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MoO₃-TeO₂ glass system for gamma ray shielding applications

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PAPER

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M S Al-Buriah¹ , Halil Arslan² , H O Tekin³ , V P Singh⁴ and Baris T Tonguc¹¹ Department of Physics, Sakarya University, Sakarya, Turkey² Electrical and Electronics Engineering, Sakarya University of Applied Sciences, Sakarya, Turkey³ University of Sharjah, College of Health Sciences, Medical Diagnostic Imaging Department, Sharjah, United Arab Emirates⁴ Department of Physics, Karnatak University, Dharwad 580 003, IndiaE-mail: harslan@sakarya.edu.trKeywords: MoO₃-TeO₂ glasses, XCOM, Geant4, gamma shielding properties**Abstract**

This paper examines the gamma-ray shielding features of some selected tellurite-based glasses in the form of xMoO₃(100-x)TeO₂ (20 ≤ x ≤ 50 mol%). Mass attenuation coefficients (μ_m) of these glasses have been calculated using Geant4 toolkit and XCOM program for photon energy range of 1 keV—1000 MeV. The correlation factor (R^2) between these two methods was found to be almost one. Shielding effectiveness for these glasses has been estimated by computing half value layer (HVL), effective atomic number (Z_{eff}), and mean free paths (MFP). It was noticed that the heavy metal oxide of TeO₂ plays an important role in improving the shielding effectiveness of the glasses. The Mo20Te80 glass has shown the promising properties to serve for gamma ray protection applications as compared with many conventional concretes and other newly developed glasses.

1. Introduction

In the field of advanced materials, glasses are promising ones due to their numerous applications as well as their physical and chemical characterizations such as ease of designing, high corrosion resistance, optical transparency, and environment friendly. Previously, many opaque materials such as concrete were suggested as shielding materials for gamma radiation, however, there are several risks and limitations associated with their use [1, 2]. Currently, glasses have the largest interest as promising radiation shielding materials to be used in the windows and doors in medical diagnostic labs, x-ray rooms and CT scans, scintillators, radiation therapy chambers, and in space technology [3, 4]. This triggered an international interest to study many glasses doping with heavy metal oxide (HMO) to serve for various gamma ray shielding applications.

The TeO₂ glasses can be used as promising materials for many applications due to their unique structure, physical and optical features [5]. Also, these glasses have a wide range of transparency (400 nm—6 μm), low melting point nature and non-hygroscopic [6]. On the other hand, the significant use of x-ray and gamma-ray in many fields requires hard efforts to search for new materials which are economic, environment friendly, and proper protective against x-ray and gamma-ray. In this regards, several authors reported different type of glasses as promising shielding materials [7–11]. Sayyed [7] proposed some HMO glasses such as PbO, MgO, Ag₂O, Nb₂O₅, ZnO and BaO for gamma-ray applications. It was found that the glasses with PbO have the lowest values of MFP, whereas the glasses with MgO have the highest values of MFP. The results were compared with some concretes. Phosphate glasses were also studied and it was noted that the substitution of ZnO by PbO improves the gamma-ray shielding ability of the glasses [11]. However, the toxicity the lead (Pb) provide strong motivations to replace Pb element by another high-Z element such as Mo, Ba, W, Bi, Gd and etc. Therefore, hard efforts are being put by many authors to find suitable substitute glasses instead of lead to find equivalent shielding properties.

The gamma-ray shielding ability of the materials can be studied by using WinXcom program, experimental measurements or Monte Carlo simulation. These methods depend on Beer–Lambert law that can applied in the case of narrow beam geometry [12, 13]. However, the experimental is restricted to limited photon energies that can not be found easily or cheaply [14–16]. The simulation technique is proposed as a strong alternative to the

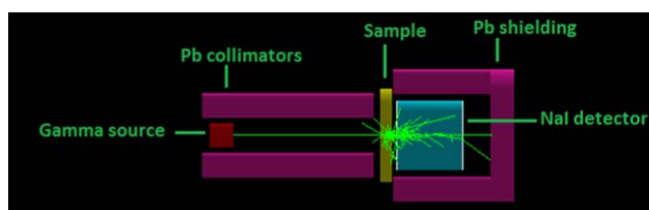


Figure 1. Geant4 simulation geometry to determine gamma shielding properties of the glasses

Table 1. Density, chemical composition and weight fractions of elements for the chosen glass samples.

