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Evaluating the dosimetry response to gamma-rays in HDPE/Bi_2O_3 and PC/Bi_2O_3 nanocomposites

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HIGHLIGHTS

- Polymer/bismuth oxide nanocomposites were considered as gamma-ray dosimeters.
- Two types of polymers were investigated crystalline HDPE, and amorphous PC.
- The change in electrical current was considered as the dosimetry response.
- PC exhibited better dispersion up to 60 wt%, while HDPE showed agglomeration at 40 wt%.
- The amorphous PC at 40 wt% exhibited a higher response rather than HDPE.

ABSTRACT

The recent emphasis on nanocomposites composed of polymers and metal oxides is largely due to their significant promise as effective radiation sensors, detectors, and dosimeters for gamma rays, X-rays, and charged particles. The sensitivity of a system may be affected by various factors, including the volume of the sensitive material, the concentration of heavy metal oxide nanoparticles, the bias voltage applied, and the degree of crystallinity within the polymer matrix. Studies have shown that a pronounced degree of crystallinity in polymers can restrict the homogeneous spread of nanoparticles. This research utilized two distinct polymer matrices, namely HDPE and PC, to create a nanocomposite that incorporated bismuth oxide nanoparticles at concentrations reaching 60 wt%. FESEM images revealed that PC exhibited better dispersion up to 60 wt%, while HDPE showed agglomeration at 40 wt%. Under a defined dose rate of 42.67 mGy.min^{-1}, and with a fixed amount of Bi_2O_3 nanofillers, the dosimetry response (measured as a change in electrical current) of PC was twice as pronounced compared to HDPE. Therefore, PC, as an amorphous polymer containing 50 wt% Bi_2O_3, may be considered a suitable candidate for dosimetry applications.

KEYWORDS
Dosimetry
Gamma-rays
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Polycarbonate
High-Density Polyethylene
Crystallinity

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1 Introduction

Scientists have recently focused their attention on polymer nanocomposites due to their promising applications in areas such as radiation protection, as well as in the fields of sensors, detection mechanisms, and dosimetry (Hosseini et al., 2022; Intaniwet et al., 2012; Kyatsandra and Wilkins, 2014; Malekie and Ziaie, 2015; Malekie et al., 2016a,b; Mehrara et al., 2021; Mosayebi et al., 2019, 2017; Safdari et al., 2022). These materials offer a variety of benefits, including their lightweight composition, flexibility, straightforward processing, tissue equivalence, and relatively affordable pricing. When exposed to gamma rays and high-energy photons, a polymer nanocomposite experiences the generation of electric charges within its sensitive volume. This charge generation is attributed to interactions including the photoelectric effect, Compton scattering, and pair production. The generation of electric charges in the form of electron-hole pairs can be effectively harnessed and converted into signals by applying a suitable electric field to the material. The interaction of gamma-rays with matter can give rise to several phenomena, including the photoelectric effect, Compton scattering, and pair production, particularly when photons possess energies exceeding 1.022 MeV. Compton scattering is notably the most common occurrence, leading to the pro-

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duction of secondary electrons in dosimeter materials as a result of photon interactions. These secondary electrons are subsequently converted into a signal by the application of an appropriate voltage across the dosimeter. It is essential to note that these secondary electrons may generate bremsstrahlung radiation near the heavy nuclei of bismuth nanoparticles, which enhances the sensitivity of the dosimeters. Therefore, an increase in the quantity of Bi_2O_3 nanoparticles within the polymer matrix results in a corresponding increase in electric current.

Due to their low densities, polymer-nanocomposites demonstrate a limited sensitivity to gamma radiation. To counteract this challenge, bismuth oxide (Bi_2O_3) nanoparticles, which have a density of 8.9 $g.cm^{-3}$ and an atomic number of $Z = 83$, are integrated into the polymer matrix. This integration is intended to enhance the sensitivity of radiation detection and dosimetry by increasing the probability of the photoelectric effect. Several studies have investigated the performance of polymer-heavy metal oxide composites in radiation detection and dosimetry applications. Notably, Intaniwet et al. conducted research on the incorporation of Bi_2O_3 nanoparticles into a semiconductor polymer to boost the sensitivity of detectors to 17.5 keV X-rays (Intaniwet et al., 2012).

