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Research Article

Experimental Comparison of PbO and BaO Addition Effect on Gamma Ray Shielding Performance of Epoxy Polymer

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Abstract

This study was devoted to investigate the gamma ray shielding abilities of the epoxy composites prepared by using two different reinforcing materials: barium oxide and lead oxide. The Epoxy/PbO and Epoxy/BaO composites were produced by using 10 wt.%, 20 wt.%, and 40 wt.% reinforcing materials to obtain low weight radiation shielders. The heaviness of the samples were compared with some conventional shielding materials such as lead, steel, concrete etc. The gamma ray shielding performance of the pure epoxy and the composites were measured experimentally by using NaI(Tl) detector. In the experiments, Ba-133 point radioactive source was also utilized as a gamma ray source with the 81 keV and 356 keV energies. As the gamma ray shielding parameters of the composites, the mass attenuation coefficient, half layer value and tenth layer value thicknesses and mean free path distance were considered. After comparison of the related parameters of the PbO and BaO added epoxy composites, it was revealed that the gamma radiation shielding performance of the PbO added epoxy composite can also be obtained by adding BaO to the epoxy. However, to achieve the same performance, BaO should be added twice as much as the PbO additive percentage. On the other hand, it was determined that 40 wt.% BaO added epoxy composite exhibits a better radiation shielding performance than steel, concrete and Gd_2O_3 nanoparticle added epoxy composite for the photons with 81 keV and 356 keV energies. Thus, a lightweight and non-toxic gamma-ray shielding material can be produced by using an Epoxy/BaO composite.

Keywords: Epoxy Composite, PbO, BaO, Mass attenuation coefficient, HVL, TVL.

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1. Introduction

Due to increasing emission of ionized radiation such as X-ray and gamma ray in medical applications, radiation protection has become a very important issue. Radiation protection can be performed by decreasing the time for exposure, increasing the distance between the radioactive source and living body, and using good radiation shielding equipment. Using effective radiation shielding materials is the best reliable way to protect from the harmful results of radiation. Lead and concrete are the most commonly utilized materials for X-ray and gamma ray protection. In recent years, alternative to lead and concrete, scientist have focused on finding alternative radiation shielding material which are transparent, non-toxic, and light weight. Polymers reinforced with elements with high atomic numbers are good candidates which fulfill these requirement mentioned above since polymers play a significant role in in primary and secondary protection against gamma radiation (Hussain, Haq, & Mohammad, 1997; Kucuk, Cakir, & Isitman, 2012). From this perspective, the radiation shielding performance of various polymer composites were researched.

Soylu et al. reinforced the hydrogen-rich, flexible and soft EVA polymer with tungsten carbide with different weight percentages varying between 50 and 70 and studied the gamma ray shielding efficiency of the related composites for the incident gamma photons emitted from the $_{131}$I, $_{137}$Cs, and $_{241}$Am radioactive sources. They found that the best gamma ray shielding performance is achieved by using 70% tungsten carbide for the EVA polymer matrix (Soylu, Lambrecht, & Ersöz, 2015). Furthermore, Li et al. compared the gamma ray shielding performance of the micro- and nano-sized Gd_2O_3 reinforced epoxy polymer composites for the photon energies ranging from 31 keV to 356 keV. They reported that the better gamma ray screening is observed for the epoxy composites include nano-sized Gd_2O_3 (Li, Gu, Wang, et al., 2017). Hou et al. also studied the gamma photon shielding ability of the basalt fiber-tungsten reinforced epoxy matrix for the photon energies of 80 keV, 356 keV, and 662 keV. They determined that the photon attenuation capability of epoxy can be enhanced by using tungsten rich basalt fiber reinforcing material in the epoxy based composite (Hou et al., 2018). Thus, a high Z metal reinforced polymer composites are considered as a new competitors of the conventional lead based radiation shielding materials. From this point of view, this study is devoted to the compare the epoxy based PbO and BaO composites' gamma ray shielding efficiency for the gamma photons emitted from $_{133}$Ba source.

