FUEL REPROCESSING: STATUS AND CHALLENGES
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Chairperson

R. COULON
France
OVERVIEW OF SPENT FUEL MANAGEMENT IN CHINA

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Abstract

This paper briefly introduces the current reprocessing situation and challenges in the world, the policy and status of the nuclear energy development and SFM for the back end of the fuel cycle in China. Chinese government has already launched the nuclear energy medium-long-term development program, the opted policy of closed fuel cycle and the technical development strategy, the projected commercial reprocessing plant. The cold uranium test for nuclear power plant spent fuel reprocessing pilot plant is finished and the radioactive test is carried out in the early this year. The R&D program of reprocessing technology is emphasized. The challenges faced by China are described.

1. THE CURRENT REPROCESSING SITUATION AND CHALLENGES IN THE WORLD

The selection of a strategy for SFM 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 countries [1].

For the ultimate management of spent fuel, the following options are being implemented or under consideration:

- The one-through cycle, i.e. the direct disposal of the spent fuel in the geologic repository;
- The closed cycle, i.e. the reprocessing of the spent fuel, recycling of the reprocessed plutonium and uranium, and disposal of the wastes from the reprocessing operations;
- The so-called “wait and see” policy, which means first storing the fuel and deciding at a later stage on reprocessing or disposal.

The major reasons for choosing the option of reprocessing have been the efficient utilization of uranium resources, appropriate conditioning of waste and further decrease waste volume and radio-toxicity. The plutonium recovered by reprocessing can be recycled in as mixed oxide (MOX) fuel, replacing a nearly equivalent amount of enriched uranium and thus avoiding the need for considerable mining and enrichment operation. In the medium-long term, the recycling of nuclear fuel, including MOX in the fast reactors, may play an important role both in the 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.

The reprocessing activities and the closed fuel cycle policy have been implemented at present in China, France, India, Japan, Russia, and UK. U.S.A. also intends to restore the closed cycle policy. Civil reprocessing has been carried out on a commercial scale for over four decades in several countries. Since the opening of the first Purex plant at Savannah Rover in 1954, the Purex process has been utilized in a variety of flow-sheets and is still being used in all commercial reprocessing plants. Reprocessing using the Purex process has become a mature...
technology with considerable experience gained from the operation of civil reprocessing plants. The current and planned reprocessing capacities in the world are shown in Table 1.

TABLE 1. THE CURRENT AND PLANED REPROCESSING CAPACITIES IN THE WORLD

<table>
<thead>
<tr>
<th>Country</th>
<th>Site</th>
<th>Plant</th>
<th>Operation time</th>
<th>Capacity(tHM/y)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>present</td>
</tr>
<tr>
<td>China</td>
<td>Jiuquan</td>
<td>RPP</td>
<td>LWR</td>
<td>60</td>
</tr>
<tr>
<td></td>
<td></td>
<td>CRP</td>
<td>LWR</td>
<td>800</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2025</td>
<td>800</td>
</tr>
<tr>
<td>France</td>
<td>La Hague</td>
<td>UP2</td>
<td>LWR</td>
<td>1994</td>
</tr>
<tr>
<td></td>
<td>La Hague</td>
<td>UP3</td>
<td>LWR</td>
<td>1990</td>
</tr>
<tr>
<td>India</td>
<td>Trombay</td>
<td>PP</td>
<td>Research</td>
<td>1964</td>
</tr>
<tr>
<td></td>
<td>Tarapur</td>
<td>PREFRE1</td>
<td>PHWR</td>
<td>1974</td>
</tr>
<tr>
<td></td>
<td>Kalpakkam</td>
<td>PREFRE2</td>
<td>PHWR</td>
<td>1998</td>
</tr>
<tr>
<td></td>
<td>Kalpakkam</td>
<td>PREFRE3A</td>
<td>PHWR</td>
<td>2005</td>
</tr>
<tr>
<td></td>
<td>Tarapur</td>
<td>PREFRE3B</td>
<td>PHWR</td>
<td>2005</td>
</tr>
<tr>
<td>Japan</td>
<td>Tokai-mura</td>
<td>PNC</td>
<td>TRP LWR</td>
<td>1977</td>
</tr>
<tr>
<td></td>
<td>Rokkasho-mura</td>
<td>RRP</td>
<td>LWR</td>
<td>2010</td>
</tr>
<tr>
<td>Russia</td>
<td>Chelybinsk</td>
<td>RT1</td>
<td>WWER-440</td>
<td>1971</td>
</tr>
<tr>
<td></td>
<td>Krasnoyarsk</td>
<td>RT2</td>
<td>WWER-1000</td>
<td>1500</td>
</tr>
<tr>
<td>UK</td>
<td>Sallafieldd</td>
<td>B205</td>
<td>GCR</td>
<td>1967</td>
</tr>
<tr>
<td></td>
<td>Sallafield</td>
<td>Thorp</td>
<td>LWR/AGR</td>
<td>1994</td>
</tr>
<tr>
<td>Total</td>
<td>capacity</td>
<td></td>
<td></td>
<td>5850</td>
</tr>
</tbody>
</table>

The near and medium term challenges for reprocessing are to achieve economic competitiveness through the reduction of the volume and radio-toxicity 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 burnups. 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.

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 radio-toxicity and the level of radioactivity of spent fuel or waste separated during
reprocessing has driven to study the feasibility and economics of the partitioning and transmutation of minor actinides and long lived fission products.

The challenges of technical innovations with a number of technical objective, including: more efficient utilization of fissile and fertile materials; enhanced proliferation resistance; 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.

In the near term; the technologically mature aqueous processing methods constitute the main path forward, while dry processes are considered as adjunct or backup processes. In the longer term, however, fuel cycle applications related to advanced reactor concepts (fast reactors, gas cooled reactors, molten salt reactors, etc) may favor the use of pyro chemical processes, as do accelerator-driven system (ADS) designed for the destruction of highly radiotoxic minor actinides.

2. NUCLEAR ENERGY DEVELOPMENT STRATEGY AND SITUATION IN CHINA

With the rapid economy development and population increment in China, the energy demands are growing rapidly. The energy structure mainly relied on fossil fuel resources is not sustainable. In order to enlarge energy supply and reduce emission of pollutants and greenhouse gases as well, Chinese government make a very important decision on the energy strategy option for adjusting energy structure and ensuring the security of the energy supply. Nuclear power is the economic, safe and clean energy. China government has already made the strategic development plan for nuclear energy in three steps from thermal reactor (TR), fast breed reactor (FBR) to fusion reactor (FR).

China nuclear power medium-long term development program in the period of 2005-2020 has been announced [2]. According to the program, the total capacity of nuclear energy in 2020 will reach 40 GWe, which will take 4% of the total gross national electricity capacity. The capacity of nuclear power plants under construction will be 18 GWe. With the increased pressure and requirements of global climate change protocol and economic development, china prepares to modify the total capacity of nuclear energy.

There are now 11 units of 6 nuclear power plants (NPPs) in operation and 24 units of 10 NPPs under construction. Table 2 indicates the information of the NPP in operation. Table 3 indicates the information of the NPP under construction or approved.

The following figure shows the NPP distribution in China, including existed and planning NPP.

The FBR development strategy is to deploy the FBR in china from the experimental FBR, demonstration FBR to commercial FBR. The projected time schedule for FBR development in the draft program is following: operation of the experimental FBR in 2010, constriction of demonstration FBR finished in around 2025, construction of the first commercial FBR finished in around 2035.
FIG. 1. NPP distribution in China.

TABLE 2. DATA OF THE NPP IN OPERATION

<table>
<thead>
<tr>
<th>Name of NPP</th>
<th>Reactor type</th>
<th>Installed capacity(MWe)</th>
<th>Operation date</th>
</tr>
</thead>
<tbody>
<tr>
<td>Qin shan (phase 1)</td>
<td>Unit 1 PWR</td>
<td>300</td>
<td>1994/04/01</td>
</tr>
<tr>
<td>Dayawan</td>
<td>Unit 1 PWR(CPY)</td>
<td>2×984</td>
<td>1994/02/01</td>
</tr>
<tr>
<td></td>
<td>Unit 2</td>
<td></td>
<td>1994/05/06</td>
</tr>
<tr>
<td>Qinshan (Phase 2)</td>
<td>Unit 1 PWR(CNP650)</td>
<td>2×650</td>
<td>2002/04/15</td>
</tr>
<tr>
<td></td>
<td>Unit 2</td>
<td></td>
<td>2004/05/03</td>
</tr>
<tr>
<td>Lingao</td>
<td>Unit 1 PWR(CPY)</td>
<td>2×990</td>
<td>2002/05/28</td>
</tr>
<tr>
<td></td>
<td>Unit 2</td>
<td></td>
<td>2003/01/08</td>
</tr>
<tr>
<td>Qinshan (Phase 3)</td>
<td>Unit 1 PHWR(CANDU)</td>
<td>2×700</td>
<td>2002/12/31</td>
</tr>
<tr>
<td></td>
<td>Unit 2</td>
<td></td>
<td>2003/07/24</td>
</tr>
<tr>
<td>Tianwan</td>
<td>Unit 1 PWR(WWER)</td>
<td>2×1060</td>
<td>2006/05/12</td>
</tr>
<tr>
<td></td>
<td>Unit 2</td>
<td></td>
<td>2007/05/14</td>
</tr>
<tr>
<td>6</td>
<td>11</td>
<td>9068</td>
<td></td>
</tr>
</tbody>
</table>
### TABLE 3. DATA OF THE NPP UNDER CONSTRUCTION OR APPROVED

<table>
<thead>
<tr>
<th>Name of NPP</th>
<th>Reactor type</th>
<th>Installed capacity (MWe)</th>
<th>FCD date</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lingao (phase2)</td>
<td>Unit 3</td>
<td>PWR(CPR1000)</td>
<td>2×1080</td>
</tr>
<tr>
<td></td>
<td>Unit 4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Qinshan 2 (extension)</td>
<td>Unit 3</td>
<td>PWR(CNP650)</td>
<td>2×650</td>
</tr>
<tr>
<td></td>
<td>Unit 4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Hongyanhe (liaoling)</td>
<td>Unit 1</td>
<td>PWR(CPR1000)</td>
<td>4×1080</td>
</tr>
<tr>
<td></td>
<td>Unit 2</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Unit 3</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Unit 4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ningde (Fujian)</td>
<td>Unit 1</td>
<td>PWR(CPR1000)</td>
<td>4×1080</td>
</tr>
<tr>
<td></td>
<td>Unit 2</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Unit 3</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Unit 4</td>
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<td></td>
</tr>
<tr>
<td>Fuqing (Fujian)</td>
<td>Unit 1</td>
<td>PWR(M310)</td>
<td>2×1000</td>
</tr>
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<td></td>
<td>Unit 2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fangjiashan (Zhejiang)</td>
<td>Unit 1</td>
<td>PWR(M310)</td>
<td>2×1000</td>
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<tr>
<td></td>
<td>Unit 2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sanmen (Zhejiang)</td>
<td>Unit 1</td>
<td>PWR(AP1000)</td>
<td>2×1250</td>
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<tr>
<td></td>
<td>Unit 2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Yangjiang (Guangdong)</td>
<td>Unit 1</td>
<td>PWR(CPR1000)</td>
<td>2×1080</td>
</tr>
<tr>
<td></td>
<td>Unit 2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Haiyang (Shandong)</td>
<td>Unit 1</td>
<td>PWR(AP1000)</td>
<td>2×1250</td>
</tr>
<tr>
<td></td>
<td>Unit 2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Taishan (Guangdong)</td>
<td>Unit 1</td>
<td>PWR(EPR)</td>
<td>2×1750</td>
</tr>
<tr>
<td></td>
<td>Unit 2</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

#### 3. REPROCESSING POLICY AND CURRENT SITUATION IN CHINA

China adopts the closed fuel cycle policy in order to meet the requirements of the healthy and sustainable development of nuclear power in China and to master the reprocessing technology.