Sample code	Density (g/cm ³)	Compositions (mol%)		Atomic composition (wt.%)		
		MoO ₃	TeO ₂	O	Mo	Te
Mo20Te80	5.251	20	80	0.225 0	0.122 6	0.652 4
Mo30Te70	5.176	30	70	0.237 6	0.185 8	0.576 6
Mo40Te60	5.018	40	60	0.250 4	0.250 3	0.499 3
Mo50Te50	4.858	50	50	0.263 5	0.316 1	0.420 4

experiment. This technique can handle all the difficulties of the experiment. Monte Carlo simulation is a widespread method to study the interaction of radiation (like gamma) with the matter by providing several models for electromagnetic interactions [17].

The objective of this work is to study the gamma-ray properties of newly developed tellurite-based glasses in the chemical form of $x\text{MoO}_3(100-x)\text{TeO}_2$ ($20 \leq x \leq 50$ mol%) [18]. All the important gamma-ray shielding parameters such as μ_m , HVL, MFP, and Z_{eff} of these glasses were obtained by using Geant4 toolkit and XCOM program for photon energy 1 keV—1000 MeV. The shielding competence of the present glasses was compared with those of conventional concrete, commercial glasses, and HMO glasses. The data in this work would be helpful to evolve new materials in the gamma-ray protection applications.

2. Materials and methods

Four tellurite-based glasses belonging to the system of $x\text{MoO}_3-(100-x)\text{TeO}_2$ with $x = 20, 30, 40,$ and 50 mol% have been reported to serve for gamma-ray protection applications. The selected glasses (taken from [18]) were coded as Mo20Te80, Mo30Te70, Mo40Te60 and Mo50Te50 for $x = 20, 30, 40,$ and 50 mol%, respectively. The conventional melt-quenching method was applied to prepare these glasses and Archimedes principle was used to measure their densities [19]. The sample code, density, and nominal composition of the glasses are shown in table 1.

2.1. Geant4 Monte Carlo simulation

GEANT4 is a radiation propagation Monte Carlo toolkit [17]. This toolkit can be used in numerous fields including high energy physics, astroparticle applications, medical applications, radiation shielding applications [20, 21]. For the radiation shielding studies, GEANT4 offers a wide range of physical models to handle all electromagnetic and nuclear interactions that may occur between the radiation and the shield material.

Figure 1 demonstrates the narrow beam geometry of the GEANT4 simulation, consisting of a point gamma source impinging on a slab of the glass. The gamma photon energies were defined in 662, 1173 and 1332keV. The thickness of the glass was ranging from 0.1—1 cm according to the gamma-ray energy. Two collimator, one is put after the gamma source and the other is kept before the detector, were also used to focus a beam and ensure that the gamma-rays travel parallel to the collimators. Also, the glass samples were modeled with respect to their atomic number, mass number, elemental weight fractions, and their densities. In this work, one million photons were gunned from monoenergetic source to hit a target of the glass sample. Then, the transmitted photons have been recorded by using sodium iodide (NaI) detector. The μ_m values of the studied glasses were then calculated

