DEVELOPMENT OF CHALLENGEABLE REPROCESSING AND FUEL FABRICATION TECHNOLOGIES FOR ADVANCED FAST REACTOR FUEL CYCLE

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ABSTRACT
R&D in the next five years in Feasibility Study Phase-2 are focused on selected key technologies for the advanced fuel cycle. These are the reference technology of simplified aqueous extraction and fuel pellet short process based on the oxide fuel and the innovative technology of oxide-electrowinning and metal-electrorefining process and their direct particle/metal fuel fabrication methods in a hot cell.
Automatic and remote handling system operation in both reprocessing and fuel manufacturing can handle MA and LLFP concurrently with Pu and U attaining the highest recovery and an accurate accountability of these materials.

Keywords: advanced fuel cycle, simplified aqueous extraction, pyro-electrochemical, sphere and vipack fuel fabrication, MA and LLFP transmutation

1. INTRODUCTION

Requisites to achieve the sustainable development for safe and public acceptant nuclear energy are being evaluated worldwide from the points of fissile material recycling, competitiveness, non-proliferation and environmental impact. The closed fuel cycle is the most promising in realizing these requisites.[1] However, fuel cycle technologies in the present closed fuel cycle, such as PUREX spent fuel reprocessing and MOX pellet fabrication, needs more flexibility and advancements to achieve the broader needs of the next generation fuel cycle.

The current Plutonium and Uranium recycling, LWR-MOX fuel, is intended to be one of the realistic solutions to break the imminent obstacles in the shortness of spent fuel storage and disposition of Pu stockpile. However, multi-recycling of Pu/U resources connected with the reprocessing of LWR-MOX spent fuel will be optimized after the introduction of "Advanced Fast Reactor (FR) Fuel Cycle system". By using FR in the advanced cycle, deteriorated Pu from LWR-MOX spent fuel can be burned again flexibly with a large amount of recovered U and also with high toxicity materials, Minor Actinides (MA) and Long Life Fission Product (LLFP), produced by the past, present and future nuclear fission energy.[2]

Key technologies for the Advanced Fuel Cycle system have been proposed and are being reviewed in Japan Nuclear Cycle Development Institute (JNC) as part of the extensive program of Feasibility Study (FS).[3][4] At the end of phase-1 in March JFY2001, several candidate technologies were screened and selected by preliminary assessment. In this paper, the direction of R&D for selected key technologies for the next five years in FS Phase-2 is introduced.

2. TECHNOLOGY CONCEPT OF ADVANCED FUEL CYCLE

Advanced Fuel Cycle concept is aimed at establishing the following social requisites in the 21st century as shown in Fig.1.

1) Safety processing features that ensure the lowest risk and reliable operation of fuel cycle system,
2) Economically competitive to the present fuel cycle,
3) More attractive system for the maximum use of U resources,
4) Minimum disposal of U resources, including Pu, MA and LLFP,
5) International acceptance by the enhanced nonproliferation process.

The basic concept of Advanced Fuel Cycle system should be constituted with simple and compact processes in order to satisfy the above requisites. When Pu, U, MA and LLFP are handled in the closed recycle process concurrently, an automatic and remote handling system operation becomes feasible while attaining high high recovery and accurate accountability of these materials.

The laboratory and bench-scale testing of such technologies will be conducted in Tokai Works of JNC by using several hundred grams of U, Pu and MA to verify and validate their feasibilities for adoption in the advanced cycle. At the end of Phase-2, these selected key technologies will be further screened and integrated into likely two most promising fuel cycle systems.
An advanced fuel cycle system chosen from the Phase-2 will then be demonstrated by testing fuel pins or assemblies in JOYO and MONJU reactors, and by processing the spent fuel pins in engineering-scale R&D facilities, such as Recycle Equipment Test Facility (RETF) in Tokai Works. These sequential approaches will address the optimization of the future fast reactor advanced fuel cycle system by the year 2015.

