

## NEUTRON RESPONSE FUNCTION OF A BONNER SPHERE SPECTROMETER WITH $^6\text{LiI}(\text{Eu})$ DETECTOR

### FUNGSI RESPON NEUTRON PADA SPEKTROMETER BOLA BONNER DENGAN DETEKTOR $^6\text{LiI}(\text{Eu})$

R. Tursinah<sup>1</sup>, Bunawas<sup>1</sup>, J. Kim<sup>2</sup>

<sup>1</sup>National Nuclear Energy Agency (BATAN), Jl. Tamansari No.71, Bandung, Indonesia

<sup>2</sup>Korean Research Institute of Standards and Science (KRISS), Daejeon 305-340, Rep. of Korea

E-mail: [rasito@batan.go.id](mailto:rasito@batan.go.id)

Received 21 March 2017, received in revised form 23 May 2017, accepted 15 August 2017

#### ABSTRACT

##### NEUTRON RESPONSE FUNCTION OF BONNER SPHERE SPECTROMETER WITH $^6\text{LiI}(\text{Eu})$ DETECTOR.

The neutron response function of  $^6\text{LiI}(\text{Eu})$  detector is needed to measure neutron fluence based on the count rates from a Bonner Sphere Spectrometer (BSS). The determination of response function of a BSS with  $^6\text{LiI}(\text{Eu})$  neutron detector has been performed using Monte Carlo MCNPX code. This calculation was performed for a BSS using scintillation detector of 4 mm × 4 mm  $^6\text{LiI}(\text{Eu})$  which was placed at the center of a set of polyethylene spheres i.e bare, 2", 3", 5", 8", 10", and 12" diameters. The BSS response functions were obtained for neutron energy range of  $1 \times 10^{-9}$  MeV -  $1 \times 10^2$  MeV in 111 energy bins and each value had an uncertainty of less than or equal to 2 %. The calculation results were compared with two response functions reported in a IAEA document of Technical Reports Series 403 (TRS-403) and calculated by Vega-Carrillo, et al. and also validated by direct measurement of  $^{252}\text{Cf}$  neutron spectra. The result show that the calculated BSS spectra are quite close to the measured spectra with a difference of 3%.

**Keywords:** Neutron response function, BSS,  $^6\text{LiI}(\text{Eu})$  detector, MCNPX

#### ABSTRAK

##### FUNGSI RESPON NEUTRON PADA SPEKTROMETER BOLA BONNER DENGAN DETEKTOR $^6\text{LiI}(\text{Eu})$ .

Fungsi respon neutron detektor  $^6\text{LiI}(\text{Eu})$  sangat diperlukan untuk menghitung fluks neutron berdasarkan nilai laju cacahnya pada Bonner Sphere Spectrometer (BSS). Telah dilakukan penentuan fungsi respon pada BSS detektor  $^6\text{LiI}(\text{Eu})$  menggunakan program Monte Carlo MCNPX. Perhitungan ini dilakukan untuk BSS dengan detektor sintilasi  $^6\text{LiI}(\text{Eu})$  ukuran 4 x 4 mm yang ditempatkan pada pusat masing-masing bola polietilen dengan diameter 0", 2", 3", 5", 8", 10", dan 12". Telah diperoleh fungsi respon BSS untuk rentang energi  $1 \times 10^{-9}$  -  $1,12 \times 10^2$  MeV yang dibagi dalam 111 energi dengan masing-masing nilai memiliki ketidakpastian di bawah 2%. Nilai fungsi respon yang diperoleh dibandingkan dengan beberapa literatur yaitu dokumen IAEA dalam Technical Reports Series 403 (TRS-403) dan hasil perhitungan oleh Vega-Carrillo. Hasil ini juga divalidasi dengan pengukuran langsung spektrum neutron  $^{252}\text{Cf}$ , yang memperlihatkan bahwa spektrum hasil simulasi mendekati sama dengan hasil pengukuran dengan perbedaan 3%.