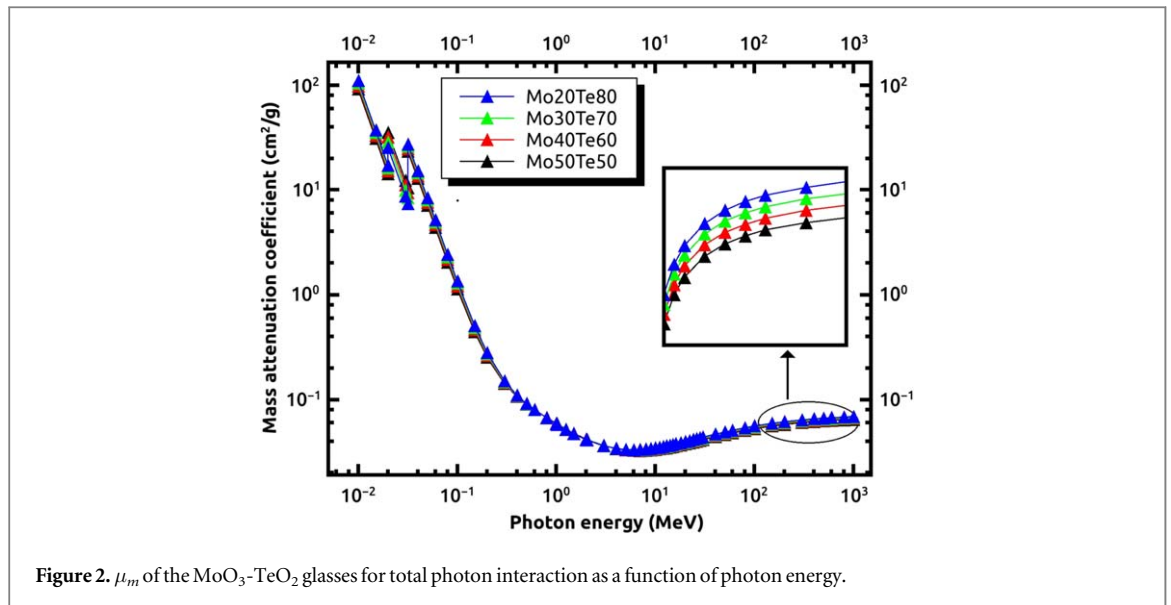


Figure 2. μ_m of the MoO₃-TeO₂ glasses for total photon interaction as a function of photon energy.

