

Effect of sodium oxide (Na₂O) glass modifier on physical properties and gamma shielding in tellurite glass systems TeO₂-ZnO-PbO-Bi₂O₃ (TZPB)

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Abstract: TZPB glasses with the composition 55TeO₂-(41-x)ZnO-2Bi₂O₃-2PbO-xNa₂O (x= 2; 2.5; 3; 3.5 mol%) have been fabricated and characterized to determine the physical properties and gamma radiation shielding parameters. The method of glass fabrication is melt quenching with the holding temperature at 900°C for 30 minutes and the annealing temperature at 256°C for 6 hours. The result of the characterization of the density and gamma shielding parameters was calculated using Phy-X PSD software. The glass density was measured using Archimedes's principle and showed a decrease from 5.79 to 5.73 g/cm³. The molar volume increased from 23.3 to 23.6 g/mol with the addition of Na₂O concentration. Gamma radiation shielding parameters, LAC, MAC, MFP, HVL, and TVL simulated with the energy range were 10⁻³-10⁵ MeV. The results of Phy-X/PSD software showed an increasing MAC, MFP, HVL, and TVL and decreasing in LAC with an increase in Na₂O concentration.

Keywords: Tellurite Glass, Gamma Shielding, Melt-Quenching, Phy-X PSD, Shielding Characteristics

1. Introduction

One electromagnetic radiation which has massive photon energy, high frequency, and short wavelength is gamma radiation. Gamma radiation can harm living cells by exposing them to high levels of radiation (AbuAlRoos *et al.*, 2020). The effects of gamma radiation are skin rashes, organ failure, genetic damage, carcinogenicity, and even death with high dose levels of exposure. Therefore, a new material is needed to reduce the effects of gamma radiation exposure.

The required characteristic of gamma radiation shielding is a material with a high density and high atomic number. The material properties are needed to absorb gamma radiation as much as possible to reduce the level of dose exposure (Alotaibi *et al.*, 2021). In this era, the current research on the development of glass materials is because materials are transparent and can be fabricated in large sizes with high spatial homogeneity, and the production costs are not expensive (Saleh *et al.*, 2022). Currently, the glass with base

tellurium dioxide (TeO₂) appears to be a shielding material for gamma radiation (Alzahrani *et al.*, 2022). The characteristic of tellurite glass is good thermal and chemical stability, low phonon energy, high density, low melting temperature, and effective photon shielding characteristics. It can potentially be a shielding material for gamma radiation (Fausta *et al.*, 2020; Almuqrin & Sayyed, 2021; Marzuki *et al.*, 2022).

In this study, the composition of the glass is 55TeO₂-(41-x)ZnO-2Bi₂O₃-2PbO-xNa₂O with x= 2; 2.5; 3; 3.5 % mol. Tellurium dioxide (TeO₂) can form glass easily and it's called glass former. This type of glass can be used as a good gamma radiation shielding material (Hanfi *et al.*, 2020). The variation composition of ZnO and Na₂O resulted in the changes in physical properties and parameters of the gamma radiation in the samples. This study aims to determine the effect of Na₂O concentration on TZBP glass for the gamma radiation shielding application.

2. Research Methods

TZPB glasses with the composition of 55TeO₂-(41-x)ZnO-2Bi₂O₃-2PbO-xNa₂O (x= 2; 2.5; 3; 3.5 mol%) have been fabricated by melt quenching technique. The first step is stoichiometric calculations to convert the glass composition from mol% to grams so that it is easily weighed with a digital balance. The glass is weighed for a mass of 10 grams. The partial masses of each glass component are shown in **Table 1**.

Table 1. Glass components in the samples

Kode Kaca	Komponen Kaca (gram)				
	TeO ₂	ZnO	Bi ₂ O ₃	PbO	Na ₂ O
G ₁	6,482	2,343	0,688	0,330	0,156
G ₂	6,476	2,311	0,687	0,329	0,195
G ₃	6,470	2,279	0,687	0,329	0,234
G ₄	6,464	2,247	0,686	0,328	0,273

The glass components that were successfully weighed were then mixed and grounded thoroughly in a mortar for ± 30 minutes until a homogeneous mixture was obtained. The glass material mixture is then put into an alumina crucible to be heated in an electric furnace. While in the furnace, the glass mixture is heated for 35 minutes at 900°C. Then all glass samples were annealed at 265°C for the next 6 hours.

