# A Novel Metal Oxide Filled Polyethylene Based Composite Shielding Material for Protection from Harmful Effects of Ionizing Electromagnetic Radiation

[*1]E. Eren Belgin, [1]G.A. Aycik
[1]Muğla Sıtkı Koçman University, Faculty of Science, Department of Chemistry, Mugla 48000, Turkey

## Abstract

In recent years, it is an important need to improve new shielding materials for protection from ionizing electromagnetic radiation. In this study metal oxide-polymer composites were produced and their ionizing electromagnetic radiation shielding effects were investigated by using HPGe detector. Powdered linear low density polyethylene was used as matrix material of the composites and filled with different amounts of powdered lead(II)oxide and tungsten(VI)oxide. The polymer reduced the heaviness of the shielding material and increased the flexibility while metal fillers acted as principle radiation attenuator in the composite. The shielding properties of the produced composites were compared with the conventional shielding material, lead. The highest radiation attenuation performance eliminated was %35 of lead while the composite had approximately 8.9 times lower density than lead and the performance was 2.5 times greater than the matrix material. Thus this non-toxic and light weight novel material could be an alternative to lead shielding.

**Key words:** Radiation shielding; polymer composites; metal fillers; radiological hazard; gamma radiation.

## 1. Introduction

Radiation is defined as the energy that can be travel through the matter and space. It is caused by self-transmutation of nuclei and can be classified as ionizing and non-ionizing according to its energy. Non-ionizing radiation has low energy while ionizing radiation like alpha, beta particles and gamma, X rays has high energy. The radiation with high energy can ionize atoms of the interacting matter. Hence to protect human and environment from harmful effects of high energy ionizing radiation appropriate shielding materials are used. Shielding material reduces the exposured dose by interacting the radiation itself and reducing the intensity of the radiation. There is no need of special shielding materials for particle radiation such as alpha and beta particles since they interact with matter strongly that makes their penetrating ability weak. However high penetrating electromagnetic radiation such as gamma and X-ray must be shielded by special materials. At the present time the most widely used electromagnetic radiation shielding materials are high density materials such as lead bricks and high density concrete. Other metal based shielding materials like tungsten, copper, bismuth, steel etc. are used as shielding material but lead superior over them due to its high density, high atomic number and low cost. Besides these lead have many important disadvantages that limits its application areas and usage such as really high toxicity and heaviness, low mechanic and chemical stability and inflexibility. Thus at the present it is an important need to improve new shielding materials and also to develop properties of the conventional shielding materials.

*Corresponding author: Ezgi Eren Belgin. Adress: Mugla Sitki Kocman University, Faculty of Science, Department of Chemistry, 48000, Mugla, TURKEY. E-mail: ebelgin@mu.edu.tr. Phone: +902522111502

In view of this, several researchers have been studying for developing new shielding materials for ionizing electromagnetic radiation. Some of them are searching for solutions by using polymer matrixes because of their flexibility and lightness. In these studies generally high atomic number fillers are used with polymer or cement matrixes. In a study, Harish et all. used unsaturated polyester resin as matrix material and used lead(II)oxide as filler. They found linear attenuation coefficient of the composite with 50% filler amount as 0.206 cm-1. Thus they reported that their composite's attenuation performance was better than cement, copper and silver [1]. Plionis et all. studied on polymer-bishmut bricks that could be used as an alternative to lead bricks and they reported their attenuation performances nearly same as lead bricks for radiation energies below 400 keV [2]. Nanomaterials are also reported as usable filler materials with polymer matrixes in literature [5, 6, 7, 8]. Also lead oxide fillers were studied with polymer matrix materials like polyethylene [9], ethylene-vinyl alkanite and ethylene-alkyl metacrilate copolymers [10], styrene butadiene rubber [11], polyethylene glycol [12], natural rubber [13], high density polyethylene [14], polystyrene [15] and epoxy [16].

In this study some metal-ceramic-polymer composites were produced and their electromagnetic radiation shielding effects were investigated. Commercial powdered polyethylene was used as matrix material of the composites and filled with different amounts of powdered lead(II)oxide (PbO) and tungsten(VI)oxide ($WO_3$). The polymer matrix was used to reduce the heaviness and increase the flexibility while metal fillers were used to increase radiation attenuation properties of the composites. The produced composites were tested gamma spectrometrically to determine their radiation shielding capacities.

