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Characterization of a new shielding rubber for use in neutron–gamma mixed fields

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Abstract A variety of formulations was investigated for the fabrication of an appropriate shielding rubber to be used in neutron–gamma mixed fields. Having considered the required mechanical properties together with tungsten as the gamma-ray absorbing element, calculations with MCNPX 2.6 code confirmed that the incorporation of 5 weight percentage (wt%) of boron carbide exhibited the best performance as a thermal neutron absorber. A series of both experimental and simulation results are provided for comparison.

Keywords Shielding · Rubber · Neutron · Gamma · MCNPX

1 Introduction

The protection against neutrons, gamma rays, and neutron–gamma mixed radiations is important in radiation medicine and biology, nuclear engineering, and space technology [1]. The problem is further complicated when dealing with applications in which the protection against both the neutrons and gamma rays is vital, such as in the detector shielding issues for prompt neutron activation analysis [2].

Since the gamma-ray absorption (the commonly used term for the absorption of photons is attenuation) increases with the mass and atomic number of the medium, tungsten is regarded as one of the best elements in gamma-ray shielding studies. Tungsten is non-toxic, exhibits a better performance in gamma-ray shielding compared to lead, and is easily dissolved in polymer substances. The neutron shielding materials, on the other hand, are very dependent on the neutron source type, its strength, and application type. Cadmium and boron are among the more well-known elements with large cross sections for thermal neutron absorption. There are some so-called fillers that can be used to convert rubber and plastic polymers into radiation shielding materials. Elasticity and ductility of a shielding material are very important properties, facilitating their use in medical and industrial applications. Radiation shielding suits and helmets, the shielding surrounding the densitometry systems, and the covering sheets of radiation-proof vehicles, are just a few examples.

Raw rubber compounds have relatively weak mechanical properties, and hence, fillers are used to enhance their properties, such as hardness, tension modulus, and fracture energy, as well as the tensile, tear, fatigue, and wear strengths. The reinforcement of elastomers and vulcanization are two basic processes for the improvement in the mechanical properties. The conventional reinforcing fillers, such as carbon, increase the tensile strength modulus and hardness, while the elongation properties decrease. Moreover, using reactive filling materials, even in small amount, causes a simultaneous increase in both the tensile modulus and strength with the elongation properties [3–5]. Although the production cost reduction is another motivation for

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using fillers such as carbon, the dark coloration of the final product has prompted some researchers to find alternative materials [6, 7].

Gwaily et al. studied the attenuation coefficients of rubber shields reinforced with boron carbide and paraffin as fillers when irradiated with neutrons and gamma rays. They showed that a combination of natural rubber with 20 phr (parts per hundred rubber) boron carbide and 60 phr of paraffin reduces the fast and thermal neutron intensities to 50 and 6%, respectively. They also showed that the addition of a maximum of 30 phr of lead does not affect the fast/slow neutron flux [8]. They also studied the neutron shielding properties of some natural rubber composites with different percentages of boron carbide. They performed a series of rheometry measurements to obtain the optimal cure conditions. Their results confirmed that the hardness increased with increasing B_4C contents, while the scorch time was reduced. The variation of attenuation coefficient with sample thickness for different concentrations of B_4C showed that the concentration of B_4C up to 20 phr increases the attenuation coefficient to the maximum value of 0.34 cm^{-1}, but higher concentrations do not show any significant further increases [9]. Gwaily et al. also studied both natural and styrene–butadiene rubbers with various concentrations of lead compounds, showing a substantial increase in the gamma attenuation coefficient.

Kaloshkin et al. studied the nanostructured compound of high molecular weight polyethylene with different concentrations of B_4C and tungsten [10]. They concluded that the use of nanoscale fillers improved both the gamma-ray shielding and mechanical properties. The studies showed that the tensile strength increased with increasing filler contents, while the elongation decreased. Their results showed that the gamma-ray attenuation coefficient reaches a maximum 3.43 cm^{-1} for 60% wt% of tungsten.