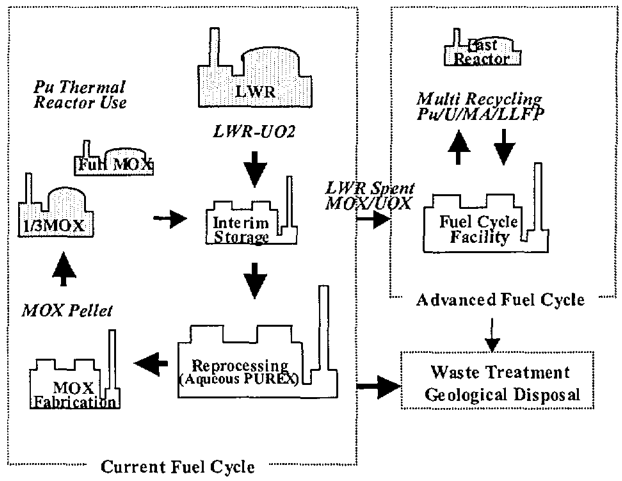

Fig.1 Advanced fuel cycle concept introduced to current fuel cycle

There are two main technologies to be developed for the advanced cycle system in Phase-2, aqueous process and dry pyro-electrochemical process shown in Fig.2.

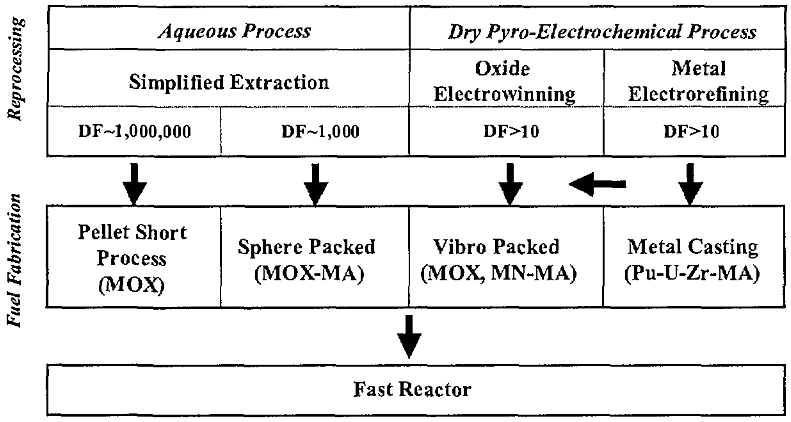

Fig.2 Selected key technologies for R&D

One is the reference technology, which is the simplified aqueous extraction and fuel pellet short process. These can be further modified and upgraded to the fast reactor specification from the present mature LWR fuel cycle technology for PUREX reprocessing and MOX pellet fabrication, which has been developed for the last 30 years and now commercialized in France, UK and Japan.

Reference technology can be harmonized easily with the current LWR fuel cycle system by handling both LWR spent UO2 and MOX fuels as well as fast reactor spent fuels. An alternation process may be sphere packed process. However, there need many technical challenges to improve economical competitiveness drastically and also to handle MA and LLFP, especially for fuel fabrication process.

The other is the more innovative technology such as oxide-electrowinning and metal-electrorefining process and their direct fuel fabrication within a hot cell. However, engineering concept in oxide and metal pyrochemical processes are a drastically different from the reference technologies; mainly, because the operating conditions of i) handling material from aqueous to molten salt, ii) temperature level from room-medium to medium-high and iii) decontamination level from high to medium-low. These are real challenges in the new world of advanced fuel cycle.
Alternative fuel types such as metal or nitride are also selected, because their in-core performances are superior to MOX under the several specified fast reactor type and coolants, such as sodium, lead-bismuth, gas, or water.

3. TECHNOLOGIES ACHIEVABLE TO ADVANCED CYCLE NEEDS

3.1 Advanced Reprocessing Technology

The basic flow sheet of key advanced reprocessing technology is shown in Fig.3 for both aqueous and dry pyro-electrochemical processes. Initial spent fuel is assumed to be the oxide type, which is currently used in LWR and FR.

