

# Radioactive Waste Management Approaches For Developed Countries

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### RADIOACTIVE WASTE MANAGEMENT APPROACHES FOR DEVELOPED COUNTRIES

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#### ABSTRACT

Nuclear power has demonstrated over the last 30 years its capacity to produce base-load electricity at a low, predictable and stable cost due to the very low economic dependence on the price of uranium. However the management of used nuclear fuel remains the “Achilles’ Heel” of this energy source since the storage of used nuclear fuel (UNF) is increasing as evidenced by 2,000 tons of UNF produced each year by the 104 US nuclear reactor units which equates to a total of 150,000 spent fuel assemblies stored in pools for at least 5 years followed by dry cask storage for about 62,000 spent fuel assemblies. Two options adopted by several countries will be presented. 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 used nuclear fuel into a geologic formation. One has to remember that only 30% of the worldwide used nuclear fuel are currently recycled, the larger part being stored (70% 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 volumetric 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 are pursued worldwide and their approaches will be highlighted.

#### INTRODUCTION

A thoroughly planned nuclear waste management program is key for the deployment and expansion of nuclear energy. As of today, two strategies are offered to countries using nuclear energy. Some countries such as the US, Sweden, Finland, and Hungary have opted for a once-through fuel cycle with the geological disposal of used nuclear fuel. Other countries such as France, Japan, Russia, UK, and India have decided to recycle UNF (Figure 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 a closed fuel cycle, which may integrate a modified PUREX process, envisions the recycling of minor actinides (MA) and fission products (FP) to be burned in a fast reactor or used as targets for transmutation in accelerator-driven systems (ADS). 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 different options available for nuclear waste management which include either the disposal of UNF or the various R&D recycling activities undertaken by different countries for improving the back end of the nuclear fuel cycle.

operations of Transuranic (TRU) waste in March 1999 and has remained an operating deep geologic disposal facility ever since that date. As the DOE Carlsbad Field Office (CBFO) gained operationally knowledge and expertise with the successful operations of WIPP, it has successfully provided numerous requests of modifications/updates to the different stakeholders, which is a testament to CBFO's ability