2. The Law Used In the Calculation of Radiation Shielding Parameters

To reveal the gamma ray shielding performance of any material, some critical parameters recognized as mass attenuation coefficient, half value layer, tenth value layer, and mean free path have to be known. These parameters can be calculated by using an experimental data of either the absorbed or scattered gamma rays from radiation shielding material. The interaction between gamma rays and a radiation shielding medium is represented mathematically by introducing the exponential decrease in the intensity of the incident beam:

$$-\frac{dI}{dx} = \mu I \qquad (1)$$

Eq. (1) is known as Beer-Lambert law. In Eq. (1), μ is the total linear attenuation coefficient of a material having the unit of cm^{-1}. The total linear attenuation coefficient corresponds to the possibility of the occurrence of an interaction between gamma ray and matter per unit length in the radiation shielding medium. The x also is the thickness of the radiation shielding.

The solution of the Beer-Lambert equation is given by Eq. (2) under ideal geometric conditions:

$$I(x) = I_{\circ} e^{-\mu x} \qquad (2)$$

The terms of $I_{\circ}$ and $I(x)$ appeared in Eq. (2) are the intensities of the incident beam and transmitted gamma ray, respectively. The mass attenuation coefficient of the material which is composed by one type of element is calculated by the ratio of total linear attenuation coefficient and mass density:

$$\frac{\mu}{\rho} = -\frac{1}{\rho x}\ln\left(\frac{I}{I_{\circ}}\right) \qquad (3)$$

If the radiation shielding material is a chemical compound or composites or a mixture of some elements, mass attenuation coefficient is calculated by Eq. (4)

$$\mu / \rho = \sum_i w_i (\mu / \rho)_i \qquad (4)$$

In Eq. (4), w_i is the fractional weight of the i th constituent in the radiation shielding material (Sayyed, 2016). In addition to mass attenuation coefficient, half value layer (HVL), tenth value layer (TVL), and mean free path (λ) play a significant role in determining the photon attenuation ability of a material. As is known, HVL is defined as the width of a material required to reduce the air kerma of an gamma ray to half its original value, TVL is the width of a material required to reduce the air kerma of an gamma ray to tenth its original value. Mean free part is the average distance at which a gamma ray travels in the radiation shielding material without any interaction (K. Singh et al., 2002; N. Singh, Singh, Singh, & Singh, 2006; S. Singh, Kumar, Singh, Thind, & Mudahar, 2008). HVL, TVL and λ parameters are calculated by using Eqs. (5), (6), and (7), respectively:

$$\mathrm{HVL} = \frac{\ln 2}{\mu} \quad (5)$$

$$\mathrm{TVL} = \frac{\ln 10}{\mu} \quad (6)$$

$$\lambda = \frac{1}{\mu} \quad (7)$$

In conclusion, a material must have high μ / ρ value, low HVL, TVL and λ values for a good radiation shielding.

3. Experimental

3.1 Materials

Liquid and unmodified bisphenol A-epichlorohydrin epoxide resin (Code: AC510 UV, Taiwan) and the hardener isophorone diamine (Code: AC510 UV, Germany) were supplied from Armor Chemical. The micro powder PbO (CAS #: 1317-36-8, ACS reagent, ≥99.0%, USA) and BaO (CAS #: 1304-28-5, 97%, USA) were also purchased from Sigma Aldrich.

3.2 Preparation of the Pure Epoxy Resin Matrix

As is illustrated in Figure 1, 1.0 g liquid and unmodified bisphenol A-epichlorohydrin epoxide resin and 0.5 g isophorone diamine hardener solution was mixed mechanically at room temperature for 15 minutes to obtain homogenous pure epoxy solution. The mixing weigth ratio of epoxide and hardener was selected as 2 bisphenol A-epichlorohydrin: isophorone diamine.

