# INFLUENCE OF SILVER OXIDE ON STRUCTURAL, PHYSICAL, ELASTIC AND OPTICAL PROPERTIES OF ZINC TELLURITE GLASS SYSTEM FOR OPTICAL APPLICATION

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The research on several properties of erbium-doped zinc tellurite glass system is thoroughly investigated recently. However, the introduction of silver oxide as a dopant plays a role on the fabrication of new optical materials specifically in optical application. By conducting the investigation, the use of melt-quenching technique is one of the techniques to fabricate a series of silver-doped zinc tellurite glass systems with the chemical composition of  $\left[\left[(TeO_2)_{0.7}(ZnO)_{0.3}\right]_{0.96}(Er_2O_3)_{0.04}\right]_{1-x}(Ag_2O)_x$  at different molar fraction of x = 0.01, 0.02, 0.03, 0.04 and 0.05. The characterization of X-ray diffraction (XRD), Fourier Transform Infrared (FTIR) spectroscopy, density, elastic and optical measurements were investigated to analyse the prepared glass samples. The result of the XRD, FTIR, density, elastic and optical properties were all reported at the room temperature. The glass samples are proven as amorphous in nature which can be seen by the XRD spectra. Besides that, the presence of functional vibration of tellurite network can be clearly observed through the analysis of FTIR spectra. The density of the glass system is found to increase with the increase of silver oxide concentration. Anyhow, the value of molar volume is noticed to be inversely proportional to the density. Therefore, the inclination in the density value causes the decrement of the molar volume which can be associated with the inclusion of silver oxide in the glass system. Plus, this also follows the theoretical formula where the density is equivalent to molar mass per molar volume. In the meantime, the ultrasonic velocity was employed in order to specify the elastic moduli of the glass systems. The elastic moduli for instance longitudinal modulus, shear modulus, bulk modulus, Young's modulus as well as other parameters come out with a varying trend against the concentration of silver oxide. The increment of the elastic moduli can be attributed to the high connectivity and rigidity of the glass structure. Nonetheless, the decrement of the elastic moduli can be associated with the breakage of bonds within the zinc tellurite glass system which impair the structure of the glass sample. The UV-Vis analysis can determine the optical properties of the prepared glass samples. The optical absorption was reported at room temperature within the wavelength ranging from 200 to 2000 nm. The optical absorption spectra unveil the fundamental absorption edge shifts to longer wavelength as the content of silver oxide increases. The values of direct and indirect band gap and metallization criterion have been evaluated and noticed to have a decreasing trend with the increasing content of silver oxide whereas the Urbach energy, refractive index, molar refraction, molar polarizability, oxide ion polarizability and optical basicity are obviously increasing as the concentration is included into the glass system.

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### 1. Introduction

Glass can be defined as an isotropic material with huge flexibility in terms of chemical composition [1]. Tellurite glasses are broadly investigated in current years due to their promising potentials in assorted of applications. The glasses are highly recommended especially in lasers,

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sensors, optical fibers, solar cells and non-linear optical applications, with suitable dopants of both rare earth and heavy metal ions doping [2].

In the field of glass technology, tellurium oxide  $(\text{TeO}_2)$  can be defined as a conditional glass former which is employed due to its low melting point, low phonon maxima and high refractive index. Good glass dopants usually come from the rare earth and heavy metal elements which have tendency to form glass easily [3]. Tellurium oxide acts as one of the most stable oxide with a melting point of 733 °C. Tellurium oxide has captivated the attentions of several researchers due to its potentials which act as good candidate for photonic and optical application. Peng et al., (2015) had stated that the tellurium oxide is a prospective candidate that applicable for a visible-band up-conversion fiber laser [4]. In addition, tellurium oxide is also interchangeable to the other glass derivatives which allow the inclusion of other compositions into the tellurite glass system at the same time. The relevance of these outstanding characteristics for the tellurite glass system is; it is recognized as an excellent material that displays slow crystallization rates, good thermal and chemical stability and low cut off phonon energy which is appropriate for the spontaneous emission [5].

Besides that, zinc oxide (ZnO) is an element that is conceived as a white powder. It is insoluble in water and acts as one of the inorganic compounds. ZnO has been widely used as an additive to some materials and products such as plastic, ceramic, glass, cement, lubricants and rubber [6]. ZnO is also applicable in semiconductor industry due to its miscellaneous desirable properties such as high electron mobility, wide band gap, strong room temperature luminescence and good transparency [7]. Based on these properties, ZnO has been utilized in transparent electrodes manufacture especially in liquid crystal displays, for energy saving or heat-protecting windows, in electronics which behaves as a thin-film transistors [8] and act as an emitting diode as well [9]. In addition, the inclusion of ZnO in the tellurite glass network also can minimize the rates of crystallization, decrease the melting point and elevate the glass forming ability [10,11,12]. The existence of ZnO in the glass formation can reduce the optical energy band gap, incline the refractive index [11] and also act as a glass stabilizer [10]. Zinc tellurite glass also has been utilized as a basis for multi-component optical glass and as a useful medium for ultra-low loss optical fibers. After all, ZnO can be deduced as a good candidate for super heavy optical flint glass [13,14].

On the other hand, erbium oxide is one of the excellent glasses doping out of all the rare elements which is commonly used in communication technology for optical signal amplification through the erbium doped fiber amplifier (EDFA). The leverage of erbium ions among the rare earth elements has drawn the attention in the field of glass science and technology due to its excellence properties as doping element in many types of glass applications in the current years [2]. A lot of researchers in the glass field have investigated several works on the influence of  $\text{Er}^{3+}$  ions on certain properties such as optical and mechanical properties. In order to improve the optical properties of tellurite glasses, erbium oxide is used as a dopant due to its unique optical properties. Erbium oxide is one of the rare earth materials that have been used comprehensively in optical and photonic applications. The lengthy study had been elucidated in order to create new laser amplifiers with an inclusion of erbium oxide. Bendamani et al., (2013) had explained that  $\text{Er}^{3+}$  can be convenient by applying the manufacture of laser amplifiers with an excellent performance compared to the other laser amplifiers [15]. Hence, erbium oxide doped with tellurite glass system will promote a good potential especially in optical applications.

Silver oxide or  $(Ag_2O)$  is another heavy metal element that is good and suitable for the preparation of glass-based materials. Silver oxide  $(Ag_2O)$  also has achieved a peculiar position out of the dopants in order to obtain better glassy materials with exact prescribed properties. Hence, the doping of tellurite glasses with silver oxide has driven the attraction towards the scientific society especially in photonics and optoelectronics application. Silver-doped glasses are appropriate materials that are useful in every single aspect such as structural, physical and chemical properties. It forms a variety of attractive characteristics which contribute to multiple of advantages. The advantages include a plenty of applications which enhance a few phenomenon's' to occur such as the silver-glass interaction, diffusion of silver into the glass, and optical response [16].