Density is a property that describes the distribution of atoms in a glass matrix. Density was measured using the pycnometer method at room temperature based on the Archimedes principle shown in Equation (1) (Marzuki *et al.*, 2020):

$$\rho_g = \frac{m_g \rho_w}{(m_3 - m_0) - (m_2 - m_1)} \quad (1)$$

Where ρ_g , ρ_w , m_g , m_0 , m_1 , m_2 , dan m_3 denote the density of glass, density of distilled water, mass of the sample, the mass of the empty pycnometer, the mass of pycnometer with the sample inside, the mass of the pycnometer filled with water and glass sample, and mass of the pycnometer filled with water. The relationship between density and molar volume is expressed in the following equation (Marzuki *et al.*, 2020):

$$\rho = \frac{\sum x_i M_i}{V_m} \quad (2)$$

Where ρ is the density of the sample, x_i is the molar fraction, M_i is the molecular weight of the oxide that forms the sample, and V_m is the molar volume. The characteristics of the sample as a gamma radiation shield were simulated with the PHY-X/PSD software. This software is available online and does not require paid access (Sakar *et al.*, 2020). To obtain sample characteristics through software, the mole fraction and density of the sample are required. Then, this density value is used by the software as a basis for determining LAC, MAC, TVL, HVL, and MFP values. The simulated energy is in the range of $10^{-3} - 10^5$ meV. Linear Attenuation Coefficient (LAC) is obtained to determine the intensity of gamma radiation that can be reduced after passing through a material (Pires, 2022). The LAC value (μ) is calculated through the following equation (Esawii *et al.*, 2022):

$$\mu = \frac{1}{x} \ln \left(\frac{I}{I_0} \right) \quad (3)$$

Where x is the thickness of the shielding material and I is the radiation intensity. The quantity that indicates the ease with which radiation penetrates a material is the Mass Attenuation Coefficient (MAC) (McAlister, 2012). The MAC value (μ_m) is obtained from a comparison of the LAC value and the mass density (ρ) expressed in the following equation (Esawii *et al.*, 2022):

$$\mu_m = \frac{\mu}{\rho} \quad (4)$$

Shielding parameter that predicted the average distance between two successive interactions respectively is Mean Free Path (MFP), and given by the following relations (Saleh *et al.*, 2022):

$$\text{MFP} = \frac{1}{\mu} \quad (5)$$

Half Value Layer (HVL) is shielding material thickness reduce the intensity of radiation to its half (50% attenuation) (McAlister, 2012). The value of this shielding parameter is shown in Equation (6) (Saleh *et al.*, 2022):

$$\text{HVL} = \frac{\ln 2}{\mu} \quad (6)$$

Tenth Value Layer (TVL) represent the thicknesses of shielding material to decrease the photon intensity to 1/10 (Pires, 2022). This parameter is calculated using the following expression (Rashid *et al.*, 2020):

$$\text{TVL} = \frac{\ln(10)}{\mu} \quad (7)$$

3. Result and discussion

Materials with high densities can absorb gamma radiation well (AbuAlRoos *et al.*, 2020). The density of the TZPB glass ($x= 2; 2.5; 3; 3.5$ %mol) was measured using a pycnometer so the effect on the addition of Na_2O concentration can be analyzed. In G_1 with a concentration of 2%mol Na_2O , a density of 5.795 g/cm^3 was obtained, while in G_4 with a concentration of 3.5%mol, a density of 5.735 g/cm^3 was obtained. The decrease in sample density was due to the substitution of the heavier ion Zn^{2+} ($A_r = 65.38$) with the lighter ion Na^+ ($A_r = 22.99$) thereby reducing the average molecular weight. Meanwhile,

the molar volume increased from 23.36 to 23.61 g/mol. Changes in density and molar volume to the concentration of Na₂O are shown in **Figure 1**.

