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The integral experiment on beryllium with D-T neutrons for verification of tritium breeding

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Abstract

A clean benchmark experiment on beryllium was performed with D-T neutrons at the FNS facility of the Japan Atomic Energy Agency. The main objective was to verify the integral data related to the tritium production on lithium isotopes. Tritium production rates, as well as activation reaction rates were measured inside the beryllium assembly that was shaped as a pseudo-cylindrical slab with an area-equivalent diameter of 628 mm and a thickness of 355 mm. Experimental results were analyzed with a three-dimensional Monte Carlo transport code MCNP-4C and FENDL/MC-2.0, JENDL-3.2/3.3 neutron transport libraries. Evaluation of reaction rates was based on the cross section data taken from the JENDL Dosimetry File and ENDF B-VI data libraries. Analysis shows that all calculation combinations (transport and activation cross section libraries) used for evaluation of reaction rates give data that is agreeable with measured values within 10%.

Keywords: Tritium production rate; Integral experiment; Beryllium; Data testing

1. Introduction

The necessity of tritium breeding for self-sufficiency of the D-T fusion reactor requires development of a blanket with an appropriate tritium breeding potential. In order to meet self-sufficiency requirements, many conceptual blanket designs involve the use of beryllium as the most vital material, due to a number of exceptional properties. From the neutronics point of view, beryllium is an excellent neutron multiplicator and a moderator with an extremely low absorption cross section for thermal neutrons. To assure that nuclear design calculations of the blanket are reliable, clean and design-oriented integral experiments, i.e. experiments on the bulk beryllium assembly and blanket mock-ups have to be completed.

Numerous clean experiments on beryllium with D-T neutrons have been performed with the main objective of investigating neutron multiplication issues

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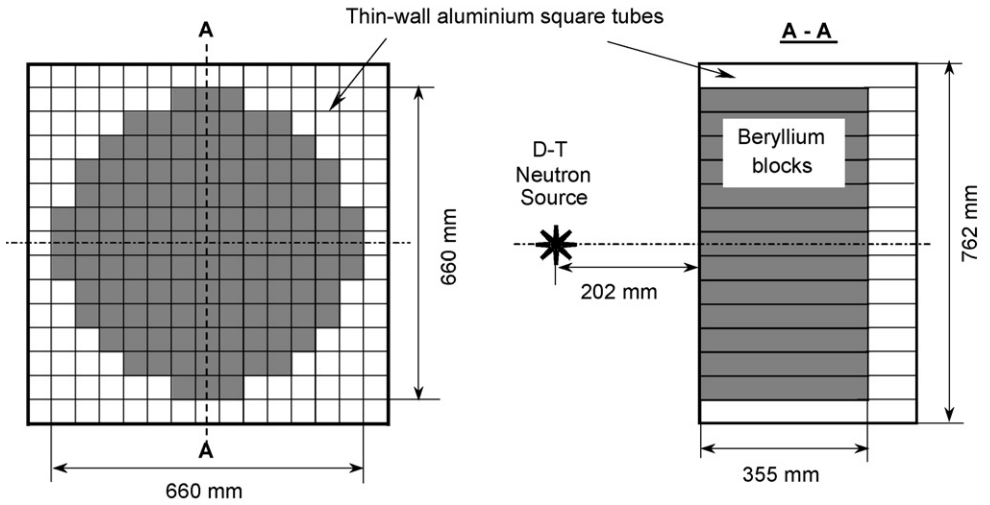

Fig. 1. Configuration of the experiment on the beryllium assembly irradiated by neutrons from the D-T source.

(overview is given in Ref. [1]). Considerably less attention was paid to tritium breeding. At the moment, it is realized that the beryllium neutron multiplication power can be suitably predicted in design calculations of breeder blankets [1]. However, observed discrepancies between calculations and experiments for the spectral neutron flux distribution [2] can affect the tritium breeding ratio. This is especially important for the present design of the solid breeder blanket of the DEMO reactor [3], since the resulting low energy neutron flux spectrum for tritium breeding via $^{6}Li(n,\alpha)^{3}H$ reaction is mostly formed by beryllium. In addition, large overestimations (20–50%) for reaction rates that are sensitive to low energy neutrons, were observed in previous experiments [4,5] on the bulk beryllium assembly. Due to importance of low energy neutrons for tritium breeding, overestimations must be examined. Parasitic absorption of thermal neutrons, due to unspecified impurities in beryllium, and a non-adequate description of the neutron thermalization process as well as the experimental model presumably have an impact on overestimations. In such circumstances, in order to fulfill the necessity for experimental verification of tritium breeding potential, the integral experiment on bulk beryllium assembly was performed. The key experimental objective was:

- to verify the accuracy of estimating the tritium production rate on lithium isotopes in the bulk beryllium assembly.

