

Overview of the international R&D recycling activities of the nuclear fuel cycle

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Abstract: Nuclear power has demonstrated over the last thirty years its capacity to produce base-load electricity at a low, predictable and stable cost due to the very low economic dependence of the price of uranium. However the management of used nuclear fuel (UNF) remains the “Achilles’ heel of this energy source since the storage of UNF is increasing as evidenced by the following number with 2,000 to 2,300 of UNF produced each year by the 104 US nuclear reactor units which equates to a total of 62,000 UNF assemblies stored in dry cask storage and 88,000 stored in pools. Alarming, more than half of US commercial reactor sites have filled their pools to capacity and have had to add dry cask storage facilities. Two options adopted by several countries will be discussed. The first one adopted by Europe, Japan and Russia consists of recycling the used nuclear fuel after irradiation in a nuclear reactor. Ninety six percent of uranium and plutonium contained in the spent fuel could be reused to produce electricity and are worth recycling. The separation of uranium and plutonium from the wastes is realized through the industrial PUREX process so that they can be recycled for re-use in a nuclear reactor as a mixed oxide (MOX) fuel. The second option undertaken by Finland, Sweden and the United States implies the direct disposal of UNF into a geologic formation. One has to remind that only 30% of the worldwide UNF are currently recycled, the larger part being stored (90% in pool) waiting for scientific or political decisions. A third option is emerging with a closed fuel cycle which will improve the global sustainability of nuclear energy. This option will not only decrease the volume amount of nuclear waste but also the long-term radiotoxicity of the final waste, as well as improving the long-term safety and the heat-loading of the final repository. At the present time, numerous countries are focusing on the R&D recycling activities of the ultimate waste composed of fission products and minor actinides (americium and curium). Several new chemical extraction processes, such as TRUSPEAK, EXAM, or LUCA processes are pursued worldwide and their approaches will be highlighted.

Keyword: Reprocessing; recycling; used nuclear fuel; nuclear repository

1 Introduction

Deployment and expansion of low-carbon energy sources such as nuclear energy implies a robust nuclear waste management program. As of today, two strategies are offered to countries using nuclear energy. Some countries such as USA, Sweden, Finland, or Canada have opted for a once-through fuel cycle (Fig. 1), or geological disposal of used nuclear fuel (UNF). Other countries such as France, Japan, Russia, UK, and India have decided to recycle UNF (Fig. 1) using the available industrial PUREX process, followed by conditioning of high level waste in borosilicate glass for final geologic disposal. A third option called closed fuel cycle, which may integrate a modified PUREX process, envisions the recycling of minor actinides (MA) and fission products (FP) to be burned into a fast reactor or used as target for transmutation in

accelerator-driven-system (ADS) (Fig. 1). Reusing nuclear materials from light water reactors (LWR) used fuel not only supplies fissile and fertile fuels needed for fast reactors but also reduces the potential radiotoxic inventory of high level waste (HLW) to be disposed of while reusing used MOX fuels. The R&D efforts on the separation of MA have been under investigation for several years worldwide. Recycling MA is not a necessity but it is an option that may contribute to optimizing closed fuel cycle strategies while contributing 1- to reducing the long-term decay heat and radiotoxic inventory of waste disposed in a geologic formation, 2- to slightly increasing the energy generated from natural uranium (above 90% vs. about 80% with recycle of U/Pu only), and 3- to possibly enhancing proliferation resistance (while increasing the decay heat and neutron source in re-fabricated nuclear fuel). In the present paper, we are giving an overview of the various R&D recycling

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activities undertaken by different countries for improving the back end of the nuclear fuel cycle. In Table 1, according to Carre and Delbecq^[1], multiple recycling of MA in fast neutron reactors, which implies their extraction from a PUREX-based raffinate, would result in accumulated amount of

transuranic elements (TRU) of 100 metric tons as opposed to a once through fuel cycle with 1,680 metric tons of accumulated TRUS over a period of 100 years from 2020 to 2120. The once-through fuel cycle will take 250,000 years to recede to the uranium ore level radiotoxicity. A modified open

