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NEUTRON ATTENUATION OF HDPE WITH CADMIUM AND LEAD OXIDES FOR RADIATION SHIELDING APPLICATION

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Abstract: The current study focused on developing composites of some materials to obtain high attenuation performance for protection against neutron and gamma radiation while maintaining strong mechanical properties. These composites based on high-density polyethylene (HDPE), with different concentrations of fillers PbO, and CdO of [5%, 10%, 15% and 20%] have been prepared. The physical and mechanical behavior of the composites was studied, in addition to study of neutron and gamma attenuation, as shielding which is the main objective of this research. The shielding properties were determined by measuring the fast neutrons and total gamma-ray attenuation coefficients respectively by $^{239}\text{Pu}-\alpha-^9\text{Be}$ neutron source. The neutron fluence rates and gamma fluxes were measured using a stilbene organic scintillator. The macroscopic effective removal cross-section Σ_R (cm^{-1}) of fast neutrons and total attenuation coefficient μ (cm^{-1}) of gamma rays has been studied experimentally. The half-value layer (HVL) and the relaxation length (λ) have been studied also. All this studies indicate that, HDPE containing PbO, and CdO as a fillers leads to having high-performance radiation shielding materials.

Keywords: Neutron attenuation, Shielding, HDPE, CdO, PbO

1. INTRODUCTION

In numerous industries, including nuclear power, radiotherapy, imaging technologies, and others, neutron sources are widely used. It served as an activation analyser in the research actions as well [1]. Because neutrons are uncharged particles, they interact to cause harm to people and the environment. There is a growing need for neutron-shielding materials. The cross-section and the energy deposited in the medium may be affecting the choice of neutron shielding material [2]. Nuclear radiation is beneficial when used properly for many purposes such as business, science, and medicine, but when not used properly, it can be dangerous to people and the environment. To keep radiation workers and environments safe, protection from undesired radiation is essential. In particular, shielding materials for ionizing radiation have been investigated. These radiations include high-energy neutrons produced by nuclear reactors and connected equipment/ facilities [3,4], fusion reactors [5], cyclotrons [6], and space radiation [7]. As well as high energy gamma radiation.

High-energy neutrons generated by neutron sources can be slowed down and absorbed in a hydrogenous medium (for slowing down) and certain absorber elements (for absorption), such as boron (B), lithium (Li), gadolinium (Gd), and others, for example, boron compounds are often inexpensive and have strong thermal neutron absorption capabilities, polymer composites containing boron are typically employed for this purpose [8]. A material with a high hydrogen concentration is a suitable option for fast neutron shielding, while boron and boron-based compounds are very effective for thermal neutron shielding. High-density polyethylene (HDPE) doped with boron (as additive) can be used as thermal neutron shielding materials. In recent studies, one of the important studies on neutron shielding is that high-density polyethylene (HDPE) was mixed with boron nitride in the form of modified boron nitride (mBN) filters and compared with other compounds in the form of BN and boron carbide (B_4C) fillers. The efficacy of HDPE/BN, HDPE/mBN, and HDPE/ B_4C compounds, the majority of which contain fillers, was demonstrated. [9]. Boron carbide was also combined with thick polyethylene and particle sizes ranging from micro

to nano, using varied weight ratios as thermal neutron shielding, [10]. In summary, boron and its derivatives have recently been employed successfully in radiation protection studies [11].

In addition, a laboratory investigation was done using a mixture of concrete and polymer (PISCC) as neutron shields, in which high-density polyethylene was crushed with three different polyethylene diameters [12,13]. Although radiation shielding investigations of polymers are critical in estimating their effect in nuclear protection applications, the number of experiments with data based on the utilization of mixed radiation fields emitted from neutron sources is limited due to complex facilities and protection requirements. To boost the hydrogen content of standard concrete mixtures, replace sand with this ingredient. It was discovered that this new form of concrete significantly improved neutron shielding qualities.[14]. Polymer Nano composites have recently been used to create shielding materials for very low-energy X-rays [15–17], However, no attempts have been made to use them to absorb thermal neutrons. However, thanks to their improved mechanical, thermal, and electrical properties, nanoparticles have been used in numerous industries over the past few decades, including electronics, pharmaceuticals, cosmetics, and biomedicine. [18]. Due to the extremely large cumulative surface area of nanofillers and the elimination of particle absorption by self-shielding, it is suggested that they increase the probability of thermal neutron interaction, So polymer Nano composites may also improve the thermal neutron absorption efficiency. [19]. Due to the high surface energy of the nanoparticles, which tends to cause them to aggregate and increase the viscosity of the polymer matrix, uniform dispersion of the nanoparticles in a polymer matrix is not easily achievable [20]. This makes the preparation of that nanomaterial very challenging.

