Bismuth Oxide Filled Polyester Composites for X-ray Radiation Shielding Applications

Farida Wahyuni*, Setyawan Purnomo Sakti, Dionysius Joseph Djoko Herry Santjojo, Unggul Pundjung Juswono

 Physics Department, Brawijaya University, Jl. Veteran 65145 Malang, East Java, Indonesia

Received: 25 November 2021
Accepted: 24 February 2022

Abstract

A new bismuth and polymer-based composite was fabricated to investigate its shielding properties against X-ray in medical applications. Bismuth oxide (Bi₂O₃) were used as the filler (99.999% trace metals basis). The unsaturated polyester (UP) resin and methyl ethyl ketone peroxide (MEKPO) were used as the composite matrix (density was 1.215 g/cm³). The open mold cast technique was used to fabricate Bi₂O₃-polyester resin composite. Silicon rubber was made as a mold formed into open cubes. The dimension of this mold was 4 cm x 4 cm with the thickness of 0.5 cm. The samples were made with different bismuth oxide concentration variations: C1 (0%), C2 (1%), C3 (2%), C4 (3%), C5 (4%), C6 (5%), and C7 (6%). The optical transmission spectra of the bismuth oxide-polyester resin composite of equal thickness (5 mm) were recorded at room temperature using optical emission spectroscopy (OES, Aurora 4000) GE-UV-NIR in the visible light region. The optical transmission spectra of the composite have been observed in the visible region (400 to 700 nm). According to the results, the samples have different attenuation coefficients. The values are varied related to the sample concentrations. The highest sample concentrations (6%) have 2.680, 2.474, and 2.197 cm⁻¹, respectively for 40, 50, and 60 kV. Besides, the lowest concentrations show 0.884, 0.827, and 0.811 cm⁻¹ for 40, 50, and 60 kV, respectively. These results show that the HVL values are varied from 0.26 to 0.85 depending on the X-ray energy and the sample concentrations. The best performance is obtained from C7, indicated by the highest linear attenuation and lowest half-value layer.

Keywords: bismuth oxide, radiation protection, shielding properties, X-ray

Introduction

In the medical sector, X and γ-rays applications are increasing due to the development of science and technology. These radiation sources are good on one side. Meanwhile, the use of these radiations in various industries is inevitable. Recent developments have been reported on their versatile application in medicine, industry, and agriculture [1-3]. For example, diagnostics and radiotherapy or research purposes allow people to be exposed to these sources [3-5]. Radiation protection systems or radiation shields are developed from environmental-friendly materials or other methods against radiation produced

*e-mail: fwahyuni77@gmail.com
in radiological examinations. Shielding application is a way to decrease the radiation effect on health or the object [6]. An appropriate radiation protection material is always necessary to protect life and other materials from negative effects due to harmful radiations such as X-rays, γ-rays, and other sources [7, 8].

In the form of lead oxide, Pb and particulate polymer composites of isophthalic resin can be used as a radiation shield for γ-rays [8-10]. However, lead has a toxicological effect on human health [11]. Lead toxicity affects neurotransmitter levels and may cause severe health issues in organ damage [12]. Another study reviewed the lead effect and toxicity in health. This study shows that lead in adults may cause an increase in blood pressure, fatigue, mood swings, slow nerve conduction, drowsiness, impaired concentration, fertility disorders, decreased sex drive, headaches, and many others [13].

Many previous studies have studied Pb-free composite shields. PVA (polyvinyl alcohol)/WO_3 composite based on the Monte Carlo N-Particle (MCNP) simulation code was fabricated as a shield for photon energy (662, 778, 964, 1170, and 1407 keV) [14]. In other similar studies, HDPE (high-density polyethylene) mixed with microsized and nano-sized cadmium oxide particles were fabricated as composites for attenuation of photon beams with energy ranging from 59.53 keV to 1408.01 keV [15]. According to this study, the nanoscale reinforced HDPE enhanced overall protection or shielding properties. Another research by Atashi et al. confirmed other materials using flexible silicone rubber/W/Bi_2O_3 by open mold cast method, in which the final composites possessed higher attenuation coefficients [6]. Furthermore, a previous study focused on fabricating an X-ray shielding using composite PVA revealed another role of Bismuth oxide [16].