The total accumulated spent fuel discharged from the reactor at present is 1300 tHM (most of them is stored at the reactor site, only very small part of them is transported to away-from-reactor storage facility in the reprocessing pilot plant by road). The projected spent fuel discharged from NPP is about 7500 tHM in 2020, 13000 tHM in 2025 for 40GW_e program.

China takes positive measures to carry out the R&D and project construction of the spent fuel reprocessing and recycling. A reprocessing pilot plant has been finished construction and is carried out the hot test in the early this year. The throughput of the pilot plant is 400 kg/d for head and end head, 300 kg/d for the chemical separation part. The storage capacity of the
pools is 550 tHM, and will be extended to 1300 tHM. The functions of the pilot plant are: 1) to demonstrate process, equipment and instrumentation for reprocessing of the spent fuel under hot condition; 2) to accumulate experiences on design, construction, commissioning and operation for a commercial reprocessing plant; 3) to supply plutonium for MOX fuel used in the experimental FBR.

The project for construction of the large-scale commercial reprocessing plant is in progress. The site investigation and pre-feasibility study have been done. The project is under approving by the state council. The capacity of the reprocessing is 800 tHM/y. the storage capacity for the pool is 3000 tHM (the first phase) and 3000 tHM (the second phase) respectively. The projected time schedule of hot test operation is 2025.

4. THE REPROCESSING AND RECYCLING TECHNOLOGY DEVELOPMENT STRATEGY IN CHINA

China has determined the development strategy of technical options for reprocessing and recycling [3].

Firstly, the spent fuel is reprocessed using the proven advanced PUREX process to recover uranium and plutonium; MA and FP entering HLLW are vitrified into solid waste; uranium and plutonium are fabricated into UOX fuel and MOX fuel respectively, and reused in the thermal reactor. This option can: 1) save natural U resource by 30%; and 2) reduce waste volume (disposal in the geological repository) to ¼ and waste radio-toxicity by about 10 times (comparing to direct disposal of spent fuel). This option is an internationally recognized technical approach for reprocessing and recycling and has a lot of safe industrial operation experiences at present.

Secondly, the spent fuel is reprocessed using the proven PUREX process to recover the uranium, plutonium and neptunium; TRU elements, LLFP and the heat-generating isotopes such as Cs-137 and Sr-90 will be separated using the HLLW partitioning process and transformed into oxide form products for interim storage and then burn in FR or ADS or disposal; plutonium is fabricated into MOX fuel, and reused in the thermal reactor first or then in the FBR. This option is transient and compromised solution for SFM. It will take less time to realize, but the process is more complicated.

Thirdly, the spent fuel is reprocessed using the integrated process considering recovery uranium, plutonium and neptunium; separation of actinide elements and LLFP, separation of actinides from lanthanides element; plutonium is fabricated into MOX fuel, and reused in the fast reactor; MA and LLPF are fabricated into target pieces, and burn/transmuted in FBR or ADS. This option can: 1) to enhance U resource usage rate greatly by about 50~60 times; 2) To reduce waste radio-toxicity by about 100 times and waste volumes very significantly; 3) to reduce isolation time from the biosphere for waste disposal.

The first large scale spent fuel reprocessing plant in china will adopt the first option after comparing and analyzing the fuel cycle technology status in china and world. That is: 1) the proven PUREX process; 2) vitrification for HLLW; 3) uranium and plutonium co-precipitation for non-proliferation purpose. The next reprocessing plant will choose the second or the third option depending on technology development in china. We will choose the international cooperation mode as one of all options to construct the first large scale reprocessing plant.
5. INTRODUCTION OF THE COMMERCIAL REPROCESSING PLANT

The main features are listed as follows [4]:

5.1. Capacity

Storage capability of spent fuel receipt and storage facility:

3000-6000 tHM (3000 t for the first phase, 3000 t for the second phase)

Capacity of reprocessing facility:

800 tHM/a Therein 4tU/d (for chemical separation parts)
5 tU/d (for head end and back end parts)

5.2. Parameters of spent fuel for design basis:

Initial U-235 enrichment 4.45% (by wt.)
Average burn up 45000 MWd/tU
Maximum burn up 55000 MWd/tU
Cooling time ≥8 years (reprocessing) ≥5 years (reception and storage)
Design basis BU for reprocessing 55000 MWd/tU

5.3. Form of product

UO$_3$ & (U+Pu)O$_2$ or PuO$_2$

5.4. The main technical requirements

(1) The reprocessing technology adopts a salt-free PUREX process with two cycles;
(2) Uranium and plutonium recovery rate should be more than 99.85%;
(3) Uranium and plutonium purification should satisfy with the product specification;
(4) HL solid wastes volume produced should be less than 0.5 m$^3$/tU (including maintenance technical wastes);
(5) Recovery and reuse of acid, water and spent solvent should be considered;
(6) A centralized equipment/vessel cooling system and heating system for whole plant should be considered;
(7) The digital control system should be adopted. The intelligent instrument and the on-line detect or monitor instrument are used as practicable as possible;
(8) A computer network management system is used for whole plant;
(9) Discharge of the gaseous and liquid waste should take consideration of the site condition;
(10) All effluences should be continuously monitored;
(11) Security and physical protection systems should be considered for whole site and nuclear material.
5.2. The main key process equipment selection

(1) Head end: Horizontal shear machine; continuously rotatory dissolver; Centrifugal separation device;
(2) Chemical separation: annular pulsed column for the co-decontamination cycle; Mixer-settler for the uranium purification cycle and spent solvent recovery; Pulsed column or centrifugal extractor for plutonium purification cycle;
(3) Uranium finishing: fluidized bed thermal denitrification reactor;
(4) Plutonium and uranium finishing: continuously operated precipitation, filtration and calcination device;
(5) HLLW vitrification: calcination furnace and meter (or cold crucible).

6. R&D OF SPENT FUEL PROCESSING TECHNOLOGY

The design, construction and operation of a reprocessing plant are complex and require not only well-demonstrated process technologies but also extensive engineering knowledge.

In order to ensure the reprocessing plant operation safe, reliable, and economic, the R&D of process technologies; plant engineering technique; key process equipment and material; process measurement and control; criticality, radiation, chemical, fire and explosion hazards control and prevention must be implemented.

We have an R&D program to master and enhance the reprocessing technology in China. Some examples are given below:

Institute of nuclear energy technology of Tsinghua University developed an innovative triakylphosphine oxides (TRPO) process to remove TRU elements from HLLW in 1992. The institute has carried out a hot test in pilot scale with genuine HLLW from the reprocessing plant for nuclear material production, which used TRPO process for separating TRU, CES (crown ether and calixrene crown ether) process for removal of Sr-90 and Cs-137 respectively, the DF is more than $1 \times 10^6$ for activity, 2000 for Sr and Cs. Now the R&D for HLLW from the NPP spent fuel reprocessing is being carried out. Experiment of Cyanex 301 process for separation Am from Lns have been done. The DF is more than $2 \times 10^6$ for activity.

R&D on partitioning is also launching in the China Institute of Atomic Energy (CIAE).

Study on the advanced Purex process and integrated separation process for processing is going on in CIAE. The advanced Purex process is a salt-free two-cycle Purex process using DMHAN (n,n-dimethyl hydroxylamine) as the reductant for Pu with MMH (methyl hydrazine) as the stabilizer, using AHA complex for uranium purification.

7. CHALLENGES FACED BY CHINA

Besides technical challenges for reprocessing faced by the world, China has faced the other unique challenges as well. Firstly, China has several kinds of reactor types, such as VVER, M310, CPR, AP1000, and EPR, Which will bring some of difficulties for spent fuel treatment, especially for spent fuel handling and shearing. Secondly, Due to the complex of the reprocessing technology, there is a long way to go to fully master the reprocessing technology, including design, construction, commissioning and operation of the large scale reprocessing plant in China. Thirdly, the dry storage of spent fuel except the Candu reactor is not yet considered. But since China has a rapid development of nuclear energy and the discharge rate of the spent fuel will increase rapidly. It is necessary to master the technology and build the dry storage facility to facilitate the choice of the storage type.
8. CONCLUSION

(1) Reprocessing using the Purex process has become a mature technology with considerable experience;
(2) Challenges for reprocessing are to achieve economic competitiveness, the adaptation of current technologies and plants to meet even more stringent national or international regulations and, to accommodate fuel performance increases;
(3) China has an ambitious and fast development nuclear power program to meet requirements of the economy development and global climate change protocol;
(4) The nuclear fuel cycle must be suitable for the nuclear energy sustainable development; the closed fuel cycle policy in china requires the reprocessing and recycling as the SFM option. To master the advanced reprocessing technology is becoming a must;
(5) The well-established Purex process will continue to be dominant in the near term, but the technical requirements to be dealt with will increase in severity with the advent of higher burnup and Mox fuels. 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;
(6) China has a desire of international cooperation to construct the first commercial reprocessing plant with the proven technology based on Purex process.

REFERENCES

THE KOREAN STRATEGY FOR NUCLEAR FUEL CYCLE

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Abstract

Since 1997, the Korea Atomic Energy Research Institute (KAERI) has been developing core pyroprocessing technology for effective SFM. Research goals are to increase equipment throughput and to minimize the volume of high level radioactive waste. The pyroprocess includes electroreduction, electrorefining, electrowinning, and a waste salt treatment system. This paper briefly addresses unit processes and related innovative technologies. Experiments on the unit processes were tested successfully, and based on the results, engineering-scale equipment has been designed for the PRIDE (PyRoprocess Integrated inactive DEmonstration facility).

1. INTRODUCTION

For the time being, PWRs will remain as the major source of nuclear power in Korea. However, the storage of the spent fuels produced from those PWRs is a big issue. The on-site spent fuel storage capacity will reach its limit by 2016. Therefore, a decision-making process for SFM is under way. Pyroprocessing is one of the promising technologies to treat a spent fuel and to reduce its volume. The basic concept of pyroprocessing is group recovery, which enhances the proliferation resistance significantly, as sole Pu cannot be separated. Pu with minor actinides, which comprise 1.4% of spent fuel, is recovered as a group. A part of U recovered from spent fuel is blended with this Pu and MA mixture, and the blended material is fabricated as the fuel of a sodium-cooled fast reactor (SFR). The Pu and MA mixture is burnt out in the SFR, resulting in transmutation of the long half-life nuclides. This reduces the waste management period by 1/1000 from 300,000 years. Cs and Sr, which are major components regarding heat load in spent fuel, are recovered during pyroprocessing, stored separately, and subsequently disposed of. The removal of transuranic elements (TRU), Cs, and Sr from spent fuel allows the repository burden to be reduced by 1/100, compared with the case without removal. The fission products (FP) are recovered and transferred to a repository. As a result of pyroprocessing, both repository efficiency and U usage are increase up to 100-fold.

2. UNITS OF PYROPROCESS

2.1. Electrolytic reduction process

As shown in Fig. 1, KAERI is currently developing an electrolytic reduction system, which is the front end of the pyroprocess, to demonstrate the laboratory scale operation of an electrolytic reduction process, and produce engineering data to be utilized in the design of the equipment, instruments, and facilities for the PRIDE.

In the electrolytic reduction process, the spent fuel oxide material from voloxidation is prepared for use in the electrorefiner by reducing the oxide to a metal [1]. The electrorefining process cannot accept oxide nuclear fuels directly, because the actinide oxides do not dissolve into the chloride salt. Also, in order to be used in an electrorefiner, materials must be electrical conductors. The metallic form produced by the electrolytic reduction process is suitable for use in the electrorefining process. In the electrolytic reducer, the feed material is placed in a fuel basket. A voltage is applied between the fuel basket cathode and a platinum
anode. This process uses a molten LiCl salt electrolyte with a small amount of Li$_2$O in solution at 650°C. Spent fuel metals are produced in the cathode basket and oxygen is evolved from the anode. Fission products, such as Cs, Sr, and Ba, are dissolved into the salt in this process, forming chloride compounds in the electrolyte salt. The metal that is collected in the cathode basket contains all of the actinides, lanthanides, and metallic fission products present in the spent fuel. After the electrolytic reduction process, the residual salt material is ~ 20 wt% of the cathode material. The cathode process for the removal of residual salts in the cathode basket of the electrolytic reducer can be added to ease the burden of the salt management process. The resulting metal is suitable for direct introduction into the electorefiner.

