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A Review of Separation Processes Proposed for Advanced Fuel Cycles Based on Technology Readiness Level Assessments

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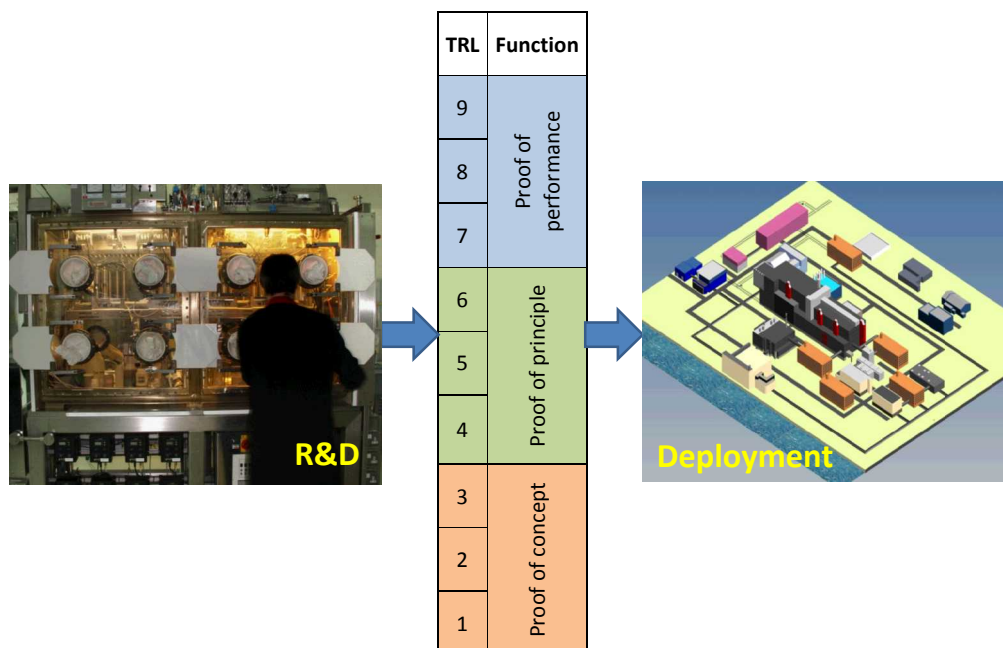
- Results of an international study
- Advanced reprocessing and minor actinide partitioning technologies
- Review of progress in R&D on separation processes for advanced nuclear fuel cycles
- Technology Readiness Level assessment of separation processes
- Status of head-end, aqueous and pyro-chemical processes reviewed

Abstract

The results of an international review of separation processes for spent nuclear fuel (SNF) recycling in future closed fuel cycles are reported. This study was made by the Expert Group on Fuel Recycling Chemistry (EGFRC) organised by the Nuclear Energy Agency (NEA) of the Organisation for Economic Co-operation and Development (OECD). A unique feature of this study was that processes were classified according to a hierarchy of separations aimed at different elements within spent fuel (uranium; uranium-plutonium co-recovery; minor actinides; high heat generating radionuclides) and also the Head-end processes, used to prepare the SNF for chemical separation, were included. Separation processes covered both wet (hydrometallurgical) and dry (pyro-chemical) processes.

A bespoke methodology suitable for assessing the technological maturities of separation processes for SNF recycling was then designed based on the well-established Technology Readiness Level (TRL) scale but adapted for spent nuclear fuel separations. The innovative feature of this was the use of a matrix approach that enabled information on both the scale of testing and materials used in testing to be combined in the evaluation of the TRL. TRLs for the Head-end, hydrometallurgical and pyrochemical processes that had been reviewed by the study have been evaluated. It is clear that, whilst more R&D is needed in most cases to raise TRLs, suitable options exist to enable the recovery of all actinide elements plus the HHRs by wet or dry processes, even up to the extraordinarily difficult isolation of americium alone. The technology readiness levels (TRL) for most processes fall in the 'proof of principle' range with TRLs between 4 and 6, although this is an evaluation at the system level and some gaps in the individual process steps may still exist that strictly would reduce the overall TRL. That is, flowsheets have been proposed for processes and tested with active materials under increasingly relevant conditions, through to initial hot tests with small quantities of SNF. The results of this work were shown to be comparable to a parallel European study that evaluated TRLs for minor actinide partitioning processes.

Graphical Abstract



List of Acronyms

Acronym	Definition	Comment
ADS	Accelerator driven system	Transmutation system based on use of an accelerator rather than critical reactor
ALSEP	Actinide-lanthanide separation	US separation process for minor actinides
AMP	Ammonium molybdophosphate	Inorganic ion exchanger
ASGARD	Advanced fuels for Generation IV reactors: reprocessing and dissolution	EU Framework Programme 7 project
ATF	Accident tolerant fuels	New fuel types under development
BTBP	Bis-triazinyl-bipyridines	Ligand for actinide/lanthanide separation
BTP	Bis-triazinyl-pyridines	Ligand for actinide/lanthanide separation
CARBEX	CARBEX: Carbonate extraction	Russian process
CCD-PEG	Chlorinated cobalt dicarbollide-polyethylene glycol	US process for caesium and strontium separation
CEA	Commissariat à l'Énergie Atomique et aux énergies alternatives	French nuclear R&D organisation
CHON	Carbon, hydrogen, oxygen, nitrogen	Principle for decomposable organic agents added to processes
CNRS	Centre National pour la Recherche Scientifique	French R&D organisation
CoDCon	Co-decontamination	US process for co-processing U and Pu
COEX™	Co-extraction of actinides	French process for co-processing U and Pu
CST	Crystalline sodium titanosilicate	Ion exchange materials
DBP	Dibutyl phosphate	TBP degradation product
DEHIBA	N,N-di-(ethyl-2-hexyl)isobutyramide	Organic ligand used in GANEX process
DF	Decontamination factor	Ratio of an impurity in a feed compared with a product
DGA	Diglycolamide	Class of organic ligands
DIAMEX	Diamide extraction	French process for MA separations
EBR	Experimental Breeder Reactor	US fast reactor
EGFRC	Expert Group on Fuel Recycling Chemistry	A group of the NEA
EU	European Union	
EXAm	Extraction of americium	French process for MA separations
FBR	Fast breeder reactor	Fast neutron reactor set up to breed plutonium
FLUOREX	Fluoride volatility solvent extraction	Japanese hybrid process of fluoride volatility and solvent extraction
FP	Fission product	Isotopes produced by nuclear fission
FPEX	Fission product extraction	US process for caesium and strontium separation
FR	Fast reactor	Generic term for fast neutron reactors
GANEX	Group actinide extraction	European processes that co-extract TRU actinides as a group

GDF	Geological disposal facility	Sub-surface radioactive waste repository
GIF	Generation IV International Forum	International forum for next generation reactor development
GNEP	Global Nuclear Energy Partnership	US initiative to develop advanced fuel cycles
HA	Highly active	Containing HHR
HAF	Highly active feed	Dissolved fuel solution that is the feed to the separations stage of a reprocessing plant
HAN	Hydroxylamine nitrate	Chemical reducing agent
HAR	Highly active raffinate	Waste stream from reprocessing containing fission products
HHR	High heat radionuclide	Caesium and strontium
HLW	High level waste	Heat generating radioactive wastes
IAEA	International Atomic Energy Agency	International organisation to promote the safe, secure and peaceful use of nuclear technologies
IFR	Integral fast reactor	US fast reactor and fuel cycle concept
IMF	Inert matrix fuel	Nuclear fuel containing a low activation matrix as carrier for the fissile material
INL	Idaho National Laboratory	US nuclear lab
KI	Kurchatov Institute	Russian national research centre
LWR	Light water reactor	Generic description for thermal reactors with H ₂ O coolant
MA	Minor actinides	neptunium, americium and curium
MDD	Modified direct denitration	A route for (U,Pu) co-conversion
MELOX		French plant for MOX fuel production
MOSART	Molten salt actinide recycler & transmuter	Russian (Kurchatov Insititute) concept for molten salt reactor
MOX	Mixed oxide	Usually referring to (U,Pu)O ₂ fuels
MSFR	Molten salt fast reactor	French CNRS concept for molten salt reactor
MSR	Molten salt reactor	Any reactor that uses molten salt fluorides / chlorides as a fuel carrier or primary coolant
MSRE	Molten salt reactor experiment	8 MWt experimental molten salt reactor at Oak Ridge National Laboratory (USA)
NEA	Nuclear Energy Agency	An agency of the OECD
NEXT	New extraction system for TRU recovery	Japanese process for reprocessing SNF
NNL	National Nuclear Laboratory	UK nuclear R&D organisation
NSC	Nuclear Science Committee	A committee of the NEA
NUEX™	New uranium extraction process	Proposed process for reprocessing SNF
OECD	Organisation for Economic Co-operation and Development	Parent organisation of the NEA
OK	Odourless kerosene	Industrial diluent
OMEGA	Options making extra gains from actinides and fission products	Japanese initiative in P&T
ORNL	Oak Ridge National Laboratory	US laboratory

P&T	Partitioning and transmutation	Fuel cycles that involve separation and burning of isotopes from radioactive wastes
PAN	Polyacrylonitrile	Polymerised organic resin
PRIDE	Pyro-process integrated inactive demonstration	Korean R&D facility for pyro-processing development
PUREX	Plutonium and Uranium Refining by EXtraction	Process used in commercial scale reprocessing
PWR	Pressurised water reactor	Type of light water reactor
R&D	Research and development	
REPA	Reprocessing-Partitioning	Russian process for reprocessing SNF
RIAR	Russian Institute of Atomic Reactors	Russian nuclear institute at Dimitrovgrad
SANEX	Selective actinide extraction	European processes that extract trivalent MA
SESAME	Selective extraction of americium by electrochemical methods	French process for americium/curium separation
SMR	Small modular reactors	Reactors less than 300 MW of modular design and manufacturing
SNF	Spent nuclear fuel	Irradiated fuel, also referred to as used nuclear fuel (UNF)
SRL	Scientific readiness level	Scale to assess state of scientific understanding of a process or technology
SSA	Specific surface area	Physical property usually measured in units of m ² /g
SX	Solvent extraction	Chemical separation method also referred to as liquid-liquid extraction
TALSPEAK	Trivalent actinide-lanthanide separations by phosphorus reagent extraction from aqueous complexes	US process for actinide/lanthanide separation
TBP	Tributylphosphate	Extractant used in the PUREX process
TddDGA	N,N,N',N'-tetradodecyl- diglycolamide	Type of DGA ligand
TODGA	N,N,N',N'-tetraoctyl-3-oxapentane-1,5-diamide	Commonly used type of DGA ligand
TRISO	Tristructural-isotropic	A type of micro fuel particle composed of a fuel kernel and outer layers
TRL	Technology readiness level	Scale for assessing technical maturity
TRU	Transuranium	Elements following U in the Periodic Table
TRUEX	Transuranium extraction	US process for MA separation
UK	United Kingdom	
UOX	Uranium oxide	Nuclear fuel material
UREX	Uranium extraction	US process for reprocessing SNF
US	United States	
VOLOX	Voloxidation	Process for oxidation of SNF and volatilisation of radionuclides
WPFC	Working Party on Scientific Issues of the Fuel Cycle	A working party of the NEA

1 Introduction

Advanced nuclear systems will generate spent (or used) nuclear fuel (SNF) that must be managed either in open or closed fuel cycles. Closing the fuel cycle to promote sustainability requires capabilities to reprocess and multi-recycle major actinides (U, Pu) into fuels for advanced reactors such as Generation IV fast reactors [1, 2]. Additionally, the separation of minor actinides (MA: Np, Am, Cm) and subsequent conversion into short-lived nuclides may be required to minimise the radioactive waste burden on geological disposal facilities (GDF) and/or improve the long-term characteristics of the wastes in the GDF [3, 4]. This is the so-called partitioning and transmutation (P&T) fuel cycle and can theoretically reduce the long-term risks associated with the use of nuclear energy [5, 6]. International R&D programmes are addressing these challenges and many advanced separation processes for major and minor actinide recovery have to date been proposed [7-11].

Within the Nuclear Energy Agency (NEA) of the OECD (Organisation for Economic Co-operation and Development) these developments are being monitored by the Expert Group on Fuel Recycling Chemistry (EGFRC) under the mandate of the Working Party on Scientific Issues of the Fuel Cycle (WPFC) which is a branch of the Nuclear Science Committee (NSC) of the NEA. The EGFRC is formed to share progress in spent fuel recycling and to make technical evaluations of the options. In this context, the EGFRC has recently completed a detailed technical assessment of the state of the art in separation processes, relevant to closed nuclear fuel cycles, including an assessment of their technical maturity. The study covered the different separation processes proposed by NEA member states (France, Japan, Korea, Russia, UK and US, as well as the EU). The outcome was a major state of the art review [12] in which the technical advantages and disadvantages of the separation processes were duly considered. Since the EGFRC study began, other reviews have been published that complemented the present study [13, 14]. The differentiating aspect of this review, however, is that it was organised by the type of processes, considering their broad roles, and also that it included head-end processes, hydrometallurgy, pyro-electrochemistry and other pyro-processing methods for different types of nuclear fuels [12]. However, the main objective of the study was to evaluate the R&D status and to understand the basis on which decisions on technology down-selection could be made. Therefore, the criteria that impact the selection of fuel recycle processes in different future closed fuel cycles, including key issues of safeguards and non-proliferation, were considered before making an assessment of technical maturity using 'technology readiness levels' (TRL) [15]. This paper reports the novel aspects of the study, specifically focusing on the methodology adopted for technology readiness assessments, coupled with the outcomes of our assessment and critical comparisons with other published TRL assessments. It is to be noted that this paper focuses on the technical readiness of the chemical flowsheets rather than the engineering technology. In moving forward, however, towards implementation it is recognised that both chemistry and engineering are inter-dependent and so must be progressed in an integrated manner.

2 Progress of separation technology and current achievement

This section outlines the separation processes that were reviewed during the NEA study. The processes are organised according to their role and aim. That is, an initial division between head-end processes, hydrometallurgy and pyro-processing followed by further sub-divisions according to the separation process function (uranium only recovery; U-Pu co-recovery; minor actinide (MA) recovery *etc.*). The processes considered are briefly described for each category, focusing on specific features and commonalities that can affect the later TRL assessment. For in-depth reviews of the separation process chemistry, readers are directed to reference [12] as well as other state of the art reviews [13, 14].

