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A new powerful D-T neutron source FNS was constructed in Japan Atomic Energy Research Institute. Tritium production-rate (TPR) distributions were measured in simulated fusion blanket assemblies using the FNS. They were pseudo-spherical lithium oxide assembly with a graphite reflector and 40 cm thick lithium-oxide slab assembly. Three methods applied to measure the TPR were (A) Dierckx's method, (B) Self-irradiation method using a LiF thermoluminescence dosimeter and (C) Liquid scintillation counting method using a sintered Li_2O pellets. Methods (B) and (C) have been developed in the JAERI. Measured TPR distributions agreed well among the three methods. It has been demonstrated that methods (B) and (C) are very useful for the fusion neutronics experiments. Experimental results support the P. G. Young's evaluation for $^{7}Li(n,n'\alpha)^{3}T$ cross section.

Keywords : Fusion Blanket, Neutronics, Tritium Production Rate, Thermoluminescence Dosimeter, Lithium Oxide, Liquid Scintillator, FNS, $^{7}Li(n,n'\alpha)^{3}T$, Cross Section

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ＦＮＳにおける模擬ブランケット体系中のトリチウム生成率分布の測定

日本原子力研究所東海研究所原子炉工学部
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（１９８３年10月26日受理）

原研に新しい強力なＤ－Ｔ中性子源であるＦＮＳが完成した。模擬核融合炉ブランケット体系中のトリチウム生成率（ＴＰＲ）分布をＦＮＳを用いて測定した。模擬体系は黒鉛反射体付き酸化リチウム球体系と40ｃｍ厚さの酸化リチウム平板体系である。ＴＰＲを(A)Dierckx の方法，(B)ＬｉＦのＴＬＤの自己照射法，(C)Li_2Oの焼結体ペレットによる液体シンチレータ法の３種類で測定した。(B)と(C)は原研で開発した方法である。測定されたＴＰＲの分布は３つの方法の間で良く一致した。(B)と(C)の方法が核融合炉のニュートロニクスの実験で有効であることを示した。実験結果はP. G. Youngの評価した^{7}Li（ｎ，ｎ′α）^{3}Ｔの断面積が妥当であることを示唆した。

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1. Introduction

The measurement of tritium production-rate (TPR) distribution in a simulated blanket assembly is one of the most important items in the fusion neutronics study. At the JAERI, integral experiments on several blanket assemblies had been carried out using the PNS-A neutron source.[1]~[7] As the neutron yield of PNS-A was not strong enough to measure the TPR, we had planned to construct a new powerful neutron source. This was named as the Fusion Neutronics Source (FNS).[8] In March 1981, the FNS was completed successfully achieving the designed specifications, i.e., intense deuteron beam was more than 20 mA at the rotating target for continuous beam operation, and pulse width and peak current were less than 2ns and more than 45 mA, respectively for pulsed beam operation.[9]

The TPR distributions were measured in two blanket assemblies. They were pseudo-spherical lithium oxide assembly with a graphite reflector (Li_2O-C) and 40 cm-thick lithium-oxide slab assembly (40 cm-Li_2O). Lithium oxide (Li_2O) has been proposed in the JAERI as a solid state tritium breeding material.[10] Benchmark experiments on the fusion blanket of Li_2O applying the FNS are necessary to check the data and method used in the neutronics design.

2. Measuring Techniques

The TPR distributions in simulated fusion blanket assemblies have been measured in several institutes.[11]~[14] The method proposed by Dierckx[15] was widely used to determine the TPR. According to this method, small Li_2CO_3 probes are irradiated in a blanket assembly. Then they are treated chemically and produced tritium is measured by a liquid scintillation counting system. Proportional counter method was also adopted using lithium metal and LiH samples.[11],[14]

In the present study, following three methods were used to measure the TPR.

(A) Dierckx's method.

(B) Self-irradiation method using a LiF thermoluminescence dosimeter (TLD).[16]

(C) Liquid scintillation counting method using a sintered Li_2O pellet.

1. Introduction

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(A) Dierckx's method.

(B) Self-irradiation method using a LiF thermoluminescence dosimeter (TLD).[16]

(C) Liquid scintillation counting method using a sintered Li_2O pellet.

Method (B) was proposed by one of the authors and this is the first chance in our application to the fusion neutronics experiments.

Method (C), that we have developed recently, has following advantages:

i) Chemical composition of pellet is just the same as the experimental blanket assembly.

ii) Sintered Li_2O pellet is fairly stable and easy to handle.

iii) Atomic density of Li per unit volume is highest among lithium compounds.

iv) Chemical treatment of pellet is very simple to make a liquid scintillation sample.

