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Polarizability of erbium-doped zinc-tellurite glasses coated with graphene oxide for telecommunication devices

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Graphene-based nanomaterials offer superior properties especially in optical performance. The existence of oxygen functionalized group in graphene active layers is beneficial to enhance the optical properties of current tellurite glass. In this work, two series of glasses denoted as ZBTEr (uncoated) and ZBTEr-GO (coated) were successfully prepared using melt-quenching and low-cost spray coating techniques. X-ray diffraction analysis was done on both glass series and confirmed the amorphous structural arrangement in the glass system. Morphological study was investigated by using Scanning Electron Microscopy (SEM) and revealed the distribution of graphene layers on the glass surface. The UV-Vis spectroscopy was carried out and several absorption bands were determined for both series of glasses. The computation of oxide ion polarizability and optical basicity were investigated based on Lorentz-Lorentz equation. The oxide ion polarizability and optical basicity for ZBTEr-GO (coated) glasses were found higher than ZBTEr (uncoated) glasses due to the existence of sp^2 graphitic attached on the oxygenic functional groups in GO layers. The metallization criterion values for ZBTEr-GO (coated) glasses were found decreased and tend to be more metallic. Hence, the deposition of graphene layers on the tellurite glasses offer a new strategy to improve the current telecommunication devices.

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1. Introduction

Graphene is a two-dimensional of single-atom-thick sheet with sp^2 bonded carbon atoms in a hexagonal structure. The use of graphene in broad optical applications display their remarkable usefulness in the field of optics, photonics, and optoelectronics devices for instance,

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modulators, filters, and solar cells [1, 2] . A single layer of graphene consists of semi metal of zero-band gap which expand the ability of any materials to achieve a high optical performance [3, 4]. On top of that, the existence of oxygen functionalize groups in graphene layers may increase the absorption properties of some materials.

The graphene oxide can be synthesized from pure graphite by using chemical approach and produce a multiple graphene oxide (GO) layers [5]. In comparison to graphene, the existence of high quantity of oxygen functionalized groups in graphene oxide possess similar hexagonal carbon structure. However, graphene oxide consists of some chemical structure such as hydroxyl, epoxy, and carboxyl that covalently bonded on the basal plane and edges, which also associated with hydrophilic property that has strongly attributed to the wide-bandgap in GO structure [6, 7]. Additionally, the wide bandgap energy takes place during oxidation process where the optical absorption of electrons in sp^2 carbon configurations oxidized uniformly and convert to sp^3 hybrid carbon [8]. Therefore, it might be implied that coating of tellurite glass with GO layers may significantly prove a successful approach to enhance the optical properties in glass materials.

Up to now, the tellurite-based glasses have been frequently reported as the best host glass for fiber lasers, waveguide, and amplifiers [9] due to its unique physical and optical properties, such as low melting point, high refractive index, low cut-off phonon energy, wide spectral transparency, and excellent infrared trasmission [10–13]. These features can be maximized through GO coating as it can be applied for future fiber optics. The inclusion of erbium oxide, Er_2O_3 in tellurite-based glasses have been investigated extensively to improve the current erbium doped fiber amplifiers (EDFA) [14–16]. Furthermore, the recent study shows a remarkable emission of Er^{3+} ions at around 2.7 µm which is highly beneficial for the laser glass [17].

The new strategy to improve the current tellurite glass through graphene oxide coating have not yet been performed so far. Therefore, we introduce a novel tellurite glass with graphene oxide coating assisted with the unique optical properties. Hence in this present work, it is envisaged that the coated-tellurite glass will offer wide-ranging applications mostly providing the major improvement in fiber optics communications network. This work covers the XRD analysis, morphological study, UV-Vis analysis, oxide ion polarizability, optical basicity and metallization criterion. The outcomes of this research may provide a new strategy to improve the current tellurite glass.

