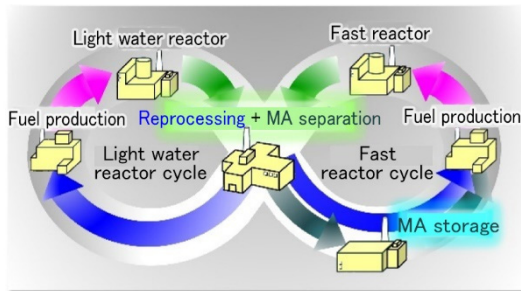


Toward a Sustainable and Eco-friendly Nuclear Fuel Cycle - Initiatives to Advanced Fuel Reprocessing Plant -



KAZUYA KISHIMOTO*¹ MINEFUMI MATSUI*²

TARO KATO*²

TAISUKE TSUKAMOTO*³

MASANORI SHIKATA*⁴ TAKUYA KONO*⁵

Japan's basic nuclear policy is to promote the nuclear fuel cycle, and the "Basic Policy for the Realization of GX"⁽¹⁾ approved by the Cabinet in 2023 states that Japan aims to develop and construct next-generation advanced reactors in addition to utilizing existing light water reactors. In line with the development of the next-generation advanced reactor, it is necessary to accelerate the consideration of reprocessing technology in order to establish the next-generation nuclear fuel cycle. Mitsubishi Heavy Industries, Ltd. has been engaged in research and development of next generation reprocessing process for many years. And in our recent effort, the principle verification of separation and direct conversion technologies have been completed by using simulated minor actinides for the study on minor actinide separation and storage technology, which is one of the core technologies. We will continue to proceed with the development toward the realization of the next-generation reprocessing process using these technologies.

1. Introduction

Japan regards nuclear power as a quasi-domestically produced energy, and has a basic policy to promote the nuclear fuel cycle, in which spent fuel is reprocessed and the recovered uranium (hereinafter referred to as U), plutonium (hereinafter referred to as Pu), etc. are effectively used, from the viewpoints of effective utilization of resources, volume reduction of high-level radioactive waste, and reduction of potential harmfulness, etc.

Based on this policy, the Federation of Electric Power Companies of Japan is promoting the introduction of the plutonium thermal use in which mixed oxide fuel (hereinafter referred to as MOX fuel) is used in light water reactors, as the immediate destination for recovered Pu, etc.⁽²⁾ As indicated in the Fast Reactor Development Policy⁽³⁾ and the Strategic Roadmap⁽⁴⁾, it is planned to utilize recovered Pu etc. in fast reactors in the future. Therefore, in addition to the light water reactor cycle, research and development of the next generation reprocessing process is required to establish the fast reactor cycle in accordance with the technological development on the power reactor side.

This paper introduces the development status of the minor actinide (hereinafter referred to as MA) separation and storage process⁽⁵⁾, which Mitsubishi Heavy Industries, Ltd. (hereinafter referred to as MHI) are conducting research and development as one of the core technologies for the next-generation reprocessing process.

2. Future vision of nuclear fuel cycle

2.1 Nuclear fuel cycle mission

The nuclear fuel cycle mainly consists of a nuclear power plant, a fuel production facility, and a reprocessing facility, and the nuclear fuel is continuously utilized in the following steps (i) to (iv).

- (i) Nuclear fuel is used to generate electricity at the nuclear power plant.

*1 Manager, Nuclear Fuel Cycle Engineering Department, Nuclear Energy Systems

*2 Nuclear Fuel Cycle Engineering Department, Nuclear Energy Systems

*3 Chemical Research Department, Research & Innovation Center

*4 General Manager, Nuclear Fuel Cycle Engineering Department, Nuclear Energy Systems

*5 Senior Project Manager, Nuclear Fuel Cycle Engineering Department, Nuclear Energy Systems

- (ii) The spent fuel is processed in the reprocessing facility, and fissile materials (U, Pu, etc.) are separated from the waste and recovered.
- (iii) The recovered fissile materials are processed into nuclear fuel at the fuel production facility.
- (iv) The nuclear fuel from (iii) is supplied to the nuclear power plant to generate electricity.

By repeating this cycle, nuclear power can be used continuously as a quasi-domestically produced energy for a long period of time. In addition, reprocessing spent fuel to reduce the volume of waste can reduce the potential harmfulness.