Prior to the experiment, an additional study concerning impurities in beryllium blocks used for assembly construction was conducted with sample and integral methods [6]. As compared with previous experiments on beryllium [4,5], beryllium composition data, experimental model description, analysis and measurement techniques were significantly improved in the present study.

2. Experiment

The integral experiment has been performed at the Fusion Neutronics Source (FNS) facility in Japan Atomic Energy Agency (JAEA). The experiment was conducted in an open geometry with a D-T point source. The facility provided good experimental conditions in regard to the neutron background, due to the large size of the target room (15 m × 15 m × 12 m). The beryllium assembly was located in front of the neutron source at a distance of 202 mm from the target, and placed inside a framework constructed with thin-wall aluminum square tubes, Fig. 1. The minimal distance between the assembly and room walls was about 6 m. In order to produce D-T neutrons, a target with a tritium activity of approximately 3.7×10^{11} Bq was bombarded by a deuteron beam with an energy of 350 keV and an intensity of 1 mA at the target. The neutron yield was determined by the associated alpha-

Table 1
Chemical composition of the S-200-F standard grade beryllium

Element	Content		Element	Content	
	ICP–MS[a]	BrushW[b]		ICP–MS[a]	BrushW[b]
Be (%)	97.9 ± 0.8	98.82	Ni (ppm)	250 ± 30	–
Li (ppm)	<1	–	Zr	S	–
B (ppm)	<3	–	Nb	S	–
C (ppm)	–	840	Mo	S	–
Mg (ppm)	110 ± 5	–	Cd (ppm)	<1	–
Al (ppm)	570 ± 50	710	Dy	S	–
Sc	S	–	W	S	–
Ti	S	–	Hg	S	–
V	S	–	Pb	S	–
Mn (ppm)	96 ± 5	–	Th (ppm)	1.5 ± 0.1	–
Fe (ppm)	1300 ± 70	1250	U (ppm)	82 ± 3	–
Co	S	–	Other max	–	400

[a] Content measured with the ICP–MS method.
[b] Data obtained from the manufacturing company, Brush Wellman Inc.

particle detector [7] with an accuracy of 3%. Neutron spectrum of the D-T source was comprehensively evaluated during research activities [8]. Irradiation of the assembly was performed for 3 days, 7 h/day, with a total neutron yield of 6.96×10^{15} neutrons. The activation technique has been used in the experiment in order to obtain the neutron spectrum indices for verification of transport calculations. Pellets and foils were inserted into the assembly during construction. After irradiation, pellets and foils were taken out of the assembly for measurement with appropriate techniques.

2.1. Beryllium assembly

An experimental assembly was constructed from beryllium blocks (S-200-F, Brush Wellman Inc., USA) and shaped as a pseudo-cylindrical slab with an area-equivalent diameter of 628 mm and a thickness of 355 mm, Fig. 1. The blocks were produced through consolidating beryllium powder by vacuum hot pressing, and afterwards machining the blocks to dimensions of 5.08 cm × 5.08 cm and a length of 2.54, 5.08 and 10.16 cm. Average density equals to 1.84 g/cm^3. Special care was paid to chemical composition of beryllium, because the composition may contain elements, which can affect the distribution of thermal neutrons, due to the parasitic absorption of thermal neutrons. Sample and integral analyses were completed in order to gain impurity information [6]. Sample analysis was completed by the ICP–MS method, and integral analysis—by the neutron pulsed method. The integral analysis was performed for all beryllium blocks used in the experiment. Results obtained in these analyses are in a reasonably good agreement. Detailed chemical composition of the beryllium sample is presented in Table 1. Some of the elements were detected, though their quantities were not measured absolutely (such elements are indicated by symbol "S"). It has been experimentally proven by the neutron pulsed method [6], that effective absorption of thermal neutrons in beryllium blocks used for assembly construction is approximately 30% higher than the calculated value, based on data specified by the manufacturing company.

