OVERVIEW OF THE HYDROMETALLURGICAL AND PYRO-METALLURGICAL PROCESSES STUDIED WORLDWIDE FOR THE PARTITIONING OF HIGH ACTIVE NUCLEAR WASTES

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Abstract

For more than 10 years, a revival of the interest for the partitioning of high active nuclear wastes (HAWs) exists worldwide in connection with possible improvements of the management of the HAWs actually produced and with futuristic nuclear fuel cycles. The main aim of the partitioning processes is to separate, from the complex mixtures of HAWs, the long-lived radionuclides (LLRNs), belonging either to the minor actinides (MAs) or to the fission products (FPs) families of elements, in order to prepare fuels and/or targets suitable for their subsequent transmutation (P&T strategy). An other possible strategy consists in the special conditioning of the separated LLRNs into stable dedicated matrices (P&C strategy).

The LLRNs considered for partitioning are the MAs, neptunium, americium and curium, but also the FPs, caesium, technetium and iodine.

Most of the partitioning processes studied so far belongs to the domain of hydrometallurgy, but, recently, a new impetus was observed in the field of pyrometallurgical processes.

The main aim of this talk is to present a brief status of the development of both "hydro" and "pyro" processes for the partitioning of LLRNs developed worldwide, with a special emphasis on the benefits/drawbacks of each process.

1. Introduction

Since the end of the 80s, a renewal of interest is observed worldwide for LLRNs partitioning techniques from nuclear wastes (HAWs). This interest is connected with two main fields:

- The conventional LWR closed fuel cycle using the PUREX process. New management methods for nuclear wastes are considered, the so-called partitioning-transmutation (P&T) and partitioning-conditioning (P&C) scenarios. For this domain, hydrometallurgy is the main route for LLRNs partitioning process development, while pyrometallurgy is also subject of some research.

- "New" fuel cycles associated with the development of fast reactors (FRs), accelerator driven systems (ADSs) and fused salt reactors (FSRs). For this field, pyrometallurgy is the main route considered for spent fuel reprocessing and wastes partitioning, while a small interest still remains for the hydrometallurgical route.

The aim of the present article is to give a brief overview of the progress realised worldwide in the recent years in the field of partitioning of LLRNs by hydrometallurgical and pyrometallurgical processes.

2. Partitioning processes: an overview

2.1 General considerations [1]

2.1.1 Target elements for the separations

Actinides: for P&T and P&C scenarios, the elements considered for partitioning are the so-called minor actinides (MAs): neptunium (Np), americium (Am), and curium (Cm), while for "new" fuel cycles scenarios, uranium (U), plutonium (Pu) and the MAs are all concerned with partitioning/reprocessing process development.

Fission products: for P&T and P&C scenarios, iodine (I), technetium (Tc) and caesium (Cs) are the three main elements considered for partitioning. Some P&C scenarios also consider the partitioning of caesium and strontium.

In Japan, the separation of the platinum group metals (PGMs) from nuclear wastes is also studied for industrial uses of the separated PGMs.

2.1.2 Goals for the separations

The most important goals for the separations are:

- Minimisation of the *long-term radiotoxic inventory* of the wastes conditioned in "conventional matrices", e.g. in glasses (removal of LLRNs of MAs and FPs families).

- Minimisation of the *heat load* of the conditioned wastes (removal of ^{137}Cs and ^{90}Sr).

- More often, *high purities* of the separated LLRNs are required, for target or fuel fabrication for subsequent transmutations of these LLRNs.

2.1.3 Consequences

Owing to the fact that efficient separation methods are needed with low losses of LLRNs and high purities of the separated LLRNs, *multi-stage processes are most often necessary.*

2.2 Hydro processes for actinides and FPs partitioning

2.2.1 Examples of separation strategies

Examples of separation strategies are given for some countries and some research organisations.

2.2.1.1 Japan

- JNC
  For FRs closed fuel cycle, JNC develops an integrated approach based on hydrometallurgical steps including the: (i) dissolution of MOX FR spent fuels with an aqueous nitric acid solution, (ii) iodine volatilisation, (iii) electrolytic extraction of technetium (Tc), palladium (Pd) and selenium (Se), (iv) crystallisation of most of the uranium contained within the spent fuel dissolution liquor in the form of hydrated uranyl nitrate, (v) single PUREX extraction step for recovery of remaining U+Pu+Np, (vi) SETFICS process for Am and Cm partitioning.

