


## Article

# Comparative X-ray Shielding Properties of Single-Layered and Multi-Layered Bi<sub>2</sub>O<sub>3</sub>/NR Composites: Simulation and Numerical Studies

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**Abstract:** This work theoretically compared the X-ray attenuation capabilities in natural rubber (NR) composites containing bismuth oxide (Bi<sub>2</sub>O<sub>3</sub>) by determining the effects of multi-layered structures on the shielding properties of the composites using two different software packages (XCOM and PHITS). The shielding properties of the single-layered and multi-layered Bi<sub>2</sub>O<sub>3</sub>/NR composites investigated consisted of the transmission factor ( $I/I_0$ ), effective linear attenuation coefficient ( $\mu_{\text{eff}}$ ), effective mass attenuation coefficient ( $\mu_{\text{m,eff}}$ ), and effective half-value layer ( $\text{HVL}_{\text{eff}}$ ). The results, with good agreement between those obtained from XCOM and PHITS (with less than 5% differences), indicated that the three-layered NR composites (sample#4), with the layer arrangement of pristine NR (layer#1)-Bi<sub>2</sub>O<sub>3</sub>/NR (layer#2)-pristine NR (layer#3), had relatively higher X-ray shielding properties than either a single-layer or the other multi-layered structures for all X-ray energies investigated (50, 100, 150, and 200 keV) due to its relatively larger effective percentage by weight of Bi<sub>2</sub>O<sub>3</sub> in the composites. Furthermore, by varying the Bi<sub>2</sub>O<sub>3</sub> contents in the middle layer (layer#2) of sample#4 from 10 to 90 wt.%, the results revealed that the overall X-ray shielding properties of the NR composites were further enhanced with additional filler, as evidenced by the highest values of  $\mu_{\text{eff}}$  and  $\mu_{\text{m,eff}}$  and the lowest values of  $I/I_0$  and  $\text{HVL}_{\text{eff}}$  observed in the 90 wt.% Bi<sub>2</sub>O<sub>3</sub>/NR composites. In addition, the recommended Bi<sub>2</sub>O<sub>3</sub> contents for the actual production of three-layered Bi<sub>2</sub>O<sub>3</sub>/NR composites (the same layer structure as sample#4) were determined by finding the least Bi<sub>2</sub>O<sub>3</sub> content that enabled the sample to attenuate incident X-rays with equal efficiency to that of a 0.5-mm lead sheet (with an effective lead equivalence of 0.5 mmPb). The results suggested that the recommended Bi<sub>2</sub>O<sub>3</sub> contents in layer#2 were 82, 72, and 64 wt.% for the combined 6 mm, 9 mm, and 12 mm samples, respectively.

**Keywords:** natural rubber; Bi<sub>2</sub>O<sub>3</sub>; X-ray shielding; simulation; multi-layered structure



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## 1. Introduction

Since the discovery of X-rays in 1895 by Wilhelm Roentgen, various applications have relied heavily on the utilization of X-ray technologies, especially X-ray imaging and X-ray irradiation in medicine, industry, material characterization, security, the arts, foods, and agriculture [1–6]. Despite their great potential and usefulness, excessive exposure to X-rays could harmfully affect the health of users and the public, with various symptoms, including nausea, skin burn, diarrhea, permanent disability, cancer, and death, depending on the exposure dose and duration as well as the sex, health condition, and age of those

exposed [7,8]. Hence, to reduce and/or prevent the risks of excessive exposure to X-rays, a radiation safety principle, namely “As Low As Reasonably Achievable” or “ALARA”, must be strictly followed in all nuclear facilities to ensure the safety of all users and the public [9].

One of the three safety measures in ALARA is the utilization of sufficient and appropriate shielding equipment; for which different applications may require different types and specific properties from the materials [10]. For example, X-ray shielding materials based on polyethylene (PE), including  $\text{Gd}_2\text{O}_3/\text{HDPE}$  and nano- $\text{ZnO}/\text{HDPE}$  composites, are suitable for applications that require exceptional strength and rigidity, such as those involving products for use as movable panels, walls, and construction parts in nuclear facilities [11,12]. On the other hand, shielding equipment, such as personal protective equipment (PPE) and covers for transporting casks, requiring exceptional flexibility, high strength, and a large amount of elongation from the materials, relies on natural and synthetic rubber composites. For example,  $\text{Bi}_2\text{O}_3/\text{NR}$ ,  $\text{Bi}_2\text{O}_3/\text{EPDM}$ ,  $\text{BaSO}_4/\text{EPDM}$ , and  $\text{W}/\text{SR}$  composites were among recently developed X-ray shielding rubber materials that offered not only effective X-ray attenuation abilities but also sufficient mechanical strength and flexibility to the users [13–16]. Notably, these mentioned examples of X-ray shielding materials are lead-free, which is presently sought-after in materials, as they could substantially reduce the risks to users from exposure to highly toxic lead (Pb) elements and compounds that are common protective fillers used for the manufacturing of X-ray and gamma shielding materials due to their economical accessibility and excellent attenuation capability [17,18].

Generally, the addition of heavy metals, including  $\text{Bi}_2\text{O}_3$ , to the main matrix is a common method to enhance the X-ray attenuation abilities of the composites, mainly due to the relatively high atomic number ( $Z$ ) and density ( $\rho$ ) of  $\text{Bi}_2\text{O}_3$  that enhance the interaction probabilities between the incident X-rays and the materials, subsequently increasing the ability to attenuate the incident X-rays of the composites [19]. Some examples showing the effects of  $\text{Bi}_2\text{O}_3$  on improving the shielding capabilities of the composites have been reported by Intom et al., who showed that the mass attenuation coefficients ( $\mu_m$ ) of  $\text{Bi}_2\text{O}_3/\text{NR}$  composites increased from 0.1324 to 0.3847 and then to 0.4779  $\text{cm}^2/\text{g}$  when the  $\text{Bi}_2\text{O}_3$  contents in the NR composites increased from 0 to 80 and then to 150 parts per hundred parts of rubber by weight (phr), respectively (determined at an energy level of 223 keV) [20]. Similarly, the report from Toyen et al. suggested that increases in the  $\text{Bi}_2\text{O}_3$  contents from 0 to 300 and then to 500 phr increased the linear attenuation coefficients ( $\mu$ ) of NR composites from 2.1 to 14.7 and then to 20.4  $\text{m}^{-1}$ , respectively (determined at an energy level of 662 keV) [13].

Nonetheless, despite the positive relationship between the contents of  $\text{Bi}_2\text{O}_3$  and the shielding properties of the composites, increases in  $\text{Bi}_2\text{O}_3$  contents may lead to undesirable reductions in the mechanical properties, such as decreased values of the tensile strength and elongation at the break of  $\text{Bi}_2\text{O}_3/\text{NR}$  composites from 14 to 7 MPa and from 630% to 500%, respectively, when the  $\text{Bi}_2\text{O}_3$  contents increase from 100 to 500 phr [13]. This behavior was observed mainly due to particle agglomerations caused by filler–filler interactions and phase separation at higher filler contents [14,21]. To alleviate or limit such drawbacks by adding high filler contents to the composites, one possible method is to prepare the materials with multi-layered structures, which would enable the pristine NR layers to better support and transfer external forces exerted on the  $\text{Bi}_2\text{O}_3/\text{NR}$  layers, consequently limiting the reduction in the overall strength of the materials [22,23].

As aforementioned, due to the competing roles of  $\text{Bi}_2\text{O}_3$  in the enhancement of X-ray-shielding properties and the reductions in mechanical properties, this work investigated appropriate multi-layered structures of  $\text{Bi}_2\text{O}_3/\text{NR}$  composites by theoretically comparing X-ray shielding parameters, consisting of the transmission factor ( $I/I_0$ ), the effective mass attenuation coefficient ( $\mu_{\text{eff}}$ ), the effective linear attenuation coefficient ( $\mu_{\text{m,eff}}$ ), the effective half-value layer ( $\text{HVL}_{\text{eff}}$ ), and the effective lead equivalence ( $\text{Pb}_{\text{eq,eff}}$ ), from 11 distinct multi-layered structures using XCOM and PHITS. In addition, the recommended  $\text{Bi}_2\text{O}_3$  contents for the multi-layered structure that produced the highest shielding properties

were also determined by finding the least  $\text{Bi}_2\text{O}_3$  contents that, when being added to the NR composites, produced the required  $\text{Pb}_{\text{eq,eff}}$  value of 0.5 mmPb. The outcomes of this work would not only provide comparative X-ray shielding properties of multi-layered products but also present promising methods to preserve the mechanical properties of shielding materials containing high contents of fillers.

