Proceedings of the Reactor Physics Asia 2015 (RPHA15) Conference
Jeju, Korea, Sept. 16-18, 2015

Study of Reactivity Control Method by Metallic Deuteride Burnable Poison for Accelerator Driven System

Hiroki Yamaguchi*, Tatsuro Takani, Naoto Aizawa and Tomohiko Iwasaki
Department of Quantum Science and Energy Engineering, Graduate School of Engineering, Tohoku University, 6-6-01-2, Aramaki-Aoba, Aoba-ku, Sendai 980-8579, Japan
* Corresponding author: hiroki_yamaguchi042@neutron.qse.tohoku.ac.jp

1. Introduction

Long-lived fission products (LLFPs) and minor actinides (MAs) are contained in a High Level Waste (HLW) and their transmutations are the effective technology for reducing a burden of its disposal. Accelerator-driven system (ADS) has been studied for transmuting. ADS is composed of a high-intensity proton accelerator, a spallation target, and a subcritical core. Since the subcritical core is employed, ADS is possible to load a large amount of MA into the core. In Japan, the Japan Atomic Energy Agency (JAEA) has proposed an 800 MWt LBE (lead-bismuth eutectic) -cooled ADS which employs a nitride fuel, and is driven by 1.5GeV proton beam [1, 2]. JAEA-proposed-ADS can transmute about 250kgMA/year [1].

There are many issues for the development of ADS and the design of beam window is the critical one. The beam window is placed on the boundary between a beam duct and the LBE target, and exposed to severe environments such as heat generation from proton beam, irradiation from the beam and neutron, and external pressure from the spallation target. To compensate the decrease of the effective multiplication factor at end-of-cycle (EOC), the beam current must be increased more than 20 mA but the complex design is necessary for the window to withstand such the high-intensity proton beam. The maximum beam current should be reduced to 14 – 16 mA if a simple hemispherical design is employed.

Reactivity control by loading the burnable poison (BP) into the core is a promising the method for reducing the maximum beam current. In the previous study, BP assembly with gadolinium (Gd) and zirconium (Zr) hydride pins is applied to the ADS core for reactivity control [3]. By loading BP assembly, maximum beam current is reduced to 14.7 mA from 21.9 mA. This result indicates that the reactivity control method by using Gd and Zr hydride is effective. However, the total amounts of transmutation of MA are decreased from 478.8 to 457.9 kg/cycle. The deterioration of the transmutation capability is the problem of this method.

Metallic hydride is used to increase the number of neutrons which is absorbed by Gd. But loading Gd into the core causes the reduction of total amounts of transmutation of MA, because moderating effect of hydrogen remains till EOC so that the number of fast neutron is decreased. One option for improving this problem is the use of deuterium instead of hydrogen. The moderation effect and the absorption of neutron of deuterium are smaller than those of hydrogen by the use of deuterium. There is some possibility to improve the ability of the reactivity control and the transmutation by adjusting the amount of thermal neutron to an appropriate amount.

This paper describes the effect of replacing hydrogen with deuterium and aims to reduce the maximum beam current and increase the amount of transmutation of MA. Chapter 2 explains the ADS model and computational method used in this study. In the chapter 3, the Gd and Zr deuteride pins are applied to the core, and the optimal BP assembly design with deuteride pins are surveyed. Finally, this study is summarized in chapter 4.

2. ADS Model and Computational Method

2.1 ADS model and structure of BP assembly

The research target model was the ADS model proposed by JAEA, which is referred to as the “reference core” in this paper. The schematic diagram of the core is shown in Fig. 1, and its main parameters are listed in Table I. LBE is used as coolant and target. Four fuel zones with 84 fuel assembly surrounds the target. The fuel is composed of plutonium-MA mixed nitride and zirconium nitride. The detail of fuel compositions is shown in Table II. One fuel assembly is composed of 391 fuel pins and 6 tie rods.

