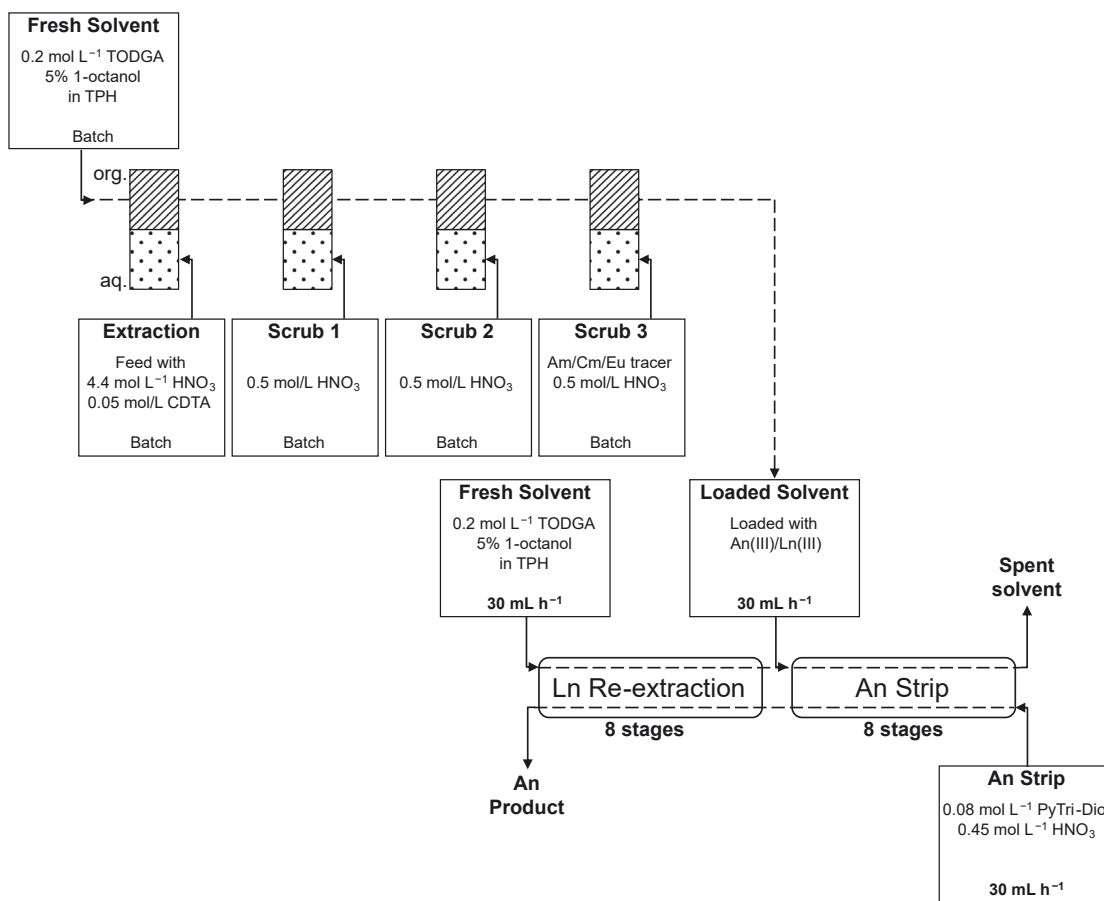


Figure 2: Flow sheet of the innovative SANEX demonstration process with PyTri-Diol.

solvent. The counter current demonstration was run in the 16-stage centrifugal contactor setup. During process operation, aliquots of the spent solvent and An product were sampled periodically to assess the evolution of the steady state. At the end of the experiment, aliquots of aqueous and organic phases were collected from each centrifugal contactor. All these samples were analyzed by gamma and alpha spectrometry, ICP-MS, and acid-base titration. Gamma measurements of ^{241}Am (60 keV) and ^{152}Eu (122 keV) were carried out using an Eurisys EGC 35-195-R germanium coaxial N-type detector and spectra were evaluated using the GammaVision Software. Samples were measured directly without further treatment. Alpha measurements were carried out for ^{241}Am (5486 keV) and ^{244}Cm (5805 keV) using an Ortec Octète-pc eight chamber alpha measurement system equipped with PIPS detectors. Sample preparation for alpha measurement was done by homogenizing a 10 μL aliquot in 100 μL of a mixture of Zapon varnish and acetone (1:100 v/v). This mixture was distributed over a stainless-steel plate obtained from Berthold, Bad Wildbad, Germany. The sample was dried under a heating lamp and annealed into the stainless-steel plate using a gas-flame burner. For stable elements, inductively coupled plasma mass spectrometry (ICP-MS) was conducted using a Perkin Elmer NexION 2000C. The ICP-MS measurements were calibrated against certified metal solution standards. Aqueous samples were measured after dilution in 1% v/v nitric acid solution without further treatment. Organic samples were measured directly in a matrix containing the surfactant Triton-X-100 in 1% v/v HNO_3 . The ICP-MS