## 2. Experimental

### *2.1. Constituents of the composites*

Linear low density polyethylene (LLDPE) was used as matrix material of the composites. LLDPE is a substantially linear thermoplastic that have significant numbers of short branches with density of 0.918-0.940 $gcm^{-3}$ and melting point of 122-124 $^oC$. LLDPE is a very flexible polymer that elongates under stress with high environmental stress cracking resistance. It has higher tensile strength, higher impact and puncture resistance than low density polyethylene that have long branching. It also has good chemical resistance while it has some disadvantages like hard processing property due to its narrow range of heat sealing. In this study matrix material LLDPE was procured commercially in powder form for this study.

In the study, powdered PbO and $WO_3$ were used as fillers of the composites. These fillers were selected due to their high atomic numbers, densities and crystal structures that increase radiation shielding performance of the material.The PbO (BDH Chemicals, England) and$WO_3$ (Sigma Aldrich, USA) was procured commercially in powder form.

### *2.2. Preparation of the composites*

All the filler and matrix materials were sieved to particle size of <25μm after oven drying to the constant weight. Mixtures with filler composition of 10, 15, 20, 25 and 30% (w/w) were prepared by using a dry powder rocking shaker at 30 rpm for 15 minutes after filler and matrix compositions were weighted sensitively. Then the mixed powders were taken into

stainless steel moulds with diameter of 21 mm and height of 5 mm. A basic melting moulding process is done in a constant temperature oven at 145°C for 12 hours. Mixture supplementation and pressurizing is done during melting to avoid from descents and spaces in the last product. Composites were taken out from the moulds after cooling and rasped to remove burrs.

### *2.3. Characterization of composites*

#### *2.3.1. Attenuation measurements*

The gamma spectrometric measurements were performed with a 110 $cm^3$ well-type HPGe detector coupled with a 64k channel analyser. The system had a resolution of 3.78 keV at 1.33 MeV gamma-ray peak of $^{60}Co$. The detector was housed in a lead shielding 10 cm thick lined with 1.5 mm thick tin and 1.0 mm thick copper in order to reduce the X-ray interferences.
The radiation attenuation measurements were done by using a point $^{60}Co$ source. The geometrical arrangement of the measurement system is given in Fig. 1.

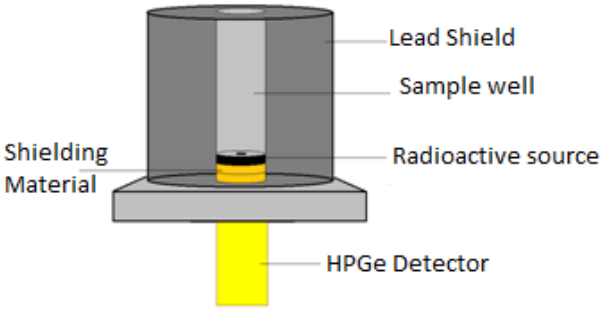


**Fig 1.** Geometrical arrangement of the measurement system

The produced composites with 10mm thickness were placed on a support in a lead shielding cylinder with 5 cm thickness as it is seen in Fig.1. The point source with known activity concentration was placed between the shielding materials and detector crystal.

The same measurement was done for lead to compare attenuation properties of lead with produced composites. At that time lead with the same geometry with the composites was placed between the detector and the point source, instead of composites.

A blank measurement was also done since knowing the gamma radiation intensity directly coming from the source would led us to calculate attenuation ratio of the materials. This time the source was placed on 10mm distance from the detector with the same arrangement seen in Fig 1 without placing any material between the source and detector. Using blank measurement results instead of the known activity of the source certificated was led us making relative calculations and eliminate the errors that could be caused from efficiency calibration of the detector.

The data acquisitions were performed for a period of 3000 seconds that gives <1% count error. The spectra were carried out using the software program Maestro-ORTEC. All the measurements were held 3 times and the mean values were used as the results.

#### *2.3.2. Attenuation rate calculations*

Every radiation type attenuates according to its energy, material properties that is passed through and thickness of the material. Attenuation rate is defined as the ratio of the incident intensity $I_0$of gamma rays to the intensity $I$ of gamma rays that successfully reaches to the detector by passing through the adsorbent material, Eq. 1.

$$F_N = I_0 / I \quad (1)$$

Where $F_N$ is attenuation rate, $I_0$ is intensity of radiation before entering to the material and I is intensity after leaving from the material.

