



Journal of Applied Sciences

ISSN 1812-5654

science
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Determination of ^{241}Am -Be Spectra using Bonner Sphere Spectrometer by Applying Shadow Cone Technique in Calibration

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Abstract: In present study, the neutron spectra of ^{241}Am -Be measured with Bonner sphere method. The system was equipped with a BF_3 thermal detector placed in the center of seven spheres with different diameter (from 8.89 to 30.48 cm). The energy response matrix of the system was calculated by using the Monte Carlo simulation from thermal energy to 18 MeV. The spectrometry system was calibrated, using the shadow cone technique. For this purpose, a new shadow cone was used to prevent direct neutrons of source than to account on the scattered neutrons. Regardless, having a smaller length for new cone, passing the direct neutrons for it was negligible and less than old cone (proposed in ISO). Taking into account the low energy resolution of the BSS system, shape of unfolded spectra from experiment was comparable with the standard ISO ^{241}Am -Be spectrum.

Key words: ^{241}Am -be spectrum, Bonner sphere, scattered neutron, shadow cone technique monte carlo method

INTRODUCTION

Neutron particles find use in a diverse array of applications in areas of physics, engineering, medicine, nuclear weapons, petroleum exploration, biology, chemistry, nuclear power and other industries (Miri-Hakimabad *et al.*, 2010; Malkawi, 2004; Halim *et al.*, 2005; Ahmed *et al.*, 2006; Adib, *et al.*, 2005). The biological effectiveness of neutrons and neutron-induced secondary particles is so high, thus uncontrolled exposure to free neutrons is hazardous (ICRP, 1996, 2007; Miri-Hakimabad *et al.*, 2009; Basyigit, 2006). On the other hand, the neutron's quality factor depends on its energy and the response of existing neutron monitors and personal dosimeters is energy-dependent. So, the spectrometric information on the neutron radiation field is very important in radiation protection.

Among the many types of neutron spectrometers that have been developed in many specialized laboratories the world, the system known as multi-sphere, or more commonly as the Bonner Sphere Spectrometer (BSS), is widely used and well recommended by some authors for applications in radiation protection field (Bramblett *et al.*, 1960; Awschalom and Sanna, 1985). BSS consists of a thermal neutron counter embedded in moderator shell (usually polyethylene) and the associated electronics in the case of an active detector like BF_3 , ^3He or scintillators like ^6Li (Eu). The BSS is applied to characterize the

neutron field from thermal to GeV (Jacobs and van den Bosch, 1980; Liu *et al.*, 1990; Thomas *et al.*, 2002).

The BSS system is very useful since it is simple to operate, portable, has an isotropic response, covers a wide range of energy and the data can be unfolded and interpreted fairly easily (Thomas and Alevra, 2002). Several different types of thermal neutron sensors have been used at the center of BSS. The purpose of this study is to present a study to prepare the BSS based on a long proportional counter filled with BF_3 gas.

In this study, the response matrix of energy for BSS with BF_3 cylindrical detector has been calculated by Monte Carlo calculations using the computer code MCNP4C (Briesmeister, 2000) because the Monte Carlo method provides a powerful tool for detailed modeling of the process and geometry. One of the methods for calibration of system and determination of scattering contribution is shadow cone technique; in this procedure, a cone places between source and detector to count the scattered neutrons. Because of applying the long BF_3 counter, contribution of scattered neutrons was sizable. Thus distance between the neutron source and sphere must be decreased; however, for determination of the scattered neutrons, a new shadow cone should be designed having an appropriate length and a negligible transmission for direct neutrons (ISO 10647, 1996).

This work mainly focuses on providing conditions for measurement of ^{241}Am -Be (α, n) spectrum and discussion

of unfolded counting from experiment by BSS in various calibrations. The measurements and calculations were compared then the detail of some different was described.

**MONTE-CARLO MODELING OF THE BSS
RESPONSE FUNCTIONS**

The Bonner Sphere Spectrometer (BSS) under consideration constitutes of a set of polyethylene spheres of different diameters: 3.5", 4.2", 5", 6.5", 8", 10" and 12". The mean polyethylene density for all spheres of the set was $0.960 \pm 0.015 \text{ g cm}^{-3}$. The central thermal detector used is a 2.54 cm diameter 28.20 cm BF_3 cylindrical proportional counter (LND2210 type). The BF_3 wall was made of copper with 0.89 mm thickness and the effective length of this counter was 25.4 cm and also the pressure of gas filling of BF_3 , according to the manufacture, was 0.92 atm at 293 K. In this detector, enrichment of ^{10}B was about 96% (Fig. 1). In each sphere a 2.55 cm diameter cylindrical hole was drilled nearly to the center of the sphere.