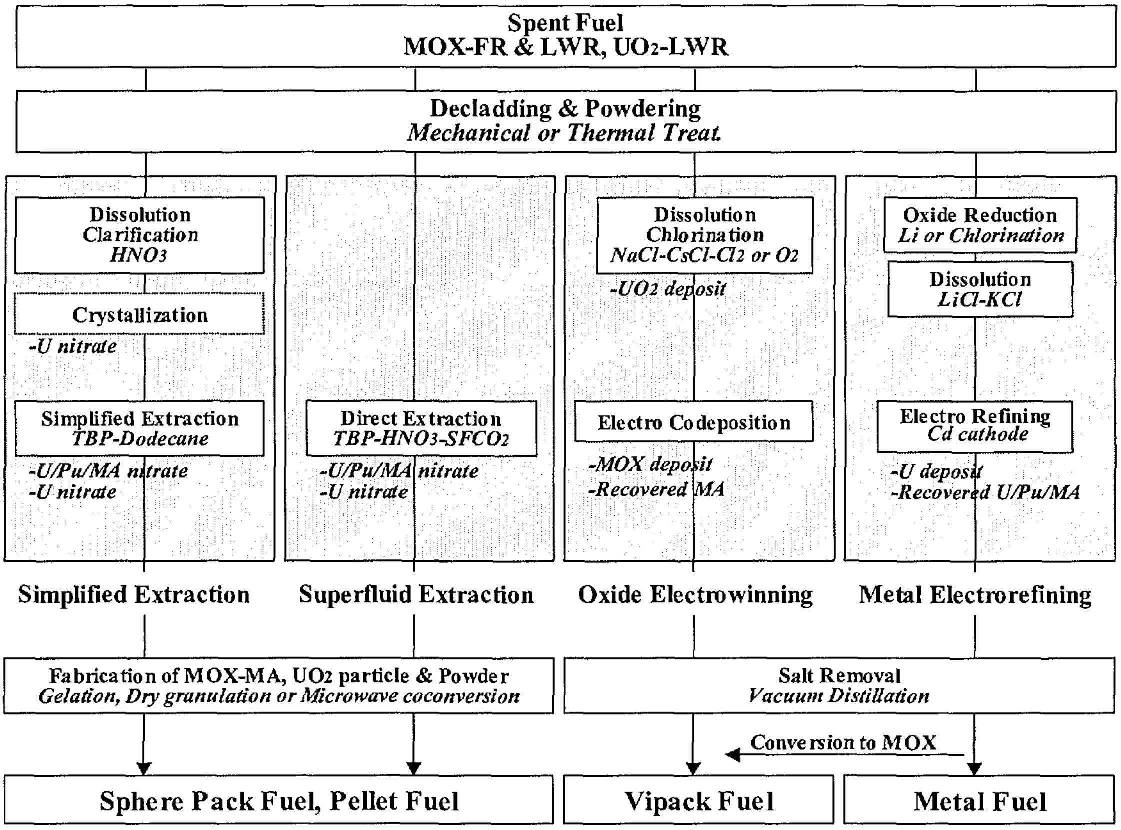

Fig.3 Advanced reprocessing flow sheet for advanced fuel fabrication

1) Simplified extraction process

Simple cycle extraction by aqueous reprocessing technology, which is a drastic modification of the current PUREX process, is the reference technology because of its realistic and extensive technical capability for the next step to industrial application. The goal is the optimization of Pu, U and MA extraction process to be as simple as possible and development of the associated equipment.

Simple flow sheet with co-recovery of U, Pu and Np was experimentally established by JNC in the MOX PUREX reprocessing. Np valence adjustment produces successful extraction of Np with U and Pu during the extraction section. All Pu, Np and a certain part of U are recovered in the mixed product stream. Specification of the products was preliminarily optimized with the flow sheet study by using spent MOX fuel. The decontamination factor (DF) of products in these simplified systems can be adjustable in the range of 1,000 to 1,000,000 considering the impact to fuel fabrication process. Achievement of higher DF product in the simple extraction is more challengeable work, which leads to the modification of current PUREX process. It can be achieved by the combination of process optimization and equipment modification. By adopting the one-cycle co-recovery process, neither separated Pu nor radiation-free nuclear material exists in any step of the entire fuel cycle; thus, the resistance against misuse and theft of Pu is greatly enhanced.

