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Status of Developments in the Back End of the Fast Reactor Fuel Cycle

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STATUS OF DEVELOPMENTS IN THE BACK END OF THE FAST REACTOR FUEL CYCLE

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Marketing and Sales Unit, Publishing Section
International Atomic Energy Agency
Vienna International Centre
PO Box 100
1400 Vienna, Austria
fax: +43 1 2600 29302
tel.: +43 1 2600 22417
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FOREWORD

The potential of a fast reactor (FR) with a closed fuel cycle for breeding fissile isotopes of plutonium (^{239}Pu and ^{241}Pu) from fertile ^{238}U and, in turn, exploiting the virtually inexhaustible energy locked in natural uranium, was realized from the very inception of nuclear energy. However, to date, FR technology has not matured enough to be economically competitive with water cooled and gas cooled thermal reactors. The present generation of nuclear power reactors is dominated mainly by light water cooled reactors and, to a limited extent, by pressurized heavy water reactors. These reactors mostly use uranium oxide fuel, containing 0.7–5% ^{235}U (the rest being ^{238}U) on a ‘once through’ basis, where only 1% (or even less) of the natural uranium resources are utilized. The rest of the uranium, mostly ^{238}U, is locked in the tailings of enrichment plants or in spent nuclear fuel (SNF). Management of the back end of the nuclear fuel cycle is becoming increasingly important for long term sustainability of nuclear energy. The plutonium by-product from SNF is best utilized in an FR, where more Pu could be bred from ^{238}U than consumed in fission. Thus, by multiple recycling of Pu with depleted, natural or reprocessed uranium, at least 60% of natural uranium resources could be utilized.

In recent years, there has been renewed interest in research and development activities on the partitioning and transmutation of SNF and the FR fuel cycle. Both aqueous and pyrochemical processes are being studied, with the common goal of ensuring ‘proliferation resistance’ by avoiding a separated fissile material stream during partitioning, in addition to safety enhancement and cost minimization. In some countries, the main motivation of the FR fuel cycle is recycling Pu in combination with ^{238}U for breeding additional Pu fuel. In other countries, advanced partitioning techniques are being developed for separation of Pu and minor actinides (MAs) (Np, Cm, Am), for burning them in FRs in order to reduce the long term radiotoxicity of SNF. The IAEA’s International Project on Innovative Nuclear Reactors and Fuel Cycles (INPRO) and the Generation IV International Forum, initiated by the United States Department of Energy, have also identified the importance of the sodium cooled fast reactor (SFR) and the related fuel cycle for multiple recycling of Pu and MAs.

On the recommendation of the IAEA Technical Working Group on Nuclear Fuel Cycle Options and Spent Fuel Management (TWGNFCO) and the INPRO Steering Committee, the IAEA initiated work on “FR fuels and fuel cycle technology”. A technical meeting on the Current Status and Future Prospects of Liquid Metal Cooled Reactor Fuel Cycle (TM-LMRs-2005) was organized in October 2005 in cooperation with IPPE, Obninsk, the Russian Federation. Thereafter, as a part of the INPRO activity, a joint study on the FR and closed fuel cycle was taken up with participation of experts from China, France, India, Japan, the Republic of Korea and the Russian Federation. This publication is based on presentations on the FR fuel cycle in these two forums. It is intended to cover exhaustively the main and emerging technologies on ‘partitioning’ and other issues related to the back end of the SFR fuel cycle.

The IAEA wishes to express its gratitude to all contributors and reviewers of this publication. Particular thanks are due to B. Raj (India) for making significant contributions and compiling the initial draft. The IAEA officers responsible for this publication were C. Ganguly, H.P. Nawada, F. Depisch, Y. Busurin and U. Basak of the Department of Nuclear Energy.

EDITORIAL NOTE

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1. INTRODUCTION

1.1. BACKGROUND

Currently, 439 nuclear power plants are operating in 30 countries and are contributing approximately 14% of global electricity generation. Of the 30 countries using nuclear power for electricity generation, 24 intend to allow new plants to be built, and, of those, the majority actively support the increased use of nuclear power, some by providing incentives. In addition, a growing number of countries are expressing interest in introducing nuclear power. The IAEA's projections indicate the world total for nuclear electrical generating capacity to be between 437 and 542 GW(e) by 2020, and between 473 and 748 GW(e) by 2030 [1]. Growth in world energy production and consumption in the next three decades is projected to be 65% higher than the growth achieved in the past 30 years with the present energy policies adopted around the world by different countries [2].

A 'nuclear renaissance' is anticipated by some observers in several countries due to the limited availability of fossil fuel resources and the need to reduce carbon dioxide emissions. Moreover, it is expected that the power industry will develop into a free market environment in the 21st century. The development of the entire power industry and nuclear power as its integral part will be influenced by the following main factors: economics, safety, radioactive waste management, non-proliferation of nuclear weapons grade material, restructuring of the electricity market, the changing structure of power resources, and environmental issues.

The growth of nuclear power will also depend on the status and maturity of nuclear fuel cycle technologies. Parallel to the renaissance of nuclear energy, the concept of the 'closed fuel cycle' is also receiving close attention. It has now been accepted that reprocessing of spent fuel and recycling of separated fissile material are essential for the effective utilization of resources and for reducing the environmental impact of nuclear power by reducing the requirements for waste repositories. Accordingly, several countries have renewed their emphasis on the closed fuel cycle as a route to sustainability of nuclear energy. There is now great interest in effective utilization of uranium as well as partitioning and transmutation (P&T) of transuranic (TRU) elements. Radioactive waste management is a key aspect which has to be resolved to assure growth of nuclear power. This is especially essential in the context of public concerns with regard to radiotoxicity of nuclear waste containing long lived radioactive isotopes.

1.2. REQUIREMENTS FOR FUTURE NUCLEAR ENERGY SYSTEMS

In the future, large scale deployment of nuclear power cannot be implemented on the basis of current technologies, considering that accessible resources of uranium are limited. New generation reactors and fuel cycle technologies should meet the following objectives:

- Efficient utilization of the available uranium (and in the future, thorium) resources without restricting their use to only fissile (^{235}U) content;
- Inherent safety, i.e. exclusion of accidents with high radioactivity release by design features and/or operational strategies of the involved technologies;
- Capability to reduce the radiotoxicity of disposed radioactive waste to a level equivalent to that of natural uranium;
- Enhanced resistance to proliferation of weapons grade nuclear materials;
- Competitive cost of new nuclear power systems in comparison with existing reactor systems and alternate energy production technologies.

In summary, the design of reactors and fuel cycles should guarantee an enhanced level of operational safety, facilitate radioactive waste management and non-proliferation of weapons grade nuclear material, and improve the economy of nuclear power in order to enhance the support of the public and investors. Future nuclear plants with innovative concepts should be based on robust and closely integrated technologies, including their respective nuclear fuel cycles. Although several countries have not adopted the closed fuel cycle for reasons relevant to their national policies, it is quite natural to predict that many countries will, in the future, adopt the closed fuel cycle since it meets several of the above objectives. The fast reactors (FRs) deployed so far have been technology demonstrators. With sufficient maturity and experience gained through operation, future innovative FRs are expected to combine the key objectives with a prime focus on economy which will make them more attractive.

1.3. INTERNATIONAL NUCLEAR INITIATIVES

Two international projects are being pursued in parallel for reactors and fuel cycles of the future nuclear energy system. One is the Generation IV International Forum initiated in 2000 [3], and the other is the International Project on Innovative Nuclear Reactors and Fuel Cycles (INPRO) initiated in 2001 [4]. Both projects aim at promoting the development of advanced nuclear power technologies which may form the basis for sustainable growth of the power industry in the 21st century.

Different FR systems, such as sodium cooled FRs (SFRs), lead cooled FRs and gas cooled FRs, are considered under the Generation IV initiative as promising options for development and deployment around 2030–2050. Under INPRO, a joint case study on the closed nuclear fuel cycle with FRs has identified a broad set of parameters of innovative SFRs that could meet the objectives of sustained nuclear energy growth.

1.4. FAST REACTORS

The following two major advantages offered by FRs can encourage Member States to opt for FR systems:

- In principle, uranium can be used in FRs almost completely by conversion to plutonium.
- FRs can be used as plutonium and minor actinide (MA) utilization machines; this is especially important considering the present day huge stockpiles of weapons grade and reactor grade plutonium as well as plutonium contained in spent nuclear fuel (SNF) of thermal reactors. FRs could also be useful for transmuting long lived radionuclides (LLRNs).

1.5. ACCELERATOR DRIVEN SUBCRITICAL SYSTEMS

The nuclear fuel cycle for accelerator driven subcritical systems (ADSs) is not conceptually different from the fuel cycle of reactor systems. Although the ADS would allow accelerated utilization of MAs in fuel by fission, this could also be used for producing energy and breeding fissile material. Several studies have been performed for the application of ADS for MA transmutation [5–8]. MA transmutation in ADSs has been studied primarily in the context of a double strata scenario, where the bulk of plutonium is recycled in commercial reactors and only small amounts of MAs are transmuted in dedicated second stratum systems.

1.6. OPEN AND CLOSED NUCLEAR FUEL CYCLES

Many countries decided to adopt the once through fuel cycle in the past for either economic considerations or for fear of proliferation of nuclear materials. Only a few countries, such as France, Japan, India, the Russian Federation and the United Kingdom have continued to reprocess spent fuel. However, there is currently widespread interest in the world for adopting closed fuel cycles. In the case of adoption of FR technology, the use of a closed fuel cycle with multiple recycling of fissile material is absolutely essential, as SNF becomes a raw material for reuse of the bred fissile content, and the remaining fissile content after irradiation remains too high to be treated as a waste. Contrary to the current situation with thermal reactor systems, where the choice between the once through and closed cycle may be justified, the FR concept is not sustainable without a closed fuel cycle.

The existing technologies and operational experience gained in the reprocessing of oxide fuels from thermal power reactors may be an excellent starting point for new reprocessing technologies adjusted to the specifics of FR oxide fuel. At the same time, there are considerable differences arising out of the use of high plutonium content fuels and the higher burnup achievable in FRs.

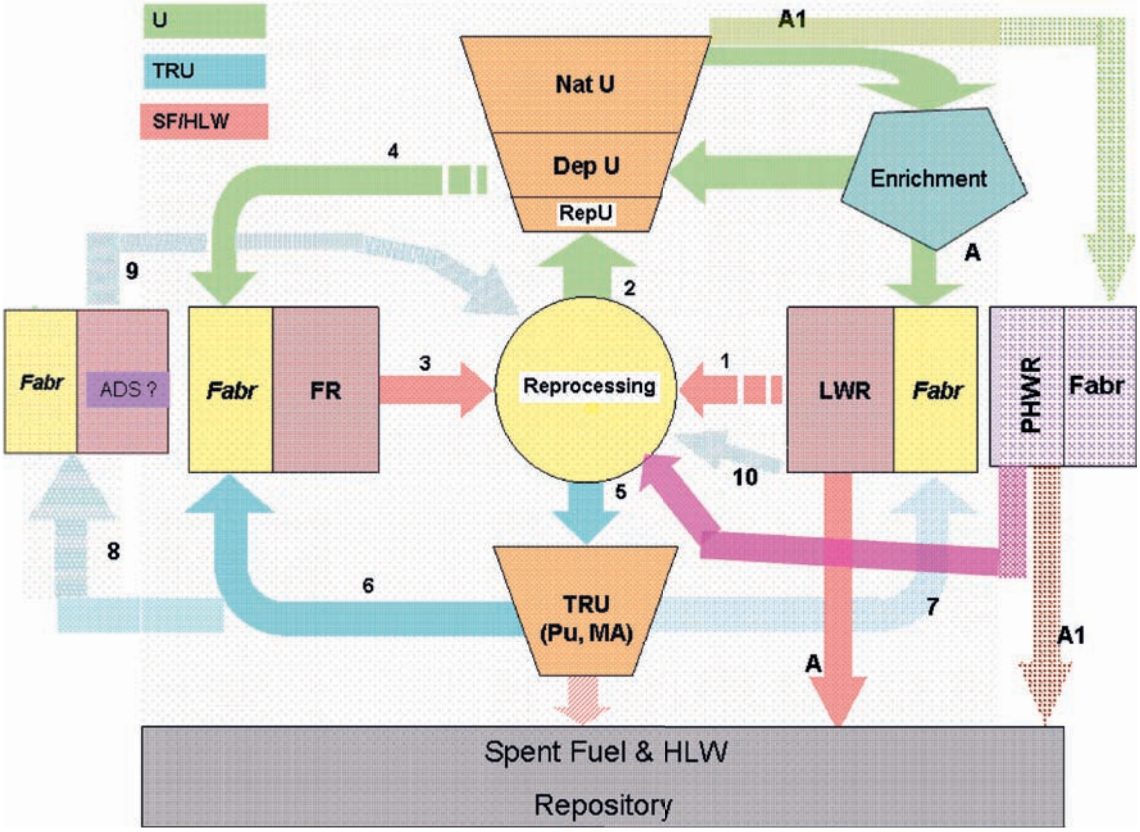


FIG. 1. Role of fast reactor fuel reprocessing in a future nuclear fuel cycle.

Closed fuel cycle (CFC) schemes in combination with FRs are emerging as the key strategy for sustainable development of nuclear energy. Conventional reprocessing technologies have limitations, namely production of pure Pu (proliferation concern) and MAs in high level liquid waste (HLLW) that lingers for millions of years after disposal. In response to these concerns, several 'advanced partitioning methods' are being developed with emphasis on co-recovery of actinides. Additionally, use of reprocessed uranium as a fuel would enhance uranium resource utilization. Two approaches are under development in aqueous partitioning processes: co-recovery of TRU elements from plutonium uranium extraction (PUREX) raffinate and alternate processes to PUREX. Pyro-processing technologies are being developed with a long term perspective. A synergetic combination of aqueous and pyro-processes constitute a critical stage of CFC development. Figure 1 depicts the potential central role of FR fuel reprocessing in future nuclear energy systems.

1.7. EXPERIENCE IN BACK END TECHNOLOGIES

Significant experience in the plant scale reprocessing of spent fuels exists from thermal neutron reactors using PUREX technology. While the PUREX process or its advanced versions may be used for aqueous reprocessing of FR oxide fuels as well, a number of additional requirements come into play due to the high Pu content of the fuel as well as the high levels of radioactivity associated with the irradiated FR fuel. Considering the need for further reducing the cooling periods and increasing the burnup due to economy and sustainability considerations, pyro-processing technologies based on inorganic molten salts are being developed as alternatives for future FRs. Pyro-processing technology is also ideally suited for metallic fuels, besides being intrinsically more proliferation resistant. While waste management technologies for FR fuel based on aqueous reprocessing will be similar to those used for thermal reactor fuels, the treatment of pyro-processed waste and its ultimate disposal require further development. Thus, the development of fuel cycle technologies will continue to be a challenging area of activity which will receive increased attention in the coming decades. The OECD Nuclear Energy Agency (OECD/NEA), in collaboration with the IAEA, has organized several meetings and published technical reports in recent years in the area of nuclear fuel cycle technology [9–11].

1.8. OBJECTIVE

The back end of the fuel cycle for FRs and thermal reactors differs mainly with respect to reprocessing operations. Technologies concerning conditioning of waste and its geological disposal are similar in the case of fuel cycles for both systems. The objective of this report is to highlight specific issues with regard to the back end of the FR fuel cycle. Thus, the report focuses more on reprocessing operations. In line with the goals of INPRO, the report also discusses emerging innovations and R&D needs for the back end of the fuel cycle, with an emphasis on reprocessing.

1.9. STRUCTURE

Following this introduction, Section 2 provides an overview of the main technologies used or proposed for use at different stages of reprocessing SNF from FRs. Section 3 describes the processes of waste generation and the characteristics and management of waste. Section 4 describes emerging technologies for the back end of the FR fuel cycle. Section 5 discusses enabling technologies for reprocessing and waste management facilities. Section 6 describes the status and prospects for the development of back end fuel cycle technologies in different countries. Section 7 analyses future R&D and innovation related to the back end of the fuel cycle. Section 8 outlines the conclusions derived from the information presented in the report. Finally, a list of resource materials related to the back end of FR fuel technologies is given in the annex.

2. OVERVIEW OF THE BASIC PROCESSES IN FAST REACTOR FUEL REPROCESSING

2.1. INTRODUCTION

Reprocessing is an important operation in nuclear fuel cycle activities, which increases the effective utilization of unburnt as well as newly generated fissile content of spent fuel. This allows utilization of the fertile component of the fuel and also reduces the volume and average decay time of the waste to be handled. Reprocessing also helps to effectively burn the MAs in FRs or accelerator driven systems after their recovery. The recovery of selected fission products (FPs) of economic value is also a possibility.

Additional requirements for reprocessing may vary, depending on national policies in areas such as non-proliferation, waste management, commercial use of radioisotopes, etc. The majority of such additional requirements are related to the needs and feasibility of further partitioning of products extracted from SNF, and may sometimes be contradictory:

- Separation of uranium and plutonium: May be attractive for many fuel fabrication concepts but less favoured from the proliferation viewpoint;
- Separation (with or without further partitioning) of MAs (mainly Np, Am and Cm) and LLRNs such as Tc: Significantly simplifies waste management, but is necessary only if further transmutation is envisaged;
- Separation of specific radioactive FPs (e.g. Cs and Sr) from the complex FP mixture: May be attractive for reducing specific heat production and activity of waste but necessary only if specific uses are identified.

Various types of chemical processes can be applied in different sequences to the flow of SNF depending upon the requirements. The emphasis on one or another criterion and certain specifics of fuel used in different types of reactors may also dictate the choice and the sequence of chemical treatment methods, thus resulting in a number of technically possible variations of a general flow sheet. Nevertheless, there are certain general features that allow reasonable categorization to be applied and the analysis to be presented in a more or less structured form. The most important features of such generalization are separate discussions for the aqueous and non-aqueous flow sheets, radically differing from each other by the basic (pyro-)chemical reactions involved, and use of the ‘classical’ PUREX process throughout the text for both comparative and reference purposes.

The importance of the PUREX process is based on its well established status as a state of the art standard for reprocessing SNF from thermal reactors, its general applicability to reprocessing of FR fuels, the significant operational experience accumulated worldwide, and being a prototype for the majority of alternative aqueous reprocessing methods proposed for spent fuel from FRs.

2.2. GENERAL CONSIDERATIONS IN FAST REACTOR FUEL REPROCESSING

FR fuel reprocessing differs from thermal reactor reprocessing in many respects [12]. The differences arise mainly from the following characteristics of spent FR fuels:

- High specific activity and decay heat generation consequent to high burnup;
- Presence of additional hardware and presence of sodium on subassemblies in the case of sodium cooled reactors;
- Enhanced formation of platinum group metals (PGMs) and insoluble residues in the fuel;
- Pyrophoric nature of carbide, nitride and metallic fuels.

The influence of the above characteristics on the different stages of the reprocessing cycle are briefly described below and evidently show the need for either serious modifications in the PUREX like aqueous processes or substituting them with alternate non-aqueous (mainly molten inorganic salts) technologies, which are currently under active development.

2.2.1. High specific activity and decay heat generation consequent to high burnup

The higher burnup achieved in FRs will necessitate shipping casks with additional shielding and probably forced cooling due to high specific power (Table 1). For the aqueous reprocessing technologies, the high specific activity causes enhanced solvent degradation, leading to problems such as phase separation, formation of cruds, incomplete stripping, etc. Centrifugal extractors, with a short residence time will have to be used to circumvent this disadvantage. Higher FP content also demands higher decontamination factors (DFs) for the uranium–plutonium fractions to avoid man-Rem problems in the fuel fabrication steps that follow.

TABLE 1. COMPARISON OF RESIDUAL DECAY HEAT OF SPENT NUCLEAR FUEL AND RADIATION DAMAGE TO SOLVENT IN THE PUREX PROCESS

Spent fuel from	Cooling time (a)	Burnup (GW·d/t HM)	Total decay heat (W/t HM)	Dose emitted by aqueous phase (Wh/L)	Dose received by solvent (Wh/L)
LWR-UOX	4	50	3.48	0.57	0.20
LWR-MOX	7	50	6.31	1.36	0.48
FR-MOX	7	185	21.77	6.72	2.40

2.2.2. High fissile content and higher plutonium content

The sizing of process equipment and utilization of neutron absorbing materials in FR fuel reprocessing plants could be considerably different from those in thermal reactor fuel reprocessing plants due to criticality considerations. The high plutonium content of the fuel also necessitates vigorous conditions for dissolution. Third phase formation during liquid–liquid extraction should also be taken into account in the design of the flow sheet and operation of the contactors.

2.2.3. Presence of additional hardware and sodium

Additional hardware, such as wrapper, has to be removed in the head-end step. The fuel subassemblies from liquid SFRs will have adhering sodium that may have to be removed prior to treatment of the spent fuel. Due to the chemical reactivity of sodium with water, safe and economic methods have to be followed.

2.2.4. Enhanced formation of platinum group metals and insoluble residues in the fuel

Due to the higher yield of PGMs in plutonium fission, some inter-metallic alloys containing significant fissile material are formed in the fuel and they remain insoluble during dissolution. The insoluble compounds also contain fissile material and require vigorous conditions for dissolution for recovery of fissile material. During the reprocessing of short cooled fuels, volatilization of ruthenium in the form of RuO_4 is significant at high acidities. The PGM may also cause phase segregation during vitrification of the high level waste (HLW).

2.2.5. Pyrophoric nature of carbide, nitride and metallic fuels

Metallic fuels, (U, Pu)C or (U, Pu)N, can react with oxygen or moisture. Head-end operations with these fuels require inert atmosphere cover during chopping operations. Additional steps may be required for carbide and nitride fuels to get PUREX compatible feed solutions.

2.2.6. Additional requirements for reprocessing

To improve economy, reduce environmental impact and address public acceptance issues, considerable R&D work is being carried out for:

- Reduction in the number of process steps;
- Reduction in the HLW volume for deep geological disposal by removal of long lived fission products (LLFPs) and recovery of MAs;
- Recovery of valuable FPs from the waste.

These additional requirements refer to both fast and thermal reactor fuel cycles. Innovative nuclear energy systems are expected to integrate the development of new processes along the above lines.

2.3. AQUEOUS REPROCESSING SCHEMES AND INDIVIDUAL PROCESSES

Starting with the Experimental Breeder Reactor I, which first became critical in 1951, over 18 FRs have been constructed and operated for a total of 385 reactor-years and three reactors are under construction as of 2007. However, the number of aqueous based FR fuel reprocessing plants constructed and operated, as well as the total reprocessing years logged, are proportionately far fewer compared to the total reprocessing years of commercial thermal reactor fuel reprocessing as well as those of research reactors in the military and civil domains.

In many countries, policy decisions taken by the respective governments abruptly stopped aqueous reprocessing operations and construction activities, or even the commissioning activities of already constructed plants.

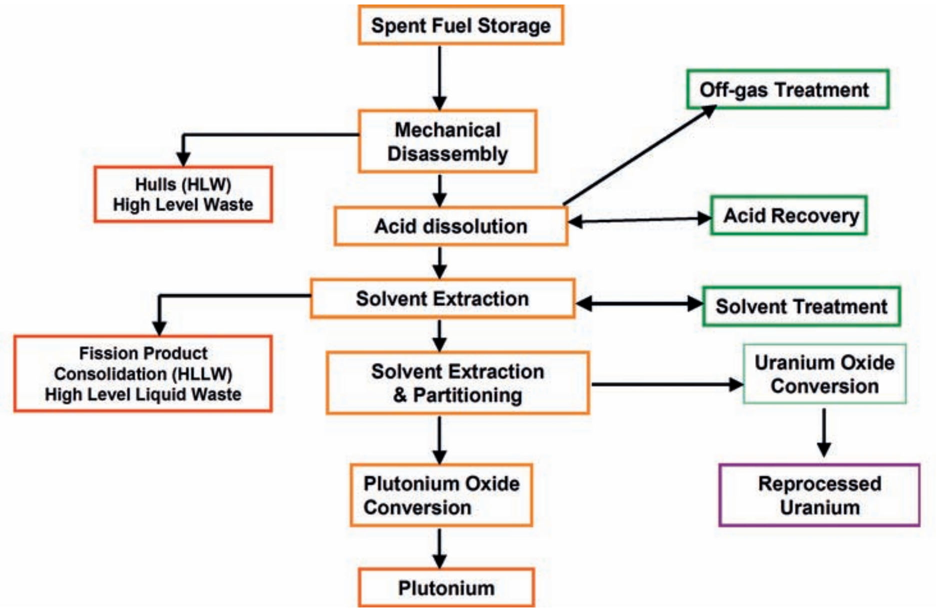


FIG. 2. Schematic flowsheet of the PUREX process.

However, the experience gained so far in this field can still be considered as worthy. Moreover, the large amount of experience gained in the aqueous reprocessing of thermal research and power reactor fuels, the continuous evolution of new processes and reagents, efforts to reduce generated waste as well as efforts to simplify the overall process provide ample confidence that pursuing FR fuel reprocessing through the aqueous route could be an attractive choice.

An excellent account of the P&T studies being carried out in Member States is available on the Internet as well as in the proceedings of several information exchange meetings conducted by the OECD/NEA (refer to the annex for additional information). Madic's [13] overview, entitled "The hydrometallurgical and pyro-metallurgical process studied worldwide for the partitioning and transmutation" covers a wide spectrum of R&D activities carried out under the European Commission Framework Programme.

In some organizations and countries (e.g. in the European Union, Japan, the Russian Federation and the United States of America), instead of developing a succession of separation processes for particular LLRNs, the integration of processes for MAs and FP extraction are studied. The following sections describe the various processes adopted to extract fissile content from spent fuel and to partition LLRNs and MAs from HLW streams. An account of the work being pursued in different countries is presented in Section 4.

2.3.1. PUREX reprocessing technology

The time tested PUREX process [14], though with some variations, has also been used for aqueous reprocessing of FR fuels. The following sections briefly describe the various steps of the PUREX aqueous reprocessing process. These are presented schematically in Fig. 2.

2.3.1.1. Head-end steps

The head-end steps in aqueous reprocessing involve disassembly, chopping the fuel pins in a single/multipin chopper and dissolution of the fuel in nitric acid. Mechanical as well as laser based techniques have been used for the removal of the wrapper. Hull monitoring for measuring residual fuel material is a standard practice adopted worldwide.

2.3.1.2. Feed clarification and conditioning

The dissolver solution is subjected to clarification by a suitable filtration step or by centrifuging to obtain feed solutions devoid of clad fines and undissolved residues which may otherwise cause severe phase separation problems and crud formation during solvent extraction. The nitric acid concentration has to be adjusted to acidities compatible with PUREX feeds, i.e. around 2–4M HNO_3, depending on the flow sheet. Considerable R&D work has been carried out in developing safer methods, such as electrolytic reduction, etc. The valence of plutonium has to be adjusted to the most extractable (IV) state prior to solvent extraction. Salt free techniques such as sparging NO_2 gas and electrolytic adjustment have replaced the old process of addition of sodium nitrite.

2.3.1.3. Co-decontamination cycle

Tri-n-butyl phosphate (TBP) has remained as the main extractant, while a wide range of diluents, such as n-dodecane, hydrogenated propylene tetramer, normal paraffin hydrocarbon mixture and odourless kerosene, have been used. The concentration of the TBP used is around 30% (vol./vol.). In the co-decontamination cycle, co-extraction of uranium and plutonium is carried out leaving FPs in the aqueous phase. Different flow sheets are followed by countries with variations in feed acidity, scrub acidity, etc. A variety of liquid–liquid contactors such as mixer–settlers, pulsed columns and centrifugal contactors are used in various plants.

The development of alternative extractants to TBP has the objective of overcoming problems of third phase formation, and residual waste generation on incineration. Notable among the alternative candidates are: long chain dialkyl amides which are completely incinerable, and higher homologues of TBP, such as tri-isoamyl phosphate (TIAP) and tri-alkyl phosphate (TAP), which do not form a third phase with plutonium and which have very low aqueous solubility [15].

2.3.1.4. Partitioning cycle

The next step after co-decontamination is partitioning, wherein plutonium is separated from uranium. A reducing agent, such as uranous nitrate or hydroxylamine nitrate, is employed in the presence of the nitrite scavenger hydrazine. The Dounreay reprocessing flow sheet used sulphuric acid for partitioning. Continuous research worldwide has led to the development of other reagents, such as iso-butyraldehyde, n-butyraldehyde, and formo and acetohydroxamic acids. However, none has been tested in actual reprocessing plants.

Co-processing of uranium and plutonium is being explored as a proliferation resistance measure in some countries. The co-stripped product can be used directly for mixed oxide (MOX) pellet preparation with suitable adjustments of the composition either by the powder pellet route or by sol-gel based methods.

2.3.1.5. Purification cycles

Subsequent to partitioning, plutonium and uranium are subjected to several purification cycles and are finally brought into the form of pure nitrate solutions.

2.3.1.6. Tail-end steps

Tail-end operations involve re-conversion of plutonium to PuO_2. This is done by precipitation as Pu(IV) oxalate and calcination in air to PuO_2. Similarly, uranium is precipitated as ammonium diuranate/ammonium uranyl carbonate and calcined to U_3O_8. Alternatively, microwave de-nitration of the uranyl-nitrate solutions to get the oxide is also in use in several plants [16, 17].

At this point, the PUREX process, in its classical version, results in two product flows, namely plutonium and uranium dioxides, the primary products in reprocessing that are directed back to fuel fabrication cycles, and a waste stream, targeted either for disposal or for further processing prior to final disposal. These additional steps can be considered to be a part of reprocessing or waste management. For the sake of clarity, in this report, the additional steps in the PUREX and PUREX-like waste stream are treated as part of fuel reprocessing, while waste management processes are assumed to start with already processed waste, i.e. after additional extraction/partitioning/separation steps.

2.4. NON-AQUEOUS REPROCESSING OF SPENT NUCLEAR FUEL

Non-aqueous methods of fuel reprocessing exclude the use of both aqueous and organic media, and are based on the use of liquid metals, molten salts or halides, usually at high temperatures. Non-aqueous methods are the potential alternative to aqueous processing methods.

Three non-aqueous methods are currently receiving the major focus of attention in different countries [18–21]:

- The molten salt electro-refining method for metallic fuels (originally developed by the Argonne National Laboratory (ANL), IL, USA);
- The oxide electrowinning method for oxide fuels (originally developed by the Russian Institute of Atomic Reactors (RIAR), Russian Federation);
- The fluoride volatility process of oxide and metallic fuels (developed by France, the Russian Federation, Japan and the USA).

These pyrochemical methods are being studied in India. The Republic of Korea is developing the molten salt electro-refining method for metallic fuels for P&T as well as the electrochemical reduction process for actinide oxides.

The advantages of non-aqueous methods are the:

- High chemical and radiation stability of molten salt;
- Ability to handle high concentrations of fissile materials (>30%);
- Absence of neutron moderators;
- Adaptability to carry out all of the processes in a single apparatus irrespective of the type of fuel (metals, oxides, nitrides, etc.);
- Absence of HLLW;
- Inherent proliferation resistance.

Non-aqueous methods enable treatment of high burnup, short cooled fuels in a smaller number of process steps compared to their aqueous counterparts. Since moderators are absent, high concentrations of fissile and radioactive materials can be handled in these processes, which make the plants based on them more compact [22, 23].

Besides the molten salt electro-refining process for metallic fuels and the oxide electrowinning process for oxide fuels, the fluoride volatility process (to a large extent) and reductive extraction in fluoride salts (to a smaller extent) are being developed for treatment of carbide and oxide fuels of gas cooled FRs, and the fluoride salts of molten salt reactors.

The basics of non-aqueous reprocessing methods are briefly outlined in the following sections, with more details in the country profile sections further on.

2.4.1. Molten salt electro-refining process

The integral fast reactor (IFR) concept and its fuel cycle were developed in the USA based on the actinide recycle programme. The IFR fuel cycle is based on reprocessing of its spent U–Pu–Zr alloy fuel using a molten salt electro-refining process.