Chemical treatment of Li_2O pellet is as follows:

i) Put the irradiated Li_2O pellet (12 mmϕ × 2 mm, ∿0.38 g for natural isotopic composition) in a 20 ml Teflon vial and add 6 ml of water. Close the vial and wait until the pellet is dissolved completely. It takes about two days at the normal room temperature.

$$Li_2O + H_2O \longrightarrow 2\ LiOH$$

The tritium trapped in the Li_2O pellet transfers to the solution.

ii) Add 0.95 ml of fluoric acid (46.5 w/o). Close the vial and wait for 2 ∿ 3 hours until the solution is cool.

$$LiOH + HF \longrightarrow LiF\downarrow + H_2O$$

iii) Centrifuge the vial to deposit the salt of LiF completely.

iv) Add 10 ml liquid scintillator (Aquazol-2) kept warm at 60°C. Then the solution becomes cloudy. Close the vial.

v) Turn the vial upside down and shake it gently and carefully not to mix the LiF salt into the solution.

vi) Return the vial to normal direction and let it settle down about 30 minutes. Then the solution becomes translucent gel and is ready for the sample to be measured by a liquid scintillator counting system.

In order to compare this method with the conventional method, the TPR in a lithium blanket assembly (See 3.3) was measured by both Li_2O and

Li_2CO_3 pellets. Natural, enriched ^6Li (95.446 atom %) and ^7Li (99.952 atom %) pellets of Li_2O and Li_2CO_3 were fabricated from very high purity powder of Li_2CO_3.[17] Total impurities in $^{N}Li_2CO_3$, $^6Li_2CO_3$ and $^7Li_2CO_3$ powder were 2.42, 175 and 93 ppm. Both Li_2O and Li_2CO_3 pellets were irradiated simultaneously in the lithium-oxide slab assembly at the distances of 10 and 20 cm from the front side along its central axis. The size and weight of Li_2O and Li_2CO_3 pellets were 12 mmϕ × 2 mm, ∿0.38 g and 13 mmϕ × 4 mm, ∿0.85 g, respectively for natural abundance.

The results were summarized in Table 1. The experimental errors were 1 ∿ 6 % for random one. The data Li_2O agree well with those of Li_2CO_3 within the error. It was demonstrated that the liquid scintillation method using a sintered Li_2O pellet can be applied to the fusion blanket experiment.

Methods (A) and (B) were adopted for the Li_2O-C assembly, and method (A) and (C) were adopted for the 40 cm-Li_2O slab assembly.

3. Experiments

3.1 Neutron Source

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 was set at the end of the beam line. This target was designed to keep its surface temperature below 200°C for 1 kW beam.[18] A 20 Ci Ti-T target was mounted on the target assembly.

Neutron yield was determined by means of the associated α-paticle detection method.[19] A small silicon surface-barrier detector (SSD) was mounted in the target assembly to detect the α-paticle of $^3T(d,n)^4He$ reaction. The distance from the target center to the slit was 157.8 cm. The directions of incident deuteron beam and detected α-particles formed an angle of about 179°. With the most careful adjustment of the parameters in the monitor system and the treatment of the data, the neutron yield can be obtained within 2.2 %.

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.[20] They

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were analyzed by Monte Carlo calculations.[21] Fairly good agreements between measurements and calculations have been obtained for the neutron energy spectra.

3.2 Li_2O-C Spherical Assembly

As the first blanket experiment using the FNS, the TPR distribution was measured in a pseudo-spherical lithium-oxide assembly with a graphite reflector (Li_2O-C). It was just the same assembly measuring absolute fission-rate distributions in the experiment previously performed with PNS-A neutron source.[7]

The Li_2O-C assembly was formed by loading Li_2O and graphite blocks in stainless steel drawer which in turn were inserted into a stainless steel lattice matrix. The horizontal section across the center of the assembly is shown in Fig. 1. The central cavity, Li_2O and graphite regions are 3.3, 22.4 and 46.8 cm respectively in effective cuter radius. Source neutrons generated at the center of the assembly.

Three types of blocks were made to sizes of 5.06 cm × 5.06 cm × (5.06 cm, 10.2 cm and 20.3 cm) long. They were fabricated by sealing one, two and four bricks, respectively, in a 0.1 mm-thick stainless steel box. The Li_2O bricks which are almost cubic, were made from Li_2O powder by cold pressing. Density of Li_2O bricks was 70 % of theoretical density.

The tritium production rate was measured by the liquid scintillation method using Li_2CO_3 pellets[15] and by the self-irradiation method of LiF thermoluminescence dosimeters (TLD).[16]

The Li_2CO_3 pellets (10 mmϕ × 5 mm, ∿0.7 g) of natural abundance and enriched ^7Li(99.952 atom %) were placed in the Li_2O blocks having a hole of 16.6 mm × 16.6 mm along the 0° direction to incident d^+ beam(See Fig. 1). After the irradiation in the assembly, the pellets were taken out and chemically treated. The number of tritium in the sample was measured by the liquid scintillation technique. The total neutron yield during the irradiation was 4.5×10^{15} neutrons.