The response of the BSS was calculated with the MCNP4C Monte Carlo Code, as the number of the ^{10}B (n, α) ^7Li reaction within sensitive detector volume per incident neutron fluence normalized to one starting particle. The irradiation geometry is based on a disk source, having the same diameter of the sphere under study, emitting a parallel neutron beam toward the sphere.

The energy response functions for seven detector configurations were calculated for different energies from

10^{-8} to 18 MeV. The discrete neutron energy values were selected at logarithmic equidistant intervals and at decade boundaries. The calculation of the response was accomplished being selected the tally F4 of MCNP4C, being considered the (n, α) reaction, associated to a card multiplier that contains the volume of the detector, the area of the disk source and the atom density (atom/barn cm) of BF_3 . For the thermal domain, the $S(\alpha, \beta)$ treatment was employed in the simulation. The statistical uncertainty was less than 4%. In Fig. 2, the energy responses of BSS system calculated with BF_3 long cylindrical proportional counter is shown.

The responses first increase, reach a maximum and then decrease. The reduced response of spheres at low neutron energies is due to capture of thermalized neutrons in the hydrogen of polyethylene. Furthermore, decrease of the response in high energies is due to escape of neutrons from polyethylene sphere. Considering the shape of the response functions, ascertain that the BSS system can be equipped with BF_3 long counter and used for neutron spectrometry.

CALIBRATION OF THE SYSTEM

There are many techniques that have been used to evaluate scattering corrections necessary for proper calibration of neutron spectrometer instruments, one of which is shadow cone technique. This technique relies upon the experimental determination of the scattered components, due to both walls reflected and air-scattered

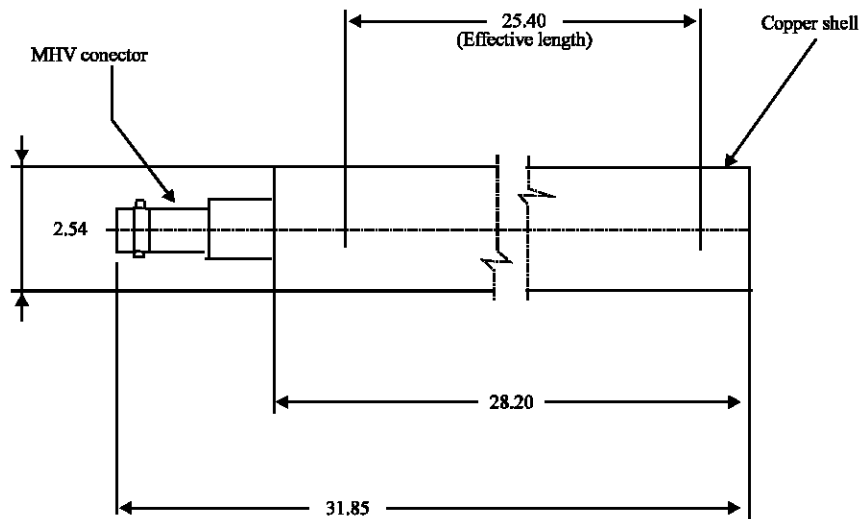


Fig. 1: Geometry of structure of BF_3 detector simulated in the calculations (all measures are in cm).

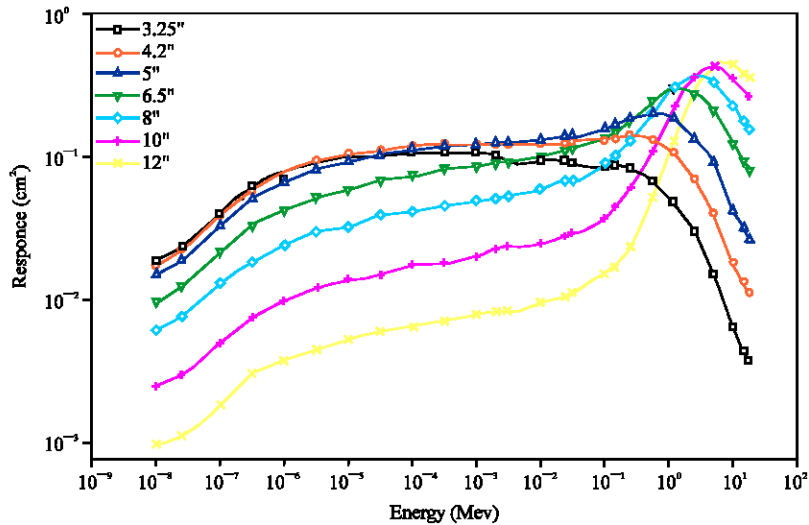


Fig. 2: Energy (MeV) response functions of BSS with BF₃ long cylindrical counter, as calculated with MCNP4C

