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# *Status and trends in spent fuel reprocessing*



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## FOREWORD

Management of spent fuel arising from nuclear power production is a crucial issue for the sustainable development of nuclear energy. While reprocessing of spent fuel was historically the favoured strategy for the back end of the fuel cycle, in the past few decades some countries have turned to other options. Specifically some countries have adopted a direct disposal or a 'wait and see' strategy, partly in response to concerns such as nuclear weapons proliferation, public acceptance and economics.

The debate on the proliferation issue associated with spent fuel reprocessing is not new. It led to the International Nuclear Fuel Cycle Evaluation (INFCE) organized by the IAEA in the late 1970s. Another IAEA initiative, the Expert Group on International Spent Fuel Management, had looked at the international issues of spent fuel management, including possible roles for the IAEA. Thereafter, the Regular Advisory Group on Spent Fuel Management was established in the early 1980s as an IAEA instrument providing surveys of the status of spent fuel management, as well as advice to the relevant IAEA programme. The work of the IAEA has continued through the Technical Working Group on Nuclear Fuel Cycle Options to which the Group was merged in 2001.

The IAEA has issued several publications that provide technical information on the global status and trends in spent fuel reprocessing and associated topics, and one purpose of the present publication is to provide an update of this information. However, the scope of this publication has been significantly expanded in an attempt to make it more comprehensive by including more information on emerging technologies. A meeting of the Scientific Forum on the topic of Fuel Cycle Issues and Challenges, held during the 48th General Conference of the IAEA, 20–22 September 2004, provided an opportunity to review and discuss several of the issues associated with spent fuel management and provided some of the input to finalize this publication.

For the preparation of this publication, an Advisory Group meeting was held in 2000, followed by consultancy meetings in 2001 and 2002, which provided initial inputs, including country reports. The contributions of all who brought valuable help in drafting and reviewing the report (listed at the end of this publication) are greatly appreciated. The IAEA officer responsible for this publication was J.S. Lee of the Division of Nuclear Fuel Cycle and Waste Technology.

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# 1. INTRODUCTION

Management of spent fuel arising from nuclear power production has long been considered an important issue due to the political, economic, and societal implications associated with it. In view of the large amount of spent fuel being progressively added to the cumulative inventory in the world, the significance of spent fuel management will continue to grow in the future.

In recognition of the importance of spent fuel reprocessing in the back end of the fuel cycle, the IAEA has provided a forum for exchange of information on the status and trends in spent fuel reprocessing since the 1970s, from which several publications have appeared [1],[2],[3].

This report updates previous IAEA technical documents which have focused on Purex based conventional reprocessing industry, and expands the scope of the analysis, by including technical information on emerging technologies associated with the reprocessing of spent fuel. This significant revision reflects growing interest in the search for innovative nuclear systems in recent years, as exemplified by international initiatives, such as INPRO<sup>1</sup> (IAEA), MICANET<sup>2</sup> (EU) and GIF<sup>3</sup> (USA), in which the technical concepts of emerging technologies are expected to play important roles in the sustainable management of spent fuel.

## 1.1. SPENT FUEL MANAGEMENT OPTIONS

In the classic approach to the back end of the fuel cycle, the closure of the fuel cycle by means of reprocessing and recycling in fast breeder reactors was regarded as a standard strategy. This strategy was reinforced by the oil shock in the 1970s, some repercussions of which are still lingering in certain countries, especially those devoid of natural energy resources.

The reprocessing and recycling strategy was based on assumptions of a rapid growth in nuclear energy and uranium demand. However, the growth in nuclear energy from the 1970s onward turned out to be more sluggish than originally assumed, and forward plans were progressively downsized. Due to these and other new realities, an increasing number of countries have abandoned the closed fuel cycle, either by turning to the once-through cycle, and adopting direct disposal of spent fuel, or by deferring a final decision on the fate of spent fuel to a future time, in effect a “wait and see” position. Evidently the Member States choosing the second option are storing spent fuel pending future developments and decisions regarding the use of nuclear power, which in particular will take into account the economics and security of energy supply, and the resolution of environmental, safety, proliferation and nuclear security concerns. While interim storage cannot be considered itself as a final solution for spent fuel management, it provides time which may enable the development of new technical options [4].

The selection of a strategy for spent fuel management is a complex decision with many factors to be taken into account including politics, economics, resource conservation, environmental protection, and public perception, the last of which has become a predominant factor in many Member States. This is mainly due to the long term implications associated with the minor actinides and fission products, as well as the fissile materials themselves, contained in spent fuel. A good example is the controversy on the issue of retrievability or reversibility of spent fuel after disposal [5]. A project was recently carried out within the 5<sup>th</sup>

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<sup>1</sup> International Project on Innovative Nuclear Reactors and Fuel Cycles.

<sup>2</sup> Michellangelo Network.

<sup>3</sup> Generation IV International Forum.



framework programme of the European Commission on the Comparison of Alternative Waste Management Strategies for Long-Lived Radioactive Wastes (COMPAS) [6].

## 1.2. REPROCESSING OF SPENT FUEL

A major reason for choosing the option of reprocessing has been the efficient utilization of uranium resources. The plutonium recovered by reprocessing can be recycled in LWRs as mixed oxide (MOX) fuel, replacing a nearly equivalent amount of enriched uranium and thus avoiding the need for considerable mining and enrichment operations.

These advantages are not so significant as long as uranium is available at a relatively low price, which has been the case in recent years. In the longer term, however, the recycle of nuclear fuel, including MOX in fast reactors, may play an important role both in global energy supply and as a technical basis for the partitioning and transmutation of minor actinides with a view to reducing environmental stress and contributing towards the sustainable use of nuclear energy. In preparation for such a future possibility, the capability of reprocessing is a desirable option to preserve. An additional consideration is that the value of fissile material recovered from reprocessing may retrieve an economic value at a future date when uranium prices, or other factors influencing total energy supply costs, such as fossil fuel prices, make reprocessing again a competitive market option [7].

However, even with the recycling of MOX fuel or the partitioning and transmutation of minor actinides, plans are still required for the disposal of wastes arising from such operations, since they do not completely eliminate the toxic radionuclides or the other radioactive wastes arising from spent fuel reprocessing [8]. It is therefore essential also to consider technical innovations in future nuclear systems that can significantly enhance the efficiency of radioactive waste management systems, as well as satisfying other criteria.

## 1.3. EMERGING TECHNOLOGIES

In the past few decades, new technical concepts for the management of spent fuel have begun to emerge, aimed at reducing the demands made on environmental resources. In the 1980s the first developments related to technology for fuel rod consolidation to reduce the volume of spent fuel assemblies to be stored [9]. A more recent example is the DUPIC cycle, which has been developed with a view to generating fresh fuel for heavy water reactors (refer to Section 3.3.2. for more details). The concept involves the thermal and mechanical treatment of spent LWR fuel by the Oxidation and Reduction of Oxides (OREOX) process which neither separates fissile materials, nor adds any enriched uranium [10]. A further example is the “melt and dilute” technique designed for the treatment of highly enriched metallic fuel [11].

Another group of technologies that has recently begun to be explored relates to the partitioning and transmutation of actinides and long-lived fission products prior to disposal in order to reduce the volume and the radiotoxicity of the waste going to a final repository. These technologies also require the development of additional facilities to separate the various actinides, and of new reactors to transmute them [12].

The efforts to develop these and other emerging technical concepts has recently been linked with the search for innovative nuclear systems, in the context of long term sustainability, as exemplified by a number of recent international initiatives [13]. One such very recent initiative, by USDOE, called AFCI (Advanced Fuel Cycle Initiative), has as an objective the development of advanced fuel cycles to support advanced fuel utilization and advanced waste

management strategies, including transmutation, and to enable the transition from current fuel cycles to those to be used with innovative reactor systems [14].

#### 1.4. PERSPECTIVES AND CHALLENGES

All of these novel technical concepts are still in the development phase. Hence it will be many decades before they are commercially available. The full potential for industrialization of most of these novel processes will require a full understanding of their technical feasibility throughout the course of their development, and, as an example, this is being specifically addressed in the AFCI. Apart from the technical development and scale-up required, all the other issues relating to the industrialization of the emerging technologies need to be addressed, such as licensing, socio-political issues, and, most of all, the decision as to whether to invest in them commercially [15]. In terms of these criteria, the advanced fuel cycle concepts, especially those involved in partitioning and transmutation, have encountered some criticism [16].

Conventional reprocessing technology will continue to play an important role as an option for spent fuel management. As technical requirements change with time, however, efforts to enhance existing technology and to apply advanced technologies will continue [17]. Several national or international initiatives have also been launched in preparation for technical innovation the next generation, looking forward beyond the current horizon of technical evolution (see 3.2).

#### 1.5. STRUCTURE OF REPORT

This report provides an overview of the status of reprocessing technology and its future prospects in terms of various criteria in Section 2. Section 3 provides a review of emerging technologies which have been attracting the interest of Member States, especially in the international initiatives for future development of innovative nuclear systems. A historical review of IAEA activities associated with spent fuel reprocessing, traceable back to the mid-1970s, is provided in Section 4, and conclusions in Section 5. A list of references is provided at the end the main text for readers interested in further information on the related topics.

Annex I summarizes the current status of reprocessing facilities around the world, including the civil operational statistics of Purex-based plants, progress with decommissioning and diagrammatic representations of the management of spent fuel arisings and of the Purex process itself. Annex II comprises country reports collected from Member States which undertake reprocessing.

## 2. REVIEW OF STATUS

### 2.1. HISTORICAL EVOLUTION

Civil reprocessing has been carried out on a commercial scale for over four decades in several countries. Today all commercial reprocessing plants are recovering material from civil nuclear reactors for recycle and the conversion of unwanted wastes into a safe form for disposal.

Irradiated nuclear fuel was first processed in the 1940s using precipitation processes to separate extremely pure plutonium for military use. Precipitation was soon displaced by solvent extraction, which is better suited to continuous, large scale, remote operation and can facilitate a 3-way separation of uranium, plutonium and fission products.

Several solvent extraction systems were explored before an efficient extraction system was identified. The combination known generically as Purex, which utilized the extractant tributyl phosphate (TBP) mixed in a largely inert hydrocarbon solvent, soon replaced all earlier solvent extraction media. The Purex process has a number of advantages including lower solvent volatility and flammability, higher chemical and radiation stability of the solvent and lower operating costs. Since the opening of the first Purex plant at Savannah River in 1954, the Purex process has been utilised in a variety of flowsheets and is still being used in all commercial reprocessing plants currently operating [18]. While the Purex technology is applicable to a various types of spent fuel, industrial applications have mainly been implemented for reprocessing of spent fuel arising from gas cooled reactors and later from LWR reactors [19].

During the 1960s, civil reprocessing of spent nuclear fuel based on the Purex process was established as a commercial business [20]. Following the opening of the first reprocessing plant, UP1, at Marcoule in France, several other reprocessing plants commenced operation during the 1960s and 1970s in Belgium, France, Germany, India, Japan, The Russian Federation, the UK and the USA. For various reasons, however, only some of these plants are still in operation now, others having been shutdown - some after only brief periods of operation (see Table 1 in Annex I).

Reprocessing using the Purex process has become a mature technology with considerable experience gained from the operation of civil reprocessing plants in several countries handling a wide variety of fuel types (see Table 1 in Annex I). As of the end of 2003, more than 89000 tHM of commercial spent fuel has been reprocessed, mostly at the two commercial plants at La Hague and Sellafield (see Table 2 in Annex I). At the present time the nominal total reprocessing capacity available is about 5000 tHM per annum. Activities range from the small scale reprocessing of fuel from research or experimental reactors to large-scale industrial plants offering an international service for standard oxide LWR, VVER and AGR fuel. The total reprocessing capacity will increase with the new plants Rokkasho-mura of 800 tHM/y nominal capacity, currently under commissioning test in Japan, which is expected to come on line soon.

The current status of industrial reprocessing is described in more detail below with particular reference to the issues that are expected to remain or become important in the future (see 2.3.2).

## 2.2. REVIEW OF TECHNOLOGY

The Purex process adopted by all industrial reprocessing plants is diagrammatically illustrated in Figure 2 of Annex II.

The technology utilized in spent fuel reprocessing facilities has improved rapidly to continuously adapt to the evolving characteristics of spent fuel and other constraints including regulatory requirements [21]. Fuel claddings are today made of harder alloys, fuel compositions have changed from metal to oxide matrix and fuel initial enrichments and burn-ups have been increased. In addition economic conditions have changed, national and international regulations are more stringent regarding safety and security while waste releases and public dose exposure limits have been lowered.

### 2.2.1. Process technologies

Commercial reprocessing relies on a series of four main technological operations: fuel handling and shearing, fuel dissolution, materials separation and purification, and finally, waste treatment and conditioning.

#### 1) Fuel unloading and shearing

There are two technologies available for unloading of spent fuel assemblies from casks:

- Wet unloading, which is well adapted for all types and sizes of casks. The wet method is reliable and flexible, but, as the casks are in contact with water, it generates larger volumes of effluents.
- Dry unloading reduces the volume of liquid effluents produced, but is less flexible.

The introduction of automated operations, for both wet and dry operations, has decreased the workers' radiological exposure [22].

Upon unloading, spent fuel is stored in pools. The main improvements, in this area, have been achieved in the purification of pool water and the optimization of operation and maintenance for the handling and storage systems. As a result of these improvements the amount of waste generated has been reduced and worker exposure reduced.

The removal of metal structures from the fuel assemblies can be performed either by chemical or mechanical processes. The process selected usually depends upon the nature of the fuel and the fuel cladding. Selective chemical dissolution of the cladding was abandoned due to the amount of high level liquid waste generated. Mechanical techniques have now been developed for use in the reprocessing of all types of fuel. For example:

- The relatively soft cladding on Magnox fuel is removed by ramming the fuel rods through a set of slit wheels and a die.
- LWR fuel pins are usually cut into short pieces with a shearing machine so that the fuel can be leached out of the cladding pieces.

#### 2) Dissolution

Dissolution is performed in a geometrically safe dissolver filled with boiling concentrated nitric acid. Dissolution can be undertaken either using a continuous process, e.g. in a rotary dissolver, or by a batch process. The batch process may be preferred over a continuous process in the case of relatively low throughput plants, because it requires less mechanical apparatus. For higher throughput plants, however, a continuous dissolver can give a technical advantage despite its mechanical complexities. In both cases nitrous vapors are recycled. By dissolution of the fuel pellets, most of the volatile and some semi-volatile fission gases are released. Gaseous effluent treatment processes have been developed to remove the major radioactive and hazardous components. Any gases released are carefully monitored to ensure their environmental impact is acceptable.

After dissolution the resultant solution is clarified using desorbers and centrifuges to remove insoluble fission products and cladding fines. Liquor clarification has been improved to increase the plant output and to reduce waste quantities and releases.

### 3) Materials separation and purification

Different types of devices may be used during the extraction cycles to realize the mixture and the separation of phases: mixer settlers, pulsed columns, and centrifuges. Mixer settlers have proved their efficiency as well as their flexibility. They are easy to design, scale-up and operate but occupy a large volume, and hence criticality control is difficult and residence times are long. Pulsed column technologies have been introduced, alongside centrifuges, to accelerate the separation process thus significantly increasing process efficiency. Pulsed columns are more difficult to design, but have a smaller volume to surface area ratio and so criticality control is easier. Further acceleration of the speed of separation has been achieved by the introduction of centrifugal contactors.

All these devices are used in today's reprocessing plants. The operators are optimizing the use of these various devices to achieve greater purity of the end products, to simplify their maintenance and to reduce the amount of effluents generated.

### 4) Waste treatment and conditioning

The aim of waste treatment and conditioning is to confine safely all the radionuclides in a non-leachable solid matrix. At present reprocessing produces two types of waste:

- Waste from the process itself in the form of a liquid solution of fission products and actinides; and
- Waste comprising hulls and end fittings from the structure of the fuel, insoluble fission products from the clarification of the highly active feed liquor, technological waste from maintenance operations and, if warranted, waste coming from the treatment of process effluents.

The reference strategy for the management of the first type of waste, classified as high level waste (HLW), is concentration of the liquid solution followed by vitrification, above ground interim storage of the resulting vitrified waste product and eventual deep geological disposal. The vitrified waste product has been internationally recognized and licensed as an acceptable waste form for the last 20 years. This process has proved to be highly flexible, since fine particles from the dissolution step and alkaline effluents from the solvent regeneration steps have additionally been incorporated into the glass matrix.

There are some differences in the approaches being used for the second type of waste, classified as intermediate level waste (ILW). The UK encapsulates uncompacted ILW in cement, whereas France is placing compacted hulls and end fittings into steel containers for return to customers. In The Russian Federation and Japan, uncompacted ILW is stored in canisters.

#### 2.2.2. Plant engineering

The design, construction and maintenance of operations at a reprocessing plant are complex and require not only well-demonstrated process technologies but also extensive engineering knowledge. In addition to the general requirements applicable to other chemical industries, there are some technical features that are unique to radiochemical plants which require special engineering standards and maintenance skills [23].

The body of engineering knowledge required for the current reprocessing plants has been established by application of the extensive experience accumulated over several decades.

Despite this knowledge and experience, the commissioning period for a new reprocessing plant can still extend over several years to enable any technical difficulties to be overcome as the plant throughput is gradually built up to the design limit. In the future the continuous evolution of reprocessing technologies and changes in the market will each bring new engineering challenges to be resolved both for the expansion of existing plants and for new build.

## 1) Plant design

There are several major factors affecting the design of a reprocessing plant, including safety, technical constraints, and economics. A fundamental legal requirement applicable to the design of any radioactive plant is the protection of workers and public from ionizing radiation sources in the plant. Both the layout of the plant site as well as that of the design of facilities are based on a consideration of the ALARA (as low as reasonably achievable) principle with risk management as well as environmental protection [24].

The multiple barrier concept is generally adopted to ensure radiological safety in the design of commercial reprocessing plants according to regulatory standards (see 2.2.3). On the basis of this concept, the radioactive materials are retained in vessels or pipes which function as the primary barrier. These vessels and pipes along with the radioactive process equipment are housed in shielded ('hot') cells which provide another containment barrier. Normal operations and remote maintenance work are usually performed in an operating area surrounding the hot cells. Access to this area is controlled and monitored. The building layout encompassing the process hot cells is segregated in distinct zones for radiological control of the facility.

The ventilation systems utilized in reprocessing plants are designed to maintain negative pressure in the higher activity zones relative to lower ones. A similar principle is applied to waste discharge systems.

An additional consideration in the design of a reprocessing plant, which is important for its operation and maintenance, is the provision of access routes. This is a complex architectural issue due to the existence of the various radiation zones contained within the plant. In addition to providing controlled access for maintenance work, adequate access will also be needed for construction, inspection, and testing.

## 2) Reliability of equipment

One of the key factors affecting the economics of spent fuel reprocessing is the availability of the plant for continuous operation which in turn is dependent on the reliability of process equipment. As the radioactive components are installed in shielded cells, maintenance of equipment requires time-consuming and costly intervention if something goes wrong.

- Mechanical Performance

The operation of mechanical equipment in the radioactive enclosure is vulnerable to failure. The maintenance and repair of the mechanical failure are often time-consuming and costly, with serious consequences for the availability of the plant. Accordingly the use of moving parts within the radioactive environment is minimized in the design of the equipment. However, since the use of moving parts is unavoidable in the design of some equipment, the capability of enhanced maintenance by remote systems has to be implemented, especially in the head end process.

Occasionally, major liquid processing components become inoperable due to clogging with solid particles such as insoluble residues. Special fluidic devices have been developed to minimize such problems. The reliability, availability and maintainability (RAM) of the major components in reprocessing plants have been significantly improved through the experience gained over the past decades, resulting in the high availability which is achieved today in commercial reprocessing plants.

- Material integrity

Corrosion or erosion of major process components, such as the dissolver or the evaporators (there is more than one evaporator associated with each reprocessing plant), can seriously affect reprocessing plant operation and thus reduce plant availability. Appropriate materials have to be selected according to the requirements of each item of equipment. In addition the integrity of all process equipment in contact with active materials has to be ensured by quality control during manufacturing, installation, inspection and testing, in order to minimize maintenance requirements and plant downtime.

Stainless steel is the standard material used in the construction of the majority of the process systems, with special materials such as titanium utilized for particularly demanding applications. All materials to be used in hot cells are subject to checks for reliability in a radiation environment. Radiation sensitive items are either located outside hot cells or locally shielded to minimize radiation effects.

Significant progress has been achieved in the development of suitable materials. However, more reliable materials are needed and R&D efforts are continuing with a view to enhancing the qualities of materials used in modern plants [25].

- Redundancy provision

Some processes in highly active sections of the plant which are particularly prone to failure may be duplicated in the plant design. This provides standby capacity for use when the original process line fails, which in turn allows more flexibility in maintenance and thereby enhances plant availability. However, such redundancy requires additional investment which may in reality be superfluous if equipment performs well.

### 3) Maintenance

Regardless of the reliability of equipment and materials, maintenance is a critical factor in enhancing the availability of radioactive facilities. Provision for maintenance is therefore an important consideration in the plant design with considerable impact on the choice of equipment and the layout of facilities. If uninterrupted operation of the plant is to be achieved, it is necessary to be fully prepared at all times to replace or repair equipment or components when they fail. Examples include having temporary replacement equipment available for important processes and being fully equipped with stand-by components. Standardization of component parts or of entire items of equipment, and even the use of off-the-shelf components, are important considerations to facilitate cost effective maintenance.

Most operational spaces are equipped with remote systems for intervention and maintenance operations if required. In addition to the basic systems like cranes and hoists and heavy duty and mechanical manipulators, dexterous robotic systems are used in special circumstances.

The design of the maintenance systems has to take into account their own maintenance in case of failure. Highly optimized maintenance operations have been developed at commercial plants through years of experience [26].

#### 4) Process measurement and control

The ability to diagnose the condition of the plant quickly is important for many reasons, such as plant control, safety, safeguards, security and operational effectiveness. Measurement and control systems, which can report precise and immediate plant parameters, are a key feature required by modern plants. It is also essential to have good sampling and analytical methods for process and quality control.

The experience in the commercial plants with process control and instrumentation over several decades of plant design and operation, together with continuous enhancement, should provide a valuable background for the design of future generations of plants [27].

#### 2.2.3. Plant safety

The principal safety objective at reprocessing facilities is the protection of operators, members of the general public and the environment against the deleterious effects of radiation by avoiding direct exposure to intense sources of radiation, preventing the spread of radioactive contamination and strictly limiting the release of radioactive materials. All fuel cycle facilities apply the concept of multiple-component protection to maintain safety and a system of successive physical barriers are used to prevent the spread into the environment of ionizing radiation, nuclear materials and radioactive substances. In addition systems of technical and organizational arrangements are employed to protect operators, the general public and the environment [28].

Reprocessing plants, and fuel cycle facilities generally, differ from reactors in several important respects, although there are some common factors. In the case of fuel cycle facilities, fissile materials and waste are handled, processed, treated and stored in easily dispersible forms throughout the entire plant using chemicals which can be toxic, corrosive or combustible. To ensure that they are safely operated, fuel cycle facilities rely to a greater extent on operator intervention and administrative controls than reactors which tend to rely more on active and passive engineered controls. Safety features based on the defence-in-depth concept needs to be implemented both in the design of the facility and in its operation. Management of the design process needs to ensure that the structures, systems and components important to safety have the appropriate technical characteristics, specifications, and material properties which are compliant with their safety functions [29].

The potential safety hazards to which fuel cycle facilities are prone include criticality excursions, radiation exposure, chemical reactions, fire and explosion [30].

#### 1) Criticality hazards

Criticality control is integral to nuclear engineering safety for nearly all installations. It is a dominant safety issue for reprocessing plants due to the large amount of fissile materials treated and the presence of water, a moderator, in many part of the plant. The control of the nature, quantity and concentration of fissile materials along the process line, the control of the geometry (dimension and shape) of the equipment used, under all conditions, and the presence



of appropriate neutron absorbers are among the preventative measures which, in combination, can be used to avoid criticality excursions.

## 2) Radiation hazards

Radiation safety is of paramount importance at reprocessing facilities. Operators are protected from the radiation by heavy shielding and containment walls which surround the radioactive sources. Human access to the radioactive areas is restricted except in special cases for maintenance or refurbishment work, which are then conducted under strict provisions and procedures for radiation protection. In modern plants, remote technology and automation are extensively used with the aim of both minimizing doses to the operators and enhancing plant performance.

## 3) Chemical hazards

Reprocessing plants are designed and operated taking fully into account the need to protect workers from the hazards associated with the use of strong acids throughout the process as well as the use of organic solvent at the extraction stage.

## 4) Fire and explosion hazards

Flammable, combustible and explosive materials, such as tributyl phosphate-organic solvent mixtures at the extraction stage and bitumen during the conditioning of radioactive waste, are used in the reprocessing process. The hazards associated with their use are considered at the design stage of reprocessing plants, and in the operational procedures, and all necessary protection measures are taken.

In addition to all the preventative measures in place in reprocessing plants, a range of emergency systems and arrangements are also maintained, and regularly exercised, to ensure that a rapid response can be deployed to any incident and its impact minimized [31].

### 2.2.4. Refurbishment, expansion, and lifetime extension

#### 1) Refurbishment and expansion

Any spent fuel management facility may undergo refurbishment or modification during its lifetime. The need for refurbishments may arise for a number of reasons such as enhancement of the facility's function, safety or economic improvements and changes in regulatory standards. Whatever the motive for the refurbishment, the facility must maintain the necessary safety criteria required for normal operating conditions, anticipated operational occurrences and design basis accident conditions both during the refurbishment and for the remainder of its lifetime.

With the evolution of demand and changes in the technical specifications of customers' fuel, the refurbishment and expansion of reprocessing facility is necessary if plants are to adapt to the changing requirements. From the outset of designing a commercial reprocessing plant it can reasonably be expected that a diverse range of refurbishment or expansion work will be undertaken during its lifetime. The optimization of implementation procedures over the years now means that the replacement of obsolete equipment and the adaptation of processes, while substantially maintaining production throughput, can be achieved more easily. For example,

the capacity of the French UP2 plant was increased from 400 tHM/y to 800 tHM/y between 1992 and 1996, with only minor interruptions to production.

As most refurbishment requires working in areas which are subject to radioactive contamination, careful planning and management of the facility configuration, as well as of the operational safety of the workers, are essential pre-requisites.

## 2) Lifetime extension

Life extension of existing nuclear facilities beyond their originally licensed period has become a key issue to nuclear operators in the world. In the USA, several nuclear utilities have recently renewed their licenses and extended the expected operational life of their nuclear power plants by 20 years. Most other utilities have taken, or are expected to take, action to follow suit.

Similar questions have been raised on the life extension of nuclear fuel cycle facilities including reprocessing plants. Most of the spent fuel management facilities in the world are licensed for 20~40 years depending on the regulatory settings of the country in which they are located. It is possible that the lifetime of reprocessing plant may need to be extended for business or other reasons and therefore need relicensing. This is not expected to present any particular problems in view of the successful relicensing already achieved in relation to refurbishment and expansion work at reprocessing plants.

### 2.2.5. Decommissioning

The final stage in the life cycle of a nuclear facility is decommissioning. With the first generation of reprocessing plants being retired from operation, experience is now being gained in decommissioning of reprocessing facilities. Decommissioning begins immediately following the final closure of a facility and continues to the point of leaving a clear site where the facility once stood. According to generally accepted principles, decommissioning operations comprise three major stages:

- Initial clean-up and preliminary decontamination, where necessary, of plant and related facilities;
- Dismantling and removal of the systems, equipment and pipework within the facilities, with decontamination as appropriate;
- Demolition or reuse (restricted or unrestricted) of buildings and structures.

The time taken to complete these stages, as well as the period between each stage, can vary considerably depending upon the type of facility, the operator and the national policy and regulations. In order to benefit from the skills and experience of operating staff, immediate dismantling and early site release may be favoured. Alternatively, deferral of later stage(s), an option called “safestore”, may be preferred. “Safestore” allows significant reduction in residual radioactivity by natural decay of the radioactive inventory, thus reducing radiation hazard during the eventual dismantling [32].

Typically an industrial reprocessing plant covers a large area and is housed in several buildings. There are a number of challenges associated with the decommissioning of these facilities, including the high radioactivity levels inside certain parts (due to fission products) and the presence of various types of contamination (alpha, beta and gamma emitting radionuclides). Consideration must be given to how the radiological hazard will change with

time, for example because of the decay of Pu-241 to Am-241. In addition the criticality hazard potential should be taken into account while decommissioning and dismantling plant areas which may contain residual plutonium and/or other fissile material. Another major consideration is the hidden presence of alpha emitters in confined areas, such as small diameter pipes, where contamination assessment is difficult (for example due to the difficulties involved in alpha measurements).

The first generation nuclear plants were built with little thought to what would happen to them once they stopped working. Experience gained during their decommissioning has, and will continue, to enable optimization of future decommissioning strategies. Accordingly, today's nuclear plants are designed and built with decommissioning in mind. Some technical information on decommissioning of reprocessing is available in the literature [33].

## 2.3. TRENDS AND ISSUES

Reprocessing has proved its effectiveness for safe and economical spent fuel management. The Purex process has been progressively and continuously improved, and this improvement accounts for the successful commercialization of reprocessing in several countries. Throughout all the stages - design, construction, testing, commissioning, operation and optimization - the operators of the plants have already proven their expertise. At each one of these stages, the feedback from experience is systematically analyzed and transferred to engineering teams in order to develop or adapt new technologies. Notwithstanding the past achievement made by the reprocessing community as described above, there are a number of issues associated with reprocessing operations in the current nuclear industry [34].

The near and medium term challenges for reprocessing are to achieve economic competitiveness through the reduction of the volume and radiotoxicity of the waste destined for ultimate disposal. An additional challenge is the adaptation of current technologies and plants to meet even more stringent national or international regulations and, at the same time, to accommodate fuel performance increases, such as higher fuel burn-ups. The experience already acquired by reprocessing plants operators in criticality control, higher throughputs, lower emissions and working with high level radiation, allows them to be confident about the adaptability of their plants to future market and regulatory changes.

