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Incorporation of neodymium, holmium, erbium, and samarium (oxides) in zinc-borotellurite glass: Physical and optical comparative analysis

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ABSTRACT

Investigating the effect of different types of rare-earth oxides on zinc borotellurite glass is important to determine the potential application in optical devices. The addition of rare-earth oxides in zinc borotellurite glass is well-known to enhance the optical properties due to the effects of 4f-4f transitions. In this work, we aim to compare the effect of different rare-earth oxides on zinc borotellurite glass denoted as ZBTNd, ZBTHo, ZBTEr and ZBTSm. The glass samples were successfully fabricated via the melt-quenched technique. The physical investigation of the glasses has been done by measuring the density and molar volume. It was found that ZBTNd glass has the lowest density than the other glasses due to the small atomic radius in neodymium oxide. High-density value for ZBTHo glass shows potential to be used as radiation shielding properties. The high value of molar volume for ZBTNd glass is advantageous for fiber optics as ZBTNd glass has good performance in elasticity. It was found that ZBTEr has a lower refractive index than the other glasses due to low dispersion characteristics. However, ZBTEr glass has good performance to be used in optical communication applications. It was found that the optical absorption shifts to a longer wavelength beginning from ZBTEr > ZBTHo > ZBTNd > ZBTSm. The optical band gap energy for ZBTEr glass is higher than the other glasses due to the Coulomb repulsion energy for erbium which is greater than neodymium and samarium and slightly higher than holmium. The pattern of electronic polarizability for all glasses was found as follows ZBTSm>ZBTNd>ZBTEr>ZBTHo. The optical basicity for ZBTEr was found highest which indicates a higher acidity, meanwhile, the ZBTNd glass has the lowest value which corresponds to a higher basicity.

Keywords: tellurite glass, rare-earth oxides, optical properties

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Introduction

There is no denying the extensive ongoing investigations on glass science and technology, which discuss discoveries in photonics and optical applications [1]. Rapid development and innovation in telecommunications enhance the production of new materials for optical fiber and laser [2]. A wide range of glass materials has often been produced to manufacture optical devices. Silicate-based glass is widely utilized as the primary core of the optical fiber. However, silicate-based glass has more than 1500 ^{0}C melting point, moderate absorbency and high signal loss [3]. The high quality of optical glass is essential for the advancement of current optoelectronic devices. Whilst tellurite-based glass is the best choice for high-quality glass materials [4].

Tellurite oxide is comprised of double triangular bipyramids which are surrounded by four oxygen atoms in the tellurite glass network. Four oxygen atoms are located at a distance of 1.95 Å from the tellurite ion [5]. Hence, the tellurite ion is located in an intermediate state between four oxygen atoms. Pure TeO_2 glass is not stable and exists in a crystal state with four coordination numbers as Te^{4+}. The addition of modifier ion in tellurite oxide, TeO_2 may lead to the formation of the glassy state and hence vitrified TeO_2. The tellurite ion in the glass network is more stable in four coordination numbers than in six coordination numbers. This trend is due to the shrinking distance between tellurite and oxide atoms, Te–O as the valence electrons increase in the glass network. Besides that, if the modifier ions have the same coordination number and size as TeO_6, the six-atom coordination state of Te^{4+} might be stable [6].

Erbium oxide is a well-known material to be used in fiber amplifiers such as erbium-doped fiber amplifier (EDFA) devices [7]. In previous research, holmium oxide has been used extensively in telecommunication and solid-state lasers [8]. Moreover, samarium oxide is one of the best lanthanide compounds to be utilized as an optical amplifier and fiber [9]. Meanwhile, neodymium oxide has the greatest interest in the area of optical materials due to its near-infrared lasing properties at around 1.06 µm [10]. Hence, the inclusion of these rare-earth oxides is beneficial to improve the optical properties of the current zinc borotellurite glass. Extensive studies have been done to incorporate these rare-earth oxides in tellurite glass. However, the detailed comparative analysis between these rare-earth oxides is rarely documented.