The powder of TLD-600(^6Li: 95.62 atom %) and TLD-700(^7Li: 99.993 atom %) purchased from Harshaw Chemical Co. Ltd. was sealed in 2 mmϕ × 12 mm Pyrex glass ampoules. Two pairs of TLD-600 and -700 were set along both 0° and 90° directions to the incident d^+ beam(See FIg. 1). For thirteen

days after the neutron irradiation to wait the full decay of activated nuclei except for tritium, the TLD's were annealed twice in the condition of 400°C and one hour. After this process, they were stored in a low background storage for 2215 hours (about three months). The thermoluminesence caused by the self-irradiation of β-rays from tritium during the keeping period were measured by a TLD reader (National UD-502B).

3.3 40 cm-thick Li_2O Slab Assembly

The size of Li_2O slab assembly was 31.4 cm in equivalent radius and 40.5 cm in thickness. The specifications of Li_2O blocks were same as those in the Li_2O-C assembly except for the thickness of stainless steel box and the density of bricks. They were 0.2 mm and 75.5 % of theoretical density, respectively. Lithium-oxide blocks were stacked to form a pancake cylinder in a frame composed by stacking thin-walled aluminum square tubes. The target was located at 10 cm from the front surface of the assembly on its central axis.

The TPR distributions along the central axis of the assembly were measured by the liquid scintillation counter method with Li_2O and Li_2CO_3 pellets. Enriched ^6Li and ^7Li pellets of Li_2O and Li_2CO_3 were adopted to measure the reaction rate of $^6Li(n,\alpha)^3T$ and $^7Li(n,n'\alpha)^3T$, separately. Enrichment, size and weight of pellets were same as mentioned in Section 2. The pellets were set at the small spaces between the Li_2O blocks along the central axis. Total neutron yields at the target were 5.18×10^{15} for the irradiations of Li_2O pellets and 1.11×10^{16} for the Li_2CO_3 pellets.

After the irradiations these pellets were treated chemically to make the samples as described in Section 2. The tritium activities in the samples were measured by a low background liquid scintillation counting system (Aloka LSC-LB1).

4. Analyses

4.1 Analysis for the Li_2O-C Assembly

A 135-group cross section set used in the calculation was GICXFNS[22] obtained from ENDF/B-IV data file except the cross section of $^7Li(n,n'\alpha)^3T$

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and Carbon. The data of P.G. Young's evaluation[23] and ENDF/B-V were adopted for ^{7}Li and C, respectively. Figure 2 shows the spherical model used in the calculation of Li_2O-C assembly. The region marked "void" in Fig. 2 consists of stainless-steel lattice and empty drawers, while only lattice framework is present in the portion designated "lattice" in Fig. 2. The homogenized nuclide densities of respective regions are tabulated in Table 2.

The neutron fluxes in the assembly were calculated by one-dimensional transport code ANISN[24] with P_5-S_{16} approximation. The result of the Monte Carlo calculation[21] was adopted as the input source neutron spectrum. For this purpose, a 135-group spectrum was generated from the angle-averaged spectrum of 24 groups in Table 3 of Ref. 21 by means of logarithmic interpolation. The isotropic neutron source was assumed in this calculation.

4.2 Analysis for the 40 cm-Li_2O Slab Assembly

The GICXFNS cross section set was also used in this analysis. The calculational model of the Li_2O slab assembly was shown in Fig. 3. The homogenized nuclide densities are given in Table 3. The two-dimensional transport code DOT 3.5[25] was used to calculate the fluxes in the assembly. The spectrum in zero-degree direction of the Monte Carlo calculation[21] was adopted as the input source spectrum. The neutron was assumed to be emitted isotropically at the target. The angular distribution of source neutrons was anisotropic actually due to the kinematics and the effect of target assembly. To follow the measured value that was normalized to unit source neutron at the target, the source normalization factor should be 1.177 in stead of unity. The factor was 4π times the summation of the zero-degree spectrum in Table 3 of Ref. 21. The first collision source (GRTUNCL code) and P_5-S_{16} approximation were employed in this analysis.

5. Results and Discussions

5.1 Li_2O-C Assembly

To estimate the TPR for $^{6}Li(n,\alpha)^{3}T$ and $^{7}Li(n,n'\alpha)^{3}T$, the measured data were processed considering the atomic densities of ^{6}Li and ^{7}Li in

and Carbon. The data of P.G. Young's evaluation[23] and ENDF/B-V were adopted for ^{7}Li and C, respectively. Figure 2 shows the spherical model used in the calculation of Li_2O-C assembly. The region marked "void" in Fig. 2 consists of stainless-steel lattice and empty drawers, while only lattice framework is present in the portion designated "lattice" in Fig. 2. The homogenized nuclide densities of respective regions are tabulated in Table 2.