This section reviews some of the main issues and challenges faced by the reprocessing industry in delivering safe, reliable and economic services to their customers.

### 2.3.1. Technical trends

As mentioned above, technology for reprocessing has shown constant evolution in the past decades and will continue to evolve in response to new trends in technical requirements. Some of them are as follows:

#### 1) Spent fuel characteristics

Spent fuel burnups have significantly been increased in the past years, mainly driven by economic reasons, resulting in commensurate impacts in fuel cycle backend including reprocessing. Due to continuous improvements in operations, reprocessing plants are now able to accept fuel with characteristics that are different from those they were originally designed for. For example, a typical LWR fuel reprocessing plant may have been designed for fuel with an initial enrichment around 3 % U-235, a burn-up of 30 GWd/t and cooled for 3

years. Today, the average LWR fuel being reprocessed has an initial enrichment reaching 3.7%, a burn-up that can reach 45 GWd/t, and requires a 4-year minimum cooling period. With the continuing increase in discharge burnup, it is expected that reprocessing plants in the future deal with still higher burnups of spent fuel [35].

This trend towards higher burn-up, and consequently to a higher initial enrichment for fresh fuel, is likely to continue, driven as it is by the desire for yet greater economies in the performance of nuclear power plants. There will be a limit to this trend, due to regulatory constraints, but generally that limit has not yet been reached. This trend has already had implications for the operation of reprocessing plants, but because of the years of operational experience the majority of reprocessing plants have been shown to be technically capable of reprocessing such fuel without any significant changes to their design [36].

There has also been a move, in Europe, at least, towards burning MOX fuel in LWRs. The extent to which this use of MOX fuel will grow is uncertain, dependent upon its comparative economics with UOX fuel and other factors, such as alternative means of disposing of separated plutonium.

Spent MOX fuel produces higher levels of radiation and heat output than spent UOX fuel, and so it presents similar challenges to reprocessing plants as high burn-up UOX. However, although reprocessing of spent MOX fuel has been demonstrated at La Hague, at present there are no plans to recycle spent MOX fuel by reprocessing routinely or on a large scale.

## 2) Logistic issues (storage and transportation)

In preparation for reprocessing of spent fuel, such logistical operations as transportation and buffer storage of spent fuel are essential processes.

- Spent fuel storage

An important consideration for spent fuel reprocessing, or any other option for spent fuel management, is the interim storage of spent fuel. The buffer storage of spent fuel at reprocessing plants plays an important logistical role in the reprocessing operation and occupies a large proportion of plant operational effort and costs. To facilitate technical flexibility for unloading and cooling, most of the spent fuel storage systems at reprocessing plants have used wet storage, but a dry unloading system has also begun to be used to some advantage at the La Hague plant [37].

- Transportation

A vital link to reprocessing is spent fuel transportation. The majority of the spent fuel transportation activities accomplished until now are associated with reprocessing. Several companies have been developed to provide the transportation service, operating a number of spent fuel transportation casks that have been licensed and employed in compliance with national and international regulations. The extensive industrial experience in spent fuel transportation accumulated in the past several decades, including an excellent safety record, will be entirely applicable to spent fuel transportation operations for other activities in the fuel cycle backend, such as interim storage and disposal. In fact, concerns are raised on the future transportation of spent fuel from reactor sites to repository is regarded as a big challenge in countries like the US, for example, because of the sheer quantity and long distance involved [38].

Most of the spent fuel transported from power plants to reprocessing plants has been by sea and/or rail, due to the weight limit on other forms of transport, and other factors. Nevertheless it is interesting to note, in this regard, that long distance (4,000 km) transportation by truck has recently been achieved in China using a dual-purpose cask (NAC-STC) weighing some 130 tons [39].

### 3) Occupational and public exposure

A combination of learning from experience and continuous improvements, modifying both plant and practice, has reduced the average employee radiation exposures at reprocessing facilities from over 10mSv to 1.5mSv per person pa over the past two decades. By comparison, the average annual exposure for airline crew is about 2mSv.

Radiation exposure of the public has also reduced, largely in line with the reductions in radioactive discharges. As the quantity of radioactivity being discharged has declined each year, the proportion of radiation exposure that is attributable to current discharges has declined. In the UK today, the average annual exposure of individuals due to radioactive discharges is less than 0.1mSv. By comparison, the average annual exposure to individuals in the UK from natural background radiation is about 2.2mSv.

### 4) Waste management

Twenty years of continuous operations in industrial reprocessing plants have led to process and waste treatment optimizations. Improved sorting procedures and increased package concentrations have allowed operational waste quantities arising from process and plant maintenance to be reduced. An example is the recent improvement resulting from fuel hulls and end fittings compaction implemented in La Hague where some three to five-fold volume reduction has been achieved.

Development of new technologies is a continuous process. At the present time developments include:

- Improvements in encapsulation matrices
- Improved sorting and categorization of waste
- Engineered transfer systems for handling  $\alpha$ -active waste
- Improvements in the vitrification process.

For example, to further increase the performance of the vitrification line, the cold crucible melter technology that was first developed in the beginning of the 1980s is now being implemented at an industrial scale. Its compact modular design enables significant cost savings on capital investment and operations. Its flexibility will allow the treatment of a range of wastes and its reliability will reduce the accumulation of the failed equipment waste stream along the process line. Another line of progress lies in the standardisation of containers, such that only a single type of waste container requires disposal [40].

#### 2.3.2. Issues associated with spent fuel reprocessing

##### (1) Disposal

The reprocessing of spent fuel should not be considered separately from the disposal of the radioactive waste deriving from the process. Safety and environmental concerns about the

long term radiotoxicity and the level of radioactivity of spent fuel or waste separated during reprocessing has driven some countries to study the feasibility and economics of the partitioning and transmutation of minor actinides and long lived fission products, for example in special “burner” reactors or in accelerators.

## (2) Recycle of plutonium and uranium

One of the major challenges facing the reprocessing industry in the future is the use of the current stockpile of separated plutonium by the prospective MOX market. If demand for MOX fuel continues to match the available fabrication capacity, it is possible that the stockpile of plutonium will be reduced in the future. It is anticipated that the future viability of spent fuel reprocessing will, at least in part, be dependent upon the demand for recycle services in the years to come [41].

Plutonium and reprocessed uranium have been successfully recycled on an industrial scale in thermal reactors for many years. Plutonium is recycled in the form of MOX fuel and experience has shown that the performance of the MOX fuel is similar to that of uranium fuel. The fuel cycle facilities required for the recycle of both plutonium and reprocessed uranium are in operation and some large scale facilities are available to offer recycle services to the international market.

The management of MOX and reprocessed uranium recycle are dependent upon the market status of uranium and enrichment services. Some countries regard reprocessed uranium and plutonium as a strategic reserve and intend to store it for possible use in the future when availability and/or the price of new uranium make their recycling a more attractive option.

## (3) Economics

Assessments of the economics of reprocessing usually compare direct disposal following interim storage against prompt reprocessing and recycle. Among the various studies performed with a view to assessing the economics of spent fuel reprocessing (or recycle of MOX fuel in thermal reactors), a well-known one is the 1994 OECD/NEA fuel cycle study. This study calculated the total fuel cycle costs, including reprocessing, as 6.23 mills/kWh (with a range of 5.17 - 7.06 mills/kWh) and the same costs, but including direct disposal instead of reprocessing, as 5.46 mills/kWh (with a range of 4.28 - 6.30 mills/kWh) [42]. The OECD/NEA report concluded that there was no significant cost difference between the prompt reprocessing and direct disposal options, especially when considered in terms of the underlying cost uncertainties and the national variations in conditions and constraints. The ultimate costs of encapsulation and disposal of spent fuel still have a large degree of uncertainty attached to them as no encapsulation or disposal facility has yet been constructed on a commercial scale. The report further noted that consideration of a much wider range of factors, including national strategy, reactor type, environmental impact, financial situation and public acceptability, will have a greater influence on the selection of a spent fuel management option.

In the years following the OECD/NEA 1994 report, fuel cycle costs have been reduced due to increased competition in the front end market and pressure for cost reductions in the back end. However the general conclusions drawn from the 1994 study are still valid as the assumptions used were by definition generic. A more recent report published by the OECD/NEA, noted that several national studies had been published over the past few years, which have demonstrated economic benefits for both fuel cycle options [43].

It seems that existing reprocessing plants will continue to operate, or not, according to national priorities and commercial decisions which take into account all of the factors identified in the OECD/NEA report, and possibly more. For future reprocessing plants a further uncertainty is introduced as to the nature of such plants and their interaction with the rest of the fuel cycle. These considerations could have a profound effect on the economics of reprocessing, and indeed that is one of the drivers of the extensive development work exploring innovative processes, as reviewed in section 3 of this report. As more is understood about the nature of, and therefore the likely costs associated with, possible future reprocessing plants further economic studies would be appropriate.

The design, construction and commissioning of a reprocessing plant is a financially “high risk” venture because of the associated high capital costs and long commissioning period. The commercial reprocessing plants operating today were underpinned by reprocessing contracts, which stemmed from legal and political national imperatives to reprocess. These circumstances gave rise to favourable cost-plus contracts and the free provision of capital for the construction of commercial reprocessing plants in Europe. Over the past few decades the economics of reprocessing have improved. As with any mature industry, benefits from improved plant efficiencies and technological developments have arisen as a result of the extensive operational experience gained. These tend to result in cost reduction. In addition, as the majority of the current reprocessing plants have been operating for many years, much of the investment has now been amortized. As a consequence, costs have decreased substantially for the large commercial plants (such as UP2/UP3 and Thorp). Against this background, competition in the reprocessing business from investment in new plant is only likely if and when there is a radical change in the market [44].

In the case of other plants, with small capacities or with irregular operation, the economics are less well known, or difficult to analyze, and are likely to be subject to greater uncertainties [45].

#### (4) Environmental impacts

Spent fuel reprocessing plants have been operating at industrial scale for several decades. During this time much knowledge has been accumulated which has resulted in significant improvements in plant safety and radiological protection.

- Radiological discharges

Substantial reductions have been achieved in the radiological discharges from reprocessing sites despite increases in reprocessing throughput. Through investment in new waste management facilities and process optimization, radioactive discharges from the Sellafield and La Hague sites have been reduced to a very small fraction of their peak levels in the 1980s.

At current levels of discharge no risk of harm to man or the environment has been proven to date. However, the industry is still under pressure to further reduce discharges from reprocessing facilities. One specific example is the OSPAR strategy which affects the two major reprocessing sites, Sellafield and La Hague. The OSPAR Strategy, as agreed by the OSPAR (Oslo-Paris) Commission meeting in Sintra, 20-24 July 1998, sets the specific objective of preventing pollution of the maritime North Sea area from ionising radiation. It was agreed that this should be achieved by 2020 through progressive and substantial reductions of discharges, emissions and losses of radioactive substances, with the ultimate

aim of concentrations in the environment being near background values for naturally occurring radioactive substances and close to zero for artificial radioactive substances [46].

The industry has agreed to this strategy provided it is based on a sound scientific approach. It also notes that it is important in considering the concept of sustainability that a global view is taken on the total radiological impact of various spent fuel management options and not just concentrating on particular narrow and local sectoral issues. For example, it is important to compare doses resulting from discharges across the world rather than simply the dose from marine discharges to one limited geographical area (the North East Atlantic).

The implementation of the objective clearly articulated in the Sintra Statement will be challenging but achievable. Current European reprocessing plants are operating safely and can meet these OSPAR obligations.

With technological improvements the reprocessing option can produce lower volumes of radioactive waste compared with those associated with direct disposal. In addition, reprocessing and recycle can contribute up to about 30% saving in natural uranium requirements and, hence, in the mining wastes generated. Overall there is little or no difference in environmental impact expected between the recycle and once through fuel cycles [47], [48].

- Return of radioactive waste

The successful return of the wastes (and products) arising from such reprocessing has been demonstrated, with the waste forms and packaging conforming to specifications agreed with the safety authority in the countries where the fuel originated.

At the present time international reprocessing services are offered by UK, French and Russian companies. International reprocessing contracts have required the satisfactory resolution of several issues such as the transport of fuel and the return of products and wastes to the countries of origin. These contracts have been subject to the full scope of international safeguards [67].

## (5) International standards

Safety requirements for reprocessing plants are reflected at national level in regulations and standards. However, there is a trend toward internationalization of safety standards for the nuclear fuel cycle in general and spent fuel management facilities in particular. This issue has been examined at the IAEA and a system of international safety standards for fuel cycle facilities is in development. A safety guide on spent fuel reprocessing facilities is also in preparation [51].

Meanwhile, a Joint Convention on the Safety of Spent Fuel Management and the Safety of Radioactive Waste Management has been agreed which entered into effect on 18 June 2001 [51]. The Joint Convention is the first international treaty relating to these areas of safety which is legally binding. It represents a commitment by States to achieve and maintain a high level of safety in the management of spent fuel and radioactive waste. The first Review Meeting of the Joint Convention was held in November 2003 [52].



Another example of an initiative to establish international standards is the development by the European Union of directives on nuclear safety and radioactive waste management. This work is still underway and is drawing on the experience of the IAEA precedents in this area [52].

#### (6) Non-proliferation and safeguards

There have been many debates on such issues such as non-proliferation, safety, economics and environmental impact associated with nuclear energy and its fuel cycles. One critical issue for reprocessing has been the risk of diversion of separated uranium and plutonium and their possible misuse for non-peaceful ends. In fact, one of the main reasons for the move towards a direct disposal policy for spent fuel management, which began in the 1970s, was concern over proliferation. Subsequently, various socio-economic concerns also influenced the move away from the reprocessing and recycle of spent fuel.

The proliferation controversy as it related to Purex-based reprocessing culminated in the latter half of the 1970s with extensive debates during the International Nuclear Fuel Cycle Evaluation (INFCE) which examined various fuel cycle concepts which might be able to mitigate the proliferation concern [53]. Although the discussions from the INFCE concluded there is no technical 'fix' to the proliferation problem, it did provide an opportunity for international recognition of some technical features that could be complementary to institutional factors. With the recent revival of nuclear issues, technical methods for deterrence to nuclear proliferation have been being revisited.

- Proliferation resistance

The technical attributes inherent in the nuclear systems and fuel cycles that increase barriers to diversion activities are regarded as intrinsic resistance in contrast to extrinsic resistance which is more artificial or institutional. Some of the major intrinsic measures examined during the INFCE exercise include such measures as ; minimizing presence of sensitive materials, reducing accessibility to sensitive materials, denaturing of the sensitive materials, enhancement of accountability or surveillance, etc. These criteria can be considered in the design of new facilities or refurbishment of operating facilities for enhancement of proliferation resistance [54].

The requirement to be proliferation resistant is one of the major criteria for innovative nuclear fuel cycles being studied in such international initiatives as INPRO, under the auspices of the IAEA. In particular, a case study in the frame of INPRO has been being conducted for application of proliferation resistance criterion to the DUPIC fuel cycle. in order to assess the proliferation resistance characteristics of the DUPIC fuel cycle using the Revised INPRO Methodology, and to develop recommendations on the further improvement of the INPRO Methodology and in the area of the proliferation resistance for the application to evaluation of innovative nuclear energy systems [57].

- Safeguarding of reprocessing plants

The objective of safeguarding is to ensure that nuclear materials are not diverted from peaceful utilization to the production of nuclear explosive devices. Safeguarding policies have been in place for more than 30 years with IAEA safeguards under the Non-Proliferation Treaty (NPT) for most nuclear material facilities in the world. Large bulk-handling facilities such as reprocessing plants are a challenge in terms of safeguards. They

are subject to extensive measures for material accountability through continuous inspection at key measurement points (KMPs), complemented by containment and surveillance verifying all transfers of nuclear materials into and out of the facilities [58].

The verification requirements of a commercial reprocessing plant call for more than 750 man-years of inspection effort compared to 6~12 for a standard LWR. No other types of nuclear facility have been the subject of as deep and as intensive safeguard studies as reprocessing plants [58]. One of the major issues which has attracted attention is the uncertainty involved in material unaccounted for (MUF) attributable to the statistical accuracy of measurements in facilities handling large quantities of material [59]. These studies were started in the late 1970s and culminated, between 1988 and 1992, in the so-called LASCAR project [60]. This study, conducted by the IAEA with direct involvement of the United States, Euratom and the principal reprocessing countries, concluded that large-scale reprocessing facilities can be safeguarded through a combination of various existing techniques such as near real time accounting (NRTA), the choice among which is largely plant-specific [61].

Reprocessing facilities are designed and constructed to satisfy all relevant national and international standards for the safekeeping of nuclear materials. The key to achieving safeguarding is accountability and transparency, resulting from communications and co-operation between the operator and safeguard authorities. Today, specific approaches are implemented in reprocessing plants, tailored to take maximum benefit from the features of these plants, such as automation, computerised systems and remote monitoring. Based on the past experience at La Hague and Sellafield, new designs can incorporate smaller material balance zones and can utilize improved technology for the measurement of processes [62]. The new reprocessing plant being commissioned at Rokkasho-Mura in Japan is an interesting case in point, with implementation of extensive safeguards measures largely automated to such extent that three-quarters of data collection are reported to be performed unattended [64].

- Institutional transparency

International reprocessing contracts implemented under bilateral agreements provide an additional degree of transparency. Plant operators must be able to demonstrate to their customers, as well as to their customers' Governments and regulatory authorities, that they are able to account fully for all an individual customer's nuclear material and that they are storing it safely and securely. Plant operators are also providing inspectors with increased access to facilities. There has never been any material diverted from civil reprocessing conducted under international contracts and operated under international safeguards.

#### (7) Physical protection

Reprocessing plants are constructed to extremely robust engineering standards and incorporate large quantities of reinforced concrete as an integral part of construction. In addition, specific physical protection measures are provided to protect material and facilities against theft and sabotage.

In addition to the prevention measures, emergency plans are maintained at reprocessing plants to ensure appropriate response to any malicious act and are exercised routinely.

## (8) Internationalization

There have been some initiatives in the past more or less successful on international co-operation in nuclear fuel cycle services, such as the Eurochemic reprocessing facility, operated under the aegis of Euratom, and the uranium enrichment enterprises like Urenco and Eurodif [64]. A series of studies conducted in the later 1970s, including the study on Regional Fuel Cycle Centres by the International Nuclear Fuel Cycle Evaluation (INFCE) and the Expert Group on International Spent Fuel Management (EG-ISFM), looked at the feasibility of international management of nuclear materials with a focus on non-proliferation [65], [66], [67]. However, most of those initiatives for multinational management of nuclear fuel cycles have not been successful for one reason or another, up to now [65],[69]. The revelation of continuing proliferation of nuclear technologies and materials have lead the Director General of IAEA to call for a revisit to the idea of multinational approach [MNA] fuel cycle management, The study group issued a report on the possible options for MNA which was circulated for discussion among the IAEA Member States [66].

As the market consolidates and greater economies of scale are sought, new or replacement fuel cycle facilities may well be constructed as regional or international joint venture facilities. Internationalization of fuel cycle facilities can offer a service on a cost competitive basis, especially for countries with small nuclear programmes. Consensus is building that facilities operating on an international basis can provide a higher degree of confidence in the implementation of nuclear material safeguard and security measures [66].

International co-operation is also an important factor in achieving energy policies consistent with global sustainable development. However short term national or local political pressures can adversely impact the implementation of these facilities. International co-operative frameworks have already been established in the nuclear energy field covering R&D, regulations and legal aspects, exchange of information, technology transfer and material trade.

## 3. EMERGING TECHNOLOGIES

Over the past decade, a renewed interest has been given to some innovative technologies for spent fuel treatment which have been emerging as ‘alternative methods’ to the conventional Purex processes. Those alternative methods making use of dry processes had previously been studied and developed in an earlier period of nuclear energy development as fuel cycle systems for fast neutron reactors. Much of such alternative R&D efforts have not been extended to industrialization, however, because the mature conventional PUREX technology was available without much additional R&D to FBR fuel processing on the one hand ,and the gradual failure of commercialization of FBR itself in the eighties on the other. Despite those circumstances, some alternative methods to the wet technology have been looked for since decades with sometimes interesting results that may deserve further attentions to develop advanced systems by dry technologies, among others :

- IFR<sup>4</sup> concept for metallic fuel recycle in the U.S.( cancelled in 1994, and now used for spent fuel treatment addressed to spent fuel disposal)

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<sup>4</sup> Integral fast reactors.

Vibropac method for oxide fuel recycle by similar treatment to the IFR in The Russian Federation ( now being planned for a test with Pu material from weapon disposition)

Advanced Fuel Recycle System concept being refined in Japan (with a view to long -term applications)

DUPIC concept being tested in cooperation between ROK-Canada-USA ( for recycle of spent L WR fuel into HWR without major refurbishment of CANDU-type power plants

Application of dry technologies could become an important new trend in some part of the backend fuel cycle area. With the recent interest in the technical possibility of transmuting actinides, some pyrochemical methods of spent fuel treatment have been revisited at some R&D centers . The dry technology has also finding its way in some complementary processes of the conventional wet technologies. The wet decontamination in the conventioanal fuel fabrication plant has now been largely replaced with dry methods to reduce liquid waste and subsequent treatment processes.

It should be noted that the definition of ‘treatment’ in a technical sense of the word applies to an extensive range of processes for spent fuel management which would follow interim storage of spent fuel. However, the term has often in the past been equated to the reprocessing option for spent fuel management.

### 3.1. BACKGROUND

For many years there has been a continuing search for alternative nuclear systems that could mitigate the adverse issues associated with current nuclear reactors. For example, the INFCE in the 1970s was an exercise to examine the proliferation risk of conventional fuel cycle concepts and to identify possible technical alternatives. The Chernobyl accident in the 1980s heightened public concern calling for reinforced efforts to improve nuclear safety, while the debate on climate change in the 1990s has drawn attention to the need to reduce carbon emissions from energy production.

It is becoming more evident that nuclear energy can be an alternative to fossil fuel use in order to avoid carbon emissions and to preserve dwindling natural resources. For these reasons, there are currently a number of Member States wishing to maintain or expand their use of nuclear power in the future. However, the future of nuclear power as an energy source is dependent upon innovative features that would mitigate the concerns witnessed in the past and be competitive in the deregulated energy market. The rationale defined for innovative technology is as follows [49]:

*“The long term outlook for nuclear energy should be considered in the broader perspective of future energy needs and environmental impacts. In order for nuclear energy to play a meaningful role in the global energy supply in the foreseeable future, innovative approaches will be required to address concerns about economic competitiveness, safety, waste and potential proliferation risks”.*

Today there are a number of innovative nuclear reactor systems under investigation by various private or public organizations worldwide with a view to looking at their feasibility to meet the future challenges of nuclear energy. Several international collaborative initiatives, such as Gen IV and INPRO, have been established to develop novel reactor systems which are expected to commence commercial operation around 2030. Some of the proposals do incorporate reprocessing and as a result a number of independent and collaborative research

programmes are now in progress to improve existing reprocessing technologies and to develop new methods for the treatment of spent nuclear fuel. Many countries, including the European Union, the United States, Japan, and the other reprocessing countries, are involved in the development of advanced aqueous processes for the recovery and separation of minor actinides. Work is also being done on improvements to the basic Purex process to reduce costs and minimize waste generation. Two expert groups chartered by the Nuclear Energy Agency of the Organization for Economic Co-operation and Development are assessing the state of the art of aqueous and pyrochemical processes for the partitioning of spent fuel and will report their findings in the near future [72], [74].

### 3.2. REQUIREMENTS FOR INNOVATIVE TECHNOLOGIES

Regardless of whether development is based on improvements of existing technology or the implementation of new technologies there are a number of common objectives. In the case of INPRO, the Terms of Reference defines the objectives of the short term as [73]:

- to help to ensure that nuclear energy is available to contribute in fulfilling, in a sustainable manner, energy needs in the 21st century;
- to bring together all interested Member States, both technology holders and technology users, to consider jointly the international and national actions required to achieve desired innovations in nuclear reactors and fuel cycles that use sound and economically competitive technology, and are based – to the extent possible – on systems with inherent safety features and minimize the risk of proliferation and the impact on the environment;
- to create a process that involves all relevant stake holders that will have an impact on, draw from, and complement both the activities of existing institutions and ongoing initiatives at national and international levels.

Some of these objectives do not necessarily complement one another and a compromise between them must be found. For example, improvements in environmental impact often have a negative impact on the overall economics of the fuel cycle [76].

#### 3.2.1. Resources management and economics

##### 1) Conservation of natural resources

At present only uranium and plutonium are recovered as products of reprocessing. Other fissile nuclides, such as americium and neptunium, are included in high-level radioactive liquid waste and immobilized for geologic disposal. New reprocessing technologies are being developed to recover other fissile nuclides with the intention of using them as fuel material.

##### 2) Improvement of fuel cycle economics

Despite the fact that reprocessing comprises only a fraction of the overall fuel cycle cost, there is an incentive to make reprocessing simpler and less expensive. The benefit gained by recycling is directly linked to the cost of natural uranium. As long as the cost of natural uranium remains at the present level, there is even greater incentive to reduce the costs of reprocessing in order to maximize the benefit of recycling. In this sense several candidate novel technologies have recently been proposed to enhance and perhaps eventually replace the traditional Purex process [75].

### 3.2.2. Environmental and waste management

#### 1) Reduction of environmental impact

In electric power generation and associated fuel cycles, the discharge of chemical (for example, carbon, sulphur and nitrogen oxides) and radiochemical (for example, iodine radioisotopes) pollutants is a sensitive issue. Even if the absence of adverse health effects has been proven, public acceptance dictates that the amount of *any* radioactive discharge be further lowered.

#### 2) Optimization of waste management

The difficulty of realizing final disposal of high level radioactive waste is partly due to the fact that it contains slowly decaying radionuclides with half-lives extending over millions of years, far beyond the imagination of the public. Separation of such radiotoxic nuclides from high level radioactive waste would contribute to wider public acceptance and further rationalization of disposal facility design. The separated radiotoxic nuclides could then be recycled in a nuclear reactor and converted into radionuclides with much shorter half-lives.

### 3.2.3. Safety

#### 1) Plant safety

New reprocessing technologies will be subject to international and country-specific safety standards. Furthermore, any organization with responsibility for design and construction of a reprocessing facility will be encouraged to achieve a higher level of safety than that required by regulatory authorities.

#### 2) Exposure to radiation

All reasonable measures will be taken to minimize exposure to radiation for both public and plant operators for any new design of a reprocessing facility.

### 3.2.4. Proliferation resistance and safeguards

Over the years, the proliferation issue has become one of the key considerations in R&D efforts on new fuel cycle concepts. An example of such an exercise is the DUPIC (Direct Use of Spent PWR fuel in CANDU reactors) concept which is being developed with a view to enhancing some fuel cycle features, including proliferation resistance. The proliferation concern is also a high priority criterion in such international projects as Gen IV (USA) and INPRO (IAEA) being initiated with a view to developing sustainable nuclear technologies (*refer to 3.2.4 above*).

#### 1) Proliferation resistance

The separation of high purity plutonium is commonly viewed as the major potential proliferation risk in reprocessing plants, if control over special nuclear materials is lost or if such plants were to be operated in the wrong hands. New recycling technologies are being designed with a degree of added resistance to proliferation by incorporating inherent features that make it more difficult to separate pure plutonium.

## 2) Safeguards

Any new reprocessing facility will have to incorporate practicable means to meet international standards for control of nuclear material. This institutional aspect is included as a basic element in the INPRO requirements as an extrinsic feature of proliferation resistance.

### 3.2.5. Public acceptance/involvement

Any new reprocessing plant will not be constructed without the agreement of local government and the public. Gaining acceptance will be especially difficult unless the public can be shown the benefits of reprocessing and the safety of the plant in a convincing and easily understandable way.

In many countries, nuclear affairs used to be enshrined in governmental control and much part of the decisions relied on technical experts. Such practices in the past have engendered problems in the communication with some stakeholders including in particular the general public and affected localities. From a recognition of the societal significance of environmental affairs, an increasing number of countries have taken political measures to enhance transparency and public involvement in addressing the issues. It is likely that this trend spread with global trend toward democratization in the future [77].

The issue of public participation has also been addressed in some international conventions like the Aarhus Convention which was initiated in 1998 with a view to provide access to information, public participation in decision-making, and access to justice in environmental matters [79].

## 3.3. REVIEW OF EMERGING TECHNOLOGIES

There are many incentives for the improvement of reprocessing technologies. As long as uranium ore prices and enrichment costs are low, the value of reprocessing may well reside in facilitating the disposal of high level nuclear waste. Technologies for partitioning and transmutation (P&T) or partitioning and conditioning (P&C) can greatly reduce the hazard of high level waste, by eliminating the longer term, most toxic radionuclides or by placing the waste in a much more durable form for long term storage.

In the near term, the technologically mature aqueous processing methods constitute the main path forward, capitalizing on the success of the well established Purex process, while dry processes are considered as adjunct or backup processes. In the longer term, however, fuel cycle applications related to advanced reactor concepts (liquid metal fast reactors, gas cooled reactors, molten salt reactors, etc) may favour the use of pyrochemical processes, as do accelerator-driven systems (ADS) designed for the destruction of highly radiotoxic minor actinides.

### 3.3.1 Aqueous technologies

Several main goals are being pursued in the further development of aqueous reprocessing technologies:

- minimizing the overall volume and/or activity of the waste to be disposed,
- recovering the long-lived radionuclides for either specific disposal or transmutation, and
- enhancement of proliferation resistance

Major radionuclides to be considered here are the minor actinides (Np, Am and Cm), long-lived fission products (<sup>129</sup>I and <sup>99</sup>Tc), and also, in some cases, the short term heat-generating isotopes such as Cs-137 and Sr-90.