In this study 3000 second data acquisition was performed for attenuation rate calculations. Intensities of the gamma rays entering to the material and leaving from the material were determined for 1173.2 and 1332 keV gamma energies of $^{60}$Co point source. Number of the counts per second (cps) recorded by the detector after placing composite materials or lead between the source and the detector were calculated by using net areas of the photopeaks. Thus cps values after leaving of gamma rays from the composite materials or lead were determined. The blank measurements, helded for the source without placing any other material between source and detector, were also analysed by the same way to calculate cps values of the gamma rays before entering to the composite materials or lead. After determination of all cps values attenuation rates were calculated by using Eq. 1.

### *2.3.3. Linear attenuation coefficient calculations*

Gamma rays interact with atoms, electrons or nuclei of the absorber material via various interaction processes and incident gamma rays can be absorbed by the material or scattered. Thus a beam of gamma rays with initial intensity of $I_0$ of primary photons will have residual intensity of I after traversing the material. This relation is given by Eq. 2.

$$I = I_0 \exp(-\mu x) \quad (2)$$

In Eq. 2 the parameter μ is total linear attenuation coefficient of the material ($cm^{-1}$) for gamma rays with appropriate energy and x is thickness of the material. According to Eq. 2, μ can be determined by using linear graph of $\ln(I/I_0)$ versus x of the material.

In the study the intensities of the gamma rays before entering to the materials and after leaving from the materials were calculated as it is mentioned in section 2.3.3 to evaluate linear attenuation coefficient.

### *2.3.4. Half value layer calculations*

Half value layer (HVL) is described as the thickness of the material at which intensity of the radi ation after interact with the material reduced to half of its energy before entering to the material. HVL is given with Eq. 3.

$$HVL = 0.693 / \mu \quad (3)$$

Where HVL value is in cm and μ is linear attenuation coefficient of the material.
In the study, the μ values for the materials were calculated as it is mentioned in section 2.3.4. Then HVL values were calculated for both composites and lead via Eq. 3.

### *2.3.5. Percent attenuation calculations*

Percent attenuation (%Att.) values of the composite materials were also calculated for basiccomparison of the attenuation performances of the composites with lead and with each other. Percent attenuation calculations were done via Eq. 4.

$$\frac{I_0 - I}{I_0} \times 100\% \quad (4)$$

Where $I_0$ is intensity of radiation before entering to the material and I is intensity after leaving from the material.

In the study, $I_0$ and I values for the materials were eliminated as it is mentioned in Section 2.3.3 and percent attenuation values were calculated by using Eq. 4.

## 3. Results and discussions

Densities, attenuation rates, linear attenuation coefficients and half value layers of the lead and produced composites were measured/calculated and results are summarized in Table 1 for 1173.2 and 1332 keV gamma energies.

$F_N$ values, linear attenuation coefficients, HVL values and % Att. values for produced composites and lead were calculated by using attenuation measurement results and Eq. 1, 2, 3 and 4, respectively as it is mentioned in Section 2.3.3 The eliminated results are summarized in Table 1 for both 1173.2 and 1332 keV gamma energies.

The linear attenuation coefficients of the composites were higher for $WO_3$ filled composites. Thus the $WO_3$ filler increased radiation attenuation performance of the polymer matrix much more than PbO filler.