2. Experimental methods

2.1. Preparation of Er^{3+}-doped zinc-tellurite glasses

Erbium ions doped zinc-tellurite glasses were prepared from high purity raw materials consist of TeO_2, B_2O_3, ZnO and Er_2O_3 powders via conventional melt-quenching method. The present glass series are composed of $(0.47(1-y))TeO_2 + (0.2(1-y))B_2O_3 + (0.29(1-y))ZnO + (y)Er_2O_3$ with the different molar concentration of Er^{3+} where y = 0.005, 0.01, 0.02, 0.03, 0.04, and 0.05 mol %, respectively. The appropriate quantities of chemical powders were precisely weighed, well mixed and transferred into a platinum crucible to undergo the stirring process about 30 minutes. The mixture was then placed to an electric furnace and being pre-heated at 400 °C in a period of 1 hour to eliminate any water content that might existing in the mixture.

Next, the sample was followed with the melting process at 900 °C for 2 hours. The cylindrical stainless-steel mould was pre-heated in the electric furnace at 400 °C. The molten was then poured into a stainless-steel mould and instantly annealed at 400 °C for 1 hour. The furnace was then switched off and the glass samples were slowly cooled down to room temperature. Eventually, the prepared glass samples were cut and polished based on the required thickness to obtain the highly transparent and flat surfaces for further optical and structural measurements.

2.2. Fabrication and deposition of GO onto glass surface via spray coating technique

Graphene oxide (GO) was synthesized from the “top-down” approach using a simpler and cheaper electrochemical exfoliation method. Briefly, two graphite rods were dipped into the dissolved electrolyte solution assisted with the commercial anionic sodium dodecyl sulphate (SDS) as a surfactant mixed in de-ionized H_2O for the dispersion process. The graphite rods were then connected to anode and cathode electrodes by applying 7.0 V direct current (DC) voltage supply and immediately allowed for exfoliation of bulk graphite transformed into GO flakes in a period of time about 24 hours.

The deposition of GO on the zinc-tellurite glass surface was applied by using spray coating technique as can be seen in Fig. 1. Afterwards, the GO-coated tellurite glass samples were then proceeded to annealing process at 400 °C for 1 hour under Ar ambient.

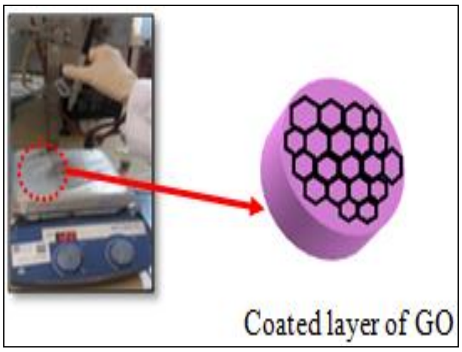

Fig. 1. Schematic diagram of transferring GO on tellurite-based glass surface.

2.3. Characterization techniques

The morphological study of GO-coated on the zinc-tellurite glass surface was investigated by using scanning electron microscopy (SEM, Hitachi SU8020). Approximately 2 mm thick glass sample was performed for the microscopic observation. The phase structure of the GO-coated zinc-tellurite glasses was characterized by X-ray diffraction (XRD) analysis on a Bruker, D2 Phaser instrument equipped with Cu-Kα emission (λ = 0.154 nm). The measurements were conducted in the 2θ range of 20-90° for identification the structural arrangement in glass materials. Ultraviolet-Visible (UV-Vis) spectroscopy was carried out using the Agilent Cary 60 Spectrophotometer with specific wavelength of 200-800 nm.

3. Results and discussion

3.1. Microstructural characterization

Fig. 2 shows the morphologies of the present GO-deposited zinc-tellurite glasses as obtained from SEM images. In general, the GO dispersion covers almost the entire part of deposited glass surface which strongly confirms the presence of GO layers through considerable non-uniform GO thickness distributions. The deposited GO layers are seemed to be transparent, folding thin layers and produced the overlapped flakes especially on the GO edges or basal plain, as indicated in Fig. 2 (a). During the oxidation process from graphite to GO, several defects occurred between the graphite layers and also interrupted the π-bond systems of graphite due to the introduction of abundant oxygen-containing functional groups of GO, thus this functional group form a larger agglomerated structures on the deposited glass surface [18, 19]. Additionally, a formation of microcrystals-ball-like in GO active layers on the glass surface is found as shown in Fig. 2 (b). The incorporation of graphene-based materials impressively affected the surface morphology which attributes to the large aggregated structures with the various oxidation degrees generated via an oxidation process. Apparently, the tendency of aggregation formed on the glass surface may be due to the strong van der walls attraction between graphene sheets thus, it leads to better exfoliation state of GO [20].

