Optimization of beam shaping assembly and thermal neutron flux measurement for BNCT

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Abstract: The pilot design of the beam shaping assembly (BSA) is optimized. The MCNP6 was used to calculate beam parameters that IAEA suggests to ensure effective and reasonable neutron capture treatment. By adding the reflector and increased density of the moderator, the BSA configuration D successfully satisfied the IAEA criteria (the epithermal neutron flux was 1.78×10^9 cm^{-2}s^{-1}, the thermal to epithermal neutron ratio was 0.031, the directionality was 0.715, the hydrogen and gamma dose per epithermal neutron were 1.84×10^{-13} Gy-cm^2 and 0.87×10^{-13} Gy-cm^2, respectively.). As the second study, the thermoluminescent dosimeter (TLD) was calibrated and verified in a nuclear reactor for the measurement of the thermal neutron flux. The conventional thermal neutron flux measurement uses gold foil or gold wire, and the measured thermal neutron flux is used to evaluate the boron dose, a dose contributing to the treatment in the boron neutron capture therapy. The use of a cadmium filter is essential because the neutron cross-section of gold has a relatively low energy range showing a 1/v shape. However, TLDs made of lithium fluoride materials have a much wider energy range in which the cross-section of lithium is 1/v, and the tendency is similar to that of boron so that the dose of boron can be evaluated more accurately than gold. For this, the thermoluminescent dosimeter was calibrated using a californium neutron source, and the calibration constant was 3.80×10^4 cm^{-2}nC^{-1}. In order to verify the measured calibration constant, a research/educational reactor of Kyunghee University was used. When the thermal neutrons of the nuclear reactor were measured using gold, it was measured as 2.17×10^5 cm^{-2}W^{-1}s^{-1}, and the TLD measured as 2.28×10^5 cm^{-2}W^{-1}s^{-1} using a cadmium filter. The thermal neutron flux was accurately measured by TLDs having a 5% difference from the one by the gold wire. In conclusion, through this study, the use of the MCNP6 code in the design of BSA can yield similar results to the actual measured values, the optimized BSA configuration was suggested, and a more accurate method of evaluating boron dose using a thermal fluorescence dosimeter was proposed.

1. Introduction

The Boron Neutron Capture Therapy (BNCT) is a binary radiation treatment modality of cancer. It delivers lethal radiation damage to the tumor cell using 1) selective accumulation of ^{10}B compound and 2) subsequent thermal neutron irradiation into the tumor region. The ^{10}B nuclei with high thermal neutron capture cross-sections release α particle and ^{7}Li ion in ^{10}B(n, α)^{7}Li reaction breakup. These products have high linear energy transfer characteristics, and their ranges are around single-cell diameter (~10 μm) in the tissue. Therefore, BNCT can selectively treat the tumor cells with the targeted boron drug delivery [1, 2].

In the BNCT, effective tumor control can be achieved by using suitable neutron beams (also highly tumor-selective and low toxic boron compounds) and accurate dose evaluation in the patient's body. The IAEA suggests the neutron beam parameters suitable for the treatments. The parameters requested from the IAEA reference report [3] are: (1) the epithermal beam intensity, $\Phi_{epi} > 1\times10^9$ cm^{-2}s^{-1}, (2) the ratio between thermal and epithermal flux (i.e., Φ_{th}/Φ_{epi}), less than 0.05, (3) the fraction of beam forward direction (i.e., forward current/flux ratio), higher than 0.7, (4) fast neutron dose and photon dose per epithermal neutron less than 2×10^{-13} Gy-cm^2. To design the AB-BNCT facilities, the Monte Carlo (MC) simulation is the essential method to evaluate these beam characteristics before construction.

After the BNCT facility is built, the dosimetry must be accurately evaluated to perform the clinical trial stage and patient treatment stage properly. The dose components in BNCT are divided into four components, boron dose, nitrogen dose, fast neutron dose, and gamma dose, and gamma dose is also divided into internal and external gamma dose depending on the location where the gamma is generated. Among them, the boron dose as the therapeutic dose is obtained from the measurement of thermal neutron flux. The conventional method to measure the thermal neutron flux in BNCT is the activation analysis method. The combination of gold wire (or foil) and cadmium are usually used to measure the thermal neutron flux. However, because of the resonance in the epithermal energy range of gold, the method using cadmium excludes the reaction rate by the epithermal neutron, and thus underestimation the reaction rate by the neutron.