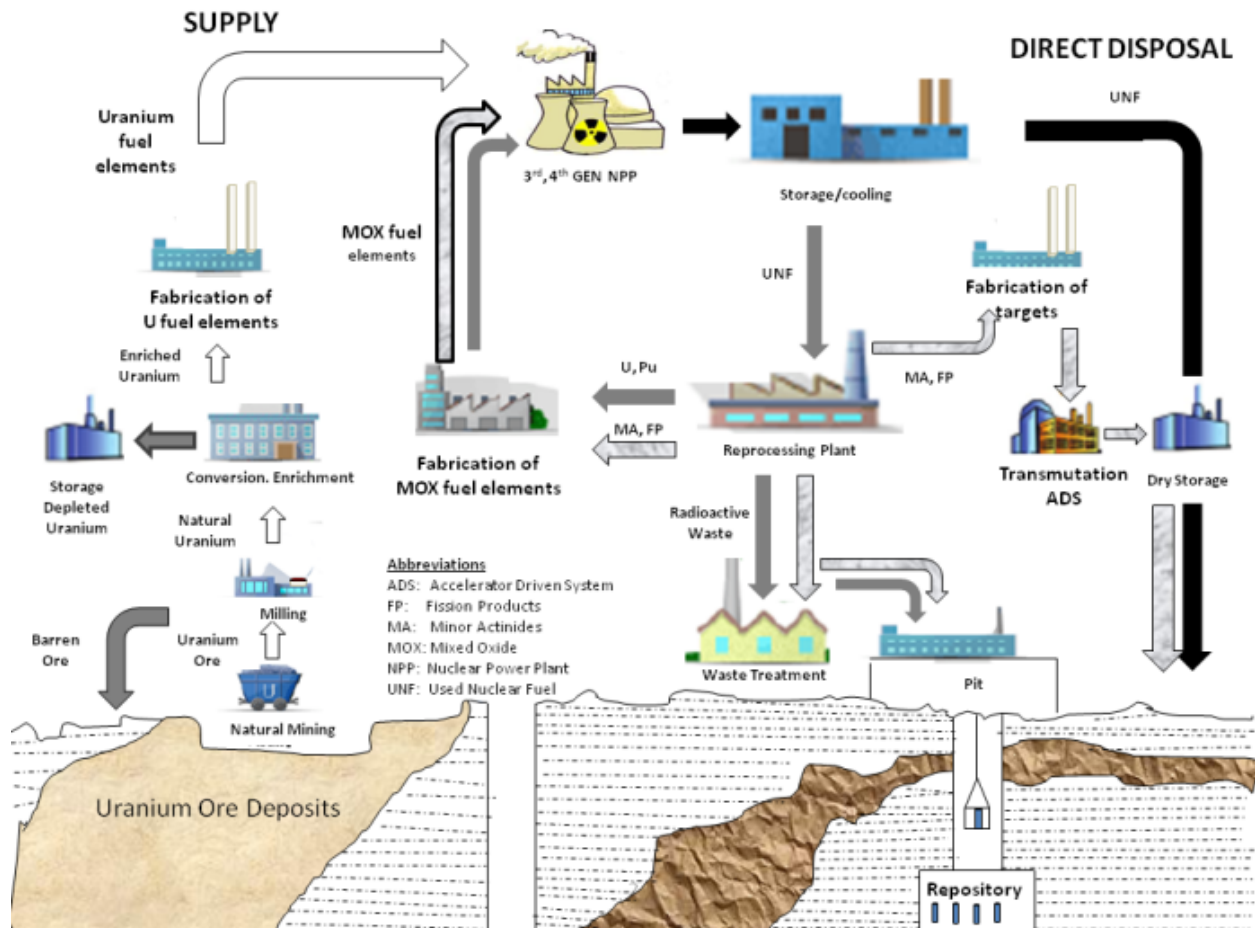


Fig.1 Nuclear fuel cycle depicting the three available strategies:
once-through fuel cycle,
modified open cycle and closed fuel cycle.