neutrons, using a shadow cone designed to prevent any neutrons passing directly from the source to the detector. The method depends critically on the design of the shadow cone and upon its position relative to the source-detector geometry. One particular design of the shadow cone consists of two parts: a front end, with the length of 20 cm entirely made of iron and a rear section, with the length of 30 cm made of polyethylene (ISO 8529-2, 2000).

Geometry-correction factor for the finite source or detector size in order to tend to unit, generally measurements are made at a distance of more than twice the shadow cone length (ISO 10647, 1996). Therefore, considering the given shadow cone, the minimum distance of source to detector must be 1.0 m.

By using the MCNP4C code, the BSS system consisting of BF₃ counter was simulated base on the library (11.5×9.0×4.0 m) with 40 cm concrete walls. In this simulation, the neutron reference spectrum was investigated ²⁴¹Am-Be source. The spectrum for this source was extracted from the standard ISO 8529-1 (2001). For determining the total count due to direct and scattered neutrons, the ²⁴¹Am-Be neutron source was considered at the center of room and at 2.10 m height; the separation distance between the neutron source and center of each polyethylene sphere was 1.15 m. The source place was chosen with the goal of minimizing the scattered radiation at the sphere position. For calculating the scattering contribution, the shadow cone (30 cm polyethylene and 20 cm iron) was placed between neutron source and sphere, while distance between the center of sphere and the back face of the cone was 50 cm. The contribution of direct neutrons was given by subtraction of total counts from scattering counts.

In these conditions, the ratio of the scattered neutrons to direct neutrons for small spheres (3.5", 4.2" and 5") was more than 2.05, while it is expected to be less than 0.4 (ISO 10647, 1996). Because of using the long counter, a lot of scattered neutrons arrived at counter from a part of BF₃ that was out of sphere. To eliminate this problem, external part of the BF₃ was covered with 3.0 cm boric acid (H₃BO₃). The scattered neutrons are prevented to arrive in external part of BF₃, due to moderation and absorption in Boric Acid (BA). The ratio of the scattered neutrons to direct neutrons decreased to 1.23. With another calculation, it became clear that less than 4% of neutrons entered to external part of counter; thus it was not necessary to use other layers.

The next stage was decreasing the separation distance between the neutron source and sphere that was limited, because of the length of shadow cone. A new shadow cone was designed with smaller length. It had 35.0 cm length and three parts: a front end, with the length of 17.0 cm constituted by iron; a middle section, with the length of 14.0 cm consisted of solution water with 5% boric acid and a rear section, with the length of 4.0 cm which was made of boric acid entirely. The emitted neutrons from source arrive at iron and they are moderated to lower energy due to inelastic scattering interaction. Then, neutrons enter to water (with 5% boric acid) and some of them are thermalized or absorbed by ¹H and ¹⁰B. Finally, according to have a high capture cross-section for ¹⁰B, thermalized neutrons are absorbed in boric acid. This design was performed to optimize the length, mass and attenuation coefficient of shadow cone. Figure 3 shows the geometrical view of the Bonner Sphere System for counting the scattered neutrons by using shadow cone.

