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Performance of PMMA/Ta2O5 Composites as Medical Radiation Shielding: WinXCom and MCNP6 Studies

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Abstract − The growing reliance on radiation in contemporary applications underscores the imperative to safeguard individuals and the environment from harmful consequences. To mitigate the adverse effects of radiation, polymer composites have begun to garner interest from researchers as potential lead-free shielding materials, largely due to their distinctive attributes, including flexibility, lightness, and environmental benignity. In this study, the gamma radiation shielding capacity of polymethyl methacrylate (PMMA) composites reinforced with varying proportions of Ta_2O_5 (5%, 10%, and 20% wt) was investigated through the utilization of Windows version of photon cross sections on a personal computer (WinXCom) software and the Monte Carlo N-Particle 6 (MCNP6) code. The alignment of the WinXCom and MCNP6 results, despite their different methodologies, provides a robust and reliable understanding of the radiation shielding performance of these composites. The present study investigated the radiation attenuation properties of PMMA/Ta_2O_5 composites about shielding coefficients, including mass attenuation coefficients (MAC), half-value layer (HVL), and effective atomic number (Z_{eff}). The findings indicated that all composites demonstrated enhanced shielding performance compared to pure PMMA. The PMMA/20% Ta_2O_5 composite exhibited MAC values of 1.22-, 1.29-, and 1.28-fold greater than those observed in the silicon-based composites. The MAC increase was observed in the PMMA/20% Ta_2O_5 composite at an energy of 81 keV. The PMMA/20% Ta_2O_5 composite demonstrated the most effective radiation shielding properties. In light of these findings, the PMMA/20% Ta_2O_5 composite can be regarded as a flexible, lightweight, and environmentally friendly shielding material, reassuring these composites' reliability in practical applications.

Keywords − *Monte Carlo simulation, radiation shielding, tantalum pentoxide, MCNP6*

1. Introduction

The utilization of ionizing radiation with a high energy level is progressively increasing across various fields, including medical diagnostics, nuclear reactors, food irradiation, research laboratories, particle accelerators, etc. In light of the detrimental impact of radiation on human health, one of the most crucial strategies to safeguard individuals from exposure to high-energy radiation is the deployment of a suitable shielding material. As the conventional shielding material, lead, with its high atomic number and density, is widely used to attenuate high-energy radiation. However, it is also toxic, inflexible, heavy, and has low mechanical strength [1]. The aforesaid disadvantages restrict the utilization of lead, particularly for those engaged in occupations within sectors such as medical diagnosis, research laboratories, and nuclear power plants, who are required to wear lead shields, such as aprons, for extended periods. This can result in significant health issues, including physical fatigue and back pain [2].

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In this context, the researchers have concentrated on producing low-weight, environmentally friendly, and flexible alternative materials that can replace lead. A substantial body of research has been conducted on the radiation attenuation performance of polymer composites that exhibit the desired properties. In these studies, polymers have been doped with high Z elements to enhance their shielding capabilities in their pure form, which cannot effectively attenuate high-energy radiation. To illustrate, the radiation-shielding performance of polyvinyl alcohol/tungsten oxide [3], high-density polyethylene/lead oxide [4], poly ether ketone/gadolinium oxide [5], poly (methyl methacrylate)/ MWCNT/Bi_2O_3 [6], and other similar composites have been the subject of extensive study. The results demonstrated that the shielding efficacy of these composites enhanced with the additive concentration. PMMA has been evaluated as a shielding material in numerous studies—for instance, Bel et al. [7] quantified the neutron and gamma radiation attenuation properties of PMMA/colemanite composites using a Cs-137 radioactive source and a Windows version of photon cross sections on a personal computer (WinXCom) software. The outcomes revealed that the shielding capability of PMMA/colemanite composites increased by 11.1% for gamma radiation and 38.56% for neutrons. Cao et al. [8] investigated the gamma radiation shielding capacity of PMMA/Bi_2O_3 composites. The gamma-ray attenuation coefficients, including MAC and (HVL), were conducted using five gamma point sources (^{57}Co, ^{60}Co, ^{109}Cd, ^{133}Ba, and ^{137}Cs). The incorporation of Bi_2O_3 particles into the PMMA matrix was observed to enhance the shielding ability of pure PMMA. It was put forth that these composites are viable candidates for use as radiation shielding materials. In a separate study, Shareef et al. [9] investigated the radiation attenuation performance of PMMA/Gd_2O_5 composites using ^{60}Co and ^{137}Cs point sources. The findings indicated that the shielding capacity of the composites exhibited a gradual increase with an increase in Gd_2O_5 concentration. Among the additives incorporated into the polymers to enhance their shielding capabilities, tantalum pentoxide (Ta_2O_5) is particularly noteworthy due to its high atomic number (Z=73) and K absorption edge value of 67.4 keV, which renders it an efficient X-ray shielding material [10].

