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ISSN: 0736-6299 (Print) 1532-2262 (Online) Journal homepage: www.tandfonline.com/journals/lsei20

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To cite this article: Andreas Geist & Giuseppe Modolo (01 Sep 2026): Thirty-Plus Years of Solvent Extraction Development for Minor Actinide Separations in Europe, Solvent Extraction and Ion Exchange, DOI: 10.1080/07366299.2026.2721488

To link to this article: https://doi.org/10.1080/07366299.2026.2721488

Published online: 01 Sep 2026.

SOLVENT EXTRACTION AND ION EXCHANGE
https://doi.org/10.1080/07366299.2026.2721488

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REVIEW

Thirty-Plus Years of Solvent Extraction Development for Minor Actinide Separations in Europe

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ABSTRACT

Recycling used nuclear fuel contributes significantly to the sustainability of nuclear energy production. The well-established PUREX process to separate uranium and plutonium is pivotal to this recycling, minimizing the need for uranium mining and milling. Additionally separating and recycling the minor actinides — most notably americium — further contributes to this goal: By reducing the long-term decay heat of the final waste to be disposed of in a deep geologic repository, its volume is utilized more efficiently. Internationally, efforts are underway to develop novel solvent extraction processes to separate americium from used nuclear fuels. For more than thirty years, collaborative European research programs have dealt with the development of solvent extraction systems and flowsheets. Initially, a suite of three consecutive processes (An(III)/Ln(III) co-extraction, An(III)-Ln(III) separation, and Am(III)-Cm(III) separation) was required to obtain an americium product. This was later reduced to two and finally only one process, directly extracting americium from the PUREX raffinate solution. Alternatively, processes to co-separate neptunium, plutonium, americium, and curium were also developed, addressing the so-called homogeneous recycling strategy. This paper summarizes the progress made in past and current EURATOM programs on actinides separations, focusing on the development of new extractants and flowsheet trials. Without steady funding and a highly motivated and collaborative team this progress certainly would not have been achieved.

KEYWORDS

EURATOM research programmes; actinides; separations; advanced nuclear fuel cycle

In memory of Ken Nash

From their first days in the field of solvent extraction and nuclear separations, the authors have associated the name *Kenneth L. Nash* with outstanding research in the field of solvent extraction. Ken was a long-standing Editor and Editor-in-Chief of *Solvent Extraction & Ion Exchange* and a frequent contributor to the journal. His contribution to the field of *f*-element separations, in the form of much-quoted reviews[1–4] and original papers,[5–8] was

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outstanding. For many years, Ken had driven the development of actinide separations by studying the thermodynamics and kinetics of known systems and developing novel improved complexing and extracting agents. In particular,[3,7] he paved the way from describing to understanding TALSPEAK ("Trivalent actinide lanthanide separation with phosphorus-reagent extraction from aqueous komplexes")[9] chemistry. Ken shared his knowledge through numerous publications in journals, conference proceedings, and books. He also trained many students and postdoctoral researchers, many of whom have become experts in solvent extraction themselves.

While writing this paper, we received the sad news that Ken had passed away. He will be greatly missed. In his honor, we gladly contribute to this Special Issue of the *Premier Solvent Extraction Journal*.

Introduction

With over 400 nuclear power reactors currently in operation, approximately 10,000 tons of used nuclear fuel are discharged annually. Most countries follow a *once-through fuel cycle*, preparing for the direct disposal of used nuclear fuel in deep geologic repositories.

Alternatively, used fuel can be *mono-recycled*, as is currently the case in France. The fuel is dissolved; uranium and plutonium are separated by the well-established PUREX ("Plutonium Uranium Reduction Extraction") process or evolutionary designs thereof.[10–13] Separated plutonium and depleted uranium are fabricated into mixed-oxide (MOX) fuel.[14] The highly radioactive raffinate solution from the PUREX process (containing the fission products and the minor actinides, neptunium, americium, and curium) is vitrified, to be finally disposed of in a deep geologic repository. Used MOX fuel may either be disposed of in a deep geologic repository or stored pending decisions on further recycling.

Repeating this cycle multiple times – *multi-recycling* – significantly contributes to the sustainability of nuclear energy production, virtually eliminating the need for uranium mining and milling with its associated environmental impact. Thus, the resource *natural uranium* is utilized much more efficiently.[15–18]

The minor actinides and some long-lived fission products could additionally be recycled – the *partitioning and transmutation* (P&T) concept.[19] P&T would result in a final waste product the radiotoxicity of which decays to the level of natural uranium within a few centuries (as compared to several 100,000 years without any separation or of several 10,000 years if only uranium and plutonium were recycled).

P&T has long been discussed as an option to fully close the nuclear fuel cycle. In the 1970s and 1980s, the P&T concept's potential was viewed with some skepticism. A 1972 report concluded on one hand that burning

fission products would not be practical; on the other hand, "When plutonium and uranium extraction efficiencies exceed 99%, a significant reduction in the long-term hazard potential of the waste can be obtained by similar removal of neptunium, americium, and curium ...".[20] Summarizing the 1977 state of P&T in OECD countries, "The major benefits of P-T, if any, would be expected to arise from the reduction in the long-term toxicity of the waste in a repository. This would, in turn, result in reduced consequences for any assumed repository release incident and might enable the repository to be built to less stringent requirements. Another advantage of P-T would be that the recycled actinides could be used as fuel in an LMFBR", (liquid-metal-cooled fast breeder reactor) "thus decreasing resource requirements. Implementation of P-T may also lead to perceived, but not necessarily real, benefits if waste disposal operations are made more acceptable to the public because of P-T".[21] The US P&T program was assessed in 1980, concluding on one hand feasibility but on the other hand stating "There are no incentives for actinide P-T, even if very conservative assumptions are used in the analysis".[22]

Following a re-think, programs were launched such as the Japanese *Options Making Extra Gains from Actinides and Fission Products* (OMEGA, initiated in 1988),[23] the French *Séparation-Incinération* (SPIN)[24,25] program (based on the French *Loi du 30 décembre 1991 sur la gestion des déchets nucléaires* – waste management act from 1991), the US *Advanced Fuel Cycle Initiative* (AFCI)[26] and similar projects in Russia, India, and China,[27] which included the P&T concept. While these programs all have their distinct flavors, a certain degree of cross-fertilization cannot be denied. For example, the processes applying selective stripping of actinides as discussed in the ACSEPT and following sections *(cf.* below) were inspired by the TALSPEAK concept.[9] Based on earlier work,[28] diglycolamides were first applied in Japan.[29] Nowadays diglycolamides are widely used to co-extract actinides and lanthanides. Polydentate nitrogen donor ligands such as bis-triazinyl-(bi)pyridines are applied universally to separate trivalent actinides from lanthanides. A breakthrough in this field[30] was achieved within the NEWPART program *(cf.* below).

The discussion relating to P&T has mainly hinged on the *reduction of long-term radiotoxicity* of the waste to be finally disposed of. However, the presence or absence of actinides in a deep geologic repository may be perceived as a lesser issue compared to the presence of some long-lived and rather mobile fission products such as I-129, C-14, Se-79, Tc-99.[31,32] Nevertheless, a further aspect deserves consideration: separating and recycling (and thus fissioning) the minor actinide americium reduces the long-term thermal power of the vitrified radioactive waste to be finally disposed of. Less heat means denser packing is possible, resulting in a more efficient utilization of the resource *final*

repository volume.[33] The benefits of a closed fuel cycle are emphasized in a recent comparison of fuel cycle strategies.[34]

In summary, the elements to be treated in a future advanced fuel cycle would be plutonium and americium; the former to reduce natural uranium consumption (and thus mining), the latter for a more efficient utilization of a final repository. Plutonium can be separated by PUREX or similar processes, and americium by novel processes which are under development but not yet applied on an industrial scale. Internationally, efforts have been made to develop processes for the separation of americium from used nuclear fuels. Such processes may be based on pyrometallurgy[35] or hydrometallurgy *(i.e.* solvent extraction). Owing to the extensive industrial experience with the PUREX process, and compatibility requirements for downstream processes, solvent extraction may be the method of choice.