**Table 1 Comparison of the characteristics of (i) one-through fuel cycle,
(ii) modified open cycle and (iii) closed fuel cycle**

	Cumulated amount of TRUs to go into final disposal (Scenario of 100 years with a nuclear fleet capacity of 60 GWe) ^[1]	Decay time of TRU to reach relative radiotoxicity level of uranium ore ^[2]	Content of HLW	Research needed
Once through fuel cycle	1680 metric tons	250,000	U, Pu, Np, Am, Cm, FP	Actinide behavior under repository condition
Modified cycle (PUREX based process)	1,230 metric tons	10,000 years	Np, Am, Cm, FP	Improved waste-forms
Closed fuel cycle (FBR fleet deployment with MA recycling)	100	300 years	FP	Advanced recycling technologies involving, Am alone, Am & Cm, Pu & MA

cycle with the fabrication of a MOX fuel (U-Pu)O₂ will see a reduced radiotoxicity by about one order of magnitude while with a closed fuel cycle, it will take 300 years for fission products to decay below the uranium ore radiotoxicity level^[2].

2 Recycling the used nuclear fuel

2.1 The PUREX and COEXTM processes

The PUREX process has been the basis for commercial UNF recycling worldwide for over 30 years^[3]. This process is based on liquid-liquid extraction of hexavalent uranium and tetravalent plutonium by a mixture of tri-n-butylphosphate (TBP) and dodecane. The PUREX process allows minor actinides (americium, curium, and a fraction of the neptunium) to remain in the high-level liquid waste, which will be vitrified. This process has demonstrated high process efficiency and reliability, while providing a high quality product for producing UO₂ and mixed oxide fuels from recycled material (Fig 2a). To reduce the risk of plutonium diversion from the PUREX process, as shown in Fig. 2a, the COEXTM process (Fig. 2b) is being developed by AREVA. This is an evolutionary process, based on the PUREX process, designed to eliminate the production of pure plutonium product stream. By not separating plutonium and by being compatible with future advanced MA recycling processes, the COEXTM

process aims at further enhancing proliferation resistance^[4]. The main characteristics of COEXTM are 1) no plutonium separation at any point of the process, and 2) co-precipitation by oxalic acid to produce a mixed solid solution of uranium and plutonium dioxide (U, Pu)O₂.

For future advanced nuclear systems, minor actinides are considered more as a resource to be recycled and transmuted than to be disposed off directly into a nuclear repository. A key feature of advanced fuel cycles technologies would be to separate MA and ultimately americium from curium. Several countries are investigating the separation of MA from a PUREX/COEXTM based process raffinate or a modified PUREX process raffinate using new extractant molecules with two potential options for actinide separations:

1- A selective separation of MA for interim storage, pending a decision regarding their transmutation in heterogeneous recycling mode either in fast reactor (blankets) or in ADS. As an example, the DIAMide EXtraction (DIAMEX)/ Selective ActiNide EXtraction (SANEX) processes developed in France^[5], TRUEX/TALSPEAK processes developed in the United States^[6-8], ARTIST process developed in Japan (Fig. 3) are aiming to separate selectively MA. 2- Other simplified processes are aiming to separate a group of actinides using an integrated fuel cycle (with