- JAERI
  JAERI proposed to separate Np and Tc during the implementation of the PUREX process. This organisation develops also, since many years, the so-called four-group partitioning process for the treatment of the wastes issuing the reprocessing by the PUREX process of UOX or MOX LWR spent fuels. This partitioning process includes the following steps: (i) MAs partitioning (Np, Am and Cm), (ii) Cs+Sr extraction, (iii) PGMs extraction. The remaining mixture of wastes constitutes the 4th category of elements of the initial mixture treated.

2.2.1.2 USA

The situation in the USA is peculiar because partitioning processes developed concern the treatment of defence wastes in particular those accumulated at DOE's Hanford site during the cold war. Several processes were developed for the partitioning of radionuclides from the wastes: (i) TRUEX process for transuranic extraction, (ii) SREX process for Sr removal and (iii) CSEX process for Cs extraction. It should be also noted that in 1999, a Report named *A Roadmap for Developing Accelerator Transmutation of Wastes (ATW) Technology* was published by the DOE [2], which considers the possible treatment of the LWR spent fuels accumulated in the USA in order to separate: (i) U for final disposition as low level waste and (ii) TRUs for burning in ATW systems. The processes considered for these separations are: (i) the UREX process, which consists in a modified PUREX process aiming to only extract U, (ii) pyrometallurgical partitioning process for TRU separation from the UREX wastes and for the ATW fuel cycle.

2.2.1.3 France (CEA)

The strategy developed by the CEA for partitioning the nuclear wastes of LWR closed cycle concerns 6 LLRNs to separate from the wastes: 3 MAs (Np, Am and Cm) and 3 FPs (I, Tc and Cs). This strategy is based on the development of successive liquid-liquid extraction processes: (i) the improved PUREX process for U, Pu, Np, I and Tc separations, (ii) the DIAMEX process for trivalent Am+Cm extraction (FP lanthanides (III), Ln, are co-extracted), (iii) the SANEX process for

Am+Cm/Ln(III) separation, (iv) the SESAME process for Am/Cm separation, (v) the CALIX-CROWN process for Cs separation.

In some organisations (e.g. in USA, Japan, Czech Republic or Russian Federation), instead of developing a succession of separation processes for peculiar LLRNs, the integration of processes for MAs and FPs extraction are studied. For example, the use of a solvent containing a mixture of cobalt dicarbollide+dioxide of diphosphine allows the combined extraction of Cs+Sr+(Am+Cm)+Ln.

2.2.2 Minor actinides partitioning

One cycle processes

- DIDPA process (JAERI, Japan)

  This process for MAs partitioning is based on the use of di-isodecylphosphoric acid (DIDPA). The extraction mechanism is the following:

  $$M^{n+} + \overline{n\,(HA)_2} \Leftrightarrow \overline{M(HA_2)_n} + n\,H^+ \tag{1}$$

  The separation of the TRU elements is done by successive stripping from the loaded solvent, including the use of diethylenetriaminopentaacetic acid (DTPA) complexing agent for actinides(III)/Ln(III) separation (TALSPEAK like process, vide infra).

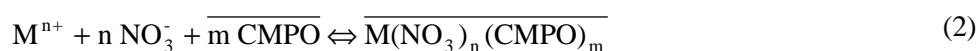 The DIDPA process was recently tested successfully in the BECKY hot-cell at NUCEF (JAERI, Tokai-Mura).

  Among the possible drawbacks of this process one can mention the: (i) required feed acid adjustment, (ii) solvent degradation and its delicate clean-up, (iii) limited solvent loading with metal ions.

