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Experimental test of neutron intensity monitor with isomer production reaction for p-Li neutron source for boron neutron capture therapy

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Introduction: Boron Neutron Capture Therapy (BNCT) is a 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 field formed with accelerators have different characteristics depending on kinds of accelerators. We thus have to characterize the field before practical use.

Method: In the authors' group, various neutronics characterization devices have been developed for our p-Li based BNCT machine named CSePT. In this paper, three neutron intensity monitor foils with an isomer production reaction for several tens to 800 keV of the p-Li neutrons were proposed, i.e., ^{107}Ag, ^{115}In and ^{189}Os.

Result and Discussion: From the experimental test results, two activation foils of ^{107}Ag and ^{115}In were confirmed to be a possible candidate as the monitor. However, the isomer production cross sections of them should be examined for practical use.

KEYWORDS

boron neutron capture therapy (BNCT), neutron intensity monitor, accelerator based neutron source, p-Li reaction, isomer/isomerization

1 Introduction

Boron Neutron Capture Therapy (BNCT) is a promising cancer therapy in future. Accumulating ^{10}B by administering a ^{10}B compound in tumor cells, neutrons are then bombarded in a neutron source facility to induce a neutron nuclear reaction with ^{10}B as Equation 1:

$$n + {}^{10}B \rightarrow \alpha + {}^{7}Li \quad (1)$$

Emitted two charged particles have energies higher than 1 MeV. The range in a human body is around 10 μm, which is mostly the same as the size of a human body cell. If ^{10}B would be accumulated only in tumor cells, only the tumor cells could be killed. BNCT is thus thought to be an excellent therapy which can treat tumor cells selectively with charged particles, sparing normal tissues.

BNCT has been carried out in nuclear reactors for more than 10 years as a clinical test in Japan. In recent years, epi-thermal neutrons (0.5eV–10 keV) have been begun to be used instead

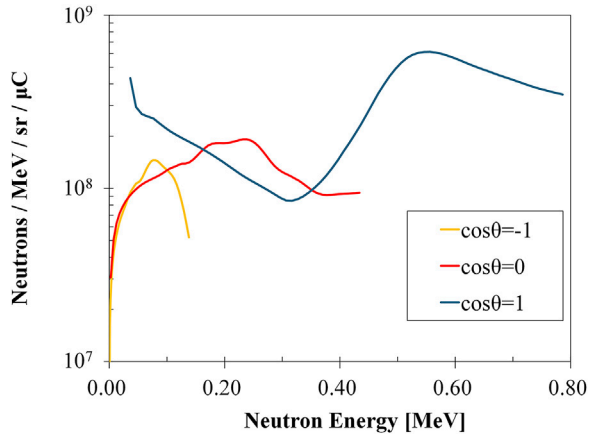

FIGURE 1
p-Li neutron spectra for various emission angles.

of thermal neutrons. Since then BNCT is recognized as a non-invasive therapy among cancer therapies. On the other hand, there is a critical problem, i.e., BNCT requires an intense epi-thermal neutron source. In the past, BNCT was performed at nuclear reactors of KUR, Kyoto Univ. (Kobayashi et al., 1990). and JRR-4, JAEA (Nakamura et al., 2011). However, especially in Japan it is not possible to construct a nuclear reactor in or close to a hospital. Under these circumstances, Japan is aiming at development and establishment of BNCT with an accelerator based neutron source (ABNS) instead of the nuclear reactor, because ABNS is safer and cheaper than nuclear reactor and can be constructed in hospitals. However, because the radiation field varies depending on kinds of accelerator and moderator, we have to conduct radiation field characterization of ABNS before practical use. As for the previous studies for this problem, basic approaches and considerations are summarized in Sauerwein's text book for BNCT (Sauerwein et al., 2012). The more recent related researches are found in the ICNCT Conference Proceedings (Koivunoro et al., 2015).

