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IONIZING RADIATION SHIELDING PROPERTIES OF TANTALUM PENTOXIDE DOPED HIGH-DENSITY POLYETHYLENE COMPOSITES: A THEORETICAL STUDY

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Abstract

The rise in the utilization of radiation across various domains necessitates the advancement of next-generation radiation shielding materials that are devoid of lead. Due to their low weight and flexibility, polymer composites are considered as environmentally friendly alternative materials that can be used instead of toxic and high-weight lead for radiation shielding. From this point of view, the present study has focused on examining the radiation shielding performance of tantalum pentoxide doped high-density polyethylene (HDPE/Ta_2O_5) composites (including 5, 10, and 20 wt% Ta_2O_5) by using WinXCom software and MCNP6 simulation. The photon energies selected corresponded to the photons emitted from the Ba-133 (81 and 356 keV), Cs-137 (662 keV), and Co-60 (1173 and 1332 keV) radioactive sources that cover X-rays along with the low-and mid-energetic gamma-rays. The mass attenuation coefficient (μ/ρ) of the composites has been calculated within the 81 keV-1332 keV photon range by utilizing WinXCom software and MCNP6 code. The other shielding parameters such as Half Value Layer (*HVL*), effective atomic number (Z_{eff}), and effective electron density (N_{eff}) have been determined. In the light of data, it has been revealed that gradual increase in Ta_2O_5 doping while improving the μ/ρ coefficients, Z_{eff}, and N_{eff} parameters, decreasing the *HVL* length of HDPE considerably. Additionally, the parameters obtained by WinXCom and MCNP6 simulation are in good agreement. the. Ultimately, the best ionizing shielding performance among the composites has been determined for HDPE/20% Ta_2O_5 composite against 81 keV photons.

Keywords: Gamma-ray shielding, HDPE, Tantalum pentoxide, WinXCom, MCNP6

1. Introduction

Ionizing radiation, a fundamental force in the realm of nuclear science and technology, has garnered significant attention due to its diverse applications in fields ranging from medical diagnostics and cancer treatment to industrial inspection and aerospace technology. While the biological effects of ionizing radiation have been extensively studied, it is equally essential to explore its broader implications and the development of innovative shielding materials to mitigate its impact. From this point of view, radiation shielding has great importance to attenuate gamma rays which have a high penetration capacity into the substance. This is due to the radiation's genetic and biological effects on human health such as DNA damage, inherited disease, organ injury, and cancer risk [1]. Conventional shielders are made of high-density materials like concrete and lead [2]. However, the usage of lead for radiation shielding material has many disadvantages such as toxicity, low flexibility, low chemical stability, and heaviness [3]. Several researchers have proposed polymer composites as shielding materials to overcome these drawbacks. Although polymers are normally inadequate to attenuate radiation due to their low Z elements contents, researchers have incorporated additives with high atomic numbers into polymer matrices. For instance, Mahmoud et al. have produced nano and micro-sized lead oxide (PbO) doped HDPE composites with additive weight fractions varying between 10% and 50%. They investigated the shielding performance of the composites by utilizing ^{60}Co, ^{137}Cs, ^{133}Ba, and ^{241}Am point radioactive sources and XCom software. The results revealed that the maximum nano PbO additive resulted in 2.084 cm^{-1} linear attenuation coefficient at 81 keV. This value is equal to almost half of the linear attenuation coefficient of lead at 81 keV. Moreover, they determined that 50 %wt nano PbO doped HDPE composite exhibited 14.57% heaviness by supposing lead normalized to 100% [4]. On the other hand, Elsafi et al. determined the gamma-ray shielding performance of PbO-doped polypropylene (PP) composites. They determined increasing μ/ρ values with enhanced PbO contribution and the maximum linear attenuation coefficient (1.86 cm^{-1}) has been recorded at 81 keV photon energy for 50 %wt PbO doped PP composite [5]. Although these studies have reached comparatively high linear attenuation coefficients, they are not completely lead-free shielding materials. High Z elements such as bismuth (Z_{Bi}=83) and tungsten (Z_W=74) have been mostly used as low toxicity lead-free shielding material by researchers. For example, Chang et al. have produced tungsten/epoxy composites with tungsten additives ranging from 30 %wt to 80 %wt. They tested the radiation attenuation performance of the composites experimentally. The result revealed that the linear attenuation coefficient of the samples increases from 0.08 cm^{-1} to 0.24 cm^{-1} with increasing weight percent from 0 %wt to 80 %wt [6]. In another study, Ambika et al. have determined radiation shielding parameters of Polyester/Bi_2O_3 composites and reported that μ/ρ values increase with Bi_2O_3 additive [7]. On the other hand, barium, zirconium, and gadolinium have also been used as lead-free radiation shielders due to their superior radiation absorption in lower photon energies [8]. For instance, Issa et al. have investigated the radiation shielding performance of the $BaTiO_3$ reinforced polyvinylalcohol (PVA) composites by using ^{241}Am, ^{133}Ba, ^{152}Eu, and ^{137}Cs point sources and MCNP code. They have determined that μ/ρ values of PVA significantly increase with $BaTiO_3$ addition [9]. Fontainha et al. also tested Poly(vinylidene fluoride–tryfluorethylene)/Zirconium dioxide composites for effective ionizing radiation shielding. They have shown that a 1 mm thick composite with 10% ZrO_2 can attenuate the X-ray beam by 60%. [10]. Also, Shreef and Abdulzahara prepared poly(methyl methacrylate (PMMA)/nano Gd_2O_3 composites. The results of this study reveal that as the nanoparticle gadolinium oxide concentration increases, μ/ρ values of the composite increase [11].

