Scientific Research Journal,
Vol 20, No 1 (2023) 13-32
https://doi.org/10.24191/srj.v20i1.19091

Optical Properties of Cerium Oxide Doped Zinc Borotellurite Glass

Hasnimulyati Laoding[1*], Halimah Mohamed Kamari[2], Azuraida Amat[3], Azianty Saroni[1]

[1]Faculty of Applied Science, Universiti Teknologi MARA Cawangan Pahang, Kampus Jengka, 26400 Bandar Pusat Jengka, Pahang.
[2]Department of Physics, Faculty of Science, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor.
[3]Department of Physics, Centre for Defence Foundation Studies, Universiti Pertahanan Nasional Malaysia, Kem Sungai Besi, 57000 Kuala Lumpur, Malaysia.

**Corresponding author's E-mail: hasnimulyati@uitm.edu.my*

Received: 13 August 2022
Accepted:12 December 2022
Online First: 17 March 2023

ABSTRACT

The addition of rare earth, zinc, and borate in tellurite glass was found can enhance the glass properties and overcome its existing weaknesses. These include improving the glass forming ability and stability. In this study, cerium-doped zinc borotellurite glasses were produced using conventional melt-quenching technique and the optical properties of the glasses were investigated. UV-VIS spectra show that the absorption edge shifted to a higher wavelength with CeO_2 addition from 0 until 0.05 mol. Consequently, the direct and indirect optical band gap was found to decrease from 3.60 to 2.07 eV and 3.28 to 1.46 eV respectively. On the contrary, the refractive index was found to increase from 2.326 to 3.017. This is attributed to the presence of cerium ions and the formation of non-bridging oxygens. Besides that, the Urbach energy found to be maximum at 0.02 mol CeO_2 and minimum at 0.00 mol CeO_2. This indicates that there are some structural disorders occur in the glass network as CeO_2 introduced into the system. The molar refraction, molar polarizability, oxide ion polarizability and optical basicity exhibited similar trend of data as the CeO_2 content was increased which signifies the relation of these parameters. Meanwhile, the metallization criterion is reported to be decreased, indicating that the glass sample become more metallized with the addition of cerium oxide.

Keywords: Borotellurite glass; Cerium; Optical characterization; Optical band gap

INTRODUCTION

Nowadays, tellurite glass has attracted the attention of many researchers due to its advantages compared to other glass formers such as silicate, phosphate, and germanate. Some of the advantages are low melting temperature, low maximum phonon energy, high host refractive index, good IR transmittance, large rare-earth doped ability, wide light transmission range from ultraviolet to mid-infrared and high dielectric constant [1,2]. However, the difficulty of tellurium dioxide (TeO_2) in forming glass on its own causes researchers to add other chemical oxides into the glass system. Borate (B_2O_3) and zinc oxide (ZnO) were found to be effective in overcoming the problem as these oxides can increase the glass forming ability of tellurite and at the same time produce glass with high stability.

Studies on borotellurite glass have been previously done by Geidam et al., [3], Faznny et al. [4], Maheshvaran et al. [5], and Berwal et al. [6]. It was reported that the glass has a low glass transition temperature, high optical non-linearity, and high chemical durability. Hence, it is suitable to be applied in optical devices. In terms of structural changes, the TeO_3 trigonal pyramids were found to form concurrently with BO_4 tetrahedra in the glass system. These structural units were connected by vertex-sharing and the excess and deficiency of positive charges compensated each other.

Cerium is one of the elements that is included in lanthanide group and is the most active among all of the rare earth elements. In the form of cerium oxide (CeO_2), it can exists in two possible valence states which are Ce^{4+} and Ce^{3+}[7]. When cerium is doped into a glass system, it can be a promising material for photonic applications due to its 5d-4f transitions. According to Singh et al., [8], it is used to make blue luminescent optical systems. Besides that, it is also used as laser active media, radiation protection of fiber optic materials and scintillation materials.

Even though there are several researches that had reported on rare-earth doped ZnO-B_2O_3-TeO_2 glass [4,9–12], however none of the studies

have used cerium oxide as their dopant. Hence, cerium oxide was doped into zinc borotellurite glass in the present work in order to study the effects that CeO_2 has on the optical properties of the glass. Based on the previous researches, it is expected for the optical band gap to decrease while the refractive index and polarizability to increase.