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To estimate the TPR for $^{6}Li(n,\alpha)^{3}T$ and $^{7}Li(n,n'\alpha)^{3}T$, the measured data were processed considering the atomic densities of ^{6}Li and ^{7}Li in

the Li_2CO_3 pellets and TLD's. Measured TPR distributions of total, $^6Li(n,\alpha)^3T$ and $^7Li(n,n'\alpha)^3T$ are shown in Fig. 4 with the calculated ones. The experimental values of TLD are normalized to the result of Li_2CO_3 pellet at 5.3 cm. Random and systematic errors for Li_2CO_3 pellet are 2 ∿ 5 % and 6.5 %, respectively. The random error for TLD is 2 ∿ 20 %.

The TPR distributions of TLD and Li_2CO_3 pellet agree well each other. The calculated TPRs agree mostly with the measured ones, however, there exist some discrepancies near the target. As a clue to clarify the difference, simple survey calculations were carried out. It became clear that the source neutron spectrum below 1 MeV affected strongly the distribution of $^6Li(n,\alpha)^3T$ near the target. Above discrepancy might be attributed to the source spectrum calculated by the Monte Carlo method. There were large differences between the calculated and measured source spectra below 1.7 MeV.[21] Further investigations about the source spectrum should be necessary to make it clear the discrepancy. The observed value of $^6Li(n,\alpha)^3T$ at 19 cm differs from the calculated one. This is caused by the irregular boundary between the Li_2O and graphite regions, and by the limitation of one-dimensional model.

As is evident from Fig. 4, the data of $^7Li(n,n'\alpha)^3T$ were represented better by Young's evaluation than that of ENDF/B-IV previously used. To confirm this, the cross section of $^7Li(n,n'\alpha)^3T$ was measured at 14.9 MeV by irradiating Li_2CO_3 pellet in the source spectrum field of the FNS. The obtained value was (0.259 ± 0.018)b.

5.2 40 cm Li_2O Slab Assembly

The experimental results are shown in Fig. 5 with the calculated ones. The measured $^6Li(n,\alpha)^3T$ and $^7Li(n,n'\alpha)^3T$ reaction rate were estimated by the same manner mentioned above. They are named as T_6 and T_7, respectively. The tritium production rates for natural abundance (T_N) are given in the same figure. The systematic and random errors are estimated to be ±5.4 % and 1 ∿ 15 %, respectively. The measured values by two different methods agree well each other.

The calculational results of T_6 are corrected for the self-shielding effect assuming that the pellet size is 12 mmϕ × 2mm and atom density of ^6Li is $6.9 \times 10^{22}/cm^3$. This correction is about 5 %.

Fairly good agreements between measurement and calculation have been obtained for T_7 and T_N, however, experimental data seem to be systematically lower than calculational results. The measured and calculated distributions for T_6 resemble each other. But differences between them are 10 ∿ 20 % and largish comparing the experimental errors of 5 ∿ 6 %.

To examine the discrepancies, the ratios of measured T_6 to T_7 are compared with those of calculations. They are shown in Fig. 6. The error bar in the figure means only random error. It is clear that the observed ratios agree well with the calculated ones. This result suggests that some systematic deviation might exist in the tritium counting system, especially in the conversion to the absolute values.

6. Concluding Remarks

(1) The tritium production-rate (TPR) distributions can be measured in various fusion blanket assemblies by using the Fusion Neutronics Source (FNS) facility. The series of the experiments on the simulated blanket assemblies based on lithium oxide will be continued.

(2) It has been demonstrated that the self-irradiation method using a LiF thermoluminescence dosimeter can obtain the TPR. The more works will be required to improve the sensitivity and to develop the calibration method for obtaining the absolute value.

(3) Liquid scintillation counting method using a sintered Li_2O pellet is very useful for our experiments because its chemical form is just the same as the breeding material.

(4) Experimental results support the Young's evaluation for $^7Li(n,n'\alpha)^3T$ cross section.

(5) Examination of the tritium counting system is in progress including the standard tritium samples.

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Fairly good agreements between measurement and calculation have been obtained for T_7 and T_N, however, experimental data seem to be systematically lower than calculational results. The measured and calculated distributions for T_6 resemble each other. But differences between them are 10 ∿ 20 % and largish comparing the experimental errors of 5 ∿ 6 %.

To examine the discrepancies, the ratios of measured T_6 to T_7 are compared with those of calculations. They are shown in Fig. 6. The error bar in the figure means only random error. It is clear that the observed ratios agree well with the calculated ones. This result suggests that some systematic deviation might exist in the tritium counting system, especially in the conversion to the absolute values.