measurements of several elements (Eu, Am, Cm) were compared with measurements from other analytical methods and found to be in very good agreement. Acid concentration was measured by titration against 0.1 mol L^{-1} or 0.10 mol L^{-1} standardized NaOH solutions using a 798 MPT Titrimo, purchased from Metrohm GmbH & Co. KG (Filderstadt, Germany).

Distribution ratios D were calculated as the ratio of activity or concentration of a metal ion in the organic phase versus the activity or concentration of the metal ion in the aqueous phase. Distribution ratios between 0.01 and 100 exhibit an uncertainty of $\pm 5\%$, while lower/higher values exhibit larger uncertainties.

To evaluate the results of the full counter current centrifugal contactor demonstration test, process decontamination factors, $DF_{\text{feed}/\text{An product}}$ were calculated according to Eq. (1), where Q is the volumetric flow rate and C is the metal ion (M) concentration. The An/M decontamination factors were calculated according to Eq. (2). For the evaluation of the demonstration test discussed in this paper, the loaded solvent composition and flow rates were considered as feed, according to Eqs. (1) and (2).

$$DF_{\text{feed}/\text{An product}} = \frac{Q_{\text{feed}} \cdot C(M)_{\text{feed}}}{Q_{\text{An product}} \cdot C(M)_{\text{An product}}} \quad (1)$$

$$DF_{\text{An/M}} = \frac{C(M)_{\text{feed}} \cdot C(\text{An})_{\text{product}} \cdot Q_{\text{feed}}}{C(M)_{\text{product}} \cdot C(\text{An})_{\text{feed}} \cdot Q_{\text{An product}}} \quad (2)$$

Table 2: Composition of the loaded solvent used as the organic feed for the centrifugal contactor test.

Component	Concentration [mg L ⁻¹ or as shown]	Component	Concentration [mg L ⁻¹]	Component	Concentration [mg L ⁻¹]
Na	≤10 ^a	Al	≤0.1 ^a	Cr	≤0.1 ^a
Fe	≤1 ^a	Ni	≤0.1 ^a	Cu	≤0.1 ^a
Se	≤0.1 ^a	Rb	≤0.1 ^a	Sr	≤0.1 ^a
Y	82	Zr	18	Mo	≤0.1 ^a
Ru	97	Rh	≤0.1 ^a	Pd	≤0.1 ^a
Ag	≤0.1 ^a	Cd	≤0.1 ^a	Sn	≤0.1 ^a
Sb	≤0.1 ^a	Te	≤0.1 ^a	Cs	≤0.1 ^a
Ba	≤0.1 ^a	La	90	Ce	314
Pr	159	Nd	612	Sm	145
Eu	33	Gd	23		
¹⁵² Eu	10.4 MBq L ⁻¹				
²⁴¹ Am	5.7 MBq L ⁻¹				
²⁴⁴ Cm	4.4 MBq L ⁻¹				
HNO ₃	0.02 mol L ⁻¹				

^adetection limit.

3 Results and discussion

The innovative-SANEX with PyTri-Diol process is based on the innovative-SANEX process using SO₃-Ph-BTP, which was demonstrated previously [31]. It uses an identical solvent composition (0.2 mol L⁻¹ TODGA and 5% v/v 1-octanol in TPH). As only 16 contactors are available in the laboratory setup, only the An stripping and Ln re-extraction sections were actually run in the centrifugal contactor battery, as PyTri-Diol is only used in those stages. The loaded solvent used as the organic feed solution for this demonstration was produced by batch contacts, resembling the extraction and scrubbing steps of the innovative-SANEX process, as these steps were already demonstrated [31] and are not within the scope of this work. Therefore, the solvent was contacted with the synthetic PUREX raffinate solution (composition given in Table 1), containing 0.05 mol L⁻¹ CDTA to prevent Zr and Pd co-extraction [31, 37]. Consecutively, the loaded solvent was washed three times with 0.5 mol L⁻¹ HNO₃. The TODGA solvent is known to co-extract a certain amount of nitric acid [65, 66], which was considered in the flow sheet development [56]. As the TODGA solvent extracts An(III) and Ln(III) even from 0.5 mol L⁻¹ HNO₃ with high distribution ratios, the radioactive tracer elements ¹⁵²Eu, ²⁴¹Am, and ²⁴⁴Cm were added in the third scrubbing step only, to reduce radiation dose to personnel. The loaded organic phase from the third scrubbing step was used as the organic feed for the centrifugal contactor test. The composition of the loaded solvent after the third scrubbing step is given in Table 2. The loaded solvent, which was prepared by batch contact here, compares well with the loaded solvent produced in

