

Investigation of The Gamma Ray Shielding Behaviour of (90-x)TeO₂—xMoO₃—10ZnO Glass System Using Geant4 Simulation Code and WinXCOM Database

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Abstract. In this study, mass attenuation coefficients (μm), transmission fractions (T), mean free path (MFP) and half value layer (HVL) of the (90-x)TeO₂—xMoO₃—10ZnO glass system ($x=10, 20$ and 30 mol%) were calculated at the gamma ray photon energies of 356 keV, 662 keV, 1173 keV and 1330 keV. Shielding parameters of the glass system were calculated from the Monte Carlo simulations carried out with the Geant4 model of a high purity germanium (HPGe) detector and from the WinXCOM database software. The used Geant4 model was validated by comparing the experimentally determined mass attenuation values of the PbO—BaO—B₂O₃ glass system to the values calculated with Geant4. The findings of the present study reveals that the glass system containing 10% mol MoO₃ exhibits better photon shielding performance compared to the glass with 30% mol of MoO₃ due to the higher amount of TeO₂ existed in its chemical formula.

Keywords: Geant4, Simulation, Glass, Photon attenuation.

(90-x)TeO₂—xMoO₃—10ZnO Cam Sisteminin Gama Işını Zırhlama Özelliklerinin Geant4 Simülasyon Kodu ve WinXCOM Veri Tabanı Kullanılarak Araştırılması

Özet. Bu çalışmada 90-x)TeO₂—xMoO₃—10ZnO cam sisteminin ($x=10, 20$ and 30 %mol) kütle zayıflatma katsayıları (μm), geçirgenlik oranları (T), ortalama serbest yol (MFP) ve yarı değer (HVL) parametreleri 356 keV, 662 keV, 1173 keV and 1330 keV gama foton enerjilerinde hesaplanmıştır. Zırhlama parametrelerini yüksek saflıkta germanium (HPGe) dedektörünün Geant4 modeli ile gerçekleştirilen Monte Carlo simülasyonları ve WinXCOM veri tabanı kullanılarak hesaplanmıştır. Geant4 modelinin geçerliliği PbO—BaO—B₂O₃ cam sistemi için deneyel olara bulunan kütle zayıflatma katsayıları Geant4 ile elde edilen sonuçlarla karşılaştırılarak sağlanmıştır. Bu çalışmanın sonuçları kimyasal formülünde yüksek oranda TeO₂ bulunmasından dolayı %10 mol MoO₃ içeren cam sisteminin radyasyon zırhlama özelliklerinin %30 mol MoO₃ cama göre daha iyi olduğunu göstermiştir.

Anahtar Kelimeler: Geant4, Simülasyon, Cam, Foton zayıflatma..

1. INTRODUCTION

Investigation of the radiation shielding properties of materials is an increasing interest for researchers due to the wide use of radioactive isotopes in diverse fields, such as nuclear power plants,

radiology and radiotherapy departments of hospitals, nuclear research and accelerator centers, agriculture, oil plants and archeology. Therefore, determination and knowledge of the radiation shielding properties of the materials used in these

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kind of places are of crucial importance to protect humans against the hazardous effects of the ionizing radiation. Gamma rays are one of the more penetrating radiation type and materials with high density and high atomic number are needed for their effective shielding. Lead is known as the best gamma ray attenuating material. Whereas, weight, price and toxicity of lead make it unpractical and uneconomical for the shielding of large areas. Recently, radiation shielding properties of different natural or synthetically produced materials, such as concrete, sand, cement, bricks, tiles, glasses, clay materials, polymers and alloys has been studied by different researchers [1,8].

Having the advantages of optical transparency and easy preparation and production make the glasses attractive for the researchers to investigate and explore their shielding properties. Numerous studies reported that glasses are promising materials when used as gamma attenuator [9,11]. Because of this, synthesis of new glass systems have been made by several groups in order to improve their shielding potential. The gamma ray attenuation characteristics of the newly synthesized glasses have been determined either by using the experimental techniques or by using the Monte Carlo simulation codes.