2.1 Head-end processes

2.1.1 Introduction

Head-end covers the range of processes needed to prepare SNF for the separation stages [16, 17]. The head-end of an advanced reprocessing plant encompasses the key activities of disassembly, capture of off-gases arising from volatile species in spent fuels and dissolution of the SNF prior to separation and recovery of specific components. Depending on the type of separation process to be used and other factors such as regulatory requirements, shearing of fuel, fission product volatilisation and recovery of zirconium from Zircaloy® used fuel cladding may also be part of the Head-end plant area. Off-gas treatment systems are a major component of the Head-end process in order to deal with radioactive gases and process chemical vapours, ensuring no unacceptable releases of radioactive or hazardous gases to the environment. Schematics of an advanced Head-end process for aqueous and pyro-chemical processing of oxide fuels, incorporating these various options, are shown in Fig. 1.

2.1.2 Processes reviewed

The study initially reviewed current technologies used in the head-end plant of industrial scale reprocessing plants before considering potential additional stages or alternative processes such as Korean processes for disassembly and decladding [18, 19]; Russian processes for decladding and fuel oxidation [20]; an American process for recovery of zirconium from fuel cladding [21, 22] and the removal of volatile fission products with off gas treatment (tritium, iodine, krypton, carbon-14, semi-volatiles and particulates [23, 24]). Based on this study, the latter area was highlighted as a topic for further evaluation and is the subject of a new, in-depth study now being made by the EGFRC. Advanced aqueous dissolution processes and head-end processes for molten salt based separations were reviewed as key stages in the preparation of the feedstock to the separations processes.

2.1.3 Head-end status

Current industrial-scale operations based on direct dissolution are implemented whilst R&D programmes have made progress in demonstrating voloxidation (VOLOX) type processes and associated off-gas trapping methods – dry processes to release and capture volatile species leading to improved control of emissions. Also, for the pulverisation of oxide fuels to powders suitable for dissolution in nitric acid or molten salts. Molten salt processes for oxide fuels will require an electro-reduction stage to generate actinide metal for electro-refining although alkali and alkaline earth metals are dissolved into the salt [25]. There are also potential benefits if

more of the spent fuel components, not just actinides, can be recycled rather than disposed as wastes, *e.g.* zirconium from Zircaloy fuel cladding and selected fission product elements [22]. The EGFR recognised also that the dry oxidation processes may have an important role to play in oxidising non-oxide fuels (*e.g.* metals, carbides, nitrides) to oxides prior to dissolution in acidic solutions for reprocessing [26, 27]. However, this study did not examine this aspect of an advanced Head-end process in detail. The recent EU-funded Framework Programme 7 project “ASGARD” did, however, address these issues and results from this project are now becoming available [28, 29]. Another key issue in aqueous processes is the need for dissolution of high plutonium content fuels using electrochemical catalysed processes, including how to manage plutonium-rich residues [30]. The overall Head-end process (comprising mechanical and dry or wet chemical stages) is of crucial importance in converting a range of possible advanced fuels into standardised and convenient forms for subsequent aqueous [16] or pyro-processing separations [25, 31]. Experience from commercial scale reprocessing using the PUREX process is that plutonium losses from Head-end also tend to dominate the overall reprocessing plant losses and this can affect the efficiency of the closed fuel cycle concepts. Whilst this review focused on the development of chemical processes, improvements to head end plant will also require addressing large scale mechanical and process engineering challenges, some of which will affect the performance of the chemical processes, such as: decladding; dissolver design; feed clarification; transport of particulates; corrosion; accountancy and criticality control. These issues are discussed in greater detail in ref. [16].

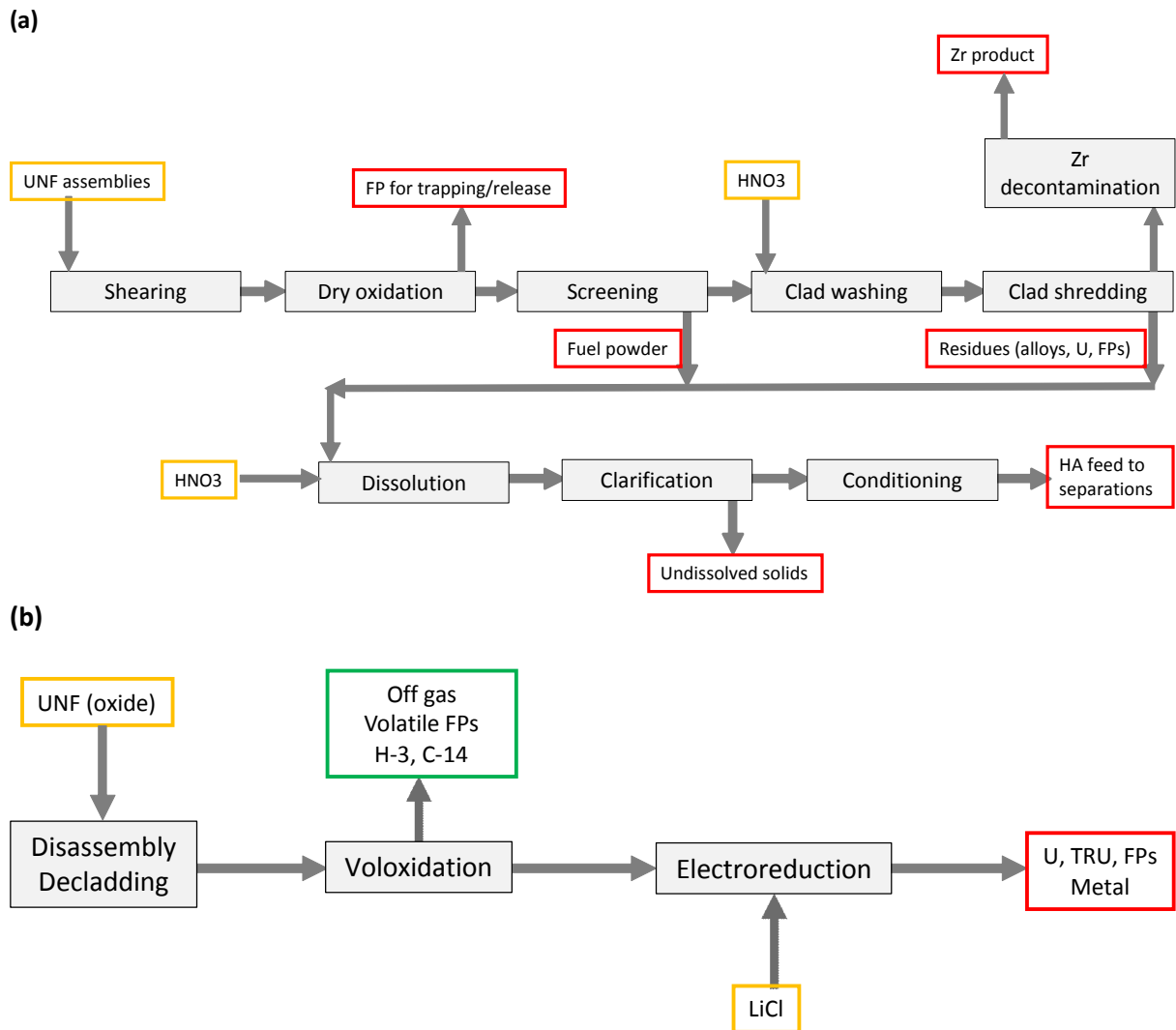


Fig. 1: Advanced head end concepts for (a) aqueous and (b) pyro- processes [12, 19]

2.2 Hydrometallurgy

2.2.1 Introduction

Hydrometallurgical separation processes were reviewed according to their function by defining a hierarchy of separations of increasing complexity and specialisation. Most processes are based on solvent extraction but other aqueous processes were included where the EGFR highlighted an interest at either the national strategic or industrial level. The hierarchy followed is:

- ⇒ Uranium only recovery
- ⇒ Major actinide co-recovery (U,Pu and U,Pu,Np since neptunium is often co-recovered with uranium and plutonium due to similarities in process chemistry)

- ⇒ TRU actinide separation processes (that use non-PUREX systems) sub-divided again into two cases:
 - MA recovery from highly active raffinate (HAR)
 - Group separations
- ⇒ Caesium and strontium separations
- ⇒ Curium separations

The different strategies that form this hierarchy are illustrated schematically in Fig. 2 and these categories of separations are described briefly below, again focusing on the chemical flowsheets rather than engineering. Although the extraction technologies used for tests are noted due to their influence on the design and performance of the flowsheets, specifically with respect to how kinetics are affected by residence times in the different types of contactors.

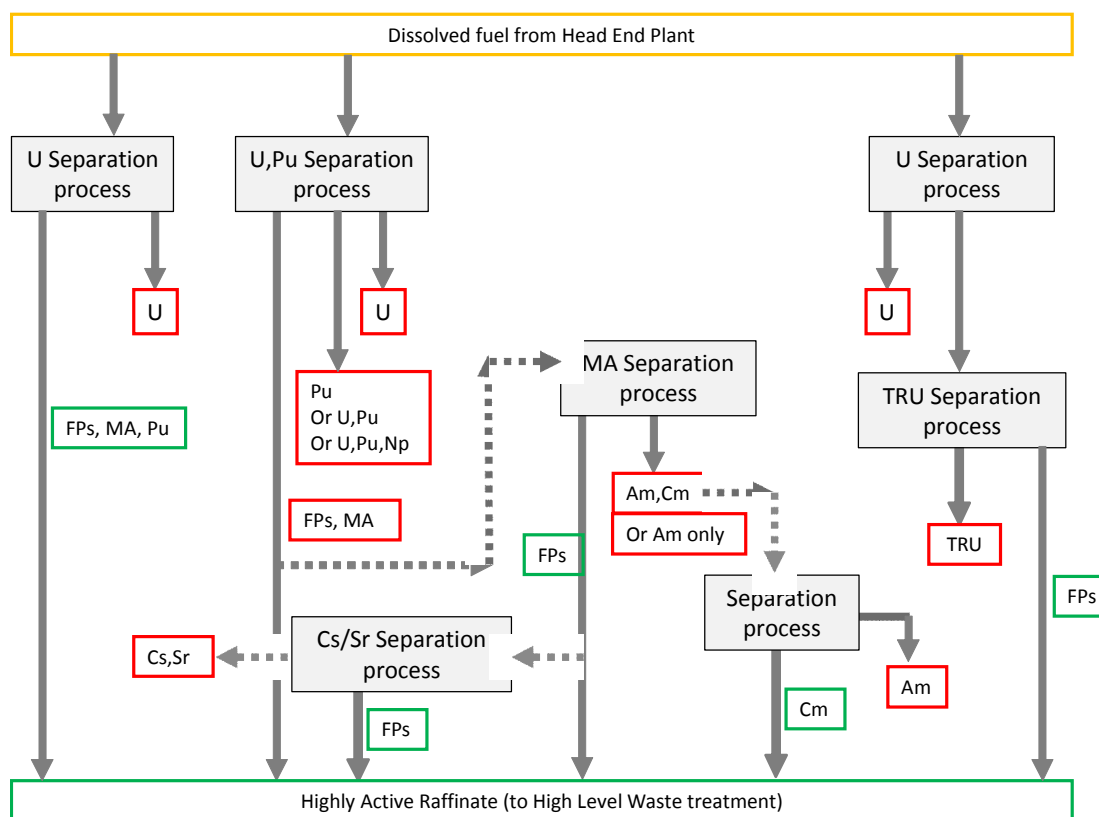


Fig. 2: Separation strategies considered in the EGFR study for aqueous recycling processes

2.2.2. Uranium Recovery

In irradiated thermal reactor fuels, ~95 wt.% of the heavy metal content (65 % of the mass of PWR fuel assemblies) is still uranium and so separation of uranium is a primary factor in reducing the volume of spent fuel that would otherwise have to be disposed of as high level waste. Reprocessed uranium can be reused as fuel and recycled into reactors or stored separately from the residual HLW. Existing separation processes have been adapted and new processes using different extractants developed for either full or partial uranium-only recoveries, usually as a first reprocessing step, in order to optimise or simplify subsequent stages.

There are two main processes in this context: (i) solvent extraction exemplified by the selective extraction of uranium in the first cycle of the GANEX process developed by CEA of France (using N,N-di(2-ethylhexyl)isobutyramide (DEHIBA) as the extractant) [32] and the US UREX process, based on a conventional tributyl phosphate (TBP) extraction cycle [33]; (ii) crystallisation for partial recovery of uranium. This is exemplified by the Japanese NEXT process and Fig. 3 illustrates how the uranium crystallisation process is incorporated into the overall schematic flow diagram in the NEXT process [7, 34].

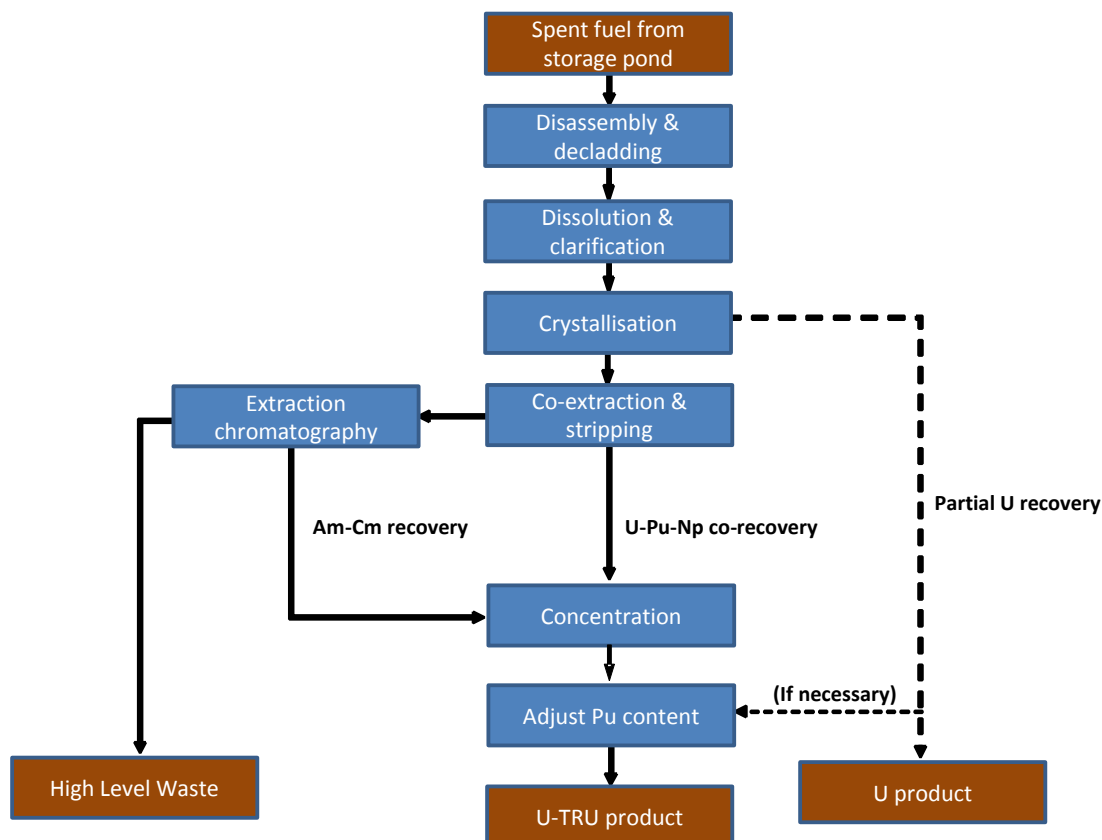


Fig. 3: Japanese NEXT process incorporating a uranium crystallisation stage [12]