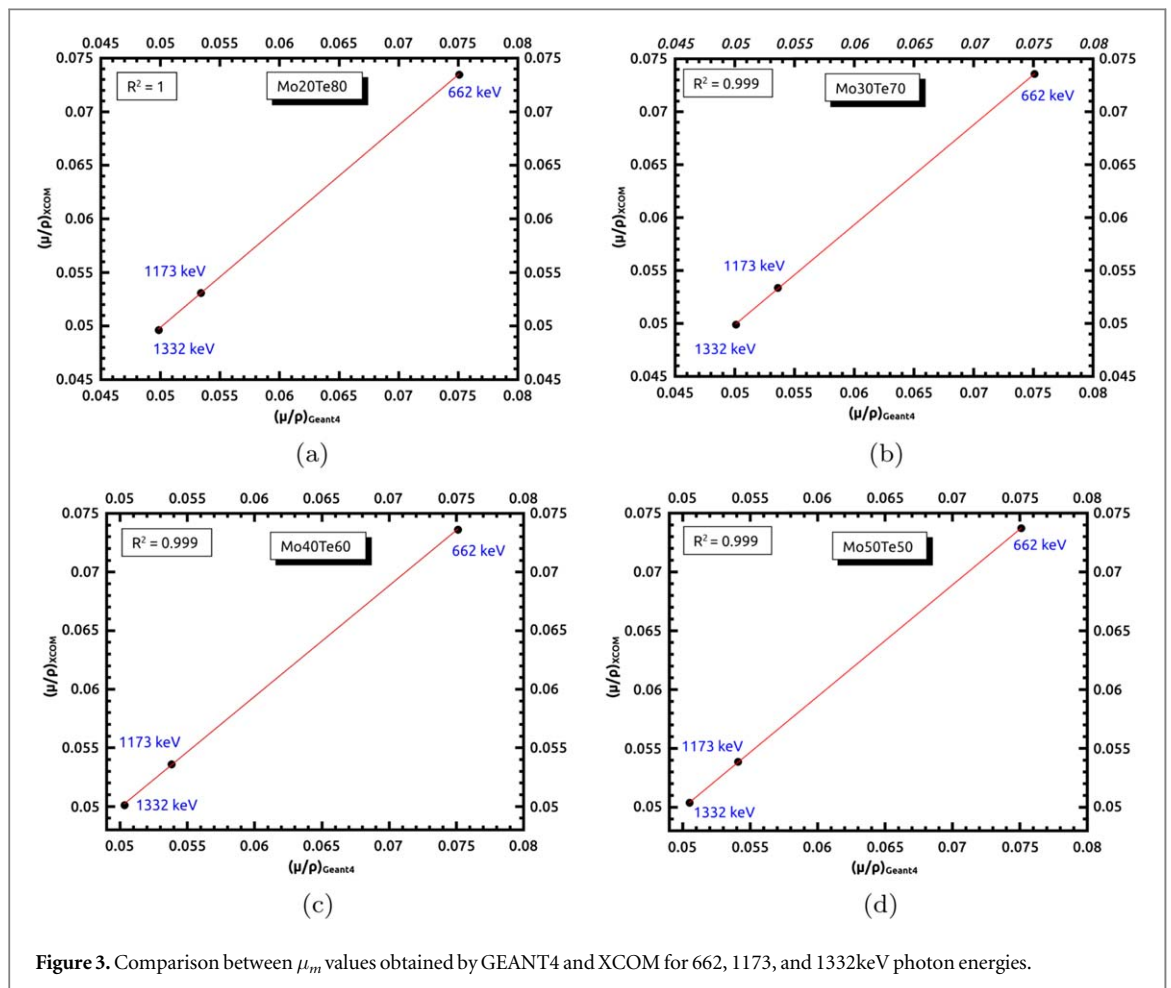


Figure 3. Comparison between μ_m values obtained by GEANT4 and XCOM for 662, 1173, and 1332keV photon energies.

according to Beer–Lambert law [21, 22]. Moreover, the theoretical values of μ_m for these glasses were obtained by using XCOM software [23].

2.2. Effective atomic number (Z_{eff})

The Z_{eff} signifies partial photon interactions with the shielding material. The Z_{eff} can be directly calculated by using the relation below [24, 25]:

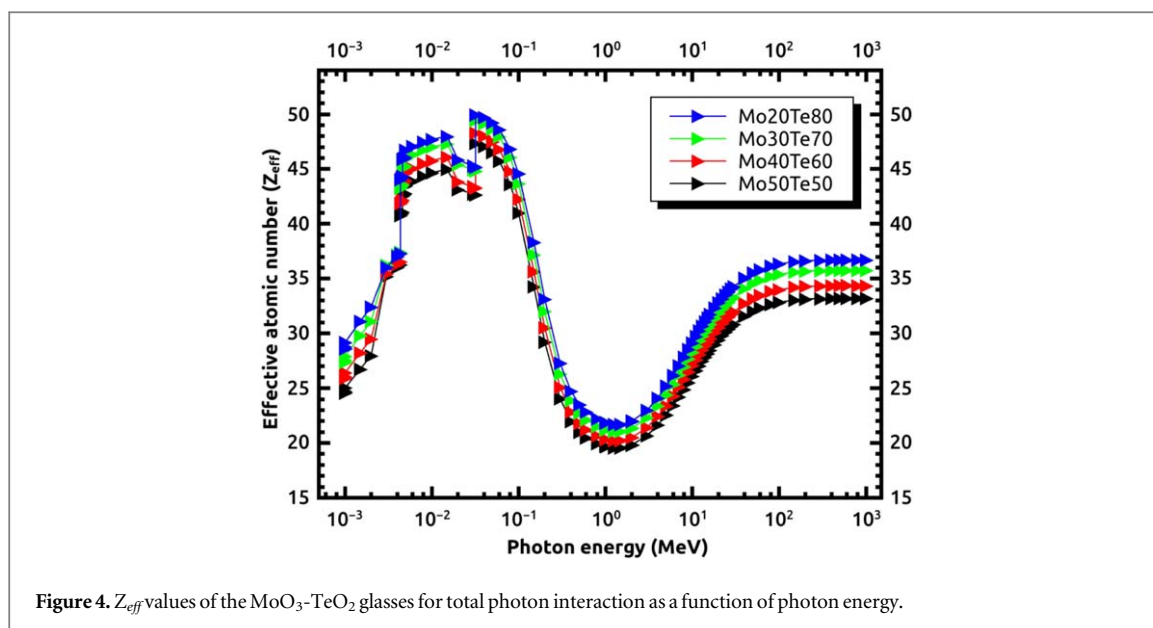


Figure 4. Z_{eff} values of the MoO_3 - TeO_2 glasses for total photon interaction as a function of photon energy.

Table 2. Z_{eff} values obtained in this study and of some tellurite glasses reported in the literature.

