Polyethylene composite with boron and tungsten additives for mixed radiation shielding

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Abstract. Mixed neutron and gamma radiations are common in many nuclear applications. Several materials can be combined to obtain a composite material that is better for mixed radiation than the individual component materials. The aim of this paper is to investigate the shielding effectiveness of a polyethylene (PE)-based composite with boron and tungsten additives. Several compositions are tested to shield against a 252Cf fission neutron source using an attenuation experiment. The composite is also manufactured using the melt-mixing method of component raw materials. Comparisons are made between the different compositions and the experimental results. Results suggest that the PE composite with 16%wt boron and 16%wt tungsten show the best mixed radiation attenuation as compared to pure PE, PE composite with 25%wt boron, and PE composite with 25%wt tungsten.

1. Introduction
Concerns of mixed neutron and gamma radiations are common in most nuclear applications. This is due to the interaction of neutrons with matter that can result in the production of secondary gamma-rays. Therefore, the shielding for gamma-rays will also need to be considered along with the neutron shielding. Many polymer-based composites have been developed to tackle this problem [1]. This is because polymer has a high content of hydrogen which is excellent in neutron moderation. Other additives are added to improve the shielding performance of the base polymer such as boron to absorb thermal neutrons, and heavy elements to absorb secondary gamma-rays.

Boron-filled composites are popular among researchers of neutron shielding because of their high macroscopic absorption cross-section for thermal neutrons. Uddin et al. (2020) [2] investigated polyethylene(PE)/boron carbide composite for up to 40% loading. They noted that although higher compositions of boron are effective, this reduces the amount of hydrogen in the polymer base which can affect the moderation of fast neutrons. Thus, they recommended 10% boron content as the best composite formulation that balances the content of boron and hydrogen. Nurazila and Megat Harun (2020) [3] showed that adding boron carbide to a high-density PE (HDPE)/natural rubber blend improved the neutron shielding performance of the base material. However, higher boron content was found to diminish the mechanical properties of the base material. Jumpee et al. (2020) [4] also used natural rubber mixed with boron powder for neutron shielding and it exhibited good neutron attenuation. Additionally, another sample with iron oxide powder was also investigated and it produced a similar
shielding performance. Although, the gamma-ray transmission was not measured which could have revealed the potential of adding the iron oxide to improve the gamma-ray attenuation of the base material.

The gamma shielding performance of a polymer-based composite can be improved by mixing heavy elements which have a higher density of electrons. Because the interaction of photons with materials usually involves electrons, heavy elements are effective in absorbing or scattering gamma-rays. Tungsten is a well-known additive element in many composite shielding studies. Alavian and Tavakoli-Anbaran (2019) [5] analyzed the effect of various sizes and proportions of tungsten particles in a low-density PE (LDPE) matrix. It was found that as the particle sizes decreased, the gamma attenuation increased. This improvement was also observed with higher proportions of the tungsten. However, the effect of particle sizes is less than that of the proportions. A similar conclusion was drawn by Asgari et al. (2020) [6] who studied the effect of tungsten and bismuth in rubber composite. Although, significant effects were only observed at higher weight percentages of the heavy elements (45-60 %). Can et al. (2021) [7] proved that regardless of the type of tungsten compounds (pure, oxide, or carbide), higher contents of tungsten are effective across a wide range of gamma-ray energies. The base polyester was made into several samples of composite with different types of tungsten compounds.

Based on the literature review, the present study aims to develop a new composite that is effective against both neutrons and gamma-rays. The base material is made from polyethylene. Boron is added to improve its neutron absorption, and tungsten is introduced to absorb gamma-rays.

2. Methods and materials
The study involves attenuation experiments with a mixed neutron and gamma-ray source. All equipment was provided by Nuclear Malaysia and Nuclear Laboratory in Universiti Teknologi Malaysia (UTM).

