Application of Epoxy/ Pb$_3$O$_4$ Composite for Gamma Ray Shielding

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ABSTRACT

This paper aims to design types of shielding materials composites to protect public and personnel from the effect of scattered radiation during radiotherapy. It was found that 20% of lead oxide (Pb$_3$O$_4$) is the maximum percent finally mixed with Epoxy preserving good mechanical properties. Thermal characterization was discussed using thermal gravimetric analysis. Mechanical properties were studied by measuring hardness. Attenuation of gamma rays was carried out on synthesized composite, while some parameters such as thickness of composite, different sources and energies were discussed. Some shielding factors were measured such as half value layer (HVL), tenth value layer (TVL), attenuation coefficient ($\mu$) and relaxation length ($\lambda$). Durability of the composite was determined by irradiation with gamma rays up to 500 KGy. Also, the change in gamma attenuation was carried out. Maximum calculated values of $\mu$ is 0.135 cm$^{-1}$, $\lambda$ is 7.61 cm, HVL is 5.66 cm and TVL is 17.06 cm were shown in case of $^{22}$Na source for 0.551 keV energy which proved the availability for using the Pb$_3$O$_4$ composite as a gamma-ray shielding.

Key words: Gamma-ray attenuation/ Shielding, Epoxy/ Pb$_3$O$_4$, Half value layer, Relaxation length

INTRODUCTION

With the increasing usage of gamma-ray active isotopes in industry, medicine and agriculture, it has now become necessary to study mass attenuation coefficients in various materials of technological and biological importance. There is always a need to develop material, which can be used under harsh conditions of nuclear radiation exposure and can act as shielding material [1]. For nuclear radiation shielding, a larger quantity of shielding material is required; therefore, study of propagation of radiation flux in shielding materials is an essential requirement for shield design [2].

The interaction of gamma-rays depends on the incoming photon energy [3], it is difficult to shield gamma-rays as it has no mass or charge. The linear attenuation coefficient, which is defined as the probability of a radiation interacting with a material per unit path length, is the most important quantity characterizing the penetration and diffusion of gamma-rays in a medium. Its magnitude depends on the incident photon energy, the atomic number and the density ($\rho$) of the shielding materials [4]. Several studies have been performed to obtain linear attenuation coefficients theoretically and experimentally for different elements, compounds and mixtures [5] for different building materials [6], concrete [7,8] and some aqueous solutions [9].

Concrete has many desirable properties from a nuclear radiation shielding point of view. These properties can be changed to suit special shielding needs [10], these needs fall into one or more of the following three categories:
Higher neutron attenuating performance, (2) Higher gamma ray attenuating performance and (3) Higher temperature performance.

Many investigators prepared polymeric composites containing inorganic additives to study their shielding properties [11-13]. One of these additives is PbS; a high electron density material [14]. Molten sulphur was loaded with lead oxide to form shielding materials. Polymeric nanocomposites with a controlled microstructure in a scale of nanometer to sub-micron have a great potential for producing highly functional materials and unusual physical properties [15–22].

In this study, preparation of Pb3O4/epoxy composite to form shielding materials was carried out. Thermal and mechanical properties proved their availability for gamma ray shielding application. Shielding of bi-line energy sources such as 22Na and 60Co were discussed and also for single line energy (137Cs). Durability of shielding materials was performed through irradiation by gamma ray till 500 kGy.

**EXPERIMENTAL WORK**

The samples were prepared using phenyl epoxy and their solidifying agents of commercial grade were purchased from Optco, Egypt. Lead oxide of analytical grade was purchased from Merck Co. The chemical formula of phenyl epoxy is shown in figure 1. It consists of standard bisphenol-A of technical purity 95% weight as a resin and poly-oxy propylene diamine as a hardener, while erosail was added to prevent fillers from sedimentation. Lead oxide filler was first dispersed with equal particulate sizes in range of nano particles and loading fractions in the epoxy formula. The mixture was stirred at room temperature and heated to 80°C for 30 minutes. The mixture was degassed to allow the entrapped air to be released, then poured with great care into the specimen moulds and left to cure. The specimen moulds were made of Teflon with different geometries. These moulds guarantee the dimensions of different shapes which suits different examinations. After 24 hours, the samples were extruded from moulds and 7 days later were processed either by shaping or machining to obtain the final ASTM dimensions

**Fig. 1: Chemical formula of phenyl epoxy**

Thermal gravimetric analysis (TGA-50, Shimadzu, Japan) was used to characterize the polymer composite thermally. Sample weight used was few milligrams, while thermal scanning was from room temperature to 600 °C.

Square shape of polymer composite of dimension not less than 20(L) x 20 (W) mm was used to be examined by analogue manual instrument of hardness tester with thin pin manufactured by Baxio USA, while durability was examined by measuring the attenuation coefficient, HVL, TVL and λ after gamma irradiation of shielding materials for 50, 100, 200 and 500kGy for the calculation of change of the previous factor.