Despite plenty of researches have been executed on the fabrication and characterization towards zinc tellurite glass system, the employment of erbium oxide and silver oxide has never been studied. The measurement on structural, physical, elastic and optical properties of the glass system also have never been declared. Therefore, via this research, a glass system with the chemical composition of [[(TeO<sub>2</sub>)<sub>0.7</sub>(ZnO)<sub>0.3</sub>]<sub>0.96</sub> (Er<sub>2</sub>O<sub>3</sub>)<sub>0.04</sub>]] <sub>1-x</sub> (Ag<sub>2</sub>O) <sub>x</sub> at different molar fraction of x = 0.01, 0.02, 0.03, 0.04 and 0.05 were identified and characterized in terms of structural and physical properties; and the effect of doping of erbium ion and silver ion on elastic and optical properties of zinc tellurite glass system then was elucidated.

## 2. Materials and methods

A series of silver-doped zinc tellurite glass systems were formed from oxide powders of tellurium (IV) oxide, TeO<sub>2</sub> (Aldrich 99.5%), zinc oxide, ZnO (Alfa Aesar 99.99%), erbium (III) oxide,  $Er_2O_3$  (Alfa Aesar, 99.9%) and  $Ag_2O$  (Alfa Aesar, 99.9+%) as raw materials by utilizing the conventional melt-quenching method. Variation amount of concentration of x = 0.01, 0.02, 0.03, 0.04 and 0.05 were employed to synthesize the glass systems. All needed amounts of raw material were mixed together.

The mixture of all the chemical powders was weighed by using an electronic weighing balance machine with an accuracy of  $\pm 0.0001$  g. The amount of chemical elements included in every composition of the glass samples are tabulated in Table 1. The entire chemical with a weight of 11 g were completely blended and put into the alumina crucible. The glass rod, spatula and alumina crucible were cleaned beforehand by using distllied water and acetone in order to get rid of the impurities. Next, the mixture was constantly stirred for about 30 minutes to form a homogenous mixture.

Molar fraction,	Weight of TeO <sub>2</sub>	Weight of ZnO	Weight of	Weight of	Total weight
Х	(±0.0001 g)	(±0.0001 g)	$Er_2O_3(\pm 0.0001)$	$Ag_2O(\pm 0.0001$	(g)
			g)	g)	
0.01	7.9536	1.7381	1.1347	0.1736	11
0.02	7.8276	1.7105	1.1167	0.3452	11
0.03	7.7030	1.6833	1.0989	0.5148	11
0.04	7.5798	1.6564	1.0814	0.6824	11
0.05	7.4581	1.6298	1.0640	0.8481	11

Table 1. Amounts of each chemical used for silver-doped zinc tellurite glass system.

The homogenous mixture in the alumina crucible was placed in the first furnace, to be heated at a temperature of 400 °C, and kept at this temperature for one hour in order to get rid of the water content in the mixture which will affect the final result. After one hour, the crucible was then moved to the second furnace for two hours at 900 °C. The mixture in the crucible was melted in the furnace. A stainless steel cylindrical shape split was used to mould the raw material which had been polished earlier in order to prevent the material from reacting with impurities. The stainless steel mould was also previously preheated at 400 °C in order to avoid thermal shock as a result of the temperature difference between the molten mixture and the mould. This process and melting process were done concurrently at the same time.

The molten mixture that had been poured into the mould was annealed at 400 °C for two hours. The purpose of the annealing process is to remove the formation of air bubbles, reduce thermal stress and enhance the mechanical stress. The furnace was then switched off and the glass sample was allowed to cool down in the furnace at room temperature. The glass samples were taken out from the furnace and kept in the bottle with silica gel to absorb any moisture.

The surfaces of the glass samples were polished by using SiC abrasive of grade 800, 1000, 1200, 2400 and 4000 in ascending order until a parallel, flat and smooth surface of the glass systems were obtained. A vernier caliper was used to measure the thickness of glass sample and to

determine the thickness suitability for elasticity measurement which should be approximately 5.0 mm. On top of that, another prepared glass sample was measured around 2.3 mm in order to achieve an excellent optical measurement. Some parts of the glass samples were ground into powder by using the plunger. Then, the glass samples were sent for structural and physical analysis.

Measurement	Uncertainties
X-ray Diffraction (XRD)	$2\theta < 1.5\%$
Fourier Transform Infrared (FTIR)	$\pm 10 \text{ cm}^{-1}$
Density	$\pm 0.001 \text{ g/cm}^3$
Ultrasonic velocities	$\pm 5 \text{ m/s}$
UV-Vis Spectrophotometer	±0.3 nm

Table 2. Uncertainties for each measurement in this work.

Table 2 shows the uncertainties of the devices used for each measurement in this work.

## 3. Results and discussions

#### 3.1 X-ray diffraction

X-ray diffraction (XRD) analysis was employed in order to determine the structural arrangement of the prepared glass sample either it is non-crystalline or crystalline phase which occur within the glass system. Fig. 1 shows the graph of XRD diffraction patterns of the prepared glass sample at different concentration of silver-doped zinc tellurite glass systems. The spectra are recorded at room temperature in the range of  $20^{\circ} \le \theta \le 80^{\circ}$ . The result of the XRD spectra for this glass series display broad diffusion at lower scattering angles proposing the presence of lack long range structural order which is displayed around  $2\theta = 30^{\circ}$ . The absence of sharp peak ratifies the non-existence of the crystalline phases [12]. Thus, the broad peaks attained from the XRD spectra affirm that the glass series are completely amorphous in nature [17]. Cai et al., (2016) and Huang et al., (2014) had also reported that the non-existence of long range atomic arrangement in the glass system proves the amorphous nature of the glass samples.



Fig. 1. XRD diffraction patterns of the prepared glass sample at different concentration of silver oxide.

#### **3.2 Fourier transform infrared spectroscopy**

Fourier transform infrared (FTIR) is one of a non-destructive method which provides the information regarding the structure and vibrational properties of the elements that exist in the glass system. Nanda et al., (2015a) had also reported that FTIR is one of the methods that can be used in determining the functional group of the elements and provide the information about the vibrational modes of the molecules which exist in the disordered amorphous materials. The FTIR spectra attained for the glass series fall in the range of 611-616 cm<sup>-1</sup> which is presented in Fig. 2. The assignments of the transmission spectra of this glass series are tabulated in Table 3. The band's positions are attributed to the tellurite network [22].

No	0.01	0.02	0.03	0.04	0.05	Assignments
	$(\pm 10 \text{ cm}^{-1})$	$(\pm 10 \text{ cm}^{-1})$	$(\pm 10 \text{ cm}^{-1})$	$(\pm 10 \text{ cm}^{-1})$	$(\pm 10 \text{ cm}^{-1})$	
1	611-616	611-616	611-616	611-616	611-616	TeO <sub>4</sub> group
						exists in all
						tellurite network
						[20]
		120 100 80 60 20 250	0.01 mol Ag 0.04 mol Ag	0.02 mol Ag 0.0 0.05 mol Ag	13 mol Ag	

 Table 3. Assignment of infrared transmission bands of the prepared glass sample at different concentration of silver oxide.