**Kata kunci:** fungsi respon neutron, BSS, detektor  $^6\text{LiI}(\text{Eu})$ , MCNPX

#### INTRODUCTION

The Bonner Spheres Spectrometer consists of a thermal neutron detector placed in the centers of moderating polyethylene spheres with different diameters. From the measured readings, information can be derived to the spectrum of the neutron field where measurements were made [1]. A Bonner sphere spectrometer (BSS) is used in radiation protection measurement because of its wide energy range (thermal to tens MeV) and easy operation [2]. A BSS can be used to measure the neutron spectrum with energy up to a few GeV usually by adding lead in the moderating spheres [3],[4],[5]. Several type of thermal neutron detector are utilized in BSS, such as  $^6\text{LiI}(\text{Eu})$  scintillator, pairs of thermoluminescent dosimeters, activation foils, track detectors, and  $\text{BF}_3$  or  $^3\text{He}$  filled proportional gas counters [6],[7],[8],[9]. The neutron response and the measured readings of the BSS are associated with the neutron count spectrum by the Fredholm integral equation in Equation 1

$$C = \int_0^E R(E) \Phi_E(E) dE \quad (1)$$

where inside a neutron field the detector produces count per time,  $C$ , that is related to the response function,  $R(E)$ , and the neutron spectrum,  $\Phi_E(E)$ .

The use of a monoenergetic neutron source for calculating the response function of a BSS is quite expensive and complex; due to the availability of monoenergetic neutron sources and the calculation can be undertaken only for several energy points [10]. This situation causes the neutron response function cannot be measured directly; and as alternative solution a Monte Carlo particle transport tools is usually used to calculate it. Monte Carlo is a statistic numerical method using random numbers to solve problems that cannot be solved by analytical methods. One of computer code that use Monte Carlo method are Monte Carlo N-Particle eXtended (MCNPX). This code was developed by Monte Carlo Team (2008) at the Los Alamos National Laboratory, USA. The aim of this work is to calculate the neutron response function of a BATAN BSS with a  ${}^6\text{LiI}(\text{Eu})$  scintillator by using MCNPX 2.6. The results will be compared with the data available in IAEA TRS-403 (Mares and Schraube) and Vega//Carrillo, et al. [11],[12] and will be validated with the measurement of  ${}^{252}\text{Cf}$  neutron spectra.

## METHODOLOGY

Calculation of response function for BSS with  ${}^6\text{LiI}(\text{Eu})$  detector using MCNPX was undertaken through three steps i.e respectively the preparation of input files, the execution of the code, and the interpretation of the results. The preparation of MCNPX input files require models of detector and BSS geometry, radiation source definition, and fluence tally model.

### Model of Detector

Bonner sphere spectrometer (BSS) used in PTKMR Neutron Laboratory have seven diameters i.e 0" (bare), 2", 3", 5", 8", 10" and 12" as shown in Figure 1. The 4 mm  $\times$  4 mm of  ${}^6\text{LiI}(\text{Eu})$  scintillator is located at the center of BSS that used as thermal neutron detector.



Figure 1. The set of BSS at PTKMR BATAN neutron laboratory

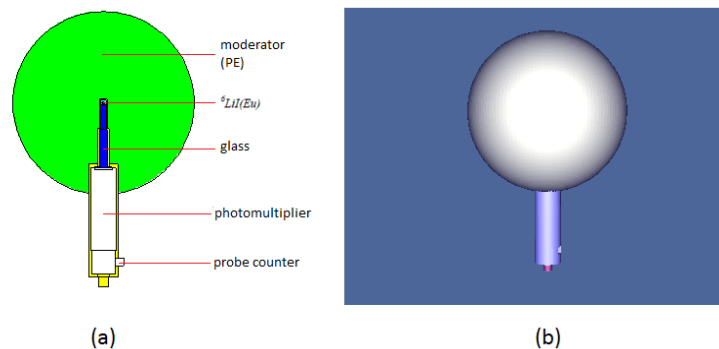


Figure 2. BSS models in MCNPX, (a) in 2D, (b) in 3D

Naturally occurring Lithium isotopes are 92.5%  ${}^6\text{Li}$  and 7.5%  ${}^7\text{Li}$ , while Iodine has only one natural isotope  ${}^{127}\text{I}$ ; all those isotopes are stable. The  ${}^6\text{LiI}(\text{Eu})$  scintillator consist of  ${}^6\text{Li}$ ,  ${}^7\text{Li}$ , and  ${}^{127}\text{I}$  isotopes, with smaller amounts of Eu as impurities [12]. The reported data in literatures are calculations based on scintillators that has been