2.2. Reaction rate measurements

Taking into account the experimental objective and the existing integral data obtained in the previous experiments [4,5] on beryllium, an appropriate set of nuclear reactions was selected (Table 2). The ^{93}Nb(n,2n)^{92m}Nb was selected to measure the distribution of high energy neutrons from the source. The ^{32}S(n,p)^{32}P and ^{7}Li(n,n′α)^{3}H reactions were selected, due to their sensitivity to fast neutrons with an energy of more than 3 MeV (this energy range is important for tritium production on lithium-7). The ^{31}P(n,γ)^{32}P and ^{6}Li(n,α)^{3}H reactions were selected due to the sensitivity to thermal neutrons, which are responsible for tritium production on lithium-6. Thermal neutrons produce tritium via the ^{6}Li(n,α)^{3}H reaction with a cross

Table 2
Dosimetry reactions

Reaction	Half-life	Abundance (%)	γ/β_{max} energy (keV)	Branch ratio (%)	Threshold (MeV)
^{93}Nb(n,2n)^{92m}Nb	10.15 days	100	934.5	99.0	9
^{7}Li(n,n′α)^{3}H	12.33 years	–	18.6	100	4
^{32}S(n,p)^{32}P	14.26 days	95.03	1710	100	3
^{31}P(n,γ)^{32}P	14.26 days	100	1710	100	–
^{6}Li(n,α)^{3}H	12.33 years	–	18.6	100	–

section which is characterized by the $1/v$ law up to about 100 keV and a resonance at 240 keV (whose contribution to the resonance integral is about 0.5%). The ^{31}P(n,γ)^{32}P reaction was selected while considering these specific characteristics for the following reactions: ^{30}Si(n,γ)^{31}Si, ^{31}P(n,γ)^{32}P, ^{51}V(n,γ)^{52}V, ^{63}Cu(n,γ)^{64}Cu, ^{164}Dy(n,γ)^{165}Dy, ^{197}Au(n,γ)^{198}Au. With regards to availability of materials containing a nuclide of the interest reaction, detectors were prepared using metal foil or pellets fabricated by cold pressing of powders. The Nb foil had a size of 10 mm × 10 mm × 0.3 mm. All pellets were fabricated with a diameter of 13 mm and a thickness of 1.0 mm. Lithium carbonate, Li_2CO_3, was used for lithium-containing pellets. To measure the reaction rates on lithium isotopes, a combination of pellets with lithium-7 (99.94%) and lithium-6 (1.18%) were used. In this case, in order to minimize the perturbation effect of thermal neutron flux in the beryllium, thin Li_2CO_3 pellets with a low enrichment of lithium-6 (1.18%) were selected. The technique for measurement of tritium production rates is described in the reference [9]. The compounds of $CH_3SO_2CH_3$ and $NH_4PH_2O_2$ were used to prepare sulfur- and phosphorous-containing pellets. These compounds were selected in order to enable measurements of ^{32}P activity in water by means of Cherenkov radiation counting [10]. Gamma activities of irradiated foils were measured with the standard technique with a Germanium detector. Major sources of uncertainties for reaction rates (RR) and tritium production rates (TPR) are listed in Table 3. As a result, all contributing factors in the quadrature gave an uncertainty of about 7% in reaction rate on lithium, and about 5%—on other nuclides.

2.3. Results

Reaction rate distributions were measured along the axis of the assembly. Reaction rates of

Table 3
Estimated maximum uncertainties for the measured reaction rate

Sources of uncertainty	Magnitude (%)	
	TPR	RR
Neutron yield	3	3
Efficiency of γ/Cherenkov-detector	–	3
Efficiency of β-detector (HTO—3%[a])	3.4	–
Uncertainty of treatment procedure	5	–
Counting statistics	1	1
Number of target atoms	0.2	0.2
Pellet weight	0.1	0.1
Irradiation, cooling and measuring time	0.1	0.1
Decay data	0.5	0.5
Total	6.8	4.7

[a] Accuracy of certified HTO standard used for calibration procedure.

