
Studies on the Shielding Properties of Transparent Glasses Prepared from Rice Husk Silica

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To cite this article:

A. G. Mostafa, H. A. Saudi, M. Y. Hassaan, S. M. Salem, S. S. Mohammad. Studies on the Shielding Properties of Transparent Glasses Prepared from Rice Husk Silica. *American Journal of Modern Physics*. Vol. 4, No. 4, 2015, pp. 149-157. doi: 10.11648/j.ajmp.20150404.11

Abstract: The glasses studied here have been prepared by the melt quenching technique from Egyptian rice husk silica. The obtained solid glasses exhibit amorphous nature and transparency. These glasses were selected to obey the following composition, (75-x) mol % RH silica. 25mol % Na₂O. x mol % Bi₂O₃, where x takes the values, 0, 5, 10, 15 and 20. It was found that, as Bi₂O₃ was gradually increased both the density and molar volume values increased. The comparison between the experimental and empirical density and molar volume values confirm that all the studied glasses are in amorphous glassy phase. Moreover, all samples showed also good shielding properties against gamma ray radiation and neutrons. The mass attenuation coefficient of gamma ray radiation increased gradually as Bi₂O₃ was gradually increased while the HVL values exhibit gradual decrease, and the sample that contains 20 mol % Bi₂O₃ appeared the efficient one at low gamma ray energy (especially at 356 KeV). On the other hand "for shielding neutrons" the sample that contains 5 mol% Bi₂O₃ appeared the best one since it exhibits the highest neutron removal cross section.

Keywords: Bismuth Oxide, Agricultural Wastes, Egyptian RH- Silica, Shielding Properties, Radioactive Sources, Fast Neutron

1. Introduction

It is worth mentioning that, radioactive sources are not dangerous to human bodies only but they are also detrimental to all sensitive laboratory equipments. Therefore, shielding is necessary to protect mankind and all equipments from penetrative neutrons and gamma-ray energies. It was known that the intensity of gamma-ray varies according to three factors: time, distance and shielding, but the most effective method to attenuate radiation is shielding [1]. Chanthima et al, [2, 3] studied the photon interactions and shielding properties of silicate glasses containing Bi₂O₃, BaO and PbO. Their results showed that silicate glass containing Bi₂O₃ exhibits the best shielding properties followed by those containing PbO and BaO, respectively, and in all cases, glasses are better than concrete. Kaewkhao and Limsuwan [4] showed that, glasses containing heavy-metal oxide such as Bi₂O₃ are promising gamma-ray shielding materials due to their high effective atomic number and strong absorption coefficient of gamma ray. Kirdsiri et al [5] studied gamma-ray shielding and optical properties of borate and silicate glasses containing Bi₂O₃, PbO

and BaO at 662 keV. They found that increasing the additive of the heavy metal oxide, the photoelectric absorption increased while Compton scattering decreased. Kurudirek et al [6] reported a comparative study of the effective atomic number for several glasses containing Pb to lead free systems. They observed that at high energy regions, the lead free glasses showed higher values of effective atomic numbers than lead based glasses. Manohara et al [7] studied the effective atomic numbers and related parameters for CaO – SrO – B₂O₃, PbO – B₂O₃, Bi₂O₃ – B₂O₃, and PbO – Bi₂O₃ – B₂O₃ glasses at photon energy ranges from 1 keV to 1 GeV. They found that for medium-Z glasses, effective atomic number was nearly constant and equal to the mean atomic number in a wide energy range dominated by Compton scattering process. Singh et al [8-12] has found a good agreement between the experimental and theoretical (calculated by the WinXCom program) values of gamma-ray mass attenuation coefficient, the Photon mean free path (MFP), the effective atomic number and the effective electron density

for ZnO – PbO – B₂O₃ glasses, investigated at photon energies of 511, 662, 1173 and 1332 keV. Singh et al [13, 14] carried out two respective studies on the radiation shielding properties of lead barium borate glass and bismuth lead borate glasses. They found that the half value layer decreased with increasing the weight fraction of Pb and Bi. Gerward et al [15, 16] studied gamma ray interaction with barium – borate – (fly ash) rice husk (RH) silica glasses by measuring the attenuation coefficient in the photon energy range of 356 – 1332 keV. They observed also good agreement between the radiation parameters obtained experimentally and theoretically.

From another point of view, Rice husk (RH) is an agricultural residue abundantly available in rice producing countries. In Egypt, a rice husk is one of the major agricultural wastes. Much of this by-product is either burnt or dumped as waste, since limited proportion has been recycled as a raw material for cement industry. But the excessive addition of RH silica to cement act to deteriorate its quality. Furthermore, the demand of cement tends to fluctuate with the construction industry while RH generation increases gradually [17].