For radiation shielding applications, as a high Z element, tantalum (Z=73), is one of the elements that attracts great interest from researchers. K absorption edge of tantalum is at 67.4 keV, this allows it more effective X-ray shielding performance than tungsten which is mostly used as a shielding material (K absorption edge is at 69.5 keV). There are a few studies using tantalum as a filler for polymer composites. For instance, Adlienė et al. tested radiation shielding performance of polymer composites including Ta and Ta_2O_5 [8], Prabhu et al.

determined radiation attenuation performance of nano and micro-sized Ta_2O_5 doped epoxy resin composites [12] and Ogul investigated radiation shielding properties of polylactic acid and acrylonitrile butadiene styrene composites doped with tantalum carbide [13]. They have reached promising results for lead-free polymeric radiation shielding materials with tantalum doping.

The main purpose of this study is to offer next-generation lead-free, flexible polymeric composites including Ta_2O_5 as radiation shielding. To achieve this objective, the shielding capabilities of HDPE/Ta_2O_5 composites have been examined using WinXCom software and MCNP6 simulation. The investigation focused on various Ta_2O_5 concentrations ranging from 5%wt to 20%wt, assessing their performance in terms of gamma-ray shielding parameters such as μ/ρ, *HVL*, Z_{eff}, and N_{eff}.

2. Material and method

2.1 Theoretical background of radiation shielding parameters

To evaluate radiation shielding performance of the composites, fundamental shielding parameters such as the linear attenuation coefficient (μ) and μ/ρ, *HVL*, Z_{eff}, and N_{eff} should be examined. The linear attenuation coefficient is defined as the probability of interaction per length of radiation. According to the well-known Beer-Lambert equation, μ is expressed by Eq.(1) [14, 15, 16, 17].

$$\mu = \frac{ln[I/I_0]}{x}\ (cm^{-1}) \qquad (1)$$

where I_0 and I is the intensities of the incident and transmitted gamma rays, respectively. The term x is the thickness of the material. The material density is an essential parameter in terms of radiation attenuation. For this reason, the definition of μ/ρ is required. μ/ρ is defined as the value of μ divided by the density of material (ρ) (see Eq. (2)).

$$Mass\ attenuation\ coefficient = \frac{Lineat\ attenuation\ coefficient}{\rho}(cm^2g^{-1}) \qquad (2)$$

Another essential radiation shielding parameter, *HVL*, has been used. It is the distance required to attenuate incoming energy to its half value and is defined by Eq. (3).

