

A Monte Carlo Study on the Shielding Properties of a Novel Polyvinyl Alcohol (PVA)/WO₃ Composite, Against Gamma Rays, Using the MCNPX Code

Kazemi F.¹, Malekie S.², Hosseini M. A.^{3*}

ABSTRACT

Background: In recent years, there has been an increased interest toward non-lead radiation shields consisting of small-sized filler particles doped into polymer matrices. In this paper, we study a new polyvinyl alcohol (PVA)/WO₃ composite in the presence of high-energy gamma photons through simulation via the Monte Carlo N-Particle (MCNP) simulation code.

Material and Methods: An MCNP geometry was first designed in the software based on real-life conditions, and the generated geometry was validated by calculating the mass attenuation coefficient and making relative comparisons with standard tables. Using the lattice card in the MCNP input file, WO₃ was considered as a filler dispersed in a PVA polymer at sizes of 10 μm and 30 nm with a weight concentration of 50 wt%. By defining 10⁶-photons emitted from point sources corresponding to 662, 778, 964, 1112, 1170, 1130 and 1407 keV energy levels, and the F4 tally used to estimate the cell average flux, the values for mass attenuation coefficient and half-value layer (HVL) were calculated.

Results: The results show that PVA/WO₃ composite can be considered to shield X and γ-rays in the mentioned energies. However, nano-WO₃ has a better ability to shield in comparison with the micro-WO₃ fillers. The differences in attenuation changed at different energy levels, ascribed to the dominance of pair production occurrence and photon interactions in the composite, which was in good agreement with previous studies.

Conclusion: Our finding showed that the composite can be considered as a lead-free shielding material.

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Keywords

Radiation Protection • Monte Carlo Method • Gamma Rays • Polyvinyl Alcohol (PVA) • Tungsten Trioxide (WO₃) • MCNP Code

Introduction

Humans have long been aware of ionizing radiation benefits as well as the dangers of being exposed to it. In medical applications, it has been several years that X-rays and high-energy gamma rays are being utilized in radiotherapy to control and treat cancerous tumors. Nevertheless, the increasing need to protect non-target tissues and the organs at risk or near cancerous tissues has led to the improvement of radiotherapy techniques [1,2]. As a solution, high-Z mate-

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rials have been utilized to fabricate traditional blocks and multileaf collimators (MLCs) in order to shape the beams better, and to reduce the absorbed dose to normal tissues. So far, lead (Pb) has been the most commonly used material due to its high atomic number (Z), low-cost and abundance [2,3]. Furthermore, lead has been extensively used as raw material in manufacturing of fixed and movable shields in diagnostic radiology, as well as nuclear medicine [2]. Nonetheless, there have been serious challenges regarding its usage, including low mechanical strength, production of secondary radiation resulting from the interaction with beams, toxicity, and heavy weight [2,4]. In addition, a study reported the risk of remaining nosocomial pathogens in the lead aprons [5]. Therefore, in recent years there has been increased demand to reduce the use of lead in shielding material, or even to replace it with a new substance [2,6].

Meanwhile, various studies have shown that composites containing micro- and nanoparticles of high-Z fillers dispersed in a conformable polymer matrix can be used to design high-energy radiation shields as an alternative to traditional shielding material [2,7]. polymeric matrix with proper filler have shown to have unique properties, due to large surface-area-to-volume ratio of fillers in the polymer matrix, and it is expected that the overall weight of a shield will decrease while its absorption/attenuation abilities improves. Furthermore, this type of material will have higher conformability and lower toxicity comparing to lead [6,8]. In this regard, extensive research has been carried out on the use of metal oxides, such as tungsten trioxide (WO_3) as a filler in different polymer matrices, and acceptable results were obtained when applied to gamma and X-ray radiation shields [8-13].

The use of composites, such as PVA/ WO_3 as an alternatives to lead is of interest from two different aspects:

Since PVA has specific properties, such as nontoxicity and non-carcinogenicity, it is be-

ing used in food production and packaging industries after its approval by the US Food and Drug Administration (FDA). It has reached a point that PVA is widely recognized as a safe substance in the manufacturing of medical equipment [14,15]. We can also point to some other interesting chemical and physical properties of PVA, such as solubility in water, crystallization in low temperatures, high thermal resistance, high mechanical tensile strength and high biocompatibility [14].