A pretreatment process of crystallization for the extraction of excess U from a dissolved MOX solution makes it possible to achieve a more compact system for U, Pu and Np extraction in the simplified PUREX. The U/Pu ratio in the mother solution after the crystallization is adequate for MOX fabrication, U and Pu partition section is deleted by co-recovery of all U and Pu.

These technologies are ready for the next demonstrating stage for MOX spent fuel reprocessing in the Chemical Processing Facility (CPF) of Tokai Works. (Figs 4 & 5)

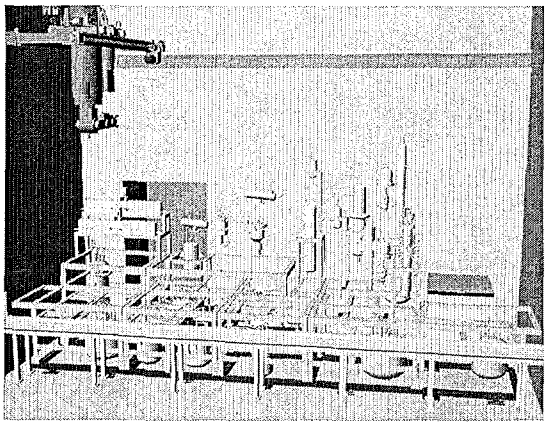

Fig.4 Plan of hot cell modification in CPF

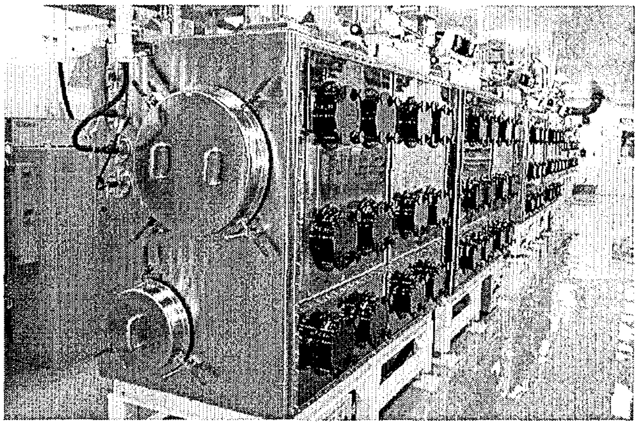

Fig.5 New GB for R&D in analysis room CPF

On the other hand, simplification and size reduction of major equipment are key issues to establish economic competitiveness of reprocessing plant. JNC has already developed new compact equipment such as a disassembly system with CO_2 laser beam, rotary kiln type continuous dissolver, centrifugal clarifier and centrifugal contactor. These were ready to be installed at the RETF, of which construction has been interrupted by the retardation of fast reactor fuel cycle demonstration.

Presently, we still envision further potential for the disassembly system to provide size reduction and to enhance the performance by applying YAG laser beam and integrating with shear. Also the centrifugal contactor has further potential to show more reliability, longer lifetime and higher throughput by modifying internal structure. These advanced equipments are still under intensive development.