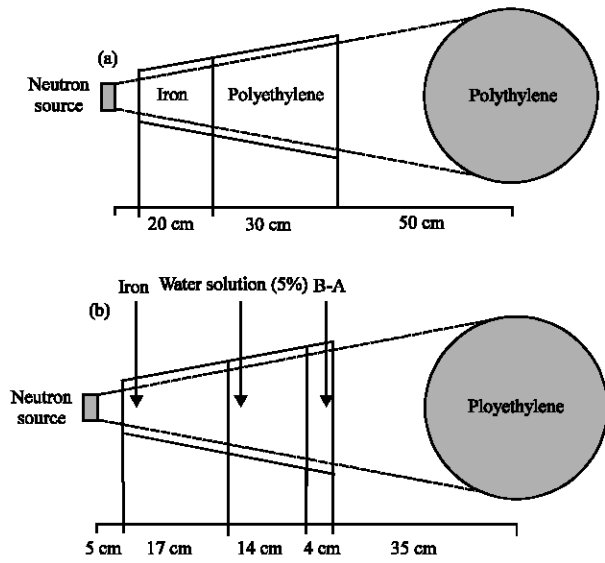


Fig. 3(a-b): Geometry of the irradiation with shadow cone for determination of scattered neutrons: (a) old shadow cone constituted by iron and polyethylene (ISO 8529-2, 2000) and (b) new shadow cone included of iron, solution of water (5%) and Boric Acid (BA)

The attenuation coefficient for a shadow cone is equal to division of detector counts in the presence of the cone to detector counts without cone while the counting must be in free space with no background. Table 1 shows the calculated attenuation coefficient for $^{241}\text{Am-Be}$ neutron source related to each cone, assuming old (50 cm) and new (35 cm) shadow cones. As in the calculation, distance between source and center of sphere with old and new shadow cones were 115 and 75 cm respectively. It can be seen that the new shadow cone has a smaller length but its attenuation coefficient is improved. Using the new shadow cone, separation distance between the $^{241}\text{Am-Be}$ source and center of sphere was considered 75.0 cm. The MCNP calculation revealed that the ratio of the scattered neutrons to direct neutrons in this separation distance, for small spheres (3.5", 4.2" and 5") and large spheres (6.5", 8" 10" and 12") was less than 0.38 and 0.25, respectively. Table 2 presents the calculated values of the ratio of scattered neutrons to direct neutrons for all spheres in all conditions for using old and new shadow cones.

MEASUREMENT

In experimental measurement by BSS system, seven spheres were positioned along axis of the centre of the source and were exposed separately

Table 1: Attenuation coefficient corresponding to each shadow cone for old (50 cm) and new (35 cm) shadow cones

Diameter (inches)	Old shadow cone (50 cm)	New shadow cone (35 cm)
3.5	3.90E-5	3.16E-5
4.2	4.24E-5	3.87E-5
5.0	4.01E-5	3.92E-5
6.5	5.96E-5	5.08E-5
8.0	6.17E-5	5.24E-5
10.0	8.65E-5	6.97E-5
12.0	1.22E-4	9.81E-5

Table 2: The calculated ratio of scattered neutrons to direct neutrons in various conditions

Diameter (inches)	Old cone without the BA	Old cone and using the BA on BF_3	New cone and using the BA on BF_3
3.5	2.345	1.418	0.380
4.2	2.178	1.313	0.361
5.0	2.055	1.231	0.330
6.5	1.890	1.140	0.303
8.0	1.680	1.020	0.280
10.0	1.340	0.940	0.260
12.0	1.080	0.911	0.250

BA: Boric acid

to fast neutrons from a $5\text{Ci } ^{241}\text{Am-Be}$ (50mm×30mm) neutron source for 1000 seconds at a distance of 75.0 cm between the source and the center of spheres. The BSS was equipped with a cylindrical 2.54 cm (diameter) by 28.2 cm (height) BF_3 (96% of ^{10}B). The detector was placed in the center of the laboratory and the source could be moved over a horizontal position at the same height as the detector. In this experiment, distance of detector from beside and behind walls of the room was 4.50 and 5.75 m, respectively.

The separation of the neutron-induced events from pulses due to noise or gamma ray induced events was performed by introducing a discrimination threshold below the lower limit of the neutron induced pulse-height distribution in MCA, whilst rejection of noise pulses did not have too much effect on the neutron sensitivity.

In order to calibration of system, the new designed shadow cone (with 35.0 cm length) was used between the source and sphere. The source-shadow cone distance was 5.0 cm. In these experiments three types of shadow cone with different obscured diameter were applied.

The resulting values from experimental measurement with different detectors for total neutrons (direct plus scattered neutrons), scattered neutrons and direct neutrons are illustrated in Fig. 4. It can be seen that with increasing diameter of spheres, the ratio of the scattered neutrons to direct neutrons (total neutrons minus scattered neutrons) is decreased. Also, for all spheres the ratio of the scattered neutrons to direct neutrons is less than 0.4.