the counter current centrifugal contactor test of the prior innovative-SANEX test [31]. Most of the feed elements were not extracted and thus not detected in the loaded solvent. In comparison to the prior test, Pd, Mo, and Sr were also not detected, while the Zr concentration was slightly, and the Ru concentration significantly higher. These effects were probably caused by a longer contact time and more efficient mixing in the batch contacts compared to the centrifugal contactor tests. The higher Zr concentrations can be explained by the very slow Zr extraction kinetics with TODGA in the presence of CDTA, which was observed before and seems to be due to very slow decomplexation kinetics of Zr-CDTA complexes in the aqueous phase [37]. The concentration of some light lanthanides was lower in the loaded solvent (esp. La and Ce), as the light lanthanides are least extracted with TODGA [20].

The An stripping and Ln re-extraction sections were run in the 1 cm annular miniature centrifugal contactor setup, installed in the laboratories of Forschungszentrum Jülich, Germany. The tested flow sheet is shown in Figure 2. The Ln stripping was not tested here, as the previous innovative-SANEX demonstration showed that Ln(III) back extraction can easily be achieved in four stages using 0.5 mol L⁻¹ citric acid at pH 3 [31].

The outlets of the centrifugal contactor battery (An product and spent solvent) were monitored during process operation by sampling and quick gamma measurements of ²⁴¹Am and ¹⁵²Eu. These preliminary gamma spectrometry results were used to assess the approach to steady state during the test. They showed very quick development of a constant ¹⁵²Eu concentration in the spent solvent, already after 60 min run time. The ²⁴¹Am concentration in the An