## 1) Enhancement of today's technology

Enhancements to existing processes are generally supported by industrial operators and deal with all steps of the reprocessing operations. One example of a novel decladding and dissolution process is the development of direct dissolution of LWR fuel and cladding by contact electrolysis by British Nuclear Fuels Ltd (BNFL). Dissolution of powdered fuel is also attracting some interest. Dissolution of powdered fuel has two rationales. It can increase the dissolution rate, which means that large scale dissolution can be achieved without serious problem, for example, from criticality safety (provided that issues such as the handling of powdered fuel and the development of sufficiently efficient off-gas treatment systems can be overcome). In addition, a more concentrated heavy metal solution can be produced in the dissolver, which facilitates the crystallization of uranium at temperatures above 0°C.

Considerable work is occurring to enhance the efficiency of material separation and purification. The addition of a crystallization step for plutonium separation is being developed by Japan Nuclear Cycle Development Institute (JNC). Direct conversion of the nitrate product to UO<sub>2</sub>, PuO<sub>2</sub>, and/or (U, Pu) O<sub>2</sub> is also being investigated, using direct thermal denitration (BNFL, JNC), plasma chemistry (Khlopin Institute), or ammonia co-precipitation (Bochvar Institute).

Among the many ideas under consideration to minimize the volume of secondary waste, the use of salt-free reagents has been widely investigated. An example is the study, by the Commissariat à l'Énergie Atomique (CEA, France), of the catalytically-mediated denitration of highly concentrated nitric acid solutions.

## 2) Actinide separation technologies

It is in this field where most of the effort is currently engaged, especially on the issue of minor actinide recovery. The main processes under investigation can be classified into two categories: one-step and two-step processes.

- One-step processes

Three liquid-liquid extraction processes are under development to recover the trivalent actinides (Am, Cm) in only one step: one in France (the PALADIN process of CEA), and two in Japan (the DIDPA process of JAERI and the SETFICS process of JNC). They are all based on the same principle: a selected extractant (DIDPA<sup>5</sup> for JAERI, CMPO<sup>6</sup> for JNC and a synergetic mixture of malonamide and HDEHP<sup>7</sup> for CEA) loads all the actinides and lanthanides in an organic phase. Transuranic elements are then successively stripped from the solvent by complexation with a carboxylic acid (DTPA<sup>8</sup>). DIDPA and PALADIN have been tested on genuine raffinate from the Purex process with some success.

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<sup>5</sup> DIDPA: Di-IsoDecylPhosphoric Acid.

<sup>6</sup> CMPO: di-isobutylphenylloctyl CarbamoylMethylPhosphineOxide.

<sup>7</sup> HDEHP: Di-EthylHexylPhosphoric acid.

<sup>8</sup> DTPA: Di-ethyleneTriaminoPentaAcetic acid.



Studies to modify the Purex process for co-extracting Pu, U, and Np are being undertaken by JNC which demonstrate the U/Pu/Np co-extraction process in laboratory scale 'hot' experiments. In the traditional Purex process, Np is partially contained in the Pu/U products from the first solvent extraction cycle. However, it is a challenge to make almost all of the Np extractable with Pu. Precise valency control of Np and Pu is a key issue for efficient recovery of Np.

Worthy of note is the UNEX process developed cooperatively by the USA and The Russian Federation for the treatment of high-level sodium bearing liquid waste. It uses a mixture of cobalt dicarbollide and CMPO to co-extract caesium, strontium and the actinides. The components are then stripped selectively by specific washing solutions. The process has been tested with real waste and application to LWR spent fuel treatment is under study.

- Two-step processes.

Two-step processes typically extract trivalent actinides and lanthanides together from the Purex raffinate and then perform a specific separation between the actinides and lanthanides. If necessary, a third step can be performed to separate americium from curium.

Co-extraction of the actinides and lanthanides can be achieved by several means. The TRUEX process uses CMPO to extract actinide and lanthanide salts from acidic feeds. Some difficulties arise with the stripping of the metal ions. The DIAMEX process uses malonamide to extract actinide and lanthanide salts from acidic feeds. Malonamide has the advantage of being fully degradable into volatile organic compounds. The process has also been tested with real high-level liquid waste by the Transuranium Institute and CEA, with encouraging results. The TRPO process is based on a family of tri-alkyl phosphine oxide extractants ( $R_3PO$ ). Tested with success on genuine high level liquid waste, its main drawbacks are the need to adjust the acidity and then back-extract with highly acidic solution.

Actinide/lanthanide separation can be achieved using several techniques. The TALSPEAK process was developed in the 1960s and is considered the reference process for actinide/lanthanide separation. After the HDEHP solvent is loaded with the mixture of An(III) + Ln(III), a selective stripping of An(III) under the action of an aqueous solution containing DTPA and a hydroxycarboxylic acid (e.g., lactic, glycolic or citric acids). The TALSPEAK process is very efficient but very sensitive to the feed acidity. A synergistic mixture made of the terdentate N-ligand, 2-(3,5,5-trimethylhexanoyl-amino)-4,6-di(pyridin-2-yl)-1,3,5-triazine (TMAHDPTZ) and octanoic acid was developed by CEA and tested on genuine solutions. It is also very sensitive to feed acidity.

The CYANEX 301 process is based on the use of acidic sulphur-bearing extractants ( $R_2PSSH$ , dialkyldithiophosphinic acid). It has already been tested on genuine solutions and showed a high efficiency for An/Ln separation. The process is, however, very sensitive to feed acidity and generates a sulphur-bearing waste that can be difficult to handle. The ALINA process is an evolution of the CYANEX process, developed to allow the use of a higher feed acidity. It uses a combination of bis(chlorophenyl) dithiophosphinic acid and tri-n-octylphosphine oxide (TOPO). The problem with sulfur-bearing waste remains.

The SANEX process has been tested successfully with real high-level liquid waste at CEA and ITU. It uses the family of bis-triazinyl-1,2,3-pyridine (BTP) extractants to enable high separation factors even with feed acidity as high as 1 M. The main drawback of the process is the poor stability of the BTP molecule.

- Americium/curium separation.

Most of the processes under development for the separation of americium from curium are based on the ability to oxidize americium to a valency higher than three while curium remains unaffected in the (III) state. In the SESAME process, americium is oxidized to Am(VI) by electrolysis (France) or ammonium persulphate (Japan) in the presence of heteropolyanions to stabilize intermediate valence states. The Am(VI) can then be selectively extracted using ordinary tributylphosphate. One drawback is that Am(VI) oxidation of TBP competes with the extraction, lowering the recovery yield. JNC developed a process based on a laboratory technique known since the 1960s: americium is electrochemically oxidized to Am(V) and then separated from curium by precipitation of the double carbonate  $K_5AmO_2(CO_3)_3 \cdot nH_2O$ . Unfortunately, the process works only in basic media. Workers in France and The Russian Federation are also investigating the selective precipitation of Am(V) ferricyanide from solutions containing curium and lanthanides.

### 3) Fission product separation technologies

Processes for recovery of certain important fission products are also under development. Radioiodine can be recovered quite efficiently from the head end dissolution step preceding the Purex process, by sparging NO<sub>x</sub> through the dissolver solution and recovering in a sodium hydroxide solution. Workers in the USA are developing a process for precipitation of NaI from the NaOH solution, thereby preparing the target form for transmutation of the I-129 present (the only radioactive iodine species in the off-gas from dissolution of spent fuel that has cooled for long times). The recovery of Tc-99 is somewhat more difficult, because even though technetium tends to co-extract with uranium, it also contaminates the plutonium product and the raffinate solution to some extent. In addition, approximately 10-20% of the technetium present in spent nuclear fuel remains in the insoluble residues following fuel dissolution. The UREX process of the USA employs acetohydroxamic acid (AHA) to suppress the extraction of plutonium by TBP; it also prevents the contamination of the raffinate stream. The presence of AHA may actually increase the proportion of Tc going to the HA raffinate by complexing the zirconium which otherwise also co-extracts with Tc, and permits very nearly quantitative stripping of Tc from the uranium product. Processes for the efficient conversion of the pertechnetate ion to technetium metal (the preferred target form for transmutation of technetium) are under development.

It may also prove advantageous in the future to extract isotopes with high rates of heat generation from the wastes to be directed to a geologic repository. Processes for extraction of caesium and/or strontium can be applied to the Purex raffinate. Japanese researchers have studied the use of inorganic sorbents, while extractants such as crown ethers (USA), cobalt dicarbollides (Czech Republic) and calix-crown ethers (France) have also been studied with some success.

### 4) Waste management

Partitioning followed by conditioning (P&C) is an intermediate strategy towards P&T. For waste management purposes the separated Np, Am-Cm could preferably be mixed with a very

insoluble matrix of the type Zirconolite, Hollandite and Perovskite known as "Synroc" and can be immobilized. Once in the embedded form, retrieval of the nuclides from the matrix is very difficult. Their solubility in geologic fluids is several orders of magnitude lower than conventionally vitrified waste.

The transmutation of most of the long lived fission products is difficult to achieve. With the practically achievable neutron fluxes the most abundant fission products  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ , with half-lives of only about 30 years, are almost "non-transmutable". However, since their radioactive life is limited to less than 300 years, they can be safely enclosed using engineered barriers.

In many cases, the necessity of an isotopic separation and difficulties in the target preparation present other important obstacles for the fission product transmutation. Long-lived fission products, which dominate the long term risk of HLW repositories, are, in order of decreasing half-life, I-129, Cs-135, Tc-99, Sn-126 and Se-79. The relative radiological importance of these nuclides varies depending on the repository concept and the type of host rock. From the characteristics it follows that, in practice, only I-129 and Tc-99 can be transmuted and the radiological impact of the other long-lived fission products can be reduced only by special conditioning and confinement. The use of high power laser in transmutation is being explored.

### 3.3.2. Non-aqueous technologies

Non-aqueous reprocessing technologies have been an elusive target of separation chemists for many years, as they have sought to capitalize on characteristics such as rapid reaction rates at high temperatures and higher critical masses in unmoderated reaction vessels. For a variety of reasons, the development of these technologies has not reached the stage of commercialization. The primary reasons for this are the technical maturity and economic success of the aqueous solvent extraction Purex process. The virtually universal deployment of the light water reactor, operating with oxide fuel, has not facilitated dry processes using metal electro-refining, but it is directly compatible with oxide electro-winning now in development in the Russian Federation. Now, however, the potential exists for the diversification of nuclear reactor types and for their application using fuels of higher rating and burn-up. For example, if there is a need to reprocess coated-particle fuel discharged from gas cooled reactors, a non-aqueous reprocessing method or a hybrid aqueous/non-aqueous process may be optimum. If fast reactors, either critical or accelerator-driven sub-critical, are utilized for transuranic burning, the specialized fuel types developed for this purpose may be best processed by non-aqueous methods. A number of countries have embarked on limited programmes for development of non-aqueous reprocessing technologies, with a number of different motivations. Much of this work can build upon past developments in the field spanning several decades.

Dry technology is also finding its way in some complementary processes of the conventional wet technologies. For example, wet decontamination in the conventional fuel fabrication plant has now been largely replaced with dry methods to reduce liquid waste and subsequent treatment processes.

#### 1) Historical perspective

In the early 1960s, fuel discharged from the EBR-II sodium-cooled fast reactor in the United States was treated by a simple pyrochemical process known as "melt refining," in which the

irradiated metallic fuel was melted to release volatile fission products. The highly-enriched uranium was recovered together with about 5 weight % noble metal fission products, which could be tolerated in the fast spectrum of this reactor. Plutonium tended to react to form a “skull” on the zirconium melting crucible and could be recovered separately. About 2.4 t of EBR-II fuel was processed by this method over a five-year demonstration period.

Researchers at the Oak Ridge National Laboratory in the USA who worked in the Molten Salt Reactor Experiment (MSRE) project in the 1960s developed pyrochemical techniques for online processing of the salt fuel, which had the nominal composition of 64.5 wt % LiF, 30 wt % BeF<sub>2</sub>, 5 wt % ZrF<sub>4</sub>, and 0.13 wt % UF<sub>4</sub>. The only step that was implemented before termination of the project was the removal of volatile fission products. More recent studies of molten salt reactor systems in the Russian Federation and in the Czech Republic envision an on-line system or by-pass system for extraction of the lanthanides, alkali metals and alkaline earth fission products from the fuel salt by electrolysis or for recovering the plutonium and minor actinides by electro-winning. The fluoride salt system offers some advantages over a chloride system by virtue of a greater possibility for reducing the lanthanide contamination of the actinide product.

The existence of volatile fluorides of uranium, neptunium and plutonium prompted the development of fluoride volatility processes for the treatment of spent fuel in the 1960s and 1970s. Considerable work was done on fluidized-bed fluorination of crushed oxide fuels, but no commercial use was made of the process beyond that in the US Midwest Fuel Reprocessing Plant, in which purified uranium was to be extracted from impure uranyl nitrate by conversion to UF<sub>6</sub> and separation of the volatile fluorides of transuranic contaminants from the uranium by fractional distillation. Unfortunately, this plant has never operated with spent fuel. More recently, in the 1980s, the Nuclear Research Institute of Czechoslovakia operated a small-scale fluoride volatility processing line for the treatment of oxide fuel discharged from the Russian BOR-60 fast reactor. A flame fluorination method was used in the process, and reasonable recovery efficiencies were achieved. The same institute, now in the Czech Republic, is studying the use of the fluoride volatility process for treatment of LWR spent fuel, in order to provide feed materials to an accelerator-driven molten salt reactor for consumption of transuranic elements.

## 2) Status of technical development.

It is most convenient to survey the new non-aqueous reprocessing technologies under development in terms of the fuel type to which they could be applied, recognizing that some processes are applicable to more than one fuel type.

- Oxide Fuels

The liquid raffinate from the Purex process has been sent to a vitrification process for immobilization of the minor actinides (Np, Am, Cm) present in the high level liquid waste stream, together with fission products, after calcination (as in the AVM process in France) or directly to the LFCM process (liquid-fed ceramic melter). With increasing emphasis being placed on reduction of the radiotoxicity of high level wastes destined for disposal in geologic repositories, renewed attention is being paid to the recovery and elimination of the minor actinides. Work is being done by CEA in France, for example, on molten salt processes for the separation of actinides from fission products. A fluorination step is used to convert the oxides to fluorides, which are then dissolved in a CaF<sub>2</sub>-MgF<sub>2</sub> mixture at about 750°C. The actinides are then recovered in a reductive extraction process whereby they are extracted from the salt into a metallic solvent (e.g., Zn, Cu-Al, or Cu-Al-Zn) in

the presence of a reducing agent (Mg). The solvent is then removed by distillation. It also appears possible to chlorinate the calcine product and then recover the minor actinides by electrochemical means after adjustment of the oxoacidity of the electrolyte salt.

Russian scientists at the Research Institute for Atomic Reactors (Dimitrovgrad) have developed and demonstrated a process known as DDP (Dimitrovgrad Dry Process) for the reprocessing of fast reactor oxide fuel. In their process, which is essentially an electro-winning process, spent oxide fuel is declad and fragmented. The powdered fuel is placed in a pyrocarbon vessel containing a molten mixture of alkali chloride, for instance, NaCl-CsCl, at about 630°C. The fuel is chemically or electrochemically dissolved and the UO<sub>2</sub>, present in large excess, is deposited at the cathode, together with a small amount of noble metal fission products, which are subsequently removed by electrolysis. A chlorine/oxygen gas mixture is then sparged through the vessel to form chlorides and oxychlorides (such as PuO<sub>2</sub>Cl<sub>2</sub>) of the actinide elements. The salt is then electrolyzed to recover uranium and plutonium. The electrolysis process, typified by the reaction  $\text{PuO}_2^{2+} + 2e^- = \text{PuO}_2$ , results in the co-deposition of uranium, neptunium and plutonium oxides at the cell cathode and liberation of chlorine at the anode. Some contamination of the deposit with americium and curium occurs. The balance of the americium and curium remains in the salt bath. An extra electrowinning step is then introduced to recover the minor actinide oxides together with remaining uranium oxide. The cathode deposits in both cases are separated from adhering salt by washing with water. The recovered mixed U-Pu-Np oxides are incorporated into fresh fuel rods by vibratory compaction. Recycle of some oxide fuel elements in the BOR-60 reactor has been accomplished. The minor actinide oxides are presumably available for transmutation as appropriate.

Workers at the Argonne National Laboratory are studying a pyrochemical processing method for treatment of spent LWR oxide fuel. After decladding and crushing, the fuel powder is placed in an electrochemical cell in which the fuel and fission product oxides are reduced to the metallic state by electrolysis in a LiCl bath containing 1 wt.% Li<sub>2</sub>O and operated at 650°C. Oxygen is liberated in the process and the reduced metals are collected in the cathode basket. This basket becomes the anode in the next step, where uranium is extracted by an electrorefining process. The metallic uranium is deposited on a solid steel cathode, and the uranium is recovered by melting at reduced pressure to volatilize off any adhering LiCl. The transuranic elements, and all but the noble metal fission products, are anodically dissolved in the electrorefining process and remain in the LiCl electrolyte salt. The transuranics (and a quantity of the remaining uranium) can be recovered in a liquid cadmium cathode, collecting about 3-5 kg of TRUs per batch. Alternatively, an electrowinning process could be used to extract the transuranics after the electrorefining step to recover uranium. The oxide fuel treatment process (now known as the PYROX process) is at a very early stage of development, but experiments have shown the technical feasibility of the various steps. The principal concern with the process is the need to deal with a large amount of uranium that has little value except as a fertile material for use in fast breeder reactors.

The Korea Atomic Energy Research Institute (KAERI) at Taejon is studying the application of a chemical reduction process using lithium metal to prepare spent LWR oxide fuel for disposal. (Reprocessing is currently circumscribed in the Republic of Korea.) Their fuel conditioning process would greatly reduce the volume of the material to be disposed in a geologic repository and could provide a front-end process for a partitioning and transmutation system known as HYPER that is currently under development.

For over ten years, KAERI have been developing the DUPIC process for direct recycling (without chemical processing) of spent PWR fuel in CANDU reactors. If discharged at relatively low burn-up, the PWR fuel is still sufficiently enriched in fissile material (about 0.6% Pu and 0.9% U-235) for use in the CANDUs. In the DUPIC process, the PWR spent fuel cladding is punctured and the fuel rod is heated to high temperature under a partial pressure of oxygen. The conversion of  $\text{UO}_2$  to  $\text{U}_3\text{O}_8$  results in volume expansion and pulverization of the fuel material, and efficient removal of the volatile fission products. The remaining powder is then pressed and sintered to form CANDU-sized fuel pellets. The DUPIC process is much simpler than conventional wet-chemistry techniques for reprocessing, and promises to be cheaper. It presents a significant anti-proliferation benefit as well, since radioactive fission products and fissile material are not separated. In addition, since the heat load of spent DUPIC fuel is similar to that of the original spent LWR fuel, disposal requirements do not increase. The limitation of the process is the motivation of PWR operators to reduce fuel cycle costs through increased fuel burn-up, which reduces the CANDU recycle value of the uranium in the spent PWR fuel.

- Metallic Fuels

When the Clinch River Breeder Reactor Project in the USA was cancelled in 1984, among the reasons given for the decision to terminate the project were the high costs of fast reactor fuel aqueous reprocessing and the proliferation issues associated with such reprocessing. Accordingly, Argonne National Laboratory proceeded with the development of a low cost, proliferation-resistant process for treating fast reactor metallic fuels, as part of the Integral Fast Reactor development program. By 1987, process development had converged on an electro-refining method in which metallic fuel pins were chopped and the fuel pin segments placed in a stainless steel mesh basket that became the anode of an electro-refining cell using a LiCl-KCl electrolyte. Application of a potential of less than one volt between the anode basket and the simple steel rod cathode results in anodic dissolution of the constituents of the spent fuel (except for the noble metal fission products, which will not form chlorides under these conditions and remain at the bottom of the anode basket). Uranium is electro-transported from the salt to the steel cathode at a rate of 3 grams per ampere-hour of charge passed. The transuranic elements will not deposit at the steel cathode as long as the TRU:U ratio in the salt is less than 100 or so, due to the higher stability of the TRU chlorides relative to  $\text{UCl}_3$ , and the resultant tendency for the back reaction such as  $\text{Pu} + \text{UCl}_3 = \text{PuCl}_3 + \text{U}$ . Recovery of the transuranic elements requires the use of a different cathode, one in which the TRUs deposit as intermetallic compounds with cadmium in a crucible containing liquid cadmium that is suspended in the electrolyte salt. Collection of several kg of transuranics is possible, corresponding to a loading in the cadmium of about 50 volume % (i.e., greatly exceeding the solubility of these materials in Cd). Deposition of the transuranics is accompanied by a certain amount of uranium, depending upon its concentration in the salt. A mixture of 70% transuranics, 30% uranium and 5% lanthanide fission products is typical.

A modified version of the electro-refining process is being used in the treatment of EBR-II fuel and blanket elements for disposal. Because this treatment is for the purpose of waste management and eventual repository disposal of the high level waste, only uranium is being recovered in the course of the treatment (i.e., the liquid cadmium cathode is not utilized). The EBR-II fuel at discharge is about 57% enriched, and the uranium deposits are melted together with depleted uranium to down blend the product to less than 20% enrichment so that it can be stored under the category of low enriched uranium (LEU).

The transuranic elements are left in the electrolyte salt, which will be periodically removed to produce a composite glass-ceramic waste form containing the TRUs and the active metal fission products. The noble metal fission products that remain in the anode basket are combined with the cladding hulls and melted together to form a metallic waste form for disposal.

Argonne scientists have developed two electro-refiners that are being used for the treatment of EBR-II fuel (~1.5 t) and blanket (~29 t). The highly-enriched driver fuel is processed in an electro-refiner with a batch size of 20 kg, whereas the depleted uranium blankets are processed in an advanced low-resistance electro-refiner with a 350-kg capacity. Both systems are being operated successfully, and the process of spent fuel treatment will continue for several more years. Even though there is very little fission product content in the blanket fuel, there is a significant quantity of plutonium present, and it may be necessary to remove the plutonium from the electro-refiner electrolyte salt before the fission product content reaches a level that would be appropriate for salt removal.

Workers at the Central Research Institute of the Electric Power Industry (CRIEPI) in Japan have followed a similar path and have performed work on actinide/lanthanide extraction in molten chloride and liquid metal (Cd,Bi) media by electro-deposition and liquid-liquid extraction. They have conducted work in Japan with uranium and with plutonium at the European Union's Institute for Transuranium Elements in Karlsruhe, Germany. Scientists at the CEA Marcoule laboratory are conducting studies with liquid metal cathodes and with reductive extraction processes.

- Coated-particle fuel

Future gas cooled reactors will almost certainly utilize TRISO-coated fuel. This fuel consists of fuel (UOX, MOX, U-Th oxides, etc.) microspheres, 50 to 300  $\mu\text{m}$  in diameter with successive coatings of porous carbon, pyrolytic graphite, silicon or zirconium carbide, and a final outer coating of pyrolytic graphite. The overall particle diameter is of the order of 800  $\mu\text{m}$ . If the fuel is to be reprocessed (and requirements on radiotoxicity reduction may mandate such reprocessing), these coating layers must be removed in order to provide reagent access to the fuel material. Both aqueous and non-aqueous processes are being developed, initially in concept only, for the treatment of these fuels.

The development of the processing technology for TRISO fuels can build to some extent on experience gained during the 1970s, when a process involving burning of the outer layers of graphite was developed. The fuel particles were crushed by passing through a set of steel rollers, exposing the fuel material for dissolution by nitric acid. The balance of the process followed a standard Purex flowsheet. Considerable problems were posed by off-gas handling, and recovery efficiencies were not particularly high.

Non-aqueous processes now being studied for application to the treatment of TRISO fuel include fluoride and chloride volatility processes, carbo-chlorination processes, and direct electrochemical dissolution. A problem common to all of these conceptual processes is the disposition of the large amount of carbon and silicon remaining from the processing of this fuel. Waste volumes can be very large, even though the fuel burn-up capability might be quite high. France and the United States are presently collaborating on the evaluation of TRISO fuel treatment processes, and there may be other collaborative efforts in the future.

- Other fuel types

A variety of different non-fertile fuels are being considered for use in partitioning and transmutation systems. Fuels currently under study are metallic (alloy of TRU with Zr), nitride cercer (dispersion of TRU nitrides in an inert matrix such as ZrN), oxide cercer (dispersion of TRU oxides in inert ZrO<sub>2</sub> or MgO), cermet (dispersions of TRU oxides or nitrides in a metal matrix such as Zr), and carbide cercer (dispersion of TRU carbide in SiC). The high content of transuranics in these fuels, together with the high zirconium content in many of them, tends to favor non-aqueous reprocessing methods.

The development of processes for treatment of these fuels is just beginning, and it is a fertile area for international collaboration. There are already some indications that direct molten salt electro-refining is technically feasible for the metallic alloy fuel and for the nitride cercer fuel. A similar process for the carbide cercer is problematic because the liberated carbon would foul the electrolyte. Non-aqueous processing may be necessary for the TRU oxide dispersion in ZrO<sub>2</sub>, and might involve a fluoride or chloride volatility process. An even more complex non-aqueous process is envisioned for the treatment of the cermet fuels. In the USA, the program for partitioning and transmutation has set a date of 2006 for selection of processing methods for specific fuel types.

### 3) Waste management scheme for pyrochemical process

In a reference pyrochemical process, the active metal fission products (Cs, Sr, etc.) will reside in the electrolyte salt together with trace amounts of actinide elements. The transition metal fission products will remain in the anodic dissolution baskets together with the cladding hulls. So, there are two waste streams to deal with: one salt and one metal. Because a chloride salt is not amenable to vitrification, a natural chloride-bearing mineral, sodalite (NaAlSi<sub>3</sub>O<sub>8</sub>·nH<sub>2</sub>O) equivalent is synthesized by mixing the fission product-loaded electrolyte salt with a zeolite (Zeolite A, Na<sub>12</sub>Al<sub>12</sub>Si<sub>12</sub>O<sub>48</sub>) and heating to temperatures near 900°C. The synthesis is catalyzed by the presence of borosilicate glass frit, which also serves to encapsulate the sodalite particles, providing an additional barrier to radionuclide release. As is the case with vitrification of high level waste, the fission product loading of the waste form is constrained by the limiting centerline temperature of the waste form and the total heat generation per unit area acceptable in the high level waste repository or interim storage site. The metallic waste stream generated by melting the baskets together with the metallic fission products and the cladding hulls is used to produce a corrosion-resistant metal alloy. This alloy is dependent in composition on the nature of the cladding material, which dominates the mass of the waste form. A metal waste form based on stainless steel cladding hulls has shown a release rate for technetium that is several orders of magnitude less than the release of technetium from spent LWR fuel.

## 3.4. FUTURE ACTIVITIES

The sustainability of nuclear power generation is vitally dependent upon an assured fuel supply and stability of fuel cycle costs. Light water reactors can be expected to remain the principal component of deployed nuclear systems for many years, so the need for reprocessing of standard oxide fuel will continue. The proportion of MOX fuel in the output of these reactors may increase over the years, and the reprocessing system must be prepared to handle the change. Continued improvements in the Purex process are expected, and



aqueous methods for separation of the minor actinides are likely to be required in the context of advanced waste management systems.

Another attractive possibility that has been explored recently is the recycle of total actinides from the spent fuel, including the minor actinides and possibly the long lived fission products, back into the reactor systems where they undergo transmutation into shorter lived nuclides as a means of improving the long term management of the radiological hazard of the waste. For this long term alternative, separation of the actinides would be required, in a similar way to the reprocessing option and renewed interest is given to some dry technologies that have been in development for recycle of spent FBR fuel. Aqueous processes complementary to Purex are being developed to treat these nuclides separately from HLW. Some dry technologies under development have an edge over the aqueous processes as they have the potential for total actinide recycle without resorting to individual separation schemes providing built-in proliferation resistance to the fuel cycle as explored in the international initiatives.

Research on utilization and transmutation of the minor actinides and long-lived fission products has led to very different concepts from the standpoint of the reactors themselves, as well as the fuel and targets, and the corresponding scenarios for nuclear reactor deployment. Nevertheless, a few constant principles underlie all of these concepts:

- Management of radioactive material flows rich in americium and curium,
- Recycling quickly after removal from the power reactor to avoid the accumulation of decay products and to limit the fuel cycle inventory,
- Implementation of robust processes to handle the high activity of the materials involved,
- Preference for compact facilities, and
- An “integral” fuel cycle concept in which the irradiation, processing and fabrication facilities are located on the same site.

In this perspective, pyrochemical processes have considerable potential. They are highly resistant to irradiation effects and are therefore suitable for use with concentrated media. This is a further incentive to develop pyrochemical processing concepts in order to confirm and better assess the expected advantages.

However, aqueous processes, due to their inherent advantages (high recovery efficiencies and well established technical maturity), remain at the forefront of reprocessing technology. Many improvements to the technology have been made over the years. The need to maintain continuity with today’s plants and to sustain their ability to adapt to new requirements demands continued development of aqueous processing methods.

In any decision on industrial scale deployment of a new technology, rigorous evaluation of this technology is needed before it can be considered ready for commercialization. Many years of large scale testing under industrial conditions are required in order to provide a basis for an informed decision on industrialization. A less disciplined approach could have dire financial consequences. It must be recalled that over two decades of military-related aqueous reprocessing, experience at very large scale was accumulated before the first commercial reprocessing plant was operational.

The scientific community must be given full support in developing advanced reprocessing technologies, because the industry should have the option for continued technical evolution that can take the form of increased profitability, more efficient resource utilization, and even greater environmental stewardship. History teaches us, however, to avoid premature

commitment, or the application of new technologies beyond the limit of their true capabilities. As new technologies progress beyond the laboratory bench and pass the test of commercial feasibility, the market place will recognize their potential. It is essential to remain open to emerging technologies, but to exercise sound engineering judgment in setting the course for future deployment. Last, but not least, among the issues to be examined prior to commercialization is the compliance with safeguards requirements for the new technology.

#### **4. IAEA ACTIVITIES IN SPENT FUEL REPROCESSING**

The IAEA activities in spent fuel management have evolved in response to the changing needs and interests of the Member States. The status and trends in the Member States through the past decades have been closely surveyed and reflected in the formulation of IAEA programmes which have dealt with a variety of technical and institutional topics. The new realities emanating from the recent trend toward globalization of market economies, together with sustainability and security issues associated with the nuclear fuel cycle options may bring a new shift in the IAEA activities.

