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Report on the comparison of SX processes for heterogeneous recycling

Authors : Mr. Andreas GEIST (KIT), Andreas GEIST (KIT)

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Summary

Deliverable D8.2, Report on the comparison of SX processes for heterogeneous recycling, discusses and compares some of the more advanced separation concepts for heterogeneous recycling developed in Europe. These are processes separating trivalent minor actinides (with a focus on americium) from the raffinate solution originating from processes such as PUREX, where U and Pu are managed separately, or an evolution of PUREX, allowing a co-recovery and a co-management of U and Pu. Six process schemes are presented, each consisting of one, two or three solvent extraction cycles. The choices for the various solvent extraction processes are briefly introduced. Next, the most suitable choices are selected, and then, the six process schemes are compared to one another. It becomes evident that not necessarily the schemes appearing the simplest at a first look are actually the most viable ones. Finally, some further directions for future R&D are proposed.

Approval

Date	By
2018-07-13 10:53:07	Mrs. Eva BOO (LGI)
2018-07-13 11:51:47	Mr. Rhodes CHRIS (NNL)
2018-10-25 20:31:27	Mr. Stéphane BOURG (CEA)

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EXECUTIVE SUMMARY

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Six process schemes are presented, each consisting of one, two or three solvent extraction cycles. The choices for the various solvent extraction processes are briefly introduced. Next, the most suitable choices are selected, and then, the six process schemes are compared to one another. It becomes evident that not necessarily the schemes appearing the simplest at a first look are actually the most viable ones.

Finally, some further directions for future R&D are proposed.

WHY RECYCLE AT ALL?

Spent nuclear fuel from reactors must be managed, firstly by being stored to reduce the radioactivity and heat generated and then treated either as a waste by disposal in a geological disposal facility (GDF) or as a resource by recycling useful components.

A question to begin with — *why recycle at all?* The three major drivers for recycling irradiated nuclear fuel from commercial reactors are:[1-3]

- Improving utilisation of resources
- Reducing long-term radiotoxicity of the final waste
- Reducing long-term heat load of the final waste

These drivers are briefly discussed below.

IMPROVING UTILISATION OF RESOURCES

In the context of nuclear energy, the resource instantly coming to mind is natural uranium. With light water reactors and a once-through fuel cycle (i.e. considering irradiated fuel a waste), natural uranium utilisation is less than 0.75% — with 99% going to waste. A small but significant increase to approximately 1% is achieved by reprocessing spent light water reactor fuel and recycling the separated plutonium and the reprocessed uranium once as MOX fuel. A further dramatic increase to > 80% is achievable by deploying multiple recycling in fast reactors. In France, for example, the stockpile of depleted uranium (approximately 300000 tons by 2025) would render further uranium mining unnecessary for centuries if multi-recycling was pursued.[4]

REDUCING LONG-TERM RADIOTOXICITY

The long-term radiotoxicity of irradiated nuclear fuel is governed by plutonium and americium. After a few hundred years, the contribution from other radionuclides is typically below that of the natural uranium required to produce nuclear fuel. Reducing the long-term radiotoxicity of the high-level waste (HLW) to be finally disposed of has been a major driver for pursuing the “Partitioning & Transmutation” strategy in the past. Simplified, the idea was to return to the geosphere less radiotoxicity than was initially removed from it.

This reasoning has never been accepted by the geological disposal community.[1, 5] Simply put, it is argued that not what is in a repository is of concern but what could find its way into the biosphere and what additional dose this would cause. Therefore, it is concluded that recycling actinides does not affect the repository safety case (except in a human intrusion scenario). It is also criticised that the liberal use of radiotoxicity-vs.-time diagrams conveyed the message that P&T would reduce the period of time HLW has to be safely stored (a statement which has indeed been made — or implied — in several P&T publications). In conclusion, the radiotoxicity aspect probably has more to do with public acceptability of nuclear energy than with technical benefits.

REDUCING LONG-TERM HEAT LOAD

Similar to its radiotoxicity, the thermal power of irradiated nuclear fuel is dominated by fission products for approximately the first century after its unloading from a reactor. Beyond that period of time, plutonium and to a lesser extent, americium are the main contributors. Thus, by removing plutonium (as done in the PUREX processes) and americium and allowing for prolonged interim storage, the remaining vitrified waste has less thermal power. This in turn allows for a denser packing of the waste containers, and a smaller repository for a given amount of HLW. Considering the cost of a repository approximately proportional to the excavated volume, this reduces the final repository's share to fuel cycle cost.

Furthermore, by reducing the volume and/or number of deep geological repositories required to store HLW, this driver also contributes to the first bullet point, “Improving utilisation of resources” — considering the space required for a repository a (scarce) resource. This may be a critical point for countries planning a long-term deployment of nuclear energy.

HOW TO RECYCLE?

Recycling of a certain value usually is not possible without prior treatment. This also holds for recycling actinides; before their re-use as reactor fuel they must be separated from fission products and, depending on the recycling strategy, from one another.

Taking full advantage of the benefits mentioned above requires multiple recycling of the transuranium elements (TRU: neptunium, plutonium, americium and curium), most notably plutonium and americium, in reactors with a fast neutron spectrum.[6] The foundations for the underlying liquid metal-cooled fast reactor technology had been laid in the 1940's and demonstrated with the Clementine reactor and later the Experimental Breeder Reactor 1 at Argonne-West, now Idaho National Laboratory.

HOMOGENEOUS AND HETEROGENEOUS RECYCLING STRATEGIES

Homogeneous recycling describes a fuel cycle in which TRU are contained together in the fuel. *Heterogeneous recycling,* in contrast, relies on MOX driver fuel (containing uranium, plutonium and optionally, neptunium) and dedicated targets containing americium (or potentially curium).

The reactors and fuels aspects related to heterogeneous and homogeneous recycling are compiled and compared against one another in a publication by the OECD-NEA.[6] As far as separation processes are concerned, heterogeneous recycling requires generating separate product streams containing uranium, plutonium and neptunium on one hand and americium (or americium and curium) on the other hand. While such separations are suitable for homogeneous recycling, too, dedicated separation processes addressing homogeneous recycling are being developed nevertheless. Such processes typically separate uranium in a first step, followed by a second step co-separating the TRU.

AQUEOUS AND NON-AQUEOUS SEPARATION PROCESSES

Of all the unit operations applicable to separating actinides from irradiated nuclear fuel, only two have successfully been studied and demonstrated:

On one hand, **solvent extraction** has successfully been applied for separating uranium and plutonium: the PUREX process is applied at the industrial scale, see the reprocessing plants at La Hague (France) and at Sellafield (UK) which are in commercial operation. Solvent extraction processes are often referred to as **hydrometallurgical** or **aqueous separations.** *Principle:* An ionic solute in an aqueous phase forms a complex with a complexing agent present in an immiscible organic phase. This complex is preferentially soluble in the organic phase; the solute is being extracted. This way, the solute is separated from other solutes not forming complexes with the complexing agent. The solute is then back extracted to an aqueous phase having a composition different from the initial aqueous phase. This way, the organic phase can be recycled to the extraction step.

