

DEVELOPMENT OF PYROPROCESSING AND ITS FUTURE DIRECTION

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Pyroprocessing is the optimal means of treating spent metal fuels from metal fast fuel reactors and is proposed as a potential option for GNEP in order to meet the requirements of the next generation fuel cycle. Currently, efforts for research and development are being made not only in the U.S., but also in Asian countries. Electrorefining, cathode processing by distillation, injection casting for fuel fabrication, and waste treatment must be verified by the use of genuine materials, and the engineering scale model of each device must be developed for commercial deployment. Pyroprocessing can be effectively extended to treat oxide fuels by applying an electrochemical reduction, for which various kinds of oxides are examined. A typical morphology change was observed following the electrochemical reduction, while the product composition was estimated through the process flow diagram. The products include much stronger radiation emitter than pure typical LWR Pu or weapon-grade Pu. Nevertheless, institutional measures are unavoidable to ensure proliferation-proof plant operations. The safeguard concept of a pyroprocessing plant was compared with that of a PUREX plant. The pyroprocessing is better adapted for a collocation system positioned with some reactors and a single processing facility rather than for a centralized reprocessing unit with a large scale throughput.

KEYWORDS : Pyroprocessing, Electrorefining, Electrochemical Reduction, Metal Fuel, Oxide Fuel, Proliferation Resistance, Safeguard

1. INTRODUCTION

Nuclear energy is the most promising option to mitigate the range of threats we face in the 21st century, including vast energy demand and growing CO₂ emissions. To ensure the stable use of nuclear energy throughout the century, the nuclear fuel cycle must be centrally positioned to ensure the effective use of uranium resources. With this in mind, ambitious countries have been developing a fast reactor fuel cycle in recent decades. However, the technological complexity and economical issues involved have hampered the commercialization of such fast reactors, meaning FFTF and EBR-II in the U.S. and Super Phenix in France were shut down and the sodium leakage of the MONJU complex adversely affected development. Following this stagnation in the 1990s, however, the revitalization of nuclear energy, the so-called the Nuclear Renaissance, has realized a policy change in the United States, where the AFCI, Generation-IV, and Global Nuclear Energy Partnership, GNEP[1] have been initiated in rapid succession. Research efforts have focused on the creation of a nuclear fuel cycle that recycles actinides to ease the burden of waste disposal and the effective use of disposal sites in those

programs. In Japan, a feasibility study for exploring a compact fast reactor cycle system was carried out from 1999 to 2005, concluding that Na-cooled FBR and an advanced aqueous reprocessing system would be the main system for commercialization from 2050, and a Na-cooled FBR with pyro-reprocessing would function as the sub-system for long term developmental efforts due to the flexibility of the metal fuel fast reactor system and its capability to satisfy a wide range of variable breeding ratios for nuclear utilization in the foreseeable future [2].

Pyroprocessing should be the optimal means of treating spent metal fuels from metal fuel fast reactors. A high breeding ratio can be achieved with metal fuel, while a breeding ratio exceeding 1.0 is also achievable using a metal fuel core without a radial blanket, from which pure Pu could be extracted. The pyrometallurgical process, itself, has potential in the sense that no additional process is required to separate TRUs and electrorefining can recover actinides, U,Pu, and minor actinides collectively, leading to strong resistance for nuclear proliferation. The Integral Fast Reactor, IFR, program started in the early 1990s has stimulated to investigate the fast reactor system with pyrometallurgical process [3]. Following the

suspension of the program in 1985 due to a U.S. policy change involving the non-use of Pu and a halt to further exploration of fuel cycle technology, the Central Research Institute of Electric Power Industry, CRIEPI, involved in the IFR program until 1985, has continued the research and development with the cooperation of the Institute of Transuranium Elements, Karlsruhe, the University of Missouri, and domestic institutes, and transmitted the activity to the nuclear world [4,5]. At the time of writing, the Korean Atomic Energy Research Institute, KAERI, who noticed the future potential of this option for spent fuel treatment, steered the use of pyrometallurgical technology [6,7]. After the return of the Nuclear Renaissance in the U.S., the pyrometallurgical process has been included in GNEP, and the process of recovering actinides by electrorefining has commenced. Currently, India and China, who expect a rapid increase in nuclear energy of 240-250GWe by around 2050, have initiated the research and development of metal fuel fast reactors with a pyrometallurgical fuel cycle. This paper reviews the latest pyrometallurgical processes including the development of the concept of material accountancy for safeguarding purposes and considers future directions of research and development.

2. ACTIVITIES OF ADVANCED FUEL CYCLE DEVELOPMENT

Advanced fuel cycles are explored not only in the U.S. and France, but also in Asian countries such as Japan, Korea, India, and China. After announcing GNEP, the U.S. has moved from a once-through fuel cycle to an improved approach based on the recycling of TRUs in spent nuclear fuels. Recycling would employ advanced fuel separation technologies, for which the aqueous process represented by the UREX+ series [8] to separate actinides and heat-generated fission products, Cs and Sr, and pyroprocessing have been investigated [9]. The aqueous process is mainly targeted for application on spent LWR fuels in order to reduce the waste volume to be disposed of in the Yucca Mountain Repository by separating uranium and heat generated species [10]. The TRUs recovered are subsequently used at the advanced recycling reactor, ARR. The pyroprocessing for the separation of uranium and TRUs has been explored at the Idaho National Laboratory to recycle fuels from ARR, which will be operated with oxide and/or subsequently with metal fuel. This technology is currently being demonstrated with the treatment of EBR-II spent fuels [11]. Through 2007, 3.4 tHM of spent fuel was treated to separate uranium by electrorefining [12].