This process flow sheet involves the following steps:

- Chopping of fuel pin containing spent fuel into pieces and loading into the electro-refining cell in a basket;
- Addition of $CdCl_2$ to the electro-refining cell at a temperature of 773 K to transfer most of the actinides, sodium and FPs as chlorides to the electrolyte (eutectic mixture of KC1 and LiCl);
- Deposition of uranium on a solid cathode (dendritic deposit);
- Introduction of a cadmium cathode in the cell as the predetermined concentration of Pu is reached in the electrolyte to deposit Pu and the remaining actinides, including an approximately equal amount of uranium on the cadmium cathode.

A cylindrical rod of low carbon steel (zirconium, molybdenum or uranium may be used) is used as the cathode for selective deposition of uranium. The higher thermodynamic stability of $PuCl_3$ compared to UCl_3 renders the deposition of plutonium on the solid cathode impossible, unless the $PuCl_3$ to UCl_3 ratio is >2 which is not realizable under normal process conditions. However, co-deposition of uranium and plutonium on a liquid cadmium cathode is enabled by the lower activity coefficient of plutonium in cadmium compared to that of uranium. A liquid cadmium cathode (liquid cadmium in a beryllium crucible) is used in the IFR reactor fuel cycle pyro-process for extraction of plutonium and other MAs due to the improved kinetics as compared to a solid cadmium cathode.

The deposit of uranium and plutonium on the cadmium cathode tends to grow and short the electrodes, hence a rotating cathode is used to compress the salt/cadmium surface and to produce a deposit without dendrites. Cathode deposits are removed from the electro-refining cell after the process is completed. Uranium is separated from the salt (in the case of a solid cathode) and TRU elements from cadmium (in the case of molten cadmium) through distillation in a retort and then melted. Ingots of materials are used for the fabrication of fuel elements using the injection casting process [24]. A major fraction of the salt as well as Cd are recycled back into the refiner, leaving only a small fraction in the waste [25].

The molten salt electro-refining process generates two waste streams: a salt stream that does not contain TRU elements and a stream of metals containing TRU element traces. Accordingly, two waste forms are produced, namely a ceramic waste form and a metal waste form which are discussed in more detail in the following sections. Schematics of the process are presented in Fig. 3.

Although the molten salt electro-refining process was developed for treating spent metallic fuels, it can be used for the reprocessing of spent nitride or carbide fuels as well. Since nitride or carbide fuels have high electrical conductivity, they could be used as the anode of an electro-refining cell, and U and Pu metals can be deposited on the cathode during the process. Studies carried out in Japan have also shown that in situ nitriding in liquid cadmium used as a cathode is possible. However, the associated technical challenges/issues to be addressed in the development of a pyrochemical process, i.e. electro-refining are listed in Table 2.

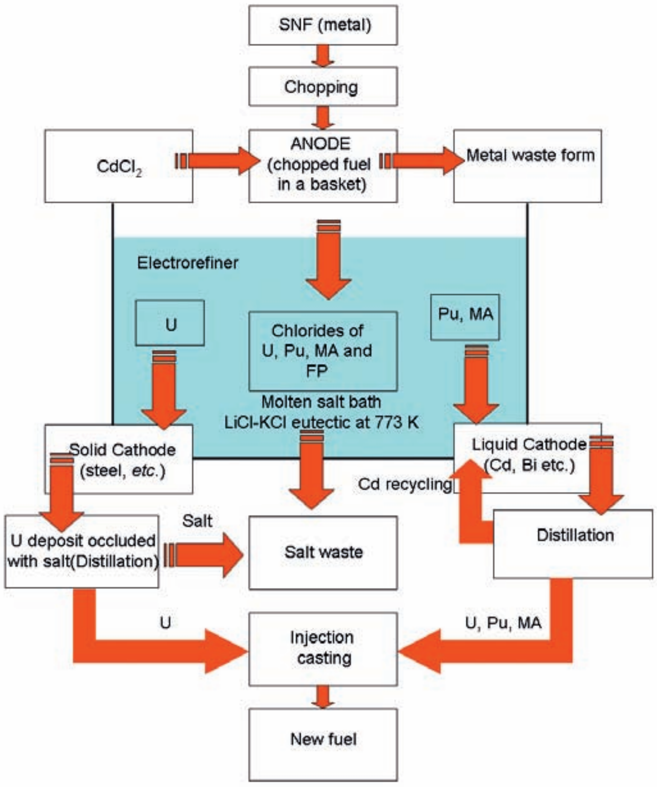


FIG. 3. Flow sheet of the molten salt electro-refining process.

TABLE 2. TECHNICAL CHALLENGES IN THE DEVELOPMENT OF PYROCHEMICAL PROCESSES

Method	Major technical issues
Metal electro-refining reprocessing method	Establishment of a criticality safety control method and material accounting system. Demonstration of the main processes, such as the TRU element recovery process by domestic tests with spent metallic fuel. Optimization of the specific salt waste treatment and confirmation of its adaptability to disposal. Development of process equipment suitable for mass production, remote handling and remote maintenance repair.
Metal casting injection method	Assurance for homogeneity of the U–Pu–MA–Zr fuel slug. Development of process equipment suitable for mass production, remote handling and remote maintenance repair.

2.4.2. Oxide electrowinning process

This process is used for the treatment of spent oxide fuels. Decladding of the fuel pin by shearing is considered to be the first step of this process and the other process steps are as follows:

- Chlorination of the fuel in the presence of molten NaCl–KCl or NaCl–CsCl to ensure complete dissolution of the fuel components in a salt.
- Electrolysis to deposit part of UO_2 free from Pu. Some FPs (Zr, Nb, Ru, Rh, Pd, Ag) are also deposited on the cathode. The U/Pu separation factor at this stage is 120–140.
- Precipitative crystallization of PuO_2 which is free from FPs that yields crystalline PuO_2 ready for vibropacking; 99.5–99.9% of Pu is collected in the PuO_2 precipitated at the bottom.
- Additional electrolysis to deposit uranium oxide: large amounts of FPs are deposited on the cathode along with UO_2 from the melt.
- Purification of electrolyte salt: this operation is carried out by introducing sodium phosphate into the melt. As a result, the impurities precipitate in the form of their respective phosphates which are insoluble in molten chlorides as well as in water. Cs, Rb and part of Sr remain in the molten salt.

The particles in the deposit have a high density of >10.7 g/cm^3 and are less than 1 mm in size. The salt covering on the deposit is washed with water and then the salt is recovered by evaporation of water for recycling. The DFs are less than 100, which is acceptable for FRs.

Omitting the second and the third step in the process allows exploiting an option with co-deposition of U and Pu oxides, which may be reasonable in the case of preparing MOX fuels in subsequent stages. The schematic flow sheet for the process (including the co-deposition option) is presented in Fig. 4.

It is also possible to design process steps to recover elements of the ruthenium subgroup from the waste. Since the process does not require stringent control of the gas atmosphere for the process, the processes are carried out in hot cells that have a normal air environment [26].

In the oxide electrowinning method, the electrolyte salt bath is proposed to be cleaned up by phosphate precipitation with Na–Cs phosphate. The FPs are precipitated as phosphates, and are then vitrified into a phosphate glass. The noble metal FPs are separated by electrolysis from the salt bath and then solidified into a metal waste form. The TRU elements remain in the electrolysis crucible.

2.4.3. Fluoride volatility process

The high radiation stability of fluorides makes the fluoride volatility process a candidate method to reprocess irradiated nuclear fuel with short cooling periods, which is especially important for breeder reactors. The difference in the volatility and the differences in their characteristics of being absorbed on the fluorides of alkaline and alkaline earth metals serve as the basis for methods of purifying uranium and plutonium from FPs. The methods of separating UF_6 and PuF_6 are based on differences in their thermodynamic stability. The process was developed in the early 1960s in the USA [27] and the former Soviet Union [28].

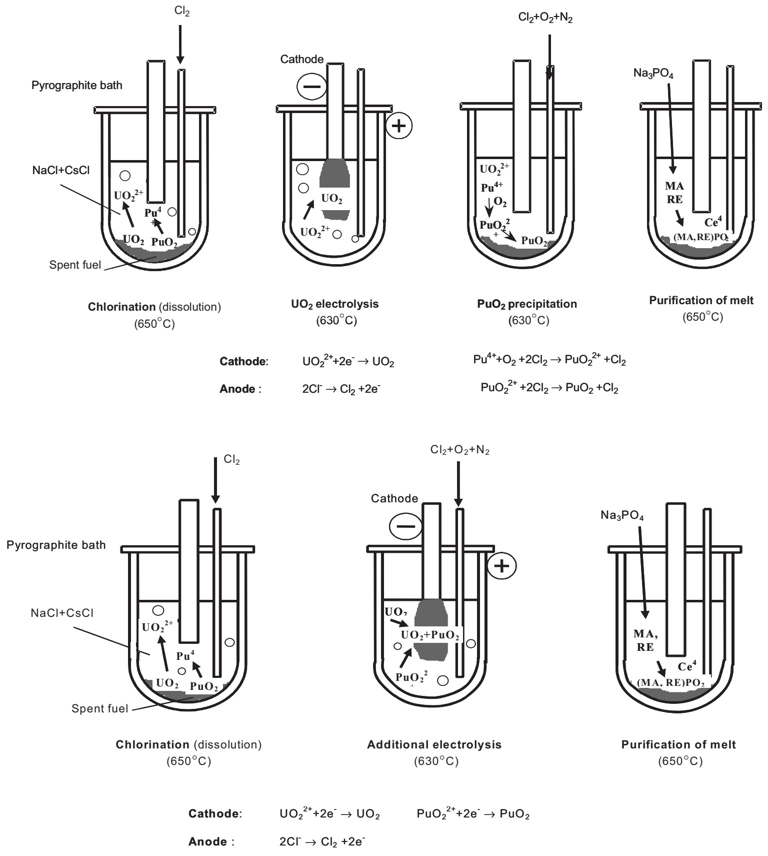


FIG. 4. Schematic flow sheet for the electrowinning process.

In this process, oxide fuels are oxidized to U_3O_8 and charged to a fluidized bed of alumina through which a fluorinating gas such as F_2, ClF_3 or BrF_5 is passed. The radioactive waste, consisting of fluorides of FPs, is produced directly in a compact form. Although the industrial production of UF_6 was mastered long ago, extensive R&D work was required to achieve efficient heat removal in sufficiently small (criticality safe) dimensions and to adequately filter the effluent gas flow, in order to optimize the fluorination process of irradiated fuel. Schematics of the process are presented in Fig. 5.

The following key problems need to be addressed for commercialization of the fluoride volatility method:

- Heat removal during fluorination of irradiated fuel;
- Purification of uranium hexafluoride from FPs;
- Concentration of FPs;
- Separation and purification of plutonium;
- Development of remote controlled equipment for high temperature processes.

The decisive advantage of the fluoride volatility process (the potential for producing non-volatile FPs in a compact form) has been experimentally demonstrated in the FREGAT (Russian abbreviation for facility for fluoride volatility reprocessing of spent fuel) facility. This special facility enabled complex studies on the reprocessing of irradiated UO_2 and MOX fuel from fast reactors. The fuel used in the tests had a burnup of about 100 GW/t HM and a short cooling period (3–6 months). The UF_6 product had a high DF from FPs ($>10^6$) after reprocessing. The total recovery of uranium from irradiated nuclear fuel was 99.4–99.6% and the extent of conversion to UF_6 was 96.1–98.3%. The losses were 0.4–0.5%. Most of the FPs (~85%) were concentrated in residues of fluorination. The HLWs were compacted by melting to cryolyte-type rocks.

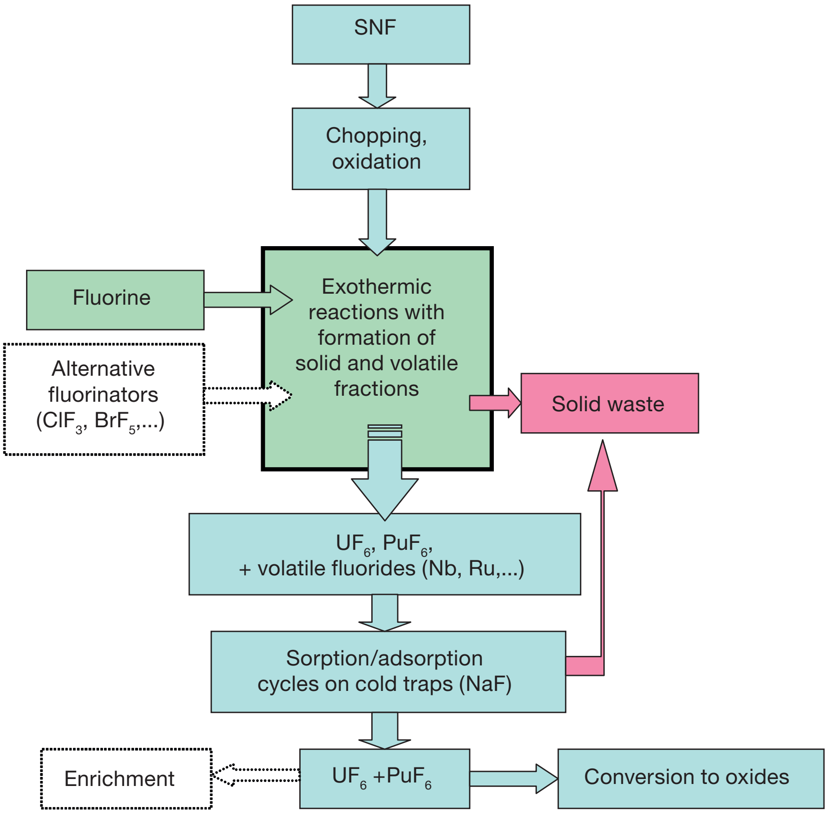


FIG. 5. Schematic flow sheet for the fluoride volatility process.

The recovery of Pu is low (89–91%) due to the instability of PuF_6. It was also proposed to carry out a low temperature fluorination of fuel. This could improve the method, especially from the point of view of decreased release of FPs and TRU elements into the gas phase.

The development of fluoride volatility processes declined in the USA following the cancellation of the MSRE project in 1973 and in the Russian Federation after the Chernobyl accident. In the Czech Republic, detailed studies were also carried out and a conceptual design of an integrated fluoride volatility plant was developed in parallel. Severe corrosion problems occurred over the course of these investigations, and in 1972 the decision was taken to abandon the fluoride volatility route for the reprocessing of irradiated nuclear fuel. In the recent past, R&D activities on the volatility process have been revived in Japan, the Russian Federation and the Czech Republic [29–38].

3. WASTE GENERATION PROCESSES, WASTE CHARACTERISTICS AND MANAGEMENT

3.1. INTRODUCTION

The management of radioactive waste requires different approaches to ensure the protection of both humans and the environment from radiation. Radioactive waste disposal systems typically include a system of barriers that act to isolate and contain the waste and, thereby, protect the environment and human health.

Radioactive waste can be classified by the level of radioactivity present (high, intermediate, low or below regulatory concern), by the dominant type of radiation emitted (alpha, beta, gamma or X ray) or by its half-life (the length of time required for the material to decay to half of its original value). Radioactive wastes can also be classified by their physical characteristics (primarily solid or liquid, but they can also exist in the gaseous state). A quantitative way of classifying radioactive waste is by specific activity or activity concentration, i.e. by the activity per quantity of waste (mass or volume). The heat generated in a sample (depends on half-life, concentration and type of radiation) can also be used for classification. Finally, waste can be classified for security and non-proliferation purposes (e.g. designated as ‘special nuclear materials’), for worker safety and for transport.

The method selected to classify radioactive waste is usually dependent on the generator of the material. For example, a nuclear power plant operator is likely to categorize waste based on the originating stream. This system is difficult to use widely because the streams differ from case to case. A classification system that takes into account qualitative considerations affecting final disposal of the conditioned waste is preferable. This report adopts the waste categories defined in the IAEA classification shown in Table 3. The classification system described in Table 3 distinguishes radioactive waste types based on two key characteristics — thermal hazard and requirements for disposal. Two types of wastes are mainly identified: HLW, and low and intermediate level waste (LILW) with short and long lifetimes [39].

3.2. MANAGEMENT OF RADIOACTIVE WASTE IN THE FAST REACTOR FUEL CYCLE

There is much experience the world over on the management of different types of waste arising from thermal reactor fuel reprocessing. Several countries have also reached technological maturity with regard to conditioning of HLW. Borosilicate glass as the matrix for immobilization of HLW has found acceptance and a vast fund of knowledge has been generated on this material. Challenges remain with regard to long term geological disposal of radioactive waste. Developmental efforts in different countries are now oriented towards reducing the cost of the operations and management of HLW in the FR fuel cycle, which has similar features to the thermal reactor fuel cycle. With the high burnup reached by FR fuels, the quantities of MAs per tonne of irradiated fuel are higher in the case of FR fuel compared to thermal reactor fuel. Similarly, the concentration of FP nuclides in the waste solution is higher by an order of magnitude as compared to the waste solution from reprocessing of thermal reactors (Table 4).

The quantities of FPs produced per tonne of discharged fuel are higher for SFR fuel than for LWR-UOX fuel. However, these quantities, with the exception of the noble metals, are comparable (e.g. Xe, Ba, La and Nd) when normalized to energy produced. Thus, for the same amount of nuclear power produced, the reprocessing of SFR fuel does not produce greater quantities of high activity FP waste than the reprocessing of LWR-UOX fuel.

TABLE 3. WASTE CATEGORIES DEFINED IN THE IAEA CLASSIFICATION

Category	HLW Deep geological disposal	LILW-LL Geological disposal	LILW-SL Surface or geological disposal
Key feature	Highly radioactive waste, containing mainly fission products, as well as some actinides, separated during reprocessing of irradiated fuel. Spent fuel, if it is declared a waste.	Waste, which, because of its radionuclide content, requires shielding but needs little or no provision for heat dissipation during its handling and transport.	Waste, which, because of its low radionuclide content, does not require shielding during normal handling and transport.
Heat generation	Any other waste with radioactivity levels intense enough to generate heat more than 2 kW/m^3 by the radioactive decay process.	<2 kW/m^3	<2 kW/m^3
Half-life		<30 a	<30 a
Other characteristic			Activity content <400 Bq/g of long lived alpha emitters.

Long lived waste (LILW-LL); short lived waste (LILW-SL)

TABLE 4. COMPARISON OF THE CHARACTERISTICS OF SPENT NUCLEAR FUELS

Spent nuclear fuel	LWR-UOX	LWR-MOX	SFR-MOX
Burnup (GW·d/t HM)	60	60	120
Enrichment	4.5% U-235	8.2% Pu	30% Pu
Cooling time (a)	5	3	5
Fission products (g/t U + Pu)			
Kr	607	265	770
Sr	1370	546	1536
Y	779	291	816
Zr	6290	3310	9356
Mo	5990	4140	11 008
Ru	4130	3690	10 900
Rh	739	1060	3235
Pd	2820	3520	8624
Te	870	705	2427
I	383	372	1070
Xe	9650	6780	16 455
Cs	4560	4220	13 619
Ba	3110	2020	5477
La	2150	1500	4094
Ce	4200	2830	7497
Pr	1970	1370	3549
Nd	7200	4670	12 562
Pm	58	104	266
Actinide (g/t U + Pu)			
U-234	229	12	96
U-235	5870	1070	1221
U-236	6240	255	323
U-238	911 000	886 000	632 270
Np-237	889	152	318
Pu-238	590	2390	1299
Pu-239	6360	23 100	143 250
Pu-240	3180	19 600	78 508
Pu-241	1640	8930	9141
Pu-242	1230	7300	4491
Am-241	504	2550	3964
Am-242m	1.3	29	66
Am-243	422	2040	396
Cm-242	0.02	2	0.2
Cm-243	1.3	12	9
Cm-244	177	923	73
Cm-245	15	111	4

In FRs, concerning the MAs, the ratio between neutron fission cross-section and neutron capture cross-section (σf:σc) is much higher than in thermal reactors. Thus, for a similar amount of energy produced, the production of MAs is relatively less in an FR-MOX fuel than in an LWR-MOX fuel. It is also to be noted that while comparing UOX with MOX irradiations, the production of Am and Cm is much higher in the case of MOX due to the initial inventory of Pu. With these characteristics, the FR route can be simultaneously used as a producer of nuclear energy and transmuter of MA.

The long term strategy for the management of HLW from FRs, thus, includes partitioning of MAs and useful FPs. The partitioning of MAs will address the long term concerns of glass as an immobilization matrix. Although the need for geological disposal will still continue, this would mean a medium depth repository instead of a deep geological repository, as conceived today. Recovery of resource metals such as caesium, strontium and PGM (Ru, Pd and Rh) will not only have an economic value but will also enable increased waste loading in the glass. In addition, partitioning of ^{137}Cs and ^{90}Sr will lead to a reduction in the long term heat generation in the vitrified waste. Partitioned waste elements may also be fixed in crystalline ceramic matrices.

The thermal load to a geological repository of nuclear waste is a combined effect of short term heat generating radionuclides, such as FPs, Cs and Sr, and long term heat generating elements, such as Pu and Am. If Cs and Sr were to be separated for temporary storage for 200 to 300 years for their decay and then were to be disposed as LLW, and group separation of actinides for their interim storage or recycling in advanced reactors were to be performed, this would greatly enhance repository capacity.

The main elements of national policy and strategy for safe management of radioactive waste are to set out clear objectives to minimize radioactive waste. The strategies adopted may also depend on the national availability of waste management competence, facilities and technology.

3.2.1. Management of low and intermediate level waste

Liquid waste is segregated into low level, intermediate level aqueous waste, alpha bearing liquid waste, organic waste and special waste. A categorization is also done for solid waste.

There are many methods of volume reduction for aqueous liquid waste, some of which are given below:

- Evaporation;
- Chemical precipitation or coagulation;
- Membrane methods;
- Ion exchange separations.

After volume reduction, the concentrate is conditioned in a suitable matrix, such as concrete before disposal.

The basic principle of management of low level liquid waste (LLLW) is largely to dilute and disperse it in large water bodies, while intermediate level liquid waste (ILLW), and low and medium level solid waste are subjected to volume reduction and conditioning.

Very low liquid waste generated in the plant during decontamination of floors, wash showers, evaporator condensates, etc., is disposed of by diluting in large water bodies such as the sea or a river. LLW and HLW are treated for removal of FP activities in a small volume, so that the remaining liquid waste of very low level can be disposed of directly. The separated FPs can be conditioned by suitable immobilization methods and stored as solid waste.

3.2.2. Management of high level waste

A three stage programme for the management of HLLW has evolved as an international practice for fuel cycles for LWRs and FRs:

- Conditioning of HLLWs, wherein radionuclides present in the aqueous stream are immobilized in suitable vitreous matrices that are inert and highly durable (resistant to chemical/aqueous attack), and can be contained in high integrity storage units which are subsequently over-packed;
- Interim storage under surveillance and cooling of over-packs containing conditioned wastes for periods ranging up to 50 years or more to allow reduction in decay heat to a level acceptable for geological disposal,

on the one hand, and to ensure integrity of the waste form and its packaging, on the other, before a commitment is made for their irretrievable disposal;

- Disposal in a deep underground repository in such a way that potentially hazardous radioactive materials are at no stage recycled back to the human environment in concentrations that could subject the biosphere to a risk considered unacceptable.

3.2.2.1. Conditioning for thermal reactor fuel reprocessing of high level liquid wastes

There is rich experience in several countries on thermal reactor HLLW conditioning, and this can be extended to FR HLLW with necessary modifications. The constituents of HLLW comprise FPs, such as ^{137}Cs, ^{90}Sr, ^{106}Ru etc., corrosion products, such as Fe, Co, Ni, Cr, Mn, etc., unextracted actinides, such as U, Pu, Am, Cm, etc., and process chemicals, such as sodium nitrate and phosphate. The composition of HLLW generated during reprocessing of SNF depends on various factors, such as type of fuel, cladding material, burnup, time of storage and reprocessing schematics.

Identification of matrices with lower temperatures of melting and tolerance for higher loading in terms of salt content as well as radioactivity content are important for the selection of a matrix. These conditioned materials are contained in high integrity storage units which are subsequently over-packed. These over-packs containing conditioned wastes are stored under surveillance for periods ranging from 50 to 100 years so that these wastes would decay down to a level such that the heat generation is low and the waste form can be stored in geological disposal sites to enable monitoring for a sufficient period. The candidate matrices for immobilization of the waste are:

- Calcines. Calcines are produced by dehydration and denitration of waste with no appreciable addition of chemicals. They are amorphous which is not considered suitable for long term disposal.
- Glasses. These are the most extensively studied and used waste form. The loading limits arise from the heat generation from the FPs, and from the concentration limits for the elements, above which the glass suffers devitrification.
- Ceramic matrices. They tolerate higher salt content and can also remain stable for extended periods of time in a geological environment.

The most widely employed form of HLLW conditioning is the vitrified glass matrix for which the technology is at an advanced stage in many countries. There are two types of glasses, namely phosphate and borosilicate glass.

Sodium borosilicate based vitreous matrices have been adopted for de facto industrial scale processing in all countries, whereas in the Russian Federation the aluminophosphate matrix has been used to some extent. A phosphate glass matrix was found to be attractive for waste containing molybdenum and sulphate. Alhough its formation temperatures are low, the process is very corrosive and the products are inferior from the point of view of long term durability.

Minor compositional adjustments are carried out on the matrix depending upon the chemical and radiochemical composition of the HLLW. Vitrified waste products made from simulated waste compositions are extensively evaluated for chemical durability, homogeneity, phase separation, thermal conductivity, viscousity, glass transition temperature, thermal stability, etc. The developed compositions of the glass are then tried out on plant scale, initially with chemically equivalent simulated waste. Some problematic elements in HLW in the case of vitrification are given in Table 5.

3.2.2.2. Conditioning of fast reactor fuel reprocessing high level liquid waste into glass

HLW from the reprocessing of spent fuel discharged from FRs will have far higher concentrations of noble metals compared to those from thermal reactors. Hence, the salt loading capacity of the matrix is likely to be reduced, resulting in an increased number of waste canisters per tonne of spent fuel reprocessed.

To minimize doubling time, fast breeder reactor (FBR) spent fuels will be reprocessed with minimum off-reactor cooling periods in some countries. Extended storage of this waste will call for large waste tank farms and the economics of storage will dictate the time of vitrification. In the case of immediate vitrification (~20 months of cooling period both in vessel and out of vessel), the HLLW will have an activity of ~400 Ci/L compared to

TABLE 5. PROBLEMATIC ELEMENTS IN A VITREOUS MATRIX

Element	Limiting solubility (kg/100 kg of glass)	Impact on vitreous product
Platinum group metals (Ru, Rh, Pd, Tc)	1.0	Precipitate out as a separate crystalline phase and act as nuclei for devitrification; 10^4–10^6 times more conductive than glass.
Rare earths + zirconium	5.0	Refractory in nature, leading to high pouring temperature; Separate out as a crystalline phase and act as nuclei for devitrification.

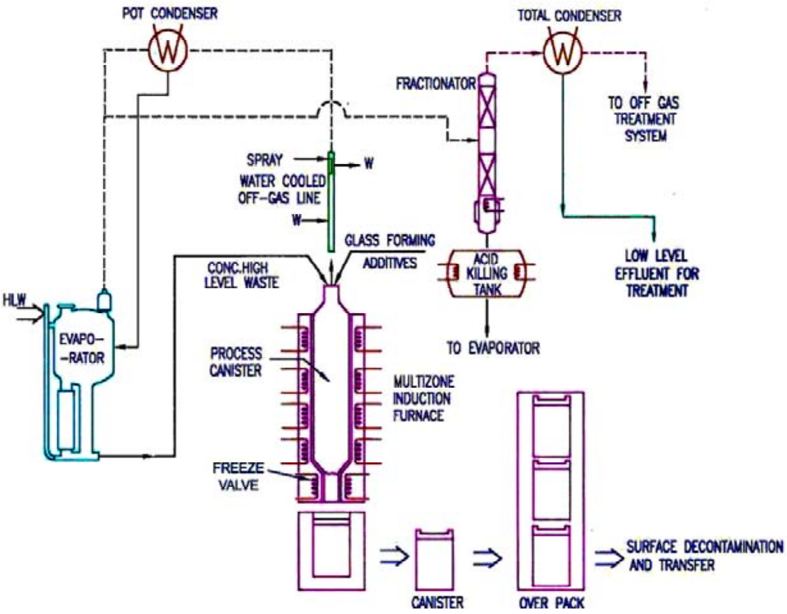


FIG. 6. Typical flow diagram of the vitrification process by induction heating.

~100 Ci/L after three years. Besides higher levels of activity, immediate vitrification will have to address the concerns arising from the volatalization of ^{106}Ru.

3.2.3. High level waste vitrification processes

Vitrification is almost a reference process for conditioning high level aqueous effluents. In this process, the solid content of the effluent is incorporated in a borosilicate glass matrix (even though phosphate glass is also used in some countries) at a temperature of around 1000°C.

The vitrification may be attained in a one step process, where the liquid waste is directly injected into the molten borosilicate bath. It can also be achieved in two steps, the first one consisting of evaporation and calcinations, and the second one involving the incorporation of the calcined residue in glass. The waste elements, thus, get fixed in the glass structure on an atomic scale.

3.2.3.1. Induction melting

A typical vitrification process (Fig. 6) consists of heating by induction of waste and glass forming additives in an induction furnace. The susceptor and the process pot are made of a high Ni–Cr alloy (Inconel-690) so as to withstand high temperature, and oxidizing and corrosive conditions. The calcined mass is fused into glass at about 950°C and is soaked at 950–1000°C for 8 h to achieve homogenization. The molten mass is then drained into

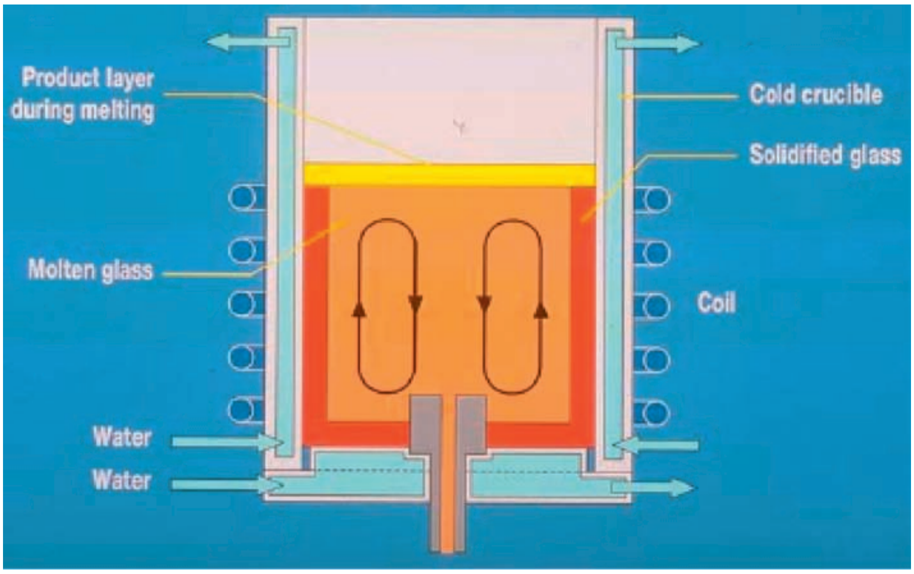


FIG. 7. *Cold crucible induction melting.*

stainless steel canisters. The vitrified waste canisters are further enclosed in secondary stainless steel containers called over-packs. Each canister may contain radioactivity up to 10^6 Ci, generating about 3–4 kW of decay heat, and hence they need to be cooled continuously to maintain a centreline glass temperature below the softening temperature in order to avoid devitrification [40].

3.2.3.2. Joule heated ceramic melter

Due to the problems of corrosion of melter walls and heating electrodes as well as the low throughput of induction melters, Joule heated ceramic melters (JHCMs) were developed. This technology has distinct advantages over conventional induction melters in the form of better product durability due to a higher achievable processing temperature, and higher throughput on account of continuous operation [41].

