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Borosilicate Glass Containing Bismuth and Zinc Oxides as a Hot Cell Material for Gamma-Ray Shielding

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Abstract Gamma-ray attenuation coefficients (the half value layer parameters, gamma – ray shielding properties) for the x ZnO - $(5-x)$ Bi_2O_3 -10 B_2O_3 -40 SiO_2 (x = 0, 5, 10, 15 and 20) at 662, 1173 and 1332 keV photon energies have been determined experimentally, using a narrow beam transmission method, as well as theoretically using the 'mixture rule' and the 'XCOM' computer software. The molar volume and FTIR investigations have been used to study the structural properties of the glass system. Optical, UV-visible, absorption measurements were performed also to investigate the transmittance aimed to investigate the performance of the glass system for using as hot cells for shielding windows in nuclear technology.

Keywords: *borosilicate glasses, FTIR measurements, radiation shielding*

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1. Introduction

Today, the use of nuclear energy and radiation is indispensable in modern society. Handling with radiation sources with safe is essential. The knowledge of gamma-ray interaction parameters such as mass attenuation coefficient, half value layer, mean free path, etc. is very important in the field of radiation shielding materials[1,2]. The most radiation shielding materials commonly used is concrete because it is inexpensive and adaptable for any construction design. There are however many drawbacks associated with the usage of concrete, such as considerable variability in its composition and water content. This variation results in uncertainty in calculations for shield design predictions of the radiation distribution and attenuation in the shield. Water contents have the disadvantages of decreasing both density and structural strength of concrete. However, its drawback is the loss of water when concrete becomes hot by absorption of energy from radiation [2,3]. Recently glass materials are one of the possible alternatives to concrete because they can be transparent to visible light and their properties can be modified by composition and preparation techniques. Silicate glasses are the most commonly available commercial glasses due to ease of fabrication and excellent transmission to visible light. Borosilicate glasses are known for having very low coefficients of thermal expansion making them resistant to thermal shock and these glasses have also the ability for an excellent transmission to visible light. Bismuth is expected to enhance the gamma-ray attenuation due to its higher atomic number. Addition of bismuth to glasses has improved also there chemical durability [4]; therefore, bismuth contributes to the stabilization of the glass structure [4]. ZnO containing optical materials have been drawing a good attention of the scientific community over world due to their very interesting optical properties in combination with their non-toxicity, non-hygroscopic nature and lower cost [5].

With these considerations in mind, we have selected this glass system for exploring the possibility of using them as hot cell material for gamma-ray shielding.

2. Theory

For photons in an attenuating medium, HVL (half value layer) is the thickness of a material required to reduce the intensity of the emergent radiation to half [6]. $I = I_o e^{-\mu_m t}$ where I_o denotes the photons with energy E, intensity without attenuation; I the photons with energy E, intensity after attenuation; $\mu_m = \mu/\rho$ (cm^2/g) is the mass attenuation coefficient and t (g/cm^2) is sample mass thickness (the mass per unit area). The total μ_m values for materials composed of multi elements is the sum of the $(\mu_m)_i$ values of each constituent element [6] by the following mixture rule $\mu_m = \sum_I w_i (\mu_m)_i$, $\boldsymbol{HVL}$=0.693/μ(cm).

3. Experimental Work

Analytically, pure grade chemicals were used to prepare the following glass samples according to the formula *:(x* ZnO - $(5-x)$ Bi_2O_3 -10 B_2O_3 -40 SiO_2 mol% where x = 0,

5, 10, 15 and 20. The batch mixtures were melted in porcelain crucibles at 1100 ° for two hours until homogeneous glasses were obtained and then annealed in a separate annealing furnace at 250 °C and then slowly cooled to the room temperature to remove any internal stresses. Samples have been obtained in circular shape of 2 cm. Glass density measurements were measured at room temperature using the standard Archimedes method, with toluene as the immersion fluid of stable density (0.866 g/cm^3).

Attenuation coefficients of the proposed glass system were measured in a narrow beam transmission geometry by using a 2″ × 2″ NaI(TI) crystal detector with energy resolution of 12.5% at 662 keV in conjunction with multi-channel analyzer (MCA). Samples were placed on specimen holder at a distance of 10 cm from source. The distance between source and detector was 20 cm. Radioactive sources ^{60}Co and ^{137}Cs having activities (10 μCi) each were used for different photon energies. Incident and transmitted intensities of photons were measured on MCA for fixed preset time for each sample by selecting a narrow region symmetrical with respect to the centroid of the photo peak. Counting time was chosen such that 10^3-10^5 counts, which were recorded under each photo peak.

