MEASUREMENT OF THE NEUTRON SPECTRUM BY THE MULTI-SPHERE METHOD USING A BF₃ COUNTER

by

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The multi-sphere method, a neutron detection technique, has been improved with a BF₃ long cylindrical counter as a thermal detector located in the center of seven spheres with a diameter range of 3.5 to 12 inches. Energy response functions of the system have been determined by applying the MCNP4C Monte Carlo code of 10^{-8} MeV to 18 MeV. A new shadow cone has been designed to account for scattered neutrons. Although the newly designed shadow cone is smaller in length, its attenuation coefficient has been improved. To evaluate the system, the neutron spectrum of a ²⁴¹AM-Be source has been measured.

Key words: neutron fluence spectrometry, Bonner sphere, response function, shadow cone

INTRODUCTION

Neutron particles play an important role in many fields of science. These particles find use in a diverse array of applications in physics, engineering, medicine, nuclear weapons, petroleum exploration, biology, chemistry, nuclear power, and other industries [1-8]. The biological effectiveness of neutrons and neutron-induced secondary particles is so high that any uncontrolled exposure to free neutrons is hazardous [9-11]. On the other hand, the neutron's quality factor depends on its energy, while the response of existing neutron monitors and personal dosimeters is energy-dependent. Thus, spectrometric information on the neutron radiation field plays a crucial role in radiation protection. Among many types of neutron spectrometers that have been developed, the system known as the multi-sphere, or more commonly, the Bonner sphere spectrometer (BSS), has been built and used by more laboratories all over the world than any other type of spectrometer available [12, 13].

It consists of a thermal detector, a set of polyethylene spheres and associated electronics in the case of an active detector like BF₃, ³He or scintillators like ⁶Li(Eu). Several passive systems have been built, *e. g.* those utilizing gold foils or TLD pairs, too. The BSS characterizes the neutron field from thermal to GeV [14-21].

The BSS system is very useful, since it is simple to operate, portable, has an isotropic response, covers a wild range of energy and data and can be unfolded and interpreted fairly easily [22]. Several different types of thermal neutron sensors have been used at the center of BSS. The purpose of this paper is to present a study on the BSS based on a long proportional counter filled with BF_3 gas.

In this work, the response functions of energy for the BSS with a BF₃ cylindrical detector have been calculated by means of Monte Carlo calculations using the MCNP4C computer code with an ENDF/B-VI neutron cross-section and $S(\alpha, \beta)$ tables for thermal neutrons [23]. The simulation included a detailed description of the geometry of the BF3 counter filled with 0.92 atm (1 atm = 1013.25 hPa) gas. By applying a long BF₃ counter, the resulting contribution of scattered neutrons was also great. Thus, the distance between the neutron source and the center of the sphere needs to be decreased. However, for a more precise determination of scattered neutrons, a new shadow cone with an appropriate length and negligible transmission of direct neutrons needs to be designed. At the NLFUM (Nuclear Laboratory of the Ferdowsi University of Mashhad), the energy spectrum for a ²⁴¹Am-Be neutron source was measured by this system. Lastly, the effect of the scattering contribution on the spectrum was studied.

THE RESPONSE MATRIX OF THE BSS SYSTEM

The reading C_i of the thermal neutron sensor inside the *i*th Bonner sphere, when exposed in a point of a neutron field, can be expressed as

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$$C_i \quad \begin{array}{c} E_{\max} \\ R_i(E) \Phi(E) dE \\ E_{\min} \end{array}$$
(1)

where $\Phi(E)$ is the neutron fluence, and $R_i(E)$ is the response function of the sphere. The response matrix of a spectrometer is a set of energy response functions of different spheres. In this work, energy response functions of the BSS system were calculated with the MCNP4C Monte Carlo code.

An ABF₃ cylindrical proportional counter with a height of 28.2 cm and diameter of 2.54 cm (LND2210 type) was located in the center of seven polyethylene $(C_2H_4)_n$ spheres with diameters of: 3.5, 4.2, 5, 6.5, 8, 10, and 12 in* (from 8.89 cm to 38.10 cm); the density of the polyethylene was 0.9 g/cm³. The BF₃ wall was made of copper with a thickness of 0.89 mm, while the effective length of this counter was 25.4 cm and the pressure of BF₃ gas filling, according to the manufacturer, was 0.92 atm at 293 K. In this detector, the enrichment of ¹⁰B amounted to about 96%.