Abdel-Aziz and Gwaily developed gamma-ray shielding materials based on styrene–butadiene rubber and lead oxide (PbO), where PbO_2 and red lead oxide (Pb_3O_4) with 87% concentration were used. The linear attenuation coefficient for the Pb_3O_4/SBR (styrene–butadiene rubber) for different gamma-ray energy sources was estimated as 0.4 cm^{-1}. The sigma value (i.e., the total linear attenuation coefficient of the composite material for gamma rays of the appropriate energy) of lead oxide/SBR compounds slowly changes with respect to the radiation dose with different change rates. The decrease in the sigma value was dependent on oxidation number; a lower oxidation number resulted in a lower sigma value [11]. Abdel-Aziz et al. investigated ethylene propylene and polyethylene compounds of low molecular weight rubbers reinforced with 47 and 57 wt% for use in neutron shielding studies. The results showed that the gamma-ray attenuation coefficient is dependent on both the formulation and thickness of the compounds [12]. Gaier et al. showed that the graphite fibers exhibit suitable mechanical properties and can be considered as substitutions for different aluminum alloys [13].

Zhong et al. investigated the radiation shields made of epoxy–polyethylene fibers of high molecular weights [14]. They used graphite nanofibers to reinforce the epoxy composites. Their results show that the mechanical, thermal wetting, and adhesion properties of the compound with polyethylene fibers of high molecular weight reinforced with fillers are improved.

Ashton-Patton et al. investigated low molecular weight polyethylene reinforced with three microsphere hollow glass borosilicate, borosilicate, and aluminum silicates [15]. They examined the prepared samples for three

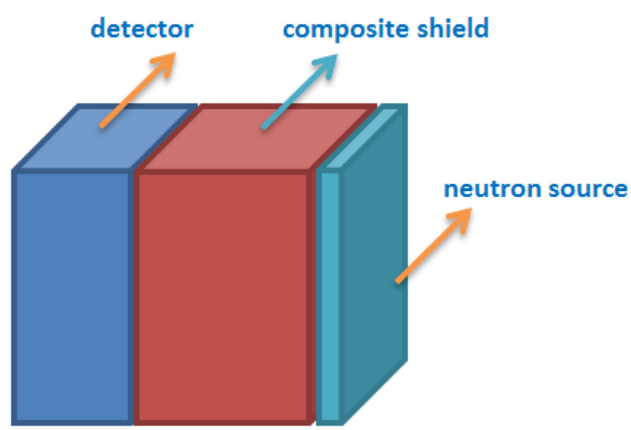

Fig. 1 (Color online) Schematic simulation setup for optimum boron content determination in typical shielding material

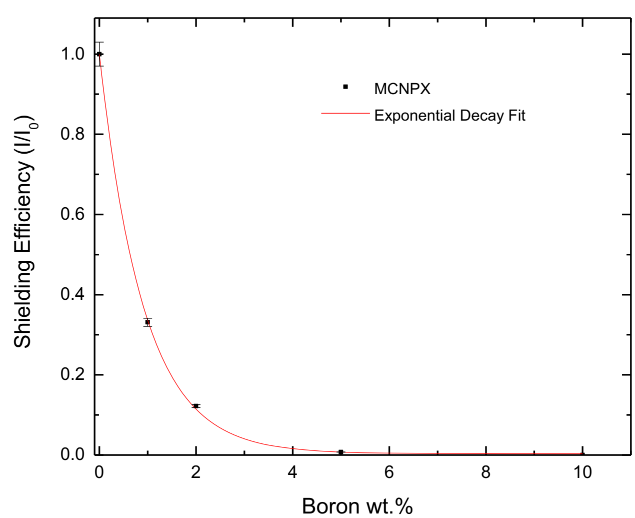

Fig. 2 (Color online) Variation of neutron counts against the boron contents in a shielding with 2 mm thickness. The neutron counts decreased to about one thousandth of its initial value by increasing the boron content to 5 wt%

Table 1 Definition of different shielding samples used in the present study

Rubber sample name	B_4C content (wt%)	Tungsten content (wt%)
W_1 (pure rubber)	0	0
W_2	5	5
W_3	5	15
W_4	5	30
W_5	5	35
W_6	5	45

different conditions with pressures and temperatures of (1) 6.51 MPa and 110 °C; (2) 3.9 MPa and 110 °C, and (3) 3.9 MPa and 120 °C. The borosilicate composites exhibited the best fractural strength in all three conditions. The use of microspheres together with the lowering of the specific weight improved the modulus. They also studied the hollow glass microsphere polyethylene to investigate the impact strength of shielding materials against high energy radiations.