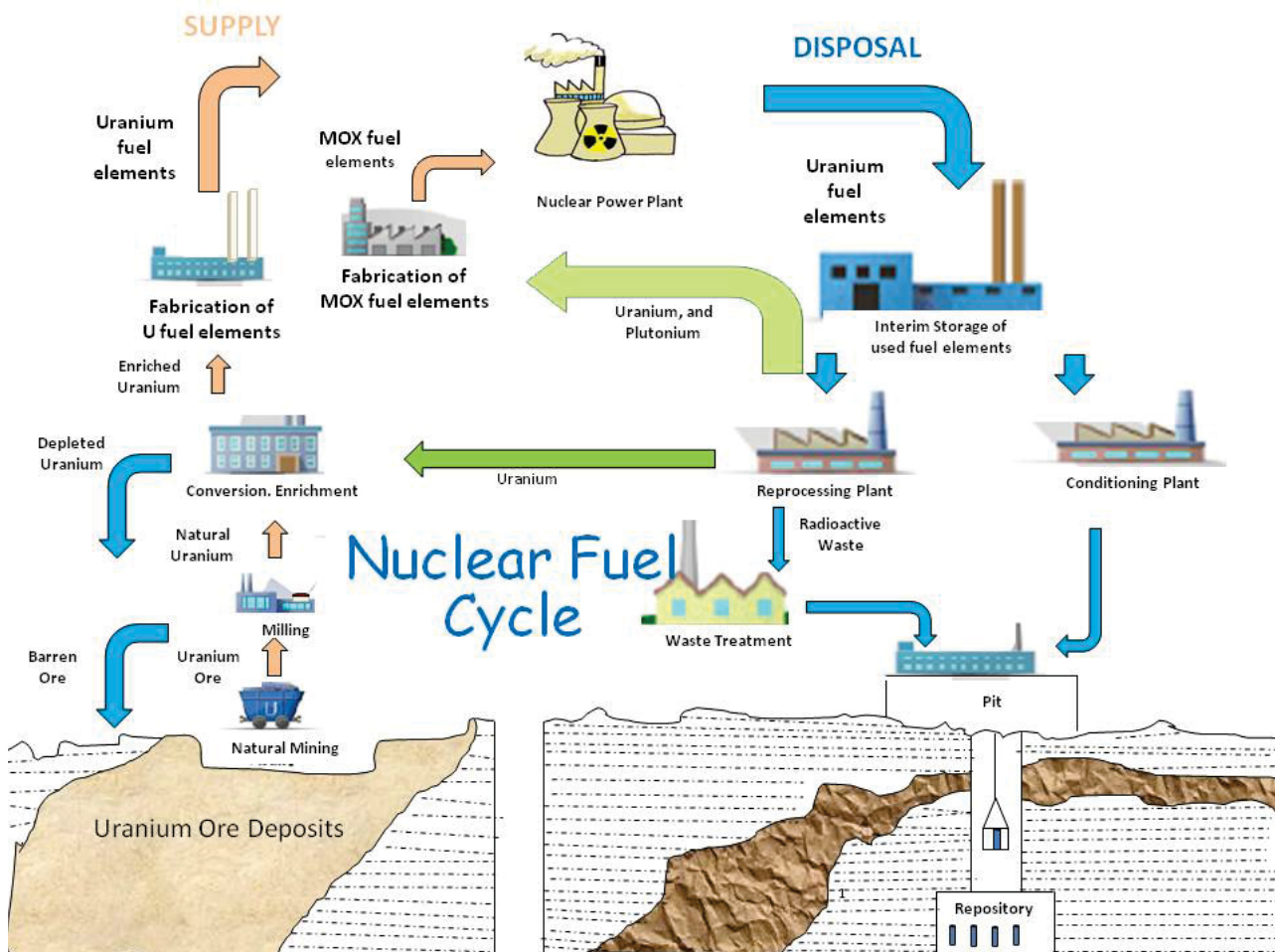


Figure 1: The nuclear fuel cycle

## DIRECT DISPOSAL OF USED NUCLEAR FUEL AND NUCLEAR WASTE

The United States has not successfully sited a repository for UNF and HLW or made any long-term fuel cycle decisions yet [1]. The Blue Ribbon Commission panel provided recommendations for developing a safe long-term solution to managing UNF. The report was issued on January 26, 2012 [2]. On the other hand, the Waste Isolation Pilot Plant (WIPP) is the world's first permitted deep geologic repository for the permanent disposal of radioactive waste. WIPP, located in southern New Mexico, began disposal

to leverage its operational expertise to provide effective enhancements to meet the nation's legacy waste challenges and improve upon operational and programmatic efficiencies in accelerating legacy waste cleanup while ensuring protection of the worker, public, and environment. 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 has reached a milestone with the application in December 2012 by Posiva, the waste management specialist jointly owned by Finnish nuclear utilities Fortum and TVO,

to construct a final repository and waste encapsulation plant at Olkiluoto [3]. The Finnish repository is located at 450 meters 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 for UNF. Sweden also expects to start constructing its repository in 2017, and SKB, the waste management organization responsible for implementing geological disposal in Sweden, has been tasked with this project, for which it has already submitted a license application. Recently, Hungary opened its radioactive waste repository 250 meters below ground at Bataapati. The first phase of the project will allow the disposal of 40,000 m<sup>3</sup> of low and intermediate radioactive waste, while in a second phase, the repository will be extended to allow the disposal of more active waste (HLW and UNF) [4]. Other countries in Europe, including the UK and Germany, are lagging behind as they search for suitable sites for their geological repositories.

## USED NUCLEAR FUEL RECYCLING

### The PUREX and COEX<sup>TM</sup> Processes

The PUREX process, based on liquid-liquid extraction of hexavalent uranium and tetravalent plutonium by a mixture of tri-*n*-butylphosphate (TBP) and dodecane, has been the basis for commercial UNF recycling worldwide for over 30 years [5, 6]. This 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 UO<sub>2</sub> and mixed oxide fuels from recycled material. To reduce the risk of plutonium diversion from the PUREX process, the COEX<sup>TM</sup> process is being developed by AREVA [7]. 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 COEX<sup>TM</sup> process aims at further enhancing proliferation resistance [6].