The challenge is that, at some point, Am(III) must be separated from the chemically similar fission lanthanides, Ln(III), some of which are strong neutron absorbers. As early as 1950,[36] the separation of Am(III) (and Cm(III)) from Ln(III) was achieved by sequential elution from an ion exchange column with hydrochloric acid, based on a stronger interaction of the soft chloride anion with Am(III) and Cm(III) compared to the Ln(III).[1] Since a chromatographic An(III)-Ln(III) separation may be viewed as "most probably a workable, but cumbersome, process",[22] the path at large turned toward solvent extraction, with the task at hand finding suitable extracting agents. The journey from the preferential complexation of Am(III) over Ln(III) by azide and by 1,10-phenanthroline[37] via tris-pyridinyl-triazines[38] to the breakthrough bis-triazinyl-pyridines[30] and similar selective nitrogen-donor extracting and complexing agents has thoroughly been reviewed, see e.g. references.[39–43]

However, a chemical system with proper selectivity does not constitute a separations process. Many more constraints such as reversible extraction (which would be precluded by too-high distribution ratios), fast kinetics, sufficient stability, loading capacity, phase disengagement, etc. must be considered. Although these parameters can be determined in shaking tube batch tests, a final test in a continuous counter-current setup is next step toward a process implementation. Thus, the most promising chemical systems have undergone flowsheet development and testing on the laboratory scale, using small mixer-settler or centrifugal contactor rigs.[2] Figure 1 shows a generic flowsheet of a counter-current process. In some cases, "hot" feed solutions prepared from dissolved used nuclear fuel have been processed.

[1]The paper states "This suggests that in addition to the interactions which the rare earths show with chloride ion perhaps americium and curium form weak covalent coordination complexes involving large numbers (say 6) of chloride ions". — a very early mentioning of a soft donor/covalence-driven An(III)-Ln(III) selectivity, a concept thoroughly discussed nowadays.

[2]While pulsed columns are the equipment of choice in commercial reprocessing plants, they have not been utilized in the framework of the EURATOM research programs discussed in this paper.

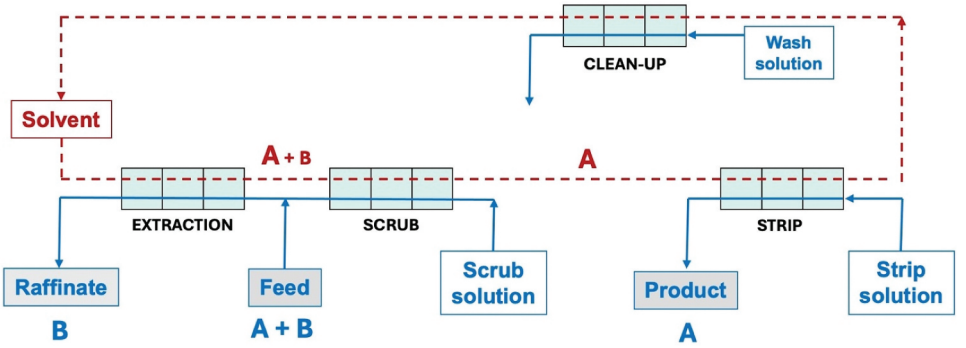

Figure 1. Typical flowsheet of a counter-current solvent extraction process. Components *A* and *B* present in the feed solution are to be separated. *A* and a fraction of *B* are co-extracted in the *extraction* section. The *scrub* section backwashes *B* from the solvent, routing it to the raffinate solution. The solvent loaded with *A* is routed to the *strip* section, where *A* is back extracted into the product solution. The solvent is recycled following clean-up. Each section consists of multiple stages.

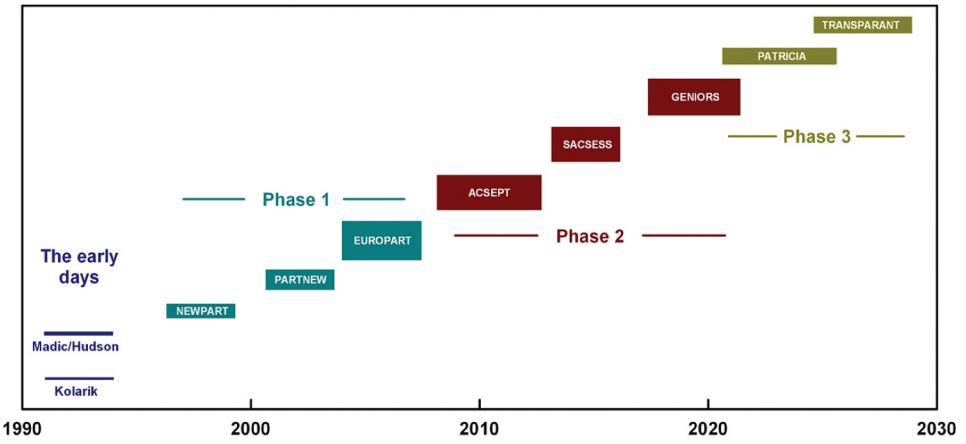

Figure 2. Timeline of the European programs on minor actinide separations. Symbol height is proportional to the number of laboratories involved in solvent extraction and related topics. Phase 1, Phase 2, Phase 3 relate to the programs' different coordinators.

Much of the process development performed in European projects has been reviewed[44,45]; a broader picture is given in references.[46–48] However, there is still a long way to go toward separating americium *on and industrial scale.*

In Europe, actinides recycling was brought back into the limelight within the framework of the SPIN program *(cf.* below). This in turn kindled the programs on (minor) actinide separations funded by the European Atomic Energy Community (EURATOM) discussed below with a focus on the development of new extractants and on flowsheet trials. Figure 2 is a graphical representation of the timeline.

Over decades, a huge body of work has been amassed. Note that many of the programs discussed included topics other than solvent extraction, which are not in this paper's scope. We chose including what – in our opinion – would make a useful summary of *Thirty-plus Years of Solvent Extraction Development for Minor Actinide Separations in Europe*.

SPIN and spin-off

The SPIN program

To comply with the French *Loi du 30 décembre 1991 sur la gestion des déchets nucléaires*, the *Séparation-Incinération* (SPIN) program[24] was initiated in the early 1990s by the French *Commissariat à l'énergie atomique* (CEA, now *Commissariat à l'énergie atomique et aux énergies alternatives)*. Within SPIN, separations strategies were devised[25] to separate some fission products identified as troublesome, plus americium. With respect to separating americium, one option would be oxidizing to Am(IV) or Am(VI),[49,50] allowing for its separation from Cm(III) and Ln(III) by common extracting agents. Another option would be co-separating An(III) and Ln(III) in a first step, followed by their separation in a second step. A third step to separate Am(III) and Cm(III) would be required.

For the latter option, as shown in Fig. 3, the PUREX process would be modified to allow for additionally separating neptunium, iodine, technetium, and zirconium. The PUREX raffinate would then be fed to a DIAMEX (Diamide extraction)[52] or TRUEX (Transuranium extraction)[53] process to co-extract Am, Cm, and fission lanthanides, separating them from other fission products and corrosion products (henceforth tagged *An(III)-Ln(III) co-extraction)*. This process's raffinate would be subjected to a cesium separation process, while the product solution would be routed to the "New extractants or TALSPEAK" box, where americium and curium would be separated from fission lanthanides (henceforth *An(III)-Ln(III) separation)*. Finally, the SESAME process[54] would separate americium (oxidized to Am(IV, VI)) from curium *(henceforth Am(III)-Cm(III) separation)*.

SPIN-off: EURATOM research programs

France's decision to reconsider the P&T strategy was the driving force that led to complementary projects in the European Union's nuclear R&D program. Without the *Loi du 30 décembre 1991*, the European research programs discussed below (which were built on the ground broken by the SPIN program) would likely never have come to happen.

The post-PUREX separation strategy adopted in those programs was

(1) An(III)-Ln(III) co-extraction followed by
(2) An(III)-Ln(III) separation and finally
(3) Am(III)-Cm(III) separation.

In the course of the programs, this rather complex three-cycle strategy (which certainly would not be the preferred choice for industrialisation due to its complexity; e.g. each of the three cycles would additionally require its own spent solvent handling, treatment, and regeneration section) was simplified to two-cycle concepts and finally to a one-cycle process directly extracting only americium from the PUREX raffinate.[55] These concepts are described and compared in a recent review.[56] Since none of cesium, neptunium, iodine, technetium, and zirconium contribute significantly to long-term radiotoxicity or long-term thermal output, separating them was no longer the primary scope.