The distribution of nanoparticles within polymer matrices presents significant challenges, especially at elevated volume fractions, as the inclusions tend to aggregate (Kim et al., 2014). Evidence indicates that significant levels of polymer crystallinity can restrict the consistent distribution of nanoparticles (Kaur et al., 2011). This investigation identifies Polycarbonate (PC), a thermoset polymer with an amorphous configuration and repeating units of -CH_2-CH (C_6H_5), as a prime candidate for producing homogeneous nanocomposites (Mehrara et al., 2021). PC is noted for its adequate radiation resistance and remarkable breakdown voltage, attributable to its aromatic structure (Chen et al., 2009; Gurkalenko et al., 2017; Mark et al., 2007). The expectation is that PC will establish more favorable connections with Bi_2O_3 nanoparticles. PC is composed of terminal groups that feature conjugated double bonds (Stix et al., 1985), along with C=O (carbonyl), C-H, phenyl, and C-O-C groups (Yadav et al., 2017). Furthermore, HDPE, characterized by a repeating unit of [C_2H_4] and classified as a thermoplastic polymer with a semi-crystalline structure, was selected to serve as the polymer matrix.

In this work, a comparative study of the dosimetry response of two nanocomposites namely PC/Bi_2O_3 and HDPE/Bi_2O_3 were carried out. The novelty of this work is investigating the effect of polymer matrix on the dosimetry response of the polymer- Bi_2O_3 nanocomposite dosimeter.

2 Research Theories

The effectiveness of detection and dosimetry is enhanced when radiation engages with materials that possess a greater attenuation coefficient at a specific energy level. Quantum efficiency (QE) is frequently characterized as (Intaniwet et al., 2012):

$$QE = \left(1 - e^{-(\frac{\mu}{\rho})\rho x}\right) \times 100 \tag{1}$$

The mass attenuation coefficient, represented as $\frac{\mu}{\rho}$, is derived from the photon cross-section database, where ρ denotes the composite density and x signifies the thickness. This coefficient was determined for the two composite materials, PC/Bi_2O_3 and HDPE/Bi_2O_3, by utilizing online resources and defining the respective compounds through the XCOM software.

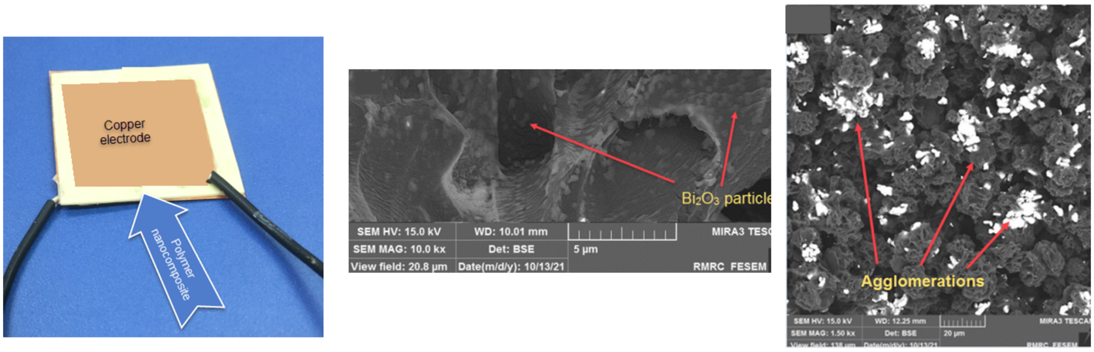
3 Experimental

In this experimental study, granules of PC and HDPE were procured from the Iranian-Khuzestan petrochemical company, exhibiting densities of 1.2 $g.cm^{-3}$ and 0.93 $g.cm^{-3}$, respectively. Bi_2O_3 nanopowders, characterized by a density of 8.9 $g.cm^{-3}$ and average particle sizes ranging from 90 to 210 nm, were sourced from Sigma-Aldrich. To summarize the methodology, the polymers were dissolved in appropriate solvents utilizing a hotplate magnetic stirrer, followed by the incorporation of Bi_2O_3 nanoparticles into the polymer solution, which were subsequently dispersed using an ultrasonic probe. The resultant samples were then subjected to hot pressing to achieve a uniform thickness of 1 mm and dimensions of 4×4 cm^2 at varying concentrations. As illustrated in Fig. 1-a, copper plates with a thickness of 100 μm were affixed to the samples using silver paste to create electrodes on both surfaces of the dosimeters. Further details regarding the fabrication process can be referenced in our previous studies (Mehrara et al., 2021; Safdari et al., 2022; Veiskarami et al., 2022).