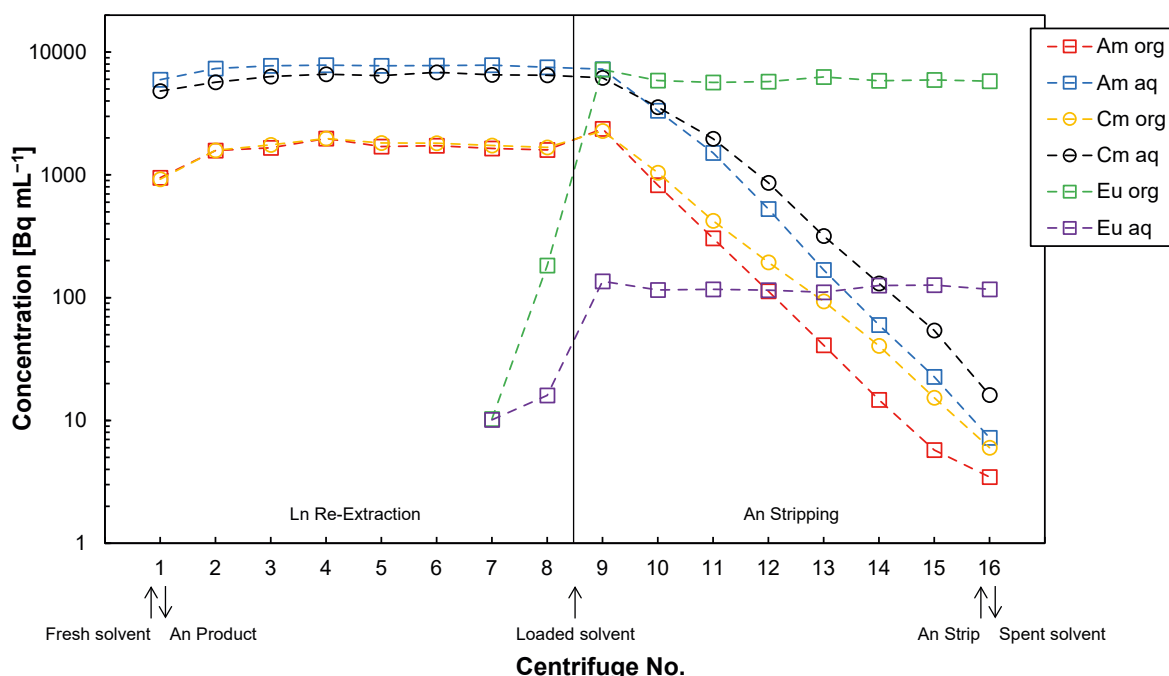


Figure 3: Am, Cm, and Eu stage profiles of the innovative-SANEX with PyTri-Diol demonstration test (^{241}Am and ^{152}Eu data from gamma spectrometry and ^{244}Cm data from alpha spectrometry measurements).

product increased more slowly and reached a plateau after 240 min. The centrifugal contactor test was continued further to enable built-up of a steady state also for the elements, which could not be monitored during the test by gamma spectrometry and stopped after 360 min. Full analysis by gamma and alpha spectrometry and ICP-MS showed that the steady-state had been reached for all elements already after 240 min. No hydrodynamic problems (phase entrainment, third phase formation, or precipitation) were observed during the process demonstration. After stopping the test (stopping pumps and contactors), the contents of the mixing chambers were quickly transferred to test tubes and centrifuged to achieve quantitative phase separation. Both phases were sampled and analyzed by gamma and alpha spectrometry and ICP-MS, and the HNO_3 concentration was measured. The results of several elements analyzed by different analytical techniques were compared to each other and found to be in very good agreement (e.g., Am profiles determined by gamma, alpha, and ICP-MS measurements were in very good agreement).

The Am, Cm, and Eu stage profiles of the innovative-SANEX with PyTri-Diol demonstration test are shown in Figure 3. Am and Cm were both stripped very well from the loaded solvent with PyTri-Diol and showed distribution ratios of ca. 0.23 and 0.28, respectively, in all stages. Both elements were efficiently routed to the An product and

separated from the lanthanides. The slightly better stripping of Am(III) in comparison with Cm(III) was observed previously [56] and is reflected in the stage profiles of stages 11–16. The An(III) distribution ratios observed here were slightly lower compared to the single centrifugal contactor test [56]. This was probably due to a slightly lower HNO_3 concentration in the loaded solvent compared to the loaded solvent used in the single centrifugal contactor test. Here, the solvent was scrubbed three times with $0.5 \text{ mol L}^{-1} \text{ HNO}_3$, while it was only scrubbed once during the single centrifugal contactor test. The average aqueous nitric acid concentration in all stages was $0.46 \text{ mol L}^{-1} \text{ HNO}_3$ and varied only slightly.

Europium was not stripped and stayed in the organic phase with distribution ratios of ca. 50. Therefore, it was routed to the spent solvent outlet. The other Ln(III) were also routed to the spent solvent, as shown in Figure 4 for the least extracted elements La, Ce, and Pr (average distribution ratios of 2.4, 3.3, and 4.6, respectively). The An(III)/Ln(III) separation with PyTri-Diol worked very well and the number of stages for An stripping and Ln re-extraction were sufficient. In comparison with the prior innovative-SANEX test using $\text{SO}_3\text{-Ph-BTP}$, four more stages were used (8 + 8 stages here, instead of 6 + 6 stages in the prior test for An stripping and Ln re-extraction), as PyTri-Diol shows a lower An(III)/Ln(III) selectivity compared to $\text{SO}_3\text{-Ph-BTP}$ [46, 51, 56].