modeled with different enrichment of  $^6\text{Li}$  of 96% to 100% [12]. In this investigation the scintillator was modeled with enrichment of  $^6\text{Li}$  of 96 %. The BSS models in MCNPX is shown in Figure 2. In general, the neutron capture cross-section for the  $^6\text{Li}$  is 940 barn [13] [14]. The large cross-section makes the detector very sensitive. Moderating spheres were modeled with a density of 939 Kg/m<sup>3</sup> and made of polyethylene. Atomic composition and physical data of different elements used to build the model were obtained from Compendium of Material Composition Data for Radiation Transport Modeling [15].

### Source Definition

In this calculations the scintillator cylindrical body was oriented normally to the source-sphere axis. Each sphere-detector combination was irradiated with a neutron beam produced by a disk-shaped neutron source. Then, a source of monoenergetic neutron was directed towards the polyethylene sphere as shown in Figure 3. Irradiations were carried out using 115 monoenergetic neutron sources with energy range from  $1 \times 10^{-9}$  MeV to  $1 \times 10^2$  MeV for each detector. The calculations were performed with the Monte Carlo code MCNPX version 2.6.0 and LA150 cross section library. The response was defined as the number of  $^6\text{Li}(n,\alpha)$  reactions per incident neutron fluence based on the track length estimation of detector fluence normalized to one starting particle.

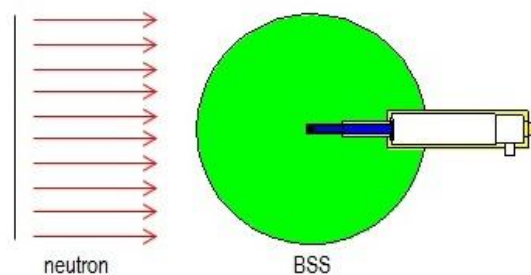


Figure 3. Disk-shape neutron source with normal direction

### Fluence Tally

MCNPX code will produce an output file containing the results of the run. The fluence can be produced by running of MCNPX input using track-length tally (F4) for neutrons (cm<sup>-2</sup>). Tally of F4 can score cell fluence (particles/cm<sup>2</sup>) and statistical errors. The response function can be calculated by multiplying the fluence with a multiplication factor. The multiplication factor with tally multiplier (FM) is one of the most important tally to convert fluence to response function. The FM value are neutron field area (cm<sup>2</sup>) multiplied by atomic density of detector ( $10^{30} \text{ m}^{-3}$ ) multiplied by detector volume (m<sup>3</sup>). The value of response function (m<sup>2</sup>) will be resulted from fluence (m<sup>-2</sup>) multiplied by FM (m<sup>4</sup>). This FM number is  $S_i N V$  that obtained from this equation:

$$R_i(E) = S_i N V \int_0^E \sigma(n, \alpha) \Phi_E(E) dE \quad (2)$$

where  $S_i = \frac{\pi}{4} d_i^2$  (m<sup>2</sup>) is the source surface area considered sphere diameter  $d_i$ ,  $N$  ( $1.74 \times 10^{28} \text{ atoms-m}^{-3}$ ) is the  $^6\text{LiI}(\text{Eu})$  crystal volumetric atom density,  $V$  ( $5 \times 10^{-8} \text{ m}^3$ ) is the crystal volume.