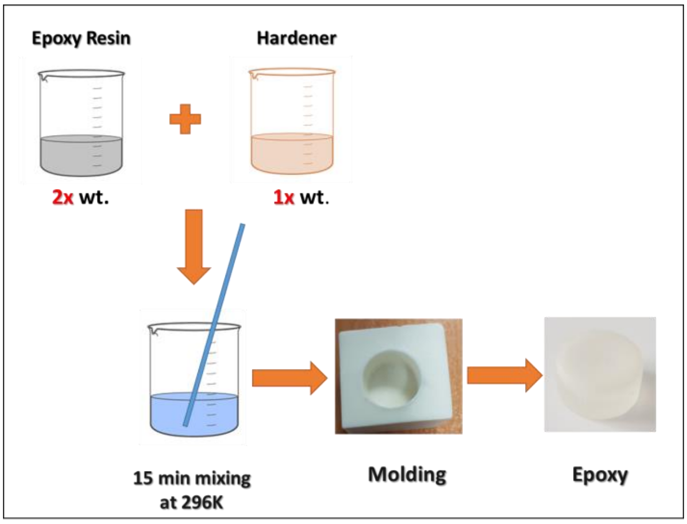

Figure 1. The preparation steps of the pure epoxy sample.

Finally, as shown in Figure 1, the solution was poured into a circular shaped teflon mold and the solution was dried in the mold for 24 h at room temperature.

3.3 Preparation of the Epoxy/BaO and Epoxy/PbO Composites

The Epoxy/BaO and Epoxy/PbO composites were also produced following the steps described above. As shown in Figure 2, initially the reiforcing materials (PbO and BaO powders) were grinded with agate mortar for 10 minutes. Then the reinforcing powders with different weight percentages were added to the 0.6-0.9 g liquid unmodified bisphenol A-epichlorohydrin epoxide resin and the mixtures were stirred in magnetic stirrer for 20 minutes. After the stirring process, the hardener was added (0.3-0.45 g) to the mixture with an appropriate masses and the resultant mixture was mixed mechanically for 15 minutes. As a last step, the final solution was poured into the mold and kept at room temperature for 24 hours.

Each samples was prepared with two different thickness for gamma ray spectroscopy experiments. While the radii of the samples were 10 mm; their thickness were measured between 1.8 mm and 2.4 mm.

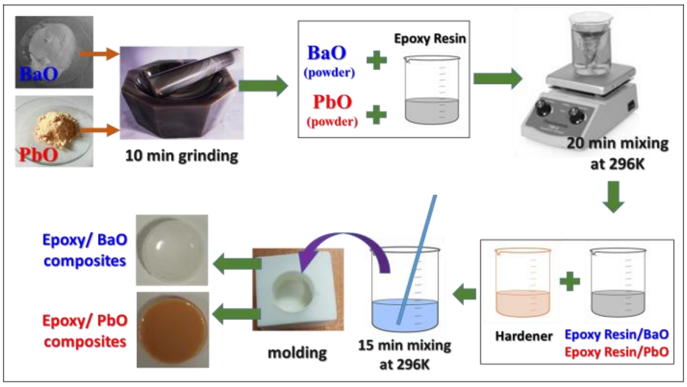

Figure 2. The preparation steps of the Epoxy/PbO and Epoxy/BaO composites.

The mass density of the radiation absorbing materials produced in the present study was calculated by using Archimedes' principle and the listed in Table 1. Additionally, the mass amounts of epoxy, PbO, and BaO in gram units were shown in Table 1.

Table 1. The mass amounts of epoxy, PbO, and BaO in the composites along with the mass density of these absorbing materials.

Sample	**Mass Amount (g)**			$\rho_{abs.\,mater.}$ **(g/cm³)**
	Epoxy+ Hardener	***PbO***	***BaO***	
Epoxy	1.500	--	--	1.255
Epoxy/10% PbO	1.350	0.150	--	1.423
Epoxy/20% PbO	1.200	0.300	--	1.633
Epoxy/40% PbO	0.900	0.600	--	1.969
Epoxy/10% BaO	1.350	--	0.150	1.388
Epoxy/20% BaO	1.200	--	0.300	1.502
Epoxy/40% BaO	0.900	--	0.600	1.763

The percentages in the heaviness of the samples were also calculated by Eq. (8):

$$\text{Heaviness}\,(\%) = \left[\frac{\rho_{absorbing\ material}}{\rho_{lead}}\right] \times 100 \qquad (8)$$

where ρ_{lead} is considered as the mass density of the reference absorbing material (lead) that has the maximum heaviness of 100%. The heaviness of each samples was compared with some of the conventional radiation shielding materials such as lead, steel, barite and ordinary concrete. For this comparison, the mass density values for the lead, steel, barite and ordinary concrete was taken as 11.34 g/cm^3, 7.85 g/cm^3, 4.48 g/cm^3, 2.3 g/cm^3, respectively (Eisenbud & Gesell, 1997; Ms, Mondal, & Tripathi, 2010). The heaviness of the samples prepared in this study was compared with the related materials in Figure 3.