### Co-Extraction of Minor Actinides and Lanthanides

For future advanced nuclear systems, minor actinides (MA) are considered more as a resource to be recycled and transmuted than to be disposed of 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/COEX<sup>TM</sup> 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. The following processes corresponding to this category are the DIAMide EXtraction (DIAMEX)/ Selective ActiNide EXtraction (SANEX) processes developed in France [8], TRUEX/TALSPEAK processes developed in the United States [9-11], or the Advanced-ORIENT cycle project developed in Japan [12].

(2) Other simplified processes are aiming to separate a group of actinides using an integrated fuel cycle (with 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).

In France, the DIAMEX/SANEX processes (Figure 2), studied at the Commissariat à l'Energie Atomique (CEA) Marcoule, aims at recovering MA selectively from PUREX raffinate by solvent extraction.

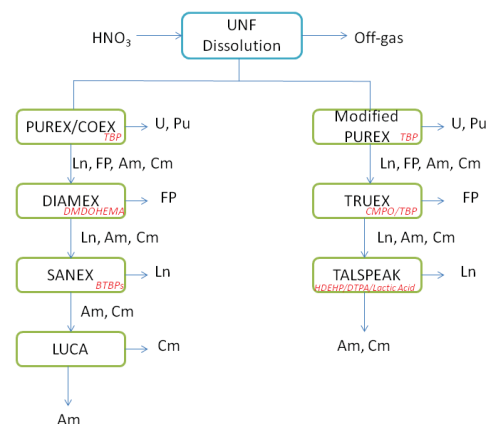
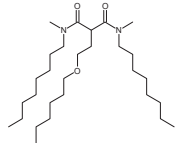
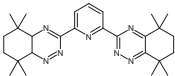
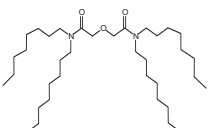


Figure 2: Advanced separation processes involving a co-extraction followed by separation of minor actinides

One of the reference molecules of the DIAMEX process is DMDOHEMA (Table 1). 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 HNO<sub>3</sub> while antagonism was noticed at lower acidity with the formation of an adduct “HDEHP-DMDOHEMA” which decreases the effective HDEHP concentration [13]. The diamide derivatives possess a good extractability and respond to the CHON concept (completely incinerable and produces no radioactive solid waste by combustion).



Table 1: Example of Ligands Used in UNF R&D Recycling

Ligand Structure	Abbreviation	Process
	<b>DMDOHEMA</b> N,N'-DiMethyl-N,N'- DiOctyl-Hexyl-Ethoxy- Malonamide	DIAMEX
	<b>CyMe4-BTBP</b> 2,6-Bis(5,5,8,8- tetrahydro-1,2,4- benzotriazin-3-yl)pyridine	SANEX
	<b>TODGA</b> N,N,N',N'- Tetraoctyldiglycolamide	ARTIST

Americium and curium are then separated from the lanthanides in a following SANEX process (in which the reference molecule is CyMe4-BTBP (Table 1). 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 [14] which showed interesting properties such as separation of trivalent actinides from trivalent lanthanides with a separation factor as high as  $SF_{Am/Eu} = 140$ . 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 (Figure 2). It uses a mixture of bis(chlorophenyl)dithiophosphinic acid ((CIPh)<sub>2</sub>PSSH) and tris(2-ethylhexyl)phosphate (TEHP) as the extractant. More than 99.5% of Cm(III) was directed to the raffinate[15]. However, considerable degradation (by oxidation) of the (CIPh)<sub>2</sub>PSSH has been observed at higher acidity (>0.5M HNO<sub>3</sub>). 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 allow the recovery of Am(III) alone [16]. In the United States, the TRUEX solvent extraction process (Figure 2) 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. The TRUEX process solvent contains

octylphenyl-N,N-diisobutylcarbamoylmethylphosphine oxide (CMPO) as the ligand while the diluents is composed of TBP and dodecane. Recent investigations [17] 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 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 [10, 11]. TALSPEAK process could be applied to a TRUEX raffinate (after adjustment) to separate TRU elements from the lanthanides (Figure 2). In Japan, the ARTIST process uses advanced amide-based extractants such as the tridentate ligand TODGA (Table 1) to separate actinides from fission products [18]. 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.