## RESULTS AND DISCUSSION

### Response Function Calculation

MCNPX code with input data of BSS geometry, radiation source, and fluence was runned using a computer at the KRISS Neutron Laboratory. In this work number of particle histories  $1 \times 10^9$  was used. To obtain statistical error of less than 2 %, for 784 input files using cluster computer 64 processors needed running time of two days. The MCNPX output are neutrons fluence on the  $^6\text{LiI}(\text{Eu})$  detectors as a function of initial neutron energy. The fluence values plotted as energy function is shown in Figure 4. In this Figure the calculated response functions are also shown, the 111 bins of energy were obtained with MCNPX, each value has an uncertainty of less or equal to 2 %.

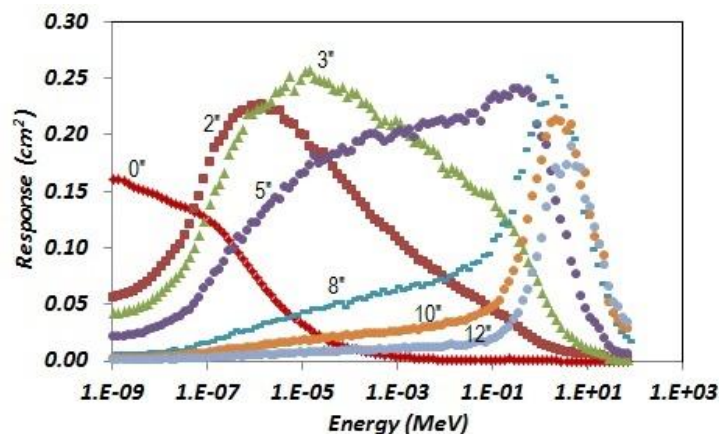


Figure 4. Response function of BSS with  $^6\text{Li}(\text{Eu})$  detectors.

Monte Carlo codes for neutron transport calculations such as MCNPX crucially rely on input data cross sections that describe the interaction of neutrons with nuclei. For neutron energies below 20 MeV, experimental cross sections data are available that are validated against experimental data[16], while for neutron energies above 20 MeV experimental cross section data are scarce, therefore intra-nuclear cascade (INC) and evaporation models are usually applied in these Monte Carlo codes[17]. For this reason, every neutron transport code is based on theoretical nuclear models to describe interactions of neutrons with nuclei in matter [16]. The calculation of BSS response functions using the Monte Carlo code and nuclear models may increase the uncertainty, in particular for neutron energies above 20 MeV[17],[18]. The response function for BSS 0" (bare) is strongly affected by the  $^6\text{Li}$  cross section shape or "no moderation". The response function for BSS of 2", 3" and 5" was affected neutron energy range  $10^{-7}$  MeV -  $10^{-1}$  MeV or epithermal neutrons, than for BSS 8", 10" and 12", response functions of detector is significant for neutron energy range  $10^{-1}$  MeV –  $10^2$  MeV or fast neutrons[12]. It can be noticed that polyethylene spheres increasing the response functions of detectors[12].

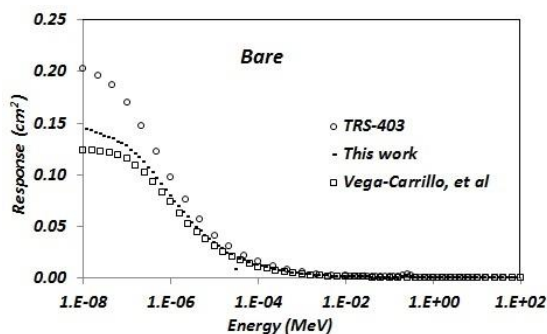


Figure 5. Neutron response function for bare

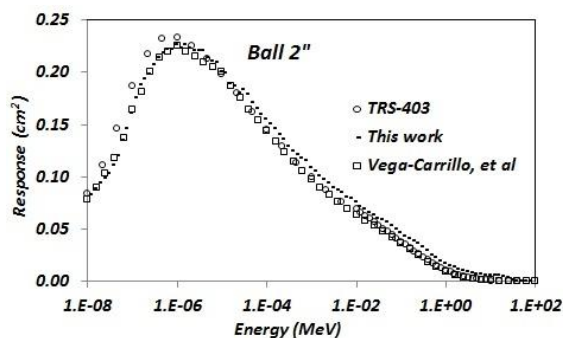


Figure 6. Neutron response function for BSS 2"