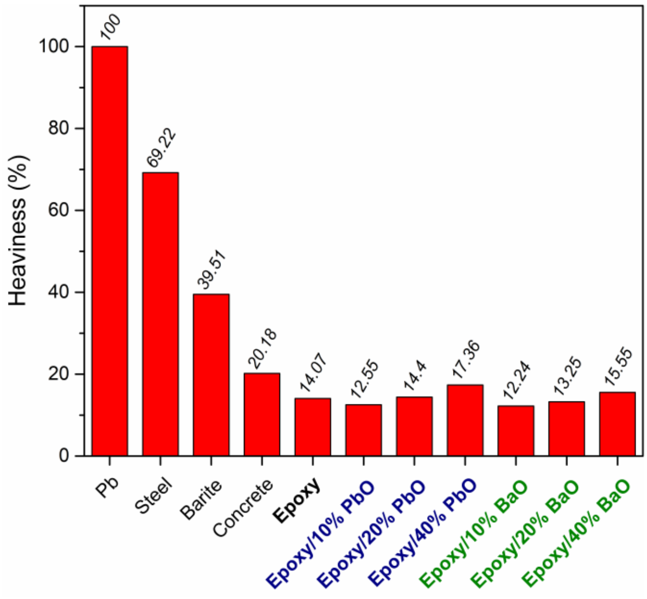

Figure 3. The heaviness of some conventional radiation shielding materials and the samples prepared in the present study.

As shown in Figure 3, the samples prepared in this study have a high low-weight advantage relative to these four conventional radiation shielders. Additionally, as it will be emphasized in Section 4, lead free epoxy composite with the higher BaO content has higher gamma ray absorption capacity than concrete at 81 keV and 356 keV.

3.4 Setup for Gamma Ray Spectroscopy Experiments

The experimental setup for determining the gamma radiation shielding ability of the samples was illustrated in Figure 4.

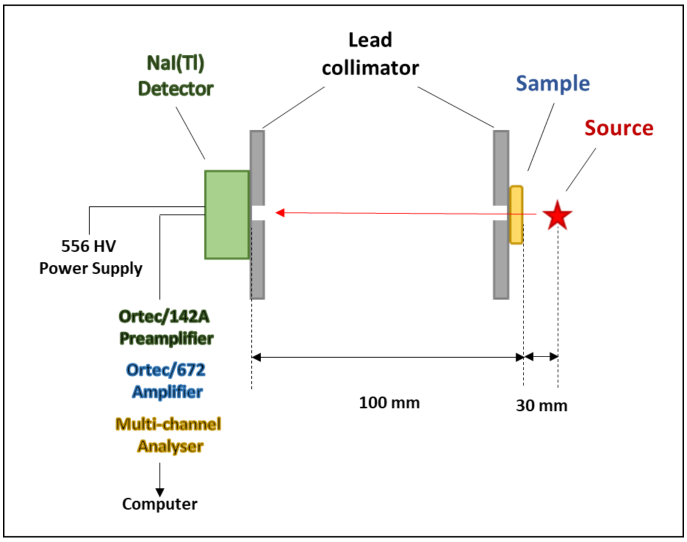

Figure 4. The experimental setup gamma ray spectroscopy measurements.