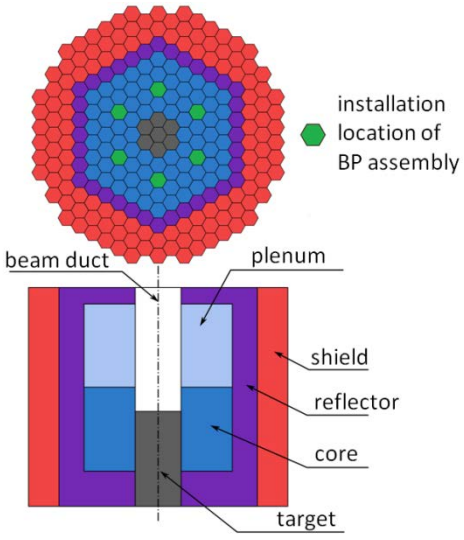

Fig. 1. ADS core model in this study

Table I. Parameters of the ADS model

Plant	Initial k_{eff}	0.97
	Fuel	(Pu+MA)N + ZrN
	Coolant inlet temperature	573.15K
	Coolant velocity	2.0m/s
Fuel assembly	Type	Hexagonal, ductless
	Pitch	233.9mm
	Width	232.9mm
	Total assembly height	3740mm
Fuel pin	Pu ratio in(Pu + MA)N	36.2vol%
	Inert matrix ratio of reference core	67.1vol%
	Pin outer diameter	7.65mm
	Thickness of cladding	0.5mm
	Pin pitch	11.48mm

Table II. Isotropic compositions of MA and Pu

Isotope	MA [wt%]	Pu [wt%]
^{234}U		0.04
^{236}U		0.01
^{237}Np	49.65	
^{238}Pu		2.38
^{239}Pu		54.48
^{240}Pu	0.32	24.19
^{241}Pu		10.85
^{242}Pu		6.96
^{241}Am	32.1	1.09
^{242m}Am	0.06	
^{243}Am	13.37	
^{242}Cm	0.03	
^{244}Cm	4.04	
^{245}Cm	0.39	
^{246}Cm	0.04	

BP assembly and fuel assembly has the same arrangement of pins. The structure of it is shown in Fig. 2. BP assembly is composed of 66 Gd-hydride pins and 331 Zr-hydride pins. Gd-hydride pin is placed in outer rim of the BP assembly, and Zr-hydride is located at the other part. BP assembly is arranged at 6 places on fuel zones (shown in Fig. 1).

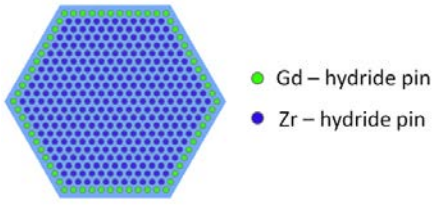

Fig. 2. Structure of BP assembly.

2.2 Computational method

MVP [4] was employed to calculate neutron flux, neutron spectrum, and effective multiplication factor (k_{eff}) of the ADS core, and the Japanese evaluated nuclear data library JENDL-4.0 [5] were adopted as the nuclear data. The total history of neutrons in eigenvalue calculations and fixed-source calculations was 10,000,000. The fuel assembly and the BP assembly are treated as a detailed structure, and the target, the reflector and the shielding are handled as a homogeneous composition. For the burnup calculation, MVP-BURN [6] was used. 21 nuclides from U-234 to Cm-244 were treated in the burnup chain model as heavy nuclides and 79 nuclides were considered as fission products and BP nuclides. The time step of burnup calculations was 100 days. The total history of neutrons in burnup calculations was 3,000,000. PHITS (Particle and Heavy Ion Transport code System) [7] were employed for the transport calculations of high-energy neutrons more than 20 MeV and the calculation results was given to MVP and MVP-BURN calculations as neutron source because MVP and MVP-BURN could perform only a calculation by energy less than 20 MeV.

3. Effect of Metallic Deuteride BP and Optimization of BP Pin Arrangement

3.1 Evaluation index and the targeted value in the design

The following evaluation indexes were set to evaluate the effect by adopting deuterium instead of hydrogen.