As an alternative material concerning lead-free material shields, in this current study, bismuth oxide and polyester were chosen for radiation protection due to their superior physical and chemical properties for an alternative X-ray protective filler. Bismuth oxide has the main substances with high density, low conductivity, low cost and is available in powder form with varied particle diameters [6-7, 17]. Thus, this study aims to get a new understanding of X-ray radiation shields using certain bismuth oxide concentrations and polyester. Hence, the samples were varied into different concentrations to obtain the best performance in protecting a medical object from X-ray radiation.

The study investigates X-ray attenuation and physical characteristics to get new information developed further for radiation shielding applications.

### Materials and Methods

#### Fabrication of Bismuth Oxide-Polyester Resin Composite

The unsaturated polyester resin was used as the composite matrix. Bi_2O_3 (202827-10G, purchased from Sigma Aldrich) was used as a filler. This powder has a 99.999% trace metals basis (aerodynamic diameter = 1 µm). Besides, YUKALAC 108B unsaturated polyester (UP) resin and methyl ethyl ketone peroxide (MEKPO) were purchased from Bratco Chemika PT, Indonesia. The UP’s density is 1.215 g/cm³. The melting point, water absorption, tensile strength are 170°C, 0.118% (24 hours), and 5.5 kg/mm², respectively. The modulus of elasticity is 300 kg/mm², while the elongation at break is 1.6%.

The open mold cast technique was used to fabricate Bi_2O_3-polyester resin composite [6]. For the first step, silicon rubber was made as a mold formed into open cubes. The dimension of this mold was 4 cm x 4 cm (thickness = 0.5 cm). The samples were made with different bismuth oxide concentrations: C1 (0%), C2 (1%), C3 (2%), C4 (3%), C5 (4%), C6 (5%), and C7 (6%). A magnetic stirrer was used to make a homogeneous mixture of bismuth oxide, methyl ethyl ketone peroxide, and polyester (rotated at 30°C, 1000 RPM, to remove bubbles inside the composite mixture). This process was conducted for five minutes (modified from [10]), to avoid solidify (optimal condition).

After uniform dispersion, bismuth oxide, polyester, and methyl ethyl ketone peroxide mixture were poured consecutively into the mold. These samples were then kept uniformly to remove the void with the help of a spatula, left for five hours (at room temperature). For the next step, the samples were removed from the mold. The produced glass samples were polished with scrubbing papers of 120, 180, 240, 320, 400 grade, and then polished again with scrubbing papers and water (600, 800, 1000, 1500, 2000, 3000, 5000 grade). The glass samples were polished with compound and glass for the final step to obtaining maximum flatness and smooth surface (Fig. 1).

![Composite samples](image_url)
Measurement of Optical Transmission Spectra

The optical transmission spectra of the bismuth oxide-polyester resin composite of equal thickness \(x\) (5 mm) were recorded at room temperature using optical emission spectroscopy (OES, Aurora 4000) GE-UV-NIR in the visible light region. The optical transmission spectra of the composite have been observed in the visible region (400 to 700 nm). The transmission measurement schematic is shown in Fig. 2. This procedure was used to measure the light transmission value before the sample, then by providing the same light source to each sample to obtain the light transparency value of each tested sample shown on the OES tool. After that, the sample was placed inside a closed chamber.

