Techniques to Measure Absolute Neutron Spectrum and Intensity for Accelerator Based Neutron Source for BNCT*)

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Boron Neutron Capture Therapy (BNCT) is a new and promising cancer therapy. At present, development of accelerator based neutron source (ABNS) is underway to be utilized as a neutron source instead of nuclear reactor. However, it is known that the neutron fields formed with accelerators have different characteristics depending on kind of accelerators. It means we have to characterize the field before practical use. In the authors' group, various neutronics characterization devices have been developed for our BNCT machine named CSePT. It includes source intensity, epi-thermal neutron flux, neutron spectrum and so on. In the present paper, such measuring techniques were overviewed.

Keywords: BNCT, neutron spectrum, epi-thermal neutron, ABNS, foil activation, proportional counter

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1. Introduction

Boron Neutron Capture Therapy (BNCT) is known to be a new cancer treatment for the next generation. Administering ^{10}B including compound, ^{10}B is accumulated in tumor cells. Neutrons are then irradiated from outside of a human body to induce a neutron-^{10}B reaction, $n + {}^{10}B \rightarrow \alpha + {}^{7}Li$. The emitted charged particles from the reaction have energy of around 1 MeV, meaning the range is ~10 μm in tissues. This is more-or-less the same as the size of a human body cell. As a result, if ^{10}B is accumulated only in tumor cells, only the tumor cells can be killed. BNCT can be regarded as a cell selective cancer therapy with charged particles.

BNCT was carried out in nuclear reactors as a clinical test so far. In recent years, epi-thermal neutrons (0.5 eV ~ 10 keV) are used instead of thermal neutrons. Since then BNCT is recognized as a less invasive therapy among cancer therapies. On the other hand, there is a critical problem, i.e., BNCT requires an intense neutron source and only two neutron sources, i.e., nuclear reactors of KUR, Kyoto Univ. and JRR-4, JAEA, were used as the neutron source. However, especially in Japan it is prohibited to construct a nuclear reactor in or close to a hospital. Under these circumstances, Japan is aiming at establishment of accelerator based neutron source (ABNS) for BNCT instead of the nuclear reactor, because ABNS can be constructed in hospitals. However, neutronics characterization of ABNS is known to be difficult, because the radiation field varies depending on kinds of accelerator and moderator. As for the previous studies for this problem, basic principles are summarized in Sauerwein's text book for BNCT [1]. The latest related researches are found in the Conference Proceedings of the International Conference on Neutron Capture Therapy (ICNCT) [2]. However, there are no established techniques available in ABNS for BNCT.

In this paper, some new measuring techniques listed below for characterization of our p-Li based ABNS-BNCT, named CSePT (Cell Selective Particle Therapy) are described, which we have been developing since 2007.

- Source neutron intensity monitor (Chap. 2)
- Epi-thermal neutron flux intensity monitor (Chap. 3)
- Neutron spectrometer (Chap. 4)

These measuring techniques would be useful in various ABNS based BNCT facilities in future. Meanwhile, in the present studies, experiments were very essential and we confirmed validity of all the techniques above experimentally in neutron source facilities. In addition, for the theoretical aspect numerical simulations are also very crucial for evaluating the techniques to compare with the experimental results. For that purpose, we utilized a general Monte Carlo N-Particle Transport Code (MCNP5) [3]. MCNP5 supported our pre-analyses and post-analyses of all the series experiments.

2. Source Neutron Intensity Monitor for p-Li Reaction with Activation Foil [4]

The ABNS-BNCT, CSePT, utilizes p-Li reaction, $p + {}^{7}Li \rightarrow n + {}^{7}Be - 1.64\,MeV$. In BNCT, ~2.5 MeV pro-

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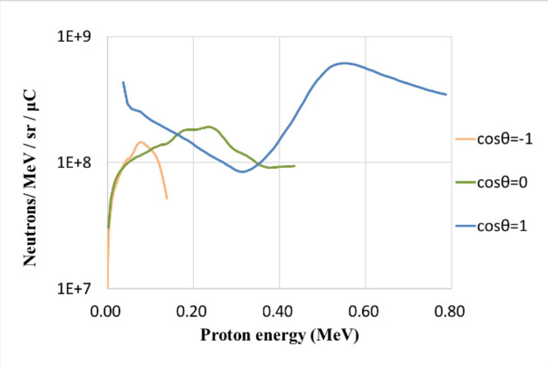