On the other hand, **non-aqueous (pyrometallurgical) processes** for separating actinides are under development. These processes are based on electrochemical methods, such as electrolysis or electrorefining, or on reductive extraction in a high temperature molten salt solvent.[7-8] Since only aqueous processes are studied in the framework of GENIORS, non-aqueous processes are not the subject of this report.

AQUEOUS SEPARATION PROCESSES ADDRESSING HOMOGENEOUS RECYCLING

Solvent extraction processes for homogeneous recycling typically comprise two cycles, a first cycle separating uranium and a second cycle separating TRU. A so-called GANEX (Grouped ActiNides EXtraction) process was developed and hot tested in France.[9-10] EURATOM projects ACSEPT and SACSESS saw the development of the EURO-GANEX[11-12] and CHALMEX[13] processes, the former having been hot tested.[14] Both processes are further developed in GENIORS. The possibility of developing a single-cycle process based on the oxidation of americium to Am(VI) is studied in the United States, co-extracting Np, Pu and Am.[15-17]

Further details on aqueous separation processes addressing homogeneous recycling are found in the literature.[18-20]

AQUEOUS SEPARATION PROCESSES ADDRESSING HETEROGENEOUS RECYCLING

Heterogeneous recycling requires separating uranium, plutonium and neptunium from americium and curium and from the fission and corrosion products. This is achieved e.g. by the well-established PUREX process (or by an evolution of PUREX developed in France,[21-22] allowing a co-recovery and a co-management of U and Pu), routing uranium, plutonium and — following some modifications to the process, also neptunium[23-24] — to the product streams. Americium and curium are directed to the raffinate solution (HAR), together with fission and corrosion products. To recover americium (or americium and curium) from HAR, a suite of solvent extraction processes have been developed internationally.[18-20, 25] A schematic overview is given in Figure 1, focussing on process schemes developed in Europe. The individual processes (i.e. the boxes in Figure 1) are briefly described below. Acronyms for extracting and complexing agents are explained in the Annex.

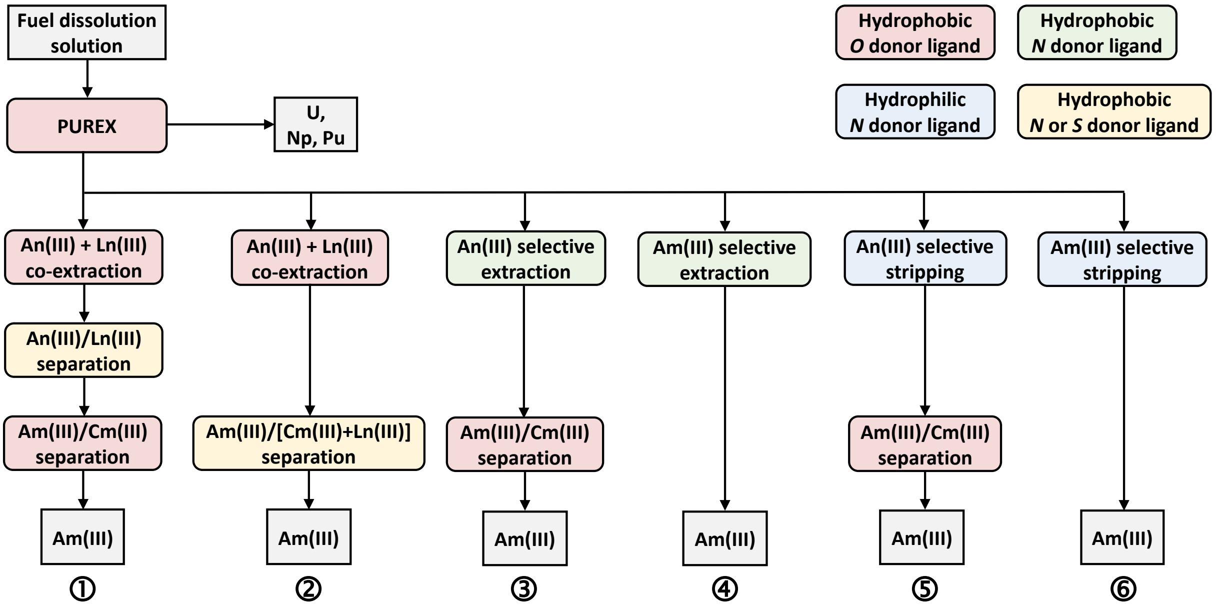


Figure 1. A schematic overview of solvent extraction processes for heterogeneous recycling.

ACTINIDES(III) AND LANTHANIDES(III) CO-EXTRACTION

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

Earlier actinides(III) separation schemes have been based on a process co-separating actinides(III) and chemically similar fission lanthanides(III) from HLW, separating them from other fission products and corrosion products. A second process follows, separating actinides(III) from lanthanides(III). The first process ideally is compatible with the high nitric acid concentrations present in HLW, typically 3–5 mol/L. Processes requiring a prior reduction of the nitric acid concentration (e.g. by partial neutralisation or by denitration) are not considered in the following discussion.

The TRUEX process,[26-27] developed at Argonne National Laboratory, USA, uses a solvent composed of CMPO + TBP in *n*-dodecane to co-extract actinides(III) and lanthanides(III).

To avoid the generation of secondary waste, extracting agents consisting of only *C, H, O* and *N* are advantageous: they and their degradation products can be incinerated without producing solid waste.[28] Thus, diamide extractants were developed at the CEA. So- called DIAMEX processes were developed and tested using spiked and hot feed solutions. Initially, DMDBTDMA[29] was used as the extracting agent which was later replaced by DMDOHEMA[30-33] due to its improved properties regarding solvent clean-up. These processes are directly compatible with the PUREX process, requiring no feed adjustment such as adjustment of the nitric acid concentration. The product solution contains actinides(III) and lanthanides(III) in dilute nitric acid. The raffinate contains non-lanthanide fission products and corrosion products plus oxalic acid and HEDTA (added to the feed and to the scrubbing solution to prevent the co-extraction of Zr, Mo and Pd) in nitric acid and is suitable for vitrification after destruction of organic complexing agents.