Therefore, it is necessary to search for new options to recycle RH. One possibility is to use RH in glass manufacturing, because of the high silica content and low transition oxide contamination. Therefore, many researches have been carried out to extract pure amorphous silica from RH [18-20].

However, in this article, a trial will be done to prepare some bismuth sodium silicate glasses, where SiO₂ was extracted from RH. The preparation of these glasses aims to produce low cost, good transparent and effective shielding materials for the harmful ray.

2. Experimental Procedure

Detailed information about the method used to extract silica from RH was found in ref. [20]

An appropriate amount of sodium carbonate, bismuth oxide and RH silica were weighed using an electric balance having accuracy of 0.0001 g. The batches were mixed well and were then melted in porcelain crucibles at 1300 °C for 2 h using an electric muffle furnace. The melts were stirred several times during melting in order to supply homogeneous glasses having the composition (75-x) mol% RH-silica. x mol% Bi₂O₃. 25 mol% Na₂O, (where 0 ≤ x ≤ 20). The melts were quenched in air between two copper plates, and just after sitting the solid glasses were directly transferred to the annealing furnace at 350 °C and the furnace was turned off and was left to cool to room temperature over-night [21]. All samples appeared transparent and exhibit pure amorphous nature.

The densities of the studied glasses were measured applying Archimedes technique, using CCl₄ as an immersing liquid of stable density = 1.593 gm/cm³ at RT. The experimental density

(ρ_{exp}) can be calculated applying equation (1),

$$\rho_{exp} = \frac{M_a}{M_a - Mc} \rho_c \quad (1)$$

Where: M_a and Mc are the masses of the sample in air and CCl₄ respectively and ρ_c is the density of CCl₄.

The empirical densities (ρ_{emp}) were also calculated applying equation (2) and were exhibited in the same figure for comparison.

$$\rho_{emp} = \sum_j \rho_j \cdot P_j \quad (2)$$

Where $\rho_1, \rho_2, \rho_3, \dots, \rho_j$ are the densities of the constituting oxides and P₁, P₂, P₃,, P_j are the percentage weight of each oxide composing such sample.

It is of interest to calculate the molar volume values, since it relates directly to the internal spatial structure of materials. However, both the experimental and the empirical molar volume values were calculated using equations (3 and 4) respectively,

$$(V_m)_{exp} = \sum (M_i / \rho_{exp}) \quad (3)$$

$$(V_m)_{emp} = \sum (M_i / \rho_{emp}) \quad (4)$$

Mass attenuation coefficient has been determined empirically by using WIN XCOM computer software developed by NIST [22–27].

The values of the total mass attenuation coefficients of the studied glasses were calculated by Win X-COM program, based on the mixture rule (equation (5)),

$$\left(\frac{\mu}{\rho}\right)_m = \sum_{i=0}^n w_i \left(\frac{\mu}{\rho}\right)_{m(i)} \quad (5)$$

Where $(\mu/\rho)_m$ is the total mass attenuation coefficient for a glass sample and w_i is the fractional weight of each oxide in such sample.

The half value layers (HVL) of the studied glass were also calculated according to the equation:

$$HVL = \ln 2 / \mu \quad (6)$$

Where μ is the linear attenuation coefficient.

3. Results and Discussion

The chemical composition of RH after chemical and thermal treatments is presented in Table (1). It was seen that the extracted silica reached 98.6% of RH, in addition to slight traces composed mainly of potassium, calcium, iron and other ignored oxides.

Table 1. Impurity analysis in wt. % and Major chemical constituents of Egyptian rice husk fly ash after treatment.

Constituents (Weight %)	SiO ₂	K ₂ O	CaO	Fe ₂ O ₃	MnO ₂	TiO ₂
Slow heating at 750 °C RH SILICA	98.6	0.23	0.688	0.28	0.042	0.08

3.1. Density (ρ) and Molar Volume (V_m)

Density is an important and accurate property, which strongly reflect the fine changes in the glass structure. Therefore the obtained experimental density values of the studied glasses were plotted as a function of Bi_2O_3 content in Fig. 1, the change in the empirical density was also depicted in the same figure, for comparison.

As was expected, both the experimental and empirical densities increased gradually as Bi_2O_3 was gradually increased. It is obvious that the empirical density values are usually

higher than those obtained experimentally. The rate of the increase of the empirical density was found to be 0.15 g/cm^3 per mol which was found to be higher than the calculated slop of the experimental one that was found to be 0.09 g/cm^3 per mol. All these factors can be taken as evidence for the homogeneity of the glassy state of the prepared samples [28].