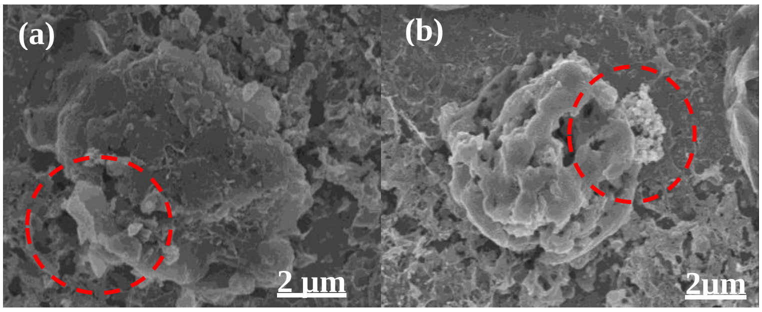

Fig. 2. SEM images of GO deposited on prepared tellurite glass surface: (a) fold-up of GO thin layers (b) microcrystals-ball-like crumpled GO.

3.2. X-ray diffraction (XRD) analysis

Fig. 3 presents the XRD pattern of GO-deposited on zinc-tellurite glass. A broad diffuse or hump pattern was observed within 20°- 40° in the 2θ scale without the existence of any sharp discrete diffraction peaks which indicates the presence of long-range structural disorder characteristics or lack of atomic orderliness in the glass network [21, 22].

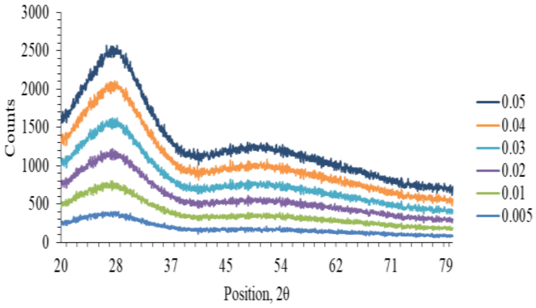

Fig. 3. XRD pattern of ZBTEr-GO (coated) glass.

3.3. Optical absorption and optical coefficient

Fig. 4 displays the optical absorption coefficient spectra of ZBTEr-GO (coated) in the range of 200-800 nm at room temperature. The absorption bands are primarily due to the electronic transitions of erbium ions originating from the ground state level of $^4I_{15/2}$ transition to the different bands of excited states level, $^4G_{11/2} + {}^2H_{9/2} + {}^4F_{5/2} + {}^4F_{7/2} + {}^2H_{11/2} + {}^4S_{3/2} + {}^4F_{9/2} + {}^4I_{9/2} + {}^4I_{11/2}$. It can be seen from the figure that the absorption coefficient increases with an increase of Er^{3+} concentration. The peak positions in the absorption bands indicate that the erbium ions are homogeneously distributed in the zinc-tellurite glass matrix [23]. The combination of sp^2 and sp^3 hybridization present in GO which bonded with carbon atoms may break the symmetry of graphene and shifts the optical absorption [24]. On the other hand, the oxygen functional groups may tunes the optical properties of ZBTEr-GO (coated), particularly in the optical absorption [25].

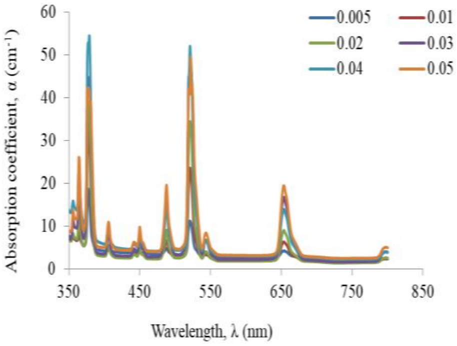

Fig. 4. Optical absorption coefficient of ZBTEr-GO (coated) glasses.