![Electrolytic reduction system](image)

**FIG. 1. Electrolytic reduction system.**

A new electrolytic reduction system equipped with a metal cathode basket, which can be linked to the electorefining process, was developed. The technology to suppress the vaporization of molten salts and enable reuse of the molten salts was verified. The bench-scale tests showed that the current density on the anode was increased from ~100 mA/cm$^2$ (old electrolytic reducer) to ~500 mA/cm$^2$ (new electrolytic reducer), thus enabling high speed electrolytic reduction. An electrolytic reduction run (17 kg UO$_2$/batch), using the new electrolytic reducer with a metal cathode basket, was successfully tested, showing an average anode current density of ~250 mA/cm$^2$ (See Fig. 2), and the basic design of an engineering scale electrolytic reducer (~50 kg UO$_2$/batch) was completed.
2.2. Electorefining process

The CERS (Continuous ElectroRefining System), which recovers pure uranium from reduced spent fuel, is composed of an electorefiner, a salt distiller, and a melting furnace. A U-chlorinator (UCI$_3$ making equipment) and a transportation system are also needed to operate the CERS. In the electorefiner, uranium deposition is initiated in molten LiCl-KCl salt with about 9 wt% UCI$_3$. Uranium dendrites are then deposited and fall from the electrode spontaneously [2], and are finally collected at the bottom of the reactor. The collected uranium deposits are transferred with a screw to a container. The uranium deposits are fed into a salt distiller, and salt in the uranium deposits is distilled by using a salt distiller. The salt-distilled uranium is melted and subsequently reformed to ingots for storage or for future use. The CERS was designed to continuously operate with a capacity of 20 kgU/day, as shown in Fig. 3.
The role of an electrorefiner is to recover uranium from the impure uranium mixture that is the product of the electrolytic reduction system. The currently installed HT (high throughput) ER is designed to have a capacity of 20 kgU/day, which has been simulated by ANSYS CFX [3]. A photograph of the HTER is shown in Fig. 4.

![FIG. 4. Continuous-type electro refiner equipped with graphite cathodes.](image)

The reduced uranium metal from the electrolytic reduction system is fed into an anode basket, which is placed at the periphery of the reactor. It rotates in order to enhance mass transfer. The cathodes, at which the uranium dendrites are deposited, are located in the core part of the reactor. At the center of the reactor is a scraper used to collect the uranium deposits that fall to the bottom of the reactor. The deposited uranium at the cathodes falls spontaneously from the solid cathode at an operation condition of 500°C. It is continuously conveyed by screw and transferred to a container. The remaining materials after depletion of the uranium from the feed material at the anode basket are noble metals, such as Fe and Mo.

2.1. **Electrowinning system**

Electrowinning technology is implemented to recover TRU from the molten salt system, a major process in the pyroprocessing technology, with proliferation resistance [4, 5]. Electrowinning technology of LCC is employed to recover group actinides such as uranium and TRU (Np, Pu, Am, Cm) in the molten salt (LiCl-KCl) transferred from the electrorefining process, which collects uranium of high purity. A cadmium distillation technology is also used to separate Cd and actinides from recovered actinide/Cd products by LCC, while residual actinide recovery (RAR) is incorporated for treatment of spent salt with low concentrations of actinides, as shown in Fig. 5. Finally, computer analysis technology is utilized to simulate the electrolytic process of the molten salt system for TRU recovery.
2.1.1. *Electrowinning reactor with liquid Cd cathode*

We designed and manufactured a lab-scale LCC electrowinner to evaluate the performance of LCC structures such as stirrer-type and mesh-type apparatuses. The experimental results of the performance tests using a mesh-type LCC structure showed that up to 8.4 wt% Cd of uranium was collected without the formation of uranium dendrites (Fig. 6).

![FIG. 6. LCC performance test results using a mesh agitator.](image)

2.1.2. *Cd distillation*

A distillation process was adopted for the separation of cadmium, since this type of physical separation process is more attractive than a chemical or dissolution process as it generates fewer secondary processes [6]. The cadmium separation takes place by evaporation of cadmium in the hot region and condensation of cadmium vapor in the cold region. The condensed solid cadmium is recycled to the electrowinning process for reuse. The apparent evaporation rate of pure cadmium was measured at various temperatures and pressures. The evaporation rate of pure cadmium was varied from 4.6–38.5 g/cm²/h in a temperature range of 500–650°C and in a vacuum pressure range of 0.5–10 torr. It increased with increasing temperature and decreasing pressure (Fig. 7). These experimental data should be very useful for designing the distiller and optimizing its operation.

![FIG. 7. Apparent evaporation rate of pure cadmium](image)
In a non-isothermal evaporation experiment, the onset evaporation temperature of cadmium alloyed with cerium was higher than that of pure cadmium (Fig. 8). It was inferred from the results that cadmium alloyed with other metals such as LCC deposits can be distilled at a higher temperature than pure cadmium.

2.1.3. RAR

For a promising pyro-partitioning process, it is necessary to find an effective method for the recovery of residual actinides from spent salt resulting from the electrowinning step prior to the removal of all the fission products in the waste salt treatment step. KAERI has established a residual actinides recovery (RAR) scheme by combining electrolysis using a LCC and oxidation of rare earth fission products using a CdCl₂ oxidant [7]. Eventually, the rare earth metal chloride products can be transferred to the waste salt treatment step. The same equipment used for LCC electrowinning can be used for RAR operation. Therefore, the RAR process has promising advantages, such as the use of compact equipment and a simple process, compared to the multi-staged counter-current reductive extraction process [8].
KAERI developed an original hybrid concept using a LCC and an oxidant based on the results of a thermodynamic approach. The approach consists of two steps. The first is a electrolysis using a LCC to collect all the residual actinides and some of the rare earth FPs so as to reduce the concentration of actinides in the molten salt. The second step is selective recovery of parts of the co-deposited rare earth FPs by oxidation (or chlorination) using CdCl₂ from a Cd alloy with the FPs, U, and TRU. Oxidation experiments involving the addition of CdCl₂ oxidant into a salt containing a molten Cd-metals alloy were carried out to confirm the residual concentration of the actinides in the salt was maintained at a value of less than 0.01 wt% (100 ppm). Fig. 9 shows cyclic voltammograms of the salt phase, which were monitored at a time interval of 30 minutes during the oxidation of Ce-U-Cd alloy with CdCl₂. Experimental results show that the residual concentration of uranium can be reduced to a value less than 100 ppm. Therefore, it was confirmed that this RAR process could be applicable to the selective removal of the actinides content from the spent LiCl-KCl salt [9].

![Cyclic voltammograms of the salt phase at a time interval of 30 minutes during oxidation of the Ce-U-Cd alloy.](image)

**FIG.9.** Cyclic voltammograms of the salt phase at a time interval of 30 minutes during oxidation of the Ce-U-Cd alloy.

### 2.1.4. Computational simulation

Multi physics electrochemical modeling, in a framework of computational fluid dynamics (CFD) code, has been proposed and dealt with in detail to simulate the electro-transport behavior in a molten-salt electrowinning system. The modeling approach utilized in this study is focused on the mass transport and current arising due to the concentration and the surface over-potential based on the cell configuration and molten-salt electrolyte turbulence. The electrowinning cell model demonstrated here has a concentrically arranged structure with an anode annulus surrounding a LCC crucible inside it (Fig. 10). This implementation with the unique feature of a potential-to-current algorithm could provide useful information for more realistic spatial variation of the electrochemical characteristics. This approach will be applied to the design of an engineering scale electrowinning system.
2.2. Waste salt regeneration and solidification system

During the pyroprocessing of LWR spent oxide fuels, two different types of waste salts are expected to be generated: (i) LiCl waste salt containing alkali and alkaline-earth (Group I/II) fission products (FPs) from the electrolytic reduction process and (ii) LiCl-KCl eutectic waste salt containing rare-earth FPs from the electrorefining (winning) process. Since these waste salts are radioactive, heat-generative, and highly soluble in water, they must be fabricated into durable waste forms that are compatible with the environment inside a geologic repository for a long time [10]. Current technology for disposing of waste salts from pyroprocessing involves non-selective total incorporation of waste salts in a zeolite matrix to form a ceramic waste form (glass-bonded ceramic waste form), which results in a significant increase of the final waste volume for disposal [11].

KAERI is working on two key R&D concepts for the development of innovative waste salt treatment technologies. The first is the minimization of waste salt generation by the removal of fission products in the waste and then recycling the cleaned salt to the main pyroprocesses. The second is an increase of safety during interim storage or final disposal by the fabrication of high-integrity final waste forms. Accordingly, KAERI has been developing various FPs
removal and waste solidification technologies such as melt crystallization, oxidative precipitation, and SAP/ZIT solidification (Fig. 11). The performance was found to be successful in small scale equipment. In addition, construction and performance evaluation of lab-scale equipment was completed in 2009. Research on modifications and resultant operation characteristics of the salt regeneration and solidification equipment will be performed in a stepwise manner according to KAERI’s long-term R&D plan, which is financially supported by the national long-term nuclear R&D program.

2.2.1. Waste salt regeneration

For the regeneration of waste LiCl salts, group I/II FPs involved in the waste salt are separated by using a melt crystallization process such as a layer melt crystallization method. This technology does not require any foreign additives or adsorption agents such as zeolite. The operation principle is based on the solubility difference of solutes (e.g. CsCl and SrCl$_2$) between molten salt and solid states. In this process, the group I/II radionuclides are concentrated in the melt phase, while the other solid phase contains relatively purified LiCl salt. This crystal phase contains a very small amount of group I/II FPs and is recycled to the electrolytic reduction process for reuse [12]. A preliminary experiment with a lab-scale apparatus showed that about 80-90% of total waste salt could be recovered in the form of a refined LiCl crystal containing less than 10-20% of the initial CsCl and SrCl$_2$ (Fig. 12).

![FIG. 12. Lab-scale layer melt crystallization equipment.](image)

In the case of rare-earth FPs involved in waste LiCl-KCl eutectic salts, they are removed from eutectic salt by oxidative precipitation and the refined salt is recovered by a salt distillation process (Fig. 13).

The rare-earth chlorides react with an oxygen bubble to convert their chemical forms into oxides or oxychlorides. After oxidation, if the molten salt settles for several hours, the oxidized rare-earth products are precipitated, and phase separation into an upper pure salt phase and a lower precipitate phase occurs [13]. The upper part can be directly recycled to the electrorefining process. The lower part is a mixture of rare-earth precipitates and residual salt. The residual salt is recovered as a pure salt by using a vacuum distillation process and is then recycled to the electrorefining process for reuse. Based on the characteristics of salt vaporization and condensation, a salt vacuum distillation and condensation apparatus
incorporating a distillation and condensation chamber was designed and its operation characteristics were determined [14]. This apparatus was subjected to the force of a temperature gradient (40°C to 920°C) at a reduced pressure (< 10 Torr) as a closed type of system. It was possible to collect the vaporized salts in only one spot, thereby minimizing salt loss and apparatus corrosion. The residual salts were vaporized and were completely separated from the rare-earth precipitates, and over 99% of the vaporized salts were recovered. During distillation, carry-over of rare-earth precipitates was not observed and the recovered salt had nearly the same composition as that of the fresh eutectic salt.

FIG. 13. Regeneration process of waste eutectic salt.

