Neutronics Analyses of the Radiation Field at the Accelerator-Based Neutron Source of Nagoya University for the BNCT Study

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Abstract: The Nagoya University Accelerator-driven Neutron Source (NUANS) is an accelerator-based neutron source by $^7$Li$(p,n)^7$Be reaction with a 2.8 MeV proton beam up to 15 mA. The fast neutrons are moderated and shaped to beam with a Beam Shaping Assembly (BSA). NUANS is aiming at the basic study of the Boron Neutron Capture Therapy (BNCT) such as an in vitro cell-based irradiation experiment using a water phantom. Moreover, the BSA is developed as a prototype of one for human treatment. We have evaluated the radiation field of NUANS by a Monte Carlo code PHITS. It is confirmed that the radiation characteristics at the BNCT outlet meet the requirement of IAEA TECDOC-1223. Additionally, the radiation field in the water phantom located just in front of the BSA outlet is calculated. In the in vitro irradiation experiment, the boron dose of 30 Gy-eq, which is the dose to kill tumor cells, is expected for 20 min of irradiation at the beam current of 15 mA.

Keywords: boron neutron capture therapy; accelerator-based neutron source; PHITS code; radiation therapy; Monte Carlo method; neutron dosimetry

1. Introduction

Boron Neutron Capture Therapy (BNCT) utilizing $^{10}$B$(n,\alpha)^7$Li reactions [1] is one of the advanced methods of cancer radiation therapy. At first, BNCT was realized by using fission reactors [2–4], but more recently, BNCT using an accelerator-based neutron source [5–8] has been initiated, which can be installed inside or adjacent to areas of hospitals.

Several types of accelerators such as an electrostatic accelerator [5], a cyclotron [6], and an RFQ linac [7] are candidates for the neutron source for BNCT. Moreover, nuclear reactions such as $^7$Li$(p,n)^7$Be, $^9$Be$(p,n)^9$Be, $^7$Li$(d,n)^8$Be, and $^9$Be$(d,n)^{10}$B are the candidates of the neutron production reaction. Generally, those accelerator-based neutron sources produce energetic neutrons typically in the MeV range; therefore, the so-called Beam Shaping Assembly (BSA) [8] is required for slowing down neutrons and neutron beam shaping suitable for the BNCT irradiation. From the point of view of BSA downsizing, lower energy of produced neutrons is preferable. The deuteron-induced reactions, $^7$Li$(d,n)^8$Be and $^9$Be$(d,n)^{10}$B, produce neutrons of 4–15 MeV due to a positive Q-value of 4–15 MeV. On the other hand, $^7$Li$(p,n)^7$B and $^9$Be$(p,n)^9$Be reactions have a negative Q-value of approximately 2 MeV. If those reactions have a sufficiently large reaction cross-section near the threshold energy, we can obtain the relatively lower energy neutrons suitable for BNCT. The reaction of $^7$Li$(d,n)^8$Be has a threshold of 1.88 MeV and a large resonance at 2.26 MeV.
The Nagoya University Accelerator-driven Neutron Source (NUANS) [9–11] is an accelerator-based neutron source using a Shenkel-type electrostatic accelerator known as the Dynamitron [12], where $^7$Li(d,n)$^8$Be is employed as a neutron production reaction. NUANS is aiming at the basic study of BNCT such as in vitro cell-based irradiation experiments, ex vivo neutron irradiation of the bone marrow for blood cancer treatments and animal BNCT treatments. The real human client treatment is out of the scope of NUANS, although the BSA has been developed as a prototype for human treatment. This paper describes the radiation field characterization by using Monte Carlo calculations using the PHITS code [13]. Some of the calculation results are validated with experimental values.