Most of the high-density polyethylene (HDPE) used in industry contains a saturated polyethylene polymer chain. All of them are used in wires, insulation materials, waterproof coatings, automotive uses, and nuclear applications in everyday life [21-23]. The HDPE high hydrogen content also contributes to its excellent neutron shielding capacity [24]. Additionally, a study by a research team has demonstrated that CdO/ HDPE can be used as a part of a composite for gamma-ray shielding material [25]. According to a different study, HDPE can be used as a matrix for embedding radioactive waste [26]. The high hydrogen concentration of HDPE makes it a natural choice for neutron shielding. Hydrogen has a medium thermal cross-section for neutrons, while boron, a typical thermal neutron shielding material, has a larger cross-section. [27]. However, compared to other commercial rubbers, high-density polyethylene (HDPE), which has a high hydrogen content, is relatively inexpensive. Several researchers have used rubbers and plastics functionalized with maleic anhydride as a coupling agent to enhance mechanical and physical behaviour. [28,29].

This work aims to evaluate the potential of high-density polyethylene (HDPE) samples as neutron shielding materials when combined with lead oxide (PbO) and cadmium oxide (CdO) as fillers. This is done by examining the mechanical and physical properties firstly, before studying the attenuation properties of these samples for shielding against radiation.

2. MATERIAL AND METHODS

Samples preparation:

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). When preparing samples, A 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 molted in a two roll mixer at 170 °C, (which is above the melting temperature of

HDPE), for 15 min with the mixer rotor speed set at 40 rpm.. After complete melting of pure HDPE, the filler – of PbO & CdO – were slowly added with continuous blending for 20 min to ensure a uniformly mixed composite. 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 with an applied pressure 10 MPa at 170°C 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 20°C. 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.

Table 1. The composite sample designations and the weight % of filler in each composite.

Sample code	HDPE (Wt %)	PbO (Wt %)	CdO (Wt %)
HDPE	100	-----	-----
2	90	5	5
3	80	10	10
4	70	15	15
5	60	20	20

■ Mechanical Testing:

Mechanical properties include tensile strength and elongation, which are important in shielding design for neutrons and gamma radiations, were conducted in the mechanical workshops of Radiation physics labs, at the Atomic Energy Authority.

3. MEASUREMENTS OF RADIATION

In this experimental work, a neutron source used is (²³⁹Pu–α–⁹Be) which is characterized by higher neutron floucnce, a compressed design, and a sensible photon contribution in the radiation scope. This source has a radioactivity of (5 Ci). The neutron and gamma spectra are measured using Stilbene scintillation detectors to evaluate the fast neutrons and total gamma spectrum transmitted through the samples of interest.

The source was placed in a lead collimator with an opening diameter of 10 mm to produce a narrow beam appropriate for measurement processes. Furthermore, the detector was placed 30 mm away from the radiation source to achieve an appropriate count rate and the samples were set in front of the detector with different thicknesses (9–45) mm as depicted in Figure 1.