## 2. Determination of X-ray Shielding Properties Using XCOM and PHITS

### 2.1. Multi-Layered Structures of $\text{Bi}_2\text{O}_3/\text{NR}$ Composites

The details and schemes of 11 distinct multi-layered structures for  $\text{Bi}_2\text{O}_3/\text{NR}$  composites with varying numbers (1–5) of layers and varying  $\text{Bi}_2\text{O}_3$  contents for each layer are shown in Table 1 and Figure 1, respectively. In order to simplify the setups for the determination of X-ray shielding properties, all samples would have the same average weight contents per thickness, i.e.,  $\sum C_i x_i / \sum x_i$  where  $C_i$  and  $x_i$  are  $\text{Bi}_2\text{O}_3$  content and thickness of the  $i$ th layer, respectively. Notably, for Figure 1, the left surface of each design was the side that faced the incident X-rays.

**Table 1.** Sample codes with details of the number of layers, thickness of each layer, and  $\text{Bi}_2\text{O}_3$  content in each layer for determination of X-ray shielding properties in  $\text{Bi}_2\text{O}_3/\text{NR}$  composites (Sample# and Layer# denote Sample Number and Layer Number, respectively).

Sample#	Number of Layers	Thickness of Each Layer (mm)	$\text{Bi}_2\text{O}_3$ Contents in Layer# (wt.%)				
			1	2	3	4	5
1	1	6.0	10	-	-	-	-
2	2	3.0	0	20	-	-	-
3	2	3.0	20	0	-	-	-
4	3	2.0	0	30	0	-	-
5	3	2.0	15	0	15	-	-
6	4	1.5	20	0	20	0	-
7	4	1.5	0	20	0	20	-
8	4	1.5	0	20	20	0	-
9	4	1.5	20	0	0	20	-
10	5	1.2	16.7	0	16.7	0	16.7
11	5	1.2	0	25	0	25	0

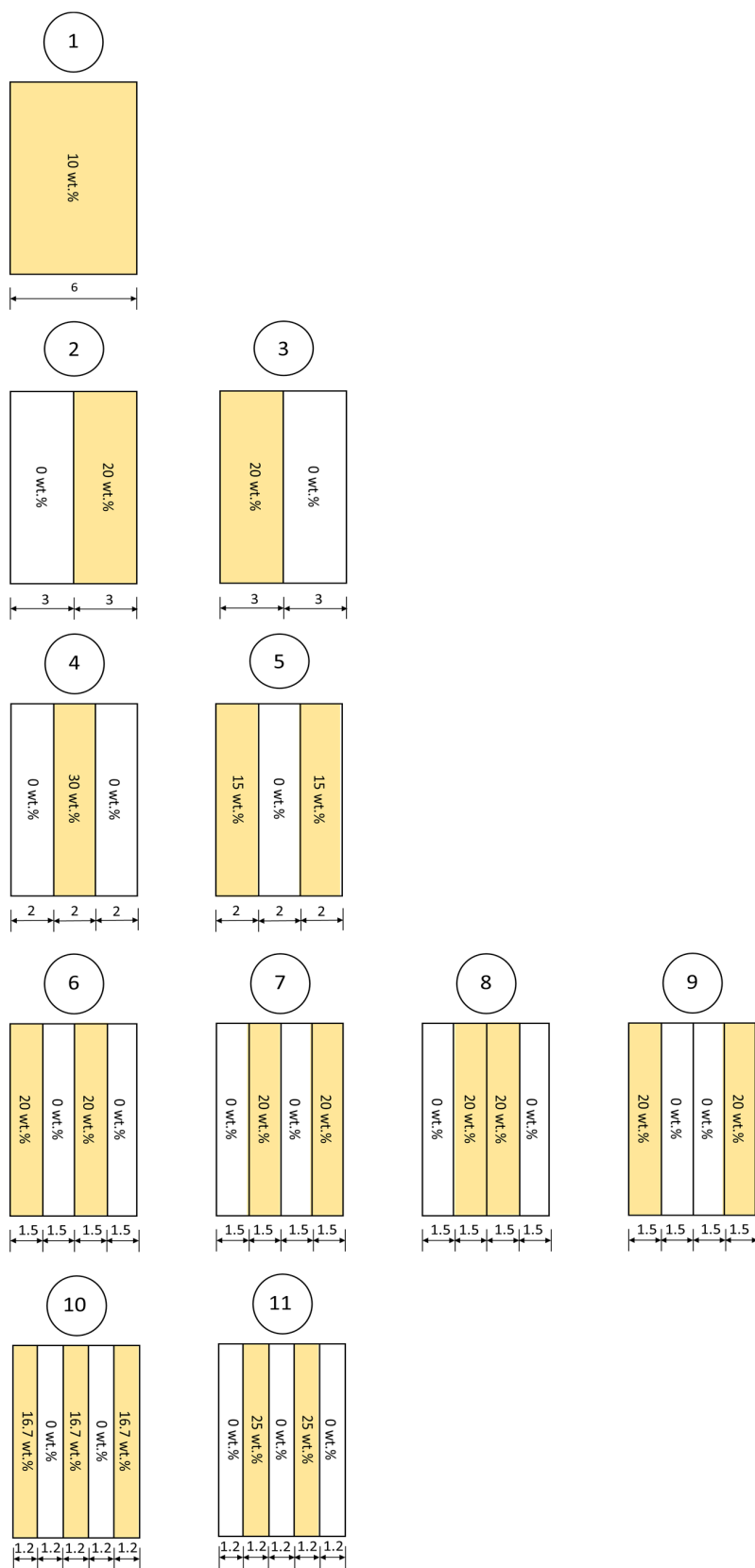
### 2.2. Determination of X-ray Shielding Properties Using XCOM

The X-ray shielding properties of all 11 multi-layered structures at the X-ray energies of 50, 100, 150, and 200 keV were numerically determined using the web-based XCOM software, provided by the National Institute of Standards and Technology (NIST) (Gaithersburg, MD, USA) [24,25]. The NIST standard reference database 8 (XGAM), released in 2010, was used as the photon cross-section database in this work and the X-ray shielding parameters were calculated from the total attenuation with the inclusion of coherent scattering [26].

In order to obtain the final transmission factor ( $I/I_0$ ) for each design, the mass attenuation coefficient ( $\mu_m$ ) for the  $\text{Bi}_2\text{O}_3/\text{NR}$  composites containing varying  $\text{Bi}_2\text{O}_3$  contents of 0, 10, 15, 16.7, 20, 25, and 30 wt.% were determined using XCOM. The details of the procedure to input material parameters and contents have been described elsewhere [24]. Then, the linear attenuation coefficients ( $\mu$ ) for each corresponding  $\text{Bi}_2\text{O}_3$  content were determined using the obtained  $\mu_m$ , following Equation (1):

$$\mu = \mu_m \times \rho \quad (1)$$

where  $\rho$  is the density of the  $\text{Bi}_2\text{O}_3/\text{NR}$  composites containing varying  $\text{Bi}_2\text{O}_3$  contents of 0, 10, 15, 16.7, 20, 25, and 30 wt.%, theoretically calculated using Equation (2):



**Figure 1.** Schemes showing single-layered and multi-layered structures and  $\text{Bi}_2\text{O}_3$  contents for 11 distinct designs for determination of X-ray shielding properties in  $\text{Bi}_2\text{O}_3/\text{NR}$  composites, where thicknesses are in millimeters and the numbers enclosed in circles represent the sample#.

$$\rho = \frac{100}{\frac{C_{\text{NR}}}{\rho_{\text{NR}}} + \frac{C_{\text{Bi}_2\text{O}_3}}{\rho_{\text{Bi}_2\text{O}_3}}} \quad (2)$$

where  $\rho_{\text{NR}}$  ( $\rho_{\text{Bi}_2\text{O}_3}$ ) is the density of NR ( $\text{Bi}_2\text{O}_3$ ), which is  $0.92 \text{ g/cm}^3$  ( $8.90 \text{ g/cm}^3$ ), and  $C_{\text{NR}}$  ( $C_{\text{Bi}_2\text{O}_3}$ ) is the weight content of NR ( $\text{Bi}_2\text{O}_3$ ) in the composites. Notably,  $C_{\text{NR}} + C_{\text{Bi}_2\text{O}_3} = 100 \text{ wt.}\%$ .

