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Investigation of Gamma Shielding Parameters for different High Density Polyethylene (HDPE) fillers.

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ABSTRACT

This research is about studying the effect of adding some oxides, specifically cadmium and lead oxides at different substitution ratios to high-density polyethylene (HDPE) on the radiation protection properties of gamma rays. Based on this, we find that the polymer is characterized by the light weight of the protective material and increased flexibility in handling, while the lead and cadmium oxide fillings play their role as basic radiation attenuators in the polymer compound. High-density polyethylene (HD-PE) polymer composite samples filled with lead and cadmium oxides in the filler weight ratios of [5%, 10%, 15% and 20%) were prepared. Then, the HD-PE samples filled with (5% by weight, 10% by weight, 15% by weight, 20% by weight) micronized cadmium oxide and micronized PBO were charectrized using scanning electron microscopy (SEM). After that they were exposed to gamma rays emitted from radioactive point sources [241Am, 133Ba, 137Cs, and 60Co],in order to determine some parameter as leaner attenuation coefficients, mass attenuation coefficients, and half value layer for HDPE composites in the wide photon energy ranges (59.53, 80.99, 121.78, 244.69, 344.28, 356.01, 661.66, 778.90, 964.13, 1173.25, 1332.50, and 1408.01) KeV, respectively Which cover low and intermediate energy ranges and this was done using a highly pure germanium (HPGe) cylindrical detector. The obtained results for the shielding properties were compared with that for pure HDPE(without fillers) to study the effect of micro-CdO and micro-PbO content on the radiation shielding properties of HDPE. The measurements showed that HDPE filled with such oxides has good shielding properties for low and medium energy gamma rays. In addition, the experimentally measured values were compared with that calculated theoretically by XCOM program and the comparison showed a remarkable agreement between them.

Introduction

Due to the continuous development in science and technology, especially in recent years, the use of radioactive sources has increased in various fields such as nuclear research centers, nuclear power plants, space research, medicine and agriculture. It was necessary to provide safe working conditions, meaning that biological radiation

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shields are necessary to provide adequate protection for workers at a reasonable cost[1].The radiation attenuation properties of a particular material are essential to determine how much protection is available and the amount of dose a person may receive if that material is used as radiation shielding [2]. It is noted that when gamma rays interact with matter, their intensity decreases as they pass through the matter. This decrease depends mainly on the type of target material in addition to the thickness of that matter [3]. The mass attenuation coefficient is an important factor that provides extensive information about photon penetration and energy deposition in biological shields. In this way, innovative alternative shield materials should be sought, preferably cheap and locally available, to meet the rapid expansion of the nuclear industry and the many applications of radioactive materials [4]. The material chosen should also be environmentally friendly, and it is sometimes preferable to use industrial waste, which has increased in production due to industrialization. Concrete remains the first practical choice for radiation protection for several reasons. The most important of these are its good mechanical properties and its ability to protect against radiation [5]. It has also been found that the polymer (as an alternative option) is characterized by reducing the weight of the protection material and increasing its flexibility [6].It is very important to determine the exact values of the interaction parameters - for the polymer as a shielding barrier - before using it in the fields of medicine, agriculture and imaging. etc [7], The mass attenuation coefficient is essentially the most important parameter for studying the interactions of gamma rays with matter. In addition to the half-value layer (HVL) and the effective atomic number, the other two are important for understanding the interaction with matter. [8],

Lead has been widely used since the past as a shield against gamma rays, but due to its high cost and heavy weight, different researchers have studied other materials for shielding against gamma rays by evaluating the interaction parameters with gamma radiation. [9]. Nowadays, and in recent years, many researches and studies have been appeared on measuring the linear and mass attenuation coefficients of related industrial materials and compounds such as building materials [10], concrete [11], cement [12], polymers, marble [13] and glass [14]. The aim of this research is to measure the gamma ray interaction parameters, namely the linear and mass attenuation coefficient, in addition to measuring the half value layer (HVL) of the high density polyethylene polymer HDPE compound filled with both lead and cadmium oxides in different substitution ratios (5, 10, 15, 20) wt% in order to study and evaluate the effectiveness of these materials as shielding against gamma rays. In this work, samples (for shielding purposes) were prepared from pure HDPE in addition to HDPE filled with lead and cadmium oxides in the filler weight ratios of (5, 10, 15, and 20) wt%.

3. Materials and methods:

3-1 Materials.

High-density polyethylene is a special type of polyethylene in that it is a monomer with a high density and a relatively high melting point. It is a thermoplastic material with a high strength ratio and represents more than 34% of the global plastics market, which is abbreviated as (HDPE). Chemically, HDPE is made up of a huge number of repeating units (known as monomers), and its chemical formula can be generalized as (C_2H_4) n. The amount of branching in HDPE is relatively low (when compared to other classes of polyethylene

Synthesis of Polymer-Matrix Composite Sheets:

Compression-molding technique was used to prepare the investigated micro sheets. Firstly, HDPE was weighted sensitively by an electrical balance (Analytical Balance, GR200, Japan) with an accuracy 0.0001g and then molten in a two roll mixer at 170oC, which is above the melting temperature of HDPE, for 15 min with the rotator speed set as 40 rpm. After complete melting of pure HDPE, the filler was slowly added with continuous blending for 20 min to ensure a uniformly mixed composite as shown in Figure (1). Fully mixed sample was then put into a stainless steel frame of dimensions (25×25×0.25 cm3) for hot-pressing between two layers of thermal Teflon to get a sheet with smooth surface. The pressing was done by using a hydraulic press