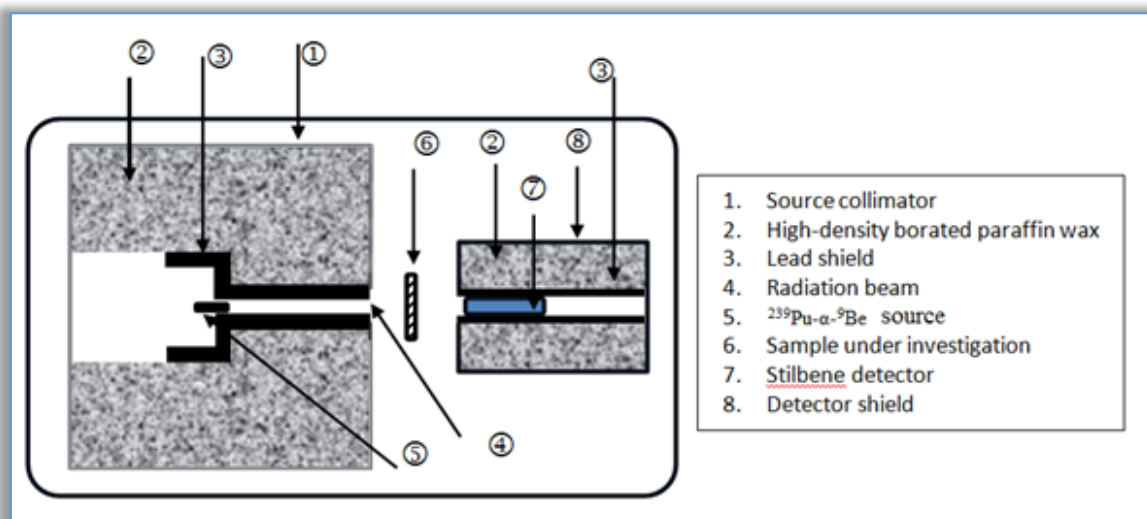


Figure.1. Schematic diagram of the experimental set up of neutron measurement.

Pulse shape differentiation (PSD) was conducted via the anticoincidence mode with a zero cross–over technique. It was essential for discriminating between neutron and gamma pulsations by treating the unbidden electron and proton pulses because of gamma–ray and neutron reactions in the scintillator, respectively [30,31]. The energy spectra of fast neutrons and gamma rays were

converted by measuring the pulse amplitude distributions of recoil protons or electrons using the two unfolding codes NSPEC and GSPEC based on double differentiation and matrix correction methods, respectively [32]. The experimental measurements have been carried out at Labs. For Developing Nuclear Techniques to Detect Landmines and Illicit Materials, Nuclear Research Center, Egyptian Atomic Energy Authority (EAEA). The spectrum of the neutron source was integrated to obtain the incident intensity (I_0) and transmitted intensity (I) for fast neutrons and total gamma-rays as a function of energy as well as thickness; according to Beer-Lambert's law (Eqs 1,2 for neutron and gamma respectively). Add to that, the corresponding shielding parameters of the fast neutrons and gamma rays, which includes both: half-value layer (HVL, cm) and relaxation length (λ , cm) were determined using Eqs (3,4) [33].

$$I = I_0 e^{-\Sigma x} \quad (1)$$

(For neutron), where Σ is macroscopic cross section (For neutron ray)

$$I = I_0 e^{-\mu x} \quad (2)$$

Where μ is attenuation coefficient (For gamma ray)

$$HVL = \frac{\ln 2}{\mu} \quad (3)$$

Where the half value layer (HVL) is defined as the thickness of material required to reduce the intensity of incoming radiation in half value

$$\lambda = \frac{1}{\mu} \text{ (for gamma) , } \lambda = 1/\Sigma \text{ (for neutron)} \quad (4)$$

The relaxation length (λ) is defined as the mean of the gap between two subsequent photon (neutron) interactions.

4. RESULTS AND DISCUSSION

This discussion covers the three important topics:

- Mechanical properties
- Neutron radiation attenuation properties
- Gamma ray attenuation properties, as follow

Mechanical properties of composites

The mechanical properties of polymers are of great importance when they are changed by adding the homogeneous distribution of inorganic fillers.[34, 35]. Figure (2) shows the effect of lead oxide (PbO) and cadmium oxide (CdO) ratios on the tensile strength and elongation at break of filler-HDPE composites.

