Benchmark Experiments on a 60 cm-thick Li$_2$O Slab Assembly and Their Analyses


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Abstract

Integral measurements of a Li₂O slab assembly have been carried out using the FNS to provide the benchmark data for checking the data and methods. The size of assembly was 63 cm in diameter and 61 cm thick. Measured items were tritium production rates of ⁶Li and ⁷Li, fission rates of ²³⁵U, ²³⁸U, ²³⁷Np and ²³²Th, reaction rates of ²⁷Al(n,α)²⁴Na, ⁵⁸Ni(n,2n)⁵⁷Ni, ¹¹⁵In(n,n')¹¹⁶In and ¹¹⁵In(n,γ)¹¹⁶In, and in-system neutron spectra. Measured values were analyzed by three transport codes, BERMUDA-2DN, MORSE-DD and DOT3.5 using nuclear data files of JENDL-3PR1 and ENDF/B-4. Observed data are consistent with each other even if the measuring methods are different. Using JENDL-3PR1, the calculations predict the tritium production rates of both ⁶Li and ⁷Li very well. For threshold reactions, there are disagreements among the results of calculations, and between the measured and calculated results. Further investigations should be necessary to make them clear.
I Introduction

To design a controlled thermonuclear reactor blanket, it is necessary to know exactly the behavior of neutrons in the blanket. The data and methods used to analyze the neutronics in the blanket should be examined by comparing the calculated results with experiments. At the JAERI, integral experiments on several blanket assemblies had been carried out using PNS-A neutron source. After the construction of the powerful neutron source named as the Fusion Neutronics Source (FNS), benchmark experiments on simulated blanket assemblies have been carried out at the JAERI. The tritium production-rate distributions were measured in two blanket assemblies.

Lithium oxide (Li$_2$O) has been proposed by JAERI's designers as a solid state tritium breeding material. Benchmark experiments on the fusion blanket of Li$_2$O applying the FNS are strongly requested to check the data and methods. In the calculation for analyzing the fusion neutronics, angular distribution of secondary neutrons should be treated accurately. Two transport codes BERMUDA-2DN and MORSE-DD have been developed at the JAERI. Double differential form cross section (DDX) are used in the two codes. And also a new nuclear data file having DDX has been evaluated by Japanese Nuclear Data Committee as JENDL-3PR1 (JENDL-3 preliminary version one).

As the benchmark data for fusion neutronics, following properties are important and desirable.

1) Measured data are absolute values to compare with their analysis directly.
2) Measured data are as many as possible to obtain a lot of informations from them.

Integral measurements of a 60 cm-thick Li$_2$O slab assembly have been done considering the properties mentioned above. Measured items and their methods are summarized as follows:

1) Tritium production-rate distributions
   i) liquid scintillation method with $^6$Li$_2$O and $^7$Li$_2$O pellets
   ii) a pair of $^6$Li and $^7$Li glass scintillators

2) Fission-rate distributions
   i) micro-fission chambers of $^{235}$U, $^{238}$U, $^{237}$Np and $^{232}$Th
ii) solid-state track recorders with $^{235}$U, $^{239}$U and $^{232}$Th foils

(3) Reaction-rate distributions of $^{27}$Al($n,\alpha$)$^{24}$Na, $^{58}$Ni($n,2n$)$^{57}$Ni, $^{115}$In($n,n'$)$^{115}$In and $^{115}$In($n,\gamma$)$^{116}$In

i) foil activation method using Al, Ni and In foils

(4) In-system neutron spectra

i) small sphere NE213 spectrometer

ii) multi-foil activation method

Response distributions of PIN diodes and thermoluminescence dosimeters were also measured as an effort to develop new techniques.

Items of (1-i), (2) and (3) are discussed in this report. The measured reaction rates were analyzed by using above two codes and JENDL-3PR1 along with DOT3.5(15) and ENDF/B-4 nuclear data file.

II Experimental Assembly and Neutron Source

The size of Li$_2$O slab assembly was 31.5 cm in equivalent radius and 61.0 cm in thickness. Lithium-oxide blocks were stacked to form a cylindrical geometry in a frame composed by stacking thin-walled aluminum square tubes. Sectional views of the Li$_2$O slab assembly and Li$_2$O blocks are shown in Fig. 1. Three types of Li$_2$O blocks were made to the sizes of 5.06 cm x 5.06 cm x (5.06, 10.12 and 20.24) cm long. They were fabricated by sealing one, two and four Li$_2$O bricks, respectively, in a 0.2 mm-thick stainless steel box. The bricks which were almost cubic, were made from Li$_2$O powder by cold pressing. Density of Li$_2$O bricks was 75.5 % of theoretical density.