Therefore, Ta_2O_5 can be deemed an appropriate compound for investigation as a radiation shielding agent, and its shielding efficacy has been evaluated in numerous studies. In a previous study, Prabhu and colleagues [11] investigated the gamma-ray attenuation potency of epoxy micro and nano Ta_2O_5 composites using ^{22}Na, ^{60}Co, ^{133}Ba, and ^{137}Cs radioactive point sources. The MAC for the epoxy composites with the highest micro and nano dopant content (30% by weight) were found to be 0.136 cm^2/g and 0.154 cm^2/g, respectively, at an energy of 0.356 MeV. In a separate investigation, Adliene and colleagues examined the X-ray shielding characteristics of polydimethylsiloxane (PDMS) silicone rubber and universal silicone rubber (UNSI)/Ta and Ta_2O_5 composites at energies range 50 to 150 keV. The researchers investigated lead equivalent values for composites. It estimated that the UNSI composite with the maximum concentration of Ta and Ta_2O_5 (50 wt%) exhibited higher values than the recommended 0.25 mmPb value. The composite with Ta exhibited a value of 0.364 mmPb, while the composite with Ta_2O_5 exhibited a value of 0.313 mmPb [10].

This study aimed to ascertain the gamma radiation shielding properties of poly (methyl methacrylate) (PMMA)/Ta_2O_5 composites to identify a lightweight, flexible, and chemically stable alternative to lead-based shielding materials. The fundamental radiation shielding coefficients, including the MAC, HVL, and Z_{eff}, were investigated using WinXCom software and MCNP6 simulation code.

2. Materials and Methods

2.1. Shielding Parameters

By the Beer-Lambert equation, the intensity of a gamma ray beam traversing a shield is observed to decrease exponentially in proportion to the chemical composition of the shield in (2.1).

$$I = I_0 e^{-\mu x} \tag{2.1}$$

In this context, the terms I and I_0 represent the intensity of the incident and transmitted gamma rays, respectively. In this context, the terms μ and x indicate the linear attenuation coefficient and the thickness of the shield material, respectively [11]. As the linear attenuation coefficient does not account for the density of the shield, the MAC (μ_m) is defined as the linear attenuation coefficient per volume (ρ).

$$\mu_m = \frac{\mu}{\rho}(\mathrm{cm^2 g^{-1}}) \tag{2.2}$$

The HVL is another shielding coefficient, defined as the length that reduces the incoming radiation to its half-value. This is expressed in (2.3) [12].

$$\mathrm{HVL} = \frac{\ln 2}{\mu}\ (\mathrm{cm}) \tag{2.3}$$

On the other hand, since the atomic number of the materials is essential for radiation shielding, the Z_{eff} is described as the atomic number of multi-element materials such as mixtures or compounds. Z_{eff} can be calculated by interpolation method according to (2.4).