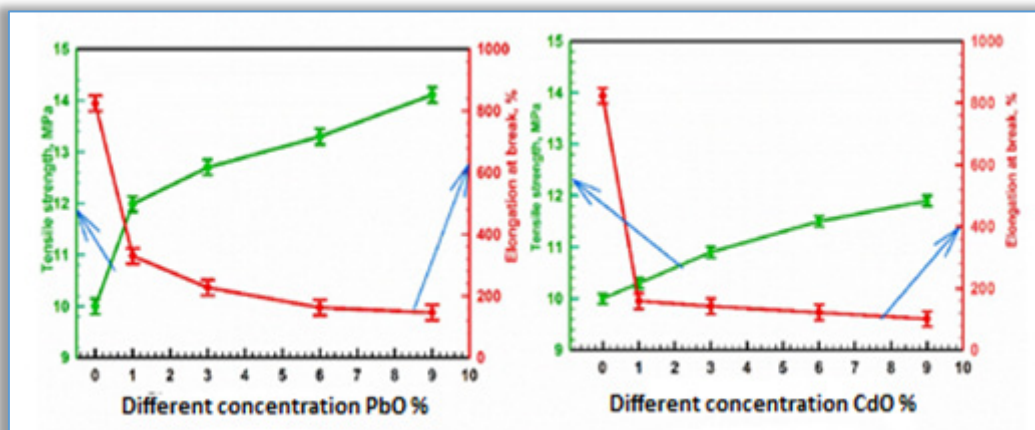


Figure 2. Variation of the mechanical properties of HDPE matrix containing different ratios of (PbO, and CdO %)

It is observed that the tensile strength of the prepared filler-HDPE composite gradually increased with increasing filler concentration, reaching a maximum concentration of 10% of PbO and cadmium oxide,(also up to 20%) It was also found that the 4.5%: 6% increase in tensile strength is attributed to the uniform distribution of fillers in HDPE composites, which improve the interfacial

interaction between them in the presence of the coupling agent saline. PbO and CdO are metal oxides that can act as activators and contribute to chemical crosslinking, resulting in higher crosslink density and tensile strength values [36, 37]. In this regard, the tensile strength values reached 11.9, 13.41, and 4.11 MPa at (5,10, up to 20) % concentrations of lead oxide (PbO) and cadmium oxide (CdO) as fillers in the high-density polyethylene (HDPE). Also, it was observed that the PbO/HDPE composites recorded the highest tensile strength (TS) values compared to the other materials, which confirms the presence of lead oxide improve the tensile properties of the PbO/HDPE composites. [38]. Furthermore, we find on the contrary the elongation at break (%) decreased as the loading of PbO, and CdO particles in the host HDPE matrix increased, which could be attributed to the loss in elasticity in HDPE as filler loadings increased.

■ Neutron- attenuation measurements:

Figure (3) displays a relation between the fast neutron fluxes ($\text{n}/\text{cm}^2\cdot\text{s}$) and their energy distributions (0.8–11 MeV) evaluated through 9, 18, 27, 36 and 45 mm thicknesses of the investigated samples.