METHODOLOGY

Ce-doped $[(TeO_2)_{0.7}(B_2O_3)_{0.3}]_{0.7}[ZnO]_{0.3}$ glasses were obtained by using the melt-quenching technique where the concentration of CeO_2 was varied from 0 until 0.05 mol. All the chemical oxides used were purchased from Alfa Aesar Company and had a high purity at 99.99 %, 97.5 %, 99.99 % and 99.9 % for TeO_2, $B2O_3$, ZnO and CeO_2 respectively. Firstly, all the chemicals were weighed in order to get the amount needed for the glass samples. Then, the chemicals were mixed and ground for 30 minutes to obtain a homogenous mixture. In this step, the grinding process is very crucial as it can help in reducing the spaces between the particles and thus prevent the formation of bubbles in the glass samples. After that, the mixture was put into an alumina crucible and placed in a furnace to undergo a pre-heating process at 400 °C. This process was done to get rid of any water vapour that might be present in the mixture and trapped during the melting process. After one hour, the crucible was transferred to another furnace for melting process at 900 °C. This process was also done for one hour. Next, the molten was poured into a stainless steel mould that has been pre-heated at 400 °C. After that, the mould was put back into the first furnace for annealing process at 400 °C for two hours. Lastly, the furnace was turned off and the glass sample was left overnight for it to cool down to room temperature. On the next day, the sample was taken out from the furnace and cut to 2 mm thickness using diamond cutter. Both sides of the glass also were polished using sandpaper to obtain clear and parallel surfaces. The optical properties of the samples were then tested using UV-1650PC UV-VIS Spectrophotometer (Shimadzu) with light wavelength in the range of 220-800 nm.

RESULTS AND DISCUSSION

Absorption spectra

The absorption spectra of cerium-doped zinc borotellurite glasses are displayed in Figure 1. In the spectra, it can be observed that there was one peak that appeared at 300 nm as cerium oxide was added. According to Kindrat et al. [13], this peak is attributed to the transition of electron from 2F5/2 to 5d1 energy level. The peak was only appeared after CeO_2 was added due to the fact that rare earth elements possess 4f electrons which are responsible for the unique optical properties of the glasses produced. This is because these electrons are shielded by the outer valence electrons and make them not participate in the bonding between the neighbouring atoms [14]. Furthermore, the existence of the peak is also evidence of the presence of cerium in the glass system. This can be attributed to the unique range of wavelength where the peak occurs for each type of atom.

Besides that, the optical absorption edge of the spectra was found to be not sharply defined. A factor that contributes to the broad absorption edge is the disorder in the glass structure [15] that caused the valence and conduction band to vary in different ways and different degrees. Hence, this can be one of the evidences that proves the amorphous nature of the samples [16]. It also shifted to the longer wavelength with the addition of CeO_2. This might be due to the changes in the oxygen bond strength in the glass system as reported by Maheshvaran et al. [5]. The modification can be related to the formation of non-bridging oxygens (NBOs) which bind electrons less tightly compared to the bridging oxygens (BOs) [17]. Hence, the oxygen bond strength decreased and caused the red shift to occur. Moreover, the shift also happened due to the presence of stable Ce^{3+} or Ce^{4+} oxidation states in the glass system. As a result, the impurity band became more extended into the main band gap [8].

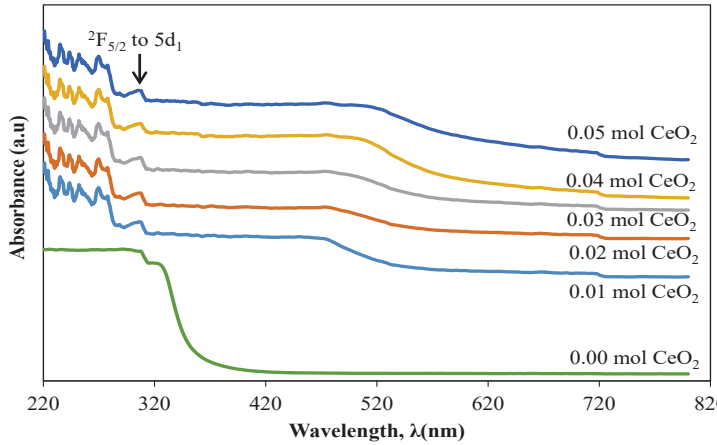

Figure 1: Absorption spectra of cerium-doped zinc borotellurite glass system

Optical band gap and Urbach energy

The optical band gap is an important parameter used to describe solid state luminescent materials. It is calculated based on absorption spectra in order to understand the optically induced transition of a material [18]. It is computed using the Mott and Davis theory [19] as shown in Equation (1);