Fig. 2. FTIR spectra of the prepared glass sample at different concentration of silver oxide.

The transmission band for pure tellurium oxide is positioned at 640 cm<sup>-1</sup>. Nevertheless, tellurium oxide subsists of two different types of functional groups which are trigonal pyramid (TeO<sub>3</sub>) and trigonal bipyramid (TeO<sub>4</sub>) [23]. The functional groups of trigonal pyramid correspond to the Te-non-bridging oxygen atoms while trigonal bipyramid corresponds to Te-bridging oxygen atom. The transmission band that lies in the range of 611-616 cm<sup>-1</sup> resembles the trigonal bipyramid group [24,25]. The variation of the composition in the glass network might also affect the shifting of the functional group's that presence in the disorder of amorphous material [23].

Meanwhile, zinc oxide (ZnO) is not found in the transmission band of the glass system. The zinc lattice in the glass system is said to be broken down which lead to the absence of zinc oxide as an additional functional group. However, at the early stage of the formation of glass, zinc oxide which act as a glass former will break down the Te-O-Te that subsequently will form coordination effects known as dangling bonds (non-bridging oxygen) (Te-O<sup>-</sup>...Zn<sup>2+</sup>...O-Te). Indirectly, this will reduce a little amount of trigonal bipyramid concentration in the glass system [26].

#### **3.3 Density and molar volume**

Density can be defined as the structural compactness as well as the strength of the amorphous materials within the glass system [27,28,29]. Pavani et al., (2012) had stated that there is a change in geometrical configuration, coordination number, cross-link density, interstices

spaces and compactness in the glass system. Densities of the prepared glass sample were evaluated using Archimedes' principle at room temperature with distilled water as an immersion liquid. Besides that, Pavani et al., (2012) had also mentioned that the density is about how tightly the ions and the ionic groups are packed together in the disordered glassy materials. Fig. 3 shows the relationship between the density and molar volume with respect to the molar fraction of silver oxide. Based on the results obtained, as the concentration of  $Ag_2O$  increases, the density of the glass samples also increases which can be related to the effect of  $Ag_2O$  in the glass system.

Molar fraction, x	Density	Molar volume
	$(\pm 0.001 \text{ g/cm}^3)$	$(\pm 0.001 \text{ cm}^3/\text{mol})$
0.01	4.467	32.871
0.02	4.578	32.267
0.03	4.714	31.516
0.04	4.822	30.987
0.05	4.971	30.324
5.0 4.9 4.7 4.7 4.7 4.6 4.5		33.5 33.0 (Jul) 32.5 32.0 31.5 Jul) 31.0 Jul) 30.5

Table 4. Density and molar volume of silver-doped zinc tellurite glass system.



0.03

0.04

0.02

0.01

30.0

0.05

The atomic weight plays an important role towards the increment of the density in the glass samples. As the concentration of silver oxide increases, the glass sample becomes more compact until there is no expansion of free volume in the glass system. In addition, there is also an effect of silver oxide in the glass system whereby the atomic weight of silver oxide is 231.74 g which is larger than tellurium oxide (159.60 g). It can be said that the presence of silver oxide in the quaternary elements of the glass system has eventually affected the density of the glass to be elevated. Noranizaha et al., (2014) had mentioned that the difference of density also might transform the structural arrangement in the glass system whereby in this work, the structural arrangement is in tightly packed condition. This can be proven by following the theoretical relationship where the decrement of molar volume is affected by a small value of molecular weight and increase in density. According to Saddeek (2004), he had also mentioned that the trend of this quaternary glass system obeys the theoretical relationship where the density and molar volume display opposite behaviour. The other reason for the decreasing trend of molar volume is the atomic radius of silver which is only 172 pm that is shorter than Te and Er which are 210 pm and 232 pm respectively. This can reduce the free volume in the glass network. The variation of density can be correlated to the difference in atomic mass of the constituents' elements [32]. The inclusion of silver oxide in the glass sample makes the glass system to become more compact and denser where the combinations of all the elements in the glass system lead the glass system to become heavier. Coelho et al., (2012) had stated that the decrement of molar volume also leads to the creation of bridging oxygen (BOs). The presence of bridging oxygen decreases the free space

in the network and makes the glass structure to become more compact in structure. This condition can be taken into consideration where there is also no element fill up within the free space region and therefore can improve the compactness of the glass system [17].

#### **3.4. Elastic properties**

Kannapan et al., (2009) and Gupta et al., (2014) had reported that the ultrasonic technique is one of the methods that is used to determine the structure of the vitreous network which is correlated to the interatomic potentials within the glass network. It can also be used to identify the structural changes in the glass network and study the elastic constant of the investigated glass sample [36]. The effect of dopant which is silver-doped glass series has been determined by using ultrasonic velocities and densities. The values of ultrasonic velocities and densities of the glasses can determine the elastic moduli which comprise the longitudinal modulus (L), Shear modulus (S), Bulk modulus (K), Young's modulus (E) and followed by other elastic parameters.

Table 5. Elastic moduli, Poisson's ratio, micro-hardness, Debye temperature, mean velocity, acoustic impedance, softening temperature and fractal bond connectivity of  $[[(TeO_2)_{0.7}(ZnO)_{0.3}]_{0.96} (Er_2O_3)_{0.04}]]_{1-x}$ (Ag<sub>2</sub>O) <sub>x</sub> at different molar fraction of silver oxide.

Elastic	0.01	0.02	0.03	0.04	0.05
parameter					
L (GPa)	54.67	61.11	51.63	49.25	64.58
G (GPa)	19.48	19.98	19.00	18.79	19.02
K (GPa)	28.69	34.47	26.29	24.20	39.22
E (GPa)	47.66	50.24	45.94	44.78	49.12
σ	0.2232	0.2571	0.2088	0.1915	0.2913
H (GPa)	3.60	3.24	3.69	3.87	2.65
$\theta_{\rm D}(^{\circ}{\rm C})$	224.43	252.92	244.67	242.86	253.53
$v_{mean}$ (m/s)	1987.48	2022.27	1941.07	1915.80	1983.72
$Z(x10^7 \text{ kg/m}^2\text{s})$	1.43	1.54	1.45	1.44	1.71
$T_s(K)$	594.32	606.68	578.04	571.58	581.32
d	2.716	2.319	2.891	3.106	1.940



Fig. 4. Variation in longitudinal velocity  $(v_L)$  and shear velocity  $(v_S)$  with respect to  $Ag_2O$  content at room temperature.

The changes in ultrasonic velocity which consists of longitudinal and shear velocity with an increment of dopant concentration are classified in Table 5 and illustrated in Figure 4. As observed from Fig. 4, both ultrasonic velocities for silver-doped glass series exhibit a similar pattern. Both longitudinal and shear velocities increase from 0.01 to 0.02 mol, decrease from 0.02 to 0.04 mol and increase from 0.04 to 0.05 mol as the dopant increases.