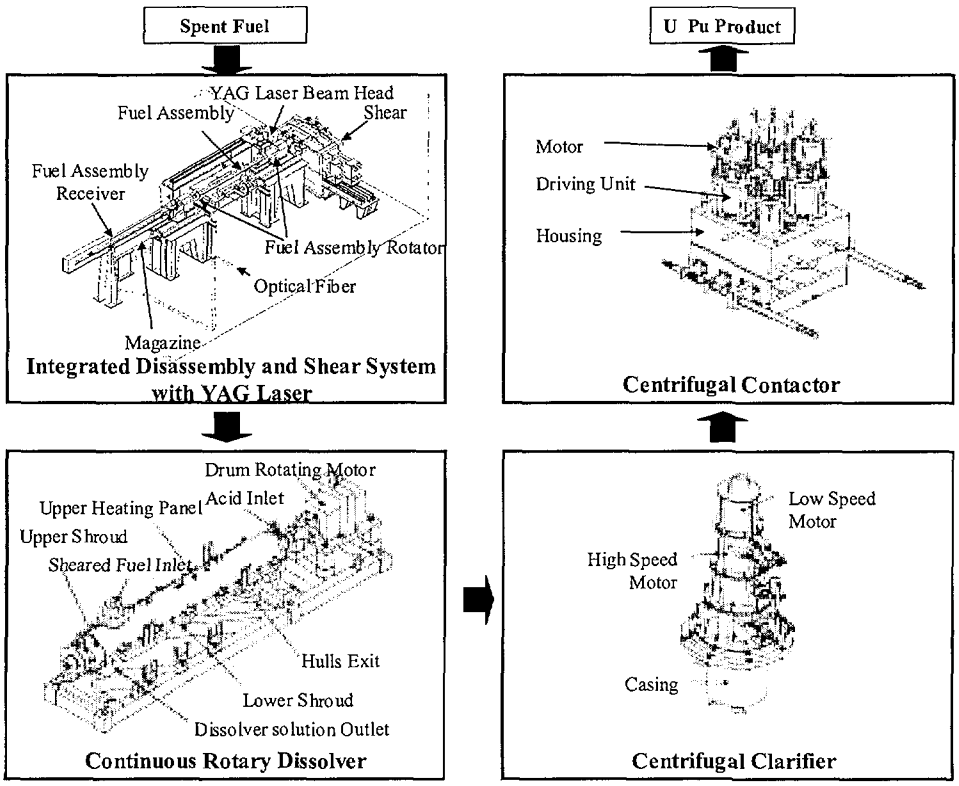

Fig.6 Major equipments under development for aqueous simplified extraction process.

2) Direct recovery of U and Pu with TBP–supercritical CO_2 mixture
Supercritical carbon dioxide (SF CO_2) is attractive for its high diffusion and easy after-treatment and has been studied for decontamination of nuclear materials. Although the fluid is nonpolar, some additives are useful to solvate polar compounds. The mixture of TBP and SF CO_2 can extract uranium nitrate.[5] Moreover, the mixture of SF CO_2 and TBP-HNO_3 solvate will react to convert U or Pu-U oxide to nitrate and extract them directly. This new extraction technique enables a drastic simplification of reprocessing process based on the PUREX process, namely merging dissolution and co-decontamination steps, and also decrease the amount of waste solutions, such as used solvent from the cut of diluent and high level liquid waste from elimination of the dissolution step.

This process eliminates the clarification, adjustment and high level liquid waste treatment steps, and reduces the size of the main extractor. The powdering step is combined with the process to obtain U_3O_8 powder, and facilitates rapid extraction with less heat generation. The TBP–HNO_3 solvent is previously prepared by contacting TBP with a concentrated HNO_3 solution. The main procedure requires high pressure, $\geq$ 12 MPa. However, the operating temperature can be lowered to 40-50 °C.

The feasibility of this process has been investigated. Direct extraction of U from oxide (U_3O_8) was confirmed by preliminary experiment. And distribution behavior of major fission products was examined. Hot parametric experiments are needed to evaluate the usefulness of this process by using MOX or its irradiated fuel. The safety issue from utilizing highly pressurized CO_2 fluid has to be cleared.

3) Dry pyro-electrochemical process
Another candidate for advanced reprocessing is the modified pyro-electrochemical processes and their equipments based on Russian-RIAR and US-ANL methods. The reprocessing technology for oxide fuel is rather focused at the transition period from the current fuel cycle to the next advanced fuel cycle. The development of these key technologies focuses on the safe, reliable, industrial scale-up of electrowinning and refining systems including an extraction process for MA and LLFP.