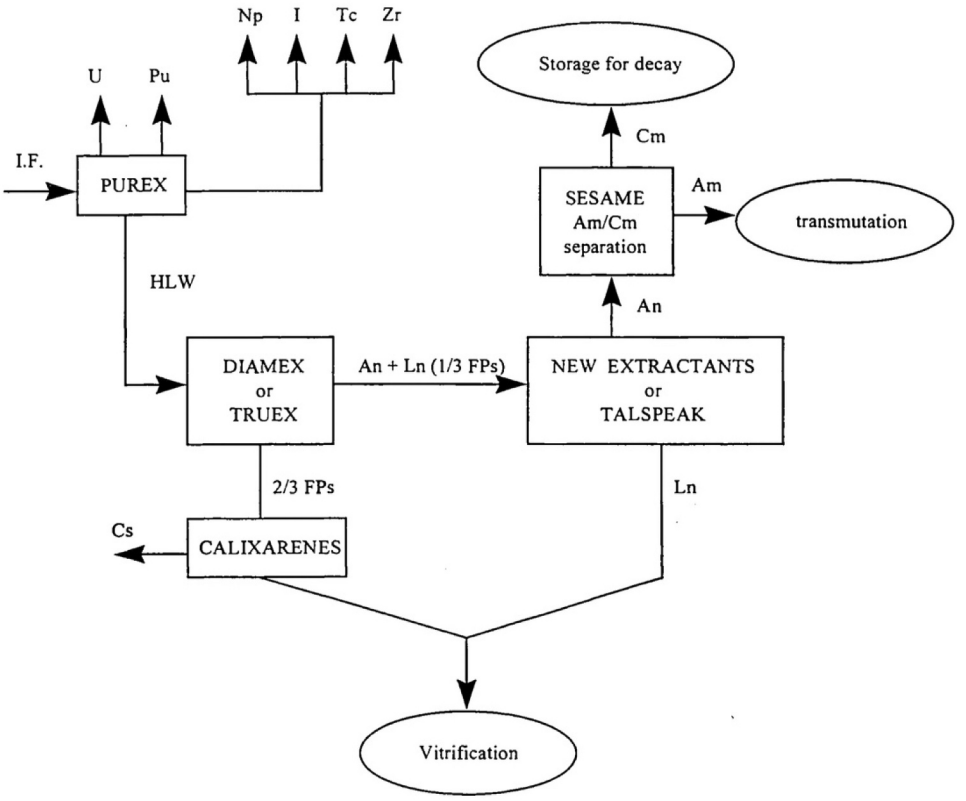

Figure 3. Scheme for separating minor actinides and some fission products from used nuclear fuel as developed during the early 1990s within the SPIN program. Figure from reference[51].

The early days: 1990–1994

Two rather small programs were executed in the early 1990s: *High-level liquid waste partitioning by means of completely incinerable extractants*[51] by Charles Madic from the CEA and Michael Hudson from Reading University, UK, and *Partition of high-level radioactive waste*[57] by Zdenek Kolarik *et al.* from *Forschungszentrum Karlsruhe*, Germany.

Madic and Hudson

Madic and Hudson[51] focused on An(III)-Ln(III) co-extraction and on An(III)-Ln(III) separation. They postulated that all chemicals used in a solvent extraction process should consist of only C, H, O, and N atoms (i.e. combustible to only gaseous products) to avoid secondary solid wastes – the *CHON* principle.

Alkylated malonamides[58] were studied for An(III)-Ln(III) co-extraction. *N,N'*-dimethyl-*N,N'*-dibutyl-2-tetradecyl-malonamide (DMDBTDMA, Fig. 4 left)[59] was selected for further process development, being applied both in cold and hot counter-current flowsheet tests.[60]

Based on earlier work,[38] 2,4,6-tris (4□alkyl-pyridin-2-yl)-1,3,5-triazine (TPTz) compounds (Fig. 4 right) were studied to separate An(III) from Ln(III). Since TPTz do not extract metal nitrates, a lipophilic anion source such as 2-bromodecanoic acid must be added to the solvent. As this conflicts with the *CHON* principle, alternative carboxylic acids such as 2-cyano and 2-hydroxy carboxylic acids were evaluated. Unfortunately, none of them was sufficiently soluble in kerosene, the diluent of choice. Separation factors for Am(III) over Eu(III) in the order of $SF_{Am/Eu}$ ≈10 were reported. Unfortunately, no distribution data for Ln(III) other than Eu(III) were reported, leaving the question unanswered whether Am(III) (and Cm(III)) could be separated from *all* fission Ln(III).

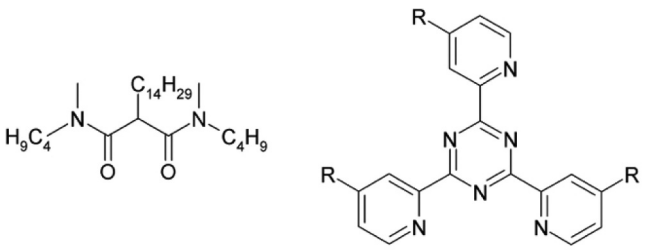

Figure 4. *N,N'*-dimethyl-*N,N'*-dibutyl-2-tetradecyl-malonamide (DMDBTDMA) and 2,4,6-tris(4-alkyl-pyridin-2-yl)-1,3,5-triazine (TPTz).

Kolarik et al.

Kolarik *et al.*[57] put less focus on An(III)-Ln(III) co-extraction, merely studying alternative modifiers for a carbamoylmethyl phosphine oxide (CMPO)[61] solvent. Regarding An(III)-Ln(III) separation, three pathways were studied: a) extracting with CMPO from media other than nitrate (e.g. a moderate increase in Am(III)/Eu(III) selectivity was observed when extracting from iodide or thiocyanate media, $SF_{Am/Eu} \approx 10$); b) extracting with di(2-ethylhexyl)dithiophosphoric acid and TBP (tri-*n*-butyl phosphate),[62] showing selectivity only in the presence of macro concentrations of Eu(III)[3]; c) extracting with nitrogen donor ligands: a vast number of ligands and synergists in various diluents were tested with separation factors typically in the order of $SF_{Am/Eu} \approx 10$, unfortunately at pH values incompatible with upstream processes.

Phase 1, 1996–2007

The late Charles Madic[63] coordinated three consecutive programs, NEWPART,[64] PARTNEW,[65] and EUROPART.[66] It was during NEWPART that the authors of this paper had the opportunity to join as postdoctoral researchers. During these years, Charles shared with us his vast knowledge and experience, and his enthusiasm catalyzed new ideas.

NEWPART: breakthrough

New Partitioning Techniques for Minor Actinides (NEWPART, 05/1996 – 04/1999)[64] joined a team of eight from France, the UK, Sweden, Germany, Italy, and the European Joint Research Centre. An(III)-Ln(III) co-extraction and An(III)-Ln(III) separation (under the acronyms of DIAMEX, diamide extraction, and SANEX, selective actinides extraction) were the topics, covering both fundamental (new extractants, distribution and thermodynamic data, structural studies) and process development studies. More than 100 molecules were synthesized and screened as extracting agents. NEWPART saw a significant breakthrough in the field of An(III)-Ln(III) separation.

An(III)-Ln(III) co-extraction

Over the years, CEA worked at optimizing the malonamide structure with respect to its solubility in kerosene diluents, loading capacity, degradation and clean-up behavior, and affinity for An(III) and Ln(III). Finally, *N,N*'-dimethyl -*N,N*'-dioctyl-2-[2-(hexyloxy)ethyl]-malonamide (DMDOHEMA, Fig. 5 left)[60,67] was selected as a new reference molecule, showing improved

[3]The same effect was observed for Cyanex 301, which was later explained by the presence of impurities, see reference[70] [Zhu *et al.* 1986] and references therein.

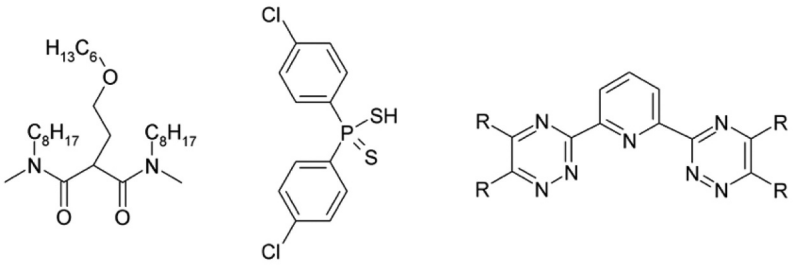

Figure 5. *N,N'*-dimethyl-*N,N'*-dioctyl-2-[2-(hexyloxy)ethyl]-malonamide (DMDOHEMA), bis(4-chlorophenyl)dithiophosphinic acid ($(ClPh)_2PSSH$), and 2,6-bis(5,6-dialkyl-1,2,4-triazin-3-yl)pyridine (BTP).

behavior regarding the criteria mentioned above. Furthermore, improvements in decontamination versus iron and ruthenium were observed.