2.2.2. Waste solidification

From the regeneration process for dirty salts, two kinds of wastes, LiCl salt residue with concentrated Group I/II FPs and rare-earth oxides, are generated. For the immobilization of waste salt, a zeolite process, developed by Argonne National Laboratory in the U.S.A., is considered a practical method for LiCl-KCl salt. Also, sodium aluminophosphate glass is well known as a glass matrix that is compatible to metal chloride, especially NaCl-KCl. Zeolite-4A containing α-cages occludes metal chlorides and is converted into sodalite as a host phase for waste salt after heat-treatment. A phosphate glass with a relatively low melting point has a high compatibility with metal chlorides, treating the waste salt at relatively higher waste loading. However, one important disadvantage of the zeolite process is a substantial increase of the final waste volume due to a limitation of the maximum salt loading into the zeolite structure. Also, when using phosphate glass for waste salt, its chemical durability is about ten times lower than that of the zeolite method. To overcome this problem, KAERI has developed an alternative technology to the zeolite process. This process, called SAP solidification, uses a new inorganic composite of silicone, aluminium, and phosphorous oxide, which can provide a series of chemical routes to stable chemical forms for waste salts [15]. The SAP material is prepared by using a conventional sol-gel method, and then mixed and reacted with molten salt residue for 16 hours at 650°C. The reaction product undergoes a final heat treatment step with
the addition of glass frit for 4 hours at around 1,150°C to form a durable ceramic waste. The rare earth oxides separated from LiCl-KCl salt can be immobilized into a durable silicate glasses or specific ceramic materials, but severe processing conditions are required to obtain a consolidated form with high waste loading. To alleviate the processing conditions, an inorganic composite (ZIT), composed of transition metal oxides and phosphates, has been developed. The composite can convert the rare earth oxides into monazite (LnPO$_4$) and the mixture can be consolidated at about 1,100°C, resulting in a durable waste form with a high density and high waste loading.

We conducted experiments applying the SAP solidification technology, and found that it provided around 3 times higher loading of waste salt into the SAP structure, thus resulting in a smaller final waste volume, while maintaining higher chemical durability and thermal stability compared to the zeolite product. The inorganic composite (ZIT) for the immobilization of rare earth oxides can alleviate the processing conditions and provide a durable waste form. To realize the immobilization method, KAERI has developed a lab-scale solidification process that consists of crushing, pulverizing, mixing/reacting, and sintering equipment. Thus far, development of the unit equipment has been ongoing to obtain proper performance and the processing conditions have been investigated to abstract the scale-up factors. The unique waste forms fabricated from this process need to be further characterized and qualified to examine whether they meet the requirements for final disposal; this work is still in progress.

3. CONCLUSION

The main goals of pyroprocess R&D in KAERI are to increase equipment throughput and to reduce the volume of final waste to be disposed of. Application of innovative technologies such as adoption of a graphite cathode in the electrorefiner and waste salt regeneration by a crystallization method was tested. Based on the results of bench- and laboratory-scale tests, an inactive engineering-scale integrated pyroprocess (PRIDE) facility with a capacity of 10 tons-U per year is planned to be constructed by the end of 2011. PRIDE will be open for international collaboration, which will be used for testing the integrity of unit processes, the adoptability of remote operability, and safeguards. Active tests in an ESPF (Engineering Scale Pyroprocessing Facility) will follow.

ACKNOWLEDGEMENTS

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COMPLEX APPROACH TO STUDY PHYSICAL FEATURES OF URANIUM MULTIPLE RECYCLING IN LIGHT WATER REACTORS

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Abstract

A complex approach was developed for studying the evolution of isotopic composition of fresh and spent light-water reactor fuel, on the basis of simultaneous simulation of neutron-physical processes, nuclide composition evolution caused by irradiation, and selective molecular transfer of components in the separation cascade. The natural uranium specific consumption for recycled uranium re-enrichment was estimated depending on 232U content in the enriched product, and on the respective burnup. Conditions of achieving equilibrium concentrations of 232U were defined; it was shown that this would require the contents of 236U in fresh fuel to be limited.

1. INTRODUCTION

Multiple usage of recycled uranium within the closed fuel cycle of light-water reactors (LWR) would make it possible to extend the resource basis of nuclear energy and to achieve certain advantages compared to once-through fuel cycle, which involves disposal of spent nuclear fuel (SNF). These advantages are due to the following factors:

(1) Natural uranium economy due to returning the uranium recycled from SNF back into the fuel cycle;
(2) Reducing of waste amounts to be disposed of: since SNF contains over 90% of uranium, without this uranium (and without plutonium separated to be subsequently used in MOX fuel) only a small amount of radioactive waste would be left for disposal, and the need of disposal sites would diminish accordingly.

Multiple usage of uranium recycled from SNF is complicated by the presence of 236U and 232U isotopes produced in the course of fuel irradiation [1–3]. 232U has a considerable impact on radiation parameters of fuel produced from the recycled uranium, while 236U manifests itself by additional neutron consumption and deteriorated neutron-physical parameters of fuel (i.e. shorter fuel campaign). Maintaining these neutronic parameters would require the impact of 236U to be compensated by increased initial concentrations of 235U. In the same time, calculations show that residual contents of 235U in the spent fuel of VVER-1000 reactors exceed those in natural uranium up to burnups of 60 MW-day/kg [4]. These assessments indicate the expediency of recycled uranium multiple application.

The goal of the present investigation was evaluation of relative consumption of natural uranium for additional uranium enrichment depending on 232U concentration in recycled uranium.
2. COMPLEX APPROACH TO STUDY PHYSICS OF URANIUM RECYCLING

In present paper the combined approach was implemented on the base of simultaneous modeling of neutron-physical processes and processes of cascade isotope separation for analysis of physical problems of usage multi-recycled uranium for VVER-1000 reactor fuel cycle [4].

The following sequence of uranium fuel irradiation scenarios was considered:

Fresh fuel fabricated from natural uranium is irradiated in the core, cooled for 10 years and reprocessed. Recycled uranium from reprocessed SNF comes to the separation cascade for further enrichment. Natural uranium is supplied to this cascade, too. Then the resulting fresh fuel containing recycled uranium is returned into the core, irradiated, cooled and reprocessed, than recycled uranium sent to the separation cascade again, etc. The total of five consecutive recycled uranium cycles was considered.

The following set of uranium recycling scenarios is considered (Figure 1): a) with restrictions on $^{232}\text{U}$ concentration in product flow, $C(^{232}\text{U})=2\cdot10^{-7}\%$, b) with restrictions on $^{232}\text{U}$ concentration in product flow $C(^{232}\text{U})=5\cdot10^{-7}\%$, c) without restrictions on $^{232}\text{U}$ concentration.

![FIG. 1. Scenario of uranium multiple recycling.](image)

Quantitative calculations of fresh fuel composition containing recycled uranium were based on the quasi-ideal cascade model, which — along with natural uranium feed flow — also enhances an additional feed flow of recycled uranium incoming to the cascade (Fig. 2) [5,6].
There is $^{236}\text{U}$ in reprocessed product, so equivalent mass concentration of $^{235}\text{U}$ in the enrichment product stream is obtained from equation

$$C_{235\text{eq}}^p = C_{235\text{nat}}^p + K_{236}C_{236}^p$$

where $C_{235\text{nat}}^p$ is mass concentration of $^{235}\text{U}$ in the enrichment product obtained from natural uranium, and $C_{236}^p$ is mass concentration of $^{236}\text{U}$ in product flow. $K_{236}$ is compensation factor.

Heterogeneous model of pin fuel cell was used for neutron-physical calculations to obtain neutron induced reaction rates. Calculations were performed with the MCNP5 Monte Carlo code. Burnup calculations were performed with ISTAR depletion code, build in Russian Research Center “Kurchatov Institute”.

3. RESULTS AND DISCUSSION

Calculations showed (Figs 3−4, Tables 1−2) that higher burnup reduce the amounts of natural uranium saved by the use of the recycled one.

If there is no restrictions of $^{232}\text{U}$ content, the burnup increasing from 48 MW-day/kg to 60 MW-day/kg reduces natural uranium economy from 15.5% to 10%. If $^{232}\text{U}$ content is restricted, the respective natural uranium economy falls lower still. The very need to dilute $^{232}\text{U}$ to obtain its admissible concentrations reduces uranium economy to 11% (for the limitation of $C_{232}^p \leq 5 \times 10^{-7}$%) or 7% (for $C_{232}^p \leq 2 \times 10^{-7}$%), assuming the burnup of 48 MW-day/kg. Burnup increased from 48 MW-day/kg to 60 MW-day/kg reduces the saved share of natural uranium from 11% to 6.5% (for $C_{232}^p \leq 5 \times 10^{-7}$%) and from 7% to 3.5% (for $C_{232}^p \leq 2 \times 10^{-7}$%), respectively.

The basic factors reducing recycled uranium attractiveness for multiple usage (besides the burning of $^{235}\text{U}$) are: the need to dilute the recycled uranium with the natural one in order to reduce the content of $^{232}\text{U}$ (if this content in fresh fuel is restricted), and the need to compensate the additional neutron absorption by $^{236}\text{U}$. 

FIG. 2. The scheme of the separating cascade with an additional flow of a feed.
TABLE 1. NATURAL URANIUM CONSUMPTION FOR FIVE CONSECUTIVE RECYCLES WITH IRRADIATION TO 48 MW-DAY/KG AND INITIAL ENRICHMENT OF 4%

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<td>&lt;2E-7%</td>
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<td>5.655</td>
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</table>

Specific uranium consumption in case of 4%-enriched fuel fabricated from natural uranium only 6.378

**FIG. 3.** Specific natural uranium consumption (a), specific recycled uranium consumption (b), and specific separation work (c) for five consecutive recycles with irradiation to 48 MW-day/kg.
TABLE 2. NATURAL URANIUM CONSUMPTION FOR FIVE CONSECUTIVE RECYCLES WITH IRRADIATION TO 60 MW-DAY/KG AND INITIAL ENRICHMENT OF 4.4%

<table>
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</table>

Specific uranium consumption in case of 4.4%-enriched fuel fabricated from natural uranium only 7.032

FIG. 4. Specific natural uranium consumption (a), specific recycled uranium consumption (b), and specific separation work (c) for five consecutive recycles with irradiation to 60 MW-day/kg.
Besides the fact that the presence of $^{236}$U in fuel makes additional fuel enrichment necessary in order to reduce the parasitic neutron absorption, this isotope is also one of the factors causing the growth of $^{232}$U concentrations in spent fuel (Figs 5–7) during the next recycles. This is due to the fact that in reactor irradiation conditions this isotope (via the short-lived $^{237}$U having a half-life of about 7 days) is the precursor of $^{235}$Np, which, in its turn, precedes $^{232}$U through the chain of radioactive transformations $^{237}$Np (n,2n) $^{236m}$Np 0.48(β−) $^{236}$Pu ($\alpha$) $^{232}$U, and precedes $^{236}$U through the chain $^{237}$Np (n,2n) $^{236m}$Np 0.52(β+) $^{236}$U (i.e. $^{236}$U is, to a certain extent, a self-precursor).

Assessments show that the contribution of $^{237}$Np (n,2n) $^{236m}$Np reaction in the formation of $^{232}$U is about 10 times higher than all contributions from other reactions taken together. For example, Fig. 7 shows results calculated for the accumulation of $^{236}$U in VVER-1000 fuel cell during four years of irradiation plus short-term cooling, both with and without account of $^{237}$Np (n,2n) $^{236m}$Np reaction.

![Np-237 concentration in case of absence restrictions on U-232](image)

**FIG. 5.** $^{237}$Np content in VVER-1000 spent fuel for five consecutive recycles with irradiation to 48 MW·day/kg.

It should be noted that though the contribution of $^{237}$Np (n,2n) $^{236m}$Np 0.52(β+) $^{236}$U chain in the formation of $^{236}$U is certainly several degrees lower than that of $^{235}$U (n,γ) $^{236}$U reaction, nevertheless – since $^{235}$U in the (n,γ) reaction is a precursor of $^{236}$U, and with account of $^{236}$U compensation with additional $^{235}$U enrichment – all nuclear transformations caused by fuel irradiation in the aggregate result in $^{236}$U production rate exceeding the rate of its burnup.
FIG. 6. $^{232}$U concentration in VVER-1000 spent fuel for five consecutive recycles with irradiation to 48 MW-day/kg and no limitation of $^{232}$U content in fresh fuel.