The gradual increase in the density values can be attributed to the replacement of one mole of RH silica (mean molecular weight = 60.08 g/mol) by one mole of Bi_2O_3 (molecular weight = 465.96 g/mol).

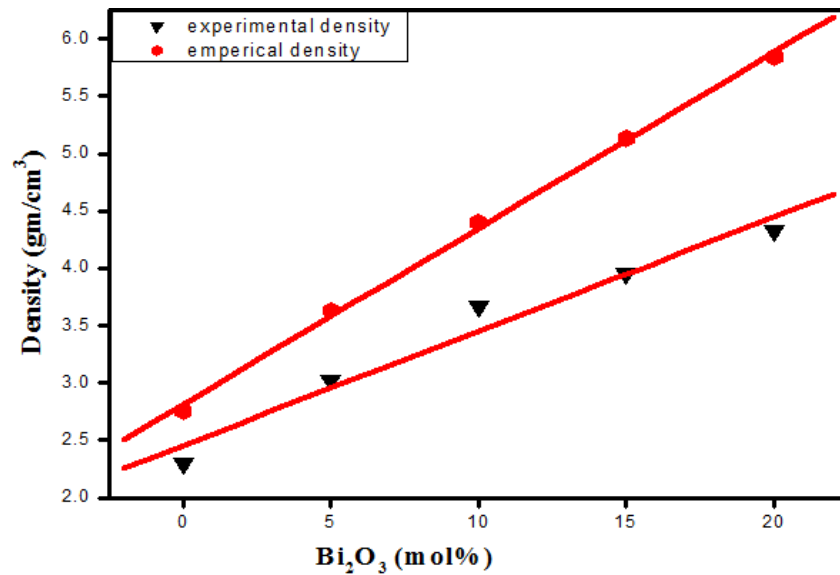


Fig. 1. The change in the experimental and empirical densities versus Bi_2O_3 content.

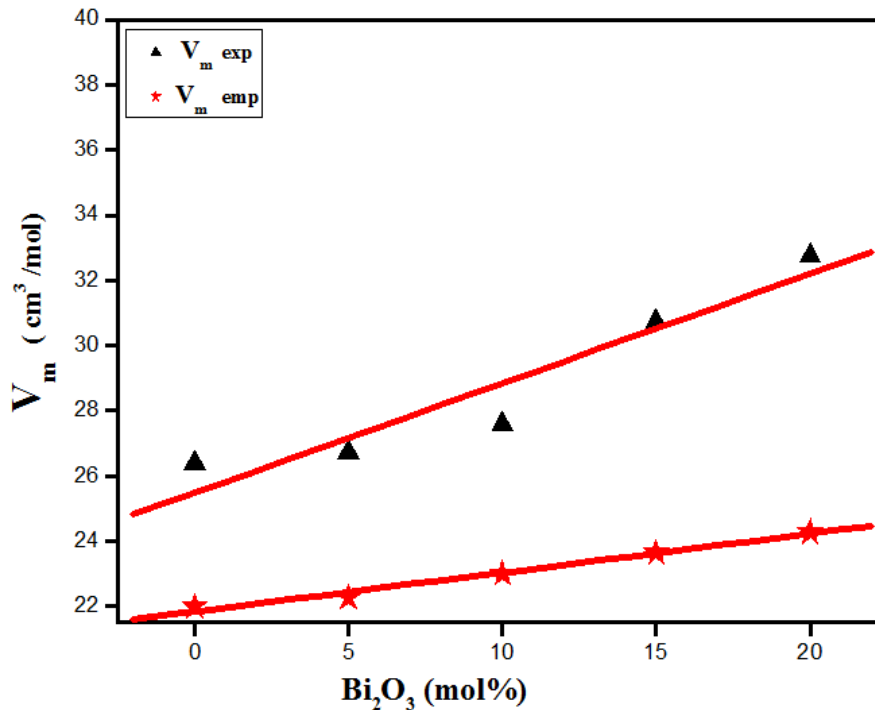


Fig. 2. The change of the experimental and empirical molar volume versus Bi_2O_3 content.

The molar volume, which is defined as the volume occupied by one mole of the material, increases from 27.38 to 32.77 cm³ mol⁻¹ as Bi₂O₃ content raises from 0 to 20 mol.%. Shelby [29] reported that the molar volume depends on the ionic radius of the modifier cations. If the ionic radius of the modifier ions (ionic radius of Bi³⁺ is 1.03 Å) is larger than the interstices of the network structure (ionic radius of Si⁴⁺ is 0.40 Å) their attraction to the oxygen ions can lead to an augment in the size of the interstices and this enhance the molar volume. Therefore Fig. (2), shows the gradual change in the molar volume of the studied glasses versus bismuth oxide content. The empirical molar volume was also represented in the same figure for comparison. A gradual increase was clearly observed in both values of the empirical and the experimental V_m.

