BB

INSTITUTE FOR NUCLEAR STUDY
UNIVERSITY OF TOKYO
Tanashi, Tokyo 188
Japan

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Using p-Be and p-Li Neutrons up to $E_n = 38$ MeV

Seiichi Shibata, Tokushi Shibata, Mineo Imamura,
Tohru Ohkubo and Shingo Satoh
Institute for Nuclear Study, University of Tokyo
Tanashi, Tokyo 188, Japan

Yoshitomo Uwamino
Institute of Physical and Chemical Research
Wako, Saitama 351, Japan

Naotake Morikawa and Norio Nogawa
Radioisotope Centre, University of Tokyo
Bunkyo-ku, Tokyo 113, Japan

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Abstract

The production cross sections of tritium from oxygen have been measured in p-Be and p-Li neutron fields installed at the sector-focusing (SF) cyclotron of the Institute for Nuclear Study (INS), University of Tokyo. Irradiations were performed with the semi-monoenergetic neutrons produced from the $^9\text{Be}(p,n)^9\text{B}$ reaction at proton energies from 20 up to 40 MeV in 2.5-MeV steps, and those from $^7\text{Li}(p,n)^7\text{Be}$ in 5-MeV steps with proton intensities of 2-5 μA , which yields neutron fluxes from 10^7 to 10^8 n/cm²/sec at the position of sample irradiation. The neutron energy spectra were determined by an NE-213 scintillation counter. The tritium produced in the water used as the oxygen target was extracted by a distillation method, and measured by a liquid scintillation counter. The excitation functions for the $^{16}\text{O}(n,t)^{14}\text{N}$ reaction, measured by the p-Be and p-Li neutrons, agreed well with each other within the experimental uncertainties. The obtained results have been compared with the general trend in the (n,t) cross sections as a function of the atomic number of the target appearing in the literature.

1. Introduction

A number of experimental data of the neutron activation cross sections for incident energies of less than 20 MeV have been reported and are summarized in the literature [1,2]. On the other hand, those for energies higher than 20 MeV are quite insufficient, because of some difficulties in preparing a monoenergetic neutron source above 20 MeV. At present evaluated data files for the higher energies exist only for a few elements [3]. The importance of these cross sections has recently been increasing for cosmo-science [4], neutron damage studies, induced activation evaluation and accelerator shielding [5]. Among fast-neutron cross sections, data for the (n,t) reactions have systematically been surveyed in the interactions of 14.6 MeV neutrons with aluminum to bismuth [6,7] as well as of fast neutrons with a continuous energy spectra produced in the break-up of 30 and 53 MeV deuterons [8,9]. The general trend in the obtained (n,t) cross sections is a constantly decreasing function of the atomic number (Z) of the target in the region of $Z = 10-16$, and a leveling off at higher Z; the probability of triton emission is relatively independent of the target nucleus in the medium-to-heavy mass region. The (n,t) cross section for very light nuclei is predicted to be exceptionally large due to deuteron and triton cluster formation which facilitates direct reactions such as deuteron pick-up and triton knock-out.

An intense neutron beam course in the energy range from 20 to 40 MeV using the ${}^9\text{Be}(p,n){}^9\text{B}$ reaction was first installed at the INS SF cyclotron. However, the energy spectrum of the neutrons produced in this field is not purely monoenergetic; a high-energy peak from the ${}^9\text{Be}(p,n){}^9\text{B}$ reaction is quite broad with a low-energy tail coming from higher excited states of ${}^9\text{B}$, which requires the unfolding method to estimate the excitation function from the measured activation rates of irradiated samples. The application of an unfolding technique brought about an unavoidable ambiguity in determining the excitation functions. Therefore, the neutron beam course using the ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction was substituted for the one using ${}^9\text{Be}(p,n){}^9\text{B}$ in order to pursue better monochromaticity of the neutron energy spectrum. By using p-Li neutrons, it became possible to determine the reaction cross sections more accurately. We are now measuring the cross sections especially for reactions with higher threshold energies than ~ 15 MeV at the p-Li neutron field in INS.