Conversely, given the crucial role of atomic number in radiation shielding, Z_{eff} is defined as the atomic number of multi-element materials, including mixtures and compounds. The Z_{eff} can be calculated using an interpolation method using the specifications in (2.4).

$$\mathrm{Z_{eff}} = \frac{\mathrm{Z_1}(\log\sigma_2 - \log\sigma_{\mathrm{eff}}) + \mathrm{Z_2}(\log\sigma_{\mathrm{eff}} - \log\sigma_1)}{\log\sigma_2 - \log\sigma_1} \tag{2.4}$$

In this context, σ_{eff} represents the effective cross-section of the shield material with σ_1 and σ_2 denoting the minimum and maximum cross-section values in the vicinity of σ_{eff}, respectively. Additionally, σ_{eff} can be defined by (2.5), contingent on the MAC [1].

$$\sigma_{\mathrm{eff}} = \frac{\left(\frac{\mu}{\rho}\right)_{\mathrm{material}}}{N_A \sum \frac{f_i}{A_i}} \tag{2.5}$$

where A_i is the atomic mass and f_i is the molar fraction of i^{th} element. The quantity N_A is defined as the Avogadro constant.

2.2. WinXCom and MCNP6

WinXCom represents the Windows operating system version of the XCom software developed by Berger and Hubbell [13]. The software can calculate the MAC values and the cross-sections of the shielding materials between 1 keV and 100 GeV per the Mixture Rule, as outlined in (2.6). By this rule, the MAC values of a given material, which may comprise various elements, are calculated as the sum of the weight ratio of each atom present [14].

$$(\mathrm{MAC})_{\mathrm{sample}} = \sum_i^n w_i (\mathrm{MAC})_i \tag{2.6}$$

where $(MAC)_i$ is MAC value and w_i is the weight of the i^{th} element. The MAC values of the studied composites were calculated by WinXCom software.

Monte Carlo N-Particle (MCNP), a computer code designed by researchers at the Los Alamos National Laboratory, is a widely utilized tool for simulating the transport of particles, including neutrons, photons, and electrons, through a material. A narrow beam geometry consisting of a detector was designed by MCNP and is illustrated in Figure 1. The MCNP6 calculations were performed using the Evaluated Nuclear Data Files (ENDF)/B-VI-Released 8 and the photatomic library MCPLIB04. The sample was situated between the detector and the radioactive source, as illustrated in Figure 1. The simulation was conducted at five discrete energy values: 81, 356, 662, 1173, and 1332 keV. The initial run was conducted without a sample to ascertain the total flux entering the detector. As shown in Table 1, a simulation was performed for the corresponding composites with varying chemical compositions.

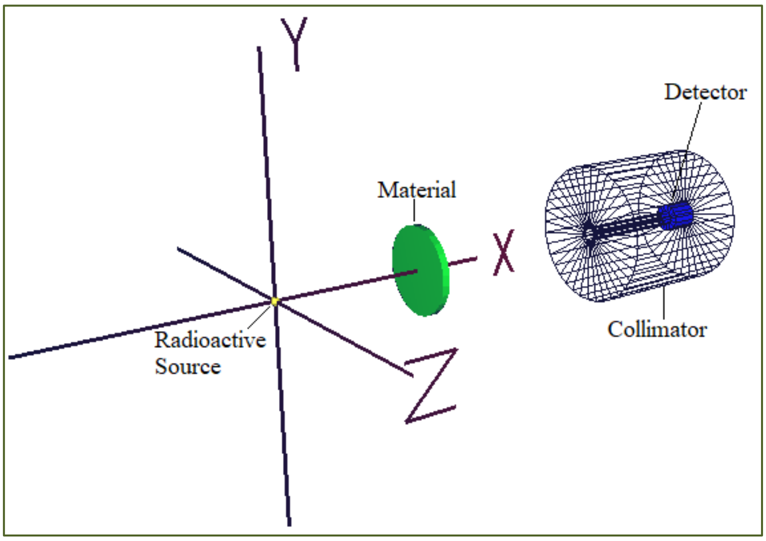

Figure 1. The geometry of the detector, source, and shielding material, as well as their placement on axes in a three-dimensional coordinate system, have been simulated using the MCNP6 software

2.3. Calculation of Radiation Shielding Parameters

To investigate the radiation shielding performance of PMMA/Ta_2O_5 composites, MAC values were calculated at energies ranging from 1 keV to 100 GeV using the WinXCom program. Furthermore, MAC values at specific energies, including 81, 356, 662, 1173, and 1332 keV, were also calculated using the MCNP6 simulation package. Other gamma ray shielding coefficients, such as HVL and Z_{eff}, were determined by (2.3)-(2.5), respectively. The elemental fractions and densities of pure PMMA and PMMA/Ta_2O_5 composites utilized in WinXCom and MCNP6 calculations are presented in Table 1.