Energy (MeV)	Mo20— Te80	Mo30— Te70	Mo40— Te60	Mo50— Te50	Ba20— Te80 ^a	Bi10— Te90 ^b	V10— Te90 ^c	Na10— Te60 ^d
0.01	47.71	47.01	45.78	44.66	47.64	49.17	40.85	39.9
0.02	45.83	45.43	43.83	43.13	48.01	29.15	29.8	41.21
0.03	45.27	44.94	43.39	42.74	47.99	28.69	29.18	41.26
0.04	49.71	49.16	48.02	47.06	49.42	65.68	59.36	79.7
0.05	49.24	48.66	47.50	46.50	49.06	64.94	58.09	78.06
0.06	48.60	47.97	46.78	45.73	48.72	63.62	56.04	75.33
0.08	46.83	46.08	44.78	43.61	47.75	59.72	50.36	67.59
0.1	44.58	43.68	42.27	40.99	46.48	30.74	43.97	58.63
0.2	33.13	32.00	30.53	29.22	38.67	27.12	23.85	29.34
0.5	23.52	22.77	21.80	20.99	29.39	23.86	15.12	16.71
1	21.87	21.23	20.39	19.71	27.47	23.16	14.01	15.28
2	21.98	21.34	20.50	19.83	27.56	23.35	14.08	15.46
3	22.99	22.31	21.41	20.68	28.69	24.16	14.74	16.43
4	24.12	23.39	22.43	21.64	29.92	25.07	15.51	17.53
5	25.20	24.43	23.41	22.58	31.07	25.96	16.28	18.6
6	26.17	25.37	24.31	23.43	32.07	26.75	17	19.6
7	27.05	26.22	25.11	24.20	32.96	27.47	17.68	20.51
8	27.84	26.99	25.85	24.90	33.74	28.12	18.3	21.36
9	28.54	27.67	26.50	25.53	34.42	28.7	18.88	22.13
10	29.16	28.28	27.08	26.10	35.02	29.22	19.41	22.83

Notes.

- ^a 80TeO₂-20BaO [27].
- ^b 90TeO₂-10Bi₂O₃ [26].
- ^c 90TeO₂-10V₂O₅ [29].
- ^d 60TeO₂-10B₂O₃-10MoO₃-10ZnO-10Na₂O [30].

$$Z_{eff} = \frac{\sum_i f_i A_i \left(\frac{\mu}{\rho}\right)_i}{\sum_j \frac{f_j A_j}{Z_j} \left(\frac{\mu}{\rho}\right)_j} \tag{1}$$

where f_i refers to the fractional abundance, and A_i is the atomic weight. Then, electron density (N_{eff}) can be calculated by the equation below [20];