The presence of silver oxide proves that this element acts as a glass modifier which modifies the glass structure in the glass system. The decrement of both longitudinal and shear velocities is correlated to the existence of dopant (silver oxide) in the glass system. Saddeek (2004) had mentioned that the inclusion of silver oxide as a fourth component into a quaternary element will modify the glass structure which will split the Te-O-Te bond. This phenomenon promotes the conversion of bridging oxygen (trigonal bipyramid) to non-bridging oxygen (trigonal pyramid) or vice versa. An addition of silver oxide into the glass interstices enables more open ions to open up which will weaken the glass structure. Obliquely, the rigidity and strength of the glass structure also will degrade. However, at certain range, both ultrasonic velocities also increase. This can be linked to the formation of bridging oxygen which is compatible towards the presence of trigonal bipyramid as proven from the FTIR result. The atomic arrangement in the glass sample will become more compact with an enhancement of rigidity as well as the strength of the glass sample.

The elastic moduli and other elastic parameters of the prepared glasses are evaluated and classified in Table 5. The change of elastic moduli with respect to the molar fraction of silver oxide is also illustrated in figures.



Fig. 5. Variation in elastic moduli (GPa) with respect to Ag<sub>2</sub>O content at room temperature.

All the elastic moduli display a fluctuating trend as the molar fraction of silver oxide is added into the glass system. Afifi et al., (2003) and Balaji et al., (2014) had reported that the increasing trend of the elastic moduli signifies the strengthening of the glass network as well as an improvement of the rigidity in the glass system. In addition, Kannapan et al., (2009) had also proposed that this phenomenon is also correlated to the increase in packing density which is attributed to the revolution in coordination number of tellurite network that leads to the conversion of trigonal pyramid to trigonal bipyramid.

There is a point where the graph exerts a declination trend as well. The steady decrease also seems to occur as the concentration of silver oxide increase. The decrement of the elastic moduli is due to the presence of non-bridging oxygen with the increase of more open structure. Consequently, the presence of silver oxide which acts as glass modifier will break the glass network structure. Sekiya et al., (1992b) also had reported that the primary structural units of tellurite glass are having high number of tellurium oxide content which mostly composed of trigonal bipyramid at which one of the equatorial sites filled up by a lone pair of electron and most of the tellurium atoms are linked at the vertices of Te-O-Te linkage. Hoppe et al., (2004) had also proposed that the inclusion of alkali oxide modifier in the tellurite glass make the connectivity of the Te-O<sub>eq</sub> and Te-O<sub>ax</sub> loosen up where the existence of trigonal bipyramid network will break down the glass network and form a non-bridging atom in both Te-O<sub>eq</sub> and Te<sub>ax</sub> bonds. The existence of silver oxide in the glass structure will break down the network structure which then leads to the decrement of the rigidity of the glass as well as reduce both ultrasonic velocities. Saddeek (2004) had proposed that the drop in rigidity contributes to a decrement in ultrasonic velocities and elevation of Poisson's ratio at the same time.



Fig. 6. Variation in Poisson's ratio with respect to  $Ag_2O$  content at room temperature.

Poisson's ratio ( $\sigma$ ) can be identified as a measurement of material's resistance towards the change in volume and shape concurrently [41,42]. For a shear-resistance material, the ratio tends to be little while for incompressible material such as rubber, the value tends to reach up to 0.5. In the prospect of glass, the value generally lies in the range of 0.1-0.4, relying upon the polymerization degree of the glass network inside the glass system. Normally, high cross-link network will produce lower Poisson's ratio or with the contrarily.

The graph of Poisson's ratio with respect to the dopant concentration is illustrated in Fig. 6. The Poisson's ratio for silver-doped glass series in this research is measured at different molar fraction. The results show that the Poisson's ratio fluctuates with the various concentration of the dopant. This parameter usually can be correlated to the cross-link density of the glass system.

The ratio that belongs to the range of 0.1 to 0.2 signifies that the glass samples have large cross-link density. Meanwhile, the ratio that exists in the range of 0.3 to 0.5 indicates that the glass systems tend to have small cross link-density [38]. The observation made by Chang et al., (2003) had justified that the relationship of the Poisson's ratio towards the deformation of the network structure that happens inside the glass system. The values which are equivalent to 0.25 define that the glass is deformed by compressing or stretching its structural units, whereas the values which is lower than 0.25 indicate the presence of any energy in the glass system which will alter its structural units via bond bending. Nevertheless, the Poisson's ratio which remains at more than 0.25 can be considered as the ions themselves are deformed under stress with the influence of network distortion. Shelby (2005) had also reported that the Poisson's ratio for the oxide glasses normally lie in the range of 0.2 to 0.3. In the current investigation, the values of Poisson's ratio of both glass series exist in the range of 0.1915 to 0.2913. This range of Poisson's ratio of the glass sample can be considered as they have both large and small cross-link density where the ions inside the glass network are deformed as stress is employed and the structural units will be altered due to an existence of energy in the glass system. In the meantime, Gaafar et al., (2009) had mentioned that the dimensionality of the glass sample is one of the components that can affect the difference of the Poisson's ratio. It can be evaluated by using fractal bond connectivity (d) that is equivalent to 4G/Ke [45]. As d increases,  $\sigma$  will decrease since the number of bonds resisting a transverse deformation also increases.



Fig. 7. Variation in micro-hardness with respect to Ag<sub>2</sub>O content at room temperature.

Micro-hardness of a glass sample can be defined as a quantity of stress required in order to remove the free volume inside the glass system. It can be evaluated by using the Poisson's ratio and Young's modulus information.

In a glass system of silver-doped glass series, the declination of the micro-hardness is due to the distortion of structural unit arrangement where there is a huge possibility for a conversion of trigonal bipyramid to trigonal pyramid or vice versa. This is also correlating towards the number of non-bridging and bridging oxygen [46]. Rajendran et al., (2002) had also mentioned that a plenty of non-bridging and bridging oxygen play a crucial role in manipulating the Young's modulus, micro-hardness, fracture toughness as well as fracture surface energy. An increase in micro-hardness can be attributed to the existence of bridging oxygen which will strengthen the Te-O bond in the glass structure. The inclusion of silver oxide in the zinc tellurite glass system will also capable to produce non-bridging and bridging oxygen which will affect the value of micro-hardness in the glass system.



Fig. 8. Variation in Debye temperature with respect to  $Ag_2O$  content at room temperature.

The determination of Debye temperature is when the temperature of all high-frequency of the lattice vibrational modes is excited. It can be employed to investigate the characteristics of the solid materials which are correlated to the atomic vibrations that are present in the glass network [36,48,49].