2. Methods

2.1. NUANS Facility

Figure 1 shows the schematic view of the NUANS facility with photographs of the Dynamitron accelerator and BSA. The maximum acceleration voltage and beam current are 2.8 MeV and 15 mA for protons, respectively. The accelerator is installed horizontally and has two beamlines: one is 20° ended line for BNCT and the other is 80° bended line for generic science and technology purpose such as neutron imaging. A solid lithium target is mounted at the end of the 20 degrees beamline and surrounded with the BSA. The target is solid lithium of $60 \times 60 \times 0.5 \text{ mm}^3$ embedded on a water-cooled copper substrate, and the solid lithium is covered with a 5 µm-thick titanium foil to prevent the contamination of lithium to the accelerator. The BSA and the irradiation room are surrounded by an additional shield of concrete walls with borated-polyethylene plates. In addition, the 20° beamline is shielded by concrete walls with borated-polyethylene plates to shield backstream neutrons from the target. In the case of the in vitro irradiation experiments, a water phantom of $200 \times 200 \times 200 \text{ mm}^3$ acrylic box filled with water is located just in front of the BSA outlet to simulate the radiation field inside a human body.

![Figure 1. Schematic view of the NUANS facility and photographs of the Dynamitron accelerator and BSA.](image)

2.2. Computational Methods

In the Monte Carlo analyses, the BSA and the additional shield are modeled and the schematic view of the BSA calculation model is shown in Figure 2. The basic concept of this BSA is described in [9], it consists of four pieces of disk assemblies, which can be separated easily for the replacement of the lithium target and the maintenance of the BSA including the replacement of the moderator/filter materials. In the previous reports [9–11], the BSA is modeled with a simple one cylinder. Here, a four disk assembly structure is modeled to evaluate the neutron and gamma-ray leak via the structural material of steel (Structural-Steel 400). The MgF$_2$ and MgF$_2$ + CaF$_2$ layers in the BSA are used to moderate...
neutrons generated at the lithium target [14]. Neutrons are collimated using boron-doped polyethylene, PE-B (10%), and LiF-doped polyethylene, PE-LiF. Lead is used to shield gamma-rays. The BSA has a nozzle made of lithium-dope epoxy to sharpen the neutron beam shape at the BSA outlet. The diameter of the nozzle outlet is 120 mm. At the foot of the nozzle, a bismuth plate is inserted to shield gamma-rays.

Figure 2. Schematic view of the computational model for the BSA.

For the simulation of the in vitro cell-based irradiation experiments, a water phantom with vials is modeled as shown in Figure 3. A 3 mm-thick acrylic box of $200 \times 200 \times 200$ mm$^3$ filled with water is located just in front of the BSA outlet. Cells and water vials are mounted in an acrylic vial sheath 17.6 mm in diameter and 180 mm in length for the cell irradiation experiment. Three vial sheaths are inserted into the acrylic sample holder brock. The position of the sample holder in the water phantom is changeable. The vials are filled with physiological saline (water with 0.9 wt. % sodium chloride).

In the radiation field calculation, we have employed the PHITS version 3.26 and the nuclear data library of JENDL 4.0 [15]. The PHITS calculations have been performed on the PC cluster consisting of 20 nodes with four cores CPU each. The number of particle histories is not smaller than $1 \times 10^9$.

2.3. Neutron and Gamma-ray Source Term at the Lithium Target

For the source term of the PHITS calculation of the radiation field, angular neutron and gamma rays are evaluated by PHITS with the proton-induced nuclear data library of ENDF/B-VII [16]. The calculation model of the lithium target is a lithium disk with 100 µm in diameter and 100 µm in thickness covered with a 5 µm thick titanium foil. The spatial profile is assumed to be a pencil beam. The incident particle is a 2.8 MeV proton. The cover foil needs to tolerate the heat deposition by the proton beam. Titanium foil is suitable material due to its high melting temperature of 1941 K. The energy spread of the 2.8 MeV proton beam by the titanium foil is approximately 30 keV.
Figure 3. Schematic view of the computational model for the water phantom with vials.

Calculated neutron and photon (gamma-ray) yields are $1.13 \times 10^{12}$ [17] and $2.27 \times 10^9$ n/s/mA, respectively. The calculated neutron yield has a large anisotropy and angular-dependent spectra. On the other hand, the calculated photon yield is isotropic within the calculation statistics. Figure 4a shows the neutron source term with angular-dependent spectra. The photon source term is isotropic with the spectrum as shown in Figure 4b.

Figure 4. Neutron and photon source terms in the PHITS calculation. (a) Neutron source term is angular dependent, on the other hand, (b) photon source term is isotropic.