^{93}Nb(n,2n)^{92m}Nb, ^{7}Li(n,n′α)^{3}H and ^{32}S(n,p)^{32}P, which are sensitive to fast neutrons, are shown in Fig. 2. Distributions show an adequate tendency of fast neutrons behavior inside the assembly, where fast neutrons decrease according to their energy thresholds. Reac-

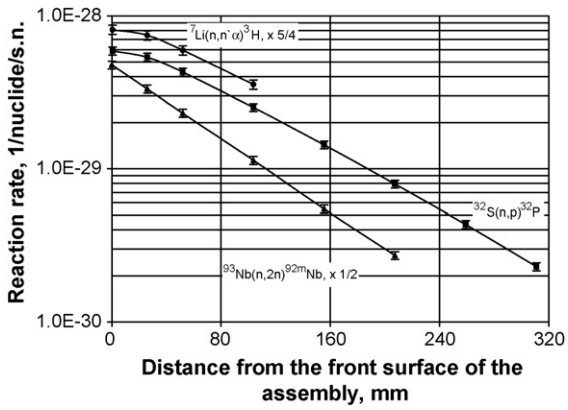

Fig. 2. Distribution of the reaction rates sensitive to fast neutrons in the beryllium assembly irradiated by neutrons from the D-T source.

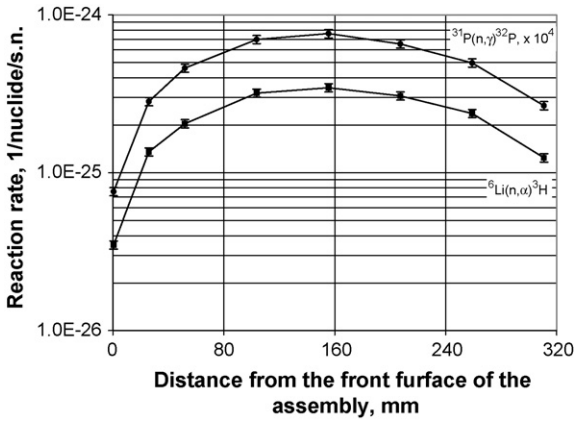

Fig. 3. Distribution of the reaction rate sensitive to slow neutrons in the beryllium assembly irradiated by neutrons from the D-T source.

tion rates of $^{6}Li(n,\alpha)^{3}H$ and $^{31}P(n,\gamma)^{32}P$, which are sensitive to slow neutrons, are shown in Fig. 3. These reactions show different tendencies, since the number of slow neutrons increases inside the assembly, due to neutron moderation and thermalization. It is important to note, that two pairs of reactions, $^{32}S(n,p)^{32}P$ with $^{7}Li(n,n'\alpha)^{3}H$, and $^{31}P(n,\gamma)^{32}P$ with $^{6}Li(n,\alpha)^{3}H$, exhibit similar reaction rate distributions. Fig. 4 shows the ratio of reaction rate for two pairs of activation reactions relevant to tritium production on lithium isotopes. It can be seen, that ratios are quite stable at various depths inside the assembly, despite the considerable change in reaction rate distributions. This observation indicates the similarity in sensitivity on the neutron spectrum for two pairs of reactions. Therefore,

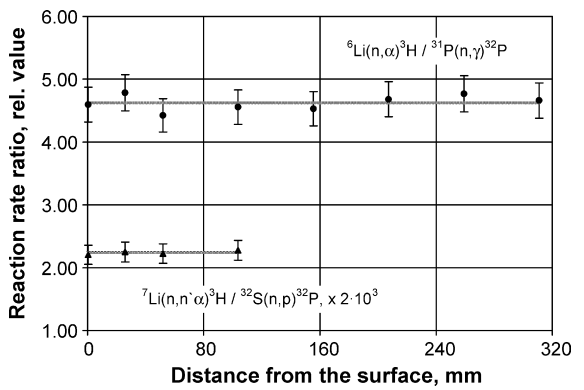

Fig. 4. Ratio of reaction rate for two pairs of activation reactions relevant to tritium production on lithium isotopes.

for future applications, the $^{32}S(n,p)^{32}P$ and $^{31}P(n,\gamma)^{32}P$ reactions can be used to obtain the integral neutron spectrum data which directly reflects tritium production via the $^{7}Li(n,n'\alpha)^{3}H$ and $^{6}Li(n,\alpha)^{3}H$ reactions, respectively.

3. Analysis

3.1. Nuclear data and the transport calculation code

The experimental results were analyzed with a three-dimensional Monte Carlo transport code MCNP-4C [11]. Transport libraries such as FENDL/MC-2.0 [12], JENDL-3.2 [13] and JENDL-3.3 [14] were used to calculate the neutron spectra. Data from neutron thermal S(α,β) tables [11] was applied for an accurate neutron transport calculation in beryllium at temperature of 300 K in the low energy range. Evaluation of reaction rates was based on the JENDL Dosimetry File, JENL-3.3 and ENDF B-VI data libraries.