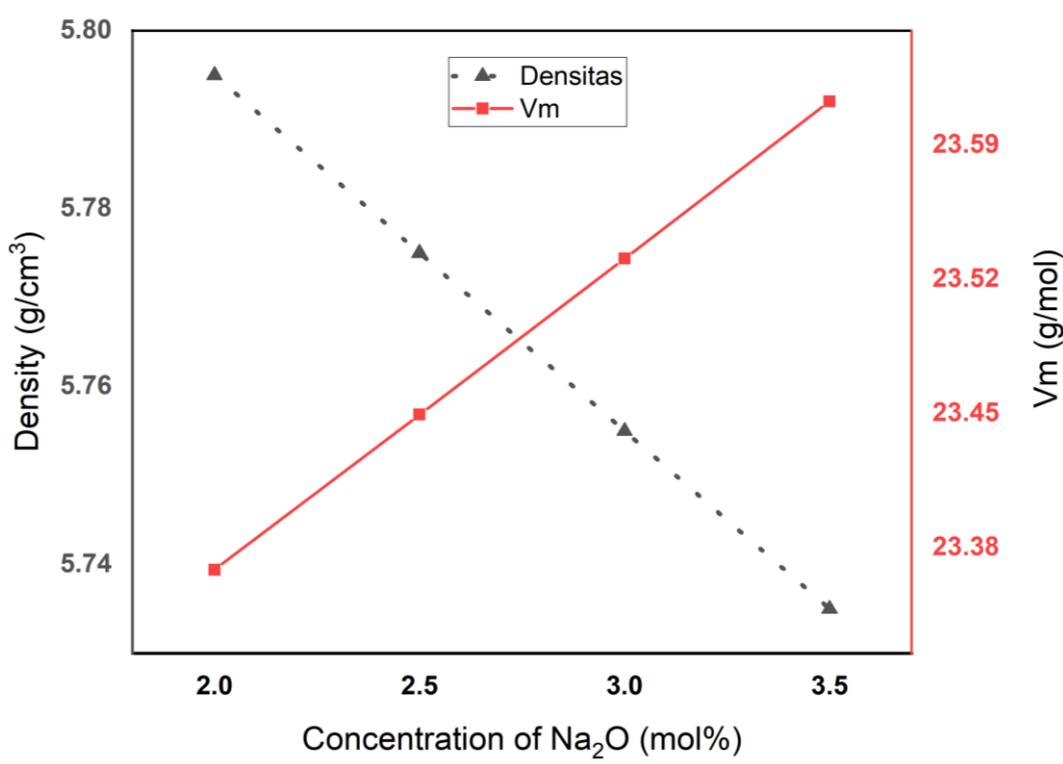


Figure 1. The effect of Na₂O concentration on the density and molar volume of glass TZBP

After obtaining the density values for the glasses, the radiation parameters can be simulated by referring to Equations (3) - (7). Linear Attenuation Coefficient (LAC) is an important parameter that depends on the density of the material used. The LAC value is shown in **Figure 2**. This value is influenced by three basic types of interactions, that is the photoelectric effect, Compton scattering, and pair production. The nature of the interaction between the radiation and the shielding material depends on the energy so various graph trends are produced in different energy zones (Algethami *et al.*, 2022). The greater the density value, the higher the sample density. The high-density value comes from a collection of atoms that have a high atomic number in a small volume so that they are tightly packed. This resulted in the LAC value also increasing (Halliwell *et al.*, 2021).

In regions with energy <0.5 MeV, the LAC value decreases drastically because the photoelectric effect predominates. Meanwhile, regions with an energy range of 0.5-1.5 MeV, it is dominated by Compton scattering, resulting in a curve with a shape that tends to be sloping to horizontally (Algethami *et al.*, 2022). To determine the effect of Na₂O concentration on the LAC value, data samples were taken at 0.005 MeV energy. Based on these data, the sample with the highest Na₂O concentration resulted in the lowest LAC value of 3384.6 cm⁻¹. This shows that the addition of too much Na₂O concentration is not suitable for use as a gamma radiation shielding material because the low molecular weight of the compound causes the density to decrease so that the LAC value also decreases (Halliwell *et al.*, 2021).