In the PHITS calculation of the BSA, the neutron and gamma-ray source term are distributed on the surface of the lithium target with an area of $60 \times 60$ mm$^2$ homogeneously. Indeed, the proton beam is scanned by the staring magnets on the lithium target surface to prevent heat concentration in the lithium plate; therefore, the assumption of the homogeneous neutron and gamma-ray emission on the lithium target surface is reasonable.

3. Results

3.1. Radiation Field Inside the Irradiation Room

In the NUANS facility, the real human treatment is out of the present scope. However, the BSA is designed as a prototype of the BSA for human treatment by BNCT. Therefore, the radiation field inside the irradiation room is important to evaluate not only the neutron...
beam quality at the BSA outlet but also shielding performance of the BSA. Figure 5 shows total neutron and gamma-ray distributions in the irradiation room, where the $z$-axis is the proton beams direction, and the $x$-axis is the horizontal direction normal to the $z$-axis. The origin of the coordinate is the center of the lithium target surface. Neutrons are emitted from the BSA outlet almost depending on $\cos \theta$, where $\theta$ is the angle against the proton beam axis. It is found that the neutron backstream is significant, and the shielding is not enough. Moreover, gamma-rays in the irradiation room are mainly from the proton beam inlet of the BSA. We presume that those gamma-rays are generated inside the BSA and via nuclear reactions with backstream neutrons and the shielding materials such as boron-doped polyethylene. Gamma-rays at the BSA outlet are sufficiently low, which is due to the bismuth plate at the foot of the nozzle. In the side direction of the BSA, neutrons and gamma-rays are well shielded. The neutron and gamma-ray streaming via steel flanges is not significant. If we need to reduce the dose rate not only by neutrons but also by gamma-rays in the irradiation room, additional shielding around the proton beam inlet is essential. Neutrons are well shielded by the auxiliary shielding walls. The absorbed neutrons generate gamma-rays in the shielding walls, and so gamma-rays are dominant outside the auxiliary shielding walls.

Figure 5. Total neutron (a) and gamma-ray distributions (b) in the irradiation room.

3.2. Radiation Field at the BSA Outlet and the Vicinity

Figure 6a shows the total neutron and gamma-ray flux distribution in the horizontal direction ($x$-axis) at the BSA outlet. The total neutron flux decreases sharply by approximately 1/100 at the edge of the BSA outlet. On the other hand, gamma-ray flux is not such a large difference between inside and outside the BSA outlet, which means that gamma-rays generated in the BSA are well shielded by the gamma-ray absorber of lead and bismuth. The neutron flux increases gradually towards the walls, which is due to the back scattered neutrons with walls and streaming neutrons from the proton beam inlet. The gradual increase in the gamma-ray flux towards the walls is mainly due to the capture gamma-rays by hydrogen and boron in the boron-doped polyethylene inner plates of walls. Figure 6b shows the neutron and gamma-ray effective dose rates derived from neutron and gamma-ray flux multiplied by the conversion coefficients recommended by ICRP60 [18]. In the region of 200 mm < $|X|$ < 600 mm, effective dose rates of neutrons and gamma-rays are almost the same. On the other hand, the effective dose rate of neutrons is dominant for the region of $|X|$ > 600 mm. The shape of the gamma-ray flux and the gamma-ray effective dose rate is different in the peripheral region, which is due to the difference in the gamma-ray spectrum.
The spectra of neutrons and gamma-rays at the center of the BSA outlet (X = 0 mm), outside the BSA outlet (X = 500 mm), and near the wall (X = 1500 mm), are shown in Figure 7. Here, the energy ranges of thermal neutron, epithermal neutron, and fast neutron are <0.5 eV, 0.5eV–10 keV, and >10 keV, respectively. At the center of the BSA outlet, neutrons in the epithermal region are dominant and the fast neutron flux of energy higher than 10 keV decreases drastically with the energy. On other hand, neutron spectra outside the BSA outlet and near the wall are almost the same, and dominant in the energy region of 0.1 eV–1 keV. For the gamma-ray spectra, some peaks of prompt gamma-rays from $\text{H}(n, \gamma)$, and $^{10}\text{B}(n, \gamma)$, and annihilation gamma-rays are identified. The gamma-rays of $^{10}\text{B}(n, \gamma)$ are believed to be primarily from the boron-doped polyethylene panels of the auxiliary shield, because gamma-rays of $^{10}\text{B}(n, \gamma)$ from boron-doped polyethylene inside the BSA, whose energy is approximately 0.5 MeV, are well shielded by lead and bismuth. A shoulder around 8 MeV is presumed to be from the neutron capture reaction with structural materials and shielding materials such as iron and lead.