2.2 Future vision of nuclear fuel cycle and technological development requirements

Figure 1 shows a future vision of the nuclear fuel cycle. Currently, with the aim of realizing the left-hand side of Figure 1, construction and operation of nuclear fuel cycle facilities with improved safety is underway, taking into account lessons learned from the Fukushima Daiichi Accident. In the medium to long term, as shown in the right-hand side of Figure 1, the next-generation reprocessing process including fast reactors and MA recovery technology will be introduced while continuing to use the light water reactor, thereby achieving the aforementioned nuclear fuel cycle mission more efficiently as shown in Figure 2.

The technological development requirements necessary to realize the medium- to long-term future vision are as follows.

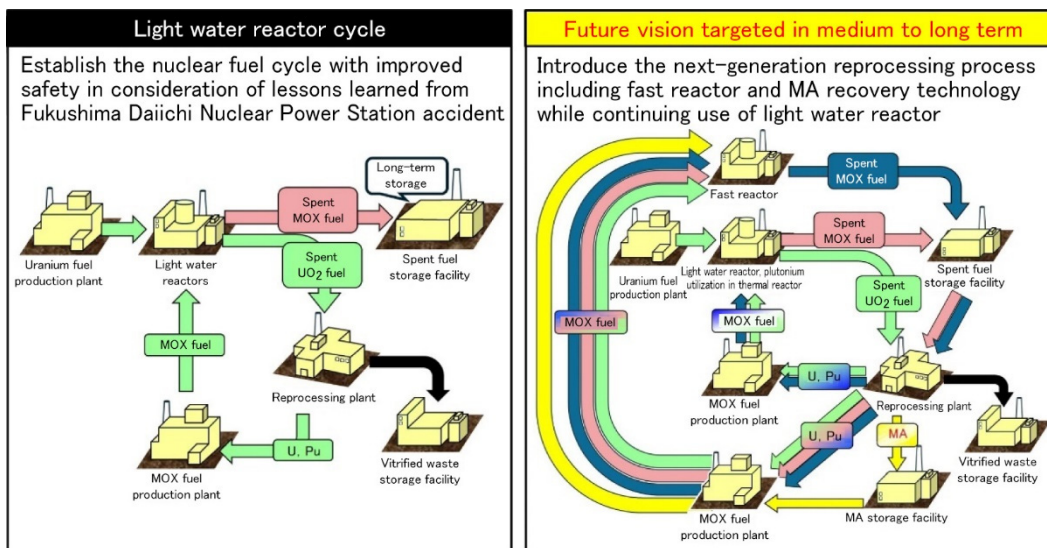


Figure 1 Future vision of nuclear fuel cycle

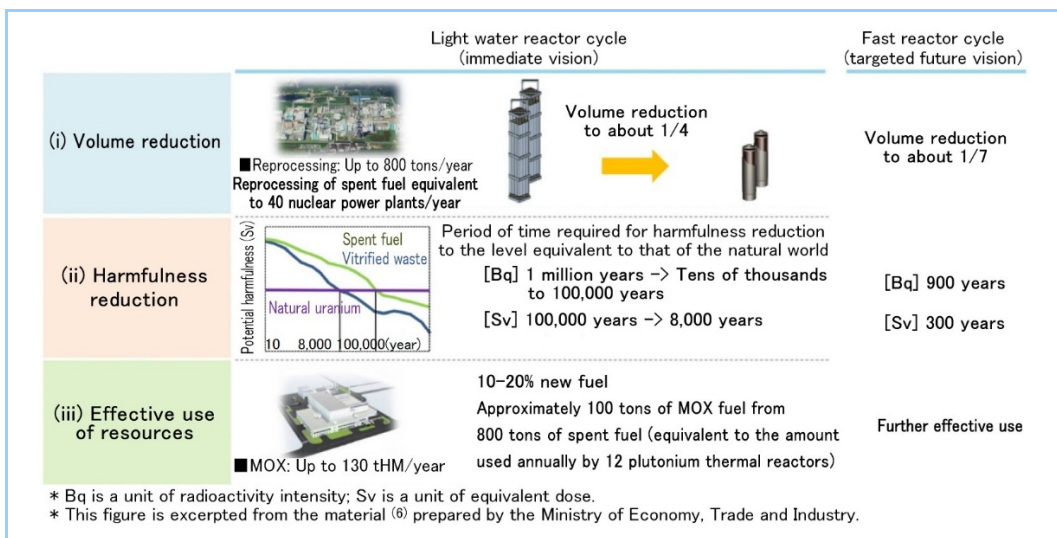


Figure 2 Advantages of nuclear fuel cycle⁽⁶⁾

(1) Reprocessing of various types of spent fuel

The next-generation reprocessing needs to reprocess spent MOX fuel and spent fuel generated from fast reactors (hereinafter referred to as “spent fast reactor fuel”) in addition to