In this study, PC/Bi_2O_3 nanocomposites were synthesized with varying concentrations of Bi_2O_3, specifically at 0, 5, 20, 40, and 50 wt%, while HDPE/Bi_2O_3 nanocomposites were produced with loadings of 0, 20, 40, and 60 wt%. The fabrication process employed a solution method. Subsequently, the samples underwent gamma-ray irradiation using a Co-60 source from the Picker V-9 at the Secondary Standard Dosimetry Laboratory (SSDL) located in Iran-Karaj, with different source to surface distances (SSDs) applied. The dosimetric response was evaluated by measuring the variation in electrical current passing through the samples, which was recorded using a SuperMax Standard Imaging electrometer set at 400 V.

4 Results and discussion

Figures 1-b and 1-c show the FESEM images of PC/Bi_2O_3 and HDPE/Bi_2O_3 nanocomposites. As can be seen from Fig. 1-b, the Bi_2O_3 nanoparticles were dispersed uniformly in the PC matrix. But, as shown in Fig. 1-c, the nanoparticles were agglomerated, which exhibited a non-uniform dispersion in the HDPE matrix. Thus, PC exhibits a better dispersion state of the inclusions in comparison with HDPE. An interpretation of this effect reveals that HDPE, as a semi-crystalline polymer, restricts



(a) (b) (c)

Figure 1: a) Fabrication of the dosimeters, and FESEM for, b) PC/Bi_2O_3, and c) HDPE/Bi_2O_3 nanocomposites.

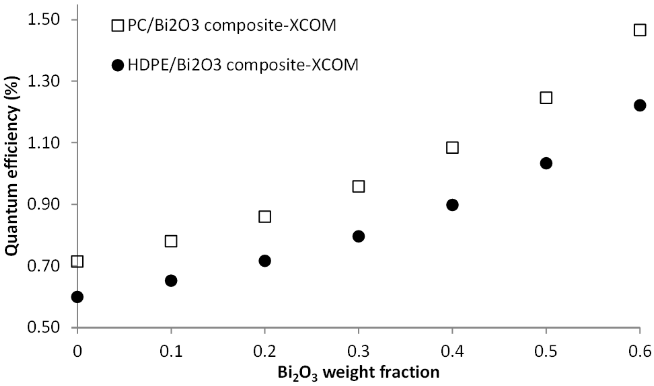
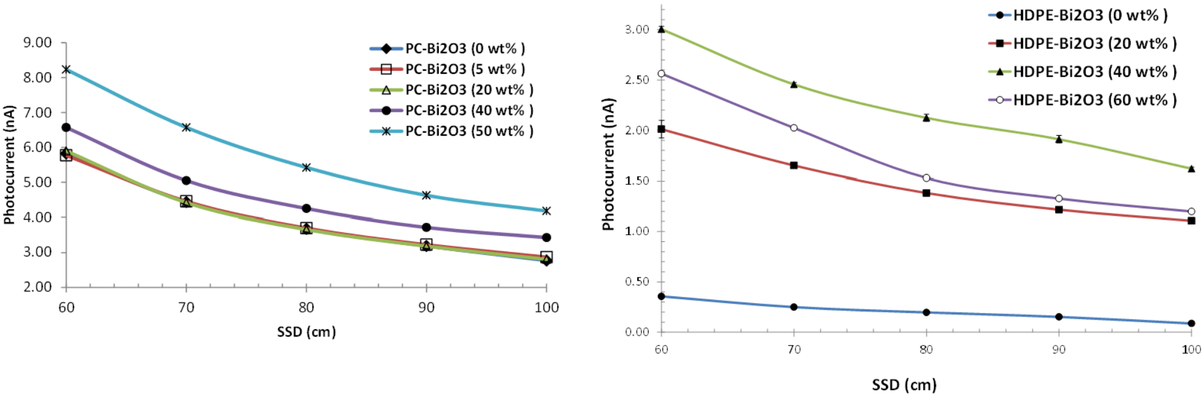


Figure 2: Quantum efficiency for PC/Bi_2O_3, and HDPE/ Bi_2O_3 composites, thickness of 1 mm at 1250 keV for different concentrations up to 60 wt% using the XCOM.