3.2. Calculation pre-analysis

Special care was paid to analysis of reaction rates that are sensitive to thermal neutrons, due to present objective and large overestimations observed in previous experiments. From this point of view, several calculation pre-analyses were completed in order to estimate the significance and impact of the following factors on the $^{6}Li(n,\alpha)^{3}H$ reaction rate data:

- room returned neutrons;
- effect of impurities in beryllium material;
- impact of perturbation of thermal neutron flux by activation detectors;
- special S(α,β) treatment for scattering cross section data of thermal neutrons.

The impact of room returned neutrons on the $^{6}Li(n,\alpha)^{3}H$ reaction rate was less that 0.5% for all detector positions located inside the assembly and about 2% for detector positions located on the assembly surface. In the present experiment, all detectors were located inside the assembly, and the influence was negligibly small. The influence of the parasitic absorption of thermal neutrons by impurities in beryllium on the

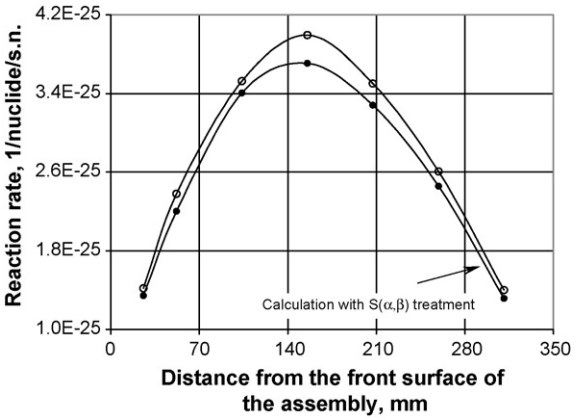

Fig. 5. Impact of S(α,β) treatment on ^{6}Li(n,α)^{3}H reaction rate inside the beryllium assembly.

reaction rate was estimated based on experimental data obtained for beryllium, utilized in the present experiment, by the integral method [6]. It was found that thermal absorption cross section of the beryllium material is approximately 30% higher than the calculated value, based on data specified by the manufacturing company for the sample. According to estimations, parasitic absorption decreases the calculated reaction rate by an average of 4%. In addition, activation pellets introduced into beryllium also caused perturbation of the thermal neutron flux and decreased the reaction rate. This effect for pellets used in the previous experiment [5] reduced the reaction rate by an average of 10%. To minimize the perturbation effect in the present experiment, thin (1 mm) pellets with low ^{6}Li content (1.18%) were used. It is also important to consider the pellet as a volume detector in the calculation model for further correct interpretation of experimental data. Another factor that has to be considered for correct evaluation of the thermal neutron spectra is a complete representation of thermal neutron scattering by solid that was encompassed by data taken from the neutron thermal S(α,β) tables (temperature of beryllium ∼300 K). Analysis shows, Fig. 5, that S(α,β) treatment decreases the reaction rate by about 6% and it is absolutely essential to use treatment in order to get a correct evaluation of the ^{6}Li(n,α)^{3}H reaction rate in the beryllium assembly. Hence, it is important to take all the above-mentioned factors into careful consideration for correct interpretation of results obtained in the present experiment.

3.3. Results

Reaction rate data gained in the experiment on the beryllium assembly irradiated by neutrons from a point D-T source was compared with appropriately calculated values. Basically, the comparison was done for calculations based on three neutron transport libraries (FENDL/MC-2.0, JENDL-3.2, JENDL-3.3) and a cross section data for activation reactions of interest taken from JENDL Dosimetry File 99, JENDL-3.3 and ENDF B-VI. Fig. 6(a)–(e) summarizes calculated to experimental rate ratios (C/E) for ^{93}Nb(n,2n)^{92m}Nb, ^{32}S(n,p)^{32}P, ^{7}Li(n,n′α)^{3}H, ^{6}Li(n,α)^{3}H and ^{31}P(n,γ)^{32}P. Except for the ^{31}P(n,γ)^{32}P reaction, reaction rates evaluated with cross sections taken from different libraries were without significant differences, therefore only JENDL Dosimetry File 99 was used to present the comparison data. For ^{31}P(n,γ)^{32}P reaction, the difference between JENDL3.3 and ENDF B-VI libraries is about 20% (Table 4). For present experimental condition, the 2200 m/s and Maxwellian spectrum average cross sections were used for comparison.