Table (1) The composite sample designations and the weight % of filler in each composite "detailed mass ratio of samples"

Sample Code	Pure HDPE (wt %)	CdO (wt %)	PbO (wt %)
HDPE	100		
2	90	5	5
3	80	10	10
4	70	15	15
5	60	20	20

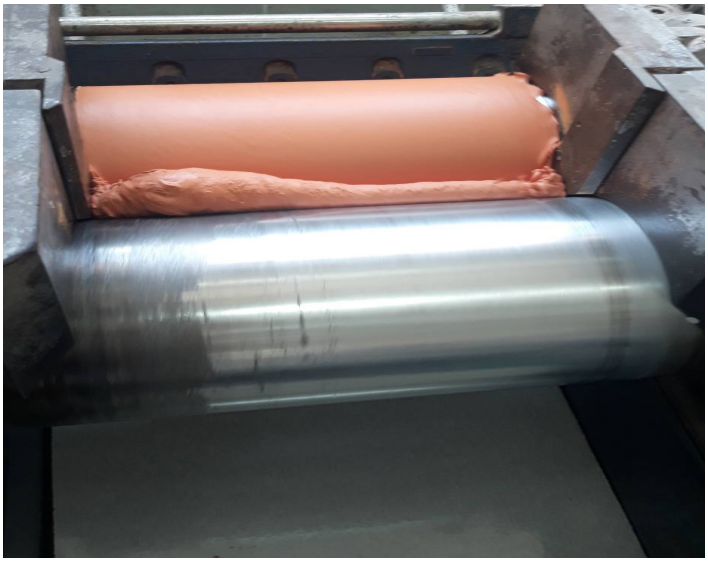


Figure (1). Uniformly mixed composite sample in a two roll mixer.

with an applied pressure 10 Mpa at170oC for 10 min. The pressure was then raised gradually up to 20 MPa for another 10 min. The sample was let in the press for 1 hour to cool down gradually by water at 20oC. Finally, the produced sheet was taken out from the mold and catted into circular samples of 8.4 cm in diameter and 0.25 cm in thickness to perform radiation-shielding tests. The composite sample designations and the weight % of filler in each composite are compiled in Table (1)

3.2. Scanning electron microscope (SEM):

In this study, scanning electron microscope (SEM) (JSM-6010LV, JEOL) was used to examine and observe the phase morphology of pure HDPE and the shape of filler dispersion with (5%, 10%, 15%, 20%) of CdO and PbO, all of which are shown in Fig. (3).

3.3. Experimental set-up:

In this study, the linear and mass attenuation coefficients of the prepared samples were measured. using a narrow beam of gamma rays was. The preparation of the experiment is depicted in Figure (2), where the samples were irradiant using sources [^{241}Am, ^{133}Ba, ^{137}Cs, and ^{60}Co].The source was enclosed in a lead container with one face aperture of 6 mm, placed behind the source collimator. Two collimators with apertures of 4 mm and 2.8 mm were placed with their front faces at a distance of 200mm and 550 mm from the source, respectively. The incident and transmitted gamma-ray intensities were determined for a fixed preset time using well calibrated Hyper Pure Germanium cylindrical detector (HPGe) from Canberra (Model GC1520) in conjunction with multichannel analyzer (MCA). To protect the detector from background radiation, the detector was contained in a cylindrical shield with a thickness of about 8 cm.

3-4 Theory:

The attenuation of gamma rays in a substance or composition of substances is a function of the following: (a) gamma energy (b) elemental composition of the attenuator substance (c) density and thickness of the attenuator substance. It is difficult to attenuate gamma rays because they have neither mass nor charge. The attenuation of gamma rays is a nonlinear function

of thickness (x) as expressed by Lambert's law [15],which state that, When the collimated beam of mono energetic gamma rays is attenuated in the matter, the transmitted intensity of
Gamma is,

$$I = I_o e^{-\mu x} \quad (1)$$

Where I is the transmitted density of gamma , I_o is the incidence intensity of gamma . X is the thickness and μ (cm $^{-1}$) is the linear attenuation coefficient of the substance. It is noted that (μ) is a function of the energy of gamma rays and depends on the density (ρ) of the substance and is expressed as a function of the mass which is known as a mass attenuation coefficient (μ_m) as follows:

$$\mu=\mu_m \rho \quad (2)$$

or μ/ρ is the mass attenuation coefficient denoted by μ_m.
The mass attenuation coefficient for a mixture of materials is:

$$(\mu/\rho)_{total} = \sum w_i (\mu/\rho)i \quad (2)$$

Where the total mass attenuation coefficient $(\mu/\rho)_{total}$ is the sum of
the mass attenuation coefficient of the individual components $(\mu/\rho)_i$ multiplied by the weight fraction w_i of component i. In the context of experimental work, the attenuation of gamma rays depends largely on the density of the sample material, meaning that more atoms and heavier atoms in the path of the photons, the more interactions per unit length of the sample. Therefore, the linear attenuation coefficient will have different values for the same material depending on its phase