Six glass pieces with composition of PbO-Li₂O-B₂O₃ were prepared by using the melt quenching technique and the shielding properties of the samples were experimentally determined at four different gamma ray energies. The results of the study showed that the increase of PbO content increases the shielding efficiency [12]. The effect of La₂O₃ on the radiation attenuation characteristics of the lanthanum calcium

silicoborate glasses is experimentally measured at eight different photon energies between 224 keV and 662 keV. It was reported that the increase of the La₂O₃ concentration results in an increase the shielding ability of the lanthanum calcium silicoborate glasses [13]. Five different samples of Bi₂O₃-BaO-B₂O₃-Na₂O quaternary glass system were produced and their structural and gamma ray shielding features were investigated experimentally by Dogra et al. [14]. It was reported that the samples have higher mass attenuation and lower half value layer compared to barite concrete at 662 keV. In addition to the experimental techniques, Monte Carlo simulations can be employed to study the radiation interaction parameters of the materials especially in the absence of the expensive experimental equipments or in the absence of the fabricated samples. Modelling of an experimental set-up consisting of a detector, a gamma ray source and an attenuator and performing the measurements in the computer environment provide an ease of use and save time. Shielding properties of 80TeO₂-5TiO₂-(15-x)WO₃-xAn_{Om} glasses were investigated by using MCNP5 code and WinXCOM. The results of this study revealed that using of 5Er₂O₃ in the places of xAn_{Om} significantly increases gamma ray and neutron shielding behavior [15]. Gamma and neutron shielding capabilities of 20BaO/SrO-(x)Bi₂O₃-(80-x)B₂O₃, where x value was changed between 10 and 60 mol, glasses has been investigated computationally by using the MCNP5 code and WinXCOM. It was published that the addition of Bi₂O₃ increases the shielding effectiveness of the glass system [16].

In this study, the complete geometry of a high purity germanium (HPGe) spectrometer produced by Canberra semiconductors was coded into Geant4 in order to determine the mass attenuation coefficients (μ_m), transmission fractions (T), half value layer (HVL) and mean free path (MFP) of the

(90-x)TeO₂—xMoO₃—10ZnO glass system from the Monte Carlo simulations. In addition to the Monte Carlo simulations, μ_m values of the selected glass system have been also determined using the WinXCOM database and compared to the results from Geant4.

2. MATERIALS AND METHODS

2.1. Theoretical Background

Mass attenuation coefficient is a substantial parameter defining the penetration and diffusion of gamma ray photons within the material. Beer-Lambert's law as given by equation (1) was used to calculate the mass attenuation coefficients of the selected glass system.

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

Where I and I_0 are the transmitted and incident photon intensities respectively, x (cm) is the thickness of the material and μ (cm^{-1}) is the linear attenuation coefficient of the sample. In order to measure the incident photon intensity, Monte Carlo simulations were performed at each selected energy without the sample placed between the source and the detector. Simulations were then repeated with the sample placed between the source and the detector to obtain the attenuated photon intensities. The logarithm of transmission [$\ln(I/I_0)$] is plotted versus sample thickness and the slope of linear fit to these data points results in linear attenuation coefficient. The mass attenuation coefficient (μ_m) is widely used for the calculation of photon transmission and energy deposition within a material. μ_m of any material is then calculated by dividing the linear attenuation coefficient with the density of sample:

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

The transmission fraction, T, is the fraction of the transmitted photon intensity (I) to the incident photon intensity (I_0). Plotting of T versus x can be used to estimate the decrease in the photon intensity with respect to the material thickness. This parameter is calculated as:

$$T = I/I_0 \quad (3)$$

The half-value layer (HVL) is the thickness of a material needed to reduce the intensity of the incident radiation by one-half. Its dependence to the linear attenuation coefficient, μ , is defined by the Eq. (4)

$$HVL = \ln(2)/\mu \quad (4)$$

The mean free path (MFP) defined as the average distance travelled by a photon between two successive interactions is equal to the reciprocal of the linear attenuation coefficient as given in next equation:

$$MFP = 1/\mu \quad (5)$$

2.2. Geant4 Model of HPGe Detector

Geant4 is a Monte Carlo simulation toolkit based on the object oriented C++ programming and used for the modeling of the detectors for the high energy, nuclear, accelerator, medical and low

energy physics applications. The Geant4 kernel manages runs, events and tracking of passage of particle through matter. It allows users to simulate all aspects of an experimental set up such as geometry of detector, primary particle generation of events, types of particles and physics processes such as electromagnetic, hadronic and decay physics that manages particle interactions [17].