3.2.3.3. Cold crucible induction melting

The development of cold crucible technology enabled overcoming the problems associated with corrosion of critical parts of the vitrification equipment. The crucible is a water cooled melter in which the glass frit and calcined waste are melted by direct high frequency induction. High specific power is directly transferred by induction to the melt and it allows high operating temperatures. The cooling of the melter wall produces a solidified glass layer that protects the melters' inner wall from corrosion and other high temperature effects. Mechanical stirring of the melt makes it possible to get a uniform composition of the glass product, and high throughput. In view of its higher temperature capability, longer lifetime, smaller overall dimensions, and easy dismantling and disposal, cold crucible induction melting (CCIM) technology (Fig. 7) is being adopted in several countries.

The characteristics that make this technology very attractive are very long melter life, no need for continuous operation of the facility, virtually no limits on upper temperature and high waste throughput. This vitrification technology is also less sensitive to waste composition [42].

3.2.4. Management of high level waste from pyrochemical reprocessing

Two types of waste forms are envisaged in the pyrochemical reprocessing of FR fuel in molten salt baths [43]:

- A metallic waste form to process the cladding hulls, Zr and noble metal FPs left behind in the anode basket of the electro-refiner;

— A ceramic waste form to immobilize the alkali, alkaline earth and rare earth FPs along with a fraction of the salt bath.

3.2.4.1. The metal waste form

The salts sticking to the contents of the anode basket are removed by distilling at 1100°C. Extra Zr is added and the metallic mixture melted at 1600°C in argon in a casting furnace to form ingots of a durable stainless steel composition containing up to 15 wt% of Zr. This constitutes the metal waste form, considered fit for disposal in geological repositories [44]. The microstructure of the SS–15Zr alloy consists of an iron-rich solid solution (ferritic phase) and an intermetallic phase $Zr(Fe,Cr,Ni)_{2+x}$. The actinides are found primarily in the intermetallic phase and, hence, are retained well within the matrix (no separate actinide phase is formed). The noble metals are distributed between the intermetallic phase and the ferritic phase [45]. No discrete noble metal phases are formed. In a typical experiment, the ingot of the waste form had a diameter of 23.75 cm (9.5 inches) and a thickness of 10–15 cm (4–6 inches), and weighed 30–40 kg. Prolonged leach tests (static immersion in leaching solution at 90°C) and electrochemical linear polarization measurements showed negligible corrosion of the waste form.

3.2.4.2. The ceramic waste form

The FPs are removed from the chloride melt by ion exchange and occlusion in zeolites (microporous crystalline aluminosilicates). The molten salt containing the FPs is equilibrated with zeolite 4A at 500°C. A portion of the cations is ion-exchanged into the zeolite matrix, and a portion (along with some salt) is occluded in the zeolite cavities. The salt-laden zeolite is further treated with a glass frit, and is subjected to hot uniaxial pressing (HUP), hot isostatic pressing (HIP) or pressureless consolidation. This treatment converts the powder mix into a glass bonded sodalite monolith, considered suitable for disposal in a geological repository. This is called the zeolite waste or glass-bonded zeolite waste or, simply, the ceramic waste form [46].

4. EMERGING TECHNOLOGIES FOR THE BACK END OF THE FAST REACTOR FUEL CYCLE

4.1. GENERAL OUTLOOK

While there is a consensus that FRs with a closed fuel cycle are required for the sustainability of nuclear energy, it is also necessary to note that any technology for electricity production needs to be safe as well as competitive in order to be successful. A number of Member States are, therefore, engaged in developing innovative FR systems, with the specific objectives of enhancing their safety and reducing the cost. The respective fuel cycle technologies are also being studied and are mainly aimed at reduced waste generation, reduction in toxicity of wastes through recovery of MAs and LLFPs, and development of better matrices for ultimate disposal of waste.

An overview of the fuel cycle programmes pursued by various Member States indicates clearly that reprocessing and waste management continue to be intense areas of research. Japan is pursuing studies on the new extraction system for TRU element recovery (NEXT) process along with a simplified pelletization route for fuel reprocessing and fuel fabrication, respectively [47]. The Advanced Fuel Cycle Initiative (AFCI) programme in the USA, as well as the Global Nuclear Energy Partnership (GNEP) programme, indicate that there is an emphasis on developing reprocessing/recycling technologies that would avoid separation of pure Pu in order to enhance proliferation resistance. In France, the COEX^{TM} process is being developed for the co-management of U and Pu from spent fuel to the fabrication of MOX fuel [48, 49].

The emerging trend is presently on development of various partitioning technologies which will aid in the immobilization of the actinides and LLFPs in appropriate matrices. Alternatively, the partitioned products could be transmuted in either FRs or ADSs. An innovative approach is to adopt pyrochemical or pyrometallurgical technologies.

4.2. PARTITIONING AND TRANSMUTATION

The area which has been receiving the highest level of attention is perhaps P&T of MAs and LLFPs, as is evident from the information exchange meeting on P&T international conferences held recently [50–52].

Partitioning of the MAs and some selected FPs is a method which would reduce the long term radiotoxicity of the residual waste components. The recovered MAs and LLFP nuclides could be recycled to the reactor for transmutation to short lived isotopes. This technique would reduce the long term radiotoxicity hazard in the HLW and shorten the time interval necessary to keep the actinide containing wastes confined in a deep geologic repository. P&T is, in principle, capable of reducing the radiotoxicity period, although a number of practical difficulties remain to be surmounted.

Future plants for back end operations can be expected to have an integrated approach to reprocessing and waste management; that is, the flow sheets have the possibility to integrate recovery of MAs and FPs in addition to fissile materials. Technologies for fabricating dedicated fuels incorporating MAs are being pursued in several countries, such as France, Japan, the Russian Federation and the USA, as well as in the European Union. The coming decades will see an emphasis on closure of the fuel cycle in the real sense, wherein the LLFPs and MAs from LWR and PWR spent fuels, would be recycled in FRs.

4.3. PARTITIONING TECHNOLOGIES

In the following sections, a general outlook is provided for the additional process steps which are generally applied to the ‘waste stream’ of a classical PUREX process after separation of the main (U and Pu) fissile content. This ‘waste’ may be characterized as a mixture of different radioactive FPs, actinides other than U and Pu, and a number of stable FPs, some of which may be of significant economic value. The basic stages of the possible additional separation/partitioning steps include:

- Co-extraction of MAs and lanthanides;
- Separation of MAs from lanthanides;
- Separation of Cm from Am;
- Extraction of short lived FPs, such as Cs and Sr, from residual waste for separate treatment;
- Extraction and purification of stable isotopes from the lanthanide fraction (mainly metals of the platinum group and rare earths).

Several extractants, such as carbamoyl methyl phosphine oxide (CMPO), diamides, diglycol amides and separation schemes, have been developed for MA recovery from HLLW. Several reagents, such as triazenes, Cyanex-301 and vinyl pyridine resins, have been developed for the separation of actinides and lanthanides after the MA extraction step.

4.3.1. Co-extraction of An(III) and Ln(III)

4.3.1.1. TRUEX process

The TRU element extraction (TRUEX) process, studied in the USA, Japan, the Russian Federation, Italy and India, which was initially developed by Horwitz (ANL) and Schulz (Hanford) in the USA in the 1980s, is based on the use of the CMPO extractant. The advantages of the TRUEX process are the following:

- It can extract An and Ln salts from acidic feeds;
- Its efficiency has been demonstrated with genuine HLW;
- A lot of data have been generated worldwide

The main drawbacks of the TRUEX process are the:

- Necessity to use a large concentration of TBP as a solvent modifier added to the solvent to prevent third phase formation;
- Need for complex stripping agents for metal ions;
- Solvent cleanup for reuse has not been demonstrated on a large scale;
- Management of salts and complex effluents.

4.3.1.2. UNEX process

The universal extraction (UNEX) process is being developed for the simultaneous extraction of caesium, strontium and actinides from acidic solutions by the Idaho National Laboratory (INL), ID, USA in collaboration with the Khlopin Radium Institute (KRI), Russian Federation. The UNEX process was first invented by researchers in the Czech Republic and later tested in the Mayak plant in the Russian Federation. This process uses a synergistic mixture containing the hexa-chloro derivative of cobalt di-carbollide (CCD), polyethylene glycol (PEG) and diphenyl-carbamoyl-methyl-phosphine oxide (d-CMPO) that extracts Cs and Sr as well as several actinides and lanthanides from HLW. CCD extracts Cs, PEG is to promote Sr ion extraction, and the CMPO extractant is adequate for the actinide/lanthanide fraction. Actinides and lanthanides are stripped with di-ethylene-triamino-penta-acetic acid (DTPA).

4.3.1.3. DIAMEX process

The diamide extraction (DIAMEX) process is studied in France, Germany, India, Italy, Japan and the USA, and is based on the use of a malonamide extractant. The main advantages of the process are:

- An and Ln salts are extracted from acidic feeds;
- Its efficiency has been demonstrated on HLLW (in France);
- No secondary solid wastes are generated owing to the “CHON” character of the malonamide extractant.

The main drawback of the DIAMEX process lies in the partial co-extraction of palladium (Pd) and ruthenium (Ru) with the MAs. A process based on a new type of diamide, i.e. tetra-octyl diglycol amide (TODGA), a terdendate ligand with better affinity for An(III) than the malonamide, has been developed at the Japan Atomic Energy Agency (JAEA), Tokai, Japan. This reagent is being tested in France and at the Institute for Transuranium Elements (ITU), Joint Research Centre, European Commission with HLW.

4.3.1.4. TRPO process

The TRPO (tri-alkyl phosphine oxide) process is based on the use of a mixture of tri-alkyl phosphine oxides ($R_3P(O)$, with R = alkyl groups) as extractant. This process has been tested successfully in China with HLW. Its main drawbacks concern the necessity to:

- Adjust the feed acidity to low level;
- Use a concentrated nitric acid solution for An(III) + Ln(III) stripping, which complicates the subsequent An(III)/Ln(III) partitioning step.

4.3.2. Separation of An (III)/Ln(III)

4.3.2.1. TALSPEAK and CTH processes

The TALSPEAK process is studied in the Russian Federation, Sweden and the USA. It was originally developed at the Oak Ridge National Laboratory (ORNL), USA, in the 1960s and then adapted (CTH process) at Chalmers University, Goteborg, Sweden. This can be considered as the reference process for An(III)/Ln(III) group separation. It is based on the use of di-ethyl hexyl phosphoric acid (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 a mixture of An(III) + Ln(III) under the action of an aqueous solution containing DTPA and a hydroxocarboxylic acid, such as lactic, glycolic or citric acids.

The advantages of this process are:

- Large amounts of data have been generated worldwide;
- Its good efficiency.

Among the main drawbacks one can cite are the:

- Necessity to reduce the acidity to low level;
- Limited solvent loading of metal ions;
- Solvent cleanup process is not yet defined;
- Management of salts and complex effluents.

4.3.2.2. CYANEX 301 process

The CYANEX 301 extractant is a dialkyl-di-thiophosphinic acid. Its use for An(III)/Ln(III) was first proposed by Zhu in China in 1995 and also studied in Germany and the USA. The main advantages of the process are:

- High efficiency for An(III)/Ln(III) separation;
- The efficiency of the process has been tested with An(III)–Ln(III) mixtures.

Nevertheless, for an efficient use of this process, the feed solution should be adjusted to pH3–5, which is not easily achieved in the industrial environment. Moreover, the solvent’s instability and difficulty in cleanup are also weak points.

4.3.2.3. ALINA process

To manage the main drawbacks of the CYANEX 301 process mentioned above, Odoj and Modolo, in Germany, proposed the use of a synergistic mixture made of bis(chlorophenyl)dithio-phosphinic acid octylphosphine oxide to perform the An(III)/Ln(III) group separation. While 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. The ALINA process was successfully tested with HLLW. The drawbacks of this process are:

- The solvent cleanup process is not yet defined;
- Generation of P- and S-bearing wastes (from the degraded extractants) which should be managed.

4.3.2.4. SANEX concept (neutral N-bearing extractants) and BTP

After the discovery by Kolarik in Germany of the potential of bis-triazinyl-1,2,4-pyridines (BTPs) for An (III)/Ln (III) separation, a process was developed and tested in the frame of the European NEWPART project. Successful hot tests were carried out both at the Alternative Energies and Atomic Energy Commission (CEA)/Marcoule and at the ITU, Karlsruhe using the n-propyl-BTP. The high efficiency of the BTP process was confirmed.

One should also mention that the feed of the n-propyl-BTP process can be acidic ($\{HNO_3\}$ = 1 mol/L). Nevertheless, while this system seems very promising, instability of the n-propyl-BTP extractant was observed. As a consequence, efforts are underway in the European Union to modify the solvent formulation to overcome this major drawback.

4.3.2.5. TMAHDPTZ-octanoic acid

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 octanoic acid was developed at CEA/Marcoule. A process flow sheet was defined and successfully tested with HLLW with good efficiency. The main drawbacks of this process are:

- Adjustment of acidity to low level is required;
- Management of the secondary wastes is not yet defined.

4.3.3. Combined processes for An/Ln extraction and separation

4.3.3.1. DIDPA process

This process for MA partitioning is based on the use of di-isodecylphosphoric acid (DIDPA). The separation of the TRU elements is done by successive stripping from the loaded solvent, including the use of DTPA complexing agent for An(III)/Ln(III) separation (TALSPEAK-like process; see Section 4.3.2.1). The DIDPA process was recently tested successfully in the BECKY hot-cell at NUCEF (JAEA, Tokai-Mura).

The drawbacks of this process are:

- Adjustment of acidity to low level is required;
- Solvent degradation and its delicate cleanup, to be addressed;
- Limited solvent loading with metal ions.

4.3.3.2. SETFICS process

The SETFICS (solvent extraction for trivalent F-elements intra-group in CMPO-complexant system) process constitutes a modification of the TRUEX process (see above) based on the use of the extractant n-octyl(phenyl)di-isobutyl-CMPO. The separation of TRU elements is done by successive stripping from the loaded solvent, also including the use of DTPA for An(III)/Ln(III) separation [53]. This process was tested with HLLW from the FR irradiated fuel in Japan [54–70].

The drawbacks of this process are:

- Limited separation efficiency for lanthanides;
- Management of salts and DTPA containing effluents.

4.3.3.3. PALADIN process

The process is being studied in France and it is based on the use of a mixture of extractants: a malonamide (DIAMEX process extractant) + HDEHP, the extractant of the TALSPEAK process. In low acid media, HDEHP serves as an extractant, while at the metal nitrate extraction step, carried out with acidic feeds from 3–5 mol/L in nitric acid, trivalent An and Ln are extracted with the malonamide. The An(III)/Ln(III) separation is performed by contacting the loaded solvent with a pH adjusted aqueous solution containing DTPA selective trivalent actinide complexing agent [55, 56].

This process was recently successfully tested in the ATALANTE facility (CEA/Marcoule). The drawbacks of this process are:

- Necessity to use two extractants;
- Adjustment of acidity to low level is required;
- Need for several scrubbing steps because of co-extraction of numerous ions;
- Solvent cleanup not yet defined;
- Management of salts and complex effluents.

4.3.4. Separation of Am/Cm

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.

4.3.4.1. SESAME process

This is being studied in France and Japan, and it utilizes the situation that in strong oxidizing conditions, Am can be oxidized from Am(III) to Am(VI). The oxidation can be achieved by electrolysis in the presence of heteropolyanions (HPAs) as a catalyst. Am(VI) can then be easily separated from Cm(III) by extraction with reagents such as TBP. The principle of the process was developed at CEA/Marcoule. At Hitachi and JAEA, Japan, the oxidation of Am to Am(VI) is obtained by the use of ammonium persulphate, and Am(VI) is then extracted by TBP. The separating americium from curium (SESAME) process exhibits a great efficiency for Am/Cm separation. Much experience was obtained by CEA on a laboratory scale [57–70]. Nevertheless, industrialization of the process faces difficulties such as:

- Instability of Am(VI);
- Generation of secondary solid waste (made of HPA constituents).

4.3.4.2. Am(V) precipitation

This process, which is being studied in Japan, uses selective precipitation. The selective precipitation of double carbonate of Am(V) and potassium (K) is one of the oldest methods for Am/Cm or Am/Ln separation. It was developed at the end of the 1960s in the USA [71]. In this method, the mixture of Am(III) and Cm(III) is dissolved in a 3.5 mol/L K_2CO_3 solution. After chemical or electrochemical oxidation of Am(III) to Am(V), Am(V) precipitates from the solution as the solid crystalline $K_5\,AmO_2\,(CO_3)_3\,nH_2O$, while Cm(III) remains in solution. After filtration, Am is separated from Cm. This process has the following advantages:

- Simplicity;
- Selectivity for Am.

The main drawbacks of the process are:

- Substantial losses of Am and Cm;
- Generation of large amounts of secondary wastes.

4.3.5. Separation of long lived fission products

4.3.5.1. Separation of iodine (^{129}I)

The separation of iodine is done just after the spent fuel dissolution step within the PUREX process. Oxidation of the iodide ion, I^-, into elemental iodine (I_2) induces its transfer to the dissolver off-gases from which iodine can be recovered through alkali washing.

4.3.5.2. Separation of 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), U(VI) and Pu(IV) by TBP is well known. Thus, the separation of the soluble fraction of Tc may be achieved through a solvent scrubbing step in the course of the implementation of the PUREX process as is done at the AREVA and Rokkasho 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.

4.3.5.3. Separation of caesium and strontium

Many processes were developed worldwide in this field, including:

- The use of inorganic sorbents, such as JAEA's 4-group partitioning process;

Crown-ether extractants, such as those used in the Sr extraction (SREX) and Cs extraction (CSEX) processes developed at ANL [72];

- Cobalt dicarbollide extractants, developed in the Czech Republic, Russian Federation and Western Europe [55, 73].
- Most of these processes were successfully tested with HLLW [74–82].

4.3.5.4. Advanced separation methods using selective ligands

The extraction techniques being developed at INL and ORNL use macrocyclic polyethers (crown ethers) which act as ligands to form stable complexes with salts of numerous metals [83]. The cyclic structure of the crown ethers creates a circular cavity of oxygen atoms which may correspond in size to a certain cation. The relative diameters of the cavity and of a cation determine the stable complex (host–guest interaction). $DCH_{18}C_6$ in n-alcohols (1-octanol) is well studied for the extraction of strontium in liquid–liquid separations. INL is experimenting with 4',4,(5')-di-(t-butyldicyclohexano)-18-crown-6.calix(4)arene-bis-(tert-octylbenzo-crown-6) ($BOBcalixC_6$) and 1-(2,2,3,3-tetrafluoropropoxy)-3-(4-sec-butylphenoxy)-2-propanol in a branched aliphatic kerosene.

In general, crown ethers have been selected for the removal of strontium, and calixarenes have been selected for separations of caesium. The ligand (3-N-(6-carboxy-methyl-picolinamide)propyloxy)calyx(6)arene(1) was selected from a series of calixarene-based picolinamide ligands as a possible candidate to be used in a small scale process for An/Ln separation under the option of advanced reprocessing of irradiated nuclear fuel. The proposed process uses a chemically complex, caesium-selective solvent system in which the caesium-selective component is a calixarene crown ether, with two crown ethers bridging the calyx(4)arene component. The specific material proposed for use at the Savannah River Site is called BoBcalixC6. Other components in the solvent system include a proprietary modifier, a tertiary amine and a hydrocarbon diluent [84, 85].

Several research groups have been synthesizing a large number of extractants (mainly calixarene and bis-crown ether derivatives) for the removal of strontium, actinides and the separation of actinides and lanthanides. Strontium complexation and extraction experiments indicate that methyloxy- and octyloxy-calyx(8)arene-octa-diethyl amide are good extractants for strontium from an acidic medium with a high selectivity with respect to sodium [81, 86–88].

4.3.6. ARTIST process

The amide based radio resources treatment with interim storage of TRU elements (ARTIST) process comprises two main steps: an exclusive isolation of uranium, a major constituent of the spent fuel; and a total recovery of TRU elements with several optional processes. Uranium separation is achieved by using a branched alkyl mono-amide extractant, which is often considered to be a promising alternative to TBP. Both actinide fractions are respectively solidified by calcination and sent for interim storage for future utilization. These separations are achieved using amidic extractants. The TRU element product is accompanied by lanthanides (Ln) and is, thus, more resistant to proliferation. The options are:

- Extractive recovery of Pu;
- Group separation of TRU elements and lanthanides;
- Isolation of Cs and Sr, respectively.

The first optional process is to be operated for producing MOX fuel. The separation of TRU elements from lanthanides is operated to obtain the target material for transmutation of TRU elements in an ADS or FR. The

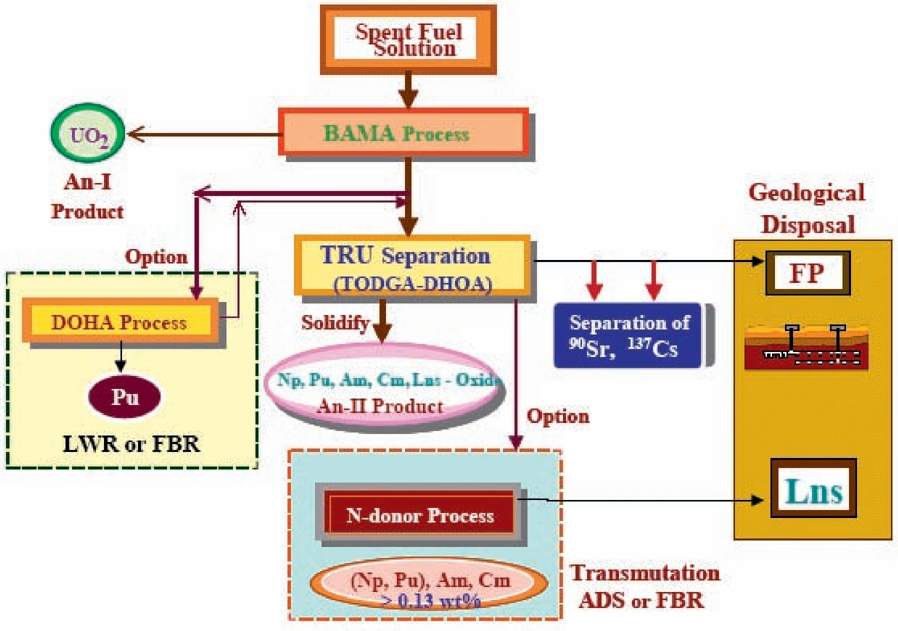


FIG. 8. Schematic of the ARTIST process.

separation of Cs and Sr is effective for reducing the cost of disposal of FPs because ^{137}Cs–^{137}mBa and ^{90}Sr–^{90}Y are the major heat-generating nuclides among FPs for the initial several hundred years. The main concept of the ARTIST process is schematically shown in Fig. 8.

4.4. CERAMIC MATRICES FOR SELECTED ACTINIDES AND LONG LIVED FISSION PRODUCTS

An attractive alternative to waste vitrification may be waste incorporation into mineral-like crystalline structures. The basic approach to achieving this is to treat waste streams with calculated quantities of chemically active additives, so that upon heat treatment and consolidation, a dense polyphase ceramic assemblage is formed that chemically binds the waste elements into known crystalline phases. These methods are often called the waste forms of the future, an emerging alternative or complement to the glass waste form [89–96].

A titanate based synthetic rock (SYNROC) is the most widely studied crystalline waste form. SYNROC was developed jointly by the Australian National University and the Australian Nuclear Science and Technology Organisation (ANSTO) in the late 1970s [90].

It is an assemblage of four mutually compatible titanate minerals, namely zirconolite ($CaZrTi_2O_7$), perovskite ($CaTiO_3$), hollandite ($BaAl_2Ti_6O_{16}$) and rutile (TiO_2). These minerals incorporate nearly all HLW elements in their crystal structure as dilute solid solutions. Many natural minerals containing radioactive elements have survived in a range of geological environments for periods up to 2000 million years. Experimental observations on these natural minerals show that they are extremely durable and resistant to chemical attack even under hydrothermal conditions [91].

Radioactive waste elements occupy specific lattice positions in the crystal structure of the minerals constituting the assemblage, depending on the atomic/ionic size, charge and bonding conditions.

The candidate mineral phases in any SYNROC type assemblage usually have relatively complex crystal structures with coordination polyhedra of different sizes and shapes; this provides different substitution schemes with a charge balance for the HLW elements, with or without minor changes in the crystal structure. The principal minerals of the SYNROC assemblage can each accommodate a range of radionuclides. Typically, hollandite can incorporate such elements as Cs, Rb and Ba; zirconolite can host U, Zr, Np, Pu and rare earths; perovskite ($CaTiO_3$) can take up Sr, Np and Pu. Rutile helps in the micro-encapsulation of minor alloy phases; it also increases the mechanical strength of the waste form. In some formulations, pyrochlore ($A_2B_2O_7$, A and B being cations), which has a crystal structure closely related to the zirconolite structure, may become the actinide host phase. The mineral phases formed and their relative amounts are determined by the additives.

The procedure reported for the SYNROC demonstration plant in Australia consists of mixing the aqueous slurry of oxide additives (precursors) with the waste solution, drying and calcinating the mixture in a reducing atmosphere at 1023 K, cold pressing the calcines in the stainless steel bellows, and finally hot pressing at 1473 K and 14 MPa pressure [97].

In recent years, a procedure involving a melting step at 1673 K, subsequent slow cooling to 1373 K in 4.5 h and further cooling to room temperature in 16 h has been proposed as a preparative procedure. Application of CCIM has also been reported. SYNROC has reached a certain level of development as a futuristic waste form, though not on a commercial scale with actual waste.

Radiation induced amorphization over geological time is a distinct possibility with the actinide bearing mineral phases. This is called ‘metamictization’. This can give rise to a volume increase (5–10%) and even microcracking of the waste form. A fractured waste with its increased surface area may increase its leachability; but a redeeming feature is that defect annealing and re-crystallization phenomena can also take place simultaneously.

Durable single phase matrices can be designed to immobilize particular waste streams containing one element or a group of chemically similar waste elements. Taking into account that advanced partitioning technologies enable the separation of rare earths and TRU elements from liquid HLW, this option becomes rather attractive. In France, zirconolite, monazite, thorium phosphate diphosphate and apatite ceramics were being developed for conditioning actinides; hollandite and apatite for caesium; titanate and phosphate matrices, and metal matrices for technetium; and apatite for iodine.

4.5. TRANSMUTATION SYSTEMS BASED ON ACCELERATOR DRIVEN SUBCRITICAL SYSTEMS

ADSs are perceived as inherently safe reactor systems that can effectively burn MAs and LLFPs. Different configurations of ADSs are being considered by various countries. In India, ADSs are also being developed as a means for thorium utilization. Studies have been reported comparing FRs with ADSs with respect to their efficacy for the transmutation of MAs and LLFPs [98].

Among various strategies for the fuel cycle, recovery and recycling of plutonium and other actinides in reactors is recognized as the best approach for reducing the toxic waste burden on the environment. Compared to the once-through fuel cycle, closed fuel cycle strategies can achieve a more than hundredfold reduction in long term waste radiotoxicity. However, this demands multiple recycling of the fuels, high fuel burnups, and very low reprocessing and fuel fabrication losses. Regarding actinide waste production and technological aspects, the FR–TRU element and the ADS–MA schemes are equally attractive. The ADS, however, has the advantage that it can burn pure MAs while avoiding deterioration of core safety characteristics.

Important technological challenges arise for the fuel cycle of a transmutation system using either an FR or ADS. This is due to the presence of high concentrations of MAs, which results in high decay heat and neutron emission. Deep burning of the fuel is an important requirement for effective transmutation.

4.6. EFFICIENCY OF MINOR ACTINIDE UTILIZATION IN FAST REACTORS

The options available for burning MAs in FRs include recycling Am and Cm heterogeneously in special ‘target’ pins or recycling homogeneously by incorporation in the fuel. The burnup achieved with the FR fuel is usually limited to about 250 GW/t HM. Thus, an effective actinide burner cannot operate in a once-through mode but rather requires a fuel cycle, which allows the fuel to be recycled many times. For the maximum burnup of 250 GW/t HM and recycle intervals of six years, it would take 96 years to achieve a hundredfold waste mass reduction. Thus, it can be concluded that an effective transmutation system needs a fully closed fuel cycle in which all actinides are recovered with nearly 100% efficiency and then recycled. To fully exploit the potential of such a system, it must be operated for an extended period of at least 100 years [5, 98, 99].

4.7. PYROCHEMICAL REPROCESSING SCHEMES

Pyrochemical processes were first investigated in the 1950s as an alternative to PUREX to increase the radiation resistance and stability of material used in extraction processes. An initial pyrochemical process was constructed at the Experimental Breeder Reactor II (EBR-II) in the USA during the 1950s. Later, many concepts for pyrochemical partitioning were developed and, in some cases, pilot plants were built and operated. Studies are under way in the USA, the United Kingdom, the Russian Federation, the Republic of Korea, Japan, India, Germany, France and the European Commission/ITU on various aspects of pyrochemical reprocessing technology. Studies are under way in France, the USA, the United Kingdom, Germany, the Russian Federation, Japan and the Republic of Korea on various aspects of pyrochemical reprocessing technology. Areas receiving high attention include direct oxide reduction on an engineering scale, optimization of the electro-refining and treatment of wastes. For the coming decade, it is expected that developments in this area will continue to focus on pyrochemical processes as alternate candidates for reprocessing. It should be noted that an integrated approach to fuel fabrication and fuel reprocessing through the ‘pyro’ route, as in the case of an IFR, has not been pursued so far by any country. India has planned to build FRs based on metallic fuels with integrated fuel cycle plants by 2025.

Several techniques that were based on molten salts were (are being) developed, namely melt-refining, volatilization, gas–solid reaction, fractional precipitation, vacuum distillation, electrodeposition, electro-refining, electrowinning and others. This technology, which has lower separation factors, can take advantage of the fact that FRs can accept higher levels of impurities in the fuel and, hence, possibly decrease the cost of reprocessing. The advantages of some of these pyro-processes are:

- Adaptability to reprocess spent fuels, including MA based fuels, with shorter cooling periods. This advantage principally relates to future fuel cycles.
- Ability to co-recover actinides largely in a single process.
- Compact plants that can accept several forms of fuels.
- Very short turn-around time for the fuel, and associated cost savings from the resulting reduction of otherwise large fissile material inventories.
- Generation of minimum TRU element waste.
- Very high intrinsic proliferation resistance for the fissile materials owing to:
  - Limited purity of the end product, which limits its direct use in nuclear weapons;
  - A built-in isotopic barrier because of the presence of isotopes with high decay heat, high spontaneous neutron yield and lethally high radiotoxicity;
  - Co-location of reprocessing and fuel fabrication facilities with the reactor.

However, the main challenges facing pyro-processes are the requirement for an oxygen- and moisture-free plant environment, and the need to develop materials that will not only withstand high radiation levels but have excellent resistance to high temperature corrosion in molten metals and molten halide salts. The development of pyrochemical processes also requires the development of state of the art equipment that is suitable for applying such processes at an industrial scale. The path to industrial utilization is, thus, perhaps longer for pyro-processes than for advanced aqueous processes [100–120].

There are several research efforts looking for ways in which different fuel cycles, both existing and future, can efficiently complement one another for sustainable nuclear energy development. Aqueous processing methods have very high throughputs and sufficient industrial maturity to handle large volumes of discharged fuels from LWRs. However, they are less suitable for reprocessing FR fuel with short turn-around times. Pyrochemical processes can handle spent fuels with short cooling times as well as with very high MA content in a significantly smaller processing facility. Furthermore, these processes can recycle TRU element waste, thus reducing the volume of TRU element waste that needs disposal. Due to impure product recovery, the process is very much proliferation resistant. Thus, synergistic combinations of aqueous partitioning steps with pyrochemical processing, as depicted in Fig. 9, could be a promising option in the future.

