

Development of remote-operating
liquid multilayer
neutron spectrometer
for boron neutron capture therapy

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Chapter 1 Introduction

This chapter introduces background and scope of this research on boron neutron capture therapy (BNCT). First, the fundamental principle of BNCT is described. Next, Kyoto University Reactor (KUR), which serves as the irradiation field in this study, is introduced. The importance of dosimetry for quality assurance (QA) in BNCT is then discussed, covering in-air, in-phantom, whole-body, and real-time beam monitoring measurements. Subsequently, the significance of neutron energy spectrum evaluation, which is a part of in-air measurement for BNCT QA, is explained, followed by an overview of representative neutron energy spectrum evaluation methods and their limitations. Finally, the key challenges addressed in this study and proposed solutions are presented, and an overall overview of this research is provided.

1.1 Principle of boron neutron capture therapy (BNCT)

Cancer treatment involves multiple therapeutic modalities, including surgery, chemotherapy, and radiation therapy. A brief overview of cancer epidemiology and radiation therapy is provided in Appendix A for general background. This section focuses on boron neutron capture therapy (BNCT) as an advanced form of radiation therapy.

Boron neutron capture therapy (BNCT) is a type of radiotherapy that targets cancer using isotope ^{10}B , which has high cross-section for capturing thermal neutrons. Prior to treatment, a boron-containing drug is administered to the patient, and it selectively accumulates in tumor cells. When the patient is subsequently exposed to a neutron beam, boron neutron capture reaction (BNCR) takes place. The ^{10}B atoms capture thermal neutrons and generate heavy charged particles, including ^7Li nuclei and alpha particles, as shown in Figure 1.1(a). These particles exhibit high linear energy transfer (LET) and have a very short range of approximately 10 μm — roughly equivalent to the diameter of a single cell — allowing them to selectively damage cancer cells.

Figure 1.1(b) shows BNCR inside biological tissue by epithermal neutron. Epithermal neutrons, upon entering the patient body, collide with hydrogen atoms (^1H) and gradually slow down, becoming thermal neutrons that can induce BNCR for the site of deeper-seated tumors. Table 1.1 describe definition of neutron energy (thermal, epithermal, fast) used in BNCT.

For BNCT to be clinically effective, two essential conditions must be satisfied: sufficient accumulation of boron compounds in tumor tissue and appropriate neutron irradiation with well-characterized beam properties. Detailed descriptions of boron compounds and neutron sources used in BNCT are provided in Appendix B and Appendix C, respectively.

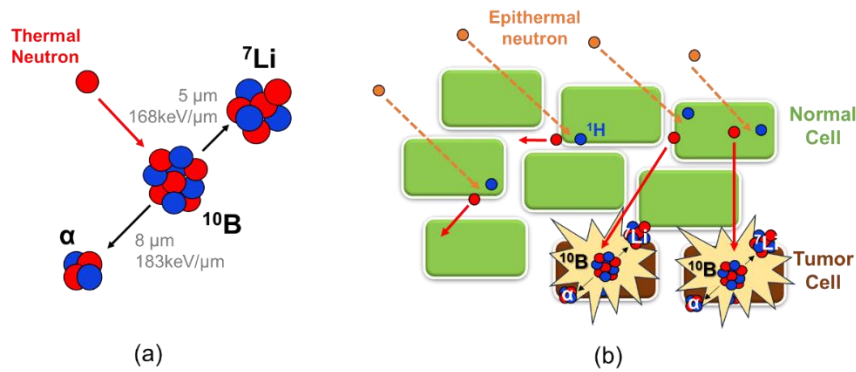


Figure 1.1: BNCT principle

(a) boron neutron capture reaction (BNCR), (b) BNCR inside biological tissue

Table 1.1: Definition of neutron energy used in BNCT

	Thermal	Epithermal	Fast
Neutron Energy	< 0.5 eV	0.5 eV – 10 keV	> 10 keV

Figure 1.2 shows the depth distribution of thermal neutron in water phantom which demonstrate as human body when irradiated with either a thermal neutron beam (0.025 eV) or highest energy of epithermal neutron beam (10 keV), normalized to the surface point. Historically, thermal neutrons were primarily used for treatment; however, in recent years, epithermal neutron beams have become standard. This shift is based on the fact that neutron beams with maximum energies around 10 keV can penetrate deeper into the body, improving the intensity of thermal neutrons at those deeper locations and effectively inducing the BNCR within tumors situated farther from the surface.

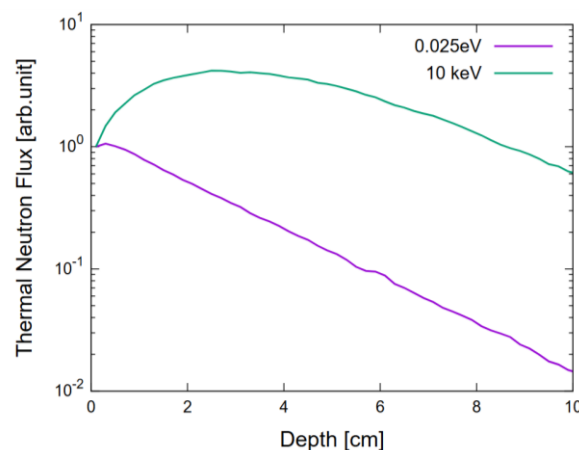


Figure 1.2: Thermal neutron distribution inside water phantom

By irradiating fast neutron energy range, thermal neutrons can penetrate deeper into tissue. However, the absorbed dose rate in normal tissue becomes high, resulting in a loss of the cancer-selective dose advantage of BNCT. Based on simulation, BNCT treatment at a 10-cm depth in the head region, the most effective neutron energy has been reported to be 10 keV. [1], [2]

1.2 Kyoto University Reactor (KUR)

In this research, the facilities inside Heavy Water Neutron Irradiation Facility (HWNIF) and E3 Prompt Gamma Analysis Facility at Kyoto University Reactor (KUR) are used as important irradiation fields. Figure 1.3 shows photo and cross-sectional view of KUR. KUR is a swimming-pool-type research reactor extensively used for experiments and research in various fields, including physics, chemistry, biology, engineering, agriculture, and medicine. KUR reached its first criticality in 1964. In February 2006, the use of high-enriched uranium (HEU; ~93% uranium enrichment) fuel was discontinued. Subsequently, in May 2010, HEU fuel was completely replaced by low-enriched uranium (LEU; ~20% uranium enrichment) fuel, and the reactor continues operation with LEU fuel to date. KUR operates at the maximum thermal power of 5 MW, and achieves an average thermal neutron flux of about $3 \times 10^{13} \text{ cm}^{-2}\text{s}^{-1}$ in the reactor core.

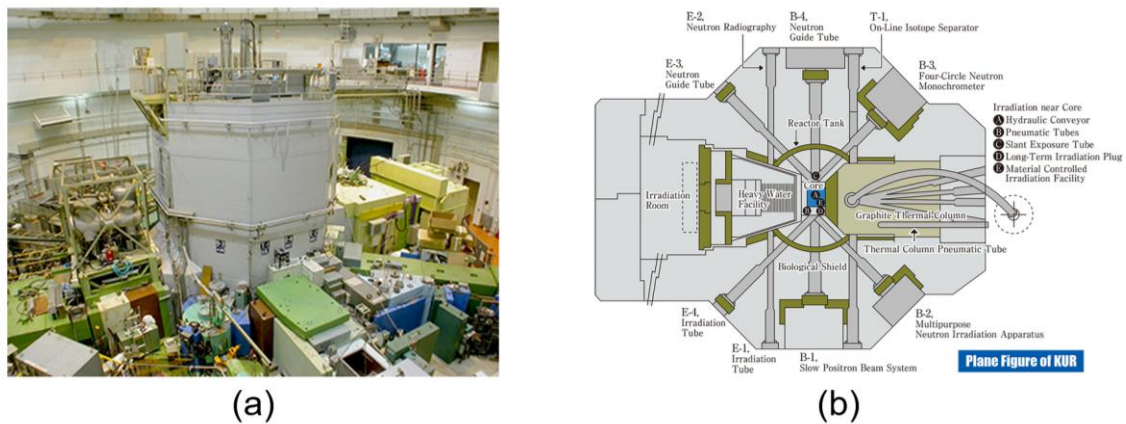


Figure 1.3: Kyoto University Reactor (KUR)

(a) top view photo, (b) horizontal cross-sectional view

Reactor operation is controlled using 4 coarse control rods and 1 fine control rod made of boron-containing stainless steel. The KUR reactor vessel is an aluminum tank 2 m in diameter, 8 m deep, and 1.2 cm thick, filled with water. The reactor core is installed at the bottom of this tank. Heat generated in the reactor core is removed by forced circulation of the tank water, transferred via heat exchangers to the secondary cooling water, and finally dissipated into the atmosphere through cooling towers.

Operation of KUR will finish by May 2026, which corresponds to the deadline for the return of spent nuclear fuel to the United States. The new 10 MW thermal power research reactor at the Monju site, Fukui prefecture is planned for western Japan nuclear research in the future. [3]

1.2.1 Heavy Water Neutron Irradiation Facility (HWNIF)

Kyoto University Reactor Heavy Water Neutron Irradiation Facility (KUR-HWNIF) has been widely used for BNCT-related research. Detailed descriptions of the facility structure and components are provided in Appendix D.

In BNCT, thermal neutron irradiation is used to treat melanomas and gliomas via open craniotomy. However, epithermal neutron beams, which can penetrate deeper with a higher thermal neutron flux within a patient's body, are currently used to treat deep-seated tumors without craniotomy. For radiobiological research on BNCT, the study of thermal and epithermal beams is necessary.

Figure 1.4 shows the neutron energy spectra of the thermal, epithermal, and mixed modes, which are the three standard irradiation modes of KUR-HWNIF. In this research, only epithermal neutron standard irradiation mode is utilized.

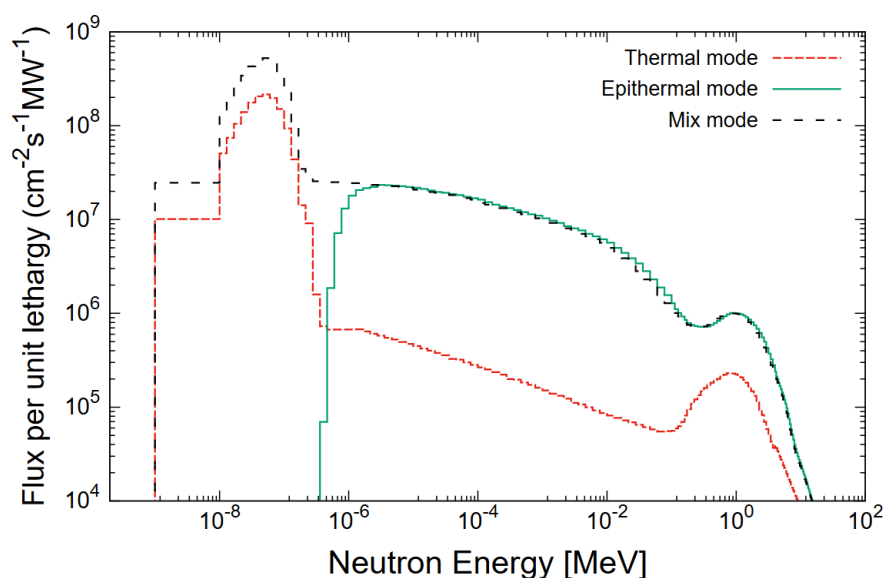


Figure 1.4: Neutron energy spectra of three standard irradiation modes in KUR-HWNIF

In the KUR-HWNIF, two different sample delivery systems can be utilized, as shown in Figure 1.5: the irradiation rail system (Figure 1.5 (a)) and the irradiation remote-carrying system (Figure 1.5 (b)). According to facility regulations, samples for the irradiation rail system must not exceed 10 cm in width, 10 cm in height, and 20 cm in length, with a weight limit of 1 kg. For the irradiation remote-carrying system, the maximum allowable sample dimensions are 100 cm × 100 cm × 100 cm, with a weight limit of less than 500 kg.



(a)



(b)

Figure 1.5: Two different sample delivery systems in KUR- HWNIF

(a) irradiation rail device, (b) irradiation remote-carrying device

1.2.2 E3 Prompt Gamma Analysis Facility

E3 Prompt Gamma Analysis Facility is designed for performing prompt gamma analysis (PGA) on research samples. It uses a highly collimated, thermal neutron beam with minimal fast neutron and gamma contamination obtained via a nickel-mirrored neutron guide from the KUR core. [4] By combining with High Purity Germanium (HPGe) detector, this facility has been used to measure ^{10}B concentrations in BNCT patient blood. In this study, the facility is used to confirm the concentration of boric acid solution, one of the liquid moderators employed in the neutron spectrometer development. It also used to confirm gamma-neutron discrimination property of neutron detector that used in this study.

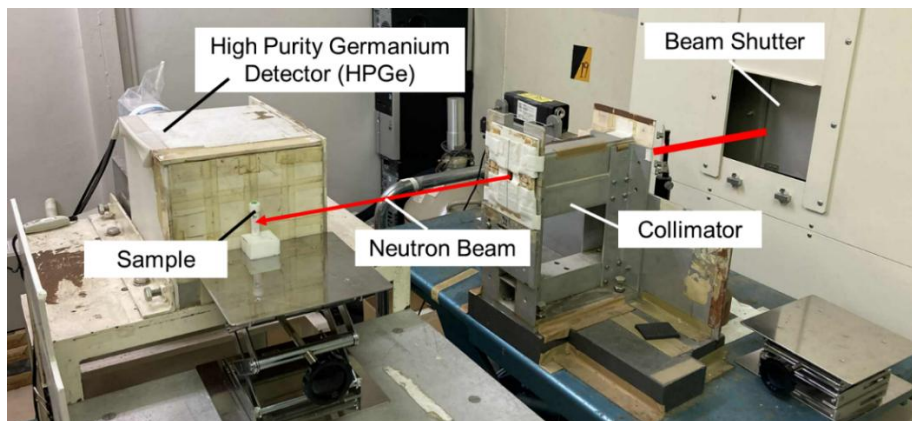


Figure 1.6: E3 Prompt Gamma Analysis Facility

The boron concentration measurement by PGA is based on the $^{10}\text{B}(n,\alpha)^7\text{Li}$ reaction, which emits 478 keV gamma ray, and the $^1\text{H}(n,\gamma)^2\text{H}$ reaction, which emits a 2220 keV gamma ray. The ^{10}B concentration in an unknown sample is determined by comparing the intensity ratio of the 478 keV and 2220 keV peaks with that obtained from a standard boron solution sample containing 50 ppm of ^{10}B .

1.3 Important dosimetry for quality assurance (QA) in BNCT

Before performing BNCT treatment at a new facility, it is necessary to confirm the characteristics of the treatment beam, or irradiation field, through appropriate measurements. Various types of measurements are required during system acceptance testing and commissioning. The results obtained during commissioning are subsequently used as reference values for quality assurance (QA) during clinical treatment.

The International Atomic Energy Agency (IAEA) report provides guidelines including accurate dosimetry for verifying beam performance, validating treatment planning calculations, and ensuring the safe clinical operation of BNCT. [5] These guidelines highlight that in-air, in-phantom, whole-body, and real-time measurements should be performed, as shown in Figure 1.7.

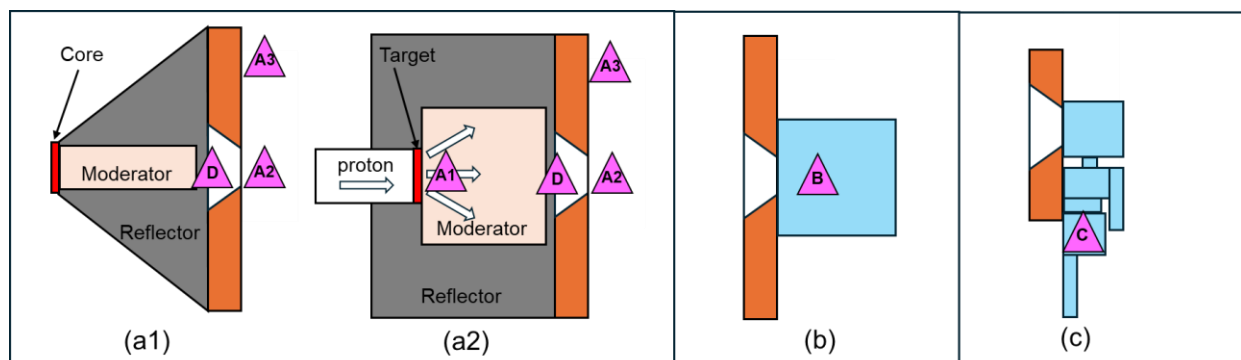


Figure 1.7: Measurement point that should be performed in BNCT (Adjusted from [5])

(a1) reactor-based BNCT upward component,

(a2) accelerator based BNCT upward component,

(b) collimator with phantom, (c) collimator with whole body phantom.

A1: neutron energy spectrum at target, A2: neutron energy spectrum at collimator exit

A3: out-of-field evaluation, B: in-phantom measurement,

C: whole-body exposure, D: real-time beam monitoring.

1.3.1 In-air measurement

- Neutron energy spectrum at target

For accelerator-based BNCT, measuring the neutron energy spectrum at the target (A1 in Figure 1.7) is critical because fast neutrons produced at different energies before entering the beam shaping assembly (BSA) significantly influence dose calculations. Accurate spectral information is therefore required to validate Monte Carlo simulations and to ensure reproducible and predictable beam characteristics during treatment.

- Neutron energy spectrum at collimator exit

Measurements at the collimator exit (A2 in Figure 1.7) are required because the therapeutic neutron field consists of mixed neutron components. The relative proportions of thermal, epithermal, and fast neutrons must be quantified to assess whether the beam meets clinical specifications, to

support accurate treatment planning, and to verify that patient irradiation is delivered under controlled and well-characterized spectral conditions.

- Out-of-field evaluation

Out-of-field evaluations (A3 in Figure 1.7) are necessary to assess unintended neutron and gamma leakage outside the treatment volume. These measurements contribute to patient safety by quantifying non-target doses, which may increase the risk of secondary cancers and other adverse effects if not properly controlled.

1.3.2 In-phantom measurement

In-phantom measurements (B in Figure 1.7) are required to determine the depth distribution of thermal neutrons, which directly contribute to the tumor dose via the $^{10}\text{B}(n,\alpha)^7\text{Li}$ reaction. Measuring the spatial and depth-dependent neutron distributions validates the dose calculations performed by the treatment planning system and confirms that the planned therapeutic dose is physically achievable in patient-like geometry.

1.3.3 Whole-body exposure

Whole-body exposure (C in Figure 1.7) assessment further ensures radiation safety by measuring neutron and gamma doses at organ-equivalent positions and comparing them with Monte Carlo simulations.

1.3.4 Real-time beam monitoring

Real-time beam monitoring (D in Figure 1.7) requires a well-calibrated relationship between the neutron output and the corresponding monitor signals. Accurate neutron measurements obtained during commissioning ensure that the system can reliably deliver the prescribed dose using monitor units.

1.4 Importance of neutron energy spectrum evaluation for BNCT QA

In this research, the focus is placed on evaluation of the neutron energy spectrum at the collimator exit, as described in Chapter 1.3. A fundamental issue in BNCT is that neutron energy spectrum can fluctuate depending on the characteristics and stability of the neutron source.

For reactor-based neutron sources, operating conditions vary due to changes in fuel composition and control rod insertion states during or between operation cycles. As a result, the neutron energy spectra for each irradiation mode can shift in both absolute intensity and relative spectral shape. In the case of KUR, uncertainties relative to nominal values have been reported to be approximately 20% for the thermal neutron flux and about 30% for the epithermal and fast neutron fluxes. [6]

In accelerator-based BNCT systems, particularly those employing lithium targets, degradation of the Li target is a significant concern. Numerical studies have shown that when the Li target thickness decreases below 50 μm , noticeable fluctuations occur in neutron energy spectrum, accompanied by a reduction in the number of useful neutrons. [7]

These findings highlight a fundamental issue in BNCT: neutron energy spectrum is not constant and can vary due to operational or structural changes in neutron source. Therefore, QA should include reliable neutron energy spectrum evaluation to ensure that beam characteristics remain consistent with those measured in commissioning process.

1.5 Neutron energy spectrum evaluation method in BNCT

Figure 1.8 describes two major methods that have traditionally been employed for neutron spectrum evaluation in BNCT, multifoil activation method and Bonner sphere spectrometer. These techniques have been widely used in many BNCT facilities as shown in Table 1.2. In addition, several newly developed neutron spectrometers have been reported, which are expected to contribute to QA in BNCT, and these approaches will also be described in this section.

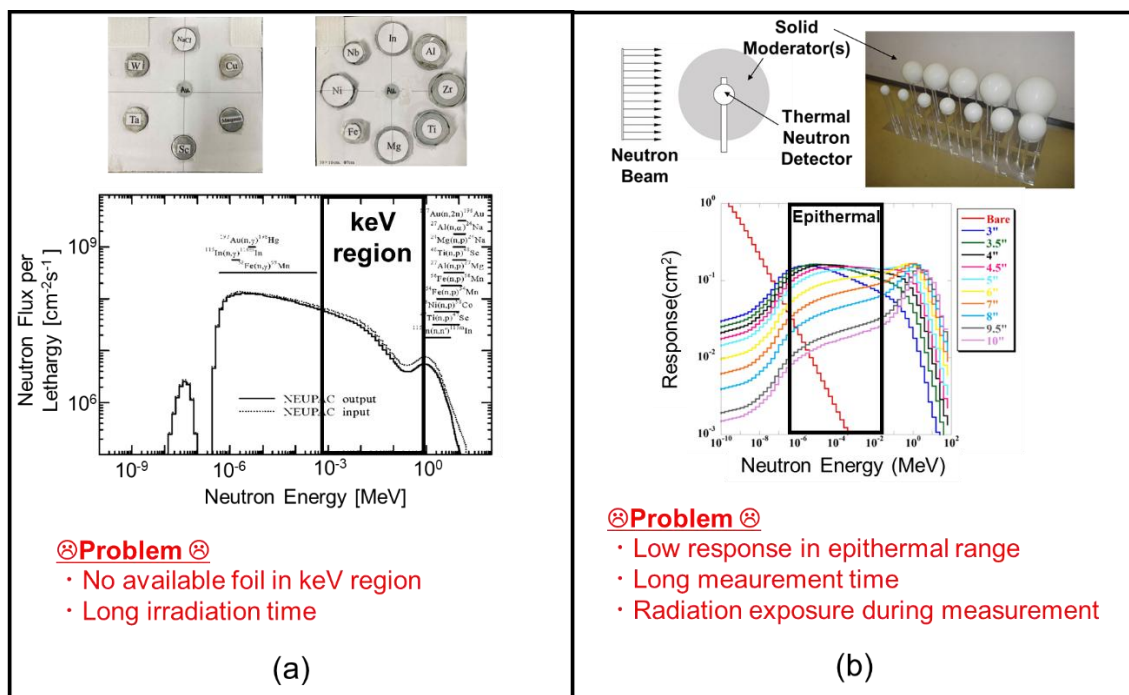


Figure 1.8: Conventional method for evaluating neutron energy spectrum in BNCT

(a) multifoil activation, (b) Bonner sphere spectrometer

Table 1.2: Neutron energy spectrum measurement situation in BNCT facilities

First Author/year	Facility/location	Ref
MULTIFOIL ACTIVATION METHOD		
Y.Harker (1992)	BMRR, USA	[8]
Y.Sakurai (2004)	KUR, Japan	[6]
I.Auterinen (2004)	BMRR, WSU and MITR, USA JRR-4, KUR, Japan HFR, Netherland FiR1, Finland RA-6, Argentina	[9]
Y.Liu (2009)	HFR, Netherland THOR, Taiwan	[10]
Y.Bae (2022)	Dawon Medax, South Korea	[11]
BONNER SPHERE SPECTROMETER		
A.Masuda (2017)	iBNCT, Japan	[12]
COMBINATION METHOD		
S.Bortolussi (2017)	TrigaMark II reactor Pavia, Italy	[13]

1.5.1 Multifoil activation method

In this method, a set of activation foils made of different elements is irradiated in neutron field. Each foil has an energy-dependent neutron reaction cross section, providing selective sensitivity to different neutron energies. After irradiation, the reaction rate is measured using gamma-ray spectrometry which is used for neutron energy spectrum evaluation.

The problem of this measurement method is that its sensitivity in the few-keV region is limited, which is a critical energy range in BNCT. [14] This limitation leads to reduced accuracy, particularly in the epithermal region. Furthermore, long irradiation time is required by using this method.

1.5.2 Bonner sphere spectrometer

Bonner sphere spectrometer consists of polyethylene spheres of various diameters with a central thermal neutron detector. By varying the moderator thickness, each sphere provides a different energy response, enabling coverage from thermal to fast neutrons. The response at higher energies can be enhanced by combining polyethylene with heavy materials such as lead or copper.

The problem of this measurement method is that it exhibits poor energy resolution in the epithermal region, which is crucial for BNCT, due to its flat response. In addition, changing solid moderators requires manual handling, increasing radiation exposure for operators.

1.5.3 Newly developed neutron spectrometer for BNCT

To address the challenges associated with Bonner sphere spectrometer in BNCT applications — particularly their poor energy resolution in the epithermal region and operational difficulties. As shown in Figure 1.9, two major approaches have been proposed: (1) modifying response functions using materials with high thermal neutron absorption, and (2) remote-operating spectrometer implementing liquid moderators and detectors for fast measurements.

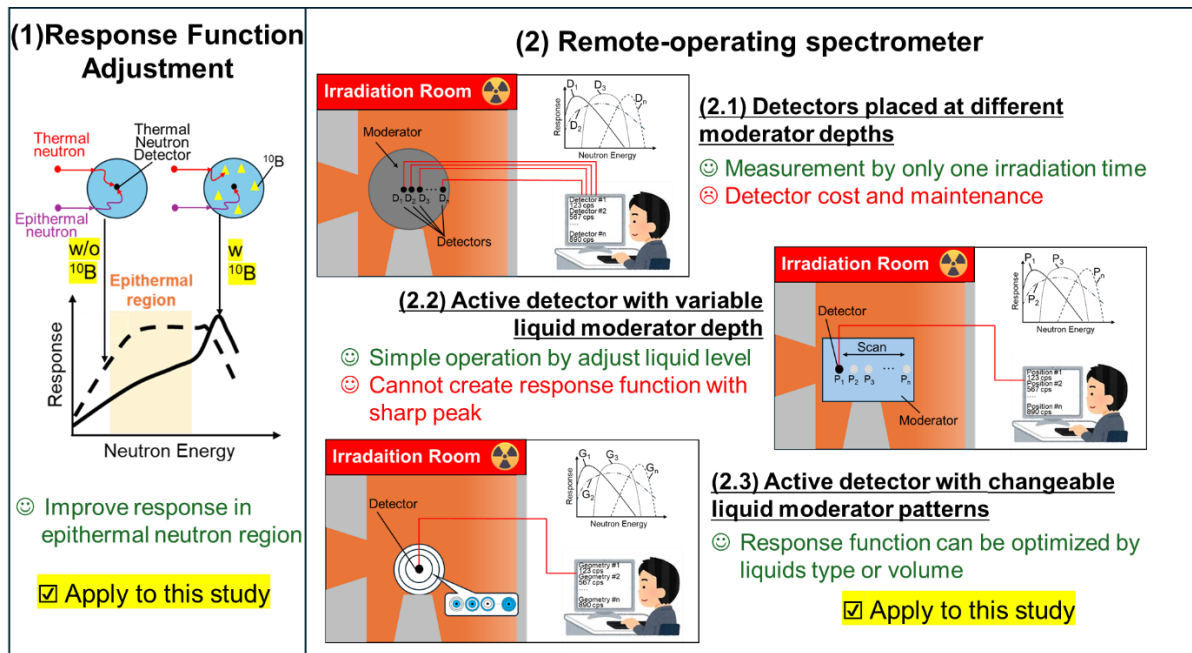


Figure 1.9: Newly developed neutron spectrometer for BNCT with different approaches

(1) Response function adjustment using thermal neutron absorbing materials

One approach for improving neutron spectrometry in BNCT is to modify the response function using thermal-neutron-absorbing materials such as boron, which suppress low-energy neutrons and enhance sensitivity in the epithermal region. Yonai et al. [15] applied this concept by inserting boron-loaded silicon rubber between polyethylene moderator layers

Another approach involves the use of liquid moderators, which provide greater flexibility than solid moderators. Tamaki et al. [16], [17], [18] investigated several liquid materials and identified $\text{B}(\text{OCH}_3)_3$ and NaBF_4 solutions — both containing high-density ^{10}B — as effective moderators for shaping the response function. Ueda et al. [19], [20] also explored moderator designs boric acid solutions, showing that adjustable ^{10}B -concentration boric acid effectively tuned the response toward the epithermal range.

In this study, a boron concentration of 0.14 wt%, as suggested by Ueda et al. [19], [20], was adopted to achieve optimized response in BNCT.

(2) Remote-operating spectrometer

Online measurement systems have been proposed to improve measurement efficiency. Different approaches enable flexible measurements while reducing operator exposure.

(2.1) Detectors placed at different moderator depths

Several spectrometers place multiple detectors at different moderator depths to obtain several response functions simultaneously. Examples include multi-layer Eu:LiCaAlF₆ (LiCAF) detector by Mizukoshi et al. [21], NCT-WES silicon-diode array by Monti et al. [22], both of which employ active detectors, as well as CsI self-activation method proposed by Kurihara et al. [23], which utilizes a passive detector, all of which enable rapid and/or real-time measurements without manually changing moderators.

However, this approach requires many detectors, resulting in high cost and increased maintenance demands.

(2.2) Active detectors with variable liquid moderator depth

Another strategy is to adjust the thickness of a liquid moderator to change response function. Tamaki et al. [17] demonstrated volume-controlled liquid moderators, and Manabe et al. [24] proposed a water-phantom-type which is routinely used in QA procedure that evaluates neutron energy spectrum from depth-dependent count profiles.

However, this approach causes only modest changes in the response functions, making it difficult to generate epithermal neutron region peaks which limit the accuracy of neutron energy spectrum evaluation in BNCT.

(2.3) Active detectors with remotely changeable liquid moderator patterns

Shiraishi et al. [25] developed a remotely operated liquid-moderator spectrometer using concentric shells filled with water or boric acid solution, allowing multiple moderator patterns to be selected without entering the irradiation room and producing a high degree of flexibility in response-function generation.

Because this approach can generate many distinct response functions and offers excellent configurational freedom, it forms the basis for the method adopted in this study.

1.6 Challenges addressed in this study and proposed solutions

Conventional neutron spectrometers used in BNCT irradiation fields face several challenges. Figure 1.10 provides an overview of the major challenges addressed in this study and summarizes the solutions implemented to overcome them. This section provides detailed descriptions of each challenge and the corresponding solutions.

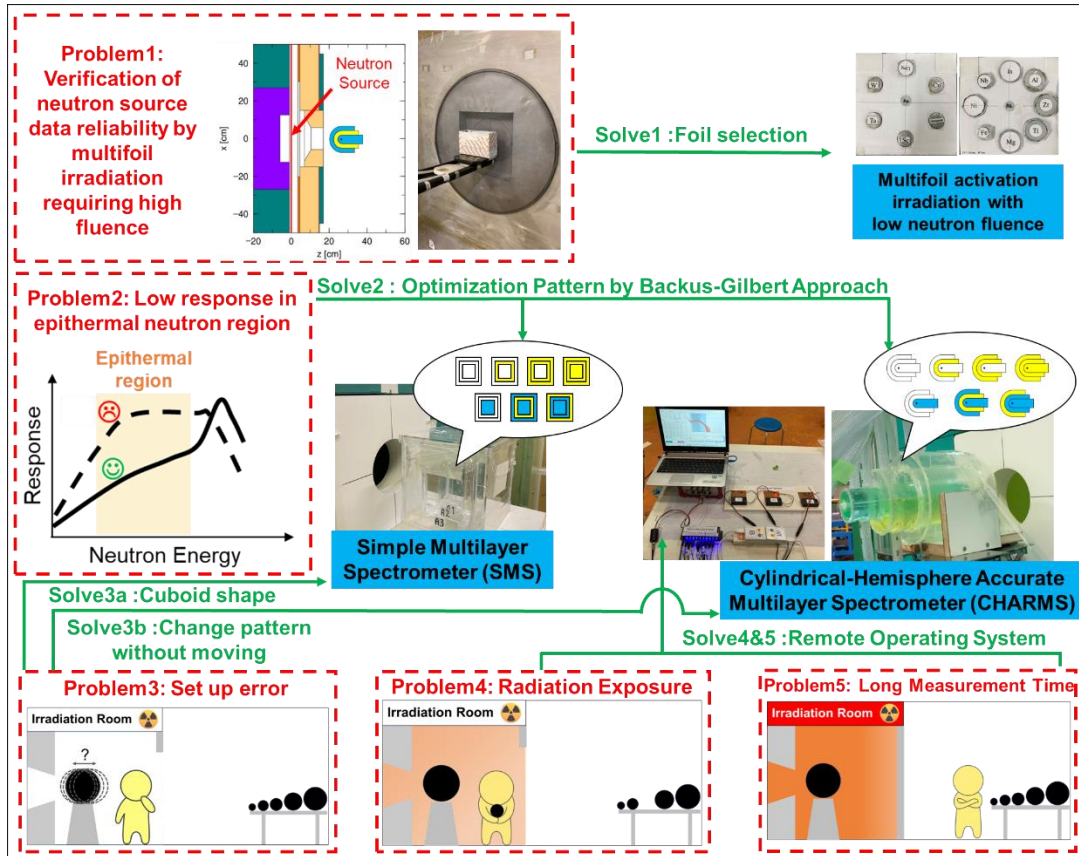


Figure 1.10: Challenges addressed in this study and proposed solutions diagram

1.6.1 Neutron source data verification by multifoil activation method

In the development of neutron spectrometers, reliable neutron source data are required for simulation calculations, including response function and initial guess spectra. In this study, the reliability of neutron source data was confirmed by using the multifoil activation method. In previous study, high neutron fluence irradiation was applied to obtain sufficient activation data. [6] However, such irradiation conditions are not practical for accelerator-based BNCT systems, where shorter irradiation times associated with lower neutron fluence are preferred.

In this study, an appropriate foil selection strategy was employed, improving the acquisition of sufficient measurement data for neutron energy spectrum evaluation even under low-fluence irradiation conditions.

1.6.2 Neutron spectrometer setup error

In conventional Bonner sphere spectrometer, moderator is made of solid materials. To change the spectrometer configuration, spheres with different radii must be exchanged, and the detector position must also be adjusted. These repeated mechanical changes can introduce setup errors, leading to a decrease in measurement accuracy. [19]

In this study, Simple Multilayer Spectrometer (SMS) with a cuboid-shaped structure is employed. This geometry is easier to set up than spherical moderators and reduces positioning errors. [26] In addition, Cylindrical-Hemisphere Accurate Remote Multilayer Spectrometer (CHARMS) system allows spectrometer configurations to be changed by remotely supplying and draining the liquid moderator without moving or replacing the spectrometer. The detector position also remains unchanged. As a result, setup errors are minimized, improving measurement accuracy and reproducibility.

1.6.3 Response function resolution in epithermal neutron region

Conventional Bonner sphere spectrometers exhibit limited sensitivity in the epithermal neutron energy region in BNCT fields, because their moderator configurations are optimized for general irradiation field rather than BNCT irradiation field.

In this study, thermal-neutron-absorbing materials containing ^{10}B (0.14 wt%) boric acid solution was introduced as one of liquid moderator to reduce the thermal neutron response and improve sensitivity to epithermal neutrons. Furthermore, by applying the Backus–Gilbert approach [27], [28], high spectral resolution over the entire neutron energy range is optimized, enabling more accurate neutron energy spectrum evaluation.

1.6.4 Radiation exposure

In conventional Bonner sphere spectrometers procedure, operators need to enter the irradiation room about six to seven times to adjust the moderator or detector, which is not desirable for radiation safety due to radiation exposure. [25]

In this study, operator exposure is reduced by using a remote operation system that combines a liquid moderator supply and drainage system with an active neutron detector based on LiCAF. By changing the spectrometer configuration and performing online neutron measurements, this system allows neutron spectrometer measurements to be carried out without entering the irradiation room during irradiation.

1.6.5 Measurement time

In conventional measurement procedures, operators cannot enter the irradiation room until radiation levels decrease to a safe level after beam shutdown. At KUR-HWNIF, this waiting time is

typically about 15 minutes after each irradiation. When the irradiation room must be entered six to seven times during a measurement session, more than one hour of experimental time is lost. [25]

In this research, by applying the remote operation system, this waiting time can be eliminated, allowing measurements to be performed more efficiently without entering the irradiation room.

1.7 Research overview

As described in Chapter 1.4, evaluating neutron energy spectrum in the irradiation field is important for BNCT, especially because neutron energy spectrum can vary depending on the neutron source operating conditions.

The aim of this research is to develop and validate a remote-operating liquid moderator neutron spectrometer system for use in BNCT irradiation fields that can overcome these limitations. The development was performed at KUR-HWNIF. To achieve this goal, a multi-step investigation was conducted:

To confirm the reliability of recently used neutron source data for simulation calculations in the development of neutron spectrometer, the conventional multifoil activation method was applied to evaluate neutron energy spectrum at KUR-HWNIF. In previous studies, high neutron fluence irradiation was employed to obtain sufficient activation data. However, such irradiation conditions are not practical for accelerator-based BNCT systems, where shorter irradiation times associated with lower neutron fluence are preferred. In this study, this issue is addressed by optimizing the selection and dimensions of activation foils, enabling the acquisition of sufficiently reaction rate suitable for neutron energy spectrum evaluation. (Chapter 2)

To achieve accurate neutron energy spectrum evaluation in BNCT irradiation field, a remote-operating neutron spectrometer was designed and optimized through Monte Carlo simulations. The Backus–Gilbert method was employed to select optimal combinations of multiple liquid moderators, enabling high-energy-resolution response functions and improving the accuracy of neutron energy spectrum evaluation compared with conventional method. (Chapter 3)

To verify the feasibility of multilayer neutron spectrometer, a proof-of-concept experiment was performed using a simplified prototype without remote operation system. The validation of this concept directly demonstrates the practicality of using multilayer liquid moderators for neutron energy spectrum evaluation in BNCT irradiation field. (Chapter 4)

To establish a practical system for BNCT QA procedure, a fully functional, remote-operating neutron spectrometer was developed and validated. A validation test confirmed that the system can measure neutron energy spectrum without requiring access to the irradiation room during measurement. This achievement demonstrates that the developed spectrometer effectively reduces radiation exposure while improving operational efficiency through quick and safe operation for BNCT. (Chapter 5)

Chapter 2 Validation of neutron source data for neutron spectrometer development

In this chapter, neutron energy spectrum at KUR-HWNIF was evaluated using the multifoil activation method to validate the neutron source data used for neutron spectrometer development. In this study, experimental design and foil selection were adapted to low neutron fluence conditions, which are also useful to accelerator-based BNCT irradiation fields. Response functions were calculated using Monte Carlo simulation with updated nuclear data libraries, and neutron energy spectrum was evaluated using unfolding techniques. The unfolded result showed variations from previous evaluation within acceptable uncertainty ranges for KUR-HWNIF operation, confirming the reliability of the neutron source data for spectrometer development.

2.1 Overview

Reliable neutron source data used in simulation calculations are important for determining response function and initial guess spectrum in neutron spectrometer development. Furthermore, in BNCT irradiation field, such data are fundamental to simulation calculations and are critical for clinical BNCT application.