spent uranium fuel from existing light water reactors that can be reprocessed at Rokkasho Reprocessing Plant (hereinafter referred to as RRP). As shown in **Table 1**, the Pu and MA content of these spent fuels is several to several dozen times higher than that of spent uranium fuel, which must be considered in the safety design of the reprocessing process (criticality safety, decay heat removal, etc.) In addition, since the fuel geometries of light water reactors and fast reactors are different, it is necessary to establish a reprocessing process that takes these differences into account.

Table 1 Characteristics, impact on reprocessing process, and issues of spent fuels

Item	Light water reactor fuel [UO ₂]	Light water reactor fuel [MOX]	Fast reactor fuel [MOX]	Impact on reprocessing process, and issues
Pu quantity	1	Approx. 6	Approx. 13	- Increased dissolution time in dissolution facility - Reflection on criticality control design (use of absorbent, shape control, etc.) - Reflection on shielding design (increasing the shielding thickness, etc.) - Reflection on decay heat design (enhancement of heat removal capability, etc.)
MA quantity	1	Am: Approx. 7 Cm: Approx. 12	Am: Approx. 7 Cm: Approx. 30	- Impact on vitrification operation (Changes in operating parameters due to increase in MA concentration, etc.) (Increase in the number of vitrified waste units due to heat generation limit, etc.)
FP quantity	1	Approx. 0.8	Approx. 2.3	- Impact on vitrification operation (Changes in operating parameters due to increase in FP concentration, etc.)

* Comparison with spent uranium fuel. For all fuels, the cooling period is assumed to be 15 years.

(2) Technology for recovering MA from high-level radioactive liquid waste

Spent fuel contains MA, which is highly radioactive for a long period of time and generates decay heat. In conventional reprocessing processes, MA is disposed of as high-level radioactive waste together with fission products (hereinafter referred to as FP), which causes an increased burden on the repository. Therefore, it is being considered to introduce MA recovery technology into the next-generation reprocessing process to prevent MA from migrating to high-level radioactive waste, thereby further enhancing the volume reduction of high-level radioactive waste and the reduction of potential harmfulness (**Figure 3**). The recovered MA can be eliminated by burning as a fast reactor fuel.