$$\alpha(\omega) = \frac{B\left(\hbar\omega - E_{opt}\right)^r}{\hbar\omega} \quad (1)$$

where $\alpha(\omega)$ is the absorption coefficient, B is the band tailing parameter, ħω is the incident photon energy, E_{opt} is the optical band gap and r is the index number used to decide the nature of the interband electronic transition causing the absorption which is ½ for direct allowed transition and 2 for indirect allowed transition. Meanwhile, the absorption coefficient can be determined from the absorbance data based on the Equation (2) [20];

$$\alpha(\omega) = 2.303\,\frac{A}{d} \quad (2)$$

where A is the absorbance and d is the sample thickness.

Referring to Equation (1), Tauc's plot can be produced and optical band gap values can be obtained by extrapolating the graph to meet at $(\alpha\hbar\omega)^2=0$ and $(\alpha\hbar\omega)^{½}=0$ for direct and indirect allowed transition respectively. Figure 2 and Figure 3 show the graph's extrapolation for the prepared glasses.

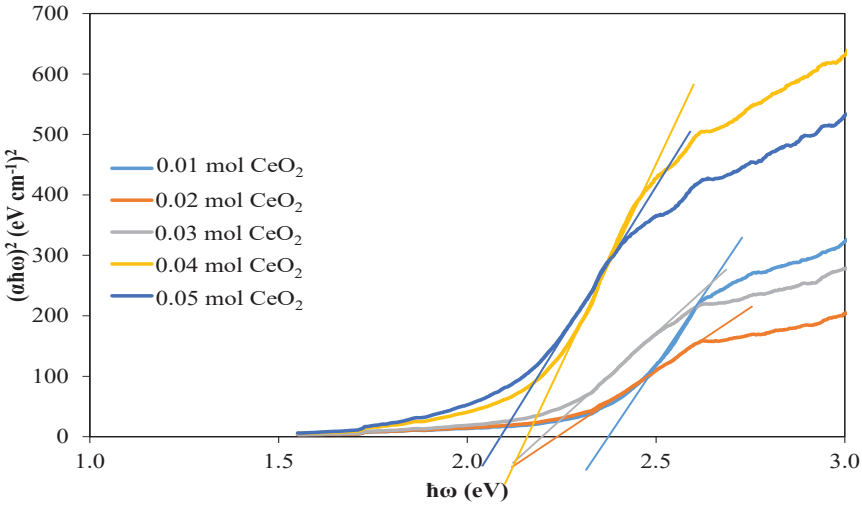

Figure 2: Plot of $(\alpha\hbar\omega)^2$ vs ħω for cerium-doped zinc borotellurite glass system

In terms of Urbach energy (ΔE), this parameter gives information regarding the structural disorder in a glass system. Similar with E_{opt}, it can also be obtained from the optical absorption coefficient data based on the equation [21];

$$\alpha(\omega) = \alpha_0 \, exp\left(\frac{\hbar\omega}{\Delta E}\right) \qquad (3)$$