This paper presents the development of an activation detector for measuring the source term of a neutron source used in BNCT. The key objective of this research is to experimentally verify the reliability of the evaluated nuclear data for the isotopes used, specifically validating the cross-sections listed in JENDL (JNDC, 1991). The experimental results presented here confirm the feasibility of this approach and benchmark the nuclear data of the monitor material. The method, which directly measures the source term of neutrons generated by the p-Li reaction at several hundred keV, has rarely been reported in BNCT-related studies. Therefore, this technique would be useful for other p-Li based ABNS-BNCT facilities.

2 Materials and methods

2.1 Activation foil selection

The ABNS-BNCT, CSePT, utilizes the following p-Li reaction as Equation 2 (Murata et al., 2015):

$$p + {}^{7}Li \rightarrow n + {}^{7}Be - 1.88\,MeV \quad (2)$$

TABLE 1 Isomers of ^{107}Ag, ^{115}In and ^{189}Os.

Nuclide	Isotope abundance	E_γ(keV)[a]	Half-life
^{107}Ag[a]	51.8	93.13	44.3 s
^{115}In[a]	95.7	336.24	4.48 h
^{189}Os[a]	16.1	30.814	5.8 h

[a]The excitation energy of isomer is the same as E_γ.

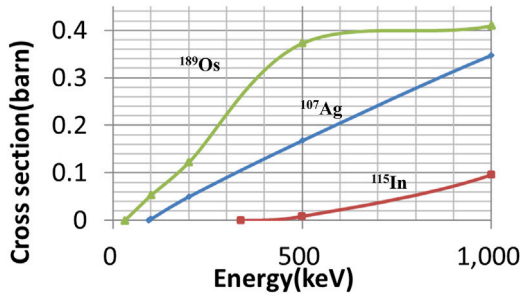

FIGURE 2
Isomer production cross sections of ^{107}Ag, ^{115}In and ^{189}Os (JNDC, 1991).

In the p-Li based BNCT, ~2.5 MeV protons are usually bombarded to a metal lithium target to produce neutrons. Figure 1 shows the neutron energy spectra (n/MeV/sr/μC) for bombarded proton energy E_p = 2.5 MeV as a function of emission angle (Drosg, 2005). The neutron energy spectrum structure is complex, i.e., the energy is lower in backward and higher in forward angles. In CSePT, an easy and convenient way to measure the absolute intensity is thus under development. The energy of neutrons to measure ranges from several tens to ~800 keV. However, it is generally known to be difficult to measure these neutrons. In this study, the foil activation method was employed to apply to convenient characterization of an intense neutron field. However, for the neutron energy range, very few activation foils are known to be available. If the energy is lower than this energy range, that is, around eV region or lower, neutron capture reaction is available. If it is in MeV or higher region, threshold reactions like (n,p), (n,α) reactions are suitable. Several tens to 800 keV are in-between capture and threshold reactions. So, we focused on isomer production reaction induced by neutron inelastic scattering. If a certain excited state of a nuclide has its own half-life, it may play a role of activation foil. Now, assuming the excited energy is E_{ex} and if the incident neutron energy is larger than E_{ex}, the nuclide may have a sensitivity to make itself excited to be an isomer. In addition, if the isomer decays by emitting a gamma-ray, the energy is also E_{ex}. If the E_{ex} is within several tens to 800 keV, that can be used for 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 (Firestone and Shirley, 1996), there exist over 20 candidate nuclides which can create an isomer by p-Li neutrons as listed in the following:

TABLE 2 Accelerator and target details.

High voltage (MV)	3.5
Beam current (μA)	3.5
Target material	Thick ^{7}LiF
Cooling medium	Air
Neutron intensity (n/sec)	~6 x 10^{8}

^{60}Co, ^{77}Se, ^{79}Se, ^{87}Sr, ^{94}Nb, ^{96}Tc, ^{99}Tc, ^{101}Rh, ^{103}Rh, ^{107}Pd, ^{107}Ag, ^{111}Cd, ^{113}In, ^{115}In, ^{117}Sn, ^{133}Ba, ^{134}Cs, ^{135}Ba, ^{154}Eu, ^{158}Tb, ^{163}Ho, ^{167}Er, ^{183}W, ^{189}Os, ^{193}Pt.