Shielding Characterization

The linear attenuation coefficient, \(\mu\), was calculated using Eq. (2) [15]. According to the Lambert-Beer law, the calculation was conducted for each sample for X-ray of appropriate energy (40, 50, and 60 kV). The sample thickness is denoted by \(x\). \(I_o\) and \(I\) show the initial photon intensity and the transmitted intensity of photons. In other words, \(\mu\) can be evaluated as the slope of the best fitting correlation between the sample thickness and intensity comparison. Another important parameter was HVL (half-value layer) thickness. This parameter was calculated using Eq. (3) [6]. PP (penetrating power) was also evaluated using Eq. (4) to evaluate the sample penetrating power against visible light [10].

\[
I = I_o e^{-\mu x}
\]

\[
\mu = \ln \left( \frac{I_o}{I} \right) / x
\]

\[
HVL = \ln 2 / \mu
\]

\[
PP = \left[1 - \left( \frac{I - I_o}{I_o} \right)\right] \times 100\%
\]

Density measurements were conducted using the Archimedes method. In this step, pure water was used as an immersing medium.

Results

Attenuation Coefficient and HVL

Evaluation of the radiation shielding properties of bismuth oxide/polyester composites concerning X-ray radiation was estimated. The attenuation coefficients were calculated theoretically. The attenuation coefficient, \(\mu\), is a common parameter investigated in developing or identifying the performance of shielding material. In this study, the attenuation coefficient is presented in Table 1. According to this table, the values are varied related to the sample concentrations. The highest sample concentrations (6%) have 2.680, 2.474, and 2.197 cm\(^{-1}\), respectively for 40, 50, and 60 kV. Besides, the lowest concentrations show 0.884, 0.827, and 0.811 cm\(^{-1}\) for 40, 50, and 60 kV, respectively. These results show a consistent trendline that a higher concentration will generate a higher attenuation coefficient. Moreover, a higher energy will also generate a lower attenuation coefficient.

Besides, HVL values are found to decrease with an increase in the filler. From Table 1, it can be noticed that C1 from all energy variations has the highest HVL value. In the other side, C7, with more concentrations, has the least HVL values, resulting in 0.26 cm, 0.28 cm, and 0.32 cm for 40, 50, and 60 kV. As the energy increases, they are able to penetrate the attenuator materials more deeply. This phenomenon will generate a higher HVL. A higher HVL value shows worse radiation protection related to the thickness requirements [6]. According to the results, it can be seen that all C7 samples do exhibit good shielding capability for the X-rays of energies 40, 50, and 60 kV, with a higher \(\mu\) and lower HVL value.

Penetrating Power

PP values were obtained by calculating the comparison between \(I\) and \(I_o\). PP was obtained by exposing the samples to visible light for different sample concentrations and energies. It is shown that PP decreases as the sample concentration decreases (Fig. 3). Besides, PP increases when the energy decreases. The table shows that the highest PP
Wahyuni F., et al.

Tungsten (W), and iron (Fe), with considerable density, have been widely used to attenuate gamma radiations due to their dominant photoelectric effect. Other studies also focused on using polymer nanocomposites as a novel material resulting in outstanding special physical and mechanical features by adding only a small amount of the nano reinforcements [20].

Lead (Pb) is commonly used as a shield since it has a high density and high efficiency in sheets, plates, laminates, bricks, foils, and others [8]. Nevertheless, the use of Pb is limited, and it is unsuitable for some specific applications that require chemical or physical stability [7]. Besides, lead is a toxic material, chemically unstable, and heavy; therefore, researchers focused on alternative lead-free materials [18]. It is urgently required to study and fabricate radiation-shielding using a novel material, including lead-free composite.

Bismuth-based nanoparticles have gained increasing attention due to their considerable dose enhancement in radiotherapy [21-22]. These materials have also been used as drug components or cancer treatment agents in radiotherapy [23-24] and contrast agents in computerized tomography scans [24]. Moreover, chemically, bismuth components dissolve in physiological conditions, removed from the body as safe soluble bismuth ions [25]. Interestingly, bismuth-based nanoparticles showed substantial radiation dose enhancement under kilovoltage X-ray beams greater than well-known gold nanoparticles, fewer adverse effects, lower costs, higher cell penetration, and higher biocompatibility than conventional radiosensitizers. Hence, they have favorably received the increasing attention of researchers [18] and can be prepared in various well-defined shapes.

