Feasibility of MA Transmutation by (MA, Zr)H_x in Radial Blanket Region of Fast Reactor and Plan of Technology Development

Kazumi IKEDA[1], Kenji KONASHI[2], Kazuo IKEDA[3], Kunihiro ITOH[3], Fumiki MIZUSAKO[4], Mutsumi HIRAI[4], Hiroaki MUTA[5], Ken KUROSAKI[5]

[1] Mitsubishi FBR Systems, Inc., Shibuya-ku, Tokyo, 150-0001, Japan

[2] Institute for Materials Research, Tohoku University, Oarai, Ibaraki-ken, 311-1313, Japan

[3] Nuclear Development Corporation, Tokai-mura, Ibaraki-ken, 319-1111, Japan

[4] Nippon Nuclear Fuel Development Co. Ltd., Oarai, Ibaraki-ken, 311-1313, Japan

[5] Division of Sustainable Energy and Environmental Engineering, Graduate School of Engineering, Osaka University, Suita, Osaka-fu, 565-0871, Japan

E-mail: kazumi_ikeda@mfbr.mhi.co.jp

Abstract. This paper shows a feasibility study of transmuting minor-actinide (MA) by MA-zirconium hydride, (MA, Zr)H_x. And it also introduces a program of technology development for the MA target. A proposed concept of the (MA, Zr)H_x target can enhance the transmutation rate. The reason is that it densely contains both of MA and hydrogen. The feature creates the neutron energy spectrum shift to promote nuclear reactions. It was assumed in calculations that 54 assemblies of Am bearing target are arranged around an active core in a 280 MW_e sodium-cooled reactor. The MA transmutation performance is significant: the irradiation of 615 days transmutes 39% of the initial Am amount, the Am transmutation rate is 67 kg/effective full power year; the transfer ratio of Am to repository reduces by one-fortieth. Integrity of the targets is predicted to be feasible on desk analysis: the maximum linear heat rate is about 310 W/cm. However, handling of the used targets is a matter of concern because the decay heat decreases to 7.5 kW/target after cooling period of 500 days.

Key Words: Transmutation, Minor actinide, Hydride, Fast Reactor

1. Introduction

This paper highlights the feasibility and efficiency of a methodology that transmutes minor-actinide (MA) by minor-actinide zirconium (MA-Zr) hydride in the blanket region of fast reactors. Nuclear energy is required to be sustainable as well as safe. Reducing MA is an important issue of radioactive waste in order to promote nuclear energy in social and technological circumstances [1]. For this reason, Japan has been developing the technologies of partitioning and transmutation since 1980s [2].

Recently, three systems have been proposed for MA transmutation: (1) conventional fast reactors, (2) innovative reactors using new type fuels, and (3) accelerator driven system. Utilizing conventional fast reactors is reasonable because of early implementation, small risk of development, and predictability of cost [3-4]. Therefore, our research will develop a technology of MA-Zr hydride target in order to transmute MA in fast reactors; and also evaluate the feasibility and efficiency.

There are two types of transmutation methodology using fast reactors: homogeneous and heterogeneous cycles. They each have good and bad points, and the selection depends on what is important for different stakeholders [5]. Japan decided to employ a low-

decontamination fuel technique [6] to achieve cost reduction in nuclear fuel cycle. Radioactive fuels will be fabricated by remote handling in manufacturing cells and MA will not be separated from Pu. If the fuel fabrication technique is realized and the safety of MA bearing reactor core is validated, the homogeneous cycle will be technologically confirmed to operate as safely as conventional reactors. On the other hand, the heterogeneous cycle is also promising in the case that Am, Cm, and lanthanoid elements can be separated; MA bearing targets can be fabricated, and it remains sound during irradiation. This system is robust because fuels can be supplied, if MA target-cycle stops. Moreover, the heterogeneous cycle can be introduced as soon as a small target-manufacturing facility starts to run.