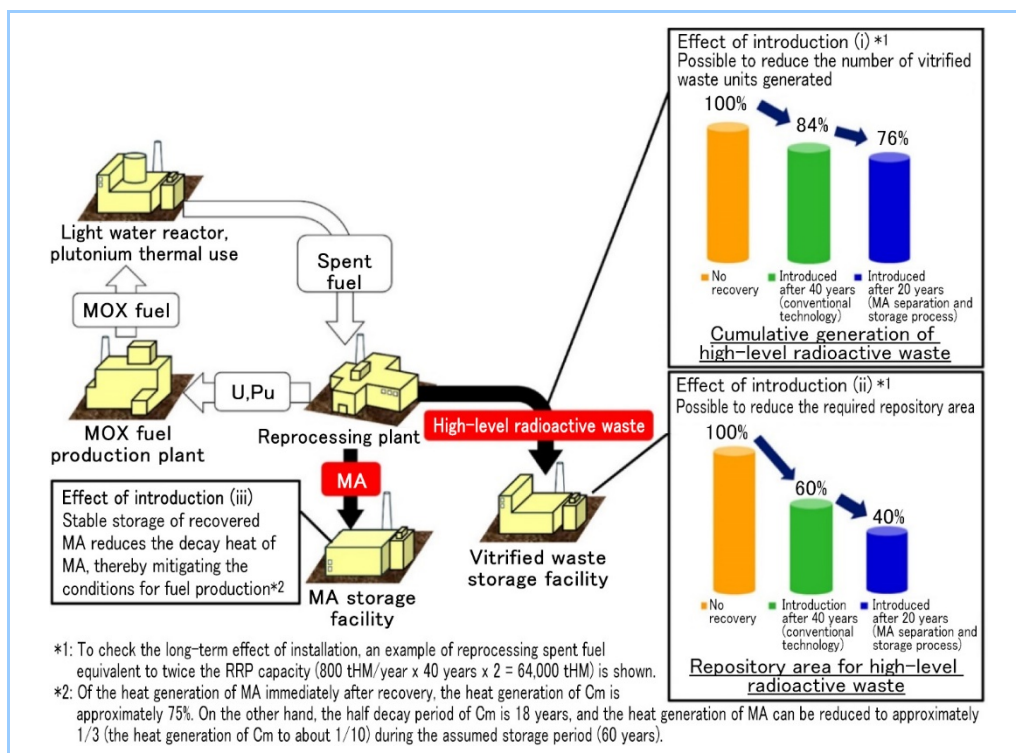


Figure 3 Effects of introducing MA separation and storage process

The following Chapter 3 describes our envisioned next-generation reprocessing process based on (1) and (2), and Chapter 4 introduces the MA separation and storage process that MHI has been developing as one of the core technologies.

3. Concept of next-generation reprocessing process

Figure 4 shows the main process plan of a reprocessing process (wet process) that can process spent light water reactor fuel (spent uranium fuel and spent MOX fuel) and spent fast reactor fuel as a typical example of a next-generation reprocessing process.

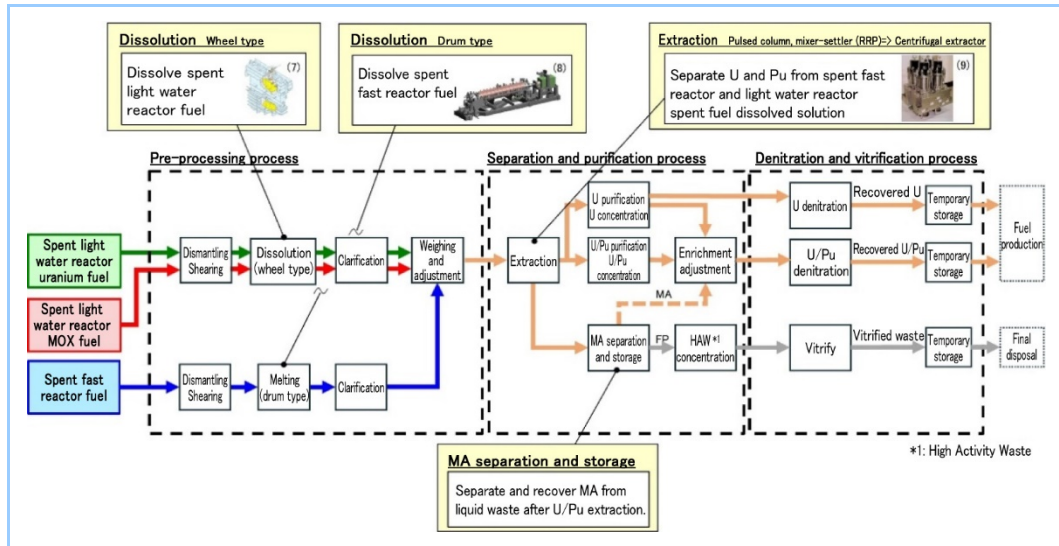


Figure 4 Main process plan applying next-generation reprocessing process

The main processes in the reprocessing process can be roughly classified into (1) to (3) as follows.

(1) Pre-processing process

Shear the spent fuel, dissolve it in nitric acid solution, and remove insoluble residues from the dissolved solution (clarification).

(2) Separation and purification process

Separate and purify the U and Pu contained in the dissolved solution by extraction technology using organic solvents, and then concentrate and adjust the degree of enrichment of the U solution and U/Pu solution (adjust the U/Pu concentration ratio by mixing). Recover and store MA before stabilization in the vitrification process, which remains with FP in the liquid waste after removal of U/Pu in the separation and purification process, and mix it with U/Pu solution as appropriate.

(3) Denitration and vitrification process

Denitrate and convert U/Pu dissolved in nitric acid solution to solid oxides, and mix the high-level radioactive liquid waste generated from the separation and purification process with molten glass to produce vitrified waste.

The above process is basically similar to RRP⁽⁷⁾, but there are major differences in the pre-processing process and the separation and purification process.