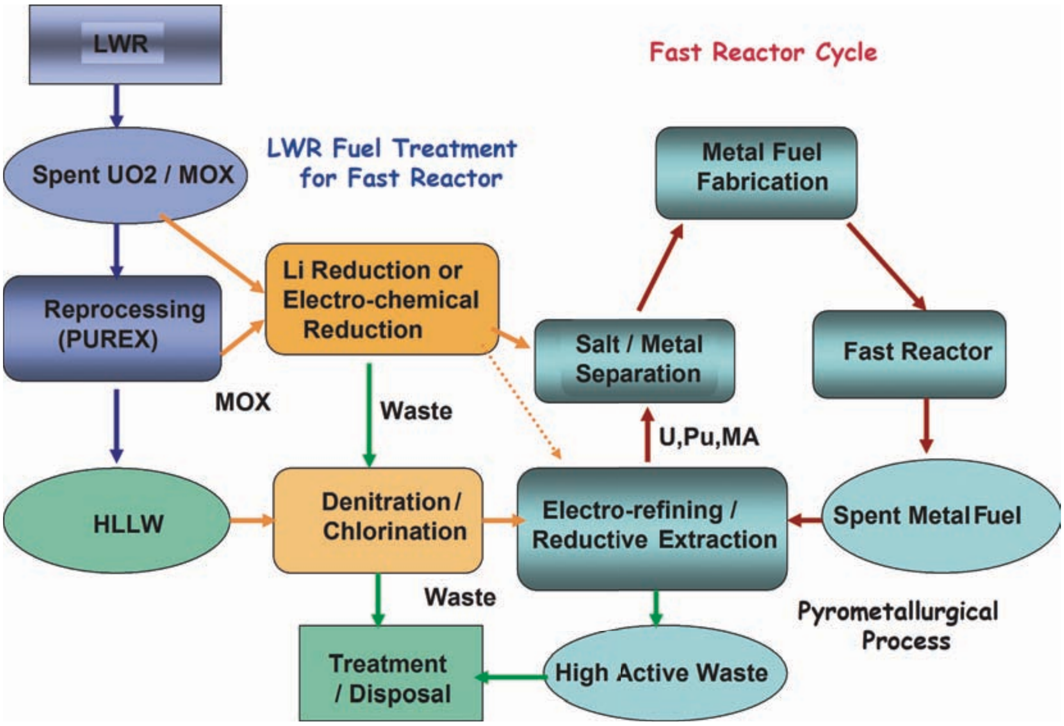


FIG. 9. Actinide recycling system by integrated oxide fuel treatment and recovery of actinides in HLLW.

4.8. SUPERCRITCAL FLUID DIRECT EXTRACTION (SUPER-DIREX)

The supercritical fluid direct extraction (Super-DIREX) system is also a candidate for study. In this process, fuel materials are directly extracted into a supercritical carbon dioxide (sf-CO_2)-TBP-nitric acid mixture from the powdered spent fuel. While this system would probably have economic advantages, many more studies are necessary to evaluate its potential.

4.9. FLUOREX PROCESS

A new FLUOREX technology has been proposed in Japan for treatment of SNF from thermal reactors and future FRs. The proposed system is a hybrid system that combines fluoride volatility methods and extraction methods. The system is expected to enable extraction of a pure U/Pu mixture with a DF > 10^7 and may also result in a pronounced reduction in the costs and waste amounts as compared to traditional methods. The final product of the FLUOREX process will be a Pu/U mixture (without further separation) directly suitable for fabrication of MOX fuel for thermal reactors and for interim storage [28, 31, 36, 121–123].

5. ENABLING TECHNOLOGIES

5.1. INTRODUCTION

This section addresses technologies for the nuclear fuel cycle with a focus on materials, robotics, process instrumentation and inspection technologies, as applicable to aqueous fuel reprocessing and waste management. It covers the developments that have taken place in the enabling technology areas of materials, welding, sensors, automation and robotics, process instrumentation and monitoring, and non-destructive evaluation, with a special emphasis on their applicability at various stages in the FR nuclear fuel cycle.

5.2. MATERIALS DEVELOPMENT

The availability of nuclear fuel reprocessing plants for uninterrupted operation depends on the quality and performance of critical engineering components, vessels and piping. To this end, materials selection, fabrication, operation, maintenance and inspection of various unit processes of reprocessing plants are of paramount importance in meeting the requirements of uninterrupted and safe operation of plants. This demand is much more stringent with respect to reprocessing plants for FBRs as they utilize high plutonium content fuels and achieve high burnup. Reprocessing plants are designed with the objective of minimizing failures, taking into account that leakages in pipes, vessels and equipment failures could lead to considerable down time of the plant. Material properties are of specific importance for improving the life of the equipment. High Pu content and higher targeted burnup of the fuel (>s150 GW·d/t HM) also require aggressive conditions for dissolution. Thus, the materials used for construction and components, such as electrodes, have to be designed to meet the demands of minimum failure and minimum maintenance of the plants.

A broad materials development programme has been in progress worldwide towards the development of construction materials for applications in spent fuel reprocessing plants [124].

Three main approaches are usually pursued in developing such materials:

- Enhancing the passive range in stainless steel by shifting the trans-passive conditions to nobler potentials;
- Improved alloys with better corrosion properties, achieved by stabilizing the passive surface films.

Stainless steel containing ultra low carbon and controlled amounts of impurities that segregate to grain boundaries has been developed. These alloys possess resistance against corrosion under diverse heat transfer conditions encountered in reprocessing plants.

To inhibit the grain boundary attack and eliminate trans-passive dissolution, an optimization process of the 304ULC SS type has been proposed by Japan [125].

This includes:

- Controlling major elements’ content (Cr and Ni);
- A decrease in Si content to between 0.1 and 0.2%;
- Limiting the minor alloying elements (P, S, B, N, etc.) through vacuum induction melting followed by electron beam melting;
- Modifying the microstructure (by thermo-mechanical treatment).

Although austenitic stainless steels of the AISI type 304L SS are the workhorse material for reprocessing plants operating with nitric acid, for most of the unit operations of PUREX based processes, international efforts have resulted in the development of advanced materials, such as nitric acid grade (NAG) special austenitic stainless steels: Ti–5%Ta, Ti–5%, Ta–1.8% Nb, Cr–W–Si alloy, Nb–W, zircaloy, etc. for highly corrosive nitric acid environments. Several incidences of failures of components made of AISI type 304L SS have been reported in SNF reprocessing plants when they were used in an HNO_3 medium beyond 8 N concentration and temperatures beyond 353 K. The failures have been attributed to inter-granular corrosion due to sensitization and segregation of impurities at grain boundaries, trans-passive dissolution of passive films and selective corrosion of the welds [126].

NAG austenitic stainless steels are alloys developed with:

- Controlled chemical composition of alloying elements;
- Modified microstructures leading to elimination of the weaker sites for passive film breakdown and dissolution;
- Enhanced strength against trans-passive dissolution leading to an extended passive state.

Several types of NAG alloys having compositions similar to AISI types 304L, 310L and several new proprietary alloys have been developed worldwide [127].

The choice of NAG SS for specific components in reprocessing plants requires attention to issues associated with fabrication, welding and maintenance. Welding of NAG alloys needs special welding consumables and requires establishment of strict welding procedures. Low carbon varieties of stainless steels have a high propensity for sigma phase and other precipitation during multipass welding of thick components (concentrators, evaporators), and it is well known that sigma phase preferentially dissolves in hot oxidizing nitric acid.

Corrosion problems due to high plutonium and neptunium in FR fuel reprocessing are severe. To address these, there are three approaches:

- Zirconium as the candidate material for dissolver and plutonium evaporators;
- Ti–5%Ta alloy as the candidate material for dissolver;
- NAG stainless steel for HLLW evaporators and acid concentrators.

Zr–Hf and Nb–W alloys are also potential materials for back end applications involving highly oxidizing conditions.

A new generation alloy has been developed in Japan by adding varying amounts of Ta and Nb to titanium, and an alloy with an optimum composition was obtained as: Ti–5% Ta–1.8% Nb [128]. The alloy exhibits significant reduction in corrosion rate, while testing in a boiling nitric acid medium. A similar alloy has been developed in India [129].

However, a holistic development of materials for equipment for the back end of the fuel cycle for FRs requires a detailed study of the corrosion and erosion behaviour in a wide variety of media representing various unit operations in reprocessing and waste management.

In addition to the construction material of equipment, a wide variety of other materials such as elastomers, cables, ceramics, etc. also need attention.

5.3. INSPECTION TECHNOLOGY

An aqueous reprocessing unit typically consists of piping systems, fuel dissolvers, evaporators, fluidic devices, mixer–settlers, pulsed and packed columns, solvent recovery systems, centrifugal contactors, dissolution units, waste storage tanks and remote handling systems. The structural integrity of all of these components is very important because various defects are possible during their service life due to exposure to hostile corrosive media. Defects also grow to critical sizes, hence disturbing the structural integrity of the components and, in this regard, non-destructive testing (NDT) plays a vital role [130–132]. A variety of NDT techniques are employed for detection, location and sizing of the defects.

In many instances, the well established NDT techniques for detection and evaluation of defects cannot be directly employed for the inspection of reprocessing plant components, essentially because of limited access and radiation. This necessitates the use of NDT methods, remote sensors and special gadgets for inspection of inaccessible components.

As corrosion is the most important degradation observed in reprocessing plants, several NDT techniques have been developed for detection and characterization of corrosion in reprocessing plant components, especially dissolver vessels, storage tanks and process vessels. NDT techniques, such as ultrasonic, laser triangulation, eddy current and flux leakage techniques, are popularly used. The choice of NDT technique depends on applicability, accessibility and suitability, in addition to the type of material inspected. It may sometimes be necessary to use a combination of two or more techniques in a complementary way.

5.3.1. Non-destructive testing techniques for on-line corrosion monitoring

Corrosion monitoring by using on-line corrosion rate measurement probes/sensors is required for evaluating the health of the various components. Components, such as electrolytic dissolvers for the dissolution of mixed carbide fuel of a fast breeder test reactor (FBTR), require high corrosion resistant materials, such as titanium, which showed a corrosion rate of less than 0.125 mm/a. However, the use of commercial purity titanium with an iron content of more than 0.05 wt% increases the corrosion rates beyond 0.5 mm/a, particularly in welded regions where metallurgical heterogeneities are generated after welding operations.

Since corrosion beyond 50% of the wall thickness of the dissolver vessel (6 mm to 3 mm) may not be permitted for operation in the plant, continuous on-line corrosion rate monitoring is necessary to assess the remaining life. In this regard, electrochemical noise (ECN), CCD cameras and eddy current probes are deployed for on-line monitoring in various corrosive–radioactive zones. The ECN method has been successfully applied to monitor corrosion damage in the underground storage tanks of nuclear waste. At the Sellafield reprocessing plant, United Kingdom, two ultrasonic devices have been developed after the conventional corrosion monitoring with electrochemical probes failed to be effective. Solid coupled probes (SCPs) and electromagnetic acoustic transducers have been used for monitoring corrosion in the dissolvers. An SCP has an accuracy better than 0.01 mm in measuring wall thickness. Corrosion monitoring probes based on measurements of electrical resistance, linear polarization resistance and ECN are available for application in the nitric acid environment. The passivating behaviour of valve metals, such as Ti, Nb, Ta, Zr and Hf, was investigated in boiling nitric acid solutions in the presence and absence of the Cr^{6+} ion as an oxidizing agent [133].

5.3.2. Laser based electro-optic tube inspection systems

For detection of corrosion, pits, wall loss, chemical/scale build-up in the dissolver vessel and accessible pipes, a laser based electro-optic tube inspection system (LOTIS) working on the optical triangulation principle is very attractive. An array based LOTIS can also be used for complete circumferential profiling without mechanical rotation of the laser sensor head. The array laser sources are sequentially pulsed, generating quantitative high resolution maps of the internal surface in radial and axial perspectives. The results of the inspection can be displayed in both tabular and graphical form for detailed analysis. A laser triangulation system has been developed in India for planar surface profiling of the titanium dissolver vessel of the FR reprocessing plants.

5.3.3. Immersion ultrasonic testing for the dissolver vessel

For assessment of wall thinning in a dissolver vessel, immersion ultrasonic testing is a possible technique. During testing, the dissolver vessel is filled with water and this water column is used as the coupling medium between the wall and transducer. Normal beam ultrasonic testing is performed with a waterproof probe operated at 20 MHz to evaluate the wall thinning. The accuracy obtained in measuring wall thickness by this method is 0.1 mm. The entire inspection is done remotely with the help of a 2-axis manipulator. The manipulator is first centred and is provided with a special drive mechanism, which can carry the probe head to different elevations and can scan circumferentially to obtain the wall thickness data [134].

5.3.4. Electromagnetic non-destructive testing of pipes and tubes

Eddy current, pulsed eddy current (PEC) and magnetic flux leakage (MFL) techniques are useful for corrosion monitoring in pipes and tubes [135]. In the PEC technique, pulsed excitation is fed to the coil, contrary to continuous sinusoids used in the conventional eddy current technique. PEC is a non-intrusive and non-contact method and is complementary to ultrasonic testing for estimating corrosion rates and trends. PEC typically achieves an in-service measurement repeatability of 0.2% (e.g. 0.02 mm on a 10 mm wall thickness) in less than one tenth of the time needed by ultrasound. Another technique to detect far-side corrosion in storage tanks is SLOFEC (saturation low frequency eddy current). The MFL technique is employed for wall thickness measurement in shells and for full surface coverage for detection of local corrosion on the invisible reverse side [136].

Local metal loss due to corrosion distorts the flux which is detected by hall effect sensors. An array of sensors covering a width of 250 mm for fast imaging/scanning, called FLOORSCANNER, is also attractive. By combining MFL and eddy current techniques into one sensor module, it is possible to detect defects on the top and bottom sides of tanks, and also discriminate them. Recently, an integrated eddy current giant-magneto-resistive sensor has been developed in India for detection of far-side inter-granular corrosion in the stainless steel walls of waste vault tanks [137]. Detection of far-side corrosion at a depth of 9 mm is feasible using this technique.

5.4. REMOTE HANDLING, ROBOTICS AND IN-SERVICE INSPECTION

Robotics, automation and remote handling technology play a crucial role in almost all facets of the nuclear fuel cycle [138–144]. The phenomenal advances in this fascinating area have been due to the various necessities that are unique to the nuclear industry, such as reducing radiation exposure during handling operations, improvements in quality with increased productivity, technologies required to facilitate remote inspection in inaccessible areas of nuclear reactor or nuclear plants, or facilitating remote repair/refurbishments at operating plants.

All operations and maintenance in the back end of the fuel cycle, such as spent fuel storage pools, reprocessing plants, plutonium based fuel manufacturing plants, temporary waste storage vaults and permanent waste repositories have to be essentially carried out remotely, owing to the prevailing harsh radiation environment. Irrespective of policies, whether it is open cycle or closed cycle adapted for the nuclear programme, remote technology still plays a major role at all stages, starting from spent fuel handling and finishing with waste disposal. The increasing amount of spent fuel from operating reactors worldwide will necessitate the addition of more spent fuel and waste management facilities for significant periods of time to come, involving continued application of remote technologies.

Another area of major application for remote technology is decommissioning of ageing nuclear power generating stations, where spent fuel inventories have to be removed first, as well as the decommissioning of the spent fuel management facilities themselves. The resurgence of nuclear energy worldwide is expected to fuel growth in remote technologies and eventually substantiate the need for implementation of the developed technologies and equipment in the nuclear energy programme.

5.4.1. Evolution in remote technologies

Remote handling technology has matured from the periscope with mirror arrangement and long-handled tools in hot cells, to such devices as through-the-wall mechanical master–slave manipulators (MSMs), electrical servo-manipulators, a dual arm anthropomorphic system with a head-aiming remote television and bilateral force reflection. The current stage of the evolution is characterized by sophisticated systems involving intelligent robotic systems with teleoperation features for a variety of nuclear applications including in-service inspection.

Worldwide use of MSMs stems from the pioneering work done at ANL. Central Research Laboratories along with ORNL developed the first force-reflecting servo-manipulator using distributed digital electronics to implement position–position reflection with multiplexed serial communications between master and slave. This model has been used over the years to perform a wide range of complex tasks for nuclear and military/space applications as well as for efficient teleoperations in highly unstructured task environments.

Advanced servo-manipulators (ASMs) [145] followed the dual arm anthropomorphic system to improve remote maintainability of the remote manipulators themselves by making them mechanically modular so that one robot system could be used to repair another. ASMs were designed to provide a foundation for telerobotics in addition to effective teleoperations. Trajectory teach-playback and automated tool-changing functions were also demonstrated. However, the United States Department of Energy (DOE) programme supporting the work was cancelled together with the nation’s breeder reactor programme before telerobotics and automation functions could be incorporated and evaluated. At this point in time, CEA focused their research on the telerobotics functionality for their concept of ‘computer-assisted teleoperations’ [146].

Remote repair and maintenance in the back end of the fuel cycle, especially in reprocessing plants, were developed in some countries due to specific conditions related to the consequences of incidents resulting in the release of radioactivity outside the cell. A well known example of remote technique applications at the head-end part of reprocessing plants is the remote inspection and maintenance operation to repair the dissolver at the Tokai-mura reprocessing plant in Japan [147]. Another example is the repair work performed at the La Hague plant in France, making use of a specially designed and built, remotely controlled system. At the Sellafield reprocessing plant, the dissolver vessel branch pipes were inspected for corrosion-induced damages and repaired using a dexterous multi-jointed manipulator called REPMAN [148].

5.4.2. National experiences

5.4.2.1. France

Considerable developments have taken place in the area of remote equipment and technologies over the years, catering to the nuclear industry, especially the spent fuel management industry, to carry out an increasing number of tasks of maintenance in active facilities and decommissioning of plants which have been shut down. Despite a lull worldwide, France has sustained an interest in providing advanced technological solutions for plants in the back end of the fuel cycle in the field of teleoperation, remote handling and robotics, through French industries such as AREVA and EDF [149–152].

R&D efforts in France have mainly been focused on the development of remote technologies, addressing requirements of existing facilities, and advanced technologies for ongoing and future remote handling applications. Much emphasis has been laid on the development of robotic equipment in addition to automation requirements for facilities in the back end of the nuclear fuel cycle, substituting human operators to reduce man-Rem consumption. The robotics developments in collaboration with French industry have resulted in application of these technologies to production plants for repair welding and contamination checking, maintenance and inspection/cleaning in emergencies following an incident. Different kinds of robotic technologies are currently used in France, primarily manipulators installed in permanent workstations with functions associated with the production process/maintenance and tele-manipulators for special tasks requiring a high degree of mobility.

For existing nuclear facilities, versatile remote handling equipment has been developed for applications such as remote maintenance in reprocessing plants. One such development is a dexterous slave arm having seven degrees of freedom with six axis force/torque sensors and an electrical force-controlled gripper intended for remote teleoperation applications in maintenance and intervention on process equipment [152]. This arm could be wall/floor-mounted or installed on a mobile platform such as a crane gantry. Such a platform makes a dual-arm intelligent teleoperation system, consisting of two dexterous arms with a radiation tolerant (10 kGy) intelligent electronics control system to carry out maintenance tasks in fuel reprocessing facilities, where standard wall-mounted mechanical tele-manipulators cannot be used. The intelligent controller provides the necessary teleoperation functionalities.

Recent developments also include long reach muti-segmented arms for reconnaissance and inspection. This is a very challenging robotic arm developed for inspection of the hot cells without viewing windows in the nuclear fuel facilities. It is equipped with on-board radiation hardened control electronics (10 kGy). This arm also has the capabilities for carrying out remote tasks such as welding, cutting and virtually any other maintenance or repair work inside cluttered hot cells.

The strategy of developing industrial robots and remote manipulators in France has resulted in reduced investment costs and high product reliability. With such industrial robots, the performance obtained in teleoperation with force feedback has been good enough to safely carry out remote high precision tasks and to handle heavier loads than those in manufacturing applications. The STAÜBLI industrial robot with a force feedback teleoperation control system has been successfully adapted to nuclear requirements and demonstrated in a real intervention at the La Hague plant.

For decommissioning, constraints of space and capacity have led to relying on hydraulic technology for the development of a remote master–slave with force feedback. The Maestro system, developed by CEA in collaboration with Cybernetix, is adapted at different configurations of facilities. The capacity and the sensitivity of the force feedback enable the operator to provide significant productivity both for maintenance and dismantling operations, thus limiting influence and risks due to the human factor. The system cured at 10 kGy has a modular architecture to facilitate its decontamination and to limit the time for maintenance.

To limit the operational dosimetry on nuclear worksites and to optimize the best scenarios for intervention according to ALARA principles, the CEA, in partnership with Euriware, has developed the NARVEOS interactive simulation software. This three dimensional modelling software of the environment allows point sources or volume associated with a gamma spectrum to be defined. The software also allows the dose rate to be calculated, in real time according to different scenarios, at any point of the hot cell, for example, and also operator dosimetry.

More recent progress in decommissioning projects has identified new requirements for low level measurements, with the objective not only of detecting the raw signal but also of quantifying the residual activity: characterization of irradiating waste, final inspection (verification of walls and floors), glove box measurements (residual activity, decontamination progress) and miscellaneous radiological characterization. These in situ measurement techniques require increasing sensitivity and resolution. The issue of background noise must also be taken into account. In this context, CEA is improving existing tools (gamma imaging), qualifying new detectors (gamma spectrometry), and developing new methods and techniques for source location and quantification (alpha imaging, high resolution spectrometry to estimate the contamination depth). Some of this work has reached the qualification stage prior to industrialization, and field tests of measurement systems are now in progress [153].

5.4.2.2. United States of America

The US DOE is vigorously addressing the problems of disposal of radioactive waste materials. Robotic and remote control systems are being considered and developed to minimize routine human exposure to radiation and hazards. The US DOE has elaborated on conceptual systems involving robotic and remote systems for addressing a wide variety of tasks for correcting problems in existing radioactive waste material storage and disposal sites [154, 155]. Automated or remote control may be used for systems handling sensors, excavating material, sorting, processing and transferring raw or processed radioactive wastes.

Robotic systems and remotely controlled manipulators are being designed or considered for a wide range of waste processing efforts, including characterization by positioning sensors within the storage tanks, over landfill areas and into ground or stored material to determine radioactivity and other hazardous factors present in areas unsafe for human entry. In addition, robotic systems are supposed to perform recovery and testing of samples from the waste site; inspection of containers; excavation and removal of material from tanks; retrieval of containers stored above ground or buried; transfer; manipulation; and opening, closing and sealing of the containers. For such purposes, a robotic system was developed and practically used for inspection of underground stainless steel tanks containing liquid waste to verify their integrity.

As an advanced remote technology, a concept for facility maintenance with a fully remotely controlled system was developed in the USA for FR fuel reprocessing due to the heavy involvement of remote operations in reprocessing. In this concept, each unit of the process equipment is designed in a module to fit in the structured position array on the hot cell floor and vertical position provided with the connectors and penetrations on the walls in the hot cell. In comparison with the conventional hot cell complex, which used to be highly compartmentalized in a number of functional cells, the fully remote hot cell concept is usually an open hall enclosure equipped with all the necessary remote monitoring and control systems for operation and maintenance.

5.4.2.3. Japan

Many kinds of remote technology have been developed in Japan to reduce radiation exposure to operators and to increase redundancy in nuclear plants. An advanced robotics technology project has been initiated with the aim of developing robots that can carry out inspection, maintenance and rescue operations or other work in the field of nuclear power plants. Basic technologies, such as a dynamic stereo vision system, an optical wireless communication system, manipulation, quadrupedal walking, on-wall locomotion, radiation resistance, a fault-tolerant robot controller and a robot health care system have been developed, and technologies have been established.

A concept known as “full remote maintenance in a large cell” [156] was conceived and developed to increase the facility operation ratio and to decrease the operator’s radiation exposure. It is needed to install a force reflecting MSM robot. In this concept, it is possible to repair failed equipment in situ using a dual arm bilateral servo-manipulator (BSM) system or to replace the equipment with a rack as a module by loading some equipment using the manipulator system and an in-cell crane. Radiation hardened electronic devices and the sensors are screened and used for the BSM system. The manipulator system includes a two arm BSM, mobile platform, remote viewing, signal and power transmission, as well as the man–machine interface subsystem, so that one BSM system could be used to repair another BSM. The manipulator system has been developed and applied to the Tokai Vitrification Facility (TVF) and the Recycle Equipment Test Facility (RETF) that adopted the full remote maintenance concept in a large cell. The concept is designed for experimental facilities such as TVF and RETF.

5.4.3. Software and electronic components

As enabling technologies, electronic multiplexers, radiation hardened embedded electronic systems using off the shelf components developed for an integral dose of 10 kGy, and force/torque sensors facilitated the successful transformation of the industrial grade robots to telerobotic systems for use in a harsh radiation environment. A radiation hardened electronics controller has been developed in France to avoid thick cables management and providing embedded intelligent control. An open and modular architecture fitting all of the needs of the telemanipulators has been developed and qualified up to 10 kGy of integrated dose. Electronic devices are required, especially for high radiation fields, such as reprocessing and high level waste treatment plants, for high radiation resistance up to 1 MGy because, in some cases, it is a design requirement to reduce replacement times.

Supplementing the efforts taken in the development of remote techniques, a generic portable software package providing shared control, and optimizing human and machine performance, has been developed. It allows increasing the speed and reliability of the intervention tasks, providing operators with teleoperation assistance functions, such as force feedback master/slave control, virtual mechanisms control and other robotics functions. Software providing fast on-line modelling of remote environments by adjusting pre-existing models or by estimating the position and orientation of the objects in three dimensional space has been developed and this software is universally adapted for all robotic systems. It provides a wide range of user friendly tools for modelling in a manual or automatic way. This software package has also been successfully applied for mobile robot navigation in unstructured environments at reprocessing plants. The realization of this tool requires software data to be devised based on the actual layout of equipment.

The efficiency and safety of remote interventions is increased by using virtual reality techniques [36, 157]. Three dimensional modelling based programming of operations, visual feedback, obstacle avoidance and operator training by means of realistic simulation are some of the benefits of this emerging technology for remote handling applications [158–160].

5.5. INSTRUMENTATION AND CONTROL SYSTEMS

5.5.1. Sensors

Purge probe sensors are still used at reprocessing plants for measuring the level and density of radioactive liquids. Radioactive liquid is transferred from one tank to another by the air lift method if the required height difference is less than six meters. Otherwise, the steam jet ejector method is used, even though dilution of process liquid may take place due to the steam.

Fibre optic spectrophotometers are usually employed for on-line measurement of valence and concentration of plutonium.

Both gamma based and neutron based criticality monitoring systems are in use for detecting unforeseen criticality in reprocessing plants. Neutron based criticality monitoring systems are more reliable if gamma background is high. This usually takes place while processing irradiated fuel of high burnup. The normal setting for a gamma based criticality alarm system is 4 mR/h or 3 mR/500 s.

Beta and gamma activity is usually measured by intelligent instrumentation systems based on Geiger–Muller counters. Multichannel analysers are useful in determining energy spectra of gamma emitted by various nuclides. The effluent lines in France are provided with an on-line measurement system for beta and gamma rays. A scintillation detector based intelligent instrument is used in France for measurement of alpha activity. On-line measurement of alpha rays will usually indicate the presence of uranium or plutonium in the process liquid. Pencil dosimeters are used to measure the dose received by a worker in a radioactive area. Thermoluminescent detector badges are popular for the measurement of cumulative dose.

Some developmental issues which are under investigation are:

- Need for monitoring plutonium due to high content in FRs, especially in head-end steps — accumulation/loss/safety/material accounting [161];
- Reduction in operator dose by reducing the number of samples to be analysed manually through introduction of on-line monitoring systems;
- Thermal and FR fuel reprocessing and similar technologies for instrumentation.

5.5.2. Reliability of instrumentation and control systems

Since computer systems are now extensively used for acquiring, processing, displaying and storing data, both hardware reliability and software reliability have to be ensured. The ‘water fall’ model is recommended for the design/development of computer systems. Detailed documentation needs to be prepared at every life cycle stage of development according to applicable IEEE standards.

All of the hardware and software are subjected to testing and verification by an independent committee. The hardware is also subjected to environmental tests, including electromagnetic compatibility according to applicable standards. The supervision software is developed according to MISRA-C guidelines. Detailed safety analysis is carried out for both hardware and software to ensure that the ‘fail safe’ criterion is honoured in these systems. For the safety instrumentation, redundancy and diversity are recommended. For example, to detect unforeseen criticality, at least three sensors and signal processing systems are usually used in parallel. The outputs are routed through appropriate voting logic for energizing an alarm in the control room.

5.5.3. Simulators and virtual reality for the reprocessing plants

In real operations, the plant operator should not try to fill a tank where the level is high, or drain a tank where the level is low. Similarly, both filling and draining a tank is not allowed at the same time. A tank cannot, at the same time, receive liquid from more than one tank. All of these conditions can be simulated for every tank in a training simulator, and the operator must be fully trained in transfer operations for radioactive liquids. The operator should also be fully trained in assessing the probability of criticality and functioning of criticality monitoring systems. The information on the La Hague plant simulator can be found in the literature.

The advanced features of a three dimensional electronic display system, animation and walkthrough software tools are extensively used in practice to view different parts of fuel recycle facilities. This helps to avoid pipeline interference during construction and ensures enough space for erection and subsequent maintenance of various equipment.

6. CURRENT STATUS OF SPENT FAST REACTOR FUEL REPROCESSING AND WASTE TREATMENT IN VARIOUS COUNTRIES

6.1. FRANCE

France constructed and operated the experimental FR RAPSODIE (40 MW(th)) from 1967 until 1983 and the prototype FR PHENIX (600 MW(th)) from 1973 until March 2009. The commercial FR Superphenix-1 (2990 MW(th)) was commissioned in 1985 and operated until 1998.

6.1.1. Aqueous reprocessing

Fast reactor fuel reprocessing in France has been carried out in pilot facilities located at Marcoule and La Hague. The first facility AT1 (Atelier de Retraitement des combustibles Rapides — Atelier Traitement 1) was at La Hague and the second — the APM facility (Atelier Pilote de Marcoule) with TOP (Traitement d’Oxydes Pilote) and TOR (Traitement d’Oxydes Rapides) treament lines — at Marcoule. All of these facilities were operated by the CEA. Some fast reactor fuel was also reprocessed at the UP2 La Hague plant between 1979 and 1984 by diluting with natural uranium fuel for graphite gas cooled reactors (UNGGs). Table 6 presents the data on quantities of FR fuel reprocessed in France. Laboratory scale reprocessing of around 20 kg of fuel from the RAPSODIE and PHENIX reactors was carried out in the Cyrano laboratory at Fontenay-aux-Roses between 1968 and 1985.

TABLE 6. FAST REACTOR FUELS REPROCESSED IN FRANCE

	Fuel	Initial (Pu/(Pu–U))	Years	Facilities	Burnup (GW·d/t HM)	Cooling time (months)	Amount (kg of (U+Pu))
RAPSODIE	1st core	25%	1969–1979	AT1	40–45	6–12	250
	Fortissimo	30%	1969–1979	AT1	50–120	5–24	660
	Fortissimo	30%	1975	APM	55–76	6–10	50
KNK I	Enriched U		1975–1976	APM	3.4–6.8	12–20	1 650
PHENIX	Enriched U		1977–1978	APM	38–45	10–30	2 300
	Pu Core I	18%	1978–1979	AT1	8–44	18	180
	PU Core I	18%	1979	APM	37	10–30	155
	Pu Core I	18%	1979–1983	UP2	23–34	38–50	7 900
	Pu Core II	25%	1979–1983	APM	36–83	14–42	6 360
	Pu Core I	18%	1983	APM	35–100	29–36	225
Total							19 730

The construction of the AT1 facility at La Hague for the reprocessing of RAPSODIE fuel was decided in 1964. The design capacity of AT1 was equivalent to one core fuel of RAPSODIE per year (134 kg of U–Pu). It began active operation in 1969 with the reprocessing of 220 fuel rods. RAPSODIE was fuelled with both plutonium and high enriched uranium (HEU). AT1 was shut down at the end of 1979 after the reprocessing of around 910 kg of irradiated fuel from RAPSODIE and 180 kg from PHENIX.