##### **4.1. HISTORICAL BACKGROUND (1970s)**

The rapid emergence of the nuclear industry around the early 1970s had given rise to active development in plans for fuel cycle services. In 1970, the IAEA published a TECDOC on spent fuel reprocessing in response to such needs in the Member States [77]. The global momentum for development of nuclear power and of fuel cycle services was accelerated by the oil-shock in the mid-1970s [81]. However, subsequently, this trend was reversed by, among others, two issues of concern: nuclear fuel supply and proliferation. This led to a series of studies under the aegis of the IAEA in search of possible solutions to the issue of nuclear materials management by an international co-operative framework.

##### **4.1.1. RFCC**

The first of the series of IAEA studies which started in 1975, and concluded in 1977, was entitled RFCC (Regional Fuel Cycle Centres) with a view to examining from various angles, such as economics, safety and safeguards, the feasibility of providing nuclear fuel cycle services, including spent fuel reprocessing, by regional centres. This study identified the relative merits of a multi-national approach to regional fuel cycle centres, including a cost saving by a factor of 2 ~ 3, in addition to the enhanced safeguards and physical protection [82].

##### **4.1.2. INFCE**

The RFCC was followed by a more comprehensive review of the conventional fuel cycles, INFCE (International Nuclear Fuel Cycle Evaluation), which lasted for a couple of years (1978~1979). Among the eight groups working for the INFCE, Working Group 4 addressed spent fuel reprocessing, including plutonium handling and recycle, while Working Group 6 was focused on spent fuel storage under the title of Spent Fuel Management [83].

As a major international study, the INFCE intended to examine the issues associated with fuel cycles, but it did not manage to produce a notable international consensus between Member States on an overall fuel cycle policy. A recommendation was nonetheless made that some issues on spent fuel management should be discussed at the expert groups established by the

IAEA; one for International Plutonium Storage (IPS) and the other for International Spent Fuel management (ISFM).

#### 4.1.3. EG-ISFM

The two expert groups (IPS and ISFM) began working in parallel to the INFCE. The two groups separated in 1978 from an initially joint group which had a mandate to come up with a concrete plan for the international management of spent fuel and plutonium. The former was terminated in 1980, while the latter had a more durable agenda which lasted until 1982 [84].

One of the results from the ISFM was the clear recognition of the important roles of spent fuel storage in the fuel cycle backend, thus providing a momentum to establish a regular program on spent fuel management in the IAEA Secretariat. The Expert Group made a recommendation, among others, to establish a standing advisory group to review and advise on the relevant IAEA programme.

#### 4.1.4. RAG-SFM

According to the recommendation referred to above, the Regular Advisory Group on Spent Fuel Management (RAG-SFM) was established and the first meeting was held in 1984. The bi-annual meetings of RAG-SFM have continued for about two decades until being merged recently with IWG-NFCO (International Working Group on Nuclear Fuel Cycle Options) which was an off-shoot from the "Symposium on Fuel Cycle and Reactor Strategies: Adjusting to the New Realities", held in 1997 [85].

The RAGM published a series of reports on the status and trends in spent fuel management in the Member States and several newsletters through the eighties and nineties.

### 4.2. THE 1980s: THE MATURING PERIOD

The post-INFCE period of the 1980s proved to be a decade of stabilization and growing maturity for the nuclear industry in general, and for the spent fuel management sector in particular, rather than one of expansion and innovation. By that time, a few countries had shifted their policy for spent fuel management from reprocessing to direct disposal, declaring spent fuel as waste. The policy shift to direct disposal from the classic option of reprocessing for plutonium recycle, although initiated by the USA as a result of proliferation concerns, was later joined by several more Member States which regarded spent fuel with its plutonium content as a liability rather than an asset. The sluggish growth in nuclear energy and the all time low price of uranium, among other factors, compounded by the increasing costs for the commercialization of fast breeder reactors (FBR), as witnessed in the 1980s, worsened the economics of plutonium recycle. Most of the ambitious FBR programmes which Member States had established earlier were cancelled and the plutonium from reprocessing was subsequently diverted to MOX for LWRs, even in those Member States still committed to the closed fuel cycle. The 1980s could be characterized as a maturing period for the nuclear fuel cycle technologies, including spent fuel reprocessing. In reflection of these trends, the IAEA organized the Symposium on the Back End of the Nuclear Fuel Cycle: Strategies and Options [86].

The Chernobyl accident, which happened in 1986, had a devastating impact on public perception of the nuclear option and to the issue of radioactive waste management, including spent fuel. The issue of public acceptance for spent fuel management was further aggravated by the globalization of green movements against the siting of facilities and the transportation

of radioactive materials. Many Member States had to defer or modify their plans for spent fuel management, due to the difficulties of securing public acceptance in the aftermath of the Chernobyl accident. The lessons learned from the 1980s gave rise to a momentum to strengthen nuclear safety, both on the national and international scale, and this was reflected in the IAEA's programmes.

A number of IAEA activities in spent fuel management had been implemented in the 1980s with a major focus on the development of storage options and technologies, resulting in a handful of relevant publications.

#### 4.3. NEW REALITIES IN THE 1990s

The demise of the cold war at the end of 1980s, and the subsequent globalization of the market economy in the early 1990s, brought some far-reaching effects to the nuclear industry and, consequentially, to spent fuel management. In many Member States, deregulation of the energy market has resulted in privatization of former public utilities, and the shrinking of the market as the internationalization of businesses led to the merger of a number of companies in the nuclear sector.

Whilst commercial reprocessing in Western Europe continued to mature with improving performance, the reprocessing enterprises in the Russian Federation were significantly affected by the withdrawal of business by Eastern European customers after the imposition of tariffs for the reprocessing service. The nuclear fuel markets were also perturbed in the 1990s by the availability of nuclear materials of military origin released as part of a treaty agreement between the USA and the Russian Federation. Reflecting these new realities, the IAEA organized the International Symposium on the Nuclear Fuel Cycle and Reactor Strategies: Adjusting to New Realities [87]. An outcome of this Symposium was the establishment of the International Working Group on Nuclear Fuel Cycle Options (IWG-NFCO).

A series of meetings was also held in the 1990s to undertake a state-of-the-art review of status and trends in spent fuel reprocessing, which resulted in a TECDOC [88]. This present publication is an extension of the IAEA activities initiated in the 1990s.

#### 4.4. NEW MILLENIUM

The needs for innovative systems was recognized by IAEA which launched the INPRO initiative, which was supported by General Conference held in 2000. Kicked off in 2001, the main objectives of INPRO are to assist Member States in the national or joint assessment of, to provide an umbrella for their development and to facilitate their deployment.

The initial efforts of INPRO had focused on the definition of user requirements for the innovative nuclear energy systems (INS). Now with participation of 22 Member States, INPRO has made progress to a stage of wrapping up Phase I by the end of this year. It is lead by Department of Nuclear Energy, but also requires expertise and support from other Departments. Currently the INPRO assessment methodology is being validated on the basis of several case studies that have been performed by INPRO members and individual [89].

The importance of technical innovation for nuclear fuel cycles for sustainable development of nuclear energy was recognized in the Scientific Forum held during the 48<sup>th</sup> General Conference of IAEA. The Forum with a topic of "Fuel Cycle Issues" was an occasion to review issues and discuss challenges of technical innovations with a number of technical

objectives, including: more efficient utilization of fissile and fertile materials; enhanced proliferation resistance through, inter alia, passive control of nuclear materials using new fuel types and configurations; greater reliance on passive safety features; and technology advances to mitigate the volume and radio-toxicity of high level and long lived wastes. These technical innovations will need to be complemented by new approaches to relevant policy and institutional issues - for example, through increased harmonization of regulatory requirements and industrial codes and standards.

## 5. CONCLUSIONS

Historically, reprocessing has been the classic option for spent fuel management. With Purex technology fully matured industrially in several countries, one-third of the global cumulative arisings of spent fuel has already been reprocessed. The balance of the global arisings is currently stored as inventory either in conventional pools or in the more recently developed dry storage systems. Reprocessing, therefore, can be regarded as the only currently proven option for spent fuel management with an end point, pending disposal in a geological repository.

Notwithstanding the success of reprocessing in spent fuel management, a growing number of countries producing nuclear power have changed their policy to direct disposal, or have deferred decisions, as concerns arose on the issue of proliferation, as was highlighted by the INFCE in the late 1970s. While these issues continue to be controversial, changing global realities have called for new deliberations on reprocessing as a technological option for the long term management of spent fuel, with possible innovations in technology and applications. It might be possible to see some technical breakthrough enabling to resolve those issues observed today on the one hand or the criteria and perceptions concerning those issues might be different from now in such way that some of them become acceptable in the new circumstances of the future : "*Waste is a commodity in the time and wrong place*". This kind of change has already been observed : the major issue of nuclear fuel cycle has shifted from front end to backend in particular to radioactive waste management. Reprocessing might in future provide a technical basis for partitioning and transmutation of minor actinides to reduce environmental stress with a view to the sustainable utilization of nuclear energy.

Regarding the future for reprocessing technology, the well-established Purex process will continue to be dominant in the near term. The reprocessing technology based on Purex has evolved over a number of decades in both scale and complexity. From the early methods, applied to low burn-up metallic fuel made from natural uranium in comparatively simple forms, highly developed counter-current solvent extraction systems have been successfully commercialized and adapted to deal with enriched high burn-up oxide fuels in fuel assemblies of sophisticated design. Although the health and safety hazards associated with reprocessing are potentially severe, a very good safety record has so far been achieved and internationally accepted standards for the protection of operators and the general public have been established.

In the mid-term future, the technical requirements to be dealt with will increase in severity with the advent of higher burn-up and MOX fuels, but the existing processes, procedures and experiences will allow the associated increase in hazard potential to be controlled and ensure the continued safe operation of future reprocessing plants.

In the longer term, however, with the implementation of advanced reactors and fuel cycle systems, such as partitioning and transmutation, novel reprocessing technology with total

actinide recycle may have to be implemented. This is mainly due to the long term implications associated with the storage and disposal of minor actinides and fission products, as well as the fissile materials, contained in the spent fuel. The efforts to develop these and other emerging technical concepts has recently been linked with the search for innovative nuclear systems, which are themselves being explored in the context of long term sustainability, as exemplified by a number of recent international initiatives, such as INPRO (IAEA), MICANET (EU) and Gen IV (USA). A recent USDOE initiative, called AFCI (Advanced Fuel Cycle Initiative), has the objective of developing advanced fuel cycles to support advanced fuel utilization and advanced waste management strategies, including transmutation, and enabling the transition from current fuel cycles to those to be used with innovative reactor systems.

In order to reflect the evolving perspective of Member States on these and associated topics, the IAEA provided a forum for review and discussion at the Scientific Forum on 'Fuel Cycle Issues and Challenges' which was held during the 48th General Conference of IAEA (21-22 Sept., 2004). The selection of a strategy for spent fuel management is a complex decision with many factors to be taken into account including politics, economics, resource conservation, environmental protection, and public perception, the last of which has become a predominant factor in many Member States.

The relative merits of spent fuel management options vary both over time and the scale under consideration ("*Waste is a commodity in the wrong time or wrong place*"). Apart from the prepaid commercial plants currently in operation, it is in a future timeframe, when uranium prices or other factors influencing total costs, such as fossil fuel prices, that reprocessing will make economic sense and become a competitive market option. As the economics of spent fuel reprocessing is sensitive to the scale of the reprocessing plant used, the globalization of market economies, which has already had profound impacts on the nuclear industry, might provide some future conditions favourable to a reprocessing business structure which is somehow globally optimized. This, in fact, would be equally true for any other spent fuel management option, including direct disposal. A new business model may have to emerge, perhaps based on a total 'cradle to grave' concept for fuel cycle services, and on a regional or international basis which could hopefully provide a breakthrough on a number of issues associated with the long term management of spent fuel.



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## Annex I

### TABLES AND FIGURES

Table I Past, current and planned reprocessing capacities in the world

COUNTRY	SITE	PLANT		OPERATION		CAPACITY	
				START	SHUTDOWN	PRESENT	FUTURE
Belgium	MOL	Eurochemic	LWR	1966	1975		
China	Jiuquan	RPP	LWR	?			25
	Lanzhou		LWR	2020			800
France	Marcoule	APM	FBR	1988	1996		
	Marcoule	UP1	GCR	1958	1997		
	La Hague	UP2	LWR	1967		800	800
	La Hague	UP3	LWR	1990		800	800
Germany	Karlsruhe	WAK	LWR	1971	1990		
India	Trombay	PP	Research	1964		60	60
	Tarapur	PREFRE 1	PHWR	1974		100	100
	Kalpakkam	PREFRE 2	PHWR	1998		100	100
	Kalpakkam	PREFRE 3A	PHWR	2005			150
	Tarapur	PREFRE 3B	PHWR	2005			150
Japan	Tokai-mura	PNC TRP	LWR	1977		90	90
	Rokkasho-mura	RRP	LWR	2005			800
Russian Federation	Chelyabinsk	RT1	WWER-440	1971		400	400
	Krasnoyarsk	RT2	WWER-1000	2020			1,500
UK	Sellafield	B205	GCR	1967	2012	1,500	
	Sellafield	Thorp	LWR/AGR	1994		900	1,000
	Dounreay	UKAEA RP	FBR	1980	2001		
USA	West Valley	NFS	LWR	1966	1972		
	Hanford	Rockwell	U metal	1956	1989		
	Savannah River	SR	U metal	1954	1989		
	Idaho Falls	R	U-Al alloy	1959	1992		
Total Capacity						4860	6845

Table II Cumulative amount of civil reprocessed spent fuel

(Status as of the end of 2003)

Country	Site	Plant	Fuel Type				TOTAL
			GCR	LWR	FBR	MO X	
Belgium	Mol	Eurochemic <sup>a</sup>	19 <sup>b</sup>	86			105
France	Marcoule	UP1	18 000 <sup>c</sup>				18 000
	La Hague	UP2/UP3		19 000	10	9.6	19 020
Germany	Karlsruhe	WAK <sup>a</sup>		180			180
India	Trombay	PP					
	Tarapur	Prefre-1					
Japan	Tokai-mura	TRP		1 000	18d		1 018
Russian Fed.	Chelyabinsk	RT-1		3 500			3 500
UK	Sellafield	B205	42 000 <sup>e</sup>				42 000
	Sellafield	Thorp		5 800 <sup>f</sup>			5 800
	Dounreay	UKAEA RP			14		14
USA	West Valley	NFS <sup>a</sup>		194			194
		Total	60 019	29 760	33	9.6	89 822

<sup>a</sup> Closed facility<sup>b</sup> CANDU, GCR and other<sup>c</sup> UNGG<sup>d</sup> Spent fuel from Fugen<sup>e</sup> Magnox<sup>f</sup> LWR/AGR

Table III. Status of decommissioning of reprocessing facilities (larger than 1 t/y capacities)

(Status as of the end of 2000)

COUNTRY	SITE	PLANT (CAPACITY,t/y)	OPERATION PERIOD	DECOMMISSIONING STATUS (PERIOD)	REMARK
Belgium	MOL	Eurochemic (60)	1966-1975	Under work (1987-)	
France	Marcoule	UP1(400)	1958-1985 -	Under work (1997-)	
France	Marcoule	APM (5)	1960-1982	Completed (-1993)	
Germany	Karlsruhe	WAK (35)	1971-1990	Under work (1997-)	
India	Trombay	PP (60)	1964- 1983-	Completed (-1977)	reoperation in 1983
Italy	Trisaia	ITREC (5)	1975-1988	In plan	
UK	Windsc ale	B-204 (?)	1952-1964	Under work (1990-2010)	
UK	Dounre ay	UKAEA RP (10)	1979-1994	In plan	
USA	West Valley	NFS (300)	1966-1972	Under work (1980-)	

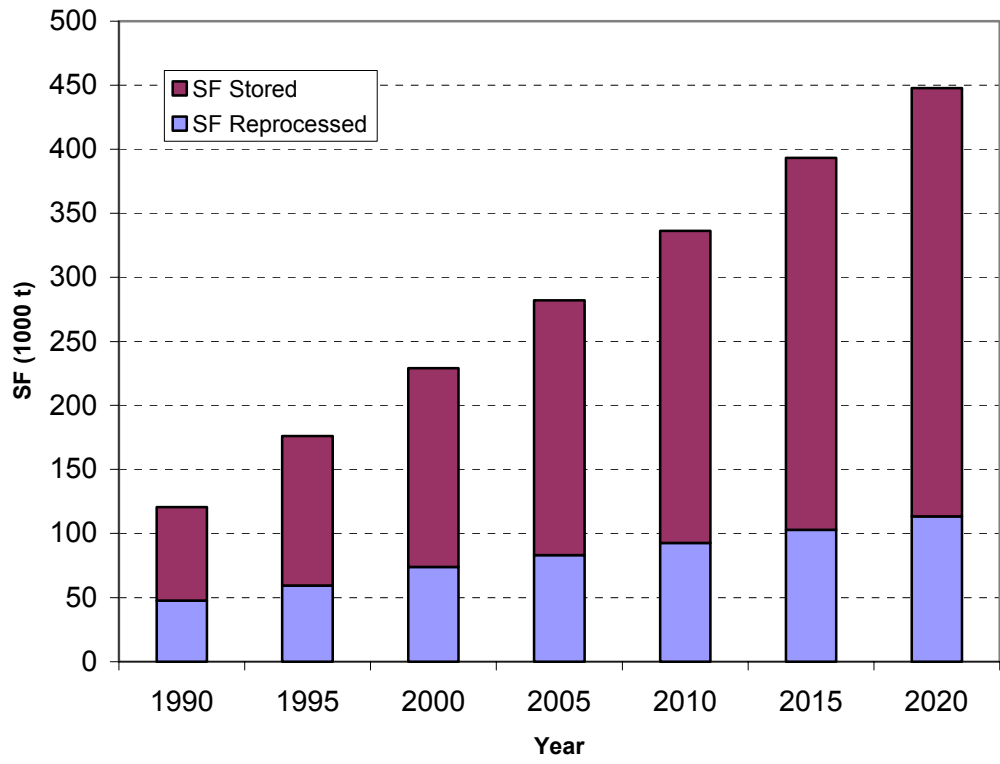


FIG. 1. Global statistics in spent fuel management.

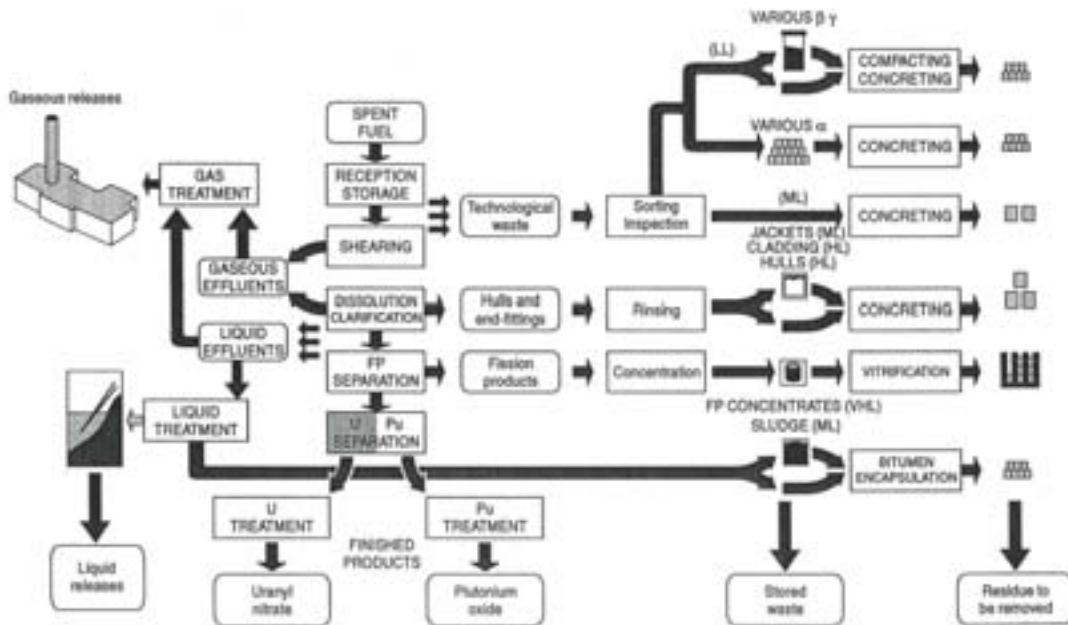


FIG. 2. Illustration of PUREX process.

## Volumes of final residues conditioned in UP3 (High level and long-lived waste after conditioning)

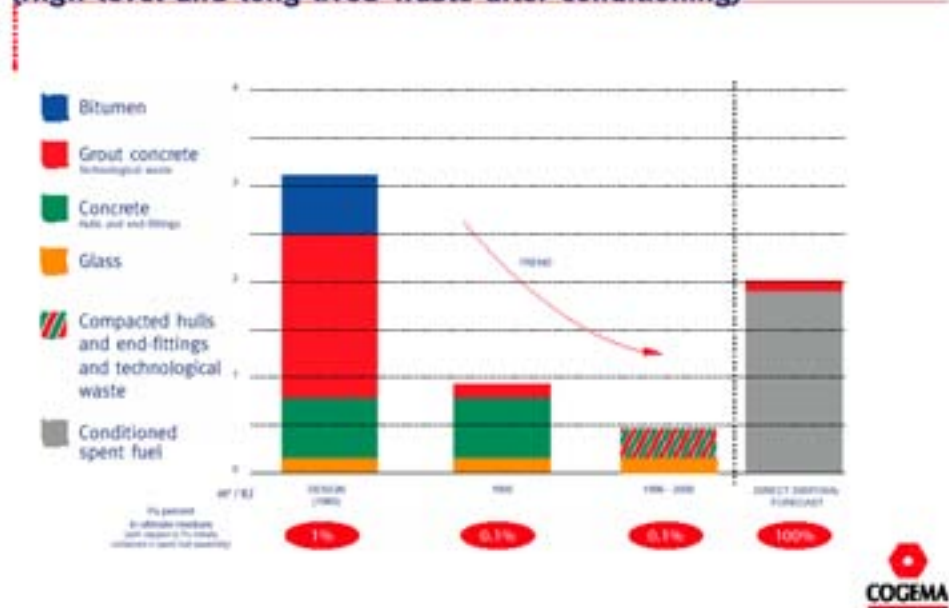


FIG. 3. Radioactive waste volume reductions at La Hague.

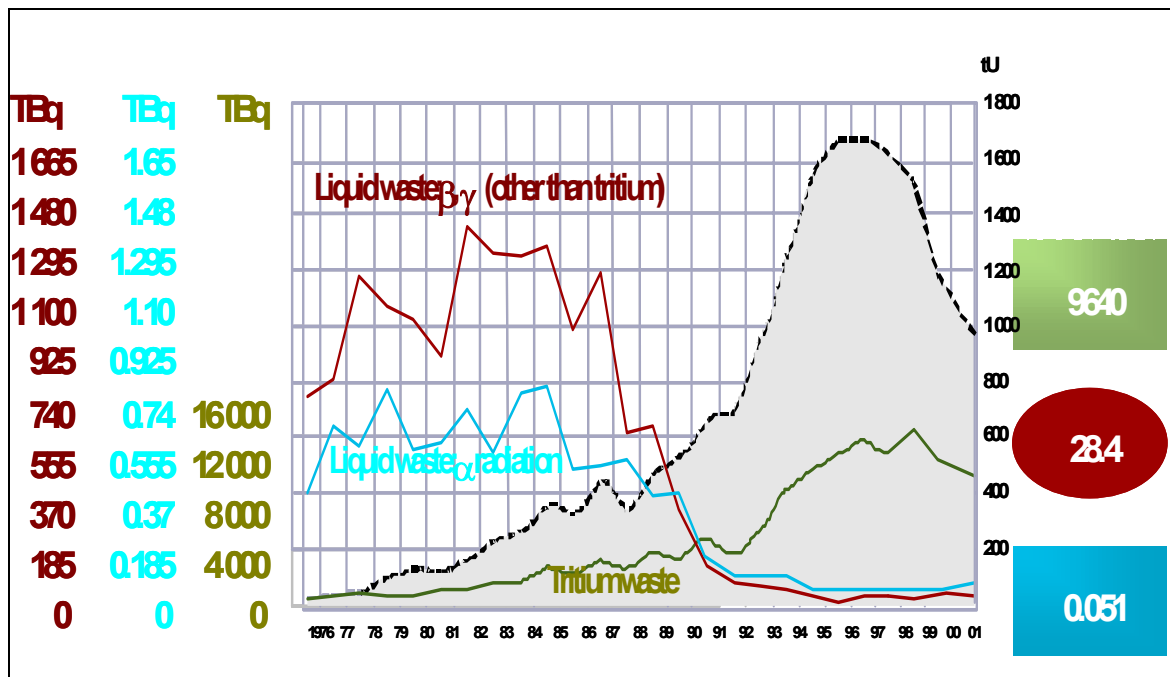
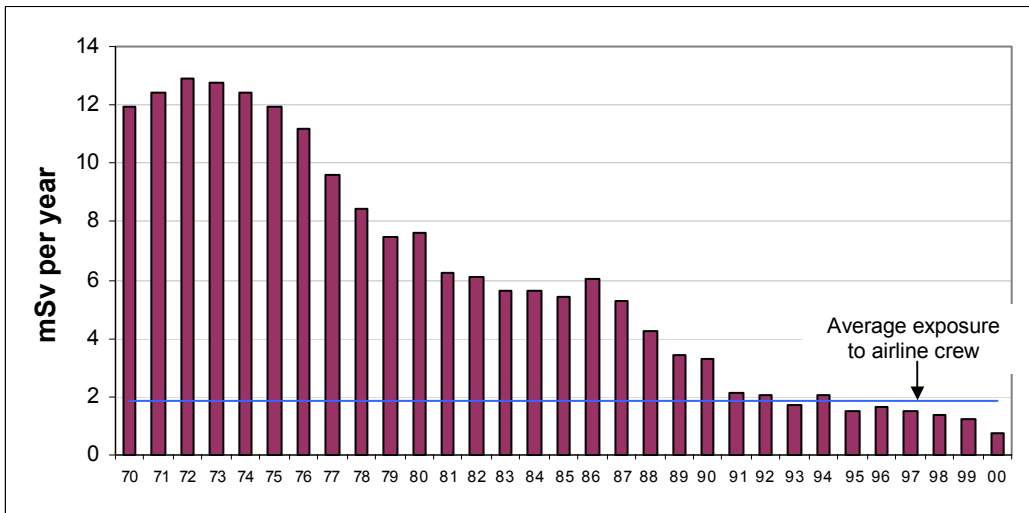


FIG 4: Average occupational exposure for employees at BNFL's Sellafield site



*FIG. 5: Liquid Discharges from La Hague site.*





**Annex II**  
**NATIONAL REPORTS**



# CURRENT STATUS OF SPENT FUEL MANAGEMENT IN CHINA

## Yuansong Liu

China National Nuclear Corporation,  
Beijing, China

### Abstract

The administration organization and the laws and regulations on spent fuel management in China are described at first. In the year of 2000, CNNC and GNPJVC signed a contract of take-over mode for spent fuel, in which CNNC will take-over the spent fuel annually since 2003. In the year of 2001, CNNC and QNPC signed another contract of take-over mode for spent fuel. Transportation by road is the realistic selections of the transportation of the spent fuel from the nuclear power stations to LNFC. As for spent fuel management, the construction of CNNC's reprocessing pilot plant is going on and R&D of spent fuel management are launching continuously.

## 1. THE ADMINISTRATION ORGANIZATION DEALING WITH SPENT FUEL MANAGEMENT IN CHINA

- The State Environment Protection Agency (SEPA) regulates issues of environmental protection caused by spent fuel management in China for. The National Nuclear Safety Administration (NNSA), subordinated to SEPA, regulates for nuclear safety of spent fuel management. SEPA and NNSA issues authorization for environmental protection and nuclear safety during the periods of siting, design, construction, commission, operation and decommission of facilities of spent fuel management. Their detail responsibilities are to approve the safety analysis report, the environmental impact assessment report, the QA program during construction, commission and operation and the emergency plan in the period of commission and operation , etc.
- The China Atomic Energy Authority (CAEA) undertakes international cooperation and exchange in nuclear field as the representative of Chinese government and is the competent authorities of nuclear industry. It is in charge of approving R&D programs of spent fuel management, and organizing the formulation of relevant national regulations, criteria, standards and requirements of spent fuel management. It issues authorization for physical protection of facilities of spent fuel management. Its SSAC (State's system of accounting for and control nuclear material) is to account for and control nuclear material in China and provide the essential basis for the application of IAEA.
- The China National Nuclear Corporation (CNNC) takes charge of siting, design, construction, commissioning, operation and decommissioning of facilities of spent fuel management. CNNC takes the ultimate responsibilities for nuclear safety and environment protection etc. depending on Chinese government's regulations, criteria, standards and requirements for nuclear safety, environmental protection, emergency plan, QA program and physical protection of nuclear facilities of spent fuel management.

## 2. LAWS AND REGULATIONS ON SPENT FUEL MANAGEMENT

China's legal system of spent fuel management consists of relevant laws, regulations, national standards (GB), and trade standards (EJ).

## 2.1 National law

The *Environment Protection Act of the People's Republic of China* was approved by the National People's Congress (NPC). The *Atomic Energy Act* and the *Radioactive Pollution Prevention Act* are now being worked out. The *Atomic Energy Act* is a supreme legal document in nuclear field to adjust and promote the atomic energy development in China. The *Radioactive Pollution Prevention Act* is a basic law for spent fuel management.

## 2.2 Administrative regulations by the state council

In October 1986, the State Council issued the *National Regulations on Supervision and Management of Safety of Civilian Nuclear Facilities*.