The French government established the Radioactive Waste Management Research Law in 1991 in order to investigate three options for rational waste disposal, comprising (1) the research into partitioning and transmutation, (2) the

study of the possibilities of reversible or irreversible disposal within deep geological formations, particularly through the construction of underground laboratories, and (3) the study of the conditioning and long-term surface storage processes for high-level and long-lived waste. After considering achievements of the last 15 years [13], a new waste management bill, detailing further research and development of partitioning and transmutation within the framework of the generation-IV system, was adopted by parliament in June 2006. President Chirac announced the construction of a Generation-IV prototype reactor by 2020. The French version of an advanced fuel cycle is a Na-cooled fast reactor with oxide fuel as a reference, but metallic fuel as a potential alternative. An electrorefining process involving an aluminum cathode in fluoride salt is under investigation [14] together with the cooperation of the 6th and 7th framework EU programs [15].

Asian countries are represented by India and China, both of which foresee a vast increase in nuclear energy demand within the first half of this century. A prototype fast reactor of 500MWe with MOX driver fuel and thorium blanket subassembly is under energetically construction in India and is expected to be commercially implemented in 2011. Following this, in 2012, construction will commence on twin plants of equivalent capacity. The spent oxide fuels will be treated by Purex-based reprocessing and construction of the facility will commence shortly. Subsequently, the operation of a further two plants of FBR with 500MWe with metallic fuel is planned. Together with this program, research and development into pyrometallurgical technology has started with the fundamental study of electrorefining, casting for fuel fabrication, and the engineering installation of an electrorefiner in a uranium glove box.

South Korea has been ambitiously involved in research and development of the pyrometallurgical process, as described earlier. Uranium is the first candidate for removal in order to reduce the waste volume by electrorefining. Engineering scale models of a voloxidizer and an electrochemical reduction cell have been developed by the use of 20kg of uranium. The facility to examine the Advanced Spent Fuel Conditioning Process, ACP should play an effective role for process verification by the use of spent fuels [7]. Furthermore, South Korea has been striving to develop a generation-IV reactor through international cooperation.

3. DEVELOPMENT OF THE METAL FUEL CYCLE

The process flow of metal fuel cycle is shown in Figure 1. After decladding and chopping of spent metallic fuel, electrorefining in chloride salt is the main method to recover uranium on a solid cathode and TRUs with uranium into a liquid cadmium cathode by transferring them from the loaded spent fuel in the anode basket. The products of uranium metal and actinide mixed alloy are