The mass attenuation coefficients of the samples were calculated by measuring an attenuated and un-attenuated intensities (I and $I_{\circ}$) given in Eq. (3) with NaI(Tl) detector. The dimension of the detector is 7.62 cm × 7.62 cm. The gamma rays emitted from the Ba-133 point radioactive source were 81 keV and 356 keV. The model of the NaI(Tl) detector was 905-4 Ortec-Amtek. The photomultiplier tube (PMT) base, digiBASE (Ortec) had also 6.3 cm diameter and 8.0 cm length. The FWHM was equal to 46 keV at 662 keV and 65 keV at 1330 keV. NaI crystal was separated from the PMT by a glass window with the thickness of 5 mm. The experimental data were analyzed by the Maestro software. Additionally it was collected into 2048 channels of the MCA. The experimental setup was also shown schematically in Figure 4.

4. Results and Discussions

The photon absorption abilities of the materials produced in the context of this study were discussed in terms of mass attenuation coefficients, HVL, TVL, and mean free path radiation shielding parameters. For this purpose, by using the experimental setup given in Figure 4, the attenuated and un-attenuated intensities (I and $I_{\circ}$) were measured experimentally for the photon energies of 81 keV and

356 keV. Then the mass attenuation coefficients of pure epoxy and epoxy based oxide composites were calculated according to Eq. (3). The additive weight percentage dependence of the mass attenuation coefficient of the epoxy was shown in Figure 5(a) and (b) for the 81 keV and 356 keV photon energies, respectively.

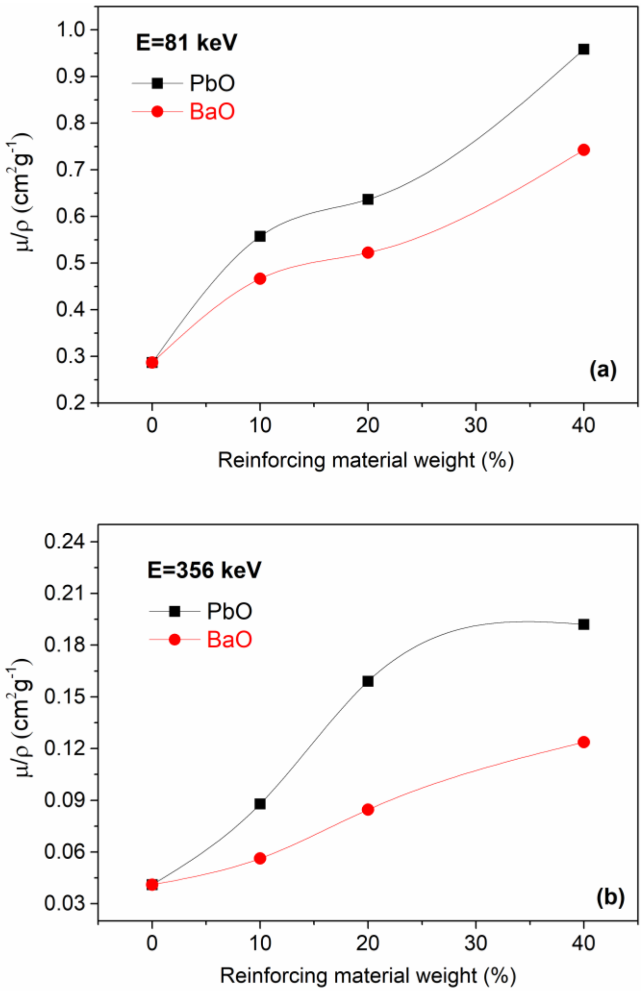

Figure 5. The variation of mass attenuation coefficient of the epoxy with different PbO and BaO reinforcing wt. percentages for the **(a)** 81 keV and **(b)** 356 keV photon energies.