2.2.3 Co-recovery of (U, Pu, Np)

Industrial scale nuclear fuel reprocessing plants produce high purity separate uranium and plutonium products. The uranium and plutonium products are purified through several solvent extraction cycles, using a process based on tributyl phosphate (TBP) diluted in a paraffinic liquid as the organic phase and nitric acid as the aqueous phase. This is the well-known PUREX process that has been very successfully used for fuel reprocessing, with only minor variations, since the 1950s [35-37]. Currently operating reprocessing plants in France, the UK and Japan are similar in design and, as an example, Fig. 4 schematically illustrates the chemical separations flowsheet that is used in the Thorp (Sellafield, UK) reprocessing plant [38]. In this process, uranium and plutonium are co-extracted into the organic phase in the first highly active (HA) cycle together with part of the neptunium. The remaining neptunium is directed to the highly active raffinate (HAR). The extracted

neptunium is separated from the uranium product stream in the second uranium purification cycle and returned to the high level waste stream [39]. The MA elements (Am, Cm, etc.) also remain with the fission product waste stream, as these are trivalent ions that are inextractable in TBP. Ultimately, the HLW, containing fission products, neptunium and minor actinides, is vitrified producing a stable glass waste form suitable for geological disposal. The EGFR considered two options in this class of processes: (i) U-Pu co-processing, as in the COEX™ process developed in France [40, 41] or (ii) U-Pu-Np co-recovery in which the main reprocessing line is adjusted to fully route neptunium with the U-Pu product, *e.g.* see [42]. The main aims for co-processing are to reduce the perceived proliferation risks by avoiding pure separated plutonium, even within the process, and to simplify separations by producing a (U,Pu) nitrate stream that is suitable for co-conversion to a mixed oxide (MOX) product [43].

Processes reviewed were the COEX™ process (France) [40], NUEX™ process (USA/UK) [44], Advanced PUREX process (UK) [45], US Co-decontamination (CoDCon or UREX+ process) [40], NEXT process (Japan) [46] and a suite of Russian processes (including the Simplified PUREX and Reprocessing-Partitioning (REPA) processes as well as some other innovative aqueous separations) [47, 48]. As an example of this class of separations and to compare with current reprocessing, Fig. 4, Fig. 5 illustrates the US Co-Decontamination (CoDCon) process that aims to simplify conventional PUREX reprocessing flowsheets to a single cycle flowsheet whilst co-processing U-Pu or U-Pu-Np (this exemplifies directions for PUREX flowsheet development being taken in advanced reprocessing R&D programmes).

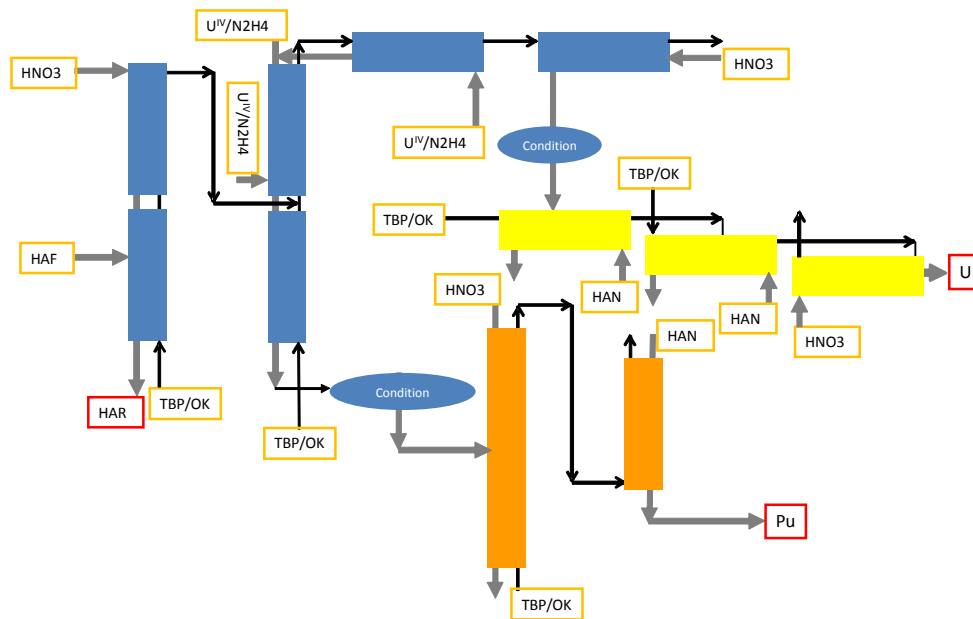


Fig. 4: Thorp solvent extraction flowsheet for commercial scale oxide fuel reprocessing (Sellafield, UK) [12]

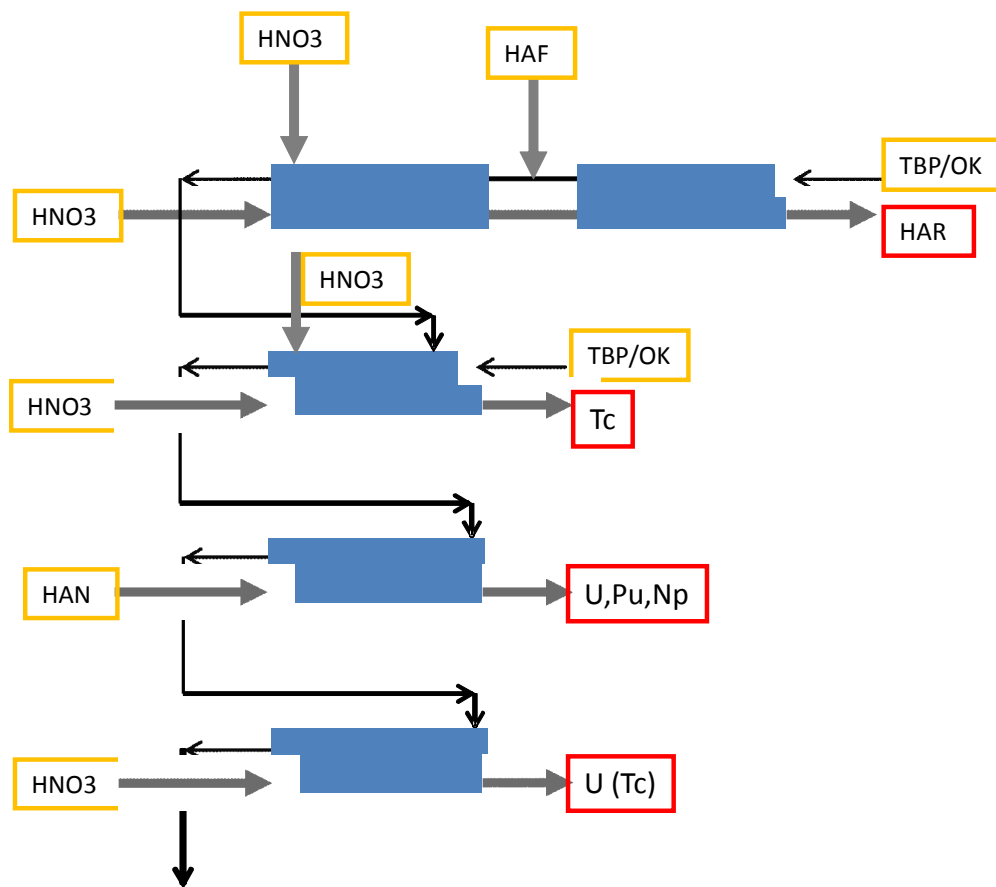


Fig. 5: US Co-Decontamination process – an example of an advanced PUREX flowsheet for future oxide fuel reprocessing [12]

2.2.4 Transuranic actinide separation processes based on new solvents

In the hierarchy of separations defined by the EGFR, the next stage was the co-recovery of mixtures of TRU actinides. By necessity this means moving away from TBP as the extractant towards new solvent systems that are capable of extracting trivalent minor actinides. This category includes the co-recovery of MA (Am, Cm) from PUREX HLW as well as grouped extractions of transuranic (TRU) actinides (co-recovery of Pu-MA, *i.e.* Pu-Np-Am-Cm).

Separation and recovery of trivalent minor actinides (Am, Cm) is a complex area with many solvent systems and variations on separation processes proposed by the international community [7-9, 49-52]. It is very difficult for any study or review to be truly comprehensive in compiling the options for new solvent systems but this study analysed a broad cross-section of options under consideration in leading NEA countries with recycle R&D programmes, including France, the EU, Japan, Russia and USA [12]. Some common themes were seen to emerge from the review such as a convergence towards amidic extractants, particularly the diglycolamide (DGA) ligands [53]. Also, the evolution towards simpler 1-cycle solvent extraction processes. Challenges remain in development of processes that are salt free in aqueous and organic phases (follow the 'CHON Principle', *i.e.* ligands comprising carbon, hydrogen, oxygen and nitrogen atoms only) and operate in the

acidic (>0.1 mol/L HNO_3) rather than high pH region for the difficult Ln/An separation stage. The latter is a major drawback of processes such as TALSPEAK which require stripping at high pH and within a rather narrow pH window and are thus difficult to operate at the engineering scale, despite recent improvements [54, 55]. The French DIAMEX-SANEX process is another classical example of the 2-cycle MA partitioning process [56, 57] whilst innovative or i-SANEX [8, 51, 58, 59] and 1 cycle (or 1c-) SANEX [8, 58, 60] are examples of the present directions towards simplified processes that are operable in the acidic region.

The other suites of processes reviewed were Grouped Actinide Extraction (GANEX) processes for full TRU co-recovery [8] and less conventional options such as a process using the zirconium salt of dibutyl phosphoric acid (DBP) developed in Russia [61]; a process based on extraction chromatography developed in Japan [62] and SNF reprocessing based on carbonates (specific focus on the Russian CARBEX process [63]).

In the context of new solvent extraction (SX) systems, the GANEX process is worthy of further explanation. The GANEX process is an alternative European concept for spent fuel recycling. The aim of the GANEX process is to co-process all the TRU actinides (Np–Cm) in one product stream [64]. It has been developed primarily for the homogeneous recycling of actinides in Generation IV fast reactor fuel cycles compared to the coupling of PUREX and MA recovery processes for the heterogeneous recycling of TRU actinides described above (homogeneous recycling is where actinides are distributed homogeneously in the fast reactor fuels whereas in heterogeneous recycling minor actinides are contained in separate targets located at specific positions in the reactor). A particular disadvantage of homogeneous recycling of actinides is that the conversion of the plutonium-containing stream to oxide products and subsequent recycle fuel fabrication will probably require shielded rather than glove box facilities. Consequently, it is potentially more complex to industrialise when compared with heterogeneous recycle where only the much smaller MA product requires shielded target fabrication; such complexities and, therefore, costs also impact reactor re-fuelling operations. From the chemistry viewpoint, the GANEX concept presents a number of clear challenges including the requirement to co-extract and co-strip actinides in oxidation states (III–VI), separation from the trivalent lanthanides, loading capacity due to high plutonium concentrations and radiolytic degradation from high burn up fast reactor fuels. There are two main variations of the GANEX process, both of which use an initial cycle to separate bulk uranium followed by a 1-cycle TRU separation. The CEA-GANEX [65] and EURO-GANEX [66–68] processes have both been successfully demonstrated with SNF. The EURO-GANEX process is interesting in that it uses TODGA in the organic phase and a novel hydrophilic bis-triazinyl pyridine (BTP) ligand in the aqueous phase to separate lanthanides from actinides in the more easily engineered acidic region rather than a high pH region commonly needed for MA separation processes like the TALSPEAK process [69] or indeed the CEA-GANEX [8, 65]; however, this is at the expense of using a non-CHON, sulphonated-BTP ligand. There are some promising developments of prospective CHON ligands to replace sulphonated-BTP but these have not yet been tested in flowsheets [70, 71]. Chalmers University (Sweden) have also reported progress towards a one cycle GANEX process based on the selective extraction of actinides but no flowsheet tests have yet been made [72–75]. One further disadvantage of several of these processes is that they propose a mixture of two agents in the organic phase (extractant and co-extractant or modifier) in order to adjust the solvent phase capacity, viscosity, extraction kinetics or other property to give an acceptable working envelope.

2.2.5 Caesium, strontium separations

^{90}Sr and ^{137}Cs are the main heat generators in the HLW, at least in the short term (first few centuries), and so separation from other radionuclides may provide further benefits in HLW disposal than are achievable by actinide recycling only. There is, therefore, some interest in developing separate management capabilities for these elements (HHR – high heat radionuclides), including separation from the high level waste with storage of

HHR in a specific facility [76]. The comparatively short half-lives of these HHRs (~30 years) makes possible decay storage with subsequent disposal in a separate facility. This can potentially lead to significant gains in economics and repository performance due, not only to improved management of the HHRs but also, the fact that the remaining fission product wastes can be solidified as medium or low-active wastes.

Chemical processes reviewed in this section for cesium and strontium separation are mainly solvent extraction methods such as those based on dicarbollide and macrocyclic polyethers [77] and the US FPEX process [78] although a broader view was taken encompassing some of the waste treatment technologies known or used globally. This broader view included processes and plants based on extraction chromatography, as developed in Japan [11], precipitation and ion exchange/adsorption with a particular focus on inorganic ion exchangers such as ammonium molybdophosphate (AMP) [79].

2.2.6 Curium separation processes

There are a range of isotopes of curium in irradiated LWR fuel, of which, for shorter storage times post-irradiation, ^{244}Cm is particularly problematic (half-life of 18.1 y). The amounts of curium produced in irradiated fuels are small. Nevertheless, curium still makes a substantial contribution to decay heat emission for several decades post-irradiation. It is also a major source of spontaneous fission neutrons and very difficult to handle in fuel fabrication operations [80]. Thus reprocessing and fuel fabrication operations for an americium-alone fuel/target would be significantly easier at the industrial scale than a mixed Am-Cm material, with benefits to the overall transmutation system. Recycle of americium-alone can still deliver the majority of the benefits to the waste disposal and geological disposal facility (GDF) footprint [81]. Reprocessing long cooled fuels (30-70 years) also benefits curium management by altering the transmutation pathways and, for some countries, may be a beneficial approach [82]. However, separation of curium from americium is extremely difficult due to their very similar chemistry. A previous report from the EGFR has focused specifically on curium management in the fuel cycle [80] and is a key reference source; the present study provided an update to that work, particularly with regards to the development of the EXAm process in France.