It is obvious from this figure that the rate of increase of the experimental V_m is higher than that of the empirical one, which can be taken also as evidence that the prepared samples are of highly homogeneous glassy phase [28].

The observed increase of V_m can be also attributed to:

1. The replacement of one Si cation (ionic radii R_i = 0.4 Å) by two Bi cations (ionic radii of each, R_i = 1.03 Å).
2. The oxygen anions concentration in the glass networks is gradually increased, since two oxygen anion (SiO₂) were replaced by three oxygen anions (Bi₂O₃). This can be confirmed by calculating the number of oxygen ion density in the studied glasses, using the following equation,

$$N_{0\text{-ions}} = \sum_i x \left[\left(\rho \frac{W_{Ti}}{M_{Wi}} \right) N_A \right] \quad (7)$$

Where ρ is the experimental density & W_i is the percentage weight of each component, M_w is the molecular weight of each molecule, N_A is Avogadro's number and x is the number of oxygen atoms in each molecule.

Fig. 3 shows that the number of oxygen ion density increased gradually as bismuth oxide was gradually increased. This result confirms the gradual extension of the oxygen network, which in turn act to increase the molar volume.

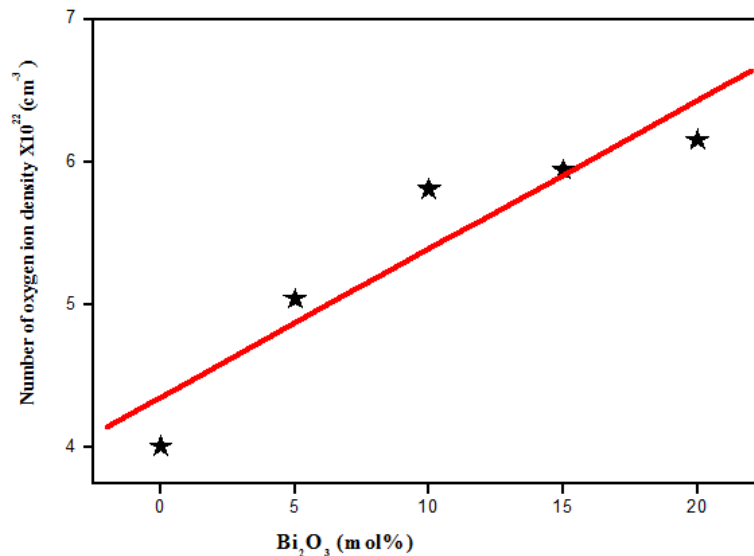


Fig. 3. The gradual change in the number of oxygen ion density versus Bi₂O₃ content.

The result of these glass samples agree well with those reported elsewhere [29-33].

Qi et al [34] have relates the density of binary silicate, borate and phosphate glasses including Bi₂O₃ to the electronic polarizability of the oxide ions and therefore the data are larger than those of binary oxide glasses containing monovalent or divalent metals (by one or two order of magnitude).

It was found that the density and molar volume increased progressively with gradual increase of Bi₂O₃. This is a similar behavior to the data obtained in ref. [35]

3.2. Gamma - Ray Attenuation Properties

3.2.1. Total Mass Attenuation Coefficient and Half Value Layer as a Function of Bismuth Oxide Content

Nowadays, owing to the universal uses of many types of

radio-active sources in different fields (medicine, industry, agriculture ... etc.), daily life becomes dangerous especially for users, since they have to be in contact to different harmful ray. However, the search for suitable shielding materials becomes now of interest. On this sense, the prepared glasses were checked whether they can be used as transparent shield or not.

The γ-ray mass attenuation coefficients [(μ/ρ)_m] have been calculated using Win X-COM program,

1. Firstly at different low γ-ray energies (356, 663, 1173 and 1332 KeV, emitted from ¹³³Ba, ¹³⁷Cs and ⁶⁰Co (for both 1173 and 1332) respectively [35]. Fig. (4) Shows the change in the γ-ray mass attenuation coefficient with the gradual increase of bismuth oxide where a gradual increase of (μ/ρ)_m, can be easily observed.
2. Secondly, Fig. (5) Shows the change in the mass

attenuation coefficient of the studied glasses at high γ -ray energies (10 up to 100 MeV). It appeared that as bismuth oxide was gradually increased, the mass attenuation coefficient increased also.

3. The HVL of the studied glasses was also calculated for the same γ -ray energies considered above. The obtained values as a function of bismuth oxide are represented in Figs (6 & 7) for low and high energies γ -ray respectively. It can be seen obviously that the HVL decreased gradually with the gradual increase of bismuth oxide content.

