# ARTICLE

# Integral Experiment on Beryllium with DD Neutrons for Nuclear Data Benchmarking

Keitaro KONDO<sup>1\*</sup>, Kentaro OCHIAI<sup>1</sup>, Yosuke TATEBE<sup>1</sup>, Takahiro YAGI<sup>2</sup>, Seiki OHNISHI<sup>1</sup>, Kosuke TAKAKURA<sup>1</sup>, Satoshi SATO<sup>1</sup>, and Chikara KONNO<sup>1</sup>

<sup>1</sup>Fusion Research and Development Directorate, Japan Atomic Energy Agency, Shirakata shirane 2-4, Tokai-mura, Naka-gun, Ibaraki 319-1195, Japan <sup>2</sup>Graduate School of Energy Science, Kyoto University, Asashironishi 2-1010, Kumatori-cho, Sennan-gun, Osaka 590-0494, Japan

An integral experiment on beryllium with DD neutrons was conducted for nuclear data benchmarking at the FNS facility of JAEA. The reaction rate distributions of <sup>115</sup>In(n,n')<sup>115m</sup>In, <sup>197</sup>Au(n, $\gamma$ )<sup>198</sup>Au, <sup>6</sup>Li(n, $\alpha$ )T and <sup>235</sup>U(n,fission) were measured inside a beryllium cylindrical assembly. The measured reaction rates were compared with the calculated values with MCNP and the latest nuclear data libraries; JENDL-3.3, ENDF/B-VII.0 and JEFF-3.1. A systematic difference was found between the C/E values of the <sup>115</sup>In(n,n')<sup>115m</sup>In reaction rate with JENDL-3.3 and with the other libraries. The angular-differential cross-section data of the elastic scattering in ENDF/B-VII.0 and JEFF-3.1 are probably inadequate. JENDL-3.3 is considered to overestimate the (n,2n) cross-section at 2.6 - 3 MeV. The calculated reaction rates for the <sup>6</sup>Li(n, $\alpha$ )T and <sup>235</sup>U(n,fission) reactions, which are sensitive to low energy neutrons, showed a large overestimation. This tendency is similar to that in the previous integral experiment on beryllium with DT neutrons conducted at FNS, but its reasons are not specified yet.

KEYWORDS: DD neutron, nuclear data, beryllium, benchmark experiment, FNS

#### I. Introduction

At the Fusion Neutronics Source (FNS) facility of JAEA, which is an intense DT neutron source for fusion neutronics studies, various integral experiments with DT neutrons for fusion reactor candidate materials have been carried out and have made a significant progress in the verification of nuclear data.<sup>1)</sup> Recently we started a new series of integral experiments with DD neutrons at FNS in order to verify nuclear data relating to DD neutrons effectively. We chose beryllium as the first material for the integral experiment with DD neutrons because of its importance as a candidate material of a fusion reactor. Several problems of nuclear data evaluated for beryllium have been pointed out in the previous benchmark experiment<sup>2)</sup> with DT neutrons. We expect that new information for verification of beryllium nuclear data would be acquired through the present integral experiment.

## **II. Experiment**

## 1. Neutron Source

The present experiment was conducted at the Fusion Neutronics Source (FNS) facility of the Japan Atomic Energy Agency (JAEA). The 80-degree line with a water-cooled fixed target was used in the present study. DD neutrons were generated by bombarding a deuteron beam of 350 keV to a deuterized titanium target. The specification of the DD neutron source was investigated based on our calculation and experiment with the activation foil method.<sup>3)</sup>

The maximum energy of produced neutrons at 0 deg. to the beam axis was 3.3 MeV. The neutron yield from the  $D(d,n)^{3}$ He reaction was determined by detecting protons from the competitive reaction, i.e., D(d,p)T reaction with small silicon solid state detectors. A typical neutron yield was approximately  $6 \times 10^{9}$ /s with the beam current of 2 mA.

## 2. Beryllium Assembly

Several types of beryllium blocks, which are standard grade beryllium blocks, were used in the present study. The density of beryllium blocks is 1.822 - 1.848 g/cm<sup>3</sup> and almost the same as the theoretical one, i.e., 1.848 g/cm<sup>3.4</sup>) A cylindrical slab (like a thick cylinder) assembly was built at the distance of 20 cm from the DD neutron source with these blocks and aluminum thin support frames as shown in **Fig. 1**. The size of the assembly was 63.0 cm in equivalent diameter and 45.7 cm in thickness.