This study aims to compare the role of holmium, erbium, neodymium and samarium oxides on the physical and optical performance of zinc-borotellurite glass. The objectives of this study are to determine the physical properties of the glasses such as density and molar volume. The optical analysis such as optical absorption, optical band gap energy, Urbach energy, electronic polarizability, optical basicity and metallization criterion is investigated. The outcomes of this study will provide a detailed comparative analysis between rare-earth oxide inclusions in zinc borotellurite glass. These reports will be useful to manufacture novel materials to be used in optoelectronic applications.

Methodology

A conventional melt-quenching method was used in the chemical formula of $\{\{[(TeO_2)_{0.70}(B_2O_3)_{0.30}]_{0.7}(ZnO)_{0.3}\}_y(RE_2O_3)_{1-y}$, RE= Ho_2O_3, Er_2O_3, Nd_2O_3 and= 0.005, 0,01, 0,02, 0.03, 0.04, 0.05). High-purity chemical powder (99 per cent purity grade) of erbium oxide, Er_2O_3 (Reacton, Alfa Aesar), holmium oxide, Ho_2O_3 (Reacton, Alfa Aesar), neodymium oxide, Nd_2O_3 (Reacton, Alfa Aesar), samarium oxide, Sm_2O_3 (Reacton, Alfa Aesar), tellurite oxide, TeO_2 (Reacton, Alfa Aesar), boron oxide, B_2O_3 (Reacton, Alfa Aesar) and zinc oxide, ZnO (Reacton, Alfa Aesar) were collected and used. The raw materials were measured with a ±0,0001 g accuracy and carefully mixed to produce 13 g of mixed powder by using an electrical balance.

The mixture was placed in the alumina crucible and preheated by an electric furnace at 400 0C for 1 hour. The aim of the preheating process is that the excess hydrogen molecules are removed from the mixture. The mixture in the alumina crucible was then melted in the second electric furnace at a temperature of 900 0C for 2 hours. During this process, the molten mixture was formed. The molten was then transferred to the cylindrical stainless-steel mold which was preheated for 1 hour at 400 0C. During the quenching process, the glass sample was formed. For the annealing process, the glass sample in a cylindrical stainless-steel mold was heated for 1 hour at 400 0C. The aim is to increase mechanical strength and remove stress during the cooling process. The glass sample was then cooled down at around 5 hours at room temperature. By using Isomet Buehler high precision low-speed saw machine, the obtained glass sample was cut to a thickness of about 2 mm. The sample was polished to get a smooth surface on either side of the sample with a various sandpaper grade (1000 grid, 1.500

grid and 2.000 grid). Shimadzu-1650PCUV-Vis spectrophotometer was used to analyze the absorption band of the glass sample.

Results and discussions

Physical comparative analysis

The density of the glass system has a significant effect on the elastic properties, the refractive index and the mechanical strength [11]. High high-density glass matrix is known to be beneficial for increasing the refractive index. In contrast, a low dense glass matrix has an excellent contribution to the elastic and mechanical properties of the glass matrix.

The density calculation can be made using the following formula:

$$\rho = \rho_t \frac{W_a}{W_a - W_t} \quad (1)$$

where ρ_t is the density of water W_a and W_t are the weight of the sample in the air and water respectively.

The change in density may be due to several factors, such as the softening or compactness of the structure, the type of doping, the coordination number and the atomic radii of the components [12]. The results of density for different types of rare-earth doping are shown in Figure 1. Figure 1 shows that neodymium oxide doping has the lowest density among doping, while holmium oxide has the highest value. Several factors may explain the large differences in density between ZBTNd and ZBTHo. The first factor is the atomic radius of the dopant, which is higher in holmium oxide than in neodymium oxide.