Options for separating americium from curium are after removal of the rest of the actinides, fission products and lanthanides (*e.g.* the SESAME process [83]) or the recovery of americium-alone from the HLW stream, *e.g.* by the EXAm process [84]. Alternative ion exchange processes under development in Japan were also reviewed [85].

2.2.7 Industrialisation issues

This review has focused on the overall development status of the separations process flowsheet. However, the separations chemistry forms only part of the technical issues that must be addressed before a process is ready for industrial application. Such related issues include, *inter alia*, process monitoring and control [86]; radiolysis [87]; corrosion [88]; solvent regeneration; mass transfer and kinetics; nitric acid recycle [89]; criticality control and other safety case challenges [90] such as flammable gas generation [91]. In nuclear fuel reprocessing and, by extension, minor actinide partitioning solvent extraction is the most well understood and trusted technology with several decades experience accumulated of successful industrial scale operations [38, 40]. However, whilst a substantial body of knowledge exists for TBP [92-94], as used in the PUREX process or variants thereof, R&D programmes will have to generate similar levels of data for the new solvents that are proposed for many of the separation processes described above. In these matters, the interactions between the process chemistry and the engineering become important [95, 96]; thus both must be developed in an integrated fashion – for example, solvent extraction contactors with short residence times require chemical

reactions with fast chemical and/or mass transfer kinetics. Changes in performance with engineering scale up must also be understood. Opportunities should be found for innovative technologies, such as *in situ* monitoring with feedback to process controls, where these can have transformative actions on safety, safeguards and efficiency [86, 97]. Any departure from nitric acid based solvent extraction processes, *e.g.* crystallisation or chromatographic separation, will bring different challenges to ultimately deploying such alternative technologies in a nuclear fuel recycling plant.

2.3 Pyro-chemical processes

2.3.1 Introduction

Pyro-chemical (dry) processes, or some combination of hydro- and pyro- processes, offer an alternative concept to hydrometallurgical processes described in the previous section [17, 98-100]. Most typically, these are considered for metallic or other non-oxide fuels and fuels with high radiation fields such as short cooled FBR fuels, particularly in concepts such as the Integrated Fast Reactor or molten salt reactors [101, 102] but other scenarios are envisaged. For instance, in the double strata concept, with the introduction of new reactor systems combinations of aqueous and pyro- (wet and dry) processes may be a suitable fuel cycle solution. In particular, if decay periods prior to reprocessing are substantially reduced then short lived fission products will significantly increase radioactivity and heat generation. Therefore, decisions on the suitable separation process(es) depend on the overall fuel cycle strategies (this is discussed further in section 3). Thus, aqueous and pyro-electrochemical partitioning do not have to be competing options for recycling spent nuclear fuels but rather should be viewed as complementary options depending on the individual national strategies and fuel cycles to be adopted [4].

Molten salt separations are high temperature pyro-chemical processes. Temperatures vary between ~500 and ~900°C with chloride based salts typically operating at lower temperatures than fluoride based salt systems. The most commonly used salt is a eutectic mixture of lithium and potassium chloride (LiCl-KCl) and electro-refining or extraction from molten salt phase into liquid metal techniques are basic pyro-processing methods for spent fuel recycling [103]. At this point, two chloride media based, pyro-electrochemical metallurgical processes have been implemented at pilot scales. These are the process developed at RIAR (Dimitrovgrad, Russia) for oxide fuels processing [104] and the US electro-refining process for metallic fuel developed for the Integral Fast Reactor (IFR) concept [101].

Amongst the NEA member countries, variations of these main processes are also under development in Europe [105], Japan [103] and Korea [19]. Other dry processes based on fluorides are of renewed interest, especially as part of molten salt reactor (MSR) concepts [106, 107]. The other main dry process, which also has several decades of R&D history, is based on the volatility of actinide fluorides [108]. These processes are described in detail in reference [12] according to a similar hierarchy of separations as used in the review of hydro- processes, although fluoride volatility and molten salt reactor systems were considered separately due to their distinctive natures. Further details can be found in other recent reviews – *e.g.* see references [14, 103, 109].

2.3.2. Uranium recovery

Uranium recovery using a pyro-electrometallurgical process is exemplified by the facility at the Idaho National Laboratory that is treating spent EBR-II reactor metal fuel [110]. Originally intended

as part of the IFR system, this plant has provided many years of operating experience of uranium recovery from metal fast reactor fuels at the pilot-scale [111]. The process uses electro-refining in a LiCl-KCl molten chloride salt. Similar developments in Korea [112] and Japan [103] are also ongoing and further details are given in reference [12].

2.3.3 Actinide co-processing

Due to similarities in redox potentials, pyro-electrochemical processing is highly suited to group separation of transuranic actinides rather than stepwise plutonium and MA separations. Chemical processing routes have been identified based on (a) selective precipitation; (b) electrochemical applications, including electrolysis or electro-refining in chloride or fluoride molten salts; (c) liquid-liquid reductive extraction between a molten salt and a liquid metal phase. Detailed reviews of European approaches using solid aluminium cathodes in chloride media [105]; French studies on liquid-liquid extraction in fluoride melts [113] as well as progress to extend the INL process to TRU recovery using liquid metal cathodes [114] were part of this study. Pyro-processing of nitride fuels were also covered as a distinct activity with R&D in progress in Japan and Russia [115-117]. Schematic flow diagrams for two example pyro-chemical process options are shown in Fig. 6. It should be noted that not all the process steps in these schemes are underpinned. For instance, the processes in Fig. 6a for reducing $AnCl_3$ and $AlCl_3$ to metals were not studied. In fact, given potential difficulties of converting $AlCl_3$ to Al metal of sufficient purity, it may be preferable to first separate uranium on an inert cathode (as in the IFR concept) and only use Al to recover plutonium and MA, with the smaller quantities of $AlCl_3$ generated being routed to wastes rather than recovered [118].

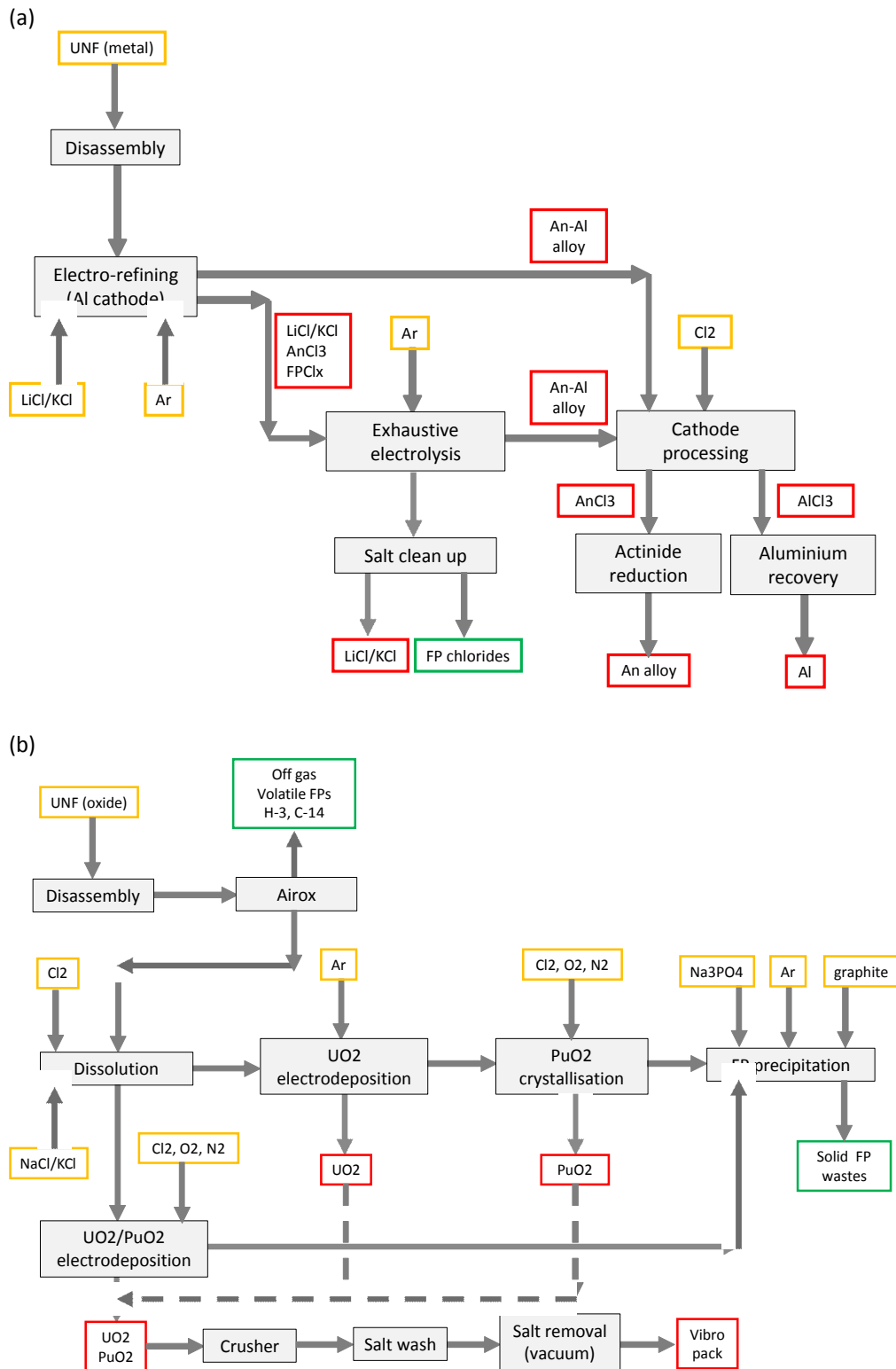


Fig. 6: Schematic flow diagrams for exemplar pyrochemical process options (a) European metal electro-refining process on solid Al cathode, [12, 105] (b) Russian electro-winning process for oxide fuels [99]

2.3.5 Separation of caesium and strontium

Separations of Group I and II elements (caesium and strontium) from LiCl salt wastes arising from the electro-reduction process have been investigated using methods such as chemical agent addition [119] and ion-exchange [120]. However, these separations from LiCl salt wastes are very difficult and the only process considered in the state of the art review was a melt crystallisation process developed in Korea [121-123]. This concept is based on differences in solubilities of the impurities between the solid (*i.e.* crystal) and melt phases during the partial crystallisation of multi-component melts. The impurities are preferentially distributed in the melt phase with >90% separation efficiency for both CsCl and SrCl₂ reported for a salt re-use rate of 90 wt.%. The main focus of the Korean programme has been on a layer crystallisation process.

2.3.6. Fluoride volatilisation processes

The “fluoride volatility” separation method relies on the ability of uranium to form the volatile hexafluoride (UF₆). In the reprocessing of spent fuel, fluorination is carried out with fluorine or alternative strong fluorinating compounds and the volatile products such as uranium and plutonium hexafluorides can be separated from non-volatile fluoride products, notably fission products. Whilst uranium readily forms volatile UF₆, plutonium can form mixtures of volatile PuF₆ and non-volatile PuF₄ depending on reaction conditions. Thermal decomposition of PuF₆ can be exploited to separate uranium and plutonium [108, 124]. Fluoride volatility methods can be classified according to types in which: (i) reactions are between fluorine and a fused salt and, (ii) there is the fluorination of a solid powder [108, 125-130].

The Molten Salt Reactor Experiment (MSRE) at Oak Ridge National Laboratory (ORNL) in the 1960-70s developed the method of bubbling fluorine into a fused fluoride salt to generate volatile fluoride products enabling the separation of uranium (as UF₆) from the molten fluoride salt carrier [128]. The molten salt was based on LiF-BeF₂ or LiF-BeF₂-ZrF₄ mixtures and complete separation of uranium from the MSRE salt was demonstrated [128].

The “Fluoride Volatility Method” was a process originally designed for Fast Breeder Reactor (FBR) fuel reprocessing based on the reaction between fluorine and powdered spent fuel oxides. It was investigated primarily in the USA, Russia, France, Czech Republic, Belgium and Japan [130] with more limited recent development in the Czech Republic, Russia and Japan [108, 125, 131]. Fluorination is usually carried out either in a fluidised bed reactor or in a flame fluorinator and high efficiencies for uranium recoveries are obtained. Plutonium recovery is more difficult due to inter-conversions of PuF₆ and PuF₄ but some promising results have been obtained, whilst the efficiencies for recovering other minor actinides remains unclear. The study [12] specifically reviewed the fluoride volatility method [129], fused salt volatilisation method [128] and the FLUOREX process [132]. Schematic flow diagrams for fluoride volatility process options are shown in Fig. 7.