We have researched a new concept of long-lived fast reactor control rod, utilizing hafnium hydride [7-10]. This R&D is also a development program of technology utilizing hydride material and fast reactor. This paper shows a preliminary result.

2. Research Program Overview

2.1. Objectives

The final objective of the R&D is enhancing MA transmutation. That is, the MA transmutation is tripled or quadrupled by the hydride target, compared with metal and oxide targets [11]. The research program from FY 2016 to FY 2019 is the first phase of the R&D.

2.2. Program of Research

The program will develop three element technologies, and will evaluate the feasibility and efficiency of the target in fast reactor fuel cycle.

2.2.1. Experimental production and measurement of MA-Zr hydride

This research will manufacture MA-Zr hydride pellets and measure the physical properties. The testing is planned to be done in RIAR (Scientific Research Institute of Atomic Reactors) of Russia in full consultation with the proposer of this R&D and Japanese specialists.

2.2.2. Observation of irradiation behaviour

Physical properties of MA-Zr hydride which is doped surrogate material of fission products, will be measured. Those of Th-U-Zr hydride irradiated at JMTR (Japan Materials Testing Reactor) will be also measured at PIE (post irradiation experiment). These data will be utilized for irradiation tests of the following phases.

2.2.3. Safety characteristics at normal operation and accidents

Hydrogen permeation rate through cladding at normal operation and ejection rate at accidents will be measured in mock tests. And it will be also observed how much oxide on MA-Zr hydride pellet surface supresses the ejection.

2.2.4. System analyses

Characteristics of MA bearing reactor core and material balance based on deployment scenarios will be calculated. Manufacturability of the MA targets will be also examined.

2.3. Research Organization

The Institute for Materials Research, Tohoku University is leading this program, and the institute, Mitsubishi FBR Systems, Inc., Nuclear Development Corporation, Nippon Nuclear Fuel Development Co. Ltd., and Osaka University are jointly conducting the studies.

3. Preliminary Study of MA Transmutation

3.1. Mark, Issues, and System Concept

The ultimate goal of the MA transmutation is to get the sustainability of nuclear energy and global circumstance. This will be realized by reducing the amount of MA, lowering the radiation exposure, and scaling down the required repository per output. The enhanced MA transmutation is better for it; the MA transmutation ratio at unloading should be high enough to suppress the transfer ratio of MA to repository. Naturally, the target should be sound. That is, the MLHR (maximum linear heat rate) and He accumulation are limited so as to keep integrity of the targets. Decay heat of the targets is also limited at fabrication and at handling. Figure 1 illustrates the scope, mark, and issues of this study.

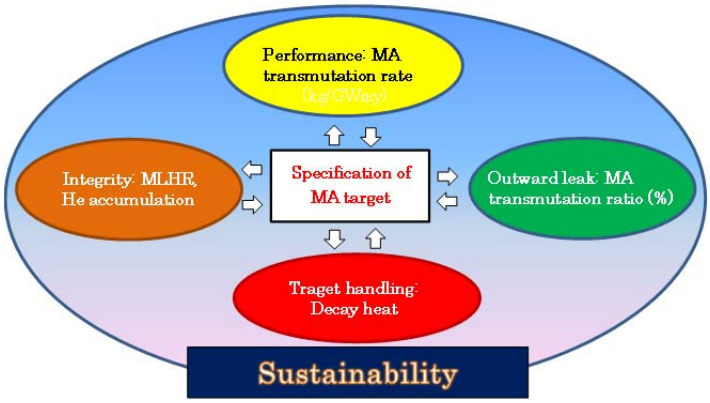

FIG. 1. Objectives of the parameter study.

Figure 2 shows the system concept of this study. Actinide elements of used fuels are separated into U, Pu, Np, Am, and Cm. U, Pu, and Np are recycled into reactor fuels; Am into MA targets. ^{244}Cm is stored to spontaneously transmute into ^{240}Pu.