The high degree of atomic radius may significantly contribute to an increase over free space in the glass matrix [13]. As a result, the compactness of the glass matrix will be reduced due to an increase in free space. In addition, the bond length between the rare earth atom and oxygen has a significant effect on the oxygen packing density in the glass matrix [14]. Table 1 shows that neodymium oxide has a higher bond length than holmium oxide, which reduces the oxygen packing density. The density of ZBTEr and ZBTSm glasses shows small differences in number compared to ZBTNd glasses. However, erbium oxide has a similar atomic radius with holmium oxide and small differences in bond length. The small degree of density of erbium oxide compared to holmium oxide can be explained by the change in the number of polyhedral coordination after the formation of the glass system [15]. Holmium oxide has a higher number of polyhedral coordination than erbium oxide, which in turn increases the density [16]. In addition, the properties of radiation shielding are highly dependent on the density value. High glass density contributes to excellent shielding properties. ZBTHo glasses therefore have a high potential to be used as shielding properties due to their high-density value.

The investigation of the molar volume of the glass system is important for the analysis of the spatial distribution of oxygen in the glass matrix [17]. The molar volume is directly affected by the density of the glass system through the compaction and expansion of the glass structure [18]. In addition, the result of molar volume is more significant in the analysis of structural changes in the glass network, except mass (m) from density, and the inclusion of equal particles for comparison between samples. The molar volume can be calculated by the following formula:

$$V_m = \sum_i \frac{x_i M_i}{\rho} = \frac{M}{\rho} \quad (2)$$

where x_i and M_i denote the molar fraction and molecular weight of the respective component and M is the total molecular weight of the composition. The obtained values for molar volume are listed in Table 1 and plotted in Figure 2. It is noted from Figure 2 that the molar volume is higher in ZBTNd than in the glasses. The interatomic spacing between the atoms may influence the molar volume in the glass network [19]. The high bond length in neodymium oxide may lead to the increase in interatomic spacing which in turn increase the molar volume. Moreover, the high number in ionic radius may increase the tendency of free expansion in the glass matrix [20]. The low number of molar volume values is obtained in ZBTHo glasses which represent the reciprocal value of density. Bulk modulus has a high dependency on molar volume which determines the elasticity of optical fiber. Based on the obtained molar volume data, ZBTHo glasses have a lower performance in elasticity than ZBTNd glasses. Hence, ZBTNd glasses is more preferable to be used for optical fiber than ZBTHo glasses.