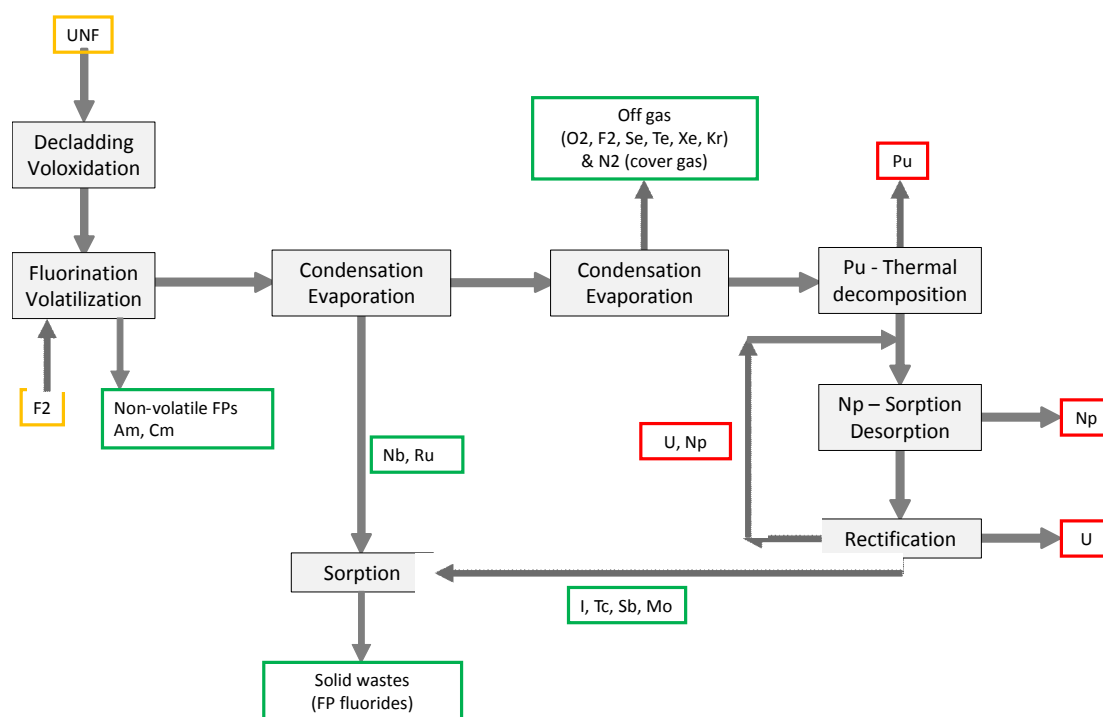


Fig. 7: Basic flow diagram for a fluoride volatility process option [12]

2.3.7. Molten salt reactors fuel separations

Molten salt reactors (MSRs) are different to conventional reactors in that the molten salt acts as both the fuel and coolant in the primary circuit. The fuel (fissile material) is dissolved in a molten salt [109]. MSRs can be used as part of either the U-Pu or Th-²³³U fuel cycles in breeder or TRU burner modes but require connection to an on-line or on-site fuel processing unit. The choice of chemical separation processes and technologies to be used for fuel processing must account for these factors [133, 134]. In particular, the separation processes have to be compatible with the molten fluoride or chloride fuel at temperatures over 500 °C and, as the fuel is processed almost immediately after leaving the reactor core, high radiation resistance is necessary. Initial developments in the 1950-1970s focused on fuel reprocessing technology for a 1000 MWe Li,Be,Th,U/F Molten Salt Breeder Reactor. Development of the fuel cycles for different MSR designs was restarted as part of GIF after 2001, building on the original investigations. The main separation processes applicable to processing the liquid molten salt fuel are [102, 135]:

- Gas extraction process (or helium bubbling)
- Fused salt volatilisation
- Molten salt / liquid metal reductive extraction
- Electrochemical separation techniques
- Vacuum distillation of molten salt
- Oxide co-precipitation.

These processes can be used to clean-up liquid fluoride MSR fuels from fission products (neutron poisons) at high temperatures after its removal from the primary circuit when the decay heat is still significant. Current

MSR designs have been proposed based on fast spectrum MSRs such as the Molten Salt Fast Reactor (MSFR) and Molten Salt Actinide Recycler and Transmuter (MOSART) [136]. Other variations also exist signalling a renewed interest in this type of system, primarily as options for development of small modular reactors (SMR). Fig. 8 indicates how the reprocessing cycle integrates with the MSR (thorium fuel cycle). Again, it is emphasised that not all process steps are fully underpinned (e.g. U, Pa electro-winning stage) and historic data may not be sufficient in this regard, particularly as designs evolve.

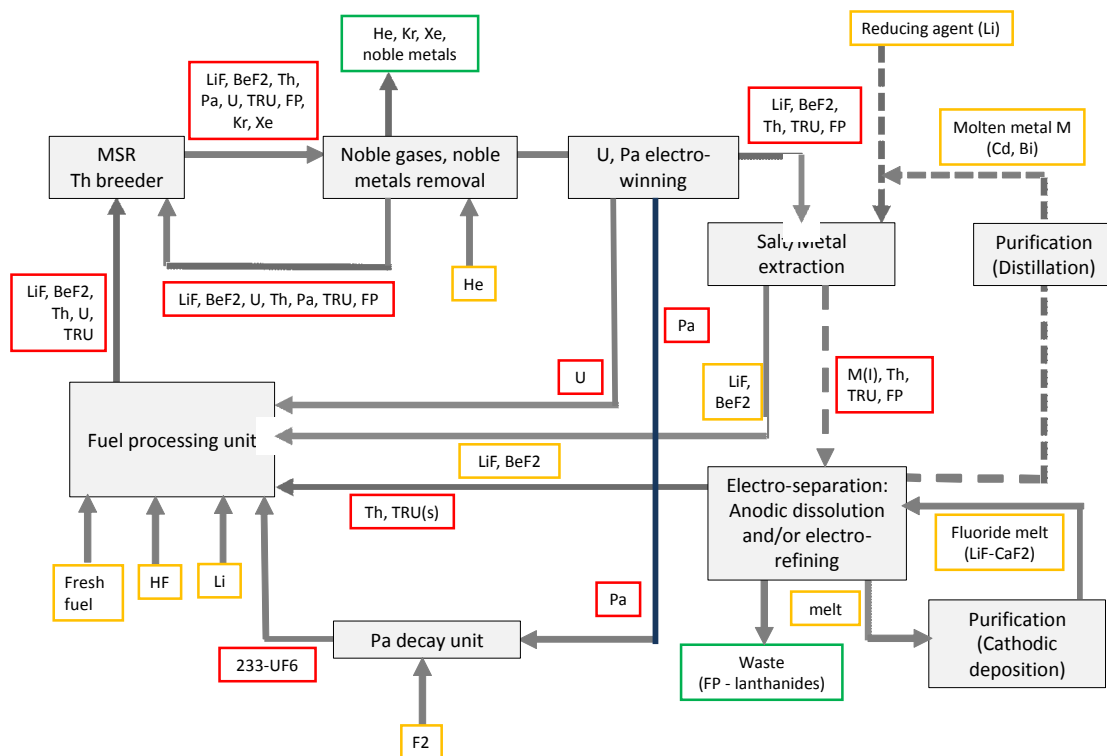


Fig. 8: The MSR fuel cycle concept for thorium breeder reactor with single fluid on-line reprocessing technology [12]

2.3.8. Industrialisation issues

The main pyro-chemical processes under development today are either based on the metal electro-refining or oxide electro-winning processes. These are small batch processes and the prospects for industrialisation of these processes present substantial challenges around, *inter alia*, salts handling; scaled up and/or continuous operations; corrosion; materials compatibility; atmosphere control; salt properties; product processing; safeguards and accountancy; process control; salt clean up; salt waste management. Whilst the core processes may be well understood, some individual stages within the overall pyro-processing schemes illustrated above are not underpinned. Understanding the fuel cycle scenarios in which pyro-processes are to be preferred over aqueous processes is a key strategic goal. Pyro-processes would bring additional regulatory challenges due to lack of past commercial scale licensing experience. The Fluoride Volatility Method has not been widely studied

in recent times and MSR require an on-line salt cleanup process, although they face many similar issues to those noted above for molten salt reprocessing.

2.4 Summary

The compilation of information on a wide range of separations processes that are under development in NEA member countries was a major study for the EGFR. The result is a comprehensive assessment of the state of the art in both hydro- and pyro- advanced reprocessing and MA or fission product partitioning methods, in the Member Countries. A special feature of this study was grouping the processes according to their function. Broadly, this hierarchy reflects the order in which sequential separations would be applied to enhance fuel cycle sustainability and reduce the waste burden, although there is an increasing tendency towards simplification. This is partly driven by the recognition that increasing the stages of separations will increase costs and secondary waste generation and multiple sequential separations, particularly for hydro-processing, will not be a commercially viable proposal. Other factors, such as reprocessing long cooled fuels, also have significant benefits in simplifying the processes and may be useful in some countries' national strategies. Some specialised fuel cycles, such as those based on MSRs, present their own unique challenges and require bespoke solutions. Based on this review and assessment of the state of the art, the objective of the study was to make a technology readiness assessment of the various options. However, it was clear from an early stage of the study that comparing processes, or even recommending which separations processes should be adopted, was not an achievable or even desirable outcome. Therefore, an additional stage of work was introduced that considered the wide ranging criteria and scenarios that would affect the choice of separation process for a future closed fuel cycle (Section 3). Following this evaluation, the EGFR proceeded to assess the TRL using an adapted 'fit for purpose' methodology (Section 4).

3 Influences on technology development and evaluation

3.1 Introduction

Having reviewed various separation processes for major (U, Pu) and minor (Np, Am, Cm) actinide recycling, the key objective of the study was to assess their technical maturity. The well-known TRL (Technology Readiness Level) scale was selected as the basis for this assessment [15]. However, before that assessment can be made some consideration must be given to the factors that influence the development of separation processes for nuclear fuel recycling through the R&D phases towards eventual industrial deployment.

3.2 Performance criteria: definition of feeds to and products from the recycling plant

The international development of advanced nuclear fuel cycles, at present, covers a wide range of reactor types and different fuels from standard thermal oxides to fast reactor fuels based on oxides, nitrides, carbides and metal alloys [137, 138]. Molten salt reactors (MSRs), with liquid fuel in a high temperature molten salt, are gaining momentum [109] as well as alternative thermal fuels such as TRISO-type fuels for high temperature reactors [139] and accident tolerant fuels (ATF) for current reactors [140, 141]. There is also some interest in thorium based fuels that could replace the established (U,Pu) fuel cycle [142, 143]. As well as the fuel matrix, these fuels will differ in other important characteristics such as burn up, initial fissile content and decay period before reprocessing. The quantities of arising spent fuels will vary and, in conjunction with the previously listed factors, this will affect the throughput and, hence, sizes of recycling plants.

Regardless, of whether aqueous or pyro- processes are selected for separations, many of the challenges presented by the wide variety of potential feed materials will be addressed within the Head-end plant area where spent fuels are mechanically prepared for reprocessing and chemically dissolved either into nitric acid or a molten salt [16]. It is a primary purpose of Head-end to prepare dissolved fuel solutions of a specified composition for separations, accepting that some parameters, such as plutonium content, may vary within specified ranges depending on the fuel characteristics. The uncertainties around feed materials, therefore, have probably the biggest impacts on the Head-end technology selection.

As well as the nature of the feeds to the reprocessing plant, the products required from the plant – that will, in turn, become recycle feeds to the fuel manufacturing plants – will also influence the selection of reprocessing technology. Future recycle plants will likely need to integrate the spent fuel reprocessing and recycled fuel production in a single entity. This is necessary primarily for safeguards and security reasons but also should deliver efficiency and economic benefits at the full system level [144]. Such integration provides the opportunity to closely align the reprocessing plant product finishing processes with the recycle fuel manufacturing processes and thus optimise operations [145]. As with fuels, there is a wide range of potential forms for products, from bulk uranium, to co-processed TRU or (U,TRU) mixtures. It is reasonable to assume that pure plutonium products will be undesirable from the safeguards attractiveness-level viewpoint based on perceived proliferation risks [144]. As noted above, MA-containing products may require shielded facilities for manufacturing and some products, such as those from pyro-electrochemical processing, may tolerate greater quantities of fission products (*i.e.* lower decontamination factors achieved in reprocessing) [146].

A range of technologies have been investigated for the conversion of product finishing processes. For an aqueous process, thermal denitration [147] is an industrial scale, reliable process for industrial scale bulk uranium conversion to oxide. Options to co-convert (U,Pu) or TRU liquid products to solids include microwave heating denitration, modified direct denitration (MDD), oxalate precipitation and gelation methods [43, 147-

149]. The metal products from electro-refining are generally cast into ingots for recycle as metal alloy fuels in fast reactors [103, 150].

Clearly, once decisions to implement advanced fuel cycles are taken, for detailed design to progress the feeds to and products from the reprocessing plant and, hence, the performance required from the separation processes, will need to be set. However, few nuclear energy programmes are, at present, at this stage and most are focused on research, developing and demonstrating future options. Consequently, separation processes cannot be easily distinguished based on performance but must be seen in the context of their purpose within the various national or international level scenarios for closing the fuel cycle. This inevitably requires a focus on flexibility and introduces some subjectivity into subsequent evaluations of technical maturity.

3.3 Constraints placed on the recycling plant(s)

Closing the fuel cycle is complex and subject to a wide range of influences. Whilst closed fuel cycles can bring a range of benefits, for instance with respect to sustainability, GDF utilisation and public acceptability, there are also a number of perceived problems that must be addressed within R&D programmes [151]. Future recycling plants designs will thus need to address these problems and manage any additional constraints that arise from them. For instance, safety and nuclear security requirements placed on design and operations by regulators will undoubtedly become ever-more challenging [90, 95]. Regulatory limits for environmental discharges and waste generation will also become tighter [6, 152] but there will be a need for flexibility to manage a more diverse range of fuel types. All these challenges will need to be addressed whilst ensuring that the economics of future recycling remain affordable [153, 154]. Affordability will drive the need to reduce capital costs by reducing plant sizes and overall footprint of the recycling site. This will require the impacts of changes to the separation processes to be assessed at the plant and even recycle site scales. Therefore, the development of the engineering technologies must progress alongside the chemical flowsheet development with a strong emphasis on technologies that intensify the processes and reduce size (area, volume, wastes). Table 1 gives examples of how reprocessing plant flowsheets can be adapted to meet these challenges of safety, sustainability, wastes, non-proliferation and improved economics.

Whilst these are major issues and so not considered in detail within the current study, it can be argued that addressing safety, environmental impact and waste management concerns is a reasonably predictable matter of meeting statutory or regulatory requirements, where success is compliance with national or international standards although some foresight is required to anticipate likely future changes. Another key issue is economics but requirements should be definable by what is an acceptable cost to the specific governments and utilities. Minimising costs to the point at which recycling is competitive with other options for managing SNF (*i.e.* storage and direct disposal) is a necessary target if the benefits of recycling are to be realised. Fuel cycle costs are a complex issue that are being addressed by other groups within the NEA and thus were outside the scope of this study [153]. Most published studies tend to agree, however, that back end fuel cycle costs whether for open or closed cycles are a small proportion of the nuclear electricity cost compared to front end activities [155-160]. Contrastingly, the EGFRC considered that the analysis of proliferation risks associated with future fuel recycling plants is somewhat different. It is a contentious and complex issue comprising both 'technical' and 'political' arguments about the extent of the risk and value of various controls, and thus needed to be investigated further in this study. This is specifically discussed in the next section.