In this paper we report on the excitation function for the ${}^{16}\text{O}(n,t){}^{14}\text{N}$ reaction measured at both the p-Be and the p-Li neutron fields. The obtained results were compared with the systematics of the cross sections for the (n,t) reactions obtained by the 14.6 MeV neutrons and the

deuteron break-up neutrons. The cross sections obtained in this work are indispensable from the view point of radiation safety, especially for estimating the tritium accumulation in the cooling water at accelerator facilities. Also, these data give valuable information concerning tritium production by secondary cosmic-ray neutrons in the upper level of the atmosphere together with data for the ${}^{14}\text{N}(n,t)$ reaction [10].

2. Experimental

2.1 p-Be neutron field

Since the target system for the p-Be neutron field used in this work has been published in detail by Uwamino et al. [11], it is briefly described here. The neutrons were produced by the interaction between protons with energies of 20-40 MeV at 2.5-MeV steps and a beryllium target with 1 mm thickness in the cases of $E_p = 20-37.5$ MeV or with 2 mm in $E_p = 40$ MeV. The incident protons lose a part of their energy at the beryllium target, and finally stop in the water coolant of the target.

The energy spectra of neutrons produced in the forward direction were measured by a $51\text{ mm}\phi \times 51\text{ mm}$ NE-213 liquid scintillation counter placed 1.1 m downstream from the beryllium target. The pulse-height distribution was converted to an energy spectrum by the unfolding method using a calculated response function. Fig. 1 shows an example of the neutron energy spectrum measured at $E_p = 40$ MeV. The energy dispersions observed in Fig. 1 are due to the energy loss of protons in the beryllium target, the existence of two reaction channels leading to ground and excited levels of ${}^9\text{B}$ and the natural line widths of the levels of ${}^9\text{B}$. The energy loss of incident protons was estimated to be 2.3 MeV (at $E_p = 37.5$ MeV) to 4.3 MeV (at 20 MeV) in 1 mm thick beryllium and 4.7 MeV (at 40 MeV) in 2 mm beryllium from the range-energy relation [12]. The spectra also include the background of the lower energy continuum, resulting from scattering and nuclear reactions in the water coolant used to stop the primary proton beam. The proton beam current during irradiation was monitored by a Faraday-cup connected to a digital current integrator. The absolute intensity of the neutron flux for each irradiation was checked by comparing the calculated radioactivities based on the neutron spectra with the observed values for the monitor reaction, ${}^{27}\text{Al}(n,\alpha){}^{24}\text{Na}$, of which cross sections were taken from Ref. [1]. The neutron fluxes obtained by the current integrator agreed with those calculated using the monitor

reaction within $\pm 10\%$ at $E_p \leq 30$ MeV and $\pm 20\%$ at the higher energies.

2.2 p-Li neutron field

The target system of the p-Li neutron field comprised a 2 mm thick Li (${}^7\text{Li}$ 99.98 % enriched) target backed by a 12 mm thick carbon (graphite) used as a beam stopper for the primary protons, as shown in Fig. 2. The lithium target was fixed in an aluminum target holder, which enabled the target to slide up and down manually. This made it possible to irradiate samples using two types of the target systems: one was a lithium-plus-carbon target; the other was an only carbon target. The proton beam current was monitored with a digital current integrator, as in the case of the p-Be field. The target system was cooled by blowing air to the target chamber during irradiation.

The neutron spectrum from this target system was measured at the time-of-flight (TOF) facility at the Cyclotron and Radioisotope Center (CYRIC) of Tohoku University, since no beam-chopping system for the TOF measurement exists in the cyclotron facility at INS. A 127 mm ϕ x 127 mm NE-213 scintillator was placed about 10 m away from the target.