However, neutron source data of KUR-HWINF for simulation calculation have the following two concerns that may have some variation.

(1) In February 2006, the use of high-enrichment uranium (HEU) fuel in the KUR was discontinued. Subsequently, in May 2010, low-enrichment uranium (LEU) fuel completely replaced HEU, and KUR has been operating with LEU fuel since then. Although previous study [29] reported that neutron energy spectrum at KUR-HWNIF did not change significantly after the fuel replacement, a highly accurate reevaluation of the neutron energy spectrum has not been performed. [30]

(2) The neutron energy spectrum was evaluated using Neutron Unfolding Code Package (NEUPAC) unfolding code [31], which employs energy-dependent group cross-section libraries for neutron dosimetry reactions based on Evaluated Nuclear Data File Version B-V (ENDF/B-V) data [32]. However, more up-to-date nuclear data libraries and advanced unfolding codes are currently available, which were not accessible at the time of the previous evaluation.

As summarized in Table 1.2, neutron energy spectrum evaluation in BNCT irradiation field has commonly been performed using the multifoil activation method. Most previous measurements were conducted in reactor-based BNCT facilities. Recently, in 2022, Y. Bae et al. [11] also reported multifoil activation measurements in accelerator-based BNCT system.

The multifoil activation method generally requires high-fluence neutron irradiation; for example, in KUR-HWNIF (reactor-based BNCT), irradiation with high neutron fluence with $1.45 \times 10^{14} \text{ cm}^{-2}$ was employed. [6] However, expectation for the widespread application of multifoil activation measurements in accelerator-based BNCT system in the future, lower neutron fluence irradiation (shorter irradiation time or lower accelerator power) condition is required. In this study, “foil selection method” is proposed to enable reliable neutron energy spectrum evaluation under reduced neutron

fluence irradiation conditions.

The main goal of this study is to confirm the reliability of neutron source data from KUR-HWNIF used in simulation calculations for neutron spectrometer development. Reliability of neutron source for simulation calculation is evaluated under lower neutron fluence irradiation conditions to enable this method at other BNCT facilities. Neutron energy spectrum evaluation at KUR-HWNIF with target variation of within 20% for epithermal neutron and 30% for fast neutron is applied to confirm the reliability of neutron source data.

2.2 Previous KUR-HWNIF neutron energy spectrum evaluation

The neutron source used in the simulation calculations for KUR-HWNIF was positioned near the bismuth gamma shielding surface, as shown in Figure 2.1(a), with the intensity distribution defined for each annular region. 11-group annular neutron source with a maximum radius of 86.25 cm was employed. This annular source distribution at the bismuth gamma shielding surface was calculated using Discrete Ordinates Transport (DOT) 3.5 code [33] with Japanese Evaluated Nuclear Data Library (JENDL) 3.3 [34] and discretized into 11 annular regions calculated by two-dimensional neutron transport calculations from the reactor core. Based on this neutron source data, neutron energy spectrum divided into 145 energy groups was calculated, as shown in NEUPAC input of Figure 2.1(b). [6], [35]

To confirm the reliability of neutron energy spectrum simulation calculation (NEUPAC input), multifoil activation irradiation experiments were conducted. The neutron energy spectrum at this location was irradiated with total neutron fluence of $1.45 \times 10^{14} \text{ cm}^{-2}$ using the set of foils listed in Table 2.1. The neutron energy spectrum was evaluated using the NEUPAC unfolding code, which employs energy-dependent group cross-section libraries for neutron dosimetry reactions based on ENDF/B-V data. The evaluated neutron energy spectrum is shown as NEUPAC output of Figure 2.1(b). Based on the NEUPAC unfolding results, the 11-group annular neutron source used for calculating the NEUPAC input spectrum was corrected, and the updated neutron source data were adopted for recent simulation calculations of KUR-HWNIF.

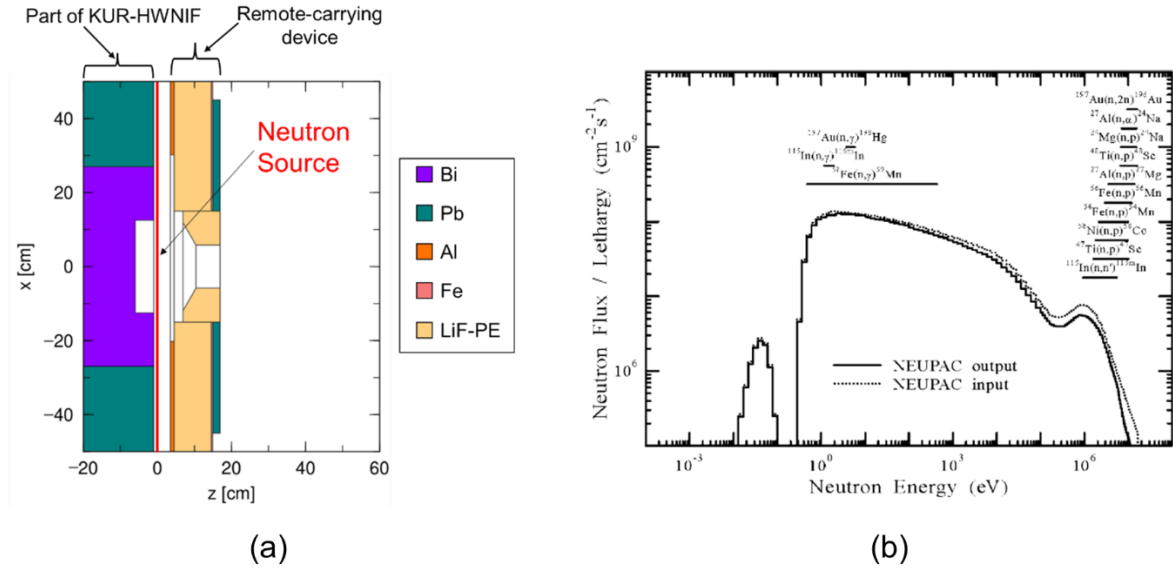


Figure 2.1: KUR-HWNIF neutron source

(a) geometry in calculation, (b) neutron energy spectrum at evaluation point

Table 2.1: Activation foils used in previous multifoil activation irradiation at KUR-HWNIF[6]

Foil	Abundance (%)	Reaction	γ-ray energy (keV) (emission rate)	Half-life
THERMAL NEUTRON CAPTURE				
¹⁹⁷ Au	100	¹⁹⁷ Au(n,γ) ¹⁹⁸ Au	411.8 (95.5%)	2.69 d
RESONANCE				
¹¹⁵ In	95.7	¹¹⁵ In(n,γ) ^{116m} In	1294(84.4%)	54.2 min
¹⁹⁷ Au	100	¹⁹⁷ Au(n,γ) ¹⁹⁸ Au	411.8 (95.5%)	2.69 d
⁵⁸ Fe	0.28	⁵⁸ Fe(n,γ) ⁵⁹ Fe	1291 (43.2%)	44.5 d
THRESHOLD				
¹¹⁵ In	95.7	¹¹⁵ In(n,n') ^{115m} In	336.3 (45.8%)	4.49 h
⁴⁷ Ti	7.3	⁴⁷ Ti(n,p) ⁴⁷ Sc	159.4 (68.2%)	3.34 d
⁵⁸ Ni	68.3	⁵⁸ Ni(n,p) ⁵⁸ Co	810.8 (99.5%)	70.9 d
⁵⁴ Fe	5.8	⁵⁴ Fe(n,p) ⁵⁴ Mn	834.8 (100.0%)	312 d
⁴⁶ Ti	8.0	⁴⁶ Ti(n,p) ⁴⁶ Sc	889.3 (100.0%)	83.8 d
²⁷ Al	100	²⁷ Al(n,p) ²⁷ Mg	1014 (29.1%)	9.46 min
⁵⁶ Fe	91.7	⁵⁶ Fe(n,p) ⁵⁶ Mn	846.8 (98.9%)	2.58 h
²⁴ Mg	79.0	²⁴ Mg(n,p) ²⁴ Na	1369 (100.0%)	14.7 h
²⁷ Al	100	²⁷ Al(n,α) ²⁴ Na	1369 (100.0%)	14.7 h
⁴⁸ Ti	73.8	⁴⁸ Ti(n,p) ⁴⁸ Sc	983.5 (100.0%)	1.82 d
¹⁹⁷ Au	100	¹⁹⁷ Au(n,2n) ¹⁹⁶ Au	355.6 (87.0%)	6.18 d
⁵⁸ Ni	68.3	⁵⁸ Ni(n,2n) ⁵⁷ Ni	1378 (77.9%)	1.50 d

2.3 KUR-HWNIF neutron energy spectrum evaluation in this study

Most activation foils used in multifoil activation method exhibit high (n, γ) reaction cross sections in the thermal neutron region due to their $1/v$ dependence, while their cross sections in the epithermal neutron region are lower because of resonance absorption. As a result, the large thermal neutron contribution tends to overshadow the responses from inelastic scattering, threshold reactions, or neutron production reactions.

In nuclear fission reactors [36], [37] with low epithermal neutron flux, cadmium-covered activation foils are commonly employed to suppress thermal neutron contributions, allowing evaluations focused primarily on fast neutrons.

In contrast, neutron spallation sources or fast-neutron reactors [38], [39] generate a large fraction of fast neutrons, which simplifies neutron energy spectrum evaluation because the influence of thermal and epithermal neutrons is relatively small.

However, in BNCT irradiation fields, even when activation foils are covered with cadmium, the overshadowing effect on fast neutron reactions cannot be sufficiently reduced. This is because cadmium does not effectively absorb epithermal neutrons, which constitute the dominant component of the neutron energy spectrum in BNCT irradiation fields. To address this issue, a foil selection method was introduced in this study, and its workflow is shown in Figure 2.2.

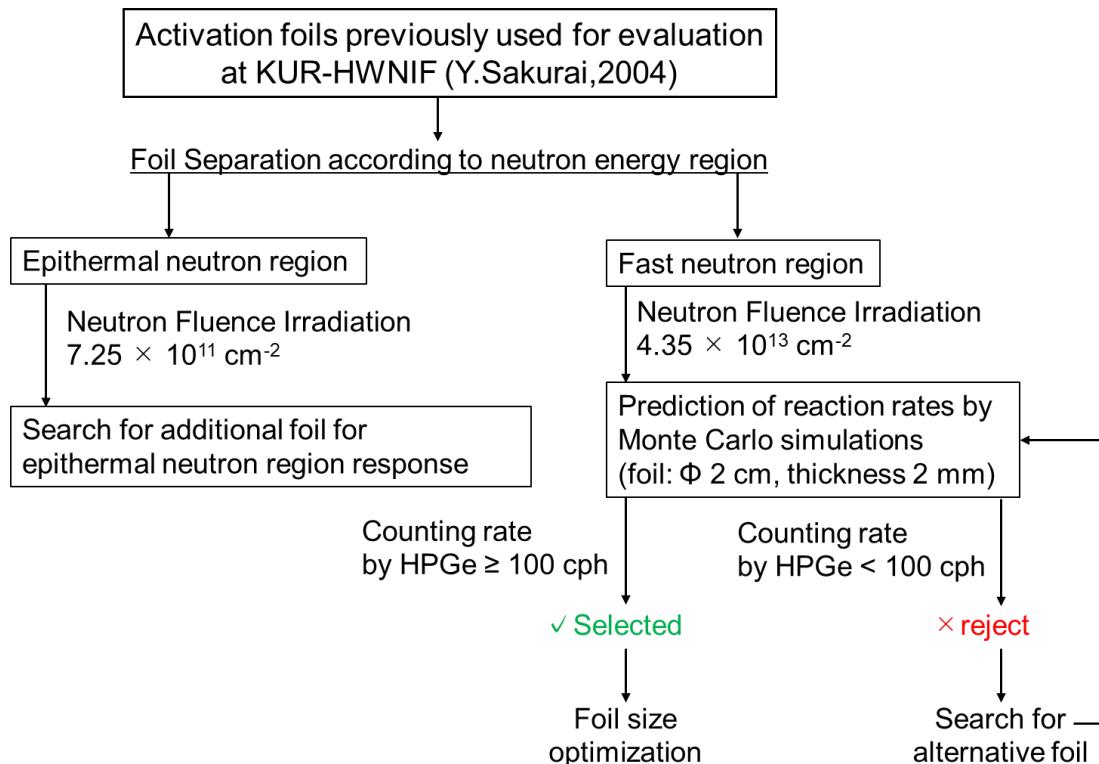


Figure 2.2: Foil selection method diagram

First, activation foils previously used for neutron energy spectrum evaluation at KUR-HWNIF were reviewed based on earlier studies. The foils were then divided into two sets according to their neutron energy sensitivity: one set sensitive to epithermal neutrons and the other sensitive to fast neutrons.

Because epithermal neutrons constitute the dominant component of BNCT irradiation fields, foils sensitive to epithermal neutrons were readily activated, allowing sufficient counting statistics to be obtained even under low-fluence irradiation conditions. In contrast, fast neutrons represent a relatively minor component of the neutron energy spectrum, and activation foils sensitive to fast neutrons therefore require higher neutron fluence to achieve adequate counting rates.

Table 2.2 summarizes the irradiation conditions, neutron fluence, and KUR operation for the epithermal- and fast-neutron evaluation foil sets in this study, compared with the previous neutron energy spectrum evaluation.

Table 2.2: Activation foil irradiation condition

	Neutron fluence (cm ⁻²)	KUR operation
Previous evaluation	1.45×10^{14}	5 MW 10 h
Epithermal neutron evaluation foil set	7.25×10^{11}	1 MW 15 min
Fast neutron evaluation foil set	4.35×10^{13}	1 MW 15 h

In previous evaluations, the epithermal-neutron-sensitive activation foil set consisted of only three reactions, $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$, $^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$ and $^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$ which was insufficient to adequately cover the entire epithermal neutron energy region. In the present study, the activation foil selection was expanded to improve sensitivity and energy coverage in the epithermal region.

Among the foils used in previous studies, $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ reaction was retained because of most widely used in multifoil activation method. In contrast, $^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$ reaction was excluded due to the unavailability of the material, and $^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$ reaction was omitted because its excessive activation under BNCT irradiation conditions resulted in overly high counting rates which can give high radiation exposure.

To compensate for these limitations, five additional activation foils were newly introduced: $^{23}\text{Na}(n,\gamma)^{24}\text{Na}$, $^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$, $^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$, $^{181}\text{Ta}(n,\gamma)^{182}\text{Ta}$, and $^{186}\text{W}(n,\gamma)^{187}\text{W}$. Table 2.3 shows six activation foils used in this study for evaluating epithermal neutron energy spectrum.

Table 2.3: Activation foils for epithermal neutron region evaluation

Foil	Reaction	Diameter (cm)	Thickness (μm)	γ -ray energy (keV) (emission rate)	Half-life
Au	$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	1.27	50	411.8 (95.5%)	2.69 d
NaCl	$^{23}\text{Na}(n,\gamma)^{24}\text{Na}$	1.27	1905	1369 (100%)	14.96 h
Manganin (11.5%Mn- Cu)	$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$	1.27	50	846.8 (98.9%)	2.58 d
Sc	$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	1.27	381	889.3 (100%)	83.8 d
Ta	$^{181}\text{Ta}(n,\gamma)^{182}\text{Ta}$	1.27	254	1121 (34.7%)	115 d
W	$^{186}\text{W}(n,\gamma)^{187}\text{W}$	1.27	127	686 (27.5%)	1 d

The counting rates of the candidate foils were evaluated assuming a standard foil geometry with a diameter of 2 cm and a thickness of 2 mm. Foils yielding counting rates higher than 100 counts per hour were selected for use, and their dimensions were further optimized if necessary. In contrast, foils with counting rates lower than 100 counts per hour were rejected, and alternative foil materials were subsequently investigated.

The threshold of 100 counts per hour corresponds to a statistical uncertainty of 10% based on Poisson statistics. This criterion was adopted because such an uncertainty level is required to ensure stable and reliable unfolding results; counting rates below this threshold lead to large statistical uncertainties, which cannot be properly handled by unfolding software used in this study.

Several activation reactions used in previous evaluations were retained in this study, including $^{115}\text{In}(n,n')^{115\text{m}}\text{In}$, $^{27}\text{Al}(n,p)^{27}\text{Mg}$, $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$, $^{47}\text{Ti}(n,p)^{47}\text{Sc}$, $^{48}\text{Ti}(n,p)^{48}\text{Sc}$, and $^{48}\text{Ti}(n,p)^{48}\text{Sc}$. These reactions provided stable responses under BNCT irradiation conditions and were therefore considered suitable for continued use. The foil dimensions were optimized in this study for enough reaction rate.

In contrast, several reactions were excluded from the present foil set due to limitations in counting statistics. The reactions $^{54}\text{Fe}(n,p)^{54}\text{Mn}$, $^{56}\text{Fe}(n,p)^{56}\text{Mn}$, $^{46}\text{Ti}(n,p)^{46}\text{Sc}$ and $^{58}\text{Ni}(n,p)^{58}\text{Co}$ were predicted to yield fewer than 100 counts per hour, even when measurements were initiated immediately after irradiation, and were therefore considered unsuitable under low neutron fluence irradiation conditions.

The $^{197}\text{Au}(n,2n)^{196}\text{Au}$ reaction, which emits a 355.6 keV gamma ray and yielded more than 100 counts per hour, was also excluded. This exclusion was due to $^{197}\text{Au}(n,2n)^{196}\text{Au}$ peak interference with the $^{197}\text{Au}(n,2n)^{198}\text{Au}$ reaction, which emits a gamma ray at 411.8 keV. The proximity of these gamma-ray peaks made it difficult to clearly separate the two contributions, thereby preventing accurate quantification of the $^{197}\text{Au}(n,2n)^{196}\text{Au}$ reaction.

To improve the sensitivity and energy coverage of the activation foil set, two additional reactions were newly introduced: $^{27}\text{Mg}(n,p)^{24}\text{Na}$ and $^{90}\text{Zr}(n,2n)^{89}\text{Zr}$. In particular, the $^{90}\text{Zr}(n,2n)^{89}\text{Zr}$ reaction was selected to enhance sensitivity to high-energy neutrons above approximately 10 MeV, which is important for evaluating the fast neutron component in BNCT irradiation fields.

Table 2.4: Activation foils for fast neutron region evaluation

Foil	Reaction	Diameter (cm)	Thickness (μm)	γ -ray energy (keV) (emission rate)	Half-life
Au	$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	1.27	50	411.8 (95.5%)	2.69 d
In	$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	2	100	336.3 (45.8%)	4.49 h
Al	$^{27}\text{Al}(n,p)^{27}\text{Mg}$	2	1000	1014 (100%)	9.46 m
	$^{27}\text{Al}(n,a)^{24}\text{Na}$			1369 (100%)	14.7 h
Mg	$^{27}\text{Mg}(n,p)^{24}\text{Na}$	2	200	1369 (100%)	14.7 h
Ti	$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	2	1500	159.4 (68.2%)	3.34 d
	$^{48}\text{Ti}(n,p)^{48}\text{Sc}$			983.5 (100%)	1.82 d
Zr	$^{90}\text{Zr}(n,2n)^{89}\text{Zr}$	2	2000	909.2 (99.0%)	3.27 d
Ni	$^{58}\text{Ni}(n,p)^{58}\text{Co}$	2	508	810.8 (99.5%)	70.9 d

2.4 Method

2.4.1 Experiment setup

All the foils were covered with 0.5-mm-thick cadmium during irradiation to mitigate the effects of thermal neutrons. The foils used in the measurements were positioned radially, with equal spacing between each foil. Because the neutron flux at the KUR-HWNIF may vary owing to the conditions in the KUR core, such as the fuel combustion and control-rod position, a gold foil with a 0.5-mm-thick cadmium cover was placed at the center of the foil attachment to monitor the neutron flux and ensure that both epithermal and fast neutron irradiation foil sets received equal irradiation with the same neutron flux as shown in Figure 2.3.

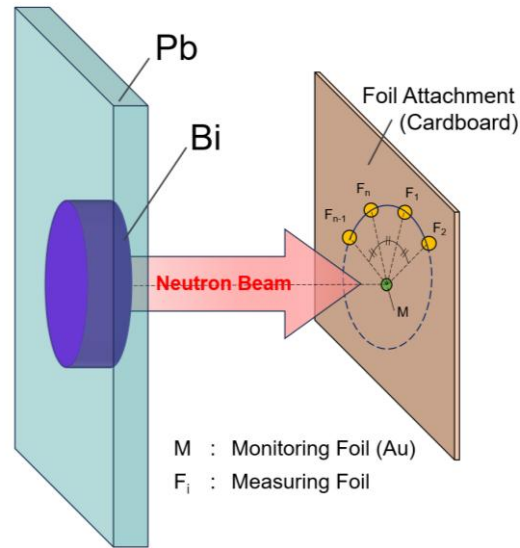


Figure 2.3: Foil arrangement during irradiation

Figure 2.4(a) shows the geometry and photograph near the irradiation field of KUR-HWNIF during the multifoil irradiation experiment. As shown in Figure 2.4(b), the evaluation point for these spectra is positioned at (0,0), which was near the bismuth gamma-ray shielding surface.

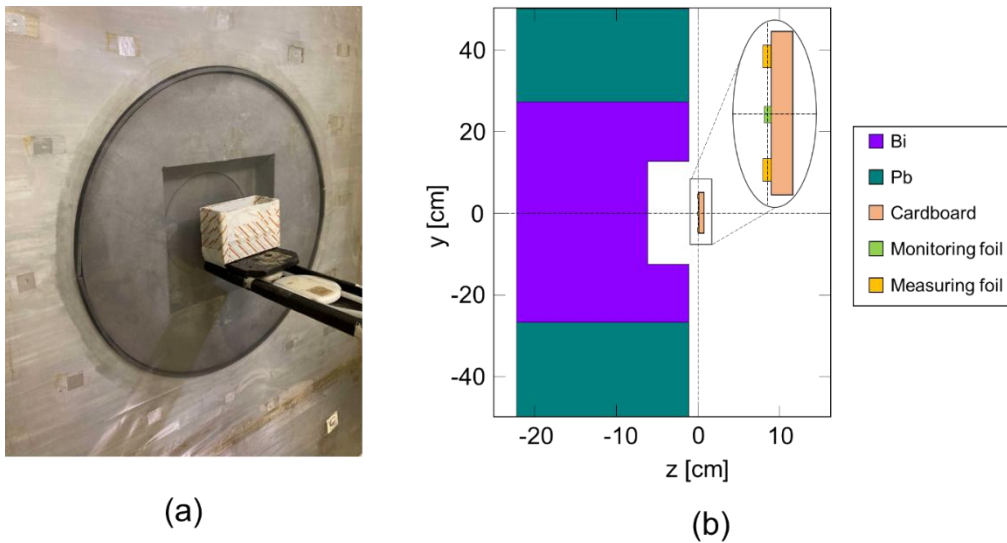


Figure 2.4: Experiment setup

(a) photograph (b) vertical cross-section

2.4.2 Activation foil measurement

The activation foil method allows the measurement of saturated activity based on the radiation induced in the foil through interactions with neutrons. Induced nuclides are generated by neutron reactions such as capture, elastic scattering, or inelastic scattering. Since these induced nuclides are unstable, they undergo radioactive decay to reach the ground state. In this study, the gamma rays emitted during decay were measured using HPGe detector to calculate reaction rate.

The reaction rate of activation foil is calculated using the following equation [40]:

$$R = \frac{\lambda T_{real} C / T_{live}}{\varepsilon I_{\gamma} e^{-\lambda T_w} (1 - e^{-\lambda T_{irrad}}) (1 - e^{-\lambda T_{real}}) M} \quad (2.1)$$

C	:	Net peak count of gamma ray
T_{irrad}	:	Irradiation time
T_w	:	Cooling time between end of irradiation and start of HPGe detector measurement
$T_{real, live}$	:	Real time and live time of HPGe detector measurement
λ	:	Decay constant
I_{γ}	:	Emission probability of gamma ray
ε	:	Full-energy peak efficiency of HPGe detector
M	:	Mass of activation foil

The detection efficiency, ε , of HPGe detector appearing in Equation (E.1) which depends on emitted gamma ray energy was evaluated experimentally; the detailed procedure is described in Appendix E.

2.4.3 Response function

The response functions of each activation were calculated using the multiplier function in Particle and Heavy Ion Transport code System (PHITS) version 3.33 [41], with simulation geometry set to be identical to experimental irradiation setup.

Based on the neutron energy group structure obtained from the previous spectrum evaluation, monochromatic neutron beams with 145 discrete energy groups were used as incident neutron sources. The JENDL-5 [42] nuclear data library was employed for most reaction calculations. However, for the evaluation of $^{115}\text{In}(n, n')^{115m}\text{In}$ reaction, the 532DOS2 [43] nuclear data library was used due to absence of corresponding data in JENDL-5.

Figure 2.5 and Figure 2.6 show the calculated response functions for epithermal and fast neutron evaluation foil set, respectively.

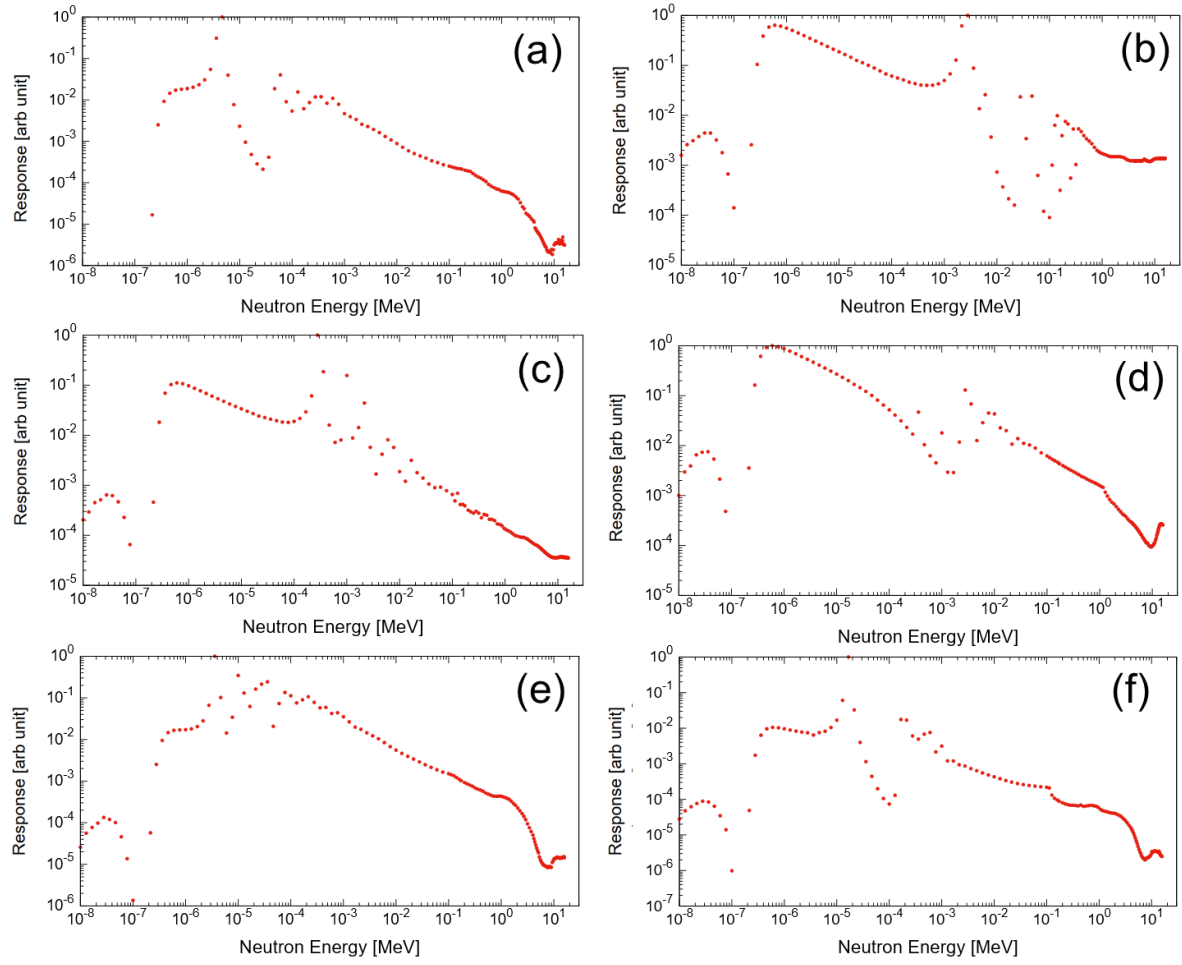


Figure 2.5: Response function calculation results of epithermal neutron evaluation foil

(a) $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$, (b) $^{23}\text{Na}(n,\gamma)^{24}\text{Na}$, (c) $^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$,
 (d) $^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$, (e) $^{181}\text{Ta}(n,\gamma)^{182}\text{Ta}$, (f) $^{186}\text{W}(n,\gamma)^{187}\text{W}$

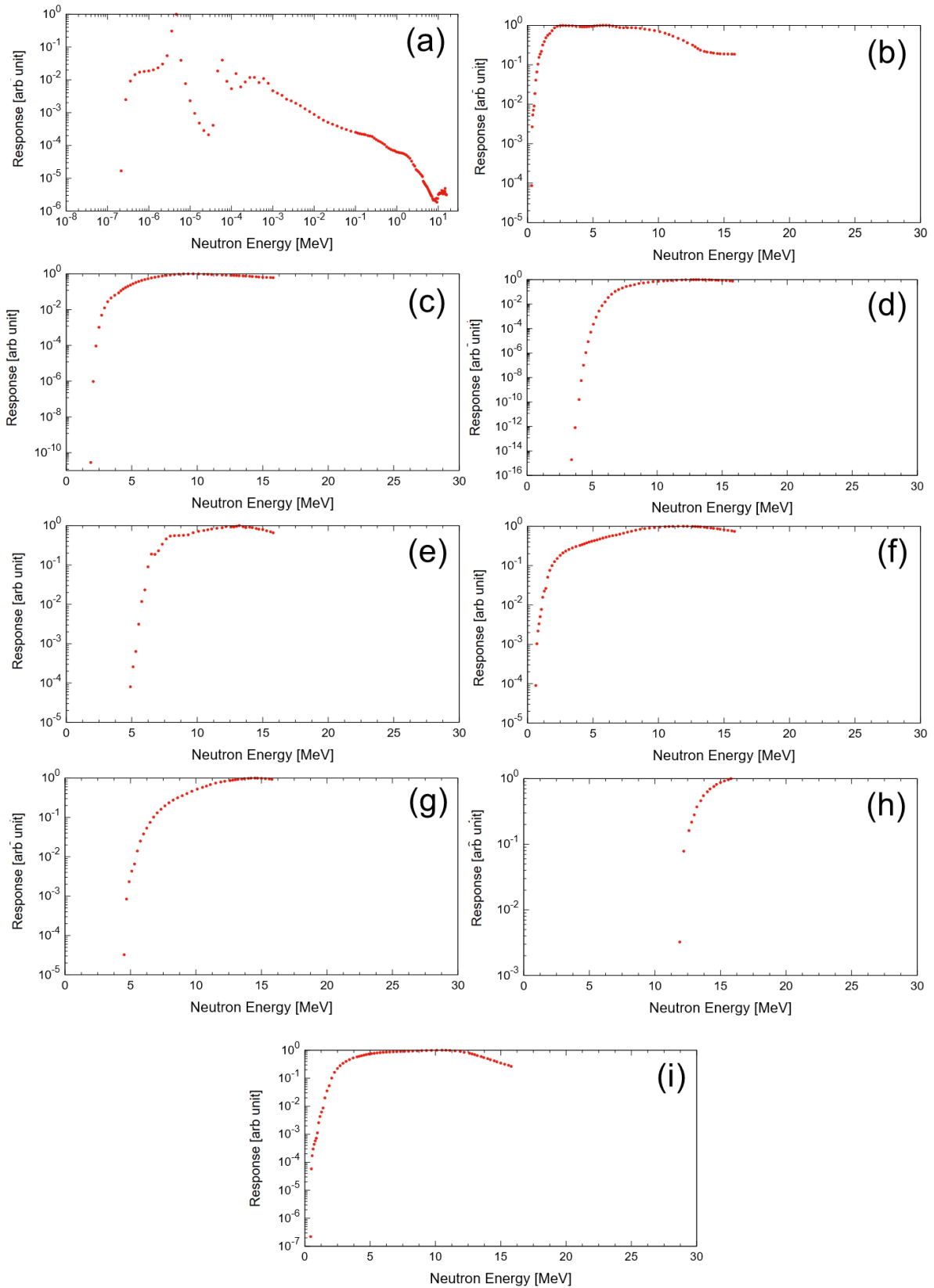


Figure 2.6: Response function calculation results of fast neutron evaluation foil

(a) $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$, (b) $^{115}\text{In}(n,n')^{115\text{m}}\text{In}$, (c) $^{27}\text{Al}(n,p)^{27}\text{Mg}$, (d) $^{27}\text{Al}(n,a)^{24}\text{Na}$, (e) $^{27}\text{Mg}(n,p)^{24}\text{Na}$,
 (f) $^{47}\text{Ti}(n,p)^{47}\text{Sc}$, (g) $^{48}\text{Ti}(n,p)^{48}\text{Sc}$, (h) $^{90}\text{Zr}(n,2n)^{89}\text{Zr}$, (i) $^{58}\text{Ni}(n,p)^{58}\text{Co}$

2.4.4 Unfolding procedure

The unfolding procedure is the process of evaluating the neutron energy spectrum $\phi(E)$ from the measured results M_i . In the activation foil method, the measurement result is defined as the saturated activity per unit mass of the foil, it is defined as the measured neutron reaction rate.

The problem of obtaining neutron spectrum from measurement result is formulated using a set of measurement data M_i ($i = 1, 2, \dots, m$)

$$M_i = \int R_i(E) \phi(E) dE \quad (2.2)$$

$R_i(E)$ is response function (or nuclear cross section) that can be calculated by using Monte Carlo simulation. To solve numerically, the equation is discretized into n energy bins, yielding:

$$M_i = \sum_{j=1}^n R_{ij} \phi_j \quad \text{where} \quad \phi_i = \int_{E_j}^{E_{j+1}} \phi(E) dE \quad (2.3)$$

Since this system is typically underdetermined ($n < m$), the solution is not unique, and unfolding methods must be applied to obtain a stable and physically meaningful neutron energy spectrum.

The experimental and calculated results were compared using the E/C ratio. For the calculated results, two cases were considered: one before and one after the unfolding procedure. The neutron energy spectrum for each case was input into Equation (2.3) to obtain the corresponding response for comparison with the experimental data.

In this study, we use the UMG (Unfolding with MAXED and GRAVEL) package, developed by Physikalisch-Technische Bundesanstalt (PTB) and distributed by the Nuclear Energy Agency (NEA), which contains two well-established unfolding codes: MAXED and GRAVEL [44]. Noted that both MAXED and GRAVEL code required prior information as initial guess spectrum ϕ_j^{DEF} which can be calculated by using Monte Carlo simulation.

● MAXED

Maximum Entropy Deconvolution (MAXED) is an unfolding code based on the maximum entropy principle. It aims to find neutron energy spectrum solution that reproduces the measurement data while remaining as close as possible to a given initial guess spectrum.

To achieve this, MAXED determines the solution spectrum ϕ_j by maximizing the entropy subject to two constraints: (1) the calculated responses reproduce the measurement data, and (2) the overall deviations between measurement and calculation result correspond to the experimental uncertainties.

The objective (entropy) function [45], [46] to be maximized is

$$S = - \sum_j \left\{ \phi_j \ln \left(\frac{\phi_j}{\phi_j^{DEF}} \right) + \phi_j^{DEF} - \phi_j \right\} \quad (2.4)$$

S becomes larger when the solution spectrum ϕ_j is closer to initial guess spectrum, thereby enforcing the principle of minimal deviation from the prior information.

The constraints are given as follows:

$$M_i + \epsilon_i = \sum_j R_{ij} \phi_j \quad (i = 1, \dots, m) \quad (2.5)$$

where, ϵ_i is the different between measurement and calculation.

$$\chi^2 = \sum_i \left(\frac{\epsilon_i}{\sigma_i} \right)^2 \quad (2.6)$$

where σ_i is the uncertainty of the measurement data, and χ^2 specifies the acceptable different between measurement and calculation for all data.

● GRAVEL

GRAVEL is an iterative unfolding method based on a modification of the SAND-II algorithm [47]. It minimizes the difference between the logarithms of the measured and calculated data using the following objective function:

$$\chi^2 = \sum_i \left(\frac{\ln M_i - \ln C_i^{(v)}}{\rho_i} \right)^2 \quad (2.7)$$

where v is iteration number, $\rho_i = \sigma_i / M_i$ is the relative standard deviation of the measurement data, and calculation data is defined as

$$C_i^{(v)} = \sum_j R_{ij} \exp(\ln \phi_j^{(v)}) \quad (2.8)$$

GRAVEL updates the logarithm of the neutron energy spectrum, $\ln \phi_j$, rather than ϕ_j itself, ensuring a positive and smoothly converging solution throughout the iterative unfolding process.

The neutron energy spectrum is updated iteratively using the formula:

$$\phi_j^{(v+1)} = \phi_j^{(v)} \exp \left\{ \frac{\sum_i W_{ij}^{(v)} \ln \left(\frac{M_i}{C_i^{(v)}} \right)}{\sum_i W_{ij}^{(v)}} \right\} \quad (2.9)$$

where, weighting factor $W_{ij}^{(v)}$ is defined as:

$$W_{ij}^{(v)} = \frac{C_i^{(v)}}{\sum_i C_i^{(v)}} \frac{M_i^2}{\sigma_i^2} \quad (2.10)$$

$W_{ij}^{(v)}$ represents the weight of detector i for energy bin j , combining both response function and the measurement reliability due to uncertainty. It determines how strongly each measured data point influences the spectrum update during iteration.

In GRAVEL, the iteration is automatically stopped when the value of χ^2 becomes reasonably small. In contrast, MAXED requires the user to specify the target χ^2 value manually. In this study, final χ^2 obtained from GRAVEL was used for the MAXED calculation.

2.5 Results and discussions

2.5.1 90% response function sensitive energy range

The characteristics of the response functions were examined using the 90% sensitive energy ranges. These ranges were determined by combining each response function with the initial guess spectrum, as shown in Figure 2.7.

Because of “foil selection method” in this study, sufficient response coverage was achieved even under low neutron fluence conditions. In particular, the introduction of the $^{23}\text{Na}(n,\gamma)^{24}\text{Na}$ and $^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$ reactions extended the epithermal neutron sensitivity down to 10^{-2} MeV, whereas previous evaluations were limited to approximately 10^{-3} MeV. Furthermore, the inclusion of the $^{90}\text{Zr}(n,2n)^{89}\text{Zr}$ reaction enabled sensitivity to neutron energies above 10 MeV. As a result, a broad and continuous response function coverage was obtained by combining different types of activation foils, except for the energy range from 10^{-2} to 1 MeV which has no available activation foil that can give response around this energy range.