Experimental measurements have been performed to investigate attenuation properties of gamma rays for the five Pb3O4/epoxy composites. Test samples with different thicknesses were arranged in front of a collimated beam emerged from gamma ray sources (5 mCi for each) 137Cs
(E1=0.662 MeV), $^{60}$Co (E2=1.173, E3=1.332 MeV) and $^{22}$Na (E4=0.2745, E5= 0.551 MeV). E1, E2, E3, E4 and E5 were mentioned to distinguish between results of $^{137}$Cs, $^{60}$Co and $^{22}$Na sources.

The gamma-ray attenuation measurements were performed using Sodium iodide NaI(Tl) scintillation detector. The detector signal was amplified and fed to the Genie 2000 (Gamma Acquisition & Analysis Software, Canberra). The transmitted gamma ray fluxes through the investigated samples at the related energies were used to estimate the linear attenuation coefficients $\mu$ (cm$^{-1}$) at definite energies. Values of mass attenuation coefficients $\mu / \rho$ (cm$^2$ g$^{-1}$) were estimated from the calculation of the composites density $^{[23,24]}$.

RESULTS AND DISCUSSION

1- Thermal gravimetric analysis

Temperature at which the loss in weight occurs is considered to be the thermal resistance of polymer $^{[25-26]}$. An improvement of thermal characteristic was detected for the composite of Pb$_3$O$_4$ with epoxy and for the blank one. The blank epoxy showed high thermal resistance reaching 550$^\circ$C above which loss of weight was abruptly noticed in the range of 550$^\circ$C to 600$^\circ$C while complete decay was noticed close to 600$^\circ$C, this is due to the homogenous distribution of lead oxide into epoxy which is confirmed also by absence of side peaks as it is seen in figure 2. The TGA curve can be divided into three sections. First section describes the working temperature of epoxy showing high stability reached to 515$^\circ$C at which loss of weight was 2%, this may be due to water loss. Second section illustrated convex zone ended at 560$^\circ$C at which loss of weight was found to be 13%. The third division showed abrupt change in weight describing the isotherm's tail which ended at 13% from the original weight at 600$^\circ$C.

![Graph of Thermogravimetric analysis](image)

Fig. 2: Thermogravimetric analysis of epoxy.

Figure 3 represented the Thermogravimetric analysis of epoxy/lead oxide composite. The Thermograph showed stability till about 350$^\circ$C at which no significant loss of weight appears (only ~5% loss). It was also found that an abrupt change in weight appears as the temperature rises from 350$^\circ$C to 400$^\circ$C. The third division as shown in figure 3 explains the change in weight (loss of weight was 17%) resulted from the raising temperature from 400$^\circ$C to 580$^\circ$C. So an improvement of thermal stability was noticed obviously by the processing of composite of Epoxy with lead oxide. The results proved the homogenous distribution of lead oxide into epoxy which is also confirmed by the absence of side peaks.
2- Shielding against gamma-rays:

The shielding of gamma rays requires large amounts of mass. They are better shielded by materials with high atomic numbers and high density, although neither effects are important compared to the total mass per area in the path of the gamma rays. For this reason, a lead shield is only modestly better (20-30%) as a gamma shield than an equal weight of another shielding material such as aluminum, concrete, or soil; the lead's major advantage is in its compactness. The higher the energy of the gamma rays are, the thicker is the required shielding. Materials for shielding gamma rays are typically measured by the thickness required to reduce the intensity of the gamma rays by one half (HVL). When a gamma ray passes through matter, the probability for absorption in a thin layer is proportional to the thickness of that layer. This leads to an exponential decrease of intensity with thickness. The exponential absorption holds only for a narrow beam of gamma rays. If a wide beam of gamma rays passes through a thick slab of concrete, the scattering from the sides reduces the absorption by the following Lambert’s law.

\[
I(d) = I_0 \cdot e^{-\mu d}
\]

where, \(\mu = N\sigma\) is the absorption coefficient, measured in \(cm^{-1}\), \(N\) is the number of atoms per \(cm^3\) in the material, \(\sigma\) is the absorption cross section in \(cm^2\) and \(d\) is the thickness of material in cm.

Fig. 3: Thermogravimetric analysis of Epoxy composite with 20 % \(Pb_2O_4\) gamma shielding

One of the most interaction mechanisms is an interaction in which an incident gamma photon loses enough energy to an atomic electron to cause its ejection, with the remainder of the original photon's energy being emitted as a new and lower energy gamma photon with an emission direction different from that of the incident gamma photon, it is termed Compton scattering. The probability of Compton scatters decreases with increasing photon energy. Compton scattering is thought to be the principal absorption mechanism for gamma rays in the intermediate energy range 100 keV to 10 MeV.

The result obtained for gamma ray intensities transmitted through slaps of irradiated lead oxide loaded epoxy are represented in figures 4, 5 and 6. The data obtained are represented as group of spatial distribution of gamma of gamma-rays. It can be shown from the figures that the gamma-rays intensities decrease as the slab thickness. The total gamma-rays are estimated from the slope of the curves as shown in table 1.
Table 1: variations of HVL, TVL, μ and λ with different sources of gamma ray.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Gamma –ray energies (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.551</td>
</tr>
<tr>
<td>μ (cm⁻¹)</td>
<td>0.135</td>
</tr>
<tr>
<td>λ (cm)</td>
<td>7.407</td>
</tr>
<tr>
<td>HVL (cm)</td>
<td>5.13</td>
</tr>
<tr>
<td>TVL (cm)</td>
<td>17.05</td>
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The materials shielding has an affinity to some extent towards the attenuation of $^{60}$Co at 1332.5 keV more than that of $^{22}$Na at less energy (1274.6 keV). The shielding materials tend to attenuate double range energies source more than single energy. The energy of single energy source ($^{137}$Cs) is less than that of double energy source ($^{22}$Na & $^{60}$Co).