The infrared absorption spectra of the glass system were measured at room temperature in the range 2000–400 cm^{-1} by a Fourier Transform infrared spectrometer (type Perken Elmer spectrometer, model RTX, FTIR) using the KBr disc technique.

Optical ultraviolet (UV) –visible absorption measurements, transmission spectra of the prepared glasses in the range 190 to 1100 nm were measured by Genway 6405 -UV-VIS spectrophotometer.

4. Results and Discussion

4.1. Structural Properties

4.1.1. Density (ρ) and Molar Volume

The density is a powerful tool capable of exploring the changes in the structure of glasses. The density is affected by the structural softening/compactness[7], change in geometrical configuration, coordination number, cross-link density and dimension of interstitial spaces of the glass. In the studied glasses, it is noted that Figure 1, the density decreases with increasing ZnO content in the glass due to the replacement of the heavy metal oxide (Bi_2O_3) by lighter oxide (ZnO). So addition of ZnO to network causes some type of structural rearrangement of the atoms. There is a possibility for the alteration of the geometrical configuration upon substitution of ZnO into the glass network. The density value of glass agrees well with that resulted from the present IR. Accordingly, the structure of the studied glasses will not be expanded, but with more compactness and low number of covalent bonds with decrease in the number of bridging oxygens. The more compactness is evident from the decreasing of the packing density as shown in Figure 1 and the decrease in covalent bonds and bridging oxygens are evident from our IR analysis as the Bi_2O_3 content decreases. This is congruent with decrease in the molar volume as the ZnO content increases. So Molar volume is also an important physical property.

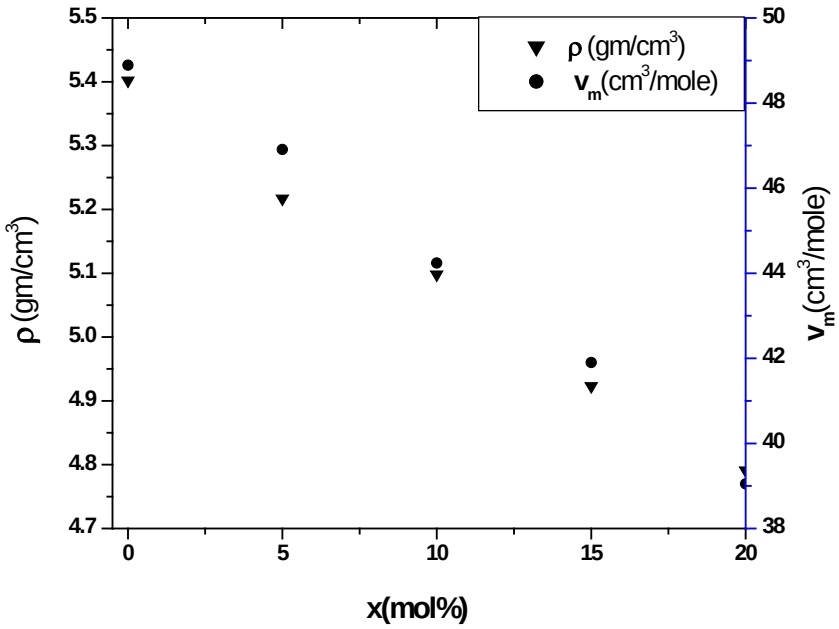

Figure 1. Dependence of the density and the molar volume of on the percentage of ZnO content

4.1.2. FT-IR Absorption Spectra Analysis

Infrared spectroscopy is a valuable tool for studying the glass network structure [8]. In the IR spectra, 2, there are two broad bands that appear around 450 and 1008 cm^{-1}. The broad bands as shown in 2 and Table 1 are an overlapping of some individual bands with each other. Each individual band is related to some type of vibration of a specific structural group. Therefore, it was necessary to use a deconvolution process [9] for separating these individual bands. Each individual band has its characteristic parameters such as its center (***C***), which is related to some type of vibration of a specific structural group, and its relative area (***A***), which is proportional to the concentration of this structural group. The deconvolution parameters of the bands for the investigated glasses are given in Table 1 and Figure 2.