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 information (JNDC, 1991).

The cross sections of them are described in Figure 2 (JNDC, 1991). The energies we have to take into account are from several tens to 800 keV. From the figure ^{189}Os has sensitivity in lower energy region and the absolute cross section value is higher than others. The threshold energy of ^{115}In is the highest, because the first excited state energy is the highest. And ^{107}Ag is in-between the two. The cross section value of ^{115}In is as a result the lowest among the three and thus ^{107}Ag is in the middle of the three. The low threshold energy means that the cross section goes up from the lower energy region as in the case of ^{189}Os, and the absolute cross section value shows commonly higher compared with other two. However, it means that the emitted gamma-ray energy is lower (30.8 keV for ^{189}Os), because it is the same as the threshold energy. Consequently, ^{189}Os seems to be the best, because the cross section shows the largest among the three. However, its emitted gamma-ray energy is critically low to measure, which is a serious problem for practical use.

In any case, the selected three foils can be utilized for the absolute intensity monitor for p-Li neutron source, because they have sensitivity in energies of interest. As mentioned above, however, each has its own feature, i.e., we have to consider their measuring purposes when we apply them to the real BNCT scene. Theoretically, they have the following characteristics for possible applications.

^{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. However, we have to notice the activity decreases very rapidly after irradiation.

^{115}In: ^{115}In can become the most accurate monitor, because the abundance is large and the emitted gamma-ray energy is fairly large compared to the 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 two problems left, one of which is difficulty in preparing a thin sample. The other is that ^{189}Os should be enriched because there exists stable ^{188}Os having abundance of 13.2%.

2.2 Experimental test

Experimental tests were carried out before practical use for CSePT. We utilized a dynamitron accelerator of Tohoku University, Japan for the experiments. The details of the dynamitron accelerator is summarized in Table 2.

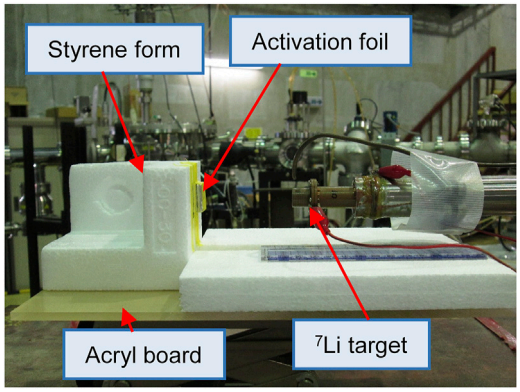

FIGURE 3
Photo of experimental setup.

The neutron intensity is fairly low compared to the real BNCT machine. Nevertheless, by the preliminary analysis the activation is confirmed to be enough large for the present irradiation experiment. In the experiment, the proton energy was 3.5 MeV and beam current was around 3 μA. The target material is a thin ^{7}LiF cooled by air to avoid distorting the source neutron spectrum. Figure 3 shows a photo of the experimental setup. A sample is placed at 7 cm from the target on the 0 deg. axis as in the figure. The absolute neutron intensity was determined by the activity of ^{7}Be produced from ^{7}Li(p,n)^{7}Be reaction, because the number of produced ^{7}Be is the same as the number of neutrons emitted. The number of neutrons in an experiment can be fixed by making difference of the activities measured before and after irradiation. The accuracy of this method is generally very high, because several hundred Bq can be obtained after only 1 h irradiation. The relative neutron intensity was monitored by a BF_3 counter located behind the sample on the 0 deg. axis.

