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ARTICLE

Analysis of Sample Worth for Dy_2O_3, Ho_2O_3, Er_2O_3 and Tm_2O_3 Measured at KUCA by MVP with Recent Version of ENDF and JENDL

Takanori KITADA[1,*], Kosuke SHIMOZATO[1], Koichi IEYAMA[1], Hidemasa OKOCHI[2] and Akio YAMAMOTO[2]

[1] *Osaka University, 2-1Yamada-oka, Suita-shi, Osaka, Japan, 565-0871*
[2] *Nagoya University, Furo-cho, Chikusaku, Nagoya, Japan, 464-8603*

Analyses were performed to validate nuclear data of some rare-earth elements by using the continuous energy Monte-Carlo code: MVP with recent version of ENDF and JENDL. The target of the analyses was the sample worth of Dy_2O_3, Ho_2O_3, Er_2O_3 and Tm_2O_3 measured at Kyoto University Critical Assembly (KUCA), and the worth was measured at two different cores named E3 (softer neutron spectrum) and EE1 (harder neutron spectrum) to evaluate the dependency of C/E values on neutron spectrum.

The cross section library such as JENDL-3.3 does not cover all of four elements, therefore the sample worth of Dy_2O_3 was analyzed with ENDF/B-VI.8, ENDF/B-VII.0 and JENDL-4.0, and that of Ho_2O_3 was analyzed with ENDF/B-VI.8 and ENDF/B-VII.0, and that of Er_2O_3 was analyzed with ENDF/B-VI.8, ENDF/B-VII.0, JENDL-3.3 and JENDL-4.0, and that of Tm_2O_3 was analyzed with JENDL-4.0.

The C/E values were within the range from 0.98 to 1.10 with relative standard deviation about 0.03. The large discrepancy of C/E value from 1.0 was not observed for all the results and this fact indicated the validity of cross section libraries of Dy, Er, Ho and Tm in ENDF/B-VI.8, ENDF/B-VII.0, JENDL-3.3 and JENDL-4.0. There was a small discrepancy observed in the case of Dy_2O_3 between E3 and EE1 cores and overestimation of Ho_2O_3 worth at both cores was observed. The analysis of JENDL-4.0 for Tm was also in good agreement with the measurements and the C/E values are around 1.0 for both E3 and EE1 cores.

KEYWORDS: rare-earth element, critical experiment, KUCA, ENDF, JENDL

I. Introduction

Utilization of high burnup fuel, which contains higher-enrichment uranium than a current fuel, is considered as one of the solution to reduce the fuel cycle cost and number of discharged fuel assemblies. Since the high burnup fuel has large excess reactivity at beginning of life, burnable poisons are used to reduce the excess reactivity at initial burnup. Characteristics of rare-earth elements are suitable for burnable poisons thus their application as burnable poison has been widely investigated. For example, gadolinia is commonly used in LWRs. Furthermore, in a context of development for over 5wt% enrichment fuel, the Er-SHB(Erbia-bearing Super High Burnup) fuel and the minimal-content gadolinia fuel are being studied as new concepts of a high burnup fuel in Japan.[1)] In this study, we focused on rare-earth elements as an advanced burnable poison for high burnup fuel.

Rare-earth elements consist of seventeen elements, scandium (Sc), yttrium (Y) and fifteen lanthanide elements, from lanthanum (La) to lutetium (Lu). Currently, gadolinium (Gd) has already used as burnable poison and erbium (Er) is recently spotlighted as a burnable poison for high burnup fuel as described in introduction. We can guess that many rare-earth elements would meet the following requirements as an advanced burnable poison for high burnup fuel:

1) Chemical affinity with nuclear fuel materials
2) More moderate capture cross section than Gd
3) Smaller reactivity suppression than Gd at BOC
4) Less residual reactivity

However, few (or no) critical experiment(s) has been carried out so far to validate the accuracy of their nuclear data and impacts on core characteristics. Therefore, critical experiments data loaded with rare earth elements will be invaluable and we will evaluate the adequacy of nuclear data of the rare-earth elements through comparison between measured data and numerical results.