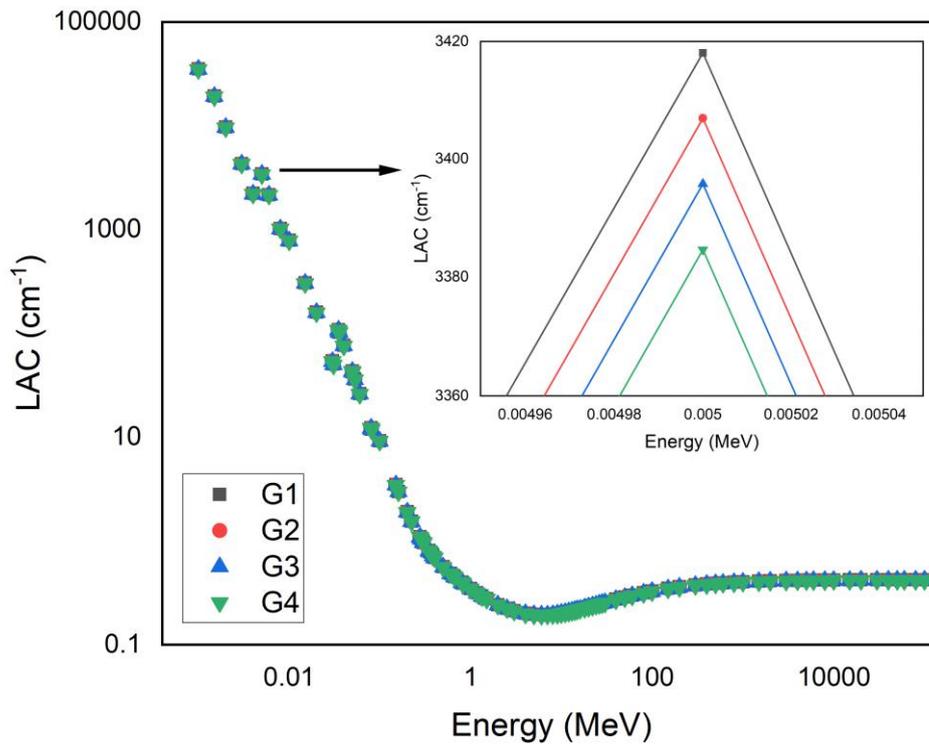


Figure 2. LAC value of TZBP glass with variations of Na₂O doping in the energy range of 10⁻³-10⁵ MeV. The insert shows the enlargement of LAC at energies (4.96 – 5.04)10⁻³ MeV.

The radiation shielding parameter that also depends on density is the Mass Attenuation Coefficient (MAC) value. These values are shown in **Figure 3**. and show the same graphic trend for MAC variation with photon energy (E) of the glass sample. In the low-energy region, the MAC value decreases drastically with increasing gamma energy. This indicates that the photoelectric effect is the dominant phenomenon that occurs so the graph shows a sharp downward trend. The more energy is added, the decrease in MAC value slows down due to the dominance of the Compton scattering phenomenon. Changes in the MAC value with the addition of Na₂O concentration were investigated at 0.356 MeV energy. The maximum value is obtained in G₄, while the minimum value is generated by G₁. The increase in the MAC value is due to the increased probability of photon interactions in the glass sample. This increase in value indicates that G₄ with 3.5% Na₂O concentration is a better shielding composition in absorbing gamma radiation.