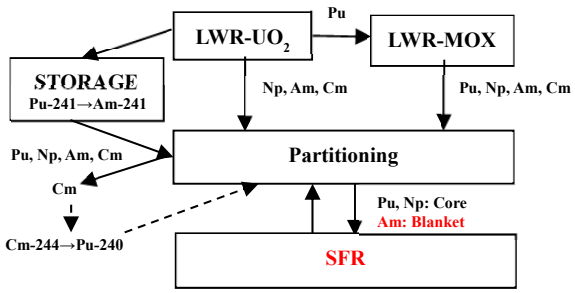

FIG. 2. Concept of nuclear fuel cycle.

3.2. Design Requirements

- ✓ The MA transmutation ratio is set to be 30% or higher.
- ✓ The decay heat is 3 kW/target or smaller at fabrication [12].
- ✓ The decay heat is 40 kW/target or smaller at unloading from reactor core, and 7.5 kW/target or smaller at sodium cleaning [5].

3.3. Calculation Condition and Methods

3.3.1. Calculation condition

The MA targets contain 61 pins as shown in TABLE I, and Am is set to be an interested element. The isotope ratio (^{241}Am, ^{242m}Am, ^{243}Am) is set to be (59.7%, 0.2%, 40.1%) (weight fraction). The specification of reactor core is shown in TABLE II, and the core configuration is drawn in Fig. 3. The length of reactor cycle is 123 EFPD (effective full power days), and the reactor core fuels are exchanged in four batches.

TABLE I: SPECIFICATION OF MA TARGET.

Items	Values
Fuel type	$(Am_{0.1}, Zr_{0.9})H_{1.7}$
Number of pins	61
MA height (mm)	930
Duct flat to flat (mm)	105
Pin diameter (mm)	12.0
Cladding thickness (mm)	0.5

TABLE II: SPECIFICATION OF SODIUM-COOLED REACTOR.

Items	Values
Active core height (mm)	930
Upper axial blanket height (mm)	300
Lower axial blanket height (mm)	350
Assembly pitch (mm)	116

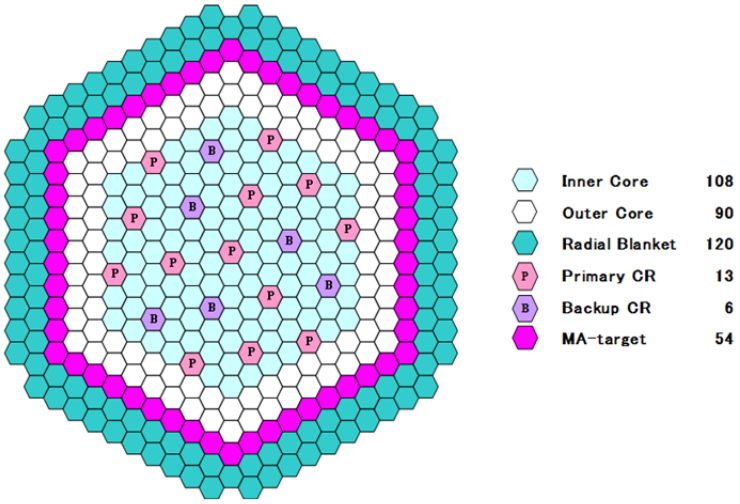

FIG. 3. Core Configuration of Sodium-cooled Fast Reactor.

3.3.2. Calculation methods

A fast reactor group constant set JFS-3-J4.0 [13], and one-dimensional burn up calculation code ODDBURN produced 7-group effective cross sections. Using these cross sections, three-dimensional diffusion calculation code TRISTAN and three-dimensional prismatic discrete ordinate code ENSEMBLE-TRIZ [14] calculated nuclear transmutation. The actinide cross sections in a nuclear data file ORLIBJ40 [15] were replaced into the one-group

cross sections produced from the nuclear calculation, and the isotope generation and depletion code ORIGEN2 [16] calculated decay heat.

