Lead oxides filled isophthalic resin polymer composites for gamma radiation shielding applications

V Harish1*, N Nagaiah2 & H G Harish Kumar3

1Department of Physics, Govt. First Grade College, Shimoga 577 201, India
2Department of Physics, Bangalore University, Bangalore 560 056, India
3Department of Physics, R V College of Engineering, Bangalore 560 056, India
*E-mail: harishvenkatreddy@radiffmail.com
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Particulate polymer composites of isophthalic resin (ISO) filled with three lead oxides (PbO, PbO2 and Pb3O4) were prepared with different filler concentrations and investigated for radiation shielding properties of gamma rays from Ba-133, Cs-137 and Co-60 point sources. Among the three composite combinations, ISO+PbO composites yield better density values than the other two due to high density of the filler which contains large lead fraction and fine dispersability in the polymer matrix. Dispersion and particle size of filler were also studied by morphological analysis of the composites using Scanning Electron Microscope. Attenuation coefficients were found to increase with the increased filler content in the composites. ISO+PbO composites have found to outperform than ISO+PbO2 and ISO+Pb3O4 composites. Excellent performance of ISO+PbO composites can be attributed to their high lead content as well as fine filler dispersion in the polymer matrix. Further, these ISO+PbO composites when compared to conventional shielding materials perform as strong contenders to barite, steel and concrete at low gamma ray energies. Even at higher gamma ray energies considered, they perform satisfactorily and are very much comparable to steel and concrete.

Keywords: Polymer composites, Radiation shielding, Gamma rays, Unsaturated polyesters, Attenuation

1 Introduction

An appropriate radiation shielding material is always necessary to protect life and other materials from degrading effect due to harmful radiations such as X-rays, γ-rays etc., emitted from unshielded sources1. High density materials such as lead bricks or high density concrete etc. are often used for this purpose. Even though lead is being superior over all other shielding materials due to its high atomic number, density and low cost2, it lacks in usage flexibility, chemical stability, mechanical strength, etc. When flexible shields are required, composites made of metal or metal oxide filled polymers are preferred. Literature survey reveals that variety of polymers with appropriate fillers have been used in preparing radiation shielding composites. In the present work, authors have chosen isophthalate resin as polymer matrix in view of its good mechanical, thermal and corrosion resistance characteristics3 and prepared its composites using three oxides of lead4 separately as filler for the purpose.

2 Experimental Details

2.1 Material Fabrication

Lead oxide powders such as lead monoxide (PbO), lead dioxide (PbO2) and lead tetroxide (Pb3O4), isophthalate resin, accelerator and catalyst were of commercial grade and used without any modifications. Composites of filler weight % of 0, 5, 10, 20, 30, 40 and 50 were prepared in laminate form using open mould casting technique at room temperature5 and post cured at 80°C. Scanning Electron Microscope (SEM) pictures of the neat and the filled composites are shown in Fig. 1.

2.2 Radiation Attenuation measurement

Generally, γ-photons incident on an absorber material may either be absorbed or scattered in a single event in various interaction processes with atoms, electrons or nuclei of the absorber, due to which a fraction of the incident photons is absorbed completely while the rest are transmitted with their full energy. Thus composites with a fine dispersion of high density filler should offer more interaction probability for photons and hence, better shielding properties. A well collimated beam of γ-rays of initial intensity I0 after traversing a thickness x of absorber will have a residual intensity I of primary photons equal to I=I0e−ux. Where u is the total linear attenuation coefficient6 (cm−1) of the absorber for γ-rays of appropriate energy. Mass attenuation coefficient6 is obtained by dividing u by density (ρ) of
the absorber. It is independent of the actual density and physical state of the absorber. Linear attenuation coefficients of the composite samples were measured for Ba-133 (80 keV and 356 keV), Cs-137 (662 keV) and Co-60 (1173 and 1332 keV) point sources using a gamma ray spectrometer (EG&G, ORTEC) with a narrow beam geometry set-up.

3 Results and Discussion

3.1 Linear attenuation coefficient

The measured values of linear attenuation coefficients increase with increasing filler concentration in all the three composites at all gamma ray energies. With reference to the filler type, the PbO filled composites have been observed to give better attenuation values than the other two fillers.

3.2 Mass attenuation coefficient

Mass attenuation coefficient facilitates an easy comparison of radiation shielding efficiency of different shielding materials. They were shown as a function of filler weight fraction in the composites for 80 keV gamma ray energy as shown in Fig. 2. Similar trend was also observed at other energies under consideration.