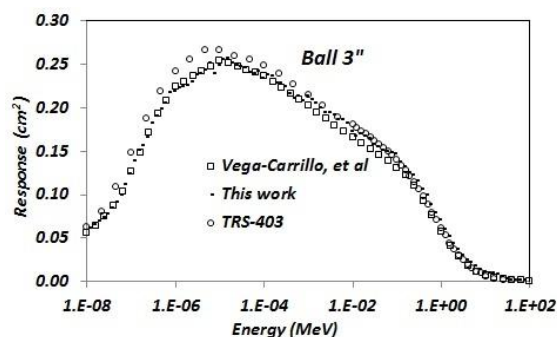


Figure 7. Neutron response function for BSS 3"

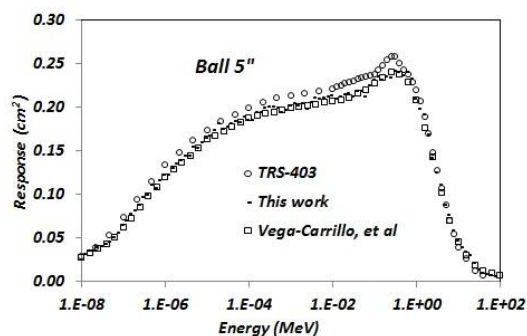


Figure 8. Neutron response function for BSS 5"

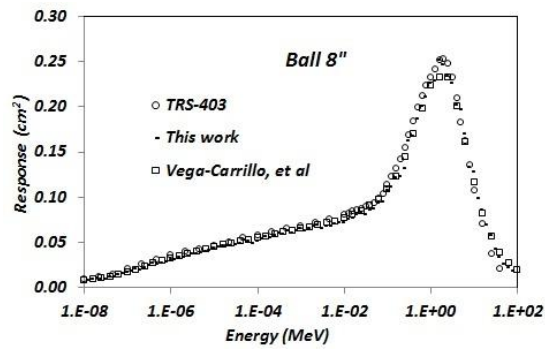


Figure 9. Neutron response function for BSS 8"

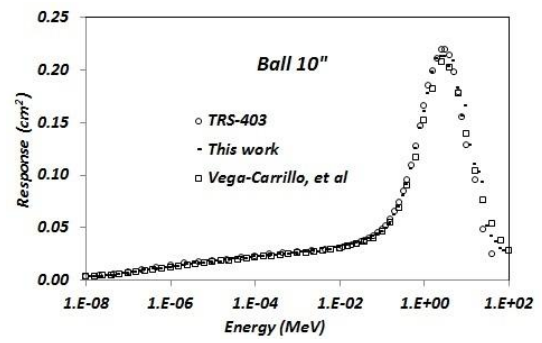


Figure 10. Neutron response function for BSS 10"

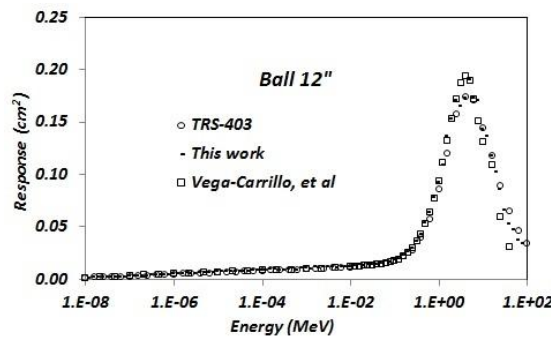


Figure 11. Neutron response function for BSS 12"