3.4. Preliminary Results

3.4.1. Nuclear characteristics of MA-Zr hydride targets

TABLE III presents micro cross sections and the amount of MA in three targets: ($MA_{0.1}$, $Zr_{0.9}$)$H_{1.7}$, ($MA_{0.1}$, $Zr_{0.9}$), and ($MA_{0.1}$, $Zr_{0.9}$)O_2. According to the table, deploying hydrogen decuples the capture cross section of ^{241}Am and ^{243}Am; hydrogenating MA-Zr alloy can also bear large amounts of MA and hydrogen in the targets. Reference [11] details the comparison.

TABLE III: MICRO CROSS SECTION AND MA INVENTORY OF TARGETS.

Characteristics	Case	($MA_{0.1}$, $Zr_{0.9}$)$H_{1.7}$		($MA_{0.1}$, $Zr_{0.9}$)		($MA_{0.1}$, $Zr_{0.9}$)O_2	
Micro Cross Section (barns)	Nuclide	Fission	Capture	Fission	Capture	Fission	Capture
	^{237}Np	0.4	12.9	0.3	1.6	0.3	2.2
	^{241}Am	0.5	24.3	0.2	1.9	0.2	2.5
	^{242m}Am	69.0	12.6	3.1	0.5	3.7	0.6
	^{243}Am	0.3	18.5	0.2	1.6	0.2	2.2
	^{242}Cm	1.0	4.0	0.7	0.5	0.7	0.7
	^{243}Cm	31.5	4.9	3.1	0.6	3.7	0.7
	^{244}Cm	0.6	10.3	0.4	0.7	0.4	1.0
	^{245}Cm	23.4	3.5	2.8	0.5	3.3	0.7
	^{246}Cm	0.4	3.3	0.3	0.5	0.3	0.7
Inventory of MA (kg)	Target	6.7		7.8		5.2	

3.4.2. Change of nuclide composition

Figure 4 plots the transmutation of ^{241}Am in the targets, and Fig. 5 does that of ^{243}Am. The irradiation of 2952 EFPD (24 cycles × 123 EFPD) changes 97% of Am into Cm, Pu, FP (fission products): ^{241}Am transmutes into predominately ^{242}Cm ($T_{1/2}$ = 162.94 days) and then ^{238}Pu; ^{243}Am into mainly ^{244}Cm ($T_{1/2}$ = 18.11 y), and then ^{245}Cm.

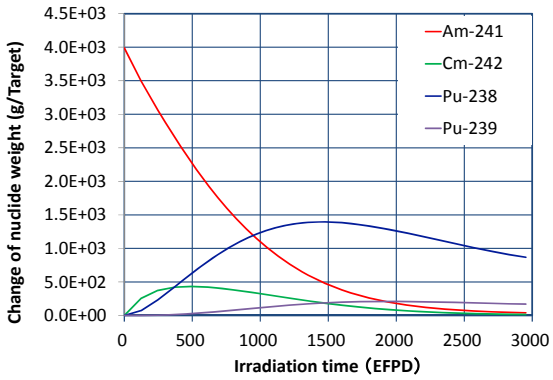

FIG. 4. Composition change by ^{241}Am transmutation in the maximum heat target.

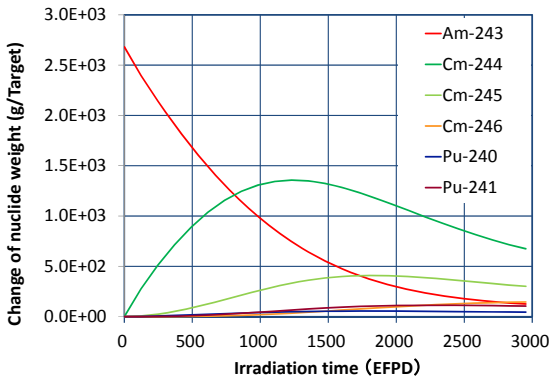

FIG. 5. Composition change by ^{243}Am transmutation in the maximum heat target.