Continuing efforts by RIAR demonstrated a successful operation of the oxide electrowinning by using several kg of spent fuel from BOR-60. Japanese electric utilities and JNC are now trying to modify these processes.
The R&D on metal electrorefining has been principally designated to Central Research Institute of Electric Power Industry (CRIEPI) in Japan. The Fuel Conditioning Facility in ANL has demonstrated its high potentiality to treat spent fuels. Metal electrorefining will be more effective for fast reactor metallic fuel cycle. When metal electrorefining process is applied to the fast reactor oxide base fuel cycle, additional processes for the initial reduction of MOX and the final oxidation of metal are required.

Fundamental studies are still required to adjust the electrowinning condition. Development of long-life component materials, including crucible material, is an issue for realization of the dry process because of the corrosive and high temperature operating conditions. Safeguardability of the dry process should be assured with the real time monitoring equipment and inspection system. The loss of fissile materials to waste should be minimized and the recovery of MA can also be further optimized. The treatment of chloric type wastes has to be guaranteed for long-term stable storage. Concept optimization for industrial-scale spent fuel pyroprocessing is important to reveal weakness of existing pyroprocesses and clear the direction of the improvement.[6]

JNC is arranging testing infrastructures in Tokai works for metal electrorefining as well as oxide electrowinning. Collaboration programs with domestic and overseas partners are in progress. Fig.7 shows the glove box equipment, which is being prepared by CRIEPI at CPF in JNC-Tokai Works to make integral Pu and U experiments related to metal electrorefining.

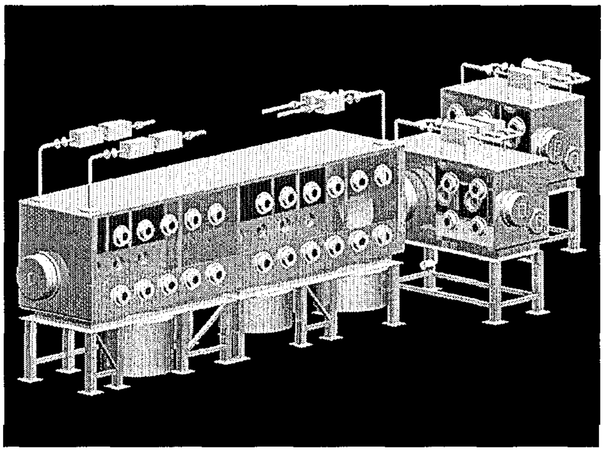

Fig.7 Metal electrorefining GB by CRIEPI in CPF

Next five years in FS pahse-2 becomes a key period to judge whether these dry processes compete with the aqueous reprocessing technology. Also the connection with to the fuel fabrication and reactor irradiation of reprocessed materials is ready to realize the advanced fuel cycle system.

3.2 ADVANCED FUEL FABRICATION

Fuel fabrication for the advanced cycle must be as simple as possible and suitable for the massive remote operation to handle radioactive materials, which are recovered from the reprocessing with low decontamination factors. Three candidates for fuel fabrication process are being investigated in FS; a simplified pellet process, vibro-packed process using particulate fuel and metal casting process. R&D for metal casing process is in progress by CRIEPI to modify the casting fuel fabrication method.