The neutron spectrum produced from the lithium target (Φ_{Li}) was estimated from the difference of the two spectra of $\Phi_{\text{Li+C}}$ and Φ_{C} produced from the Li+C and C target systems, respectively,

$$\Phi_{\text{Li}}(E) = \Phi_{\text{Li+C}}(E) - \Phi_{\text{C}}(E). \quad (1)$$

The monoenergetic peak neutrons produced by ${}^7\text{Li}(p,n)$ correspond to the reaction rate for the production of the ground and the first excited (0.43 MeV) states of ${}^7\text{Be}$, because the higher excited states of ${}^7\text{Be}$ decay through proton and alpha-particle emissions. Therefore, the absolute value of the monoenergetic peak neutron spectrum at 0° ($\Phi(E_p)$), which is needed to estimate production cross section, can be obtained from the ${}^7\text{Be}$ yield produced by ${}^7\text{Li}(p,n)$ at 0° as follows:

$$\Phi(E_p) = N({}^7\text{Li})I_p \left(\frac{d\sigma}{d\Omega} \right)_{\theta=0}, \quad (2)$$

where $N({}^7\text{Li})$ is the number of ${}^7\text{Li}$ atoms in the target, I_p the number of incident protons and $(d\sigma/d\Omega)_{\theta=0}$ the differential cross section of ${}^7\text{Li}(p,n)$ in the monoenergetic peak at 0° . Also, the number of ${}^7\text{Be}$ produced from ${}^7\text{Li}(p,n)$ ($N({}^7\text{Be})$) is given by

$$N({}^7\text{Be}) = N({}^7\text{Li})I_p\sigma. \quad (3)$$

Here, σ is the total cross section of ${}^7\text{Li}(p,n){}^7\text{Be}$, that is

$$\sigma = \int_{4\pi} \left(\frac{d\sigma}{d\Omega} \right) d\Omega. \quad (4)$$

By combining Eqs. (2) and (3), the peak neutron yield at 0° can be obtained as

$$\Phi(E_p) = N({}^7\text{Be}) \left(\frac{d\sigma}{d\Omega} \right)_{\theta=0} / \sigma. \quad (5)$$

The relative angular distribution of the peak neutrons $((d\sigma/d\Omega)_{\theta=0}/\sigma)$ was measured by rotating the angle of the incident proton beam from 0 to 125° at $E_p = 20$ –40 MeV using the beam swinger system at CYRIC. The radioactivity of ${}^7\text{Be}$ produced in the lithium target was measured by gamma-spectrometry with a Ge detector.

Fig. 3(a) shows two neutron spectra from the Li+C and the C targets measured at $E_p = 40$ MeV, and (b) the spectrum for the lithium target obtained by the difference of the two spectra in Fig. 3(a). The monoenergetic peak neutrons having ~ 2 MeV of the full width at half-maximum (FWHM) belong to the ground and the first 0.43 MeV excited states of ${}^7\text{Be}$. The energy dispersion for the peak neutrons is almost attributed to the energy loss of incident protons with the lithium target thickness. The contribution of the peak neutron yields from the C(p,n) reaction is negligibly small, because the threshold energy of the ${}^{12}\text{C}(p,n)$ reaction is 19.6 MeV and the abundance of ${}^{13}\text{C}$ is only 1.1%. The threshold energy of ${}^{13}\text{C}(p,n)$ is 3.23 MeV.

The normalization of the neutron spectrum measured at CYRIC to the neutron flux during sample irradiation for the activation at INS was done using the radioactivity of ${}^7\text{Be}$ produced in the lithium target. The details concerning the p-Li neutron field have been reported in Refs.[13,14,15].

2.3 Irradiation

About 3g of purified water used as the oxygen target in an acrylic container of 2 cm ϕ x 1 cm was irradiated with aluminum beam monitors by p-Be neutrons at $E_p = 20$ –40 MeV in 2.5-MeV steps and by p-Li ones in 5-MeV steps. The samples were placed 20 cm away from the beryllium target and 15 cm from the lithium, respectively. The typical irradiation time was 5 hours with a proton beam intensity of $\sim 2 \mu\text{A}$. Under this irradiation condition, the neutron fluxes were 10^7 – 10^8 n/cm²/sec in both neutron fields.

