Effects of particle size and weight percentage of heavy metal elements on photon shielding efficiency of reinforced polymer composites

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ABSTRACT

Background: Polymer composites when reinforced with heavy metals in the form of micro/nano particles are efficient gamma- and X-ray shielding materials providing such advantageous features as cost-effectiveness, light-weight factor, flexibility, non-toxicity, conformability over conventional shields. Materials and Methods: In this paper the influence of weight percentages and particle sizes of bismuth and tungsten contents of reinforced composites, i.e., bismuth/rubber (Bi/R) and tungsten/rubber (W/R) shields, in their photon absorption capabilities for photon energies, ranging from 40 to 350 keV, were investigated using both MCNPX simulations and measurements. The Bi/R and W/R polymer composites were prepared by mixing rubber with different weight percentages of bismuth and tungsten powders before exposing them to $^{152}$Eu gamma-rays. Then, the shielding efficiencies or photon transmission fractions, half-value layer (HVL) and tenth-value layer (TVL) of the samples were determined. Results: The simulation results confirmed that the particle size of the heavy element plays an important role in the shielding efficiency, especially at low photon energies. The decrease in the particle size of shielding material in each weight percentage improved the radiation shielding features. Therefore, the results supported the feasibility of nano-sized composite applications for shielding against low-energy photons, especially in diagnostic studies such as mammography. Conclusions: At low photon energies, especially around 40 keV, a considerable decrease in photon flux was achieved by incorporating nano-sized heavy elements in a polymer matrix.

Keywords: Atomic number, MCNPX simulation, particle size, photon, polymer composite, shielding.

INTRODUCTION

Many researchers worldwide have been conducted to find new materials and compounds as alternatives to traditional shielding materials. These researchers have developed polymer micro/nano composites using non lead metals for radiation shielding applications. Polymer composites when reinforced with heavy metal in form of micro/nano particles provides many advantages including cost-effectiveness, light weight factor, flexibility, non-toxicity, conformability, etc. over conventional materials.

Many researchers have studied the effects of the type, weight percentages and particle size of radiation absorbing materials on the radiation shielding properties for X-ray, gamma and neutron attenuation (1-5).

Botelho et al. (2011) compared the micron and nano size of CuO on the X-ray absorption
and concluded that, the nano size CuO absorb more radiation when exposed to low energy X-ray \(^6\). Also, Künzel et al. (2011) showed that the samples with nano size of CuO present a greater potential in absorbing low energy X-ray compared to micro size CuO \(^7\). Kaloshkin et al. (2012) studied the nano composites based on UHMWPE with W and B4C nano powders. According to this research nano-size fillers increase the gamma-ray protection properties \(^8\). Alymov et al. (2005) concluded that the shielding materials containing nano powders provide high protective characteristics against X-rays and thermal neutrons \(^9\).

So the general trend seems to be towards the use of nano particles in radiation shielding as a promising way to develop the radiation protective materials in nuclear industries \(^10\)-\(^12\). High Z materials attenuate low energy X-rays through photoelectric effect. Lead (Z=82) is considered as the most effective material for protection against X-ray exposure, however exposure to lead may lead to several health problems. There is an increasing demand for lead free polymer-based materials composite. High Z, non-lead particles such as tungsten and bismuth can be used for conventional lead/polymers composite \(^13,14\).

The present study aimed to investigate, (1) the effects of particle type and weight percentages of attenuator particles on the absorption ability of shielding layer against gamma rays in the energy range of \((40 \text{ to } 350 \text{ keV})\), (2) the effects of particle size in the range of \((100 \text{ nanometer to } 100 \text{ micrometer})\) on the absorption ability of shielding layer against gamma rays in the energy range of \((40 \text{ to } 350 \text{ keV})\), and (3) comparing the shielding effectiveness of the composite material used in the present work at different photon energies.

**MATERIALS AND METHODS**

**MCNPX simulation**

Shielding efficiency of bismuth/Rubber and tungsten/Rubber polymer composite, each with the different particle size and weight percentages of attenuator were simulated by utilizing the MCNP computer code. The MCNP code is an internationally recognized code for analyzing of neutron and gamma-ray transports using the Monte-Carlo method which is developed and maintained by Los Alamos National Laboratory \(^15\). MCNP simulation is unable to calculate the radiation shielding efficiency depending on the particle size. In this simulation, to define particle size simulation the lattice card were used. To define different size of spherical particle in a regular pattern following steps were followed:

1. Bismuth and tungsten particles defined as spheres located in the center of cubes in the rubber matrix.
2. The diameter of spheres was selected according to different bismuth and tungsten particle sizes of \(100 \mu\text{m}, 10 \mu\text{m}, 1 \mu\text{m}\) and \(100 \text{nm}\).
3. For each bismuth/Rubber and tungsten/Rubber polymer composite, the bismuth and tungsten powder with \(15, 30, 45\) and \(60 \text{wt\%}\) along with the rubber matrix were defined.
4. Shielding efficiency of bismuth/Rubber and tungsten/Rubber each with different weight percentages and size of particles against gamma rays of energies \((40, 60, 120 \text{ and } 350 \text{ keV})\) were calculated and determined by the code. The measurement data were analyzed with origin 2019b software and expresses as mean \(\pm SD\) (standard deviation).