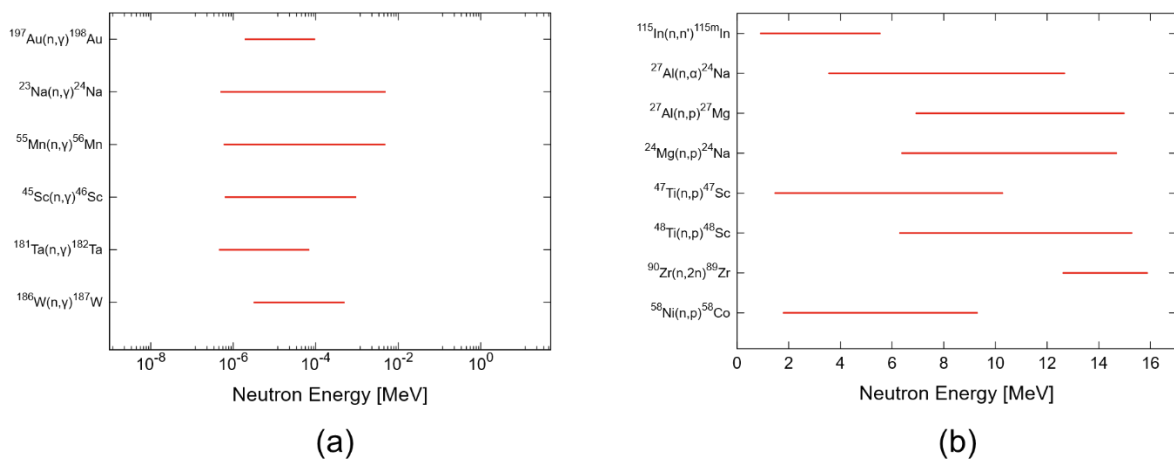


Figure 2.7: 90 % sensitive energy ranges

(a) epithermal neutron evaluation foil, (b) fast neutron evaluation foil

2.5.2 HPGe detector measurement

The gamma rays were measured using HPGe detector (ORTEC, GEM20P4-70). A multi-channel analyzer (Amptek, MCA-8000D) was used to analyze the gamma-ray spectra, receiving signals from the HPGe detector.

Figure 2.8 and Figure 2.9 show the gamma spectrum measurement results for epithermal and fast neutron evaluation foils, respectively, where the horizontal axis represents the gamma-ray energy and the vertical axis represents the counts.

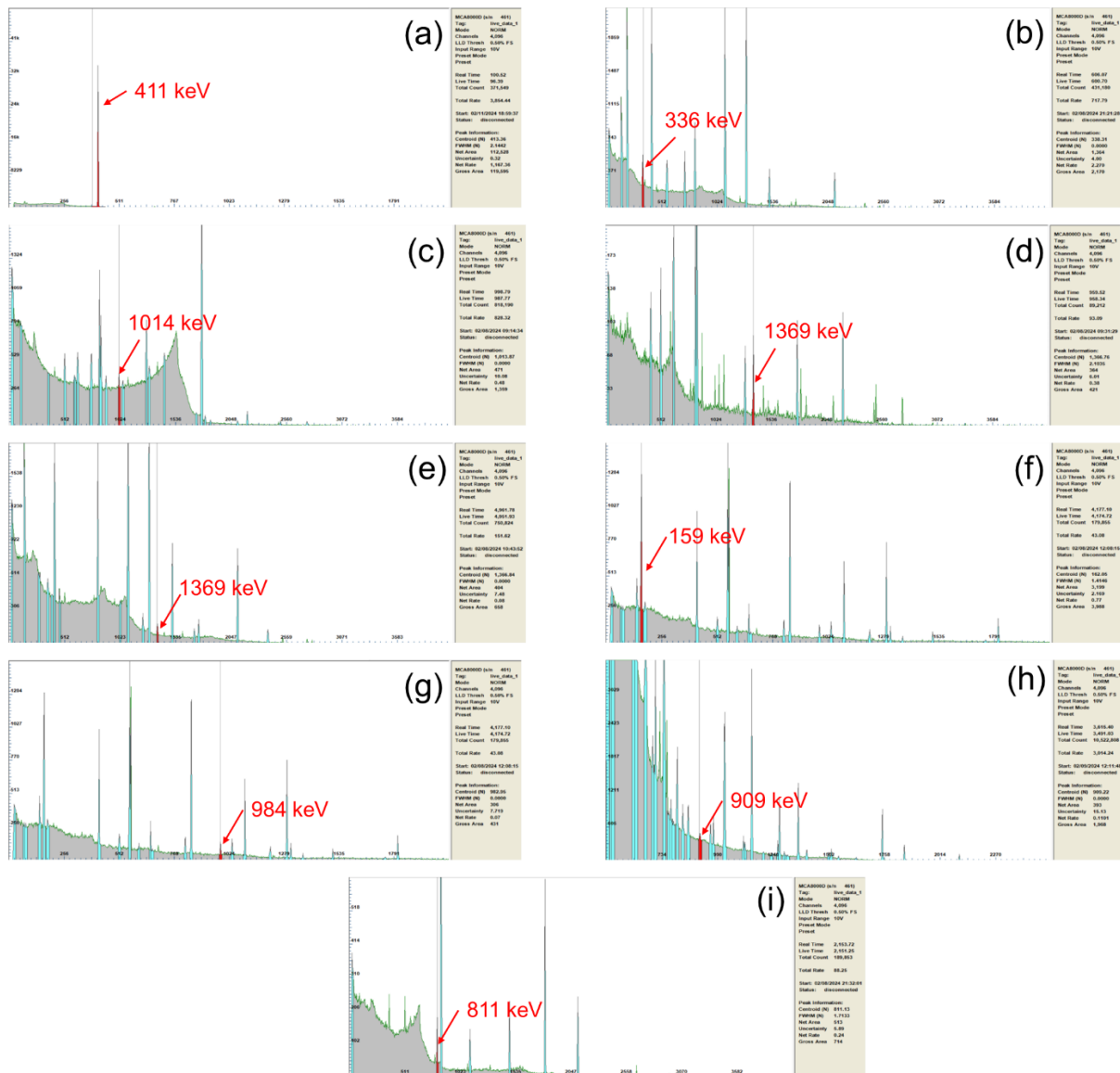


Figure 2.9: Gamma spectrum measurement results for fast neutron evaluation foil

(a) $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$, (b) $^{115}\text{In}(n,n')^{115\text{m}}\text{In}$, (c) $^{27}\text{Al}(n,p)^{27}\text{Mg}$, (d) $^{27}\text{Al}(n,a)^{24}\text{Na}$, (e) $^{27}\text{Mg}(n,p)^{24}\text{Na}$,
(f) $^{47}\text{Ti}(n,p)^{47}\text{Sc}$, (g) $^{48}\text{Ti}(n,p)^{48}\text{Sc}$, (h) $^{90}\text{Zr}(n,2n)^{89}\text{Zr}$, (i) $^{58}\text{Ni}(n,p)^{58}\text{Co}$

The HPGe detector measurement conditions and reaction rate epithermal and fast neutron-evaluation foils are presented in Table 2.5 and Table 2.6, respectively. For each foil, the data required to calculate reaction rate — including HPGe detector efficiency, foil weight, cooling time, measurement time, and gamma peak area — are provided. The measurement uncertainty recorded from the HPGe detector includes not only statistical uncertainty based on Poisson distribution, but also uncertainty from peak fitting procedures.

The reaction rate for both irradiation durations of the gold foil irradiation which used as neutron flux monitoring indicate no significant difference in the neutron flux between epithermal- and fast-neutron-sensitive activation foils.

Table 2.5: HPGe detector measurement condition and reaction rate result of epithermal neutron evaluation foils

Foil	Reaction	HPGe detector efficiency [%]	Weight [mg]	Cooling time [s]	Measurement time		Peak area	Reaction Rate [s ⁻¹ g ⁻¹]	Measurement uncertainty [%]
					T _{real} [s]	T _{live} [s]			
Au	¹⁹⁷ Au(n,γ) ¹⁹⁸ Au	6.15	7.12	3.33 × 10 ³	242.26	241.8	12017	4.48 × 10 ⁷	0.92
NaCl	²³ Na(n,γ) ²⁴ Na	1.57	475.55	8.93 × 10 ⁴	7206.43	7202.73	13421	1.82 × 10 ⁵	0.91
Manganin (11.5%Mn-Cu)	⁵⁵ Mn(n,γ) ⁵⁶ Mn	2.71	52.94	1.10 × 10 ³	299.94	298.74	10266	3.56 × 10 ⁶	1.02
Sc	⁴⁵ Sc(n,γ) ⁴⁶ Sc	2.56	143.18	1.55 × 10 ³	663.11	662.9	934	4.46 × 10 ⁶	3.43
Ta	¹⁸¹ Ta(n,γ) ¹⁸² Ta	1.97	510.82	2.34 × 10 ³	847.53	845.54	3207	1.73 × 10 ⁷	1.89
W	¹⁸⁶ W(n,γ) ¹⁸⁷ W	3.44	361.01	3.64 × 10 ³	116.72	114.42	13913	1.81 × 10 ⁷	0.86

Table 2.6: HPGe measurement condition and reaction rate result of fast neutron evaluation foils

Foil	Reaction	HPGe detector efficiency [%]	Weight [mg]	Cooling time [s]	Measurement time [s]		Peak area	Reaction Rate [s ⁻¹ g ⁻¹]	Measurement uncertainty [%]
					T _{real} [s]	T _{live} [s]			
Au	¹⁹⁷ Au(n,γ) ¹⁹⁸ Au	6.15	7.25	2.94 × 10 ⁵	1.01 × 10 ²	9.64 × 10 ¹	112528	4.43 × 10 ⁷	0.32
In	¹¹⁵ In(n,n') ^{115m} In	7.75	241.69	4.38 × 10 ⁴	6.07 × 10 ²	6.06 × 10 ²	1364	2.01 × 10 ³	4.00
Al	²⁷ Al(n,p) ²⁷ Mg	2.21	853.44	1.63 × 10 ²	9.99 × 10 ²	9.88 × 10 ²	471	1.84 × 10 ²	10.08
	²⁷ Al(n,α) ²⁴ Na	1.57		1.18 × 10 ³	9.60 × 10 ²	9.58 × 10 ²	364	5.71 × 10 ¹	6.01
Mg	²⁷ Mg(n,p) ²⁴ Na	1.57	101.35	5.52 × 10 ³	4.96 × 10 ³	4.95 × 10 ³	404	1.42 × 10 ²	7.48
Ti	⁴⁷ Ti(n,p) ⁴⁷ Sc	18.10	1870.07	1.06 × 10 ⁴	4.18 × 10 ³	4.17 × 10 ³	3199	3.85 × 10 ²	2.17
	⁴⁸ Ti(n,p) ⁴⁸ Sc	2.29		1.06 × 10 ⁴	4.18 × 10 ³	4.17 × 10 ³	306	1.16 × 10 ¹	7.71
Zr	⁹⁰ Zr(n,2n) ⁸⁹ Zr	2.50	3158.35	9.72 × 10 ⁴	3.62 × 10 ³	3.49 × 10 ³	393	2.87 × 10 ¹	15.13
Ni	⁵⁸ Ni(n,p) ⁵⁸ Co	2.85	1425.02	4.44 × 10 ⁴	2.15 × 10 ³	2.15 × 10 ³	513	1.43 × 10 ³	5.89

2.5.3 Neutron energy spectrum evaluation

Figure 2.10 shows unfolded neutron energy spectra from the UMG unfolding package using the MAXED and GRAVEL codes compared with initial guess spectrum which is previous neutron energy spectrum evaluation result for KUR-HWNIF.

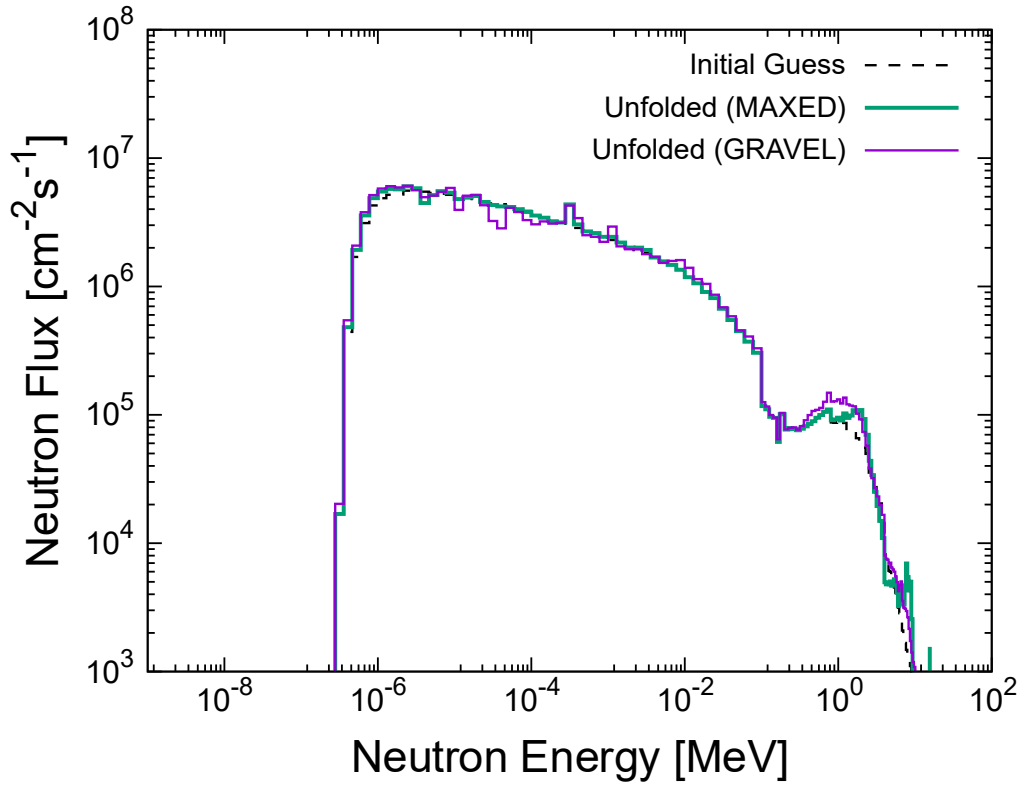


Figure 2.10: Evaluated neutron energy spectrum using UMG unfolding package

In MAXED and GRAVEL, before the unfolding process, it is possible to choose whether to apply a scaling factor — determined by the code based on the measured data — to the entire spectrum. In this study, the scaling factor obtained from MAXED and GRAVEL was 0.93, indicating that the evaluated total neutron energy spectrum was about 7% lower than the previous one. At KUR, the uncertainties in the nominal values are approximately 20% for thermal neutron flux and around 30% for epithermal and fast neutron fluxes [6]; therefore, 7% difference is considered acceptable.

In the MAXED unfolding results, irregularities were observed in the neutron energy spectrum at approximately 10 MeV, a common phenomenon that has been observed in other studies [48], [49]. One possible reason for these irregularities in the unfolded spectrum is the large measurement uncertainty of the high-fluence irradiation foils used to evaluate the neutron energy spectrum in the fast neutron region, compared to the epithermal neutron evaluation irradiation foils. This problem could be addressed by increasing the irradiation time for fast neutron evaluation irradiation foils, thereby reducing the measurement uncertainty.

Based on the unfolding results, the epithermal- and fast-neutron fluxes are summarized in Table 2.7. In the epithermal neutron region, the neutron flux received from the MAXED and GRAVEL unfolding decreased by 4.23% and 6.12%, respectively. In contrast, in the fast- neutron region, the neutron flux received from the MAXED unfolding decreased by 4.34 %, whereas that received from the GRAVEL unfolding increased by 5.62%. According to Y.Sakurai et al. [6], the uncertainties in the nominal values are almost 30% for the epithermal- and fast-neutron fluxes, indicating that these unfolding results are acceptable.

Table 2.7: Unfolding results of epithermal- and fast-neutron fluxes and neutron absorption dose rate

Spectrum	Epithermal neutron flux (cm ⁻² s ⁻¹)	Fast neutron flux (cm ⁻² s ⁻¹)	Neutron absorption dose rate (Gy s ⁻¹)
Initial guess	1.49×10^8	9.75×10^6	8.59×10^{-5}
MAXED unfolding	1.43×10^8 (−4.23%) ¹	9.33×10^6 (−4.34%)	8.83×10^{-5} (−2.76%)
GRAVEL unfolding	1.40×10^8 (−6.12%)	1.03×10^7 (+5.62%)	9.73×10^{-5} (+10.16%)

The neutron absorption dose rate at the evaluation point position was also calculated. As we consider the point evaluation, the issue of electronic equilibrium can be neglected. The results are presented in Table 2.7. The DS02 kerma factor [50] was used for the calculations, which considers the composition of soft tissues of the body in ICRU Report 44 [51]. The neutron energy spectrum from the GRAVEL unfolding results, which showed a better representation of the reevaluated neutron spectrum of KUR-HWNIF, yielded a 10.16% higher neutron absorption dose than that of the previous evaluation. For the GRAVEL unfolding code results, the epithermal neutron flux and fast neutron flux have increased; however, the neutron absorption dose has decreased. The reason is that at energies over 1 MeV, the DS02 kerma factor increases rapidly, indicating that neutrons with 1 MeV are dominant for the neutron absorption dose. Neutron energies exceeding 1 MeV exhibit a lower flux per lethargy compared to the previous evaluation, which explains why the neutron absorption dose has decreased from the previous evaluation. This is further evidence of the importance of evaluating the neutron energy spectrum, not just the neutron flux in each neutron energy region.

¹ Percentage variation from the initial guess spectrum

2.5.4 E/C ratio

E/C ratio of epithermal and fast neutron sensitive foil are presented in Figure 2.11 and Figure 2.12 respectively.

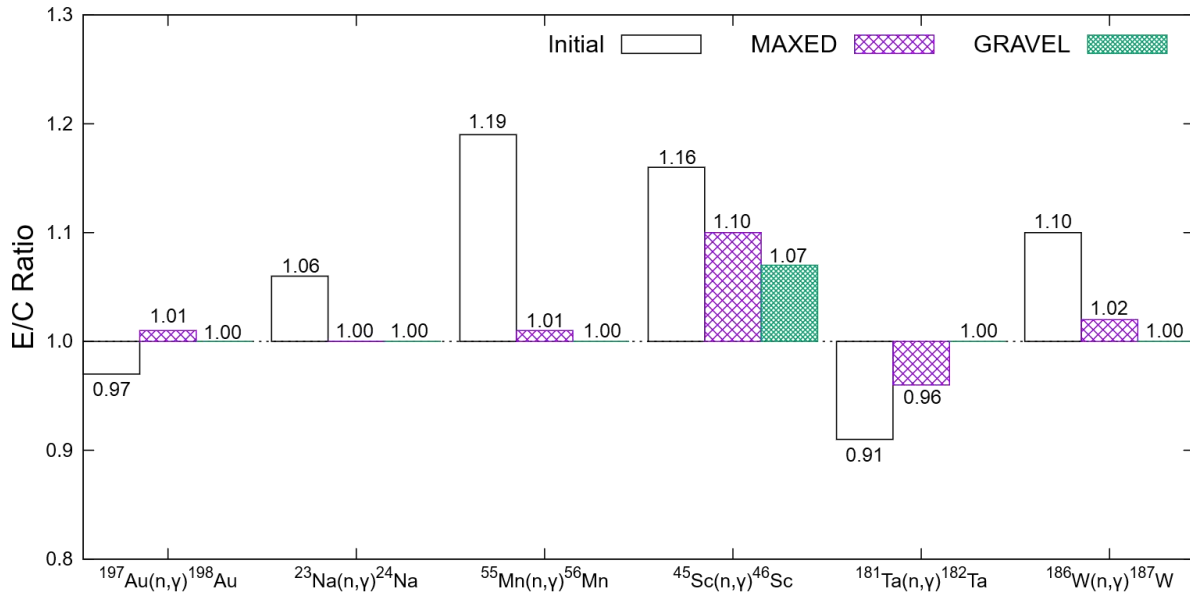


Figure 2.11: E/C ratio for epithermal neutron sensitive foil

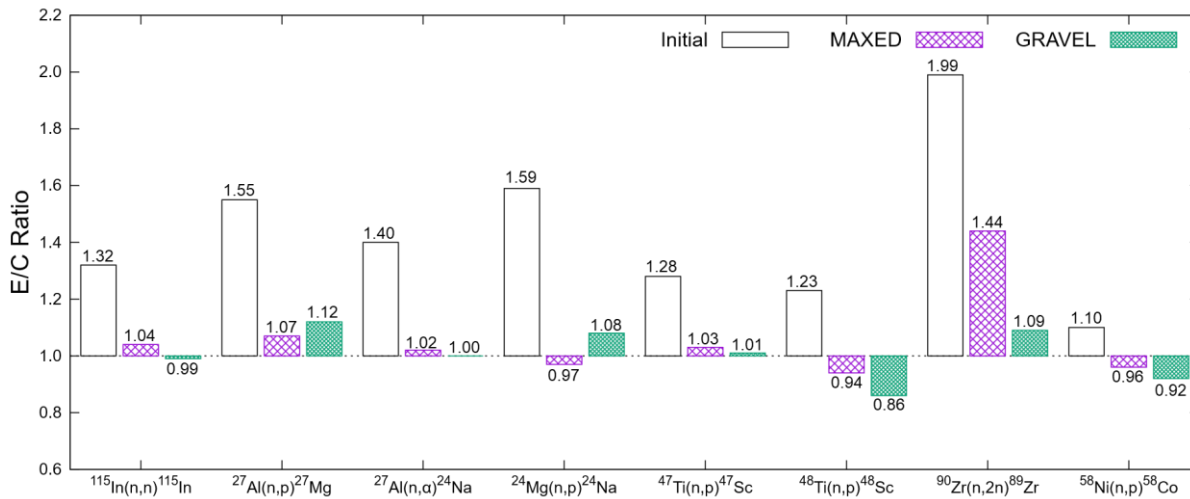


Figure 2.12: E/C ratio for fast neutron sensitive foil

The E/C ratios of the unfolding results were close to 1.0. This trend indicates that the unfolding process was effective in adjusting the neutron energy spectrum to align with the experimental results. In the MAXED unfolding results, the E/C ratio for $^{90}\text{Zr}(n,2n)^{89}\text{Zr}$ was significantly far from 1.0 compared to the GRAVEL results. Thus, GRAVEL exhibited a better ability than MAXED in adjusting the neutron energy spectrum. In addition, irregularities were observed in the neutron energy spectrum of the MAXED unfolding results but not in the GRAVEL results. The fast neutron

evaluation irradiation foils used contain a large measurement uncertainty. Specifically, the foil used for measuring $^{90}\text{Zr}(n,2n)^{89}\text{Zr}$ exhibited the largest measurement uncertainty at 15.13 %. We consider that the MAXED unfolding code is more sensitive to large measurement uncertainties compared to GRAVEL, as shown by the neutron energy spectrum results and the E/C ratio for $^{90}\text{Zr}(n,2n)^{89}\text{Zr}$. In the BNCT irradiation field, in the fast neutron region, measurement data with large measurement uncertainties need to be used as information for the unfolding process. To achieve reasonable unfolded results, the GRAVEL unfolding code better represents neutron energy spectrum of the KUR-HWNIF.

2.6 Conclusion

To confirm the reliability of neutron source data from KUR-HWNIF under low neutron fluence conditions, the neutron energy spectrum was evaluated using the multifoil activation method with “foil selection method”.

Because the unfolding analysis performed with the UMG package cannot adequately account for large statistical uncertainties in the measured activities, foils expected to yield poor counting statistics were excluded at the selection stage. Specifically, a threshold of 100 counts per hour (10% statistical uncertainty based on Poisson statistics) was applied to screen out reactions with excessively large counting uncertainties. This approach enabled reliable spectrum evaluation under reduced neutron fluence conditions, with the neutron fluence decreased by approximately 70%, making it possible for application in accelerator-based BNCT irradiation fields.

The neutron energy spectrum obtained through unfolding analysis showed 10% maximum variations compared to the initial guess, i.e., the spectrum before the fuel low-enrichment. These variations were which is within the target uncertainty ranges for KUR-HWNIF operation (within 20% for epithermal neutrons and 30% for fast neutrons). These results demonstrate that reliable neutron source data for simulation calculation can be used for neutron spectrometer development and other simulation-related BNCT research.

Chapter 3 Response function optimization for liquid multilayer neutron spectrometer

In this chapter, the optimization study of the Cylindrical Hemisphere-Accurate Remote Multilayer Spectrometer (CHARMS) is described. In the conventional Bonner sphere spectrometer, the neutron energy spectrum is evaluated by replacing solid moderators of different thicknesses. However, an alternative approach using multilayer liquid moderators is proposed. Building on this concept, CHARMS employs multilayer liquid moderators and applies the Backus–Gilbert method to select optimal moderator combinations, thereby achieving response functions with higher energy resolution. Simulation calculation results demonstrate that, for BNCT irradiation fields, the performance of CHARMS with Backus–Gilbert–based optimization shows superior performance compared to the conventional Bonner sphere method.

3.1 CHARMS structure

Figure 3.1 shows the structure of the Cylindrical-Hemisphere Accurate Remote Multilayer Spectrometer (CHARMS). The system was designed to operate from outside the irradiation room by combining a liquid moderator with an active neutron detector. LiCAF neutron detector with dimensions of 0.3 mm × 0.3 mm × 0.3 mm is placed at the center, surrounded by three layers of liquid moderator.

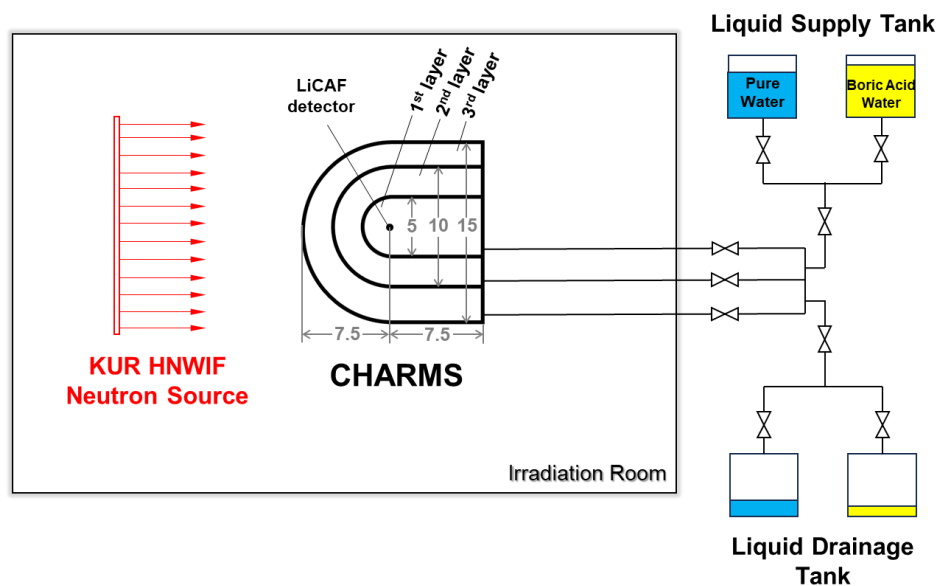


Figure 3.1: Structure of CHARMS with liquid moderator supply and drainage system (unit: cm)

3.1.1 Liquid moderator

Two types of liquids, pure water and a boric acid solution, were used as neutron moderators in the respective layers. In addition, an empty layer (air) was also considered, resulting in 27 ($= 3^3$) injection patterns for CHARMS.

Based on the solubility limit of boric acid in water, which is 47.2 g/L at 20 °C [52], the maximum achievable concentration, corresponding to 0.14 wt% ^{10}B boric acid, was adopted in this study.

3.1.2 Container

Acrylic was selected as the material for the liquid moderator containers in this study because it is transparent and widely used in BNCT QA procedures. Its transparency allows visual confirmation of the liquid moderator condition.

As shown in Figure 3.1, the container shapes are hemispherical and cylindrical, with diameters of 5 cm, 10 cm, and 15 cm, since commercially available acrylic hemispheres are generally limited to sizes larger than 5 cm. The acrylic thickness is 3 mm. The front side of the liquid moderator container is designed as a hemisphere facing the irradiation field to maintain a symmetrical geometry, similar to that of a conventional Bonner sphere spectrometer. In contrast, the rear part of the container is cylindrical to facilitate liquid moderator injection and drainage. Although this design introduces asymmetry in the cylindrical section, it provides practical advantages for prototyping, as described in Chapter 5.

In this chapter, the acrylic liquid moderator container is not considered in PHITS simulations in order to reduce the computational time.

3.1.3 Neutron detection

LiCAF with dimensions of 0.3 mm $\times$ 0.3 mm $\times$ 0.3 mm, together with its electronic analysis system, is used. A detailed description will be provided in Chapter 5.

3.2 Neutron spectrometer selection study

The structure of CHARMS allows a maximum of 27 liquid moderator injection patterns. In conventional practice, Bonner sphere spectrometers with 6–7 different sizes and distinct response functions are typically used. [53] In this study, a systematic approach is proposed to select suitable liquid moderator injection patterns for accurate neutron energy spectrum evaluation. The following section summarizes previous evaluation methods that are applicable to the present research.

3.2.1 Energy resolution evaluation by using Backus-Gilbert approach

It is difficult to evaluate the energy resolution of a neutron spectrometer by visually judging response functions. To address this issue, Backus and Gilbert [28], [54] introduced mathematical procedure for quantifying resolving power in the context of geophysical inverse problems. In this study, their approach is applied to evaluate the energy resolution of the neutron spectrometer using only the response functions information. This method was used to evaluate energy resolution of neutron spectrometer set [19], [27] and long counter detector [55].

The most accurate way to evaluate the neutron flux at a reference energy E_0 would be to use a response function that behaves like a Dirac delta function at E_0 . However, this is not possible for an actual neutron spectrometer. The Backus–Gilbert approach overcomes this limitation by combining multiple response functions with different weights to form an averaging kernel, which has a peak that approximates a delta function at E_0 . The averaging kernel $A(E_0, E)$ at the reference energy E_0 is expressed as a linear combination of response functions:

$$A(E_0, E) = \sum_{i=1}^n a_i(E_0) R_i(E) \quad (3.1)$$

Figure 3.2 schematically shows Backus–Gilbert approach, in which multiple response functions are combined with weighting coefficients to form an averaging kernel centered at the reference energy E_0 .

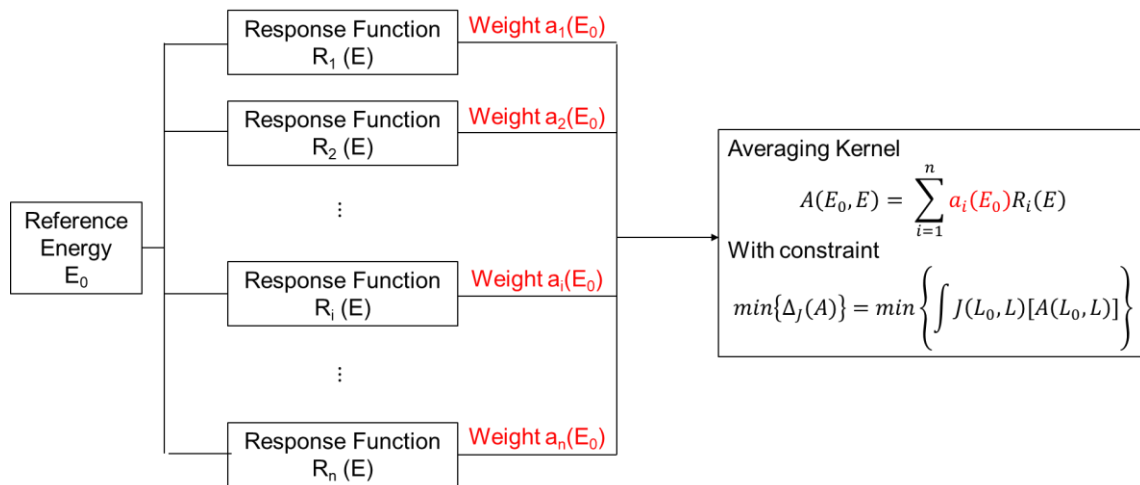


Figure 3.2: Energy resolution evaluation by Backus-Gilbert approach overview

Because the response function and neutron energy spectrum of a spectrometer are typically represented on a logarithmic energy scale, it is convenient to introduce a change of variables from energy E to lethargy E , defined as:

$$L = \log_{10} E \quad (3.2)$$

By expressing all quantities in terms of lethargy, the averaging kernel can be evaluated around a reference point L_0 .

To quantify how narrowly the kernel is concentrated around L_0 , an evaluating function is introduced as:

$$J(L_0, L) = (L_0 - L)^2 \quad (3.3)$$

This function is zero at $L = L_0$ and increases as the neutron energy deviates from this point.

Based on this function, a measure of the kernel's spread can be defined, providing a criterion for energy resolution as follows:

$$\Delta_J(A) = \int J(L_0, L)[A(L_0, L)]^2 \quad (3.4)$$

The coefficients $a_i(E_0)$ in equation (3.1) are determined by minimizing $\Delta_J(A)$ under a normalization constraint using the Lagrange multiplier method. This procedure yields a linear combination of response functions that provides the closest possible approximation to a delta function.

3.2.2 Rectangular spectrum evaluation method

For the rectangular spectrum evaluation method, once the response function and measurement data set based on an assumed synthetic rectangular neutron spectrum centered at the reference energy E_0 are prepared, the energy resolution can be evaluated through a spectral test, as shown in Figure 3.3(a).

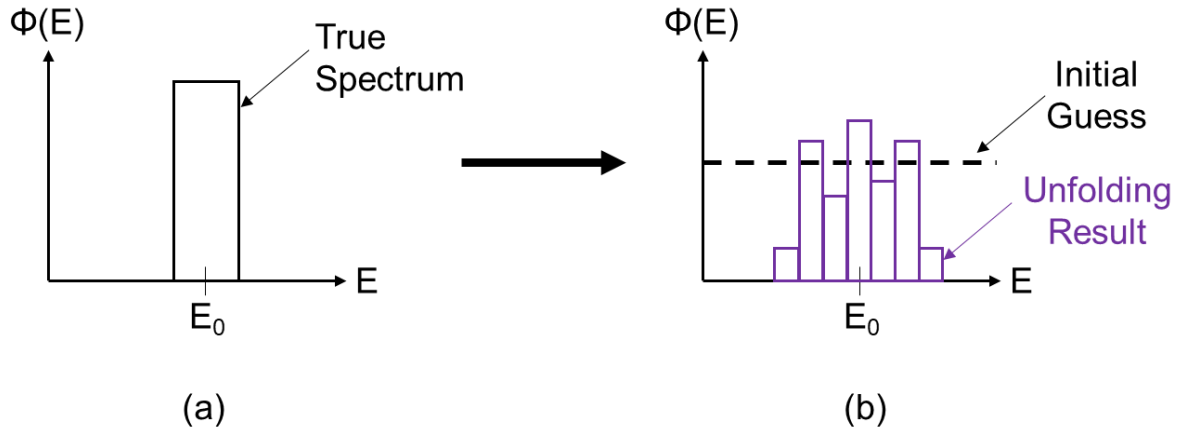


Figure 3.3: Rectangular spectrum evaluation method overview

The unfolding is performed using a flat, uniform initial guess spectrum, and the result example is shown in Figure 3.3(b). The similarity between the unfolded spectrum and the original rectangular spectrum is then analyzed. The closer match in spectral shape indicates better energy resolution at the reference energy E_0 .

This method was used to evaluate developed novel neutron spectrometers compared to conventional one. [19], [56]

3.2.3 Theoretical and unfolded result spectrum comparison method

This method evaluates how accurately a prepared neutron spectrometer can perform neutron unfolding when the measurement data set is generated by theoretical neutron energy spectrum.

To assess the difference between the derived and theoretical neutron spectra, an assessment function A is used.

$$A = \sqrt{\frac{1}{\log\left(\frac{E_n}{E_0}\right)} \sum_{j=1}^n \left[\int_{E_{j-1}}^{E_j} \left\{ \ln \left(\frac{\Phi_m(E)}{\Phi_T(E)} \right) \right\}^2 \cdot d(\log E) \right]} \quad (3.5)$$

Here, $\Phi_m(E)$ denotes the derived neutron spectrum, $\Phi_T(E)$ the theoretical neutron spectrum, and E_n and E_0 represent the maximum and minimum neutron energy values of the spectrum, respectively. A smaller value of A indicates better agreement between the two spectra.

This method was used to evaluate novel neutron spectrometer unfolding performance based on with [24], [25] and without [18] initial guess neutron spectrum information.

3.3 CHARMS liquid moderator injection patterns selection

3.3.1 Response function calculation

Figure 3.4 shows the geometry used for calculating the response functions. The response functions for the 27 different liquid moderator injection patterns were calculated using PHITS. In these calculations, monoenergetic neutron plane sources with different energies were employed as incident neutron beams.

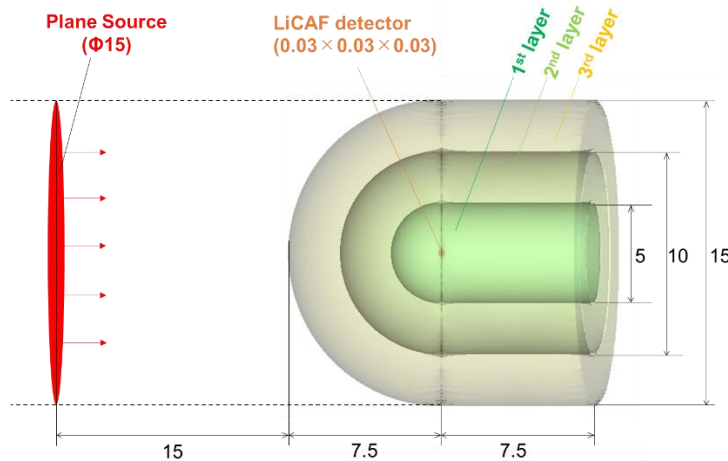


Figure 3.4: PHITS simulation geometry for response function calculation (unit: cm)

In Chapter 2, 145-group neutron energy structure was applied for the incident neutron irradiation. Ideally, the same 145-group structure would also be desirable in the present study. However, the use of such many neutron energy groups results in long computation times and introduces

difficulties in the unfolding process, since the number of energy groups becomes too large relative to the limited number of available CHARMS liquid moderator injection pattern.

Therefore, in this study, the neutron energy groups were combined into five-group intervals, resulting in a reduced 29-group structure for the calculation of the response functions.

3.3.2 Evaluation method that used for selection in this study

In Section 3.2, three possible evaluation methods for neutron spectrometer measurements were proposed: (i) energy resolution evaluation using the Backus–Gilbert approach, (ii) rectangular spectrum evaluation, and (iii) comparison between theoretical and unfolded spectra.

Among these three methods, Backus–Gilbert approach is the only one that relies only on the response functions, whereas the other two methods also depend on the unfolding procedure. Since different unfolding algorithms can lead to variations in the results, this study adopts the Backus–Gilbert energy resolution evaluation method.

To achieve high energy resolution across different neutron energy ranges in BNCT irradiation field, a two-pattern combination was selected for the thermal neutron region ($L_0 = -8, -7$), a three-pattern combination for the epithermal region ($L_0 = -6, -5, -4, -3, -2$), and a two-pattern combination for the fast neutron region ($L_0 = -1, 0$). The procedure for selecting the two-pattern combination in the thermal neutron region is as follows:

- (1) For all possible combinations (${}_{27}C_2$) of selecting two liquid moderator injection patterns out of the 27, by using energy resolution evaluation using the Backus–Gilbert approach Δ_J was calculated for $L_0 = -8, -7$. The total resolving power, $\Delta_{J,total}$ was defined as the sum of these values.
- (2) Since a smaller $\Delta_{J,total}$ corresponds to higher energy resolution, the combination with the minimum $\Delta_{J,total}$ was selected.

The same procedure was followed for epithermal and fast energy neutron region combination selection.