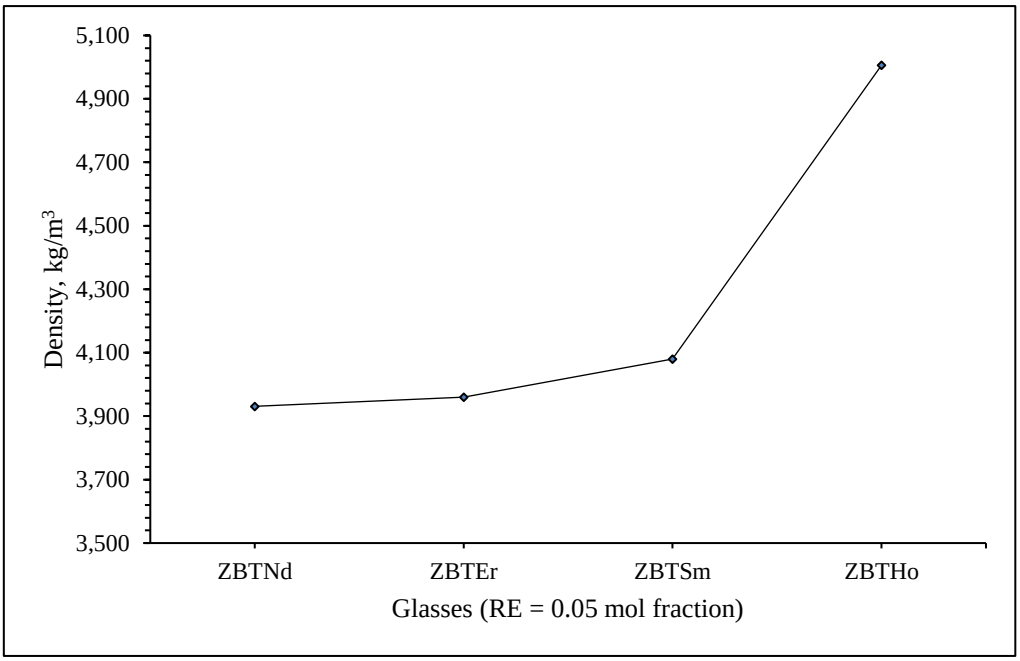

Figure 1 - Density of ZBTNd, ZBTEr, ZBTSm and ZBTHo glasses

Table 1 - Density of ZBTNd, ZBTEr, ZBTSm and ZBTHo doped tellurite glass

Glasses	Density (kg/m^3)	Molar Volume (m^3/mol)	Atomic radius (pm)	Bond length	Ionic radius
ZBTNd	3.931	128.200	-	-	-
ZBTHo	5.006	26.043	-	-	-
ZBTEr	3.960	32.955	-	-	-
ZBTSm	4.080	31.570	-	-	-
Single constituents					
TeO_2	-	-	140	1.974	2.210
B_2O_3	-	-	85	1.236	0.230
ZnO	-	-	135	1.975	0.740
Nd_2O_3	-	-	185	2.559	0.983
Ho_2O_3	-	-	175	2.385	0.901
Er_2O_3	-	-	175	2.310	0.890
Sm_2O_3	-	-	185	2.509	0.958

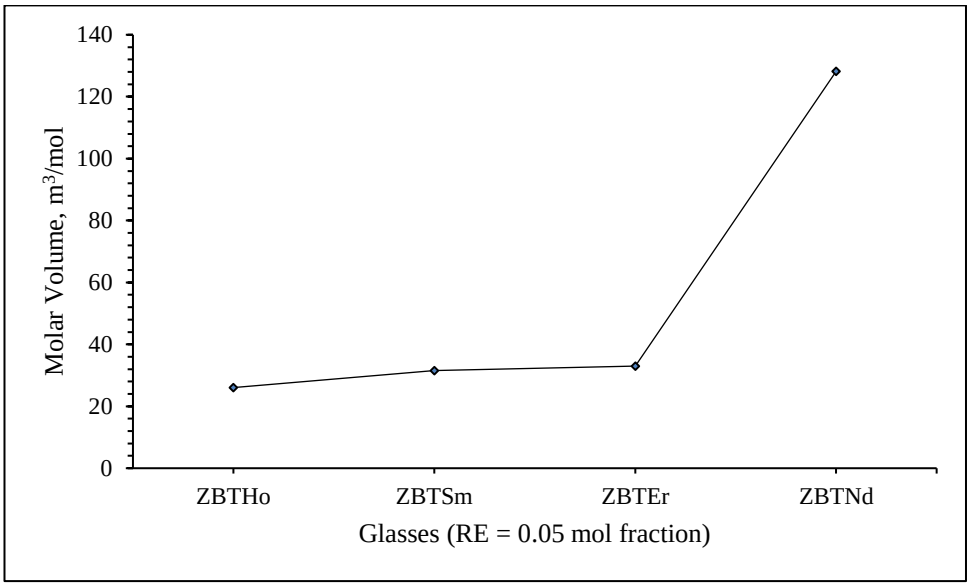

Figure 2 - Molar volume of ZBTHo, ZBTSm, ZBTEr and ZBTNd glasses

Refractive index

The refractive index is an important parameter to be used in a variety of applications such as smart glass, lenses, optical fiber and electronic displays [21]. The structure of an amorphous glass network is closely associated with the amount of non-bridging oxygen in the glass network. The existence of non-bridging oxygen will contribute to improving the polarization of materials which, in turn, may impact the value of refractive indexes. There are two effects of polarization on the transmission of light in a medium;

1. Some light transmission may be absorbed,
2. The delay in the velocity of light waves passes through the medium.

The large polarizability of the glass system minimizes the velocity of light propagation in a medium which, in turn, generates a high refractive index. The results of the refractive index are shown in Table 2 and shown in Figure 3. Figure 3 shows that the highest refractive index is the ZBTSm glasses.