## 2.3 Rules and regulations by governmental departments

This refers to the rules and regulations issued by the National Nuclear Safety Administration (NNSA) or jointly issued by the NNSA and other department(s) of the State Council. So far, the *Detailed Implementation Rules for the National Regulations on Supervision and Management of Safety of Civilian Nuclear Facilities*, the *Provisions on Safety of Civilian Nuclear Fuel Cycle Facilities*, the *Environmental Policy on Intermediate and Low-level Radioactive Wastes Disposal*, and the *Provisions on Radioactive Waste Management*, etc. have been promulgated in succession.

## 2.4 Guiding documents

The technical specifications and standardization of spent fuel management have been getting replenished and revised.

## 3. SPENT FUEL ARISING IN CHINA

Up to now, there are four nuclear power units with a total capacity of 2700MWe in China. About 60 tHM spent fuels are produced this year and about 80 tHM spent fuels will be produced from next year. About 400 tHM spent fuels are stored in the reactor pools. Moreover, other eight units with a total capacity of 6000MWe are under construction. It is estimated that, after 2005, about 199 tHM PWR spent fuels and 198 tHM CANDU spent fuels will be produced each year.

## 4. CONTRACTS OF TAKING-OVER SPENT FUELS FROM GNPJVC AND QNPC BY CNNC

CNNC and Guangdong Nuclear Power Joint-Venture Co.(GNPJVC) signed a contract of taking-over spent fuels in March of 2000 and CNNC and Qinshan Nuclear Power Co.(QNPC) signed a contract of taking-over spent fuels in the year of 2001. According to the contract with GNPJVC, CNNC will take-over the spent fuel annually from the year of 2003 to the year of 2015. The initial fuel assembly enrichment is less 4.6%, the maximum burn-up is less 55GWd/tHM and the spent fuels are at the reactor pool for 8 or more than 8 years. In the contract with QNPC, The initial fuel assembly enrichment is less 4.0%, the maximum burn-up is less 45GWd/tHM and the spent fuels are at the reactor pool for 5 or more than 5 years.

In the two contracts the reprocessed Pu, U and all fission products will remain in CNNC. Both sides of the contracts agree that the proprietary, the risk and the nuclear responsibility of the spent fuel will shift to CNNC after spent fuel is hooked on the vehicle of CNNC. CNNC will be responsible for applying for licenses or authorizations for transportation, interim-storage and reprocessing of the spent fuel from China regulatory authority.

## 5. TRANSPORTATION OF SPENT FUEL

### 5.1 Regulatory system

The Chinese Regulations for the Safe Transport of Radioactive Materials—GB11806-89, which technical index is as same as IAEA SS No.6 (1985), was issued on November 21, 1989 and the Chinese Regulations of Surveillance and Management for the Safe Transport of Radioactive Materials is waiting for approval .

As for transportation of spent fuel, NNSA is responsible for approval of certificate of transportation, spent fuel package-design and compliance of certificate (COC), SEPA for approval of Environmental Impact Report and the Ministry of Public Security and CAEA for the physical protection of nuclear material. CNNC is responsible for transportation and procurement of flasks.

### 5.2 Preparation work

Everclean Environmental Engineering Corp., subordinated to CNNC, is in charge of transportation of spent fuel in China. NAC(U.S) has obtained the bid of two casks for transport of spent fuel of GNPJVC. The two casks is being fabricated in Spain. The quality assurance program of transportation of GNPJVC's spent fuel has been worked out and is waiting for approval. The construction of railway terminal for receiving the spent fuel is nearly finished at Lanzhou Nuclear Fuel Complex(LNFC).

### 5.3 Realistic scheme

Two shipments of spent fuel assemblies of a research reactor from China Institute of Atomic Energy in Beijing to LNFC were carried out by road respectively in July 1995 and June 1996. It has shown that transportation by road is safe in China. Feasibility studies on road transportation of spent fuel from Daya Bay Nuclear Power Station and Qinshan Nuclear Power Station to LNFC have been made and the route have been investigated. And so the spent fuels from Daya Bay Nuclear Power Station and Qinshan Nuclear Power Station will be transported by road, but about 10km distance of rail in the scope of LNFC is used for transfer of casks from road trailer to railcar by LNFC's crane and be introduced into fuel receipt building by a railcar.

## 6. STATUS OF SPENT FUEL MANAGEMENT

### 6.1. Centralized wet storage facility

Spent fuels produced by nuclear power plants should be temporarily stored at on-site pools for at least 5 years in China, and then they will be transported to a centralized wet storage facility

for interim storage and to be reprocessed finally. At present, the centralized wet storage facility with a capacity of 550 tHM at Lanzhou Nuclear Fuel Complex (LNFC), as a part of the pilot reprocessing plant, is in cold commission and is expected to receive spent fuels in Oct. of 2003.

## 6.2. Construction of pilot reprocessing plant

A pilot reprocessing plant is under construction at LNFC. The functions of the plant are: 1. to demonstrate process, equipments and instrumentation for reprocessing of spent fuel from nuclear power plant under hot conditions; 2. to accumulate experience on design, construction, commission and operation for a commercial reprocessing plant in future; 3. to supply plutonium for MOX fuel; etc.

The main reprocessing facility of the pilot plant has the maximal flow rate of 400kgHM/day with a modified PUREX process. The completion rate of construction of the main reprocessing facility is over 90% now. The pilot plant will avail itself a continuous production capacity of 100 tHM/ y after reprocessing 50 tHM spent fuels for a project of R&D and being replenished.

## 6.3. Treatment and disposal of i/llw

### 6.3.1 Disposal of ILLW by hydraulic fracture

China began the studies on disposal of ILLW by hydraulic fracture since 1980s. After studies of geology and hydrogeology for a long time and demonstration tests of hydraulic fracture with radioactive isotope tracer for many times, the way of hydraulic fracture for disposal of ILLW is proved safe, reliable and comparatively economic in China. A disposal project has been put into operation since 1993.

### 6.3.2 Treatment of ILLW by bulk pouring cementation

The construction of bulk pouring cementation plant has been completed. A solidification and disposal test for simulated liquid wastes has been carried out. The trial run for real liquid wastes is being prepared recently.

### 6.3.3 Near-surface disposal of I/LSW

Following the policy of regional disposal of I/LSW in China, several national regional I/LSW disposal sites have been set up, such as the northwest I/LSW repository and Guangdong Beilong repository. These sites are run independently by the China Nuclear Everclean Corp..

## 6.4. Treatment and disposal of HLW

### 6.4.1 Vitrification of HLLW

Cold rig tests of vitrification have been completed by German technology. ***Now the preparation work is being done to put the establishment of a vitrification workshop under an authorized plan.***

## 6.4.2 Deep geological disposal of HLW

Studies on HLW disposal were started in China in 1980s. And a plan for deep geological disposal of HLW was mapped out, which consists of four stages, i.e. technical preparedness, geological research, on-site test and construction. From 1985, great progress has been made in the field of HLW research. Siting for repository and underground laboratory, and the feasibility study has been completed. The northwest region of China was primarily determined as one of the most likely candidate sites. Two wells as deep as 700 m and 500 m respectively were drilled in the above-mentioned region for carrying out multi-disciplinary research including hydrology and geology.

## 6.5. Criticality experiment lab

A project of modern criticality experimental lab. has been applied by CNNC to COSTIND. The new lab. will provides a experimental platform to solve criticality issues in the nuclear fuel cycle for future, including in the field of spent fuel treatment and management, in China. Now CNNC are waiting for approval.

## 7. R&D OF SPENT FUEL TREATMENT

### 7.1. Partitioning and transmutation process

Institute of Nuclear Energy Technology of Tsinghua University developed an innovative a mixed trialkylphosphine oxides (TRPO) process to remove transuranium elements from HLLW in 1992 in the world. Now the institute has carried out a hot test of total partition with genuine HLLW, which used TRPO process for separating TRU and  $^{99}\text{Tc}$ , CES (crown ether) process for removal of  $^{90}\text{Sr}$ , and KTiFC (potassium titanium hexacyanoferrate) ion exchanger process for segregating  $^{137}\text{Cs}$ . The decontamination factor for total activity is 588, that for  $^{99}\text{Tc}$ ,  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  is 125, >2500 and >200 respectively.

Experiments for separation of trivalent Am from Lns (from La to Gd) have been done with Purified Cyanex 301 Extraction. The decontamination factor of Am from Lns is higher than 106.

*R&D on partitioning is also launching in the Institute of Atomic Energy of China of CNNC. They use podamine as an extractant to remove TRU from HLLW.*

### 7.2. Advanced purex process and integrated separation process for reprocessing

Study on improved or enhanced Purex process is going on to realize a single solvent extraction cycle and salt-free in the Institute of Atomic Energy of China (IAEC). An ideal integrated reprocessing process is being studied also in IAEC, which considering the separation of U Pu, long lived radionuclides and strong  $\beta$   $\gamma$  fission product in a reprocessing process of spent fuels.



### 7.3. Mox

An experimental fast reactor with a capacity of 65MWt (20MWe) is under construction. According to a plan, plutonium from the pilot reprocessing plant should be used for MOX fuel of the FBR and CNNC is going on R&D of MOX for FBR.

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# STATUS OF SPENT FUEL REPROCESSING IN FRANCE

**M. Giroux**

COGEMA - DCDI/DM,  
Velizy, France

## Abstract

France made the choice of a closed nuclear fuel cycle from the start of its nuclear program and developed reprocessing-recycling capacities accordingly. This choice was confirmed in the nineties, following the Nuclear Waste Act of December 30<sup>th</sup> 1991. Presently spent fuel is reprocessed and recovered fissile materials are recycled in 900 MWe PWR reactors. By October 2002, more than 18 000 t of LWR spent fuel and 18000 t of GCR fuel had been reprocessed in French facilities, while more than 1200 tHM of MOX fuels had been fabricated in Europe. Reprocessing and MOX fuel fabrication are performed for French and foreign utilities. Safety and waste minimization are a constant objective. Other targets for the medium term are further improvements in MOX fuel performance and use and in waste management. For the longer term, research on new scenarios for enhanced waste management are actively pursued in the framework of the 1991 Act.

In France, reprocessing and recycling are mature industrial technologies, which address environment preservation and non-proliferation concerns.

## 1. FOREWORD

France made the choice for a closed cycle very early on in the development of its nuclear program. This strategic choice was driven with the aim of increasing the country's energy independence, while available domestic uranium resources were in limited amount. This goal has been achieved. Since the beginning of the nineties, nuclear power has been fulfilling about three-quarters of the electricity demand in France, a sizeable contribution to the decrease in foreign supply dependency for its energy requirements. An added benefit is the avoided emission of about 400 Mt of CO<sub>2</sub> a year, thanks to the nuclear energy contribution.

In 1991, taking into account the fact that a final disposal site of HLW was yet to be selected, the French Parliament adopted a law (the Nuclear Waste Act of December 30<sup>th</sup> 1991) which targets 2006 for the approval of a national policy concerning the future of high-level and long-lived radioactive waste.

In 1997 the French Government confirmed the French nuclear policy with the following basic guidelines:

1. a key role for nuclear energy generation,
2. the supply of reprocessing and recycling services for national and international utilities,
3. the recycling of plutonium in EDF 900 Mwe PWR,
4. the closing down of Superphenix (FNR 1250 MWe) and the restart of Phenix (FNR 250 MWe) two reactors which were part of the recycling scheme.

ANDRA, the agency in charge of waste repositories, CEA (Atomic Energy Commission) in charge of research, COGEMA operating the reprocessing and recycling capacities and EDF, the utility, all adapted their strategies to this new deal.

ANDRA is operating the Soulaïnes low level waste disposal facility. In 1999, ANDRA was authorized to build in the Bure area an underground research laboratory for HLW and ILW disposal.

CEA is carrying research on advanced nuclear technologies, including those related waste management as required by the 1991 Act.

COGEMA has the capacity to reprocess 1700 t/year spent fuel in its La Hague plant and to market about 180 t/year of MOX fuel to French and foreign customers.

EDF, operates 58 reactors for an installed capacity of 63 GWe net. They generated around 400 TWh net in 2001, contributing to more than three quarters of the total electricity produced in France. About 1100 t of spent fuel are unloaded each year.

FRAMATOME ANP builds reactors and supplies UO<sub>2</sub> fuels assemblies.

ASN, the nuclear safety Authority, is in charge of the safety controls.

## 2. BACK END OF FUEL CYCLE IN FRANCE

### 2.1. Recycling

In France, spent fuel reprocessing takes place in order to recycle the fissile materials recovered and to condition the final residues. EDF's strategy is to adapt the spent fuel reprocessing rate to the recycling capability of its licensed reactors in order to avoid the stockpiling of separated plutonium. Initially plutonium was recycled as MOX fuel in FNRs. Today MOX recycling is limited to LWRs as the FNRs deployment is mothballed, following the closing down of the Superphenix prototype. Reprocessed uranium recycling has been demonstrated in two reactors at the Cruas NPP.

Reprocessed uranium is converted in COMURHEX plants at Pierrelatte, either as U<sub>3</sub>O<sub>8</sub> for interim storage, or as UF<sub>6</sub> for re-enrichment abroad. The enriched UF<sub>6</sub> is then converted in the FBFC Romans plant (capacity 150 tHM/y) as UO<sub>2</sub> fuel. Since the mid eighties, EDF has been loading reprocessed uranium fuel assemblies in Cruas reactors .

The first plutonium recycling in France took place in the Rapsodie research reactor about forty years ago. The Cadarache Plant has been fabricating MOX fuel since 1961, starting with Rapsodie fuel. Cadarache then fabricated MOX fuel for Phenix and Superphenix fast reactors. In 1987, the Cadarache plant was upgraded to produce MOX fuel assemblies for LWR. Its 40 t/y capacity was reached by the mid nineties.

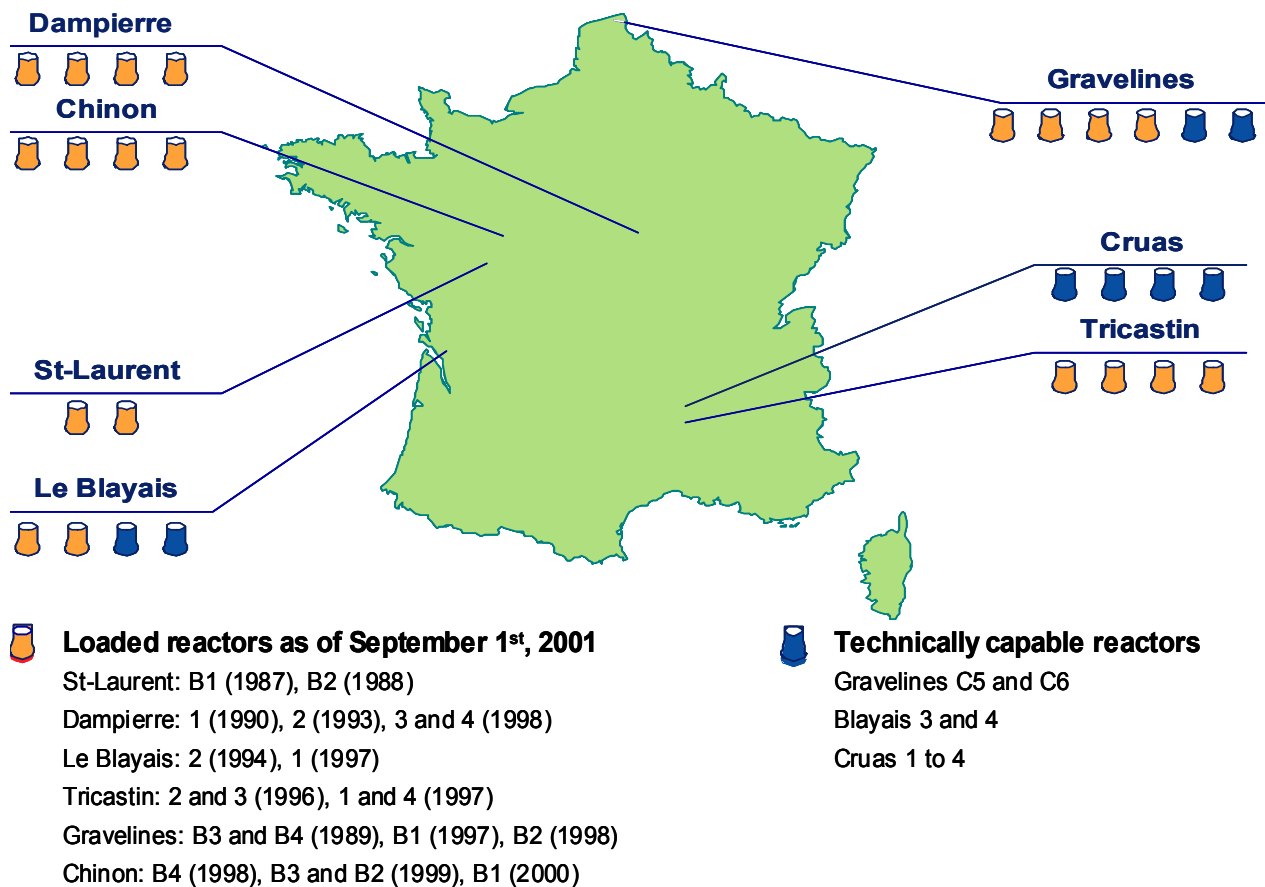
To face the growing demand for MOX fuel, the design of a new plant, the MELOX facility, was initiated in 1986. The Advanced MIMAS process was chosen for the initial stage of the powder preparation, as it allows to recycle in the process powders or sintered pellets recovered along the production line. The construction work started in 1990 and the plant was commissioned in March 1995. The plant reached its current production license of 100 tHM/y by 1997. In a March 1998 test campaign, the higher production potential of the MELOX facility was demonstrated with 20 t of MOX fuel produced during one month. The full benefit of this potential will be utilized once the CADARACHE plant is closed down as requested by the French Safety Authority and its production transferred to MELOX. The license to increase the production of MELOX should be granted by 2003.

MOX fabrication lines are qualified by French and foreign fuel vendors: FRAMATOME ANP, JNFL, MHI and NFI.

In France, today, EDF loads MOX fuel in 20 of its 900 MWe PWR's (Figure 1). As of December 31<sup>st</sup> 2001, the total quantity of MOX fuel delivered to EDF had reached 1648 assemblies. MOX fuel loaded in French PWR's is licensed for an average burn-up of about 40 GWd/tHM while UO<sub>2</sub> fuel is licensed for higher burn-ups. Studies on MOX fuel in core behavior are undertaken to further optimize its use and to allow it to be licensed to reach an average burn-up equivalent to UO<sub>2</sub> fuel.

In France, experimental MOX elements are being tested up to 57 GWd/tHM with a target of 70 GWd/tHM in mind. Elsewhere in Europe, commercial MOX fuel has already been licensed to reach burn-ups higher than 50 GWd/tHM.

Each year, EDF loads 100 t of MOX fuel in its reactors. The amount of plutonium needed requires the yearly reprocessing of about 800-900 t of spent fuel. Unloaded spent fuel on top of that is stored in COGEMA's pools at the La Hague plant.



In France : 58 reactors in operation, 20 « moxidized » reactors

FIG. 1. French MOX use in reactors.

## 2.2. Spent fuel interim storage and transport

In France, all spent fuel assemblies unloaded from commercial reactors are stored for a few years cooling time in pools at reactor site. They can then be transported to the pools of COGEMA's reprocessing plant at La Hague for further treatment. The bulk of it, in the form

of standard UO<sub>2</sub> spent fuel, is reprocessed after a short additional cooling period. Spent MOX fuel and spent fuel made of reprocessed uranium are presently stored.

Authorized capacity for COGEMA's pools at the La Hague plant is about 14,000 tHM. Licensing is under way to increase the authorized capacity. This large capacity ensures the safe storage of customers' spent fuel before reprocessing.

Some special fuel elements, which will not be reprocessed in the near future, are stored at Cadarache in CASCAD, a dry modular interim storage designed by SGN.

Transports of French and foreign spent fuel to COGEMA's reprocessing plant at La Hague are performed by Cogema Logistics. The company has acquired a large experience in metallic casks and dry transfer technologies, allowing it to develop dry metallic storage & transport casks and to market concrete cask technology to foreign customers.

### **2.3. Reprocessing**

In France, GCR's (Natural Uranium Graphite Gas Reactors), FBR's, PWR's and RTR's (Research and Test Reactors) have been or are in operation. The various spent fuel coming from these different types of reactors have all been successfully reprocessed in the past 40 years in 3 reprocessing plants: UP1, UP2 and UP3. The experience gained by the French industry has benefited foreign customers. More than 18,000 tHM of GCR fuel and 18,000 tHM of LWR fuel have been reprocessed to date.

#### UP1 (COGEMA Marcoule)

The UP1 reprocessing plant, located in Marcoule (Rhône Valley), started operations in 1958. Civilian operations in the plant began in 1965 with the reprocessing of spent nuclear fuel from French and foreign GCR reactors. The plant, whose operations are based on the PUREX process, was managed by the CEA until 1976 and by COGEMA after 1976. In 1978 the first French vitrification facility (AVM) was added to the UP1 plant to stabilize HLW waste.

UP1 reprocessed the last GCR fuel assemblies in late 1997 and was shut down in 1998. An Economic Interest Group for the management of all the decommissioning operations was set up by CEA, COGEMA and EDF. The closing down phase (MAD: mise à l'arrêt définitif), including advanced rinsing and removal of nuclear material, was completed in 2002 and total dismantling should be accomplished within 30 years, funded mainly by EDF and CEA.

#### COGEMA La Hague (present licence status)

UP2 and UP3 reprocessing plants are located on the same site at La Hague (Normandy). UP2 was commissioned in 1966 to reprocess GCR fuel. In 1976 a new head-end (HAO) has been added to UP2 to reprocess LWR fuel. In 1994 this plant (also known as UP2-400) was completely upgraded to treat the growing quantity of French PWR spent fuel. This new facility is known as UP2-800. In 1989, the UP3 plant was commissioned to reprocess foreign spent fuel. Each plant is operating under its own license.

In 1999 COGEMA submitted to the French authorities a request to modify the existing operating licenses. The public inquiry completed in 2000 endorsed the proposal to increase the licensed capacity of each plant from 800 tHM to 1,000 tHM while limiting the site

production to 1700 tHM per year. The governmental licence approval is expected in the coming months. This new La Hague Plant will be operated, no more on two dedicated plants, but like one multi services plant.

COGEMA's La Hague plant reprocessed 17,244 tHM (as of January 1<sup>st</sup>, 2002) of which 6,904 t in UP3 and 10,340 t in UP2. UP3 production was dedicated to foreign customer's fuel while the bulk of the UP2 input is domestic fuel. Details are provided in Table I and figure II.

Table I UP2/UP3 cumulated production (as of January 1<sup>st</sup>, 2002)

	Total reprocessed MTU
France	8272
Germany	4470
Japan	2944
Switzerland	619
Netherlands	268
Belgium	671
<b>Total</b>	<b>17244</b>

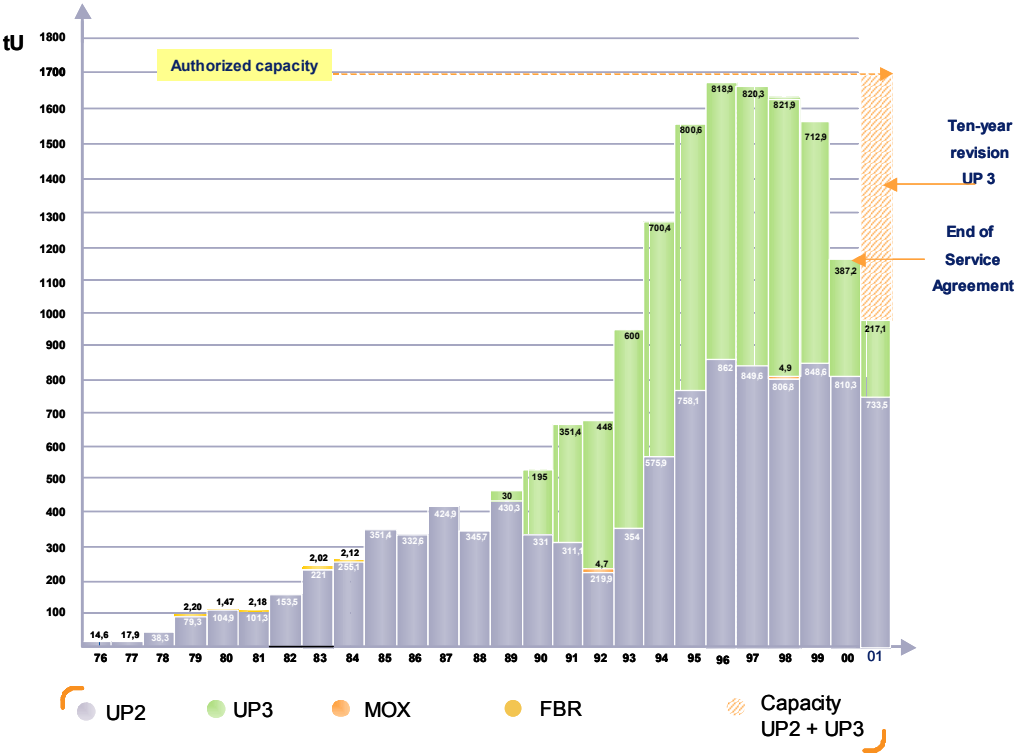


FIG. 2. Reprocessing Performance : COGEMA La Hague Plants.

This high-performance record has been achieved while minimizing the impact on the environment and public health. See figures 3 and 4.

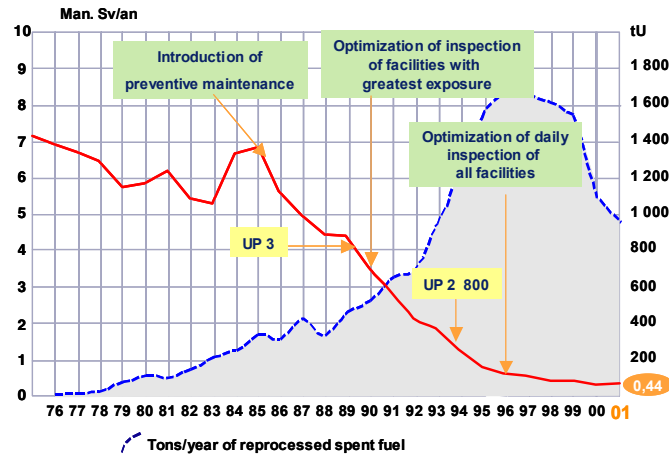
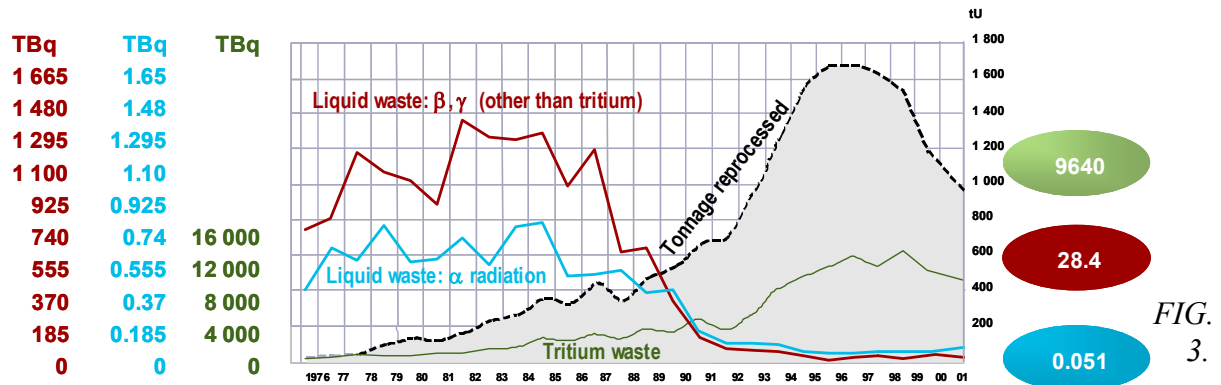


FIG. 3. Total exposure for UP2+UP3 plant.



UP2 and UP3 plant release into the sea.

## 2.4. Plant improvements

UP2 and UP3 plant operations are based on the PUREX process. Their construction was decided in the seventies and massive production took place in the nineties, during which time plant operations were continuously improved. The uranium and plutonium separations factor has now reached 99.88%, leaving in the vitrified matrix only about one gram of plutonium for each kilo recovered, and contributing to the minimization of waste volume.

### New Effluent Management

Thanks to the extraction process performance levels, the volume of low-and medium activity effluents produced are much below the estimates made during the facilities' design phase. The new management allows better sorting of the effluents, based on their radioactive and chemical content. After evaporation, concentrates containing most of the effluents' radioactivity are now vitrified with high-activity wastes without increasing the volume of glass produced (see Table II and Figure 5). Thanks to this process, there is no more ILW to bitumenize prior effluent discharge to the sea, as was previously the case.

Table II Reprocessing by-product activity

Percentage of activity	$\alpha$	$\beta \gamma$ (except tritium)
Vitrified waste	99.5	97.6
Hulls and end-fittings	0.4	2.3
Total process waste	99.9	99.9
Technological waste	0.1	0.1

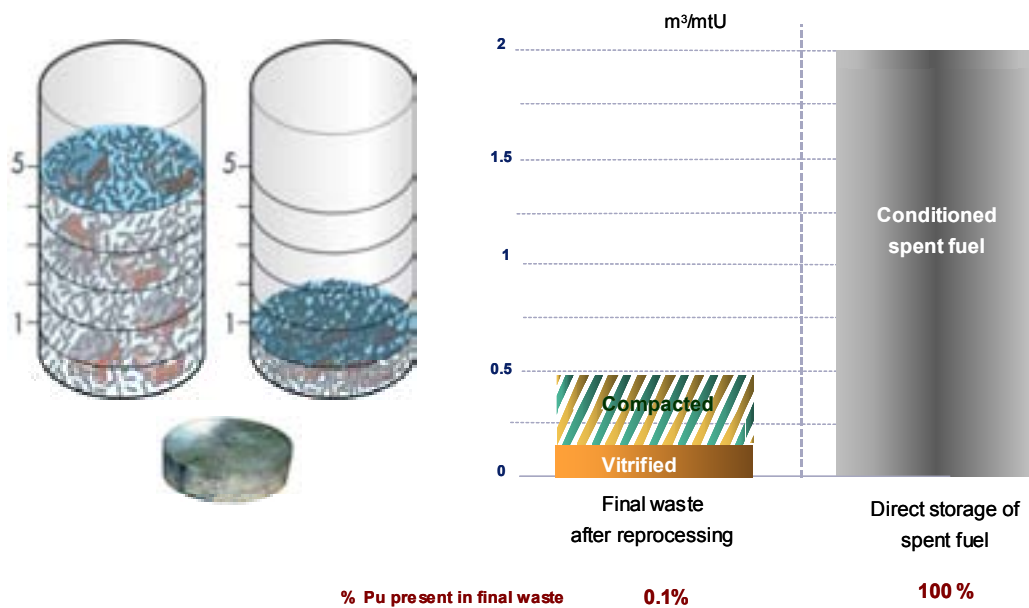


FIG. 5. Volumes of final residues conditioned in UP3.