Harrison et al. studied the mechanical and shielding properties of high molecular weight polyethylene composites reinforced with boron nitride [16]. The addition of 15 volume percent of pure boron nitride and signed boron nitride increased the composite modulus from the initial value of 588 to 735 and 856, respectively. They compared the shielding properties of polyethylene with 2 wt% of boron nitride with high molecular pure polyethylene and aluminum against few MeV neutrons and 120 GeV protons. In their studies, high molecular weight polyethylene and polyethylene/boron nitride showed similar shielding behaviors to aluminum.

El-Sayed Abdo et al. studied the neutron irradiation on polyethylene/lead/natural fiber composites [17]. They fabricated different composites made of plastic, lead oxide, boron carbide, and natural rubber with 5, 10, 15, and 50 cm thickness, respectively. The fabricated shields were irradiated with reactor-originating neutrons, and the neutron macroscopic cross sections were measured and compared with results from calculations.

Korkut et al. also studied the neutron shielding properties of samples with different boron contents [colemanite ($2CaO:3B_2O_3:5H_2O$), ulexite ($Na_2O:2CaO:5B_2O_3:16H_2O$), and tincal ($Na_2O:2B_2O_3:10H_2O$)], showing that the shielding properties improve as the boron content increases [18].

Singh et al. investigated the gamma-ray and neutron shielding properties of silicate and borate heavy metal oxide glasses; it demonstrated that bismuth silicate glass

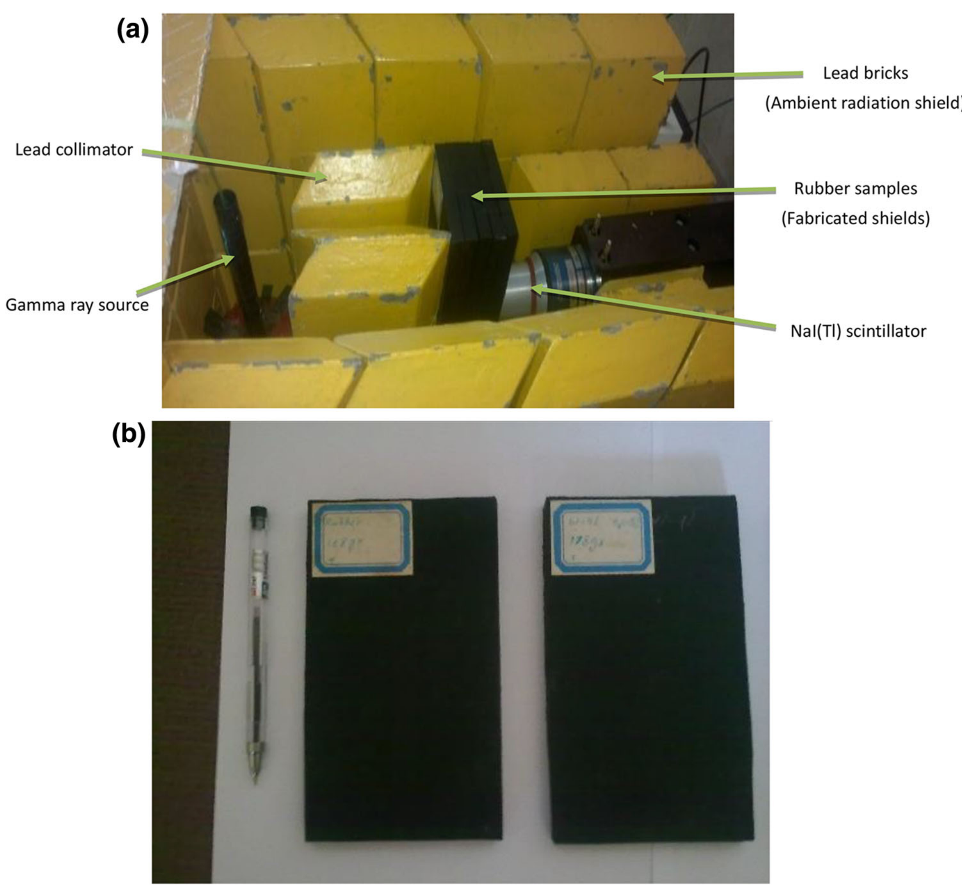

Fig. 3 (Color online) **a** Detection setup for the measurement studies including the NaI scintillation detector (right), gamma-ray source (left), and fabricated shields (middle), together with the lead collimator and ambient radiation shield. **b** Two samples of fabricated shields

has superior shielding properties and is suitable for the replacement of lead glasses [19].