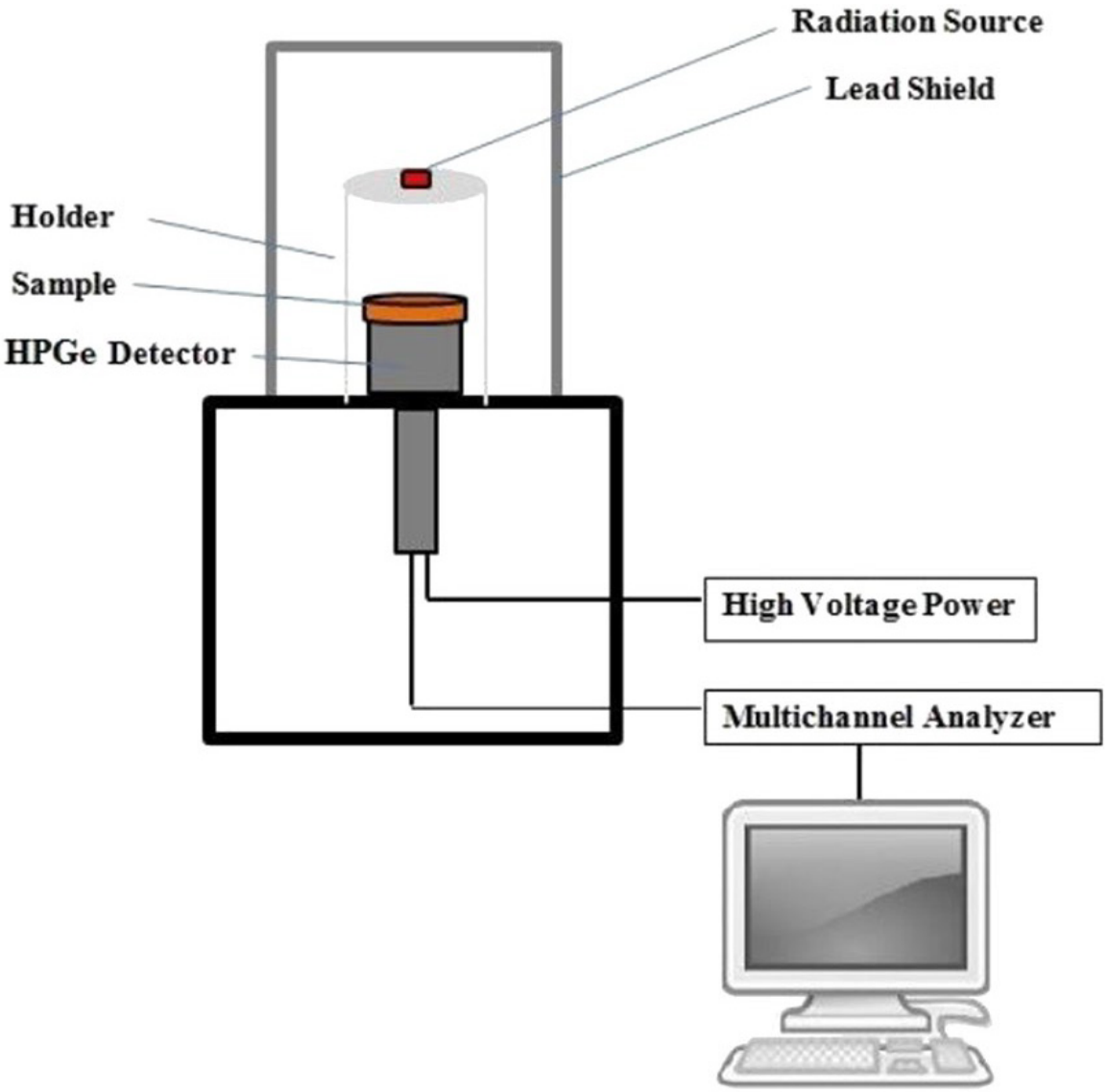


Fig. (2). The experimental setup for examining γ-ray shielding property.

(solid, liquid, or vapor) as well as its temperature [16]. For this reason, it is more convenient in practice to deal with the mass attenuation coefficient μ/ρ by dividing μ by the density ρ of the sample. Where the mass attenuation coefficient is, $\mu_m = (\mu/\rho)$

Finally, the half value layer (HVL), which is the thickness at which the transmitted intensity is one half the initial value, or The HVL (is the thickness of shielding material needed to reduce the incident intensity of the gamma ray to its half).It can be calculated from the linear attenuation coefficient like this ,

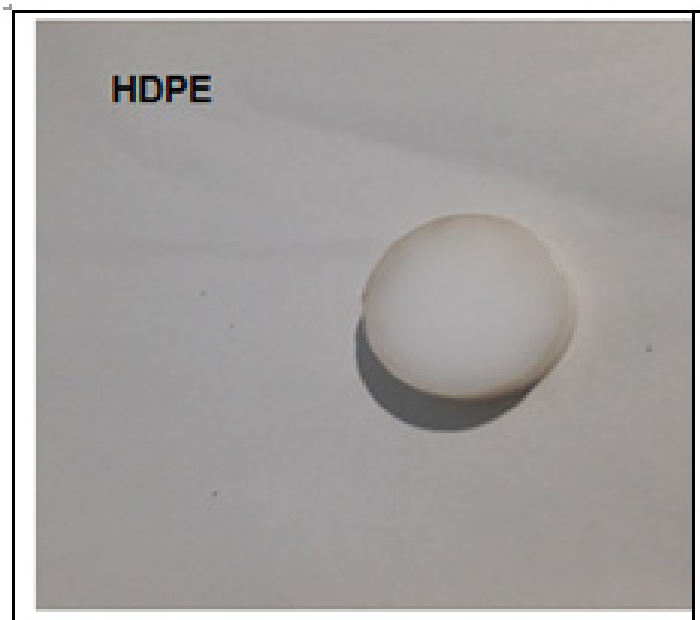
$$HVL = \frac{ln2 ln2}{\mu \quad \mu} \qquad (3)$$