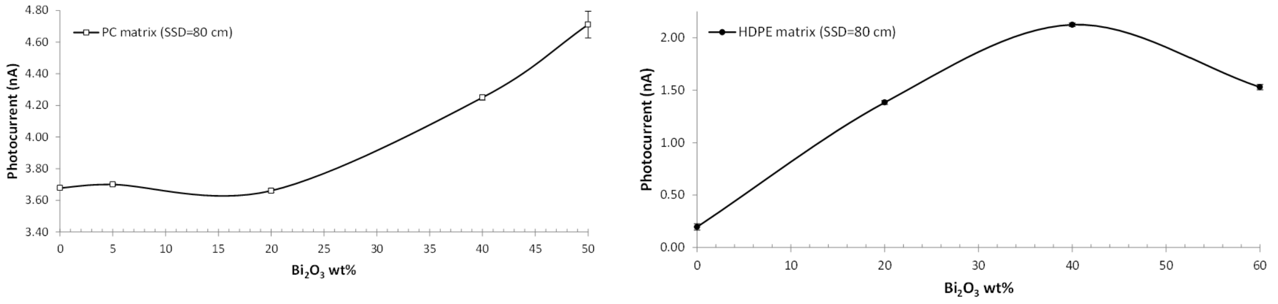
(a) (b)

Figure 3: Average photocurrent vs. SSD for (a) PC/Bi_2O_3, and (b) HDPE/ Bi_2O_3 nanocomposites.

the uniform distribution of nanoparticles, which can be attributed to the presence of crystallites.

Figure 2 displays the predicted quantum efficiencies of a 1 mm thickness for the two composites namely PC/Bi_2O_3, and and HDPE/Bi_2O_3 at 1250 keV with various concentrations of the inclusioins using the XCOM program. For both composites, as the Bi_2O_3 concentration increases, the QA enhances consequentntly. Also, PC due to higher density (1.2 g.cm^{-3}) in comparision with the HDPE (0.93 g.cm^{-3}) exhibits the higher QE.

Figures 3-a and 3-b exhibit the average photocurrent vs. SSD for PC/Bi_2O_3, and HDPE/Bi_2O_3 nanocomposites, respectively. As the Bi_2O_3 wt% increases, the dosimeter response enhances subsequently. As can be seen from Fig. 3-a, there isn't any significant difference in the electric current for the percentages of 0, 5 and 20 wt% for PC/Bi_2O_3 nanocomposites. It can be inferred that there is a critical weight percentage of the heavy metal



(a) (b)

Figure 4: Dosimetry response at a fixed SSD=80 cm for a) PC/Bi_2O_3, and b) HDPE/Bi_2O_3 nanocomposites.

oxide nanoparticles into the PC matrix, in which for concentrations greater than 20 wt%, the amount of electric current increases subsequently. Furthermore, the distribution states of the inclusions within the PC matrix may have been affected by agglomeration at 5 and 20 wt%. This is evident as the electric current generated by samples with 0, 5, and 20 wt% are closely similar.

As shown in Fig. 3-b, for HDPE/Bi_2O_3 nanocomposite, this trend is valid up to 40 wt%; but for 60 wt% sample, the dosimeter response decreases by 20-30% in comparison with the 40 wt% sample. This phenomenon can be explained by the semi-crystalline characteristics of the HDPE matrix, which result in agglomeration at elevated reinforcement loadings, specifically at 60 wt%. This agglomeration subsequently leads to a reduction in the response of the dosimeter.

The dosimetry response of PC/Bi_2O_3 and HDPE/Bi_2O_3 nanocomposites for various Bi_2O_3 wt% at the fixed SSD=80 cm which is equal to dose rate of 42.67 mGy/min is depicted in Fig. 4. As can be seen from Fig. 4-a, the dosimetry response for PC/Bi_2O_3 nanocomposite is linear between 20 to 50 wt%. But, as shown for in Fig. 4-b, for HDPE it was increases linearly up to 40 wt%, then decreases 25% at 60 wt%. This is due to agglomeration of the inclusions in the semi-crystalline polymer of HDPE.

5 Conclusions

This study examined the influence of the polymer matrix on the sensitivity of an innovative radiation dosimeter that utilizes polymer-heavy metal oxide nanocomposites. For this purpose, two composites namely PC/Bi_2O_3 and HDPE/Bi_2O_3 were considered with different crystallinity degree of the polymer matrices. Quantum efficiency was evaluated for both composites using the XCOM. It was concluded that PC with higher density exhibited higher efficiency at various concentrations in comparison with the HDPE. FESEM images demonstrated that agglomeration happened at the composites containing HDPE as a semi-crystalline polymer matrix. This is due to the fact that crystallinity can hinder the homogenous dispersion of the nanoparticles. So, PC/Bi_2O_3 has potential application to be used as a real-time dosimeter for therapeutic dose level.

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Conflict of Interest

The authors declare no potential conflict of interest regarding the publication of this work.

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