**Table 1.**Attenuation rates, linear attenuation coefficients, HVL values of materials for 1173.2 keV and 1332 keV.

| Sample | 1173.2 keV Energy | | | | 1332 keV energy | | | |
|---|---|---|---|---|---|---|---|---|
| | **$F_N$** | **μ ($cm^{-1}$)** | **HVL (cm)** | **Att. (%)** | **$F_N$** | **μ ($cm^{-1}$)** | **HVL (cm)** | **Att. (%)** |
| Lead(pure) | 1,895 | 0,064 | 10,845 | 47,217 | 1,717 | 0,054 | 12,814 | 41,773 |
| LLDPE(pure) | 1,086 | 0,008 | 84,070 | 7,913 | 1,062 | 0,006 | 114,659 | 5,865 |
| LLDPE+10%PbO | 1,128 | 0,012 | 57,617 | 11,332 | 1,107 | 0,010 | 68,073 | 9,679 |
| LLDPE+15%PbO | 1,124 | 0,012 | 59,279 | 11,033 | 1,125 | 0,012 | 58,857 | 11,108 |
| LLDPE+20%PbO | 1,151 | 0,014 | 49,405 | 13,088 | 1,122 | 0,011 | 60,327 | 10,852 |
| LLDPE+25%PbO | 1,152 | 0,014 | 48,874 | 13,220 | 1,123 | 0,012 | 59,721 | 10,956 |
| LLDPE+30%PbO | 1,149 | 0,014 | 50,004 | 12,941 | 1,109 | 0,010 | 67,025 | 9,823 |
| LLDPE+10% $WO_3$ | 1,164 | 0,015 | 45,678 | 14,077 | 1,140 | 0,013 | 52,853 | 12,289 |
| LLDPE+15% $WO_3$ | 1,142 | 0,013 | 52,323 | 12,405 | 1,129 | 0,012 | 57,325 | 11,387 |
| LLDPE+20% $WO_3$ | 1,150 | 0,014 | 49,518 | 13,060 | 1,128 | 0,012 | 57,453 | 11,363 |
| LLDPE+25% $WO_3$ | 1,198 | 0,018 | 38,341 | 16,535 | 1,171 | 0,016 | 43,823 | 14,627 |
| LLDPE+30% $WO_3$ | 1,184 | 0,017 | 41,014 | 15,546 | 1,146 | 0,014 | 51,009 | 12,703 |

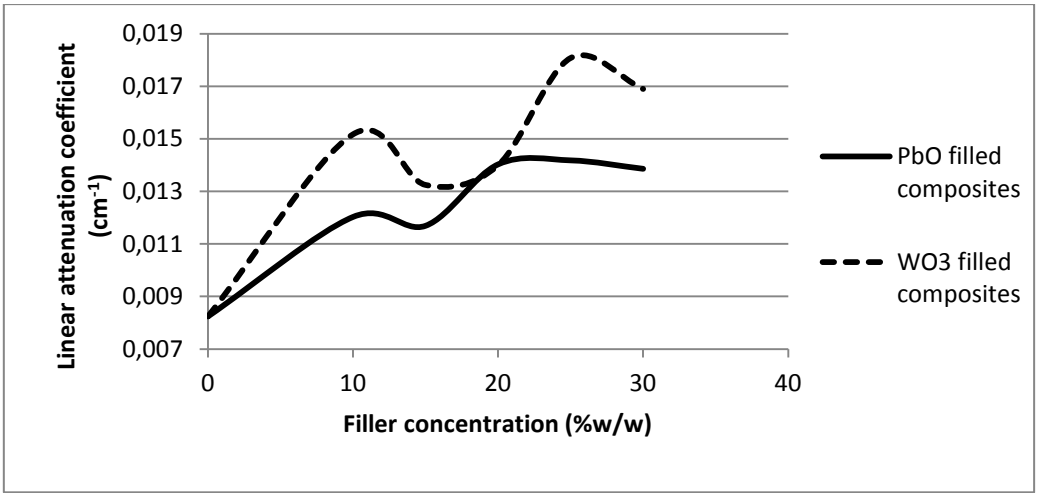


**Fig 2.** Change of linear attenuation coefficient of composites with respect to filler type and filler concentration

Generally a regular increment tendency is expected for attenuation performances of the composites as the filler concentration is increased. But as it is seen in Fig. 2 There is a performance decrement for both filler types between 10-20% filler concentration and above 30% filler concentration. Because both of chemicals showed the same tendency, this is not resulted in experimental error sources, it may be sourced from the chemical bonding in these filler concentrations.

Another way for comparison of composites radiation attenuation performances was comparing HVL values of the composites, Fig. 3.