Fig. 1 Schematic view of the experimental arrangement

<sup>\*</sup>Corresponding Author, E-mail: kondo.keitaro@jaea.go.jp

<sup>©</sup> Atomic Energy Society of Japan

## 3. Dosimetry Detectors

Several reaction rate distributions were measured along the central axis of the assembly with the following three methods:

# (1) Activation Foils

The <sup>115</sup>In(n,n')<sup>115m</sup>In and <sup>197</sup>Au(n, $\gamma$ )<sup>198</sup>Au reaction rate distributions were measured with the activation foil method. The In foils used were 10 mm square and 1 mm in thickness. The Au foils used were 10 mm square and 1  $\mu$ m in thickness. Most of the In foils were covered with Cd foils of 0.5 mm in thickness in order to reduce thermal neutrons and measured after cooling for several hours, because <sup>116</sup>In produced via (n, $\gamma$ ) reaction emits a large number of gamma-rays disturbing the measurement.

## (2) Li<sub>2</sub>CO<sub>3</sub> Pellets

The <sup>6</sup>Li(n, $\alpha$ )T reaction rate distribution was measured with <sup>6</sup>Li-enriched (95.5 atom%) Li<sub>2</sub>CO<sub>3</sub> pellet disks of 13 mm in diameter and 1 mm in thickness. These pellets were placed inside the assembly. After the irradiation, the pellets were dissolved with the binary-acid method<sup>5</sup> and samples of the pellet solution with a scintillation cocktail were prepared. The produced activities of tritium in the samples were measured with a liquid scintillation counter, and the reaction rates were deduced.

#### (3) Micro-fission Chamber

The  $^{235}$ U(n,fission) reaction rate distribution was measured with a micro-fission chamber coated with about 4 mg of  $^{235}$ U oxides, the size of which was 6.25 mm in outside diameter and 25.4 mm in active length. The chamber was inserted in beryllium blocks with a central hole of 21 mm in diameter and moved along the central axis inside the assembly. The space forward of the detector was filled with usual blocks. The absolute fission rate was obtained with a measured count and an effective  $^{235}$ U atom number determined in the previous experiment.<sup>6</sup>

The <sup>115</sup>In(n,n')<sup>115m</sup>In reaction is sensitive to neutrons above around 0.5 MeV because of its threshold energy of 0.34 MeV and thus indicator of the validity of nuclear data in the MeV region. The <sup>197</sup>Au(n, $\gamma$ )<sup>198</sup>Au, <sup>6</sup>Li(n, $\alpha$ )T and <sup>235</sup>U(n,fission) reactions are sensitive to low energy neutrons and affected both by nuclear data of the whole energy range and by thermal neutron scattering law data, namely S( $\alpha$ , $\beta$ ) data.

## **III. Analysis**

The reaction rates inside the assembly were calculated with the MCNP5  $code^{7}$  and the latest nuclear data libraries: JENDL-3.3<sup>8</sup>, ENDF/B-VII.0<sup>9</sup> and JEFF-3.1<sup>10</sup>. In the calculation, the experimental assembly, activation foils and Li<sub>2</sub>CO<sub>3</sub> pellets were precisely modeled and their reaction rates were obtained using the track length tally (F4) function of MCNP with JENDL Dosimetry File 99<sup>11</sup>. The source

term of the calculation is described in Ref. 3. The SAB2002 library<sup>12)</sup> was used in the calculation as  $S(\alpha,\beta)$  data of beryllium metal.

# **IV. Results and Discussion**

# 1. <sup>115</sup>In(n,n')<sup>115m</sup>In Reaction Rate

**Figure 2** shows the measured and calculated reaction rate distributions of <sup>115</sup>In(n,n')<sup>115m</sup>In, where the measured value was obtained up to 23-cm depth inside the assembly. **Figure 3** shows calculated to experimental ratios (C/E) of the <sup>115</sup>In(n,n')<sup>115m</sup>In reaction rate. The calculation with JENDL-3.3 underestimates the measured value by approximately 5%. The calculations with ENDF/B-VII.0 and JEFF-3.1 show rather good agreement with the experiment within the experimental error, but an underestimation is obvious on the surface. There is a systematic difference of around 10% between the C/E values with JENDL-3.3 and with the other libraries, ENDF/B-VII.0 and JEFF-3.1.