In this work, the simulated detector geometry is based on the HPGe detector produced by the Canberra semiconductors. The core of the detector consists of a 2.2 kg HPGe crystal installed as an ionization detector. The crystal is 82 mm in diameter and 81.5 mm in height. The core cavity of crystal is 10.5 mm in diameter and 62.5 mm in height. The detector has a 1 mm thick copper endcap window placed 6.3 mm from the top of germanium crystal. The thickness of the crystal dead layer is 0.8 mm. Fig. 1 shows the Geant4 model of the HPGe detector geometry.

As shown in Fig. 2, the detector and the gamma ray source were surrounded with lead shields. A cavity with 3 mm in diameter and 50 mm in height is defined at the center of the both shields to create a narrow beam in order both to ensure that the Beer-Lambert's law is valid and to minimize the variance and systematic errors. The chemical composition in mol% and weight fractions of elemental composition along with the densities of $(90-x)\text{TeO}_2-x\text{MoO}_3-10\text{ZnO}$ (shortly referred as TMZ) glass system has been coded into Geant4 according to the values as given in Table 1 [18]. During the simulations, the thickness of the glass sample was varied from 1 cm up to 5 cm with 1 cm increment and simulations were repeated for the new thickness. The glass sample was placed between the source and the detector as an attenuator and the mass attenuation values of the TMZ glass systems have been reported at the gamma ray energies of 356 keV (^{133}Ba), 662 keV (^{137}Cs), 1173 keV (^{60}Co) and 1330 keV (^{60}Co).

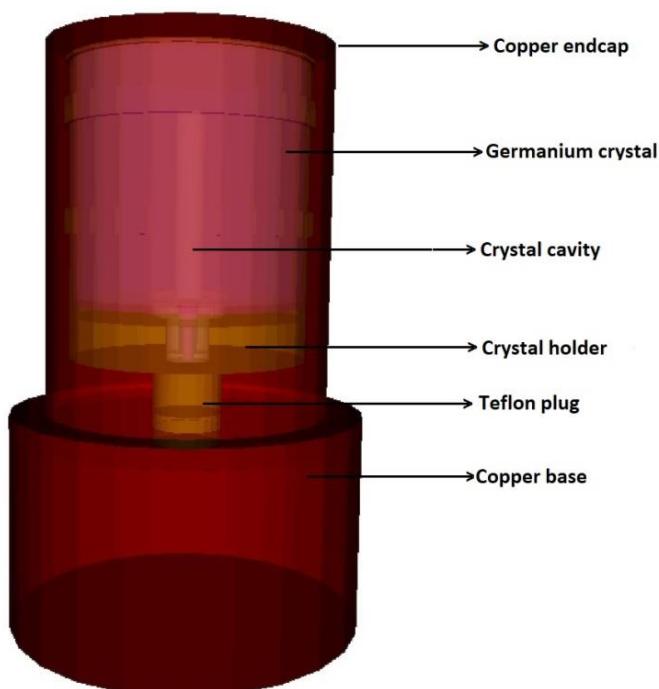


Figure 1. Geant4 model of the HPGe detector.