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

Z_{eff} and N_{eff} are other crucial radiation shielding parameters that should be calculated to evaluate shielding performance of the materials. There are several effective methods to calculate Z_{eff} value. However, when the studies are examined, it is seen that the effective atomic numbers calculated by the Direct, interpolation and Auto-*Z*eff methods show good results and are in harmony with the experimental results, especially in the Compton scattering and pair production energy regions. For this reason, the interpolation method was used in this article. The interpolation method is one of the most used methods. It is implemented by Eq. (4) [18, 19, 20, 21].

$$Z_{eff} = \frac{Z_1(log\sigma_2 - log\sigma_{eff}) + Z_2(log\sigma_{eff} - log\sigma_1)}{log\sigma_2 - log\sigma_1} \qquad (4)$$

where σ_1 and σ_2 symbolize the cross-section of the lowest and the highest value near σ_{eff}, respectively. Moreover, Z_1 and Z_2 represent to the atomic numbers of the elements corresponding to the cross-sections of σ_1 and σ_2, respectively. On the other hand, σ_{eff} is the total atomic cross-section of the material and it is defined by Eq. (5).

$$\sigma_{eff} = \frac{(MAC)_{material}}{N_A \sum f_i/A_i} \qquad (5)$$

where N_A is Avogadro constant, A_i are the atomic mass and f_i is the molar fraction of ith element. N_{eff} is calculated by Equation 6 and it is closely related to Z_{eff}.

$$N_{eff} = N_A \frac{Z_{eff}}{<A>} \qquad (6)$$

where N_A is Avogadro constant and effective atomic mass value is defined by Eq. (7) [22, 23].

$$< A >= \sum n_i A_i / n \qquad (7)$$

2.2 WinXCom calculation

WinXCom is the adopted version of XCom computer program produced by Hubbell and Berger for Windows operating system [24]. As is known, WinXCom is a computer program that calculates the cross-sections for photon interactions with the elements and compounds of the periodic table. In WinXCom, mixing rule is valuable tool for quickly estimating the behavior of composite materials without the need for complex simulations or experiments. However, they are approximations and may not capture all nuances of radiation interactions in composite materials. The data obtained with WinXCom for a composite may differ in the range of ±0.5%-7% from the experimental data of the same material [16, 25]. This difference is largely due to the fact that the amount of elements theoretically assumed to be in the relevant composite does not coincide with the amount of material in the experimentally produced one.
μ/ρ values of the HDPE/Ta_2O_5 composites have been calculated by the mixture rule (see Eq. (8)) implementing WinXCom software at energies between 1 keV and 100 GeV.

$$(\mu/\rho)_{alloy} = \sum_i^n w_i (\mu/\rho)_i \qquad (8)$$

where w_i is the weight of ith element and $(\mu/\rho)_i$ is μ/ρ value of the ith element.

2.3 Monte Carlo N-Particle simulation

Monte Carlo N-Particle (MCNP) is a commonly used computer code to simulate radiation motion. It is based on the Monte Carlo method, which uses random numbers to simulate the behavior of individual particles as they interact with a material. The code traces the path particles take as they travel and interact with the material, creating a detailed diagram of the radiation's motion until the particle disappears. By simulating large numbers of particles, MCNP can accurately predict radiation behavior in complex geometries and materials.

MCNP6 is the latest version of the MCNP code developed by Los Alamos National Laboratory. It includes many improvements, such as faster execution time and improved accuracy for certain types of simulations. MCNP6 is a useful tool that can be used for a wide variety of radiation transport simulations, including nuclear reactor design, medical physics applications, and calculations of particle physics. The MCNP6 can also be utilized to predict the amount of dose reduction provided by various shielding configurations and to improve the design of the shielding for a particular radiation source and exposure scenario. In MCNP, the factors that affect the difference between experimental data and simulation are simulation code and implementation, input data, number of particles (statistics), energy range, geometry and boundary conditions, simulation parameters, modeling assumptions, verification and validation, computational resources and code validation. In Figure 1, Monte Carlo simulation geometry used in this study is given.