Table 1: Requirements for advanced reprocessing (reprinted from ref.[12] by permission of the OECD-NEA)

Strategic objective	Fuel cycle requirement	Potential implications for reprocessing plant flowsheets
Process safety	<ul style="list-style-type: none"> o Geometrically safe o Safer processes 	<ul style="list-style-type: none"> o Engineering design o Continuous vs. batch processes o Non-aqueous processes o Low temperature & pressure processes o Avoid specific reagents, <i>e.g.</i> hydrazine o Fundamental understanding of process chemistry o Modelling and simulation
Sustainability	Plutonium multi-recycling	<ul style="list-style-type: none"> o Dissolution of high Pu content fuels o High Pu recovery yield o High Pu concentrations in plant o High solvent loading, risk of 3rd phase o Increased solvent degradation and radiolysis o Ruthenium decontamination in short cooled fuels
Waste management & environmental impact	<ul style="list-style-type: none"> o Minor actinides multi-recycled for transmutation (P&T) and Plutonium Multi-recycling o Reduced impact of fuel cycle on GDF (geological disposal facility) footprint (radiotoxicity and heat loading) o Reduced waste generation and lower environmental impact 	<ul style="list-style-type: none"> o >99% recovery of Np from flowsheet o Options needed for >99% recovery of Am+Cm and Am alone from HLW o Cs, Sr separations or decay storage o Reduced number and volumes of aqueous waste streams o Ideally CHON reagents (<i>e.g.</i> salt free solvent washing) and solvents or, if not, compatibility with waste forms. o Target “near-zero” emissions o Capture of volatile species (³H, ¹⁴C, ⁸⁵Kr, ¹²⁹I) o New wasteforms
Non-proliferation	<ul style="list-style-type: none"> o Increased physical safeguards and monitoring o Integrated SNF storage, separations and recycle fuel/targets fabrication 	<ul style="list-style-type: none"> o (U+Pu) or (U+Np+Pu) co-processing in Advanced PUREX flowsheets o Increased fission and/or minor-actinide product contamination of Pu product o Near real time accountancy, on-line process monitoring o Sampling and analysis o Proliferation resistance assessment methodology

Economic	<ul style="list-style-type: none"> o Reduced capital costs through smaller plant footprint o Greater flexibility of processes 	<ul style="list-style-type: none"> o Intensified processing o Fit-for-purpose equipment for solvent extraction o Single cycle reprocessing o Fewer waste streams o Integrate with waste and fuel fabrication plants o Process LWR, MOX and fast reactor fuels o Feed variations (carbide, nitride and metal fuels as well as oxides) o Higher burn ups o Short cooled fast reactor or very long cooled LWR fuels – depends on scenario
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3.4 Non-proliferation aspects

A number of basic technical issues related to “proliferation resistance” or, perhaps more precisely, the perception of proliferation risks in closed fuel cycles were considered in this study without specifying separations processes. These were:

- Systems studies, including isotopics
- Impacts of separating plutonium
- Engineered safeguards applied
- Uncertainties in non-proliferation assessments

Systems studies are an appropriate means to examine the proliferation risks associated with various nuclear fuel cycle options. The fissile isotopes that are of primary proliferation concern are: ^{235}U , ^{239}Pu , ^{241}Pu for the (U, Pu) fuel cycle and ^{233}U for the (Th, U) fuel cycle [142]. Plutonium is of special interest as 50–80 % of the total plutonium content is fissile, depending on burnup, and plutonium can be isolated by chemical means rather than requiring isotopic separations and enrichment [161, 162] although these other parts of the fuel cycle may also present proliferation risks [163]. Other actinides and fission products in spent fuel or reprocessed uranium and plutonium products can actually reduce the proliferation risks (or “attractiveness level” of the materials) by providing a radiation barrier [144]. That is, the radiation emitted by the material makes it more difficult to divert for misuse.

In the open or once-through fuel cycle, where SNF is directly disposed, the intrinsic radiation field is widely considered to be the primary safeguard. However, this ‘radiation barrier’ decreases exponentially with storage time due to the radioactive decay of short-lived and intermediate-lived radioactive fission products. Thus, after a period of several decades, the SNF becomes more vulnerable (proliferation risk increases) to the point at which the radiation barrier is insufficient to maintain this self-protection [155, 164].

So, it can be concluded from systems studies that there are no intrinsic safeguards for plutonium even when retained in unseparated form in SNF. Whilst unseparated plutonium might have a lower “attractiveness level” than pure plutonium, it can still be recovered and this becomes easier with storage time. Hence, physical protection and other safeguards are necessary for plutonium in all forms, even within SNF. So locations in

which SNF is long term stored or directly disposed should also be protected because recovery of the plutonium remains possible.

The concept of “safeguards-by-design” is a valuable tool to ensure engineered safeguards and physical protection requirements are incorporated from the outset. Using this concept, integrated spent fuel treatment facilities are proposed [155]. That is, for the closed fuel cycle, facilities for spent fuel storage, reprocessing and fuel manufacturing are co-located within an integrated secure recycle plant. The fissile material (*i.e.* plutonium) is then controlled as it only enters and leaves the Recycle Plant as large, heavy, easily accountable fuel assemblies (irradiated or non-irradiated). Wastes and personnel movements can be monitored and use of “near-real-time” plutonium accountancy to track movements within the plant, can then be utilized to the greatest extent technologically possible to maintain safeguards. So the choice of separation technologies can be made for reasons of process efficiency, technical maturity and economics rather than proliferation risk.

MSRs are a potential exception in that for on-line reprocessing of MSR fuels the configuration and properties of the process are probably the main safeguarding methods and whilst ^{233}U can be separated by on-line reprocessing, there are intrinsic difficulties due to the high radiation fields present that provide additional barriers.

Any facilities dealing with nuclear materials in open or closed cycles, therefore, present proliferation risks that will require mitigation through physical protection and materials safeguards. Hence, it can be assumed that all legitimate operations must be under safeguards and accountancy regimes that comply with international norms and oversight, such as those of the IAEA [165, 166]. Measures that increase the ‘defence in depth’ by adding further barriers should thus be seen as useful enhancements to processes but not critical to meeting non-proliferation and security requirements. Such enhancements are a useful aim of R&D. Proliferation and physical security risk analysis of separation technologies using methods that can provide quantitative or relative comparisons [144, 167] is another useful R&D aim, particularly where additional barriers to proliferation can be identified that would add to the defence in depth.

3.5 Down selection

It is concluded that clear-cut comparisons between separations processes, with the associated goal of down-selecting options, are not really possible at this time. This is because specific design criteria are not established for the majority of R&D programmes. Hence, a wide range of options have been developed for various future nuclear energy scenarios. Furthermore, there are other factors such as cost, public acceptability, proliferation risks and environmental impact that will influence decisions regarding nuclear fuel cycle options. Also, the conclusions from national level fuel cycle analyses change over time due to changing political, economic and social conditions as well as technical advances.

Nevertheless, the choice of separation process(es) for nuclear fuel recycling is probably the critical factor in realising the benefits of and addressing the barriers to, closing the nuclear fuel cycle. Therefore, from this phase of the study, the EGFRC concluded that there is a clear need to progress the technical development of separation processes through R&D, specifically to:

- (i) *“develop methodologies to compare fuel cycle options against the different pathways for nuclear energy”;*
- (ii) *“prove technical feasibility of leading options for a range of likely fuel cycle scenarios (up to TRL 6)”;*
- (iii) *“demonstrate selected technologies at engineering scales with spent fuels (TRL 7-8)”.*

The needs for technology demonstration and the metrics to assess technology readiness form the next chapter of this paper.

4. Analysis of technology readiness for chemical separation processes

4.1 Introduction

The section above concluded that there was a need to develop methodologies that enable fuel cycle options, including separation processes, to be compared and for technical maturity to be demonstrated. To develop a methodology and apply it in a self-consistent manner to the range of separation processes reviewed in section 2 was the final aim of the EGFR's study. It is widely appreciated that technical maturity can be conveniently assessed using the well-known and internationally recognisable Technology Readiness Level (TRL) system [15, 95]. In our study, TRLs were assessed for the individual separation processes without broader consideration of how they fit within the full recycling plant layout or the readiness of the engineering design and equipment needs. In this regard, the simplifying view was taken that most of the separation processes would be operable using established process technologies, if necessary. However, it is recognised that R&D in engineering is needed to deliver some of the key objectives such as process safety, efficiency (cost savings) and safeguards. Also, the assessments made here are at the 'system' level not at the component level; that is, some individual steps within a separation process (which we have assessed as a whole system), may actually be at a lower TRL than that at which we have judged the overall system to be – whereas a stricter interpretation of TRL assessment would rate the overall system to have a TRL equal to the lowest TRL of any component. For the present exercise we did not consider that to be a useful output to aid a general understanding of the technological status of the range of advanced separation processes currently under development internationally.

4.1.1 Definition of TRLs for Comparing Separations Processes

The TRL scale is primarily aimed at development and testing of engineered components and runs from level 1 – initial generation of the idea – through to level 9 – routine use of the component in the operational environment for which it was designed. Levels 1-3 are generally basic studies collectively designated "proof of concept", levels 4-6 are the development of the component under increasingly realistic conditions – "proof of principle" – and levels 7-9 ("proof of performance") take the component through operational tests to full deployment in the system for which it was designed. Prior to application, therefore, the TRL definitions needed to be adapted to be relevant to assessing chemical processes for nuclear fuel recycling whilst retaining the key features of each stage, see Table 2 for the TRLs defined and used in this study. The NEA Expert Group on Innovative Fuels have performed a similar exercise to assess the R&D status of transmutation fuels in which an innovative matrix approach was developed [138]. Our study also developed a matrix method in order to develop an understanding of both the scale at which processes had been tested and what type of materials (feeds) these tests used. This was considered necessary to reflect the reality of R&D programmes for separation process development. For example, non-active or trace active simulants are commonly used for fundamental studies at lab scales where an understanding of how specific metal ions interact with an extracting ligand are needed but high activity spent fuel solutions are rarely, if ever, used for such basic studies. Flowsheet tests with spent fuel solutions, however, are necessary to demonstrate the process under realistic conditions (TRL5-7). The matrix developed and how the TRLs, defined in Table 2, map onto the matrix are illustrated in Table 3. In this approach, achieving a specific level is through a combination of studies at the appropriate scale using appropriate feed materials. Process development, therefore, typically progresses in a diagonal fashion from bottom left to top right of the matrix as the TRL rises. The matrix thus provides evidence underpinning the choice of TRL that reduces any subjectivity in the expert judgments made (re-emphasising

that the analysis was made at the system level considering the status of the basic separation process as a whole and accepting that technological or knowledge gaps may remain in individual stages). An example of the assessments made is given for Russian head-end processes in Table 3 where dark blue shaded squares indicate progress against the matrix supporting a claimed TRL of close to 7.

Having defined the methodology to be adopted, the process followed was that the most appropriate experts from the EGFRC were selected for each process requiring assessment and the individual assessments were then moderated by the full group to, as far as possible, ensure consistency across the full spectrum of assessments. Each assessment was supported by a short evidence statement (the various individual assessments are included in reference [12] except for a small number of new assessments reported here). The results of the assessments are summarised in Figs. 10-12 for:

- Head-end processes
- Hydro-metallurgical processes (sub-divided as in section 2)
- Pyro-chemical processes (sub-divided as in section 2)

Table 2: Summary of TRL definition for separation processes (adapted from ref. [12])

TR L	Function	Definition
9	Proof of performance	Multiple years of operational experience established at industrial scale.
8		Full scale process demonstrated in a limited operational environment.
7		Prototype system demonstrated under conditions fully representative of operations.
6	Proof of principle	Engineering or pilot scale testing of technology component or process step. Process flowsheets proven through hot tests using spent fuel. Process models validated.
5		Technology component or process step validated at bench scale under relevant conditions. Process models developed. Proof of principle hot tests using spent fuel.
4		Technology component or process step validated under laboratory conditions. Tests performed using active materials in simulated feeds. Fundamental properties measured.
3	Proof of concept	Lab scale tests to prove concepts, fundamental data obtained
2		

		Technology application developed and options investigated
1		Initial concepts are proposed and basic principles established

Table 3: Matrix approach to TRL evaluation: example from Head-end Processes — Russian process for decladding, fuel oxidation and release of tritium indicating TRL 6-7 applies (adapted from ref.[12])

Commercial feeds				TRL 8-9
Irradiated fuels or targets		TRL 6-7	TRL 7	
Full simulants	TRL 3-4	TRL 4-5	TRL 6	
Partial simulants or representative materials	TRL 2-3	TRL 3-4		
Inactive or trace active	TRL 1-2			
	Fundamental studies	Flowsheet tests	Maloperation studies	Full scale operations

4.2 Head-end processes

Current industrial processes for head-end operations, disassembly of irradiated fuels and dissolution in nitric acid, are obviously TRL 9. Of the other processes described in section 2, the dry processes for pre-converting spent fuel into a powder with oxidation of UO_2 to U_3O_8 and concomitant release of volatile products are in the range 5-6, possibly up to TRL 7. Such dry processes can be precursors for either nitric acid or molten salt dissolution. With regards to the abatement of off gases (tritium, iodine-129, krypton-86 and carbon-14), processes that capture these species from off gas streams and immobilise them in waste forms for disposal are under development and again TRLs are in the range 5-6. An additional dry process for zirconium recovery from hulls is evaluated at TRL 6-7.

So, the head-end processes evaluated within this study are fairly well developed with TRLs generally in the range of 5 to 6, possibly higher when considering UOx fuel oxidation. They are thus close to the point at which industrialisation could be considered, at least for thermal UOx fuel reprocessing. Head-end processes for fast reactor reprocessing, non-oxide based fuels and high plutonium content fuels are at a less mature level. Also, the mechanical operations within Head-end make a substantial contribution to the cost and complexity of aqueous reprocessing and further R&D is needed to optimise the engineering of head-end plant.

4.3 Aqueous separations

Current industrial processes for chemical separations, that is solvent extraction using the PUREX process, are obviously TRL 9. COEX™, Simplified PUREX and NUEX™ are really evolutions of the PUREX process and, although the published data underpinning these processes are variable, they can be considered to be of rather high technical maturity with a degree of proven performance due to their basic similarity to the conventional PUREX process. At the system level, TRLs are in the range 6-8 but it is recognised that some stages or components may strictly be of lower technical readiness and require further R&D. For instance, if centrifugal contactors are to replace pulsed columns and/or mixer-settlers; control of mixed U-Pu products and the behaviours of any new reagents proposed for the processes.

There are then a range of more radical PUREX variations including the NEXT process (U crystallisation and the 2nd solvent extraction step), UREX co-decontamination (CoDCon) and Advanced PUREX that are broadly in the proof of principle range (TRL 4-6). These processes offer more substantial gains in aspects such as economics, waste generation and proliferation resistance but are correspondingly less mature although there is a reasonably high confidence that such processes could eventually be industrially deployable. Demonstration of these processes in increasingly realistic environments and with process equipment is needed to raise TRLs – proving key issues such as fission product decontamination factors in the single cycle; control of the U/Pu ratio in mixed products; neptunium and technetium behaviours; solvent recycling and process monitoring technologies. The non-PUREX uranium only separation processes (GANEX first cycle and crystallisation) are comparatively simple and demonstrations to date indicate a TRL of ~5 but similar issues still require addressing in R&D programmes.

It is notable here that there are some options (NEXT; Russian REPA and Co-crystallisation processes) which utilise crystallisation in place of solvent extraction. This would be a significant technological change in nuclear fuel reprocessing at the industrial scale and so these processes are still in the TRL range of 4-5 although hot tests have been performed in some cases.