- SETFICS (JNC, Japan)

  The SETFICS process constitutes a modification of the TRUEX process (vide infra) based on the use of the extractant di-isobutyl-phenyl-octylcarbamoylmethylphosphine oxide

  $$(\Phi C_8 H_{17} P(O) CH_2 C(O)(i\text{-}C_4 H_9)_2 = CMPO)$$

  The extraction mechanism is as follows:

  $$M^{n+} + n\,NO_3^- + \overline{m\,CMPO} \Leftrightarrow \overline{M(NO_3)_n(CMPO)_m} \tag{2}$$

  The separation of TRUs is done by successive stripping from the loaded solvent, including also the use of DTPA for An(III)/Ln(III) separation. This process has not been tested yet with genuine HAWs. The possible drawbacks of this process are: (i) the limited stripping efficiency, (ii) the management of salts and DTPA containing effluents.

- PALADIN (CEA, France)

  This process is based on the use of a mixture of extractants: a malonamide (DIAMEX process extractant)+di-ethylhexylphosphoric acid (HDEHP), the extractant of the TALSPEAK process.

The extraction and separation mechanisms are the following:

Extraction:

$$M^{3+} + 3NO_3^- + \overline{2\,DIAM} \Leftrightarrow \overline{M(NO_3)_3(DIAM)_2}$$ (3)
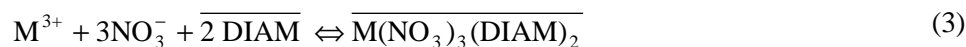
An(III)/Ln(III) separation: done after contacting the loaded solvent with a pH adjusted aqueous solution containing DTPA selective trivalent actinide complexing agent. For pH range HDEHP is the extractant, while at the metal nitrate extraction step, carried out with acidic feeds 3 to 5 mol/L in nitric acid, trivalent An and Ln are extracted with the malonamide.

This process was recently successfully tested in the ATALANTE facility (CEA/Marcoule, France).

The possible drawbacks of this process are the: (i) necessity to use 2 extractants, (ii) need of pH adjustment, (iii) co-extraction of numerous ions, (iv) solvent clean-up not yet defined.

Multi-cycle processes

- 1st step: An+Ln co-extraction

TRUEX (USA, Japan, Russian Federation, Italy and India)

This process is based on the use of the CMPO extractant. This process was developed by Horwitz (ANL) and Schulz (Hanford) in the USA in the 80s. The advantages of the TRUEX process are the following: (i) it can extract An (and Ln) salts from acidic feeds, (ii) its efficiency has been demonstrated with genuine HAWs, (iii) a large experience has been gained worldwide. The main drawbacks of the TRUEX process are the: (i) necessity to use large concentration of tri-n-butylphosphate (TBP) as solvent modifier added to the solvent to prevent third phase formation, (ii) stripping of metal ions which are not so efficient, (iii) delicate solvent clean-up.

DIAMEX (France, Italy, Germany, Europe, Japan, USA and India)

This process is based on the use of a malonamide extractant. The main interests of the process are: (i) An (and Ln) salts can be extracted from acidic feeds, (ii) its efficiency has been demonstrated on genuine HAWs, (France, Europe), (iii) no secondary solid wastes generated owing to the CHON character of the malonamide extractant. Its main drawback relies in the partial co-extraction of palladium (Pd) and ruthenium (Ru) with the MAs.

A process based on a new type of diamide, a diglycolamide (DGA), which is a terdendate ligand having better affinity for An(III) than the malonamide, is under development at JAERI (Tokai, Japan).

TRPO (INET, Tsinghua University, China)

The TRPO process is based on the use of a mixture of tri-alkyl phosphine oxides (R3P(O), with R = alkyl groups) as extractant. This process has been tested successfully in China with genuine HAW. Its main drawbacks concern the necessity: (i) to adjust the feed acidity, (ii) to use a concentrated nitric acid solution for An(III)+Ln(III) stripping, which complicates the subsequent An(III)/Ln(III) partitioning step.

- 2nd step: An(III)/Ln(III) separation

TALSPEAK and CTH processes

The TALSPEAK process, developed at ORNL (USA) in the sixties and then adapted (CTH process) at Chalmers University, Göteborg, Sweden, can be considered as the reference process

for An(III)/Ln(III) group separation. It is based on the use of HDEHP as extractant and DTPA as the selective An(III) complexing agent. The An(III)/Ln(III) separation is performed by the selective stripping of An(III) from the HDEHP solvent loaded with the mixture of An(III)+Ln(III) under the action of an aqueous solution containing DTPA and an hydroxocarboxylic acid, like lactic, glycolic or citric acids. The advantages of this process are: (i) the large experience gained worldwide, (ii) its good efficiency. Among the main drawbacks one can cite: (i) the necessity to adjust the pH of the feed, (ii) the limited solvent loading of metal ions, (iii) the difficult solvent clean-up.