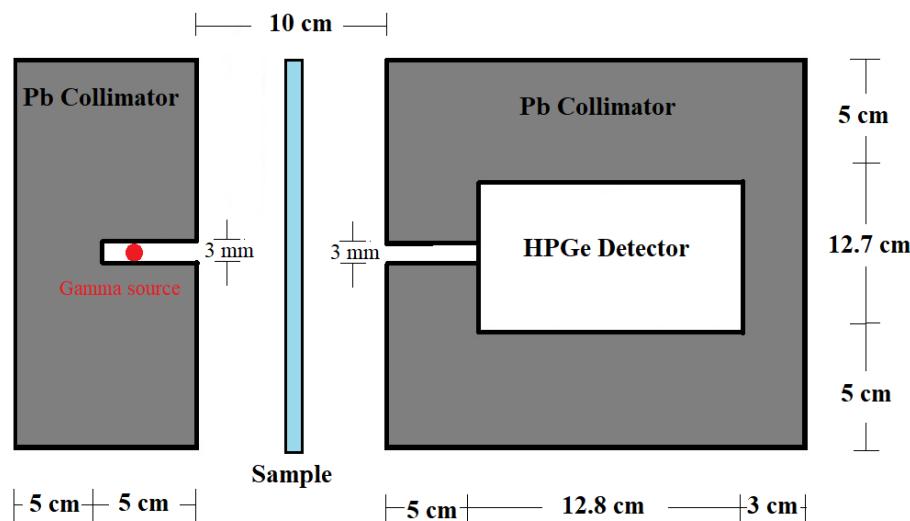


Figure 2. Schematic of the Geant4 set-up used to perform Monte Carlo simulations

Table 1. The chemical composition and density of (90-x)TeO₂—xMoO₃—10ZnO glass system

Code	Glass sample in mol (%)	Mass fraction (%)				Density (gr/cm ³)
		Te	O	Mo	Zn	
TMZ10	80TeO ₂ —10MoO ₃ —10ZnO	67.9	21.3	6.45	4.35	5.103
TMZ20	70TeO ₂ —20MoO ₃ —10ZnO	60.0	22.6	13.0	4.4	5.066
TMZ30	60TeO ₂ —30MoO ₃ —10ZnO	52.0	24.0	19.56	4.44	4.976

2.3. WinXCOM Database Software

WinXCOM database can also be used to calculate μ_m values of any material and this software does not require any experimental setup [19]. Calculation of the μ_m values is based on the mixture rule given by the following equation.

$$(\mu_m)_{glass} = \sum_i^n w_i (\mu_m)_i \quad (6)$$

Where w_i and $(\mu_m)_i$ are the weight fraction and mass attenuation coefficient of the i^{th} element respectively.

3. RESULTS AND DISCUSSION

3.1. Validation of the Geant4 model

Prior starting the Monte Carlo simulations for the (90-x)TeO₂—xMoO₃—10ZnO glass system, the validity of the Geant4 model was proven by comparing the experimentally obtained mass attenuation values of 20MoO₃-xBi₂O₃-(80-x)B₂O₃ glasses ($x = 10, 20, 30, 40$ mol%) [20] to the Geant4 based simulation results. The findings from the both studies are shown in Table 2. The relative difference of the experimental and Geant4 results vary between 0.7% - 12.4%. From the validation of the Geant4 model, we are confident about the reliability of the values determined as the shielding parameters of (90-x)TeO₂—xMoO₃—10ZnO glasses and any other glass system which has not been investigated previously.

Table 2. Mass attenuation coefficients of $\text{MoO}_3\text{-Bi}_2\text{O}_3\text{-B}_2\text{O}_3$ (MoBiB) glasses calculated with Geant4 and comparison to experimental data [20] for the validation of the Geant4 model.

Glass sample	356 keV		662 keV		1173 keV		1330 keV	
	Geant4	Exp.	Geant4	Exp.	Geant4	Exp.	Geant4	Exp.
MoBiB10	0.2402 ± 0.0092	0.2136 ± 0.0128	0.1029 ± 0.0052	0.0977 ± 0.0048	0.0657 ± 0.0033	0.0594 ± 0.0071	0.0512 ± 0.0053	0.0549 ± 0.0028
MoBiB20	0.2269 ± 0.0098	0.2201 ± 0.00134	0.1055 ± 0.0046	0.0986 ± 0.0059	0.0607 ± 0.0050	0.0603 ± 0.0029	0.0518 ± 0.0050	0.0554 ± 0.0029
MoBiB30	0.2354 ± 0.012	0.2234 ± 0.0136	0.1078 ± 0.0050	0.0996 ± 0.050	0.0599 ± 0.0046	0.0605 ± 0.0029	0.0581 ± 0.0044	0.0555 ± 0.0029
MoBiB40	0.2434 ± 0.0080	0.2302 ± 0.0138	0.0973 ± 0.0046	0.1011 ± 0.051	0.0579 ± 0.0039	0.0610 ± 0.0030	0.0513 ± 0.0042	0.0556 ± 0.0029

