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Resource Evaluation of Heavy Rare Earth Derived from the Spent Gd_2O_3 Burnable Poison in LWRs

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Abstract: A burnup calculation has been performed to evaluate heavy rare earth (terbium—Tb, dysprosium—Dy) production in spent gadolinium oxide (Gd_2O_3) installed as a BP (burnable poison). Each amount of Tb and Dy production derived from the BP has been about 30-40 times larger than those created in FP (fission products). Required cooling time to achieve exemption level on radioactivity concentration produced Tb and Dy derived from the BP are much shorter (BP-Tb: 7.9y, BP-Dy: < 0.1y) than those created in FP (FP-Tb: 3,616y, FP-Dy: 6.9y). However, the BP is mixed homogeneously with UO_2 pellet in current nuclear fuel system of LWRs (light water reactors), and hence mixing of FP cannot be not avoided. In such a mixture case, the required cooling time of recovered Tb will become 2,653y and that of recovered Dy be 4.8y. For this reason, recovered Tb is unlikely to be resource for utilization, while recovered Dy must be the resource provided the precise separation from the other FP.

Key words: Burnable poison, heavy rare earth, Tb, Dy, fission products, Adv.-ORIENT Cycle.

1. Introduction

Japan is one of the major RE (rare earth) consumer nations in the world [1], and in addition approximately no production of RE, domestically. The supply country of heavy RE (terbium—Tb, dysprosium—Dy) is specifically localized in the world. Thus those are recognized as critical materials for high supply risk in the future.

By the way, PGM (platinum group metals) and other useful elements are created as FP (fission products) in the spent nuclear fuels. A new concept Adv.-ORIENT Cycle project defines these elements as "NRM (nuclear rare metals)", and aim at utilization of them [2]. In this project, heavy RE (Gd-Lu) has not been deeply estimated, because of their rather small fission yields [3]. On the contrary, BP (burnable poison) composed of gadolinium oxide (Gd_2O_3) is loaded in LWR (light water reactor) fuel with large amount in order to control excess reactivity at the beginning of reactor's operation cycle, and hence is supposed that, heavy RE elements must be produced abundantly by neutron capture of Gd followed by some β^- disintegrations as shown in Fig. 1. In this paper, the resource ability of heavy RE, Tb and Dy, by burnup of Gd_2O_3 BP has been studied calculatively to show quantitative and radiochemical qualitative properties of them for the utilization.

2. Calculation Method

The 9 × 9 lattice BWR (boiling water reactor) fuel assembly [4] and the 17 × 17 lattice PWR (pressurized water reactor) fuel assembly have been analyzed to evaluation of the rare earth production [5] as shown in Fig. 2. The void rate in the BWR fuel assembly assumed to be constant (40%) in axial. The BP formed of Gd_2O_3 is mixed homogeneously with several parts of UO_2 pellets in both fuel assemblies, which is the

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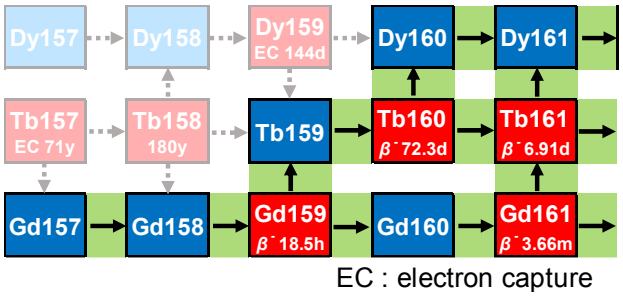


Fig. 1 Burnup process of BP (color online).

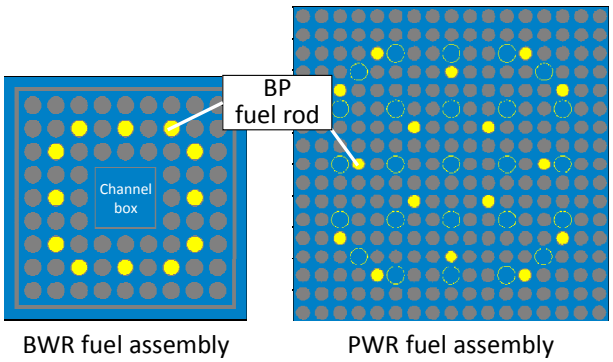


Fig. 2 BP fuel rods layout (color online).

Table 1 Specifications of BWR and PWR fuel assembly.