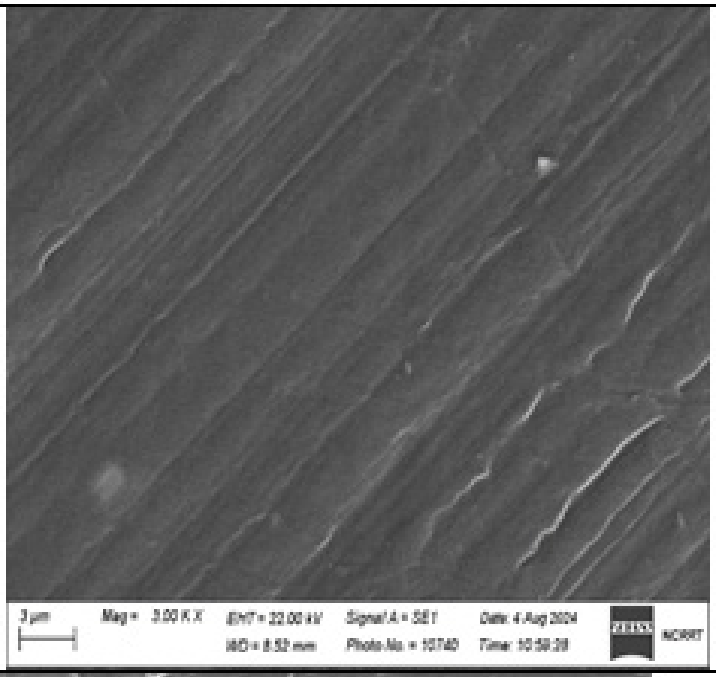
It should be noted that the interaction of gamma rays with matter occurs through three mechanisms: (1) the photoelectric effect (dominants < 50 KeV). (2). The Compton scattering(dominants 100 KeV - 10 MeV). (3) The pair-production which occurs for photons with energies higher than 1.02 MeV

4- Results& Discussion:

4-1 Scanning electron microscope (SEM).

Fig (3) depicts the electron microscope images of all samples (every sample with its SEM micrographs) with different Wight fractions (5%, 10%, 15 % and 20%) of CdO/HDPE and PbO/HDPE composites in addition to the HDPE pure which were used in the experiment, starting with pure high-density polyethylene, then polyethylene filled with both lead


HDPE


3 µm
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EHT = 22.00 kV
WD = 8.52 mm
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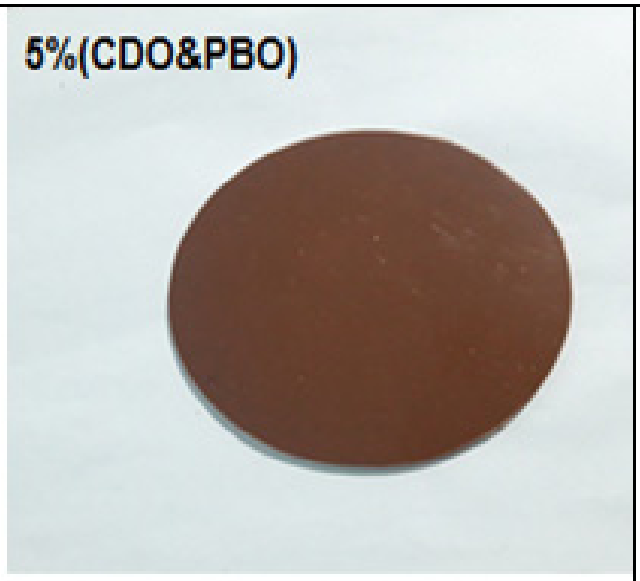
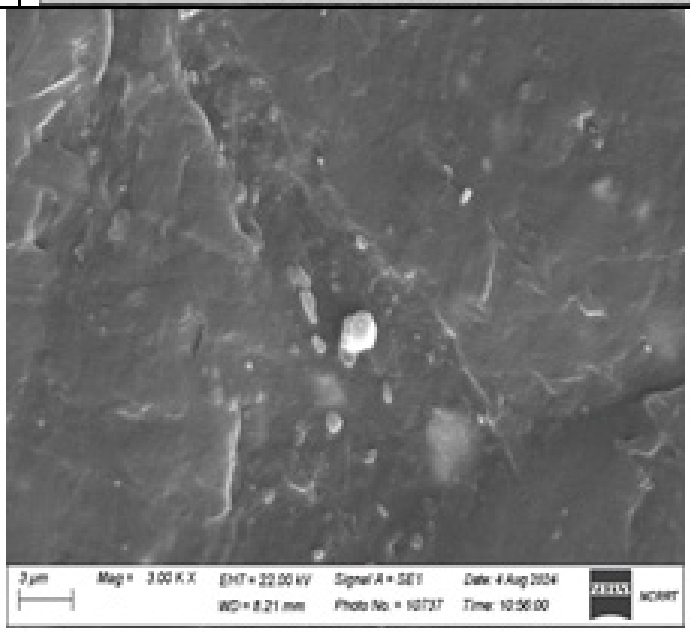
NORIT