The response comparison between this work, IAEA TRS-403 and Vega-Carrillo, et al. are shown in Figure 5-11. In this work was done using MCNPX 2.6 with the ENDF66A/B.VI.1 and LA150 as cross-section libraries. The calculations of IAEA TRS-403 was done using MCNP4B[11], while Vega-Carrillo, et al. using MCNP4C and MCNPX. In Vega-Carrillo, et al. calculation for neutrons energy  $2.5 \times 10^{-8}$  MeV to 30 MeV was done using the ENDF/B-VI cross sections library and for energy 30 MeV to 100 MeV the response was obtained using the MCNPX code and the LA150 cross section library. In the IAEA TRS-403 calculation, neutron energy of  $1 \times 10^{-9}$  MeV –  $4.35 \times 10^2$  MeV divided in 51 energy bins, while the Vega-Carrillo, et al. divided in 23 energy bins. The both calculation was use normal direction beam source model and each value has an uncertainty less or equal to 3%[12].

The response function of Bare was shown in Figure 5 for neutron energy lower than  $10^{-7}$  MeV in this calculation was higher than Vega-Carrillo, et al., while in TRS-403 has not been calculated. This different values was affected from different cross-section library have been used. For BSS 2" and 3" the response function is in a good agreement and the influence of  $^6\text{Li}$  cross section shape is lost. In Figure 6 and 7 can be noticed that as the sphere's diameter is increased the response functions tend to decrease for thermal and epithermal neutrons, and the response's maximum are shifted to higher energies. The TRS-403 response's was a little bit higher than others for thermal and epithermal energy.

For Ball 2" to 12" the main differences are in the low energy region and for neutrons whose energy is larger to 20 MeV, i.e. those values calculated using MCNPX. Probable explanation of this difference is attributed to the cross sections utilized by Mares and Schraube for neutrons beyond 20 MeV[12]. They utilized the HIGH library, while in this work and Vega-Carrillo, et al. it were utilized those included in MCNPX.

A chi-square test ( $\chi^2$ ) can be used to see if there is a relationship between two categorical variables. In this calculation it was used chi-square to test for a statistically significant relationship between BSS response functions from this work, TRS-403, and Vega-Carrillo, et al. calculations. The test was done using  $\alpha = 0.05$ ; with  $\chi^2$  in the chi-square table (critical value) is 0.02. The  $\chi^2$  value of BSS shown in the Table 1 are lower than critical

value. These results showed that the response function of BSS for this work, TRS-403, and Vega-Carrillo, et al. calculations are same. Therefore, according to Chi-square test, the differences between three calculations are not significant.

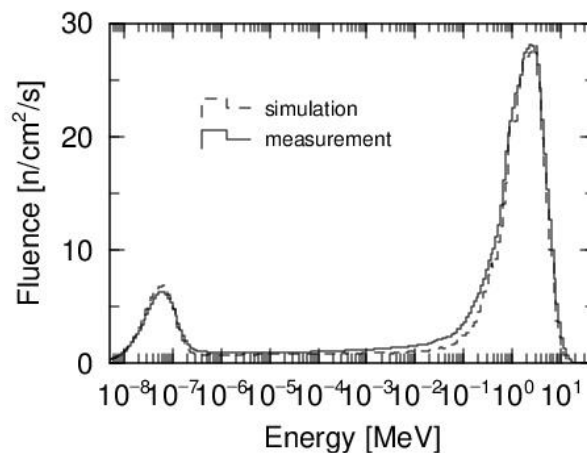
### Measurement of $^{252}\text{Cf}$ neutron spectra

To validate these response values calculated using MCNPX code, the experiment was done in the KRISS neutron calibration room with took place the BSS at 150 cm from the  $^{252}\text{Cf}$  neutron standard source. Based on KRISS calculation result, neutron emission rate of this source is  $1.37 \times 10^8$  n/s and fluence at the BSS position is  $4.89 \times 10^6 \text{ m}^{-2} \text{ s}^{-1}$ . Measurement of  $^{252}\text{Cf}$  neutron spectra using BSS  $^6\text{Li}(\text{Eu})$  at the KRISS neutron calibration room was shown in Figure 12.