FIG. 7. $^{232}$U concentration during irradiation in VVER-1000 fuel cell, with and without account of $^{237}$Np(n,2n)$^{236}$Np reaction.
Correlations registered in $^{232}\text{U}$ and $^{236}\text{U}$ isotopic contents in fresh and spent fuel (Figs 8–9) indicate that $^{236}\text{U}$ accumulated by the end of irradiation cycle serves as an additional source of $^{232}\text{U}$ formation during the next irradiation cycle.

The impact of $^{236}\text{U}$ on natural uranium consumption is to some extent reflected in Fig. 3 and Table 1, which shows a drastic, almost two-fold, reduction of natural uranium economy during the transition from the first recycle to the second one (respective consumption increases by 5%) for $P_{^{232}\text{U}^{2}} \leq 2 \cdot 10^{-7}\%$; a similar picture can be observed in Fig. 4 and Table 2 for $P_{^{232}\text{U}^{2}} \leq 5 \cdot 10^{-7}\%$.

**FIG. 8.** Content of $^{232}\text{U}$ and $^{236}\text{U}$ in fresh and spent fuel of VVER-1000 reactor during five consecutive recycles with irradiation to 48 MW·day/kg.

**FIG. 9.** Content of $^{232}\text{U}$ and $^{236}\text{U}$ in fresh and spent fuel of VVER-1000 reactor during five consecutive recycles with irradiation to 60 MW·day/kg.
However, this effect takes account only of the impact of $^{236}\text{U}$ as an additional $^{232}\text{U}$ source while transferring to the next recycle, and no account of the additional enrichment compensation effect. Since it is necessary to compensate the parasitic neutron absorption, the presence of $^{236}\text{U}$ also results in additional natural uranium and separation work expenditures. Tables 3–4 provide estimated additional separation work and natural uranium consumption for the case of unlimited $^{232}\text{U}$ contents.

**TABLE 3.** SPECIFIC NATURAL URANIUM CONSUMPTION AND SEPARATION WORK FOR FIVE CONSECUTIVE RECYCLES, WITH AND WITHOUT ACCOUNT OF $^{236}\text{U}$ COMPENSATION (BURNUP 48 MW-DAY/KG)

<table>
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<th>Specific natural uranium consumption</th>
<th>Specific excess consumption of natural uranium for $^{236}\text{U}$ compensation, %</th>
<th>Separation work amount, SWU</th>
<th>Excess separation work for $^{236}\text{U}$ compensation, %</th>
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</table>

**TABLE 4.** SPECIFIC NATURAL URANIUM CONSUMPTION AND SEPARATION WORK FOR FIVE CONSECUTIVE RECYCLES, WITH AND WITHOUT ACCOUNT OF $^{236}\text{U}$ COMPENSATION (BURNUP 60 MW-DAY/KG)

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<td>6.312</td>
<td>5.876</td>
<td>6.91</td>
<td>10.601</td>
</tr>
<tr>
<td>5</td>
<td>6.327</td>
<td>5.856</td>
<td>7.44</td>
<td>10.642</td>
</tr>
</tbody>
</table>
The data from table 3 shows that, for 4.0% enrichment and 48 MW-day/kg burnup, additional consumption of natural uranium determined by the need to compensate the presence of $^{236}\text{U}$ lies between 3.5% and 9% and additional separation work — between 4 and 10% (depending on the recycle number).

For 4.4% enrichment and 60 MW-day/kg burnup, additional consumption of natural uranium at the fifth recycle lies between 3 and 7.44%, and additional separation work — between 3 and 8%, depending on the specific recycle number. This slight reduction of additional natural uranium consumption compared to 4.0% enrichment and 48 MW-day/kg burnup is due to the fact that the specific consumption of recycled uranium in this case is lower (Figs 2–3), as well as to some specific features of separation processes running in the cascade, discussion of which is beyond the frames of this study.

In the same time, the results obtained (Figs 7 and 8) show that — due to the need to dilute the recycled uranium to admissible $^{232}\text{U}$ concentrations — the chosen recycled uranium re-enrichment pattern coupled with restricted $^{232}\text{U}$ concentrations in fresh fuel imposes natural restrictions on $^{236}\text{U}$ content in fresh fuel, which, in turn, restrict $^{232}\text{U}$ and $^{236}\text{U}$ accumulation in spent fuel and drive their concentrations to the equilibrium.

In the calculations performed within this study, the above effect was produced by mixing recycled and natural uranium in the separation cascade; however, in principle it could be also produced by $^{236}\text{U}$ separation. The latter case, due to possible growth of separation work amount, could result both in additional economy of natural uranium and in reduced accumulation of $^{232}\text{U}$ in the process of fuel irradiation, thus facilitating uranium multiple recycling, as well as improving the environmental conditions of the fuel cycle.

4. CONCLUSION

This study develops a complex approach to the analysis of evolving isotopic compositions of fresh and spent light-water reactor fuel, on the basis of coordinated simulation of neutronic processes, nuclide composition evolution caused by irradiation, and selective molecular transfer of components in the separation cascade.

The following regularities were analyzed using this complex approach:

(1) Impact of recycles’ number on specific natural uranium consumption and separation work amount, depending on burnup rate and initial fuel enrichment;

(2) Dependence of specific natural uranium consumption on limitations of $^{232}\text{U}$ content in fresh fuel;

(3) Specific features of achieving the equilibrium fuel composition under limitations of $^{232}\text{U}$ content.

Analysis of calculated results allows the following conclusions to be formulated in connection with the scenarios considered:

(1) Multiple usage of recycled uranium without any limitations imposed on the content of $^{232}\text{U}$ isotope allows natural uranium consumption to be reduced by about 16% and 10% for the respective burnup rates of 48 and 60 MW-day/kg;

(2) In case the content of $^{232}\text{U}$ is limited (by $2 \times 10^{-7}$ mass %), natural uranium economy reduces to 7% and 3.5% for the respective burnup rates of 48 and 60 MW-day/kg;

(3) Identified conditions of achieving equilibrium concentrations of $^{232}\text{U}$ show that this would require the contents of $^{236}\text{U}$ in fresh fuel to be limited (in the given study this
limitation occurred naturally as a result of recycled uranium diluted to admissible $^{232}$U concentrations;

(4) Excess amounts of natural uranium spent on $^{236}$U compensation, depending on the specific recycle number, lie between 3.5 and 9% for the burnup of 48 MW-day/kg, and between 3 and 7% – for the burnup of 60 MW-day/kg;

(5) Results of this study lead to the conclusion that development of $^{236}$U separation technology could reduce accumulation of $^{232}$U in the LWR fuel cycle and improve natural uranium economy.

REFERENCES


INTRODUCTION OF THE LARGE SCALE SPENT FUEL REPROCESSING PLANT IN CHINA

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Abstract

According to Chinese nuclear power development program, nuclear power has entered into the period of rapid development, which means that a large amount of spent fuel will be discharged from NPPs. China has always pursued the reprocessing technical route on the management policy of spent fuel. The construction of the large-scale spent fuel reprocessing plant is an important activity on the management of the spent fuel, and it is an important assurance of sustainable development of nuclear energy in China. China has accumulated a great deal of experience in construction of reprocessing plant for the production reactor spent fuel and the pilot reprocessing plant for power reactor spent fuel, established a technical team of R&D, design, construction and operation in reprocessing. Furthermore, Chinese government has approved the R&D program, further intensifying R&D efforts in reprocessing technology, which will make China confident that the large plant can be successfully constructed. At present, the project is implemented by China National Nuclear Corporation (CNNC). The preliminary feasibility study report and the project proposal have been submitted to the government, according to the results of the preparatory works. The product form, plant capacity, main technical options, site characteristics, R&D of reprocessing and several considerations for the project etc. are presented in this paper.

1. FOREWORD

Any country must consider the problems of SFM while developing nuclear power, China is no exception. At present, nuclear power in China has entered into the period of rapid development. According to the development target, the installed capacity of nuclear power will reach 40 GWe by 2020; the total inventory of the spent fuel discharged from NPPs will reach 7,000 t by 2020, and 20,000 t by 2030. The rapid development of nuclear power will make the problems of SFM more prominent. China has always pursued the reprocessing technical route on the management policy of spent fuel. Early in 1983, China had established the strategy that nuclear power development necessitates the development of reprocessing accordingly. The construction of the pilot plant for reprocessing the power reactor spent fuel (hereinafter referred to as “the pilot plant”) has begun since 1990s. In 2005, relevant Chinese government authorities organized a symposium on the construction of the large-scale NPP spent fuel reprocessing plant, and the construction of the large-scale spent fuel reprocessing plant (hereinafter referred to as “the large plant”) was put on government’s agenda for the first time. Later, the preparatory works start up completely. The construction of the large plant is an important activity on the management of the spent fuel, and it is an important assurance of sustainable development of nuclear energy in China.

2. BASIC INFORMATION OF THE LARGE PLANT

2.1. Product form

Product form of the large plant: UO$_3$ and (U)PuO$_2$. 

2.2. Plant capacity

According to the China’s nuclear power development program, the amount of discharged spent fuel will reach ~1300 t (U before irradiation, the same below), and the total amount of spent fuel accumulated will reach ~12800 t by 2025. In addition, through comprehensive analysis of the construction experiences in foreign commercial spent fuel reprocessing plants, reprocessing capacity of 800 t/a is suitable.

Therefore, the plant capacity is:

- Storage capacity: 3000–6000 tU
- Reprocessing capacity: 800 tU/a

2.3. Main technical options

2.3.1. Design basis spent fuel

The design basis spent fuel is AFA-3G spent fuel.

- Cladding material: M5 alloy
- Pellet material: UO2
- $^{235}$U initial enrichment: 4.45% (by weight)
- Burnup: (average) 45000 MWD/tU; (max.) 55000 MWD/tU
- Cooling time: ≥8 years

2.3.2. Process options

The wet unloading method will be used to unload the spent fuel into storage pool for storage. The main process option will be proved PUREX process, and the technical improvements since 1990s will be considered. The waste treatment facilities and the main process facility will be constructed at the same time, waste minimization will be ensured by advanced waste treatment process. At the same time, the measures such as liquid waste recycling, solvent rectification and nitric acid recovery, etc. will be fully considered.

2.3.2.1. Options of spent fuel receipt and storage

The wet unloading method will be used. The spent fuel assemblies will be stored in storage rack in storage pool considering the anti-seismic requirement.

Decay heat generated by the spent fuel assemblies in storage pool will be evacuated by pool water cooling and cleaning system, maintaining the temperature of the pool water not exceeding 40°C, keeping the radioactive concentration below the limit.

2.3.2.2. Reprocessing process options

(a) Mechanical and chemical head-end:
- Spent fuel assemblies will be chopped by horizontal shearing machine. The chopped sections of fuel will be continuously dissolved in dissolver. Dissolver solution will be clarified by settling centrifuge, after adjustment and precision measurement, prepared as 1AF feed solution;

(b) Chemical separation and purification:
- Chemical separation and purification of the main process will be performed by proved PUREX two cycle flow (Fig.1), main process steps include:
LARGE SCALE SPENT FUEL REPROCESSING PLANT IN CHINA

(i) Co-decontamination and separation: 1AF and 1AX counter currently contacted in 1A, U and Pu in 1AF feed solution extracted into organic phase, and most of FPs remain in raffinate, so U and Pu co-decontaminated from FPs. Pu stripped into aqueous phase by U4+ in 1B, so Pu separated from U. U stripped into aqueous phase by dilute nitric acid in 1C;

(ii) U purification: 2DF and 2DX counter currently contacted in 2D. U in 2DF feed solution extracted into organic phase, remove Pu, Np and Ru from U. U stripped into aqueous phase by dilute nitric acid;

(iii) Pu purification: 2AF and 2AX counter currently contacted in 2A, Pu in 2AF feed solution extracted into organic phase; remove U and FPs from Pu. Pu stripped into aqueous phase by reducing stripping solution (HAN).