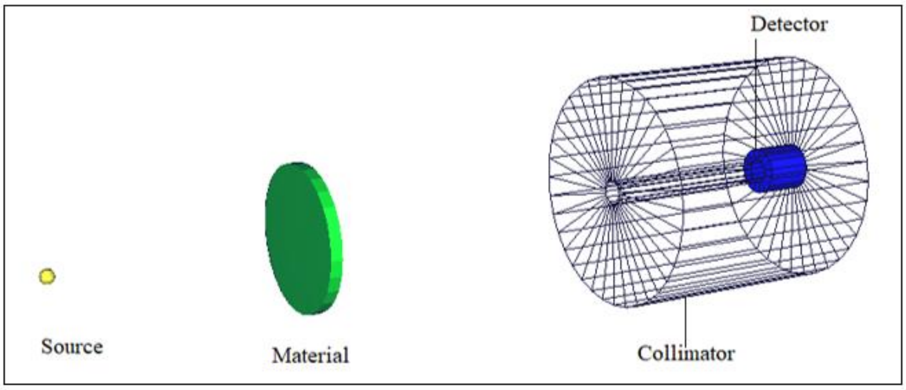

Figure 1. Monte Carlo Simulation Geometry

μ/ρ values of HDPE and HDPE/Ta_2O_5 composites have been calculated by utilizing MCNP6 simulation code for specific photon energies such as 81, 356, 662 1173, and 1332 keV. These photon energies are the photons emitted from the Ba-133 (81 and 356 keV) , Cs-137 (662 keV), and Co-60 (1173 and 1332 keV) radioactive sources.

3. Results and discussions

3.1 The mass attenuation coefficient values of the HDPE/Ta_2O_5 composites

The μ/ρ values of the pure HDPE and HDPE/Ta_2O_5 composites have been determined by both WinXCom and MCNP6 at specific gamma energies ranging from 81 keV to 1332 keV. The results have been shown in Table 1.

Table 1. μ/ρ values of HDPE/Ta_2O_5 composites.

MAC (cm^2g^{-1})	Sample	HDPE	HDPE/5 %Ta_2O_5	HDPE/10% Ta_2O_5	HDPE/20% Ta_2O_5
81 keV	**WinXCom**	0.182	0.475	0.768	1.355
	MCNP6	0.200	0.498	0.792	1.366
356 keV	**WinXCom**	0.114	0.119	0.123	0.132
	MCNP6	0.116	0.121	0.125	0.134
662 keV	**WinXCom**	0.088	0.088	0.088	0.089
	MCNP6	0.088	0.088	0.088	0.089
1173 keV	**WinXCom**	0.067	0.067	0.066	0.065
	MCNP6	0.088	0.088	0.088	0.089
1332 keV	**WinXCom**	0.063	0.062	0.062	0.061
	MCNP6	0.088	0.088	0.088	0.089

According to Table 1, it has been determined that the variations between WinXCom and MCNP6 values remain acceptable for pure HDPE and the composites. Therefore, it could be deduced that there is an agreement between WinXCom and MCNP6 results. On the other hand, μ/ρ values increase gradually with Ta_2O_5 additive for the low photon energies such as 81 keV and 356 keV. It can be seen from Table 1 that the highest μ/ρ value has been determined for the HDPE/20% Ta_2O_5 composite. The μ/ρ value for the related composite (1.33 cm^2/g) is 7.47 times higher than that of the pure HDPE (0.182 cm^2/g) at 81 keV. Moreover, HDPE/10% Ta_2O_5 and HDPE/5% Ta_2O_5 composites have 4.21 and 2.60 times higher μ/ρ values than that of the pure HDPE at 81 keV, respectively. These results indicate that the incorporation of Ta_2O_5 into HDPE matrix improves the radiation shielding performance of the HDPE. However, for each sample, when the photon energy is increased from 81 keV to 1332 keV, a significant decrease in μ/ρ values has been observed. In addition, the increase in the Ta_2O_5 contribution cannot help to enhance the μ/ρ values, especially 1173 keV and 1332 keV. The decrease in μ/ρ values due to increasing photon energy and the increase in μ/ρ value due to increasing doping compound at low photon energies can be explained as follows. At photon energies as low as 81 keV and 356 keV, the dominant material-radiation interaction mechanism is the photoelectric interaction mechanism, and the photoelectric cross section strongly depends on the n^{th} power of the inverse of the incoming photon energy ($\sigma_{photoelectric} \propto E^{-3.5}$) [26]. This dependence characterizes itself by increasing μ/ρ values with the contribution of the higher effective atomic number compound increasing at low photon energies. On the other hand, as the incoming photon energy increases (662 keV and above), the dominant photoelectric interaction mechanism is replaced by Compton scattering, and accordingly, lower μ/ρ values are observed. It's a known fact that the cross section for Compton scattering increases in inverse proportion to the incident photon's energy ($\sigma_{Compton} \propto E^{-1}$) [27].