In addition, tungsten is known as a high-Z metal with a lower density than lead that ultimately produces less secondary radiation. Moreover, in micro/nano sizes, tungsten can result in reduced weight due to its desirable dispersion in the matrix (increased surface-to-volume ratio) [2]. Furthermore, several studies have stated that metallic fillers such as WO_3 embedded in the PVA polymer can enhance physical properties like thermal, mechanical, optical, and electrical in comparison to its macroscales. [16-22].

Considering the stated properties of PVA/ WO_3 , our aim was to study the absorption and attenuation properties of this composite in high gamma radiation energy (^{60}Co , ^{137}Cs , and ^{152}Eu point sources) through a Monte Carlo simulation code. To the best of our knowledge, this is the first study on PVA/ WO_3 properties as a radiation shielding material in the stated energy spectrums.

Material and Methods

The present study deals with a PVA/ WO_3 composite consisting of polyvinyl alcohol (PVA) with the chemical formula $\text{C}_2\text{H}_4\text{O}$ and the density of 1.19 g/cm^3 as the matrix, and tungsten oxide with the density of 7.16 g/cm^3 as the polymer filler in 30 nm and 10 μm sizes.

Similar studies showed that the WO_3 filler with a weight percentage of 50 wt% had an interesting performance in attenuation/absorption of high-energy gamma photons [8,11]. Therefore, similar weight percentage was selected (including 50% PVA and 50% WO_3

with 2.04 g/cm³ density), and the Monte Carlo N-particle Transport Code (MCNPX 2.6.0) was used to carry out the simulation, which will be explained as follow.

In this research, a narrow beam geometry was designed in MCNPX, presented in Figure 1, which represents the real geometry in the Secondary Standard Dosimetry Laboratory (SSDL) located in Karaj, Iran.

As can be seen, two collimators were used; the primary collimator was a lead cylinder 30 cm in diameter, with a flat bottom base and an inner diameter of 6.1 cm, placed 40 cm from a point source. The second collimator was placed behind the first one, this one being a lead cylinder with two cavities with circular cross-sections, one on the surface of the top base, 11 cm in diameter and the other one on the side, 5.6 cm in diameter. The incident particles were detected via a NaI detector with 3×3 inch dimensions placed inside the cavity (see Figure 1). Also, the F4 tally type was utilized in the code. This tally allows us to estimate the averaged flux over a cell [23], which represents the particles interaction rate per unit volume (1 cm³).

The radiation source defined in the code was

a point source that released 10⁶ photon gamma rays at photon energies of 662, 778, 964, 1112, 1170, 1130, and 1407 keV [8,11]. To validate the output results in the mentioned geometry, a pure aluminum with 2 mm thickness was defined in the geometry as a target, and its mass attenuation coefficient was calculated through the code at 662 keV photon energy. At the end, the obtained values were compared with standard tables. Note that the relative error reported after each execution did not exceed 0.05% in any of the cases.

As shown in Figure 1, the attenuation and absorption properties of PVA/WO₃ composite were studied using a cube 8×8 cm and 0.2 cm thickness placed at a distance of 60 cm. Using the lattice card, WO₃ particles were dispersed homogeneously in the PVA polymer matrix as 10 μm and 30 nm spherical particles. For better clarification, screenshots of the simulated composite in both micro and nano filler sizes are shown in Figure 2.

Consider I and I_0 as the program outputs in the presence and absence of a sample, and x denote the sample thickness (0.2 cm in here), thus the linear attenuation coefficient (μ) can be expressed based on the Lambert-Beer law