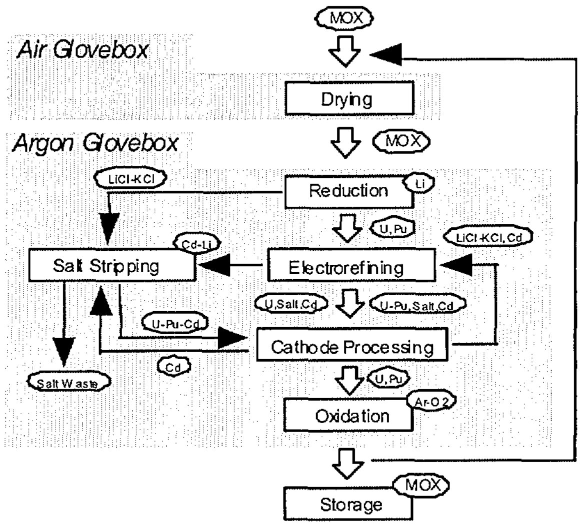

Fig.8 Experimental flow sheet for metal electrorefining

1) Simplified pellet process

The present MOX pellet fabrication process in a globe box becomes a mature technology applicable to LWR-MOX commercialization. However, this process must be modified to fabricate low DF fuels in a remote operation mode. Simplified pellet process is the shortest route in which MOX powder adjusted Pu content is co-converted directly by microwave heating process from Pu, U and Np nitric acid solutions for the next pelletizing process with minimum pre-treatment of powder. The key in this technology is to prepare the well-homogenized and controlled powder to obtain a high throughput.

When the simplified pellet process is applied to low DF products, the pellet design specification should be relaxed to realize a hot cell operation and also to match an increase of impurity level. JNC is conducting basic parameter tests to optimize powder characteristics and fabricate MOX pellet with the preliminarily simplified process.

2) Sphere and Vibro-Packed Fuel Fabrication

Sphere and Vibro-packed fuel fabrication method is a most important technology common to the particulate fuel products from both aqueous and dry reprocessing in advanced fuel cycle. The concept of vibro-packed fuel itself was introduced about 40 years ago. The key issue is the selection and optimization of particulate MOX-MA fuel fabrication methods. These are gel precipitation[7], dry granulation process in simplified PUREX and MOX co-precipitation in oxide electrowinning.

The controlled fabrication method of two or three size distribution of granular particles is important to achieve an equivalent smear density with MOX pellet pin of about 80%TD. The challengeable technology is to get the smaller size particle of higher Pu content MOX-MA fuel controlled less than 100μm in diameter by remote operation. Lessons from the past experiences in BNFL and RIAR for the vibro-packed fuel suggest that fuel pin quality assurance should be optimized for the continuous operation of manufacturing and inspection process. Laser scanning system to guarantee the quality of particle fuel and three-dimensional CT scan system to check the smear density distribution in a fuel pin are still under basic investigation.[8] The prevention of fuel-cladding chemical interaction (FCCI) is another issue. Controlling oxygen potential under an irradiation condition is necessary to achieve a high burnup capability. Both the initial conditioning of oxygen-to-metal ratio of MOX particulate and the mixing of oxygen getter with fuel particle are candidates.

4. OPTIMIZATION OF MA RECYCLE

Advanced closed cycle system coupled with the prospective use of fast reactors can realize both maximum use of U resources and minimum generation of waste via multi-recycling of U, Pu and MA. However, burning of MA and LLFP is still a basic stage to recover and handle in both reprocessing and fuel manufacturing.[9][10] In JNC, Np-MOX and Am-MOX pellet were fabricated and now examine physical properties to get an irradiation licensing in JOYO. Np and Am transmutation, however, should be optimized in the irradiation of advanced long-life fuel admixing with low DF Pu and U products in a homogeneous recycle concept.

When recovery of MA is considered in the reprocessing, it is necessary to compromise the equivalent recovery rate between Pu and MA. A level of 99.9% for recovery, which means 0.1% reprocessing loss, is recommended to decrease a long-term radio-toxicity drastically.[11] This is an important index to design the advanced reprocessing system.

5. CONCLUSION

Advanced fuel cycle option is expected to assure a virtually inexhaustible energy supply for our next generations without entailing economically and environmentally unbearable burdens nor the risk of proliferation.

Medium and long term R&Ds are needed with much resources to develop technologies for advanced fuel cycle. Both advanced reprocessing of dry pyro-electrochemical type and advanced fuel fabrication of particle fuel are challengeable key technologies to realize it. It is necessary to accumulate database for the selection and optimization of the next industrial level of such technologies.

Under the worldwide retardation of nuclear power R&D, international collaborations are necessary to develop advanced fuel cycle technologies. The globalization effect of safety, environmental restriction tends to establish an international standardization even in the technologies for the next generation fuel cycle system.

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