3.4. Oxide ion polarizability

The oxide ions can be classified as the charge carriers in conducting materials and possess highly polarizable ions which often has been ignored in materials. The negative charge of glass materials depends on the structural arrangement and the nature of constituent ions. The number of electrons in oxides are greater than cation charges. The relationship between the $\sqrt{\mathrm{E_g}}$ and $1-\frac{\mathrm{R_m}}{\mathrm{V_m}}$ for a simple oxides based on the following equation [26].

$$\mathrm{E_g} = 20\left(1-\frac{\mathrm{R_m}}{\mathrm{V_m}}\right)^2 \tag{1}$$

where $\mathrm{E_g}$ denotes as energy gap, $\mathrm{R_m}$ presents the molar reftaction, and $\mathrm{V_m}$ is the molar volume of the glasses. The oxide ion polarizability can be determined by substitute the equation (1) into equation (2) as indicated in the following relation:

$$\alpha_{\mathrm{O_2^-}}(\mathrm{E_g}) = \left[\frac{\mathrm{V_m}}{2.52}\left(1-\sqrt{\frac{\mathrm{E_g}}{20}}\right)-\sum\alpha_i\right]\left(\mathrm{N}_{\mathrm{O_2^-}}\right)^{-1} \tag{2}$$

where $\alpha_{\mathrm{O_2^-}}(\mathrm{E_g})$ is the oxide ion polarizability, $\sum\alpha_i$ refer as the molar fraction polarizability, and $\mathrm{N}_{\mathrm{O_2^-}}$ is the number of oxide ions. Fig. 5 presents the variation of oxide ion polarizability for all synthesized ZBTEr and ZBTEr-GO (coated) glasses along with the different concentration of $\mathrm{Er^{3+}}$ ions, meanwhile, Table 1 summarizes the calculated energy gap, $\mathrm{E_g}$ and oxide ion polarizability, $\alpha_{\mathrm{O_2^-}}$ values for ZBTEr and ZBTEr-GO (coated) glasses. The oxide ion polarizability values show an increasing trend in the range 1.903 to 3.495 $\mathrm{Å^3}$ for ZBTEr-GO (coated) glasses. Meanwhile, the uncoated glass falls in the range of 3.306 to 3.472 $\mathrm{Å^3}$ [27]. However, it is observed from the plotted graph that the oxide ion polarizability values for ZBTEr-GO (coated) are decreased gradually at 0.005 molar fraction. This trend is resulted from the lack of homogeneity in GO distribution on certain area of glass surface. Moreover, it is clearly observed that the ZBTEr-GO (coated) becomes less polarized which may be correlated to the role of oxygenated functional groups existing in GO layers. In the meantime, the high degree content of oxygenated functional groups in GO active layers enhanced the energy bandgap and increase the optical absorption of ZBTEr-GO (coated). Therefore, the GO layers may shift the oxide ion polarizability.

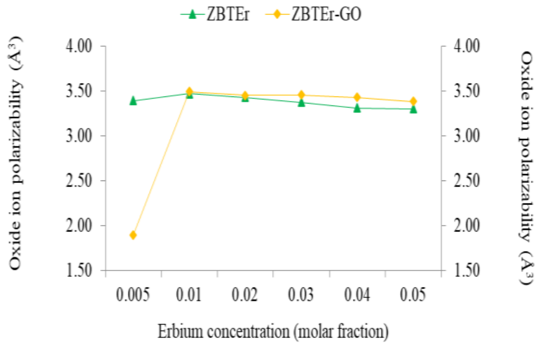

Fig. 5. Variation of energy gap-based oxide ion polarizability values for ZBTEr and ZBTEr-GO (coated) glasses.

Table 1. Comparison of energy gap-based oxide ion polarizability values between ZBTEr and ZBTEr-GO (coated) glasses.