This study attempts to estimate the adequacy of nuclear data and the impact of the rare-earth elements on core characteristics through critical experiments. Critical experiments using the rare-earth elements (Dy, Ho, Er and Tm), which are considered as a candidate of advanced burnable poison, were carried out at Kyoto University Critical Assembly (KUCA) from mid to late October in 2009. In this paper, we describe outline of the critical experiments with rare-earth elements at KUCA for the convenience sake.

The following Section II will describe the outline of the critical experiments. The experimental results will be shown in Section III. The calculation results obtained by the continuous energy Monte-Carlo code MVP will be compared in Section IV. In Section V, some conclusions will be provided.

*Corresponding author, E-mail:kitada@see.eng.osaka-u.ac.jp

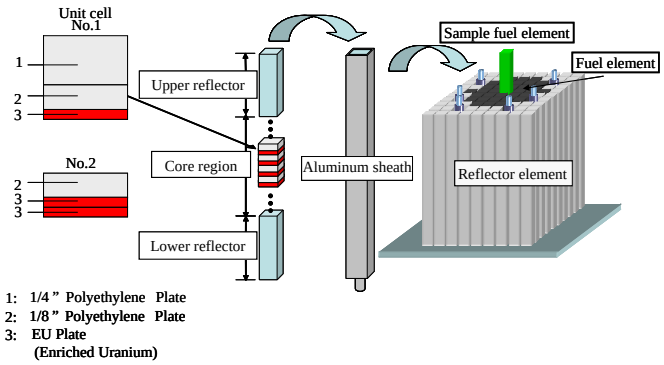

Fig. 1 A schematic view of solid moderated core (B core)

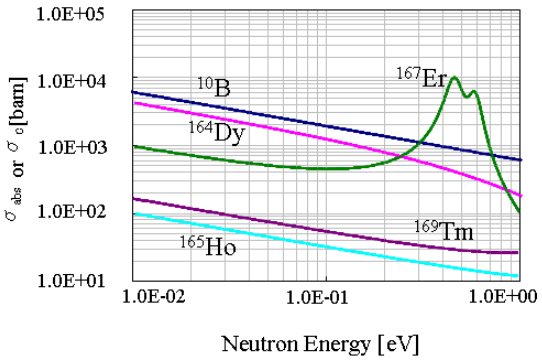

Fig. 2 Absorption cross section of ^{10}B, ^{164}Dy, ^{165}Ho, ^{167}Er and capture cross section of ^{169}Tm

II. Critical Experiment

1. Experimental Core

Kyoto University Critical Assembly (KUCA) consists of two solid moderated cores (A core and B core) and one light water moderated core (C core). In this critical experiment, a solid moderator core (B core) was used. A schematic view of the present critical experiment with the B core is shown in **Fig. 1**. A fuel element of the B core consists of combination with 1/16 inch thick 93wt% ^{235}U/U uranium-aluminum plate (EU), 1/4 and 1/8 inch thick polyethylene moderator plates (1/8"P, 1/4"P). These plates have 2x2 inch square shape. The combination of these plates is called "unit cell", whose typical thickness is 1-2 cm. A fuel element consists of stack of unit cells (core region is about 40 cm) and is sandwiched with the upper and lower polyethylene reflector blocks (about 50 cm each). The stacked unit cells and reflectors are inserted into an aluminum sheath tube. The B core is composed of the fuel elements and the reflector elements which are arranged on the periphery.

2. Treatment of Rare-Earth Element

In this experiment, the number of rare-earth elements to be measured was limited due to available machine time. To determine measured rare-earth elements, we firstly excluded the elements which have very small absorption cross section and have very large cross section (e.g. Gd). Therefore, we picked up following four elements: dysprosium (Dy), holmium (Ho), erbium (Er) and thulium(Tm). The atomic numbers of these elements are sequential and a burnup chain is formed by these elements. So it was considered that systematic data of these rare-earth elements would be expected. It is worth nothing that cross sections of Ho and Tm are small, but they were chosen since they are produced from the capture reaction of Dy and Er, respectively, which are considered as candidates of burnable absorber. Finally, we decided to measure the data of five elements: Dy, Ho, Er, Tm, and boron (B). Boron was used to estimate systematic error in experiments as a reference element.