FR fuel reprocessing at Marcoule began in 1974 when the APM facility, which had previously treated fuel from UNGGs, was refurbished for the reprocessing of RAPSODIE fuel. The design capacity of the TOP line was 10 kg of fuel per day. The TOP line was shut down at the end of 1976 after reprocessing one RAPSODIE core.

In 1979, the reprocessing campaigns of FR fuels began at the UP2 plant. The fuel was sheared and dissolved in the presence of gadolinium in a separate facility, and then treated in U–Pu extraction cycles in dilution with UNGG fuel dissolver solution. A total of around 10 t of PHENIX fuel was treated at UP2 between 1979 and 1984.

In 1978, the CEA decided to refurbish the TOP facility and to extend the capacity of the main line to 6 t/a (TOR line) and to create a second line for the R&D technology studies. The new APM facility was brought into operation in 1988 and was used to qualify the technologies of the head-end process in the TOR facility and the technologies of the extraction of U–Pu and the conversion of Pu to PuO_2. The equipment of the TOR main line was designed for:

- Fuel receipt and storage;
- Mechanical pretreatment (subassembly);
- Dissolution using a batch dissolver;
- Off-gas treatment;
- Feed clarification;
- Separation and storage of insoluble residues;
- Packing of hulls;
- Measurement of fissile inventory in the process for nuclear material accounting;
- Pulsed columns for solvent extraction.

The equipment of the R&D line was designed for:

- Mechanical treatment, such as spacer wire removal, new choppers;
- Continuous helicoidal dissolver;
- Pulsed filters for feed clarification;
- Iodine traps;

— Krypton-85 cryogenic separation;
— Treatment and packing of hulls and insoluble residues.

Between January 1988 and January 1991, about 5 t of PHENIX fuel and KNK-II German FR fuel were reprocessed in the APM facility [162].

The deployment of LWRs in France in the 1980s and the start up of new La Hague reprocessing plants led to a halt in the R&D programme on the reprocessing of SFR-MOX fuel for around fifteen years. In 2006, the new French law on P&T of LLRN saught to connect this research with the research on fast neutron reactors. Within this framework, the CEA launched a new R&D programme on the SFR fuel cycle and the multi-recycling of uranium and plutonium in the reactor core.

In the framework of the French law on P&T of LLRNs, studies have been carried out since 1991 on the separation of minor actinides. The strategy is based on the development of successive liquid–liquid separation processes:

— The advanced PUREX process for the separation of U, Pu, Np, and FPs I and Tc;
— The DIAMEX process for the co-extraction of trivalent Am + Cm and lanthanides from the PUREX/COEX raffinate;
— The selective actinide extraction (SANEX) process for the partitioning of Am + Cm/Ln.

6.1.2. Non-aqueous reprocessing

The ATTILA facility was built at the nuclear centre of Fontenay-aux-Roses in 1967 for the development of the fluoride volatility process for the separation of U and Pu from irradiated fuel. Initially, the facility was intended for studies on fluorination of fuel based on HEU (uranium–zirconium and uranium–aluminium alloys) with practically no plutonium content. It was proposed that the facility only be used for separation and purification of uranium.

The fluoride volatility process of SNF for the separation of plutonium was optimized in France from 1967 to 1972 with treatment in the ATTILA facility of $UPuO_2$ fuel from RAPSODIE, irradiated at 50 GW·d/t HM and with a cooling time of 6 months. The fuel contained up to 25% plutonium dioxide.

Uranium and plutonium recovery yields were 97.45% and 95–97.5%, respectively. The uranium purification ratio was 10^8 and the plutonium purification ratio was 10^3 [163]. This project was implemented jointly with the Russian Federation in the FREGAT facility at RIAR. The development of this process in France was stopped at the beginning of the 1980s.

Currently, pyrochemical processes are being considered in France as an alternative route for the recycling of high content MA transmutation targets. Within this framework, the CEA has undertaken research programmes on MA separation in collaboration with Japan, the Russian Federation and the USA.

6.1.3. High level waste management

France has significant experience in reprocessing FR fuels and vitrification of HLLW. Since 1978, about 5000 t of ‘glass’ has been produced using an induction heat crucible. A cold crucible induction melter will be in commercial operation in the La Hague facilities in 2010. An added feature of a cold crucible induction melter is the melt stirring tool which improves the quality of the vitrified product by avoiding inhomogeneities.

ANDRA is the French national agency responsible for radioactive waste management. In July 2001, ANDRA signed a four year contract with the French Government for the definition of a ‘global approach’ in waste management [164]. The agreement envisaged the creation of a very low level waste (VLLW) repository, the construction of research laboratories for studies on HLLW geological disposal and studies on radium bearing and graphite waste disposal.

The VLLW disposal is built and commissioned for 30 years at the Soulaines site.

A deep underground research laboratory has been built at the Bure site (eastern France) for studies of LLRN migration in clay. ANDRA has also initiated studies for defining which types of disposal canisters are suitable for future deep underground repositories.

Work has also been carried out to evaluate the long term performance of vitreous waste. These experimental and theoretical modelling studies have provided the scientific basis for evaluating the durability of confinement in interim storage (physico-chemical properties of the materials and their state at the time of recovery and handling operations) and in disposal (under conditions of long term alteration by ground water) [165].

6.2. GERMANY

In Germany, studies on reprocessing of FR fuels were initiated in the 1960s at the Institute of Hot Chemistry, Karlsruhe Nuclear Research Centre within the framework of the fast breeder project funded at that time. Without attributing any specific preference, techniques such as molten salt, volatilization, chlorination and fluorination were studied, with the idea of building a joint reprocessing facility called “Scharade” akin to the EBR-II facility in Idaho, USA. However, in 1964 and 1965, the decision was taken to implement the aqueous route and, thus, planning and construction of an experimental scale aqueous reprocessing facility (MILLI) was started and it had its first high active operation in 1971 [166]. This facility had a capacity of 1 kg of fuel per day per extraction cycle, which was safe against criticality by geometry. The facility was also an alpha-tight one, with remote manipulators to take up to 3×10^5 γ-Ci per cell. It comprised three extraction cycles with mixer–settlers as contactors, a co-decontamination cycle, one partitioning cycle and the third purification cycle, wherein plutonium or uranium were processed alternately.

The MILLI facility offered sufficient chemical experience of reprocessing advanced fuel elements using the PUREX technique, processing test irradiation samples from the Dounreay Fast Reactor (DFR) and LWR fuel specimens of burnup up to 40 GW·d/t HM. In the first half of 1974, fuel with 15% PuO_2 from DFR with a burnup of 60 GW·d/t HM was processed using the PUREX process in the MILLI facility, adopting the PUREX flow sheet. Several dissolution tests and reprocessing of fuel specimens of high burnup were also carried out in 1976 and 1977. The facility had three banks of sixteen unit mixer–settlers capable of processing 500–600 g of metal per batch.

Between 1967 and 1973, the Gesellschaft für Kernforschung and the Gesellschaft für Wiederaufarbeitung von Kernbrennstoffen pursued a joint development programme on issues related to the reprocessing of high plutonium content MOX fuels of FRs, such as SNR-300. The important aims of this collaborative work focused on the breeder head-end stage for WAK (Karlsruhe Reprocessing Plant) and increasing plutonium throughput.

The development work carried out at WAK tried to achieve solutions to the following problems associated with handling high Pu content, high specific power and high burnup fuels:

- Cooling of discharged fuel elements during storage and transport;
- Removal of adherent sodium;
- Disassembling and cutting system;
- Off-gas retention;
- Complete dissolution of PuO_2 fraction;
- Complete co-extraction of plutonium with uranium;
- Plutonium/uranium partitioning avoiding excess addition of reducing agent and resulting process volume increase;
- Extractive plutonium decontamination under criticality restrictions;
- Plutonium concentration techniques including extraction/distillation;
- Removal of insolubles from the feed solution;
- Overcoming extraction failures due to solvent degradation;
- Reducing radiolysis by reducing contact time;
- Separation of the solvent degradation products.

Important outcomes of this development work were:

- A flow sheet with 0.1% loss of plutonium in raffinate with safe concentration control of plutonium;
- A diaphragmless in situ electrolytic mixer–settler for plutonium partitioning;
- Electrolytic oxidation cells for adjustment of the plutonium valence to the IV state in the feed solutions avoiding use of $NaNO_2$, NO_2 or N_2O_4;

- A metal frit feed clarifier;
- Measurement of the extent of degradation of the solvent and the rate of by-product formation as a function of burnup;
- Adopting high acid (3–4M HNO_3) extraction conditions and high U/Pu saturation (>70%) to avoid emulsification and phase separation problems;
- Extensive data on mass transfer kinetics of U/Pu to estimate the minimum contact time needed to ensure effective extraction, at the same time reducing solvent degradation;
- Identification of the products of solvent degradation under high radiation exposure and ways to remove them and restore the solvent;
- Decisions not to follow voloxidation due to the problems of plutonium segregation and difficulties in dissolution.

The studies in the WAK facility provided innumerable inputs to the reprocessing community. The plant was shut down in 1980 to replace a leaking dissolver after processing around 114 t HM from various sources. In October 1982, it again went on-stream with a new dissolver, disassembly machine, stainless steel steam lines and large lead-shielded boxes. Considerable work on modelling uranium–plutonium partitioning by solvent extraction and computer simulation to calculate multicomponent distribution data for U(IV), U(VI), Pu(IV), Pu(III), HNO_3 and hydrazine was also carried out at WAK. Ultimately, WAK was shut down in 1990 due to political decisions.

A waste vitrification plant, PAMELA, was built at Mol, Belgium, as a joint project between Germany and Belgium in 1985 [167]. This was to demonstrate the liquid-fed ceramic melter, processing the Eurochemic HLW. Similar vitrification plants were to be built at reprocessing plants in Germany. Due to changes in governmental policies towards nuclear energy, these plans were not actually realized and Germany currently relies on the capabilities of France and the United Kingdom for treating waste from German reactors.

In September 2000, the Government permitted the resumption of the transport of SNF to La Hague, France. Such transports out of the country were banned in 1998. Subsequently, transports to Sellafield, United Kingdom, were permitted. Six casks of vitrified HLW were transported from France to Germany’s Gorleben interim waste storage facility in March 2001. In August 2001, the federal radiation protection agency gave approval for an intermediate on-site storage facility for spent fuel from the Philippsburg nuclear power plant. The spent fuel will be contained in specially built transport casks. Spent fuel from unit two will be stored in twelve canisters for a five year period. Similarly, building of on-site or near-site spent fuel storage sites at all nuclear power units has been planned, pending the construction of a national repository by 2030.

6.3. INDIA

India initiated an FR programme around the late 1960s. The design and construction of the first experimental reactor FBTR (40 MW(th), 12 MW(e)) at Kalpakkam, Tamil Nadu, was initiated in the early 1970s with French collaboration. The reactor uses a unique, first of its kind, mixed carbide fuel with the composition $(U_{0.3}, Pu_{0.7})C$, with some sesqui-carbide content. The design and construction of a co-located pilot reprocessing facility was also initiated in parallel.

6.3.1. Aqueous reprocessing

The development of FR fuel reprocessing was planned to be established in four phases: the first one with the aim of developing the process, equipment and systems, and performing non-radioactive engineering scale experiments; the second phase aimed at setting up a pilot plant to process irradiated fuel; the third phase involved construction and operation of a demonstration facility; and the final phase involved construction and operation of a reprocessing plant for an FR with a capacity of 500 MW(e). The demonstration fast reactor plant (DFRP) is at an advanced stage of construction for the regular reprocessing of FBTR fuel, with provisions to also reprocess a few subassemblies of a prototype FBR (PFBR–500 MW(e)).

6.3.2. CORAL facility

After extensive development work on the process and equipment in the first phase, the second phase of the FR fuel reprocessing is, at present, being carried out at the CORAL (COmpact Reprocessing of Advanced fuels in Lead cells) facility. In this facility, the PUREX process was adopted, employing 30% TBP in a heavy normal paraffin mixture with three cycles of solvent extraction. CORAL is a lead-shielded facility, with a containment box in which chopper, electrolytic dissolver, feed clarification centrifuge and centrifugal contactors are located. The containment box is equipped with an in-cell crane, MSMs and several hot cell gadgets for remote operation, maintenance and replacement.

The CORAL facility was commissioned in December 2003 with the receipt of spent fuel pins from FBTR. Since then, several campaigns have been carried out with fuel pins, starting with very low burnup to as high as 155 GW·d/t and also with different cooling periods. Recently, the facility completed a campaign with a batch of fuel pins having a burnup of 155 GW·d/t and less than a two year cooling period.

A brief description of the individual operations at the CORAL facility is provided below.

6.3.2.1. Fuel handling

Spent subassemblies from the FBTR are received in the hot cells of the adjoining radio-metallurgy laboratory and sodium is removed by washing with ethyl alcohol. Mechanical de-wrapping is done and the individual fuel pins are loaded into canisters and transported in alpha-tight containers to the CORAL facility.

6.3.2.2. Fuel pin chopping

Single pin chopping is performed in a single pin chopper with a modular design to enable remote maintenance.

6.3.2.3. Dissolution

Initially, there were doubts about the complete solubility of the pins using the usual nitric acid refluxing. At the same time, dissolver solution derived from (U, Pu)C using organic acids had the potential to interfere with the normal PUREX extraction/stripping phase and could also cause phase separation problems. Extensive studies on dissolution and solvent extraction with an actual carbide dissolver solution indicated that refluxing in nitric acid would enable complete dissolution and problem free extraction and stripping.

6.3.2.4. Feed clarification

The dissolver solution was clarified using an air turbine operated centrifuge. Centrifuge maintenance was performed using remotely operable gadgets. The bowl did not show any measurable quantity of undissolved solids.

6.3.2.5. Solvent extraction

CORAL is used extensively to validate various design concepts of centrifugal extractors apart from evaluating the process flow sheet. For example, two types of centrifugal extractors, one with a fixed and the other with a remotely adjustable weir were employed. Modifications of the extractors were carried out remotely to improve performance during the campaigns.

6.3.2.6. Partitioning and reconversion

The co-decontaminated uranium–plutonium nitrate solution is subjected to an oxalate precipitation step, where Pu oxalate is a precipitate leaving the U in the supernatant. The oxalate precipitate is filtered and calcined to get PuO_2. The supernatant is precipitated with ammonium hydroxide to get ADU which is calcined to get uranium oxide powder.

CORAL has provided extensive experience in the design of key equipment for FR fuel reprocessing as well as information required for fine tuning the process flow sheet. These are vital for the DFRP and PFRP reprocessing plants.

6.3.3. Demonstration fast reactor plant

A demonstration FR fuel reprocessing plant is currently under construction. This has more than ten shielded cells. This facility will incorporate the laser dismantling facility for the wrapper removal. Chopper designs are modified to provide the increased throughput. The thermosyphon dissolver design incorporates features to reduce the hull losses. It is planned that feed conditioning is carried out using electrolytic methods. This feature will also improve the dissolution of Pu-rich MOX fuel from the FBTR. Modified versions of centrifugal extractors will be used for all of the three cycles. This plant will have the capacity to reprocess around 35 SA of FBTR. The plant will also demonstrate the reprocessing of PFBR subassemblies. Cold commissioning trials are scheduled in late 2010.

6.3.4. FRP

The reprocessing plant for the PFBR is currently at the design stage. This will have a capacity to reprocess 15 t/a which can be expanded to 50 t/a in the future.

6.3.5. R&D on aqueous reprocessing

Modelling and computer simulation are cost effective means of reducing the number of active experiments needed to finalize the process flow sheets. Significant expertise has been developed at the Indira Gandhi Centre for Atomic Research (IGCAR) in basic modelling as well as in the development of computer codes related to nuclear fuel reprocessing flow sheet analysis and development. The computer codes developed are SIMPSEX, PuTHEX and SIMPUREXE, which deal with high plutonium extraction, U–Pu–Th separation, and the PUREX operation with alternate extractants such as DOHA (di-octyl hexanamide) and TIAP, respectively. A code, SIMPACTR (SIMulation Program for ACTinide Recovery), has been developed to simulate actinide recovery from HLW, covering all solvent extraction processes. The data needed for modelling are equilibrium distribution ratios of the metal ions concerned, with densities of the organic and aqueous phases.

The currently available expertise also includes modelling of Tc behaviour in the PUREX process (including its co-extraction with U, Pu and Zr), computer simulation of Tc rejection flow sheets, improved modelling of Np distribution data and disproportional behaviour of Np(V) under PUREX conditions, distribution coefficient models for selective FPs, modelling of bulk level Pu polymerization, as well as modelling of third phase formation in TBP and other solvent extraction systems of interest.

Considerable work has been carried out in IGCAR, Kalpakkam, on the development of alternate TAPs. Several higher homologues of TBP, such as TIAP, tri-n-amyl-phosphate, tri-2-methyl-1-butyl-phosphate and tri-n-hexyl-phosphate, have been synthesized in-house, characterized and their suitability for the reprocessing applications assessed. These phosphates are devoid of the problems associated with TBP, such as high aqueous phase solubility and third phase formation, while extracting tetravalent actinides.

Long chain dialkyl monamides have been developed and suggested as extractants for thermal reactor reprocessing due to their advantageous properties, such as:

- Complete incinerability;
- Easy partitioning with mere acidity variation;
- Formation of non-deleterious degradation products, etc.

At the Bhabha Atomic Research Centre (BARC), Mumbai, work on several dialkyl monamides has been carried out for applications in PUREX and also for U–Th separation.

R&D in India on MA partitioning started in the 1990s [168]. At BARC, basic data were generated for the extraction of actinides, and a few fission and corrosion products using TRUEX solvent (0.2 M CMPO + 1.2 M TBP in dodecane). Subsequent studies examined the extraction and separation of actinides from synthetic and actual high level aqueous raffinate waste (HLW), sulphate bearing wastes at low acidity of about 0.3M, non-sulphate wastes originating from a pressurized heavy water reactor (PHWR) and FBR both in 3 M HNO_3, and actual HLW solutions generated from reprocessing of the research reactor fuels at the Centre. Mixer–settler experiments employing a six stage unit with synthetic sulphate-bearing and PHWR–HLW were also carried out. After pretreatment with 30% TBP to reduce the concentrations of U, Np and Pu, the raffinate containing the remaining U, Np, Pu, the trivalent actinides and lanthanides (at a total acidity of 3 M) was the feed for a subsequent mixer–settler experiment using 0.2 M CMPO + 1.2 M TBP in dodecane.

In IGCAR, several precursory mixer–settler runs were carried out in connection with identifying a suitable flow sheet for the recovery of MAs from the HLW solution generated from FBTR fuel reprocessing in the CORAL facility. The runs employed both TRUEX solvent and 0.2 M TODGA + 1 M DHOA (di-hexyl octanamide) in a normal paraffin hydrocarbon mixture, employing simulated PHWR waste solution as the feed.

6.3.6. Non-aqueous reprocessing

Intensive R&D has been carried out in India on the development of pyrochemical processes for oxide as well as metallic fuels. Studies on the pyroprocess for oxide fuels have established the feasibility of electro-refining for UO_2. The annular pellets of UO_2 were used as the anode and the purified UO_2 was electro-transported and deposited on a graphite cathode through a molten salt electrolyte containing the oxychloride of uranium. It has also been established that the eutectic salt of $MgCl_2$–NaCl–KCl could be a better alternate electrolyte to the CsCl–NaCl mixture used in the RIAR process of oxide electrowinning. The advantages offered by the alternate electrolyte are lower operating temperatures due to a lower melting temperature and lower cost.

It is planned that fuel cycle technology for metallic fuels will be introduced into the Indian FR system during the next decade. Thus, R&D on the molten salt electro-refining process has been in progress, using a laboratory scale facility. Besides gaining experience in the various steps of the molten electro-refining process, determination of the separation factors for Zr, Ce and Pd have been a research focus. Studies of plutonium based alloys have also been in progress. A demonstration facility is being set up to generate experience in carrying out the process with kilogram amounts. The facility will also be used to demonstrate the remote operations ability of the process.

6.3.7. Waste management

Partitioning of HLW is presently being addressed in India using the hydro-metallurgical routes involving multistep extraction employing different solvent systems.

The partitioning technology involves the separation and recovery of:

— MAs and rare earths;
— Cs and Sr;
— PGMs.

R&D studies have resulted in the synthesis, production and application of extractants such as CMPO (octyl-(phenyl)N-N-di-isobutyl carbomyl methyl phosphine oxide) and diglycol amide based TEHDGA (tetra (2-ethyl hexyl diglycolamide)) solvent systems. Bulk synthesis of both of these solvents at the required purity has been carried out, and suitable processes for their use in an actual application have been developed. Table 7 outlines the programme under development in India.

The Indian experience with FR HLW is not extensive, although considerable expertise exists in managing HLW from PUREX cycles related to thermal reactors [169]. The main features of the adopted approach are described below.

6.3.7.1. Raffinate evaporation

The first cycle raffinate is generally not evaporated in view of its high activity levels. The second and third cycle raffinates are concentrated and mixed with that from the first cycle. The acidity of the evaporator concentrate is limited to 4 N due to corrosion considerations for the stainless steel storage tanks. Since the acidity of the raffinate is around 4 N, acid killing becomes imperative prior to concentration in order to achieve volume reduction. Prior to evaporation, it is ensured that the raffinates are free of entrained and dissolved organics.

TABLE 7. PARTITIONING OF HLLW — INDIAN SCENARIO

Partitioning technology
A) Minor actinides and rare earths
A.1) Separation of trivalent actinides (An(III) and Ln(III)) using CMPO and diamides
A.2) Selective separation of actinides, An(III) from lanthanides, Ln(III)
B) Recovery of Cs
B.1) Sorptive route: Ammonium molybdophosphate (AMP)
B.2) Extraction route: Crown ethers
C) Recovery of Sr
C.1) Sorptive route: Poly-antimonate, silica-titanate
C.2) Extraction route: Crown ethers
D) Recovery of platinum group metals
D.1) Supported liquid membranes
D.2) Electrochemical separation

6.3.7.2. High level waste vitrification

Like most other countries engaged in the field of HLW vitrification, India was attracted by the apparent simplicity of the pot glass process and initiated developmental work on similar lines during the early 1970s. The objective was to carry out a thorough evaluation of the process for subsequent treatment of actual radioactive waste on a routine basis. This culminated in setting up and operating the first waste immobilization plant (WIP) at Tarapur. The second vitrification facility was set up at BARC to handle waste from various research reactors. A third WIP is under construction at Kalpakkam for the management of HLW generated from the reprocessing of fuel from PHWRs and FRs.

A solid storage and surveillance facility has been set up at Tarapur for interim storage of vitrified HLW. An elaborate programme on deep geological disposal is also being pursued in order to realize the third stage of the waste disposal programme.

6.3.7.3. Melter technologies for vitrification

The transition from induction melters to JHCMs is under way in India. The JHCM adopted at the Tarapur site has external dimensions of 1.5 m × 1.5 m × 1.8 m. The main refractory is a corrosion resistant alumina–zirconia–silica ceramic backed up with layers of insulating materials encased in a stainless steel box on suitable structural supports. The melter cavity has a hold up volume of 125 L, which is sufficient to provide a mean residence time of around 25 h at the normal glass production rate of 1.5 kg/h. This ceramic melter has successfully vitrified several cubic meters of HLW.

A CCIM system is also under development for radioactive waste vitrification. This melter will also address the vitrification of thorium bearing wastes from AHWR fuel, which needs higher melting temperatures. The equipment has been demonstrated on an industrial scale with simulated HLW.

6.4. JAPAN

The need for the development of FR fuel cycle technologies as a basis for a future energy system of the next generation was recognized in Japan from the outset of nuclear power programmes in the country. To investigate a wide range of technical options for FRs and related fuel cycle technologies, a feasibility study on commercialized FR cycle systems (F/S) was launched. Phase I of the feasibility study was carried out from 1999 to 2000. Based on the study, fuel reprocessing methods, such as advanced aqueous, oxide electro winning and metal electro-refining, and fuel fabrication methods, such as simplified pelletizing, sphere-packing, vibropacking, coated particle and injection casting, were selected for further development.

In phase II (2001–2005), a design study of FR cycle concepts, the development of significant technologies necessary for the feasibility evaluation, and confirmation of the key technical issues were performed to arrive at a decision on the promising candidate concepts for commercialization. The interim report was published in 2004 to indicate the perspective of the characteristics and performance of the candidates. These studies have indicated that the combined system of an SFR with MOX fuel, advanced aqueous reprocessing called NEXT, and a simplified pelletizing fuel fabrication are considered to be the most promising for commercialization. Metallic fuel is considered to have the potential merit of improving the core performance of SFRs. The results of phase II (completed by March 2006) have been used to develop a perspective of the promising concepts, the R&D plan until around 2015, and the key issues for commercialization [103].

Regarding the fuel cycle technology for metallic fuels of FRs, a combined system of metal electro-refining for reprocessing and injection casting for fabrication is considered to be the candidate. The fuel cycle facilities will be designed as integrated reprocessing and fuel fabrication plants, whose systems are simple and compact, so as to be suitable for remote operation and maintenance, considering that pure Pu never appears at any stage.

JAEA launched the “Fast Reactor (FR) Fuel Cycle Technology Development (FaCT)” project in cooperation with the Japanese electric utilities in 2006. The FaCT project is based on the conclusion of the F/S project.

Currently, R&D of innovative technologies and a design study of a commercial facility are being implemented to decide on the adoption of innovative technologies by 2010, and to judge the applicability of innovative technologies and conduct the conceptual design of demonstration and/or commercial facilities, which would be possible to achieve the development target, by 2015.

The NEXT process is composed of several processes and technologies, and the following six issues have been identified as the innovative technologies to be developed corresponding to each process step: (1) disassembling and shearing, (2) highly effective dissolution, (3) effective U pre-recovery by crystallization, (4) U–Pu–Np co-recovery, (5) MA recovery by extraction chromatography and (6) a salt-free process for waste reduction.

6.4.1. Aqueous processing (before launching the F/S)

JAEA commissioned the experimental FR JOYO (140 MW(th)) in 1977, and a demonstration FR MONJU (714 MW(th)) in 1994. A sodium leak accident occurred in MONJU in 1995 and many modifications were completed by May 2007. The functions of modified and/or long term isolated systems of the entire plant are being checked for the restart.

JAEA has been pursuing FR fuel reprocessing technology development since 1975. In 1981, JAEA completed the construction of a new major R&D facility at the Tokai site, called the Chemical Processing Facility (CPF). CPF has two hot cell lines, the first designed for studying the application of the PUREX process to irradiated FR fuels and the second to study HLW vitrification. CPF started operation in 1982, with laboratory scale hot experiments on reprocessing MOX fuel from JOYO with a burnup of 4.4 GW·d/t HM. Operational experience in CPF indicated satisfactory fuel dissolution and diminished crud formation with clarified feed solutions. Extensive studies were carried out with irradiated JOYO fuel to explore the solubility behaviour and to assess the insoluble residue content, as well as the formation of Pu(VI) in the dissolver solution.

The PUREX solvent extraction flow sheet was adopted using 30% TBP/n-dodecane with a multistage mixer–settler. The computer code MIXSET was developed to simulate the dynamic behaviour of the PUREX process with mixer–settlers for the Tokai facility. The calculated concentration profiles were in good agreement with the experimental results [170].

Approximately 10 kg HM of FR spent fuels with a burnup of approximately 40–50 GW·d/t HM in early times, and approximately 100 GW·d/t HM at peak times, have been treated in CPF.

Before the F/S was launched, JAEA developed an integrated approach for FRs with a closed fuel cycle based on an aqueous process including the:

- Dissolution of the MOX FR spent fuels with an aqueous nitric acid solution;
- Iodine volatilization;
- Electrolytic extraction of Tc, Pd and Se;

- Crystallization of most of the uranium contained in the spent fuel dissolution liquor in the form of uranyl nitrate hexahydrate crystal;
- A single PUREX extraction step for recovery of the remaining U, Pu and Np;
- SETFICS process for Am and Cm partitioning.

JAEA projected to construct the RETF for the purpose of conducting the engineering scale hot test of the advanced process and equipment for FR spent fuel reprocessing based on the PUREX process. The construction of RETF was started in 1995. After the incidents of MONJU in 1995 and the Tokai bituminization plant in 1997, it was decided to suspend construction of RETF and to remain flexible in order to appropriately reflect the progress being made in the development of FR reprocessing technologies. The first stage of construction of RETF was completed in 2000 and the future utilization plan is now under investigation considering the progress of the FaCT project.

For many years, JAEA has been developing the so called four group partitioning process for treatment of the waste stream resulting from the reprocessing flow of the standard PUREX process for UOX or MOX LWR spent fuels. This partitioning process includes the following steps associated with the respective four groups:

- MA partitioning (Np, Am and Cm);
- Cs and Sr extraction;
- PGM extraction.

The remaining mixture of the wastes constitutes the fourth category of elements of the initial mixture treated. JAERI has also proposed to separate Np and Tc during the implementation of the PUREX process.

6.4.2. Integrated approach based on the FaCT project

JAEA is currently developing an integrated approach to back end FR fuel cycle technologies (the FaCT project), allowing optimizing FR advantages through combining the SNF reprocessing stage with manufacturing new fuel elements/assemblies from the reprocessed fuel. The main concept in the FaCT project, which should be developed principally for the most promising fuel cycle system for commercialization, is the combination of a NEXT process with a simplified pelletizing fuel fabrication, and an SFR with MOX fuel.

The facilities are designed to be a unification of simplified and compacted reprocessing and fuel fabrication steps, well suited for remote operation and maintenance. The basic requirement assumes that pure Pu will never appear in any processes. For this purpose, an essential and challengeable technical issue is to establish a remote fabrication system to handle the fuel materials containing MA, degraded Pu and FPs with high levels of radiation and heat generation in a hot cell.

The NEXT system (Fig. 10) consists of a simplified solvent extraction process for U, Pu and Np with the addition of a U crystallization step and an MA (Am, Cm) recovery process by extraction chromatography. In this process, a highly concentrated dissolver solution is chilled and around 70 wt% of U is separated from the solution as crystals of uranyl nitrate hexahydrate to obtain a residual solution with the concentration ratio of Pu to U necessary for a new MOX fuel. The size of the equipment and the amount of solvent required for the subsequent extraction process are, thus, significantly decreased. The single-cycle solvent extraction, without further partitioning and purification steps, recovers a U, Pu and Np mixed solution with a moderate DF $\geq 10^4$. A centrifugal contactor, which has advantages in terms of compactness, smaller holdup volume and high separation performance, is used for the extraction process.

For recovering both Am and Cm from the raffinate, an extraction chromatography method with a more compact size and a better DF for lanthanide FPs will be adopted, based on a comparitive study of the solvent extraction method SETFICS and others. An adsorbent, impregnated with several candidates of solvent, such as CMPO, TODGA, HDEHP, i-Hex-BTP, is being experimentally investigated for actinide (III) recovery from a highly acidic solution and actinide (III)/lanthanide (III) separation. The use of non-flammable microporous silica (SiO_2) provides an adsorbent with faster kinetics, less organic material, and also with good separation during large scale operation.

A bird’s eye view of an integrated recycle plant with a 200 t HM/a throughput and a cross-sectional view of the NEXT part are shown in Fig. 11.