The declination of the Debye temperature can be supported by the inclusion of silver oxide in the glass system which will produce non-bridging oxygen and lead to the loose packing structure in the vibration of the lattice. The formation of triangular pyramid from triangular bipyramid promotes the presence of non-bridging oxygen with more inclusion of silver oxide in which the covalent bond of the atoms will become weaker. This phenomenon will make the glass less rigid and affect the Debye temperature at the same time. However, the increment of Debye temperature can be a reason towards the strengthening of the glass structure in which the conversion of trigonal pyramid to trigonal bipyramid is occurred. This creates more bridging oxygen in the glass atoms [50]. Other than that, the increment in Debye temperature also promotes the elevation of the rigidity in the glass structure. This is relatable to the inclination of lattice vibration of the glass network [48].



Fig. 9. Variation in acoustic impedance with respect to  $Ag_2O$  content at room temperature.

Acoustic impedance can be determined by the transmission and reflectance measurement of the sound energy in the glass specimen. It is reliable on the sound velocity and also the density of the material.

Fig. 9 shows the fluctuation of the acoustic impedance as the dopant increases. At point where the acoustic impedance increases, the glass samples become more rigid and have strong connection within the glass network due to the increasing number of bond per unit volume. Other than that, the denser glass sample will also affect the compactness of the glass sample which will produce larger impedance to the ultrasonic waves. This circumstance can be a reason that a denser material tends to have large molecular weight. This can lead to a huge inertia and massive resistance in order to change its state of motion.

However, the decrement of acoustic impedance is due to the capability of the ultrasonic wave to propagate. This can be emphasized that a material with low density that comes from low molecular weight will result in a small inertia of the molecules to occur in the glass system. Hence, it is easier for the ultrasonic wave to vibrate the atoms and molecules yet the acoustic impedance will get affected and eventually decrease.



Fig. 10. Variation in softening temperature with respect to  $Ag_2O$  content at room temperature.

Softening temperature is one of the elastic parameter that is able to determine the changes of viscous flow to plastic flow. This parameter is crucial in order to identify the temperature stability of the glass system. Usually, massive softening temperature of the glass promotes better stability in the elastic properties of the glass sample.

In the investigation, the decrement of softening temperature with an inclusion of silver oxide can be attributed to the less rigidity of the glass sample. This can be supported by the presence of non-bridging oxygen in the glass system which requires less energy to break the bond in the glass network [51].

The increment of the softening temperature can be attributed to the presence of trigonal bipyramid which leads to the presence of bridging oxygen. At the points where the increment of softening temperature starts to occur, it can cause strong connectivity of the glass network and improve the rigidity of the glass sample as well [52].

### 3.5. Optical properties

The investigation of optical absorption spectra gives some information regarding the band gap and Urbach energy of crystalline or even disordered glassy materials. It also contributes the information of atomic vibration and electronic states of atoms of the materials [53]. The optical absorption of the prepared glass system was attained by employing UV-Visible spectroscopy within the wavelength of 200-2000 nm at room temperature. The outcome of the glass series is presented in Figure 11. It is noticed that the absorption edge of the glass series is not sharply defined due to the non-crystalline nature [23]. It can be seen clearly that absorption edge shifts to longer wavelength as the concentration of silver oxide is increasing.



Fig. 11. UV-Visible absorption spectra with respect to  $Ag_2O$  content at room temperature.

Table 6. Direct energy band gap,  $E1_{opb}$  Indirect energy band gap,  $E2_{opt}$  and Urbach energy,  $\Delta E$  of silver-doped zinc tellurite glass system.

Molar fraction, x	Direct energy band gap,	Indirect energy band	Urbach Energy, $\Delta E$
	$E1_{opt}$	gap, E2 <sub>opt</sub>	(±0.3 eV)
	(±0.3 eV)	(±0.3 eV)	
0.01	3.505	3.362	0.1884
0.02	3.495	3.235	0.1556
0.03	3.390	3.181	0.3500
0.04	3.371	3.141	0.3889
0.05	3.231	3.015	1.000

Molar fraction, x	Refractive index, n	Molar refraction, Rm	Molar polarizability, $\alpha_m$ (Å <sup>3</sup> )	Oxide ion polarizability, $\alpha_0^{2-}$	Optical basicity, $\Lambda$	Metallization crtiterion, M
0.01	2.306	19.40	7.70	3.629	1.210	0.410
0.02	2.339	19.31	7.66	3.604	1.207	0.402
0.03	2.349	18.94	7.52	3.515	1.195	0.399
0.04	2.361	18.71	7.42	3.457	1.187	0.396
0.05	2.391	18.48	7.33	3.399	1.179	0389

*Table 7. Refractive index, Molar refraction, Molar polarizability, Oxide ion polarizability, Optical basicity and Metallization criterion of silver-doped zinc tellurite glass system.* 



Fig. 12. Plot of direct energy band gap with respect to Ag<sub>2</sub>O content at room temperature.



Fig. 13. Plot of indirect energy band gap with respect to Ag<sub>2</sub>O content at room temperature.

Based on the absorption spectra, the coefficient data are calculated and the graph of Tauc's plot is obtained. The extrapolation of the graphs provides both direct and indirect optical band gap information for the silver-doped glass series. The graphs are displayed in Figure 12 and 13 for silver-doped glass series respectively. The graph of  $(\alpha\hbar w)^2$  against  $\hbar w$  provide the direct optical band gap data while  $(\alpha\hbar w)^{1/2}$  against the  $\hbar w$  provides the indirect optical band gap data. All data are enumerated in Table 6.



Fig. 14. Plot of direct and indirect energy band gap with respect to Ag<sub>2</sub>O content at room temperature.

As mentioned earlier, the analysis of the optical spectra of all the glass samples that fitting the curve in Fig. 12 and 13 provide good fit for n=2 indicating indirect transition and n=1/2 for direct transition. The values of direct optical band gap lie within the range of 3.231 to 3.505 eV while the values of indirect optical band gap are within 3.015 until 3.362 eV. The values for both direct and indirect optical band gaps exhibit trends of decreasing as silver oxide is included in the glass system. The decreasing phenomenon may be associated with the changes in structure that occurred due to the increase of the disorder and as a result, there is more extension of the localized state within the gap [54.55]. This lead to the formation of non-bridging oxygen which is already present due to the role of silver oxide that acts as a glass modifier. This makes the glass structure to become randomized in which the inclusion of silver oxide causes the breaking of regular structure of the tellurite network that will reduce the optical band gap [56]. It is also relevant that the addition of silver oxide in the glass system will cause a conversion of trigonal bipyramid to trigonal pyramid which promotes the presence of more non-bridging oxygen in the glass network. The electrons in bridging oxygen in the glass system are less tightly bound as compared to bridging oxygen. Thus, less energy is required to break down the bond within the glass network yet the optical band gap decreases at the same time [57,58].



Fig. 15. Plot of  $\ln \alpha$  (cm<sup>-1</sup>) with respect to photon energy,  $\hbar w(eV)$ .