Factors that affect the value of the refractive index are the characteristics of dispersion, doping coordination number, non-bridging oxygen, electronic cloud density, polarization and density [22]. Based on previous data, the density of the ZBTHo glasses is higher than that of the ZBTSm glasses. It is therefore presumed that the refractive index of the ZBTHo glasses must be higher than the rest of the series of glasses. However, the density of the glass system is not the only factor affecting the value of the refractive index.

Cation polarization values for single samarium oxide and holmium oxide are 1.16 Å and 0.91 Å respectively**.** The high number of cation polarizability can therefore contribute to the increase in the refractive index value. The high value of the refractive index is beneficial to produce fiber optics as it widens the angle of reflection by increasing the critical angle. The lowest refractive index value is found in the ZBTEr glasses due to the low cation polarization (0.89 Å) compared to the other dopants. Another possibility is that ZBTEr glasses may have low dispersion characteristics compared to other dopants as one of the important parameters for optical communication is the dispersion characteristic.

High-dispersion characteristics may reduce the performance of optical fiber by increasing the optical pulse and limiting the information-carrying capacity of the fiber. It can therefore be justified that ZBTEr glasses are also a good choice to produce fiber optics, particularly as a core material. In addition, the amount of non-bridging oxygen may contribute to the variation of the refractive index as the existence of lone-pair electrons is high in non–bridging oxygen. Based on the trend of the refractive index, the ZBTSm glasses produce high levels of non-bridging oxygen compared to ZBTHo, ZBTNd and ZBTEr glasses.