An experimental channel — a set of square tube and drawer made of 0.2 mm-thick stainless steel — and special-sized Li$_2$O blocks were prepared for saving the changing time of detector position and for minimizing the personnel exposure for experimentalists. This experiment channel was set at the central axis of the assembly.

The 80° beam line in the first target room of the FNS facility was used in the present experiments. A high speed water-cooled target assembly (16) was set at the end of the beam line. A 20 Ci Ti-T target was mounted on the target assembly. Neutrons were generated at the distance of 20 cm from the surface of assembly on its central axis.
Neutron yields were determined by means of the associated α-particle detection method. A small silicon surface-barrier detector was mounted in the target assembly to detect the α-particle of $^3$T(d,n)$^4$He reaction. Source characteristics — neutron yield, angular distribution and spectra — of this target assembly were measured by the time-of-flight method, the foil activation method and an NE213 spectrometer. Measured neutron yields agreed well each other within the experimental errors. They were analyzed by Monte Carlo calculations. Fairly good agreements between measurements and calculations were obtained for the neutron energy spectra.

III Measurements

1. Tritium Production-Rate (TPR) Distributions

The TPR distributions were measured by the liquid scintillation counting method with sintered Li$_2$O pellets. The size and density of pellet were 12 mm dia. x 2 mm and about 83 % T.D., respectively. Enriched $^6$Li(95.446 atom %) and $^7$Li(99.952 atom %) pellets were adopted to obtain the reaction of $^6$Li(n,α)$^3$T and $^7$Li(n,n'α)$^3$T, separately. The pellets were placed at the small spaces between the Li$_2$O blocks along the central axis. Total neutron yield at the target was $9.34 \times 10^{15}$ for the irradiation.

Irradiated pellets were dissolved in 6.5 ml of water. It took a couple of days to get complete solution. The solution was distilled with an apparatus shown in Fig. 2. As the first step, the solution was poured into a "boiling flask" and was frozen in a liquid nitrogen dewar before the apparatus was evacuated by a rotary pump. After complete evacuation, the flask was warmed up with a water bath of about 80°C and a "condensing flask" was, in turn, cooled down with a liquid nitrogen dewar. The 6 ml of the collected water was pipetted into a 20 ml vial. Then, 4 ml of ethylalcohol and 10 ml liquid scintillation cocktail were added in it. The addition of ethylalcohol made liquid (single phase) samples clear and stable. The tritium activities in the samples were measured by a low background liquid scintillation counting system (Aloka LSC-LBI).

To estimate the tritium escaped during the irradiation and
chemical treatment, the same type pellets covered with aluminum foils were placed near the rotating target of the FNS. The foils, in which recoil tritons were trapped during the irradiation, were treated chemically and the tritium was measured by a radio gas chromatograph. The HT gas undissolved in the solution was extracted by the D₂ gas bubbling method. The total escaped tritium rates were (6.1 ± 1.3) % and (7.1 ± 0.9) % for ⁹Li₂O and ⁷Li₂O pellets, respectively. After the correction of escaped tritium, the TPRs of ⁹Li and ⁷Li were obtained considering the atomic ratios of pellets.

2. Fission-Rate Distributions

The absolute fission-rates of ²³⁵U, ²³⁸U, ²³⁷Np and ²³²Th were measured with four micro-fission chambers coated respectively with the oxides of ²³⁵U, ²³⁸U, ²³⁷Np and ²³²Th. The chambers were Type FC4A manufactured by 20th Century Electronics. These chambers were calibrated using 15 MeV neutrons produced by the 80° beam line of the FNS. All the counter traverses were made along the experimental channel. The fission rates of the ²³⁵U and ²³⁸U chambers were measured simultaneously by placing the two chambers side by side in the same hole. Similar measurements of fission rate were carried out on the ²³⁷Np and ²³²Th chambers. As the detectors were successively withdrawn along the central axis, the blocks with experimental hole were replaced by solid blocks of the same material, to minimize neutron streaming. The remaining part of the hole on the back side of the detector served to pass the instrument lead wires.