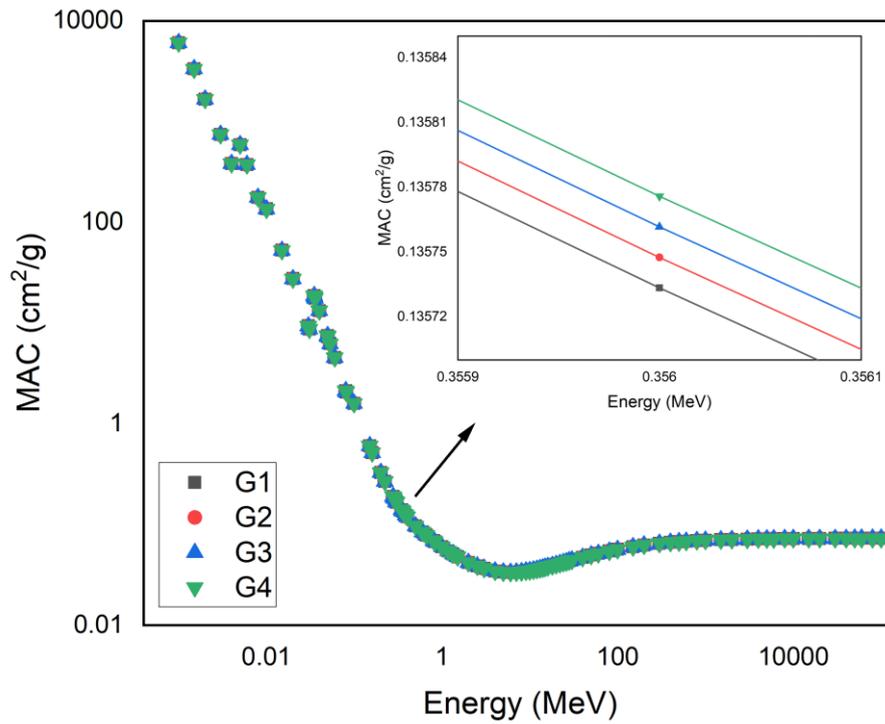


Figure 3. MAC value of TZBP glass with variations of Na₂O doping in the energy range of 10⁻³-10⁵ MeV. The insert shows the MAC magnification at energies 0.355-0.356 MeV.

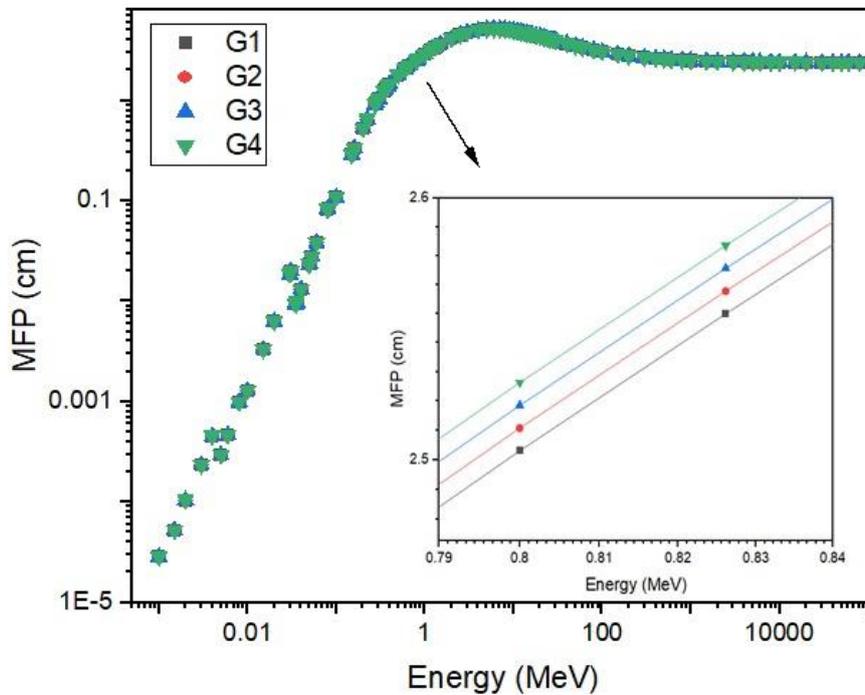


Figure 4. Comparison of Mean Free Paths (MFP) of TZBP glass system doped with various Na₂O at range energy 10⁻³-10⁵ MeV. The insert shows the MFP magnification at energies of 0.79-0.84 MeV.

Also, another studied parameter related to shielding aspects is mean free path (MFP) or relaxation length. The change of mean free path (MFP) with gamma energy is presented in **Figure 4**. There are three possible events due to the interaction of radiation with matter, photoelectric effect, Compton scattering, and pair production (Liang *et al.*, 2020). The interaction of radiation with the shielding material is influenced by the amount of energy generated in the range of energy values and variations in the trend graph are obtained. The phenomenon which occurs in the energy of more than 1 MeV was pair production (Xue *et al.*, 2021). From **Figure 4**, the pair production interactions that produce electrons and positrons occur in the energy range above 50 MeV. Besides that, based on the results of the graph obtained, shows that the MFP of G₁ glass is less than other prepared samples. Therefore, the comparative curves of MFP for samples indicate that G₁ glass composition is much better for radiation shielding relative to different samples of this glass system