SANEX concept (acidic S-bearing extractants)

– CYANEX 301 process (China, USA, Germany)

The CYANEX 301 extractant consists in a dialkyldithiophosphinic acid (R2PSSH, with R = an alkyl group). Its use for An(III)/Ln(III) was first proposed by Zhu at Beijing (China) in 1995. The main interest of the process relies in: (i) the large efficiency for An(III)/Ln(III) separation, (ii) the fact that the process has been tested with genuine An(III)+Ln(III) mixtures. Nevertheless, for an efficient use of this process the feed solution should be adjusted to pH 3 to 5, which is not so easy to carried out industrially. Moreover, the solvent clean-up is also a weak point.

– ALINA process (Germany)

To cope with the main drawbacks of the CYANEX 301 process mentioned above, Odoj and Modolo at Jülich (Germany) proposed the use of a syngergistic mixture made of bis(chlorophenyl)dithio-phosphinic acid ((ClΦ)2PSSH)+tri-n-octylphosphine oxide (TOPO) to perform the An(III)/Ln(III) group separation. If the separation factors between An(III) and Ln(III) are less than those observed with CYANEX 301, the concentration of nitric acid in the feed can be as high as 1.5 mol/L, which makes the ALINA process more attractive than the CYANEX 301 one. The ALINA process was successfully tested with genuine wastes. The possible drawbacks of this process are: (i) the solvent clean-up process not yet defined, (ii) the generation of P- and S-bearing wastes (from the degraded extractants) which should be managed.

SANEX concept (neutral N-bearing extractants)

– BTPs (Germany, France, Europe)

After the discovery by Kolarik at FZ Karlsruhe (Germany) of the astonishing properties of the bis-triazinyl-1,2,4-pyridines (BTPs) for An(III)/Ln(III) separation, a process was readily developed and tested in the frame of the European so-called NEWPART project [3]. Successful hot tests were achieved both at the CEA/Marcoule and at the ITU in Karlsruhe using the n-propyl-BTP. Large efficiency of the BTP process was obtained. One should mention also that the feed of the n-propyl-BTP process can be acidic ([HNO$_3$] = 1 mol/L). Nevertheless, even if this system seems very promising, an instability of the n-propyl-BTP extractant was observed. As a consequence, efforts are underway at the CEA to modify the solvent formulation to suppress this major drawback.

– TMAHDPTZ+octanoïc acid (CEA, France)

A synergistic mixture made of the terdendate N-ligand, 2-(3,5,5-trimethylhexanoylamino)-4,6-di-(pyridin-2-yl)-1,3,5-triazine (TMAHDPTZ), and octanoïc acid was developed at

CEA/Marcoule. A process flowsheet was defined and successfully tested with genuine effluent with good efficiency. The main drawbacks of this process are: (i) the required pH adjustment of the feed, (ii) the management of secondary wastes not yet defined.

- 3rd step: Am/Cm separation

For this step, processes based on the selective oxidation of Am at the +VI or +V oxidation states are developed, the curium remaining unchanged as Cm(III), allowing simple Am/Cm separation processes to be defined.

SESAME process (CEA, France, Hitachi, Japan)

In strong oxidising conditions, Am can be oxidised from Am(III) to Am(VI). This can be done, for example, by electrolysis in the presence of heteropolyanions (HPA) acting as catalyst. The so-generated Am(VI) can be separated from Cm(III) by extraction, for example by TBP. This is the principle of the so-called SESAME process developed at CEA/Marcoule. At Hitachi (Hitachi city, Japan), oxidation of Am to Am(VI) is obtained by the use of ammonium persulphate. Then, Am(VI) is extracted by TBP. The SESAME process exhibits a great efficiency for Am/Cm separation. A large experience was obtained at the CEA at pilot scale during the last twenty years with a SESAME like process (kg amounts of ^{241}Am were purified). Nevertheless, the industrialisation of the process is faced with difficulties such as: (i) the instability of Am(VI), (ii) the non-easiness to develop a multi-stage process, (iii) the generation of secondary solid waste (made of HPA constituents).