Three counter-current tests (cold, spiked, and hot) were performed using DMDBTDMA, and a cold test using DMDOHEMA. These tests demonstrated *i. a.* improved performance of DMDOHEMA regarding the unwanted co-extraction of zirconium, molybdenum, ruthenium, and iron.[64,68,69]

An(III)-Ln(III) separation

Significant breakthrough was achieved in the field of An(III)-Ln(III) separation. Two chemical systems were introduced which extract Am(III) and Cm(III) from nitric acid concentrations of >0.5 mol/L, with excellent selectivity over all Ln(III).

Based on work by Zhu *et al.*[70] and Jarvinen *et al.*,[71] systems comprising novel aromatic dithiophosphinic acids were developed.[72,73] A synergistic mixture of bis(4-chlorophenyl)dithiophosphinic acid ($(ClPh)_2PSSH$), Figure 5 center) and TOPO (tri-*n*-octyl phosphine oxide) in *tert.*-butyl benzene proved superior, with separation factors for Am(III) over Ln(III) of $SF_{Am/Ln} \approx 30$ at a nitric acid concentration of 0.5 mol/L. Despite not being *CHON*, this represents a highly promising system for An(III)-Ln(III) separation.

Furthermore, alkylated bis-triazinyl-pyridines (BTP, Fig. 5 right) were investigated. These were the first *CHON* nitrogen-donor extractants to extract Am(III) selectively over Eu(III) ($SF_{Am/Eu} > 100$) without the need for a lipophilic anion source such as 2-bromocarboxylic acid from ≈1 mol/L HNO_3.[30,74] Two hot counter-current tests using *n*-Pr-BTP (Fig. 5 right, R = *n*-C_3H_7) were performed which unfortunately suffered from *n*-Pr-BTP degradation.

PARTNEW: evolution

New Solvent Extraction Processes for Minor Actinides (PARTNEW, 09/2000 – 08/2003)[65] saw the NEWPART consortium strengthened by the

addition of two new partners from Spain, building on the breakthrough advances from NEWPART. The field of An(III)-Ln(III) co-extraction saw diglycolamide extracting agents replace the malonamides. BTP's limited stability manifested during counter-current process tests; second-generation BTPs resolved this stability issue. Finally, processes separating Am(III) and Cm(III) were developed and tested.

An(III)-Ln(III) co-extraction

With DMDOHEMA having been selected as the reference extractant in NEWPART, its radiolytic and hydrolytic study degradation was studied in detail, and solvent clean-up procedures were established.[67] Hot counter-current process test using 16 stages was performed successfully,[69] one of them using a feed solution concentrated by a factor of approximately 10 (high active concentrate, HAR).[75] This volume reduction is advantageous as it reduces the size of the installation, the amount of waste and thus the costs of the process.

PARTNEW saw the well-known *N,N,N',N'*-tetra-*n*-octyl-diglycolamide (TODGA, Fig. 6 left), first studied by Sasaki *et al.*[29] for An(III)-Ln(III) co-extraction, introduced to Europe.[76] It is worth noting that TODGA[77] (together with its branched-chain analogue T2EHDGA[78,79]) has become an ingredient to almost any solvent extraction-based minor actinide separations concept around the world. TODGA has henceforth replaced DMDOHEMA for An(III)-Ln(III) co-extraction. The main reason was TODGA's much better affinity for An(III). While this is not mandatory for An(III)-Ln(III) co-extraction (indeed, DMDOHEMA performed excellently[69,75]), this would later become a prerequisite for the development of processes based on selective An(III) stripping, see below.

TODGA dissolved in kerosene is prone to third phase formation when loaded with Ln(III). Third phase formation can be suppressed by adding modifiers such as TBP, *N,N*-dihexyloctanamide or lipophilic alcohols.[80–82]

Using a solvent comprising 0.2 mol/L TODGA +0.5 mol/L TBP (as phase modifier) in kerosene, two spiked counter-current process tests were performed successfully, proving its process applicability.[83] As with prior tests using DMDOHEMA, oxalic acid and HEDTA were added to the feed solution

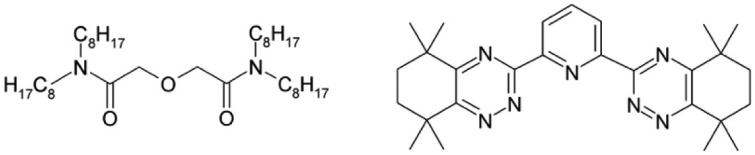

Figure 6. *N,N,N',N'*-tetra-*n*-octyl-diglycolamide (TODGA) and 2,6-bis(5,5,8,8-tetramethyl-5,6,7,8-tetrahydro-benzo-1,2,4-triazin-3-yl)-pyridine ($CyMe_4$-BTP).

to mask zirconium, molybdenum, and palladium. Sixteen stages, as used in the first test, proved to be insufficient with respect to strontium and zirconium rejection and stripping efficiency. This was improved in the second test by increasing the number of stages to 28. Finally, a hot test was performed with 32 stages.[84] Am(III) and Cm(III) losses to the raffinate solution were < 0.01%, and the only product contamination was ≈1% of the ruthenium inventory. All other non-Ln(III) contaminants were < 0.1% of the feed inventory.

Finally, a solvent comprising 0.2 mol/L TODGA +5%$_{vol.}$ (0.32 mol/L) 1-octanol in a kerosene diluent[85,86] was chosen as the baseline *CHON* solvent for An(III)-Ln(III) co-extraction in later EURATOM programs, see below. In contrast to the TODGA-TBP solvent,[80] this solvent is *CHON* compliant, and it extracts less nitric acid compared to other TODGA solvents.[81,82]

An(III)-Ln(III) separation

More in-depth investigations revealed *n*-Pr-BTP' prohibitively low chemical stability.[64,87] Better results were achieved with *i*-Pr-BTP (Fig. 5 right, R = *i*-C_3H_7), which consequently was applied in two counter-current hot tests, the second one with solvent recycling. Excellent separation was achieved, but *i*-Pr-BTP still suffered some degradation.[88] This degradation is initiated by an attack on the alkyl moieties' alpha protons.[89]

Thus, BTPs without such protons were expected to be more stable. Synthesizing a BTP with *tert.*-butyl moieties was unsuccessful. Finally, 2,6-bis(5,5,8,8-tetramethyl-5,6,7,8-tetrahydro-benzo-1,2,4-triazin-3-yl)-pyridine ($CyMe_4$-BTP, Fig. 6 right) was synthesized,[90] showing sufficient stability. Unfortunately, $CyMe_4$-BTP is a too strong extracting agent, resulting in problems stripping An(III).

The synergistic system, $(ClPh)_2PSSH$ + TOPO,[73] was successfully applied in a process test in a 24-stage centrifugal contactor setup.[91] Less than 0.1% of the Am(III) inventory was lost to the raffinate, and only 2% of the Ln(III) inventory was routed to the An(III) product solution. A larger fraction (≈23%) of the Cm(III) inventory was routed to the raffinate, which however is not an issue since Am(III) and Cm(III) require downstream separation anyway.

Am(III)-Cm(III) separation

The final step, Am(III)-Cm(III) separation, had not been addressed in prior programs. Due to somewhat mixed experience, separation based on Am oxidation was not in the scope, other than *e.g.* in the US.[49,50] PARTNEW studied two systems for this task: DMDOHEMA and a synergistic mixture of $(ClPh)_2PSSH$ and tris (2-ethylhexyl) phosphate (T2EHP, Fig. 7).

DMDOHEMA has a small selectivity for Am(III) over Cm(III), $SF_{Am/Cm}$ ≈1.6–1.8.[92] Given a sufficient number of stages, a separation thus is viable. Indeed, a hot test with 56 stages yielded very good results: approximately 0.6%

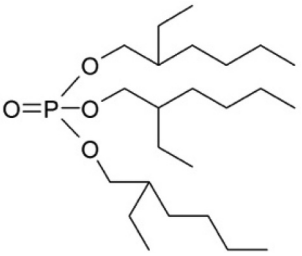

Figure 7. Tris(2-ethylhexyl) phosphate (T2EHP).

of the Am(III) inventory was routed to the Cm(III) raffinate, and approximately 0.7% of the Cm(III) inventory to the Am(III) product.[93]

During the studies on $(ClPh)_2PSSH$, numerous phosphine oxide or phosphate compounds were tested as synergists. By replacing TOPO with T2EHP, an unprecedented Am(III)-Cm(III) selectivity, $SF_{Am/Cm} \approx 8$, was obtained for the synergistic $(ClPh)_2PSSH$ system.[94] This system was later successfully applied in a 24-stage counter-current process test, see the *ACSEPT* section below.