6. Concluding Remarks

(1) The tritium production-rate (TPR) distributions can be measured in various fusion blanket assemblies by using the Fusion Neutronics Source (FNS) facility. The series of the experiments on the simulated blanket assemblies based on lithium oxide will be continued.

(2) It has been demonstrated that the self-irradiation method using a LiF thermoluminescence dosimeter can obtain the TPR. The more works will be required to improve the sensitivity and to develop the calibration method for obtaining the absolute value.

(3) Liquid scintillation counting method using a sintered Li_2O pellet is very useful for our experiments because its chemical form is just the same as the breeding material.

(4) Experimental results support the Young's evaluation for $^7Li(n,n'\alpha)^3T$ cross section.

(5) Examination of the tritium counting system is in progress including the standard tritium samples.

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Table 1 Comparison between Li_2O and Li_2CO_3 pellets

		Li_2O	Li_2CO_3	Li_2O / Li_2CO_3
10 cm	^7Li	4.555×10^{-29}	4.742×10^{-29}	0.961
	^6Li	1.469×10^{-28}	1.538×10^{-28}	0.955
	N_{Li}	5.588×10^{-29}	5.507×10^{-29}	1.015
20 cm	^7Li	1.377×10^{-29}	1.272×10^{-29}	1.083
	^6Li	8.346×10^{-29}	9.142×10^{-29}	0.913
	N_{Li}	1.878×10^{-29}	1.771×10^{-29}	1.060

* Unit : Tritium / Li atom · source neutron

Table 2 Nuclide densities in individual regions for Li_2O-C assembly

Nuclide	Nuclide density (10^{24} atoms/cm^3)			
	Void	Li_2O	Graphite	Lattice
^6Li		3.355×10^{-3}		
^7Li		4.1855×10^{-2}		
O		2.2605×10^{-2}		
C			6.9298×10^{-2}	
Mn	8.185×10^{-5}	9.632×10^{-5}	8.185×10^{-5}	5.632×10^{-5}
Fe	6.349×10^{-3}	7.030×10^{-3}	6.349×10^{-3}	4.159×10^{-3}
Ni	7.303×10^{-4}	8.106×10^{-4}	7.303×10^{-4}	4.821×10^{-4}
Cr	1.751×10^{-3}	1.935×10^{-3}	1.751×10^{-3}	1.161×10^{-3}

Table 3 Nuclide densities in individual regions for Li_2O slab assembly

Nuclide	Nuclide density (10^{24} atoms/cm^3)	
	Air	Li_2O
^6Li		4.152×10^{-3}
^7Li		5.190×10^{-2}
O	1.040×10^{-5}	2.803×10^{-2}
N	3.881×10^{-5}	
Mn		1.778×10^{-5}
Fe		1.262×10^{-3}
Ni		1.534×10^{-4}
Cr		3.473×10^{-4}