As a reference, Table 1 summarizes the major parameters of the radiation field at the BSA outlet compared with those recommended by the IAEA TECDOC-1223 [1], where the proton beam current is assumed to be the maximum value of 15 mA while this system is not used for BNCT treatment and the beam was affected by the auxiliary shielding. Thermal neutron flux ratio, the ratio of epithermal neutron current to epithermal neutron flux, and gamma-ray dose per epithermal neutron flux meet the IAEA recommendations. Especially, the thermal neutron flux ratio is approximately 1/10 of the IAEA recommendation, which indicates that the thermal neutron is well suppressed in the BSA. The epithermal neutron flux is 20% lower than that recommended by IAEA. However, IAEA TECDOC-1223 describes that flux of $5 \times 10^8$ is acceptable with longer irradiation time. Therefore, we believe that the epithermal neutron flux of $8 \times 10^8$ is an acceptable level. Fast neutron dose per epithermal neutron flux is 2.6 times larger than that of the IAEA recommendation, however, it is an almost middle value of the range of $(2.5–13) \times 10^{-13}$ for the existing facilities. In the in vitro cell-based irradiation experiments and ex vivo neutron irradiation experiments, we will use a water or polyethylene phantom, where fast neutrons will be moderated immediately as described in Section 3.3. Therefore, we believe that the ratio of the fast neutron dose per epithermal neutron flux is not so critical [19]. Consequently, we evaluate that the radiation field at the BSA outlet of NUANS meets the IAEA recommendations.
except epithermal neutron flux and fast neutron dose per epithermal neutron flux; however, those two values are at acceptable levels.

![Image of neutron and gamma-ray spectra](a) Neutrons and (b) gamma-rays at the center of the BSA outlet (X = 0 mm), outside the BSA outlet (X = 500 mm), and near the wall (X = 1500 mm).

**Figure 7.** Spectra of (a) neutrons and (b) gamma-rays at the center of the BSA outlet (X = 0 mm), outside the BSA outlet (X = 500 mm), and near the wall (X = 1500 mm).

**Table 1.** Major parameters of the radiation field at the BSA outlet compared with those recommended by the IAEA TECDOC-1223.

| Parameters                                      | PHITS Calculation          | IAEA Recommendation |
|------------------------------------------------|----------------------------|----------------------|
| Epithermal neutron flux (1/cm²/s) \(\Phi_{\text{epi}}\) | \((8.04 \pm 0.03) \times 10^8\) (at 15 mA) | \(\geq 1 \times 10^9\) |
| Fast neutron flux (1/cm²/s) \(\Phi_f\)            | \((1.42 \pm 0.01) \times 10^8\) (at 15 mA) | No recommendation    |
| Thermal neutron flux (1/cm²/s) \(\Phi_{\text{th}}\) | \((5.31 \pm 0.53) \times 10^6\) (at 15 mA) | No recommendation    |
| Thermal neutron flux ratio \(\Phi_{\text{th}}/\Phi_{\text{epi}}\) | \(0.0066 \pm 0.0004\) | \(\leq 0.05\)        |
| Ratio of Epithermal neutron current to Epithermal neutron flux \(C_{\text{epi}}/\Phi_{\text{epi}}\) | \(0.726 \pm 0.006\) | \(\geq 0.7\)         |
| Fast neutron dose per epithermal neutron flux (Gy cm²) \(D_f/\Phi_{\text{epi}}\) | \((5.28 \pm 0.08) \times 10^{-13}\) | \(\leq 2 \times 10^{-13}\) |
| Gamma-ray dose per epithermal neutron flux (Gy cm²) \(D_{\gamma}/\Phi_{\text{epi}}\) | \((1.00 \pm 0.19) \times 10^{-13}\) | \(\leq 2 \times 10^{-13}\) |