- Burnup reactivity swing
  Burnup reactivity swing was defined in the following equation.

$$\text{Burnup reactivity swing } [\%\Delta k/kk'] = \frac{k_{eff\,MAX} - k_{eff\,MIN}}{k_{eff\,MAX} \times k_{eff\,MIN}} \times 100 \qquad (1)$$

When the change of k_{eff} during the burnup step is large, Burnup reactivity swing becomes large. This means that the maximum beam current must be increased to supplement the decrease of k_{eff}. This is the important index to judge whether it is possible for reactivity control by using the metallic deuteride BP.

- Maximum beam current
  The BP assembly was designed for the purpose of the reduction of the maximum beam current. This study aimed to reduce it less than 15mA.

- Amount of transmutation
  The amount of transmutation had decreased by the reactivity control using the BP in the previous study. Realizing the transmutation amount which is larger than that of the reference core is another purpose in this study.

3.2 Effect of the use of deuterium instead of all hydrogen

In this analysis, deuterium was employed instead of all hydrogen on the BP assembly which was used in the previous study. The reference core case (reference core), using metallic hydride case (GdH + All ZrH), and using metallic deuteride case (GdD + All ZrD) were compared

and the effect of the deuterium was investigated. The volume ratio of the BP pin is 20% of GdD and 80% of ZrD.

The result of the change of k_{eff} every case is shown in Fig. 3. The amount of transmutation became 485.2 kg which nearly equaled to that of reference core. This result indicated the improvement of the ability of transmuting. However, burnup reactivity swing was close to that of the reference core, and the maximum beam current was 21.6 mA which exceeded 15 mA. Consequently, the GdD + All ZrD case was not effective for the reactivity control. Neutron Spectra of the BP assembly in each core are shown in Fig. 4. In the thermal neutron region of GdD + All ZrD, the peak was not seen. This was because fast neutrons were not moderated enough for small moderation effect. Thus, the method to replace all hydrogen with deuterium is not effective for the reactivity control.

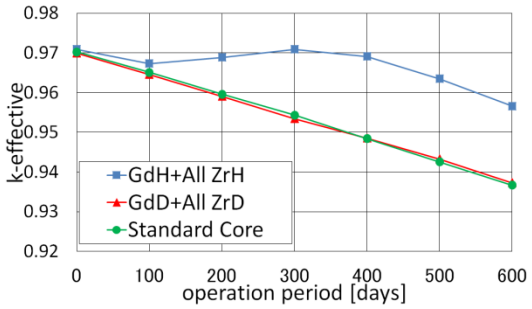

Fig. 3. Change of k_{eff} in every case.

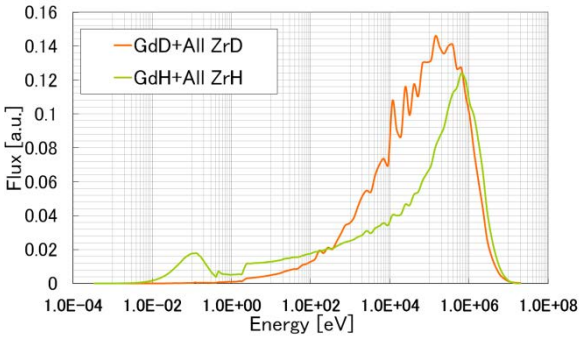

Fig. 4. Neutron Spectra of the GdH + All ZrH and GdD + All ZrD.

3.3 Optimization of BP pin arrangement in the BP assembly

In the foregoing section, it was not the effective method by replacing all hydrogen with deuterium. Loading both ZrH pin and ZrD pin in the BP assembly was investigated, and BP assembly was optimized by coordinating the loading number in this section. Two patterns for optimizing the arrangement of BP pin were discussed. We defined the loading ZrD pin from the outer row to inner row as pattern 1, and loading it from inner to outer as pattern 2. The schematics of loading patterns are shown in Fig. 5. Because there are ten BP pin rows in the assembly, 18 loading patterns existed of all loading patterns. The evaluation indexes mentioned above were compared by these 18 patterns for examining better design specifications. As well as the foregoing section, Gd was used for BP, and the volume ratio of the BP pin is 20% of GdD and 80% of ZrD.