3.4.3. Transfer ratio to repository

Figure 6 shows the relation of the transfer ratio of Am to repository and the transmutation ratio. The transfer ratio is calculated in the case of recycling Am as follows.

$$L_{cum} = (1 - R_{recyc.}) / Tr \qquad (1)$$

where

L_{cum}: Transfer ratio of Am to repository,

$R_{recyc.}$: Recovery ratio of Am at recycling (99% is assumed),

Tr: Transmutation ratio of Am target at unloading.

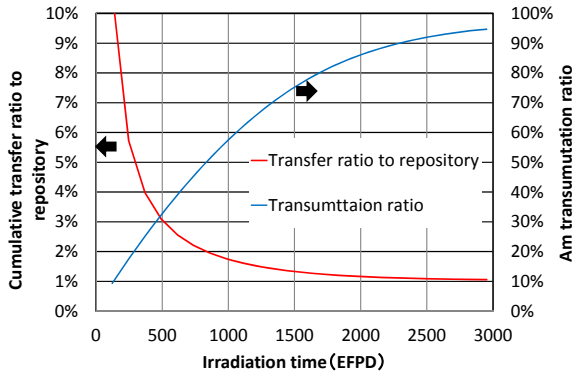

FIG. 6. Transfer ratio of Am to repository by recycling loss.

The average Am transmutation ratio attains 30% at 500 EFPD as shown in Fig. 6. And it becomes 39% at 615 EFPD (5 cycles × 123 EFPD) and the transfer ratio of Am to repository reduces by one-fortieth, in the case that the recovery ratio of Am at recycling is 99%. Further, the irradiation of 2952 EFPD transmutes 95% of the initial amount, in which the transfer ratio becomes about 1%.

3.4.4. Average number of Am recycling

Figure 7 indicates the average number of Am recycling up to transmutation. That is, as the transmutation ratio increases, the required number of recycling decreases. The average number is 2.6 at the irradiation length of 615 EFPD, and 1.1 at 2952 EFPD as shown in Fig. 7. It should be noted that the small number of recycling reduces the recycle cost in that the number of targets to be fabricated and the amount of reprocessing decrease.

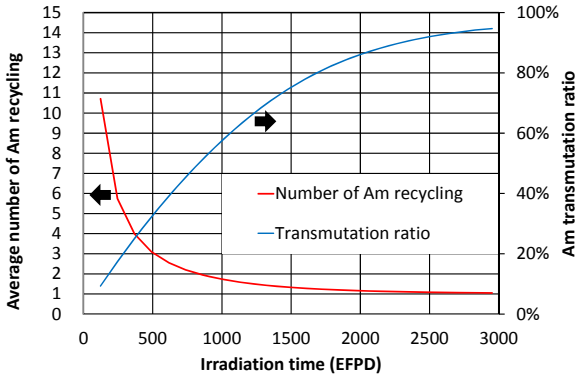

FIG. 7. Average number of Am recycling up to transmutation.

3.4.5. Am transmutation rate

Figure 8 shows the Am transmutation rate and transmutation ratio. The Am transmutation rate is 67 kg/EFPY (effective full power year) at reprocessing with irradiation length of 615 EFPD. Naturally, as the transmutation ratio increases, the transmutation rate decreases, and the number of required fast reactor increases.

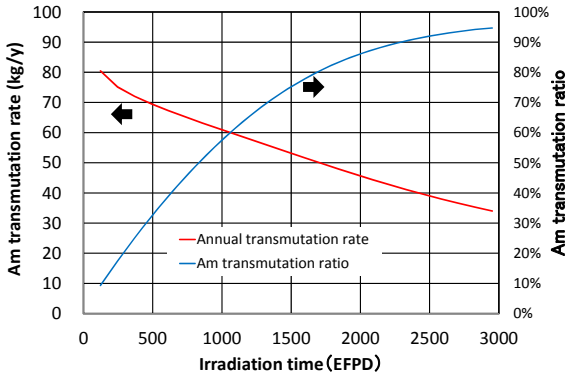

FIG. 8. Am transmutation rate.