As is clearly observed that since pure epoxy consists of elements with low atomic numbers (H, C, O and N), it has the lowest photon absorption ability which manifests itself as the lowest mass attenuation coefficients for the photon energies of 81 keV and 356 keV among other samples prepared (Li, Gu, Zhang, et al., 2017). However, it was revealed that the photon absorption performance of epoxy can be increased by using high Z metal oxide materials such as PbO and BaO at both photon energies (See Figure 5(a) and (b)). Moreover, it is worth to note that since $_{82}$Pb has higher atomic number than $_{56}$Ba, when the addition of the same weight percentages of PbO and BaO to epoxy is compared in the context of μ/ρ , PbO contribution to epoxy results in higher μ/ρ coefficient than that of BaO. On the other hand, it is a remarkable point that when the BaO additive percentage exceeds 40 wt. % in the epoxy matrix, the μ/ρ coefficient is greater than the μ/ρ coefficient of the 20 wt.% PbO doped epoxy (see Table 2). In other words, the better gamma ray shielding performance could be obtained with 40 wt.% BaO addition to epoxy instead of using 20 wt.% toxic PbO in the epoxy matrix. In table 2, the mass attenuation coefficients of the samples with the highest PbO and BaO additive percentage produced in the present work were compared with some conventional gamma ray shielding materials. According to Table 2, it was also determined that Epoxy / 40% BaO composite shields the gamma ray better than some traditional gamma ray screeners such as steel, bricks, cements, concrete, and Epoxy/Gd_2O_3 nanocomposites at both 81 keV and 356 keV. On the other hand, it was determined that 40 wt. % lead oxide added composite shows the best performance among the materials listed in Table 2 for both photon energies.

Table 2. Mass attenuation coefficients of some traditional radiation shielding materials reported in scientific literature along with the epoxy composite with high PbO and BaO content for the photon energies of 81 and 356 keV.

Material	**Mass attenuation coefficient (cm^2g^{-1})**	
	81 keV	*356 keV*
Steel SS304L (V. P. Singh, Medhat, & Shirmardi, 2015)	0.587*	0.099
Epoxy/20.1 wt.% Gd_2O_3 nanocomposite (Li, Gu, Wang, et al., 2017)	0.911*	0.103
Bricks (Tekin & Manici, 2017)	0.183	0.098
Cements (Tekin & Manici, 2017)	0.211	0.096
Concrete (Tekin & Manici, 2017)	0.200	0.101
Epoxy/40 % BaO	**0.743**	**0.123**
Epoxy/40 % PbO	**0.958**	**0.192**

* *The mass attenuation coefficient was reported for 80 keV* photon energies.

According to Figure 5, the mass attenuation coefficient of the samples decreased when the photon energy increases. While the high μ/ρ coefficient for the low photon energies is attributed to the high possibility of occurrence of photon absorption via photoelectric effect, the considerable low μ/ρ coefficient observed for the high photon energies is due to the reduced possibility of photoelectric effect and occurrence of Compton scattering. In Compton scattering process, the photons with high energy are attenuated via Compton scattering and the scattered photons with low energy are also absorbed by photoelectric effect (Li, Gu, Wang, et al., 2017). Hence, the mass attenuation coefficient decrease for the high energetic photons. On the other hand, the possibility of the occurrence of photon absorption via photoelectric effect is directly proportional to the Z^3 (Azman, Siddiqui, Hart, & Low, 2013). From this respect, determination of the higher mass attenuation coefficients for the epoxy composite including of lead oxide relative to that of the composite having barium oxide content is a natural consequence of this reality.

To determine the photon absorption ability of a radiation shielding material, along with the mass attenuation coefficient, the critical thicknesses of HVL and TVL are commonly used. The reinforcing material weight dependencies of HVL and TVL were shown in Figure 6 and 7 for the epoxy composites including PbO and BaO with different wt. percentages for the incident photon energies of 81 keV and 356 keV.