The uranium in the ²³⁵U micro fission chamber contained 7 % ²³⁸U, and that in the ²³⁸U chamber 0.044 % ²³⁵U, for which appropriate corrections were brought to the observed fission rates. In the ²³⁷Np chamber, the neptunium contained 0.1 % plutonium, mostly ²³⁹Pu. The fission of this plutonium was corrected with its reaction rate estimated from that of ²³⁵U assuming an average ratio of 1.26 between the cross sections of ²³⁹Pu and ²³⁵U. The error due to this assumption should be small. The corresponding amount of impurity for ²³²Th chamber was 0.1 %. Since its composition was unknown, the results obtained on ²³²Th were not corrected.
3. Reaction-Rate Distributions of $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$, $^{58}\text{Ni}(n,2n)^{57}\text{Ni}$, $^{115}\text{In}(n,n')^{115}\text{In}$ and $^{115}\text{In}(n,\gamma)^{116}\text{In}$

Reaction-rates of $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$, $^{57}\text{Ni}(n,2n)^{56}\text{Ni}$, $^{115}\text{In}(n,n')^{115}\text{In}$ and $^{115}\text{In}(n,\gamma)^{116}\text{In}$ were measured by the foil activation method. The sizes of Al and Ni foils were 15 mm dia. x 1 mm, while the size of In foil was 10 mm x 10 mm x 0.1 mm. The foils were set in the same manner that was used in the irradiation of Li$_2$O pellets. The irradiation time and total neutrons generated at the target were about 12 hours (42900 s) and $1.20 \times 10^{16}$, respectively. The neutron yield rate was monitored continuously for the correction of flux change with time.

After the irradiation, the activities of foils were measured by a 60 cm$^3$ Ge(Li) detector. The measured $\gamma$-ray count was converted to the reaction rate using the data of neutron yield, detector efficiency, branching ratio, half-life and foil weight. The results were corrected for sum-peak, self-absorption and neutron flux fluctuation.

IV. Analyses

The neutron fluxes in the assembly were calculated by using the three transport codes DOT3.5, BERMUDA-2DN and MORSE-DD. The latter two codes have been developed recently at the JAERI, which do not use Legendre expansion method but use double differential form cross sections (DDX). The ENDF/B-4 and JENDL-3PR1 (JENDL-3 preliminary version one) were used as the basic nuclear data. The features of each code and calculational conditions are summarized in Table I.

The assembly is modeled as a cylindrical slab geometry composed of Li$_2$O, canning and support materials. The size of assembly is 63 cm in diameter and 61 cm thick. The isotropic point source is placed at the distance of 20 cm from the front surface of assembly. The source neutron spectrum at the target position was calculated by a Monte Carlo method. The first collision source method was adopted for DOT3.5 and BERMUDA-2DN. The track length estimator was used in MORSE-DD. As the reaction cross sections for the analysis of foil activation method, the data of ENDF/B-5 dosimetry file were used except for $^{58}\text{Ni}(n,2n)^{57}\text{Ni}$. The cross section of $^{58}\text{Ni}(n,2n)^{57}\text{Ni}$ has been measured recently at the FNS. This data was adopted in this analysis.
V. Results and Discussions

1. Tritium Production-Rate Distributions

The values of C/E (ratio of calculated to measured values) for TPR are shown in Figs. 3 and 4 for $^6$Li and $^7$Li, respectively. The result of experimental error analysis is summarized in Table II. The range of experimental error is given in the figures. The measured values were corrected for the tritium decay between the irradiation and the tritium counting, and for the escaped tritium from the pellet during the irradiation and the dissolution of pellet.

For the TPR of $^6$Li, the results of DOT3.5 and MORSE-DD using the JENDL-3PR1 agree well with those of the experiment over whole region, while BERMUDA gives the higher values by a few percents than the others. It is clear that the calculational results using the JENDL-3PR1 predict the TPR of $^6$Li very well comparing with those using the ENDF/B-4, especially in the region near the front surface. In the region near the rear surface, the room return effect and the self-shielding effect become important. The room return effect from the wall and floor of experimental room was estimated by using MORSE-DD. The contribution to TPR is estimated to be 6.9, 3.8, 2.4 and 1.6 % at the distances of 55.8, 50.7, 45.6 and 40.5 cm, respectively. The corrections for these effect are not made in the figure. From a rough estimation, the self-shielding effect is a few percents except for the data near the surfaces. This effect will be examined in near future.