The observed increase in the γ -ray mass attenuation coefficient as well as the corresponding decrease in the HVL of the studied glasses can be attributed to the gradual replacement of RH silica by Bi_2O_3 . This can be understood logically by knowing that the γ -ray mass attenuation coefficient of bismuth is higher than that of silicon, while the HVL of bismuth is smaller than that of silicon. The μ/ρ (cm^2/g) for bismuth and silicon as well as for oxygen and sodium are

exhibited in Table 2 for comparison [36].

Table 2. $(\mu/\rho)_m$ (cm^2/g) and HVL (cm) for bismuth and silicon at different low γ -ray energies [36].

Energy(KeV)	$(\mu/\rho)_m$ (cm^2/gm)			
	356 KeV	662 KeV	1173 KeV	1332 KeV
Bi	0.2960	0.1130	0.0626	0.0569
O	0.1000	0.0773	0.0588	0.0551
Si	0.1010	0.0773	0.0588	0.0551
Na	0.0963	0.0740	0.0563	0.0528

Generally, it can be stated that, all the studied glasses can be used as gamma-ray shield and to capsule nuclear radio-active wastes before interment underground. These samples work well for law than for high gamma-ray energies. It is worth to state also that the sample containing 20 mol% Bi_2O_3 appeared to be of high suitability to act as shield for the 356 keV gamma-ray energy, since it presents the highest μ/ρ coefficient and lowest HVL values.

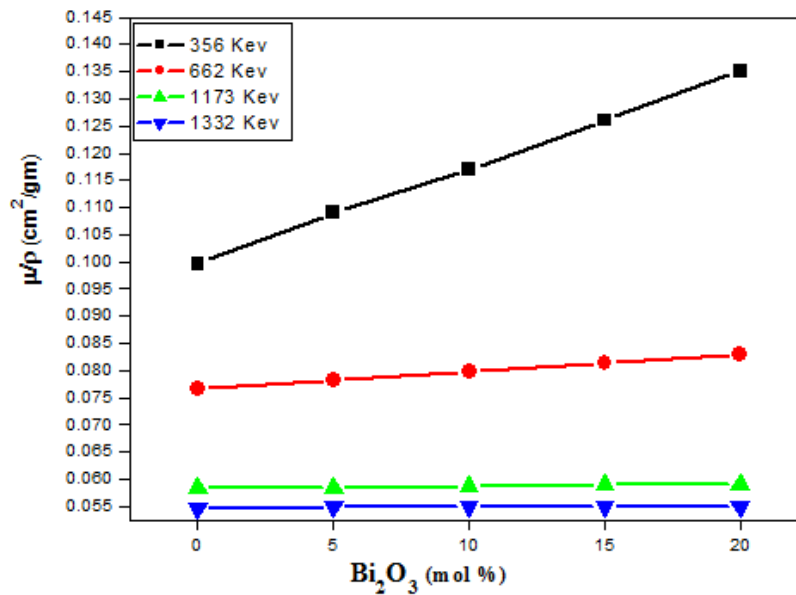


Fig. 4. The mass attenuation coefficient versus Bi_2O_3 content for different low γ -ray energies.

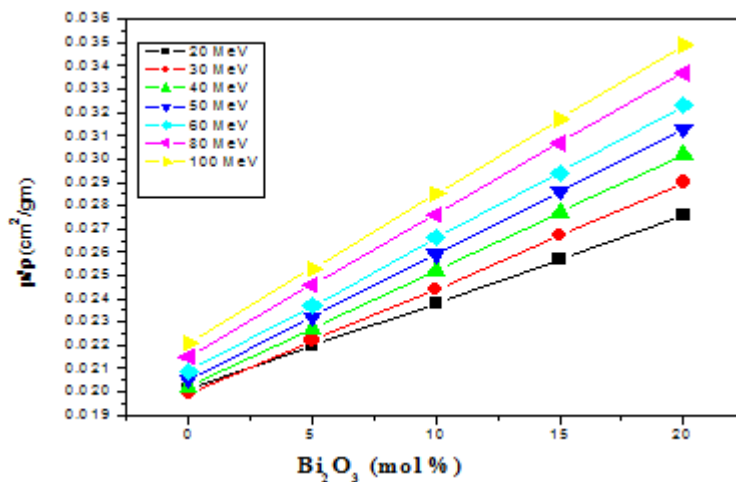


Fig. 5. The mass attenuation coefficient versus Bi_2O_3 content for different high γ -ray energies.