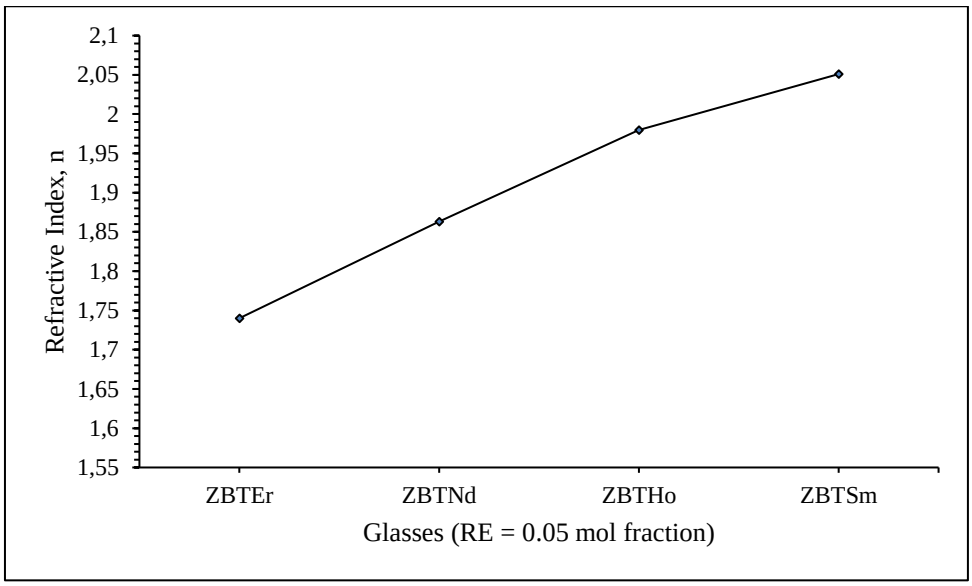

Figure 3 - Refractive index of ZBTEr, ZBTNd, ZBTHo and ZBTSm glasses

Table 2 - Refractive index of ZBTEr, ZBTNd, ZBTHo and ZBTSm glasses

Glasses	Refractive index
ZBTEr	1.740
ZBTNd	1.863
ZBTHo	1.980
ZBTSm	2.051

Optical absorption and band gap energy

Optical absorption spectra have a significant role in determining the properties of the electronic structure of non-crystalline solids. The optical absorption in glass materials can occur by two mechanisms which are electronic polarization and the excitation of an electron from the valence band to the conduction band [23]. The occurrence of optical absorption by electronic polarization only depends on the light frequency in the constituent atoms. Meanwhile, the process of optical absorption during the transition of an electron from valence to conduction band depends on the electronic band structure of semiconducting materials.

The promotion of an electron from the valence band to the conduction band is due to the absorption of photon energy from the electron. Figure 4 shows the optical absorption spectra for ZBTSm, ZBTNd, ZBTHo and ZBTEr glasses. It can be seen from the figure that the optical absorption shifts to a longer wavelength beginning from ZBTEr > ZBTHo > ZBTNd > ZBTSm. In comparison, the absorbance is higher at the lower wavelength and lower at the higher wavelength for all glasses indicating an increase in the absorption factor.

Sharp peaks are observed in the absorption spectra of all dopants leading to the excitation of electrons from the ground to the excitation state. This trend can be explained by the type of orbital in rare–earth oxide. It is well established that rare earth oxides have 4f orbital, which are localized due to their strong intra-atomic Coulomb association [24]. The 4f-4f orbital in rare earth materials leads to a strong magnetic dipole and an electric dipole force which, in turn, produces sharp peaks in the absorption spectra [25].

The investigation of the optical band gap is important, as the tendency of glass materials to be more applicable in semiconductor devices is expected. Optical absorption in glass materials may arise through two different mechanisms, e.g. electronic polarization and electron excitation from the valence band to the conduction band. The occurrence of optical absorption by electronic polarization relies only on the frequency of light in the constituent atoms. Meanwhile, the process of optical absorption during the transition from valence to conduction band depends on the electronic band structure of semiconductor materials. The promotion of the electron from the

valence band to the conductive band is due to the absorption of photon energy from the electron.

There are two types of optical band gaps that work in both crystalline and amorphous structures that are direct and indirect optical band gaps. In the case of glass materials, the indirect optical band gap is more accurate in describing the behavior of electronic transitions due to the long-range structural disorder in the glass matrix.

The absorption coefficients near the absorption edge of the glass samples are calculated by:

$$\alpha(\lambda)=2.303\frac{A}{d} \quad (3)$$

Where A applies to absorption and d refers to the thickness of the glass samples. The absorbance of the glass materials often impacts the absorption coefficient with a directly proportional behavior to the absorption coefficient value. More evidence of the electronic states of the glass system is given by the higher energy components of the spectral region corresponding with the electronic transition. Electrons are excited through photon absorption from a filled band to an empty band. The following formula can be used to measure photon energy

$$\hbar\omega=\frac{\hbar}{2\pi}(2\pi f) = hf = \frac{\hbar c}{\lambda} \quad (4)$$

where c=2.9979 x 108 m/s and ħ = 4.14 x 10-15 eVs. As a result of the electron transitions, the absorption coefficient α(to) increases significantly. The relationship between α (a), the photon energy of the incident radiation, and the absorption coefficient, can be written as follows:

$$\alpha(\omega)=\frac{B(\hbar\omega-E_{0pt})^n}{\hbar\omega} \quad (5)$$

Where the tailing parameter is denoted as B, Eopt is the optical bandgap energy, n is the type of transition where n=2 in the indirect transition, n=1/2 in the forbidden indirect transition, n=1/3 in the prohibited direct transition, and n=1/3 in the forbidden direct transition. Electromagnetic waves interact with electrons in the valence band, which are raised across a fundamental gap in the conductive band in direct and indirect transition cases [25].