(c) Finishing:

(i) U finishing: 2EU from U purification concentrated by evaporator, the concentrate denitrated by fluidized bed, finally converted to UO$_3$ product;

(ii) (U)Pu finishing: precipitation, filtration and calcination of (U)Pu oxalate, finally converted to (U)PuO$_2$ product.

FIG. 1 Block flow diagram of the main process of the large plant.
2.3.2.3. Radioactive waste treatment options

(a) Radioactive gaseous waste treatment:
Radioactive gaseous waste treatment system removes NOx, radioactive aerosols, C and I from process off-gas through scrubber, demister, dust remover, absorption column, HEPA and iodine filter, making them below release limit, then releasing through stack into atmosphere;

(b) Radioactive liquid waste treatment:
(i) Process HLLW treatment system: HLLW will be concentrated by evaporation, condensates will be evaporated twice, concentrates will be vitrified, the resulting condensates will be partly recycled into process, the remaining part will be treated in waste treatment facility;
(ii) Process ILLW treatment system: ILLW will be concentrated by evaporation, condensates will be evaporated again, concentrates will be combined into HLLW, the resulting condensates will be partly recycled into process, the remaining part will be treated in waste treatment facility;
(iii) Non-process IL-LLLW treatment system: The non-process acidic and alkaline ILLW will be concentrated by evaporation, concentrates will be cemented, condensates will be evaporated. The non-process LLLW will be concentrated by evaporation, concentrates will be cemented;
(iv) Spent solvent treatment system: the used solvents will be separated by vacuum flash evaporation and vacuum rectification. The distillates of TBP and kerosene will be made up to be recycled into process. The TBP residue will be pyrolyzed, the resulting ashes will be cemented;

(c) Radioactive solid waste treatment:
The various solid wastes from reprocessing plant will be monitored, classified and treated. The conditioned IL-LL solid waste packages and cemented forms will be directed to IL-LL waste disposal site for final disposal. The conditioned α and HLW solid packages will be temporarily stored pending final deep geological disposal.

2.4. Type selection of main equipment

The following types of main equipment will be used in the large plant:

| Shearing machine: | Horizontal shear machine |
| Dissolver: | Continuous dissolver |
| Feed clarification: | Settling centrifuge |
| Extraction equipment: | Pulsed extraction column, mixer-settler, centrifugal extractor |
| U finishing: | Fluidized bed |
| (U)Pu finishing: | Precipitator, filter and calcination furnace |

2.5. Construction planning

The spent fuel storage facilities will start to receive spent fuel in 2018, and the reprocessing facilities will start up in 2025.

3. PROGRESS OF THE PROJECT

At present, the project is implemented by China National Nuclear Corporation. The phase results have been obtained in the preliminary feasibility study and foreign cooperation, etc.
Siting is the key point of the preparatory works. Siting is performed both in the coast and inland area according to the requirements of nuclear safety laws. At present, the preliminary survey work of the inland site has been performed. The preliminary feasibility study works such as technical options, environment impact and safety assessment, investment estimation and economic analysis etc. have been performed simultaneously. The preliminary feasibility study report and the project proposal have been submitted to government for approval.

In order to speed up the construction, China welcomes the countries or the enterprises which have reprocessing ability to cooperate with China, if the conditions are permissible. At present, CNNC has contacted and discussed on the potential cooperation with relevant organizations and enterprises of Russia, France and UK.

4. SITE CHARACTERISTICS

The characteristics of the inland site are as follows:

— The site is located in Gansu province;
— The topography of the site is flat, and the slope of natural topography is about 1%. The landform is gently inclined Gobi plain and hilly ground;
— The site is far away from densely populated areas and the city of 100,000 people, the population density is very low within 20km;
— There are no industrial, agricultural, culture and health facilities within 15 km of the site.

According to the results of the preliminary survey work, this site meets the siting requirements of reprocessing plant.

5. R&D OF REPROCESSING

China had successfully constructed the reprocessing plant for the production reactor spent fuel, its technical performance and process approached or reached the international level of the time. At present, the hot test of the pilot reprocessing plant for power reactor spent fuel, which is self-designed and constructed by China, has gain preliminary success. China has preliminarily mastered the design and construction technology of the spent fuel reprocessing plant, and established a technical team of R&D, design, construction and operation of reprocessing.

However, the spent fuel reprocessing is a complicated system engineering. In order to improve the reprocessing technology of China, and assure the successful construction of the large plant, it is necessary to further increase the R&D strength of reprocessing technology. At present, Chinese government has approved the R&D program in order to research key reprocessing technology, break through the core technology in design, process, key equipment and materials, analysis, monitoring and control, nuclear and radiation safety, waste treatment etc., wishing to enable China’s reprocessing technology reach the world’s advanced level nowadays.
6. SEVERAL CONSIDERATIONS FOR THE PROJECT

6.1. Reprocessing object

The types of technology used in the constructed, being constructed and proposed NPPs are many due to historical reason, and the technical parameters of fuel assemblies are different. In addition, as the nuclear power technology is continuously upgraded, the performance of fuel assemblies is changing. How to plan the reprocessing of various types of the spent fuels is one of the problems must be considered while constructing the large plant.

One of the considered options is to reprocess AFA spent fuel in the first large plant, reprocessing the same fuel in batch mode. The spent fuel from future China’s main reactors will be reprocessed in the second large plant.

6.2. Product outlet

The main products of reprocessing plant are U and Pu, the main outlet of which is to recycle them in thermal or fast reactors after being manufactured into UO$_2$ or MOX fuel.

There are two options of industrial application for the Pu product: (i) first recycled into thermal reactors, then into fast reactors; (ii) directly recycled into fast reactors. Apparently, the efficiency of option (ii) is better. But which option will be adopted by that time will depend on the development of China’s fast reactors. Chinese government is undergoing the study for the problem of the outlet of the Pu product; one of the options is to recycle them into fast reactors directly.

7. CONCLUDING REMARKS

China has decided to construct a large-scale spent fuel reprocessing plant, which is an important activity on the management of spent fuel in China.

China has preliminarily mastered the design and construction technology of spent fuel reprocessing plant, established a technical team of R&D, design, construction and operation of reprocessing. Besides, Chinese government has approved the R&D program, further intensifying R&D efforts in reprocessing technology. China has confident that the large plant can be successfully constructed.

In order to speed up the construction, China welcomes the countries or the enterprises which have reprocessing ability to cooperate with China, if the conditions are permissible.
Abstract

There is renewed domestic interest in establishing spent nuclear fuel recycle in the U.S. after about a 30 year hiatus. Introduction of safe, proliferation-resistant, and economical civilian nuclear fuel cycles, especially the reprocessing step, in the U.S. poses numerous technical, social, and regulatory challenges. Initially, fuel recycle activities are expected to focus on light water reactor fuels, but it is anticipated that recycle of fuel from advanced reactors such as liquid-metal-cooled reactors and gas-cooled reactors will follow. Proposed reprocessing technologies include processes for removing heat-producing and high-risk fission products and actinides from waste streams prior to disposal. Proposed reprocessing processes and operations raise a range of issues identified in this paper that would require new and revised regulations to effectively and efficiently ensure their safety. The NRC prepared a report (NUREG-1909) documenting the background, status, and potential future issues concerning recycle of spent nuclear fuel that is summarized in this paper. In response to the issues, the NRC Commissioners, and other stakeholders, the NRC staff has conducted two analyses to identify and prioritize regulatory gaps for spent fuel reprocessing facilities and held public meetings to obtain stakeholder input. The NRC staff is now working on a revised regulatory framework for reprocessing facilities with a goal of completing the revisions by FY 2012. This paper summarizes the contents of NUREG-1909 and the activities of the NRC staff to update the regulatory framework in to address the issues that have been identified.

1. INTRODUCTION

A desirable method for disposition of the spent nuclear fuels (SNF) from the 104 operating commercial Light Water Reactors (LWRs) and from projected future reactors in the U.S. would meet the following goals: 1) increasing utilization of fissile and fertile materials by recycling them, 2) reducing the risk of proliferation by avoiding production of a pure plutonium stream, 3) reducing release of radionuclides that are predicted to result in the highest dose to humans, e.g., Np-237, Te-99, and I-129, from repositories, and 4) remove long-lived, heat-producing radionuclides, e.g., Cs-137, Sr-90, and Am-241 from high-level waste going to repositories. To meet these goals the Department of Energy (DOE) is conducting a research and development initiative to develop innovative recycle processes leading ultimately to advanced nuclear fuel recycle facilities; advanced nuclear reactors for irradiating neptunium, plutonium, americium and curium; and a fuel cycle research facility to develop advanced recycle technologies.

In response to DOE’s fuel recycle initiative the Nuclear Regulatory Commission’s (NRC’s) Advisory Committee on Nuclear Waste and Materials prepared a white paper on advanced recycle facilities [1] summarized in this paper that covered the following topics:

- Nuclear fuel designs;

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1 Recycle involves SNF reprocessing, fabrication of new fuel, management of wastes generated by these activities, and storage of SNF and wastes.
• Historical, current, and advanced SNF recycle processes and facilities;
• Recycle facility siting and design;
• Advanced spent fuel recycle initiatives;
• Regulation and licensing of fuel recycle facilities;
• Important issues associated with licensing and regulation of fuel recycle facilities and operations.

In addition, appendices were included that covered a description of the Purex process in the Barnwell nuclear fuel plant and estimates of the volume of and radionuclide distribution in wastes from advanced aqueous reprocessing facilities.

The GNEP provided the initial impetus for the NRC to begin considering the adequacy of the existing regulatory framework for SNF recycle in 2006. In 2008 the GNEP was redirected by Congress so that it does not involve near-term deployment of industrial-scale recycle facilities but subsequently nuclear industry companies provided the impetus for the NRC’s activities by indicated their intent to submit a license application for a SNF reprocessing facility in the 2013-2014 timeframe. Based on the results of the white paper, direction from the NRC Commissioners, and input from stakeholders outside the NRC, the NRC staff has initiated an effort to promulgate the regulatory framework by 2013.

The NRC’s regulations, sometimes called rules, impose requirements that licensees must meet to obtain or retain a license or certificate to use nuclear materials or operate a nuclear facility. These regulations govern the transportation of materials; the use of materials at such nuclear facilities as power plants, research reactors, uranium mills, fuel facilities, and waste repositories; and the use of materials for medical, industrial, and academic purposes. The process of developing regulations is called rulemaking. The NRC’s effort to revise the regulatory framework for reprocessing will be accomplished primary through the rulemaking and guidance development process.

Activities that have been completed and which are discussed in this paper are:

• Performing a first-order analysis to identify regulatory gaps;
• Performing a more detailed follow-on regulatory gap analysis that identified and prioritized 23 gaps in four categories;
• Held public meetings on a white paper by the Nuclear Energy Institute (NEI) and on revising the regulatory framework.

Subject to budget considerations, the NRC staff plans to complete the revised regulatory framework in FY 2012 using a process that provides extensive opportunities for public involvement.

2. HISTORICAL OVERVIEW OF SNF REPROCESSING

2.1. U.S. Defence reprocessing plants

SNF reprocessing started as a U.S. wartime activity for the recovery of plutonium from production reactors for use in nuclear weapons. Major fuel reprocessing plants were constructed in Washington at the Hanford site and in South Carolina at the Savannah River site. The first large-scale plutonium reprocessing was carried out using a precipitation process based on very small scale chemical studies using bismuth phosphate as a carrier for plutonium (the Bismuth Phosphate Process). Subsequently improved processes based on solvent
extraction were developed and implemented. The first large-scale solvent extraction process was based on methyl isobutyl ketone as a solvent (the HEXONE process); subsequently the HEXONE process was displaced by an improved process based on solvent extraction of plutonium and uranium into tri-n-tributyl phosphate, commonly known as TBP (the PUREX process). The Bismuth Phosphate, HEXONE and PUREX processes produced large amounts of wastes\(^2\) that have proven to be difficult to manage as attempts are made to empty the large waste storage tanks during site clean-up efforts. After WWII the Atomic Energy Commission (AEC) encouraged commercial SNF reprocessing in the U.S. and the U.S. government encouraged reprocessing overseas.