Figure 2 shows neutron absorption or capture cross section of target elements in the thermal energy range (0.01 eV~1 eV). The absorption cross sections of ^{10}B, ^{164}Dy, ^{165}Ho and ^{167}Er, are based on ENDF/B-VI.8 and that of ^{169}Tm is based on the NG-ATLAS.

Fig. 3 The packing of rare-earth elements (left): the loading of Al sample case to sample fuel element (right)

3. Al Sample Case and Loading of Rare-Earth Elements

The rare-earth elements were used in the oxide form in the present experiments because they are chemically stable and the number density can be adjusted by diluting them into other material. Rare-earth elements and B oxide powders were diluted with alumina (Al_2O_3) as "samples" to suppress the spatial self-shielding effect of the elements, and their samples were packed into Al sample cases (50.8 mm × 50.8 mm × 10.0 mm) shown in **Fig. 3**. The Al sample case was inserted in the center of fuel element ("sample fuel element") shown in Fig. 3. The fuel element at the center of core was exchanged with the "sample case" in which rare-earth element was loaded.

4. Measurements of Reactivity Worth of Rare-Earth Elements

This experiment was carried out in two cores, 36 unit cells of 3/8"P EU (E3: B3/8"P36EU(3) core) and 60 unit cells of 1/8"P EU-EU(EE1: B1/8"P60EU-EU(5) core). The arrangement of the two cores is shown in **Fig. 4**. The "polyethylene reflector with void tube" and "partially loaded fuel element" is used to adjust the excess reactivity of a core to be approximately 0.25%Δk/k because the reactivity worth of the rare-earth sample was adjusted to approximately 0.15%Δk/k. These two cores were selected since these have different spectrum. The E3 core has soft spectrum and the EE1 core has hard spectrum. **Figure 5** shows the unit cell-averaged neutron spectrum of the two cores.

III. Results of the Experiment

In this experiment, sample worth of the rare-earth elements was evaluated from the difference of excess reactivity

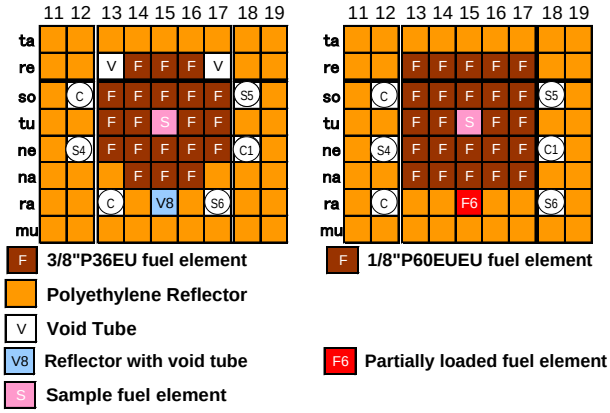

Fig. 4 Arrangement of the two cores(left- E3:B3/8”P36EU(3) core, right- EE1: B1/8”P60EU-EU(5) core)

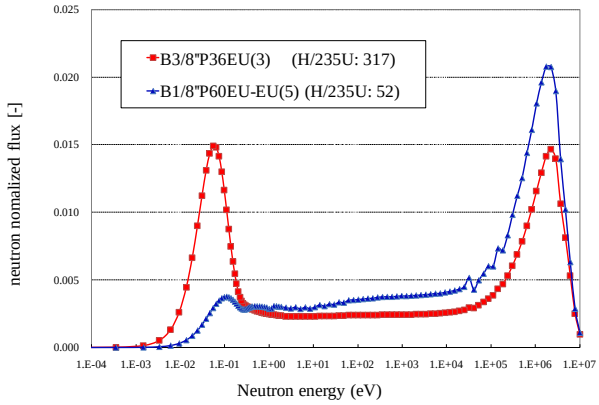

Fig. 5 Neutron spectrum of each unit cell

of the cores with and without the rare-earth sample, expressed as

$$\delta\rho = \rho - \rho', \quad (1)$$

where $\delta\rho$ represents the reactivity worth of a target element, and ρ is the excess reactivity without the target elements (only Al sample case) and ρ' is excess reactivity with the target element. The sample worth measurement was performed several times and the worth was obtained by averaging the results.