5%(CDO&PBO)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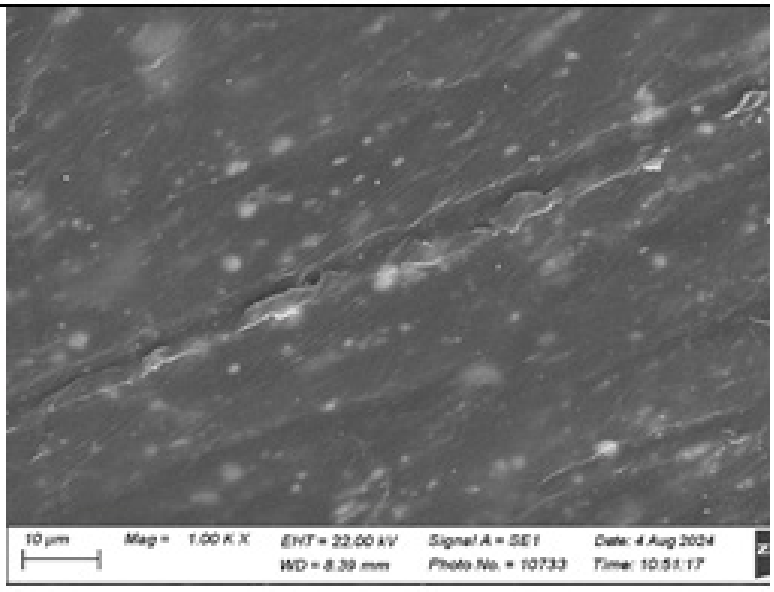
10%(CDO&PBO)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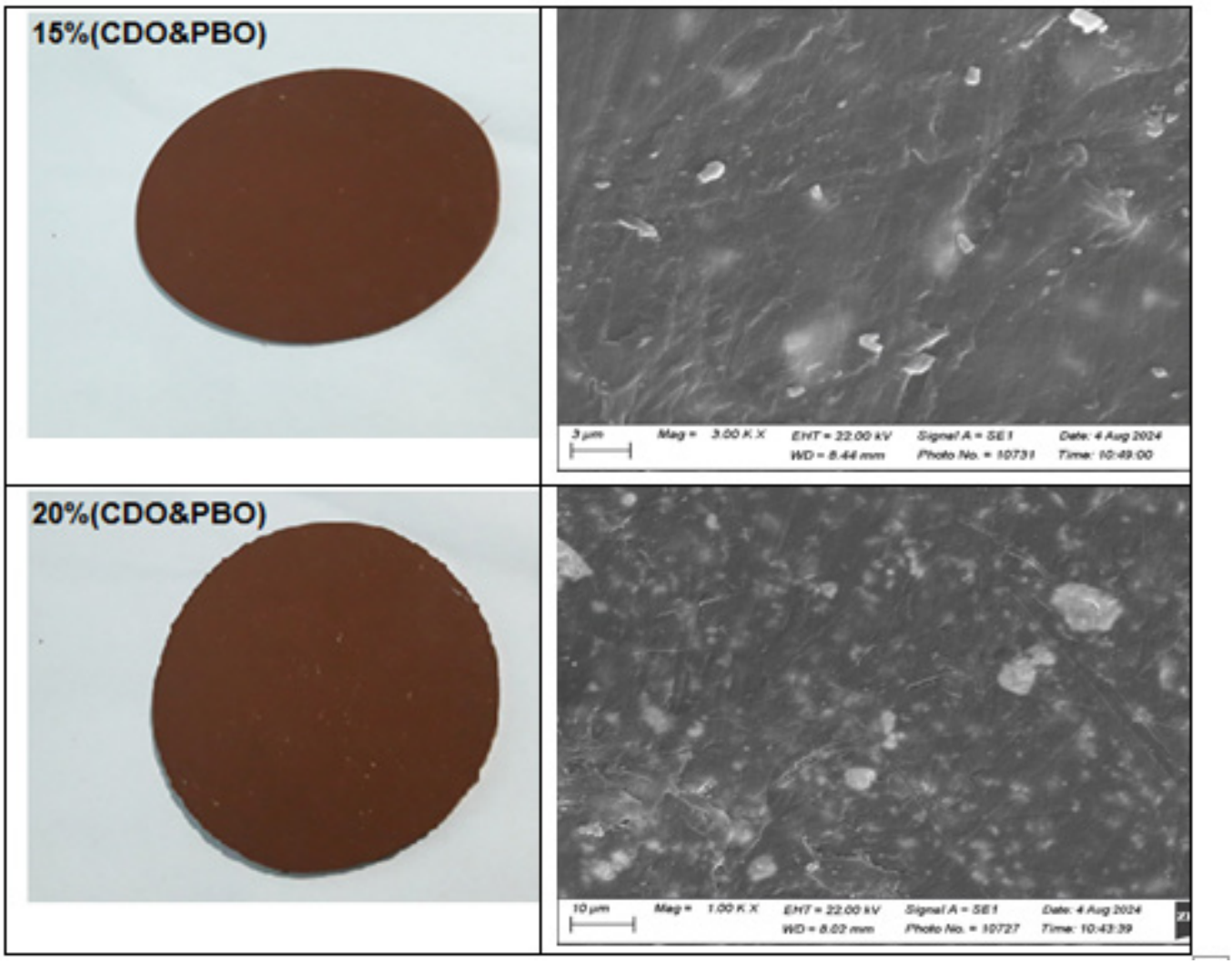


Fig (3) SEM images of Pure HDPE and HDPE/CDO and HDPE/PBO with different Wight fractions

Table(2). The variation of the parameters μ, μm and HVL either experimentally or theoretically as a function of γ energy for (micro CdO + micro PbO)/HDPE composites at different filler loadings.