The pre-processing process of the next-generation reprocessing process differs from that of RRP in that the processing line for spent light water reactor fuel is separated from that for spent fast reactor fuel. This is because of the difference in the equipment design policy (equipment type), such as disassembly method and subcriticality control method, due to the difference in the shape and composition of the spent fuel, and also because of the difference in the operational constraints, such as the fact that the spent fast reactor fuel has more insoluble Pu, which requires a longer dissolution time than the spent light water reactor fuel. As for the specific equipment type of the dissolver, which is the main equipment of the pre-processing process, the spent light water reactor fuel processing line will use a wheel type dissolver (same type as RRP) that can perform large-scale processing, while the spent fast reactor fuel processing line will use a drum type dissolver that has better subcriticality control. The fuel types of spent uranium fuel and spent MOX

fuel, which are both spent light water reactor fuels, are the same, so it is planned to process them by using the same equipment with process conditions adjusted accordingly.

The separation and purification process of the next-generation reprocessing process is planned to introduce a centrifugal extractor developed by Japan Atomic Energy Agency instead of the extractors used in RRP (pulsed column and mixer-settler). The size of the centrifugal extractor is small and it holds less process solution, which is superior to the pulsed column and mixer-settler in terms of criticality safety control and radiation-induced solvent degradation, which are concerns due to the increase in Pu and MA.

Furthermore, the most significant feature of the separation and purification process is the introduction of a new MA separation and storage process currently under technology development. The next-generation reprocessing process will be able to separate and recover MA from the extraction liquid waste using this technology (see Chapter 4), in contrast to the RRP, where the liquid waste after U/Pu extraction is vitrified to solid waste.

4. Status of technological development of MA separation and storage process

4.1 Concept of MA separation and storage process

Many domestic and foreign companies and research institutes have so far developed technologies to recover highly harmful MA from high-level radioactive waste and burn it using fast reactors and other equipment. However, these technologies were based on the assumption that MA recovered at a reprocessing facility would be burned in a fast reactor immediately, and therefore MA recovery would take place only after the commercialization of fast reactors. For this reason, there is an issue that MA from high-level radioactive wastes generated before the introduction of fast reactors cannot be separated and removed. In addition, conventional MA recovery processes require operations such as back-extraction and elution when MA trapped in the organic phase or adsorbent is processed in the next step, resulting in the generation of several tens of times the amount of liquid waste as secondary waste compared to the high-level radioactive waste to be processed, and there are concerns about the increase in process load due to storage and processing of this liquid waste.

To solve these problems, MHI has been working on the technological development of an MA separation and storage process, which is a new process that introduces a direct conversion process to convert MA recovered as organometallic complexes in the organic phase directly into oxides without back-extraction, and a temporary storage process to stably store the oxides containing MA over the medium to long term (**Figure 5**). The features of the MA separation and storage process MHI is developing are as follows.

- The MA recovered as organometallic complexes is directly converted without back-extraction, thereby reducing secondary waste generated by the back-extraction process.
- The recovered MA is made into fluorite structural oxide, which is chemically stable over a wide temperature range and has a chemical form similar to that of nuclear fuel pellets, thereby ensuring mid- to long-term stability against temperature rise due to decay heat originating from MA.
- The MA that has been made into fluorite structural oxide can be dissolved in nitric acid in the same way as nuclear fuel pellets, which facilitates the processing of MA in the storage when it is processed into fuel.
- The decay of curium (Cm), a short-half-life radionuclide, during the storage period reduces the decay heat, thus mitigating the cooling conditions (reduction of decay heat to about 1/3 to 1/5) at the fuel production facility that handles MA after storage.

Based on the above features, the implementation of the MA separation and storage process will make it possible to introduce MA recovery as early as possible, regardless of when fast reactors are put to practical use, and is expected to improve the reduction effect on the amount of waste and potential harmfulness. In addition, it will be possible to reduce secondary waste generated by MA recovery and mitigate cooling conditions in MA fuel production, thereby reducing the overall processing load of the process.