3.3.3 Liquid moderator injection selection result

Using the energy resolution evaluation based on the Backus–Gilbert approach, this section describes the results of selecting liquid moderator injection patterns for CHARMS. For clarity, the following notation is used to represent the liquid moderator injection patterns in CHARMS: “w” denotes water layer, “b” denotes boric acid layer, and “a” denotes empty layer (air). These notations are written in the order of the first, second, and third layers, as shown in Figure 3.4.

From the total of 27 liquid moderator injection patterns, the Backus–Gilbert energy resolution analysis selected the two-pattern combination “aba” and “waa” for the thermal neutron region. For the epithermal neutron region, the three-pattern combination “abb”, “wbw”, and “wwb” was identified as optimal, whereas for the fast neutron region, the two-pattern combination “abb” and “bbb” was selected.

Notably, the pattern “abb” was selected for both the epithermal and fast neutron regions, resulting in a total of only six liquid moderator injection patterns being required for neutron spectrum evaluation across all energy regions.

Figure 3.5 summarizes the optimized liquid moderator injection patterns for the different neutron energy regions.

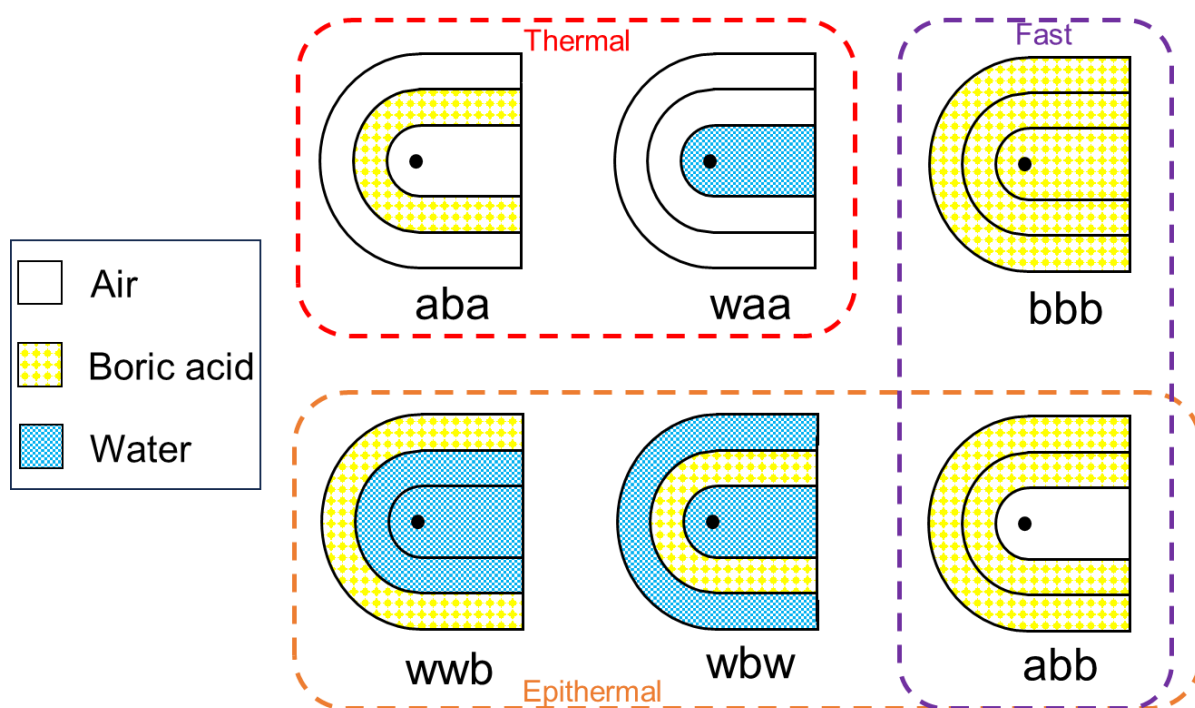


Figure 3.5: Optimized CHARMS liquid moderator injection pattern in each neutron energy region

The details of the optimization results and the characteristics of the selected injection patterns for each neutron energy region are described in the following subsections.

- Thermal neutron region

Table 3.1 shows the values of Δ_J and J at $L_0 = -8$, and $L_0 = -7$, together with the corresponding composite response function equations of “aba” and “waa”, which is selected for thermal neutron sensitive pattern in CHARMS.

Based on these results, the optimal combination that minimizes Δ_J was selected for each reference lethargy. The corresponding averaging kernels are shown in Figure 3.6(a), no distinct peak is formed at $L_0 = -8$ because none of the CHARMS response functions exhibit a sharp response in this energy region. As a result, the averaging kernel is broad, leading to a large Δ_J value and consequently poor energy resolution at this point. To improve sensitivity in this lethargy region, all-air configuration (“aaa”), which provides high sensitivity to the low thermal neutron energy region, is additionally incorporated in the CHARMS measurement pattern presented in Chapter 4 and Chapter 5.

In contrast, at $L_0 = -7$ (Figure 3.6(b)), a clear peak is observed at the reference lethargy. In this case, the averaging kernel is more strongly localized around $L_0 = -7$, resulting in a smaller Δ_J value and improved energy resolution compared with that at $L_0 = -8$.

Table 3.1: Values of Δ_J and J and composite response function equations in thermal neutron region

Lethargy	Δ_J	J	Composite response function equation
-8	0.96	-6.28	$-0.50R_{aba} + 1.37R_{waa}$
-7	0.74	-6.39	$-0.55R_{aba} + 1.40R_{aba}$

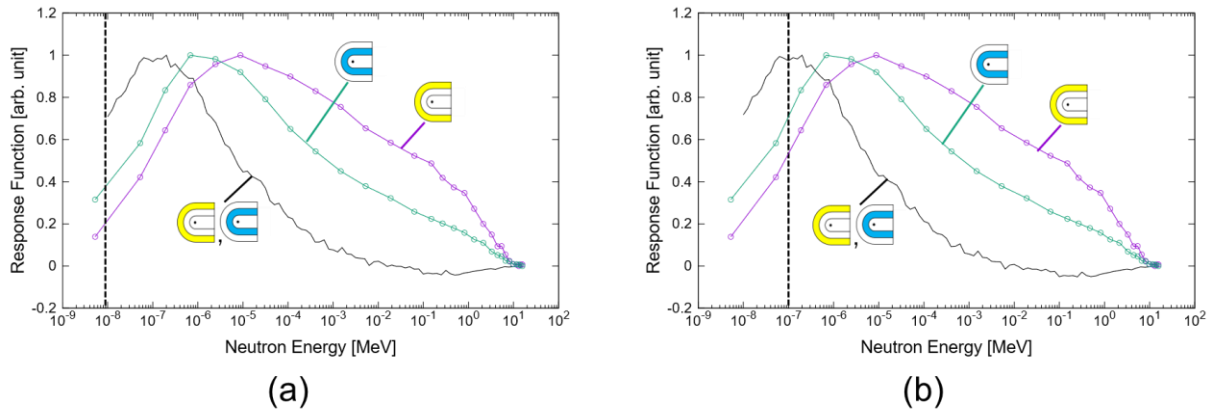


Figure 3.6: Averaging kernel results of best combination for thermal neutron region

(a) $L_0 = -8$, (b) $L_0 = -7$

- Epithermal neutron region

Table 3.2 shows the values of Δ_J and J at $L_0 = -6, -5, -4, -3, -2$, together with the corresponding composite response function equations of “abb”, “wbw” and “wwb”, which is selected for epithermal neutron sensitive pattern in CHARMS.

Table 3.2: Values of Δ_J and J and composite response function equations in epithermal neutron region

Lethargy	Δ_J	J	Composite response function equation
-6	1.37	-5.81	$1.56R_{abb}-1.61R_{wbw}+0.17R_{wwb}$
-5	1.53	-4.91	$1.60R_{abb}+1.15R_{wbw}-2.56R_{wwb}$
-4	1.61	-3.96	$1.19R_{abb}+2.53R_{wbw}-3.47R_{wwb}$
-3	1.75	-2.96	$1.40R_{abb}+1.22R_{wbw}-1.90R_{wwb}$
-2	1.60	-2.22	$1.05R_{abb}-1.15R_{wbw}+0.94R_{wwb}$

The corresponding averaging kernels are shown in Figure 3.7. At lower lethargy values ($L_0 = -6, -5$), the kernels exhibit narrower peaks and smaller Δ_J values, indicating better energy resolution. At higher lethargy values ($L_0 = -4, -3, -2$), the kernels become broader, resulting in larger Δ_J values and lower resolution.

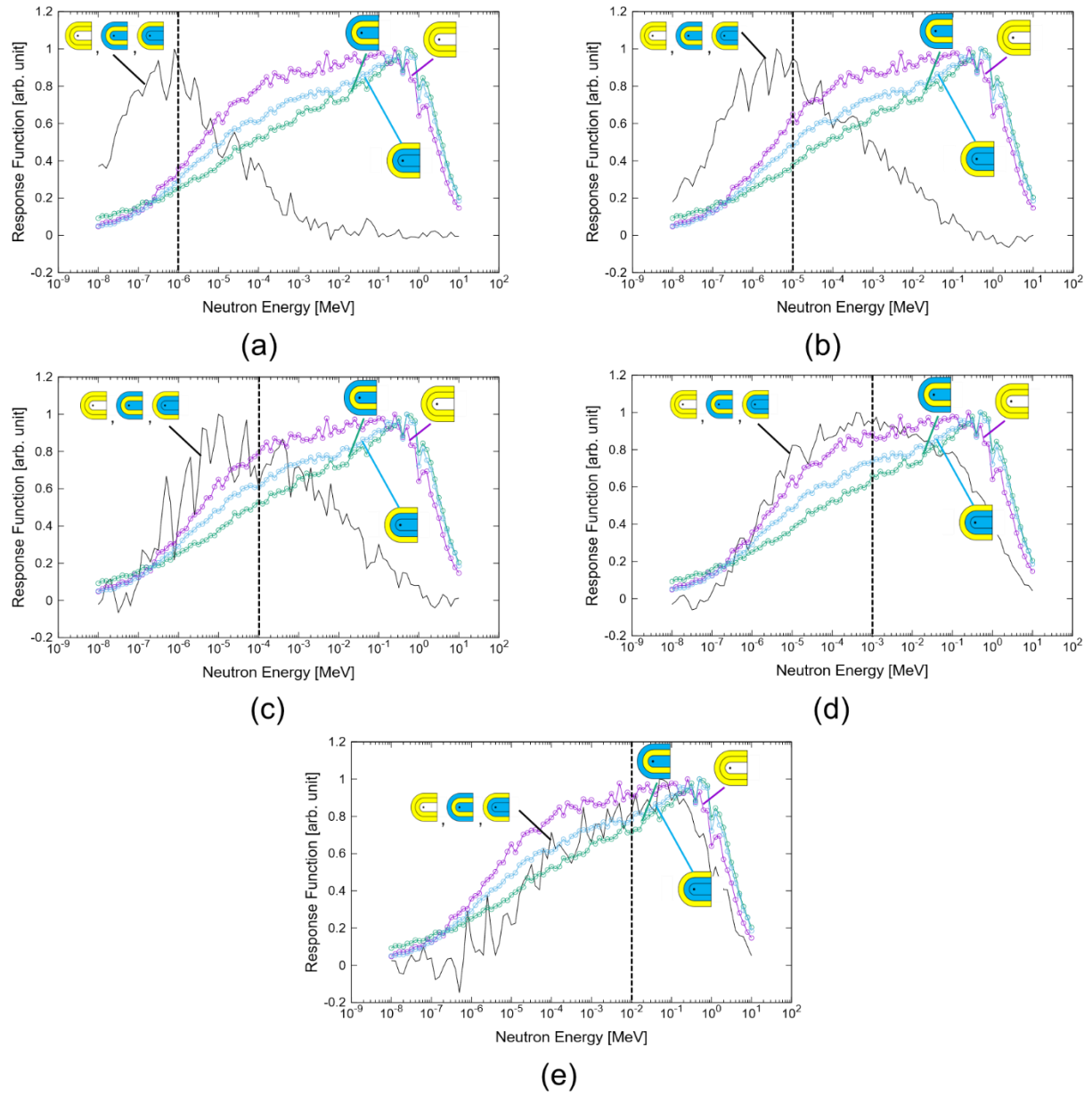


Figure 3.7: Averaging kernel results of best combination for epithermal neutron region

(a) $L_0 = -6$, (b) $L_0 = -5$, (c) $L_0 = -4$, (d) $L_0 = -3$, (e) $L_0 = -2$

- Fast neutron region

Table 3.2 shows the values of Δ_J and J at $L_0 = -1$, and $L_0 = -0$, together with the corresponding composite response function equations of “abb”, and “bbb”, which is selected for fast neutron sensitive pattern in CHARMS.

Table 3.3: Values of Δ_J and J and composite response function equations in fast neutron region

Lethargy	Δ_J	J	Composite response function equation
-1	1.26	-1.16	$-0.35R_{abb}+1.29R_{bbb}$
0	0.80	-0.88	$-0.45R_{abb}+1.38R_{bbb}$

In this region, the kernel peaks are located close to the reference lethargy values as shown in Figure 3.8, and the corresponding Δ_J values are relatively small compared with those in the thermal and epithermal regions, showing better resolution in the fast neutron energy range.

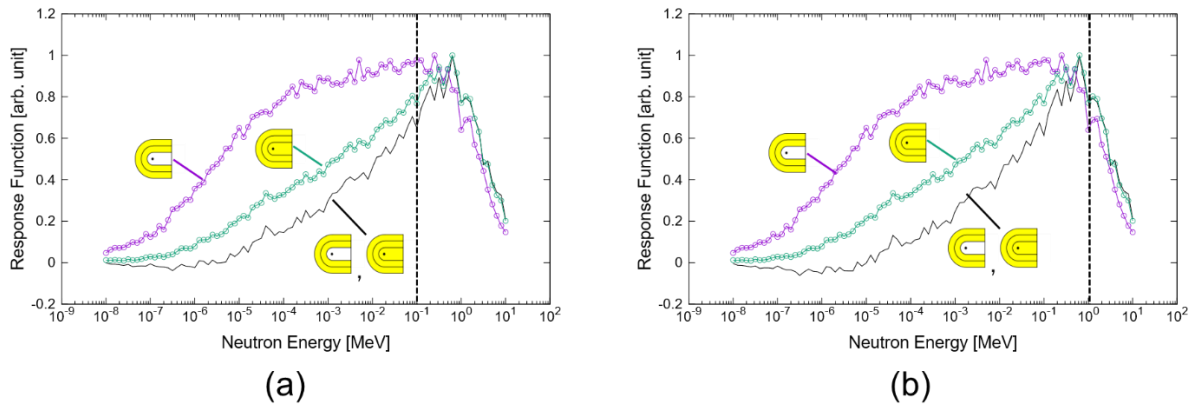


Figure 3.8: Averaging kernel results of best combination for thermal neutron region

(a) $L_0 = -1$, (b) $L_0 = 0$

Finally, Figure 3.9 summarizes the selected response functions, normalized to their respective peak values, for comparison across different energy regions. Table 3.4 summarizes the full width at half maximum (FWHM) values of the response functions for the novel combinations shown in Figure 3.9. The FWHM values were evaluated from the normalized response functions and are used as a quantitative indicator of energy resolution for comparing with conventional method.

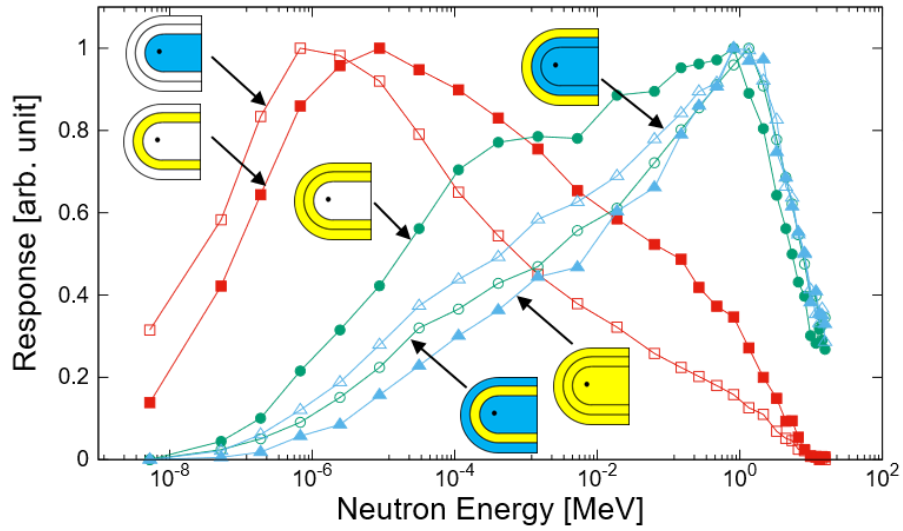


Figure 3.9: Optimized CHARMS liquid moderator injection pattern response function (peak normalization)

Table 3.4: FWHM values of response functions for optimized CHARMS liquid moderator injection pattern

Liquid moderator injection pattern	FWHM (Energy: MeV)
aba	6.34×10^2
waa	4.08×10^{-4}
abb	4.43×10^0
wbw	6.62×10^0
wwb	6.63×10^0
bbb	8.08×10^0

3.4 CHARMS evaluation test in BNCT irradiation field by simulation

The aim of this study is to compare the performance of CHARMS using a conventional method described in Subsection 3.4.1 with that of a novel method (Optimized CHARMS liquid moderator injection pattern) described in Section 3.3 for evaluating the neutron energy spectrum at KUR-HWNIF. The comparison included response function and its unfolding test result.

3.4.1 Conventional CHARMS liquid moderator injection method

An increase in the thickness of the pure water and boric acid liquid moderator was considered using a conventional Bonner spectrometer. Changing the injection pattern for each layer was considered in CHARMS.

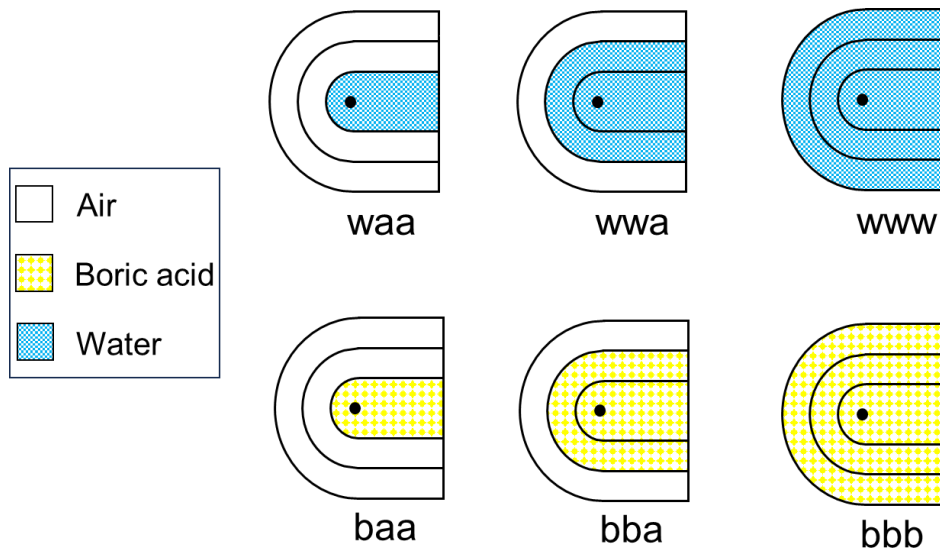


Figure 3.10: Conventional CHARMS liquid moderator injection pattern geometry

The selected patterns, “baa,” “bba,” “bbb,” “waa,” “wwa,” and “www,” as shown in Figure 3.10, were used for the spectrum unfolding at the KUR-HWNIF. The response function calculation result for conventional method is shown in Figure 3.11. Table 3.4 summarizes the full width at half maximum (FWHM) values of the response functions for the novel combinations shown in Figure 3.11.

A quantitative comparison of the FWHM values indicates that the novel combinations (Table 3.4) exhibit a smaller average FWHM than the conventional combinations (Table 3.5). In the epithermal neutron region, conventional combinations such as “wwa” and “bba” show relatively large FWHM values, indicating broadly distributed energy responses. In contrast, the novel combinations tend to suppress such broadening and provide more confined energy widths.

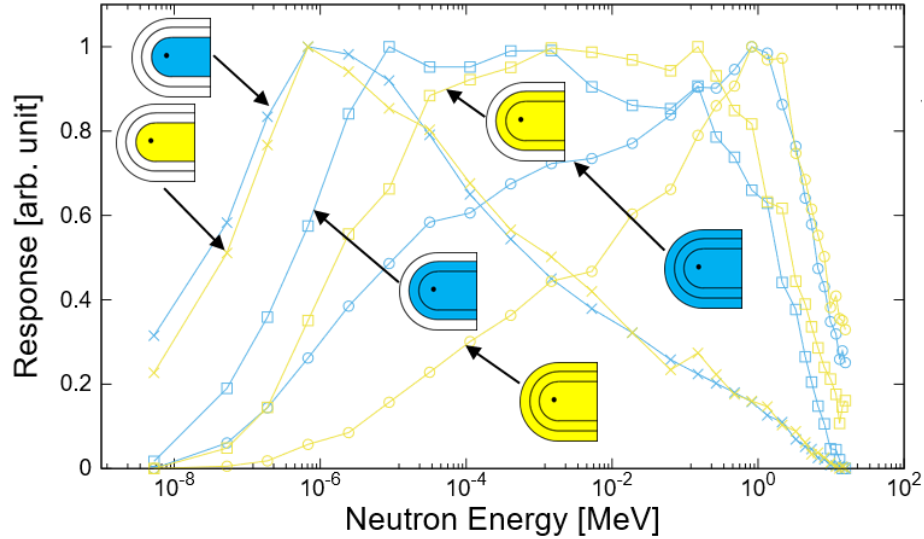


Figure 3.11: Conventional CHARMS liquid moderator injection pattern response function (peak normalization)

Table 3.5: FWHM values of response functions for conventional CHARMS liquid moderator injection pattern

Liquid moderator injection pattern	FWHM (Energy: MeV)
baa	1.47×10^{-3}
bba	2.14×10^0
bbb	8.08×10^0
waa	4.08×10^{-4}
wwa	1.34×10^0
www	5.42×10^0

3.4.2 KUR-HWNIF spectrum unfolding

In this study, the neutron energy spectrum of the KUR-HWNIF was used in the unfolding test. CHARMS with the selected combination for patterns using the novel method were placed on the beam center to calculate the reaction rate for ${}^6\text{Li}(n,\alpha){}^3\text{H}$. The distance between CHARMS surface and collimator surface was set to 4 cm. Figure 3.12 shows the calculation geometry for CHARMS. The annular neutron source radius decreased to 13.5 cm to save calculation time.

Using the reaction rate calculation results, an unfolding test was performed using the MAXED code in the UMG package [57]. Using the MAXED code, the initial guess of the solution must be provided by the user. First, the neutron energy spectrum at LiCAF position (true spectrum) was used as one of initial guess spectrum.

As shown in Figure 3.13, the true spectrum is divided into four regions: thermal neutrons, low-energy epithermal neutrons, high-energy epithermal neutrons, and fast neutrons. For each region, initial guess spectra were created by increasing the neutron flux by 50%. These different modified

spectra were also used as initial guess spectra for the unfolding test to evaluate the spectrum correction ability.

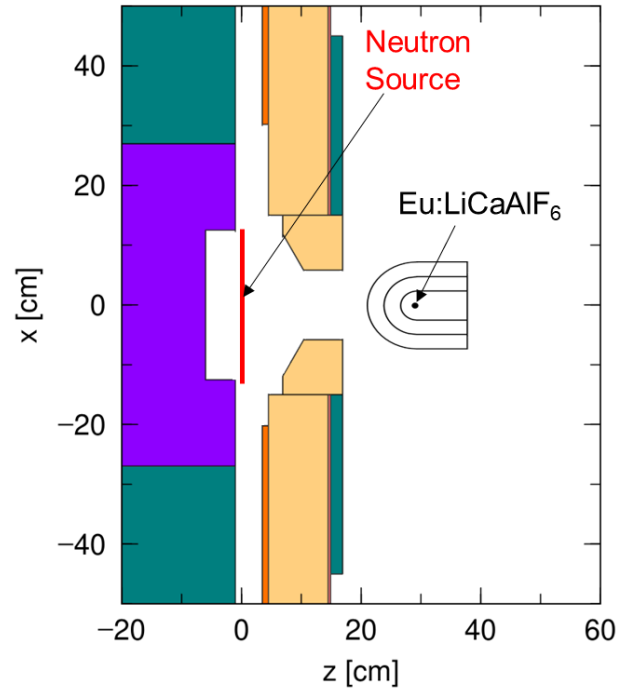


Figure 3.12: Calculation geometry for CHARMS

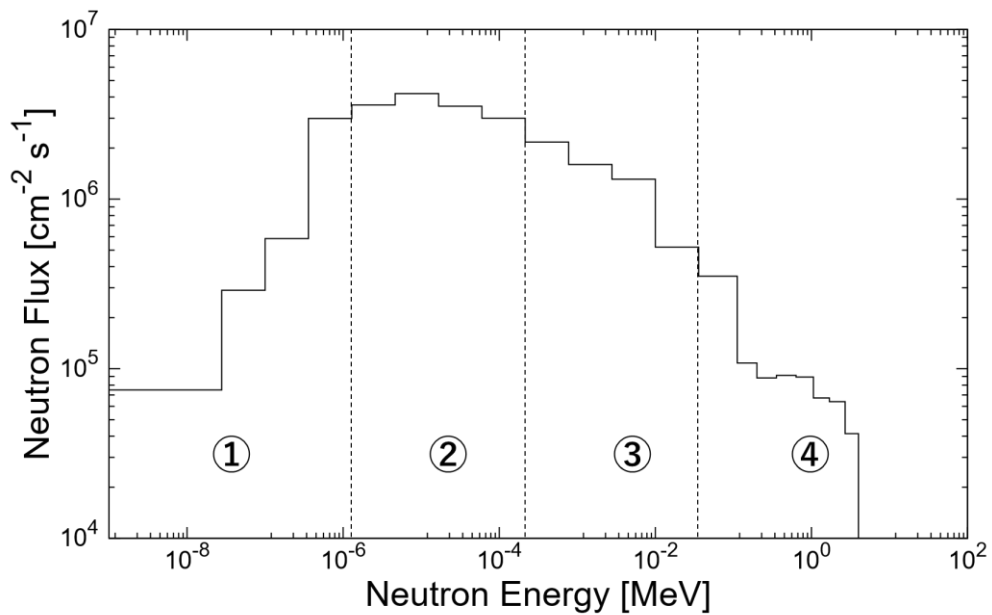


Figure 3.13: Neutron energy spectrum at LiCAF position

- ① Thermal: below 1.29×10^{-6} MeV, ② Low-energy epithermal: 1.29×10^{-6} – 2.15×10^{-4} MeV,
 ③ High-energy epithermal: 2.15×10^{-4} – 3.59×10^{-2} MeV, ④ Fast: over 3.59×10^{-2} MeV

3.4.3 Response function comparison between novel and conventional method

Figure 3.9 and Figure 3.11 shows the response functions of selected liquid moderator injection patterns using both the novel and conventional methods with normalization at the peak value respectively. A comparison between the two methods shows that the proposed method does not have a flat part in any response function for all injection patterns. Additionally, the novel method exhibited varying slope increments compared with the conventional method.

3.4.4 KUR-HWNIF spectrum unfolding result between novel and conventional method

Using the MAXED code, the true spectrum was unfolded using the novel and conventional methods. Figure 3.14 shows the unfolding results of the true KUR-HWNIF spectrum. The unfolding error is defined as the error between the unfolded spectrum and the true spectrum. From Figure 3.15, it can be observed that the novel method can obtain a lower unfolding error than the conventional method, particularly in the high epithermal and fast neutron regions.

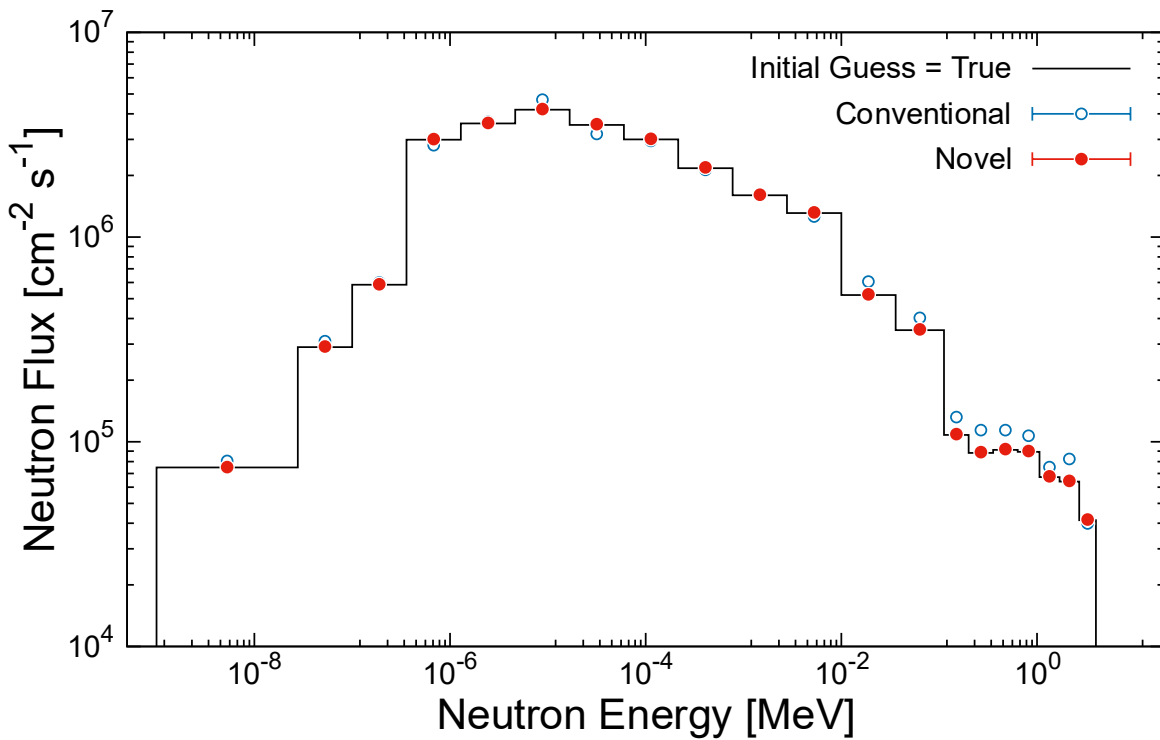


Figure 3.14: Unfolding results for KUR-HWNIF true spectrum by using novel and conventional method

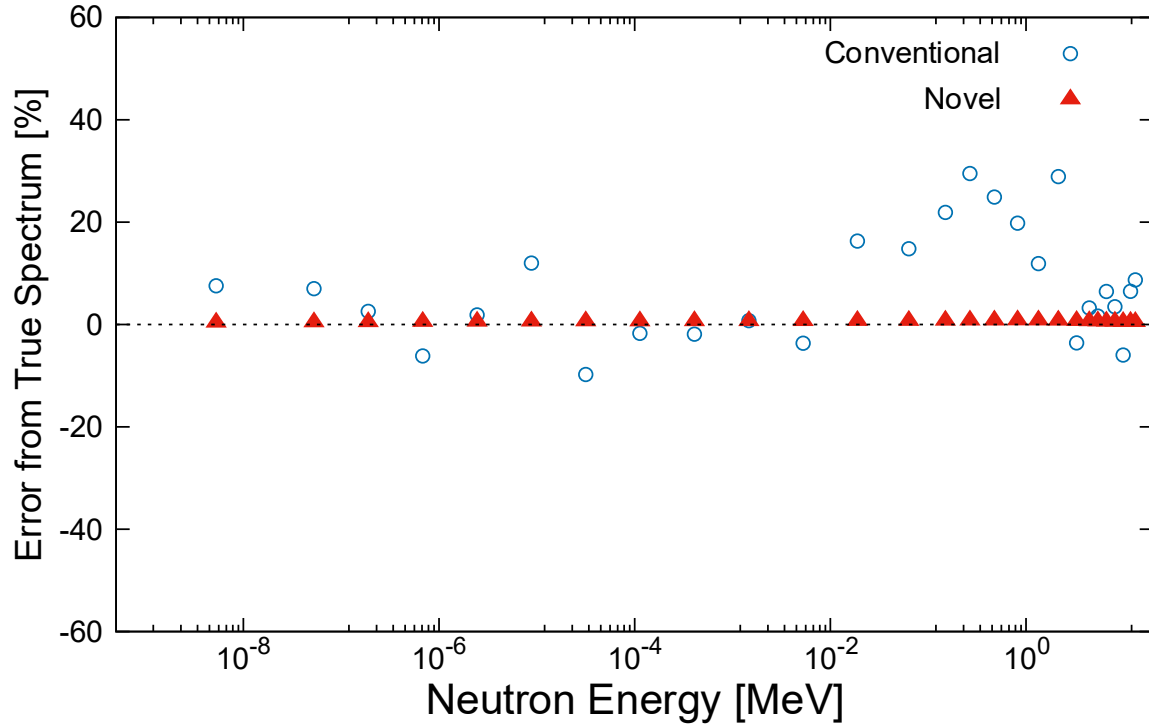


Figure 3.15: Unfolding error results for KUR-HWNIF true spectrum by using novel and conventional method

Figure 3.16—Figure 3.22 show the unfolding results and their error from true spectrum obtained using different modified spectra as initial guesses. Both the novel and conventional methods show that the increased neutron flux part of the initial guess approaches the true spectrum, indicating that some correction has occurred. However, an error was created in the unchanged neutron flux part of the initial guess, which indicated that proper unfolding had been achieved.

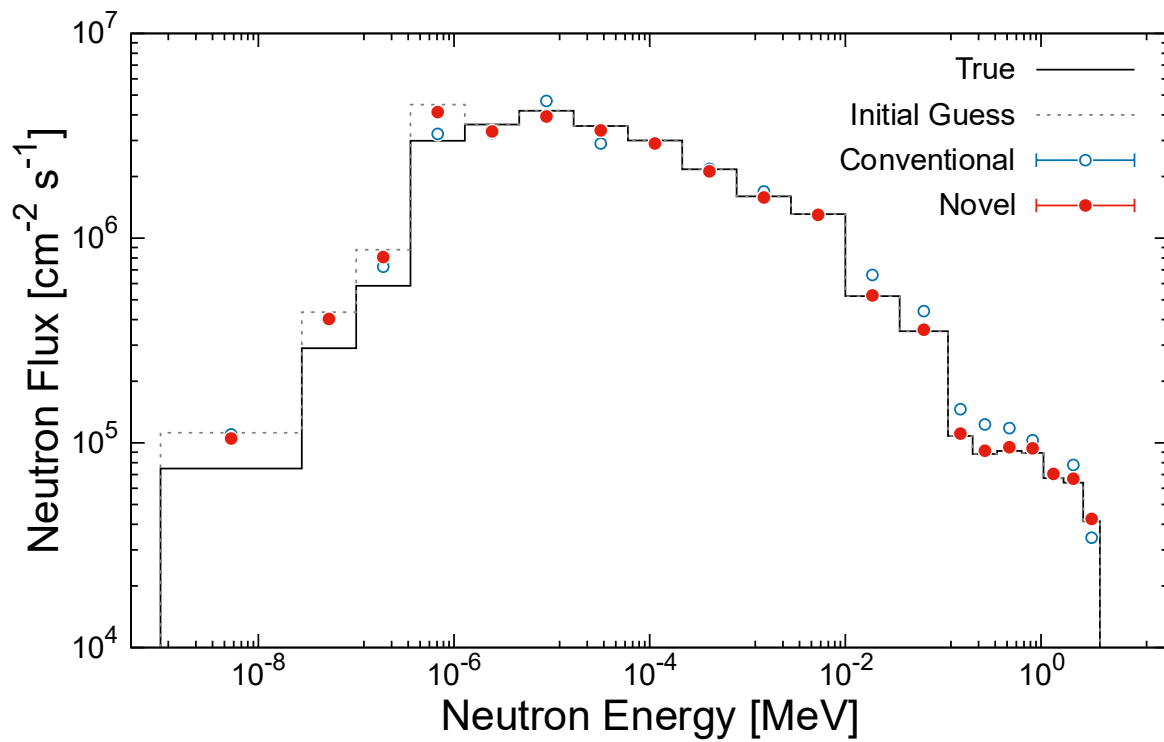


Figure 3.16: Unfolding result for the initial guess for the increase of thermal neutron flux by 50 %

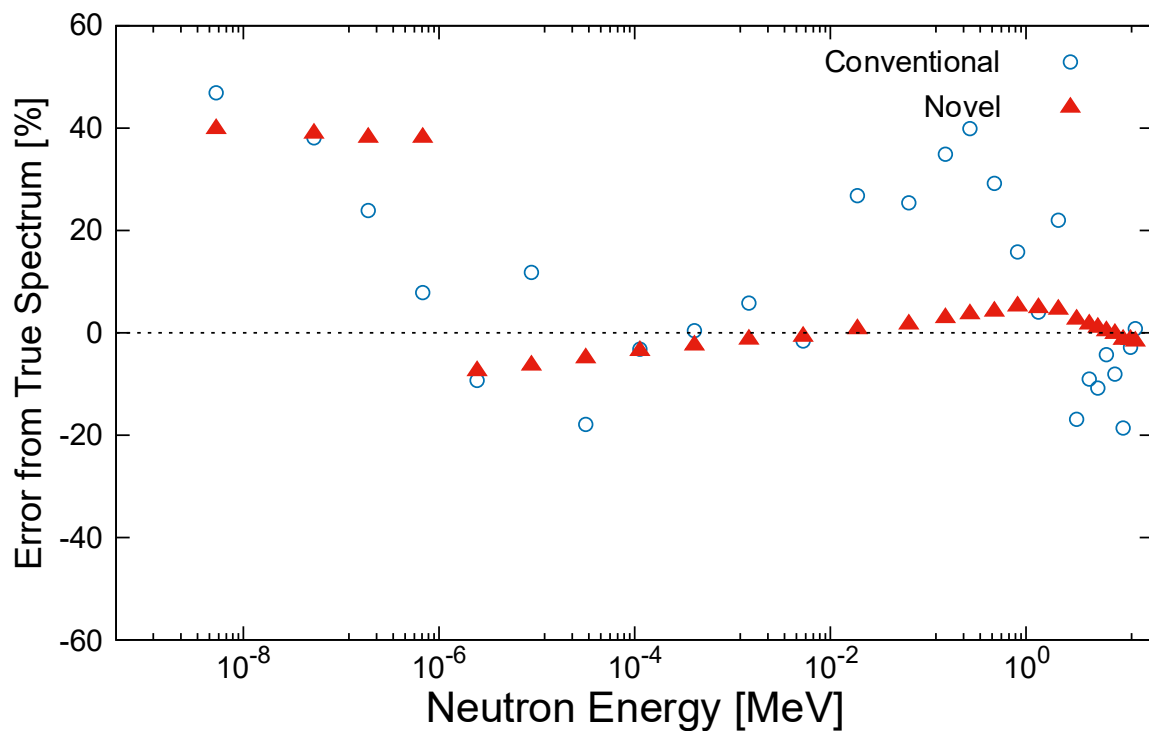


Figure 3.17: Unfolding error result for the initial guess for the increase of thermal neutron flux by 50 %