Section 2 described the wide range of MA separation processes that are under development and their different purposes; *i.e.* actinide-lanthanide co-recovery, actinide/lanthanide separation, americium/curium separation *etc.* The most mature processes with TRL 6-7 are those that employ two solvent extraction cycles to co-separate trivalent actinides and lanthanides away from the remaining fission products in the HLW before separating them using actinide selective agents (*e.g.* DIAMEX; TRUEX-TALSPEAK). Arguably, the French DIAMEX-SANEX combined process is close to this level, assessed here as TRL 5. Given such success in the initially challenging goal of developing processes for recovering trivalent MAs, R&D is now generally focused on simplifying these options into one solvent extraction cycle that can be directly applied to PUREX highly active raffinate. These are obviously less mature, with European SANEX processes, of which there are a number of variations, having TRLs around 4, as is the case for other proposed processes; *inter alia* US ALSEP and Advanced TALSPEAK (or TALSQUEAK), Japanese TddDGA, Russian DGA or diamide based processes. Some key technical issues remain to be addressed with these MA separation processes, such as: slow kinetics; loading capacity; third phase formation; solvent properties; use of non-CHON molecules and difficult to control actinide stripping at high pHs.

It is seen in section 2 that there are substantial challenges in the co-extraction of all TRU actinides for homogeneous recycling scenarios, as proposed in the GANEX process [66]. At present, two options have been developed and hot tested and so have reached a similar stage of technology readiness (TRL 4-5): CEA-GANEX and EURO-GANEX whilst the more ambitious CHALMEX option remains in the proof of concept phase (TRL 2). Significant issues remain though for R&D before homogeneous recycling is a realistically deployable concept using the GANEX process.

Processes for caesium and strontium removal from wastes cover a wide range of applications. Some are already used industrially in nuclear waste treatment (TRL >6) although those specifically being developed for

treating HLW from reprocessing (rather than decontamination of dilute effluents) such as the CCD-PEG, FPEX and extraction chromatography are still in the TRL 4-6 range (proof of principle).

For americium/curium separations, the French EXAm process is judged to be the most well developed (TRL 6) but this process relies on a very small difference in the separation factor such that 68 stages were used in the flowsheet test [8].

This study has focused on the main hydrometallurgical separation processes being developed in NEA member countries. TRLs vary widely depending on the type of separation being targeted and the ambition. For example, processes for (U,Pu) reprocessing that are evolutions of the PUREX process could probably be industrialised now whereas more radical processes, with lower TRLs, still require significant R&D to prove the performance that would be achievable in an operational environment. MA partitioning is in a similar state with some options having high TRLs but with major drawbacks (difficult engineering; expensive; wastes *etc.*). More attractive options such as single cycle flowsheets and americium-only extractions are generally lower TRL and require further R&D before deployment can be considered. Nevertheless, it is clear that substantial improvements to the separation processes at the heart of nuclear fuel reprocessing can be made and that the extraordinarily difficult separation of trivalent actinides from lanthanides in HLW is technically feasible.

4.4 Pyro-chemical separations

The US electro-chemical process at the Idaho National Laboratory has demonstrated uranium recovery from EBR-II fuels and, together with the Russian RIAR process, is the most well demonstrated pyro-process for spent fuel – both are evaluated to be at TRL 7. Korean processes, that include electro-reduction of oxide fuels prior to electro-refining, are in range TRL 3-4 overall but the PRIDE facility will generate experience with uranium-active processing at the pilot scale moving them into TRL 4-6.

Actinide co-recovery processes are lower TRL (apart from the Russian RIAR pyro-chemical process noted above). Other dry options such as fluoride volatility are generally TRL 3-4.

Whilst batch operations, especially for uranium recovery and metallic fuels, are well proven at the pilot scale, pyro-processing for larger scale reprocessing of spent fuels with recycle of TRU is further from industrialisation. Ancillary processes are needed for oxide fuels (electro-reduction) and salt waste treatment and some of the intermediate process steps in the overall schemes are not yet underpinned, requiring technology development. Nevertheless, promising options exist that have been developed to the proof of principle stage and, hence, pyro-processes offer complementary technology for actinide recovery that may be better suited to some fuel cycle scenarios, *e.g.* recycle of FBR metal fuels, and of course MSRs.

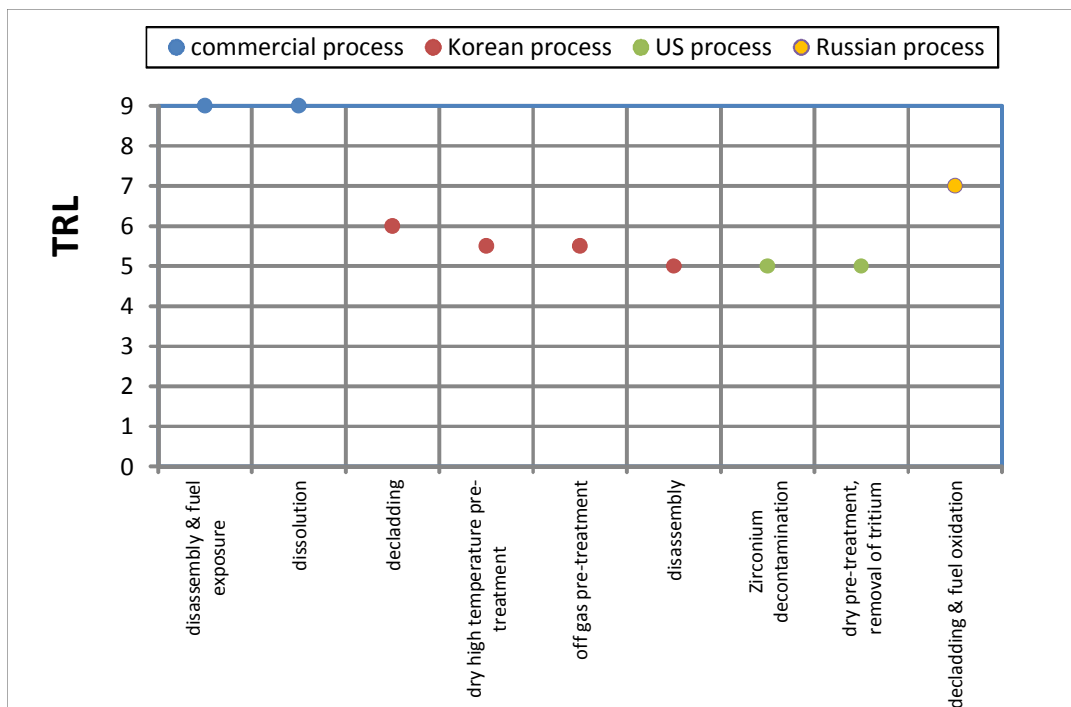


Fig. 10: Summary of TRLs for head-end processes (adapted from ref.[12])

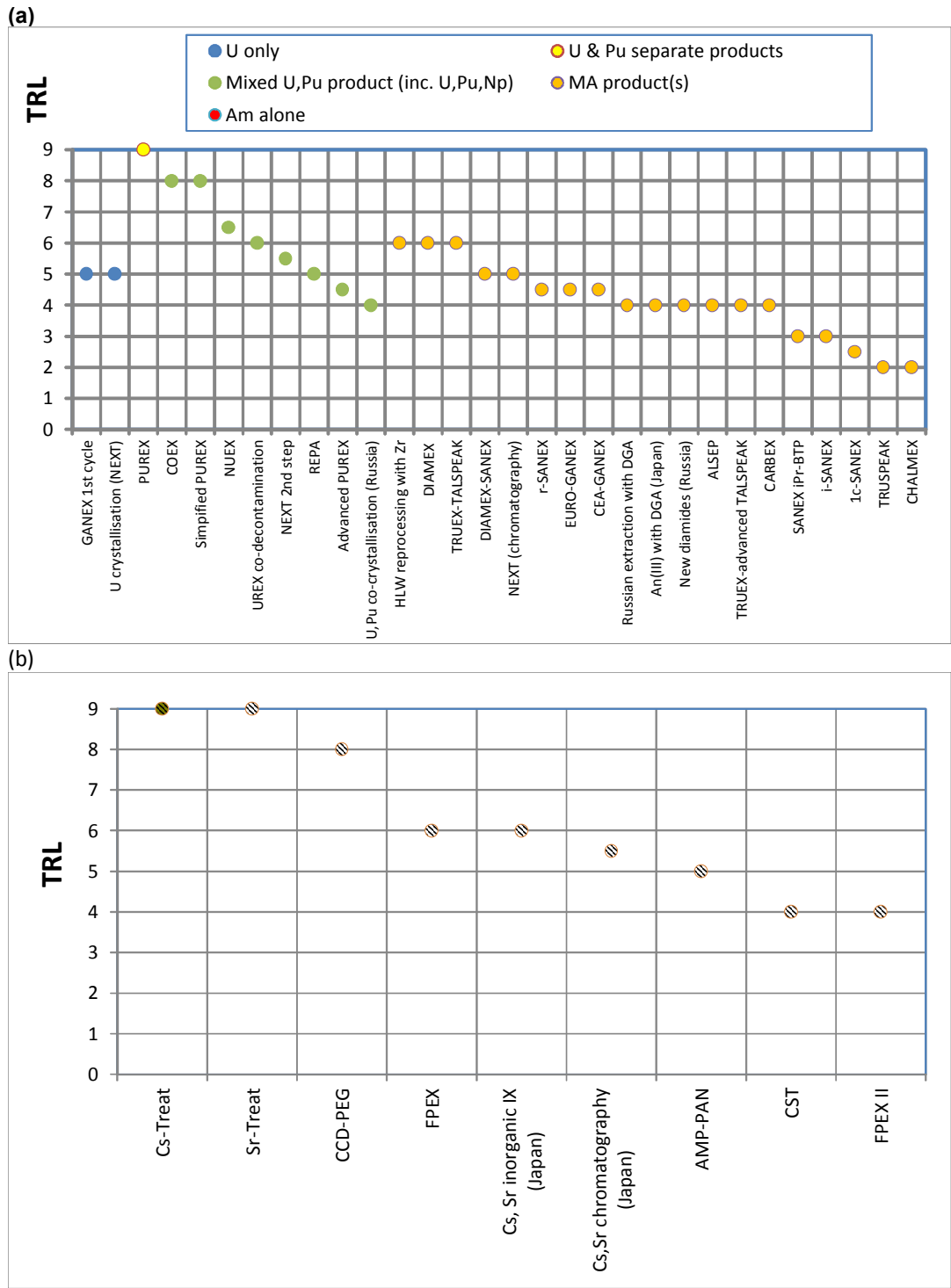


Fig. 11: Summary of TRLs for aqueous processes (a) actinide separation processes and (b) HHR (Cs, Sr) separation processes (adapted from ref.[12])

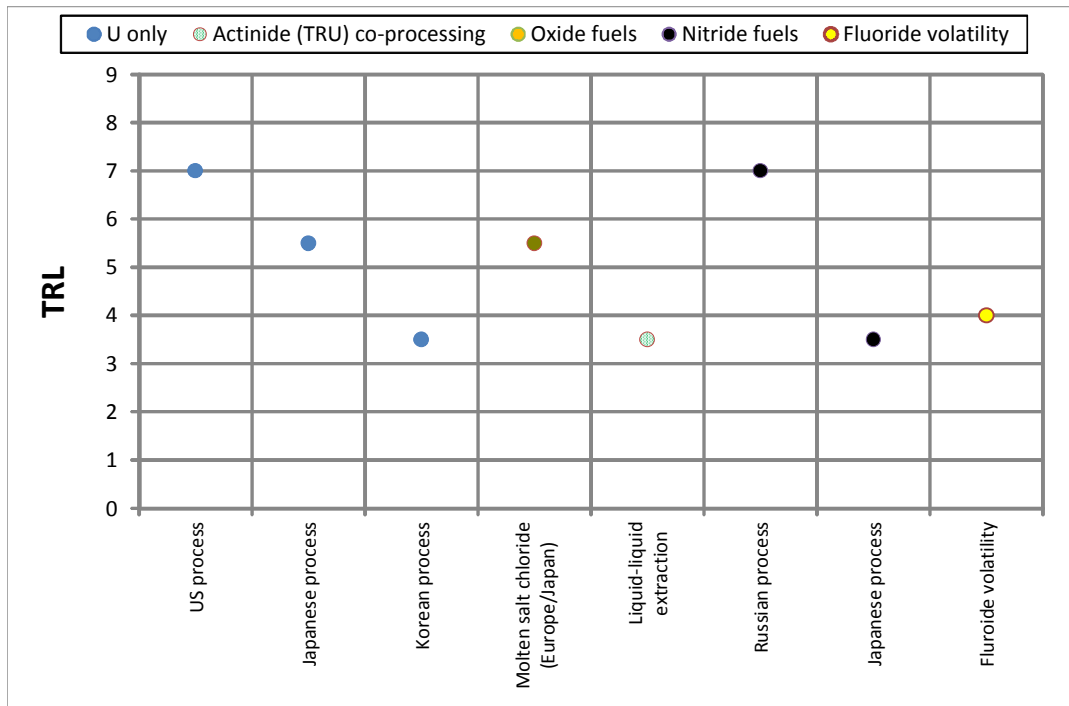


Fig. 12: Summary of TRLs for pyrochemical separation processes NB: Head-end dry (pyrochemical) processes are shown in Figure 10 (adapted from ref.[12])

4.5 Raising TRLs

A range of R&D activities are needed to develop advanced reprocessing and recycling technologies to the point at which they can be considered ready for industrialisation. The TRL scale is a convenient and now widely understood means of tracking the development through the different stages. As seen above (Table 2), the TRL scale moves technologies or process development through proof of concept (TRL 1-3), proof of principle (TRL 4-6) to proof of performance (TRL 7-9). The methodology adopted here (Table 3) indicates how actinide separation process development must also increase in complexity from fundamental studies to flowsheet testing and safety studies before full scale operations. Whilst the experimental studies become more realistic of the industrial process, the materials being studied also must become more representative. So early basic research on simulants or surrogate materials to prove the concept (TRL 3) must evolve into flowsheet testing – firstly with simulated feeds, that include plutonium and minor actinides, before demonstration tests with small quantities of spent fuel to reach ~TRL 6 (proof of principle). Therefore, a chemical process is most efficiently developed by ascending diagonally across this matrix from bottom left to top right corners. In combination with experimental studies, process modelling, flowsheet simulation and engineering design are also needed before demonstration of feasibility. Experimental studies for process chemistry and chemical engineering development in general will utilise different facilities and scales. Lab-scale facilities for the use of radioactive materials including plutonium, minor actinides and small quantities of spent fuels, are required to prove the flowsheet under realistic radiation conditions. Development of equipment and engineering, however, may require larger scale rigs but only in non-active or uranium-active facilities. Computer based models and simulations are powerful methods to accelerate the R&D, reducing the number of costly active experiments to

be done when developing chemical flowsheets and also to bridge the scales between small scale active and large scale non-active testing of engineering and technology, *e.g.* effects on mass transfer. Also, in this middle development phase, process safety studies will be needed to understand the resilience of the flowsheet, ensuring the processes are suitable for ultimate industrialisation. Reaching TRL 6, having addressed chemistry and engineering and proven technical feasibility under realistic conditions at small scales, is the usual R&D goal of advanced reprocessing and recycling programmes that aim to introduce new separation processes for future fuel cycles [151]. Prior to building a commercial scale plant, however, an intermediate stage of a small scale demonstration plant may be an attractive option to fully prove feasibility in a representative environment (TRL 7). The demonstration plant can also be used to disposition legacy or orphan fuels from past operations, as is occurring in the pyro-processing of spent EBR-II fuels at the INL. There are, however, step changes in the investments needed to move from a laboratory scale R&D programme (TRL 1-6) to a pilot plant and eventually commercial operations (TRL 8-9). The future development of these separation processes should also look to innovative technologies that can be imported from the non-nuclear chemical industry and other industries that can deliver cost reductions; these may include on-line analysis and automated process control, robotics or digital systems [168].