Fig. 1 p-Li neutron spectra for various emission angles cited from DROSG-2000 [5].

tons are usually bombarded to a metal lithium target to produce neutrons. Figure 1 shows the neutron energy spectra as a function of emission angle cited from DROSG-2000, IAEA [5]. The neutron energy spectrum shows complex, i.e., the energy is lower in backward angles and higher in forward angles. In the authors' group, an easy and convenient technique to measure the absolute source neutron intensity is under development. The energy of neutrons to measure ranges from several tens to around 800 keV. However, it is generally known to be difficult to measure such energetic neutrons. In this study, the foil activation method was employed to apply to an intense neutron field characterization. However, for the neutron energy range, very few activation foils are known to be available. If it is in eV region, neutron capture reaction is available. If it is in MeV or higher region, threshold reactions like an (n, p) reaction are suitable. The problem is that several tens to 800 keV are in-between capture and threshold reactions. So, we focused on isomer production reaction induced by inelastic scattering. If a certain excited state of a nuclide has its own half-life, it could in principle be utilized as an activation foil. Now, assuming the excitation energy is E_{ex} and if the incident neutron energy is larger than E_{ex}, the nuclide may be excited and have a sensitivity to the neutron. In addition, the emitted gamma-ray energy is exactly the same as E_{ex}. If the E_{ex} of the nuclide is within several tens to 800 keV corresponding to the p-Li reaction, the nuclide can be used and the emitted gamma-rays can be measured with a germanium semiconductor detector easily.

According to Table of Isotopes [6], there exist over 20 nuclides which can create an isomer by p-Li neutrons. Among them, taking into account their half-lives, threshold energies, reaction cross sections and energy dependence, we finally selected three candidate nuclides of ^{107}Ag, ^{115}In, ^{189}Os. Table 1 summarizes their basic feature.

Before the practical use, we have to confirm the validity by experimental tests. We carried out irradiation experiments at Fast Neutron Laboratory (FNL) of Tohoku University, Japan. The FNL facility has a dynamitron accelerator. Using the accelerator, applicability of ^{107}Ag and ^{115}In foils was confirmed by p-Li neutron irradiation experiments. Practically, we determined experimentally the p-Li source intensity from the measured activities of the monitor foils, that is, ^{7}Be activity of the target produced by ^{7}Li(p, n)^{7}Be reaction [7]. The features of the candidate activation foils are summarized as follows:

Table 1 Isomeric description of ^{107}Ag, ^{115}In and ^{189}Os.

Nuclide	E γ (keV)	Half-life
^{107}Ag	93.13	44.3 sec
^{115}In	336.24	4.48 hrs
^{189}Os	30.814	5.8 hrs

^{107}Ag: Because of its short half-life, ^{107}Ag is the most convenient monitor for an intense neutron source of the real p-Li based ABNS-BNCT machine. We should notice however that the activity decays very rapidly after irradiation.

^{115}In: ^{115}In can be the most accurate monitor, because the abundance is large and the emitted gamma-ray energy is fairly large compared to other two.

^{189}Os: ^{189}Os is the best monitor for measuring in the backward angle with respect to the proton beam, because the cross section in low energy region is large. However, there are some problems left: 1) Difficult to prepare a thin sample. 2) Osmium has toxicity. 3) ^{189}Os should be enriched because stable isotope of ^{188}Os having abundance of 13.2% produces ^{189}Os by (n, γ) reaction.

3. Epi-Thermal Neutron Flux Intensity Monitor with Activation Foil [8, 9]

In BNCT, epi-thermal neutrons are commonly used except surface cancers. Epi-thermal neutrons can be obtained in nuclear reactors, and will be produced by ABNS in near future. In ABNS, the neutron spectrum changes depending on the target material, kind of incident particle and its energy and so on. It means epi-thermal neutron fields are not exactly the same with each other. We hence have to characterize the epi-thermal neutron field before practical use. The characteristics of the field include mainly the energy spectrum and absolute flux intensity. In this chapter, the technique to measure the absolute epi-thermal neutron flux intensity is detailed.