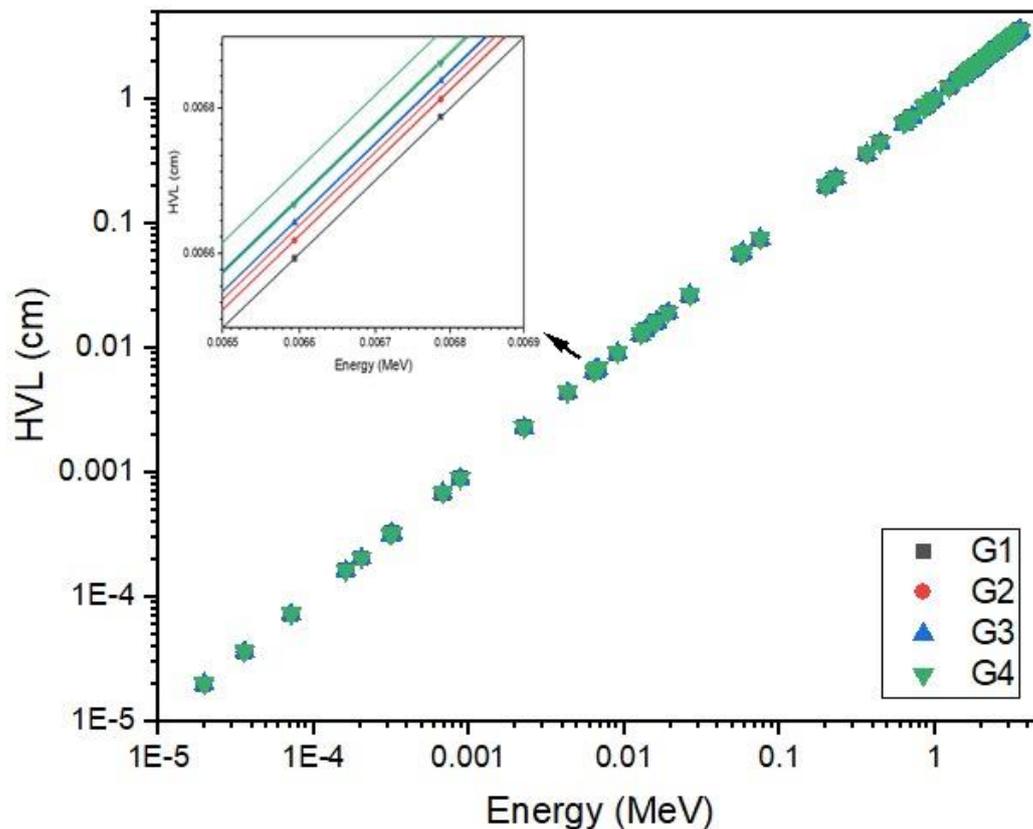


Figure 5. Comparison of Half Value Layer (HVL) of TZBP glass system doped with various Na₂O at range energy 10⁻³-10⁵ MeV. The insert shows the enlargement of HVL at energies (6.5-6.9)10⁻³ MeV.

The radiation shielding property parameter related to the thickness of the glass sample is the half-value layer (HVL). The HVL is the thickness of the radiation shield needed to reduce the intensity of the incoming photons by 50% (Eid *et al.*, 2022). If the HVL is low, it indicates that the glass is better for use as a radiation shield (Waly *et al.*, 2018). The

estimated values of the Half Value Layer (HVL) as a function of gamma photon energy for four glass samples have been shown in **Figure 5**. Based on glass samples, the HVL increases in the order glass system of G₁, G₂, G₃, and G₄. Thus, based on absorber thickness, the best shielding material glass is the G₁ glass system with the lowest HVL value to other samples of this glass system. This is due to the high content of higher atomic number elements i.e. tellurium (Z = 52) that increases the photoelectric effect with absorbing glass material.