The primary object of this study is to optimize the pilot beam shaping assembly (BSA) design and develop the method to estimate the thermal neutron flux with TLDs. The MCNP6 was used as a result of the previous study [4]. The TLDs were irradiated at the research/educational reactor and the measured thermal neutron flux was compared with the gold activation analysis.

2. Materials and Methods

2.1 BSA optimization

The beam characteristics of the pilot design calculated in the previous chapter did not fully satisfy the IAEA criteria [3]. In detail, the epithermal neutron flux, directionality, thermal to epithermal neutron flux ratio, and gamma dose were satisfied, but the hydrogen dose was not. For the optimization, the pilot design was modified as described in Table 1 except the proton beam parameters (8 MeV and 10 mA), the beryllium target shape (2-cm thick), and the size of the beam exit port (12-cm in diameter). Accordingly, the thickness and material combination of BSA components, such as moderator, collimator, reflector, etc., were changed to satisfy IAEA criteria. The necessary properties of the material constituting the reflector should have high scattering cross-sections and high atomic masses to reduce energy transfer during scattering. On the other hand, the materials of the moderator should be low atomic masses to effectively moderate the fast neutron energy.

Table 1 shows the dimensions of these configurations. From the pilot design to configurations A and B, the primary design objective was to increase the neutron population at the beam exit port by reflecting the back-scattered neutrons. Then the neutrons were effectively moderated in the configurations C and D.

Table 1. The configuration of the five BSA designs

BSA Configuration	Materials & thickness [cm] at the central axis of BSA	Major Modification
Pilot design	W 3, Fe 9.5, AlF_3 52, Pb 2	Reference [4]
A	W 3, Fe 17, AlF_3 44.5, Pb 2	Add Al near the target
B	Fe 20, AlF_3 44.5, Pb 2	Switch W to Fe
C	Fe 20, AlF_3 44.5, Pb 2	Extend Al up to 0 cm Increase in AlF_3 density
D	Fe 20, AlF_3 44.5, Bi 2	Switch Pb to Bi Add LiF sheet prior to the exit port

2.2 TLD reading

The 21 TLD-600 and 21 TLD-700 (Harshaw, USA) chips were prepared for this study. Its diameter was 4.5 mm and the thickness was 0.6 mm. The TLDs used in this study are lithium fluoride based materials (LiF:Mg,Ti). TLD-600 was enriched by ^{6}Li (95.6%) whereas TLD-700 was enriched by ^{7}Li(99.99%) [5]. Due to the difference in neutron cross-section of ^{6}Li and ^{7}Li, TLD-600 is sensitive to neutrons and TLD-700 is insensitive to neutrons [6]. The TLDs were annealed with an electric furnace at 400°C for 1 hour followed by 100°C for 2 hours to eliminate the residual signals before the irradiation.

The readout of TLDs was done by using Harshaw TLD reader (Model 3500). The TLDs were linearly heated from 50 °C to 300 °C at 10 °C/sec. The acquisition time was 33.3 seconds.

The TL signal was converted in the unit of electric charges by integrating the glow curves from channels 72 to 200. The thermal neutron induced TL signal was separated by using the combination of TLD-600, TLD-700 and cadmium sheets with their different interaction probabilities of neutrons and gamma rays.

2.3 TLD validation

To evaluate whether the calculated TLD calibration factor is valid, a research/educational reactor at Kyunghee University, known as a thermal neutron source, was used. The thermal neutron flux measured through the applying calibration factors of TLDs was compared with the thermal neutron flux using the conventional gold wire activation method. At this time, the thermal neutron flux is the Westcott fluence.

In this study, measurements were made using a Cd sheet that absorbs thermal neutrons with high probability. Therefore, if Cd sheet is placed in the existing beam port in AGN-201K, it absorbs too many thermal neutrons and lowers the criticality. Due to the insufficient margin of the criticality of the educational/research reactor, the neutron irradiation was conducted at the top of the vessel far from the core.