**Figure 12.** (a) KRISS neutron calibration room, and (b) measurement of  $^{252}\text{Cf}$  neutron spectra using BSS

A commercial Ludlum Model 2200 scaler ratemeter is used to count the BSS after irradiation. Using UMG 3.3 unfolding code and BSS calibration factor of (1.53), the obtained the  $^{252}\text{Cf}$  neutron spectra from measurement are compared with the simulation results as shown in Figure 13. The total neutron fluence obtained with the BSS  $^6\text{Li}(\text{Eu})$  is  $8.46 \times 10^6 \text{ m}^{-2} \text{ s}^{-1} \pm 3\%$ . This total fluence is accumulation from neutron source and air scattered neutron and room-reflected neutrons from the walls, ceiling and floor. A shadow cone was used to prevent direct neutrons of source and only measured of neutron scattering. From this measurement obtained scattered neutron is  $3.73 \times 10^6 \text{ m}^{-2} \text{ s}^{-1} \pm 5\%$ , so neutron fluence is  $4.73 \times 10^6 \text{ m}^{-2} \text{ s}^{-1} \pm 4\%$ . The simulated BSS spectra were quite close to the experimental measured spectra ( $4.89 \times 10^6 \text{ m}^{-2} \text{ s}^{-1}$ ) with different up to 3%.



**Figure 13.** Measured  $^{252}\text{Cf}$  neutron spectra compared with simulated

## CONCLUSION

The response function of a conventional Bonner Sphere Spectrometer was carefully calculated with 111 discrete energy points from  $1 \times 10^{-9}$  MeV to  $1 \times 10^2$  MeV with an uncertainty of less than 2%, by simulation based on a MCNPX code. The simulation results was compared with two other response functions reported in the IAEA document in Technical Reports Series 403 (TRS-403) and calculated Vega-Carrillo, et al. Based on this comparison and chi-square test results, it was shown that the calculated response functions was in a good agreement with the two other response functions. The superiority of this work is on the number of energy bins and the lower uncertainty of the BSS responses. The spectra response of the BSS to the emitted neutrons by an  $^{252}\text{Cf}$  neutron source was also calculated by simulation, and the results are quite close to the spectra measured directly with a difference of 3%. This fact proves that the simulation of the BSS's response to neutrons by MCNPX is quite accurate.

## ACKNOWLEDGMENT

This work from joint cooperation between National Nuclear Energy Agency (BATAN) and Korean Research Institute of Standards and Science (KRISS). The authors gratefully acknowledge given to BATAN and KRISS to the financial and facilities support in this work.

## REFERENCES

- [1] J. M. Ortiz-Rodríguez, A. Reyes Alfaro, A. Reyes Haro, J. M. Cervantes Viramontes, and H. R. Vega-Carrillo, "A neutron spectrum unfolding computer code based on artificial neural networks," *Radiat. Phys. Chem.*, vol. 95, pp. 428–431, 2014.
- [2] T. Ogata, S. Kudo, Y. Watanabe, T. Muramatsu, H. Yamamoto, S. Iwai, S. Takagi, H. Harano, T. Matsumoto, and J. Nishiyama, "The calibration of bonner sphere spectrometer," *Radiat. Prot. Dosimetry*, vol. 146, no. 1–3, pp. 107–110, 2011.
- [3] A. Masuda, T. Matsumoto, H. Harano, J. Nishiyama, Y. Iwamoto, M. Hagiwara, D. Satoh, H. Iwase, H. Yashima, T. Nakamura, T. Sato, T. Itoga, Y. Nakane, H. Nakashima, Y. Sakamoto, C. Theis, E. Feldbaumer, L. Jaegerhofer, C. Pioch, V. Mares, A. Tamii, and K. Hatanaka, "Response measurement of a Bonner sphere spectrometer for high-energy neutrons," *IEEE Trans. Nucl. Sci.*, vol. 59, no. 1 PART 2, pp. 161–166, 2012.
- [4] M. Marek and L. Viererbl, "Bonner sphere spectrometer for characterization of BNCT beam," *Appl. Radiat. Isot.*, vol. 69, no. 12, pp. 1918–1920, 2011.
- [5] R. Bedogni, C. Domingo, K. Amgarou, M. De-San-Pedro, A. Esposito, A. Gentile, and A. Pola, "Spectrometry of 50 and 100 MeV quasi monochromatic neutron fields with extended range Bonner spheres," *Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip.*, vol. 746, pp. 59–63, 2014.
- [6] R. Bedogni, A. Esposito, A. Gentile, M. Angelone, and M. Pillon, "Comparing active and passive bonner sphere spectrometers in the 2.5 MeV quasi mono-energetic neutron field of the ENEA Frascati neutron generator (FNG)," in *Radiation Measurements*, 2011, vol. 46, no. 12, pp. 1757–1760.
- [7] S. Garny, V. Mares, H. Roos, F. M. Wagner, and W. Rühm, "Measurement of neutron spectra and neutron doses at the Munich FRM II therapy beam with Bonner spheres," *Radiat. Meas.*, vol. 46, no. 1, pp. 92–97, 2011.
- [8] W. H. Chu, J. H. Lan, T. C. Chao, C. C. Lee, and C. J. Tung, "Neutron spectrometry and dosimetry around 15 MV linac," in *Radiation Measurements*, 2011, vol. 46, no. 12, pp. 1741–1744.
- [9] G. Medkour Ishak-Boushaki, K. Boukeffoussa, Z. Idiri, and M. Allab, "Thick activation detectors for neutron spectrometry using different unfolding methods: Sensitivity analysis and dose calculation," *Applied Radiation and Isotopes*, vol. 70, no. 3, pp. 515–519, 2012.