![Graph showing the variation of gamma intensity with unirradiated lead oxide loaded epoxy using $^{60}$Co source.]

**Fig. 4:** The variation of gamma intensity with unirradiated lead oxide loaded epoxy using $^{60}$Co source.

![Graph showing the variation of gamma intensity with unirradiated lead oxide loaded epoxy using $^{7}$Cs source.]

**Fig. 5:** The variation of gamma intensity with unirradiated lead oxide loaded epoxy using $^{7}$Cs source.
Fig. 6: The variation of gamma intensity with unirradiated lead oxide loaded epoxy using $^{22}$Na source.

Table 2 presents the values of half-value layer and linear attenuation coefficient for different gamma energies in ordinary concrete, steel and lead shielding materials\cite{27,28} with the present data for comparison. By comparing the attained results by polymer composite with concrete, Cs-137 and Co-60 shielding, it showed very close results, however, composite polymer has an advantage over concrete because of its mobility, less electrical properties and the ability to avoid neutron emission. It neither makes magnetic nor electrical field influencing human health of the users and/or patients.

**Table 2: Half-value layer and linear attenuation coefficient for different gamma energies in ordinary concrete, steel and lead shielding materials.**

<table>
<thead>
<tr>
<th>Radioisotope</th>
<th>Gamma Energy (MeV)</th>
<th>Half-Value Layer (cm)</th>
<th>Linear attenuation coefficient, $\mu$ (cm$^{-1}$)</th>
</tr>
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<tbody>
<tr>
<td></td>
<td></td>
<td>Ordinary Concrete</td>
<td>Steel</td>
</tr>
<tr>
<td>Cesium-137</td>
<td>0.661</td>
<td>4.8</td>
<td>1.6</td>
</tr>
<tr>
<td>Cobalt-60</td>
<td>1.1732, 1.3325</td>
<td>6.6</td>
<td>2.1</td>
</tr>
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</table>

Gamma irradiation is an efficient factor of the polymer composite to be used as shielding materials. Measuring $\mu$ of the composites at different doses of gamma irradiation can be considered. The change in linear attenuation coefficient ($\Delta\mu$) for the polymer composite samples irradiated with different gamma irradiated doses is shown in table 3. From the table, It can be seen that there is a dramatic change in attenuation coefficient for all gamma sources. Increasing irradiation doses conducts less attenuation coefficient while first step of irradiation (50 kGy) leads to the maximum decreasing percentage. Gradual decrease of the shielding was noticed with the least steps of irradiation doses (100, 200 and 500 K Gy). Polymer composite shielding materials have maximum durability to gamma irradiation for single energy source used (Cs-137). Regarding bi-energy line source; the composite is effective for higher energy than lower one (for Co-60 and Na-22). In general the polymer composite is preferred to be a shielding material of higher energy for long times of work (over 100 KeV) than that of lower energy source.
Table 3: The change in linear attenuation coefficient ($\Delta \mu$) for the polymer composite samples irradiated with different gamma irradiated doses.

<table>
<thead>
<tr>
<th>Irradiation dose, kGy</th>
<th>Gamma-ray energies (MeV)</th>
<th>0.551</th>
<th>0.661</th>
<th>1.1732</th>
<th>1.2745</th>
<th>1.3325</th>
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<td>44</td>
<td>61</td>
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Irradiation by gamma rays of Epoxy/lead oxide composite plays a very important role in hardness. Hardness increases with irradiation to the level of 100 KGY which is due to complete cross-linking of the polymer and lead oxide. Hardness begins to decrease by the increasing irradiation dose which may be explained by some deterioration of the composite started to occur. Decreases of hardness is very low by irradiation with 200 KGY while it decreased more significantly as it is seen in figure 7 by irradiation to 500 KGY. Hardness decreases from starting point (zero irradiation) to 500 KGY does not exceed 4.6 %.

Figure 6: Variation of hardness of shielding materials after irradiation with different doses.

CONCLUSION

The present work includes synthesis of lead oxide/epoxy composite for gamma irradiation shielding purpose, which has more advantages than that of the traditional shielding materials by its mobility, less reactivity and less electrical properties. It revealed good thermal stability and mechanical properties which increased their availability for applications. Shielding of gamma rays has a good efficiency in the range of 660 to 1371 keV. Shielding of bi-energy source is more efficient than single energy source, while by the irradiation of shielding materials by single energy source, it will be more durable than that by irradiation of bi-energy source. The hardness decrease as a result of gamma irradiation of materials used in attenuation up to 500 kGy irradiation is very small which could be neglected.
REFERENCES