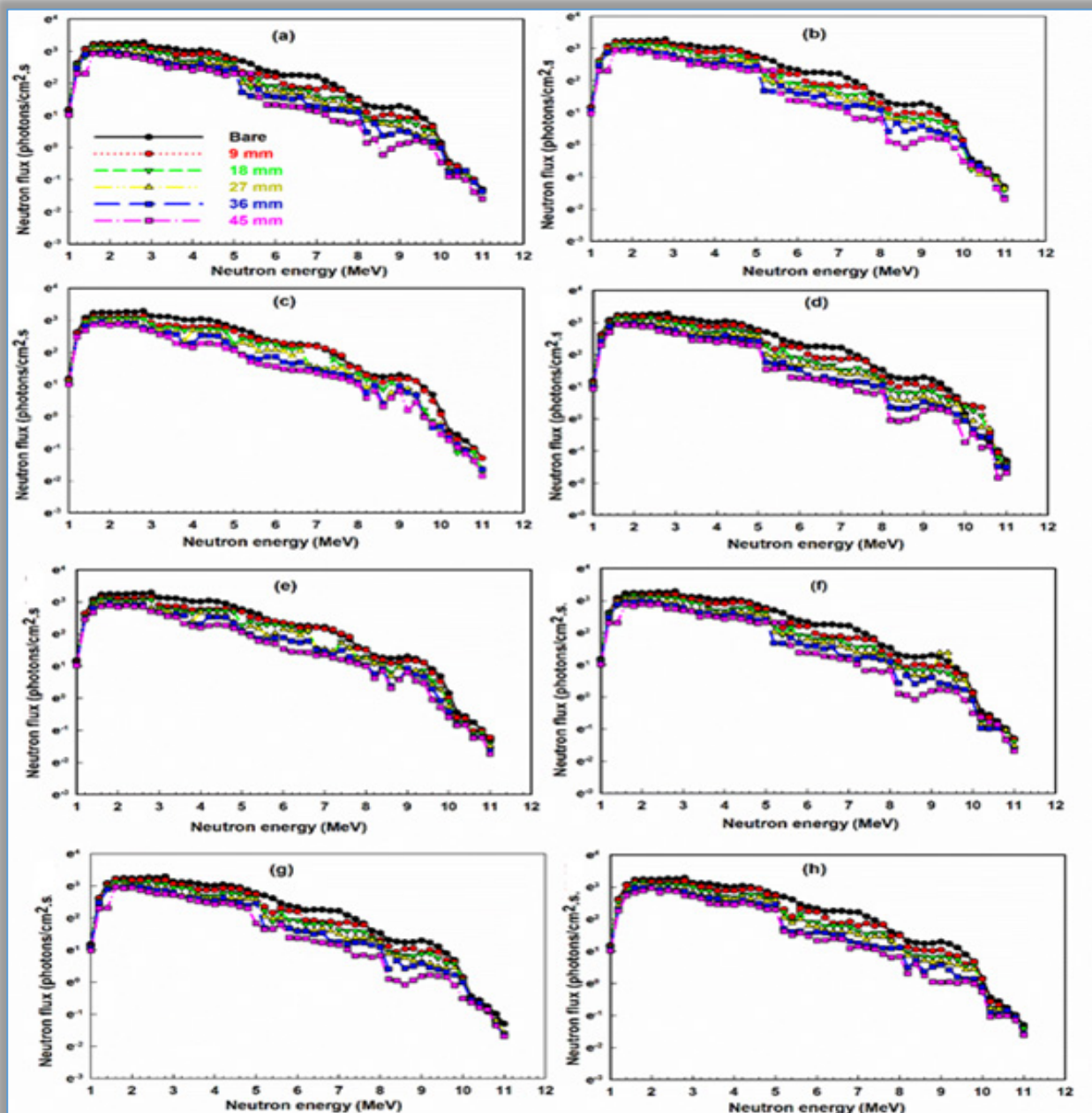


Figure 3. Measured fast neutron spectra behind different thickness of unfilled HDPE (a), HDPE matrix filled with 4 different ratios of PbO/ HDPE structure (b, c, d & e), and also different ratios of CdO/ HDPE structure (f, g & h).

The formats of fast neutron spectra for all studied thicknesses are nearly the same and possess similar profiles such as the bare neutron spectrum in fig (3 a). Note in the following Figures (3, b, c, d & e), it is clear that the greater the sample thickness, the less the fast neutron fluxes gradually increase for all neutron energies. At a low energy range of ~1.5–2.5 MeV, there is an increase in the flux intensity of fast neutrons at all spectra for all investigated samples. These high neutron fluxes proceed from the fission of plutonium and the (n, 2n) reaction with Be of the used (Pu- α -Be) source in the measurements [39, 40]. At low energies of 3–4 MeV, there is more distinctness with increasing the thickness of samples.

A high flux of fast neutrons which is still observed in the energy range of 4–5 MeV, indicating a small effective removal cross-section (Note that the removal cross section refers to the parameter that represents the amount of attenuation of the neutron flux and is symbolized by Σ). On the other hand, it is noted that the highest effective cross-section for removal is in the energy region. From 5 to 11 MeV, here it is noted that the low flux may be due to the reaction of $^9\text{Be}(\alpha, n)^{12}\text{C}$ to produce high-energy alpha particles. [41–43]. Therefore, these materials can contribute to enhancing the attenuation of fast neutrons, which is currently represented by high-density polyethylene (HDPE) [44, 45]. In addition, it has been found that the addition of 10% PbO/HDPE (as well as CdO/HDPE) represents the best additive to enhance fast neutron attenuation.