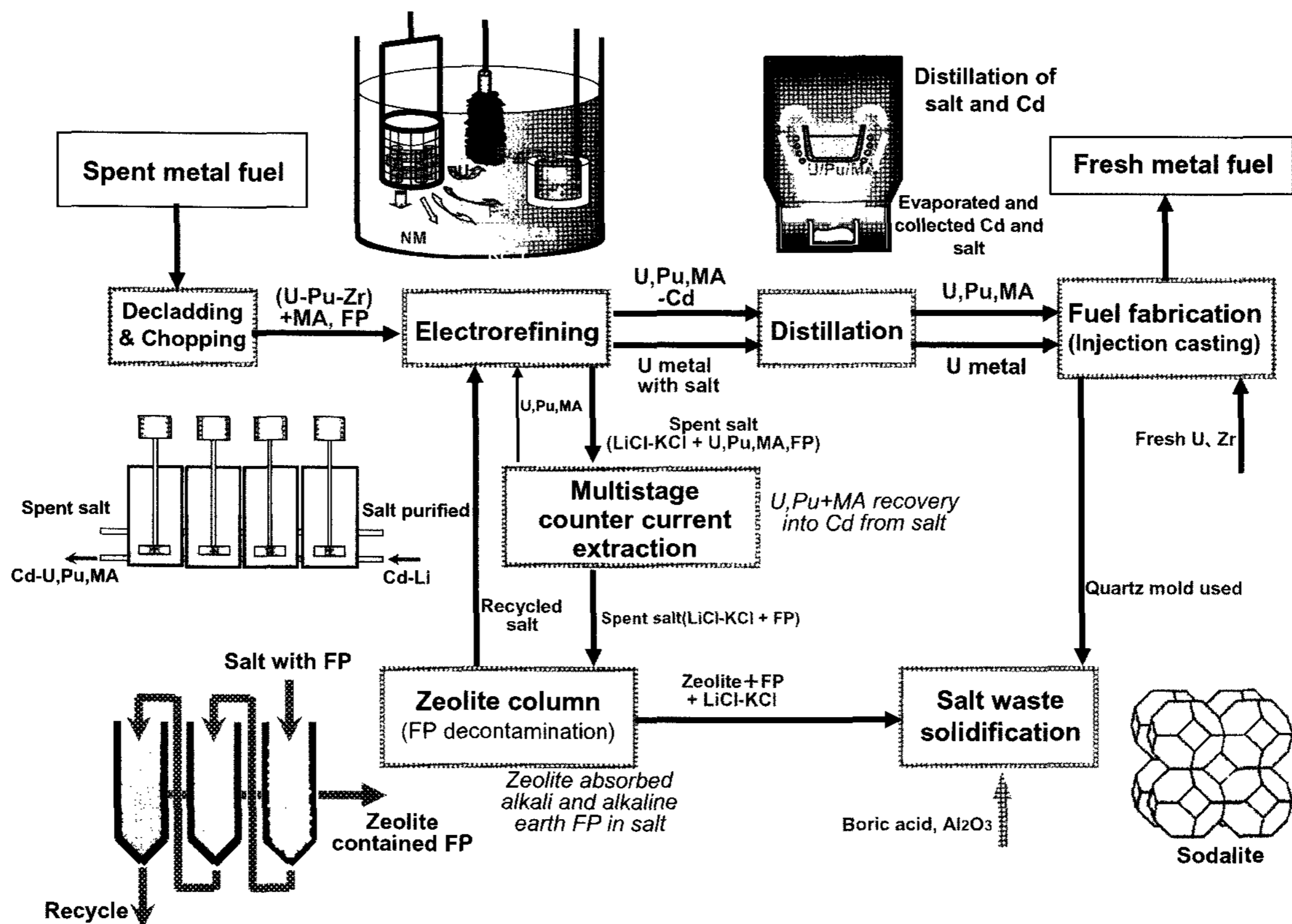


Fig. 1. Process Flow of Pyroprocessing of Metal Fuel Cycle

obtained by distilling salt or cadmium in cathode products, and, then, injection casting is employed to fabricate new fuels. U-Pu-Zr with minor actinides has been electrorefined for process verification, as shown in Figure 2 [16]. Figure 2 reveals the inside of the caisson, where the electrorefining apparatus is set up in a hot cell. Post-electrorefined metal fuel in an anode basket and cathode products in solid and cadmium cathodes are also given. Experimental data verifies that the americium is proportionally recovered with plutonium and that more than 10wt% of Pu can be collected into a liquid cadmium cathode via the formation of intermetallic $Pu_{1-x}Am_xCd_6$ compounds with high current efficiency [17,18]. The engineering model of an electrorefiner for practical application has been developed with the use of uranium [19]. Figure 3 shows the schematic presentation of the electrorefiner by which several kilograms of uranium were recovered at the solid cathode through transport from anode baskets and scraped in a bottom dish of the electrorefiner. Besides the main processes, waste treatment is another essential process to minimize the salt waste to be disposed of. The salt contained small amount of actinides and majority of fission products after use in the electrorefining process should be treated to remove the actinides. The counter current extraction is applied in a $MCl_x-LiCl-KCl/Cd$ or $/Bi$ system. Thermodynamic

measurements indicate a higher separation factor between actinides and rare earths within a $MCl_x-LiCl-KCl/Bi$ system [20]. Subsequently, the salt is scrubbed to remove fission products through a zeolite column. The eutectic salt regenerated is recycled into the main process, and the zeolite with fission products is sintered to create a sodalite, which is stable and durable upon disposal. For development of the transport technology of molten salt and liquid cadmium at 773 K, transport devices were installed in an Ar atmosphere glove box. The transportation of molten salt to a salt treatment system and of liquid cadmium to a distillation furnace from an engineering- scaled electrorefiner are demonstrated by the use of centrifugal pumps [21].

4. APPLICATION OF THE PYROMETALLURGICAL PROCESS TO OXIDE FUEL

Pyroprocessing can be applied to spent oxide fuel, especially high actinide fuel or short cooling fuel. The electrochemical reduction makes oxides into metals within a $LiCl$ melt with the addition of Li_2O . Oxides loaded into the cathode basket are reduced to produce oxygen gas or CO_2 at the anode with platinum metal or pyro-graphite. After reduction, the anode products are