For the above purpose, we adopted the foil activation method. Generally, the cross section of the activation foil has its own energy dependence. It indicates, if the energy spectrum is not known in a neutron field, we cannot estimate the absolute intensity in the field. We thus attach some neutron filter to the activation foil so as to make the sensitivity of the foil equal in the whole neutron energy range.

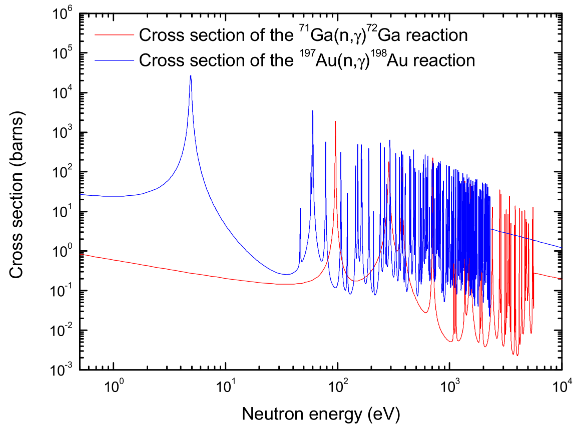

Fig. 2 Capture cross section of gold and gallium [11].

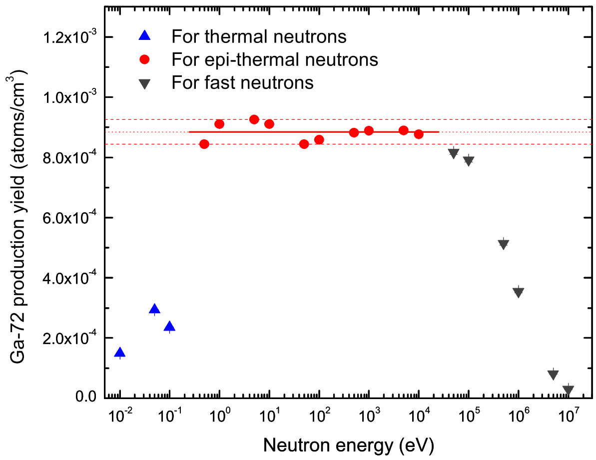

Fig. 3 Designed sensitivity of ^{71}Ga(n, γ) reaction.

In the lower energy region, gold foil is commonly used as a neutron monitor with ^{197}Au(n, γ)^{198}Au reaction. Since this reaction has a large resonance at 4.9 eV, the gold foil is very suitable for thermal neutron measurement. However, in the epi-thermal energy region, the sensitivity becomes lower. A usual way to solve this problem is to employ a neutron moderator of like polyethylene arranged surrounding the gold foil in order to moderate fast neutrons and to make the foil absorb the slowing down neutrons. In the present design, we used a spherical polyethylene as moderator to avoid angle dependence of incident neutrons. On the other hand, to remove incident thermal neutrons, the monitor body of polyethylene is covered with a cadmium sheet. However, due to this cadmium sheet, lower energy neutrons in the epi-thermal neutron energy region are also absorbed and removed unexpectedly. To avoid this phenomenon, it is essential to make the cadmium sheet thin. However, gold has a large resonance at 4.9 eV. The energy dependent sensitivity curve is deformed accordingly if it is too thin.

For these reasons, we next examined other seven candidate activation foils, ^{151}Eu, ^{127}I, ^{115}In, ^{71}Ga, ^{55}Mn, ^{37}Cl and ^{23}Na, which can be activated by epi-thermal neutrons and the half-lives and emitted gamma-ray energies of which are appropriate. As the result of examination, it was found that ^{71}Ga showed the flattest sensitivity. In Fig. 2 the cross section curve is described together with that of ^{197}Au. The point to choose ^{71}Ga is that the capture reaction cross section shows not so rapid change in the lower energy region and has no wide and big resonances in epi-thermal energy region. As for the dimensions of the monitor, the diameter of the spherical polyethylene was finally fixed to be 7.1 cm and the outside cadmium sheet thickness was 0.05 mm. The designed sensitivity calculated by MCNP5 is shown in Fig. 3. The sensitivity fluctuation is suppressed within 5% in the epi-thermal energy region as in the figure. In practical applications, however, because the melting point of gallium is very low, i.e., as low as 30°C, we decided to use GaN, though it is a little expensive.