Based on early work by Stephan at al.,[34-35] diglycolamide extracting agents were proposed by Sasaki et al.[36-38] for the co-extraction of actinides(III) and lanthanides(III). Spiked[39] and hot[40] process tests using a TODGA solvent (0.2 mol/L TODGA + 0.5 mol/L TBP in TPH)[41] were performed in Jülich and at the ITU (now JRC Karlsruhe), respectively. 16 + 16 stages centrifugal contactor setups were used. In the spiked test, actinides(III) concentrations in the raffinate were below the limit of detection. The product solution, containing actinides(III) and lanthanides(III), was very clean, containing as little as 0.1% of the Zr and Pd present in the feed, and 1.8% of Ru. Similar results were obtained in the hot test, with Am(III) and Cm(III) raffinate decontamination factors of ≈ 40,000. The raffinate solution, containing non-lanthanide fission products and corrosion products plus oxalic acid and HEDTA, is suitable for vitrification. The product solution contains actinides(III) and lanthanides(III) in 0.01 mol/L HNO_3.

ACTINIDES(III)/LANTHANIDES(III) SEPARATION

An(III)/Ln(III) separation

The product solution from above processes, containing Am(III), Cm(III) and Ln(III) (more exactly, Y(III), La(III) and the “fission lanthanides”, Ce(III)–Dy(III)), is subjected to further processes to obtain a pure Am(III) + Cm(III) fraction. Since actinides(III) and lanthanides(III) behave chemically similar, they cannot be separated using usual extracting agents which co-ordinate via oxygen donor atoms. Separation is based on the fact that actinides(III) form slightly stronger complexes with 'soft donor atoms as do lanthanides(III).[42-43] Thus, extracting agents coordinating via nitrogen or sulphur donor atoms should extract actinides(III) preferentially over lanthanides(III). Earlier work reported such extracting agents;[44-46] however, these required too low aqueous phase acidity (pH ≥ 3) to be successfully applied for process development.

Breakthrough in this field was achieved in 1999 with the development of aromatic dithiophosphinic acids[47] in Jülich and of alkylated 2,6-bis(1,2,4-triazin-3-yl)-pyridines (BTPs)[48] in Karlsruhe, enabling the selective extraction of actinides(III) from ≥ 0.5 mol/L HNO_3.

A synergistic solvent comprising $(ClPh)_2$-PSSH acid and TOPO in *tert.*-butyl benzene was used in the ALINA process.[49] With a 24-stage centrifugal contactor setup, less than 0.1% of Am(III) reported to the raffinate and approximately 2% of the Ln(III) reported to the product solution. 22% of Cm(III) were lost to the raffinate; more stages would have been required to achieve better Cm(III) recovery. The ALINA process required a feed acidity of ≤ 0.5 mol/L, making it directly compatible to DIAMEX or TODGA actinides(III)-lanthanides(III) co-extraction processes. The product solution contained actinides(III) in 1.5 mol/L HNO_3, the raffinate contained lanthanides(III) in 0.6 mol/L HNO_3 and is suitable for vitrification.

Process tests using BTP extracting agents were less successful due to the limited chemical and radiolytic stability of early BTP compounds.[50] A chemically and radiolytically rather stable BTBP, $CyMe_4$-BTBP was developed at Reading University, UK.[51-52] Spiked[53] and hot[54] tests using $CyMe_4$-BTBP were performed in 20 or 16-stage centrifugal contactor setups. Very clean Am(III) + Cm(III) product solutions were obtained, containing only small fractions of the Ln(III) inventory. This process requires a feed nitric acid concentration of 1.3–2 mol/L, making it directly compatible to DIAMEX or TODGA actinides(III)-lanthanides(III) co-extraction processes following feed nitric acid concentration adjustment by addition of concentrated nitric acid. The product solution contained actinides(III) in a 0.5 mol/L glycolate solution, the raffinate contained lanthanides(III) in ≈ 1 mol/L HNO_3 and is suitable for vitrification.

AMERICIUM-CURIUM SEPARATION

Am(III)/Cm(III) separation

Processes yielding an actinides(III) product solution containing both americium and curium require an additional separation if only americium is to be recycled. Due to the similar chemical behaviour of Am(III) and Cm(III), most extracting agents have a low selectivity (most of the compounds studied showing a separation factor of approximately 1.6).

Nevertheless, separation is achieved by using a sufficient number of stages. The CEA performed an Am(III)/Cm(III) separation test with the extracting agent DMDOHEMA. A flow-sheet with 56 stages yielded decontamination factors of approximately 150.[55]

This process requires a feed nitric acid concentration in the range of 3 mol/L. The impact of any complexing agents present in the feed solution (coming from an upstream process) would have to be assessed. The raffinate solution (Cm(III) in nitric acid) is suitable for vitrification. The product solution is Am(III) in dilute nitric acid.

A different approach exploits the fact that in contrast to Cm(III), Am(III) can be oxidised to higher valences, enabling separation by common extracting agents.[56] This approach was studied at the CEA[57-58] but abandoned later due to issues with upstream compatibility. Nevertheless, new processes based on this concept are currently under development in the USA.[15-17]

AMERICIUM(III)/[CURIUM(III) + LANTHANIDES(III)] SEPARATION

Am(III)/[Cm(III)+Ln(III)] separation

Extracting only Am(III) from the product solution of processes such as TRUEX or DIAMEX requires a solvent preferentially extracting Am(III) over Cm(III) and Ln(III) — the other fission and the corrosion products already being separated in the upstream process.

Jülich developed a synergistic system comprising $(ClPh)_2PSSH$ and T2EHP, yielding an Am(III)/Cm(III) separation factor of approximately 7.[59] Applying this system in a 24-stage LUCA flow-sheet, the raffinate decontamination factor was > 1000 (i.e. less than 0.1% of americium was lost to the raffinate) and the product decontamination factor was 200 (i.e. 0.5% of curium reported to the americium product solution).[60] The LUCA process required a feed nitric acid concentration of < 0.1 mol/L; additionally, 0.5 mol/L $NaNO_3$ was added to warrant satisfactory phase separation in the extraction section. The product solution contained Am(III) in ≈ 0.7 mol/L HNO_3, the raffinate contained Cm(III) and lanthanides(III) in ≈ 0.1 mol/L HNO_3 + 0.15 mol/L $NaNO_3$. The impact of $NaNO_3$ on raffinate vitrification should be discussed.

Alternatively, systems based on *N*-donor extracting agents such as $CyMe_4$-BTBP[52] or $CyMe_4$-BTPhen,[61] combined with water soluble diglycolamides,[62] could be used to perform this separation. However, no process using such a system has so far been developed and tested. Such processes would require a feed nitric acid concentration of ≈ 3 mol/L. The raffinate would contain Cm(III), lanthanides(III) and TEDGA in nitric acid and should be suitable for vitrification. The product would be Am(III) in a solution of glycolate, citrate or similar.