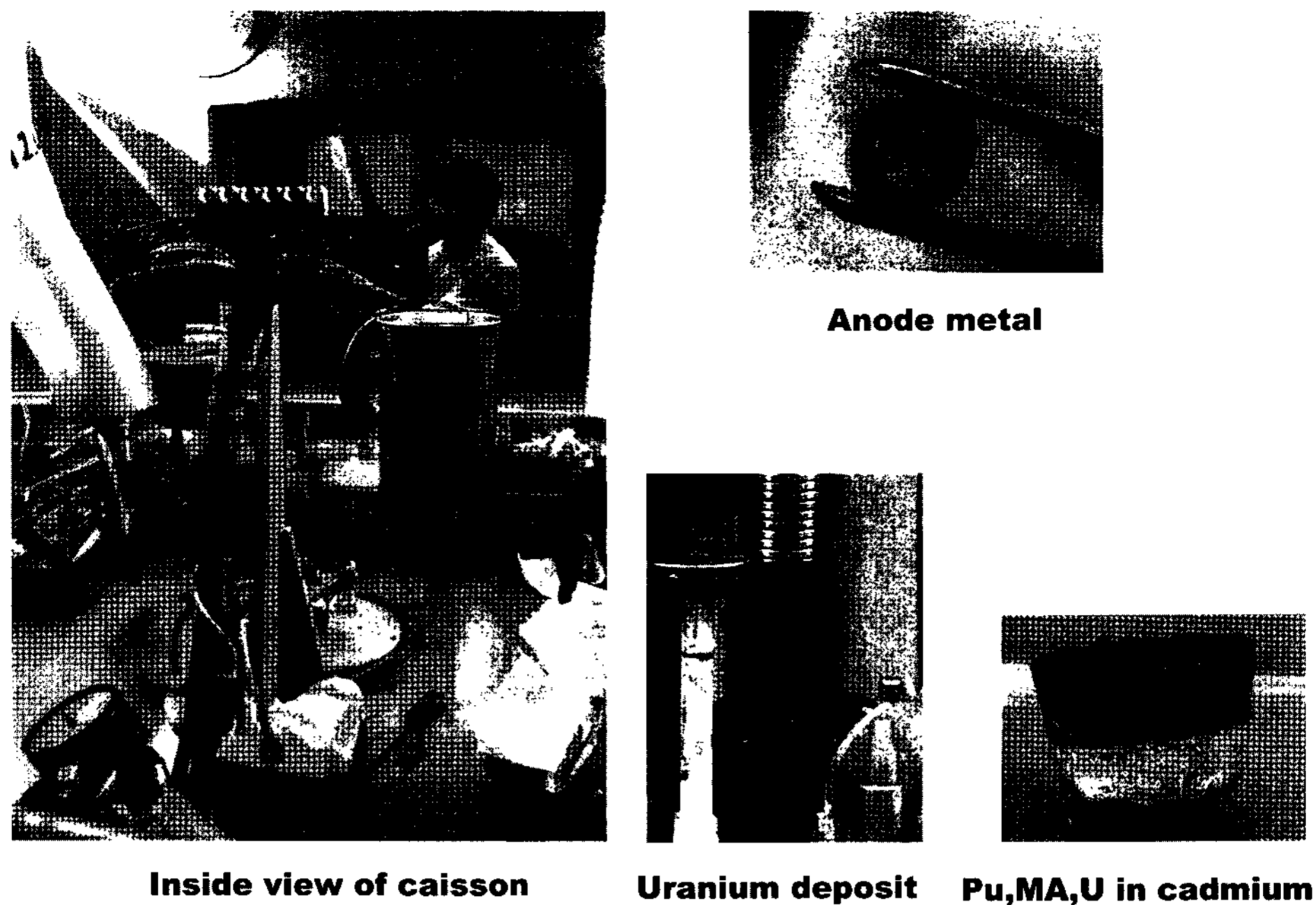


Fig. 2. Set up of Caisson in Hot Cell for Process Verification

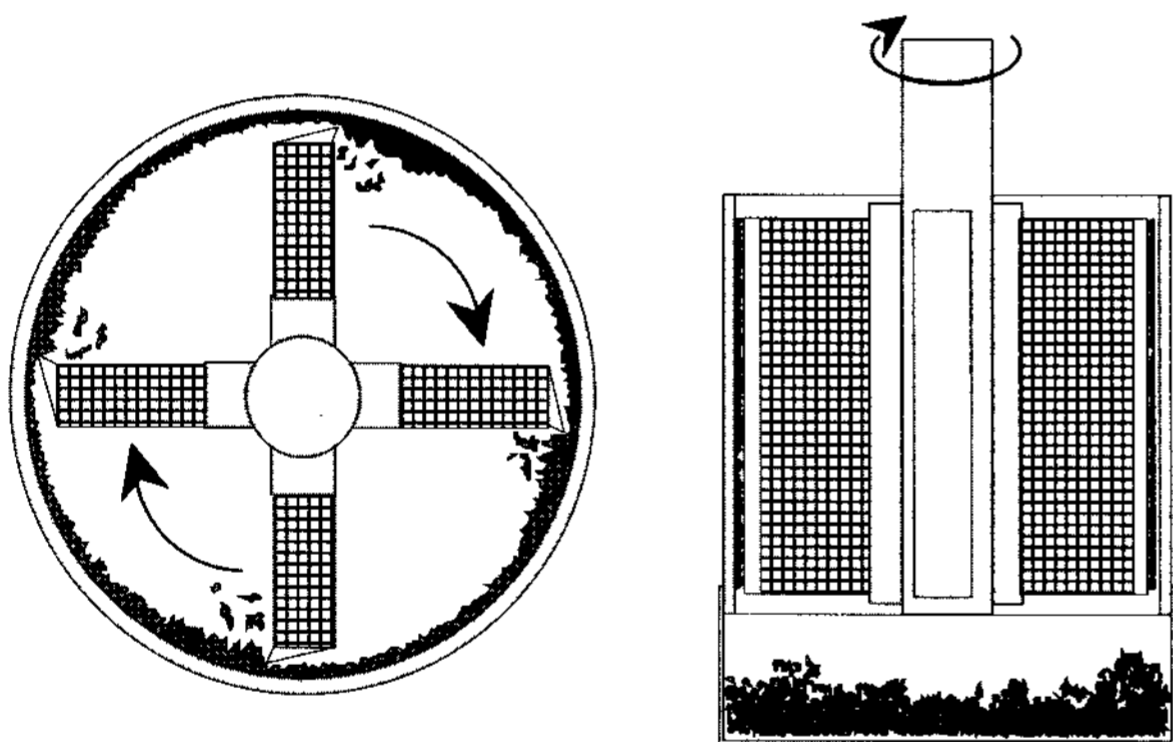


Fig. 3. Development of Engineering Model of Electrorefiner

conveyed to the electrorefiner to separate actinides as mentioned in the previous section. Experimental studies verify the complete reduction of actinides electrochemically on UO_2 , $(U,Pu)O_2$, and UO_2 doped stable fission products and irradiated $(U,MA)O_x$ [22-24]. Figure 4 shows a schematic presentation of an electrochemical reduction cell, and the change of the morphology of $MOX((U-40Pu-5Np)O_2)$ electrochemically reduced in a $LiCl$ bath at 923K. The dense surface changed to a porous structure with agglomerated particles. The experiment suggests that the reduction proceeds more rapidly in Pu containing material and porous media than in UO_2 and highly dense materials. The significant influence of the feed oxide on

effective reduction is, therefore, amply demonstrated. The cathode basket also plays an important role in ensuring effective reduction and maintaining stable thermal conductivity. Commercially irradiated MOX fuel will be supplied in the next experimental trial. Electrowinning of the UO_2 prior to the electrochemical reduction reduces the volume to be treated in the reduction process and enables the adjustment of the product composition for fuel fabrication. A eutectic salt of $LiCl-KCl$ is used for the UO_2 collection on the solid pyrographite cathode. The estimated material balance through electrowinning to electrorefining is given in Figure 5, as well as the product composition in Table 1 for spent UO_2 and MOX fuels with burnups of 45GWd/t and 60GWd/t, respectively. The uranium content in the actinide mixture in a liquid cadmium cathode to be supplied as part of the main resources for fuel fabrication can be varied by changing the recovery amount of uranium during electrowinning and electrorefining on the solid cathode.

5. EVALUATION OF THE PYROMETALLURGICAL TREATMENT PRODUCT FROM THE PERSPECTIVE OF PROLIFERATION RESISTANCE AND THE SAFEGUARD CONCEPT

The pyrometallurgical process produces Pu together with other actinides, U, Np, Am, and trace amount of fission products, when blanket fuels are treated with driver fuels. The product can be handled only when biologically shielded. Table 2 shows the radiation increase of 1g of a one-year-