2.4 Sample preparation for liquid scintillation counting

The tritium produced in the water was extracted by a distillation method using the device shown

in Fig. 4 [16]. The irradiated sample was transferred to flask-I and frozen by liquid nitrogen. After connecting to an empty flask-II, it was evacuated to 10 Pa while being kept in the frozen state by liquid nitrogen. After evacuation, flask-II was soaked in liquid nitrogen while flask-I was heated by an infrared lamp. The frozen water in flask-I was then sublimated and trapped in flask-II. The water in flask-II was transferred into a 20 ml glass vial and mixed with the scintillator, PACKARD PICO-FLUOR LLT, which was specially developed for counting samples containing low-level tritium radioactivity. The tritium measurements were performed using the liquid scintillation counter, PACKARD 2000CA/LL, at the Radioisotope Centre, University of Tokyo. The typical counting time was 500 min.

3. Results and Discussion

The average cross section (σ_{E_p}) for the energy range of the peak neutrons is calculated as follows. The reaction rate R is given by

$$R = N \int_{E_{th}}^{E_{max}} \sigma(E) \Phi(E) dE, \quad (6)$$

where N is the number of target atoms, E_{th} the threshold energy of the reaction, E_{max} the maximum energy of monoenergetic peak neutrons, $\sigma(E)$ the activation cross section and $\Phi(E)$ the neutron energy spectrum. Since the threshold energy of the $^{16}\text{O}(n,t)$ reaction (15.4 MeV) is lower than the minimum energy of peak neutrons (E_{min}) produced at the incident proton energy region of 20–40 MeV, R consists of two parts; one is induced by the peak energy neutrons and the other by the low-energy neutrons:

$$R = N \int_{E_{th}}^{E_{min}} \sigma(E) \Phi(E) dE + N \sigma_{E_p} \Phi_{E_p}, \quad (7)$$

where Φ_{E_p} is the neutron flux at peak neutron energy. Then, σ_{E_p} is calculated using

$$\sigma_{E_p} = \frac{R - N \int_{E_{th}}^{E_{min}} \sigma(E) \Phi(E) dE}{N \Phi_{E_p}}. \quad (8)$$

Since Φ_{E_p} can be obtained from Eq. (5), we must determine the second integration term in the numerator. The calculation was performed by a simple iterative procedure starting from the lowest energy, moving to a higher energy and repeating the process until the value for σ_{E_p} became constant.

The results for tritium production at the p-Be and p-Li neutron fields are summarized in Tables 1 and 2, respectively. The excitation functions of $^{16}\text{O}(n,t)^{14}\text{N}$ obtained for both neutron fields are shown in Fig. 5. The two results agreed with each other within the experimental uncertainties, although the one at $E_n \approx 30$ MeV, obtained in the p-Be field, is apparently lower than the expected value. The cross sections increase from the threshold energy and gradually decrease after the peak position (~ 13 mb) around $E_n = 25$ MeV. The obtained excitation function in $20 \text{ MeV} < E_n < 25 \text{ MeV}$ is almost the same in shape as those in $15 \text{ MeV} < E_n < 20 \text{ MeV}$ of other (n,t) reactions appearing in Ref. [17]. There are no data for the tritium production from oxygen, except for the integral cross-section of 8.5 ± 1.5 mb, measured by 53-MeV deuteron break-up neutrons by Qaim and Wölffe [9], because of the higher threshold energy of the $^{16}\text{O}(n,t)$ reaction. By considering the break-up neutron energy spectrum, in which the maximum intensity occurs at ~ 22 MeV and FWHM is 15.8 MeV, their cross section seems to be consistent with our results (also plotted in Fig. 5).