ACTINIDES(III) SELECTIVE EXTRACTION

An(III) selective extraction

The 1-cycle SANEX process was developed at Jülich to directly separate Am(III) and Cm(III) from HLW.[63] A solvent containing $CyMe_4$-BTBP and TODGA in a kerosene/octanol diluent was used to extract Am(III) and Cm(III). The unwanted co-extraction of Zr(IV) and Mo(VI) was suppressed by adding oxalic acid to the feed. A flow-sheet was developed[64] and tested[65] using a spiked surrogate feed solution. This flow-sheet managed Pd(II) (which is co-extracted with

Am(III) and Cm(III)) in a selective Pd(II) stripping section using L-cysteine before stripping Am(III) and Cm(III) using a glycolate solution. Ni(II) and Cd(II), which were also co-extracted, stayed in the organic phase. A product solution was obtained containing ≥ 99.8% Am(III) and ≥ 99.4% Cm(III), 9% Y(III) and 0.8% Pd(II). Most other elements were well below 0.1%.

The 1-cycle SANEX process directly uses HAR as a feed solution; nitric acid concentration adjustment is not required. The product solution contains actinides(III) in a 0.5 mol/L glycolate solution, the raffinate contains fission products and oxalic acid in ≈ 2 mol/L HNO_3. A second raffinate solution is generated, containing Pd(II) + 10 mmol/L L-cysteine in 1 mol/L HNO_3. The sulphur content of this solution may pose problems during vitrification.

AMERICIUM(III) SELECTIVE EXTRACTION

Am(III) selective extraction

Based on their experience with the 1-cycle SANEX process,[63-65] Jülich developed a solvent extraction system combining $CyMe_4$-BTPhen and TEDGA and using bimet to suppress Pd(II) and Ag(I) extraction.[62] With Am(III)/Cm(III) separation factors in the range of 2–5, depending on experimental conditions, this system has the potential to extract only Am(III) from HAR. So far, no flow-sheet using this system has been developed and tested.

Such a process would require a feed nitric acid concentration of ≈ 3 mol/L, depending on the TEDGA concentration. That is, HAR could directly be used as a feed solution. The raffinate would contain Cm(III), fission and corrosion products, TEDGA and bimet in ≈ 2 mol/L. The product would be a solution of Am(III) in a 0.5 mol/L glycolate solution. As with the 1c-SANEX process, the sulphur content of the raffinate solution may pose problems during vitrification.

ACTINIDES(III) SELECTIVE STRIPPING

An(III) selective stripping

Based on the TALSPEAK process[66-67] developed at Oak Ridge National Laboratory in the 1960's, various processes were developed using chemistry that places selectivity for actinides(III) over lanthanides in the aqueous phase rather than using selective extracting agents. These processes use aqueous solutions containing aminopolycarboxylates such as DTPA or HEDTA to strip actinides(III) from a solvent loaded with actinides(III) and lanthanides(III). To make extraction from HAR (with its rather high nitric acid concentration) viable, a neutral extracting agent such as CMPO, DMDOHEMA or TODGA is required. Lanthanides(III) must remain in the organic phase under the low acidity conditions of the actinides(III) stripping by aminopolycarboxylates. This is achieved either by adding an acidic extracting agent to the solvent or by adding nitrate salt to the aminopolycarboxylate solution.

TALSPEAK-based processes such as TRUSPEAK,[68] advanced TALSPEAK,[69-71] ALSEP,[72-73] have been developed and tested in the USA. Solvents used comprise a neutral (CMPO or T2EHDGA) and an acidic (HDEHP or HEHEHP) extractant. Citrate-buffered solutions of DTPA or HEDTA are used to strip actinides(III). By synthesising systematically modified aminopolycarboxylates,[74-75] TALSPEAK chemistry is continuously being improved. Furthermore, the Japanese SETFICS[76-77] process is based on TALSPEAK chemistry.

Several versions of the so-called DIAMEX-SANEX process were developed and hot tested at the CEA.[55] These processes used a solvent comprising DMDOHEMA and HDEHP in TPH. A solution of HEDTA in a citrate buffer was used to strip actinides(III). Later a so-called i-SANEX process was developed and hot tested at CEA.[78] A TODGA based solvent (0.2 mol/L TODGA + 0.5 mol/L TBP in TPH)[41] was used; actinides(III) were selectively stripped using a solution of DTPA in a malonate buffer, containing $NaNO_3$ to keep the lanthanides(III) in the solvent.

To avoid the addition of an acidic extracting agent or of nitrate salt, stripping agents were developed that cope with nitric acid concentrations sufficient to keep lanthanides(III) in a solvent containing only a neutral extracting agent. In the framework of ACSEPT and SACSESS, such systems have been developed, based on a TODGA solvent (0.2 mol/L TODGA + 5% 1-octanol in TPH).[79-80]

The first one, developed at KIT and tested in Jülich, used SO_3-Ph-BTP to strip actinides(III).[81-82] A successful spiked process test was performed using a 16 + 16 stages centrifugal contactor setup.[83] Am(III) and Cm(III) losses were in the range of 0.1%. Ln(III) in the actinides(III) product solution were < 0.1%. Furthermore, the product contained low concentrations of Ru (0.4%), Zr (0.1%), Mo (0.1%) and Sr (0.3%).

Due to concerns with the sulphur content of the stripping agent, SO_3-Ph-BTP, an improved system was developed at Politecnico di Milano and Università di Parma. This system uses PTD to strip actinides(III).[84-85] A flow-sheet for a centrifugal contactor run has been calculated;[86] a counter-current flow-sheet test will be performed in the near future.

These two processes are compatible with a PUREX process, i.e. HAR can be used as a feed solution without prior nitric acid concentration adjustment. The raffinate solutions are suitable for vitrification. Product solutions are Am(III) and Cm(III) in nitric acid containing either SO_3-Ph-BTP or PTD. The use of SO_3-Ph-BTP gives rise to sulphur-containing solid waste, which should be avoided. PTD, on the other hand, is a *CHON* compound, allowing for its destruction to gaseous products.

AMERICIUM(III) SELECTIVE STRIPPING

Am(III) selective stripping

Using slightly different aqueous phase ligands than those described in the "actinides(III) selective stripping" section above allows for selectively stripping only Am(III) from an organic phase containing Am(III), Cm(III) and Ln(III).