## 2.5. Hulls and end fittings compaction (ACC and ECC UNITS)

The ACC compaction facility, which started operation in 2002, allows four-fold reductions in waste volume resulting from fuel reprocessing. Structural parts of fuel elements (hulls and end-fittings) with long-lived radio-nuclides, which were conditioned in grout until 1995, are now compacted and packaged in universal canisters also used for vitrified waste. Each canister is filled with 5 to 7 discs according to their thickness, in order to produce less than 1.5 Universal Canister per ton of reprocessed spent fuel. Up to 2,400 canisters yearly can be produced at the ACC.

The Universal Canisters once produced in the ACC are sent to the ECC interim storage facility before final shipment to COGEMA customers.

## 2.6. Vitrification

COGEMA has more than 20 years industrial experience using hot crucible melter technology in three different vitrification facilities: AVM in Marcoule, R7 and T7 (respectively



commissioned in 1989 and 1992) at the La Hague site. By 2000, COGEMA's vitrification facilities had produced over 10,000 canisters containing about 4,000 tons of vitrified HLW, more than three quarters of which were from R7/T7 (3 G C of  $\beta$  and  $\gamma$  activity).

After ten years of operation in R7 and T7, each with a nominal capacity of more than 550 glass canisters per year, the life time of the melters exceeds design basis values by a factor of two. The feedback from active operation and R&D results have led to further improve the process in areas such as glass characteristics, associated technologies, operation and maintenance.

To broaden the performance range of the vitrification process, CEA and COGEMA, with SGN, have developed the use of the cold crucible melter (CCM) technology. It leads to a virtually unlimited equipment service life and allows a greater flexibility with respect to waste composition. The high specific power directly transferred to the melt by induction helps in adjusting glass temperatures.

To increase throughput and flexibility (with respect to liquid feed as well as solid feed acceptance criteria), the ACCM (advanced CCM) variant was developed. This design allows (in a single step process) to directly feed the melter with solid or liquid waste flow, while improving the glass throughput capacity.

## **2.7. Broadening the range of operation**

The reference fuel considered for the design and start-up of the UP3 and UP2-800 plants was uranium oxide fuel initially enriched to 3.25% in  $U_{235}$ , with a burn-up of 33 GWd/t and cooled for 3 years. However, right from the very beginning of the La Hague site, COGEMA was authorised, based on supporting studies, to use the HAO facility to reprocess other types of fuels. The UP3 and UP2-800 plants today process PWR and BWR fuels initially enriched to 3.7% in  $U_{235}$ , with burn-up reaching 45 GWd/t, cooled for a minimal 4-year period. COGEMA's La Hague reprocessing plants are technically capable to reprocess the new generations of spent fuels<sup>9</sup> without any significant modifications.

The new licences to be published end 2002 will enlarge the scope of Reprocessing (Burn up, type of fuel elements etc....).

## **3. CONCLUSIONS**

France has made the choice of a closed nuclear fuel cycle from the start of its nuclear program and developed reprocessing-recycling capacities accordingly. The industry has matured and the facilities, which have been continually improved, continue to offer French and foreign customers reliable services in accordance with environmental protection and safety principles. In addition to research efforts designed to continuously improve the processes in existing facilities (in terms of performance, reliability and economics), a comprehensive R&D programme was launched under the guidelines of the Nuclear Waste Act of December 30<sup>th</sup> 1991. Addressing three domains (separation-transmutation, disposal in deep geological formations, conditioning and long term interim storage), this programme intends to assess a broad set of possible techniques and strategies to further improve short- and long term waste management.

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<sup>9</sup> Higher burn-up UO<sub>2</sub> and MOX used fuels.

## STATUS OF SPENT FUEL TREATMENT IN JAPAN

### T. Koyama

Japan Nuclear Cycle Development Institute (JNC),  
Tokai Works, Waste Management and Fuel Cycle Research Center,  
Tokai-mura, Ibaraki-ken, Japan

#### Abstract

Recent changes concerning the treatment of spent fuel in Japan are reviewed. The JNC Tokai Reprocessing Plant totally processed 1,000 t of spent fuel. The construction of the JNFL Rokkasho reprocessing plant is proceeded and now scheduled to start hot operation in 2005. The JNC-led feasibility studies of Phase II on the commercialized fast reactor (FR) cycle system are underway. Both aqueous and dry reprocessing technologies are under intensive development for establishing FR cycle.

### 1. INTRODUCTION

On September 30th of 1999, the criticality accident occurred at Tokai plant of JCO Co., Ltd. in Tokai-mura. Two workers died even after extensive medical treatment. The accident gave a serious and negative effect on the social environment for utilization of nuclear energy. All of nuclear-related organizations worked to recover public confidence, and Japanese policy to proceed with nuclear power has been maintained. The report reviews the status of the nuclear fuel cycle in Japan as of October 2002, and the changes from the situation described in the previous report in 2000 [1].

### 2. LONG TERM PROGRAM ISSUED IN 2000

Under the circumstance that was influenced by the aftermath of the criticality accident, "Long term Program for Research, Development and Utilization of Nuclear Energy" was issued in November 2000 by the Atomic Energy Commission, Japan [2]. The Long term Program Council discussed with opening to the public all of the meetings including that of its six subcommittees for transparency. Concerning to the nuclear fuel cycle industry, the following is the important direction of the Program:

- MOX fuel utilization in light water reactors (LWR) is being steadily carried out.
- Future MOX fuel that will contain plutonium recovered in Japan is being manufactured at a domestic fabrication plant.
- All spent fuel will be domestically reprocessed in principle.
- The private nuclear operators are expected to make steady progress with the construction and operation of the Rokkasho-mura facilities including Rokkasho Reprocessing Plant of Japan Nuclear Fuel Limited, JNFL.
- The Japan Nuclear Cycle Development Institute (JNC) should conduct demonstration tests on reprocessing for high burn-up fuel, spent MOX fuel from LWR at its Tokai Reprocessing Plant.
- The next reprocessing plant is expected to reprocess not only spent uranium fuel but also high burn-up fuel and spent MOX fuel from light water reactors, besides fuel from fast breeder reactors based on future research and development. The new plant project will be discussed around 2010.
- Commercial operation of storage facilities is being prepared by 2010 based on the law of intermediate storage that was enacted in 1999.

- And the direction for research and development of FBR cycle technology is summarized as the following:
- FBR cycle technology has some of the greatest potential among technological options.
- First, the option that is based on MOX fuel and sodium cooling should be assessed.
- The prototype fast breeder reactor "Monju" is considered as the core of research and development activities for FBR cycle technology, and its operation will be quickly resumed.
- In view of the technical diversity of FBR cycle technology, the research and development projects will be given sufficient flexibility and will investigate multiple options.
- The ongoing "Feasibility Study on a Commercialized FBR Cycle System", undertaken by the Japan Nuclear Cycle Development Institute with the cooperation of electric utilities and other interested parties, will be continued to examine such aspects as reactor, reprocessing and fuel fabrication technologies.

At the present, the parties relating nuclear energy industry and also universities are continuing their activities on the basis of the Long term Program.

### 3. LWR FUEL REPROCESSING

#### 3.1. Tokai Reprocessing Plant, JNC

Since April 1997, just after the accident of bitumen solidification facility in Tokai Reprocessing Plant, the plant stopped its operation. In July 2000, Government approved the change of the license for reprocessing; namely, construction of Low-level Waste Storage Facility (LWSF) and treatment of the uranium solution resulted from the criticality accident of JCO. After the local government of Ibaraki prefecture and Tokai village admitted the restart of plant operation in November 2000, it started for treatment of the active solution of the JCO accident, and then also the spent fuel from "Fugen" reactor of JNC. Four campaigns were completed and the amount of spent fuel reprocessed reached 1,000 t from the beginning of the operation in 1977. The 15th periodical plant check is now underway. The summary of the operation is shown in Table I.

Table I Summary of recent operation of Tokai Reprocessing Plant

Campaign	Period	Amount of spent fuel reprocessed, t	Number of Vitrified waste produced
00-2	Nov '00–Dec '00	2.8	6
01-1	Mar '01–Jun '01	23.9	23
01-2	Oct '01–Dec '01	12.1	8
02-1	Mar '02–Jul '02	22.3	–

After the issue of the Long term Program by the Atomic Energy Commission of Japan, JNC revised its program as the "Medium- and Long term R&D Project and Management Plan" in July 2001 [3]. The program describes the long term perspective and the plan for the next five years. The following is the important items related to reprocessing LWR fuels.

- Spent fuel reprocessing including "Fugen" MOX fuel will be continued.
- Tests using fuels of high burn-up, approximately 32 GWD/t, will be carried out.

- Expertise and operation/maintenance experience will be provided for JNFL in order to promote the establishment of the commercial reprocessing plant.

Concerning to the third item of supporting JNFL, JNC and JNFL made a contract on the technical support in July 2002. The contract is valid for July 2005, namely the end of the test operation.

### 3.2. Rokkasho Reprocessing Plant, JNFL

The construction of fuel receiving and storage facility was completed in 1999. The facility can store 3,000 t uranium of spent fuel. About 640 t of spent fuel is now stored. The summary of the status is shown in Table II. The construction work of the main reprocessing plant will be completed in July 2005. As of the end of July 2002, about 89 % of construction work was already made.

The water-flow testing was started at the head end building of the plant in April 2001. The testing steps are the following; chemical test, uranium test and active test using spent fuel. The completion of test and construction is scheduled in July 2005. JNC will technically support them based on the contract mentioned above.

A water leak from the storage pool for PWR spent fuel was found in 2001. The fuel that had been stored were moved to one of other pools, and the point of the leakage was, at first, regarded as some part of the welding line by initial investigation. However, recently, it was found that identified holes were not penetrating the stainless wall. The investigation will be continued.

Table II Summary of spent fuel received at Rokkasho Reprocessing Plant, August 2002

Type	Total amount of spent fuel received, t	Number of fuel assemblies received
BWR	359	2,050
PWR	288	660

## 4. RESEARCH AND DEVELOPMENT FOR FUTURE REPROCESSING

### 4.1. “Feasibility Study on Commercialized Fast Reactor Cycle Systems”

#### 4.1.1 Outline of the feasibility study

The requirements for the future fast reactor (FR) cycle are considered to be as follows: 1) safety of reactor and fuel cycle (safety-in-design), 2) economic competitiveness as an energy system to a level at least comparable to the future LWRs, 3) efficient utilization of uranium resources, 4) reduction of environmental burden, 5) enhancement of nuclear non-proliferation. The detail is summarized in Table III.

In order to establish a commercialized FR cycle system that meets the above objectives, the “Feasibility Study on Commercialized Fast Reactor Cycle Systems” was started in Japan at the beginning of July 1999 [4]. The feasibility study is being carried out in four phases as shown in Figure 1. The Phase I study was started in July, 1999 with the participation of parties concerned in Japan, i.e., the electric utilities, CRIEPI (Central Research Institute of

Electric Power Industry), and JAERI (Japan Atomic Energy Research Institute). Technical options were reviewed and evaluated, and some candidate concepts for the FR cycle were selected.

During 5 years of the Phase II study, candidate concepts selected in Phase I will be secondary screened based on experimental data and further research. The Phase II study will be concluded within Japanese fiscal year 2005. Through the evaluation, the essential subjects for research will be identified.

After the completion of Phase II study, research and development activities will be carried out for making a conceptual and detailed design study, of course, with experimental study. The results of the study will be checked and reviewed every five years for making decision to proceed to the next step of development. These R&D activities shall aim to make a highly attractive and competitive FR cycle concept by around 2015.

Table III Development target of FR cycle system

Requirement	Technical target
Ensuring safety	<ul style="list-style-type: none"> <li>• Reactor               <ul style="list-style-type: none"> <li>– Prevent the event causing reactor core damage. Assuming that it would happen, it should end naturally without huge mechanical energy discharge</li> </ul> </li> <li>• Fuel Cycle               <ul style="list-style-type: none"> <li>– Take sufficient measures against criticality safety and confining function</li> <li>– Take sufficient measures, taking into account characteristics of using material (chemical activity, toxicity etc.) and process conditions ( operating temperature etc. )</li> </ul> </li> </ul>
Economic competitiveness	<ul style="list-style-type: none"> <li>• Attain economic competitiveness to future LWRs</li> <li>• Cost Targets               <ul style="list-style-type: none"> <li>– Construction Cost: 0.2 million yen/kWe</li> <li>– Reprocessing Cost: 0.27 million yen /kgHM</li> <li>– Fuel Fabrication Cost: 0.16 million yen /kgHM</li> </ul> </li> </ul>
Efficient Utilization of Resources	<ul style="list-style-type: none"> <li>• Variety Utilization using rich neutrons</li> <li>• TRUs burning and LLFP incineration</li> <li>• Recycled LWRs Pu Burning</li> </ul>
Reduction of Environmental Burden	<ul style="list-style-type: none"> <li>• Reduce the amount of radioactive waste for disposal by TRUs burning and LLFPs transmutation</li> <li>• Reduce generation of radioactive waste from plant operation and maintenance dismantling</li> </ul>
Enhancement of Nuclear Non Proliferation	<ul style="list-style-type: none"> <li>• No existing pure plutonium in the FR cycle system</li> <li>• Enhanced Design for physical protection and safeguard</li> </ul>

Table IV Evaluated result of FR System technology in the Phase I study

Reactor System	Reactor Type	Fuel Type		
		MOX	Nitride	Metal
Sodium	Large-scale tank type			
	Large-scale loop type	A	B	A
	Medium-scale modular type	A		
Heavy Metal	Large-scale	B	A	A
	Medium-scale modular type (Pb-Bi)	A		
Gas	Carbon dioxide gas cooled pin type fuel			
	Helium gas cooled pin type fuel	B	A	C
	Helium gas cooled coated particle fuel	A		
Water	BWR system			
	PWR system	A	–	–
	Supercritical pressure	A		
Small-scale	Sodium-cooled	B	A	A
	Heavy metal-cooled	A		

A: Selected technology for Phase II study

B: Technology to be reevaluated in the Phase II by reviewing results obtained in the other studies

C: Scope of the study in the Phase I except A and B

–: Out of scope in the Phase I

#### 4.1.2 Summary of Phase I study

The Phase I study was concluded in March 2001 [5]. In this section, the important results are reviewed.

The results of study on the FBR system are summarized in Table IV. Sodium cooled large and medium scale reactors are feasible and are expected that they provide possibility to attain economy comparable to the LWR system in the future. Large-scale advanced loop type reactor was chosen as the most promising. As a fuel, MOX and metal form are selected. Among Heavy metal cooled reactors, medium scale Pb-Bi cooled reactor combined with nitride and metal fuel was selected. From gas cooled reactors, helium gas cooled reactor that uses coated particle of nitride was selected. Water-cooled system employing MOX fuel will be further researched, however it is weak in uranium utilization.

Reprocessing system is classified into two groups: aqueous and pyroprocess. The aqueous process is based on the PUREX process and combined with the SETFICS process that is a variation of the TRUEX process for actinides(III) recovery. As pyroprocess, oxide-electrowinning process, metal-electrorefining process and fluoride volatility method were evaluated. As a result, JNC will mainly conduct experimental study of oxide-electrowinning and metal-electrorefining processes.

Table V Evaluated result of FR fuel cycle system technology in the Phase I study

Technology		Fuel Type			
		MOX	Nitride	Metal	
Reprocessing	Aqueous process	A	A	–	
	Pyroprocess	Oxide electrowinning	A	C	C
		Metal electrorefining	A	A	A
		Fluoride volatility method	B	B	B
Fuel Fabrication	Pelletizing process	A	A	–	
	Vibration compaction	Gelation method	A	A	–
		Oxide electrowinning compatible	A	C	–
		Metal electrorefining compatible	A	A	–
		Fluoride volatility method compatible	B	B	–
	Casting	–	–	A	

- A: Selected technology for Phase II study
- B: Technology to be reevaluated in the Phase II by reviewing results obtained in the other studies
- C: Scope of the study in the Phase I except A and B
- : Out of scope in the Phase I

#### 4.1.3 Summary of Phase II study

In the Phase II study on fuel cycle systems (reprocessing systems and fuel fabrication systems), the conceptual design is to be developed, incorporating innovative technologies in the candidate concepts extracted in the Phase I study, and elemental technology experiments are to be performed to clarify the feasibility of the principal technologies and to acquire the quantitative data necessary for screening the candidate concepts.

The framework of research activities of the Phase II is shown in Figure 2. During the first three-year until 2003, research activities will be focused on the design of the candidate concepts and elemental tests of key technologies. Outcomes of the activities as an interim summary will be checked and reviewed, and the research for fiscal 2004 to 2005 will be conducted in order to narrow down to two or three promising concepts for the FR cycle.

##### (1) Aqueous process

For the aqueous process development, the reference process that is based on the PUREX and the SETFICS process is investigated as well as some alternative process. The block flow diagram of the aqueous process is shown in Figure 3. The spent fuel is dissolved into nitric acid solution. The resulted dissolver solution should be highly concentrated in nitric acid and heavy metal for crystalline step that recovers uranium hexahydrate. Accordingly, the dissolving procedure should be improved to attain higher U and Pu concentration within practical dissolution time. In order to accelerate the rate of dissolution, the increase of surface area of the fuel is one of an effective method. The process that consists of powdering an oxide fuel and dissolution with nitric acid has been investigated. The crystallization step helps to adjust the ratio of Pu to U for eliminating Pu partitioning in the PUREX process. The PUREX process recovers a mixed product of U, Pu and Np. The long-lived radionuclides, Am and Cm, are extracted with CMPO–TBP mixed solution from the raffinate of the

PUREX. In the extraction step, rare earth elements are simultaneously recovered due to the chemical similarity. The actinides(III) can be selectively stripped with a solution that contains DTPA and sodium nitrate while light lanthanides is held in the organic phase. A product solution of Am and Cm with a part of lanthanides is obtained. In order to evaluate the feasibility of the chemical process, some experiments were conducted.

The crystallization step shall treat the dissolvor solution containing Pu besides U. The valence of the Pu influences its behavior while U crystallizes as UNH. JNC cooperates with AEA Technology in UK for conducting experiments using U–Pu mixed solution and real dissolvor solution. Figure 4 shows the appearance of UNH crystal recovered. When Pu exists as Pu(IV), Pu was decontaminated with other typical fission products, such as Cs and Eu. On the other hand, Pu(IV) crystallizes with UNH [6]. As far as valency adjustment of Pu is performed prior to crystallization, it was confirmed that Pu can be decontaminated and retained in the solution.

The SETFICS process for actinides(III) recovery requires concentrated salting-out reagent. Decreasing the amount of secondary waste is preferred. Replacing sodium nitrate with a salt-free reagent, hydroxylamine nitrate, was evaluated by an inactive counter-current experiment using mixer-settlers and the possibility of adoption of the salt-free technology for the process was confirmed.

Besides the reference process, some alternative technique were also investigated; the supercritical fluid direct extraction method as the alternative for the PUREX, the amine extraction method as the alternative for the SETFICS, and the extraction chromatography method for the SETFICS and liquid-liquid contactors.

In order to conduct experiments using irradiated fuel, Chemical Processing Facility (CPF) was improved in its cell and glove boxes. The installation and refurbishment work was completed in March 2002. It is scheduled to start hot experiments late this year.

## (2) Pyroprocess

Regarding the oxide electro-winning method, evaluation of safety design, such as rationalization of waste process and identification of countermeasures for accidents concerning electrowinning equipment proceeded, and, among the principal subjects, the elemental technology tests for MOX electrowinning co-precipitation and chlorination dissolution technologies were carried out.

Regarding the MOX electrowinning co-precipitation technology, the electrowinning co-precipitation tests under Ce coexistence which might affect processing efficiency were carried out at RIAR in Russia and U and Pu were confirmed to be recoverable without being greatly affected even in high-concentration Ce conditions, and the possibility of technical feasibility was confirmed.

Regarding the chlorination dissolution technology, appropriate dissolution conditions etc. were identified by the chlorination dissolution basic tests using Fe, Rh, and other elements which are considered to inhibit dissolution or platinum group metals.

Regarding the metal electrowinning method, further rationalization of the apparatus and equipment was achieved by deletion of the chlorine gas system through adoption of the Li re-oxidization method and increase of the processing capacity by increasing the number of



electrodes per set, and, among principal subjects, the elemental technology tests for electrowinning deposit and the Cd cathode treatment were carried out.

In small-scale process feasibility tests which have been carried out by CRIEPI in collaboration with the Institute for Transuranium Elements (ITU) of the EU, the electrowinning refining tests for U-Pu-Zr ternary alloy fuel containing MA (non-irradiated) were performed and fundamental data, such as recovery ratio, were obtained. Also, tests for recovery of Pu by liquid Cd cathode were carried out using U/Pu ratio in salt as a parameter. It was confirmed that recovery of heavy metals at a concentration exceeding the design value (10 wt%) is possible and the possibility of rationalization of the system was confirmed. Moreover, glove boxes are being installed at CPF for Pu tests.

### (3) Fuel fabrication

Regarding fuel fabrication systems, taking integrity with reprocessing systems into account, the conceptual designs of the principal equipment were executed, applying a simplified pelletizing method, a vibration compaction method and a casting method, considering low-decontamination TRU fuel with high heat generation and high dose radiation, and the elemental technology tests were carried out.

Regarding the simplified pelletizing method corresponding to aqueous reprocessing, MOX powder storage hopper was identified to be the portion greatly affected by heat generation due to handling of low-decontamination TRU fuel and it was clarified that heat is removable by attaching fins to the inside of the hopper. Moreover, design evaluation for the main equipment such as turntable-type equipment for removing nitric acid was promoted and the elemental technology tests for molding and powder transportation technologies, which are principal subjects, were carried out.

The lubrication performance tests of the DAI lubricating-type molding machine using MOX powder were carried out and the possibility of feasibility of the molding technology was confirmed on an engineering scale. Also, cold tests were carried out in order to determine the air current powder transportation system and the relation between the various air current transportation methods and the shape variation and molding characteristics of the transported materials before and after transportation was clarified.

Regarding fabrication of low-decontamination TRU fuel, tests for fabricating MOX pellets containing Am in the remote fabrication equipment in cells started and it was confirmed that there were no problems in remote operability. Moreover, the homogeneity etc. of Np in MOX was confirmed by measurement of physical properties of MOX containing Np.

Regarding the vibration compaction method, design evaluation was carried out to realize remote automation in cells as a response to low-decontamination TRU fuel, and it was clarified that degradation due to dripping of solution and re-oxidization of fuel particles could be prevented. Also, in order to improve the compaction ratio, which is one of the principal subjects to be addressed, simulation tests were performed using the simulated granule or UO<sub>2</sub> granule produced by the electrowinning method to clarify the relation between parameters such as particle shape factor and the compaction density, and the possibility of achieving the compaction density of 80 %, which is a design requirement, was confirmed.

In collaborative research with PSI of Switzerland, irradiation tests to confirm the fuel behavior in the case of the vibration compaction method (granule fabrication by the internal gelling method corresponding to the aqueous method) are being prepared. Regarding the

external gelling method, by means of heat-resistance, acid-proof and radiation-proof tests using simulated solutions, temperature and concentration conditions to maintain raw liquid nitric acid for appropriate dripping were clarified. The number of units of injection molding equipment was reduced from five to four for rationalization of the fuel processing procedure and the design of injection molding equipment suitable for remote operation in cells is under way as a response to low-decontamination TRU fuel.

Regarding the injection molding technology, the U-Zr injection tests (about 50 slag/injection) for circa 20 kg engineering scale were carried out by CRIEPI and the operation parameters to satisfy the required specification of the slag product were clarified.

#### **4.2. Recycle Equipment Test Facility (RETF), JNC**

The first phase of the construction was completed in June 2000. The RETF plan is mentioned in the plan of JNC [3]. The facility will remain flexible in order to appropriately reflect the progress being made in the development of FR reprocessing technologies. Further RETF plan will be formulated by around 2003 remaining consistent with the results of the "Feasibility Studies on Commercialized FR Cycle System".

#### **4.3. Research and development funded by METI**

The Institute of Applied Energy (IAE) started research and development on innovative nuclear technology for practical use since 2000. This enterprise collects publicly new promising technologies for study and is funded by the Ministry of Economy, Trade and Industry (METI).

In the 2000 for the first year, reprocessing and partitioning technologies, the "FLUOREX" method for U and Pu recovery and long-lived radionuclides partitioning, were selected for research and development. In the second year 2001, the "Super-DIREX" process utilizing supercritical carbon dioxide was nominated.

The party of Hitachi Ltd., Saitama Univ. and Nagoya Univ. studies on a hybrid reprocessing process that consists of fluoride volatilization and the PUREX process. The FLUOREX process developed to improve economy of reprocessing. A spent fuel decladded is fluorinated to convert uranium oxide into  $UF_6$  and the remained U and Pu is dissolved into nitric acid and processed by the PUREX. The expected decontamination factor for uranium fluoride product is expected as  $> 10^7$  that enables to directly enrich it for further recycling. The technological subjects are those; selective fluorination of uranium, recovery of fluorine and dissolving remained fluoride of U and Pu into nitric acid solution.

The party of Tokyo Institute of Technology, Mitsubishi Material Co. and Japan Atomic Energy Research Institute (JAERI) studies on long-lived radionuclides; Am, Cm and I-129. Recovery of Am and Cm is attained by using soft donor extractants such as  $N,N,N',N'$ -tetraoctyl-3-oxapentane-1,5-diamide (TODGA) and  $N,N,N',N'$ -tetra (methylpyridyl) ethylenediamine (TPEN). TODGA is used for actinides(III) and lanthanides recovery from highly acidic waste solution, whereas TPEN for actinides(III)/lanthanides separation. TPEN is combined with acidic extractant for synergistic separation. Iodine-129 is recovered from dissolvor off-gas and separated from C-14 by a thermal swing adsorption process.

The party of Mitsubishi Heavy Industry Co., Nagoya university and JNC studies on a reprocessing process that utilizes supercritical carbon dioxide ( $sc\text{-CO}_2$ ) instead of normal

paraffinic hydrocarbon. A spent fuel is first voloxidated and contacted with  $\text{sf-CO}_2$  containing tributylphosphate (TBP) and nitric acid. Uranium and plutonium will be selectively extracted into  $\text{sf-CO}_2$  phase with remaining fission products in solid state. The process enables to combine dissolution and co-decontamination steps and eliminates wastes originated from nitric acid dissolution and paraffinic diluent. The feasibility of this process will be examined.

#### 4.4. Other activities in Japan

Chromatographic separation is limited in analytical and special purposes due to the degradation of organic matrix of adsorbent. Institute of Research and Innovation (IRI) studies application of macro porous silica based adsorbent such as anionic exchanger and adsorbent that various extractants are impregnated. The advantages of those special adsorbent are less degradation products by radiolysis and fast elution. As a reprocessing of spent fuel, anionic exchange process has been investigated [7]. From the highly active eluate, extraction chromatography is also studied. HDEHP, CMPO, Cyanex 301 and R-BTP were already examined [8, 9].

The Tachimori's group of JAERI proposed the ARTIST process for spent fuel reprocessing [10]. The separation process intends to recover actinides including U, Pu, Np, Am and Cm by utilizing amide extractants. The process recovers both U and the other actinides mixed products that will be stored for further treatment in the future. For recovery of uranium, a branched monoamide extractants will be used because it preferentially extracts U(VI) compared with Pu(IV) and enables selective recovery of uranium from dissolvor solution. In order to recover other actinides after U recovery, a newly developed extractant TODGA is effective. If necessary, the actinides product without uranium will be treated with monoamide for Pu recovery and with other extractant for actinides(III)/lanthanides(III) separation.

Nitride fuel is attractive for its thermal conductivity and safety aspect. Aqueous reprocessing requires N-15 recovery by voloxidation. On the other hand, nitride fuel can be treated by pyroprocess owing to its electric conductivity and free energy for chloride production. JAERI conducts basic study on pyrochemical process for nitride fuel and found N contained in fuel can be recovered and recycled [11].

JNC also proposed another recycling system based on the concept that it should reject unneeded elements from spent nuclear fuel for recycling. The conceptual recycling system was named as the ORIENT (Optimization by Removing Impedimental Elements) cycle [12]. First, elements contained in a FBR fuel was classified into 4 categories of routing, namely core fuel, vitrified waste, transmutation and low level waste, according to the property of reaction with neutron, radioactivity, fuel fabrication and vitrification. As a result, actinides (U, Np, Pu, Am and Cm) and long-lived radionuclides will be recycled as a fuel. Fission products that has short half lives and is stable will be treated into low level waste other than vitrified waste. Although the reprocessing process is under discussion, both aqueous and pyroprocess was proposed as candidates for discussion.