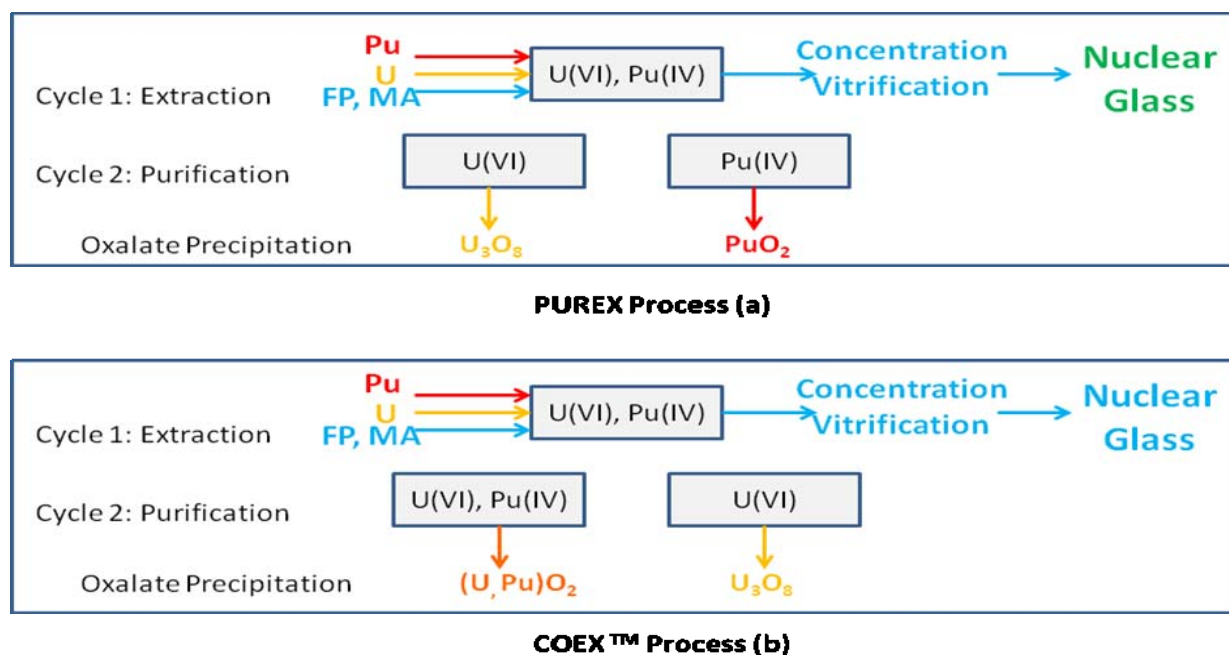
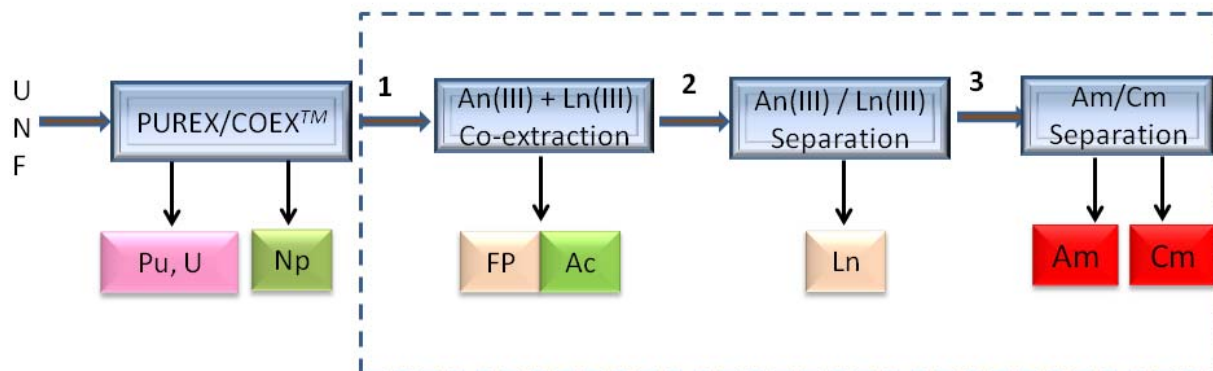


Fig.2 Global recycling scheme of PUREX (a) and COEXTM (b) processes.



After PUREX/COEX™ process

Step 1- Co-extraction of trivalent actinides An(III), and trivalent lanthanides Ln(III)

Step 2- Separation of trivalent actinides An(III) from trivalent lanthanides Ln(III)

Step 3- Separation of trivalent americium (Am(III)) from trivalent curium (Cm(III))

Fig. 3 Minor actinides separation strategy.

online fuel recycling and re-fabrication) with the prospect of their homogeneous recycling in a fast reactor (i-SANEX and GANEX processes developed in Europe, TRUSPEAK process developed in the USA).

2.2 Advanced recycling technologies

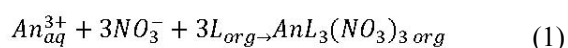
Fundamental R&D research activities are prerequisite to the development of efficient advanced separation processes especially for trivalent actinides/lanthanides separation. It is essential that lanthanides be separated from minor actinides as lanthanides compete with them for neutrons in both thermal and fast reactors, thus limiting efficiency. The chemical similarities for trivalent actinides and lanthanides (*e.g.* strong hydration and similar ionic radii) make separation very challenging. To accomplish the separation, extractants with soft donors atoms like sulfur or nitrogen have been tested to take advantage of the enhanced covalent bonding contribution that can occur with actinides. This

stronger interaction strength of actinides for soft donor atoms (N, S, Cl) is used worldwide in designing and synthesizing new ligands.