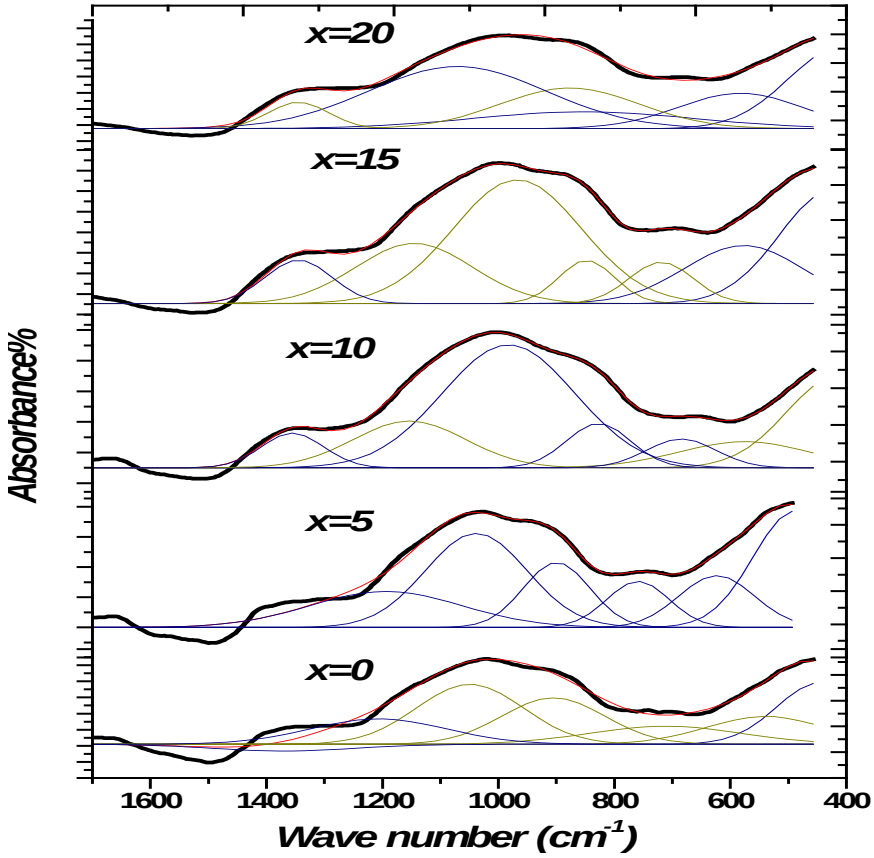

Figure 2. IR spectra of *:(5- x) Bi_2O_3–x ZnO–1 0B_2O_3–40 SiO_2* glasses and their deconvolution, which show the difference between the experimental and simulated curves

Table 1. Deconvolution parameters of the IR spectra of the glass system *:(0.5- x) Bi_2O_3–x ZnO–0.1 B_2O_3–0.4 SiO_2. C* is the component band center and *A* is the relative area (%) of the component band

x	400- 800				800-1200				1200-1600	
	C	A	C	A	C	A	C	A	C	A
0	450				1008					
	447	45	609	13	885	7	1013	13	1368	15
	715	9			1060	8				
5	450				1008					
	447	36	576	18	885	11	940	18		
	715	15			1064	12				
10	450				1028					
	447	33	607	14	885	8	1018	19		
	715	13			1065	11			1375	13
15	450				1008					
	447	33	609	17	885	9	981	31	1364	12
	721	11			1070	11				
20	440				1008					
	447	33	608	17	885	4	983	32	1376	11
	725	9			1075	14				

The IR analysis of the glass system indicates that the band around 450 cm^{-1}, which appears in all glasses, is due to overlapping of Bi–O bending vibrations in BiO_6 octahedral units and Si-O-Si bending in SiO_4 tetrahedra [4,10]. The Intensity of this band is decreasing with increasing the contents of ZnO because of decrease in BiO_6 group. At the same time, a new band has been observed at 576cm^{-1} in sample with 5 ZnO which is due to the presence of Zn^{+2} ions which act as modifier in ZnO_4 units overlapping with the vibrations of the BiO_6 octahedral unit [4] in a location with the ring structure of SiO_2 [10,11]. The absorption band in the region 610–623 cm^{-1} may be due to Bi- O^- stretching vibrations (non-bridging oxygen in BiO_6 units. The band at 699–712 cm^{-1} is assigned to the bending vibrations of B–O linkages in the borate network [4,12]. A weak band has been observed at 885 cm^{-1} which is due to non bridging oxygens of BO4 groups overlapping with the symmetrical stretching vibrations of the Bi–O bond in the BiO_3, ZnO bond stretching of the tetrahedral ZnO_4 units and Si- O^- stretching with non-bridging oxygen [4,11,12]. So increasing the contents of ZnO, help for conversion of BO_3 to BO_4 groups and extra oxygen's from Bi_2O_3 helps to form ZnO_4 units in glass. This reveals the decrease of BiO_3 units and the formation of more tetrahedral BO_4 and ZnO_4 groups. The band in the region 1006–1028 cm^{-1} is overlapping the symmetric stretching vibration of SiO_4, BO_4 tetrahedral units [4,11]. The band in the region 1060–1075 cm^{-1} is attributed to the the Si- O- Si anti symmetric stretching vibrations of bridging oxygen in tetrahedra and BO_4 units [13]. Upon increasing the zinc content the band at about 1060 cm^{-1} shifts to higher wave number 1075cm^{-1}. The increasing of intensity of this band indicates an increase of BO_4 groups. The band present around 1370cm^{-1} is due to stretching and bending vibration BO_3 groups [14]. The Spectra reveal that the intensity of the band around 1370 cm^{-1} decreases and corresponding to it, the intensity of the band between 800 and 1200 cm^{-1} increases. This reveals that an addition of zinc modifies the glass network.