After the design, to confirm the performance of the present monitor, we made a prototype monitor and carried out an irradiation experiment at OKTAVIAN facility of Osaka University, Japan. The OKTAVIAN is a DT neutron source for fusion neutronics studies. At first we developed an epi-thermal neutron field with the DT neutron source combined with lead neutron multiplier and graphite moderator. Then irradiation experiments were performed to activate ^{71}Ga via (n, γ) reaction in the monitor. The activity of ^{72}Ga was measured after the irradiation and compared with the calculation result to discuss the validity of the monitor. As a result, the agreement was acceptable to show that the monitor would be utilized in real scenes of ABNS-BNCT [10].

4. Epi-Thermal Neutron Spectrometer with Proportional Counter [12, 13]

Measurement of neutron spectrum in the epi-thermal region is difficult. Generally, Bonner sphere, multi-foil method and so on are utilized [14]. With the Bonner sphere, several-time measurements are required with several neutron moderators arranged surrounding the Bonner detector. In this case, each Bonner detector with a different thick moderator has a different sensitivity to a given neutron spectrum. With several measured results, the neutron spectrum can be estimated by solving a so-called inverse problem with an appropriately evaluated detector response for neutron energy. In case of the multi-foil method, several kinds of foils are used, which have different sensitivities for neutron energy to produce radioactive isotopes. By combining several suitable foils covering their own sensitive neutron energy in a neutron field of interest, the neutron spectrum can be estimated by measuring their activities after neutron irradiation. These techniques are already established, however problems are known, i.e., the energy

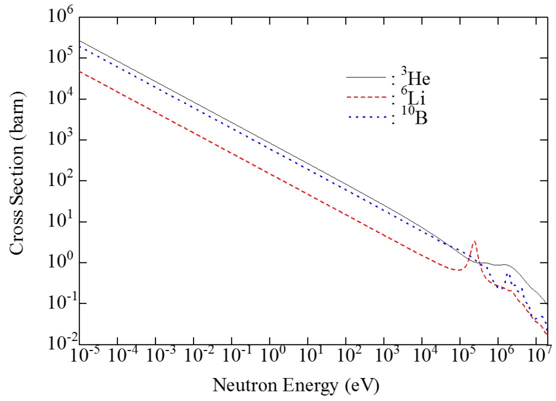

Fig. 4 Charged particle emission reaction of ^{3}He, ^{6}Li and ^{10}B [11].

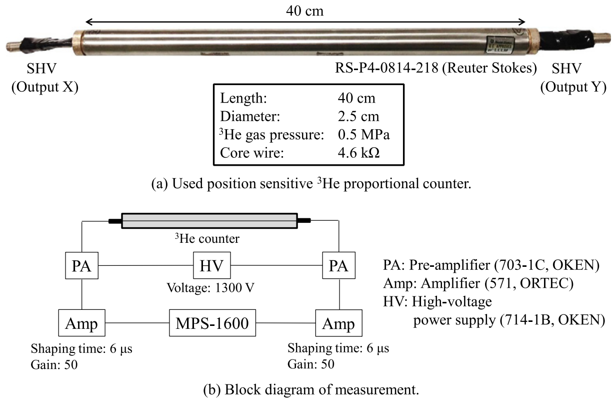

Fig. 5 Neutron detector used in this study.

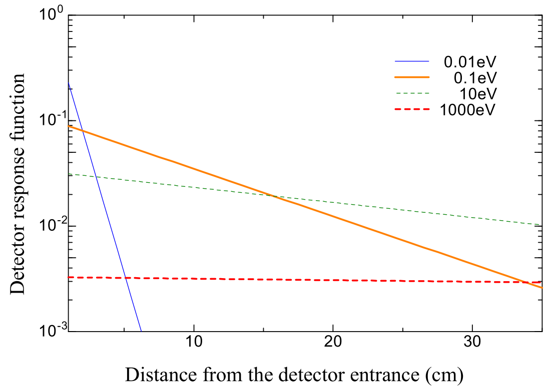

Fig. 6 Typical detection depth distribution.

resolution is not so high and the result is strongly dependent on the initial guess.