2.2.1 Co-extraction followed by separation of trivalent actinides and trivalent lanthanides

In France, the DIAMEX/SANEX processes (Fig. 4), studied at the Commissariat à l'Énergie Atomique (CEA) Marcoule, aims at recovering MA selectively from PUREX raffinate, by solvent extraction. One of the reference molecule of the DIAMEX process is N,N'-DiMethyl-N,N'-DiOctyl-Hexyl-Ethoxy-Malonamide (DMDOHEMA). This malonamide is used alone as the extractant, whereas it is mixed with di(2-ethylhexyl)phosphoric acid (HDEHP), an acidic extractant, in the other parts of the process. Synergetic effects have been observed when mixing DMDOHEMA with HDEHP at 0.5M – 1M HNO₃ while antagonism was noticed at lower acidity with the formation of an adduct "HDEHP-DMDOHEMA" which decreases the effective HDEHP concentration [9]. The diamide derivatives possess a good

extractability and respond to the CHON concept (completely incinerable and produces no radioactive solid waste by combustion). Americium and curium are then separated from the lanthanides in a following SANEX process (in which the reference molecule is 2,6-Bis(5,5,8,8-tetrahydro-1,2,4-benzotriazin-3-yl)pyridine (CyMe4-BTBP)). Several nitrogen terdentate ligands were studied but the most exciting results were obtained with the pyridine-bis-1,2,4-triazines derivatives (BTPs), first synthesized by Kolarik [10] and showed interesting properties such as separation of trivalent actinides from trivalent lanthanides with a separation factor as high as $SF_{Am/Eu} = 140$ and this for high aqueous nitric acid concentration. An(III) extraction proceeds through a solvation mechanism involving three molecules of BTP, as shown in the following reaction (1):



with $L = BTP$.

So far, around 140 new compounds have been synthesized in Europe and screened following standardized protocols. 75% of these compounds are lipophilic extracting agents whereas the remaining 25 % are hydrophilic complexing agents [11]. The Lanthaniden Und Curium Americium (LUCA)

process is developed in Germany for the selective separation of Am(III) from solution issued either from the DIAMEX-SANEX processes or the GANEX process. It uses a mixture of bis(chlorophenyl)dithiophosphinic acid ((ClPh)₂PSSH) and tris(2-ethylhexyl)phosphate (TEHP) as the extractant. More than 99.5% of Cm(III) was directed to the raffinate [12]. However, considerable degradation (by oxidation) of the (ClPh)₂PSSH has been observed at higher acidity (>0.5M HNO₃). Since 2008, the development and validation of the EXAm process (Extraction of Americium) is being pursued in France. Tridentate hydrosoluble tetraethyldiglycolamide (TEDGA) has been introduced in the EXAm process to complex Cm(III) and heavier lanthanides and allows the recovery of Am(III) alone [13]. In the United States, the TRUEX solvent extraction process (Fig. 4) is capable of separating, with very high efficiency, small quantities of transuranic elements (*e.g.*, Np, Am, Pu, Cm) from aqueous nitrate or chloride solutions that are typically generated in reprocessing plant operations or in plutonium production and purification operations [14]. The TRUEX process solvent contains octylphenyl-N,N-diisobutylcarbamoylmethyl phosphine oxide (CMPO) as the ligand while the diluents is composed of TBP and dodecane. Recent