2.2. U.S. commercial reprocessing plants

Partly as a consequence of encouragement by the U.S. government, three commercial reprocessing initiatives were started. All three of these plants were based on the PUREX Process, as are all commercial reprocessing plants worldwide. Figure 1 is a greatly simplified diagrammatic representation of the universally used PUREX Process.

The first commercial reprocessing plant was the Nuclear Fuel Services West Valley Plant. This plant was built in western New York and had a nominal throughput of 300 metric tons of initial heavy metal (MTIHM) per year. This plant operated for about seven year until required upgrades proved to be too costly and the plant was shut down. The second plant was the General Electric Plant in Illinois. This plant had design flaws and was only operated with uranium during “cold” tests before being shut down. The third plant was the Barnwell Nuclear Fuel Plant with a projected capacity of 1500 MTIHM per year. As a result of proliferation concerns, actions were taken by Presidents Ford and Carter that led to the plant never being licensed or operated with SNF.

2.3. International reprocessing plants

Other countries did not follow the U.S. lead in eschewing SNF reprocessing and, in fact, aggressively pursued reprocessing, both for weapons production and for commercial gain. At the present time France, U.K., Russia, Japan, India and China have publicly declared operating reprocessing plants and Israel is known to have an undeclared plant. Germany and Belgium had reprocessing plants that are now inactive or shut down. Both France and U.K. operate commercial reprocessing plants for both foreign and domestic SNF. Table 1 summarizes the capacity of civil (non-weapons) reprocessing plants that are operating or planned.

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\(^2\) The wastes in the high-level waste storage tanks are predominantly sodium nitrate, sodium hydroxide, iron oxides, and sludges comprised of oxides/hydroxides of the fission products and actinide elements.
3. CURRENT REPROCESSING ACTIVITIES

There are several types of SNF from a variety of different reactor types being processed in commercial reprocessing plants. The most common SNF is from LWRs, of which the most prevalent is the PWR. The next most prevalent reactor type is the boiling water reactor (BWR). These reactors both use uranium oxide fuel in Zircaloy clad fuel rods supported by hardware composed of Zircaloy, Inconel, and stainless steel.

BWR fuel is similar to PWR fuel in most respects except that its horizontal dimensions are smaller and its sides are surrounded by a sheet of Zircaloy. From the point of view of reprocessing it is very nearly the same as PWR fuel. Reprocessing, as illustrated in Fig. 1 for the PUREX process, involves dissolving the uranium dioxide “meat” in nitric acid and extracting uranium and plutonium into TBP. Other actinide elements and fission products remain in an acidic waste stream. The hardware accounts for about one fourth of the total weight of an assembly and comprises a separate radioactive waste stream that must be disposed of. During the SNF reprocessing dissolution step significant quantities of radioactive gases such as Kr-85, I-129, and Carbon-14 dioxide are released into the plant off-gas system. Tritium (H-3 or T) reacts with water to produce HTO unless removed as T₂ gas prior to dissolution.

FIG. 1. Greatly simplified diagrammatic representation of the PUREX process.
3. ACCIDENTS OCCURRING IN RECYCLING SNF

Spent nuclear fuel reprocessing has not been without accidents resulting in the release of radioactivity. In addition, deliberate release of Kr-85 and C-14 has been practiced as has release of some radionuclides from reprocessing plants abroad into the sea. Table 2 lists occurrences and types of some accidents at SNF reprocessing plants and sites.

### TABLE 1. OPERATING AND PLANNED REPROCESSING PLANTS IN OTHER COUNTRIES

<table>
<thead>
<tr>
<th>Country</th>
<th>Location</th>
<th>Scale</th>
<th>Rated Capacity, MTIHM/yr</th>
<th>Source of Feed Material</th>
</tr>
</thead>
<tbody>
<tr>
<td>China</td>
<td>Lanshou</td>
<td>Pilot Plant</td>
<td>0.1</td>
<td>PWR.HWRR</td>
</tr>
<tr>
<td>France</td>
<td>LaHague UP2-800</td>
<td>Commercial</td>
<td>850</td>
<td>LWR</td>
</tr>
<tr>
<td>France</td>
<td>LaHague UP3</td>
<td>Commercial</td>
<td>850</td>
<td>LWR</td>
</tr>
<tr>
<td>India</td>
<td>Kalpakkam Reprocessing Plant (KARP)</td>
<td>Demonstration</td>
<td>100</td>
<td>PHWR</td>
</tr>
<tr>
<td>India</td>
<td>Lead Minicell Facility (LMF)</td>
<td>Pilot Plant</td>
<td>n/a</td>
<td>FBTR</td>
</tr>
<tr>
<td>India</td>
<td>Power Reactor Fuel Reprocessing Plant (PREFRE)</td>
<td>Demonstration</td>
<td>100</td>
<td>PHWR, LWR</td>
</tr>
<tr>
<td>India</td>
<td>Fast Reactor Fuel Reprocessing Plant</td>
<td>Commercial</td>
<td>n/a</td>
<td>FBTR</td>
</tr>
<tr>
<td>Japan</td>
<td>Rokkasho Reprocessing Plant</td>
<td>Commercial</td>
<td>800</td>
<td>LWR</td>
</tr>
<tr>
<td>Japan</td>
<td>JNC Tokai Reprocessing Plant</td>
<td>Demonstration</td>
<td>210</td>
<td>LWR</td>
</tr>
<tr>
<td>Russia</td>
<td>Research Institute Atomic Reactors (RIAR)</td>
<td>Pilot Plant</td>
<td>1</td>
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</tr>
<tr>
<td>Russia</td>
<td>RT-1, Combined Mayak</td>
<td>Commercial</td>
<td>400</td>
<td>VVER-440</td>
</tr>
<tr>
<td>United Kingdom</td>
<td>British Nuclear Fuels Limited (BNFL): B205</td>
<td>Commercial</td>
<td>1500</td>
<td>U Metal (MAGNOX)</td>
</tr>
<tr>
<td>United Kingdom</td>
<td>BNFL Thermal Oxide Reprocessing Plant</td>
<td>Commercial</td>
<td>1200</td>
<td>LWR, AGR OXIDE</td>
</tr>
</tbody>
</table>


4. STATUS OF THE EXISTING REGULATORY FRAMEWORK FOR REPROCESSING

There are a number of existing regulations for licensing fuel reprocessing facilities. Under current U.S. government regulations, various parts of a SNF reprocessing facility would have to meet the requirements of following regulations: 10 CFR Part 50, 10 CFR Part 70, 10 CFR Part 30, 10 CFR Part 40, 10 CFR Part 73 and 10 CFR Part 74. The primary licensing
regulation, 10 CFR Part 50 “Domestic Licensing of Production and Utilization Facilities,” has evolved to focus on licensing LWRs. The NRC has used Part 70 to license fuel fabrication facilities, and this regulation was the basis for licensing the mixed oxide fuel fabrication plant in Wilmington, SC. 10 CFR Part 70 allows for a one-step licensing process.

While licensing under these regulations is possible, the result would be both inefficient and complex because the regulations would have to be substantially modified to account for the features of a reprocessing plant (e.g., significant amounts of reactive chemicals) while still being applicable to other facilities. These differences lead to the need for substantial changes to existing regulations or to new regulations. In 2007 the NRC directed its staff to begin developing the primary regulatory framework to license SNF reprocessing facilities using an option based on 10 CFR Part 70, “Domestic Licensing of Special Nuclear Material.” Advisory committees to the NRC have recommended that a risk-informed, performance-based approach based on a probabilistic risk assessment (PRA) is preferred for licensing a future reprocessing. In a risk-informed, performance-based regulatory approach risk is an important consideration, but other things such as cost and environmental considerations should be balanced against risk reduction. The NRC has defined risk-informed regulation [4].

<table>
<thead>
<tr>
<th>Type of accident</th>
<th>Liquid releases</th>
<th>Gaseous releases</th>
<th>Occurrence</th>
</tr>
</thead>
<tbody>
<tr>
<td>Criticality in dissolver tank</td>
<td>X</td>
<td>X</td>
<td>Windscale, 1973 Tokai, 1999a</td>
</tr>
<tr>
<td>Leak of discharge pipe; breach in a tank</td>
<td>X</td>
<td>La Hague, 1979-80 Sellafield, 1983</td>
<td></td>
</tr>
</tbody>
</table>

aThe September 1999 accident at Tokai Mura did not involve a reprocessing plant but is a type of accident that could occur in a reprocessing plant.

5. ISSUES ASSOCIATED WITH LICENSING AND REGULATING ADVANCED FUEL RECYCLE FACILITIES IDENTIFIED IN THE WHITE PAPER

In addition to establishing the approaches to use for the primary licensing regulations for fuel recycle facilities it will be necessary to evaluate the impact that fuel recycle facilities and operations may have on other regulations that may be invoked or need to be developed. Fuel recycle facilities and other aspects of recycle that fall into this category include:
• New products, effluents, and wastes: cladding waste, Kr-85, I-129, C-14, H-3, recovered uranium, and transuranic (GTCC) wastes;
• Waste classification: determining the class of some of the novel wastes in the foregoing bullet in the present waste classification system;
• Waste forms: how to stabilize long-lived, mobile radionuclides (I-129, C-14) or non-reactive radionuclides (e.g., Kr-85);
• Distribution of radionuclides in various streams: how do key radionuclides such as I-129 distribute in the process streams of a highly integrated reprocessing facility;
• Disposal technology: identification of an appropriate disposal technology for wastes containing the radionuclides identified in the first bullet and revised or new licensing regulations for the disposal facility;
• Repository licensing: impacts of separate waste forms containing the radionuclides in the first bullet and the substantial reduction in heat load on the repository if essentially no cesium, strontium, and the actinides are in the wastes sent to the repository;
• Uranium handling and disposal facilities: licensing regulations for facilities to recycle or dispose of recovered uranium containing U-232, U-236, and trace contaminants;
• Cesium and strontium storage and disposal: licensing regulations for the long-term storage facility contemplated by DOE to allow radiocesium and radio strontium to decay to Class C concentration and for in situ closure of the facility or alternative disposal of its contents;
• Transuranium element storage and disposal: safeguards, security, and safety for transuranium elements containing substantial amounts of americium and curium stored awaiting recycle;
• Integrated plant performance of very complex plants: how to account for the many recycle streams in highly integrated facilities;
• Decontamination and decommissioning (D&D): how to design and operate the plant to facilitate D&D;
• Sigma inventory differences: reconciling inconsistencies among IAEA, NRC and DOE requirements for limits on the permissible significant (sigma) plutonium inventory differences.

6. OTHER IMPORTANT ISSUES RELATED TO LICENSING

In the 1970s DOE’s predecessor agencies began a generic environmental impact statement (GEIS) and the Environmental Protection Agency (EPA) began to develop standards for radionuclide releases and environmental radiation protection standards for reprocessing facilities. The GEIS effort ended with the publication of the GESMO document, but limitations in the scope of GESMO advances in technology and risk assessment techniques during the last few decades make it essentially irrelevant to what DOE and the commercial SNF recycle industry is contemplating. DOE initiated a proposal [5] for a follow-on Programmatic Environmental Impact Statement (PEIS) but this was recently suspended.