### Simplified Processes for Minor Actinides Separation

As depicted in Figure 3, new advanced recycling technologies aimed at simplifying processes with one single extraction step following a PUREX-based process.

In Europe, alternative concepts to the PUREX-DIAMEX-SANEX processes are being developed with either a one-cycle SANEX process or a new GANEX process (Figure 3). The one-cycle SANEX (innovative SANEX or *i*-SANEX) separates MA directly from a PUREX raffinate by combining TODGA and SO<sub>3</sub>-Ph-BTP molecules. 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 SO<sub>3</sub>-Ph-BTP [19]. The new GANEX 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 (Figure 3). The solvent consists of a mixture of TODGA and DMDOHEMA in Exxsol D80. The undesired co-extraction of some fission products is realized by adding 1,2-cyclohexanediaminetetraacetic acid (CDTA). In the United States, Lumetta et al. [20] are developing a single process called TRUSPEAK process 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

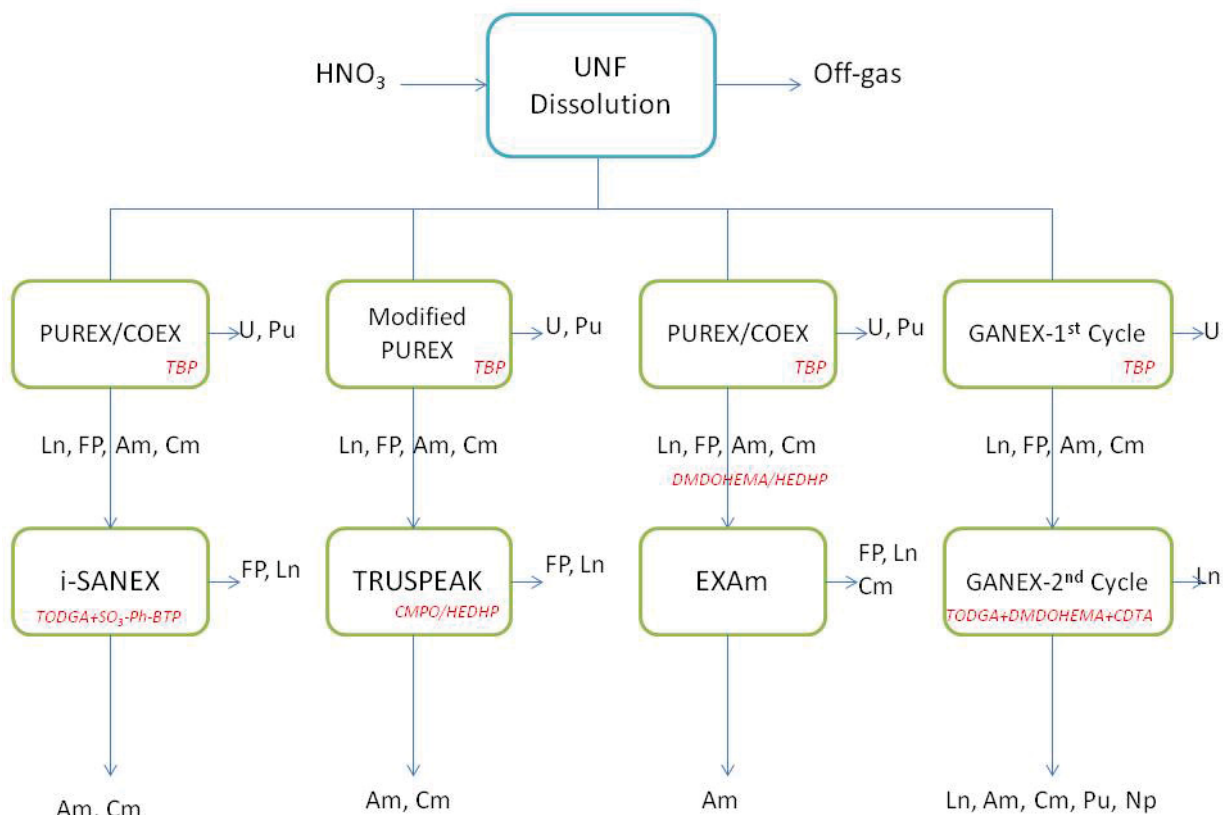


Figure 3: Simplified processes for minor actinides recovery

TALSPEAK solvent HDEHP in dodecane. In doing this, it is envisioned that the CMPO chemistry would dominate under conditions of high acidity ( $\geq 1$  M  $\text{HNO}_3$ ), 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  $\sim 3$  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 (TRU) versus the lanthanides causes the TRU to be selectively stripped into the aqueous phase, thereby separating them from the lanthanides.

### Recycling of actinides and fission products

The UREX process was the first solvent extraction operation in a group of separation processes generically called UREX + that has been developed in the US as a baseline set of separation operations to partition the components of UNF to enable recycle of the actinides and more efficient disposal of the fission products [21]. Recently there has been some interest in the application of simple hydrophilic hydroxamic ligands such as acetohydroxamic acid (AHA) for the stripping of tetravalent actinides in UREX process flowsheet. This