Based on the results of the above experimental researches undertaken in recent years, a series of simulation studies (Sect. 2.1) and corresponding measurements (Sect. 2.2) were performed to obtain the most appropriate shielding material by incorporating different weight percentages of tungsten and boron carbide, as summarized in Sect. 3.

2 Experimental studies and results

2.1 Materials

In this study, to manufacture the most appropriate shielding rubber for use in the neutron–gamma mixed field, different wt% of B_4C (as the thermal neutron absorber) and tungsten (as the gamma-ray absorber) were added to pure rubber. The B_4C (98%, < 10 μm, Sigma-Aldrich [20]), tungsten fine powder (99.99%, 10 μm, Sigma-Aldrich [21]), and pure rubber from Iran Polymer and Petrochemical Institute (IPPI) [22] were purchased for the experimental studies.

2.2 Simulation studies

In this research, using Monte Carlo code, MCNPX 2.6 [23], the detection setup including a neutron source, radiation shield, and neutron counter was modeled, as shown in Fig. 1. The neutron source was ^{241}Am–Be with energies ranging from thermal up to 11.3 MeV and was ideally collimated to form a surface-type source (50 cm^2 in area) that emitted neutrons in the negative *x* direction. The neutron flux for different thicknesses of shielding materials, such as water, paraffin, polyethylene, concrete, iron,