EPA’s efforts ultimately produced Title 40, Part 190 of the Code of Federal Regulations (40 CFR 190). Of special relevance to reprocessing is 40 CFR 190.10(b) which limits release of Kr-85 and I-129. The NRC adopted the same limits as 10 CFR 20.1301(e). Studies of release limits for C-14 and H-3 had begun but ceased when President Carter decided to defer commercial SNF reprocessing. Because the technologies of obtaining the specified EPA release limits have not been industrially demonstrated they may be uneconomical or impractical for commercial practice in the near term.
7. REVISING THE REGULATORY FRAMEWORK FOR LICENSING A SPENT NUCLEAR FUEL REPROCESSING FACILITY

8.1. The global nuclear energy partnership and spent fuel reprocessing

In February 2006, the Administration announced the Global Nuclear Energy Partnership (GNEP), part of the President’s Advanced Energy Initiative aimed at advancing three identified ways to meet the challenge of generating more electricity - clean coal technology, advanced emission-free nuclear power, and renewable resources, such as solar and wind. The international aspect of GNEP involved the development of a framework to manage spent nuclear fuel, enhance the nuclear energy option, and provide safe and secure nuclear power to foreign countries.

In August 2006, the Department of Energy (DOE) sought expressions of interest from the nuclear industry for constructing spent fuel reprocessing and transmutation fuel fabrication facilities. DOE subsequently established contracts with four industry consortia headed by EnergySolutions, International Nuclear Recycling Alliance (INRA), General Atomics, and General Electric-Hitachi (GEH), for the purpose of assessing options to close the nuclear fuel cycle in the U.S. In January 2008, each of the consortia provided DOE conceptual designs, cost bases, and schedules for the Consolidated Fuel Treatment Centre (CFTC), a recycling centre that would reprocess SNF, and the Advanced Burner Reactor (ABR), a new reactor that generates electricity while converting long-lived transuranic radionuclides contained in fuel manufactured at the CFTC into short-lived radioactive waste [6, 7, 8, 9]. The consortia updated their proprietary submittals and provided these to DOE on June 30, 2008. DOE subsequently provided these to the NRC.

The four proposals DOE received from various industry consortia ranged from aqueous based reprocessing methods with light water reactor (LWR) recycling of the fuel (as mixed oxide {MOX} fuel) to pyroprocessing technologies with ABR recycling of the fuel.

In FY 2009 Congress redirected all funding of domestic GNEP and directed DOE to transition its focus from a combination of R&D with near-term commercial deployment of recycling facilities and a fast reactor demonstration project to a purely R&D approach under the long-term, science-based Fuel Cycle Research and Development (FCR&D) program which includes recycling.

8.2. Revising the regulatory framework after the domestic GNEP program

Initially, NRC believed that a new U.S. reprocessing facility would be the result of the DOE’s GNEP program. Before investing significant NRC resources, the Commission directed the NRC staff to await the Energy Secretary’s decision regarding a path forward for the chosen spent fuel reprocessing technology. The Energy Secretary deferred indefinitely making a decision on GNEP.

In mid-2008, two nuclear industry companies informed the agency of their intent to seek a license for a reprocessing facility in the U.S. An additional company expressed its support for updating the regulatory framework for reprocessing, but stopped short of stating its intent to seek a license for such a facility. At the time, the staff also noted that progress on some GNEP initiatives had waned and it appeared appropriate to shift the focus of the staff’s efforts from facilities specific to GNEP to a more broadly applicable regulatory framework for commercial reprocessing facilities.

As a result, the staff concluded GNEP should no longer be the impetus for considering
reprocessing-related activities and it was appropriate to devote limited resources at a pace consistent with industry interest and commitment, to develop an appropriate, effective, and efficient regulatory framework for licensing a potential commercial spent nuclear fuel reprocessing facility.

The NRC still maintains an interagency agreement (IA) with DOE to allow NRC staff to develop its knowledge base in the area of recycling and provide DOE regulatory insights into key safety, safeguards and security topics to inform DOE’s FCR&D program. The IA was recently extended to September 30, 2010.

8.3. Industry initiatives associated with spent nuclear fuel reprocessing

In July 2008, NRC staff was made aware that the Nuclear Energy Institute (NEI) had established its “Closing the Fuel Cycle Task Force”. The task force’s primary objective is to facilitate implementation of a regulatory structure to license reprocessing facilities and the associated fuel fabrication facilities. A secondary objective is addressing the regulatory framework for the ABR. The task force initially determined that the primary objective needs to be implemented by 2012 to be in alignment with industry’s interest/intent. The NEI task force has prepared a white paper on the regulatory framework for recycling SNF [10] and the NRC held a public meeting to obtain comments on the paper in February 2009.

9. REGULATORY FRAMEWORK DEVELOPMENT

9.1. Initial regulatory gap analysis

Currently, 10 CFR Part 50, “Domestic Licensing of Production and Utilization Facilities”, provides the licensing framework for production and utilization facilities. Although a reprocessing facility is one type of production facility, its industrial processes are more akin to fuel cycle processes than to production or utilization facility processes, such as a power reactor. Most fuel cycle facilities are licensed to operate under 10 CFR Part 70, “Domestic Licensing of Special Nuclear Material”. Therefore, in accordance with Commission direction to the NRC staff [3], the staff completed an initial regulatory gap analysis [11] focused on necessary changes to 10 CFR Part 70 considering requirements, where appropriate, from Part 50, as the basis for a revised reprocessing regulatory framework. The goal of this “gap” analysis was to identify the regulatory gaps and not to devote NRC resources to identify potential solutions.

The results of first-order gap analysis confirmed that: 1) although current regulations could be used to license a reprocessing facility, new regulations will be required to enhance the effectiveness and efficiency of regulatory oversight and 2) it would neither be effective nor efficient to revise Part 50 to license modern reprocessing facilities.

Over the years, Part 50 has essentially evolved to be a LWR specific regulation and the resources needed to address a modern reprocessing facility in Part 50 would be extensive and not timely considering industry’s interest. Furthermore, results of the regulatory gap analysis indicated that Part 70 currently does not adequately address all the hazards associated with the reprocessing of spent nuclear fuel including but not limited to, an increase in radiological risk and the complex process streams which are different than the uranium fuel processing facilities process streams for which Part 70 was most recently revised in 2000. The hazards and risks will manifest themselves in different ways throughout a reprocessing process and their contributions will be considered as staff prepares the technical basis document and
begins the development of new regulations. NRC staff has considered examples of technology-neutral regulatory frameworks currently under consideration. These examples were considered for their applicability for reprocessing facilities. Additionally, the staff considered that the existing Part 70 currently regulates many different types of fuel cycle facilities and provides a model of a regulation capable of licensing several different types of facilities. As such, the NRC staff believes that it is possible to either include a new subpart to Part 70 that would provide new regulatory requirements for reprocessing facilities, or create a new Part specific for reprocessing. These new regulations could be capable of licensing aqueous separation techniques, and, potentially, non-aqueous techniques. Further, the unique design and safety issues associated with a reprocessing facility could be efficiently tailored and consolidated in a new Part without unnecessarily complicating the existing 10 CFR Parts 50 and 70. The form of the new regulations will be determined upon completion of the technical basis document.

9.2. Second regulatory gap analysis

The second regulatory gap analysis was completed in March 2009 [12]. Building on the first-order gap analysis, it considered several other documents, including: NUREG-1909 [1], correspondence from the Union of Concerned Scientists [12], and the NEI white paper [10]. The results are discussed in detail in [13]. Briefly, the staff identified 23 regulatory gaps and categorized each of the identified gaps in one of four categories:

(i) Lack of regulations;
(ii) Existing regulations pose a significant hindrance or regulatory burden to effective and efficient licensing;
(iii) Gaps resulting from potentially licensing a production facility under Part 70 (versus Part 50);
(iv) Requirements exist, but modifications may be needed for clarity.

Fourteen gaps were identified a high priority gaps; five identified as moderate priority gaps; and, four identified as low priority gaps. High priority gaps are those that must be resolved to establish an effective and efficient regulatory framework. An example of a high priority gap is Gap 2, “independent storage of high level waste” which describes the lack of available independent waste storage options to accommodate solidified high level waste. The staff will pursue high priority gaps in the technical basis development to follow.

Moderate priority gaps are those that should be resolved, but are not essential, at this stage. An example of a moderate priority gap is Gap 15, “Waste confidence for reprocessing facilities”. Gap 15 details that the existing waste confidence rule does not apply to reprocessing facilities. Because applicants for reprocessing facility licenses can address long-term storage of their waste in their environmental reports, resolution of Gap 15 was not determined to be essential at this point. However, the effectiveness and efficiency of the regulatory process could be enhanced by resolving this gap through rulemaking. Moderate priority gaps will be addressed in the technical basis development, in conjunction with the high priority gaps, if sufficient resources are available.

Low priority gaps could be resolved, but are determined to be not essential. An example of a low priority gap is Gap 20, “Advanced fuel cycles and transuranic special nuclear material (SNM) classification”. Gap 20 details the need to expand SNM requirements to other materials in order to accommodate reprocessing technologies. The Commission did not previously support this expansion, as stated in the Staff Requirements Memorandum to
SECY-08-0059 [14], and this gap will not be pursued in the reprocessing technical basis. Staff has determined that for the reprocessing framework development, low priority gaps are not essential and will not be pursued in the technical basis development, unless the Commission directs the staff to do so.

9.3. The rulemaking process

The NRC’s regulations, sometimes called rules, impose requirements that licensees must meet to obtain or retain a license or certificate to use nuclear materials or operate a nuclear facility. These regulations govern the transportation of materials; the use of materials at such nuclear facilities as power plants, research reactors, uranium mills, fuel facilities, and waste repositories; and the use of materials for medical, industrial, and academic purposes. The process of developing regulations is called rulemaking.

The NRC considers public involvement in the agency's activities to be a cornerstone of strong, fair regulation of the nuclear industry. For that reason, the NRC has a long-standing practice of conducting its regulatory responsibilities in an open manner, and keeping the public informed of the agency's regulatory, licensing, and oversight activities. Toward that end, our regulatory process provides a variety of opportunities, shown in Fig. 2, for citizens to be heard. For example, we announce public meetings on our Web site to enable interested members of the public to participate. The NRC also encourages public involvement in rulemaking, provide related information on our Rulemaking Dockets page, and provide opportunities for public involvement in hearings. See the Public Meetings and Involvements link on the NRC web page [15] and NUREG/BR-0215 [16] for general information about the available opportunities for public involvement.

10. RESOURCES AND STAKEHOLDER INVOLVEMENT

Although the revised regulatory framework for an advanced burner reactor is no longer included in the scope, the staff has reviewed its estimates for completing the regulatory framework for reprocessing and concluded that the activity will: (1) be more comprehensive than originally envisioned; (2) will involve resolution of several complex technical and policy-related issues; (3) will entail the development of new and substantive regulatory guidance; and (4) will require extensive stakeholder involvement.
As stated in industry correspondence, industry’s intent is to submit an application for a reprocessing facility in the 2013-2014 timeframe. To be prepared to review a potential application in that timeframe, the staff planned to complete the revised regulatory framework in FY 2012. The staff estimates that in order to complete the rulemaking activities in FY 2012, a total of approximately 15–20 FTE and $1.5–$2.0 million dollars will be needed in the FY 2010–2012 period. The staff recognizes that resolution of several policy and technical issues, independent of the resources available, may inform the final schedule for revising the reprocessing regulatory framework, such as the Secretary of Energy’s plan to create a commission to study alternatives to a nuclear waste repository at Yucca Mountain.

The process for revising the regulatory framework began in 2006. The initial pace was slow and will continue consistent with industry commitment and the NRC’s staff allocated resources. Fig. 3 depicts a timeline that shows several significant interactions and milestones related to the ongoing development of the revised regulatory framework.

The staff plans to continue to appropriately engage stakeholders during the development of the technical basis, achieving transparency and openness in the regulatory process. Completion of the technical basis will be contingent on the availability of resources, which the Commission will decide in the development of the Agency budget for fiscal year 2011.
FIG. 3. Schedule, key interactions, and milestones for updating the regulatory framework for spent fuel reprocessing.
REFERENCES