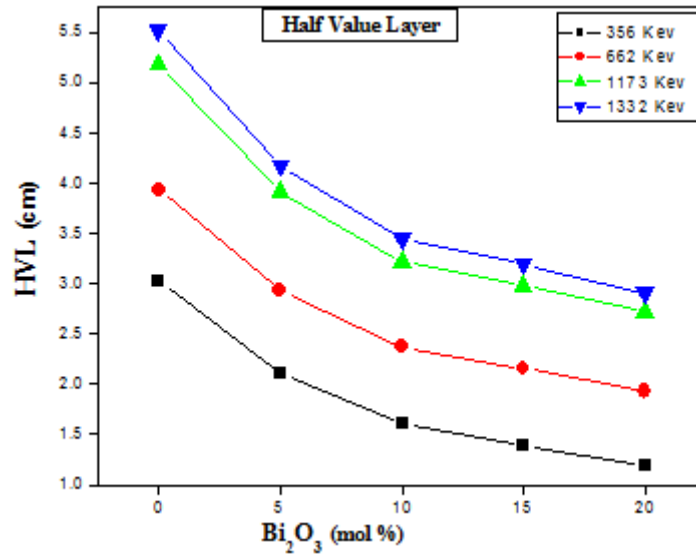


Fig. 6. The HVL of the investigated glasses versus Bi₂O₃ content for different low γ -ray energies.

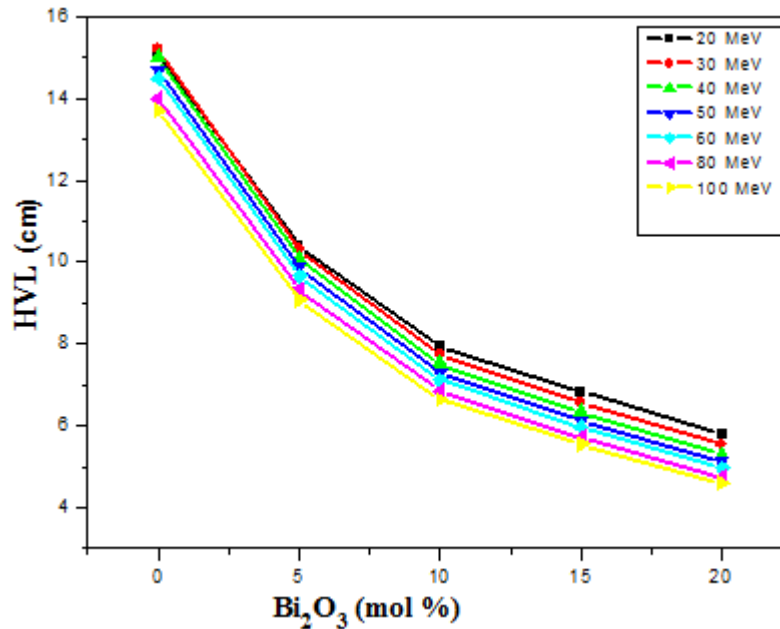


Fig. 7. The HVL of the investigated glasses versus Bi₂O₃ content for different high γ -ray energies.

3.2.2. Total Mass Attenuation Coefficients as a Function of Incident Gamma Ray Energies [37-39]

The mass attenuation coefficient (μ/ρ)_m for the studied glass samples, has been presented graphically in Fig. 8 as a function of the incident gamma-ray energy. It is clear that, at low incident photon energies (0.001-0.20 MeV), (μ/ρ)_m decreases rapidly from 3.24×10^3 at 0.001 MeV to 4.87×10^{-1} at 0.20 MeV. When the incident photons energy increased at the intermediate energy range (0.20 MeV-20 MeV), it decreases slowly. Finally, at high energy regions ($E > 20$ MeV), it appeared to be approximately stable. This behavior can be explained by the fact that the dominance of different interaction processes of photon with matter (photoelectric absorption, Compton scattering and pair production in nuclear field and in electronic field) is not the same for different

photon energies.

Fig. 9 shows the partial mass attenuation coefficients of different photon interaction processes for the studied glass system. It is clear that at low energy region, the photoelectric absorption is the dominant process and the contribution of other processes is negligible. Also, it can be noted that the partial mass attenuation coefficients of the photoelectric absorption decreases rapidly and its contribution becomes negligible starting from 0.20 MeV, because its effective cross section is inversely proportional to the incident photon energy [40]. Therefore, the fast decrease of the total mass attenuation coefficient at low energy range is caused by this effect.