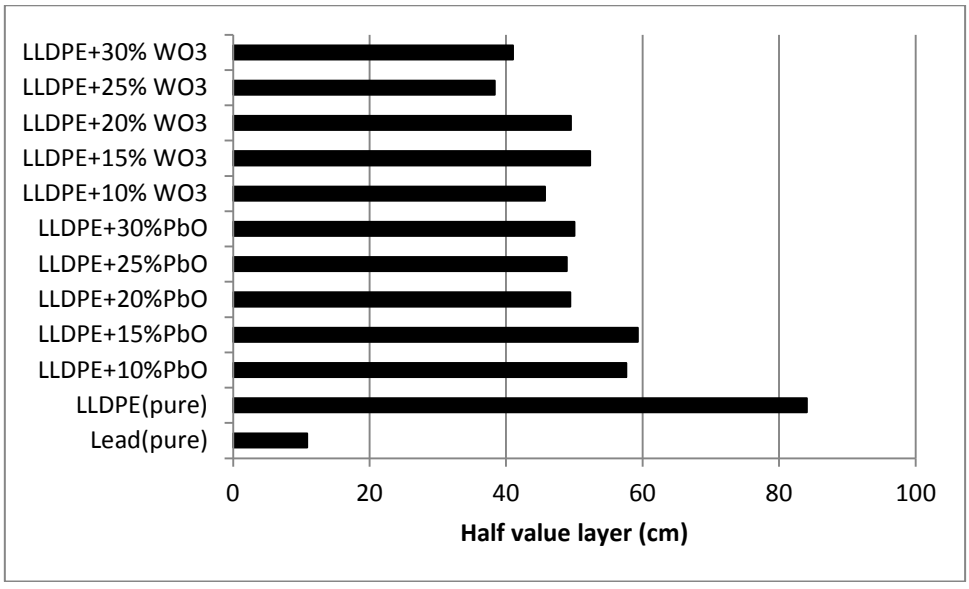


**Fig. 3.** Half value layers of the composites

According to the results, for attenuation of radiation with certain energy to a definite (half of its initial intensity) intensity 38 cm of the 25% $WO_3$ filled composite (the composite with best performance) is necessary while 11 cm of pure lead has enough thickness. Thus nearly 3.5 cm of produced composite has the same radiation attenuation performance with 1 cm pure lead.

On the other hand, the produced composite is nearly 8.9 times lighter than pure lead ($11.34 gcm^{-3}$) with density of 1.28 $gcm^{-3}$. As a result in the case of using suggested composite the heaviness of the shielding material would be reduced nearly 2.5 times for same radiation attenuation performance.

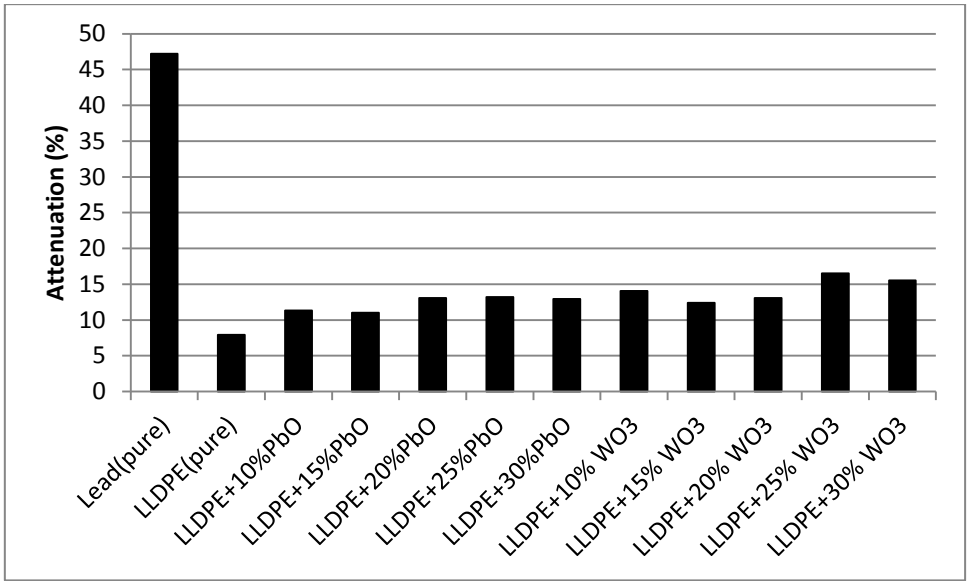


**Fig 4.** Percent attenuation of the composites

The percent attenuation performances for 1 cm of all the composites are shown in Fig. 4. The performance of the matrix material (LLDPE) was increased with metal oxide filler additions. This increment is nearly 2.5 times for the composite showing best attenuation performance. Also this composite showed 35% of performance of pure lead, Table 1.

## 4. Conclusion

The produced composites have superiority on lead with non-toxicity, flexibility, high chemical and mechanic stability because of the thermoplastic matrix material (LLDPE) of the composites.

This non-toxic, flexible and nearly 2.5 times light weight novel material could be an alternative to conventional shielding material lead by considering disadvantages of lead like really high toxicity and heaviness, low mechanic and chemical stability and inflexibility. This shield could be used for both stationary applications as radioactive source shielding and also mobile applications because of its light weight. Light weight, flexible and comfortable clothing shield materials could be produced for radiation workers by using this material. Thus this will be very effective result by ionizing-radiation workers and by the comfort of general population.

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