3.2. Photon Shielding Behavior of $\text{TeO}_2\text{-MoO}_3\text{-ZnO}$ Glass System

The Geant4 and WinXCOM results of μ_m values for the $(90-x)\text{TeO}_2\text{-}x\text{MoO}_3\text{-}10\text{ZnO}$ glass ternary ($x = 10, 20, 30$ mol%) are given in Table 3. The relative differences (RD%) between the results from the both toolkits were calculated by using the following formula;

$$\text{RD} (\%) = \frac{R_G - R_W}{R_W} \times 100 \quad (7)$$

where R_G and R_W is the results from Geant4 and WinXCOM respectively.

As can be seen from the results presented in Table 3, the relative difference (RD) varies between 0.8% and 4.1% and the results from Geant4 and WinXCOM are in agreement. Furthermore, it is observed that μ_m decreases with the increasing photon energy. This trend in μ_m is expected since the high energy photons have more penetration and diffusion ability within materials. Furthermore, in order to estimate the mass attenuation values at the photon energies which were not simulated, a four parameter (p_0, p_1, p_2, p_3) exponential fit in the form of $\mu_m = p_0 + p_1 \cdot \exp(p_2 + p_3 \cdot E)$ has been applied to the mass attenuation data calculated for all TMZ glass samples to obtain an equation to reproduce the mass attenuation coefficients at any desired energy within the range between 356 and 1330 keV. As shown in Table 4, the mass attenuation values calculated from the best fitted equations at the photon energies of 427.9 keV (^{125}Sb), (^{134}Cs), 834,8 keV (^{54}Mn), 1274.6 keV (^{22}Na) are in agreement with the results calculated using Geant4. It can therefore be concluded that these equations are reliable to calculate the mass attenuation of TMZ glass samples at any desired energy between 356 and 1330 keV.

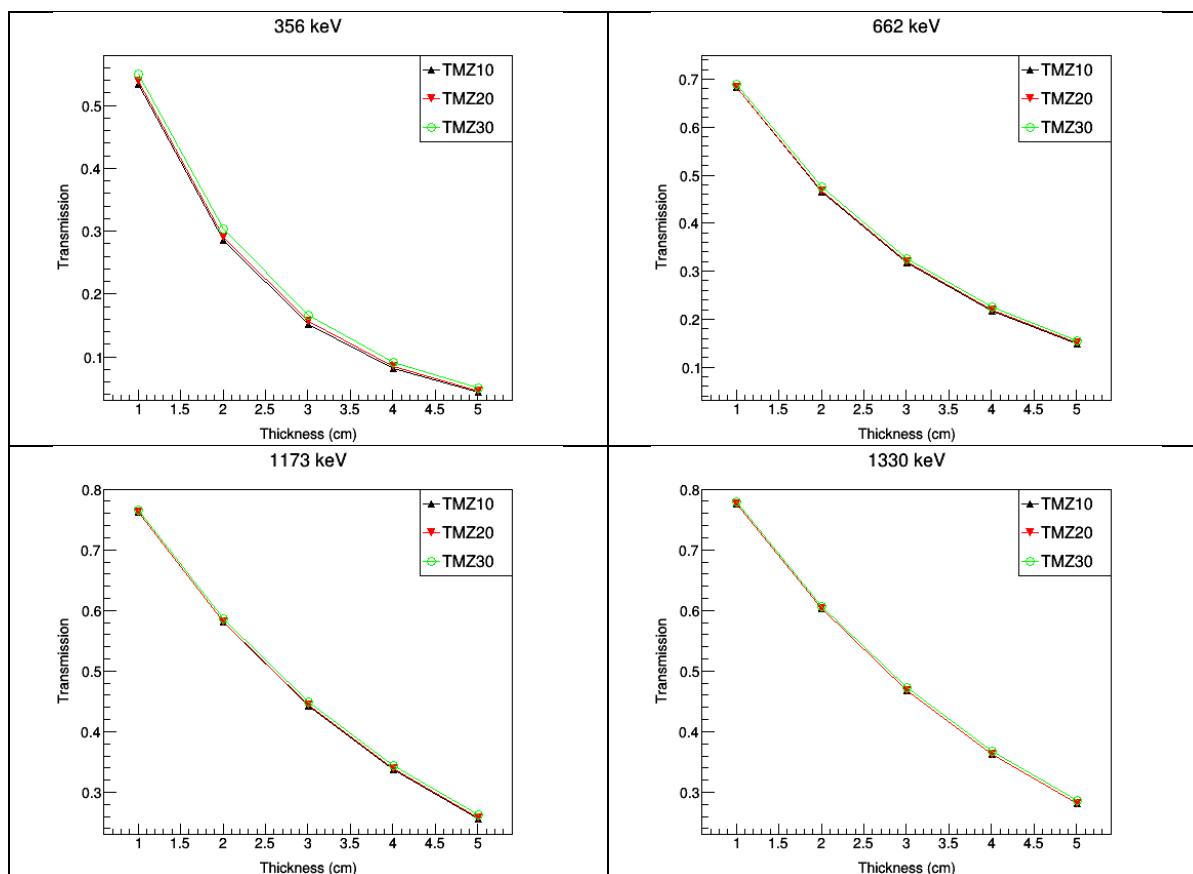
Table 3: Comparison of mass attenuation coefficients (cm^2/g) of the $\text{TeO}_2\text{-MoO}_3\text{-ZnO}$ (TMZ) glass ternary obtained using Geant4 simulations and WinXCOM