Moreover, when the μ/ρ values given in Table 1 are compared with epoxy/10% Ta_2O_5, another polymer composite reinforced with micro-sized Ta_2O_5, the experimental and WinXCom $\mu/$ values of the related composite have been found to be as 0.119±0.003 cm^2/g and 0.116 cm^2/g for photons having 356 keV energy, respectively. Similarly, the μ/ρ values of 0.127±0.006 cm^2/g (exp.) and 0.126 cm^2/g (WinXCom) for 356 keV for the epoxy/20% Ta_2O_5 composite have been reported by Prabhu et al. [12] As is tabulated in Table 1, all of these values are below the values predicted for 356 keV photon energy for HDPE/10% Ta_2O_5 and HDPE/20% Ta_2O_5 composites. In this context, it can be thought that micro-sized Ta_2O_5 additive gives HDPE a better radiation shielding ability than epoxy. On the other hand, when the μ/ρ values in Table 1 are compared at 81 keV for another HDPE-based composite, it is seen that the experimental μ/ρ values of the composites containing 10% and 20% micro-sized CdO lag behind the μ/ρ values determined in this study. Namely; the μ/ρ values at 81 keV were determined as 0.3822 cm^2/g and 0.59226 cm^2/g for HDPE-based composites containing 10% and 20% micro-size CdO by Ktahib et al., respectively [3]. These values are below the values expressed in Table 1 as 0.768 cm^2/g for HDPE/10% Ta_2O_5 and 1.355 cm^2/g for HDPE/20%Ta_2O_5 at 81 keV. In this context, it can be stated that the Ta_2O_5 additive has a promising potential to improve the ionizing radiation shielding properties of HDPE.

Figure 2 also depicts the variation of μ/ρ values calculated by WinXCom for an incident photon energy between 1 keV and 1 MeV for HDPE/Ta_2O_5 composites. As shown in Figure 2, due to its low-Z element content, pure HDPE displays the lowest μ/ρ value compared to composites at all photon energies.

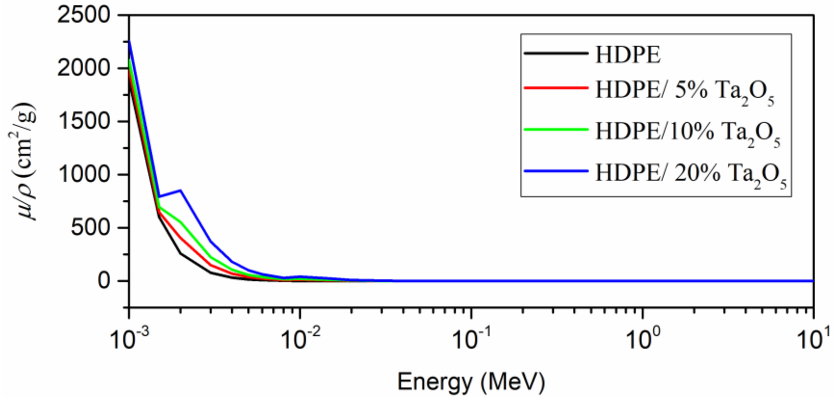

Figure 2. μ/ρ values of the pure HDPE and HDPE/Ta_2O_5 composites.

In Figure 2, it can be seen that μ/ρ values of the composites decrease with increasing photon energy. This tendency of μ/ρ values is in good agreement with studies in the scientific literature [7, 8, 12]. This behavior of μ/ρ versus photon energy curves is related to photon-matter interaction. While the probability of interaction is sufficiently high at lower energies, it decreases at higher photon energies [3].

3.2 Half-value layer lengths of the HDPE/Ta_2O_5 composites

HVL values of the pure HDPE and HDPE/Ta_2O_5 composites have been calculated by Eq. (3). The variations of *HVL* values with respect to both increasing Ta_2O_5 contribution and photon energy have been illustrated in In Figure 3, the *HVL* values of the samples at several specific energies have been determined by using WinXCom findings.