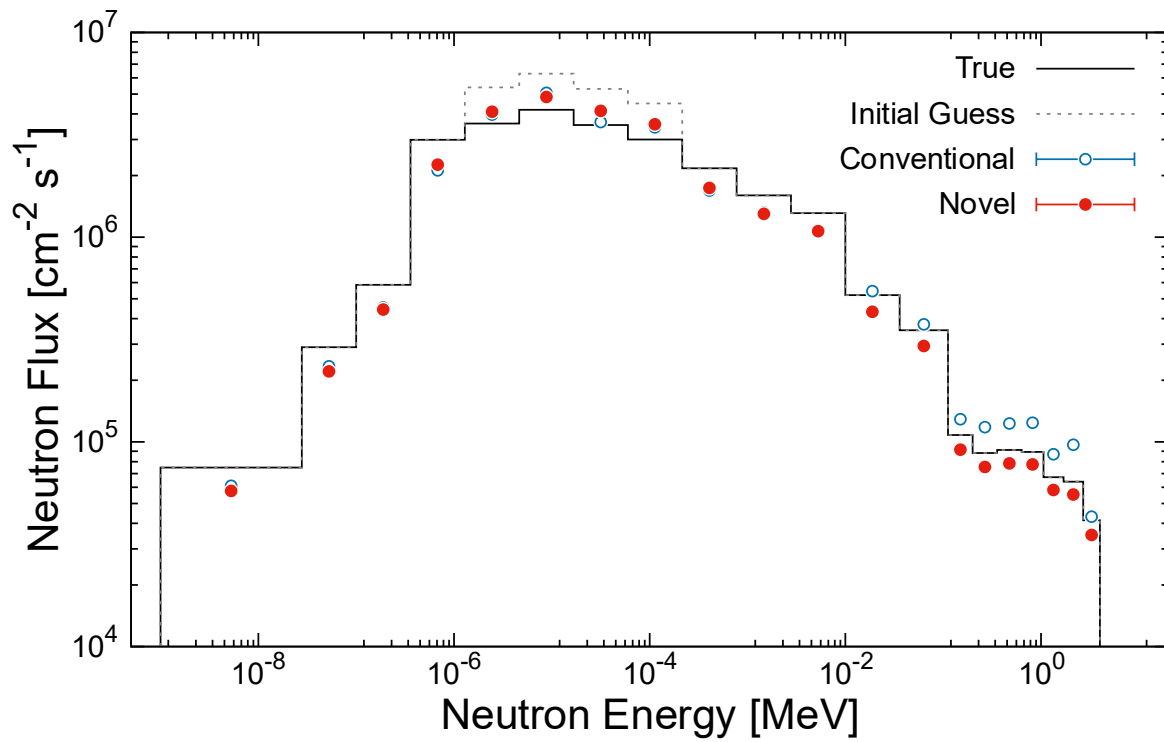


Figure 3.18: Unfolding result for the initial guess for the increase of low energy epithermal neutron flux by 50 %

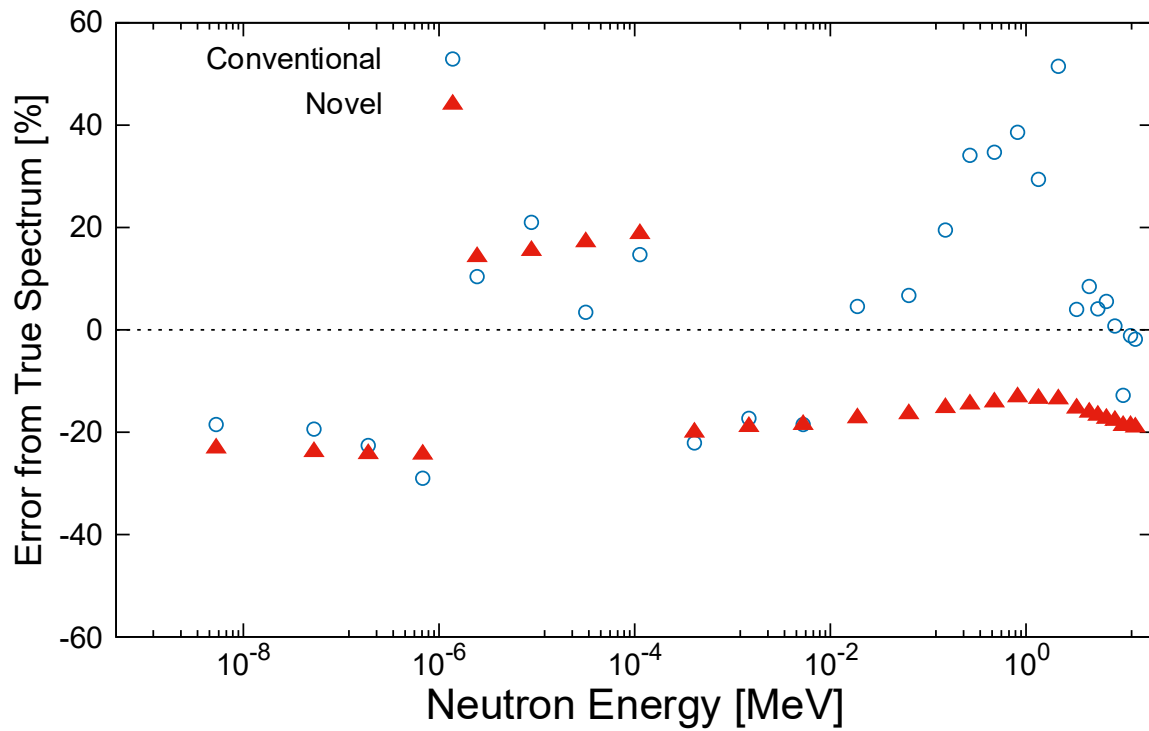


Figure 3.19: Unfolding error result for the initial guess for the increase of low energy epithermal neutron flux by 50 %

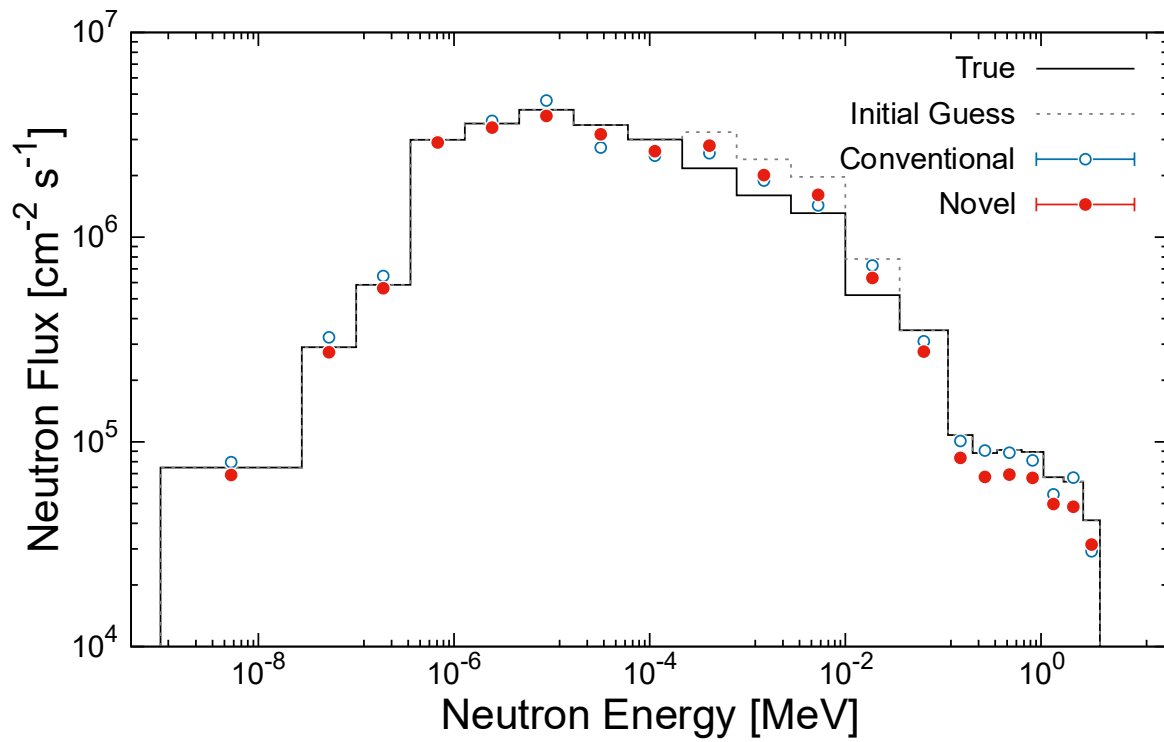


Figure 3.20: Unfolding result for the initial guess for the increase of high energy epithermal neutron flux by 50 %

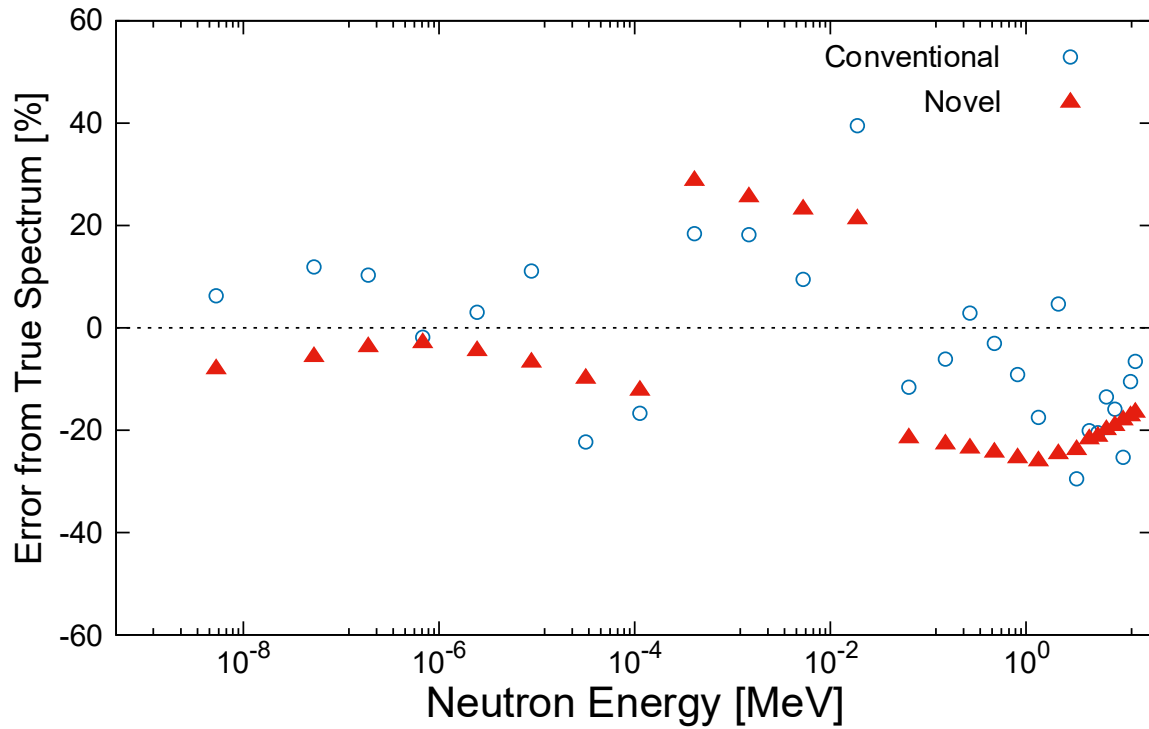


Figure 3.21: Unfolding error result for the initial guess for the increase of high energy epithermal neutron flux by 50 %

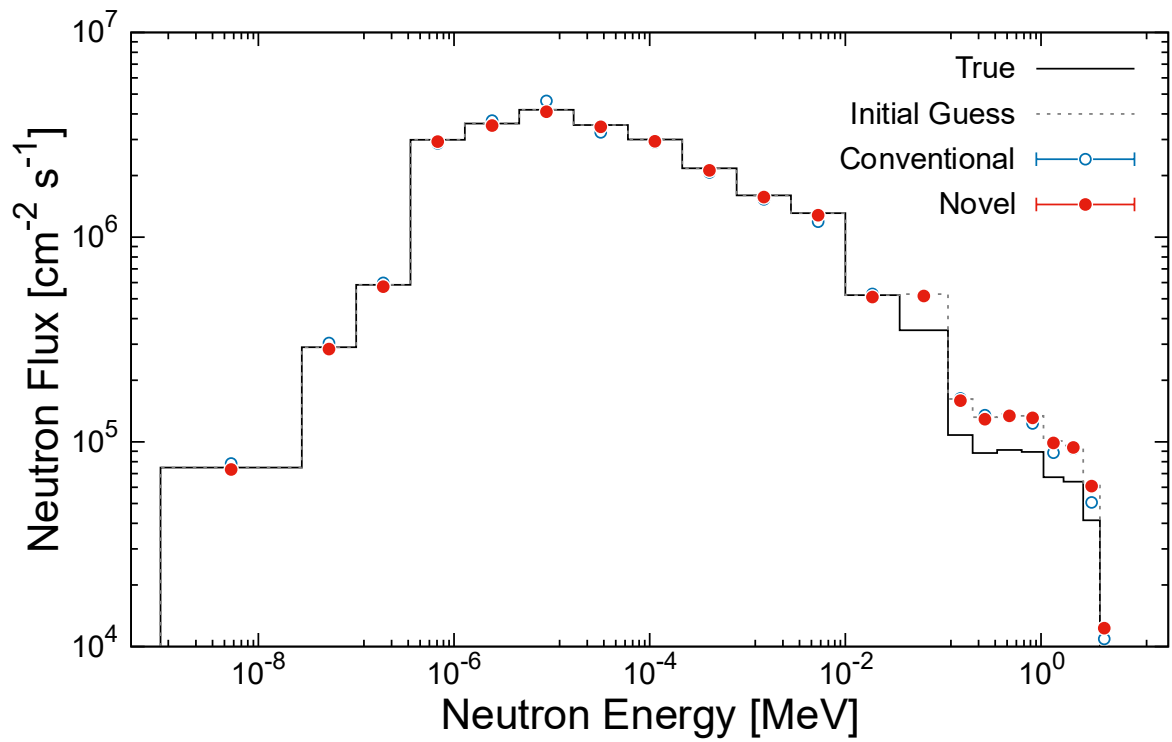


Figure 3.22: Unfolding result for the initial guess for the increase of fast neutron flux by 50 %

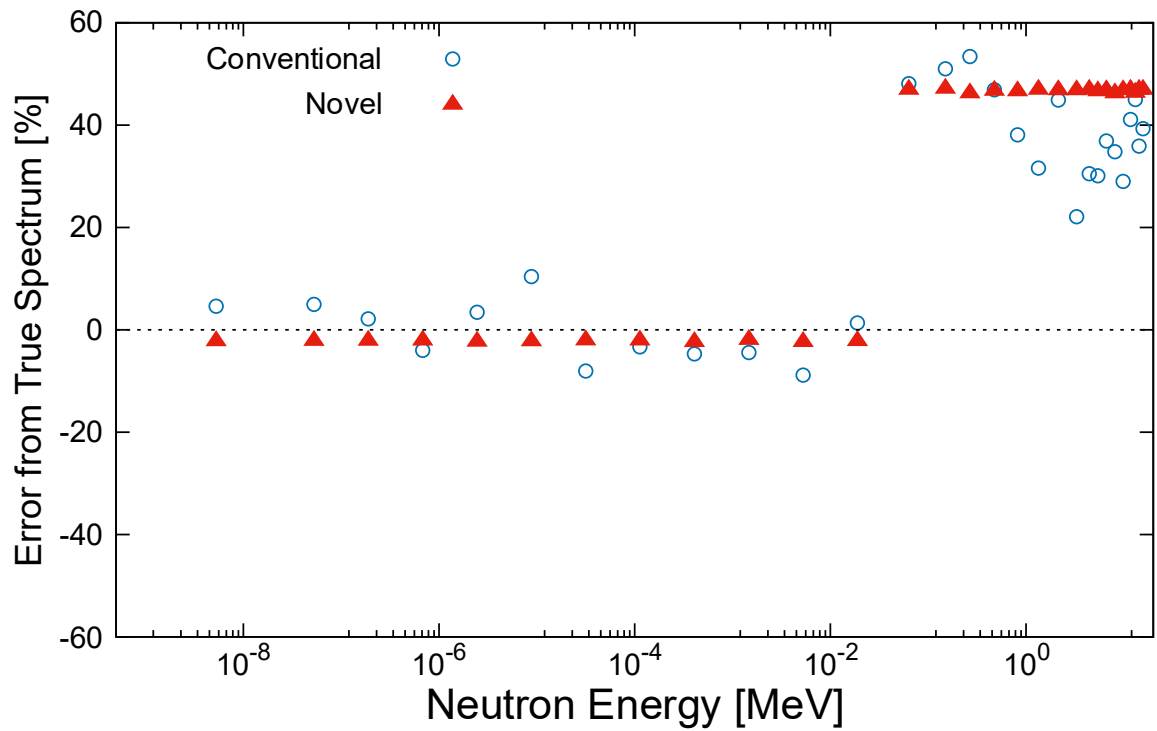


Figure 3.23: Unfolding error result for the initial guess for the increase of fast neutron flux by 50 %

In Figure 3.16, with 50% increase in neutron flux in the thermal neutron region from the initial guess, the conventional method exhibits a wide range of errors, ranging from 17%—40%. In contrast, the novel method showed a much narrower error range, ranging from 38%—39%. These results were also observed when the neutron flux was increased by 50 % in the low-energy epithermal neutron, high-energy epithermal neutron, and fast neutron regions, as shown in Figure 3.18—Figure 3.23. Therefore, the novel method proved to be more stable than the conventional method in correcting regions with incorrect neutron energy spectra.

For the unchanged neutron flux part, it was observed that neutron flux oscillations occurred. The conventional method exhibited more intense oscillations than the novel method.

3.5 Conclusion

Using CHARMS, the higher-energy-resolution response function with the novel method than with the conventional method was obtained. The novel method provided more stable and accurate unfolding results for different types of initial guess spectra compared to the conventional method. CHARMS with a novel method such as the approach of Backus-Gilbert for selecting a combination of liquid moderators is promising for evaluating the neutron energy spectrum in the BNCT irradiation field.

Chapter 4 Validation test of liquid multilayer neutron spectrometer

In Chapter 3, the optimization study of CHARMS in BNCT irradiation field was conducted using simulation calculations. The final goal of CHARMS is to be applied to BNCT QA with a remote-operating system. However, before realizing the full-scale CHARMS system with remote operation, it is necessary to confirm and verify the feasibility of multilayer moderator spectrometer under BNCT irradiation field.

To achieve this, Simple Multilayer Spectrometer (SMS) was designed, constructed, and experimentally tested. Unlike CHARMS, which requires a liquid supply and drainage system for remote-operating system, the SMS was developed as a passive and compact device that retains the essential concept of CHARMS. This design enabled experimental validation of multilayer spectrometer concept in BNCT irradiation field.

4.1 Simple Multilayer Spectrometer (SMS)

4.1.1 Container

In Chapter 3, CHARMS container is the combination of a cylindrical body coupled with a hemispherical front surface. The hemispherical section provided high symmetry and minimized directional dependence when exposed to an irradiation field. However, several research groups have reported neutron spectrometers using simpler, non-symmetric geometries such as cylinders [58] and cuboids [26]. These shapes have not yet been fully verified in BNCT irradiation fields, but they offer the potential for easier fabrication and handling.

In particular, the cuboid configuration presents two notable benefits. First, its structural simplicity eliminates the need for additional fixtures that might otherwise increase measurement uncertainty during irradiation [19]. Second, in comparison with a sphere of similar size, the cuboid provides extra moderator volume due to its corner regions, which can increase response at higher neutron energies [59].

Based on these considerations, Simple Multilayer Spectrometer (SMS) was designed as a cuboidal assembly (Figure 4.1). The SMS consists of three acrylic moderator containers, labeled A1, A2, and A3, with outer dimensions of 5 cm × 5 cm × 15 cm, 10 cm × 10 cm × 15 cm, and 15 cm × 15 cm × 15 cm, respectively. The acrylic walls are 3 mm thick, and each container is sealed with a lid combining an expansion structure to hold an activation foil at the central position. The containers can be assembled in sequence, and the inter-container gaps are filled with liquid moderators. In this work, pure water and boric acid were employed, and in some cases empty gaps were left to act as air layers.

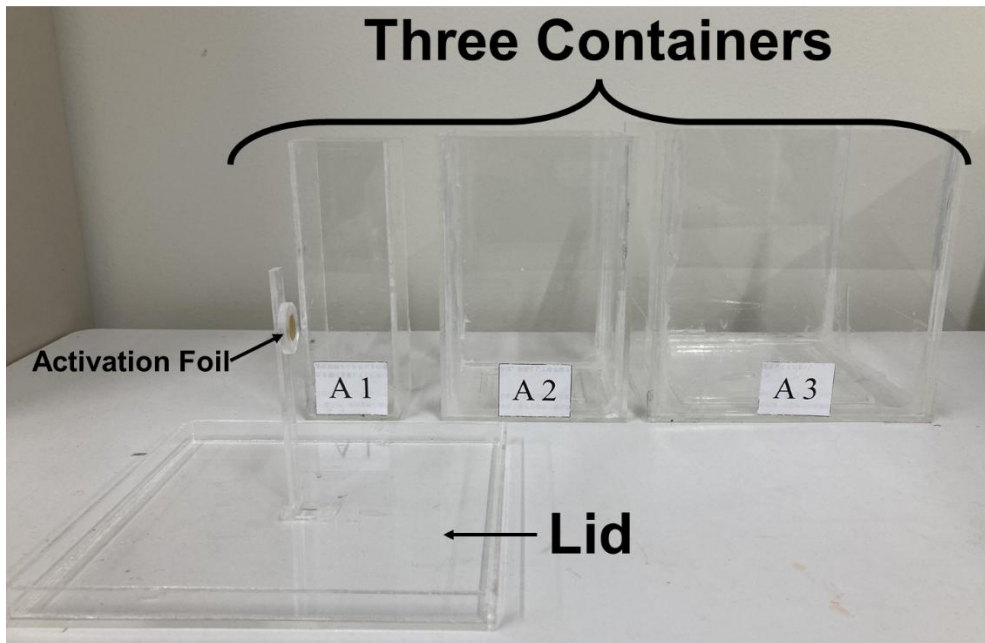


Figure 4.1: Simple multilayer spectrometer (SMS) components

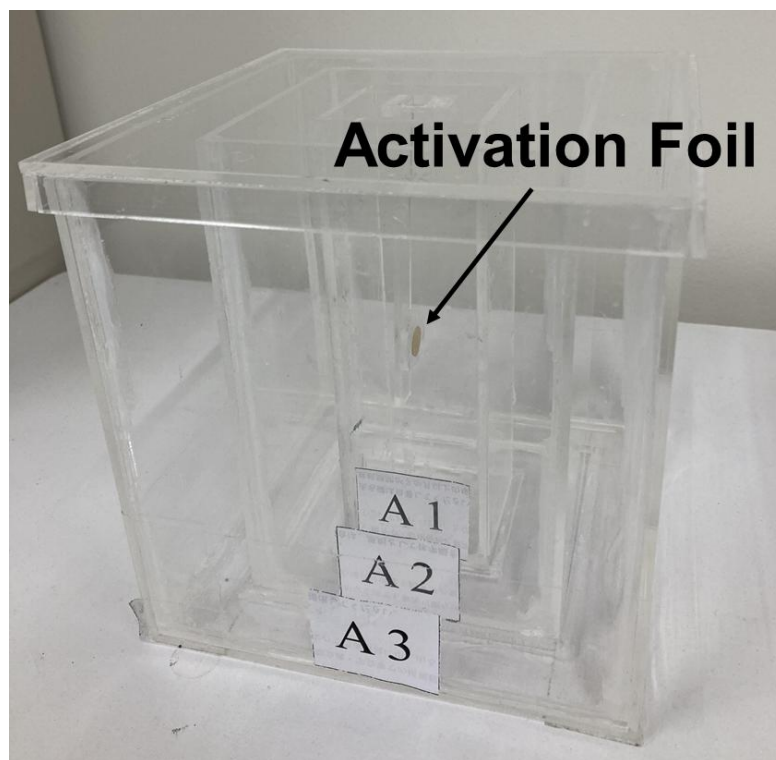


Figure 4.2: Assembly of SMS components

4.1.2 Neutron detection

The original CHARMS design incorporated an online neutron detection system using LiCAF detector, together with a liquid supply and drainage system to enable remote operation. In the present study, these components were intentionally omitted in SMS in order to facilitate a fundamental verification experiment.

Instead of online detection, thermal neutron reaction rates were evaluated using activation foils, which are widely recognized as reliable neutron detectors exhibiting an approximately $1/v$ response in the thermal neutron energy region. Thin circular gold foils with a diameter of 1 cm and a thickness of 0.05 mm were employed in this study.

To separate the thermal and epithermal neutron contributions, cadmium-covered gold foils were additionally prepared. Each cadmium cover had a thickness of 0.5 mm and a diameter of 1.2 cm, slightly larger than that of the gold foil, ensuring complete shielding of the foil edges. For each moderator injection pattern, SMS was irradiated twice: once with a bare gold foil and once with a cadmium-covered gold foil placed at the central position of the moderator.

Gold foils were activated via the $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ reaction, and the reaction rate was determined by measuring the 411 keV gamma ray emitted from ^{198}Au . This reaction exhibits a large cross section in the thermal neutron region, while also possessing resonance structures in the epithermal neutron region. Since BNCT irradiation fields contain a substantial fraction of epithermal neutrons, these resonance contributions can introduce significant uncertainty in the direct evaluation of the reaction rate using bare gold foils alone. To mitigate this effect, the thermal neutron reaction rate was evaluated using the cadmium difference method. By subtracting the reaction rate measured with a cadmium-covered gold foil from that measured with a bare gold foil, the contribution from neutrons with energies below the cadmium cutoff energy (approximately 0.1 eV) can be estimated. The cadmium ratio is defined as

$$RR_{Cd} = \frac{R_{bare}}{R_{Cd}} \quad (4.1)$$

where R_{bare} is the reaction rate measured with a bare gold foil, which responds to neutrons over the entire energy range, and R_{Cd} is the reaction rate measured with a cadmium-covered gold foil, which mainly responds to epithermal and fast neutrons. Note that, The reaction rate R was calculated according to Equation (E.1).

Based on the cadmium ratio, the reaction rate per unit mass due to thermal neutrons below 0.1 eV, R_{th} , can be expressed as

$$R_{th} = \frac{R_{bare}(1 - 1/RR_{Cd})}{F_{Cd}} \quad (4.2)$$

where F_{Cd} is the self-shielding correction factor determined by the thickness of the gold foil. In this study, a gold foil thickness of 50 μm was used, resulting in an F_{Cd} value of 0.95.

4.2 Simulation Calculation

4.2.1 Optimized SMS liquid injection patterns

Six liquid moderator injection patterns (“aba,” “abb,” “bbb,” “waa,” “wbw,” and “wwb”) were selected as shown in Figure 4.3. Additionally, the “aaa” pattern was included because it provides improved resolution in the low energy thermal neutron region. The “aaa” pattern corresponds to the empty layer condition, in which neutrons can directly interact with the gold foil without passing through any liquid moderator.

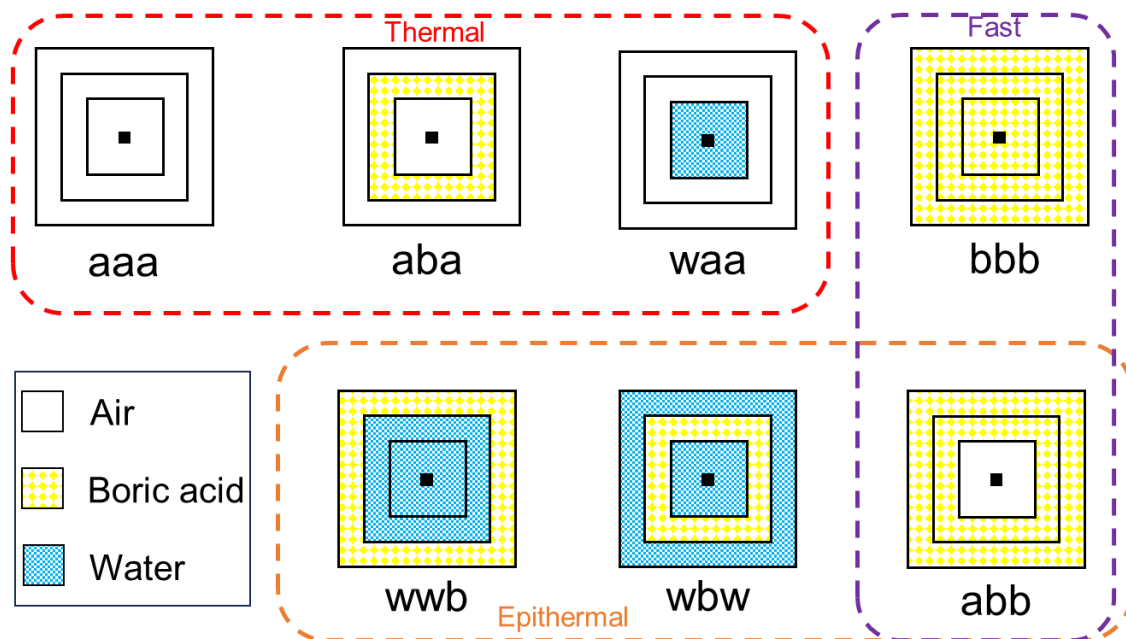


Figure 4.3: Optimized SMS liquid moderator injection patterns in each neutron energy region

4.2.2 Calculation geometry

Figure 4.4 shows the SMS response function calculation geometry used in PHITS. In this calculation, the acrylic cuboid containers were included. To obtain the initial guess spectrum, geometry without the SMS structure was applied, and neutron flux tallies were performed at the position of the gold foil.

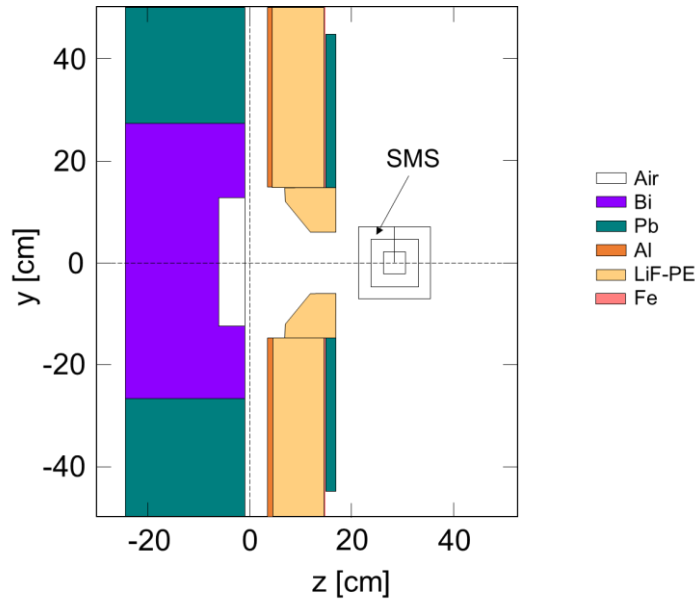


Figure 4.4: SMS calculation geometry

Figure 4.5 shows irradiation set-up of SMS in three dimensions. The dimension of the SMS is designed based on the structure of our previous study, CHARMS. CHARMS comprises 3 layers, with the outermost layer having a diameter of 15 cm. To ensure that the response function of the SMS closely matches that of CHARMS, a dimension of 15 cm $\times$ 15 cm $\times$ 15 cm for the SMS was selected. This dimensional consistency helps ensure alignment and comparability between the two systems. On the other hand, the collimator size of KUR- HWNIF is 12 cm, which is typical collimator size for BNCT. The collimator diameter was smaller than SMS height, so SMS were placed 4 cm apart for the surface of collimator in order to reduce the effects of the collimator edge. Because the neutron beam is not perfectly parallel, the neutrons spread out depending on their distance from the collimator surface.

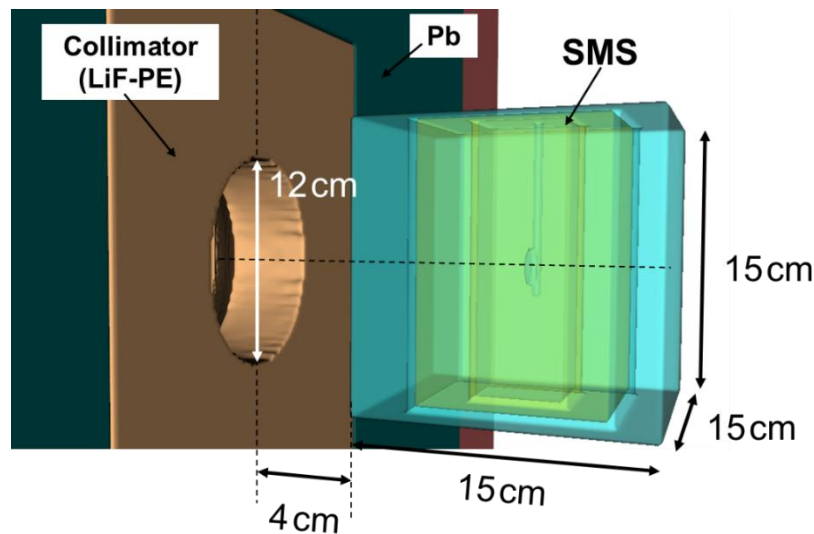


Figure 4.5: Three-Dimensional Irradiation condition of SMS at KUR-HWNIF

4.2.3 Response function

The response functions of the seven selected liquid injection patterns were calculated by PHITS. In these calculations, the incident neutron beam was modeled as monoenergetic neutrons with energies between 10^{-9} eV and 31.6 MeV. The energy range was divided into 42 logarithmically spaced bins, and for each bin the transport and interaction of neutrons through the SMS geometry were evaluated.

A different energy grouping from that used in the CHARMS optimization study was adopted. In the optimization process, using overly fine energy intervals in the fast neutron region led to instability in the unfolding results. Therefore, a wider bin width was applied in the fast energy region, consistent with that of the other regions, to ensure stable unfolding results.

The thermal neutron reaction rate of a gold foil located at the neutron spectrometer center was obtained from the difference in responses between bare and cadmium-covered foils. This approach enabled the isolation of the <0.1 eV neutron contribution, ensuring that only the thermal component was considered in the response function. To satisfy the requirements of the SMS neutron detection system, the tally focused on the $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ saturated activity for neutrons below 0.1 eV.

Figure 4.6 shows the comparison of the normalized response functions with normalization at the peak value results of the SMS and CHARMS. In all liquid injection patterns, the shapes of the response functions of SMS are similar to those of CHARMS. Therefore, neutron spectrometer verification of CHAMRS can be confirmed using the SMS.

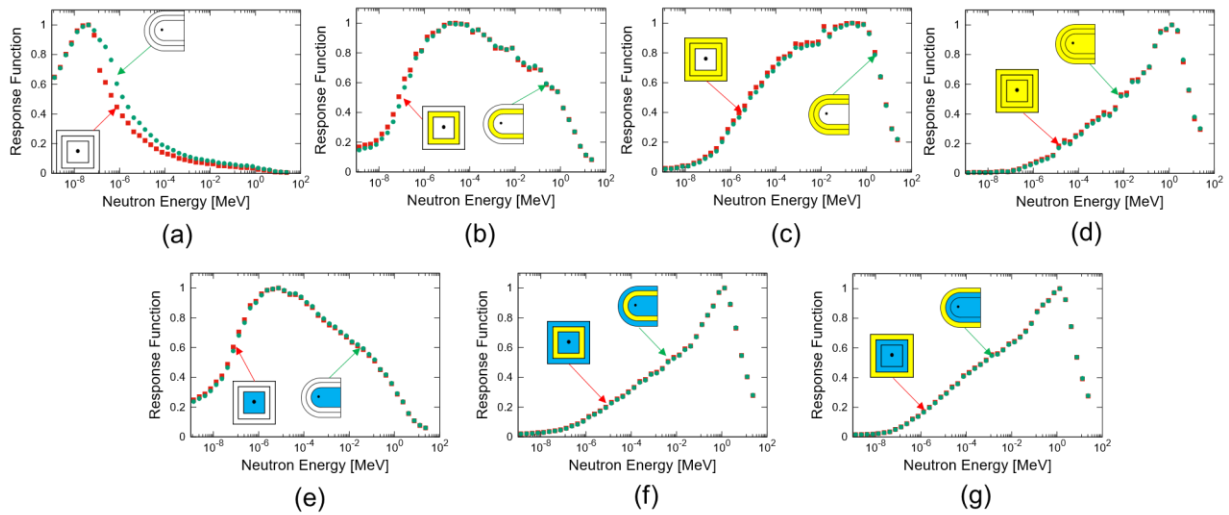


Figure 4.6: Response function of SMS compared with CHARMS

(a)“aaa”, (b)“aba”, (c) “abb”, (d)“bbb”, (e)“waa”, (f)“wbw”, (g)“wwb”

4.3 SMS irradiation experiment

Figure 4.7 shows the experimental setup for SMS measurements at the KUR-HWNIF. Each liquid moderator injection pattern was irradiated separately for the bare and cadmium-covered gold foils. The irradiation time for measuring the neutron spectra at a distance of 11.5 cm from the collimator surface was 15 min. The 412 keV gamma rays emitted from the activated gold foil were measured using HPGe detector.

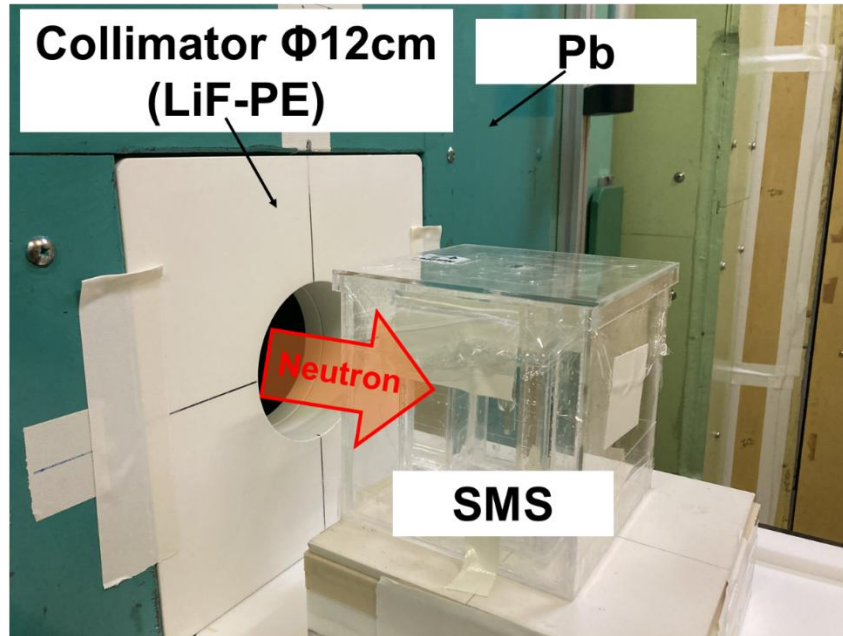


Figure 4.7: SMS irradiation experiment setup

4.4 Results and discussion

4.4.1 HPGe detector measurement

Table 4.1 and Table 4.2, show the measured reaction rate of gold foils for the bare and cadmium-covered cases, respectively. From these values, the thermal neutron reaction rate of gold foils for each liquid moderator injection pattern was calculated, and the results are summarized in Table 4.3. The reaction rates were in the range of 10^5 — 10^6 s⁻¹g⁻¹.

Patterns containing a larger fraction of boric acid, such as “abb” and “bbb,” showed lower thermal neutron reaction rates compared with patterns filled with only water or with empty layers. This reduction is due to the strong absorption of thermal neutrons by boron through the $^{10}\text{B}(n,\gamma)^7\text{Li}$ reaction. As a result, liquid moderator injection pattern that contain larger volume of boric acid suppress the thermal neutron component, while water-filled or empty configurations allow higher thermal neutron intensities to reach the gold foil.