approach is based on the high coordinating ability of hydroxamic acids with tetravalent actinides,  $\text{Np}^{4+}$  and  $\text{Pu}^{4+}$  compared with hexavalent uranium,  $\text{UO}_2^{2+}$ . Thus, the use of AHA offers a route for controlling neptunium and plutonium in the UREX process by complexant-based stripping of  $\text{Np(IV)}$  and  $\text{Pu(IV)}$  from TBP solvent phase, while  $\text{U(VI)}$  ions are not affected by AHA and remain solvated in the TBP phase. The UREX process selectively extracts uranyl nitrate ( $\text{UO}_2(\text{NO}_3)_2$ ) and pertechnetate ( $\text{TcO}_4^-$ ) from dissolving used fuel in nitric acid into a mixture of TBP and hydrocarbon diluent as  $\text{UO}_2(\text{NO}_3)(\text{TcO}_4) \cdot 2\text{TBP}$  species, and back extracts these species into diluted nitric acid. As depicted in Figure 4, the following steps of the UREX process were composed of different processes allowing the selective extraction of fission products such as Sr-90 and Cs-137, as well as MA. The R&D of the UREX+ process has been abandoned because of the complexity of the different waste streams. Japan is developing the Advanced-Orient Cycle project [12] which proposed the separation, transmutation of FP and MA and utilization of rare earth metals such as the light platinum group metals (Ru, Rh, and Pd) for fast breeder reactor fuel cycle [12]. It also includes the separation of Tc-99.

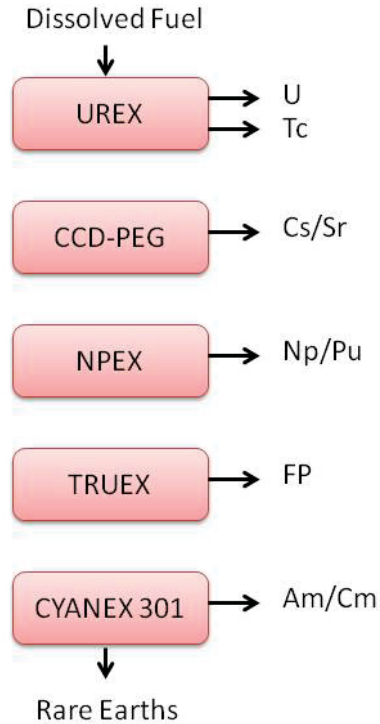


Figure 4: The UREX+ Process

The laboratory-scale experiments are based on ion exchange resins and one catalytic electrolytic extraction step. The feasibility of the process is being evaluated and results can be found in [23].

## CONCLUSION

According to Carre and Delbecq [22], multiple recycling of MA in fast neutron reactors, which implies their extraction from a PUREX-based raffinate, would result in an accumulated amount of TRU elements of 100 metric tons as opposed to a once through fuel cycle with 1,680 metric tons of accumulated TRU 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 cycle with the fabrication of a MOX fuel (U-Pu)O<sub>2</sub> 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. Recovering minor actinides from UNF is key not only to decreasing 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.

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