The response of the BSS was calculated as the number of counts per incident neutron fluence. The irradiation geometry is based on a disk source, having the same diameter of the sphere under study. All neutron tracks were parallel to the source-detector axis and fully included the entire surface of the moderating sphere (fig. 1).



Figure 1. General view of the geometric set-up simulaed in the calculations

The energy response functions for seven detector configurations were calculated for energies from 10^{-8} MeV to 18 MeV. Discrete neutron energy values were selected at logarithmic equidistant intervals and at decade boundaries. The calculation of the response was accomplished by selecting the tally F4 of the MCNP4C code, considered to be 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 BF₃. 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 the BSS system calculated with a BF₃ long cylindrical proportional counter are shown.

In order to compare the calculated response matrix with another counter, energy response functions



Figure 2. Energy response functions of the BSS with a BF₃ long cylindrical counter, as calculated by MCNP4C

for spheres of diameters of 3.5, 5, and 12 in with a ³He detector (typical SP9) [18], are plotted together in fig. 3. For the ³He spherical counter with a diameter of 3.2 cm, the gas filling was assumed to be 172 kPa. The responses were normalized to the value corresponding to the energy of 5.0 MeV for a 12 in sphere. This was necessary because of the different sensitivities of the respective thermal neutron detectors.

It is obvious from fig. 3 that the response matrices are approximately alike in shape; however, in the case of BF₃, the scale of the response function for small spheres, *i. e.*, those of 3.5 and 5 inches, is less than the one for spheres of the same size equipped with ³He. This may be explained by means of the long counter. When a long BF₃ is located in the center of the sphere, a part of the counter is out of the sphere; therefore, the efficiency of the detector decreases. Also, the beginning of the ³He gas is at a distance of about 1.6 cm from the center of the sphere toward the source, while the beginning of the BF₃ gas is at a distance of around 1.5 cm from the center of the sphere backward from the source. Considering the shape of the response functions, ascertain that the BSS system is equipped with a BF₃ long counter.



Figure 3. Comparison between the energy response functions of BF₃ and ³He [18] for 3.5, 5, and 12 in. To normalize the responses of ³He, they have been multiplied by a factor of 0.213

^{* 1} in or 1" means 2.54 cm

DESIGN OF THE NEW SHADOW CONE

In an ideal irradiation facility in free space, with no background due to air and room scattering and no source anisotropy, a detector would exhibit the so called free-field response. In practice, when the BSS system is exposed to a neutron source, the count in the thermal detector is due to direct (un-collided) and scattered neutrons. The contribution of room-reflected neutrons to the response of the detector may be significant, particularly if the counter is sensitive to low energy neutrons resulting from room scatter [24].

There are many techniques that have been used to evaluate scattering corrections necessary for the proper calibration of neutron spectrometer instruments, one of which is the shadow cone technique. This technique relies on the experimental determination of scattered components, due to both wall-reflected and air-scattered neutrons, and a shadow cone designed to prevent any neutrons passing directly from the source to the detector. The said method depends critically on the design of the shadow cone and its position relative to the source-detector geometry. One particular design of the shadow cone consists of two parts: a front end, 20 cm long and entirely made of iron and a rear section 30 cm long and made of polyethylene [25].

In order to tend to the unit, generally, the geometry-correction factor for the finite source or detector size, measurements are made at a distance of greater than twice the shadow cone length [26]. Therefore, considering the given shadow cone, the minimum source-to-detector distance must be no less than 1.0 m.