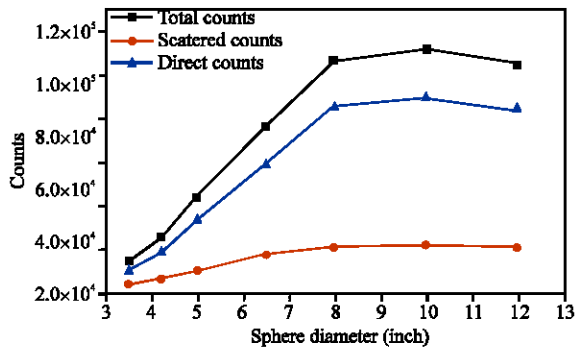


Fig. 4: Experimental measurement determined for different detectors in 1000 sec

The results obtained by the BSS system were unfolded with a modified version of the code SANDII (McElroy *et al.*, 1967). This code uses an iterative perturbation method to obtain a best fit neutron flux spectrum for a given input set of measured detector counts. The procedure consists of a flux spectrum that serves as the initial approximation to a solution.

The direct counts of the ²⁴¹Am-Be source and energy response functions that have been calculated with MCNP, were used as input values in SANDII. Therefore, in this calculation the neutron spectrum is determined at position of the detector. Also, another experiment with old shadow cone (30 cm polyethylene and 20 cm iron) was done. In this experiment, the separation between ²⁴¹Am-Be source and center of the sphere was 115 cm and external part of BF₃ was covered with 3.0 cm boric acid.

Figure 5 illustrates the experimental neutron spectrum of ²⁴¹Am-Be source that unfolded with the program SANDII, in comparison with ISO standard spectrum (ISO 8529-1, 2001) using the new shadow cone (35 cm) and old shadow cone (50 cm). As it can be seen a satisfactory agreement has been obtained for the spectrum ²⁴¹Am-Be source using the new shadow cone, taking into account the low energy resolution of the BSS system. In measurement applying the old shadow cone, spectrum increases in low energies values and spectrum decreases in energies values. Also, the peak of spectrum extravagates to lower energy.

This deviation can be explained by having a great ratio of scattered neutrons to direct neutrons. Diverting the shape of spectrum in low energies happens due to higher excessive increase of this ratio for small spheres and an increase in count error. In general one can conclude that when scattering contribution increases, the spectrum deflects to lower energies.

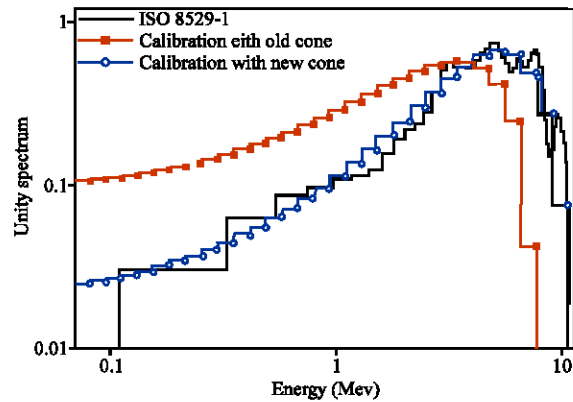


Fig. 5: Comparison between the experimental neutron spectrums for ²⁴¹Am-Be, using the new shadow cone (35 cm) and old shadow cone (50 cm) and the ISO standard spectrum

SUMMARY AND DISCUSSION

Computational and experimental evaluations of BSS calibration were performed with a ²⁴¹Am-Be (α,n) source. First, the response function of BSS system equipped with BF₃ calculated by Monte Carlo simulation. The general form of response ascertained that this detector can be a proper choice for use in BSS. For calibration of spectrometer, in order to determine the scattered neutrons, the shadow cone method was applied. Because of limitation in reducing of distance between source and sphere, proposed cone by ISO was unsuitable for this set-up and with use of this cone the ratio of scattered neutrons to direct neutron was greater than standard value. Therefore, a shadow cone with better performance and lower length was used. Then, to decrease the scattered neutrons, the rear part of the BF₃ long counter that is out of sphere has been covered with an appropriate thickness of boric acid.

An experimental limited to seven spheres and applying the new shadow cone for calibration has been performed with ²⁴¹Am-Be source in the library. Although the BSS system has inherently poor energy resolution, the results shown herein indicate a satisfactory agreement. Another experiment revealed that the shape of spectrum would be changed due to increase in scattering contribution. So, by using BSS system equipped to a long detector, base on calibration with new cone, the ²⁴¹Am- Be spectra can be obtained fairly reasonably.

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