So the TRL assessments made in this work provide a useful, or at least relative, scale of the technical maturity of a wide range of separation processes proposed by OECD-NEA member states, at least. In turn, this will assist stakeholders in understanding which options are closest to deployment for various fuel cycle scenarios and what gaps need to be addressed in order to raise technology readiness and underpin decision making. This will be further aided by more targeted and detailed TRL assessments of sub-processes or process stages enabling a deeper understanding of the technical maturity of various options, thus the R&D needs of each and correspondingly prioritisation of resources.

4.6 Comparison with other assessments

Despite TRLs being a widely recognised and oft-quoted system for assessing technological maturity, there are very few similar assessments of separation processes in the published literature. Indeed, this was one motivation for the present study. However, the “SACSESS” (Safety of Actinide Separation Processes) project, funded by the European Union’s Framework Programme 7 [169, 170] reported a roadmapping exercise in 2015 [171]. Their definitions of TRLs were similar to those used in this work, divided into groups: Proof of Concept (TRL 1-3); Proof of Principle (TRL 4-6) and Proof of Performance (TRL 7-9) – see Table 4. Ikeda and co-workers have also made a technology readiness assessment of P&T technologies (fuels, reactors, reprocessing/partitioning), again differentiating between the phases at TRL 1-3, TRL 4-6 and TRL 7-9 [172]. Their definitions, which were based on those defined by the Global Nuclear Energy Partnership (GNEP), are also given in Table 4. The SACSESS definitions are close to those used in this study but those of Ikeda differ substantially at TRL 3-4. At TRL 3 Ikeda requires flowsheet tests with simulant feeds and bench scale batch tests with irradiated materials and for TRL 4 engineering scale tests using simulant materials. Ikeda also introduces a TRL 4’ which, for hydro-processes, requires hot testing flowsheets with solvent recycling. TRL 5 involves engineering scale tests that complete the “development of elemental technologies” whilst TRL 6 demonstrates integrated technologies with hot testing of full scale units. This represents a more demanding ramp up during the proof of principle phase than the scales used by either the SACSESS or present studies. It is explicit in their report that the evaluation considered the quantities and types of materials used in testing as well as the scale of the testing – definitions for bench, lab, engineering and commercial scale tests were tabled. This compares nicely with the present work where these issues were also considered in the evaluation but by the application of our more flexible matrix based approach.

The SACSESS project adopted a three step process to evaluate the TRLs:

1. Mapping of actinide separation processes – identify the main processes developed globally and select the most promising
2. Review the selected processes – compile key data such as process objective; responsible organisations; process description and current status. Five performance indicators were then assessed and compared:
 - a. Elements removed
 - b. Main solvent used
 - c. Number of stages
 - d. Evolution of the waste volume
 - e. TRL
3. Interviews with scientific experts involved in the SACSESS project to refine the evaluation

The Japanese study used a task force of the Research Committee on P&T managed under the auspices of the Atomic Energy Society of Japan. A two-step approach was taken with an initial assessment by the experts of the task force followed by a second review with the Japan Atomic Energy Commission [172].

The list of processes evaluated by SACSESS and Japanese studies is given in Tables 5-6 and the TRLs are compared with the present work where there is overlap. In the SACSESS study, for hydrometallurgical processes the scope was limited to processes that recover minor actinides either in heterogeneous or homogeneous recycle modes (Table 5). (U,Pu) separations, in the heterogeneous recycle scenario, were not considered as it was assumed that the PUREX process (or variations thereof) were industrially mature. A broader perspective was taken with respect to pyro-metallurgical processes because of the way these can be designed to operate in batch mode with electrochemical recovery of different combinations of actinides (Table 6). The authors of this report concluded that a plethora of processes and process variants for minor actinide recycling have been developed. They highlighted that the DIAMEX-r-SANEX combination provides a complete P&T solution and that the ARTIST process, despite a low TRL at present, is promising and should be a priority for further development. In Europe, the current reference processes for minor actinide heterogeneous recycling are the innovative (i-) SANEX (for Am, Cm recovery) and EXAm (for Am only recovery) and the GANEX process for homogeneous recycling – the CEA-GANEX in France and the EURO-GANEX process at the European level [8, 169, 173, 174].

The Japanese study assessed a limited number of hydrometallurgical and dry processes that were being developed in Japan. Aqueous processing (hydrometallurgy) was stated as the preferred technology for oxide fuels in fast reactor reprocessing (homogeneous and heterogeneous recycling modes) and molten salt processes for metal or nitride fuels in fast reactor or ADS-double strata scenarios respectively. For aqueous processes only the separation technologies were assessed whereas for the dry processes the assessment also included the head-end and waste management. Whilst the Japanese study evaluated TRLs for only a few partitioning processes compared to the present work and the SACSESS study, the difference is that they assessed the fuel cycle system, *i.e.* fuel, reactor/ADS and partitioning. Ikeda and co-workers concluded that the FR-homogeneous fuel cycle system using oxide fuel was at TRL 3-4 compared to that with the metal fuel which was at TRL 3. The FR-heterogeneous fuel cycle system using MA oxide targets was assessed to be in the range TRL 2-4 mainly due to the low TRL of the oxide target fabrication; partitioning in this cycle was assessed to be TRL 3. The ADS-double strata concept was assessed at TRL 2-3 with the partitioning again at the higher end compared to the fuel fabrication. Integrated tests – *i.e.* the full cycle of partitioning MA from irradiated fuels →

fuel fabrication → irradiation – at lab scale were proposed by Ikdea *et al.* to accelerate technology system development.

From Tables 5-6 it is clear that some differences exist between the three assessments. Given the large number of separation processes reported in the literature, no study is likely to be able to cover all the variations. The Japanese study generally attributes lower TRLs to the processes due to a much more demanding set of criteria, particularly in the Proof of Principle stage (*i.e.* moving from TRL 3 to TRL 6). The SACSESS study and the present work are similar in style, focusing on a ‘system-level’ assessment of the separation process, and broadly in agreement. The main differences (hydrometallurgy) are in evaluations of the EXAm and ALSEP processes but as these are the current focal points of French and US R&D programmes in MA partitioning, respectively, then it is not too surprising that judgements vary. For pyro-processes, the main disagreement is the assessment of the TRL for electro-refining undertaken in Europe but the TRL attributed to this in the present OECD-NEA study includes all the related progress made in Japan. In conclusion, comparing the actual TRL values between studies is probably not so important as long as each study is self-consistent – rather understanding the relative technological status is the useful measure to assist decision-making and/or prioritisation of resources. Furthermore, the TRL assessment is just one tool in the broader evaluation of separation processes for future closed fuel cycles where factors such as economics, proliferation risk, safety, environmental impacts and waste management are equally or even more important in the down-selection of technologies and prioritisation of R&D funds [151].

Table 4: Summary of TRL definitions for partitioning processes used by the SACSESS project and Ikeda *et al.* [171])

TR L	Function	Horizon 2020 definition (as used in the SACSESS Roadmap)	Ikeda <i>et al.</i>
9	Proof of performance	Actual system prove in operational environment (Industrial process)	
8		System complete and qualified (Pilot scale)	
7		System prototype demonstration in operational environment (Engineering scale)	
6	Proof of principle	Technology demonstrated in relevant environment (Hot tested with larger scale)	Integrated tests of engineering technologies and full scale hot tests of units
5		Technology validated in relevant environment (Hot tested)	Engineering scale test with spent fuels
4		Technology validated in lab (Cold tested)	Laboratory scale continuous operation test using spent fuels (4' or 4+) Engineering scale test with unirradiated material
3	Proof of concept	Experimental proof of concept (Modelled)	Flowsheet based on technological data
2		Technology concept formulated (Demonstrated)	Materialization of concept
1		Basic principles observed (Idea)	

Table 5: Comparison of TRL evaluations made by the SACSESS project, Ikeda *et al.* and the present work [171]): hydrometallurgical separations

Process	SACSESS Roadmap	Ikeda <i>et al.</i>	Present work
TRUEX	5-7		6
DIAMEX	6-7		6
TRPO	4-5		-
ARTIST	2-3		-
DIDPA	5-6	3	-
SETFICS	3-4	3	-
r-SANEX	4-5		4-5
i-SANEX	4-5		3
1c-SANEX	3-4		2-3
CYANEX-301	4-5		-
TALSPEAK	4-5		6
TRUSPEAK	2-3		2
Advanced TALSPEAK (TRUSQUEAK)	2-3		4
ALSEP	2-3		4
EXAm	4-5		6
GANEX (1 st cycle)	6-7		5
CEA GANEX	4-5		4-5
EURO-GANEX	4-5		4-5
CHALMEX	2-3		2
NEXT process (co-crystallisation)		3	-
NEXT process (co-extraction)		3	5-6
NEXT process (extraction chromatography)		3	5
DGA method (Japan)		2	4

Table 6: Comparison of TRL evaluations made by the SACSESS project, Ikeda *et al.* and the present work [171]): pyrometallurgical separations

Process	SACSESS Roadmap	Ikeda <i>et al.</i>	Present work
US electro-refining (U only recovery)	8-9		7
US electro-refining (TRU recovery)	5-6		-
EU electro-refining	2-3		5-6
Liquid-liquid reductive extraction	3-4		3-4
DDP (Dimitrovgrad Dry Process)	8-9		7
ACPF (Korean Process)	3-4		3-4
Japanese electrorefining		3	5-6

Head end (Japan)		2	-
Salt waste treatment (Japan)		2	-
Japanese nitride process (Japan)		3	3-4

5. Conclusions

A major study has just been concluded by the EGFRFC under the auspices of the Nuclear Science Committee of the NEA. This study reviewed progress on separation chemistry processes that are needed for actinide recycling in the future implementation of fully closed fuel cycles. The state-of-the-art in separation processes was described with technology options classified by a hierarchy of separations based on different levels of separation. The overall aim of the study was to assess the technological readiness of the various reprocessing techniques so a fit-for-purpose methodology, based on the well-known TRL scale, was established; this adopted a similar approach to an earlier assessment of innovative fuels made by another Expert Group of the NEA [138]. The approach adopted a matrix style assessment in which information on both the scale of testing and the materials used in testing could be combined to evaluate the TRL achieved. This assessment will help inform international perspectives for future R&D needed to raise technology readiness of recycling options for future closed fuel cycles and ultimately down-selection of technologies as R&D moves towards implementation. The study assessed initial Head-end processes as well as both aqueous based process steps (hydrometallurgy) and dry pyro-chemical processing steps. Although the use of TRLs is widely quoted, there are few similarly broad-ranging studies of technology readiness of actinide separation processes in the published literature. Results from the main alternative study, made under the European SACSESS project [171], were compared to the present work and conclusions were shown to be broadly similar giving added confidence in the findings of this study although it is emphasised that self-consistency within a study is more important to provide the relative understanding of technical maturity of processes. Also, it is re-iterated that the EGFRFC and SACSESS studies both made their assessments at the system level and there may be individual process stages that are at lower TRLs. Thus more detailed analyses of leading options are now required.

Furthermore, the technical performance of the separation processes was shown to be only part of the overall R&D requirements. Proliferation resistance, waste minimisation, environmental impact, process safety, public acceptability and economics all must be considered. Also, given the global uncertainties in how nuclear energy will be utilised in the 21st century, the development of the associated fuel cycles must be sufficiently flexible to address a wide variety of possible scenarios. The review of processes indicates that, whilst more R&D is needed in most cases to raise TRLs, suitable options exist to enable the recovery of all actinide elements plus the HHRs by wet or dry processes, even up to the extraordinarily difficult isolation of americium alone. The technology readiness levels (TRL) for most processes fall in the proof of principle range with TRLs between 4 and 6. That is, flowsheets have been proposed for processes and tested with active materials under increasingly relevant conditions, through to initial hot tests with small quantities of SNF.

The NEA study concluded that *“Although the different processes have not reached the same level of maturity it must be noticed that, as of today, no “showstopper” has been detected that could jeopardize their development at the industrial scale. Further development will clearly be required to bring these separation processes to the point at which they can be industrialised”*.

Consequently, whether decisions are taken to close the fuel cycle and which technologies are to be selected will be most heavily influenced by the national fuel cycle scenarios depending on factors such as reactor and fuel type as well as positions taken on GDF capacities, nuclear non-proliferation, financing and regulation. This study did not consider these aspects in detail although some evaluation was made of the non-proliferation requirements which were shown to be largely due to perceived requirements of key stakeholders rather than technological benefits as all options (including no recycle) will require physical protection and safeguards.

It is to be emphasised that the development of associated back-end fuel cycle technologies must proceed in parallel with advanced fuels and reactor designs. This is obviously essential for sustainable closed fuel cycles

but, overall, the safe, efficient and economic management of spent nuclear fuel and associated wastes are key ingredients to the public acceptability, affordability and technological success of deploying any new reactors.

Given the potential benefits compared to the open fuel cycle, developing options for closing the fuel cycle in the longer term must be supported internationally although it is clear from this study that R&D is still needed to develop separation processes that are ready for industrial deployment and that realise these benefits whilst addressing the perceived risks and down-sides of SNF reprocessing. It is also obvious from the study that greater international co-operation between countries in the development of these processes can accelerate progress. A good example of international collaboration in process development is the succession of European Union funded projects from “NEWPART”, “PARTNEW”, “EUROPART” in Framework Programmes (FP) 4, 5, 6 to “ACSEPT” and “SACSESS” in FP7 and the new “GENIORS” project in the present EU Horizon 2020 programme [8, 173].

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