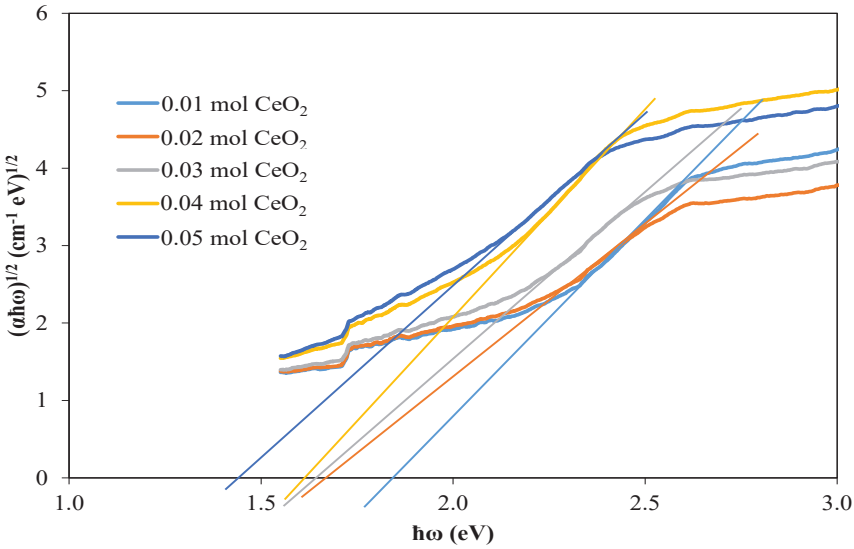

Figure 3: Plot of $(\alpha\hbar\omega)^{1/2}$ vs $\hbar\omega$ for cerium-doped zinc borotellurite glass system

where α_0 is a constant. From this equation, the graph of ln α vs $\hbar\omega$ can be plotted and ΔE is the reciprocal of the graph's slope. All data for the optical band gaps and Urbach energy are recorded in Table 1 and illustrated in Figure 4 and Figure 6.

Table 1: Direct and indirect optical band gap of cerium-doped zinc borotellurite glass system

CeO_2 molar fraction (mol)	Direct E_{opt} (eV)	Indirect E_{opt} (eV)	Urbach energy, ΔE (eV)	**Ref.**
0.00	3.60	3.28	0.1850	**[21]**
0.01	2.39	1.81	0.3789	
0.02	2.25	1.68	0.6249	
0.03	2.21	1.60	0.4609	
0.04	2.16	1.57	0.3922	
0.05	2.07	1.46	0.5339	

The indirect optical band gaps for cerium-doped glasses were found to be ~3.28, 1.81, 1.68, 1.60. 1.57 and 1.46 eV for 0.00, 0.01, 0.02, 0.03, 0.04 and 0.05 mol of CeO_2 respectively. Meanwhile, the direct optical band gaps were found to be in the range of 2.07 until 3.60 eV. As 0.01 mol of CeO_2 was added to the glass system, it shows a drastic decrease in both indirect and direct Eopt and steadily decrease as CeO_2 added up to 0.05 mol. This trend can be related to the weaker bond strength of the additional cerium oxide (Ce-O: 111.45 N/m) compared to the other oxides (Te-O: 219.00 N/m; B-O: 668.42 N/m; Zn-O: 216.37 N/m) that were used in this study. As a result, the average bond strength of the glass system will decrease, thus leading to the reduction of conduction band energy as well. This is due to the fact that as the average bond strength decreases, the valence electron will become loosely bound in the atoms. Hence, a lower amount of energy is required for an electron in the valence band to be excited into the conduction band and cause the optical band gap to become smaller [22].

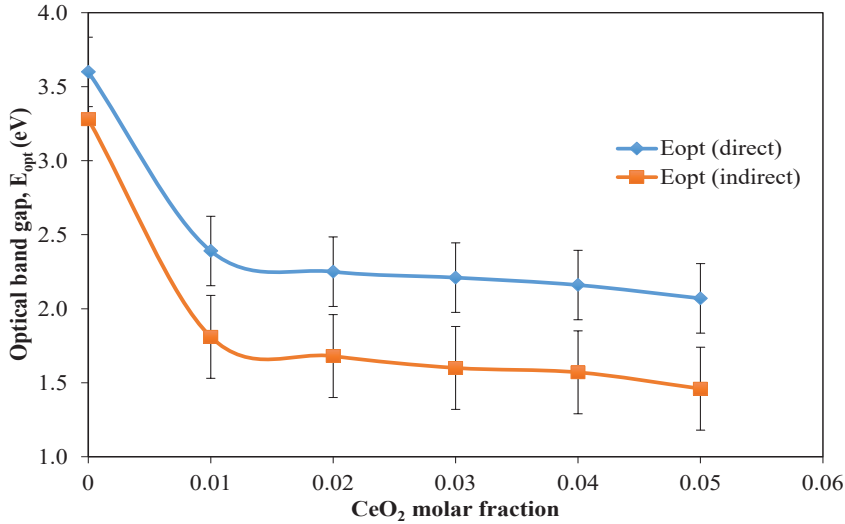

Figure 4: Direct and indirect optical band gap of cerium-doped zinc borotellurite glass system

Another reason that might contribute to the reduction of E_{opt} is the changes in the glasses' colour as cerium was added. As shown in Figure 5, the colour of the glass samples changed from light yellow into brownish-orange with the addition of CeO_2. When the glass becomes darker, it

indicates that the minimum wavelength of light that can be absorbed by the glass is increased [23]. As a result, the optical band gap of the glass samples is decreased.