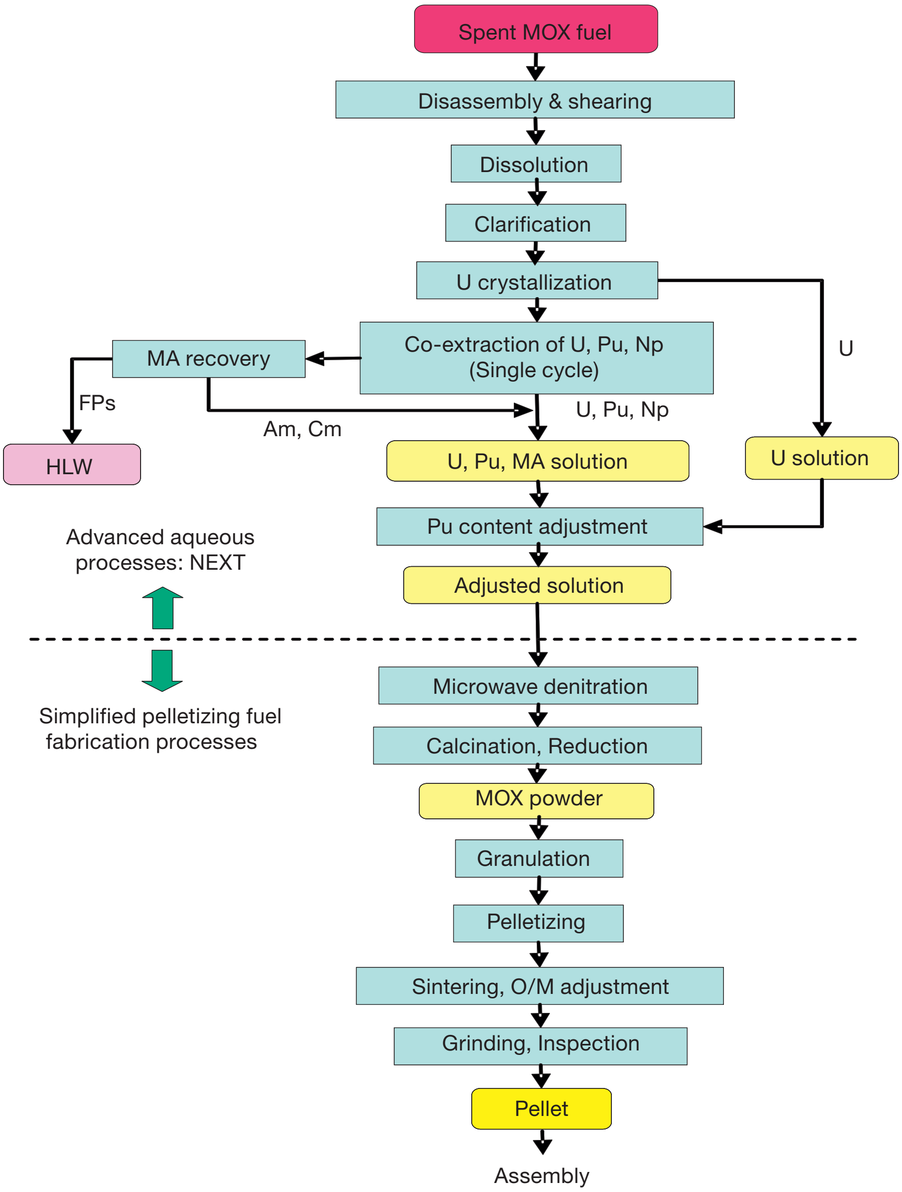
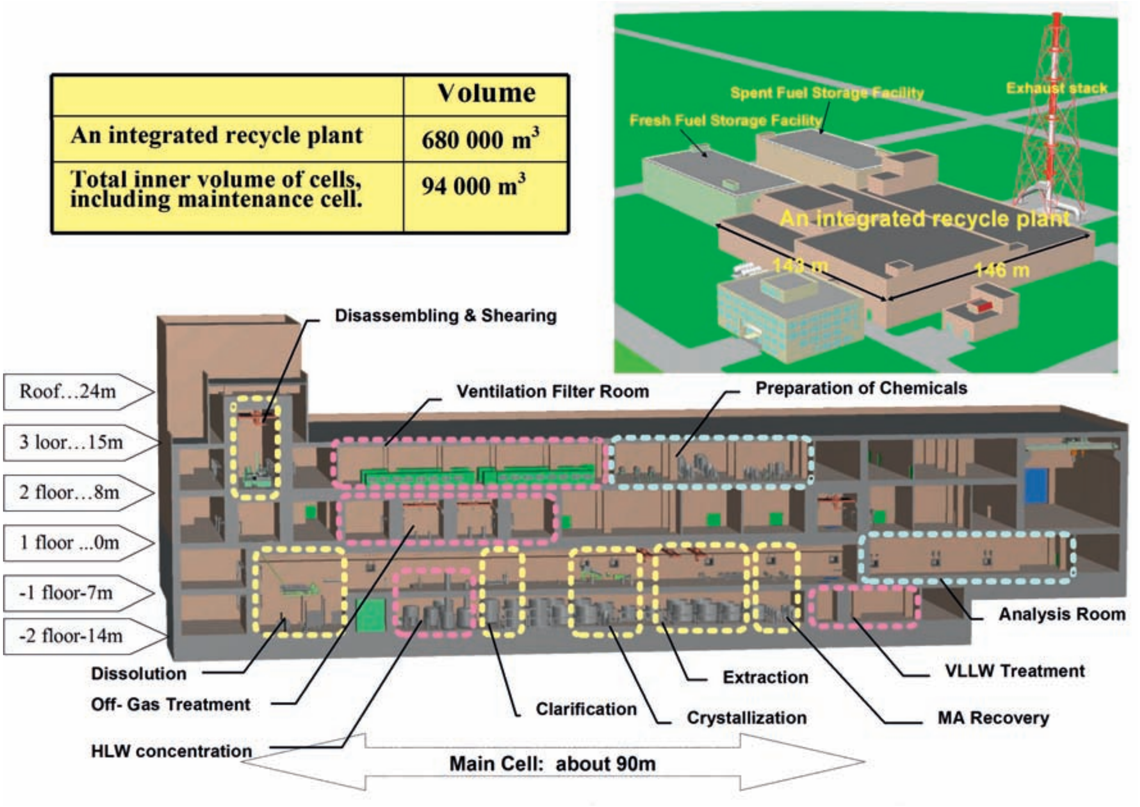


FIG. 10. Process flow of advanced aqueous reprocessing (NEXT) and simplified pelletizing.

The NEXT reprocessing system allows for a reduction in costs for both construction and operation in comparison with a conventional PUREX system. Regarding the environmental impact, the volume of vitrified waste (HLW) normalized by the specific power generation will be reduced to approximately 60% compared to that from LWR fuel reprocessing, because the heat generation from actinides could be suppressed and this allows maximization of FP content to their solubility limit in the glass phase. The NEXT system also reduces the amount of low level waste (LLW) for the following reasons:

- Simplifying the main reprocessing processes;
- Use of salt-free reagents, such as hydrazine oxalate and hydrazine carbonate which are easily decomposed into gas, thus decreasing secondary solid waste;
- Categorizing liquid waste into very low and high radioactive wastes by sequential concentration and evaporation, which simplifies the effluent treatment system and the solidification process.

Regarding enhancement of nuclear proliferation resistance, co-extraction of U, Pu and Np, and mixing them with Am and Cm decreases the product attractiveness for potential proliferators, and the low DFs admissible in the remotely operated facilities impede illegal access to the products due to high radiation and heat generation levels.



	Volume
An integrated recycle plant	680 000 m^3
Total inner volume of cells, including maintenance cell.	94 000 m^3

FIG. 11. Bird's eye view of an integrated recycle plant with a 200 t HM/a throughput and a cross-sectional view of the NEXT reprocessing part.

Japan is also developing a process called Super-DIREX based on supercritical fluid extraction.

One more process considered at JAEA implements an approach combining such features as using amides instead of the traditional PUREX TBP extractant, and flexibility in adjusting the process to different transmutation/waste management scenarios. The concept was named ARTIST (see Section 4.3.6).

6.4.3. Non-aqueous processing

Over the years, Japan has carried out research on pyrochemical processes, and made the following improvements in the electro-refining process for metallic fuels originally developed at ANL:

- Introduction of a process step for distillation of bond sodium in the fuel pin prior to electro-refining to avoid the introduction of the excess Na into the electro-refiner. The distilled Na could be oxidized and utilized as an ingredient in glass materials of the salt waste form.
- Recycling of a large amount of salt after it is passed through the zeolite column. This reduces the salt waste volume.
- Design of a rectangular electro-refiner with multiple electrode assemblies for simultaneous recovery of U on the solid cathodes and a U–Pu–MA mixture in the liquid Cd cathodes, and a continuous cathode processor to enhance economy, operability and throughput.

Japan is developing the closed fuel cycle for both metallic and nitride fuelled fast burners with corresponding pyroprocesses. With respect to electro-refining reprocessing for metallic fuels, feasibility is basically assured for the main process technologies such as electro-refining, via laboratory and/or engineering tests performed at ANL, CRIEPI, JAEA and ITU.

CRIEPI and JAEA are developing a closed fuel cycle for metal-fuelled FRs assuming its integration with the oxide fuel reprocessing from LWR. LWR spent oxide fuel will be subjected to electrochemical reduction to metals, and further pyrochemically processed to prepare the feed for the U–Pu–MA–Zr fuel of the FRs. With this objective, CRIEPI has drawn up a programme up to 2015 for developing a molten salt electro-refining process. In this programme with JAEA, the irradiation of U–Pu–Zr alloy fuels prepared by injection casting is performed in the JOYO reactor to a burnup of 200 GW/t HM, with linear heat rates (LHRs) of 500 W/cm. U–Pu–MA–Zr alloy fuels, prepared by arc melting, are irradiated in a Phoenix reactor, under the collaborative METAPHIX programme with ITU to a burnup of 100 GW/t HM with an LHR of 350 W/cm. The irradiated samples then undergo pyroprocessing. Developing an engineering scale electro-refiner model is also part of the programme.

JAEA is pursuing the development of the closed fuel cycle based on molten salt electro-refining of nitride fuels proposed for ADSs, liquid metal and gas cooled FRs. Studies have shown that the electro-refining process is suitable for recovering ^{15}N, as well as for preparation of nitride powders by nitriding the U–Pu–MA–RE deposited on a liquid cadmium cathode.

Cost analysis showed that the process can meet target costs, with smaller capacity plants being more economical. However, there are still significant technical challenges for the commercialization of the system. Development of the system has been undertaken under international cooperation with the USA and other countries because the facilities for development are not available in Japan.

Further development of these fuel cycle technologies will be continued in Japan and the various aspects surrounding the commercialization of FR cycle systems will be more clearly understood by around 2015. The commercialization promotion stage will be the next step, providing the basis for economic estimates of commercial size system performance. A systematic R&D plan has been established for this.

R&D of the combined system of metal electro-refining and injection casting, which is compatible with metallic fuel, will be continued. For this concept, specific items which are less developed than those of the aqueous system or those with relatively low adaptability to design targets, such as main equipment development and optimization of the solid waste volume, will be preferentially performed. A design study for the commercialized fuel cycle facility will also be carried out to compare system performance with that of aqueous systems.

Japan is also developing a new technology called FLUOREX (see Section 4.9).

6.4.4. Waste management

At present, commercial reprocessing of SNF for Japan is done in Europe, and the vitrified HLW is returned to Japan. Currently, shipments from France and the United Kingdom arrive at Rokkasho-mura in sequential batches. More than 1300 canisters had been successfully delivered to the site as of March 2009 [171].

In March 2001, Japan Nuclear Fuels Ltd proposed the construction of an additional facility to store vitrified HLW at the Rokkasho-mura site to take over from the existing waste storage centre, which was expected to be filled by the end of 2005. In October 2000, the Nuclear Waste Management Organization was formed to implement geological disposal of the waste. Construction of a repository in granite or sedimentary rock is planned for the 2030s.

Regarding HLW management from FRs, JAEA has carried out a feasibility study of several FR cycle systems, including HLW management aspects associated with four reprocessing routes, namely the advanced aqueous process proposed by JAEA, the oxide electrowinning method developed by RIAR, the metal electrowinning method developed by ANL, and the fluoride volatility method.

A tenfold larger waste volume in comparison with dry methods was calculated for the SETFICS process as it produces large amounts of sodium containing HLLW. This indicates the necessity for improvements in the SETFICS process. Moreover, it was shown that the amount of TRU element waste would, under all ‘dry’ scenarios, not be more than one-fifth as compared to aqueous processes.

In the metal electro-refining method, the salt is cleaned by using a zeolite column system. The zeolite occluded by FP chlorides is heated with glass forming material and converted to sodalite, the final disposal form.

Thus, in this study, the salt waste is mainly sought to be fixed as borosilicate glass, with the exception of the metallic waste form for the noble metals from the oxide electro winning route. For the fluoride volatility and the metal electro-refining methods, the calculated waste volumes are almost the same as those of the conventional PUREX process. In the case of oxide electro winning, larger vitrified waste volumes are expected due to the limited solubility of Cs in glass; this is also true for metallic fuel processing. However, TRU element waste volumes will be reduced to less than a quarter of those from the advanced aqueous route, since no liquid waste is generated.

For the fluoride volatility and the metal electro-refining methods, the calculated waste volumes are almost the same as those of the conventional PUREX process.

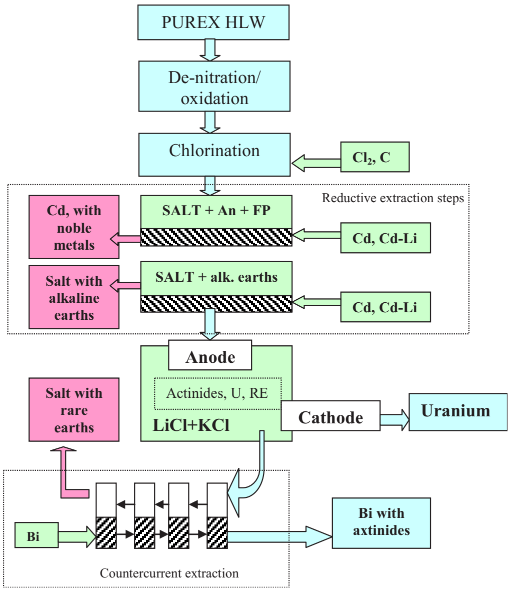


FIG. 12. Flow diagram of CRIEPI's dry separation process, Japan.

The CRIEPI's dry separation process consists of six stages: de-nitration, chlorination, dissolution, reducing extraction, electrolytic refining and waste treatment (Fig. 12).

The above flow sheet is based on the reductive extraction and electrolytic refining processes that have been developed on a laboratory scale in a collaborative project between the USA and Japan. Laboratory studies using a solid Cd cathode have shown that the set goals (separation of 99% of each actinide) are achieved in the TRUMP-S (TRansUranic Management through Pyropartitioning Separation) process although the product purity will require recycling of cathode deposits containing a few per cent of americium [172]. A conceptual design of a facility based on this technology, with a capacity of 0.25 t HM/d, is being developed in Japan for treatment of FR oxide fuel [173].

6.5. REPUBLIC OF KOREA

KAERI has been developing an advanced spent fuel conditioning process (ACP) since 1997, based on a pyrochemical process, which has a number of potential advantages, such as compactness, simplicity and low cost. The ACP reduces the volume, radiotoxicity and heat load of the spent oxide fuels discharged from the current fleet of commercial PWRs and eventually creates a facile form of a metallic ingot for disposal.

An electrolytic reduction process has been developed as a key unit of the ACP, which consists of several steps, including voloxidation, electrolytic reduction and smelting. In the ACP concept, the air oxidation, called voloxidation, enables the pulverizing of spent fuel, enhancing the electrolytic reduction rate due to the surface area increase of spent fuel. The spent oxide fuel, which is voloxidized in advance, is converted to metallic form by an electrochemical method in $LiCl–Li_2O$ molten salt. The produced metal is smelted to be prepared in a metal ingot form. Gaseous FPs, such as I_2, Kr and Xe, are released during the voloxidation step. In addition, salt-soluble FPs, such as Cs, Sr, etc., are dissolved and removed from the SNF during an electrolytic reduction step. A facility for demonstration of the ACP process at 20 kg HM/batch scale has been built.

KAERI is also carrying out another R&D project on P&T of LLRNs, focusing on the development of a pyroprocess based on electro-refining of actinides. In the electro-refining process, most of the actinides as well as FPs are electrochemically dissolved from the anode into molten LiCl–KCl salt, and then uranium and TRU elements are selectively recovered by using two types of cathodes. As part of the pyrochemical partitioning studies for P&T purposes, various electrochemical experiments using a solid cathode and a liquid cadmium cathode have been carried out to investigate the electrochemical behaviour of uranium and rare earth elements in the electro-refining system. On the basis of this fundamental study, electro-refiners of 100 g U/batch and 1 kg U/batch were developed successfully in 2003 and 2006, respectively. Currently, KAERI's unique continuous high throughput

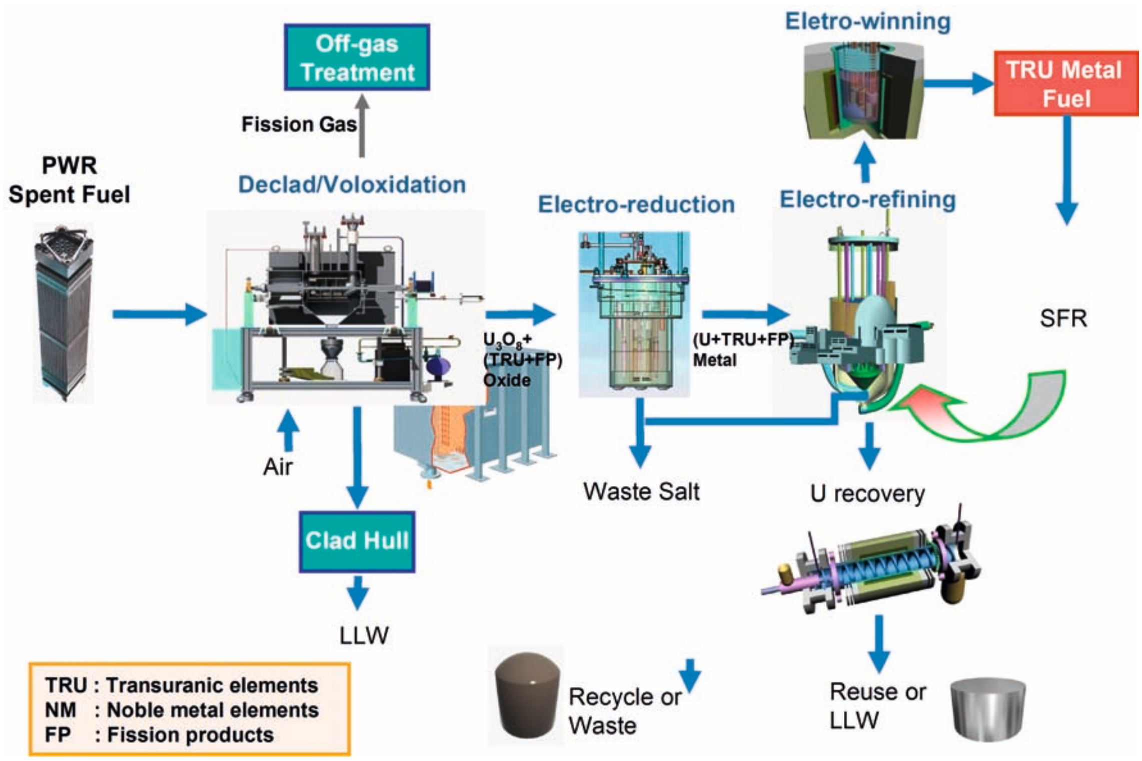


FIG. 13. Pyroprocessing flow being developed in the Republic of Korea.

electro-refiner is under development to embody a capacity of 20 kg U/batch. The continuous concept in this refiner was realized by means of a graphite cathode system.

For the recycling of a used salt during the pyroprocess, batch type ion exchange technology has been developed to remove the radioactive nuclides, such as Cs and Sr, in a waste salt. Especially, the final waste volume may be drastically decreased by selective removal of FP nuclides in the waste salt through an oxidation/precipitation reaction. KAERI has established the basic concepts of waste salt minimization by recycling of the refined salt.

In December 2008, the Korea Atomic Energy Commission approved a long term R&D plan for the development of a sodium FR and pyroprocessing technology in order to utilize the benefits of nuclear energy and manage the burden of spent fuel in a proliferation resistant manner. The main efforts for developing pyroprocessing are directed at the electrolytic reduction of oxide fuel and electro-refining, targeting the reduction of the volume, heat load and toxicity of the spent fuel, and application to sodium FR systems as the recycling and transmutation methods for closing the fuel cycle as shown in Fig. 13.

Future technology development of a pyroprocess includes four steps: development of the key technologies in the first step (2007–2009), demonstration and verification of an engineering scale facility (2010–2016), installation of a demonstration facility (2017–2025) and operation of the pyroprocessing facility after 2026.

6.6. RUSSIAN FEDERATION

This country has built and operated several experimental and power FRs, of which BR-10 (8 MW(th)) was an experimental facility and became critical in the year 1958. The industrial pilot reactors BOR-60 (55 MW(th)) and BN-600 (1470 MW(th)) were commissioned in 1968 and 1980, respectively. The industrial reactor BN-350 in Kazakhstan (750 MW(th)) was commissioned in 1972 and was closed in 1999. There are plans to commission the commercial reactor BN-800 (under construction) in 2012.

6.6.1. Aqueous reprocessing

The closed fuel cycle was pursued by Russia, with the aim of reprocessing all WWER reactor fuels from Russia and its client states, and recycling plutonium in the FRs. The RT-1 facility, based on a PUREX cycle, was constructed at the MAYAK Reprocessing Plant at Chelyabinsk-65 (now Ozersk). The facility reprocessed spent fuels from smaller WWER-210 and WWER-440 power reactors and was also used for reprocessing spent fuel from the FRs BN-600 (at Yekaterinburg) and BN-350 (at Mangyshlak Peninsula) [174]. The RT-1 plant was designed for a reprocessing capacity of up to 400 t/a.

In 1975, it was decided to construct a second reprocessing facility RT-2, at Krasnoyarsk, with a capacity of 1000 t/a to reprocess fuel from WWER-1000s and other reactors. However, construction was impeded by policy decisions and shortage of funds, and by 1985 only 30% of construction had been completed (present status is indefinite).

Considerable research and development work has been pursued in the Russian Federation towards alternative extractants in place of TBP in the PUREX cycle, MA separation, and recovery of ^{137}Cs, ^{90}Sr and PGEs from HLW.

6.6.2. Non-aqueous reprocessing

Russia developed both the pyro-electrochemical process and the fluoride volatility process. RIAR formulated and experimentally validated the basic steps of an advanced closed fuel cycle, based on mutually compatible reprocessing of uranium–plutonium oxide fuel and fabrication of fuel elements and fuel assemblies. These steps are:

- Implementation of dry pyro-electrochemical processes in molten salt (the so called Dimitrovgrad dry process (DDP)) for reprocessing spent fuel and waste treatment;
- Production of poly-dispersed crystalline oxide fuel particles (both MOX or individual oxides of U and Pu) having nearly the theoretical density necessary for immediate vibrocompaction in the fuel pin cladding;
- Implementation of the vibrocompaction method to fabricate fuel elements using granulated fuels;
- Use of automated and remotely controlled equipment for fuel reprocessing and manufacturing of fuel elements and fuel assemblies.

The reprocessing technology for the spent oxide fuel is ready for commercialization. The pyro-electrochemical DDP is being developed for application in the reprocessing of spent MOX fuel and nitride fuel from the BREST (Russian lead cooled fast reactor) reactor. The most important results were obtained in experiments for treatment of irradiated MOX fuel of the BOR-60 reactor in kilogram quantities with a burnup of 210 to 240 GW·d/t HM. The process temperature is in the range of 900 to 920 K [175].

In collaboration with the Bochvar Institute, Russian Federation, RIAR carried out studies on various pyrochemical process options for reprocessing nitride fuels. The most studied method is electrochemical treatment of nitride fuel in relatively low temperature molten chloride salts: anodic dissolution of uranium–plutonium nitride fuel pellets and deposition of uranium and plutonium metals in an electrolytic cell in the temperature range of 773–823 K. The electrolyte is a eutectic mixture of LiCl and KC1 containing UCl_3 and $PuCl_3$. Excellent recovery of uranium and plutonium has been reported: only 0.4 mass% of uranium and 0.05 mass% of plutonium remain in the molten chlorides [176]. These studies have shown that co-deposition of neptunium and americium with uranium and plutonium is feasible.

The Russian Federation has proposed carrying out the following studies related to the development of non-aqueous technologies per se as well as for subsequent additional applications:

- Studies on the behaviour of noble impurities (Ru, Rh, Pd), Tc and other FPs in the pyrochemical processes;
- Investigation of methods for fractionation of FPs by directed crystallization, extraction in the salt–metal system and liquid cathode deposition [177];
- Production of oxide, nitride and metallic type fuel with MAs (for transmutation purposes);
- Reprocessing of BREST reactor nitride fuel;
- Reprocessing of FR metal fuel;
- Waste partitioning to extract TRU elements for transmutation;
- Reprocessing of uranium fuel from the research reactors (to solve the problem of handling unconventional SNF).

An international collaboration between the Russian Federation, Japan, France, the Republic of Korea and the USA has been initiated for the development of dry processes.

Significant efforts have been undertaken in Russia in developing fluorination methods of re-processing fuel from the FR. A combination of studies for treatment of irradiated uranium and uranium–plutonium MOX fuel of FRs (BOR-60 irradiated fuel with a burnup of about 100 GW·d/t HM) was undertaken at the FREGAT, FREGAT-2 and KS-2 facilities. The flowchart of the FREGAT facility is similar to that of the ATTILA facility (France) and includes separation and purification of one target component (uranium).

A major advantage of the fluoride volatility process is that UF_6 can be used directly for re-enrichment. In addition, hexafluorides can be transformed into dioxides just after reprocessing by pyro-hydrolysis in a gaseous mixture of $H_2O + H_2$. This process was developed at RIAR. After irradiated fuel reprocessing, UF_6 was converted to granulated UO_2, which was used for manufacturing vibropacked fuel pins. The manufactured fuel elements were subsequently irradiated in the BOR-60 reactor up to a burnup of 156 GW/t HM.

Cost studies carried out in the project and at design institutions of Rosatom have shown economical advantages of the fluoride volatility method of spent fuel reprocessing as compared to aqueous extraction technology, in particular for fuels with high plutonium content. Thus, the Russian Federation has proposed using pyroprocess based MOX fuel production for manufacturing MOX fuel for BN-600 and BN-800. An integrated fuel cycle facility for BN-800 has also been planned.

In the Russian Federation, the fluoride volatility process has been developed up to the pilot commercial level and forms part of a combined flow sheet for a new RT-2 plant. Studies have proven the feasibility of advanced uranium purification (to a level of 10^7) [32, 178].

6.6.3. High level waste management philosophy and practices in the Russian Federation

Recently, Poluektov (Bochvar Institute) presented a fairly detailed account of the waste management philosophy and practices in the Russian Federation. The nuclear activities, so far, have resulted in the accumulation of a large amount of high and medium level radioactive waste.

Currently, the PUREX process leads to the generation of large amounts of liquid radioactive waste: 4.5 m^3 of liquid HLW, 150 m^3 of intermediate level waste (ILW) and up to 2000 m^3 of LLW from the reprocessing of 1 t of SNF. However, new technologies of SNF reprocessing are under development, with the aim of substantially reducing radioactive waste. In any case, almost all FPs and MAs are intended to be removed from fuel upon reprocessing. The initial conditions for the development of waste management standards were the general principles of safety, which were discussed and refined in international discussions organized by the IAEA.

The solid and solidified LLW and short lived ILW are directed to near-surface disposal. LLLW and short lived ILW are intended for deep underground disposal. The solid and solidified HLW and ILW with high specific power generation containing no LLRNs are intended for long term storage with forced cooling and subsequent disposal, and the solid and solidified HLW and ILW containing LLRNs are intended for deep geological disposal. The principles, criteria and general safety requirements of radioactive waste disposal in the Russian Federation are contained in the regulatory document NP-055-04 “Radioactive waste disposal: Principles, criteria and general safety requirements — Federal norms and rules (2004)” both for near-surface and deep geological disposal.

6.6.3.1. Treatment of liquid waste

The first industrial facility for vitrification at the MAYAK reprocessing plant was commissioned in 1987. To date, more than 20 000 m^3 of liquid HLW, with a total activity of more than 450 million Ci, have been treated there, and more than 4000 t of glass have been produced and filled into about 8800 cans, and directed to interim storage. Operation of the melter EP-500/3 is now in progress. Schematic views of the vitrification facility and subsequent storage facility are presented in Figs 14 and 15.

CCIM technology has been under development in Russia since the end of the 1970s. Different schemes for liquid processing and immobilization were tested using CCIM. The calcination of liquid waste to generate dry oxides is the first stage of treatment. A CCIM pilot facility is planned at PO MAYAK for HLW immobilization. The Russian Federation and France have a cooperative programme for the development of conditioning technologies within the framework of a bilateral agreement between Rosatom and the CEA.

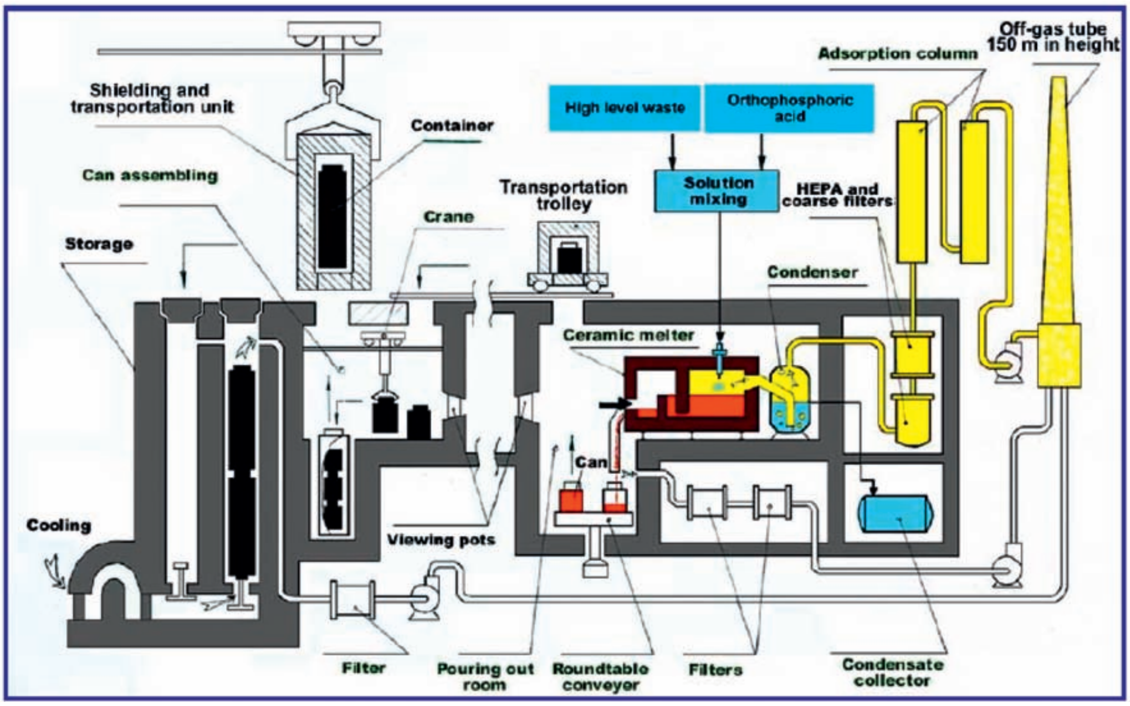


FIG. 14. Schematics of the vitrification complex facility with an EP-500 melter at MAYAK.

FIG. 15. Storage facility for vitrified waste at MAYAK.

6.6.3.2. Treatment of molten salt wastes by phosphate precipitation

The Russian Federation has spearheaded the idea of precipitating FP elements from the salt waste as a mixture of simple or complex phosphates by treating the salt melt with alkali phosphate. The separated crystalline phosphates consist of durable sodium–zirconium phosphate, monazite, apatite, etc. They constitute a ceramic waste form or will be converted to a phosphate glass waste form.