Fig. 16. Variation of Urbach energy with respect to Ag<sub>2</sub>O content at room temperature.

Urbach energy ( $\Delta E$ ) is obtained by calculating the reciprocal of the gradient for the graphs in Fig. 15 while Fig. 16 shows the variation of the Urbach energy with respect to silver oxide within the glass system. The data for this parameter of this glass series is recorded in Table 6.

The trend can be viewed as increases as the dopants increase. This can be attributed to the increase in defects within the glass system [59]. Thus, the increase in Urbach energy with the addition of silver oxide leads to the increase in number of defects. The increment in this parameter can also be proven by theory made by Mott and Davis (1979) where the extension of the localized state near mobility edge increases as the amorphous of the structure increases.



Fig. 17. Variation of refractive index with respect to Ag<sub>2</sub>O content at room temperature.

Refractive index is one of the most crucial properties in optical glasses. Hence, numbers of researchers have carried out the investigation to correlate the refractive index and glass composition. Fig. 17 shows the variation of refractive index with a concentration of silver oxide in zinc tellurite glass system. The obtained data of refractive index for the glass series are tabulated in Table 7. It can be seen that the trend of refractive index is increasing with increasing concentration of silver oxide. The refractive index can be considered to be linearly proportional to the density. The earlier information of the density for the glass series reveals that the density is increasing with an increasing number of refractive index [1]. Besides that, the refractive index is also affected by the polarizability of the glass system. High polarity of silver ions which is 2.4 A<sup>3</sup> is larger than the tellurium ions (1.595 A<sup>3</sup>), zinc ions (0.283 A<sup>3</sup>) and erbium oxide (2.253 A<sup>3</sup>) in the tellurite glass network which will break the Te-O-Te (bridging oxygen) and hence increase the non-bridging oxygen in the glass system [61]. The non-bridging oxygen influences the polarizability of the ions which may affect the refractive index. The increasing number of non-bridging oxygen will increase the polarizability and refractive index at the same time [62]. Abdel-baki *et al.*, (2006) had

suggested that the high coordination number of ions may enhance the refractive index of the glass materials [40]. Silver ions have high coordination numbers in which will increase the number of refractive index. The dual nature of silver oxide which acts as a network modifier will occupy the network former position that increases the refractive index. The structural variations that occur in the glass system are can be attributed to the conversion of trigonal bipyramid to trigonal pyramid. This occurence will increase the refractive index which can be associated to a high polarizability phenomenon taking place. This phenomenon will also enhance the number of bridging oxygen to develop.



Fig. 18. Variation of molar refraction with respect to Ag<sub>2</sub>O content at room temperature.

Molar refraction relies on the polarizability of the material. As can be viewed from Table 7, the molar refraction increases with an inclusion of silver oxide in the glass system. The use of molar refraction is pushing through the role of ironic packing in regulating the refractive index. Since the refractive index has same trend towards the molar refraction, it can be concluded that a small molar volume will elucidate large number of refractive index in the glass system that consist of ions in similar polarizability [44].



Fig. 19. Variation of molar polarizability with respect to Ag<sub>2</sub>O content at room temperature.

Fig. 19 shows the effect of increasing the silver oxide concentration on the molar polarizability of the glass system. The evaluated data of molar polarizability in the glass series is listed in Table 7. From Table 7, it can be observed that the values of molar polarizability are found to attain the rising trend as the amount of silver oxide increases. The refractive index has a good correlation with molar polarizability whereby the molar polarizability displays direct relationship with the refractive index. This result can be supported by the recent report which states that the behaviour between refractive index and the polarizability are directly proportional to each other

[64]. Therefore, high polarizability of the glass system promote to the massive value of refractive index. Molar polarizability is affected by a number of non-bridging oxygen in the glass material. The non-bridging oxygen has huge polarizability than bridging oxygen. Therefore, large number of non-bridging oxygen will elevate the number of molar polarizability or contrariwise.



Fig. 20. Variation of oxide polarizability with respect to  $Ag_2O$  content at room temperature.

The evaluated data of oxide ion polarizability of the silver oxide doped with zinc tellurite glass system are tabulated in Table 7. Fig. 20 shows the trend of oxide ion polarizability against the concentration of silver oxide. This figure shows the consistent trend of oxide ion polarizability along with the concentration of dopant. After all, this may be due to the different of structural changes in the glass system as the concentration of silver oxide increases. Not only that, the oxide ion polarizability is also affected by the changes in optical band gap energy. The decreasing of optical band gap energy leads to the increasing value of oxide ion polarizability [65].



Fig. 21. Variation of optical basicity with respect to Ag<sub>2</sub>O content at room temperature.

Theoretically, the optical basicity ( $\Lambda$ ) is a measurement of electron donor power of anions in a medium. The anions in this current research refer to the oxygen ions since all the chemicals used are in oxide forms. Besides that, it is also stated by Elkhoshkhany et al., (2014) that this parameter helps in the estimation of the type of bonding that exists in a glass network. This is due to the fact that the formation of bonds in a system is dependent on the donation or sharing of electrons. Thus, as the electron donor power and  $\Lambda$  increases, this will favour to the formation of ionic bonding and cause the glass to become more basic. The reduction of electron donor power tends to produce covalent bonding and the glass will become more acidic.

The obtained values of optical basicity for the glass series are tabulated in Table 7. The plotted graph of optical basicity versus silver oxide is illustrated in Fig. 21. Table 7 reveals that there is a similar trend of optical basicity along with the oxide ion polarizability. Thus, it can be

concluded that the optical basicity has a direct relationship with the oxide ion polarizability. The optical basicity of the two glass series are in high values as they have higher polarizability. In accordance to previous indication about the trend of  $\Lambda$ , the increase of optical basicity implies that the glass samples become more ionic and the electron donor power of oxygen atoms increase. This can be attributed to the inclination of glass polarizability and the amount of non-bridging oxygen (NBOs) within the glass system. As the NBOs increase, the ionic bonding will be improved in which will cause the glass to become more basic. This reasoning is also affirmed by the formation of TeO<sub>3</sub> structural units together with the increment of non-bridging oxygen at that point. Besides that, Lakshminarayana et al., (2008) had also stated that the increment of NBOs results in the inclination of negative charges around the cation and thus increases the optical basicity of the glasses. Polarizability ( $\alpha$ ) and  $\Lambda$  are related where both of these parameters are directly proportional to one another. This is due to the fact that polarization means the distortion of the electron cloud of an anion towards a cation. Hence, as the electron donor power of oxygen ions or optical basicity increases, the tendency of the polarization process to occur becomes easier. The other factor that could result in the inclination of  $\Lambda$  is the weaker bond strength of the dopants [68].



Fig. 22. Variation of metallization criterion with respect to  $Ag_2O$  content at room temperature.