Experiments were carried out for ^{107}Ag and ^{115}In. For ^{189}Os, it was not possible to prepare a thin sample. Irradiations were performed at 0 deg. and 90 deg. directions. Since the half-life of ^{107}Ag* is short, we adopted 5 min irradiation, 1 min Cooling and 5 min Measurement. The reason of 1 min Cooling is that it takes time to remove the sample and set it in a germanium semiconductor detector. We repeated this irradiation and measurement 20 times. After the experiment, we decided to exclude the data of 90 deg., because of the low measurement accuracy. For ^{115}In, the irradiation time was 19,000 s for 0 deg. and 27,600 s for 90 deg. Cooling of ^{116}In was not considered because the contribution was found to be very small. The measurement started after ~10 min After irradiation, and the measurement time was 48,000 s.

Neutron transport analysis of the experiment was carried out with MCNP5 (MCNP Team, 2005). In the analysis, JENDL-Activation Cross Section 96 (JENDL/A-96) (JNDC, 1991) and International Reactor Dosimetry and Fusion File (IRDFF) (Capote et al., 2012) were employed as activation cross section libraries. As for the neutron source term of p-Li reaction, that is, angle dependent neutron spectrum, DROSG-2000 was adopted. And the absolute neutron intensity was taken from the value

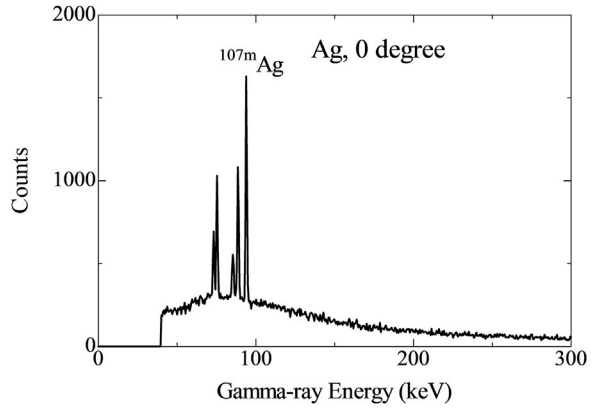

FIGURE 4
Pulse height spectrum for Ag (0 deg.).

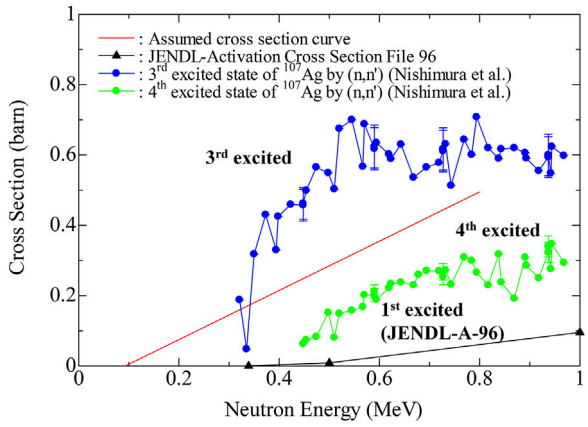

FIGURE 6
Inelastic scattering cross sections of ^{107}Ag.

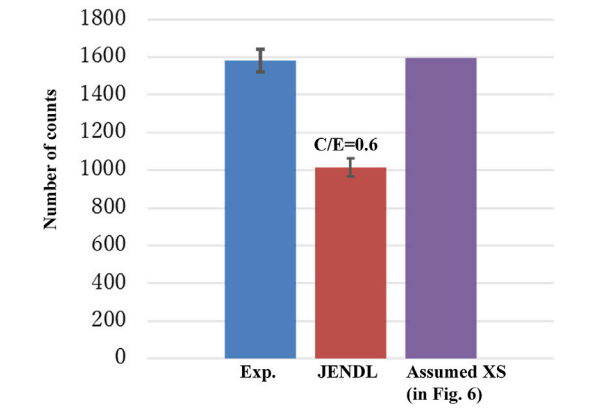

FIGURE 5
Number of counts of ^{107}Ag* at 0 deg. C/E = JENDL/Exp.