The calculations of ^{92}Nb(n,2n)^{92m}Nb reaction rate agree with the measured values within 10%, producing results identical with the previous experiment [5]. This observation is the comparison point between experiments on the beryllium assembly performed at FNS laboratory. A reasonable agreement, within 10%, was obtained for ^{32}S(n,p)^{32}P and ^{7}Li(n,n′α)^{3}H reaction rates that are sensitive to fast neutrons with an energy of more than 3 MeV for all calculation combinations with neutron transport and dosimetry libraries. The limited number of positions for the ^{7}Li(n,n′α)^{3}H reaction rate measurement was due to low contribution of tritium activity from ^{7}Li(n,n′α)^{3}H reaction. Con-

Table 4
Activation cross section data [15] for ^{31}P(n,γ)^{32}P reaction

	Thermal cross section, *b*	
	2200 m/s	Maxwellian spectrum average (energy range 10^{-5} to 10 eV)
Cross section library		
ENDF/B-VI	0.1995	0.1752
JENDL-3.3	0.1663	0.1473
Cross section ratio	1.20	1.19

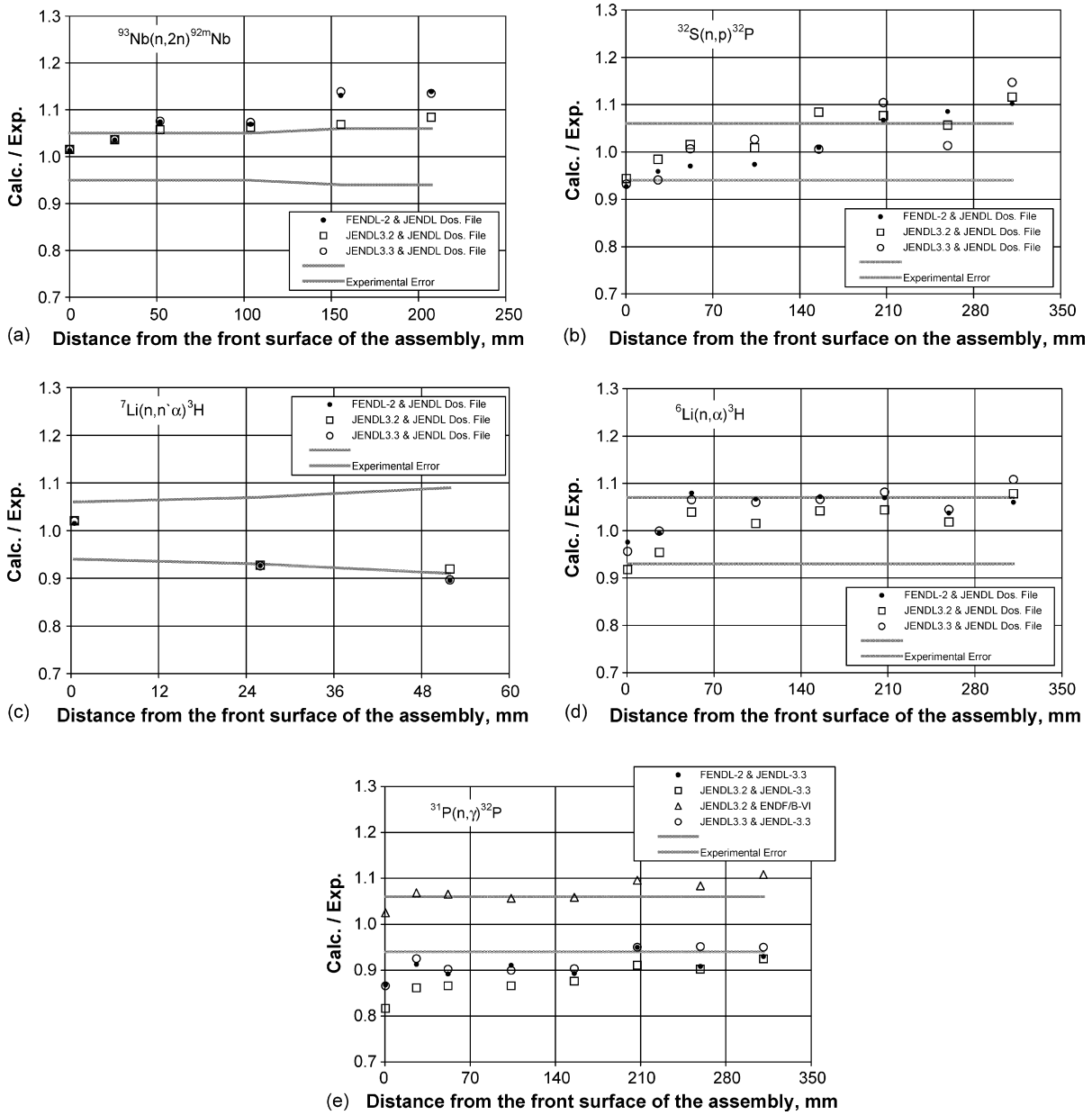

Fig. 6. (a–e) The calculated to experimental rate ratio for activation reactions in the beryllium assembly irradiated by neutrons from the D-T source.

sidering the C/E data obtained for ^{93}Nb(n,2n)^{92m}Nb and ^{32}S(n,p)^{32}P reactions, it is possible to conclude that there is an insignificant increasing trend of C/E values from 1.0 to 1.1 with the depth inside the assembly. A similar observation was obtained from previous analyses [5] for most threshold-type reactions (such as ^{27}Al(n,α)^{24}Na, ^{56}Fe(n,p)^{56}Mn and ^{58}Ni(n,p)^{58}Co). The reason for this tendency is not clear.

The ^{6}Li(n,α)^{3}H and ^{31}P(n,γ)^{32}P reactions have a similar sensitivity to thermal neutrons and such

finding is supported by the constant reaction rate ratio obtained in the present experiment (Fig. 4). Therefore, it is expected that C/E values for both reactions have to show the same tendency, assuming that the cross section data for reactions is well known. It is possible to conclude according to obtained data, Fig. 6(d)–(e), that calculated reaction rates are in a reasonable agreement with experimental values. The calculations of $^{6}Li(n,\alpha)^{3}H$ reaction rate agree with the measured values within 10% for all considered calculation combinations. The