$$N_{eff} = N_A \frac{nZ_{eff}}{\sum_i n_i A_i} \text{ (electrons/g)} \tag{2}$$

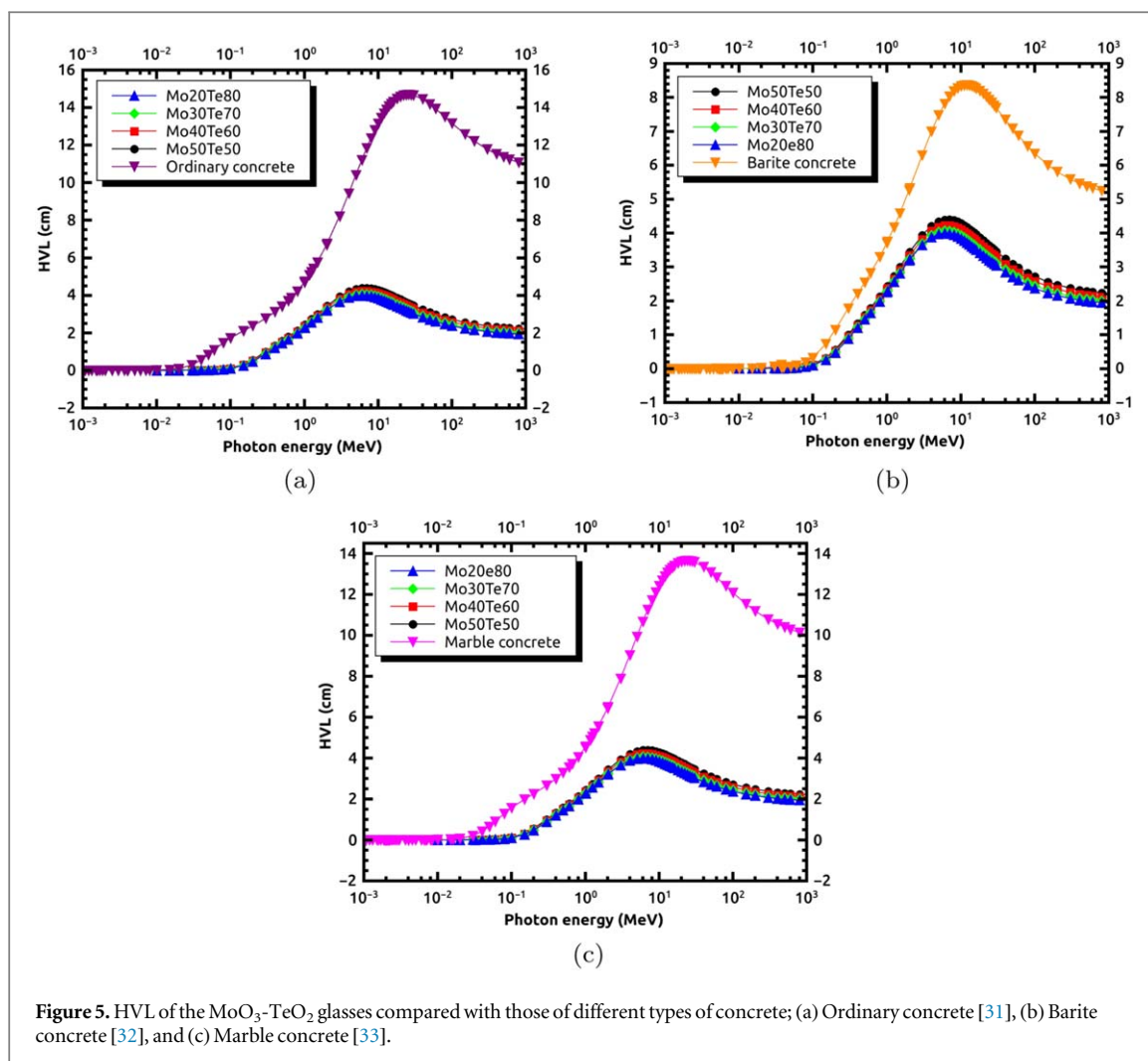


Figure 5. HVL of the MoO₃-TeO₂ glasses compared with those of different types of concrete; (a) Ordinary concrete [31], (b) Barite concrete [32], and (c) Marble concrete [33].

2.3. Half value layer (HVL)

The HVL is a certain thickness of certain shielding material necessary to reduce gamma-ray intensity to half of its original value. The HVL depends on the energy of gamma-ray and on the type of the shielding material. The following relation gives the numerical value of HVL [7, 26]:

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

2.4. Mean free path (MFP)

The MFP is defined by the distance travelled between two gamma-ray collisions. The MFP depends also on the energy of gamma-ray and on the type of the shielding material. The following relation gives the numerical value of MFP [27]:

$$\text{MFP} = \frac{1}{\mu} \quad (4)$$

3. Results and discussion

The mass attenuation coefficients of the tellurite glasses in the form $x\text{MoO}_3-(100-x)\text{TeO}_2$ with $x = 20, 30, 40,$ and 50 mol% have been determined by using Geant4 toolkit and XCOM software. The obtained results of μ_m have been plotted in the energy range of 1 keV-1000 MeV as shown figure 2.

It can be seen that the μ_m values of the present glasses depend on the energy of gamma-ray as well as on the atomic composition of the glass sample. The values of μ_m were observed to be large when the photon energy is small as well as when the Te content is considerable in the glass sample. When the photon energy is small (0.001–0.5 MeV), one can notice a considerable decrease in μ_m values due to the dominance of the photoelectric