Erbium concentration	Energy gap (E_g) (ZBTEr)	Energy gap (E_g) (ZBTEr-GO)	Oxide ion polarizability, $\alpha_{O_2^-}$ (Å^3) (ZBTEr)	Oxide ion polarizability, $\alpha_{O_2^-}$ (Å^3) (ZBTEr-GO)
0.005	3.348 [27]	3.477 [28]	3.396 [27]	1.903
0.01	3.025 [27]	3.444 [28]	3.472 [27]	3.495
0.02	3.302 [27]	3.495 [28]	3.430 [27]	3.453
0.03	3.262 [27]	3.450 [28]	3.376 [27]	3.461
0.04	3.440 [27]	3.392 [28]	3.316 [27]	3.433
0.05	3.380 [27]	3.454 [28]	3.306 [27]	3.385

3.5. Optical basicity

Optical basicity is defined as an electron donating ability of a material. The glass materials containing oxide ions with high electron density is regarded to be in high basicity [29]. The theoretical calculation of optical basicity, Λ_{th} for different type of component glasses can be identified based on the following expression:

$$\Lambda = X_1\Lambda_1 + X_2\Lambda_2 + \ldots\ldots + X_n\Lambda_n \tag{3}$$

where X_1, X_2, ……., X_n represent the equivalent fractions of each oxides which attribute to the overall material stoichiometry and Λ_1, Λ_2, ……, Λ_n denote as the optical basicity of each individual oxides in the glass system [30]. The values of optical basicity for single element in the glass system are: Λ (Er_2O_3) = 0.929, Λ (TeO_2) = 0.93, Λ (ZnO) = 0.82, and Λ (B_2O_3) = 0.425. Borate oxide is considered to have less basicity than the other oxides. Moreover, the optical basicity of tellurite oxide is higher than erbium oxide which is due to high value of ionic radius in tellurite ions. The optical basicity of the glass system can be obtained as follows [31]:

$$\Lambda = 1.67\left(1 - \frac{1}{\alpha_{O_2^-}}\right) \tag{4}$$

Based on Fig. 6 and Table 2, the optical basicity values for ZBTEr-GO (coated) glasses are found to increased from 0.793 to 1.192. Meanwhile, the optical basicity of ZBTEr [27] falls in range of 1.165 to 1.189. The increasing pattern of optical basicity for ZBTEr-GO (coated) is affected by the high number of oxygenated functionalized groups in GO active layers which contributes to the less acidity. The presence of sp^2 graphitic due to the attached oxygen functional epoxy and hydroxyl groups through interlayer of hydrogen bond located on the basal plane [32] causes the higher covalent bond which expected to give the increment in optical basicity values. However, the less value of optical basicity at 0.005 molar fraction for ZBTEr-GO (coated) may be related to the inhomogeneity of GO layers on the glass surface. Furthermore, the less number of negative charge in the oxygen atoms may leads to the lower covalency for cation-oxygen linking in glass network system [33].

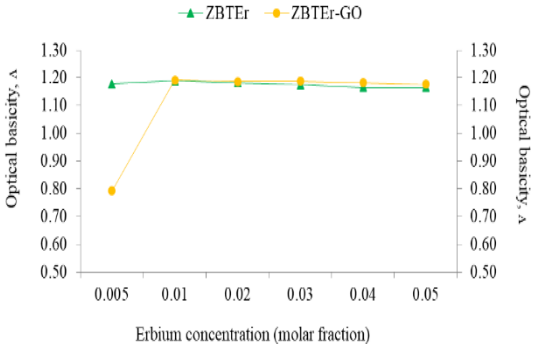

Fig. 6. Variation of optical basicity values for ZBTEr and ZBTEr-GO (coated) glasses.

Table 2. Comparison of optical basicity values between ZBTEr and ZBTEr-GO (coated) glasses.