Glass Sample	356 keV		662 keV		1173 keV		1330 keV		RD			
	Geant4	XCO M	%	Geant 4	XCO M	%	Geant 4	XCO M	%			
TMZ10	0.120 ± 0.003	0.123	2.4	0.0763 ± 0.0006	0.0750	1.7	0.0520 ± 0.0005	0.0533	2.4	0.0487 ± 0.0004	0.0497	2.0
TMZ20	0.123 ± 0.004	0.122	0.8	0.0744 ± 0.0004	0.0750	0.8	0.0524 ± 0.0006	0.0535	2.0	0.0491 ± 0.0005	0.0500	1.8
TMZ30	0.125 ± 0.003	0.120	4.1	0.0740 ± 0.0004	0.0749	1.2	0.0527 ± 0.0005	0.0537	1.8	0.0497 ± 0.0005	0.0502	1.0

Table 4. Best fitted equations for estimation of mass attenuation and comparison to Geant4 results.

Glass Sample	Best Fitted Equation	Energy (keV)	Mass Attenuation Coefficient (cm^2/g)		Percentage Difference
			Fitted Equation	Geant4	
TMZ10	$\mu_m = 0.0437 + 0.0939 \cdot \exp(0.777 - 0.00276 \cdot E)$	427.9 (^{125}SB)	0.106	0.110	3.6
TMZ20	$\mu_m = 0.0460 + 0.214 \cdot \exp(0.125 - 0.00324 \cdot E)$	834.8 (^{54}Mn)	0.0622	0.0628	0.9
TMZ30	$\mu_m = 0.0475 + 0.235 \cdot \exp(0.144 - 0.00353 \cdot E)$	1274.6 (^{22}Na)	0.0505	0.0499	1.2

Fig. 3 shows the plot of the transmission values of TMZ glasses with respect to the glass thickness at the photon energies of 356 keV, 662 keV, 1173 keV and 1330 keV. Again, due to the penetration abilities of the high energy photons, a big difference is observed between 356 keV and 1330 keV. Transmission increases with the increase of photon energy. At the photon energies of 356 and 1330 keV the transmission values of 5 cm thick TMZ10 glass are 0.0433 and 0.281 respectively. Fig. 3 also shows that, among the simulated glasses, the lowest transmissions for all photon energies occur for the TMZ10 sample and highest transmissions occur for the TMZ30 sample. This behaviour is due to the higher fraction of TeO_2 existed in TMZ10 glass and the atomic number of Te ($Z = 52$) is higher than the atomic number of Mo ($Z = 42$). Since the decrease in the Te content in TMZ30 is compensated by the increase in Mo atoms, the transmission values for all glasses are close to each other at the selected energies. Furthermore, for the photon energies starting from 1100 keV, pair production is the dominating photon interaction mechanism and above this energy interaction mechanism becomes almost independent of chemical structure.