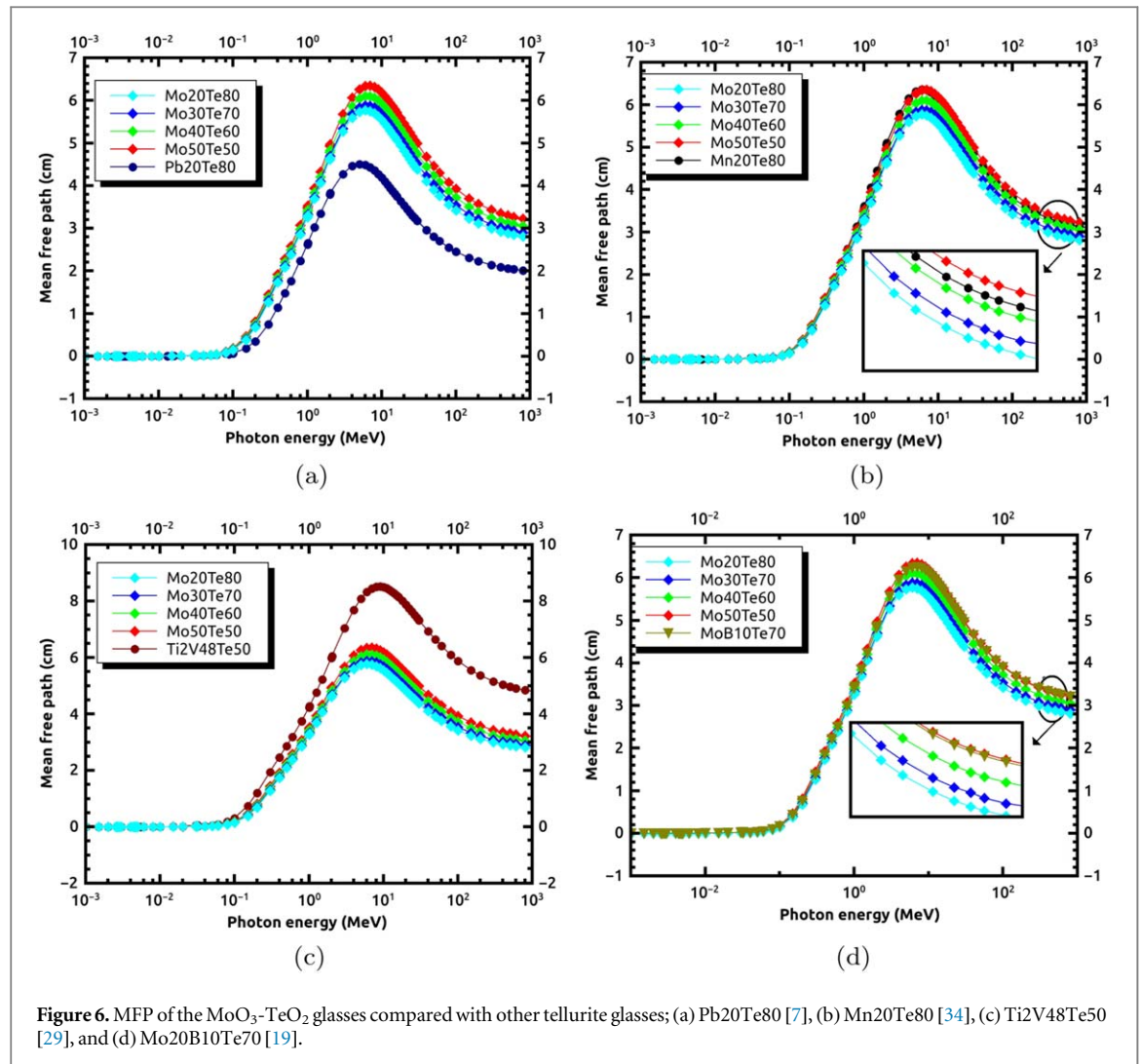


Figure 6. MFP of the $\text{MoO}_3\text{-TeO}_2$ glasses compared with other tellurite glasses; (a) $\text{Pb}_{20}\text{Te}_{80}$ [7], (b) $\text{Mn}_{20}\text{Te}_{80}$ [34], (c) $\text{Ti}_2\text{V}_{48}\text{Te}_{50}$ [29], and (d) $\text{Mo}_{20}\text{B}_{10}\text{Te}_{70}$ [19].

absorption process in this energy range. By increasing the photon energy (0.5–4 MeV), the decrease is still in the values of μ_m but in this case a slight decrease was observed due to the Compton effect. By more increase in the photon energy (4–1000 MeV), a very trivial increase can be seen in the values of μ_m due to the process of creation and annihilation of electron-positron pairs. Moreover, some abrupt discontinuities can be seen in the figure 2 due to the photoelectric effect near the absorption K-edge of Mo and Te elements at 20 keV and 31.8 keV, respectively. Among our proposed glasses, the one coded as $\text{Mo}_{20}\text{Te}_{80}$ have a higher μ_m due to the $\text{Mo}_{20}\text{Te}_{80}$ has higher weight fraction of the heavy metal oxide (TeO_2 , 80 mol%). The present glasses offer very large μ_m comparing to commercial glasses [28]. In order to confirm the accuracy of the present μ_m values, we carried out the Geant4 simulations at some photon energies such as 662, 1173, and 1332keV. The comparison of μ_m values obtained by Geant4 and XCOM codes is shown in figure 3.