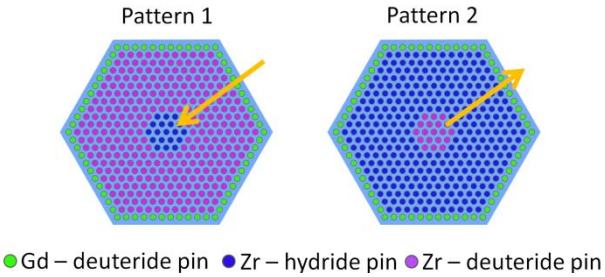

Fig. 5. Examples of the BP assembly by optimizations.

The change of k_{eff} and the spectrum in pattern 1 are shown in Figs. 6 and 7. From the viewpoint of the reactivity control, when only BP pins are replaced hydrogen with deuterium (GdD + ZrH), minimum burnup reactivity swing of 1.91 %Δk/kk' was obtained. The amount of transmutation of this case was 458.8 kg, which was similar to that of GdH + All ZrH case. The maximum beam current was 15.8 mA which was larger than that of GdH + All ZrH case. In the case of replacing hydrogen with deuterium in more than eight rows from the outer row (GdD + ZrD outside 8 row and GdD + All ZrD), the amount of transmutation of this case was closed to a reference core. However, the maximum beam current was 21.9 mA. From these results it was less possible for effective reactivity control with pattern 1. This was because thermal neutrons in the BP pin decreases by placing deuterium at the near position of BP pins. Arranging small quantities of deuterium near the BP pin led to decrease the neutron absorption of the Gd.

Next, the change of k_{eff} and the spectrum in pattern 2 are shown in Figs. 8 and 9. At the point of view of the reactivity control, burnup reactivity swing is minimized with 1.6 %Δk/kk' in the case of two rows from the inside are replaced hydrogen with deuterium (GdD + ZrD inside 2 row). The amount of transmutation of this case was 461.6 kg, which is similar to that of GdH + All ZrH case. The maximum beam current is 15 mA which achieves the target value. Thus, it is possible to control reactivity by pattern 2. At the point of view of the amount of transmutation, the maximum beam current become 16.9 mA when more than eight rows from the inside are replaced hydrogen with deuterium (GdD + ZrD inside 8 row and GdD + All ZrD). The amount of transmutation of this case was equal to a reference core (483.6 kg). From the above-mentioned result, pattern 2 was the effective method for improvement of the reactivity control and amount of transmutation.

Because of improving burnup reactivity swing and a maximum beam current in comparison with the reference core by applying the optimization of pattern 2, reactivity control was possible by substituting part of hydrogen of the BP assembly for deuterium. But reducing the maximum beam current was incompatible

with increasing the amount of transmutation. There are needs to change constitution of the BP assembly, in accordance with the performance requirement.

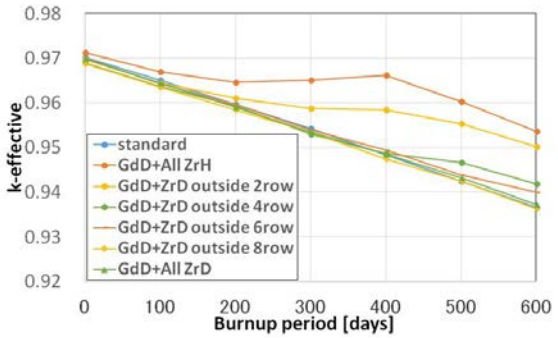

Fig. 6. Change of k_{eff} in pattern 1.

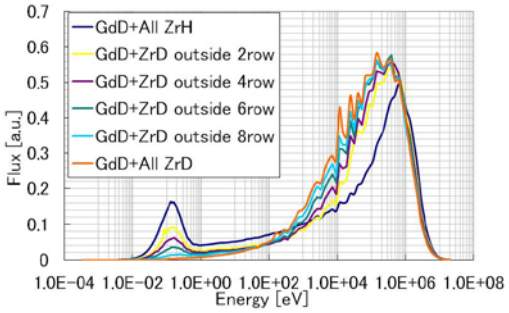

Fig. 7. Neutron Spectra of pattern 1.