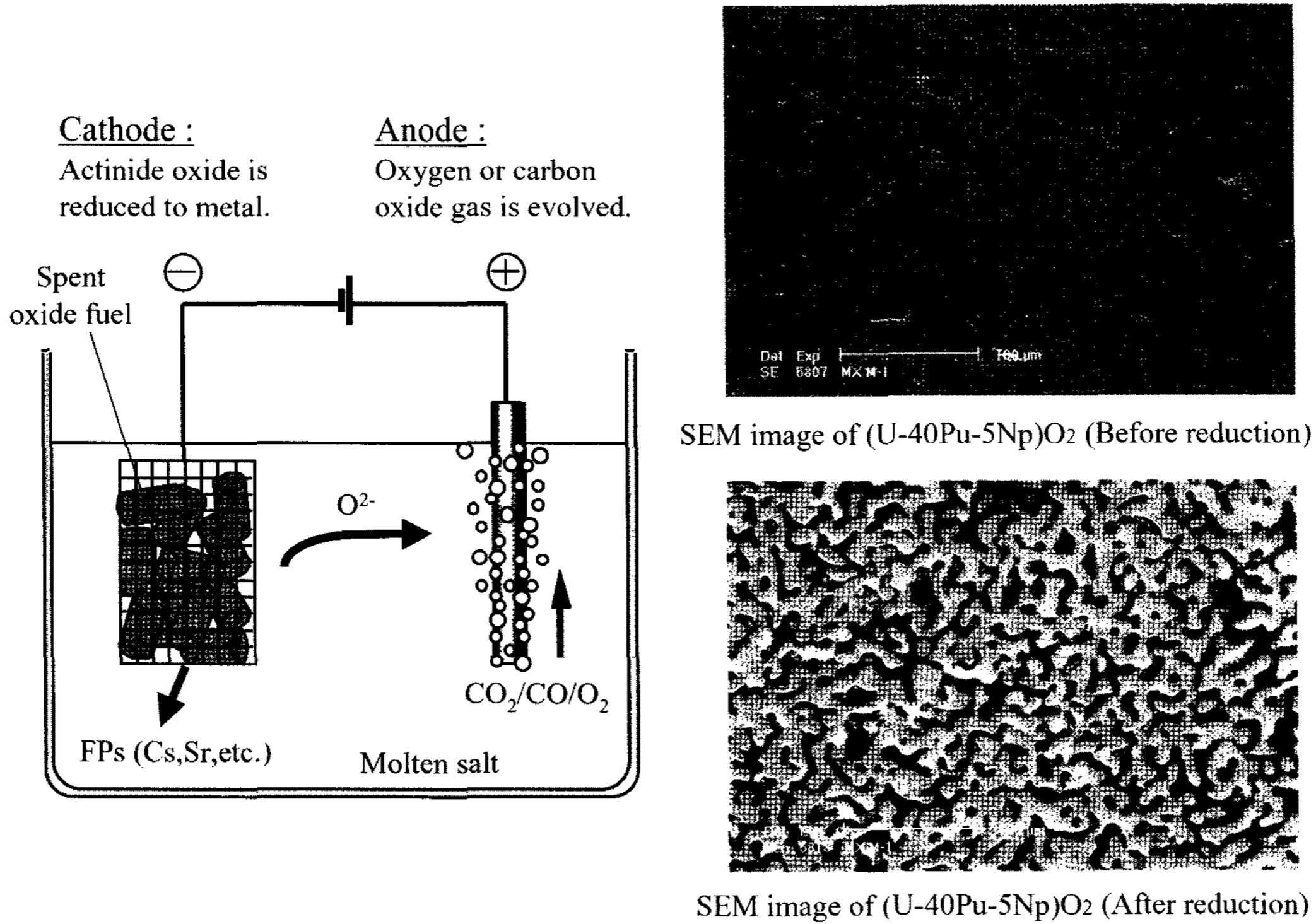


Fig. 4. Schematic Presentation of Electrochemical Reduction and SEM Image of the Electrochemically Reduced (U-Pu-Np)O₂

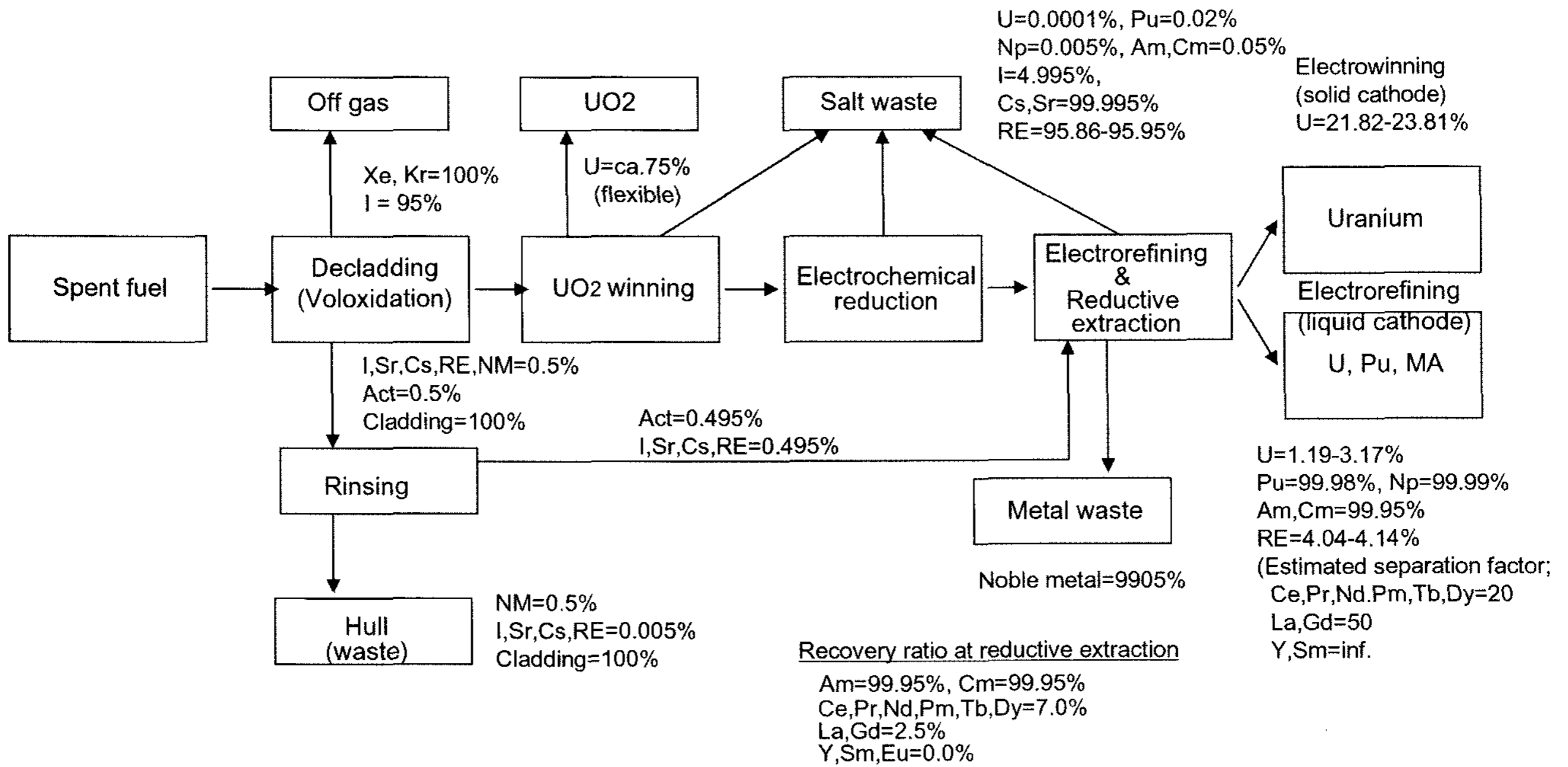


Fig. 5. Estimated Material Flow for the Treatment of Oxide Fuel

Table 1. Expected Composition of Product by Electrowinning for Metal Fuel Fast Reactor