3. Results and discussion

3.1 BSA optimization

The hydrogen dose per epithermal neutron of the pilot design was 6.80×10^{-13} Gy-cm^2, exceeding the IAEA criteria of 2.0×10^{-13} Gy-cm^2 as shown in Table 2. Therefore, it is important to increase the epithermal neutron flux or to decrease the fast neutron fluence to reduce the hydrogen dose.

From the MC simulations, it was noticed that there were many neutrons from the target directed toward the front of the BSA (opposite to beam exit port), which led to beam loss at the beam exit port. Therefore, the first modification of BSA optimization (i.e., BSA configuration A) was to further increase the neutron flux at the beam exit port by adding a material that can act as a reflector on the front side of the target. Materials widely used as reflectors are Pb, PbF_2, and Bi [3]. However, the scattering cross-section of Pb is high, which is an appropriate characteristic for the purpose of the reflector, but the energy of the neutron generated in the ^{9}Be target of this study was not adequately moderated resulting in a high hydrogen dose at the beam exit port. Therefore, it was decided to expand the first aluminum of the pilot design back to the entrance.

Table 2. Beam characteristics of various BSA designs in terms of IAEA criteria [3]. (ϕ_{epi} is epithermal neutron flux and ϕ_{th} is thermal neutron flux) The iBNCT and Nagoya data were from the IAEA technical

meeting [7].

BSA configuration	ϕ_{epi} [cm^{-2} s^{-1}]	ϕ_{th} / ϕ_{epi}	Hydrogen dose / ϕ_{epi} [Gy-cm^2]	Photon dose / ϕ_{epi} [Gy-cm^2]	Directionality
IAEA recommendation	> 1×10^9	< 0.05	< 2×10^{-13}	< 2×10^{-13}	> 0.7
Pilot design (8 mA)	1.40×10^9	0.015	6.80×10^{-13}	0.69×10^{-13}	0.780
A (8 mA)	2.14×10^9	0.011	8.64×10^{-13}	0.39×10^{-13}	0.715
B (8 mA)	2.97×10^9	0.012	8.55×10^{-13}	0.40×10^{-13}	0.714
C (8 mA)	1.92×10^9	0.045	1.78×10^{-13}	0.91×10^{-13}	0.714
D (8 mA)	1.78×10^9	0.031	1.84×10^{-13}	0.87×10^{-13}	0.715
iBNCT (5 mA)	1.80×10^9	0.018	3.75×10^{-13}	0.28×10^{-13}	-
Nagoya (15 mA)	1.02×10^9	0.073	2.00×10^{-13}	3.50×10^{-13}	-

In configuration A, the epithermal neutron flux was increased from 1.40×10^9 cm^{-2}s^{-1} to 2.47×10^9 cm^{-2}s^{-1}, but the hydrogen dose was also increased from 6.80×10^{-13} Gy-cm^{-2} to 8.64×10^{-13} Gy-cm^{-2} due to the increase in the fast neutron flux. The directionality was decreased from 0.780 to 0.715 due to the neutrons reflected back to the beam exit port.

In configuration B, the tungsten adopted in the pilot design was replaced with Fe in order to increase the moderation effect. As a result, the epithermal neutron was increased by 38%, while the hydrogen dose was decreased. However, the hydrogen dose was still four times more than the IAEA criteria.

In configuration C, reducing the number of fast neutrons was pursued by extending the aluminum on the side to the entrance. Further, the pressured powder AlF_3 used in the pilot design was replaced with sintering and sputtering AlF_3. This increased the density of AlF_3 from 1.58 to 3.02 g/cm^3. This configuration significantly lowered the fast neutron dose below the IAEA criteria but increased thermal neutrons, which were almost close to the IAEA criteria.