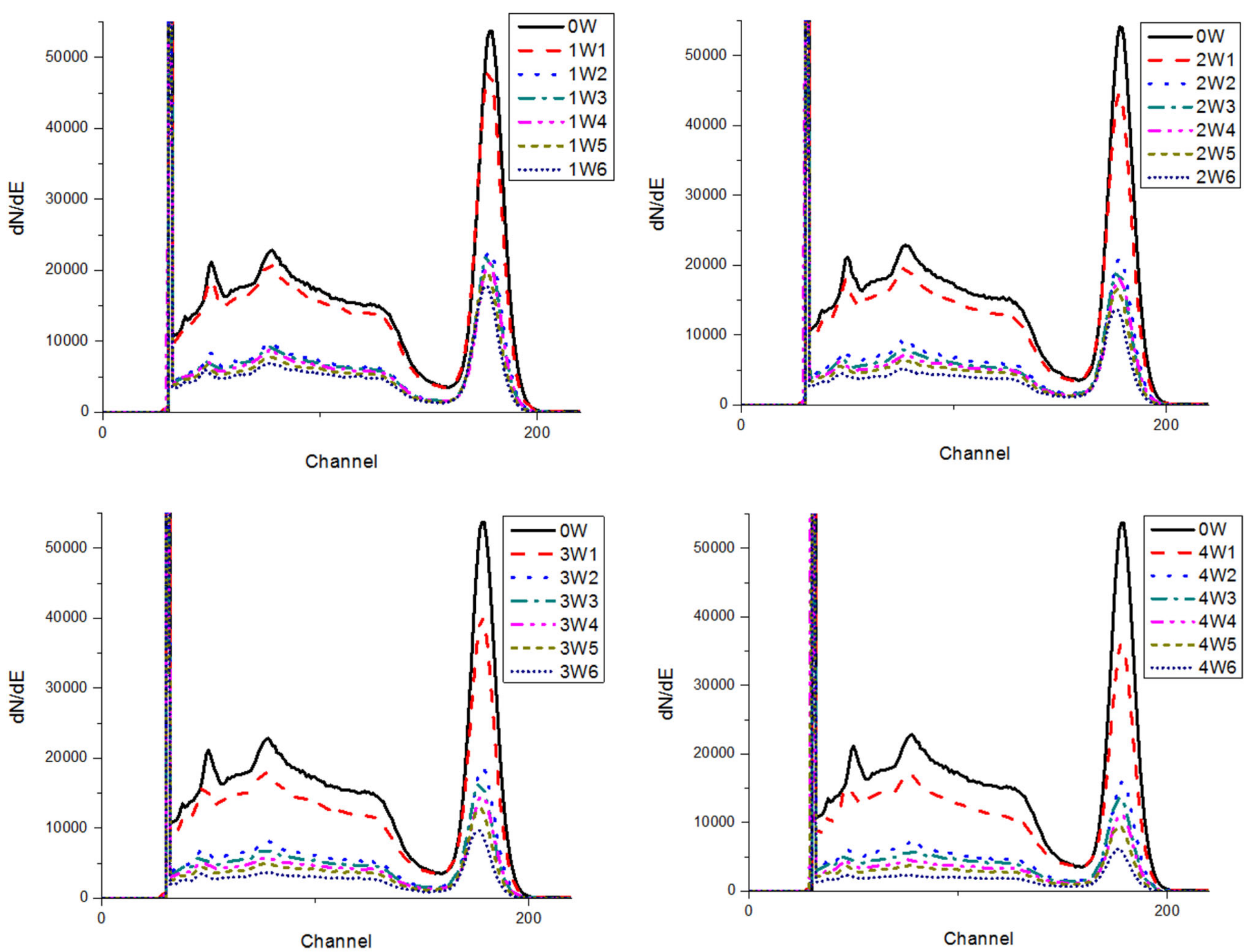

Fig. 4 (Color online) NaI(Tl) spectra of transmitted ^{137}Cs gamma rays through four different thicknesses (1, 2, 3, and 4 cm) of fabricated rubber shield samples with different wt% of tungsten (*n*W*m* stands for the *n* cm-thick rubber shield of sample number *m*)

graphite, borated polyethylene, and B_4C–polyethylene, was calculated. Boron carbide had the best performance among all shielding materials considered.

To find the optimum boron carbide content, different percentages of B_4C with a variety of thicknesses were considered. As expected, the studies showed that the detector counts decreased with thickness but reached a minimum when 5 wt% of B_4C was used (see Fig. 2). The value $(I/I_0) \times 100$ is the shielding efficiency representing the remaining percentage of the initial intensity after traveling a thickness x. Higher percentages of B_4C did not significantly change the detector counts, and it was concluded that the maximum 5 wt% of B_4C was the optimum value.

The use of boron as a neutron absorber in polyethylene produces ^{13}C de-excitation 478 keV gamma rays, in addition to the 2.2 MeV gammas from the hydrogen which must be further removed by using heavy elements, such as tungsten and bismuth.

2.3 Experimental studies

2.3.1 Fabrication of shielding material

The rubber shield used in this study contained 5 wt% of boron carbide for absorbing thermal neutrons, while different weight percentages of tungsten were incorporated for the gamma-ray attenuation, as listed in Table 1.

Two types of rubber materials, natural and artificial rubbers known as CR/NR, were reinforced with MGO/DPG/MBTS/S, ZnO/St asid/Wax/TMQ/4010NA, and W550/840 Oil/ETU/Anti Ox.Sp. These additives were prepared in a laboratory two-roll mill with 50 rpm at 30–40 °C mixing temperatures. The samples were fabricated with a hot press using compression molding in the form of five sheets with dimensions 1 cm × 9 cm × 16 cm. The hard phase of the boron carbide was chosen for the fabrication of the elastic sheets. The prepared sheets exhibited excellent heat-resistant, ozone-resistant properties, and comparatively low weight.

2.3.2 Radiation measurements

- Gamma-ray attenuation coefficient measurements

A 100 micro-Curie ^{137}Cs gamma source and an NaI(Tl) scintillator (with 300 s acquisition live time) were used for the measurements (Fig. 3). The spectra of different thicknesses (1, 2, 3, and 4 cm) of the fabricated samples are shown in Fig. 4, while Fig. 5 shows the logarithm of the relative intensity of the transmitted gamma rays against the sample thickness. The linear fit of the data in Fig. 5 provides the gamma attenuation coefficients for the samples with 5 different wt% of tungsten (see Fig. 6 for comparison). After carefully modeling the experimental setup of Fig. 3a, the MCNPX simulation was used to study the variation of the transmitted gamma rays versus the absorber shield thickness (for example, the W_2 sample). The comparison between measurement and simulation data exhibited very good agreement (Fig. 7). The simulation relative errors for all data points were less than 3%. We