EUROPART: dissipation

The *European research program for the partitioning of minor actinides from high active wastes arising from the reprocessing of spent nuclear fuels* (EUROPART)[66] ran from 01/2004 to 06/2007. It merged the former PARTNEW, CALIXPART,[95] and PYROREP[96] members to a comparatively large consortium of 26 partners, 21 of which were involved in solvent extraction studies. With this, additional countries Czech Republic, Belgium, the Netherlands, and Poland joined. Japan and Australia joined as associated partners without EURATOM funding.

The development related to solvent extraction saw proven compounds such as diglycolamidess, picolinamides, CMPO, etc. bonded to cobaltabisdicarbollide (COSAN) or pre-organized on aromatic or calixarene platforms. To the authors' little surprise, none of almost 100 such pre-organized compounds exhibited improved, let alone useful, properties. The general issue was their extremely strong affinity, as expected for entropic reasons.

On a positive note, the promising BTP compounds evolved to bis-triazinyl-bipyridines. The idea was that such quadridentate extracting agents would form 1:2 complexes rather than the 1:3 complexes formed by BTPs. When the extracting agent degrades due to radiolysis, its decrease in concentration would have a smaller effect on distribution ratios in the case of a 1:2 complex than in the case of a 1:3 complex.

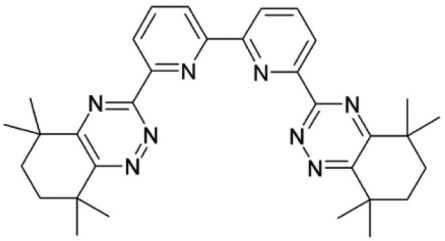

Figure 8. 6,6′-bis(5,5,8,8-tetramethyl-5,6,7,8-tetrahydro-benzo-1,2,4-triazin-3-yl)-2,2′-bipyridine ($CyMe_4$-BTBP).

This concept together with the stability improvement experienced with $CyMe_4$-BTP led to the synthesis of 6,6'-bis(5,5,8,8-tetramethyl-5,6,7,8-tetrahydro-benzo-1,2,4-triazin-3-yl)-2,2'-bipyridine ($CyMe_4$-BTBP, Fig. 8).[97] Fortunately, $CyMe_4$-BTBP exhibits usable selectivity and extraction strength, extracting Am(III) and Cm(III) from approximately 1 mol/L HNO_3 selectively over Ln(III). Stripping is feasible using e.g. glycolate solutions. Its slow kinetics could be overcome by adding DMDOHEMA[98] or TODGA[99] as phase transfer catalyst. Indeed, a highly successful hot flowsheet test was performed in a 16-stage centrifugal contactor rig.[100] Am(III) and Cm(III) losses to the raffinate were 0.01% and 0.09%, respectively, and as little as 0.01% of the Ln(III) inventory was routed to the An(III) product solution.

$CyMe_4$-BTPB has been the European reference extracting agent for An(III)-Ln(III) separations for many years. The journey from the unstable first-generation BTPs via $CyMe_4$-BTP to $CyMe_4$-BTBP took approximately 7 years. Several reviews recapitulate this development.[40–42]

Phase 2, 2008–2021

The following programs, ACSEPT,[101] SACSESS,[102] and GENIORS,[103] were coordinated by Stéphane Bourg from the CEA. With the most promising chemical systems established in past programs, the focus shifted to consolidating process options, stability and safety studies, and fuel cycle integration.

ACSEPT: refocus

The *Actinide Recycling by Separation and Transmutation* (ACSEPT, 03/2008 – 09/2012)[101] program covered hydrometallurgical and pyrometallurgical separations, involving 34 teams from 15 countries (now including Portugal and Switzerland), approximately 22 of them involved in hydrometallurgy. Approximately 140 new compounds were synthesized and screened in the *hydrometallurgy* domain. The experience gained during this and earlier programs allowed narrowing down to a portfolio of three proven molecule cores,

TODGA, BTP, and BTBP. An(III)-Ln(III) co-extraction was no longer in the scope, and work focused on An(III)-Ln(III) separation and on *Grouped Actinide Extraction* (GANEX), the latter supporting homogeneous recycling strategies.[104]

An(III)-Ln(III) separation

Three concepts for separating An(III) from Ln(III) were studied: (1) regular An(III)-Ln(III) separation using $CyMe_4$-BTBP, (2) direct extraction of An(III) from PUREX raffinate, skipping the upstream An(III)-Ln(III) co-extraction process, and (3) selective An(III) stripping using hydrophilic ligands, based on the TALSPEAK concept.

(1) Since DMDOHEMA was dropped in favor of TODGA *(cf.* PARTNEW), the latter was also used as a phase transfer catalyst to improve the slow kinetics of $CyMe_4$-BTBP. A spiked 20-stage flowsheet test was performed using a solvent of 15 mmol/L $CyMe_4$-BTBP and 5 mmol/L TODGA in 1-octanol. Am(III) and Cm(III) losses to the raffinate were 0.08% and 0.3%, respectively, and approximately 0.1% of the Ln(III) inventory was routed to the An(III) product solution.[99]

(2) An(III) are directly extracted from PUREX raffinate into a solvent of 15 mmol/L $CyMe_4$-BTBP and 5 mmol/L TODGA in kerosene/1-octanol. Zr and Mo are masked with oxalic acid, Pd extraction is suppressed by adding L-cysteine.[105] A spiked counter-current test of a 32-stage flowsheet[106] was successfully run using centrifugal contactors.[107] The product solution contained ≥ 99.8%, ≥99.4%, and 0.8% of the respective Am(III), Cm(III), and Pd(II) inventories, demonstrating the system's potential.

(3) The CEA developed a process co-extracting An(III) and Ln(III) into a TODGA + TBP solvent, followed by selective An(III) stripping using DTPA (diethylenetriaminepentaacetic acid) in a buffered nitrate solution (nitrate is required to keep Ln(III) in the solvent at low acidity). A successful hot test was performed.[108] Alternatively, a simplified system was developed: An(III) and Ln(III) are co-extracted from a PUREX raffinate into the baseline TODGA solvent (0.2 mol/L TODGA +5%$_{vol.}$ 1-octanol in kerosene), and An(III) are selectively stripped from the loaded solvent using a water-soluble stripping agent, 2,6-bis(5,6-di(3-sulphophenyl)-1,2,4-triazin-3-yl)-pyridine tetrasodium salt (SO_3-Ph-BTP, Figure 9). Ln(III) are kept in the solvent by a sufficiently high nitric acid concentration in the stripping solution, avoiding the addition of nitrate salt.[109] Excellent selectivity was obtained, $SF_{Eu/Am} \approx 800$, $SF_{La/Am} \approx 50$.

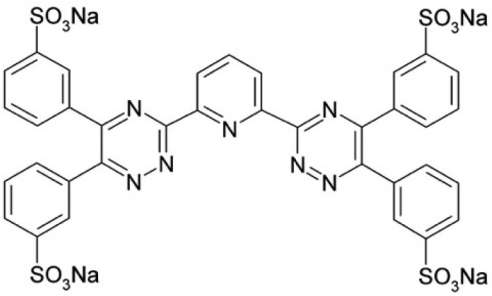

Figure 9. 2,6-bis(5,6-di(3-sulphophenyl)-1,2,4-triazin-3-yl)-pyridine tetrasodium salt (SO_3-Ph-BTP).

Concepts (2) and (3) represent a significant simplification of the overall scheme to sequester Am(III) by skipping the An(III)-Ln(III) co-extraction process.[56] Particularly the TODGA/SO_3-Ph-BTP system[109] was a major step forward: In contrast to stripping agents such as DTPA, SO_3-Ph-BTP allows stripping An(III) at nitric acid concentrations sufficient to keep Ln(III) in the solvent. TALSPEAK-based processes require a pH buffer and adding a nitrate salt or an acidic extracting agent to keep Ln(III) in the solvent under low-acidity stripping conditions. Although the introduction of a non-*CHON* stripping agent is disadvantageous, the TODGA/SO_3-Ph-BTP system is simple and efficient.

Am(III)-Cm(III) separation

Although initially intended to separate Am(III) from Cm(III), the $(ClPh)_2$ PSSH + TEHP system developed in PARTNEW indeed is capable of separating Am(III) from both Cm(III) and Ln(III).[94] Thus, it can directly process the product solution from an An(III)-Ln(III) co-extraction process, skipping An(III)-Ln(III) separation.