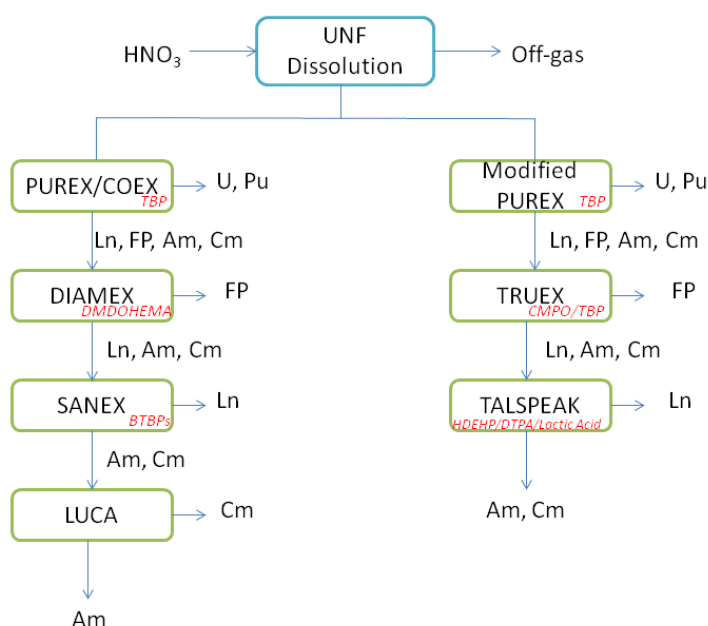


Fig. 4 Processes involving a co-extraction followed by separation of minor actinides.

investigations ^[15] evaluated the effects of radiolytic degradation upon the efficacy of the TRUEX flowsheet and showed that gamma irradiation adversely impacted flowsheet performance as measured by the decreasing americium and europium distribution ratios in the extraction section as accumulated dose increased. The TALSPEAK process (Fig. 4) relies on the aqueous soluble complexing agent diethylenetriaminepentaacetic acid (DTPA) to selectively retain actinides in the aqueous phase while the lanthanides are extracted by HDEHP in the presence of DTPA, thereby achieving a separation of the lanthanides from the actinides ^[7]. TALSPEAK process could be applied to a TRUEX raffinate (after adjustment) to separate TRU elements from the lanthanides (Fig. 4).

2.2.2 Simplified recovery of minor actinides from PUREX or modified PUREX process raffinate

In Japan, the ARTIST process uses advanced amide-based extractants such as the tridentate ligand *N,N,N',N'*-Tetraoctyldiglycolamide (TODGA) ^[16] to separate actinides from fission products. TODGA has good radiolytic and hydrolytic stability properties which makes it a strong candidate for trivalent actinides separation. The separation produces two streams, U- and TRU- bearing phases that are then solidified and stored for future treatment. The

radiolytic degradation products of TODGA were determined to be *N,N*-dioctyldiglycolamic acid, *N,N*-dioctylamine, and various *N,N*-dioctylmonoamides, which were formed by ruptures of amide- and ether-bonds in TODGA. The radiolysis of TODGA was enhanced in the presence of *n*-dodecane ^[17]. In Europe, alternatives concept to PUREX-DIAMEX-SANEX processes are being developed with either a one-cycle SANEX process or a new GANEX process (Fig. 5). The one-cycle SANEX (innovative SANEX or *i*-SANEX) separates MAs directly from a PUREX raffinate by combining TODGA and SO₃-Ph-BTP molecules ^[18]. The benefit with this process is the reduction of process steps from two to one compared with the DIAMEX+SANEX concept. Here the trivalent actinides and trivalent lanthanides are first co-extracted, leaving the remaining fission and corrosion products in the raffinate, followed by a selective stripping of the actinides by SO₃-Ph-BTP ^[19]. The new GANEX ^[20] aims at the recovery of all transuranium elements from the high activity used nuclear fuel dissolution solution. It proceeds in two cycles. The first cycle extracts selectively uranium while the second cycle achieves the co-extraction of all transuranium elements (Fig. 5). The solvent consists of a mixture of TODGA and DMDOHEMA in Exxsol D80. The undesired co-extraction of some