Table 1. The fraction of elements of PMMA and PMMA/Ta2O5 composites

Sample	Element (wt%)				Density (g/cm^3)
	H	C	O	Ta	
PMMA	0.08054	0.59985	0.31961	--	1.180
PMMA/5% Ta_2O_5	0.07651	0.56986	0.31268	0.04095	1.531
PMMA/10% Ta_2O_5	0.07249	0.53986	0.30575	0.08190	1.882
PMMA/20% Ta_2O_5	0.06443	0.47988	0.29190	0.16379	2.584

3. Results and Discussion

3.1. MAC Values of PMMA and PMMA/Ta2O5 Composites

The MAC is one of the most significant parameters for evaluating the radiation attenuation properties of various materials. It characterizes the capacity of a shielding material to absorb radiation, which is contingent upon the density of the sample and the energy of the incident photons. A higher MAC value indicates an enhanced shielding capability. Figure 2 illustrates the variations in the MAC of pure PMMA and PMMA/Ta_2O_5 composites as a function of increasing photon energy.

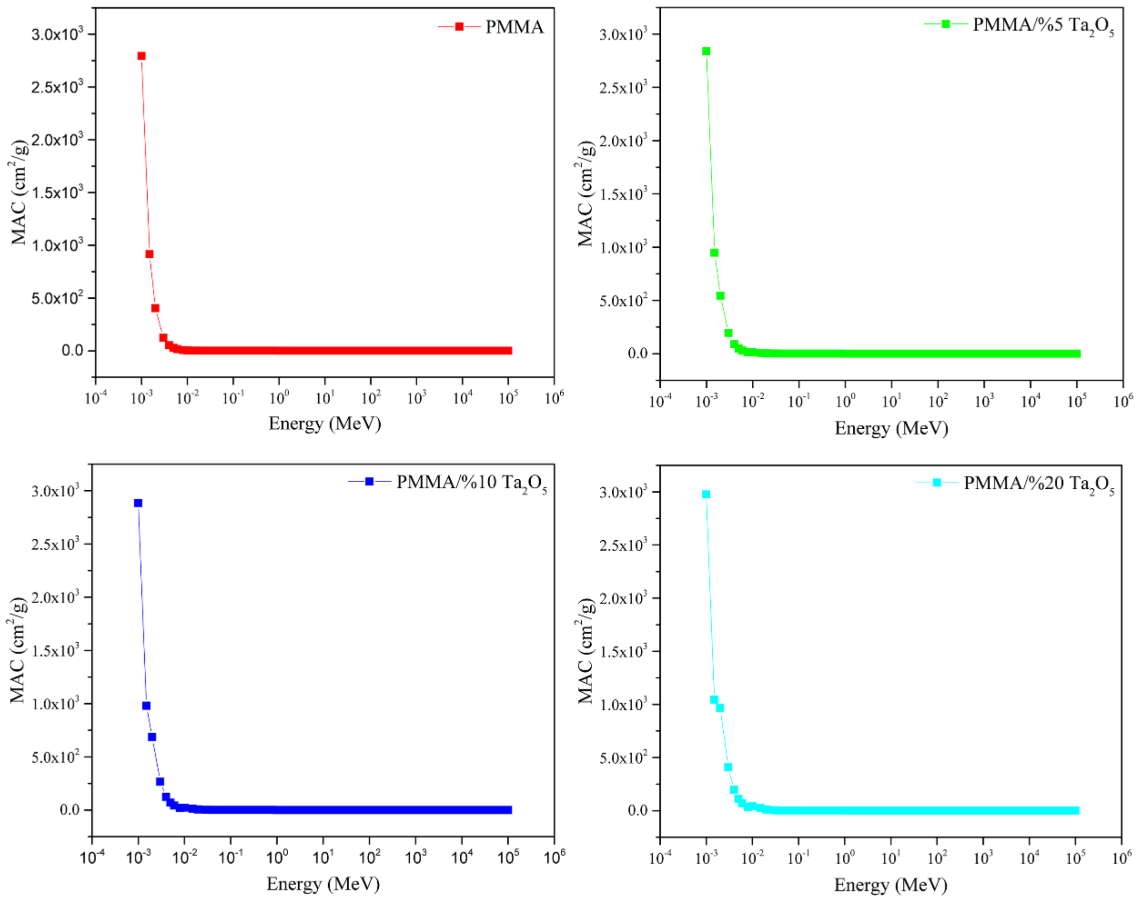

Figure 2. MAC values of PMMA and PMMA/Ta_2O_5 composites as a function of the photon energy

As illustrated in Figure 2, the measured MAC values exhibit a general decrease with increasing photon energies. This phenomenon is contingent upon the interaction mechanism between photons and matter. At low energies, the cross-section for the photoelectric interaction is sufficiently high such that the absorbing medium absorbs nearly all incident photons. This is evidenced by the elevated values observed on the MAC-Energy graph. As the incident photon energy increases, Compton scattering becomes the dominant interaction mechanism, leading to a decline in photon interaction possibilities. At high energy levels, the graph exhibits an exponential decline due to the prevalence of pair production as the dominant interaction mechanism with energy dependence [15]. The MAC values, calculated using the WinXCom software and the MCNP6 code at various energies, are presented in Table 2.

Table 2. The following chart presents the MAC values of PMMA/Ta_2O_5 composites for various energies

Sample	MAC (cm²/g)				
	WinXCom/MCNP6 81 keV	**WinXCom/MCNP6 356 keV**	**WinXCom/MCNP6 662 keV**	**WinXCom/MCNP6 1173 keV**	**WinXCom/MCNP6 1332 keV**
PMMA	0.174/0.192	0.108/0.111	0.083/0.083	0.063/0.083	0.059/0.083
PMMA/%5 Ta_2O_5	0.468/0.492	0.113/0.115	0.083/0.083	0.063/0.083	0.059/0.083
PMMA/%10 Ta_2O_5	0.762/0.785	0.117/0.120	0.084/0.083	0.063/0.083	0.059/0.083