Sample	Energy (keV)	Linear Attenuation Coefficient (cm^{-1})	Half Value layer (cm)	Density	Mass Attenuation Coefficient ($cm^2 g^{-1}$)		
					Measured	XCOM	Δ%
Pure HDPE	59.53	0.17834	3.887	0.944 ± 0.008	0.18892	0.18880	0.06%
	80.99	0.16774	4.132		0.17769	0.17690	0.45%
	121.78	0.15209	4.557		0.16111	0.16070	0.26%
	244.69	0.12313	5.629		0.13043	0.13040	0.03%
	344.28	0.10901	6.359		0.11548	0.11520	0.24%
	356.01	0.10777	6.432		0.11416	0.11380	0.32%
	661.66	0.08303	8.348		0.08796	0.08802	-0.07%
	778.90	0.07733	8.963		0.08192	0.08174	0.22%
	964.13	0.06972	9.942		0.07386	0.07387	-0.02%
	1173.23	0.06363	10.893		0.06740	0.06708	0.48%
	1332.50	0.05957	11.636		0.06310	0.06283	0.44%
	1408.01	0.05783	11.986		0.06126	0.06107	0.31%

Content of Table(2).

Sample	Energy (keV)	Linear Attenuation **Coefficient** (cm^{-1})	**Half Value layer (cm)**	**Density**	**Mass Attenuation Coefficient** ($cm^2\ g^{-1}$)		
					Measured	**XCOM**	**Δ%**
HDPE filled with 5 wt% Micro CdO + 5 wt% Micro PbO	59.53	0.67002	1.035	1.041 ± 0.006	0.64363	0.64360	0.00%
	80.99	0.38203	1.814		0.36698	0.36620	0.21%
	121.78	0.34704	1.997		0.33337	0.33340	-0.01%
	244.69	0.16046	4.320		0.15414	0.15400	0.09%
	344.28	0.12848	5.395		0.12342	0.12320	0.18%
	356.01	0.12594	5.504		0.12098	0.12090	0.07%
	661.66	0.09137	7.586		0.08777	0.08796	-0.21%
	778.90	0.08466	8.187		0.08133	0.08116	0.20%
	964.13	0.07609	9.110		0.07309	0.07293	0.22%
	1173.23	0.06882	10.072		0.06611	0.06600	0.17%
	1332.50	0.06445	10.755		0.06191	0.06175	0.26%
	1408.01	0.06243	11.103		0.05997	0.06001	-0.06%
HDPE filled with 10 wt% Micro CdO + 10 wt% Micro PbO	59.53	1.26294	0.549	1.148 ± 0.009	1.10012	1.09800	0.19%
	80.99	0.63713	1.088		0.55499	0.55560	-0.11%
	121.78	0.58234	1.190		0.50726	0.50610	0.23%
	244.69	0.20363	3.404		0.17738	0.17760	-0.12%
	344.28	0.15126	4.582		0.13176	0.13120	0.43%
	356.01	0.14665	4.727		0.12774	0.12800	-0.20%
	661.66	0.10114	6.853		0.08810	0.08790	0.23%
	778.90	0.09252	7.492		0.08059	0.08059	0.00%
	964.13	0.08243	8.409		0.07180	0.07199	-0.26%
	1173.23	0.07464	9.287		0.06502	0.06492	0.15%
	1332.50	0.06982	9.928		0.06082	0.06068	0.23%
	1408.01	0.06789	10.210		0.05914	0.05896	0.30%
HDPE filled with 15 wt% Micro CdO + 15 wt% Micro PbO	59.53	2.00860	0.345	1.259 ± 0.004	1.55104	1.55300	-0.13%
	80.99	0.96821	0.716		0.74765	0.74490	0.37%
	121.78	0.87830	0.789		0.67822	0.67880	-0.08%
	244.69	0.26179	2.648		0.20215	0.20120	0.47%
	344.28	0.17992	3.853		0.13893	0.13920	-0.19%
	356.01	0.17546	3.950		0.13549	0.13510	0.29%
	661.66	0.11380	6.091		0.08788	0.08785	0.03%
	778.90	0.10363	6.689		0.08002	0.08001	0.02%
	964.13	0.09226	7.513		0.07124	0.07105	0.27%
	1173.23	0.08272	8.379		0.06388	0.06384	0.06%
	1332.50	0.07706	8.995		0.05951	0.05960	-0.16%
	1408.01	0.07511	9.228		0.05800	0.05791	0.16%

Content of Table(2)

	59.53	2.93492	0.236		2.00747	2.00800	-0.03%
	80.99	1.36750	0.507		0.93536	0.93430	0.11%
	121.78	1.24372	0.557		0.85070	0.85140	-0.08%
	244.69	0.32945	2.104		0.22534	0.22480	0.24%
HDPE filled with 20 wt% Micro CdO + 20 wt% Micro PbO	344.28	0.21595	3.210	1.462 ± 0.005	0.14771	0.14720	0.35%
	356.01	0.20871	3.321		0.14276	0.14230	0.32%
	661.66	0.12861	5.390		0.08797	0.08779	0.20%
	778.90	0.11637	5.956		0.07960	0.07944	0.20%
	964.13	0.10231	6.775		0.06998	0.07011	-0.19%
	1173.23	0.09172	7.557		0.06274	0.06276	-0.04%
	1332.50	0.08582	8.077		0.05870	0.05853	0.29%
	1408.01	0.08318	8.333		0.05689	0.05685	0.08%
1408.01	0.07511	9.228		0.05800	0.05791	0.16%	