The EXAm process[87-88] developed at the CEA used a DMDOHEMA + HDEHP solvent to co-extract actinides(III) and lanthanides(III). Cm(III) and heavier Ln(III) were scrubbed from the organic phase using TEDGA (a water soluble diglycolamide). Co-extracted Pd, Mo and Ru were scrubbed with citrate. Am(III) was stripped using a solution containing DTPA and malonate at pH = 2.5. Finally, light Ln(IIII) and Fe(III) were stripped into a solution containing TEDGA and oxalic acid. A successful hot test was performed in mixer-settler batteries. > 99% of Am(III) were extracted from the feed solution and 98.3% were recovered in the product solution, containing 0.2% Cm(III) (i.e. $DF_{\mathrm{Am/Cm}} = 500$). A further hot EXAm test was performed at the CEA, using a concentrated HAR (HAC) feed solution. Approximately 2.5 g americium was recovered and converted into $UAmO_2$ to be irradiated in a materials test reactor.[89]

An improved system was later developed at the CEA and in the SACSESS project: the EURO-EXAm system used a TODGA solvent to co-extract actinides(III) and lanthanides(III); TPAEN was used to selectively strip Am(III) from the loaded solvent.[90-92] Unfortunately, precipitation problems were encountered during counter-current centrifugal contactor runs performed during SACSESS.

By replacing the actinides(III) stripping agent, SO_3-Ph-BTP (used in one of the processes involving selective stripping of actinides(III)[83]), with the respective SO_3-Ph-BTBP, Am(III) can selectively be stripped from a TODGA solvent.[93-94] As a contribution to GENIORS, a flow-sheet is currently being developed and an "AmSel" process test will be performed in the centrifugal contactor setup at Jülich in the near future.

All of the above processes directly use HAR as their feed solution, requiring no prior nitric acid concentration adjustment. The raffinate solutions are compatible with vitrification. The EXAm product solution contains Am(III), HEDTA and citrate. The EURO-EXAm product solution contains Am(III) and TPAEN in $HNO_3/NaNO_3$. The AmSel product solution is Am(III) and 10–20 mmol/L SO_3-Ph-BTBP in ≈ 0.8 mol/L HNO_3. However, the sulphur content in the AmSel raffinate solution creates a waste problem.

COMPARISON OF PROCESS SCHEMES

In early European projects the technical challenges in developing ligands and potential processes that could achieve actinide(III)-lanthanide(III) separation were huge. Following the extraordinary successes of these projects, later projects (ACSEPT,[95] SACSESS[96]) have focused on defining and testing 'reference' processes. So far, the selection of such reference processes has been dominated by the process chemistry, i.e. considering properties such as separation factors, radiolytic stability, kinetics, CHON molecule design etc. In most cases, solvent extraction flows-heet tests have confirmed the performance of these references processes. In the most recent project (SACSESS[96]) and now in GENIORS, these reference processes must be compared against each other and with a greater consideration of the engineering issues that will determine whether they can be industrialised (safety, solvent clean up, choice of contactors, interfacing with other parts of the process etc.) and, very importantly, the overall complexity – at this stage of R&D it is assumed that simpler processes will be more economic to deploy. This will enable GENIORS and future projects to be able to focus limited resources most efficiently on the leading options and their optimisation to the point at which industry could take over implementation.

Looking at the different process schemes to separate Am(III) from HAR discussed above (Figure 1), schemes ④ and ⑥ appear the simplest (adding one box to PUREX), scheme ① the most complex (adding three boxes), schemes ②, ③ and ⑤ intermediate (two additional boxes). However, Figure 1 represents any of the solvent extraction cycles as one box. Each box contains extraction, scrubbing and stripping stages, spent solvent regeneration cycles and various auxiliary processes. The arrows connecting the boxes in reality will be more than a simple pipe; buffer tanks, feed adjustment, addition or destruction of auxiliary chemicals may be required to connect the solvent extraction cycles to one another. Thus, evaluating the different process schemes requires taking into account many aspects such as number of stages required to achieve certain decontamination factors, waste generation, upstream and downstream compatibility, solvent clean-up, safety of operation etc.

To assess the different process schemes ①–⑥, optimum individual solvent extraction processes (the boxes in Figure 1) are selected in case of more than one option. Then, the schemes ①–⑥ are compared to each other, taking into account the criteria mentioned above.

SELECTION OF OPTIMUM PROCESSES

ACTINIDES(III) AND LANTHANIDES(III) CO-EXTRACTION

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

Both the DMDOHEMA[30] and TODGA[40] based processes have thoroughly been evaluated in Europe. They are compatible with PUREX. The DMDOHEMA process is fully *CHON* compliant. To make the TODGA system[41] *CHON* compliant, TBP is replaced by 1-octanol.[79-80]

Similar performance was obtained for the two processes regarding decontamination factors and throughput. Reasonably fast kinetics[97] are advantageous. However, the TODGA process used significantly more stages (32 vs. 16 for the DMDOHEMA process). This is explained by a conservatively sized stripping section (16 stages were used). Indeed, the concentration profiles indicate that approximately six stripping stages would have sufficed.

Regarding the consumption of auxiliary chemicals (oxalic acid, HEDTA), higher concentrations were used in the TODGA test. However, lower concentrations of oxalic acid were used in a more recent process using a TODGA solvent.[83]

In conclusion, the two processes perform similarly, regarding both separation performance and radiolytic stability of the solvents applied.[33, 37, 98-99] A decision may be based on the solvent clean-up procedures required to maintain a continuously efficient and safe process performance.

ACTINIDES(III)/LANTHANIDES(III) SEPARATION

An(III)/Ln(III) separation

Two processes to separate actinides(III) from the product solution of DMDOHEMA[30] or TODGA[40] co-extraction processes showed promising results: the ALINA process[49] and the CyMe$_4$-BTBP based processes.[53-54]

The ALINA process[49] has good selectivity and fast kinetics,[100] making it a rather compact process. Equipment would be of similar size to the used in an upstream DMDOHEMA or TODGA co-extraction process. However, the solvent contains a neutral (TOPO) and an acidic ((ClPh)$_2$PSSH) extracting agent, possibly complicating solvent clean-up. Furthermore, the sulphur content will give rise to solid waste originating from solvent clean-up.

In contrast, the CyMe$_4$-BTBP based processes[53-54] are fully *CHON* compliant. A disadvantage is the rather slow kinetics[101] of solvents based on CyMe$_4$-BTBP: While the ALINA process had a feed flow-rate of 50 mL/h, the CyMe$_4$-BTBP based processes had feed flow-rates of 10 mL/h, using similarly sized equipment. In consequence, a CyMe$_4$-BTBP based process would require larger equipment to handle the product stream from an upstream DMDOHEMA or TODGA process. Since the actinides(III) product solution contains glycolate, the downstream compatibility needs to be assessed.

Considering the features discussed above, the CyMe$_4$-BTBP based processes were selected the European baseline process for separating actinides(III) from DIAMEX or TODGA process product solutions.

AMERICIUM(III)/[CURIUM(III) + LANTHANIDES(III)] SEPARATION

Am(III)/[Cm(III)+Ln(III)] separation

So far, one process has been developed and tested on a spiked feed, the LUCA process.[60] This process requires a comparatively low number of stages due to its good selectivity for Am(III) over Cm(III) and its fast kinetics. However, the solvent contains a neutral (T2EHP) and an acidic ((ClPh)$_2$PSSH) extracting agent, possibly complicating solvent clean-up. Furthermore, the sulphur content will give rise to solid waste originating from solvent clean-up.