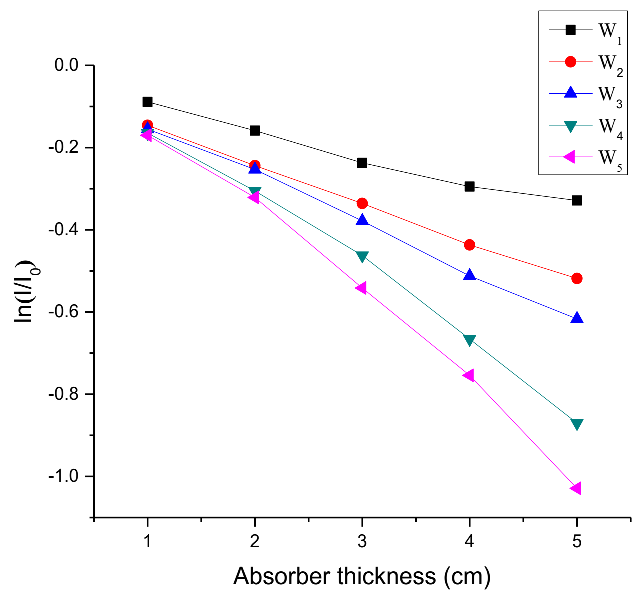

Fig. 5 (Color online) Variation of $\ln(I/I_0)$ with sample thickness. The slopes of the fitted lines are the attenuation coefficients shown in Fig. 6

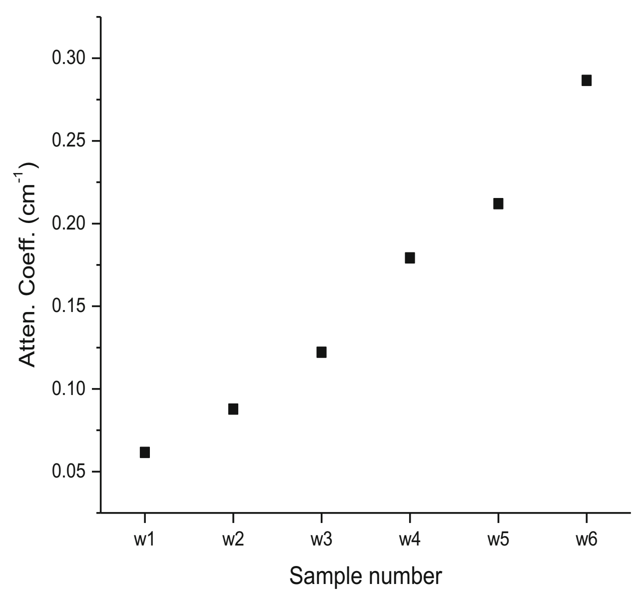

Fig. 6 (Color online) Gamma attenuation coefficient for the rubber shield samples for different wt% of tungsten

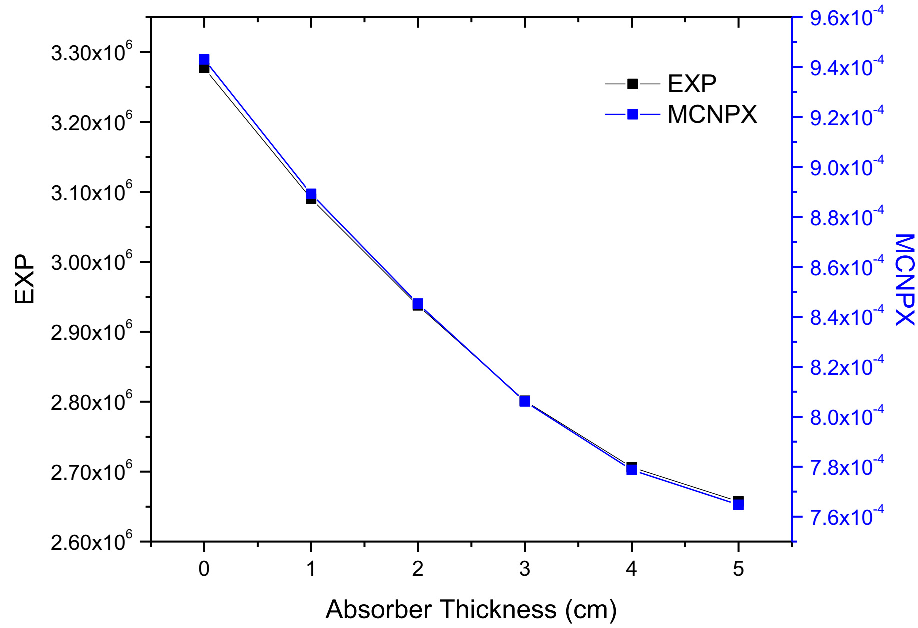

Fig. 7 (Color online) Experimental versus simulation results of the transmitted gamma-ray intensity for different thicknesses of the sample W_2