PMMA/%20 Ta_2O_5	1.349/1.360	0.127/0.130	0.085/0.085	0.062/0.085	0.058/0.085

As illustrated in Table 2, the results produced by WinXCom are closely aligned with those obtained by MCNP6. The MAC values of the composites demonstrate a gradual increase with the addition of Ta_2O_5 at a constant energy value for low-energy particles (81, 356, and 662 keV). The minimum MAC levels are observed in the case of pure PMMA, which contains low Z element components such as C, H, and O. The gradual increase in MAC values with the incorporation of Ta_2O_5 is attributed to adding high Z elements into the PMMA matrix. The maximum increase in MAC has been determined to occur in the PMMA/20% Ta_2O_5 composite at 81 keV. The MAC value of this composite is 7.75 times greater than that of pure PMMA at this energy value. Furthermore, all composites exhibit larger MAC values than pure PMMA, indicating that the addition of Ta_2O_5 enhances the radiation shielding capacity of PMMA.

The increase in MAC values for composite materials is observed as 2.69-7.75 times greater than that of pure PMMA at an energy of 81 keV. Conversely, MAC values remain relatively constant at high energies above 662 keV, suggesting that the shielding material is inadequate for attenuating high-energy radiation. In contrast, the MAC values of the composites were compared with those of other shielding materials that had been the subject of scientific study. As an instance, the mass attenuation coefficient of the 304 L stainless steel sample was calculated as 0.100 cm^2/g at 356 keV energy by Buyukyildiz et al. The PMMA/20%Ta_2O_5 composite prepared in this study exhibited a MAC value of 0.127 cm^2/g at 356 keV, indicating that this composite exhibited a 1.27 times greater attenuation capacity than the steel sample [16]. In a separate study, Verdipoor et al. employed MCNP simulation to calculate the MAC values of silicon resin composites loaded with tungsten trioxide (WO_3), lead oxide (PbO), and bismuth oxide (Bi_2O_3). The maximum MAC values in the maximum additive-doped composites were determined to be 0.1039, 0.0980, and 0.0989 cm^2/g for the 0.5 wt% WO_3-0.5 wt% silicon resin, 0.5 wt% PbO-0.5 wt% silicon resin, and 0.5 wt% Bi_2O_3-0.5 wt% silicon resin composite, respectively, at 356 keV energy [17]. PMMA/20%Ta_2O_5 composite showed 1.22-, 1.29- and 1.28-times bigger MAC values than these silicon-based composites.