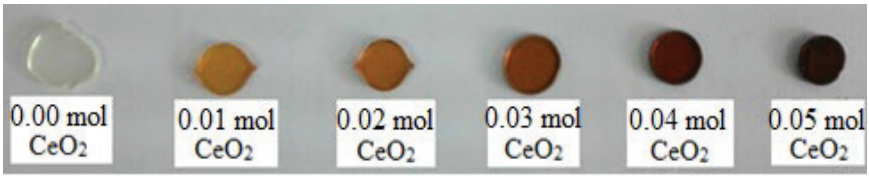

Figure 5: Glass samples with composition of $\{[(TeO_2)_{0.7}(B_2O_3)_{0.3}]_{0.7}[ZnO]_{0.3}\}_{1-x}\{CeO_2\}_x$

Besides that, the creation of non-bridging oxygens (NBOs)[24] can also become a factor to the decline of optical band gap for the prepared glasses. This is because NBOs are not bonded with another atoms. Hence, they hold valence electrons less tightly compared to the bridging oxygens (BOs)[25]. As a result, the valence band will go further away from the nucleus and nearer to the conduction band. This in turn causes the optical band gap to be decreased.

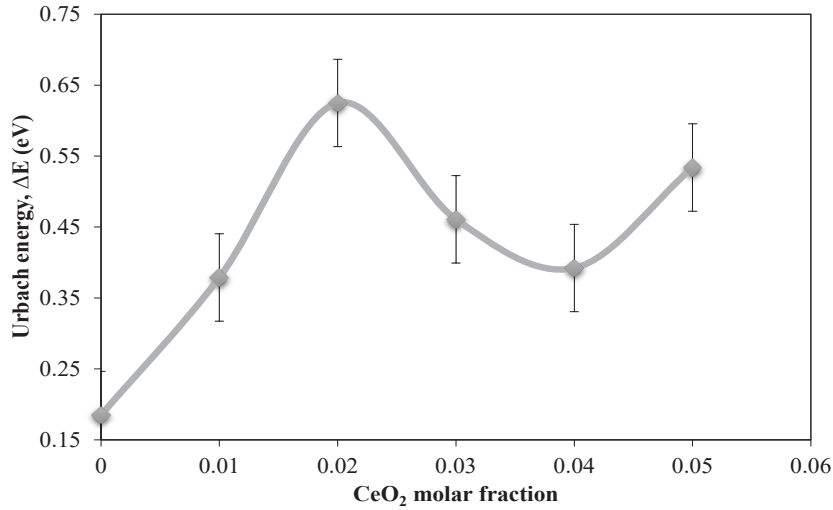

Figure 6: Urbach energy (ΔE) of cerium-doped zinc borotellurite glass system

The Urbach energy (ΔE) of the glass samples were found to vary in the range of 0.1850 to 0.6249 eV (Table 1). Besides the fact that this parameter tells about the structural disorder in a glass network, Azlan et al. [21] reported that it is also related to the width of localized states. As ΔE increases, the amount of structural disorder in a glass system inclines and there is a high probability for the weak bonds to turn into defects. As a result, the glass network become less stable and decreases in its connectivity. On the other hand, the decrease of Urbach energy at 0.03 and 0.04 mol addition of CeO_2 (Figure 6) reflects that the structural disorder is decreasing as the glass becomes more stable and decrease in its fragility nature [26,27]. This might be attributed to the formation of $BTeO_3$ and $BTeO_5$ structural units that contribute to the increment in the connectivity of the glass system and cause the defects to be minimized [5].

According to Silins [28], there are three types of defects that can be present in a glass system which are intrinsic defects, impurity defects and intrinsic impurity defects. Intrinsic defects are defects that arise from the atomic size's local deviation from a short-range order. Impurity defects come from isolated impurity atoms in a glass network. Meanwhile, intrinsic impurity defects are isolated impurity atoms that are attached to the intrinsic defects. It is also stated that the latter type of defect is the characteristic of a doped glass and believed to be the type of most defects in the glasses under study.