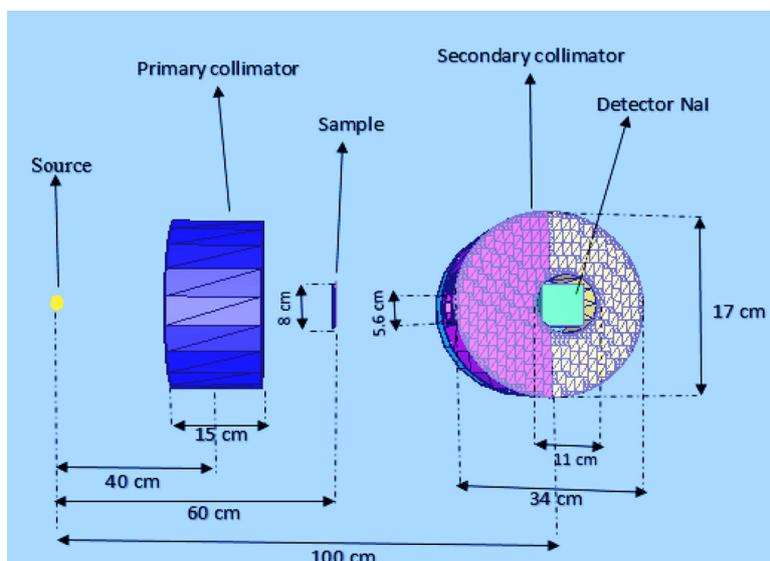


Figure 1: Layout of the radiation system and geometry in the simulation code including a point source, the first and secondary collimators, and a NaI detector to calculate incidence particle flux.

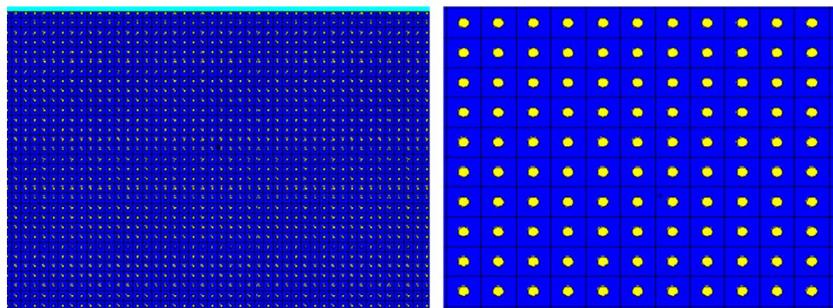


Figure 2: Images obtained from lattice creation based on a sample of 50 wt% PVA/WO₃ composite; the images show the even dispersion of tungsten trioxide particles in the polyvinyl alcohol matrix; the WO₃ particles were in the form of spheres with radii of 30 nm (left) and 10 μm (right).

[24] as follow:

$$\mu = \frac{\text{Ln}(I_0/I)}{x} \quad (1)$$

Note that the linear attenuation coefficient (in cm⁻¹) is dependent on the photon energy and the absorber material. Therefore, by dividing it by the sample's density (μ/ρ), we can define the mass attenuation coefficient independent of the used material. The half-value layer (HVL) is calculated through the following equation:

$$\text{HVL} = \frac{\text{Ln}(2)}{\mu} \quad (2)$$

Results

In this research, the attenuation and absorption properties of a PVA/WO₃ composite (50 wt% of each compound) against high-energy photons emitted from ⁶⁰Co, ¹³⁷Cs and ¹⁵²Er point sources were studied through simulation by MCNPX. Hence, a geometry was first generated based on a real-life condition, and the mass attenuation coefficient of pure aluminum was calculated through the program output as 0.0781 cm²/g, which is in good agreement with NIST tables (0.0782 cm²/g) [25]. This minor difference indicates the validity of geometry used in our simulation. Table 1 provides the elemental mass fractions and the density of the composite under study.

Table 2 presents the calculated mass attenuation coefficients for the micro- and nano-composites, along with the mass attenuation increase rate ($\Delta = (\mu/\rho_{\text{nano}} - \mu/\rho_{\text{micro}}) / \mu/\rho_{\text{micro}}$). Furthermore, Figure 3 shows the changing

Table 1: Elemental mass fractions of 50 wt% PVA/WO₃ ($d=2.04$ g/cm³).

Element	Concentration
H	0.08
C	0.227
O	0.2
W	0.3

Table 2: Calculated mass attenuation coefficients and their improvement rates (Δ) for PVA/WO₃.