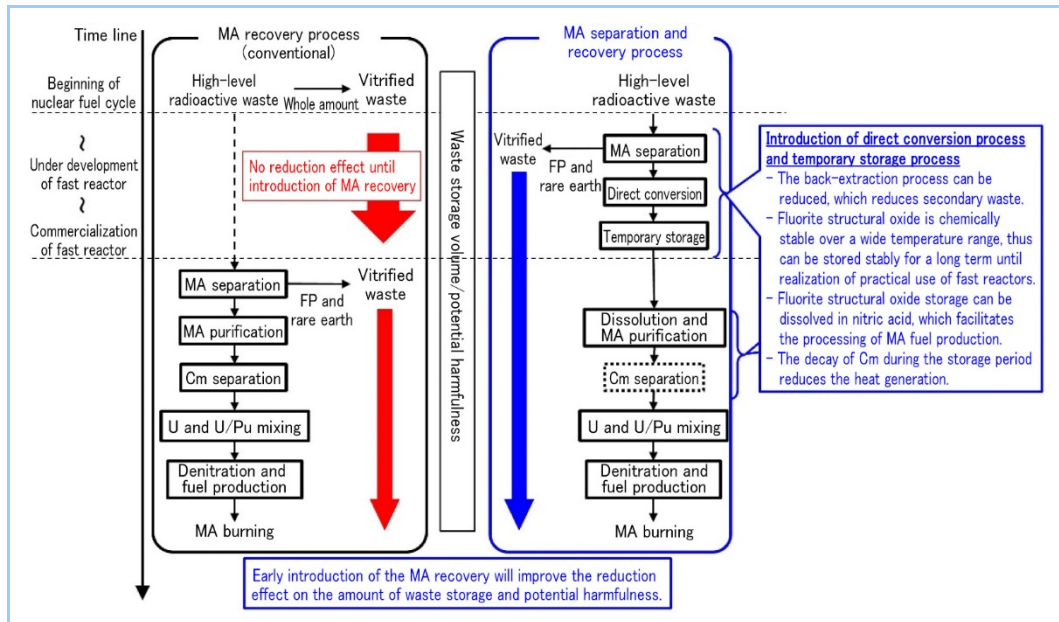


Figure 5 Comparison of conventional MA recovery process and MA separation and storage process

4.2 Major technologies for MA separation and storage process

Figure 6 shows the schematic flow of the MA separation and storage process. The major technologies of the MA separation and storage process consist of (1) MA separation technology and (2) direct conversion technology. MHI is developing proprietary MA separation and direct conversion technologies using flame-retardant low vaporization heat diluent as core technologies for stable storage of MA and reduction of secondary waste. The following sections summarize the major technologies.

(1) MA separation technology

The conventional extraction separation process mainly uses n-dodecane, which is flammable (flash point: 70°C), as a diluent, and thus requires fire prevention measures. In addition, the recovery of MA and other substances from the organic phase after extraction requires back-extraction, which results in a problem with the generation of secondary wastes such as liquid waste. Therefore, MHI is developing MA separation technology using flame-retardant low vaporization heat diluents such as hydrofluorocarbons (HFCs), which are safe due to their flame-retardant properties and can be easily recovered by evaporation due to their low vaporization heat. In addition, we use diglycolamide (DGA) based, nitrilotriacetamide (NTA-amide) based, and other extractants, which are composed of the four elements of carbon, hydrogen, oxygen, and nitrogen (CHON elements) for easy oxidative decomposition processing, to facilitate oxidation and decomposition of the remaining extractants in the subsequent direct conversion process.

(2) Direct conversion process

The MA separation and storage process requires technology to convert recovered MA into a chemically stable form over a wide temperature range, since it is assumed that the recovered MA will be stored for a medium to long period of time. MHI adopts a fluorite-structured oxide of UO_2 , which is chemically stable and has been well proven in the nuclear fuel field, as the storage form, and are developing a direct conversion technology to produce an oxide in which MA is solidly dissolved in a fluorite structure composed mainly of UO_2 by burning recovered MA in a reducing atmosphere with the addition of U. As described in section (1), the MA separation and storage process can recover the diluent by distillation and oxidatively decompose the extractant to gases such as CO_2 and H_2O by directly converting the organic phase after extraction and separation using an extractant composed of flame-retardant low vaporization heat diluent and CHON elements in the MA recovery process. This reduces the back-extraction operation required in the conventional extraction separation process, reduces secondary waste, and also suppresses the generation of gases derived from residual

organic matter during storage.

As described above, the direct conversion technology involves three steps of processing after adding U to the post-extraction and separation organic phase: (1) recovery of diluent, (2) decomposition and removal of CHON elements, and (3) fluorite structuring.