By using the MCNP4C code, the BSS system consisting of a BF₃ counter was simulated based on the NLFUM (11.5 m × 9.0 m × 4.0 m) with 40 cm concrete walls. In the simulation, the neutron reference spectrum investigated an ²⁴¹Am-Be source (50 mm in height and 30 mm in diameter). The spectrum for this source was extracted from the standard ISO 8529-1 [27]. For determining the total count due to direct and scattered neutrons, the ²⁴¹Am-Be neutron source was considered to be at the center of the NLFUM, at a

height of 2.1 m; the separation distance between the neutron source and the center of each polyethylene sphere was 1.15 m. The position of the source was chosen with the aim of minimizing scattered radiation within the sphere itself. For calculating the contribution of scattering, the shadow cone (30 cm polyethylene and 20 cm iron) was placed between the neutron source and the sphere, while the distance between the center of the sphere and the back face of the cone was 50 cm. The contribution of direct neutrons was given by subtracting the scattered counts from the total count.

Since the probability of a source neutron scattered from the walls of the laboratory to be detected by the detector was small, the forced collision method was used for scoring detector cells in order to increase the number of collisions that may produce large contributions in the detector. This technique forces the particles to undergo collisions by splitting them into collided and un-collided parts and then adjusting the weight according to collision probability.

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 [26]. Because the long counter was used, a lot of scattered neutrons reached the counter from a part of BF₃ that was out of the sphere. To eliminate this problem, the external part of the BF3 was covered with 3.0 cm of boric acid (H₃BO₃). In this manner, the scattered neutrons are prevented from reaching the external part of BF₃, due to the moderation and absorption into the boric acid. The ratio of scattered neutrons to direct neutrons decreased to 1.23. With another calculation, it became clear that less than 4% of neutrons entered the external part of the counter; thus, it was not necessary to use additional layers. Table 1 presents the numerical values for all calculations.

The next stage was decreasing the separation distance between the neutron source and the sphere, limited by the length of the shadow cone. A new shadow cone, smaller in length, was designed. It was 35.0 cm long and made in three sections: the front part, made of iron, 17.0 cm long; middle section, consisting

Table 1. Calculated values of the total counts scattered counts and contribution of neutrons arriving to the external part of BF₃, for the old shadow cone without and with boric acid (B-A) on the external part of BF₃, and the new shadow cone with boric acid on the detector; the counts were for 10⁶ neutrons emitted from an isotropic ²⁴¹Am-Be source

	Old cone without the B-A on BF ₃			Old cone, with B-A on BF ₃			New cone, with B-A on BF ₃		
Diameter [in]	Total	Scattered	Interring the external par of BF ₃ [%]	Total	Scattered	Interring the external par of BF ₃ [%]	Total	Scattered	Interring the external par of BF ₃ [%]
3.5	0.853	0.598	35.9	0.370	0.217	3.9	0.483	0.133	2.1
4.2	1.230	0.843	22.6	0.849	0.482	3.3	1.079	0.286	1.7
5	1.845	1.241	16.5	1.432	0.790	2.8	1.968	0.488	1.6
6.5	3.037	1.986	10.1	2.711	1.444	2.1	3.921	0.912	1.1
8	3.455	2.166	7.3	3.111	1.571	1.5	5.023	1.098	0.9
10	3.974	2.276	5.2	3.779	1.831	1.1	5.424	1.118	0.6
12	3.575	1.856	4.7	3.376	1.609	0.9	5.103	1.020	0.5



of water and 5% boric acid, 14.0 cm long; and a rear section, 4.0 cm long, entirely made of boric acid (fig. 4).

Figure 4. The newly designed shadow cone; front end, 17.0 cm in lenght,

made of iron; middle section, 14.0 cm

in length, consisting of water with 5% boric acid; rear section, 4.0 cm in lenght, entirely composed of boric acid, the ²⁴¹Am-Be source has 50 mm in height and 30 mm in diameter

The neutrons emitted from the source reach the iron and are moderated to a lower energy due to the inelastic scattering interaction. Following this, neutrons enter the water (with 5% boric acid), and some of them are thermalized or absorbed by ¹H and ¹⁰B. Finally, in order to achieve a high capture cross-section for ¹⁰B, thermalized neutrons are absorbed by the boric acid. This design optimizes the length, mass, and attenuation coefficient of the shadow cone.