Our study results indicate that bismuth-based material has a good performance for radiation protection. As supported by a previous study, Bi$_2$O$_3$ fillers in the polycarbonate matrix increased the attenuation coefficients of the composites significantly. Interaction between the radiation and matter is also required to obtain a good and proper shield performance [19]. As found generally, heavy metals such as lead (Pb), tungsten (W), and iron (Fe), with considerable density, have been widely used to attenuate gamma radiations due to their dominant photoelectric effect. Other studies also focused on using polymer nanocomposites as a novel material resulting in outstanding special physical and mechanical features by adding only a small amount of the nano reinforcements [20].

Table 1. Linear attenuation coefficients and HVL values for all samples with energy variations.

<table>
<thead>
<tr>
<th>Samples</th>
<th>40 kV</th>
<th>50 kV</th>
<th>60 kV</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\mu$ (cm$^{-1}$)</td>
<td>HVL (cm)</td>
<td>$\mu$ (cm$^{-1}$)</td>
</tr>
<tr>
<td>C1</td>
<td>0.88</td>
<td>0.78</td>
<td>0.83</td>
</tr>
<tr>
<td>C2</td>
<td>1.29</td>
<td>0.54</td>
<td>1.20</td>
</tr>
<tr>
<td>C3</td>
<td>1.48</td>
<td>0.47</td>
<td>1.39</td>
</tr>
<tr>
<td>C4</td>
<td>1.93</td>
<td>0.36</td>
<td>1.82</td>
</tr>
<tr>
<td>C5</td>
<td>2.20</td>
<td>0.32</td>
<td>2.07</td>
</tr>
<tr>
<td>C6</td>
<td>2.51</td>
<td>0.28</td>
<td>2.26</td>
</tr>
<tr>
<td>C7</td>
<td>2.68</td>
<td>0.26</td>
<td>2.47</td>
</tr>
</tbody>
</table>

Fig. 3. Penetrating power level for all energy and sample concentration variations.

Discussion

In the medical sector, radiation technology is used in a wide range of applications. Increasing the utilization of hazardous radiations, like $X$ and $\gamma$-rays sources hospitals and research centers or other medical sectors for diagnostic and therapeutic applications, has provided a much more unsecured place for personnel. Hence, there will be a need to design an appropriate radiation shielding, depending on the type of ionizing radiation, to reduce the radiation dose in the intended site [18]. Interaction between the radiation and matter is also required to obtain a good and proper shield performance [19]. As found generally, heavy metals such as lead (Pb),
bismuth oxide nanoparticles and multi-walled carbon nanotubes (MWCNT) were developed as poly (methyl methacrylate) (PMMA) to fabricate the nanocomposite for electron-beam shielding applications [19]. Another previous study shows the stabled thermal properties, chemical structures, and morphology of polyimide and Bi$_2$O$_3$ for radiation shielding composites [26].

**Conclusions**

The research results show a consistent trendline of linear attenuation coefficient that a higher concentration will generate a higher attenuation coefficient. Besides, a higher energy will also generate a lower attenuation coefficient. It is indicated that all C7 samples (the highest concentration) do exhibit good shielding capability for the X-rays of energies 40, 50, and 60 kV, with a higher linear attenuation coefficient and lower HVL value. Moreover, the penetrating power calculation clearly shows that the level decreases as the bismuth oxide - polyester concentration decreases. The values are 33.33% to 73.81%, influenced by the energy and concentration variations. A higher composite concentration has a better shielding performance than a lower concentration, indicated by a higher PP value.

**Acknowledgments**

All authors would like to offer special thanks to Sensor Laboratory, Physics Department, Brawijaya University, Indonesia.

**Conflict of Interest**

The authors declare no conflict of interest.

**References**