The $(ClPh)_2$PSSH + TEHP system was applied in a 24-stage counter-current process test. An Am(III) product containing < 5% of the Cm(III) inventory was obtained, and <0.1% Am(III) was lost to the raffinate.[110]

Homogeneous recycling – the GANEX processes

The *Grouped Actinide Extraction* (GANEX) concept[111,112] was developed by the CEA within their national R&D program to address homogeneous recycling strategies[104] and was later introduced to the European programs. The GANEX first cycle extracts uranium from the fuel dissolution solution, and the second cycle co-separates the group of transuranium (TRU) elements, neptunium, plutonium, americium, and curium. Three alternative GANEX 2^{nd} cycle processes were developed: one (CHALMEX) based on selective TRU extraction and two (CEA-GANEX and EURO-GANEX) based on selective TRU stripping.[113]

GANEX 1st cycle, U extraction: In a first step, uranium is extracted from the fuel dissolution solution. This step yields a pure uranium product, which is required to adjust the U/TRU ratio in the final fuel. Uranium, present as UO_2^{2+}, is extracted using *N,N*-di-(ethyl-2-hexyl)isobutyramide (DEHiBA) dissolved in kerosene. TRU and fission and corrosion products are routed to the raffinate, to be treated in the GANEX 2nd cycle. Flowsheets were designed, and two successful hot tests were performed: the first at the CEA (within their national R&D program) using irradiated UOx fuel[114]; the second one (as part of ACSEPT) using $(U_{80}Pu_{20})Ox$ fast reactor fuel.[115]

GANEX 2nd cycle development at the CEA:[116] Based on CEA's experience with the DIAMEX-SANEX concept,[93] a system was developed that can additionally handle Pu and Np. TRU, Ln(III) and some fission and corrosion products (Zr, Mo, Tc, Fe) are co-extracted into a solvent of DMDOHEMA and di-(2-ethyl-hexyl)phosphoric acid (D2EHPA) in kerosene, followed by (1) Mo, Tc stripping, (2) TRU stripping using hydroxyethylethylenediaminetriacetic acid (HEDTA), citric acid and hydroxyurea (the latter to reduce Np to the unextractable (V) state), and (3) Ln(III), Zr, Fe stripping. A 24-stage flowsheet was hot-tested with very good results, using the 1st cycle[114] raffinate.

EURO-GANEX 2nd cycle: aiming at simplifying above flowsheet, the EURO-GANEX system was developed in ACSEPT, based on the SO_3-Ph-BTP system.[109] TODGA is used to co-extract TRU(III,IV,VI) and Ln(III). To manage a Pu(IV) feed concentration of at least 10 g/L, the solvent had to be optimized. A sufficient Pu(IV) loading capacity is achieved with a solvent containing 0.2 mol/L TODGA and 0.5 mol/L DMDOHEMA in kerosene.[117,118] TRU are stripped into a solution of SO_3-Ph-BTP (to strip Am and Cm) and acetohydroxamic acid (AHA, to strip Np and Pu)[119] in nitric acid (to keep Ln(III) in the solvent). The system's development is described in detail in reference.[120] A 32-stage flowsheet was calculated and a "Pu-active" test (with a synthetic feed solution containing 10 g/L Pu) performed.[121] Based on the experience from this test, a consolidated flowsheet was designed and hot-tested (including the GANEX 1st cycle) on a feed solution containing ≈100 g/L U and ≈23 g/L Pu (1st cycle feed) and ≈10 g/L Pu (2nd cycle feed).[115] Total TRU losses across the two cycles were as low as 3.3% Np, 0.26% (Pu), and 0.11% (Am). Shortcomings are the use of a solvent containing two extracting agents and of a non-*CHON* stripping agent, SO_3-Ph-BTP.

CHALMEX 2nd cycle: Obviously, an even simpler concept would be to extract only TRU from the 1st cycle raffinate, rejecting all other elements. $CyMe_4$-BTBP has already been applied in a flowsheet directly extracting Am(III) from a PUREX raffinate simulate,[105–107] and consequently was selected for the CHALMEX process.[122] TBP was added to manage Pu extraction.[123] To solve the solubility and kinetics issues associated with

$CyMe_4$-BTBP, various diluents were assessed, and finally, phenyltrifluoromethylsulfone (FS-13)[124] was selected. Co-extraction of some fission and corrosion products by $CyMe_4$-BTBP is an issue; Mo, Zr and Pd are masked by adding bimet[125] and D-mannitol to the feed solution. Improvements toward making the CHALMEX process *CHON* are underway, *e.g.* by replacing TBP with a monoamide extractant.[126] To date, the CHALMEX system has not been flowsheet-tested.

SACSESS: safety first

In December 2011, Japanese colleagues held an excellent *GLOBAL* conference in Chiba, Japan. During that event, which was heavily shadowed by the Fukushima Dai-ichi accident, the coordinator of the Phase 2 programs and one of this paper's authors sat together and drafted a follow-up program to ACSEPT. Safety had to be an important keyword, and *Safety of Actinide Separation Processes* (SACSESS, 03/2013 – 02/2016)[102] was the name. The focus accordingly shifted toward maloperations and stability studies, and a detailed safety analysis of the EURO-GANEX process developed in ACSEPT was executed. Under a collaboration agreement with the US Department of Energy, joint experiments with colleagues at Idaho National Laboratory were performed, and joint workshops on kinetics and on americium chemistry were organized. During these days, there also was an informal nevertheless intensive exchange with the US *Sigma Team for Advanced Actinides Recycle* (STAAR, see reference[127]) led by Bruce Moyer, predominantly on Am(III)-Cm(III) separation using nitrogen-donor extracting agents. SACSESS covered both pyrometallurgical and hydrometallurgical separations, with 19 out of 26 partners involved in hydrometallurgy.

An(III)-Ln(III) separation

A spiked flowsheet test using the TODGA/SO_3-Ph-BTP system[109] developed in ACSEPT was run in a centrifugal contactor rig. With a total of 32 stages, ≥99.8% An(III) were recovered together with small amounts of impurities, 0.4% Ru, 0.3% Sr, and 0.1% Ln(III).[128] Despite its drawback of not being *CHON*, the potential of a simple system was successfully demonstrated: one extracting agent plus modifier, one stripping complexant, nitric acid to keep Ln(III) in the solvent. *Trans*-1,2-diaminocyclohexane-*N,N,N',N'*-tetraacetic acid (CDTA)[129] must be added to the feed and scrub solutions to mask Zr and Pd.

Consequently, efforts were made finding a *CHON* alternative to SO_3-Ph-BTP. Earlier attempts hydrophilising BT(B)P showed unfavorable results.[130] A breakthrough achievement was the development of 2,6-bis[1-(3-hydroxypropyl)-1,2,3-triazol-4-yl]pyridine[131] (PTD, Fig. 10). This *CHON* compound selectively strips An(III) from the baseline TODGA solvent (0.2

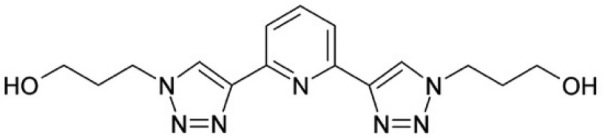

Figure 10. 2,6-bis[1-(3-hydroxypropyl)-1,2,3-triazol-4-yl]pyridine (PTD).

mol/L TODGA +5%$_{vol.}$ 1-octanol in kerosene) loaded with An(III) and Ln(III) at nitric acid concentrations of approximately 0.5 mol/L. This system was later tested in a centrifugal contactor rig, see the GENIORS section below.

Am(III)-Cm(III) separation

Extracting Am(III) directly from the PUREX raffinate drastically simplifies the overall separation scheme. To this, CEA developed the *Extraction of Americium* (EXAm) process[55] as a part of their national R&D program and thus outside of SACESSS. Combining DMDOHEMA's preference for lighter *f*-element ions with the preference of *N,N,N',N'*-tetraethyldiglycolamide (TEDGA)[132,133] for heavy ones yields a selectivity for Am(III) over Cm(III) of $SF_{Am/Cm}$ ≈2.5, allowing for preferential extraction of Am(III). Am(III) is then separated from the light Ln(III) (which are co-extracted) by stripping into a HEDTA/citrate or DTPA/malonate solution, *i.e.* TALSPEAK chemistry. To keep the light Ln(III) in the organic phase during Am(III) stripping, the solvent also contains D2EHPA. A hot test was run using 68 stages of laboratory-scale mixer-settlers (with 32 stages devoted to Am(III)-Cm(III) separation). >98% of Am(III) was recovered with decontamination factors of 505 and 340 over Cm(III) and Nd(III), respectively.[55]

Goal within SACSESS was simplifying the EXAm process: TODGA was to be used as extracting agent (to co-extract An(III) and Ln(III) while rejecting other fission products and corrosion products), and stripping agents were to be found which would (1) strip Am(III) selectively over Cm(III) and Ln(III) and (2) remain active in nitric acid having a concentration sufficiently high to keep Ln(III) in the solvent during stripping.