Am(V) precipitation (JNC, Japan)

The selective precipitation of double carbonate of Am(V) and potassium (K) is one of the oldest method for Am/Cm or Am/Ln separations, developed at the end of the 60's in the USA. This method requires the use a 2 mol/L K_2CO_3 solution in which the mixture of Am(III) and Cm(III) is dissolved. After chemical or electrochemical oxidation of Am(III) to Am(V), Am(V) precipitates from the solution as the solid crystalline $K_5AmO_2(CO_3)3$ nH_2O, while Cm(III) remains in solution. After filtration, Am is separated from Cm. This process: (i) is simple, (ii) is selective for Am, (iii) has been largely used worldwide. The process main drawbacks are: (i) the Am losses with Cm, which are not so low, (ii) the fact that it exists only one stage for the process, (iii) the large amounts of secondary wastes generated.

2.2.3 Fission products partitioning

Iodine (^{129}I)

The separation of iodine is done just after the spent fuel dissolution step within the PUREX process. Oxidation of iodide ion, I^-, into elemental iodine (I_2) induces its transfer to the dissolver off-gases (DOGs) where iodine is recovered through DOGs basic washing. To recover most of the iodine spent fuel inventory at that step, slight improvement of the efficiency of the transfer of iodine from the dissolution liquor to the DOGs seems required.

Technetium (^{99}Tc)

The soluble fraction of Tc contained in the spent fuels exists in the dissolution liquor as Tc(VII) (TcO_4^-). Its co-extraction with Zr(IV), then U(VI) and Pu(IV), by TBP is well known. So, for example, the separation of the Tc soluble fraction is achieved through a solvent special scrubbing step in the course of the implementation of the PUREX process at COGEMA La Hague reprocessing plants. If high Tc partitioning yield is required, the main problem concerns the recovery of the Tc fraction that is

contained within the solid insoluble residues remaining after spent fuel dissolution. A special process is required for this Tc recovery, which actually does not exist.

Caesium and strontium or caesium alone

Many processes were developed worldwide in this field, including the use of:

– Inorganic sorbents, like for the JAERI's 4 group partitioning process.

– Crown-ether extractants, like for the SREX and CSEX processes developed in the USA (ANL).

– Cobalt dicarbollide extractants, as developed in Czech Republic, Russia and Western Europe.

– Calix-crown extractants, as developed in France, Western Europe and in the USA.

Most of these processes were successfully tested with radioactive effluents.

2.3. Pyro processes for actinide partitioning [4]

2.3.1 Selected media and possible separation techniques

Selected media

Most of the "pyro" processes developed so-far are based on the use of one or two of the following high temperature liquid phases:

- Fused salts. The most popular fused salts studied are:

  – Molten chloride eutectic, such as LiCl+KCl.

  – Molten fluoride eutectic, such as $LiF+CaF_2$.

- Fused metal, such as Cd, Bi, Al, etc.

Separation techniques

To partition the actinides contained within the fused salt baths, three main techniques are studied and developed:

- Actinide electrodeposition on solid (pyrographite or metal) or liquid metal cathodes.

- Liquid-liquid extraction of actinide between fused salt bath and a metal bath containing a reductive metal solute (Li for example).

- Actinide oxide precipitation from the fused salt under the proper control of the oxygen thermodynamic activity within the salt bath.

2.3.2 Examples of strategies and "pyro" processes

2.3.2.1 USA (ANL, Chicago)

A "pyro" process was developed at ANL in relation with the treatment of FR metallic fuels (EBR II's type) for stabilisation of these Na bonded fuels. The aims of the process is limited. It consists in the separation of the spent fuel into three major fluxes: (i) most of the uranium as a low level waste,

(ii) cladding+noble metals+Zr as metallic waste, (iii) TRUs+FPs+Na+salt incorporation into a zeolithe matrix in order to obtain a ceramic waste after hot pressing. The key step consists, after the oxidative dissolution of the spent metallic fuel in LiCl+KCl eutectic bath at 500°C, into the separation of most of the uranium by electrorefining on a solid cathode. A demonstration campaign involving the treatment of 100 core assemblies (0.4 ton of spent HEU) and 25 blanket assemblies (1.2 tons of spent depleted U) was successfully carried out at Argonne West in the recent years. License for pyroprocessing the whole EBR II spent fuel inventory was obtained recently.