### 3.3. Radiation Field in the Water Phantom and Vials

As described in Section 2, cells are irradiated in the water phantom, where thermal neutron flux is the most important parameter because \(^{10}\text{B}(n, \alpha)\) reaction has a large cross-section for thermal neutrons. Figure 8 shows the thermal neutron flux distributions in the horizontal plane at the beam axis level for the cases of the water phantom only, and that with a sample holder with vials where the center of the sample holder is 30 mm distant from the water phantom surface. Incident neutrons are thermalized rapidly in the water and the thermal neutron flux maximizes around 20–30 mm from the surface of the phantom.
It is found that the acrylic sample holder distorts the thermal neutron distribution slightly. The high thermal neutron flux region is elongated toward the Z-direction, which is due to lower hydrogen atomic density of acrylic than that of water.

Figure 8. Thermal neutron flux distributions in the horizontal plane at the beam axis level for the cases of (a) the water phantom only and (b) the water phantom with a sample holder with vials.

Figure 9 shows thermal neutron, epithermal neutron, fast neutron, and gamma-ray flux, and $^{10}\text{B}(n, \alpha)$ reaction rate distributions on the beam axis as a function of the distance from the water phantom surface for the cases of the water phantom only and that with a sample holder with vials. It is clearly shown that the thermal neutron flux maximizes at around 20–30 mm from the surface of the water phantom. Additionally, gamma-ray flux reaches its maximum around 20–30 mm, showing that gamma-rays are mostly generated by the thermal neutron capture reactions of hydrogen. Fast neutron and epithermal neutron fluxes decrease almost exponentially with the distance from the surface of the water phantom. An increase in the fast and epithermal neutron flux is observed near the back end of the phantom, which is caused by neutrons from the backside of the water phantom because the irradiation room is very small. The acrylic sample holder reduces thermal neutron and gamma-ray fluxes slightly (typically 10%); however, the sample holder does not affect epithermal and fast neutron fluxes. Therefore, we can choose the sample holder location to meet the desired parameters such as ratios of the fast neutron flux to the thermal neutron flux and the gamma-ray flux to the thermal neutron flux. We can conclude that the current sample holder location, where the center of the vial is 30 mm from the surface of the water phantom, is almost optimum to obtain the highest thermal neutron flux.

The radiation field of each vial at 1 mA operation is listed in Table 1, where the center of the vial is 30 mm from the water phantom surface. The vial numbers correspond to those in Figure 3. These values have good symmetry in the right/left direction. The values in the bottom vials are approximately 3% larger than those in the top vials, which might be explained by the neutron and gamma-ray flux reflected by the floor. The thermal neutron fluxes inside the top and bottom vials are approximately 10% lower than that of the center vial. Those in the four corners are approximately 20% lower. Fast neutron flux and gamma-ray flux are approximately 1/10 of the thermal neutron flux in the same vial. Based on this table, we can make an irradiation plan of cells.

Vial numbers correspond to those in Figure 3.
4. Discussion

For the basic study of BNCT at NUANS such as the in vitro cell-based irradiation tests, the radiation fields around the BSA including the BSA outlet and in the water phantom have been evaluated by a Monte Carlo code PHITS. To confirm the calculation validity, thermal neutron flux distribution on the beam axis in the water phantom has been measured with the small $^6$Li/Eu: CaFe scintillator detector [20] at the 0.25 mA operation as shown in [17], where we obtained a good agreement, typically within 6% deviation, between the calculation and the measurement.

We chose low-activation materials for the BSA. The gamma-ray dose measurement by an ionization chamber located at the BSA outlet confirmed that the gamma-ray dose due to activated materials was negligibly small compared with the prompt gamma-ray dose, by the comparison of the doses during proton beam on and just beam off.