The value of  $(I/I_0)_i$  for the  $i$ th layer was calculated from its corresponding  $\mu$  using Equation (3):

$$\left(\frac{I}{I_0}\right)_i = e^{-\mu x_i} \quad (3)$$

where  $x_i$  is the thickness of the  $i$ th layer for each design shown in Table 1. Then, the final  $I/I_0$  value for each sample was calculated by multiplying individual  $(I/I_0)_i$  values from each layer, according to Equation (4):

$$\frac{I}{I_0} = \prod_{i=1}^n \left(\frac{I}{I_0}\right)_i \quad (4)$$

where  $n$  is the number of layers in the sample and  $i$  is  $1, 2, \dots, n$ .

Lastly, the effective linear attenuation coefficient ( $\mu_{\text{eff}}$ ), the effective mass attenuation coefficient ( $\mu_{\text{m,eff}}$ ), and the effective half-value layer ( $\text{HVL}_{\text{eff}}$ ), which represented the overall X-ray shielding properties for each design, were determined using Equations (5)–(7), respectively:

$$\mu_{\text{eff}} = \frac{-\ln\left(\frac{I}{I_0}\right)_i}{x_i} \quad (5)$$

$$\mu_{\text{m,eff}} = \frac{\mu_{\text{eff}}}{\rho_{\text{eff}}} \quad (6)$$

$$\text{HVL}_{\text{eff}} = \frac{\ln(2)}{\mu_{\text{eff}}} \quad (7)$$

where  $\rho_{\text{eff}}$  is the effective density of the sample, calculated using Equation (8):

$$\rho_{\text{eff}} = \frac{\sum_{i=1}^n \rho_i x_i}{\sum_{i=1}^n x_i} \quad (8)$$

where  $\rho_i$  and  $x_i$  are the density and the thickness of the  $i$ th layer, respectively. Notably, for further determination, the values of  $\mu$  for a pure Pb sheet at X-ray energies of 50, 100, 150, and 200 keV were also determined using XCOM. Furthermore, the effective percentage by weight ( $C_{\text{eff,Bi}_2\text{O}_3}$ ) of  $\text{Bi}_2\text{O}_3$  in different multi-layered samples (sample#2–sample#11) was also determined using Equation (9), which was derived from Equation (2):

$$\rho_{\text{eff}} = \frac{100}{\frac{100 - C_{\text{eff,Bi}_2\text{O}_3}}{\rho_{\text{NR}}} + \frac{C_{\text{eff,Bi}_2\text{O}_3}}{\rho_{\text{Bi}_2\text{O}_3}}} \quad (9)$$

### 2.3. Determination of X-ray Shielding Properties Using PHITS

In order to verify the X-ray shielding properties obtained using XCOM, the final  $I/I_0$  values were also determined for all multi-layered structures using PHITS by setting up the incident X-ray beam with a diameter of 1 mm pointing directly to the center of each sample, having a surface area of  $20 \text{ cm} \times 20 \text{ cm}$  and a combined thickness of 6 mm. This setup would minimize the possible overestimation of the final  $I/I_0$  value caused by build-up effects [27]. In addition, the detector with a 100% detection efficiency was set up to capture all primary transmitted X-rays. Further details of the PHITS setup are provided

elsewhere [10,11]. The percentages of difference (%Difference) between the final  $I/I_0$  values obtained from XCOM and those from PHITS were determined, following Equation (10):

$$\%Difference = \frac{\left| \left( \frac{I}{I_0} \right)_{XCOM} - \left( \frac{I}{I_0} \right)_{PHITS} \right|}{\left( \frac{I}{I_0} \right)_{XCOM}} \times 100\% \quad (10)$$

where  $(I/I_0)_{XCOM}$  and  $(I/I_0)_{PHITS}$  are the effective transmission factors of the  $\text{Bi}_2\text{O}_3/\text{NR}$  composites obtained from XCOM and PHITS, respectively.

#### 2.4. Determination of Effective Lead Equivalence and Recommended Contents of $\text{Bi}_2\text{O}_3$

The values of effective lead equivalence ( $\text{Pb}_{\text{eq,eff}}$ ) at X-ray energies of 50, 100, 150, and 200 keV for the multi-layered  $\text{Bi}_2\text{O}_3/\text{NR}$  composites offering the highest final  $I/I_0$  values among all 11 designs were calculated, following Equation (11):

$$\mu_{\text{Pb}} \text{Pb}_{\text{eq,eff}} = \mu_{\text{NR,eff}} x_{\text{NR}} \quad (11)$$

where  $\mu_{\text{Pb}}$  is the linear attenuation coefficient of a pure Pb sheet,  $\mu_{\text{NR,eff}}$  is the effective linear attenuation coefficient of multi-layered  $\text{Bi}_2\text{O}_3/\text{NR}$  composites, and  $x_{\text{NR}}$  is the combined thickness of the multi-layered  $\text{Bi}_2\text{O}_3/\text{NR}$  composites, which varied from 6 to 9 to 12 mm. Notably, the  $\text{Bi}_2\text{O}_3$  contents for the determination of  $\text{Pb}_{\text{eq,eff}}$  were varied up to the maximum content of 90 wt.% and the  $\mu_{\text{Pb}}$  values were 90.9, 62.7, 22.8, and  $1.13 \text{ cm}^{-1}$  at X-ray energies of 50, 100, 150, and 200 keV, respectively, determined using XCOM.

To determine the recommended  $\text{Bi}_2\text{O}_3$  contents, the values of  $\text{Pb}_{\text{eq,eff}}$  for all conditions obtained from the previous steps were plotted against their corresponding  $\text{Bi}_2\text{O}_3$  contents. Then, a horizontal straight line with a  $\text{Pb}_{\text{eq}}$  value of 0.5 mmPb (the common requirement for X-ray shielding equipment in general nuclear facilities) was plotted and the points of intersection were noted for each thickness (6, 9, and 12 mm), which represented the least  $\text{Bi}_2\text{O}_3$  contents providing the composites with a  $\text{Pb}_{\text{eq}}$  value of 0.5 mmPb, and could be regarded as the recommended  $\text{Bi}_2\text{O}_3$  contents for the actual production.

### 3. Results and Discussion

#### 3.1. Values of $\mu_m$ , $\mu$ , and $\rho$ for $\text{Bi}_2\text{O}_3/\text{NR}$ Composites

The values of the numerically determined  $\mu_m$ ,  $\rho$ , and  $\mu$  for the single-layered  $\text{Bi}_2\text{O}_3/\text{NR}$  composites with varying  $\text{Bi}_2\text{O}_3$  contents of 0, 10, 15, 16.7, 20, 25, or 30 wt.% at X-ray energies of 50, 100, 150, and 200 keV are shown in Tables 2–4, respectively. The results shown in Table 2 indicated that the values of  $\mu_m$  tended to increase with increasing  $\text{Bi}_2\text{O}_3$  content but decreased with increasing X-ray energy. The positive relationship between  $\mu_m$  and filler contents was mainly due to the high atomic number ( $Z$ ) of Bi and the much higher density ( $\rho$ ) of  $\text{Bi}_2\text{O}_3$  compared to those of NR, resulting in substantially enhanced interaction probabilities between the incident X-rays and the materials through the very effective and dominant X-ray interaction, namely photoelectric absorption, which subsequently improved the overall X-ray shielding properties of the composites with the addition of  $\text{Bi}_2\text{O}_3$ . The behavior could be mathematically explained by considering the relationship between the photoelectric cross-section ( $\sigma_{\text{pe}}$ ), atomic numbers ( $Z$ ) of elements in the composites, and the frequencies ( $\nu$ ) of incident X-rays, following Equation (12):

$$\sigma_{\text{pe}} = \frac{Z^n}{(h\nu)^3} \quad (12)$$

where  $h$  is Planck's constant [11].