Finally, in configuration D, the thermal/epithermal flux ratio was reduced by placing a 3-mm thick sheet of LiF near the beam exit port to absorb the thermal neutrons. In addition, in order to increase epithermal neutron, Bi was used instead of Pb at the end of the AlF_3 moderator. Bi can shield gammas as much a Pb but passes epithermal neutrons better than Pb. Bi was placed at a distance from the beam exit port because it generates ^{210}Po by neutron capture reactions, emitting alphas [3]. The beam characteristics of configuration D fully satisfied all of the IAEA criteria. In particular, the epithermal neutron flux was comparable to that of iBNCT [7], even satisfying the hydrogen dose criteria. The beam characteristics of the other groups (Table 2) which have accelerator-based BNCT facilities need further optimization in the unwanted dose distribution by recoiled protons or gammas.

3.2 TLD validation

The thermal neutron flux measured by the gold wire activation method is 2.00×10^5 cm^{-2}s^{-1}W^{-1}. The self-shielding correction factor of 0.2 mm diameter gold wire was 0.928 by MCNP6 simulations [8]. The corrected value was 2.17×10^5 cm^{-2}s^{-1}W^{-1} and it used as the standard for thermal neutron flux measurement.

Table 3 shows the thermal neutron flux at the irradiation site of the reactor by multiplying the TLD reading value obtained in the TLD validation setup by the calibration factor. In 4 measurements, when the thermal neutron flux was normalized to the reactor power and irradiation time, the average value was 2.28×10^5 cm^{-2}W^{-1}s^{-1}. The thermal neutron flux measured by TLD showed an average difference of 5% with the gold wire method.

Table 3. The TLD readings are normalized to reactor power and irradiation time and corrected by the neutron self-shielding correction factor.

	TLD-600	TLD-700	Neutron induced readings
Bare [nC W^{-1}s^{-1}]	6.15 ± 0.63	0.04 ± 0.003	6.11 ± 0.63
Cd covered [nC W^{-1}s^{-1}]	0.26 ± 0.03	0.16 ± 0.01	0.10 ± 0.03
Thermal neutron induced reading [nC W^{-1} s^{-1}]			6.01 ± 0.63
Calibration factor [cm^{-2} nC^{-1}]		$(3.80 \pm 0.425) \times 10^4$	
Thermal neutron flux [cm^{-2}W^{-1} s^{-1}]		$(2.28 \pm 0.35) \times 10^5$	

The calibration factor that converts TLD readings to thermal neutron flux is aided by MC simulations. The calculated thermal neutron flux is divided by TLD readings in the calibration process. The uncertainties are classified as Type A or Type B [9]. Type A evaluation of uncertainties is from the statistical analysis, such as the standard deviation of MC results and readings of TLD chips in the calibration process. On the other hand, Type B evaluation is other than the statistical analysis such as the specifications by manufacturer and previously defined values. The combined uncertainties of the calibration factor and validation process were 10.91% and 15.13% (coverage factor, *k*=1), respectively (Table 4).

Table 4. Estimated uncertainties of calibration factor and validation.

Component	Uncertainties (%)	
	Type A	Type B
Heater reproducibility (σ_{Heat})		1%
Photomultiplier stability (σ_{PMT})		2%
High voltage stability ($\sigma_{Voltage}$)		0.005%
TLD reading in calibration process ($\sigma_{TLD\_Cali.}$)	10.65%	

MC results (σ_{MC_Cali})	0.71%
***Calibration factor ($\sigma_{Cali.}$)**	10.91%
TLD reading in validation process ($\sigma_{TLD\_Vali.}$)	10.49%
****Validation (σ_{Vali})**	15.13%

* $\sigma_{Cali.} = \sqrt{\sigma^2_{Heat} + \sigma^2_{PMT} + \sigma^2_{Voltage} + \sigma^2_{TLD\_Cali.} + \sigma^2_{MC\_Cali.}}$

** $\sigma_{Vali.} = \sqrt{\sigma^2_{Cali.} + \sigma^2_{TLD\_Vali.}}$

4. Conclusions

In this study, the BSA was optimized, and the alternative thermal neutron flux measurement was successfully established. The TLD-600, TLD-700 and cadmium sheets were used to validate the calibration factor. The established method enables to estimate thermal neutron flux without complex instruments used in the conventional method. The thermal neutron flux measured by TLD showed an average difference of 5% with gold activation analysis which means TLD measurements showed fair agreement with the conventional method.

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