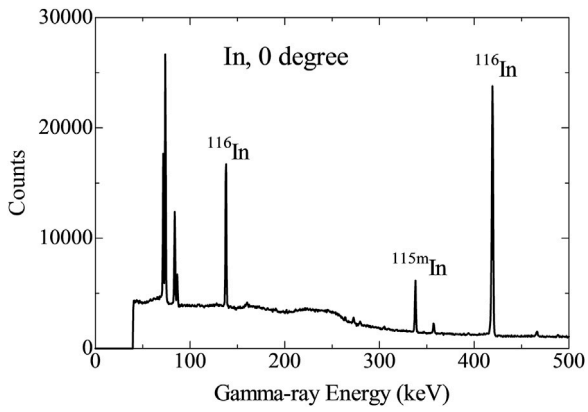

FIGURE 7
Pulse height spectrum for In (0 deg.).

determined by ^{7}Be in the target described earlier. The calculation model was made to accurately simulate the experimental system in Figure 3, including lithium target, activation foil, styrene form and so on. To estimate activity of the foil, track length estimator, F4, was used in MCNP5, until the uncertainty reach to less than 1%.

3 Results and discussion

Figure 4 shows an example of measured pulse height spectrum for Ag. This is the case for 0° after 20 times irradiation. A 93 keV peak is clearly observed. Other background gamma-rays from radioisotopes created by (n,γ) reaction do not affect the measurement as shown in the figure, because their energies are quite higher than 93 keV. Figure 5 shows the comparison result of ^{107}Ag between experiment and calculation. The *y*-axis is the number of measured gamma-ray counts. JENDL shown in the figure means the calculated result with JENDL/A-96. The ratio of the Calculated counts to the Experimental counts (C/E) is 0.6 and fairly low, indicating the cross section shows large underestimation. Figure 6 describes the cross section curve of ^{107}Ag inelastic scattering reaction. Similar to the result of Figure 5, JENDL/A-96 shows very small cross section value compared to those of other excited levels of third and fourth level excitations measured by Nishimura (Nishimura et al., 1965). If we use a similar cross section value of Nishimura's data shown in a red curve in Figure 6, the calculation value shows almost an equal value to the experiment within 1% difference. In any case, it shows that the evaluation was confirmed to be substantially low and should be examined and revised.

Figure 7 shows an example of 0° for In. A 336 keV peak is observed separately, though it is known that gamma-rays from ^{116}In can normally be measured simultaneously. It means that the influence of ^{116}In is acceptably small. In this measurement, the cooling time is not taken into account explicitly, i.e., around 10 min, suggesting ^{116}In would not be a serious problem in real applications. The comparison results of ^{115}In for 0 deg. and 90 deg. are shown in Figures 8, 9, respectively. For 0 deg., the agreement with the result of

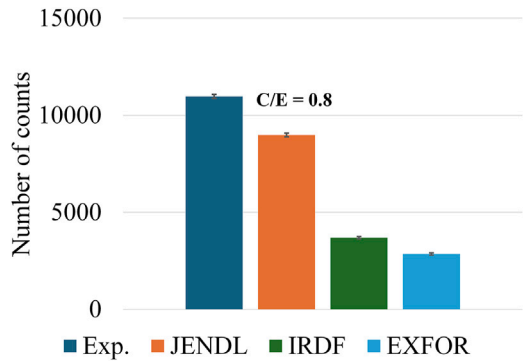

FIGURE 8
Number of counts of ^{115}In* at 0 deg. C/E = JENDL/Exp.

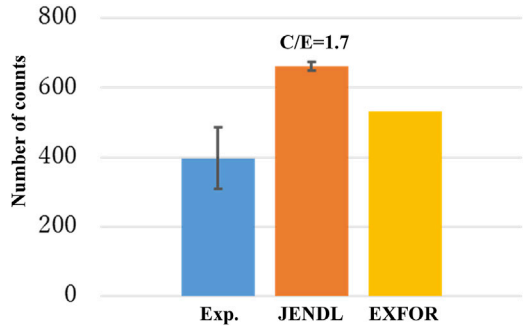

FIGURE 9
Number of counts of ^{115}In* at 90 deg. C/E = JENDL/Exp.