In addition, the HVL parameters of the samples were examined to ascertain their shielding efficacy. The term HVL is defined as the length required to reduce the intensity of incident radiation to a level that is half of its original value. Lower HVL values indicate that the radiation can traverse a shorter distance through the absorbing medium, which enhances radiation attenuation ability. Figure 3 compares the HVL values of PMMA/Ta_2O_5 composites for a range of gamma energies.

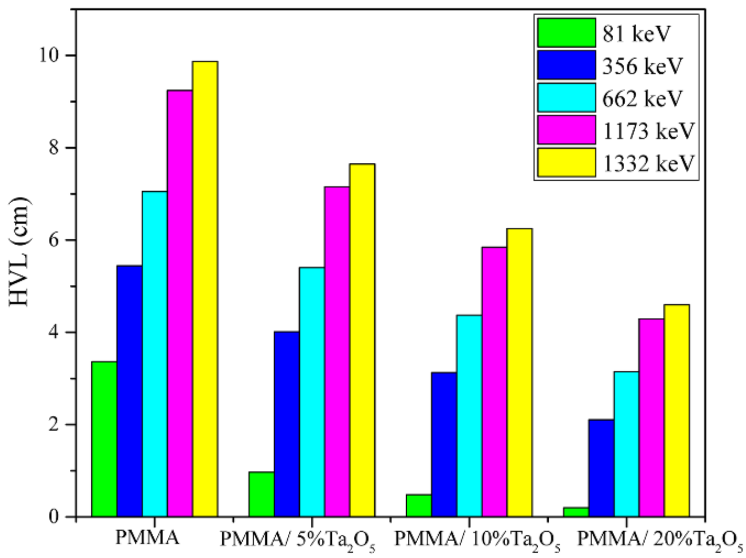

Figure 3. HVL values of PMMA/Ta_2O_5 composites at various photon energies

As illustrated in Figure 3, the HVL values demonstrate an increase in photon energies and a decline with increasing Ta_2O_5 doping concentration. The elevated HVL values at higher energies suggest that radiation penetration increases with photon energy. Consequently, a greater length is necessary to achieve the desired reduction in radiation. It can thus be concluded that the samples exhibit superior shielding performance at low energies. By the MAC values of the composites, the lowest HVL value, indicative of the optimal shielding performance, is observed in the PMMA/20% Ta_2O_5 composite at all energy values. Furthermore, all of the composites exhibit lower HVL values than pure PMMA. The HVL value of the PMMA/20% Ta_2O_5 composite is 16.9 times smaller than that of pure PMMA, indicating that this composite exhibits the best shielding performance.

Z_{eff}, defined as the average atomic number of multi-element materials, is another crucial parameter for evaluating the gamma-ray shielding capability of PMMA/ Ta_2O_5 composites. The variation of Z_{eff} of pure PMMA and PMMA/ Ta_2O_5 composites with increasing energy is illustrated in Figure 4.