calculations of $^{31}P(n,\gamma)^{32}P$ reaction rate agree with the measured values within 10% for the reaction cross section data taken from the ENDF/B-VI library, although they show an insignificant underestimation, approximately 10%, for data taken from the JENDL-3.3 library. The observed difference between ENDF/B-VI and JENDL-3.3 libraries is around 20%. This experimental value is in good agreement with the estimation based on the cross section data, Table 4.

Considering all C/E values obtained in the present experiment it is possible to conclude that the calculation of design-relevant nuclear response, such as tritium production rate on lithium isotopes, using FENDL-2 or JENDL-3.2/3.3 provides an acceptable agreement with the experimental data.

4. Conclusions

A clean benchmark experiment on beryllium with the main objective of verifying the integral data related to tritium production on lithium isotopes was executed. In order to meet the objective the $^{7}Li(n,n'\alpha)^{3}H$ and $^{6}Li(n,\alpha)^{3}H$ reaction rates were measured within an accuracy of less than 7%. In addition to these reactions, the $^{32}S(n,p)^{32}P$ and $^{31}P(n,\gamma)^{32}P$ reactions that exhibit the neutron spectrum sensitivity similar to the $^{7}Li(n,n'\alpha)^{3}H$ and $^{6}Li(n,\alpha)^{3}H$, respectively, were measured in order to provide additional verification of integral parameters related to tritium breeding. The similarity of neutron spectrum sensitivity of reactions was demonstrated in the experiment.

Experimental analyses were performed using a three-dimensional Monte Carlo transport code MCNP-4C with FENDL/MC-2.0, JENDL-3.2/3.3 transport libraries. It is absolutely essential to use the data from neutron thermal $S(\alpha,\beta)$ tables for correct evaluation of the $^{6}Li(n,\alpha)^{3}H$ reaction rate. Evaluation of reaction rates was based on the JENDL Dosimetry File 99, JENDL-3.3 and ENDF B-VI data libraries. All calculation combinations (transport and activation cross section libraries) used for evaluation of reaction rates give data that is agreeable with measured values within 10%. Results for reactions with a similar neutron sensitivity show a reasonable consistency of obtained C/E values.

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