A possible process based on CyMe$_4$-BTBP[52] or CyMe$_4$-BTPhen[61] in combination with TEDGA[62] has some promise but so far is only a concept (TRL 1). The rather slow kinetics[101] of solvents based on CyMe$_4$-BTBP or CyMe$_4$-BTPhen will require larger contactors.

ACTINIDES(III) SELECTIVE STRIPPING

An(III) selective stripping

The TALSPEAK based processes developed in the USA[68-75] and the French DIAMEX-SANEX process[55] rely on a solvent containing a neutral and an acidic extracting agent. Processes using a solvent containing only one extracting agent are advantageous with respect to solvent clean-up. This narrows down the choice to the CEA i-SANEX process,[78] the SO_3-Ph-BTP process[83] and the PTD system.[86]

The SO_3-Ph-BTP process,[83] despite its excellent performance, has a serious disadvantage, actually being a show stopper: the actinides(III) product solution contains the stripping agent, SO_3-Ph-BTP. Separating actinides(III) from SO_3-Ph-BTP would require an additional solvent extraction cycle and produce a sulphur containing secondary waste. Alternatively, recycling SO_3-Ph-BTP back to the process would require another solvent extraction cycle. With this, scheme ③, if implementing the SO_3-Ph-BTP process, actually becomes more complicated than e.g. scheme ①.

The CEA i-SANEX process[78] was successfully hot tested. This process has two drawbacks: a rather pronounced pH dependence of the actinides(III) stripping section's performance and the requirement of adding nitrate salt to keep lanthanides(III) in the solvent during actinides(III) stripping.

Finally, the PTD system[86] shows promise. It is fully *CHON* compliant, nitric acid is used to keep lanthanides(III) in the solvent during actinides(III) stripping, the kinetics is reasonably fast. Pending a successful process demonstration, this system has the potential of becoming the European reference flow-sheet for a process based on the selective striping of actinides(III) from a solvent loaded with actinides(III) and lanthanides(III).

AMERICIUM(III) SELECTIVE STRIPPING

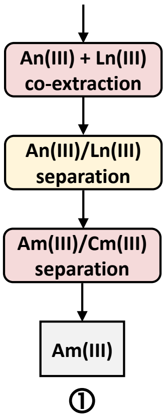
Am(III) selective stripping

So far, only the EXAm process[87-88] has been demonstrated. All three processes discussed — EXAm, EURO-EXAm[90-92] and AmSel[93-94] — each have significant drawbacks: EXAm uses a solvent containing an acidic and a neutral extracting agent. This may require a more complex solvent clean-up cycle. EURO-EXAm, further to not yet having been demonstrated, requires adding nitrate to the Am(III) stripping solution. AmSel uses a sulphur-containing Am(III) stripping agent, SO_3-Ph-BTBP, resulting in the same disadvantages the SO_3-Ph-BTP process[83] suffers from. Further basic research is needed to identify optimised options for americium(III) selective systems.

COMPARISON OF SCHEMES ①–⑥

Considering the above discussions, the most promising variations of schemes ①–⑥ are combined from the individual solvent extraction processes, and the schemes are assessed and compared against one another.

SCHEME ①



This scheme adds three solvent extraction processes to the PUREX process: one process to co-extract actinides(III) and lanthanides(III) from the HAR solution, one to separate actinides(III) from lanthanides(III) and one to separate Am(III) from Cm(III).

The DMDOHEMA[30] and TODGA[40] based processes have similar performance. Hence, no clear recommendation is made as to which process is most favourably used to co-extract actinides(III) and lanthanides(III) from HAR. However, a selection may be made depending on which process is used to separate Am(III) from Cm(III), see below.

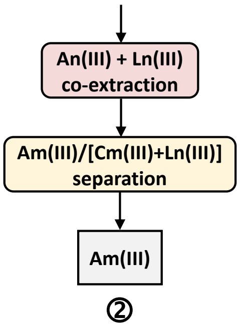
To separate actinides(III) from lanthanides(III), the CyMe$_4$-BTBP process is the European reference and baseline process. Whether to add TODGA[53] or DMDOHEMA[54] as phase transfer catalyst again should be decided based on which extracting agent is used in the first and third solvent extraction processes, in order to minimise the number of extracting agents in use. However, the relatively slow kinetics of these processes will require somewhat larger equipment. Finding a solvent extraction system with faster kinetics but otherwise comparable performance would greatly help.

To separate Am(III) from Cm(III), the DMDOHEMA process[55] is suitable. To make this process more compact (i.e. reduce the number of stages), the addition of TEDGA should be considered, which would help increase the Am(III)/Cm(III) selectivity, see the EXAm process.[87-88]

A possibly promising direction of research would be developing an Am(III)/Cm(III) separation process using TODGA (which extracts Cm(III) slightly better than Am(III)) as extracting agent and searching for water soluble complexing agents with the reverse selectivity.

In conclusion, scheme ① appears the most complex. However, using the same extracting agent in the first and in the third solvent extraction cycles would reduce the number of solvent clean-up cycles to two. This way, this scheme would be less complex than it appears at first sight.

SCHEME ②



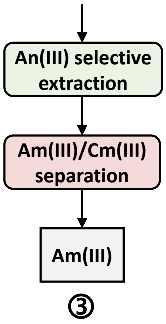
This scheme adds two solvent extraction processes to the PUREX process, one process to co-extract actinides(III) and lanthanides(III) from the HAR solution and one to separate Am(III) from Cm(III) and the lanthanides(III).

Regarding the first process, again, both the DMDOHEMA[30] and TODGA[40] based processes appear similarly suitable.

Using the LUCA process[60] to separate Am(III) from Cm(III) and lanthanides(III) is not optimal since it uses a mixture of an acidic and a neutral extracting agent; furthermore, one of them contains sulphur. Alternative processes, based on CyMe$_4$-BTBP, may be a possibility. However, besides not having been developed and demonstrated, such processes will suffer from slow kinetics.

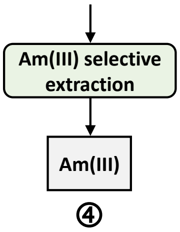
In any case, scheme ② involves two different solvents, requiring two solvent clean-up cycles.

SCHEME ③



This scheme adds two solvent extraction processes to the PUREX process, one process to extract actinides(III) from the HAR solution and one to separate Am(III) from Cm(III). The bottleneck of this scheme is the extraction of actinides(III) by the 1c-SANEX process,[63-65] for two reasons: (a) the slow kinetics will require comparatively large equipment; (b) the use of a sulphur-containing Pd(II) stripping agent continuously routes sulphur to the raffinate, creating issues with vitrification.