Fig. 6 shows the trend of the tritium production cross sections around $E_n = 20$ MeV as a function of the atomic number (Z) of the target element. The tritium formation cross sections decrease with an increase of target Z in the region of $4 \leq Z \leq 25$, and are almost constant at higher Z . For the $^{16}\text{O}(n,t)$ reaction, the cross-section value at $E_n = 25$ MeV is plotted in this figure, since the threshold energy of the $^{16}\text{O}(n,t)$ is ~ 5 MeV higher than those of (n,t) reactions from Be, Al, V and Co targets. The cross section at $E_n \approx 15$ MeV is shown in the figure for the $^{14}\text{N}(n,t)$ reaction [10] for a similar reason to that mentioned above; the threshold energy of this reaction is 5 MeV smaller than other (n,t) reactions. The plotted data from Be to Co targets correspond to the cross sections at a neutron energy of $\sim (E_{th} + 10)$ MeV. Our result for $^{16}\text{O}(n,t)$ is fitted well with the value predicted from the systematics of (n,t) reactions. Conclusively, it is noted that the (n,t) cross sections for light elements are exceptionally large probably due to direct interactions, and that the probability of emission of a triton is relatively independent of the target nucleus in the medium- and heavy-mass regions [9,17].

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References

1. Mclane, V., Dunford, C. L., Rose, P. F.: *Neutron Cross Sections*, (Academic Press, 1988).
2. Manokhin, V. N., Pashchenko, A. B., Plyaskin, V. I., Bychkov, V. M., Pronyaev, V. G.: *Handbook on Nuclear Activation Data*, Technical Reports Series No. 273, 305-411 (IAEA, 1987).
3. ENDF/B-VI (Evaluated Nuclear Data File, Version 6), National Nuclear Data Center, Brookhaven National Laboratory (1993).
4. Imamura, M., Nagai, H., Takabatake, M., Shibata, S., Kobayashi, K., Yoshida, K., Ohashi, H., Uwamino, Y., Nakamura, T.: Nucl. Instr. and Meth. B52, 595-600 (1990).
5. Uwamino, Y., Sugita, H., Kondo, Y., Nakamura, T.: Nucl. Sci. Eng. 111, 391-403 (1992).
6. Qaim, S. M., Stöcklin, G.: Nucl. Phys. A257, 233-241 (1976).
7. Sudár, S., Csikai, J.: Nucl. Phys. A319, 157-164 (1979).
8. Wölfe, R., Khatun, S., Qaim, S. M.: Nucl. Phys. A423, 130-138 (1984).
9. Qaim, S. M., Wölfe, R.: Nucl. Phys. A295, 150-162 (1978).
10. Suhaimi, A., Wölfe, R., Qaim, S. M., Warwick, P., Stöcklin, G.: Radiochim. Acta 43, 133-138 (1988).
11. Uwamino, Y., Ohkubo, T., Torii, A., Nakamura, T.: Nucl. Instr. and Meth. A271, 546-552 (1988).
12. Janni, J. F.: Atomic Data and Nucl. Data Tables 27, 147-339 (1982).
13. Soewarsono, T. S.: Thesis, Tohoku Univ. (1993).
14. Nakamura, T., Soewarsono, T. S., Uno, Y., Nakao, N., Uwamino, Y., Imamura, M., Ishikawa, T., Tanaka, S., Nakashima, H., Tanaka, S., Baba, M., Iwasaki, T.: *Proc. of 8th Int. Conf. Radiation Shielding*, (American Nuclear Society, Inc., 1995) p.264-p.271.
15. Uwamino, Y., Sugita, H., Soewarsono, T. S., Kondo, Y., Nakamura, T.: *Proc. of Specialists' Meeting on Neutron Cross Section Standards for the Energy Region above 20 MeV*, (The Svedberg Laboratory, Uppsala and OECD Nuclear Energy Agency, 1991) p.221-p.232.
16. Shibata, S., Kawakami, H., Kato, S., Nisimura, K., Ohshima, T., Sugai, I., Ukai, K., Yasuda, T., Morikawa, N., Nogawa, N., Haga, K., Shoji, Y., Nagafuchi, T., Naito, F., Suzuki, T., Shigeta, M., Iwahashi, M., Hisatake, K., Fujioka, M., Matsuda, T.: Nucl. Instr. and Meth. A350, 263-274 (1994).
17. Wölfe, R., Qaim, S. M., Liskien, H., Widera, R.: Radiochim. Acta 50, 5-9 (1990).