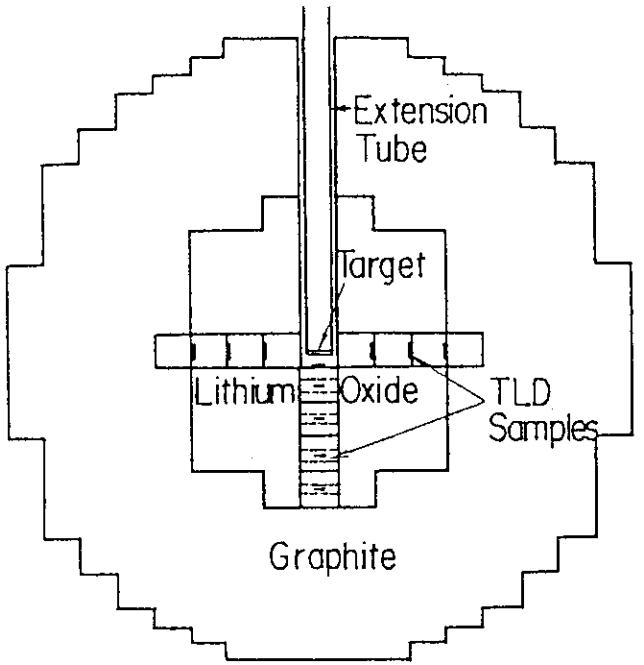

Fig. 1 Horizontal section across the center of Li_2O-C assembly

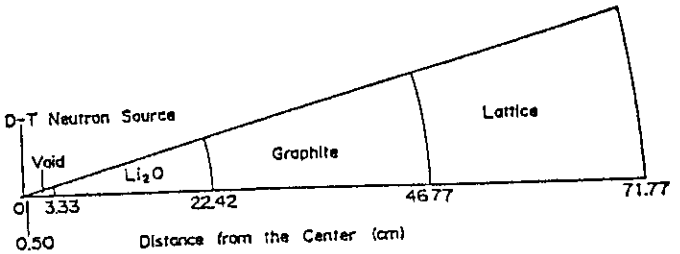

Fig. 2 Calculational model of Li_2O-C assembly