6.7. UNITED KINGDOM

6.7.1. Aqueous processing

Work on FR fuel reprocessing started in the United Kingdom along with work on the demonstration fast reactor [179] in the late 1950s. Initially, the required basic R&D work was carried out at Harwell as well as at Dounreay laboratories. A dedicated plant to process the fuel of the DFR was set up in 1961 and it reprocessed 10 t of DFR fuel.

The DFR plant was housed in a building 130 ft long, 85 ft wide and 40 ft tall, and was divided into two long sections. The larger section comprised the caves, cells, their service and sampling systems, and the access area, also termed the active area. The smaller section enclosed instrumentation and control systems, and the inactive feed system area. The plant was originally designed to process enriched ^{235}U (75%) alloy fuel, which was chopped into slugs after mechanical stripping of the niobium cladding to the dissolver. The dissolution was performed in nitric acid medium and the process adopted was the PUREX process with 20% TBP/odourless kerosene. The contactors were mixer–settlers with a 4 in diameter to avoid criticality. Three cycles of solvent extraction gave FP DFs of 10^7 to 10^8, with the first cycle alone providing around 10^4 to 10^5. A fourth purification cycle for plutonium passed pure plutonium to a glove box and was not regularly used due to low production in the HEU fuel.

In 1972, the United Kingdom made a decision to reprocess the prototype fast reactor (PFR) fuel at the Dounreay reprocessing plant. Analysis of the existing facilities compared to the requirements of PFR reprocessing — a 10^3 increase in plutonium throughput and an order of magnitude increase in FP activity compared to DFR — demonstrated the need for extensive modifications of the existing FR reprocessing plant and the construction of waste management facilities for the storage/treatment of plutonium contaminated waste. The process was based on removal of sodium from the irradiated subassembly, disassembly to individual fuel pin, single pin chopping, nitric acid dissolution and a PUREX type flow sheet in a chemical separation plant equipped with mixer–settlers. Discharge of the low active liquid effluent into the sea was authorized for the Dounreay plant. An additional liquid effluent treatment plant was constructed to ensure that discharges were not significantly different from the PFR reprocessing operations. After the shutdown of the PFR in 1994 following withdrawal of Government support for the FR programme, the Dounreay reprocessing plant was shut down in 1996 after developing a leak in the dissolver and is currently being decommissioned. The majority of the fuel from the PFR was reprocessed at this facility and plutonium products were sent to Sellafield for storage. Options for reprocessing the rest of the PFR fuel are still under evaluation.

Until the recent PFR reprocessing campaign, only laboratory scale dissolution trials on both unirradiated and irradiated $(Pu/U)O_2$ had been carried out by both co-precipitation and physically mixed fabrication routes. The laboratory work indicated that freshly manufactured, co-precipitated fuel was not completely soluble in nitric acid under reflux conditions, with insoluble residues in the range of 0.9–5.6%. For physically mixed fuel, residues were in the range of 0.33–1.58%. Examination of residues from laboratory scale (100 g) dissolution studies of samples from DFR irradiated co-precipitated $(Pu/U)O_2$ fuel pins indicated lower residues of 0.5–1.0% as compared to those obtained from comparable unirradiated dissolution studies. The irradiated fuel dissolved smoothly, essentially going to completion in about 8 h. Air driven centrifuges (5 cm diameter, 25 cm long) designed and developed specifically with an active remote maintenance possibility with a maximum capacity of 500 g were used for feed clarification.

6.7.2. Non-aqueous processing

The United Kingdom pyrochemical programme is led by British Nuclear Fuels Ltd (BNFL). BNFL carried out a number of laboratory scale studies on molten salts. In October 2001, a new, larger BNFL molten salt project was initiated with a seven year time frame. Electro-refining experiments were carried out at AEA Technology (AEA-T), Harwell, as part of a collaborative research project (PYROREP) under the European Commission 5th Framework Programme. Pyrochemical test work was performed at bench scale in the new BNFL Technology Centre at Sellafield. BNFL also participated in the European Commission 6th Framework Programme (2002–2006), which is the European Union programme for research and technological development and a major tool to support the creation of the European Research Area. Pyrochemical separation studies are under way in various universities as well.

BNFL has designed, constructed and tested a 1 L capacity electro-refiner using chloride salts as electrolyte and associated reductive extraction equipment. These have been used to measure various kinetic and thermodynamic data as well as to explore and develop further partitioning process features. The maximum inventory of Pu was 50 g. Although the pyrochemical separations data obtained were insufficient for arriving at a flow sheet for the partitioning processes, they did generally confirm the feasibility of several key separation steps.

BNFL has subsequently addressed issues associated with the oxide reduction step by chemical and electrochemical means. Non-radioactive electrochemical reduction experiments were carried out. In addition to participation in the PYROREP programme, BNFL has undertaken additional studies on electro-refining. Practical electrochemical measurements in nitride systems were undertaken by assignment to JAERI. A contract placed at a US university examined the pyrochemical separation of uranium from the fuel cladding materials to determine the DFs and how the electrochemical dissolution of the cladding material affects the behaviour of the melt system. A number of studies are ongoing to examine separation of the FPs from the salt. Extensive studies have now been completed by BNFL on the treatment of various simulant salt wastes with phosphate precipitants. It is proposed to explore zeolite and other ion exchange materials in the future. BNFL has examined incorporation of the inactive waste simulants, and has attempted to optimize the waste loading [110]. A number of waste forms including glasses, glass–ceramics and ceramics are considered.

UKAEA, Harwell carried out several studies for the development of pyrochemical processes in the 1960s, which include studies on electrowinning of crystalline uranium oxide from molten salts at pilot plant scale [111]. AEA-T, the privatised commercial division of UKAEA, has carried out contract research services for international research institutions, such as CRIEPI and KAERI [116–118]. Studies on the reduction of uranium, plutonium, americium and neptunium oxides, and unirradiated MOX pellets using lithium metal were performed for CRIEPI. KAERI and AEA-T have collaboratively conducted electro-refining studies at Harwell on the electrolysis of actinide and lanthanide fluorides in an LiF–NaF–KF melt.

Pyrochemical processing is seen in the United Kingdom to be at a pre-competitive stage and it is felt that pyroprocessing plants are more likely to be installed in the longer term. A first radioactive demonstration facility is anticipated in the period 2015–2020. BNFL and JAEA have a technical collaboration in the industrialization of pyrochemical separations. It is proposed to perform a transparent design procedure including peer review and recommendation of a small number of favoured candidates later to be reduced to one. Design objectives have been based on the use of high fissile content fuel, taking into account severe criticality safety constraints and with an initial facility throughput of 50 t HM/a and a possible later deployment at 200 t HM/a. It has been proposed to design, construct and test a one-fifth scale pilot plant, using depleted U and alkali chlorides.

6.7.3. Waste management

Several aspects of handling radioactive solid waste arising from PFR reprocessing were reported in 1981 [27]. An integrated system of waste management facilities was commissioned at Dounreay for this purpose. The solid wastes were categorized as high active, low active, high active plutonium contaminated and low active plutonium contaminated. A number of NDA techniques were developed for the waste categorization. Processing of HLLW was not indicated.

BNFL obtained a licence from the Nuclear Installations Inspectorate to commence active commissioning of the third line at its Waste Vitrification Plant at Sellafield in December 2001. Active liquor was introduced in January 2002. This will enable BNFL to meet its plan to convert liquid HLW to borosilicate glass blocks for long term storage [164].

6.8. UNITED STATES OF AMERICA

6.8.1. Aqueous processing

Due to the previous strategic US decision on treating SNF as waste and not pursuing the reprocessing option, development work for the FR fuel cycle was only performed in a few laboratories, although interest is now increasing again. ORNL together with ANL have been influential in promoting the wider use of centrifugal contactors (favoured due to the high fissile content and decay power of FR fuel materials), associated remote handling systems and hardware prototypes for most unit operations in the reprocessing conceptual designs in the context of their development of the Consolidated Fuel Reprocessing Program.

There is limited experience with reprocessing tests on the Fast Flux Text Facility (FFTF) MOX fuel. ORNL has undertaken small tests on laboratory scale dissolution and solvent extraction of MOX fuel irradiated to 220 GW/t HM burnup at around 2 kg batch scale [180–186].

The initiative called the breeder reprocessing engineering test (BRET) was started in the 1980s with a focus on the developmental activity of the US DOE to demonstrate breeder fuel reprocessing technology while closing the fuel cycle for the FFTF. The process was supposed to be installed at the existing Fuels and Materials Examination Facility (FMEF) at the Hanford Site, Richland, Washington. The major objectives of BRET were to:

- Develop and demonstrate reprocessing technology and systems for breeder fuel;
- Close the fuel cycle for the FFTF;
- Provide an integrated test of breeder reactor fuel cycle technology — reprocessing, safeguards and waste management.

BRET was a joint effort between the Westinghouse Hanford Company and ORNL. Improved processes and components for BRET were identified and developed as well as the design, procurement and development of prototypic equipment. The integrated testing of the process equipment and flow sheets was conducted with the goal of prototyping a full scale pilot reprocessing plant, and also for testing prototypes of specific complex components, operating in a remote mode. Head-end process and laboratory scale development efforts were reported. The development of off-gas treatment processes has generic application to fuel reprocessing; progress of this work is also reported [187–189].

The partitioning processes developed in the USA were mainly aimed at treatment of defence wastes, in particular those accumulated at the US DOE’s Hanford site during the cold war. Initial development studies on TRUEX used a dissolved sludge waste that simulated the insoluble HLW sludge from Hanford storage tanks. All subsequent studies used real waste solutions of TRU element analytical waste generated at ANL and the New Brunswick Laboratory. The TRUEX process was evaluated at the Lockheed Martin Idaho Technologies Company for the separation of the actinides from acidic HLW. Several TRUEX demonstration runs have been performed on sodium-bearing waste.

Several processes were developed for the partitioning of radionuclides from the wastes:

- TRUEX process for TRU element extraction;
- SREX process for Sr removal;
- CSEX process for Cs extraction.

The areas that would influence next generation aqueous fuel reprocessing for conceptual design of the Advanced Fuel Cycle Facility (AFCF) and the Consolidated Fuel Treatment Centre (CFTC) have been recently reviewed by Hebditch et al. [188].

The main factors driving the conceptual design of AFCF/CFTC are summarized in Fig. 16.

6.8.2. Combined aqueous/dry process under the “UREX+” concept

The USA has also been developing the flow sheets for pyrochemical processing of fuels for accelerator based transmutation of waste (ATW) [6]. It was originally envisioned that the system resulting from this development programme would deal with large quantities of commercial LWR irradiated fuel currently stored in the USA (over

FIG. 16. Schematic of drivers for AFCF/CFTC conceptual design.

40 000 t HM now, and over 100 000 t HM by 2020). In the initial formulation of the US ATW programme, it was assumed that all TRU elements contained in LWR irradiated fuel would be directed to an accelerator driven subcritical reactor for fission. An aqueous separation method was chosen for initial partitioning of LWR irradiated fuel which incorporates a solvent extraction process for extraction of pure uranium and technetium, but leaves the TRU elements and other FPs in the first cycle raffinate. That stream is calcined to produce TRU elements and FP oxides, which are fed to a pyrochemical process (known as PYRO-A) for separation of the TRU elements from FPs. In this process, the oxides are electrochemically reduced to metallic form for TRU element/FP separation. Several methods for oxide reduction were considered, including chemical reduction by lithium or calcium and direct electrochemical reduction. The chosen electrochemical reduction process is carried out in a LiCl carrier salt containing a small concentration of Li_2O, which then becomes the electrolyte for the next step, electro-refining, in which FPs are separated from the TRU elements.

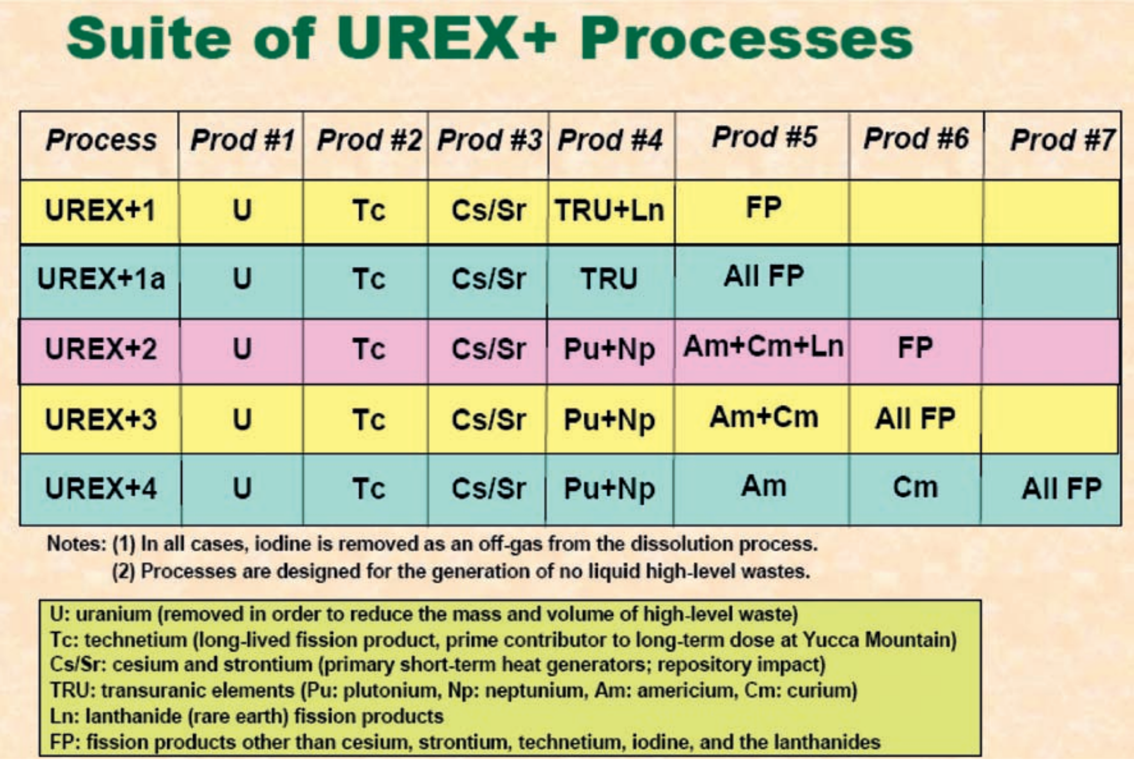
A reductive extraction process might also serve to accomplish separation of FPs from TRU elements. A process incorporating this method is under development. The oxide product is chlorinated in a LiCl–KCl carrier salt and the salt is then contacted with a molten dilute alloy of lithium in cadmium. The TRU elements are extracted into the cadmium and the FPs remain in the salt. The TRU elements are then recovered by distilling off the cadmium.

Primarily oriented on reprocessing HLW from LWRs, the reprocessing scheme should have been providing separation of U for final disposition as LLW, and TRU elements for burning in dedicated systems.

The processes considered for these separations are:

- The uranium extraction (UREX) process, which consists of a modified PUREX process aiming to only extract U;
- A pyro-metallurgical partitioning process for TRU element separation from the UREX wastes and for the ATW fuel cycle.

This UREX based separation process has become one of the key attributes in the development plan of the US DOE AFCI which is the fundamental technical edifice for the development of the GNEP. The main characteristic feature of the UREX process is the separation, at the first stage, of pure uranium which constitutes the bulk of the mass of the spent fuel and the removal, at an early stage, of the main heat-generating radionuclides, such as Cs and Sr. The resulting ‘waste’ stream, thus, contains all actinides including Pu and the bulk of the FPs, with the exception



Suite of UREX+ Processes

Process	Prod #1	Prod #2	Prod #3	Prod #4	Prod #5	Prod #6	Prod #7
UREX+1	U	Tc	Cs/Sr	TRU+Ln	FP		
UREX+1a	U	Tc	Cs/Sr	TRU	All FP		
UREX+2	U	Tc	Cs/Sr	Pu+Np	Am+Cm+Ln	FP	
UREX+3	U	Tc	Cs/Sr	Pu+Np	Am+Cm	All FP	
UREX+4	U	Tc	Cs/Sr	Pu+Np	Am	Cm	All FP

Notes: (1) In all cases, iodine is removed as an off-gas from the dissolution process.
(2) Processes are designed for the generation of no liquid high-level wastes.

U: uranium (removed in order to reduce the mass and volume of high-level waste)
Tc: technetium (long-lived fission product, prime contributor to long-term dose at Yucca Mountain)
Cs/Sr: cesium and strontium (primary short-term heat generators; repository impact)
TRU: transuranic elements (Pu: plutonium, Np: neptunium, Am: americium, Cm: curium)
Ln: lanthanide (rare earth) fission products
FP: fission products other than cesium, strontium, technetium, iodine, and the lanthanides

FIG. 17. Details of UREX+ processes.

of Tc, Cs and Sr. Further processing of this stream becomes optional depending on the specifics of the chosen transmutation scenario. The full set of such options is illustrated in Fig. 17.

The raffinate of the extraction containing the FPs and the TRU elements (which also includes plutonium) is calcined and transferred to the pyrochemical section of the plant. Each of the UREX+ processes, shown above in Fig. 17, has been designed for different applications. The UREX+1 and UREX+1a processes are group extractions of the TRU elements, in which there is no further separation of the TRU elements. These processes are intended for use in a single-tier system, with the TRU elements being directed to fast spectrum reactors as a group for burning.

The UREX+3 and UREX+4 processes can be used for FR recycling of plutonium and neptunium. Under this option, the recycling fuel fabrication will not require a hot cell environment and glove box standards will be sufficient. Additionally, such fuel is easier qualified due to the large relevant database on fuel performance from prior testing programmes carried out in EBR-II and FFTF.

The UREX+4 process provides the added option of burning americium in specially designed target assemblies. The residual FPs remaining after these separation steps are comparatively benign (low radiation level and heat generation rate) and can be immobilized at high concentrations in durable ceramic waste forms.

Since the initial dissolution in the UREX process is performed at lower acidity, it may result in greater amounts of undissolved solids or require a longer reaction time. Although spent fuel dissolution is a decades old technology, conditions that minimize the formation of insoluble solids or increase the solids recycle rate must be fully defined.

These different UREX processes (UREX+1, +1a, +2, +3, +4) are being developed, keeping in view different transmutation scenario possibilities. For example, UREX+1 and UREX+1a are designed for homogeneous recycling of all TRU elements to fast spectrum reactors, while UREX+2, +3 and +4 are designed for heterogeneous recycling, possibly as an evolutionary step, to preclude the need for remote fabrication of fuel.

6.8.3. Other non-aqueous technologies

The quest for pyrochemical alternatives to aqueous reprocessing has been under way in the USA since the late 1950s [35]. Approaches examined at various levels of development and for a variety of fuels include alloy melting, FP volatilization and adsorption, fluoride and chloride volatility methods, redox solvent extractions between liquid salt and metal phases, precipitation and fractional crystallization, and electrowinning and electro-refining of actinide metals and oxides. The processes that won through to greatest acceptance internationally are considered to be metal electro-refining, oxide electrowinning and fluoride volatility. The first one of these has been the most successful in the USA.

Many studies were carried out by the Brookhaven National Laboratory (BNL), NY, ANL and ORNL for the chemical processing of irradiated fuels through the use of the fluoride volatility process. ORNL has treated kilogram quantities of irradiated fuels including: Zr–HEU submarine type, HEU–Al research reactor type and salt from the ORNL molten salt reactor experiment [37]. Development of the fluoride volatility processes decreased in the following the cancellation of the MSRE project in 1973.

The phenomenon of electrolytic production of solid UO_2 from molten UO_2Cl_2 was discovered in the 1950s, [38]. An oxide electrowinning process was developed at Hanford Laboratories as the salt cycle process but terminated there in the mid-1960s after processing irradiated thermal oxide fuels at a 0.1 m^3 salt batch pilot scale [25]. Oxidative–reductive extractions between the molten metal and the salt phases have been the basis of a number of fuel cycle processes for experimental reactor concepts with molten fuel types. BNL studied the pyrochemical separation of uranium and FPs using redox extractions between chloride salts and molten bismuth for the liquid metal fast reactor with ^{235}U dissolved in Bi as fuel [190]. Los Alamos National Laboratory tested pyrochemical extraction methods to separate FPs from molten plutonium fuel in the LAMPRE system [191]. ORNL investigated liquid bismuth as media using lithium as the reductant for separation of U and Pa from FPs, and from salt in the MSRE [192].

Reprocessing and re-fabrication of 35 000 fuel elements (~2300 kg of short cooled fuels) for EBR-II took place at ANL-West in the mid to late 1960s using a melt refining process (Fig. 18) based on volatilization and selective reaction with the crucible material [193]. The typical ‘turn around time’ (the time starting from getting discharged fuel from the reactor, including reprocessing and re-fabrication, and ending with return to the reactor) was around 50 d. However, the process was considered to be incomplete as it leaves behind about 7% of the heavy metal. These residuals were set aside for a subsequent recovery step — one that was never made part of the process. Moreover, the metal fuel burnup was limited to about 10–20 GW·d/t HM in the early years of EBR-II operation.

Further development of this process was discontinued when EBR-II became a test bed for oxide fuels. Much of the ANL nuclear pyrochemical development work after 1960 was based on the salt-transport process using zinc and other low melting metals as solvents for extractions between metals and salts; this has been reviewed by Steunenberg et al. [194]. However, the disadvantages of this method include difficulties in phase separations and solvent removal, and a limited variety of construction materials compatible with zinc. This work preceded electro-refining of irradiated nuclear fuels as identified by Burris et al. [195], who presented the early proposals for the IFR fuel recycling system in the mid to late 1980s. Halides slagging and electro-refining were first proposed, and then it was found that anodic dissolution obviated the need for slagging and/or chemical dissolution in liquid cadmium. These aspects of IFR irradiated fuel processing are discussed by Hannum [24].

In the IFR process, electro-refining of irradiated metallic fuel is performed in LiCl–KCl salt. The IFR project was terminated in 1994 but the technology provided the basis for a fuel conditioning treatment to manage irradiated fuels at the ANL-West site.

A modified pyro-treatment version — electrometallurgical treatment (EMT) — has been selected by the US DOE to treat 25 t of SNF from the EBR-II FR at a facility at INL. A three year programme to demonstrate EMT technology using the EBR-II SNF was completed at ANL in 1999. The core fuel was a U–Zr alloy, the burnup was 100 GW·d/t HM and the fuel blanket was sodium-contaminated metallic uranium. One hundred core fuel subassemblies (containing 400 kg of HEU) and 13 blanket fuel assemblies (containing 600 kg of depleted uranium) have been treated. EMT technology comprises four steps for the treatment of sodium-contaminated SNF:

- Electrolyte purification with neutralization of sodium and removal of uranium from SNF;
- Melting with casting of uranium ingots for storage;
- Preparation for removal of TRU element waste and soluble FPs as well as treatment of waste with generation of ceramic forms;
- Treatment of metal waste with preparation for removal of fuel cladding and insoluble FPs.

The LiCl–KCl eutectic salt stream accumulates the FPs (alkali metals, alkaline earths and rare earths) from the reprocessed fuel, and sodium from the sodium-bonding of the fuel pin. The efficacy of the molten salt,

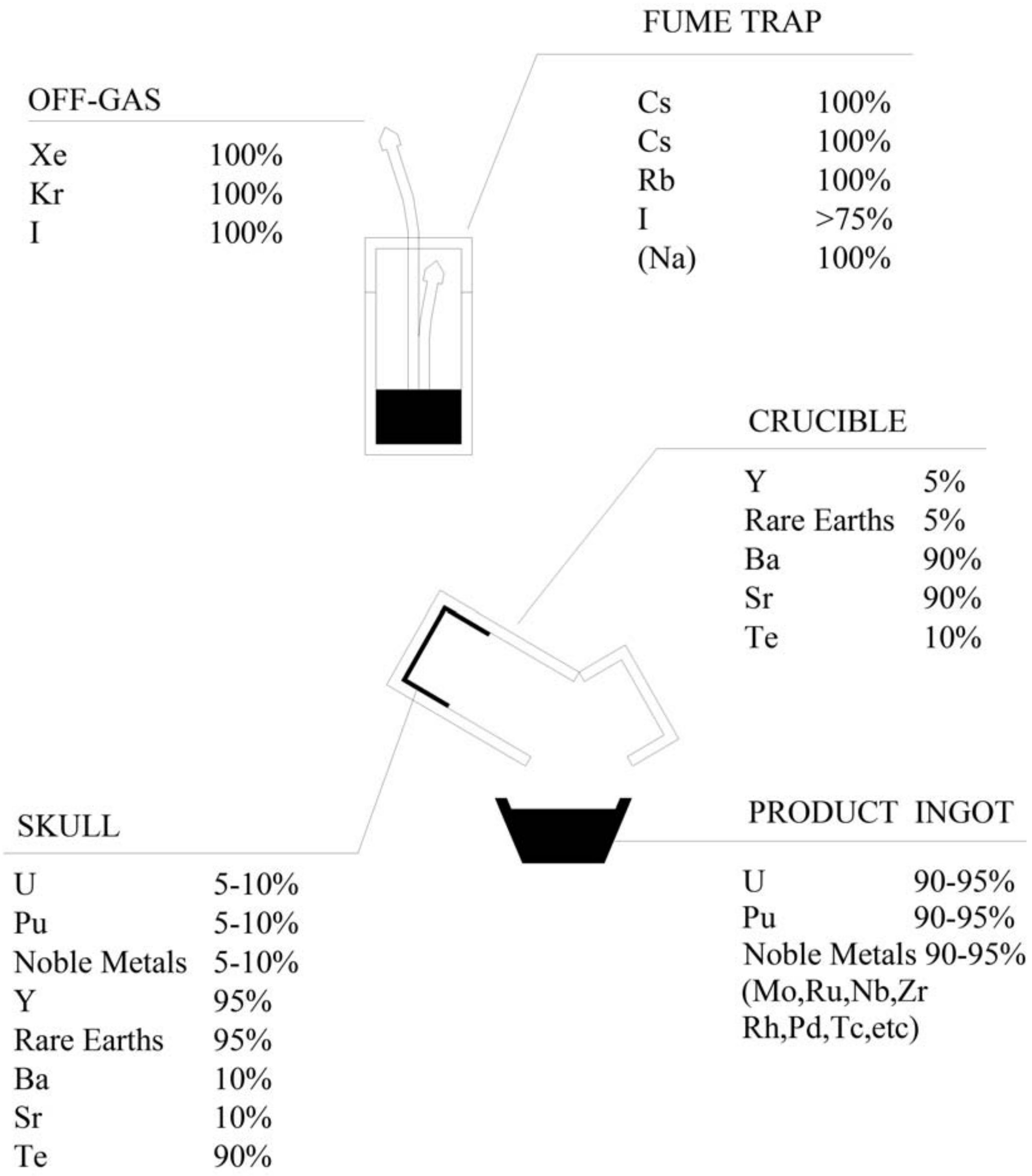


FIG. 18. Schematic description of the melt-refining process for spent metallic fuel.

therefore, comes down progressively. The FPs can be removed from the salt to a considerable extent by ion exchange with suitable zeolites, and the cleaned salt stream can be sent back to the electro-refiner. After several cycles, the entire FP-laden salt will have to be rejected and disposed of as a waste.

A licence for pyroprocessing of all of the EBR-II spent fuel has been obtained. EMT technology can also be used for treating oxide fuels with addition of the reduction process to the flow sheet [43, 196].

6.8.4. Future fuel reprocessing scenarios

The future scenarios could be operation of an ADS subcritical reactor to transmute waste with significant fuel burnups, ~300 GW·d/t HM, using a non-fertile fuel to preclude the generation of additional TRU elements. After discharge, the fuel would be processed to recover unburned TRU elements for recycle and to extract newly-generated LLFPs (i.e. Tc and I) for transmutation in the thermalized regions of the reactor. As it was economically desirable to process the irradiated transmuter fuel with short cooling times, and because the amount of the fuel to be processed annually contains a comparatively small amount of heavy metal (relative to the annual throughput of heavy metal in LWR irradiated fuel), pyrochemical processing was chosen for the treatment of the transmuted fuel.

Initially, a metal alloy fuel (TRU element–Zr) was assumed as the transmuter fuel. The volume fraction of Zr was assumed to be large (about 90%) and this large quantity of Zr coupled with high radiation and decay heat levels for the fuel drove the process selection towards pyrochemistry.

The process initially selected, known as PYRO-B, involved the digestion of the zirconium matrix as the first step. Entrainment of small amounts of cadmium in the recovered $ZrCl_4$, however, led to abandonment of the PYRO-B process in favour of a direct electro-refining process in which the TRU elements are electro-deposited on a steel cathode and zirconium is left behind with noble metal FPs and incorporated as a matrix material in the metal waste form.

The objectives of the Global Nuclear Energy Partnership Technical Demonstration (GNEP-TD) Programme include demonstration of advanced fuel cycles, namely the pyrochemical process based on molten salt electro-refining for the spent transmuter fuel from the advanced burner reactor (ABR) which is an SFR. The AFCF planned under the programme is aimed at the development and demonstration of the remote refabrication technology for TRU element fuels of the ABR and pyroprocessing technology for the spent ABR fuels from bench scale to engineering scale and the capacities of the respective plants would be eight lead test assemblies/a and ~1 t/a, respectively.

Efficiency tests were conducted after the integration of the main steps in electro-refining as well as cathode product consolidation steps in a 50 kg batch using TRU element based fuel. Experiments showed significantly lower losses compared to theoretical estimates.

The recent technological development efforts at INL range from laboratory scale studies of new processing concepts, such as the electrolytic reduction of spent LWR oxide fuel, to the engineering scale demonstration of high throughput uranium electro-refining. To accommodate processing of other types of fuels, namely oxide spent fuel, in this non-aqueous process, the head-end treatment is modified. For example, an initial Li/Li_2O step can be added to convert the oxide fuel to the metallic form. A head-end processing step (termed DEOX for its emphasis on de-cladding via oxidation) is being developed for the treatment of spent oxide fuel by pyroprocessing techniques. The head-end step employs high temperatures to oxidize UO_2 to U_3O_8, resulting in the separation of fuel from cladding and the removal of volatile FPs. An electrolytic reduction process for spent LWR oxide fuel is being developed at INL as part of an integrated process for the pyrochemical treatment of spent nuclear oxide fuel in conjunction with the DEOX process in collaboration with KAERI under the international nuclear energy research initiative [104, 105, 197].

6.8.5. High level waste management

Spent fuel reprocessing is not currently performed in the USA on a commercial scale. The emphasis remains on spent fuel storage. Vitrification of some defence wastes has, however, been carried out. Efforts to clean up the accumulated wastes in old weapons laboratories by converting them into glass or glass–ceramic matrices are also underway.

US law requires the US DOE to develop a temporary storage facility, a transport system and a permanent repository for used nuclear fuel. An initiative to find an interim storage for used fuel has been undertaken by Private Fuels Storage Limited (PFS). PFS is a consortium of eight US utilities to build the interim storage facility on the Goshute Native American reservation in Utah. Construction of the facility was thwarted by the formal opposition of the State of Utah. PFS continues to work on other aspects of the project, including the container and mode of safe transport.

The Nuclear Waste Policy Act of 1982 and its 1987 amendments provide for the US DOE to conduct research and then construct an underground repository for the permanent storage of used nuclear fuel and HLW from US utilities. Yucca Mountain, a region of the Nevada Test Site was selected for study as a possible repository site. Good progress has been made on this research, but there is stiff political opposition from the state of Nevada. The Yucca Mountain programme experienced major progress in 2001. The preliminary site suitability evaluation report was published in August 2001. This report confirmed that the site and the engineered facility for the repository could meet the national radiation safety standards for protecting the public’s health and the environment [198].

Currently, there is renewed interest in the use of metallic fuel in FRs in view of the possibility of achieving better breeding ratios and also actinide burning. EBR-II has accumulated a lot of operating experience with metallic fuel, both with respect to its in-pile behaviour, and reprocessing and waste management.