EURO-EXAm: Combining the baseline TOGDA solvent with *N,N,N',N'*-tetrakis[(6-carboxypyridin-2-yl)methyl]ethylenediamine (TPAEN, Fig. 11 left) as Am(III) stripping agent gave improved selectivity ($SF_{Cm/Am} \leq 4$) in nitric acid concentrations up to 0.2 mol/L.[134] Unfortunately, irreproducible precipitations during centrifugal contactor tests led to the decision to focus on alternative stripping agents.

AmSel: As an alternative, the *Americium Selective Extraction* (AmSel) system[135] was developed, replacing TPAEN with 6,6'-bis(5,6-di(3-sulphophenyl)-1,2,4-triazin-3-yl)-2,2'-bipyridine tetrasodium salt (SO_3-Ph-BTBP, Fig. 11 right). With $SF_{Cm/Am}$ ≈2.5, the selectivity was comparable to that of the original EXAm process, however at much higher nitric acid

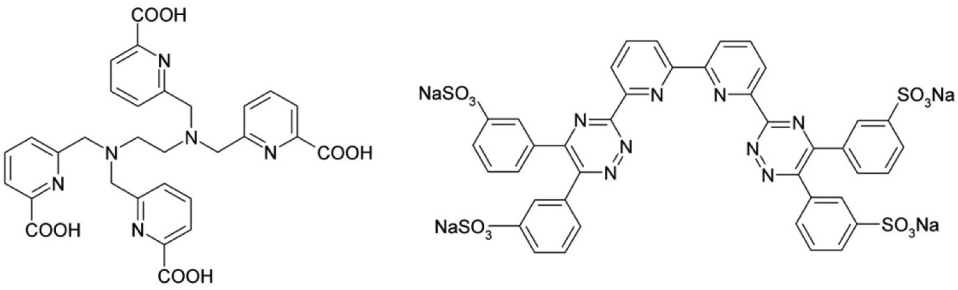

Figure 11. *N,N,N′,N′*-tetrakis[(6-carboxypyridin-2-yl)methyl]ethylenediamine (TPAEN) and 6,6′-bis (5,6-di(3-sulphophenyl)-1,2,4-triazin-3-yl)-2,2′-bipyridine tetrasodium salt (SO_3-Ph-BTBP).

concentrations of ≈0.7 mol/L. Despite AmSel not being *CHON*, the system shows some promise. SO_3-BTPhen,[136] the phenanthroline analogue of SO_3-Ph-BTBP, shows slightly improved selectivity, $SF_{Cm/Am}$ ≈3, in combination with the baseline TODGA solvent.

We would like to point out that the state of Am(III)-Cm(III) separation was reviewed in 2021 by our colleagues from Lomonosov Moscow State University, Moscow, and Bhabha Atomic Research Centre, Mumbai.[137] A more recent review is also available.[138]

Stability studies

Extracting agents to be applied in advanced actinide recycling schemes are in prolonged contact with nitric acid and are exposed to significant radiation doses and hence must be sufficiently robust.

Extending earlier work,[139] both the baseline TODGA solvent in contact with nitric acid and the TODGA/SO_3-Ph-BTP system[109] were gamma-irradiated under realistic conditions in the radiolysis test loop at Idaho National Laboratory.[140] An(III) and Ln(III) distribution ratios remained essentially unchanged to maximum absorbed doses of 0.9 MGy (TODGA/HNO_3) and 174 kGy (TODGA/SO_3-Ph-BTP).

EURO-GANEX safety analysis

The EURO-GANEX 2nd cycle was used as a reference process to carry out a detailed safety analysis. In a first step, a safety assessment methodology was defined. This methodology covered both normal operations and mal-operations. One specific maloperation, reducing the scrub acidity from the normal concentration of 0.5 mol/L to 0.05 mol/L, was selected to observe its effect on Pu and Am extraction.[141] The malop flowsheet was tested in a centrifugal contactor rig (10 extraction stages, 6 scrub stages) with a feed containing 10 g/L Pu. With normal operations steady state attained, the scrub acidity was reduced. Surprisingly, this dramatic process upset did not result in the backwashing and recycle of plutonium in the extract-scrub

contactors. A constant plutonium loading of the solvent was maintained under both normal and maloperation conditions, demonstrating the process's robustness. The EURO-GANEX safety analysis was carried forward to the GENIORS program.

GENIORS: integration

In continuity of SACSESS, *GEN IV Integrated Oxide Fuels Recycling Strategies* (GENIORS, 06/2017 – 05/2021) addressed the reprocessing and manufacture of MOX fuel potentially containing minor actinides.[103] With the most promising solvent extraction systems developed and mostly tested, the focus shifted toward system and safety studies and integrating the separations processes into the fuel cycle. Process development addressed flowsheet-testing PTD as a *CHON* alternative to SO_3-Ph-BTP and simplifying the EURO-GANEX process. Twenty of 24 partners were involved in topics related to hydrometallurgy, from dissolution to separation to conversion. Unfortunately, the last year of GENIORS was essentially thwarted due to COVID restrictions.

An(III)-Ln(III) separation

To evaluate the applicability of PTD[131] (Fig. 10) as a *CHON* alternative to SO_3-Ph-BTP (Fig. 9), a flow-sheet was developed[142] and tested using a spiked feed solution.[143] The baseline TODGA solvent (0.2 mol/L TODGA +5%$_{vol.}$ 1-octanol in kerosene) was loaded with An(III) and Ln(III) in batch contacts. The An(III) stripping and the Ln(III) re-extraction sections were run in a 16-stage centrifugal contactor setup. The product solution contained ≥ 99.9% of the An(III) inventory and ≤0.1%, 0.25%, and 1.7% of the respective Ln(III), Zr(IV), and Ru(III) inventories, relative to the feed solution. Later, a full 32-stage flow-sheet trial with an Am(III) feed concentration of approximately 4 mmol/L was run, including spectroscopic on-line monitoring.[144] This test corroborated the results from the prior test. These excellent results qualify PTD as a viable *CHON* replacement for SO_3-Ph-BTP.

EURO-GANEX simplification

A major drawback of the EURO-GANEX 2nd cycle chemistry[120] as developed in ACSEPT is the solvent containing two extracting agents. This was resolved by replacing TODGA with *R,S*-dimethyl-*N,N,N',N'*-tetra-*n*-decyl-diglycolamide (mTDDGA, Fig. 12), no longer requiring the addition of DMDOHEMA to achieve sufficient plutonium loading capacity.[145] Besides improving the loading capacity, decontamination from fission products was also improved. Furthermore, to make this process *CHON*, SO_3-Ph-BTP (Fig. 9) was dropped in favor of PTD (Fig. 10). The mTDDGA/PTD system was never developed up to a level making it applicable to a flowsheet trial.

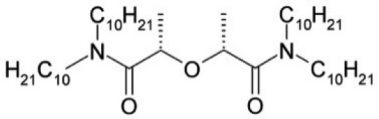

Figure 12. *R,S*-dimethyl-*N,N,N′,N′*-tetra-*n*-decyl-diglycolamide (mTDDGA).

Nevertheless, its radiolytic resistance was thoroughly evaluated under static[146] and dynamic conditions, the latter in an inter-institutional study in collaboration with Idaho National Laboratory.[147]

Further topics

Numerous further topics (*cf.* www.geniors.eu/public-library) addressing process safety and integration were covered, among these: The baseline TODGA and the EURO-GANEX 2nd cycle solvents were irradiated to quantify the rate of hydrogen generation. G_{H2} values of 3.51 and 3.41 molecules/100 eV respectively were determined.[148] The clean-up and recycling of degraded TODGA solvents was studied and purification strategies developed.[149] A sophisticated equilibrium model to calculate nitric acid extraction into the TODGA solvent modified with 1-octanol was established.[150] A concept design for a EURO-GANEX plant was made and safety-reviewed. Using a "sim-plant" engineering simulation tool, the concept EURO-GANEX flowsheet was shown to perform well compared to PUREX and Advanced PUREX. This exercise also outlined key areas for improvement to the flowsheet.[151]

Certainly, the second *Radical Behaviour* workshop held in Würzburg, Germany, was a highlight. Approximately 40 scientists were discussing radiation chemistry related to the field of nuclear fuel cycle solvent extraction for 2 days.