Figure 1 shows the schematic of the designed geometry in MCNP code. Gamma source with energies of \(40, 60, 120 \text{ and } 350 \text{ keV}\) were calculated and determined by the code. The measurement data were analyzed with origin 2019b software and expresses as mean \(\pm SD\) (standard deviation).

Figure 1. A schematic diagram for MCNPX simulation.
**Experimental Measurements**

**Sample preparation**

In order to construct the elastomer shielding material, the chloroprene rubber/natural rubber (NR/CR) combination and other additives have been used as following (table 1):

1. CR/NR, 2. MGO/DPG/MBTS/S/ETU, 3. ZNO/ST.Acid/Wax/TMQ/4010NA and 4. N550/840 Oil/Anti Ox.Sp.

The first group materials are double-bond polymers basically used to improve elastomer properties, such as flexibility, mechanical performance and filling in the ozone condition; whilst the second group materials are added to the mixture in order to facilitate low-temperature curing and to provide better homogeneity. The third group materials are accelerators and actuators, which are responsible for assisting the curing chain and accelerating the curing process. The fourth group materials are primary fillers including soot and aromatic oil.

Having prepared the above mixture, one has to add and mix appropriate heavy elements (i.e., lead, bismuth and tungsten) to enhance photon attenuation. Different weight percentages (0, 15, 30, 45 and 60) of lead, bismuth and tungsten have been added using the Schwabenthan Polymer 200L roller. The rheometry studies have been undertaken with the raw samples at temperature 190°C in order to define the curing time, pressure and temperature. Finally, it has been decided to prepare the samples with 200mm×200mm×2mm dimensions and at 160°C and 25 MPa temperature and pressure, respectively.

In order to study the quantity and homogeneity of distribution of heavy metal in polymeric matrix, the SEM and E-DAX analysis have been conducted by VEGA-TESCAN system. The SEM images of samples consisting of heavy metal are exhibited in figure 2 which confirms the homogeneity of heavy element distribution in the sample.

**Table 1. Constituent materials in base polymer matrix used in this study.**

| Material       | Weight Percent | Manufacturing Country | Brand/Chemical Acronym |
|----------------|----------------|-----------------------|------------------------|
| CR             | 35             | Germany               | -                      |
| NR             | 65             | Malaysia              | SMR-20                 |
| MgO            | 3              | China                 | -                      |
| ZnO            | 5              | Iran                  | -                      |
| Stearic acid   | 2              | Germany               | RGA                    |
| Soot           | 40             | Iran                  | N550                   |
| Aromatics Oil  | 20             | Iran                  | -                      |
| Paraffin Wax   | 15             | Iran                  | Antilux                |
| Antioxidant SP | 2.3            | Slovakia              | Dusantox               |
| TMQ            | 1.6            | Germany               | CZ/EG-C                |
| 4010 Na        | 1.8            | China                 | -                      |
| Vulkacit D     | 1              | China                 | DPG                    |
| Vulkacit DM    | 1.2            | China                 | MBTS                   |
| ETU            | 2.6            | China                 | -                      |
| Sulfur         | 3.5            | Iran                  | -                      |
| Tungsten       | ---            | Russia                | W                      |
| Bismuth        | ---            | Russia                | Bi                     |

**Figure 2.** The SEM and E-DAX images of constructed shielding material.
Shielding efficiency measurements

A 2”×2” right cylindrical NaI (Tl) homemade scintillator with an input voltage of 650-700 volts (2”×2” NaI (TL) Detector includes a 2”×2” cell manufactured by Scintitech, a XP2020 PMT and a VD124 K voltage divider, both manufactured by Photonis) has been used for the gamma-ray spectroscopy. The measurements with and without constructed shielding materials (containing different percentages of heavy elements) have been carried out using the data acquisition electronics shown in figure 3. The high voltage has been set to +700V and the analog-to-digital convertor (ADC), pre-amplifier and multi-parameter analyzer are ORTEC 4010, 3000 and 4201, respectively. The pulse-height spectrum of NaI scintillator has been recorded in order to calculate the area under the photopeaks using a dedicated data acquisition software (DAS). Next, the attenuation coefficients, half-value layers (HVLs) and tenth-value layers (TVLs) have been calculated as listed in tables 2.