Refractive index, molar refraction, molar and oxide ion polarizability

Refractive index (*n*) is another important parameter in determining the optical properties of glasses. It generally measures the ratio of light velocity in the vacuum to that in a medium. As it is known that the velocity of light in a denser medium is slower than in a less dense medium, it can be expected that materials with a higher density possess a higher refractive index compared to materials with low density. In the present study, this parameter was calculated (Equation 4) from the optical band gap data using the relation that proposed by Dimitrov and Komatsu [29];

$$1 - \frac{n^2 - 1}{n^2 + 2} = \left(\frac{E_{opt}}{20}\right)^{\frac{1}{2}} \quad (4)$$

where n is the refractive index while E_{opt} is the optical band gap.

Molar refraction reveals the contribution of ionic packing to the overall refractive index of a glass. Based on previous researchers, it is directly related to the polarizability of constituent ions that exist in a glass sample, along with the refractive index, density and average molecular weight of the glass [30]. Equation (5) was used to calculate molar refraction.

$$R_m = V_m \left(\frac{n^2 - 1}{n^2 + 2}\right) \quad (5)$$

where R_m is the molar refraction, V_m is the molar volume and n is the refractive index.

Polarizability is a measure of the response of electrons to an electromagnetic field, which is the distortion of electron cloud. This parameter can affect the refractive index of a glass in a way that when the electron cloud is distorted, the electrons will lump together and interact with the light that penetrates the glass. As a result, the light velocity is reduced, and the refractive index rises. There are two types of polarizabilities that can be calculated which are molar polarizability (α_m) and oxide ion polarizability ($\alpha_{O^{2-}}$). The Equations (6) and (7) were used to compute the parameters.

$$R_m = \frac{4}{3}\pi N_A \alpha_m \quad (6)$$

$$\alpha_{O^{2-}} = \frac{\left[\left(\frac{R_m}{2.52}\right) - \sum_i x_i p_i \alpha_i\right]}{\sum_i x_i q_i} \quad (7)$$

where N_A is the Avogadro's constant, x is the molar fraction, p is the number of cations, α is the ionic polarizability and q is the number of oxygen atoms.

Data on the refractive index (n) shows an increasing trend as CeO_2 was added into the glass network (Figure 7). Meanwhile, molar refraction (R_m), molar polarizability (α_m) and oxide ion polarizability possessed similar trends which are decreased at 0.01 mol of CeO_2 and increased beyond that (Figure 7 and Figure 8). The variation of n can be related to the increment of density as stated in the earlier expectation. The same rule was found to be applied to the glass under study. When cerium oxide with high molecular weight was incorporated into the glass network, it caused the density of the glass to increase as well as its refractive index as confirmed by the data of density in Table 2.

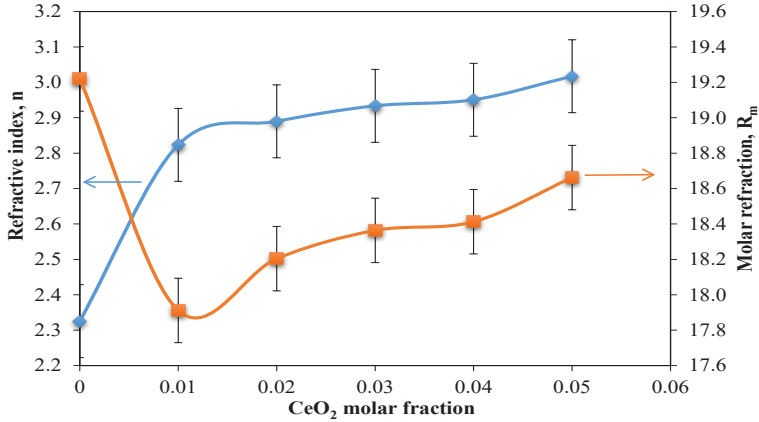

Figure 7: Variation of refractive index and molar refraction of cerium-doped zinc borotellurite glass system