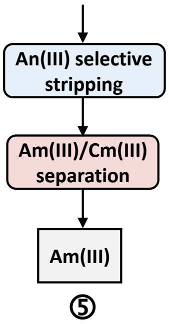
SCHEME ④



This scheme adds a single solvent extraction process to the PUREX process. This process ("Am(III) selective extraction") would directly extract only Am(III) from the HAR solution. The only chemical system developed so far[62] shares the drawbacks of the 1c-SANEX process (see scheme ③) regarding kinetics and sulphur-containing waste. Furthermore, a flow-sheet based on this system has not yet been developed and tested.

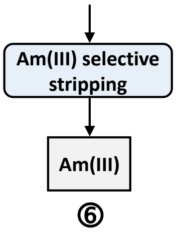
SCHEME ⑤

This scheme adds two solvent extraction cycles to the PUREX process. The first one would most favourably use the PTD system[86] to separate actinides(III) from the HAR solution. Regarding the second process to separate Am(III) from Cm(III), see the discussion on scheme ①. Again, developing a TODGA-based Am(III)/Cm(III) separation system would be beneficial since the PTD system[86] also uses TODGA: only one solvent clean-up cycle would be necessary to regenerate the solvent from both cycles.



SCHEME ⑥

This scheme is very compact as it adds only one solvent extraction cycle to the PUREX process. However, so far no Am(III) selective stripping process without more or less substantial drawbacks is available.



CONCLUSIONS

Several solvent extraction processes addressing heterogeneous recycling have been developed and demonstrated in Europe. Based on the most promising of these solvent extraction processes, six schemes for separating americium from PUREX HAR were composed and compared against one another. The following conclusions are drawn, based on the current state of the art:

1. Schemes ③ and ④, if relying on the processes developed and demonstrated so far, create a sulphur-containing raffinate solution. This is expected to cause problems with the vitrification of these solutions. Therefore, schemes ③ and ④ appear the least suitable.
2. Scheme ⑥ appears promising as it adds only one additional solvent extraction process. Unfortunately, so far, none of the systems available for developing such a process is without drawbacks of varying significance.
3. Scheme ② appears promising but so far has only been demonstrated using an extraction system which is not *CHON* compliant.
4. Scheme ① appears the most complex. However, using the same extracting agent in the first and in the third solvent extraction cycles would reduce the number of solvent clean-up cycles to two. This way, this scheme would be less complex than it appears at first sight. The second solvent extraction cycle (actinides(III)/lanthanides(III) separation) could benefit from developing a system with faster kinetics.
5. Finally, **scheme ⑤ appears the most promising option** given that an actinides(III) selective extraction process using PTD will be successfully demonstrated (which is to

be expected for the near future). In case an Am(III)/Cm(III) separation system based on the use of TODGA becomes available, scheme ⑤ will be rather compact, adding two solvent extraction processes with one common solvent clean-up cycle.

FURTHER DIRECTIONS FOR R&D

Based on the discussion of the various solvent extraction cycles and process schemes, some directions for future R&D emerge. These aim at improving and simplifying heterogeneous recycling.

Scheme ① would benefit from a nitrogen donor extracting agent with properties similar to those of CyMe$_4$-BTBP but with substantially faster kinetics. CA-BTP[102-103] appeared promising; however, no actinides(III)/lanthanides(III) separation process using CA-BTP has been developed and tested.

Scheme ⑥, being one of the simplest schemes, would become viable if a water soluble *CHON* nitrogen donor ligand with the following properties was found: it must form stronger complexes with Am(III) than with Cm(III) and lanthanides(III) and it must remain efficient in ≈ 1 mol/L nitric acid.

Schemes ① and ⑤ use TODGA to co-extract actinides(III) and lanthanides(III) from HAR. Furthermore, they rely on a solvent extraction process to separate Am(III) from Cm(III). Developing an Am(III)/Cm(III) separation process using TODGA (which extracts Cm(III) slightly better than Am(III)) as extracting agent and searching for water soluble complexing agents with the reverse selectivity would simplify these schemes: this way, a common solvent clean-up cycle could be used.

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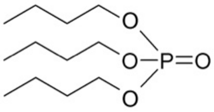
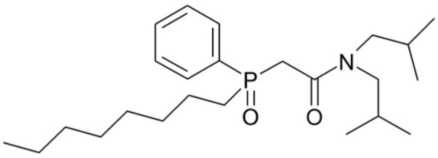
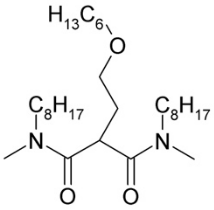
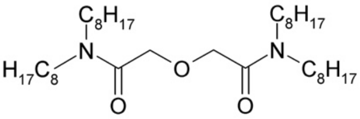
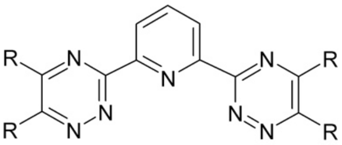
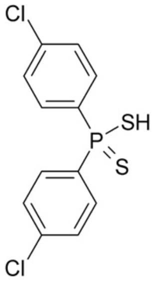
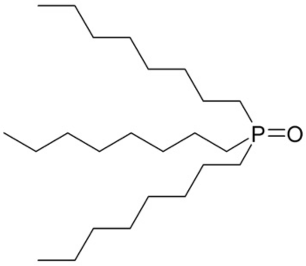
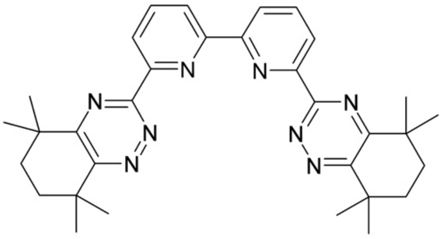
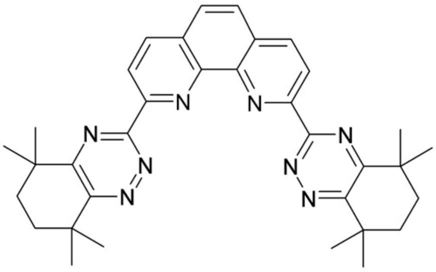
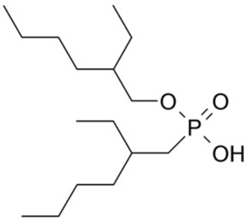
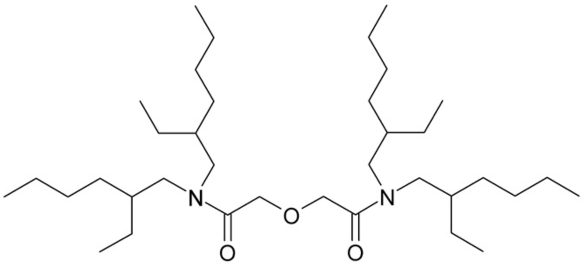
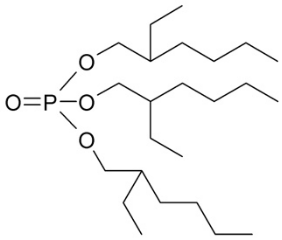
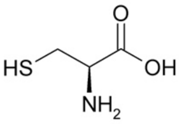
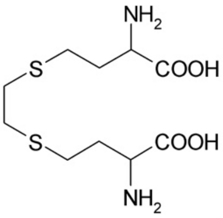
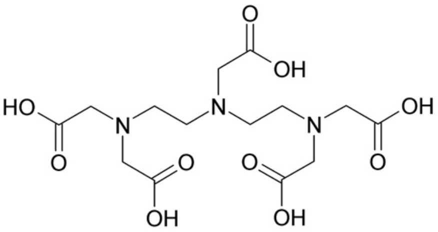
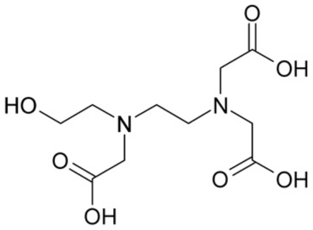
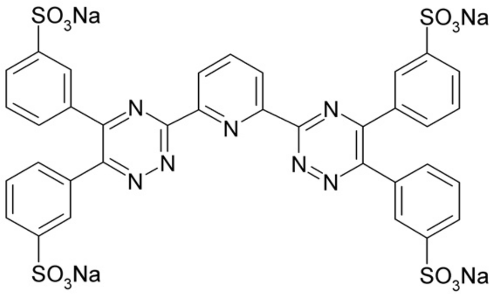
APPENDIX: LIST OF COMPOUNDS

