Neutron attenuation and mechanical properties of polymer composites filled with boron carbide particles

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Abstract. In this work, neutron attenuation and mechanical properties of thermoplastic natural rubber (TPNR) blend filled with boron carbide (B4C) have been studied as a function of filler loading. Thermoplastic natural rubber of high density polyethylene/natural rubber (HDPE/NR) blend with different amounts of boron carbide (5–20 wt%) have been prepared via melt blending method. All samples were subjected to neutron transmission and tensile tests. The results showed that neutron shielding performance of the composites were improved significantly with the addition of B4C into TPNR matrix. On the other hand, mechanical properties of the composites were found to decrease with increasing filler loading.

1. Introduction
Recently, polymer-based composites have gained much attention by the radiation protection industry due to their excellent properties such as lightweight, good processability, good flexibility and low manufacturing cost [1]. For neutron radiation, polymers served as an ideal neutron shielding material because of their high hydrogen content. Neutron shielding requires low atomic number elements such as hydrogen with high scattering cross sections that can effectively moderate or thermalize incident neutrons.

Polyethylene is one of the effective neutron shielding materials and displays excellent attenuation behavior because of its hydrogen content (14% by weight) and its density [2, 3]. It is inexpensive and resistant to many chemicals and has hydrophobic nature which reduces the possibility of a superficial contamination to a minimum. In polyethylene, the hydrogen capturing of thermal neutrons is through the \( ^1\text{H}(n, \gamma)^2\text{H} \) reaction which has a cross section of 0.33 barn for neutrons in thermal equilibrium at room temperature \( (E_n = 0.027 \text{ eV}) \).

Elements with high absorption cross sections for thermal neutrons like boron, cadmium and gadolinium are often dispersed in the shielding material to absorb moderated/thermalized neutrons. Adding boron in polyethylene can reduce the dose from secondary gamma production from radiative capture \((n, \gamma)\). Boron is an excellent choice to absorb neutrons due to its high neutrons cross section of boron-10 isotope which is 3840 barns [4]. The capturing of thermal neutrons by boron is through the \( ^{10}\text{B}(n,\alpha)^7\text{Li} \) reaction. Another advantage of the boron-10 isotope is that the neutron capture reaction is creating alpha radiation that is easily stopped by the material.

In this study, thermoplastic natural rubber (TPNR) composites consist of high density polyethylene/natural rubber (HDPE/NR) blend were fabricated for neutron shielding materials. TPNR which has intermediate properties between rubber and plastic provide flexibility in shaping and making it attractive to be used as a matrix in a composite material. The unique properties of elastic and rigid of TPNR could
provide wide area coverage that require radiation shielding. Boron carbide (B₄C) with high thermal neutron cross section was used as filler in order to provide shielding effect against thermal neutrons [5-7]. Then the attenuation and mechanical properties of the TPNR/B₄C composites were studied as a function of filler loading.

2. Materials and method

2.1. Sample preparation
Thermoplastic natural rubber (TPNR) blend was prepared via melt blending of natural rubber and high density polyethylene (NR/HDPE) with the ratio of 60/40. Different amounts of boron carbide (B₄C) powders were added to get composites with different concentrations (5, 10, 15 and 20 wt%). Liquid natural rubber (LNR) was used as a compatibilizer. Mixing was done using an internal mixer (Haake Rheomix 600P) with a processing mixing temperature at 135 °C, a rotor speed of 55 rpm and mixing time of 15 minutes. Then, the composite compound was moulded in a compression moulding machine at 135 °C to form a slab (15 cm x 15 cm).

2.2. Neutron transmission test
Neutron transmission tests were conducted in TRIGA reactor Malaysian Nuclear Agency. All samples with 1 mm thickness were exposed to thermal neutron for equal interval of time (2 minutes) using the thermal neutron beam. Composite samples were installed at the beam port located 719 cm from the source. The transmitted neutron intensity (I) for each composite was determined by recording the corresponding counts using a fission chamber detector (Model 3053, LND Inc.). The incident neutron intensity (I₀) was determined when no composite material is in place. Total neutron transmission (I/I₀) and macroscopic cross section (Σ) of the composites were calculated using Beer-Lambert Law (equation 1) [8] where x is the thickness of the samples. Half value layer (HVL) of the samples were also calculated using equation 2.

\[ I = I_0 e^{-\Sigma x} \]  
\[ HVL = \frac{\ln 2}{\Sigma} \]

2.3. Tensile test
Five dumbbell-shaped specimens for each sample type were cut according to ASTM D638-91a with 1 mm thickness for tensile test. Tensile strength and elongation at break were measured using Instron 8874 tensile test machine with cross-head speed of 50 mm/min at room temperature. The test was performed according to ASTM D638 with maximum load of 5000 N. The average of the values was taken.