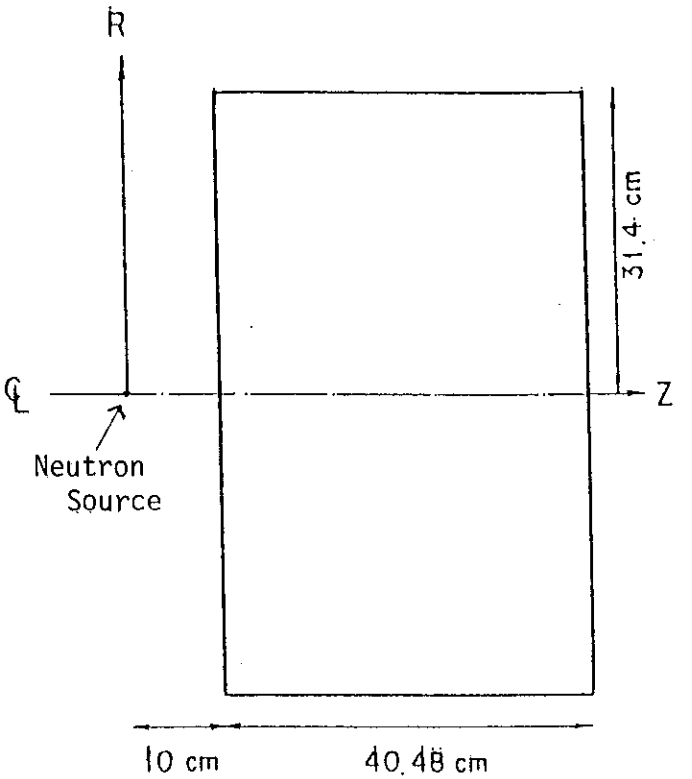

Fig. 3 Calculational model of 40 cm thick Li_2O slab assembly

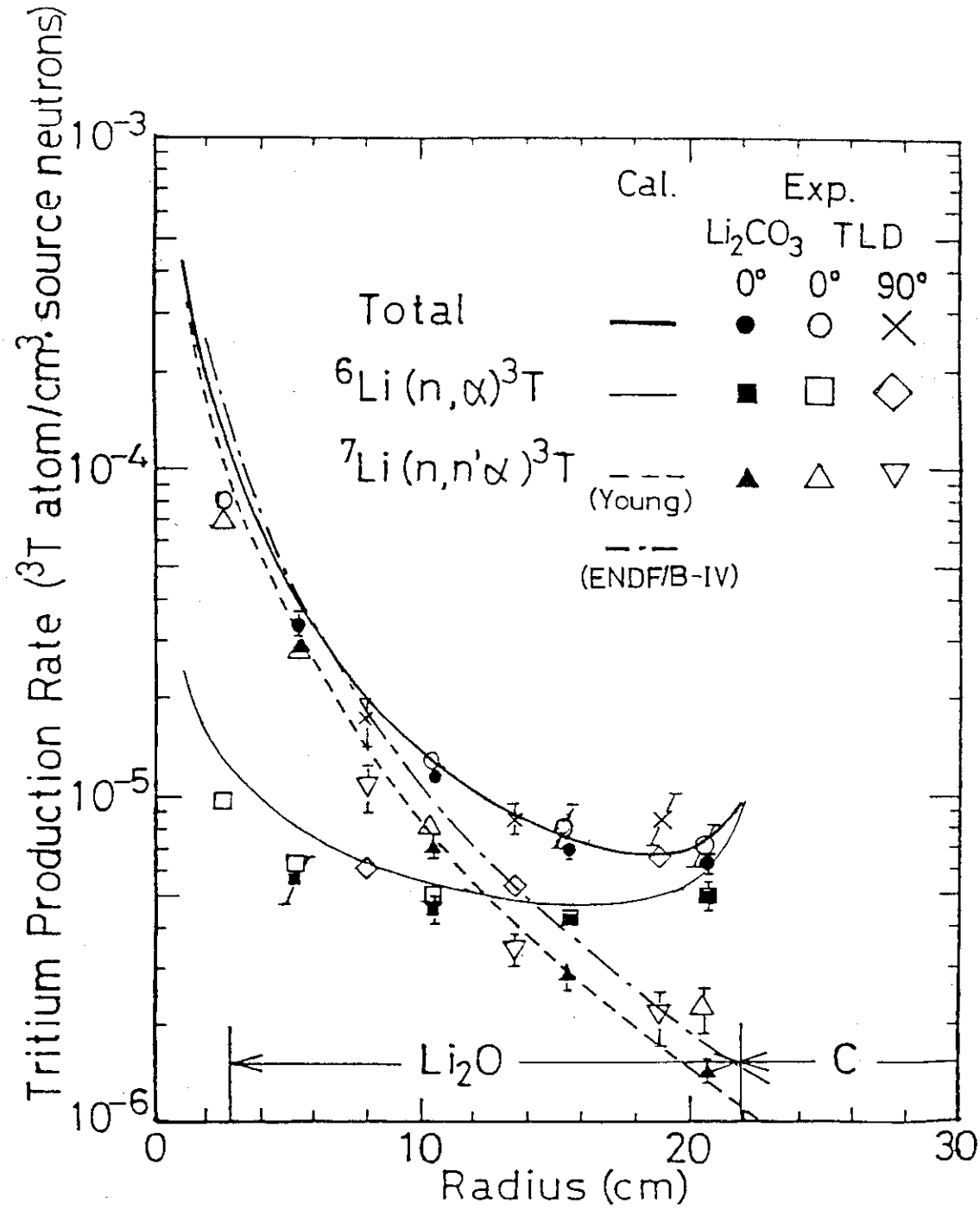

Fig. 4 Tritium production-rate distribution in Li_2O-C assembly