## 5. CONCLUSIONS

The MOX fuel was arrived at BNFL from Takahama Power Plant of Kansai Electric Power Co. on September 17th 2002. The Shipment was completed and was due to the problem of manufacturing report. In this August, another problem relating to the report of regular checking at power plant of Tokyo Electric Co. was found out. Similar problem was reported

from other electric power companies. This has affected on the public confidence especially at the area that has nuclear power plant. Some governors already stated that utilizing MOX at existing LWR had to be prolonged. As a result, it is difficult to predict when the use of MOX fuel will start and is doubtful whether this will not this cause delay on commercial storage and reprocessing of spent fuel.

In December 2001, the government released a program on the improvement of special public institutions. In the program, JAERI and JNC shall be merged. The detail of merge shall be declared within 2004 Japanese fiscal year. The direction of new organization as well as future research and development program will be discussed in detail.

The secondary investigation of the submitted application for change of "Monju" prototype FR was started in May 2002 by the Atomic Energy Commission and Nuclear Safety Commission after the acceptance by the Nuclear and Industrial Safety Agency. It is expected that the investigation will be completed without delay. This will contribute not only to the progress of FBR cycle development to the next stage but also to steady growth of LWR fuel recycling including MOX fuel.

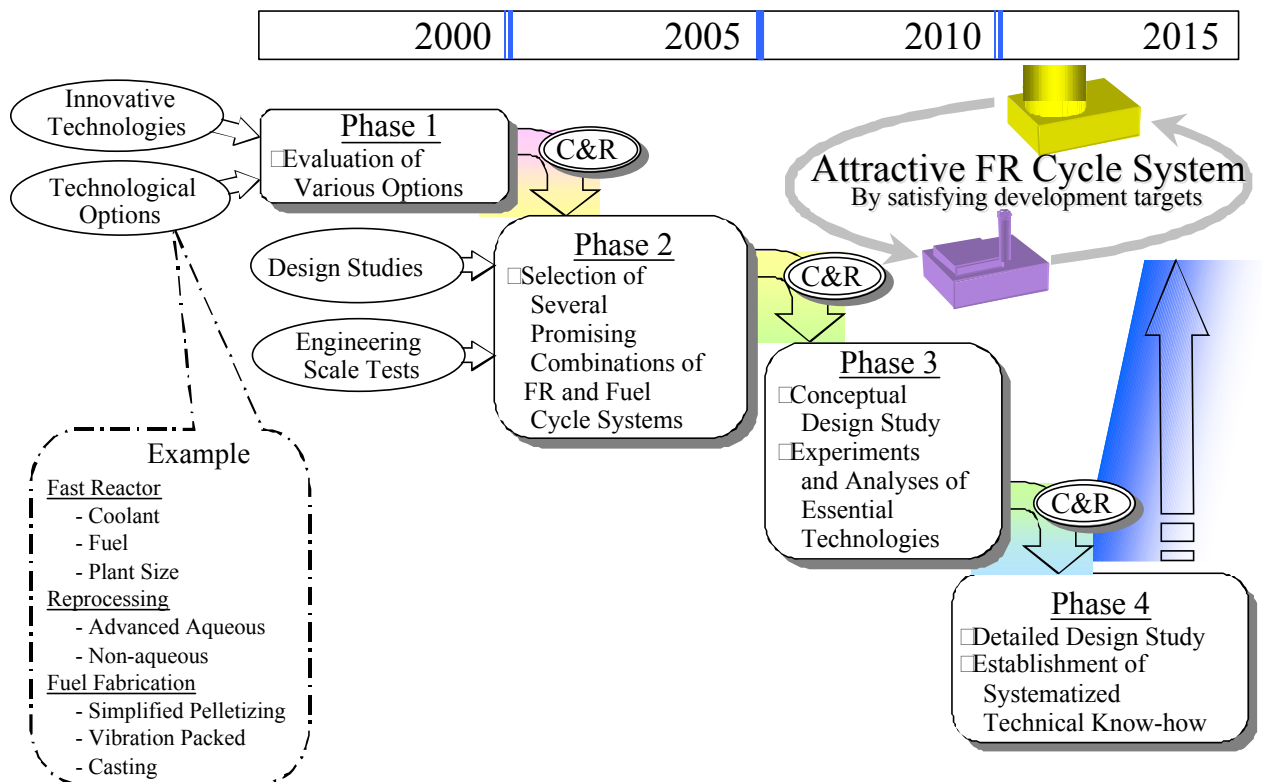


FIG. 1 Development of the FR Cycle System.

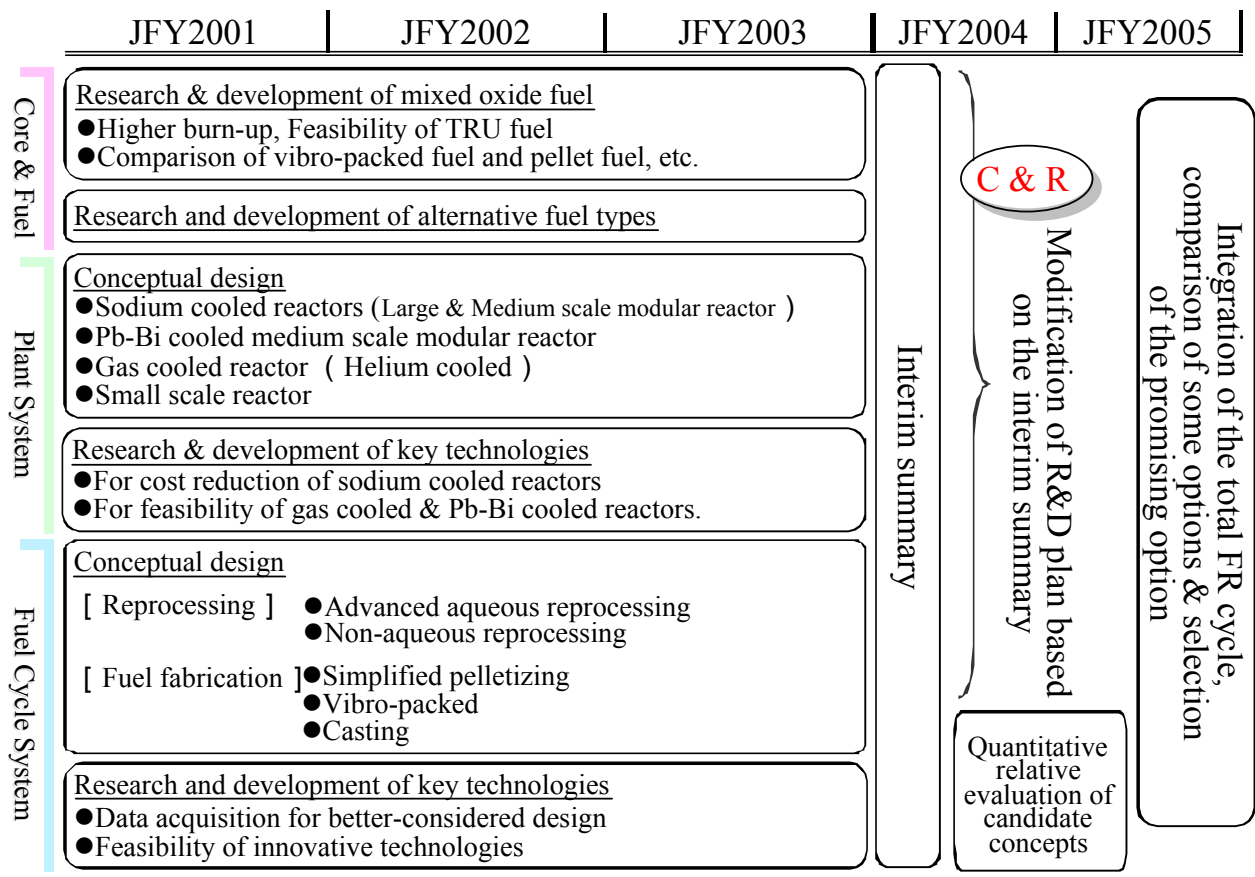


FIG. 2. Scope of the Phase II Feasibility Study.

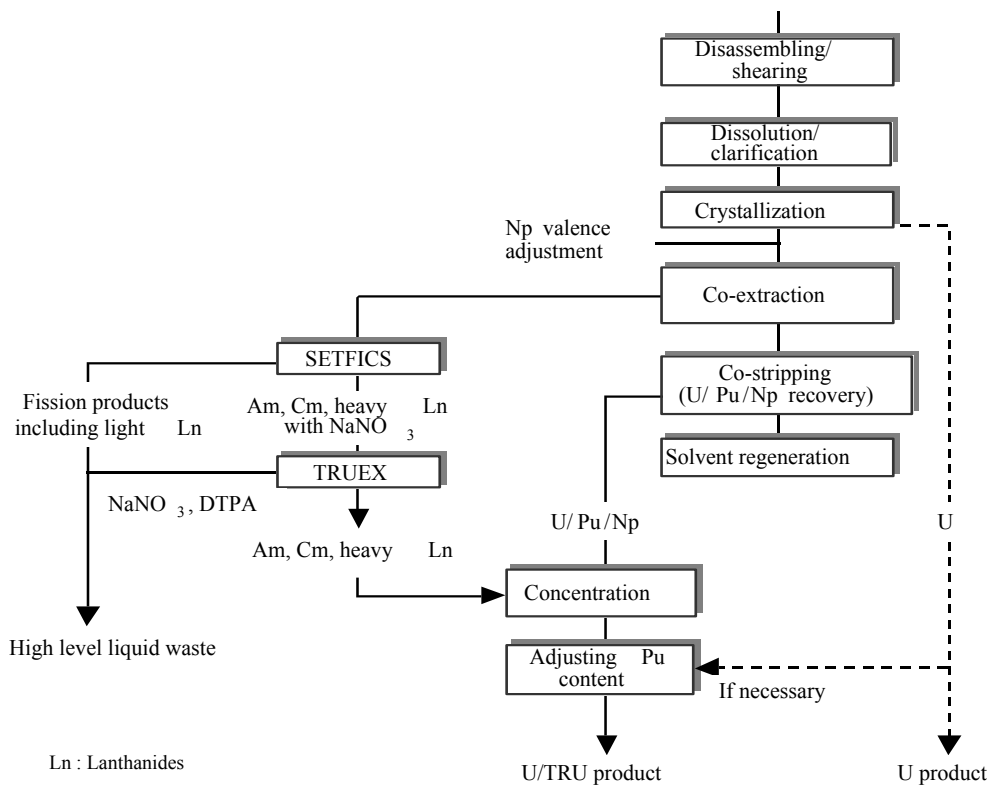


FIG. 3 The reference process for aqueous reprocessing.



a) U(VI) and Pu(IV)



b) U(VI) and Pu(VI)

*FIG. 4. The appearance of the uranium crystallization. a) UNH crystal from the real dissolver solution, Pu(IV) was rejected. b) UNH crystal with Pu(VI) nitrate. The color was orange instead of yellow of UNH.*

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## SPENT FUEL TREATMENT IN THE RUSSIAN FEDERATION

**T.F. Makarchuk**

ICC Nuclide, St-Petersburg, Russian Federation

**Ev.G. Kudryavtsev**

Minatom, Moscow, Russian Federation

### INTRODUCTION

A strategic policy of the development of nuclear industry in Russia is the closure of nuclear fuel cycle, which must result in a more complete use of natural nuclear fuel and artificial fissile materials generated in operation of nuclear reactors (plutonium, etc.), ensuring minimization of waste from fuel reprocessing, approximation to equivalency in radiation and migration terms of initial natural fuel and waste which is subjected to final disposal. The main idea is to provide the greatest possible, in terms of volume and nomenclature, reprocessing of irradiated fuel at the radiochemical facilities both existing and under construction.

The mode of thought of Minatom concerning the spent nuclear fuel (SNF) management is generalized in the concept which systematically describes the problems of management of SNF with due allowance for its importance for competitive development of the closing technology of nuclear fuel cycle. The implementation of technical solutions incorporated in the concept of SNF management will require investments to the amount of more than 3 billion US dollars for construction of basic facilities, upgrading of available facilities, performance of research as well as for designing.

In July 2001 the President of the Russian Federation approved a package of few laws – new and amended – On using of Atomic Energy, and On Radiation Remediation, then in January 2002 new one - On Environment Protection, The adoption of these documents will allow not only to establish a reliable basis for development of the entire complex of closing nuclear fuel cycle but also to resolve the environmental problems including those of SNF management.

In compliance with the long term Russian strategy of development of nuclear industry an cost-effective use of plutonium energy potential requires to postpone reprocessing of SNF from light-water reactors for tens of years until the possibility will exist of using the retrieved plutonium for initial load into new generation fast reactors which will form the basis of large scale nuclear power industry. Thus along with upgrading and establishment of new radiochemical plants which are planned to be put into operation after 2020, another important issue is a reliable long term storage of all kinds of spent fuel except those which are reprocessed on industrial scale at the RT-1 (Mayak) plant. The output of the Mayak plant in 2001 was more than 100 t/year, by 2007 an increase to 300 t/year is expected. By 2002 the total amount of SNF retrieved from reactors exceeded 15,000 t U with total activity more than  $300 \times 10^{18}$  Bq with an annual discharge of about 710 tHM.

Currently, approximately 3300 t U has been reprocessed at the Mayak plant. With the limited amount of reprocessing the problem of SNF storage increasingly gains momentum: taking into account the increased defuelling from decommissioned power plants, nuclear powered vessels, propulsion and research reactors, the existing SNF stores are expected to be filled by



2007. One should also consider that the technology now in use for storing fuel in water pools for several dozens of years does not fully meet modern safety requirements while a small reprocessing plant at PO Mayak has been in operation since 1977 and requires a technical upgrading and reconstruction. Principally new technologies of SNF reprocessing are under development. In this context at the moment the technical design solutions of the radiochemical plant RT-2 at MCC (Krasnoyarsk) are being re-evaluated.

## SOME CHANGE IN LEGAL PROVISIONS

The State Duma of the Russian Federation in June 2001 adopted a package of Federal laws including amendment to the law “On use of atomic energy” as well as new law “On special environmental programs for remediation of radioactivity contaminated areas”. In July 2001 the President of the Russian Federation approved a package of Federal laws which allow to expand services rendered by Russian companies in the field of management of SNF, namely, to accept SNF from foreign NPP for the interim storage and reprocessing. In January 2002 the renewed law “On environmental protection” was approved.

During last decades Russia has accumulated a lot of environmental problems due to the development of defence nuclear programs. Receipt of foreign SNF for reprocessing with the initial long term storage over the period of 20-40 years will allow funding the remediation activity as well as to develop and update the nuclear industry infrastructure for SNF management.

### 1. SPENT FUEL TREATMENT – STATE AND HORIZONS

The basic milestones of the SF management development include:

- creation of reliable systems for long term monitored fuel storage;
- development of fuel reprocessing;
- balanced inclusion of SF recycle products into fuel cycle.

Currently, 30 power units of total capacity more than 22,0 GWt are operating at 10 nuclear power plant sites. New WWER-1000 unit was put into operation at the Rostov NPP in 2001. Several additional units are under construction. Table I lists the quantities of SF arising from operating reactors of various types.

Table I Types and amount of accumulated SNF from reactors

<b>Reactor type</b>	<b>WWER-440</b>	<b>WWER-1000</b>	<b>RBMK-1000</b>	<b>BN-600</b>	<b>EGP-6</b>
<b>Factors</b>					
Number of power units	6	8	11	1	4
Annually discharged amount of SNF from 1 unit/total, tU	12.0/70	21.0/170.0	40.0/450.0	13.0	1.5/6.0
Amount of SNF at storage at NPPs sites, tU	263.0	643.0	9500.0	66.0	120.0

### WWER-440, BN-600

After 3-5-year cooling in AR-pools the WWER-440, and BN-600 fuel is shipped to the RT-1 plant for chemical reprocessing. The transportation is carried out in transportation packages in keeping with the rate of SF formation. Defective spent fuel assemblies are stored in special cans in the AR-pools.

### WWER-1000

The SF from WWER-1000 reactors cooled for 3-5 years is shipped from the NPP sites to the centralized storage facility at the RT-2 site. Currently this facility of the design capacity 6,000 tU is 55% filled (3,200 tU). By 2005-2007 the design capacity will exhaust. The spent fuel has been stored in water for 20 years and future plans call for this facility modification to increase the storage capacity to 8 400 tU through the use of fuel baskets of the higher capacity and the construction of adjoining building. These measures will prolong SF reception for another 3-5 years. The start of WWER-1000 fuel reprocessing is expected after the completion of RT-1 modification (by 2007), while the commissioning of RT-2 meant for full-size reprocessing of this fuel is expected beyond 2020. Therefore, it is necessary to get a more accurate assessment of permissible wet storage duration and to provide additional dry storage capacity.

### RBMK-1000

RBMK-1000 SF is not reprocessed and is currently stored in AR- cooling pools and wet AFR facilities at the NPP sites. The existing storage facilities will provide for, at most, 5 years of power unit operation. The fuel arisings at the NPP sites have exceeded 9500 tU. 3,000 FAs are leaking and stored in the special cans at reactor pool. In the future the long (~10 meter) assemblies will be separated into two fuel bundles in special hot cells at the NPP sites and stored in the dual-purpose metal-concrete casks (MCC), then transported to the Centralized dry storage facility at the Krasnoyarsk site. It is expected that the first stage of the facility will come into service by 2006-2007.

### EGP-6

Four EGP-6 reactors at Bilibino NPP shall be shut down in 2004 according to the design (in case their service life is not extended). The total mass of SF is 164 tU (6500 FAs). There are no leaking fuel assemblies. Of the three available cooling pools existing at the NPP site two have already been filled and these are used as dry storages.

### AMB

Two AMB reactors of the Beloyarsk NPP were shut down in 1989. The fuel – 190 tU in 5,000 FAs was discharged from the reactors and stored in dry canisters and AR-pools at the reactor site; and in the cooling pools of the RT-1 plant (76 tU in 2200 FAs). The major portion of FAs is damaged.

The basic option of AMB fuel management is connected with removal the fuel from cooling pools into dry storage facility. This necessitates the development of the technology for fuel transforming into safe state with the aim of subsequent transportation for extended storage.

Figures 1 and 2 shows the management schemes for SF of various types. Scheme 1 includes WWER-1000 SF, which will be reprocessed at the RT-1 after its modernization. The concept defines the immediate (2007) and long term (2025-2030) prospects of its realization.

In the nearest future consideration shall be given to the feasibility study of permissible storage times (to 50 years) and optimal conditions for dry storage of intact and defective fuel. For safety reasons leaking FAs to be reprocessed are accommodated in sealed cans.

The storage mode should provide the possibility of fuel retrieval for integrity control or reprocessing or conditioning for final disposal.

Hence, the dry storage must provide the possibility of fuel condition monitoring during storage. A dry storage facility for WWER-1000 and RBMK-1000 will be constructed at the Krasnoyarsk site by 2005-2007. Its capacity will amount to 33,000 tU.

After the completion of the future RT-2 plant will treat WWER-1000 SNF (which currently is being accumulated in the MCC storage facility), foreign PWR and BWR, and, perhaps, Russian RBMK reactors. The plant commissioning is planned after 2020, the design productive capacity is 1500 tU of annually. Reprocessing of conditioned and failed SFA is anticipated. The recovered uranium will be shipped for re-enrichment and plutonium for use in future fast reactors.

With allowance made for priorities on safety and economics the work is under way on a further development of the design solutions for SNF processing which will provide conditioning, environmentally safe storage, disposal and/or transmutation of fissile nuclides and radioactive elements.

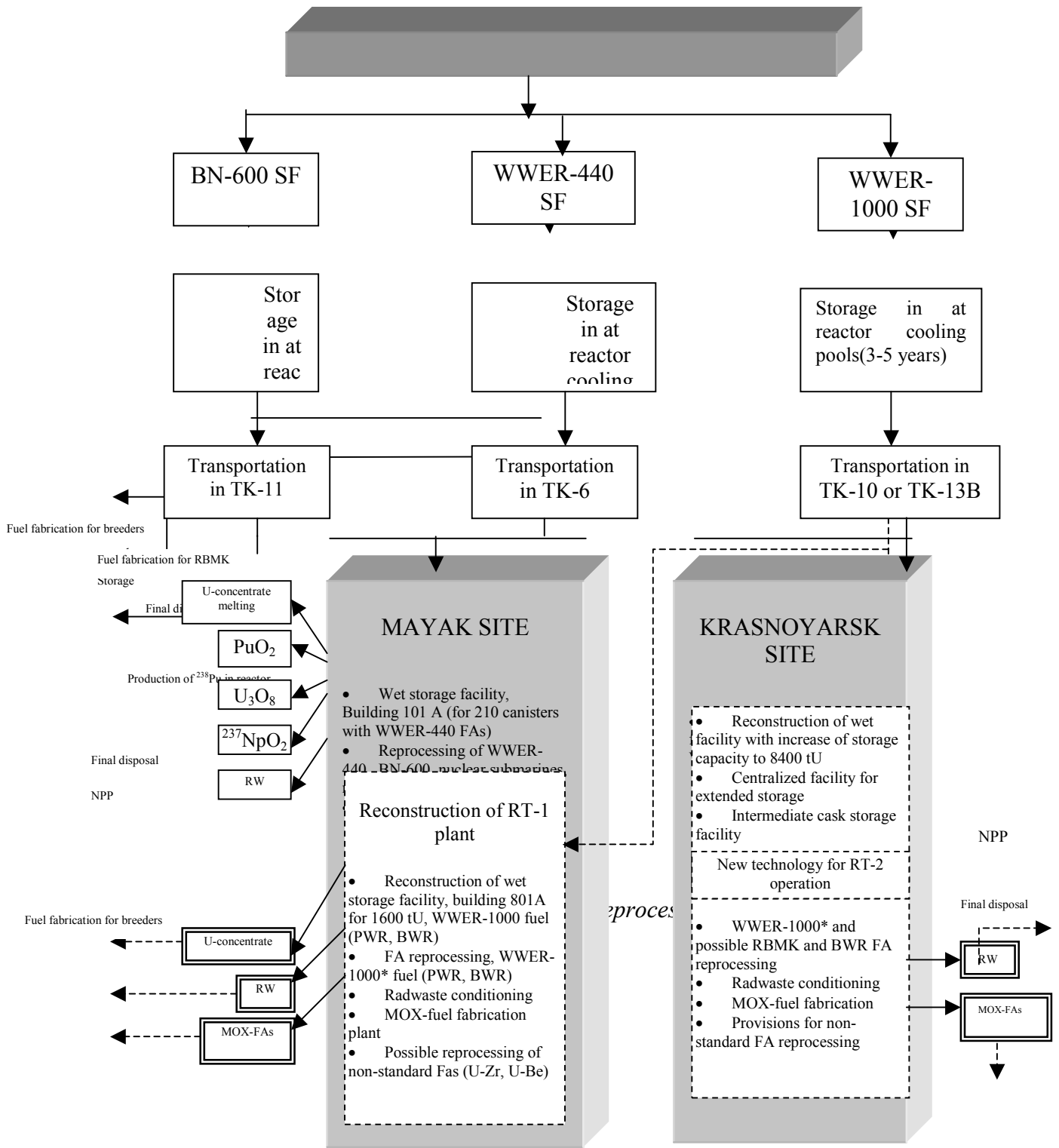
## 2. SNF REPROCESSING

The radiochemical reprocessing of SNF at present is carried out at RT-1 plant of PO Mayak near Chelyabinsk, and will proceed after its reconstruction and upgrading. In the future SNF will be reprocessed at RT-2 plant at MCC site near Krasnoyarsk.

The existing RT-1 plant with solvent extraction technology is able to reprocess the following types of spent fuel:

- SFA based on ceramic  $\text{UO}_2$  fuel with zirconium alloy claddings;
- SFA based on ceramic  $\text{UO}_2$  fuel with different steel claddings;
- SFA based on dispersed  $\text{UAl}_3 - \text{Al}$  and  $\text{UO}_2 - \text{Al}$  compositions in aluminum alloy cladding, initial enrichment up to 90% of uranium-235;
- SFA based on dispersed uranium dioxide in copper-magnesium matrix fuel composition with enrichment up to 90% in uranium-235, steel cladding.

For processing of SFA from research reactors BOR-60, BR-10 the electrochemical treatment technology is under development in NIAR (Dimitrovgrad).



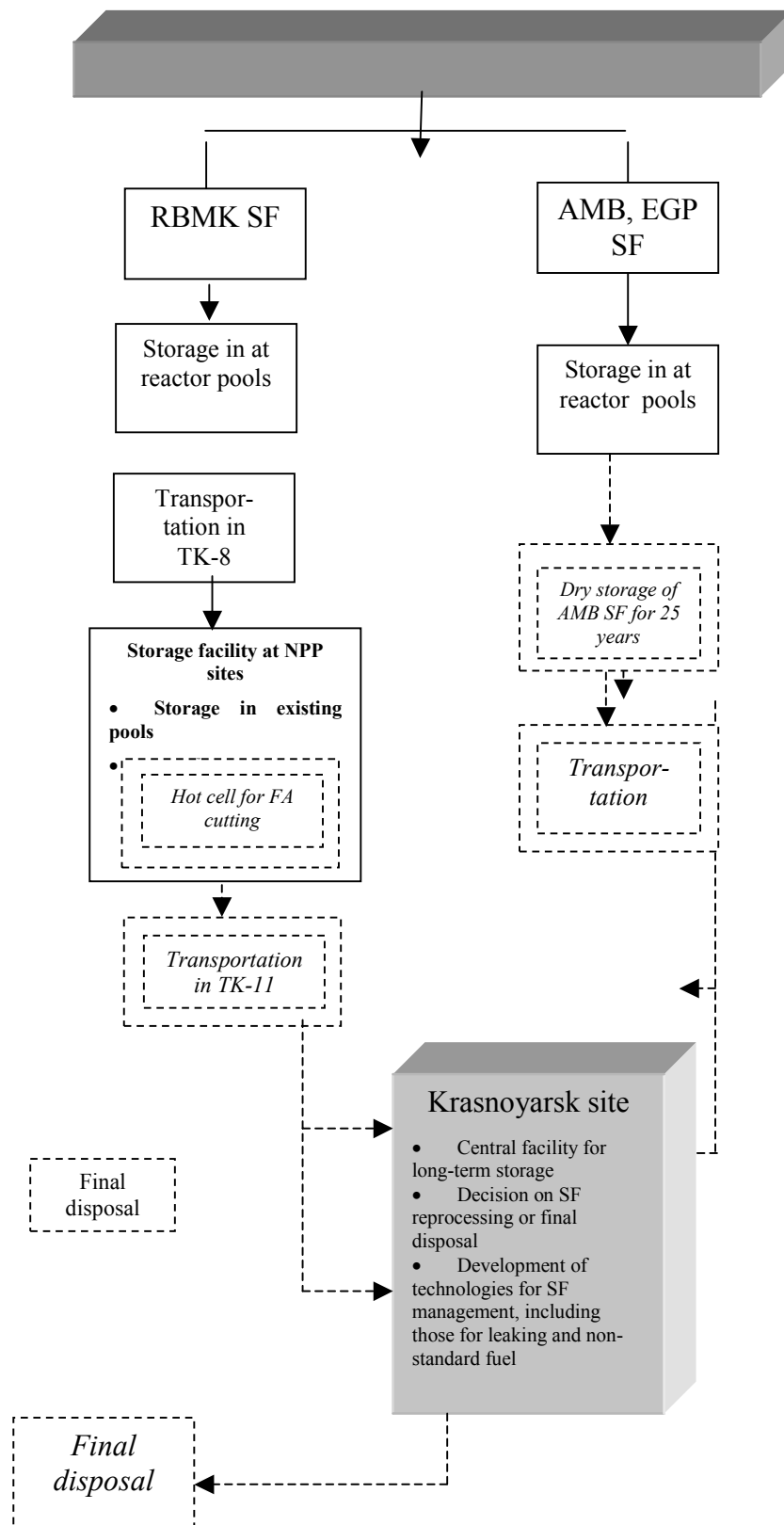


FIG. 2. Management with stored fuel with deferred decision.

The design output of the RT-1 plant by WWER-440 SNF is 400 tons a year. The reprocessing process (“modified purex”) includes dissolution of spent fuel with a subsequent separation of plutonium and uranium from fissile products and minor actinides. A detailed description of the technology of acceptance, SNF buffer storage, SNF shearing, dissolution, clarification and extraction purification was presented in a report at the IAEA meetings (see: IAEA-TECDOC-1103).

The end products from SNF reprocessing are:

- melt of uranyl nitrate hexahydrate enriched to 2.0-2.4% in  $U^{235}$  (due to mixing of RepU from WWER with highly enriched uranium from reprocessing of BN and propulsion reactors fuels);
- uranium oxides with enrichment of 17-21% in  $U^{235}$ ;
- neptunium dioxide;
- plutonium dioxide.

Reprocessed uranium is used mainly for RBMK fuel manufacturing. Neptunium dioxide is used for production of  $Pu^{238}$  isotope. Plutonium dioxide in a special package is transferred for storage.

SNF reprocessing is accompanied with a production of RadWastes which are subjected to treatment. At the present time at the plant the technology of high-level liquid wastes partitioning is used to separate cesium and strontium. Cesium- and strontium-containing solutions transferred to a vitrification facility.

Due to decrease of amount of WWER-440 SNF shipped for reprocessing the plutonium production is reduced from 2.7 t/year to 1,0 t/year.

The RT-1 plant is in operation more than 25 years. To replace equipment and upgrade obsolete waste management technologies it is essential to upgrade the plant. Reconstruction will also provide an opportunity to reprocess SNF from WWER-1000 as well as foreign PWR and BWR fuels. The modernization is planned to be completed around 2007.

### 3. MANAGEMENT OF RADWASTES ARISING FROM REPROCESSING

Current management practice for liquid middle level waste and liquid high level waste (HLW) from SNF reprocessing at RT-1 plant involves vitrification of the wastes in EP-500 ceramic melter with capacity 500 liter of concentrated HLW per hour. Alumophosphate radioactive glass form is produced using direct evaporation-calcination-vitrification technology. Vitrified wastes are placed in steel canister and are stored in a dry vault-type storage facility. The semi-industrial facility for partitioning of high-level wastes was put in operation at RT-1 in August 1996 first in the world. The detail description of management of RadWastes was presented in a report at the meeting of Working Group (see: IAEA-TECDOC-1103).