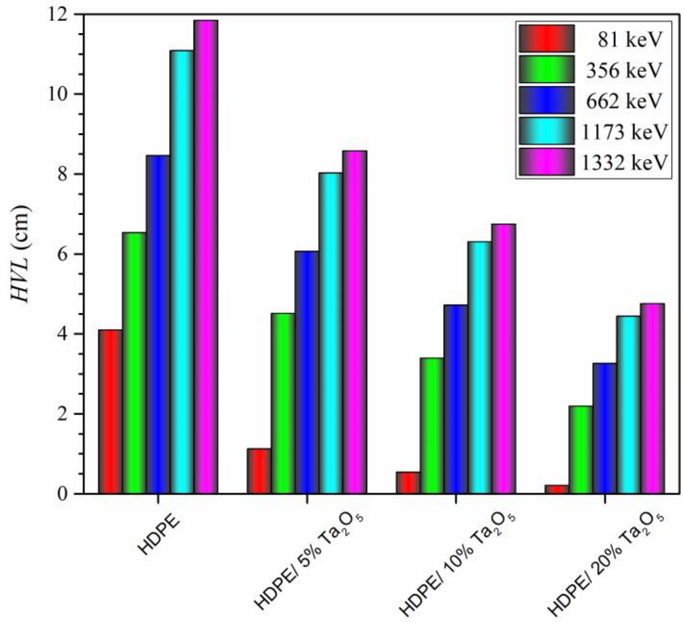

Figure 3. HVL values pure HDPE and HDPE/Ta_2O_5 composites.

As shown in Figure 3, *HVL* values decrease gradually with increasing Ta_2O_5 additive. The pure HDPE material exhibited the highest *HVL* value, while the HDPE/20% Ta_2O_5 composite demonstrated the lowest *HVL* value. This tendency is quite compatible with the results related to μ/ρ values of the composites. As is known, *HVL* indicates the distance required to reduce incoming energy to its half value. So, the lower *HVL* values correspond to better radiation shielding performance. From this aspect, it can be deduced that with increasing Ta_2O_5 addition, *HVL* values decrease and the shielding ability of the composites improves. So, the increase in μ/ρ values (Figure 2) and the decrease in the *HVL* values with Ta_2O_5 addition are coherent with each other. This can be associated with the increase of high Z element content in the composites due to Ta_2O_5 doping. Moreover, when the *HVL* values (3.39 cm and 2.19 cm) of HDPE/10% Ta2O5 and HDPE/20% Ta_2O_5 composites for 356 keV are compared with the 4.58 cm and 3.92 cm values of epoxy/10% Ta_2O_5 and epoxy/20% Ta_2O_5composites at the same energies by Prabhu et al. [12], again it can be confirmed that the combination of Ta_2O_5 with HDPE results in better radiation shielding. The present results also showed that the composite with the best radiation shielding performance was HDPE/%20 Ta_2O_5. Moreover, it can be concluded that Ta_2O_5 addition improves the radiation shielding performance of pure HDPE.

3.3 The effective atomic number values of the HDPE/Ta_2O_5 composites

Z_{eff} and N_{eff} values of the pure HDPE and the HDPE/Ta_2O_5 composites for the increasing incident photon energies from 1 keV to 100 GeV have been calculated with Eqs. (4) and (6) by using WinXCom software. Figures 4 and 5 illustrate the variation of Z_{eff} and N_{eff} values with increasing photon energy, respectively.