4.2. Gamma-ray Shielding Properties

The experimental values of mass attenuation coefficients along with theoretical values are shown in Figure 3. A good agreement has been observed between experimental and theoretical values and the discrepancies are within the experimental errors. The results of mass attenuation coefficients decrease with the mol% of ZnO which may be due to the decrease in the weight fraction of higher atomic number constituent (Bi) as compared to other elements. The mass attenuation coefficients for 5 to 15 ZnO content is nearly linear, This is due to the presence of Zn^{+2} ions in glass structure which modifies the borosilicate glass network by converting more BO_3 to BO_4 where extra oxygen's from Bi_2O_3 helps to form ZnO_4 units in glass. This reveals the decrease of BiO_3 units and the formation of more tetrahedral BO_4 and ZnO_4 groups.

HVL is used to describe the effectiveness of γ-ray shielding [4]. Figure 4 show the behavior of the HVL for glasses with different amounts of ZnO and different γ-energies. This figure indicates that the half value layer (HVL) increases with decreasing weight fractions of Bi_2O_3 in this glass system. This is due to the lower values of mass attenuation coefficients and densities for glass samples. For all glass system higher γ-ray energies need more HVL of glasses.

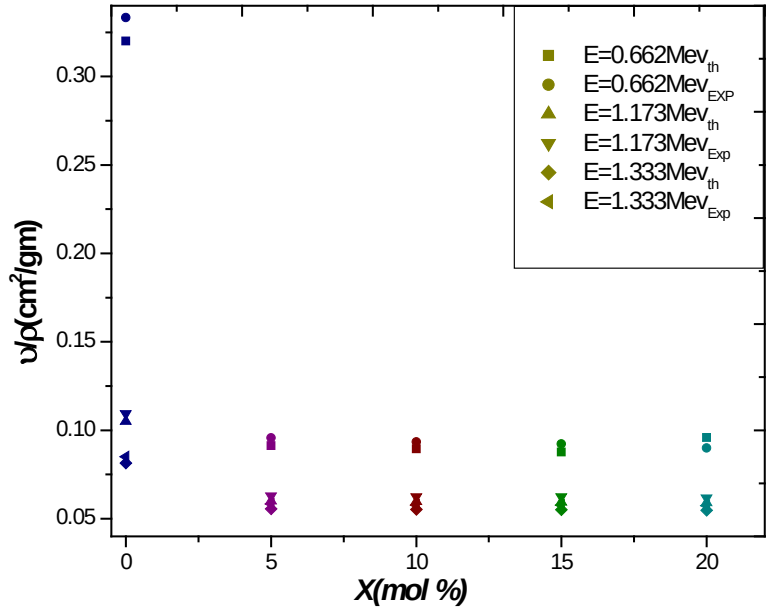

Figure 3. Experimental and theoretical values of mass attenuation coefficients (μ_m) as function of mol% of ZnO in the glass system at 662, 1173 and 1332 keV