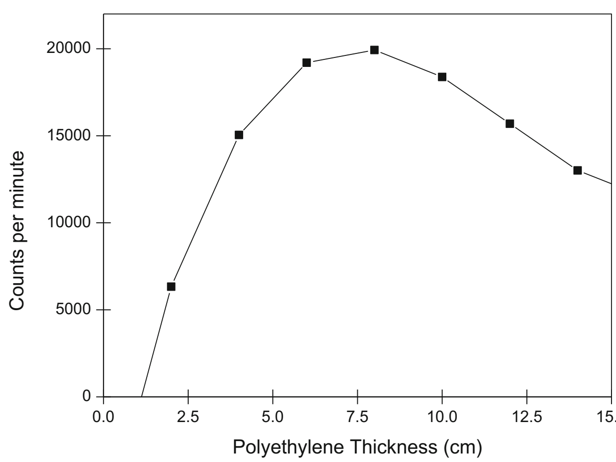

Fig. 8 Variation of thermal neutron counts with polyethylene thickness

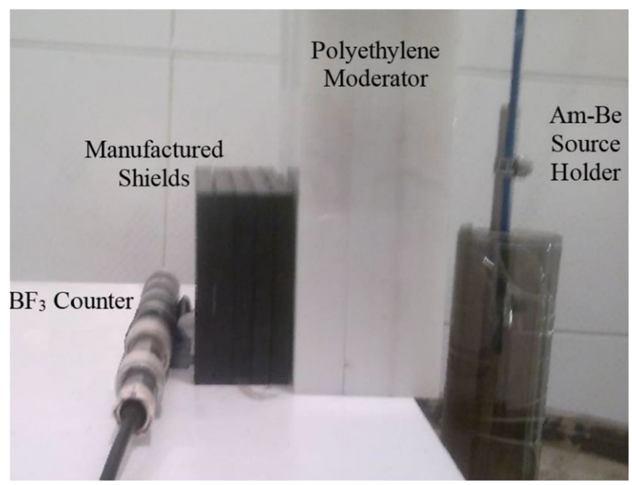

Fig. 9 (Color online) Experimental setup for the neutron absorption measurements of the fabricated samples

note that the MCNPX data were for one primary particle, while the experimental data comprised of the actual detector count rate that includes the source activity and detector efficiency.

- Neutron absorption measurements

A 100 micro-Ci Am–Be neutron source and BF_3 proportional counter were used for the measurements. The source-to-detector distance remained unchanged (30 cm) during the measurements, and a 7.5-cm polyethylene sheet was used as a neutron moderator. The optimum thickness of the polyethylene neutron moderator was obtained by plotting the thermal neutron counts registered by the BF_3 counter against the moderator thickness. As shown in Fig. 8, there is peak at 7.5 cm. This value also coincided with the MCNPX simulation results.

The thermal neutron absorption characteristics for the fabricated rubber samples were investigated by simply inserting the rubber samples between the Am–Be neutron source and BF_3 counter as shown in Fig. 9. The experimental data for the samples with different weight percentages of tungsten are illustrated in Fig. 10. Similar to the gamma rays (Fig. 5), the intensity followed the exponential decay function, $I(x) = I_0 \exp(-\alpha x)$. The constant α is the attenuation coefficient (denoted by μ) and the macroscopic cross section (denoted by Σ) for the gamma rays and neutrons, respectively. Therefore, $\ln(I(x)/I_0)$ is always negative.