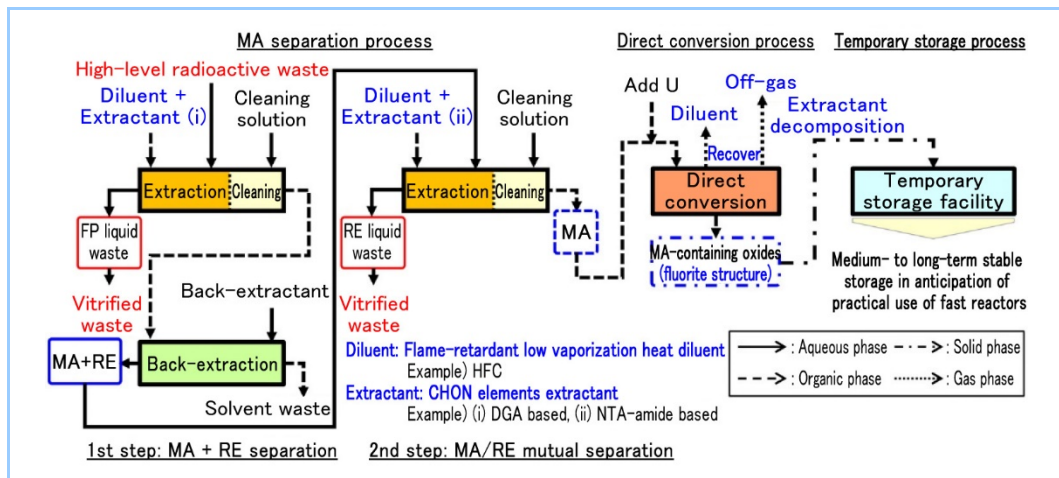


Figure 6 Schematic flow of MA separation and storage process

4.3 Status of technological development of MA segregation and storage process

As for the technological development of the MA separation and storage process, we have completed a conceptual study of the process and verification of the principle using simulated MA, such as neodymium (Nd) and europium (Eu). In addition, we are proceeding with the evaluation of the effects of elements contaminated in MA, such as FP and rare earth (hereinafter referred to as RE), considering the composition of high-level radioactive waste, and the acquisition of test data for hot tests, as efforts toward practical application. Specifically, as for MA separation technology, we are acquiring performance evaluation data for the development of MA separation process using decafluoropentane, a flame-retardant low vaporization heat diluent, and tetra-2-ethylhexyl-diglycolamide, a CHON element extractant, and we have verified as a result of americium (Am) extraction equilibrium tests that highly efficient recovery of more than 90% is possible at the nitric acid concentration assuming high-level radioactive waste (Figure 7). As for the direct conversion technology, we have conducted feasibility verification tests using Nd and Eu simulating MA for the three steps of the processing: (1) recovery of diluent, (2) decomposition and removal of CHON elements, and (3) fluorite structuring. In the steps (1) and (2), high diluent recovery and CHON decomposition rates were achieved by burning in an atmospheric atmosphere, and fluorite structuring was also verified. Furthermore, in the step (3), by burning in a reducing atmosphere with hydrogen gas flow, it was verified that fluorite structuring with high crystallinity is possible under the condition that Nd, which is a simulated MA, and zirconium (Zr), which is a contaminated element, are contained by 30 to 40% with respect to U (Figure 8).

Currently, we are also developing a mutual separation process for MA/RE to reduce the RE co-extraction rate (a countermeasure against the increase in storage volume due to RE accompaniment) in order to further improve economic efficiency. The flame-retardant low vaporization heat diluent that is currently being considered as a candidate falls under the category of chlorofluorocarbon substitutes, but we will continue to screen diluents with low GWP (global warming potential) and flame-retardant low vaporization heat in order to reduce emissions of chlorofluorocarbons as a countermeasure against global warming. In addition, we are planning to conduct performance evaluations of compositions that simulate high-level radioactive waste in consideration of the fact that high-level radioactive waste contains a large number of elements, although we have so far conducted principle verification and performance evaluations by testing single elements and mixtures of a few elements.