**Figure 3.** Transmission factor versus the thickness of $\text{TeO}_2\text{—MoO}_3\text{—ZnO}$ (TMZ) glass system at 356, 662, 1173 and 1330 keV photon energies.

The two important parameters characterizing the shielding ability of the TMZ glass system are the mean free path (MFP) and the half value layer (HVL) of the glass system. It is important to mention here that the lesser the value of MFP and HVL of given material, the better the shielding properties it would possess and thanks to the lower value of MFP and HVL, better shielding performance can be obtained by using a thinner layer of the selected material. Fig. 5 and Fig. 6 show the plot of MFP and HVL values of the TeO_2 — MoO_3 — ZnO glass system versus the photon energies. As can be seen from these two plots, TMZ10 has the lowest and TMZ30 has the highest values of MFP and HVL at the given photon energies. Depending on the close atomic numbers of Te and Mo elements, the observed differences for the MFP and HVL values are around 5.3% at 356 keV and 1.6% at 1330 keV for the 10% and 30% MoO_3 concentrations respectively.

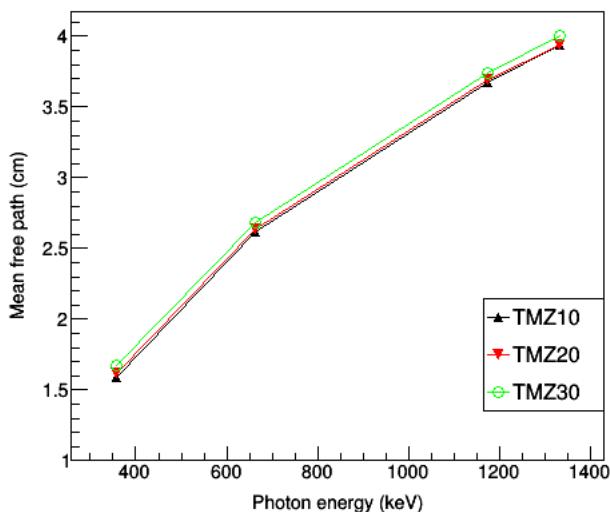


Figure 4. Mean free path of the TeO_2 — MoO_3 — ZnO glass system as a function of photon energy.

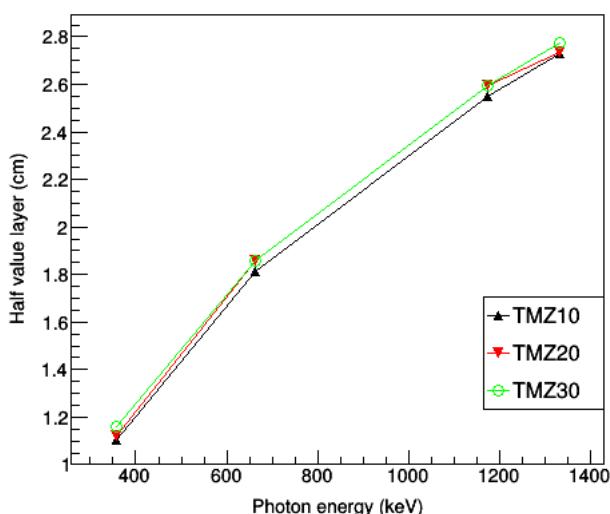


Figure 5. Half value layer of the TeO_2 — MoO_3 — ZnO glass system as a function of photon energy.