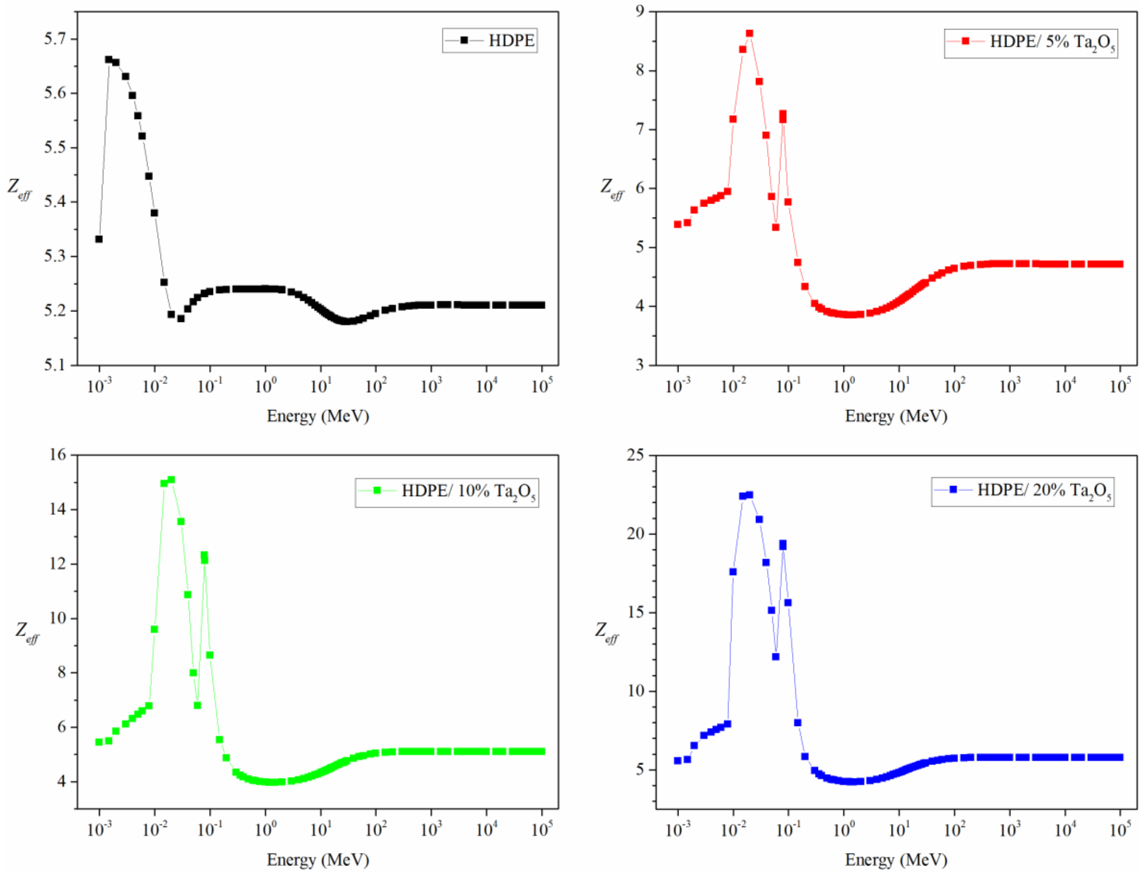

Figure 4. The variation of Z_{eff} with incident photon energy for a) HDPE, b) HDPE/5% Ta_2O_5, c) HDPE/10% Ta_2O_5, and d) HDPE/20% Ta_2O_5