2.3.2.2 Russian Federation (RIAR, Dimitrovgrad)

The pyro-process developed at RIAR concerns the treatment of spent oxide fuels (UOX and MOX) in order to recover U and Pu for MOX fuel re-fabrication by the vibro-compaction process.

The spent oxide fuel is dissolved by chloration in a Li, Na, K, Cs chloride fused salt bath at 650-700°C. Separation of U, Pu or mixture of U+Pu from the salt bath can be obtained by electrodeposition or precipitation. For example:

- U can be separated as UO_2 (which is a good electric conductor) by electrodeposition on a cathode made of pyrographite, while chlorine gas is generated at the anode.

- As PuO_2 is a bad electric conductor, it cannot be electrodeposited on solid cathode. But PuO_2 can be selectively separated by precipitation after bubbling a mixture of Cl_2+O_2 gases into the fused salt bath.

- Under the addition of a mixture of Cl_2+O_2 gases into the fused salt bath, which stabilises Pu as oxychlorides, electrolysis generates a deposit of $(U,Pu)O_2$ onto the pyrographite cathode while chlorine gas evolves at the anode.

An important experience with spent fuel pyroprocessing has been obtained at RIAR with the treatment of:

- 3.3 kg of UO_2 spent fuel (1% burn-up) from the VK-50 reactor, done in 1968.

- 2.5 kg of UO_2 spent fuel (7.7% burn-up) from the BOR-50 reactor, done in 1972-73.

- 4.1 kg of $(U,Pu)O_2$ spent fuel (4.7% burn-up) from the BN-350 reactor, done in 1991.

- 3.5 kg of $(U,Pu)O_2$ spent fuel (21-24% burn-up) from the BOR-60 reactor, done in 1995.

2.3.2.3 Japan

- CRIEPI

  The "pyro" process developed at CRIEPI concerns both the treatment of spent fuels from LWRs (oxide fuels) or FRs (metallic fuels) and the partitioning of TRU elements from the wastes issuing the reprocessing of spent LWR fuels by the PUREX process. The fused salt selected is the LiCl+KCl eutectic in which the spent fuels or the oxides of high active wastes are dissolved by a carbo-chloration technique. After dissolution, U can be electrodeposited as a metal on a solid cathode, then the TRUs can be recovered by electrolysis using a liquid cadmium or bismuth cathode. CRIEPI is also studying the partitioning of MAs by liquid-liquid extraction using a LiCl+KCl salt bath and Cd or Bi metallic solvents containing Li as a reducing agent. In this case, liquid-liquid extraction corresponds to the reductive transfer of a metal from the salt bath, where it exists as M^{n+} cation, to the metal solvent, where M exists as a M^0 solute.

The equation of the extraction reaction can be written as follows:

$$M^{n+}_{(salt)} + n\,Li^0_{(Cd\ or\ Bi\ metal\ phase)} \Leftrightarrow M^0_{(Cd\ or\ Bi\ metal\ phase)} + n\,Li^+_{(salt)} \qquad (4)$$
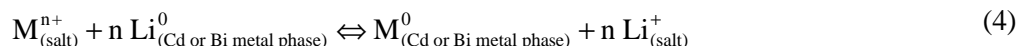
Large expertise has been gained by CRIEPI in this field but only with surrogates of actinides. A joint CRIEPI-ITU programme is under way to test the process with actinides.

- JAERI

    JAERI is studying pyroprocessing for the possible treatment of nitride, oxide or metallic spent fuels in order to prepare nitride fuels enriched with ^{15}N for FRs. After dissolution of the spent fuels into a LiCl+KCl eutectic salt bath, the actinides will be electrodeposited on solid or liquid (Cd) cathodes. The recovered actinide metals will then be converted into actinides nitrides after their dissolution in liquid cadmium. The nitruration agent will be N_2 or Li_3N.