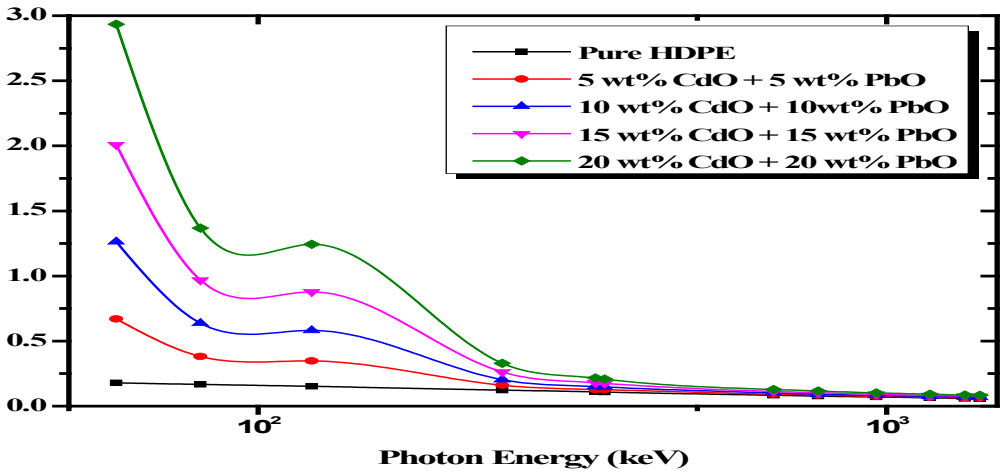


Figure (4) The linear-attenuation μ (cm-1) for different wt% of the fillers

linear-attenuation coefficient (μ, cm^{-1}) as well as the mass-attenuation coefficient (cm^2g^{-1}) of the absorber (sample targets) for gamma-ray of certain energy was evaluated by the Eq. (1).

$$\mu = (1/x) \ln (I/I_o)$$

And $\mu_m = 1/(\rho x) \ln (I/I_o)$

Where x is the thickness of the sample, I and I_o are the number of counts recorded in the detector with and without the sample target, respectively. Table (2) lists out the values of measured density (ρ), linear-attenuation coefficient (μ, cm^{-1}), mass-attenuation coefficient (μ_m, cm^2g^{-1}) and theoretical values of (μ_m, cm^2g^{-1}) evaluated from XCOM program, in addition to the discrepancy Δ% (between the measured and theoretical values of μ_m) for HDPE pure, and CdO/ HDPE, PbO/ HDPE composites at the energies (59.53, 80.99, 121.78, 244.69, 344.28, 356.01, 661.66, 778.90, 964.13, 1173.25, 1332.50, and 1408.01) KeV, of the incident photon. From the listed results in Table (2), it is evident that, the ρ value

of the composites increases dramatically with increasing the wt% of the fillers (PbO &CdO) because of the dispersion of the high density and high Z number filler material within the low density HDPE matrix. Consequently, incorporating PbO and CdO into HDPE leads to an increase in its packing density. Based on the listed data in Table (2), and also from the graph shown in the figure (4).it is evident that the measured values of μ increase with increasing the PbO & CdO content in the polymeric composite at all gamma-ray energies, and this trend is mainly assigned to the high absorbability of PbO & CdO to gamma-ray radiation [17], In addition, μ for various photon interaction processes at the start is high and then decreases sharply with increases the photon energy up to 100 keV for all investigated composite materials due to the dominance of three main processes of incident photon energies.

oxide and cadmium oxide with different weight fractions. We notice that the micro particles of both lead oxide and cadmium oxide take the form of flakes and are distributed throughout the polyethylene matrix and have an average size in the range between 0.6 to 0.9 μm. It is also noted that they appear more densely as we move to higher concentrations of the two oxides.

4-2 Measurement of linear and mass-attenuation coefficient for γ-ray with different energies:

In the course of experiment, the gamma-ray attenuation was measured with and without polymer target. The l the The obtained results of mass attenuation confident μ_m are listed in Table (2), and also from the graph-in three dimensions- shown in the figure (5) It is evident that, μ_m increase with the increase in filler weight fraction at all gamma-ray energies. This may be attributed to increasing lead and cadmium contents in the polymeric composite and the filler dispensability in the polymer matrix as well. These observations indicate that PbO &CdO filled composites perform is better at all the gamma-ray energies than HDPE itself. Moreover, μ_m is a function of the photon energy as given in Table (2) and evident in figure (5). With an increase in energy (from 59.53 up to 1408.01keV), μ_m decreases for each weight fraction of filler in the composite. The interaction cross-sections decrease with the increase in the gamma-ray energy. The cross-sections for photoelectric-interactions are adequately high at energies lower than 50 KeV in most