3. Results and discussion

3.1. Neutron attenuation
The neutron shielding characteristics of TPNR/B₄C composites at various filler loadings (5-20 wt%) were determined. Figure 1 shows neutron transmission of TPNR/B₄C composites as a function of filler loading and the values are listed in Table 1. It is clear from the figure that neutron transmission value decreases as filler loading increases. TPNR matrix have neutron transmission value of 0.674. The incorporation of B₄C filler into TPNR matrix decreases the neutron transmission and the value reaches to 0.302 at 20 wt% of B₄C.
The results for macroscopic cross section and the half value layer (HVL) are shown in Table 1. It can be seen that the highest value comes from a composite containing 20 wt% of B\textsubscript{4}C. The attenuation of the 20 wt% B\textsubscript{4}C composite is about 217% higher than the TPNR. A significant increase in macroscopic cross section as the B\textsubscript{4}C filler loading increases means that the probability of interaction increases as B\textsubscript{4}C content became higher. Therefore, more neutron can be absorbed by shielding material. Increasing of macroscopic cross section value of TPNR composites with B\textsubscript{4}C loading is due to the high thermal neutron absorption cross section of boron-10 isotope in B\textsubscript{4}C. Natural boron consists primarily of two stable isotopes, \textsuperscript{11}B (80%) and \textsuperscript{10}B (20%) and its (n, α) reaction cross section for thermal neutrons is about 3840 barns.

Increasing the percentage of B\textsubscript{4}C filler also causes the half value layer (HVL) to decrease. Half value layer indicates the required thickness of an absorber to reduce the radiation level to half of its initial value [9]. In this work, HVL decreases from 0.207 cm to 0.065 cm with B\textsubscript{4}C loading increase from 0 wt% to 20 wt% indicating an improvement of the attenuation properties of the composites. The concept of HVL is very useful in doing rapid, approximate shielding calculations [10].

### Table 1. Neutron attenuation properties of TPNR composites at various B\textsubscript{4}C loadings.

| Filler loading (wt%) | Neutron transmission \( I/I_o \) | Macroscopic cross section (cm\(^{-1}\)) | Half value layer (cm) |
|----------------------|----------------------------------|---------------------------------------|----------------------|
| 0                    | 0.674                            | 3.343                                 | 0.207                |
| 5                    | 0.543                            | 5.085                                 | 0.136                |
| 10                   | 0.428                            | 7.015                                 | 0.099                |
| 15                   | 0.352                            | 8.692                                 | 0.080                |
| 20                   | 0.302                            | 10.604                                | 0.065                |

3.2. **Mechanical properties**

Mechanical properties of the radiation shielding materials are important to minimize degradation of the materials during usage. The tensile strength and elongation at break of TPNR/B\textsubscript{4}C composites at different B\textsubscript{4}C loading are shown in Figure 2 and Figure 3 respectively and the values are summarized in Table 2. From Figure 2, it can be seen that the tensile strength decreases by loading B\textsubscript{4}C into TPNR
matrix. The tensile strength decreases from 3.9 MPa to 2.74 MPa when TPNR is loaded with 5 wt% of B₄C. Only 1.82 MPa of tensile strength is observed at 20 wt% loaded samples.

Small amount of filler gives better strength to the composites might be because of good rubber-filler interaction occur as a result of good dispersion of the filler particles into the matrix. Wang and Chen [11] reported in their study that lower amounts of filler can easily dispersed in the composites thus produced a significant improvement in the filler-matrix interfacial bonding. Better dispersion of the filler in the matrix may increase the efficiency of the stress transfer from matrix to filler phase. At higher filler loading, filler particles are no longer wetted properly by the TPNR matrix resulting in poor interfacial adhesion. In addition, the agglomeration and the aggregation of the filler particles in the TPNR matrix may have formed at higher filler loading and it caused interruption to the filler-matrix bonding [12, 13]. Thus, it will produce stress concentration points leading to early failure.

Figure 2. Tensile strength of the composites as a function of filler loading.

Figure 3. Elongation at break of the composites as a function of filler loading.
Elongation at break of the composites shows similar trend like tensile strength. As the filler loading increases, the elongation at break tend to decrease. The elongation at break value decreases from 162.96% to 55.55% by loading 5 wt% of B₄C. The lowest value of 21.29% can be observed at maximum filler loading of 20 wt%. The decreasing of elongation at break at higher filler loading is due to the enhancement in rigidity of the composites [14]. Azahari et al. [15] stated that the reduction in elongation at break may possibly affected by the attractive forces between the fillers and the polymer molecules. The forces restrict the free mobility of the polymer chains to form a cross-linked network. Thus, it will increase the resistance to stretch upon the application of strain.

Table 2. Tensile properties of TPNR/B₄C composites at different B₄C loadings.

| Filler loading (wt%) | Tensile strength (MPa) | Elongation at break (%) |
|----------------------|------------------------|------------------------|
| 0                    | 3.9                    | 162.96                 |
| 5                    | 2.74                   | 55.55                  |
| 10                   | 1.88                   | 25.01                  |
| 15                   | 1.85                   | 22.23                  |
| 20                   | 1.82                   | 21.29                  |

4. Conclusion
TPNR/B₄C composites with various B₄C loadings were successfully fabricated and studied. Neutron transmission and tensile tests were conducted to investigate the neutron attenuation and mechanical properties of TPNR/B₄C composites. It has been found that neutron transmission and half value layer of composites decreased while macroscopic cross section increased indicating an improvement of the attenuation properties of the composites with the increase of B₄C loading into TPNR matrix. On the other hand, tensile strength and elongation at break of the composites were decreased with increasing filler loading.

5. References
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