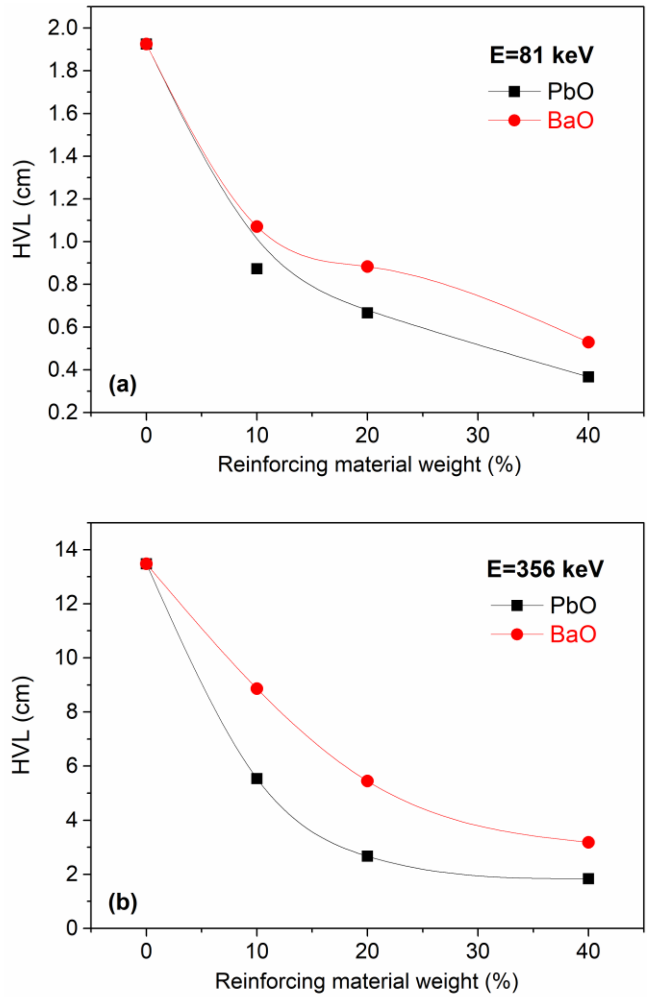

Figure 6. The HVL of the epoxy based composites at **(a)** 81 keV and **(b)** 356 keV photon energies.

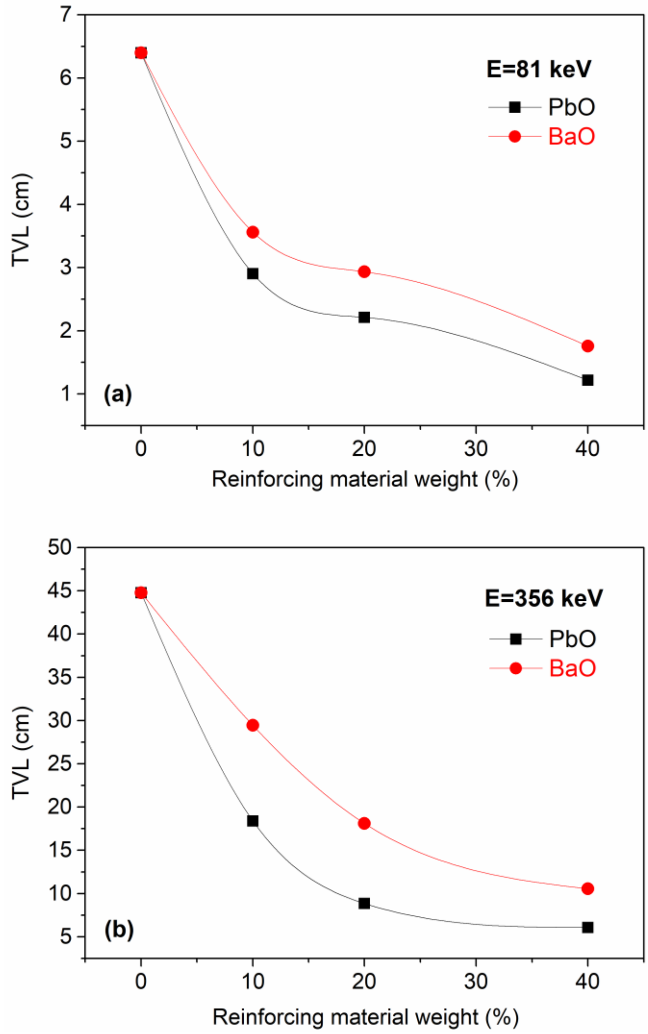

Figure 7. The TVL of the epoxy based composites at **(a)** 81 keV and **(b)** 356 keV photon energies.

In Figure 8, the variations of mean free path of the epoxy with reinforcing materials were also given for the 81 keV and 356 keV. When the HVL, TVL and mean free path parameters of all samples are interpreted together for both incident photon energies, the trends of the curves given in Figures 6-8 are in good agreement with the findings deduced from the mass attenuation coefficient versus reinforcing material weight curves shown in Figure 5. Since the values of HVL, TVL and λ parameters are accepted as the indicators of the radiation shielding performance of the materials, the lower HVL, TVL and λ values implies the better radiation shielding performance due to lesser volume requirements of the samples.