For this problem, we have been developing two spectrometers, one of which is a proportional counter based epi-thermal neutron spectrometer and the other is a Bonner sphere with a liquid moderator. In this paper the former is detailed. The latter is described briefly as follows: A special Bonner sphere was proposed using a liquid moderator/absorber instead of solid moderator like polyethylene, by which the number of moderators can be increased in principle. As a result, the number of energy group can also be increased, i.e., the energy resolution can be improved substantially. In our case, the number of moderators is around 50. The details can be found elsewhere [15].

Generally, it is hard to measure neutron energy if the energy is low. In the low energy region, the energy difference is a very tiny amount. We thus need to find a physical quantity to have a large sensitivity to the small energy difference. In this study, we employed neutron nuclear reaction cross section as the physical quantity, which can change largely reflecting and expanding the tiny energy change. For example, ^{3}He, ^{6}Li and ^{10}B are known to be available, in which the reaction cross sections are exceptionally large and in addition change drastically against neutron energy as shown in Fig. 4 [15]. ^{3}He and ^{10}B are commonly utilized as a neutron detection medium.

In this study, we selected a ^{3}He detector. Figure 5 shows the photo and electronic circuit of the detector. High voltage is impressed from both ends to obtain two signals. This kind of detector is known to be a position sensitive counter. In the position sensitive counter, the output amplitude of one side out of the two ends is determined by the resistance of the center wire of the detector. The resistance changes depending on the detection position, because the resistance is proportional to the distance from the detection position to each end. If a neutron is detected, by making a ratio of the both signals, the detection depth (position) can be determined.

Now assuming that one low energy neutron is entering from one side in parallel to the detector axis, it can be detected in a shallow position, i.e., close to the entrance surface, because the cross section in the lower energy region is higher as shown in Fig. 4. If the neutron energy is higher, it will be detected in a deeper position. When we think of the detection depth distribution after a lot of neutron incidences to the counter, the distributions for low and high energy neutrons are different with each other. The typical detection depth distributions are described in Fig. 6. If we prepare the detector responses (detection depth distributions) like Fig. 6 for each neutron energy, the measured detection depth distribution can be converted to the energy spectrum by solving an inverse problem with the estimated detector response function.

In the authors' group, for the last few years experimental validation of the developed spectrometer has been carried out to confirm the reproducibility performance of the neutron spectrum including the epi-thermal energy region. Practically, we first designed and developed a thermal/epi-thermal column with a DT neutron source of OKTAVIAN of Osaka University, Japan. This is a similar column to the one shown in Chap. 3, but in Chap. 3 the spectrum was designed to have a little harder neutron spectrum over 10 keV, while the column shown here has a clear epi-thermal spectrum covering 0.5 eV to 10 keV by using AlF_3 and Teflon moderators. Figure 7 shows the devel-

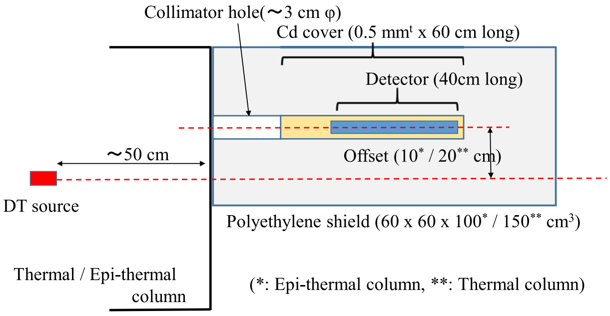

Fig. 7 Experimental arrangement of the spectrometer validation.