With an increase in energy (say 80 keV to 1337 keV) the mass attenuation coefficients decrease at each filler concentration in all the three composites. The interaction cross-sections decrease with increase in gamma ray energy. The cross-sections for photoelectric interactions are sufficiently high at energies lower than 500 keV in most of the absorbing materials. Between 100 keV to 10 MeV, cross-sections for Compton scattering are significant and above 2 MeV pair production process becomes dominant. Only in photoelectric absorption, the incident gamma photons which interact are completely absorbed and as many photons are straight away removed from the incident flux. But the situation is not so in Compton scattering and pair production, where photons are not completely absorbed.

The mass attenuation coefficients increase with increase in filler concentration in all the three composites. This fact may be attributed to the increasing lead content in the composite as well as the filler dispersability in the polymer matrix. These observations show that PbO filled composites perform better at all the gamma ray energies than the other two composites.

3.3 Effect of lead content

Theoretical mass attenuation coefficients for each element present in the composites (say, carbon, hydrogen, lead and oxygen) were also evaluated using WinXCom code and XMuDat code. The results shown in Fig. 3 depict that, within the gamma ray energy limits chosen for the experiment, the contributions to mass attenuation coefficients from carbon, hydrogen and oxygen present in the composites appear more or less constant and further do not vary appreciably with an increase in their concentration because of lack of any resonance absorption edges for photoelectric absorption. But, in case of lead the mass attenuation coefficients vary with gamma ray energies. Hence, the mass attenuation coefficients are expected to increase with an increase in lead concentration in the composites. In addition to
this, larger values observed at low energy are due to the presence of a resonance absorption peak called K-edge for photoelectric absorption at about 88keV. Hence the effective mass attenuation coefficients of the composites depend on relative concentrations of each of the individual elements present in the composites and further depend largely on the lead concentration.

The mass attenuation coefficients as a function of gamma ray energies are shown in Figs 4 to 6 for ISO+PbO, ISO+PbO$_2$ and ISO+Pb$_2$O$_3$ composites, respectively. Figs 4-6 show that mass attenuation coefficients depend on the gamma ray energy which have larger value at lower energies and decrease rapidly towards higher energies. At all energies, the PbO filled composites show better values of mass attenuation coefficients when compared to other two fillers at all concentrations.

### 3.4 Half Value Layer
Discussion made so far is for comparative purpose. But much before these, we need the half value layer (HVL) values which decides applicability of these materials for radiation shielding applications. HVL is nothing but the thickness of the radiation shielding material required to either absorb or reduce the incident intensity of the gamma radiation to its half at a given energy. Numerically,

$$\text{HVL} = \frac{\ln 2}{\text{Linear attenuation coefficient}}$$

The HVL values for the conventional shielding materials as well as the ISO+PbO composites were evaluated for different gamma ray energies considered as shown in Fig. 7. It is evident that performance of lead and barite concrete is exceptional.
at all energies. The HVL values of steel and concrete are not so encouraging at low energies but appreciable and comparable to those of barite concrete at higher energies. In case of ISO+PbO composites, the composite with 50% filler performs exceptionally well with HVL values comparable to those of barite concrete and much better than steel and concrete at lower gamma energies. At higher energies, its HVL values are very much equal to those of concrete. Further, in ISO+PbO composites, with a decrease in filler concentration, obviously the HVL values are expected to increase at all energies. Composites with filler concentration of 30% and above have very good HVL values than those of steel and concrete at low gamma ray energies.

3.5 Heaviness

Finally, the beauty of the polymer composites lies with their lightness. In general, the composites are known for exceptionally high performance against their very low density or lightness. To verify the heaviness of the ISO+PbO composites, lead was assumed as standard and normalized to 100%. With reference to lead, the % of heaviness of the other conventional shielding materials along with ISO+PbO composites was evaluated using the following relation as shown in Fig. 8.

\[
\text{% of heaviness} = \left( \frac{\text{density of material}}{\text{density of lead}} \right) \times 100
\]

With lead at 100% heavier than other shielding materials under consideration, steel is at 69.47%, barite at 31.42% and concrete only at 21.23% heavier of lead. In case of ISO+PbO composites, even 50% filled composite is only 19.36% of lead while unfilled polymer is only 10.62% of lead. These results prove that the polymer composites considered exhibit excellent lightness when compared to conventional radiation shielding materials such as lead, steel, barite and concrete. At the same time, their performance as radiation shielding materials is appreciable particularly at higher filler concentrations (preferably 30% and above) and for low energy gamma rays.

4 Conclusions

Among the three composites, ISO+PbO composites show better density, fine filler dispersability and perform better than other two. Their performance is comparable to barite, steel and concrete at low gamma ray energies and perform satisfactorily at higher gamma ray energies considered.

References