In the case of in vitro cell-based irradiation tests, the absorbed dose of a tumor cell including 87.5 ppm of $^{10}$B can be derived from the thermal neutron flux. Absorbed dose on human tissue ($D_{eq}$) after accounting for biological effect is represented by:

$$D_{eq} = \text{RBE (or CBE)} \times D$$  \hfill (1)

where RBE is the relative biological effectiveness; CBE is the compound biological effectiveness for an alpha particle from $^{10}$B(n, $\alpha$)$^7$Li reactions; and D is the absorbed dose. The CBE value of the boron dose is 3.8, assuming the boron transporting drug of BPA (boronophenylalanine) [21]. The absorbed dose by the boron capture reaction is approximately $2 \times 10^{-3}$ Gy-eq/s/mA or 7 Gy-eq/h/mA for vial #1. The lower limit dose to kill tumor cells is considered to be 30 Gy-eq; thus, we can obtain the dose by 20 min irradiation at the 15 mA operation. Therefore, we can perform the in vitro cell-based irradiation tests at NUANS.

As shown in Table 2, we found non-uniformity in the thermal neutron flux for each vial. If we obtain the uniform thermal neutron flux for nine vials, we can irradiate nine samples simultaneously under the same radiation field. Now, we are trying to modify the sample holder numerically to provide the uniform thermal neutron flux for nine vials.
Table 2. The radiation field of each vial at 1 mA operation in the case of vial center location of 30 mm from the phantom surface. The vial number corresponds to Figure 3.

| Vial number | #1       | #4       | #7       |
|-------------|----------|----------|----------|
| Thermal neutron flux \( \times 10^7 / \text{cm}^2 \text{s/MA} \) | 6.49 ± 0.01 | 7.17 ± 0.01 | 6.49 ± 0.01 |
| Fast neutron flux \( \times 10^6 / \text{cm}^2 \text{s/MA} \) | 6.20 ± 0.02 | 7.36 ± 0.02 | 6.40 ± 0.02 |
| Gamma-ray flux \( \times 10^6 / \text{cm}^2 \text{s/MA} \) | 7.37 ± 0.01 | 7.87 ± 0.01 | 7.41 ± 0.01 |

| Vial number | #2       | #5       | #8       |
|-------------|----------|----------|----------|
| Thermal neutron flux \( \times 10^7 / \text{cm}^2 \text{s/MA} \) | 7.77 ± 0.01 | 8.49 ± 0.01 | 7.75 ± 0.01 |
| Fast neutron flux \( \times 10^6 / \text{cm}^2 \text{s/MA} \) | 6.89 ± 0.02 | 7.89 ± 0.02 | 7.05 ± 0.02 |
| Gamma-ray flux \( \times 10^6 / \text{cm}^2 \text{s/MA} \) | 8.40 ± 0.01 | 9.01 ± 0.01 | 8.82 ± 0.01 |

| Vial number | #3       | #6       | #9       |
|-------------|----------|----------|----------|
| Thermal neutron flux \( \times 10^7 / \text{cm}^2 \text{s/MA} \) | 6.69 ± 0.01 | 7.38 ± 0.01 | 6.71 ± 0.01 |
| Fast neutron flux \( \times 10^6 / \text{cm}^2 \text{s/MA} \) | 6.23 ± 0.02 | 7.30 ± 0.02 | 6.38 ± 0.02 |
| Gamma-ray flux \( \times 10^6 / \text{cm}^2 \text{s/MA} \) | 7.52 ± 0.01 | 8.09 ± 0.01 | 7.54 ± 0.01 |

5. Conclusions

By Monte Carlo calculations using the PHITS code, radiation fields around the BSA including the BSA outlet have been evaluated. The radiation field at the BSA outlet meets almost all of the IAEA recommendations. The radiation fields in vials embedded in the water phantom have also been evaluated. We now have the prospect to kill tumor cells in vials by BNCT with 20 min irradiation at 15 mA operation.

Author Contributions: Development of NUANS, K.W., K.T., Y.K. and A.U.; lithium target development, S.H.; modeling, Y.T.; software and computational infrastructure, A.Y.; measurement, S.Y.; computation, visualization, and writing, T.N.; review and editing, Y.K.; project administration, K.T. All authors have read and agreed to the published version of the manuscript.

Funding: This research was done as a part of the collaborative work between Nagoya University and YAGAMI inc.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Acknowledgments: Authors wish to thank Y. Tsurita for his effort to the NUANS operation and maintenance. One of authors (T.N.) wish to thank the PHITS office of JAEA for their technical suggestion on the PHITS code.

Conflicts of Interest: The authors declare no conflict of interest.

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