The absorption coefficient is calculated near the absorption edge of all the different glass samples. Amorphous materials are well known to fit equation (3) where n = 2 which is an indirect transition. The equation (3.8) is therefore converted to the following equation:

$$((\alpha\hbar\omega)^2=B(\hbar\omega-E_{opt}) \quad (6)$$

The optical band gap can be used to provide insight into the state of solid-state materials. The Urbach energy (ΔE) be computed by the plot of the logarithm of the absorption coefficient (a(v)) as a function of the photon energy of the amorphous material as given below:

$$\alpha(\upsilon)=ßexp(\frac{\hbar\upsilon}{\Delta E}) \quad (7)$$

Where ß is a constant, h is the plank constant, v is the frequency of the photon, and ΔE is the Urbach energy (Maheshvaran et al., 2013).

Figure 5 shows the optical band gap pattern for ZBTSm, ZBTNd, ZBTHo and ZBTEr glasses and the data are listed in Table 3. It can be seen from Figure 5 that the ZBTEr glass has the highest number of optical band gaps than the other glasses. The variations in optical band gap can be explained by the Coulomb repulsion energy between the glasses. The Coulomb repulsion energy for rare-earth is shown in Figure 6. It is noted that the Coulomb repulsion energy for erbium is greater than neodymium and samarium and slightly higher than holmium.

Coulomb repulsion energy confines the electron in orbital which affects the mobility of electrons to be excited from the ground state to the excited state [26]. Hence, it can be justified that the higher number in the optical band gap may be due to the Coulomb repulsion energy which is greater in erbium. ZBTSm glass has the lowest number of optical band gaps which reflects the lower number in Coloumb repulsion energy. Besides that, the 4f states for rare-earth may affect the excitation of electrons to the conduction band. The unoccupied 4f orbitals may experience the energetic up-shift to the conduction band which improves the optical band gap for ZBTSm [27]. The highest optical band gap value for ZBTEr reflects the lower number of unoccupied 4f orbitals. However, all glasses fall in the range of semiconductor energy gap which is compatible to be used in semiconductor applications.

Figure 7 and Table 3 depicted and listed the values of Urbach energy for the glasses. The exponential area in the absorption coefficient and near to the optical band edge is called as Urbach tail.

The appearance of the Urbach tail is due to the localized states in the amorphous structure which is extended in the band gap. The localized states are associated with the disorders of structure which gives the information of defects in the glass system. The high number of Urbach energy indicates the higher tendency of the weak bond in the glass structure to be converted to defects. It can be seen from Figure 7 that the ZBTSm glass has the highest number of Urbach energy which reflects the fragility of the glass structure. The lower value of Urbach energy in ZBTEr glass shows that the glass structure is less likely to convert the weak bond into defects. Hence, ZBTEr is more stable than the other glasses.