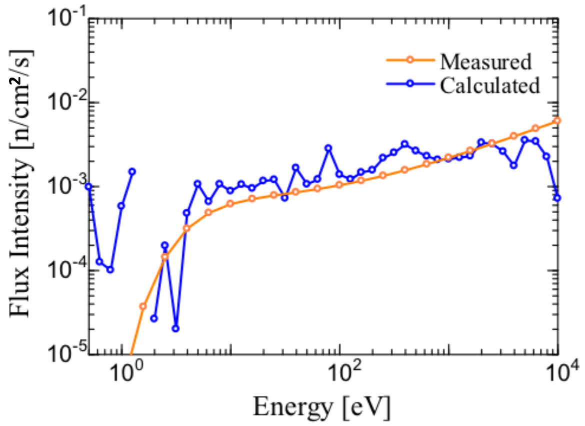

Fig. 8 Comparison of measured and calculated spectra.

oped thermal/epi-thermal neutron column (left hand side) and the spectrometer (right hand side). We then carried out the validation experiment of the spectrometer with the system in Fig. 7. A two-dimensional detection depth distribution derived from signals obtained from the both ends of the spectrometer was measured and the neutron spectrum was estimated with the measured detection depth distribution and the response function. Figure 8 is an example of comparison result of the measured and calculated spectra. The result shows an acceptable agreement between them, indicating the epi-thermal neutron spectrum can be measured directly in a field of BNCT. We are now planning to use the present spectrometer for our CSePT machine.

5. Conclusion

New essential neutronics characterization methods for ABNS-BNCT developed by the authors' group were overviewed, i.e., source neutron intensity monitor, epi-thermal neutron flux intensity monitor and neutron spectrometer. These were known to be difficult to realize especially in the epi-thermal energy region in BNCT. The developed techniques would be utilized in Osaka University ABNS-BNCT machine (CSePT) as well as other ABNS based BNCT facilities in near future.

[1] W.A.G. Sauerwein, A. Wittig, R. Moss and Y. Nakagawa (Eds.), *Neutron Capture Therapy: Principles and Applications* (Springer, 2012).

[2] H. Koivunoro, S. Green, I. Auterinen and M. Kulvik (Ed.), Proc. the 16th International Congress on Neutron Capture Therapy (ICNCT-16), Applied Radiation and Isotopes **106**, 1-264 (2015).

[3] MCNP Team, "MCNP5-1.40 RSICC Release Notes", LA-UR-05-8617 (2005).

[4] I. Murata, Y. Otani and F. Sato, Appl. Radiat. Isot. **106**, 75 (2015).

[5] M. Drosg, "DRORG-2000: Neutron Source Reactions", Nuclear Data Service, IAEA (2005).

[6] R.B. Firestone, V.S. Shirley (Ed.), *Table of Isotopes, Eighth Ed. Vol. I, II* (John Wiley and Sons, Inc., 1996).

[7] Y. Otani, F. Sato and I. Murata, "Experimental test of neutron intensity monitor with activation foil for p-Li neutron source", 8th Young Researchers BNCT Meeting, Sept. 13-17, 2015, Pavia, Italy (2015).

[8] X. Guan, M. Manabe, I. Murata and T. Wang, J. Nucl. Sci. Technol. **52**, 503 (2015).

[9] X. Guan, I. Murata and T. Wang, J. Nucl. Sci. Technol. **53 (8)**, 1112 (2016).

[10] X. Guan, M. Manabe, S. Tamaki, S. Liu, F. Sato, I. Murata and T. Wang, Appl. Radiat. Isot. **113**, 28 (2016).

[11] K. Shibata, O. Iwamoto, T. Nakagawa, N. Iwamoto, A. Ichihara, S. Kunieda, S. Chiba, K. Furutaka, N. Otuka, T. Ohsawa, T. Murata, H. Matsunobu, A. Zukeran, S. Kamada and J. Katakura, J. Nucl. Sci. Technol. **48 (1)**, 1 (2011).

[12] I. Murata and H. Miyamaru, Nucl. Instrum. Methods Phys. Res. **A589**, 445 (2008).

[13] I. Murata and T. Obata, Plasma Fusion Res. **9**, 4401107 (2014).

[14] G.F. Knoll, *Radiation Detection and Measurement, Fourth Edition* (John Wiley & Sons, Inc., 2010). ISBN: 978-0-470-13148-0

[15] S. Tamaki, F. Sato and I. Murata, Appl. Radiat. Isot. **106**, 41 (2015).