Energy (keV)	$\mu/\rho(\text{cm}^2/\text{g})$		$\Delta(\%)$
	nano	micro	
662	0.0599	0.0532	12.6
778	0.0550	0.0487	13.0
964	0.0489	0.0436	12.1
1112	0.0443	0.0412	7.6
1170	0.0432	0.0401	7.7
1330	0.0408	0.0368	11.0
1407	0.0391	0.0349	12.0

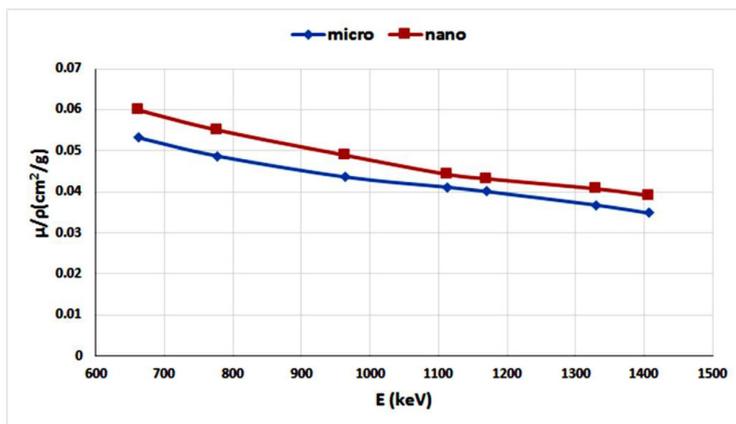


Figure 3: Diagram shows the changes in mass attenuation coefficients of the micro- and nano-composites under study based on energy level.

trend in mass attenuation coefficient of the sample based on 662, 778, 964, 1112, 1170, 1130 and 1407 keV photon energies for different WO_3 particle sizes. Results indicate that increasing energy will reduce mass attenuation coefficient in both composite samples. Overall, the coefficients are higher for nano-composite in comparison with the micro-composite, suggesting a better attenuation. A better shielding performance of nanocomposite can be attributed to higher surface-to-volume ratio and a better dispersion in the polymer matrix, owing to larger photon interaction cross-section that results in a higher mass attenuation coefficient. This is consistent with the results of previous studies on WO_3 doped into other polymer matrices [8,11].

However, the Δ values were different at different energy levels (7-13%), which could be due to changes in the possibility of photon interactions with materials, such as the Photoelectric and Compton effects. Moreover, at higher energies (>1022 keV), this incident could be explained by the dominance of pair production occurrence [11]. Figure 4 illustrates the photon shielding performance of the composite under study expressed in terms of HVL. A description for the increased HVL values at higher energy levels could be related to the gamma ray increased energy, which is the necessary thickness to split the incident photons in half. This again shows the relative superiority of the nanocomposite in attenuation/absorption of photons. We must note that

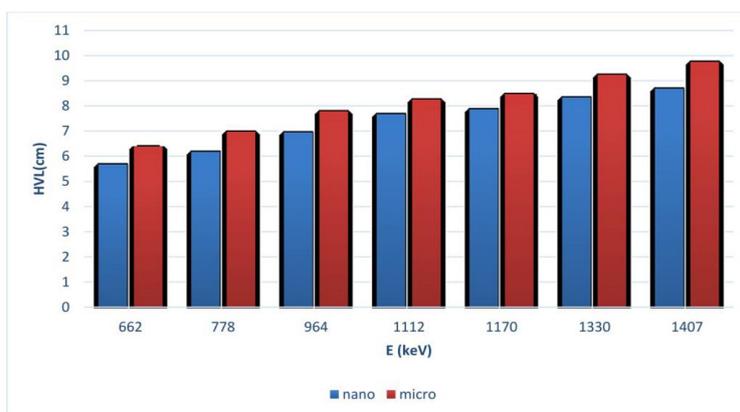


Figure 4: Calculated HVL values in different photon energies for the micro- and nanocomposite.

despite the different HVL values calculated at different energy levels, the reduction in the shielding thickness was almost constant at all energies (~10%).

In Figure 5 the mass attenuation coefficient of 50 wt% WO_3 /PVA composite is plotted over a wide range of energy using XCOM [26], in the energies near the K-edge of tungsten (70 keV), the amount of attenuation coefficient increased due to the photoelectric effect while for higher energies the amount of μ/ρ decreased gradually. For higher energies, Compton scattering and pair production are two dominant events.