The rare-earth sample mass was determined through preliminary calculations by Monte-Carlo code MVP[4] to adjust the sample worth about 0.15%Δk/k. The rare-earth sample in the sample case is diluted by alumina (Al_2O_3), to mitigate the spatial self-shielding effect of the rare-earth sample since Al_2O_3 has small impact on the excess reactivity.

The excess reactivity of each core was measured by the period method. Firstly, we draw the reactivity curve on the C1 control rod in Fig. 4 by repeating the excess reactivity measurement. Then the excess reactivity of a core loaded with or without rare-earth sample was evaluated by the position of C1 at critical point.[5] The effective delayed neutron fraction and the prompt neutron life time evaluated by the deterministic code SRAC[6] were used to evaluate the reactivity in %Δk/k.

Table 1 Sample worth of the rare-earth elements

Core Type		Dy_2O_3	Ho_2O_3	Er_2O_3	Tm_2O_3
E3 core	mass(g)	0.95	3.50	9.00	6.00
	sample worth (%⊿k/k)	0.174 [4]	0.160 [4]	0.184 [4]	0.171 [4]
	measurement error (%⊿k/k)	±0.004	± 0.003	± 0.002	± 0.004
EE1 core	mass(g)	5.50	23.50	14.00	22.00
	sample worth (%⊿k/k)	0.156 [5]	0.123 [5]	0.160 [7]	0.147 [5]
	measurement error (%⊿k/k)	± 0.001	± 0.001	± 0.002	± 0.002

Table 2 Analysis results of rare-earth sample worth (E3: B3/8”P36EU (3) core)

element	C/E value			
	JENDL-4.0	JENDL-3.3	ENDF/B-VII.0	ENDF/B-VI.8
Dy_2O_3	1.053 (±0.029)	-	1.074 (±0.029)	1.100 (±0.032)
Ho_2O_3	-	-	1.053 (±0.025)	1.094 (±0.028)
Er_2O_3	1.041 (±0.019)	1.017 (±0.019)	1.023 (±0.019)	1.024 (±0.022)
Tm_2O_3	1.005 (±0.021)	-	-	-

Table 3 Analysis results of rare-earth sample worth (EE1: B1/8”P60EU-EU (5) core)

element	C/E value			
	JENDL-4.0	JENDL-3.3	ENDF/B-VII.0	ENDF/B-VI.8
Dy_2O_3	1.027 (±0.019)	-	1.018 (±0.019)	1.028 (±0.024)
Ho_2O_3	-	-	1.028 (±0.025)	1.076 (±0.030)
Er_2O_3	1.004 (±0.020)	0.989 (±0.020)	0.972 (±0.020)	0.965 (±0.026)
Tm_2O_3	0.991 (±0.022)	-	-	-

Table 1 shows the mass of each element in the sample case and the average of sample worth on two cores. Ho_2O_3 for EE1 core was not able to be packed up to the expected mass due to the limitation of the space in the Al sample case. The number in parentheses is the measurement frequency. The measurement error in Table 1 is one standard deviation of measured sample worth. The reactivity worth was calculated by Eq. (1).

IV. Comparison of Calculation and Experimental Results

Calculation was performed by the continuous energy Monte Carlo code MVP with rigorous treatment of the core geometries and material compositions as much as possible. To evaluate the reactivity worth, two independent Monte Carlo calculations with 2,500 histories were performed. One is the case with the target elements, and another is the case without the target element without all control rods. The calculated two k_{eff}’s are used to evaluate the worth like as Eq. (1). The cross section libraries used in the MVP analyses were ENDF/B-VI.8, ENDF/B-VII.0, JENDL-3.3 and JENDL-4.0. We evaluated the sample worth of rare-earth by using these cross section libraries for the comparison. Analysis results of E3: B3/8”P36EU(3) core are shown in **Table 2** and **Fig. 6**. Analysis results of EE1: B1/8”P60EU-EU(5) core are shown in **Table 3** and **Fig. 7**.