3.4.6. Maximum linear heat rate

Figure 9 plots the history of MLHR of the maximum heat target. The MLHR becomes 310 W/cm at 615 EFPD and the peak is 500 W/cm.

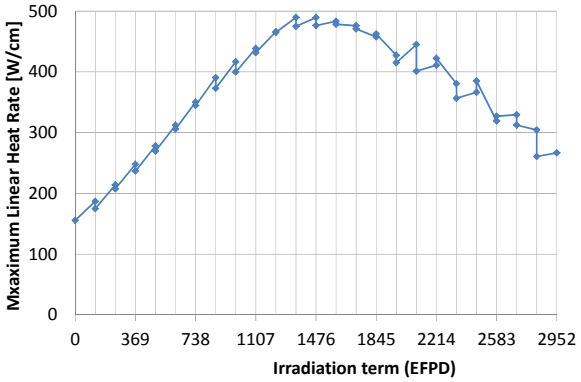

FIG. 9. Maximum linear heat rate of Am target.

3.4.7. Gas accumulation

Figure 10 shows the production of noble gas and volatile elements in the maximum heat target. That is, He is produced from α decay of ^{242}Cm and ^{244}Cm; Br, Kr, I, Xe, and Cs from fission reaction. That becomes seven moles (atom) at 615 EFPD, as shown in Fig. 10. Assuming that the plenum capacity of pins is 100 cc, and the release ratio is 100%, the average pressure of plenum becomes 7 MPa at 500˚C. Furthermore, the irradiation of 2952 EFPD produces 27 moles. On the other hand, if the release ratio is low, swelling of the pellets and PCMI (pellet clad mechanical interaction) are concerned.

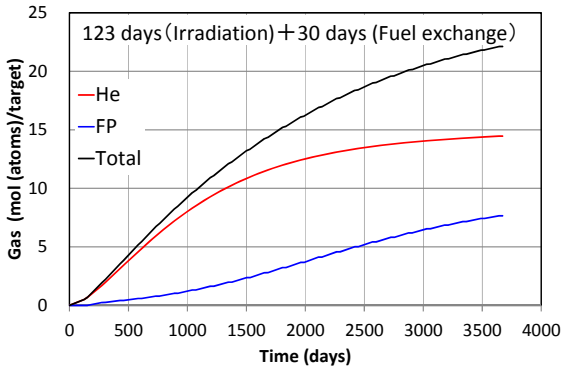

FIG. 10. Accumulation of noble gas and volatile elements in the maximum heat Am target.

3.4.8. Decay heat of Am target before and after irradiation

The decay heat of an unirradiated target is 470 W/target at the set isotope ratio. Figure 11 shows the decay heat of the maximum heat target after the irradiation of 615 EFPD; it becomes smaller than 40 kW/target after cooling period of 50 days, and 7.5 kW/target after 500 days.

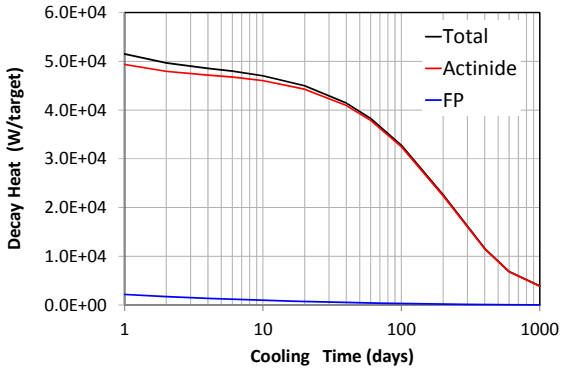

FIG. 11. Decay heat of the maximum heat Am target after the irradiation of 615 EFPD.