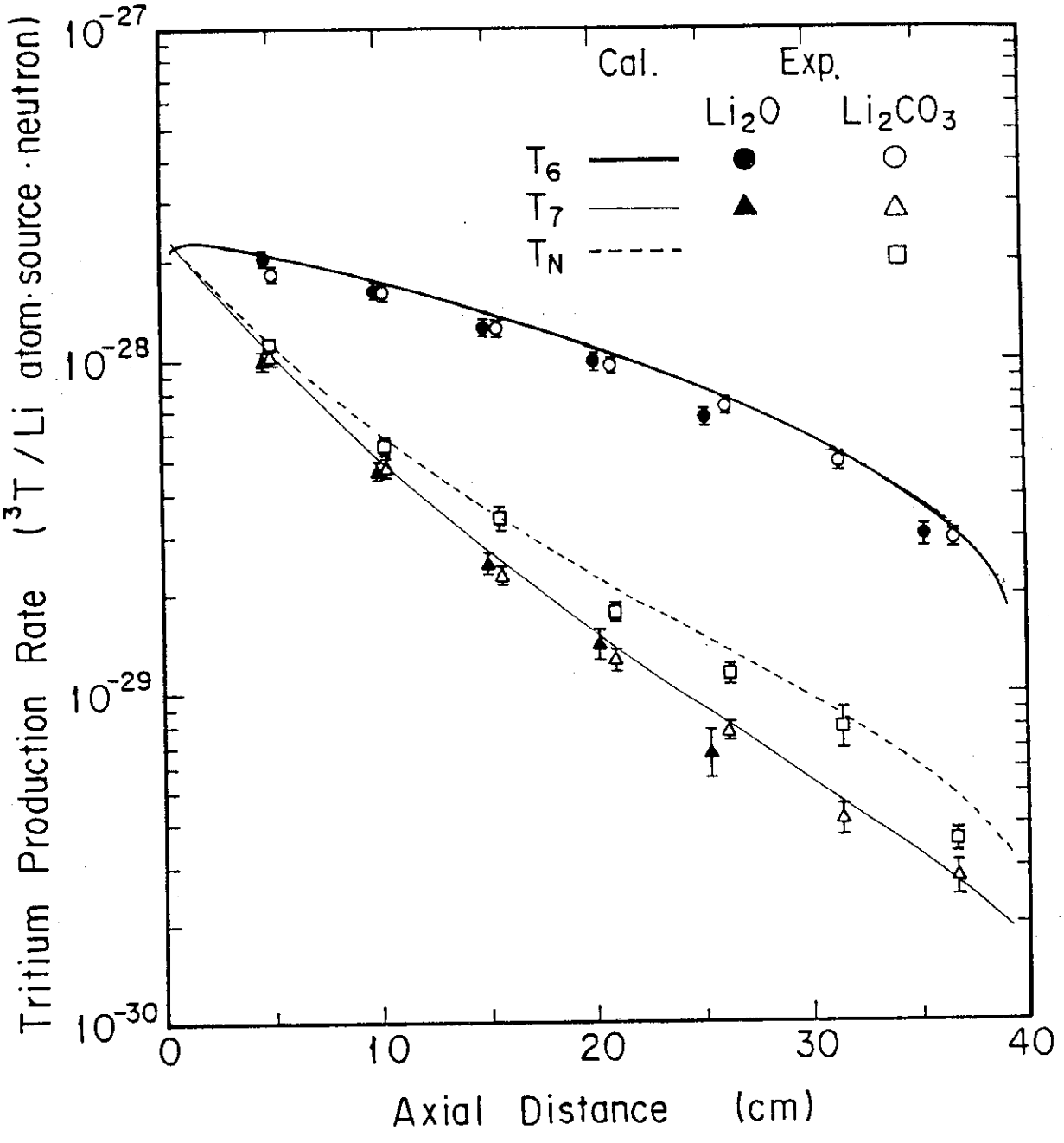

Fig. 5 Tritium production-rate distribution in 40 cm thick Li_2O slab assembly

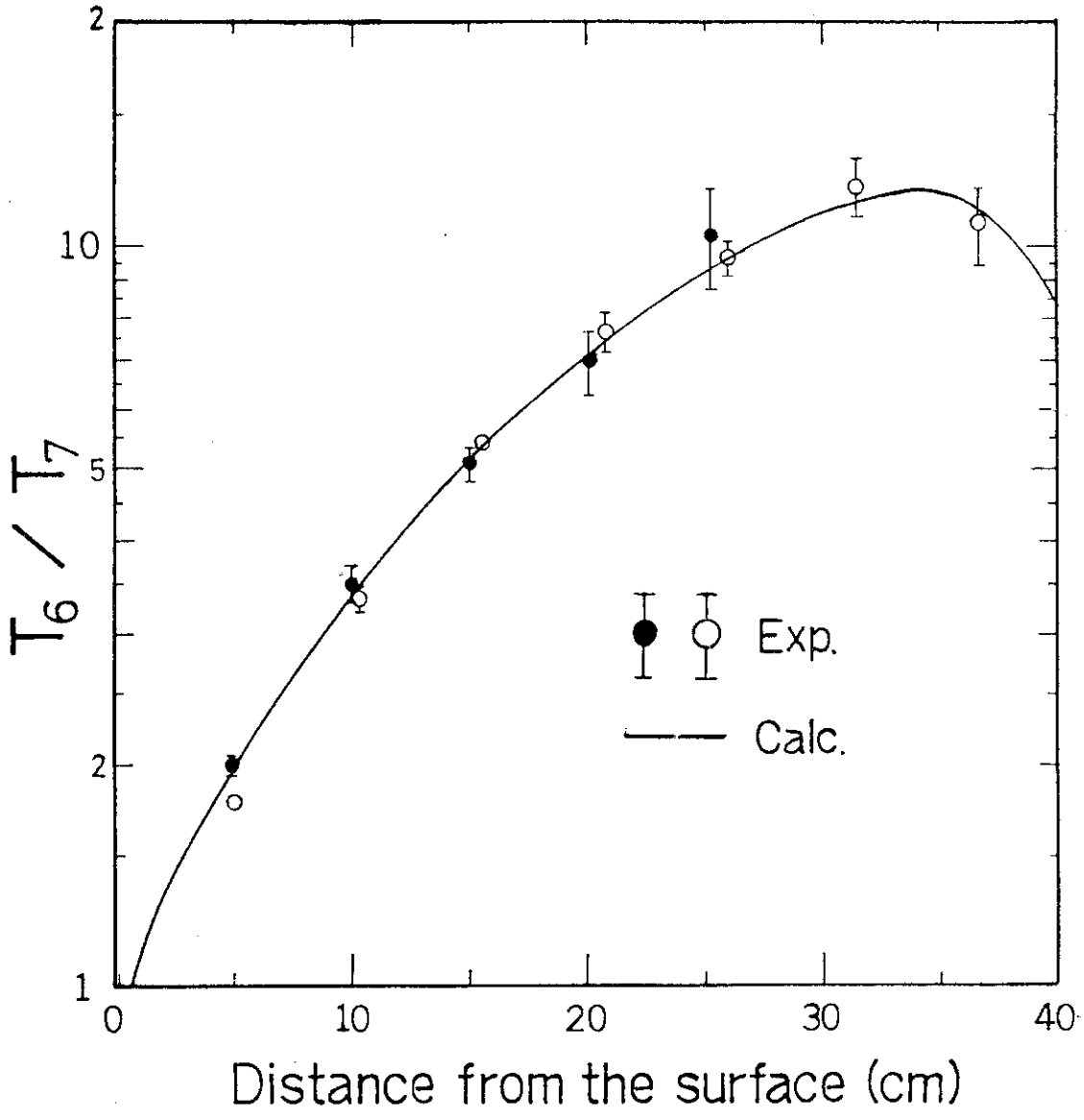

Fig. 6 T_6 / T_7 ratio distribution in 40 cm thick Li_2O slab assembly