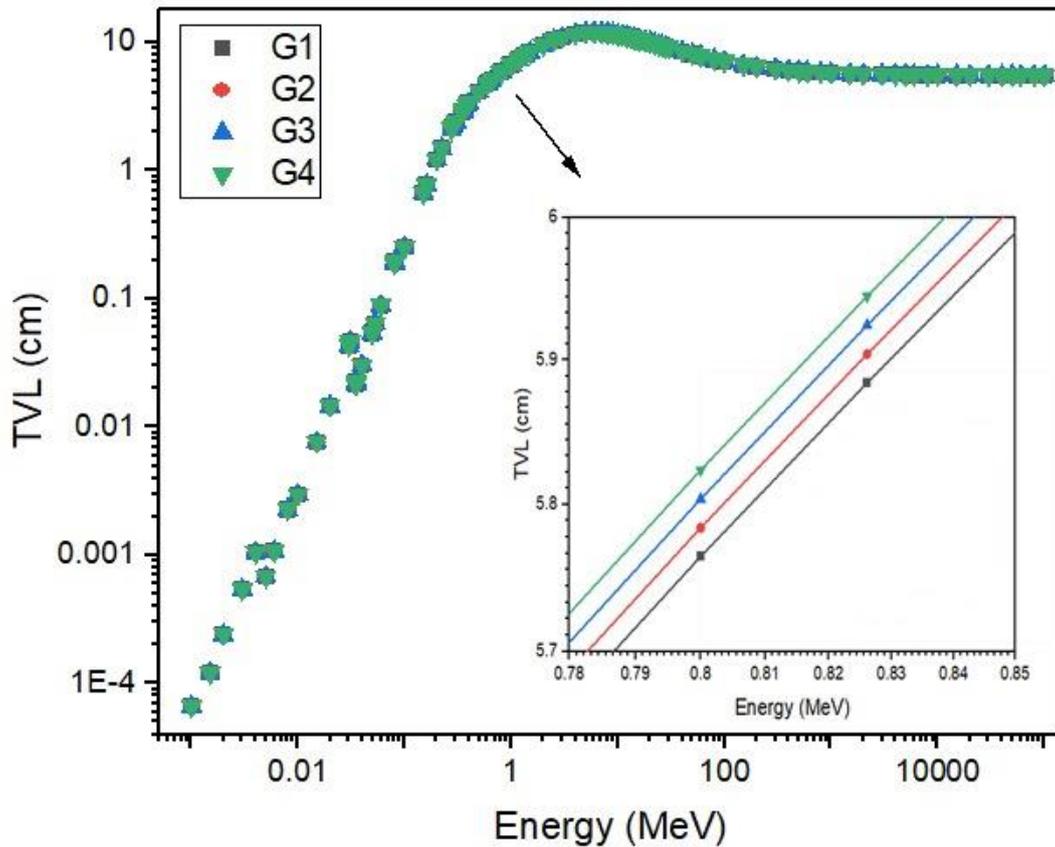


Figure 6. Comparison of Tenth Value Layer (TVL) of TZBP glass system doped with various Na₂O at range energy 10⁻³-10⁵ MeV. The insert shows the TVL magnification at energies 0.78-0.85 MeV.

The radiation shielding property parameter related to the thickness of the glass sample is the tenth value layer (TVL). The TVL is the thickness of the radiation shield needed to reduce the intensity of the incoming photons by 10% (Rashid *et al.*, 2020). If the TVL is low, it indicates that the glass is better for use as a radiation shield. The estimated values of the Tenth Value Layer (TVL) as a function of gamma photon energy for four glass samples have been shown in **Figure 6**. There are three possible events due to the interaction of radiation with matter, are the photoelectric effect, Compton scattering, and pair production (Liang *et al.*, 2020). One of the phenomena that occur in the production of pairs may occur in the energy range of more than 1 MeV. From **Figure 6**, the pair production interactions that produce electrons and positrons occur in the energy range

above 70 MeV. Besides that, based on glass samples, the TVL increases in the order glass system of G_1 , G_2 , G_3 , and G_4 . Thus, based on absorber thickness, the best shielding material glass is the G_1 glass system with the lowest TVL value to other samples of this glass system.

4. Conclusion

TZBP glass system with the composition $55\text{TeO}_2-(41-x)\text{ZnO}-2\text{Bi}_2\text{O}_3-2\text{PbO}-x\text{Na}_2\text{O}$ where $x = 2; 2.5; 3; 3.5$ mol% has been fabricated with the holding temperature at 900°C for 30 minutes. The density decreased from 5.79 to 5.73 g/cm^3 , oppositely the molar volume increased from 23.36 to 23.61 g/mol due to the substitution of the heavier ion Zn^{2+} ($A_r = 65.38$) with lighter ion Na^+ ($A_r = 22.99$) which reduce the glass average molecular weight. According to the density results, the radiation shielding parameters were calculated using Phy-X/PSD software. The simulation process was carried out using a range of energy 10^{-3} - 10^5 MeV. Based on the simulation, the LAC decreased, whereas an increase of MAC, MFP, HVL, and TVL with the addition of Na_2O . It can be concluded, the addition of a glass modifier with light molecular weight such as Na_2O into the glass structure caused radiation shielding characteristics reduced. The TZBP glass with less Na_2O is more potential to be applied in gamma radiation shielding.

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