In a free space with no background, the detector counts being M_0 without the shadow cone, and M when the shadow cone is between the source and the sphere, the attenuation coefficient will be M/M_0 . Table 2 shows the calculated attenuation coefficient for a ²⁴¹Am-Be neutron source and half angle (the angle between the axis and the boundary surface of the cone) related to each cone, assuming old (50 cm) and new (35 cm) shadow cones. As can be seen, the new shadow cone is smaller in length, but its attenuation coefficient has improved.

Using the new shadow cone, the separation distance between the 241 Am-Be source and the center of the sphere was considered to be 75.0 cm. The MCNP calculation revealed that the ratio of scattered neutrons to direct neutrons at 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.

SPECTRAL MEASUREMENT

For testing purposes, the multi-sphere method in combination with the newly designed shadow cone was applied to known neutron-energy spectra. A ²⁴¹Am-Be source (50 mm in height and 30 mm in diameter) was used. The measurement was performed at the NLFUM. The detector was placed in the center of the laboratory with the source in a position which allowed it to be moved along a horizontal line at the same height as the detector. In this experiment, the distance of the detector from side walls and the rear and front of the room was 4.50 m and 5.75 m, respectively.

The counts of the detectors were determined at a source-sphere distance of 75.0 cm. The separation of 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 the rejection of noise pulses did not have a significant effect on neutron sensitivity.

In order to account for the scattered neutrons, the newly designed shadow cone (35.0 cm in length) was used between the source and the sphere. The source-shadow cone distance was 5.0 cm. In this experiment, three types of shadow cones with different obscured diameters were applied, fig. 5(a).

The resulting values of these experimental measurement with different detectors for total (direct plus scattered neutrons) and scattered neutrons are listed in tab. 3. Figure 6 shows the comparison between direct

Table 2. Attenuation coefficient and opening angle corresponding to each of the old (50 cm), and new (35 cm) shadow cones; as calculated, the distances between the source and the center of the sphere for both the old and new shadow cones were 115 cm, and 75 cm, respectively

Diamatar [in]	Old shadow	cone (50 cm)	New shadow cone (35 cm)		
	<i>M</i> / <i>M</i> _o Half-angle (degrees)		$M/M_{ m o}$	Half-angle (degrees)	
3.5	$3.90 \ 10^{-5}$	0.97	3.16 -5	1.49	
4.2	$4.24 10^{-5}$	1.41	$3.87 \ 10^{-5}$	2.16	
5	$4.01 10^{-5}$	1.92	$3.92 \ 10^{-5}$	2.94	
6.5	$5.96 \ 10^{-5}$	2.86	$5.08 \ 10^{-5}$	4.39	
8	$6.17 \ 10^{-5}$	3.81	$5.24 \ 10^{-5}$	5.83	
10	8.65 10 ⁻⁵	5.07	$6.97 \ 10^{-5}$	7.74	
12	$1.22 \ 10^{-4}$	6.32	9.81 10 ⁻⁵	9.64	



Figure 5. The set-up of the measurement with the BSS system at the NLFUM; the system was placed at a height of 2.1 m, while the distance of the detector from the side walls and those at the front and rear ends of the room was 4.50 m, and 5.75 m, respectively

(a) with new shadow cone, (b) with old shadow cone

Table 3. Experimental measurement determined for different detectors in 1000 seconds

Detector	Sphere diameter [in]	Total c	ount	Scattered	count
1	3.5	14565		4152	64
2	4.2	26001	161	7028	84
3	5	44781	212	10593	103
4	6.5	76554	277	17377	132
5	8	106478	326	21268	146
6	10	111897	335	22276	149
7	12	105030	324	20959	145



Figure 6. Comparison of the direct count rates measured by the BSS system and those calculated as a function of the sphere diameter

count rates measured by the BSS and those calculated by the ISO standard neutron spectrum for ²⁴¹Am-Be. The measurement and calculation are in good agreement if the experimental errors are taken into account. For some spheres, the measured direct count rates are slightly greater than the values established by calculation. This can be attributed to the overshadowing of these spheres during the measurement.