Spent fuel	Uranium (wt%)	Plutonium (wt%)	Minor actinide (wt%)	Rare earth (wt%)
UO ₂ , 45GWd/t	46.5	46.5	4.6	2.3
UO ₂ , 60GWd/t	45.7	45.7	5.9	2.7
MOX, 45GWd/t	47.0	47.0	5.2	0.8
MOX, 60GWd/t	46.4	46.4	6.0	1.2

old TU mixture with a fission product compared to 1g of pure typical LWR Pu or weapon-grade Pu [25]. The radiation doses will deter any diverter and make it impossible to convert the fuel directly into a nuclear explosive without sophisticated purification. If the spent fuel is processed in due time, we can expect a decay heat of several 100W, enough to melt the explosive detonator contained in a nuclear device. Nevertheless, institutional measures must be taken to ensure proliferation-proof plant operations. The blanket fuel of FBRs contains weapon-grade Pu therefore, the chopped blanket fuel must be mixed with driver fuel and electrorefined in a common electrorefiner. As long as the overall extent is unknown, repeated electrorefining will lead to a cleaner product usable as a nuclear explosive, meaning any recycling in the plant must be carefully monitored.

Table 2. Comparison of Radiation Emitted by the Pyrochemically Produced TRU, from LWR Pu and Weapon-Grade Pu

Radiation type	Per g of TRU	Per g of LWR Pu	Per g of weapon-grade Pu
Alpha-activity (Ci)	1.53	0.85	0.09
n-activity (n/sec)	8.8E+04	1.4E+03	1.3E+02
photons/sec(Gamma radiation)	1.1E+10	5.0E+09	2.9E+08
MeV/sec	2.3E+08	5.1E+07	3.1E+06
Decay heat(W)	0.052	0.027	0.003