2.1. Fabrication of composite shield
The raw materials were purchased in the form of polyethylene (PE) pellets and powders of pure boron and tungsten. The new composite material was fabricated using the melt mixing method based on the configuration shown in Table 1. Firstly, the PE pellets were melted in a Haake internal mixer at 150 °C. After two minutes, the boron and tungsten powders were added. Eight minutes later, the composite mix was extracted and transferred to the hot press machine. The composite was then pressed into a 15 cm × 15 cm × 0.2 cm mold at 180 °C. After it has cooled, it was cut into 3 cm × 3 cm slabs. For dose rate measurement, five slabs were stacked together to form a 1-cm thick shield slab as shown in Figure 1.

| Sample | Composition (%wt) |
|--------|-------------------|
| PE100  | PE: 100, B: -      |
| B25    | PE: 75, B: 25, W  |
| W25    | PE: 75, B: -      |
| BW32   | PE: 68, B: 16, W: 16 |

2.2. Attenuation experiment
The shielding experiment was carried out using a Californium-252 fission neutron source in UTM. At the time of the experiment, the source strength was calculated to be 171 kBq or 4.6 µCi. Since the source is relatively weak, the shield was positioned immediately beside the source and the detector was placed 5 cm away from the source as shown in Figure 2. The background radiation was measured before the experiment to obtain the true value of the dose rate measurements.
3. Results and discussion

3.1. Physical density of samples
Theoretical density has upper bound and lower bound values which were calculated using Equations (1) and (2) respectively [8]. Table 2 shows the comparison between the theoretical density and the actual density of all samples. The actual density was obtained by measuring the weight of each sample using a digital balance and dividing the weight by the volume of the sample. It can be observed that the B25 and W25 composites have an actual density that is within the range of the theoretical density. However, BW32 is shown to be lower than its theoretical density. This might be due to the way the boron and tungsten atoms are arranged inside the PE matrix, reducing the amount of matter per cubic cm of the composite as illustrated in Figure 3.

\[ \rho_C = \rho_{PE} w_{PE} + \rho_B w_B + \rho_W w_W \]  
\[ \rho_C = \frac{1}{\frac{w_{PE}}{\rho_{PE}} + \frac{w_B}{\rho_B} + \frac{w_W}{\rho_W}} \]  

where \( \rho_C \) is the density of composite (g/cm\(^3\)) and \( \rho_{PE} \) is the density of polyethylene (0.86 g/cm\(^3\)).
$\rho_B$ density of boron (2.34 g/cm$^3$)
$\rho_W$ density of tungsten (19.25 g/cm$^3$)
$w_{PE}$ weight fractions of polyethylene
$w_B$ weight fractions of boron
$w_W$ weight fractions of tungsten

### Table 2. Densities of samples

| Sample | Theoretical density (g/cm$^3$) | Measured density (g/cm$^3$) |
|--------|-------------------------------|-----------------------------|
|        | Lower bound | Upper bound |                  |
| PE100  | -            | -            | 0.86              |
| B25    | 1.02         | 1.23         | 1.03              |
| W25    | 1.13         | 5.46         | 1.16              |
| BW32   | 1.15         | 4.04         | 1.00              |

**Figure 3.** When tungsten atoms are added into the matrix of polyboron (a), the amount of matter in the same given volume decreases as shown in (b). Note that this is an oversimplification since polyethylene molecule is a long chain of $(C_2H_4)_n$ monomers.

### 3.2. Shielding performance of samples

Figure 4 shows the transmission ratio for all samples. In terms of neutron shielding, B25 is the best which might be due to its high boron content. However, at 5 cm, there is an increase in the neutron dose rate from B25. This might be an effect of buildup in which the neutrons are reflected by the shield atoms towards the detector. The second best is the pure polyethylene PE100. This might be because it has the highest hydrogen content among all samples which are very effective in scattering neutrons. This is also the reason why the new composite BW32 is the worst neutron shield. It has the lowest hydrogen content which affected its capability in slowing down the neutrons. Therefore, its resulting neutron dose rates are higher than the other samples. Nevertheless, all samples show a decrease in their transmission ratio when their thickness increases.

Table 3 shows the comparison of the neutron shielding performance between the composites in this study and previous studies. The removal cross-sections $\Sigma_R$ are calculated using Equation 3, while the mass removal cross-sections are obtained by dividing $\Sigma_R$ by density. B25 has the highest mass removal cross-section than the past studies due to having the highest boron content of 25% by weight. W25 is expectedly worse than the past studies due to having no boron content. This is also the case with the new BW32. Although it has more boron than the past studies, it has less hydrogen content due to the additional content of tungsten. This lessens the effect of neutron moderation, resulting in higher neutron dose rates. Nevertheless, this new composite has good mixed radiation shielding performance as discussed later.
\[
\Sigma_R = \frac{1}{x} \ln \frac{D_0}{D}
\]

where
- \(\Sigma_R\): fast neutron removal cross-section \((\text{cm}^{-1})\)
- \(x\): shield thickness (cm)
- \(D_0\): initial neutron dose rates \((\text{mSv hr}^{-1})\)
- \(D\): final neutron dose rates \((\text{mSv hr}^{-1})\)