In order to see the dependence of the mass attenuation coefficient and half value layer on the fraction of MoO_3 existed in the selected glasses, these parameters were plotted versus the 10%, 20% and 30% mol of MoO_3 and the dependencies were shown in Fig. 6 and Fig. 7 respectively. For 10% and 30% mol of MoO_3 , the mass attenuation values change from 0.120 to 0.125 at 356 keV and from 0.0487 to 0.0497 at 1330 keV respectively. In the same way, the half value layers change from 1.104 to 1.162 at 356 keV and from 2.732 to 2.775 at 1330 keV. These findings also support that the decrease occurred for Te atoms are partially compensated by the increase in the Mo atoms in the glass structure. Therefore, mass attenuation values for the minimum and maximum MoO_3 concentrations do not show a significant change.

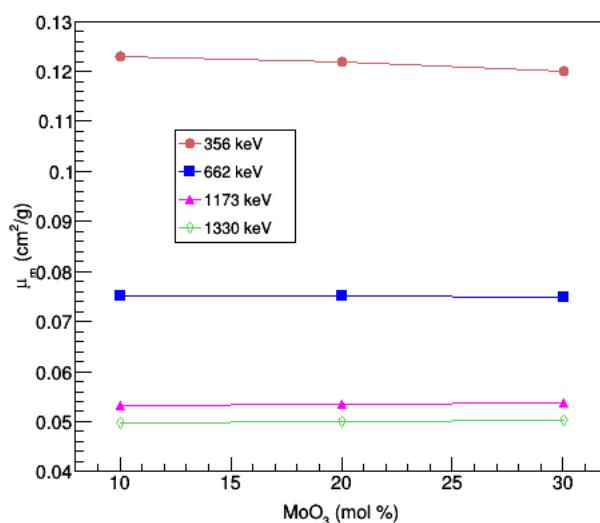


Figure 6. Dependence of mass attenuation values on the MoO_3 amount in the glasses

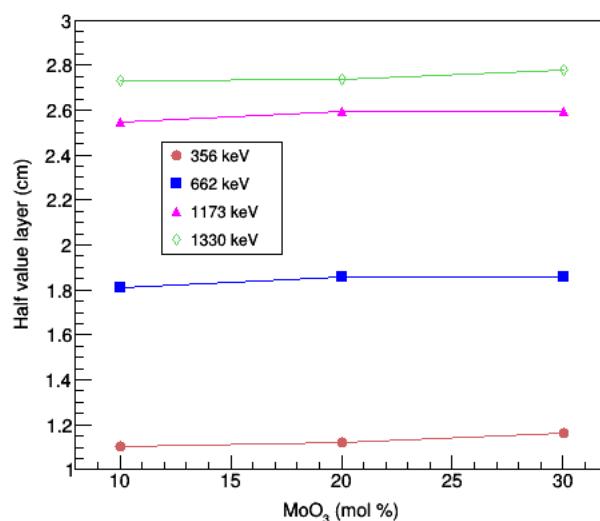


Figure 7. Dependence of half value layer on the MoO_3 amount in the glasses

4. CONCLUSION

In this study the photon shielding parameters of $(90-x)\text{TeO}_2-x\text{MoO}_3-10\text{ZnO}$ glass ternary ($x = 10, 20, 30 \text{ mol\%}$) was investigated computationally using Geant4 and WinXCOM database software to exploit the shielding potential of the selected glass system. Monte Carlo outputs of the Geant4 have been analyzed and the following conclusions were arrived;

- Geant4 model of HPGe detector can be a reliable alternative to the experimental techniques especially in the absence of equipments or material samples to determine the shielding capability of any other materials.
- Mass attenuation coefficients determined using Geant4 and WinXCOM agrees well. Mass attenuation values are high at lower energy region and they decrease as the photon energy increases and stay almost unchanged at the energy region above 1100 keV. This trend observed in mass attenuation coefficients can be explained by the photon interaction mechanisms at low, medium and high energy regions in terms of photoelectric effect, Compton scattering or pair production respectively.
- Thanks to the function obtained from the best fit applied to the simulated data, mass attenuation values at any desired energy can be calculated.
- Increase of the MoO_3 fraction from 10% to 30% in the glass structure causes an increase in the transmission fraction, half value layer and mean free path of the selected glass system and therefore weakens the shielding ability.

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