### RESULTS

In this study, a series of radiation shields have been designed using rubber elastomers with different weight percentages (15, 30, 45 and 60%) of bismuth and tungsten each with different particle size from 100 µm to 100 nm. The effect of size of bismuth particles at constant 15% wt on the gamma-ray have been shown in figures 4a. Also in figures 4b gamma-ray flux versus different size of tungsten particles at constant 15% wt tungsten Rubber have been shown.

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**Table 2.** Measured photon attenuation coefficients for bismuth and tungsten using 512Eu gamma-rays.

| Element | Weight percent (%) | TVL (mm) | HVL (mm) | Attenuation Coefficients (1/mm) |
|---------|------------------|----------|----------|-------------------------------|
|         |                  | Eu-152   | Eu-152   | Eu-152                        |
| Bismuth | 15               | 344 keV  | 122 keV  | 40 keV                        |
|         |                  | 7.53     | 3.24     | 2.31                          |
|         |                  | 0.092    | 0.214    | 0.3                            |
|         | 30               | 7.24     | 3.54     | 2.18                          |
|         |                  | 0.2      | 0.318    | 0.651                         |
|         | 45               | 7.52     | 3.67     | 1.27                          |
|         |                  | 0.306    | 0.628    | ...                           |
|         | 60               | 4.30     | 4.00     | 1.30                          |
|         |                  | 0.535    | 0.576    | ...                           |
| Tungsten| 15               | 11.51    | 7.24     | 3.47                          |
|         |                  | 0.75     | 0.535    | 0.628                         |
|         | 30               | 16.10    | 4.30     | 4.85                          |
|         |                  | 1.30     | 0.353    | 0.414                         |
|         | 45               | 9.28     | 2.26     | 2.97                          |
|         |                  | 0.248    | 0.318    | 0.651                         |
|         | 46               | 6.87     | 5.26     | 2.07                          |
|         |                  | 0.335    | 0.438    | 0.131                         |

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![Figure 3. Block-diagram of electronic modules used in the measurements.](image-url)

![Figure 4. Variation of photon flux against additive particle size for a sample of 15% Bi (a) and 15% W (b) for photon energies of 40 keV to 350 keV.](image-url)
The gamma–ray flux as a function of size of bismuth particles at constant 30% wt have been shown in figures 5a. Also gamma–ray flux as a function of size of bismuth particles at constant 30% wt of tungsten/Rubber have been shown in figures 5b.

The gamma–ray flux as a function of size of bismuth particles at constant 45% wt and 60% wt have been shown in figures 6a and 7a. Also gamma–ray flux as a function of size of bismuth particles at constant 45% wt and 60% wt of tungsten/Rubber have been shown in figures 6b and 7b.
DISCUSSION

As it can be seen in series of figures (2 a,b to 5 a,b), the gamma-ray flux received to the detector has been reduced with increasing the weight percentages of heavy elements (Bi and W). Also, at a specific weight percentage, the photon flux has been reduced by decreasing the particle size especially at low gamma energy of 40 keV.

The results also confirm that at photon energy of 40 keV, bismuth exhibit larger attenuation coefficient compared to tungsten mainly due to higher photoelectric cross section \[^{17}\]. Similar behavior can be seen at photon energy of 60keV.

When the photon energy reaches to 120 keV, the photoelectric effect is less dominant therefore the attenuation dependency on the cross-section weakens which results in almost similar attenuation coefficients for both tungsten and bismuth.

At photon energy of 130 keV, Compton interaction is dominant and because the Compton cross section varies smoothly with atomic number, both bismuth and tungsten represent similar behavior at this energy region.

The results of photon attenuation coefficients measurement for the bismuth/Rubber and tungsten/Rubber polymer composite samples with different percentages of particles against gamma rays from \(^{152}\)Eu source have been also measured as listed in tables 2.

CONCLUSIONS

The MCNPX simulation results show that at low energies, reducing the particle size increases the radiation absorption. The value of photon flux reduction reaches to 22% at 40keV, whilst it remains below 6% at 60 keV. It has been also observed that the heavy element particle size represents non-significant role in reducing the gamma flux received at the detector position with increasing the gamma ray energy.

Overall, the comparison between MCNPX simulations and measurements, which are in promising agreement, confirm that the linear attenuation coefficient increases with increasing the heavy element additives. At low photon energies (especially around 40 keV) dominate decreases in photon flux obtained by using nano size attenuator in polymer matrix.

Conflicts of interest: Declared none.

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