**Table 3. Comparison of composite neutron shielding performances**

| Author                               | Composition      | B (wt%) | \(\rho\) (g/cm\(^3\)) | \(\Sigma_R\) (cm\(^{-1}\)) | \(\Sigma_R\rho\) (cm\(^2\)/g) |
|--------------------------------------|------------------|---------|-------------------------|-----------------------------|-------------------------------|
| Sazali et al. (current)              | PE100            | 0       | 0.86                    | 0.1069                      | 0.1243                        |
|                                      | B25              | 25      | 1.03                    | 0.1414                      | 0.3733                        |
|                                      | W25              | 0       | 1.16                    | 0.0865                      | 0.0746                        |
|                                      | BW32             | 16      | 1.00                    | 0.0520                      | 0.0520                        |
| Uddin et al. (2020)                  | HDPE+B           | 10      | 1.03                    | 0.0981                      | 0.0948                        |
| Abd Elwahab et al. (2019)            | HDPE+Borax       | 7.72    | 2.08                    | 0.1570                      | 0.0754                        |
| El-Khayatt (2010)                    | PE+B             | 8.97    | 1.60                    | 0.0953                      | 0.0596                        |

In Figure 4(b), all samples exhibit a decrease in gamma dose rates as their thickness increases, except W25 which shows a slight increase in transmission from 1 cm to 3 cm. This might be due to the inelastic scattering of neutrons and neutron capture by tungsten atoms \((E_\gamma = 0.13, 0.48, \text{and} 0.69 \text{ MeV})\). It is expected to be the best gamma shield due to it having the highest tungsten additive. However, this is only observed at 1 cm thickness. At greater thicknesses, it is outperformed by B25 and BW32. B25 shows a steady decline in gamma-ray transmission. BW32 has the lowest gamma dose rates at 2-4 cm except at 1 cm and 5 cm. These gamma shielding performances might have been affected by the secondary gamma-rays generated from the scattering of neutrons and neutron capture by hydrogen \((E_\gamma = 2.2 \text{ MeV})\), boron \((E_\gamma = 0.48 \text{ MeV})\), and tungsten. The worst gamma shield was PE100. Even though it has good neutron attenuation as seen in Figure 4(a), it is not able to absorb the gamma-rays effectively without any additives.

Finally, in terms of mixed radiation attenuation, the new BW32 is the best among all samples as shown in Figure 5. Although it does not have the best neutron attenuation (Figure 4(a)), its combined neutron and gamma-ray transmission ratio is the lowest. This proves the effectiveness of combining various elements to improve the base material. In Figure 5, polyethylene by itself has poor mixed radiation performance as compared to others. By adding boron and tungsten, the transmitted radiations of both neutrons and gamma-rays can be lowered.
4. Conclusion

Four samples of shielding are fabricated and tested through an attenuation experiment with a mixed neutron and gamma-ray source, $^{252}$Cf. The first sample PE100 is pure polyethylene and it is used as the control to observe the effect of the additives in the composite samples. PE100 shows good neutron attenuation but has the worst gamma-ray attenuation. The second sample is B25, a polyethylene-based composite with 25% by mass of boron. It has the best neutron attenuation but at 5 cm, the transmitted dose rate increases due to the buildup effect. Its gamma transmission decreases almost linearly with

![Transmission ratio per shield thickness for neutron (a) and gamma-rays (b).](image1)

![Transmission ratio per shield thickness for neutron + gamma-rays.](image2)
thickness. The third sample W25 is also a composite with a PE base and tungsten powder (25% by mass). It is slightly worse than PE100 in terms of neutron shielding due to lower hydrogen content. Contrary to expectation, W25 does not exhibit the best gamma attenuation, and the gamma dose rates even increase slightly at thicknesses of 1-3 cm probably due to inelastic scattering of neutrons. Finally, BW32 shows the best mixed neutron and gamma-ray attenuation, proving the advantage of combining various elements to improve the shielding capabilities of the base material. Comparisons to previous studies are also provided. Future researchers may expand this study by investigating and improving its structural strength or other relevant properties. The methodology can be improved by using radiation sources of higher activities and using narrow beam geometry to identify the buildup factor.

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