### 4. EMERGING TECHNOLOGIES

Studies of new nuclear cycle technologies are related to the use of molten salts – alkaline metal chlorides and fluorides. Development of this option is motivated by search for simpler processes and more compact equipment systems. Molten salts as ionic liquids possess the unique technological features: no need for diluents, high dissolubility of fuel components, high resistance to irradiation. The investigation of characteristics of haloid melts in order to

use them in nuclear technologies started in 40-s. Up to date an extensive data set on chemistry and technology of chloride melts has been gained. Chemistry of U, Pu, Th, Zr, Nb, Mo, Ru and rare elements has been studied in detail. These fundamental studies have been used as the basis for the technology of treatment of spent fuel from fast reactors by pyroelectrochemical method developed by NIIAR.

The basis for the technology is a concept of separation and clearing of plutonium dioxide as a matrix for incorporation of a number of radionuclides, and subsequent storage. Treatment includes fuel dissolution, electrochemical precipitation of  $UO_2$  with capture of specific fission products. To concentrate the remaining nuclides (except cesium-137 and, partially, strontium-90) they are precipitated from NaCl-KCl melt in form of phosphates. Studies of products and reprocessing waste have been completed. The successful performance of investigations and experiments on treatment of BOR-60 spent fuel has allowed to elaborate the technology of fast reactor fuels reprocessing.

More than 20-year experience of investigating electrochemical technology of treatment of oxide fuel of nuclear reactors, technology of manufacturing fuel elements with vibropacked fuel, and results of fundamental researches have made possible to develop a technological process of converting metal plutonium into oxide fuel. It has been verified that pyrometallurgical process in salt melts allows in the shortest possible time to convert metal plutonium into oxide fuel which might be used for manufacturing fuel elements both with vibropacked and pellet fuel, results of these works are reviewed in selection of optimum technology of SNF treatment at RT-2 plant.

## 5. CONCLUSION

By now the technological potential and experience of SNF treatment has been gained which guarantees the safety of personnel, public and environment. The gained technical potential is based on a long-standing experience of treatment of SNF from WWER-440 and BN-600 reactors as well as propulsion and research reactors.

Treatment of the most part of accumulated SNF is postponed till the serial construction of new generation of fast reactors. The most essential tasks in the area of radiochemical treatment of SNF are as follows:

- improvement of the existing reprocessing technology in order to waste reduction;
- creation of new technologies for long-lived radionuclides management (P&T);
- refinement of “dry” technologies – electrochemical and pyrometallurgic processing of SNF;
- development and improvement of technologies for minimization of RW quantities;
- design and construction of large central dry storage facility for SNF.

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## STATUS OF SPENT FUEL TREATMENT IN THE UNITED KINGDOM

**M.J. Dunn, J.E. Hatter**

British Nuclear Fuels plc.,  
Spent Fuel Services,  
Risley, Warrington, United Kingdom

### Abstract

Nuclear power has been used to generate electricity in the UK since the 1950s. A number of reactor and fuel types have been developed and are currently in use, requiring different spent fuel treatment routes. Government policy on spent fuel management in the UK is that it is for the owners of the spent fuel to decide on the appropriate spent fuel management option, based on their commercial judgement, subject to meeting the necessary regulatory requirements. This paper reviews the spent fuel treatment technology along with the associated waste management and recycle facilities currently in use in the UK.

### Background and general issues

Nuclear power in the UK represents 17% of the installed capacity, but currently supplies about 25% of the electricity produced. The generating capacity comprises 8,320 MW AGR and 1,200 MW PWR operated by British Energy's (BE) subsidiaries British Energy Generation Ltd. (BEGL) and British Energy Generation (UK) Ltd. (BEG(UK)L), and 2,650 MW Magnox operated by British Nuclear Fuels plc (BNFL). The details of the nuclear power stations currently in operation in the UK are given in Table I.

Table I: Operational Nuclear Power Stations in the UK

Name	Type	No. of Reactors	Net Capacity (MW)	Start of Operation	Magnox closure date & AGR/PWR Accountancy Lifetime (Years)
Calder Hall	Magnox	4	200	1956	2003
Chapelcross	Magnox	4	200	1959	2005
Dungeness A	Magnox	2	440	1965	2006
Sizewell A	Magnox	2	420	1966	2006
Oldbury	Magnox	2	440	1967	2008
Wylfa	Magnox	2	950	1971	2010
Hinkley Point B	AGR	2	1170	1976	35
Hunterston B	AGR	2	1240	1976	35
Dungeness B	AGR	2	1140	1983	25
Hartlepool	AGR	2	1180	1983	30
Heysham 1	AGR	2	1100	1983	30
Heysham 2	AGR	2	1240	1988	35
Torness	AGR	2	1250	1988	35
Sizewell B	PWR	1	1200	1994	40



No new nuclear capacity is currently under construction, however, three recently published UK Government and Parliamentary reports on energy were all positive about the future role of nuclear energy.<sup>10</sup>

Throughout the long history of nuclear power in the UK the dominant form of spent fuel treatment has been reprocessing. Reprocessing has been carried out on a commercial scale in the UK at Sellafield since 1952. Initially, the driver for reprocessing in the UK was to separate plutonium for the military programme, but as the civil nuclear power programme began to expand, the driver became more one of resource utilisation with the anticipation that the separated plutonium would ultimately be recycled in fast reactors.

The first reprocessing plant at Sellafield, B204, was built to reprocess uranium metal fuel from the atomic piles. This plant operated until 1964, when a new larger reprocessing plant, B205, was constructed to reprocess spent fuel from the eleven Magnox nuclear power stations that were constructed in the UK; as well as fuel from two other Magnox stations, one in Japan and the other in Italy. This Magnox reprocessing plant has been reliably and successfully operated for over 30 years and still provides the reprocessing capacity for the UK's Magnox reactors. As of March 2002, the Magnox plant has reprocessed in excess of 40,000 tonnes of fuel.

Modifications were made to B204 head end plant in the late 1960's to allow the metal fuel reprocessing facilities to reprocess oxide fuel from Light Water Reactors commercially. B204 reprocessed some 90 tonnes of oxide fuel between 1969 & 1973. However, following an incident in 1973, B204 was shut down and never re-opened. Plans were then drawn up for a dedicated plant to reprocess both AGR and overseas LWR fuel. Following a lengthy Public Inquiry in the late 1970's permission was granted for the construction of the Thermal Oxide Reprocessing plant now known as Thorp. Thorp began operation in 1994 and the order book for Thorp is full for the first ten years of operation as shown in Table II below. As of March 2002, over 3,800 tU of this fuel has been reprocessed so far.

Table II: Thorp's Order Book for the First Ten Years of Operation (7000tU)

Country	Quantity of Fuel (tU)	Fuel Type
UK	2158	AGR
Germany	969	LWR
Japan	2673	LWR
Switzerland	422	LWR
Sweden	140	LWR
Spain	145	LWR
Netherlands	53	LWR
Canada	2	LWR
Italy	143	LWR
Reserved Capacity	295	-
Total	7000	-

Business has also been secured for the second 10 years of operations from customers in the UK and Germany. The total value of Thorp's order book is in excess of £12 billion.

<sup>10</sup> PIU report to the Energy Review; House of Lords Select Committee report on the security of Energy Supplies in the EU; House of Commons Trade and Industry Committee on the security of UK energy supplies.

In 2001, the first commercial scale Mox fuel fabrication facility in the UK, Sellafield Mox Plant (SMP), was granted authorisation to operate. SMP is physically linked to Thorp, providing a truly integrated recycling facility and eliminating the need for the transport of separated plutonium to a separate facility. The plant already has sufficient contracted business to reach break-even, in terms of economic justification.

### Spent Fuel Treatment

The drivers behind the choice of spent fuel management route for the different fuel types within the UK are detailed below. It should be noted that Government policy in the UK is that it is for the owners of the spent fuel to decide on the appropriate spent fuel management option based on their own commercial judgement, subject to meeting the necessary regulatory requirements. The main factors which have determined UK utilities decisions on spent fuel management, to date, have been based predominately on the technical considerations of the spent fuel characteristics, economic attractiveness of the options & at reactor site spent fuel storage capacities.

### Magnox Fuel

Magnox fuel elements consist of bars of natural uranium metal, approximately 1m long, which are clad in a magnesium alloy (giving rise to the name Magnox). The Magnox system was designed with a wet discharge routes and interim pond storage of fuel in anticipation of early reprocessing. Wylfa, which utilises a wet discharge route also, has an at-reactor dry storage facility built to guard against any interruption to reprocessing activities at Sellafield. Magnox fuel is reprocessed after about 6 months storage.

The reprocessing of Magnox fuel takes place at the B205 facility at Sellafield, which has been operational since 1964. This facility, which has a nominal capacity of 1500 tU per year, utilises the PUREX process to separate plutonium and uranium from the waste fission products and actinides. The plutonium and uranium are converted to PuO<sub>2</sub> and UO<sub>3</sub> powders, respectively and stored at Sellafield pending recycle in fuel.

The highly active waste liquor containing fission product waste is stored in cooled tanks prior to vitrification. The intermediate level waste, principally Magnox cladding, is sent for treatment at the Magnox Encapsulation Plant (MEP) where it is encapsulated in a cement grout and placed in steel drums. Drummed wastes are stored in purpose built stores in anticipation of disposal in a deep geological repository.

The B205 facility is due to close around 2012 once all the Magnox fuel has been reprocessed.

### AGR Fuel

AGR fuel pins are approximately 1m long and consist of enriched UO<sub>2</sub> pellets clad in a stainless steel tube. The fuel elements consist of 36 pins arranged in a circular lattice and sheathed in a graphite sleeve. The AGR power stations have very small at-reactor pond stores, as early reprocessing was envisaged during the design of the reactors, and hence all spent AGR fuel is sent to Sellafield where it is stored underwater. The contractual relationship between BNFL & BE covers the lifetime arisings of AGR fuel. It provides for a near maximum commitment to reprocessing over the first two decades of Thorp operation. Options for further reprocessing following the first 20 years of Thorp operation or long term storage also exist under the terms of the contracts.

Prior to reprocessing, the AGR fuel elements are dismantled in a purpose-built facility at Sellafield and the pins are placed into thin walled slotted cans, which can then be fed into the shearing facility of Thorp. Thorp, in common with the Magnox reprocessing facilities, utilises the PUREX process to separate out the uranium and plutonium from the wastes. The uranium and plutonium are converted within the plant to  $UO_3$  and  $PuO_2$  powders respectively and both products are retained within purpose built stores in the plant, awaiting customers decision on recycle utilisation.

As with the Magnox reprocessing facilities, the highly active liquor from Thorp reprocessing is stored in cooled tanks prior to vitrification. Intermediate level waste, in the form of cladding and associated components, are encapsulated in a cement grout and sealed in steel drums where appropriate. Drummed wastes are stored in purpose built stores in anticipation of disposal in a deep geological repository.

#### PWR Fuel

Currently, there is only one PWR in the UK which is Sizewell B. The spent fuel storage pond at Sizewell B was designed to accommodate 18 years spent fuel arisings but was reconfigured to accommodate 30 years spent fuel arisings. BE will consider in due course arrangements for further management of spent PWR fuel in the light of the prevailing commercial and regulatory environment.

#### SGHWR and WAGR Fuel

Currently, some 160 tU of fuel from the SGHWR and WAGR prototype reactors is being stored at Sellafield. UKAEA have signed a Memorandum of Understanding with BNFL for the reprocessing of this fuel through Thorp.

#### Fast Reactor Fuel

Following the withdrawal of Government support for the project, the Prototype Fast Reactor (PFR) at Dounreay in Scotland was shut down in March 1994 and is currently being decommissioned. The majority of the fuel from the PFR has been reprocessed in a mixed oxide reprocessing plant at Dounreay since 1979, with the plutonium arisings transferred to Sellafield for storage. This plant was closed in 1996 when the main dissolver developed a leak. The UKAEA and the Government are currently evaluating options for the future management of the remaining PFR fuel.

#### Treatment and Disposal of Radioactive Waste

Following the vitrification of the high level waste arising from both Magnox and Oxide reprocessing, the glass blocks are encased in stainless steel drums and transferred to a vitrified product store. The vitrified waste is cooled using passive air circulation in the store.

Similarly, the intermediate level wastes from both the Magnox and AGR reprocessing operations are stored in engineered stores following encapsulation in cement in steel drums (wastes arising from dismantling operations for AGR fuel do not require encapsulation and are stored in steel drums).

Solid low level waste is placed in steel drums, compacted and put into half-height ISO containers which are then disposed of at the nearby Drigg facility where they are buried in

concrete lined vaults. Liquid low level waste is treated by a number of plants to remove as much of the radioactivity as practicable before being discharged to the sea.

The UK Government is currently undertaking a consultation exercise on radioactive waste management. The process should ultimately lead to the identification and implementation of a management policy for HLW and ILW in the United Kingdom.

The overseas reprocessing customers have been offered the choice of receiving back all categories of wastes or substituting low and intermediate level wastes for a radiologically equivalent quantity of vitrified high level wastes. With this option a customer would receive back a single form residue, Vitrified High Level Waste.

Over the past two decades, BNFL has successfully pursued a strategy of reducing radioactive discharges from all its sites in the UK, particularly Sellafield. Since the 1970's, through investment in new waste management facilities, radioactive discharges from Sellafield site have been reduced to less than 1% of their peak levels in the 1970's.

### Recycle of Uranium

Over 15000 tU of the uranium recovered by Magnox reprocessing has been recycled and about 1650 tU of AGR fuel has been produced from this material. The recycle of reprocessed uranium from Magnox fuel currently has limited strategic benefit as assessed against alternative commercial options. The uranium market conditions are such that further Magnox Depleted Uranium (MDU) recycle is not attractive at present. Recycle of the higher residual enrichment product from Thorp is more economically attractive. BNFL is currently examining a range of recycled uranium opportunities with a number of customers.

### Recycle of Plutonium

At 31 December 2001, there was 79.9 tes of unirradiated separated plutonium in product stores at UK reprocessing plants. Some 17.1 tes of this material is held for overseas customers and the remainder is UK owned. Plutonium is continuing to be stored safely and securely under international safeguards at Sellafield. The UK Government policy on the utilisation of plutonium is that it is for the plutonium owner to choose its preferred management option subject to meeting the necessary environmental and regulatory requirements. BE will consider in due course the feasibility of recycling plutonium as MOX fuel at Sizewell B.

### The Prospects for Direct Disposal

The current policy is to reprocess all the Magnox Fuel and hence it is not anticipated that any Magnox fuel will be directly disposed of. It is expected that some AGR fuel will be prepared for long term interim safe storage, as BE's current contracts with BNFL allow for some long term storage as required. This fuel together with PWR fuel may ultimately be directly disposed of rather than reprocessed.

### Future Developments in Spent Fuel management Technology

With the high commitment to reprocessing in the UK, BNFL is actively investigating improvements in reprocessing technology both in the short term, to optimise the operation of current reprocessing plants, and in the longer term, looking towards the next generation of reprocessing plants. BNFL have invested in a new research facility, the "BNFL Technology

Centre” at the Sellafield site, which demonstrates the UK’s long term commitment to research, technology development and innovation in all areas of the fuel cycle. The integrated facilities will consist of high active cells, high active laboratories, low active and inactive laboratories and uranium-active rig hall. Construction of the facility is well advanced with occupation and operations now started.

Shorter-term research work is concentrated on process optimisation and improvement looking at such measures as reagent consumption, energy usage, further reduction of discharges, feed clarification and waste reduction. In addition, work is proceeding into enhancing the process envelope to allow a greater range of fuel types and histories to be accepted. As a result of the OSPAR meeting in July 1998, the UK government has published its draft strategy showing how it will further reduce radioactive discharges into the marine environment by 2020 and much work is now focused on providing ways to meet these targets.

It is anticipated that the Purex process will be the major reprocessing technology for the next few decades. However, novel reprocessing technologies are also being developed which incorporate alternative chemical separation processes, such as molten salts. These pyrochemical processes, based on chemical and electrochemical reactions between metals, molten salts and other “dry” media, are being designed for the recycle of fast reactor fuel and other waste treatment process for which the Purex technology is not specifically design. The novel processes provide a lower degree of decontamination, which corresponds to the prediction of relaxed specification for recycled uranium and plutonium in future generating plants. It is also expected that through the elimination of the aqueous reagents and the associated waste streams the processes may have environmental attractions. Significant technical work is still required to commercialise these processes. One of the major issues to be considered is their batch nature and the processing approach required to facilitate high throughput plants.

## **REVIEW OF SPENT FUEL REPROCESSING IN THE UNITED STATES OF AMERICA**

**J. Laidler**

Argonne National Laboratory,  
Argonne, Illinois, United States of America

Reprocessing in the United States began in the wartime Manhattan Project with recovery of plutonium from irradiated metallic uranium fuel discharged from the Hanford production reactors. Such reprocessing continued to large plants at Hanford and the Savannah River site until the early 1980s, when it was determined that the stockpile of separated military plutonium was more than adequate for defense needs. Shutdown of these facilities was essentially complete by the end of the 1990s.

In 1953, the U.S. instituted the Atoms for Peace Program. This program brought nuclear technologies out of the cloak of secrecy and encouraged peaceful uses of nuclear energy throughout the world. The objective of the Atoms for Peace Program was to promote the domestic and international exploration, development, and advancement of the technology necessary to build and operate reliable, economic nuclear power plants; to provide cooperative assistance in establishing a self-sufficient nuclear power industry; and to ensure the development and use of nuclear energy in electric power production. The agreements implementing this program allowed a sharing of information about industrial applications of nuclear energy, including nuclear fuel reprocessing techniques, while discouraging nuclear weapons proliferation. This change in U.S. policy set the stage for the International Atomic Energy Agency (IAEA), and promulgation of the Nuclear Nonproliferation Treaty (NPT).

To insure a self-sufficient, domestic commercial nuclear power industry, the U.S. Atomic Energy Commission (AEC) encouraged the transfer of nuclear fuel reprocessing from the federal government to private industry. As a result of this policy, three commercial reprocessing facilities were built in the U.S.: General Electric's Midwest Fuel Recovery Plant at Morris, Illinois; the Allied General Nuclear Services (AGNS) plant at Barnwell, South Carolina; and Nuclear Fuel Service's facility located near West Valley, New York.

The NFS West Valley facility was the first and only private plant in the U.S. to reprocess spent nuclear fuel. The West Valley facility was a PUREX process plant with a design capacity of 300 tons of fuel per year. It operated commercially from 1966 to 1972. The two other commercial reprocessing facilities were built, but never operated. The General Electric Midwest Fuel Recovery Plant (also 300 tons per year) at Morris, Illinois, adjacent to the site of the Commonwealth Edison Company Dresden reactors, was completed at a cost of \$64 million but was declared inoperable in 1974. In 1970, Allied General Nuclear Services (AGNS) began construction of a 1500 tons per year reprocessing plant at Barnwell, South Carolina, adjacent to the DOE Savannah River site. The Barnwell facility was due to begin operation in 1974, but following delays in construction and licensing, it still had not been completed or licensed when in 1977 President Carter decided to defer indefinitely all reprocessing of commercial irradiated fuel. That decision was based on non-proliferation grounds and remained in effect until President Reagan lifted the restrictions on commercial reprocessing in 1981. By that time, however, uranium prices were stable at very low levels and the private sector did not choose to enter into a reprocessing enterprise that was economically questionable and subject to interruption by political policy changes in future administrations.

In 1982, the successor agency to the AEC, the U.S. Department of Energy, made a commitment to accept spent nuclear fuel from utility companies operating commercial nuclear power stations. This eliminated any remaining incentive on the part of the utilities to invest in reprocessing. When the Clinch River Breeder Reactor Project was canceled in 1984, another incentive for the development of a commercial reprocessing industry was removed. Subsequently, President Clinton issued a directive on reprocessing that stated that the U.S. would not engage in civilian reprocessing for the purpose of recovering plutonium for use in commercial reactors. This policy remains in effect, even though the National Energy Policy issued by the administration of President Bush in 2001 provided some indication of a policy change by stating that the U.S. should re-examine its policies to allow for research, development and deployment of fuel conditioning methods that reduce waste streams and enhance proliferation resistance. The policy document reaffirms the position that the U.S. will continue to discourage the accumulation of separated plutonium worldwide, and proposes that the U.S. consider the collaborative development of reprocessing and fuel treatment technologies with international partners having highly developed fuel cycles and a record of close cooperation.

Commercial reactors in the United States presently generate about 2,000 tonnes of spent fuel per year, and the quantity of spent fuel now in temporary surface storage is about 50,000 tonnes, approaching the legislated limit of 63,000 tonnes for emplacement in the proposed Yucca Mountain repository. In the absence of a mechanism to cap the accumulation of spent fuel while still maintaining a nuclear power infrastructure capable of supplying at least 20% of the nation's electricity, it is conceivable that a second repository would be needed before the first is filled. Although the national policy restriction on commercial reprocessing remains in effect at present, the U.S. is engaged in the development of the technologies necessary for partitioning and transmutation of spent nuclear fuel, for the purpose of facilitating the geologic disposal of high-level wastes generated in the future.

In the partitioning and transmutation scenario, an integrated multi-tier nuclear energy system is envisioned, wherein the fleet of commercial nuclear power plants (categorized as Tier 0) would generate spent fuel at a rate of at least 2,000 tonnes per year. These Tier 0 reactors can be advanced LWRs, gas cooled reactors, or futuristic reactors (e.g., the Generation IV variety). The mix of these reactor types may evolve over time, but for the present it is assumed that the Tier 0 reactors will primarily be current-generation and advanced LWRs. The spent fuel discharged from the Tier 0 reactors will be reprocessed to recover plutonium, which (possibly together with neptunium) will be recycled to thermal burner reactors comprising Tier 1 of the multi-tier system. The Tier 1 reactors could be MOX-burning advanced LWRs or plutonium/neptunium-burning gas cooled thermal reactors. These Tier 1 reactors would be electricity generators, with the added mission of plutonium (and also perhaps neptunium) burning. It is assumed that the plutonium and neptunium recovered in the course of reprocessing the discharged spent fuel from Tier 1 advanced LWRs would be multi-recycled, while the fuel in Tier 1 gas-cooled reactors may be capable of sufficiently high burnup of fissile plutonium and neptunium that the fuel could be on a once-through cycle. In either case, fuel finally discharged from the Tier 1 reactors would be reprocessed to recover the transuranic elements. These materials would be combined with the minor actinides recovered during Tier 0 fuel reprocessing and sent to a Tier 2 transmuter reactor for complete fissioning.

The Tier 2 reactor is assumed to be a fast spectrum reactor, to exploit the higher fission cross sections of the minor actinides at high neutron energies. These reactors can be either critical fast reactors or subcritical accelerator-driven reactors. With appropriate design of the overall

system, the support ratio 2 for Tier 2 reactors can be reasonably large, on the order of 15 to 16. That is, about 6 to 7 GWt of Tier 2 reactors to consume the annual net transuranic output of 100 GWt of combined (Tier 0 + Tier 1) reactors. This is considered to be economically feasible, given the assumed higher capital cost of the Tier 2 reactors.

The reprocessing technologies that would be utilized in this multi-tier system are currently underdevelopment, and final selection of the processes for treatment of Tier 0, Tier I and Tier 2 fuels will not be made until 2006. It is most likely, however, that advanced LWR fuel (either UOX or MOX) will be processed by an aqueous solvent extraction method similar to the PUREX process. It will be necessary to extract plutonium (and probably neptunium) for recycle to the Tier I reactors, and to recover americium and curium for burning in the Tier 2 reactors. It is presently planned that iodine and technetium will be recovered during all spent fuel processing, with these elements to be sent to a thermal spectrum reactor for transmutation to stable xenon and ruthenium, respectively.

Both aqueous and non-aqueous methods for reprocessing discharged gas cooled reactor fuel are under development. A challenge for these processes is the attainment of high recovery efficiencies for the transuranic elements and the minimization of waste generation. The latter is a difficult problem due to the large quantities of carbon and silicon (or zirconium) present in the coated-particle gas cooled reactor fuel.

Tier 2 fuels are likely to be non-fertile inert matrix fuels. They would be multirecycled in order to achieve the required level of transuranic fissioning. They would probably contain significant amounts of zirconium. As such, they are not particularly suited to conventional aqueous processing, so pyrochemical processes are being studied. Support ratio is defined here as the ratio of GWt in (Tier 0 + Tier 1) to the GWt in Tier 2 that would be required for a total system in which there is no net annual accumulation of transuranics for the initial Dcof of the fuel. Although there is no need in Tier 2 for separation of the transuranic elements, the typical pyrochemical process does not produce a complete decontamination of lanthanide fission products from the transuranic product stream. If the lanthanide content is too high for acceptable table neutronics in the Tier 2 reactor, it may be necessary to resort to a hybrid process including an aqueous step to remove the lanthanides.

In order to meet the current program goal that the high-level waste sent to a geologic repository be no more toxic, after a decay period of less than 1,000 years, than the original uranium used to produce the fuel, it is necessary to recover 99.5-99.9% of the transuranic elements present in the discharged fuel stream. This requires high recovery efficiencies in both the chemical separations and recycle fuel fabrication operations, and efforts are being directed toward the development of fuels, fuel refabrication methods, and chemical processing methods that will meet these challenges.

The deployment of an integrated multi-tier system obviously will not occur in the near term. It is possible, however, that steps toward initiation of a partitioning and transmutation system could be taken within a decade or so, by processing LWR spent fuel for MOX recycle in existing commercial LWRs. Some of the groundwork for this step is already being done in the form of the Fissile Materials Disposition Program, which will consume excess weapons plutonium in selected commercial reactors. A form of chemical processing of the weapons plutonium is required for its use in MOX fuel, but the recovery and recycle of plutonium from commercial spent fuel would require a landmark change in national policy toward reprocessing, and it is impossible to gauge the likelihood of the implementation of such a policy change. For the moment, the course of action will be to develop and assess the necessary technologies in order that they can be available if called upon in the future.





## CONTRIBUTORS TO DRAFTING AND REVIEW

### CHINA

Yuansong Liu

China National Nuclear Corporation (CNNC),  
Division of Nuclear Fuels,  
P.O. Box 2102-10, Beijing 100 822  
Tel.: 0086-10-6851 6507  
Fax.: 0086-10-6857 1355  
E-Mail: liuys@cnncc.com.cn

### FRANCE

Brossard, P.

CEA/Valrho,  
Service des Procédés de Haute-Activité,  
B.P. 171,  
F-30207 Bagnols sur Ceze Cedex, France  
Tel.: 0033 4 66796561  
Fax.: 0033 4 66791474  
E-Mail: brossardp@amandine.cea.fr

Deroubaix, D.

COGEMA, DSI/MA,  
2, rue Paul Dautier,  
B.P. 4, F-78141 Velizy Cedex  
Tel.: 33 1 39 26 37 15  
Fax.: 33 1 39 26 27 73  
E-Mail: dderoubaix@cogema.fr

Giroux, M

COGEMA, DSI/MA,  
2, rue Paul Dautier ,  
B.P. 4, F-78141 Velizy Cedex  
E-Mail: mgiroux@cogema.fr

### INDIA

Kansra, V.P.

BARC,  
Nuclear Recycle Group,  
Trombay, Mumbai 400 085  
Tel.: 0091 22.550.5050-3112  
Fax.: 0091 22.550 5275  
E-Mail: kansravp@hotmail.com

### JAPAN

Koyama, T.

JNC,  
Advanced Fuel Technology Division,  
Tokai Works, 4-33 Tokai-mura, Naka-gun,  
Ibaraki-ken 319-1194  
Tel.: 0081 292821111  
Fax.: 0081 292820685  
E-Mail: koyama@tokai.jnc.go.jp

Kosugi, Mr.

JNC,  
Advanced Fuel Technology Division,  
Tokai Works, 4-33 Tokai-mura, Naka-gun,  
Ibaraki-ken 319-1194  
Tel.: 0081 292821111  
Fax.: 0081 292820685  
E-Mail: kosugi@tokai.jnc.go.jp

## **RUSSIAN FEDERATION**

T. Makarchuk

ICC Nuclide, Ministry for Atomic Energy,  
Lesnoy prospect, 64,  
194100 St. Petersburg  
Tel.: 0078125428186  
Fax.: 0078125426228  
E-Mail: nuklide@online.ru

## **UNITED KINGDOM**

Dunn, M.

British Nuclear Fuels,  
Spent Fuel Commercial Department,  
Hinton House, Risley,  
Warrington, Cheshire WA3 6AS  
Tel.:0044 1925 832136  
Fax.:0044 1925 833 213  
E-Mail: mike.dunn@bnfl.com

Hatter, J.

British Nuclear Fuels,  
Spent Fuel Commercial Department,  
Hinton House, Risley,  
Warrington, Cheshire WA3 6AS  
Tel.:0044 1925 832136  
Fax.:0044 1925 833 213  
E-Mail: justine.e.hatter@bnfl.com

## **UNITED STATES OF AMERICA**

Laidler, J.J.

Chemical Technology Div., Bld205,  
Argonne National Lab., Argonne, IL 60439-4837  
+1-630-2524479  
+1-630-9724479  
E-Mail: laidler@cmt.anl.gov

## **IAEA**

Hioki, K.

International Atomic Energy Agency,  
Wagramer Strasse 5, A-1400, Vienna, Austria  
+43-1-2600-33767  
+43-1-26007-22767  
E-Mail: K.Hioki@iaea.org

Lee, J.S.

International Atomic Energy Agency,  
Wagramer Strasse 5, A-1400, Vienna, Austria  
+43-1-2600-33767  
+43-1-26007-22767  
E-Mail: [J.S.Lee@iaea.org](mailto:J.S.Lee@iaea.org)

Legoux, P.

International Atomic Energy Agency,  
Wagramer Strasse 5, A-1400, Vienna, Austria  
+43-1-2600-33767  
+43-1-26007-22767  
E-Mail: [P.Legoux@iaea.org](mailto:P.Legoux@iaea.org)