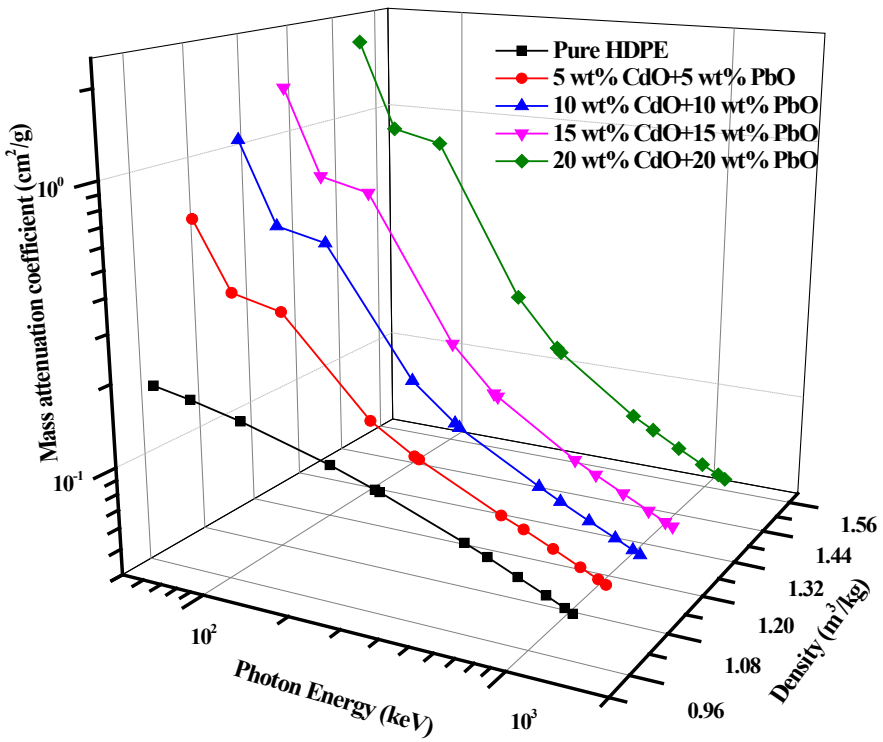
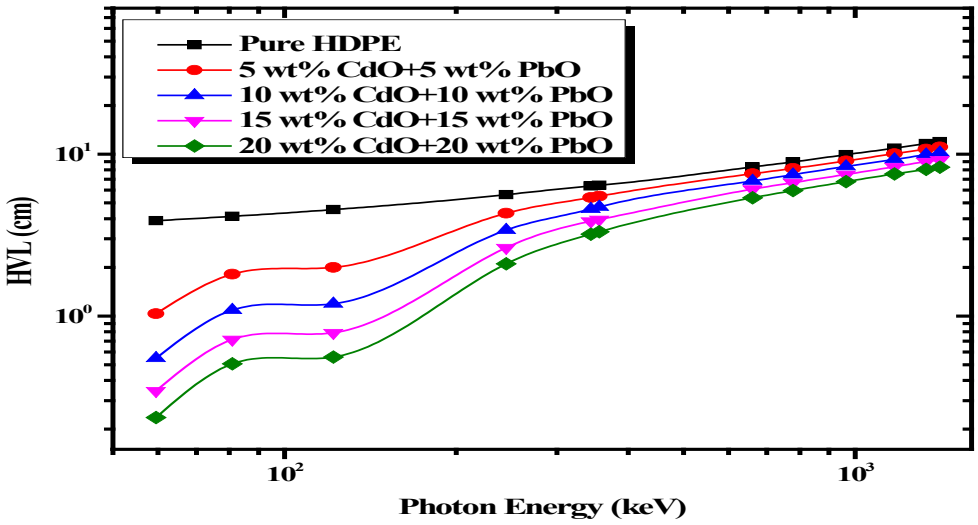


Figure (5) Mass attenuation as a function of photon energy and target density.



Figure(6).Half value layer (HVL),as a function of photon energy

absorbing materials. Between 100 KeV to 10 MeV, cross-sections for Compton-scattering are significant and above 2 MeV pair-production process becomes dominant [18]. 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 it is not the case in Compton-scattering and pair-production, where photons are not completely absorbed [19]. And since the mass attenuation coefficient play the largest role in designing radiation shields against the gamma rays, so we must notice the influence of these parameter with both photon energy and the increase in the perentage of filling material or in other words the density of the sample material, and this is evident in translation of data in the table to the three -dimensional graphic figure (5)

It is also noted from the magnitude of Δ% that, the measured mass-attenuation coefficients are in close agreement with the database of the reference mass -attenuation coefficients using XCOM program.

Half value layer (HVL), is an important parameter in the study of radiation shielding. It is clear from the table and from the figure (6) the extent of its influence by the photon energy as well as the filler contents.

4. Conclusion

The polymer-matrix composites based on HDPE with PbO& CdO using various weight fractions were successfully fabricated for use in the gamma-ray shielding applications. PbO and CdO were synthesized by simple, economic and productive techniques. The attenuation coefficients were identified to increase with the increase in the contents of PbO and CdO. The photoelectric absorption and Compton-scattering were identified as the main absorption processes. A comparative study was performed to determine the linear and mass attenuation coefficients for HDPE,and PbO / HDPE , CdO / HDPE. The results revealed that improvement was attained using all investigated gamma-ray energies. The PbO / HDPE and CdO/HDPE were characterized as promising alternative candidates for applications in the gamma-ray shielding over a wide range of energies due to ease of processing, good dispersion and flexibility. The outlined tends could be used for both stationary applications as radioactive source shielding and also mobile applications. Comfortable clothing shield materials could be produced for radiation workers by using these materials.

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