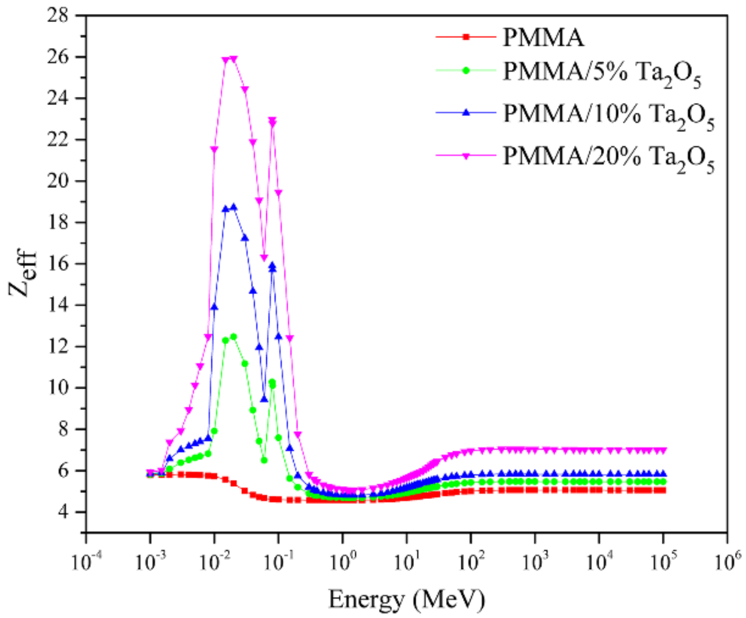

Figure 4. The variation of Z_{eff} of PMMA and PMMA/Ta_2O_5 composites with gamma energies

As illustrated in Figure 4, all composites and pure PMMA display Z_{eff}-energy plots consistent with those observed in other studies in the literature [1,18,19]. This characteristic of Z_{eff}'s behavior about photon energy is associated with the interaction between radiation and matter. In these graphs, Z_{eff} tends to have maximum values at the low-energy region due to the dominance of the photoelectric event as the interaction mechanism, which depends on the atomic number Z. As the energy of the gamma photon increases, Compton scattering becomes the dominant interaction, whose cross-section is independent of Z, which manifests as a decrease in the graph. Nevertheless, there is a sudden rise in the Z_{eff} graphs of the composites in this area at approximately 67 keV. This phenomenon is attributed to the K absorption edge of Ta_2O_5. Furthermore, at elevated energies, the Z_{eff} values demonstrate an increase due to the dominance of pair generation over Compton scattering [20]. As illustrated in Figure 4, the Z_{eff} values of the composites exhibit a discernible increase with the addition of Ta_2O_5. This increase in Z_{eff} values with Ta_2O_5 addition can be attributed to the rise in the concentration of high Z elements in the composites. The lowest Z_{eff} value is observed for the pure PMMA, while the highest is for the PMMA/20%Ta_2O_5 composite. Thus, the PMMA/20%Ta_2O_5 composite exhibits Z_{eff}'s most effective gamma ray attenuation performance.

4. Conclusion

Lead has been employed for decades to protect high-energy radiation, yet it presents several drawbacks, including toxicity, high weight, and low processability. In light of these limitations, researchers have directed their attention toward developing alternative materials to address the shortcomings associated with lead. It is anticipated that these alternative materials will be light in weight, environmentally friendly, and sufficiently flexible to meet the requirements of the intended application. Polymer composites represent a class of materials that provide the aforementioned desired properties. This study investigated the gamma-ray shielding capacity of PMMA/ Ta_2O_5 composites as a potential lead-free shielding material. The radiation shielding capacity of the composites was determined using WinXCom software and the MCNP6 code. The findings indicated that the incorporation of Ta_2O_5 into PMMA led to an enhancement in radiation shielding parameters. The MAC values increased with incorporating Ta_2O_5, exhibiting a 7.75-fold enhancement compared to pure PMMA for the PMMA/20% Ta_2O_5 composite at 81 keV gamma energy. Furthermore, the PMMA/20% Ta_2O_5 composite exhibited the lowest HVL and the highest Z_{eff} value among the composites. From this perspective, the

PMMA/20% Ta_2O_5 composite can be considered a flexible, lightweight, and environmentally friendly shielding material. Nevertheless, further studies could be conducted on additional properties of this material, such as its mechanical and thermal resistance. Furthermore, investigating its efficacy for various radiation energies and types would facilitate the multi-purpose utilization of the material.

Author Contributions

The author read and approved the final version of the paper.

Conflicts of Interest

The author declares no conflict of interest.

Ethical Review and Approval

No approval from the Board of Ethics is required.

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