- [10] J. Cao, X. Jiang, C. Jiang, H. Cao, and Y. Zejie, "Calculation of Response Function for Bonner Sphere Spectrometer," *Plasma Sci. Technol.*, vol. 17, no. 1, pp. 2013–2016, 2015.
- [11] International Atomic Energy Agency, *Compendium of neutron spectra and detector responses for radiation Protection purposes*, IAEA Technical Reports Series 403. Vienna, Austria: International Atomic Energy Agency, 2001.
- [12] J. I. G. Hector R. Vega-Carrillo, I. Donaire, E. Gallego, E. Manzanares, A. Lorente, M.P. Iniguez, A. Martin, dan, "Calculation of response matrix of a BSS with 6LiI(Eu) scintillator," *Rev. Mex. Fis.*, vol. S 54(1), pp. 57–62, 2008.
- [13] K. Nafisah and M. Rachid, "Neutron and Scintillator Gamma-ray Detection using a Cs2LiYCl6 Scintillator," *EPJ Web Conf.*, vol. 11018, pp. 4–7, 2014.
- [14] S. Esteban, C. Fleita, T. Ino, and H. Otono, "Measurement of fast neutron detection efficiency with 6 Li and 7 Li enriched CLYC scintillators," *J. Phys. Conf. Ser.*, vol. 763, pp. 1–5, 2016.
- [15] R. J. McConn, C. J. Gesh, R. T. Pagh, R. A. Rucker, and R. G. Williams, *Compendium of Material Composition Data for Radiation Transport Modeling*. 2011.
- [16] W. R. C. Pioch, V. Mares, "Influence of Bonner sphere response functions above 20 MeV on unfolded neutron spectra and doses," *Radiat. Meas.*, vol. 45(10), pp. 1263–1267, 2010.
- [17] W. Rühm, V. Mares, C. Pioch, S. Agosteo, A. Endo, M. Ferrarini, I. Rakhno, S. Rollet, D. Satoh, and V. H, "Comparison of Bonner sphere responses calculated by different Monte Carlo codes at energies between 1 MeV and 1 GeV – Potential impact on neutron dosimetry at energies higher than 20 MeV," *Radiat. Meas.*, vol. 67, pp. 24–34, 2014.
- [18] C. Pioch, V. Mares, E. . Vashenyuk, Y. . Balabin, and W. Rühm, "Measurement of cosmic ray neutrons with Bonner sphere spectrometer and neutron monitor at 79°N," *Nucl. Instruments Methods Phys. Res. Sect. A*, vol. 626, pp. 51–57, 2011.