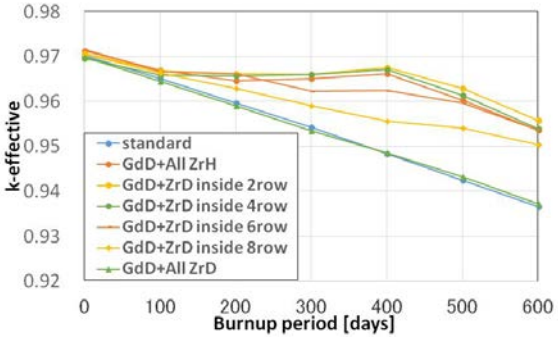

Fig. 8. Change of k_{eff} in pattern 2.

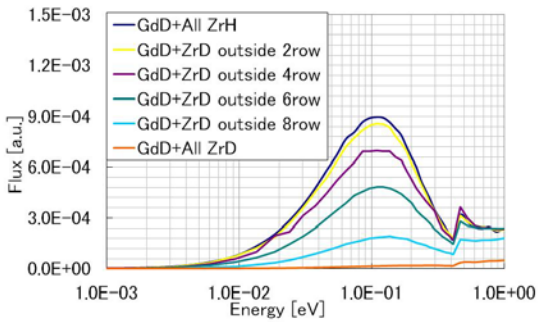

Fig. 9. Neutron Spectra of pattern 2.

4. Conclusion

A reactivity control method by using metallic deuteride instead of metallic hydride on the BP assemblies was studied for reducing the maximum beam current and increasing the total amounts of transmutation of MA. By comparing evaluation indexes, the optimal structure of the BP assembly was investigated.

When all hydrogen were replaced with deuterium, the amount of transmutation becomes 485.2kg. But burnup reactivity swing was close to the reference core and the maximum beam current was 21.6 mA. This case was not effective for the reactivity control.

Also loading both ZrH pin and ZrD pin in the BP assembly was investigated. In the case of loading ZrD pin from the outer row to inner row, the maximum beam current was larger than 15 mA in all cases. This pattern was not effective for reactivity control. When ZrD pins were loaded from the inner row to outer row, the amount of transmutation was 461.6 kg and the maximum beam current was 15 mA in the case of replacing hydrogen with deuterium on two rows from the inside. Also when more than eight rows from the inside were replaced hydrogen with deuterium, the maximum beam current became 16.9 mA and the amount of transmutation was 483.6 kg. From these results, it was revealed that realizing either reduction of the maximum beam current or increase of the amount of transmutation was possible by loading the metallic deuteride.

References

1. Tsujimoto K, Sasa T, Nishihara K, Takizuka T, Takano H, “Accelerator- driven system for transmutation of high-level waste,” *Prog Nucl Energy*., **37**, 339-344 (2000).
2. Tsujimoto K, Sasa T, Nishihara K, Takizuka T, Takano H, “Neutronics design for lead-bismuth cooled accelerator-driven system for transmutation of minor actinide,” *J Nucl Sci Technol*., **41**, 21-36 (2004).
3. Aizawa N, Iwasaki T, Watanabe Y, Takani T, “Study of reactivity control method by the application of gadolinium hydride to accelerator-driven system,” *J Nucl Sci Technol*., available online (2015).
4. Nagaya Y, Okumura K, Mori T, Nakagawa M, “MVP/GMVP II: general purpose Monte Carlo codes for neutron and photon transport calculations based on continuous energy and multigroup methods,” JAERI 1348, Tokai-mura(Japan): Japan Atomic Energy Research Institute (2005).
5. Shibata K, et. al., “JENDL-4.0: a new library for nuclear science and engineering,” *J Nucl Sci Technol*., **39**, 1-30 (2011).
6. Okumura K, Mori T, Nakagawa M, Kaneko K, “Validation of a continuous-energy Monte Carlo burn-up code MVP-BURN and its application to analysis of post irradiation experiment,” *J Nucl Sci Technol*., **37**, 128-138 (2000).
7. Iwase H, Niita K, Nakamura T, “Development of general-purpose particle and heavy ion transport Monte Carlo code,” *J Nucl Sci Technol*., **39**, 1142-1151 (2002).