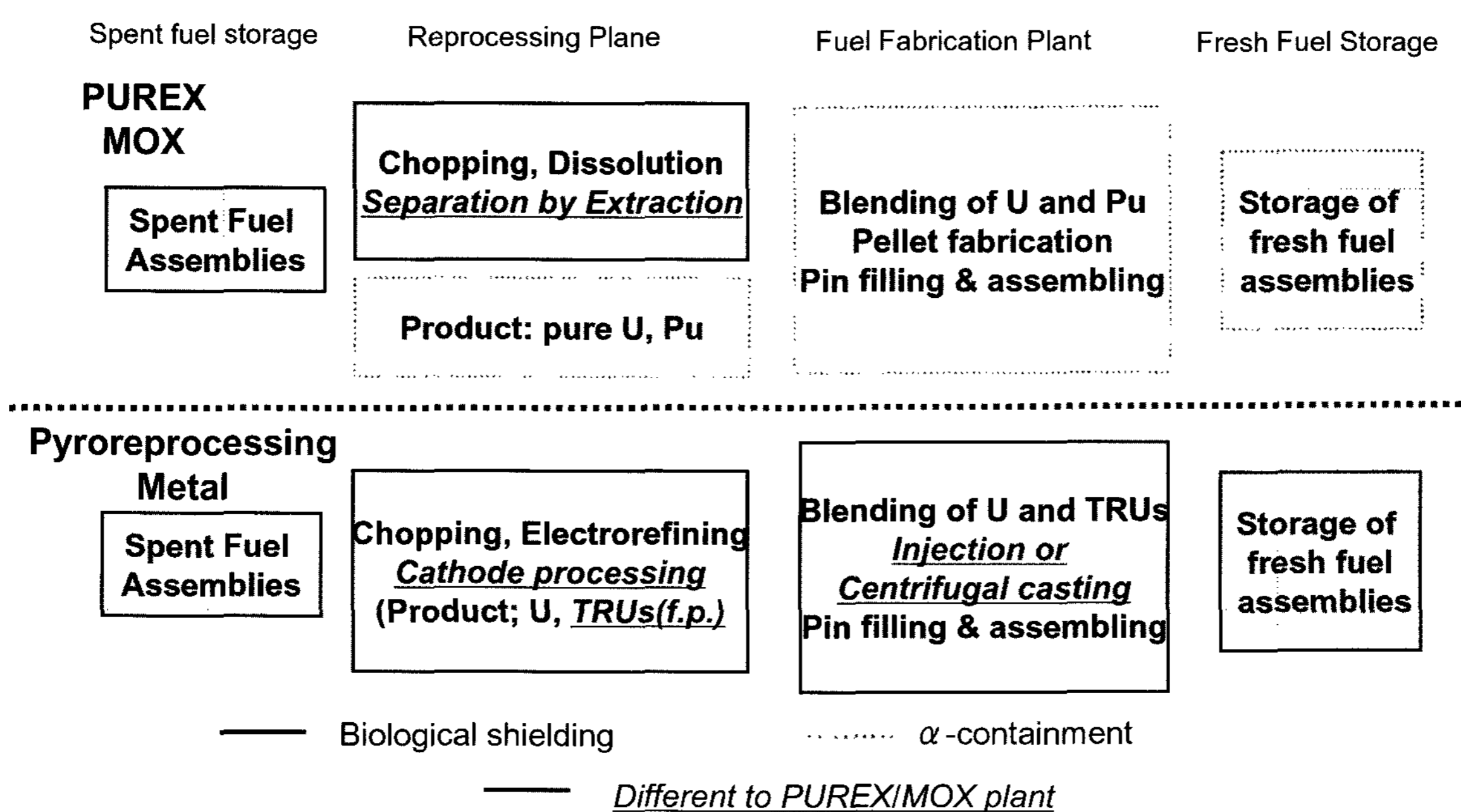


Fig. 6. Comparison of Safeguard Concept Between Aqueous and Pyroprocessing

Figure 6 shows a comparison between a pyroprocessing plant and a PUREX plant, revealing a similar structure: spent fuel storage, reprocessing plant, fuel fabrication plant, and fresh fuel storage. From the viewpoint of safeguardability in terms of the storage of spent and fresh fuels, the same safeguard measures (containment and surveillance, C&S measures, as well as item counting) can be applied. Pyrochemical processing plants for commercial spent nuclear fuel reprocessing are more compact than aqueous reprocessing plants of the same throughput. The compactness of the pyroprocessing plant mitigates the physical protection measures to be applied. Hot-cell blocks are ideal for physical protection, while the higher radiation stability of the chemical reactants facilitates the processing of spent fuels with short cooling. Plants are usually involved in both the reprocessing of spent fuel and fabrication of fresh fuel. Due to the high radiotoxicity of transuranics, the strong neutron and gamma radiation emitted by the spontaneous fission of actinides, and the decay of fission products, the process takes place within a single alpha tight enclosure with concrete shielding. The hot cell complex is divided into three mass balance areas:

- The first comprises the spent fuel storage and the element chopper
- The second the electrorefining of the fuel
- The final consists of the fuel production venue with storage of the fabricated fuel.

Between the areas are key measurement points, KMP, which account for the transfer of nuclear material. All entries and outlets are subjected to C&S measures. Similarly to the DUPIC project, the interior of the hot cells are monitored by cameras to survey any clandestine activities. In the first material balance area, MBA, item counting together with non-destructive analysis, NDA is applied, while in the other two near real-time accounting methods, NRTA. Immediately stored products are under seal.

6. FUTURE DIRECTION OF PYROPROCESSING

Pyrometallurgical technology has the potential to treat spent fuels, not only metal fuel, but also oxide fuel, especially MOX fuel with high heat generation, because of the non-use of degradation reactants by radiation. Metal fuel that has been well fitted with the pyroprocessing can achieve a high breeding ratio exceeding 1.3, making metal fueled FBR a potentially feasible nuclear system for forthcoming generations in India and China, both of which foresee enormous future energy demand. As proposed in the IFR program, the metal fuel cycle is positioned as a Generation-IV system in the U.S. Currently, extensive activity of research and development has been initiated in those states with the potential to recycle minor actinides in mind, as represented with the Actinide Recycle Reactor, ARR. Meanwhile, pyroprocessing is deployed as a batch-wise operation and assembled with compact devices,

which is better adapted for a collocation system positioned with some reactors and a single processing facility rather than for a centralized reprocessing unit with large scale throughput. This system is more advantageous in terms of proliferation resistance because fissile materials, except for externally-supplied uranium and trace amounts of actinides in wastes, can be confined to a single monitored site without any transportation of TRUs within public spaces, which facilitates safeguarding efforts. This is more acceptable in certain countries where insufficient grid lines have been improved. The further potential of metal-fueled fast reactors can maintain a breeding ratio of 1.0 without any blanket fuel. This indicates that no sources exist to produce Pu with high purity without changing the process.

Last but not least, the direction of R&D for practical installation is suggested. The current stage of technology features the exploitation of the engineering model and its application. The process should be verified and optimized by the use of genuine material with a scale of hundreds of kilograms. In parallel, extensive efforts will be required to explore an engineering scale installation with a transport system of melts, molten salt and liquid metal, and a heating system with cooling and ventilation of inert atmosphere. A concept of an operational system must be established in which a computational simulation as well as a design study are effective components.

7. SUMMARY AND CONCLUSION

An advanced fuel cycle with a next generation fast reactor mitigates the energy issue emerging this century by satisfying environmental safety and proliferation resistance. Not only the U.S. and France, but also Asian countries are significantly investigating the scope of improvement of current technology and innovative technology. For example, pyroprocessing has been investigated in nations like China and India, which will have to satisfy vast future energy demand and for the fuel cycle technology of actinide burning by metal fast reactors like the GNEP program. Intensive research has been carried out on electrorefining, electrochemical reduction, injection casting, and the reductive extraction of actinides. Verification by the use of irradiated materials and development engineering scale installation are current issues of interest. From a technological achievement perspective, a decade would be sufficient to design a pilot scale facility of 50kg/batch/ device by completing the remote handling and melt transfer line. Another emerging issue is the establishment of a credible safeguard concept in order to ensure the proliferation resistance. The ideal image is a collocation of fast reactors and a pyroprocessing plant from where the wastes without actinides may be evacuated. Lastly, a metal fuel cycle with pyroprocessing with potentials, such as a high breeding ratio and no additional process to recover

minor actinides, can provide a future nuclear system which satisfies the need for compactness, strong proliferation resistance, and environmental friendliness.

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