Table 4.1: HPGe detector measurement condition and reaction rate result of bare gold foil irradiation set

Pattern	Weight [mg]	Cooling time [s]	Measurement time		Peak area	Reaction rate [s ⁻¹ g ⁻¹]	Measurement uncertainty [%]
			T _{real} [s]	T _{real} [s]			
aaa	73.25	7.09 × 10 ³	1.47 × 10 ²	1.47 × 10 ²	10301	5.97 × 10 ⁶	0.99
aba	74.63	9.97 × 10 ²	2.27 × 10 ²	2.27 × 10 ²	10125	3.66 × 10 ⁶	0.99
abb	74.13	5.11 × 10 ³	6.33 × 10 ²	6.32 × 10 ²	10046	1.33 × 10 ⁶	1.00
bbb	73.86	6.51 × 10 ³	1.77 × 10 ³	1.77 × 10 ³	10313	4.91 × 10 ⁵	0.98
waa	73.60	1.20 × 10 ³	9.65 × 10 ¹	9.61 × 10 ¹	10123	8.76 × 10 ⁶	0.99
wbw	72.56	2.79 × 10 ³	6.61 × 10 ²	6.61 × 10 ²	10570	1.36 × 10 ⁶	0.97
wwb	73.73	7.29 × 10 ³	2.54 × 10 ²	2.53 × 10 ²	10159	3.39 × 10 ⁶	0.99

Table 4.2: HPGe detector measurement condition and reaction rate result of cadmium cover gold foil irradiation set

Pattern	Weight [mg]	Cooling time [s]	Measurement time		Peak area	Reaction rate [s ⁻¹ g ⁻¹]	Measurement uncertainty [%]
			T _{real} [s]	T _{real} [s]			
aaa	74.70	1.39 × 10 ³	2.04 × 10 ²	2.04 × 10 ²	9460	3.81 × 10 ⁶	1.03
aba	74.22	1.09 × 10 ³	5.15 × 10 ²	5.15 × 10 ²	10112	1.62 × 10 ⁶	0.99
abb	73.79	5.36 × 10 ³	1.75 × 10 ³	1.75 × 10 ³	10005	4.81 × 10 ⁵	1.00
bbb	73.21	1.13 × 10 ³	4.68 × 10 ³	4.68 × 10 ³	9845	1.77 × 10 ⁵	1.01
waa	73.48	1.59 × 10 ³	5.26 × 10 ²	5.26 × 10 ²	10426	1.65 × 10 ⁶	0.98
wbw	74.16	6.87×10 ³	6.49×10 ³	6.49×10 ³	9763	1.27×10 ⁵	1.01
wwb	74.01	8.92×10 ²	5.89×10 ³	5.88×10 ³	9733	1.38×10 ⁵	1.01

Table 4.3: Experimental results of thermal neutron reaction rate result of gold foils for SMS injection patterns.

Pattern	Thermal neutron reaction rate	Statistical Error(%)
aaa	1.56×10^6	1.40
aba	1.74×10^6	1.41
abb	7.59×10^5	1.42
bbb	2.80×10^5	1.42
waa	6.81×10^6	1.40
wbw	1.21×10^6	1.37
wwb	3.23×10^6	1.41

4.4.2 Neutron energy spectrum evaluation

Figure 4.8 shows the initial guess and the unfolded neutron spectrum result. Both the MAXED and GRAVEL unfolding codes exhibit the same trends following the neutron energy spectral evaluations. The fluxes of thermal neutrons and fast neutrons were higher than those of the initial guess spectrum.

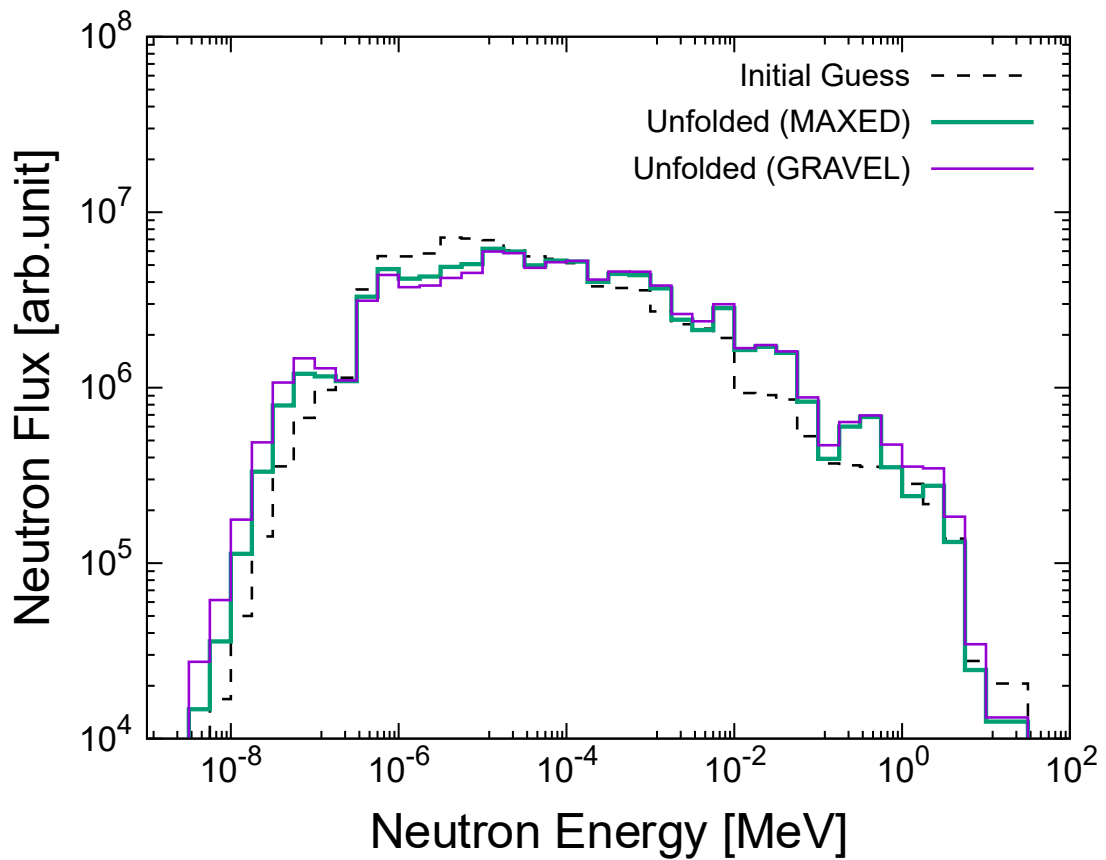


Figure 4.8: Evaluated neutron energy spectra by using SMS

4.4.3 E/C ratio

In Figure 4.9, SMS injection patterns are arranged in order of their sensitivity to neutron energy. Based on the characteristics of the response functions in Figure 4.6, liquid moderator injection patterns with higher sensitivity to lower-energy neutrons are placed on the left, while those with higher sensitivity to higher-energy neutrons are placed on the right.

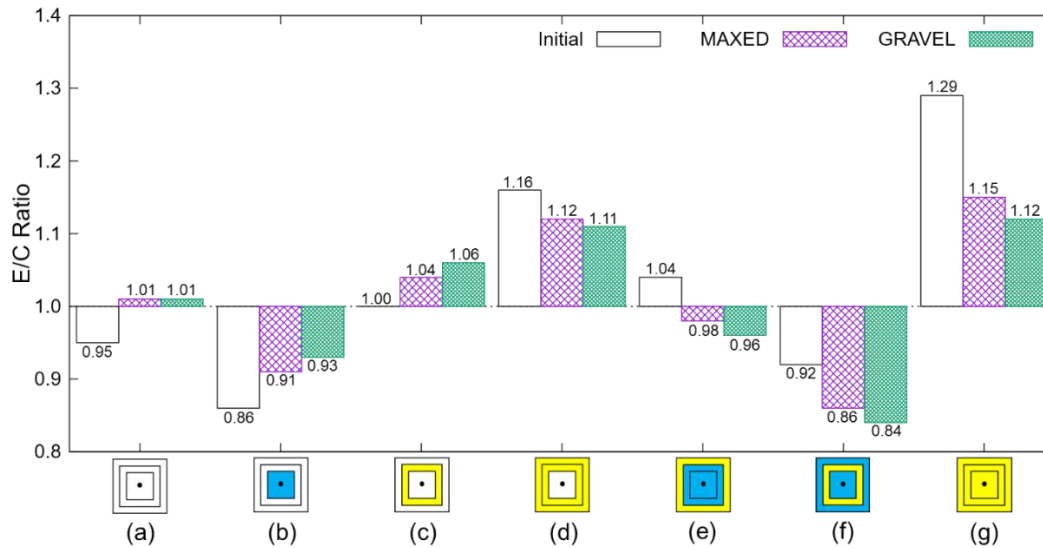


Figure 4.9: SMS injection patterns E/C ratio

(a)“aaa”, (b)“waa”, (c) “aba”, (d)“abb”, (e)“wwb”, (f)“wbw”, (g)“bbb”

E/C ratio for the initial guess, MAXED unfolding, and GRAVEL unfolding fell within the ranges of 0.86–1.29, 0.86–1.15, and 0.85–1.12, respectively. The unfolding results showed that the E/C ratio approached closer to the initial guess, indicating successful unfolding. However, an error in the E/C ratio of approximately 15% persisted.

Although the response functions of “bbb,” “wbw,” and “wwb” exhibit similar sharp peaks in the fast neutron region as shown in Figure 4.6, their E/C ratios for the initial guess differed. This phenomenon indicates an irregular result that was not expected. The effect likely arises from differences in the neutron beam intensity and angular distribution in the BNCT irradiation field compared to other neutron irradiation fields, which generally have uniform beams.

4.4.4 Challenge of neutron spectrometer development in BNCT irradiation field

In the response function calculation for CHARMS, neutron source — including its intensity and angular distribution — was incorporated to address this issue. However, the problem could not be resolved, possibly because the neutron source definition did not accurately represent the conditions at the CHARMS evaluation position, or because PHITS may not correctly calculate the response function under these conditions.

Figure 4.10 shows thermal neutron intensity calculated by PHITS at KUR-HWNIF for two irradiation configurations: (a) with the collimator and (b) without the collimator. In particular, the

presence of the collimator introduces non-uniform beam conditions (Position a in Figure 4.10), which can limit the accuracy of the neutron energy spectrum evaluation. For the full-scale CHARMS system, measurement setups under more uniform beam conditions will be considered, such as measurements performed at positions farther from the collimator (Position b in Figure 4.10) or configurations without a collimator (Position c in Figure 4.10). These approaches are expected to improve accuracy of the response functions calculation result and enhance the accuracy of neutron energy spectrum evaluation.

In the full-scale CHARMS experiment (Chapter 5), irradiation without a collimator, which provides a more parallel neutron beam than the collimated configuration, is considered to achieve better results in spectrometry study for BNCT.

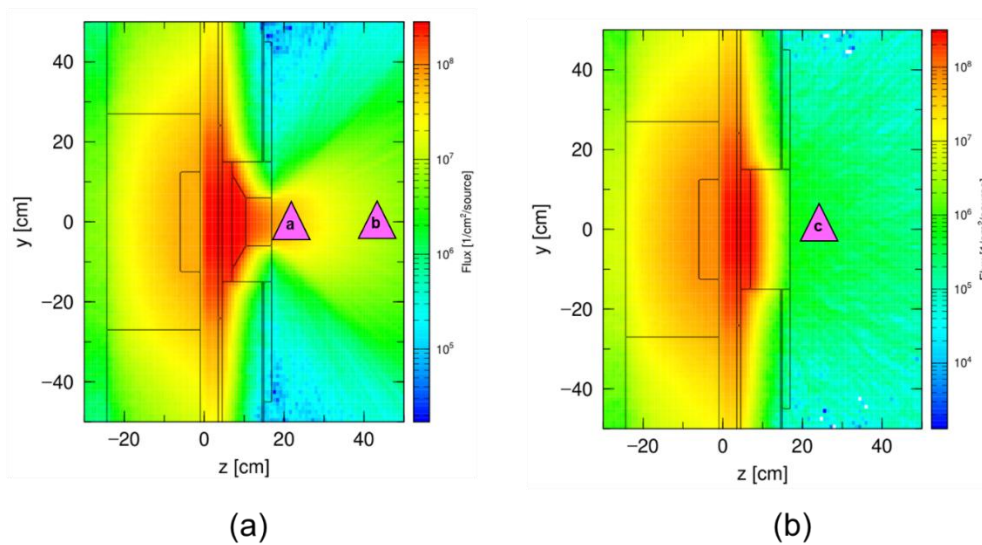


Figure 4.10: Thermal neutron intensity in KUR-HWNIF

(a) with collimator irradiation setup, (b) without collimator irradiation setup

4.5 Conclusion

A moderator combination–selection-based multilayer neutron spectrometer was verified using the SMS. Using SMS, the neutron energy spectrum in the epithermal irradiation mode at a distance of 11.5 cm from the collimator of the KUR-HWNIF was evaluated with an uncertainty below 15%. The results of this study demonstrate the effectiveness of the moderator combination–selection-based multilayer neutron spectrometer for evaluating the neutron energy spectrum in the BNCT irradiation field.

However, the E/C results indicated irregularities that likely originated from non-uniform neutron beam intensity and angular distribution in the BNCT irradiation field, which differ from those in ideal uniform-beam conditions. This finding provides an important hint for the next full-scale CHARMS experiment, suggesting that irradiation conditions with reduced intensity and angular variations should be employed for better results in spectrometry study for BNCT.

Chapter 5 Development and validation of remote-operating liquid multilayer neutron spectrometer for quality assurance

In Chapter 3, CHARMS was optimized through simulation, and the liquid moderator injection patterns were selected to obtain response functions with improved energy resolution, enabling more accurate evaluation of the neutron energy spectrum. In Chapter 4, SMS was constructed and tested as a proof of concept, confirming the validity of the multilayer moderator approach in the BNCT irradiation field with careful consideration of intensity and angular property of BNCT neutron irradiation field. This chapter describes the development and establishment of the full-scale CHARMS system with remote operation capability for use in BNCT quality assurance (QA) procedures.

5.1 Update structure of CHARMS

Figure 5.1 shows update CHARMS structure. In Chapter 3, the structure of liquid moderator container of CHARMS was designed. the length of cylinder part of the liquid moderator container was designed to be 10 cm. However, the difficulties with the liquid injection and drainage operation was found, as the liquid moderator inside the container could not be injected or drained effectively. As a result, the cylindrical parts of the first layer (innermost) and the second layer (middle) were extended to 20 cm and 15 cm respectively.

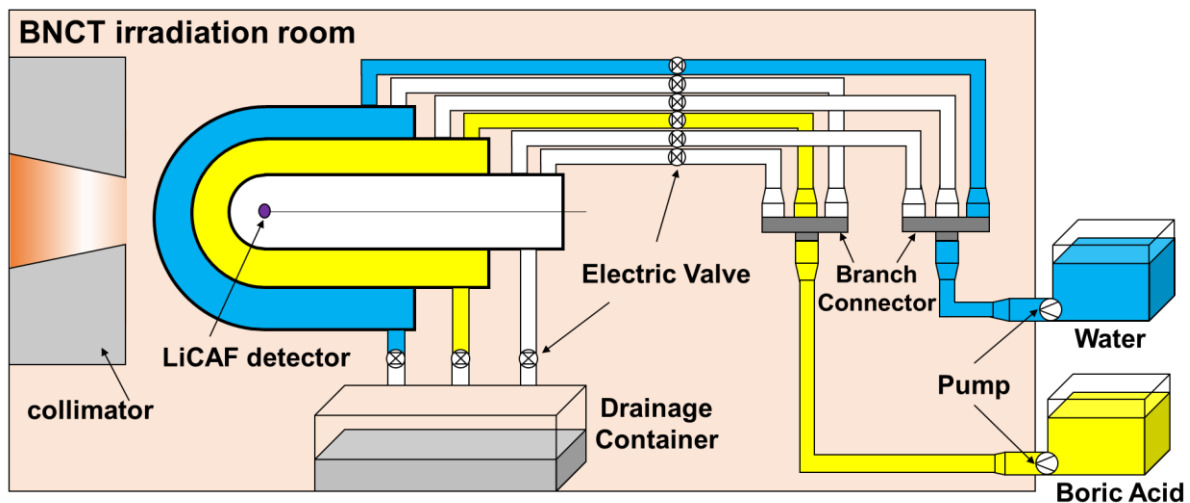


Figure 5.1: Update CHARMS Structure

Figure 5.2 shows the structure of the CHARMS liquid moderator containers, which consist of three different sizes (I–III), and a neutron detector insertion tube (IV) designed as a nested structure for easy assembly. Containers I–III are composed of cylindrical sections with lengths of 10 cm, 15 cm, and 20 cm, combined with hemispherical sections of 15 cm, 10 cm, and 5 cm in diameter, respectively. These containers serve as space for the liquid moderators. Container IV is a cylindrical

tube with a length of 21 cm, designed to insert LiCAF neutron detector. After assembly, three layers of liquid moderators can be injected, enabling CHARMS to realize various moderator injection patterns. Furthermore, as shown in Figure 5.1, the drainage container was installed inside the irradiation room, in contrast to the design expectation described in Figure 3.1.

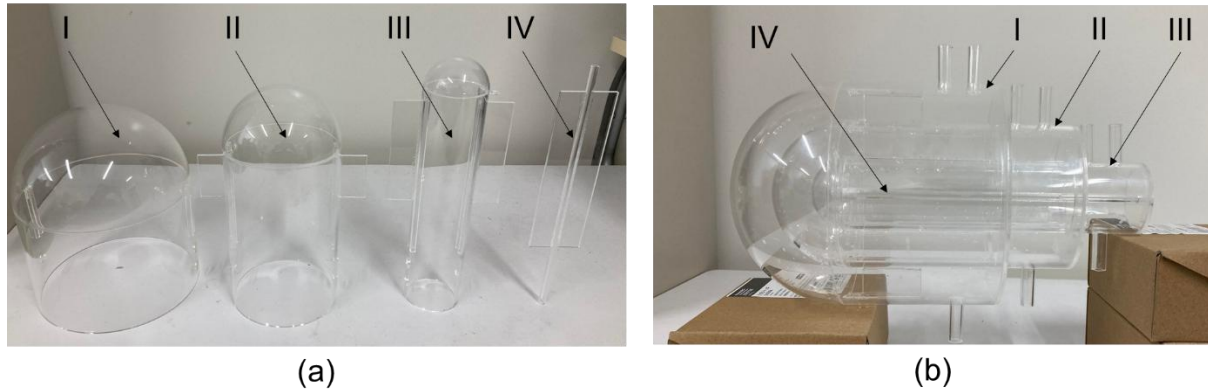


Figure 5.2: CHARMS acrylic container structure
(a) before assembly, (b) after assembly

5.2 CHARMS system

To implement CHARMS for the BNCT QA protocol, we constructed three systems: a liquid moderator supply and drainage, neutron detection, and monitoring (Figure 5.3), as described below.

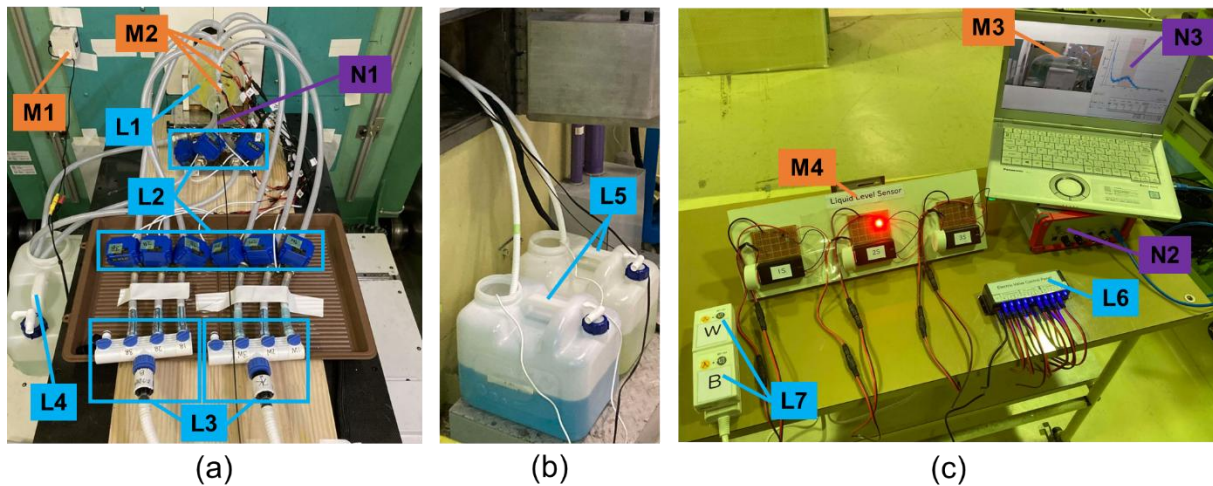


Figure 5.3: CHARMS structure at KUR-HWNIF BNCT irradiation field.

(a) equipment inside irradiation room, (b) liquid moderator supply outside irradiation room
(c) on-desk controller outside irradiation room

5.2.1 Liquid moderator supply and drainage system

CHARMS liquid moderator supply and drainage system allowed liquid moderators to be injected into or drained from the container inside the irradiation room, with the control operated remotely from outside the irradiation room. At the operating desk, two pump controllers manage the injection of the liquid moderators, pure water and boric acid, from the supply container into the acrylic container. In addition, the electric valves inside the irradiation room, which are controlled by a control panel at the operation desk, allow the injection or drainage of the liquid moderator into the desired layer separately. For the liquid moderator drainage, we took advantage of the natural free-fall property by positioning a drainage container at the bottom of the CHARMS and using an electric valve to control the flow. Table 5.1 gives details of equipment in liquid moderator supply and drainage system.

Table 5.1: Equipment in liquid moderator supply and drainage system

Label in Figure 5.3	Equipment
L1	Liquid moderator acrylic container (Figure 5.2(b))
L2	9 Electric valves (U.S. Solid, USS-MSV00010-G)
L3	2 Branch connectors (LIFELEX, M22820A)
L4	Drainage container
L5	liquid moderator (water and boric acid) supply containers with 2 pumps (LIFELEX, BP-104N)
L6	Electric valve control panel (OTraki, B07Rddb8QQ)
L7	Pump controller (LIFELEX, BP-104N)

5.2.2 Neutron detection system

Table 5.2 gives details of equipment in neutron detection system.

Table 5.2: Equipment in neutron detection system

Label in Figure 5.3	Equipment
N1	Optical fiber connected with a LiCAF scintillator and Photo Multiplier Tube (C&A, FD-L(6-6)Q1010)
N2	Multi-channel Analyzer (ZMCAN-CH02-02)
N3	Pulse Height distribution display

Figure 5.4 (a) and (b) shows neutron detection system in diagram and actual item respectively. When neutron reached Eu:LiCaAlF_6 (LiCAF) scintillator with dimensions of $0.6 \text{ mm} \times 0.6 \text{ mm} \times 0.6 \text{ mm}$ (FD-L(6-6)Q1010, C&A corporation), the light produced from the ${}^6\text{Li}(n,\alpha){}^3\text{H}$ nuclear reaction is transmitted through an optical fiber to a photomultiplier tube (PMT). The PMT output is then

processed by a 2 channel multi-channel analyzer (MCA) (ZMCAN-CH02-02, ANSeeN) that incorporates high voltage power supply, amplifier, shaper, and analog-to-digital converter. The count rate recorded by the LiCAF scintillator primarily reflects neutron interactions with ^6Li which highly sensitive to thermal neutron energy region.

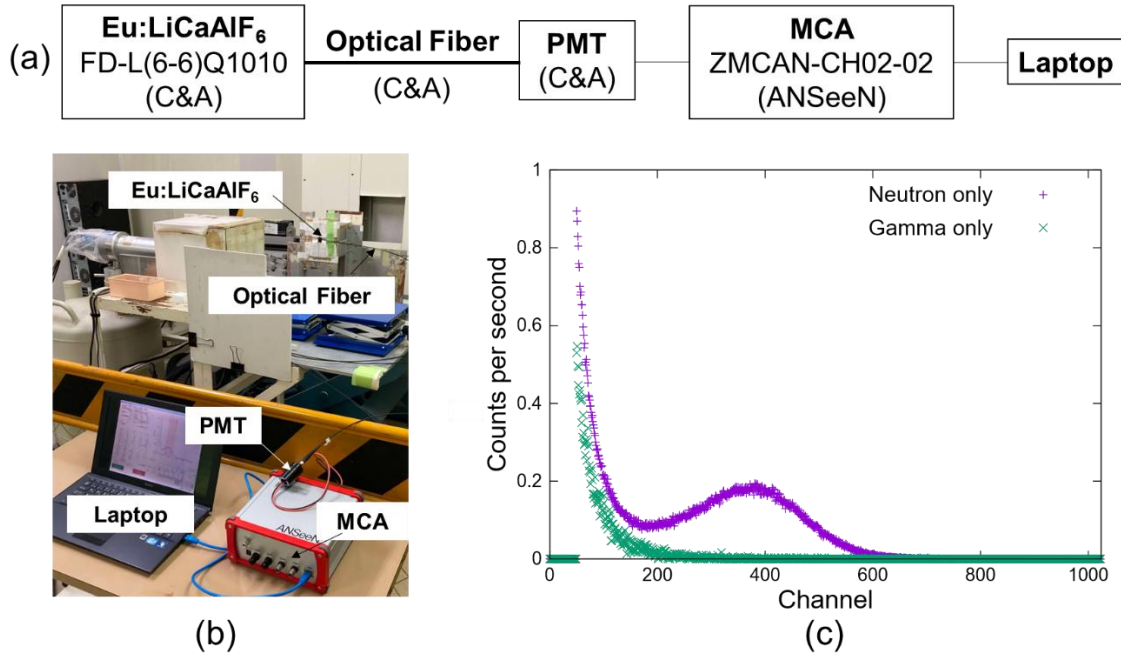


Figure 5.4: LiCAF detector

(a) configuration, (b) actual item,

(c) pulse-height distribution demonstrating effective gamma–neutron discrimination

Figure 5.4 (c) shows pulse-height spectra measured by the neutron detection system at E3 Prompt Gamma Analysis Facility of KUR. To confirm the gamma–neutron discrimination capability of the detector, two measurements were performed: irradiation with the detector exposed to neutrons only (Neutron only in Figure 5.4 (c)), and irradiation with a cadmium plate placed in front of the detector to suppress neutrons and generate a gamma-ray–dominant field (Gamma only in Figure 5.4 (c)).

Gamma-ray events were mainly observed below approximately 200 channels, while the neutron-induced Gaussian peak appeared above this region. Although the spectrum contains some gamma-ray events, particularly in the low-channel region, the main peak located at approximately 320–330 channels was fitted with a Gaussian function. The neutron reaction rate was defined as the count rate from the mean value up to one standard deviation on the positive side of the peak in order to exclude gamma-ray contributions, where σ represents the standard deviation of the fitted distribution. Figure 5.5 shows an example of a measurement obtained with the CHARMS system at KUR-HWNIF using this fitting method.

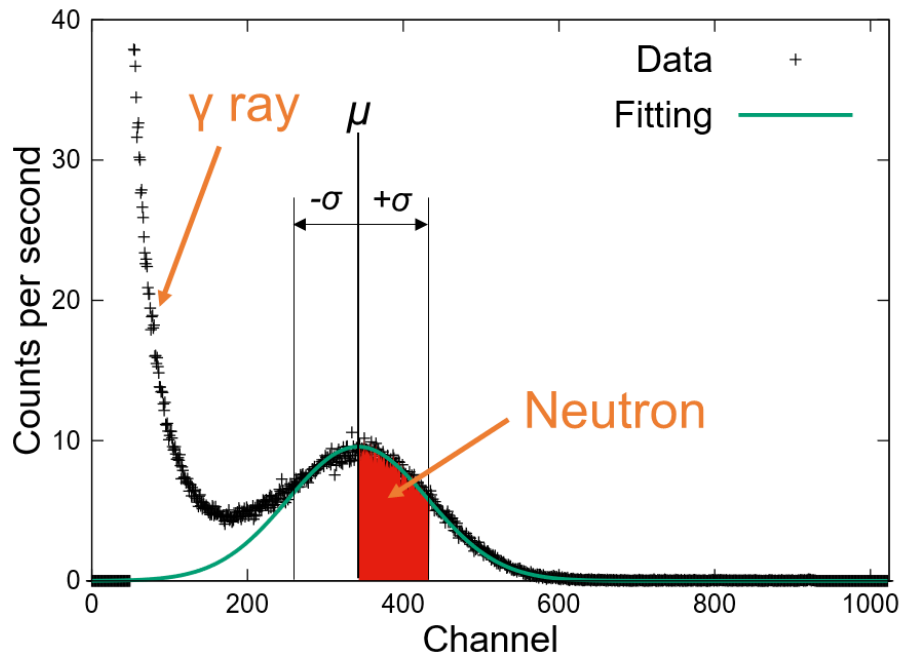


Figure 5.5: Pulse-height distribution measured with the LiCAF detector under BNCT irradiation conditions, showing Gaussian fitting used for neutron event extraction

5.2.3 Monitoring system

Monitoring the status of the liquid moderator inside irradiation room is important. Because direct observation is not possible from the outside, a camera was installed to provide the real-time liquid moderator situation of CHARMS. In addition, liquid moderator level sensors were positioned at the top of each layer, and when the liquid moderator reached the top, a signal was sent to an external control panel. This monitoring system is critical for effective operation of the liquid moderator supply and drainage system. Table 5.3 gives details of equipment in monitoring system.

Table 5.3: Equipment in monitoring system

Label in Figure 5.3	Equipment
M1	Camera with neutron shielding
M2	Liquid level sensors
M3	Real time video display
M4	Liquid level sensor display

5.3 Simulation calculation

5.3.1 Initial guess spectrum

The initial guess spectrum was calculated using 11-group annular neutron source with a radius of 13.5 cm of KUR-HWNIF under two different BNCT irradiation field conditions: with and without a

collimator. As described in Subsection 4.4.4 about non-uniform neutron irradiation field of BNCT, without collimator irradiation is introduced in this study. Figure 5.6 shows the irradiation geometry of CHARMS at the KUR-HWNIF with and without collimator irradiation, respectively. In these calculations, CHARMS and its wooden support structure were excluded to obtain the in-air neutron energy spectrum.

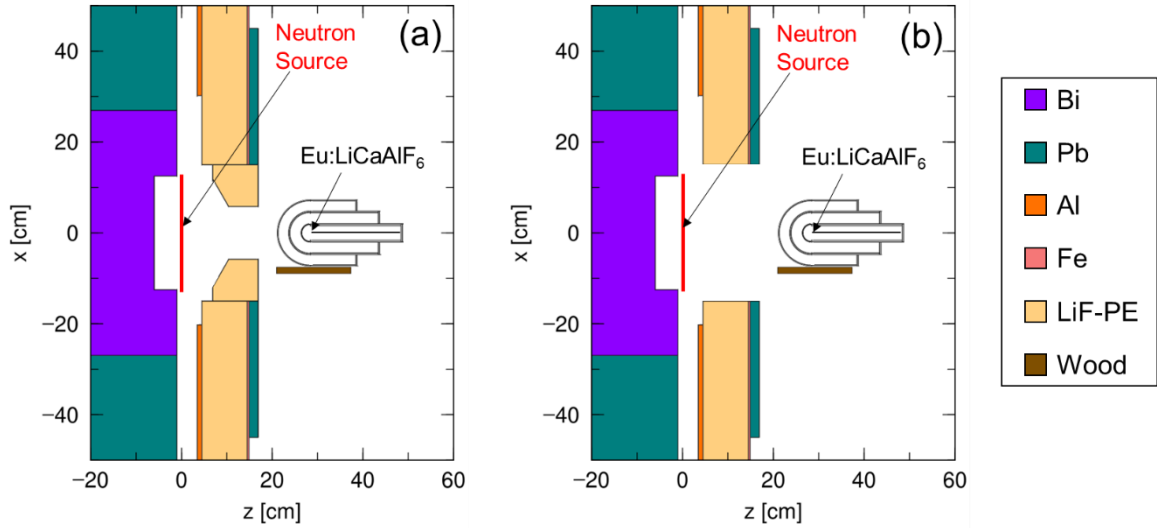


Figure 5.6: Calculation geometry of CHAMRS setup at KUR-HWNIF BNCT irradiation field
(a) with collimator, (b) without collimator

Figure 5.7 shows the calculated initial guess for the neutron energy spectrum under both with and without collimator irradiations. Owing to the effect of the collimator, the total neutron flux decreased by a factor of approximately three, whereas the number of thermal neutrons increased slightly because of the moderation process in the collimator.

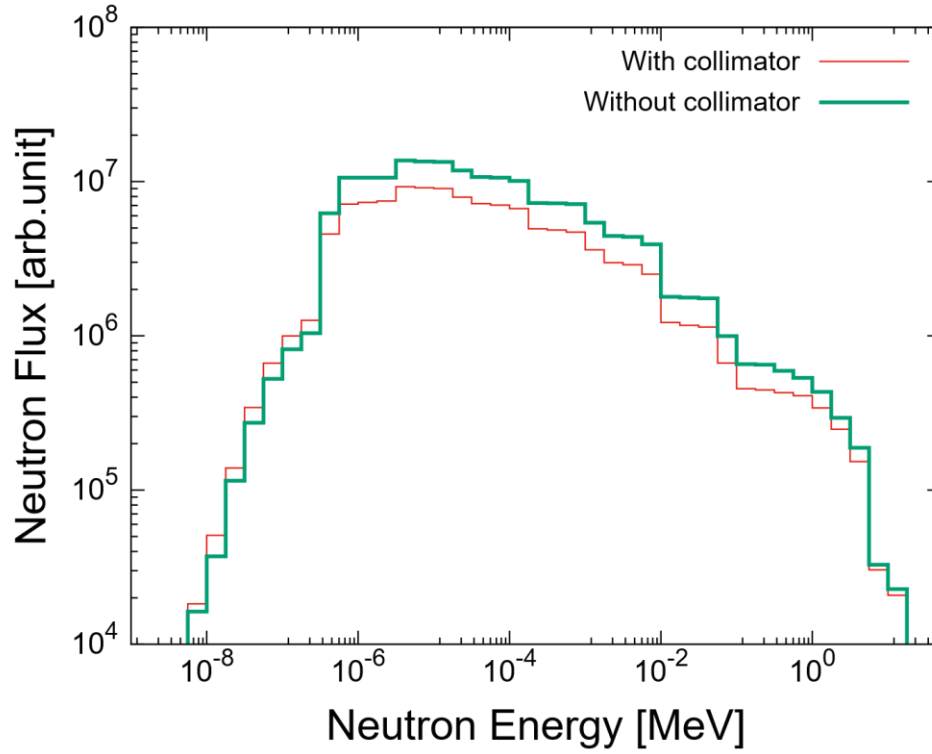


Figure 5.7: Initial guess of neutron energy spectrum calculation result

5.3.2 Response function

The responses of the seven liquid moderator injection patterns of CHARMS were calculated by PHITS using the irradiation geometry shown in Figure 5.6 (a) and (b) with and without collimator irradiation, respectively. The incident neutron beam consisted of monoenergetic neutrons with energies ranging from 10^{-9} to 31.6 MeV, divided into 42 energy bins equally spaced on a logarithmic scale same with SMS response function calculation.

Figure 5.8 shows the calculated response function of seven CHARMS liquid moderator injection patterns with and without collimator irradiation setup. The shapes of the response functions for both the with and without collimator conditions were similar; however, the presence of the collimator resulted in a relatively higher response function in the fast-neutron region. This effect is attributed to the material properties of the collimator, composed of LiF–polyethylene, which serves to thermalize a greater fraction of high-energy neutrons compared to the condition without a collimator. Since the CHARMS system employs a thermal neutron detector, fast neutrons that are moderated to thermal energies by the collimator contribute more significantly to the thermal neutron under the with-collimator condition.

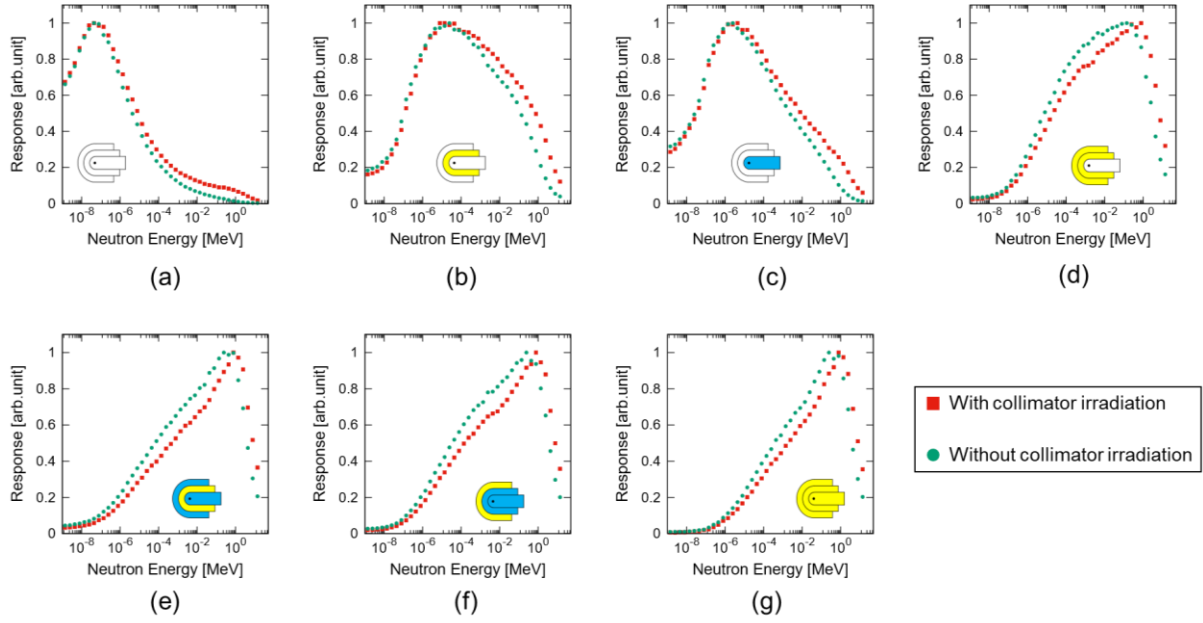


Figure 5.8: Response function of CHARMS

(a)“aaa”, (b)“aba”, (c) “waa”, (d)“abb”, (e)“wbw”, (f)“wwb”, (g)“bbb”

5.4 Irradiation experiment

Figure 5.9 (a) and (b) show the CHARMS irradiation experiments performed with and without a collimator, respectively. Both conditions were investigated because they are routinely employed at our facility: the configuration with a collimator is primarily used in clinical research, yielding a forward-directed neutron beam intensity, whereas the configuration without a collimator is applied in biological irradiation, providing a nearly isotropic neutron beam. To validate CHARMS in routine BNCT QA procedures, irradiation experiments were conducted three times over three separate months.