	BWR assembly	PWR assembly
Lattice	9 × 9	17 × 17
BP fuel rods	12 rods	16 rods
BP loading form	Gd_2O_3: 5wt.% Gd_2O_3: 6wt.% (mixed homogeneity with UO_2 pellet)	
Amount of Gd loading	1,392 (g/assembly)	1,751 (g/assembly)
Void rate	40% (flat)	-
Burnup	45 (GWd/tHM)	

Table 2 Natural abundance of Gd nuclides.

	Natural abundance (%)
^{152}Gd	0.20
^{154}Gd	2.18
^{155}Gd	14.80
^{156}Gd	20.47
^{157}Gd	15.65
^{158}Gd	24.84
^{160}Gd	21.86

ordinary loading method of the BP in the current LWRs fuel system. Table 1 lists the specifications of the BWR and PWR fuel assembly, and Table 2 presents a natural abundance of Gd nuclides.

A burnup calculation have been performed using a continuous energy 3-dimensional Monte Carlo code, MVP [6] with its burn-up calculation routine MVP-BURN [7] and JENDL-4.0 [8] library. A standard burnup chain for thermal reactor "u4cm6fp50bp16T" has been modified by using the corresponding parts of the more detailed burnup chain "th2cm6fp193bp6T" to evaluate production of Tb and Dy from Gd.

The radioactivity concentration of these elements have been evaluated the ORLIBJ40 package [9], which is a combination of ORIGEN2.2 code as an isotope generation and depletion calculation code and 1-group cross section library based on JENDL-4.0.

3. Results and Discussion

3.1 Production of Tb and Dy

Fig. 3 indicates two types of Tb production. One is the production of Tb produced by the (n, γ) reaction of Gd nuclides and successive β^- disintegration. The Tb produced in BP is noted as BP-Tb hereafter. Another is the production of Tb created as FP of fuel in the same BWR assembly (FP-Tb). Fig. 4 plots the production of Dy produced by the (n, γ) reaction of Gd and Tb nuclides and successive β^- disintegrations (BP-Dy), and that created as FP (FP-Dy) by the burnup of fuel in it.

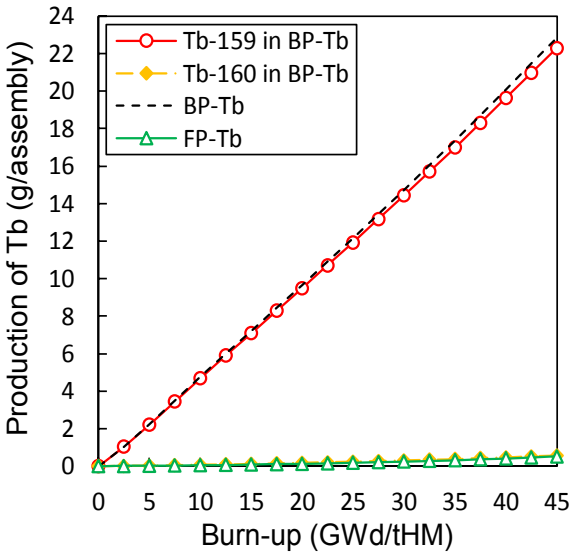


Fig. 3 Production of Tb in BP and fuel in the BWR assembly (color online).

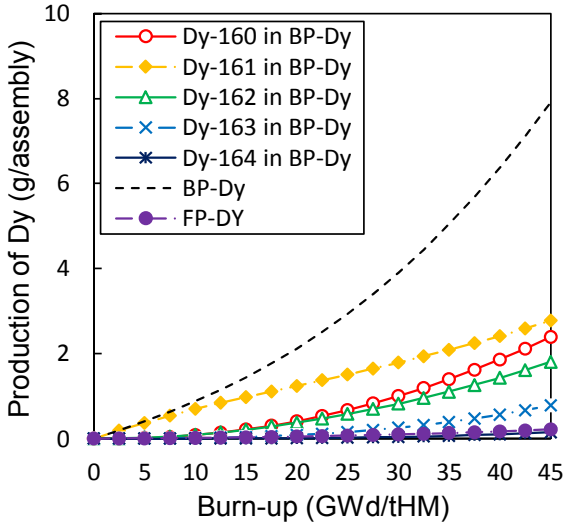


Fig. 4 Production of Dy same in above (color online).