**Table 2.** Mass attenuation coefficients ( $\mu_m$ ;  $\text{cm}^2/\text{g}$ ) of  $\text{Bi}_2\text{O}_3/\text{NR}$  composites with varying  $\text{Bi}_2\text{O}_3$  contents of 0, 10, 15, 16.7, 20, 25, and 30 wt.% at the X-ray energies of 50, 100, 150, and 200 keV.

X-ray Energy (keV)	$\text{Bi}_2\text{O}_3$ Content (wt.%)						
	0	10	15	16.7	20	25	30
50	0.2047	0.9379	1.3050	1.4290	1.6710	2.0380	2.4040
100	0.1683	0.6677	0.9174	1.0020	1.1670	1.4170	1.6670
150	0.1501	0.3233	0.4099	0.4393	0.4965	0.5831	0.6696
200	0.1371	0.2174	0.2575	0.2712	0.2976	0.3378	0.3779

**Table 3.** Calculated densities ( $\rho$ ) of  $\text{Bi}_2\text{O}_3/\text{NR}$  composites with varying  $\text{Bi}_2\text{O}_3$  contents of 0, 10, 15, 16.7, 20, 25, and 30 wt.%.

$\text{Bi}_2\text{O}_3$ Content (wt.%)	Density ( $\text{g}/\text{cm}^3$ )
0	0.920
10	1.011
15	1.063
16.7	1.082
20	1.121
25	1.186
30	1.259

**Table 4.** Linear attenuation coefficients ( $\mu$ ;  $\text{cm}^{-1}$ ) of single-layered  $\text{Bi}_2\text{O}_3/\text{NR}$  composites with varying  $\text{Bi}_2\text{O}_3$  contents of 0, 10, 15, 16.7, 20, 25, and 30 wt.% at X-ray energies of 50, 100, 150, and 200 keV.

X-ray Energy (keV)	$\text{Bi}_2\text{O}_3$ Content (wt.%)						
	0	10	15	16.7	20	25	30
50	0.1883	0.9478	1.3872	1.5457	1.8732	2.4167	3.0255
100	0.1548	0.6747	0.9751	1.0838	1.3082	1.6803	2.0979
150	0.1380	0.3267	0.4357	0.4751	0.5565	0.6914	0.8427
200	0.1261	0.2197	0.2737	0.2933	0.3336	0.4005	0.4755

Notably,  $\nu$  and the X-ray energy ( $E$ ) are directly proportional to each other as shown in Equation (13):

$$E = h\nu \quad (13)$$

Equations (12) and (13) also depict that the interaction probabilities between the incident X-rays and the materials are inversely proportional to  $\nu^3$  or  $E^3$ ; for which the results in Table 2 clearly illustrate this effect, as evidenced by the lowest  $\mu_m$  values being observed at the X-ray energy of 200 keV [28].

Table 3, which shows the calculated densities ( $\rho$ ) of a single-layered  $\text{Bi}_2\text{O}_3/\text{NR}$  composite with varying  $\text{Bi}_2\text{O}_3$  contents of 0, 10, 15, 16.7, 20, 25, and 30 wt.% that were used for the determination of the linear attenuation coefficient ( $\mu$ ), suggested that the density of the NR composites increased with increasing  $\text{Bi}_2\text{O}_3$  contents, which is mainly due to the much higher  $\rho$  value of  $\text{Bi}_2\text{O}_3$  ( $\rho_{\text{Bi}_2\text{O}_3} = 8.90 \text{ g}/\text{cm}^3$ ) than for NR ( $\rho_{\text{NR}} = 0.92 \text{ g}/\text{cm}^3$ ). Using the results shown in Tables 2 and 3, and Equation (1), the values of  $\mu$  for all the single-layered  $\text{Bi}_2\text{O}_3/\text{NR}$  composites with varying  $\text{Bi}_2\text{O}_3$  contents of 0, 10, 15, 16.7, 20, 25, and 30 wt.% were determined, and the results are shown in Table 4, which indicates similar behavior as for  $\mu_m$  (Table 2). However, more pronounced effects of  $\text{Bi}_2\text{O}_3$  on the enhancement of  $\mu$  were observed compared to those for  $\mu_m$  due to the simultaneous roles of  $\text{Bi}_2\text{O}_3$  in increasing both the  $\mu_m$  and  $\rho$  values of the composites, which further amplified the values of  $\mu$  at higher  $\text{Bi}_2\text{O}_3$  contents (Equation (1)).

### 3.2. Final $I/I_0$ of Multi-Layered $\text{Bi}_2\text{O}_3/\text{NR}$ Composites

Tables 5–8 show the transmission factors ( $I/I_0$ ) for each layer as well as the final  $I/I_0$  values of the 11 multi-layered  $\text{Bi}_2\text{O}_3/\text{NR}$  composites at X-ray energies of 50, 100, 150, and 200 keV, respectively, and Figure 2 shows the schematic representation of relative X-ray intensities for each layer of some designs at the X-ray energy of 50 keV. All the results suggested that the NR layers containing  $\text{Bi}_2\text{O}_3$  could attenuate X-rays with higher efficiencies than those without  $\text{Bi}_2\text{O}_3$  due to the much higher  $\mu$  values of  $\text{Bi}_2\text{O}_3/\text{NR}$  composites (Table 4), especially those with higher  $\text{Bi}_2\text{O}_3$  contents, that better interacted and attenuated incident X-rays. Furthermore, the results revealed that the final  $I/I_0$  values for the composites had larger transmitted X-ray intensities at higher X-ray energies (for the same sample#). This behavior could be explained using Equation (12), which suggested that the interaction probabilities, as well as their X-ray attenuation capabilities, decreased with increasing X-ray energies, resulting in more X-rays being able to escape the materials.