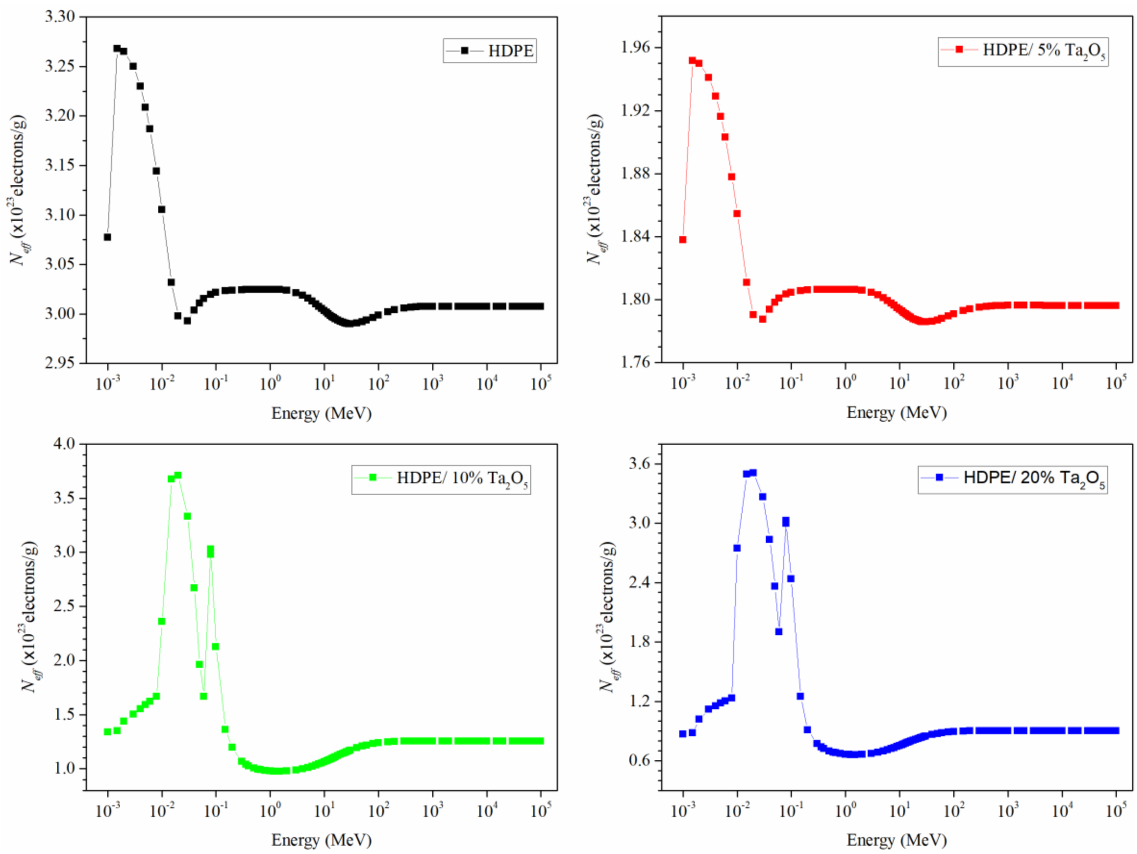

Figure 5. The variation of N_{eff} with incident photon energy for a) HDPE, b) HDPE/5% Ta_2O_5, c) HDPE/10% Ta_2O_5, and d) HDPE/20% Ta_2O_5

Since N_{eff} is directly proportional to Z_{eff}, the tendencies of the Z_{eff} and N_{eff} curves of the pure HDPE are the same [18, 28, 29]. Higher Z_{eff} and N_{eff} values indicate better gamma ray attenuation. As shown in Figure 4 and Figure5, Z_{eff} and N_{eff} values increase gradually with the Ta_2O_5 additive. Among the composites, the highest Z_{eff} and N_{eff} values have been observed for the HDPE/20% Ta_2O_5 composite. This result indicates the best shielding performance for HDPE/20% Ta_2O_5 composite which is consistent with its greatest μ/ρ and lowest *HVL* values. This could be related to the incorporation of high Z particles into the polymer matrix. On the other hand, as can be seen from both Figure 4 and Figure 5, especially for 10% and 20% Ta_2O_5 contributions, Z_{eff} and N_{eff} first decreased with increasing photon energy between 17.5 keV and 87.7 keV in the related graphics, and then increased with increasing photon energy around 60 keV. This was observed concerning the K-edge discontinuity reported around 67 keV for Ta [30].

4. Conclusions and recommendations

In the scientific literature, it can be seen that polymers incorporated by high Z particles are unique materials for gamma radiation shielding due to their superior properties such as low weight, flexibility, and environmental friendliness. Motivated by these advantages, this study aims to examine the radiation shielding performance of HDPE polymer composites doped with Ta_2O_5.

The radiation shielding performance of the composites has been investigated by the most commonly used radiation shielding parameters such as μ/ρ, *HVL*, Z_{eff}, and N_{eff} by utilizing

WinXCom software and MCNP6 simulation. The findings revealed that MAC value of the HDPR can be increased by doping it with Ta_2O_5. Furthermore, the HDPE/20% Ta_2O_5 composite showed the highest μ/ρ values among the composites. This composite showed a 7.47 times higher μ/ρ value than that of pure HDPE. *HVL* values for all composites diminished as the concentration of Ta_2O_5 increased. In accordance with both μ/ρ and *HVL* values, the best radiation shielding performance has been determined for the HDPE/20% Ta_2O_5 composite. Moreover, the incorporation of Ta_2O_5 into the HDPE matrix has been shown to gradually increase Z_{eff} and N_{eff}, which can be attributed to better shielding performance. As a consequence, the HDPE/Ta_2O_5 composite having the highest Ta_2O_5 contribution can be suggested as a promising shielding material against X-ray and low-energy gamma-ray (up to 356 keV) that are utilized in both diagnosis and treatment in medicine.

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