From this figure it is seen that there is strong correlation between the between the theoretical and simulation values. Such that we found the correlation factor (R^2) of the μ_m values to be almost one. The calculated μ_m values were then used to compute Z_{eff} values which were plotted in figure 4.

Obviously, the Z_{eff} values are large at the low energies, small at the intermediate energies, and constant at the high energies. Also, there is n abrupt change occurs at 20-32 keV due to the absorption edges of Mo and Te elements. Moreover, the highest Z_{eff} value was found for $\text{Mo}_{20}\text{Te}_{80}$ glass and the lowest Z_{eff} value was noted for $\text{Mo}_{50}\text{Te}_{50}$ glass. This is because of that the interaction of photon with matter is directly related to Z (atomic number). For example, the photoelectric interactions rely on Z^4 , the Compton interactions rely on Z, and the pair production interactions rely on Z^2 . Thus, the highest value of Z_{eff} was noted for $\text{Mo}_{20}\text{Te}_{80}$ glass, which contains the highest weight fraction of Te. Since a higher Z_{eff} refers to a better shield against gamma-rays, $\text{Mo}_{20}\text{Te}_{80}$ is found to be a superior shielding material among the studied glass samples. The Z_{eff} values of the studied glasses were compared with different promising tellurite glasses in table 2.

It is clear that the Z_{eff} values of the studied glasses are higher than those of $90\text{TeO}_2-10\text{V}_2\text{O}_5$ [29] and $60\text{TeO}_2-10\text{B}_2\text{O}_3-10\text{MoO}_3-10\text{ZnO}-10\text{Na}_2\text{O}$ [30]. On the other hand, the Z_{eff} values of the studied glasses are lower than those of $90\text{TeO}_2-10\text{Bi}_2\text{O}_3$ [26], and comparable with those of $80\text{TeO}_2-20\text{BaO}$ [27].

The HVL values of the present glasses were calculated and then plotted along with those of ordinary concrete, barite concrete and marble concrete in figure 5. In gamma-ray applications, the lower HVL values indicate to the higher shielding ability of the materials. From figure 5, it can be seen that the HVL values of the present glasses are very small at the low photon energies. Then, HVL values increase as the photon energy increases and a peak was observed around 10 MeV for all studied glasses. Thereafter, the HVL values of the present glasses decrease with increasing the photon energy. Such behavior of HVL values with incident photon energy can be explained similarly to the aforementioned discussion of μ_m . The HVL values of the $\text{MoO}_3\text{-TeO}_2$ glasses were compared with those of different types of concrete; (a) Ordinary concrete [31], (b) Barite concrete [32], and (c) Marble concrete [33]. It is seen obviously that the shielding properties of the present glasses are better than those of conventional concrete and thus these glasses can serve as shielding materials for gamma-rays applications. The MFP values of the studied glasses are calculated and then plotted along with those of collected tellurite glasses in figure 6.

The MFP values of the studied glasses were observed to be low at the low energies. By the increasing of the photon energy the MFP values increase very fast, so in the shielding applications it is better to increase the thickness of the glass because the gamma-ray can penetrate the glass more deeply. Moreover, figure 6 reveals that the present glasses have better shielding properties than those of $\text{Ti}_2\text{V}_4\text{Te}_5\text{O}_{20}$ glass [29], lower than those of $\text{Pb}_2\text{OTe}_8\text{O}_{20}$ [7] glass and comparable with those of $\text{Mn}_2\text{OTe}_8\text{O}_{20}$ [34] and $\text{Mo}_2\text{OB}_{10}\text{Te}_7\text{O}_{20}$ glasses [19].

4. Conclusion

In the present work, four glasses belonging to the system of $x\text{MoO}_3(100-x)\text{TeO}_2$ ($20 \leq x \leq 50$ mol%) have been investigated for gamma shielding applications. The μ_m values of these glasses were obtained by Geant4 Monte Carlo simulation and by XCOM program. The Monte Carlo simulation and XCOM results were comparable. The shielding effectiveness parameters such as Z_{eff} , HVL and MFP were computed. The presence of TeO_2 reduces MFP values and improves the gamma-ray shielding effectiveness. $\text{Mo}_2\text{OB}_{10}\text{Te}_7\text{O}_{20}$ glass was found to be most promising shielding materials as compared with many conventional concretes and other newly developed glasses.

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