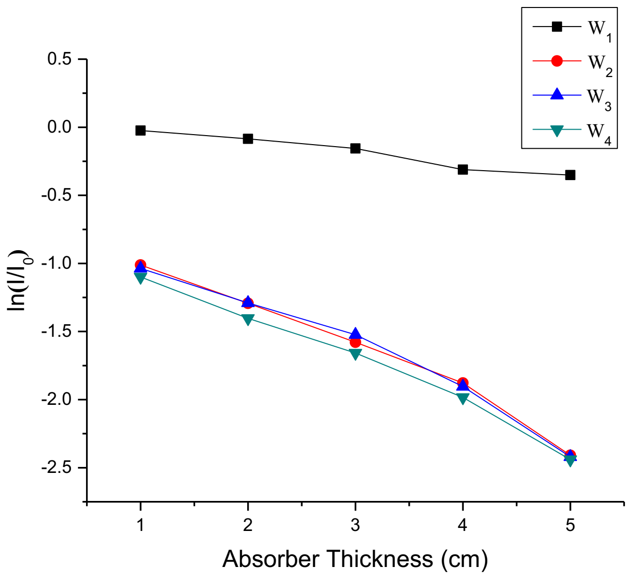

Fig. 10 (Color online) Variation of thermal neutron absorption coefficients against absorber thickness for four different fabricated samples measured with the BF_3 counter

As the boron carbide contents of the samples remained the same, the neutron absorption remained almost unchanged. The data confirmed that for every 5 wt% of boron carbide, the thermal neutron absorption coefficient doubled.

2.3.3 Mechanical tests

A number of mechanical tests were performed on the fabricated rubber shields, and some of these are listed below:

- The tensile as well as elongation-at-break tests showed that the tensile strength increased, by increasing up to 5 wt% of tungsten.
- The compression set results showed that by increasing the weight percentage of tungsten up to 30%, the viscoelastic damping increased.
- An increase in the fillers increased the hardness.
- The presence of fillers decreased the viscoelastic energy loss at the matrix–filler interface. The loss increased in the samples and the elasticity decreased.
- When the tungsten weight percentage increased from 30 to 45%, the cure system was affected and the cross-link density subsequently decreased.
- The rheometry results showed that for a tungsten weight percentage of less than 35, the scorch time and optimum cure time (t_{95}) were not significantly affected.

The mechanical test results are listed in Table 2.

3 Concluding remarks

In this research, a variety of materials, such as water, paraffin, polyethylene, concrete, iron, graphite, borated polyethylene, and boron carbide used as appropriate neutron shielding materials, were selected as candidates for neutron shielding studies. Having chosen rubber, due to its physical properties, the thermal neutron flux for different thicknesses of the manufactured shields was modeled with the MCNPX 2.6 code. The simulation results showed that the boron carbide provided the best performance. Moreover, the obtained optimum weight percentage of boron carbide was 5 wt%. We chose tungsten as the gamma-ray

Table 2 Mechanical test results for the fabricated rubber shields

Mechanical test	Maximum value	Tungsten (wt%)	Minimum value	Tungsten (wt%)
Hardness (shore A)	66.36	80	57.03	0
Compression set (%)	12	30	8.8	80
Abrasion strength (%)	31.94	80	12.93	0
Resilience (%)	47	0	37	80
Tensile strength (MPa)	8.678	5	3.639	80
Elongation (%)	268.705	5	196.802	35
Rheometry (t_{95}) (min)	6.31	15	4.65	35

Table 3 Variation of gamma flux attenuation in three different shielding samples

	Sample W_1					Sample W_2					Sample W_3				
Sample thickness (cm)	1	2	3	4	5	1	2	3	4	5	1	2	3	4	5
Decrease percentage of ^{137}Cs gamma-ray flux	9	16	38	60	80	11	20	42	63	87	14	24	45	64	88

Table 4 Transmitted neutron flux for different thicknesses of rubber shield samples

Shielding thickness (cm)	W_1	W_2	W_3	W_4	W_5	W_6
1	85	35	33	31	30	29
2	78	27	27	24	22	21
3	67	20	21	19	16	15
4	57	17	14	13	12	12
5	55	13	9	8	7	6

absorbing nuclei. The results of the gamma attenuation coefficients are listed in Table 3, and the variations of the thermal neutron flux are given in Table 4.

To produce rubber shields with improved mechanical properties, i.e., high flexibility and filling ability, natural and polar rubbers are ideal. However, to provide better resistance against ozone and acid, CR and non-polar rubbers are used. The combination of CR and NR forms a chemical multiple bond and provides a better compatibility and enhanced filling ability.

In conclusion, the most appropriate shielding material for use in a neutron–gamma mixed field was found by incorporating 5 wt% of boron carbide and different weight percentages of tungsten, which was varied depending on the application type and required mechanical properties.

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