Finally, values and data shown in the table (2) which were calculated according to equations (1,2,3,4) showed that the value of the removal cross section Σ appears to be somewhat higher for CdO/HDPE composites compared to the other samples. In addition, the other shielding parameters (HVL and λ) are shown in the same table which indicates the change in the values of these parameters as a result of the difference in the concentration of additive fillers, which in turn show an improvement in the attenuation of fast neutrons.

Table 2. Measured Σ , HVL and λ for the investigated samples

No	sample	Σ (cm ⁻¹)	HVL (cm)	λ (cm)
1.	HDPE	0.131	5.291	7.634
2.	HDPE-5% PbO	0.132	5.251	7.576
3.	HDPE-10% PbO	0.133	5.212	7.519
4.	HDPE-15% PbO	0.134	5.173	7.463
5.	HDPE-20% PbO	0.136	5.097	7.353
6.	HDPE-5% CdO	0.133	5.212	7.519
7.	HDPE-10% CdO	0.135	5.134	7.407
8.	HDPE-15% CdO	0.136	5.097	7.353
9.	HDPE-20% CdO	0.138	5.023	7.246

■ Gamma – attenuation measurements:

Figure 4 shows the intensities of the total gamma flux (photons/cm².s) measured through the 9, 18, 27, 36 and 45 mm thicknesses of the investigated samples for total gamma-ray energies.

The total gamma spectra are almost the same and have a similar profile to the bare gamma spectrum in fig (4, a). Note in the following Figures (4, b, c, d & e), it is obvious that the total gamma-ray flux intensities decrease with increasing the thickness of the investigated samples. Although the total gamma-ray flux intensities were observed to decrease with increasing thickness of the studied samples, It is noted that there are peaks observed in the gamma spectra of the entire studied sample produced in different shapes at different energies. This may be attributed to one or more of the following three causes: first, isotopic impurities in the radiation source, second, excitation of lead- ^{208}Pb from the colliders to its ground state, and third, the background shape of the chamber, ^{208}Tl . [46–48]. Additionally, the total gamma ray attenuation parameters (μ , HVL, and λ) for all studied samples indicate that, the higher the values of additives fillers as well as thicknesses, the higher the values of attenuation coefficients (μ) and the lower the values of HVL and λ , This in turn confirms an improvement in the radiation attenuation properties of the samples under study, all of this is shown in Table 3.