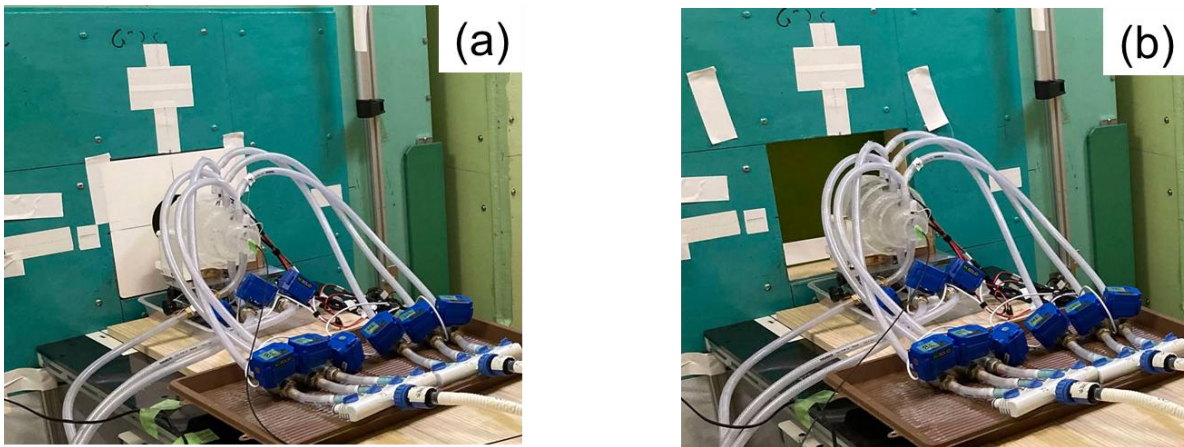


Figure 5.9: CHARMS irradiation experiment setup at KUR-HWNIF

(a) with collimator, (b) without collimator

5.5 Result and discussion

5.5.1 Measurement time

To reduce the measurement uncertainty, the CHARMS neutron detection system is operated until the neutron count exceeds 10,000, which corresponds to an uncertainty of less than 1%. Table 5.4 shows the measurement times for CHARMS. The combined time for both with and without collimator irradiation conditions was less than 10 min.

The measurement times for both the with and without collimator irradiations remained under 10 min, maintaining an uncertainty of neutron count less than 1%. This rapid measurement capability suggests that CHARMS is well suited for routine QA procedures, where efficiency is essential.

Table 5.4: Each CHARMS liquid moderator injection pattern measurement time

Injection Pattern	Measurement time [s]	
	With collimator irradiation condition	Without collimator irradiation condition
aaa	17	8
aba	24	10
abb	65	27
bbb	160	60
waa	10	6
wbw	40	15
wwb	15	7
Total	331	133

5.5.2 Stability of CHARMS during measurements

To assess the stability of the CHARMS liquid moderator injection and drainage system, the neutron counts were monitored at counts were monitored at ten equally spaced time points throughout the irradiation for each liquid moderator geometry. Figure 5.10 shows neutron counts under collimator irradiation conditions in a1, b1, and c1, and under conditions without collimator irradiation in a2, b2, and c2, corresponding to the first, second, and third measurements, respectively. The majority of the counts in each interval fell within the $\mu \pm \sigma$ range, where μ represents the average neutron count across the ten intervals and σ is the standard deviation based on Poisson distribution.

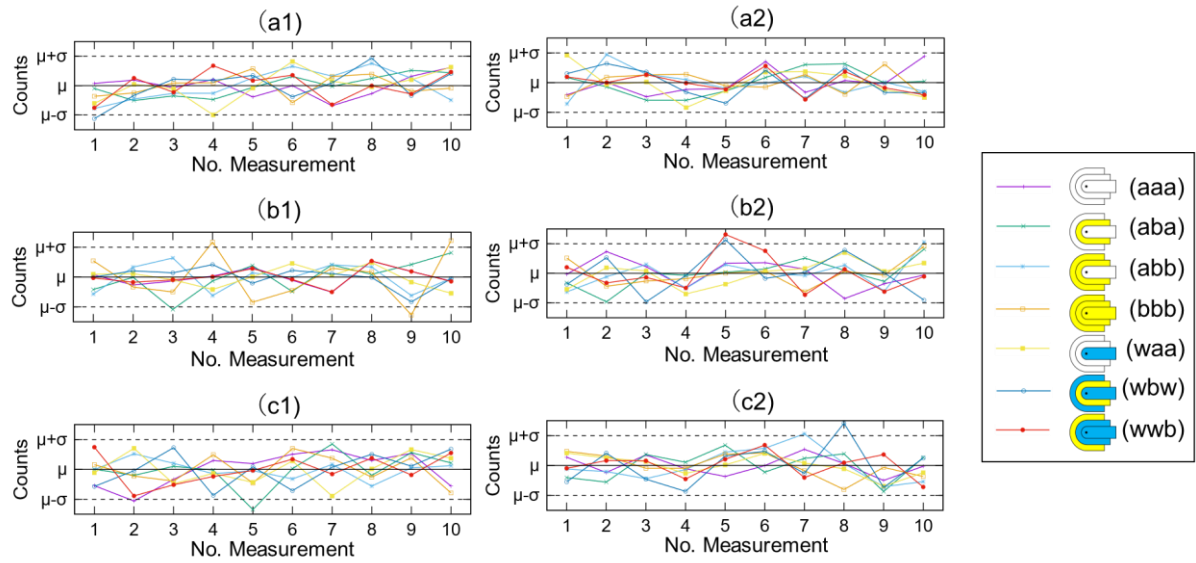


Figure 5.10: Variability of neutron detection of CHARMS

First measurement: (a1) with and (a2) without collimator

Second measurement: (b1) with and (b2) without collimator

Third measurement: (c1) with and (c2) without collimator

The stability tests showed that the liquid moderator injection and drainage system stably injected and drained the liquid moderator, yielding a consistent neutron detection signal throughout the measurement period, as evidenced by the neutron counts over ten intervals remaining within one Poisson-derived standard deviation.

5.5.3 LiCAF detector measurement

Table 5.5 summarizes the thermal neutron response rates obtained from the LiCAF detector in three different measurement both with and without collimator irradiation setup. The response rates were derived from the fitted pulse-height spectra using the predefined region of interest (ROI), while the detailed raw data and fitting procedure are described in Appendix F.

Table 5.5: Thermal neutron response rate from LiCAF detector (arb.unit)

Injection pattern	First measurement		Second measurement		Third measurement	
	With collimator	Without collimator	With collimator	Without collimator	With collimator	Without collimator
aaa	3.11×10^3	6.91×10^3	2.78×10^3	6.46×10^3	2.75×10^3	6.42×10^3
aba	2.43×10^3	5.29×10^3	2.13×10^3	4.86×10^3	2.11×10^3	4.82×10^3
abb	8.49×10^2	2.02×10^3	7.54×10^2	1.79×10^3	7.32×10^2	1.84×10^3
bbb	3.34×10^2	8.42×10^2	3.02×10^2	7.85×10^2	3.00×10^2	7.93×10^2
waa	7.73×10^3	1.74×10^4	6.79×10^3	1.70×10^4	6.88×10^3	1.64×10^4
wbw	1.51×10^3	3.64×10^3	1.30×10^3	3.23×10^3	1.30×10^3	3.27×10^3
wwb	3.72×10^3	7.69×10^3	3.15×10^3	6.89×10^3	3.26×10^3	6.93×10^3

Table 5.6 summarizes the measurement-to-measurement variation of the thermal neutron response rates obtained from three repeated measurements. For all patterns, the coefficients of variation were within approximately 6–9% under collimated conditions and 3–7% without the collimator, indicating good reproducibility of the LiCAF detector response. The non-collimated measurements generally exhibited smaller coefficient of variation (CV) values, reflecting the higher neutron intensity and improved counting statistics.

From QA perspective, variations within 5% are considered acceptable. While some measurements slightly exceeded this threshold, the results remain acceptable when setup-related uncertainties are considered.

Table 5.6: Measurement-to-measurement variation of the thermal neutron response rates

Injection pattern	With collimator		Without collimator	
	Mean $\pm$ Standard derivation	CV (%)	Mean $\pm$ Standard derivation	CV (%)
aaa	$(2.88 \pm 0.20) \times 10^3$	6.94	$(6.60 \pm 0.27) \times 10^3$	4.12
aba	$(2.22 \pm 0.18) \times 10^3$	8.06	$(4.99 \pm 0.26) \times 10^3$	5.22
abb	$(7.78 \pm 0.62) \times 10^2$	7.99	$(1.88 \pm 0.12) \times 10^3$	6.42
bbb	$(3.12 \pm 0.19) \times 10^2$	6.11	$(8.07 \pm 0.31) \times 10^2$	3.83
waa	$(7.13 \pm 0.52) \times 10^3$	7.27	$(1.69 \pm 0.05) \times 10^4$	2.97
wbw	$(1.37 \pm 0.12) \times 10^3$	8.85	$(3.38 \pm 0.23) \times 10^3$	6.69
wwb	$(3.38 \pm 0.30) \times 10^3$	8.95	$(7.17 \pm 0.45) \times 10^3$	6.29

5.5.4 Neutron energy spectrum evaluation

Figure 5.11—Figure 5.13 show the three unfolded neutron energy spectra for the irradiation without beam collimation in three different measurement time. In this study, determining the absolute neutron energy spectrum from the measured neutron count was not possible because variations in parameters such as the power supply voltage, temperature, and other conditions at different measurement times affect the response of the neutron detection system. Therefore, scaling factor was not applied to unfolding process and total neutron flux was normalized to 1.0.

For collimator irradiation setup, regular unfolding result was not received which will be explained in Subsection 5.5.5.

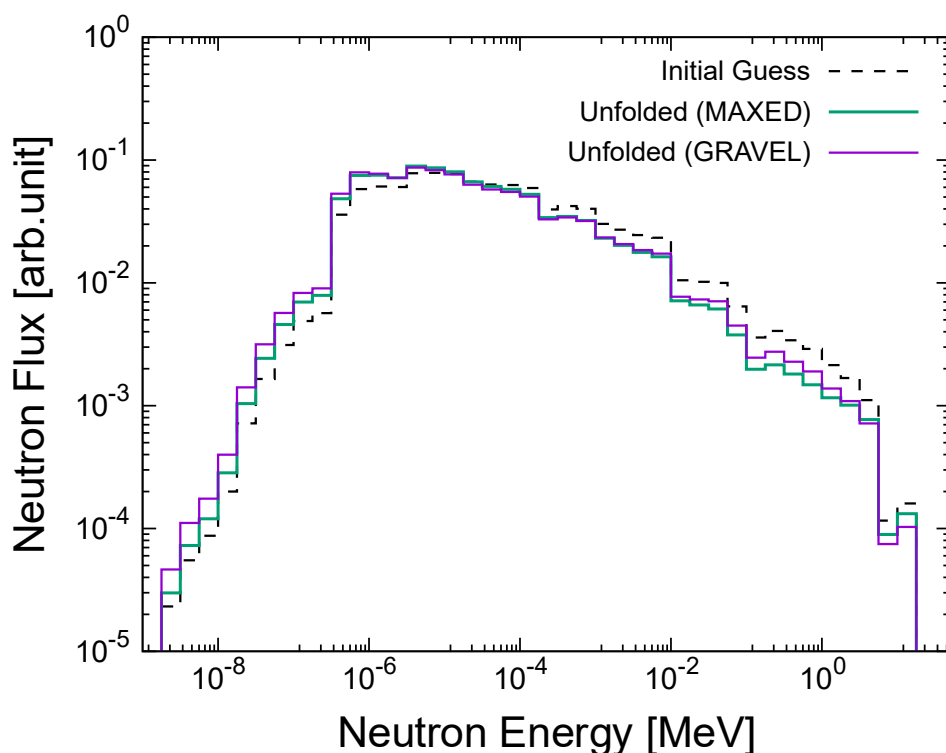


Figure 5.11: First measurement of evaluated neutron spectrum by using CHARMS

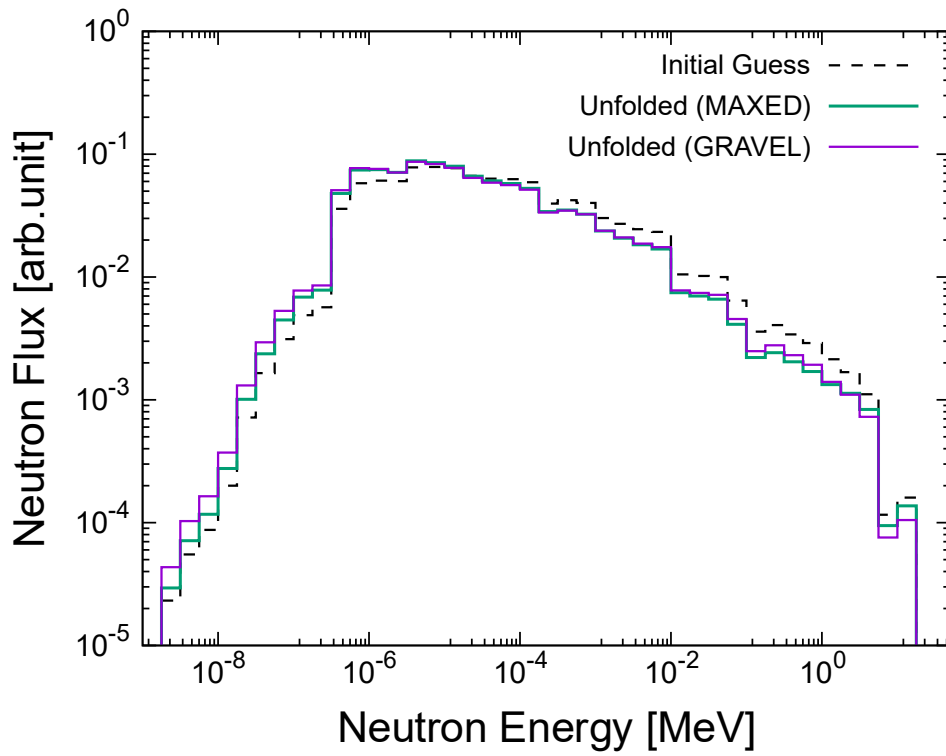


Figure 5.12: Second measurement of evaluated neutron spectrum by using CHARMS

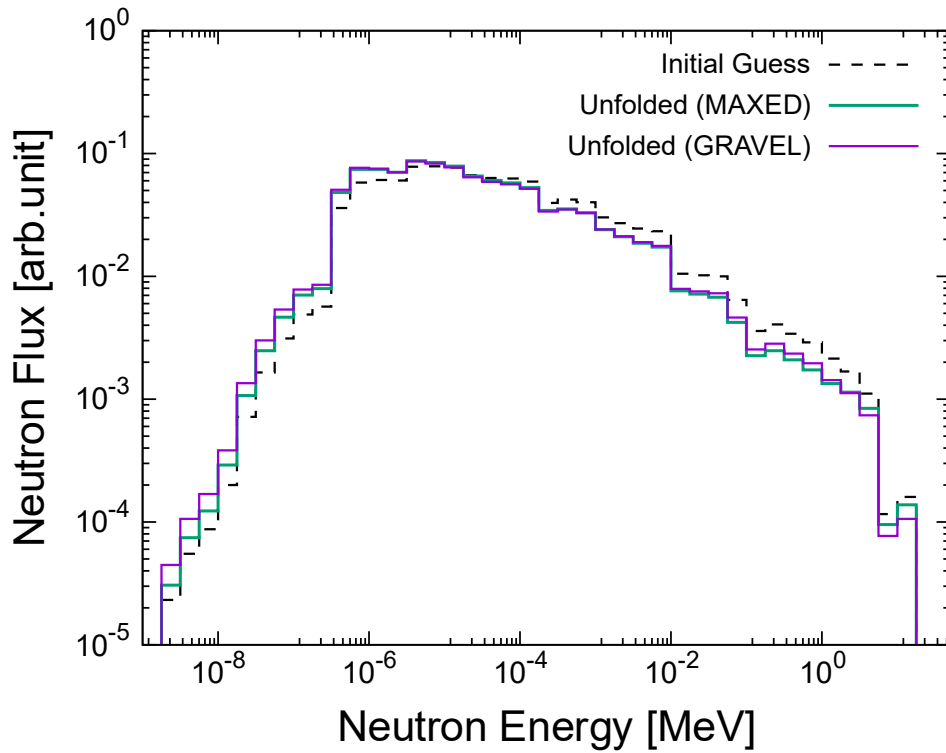


Figure 5.13: Third measurement of evaluated neutron spectrum by using CHARMS

5.5.5 E/C ratio

Figure 5.14—Figure 5.16 presents the E/C ratios for three different irradiation times with and without collimator irradiation. The E/C ratios were normalized by dividing each value by E/C data of “bbb” liquid moderator injection pattern because absolute neutron detection result cannot obtain by LiCAF.

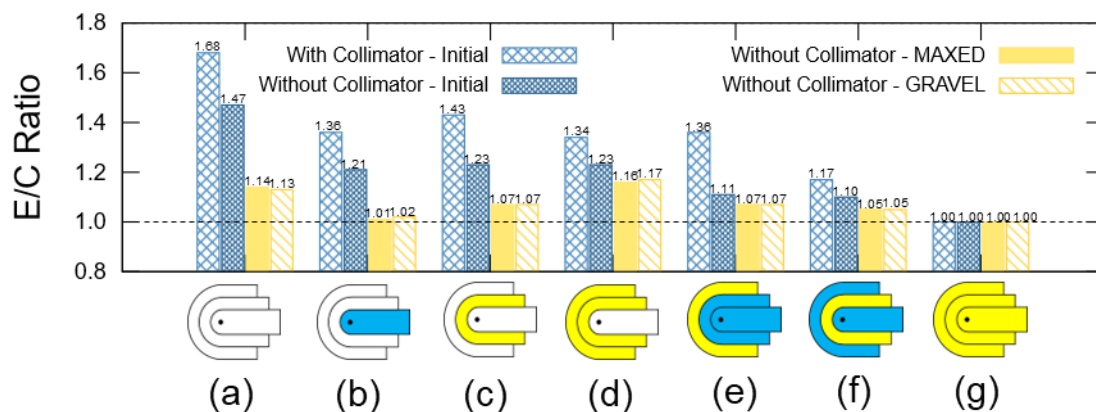


Figure 5.14: E/C ratio of the first measurement

(a)“aaa”, (b)“waa”, (c) “aba”, (d)“abb”, (e)“wwb”, (f)“wbw”, (g)“bbb”

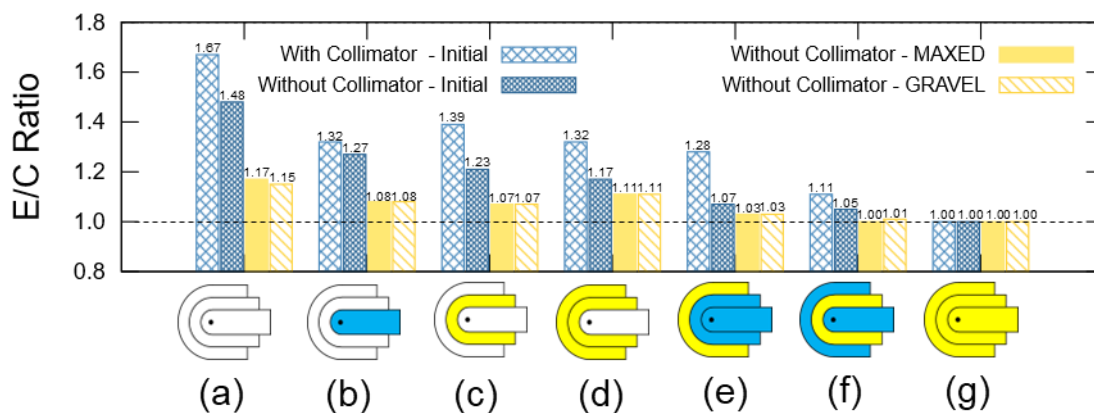


Figure 5.15: E/C ratio of the second measurement

(a)“aaa”, (b)“waa”, (c) “aba”, (d)“abb”, (e)“wwb”, (f)“wbw”, (g)“bbb”

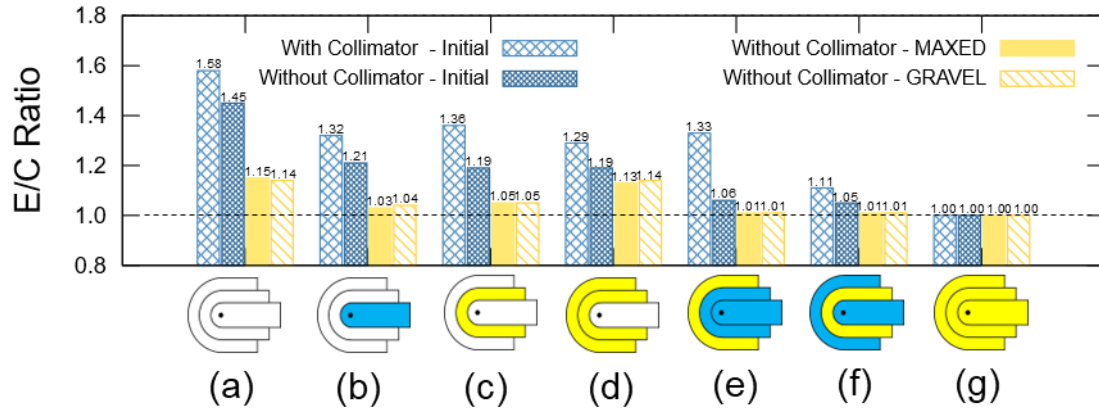


Figure 5.16: E/C ratio of the third measurement

(a)“aaa”, (b)“waa”, (c) “aba”, (d)“abb”, (e)“wwb”, (f)“wbw”, (g)“bbb”

Only the initial guess of neutron energy spectrum E/C ratios was obtained under the with-collimator condition, and neither the MAXED nor the GRAVEL unfolding codes produced a proper unfolded spectrum that differed significantly from the initial guess. In contrast, for the irradiation setup without a collimator, the proper unfolded spectra were consistently obtained across all three measurement sessions.

Liquid moderator injection pattern that are sensitive to lower neutron energies tend to yield higher E/C ratios. Specifically, pattern “aaa”, which was highly sensitive to low-thermal neutrons, exhibited the highest E/C ratio. The liquid moderator geometries that are sensitive to high-thermal neutrons (“aba” and “waa”) and epithermal neutrons (“abb”) have intermediate E/C values, whereas those sensitive to high-energy epithermal neutrons (“wbw”) and fast neutrons (“wwb”) exhibit slightly lower E/C ratios. The response functions for the “abb”, “wbw”, and “wwb” injection pattern are very similar; in an ideal case, their E/C ratios are identical.

However, in the “abb”, one of liquid moderator layers is left unfilled; it is more affected by variations in the BNCT neutron beam intensity and angular distribution than “wbw” and “wwb”. Because this study considered only the energy distribution, these additional effects led to the E/C ratio evaluation results. For the collimator irradiation condition, the order of the E/C values was similar to that without the collimator. However, the E/C value for pattern “wwb” was irregularly high, almost matching the thermal neutron “waa” and epithermal “abb” sensitive patterns. This suggests that the intensity and angular distribution of the BNCT irradiation field play significant roles even in without collimator irradiation setup.

5.6 Conclusion

In this study, the validity of CHARMS as a QA tool for BNCT was confirmed. The rapid measurements and stable operation of the liquid moderator injection and drainage subsystem show that CHARMS is well suited for routine BNCT QA procedures, eliminating the need for moderator replacement, which can minimize radiation exposure. A comparison under different irradiation conditions — with and without collimator — revealed that E/C ratios vary with the neutron-energy sensitivity of the liquid moderator injection patterns.

The variations in the BNCT neutron beam intensity and angular distribution influenced the E/C results were found, particularly under collimator irradiation conditions where a reliable energy spectrum could not be obtained. Future work will address the impact of BNCT neutron beam intensity and angular distribution, with the aim of improving neutron energy spectrum evaluation as an important consideration for clinical BNCT application.

Chapter 6 Summary

This chapter summarizes how the challenges identified in Chapter 1.6. were addressed through the results presented in Chapter 2 - Chapter 5, In addition, future perspectives for further development of the neutron spectrometer system are briefly discussed.

6.1 Improvement in neutron source data reliability

As discussed in Subsection 1.6.1, conventional neutron source data verification by the multifoil activation method has relied on high-fluence irradiation conditions.

In Chapter 2, this study demonstrated that, by optimizing foil selection and geometry, reliable neutron energy spectrum evaluation can be achieved even under reduced neutron fluence conditions approximately 70% compared with that used in previous high-fluence studies. A threshold of 100 counts per hour was introduced as a screening criterion to exclude activation reactions with excessively large statistical uncertainties. Based on Poisson statistics, this threshold corresponds to a statistical uncertainty of approximately 10%, ensuring sufficient counting reliability for subsequent spectrum unfolding. By applying this criterion, reactions that are impractical under low-fluence irradiation conditions were systematically excluded from the analysis.

The evaluated neutron energy spectra showed agreement within approximately 20% for epithermal neutrons and 30% for fast neutrons, satisfying the target accuracy defined for spectrometer development purposes. This result confirms that neutron source data suitable for simulation calculations can be obtained without requiring high-fluence irradiation, thereby improving the applicability of the method to accelerator-based BNCT systems.

However, several limitations of the multifoil activation method remain and may affect the accuracy of neutron energy spectrum evaluation. In particular, in the neutron energy range from approximately 10 keV to 1 MeV, no activation foil with a sufficiently sharp and high sensitivity response is currently available, resulting in reduced spectral sensitivity in this region. One possible approach to mitigate this limitation is the use of additional moderating or filtering materials in combination with activation foils to broaden the effective response into this energy range. Such modifications, however, require careful optimization to ensure that the added materials remain sufficiently small so as not to disturb the original neutron field. Further studies are therefore needed to optimize material type, thickness, and geometry.

6.2 Reduction of setup-related measurement errors

Chapter 1.6.2 addressed setup errors arising from repeated mechanical changes in conventional Bonner sphere spectrometers. SMS and the CHARMS concept were introduced to mitigate these issues.

For the SMS configuration (Chapter 4), the detector position was fixed and the cuboid-shaped moderator container enabled consistent and straightforward placement, resulting in highly stable measurement conditions across repeated setups. No systematic variation attributable to detector repositioning was observed, indicating that setup-related uncertainties were negligible.

For the CHARMS system (Chapter 5), experimental results further demonstrated improved measurement reproducibility achieved by maintaining a fixed detector position while changing spectrometer configurations through liquid moderator injection and drainage. To evaluate the stability of the liquid moderator control system, neutron counts were monitored at ten equally spaced time points during the first, second, and third measurements. In all cases, the majority of the measured counts fell within the $\mu \pm \sigma$ range, indicating stable neutron detection throughout the measurement period.

These results suggest that setup-related uncertainties were effectively suppressed compared with conventional procedures that require repeated physical reconfiguration of neutron spectrometer.

6.3 Enhancement of epithermal neutron energy resolution

As discussed in Subsection 1.6.3, limited sensitivity in the epithermal neutron region is a major drawback of conventional neutron spectrometers in BNCT fields.

In Chapter 3, the introduction of boric-acid-containing liquid moderators and the optimization of response functions using the Backus–Gilbert approach led to a clear improvement in spectral resolution. The optimized response functions exhibited reduced full width at half maximum (FWHM) values in the epithermal neutron region compared with conventional configurations, indicating a more confined energy response. This improvement enabled more stable and reliable neutron spectrum unfolding result, which is critical for BNCT beam characterization.

In this study, the optimization was performed based on measurements in BNCT irradiation fields, with emphasis on the epithermal neutron region. However, for other neutron fields, such as fast reactors, greater emphasis on fast neutron sensitivity would be required. The Backus–Gilbert approach allows the optimization conditions to be flexibly adjusted according to the target neutron energy range, which is a key advantage of this method and enables its application to a wide variety of neutron fields.

6.4 Reduction of operator radiation exposure

Subsection 1.6.4 highlighted radiation exposure to operators during repeated entry into the irradiation room. In Chapter 5, the remote-operated CHARMS system demonstrated that spectrometer configuration changes and neutron measurements could be performed without entering the irradiation room during irradiation.

By eliminating approximately six to seven room entries per measurement session, operator exposure was substantially reduced. Although precise dose reduction was not quantified in this study, the qualitative reduction in occupational exposure represents a significant improvement in radiation safety.

6.5 Improvement in measurement efficiency

As discussed in Subsection 1.6.5, conventional measurement procedures at KUR typically require a waiting time of about 15 minutes after each irradiation, resulting in more than one hour of lost experimental time per session. The remote operation system introduced in this study eliminated the need for post-irradiation room entry, enabling continuous and efficient measurements. This improvement not only increased experimental efficiency but also enhanced measurement consistency by reducing interruptions and setup variability.

In addition, the LiCAF detector itself does not rely on remote placement, but its sensitivity can be adjusted by selecting an appropriate crystal size according to the neutron intensity at the measurement location. This flexibility enables shorter measurement times while maintaining sufficient counting statistics. Future studies will therefore focus not on remote operation of the detector itself, but on establishing guidelines for selecting LiCAF detectors optimized for specific irradiation fields and measurement conditions.

In conclusion, incorporating the multilayer moderator concept with the Backus–Gilbert optimization method and remote operation into neutron spectrometer development enables accurate and rapid evaluation of neutron energy spectra in BNCT irradiation field compared to conventional evaluation method. It is expected to make a significant contribution to BNCT in terms of efficiency, safety, and flexibility as a practical tool for routine QA.

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Appendix A Cancer and radiation therapy

A.1 Cancer

Figure A.1 shows the long-term trends in mortality rates for major causes of death in Japan from 1960 to 2022. Until the middle of 20th century, infectious diseases such as pneumonia, tuberculosis, were the leading causes of death. However, after World War II, these infectious diseases sharply declined, and lifestyle-related diseases — particularly malignant neoplasms (cancer), heart disease, and cerebrovascular disease — became the dominant causes of mortality. Since 1981, cancer has consistently been the top cause of death. In 2022, there were 385,797 cancer deaths, with a mortality rate of 316.1 per 100,000 population, accounting for about 24.6% of all deaths.

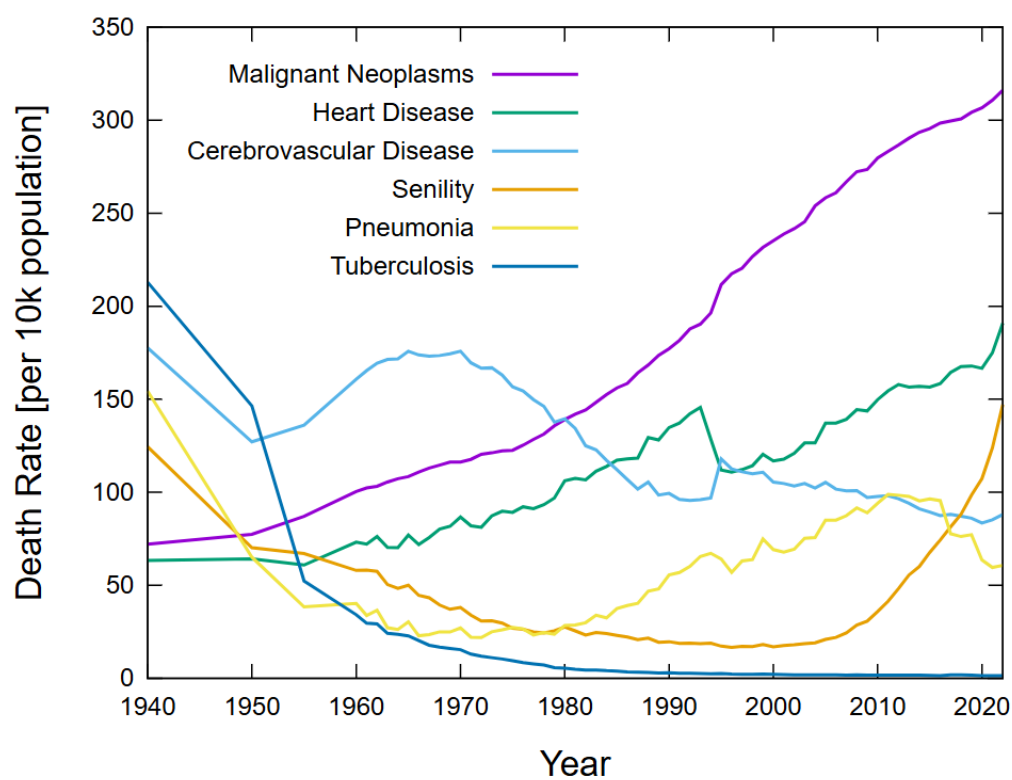


Figure A.1: Trends in crude mortality rate for leading causes of death (1940-2022) [60]

In 2022, the total number of cancer deaths in Japan reached approximately 380,000, with male deaths about 1.4 times higher than female deaths. Table A.1 shows the top five cancer sites by number of deaths in 2022. Among males, the leading cancer site was lung (24.1% of male cancer deaths), while among females, the most common was colon/rectum (15.4% of female cancer deaths). When combining both genders, lung cancer ranked first overall.

Table A.2, on the other hand, presents the number of cancer incidence cases by site in Japan. Notably, the most recent available update for incidence data is from 2019, based on the National Cancer Registry system. The total number of incidence cases was approximately 1,000,000, with male incidence about 1.3 times higher than female incidence. For males, the most frequently

diagnosed cancer was prostate cancer (16.7% of male incidence cases), whereas for females, breast cancer ranked first (22.5% of female incidence cases). When combining both sexes, colon/rectum cancer ranked first overall.

Table A.1: Number of Deaths, by Cancer Site (2022) [60]

	1 st	2 nd	3 rd	4 th	5 th
Male	Lung	Colon/Rectum	Stomach	Pancreas	Liver
Female	Colon/Rectum	Lung	Pancreas	Breast	Stomach
Total	Lung	Colon/Rectum	Stomach	Pancreas	Liver

Table A.2: Number of Incidence, by Cancer Site (2019) [60]

	1 st	2 nd	3 rd	4 th	5 th
Male	Prostate	Colon/Rectum	Stomach	Lung	Liver
Female	Breast	Colon/Rectum	Lung	Stomach	Uterus
Total	Colon/Rectum	Lung	Stomach	Breast	Prostate

Over the past century, advances in medical science have led to the development of various strategies to fight cancer. Cancer treatment is generally categorized into three main pillars:

- Surgical therapy, which removes the tumor itself from the body using instruments such as scalpels
- Chemotherapy, which delivers anticancer agents intravenously or orally to suppress and destroy tumor cell growth throughout the body
- Radiation therapy, which uses the biological effects caused by the energy absorbed from irradiated radiation within the body.

In addition to these, other approaches such as hyperthermia therapy and immunotherapy are also employed.

A.2 Radiation therapy

In this research, the primary focus will be on radiation therapy, which can be subdivided into three main methods.

- External beam radiation therapy

This method delivers radiation from outside the body to target tumors internally. The most commonly used device is the linear accelerator, which generates high-energy X-rays or electron beams. In recent years, particle therapy such as proton therapy, heavy ion therapy and boron neutron capture therapy (BNCT) have emerged, providing superior cancer dose concentration and biological effectiveness.

High-precision techniques — including intensity-modulated radiation therapy (IMRT), stereotactic

body radiation therapy (SBRT), image-guided radiation therapy (IGRT), and motion tracking systems — allow for accurate dose delivery while minimizing damage to surrounding healthy tissues.

- Brachytherapy

This method involves placing sealed radioactive sources directly inside or near the tumor. The delivered dose is determined by the source's position and dwell time. There are two main types of brachytherapy, intracavitary and interstitial brachytherapy. Intracavitary brachytherapy is used for tumors located within body cavities, such as cervical, esophageal, or rectal cancers, where the radioactive sources are positioned inside the cavity. In contrast, interstitial brachytherapy is applied to solid tumors, including prostate cancer, tongue cancer, or breast cancer, where applicators are inserted directly into the tissue to deliver the radioactive sources.

- Nuclear medicine

This approach uses non-sealed radioactive isotopes, administered orally or intravenously, which selectively accumulate in specific tumors or organs. It combines both local and systemic treatment effects and is commonly employed for treating thyroid cancer and multiple bone metastases.

Appendix B Boron compound

Figure B.1(a) and (b) show the chemical structures of Mercapto-undecahydro-closo-dodecaborate (BSH) and 4-borono-L-phenylalanine (BPA), respectively, which are mainly used in clinical BNCT.

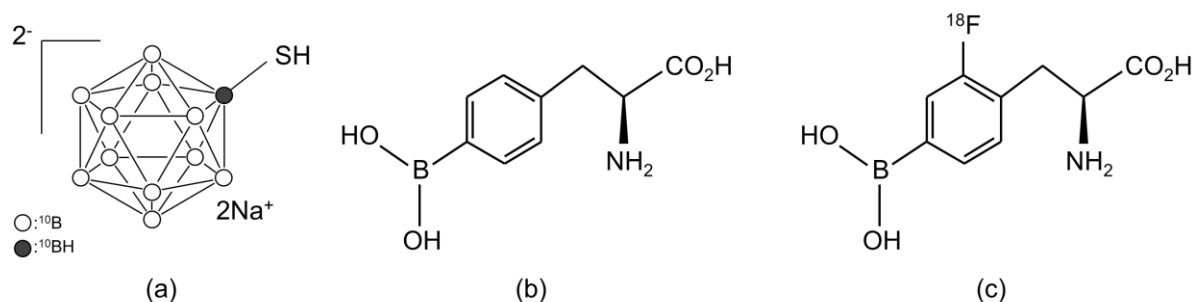


Figure B.1: Boron agents and diagnostic compound used in BNCT

(a) BSH, (b) BPA, (c) ¹⁸F-FBPA

BSH is a boron compound containing a high number of boron atoms (12 per molecule) but exhibits low tumor selectivity. It is primarily used for brain tumors because it can passively accumulate in tumor tissues, even without specific cellular targeting. Normally, BSH cannot effectively cross the blood–brain barrier (BBB). However, it mainly reaches tumors when the BBB has been compromised or disrupted in case of glioblastomas.

BPA, on the other hand, is a boronated amino acid derivative designed for selective tumor uptake, particularly in cancers with elevated amino acid transport activity, such as melanomas and head-and-neck cancers. Notably, BPA can cross the BBB via active amino acid transport mechanisms, making it a promising agent for targeting certain brain tumors. Its tumor-to-normal tissue boron concentration ratios typically range between 3 and 4, providing greater selectivity compared to BSH.

¹⁸F-FBPA (4-borono-2-[¹⁸F]fluoro-L-phenylalanine) is a fluorine-18 labeled derivative of BPA, developed for Positron Emission Tomography (PET) imaging to non-invasively assess the distribution and accumulation of BPA in the body before BNCT. It allows to estimate tumor ¹⁰B concentration and predict each patient tumor-to-normal tissue uptake ratio. ¹⁸F-FBPA helps ensure that sufficient boron delivery to the tumor is achieved for effective BNCT.

Appendix C Neutron source

In 1932, Chadwick and colleagues discovered the neutron [61], and in 1936, Locher proposed a cancer treatment method using neutron-induced nuclear reactions [62]. To make this treatment feasible, the neutron source is just as important as the boron compound. Over time, neutron sources have been developed across four generations: the first generation in the United States, the second generation in Japan, the third generation using epithermal neutrons, and the fourth generation employing accelerator-based systems [63]. Current status of reactor-based and accelerator-based BNCT facility is describe in Table C.1 and Table C.2 respectively.

The first generation neutron source for BNCT is around 1950s, when the world's first BNCT treatment was conducted at the Brookhaven Graphite Research Reactor (BGRR) in 1951, followed by treatments at the Massachusetts Institute of Technology Reactor (MITR) starting in 1959. During this period, thermal neutron irradiation was applied at both facilities; however, by 1961, the United States suspended BNCT treatments due to poor clinical outcomes related to issues with ^{10}B compounds and neutron beam quality. Note that, later in 1994, BNCT treatments were resumed at both facilities, using epithermal neutrons. The Brookhaven National Laboratory program was terminated in 1999 due to reactor decommissioning, while activities at Massachusetts Institute of Technology have been on hold since 2003.