It can be noted also that when the incident photon energy is between 0.20 and 20 MeV, Compton scattering process (especially incoherent) becomes the dominant mechanism, and its partial mass attenuation coefficients increases when the

energy is between 1 and 20 keV. But it is smaller than the partial mass attenuation coefficients of the photoelectric absorption. Then, it becomes almost constant up to 150 keV and from this value it decreases slowly. The behavior of this coefficient is due to the fact that the cross section of Compton scattering process is inversely proportional to the incident photons energy [41]. Therefore, the slow decrease in $(\mu/\rho)_m$ values in the intermediate energy can be explained by the dominance of the Compton scattering process. Finally, in the

high energy region, the pair-production process becomes dominant. The partial mass attenuation coefficient of this process is zero for an energy between 1 keV and 1.02 MeV. Then, it increases linearly with the increase of energy and when the energy is 24 MeV it becomes equivalent to the partial mass attenuation coefficients of the Compton scattering process and from 100 MeV it becomes almost constant. So, this may explain why $(\mu/\rho)_m$ remains almost constant in the high photon energy region.

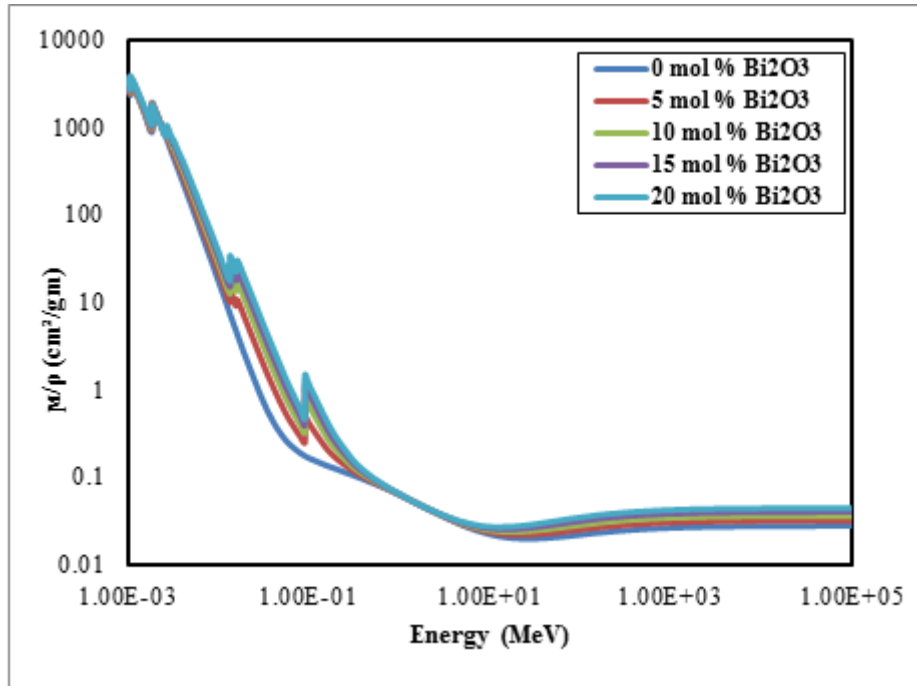


Fig. 8. The mass attenuation coefficient for the studied glasses as a function of the incident gamma ray energies.

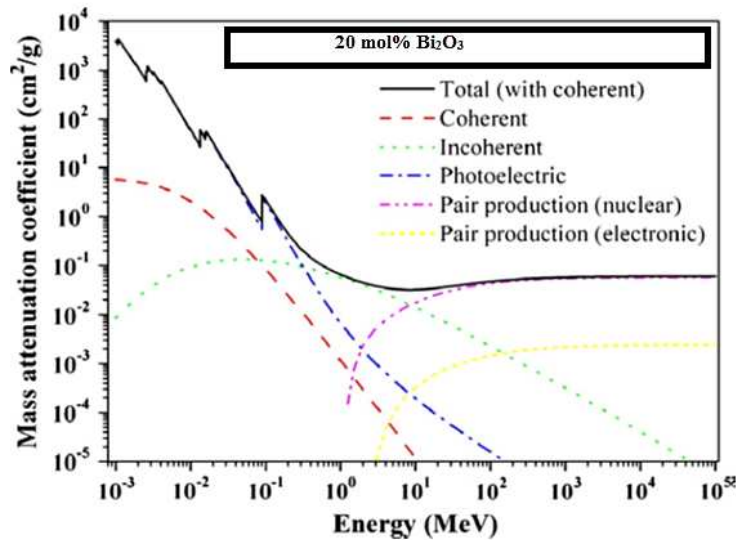


Fig. 9. The variation of mass attenuation coefficient for 20 mol% Bi_2O_3 glass sample with photon energy for total and partial interactions (with coherent).