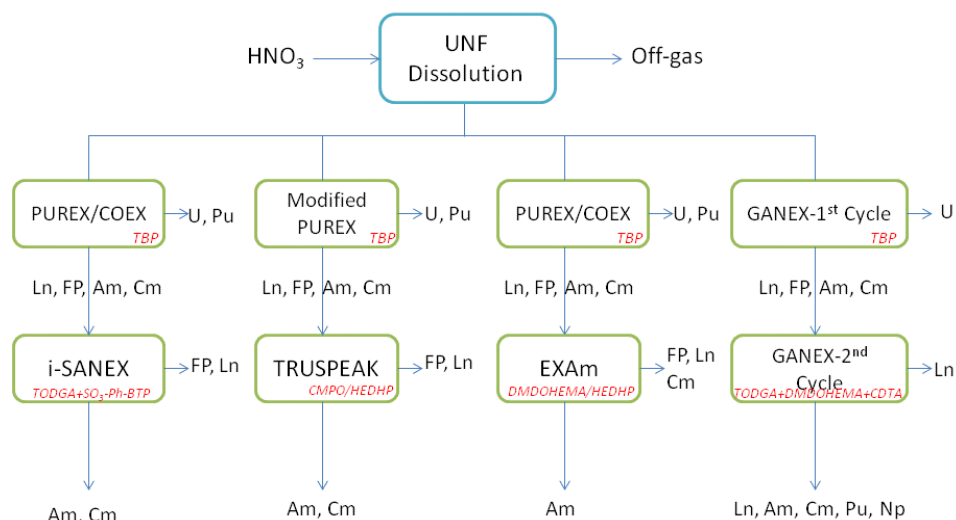


Fig. 5 Simplified processes for minor actinides recovery.

fission products is realized by adding 1,2-cyclohexanediaminetetraacetic acid (CDTA). In the United States, Lumetta et al.^[21] are developing a single process called TRUSPEAK process (Fig. 5) which combines the attributes of TRUEX and TALSPEAK processes. The extractants from both processes can be combined into a single process solvent to separate 1) the lanthanides and actinides from acidic high-level waste (HLW) and 2) the actinides from the lanthanides in a single solvent extraction process. CMPO is combined with the TALSPEAK solvent HDEHP in dodecane. In doing this, it is envisioned that the CMPO chemistry would dominate under conditions of high acidity (≥ 1 M HNO_3), resulting in co-extraction of the transuranium elements and lanthanide elements into the organic phase. After suitable scrubbing steps, contacting the loaded solvent with a buffered DTPA solution at pH ~ 3 to 4 will result in condition in which the HDEHP chemistry dominates, and the system will behave in a manner analogous to a reverse TALSPEAK process. The greater affinity of DTPA for the transuranium ions (TRUs) versus the lanthanides causes the TRUs to be selectively stripped into the aqueous phase, thereby separating them from the lanthanides.

3 Direct disposal of used nuclear fuel

Advanced recycling technologies do not eliminate the need for eventual permanent disposal of radioactive wastes, and therefore, in all cases, a geologic repository will be needed for final disposal of nuclear wastes. In Europe, the directive on the management of UNF and radioactive waste was adopted in July 2011 by the EU council. Consequently each member state is required to have long-term plans in place for managing nuclear waste by 2015. Finland, for example, has chosen a once-through fuel cycle and is building a national repository at 1,600 feet below ground at Olkiluoto Island. For the next two years, engineers will test water flows and durability of the Onkalo tunnel, and from 2014 bedrock will be blasted away to open the site in 2020, making it the world's first permanent nuclear waste repository. Neighboring Sweden also expects to start constructing its repository in 2017, and SKB has been tasked with this project, for which it has already submitted a license application. Other countries in Europe, including the UK and Germany, are lagging

behind as they search for suitable sites for their geological repositories. The United States have not successfully sited a repository for UNF and HLW or made long-term fuel cycle decision yet^[22]. The Blue Ribbon Commission panel provided recommendations for developing a safe long-term solution to managing UNF. The report was issued on January 26th 2012^[23].

4 Conclusion

As discussed, recovering minor actinides from UNF is key not only to decrease the long-term radiotoxicity and heat-loading of the repository but also to manage the size of the repository. Countries are investigating minor actinides separation and integrated repository science as well. But it is important to keep in mind that developing waste management policies that incorporates either advanced recycling technologies or direct disposal of UNF requires public consultancy with the participation of all stakeholders involved in the decision making process. Each country is different and has to consider its own political constraints but whatever the option is, it should provide a safe and secure environment for the generations to come.

Nomenclature

ADS	Accelerator-Driven-System
COEX TM	CO-Extraction
DIAMEX	DIAMide Extraction
EXAm	EXtraction of Americium
FP	Fission Products
GANEX	Group ActiNide Extraction
HLW	High Level Waste
LUCA	Lanthanide Und Curium Americium
MA	Minor Actinides
MOX	Mixed Oxide fuel
SANEX	Selective ActiNide Extraction
TALSPEAK	Trivalent Actinide – Lanthanide Separation by Phosphorous reagent Extraction from Aqueous Komplexes
TRU	TRansUranic elements
TRUEX	TRansUranics Extraction
UNF	Used Nuclear Fuel

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