The second generation neutron source for BNCT is between began in 1980s and continued into around 1990s, focusing entirely on thermal neutron irradiation. However, in 1968, Hatanaka initiated BNCT treatments for malignant brain tumors using BSH boron compound at the Hitachi Training Reactor (HTR), achieving improved clinical outcomes compared to the first generation result. BNCT treatments in Japan were conducted at several reactors, including Musashi Institute of Technology (MuITR), Japan Research Reactor 2 (JRR-2), Japan Research Reactor 3 (JRR-3), Japan Research Reactor-4 (JRR-4), and Kyoto University Reactor (KUR). Across these facilities, BNCT patient were treated, often under craniotomy to improve dose delivery to deep-seated tumors.

The development of third-generation neutron sources for BNCT began in 1980s and continued into around 2000s, introduced the use of epithermal neutrons, which can penetrate deeper into the body and eliminate the need for craniotomy. In Japan, KUR and JRR-4 were upgraded to enable epithermal neutron irradiation for clinical trials. In Europe, clinical BNCT trials were launched in the Netherlands, Finland, the Czech Republic, Sweden, and Italy. Argentina began clinical treatments using a combination of thermal and epithermal neutrons for melanoma. Taiwan also started BNCT for head and neck tumors using epithermal neutrons, with more than 500 patients treated to date. In China, a low-power, compact nuclear reactor suitable for installation within hospitals was proposed.

The fourth generation neutron source for BNCT began around 2000s and continue to the present, Because accelerators are easier to install in hospitals compared to reactors, they have gained widespread attention. Following the success of C-BENS, the NeuCure system was implemented at South Tohoku BNCT Center and Kansai BNCT Center, and BNCT became an insurance-covered

treatment for unresectable, locally advanced, and recurrent head and neck carcinomas in Japan starting in 2020 [64]. Recently, not only in Japan, numerous accelerator-based BNCT projects have emerged worldwide.

Table C.1: Current status of reactor based BNCT which perform clinical treatment [65], [66], [67]

Reactor/Institution/Location	Reactor Power (MW)	Neutron Mode	Clinical Trial number	Period
AMERICA				
BGRR, Brookhaven National Laboratory, USA	28	Thermal	28	1951-1958
BMRR, Brookhaven National Laboratory, USA	5	Thermal	17	1959-1961
BMRR-M, Brookhaven National Laboratory, USA	5	Epithermal	54	1994-1999
MITR, Massachusetts Institute of Technology, USA	3	Thermal	18	1953-1961
MITR, Massachusetts Institute of Technology, USA	3	Epithermal	24	1994-1999
MITR, Massachusetts Institute of Technology, USA	3	Epithermal	6	2002-2003
ASIA				
HTR, Hitachi, Japan	0.1	Thermal	13	1968-1974
MuITR, Musashi Institute of Technology, Japan	0.1	Thermal	108	1977-1989
JRR-3, Japan Atomic Energy Research Institute, Japan	10	Thermal	1	1969
JRR-2, Japan Atomic Energy Research Institute, Japan	10	Thermal	13	1990-1996
JRR-4, Japan Atomic Energy Research Institute, Japan	3.5	Thermal	2	2004-2010
		Mixed	16	1999-2010
		Epithermal	87	2003-2010
KUR, Kyoto University Research Reactor Institute, Japan	5	Thermal	16	1996-2019
		Mixed	17	1997-2002
		Epithermal	490	2001-2020
THOR, National Tsing Hua University, Taiwan	2	Epithermal	500+	2010-
IHNI-1, Beijing Nuclear Industry Hospital, China	0.03	Epithermal	3+	2014-
		Thermal		

EUROPE				
HFR-Petten, Netherlands Energy Research Foundation, Netherlands	45	Epithermal	30	1997-
FiR-1, Technical Research Centre of Finland, Finland	0.25	Epithermal	314	1997-2011
LVR-15, Nuclear Research Institute Rez, Czech	10	Epithermal	2	2000
R2-0, Studsvik, Sweden	1	Epithermal	52	2001-2005
TrigaMark II reactor Pavia, Applied Nuclear Energy Laboratory of the University of Pavia, Italy	0.25	Thermal	2	2001-
SOUTH AMERICA				
RA-6, Bariloche Atomic Center, Argentina	0.5	Thermal	9+	2003-

Table C.2: Current status of accelerator based BNCT [5], [68]

Project/Institution/Location	Accelerator type	Target reaction	Beam energy (MeV)	Current goal (mA)	Final power (kW)	Current Status	Ref
ASIA							
C-BENS, Kyoto University Research Reactor Institute, Japan	Cyclotron	${}^9\text{Be}(p,n)$	30	1	30	Experimental Usage	[69]
NeuCure, Southern Tohoku BNCT Research Center, Japan	Cyclotron	${}^9\text{Be}(p,n)$	30	1	30	Clinical Treatment	[70]
NeuCure, Kansai BNCT Medical Center, Japan	Cyclotron	${}^9\text{Be}(p,n)$	30	1	30	Clinical Treatment	[71]
CICS-1, National Cancer Center Hospital, Japan	RFQ	Solid ${}^7\text{Li}(p,n)$	2.8	20	50	Clinical Trial	[72]
CICS-2, Edogawa Hospital, Japan	RFQ	Solid ${}^7\text{Li}(p,n)$	2.8	20	50	Clinical Trial	[73]
iBNCT, Ibaraki Neutron Medical Research Center, Japan	RFQ+DTL	${}^9\text{Be}(p,n)$	8	10	80	Clinical Trial	[74]
NUANS, Nagoya University, Japan	Dynamitron	Solid ${}^7\text{Li}(p,n)$	2.8	15	42	Experimental Usage	[75]
nuBeam, Shonan Kamakura General Hospital, Japan	Electrostatic	Solid ${}^7\text{Li}(p,n)$	2.6	30	78	Commissioning	[76]
NeuBoron, Xiamen Humanity Hospital, China	Electrostatic	Solid ${}^7\text{Li}(p,n)$	2.5	10	25	Clinical Trial	[77]
D-BNCT01, Dongguan Neutron Science Center, China	RFQ	Solid ${}^7\text{Li}(p,n)$	3.5	5	17.5	Experimental Usage	[78]
D-BNCT02, Dongguan People's Hospital, China	RFQ	Solid ${}^7\text{Li}(p,n)$	2.8	20	56	Clinical Trial	[79]
CIAE AB-BNCT, China Institute of Atomic Energy, China Tai'an Central Hospital, Tai'an, China	Cyclotron	${}^9\text{Be}(p,n)$	14	1	14	Experimental Usage	[80]
LU AB-BNCT, Fujian Medical University Union Hospital, China	RFQ	Solid ${}^7\text{Li}(p,n)$	2.6	30	78	Commissioning	[81]
X-TANS, Xi'an Jiaotong University, China	RFQ	Solid ${}^7\text{Li}(p,n)$	2.5	10	25	Under Development	[82]
NeuCure, Boao BNCT Center, China	Cyclotron	${}^9\text{Be}(p,n)$	30	1	30	Under Construction	[83]
Heron, China Medical University Hsinchu Hospital, Taiwan	Cyclotron	${}^9\text{Be}(p,n)$	30	1	30	Clinical Trial	[84]
Heron, Taipei Veterans General Hospital, Taiwan	Cyclotron	${}^9\text{Be}(p,n)$	30	1	30	Under Development	[85]
A-BNCT, Dawon Medax, South Korea	RFQ-DTL	${}^9\text{Be}(p,n)$	10	8	80	Clinical Trial	[11]

KIRAMS AB-BNCT, KIRAMS, South Korea	Electrostatic	$^9\text{Be}(p,n)$	2.4	10	24	Under Construction	[86]
SARAF-LiLiT, Soreq Nuclear Research Center, Israel	RFQ-DTL	Liquid $^7\text{Li}(p,n)$	2.5	20	50	Under Development	[87]
EUROPE							
VITA, Budker Institute, Russia	Electrostatic	Solid $^7\text{Li}(p,n)$	2.0-2.3	10	23	Experimental Usage	[88]
VITA, Blokhin Cancer Center, Russia	Electrostatic	Solid $^7\text{Li}(p,n)$	2.3	7	20	Under Construction	[89]
nuBeam, Helsinki University Hospital, Finland	Electrostatic	Solid $^7\text{Li}(p,n)$	2.6	30	78	Clinical Trial	[90]
nuBeam, Birmingham University, UK	Electrostatic	Solid $^7\text{Li}(p,n)$	2.6	30	78	Experimental Usage	[91]
NeMeSis, Granada University Hospital, Spain	Electrostatic	Solid $^7\text{Li}(p,n)$	2.1	30	6.3	Under Development	[92]
Alphabeam, CNAO, Italy	Electrostatic	Solid $^7\text{Li}(p,n)$	2.5	10	25	Under Construction	[93]
Legnaro-RFQ, Legnaro National Laboratory, Italy	RFQ	$^9\text{Be}(p,n)$	5	30	150	Under Construction	[94]
E_LibANS, University of Torino, Italy	Electron LINAC	$\text{Pb}(\gamma,n)$	18	?	?	Experimental Usage	[95]
SOUTH AMERICA							
ESQ, CNEA, Argentina	ESQ	$^9\text{Be}(d,n)$, $^{13}\text{C}(d,n)$	1.45	30	43	Under Development	[96]

Appendix D Detailed structure of Heavy Water Neutron Irradiation Facility

Figure D.1(a) shows an external photograph and a cross-sectional view of Heavy Water Neutron Irradiation Facility installed at Kyoto University Reactor (KUR-HWNIF). The essential structural components of the KUR-HWNIF relevant to BNCT showing in Figure D.1(b) are described below.

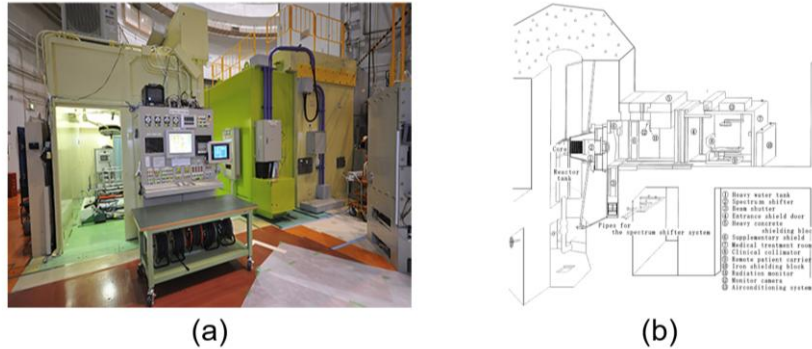


Figure D.1: Heavy Water Neutron Irradiation Facility (HWNIF)

(a) photo outside HWNIF with control panel, (b) vertical cross-sectional view [35]

- Heavy water tank

The heavy water tank is installed close to the reactor core and contains a moderator designed for producing epithermal neutrons. This moderator is composed of a mixture of aluminum and heavy water (80% aluminum and 20% heavy water by volume). This combination effectively removes fast neutrons while allowing epithermal neutrons with energies around tens of keV to pass through, optimizing the neutron energy spectrum for BNCT.

- Spectrum shifter

The spectrum shifter consists of three separate tanks, 10 cm, 20 cm, and 30 cm thick, arranged sequentially starting from the reactor core side. Heavy water in each tank can be independently supplied or drained, allowing the total heavy-water thickness to be adjusted in increments of 10 cm, ranging from 0 cm up to 90 cm. By changing the heavy water thickness, the neutron energy spectrum can be precisely controlled. Next to the spectrum shifter, a 30.5 cm thick heavy water shutter is installed, functioning as a part of the radiation shielding system.

- Thermal neutron filter and bismuth gamma shielding

Two types of thermal neutron filters are installed outside the heavy water shutter. These filters can be exchanged and serve to control the thermal neutron component in the beam.

By combining the presence or absence of heavy water in each layer of the spectrum shifter and heavy water shutter, along with the open or closed states of the two cadmium filters, up to 64 different irradiation modes can be configured. The bismuth gamma shielding, located next to the thermal neutron filters, has a thickness of 23.4 cm and is designed to block gamma rays produced from the reactor core.

Appendix E High purity germanium (HPGe) detector efficiency

The efficiency calibration was carried out using a mixed radionuclide standard source (Isotrack, BE-8302), which contains twelve encapsulated radioisotopes.

Among the twelve radioisotopes included in the calibration source, only six were detectable under the present measurement conditions. Measurement result is shown in Table E.1. In this study, γ -ray energies below 300 keV were not considered; therefore, the efficiency calibration was performed using the γ -ray peaks emitted from ^{137}Cs , and ^{60}Co (both 1.173 MeV and 1.332 MeV).

Table E.1: Measurement result of six detectable radioisotopes

Nuclide	Energy [keV]	Half life	Emission Rate [s^{-1}]		Net rate [cps]	Efficiency [%]
			ref date	meas. date		
Am-241	59.5	432.6 y	1.23×10^3	1.22×10^3	2.63×10^1	2.15
Cd-109	88	461.9 d	6.12×10^2	4.83×10^1	4.48×10^0	9.28
Co-57	122.1	271.74 d	5.40×10^2	7.20×10^0	2.14×10^0	24.88
Cs-137	661.7	30.08 y	2.35×10^3	2.11×10^3	7.60×10^1	3.60
Co-60(1)	1173.2	1925.28 d	3.29×10^3	1.79×10^3	3.31×10^1	1.85
Co-60(2)	1332.5		3.30×10^3	1.79×10^3	2.94×10^1	1.64

Pulse-height spectra were measured for the calibration source, and the detection efficiency was evaluated for each identified γ -ray peak. The efficiency curve was determined by fitting the experimental data using the fitting function given in Equation (E.1)(E.1).

$$\log_{10}(\text{Efficiency}[\%]) = A \times \log_{10}(E[\text{keV}]) + B \quad (\text{E.1})$$

The fitting was performed, and the results are shown in Table E.2 and Figure E.1

Table E.2: Fitting result of equation (E.1)

	value	error
A	-1.137	0.031
B	3.762	0.093
R^2	0.99	

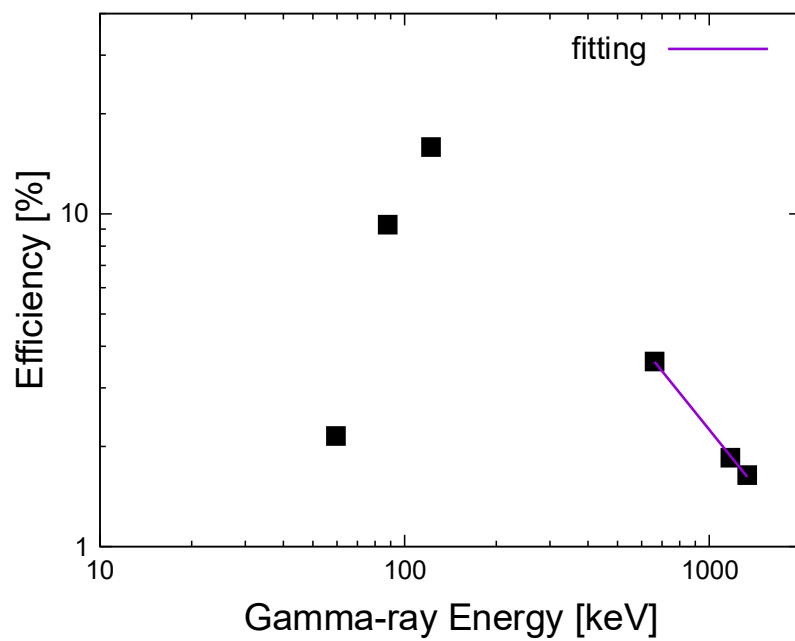


Figure E.1: Detection efficiency as a function of γ -ray energy with fitting line

Appendix F LiCAF detector fitting result

The Gaussian fitting results of the pulse-height spectra are summarized for three different measurement times, both with and without the collimator. The fitting was performed using the Solver function in Microsoft Excel, where the region of interest (ROI) defined from the mean value μ to $\mu + \sigma$ in order to focus on neutron-induced events and suppress gamma-ray contributions.

For all measurement times, a clear Gaussian peak corresponding to neutron events was observed, and stable fitting results were obtained under both irradiation conditions. Measurements conducted without the collimator exhibited higher count rates due to the broader neutron beam and increased neutron intensity, whereas measurements with the collimator showed reduced count rates, reflecting beam shaping and intensity attenuation by the collimation system.

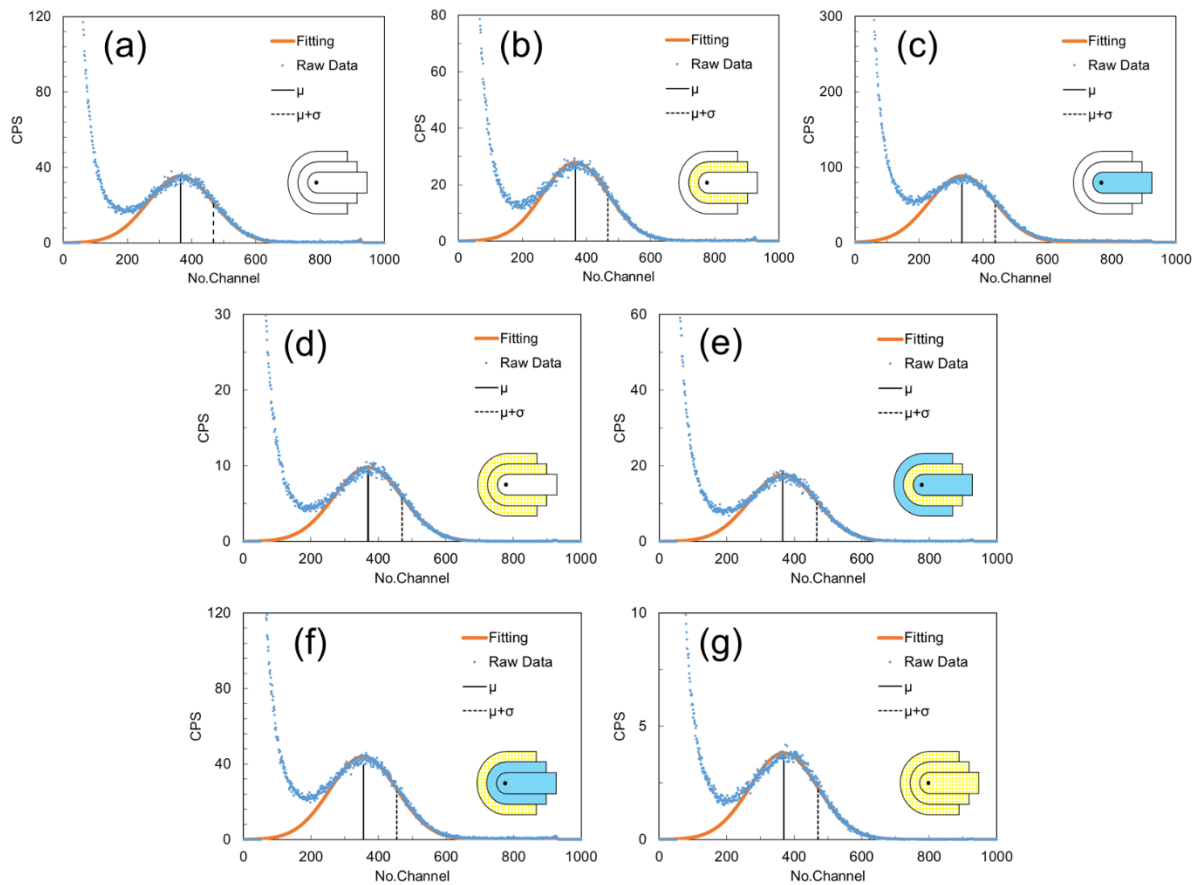
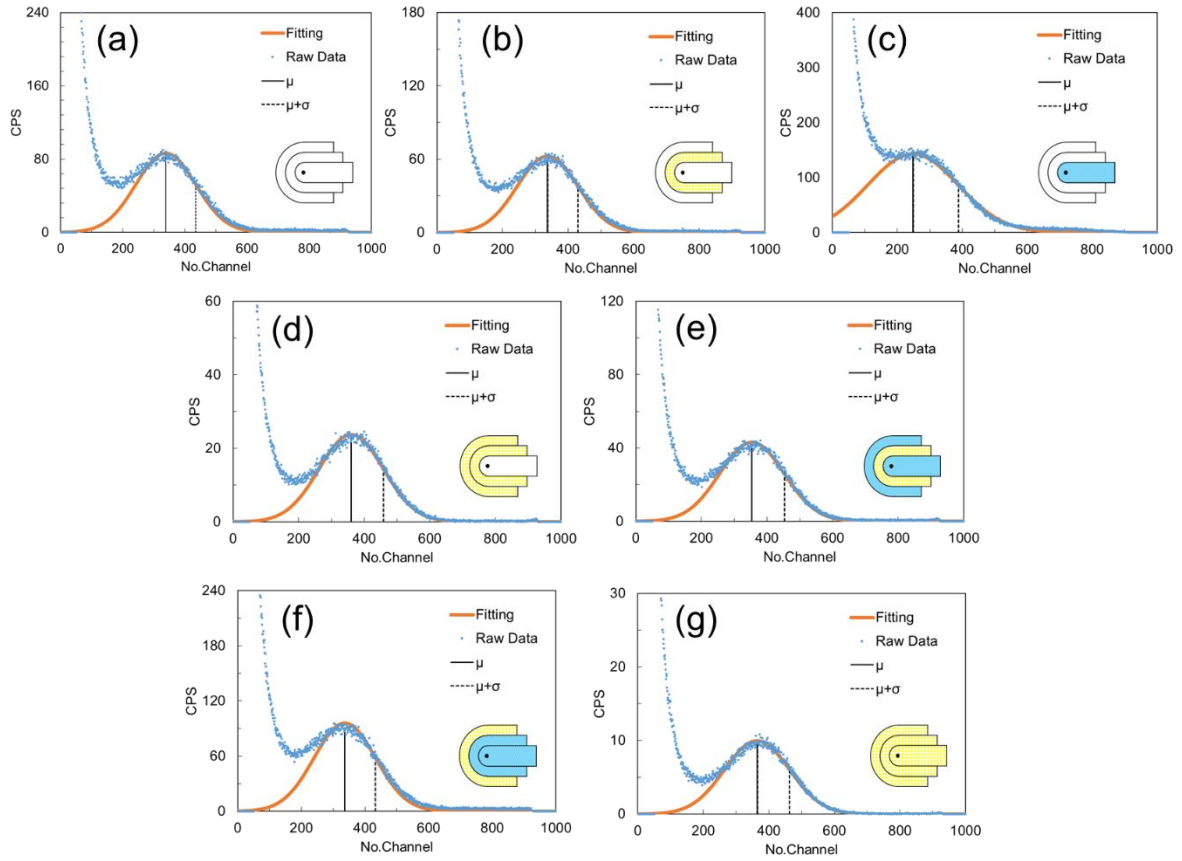


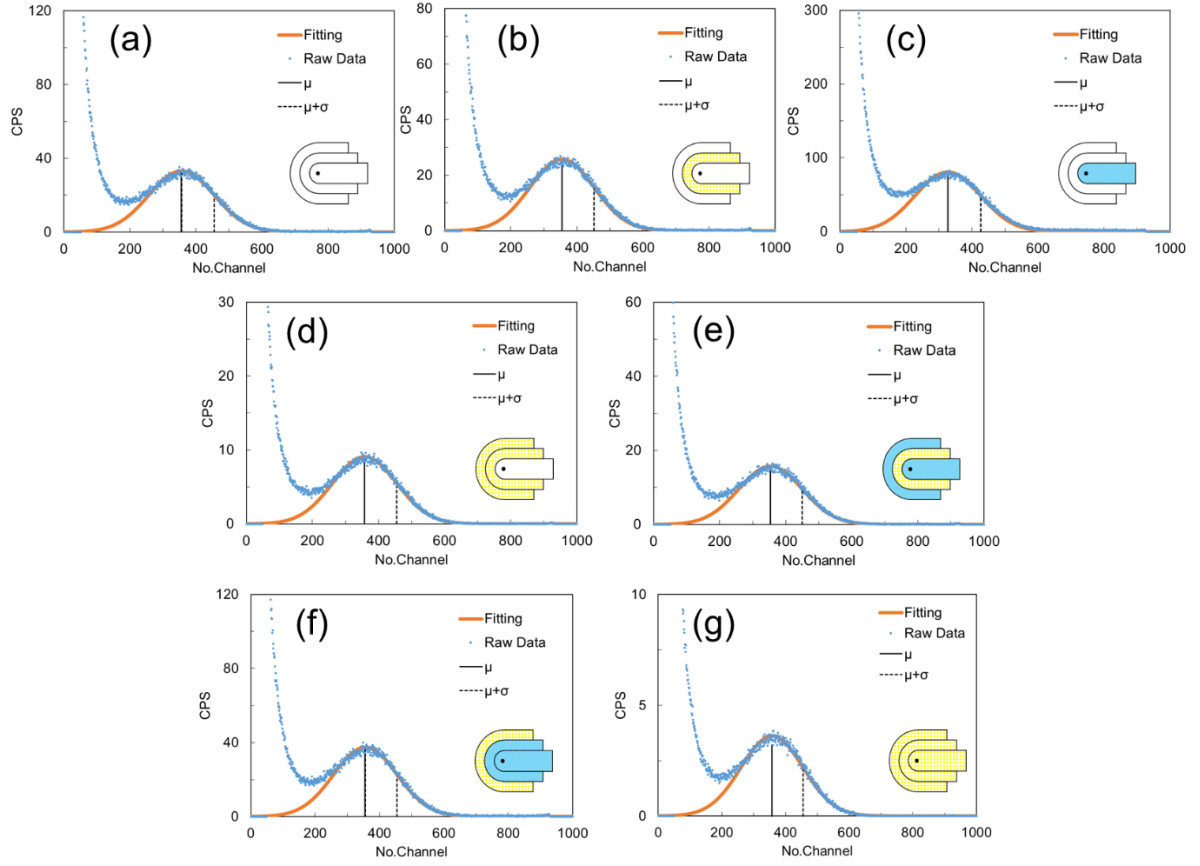
Figure F.1: LiCAF detector fitting results from the first measurement with collimator irradiation setup

(a)“aaa”, (b)“aba”, (c) “waa”, (d)“abb”, (e)“wbw”, (f)“wwb”, (g)“bbb”



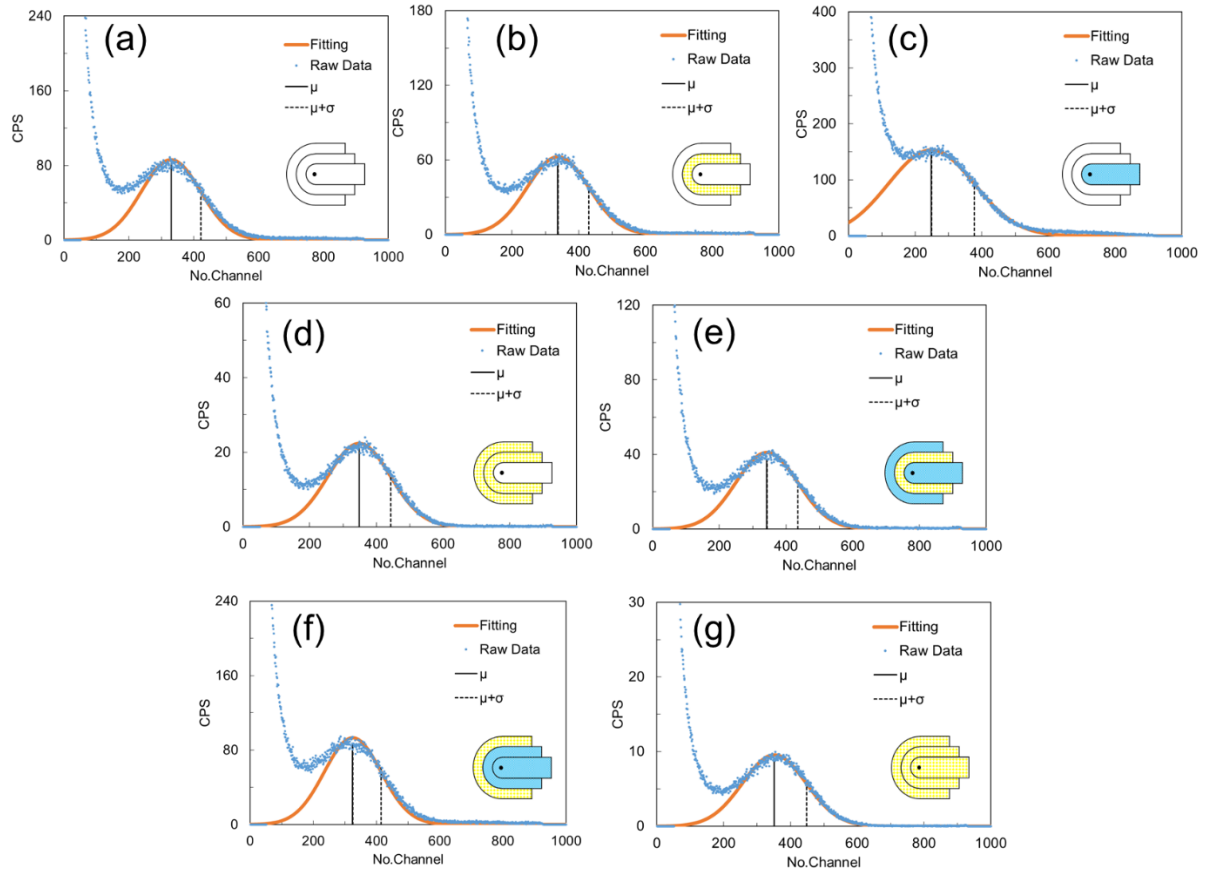
**Figure F.2: LiCAF detector fitting results from the first measurement
without collimator irradiation setup**

(a)“aaa”, (b)“aba”, (c) “waa”, (d)“abb”, (e)“wbw”, (f)“wwb”, (g)“bbb”



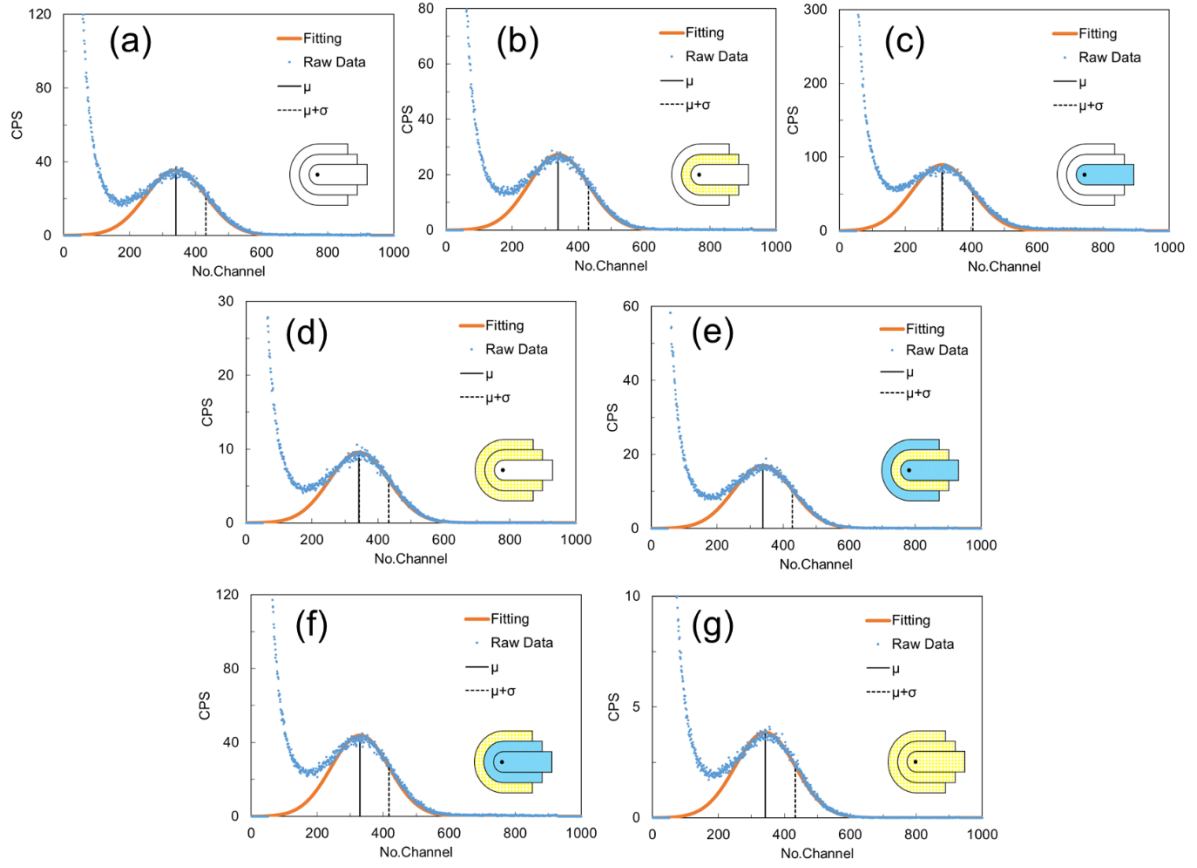
**Figure F.3: LiCAF detector fitting results from the second measurement
with collimator irradiation setup**

(a)“aaa”, (b)“aba”, (c) “waa”, (d)“abb”, (e)“wbw”, (f)“wwb”, (g)“bbb”



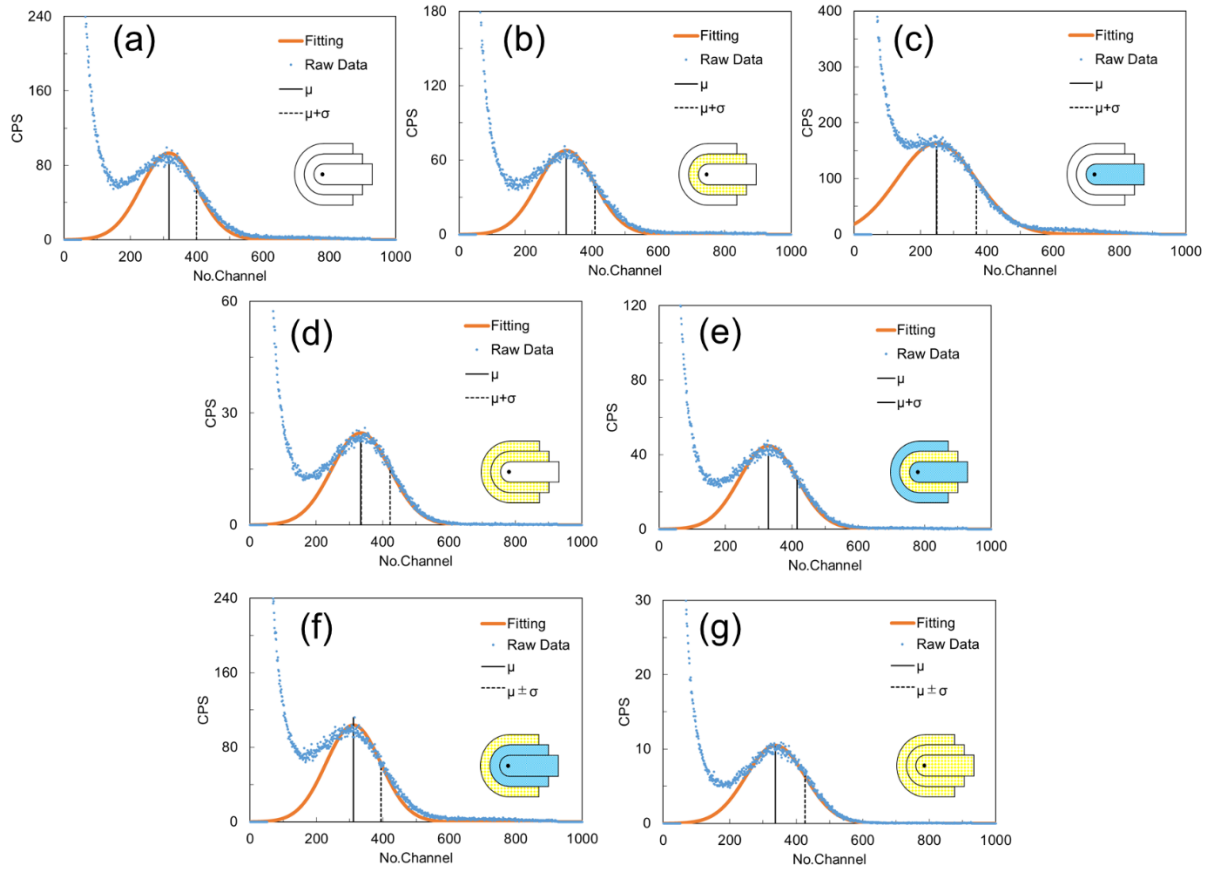
**Figure F.4: LiCAF detector fitting results from the second measurement
without collimator irradiation setup**

(a)“aaa”, (b)“aba”, (c) “waa”, (d)“abb”, (e)“wbw”, (f)“wwb”, (g)“bbb”



**Figure F.5: LiCAF detector fitting results from the third measurement
with collimator irradiation setup**

(a) "aaa", (b) "aba", (c) "waa", (d) "abb", (e) "wbw", (f) "wwb", (g) "bbb"



**Figure F.6: LiCAF detector fitting results from the third measurement
without collimator irradiation setup**

(a) "aaa", (b) "aba", (c) "waa", (d) "abb", (e) "wbw", (f) "wwb", (g) "bbb"

Publication

1. J. Prateepkaew and Y. Sakurai, "Reevaluation of neutron energy spectrum in Heavy-Water neutron irradiation facility of Kyoto University research Reactor using multifoil activation method," *Nucl Instrum Methods Phys Res B*, vol. 557, Dec. 2024, doi: 10.1016/j.nimb.2024.165555. (Chapter 2)
2. J. Prateepkaew, N. Matsubayashi, T. Takata, H. Tanaka, and Y. Sakurai, "Feasibility of multilayer neutron spectrometer with Backus and Gilbert approach using moderator-combination selection for energy-resolution optimization in Boron Neutron Capture Therapy irradiation field," *Nucl Instrum Methods Phys Res A*, vol. 1059, Feb. 2024, doi: 10.1016/j.nima.2023.168948. (Chapter 3)
3. J. Prateepkaew, T. Takata, H. Tanaka, and Y. Sakurai, "Experimental verification of liquid multilayer spectrometer in neutron irradiation field for boron neutron capture therapy," *Nucl Instrum Methods Phys Res A*, vol. 1065, Aug. 2024, doi: 10.1016/j.nima.2024.169534. (Chapter 4)
4. J. Prateepkaew, N. Matsubayashi, T. Takata, H. Tanaka, and Y. Sakurai, "Quality assurance validation of remote neutron spectrometer for boron neutron capture therapy," *Med Phys*, vol. 52, no. 8, Aug. 2025, doi: 10.1002/mp.18029. (Chapter 5)
5. J. Prateepkaew, N. Matsubayashi, T. Takata, H. Tanaka, and Y. Sakurai, "Characteristic comparison between two different neutron spectrometers for boron neutron capture therapy irradiation field developed at Heavy Water Neutron Irradiation Facility of Kyoto University Research Reactor," *Applied Radiation and Isotopes*, vol. 219, May 2025, doi: 10.1016/j.apradiso.2025.111717. (Chapter 4, Chapter 5)