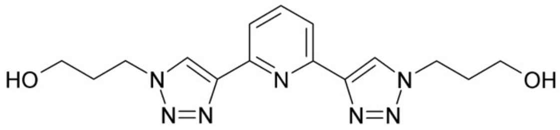
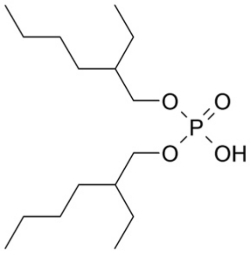
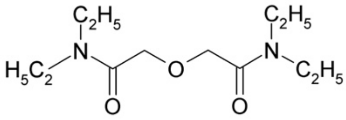
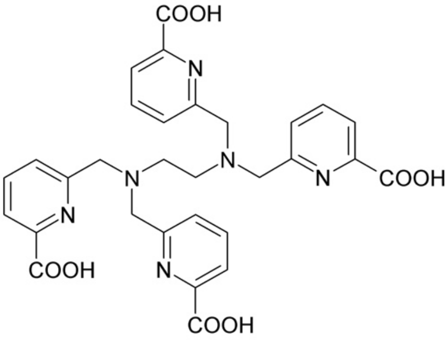
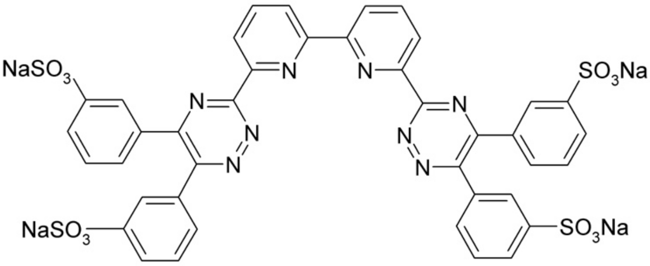
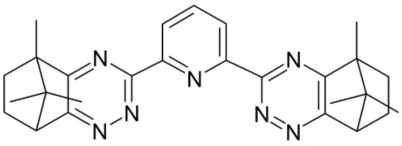
The structures and names of extracting and complexing agents mentioned in the report are given in below table, in order of appearance in the text.

Compound	Acronym	Name
	TBP	tri-*n*-butyl phosphate
	CMPO	*n*-octyl(phenyl)-*N,N*-diisobutyl-carbamoylmethyl-phosphine oxide
	DMDOHEMA	*N,N'*-dimethyl-*N,N'*-dioctyl-2-(2-hexyloxy-ethyl)-malonamide
	TODGA	*N,N,N',N'*-tetra-*n*-octyl diglycolamide
	BTP	2,6-bis(5,6-dialkyl-1,2,4-triazin-3-yl)-pyridine
	(ClPh)$_2$PSSH	bis(chlorophenyl)dithio-phosphinic acid

Compound	Acronym	Name
	TOPO	tri-*n*-octylphosphine oxide
	CyMe$_4$-BTBP	6,6’-bis(5,5,8,8-tetramethyl-5,6,7,8-tetrahydro-benzo-1,2,4-triazin-3-yl)-2,2’-bipyridine
	CyMe$_4$-BTPhen	2,9-bis(5,5,8,8-tetramethyl-5,6,7,8-tetrahydro-benzo-1,2,4-triazin-3-yl)- 1,10-phenanthroline
	HEHEHP	2-ethylhexylphosphonic acid mono-2-ethylhexyl ester
	T2EHDGA	*N,N,N’,N’*-tetrakis-(2-ethylhexyl) diglycolamide

Compound	Acronym	Name
	T2EHP	tris(2-ethylhexyl) phosphate
	L-cysteine	L-cysteine
	bimet	(2S,2’S)-4,4’-(ethane-1,2-diylbis(sulfanediyl))-bis(2-aminobutanoic acid)
	DTPA	diethylenetriamine-pentaacetic acid
	HEDTA	*N*-(hydroxyethyl)-ethylenediamine-triacetic acid
	SO_3-Ph-BTP	2,6-bis(5,6-di(3-sulphophenyl)-1,2,4-triazin-3-yl)-pyridine tetrasodium salt

This project has received funding from the EURATOM research and training programme 2014–2018 under grant agreement No. 730227

Compound	Acronym	Name
	PTD	2,6-bis[1-(propan-1-ol)-1,2,3-triazol-4-yl]-pyridine
	HDEHP	bis(2-ethylhexyl)-phosphate
	TEDGA	*N,N,N′,N′*-tetraethyl diglycolamide
	TPAEN	*N,N,N′,N′*-tetrakis-[(6-carboxypyridin-2-yl)-methyl]ethylene-diamine
	SO_3-Ph-BTBP	6,6′-bis(5,6-di(3-sulphophenyl)-1,2,4-triazin-3-yl)-2,2′-bipyridine tetrasodium salt
	CA-BTP	Bis-2,6-(5,6,7,8-tetrahydro-5,9,9-trimethyl-5,8-methano-1,2,4-benzotriazin-3-yl)-pyridine

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