4. Conclusions

This paper has revealed the feasibility and efficiency of the proposed methodology that transmutes MA by MA-Zr hydride targets in the blanket region of a 280 MW_e fast reactor. This study is a part of the R&D program that is developing the element technologies of MA-Zr hydride target. The preliminary analysis yielded the following results.

(1) The proposed concept of the MA-Zr hydride target has been confirmed to enhance the MA transmutation rate. That is, deploying hydrogen enhances tenfold the absorption cross sections of ^{241}Am and ^{243}Am. Hydrogenating MA-Zr alloy can produce large amounts of

MA and hydrogen in the target in place of moderator pins. As a result, the Am transmutation rate is 67 kg/EFPY at the irradiation length of 615 EFPD in the case that 54 targets are arranged.

(2) It has been confirmed that the deployment of the targets reduces the transfer ratio of MA to repository by one-fortieth at the irradiation length of 615 EFPD. In addition, the proposed concept was suggested to reduce the MA recycling cost: the reprocessing amount of Am target becomes smaller because the required number of recycling is 2.6 (See 3.4.4).

(3) It has been predicted that the targets might remain sound under irradiation for 615 EFPD: the MLHR is 310 W/cm; and the pressure of gas plenum is 7 MPa, assuming 100 cc of the plenum capacity of target pins, and 100% of the release ratio. It should be noted that the acceptable MLHR has never been determined due to a lack of physical property data, and the plenum of target pins has not been designed yet.

(4) The handling of the used targets is a matter of concern: the decay heat of the maximum heat target after the irradiation of 615 EFPD becomes smaller than 40 kW/target after cooling period of 50 days and 7.5 kW/target after 500 days. It has been confirmed that the decay heat of the Am target is not a serious fabrication issue because it is 470 W/target, which is smaller than the limited value 3 kW/target.

(5) The large volume of He and fission gas is a serious technological design problem in the case of irradiation of 2952 EFPD. That is, the deep burnup of Am is challenging in the integrity of the Am targets, although it has advantages in the reduction of radiotoxicity in repository and the recycling cost.

Therefore, it has been concluded that the proposed concept can improve the performance of MA transmutation and the sustainability of nuclear energy in social and technological circumstance. However, in the case of the set specification of Am targets and irradiation length of 615 EFPD, the handling of the used targets is a matter of concern. In the case of the irradiation length of 2952 EFPD, the accumulation of fission gas and He highlights the plenum design of pins and PCMI. A parameter study of the target specification is presently underway, aiming to outline optimized MA transmutation.

ACKNOWLEDGMENTS

The authors thank Mr. Suguru Tada (IX Knowledge Inc.) for technical assistance. The present study includes the result of “Development of MA-Zr hydride for early realization of transmutation of nuclear wastes” entrusted to Tohoku University by the Ministry of Education, Culture, Sports, Science, and Technology of Japan (MEXT).

REFERENCES

[1] NUCLEAR ENERGY AGENCY, Potential Benefits and Impacts of Advanced Nuclear Fuel Cycles with Actinide Partitioning and Transmutation, NEA no. 6894, OECD-NEA, Paris, ISBN 978-92-64-99165-1 (2011).

[2] IKEDA, K., KOYAMA, S., KURATA, M., MORITA, Y., et al., “Technology Readiness Assessment of Partitioning and Transmutation in Japan and Issues toward Closed Fuel Cycle,” Prog. Nucl. Energy, **74** (2014) 242-263.

[3] U.S. GOVERNMENT ACCOUNTABILITY OFFICE, Nuclear Fuel Cycle Options DOE Needs to Enhance Planning for Technology Assessment and Collaboration with Industry

and Other Countries, GAO-12-70, GAO, Washington D. C. (2015).