Erbium concentration	Optical basicity, Λ (ZBTEr)	Optical basicity, Λ (ZBTEr-GO)
0.005	1.178 [27]	0.793
0.01	1.189 [27]	1.192
0.02	1.183 [27]	1.186
0.03	1.175 [27]	1.188
0.04	1.166 [27]	1.183
0.05	1.165 [27]	1.177

3.6. Metallization criterion

Metallization criterion is used to determine the tendency of glass to be more metallic or insulating nature. According to metallization theory proposed by Herzfeld [34], if the glass materials have ratio of $\frac{R_m}{V_m} < 1$, then the materials have high tendency to be more non-metallic characteristics, meanwhile, if the glass materials falls in ratio of $\frac{R_m}{V_m} = 1$ or $\frac{R_m}{V_m} > 1$, then the glass materials have high tendency to be more metallic. Hence, the following relation is applied to calculate the metallization criterion value as shown:

$$M = 1- \frac{R_m}{V_m} \tag{5}$$

where R_m known as molar refraction and V_m is the molar volume of prepared glasses. The metallization criterion can be identified as proposed by Dimitrov and Sakka [35] by using the following expression:

$$M\,(n_0) = 1- \left[\frac{n_0^2-1}{n_0^2+2}\right] \tag{6}$$

Fig. 7 and Table 3 present the metallization criterion-based refractive index values for ZBTEr and ZBTEr-GO (coated) glasses along with Er^{3+} ions content. The decreasing trend in metallization criterion is found in ZBTEr-GO (coated) and falls in the range of 0.318 - 0.324. Meanwhile, the metallization criterion for ZBTEr glasses falls in the range of 0.597 - 0.607 [27]. The lower value of metallization criterion for ZBTEr-GO (coated) can be due to the wide band gap in GO layers and higher number of oxygens. The high density of oxygens in GO layers with strong covalent bond has a significant impact on the metallizing of tellurite glass. It can be justified that the GO layers may act as an insulating material. Hence, high number of oxygen functionalized groups in GO layers [36] may potentially tunes the optical bandgap and shifts the metallization criterion for tellurite glass.

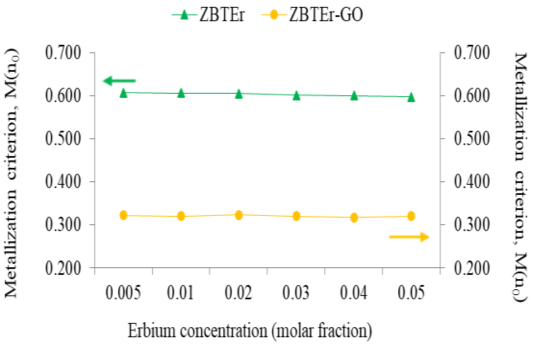

Fig. 7. Variation of refractive index-based metallization criterion values for ZBTEr and ZBTEr-GO (coated) glasses.

Table 3. Comparison of refractive index-based metallization criterion values between ZBTEr and ZBTEr-GO (coated) glasses.

Erbium concentration	**Refractive index, *n* (ZBTEr)**	**Refractive index, *n* (ZBTEr-GO)**	**Metallization criterion, M(n_0) (ZBTEr)**	**Metallization criterion, M(n_0) (ZBTEr-GO)**
0.005	1.716 [27]	2.279 [28]	0.607 [27]	0.323
0.01	1.718 [27]	2.287 [28]	0.606 [27]	0.321
0.02	1.721 [27]	2.275 [28]	0.605 [27]	0.324
0.03	1.730 [27]	2.285 [28]	0.601 [27]	0.321
0.04	1.732 [27]	2.299 [28]	0.600 [27]	0.318
0.05	1.740 [27]	2.285 [28]	0.597 [27]	0.321

3. Conclusion

In summary, the surface morphology, structural and optical properties of ZBTEr-GO (coating) glasses had been studied and compared with ZBTEr glasses. XRD analysis proved the absence of sharp peaks and showed that the glass samples are amorphous in nature. The morphology study revealed the existence and distribution of GO layers on glass surface. The oxide ion polarizability and optical basicity values were found higher in ZBTEr-GO (coating) due to the presence of high number of oxygens which also increase the energy bandgap in glass materials. The metallization criterion values of ZBTEr-GO (coating) glasses were found to decrease which may be correlated to the existing sp^2 graphitic attached on the oxygen functional epoxy and hydroxyl groups via oxidation. Based on these results, GO active layers on the glass surface may be used to tune the optical properties in tellurite glass system and beneficial to improve the current fiber optics.

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