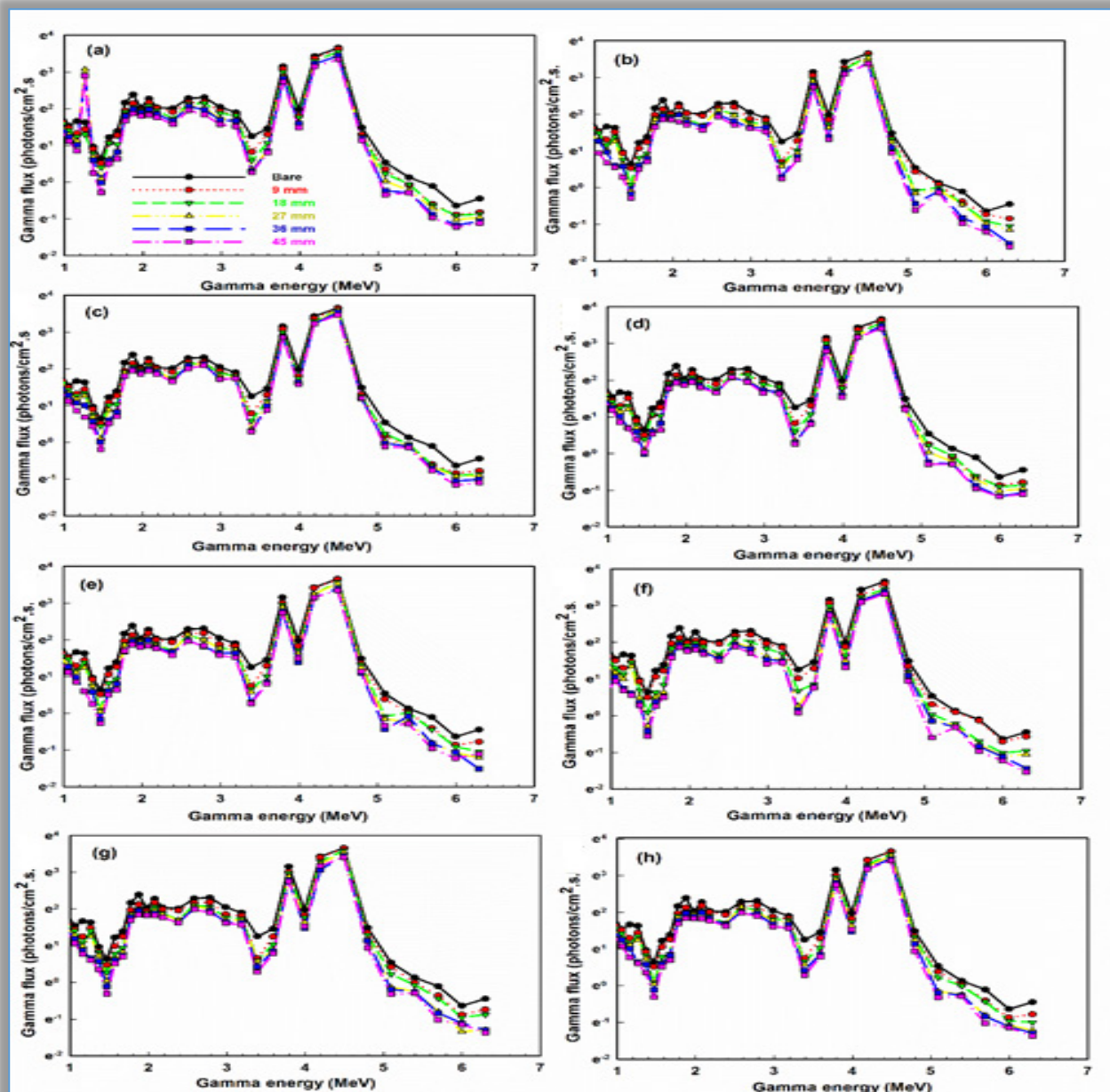


Figure 4. Distribution of total gamma spectra behind different thickness of unfilled HDPE (a), HDPE matrix with 4 different ratios of PbO/ HDPE structure (b, c, d & e), and also different ratios of CdO/ HDPE structure (f, g & h).

Table 3. The measured value of μ , HVL and λ for total gamma rays.

No	Sample	μ (cm ⁻¹)	HVL (cm)	λ (cm)
1.	HDPE	0.082	8.453	12.195
2.	HDPE-5% PbO	0.103	6.729	9.709
3.	HDPE-10% PbO	0.110	6.301	9.090
4.	HDPE-15% PbO	0.115	6.027	8.696
5.	HDPE-20% PbO	0.119	5.825	8.403
6.	HDPE-5% CdO	0.097	7.146	10.309
7.	HDPE-10% CdO	0.100	6.931	10.00
8.	HDPE-15% CdO	0.109	6.359	9.174
9.	HDPE-20% CdO	0.112	6.189	8.929

5. CONCLUSION

The aim of the current work focused on studying the modifications in the mechanical properties and attenuation properties of neutron radiation as well as gamma radiations of the developed composites based on high-density polyethylene with different concentrations of additives of both lead oxide and cadmium oxide as fillers, this is to be used as shields for protection against this

radiation. It was observed experimentally that the incorporation of lead oxide and cadmium oxide led to a gradual increase in the tensile strength values of high-density polyethylene. Moreover, the elongation at break decreased and the hardness increased significantly with increasing loading of lead oxide and cadmium oxide particles in the high-density polyethylene samples. But to study these composites as shields against neutron and gamma radiation, this was done experimentally by exposing them to a neutron source ($^{239}\text{Pu}-\alpha-^9\text{Be}$). It was found that the increase in the additional concentration of both lead oxide and cadmium oxide gives a significant improvement in the attenuation coefficients. For both neutron radiation as well as gamma radiation, and thus a clear improvement appeared in the values of other shielding parameters (HVL as well as λ). Therefore, the obtained results showed that HDPE composites filled with PbO and CdO act as effective neutron shielding materials.

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