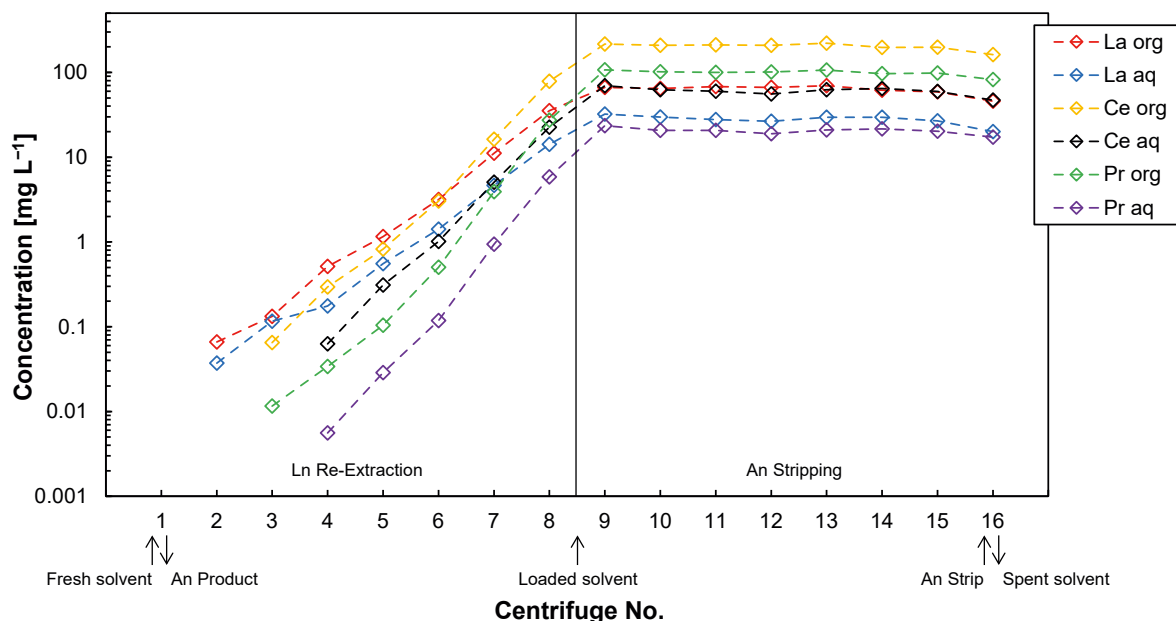


Figure 4: La, Ce, and Pr stage profiles of the innovative-SANEX with PyTri-Diol demonstration test (data from ICP-MS measurements).

Table 3: Mass balances, recoveries, process, and An/M decontamination factors (DF) obtained during the innovative-SANEX with PyTri-Diol test (only the elements detectable in the loaded solvent are shown, cf. Table 2).

Component	An product [%]	Spent solvent [%]	$DF_{\text{feed}/\text{An product}}$	$DF_{\text{Am}/\text{M}}$	$DF_{\text{Cm}/\text{M}}$
Y	≤ 0.1	≥ 99.9	≥ 5000	≥ 5000	≥ 5000
Zr	10	90	10	11	10
Ru	5	95	20	22	20
La	≤ 0.1	≥ 99.9	≥ 4000	≥ 4000	≥ 4000
Ce	≤ 0.1	≥ 99.9	≥ 5000	≥ 5000	≥ 5000
Pr	≤ 0.1	≥ 99.9	≥ 5000	≥ 5000	≥ 5000
Nd	≤ 0.1	≥ 99.9	≥ 5000	≥ 5000	≥ 5000
Sm	≤ 0.1	≥ 99.9	≥ 5000	≥ 5000	≥ 5000
Eu	≤ 0.1	≥ 99.9	≥ 5000	≥ 5000	≥ 5000
Gd	≤ 0.1	≥ 99.9	≥ 5000	≥ 5000	≥ 5000
^{241}Am	≥ 99.9	≤ 0.1	1.0	1.0	0.9
^{244}Cm	≥ 99.9	≤ 0.1	1.0	1.1	1.0

The An(III) product was very clean with only very low contaminations as shown in Table 3. The only measurable contaminants in the product were Zr and Ru. The Zr and Ru stage profiles are shown in Figure 5. Both elements showed high distribution ratios in the An stripping section and low distribution ratios in the Ln re-extraction section and the stage profiles were flat in the corresponding sections. Comparable stage profiles were observed for Ru in the prior innovative-SANEX process (the Zr concentration was too

low) [31]. For both elements, this behavior could be explained by the existence of different Zr and Ru species, which have different solubility in the aqueous and organic phases, respectively. The existence of different zirconium and ruthenium species and their different extraction behavior in solvent extraction systems has been studied previously (mainly under PUREX process conditions), ascribed to different hydroxide, nitrate, nitrite and other nitrous oxide (mixed) complexes [35, 67–71]. Further process development has to be conducted to address these issues, although the higher Zr concentration in the current process demonstration was due to the batch preparation of the loaded solvent and is expected to be less impacting in a counter current process including also the extraction and scrubbing stages. The loaded solvent in the prior innovative-SANEX process contained much lower Zr concentrations [31].