Figure Captions

Fig. 1 Neutron energy spectrum at a p-Be neutron field ($E_p=40$ MeV).

Fig. 2 Target system for a p-Li neutron field.

Fig. 3 (a) Neutron energy spectra obtained from Li+C and C targets ($E_p=40$ MeV). (b) Neutron energy spectrum for Li target ($E_p=40$ MeV), the difference of the spectra from Li+C and C targets.

Fig. 4 Device for evaporating an irradiated water sample. See the text for details.

Fig. 5 Excitation function for $^{16}\text{O}(n,t)^{14}\text{N}$ reaction. ($\square$): p-Be neutron, ($\circ$): p-Li neutron, and ($\triangle$): deuteron break-up neutron with continuous energy spectrum (FWHM=15.8 MeV) [9].

Fig. 6 Trend in the tritium formation cross section at $E_n \approx 20$ MeV. ($\circ$): this work, ($\bullet$): [17], ($\triangle$): [10]. The solid curve is to guide the eye.

Table 1. Cross sections for $^{16}\text{O}(n,t)$ reaction obtained at p-Be neutron fields

E_p (MeV)	Proton intensity (Coulomb)	Peak neutron yield ^{a)} ($10^{12} \text{ n/cm}^2/\text{Coulomb}$)	Tritium produced ^{b)} (dpm/sample)	Cross section (mb)	$E_n^c)$ (MeV)
22.5	0.0465	5.56	4.69 ± 2.31	1.76 ± 0.87	15 - 21
25	0.0680	5.59	20.55 ± 1.20	7.35 ± 2.80	21 - 24
27.5	0.0518	7.15	30.35 ± 1.79	11.2 ± 2.2	24 - 26
30	0.0812	8.23	88.10 ± 3.90	13.5 ± 2.6	26 - 29
32.5	0.0428	8.09	45.21 ± 2.11	7.45 ± 1.27	29 - 31
35	0.0346	6.02	52.16 ± 1.68	12.3 ± 2.3	31 - 34
37.5	0.0201	7.58	27.70 ± 1.89	8.36 ± 1.65	34 - 36
40	0.0406	18.7	129.7 ± 3.5	7.66 ± 1.49	36 - 39

a) Peak neutron yield at 20 cm from the Be target.

b) Corrected for the blank.

c) Energy range for which cross section is evaluated.

Table 2. Cross sections for $^{16}\text{O}(n,t)$ reaction obtained at p-Li neutron fields

E_p (MeV)	Target ^{a)}	Proton intensity (Coulomb)	Peak neutron yield ^{b)} ($10^{12} \text{ n/cm}^2/\text{Coulomb}$)	Tritium produced ^{c)} (dpm/sample)	Cross section ^{d)} (mb)	E_n ^{e)} (MeV)
20	Li + C	0.0460	3.805	3.86 ± 0.41	0.88 ± 0.31	15.6 - 19.3
	C	0.0364	—	1.68 ± 0.36		
25	Li + C	0.0508	4.376	11.49 ± 0.79	4.12 ± 0.66	19.9 - 23.4
	C	0.0395	—	0.72 ± 0.76		
30	Li + C	0.0586	5.418	51.81 ± 3.31	12.8 ± 1.8	25.7 - 29.3
	C	0.0599	—	3.13 ± 2.76		
35	Li + C	0.0206	5.382	26.56 ± 0.84	12.9 ± 1.8	31.0 - 34.4
	C	0.0199	—	5.51 ± 0.78		
40	Li + C	0.0358	7.080	55.68 ± 0.92	10.5 ± 1.6	36.4 - 39.8
	C	0.0273	—	5.42 ± 0.38		

a) Target system for neutron production.

b) Peak neutron yield at 15 cm from the Li target.

c) Corrected for blank.

d) Cross section is calculated from the difference of the tritium produced from Li+C and C targets after normalizing the proton beam intensity.

e) Energy range for which cross section is evaluated.

Fig. 1

Fig. 2

Fig. 3 (a)

Fig. 3 (b)

Fig. 4

Fig. 5

Fig. 6