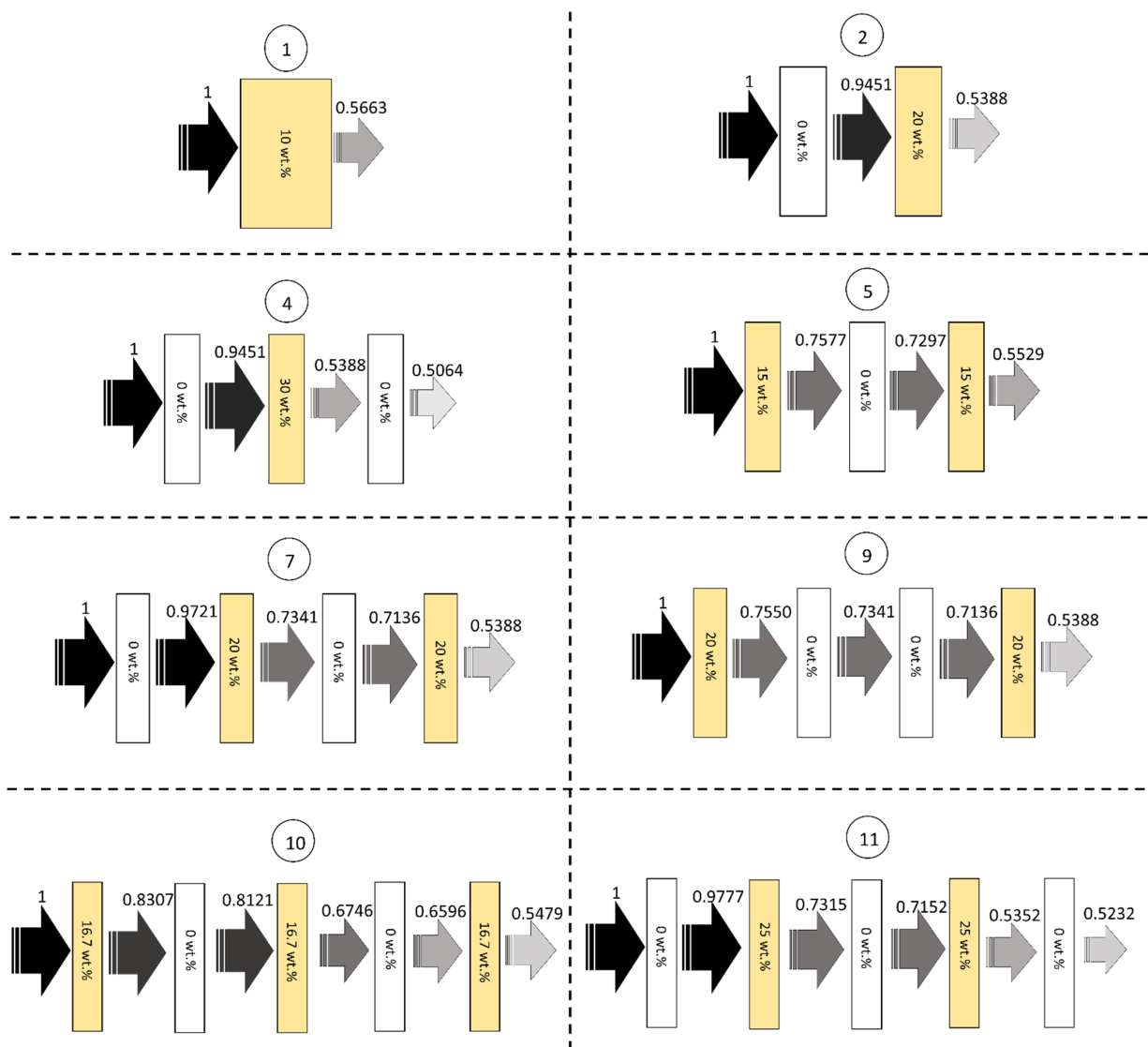
**Table 5.** Relative X-ray intensities for each layer of multi-layered  $\text{Bi}_2\text{O}_3/\text{NR}$  composites at an X-ray energy of 50 keV (Sample# and Layer# denote Sample Number and Layer Number, respectively).

Sample#	Number of Layers	Thickness of Each Layer (mm)	Relative X-ray Intensities for Layer#				
			1	2	3	4	5
1	1	6.0	0.5663	-	-	-	-
2	2	3.0	0.9451	0.5388	-	-	-
3	2	3.0	0.5701	0.5388	-	-	-
4	3	2.0	0.9630	0.5258	0.5064	-	-
5	3	2.0	0.7577	0.7298	0.5529	-	-
6	4	1.5	0.7550	0.7340	0.5542	0.5388	-
7	4	1.5	0.9722	0.7340	0.7136	0.5388	-
8	4	1.5	0.9722	0.7340	0.5542	0.5388	-
9	4	1.5	0.7550	0.7340	0.7136	0.5388	-
10	5	1.2	0.8307	0.8121	0.6746	0.6596	0.5479
11	5	1.2	0.9777	0.7315	0.7152	0.5352	0.5232

**Table 6.** Relative X-ray intensities for each layer of multi-layered  $\text{Bi}_2\text{O}_3/\text{NR}$  composites at an X-ray energy of 100 keV (Sample# and Layer# denote Sample Number and Layer Number, respectively).

Sample#	Number of Layers	Thickness of Each Layer (mm)	Relative X-ray Intensities for Layer#				
			1	2	3	4	5
1	1	6.0	0.6671	-	-	-	-
2	2	3.0	0.9546	0.6447	-	-	-
3	2	3.0	0.6754	0.6447	-	-	-
4	3	2.0	0.9695	0.6373	0.6178	-	-
5	3	2.0	0.8228	0.7977	0.6564	-	-
6	4	1.5	0.8218	0.8030	0.6599	0.6447	-
7	4	1.5	0.9770	0.8030	0.7845	0.6447	-
8	4	1.5	0.9770	0.8030	0.6599	0.6447	-
9	4	1.5	0.8218	0.8030	0.7845	0.6447	-
10	5	1.2	0.8780	0.8619	0.7568	0.7428	0.6522
11	5	1.2	0.9816	0.8023	0.7876	0.6438	0.6319





**Figure 2.** Schemes showing relative X-ray intensities for each layer of sample#1, sample#2, sample#4, sample#5, sample#7, sample#9, sample#10, and sample#11, at the X-ray energy of 50 keV. The numbers enclosed in circles represent sample#.

**Table 7.** Relative X-ray intensities for each layer of multi-layered Bi<sub>2</sub>O<sub>3</sub>/NR composites at an X-ray energy of 150 keV (Sample# and Layer# denote Sample Number and Layer Number, respectively).

Sample#	Number of Layers	Thickness of Each Layer (mm)	Relative X-ray Intensities for Layer#				
			1	2	3	4	5
1	1	6.0	0.8220	-	-	-	-
2	2	3.0	0.9594	0.8119	-	-	-
3	2	3.0	0.8462	0.8119	-	-	-
4	3	2.0	0.9728	0.8219	0.7995	-	-
5	3	2.0	0.9165	0.8916	0.8172	-	-
6	4	1.5	0.9199	0.9010	0.8289	0.8119	-
7	4	1.5	0.9795	0.9010	0.8826	0.8119	-
8	4	1.5	0.9795	0.9010	0.8289	0.8119	-
9	4	1.5	0.9199	0.9010	0.8826	0.8119	-
10	5	1.2	0.9446	0.9291	0.8776	0.8631	0.8153
11	5	1.2	0.9836	0.9052	0.8904	0.8195	0.8060

**Table 8.** Relative X-ray intensities for each layer of multi-layered Bi<sub>2</sub>O<sub>3</sub>/NR composites at an X-ray energy of 200 keV (Sample# and Layer# denote Sample Number and Layer Number, respectively).

Sample#	Number of Layers	Thickness of Each Layer (mm)	Relative X-ray intensities for Layer#				
			1	2	3	4	5
1	1	6.0	0.8765	-	-	-	-
2	2	3.0	0.9629	0.8712	-	-	-
3	2	3.0	0.9048	0.8712	-	-	-
4	3	2.0	0.9751	0.8866	0.8645	-	-
5	3	2.0	0.9467	0.9231	0.8740	-	-
6	4	1.5	0.9512	0.9334	0.8878	0.8712	-
7	4	1.5	0.9813	0.9334	0.9159	0.8712	-
8	4	1.5	0.9813	0.9334	0.8878	0.8712	-
9	4	1.5	0.9512	0.9334	0.9159	0.8712	-
10	5	1.2	0.9654	0.9509	0.9180	0.9042	0.8729
11	5	1.2	0.9850	0.9388	0.9247	0.8813	0.8680

Among the 11 multi-layered designs, sample#4, which has a three-layered structure, had the lowest final  $I/I_0$  values of 0.5064, 0.6178, 0.7995, and 0.8645 at X-ray energies of 50, 100, 150, and 200 keV, respectively, while sample#1, a single-layered structure, had the highest final  $I/I_0$  values of 0.5663, 0.6671, 0.8220, and 0.8765 at X-ray energies of 50, 100, 150, and 200 keV, respectively. Based on the results from these two designs, the multi-layered structure exhibited higher X-ray shielding capabilities by as much as 10.5, 8.7, 4.0, and 2.1% compared to a single-layered structure, determined at X-ray energies of 50, 100, 150, and 200 keV, respectively. Specifically, for sample#4, its highest X-ray attenuation capability was due to its highest effective density and effective percentage by weight of Bi<sub>2</sub>O<sub>3</sub> contained in the sample, determined using Equations (8) and (9); for which the results of both parameters for all designs are shown in Table 9. The larger values of both quantities in multi-layered structures were mainly due to the much higher density of Bi<sub>2</sub>O<sub>3</sub> particles in comparison with that of the NR matrix (for instance, adding 20 wt.% of Bi<sub>2</sub>O<sub>3</sub> to layer#2 in sample#4 would require much less volume than removing 20 wt.% of NR, resulting in a considerable reduction in the total volume and subsequently the increase in the density of the sample). These effects then enabled sample#4 to have more Bi atoms to interact with incoming X-rays through the photoelectric absorption than that of sample#1.