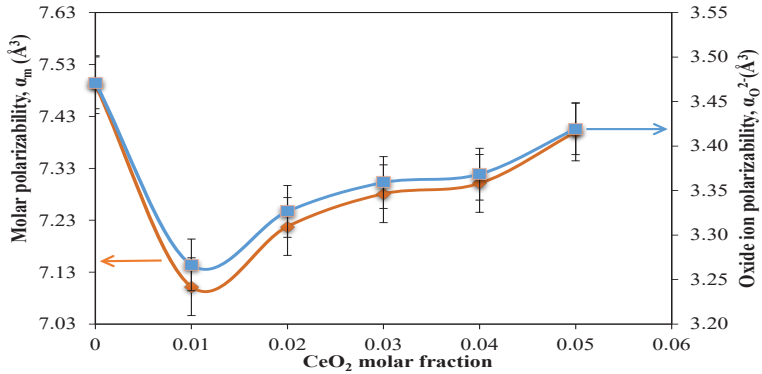

Figure 8: Variation of molar and oxide ion polarizability of cerium-doped zinc borotellurite glass system

Table 2: Density (ρ), molar volume (V_m), refractive index (n), molar refraction (R_m), molar (α_m) and oxide ion polarizability (α_O^{2-}) of cerium-doped zinc borotellurite glass system

CeO_2 molar fraction (mol)	Density, ρ (g/cm³)	Molar volume, V_m (cm³/mol)	n	R_m	α_m (Å³)	α_o^{2-} (Å³)
0.00	3.69	31.75	2.326	18.892	7.491	3.471
0.01	4.60	25.62	2.823	17.911	7.102	3.267
0.02	4.62	25.63	2.890	18.204	7.218	3.327
0.03	4.64	25.61	2.934	18.364	7.281	3.359
0.04	4.67	25.58	2.951	18.413	7.301	3.368
0.05	4.69	25.57	3.017	18.662	7.400	3.419

In addition, the refractive index is also affected by the molar refraction of the glass, polarizability of the cation, coordination number of the ion and oxide ion polarizability of the glasses [31]. This is confirmed by the data displayed in the figures above where n, R_m, α_m and α_O^{2-} have a similar trend from 0.01 until 0.05 mol of cerium oxide. The increment of molar and oxide ion polarizability can be attributed to the presence of highly polarizable ions that are NBOs and Ce. NBO bonds with the characteristics of a greater ionic character and lower bond energies cause them to have a higher polarizability and cation refractions [32]. The lower bond energy and cation refractions can be related in terms of the weak interaction between cation and anion in NBO bonds. Hence, electrons in the cation will become loosely bound and easily polarized. As these electrons interact with incident light, they will reduce the speed of light and caused the cation refractions to become higher. Meanwhile, CeO_2 has a high polarizable cation (0.702 Å^3), low unit field strength and hence, a low influence to its electron cloud charge. As cerium replaced B^{3+} (0.002 Å^3) and Zn (0.29 Å^3) that have lower polarizability, this led to the elevation of molar refraction, molar polarizability as well as the refractive index [33]. Moreover, the growth of cross-links in the glass network also contributes to the rise of n as the addition of cross-linking is directly proportional to the density of the sample.

Optical basicity and metallization criterion

Optical basicity (Λ) represents the average electron donor power of oxygen ions in a medium. Thus, the increment of optical basicity shows that the electrons are easily polarized. Besides that, it also gives an insight about the nature of bonding, either ionic or covalent in a glass system. This parameter can be calculated using the data of oxide ion polarizability as shown in Equation (8).

$$\Lambda = 1.67\left(1 - \frac{1}{\alpha_{O^{2-}}}\right) \tag{8}$$

Another parameter that can be investigated is the metallization criterion (*M*). This parameter shows either our glass tend to have a metallic or non-metallic nature according to the theory on metallization as proposed by Herzfeld [34]. M is computed using the data on molar refraction and also molar volume as shown in Equation (9).

$$M = 1 - \frac{R_m}{V_m} \tag{9}$$

In this study, the variation of optical basicity followed the trend of oxide ion polarizability where it was found to decrease at 0.01 mol of CeO_2 and increase beyond that. This shows that these two factors also contributed to the variation of α_m and R_m as well as the increment of n. Furthermore, the reduction of Λ indicated that the electron donor power of the oxygen ions was decreasing. Hence, covalent bonds tend to produce in the glass network. This is due to the fact that bonding formation is caused by the sharing and donation of electrons. As the ability of oxygen ions to donate electron is decreasing, it will tend to share its electron which causes covalent bonds to form. Meanwhile, the increment of optical basicity from 0.01 until 0.05 mol of cerium oxide signified that the electron donating ability of the oxygen ions had also inclined and lead to the formation of ionic bonding. This increment is also believed to be one of the factors that contribute to the reduction of E_{opt} as presented earlier.