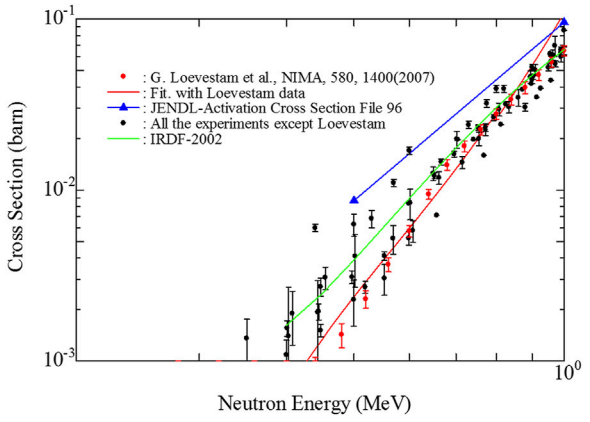

FIGURE 10
Inelastic scattering cross sections of ^{115}In.

JENDL was better than that of IRDF-2002 and fitting curve based on experimental value from EXFOR, and was acceptable, that is, prediction of the source intensity can be made in 20% accuracy. For 90-degree direction, as shown in Figure 9, JENDL shows a significantly higher value than the experimental result. Unfortunately, the experimental accuracy is a little low for the following reason: as shown in Figure 1, the neutron energy at 90° in this experiment was theoretically below approximately 400 keV. However, as indicated in Figure 2, the cross section of 115 In at this energy level was very small, resulting in minimal activation of the experimental sample. Consequently, the statistical error in the measurement was relatively large. To address this issue, it would have been necessary to extend the irradiation time or perform repeated irradiations. Figure 10 describes the cross section of JENDL/A-96 together with experimental values cited from EXFOR (Otuka et al., 2014). As expected in Figure 9, JENDL/A-96 shows larger cross sections than the experimental values. Red plots in EXFOR data were measured results obtained by Lövestam (Lövestam et al., 2007), which were the latest experimental values. If we employ the red curve fitted from Lövestam's data, the agreement is really improved as shown in Figure 9. It is thus suggested that the JENDL/A-96 data should be improved. In the figure the curve of IRDFF is also shown. The IRDFF is a little larger than Lövestam's data, indicating the IRDFF might be a little too large suggested from the present experiment.

4 Conclusion

In Osaka University, a new p-Li based ABNS-BNCT (CSePT) project is underway, and various neutronics devices are being developed for characterization of CSePT. In the present paper, a source neutron intensity monitor for the p-Li neutron source was investigated. As a result, the following three foils could be applied for the monitor depending on measuring purposes in the real BNCT scene:

^{107}Ag: Most convenient in a short measuring time.

^{115}In: Highest measuring accuracy.

^{189}Os: Only available for backward angle.

Also from the experimental tests, it was found that the activation reaction cross section data of JENDL-A-96 should be improved for ^{107}Ag and ^{115}In, while IRDFF was acceptable, for real applications of BNCT, because the accuracy of the performance check is quite important for real BNCT machines.

In the next step, we should check isomer production cross sections especially in lower energy region below 1 MeV for ^{107}Ag and ^{115}In. And it is necessary to investigate possibility of ^{189}Os for backward angle measurement. Our BNCT machine (CSePT) will be constructed in near future and will be operated with our developing neutronics devices for clinical tests.

Data availability statement

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

Author contributions

ST: Conceptualization, Investigation, Methodology, Validation, Visualization, Writing–original draft, Writing–review and editing. YO: Conceptualization, Investigation, Methodology, Validation, Visualization, Writing–original draft, Writing–review and editing. SK: Investigation, Writing–review and editing. FS: Methodology,

Supervision, Validation, Writing–review and editing. IM: Conceptualization, Methodology, Supervision, Validation, Writing–review and editing.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

The author(s) declared that they were an editorial board member of Frontiers, at the time of submission. This had no impact on the peer review process and the final decision.

Publisher's note

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