**Table 9.** Effective densities and effective percentages by weight of Bi<sub>2</sub>O<sub>3</sub> for all 11 multi-layered Bi<sub>2</sub>O<sub>3</sub>/NR composites (Sample# denotes Sample Number).

Sample#	Effective Density (g/cm <sup>3</sup> )	Effective Percentage by Weight (wt.%)
1	1.011	10.00
2	1.021	10.98
3	1.021	10.98
4	1.033	12.19
5	1.015	10.47
6	1.021	10.98
7	1.021	10.98
8	1.021	10.98
9	1.021	10.98
10	1.017	10.64
11	1.026	11.55

In addition, Equation (3) could be modified for the calculation of  $I/I_0$  as Equation (14):

$$\frac{I}{I_0} = e^{-\sum_i^N \mu_i x_i} \quad (14)$$

where  $\mu_i$  is the linear attenuation coefficient of the  $i$ th layer,  $x_i$  is the thickness of the  $i$ th layer, and  $N$  is the total number of layers in the composites [29], which depicted that the values of  $\sum_i^N \mu_i x_i$  for the multi-layered structures (using information from Tables 1 and 4) were larger than that of the single-layered sample. For instance, sample#4 had the value of  $\sum_i^N \mu_i x_i$  of 0.6804, while sample#1 had the value of 0.5687, leading to a lower  $I/I_0$  and better X-ray shielding capabilities in sample#4. Furthermore, the results showed that rearranging layers of the samples having the same  $\text{Bi}_2\text{O}_3$  contents and numbers of layers did not have effects on X-ray shielding capabilities. For instance, sample #2 and sample #3, as well as samples #6–#9, had the same values of  $I/I_0$ , regardless of how the layers were arranged. This was due to the values of  $\sum_i^N \mu_i x_i$  being the same for all of them.

Table 10 shows the final  $I/I_0$  values of all 11 multi-layered structures using XCOM and PHITS, as well as their corresponding %Difference values for these two methods. The comparisons indicated that the results obtained from both methods were in good agreement, with the largest %Difference value being 4.78% and the average %Difference being 2.24%. Consequently, the values obtained from XCOM and PHITS could be further used for the determination of other parameters, including  $\mu_{m,eff}$ ,  $\mu_{eff}$ ,  $\text{HVL}_{eff}$ , and  $\text{Pb}_{eq,eff}$ . Another interesting outcome from Table 9 was that the final  $I/I_0$  values from PHITS seemed to be slightly higher than those from XCOM. This could have been due to factors, such as backscattering and the rescattering of X-rays inside the materials, resulting in an increase in the transmitted X-rays and a subsequent underestimation of the theoretical or ideal X-ray attenuation capabilities of the composites in the results obtained from PHITS [30].

**Table 10.** Comparative final transmission factors ( $I/I_0$ ) of 11 multi-layered structures of  $\text{Bi}_2\text{O}_3/\text{NR}$  composites at X-ray energies of 50, 100, 150, and 200 keV using XCOM and PHITS and their corresponding percentage differences (Sample# denotes Sample Number).

Sample#	XCOM				PHITS				%Difference			
	Final Transmission Factor ( $I/I_0$ ) at X-ray Energy (keV)											
	50	100	150	200	50	100	150	200	50	100	150	200
1	0.5663	0.6671	0.8220	0.8765	0.5732	0.6681	0.8258	0.8785	1.22	0.15	0.46	0.23
2	0.5388	0.6447	0.8119	0.8712	0.5518	0.6483	0.8498	0.8964	2.42	0.55	4.67	2.89
3	0.5388	0.6447	0.8119	0.8712	0.5578	0.6756	0.8196	0.8946	3.54	4.78	0.95	2.69
4	0.5064	0.6178	0.7995	0.8645	0.5289	0.6371	0.8255	0.8865	4.44	3.11	3.25	2.54
5	0.5529	0.6564	0.8172	0.8740	0.5766	0.6589	0.8443	0.8871	4.27	0.38	3.32	1.50
6	0.5388	0.6447	0.8119	0.8712	0.5428	0.6480	0.8226	0.8735	0.75	0.50	1.32	0.27
7	0.5388	0.6447	0.8119	0.8712	0.5494	0.6547	0.8432	0.8898	1.97	1.54	3.85	2.14
8	0.5388	0.6447	0.8119	0.8712	0.5471	0.6452	0.8461	0.8917	1.55	0.08	4.22	2.36
9	0.5388	0.6447	0.8119	0.8712	0.5596	0.6522	0.8480	0.8789	3.87	1.15	4.45	0.89
10	0.5479	0.6522	0.8153	0.8729	0.5550	0.6723	0.8523	0.8861	1.30	3.08	4.54	1.50
11	0.5232	0.6319	0.8060	0.8680	0.5297	0.6384	0.8343	0.9039	1.24	1.03	3.51	4.13

### 3.3. Values for $\mu_{eff}$ , $\mu_{m,eff}$ , and $\text{HVL}_{eff}$ of Multi-Layered $\text{Bi}_2\text{O}_3/\text{NR}$ Composites

Table 11 shows the values of  $\mu_{eff}$ ,  $\mu_{m,eff}$ , and  $\text{HVL}_{eff}$  for the 11 multi-layered  $\text{Bi}_2\text{O}_3/\text{NR}$  composites at X-ray energies of 50, 100, 150, and 200 keV, determined using Equations (5)–(7) and the effective densities ( $\rho_{eff}$ ) of the samples shown in Table 9. The results indicated that similar to those of the final  $I/I_0$  (Table 10), sample#4 had the most efficient X-ray shielding properties as well as  $\rho_{eff}$ , as evidenced by its highest values of  $\mu_{eff}$ ,  $\mu_{m,eff}$ , and  $\text{HVL}_{eff}$  compared to the other designs.