The numbers in parentheses in Tables 2 and 3 and the er-

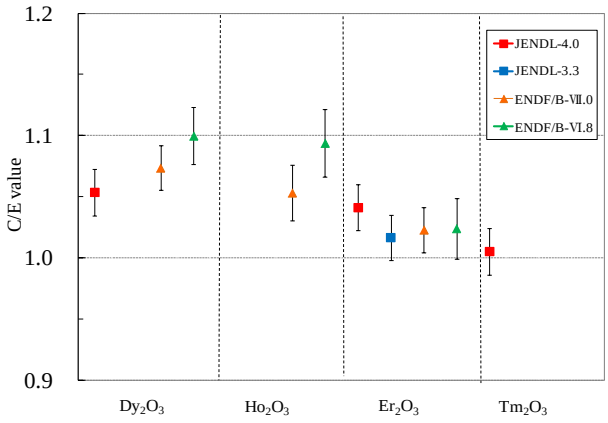

Fig. 6 C/E value of reactivity worth in MVP calculation (E3: B3/8"P36EU(3) core)

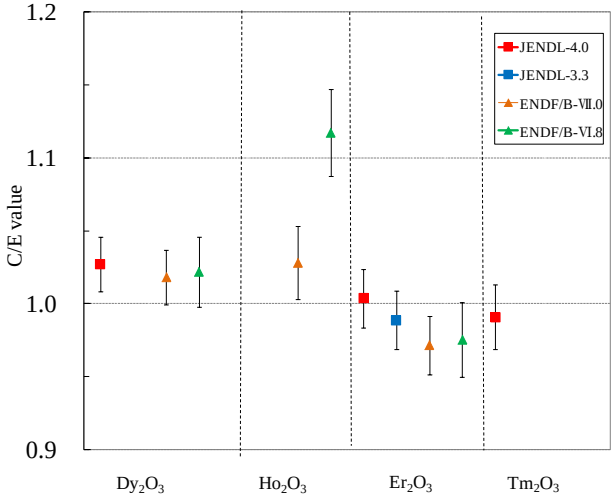

Fig. 7 C/E value of reactivity worth in MVP calculation (EE1: B1/8"P60EU-EU(5) core)

ror bar in Figs. 6 and 7 represent absolute error of C/E value from measurement error and MVP calculation error.

As shown in Figs. 6 and 7, all results of C/E value are roughly around 1.0 (from 0.98 to 1.10) by considering a standard deviation of about 0.03, which is evaluated from the experimental errors and the statistical error of MVP calculation.

Remarkable difference in C/E value among cross section libraries could not be observed for all elements and cores. However there is a spectrum dependency on C/E value of Dy_2O_3, the value at E3 is more than 1.0 although the value at EE1 is around 1.0. The C/E value of Ho_2O_3 is slightly more than 1.0 at both cores and this shows the overestimation of Ho_2O_3 worth calculated with ENDF/B-VI.8 and ENDF/B-VII.0.

However, the experimental errors are relatively large to discuss further difference, therefore we are planning the additional experiments to reduce the experimental errors.

V. Conclusions

We have measured the reactivity worth of the rare-earth elements at Kyoto University Critical Assembly (KUCA). The measured elements are Dy, Ho, Er and Tm by packing them into an Al sample case with Al_2O_3 as diluting material to decrease the spatial self-shielding effect of the element. Two different cores, which have different neutron spectra, were assembled to estimate the dependency of the sample worth on neutron spectrum.

The large discrepancy of C/E value from 1.0 is not observed for all the results and this fact shows that the cross section data of Dy, Er, Ho and Tm in the libraries (ENDF/B-VI.8, ENDF/B-VII.0, JENDL-3.3 and JENDL-4.0) has a good reliability to evaluate reactivity worth of those elements. There is a small discrepancy observed in the case of Dy_2O_3 between E3 and EE1 cores and overestimation of Ho_2O_3 worth at both cores is expected.

The experimental errors are relatively large compared to the difference among the results, therefore further experiment is planning to reduce the errors for more detailed discussion about the difference.

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