In conclusion, the photons having high energy will have more penetration within the epoxy based composites comparing to the photon having low energy. When the results are compared with a commercial polymer composite consisting of 15 wt.% polypropylene copolymer, 75 % iron oxide powder and 10 wt.% impact modifier (QUEO 8210), since the commercial polymer composite has the mean free path values of 2.549 cm (81 keV) and 4.694 cm (356 keV) and the HVL values of 1.767 (81 keV) and 3.254 (356 kev) (Büyükyıldız et al., 2018); it was revealed that both epoxy composites include 40 wt.% PbO or 40 wt.% BaO reinforcing material exhibit better gamma radiation shielding than the commercial composite for each photon.

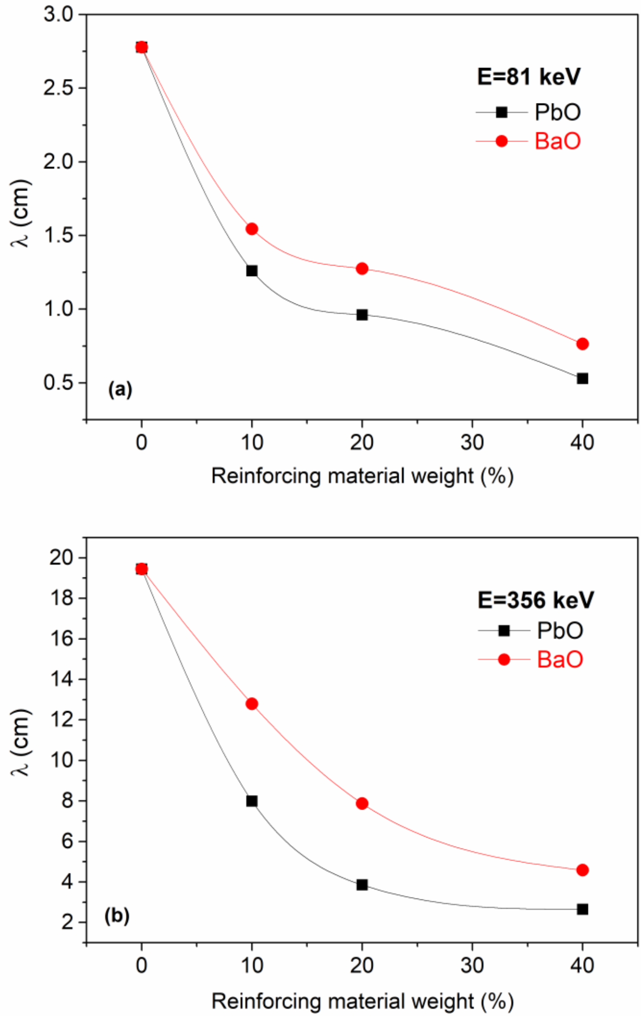

Figure 8. The change in the mean free path of the epoxy due to increasing PbO and BaO wt. % for the incident photon energies of **(a)** 81 keV and **(b)** 356 keV.

Additionally, as expected the Epoxy /PbO composites have lower HVL, TVL and λ parameters than that of Epoxy/PbO composites for both incident photon energies. In the light of all results, Epoxy/40 wt.% BaO composite can be suggested as an eco-friendly and low weight radiation shielding material alternative to some traditional radiation shielders.

5. Conclusions

In this study, the mass attenuation coefficient, half value layer, tenth value layer and mean free path of the epoxy polymer based composites which includes both lead oxide and barium oxide with different weight percentages were determined experimentally for the incident photon energies of 81 keV and 356 keV emitted from Ba-133 point radioactive source. The results related to the PbO and BaO added epoxy composites were compared with both each other and some conventional gamma ray shielding materials. As a result of this comparative study, it was deduced that 40 wt.% BaO added epoxy composite can be suggested as a non-toxic and low heaviness promising material for low energetic gamma radiation shielding material.

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