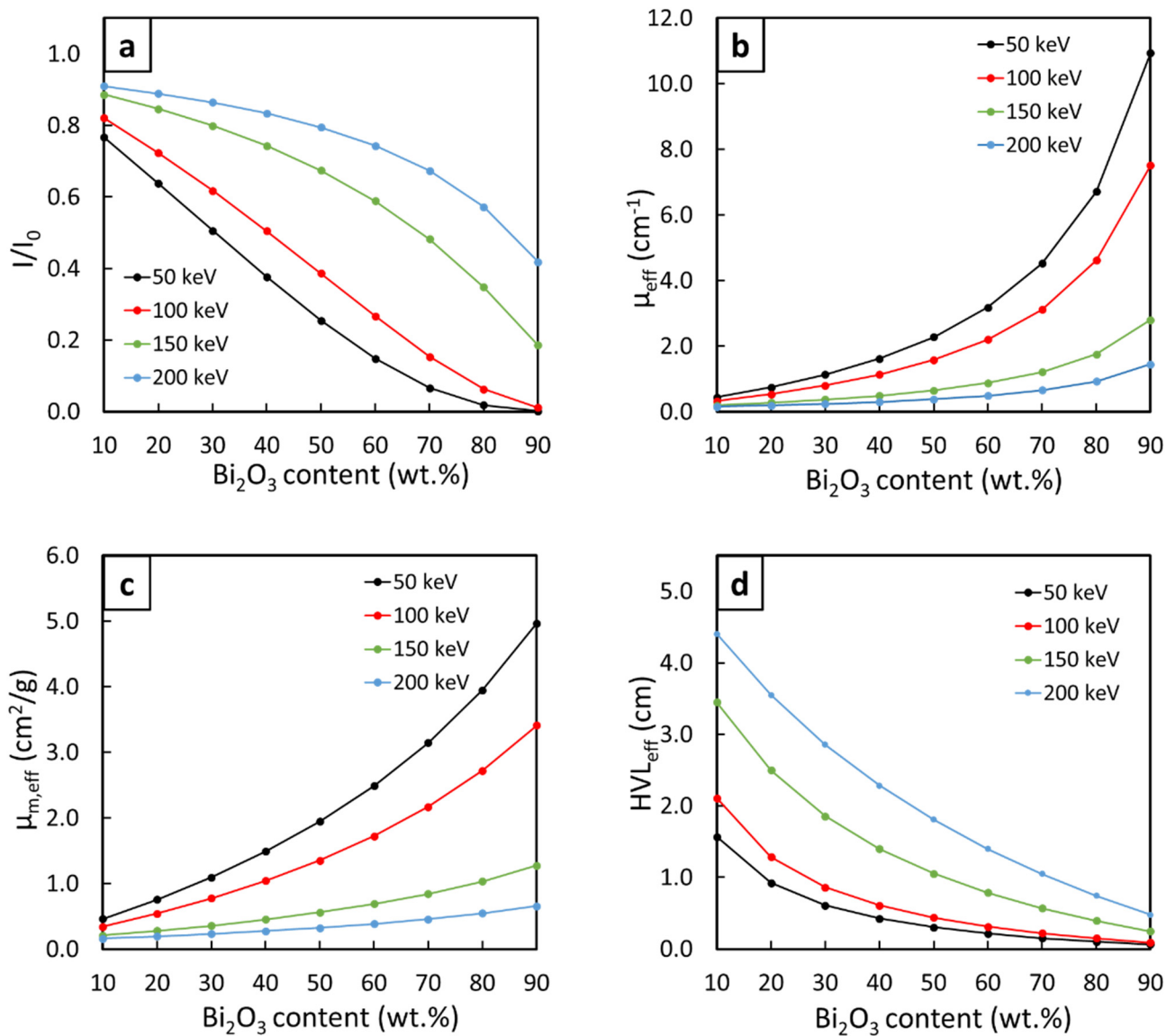
**Table 11.** Values for  $\mu_{\text{eff}}$ ,  $\mu_{\text{m,eff}}$ , and  $\text{HVL}_{\text{eff}}$  of 11 multi-layered  $\text{Bi}_2\text{O}_3/\text{NR}$  composites at X-ray energies of 50, 100, 150, and 200 keV (Sample# denotes Sample Number).

Sample#	$\mu_{\text{eff}}$ ( $\text{cm}^{-1}$ )				$\mu_{\text{m,eff}}$ ( $\text{cm}^2/\text{g}$ )				$\text{HVL}_{\text{eff}}$ (cm)			
	50 keV	100 keV	150 keV	200 keV	50 keV	100 keV	150 keV	200 keV	50 keV	100 keV	150 keV	200 keV
1	0.9479	0.6748	0.3267	0.2197	0.9379	0.7195	0.4541	0.4838	0.7313	1.0272	2.1215	3.1549
2	1.0308	0.7315	0.3473	0.2299	1.0101	0.7243	0.4796	0.4793	0.6724	0.9475	1.9956	3.0153
3	1.0308	0.7315	0.3473	0.2299	1.0101	0.7243	0.4796	0.4793	0.6724	0.9475	1.9956	3.0153
4	1.1341	0.8025	0.3730	0.2426	1.0980	0.7309	0.5103	0.4755	0.6112	0.8637	1.8585	2.8569
5	0.9876	0.7017	0.3365	0.2245	0.9727	0.7214	0.4664	0.4814	0.7019	0.9878	2.0599	3.0873
6	1.0308	0.7315	0.3473	0.2299	1.0101	0.7243	0.4796	0.4793	0.6724	0.9475	1.9956	3.0153
7	1.0308	0.7315	0.3473	0.2299	1.0101	0.7243	0.4796	0.4793	0.6724	0.9475	1.9956	3.0153
8	1.0308	0.7315	0.3473	0.2299	1.0101	0.7243	0.4796	0.4793	0.6724	0.9475	1.9956	3.0153
9	1.0308	0.7315	0.3473	0.2299	1.0101	0.7243	0.4796	0.4793	0.6724	0.9475	1.9956	3.0153
10	1.0028	0.7122	0.3403	0.2265	0.9860	0.7224	0.4712	0.4807	0.6912	0.9732	2.0366	3.0608
11	1.0797	0.7650	0.3594	0.2359	1.0520	0.7272	0.4943	0.4773	0.6420	0.9061	1.9284	2.9382

### 3.4. X-rays Shielding Properties and Recommended $\text{Bi}_2\text{O}_3$ Contents of Three-Layered $\text{Bi}_2\text{O}_3/\text{NR}$ Composites (Sample#4)

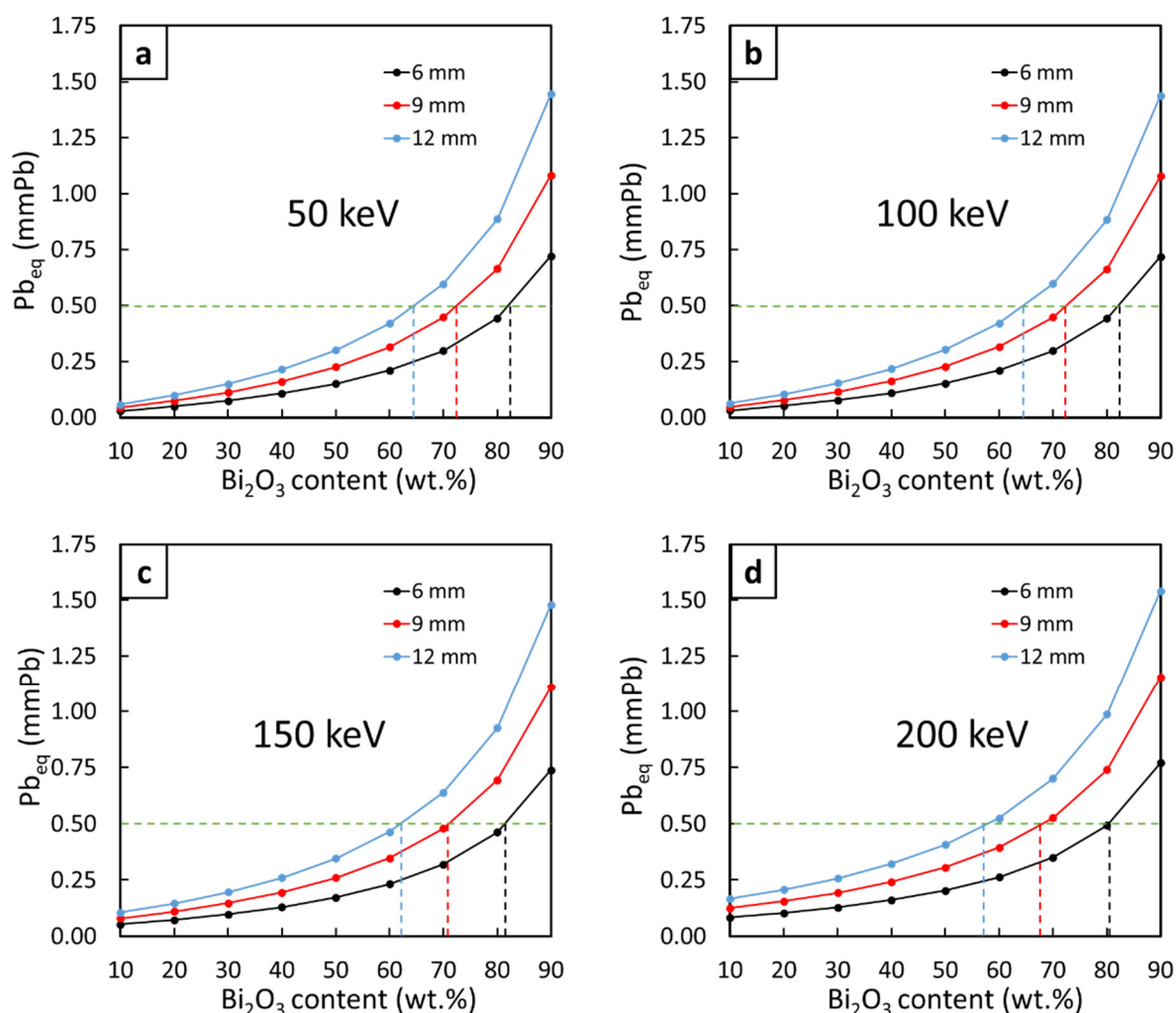
Figure 3 shows the values of the final  $I/I_0$ ,  $\mu_{\text{eff}}$ ,  $\mu_{\text{m,eff}}$ , and  $\text{HVL}_{\text{eff}}$  of the three-layered  $\text{Bi}_2\text{O}_3/\text{NR}$  composites (sample#4, which provided higher X-ray shielding properties compared to the other designs), with varying  $\text{Bi}_2\text{O}_3$  contents in layer#2 (middle layer) from 10 to 90 wt.% in 10 wt.% increments and a fixed combined thickness of 6 mm, determined at X-ray energies of 50, 100, 150, and 200 keV. The results indicated that the ability to attenuate incident X-rays greatly improved with increasing  $\text{Bi}_2\text{O}_3$  contents, as evidenced by the decreases in the values of  $I/I_0$  and  $\text{HVL}_{\text{eff}}$  and the increases in  $\mu_{\text{eff}}$  and  $\mu_{\text{m,eff}}$  with increasing contents. On the other hand, the overall shielding properties of the composites tended to decrease with increasing X-ray energy, as the lowest (highest) values of  $\mu_{\text{eff}}$  and  $\mu_{\text{m,eff}}$  ( $I/I_0$  and  $\text{HVL}_{\text{eff}}$ ) were observed at an X-ray energy of 200 keV. These two sets of behavior could be explained using Equation (12), which states that the photoelectric cross-section ( $\sigma_{\text{pe}}$ ) (the ability to attenuate X-rays) is directly proportional to  $Z^n$  while being inversely proportional to  $\nu^3$  ( $E^3$ ), resulting in enhanced (lower) shielding properties at higher filler contents (X-ray energies).