Discussion

Considering the results of the present study, it seems necessary to express a few points with regards to the previous literature:

First, we must point to the WO_3 filler and its attenuation and absorption properties against X-ray and gamma photons in different polymer matrices. Recently, studies were conducted on WO_3 /Epoxy composites in the presence of low-energy X-ray sources through simulation and experiment, and the effects of WO_3 particle size was also investigated. In this research, in addition to reaching a good agreement between experimental and simulated data, the higher attenuation of the nanocomposite was reported [27,28]. However, in other studies at higher energy spectrum, the researchers had

only attempted to explore the absorption and attenuation properties of the relative composites via simulation. For instance, one study dealt with a 50-wt% WO_3 /E44 Epoxy composite by simulation at a wide range of gamma energies, and reported the relative superiority of the shielding properties of nanoparticles in comparison with micro-particles, even in high energy spectrums [8]. Furthermore, it has been shown that after doping concrete with micro- and nanoparticles of WO_3 , more desirable absorption properties were observed in comparison to pure concrete, and those properties are even much better with nanoparticles [10]. Therefore, in line with the mentioned studies, a 50-wt% PVA/ WO_3 composite was able to show a good attenuation/absorption in the current research, and the size of filler particles influenced the composite's shielding properties. In an experimental research on WO_3 /Epoxy against high-energy sources, tungsten trioxide micro-particles were tested via practical experiments with ^{60}Co irradiation. In the mentioned study, researchers emphasized on the necessity to preserve the mechanical and thermal properties of the composite, while increasing the weight percentage while showing that a 50% weight concentration (similar to the current research) would maintain the mechanical and thermal properties to a great degree in addition to being radiation absorbent [7].

These results compel us to conduct further

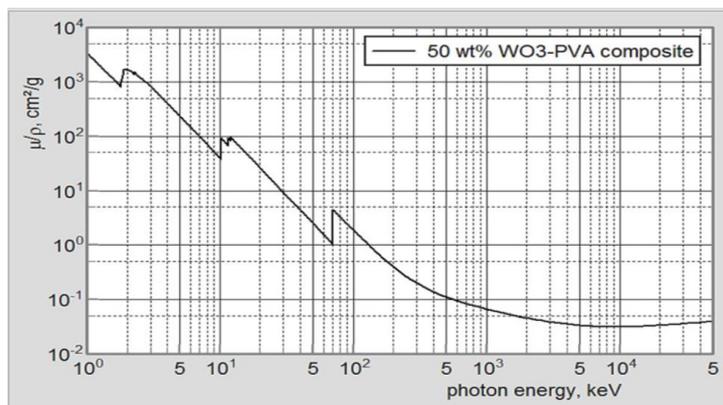


Figure 5: Mass attenuation coefficient of 50 wt% WO_3 /PVA composite in different energies using XCOM.

experimental research by considering all the mechanical, thermal and radiation shielding properties of the PVA/WO₃ composite, to make relative comparisons with the findings of our simulation.

Additionally, our search in the literature showed that despite the unique properties of PVA, such as low toxicity and high environmental compatibility, it has not been thoroughly studied as a polymer matrix in lead-free radiation shields [21]. Recent studies have produced acceptable results with PVA nanocomposites thin films doped with various nano-sized metal fillers in relatively low weight concentrations (<10 wt%) [19,20,22]. Nonetheless, it seems necessary to study the shielding properties of such composites in the presence of low-energy photons. As far as we know, this is the first study that has dealt with the PVA/WO₃ composite as a shielding material for high-energy gamma sources; hence, the authors recommend further research on this subject.

Conclusion

In this study, the attenuation/absorption properties of a new PVA/WO₃ composite were investigated in the most commonly-used photon spectrums corresponding to ⁶⁰Co, ¹³⁷Cs and ¹⁵²Er point sources through simulation via the MCNPX code. At first, the validity of the designed geometry was confirmed. Then, doping PVA with micro- and nano-sized WO₃ fillers with a 50-wt% weight concentration and calculation of important quantities, such as the mass attenuation coefficient and HVL yielded a promising results. Moreover, the effect of particle size was also assessed on the shielding properties of the composite under study, and it was revealed that nano-sized particles cause more improvement than micro-particles at all energies. Overall, good level of agreement was obtained with previous studies.

Despite special properties of PVA such as extremely low toxicity, high environmental compatibility and certain useful applications as well as the unique properties of WO₃ in at-

tenuation of high-frequency photons, it seems that research in this area is still in its primary stages; hence further studies are warranted.

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Conflict of Interest

None

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