- JNC

    JNC is also engaged in the development of pyro processes aiming to reprocess FR spent fuels. The method selected are similar to those studied by CRIEPI: (i) choice of LiCl+KCl eutectic bath, (ii) electrodeposition method, (iii) liquid-liquid extraction between salt bath and liquid Cd metal.

2.3.2.4 France

Two years ago, the CEA has launched a programme dedicated to the partitioning of MAs by "pyro" processing. A research team was created at CEA/Marcoule and special hot facilities have been created. The programme selected is rather wide. It will consider both chloride and fluoride melts and the most important separation techniques known to be effective in "pyroprocessing", i.e. (i) electrodeposition, (ii) oxide precipitation, (iii) liquid-liquid extraction between fused salt bath and a metallic solvent. The results obtained to-date concern mainly the basic understanding of the chemistry of actinides (U, Pu and Am) in solution in the fused melts. Process developments are also underway and active tests on irradiated objects are foreseen to be done before 2005.

2.3.2.5 Czech Republic

At Rez Institute, Czech scientists are developing a process based on the dual use of actinide hexafluoride volatilisation and pyroprocessing of the wastes from a fluoride melt. This research programme is connected with the interest of Czech Republic for the development of the molten salt reactor (MSR) technology. Facilities are under construction at Rez Institute for testing the pyro-processes.

3. Conclusions and perspectives

3.1 Conclusions

Numerous concepts have been consolidated or newly developed during the last few years, both in "Hydro" and "Pyro" processing of HAWs or spent fuels and targets for "new" nuclear systems.

Tests on "real objects" were carried out successfully in several countries, including the EBR II demonstration test at Argonne-West (UA) on pyroprocessing of FRs spent fuels.

In the domain of "Hydro", blooming of concepts is observed. Multi-step processes look promising but most of the systems developed so far appear complex. Efforts to simplify the processes seem required.

In the domain of "Pyro", strong consolidations of "old concepts" were obtained, including fluoride volatilisation.

3.2 Perspectives

- Hydro

    It seems important to work in order to increase the "simplicity" and "compacity" of the MAs and LLFPs separation processes. Some routes for improvement can be proposed:

    – One cycle process.

    – Consideration of High Active Concentrates instead of High Active Raffinates for process development (large volume reduction factor).

    – Integration of MAs and LLFPs separation processes.

    – Consideration of possibly new LLFPs for partitioning.

    – Maintaining alive the "CHON principle" (minimisation of secondary solid wastes).

- Pyro

    Directions for improving the processes appear to be:

    – Minimisation of TRU losses in wastes and increase of the purities of the separated.

    – Actinides which can be obtained through the combined use of several separation techniques and multi-stage techniques.

    – The waste problem, which is mostly corrosion related owing to the aggressive.

    – Character of the media and the high process temperatures, needs to be precisely estimated.

    – Consideration of the possible separation of LLFPs.

- Collaborations

    It seems a pressing necessity to maintain, or best to increase, the collaborations in this complex field at:

    – National levels: maintain or create network(s) between academic and applied research bodies. As example in France it exist two networks working under the auspices of the December 1991 Nuclear Waste Act: the so-called PRACTIS and NOMADE Groupes de Recherches.

    – Bi-national levels: numerous collaborations exist, e.g. CRIEPI-ANL, CEA-JNC, CEA-JAERI, etc.

    – Regional level. As example at the European level it exists common works partly financed by the EU, e.g. the PARTNEW, CALIXPART and PYROREP programmes within the 5th FWP of EU (2000-2003). The role of ITU at Karlsruhe is also very important for European and wider collaborations,

– At the International level, the roles of OECD/NEA for Workshops and Working Parties managements and also of IAEA appear essential.

So, within a few years, one predicts that a large array of robust "hydro" and "pyro" processes will be available for the definition of new scenarios for the management of nuclear wastes generated through LWRs and FRs closed fuel cycles, but also for the fuel cycles of futuristic nuclear systems, such as the MSRs or the ADSs.

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