#### 2. Reaction Rates for Low Energy Neutrons

**Figure 4** shows the measured and calculated reaction rate distributions of the  ${}^{197}Au(n,\gamma){}^{198}Au$ ,  ${}^{6}Li(n,\alpha)T$  and  ${}^{235}U(n,fission)$  reactions, all of which are sensitive to low energy neutrons. The differences among the calculated



Fig. 2 Reaction rate distributions of  ${}^{115}In(n,n'){}^{115m}In$ 



**Fig. 3** C/E of the  ${}^{115}$ In(n,n') ${}^{115m}$ In reaction rate

reaction rates with the three nuclear data libraries were around 5% for all the three reactions, where JENDL-3.3 systematically gave the largest values and ENDF/B-VII.0 the smallest. **Figure 5** shows the C/Es of these reactions with JENDL-3.3. The reaction rates calculated for the <sup>6</sup>Li(n, $\alpha$ )T and <sup>235</sup>U(n,fission) reactions show a very large overestimation; 30% for <sup>6</sup>Li and 60% for <sup>235</sup>U. This tendency is similar to that in the beryllium integral experiment<sup>2)</sup> with DT neutrons previously carried out at FNS.



Fig. 4 Reaction rate distributions of  ${}^{197}Au(n,\gamma){}^{198}Au$ ,  ${}^{6}Li(n,\alpha)T$  and  ${}^{235}U(n,fission)$ 



3. Reason of the Discrepancy in the Calculation

In order to find the reason of the above discrepancy appeared in the C/E values, we investigated differences

among nuclear data for beryllium evaluated in the libraries and their impact on the calculated C/E value. Several modified libraries were made by exchanging a part of the evaluated libraries reaction by reaction. ACE files of the modified libraries for MCNP were produced with the NJOY99.259 code<sup>13</sup>.

First, we found there was a systematic difference between JENDL-3.3 and the other libraries in the evaluation of the angular-differential cross-section (ADX) of the elastic scattering at 2.7 - 3.4 MeV. **Figure 6** shows a typical ADX

of the elastic scattering in these libraries with the experimental data measured by Baba et al.<sup>14)</sup> When the ADX in JEFF-3.1 is replaced by that in JENDL-3.3, the C/E tendency of the <sup>115</sup>In(n,n')<sup>115m</sup>In reaction rate changes by approximately 10% as shown in **Fig. 7**. As for the ADX in ENDF/B-VII.0, the same trend appears. The C/E value on the surface becomes better when the ADX in JENDL-3.3 is used. Thus, the ADX of the elastic scattering at this energy region of JENDL-3.3 would be better than those of ENDF/B-VII.0 and JEFF-3.1.

Second, a difference of the (n,2n) reaction cross-section exists near its threshold energy of around 3 MeV between the libraries as shown in Fig. 8, which includes the experimental data measured by Holmberg et al.<sup>15)</sup> When the (n,2n) cross-section data in JENDL-3.3 are replaced by those in the other libraries, the C/E values of the <sup>115</sup>In(n,n')<sup>115m</sup>In reaction rate become closer to 1 as shown in Fig. 9. The present result suggests that JENDL-3.3 would overestimate the (n,2n) cross-section at 2.6 - 3 MeV. As noted in the former section, JENDL-3.3 tends to bring the larger reaction rates for low energy neutrons. If the (n,2n)reaction cross section data of JENDL-3.3 were replaced by those in the other libraries, all of the calculated reaction rates for low energy neutrons also decreased by 5%. However, the large overestimation of the  ${}^{6}Li(n,\alpha)T$  and <sup>235</sup>U(n,fission) reaction rates cannot be explained from the



Fig. 6 ADX of elastic scattering of beryllium at 3.00 MeV



**Fig. 7** Comparison of the C/E values for the <sup>115</sup>In(n,n')<sup>115m</sup>In reaction rate with modified nuclear data libraries, in which the ADX of elastic scattering is replaced by that in JENDL-3.3



Fig. 8 The (n,2n) reaction cross-section at the DD-neutron energy region



Fig. 9 Comparison of the C/E values for the  $^{115}In(n,n')^{115m}In$  reaction rate with modified JENDL-3.3 libraries, in which the (n,2n) cross-section is replaced by that in the other libraries.

difference. Although there is no difference among the libraries, its origin might be in nuclear data of the lower energy region. Also thermal neutron scattering law data can affect the result, thus further examination for these data should be required.