The results obtained by the BSS system were unfolded with a modified version of the SANDII code [28]. 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 by MCNP were used as input values in SANDII. Therefore, in this calculation, the neutron spectrum is determined at the position of the detector. Also, to evaluate the effect of the contribution of scattering on spectrometry, another experiment with an old shadow cone (30 cm polyethylene and 20 cm iron) was performed. In this experiment, the separation between the ²⁴¹Am-Be source and the center of the sphere was 115 cm, and the external part of the BF₃ was covered with 3.0 cm of boric acid, fig. 5(b).

Figure 7 illustrates the experimental neutron spectrum of the ²⁴¹Am-Be source that unfolded with the SANDII program, in comparison with the ISO standard spectrum [27], using the new shadow cone (35 cm) and old shadow cone (50 cm). As can be seen, a satisfactory agreement has been obtained for the spectrum ²⁴¹Am-Be source using the new shadow cone, if the low energy resolution of the BSS system is taken into account. In the measurement with the old shadow cone, the spectrum increases in low energies



Figure 7. 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

values and decreases with higher energies values. Also, the peak of the spectrum shifts to lower energies.

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

The fluence-average neutron energy, E_{Φ} , was obtained as follows

$$E_{\Phi} = E\Phi(E)dE$$
 (2)

where $\Phi(E)$ is the neutron fluence. Table 4 presents the fluence-average neutron energy corresponding to the use of new and old shadow cones, and compares the result with reference to values and the report of Bedogni *et al.*, [29]. As can be seen, the result for using the new shadow cone is in agreement with the reference.

 Table 4. Comparison of the fluence-average neutron energy

	E_{φ} [MeV]	$\frac{E_{\varphi} E_{\varphi, \text{ref}}}{E_{\varphi, \text{ref}}} 100$
Reference	4.16	0
With old cone	1.76	57.96
With new cone	4.05	2.64
Bedogni report	4.24	1.92

CONCLUSIONS

A Bonner sphere spectrometer system based on the BF₃ long cylindrical proportional counter has been developed. The energy response functions of the spectrometer have been accurately simulated from 10^{-8} MeV to 18 MeV. Although the scale of the response decreased for small spheres in comparison to the ³He response; the shape of the response was proper for spectrometry standards.

One of the main aims of this work is the development of a system which would be less affected by scattered neutrons. To decrease the effect of scattered neutrons, the rear part of the BF3 long counter on the outside of the sphere has been covered with 3.0 cm of boric acid. In the next stage, to decrease the separation distance between the neutron source and sphere, a new shadow cone of a smaller length and improved attenuation coefficient was designed. An experimental validation limited to seven spheres, applying the new shadow cone for calibration, was performed via a ²⁴¹Am-Be source. Although the BSS system has an inherently poor energy resolution, the results presented herein indicate a satisfactory agreement, since the deviation from the fluence and neutron energy average of the reference value is low. Another experiment revealed that the shape of the spectrum will be changed due to the increase in scattering contribution. Therefore, the BSS system equipped with the BF₃ long cylindrical counter, if the said break throughs are applied, can be used in neutron fluence spectrometry.

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Рахим КАБАЗ, Хашем МИРИ ХАКИМАБАД

МЕРЕЊЕ НЕУТРОНСКОГ СПЕКТРА МУЛТИСФЕРНИМ ПОСТУПКОМ СА УПОТРЕБОМ ВF3 БРОЈАЧА

Мултисферна метода, једна техника детекције неутрона, унапређена је употребом дугог цилиндричног BF₃ бројача као термичког детектора, смештеног у центар седам сфера са пречницима од 3.5 до 12 инча. У интервалу од 10⁻⁸ MeV до 18 MeV, одређене су функције енергетског одзива система применом MCNP4C Монте Карло програма. Нови конусни заклон пројектован је ради урачунавања расејаних неутрона. Мада краћи по дужини, новопројектовани конус има унапређен коефицијент слабљења. Ради оцене система измерен је неутронски спектар једног ²⁴¹Am-Ве извора.

Кључне реч: сūекшромешрија неушронског флуенса, Бонерова сфера, функција одзива, конусни заклон