The abundance of ^{159}Tb (stable) is 97% of total BP-Tb which is produced by (n, γ) reaction of ^{158}Gd followed by β^- decay disintegration.

$$^{158}\mathrm{Gd}\ (n,\ \gamma)^{159}\mathrm{Gd} \rightarrow \underline{^{159}\mathrm{Tb}} + \beta^-(18.5h)$$

On the other hand, the main nuclide of produced BP-Dy is ^{161}Dy, which is produced by (n, γ) reaction of ^{160}Gd with subsequent two β^- disintegrations, or (n, γ) reaction of ^{160}Dy.

$$^{160}\mathrm{Gd}\ (n,\ \gamma)^{161}\mathrm{Gd} \rightarrow {}^{160}\mathrm{Tb} + \beta^-(3.66m)$$

$$^{160}\mathrm{Tb} \rightarrow \underline{^{161}\mathrm{Dy}} + \beta^-(6.91d)$$

$$^{160}\mathrm{Dy}(n,\ \beta^-) \rightarrow \underline{^{161}\mathrm{Dy}}$$

Table 3 presents a summary of production of Tb and Dy at the end of burnup of the BWR and PWR assembly. In the BWR assembly, production of BP-Tb is 44.8 times larger than that of FP-Tb and, production of BP-Dy is also 38 times larger than that of FP-Dy.

This result is used to estimate the total production of Tb and Dy those can be produced potentially from operation of nuclear reactors in Japan under the following assumptions:

(1) Utilization factor of all reactors is 100%;

(2) All reactors are loaded with the fuel assemblies described above;

(3) Fuel exchange of all reactors is conducted by four batches scheme annually.

Fig. 5 shows annual and total production of Tb and Dy produced potentially from the operation of nuclear reactors in Japan from A.D. 1990 to 2010. Annual production of Tb is 146.44 (kg/y), and that of Dy is 146.44 (kg/y) as of A.D. 2010. In addition, total production of Tb from A.D. 1990 to 2010 is 2,739 (kg/y), and that of Dy is 1,059 (kg/y) as of A.D. 2010.

Table 3 Production of BP-Tb and BP-Dy.

	BWR assembly	PWR assembly
(a) Amount of Gd loading	1,392 (g/assembly)	1,751 (g/assembly)
(b) Production of BP-Tb	22.85 (g/assembly)	31.40 (g/assembly)
Production rate of BP-Tb (b)/(a)	1.64%	1.79%
(c) Production of BP-Dy	7.90 (g/assembly)	11.25 (g/assembly)
Production rate of BP-Dy (c)/(a)	0.56%	0.64%
Production of FP-Tb	0.51 (g/assembly)	1.60 (g/assembly)
Production of FP-Dy	0.22 (g/assembly)	0.70 (g/assembly)

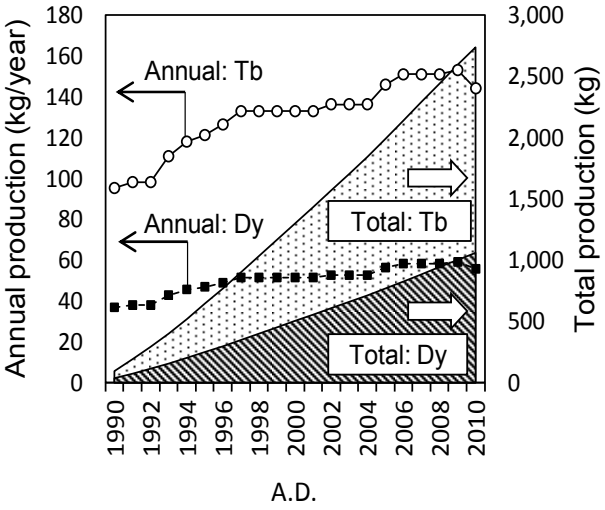


Fig. 5 Estimated production of Tb and Dy produced potentially from operation of nuclear reactors in Japan.

3.2 Radioactivity Concentration

Fig. 6 plots the radioactivity concentration of BP-Tb and FP-Tb in the BWR fuel assembly after the end of burnup. Fig. 7 shows the radioactivity concentration of BP-Dy and FP-Dy in BWR fuel assembly after the end of burnup. The decrease of radioactivity concentration of BP-Tb is faster than that

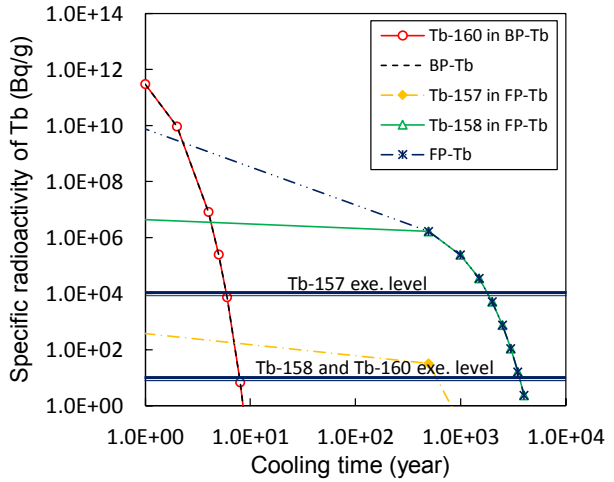


Fig. 6 Radioactivity concentration of Tb vs. cooling time (color online).