#### V. Conclusion

The integral experiment with the DD neutron source we have started at FNS is a powerful tool to verify the accuracy of nuclear data induced with DD neutrons. In the integral experiment on beryllium, we found a systematic difference of around 10% between the C/E values of the <sup>115</sup>In(n,n')<sup>115m</sup>In reaction rate with JENDL-3.3 and with ENDF/B-VII.0 and JEFF-3.1. The ADX of the elastic scattering at the DD-neutron energy region significantly affects the calculated  $^{115}\text{In}(n,n')^{115m}\text{In}$  reaction rate, and the ADX data in ENDF/B-VII.0 and JEFF-3.1 are probably not good. JENDL-3.3 is considered to overestimate the (n,2n)cross-section at 2.6 - 3 MeV. The calculated reaction rates for the <sup>6</sup>Li( $n,\alpha$ )T and <sup>235</sup>U(n,fission) reactions showed a large overestimation. We will try to find reasons for the overestimation by comprehensively analyzing the

experiments with DT and DD neutrons at FNS.

## Acknowledgment

The authors wish to acknowledge the FNS staff for their excellent operation of the FNS accelerator: Messrs. C. Kutsukake, S. Tanaka, Y. Abe, Y. Oginuma and M. Kawabe.

#### References

- F. Maekawa, C. Konno, Y. Kasugai, et al., "Data collection of fusion neutronics benchmark experiment conducted at FNS/JAEA," JAERI-Data/Code 98-021, Japan Atomic Energy Research Institute, (1998).
- C. Konno, K. Ochiai, M. Wada, S. Sato, "Analyses of fusion integral benchmark experiments at JAEA/FNS with FENDL-2.1 and other recent nuclear data libraries," Fusion Eng. Des., 83, 1774 (2008).
- K. Kondo, K. Ochiai, C. Kutsukake, C. Konno, "Characterization of the DD-neutron source for the 80 degrees beam line of the Fusion Neutronics Source (FNS)," JAEA-Technology 2008-088, Japan Atomic Energy Agency, (2009).
- Y. M. Verzilov, K. Ochiai, S. Sato, et al., "Analysis of impurities in beryllium affecting evaluation of the tritium breeding ratio," JAERI-Research 2004-005, Japan Atomic Energy Research Institute, (2004).
- Y. Verzilov, F. Maekawa, Y. Oyama, "A novel method for solving lithium carbonate pellet by binary-acid for tritium production rate measurement by liquid scintillation counting technique," J. Nucl. Sci. Technol., 33, 390 (1996).
- H. Maekawa, Y. Ikeda, Y. Oyama, et al., "Benchmark experiments on a 60 cm-thick graphite cylindrical assembly," JAERI-M 88-034, Japan Atomic Energy Research Institute, (1988).
- F.B. Brown, R.F. Barrett, T.E. Booth, et al., "MCNP version 5," Trans. Am. Nucl. Soc., 87, 273 (2004).
- K. Shibata T. Kawano, T. Nakagawa, et al., "Japanese Evaluated Nuclear Data Library Version 3 Revision-3: JENDL-3.3," J. Nucl. Sci. Technol., 39, 1125 (2002).
- M. Chadwick, P. Oblozinsky, M. Herman, et al., "ENDF/B-VII.
  0: Next generation evaluated nuclear data library for nuclear science and technology," Nuclear Data Sheets, 107, 2931 (2006).
- A. J. Koning, R. Forrest, M. Kellett, et al., "The JEFF-3.1 nuclear data library," JEFF Report 21, OECD Nuclear Energy Agency, (2006).
- K. Kobayashi, T. Iguchi, S. Iwasaki, et al., "JENDL Dosimetry File 99 (JENDL/D-99)," JAERI 1344, Japan Atomic Energy Research Institute, (2002).
- R. C. Little, R. E. MacFarlane, "SAB2002—An S(a,b) library for MCNP," LA-UR-03-808, Los Alamos National Laboratory, (2003).
- R.E. MacFarlane, D.W. Muir, "The NJOY nuclear data processing system Version 91," LA-12740-M, Los Alamos National Laboratory, (1994).
- 14) M. Baba, T. Sakase, T. Nishitani, et al., "The interactions of fast neutrons with <sup>9</sup>Be," Proc. Int. Conf. Neutron Phys. Nucl. Data, Harwell, UK, Sep. 25-29, 1978, 198 (1978).
- 15) M. Holmberg, J. Hansén, "The (n, 2n) cross section of <sup>9</sup>Be in the energy region 2.0–6.4 MeV," Nucl. Phys. A, 129, 305 (1969).