3.3. Fast Neutron Removal Cross Section

Fast neutron attenuation is described by a parameter called the "removal cross-section", denoted by Σ_R (cm^{-1}). It is

different from the total macroscopic cross-section but it has only a fraction of the macroscopic cross section. The removal cross-section presents the probability that a fast or fission-energetic neutron undergoes a first collision, which

removes it from the group of penetrating un-collided neutrons [41-43]. Indeed, in the MeV-energy region, the absorption cross-section of neutrons is very low compared to the scattering cross-section. In fact, the fast neutrons are not directly absorbed during their passage through the shielding hydrogenated, but they slow primarily by successive elastic collisions with the nuclei of light elements and when their energy is in the order of the thermal energy (0.025 eV), they are absorbed by the nuclei of heavy elements via interaction radiative capture [43].

Generally, shielding materials are chemical compounds or mixtures, their macroscopic removal cross-section can be obtained by calculating their $(\Sigma R)_C$ of their constituent

elements applying the following equation, [41-43],

$$(\Sigma R)_C = \sum_i w_i (\Sigma R/\rho)_i \quad (8)$$

Where W_i , ρ and $(\Sigma R/\rho)_i$ are respectively the partial density (g cm^{-3}), density and mass removal cross section of the i th constituent [44]. Fig. (10) Shows the mass removal cross-sections $(\Sigma R)_C$ as a function of Bi_2O_3 concentration. The calculated values of removal cross-sections $(\Sigma R)_C$ show that the sample contained 5 % Bi_2O_3 has the largest removal cross-section and the sample no. 1, (Bi_2O_3 free sample) has lowest one as shown in Fig. (10). Therefore, the addition little of Bi_2O_3 improves the removal cross section values of these glasses.

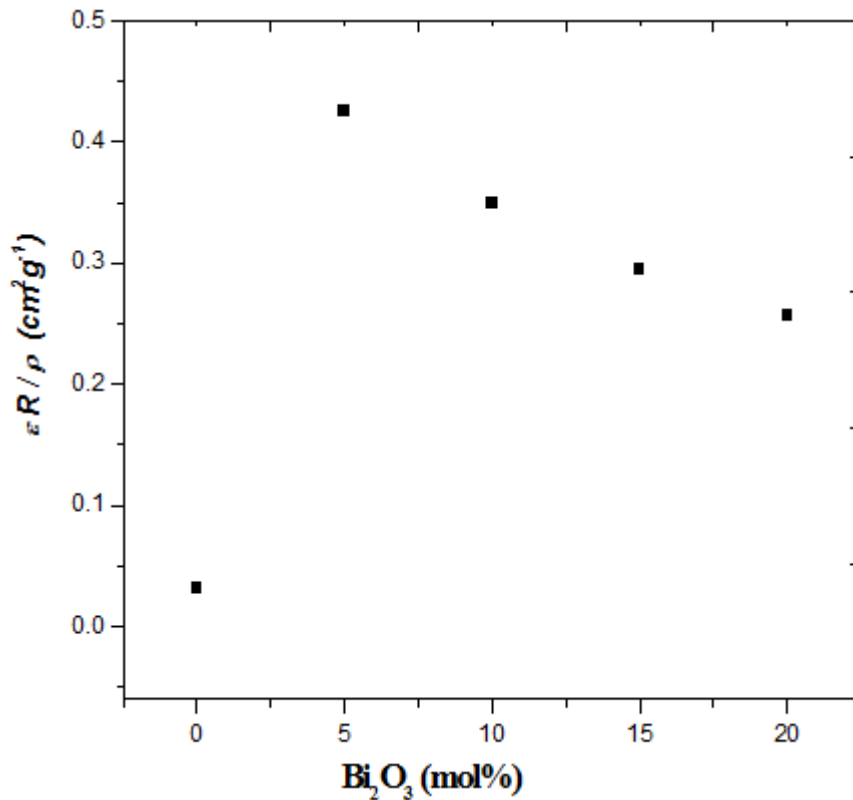


Fig. 10. The removal cross-section $(\Sigma R)_C$ of fast neutrons for the studied glass system.

4. Conclusions

- High pure Silica content can be extracted from RH after chemical and thermal treatments, and homogenous transparent glasses can be manufactured using such RH silica for various scientific and industrial applications.
- The introduction of Bi_2O_3 in elevated amounts increases gradually the density and the molar volume of the studied samples.
- According to the shielding properties, gamma-ray shielding glass can be prepared from RH silica as host glass doped with bismuth oxide. It is seen that, as Bi_2O_3 was increased the gamma-ray mass attenuation coefficient increased and consequently the HVL decreased. Among all glasses the sample that contains 20 mol% Bi_2O_3 is the best one for attenuate 356 keV gamma

ray energy, it seen also that, the sample that contains 5 mol% Bi_2O_3 is the efficient one for attenuate neutrons since it show the highest $(\Sigma R)_C$, in comparison to all other glasses.

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