Metallization criterion (M) parameter provides an overview regarding the electrical conduction ability of a glass sample. Besides that, it can also be related to the optical band gap (Eopt) of the glass sample [69]. In the present study, the data of M for zinc tellurite glass series with the addition of silver oxide are recorded in Table 7 and displayed in Figure 22. It is found that the trend of this parameter is following the trend of Eopt. All the values which are more than 0, indicate the insulating behaviour of the glasses under investigation.

The reduction of metallization criterion indicates that the valence and conduction bands become wider, hence contributing to the decrease of  $E_{opt}$  [66,70]. It is previously mentioned that when M is equivalent to 0, the glass sample is said to achieve the metal state. Therefore, the continuous reduction of M as silver oxide is added into the glass system suggests that the glass sample exhibits lesser insulating behaviour with the incorporation of silver oxides.

### 4. Conclusion

A series of zinc tellurite glass system doped with silver oxide with a chemical formula of  $[[(TeO_2)_{0.7}(ZnO)_{0.3}]_{0.96}$   $(Er_2O_3)_{0.04}]]_{1-x}$   $(Ag_2O)_x$  whereby x = 0.01, 0.02, 0.03, 0.04 and 0.05 were fabricated by applying melt-quenching method. All of the glass samples are amorphous in nature since there is no sharp peak observed from the XRD analysis. FTIR analysis indicates the existence of the TeO<sub>4</sub> units' stretching vibrations in the glass samples. The density of the glass samples rises from the molar fraction 0.01 until 0.05. The increment in density is attributed to the excess of oxygen in the glass systems and also due to the heavier molecular weight of silver oxide as compared to other elements in the glass system. The molar volume of glass samples shows the

opposite behaviour to the density. The pulse echo overlap method at 5 MHz was used to determine the elastic properties of the glass samples by evaluating the longitudinal and shear velocity and the results show a fluctuating trend as expected. Due to the creation of BO that produces strong connectivity and rigidity of the glass, all the values including longitudinal velocity, shear velocity, longitudinal modulus, shear modulus, bulk modulus, Young's modulus, Poisson's ratio, hardness, Debye temperature, acoustic impedance and softening temperature increase with respect to the content of silver oxide. At the same time, when the value of the related moduli decreases, it is because of the conversion of BO to NBO which is considered to distract the glass network and decrease the rigidity as well as the connectivity of the glass a network. Elastic properties may be affected by the structural changes that correspond to the variation of the molar fraction. In this research, the behaviour of elastic properties at different molar fraction is also related to the density and molar volume. For optical analysis, the UV-VIS absorption spectra, band gaps, Urbach energy, refractive index, molar refraction, molar polarizability, oxide ion polarizability, optical basicity and metallization criterion were analysed.

The optical absorption edges do not show any sharp peaks which indicate that the glass samples are amorphous in nature. In addition, the values of direct and indirect energy band gap are noticed to be decreased in trend as the molar fraction of silver oxide increases. This is due to the formation of non-bridging oxygen in the glass network which causes the glass structure to become less order. On the other hand, the value of Urbach energy is viewed to increase as more silver oxide is added into the glass system. This is due to the increasing amount of defects which act as nuclei for crystallization, when the silver oxide content in the glass samples increases. This can be supported by the proportionality of Urbach energy to the number of defects in the glass network. Besides that, the other parameters such as refractive index, molar refraction, molar polarizability, oxide ion polarizability, optical basicity and metallization criterion are observed to increase as the silver oxides are added into the glass system.

This behaviour is due to the increasing formation of NBO atoms which are more polarizable than the BO atoms, the effect of large concentration of heavy metal ions (silver ions) which leads to the formation of dense packing of heavy metal modifiers into the host material and the size of the cation of silver ion which has high polarizability value. A small value of metallization criterion lead to wider conduction and valence bands which will reduce the band gap at the same time. Small amount of metallization criterion lead the glass to become less insulator and more metallic. Therefore, the elastic and optical properties of the fabricated silver-doped zinc tellurite glass system show high performance of the glass system, making it highly attractive for optical application.

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#### References

- [1] R. El-Malawany, Tellurite glasses (2002).
- [2] M. K. Halimah, W. M. Daud, H. A. A. Sidek, A. W., Zaidan, A. S. Zainal, Materials Science-Poland 28(1), 173 (2010).
- [3] Z. A. S., Mahraz, M. R. Sahar, S. K. Ghoshal, Journal of Molecular Structure 1072, 238 (2014).
- [4] S. Peng, L. Wu, B. Wang, F. Yang, Y. Qi, Y. Zhou, Optical Fiber Technology 22, 95 (2015).
- [5] V. P. Tuyen, B. Sengthong, V. X. Quang, P. Van Do, H. Van Tuyen, L. X. Hung, T. Hayakawa, Journal of Luminescence 178, 27 (2016).
- [6] A. H. Battez, R. González, J. L. Viesca, J. E. Fernández, J. D. Fernández, A. Machado, J. Riba, Wear 265(3-4), 422 (2008).
- [7] K. Takahashi, A. Yoshikawa, A. Sandhu, Springer-Verlag Berlin Heidelberg. 239, 1 (2007).