Some details of the ANL experiments on the ceramic waste form are described here. Laboratory scale experiments were started in 1986. HUP was carried out in graphite dies (990 K, 28 MPa), and HIP in 2.5 cm diameter stainless steel bellows (873–1023 K, 170 MPa, 1 h). Engineering scale HIP experiments using inactive materials were carried out (1996) in steel cans with a diameter of 11 cm, a height of 23 cm and a volume of 2300 cm^3. The HIP can was provided by ANSTO under a development agreement. In the subsequent experiments, temperatures in HIP experiments were raised to 1123–1173 K. The MCC-1 leach tests (static test in de-ionized water at 363 K up to 28 d) have shown the leaching behaviour to be comparable with that of the standard borosilicate glass waste form.

The technology to compress high level radioactive waste in a remotely operated hot cell was developed jointly by ANSTO and ANL in 1997. Remediation of nuclear waste at the Hanford weapons facility has also been taken up by ANSTO. A SYNROC/glass composite waste form was tested for simulated insoluble HLW sludges at Hanford [199].

Operations with the radioactive materials commenced using the parameters developed from inactive testing. The salts were taken from the Mark IV electro-refiner after treatment of one hundred driver subassemblies (EBR-II). The charge contained 9.4 wt% uranium chlorides, 0.5 wt% TRU element chlorides and 5.2 wt% FP chlorides. Four kilograms of salt were taken and mixed with 36 kg of zeolite in a V-mixer (thus, 10 wt% salt in zeolite); 10 kg of borosilicate glass frit was added subsequently and blended in the same mixer assembly. With this 50 kg of material, ten HIP experiments were carried out using cans 11 cm in diameter.

Thus, the production of a ceramic waste form was demonstrated at ANL with radioactive materials on a nearly realistic scale. Based on the technology demonstration work on 1 t of spent metallic fuel, ANL has estimated that their programme of electro-metallurgical processing of 25 t HM in the period 2005–2017 will produce 5.85 t of metallic waste form (3.8 m^3 disposal volume inclusive of package) and 51.2 t of ceramic waste (33 m^3 disposal volume inclusive of package). These waste volumes are still considered economical when compared to the direct disposal of spent fuel. The HEU metal in the spent fuel would require dilution by depleted uranium due to criticality considerations. This would have generated a total waste volume of 309 canisters, whereas reprocessing would lead to a waste volume of only 59 canisters.

6.8.5.1. *Alternative to hot isostatic pressing: Pressureless sintering*

Carrying out HIP under hot cell conditions is cumbersome. ANL has developed an alternative procedure, called pressureless sintering, to produce the ceramic waste form. The zeolite, waste-laden salt and the glass binder are mixed together, and poured into graphite moulds that are passed through a tunnel kiln at a fixed temperature and time (1123 K, 4 h, 1 atm pressure argon). For 1 kg samples, the furnace temperature is raised at 10 K/min to 773 K where it is held for about 2 h. It is then raised to 1198 K for the final 15 h. Specimens of 140 kg were made by holding the furnace at 1198 K for 100 h [120]. Initial studies indicated that a higher vol% of glass binder (50%) is needed to fabricate a reproducible waste form, and that the density and leach behaviour are the same as those of the HIP product.

7. R&D AREAS FOR FUTURE FUEL CYCLE TECHNOLOGIES

7.1. INTRODUCTION

Reprocessing and waste management have been especially vibrant areas of international R&D efforts because of the need to recover valuable fissile material for recycling. Immense opportunities are available for basic and applied research work in these areas. This is due to the challenges posed by the presence of a number of elements from various groups in the periodic table in the solution of irradiated fuel, and the high level of beta and gamma radiation. The use of a variety of organic as well as all aqueous reagents, and the need for remote operations and maintenance also provide opportunities for innovation. The progress made by various research groups in this area have been covered in many international meetings, some of which have been dedicated to reprocessing and waste management, such as the “RECOD”, “ATALANTE”, “IPRC” and “Waste Management” series of conferences. The “GLOBAL” series of meetings have also provided valuable information regarding the recent R&D activities relevant to the FR fuel cycle.

However, as compared to thermal reactor systems, reports specific to R&D on the FR fuel cycle have been limited, due to the fact that FR programmes are currently only being pursued by a select group of countries. However, separation sciences, which form the heart of reprocessing and waste management, lend themselves to a variety of R&D areas which are studied even in countries where no nuclear fuel cycle activities are carried out.

7.2. REPROCESSING AND WASTE MANAGEMENT

Aqueous reprocessing is still the preferred route for recovery of fissile material from thermal reactor fuels. In the case of FRs, fuels irradiated to burnup levels as high as 150 GW·d/t HM and cooling times as short as six months have been reprocessed by the aqueous route. However, the burnup targets for FR fuels are now as high as 200 GW·d/t HM and the fuels have to be reprocessed after relatively short periods of cooling. There is no commercial experience with reprocessing of fuels irradiated to such high burnup levels. The high radiation fields and high plutonium content of these fuels will provide a number of challenges to process chemists and engineers in designing extractants and developing flow sheets.

The nuclear fuel cycle for FRs has been a subject of intense R&D in a number of countries. Besides the development of indigenous technologies for the various steps in the fuel cycle, such as fuel fabrication, reprocessing and waste management, R&D in the fuel cycle is important for establishing a robust fuel cycle for FRs which would not only address economy, safety and proliferation resistance, but also the main concerns of the public, which relate to the presence of high concentration and quantities of plutonium and FPs in the reprocessing and waste management steps for FRs.

The key drivers for innovation and R&D in reprocessing can be listed as follows:

- Improving the economy of operations;
- Enhancing the operational life of the equipment and availability of the reprocessing plant;
- Reduction in waste volumes;
- Maintaining a high level of safety;
- Attaining an ‘ideal’ process flow sheet which would recover valuable FPs present in the irradiated fuel;
- Enhancing proliferation resistance.

The above key drivers relate to the following areas of R&D in reprocessing:

- Studies on the alternate flow sheets for the main process where the fissile material is recovered;
- Studies on the alternate flow sheets for the auxiliary processes, such as MA partitioning, FP recovery, etc;
- Development of materials with improved corrosion resistance;
- Development of equipment which is more amenable for remote operations and maintenance, and especially equipment with maintenance-free operations;
- Modelling and simulation of flow sheets as well as of equipment operations, so that the number of experiments involving radioactive materials can be reduced;
- Development of instrumentation for on-line nuclear material accounting, on-line flow monitoring, characterization of process streams, etc;
- In-service inspection and repair technologies.

The aqueous reprocessing route for FR fuels based on oxide, carbide and nitrides has mainly been through the PUREX process which utilizes TBP as the extractant for separating uranium and plutonium. The high burnup levels achieved in FRs have provided the impetus for studies on extractants that are stable at high radiation levels and produce degradation products that do not interfere with the extraction and can be removed easily. TBP has been the mainstay of the process and has been universally accepted as a nearly ideal extractant. However, a few features of TBP such as “red oil” formation, tendency for degradation with products not easily amenable for treatment, tendency for third phase formation during the extraction of plutonium, and issues related to ultimate disposal of the extractant have provided the impetus for the development of alternate extractants to replace TBP.

Higher homologues of tri-butyl phosphates have been studied in some countries. TIAP has, in fact, been used as the extractant for reprocessing of FR fuel in the Russian Federation [198]. However, the experience with alternate TAPs has been limited and it remains to be seen whether the advantages, in terms of lesser aqueous solubility and tendency for third phase formation, will be offset by other factors.

Since the 1980s, alternate extractants based on amides have been studied in many countries, especially France, as replacements for tri-butyl phosphate. Besides possessing extraction properties similar to tri-butyl phosphates, the amides have other advantages, such as low aqueous solubility and the possibility of uranium–plutonium partitioning without reduction of plutonium to the lower oxidation state. The amides contain only the elements C, H and N, and O atoms, and are, therefore, completely incinerable. Flow sheets based on the amides have been tested in various countries including France. However, amides are yet to be deployed even in reprocessing plants for thermal reactor fuels, and considerable experience on an industrial scale is required before they are accepted as a replacement for TBP. In addition, a number of candidate amides have been studied, and an evaluation leading to the identification of a specific amide extractant is necessary. However, because of their inherent advantages, development of amides can be expected to be a focal point of international efforts with respect to the main flow sheet for reprocessing.

The solvent extraction and ion exchange processes used in reprocessing generate significant quantities of aqueous wastes. Reduction in the number of purification cycles is one approach for addressing this issue. This will also be aided by adopting extractants that promise higher DFs from FPs. Development of processes that generate reduced quantities of aqueous waste and recovery of actinides and FPs from such wastes is an area of intense R&D. Similarly, reduction of alpha-bearing solid waste by suitable decontamination techniques without generation of secondary waste is another issue. The existing processes also use organic reagents (e.g. N-dodecane as the diluent in the PUREX process) which have to be destroyed after repeated use.

The equipment designs of the PUREX process have to meet the demanding requirements of FR fuel reprocessing. In particular, fuel disassembling, dissolver, feed clarifiers and solvent extraction equipment, such as centrifugal extractors and conversion equipment, are a few examples which require intense R&D.

7.3. ACTINIDE PARTITIONING FROM HIGH LEVEL WASTE

The recovery of MAs and LLFPs is essential for reducing repository volume requirements. In the late 1980s, studies at ANL resulted in the development of a unique extractant — N-octyl, Phenyl N, N — diisobutyl CMPO for the recovery of actinides from HLW solutions. This extractant can be used for recovery of actinides from the high level aqueous waste generated in thermal reactor as well as in FR fuel reprocessing. Flow sheets based on CMPO have been developed and demonstrated on a pilot plant scale in Europe as well as in other countries. However, there is no reprocessing plant in the world which has integrated MA partitioning with the main flow sheet of reprocessing. Newer extractant systems, such as diglycolamides, have since emerged, which promise better extraction characteristics. Experience with these extractants is once again limited to lab scale experiments.

The separation of actinides from lanthanides in the mixture, which is obtained by the separation processes described above, is another area where work is still in the laboratory domain. Several extractants, such as HDEHP, BTP, CYANEX 301, etc., have all been considered as candidates, but there is no consensus yet on which one of these would be the most suitable extractant. In addition, there has been no techno-economical evaluation of the separation of MAs and their subsequent utilization.

France along with other OECD countries has carried out a number of studies on the development of flow sheets for partitioning of MAs. An international review carried out recently on this programme, emphasized the importance of further studies before the partitioning flow sheets can be considered for commercial applications.

It is also well known that the HLW solution produced by reprocessing of FR fuels contains a wealth of isotopes such as ^{137}Cs and ^{90}Sr that could be used for societal applications. The routine recovery of large quantities of ^{137}Cs has been reported from the Russian Federation. However, there are no reports on recovery of this isotope from irradiated FR fuels. Plutonium-based fuels at high burnup contain significant quantities of palladium, whose isotopes are stable (except ^{107}Pd which has a half-life of 6.5×10^6 years and emits soft beta and gamma radiation). Recovery of palladium could not only help to avoid the problems in waste immobilization arising from the incompatibility of Pd with the glass matrix, but also yield significant quantities of palladium for many industrial applications. It is conceivable that in the future, the elements of waste management involving recovery of MAs and FPs could be integrated with fuel reprocessing. However, these flow sheets must enable recovery of the desired elements with a high degree of decontamination of others.

The large scale deployment envisaged for FRs would require setting up reprocessing plants which would handle large inventories of fissile material. Both from the point of view of safety and process efficiency, as well as for addressing the concern of proliferation of nuclear materials, more accurate and reliable on-line monitoring equipment and nuclear material accounting techniques would have to be developed. The timely detection of accumulation or loss of significant quantities of fissile material in process equipment and systems in a large capacity reprocessing plant is still a challenge.

For a comprehensive and sustained growth of fuel cycle facilities, it is inevitable that the technology is assessed vis-à-vis nuclear reactors, in which domain considerable standardization on an international scale has taken place. Emphasis on the increased lifespan of reprocessing and waste management plants in line with reactor systems would, thus, demand more intensive R&D on materials, fabrication processes and in-service inspection technologies, including on-line corrosion monitoring of critical equipment such as dissolvers and evaporators. Regulatory processes for the fuel cycle facilities can be expected to include a probabilistic safety assessment as applied to reactor systems. With ever increasing emphasis on the reduction of exposure, increased automation, robotics and radiation hardened electronics would need to be deployed in future fuel cycle facilities. This would become especially crucial when plants actually start handling recycled plutonium.

7.4. PYROCHEMICAL REPROCESSING

Pyrochemical reprocessing has been recognized to be an important alternative to the aqueous reprocessing route adopted presently. The basic features of some pyrochemical routes and their advantages were discussed in Section 2. Pyrochemical processing of metallic fuel as well as oxide fuels has been demonstrated in the USA and the Russian Federation, respectively. A Japanese study has concluded that at a process scale up to 50 t/a, the pyrochemical processing route will be more economical than the aqueous reprocessing route. The constraint for processing capacity via the pyrochemical route arises from the fact that it is basically a batch process. It remains to be seen whether the pyrochemical process can be made continuous.

The use of molten chloride salts as a medium for processing requires operation at high temperatures (typically 500–550°C). The development of alternative salt media for achieving lower operating temperatures, the development of materials and coatings that are compatible with molten chloride salts over extended periods of time, processes for recovery of residual actinides and selected LLFPs from waste salts are areas of R&D, which can make the pyrochemical processes more competitive and better address waste management issues. The management of salt waste arising from the pyrochemical process, including its immobilization in a glass matrix, has also been studied on a lab scale, and much more work is required before such technology can be realized on a commercial scale.

The experience with pyrochemical reprocessing so far has been on a very small scale as compared to aqueous reprocessing. It can be expected that many more areas of R&D could arise as pyrochemical reprocessing enters into the commercial domain. However, considering its promise with regard to proliferation resistance and its ability to handle high burnup short cooled fuel, R&D in this area is bound to provide breakthroughs for the development of innovative FR systems.

7.5. OTHER APPROACHES TO REPROCESSING

A paradigm shift in reprocessing technologies could be related to the use of so called ‘green’ processes that can nearly eliminate the generation of aqueous waste or use environmentally friendly reagents. An example of such an approach is the use of supercritical fluid extraction (SFE) with CO_2, which is emerging as an important area of research for waste management, due to increasing awareness about the environmental impact of using organic solvents. The Super-Direx process has been proposed, by Japanese researchers, for reprocessing irradiated fuels by SFE [200]. This process is being improved to enable simultaneous recovery of MAs along with U and Pu. At KRI, Russian Federation, studies with freons as super critical media are being pursued for reprocessing applications.

However, these studies are at a preliminary stage and considerable R&D also has to be carried out to address safety issues before the actual use of SFE in hot cells will become possible [201].

Room temperature ionic liquids (RTILs) are emerging as versatile media for recovery processes. Due to their low volatility, non-flammable nature and stability to radiation, their use in separations has evolved as an important area of R&D. Extraction of actinides and selected FPs using ionic liquids as diluents in place of n-dodecane has recently been reported. RTILs possess an additional advantage in that their physical and chemical properties can be tailored by altering the functional groups. However, RTILs are expensive as compared to dodecane, and are yet to be deployed on a large scale in recovery processes for the nuclear fuel cycle. Considering their important advantages, it could be expected that RTILs will continue to be explored for reprocessing and waste management operations, and sustained R&D could result in applications in the coming decades.

Reducing the salt content in waste streams, especially from off-gas scrubbing, solvent washing and chemical analysis, is another area of R&D.

7.6. WASTE IMMOBILIZATION

The volume of high level aqueous waste produced in reprocessing is mainly dictated by the FP content, which again is limited by the achievable loading in the matrix for waste immobilization. The loading limits arise from heat generation from the FPs, and from the concentration limits for the elements, above which the glass suffers de-vitrification. Thus, in the case of the FR fuel cycle, there is an incentive for development of waste matrices that can tolerate higher salt content and can also remain stable for extended periods of time in a geological environment. Ceramic matrices for waste immobilization have been under development in many countries for this purpose. However, there is no industrial scale experience on immobilization of waste in ceramic matrices. Identification of matrices with lower melting temperatures, and tolerance for higher loading in terms of salt content as well as radioactivity content are important areas of innovation.

Finally, perhaps the most important subject related to the fuel cycle, which has occupied the public mind, is the ultimate disposal of radioactive waste in a geological repository. Public acceptance can only be achieved by developing and implementing technologies that could ensure near-quantitative recovery of LLRNs from HLW, so that the monitoring period for the waste could be reduced to a few hundred years. In addiiton, effective recovery of these elements would permit a significant reduction in the requirements for repository space and enable public acceptance, which could otherwise be a limiting factor for the growth of reactor systems. However, independent of the approach to waste management, studies on migration of LLRNs in the environment, development of stable waste forms and models that could reliably predict the long term behaviour of waste forms will continue to be important topics of research.

8. CONCLUSION

It is very likely that a ‘nuclear renaissance’ will take place in several countries in the coming decades. Confidence in the nuclear renaissance is enhanced due to the high performance and safety record of nuclear power plants and increasing environmental concerns, combined with the issues of sustainability of energy resources compatible with economic aspirations. The production of electricity through nuclear fission is expected to increase from the present level of 370 GW(e) to 500 GW(e) by 2030 and 2000 GW(e) by 2050 [202].

Fast spectrum reactors and associated fuel cycle facilities will play an important role to enable nuclear energy systems to make enhanced contributions to the production of energy and management of nuclear wastes beyond 2020. FRs will act as breeders for enhancing nuclear energy growth, or burners to effectively reduce the stockpile of fissile materials and MAs, depending on national approaches. It is, however, clear that, independent of the manner in which the nuclear scenario evolves, fuel cycle technologies will play a significant role in the growth of FRs, for both effective utilization of fissile materials and management of radioactive waste.

Closing of the fuel cycle is accepted by a large number of countries with mature nuclear technologies as an important element of future nuclear energy scenarios. Only a few countries possess the technologies for U–Pu fuel fabrication, fuel reprocessing and HLW management. Commercial scale reprocessing experience with FR fuels does not exist in any country. Even reprocessing of thermal reactor fuels is only being pursued by a few countries, namely France, Japan, the United Kingdom, the Russian Federation and India. This situation is likely to change in the future as closing the fuel cycle with FRs is necessary for sustainability reasons. Furthermore, fuel cycle technologies may include recovery of MAs and FPs for transmutation into short lived radionuclides. Along with the development of ADS systems, technologies for effective isolation of the MAs and FPs, and technologies for incorporating them into fuels for burning in FR and ADS systems will emerge as intense areas of R&D.

The contribution of fuel cycle operations to the cost of nuclear power depends not only on the technologies employed but also on the pricing policies adopted by various countries. The cost of nuclear fuel cycle operations constitutes 5–20% of unit energy costs. While this must be considered as a small contribution, additional efforts shall be made to further lower these costs by developing advanced technologies. The other key issues, such as the effect of fuel cycle operations on waste management, environmental impact and proliferation resistance will also be of primary importance for future developments. It is generally perceived that fuel cycle technologies are less proliferation resistant as compared to nuclear reactor systems, that they require higher levels of maturity in complex technologies, and, therefore, may not be readily accepted by all countries that would like to pursue FR programmes. Thus, the possibility of establishing multinational fuel cycle facilities could become an important area of international endeavour. At the same time, countries that would like to master fuel cycle technologies in order to achieve self-sufficiency with regard to the generation of nuclear power will probably continue intensive R&D activities on their own.

It can be concluded that the scale of sharing available data on operating experience and development of international standards for various equipment and processes used in the nuclear fuel cycle is not adequate. This issue needs further emphasis and focus. Certain aspects of fuel cycle technologies, such as safety, waste management and proliferation resistance, could lend themselves for wider international collaboration. The IAEA is, thus, well positioned to play an important role in the development of FR fuel cycle technologies, around the world.

An examination of the various facets of FR fuel cycle technologies indicates that paradigm shifts are not expected in the head-end steps of the fuel cycle. However, with the limited experience in the aqueous route of FR fuel reprocessing and the increasing laboratory scale experience with pyrochemical reprocessing, and with the increasing emphasis on achieving high burnup, it is to be expected that R&D programmes will attempt to identify the relative merits and disadvantages of these two approaches. Ultimately, the choice of reprocessing technology may be as much governed by the issues of safety and proliferation resistance as by economic considerations.

This book has attempted to provide an overview of various aspects of fuel cycle technologies for FRs, developed in Member States in the past, and also to highlight the current programmes and future plans of Member States. Additional reading materials are recommended in the annex, which could enable the reader to gain a better appreciation of the programmes in various countries. The data provided in the publication suggest that there is much convergence in the approaches of various countries alongside with interesting variations that reflect the difference in national approaches. R&D areas which are presently being addressed by various countries as well as emerging areas, in which more R&D is expected, have been described in this report. The description of R&D priorities is not intended to be comprehensive; breakthroughs in any of these technologies could alter the processes and equipment used in fuel cycle facilities.

Finally, the basic research and theoretical studies on a variety of subjects, including materials and equipment development, inspection technologies, etc., are likely to provide the breakthroughs needed for the rapid growth of fuel cycle technologies. It is essential to harmonize approaches towards the safety assessment of nuclear facilities, so that they can be designed to withstand natural, as well as unnatural environments for a lifetime of about 60 years, following the extended lifetime trends in reactor systems. International collaboration for the evolution of approaches and the safety guidelines is a key to success in this endeavour.

International programmes, such as INPRO and GIF, provide impetus for the development of innovative fuel cycle technologies. The success of these programmes is vital to catalyse the growth of robust, safe, economical, environmentally acceptable and proliferation resistant fuel cycle technologies.

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ABBREVIATIONS

ABR	Advanced burner reactor
ACP	Advanced spent fuel conditioning process
ADS	Accelerator driven subcritical system
AFCF	Advanced Fuel Cycle Facility
AFCI	Advanced Fuel Cycle Initiative
ANL	Argonne National Laboratory
ANSTO	Australian Nuclear Science and Technology Organization
APM	Atelier Pilote de Marcoule
ARTIST	Amide based radio resources treatment with interim storage of TRU
ASM	Advanced servo-manipulator
ATW	Accelerator based transmutation of waste
BARC	Bhabha Atomic Research Centre
BNFL	British Nuclear Fuels Ltd
BNL	Brookhaven National Laboratory
BREST	Russian lead cooled fast reactor
BRET	Breeder reprocessing engineering test
BSM	Bilateral servo-manipulator
BTP	Bis-triazinyl-1,2,4-pyridines
CCD	Chlorinated cobalt di-carbollide
CCIM	Cold crucible induction melting
CEA	Alternative Energies and Atomic Energy Commission
CFC	Closed fuel cycle
CFTC	Consolidated Fuel Treatment Centre
CMPO	Carbamoyl methyl phosphine oxide
CORAL	Compact reprocessing of advanced fuels in lead cells
CPF	Chemical Processing Facility
CSEX	Cs extraction
DDP	Dimitrovgrad Dry Process
DF	Decontamination factor
DFR	Dounreay Fast Reactor
DFRP	Demonstration fast reactor plant
DIAMEX	DIAMide EXtraction process
DHOA	Di-hexyl octanamide
DOHA	Di-octyl hexanamide
DIDPA	Di-isodecylphosphoric acid
DOE	Department of Energy (USA)
DTPA	Diethylene-triamine-penta-acetic acid
EBR-II	Experimental Breeder Reactor-II
ECN	Electrochemical noise
EMT	Electrometallurgical treatment
FBR	Fast breeder reactor
FBTR	Fast breeder test reactor
FFTF	Fast Flux Text Facility
FMEF	Fuels and Materials Examination Facility
FR	Fast reactor
FREGAT	Facility for fluoride volatility reprocessing of spent fuel
FP	Fission product
GNEP	Global Nuclear Energy Partnership
HDEHP	Di-ethyl hexyl phosphoric acid
HEU	High enriched uranium
HIP	Hot isostatic pressing
HLLW	High level liquid waste

HLW	High level waste
HPA	Heteropolyanions
HUP	Hot uniaxial pressing
IFR	Integral fast reactor
IGCAR	Indira Gandhi Centre for Atomic Research
ILW	Intermediate level waste
INL	Idaho National Laboratory
INPRO	International Project on Innovative Nuclear Reactors and Fuel Cycles
ITU	Institute for Transuranium Elements
JAEA	Japan Atomic Energy Agency
JHCM	Joule heated ceramic melter
KRI	Khlopin Radium Institute
LHR	Linear heat rates
LILW	Low and intermediate level waste
LLFP	Long lived fission product
LLRN	Long lived radionuclide
LLLW	Low level liquid waste
LLW	Low level waste
LOTIS	Laser based electro-optic tube inspection system
LWR	Light water reactor
MA	Minor actinide
MFL	Magnetic flux leakage
MOX	Mixed oxide
MSM	Master–slave manipulator
NAG	Nitric acid grade
NDT	Non-destructive testing
NEXT	New extraction system for TRU recovery
ORNL	Oak Ridge National Laboratory
PEC	Pulsed eddy current
PEG	Polyethylene glycol
PFR	Prototype fast reactor
PFS	Private Fuels Storage Limited
PGM	Platinum group metal
PHWR	Pressurized heavy water reactor
P&T	Partitioning and transmutation
PUREX	Plutonium uranium extraction
PWR	Pressurized water reactor
RETF	Recycle Equipment Test Facility
RIAR	Research Institute of Atomic Reactors
RTIL	Room temperature ionic liquid
SANEX	Selective actinide extraction
SCP	Solid coupled probe
SETFICS	Solvent extraction for trivalent F-elements intra-group in CMPO-complexant system
SESAME	Separating americium from curium
SFE	Supercritical fluid extraction
SFR	Sodium cooled fast reactor
SIMPACTR	SIMulation Program for ACTinide Recovery
SLOFEC	Saturation low frequency eddy current
SNF	Spent nuclear fuel
SREX	Sr extraction
SYNROC	Synthetic rock
TAP	Tri-alkyl phosphate
TBP	Tri-n-butyl phosphate
TIAP	Tri-isoamyl phosphate

TMAHDPTZ	2-(3,5,5-trimethylhexanoylamino)-4,6-di-(pyridin-2-yl)-1,3,5-triazine
TODGA	Tetra-octyl diglycol amide
TOP	Traitement d’Oxydes Pilote
TOR	Traitement d’Oxydes Rapides
TRPO	Tri-alkyl phosphine oxide
TRU	Transuranic
TRUEX	TRU extraction
TRUMP-S	TRansUranic Management through Pyropartitioning Separation
TVF	Tokai Vitrification Facility
UNEX	Universal extraction
UNGG	Graphite gas cooled reactors
UREX	Uranium extraction
VLLW	Very low level waste
WAK	Karlsruhe Reprocessing Plant
WIP	Waste immobilization plant

CONTRIBUTORS TO DRAFTING AND REVIEW

Baldev, R.	Indira Gandhi Centre for Atomic Research, India
Busurin, Y.	International Atomic Energy Agency
Depisch, F.	International Atomic Energy Agency
Ganguly, C.	International Atomic Energy Agency
Ghosh, S.	Indira Gandhi Centre for Atomic Research, India
Koganti, S.B.	Indira Gandhi Centre for Atomic Research, India
Kormilitsyn, M.	Russian Institute of Atomic Reactors, Russian Federation
Koyama, T.	Japan Atomic Energy Agency, Japan
Kudryavtsev, Ye.	State Atomic Energy Corporation Rosatom, Russian Federation
Kutty, K.V.G.	Indira Gandhi Centre for Atomic Research, India
Masson, M.	Alternative Energies and Atomic Energy Commission, France
Nagarajan, K.	Indira Gandhi Centre for Atomic Research, India
Natarajan, R.	Indira Gandhi Centre for Atomic Research, India
Nawada, H.P.	International Atomic Energy Agency
Prabhakara Reddy, B.	Indira Gandhi Centre for Atomic Research, India
Rao, P.R.V.	Indira Gandhi Centre for Atomic Research, India
Srinivasan, T.G.	Indira Gandhi Centre for Atomic Research, India
Swaminathan, P.	Indira Gandhi Centre for Atomic Research, India
Wattal, P.K.	Bhabha Atomic Research Centre, India

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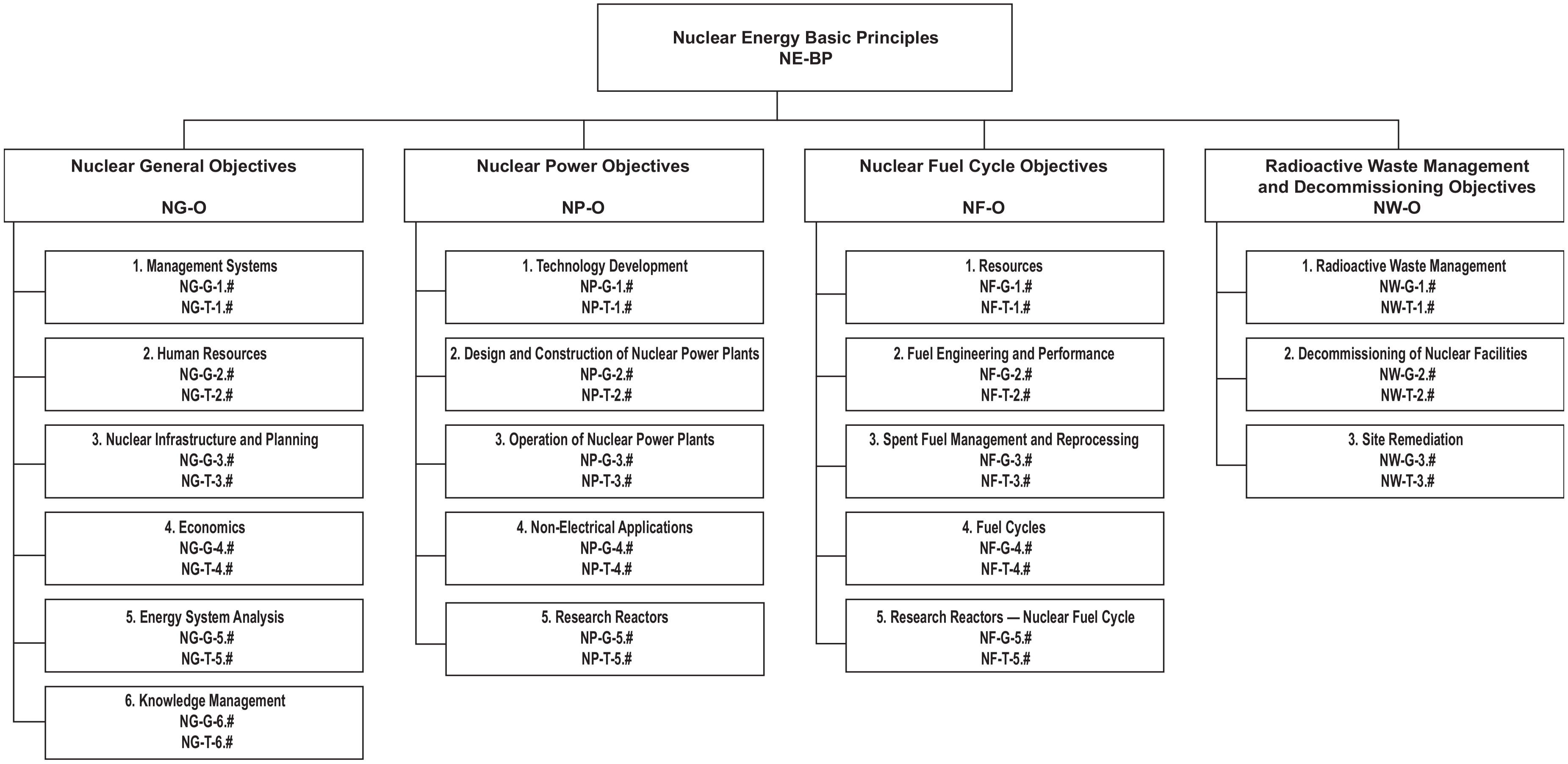

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