For the TPR of $^7$Li, the results of three codes using the JENDL-3PR1 agree very well with the measured values within the experimental error, while those using the ENDF/B-4 are higher than the measured values by about 20 %. The data in the region of 35 ~ 81 cm could not be measured because the TPR was very low and the tritium count was close to the background of the liquid scintillation counting system.

2. Fission Rate and Reaction Rate Distributions

The values of C/E for fission rate are shown in Figs. 5 ~ 9 for $^{235}$U, $^{237}$Np, $^{238}$U and $^{232}$Th, respectively. Experimental error was estimated to be about 5 % except for a few data of $^{232}$Th. The
reaction rate measured by the foil activation method are also compared with those calculated by DOT3.5 as the value of C/E. They are shown in Figs. 10-13 for $^{58}$Ni(n,2n)$^{57}$Ni, $^{27}$Al(n,$\alpha$)$^{24}$Na, $^{115}$In(n,n')$^{115}$In and $^{115}$In(n,$\gamma$)$^{116}$In, respectively. Experimental errors were from 2.8 to 10.3% depending mainly on the statistical error in the $\gamma$-ray measurement.

The room return effect for the fission rate of $^{235}$U is estimated to be 7.4, 3.8, 2.2 and 1.4% at the distances of 57.2, 52.1, 47.1 and 42.0 cm, respectively. The tendency of C/E curve for $^{235}$U is very close to that for TPR of $^6$Li. The calculated reaction rates of $^{58}$Ni(n,2n)$^{57}$Ni agree very well with measured ones for both JENDL-3PR1 and ENDF/B-4. For other reactions such as $^{238}$U(n,f) and $^{27}$Al(n,$\alpha$)$^{24}$Na, there are differences not only between the calculational results of JENDL-3PR1 and ENDF/B-4 but also among the codes. Further investigations should be necessary to make clear above discrepancies among calculations and between the measured and calculated values.

VI Summary

(1) Integral experiments on a 60 cm-thick Li$_2$O slab assembly have been down. Four types of fission rates and four types of reactions were measured by micro-fission chambers and foil activation method. As those reactions have various response, useful informations can be obtained from the measured values.

(2) Measured data are consistent with each other even if the measuring methods are different. They can be used for checking data and method as the benchmark data.

(3) Using JENDL-3PR1, the calculations predict the tritium production rates of both $^6$Li and $^7$Li very well.

(4) For threshold reactions, there are disagreements among the results of calculations, and between the measured and calculated results. Further investigations should be necessary to make them clear.
Acknowledgement

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References


(5) idem.: ibid. 14, 219 (1977)


(14) Shibata, K., et al.: private communication


(20) Ikeda, Y., et al.: To be published
Table I Features of each code and calculational conditions

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*1 for ENDF/B-4 cross section set GICXFNS1(24)
*2 for JENDL-3PR1 GICXJ3
*3 collapsed group constant from a 121 group set
Fig. 1 Sectional views of Li$_2$O slab assembly and Li$_2$O blocks
Table I  Error analysis for the TPR measurement by liquid scintillation method with sintered Li$_2$O pellet

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<td>Self-Shielding</td>
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Fig. 2  A sketch of distillation apparatus
Fig. 3 Comparison of C/E values for tritium production rate of $^6$Li

Fig. 4 Comparison of C/E values for tritium production rate of $^7$Li
Fig. 5 Comparison of C/E values for fission rate of Th-232

Fig. 6 Comparison of C/E values for fission rate of U-238
Fig. 7  Comparison of C/E values for fission rate of Np-237

Fig. 8  Comparison of C/E values for fission rate of U-235
Fig. 9 Comparison of C/E values for reaction rate of $^{27}$Al($n,\alpha$)$^{24}$Na

Fig. 10 Comparison of C/E values for reaction rate of $^{56}$Ni($n,2n$)$^{57}$Ni
Fig. 11 Comparison of C/E values for reaction rate of $^{115}\text{In}(n,\gamma)^{116}\text{In}$

Fig. 12 Comparison of C/E values for reaction rate of $^{115}\text{In}(n,n')^{115m}\text{In}$