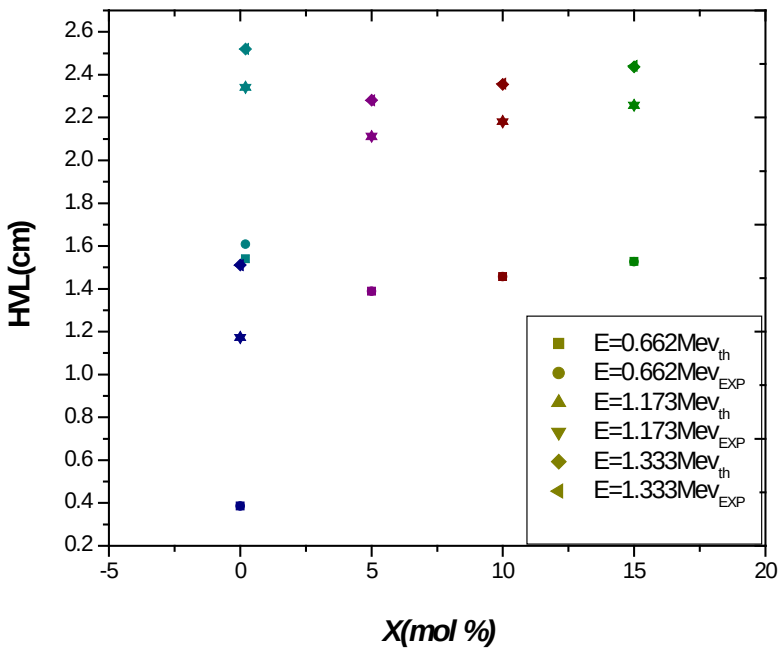

Figure 4. HVL function of mol% of ZnO in the glass system at 662, 1173 and 1332 keV

4.3. UV–Visible Transmission Spectra

Spectra of all prepared samples follow one common pattern, where an edge extended over wide wavelength range is observed Figure 5. This is in consistence with the non-crystalline nature of the glasses. The cut-off wavelength (λ_c) exhibit red shift with increasing ZnO content. Such behavior can be accounted for by increasing

the number of non-bridging oxygen. The same behavior was observed by increasing ZnO content in bismuth borate glass [15]. A shoulder at 663 nm was observed and broadened with increasing ZnO content. It is evident from the figure that, the percentage of transmission decreases with increasing ZnO content except the sample with x = 5 ZnO which shows high percentage of transmittance in the visible region of the spectra (656 nm to 998 nm) Furthermore, sample with 20 ZnO content show a strong absorption in the UV region up to 530 nm. From IR results it can explain the transmission by the overlapping of the vibrations of the Bi–O bonds in the BiO_6 octahedral unit with deformation modes of network structure and vibration of chemical bond Zn^{2+} - O^{2-} in a location of with the ring structure of SiO_2 at 575 cm^{-1}. So x=0.05 make this concentration is more transparent than others.

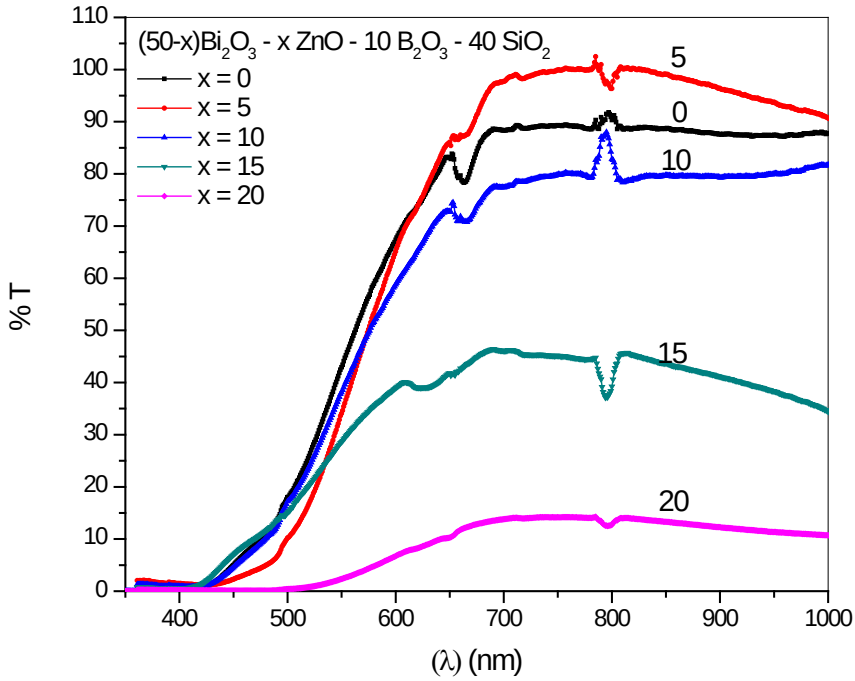

Figure 5. The optical Transmission as a function of wavelength in the UV region of the prepared glasses

5. Conclusion

The results of ZnO doped in bismuth borosilicate glass samples with chemical composition *:(5- **x**) Bi_2O_3–**x** ZnO–10 B_2O_3–40 SiO_2* proved that the density, molar volume and mass attenuation coefficient of glass samples were decreased along with increasing the concentration of ZnO, due to high molecular weight of Bi_2O_3 compared with ZnO. The FTIR spectra supported also the structural changes with composition. The UV–VIS transmission spectra of these glasses show that the system is transparent especially at concentration with ZnO =5 due to the formation of Zn^{2+} - O^{2-}. So small amount of ZnO (~5%) is necessary to make the above glass system is transparent and good attenuating to γ-rays at the same time.

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