The separation from Ln(III) worked very well and the corresponding An(III)/Ln(III) decontamination factors were very high ($\geq 4,000$, Table 3). The lanthanide distribution ratios in the An stripping and Ln re-extraction sections were well above one ($D_{\text{La}} \geq 2.0$) and followed the known extraction pattern (i.e. increasing distribution ratios with decreasing ionic radius) [20, 56]. The Ln(III) distribution ratios observed in the present demonstration test and stage profiles in the An stripping and Ln re-extraction sections are comparable to the ones of the prior innovative-SANEX test [31]. The performance of the present

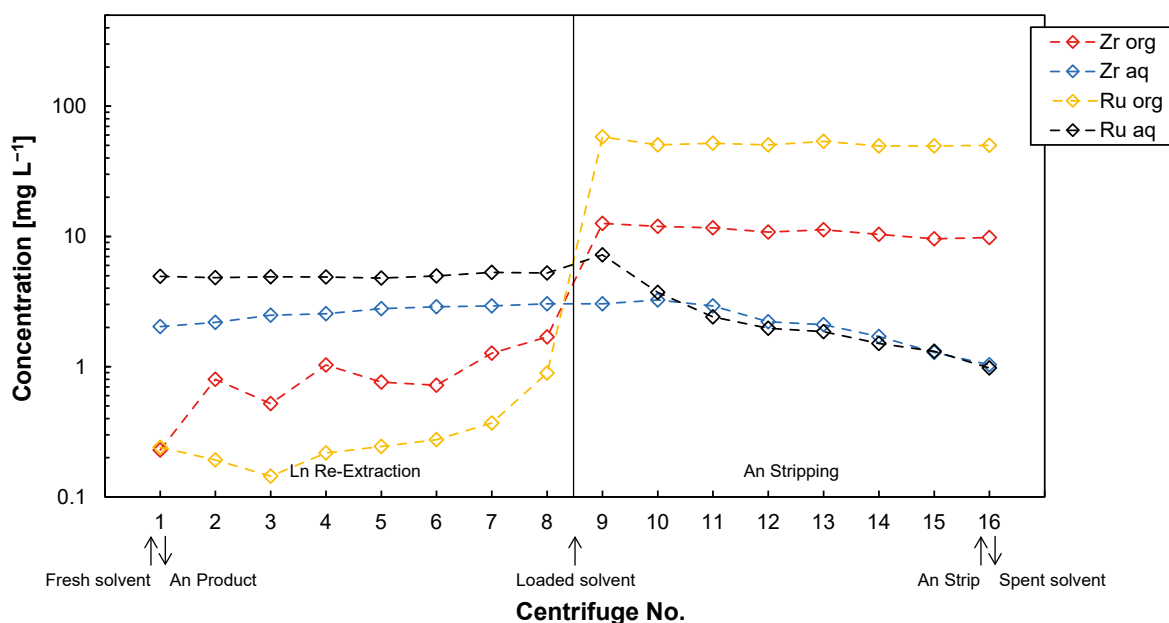


Figure 5: Zr and Ru stage profiles of the innovative-SANEX with PyTri-Diol demonstration test (data from ICP-MS measurements).

demonstration test exceeded the expectations from flow sheet calculations [56]. The use of six stages each for An stripping and Ln re-extraction (as in the prior innovative-SANEX test) would probably result in a loss of 0.5% Am and 0.9% Cm to the spent solvent, but a contamination of the An(III) product with light lanthanides would still be negligible (ca. 0.1% La(III), all other Ln(III) even lower). PyTri-Diol therefore is a very good alternative stripping agent for the recovery of An(III) from PUREX raffinate, which also fulfills the CHON criterion.

4 Conclusions

The innovative-SANEX with PyTri-Diol process demonstration described in this paper showed excellent performance for the recovery of An(III) from a simulated feed solution using the same TODGA-based solvent as in the prior innovative-SANEX process. With the new process, the separation of An(III) in a single cycle becomes possible using chemicals made of C, H, O, and N atoms, using only four stages more than in the prior process. This is a great improvement over the prior innovative-SANEX process, which used a sulfur-containing stripping agent. Therefore, the generation of secondary radioactive waste during process operation can be reduced drastically. The successful demonstration of the use of PyTri-Diol also promises its usability in other processes, e.g., EURO-GANEX [48], TRU-SANEX [49] or TALSPEAK [72].

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Conflict of interest statement: The authors declare no conflicts of interest regarding this article.

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