In terms of metallization criterion, it was found that the value of *M* had reduced from 0.415 to 0.270 as Ce was varied. This indicated that the addition of cerium oxide had caused the width of valence and conduction

band to become large and resulted in the decrement of E_{opt} as depicted in Figure 4. According to Dimitrov and Komatsu [35], the M value which is lower than 1 implies that the glass samples possessed a non-metallic nature. However, as the nonmetal-metal transition occurs when $1\text{-}R_m/V_m = 0$, the decrement of M means that the prepared glasses became more metallized with the addition of CeO_2 [33].

Table 3: Optical basicity and metallization criterion of cerium-doped zinc borotellurite glass system.

CeO_2 molar fraction (mol)	Optical basicity, Λ	Metallization criterion, *M*
0.00	1.189	0.4151
0.01	1.159	0.3008
0.02	1.168	0.2898
0.03	1.173	0.2828
0.04	1.174	0.2802
0.05	1.182	0.2702

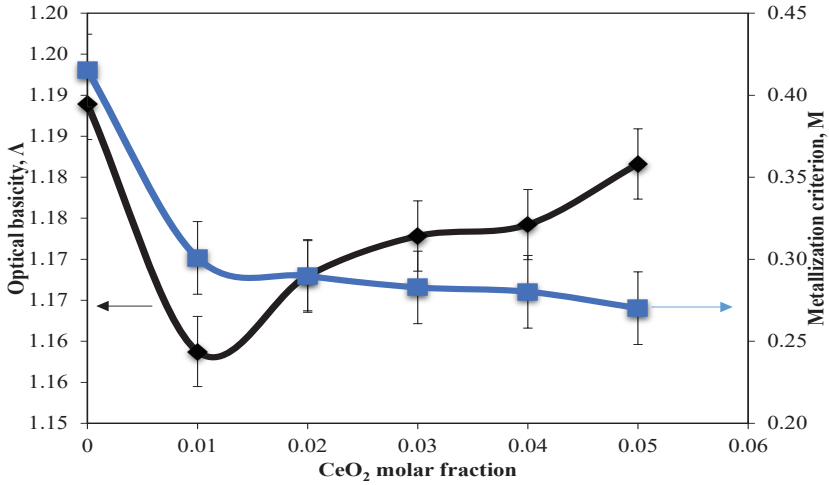

Figure 9: Variation of optical basicity and metallization criterion of cerium-doped zinc borotellurite glass system

CONCLUSION

In conclusion, the glass forming ability of tellurite glass greatly improved in this study where clear and bubbleless cerium-doped zinc borotellurite glass have been successfully produced. The optical properties of the glass samples have been studied in detail. UV-VIS spectra of the glasses show that the absorption edge shifted towards a higher wavelength with the inclusion of CeO_2. This is attributed to the formation of non-bridging oxygens in the glass system and also the presence of Ce^{3+} and Ce^{4+} oxidation state. The direct and indirect optical band gap of the glasses exhibited a decreasing trend with the addition of CeO_2. This was expected as the absorption edge was shifted and the glasses' colour also changed. The Urbach energy, molar refraction and polarizability showed some variation with CeO_2 content. The increment of refractive index indicated that this parameter is related to density, molar refraction and also polarizability. Optical basicity varies in a similar way to oxide ion polarizability. Meanwhile, the reduction of metallization criterion signified that the glass samples became more metallized with the increments of cerium oxide.

DECLARATION OF COMPETING INTEREST

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

ACKNOWLEDGEMENTS

The authors appreciate the financial support from Ministry of Higher Education and Universiti Pertahanan Nasional Malaysia with Short Term Grant SF0123-UPNM/2022/SF/SG/1 and UPNM/2022/GPJP/SG/3, Universiti Putra Malaysia for providing the authors with the usage of laboratory facilities and Universiti Teknologi MARA for providing e-Article Review Tracking System (eRTS) to review this manuscript.

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