[4] WIGELAND, R., TAIWO, T., LUDEWIG, H., TODOSOW, M., et al., Nuclear Fuel Cycle Evaluation and Screening, INL/EXT-14-31465, INL, Idaho Falls (2014).

[5] NUCLEAR ENERGY AGENCY, Homogeneous versus Heterogeneous Recycling of Transuranics in Fast Nuclear Reactors, OECD-NEA, Paris, ISBN 978-92-64-99177-4 (2012).

[6] SATO, K., HIGUCHI, T., KOIKE, K., et al., Feasibility Study on Commercialized Fast Reactor Cycle Systems Technical Study Report of Phase II (2) Nuclear Fuel Cycle Systems, JAEA-Research 2006-043, JAEA, Tokai-mura, Ibaraki-ken, Japan (2006) [in Japanese].

[7] IWASAKI, T. and KONASHI, K., "Development of Hydride Absorber for Fast Reactor - Application of Hafnium Hydride to Control Rod of Large Fast Reactor-," J. Nucl. Sci. Technol., 46, **8** (2009) 874-882.

[8] IKEDA, K., MORIWAKI, H., OHKUBO, Y., IWASAKI, T., KONASHI, K., "Application of Hafnium Hydride Control Rod to Large Sodium Cooled Fast Breeder Reactor," Nucl. Eng. Des., **278** (2014) 97-107.

[9] KONASHI, K., IWASAKI, T., ITOH, K., HIRAI, M., et al., "Study on an Innovative Fast Reactor Utilizing Hydride Neutron Absorber - Final Report of Phase I Study," Proc. ICAPP2010, San Diego (2010) 2098-2104 (CD-ROM).

[10] KONASHI, K., ITOH, K., KIDO, T., KOSAKA, Y., SEINO, S., "Development of Hydride Neutron Absorber for Fast Reactor -Irradiation Experiment on Hydride Neutron Absorber in BOR-60-, Proc. ICAPP 2013, Jeju Island, Korea (2013) (CD-ROM).

[11] KONASHI, K., IKEDA, K., ITOH, K., HIRAI, M., et al., "Enhancing MA Transmutation by Irradiation of (MA, Zr)H_x in FBR Blanket Region," Proc. Global 2015, Paris (2015).

[12] ITOH, K., IKEDA, K., HISHIDA, K., KURODA, T., et al., "Development of the Cooling Technology on TRU Fuel Pin Bundle during Fuel Fabrication Process, (1) Whole Study Plan and Fabrication of Test Apparatuses," Proc. ICAPP'08, Anaheim (2008) (CD-ROM).

[13] SUGINO, K., JIN, T., HAZAMA, T., NUMATA, K., Preparation of Fast Reactor Group Constant Sets UFLIB.J40 and JFS-3-J4.0 Based on the JENDL-4.0 Data, JAEA-Data/Code 2011-017, JAEA, Tokai-mura, Ibaraki-ken, Japan (2011) [in Japanese].

[14] IKEDA, K., MORIWAKIA, H., NAKAZATO, W., "Nuclear Calculation Methodology and Development of 3-D Transport Nuclear Design Code," Fast Reactors and Related Fuel Cycles: Challenges and Opportunities (Proc. FR'09, Kyoto, 2009), IAEA, Vienna, ISBN 978-92-0-102410-7, Paper 06-32.FP (2012) (CD-ROM).

[15] OKUMURA, K., SUGINO, K., KOJIMA, K., JIN, T., OKAMOTO, T., KATAKURA, J., A Set of ORIGEN2 Cross Section Libraries Based on JENDL-4.0: ORLIBJ40, JAEA-Data/Code 2012-032, JAEA, Tokai-mura, Ibaraki-ken, Japan (2012) [in Japanese].

[16] CROFF, A.G., ORIGEN2-A Revised and Updated Version of the Oak Ridge Isotope Generation and Depletion Code, ORNL-5621, ORNL, Oak Ridge (1980).