Phase 3, 2020–2028

The Belgian Nuclear Research Centre (SCK CEN) coordinated PATRICIA[152] and currently coordinates its follow-up, TRANSPARANT.[153] These programs focus on the separation of americium from used fuel, on experimental and fuel performance code development related to americium-bearing fuel under irradiation, and on safety related research supporting the licensing process of MYRRHA[154] as a dedicated accelerator driven transmuter demonstrator.

Combining "P&T" into one program was a major departure from the previous projects, which were fully dedicated to separations. In consequence, the budget available for separations studies was drastically reduced, and only nine core partners (from France, the UK, Sweden, Spain, Italy, Belgium, and Germany) from the previous programs could be maintained in the *separations*

domain of PATRICIA and TRANSPARANT. Work thus had to focus on a single process. Since Am(III)/Cm(III) separation was the least mature, it was selected for further study, continuing where one has left off with the EXAm[55,155] and AmSel[135] processes. The goal is to design a robust process yielding a solid precursor material suitable for americium target fabrication.

PATRICIA: Am(III) only

The *Partitioning and Transmuter Research Initiative in a Collaborative Innovation Action's* (PATRICIA,[152,156] 09/2020 – 08/2025) *separations* domain developed and tested processes to extract Am(III) from a PUREX raffinate solution and convert it to a solid precursor material suitable for target of fuel fabrication.

The TODGA/SO_3-Ph-BTBP AmSel system[135] developed in earlier programs was selected as a reference system to be used in a counter-current lab scale flowsheet test and to be studied in more detail. To this, numerous thermodynamic and kinetic data were collected. The system's behavior under irradiation was also studied in detail.[157,158] Finally, a flowsheet was calculated and tested in a centrifugal contactor setup. Only the Am(III) stripping/[Cm(III) + Ln(III)] re-extraction was run using 16 stages; the respective extraction/scrubbing having been demonstrated successfully earlier.[128] Am(III) was efficiently separated from Ln(III). However, significant build-up of Am(III) between the Am(III) stripping section and the Cm(III) + Ln(III) re-extraction section occurred, and only 45% of the Am(III) inventory, together with 5% Cm(III) was routed to the product solution.[159] Obviously, the flowsheet requires further optimization, including increasing the number of stages.

Further goal was finding a *CHON* AmSel system. Inspired by an earlier study,[160] the central pyridine in PTD[131] was replaced by bipyridine to make 6,6'-bis[1-(3-hydroxypropyl)-1,2,3-triazol-4-yl]pyridine-2,2'-bipyridine (PrOH-BPTD, Fig. 13).[161] As expected, PrOH-BPTD combined with the baseline TODGA solvent (0.2 mol/L TODGA +5%$_{vol.}$ 1-octanol in kerosene)

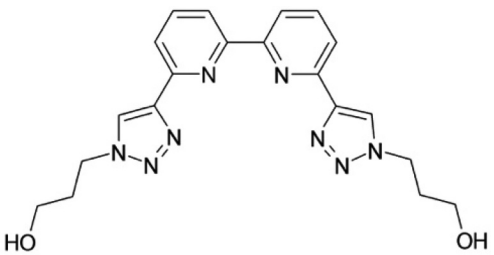

Figure 13. 6,6'-bis[1-(3-hydroxypropyl)-1,2,3-triazol-4-yl]pyridine-2,2'-bipyridine (PrOH-BPTD).

separates Am(III) from Cm(III) and Ln(III), albeit with a moderate selectivity of $SF_{Cm/Am} \approx 2$. Alternatively, molecules with a phenanthroline moiety replacing the central bipyridine unit were synthesized and screened, with comparable results.

TRANSPARANT: selectivity

Technological Research Action Necessary for Safe Partitioning and Nuclear Transmutation (TRANSPARANT,[153] 09/2024 – 11/2028) is the direct follow-up program to PATRICIA. Its *separations* domain maintains both the consortium (with the UK partners no longer receiving EURATOM funding due to Brexit) and the scope, with coordinating tasks being handed over to the next generation of scientists.

The goal is to further develop and optimize the TODGA/PrOH-BPTD baseline system[161] developed in PATRICIA. A major objective is to improve the relatively low selectivity, $SF_{Cm/Am} \approx 2$, while maintaining sufficient selectivity between Am(III) and the light Ln(III). To this end, slight structural modifications are being made to both the extracting agent (TODGA)[162,163] and the stripping agent (PrOH-BPTD), and their effect on selectivity assessed. Furthermore, a flowsheet will be designed to finally apply an optimized system in a counter-current process demonstration.

Conclusions

Recycling uranium and plutonium allows for a more efficient utilization of the resource *natural uranium*. Additionally recycling americium can result in a more efficient utilization of the resource *final repository volume.*

To this, concepts for separating americium from PUREX raffinate solutions have been developed within European collaborative projects for more than three decades. Hundreds of novel extracting agents have been synthesized and screened. Schemes each consisting of three, two, or one post-PUREX solvent extraction cycles[56] to obtain a pure americium product have been designed and demonstrated on the lab scale up to a "technical readiness level" of TRL = 5.[48] Additionally, GANEX processes addressing homogeneous recycle have been developed and demonstrated.[113] This way, substantial progress has been achieved in the field of actinide separations.

Obviously, a great deal of liquid–liquid distribution data has been collected during the execution of these projects, much of which have been entered into *The International Database for Extractant Ligands* (IDEaL).[164] Addressing some shortcomings of the IDEaL database, the *Separation Archive for Elements* (SAFE) database[165] was created under an effort led by Los Alamos National Laboratory, USA. Continued population and curation of SAFE will be essential

to maximize its long-term value for the separation science community. Please help improving this database!

The successful execution of the research discussed in this paper hinged on three factors: (1) continuity, which was maintained through steady financial support from the European Atomic Energy Community (EURATOM); (2) highly motivated teams from the contributing laboratories across Europe, and (3) a "collaboration rather than competition" mindset. Countless discussions (not: on-line meetings) both within the projects and with many excellent scientists from around the globe helped promoting the science. However, numerous ideas for improvement have been discussed, but never pursued due to a lack of funding.

In conclusion, these EURATOM programs have created a close-knit European actinide separations community, which, in all modesty, has been at the forefront of minor actinide separations. Starting out with a concept based on three post-PUREX processes to obtain an americium product, this number has been reduced to one process. Furthermore, a suite of GANEX processes have been developed, addressing homogeneous recycling. A portfolio of novel extracting and complexing agents is now at hand, providing a toolbox to develop even better processes.

This success is expected to continue with the next generation of young scientists at the helm. However, we must emphasize that this success is at risk with any further reduction in funding.

Acknowledgments

This paper is based on an oral presentation given at the 23rd International Solvent Extraction Conference (ISEC 2025), Melbourne, Australia, 29 September – 2 October 2025.

Our thanks go to a very long list of people. Our *special* thanks go to our dear colleagues and friends we have had the chance of working, discussing, and having an occasional beer with: Stéphane Bourg, Melissa Denecke, Christian Ekberg, Amparo Espartero, Hitos Galán, Bruce Hanson, Clément Hill, Rikard Malmbeck, Bruce Mincher, Bruce Moyer, *Ken Nash*, Petra Panak, Robin Taylor, Terry Todd. Finally, a *thank you* to the Guest Editors of this Special Issue, Mark Jensen and Peter Zalupski.

Author contributions

CRediT: **Andreas Geist:** Conceptualization, Writing – original draft, Writing – review & editing; **Giuseppe Modolo:** Writing – original draft, Writing – review & editing.

Disclosure statement

No potential conflict of interest was reported by the author(s).

Funding

We are grateful for the continuous funding through EURATOM since the 1990's. The work discussed in this paper received EURATOM funding via the following contracts: Partition of high-level radioactive waste, contract FI2W-CT90-0047; High-level liquid waste partitioning by means of completely incinerable extractants, contract FI2W-CT91-0112; NEWPART, contract FI41-CT-96-0010; PARTNEW, contract FIKW-CT200-00087; EUROPART, contract FI6W-CT-2003-508854; ACSEPT, contract FP7-CP-2007-211267; SACSESS, grant agreement No. 323282; GENIORS, grant agreement No. [755171]; PATRICIA, grant agreement No. [945077]; TRANSPARANT, grant agreement No. [101166386.

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