- [8] K. Nomura, H. Ohta, K. Ueda, T. Kamiya, M. Hirano, H. Hosono, Science 300(5623), 1269 (2003).
- [9] A. Bakin, A. El-Shaer, A. C. Mofor, M. Al-Suleiman, E. Schlenker, A. Waag, Physica Status Solidi (c) 4(1), 158 (2007).
- [10] M. F. Faznny, M. K. Halimah, M. N. Azlan, Journal of Optoelectronics and Biomedical Materials 8(2), 49 (2016).
- [11] S. S. Hajer, M. K. Halimah, Z. Azmi, M. N. Azlan, Chalcogenide Letters 11(11), 1 (2014).
- [12] Z. A. S. Mahraz, M. R. Sahar, S. K. Ghoshal, M. R. Dousti, Journal of Luminescence 144, 139 (2013).
- [13] M. K. Halimah, M. N. Azlan, S. Z. Shafinas, Advanced Materials Research 1112, 7 (2015).
- [14] H. A. A. Sidek, S. Rosmawati, Z. A. Talib, M. K. Halimah, W. M. Daud, American Journal of Applied Sciences 6(8), 1489 (2009).
- [15] M. N. Azlan, M. K. Halimah, H. A. A. Sidek, Journal of Luminescence 181, 400 (2017).
- [16] F. Gonella, Ceramics International 41(5), 6693 (2015).
- [17] M. R. Dousti, M. R. Sahar, S. K. Ghoshal, R. J. Amjad, A. R. Samavati, Journal of Molecular Structure 1035, 6 (2013).
- [18] M. Cai, Y. Lu, R. Cao, Y. Tian, S., Xu, J. Zhang, Optical Materials 57, 236 (2016).
- [19] B. Huang, Y. Zhou, F. Yang, L. Wu, Y. Qi, J. Li, Optical Materials 51, 9 (2016).
- [20] A. M. Noorazlan, H. M. Kamari, S. S. Zulkefly, D. W. Mohamad, Journal of Nanomaterials 2013, 168 (2013).
- [21] K. Nanda, N. Berwal, R. S. Kundu, R. Punia, N. Kishore, Journal of Molecular Structure 1088, 147 (2015).
- [22] M. N. Azlan, M. K. Halimah, S. Z. Shafinas, W. M. Daud, Materials Express 5(3), 211 (2015).
- [23] P. G. Pavani, K., Sadhana, V. C. Mouli, Physica B: Condensed Matter 406(6-7), 1242 (2011).
- [24] A. Kaur, A. Khanna, F. González, C. Pesquera, B. Chen, Journal of Non-Crystalline Solids 444, 1 (2016).
- [25] K. Maheshvaran, P. K. Veeran, K. Marimuthu, Solid State Sciences 17, 54 (2013).
- [26] Y. B. Saddeek, E. R. Shaaban, H. M. Moustafa, Physica B: Condensed Matter 403(13-16), 2399 (2008).
- [27] V. V. Gowda, Physica B: Condensed Matter 456, 298 (2015).
- [30] P. G. Pavani, M. Prasad, V. C. Mouli, Journal of Alloys and Compounds 527, 5 (2012).
- [31] P. Pascuta, E. Culea, Materials Letters **62**(25), 4127 (2008).
- [32] A. Noranizaha, K. Azmanb, H. Azhanb, E. S. Nurbaisyatula, A. Mardhiahb, J. Teknologi 69(6), 1 (2014).
- [33] Y. B. Saddeek, Materials Chemistry and Physics 83(2-3), 222 (2004).
- [34] M. A. Sidkey, R. El Mallawany, R. I. Nakhla, A. A. El-Moneim, Journal of Non-Crystalline Solids 215(1), 75 (1997).
- [35] J. Coelho, C. Freire, N. S. Hussain, Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy 86, 392 (2012).
- [36] A. N. Kannappan, S. Thirumaran, R. Palani, ARPN Journal of Engineering and Applied Sciences 4(1), 27 (2009).
- [37] G. Gupta, A. D. Sontakke, P. Karmakar, K. Biswas, S. Balaji, R. Saha, K. Annapurna, Journal of Luminescence 149, 163 (2014).
- [38] M. S. Gaafar, N. A. El-Aal, O. W. Gerges, G. El-Amir, Journal of Alloys and Compounds 475(1-2), 535 (2009).
- [39] H. Afifi, S. Marzouk, Materials Chemistry and Physics 80(2), 517 (2003).
- [40] S. Balaji, K. Biswas, A. D. Sontakke, G. Gupta, D. Ghosh, K. Annapurna, Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy 133, 318 (2014).
- [41] T. Sekiya, N. Mochida, A. Ohtsuka, M. Tonokawa, Journal of Non-Crystalline Solids 144, 128 (1992).
- [42] U. Hoppe, E. Yousef, C. Rüssel, J. Neuefeind, A. C. Hannon, Journal of Physics: Condensed Matter 16(9), 1645 (2004).
- [43] P. Sellappan, V. Jayaram, A. H. Chokshi, C. Divakar, Journal of the American Ceramic Society 90(11), 3638 (2007).

- [44] T. Rouxel, Journal of the American Ceramic Society 90(10), 3019 (2007).
- [45] K. H. Chang, T. H. Lee, L. G. Hwa, Chinese Journal of Physics 41(4), 414 (2003).
- [46] J. E. Shelby, Introduction to Glass Science and Technology 1 (2005).
- [47] Y. B. Saddeek, Glass Physics and Chemistry **38**(4), 373 (2012).
- [48] A. A. El-Moneim, I. M. Youssof, L. A. El-Latif, Acta Materialia 54(14), 3811 (2006).
- [49] V. Rajendran, A. N. Begum, M. A. Azooz, F. H. El Batal, Biomaterials 23(21), 4263 (2002).
- [50] M. M. Elokr, Y. M. AbouDeif, Journal of Molecular Structure 1108, 257 (2016).
- [51] S. Y. Marzouk, Physica B: Condensed Matter **405**(16), 3395 (2010).
- [52] Y. B. Saddeek, Materials Chemistry and Physics 91(1), 146 (2005).
- [53] R. El-Mallawany, Materials Chemistry and Physics 60(2), 103 (1999).
- [54] D. Zhu, C. S. Ray, W. Zhou, D. E. Day, Journal of Non-Crystalline Solids 319(3), 247 (2003).
- [55] H. K. Obayes, H. Wagiran, R. Hussin, M. A. Saeed, Materials & Design, 94, 121 (2016).
- [56] N. F. Mott, E. A. Davis, Clarendon, Oxford, 465 (1979).
- [57] I. Shaltout, Y. I. Tang, R. Braunstein, E. E. Shaisha, Journal of Physics and Chemistry of Solids 57(9), 1223 (1996).
- [58] S. Sindhu, S. Sanghi, A. Agarwal, V. P. Seth, N. Kishore, Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy 64(1), 196 (2006).
- [59] A. I. Sabry, M. M. El-Samanoudy, Journal of Materials Science 30(15), 3930 (1995).
- [60] M. Farouk, A. A. El-Maboud, M. Ibrahim, A. Ratep, I. Kashif, Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy 149, 338 (2015).
- [61] R. S. Chakradhar, G. Sivaramaiah, J. L. Rao, N. O. Gopal, Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy 62(4-5), 761 (2005).
- [62] I. Fanderlik, Glass Science and Technology, 1983.
- [63] D. Linda, J. R. Duclère, T. Hayakawa, M. Dutreilh-Colas, T. Cardinal, A. Mirgorodsky, P. Thomas, Journal of Alloys and Compounds **561**, 151 (2013).
- [64] B. Eraiah, Bulletin of Materials Science 29(4), 375 (2006).
- [65] M. Abdel-Baki, F. El-Diasty, Current Opinion in Solid State and Materials Science **10**(5-6), 217 (2006).
- [66] S. B. Mallur, T. Czarnecki, A. Adhikari, P. K. Babu, Materials Research Bulletin 68, 27 (2015).
- [67] V. Dimitrov, T. Komatsu, J. Univ. Chem. Technol. Metall 45(3), 219 (2010).
- [68] N. Elkhoshkhany, R. Abbas, R. El-Mallawany, Ceramics International 40(8), 11985 (2014).
- [69] G. Lakshminarayana, R. Yang, J. R. Qiu, M. G. Brik, G. A. Kumar, I. V. Kityk, Journal of Physics D: Applied Physics 42(1), 015414 (2008).
- [70] E. Duval, A. Boukenter, T. Achibat, Journal of Physics: Condensed Matter 2(51), 10227 (1990).
- [71] V. Dimitrov, S. Sakka, Journal of Applied Physics 79(3), 1736 (1996).
- [72] R. Jose, T. Suzuki, Y. Ohishi, Journal of Non-Crystalline Solids 352(52-54), 5571 (2006).