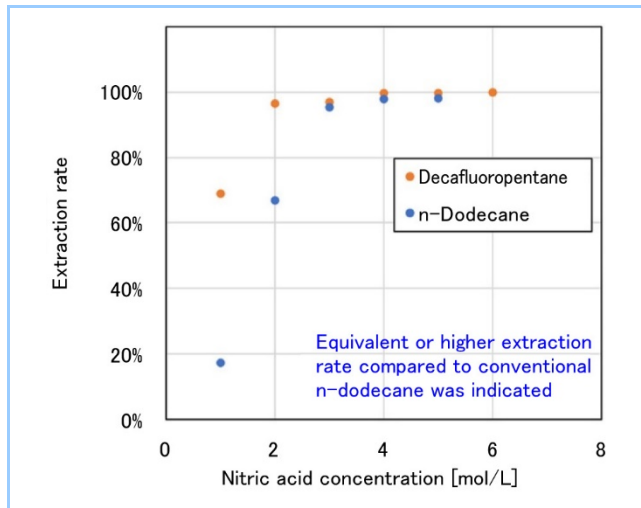


Figure 7 Am extraction performance with low vaporization heat diluent and CHON element extractant

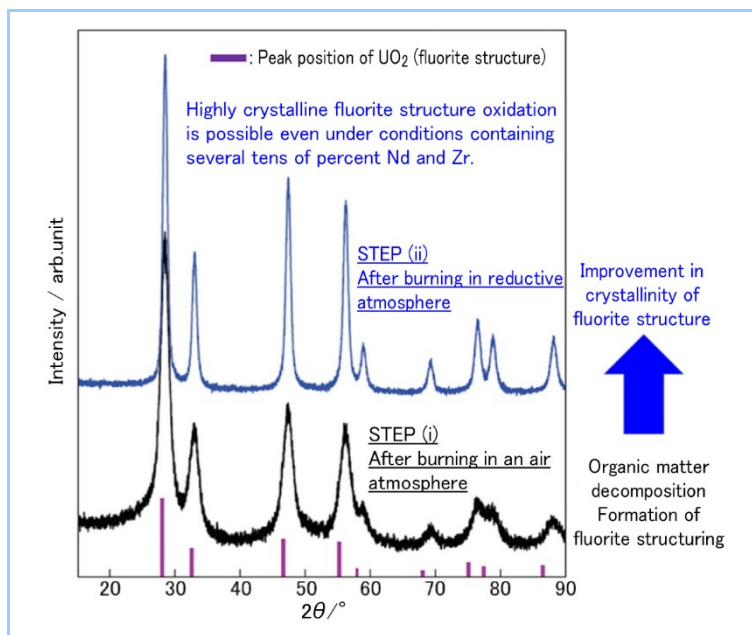


Figure 8 X-ray diffraction (XRD) patterns after direct conversion for Nd and Zr

5. Conclusion

This report presented a future vision of the next-generation nuclear fuel cycle in line with the transition from light water reactors to fast reactors, aiming at effective use of resources through steady reprocessing of spent fuel, volume reduction of high-level radioactive waste, and reduction of potential harmfulness, and described the requirements for next-generation reprocessing facilities necessary to realize this vision, as well as the importance of new technology (MA separation and storage process).

In particular, the MA separation and storage process that MHI is developing enables medium- to long-term storage of recovered MA by making it into a chemically stable fluorite-structured oxide over a wide temperature range, making it possible to start MA recovery as early as possible and increase the reduction effects on the volume and potential harmfulness of high-level radioactive waste.

MHI will steadily work on technological development toward the realization of the next-generation fuel cycle, with a view to the future of nuclear power generation in the mid- to long-term.

References

- (1) Ministry of Economy, Trade and Industry, The Basic Policy for the Realization of GX - A roadmap for the next 10 years -, (2023)
- (2) The Federation of Electric Power Companies of Japan, The New Pluthermal Program, (2020)
- (3) Nuclear Ministerial Meeting, Policy for Fast Reactor Development, (2016) (in Japanese)
- (4) Nuclear Ministerial Meeting, Strategic Roadmap, (2022) (in Japanese)
- (5) Mitsubishi Heavy Industries, Ltd., JP, 7155031, B Method for reducing disposal load of high-level radioactive waste (in Japanese)
- (6) Ministry of Economy, Trade and Industry, Efforts to Establish the Nuclear Fuel Cycle and Future Considerations (in Japanese)
- (7) Japan Nuclear Fuel Limited, <https://www.jnfl.co.jp/en/>
- (8) Japan Atomic Energy Agency JAEA Technology Seeds, https://rdreview.jaea.go.jp/review_en/2019/e2019_1_9.html
- (9) T. Washitani et al., Development of centrifugal extractor for fast reactor fuel reprocessing, JNC Technical Review, No.21 (2003) (in Japanese)