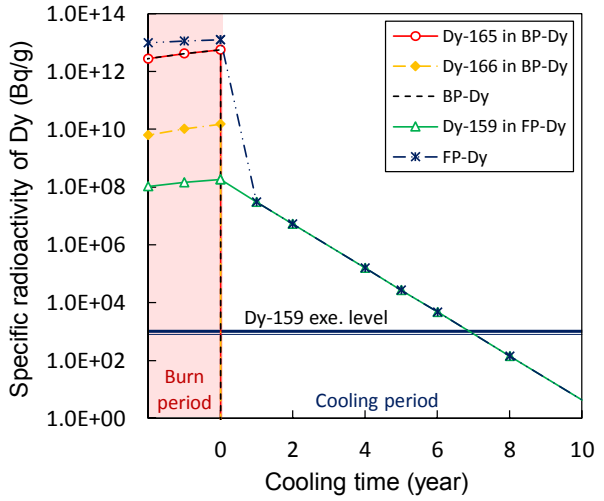
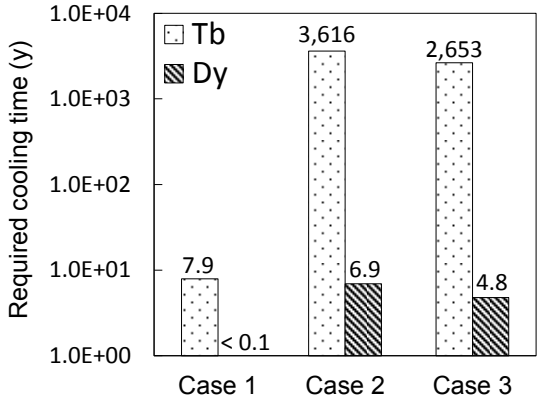


Fig. 7 Radioactivity concentration of Dy vs. cooling time (color online).

Table 4 Exemption level of Tb and Dy nuclides.

	Exemption level (IAEA basic safety standards) (Bq/g)
^{157}Tb	1.0E+04
^{158}Tb	1.0E+01
^{160}Tb	1.0E+01
^{159}Dy	1.0E+03

of FP-Tb because the radioactive nuclide ^{158}Tb, which is created as FP only, has a long life-time ($T_{1/2}$ = 180y). On the other hand, the decrease of radioactivity concentration of both BP-Dy and FP-Dy are relativity fast because the radioactive nuclides of Dy have short life-time regardless of create process. Table 4 shows the exemption level about the radioactivity



Case 1	Products derived from BP
Case 2	Products created as FP
Case 3	Products extracted in current fuel system

Fig. 8 Required cooling time of recovered Tb and Dy.

concentration regulated by IAEA (International Atomic Energy Agency) [10]. The required cooling time to achieve the exemption level is 7.9y for BP-Tb, while that time is 3,616y for FP-Tb. However, BP is mixed homogeneity with UO_2 pellet in the current nuclear fuel system as stated previously, thus the contamination of ^{158}Tb in FP-Tb cannot be avoided. Considering the current nuclear fuel system, the required cooling time of Tb is 2,653y as shown in Fig. 8. On the other hand, the required cooling time of Dy under the current nuclear fuel system is 4.9y. Then, Tb is not realistic, while Dy can be employed as the resource from the view point.

4. Conclusions

The burnup calculation has been performed to evaluate of resource ability of Tb and Dy, which produced by the burnup of Gd_2O_3 BP. The amount of Tb and Dy, transmuted by the (n, γ) reaction of Gd and its subsequent some β^- disintegrations are about 30-40 times larger than those created as FP. Required cooling time to achieve the exemption level of the radioactivity concentration of Tb and Dy recovered from the BP is much shorter (BP-Tb: 7.9y, BP-Dy: < 0.1y) than that created in FP (FP-Tb: 3,616y, FP-Dy: 6.9y). The BP is mixed homogeneity with UO_2 pellet in the current nuclear fuel system, where BP-Tb and

FP-Tb cannot be separated. Under the current nuclear fuel system, the required cooling time of Tb is 2,653y and that of Dy is 4.8y. Then, the use of Tb is not realistic but Dy can be used as the industrial from the view point of radioactivity concentration. From these results, it is concluded that, a new fuel system which separate BP from fuel is required to use recovered Tb as the industrial resource.

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