The  $\text{Pb}_{\text{eq,eff}}$  values of the three-layered  $\text{Bi}_2\text{O}_3/\text{NR}$  composites (sample#4) with varying  $\text{Bi}_2\text{O}_3$  contents in layer#2 (middle layer) from 10 to 90 wt.% in 10 wt.% increments and varying combined thicknesses of 6, 9, and 12 mm, are shown in Figure 4. The results indicated that the least  $\text{Bi}_2\text{O}_3$  contents in layer#2, which could be regarded as the recommended  $\text{Bi}_2\text{O}_3$  contents, that provided the three-layered NR composites with the required  $\text{Pb}_{\text{eq}}$  of 0.5 mmPb, were 82, 72, and 64 wt.% for the combined thicknesses of 6, 9, and 12 mm, respectively. The decreases in the recommended  $\text{Bi}_2\text{O}_3$  contents with thicker samples were due to more Bi atoms being available in thicker materials (with the same filler content) to interact with incident X-rays, subsequently reducing the required  $\text{Bi}_2\text{O}_3$  contents in layer#2. Notably, while it is possible to prepare NR composites with a 90 wt.% of fillers, as reported by Gwaily et al. who prepared Pb/NR composites for gamma shielding with the Pb contents up to 2000 phr (~95 wt.%) [31], difficulties in the sample preparation process, as well as possible substantial reductions in mechanical properties, could limit the processibility of multi-layered composites with very high filler contents. Consequently, for applications that allow space for thicker materials, lower recommended  $\text{Bi}_2\text{O}_3$  fillers, such as those in 9 mm and 12 mm samples, should be considered to ease the difficulty and preserve the mechanical properties and product flexibility.



**Figure 3.** (a) Final values of  $I/I_0$ , (b)  $\mu_{\text{eff}}$ , (c)  $\mu_{\text{m,eff}}$ , and (d)  $\text{HVL}_{\text{eff}}$  of three-layered  $\text{Bi}_2\text{O}_3/\text{NR}$  composites (sample#4) containing varying  $\text{Bi}_2\text{O}_3$  contents from 10 to 90 wt.% in layer#2 (middle layer) and a fixed combined thickness of 6 mm, determined at X-ray energies of 50, 100, 150, and 200 keV.

In order to understand how the developed multi-layered structure (sample#4) performed with respect to previously reported materials, the results revealed that sample#4 in this work with the  $\text{Bi}_2\text{O}_3$  content of 90 wt.% in layer#2 (middle layer) exhibited the  $\mu$  value of  $7.51 \text{ cm}^{-1}$  (at 100 keV), while the dimensionally-enhanced wood/ $\text{Bi}_2\text{O}_3/\text{NR}$  composites and  $\text{Gd}_2\text{O}_3/\text{NR}$  composites with a total  $\text{Bi}_2\text{O}_3$  content of 50 phr (approximately equal  $\text{Bi}_2\text{O}_3$  content in the sample as those in sample#4) but with a single-layer structure, had the  $\mu$  values of 2–3 and  $2.6 \text{ cm}^{-1}$  (at 100 keV), respectively [11,32]. These comparisons clearly indicate that the use of a multi-layered structure had great potential to substantially improve the X-ray shielding properties of the products.



**Figure 4.** Effective  $Pb_{eq}$  of three-layered  $Bi_2O_3$ /NR composites (sample#4) with varying  $Bi_2O_3$  contents from 10 to 90 wt.% in layer#2 (middle layer) and varying combined thicknesses of 6, 9, and 12 mm, determined at X-ray energies of (a) 50, (b) 100, (c) 150, and (d) 200 keV. The green dotted lines represent the common requirement of 0.5 mmPb used as a benchmark for this work and the blue, red, and black dotted lines represent the least  $Bi_2O_3$  contents providing the composites with the  $Pb_{eq}$  of the required 0.5 mmPb for varying thicknesses.

#### 4. Conclusions

This work theoretically compared the X-ray shielding properties of single-layered and multi-layered  $Bi_2O_3$ /NR composites by determining various shielding parameters ( $\mu_{eff}$ ,  $\mu_{m,eff}$ ,  $HVL_{eff}$ , and  $Pb_{eq,eff}$ ). In total, 11 different single-layered and multi-layered designs were used to investigate the X-ray attenuation capabilities at X-ray energies of 50, 100, 150, and 200 keV. The results indicated that the layers with higher  $Bi_2O_3$  contents had better shielding abilities than those with lower contents and the three-layered structure (sample#4), with the layer arrangement of pristine NR (layer#1)- $Bi_2O_3$ /NR (layer#2)-pristine NR (layer#3), had the highest overall X-ray shielding properties among the designs investigated, due to its highest effective  $Bi_2O_3$  content, offering enhanced X-ray shielding properties of 10.5, 8.7, 4.0, and 2.1% compared to those of a single-layered structure (sample#1). Additionally, further investigation by varying the  $Bi_2O_3$  contents in layer#2 (the middle layer) of sample#4 from 10 to 90 wt.% in 10 wt.% increments revealed that the X-ray shielding properties could be further enhanced by increasing the  $Bi_2O_3$  contents; for which the recommended filler contents for actual production, determined from the common required  $Pb_{eq}$  value of 0.5 mm Pb, were 82, 72, and 64 wt.% for sample combined thicknesses of 6, 9, and

12 mm, respectively. The overall outcomes of this work reported not only the comparison of X-ray shielding properties of single-layered and multi-layered Bi<sub>2</sub>O<sub>3</sub>/NR composites but also presented potential methods to limit the reduction in mechanical properties and flexibility of the composites containing high filler contents.

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