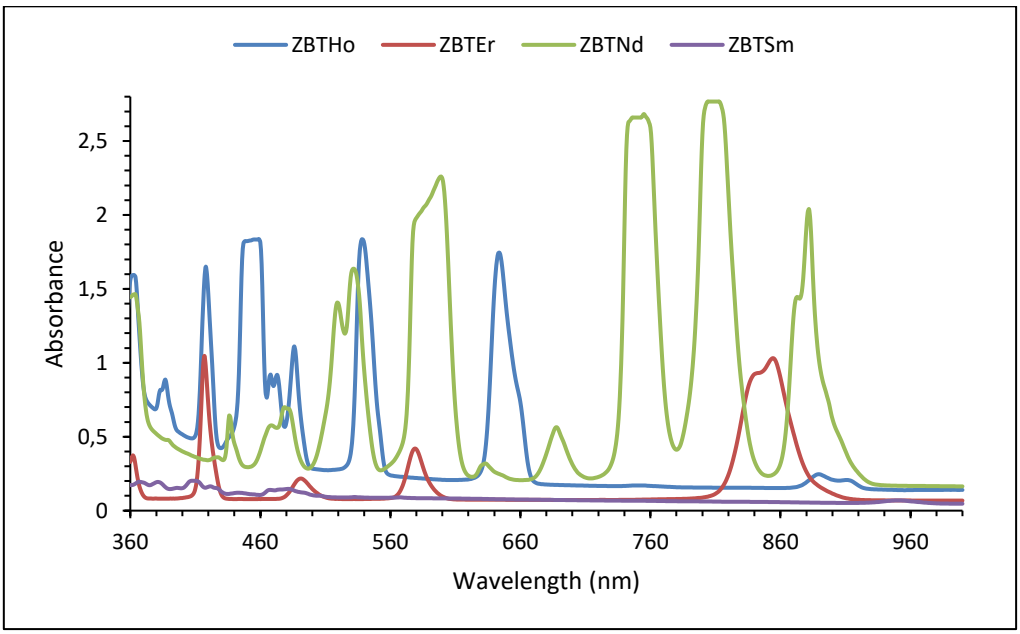

Figure 4 - Optical absorption of ZBTHo, ZBTEr, ZBTNd and ZBTSm glasses

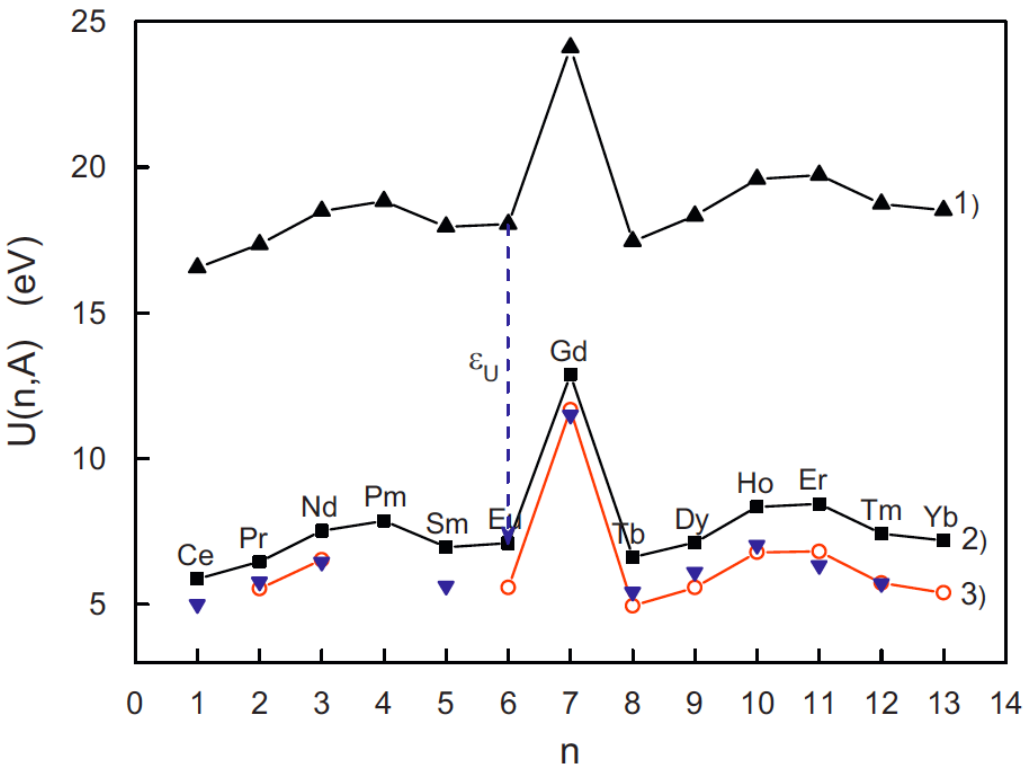

Figure 6 - Coulomb repulsion energy for rare-earth. Reproduced figure from **[27]**

Figure 5 - Optical band gap energy of ZBTSm, ZBTNd, ZBTHo and ZBTEr glasses

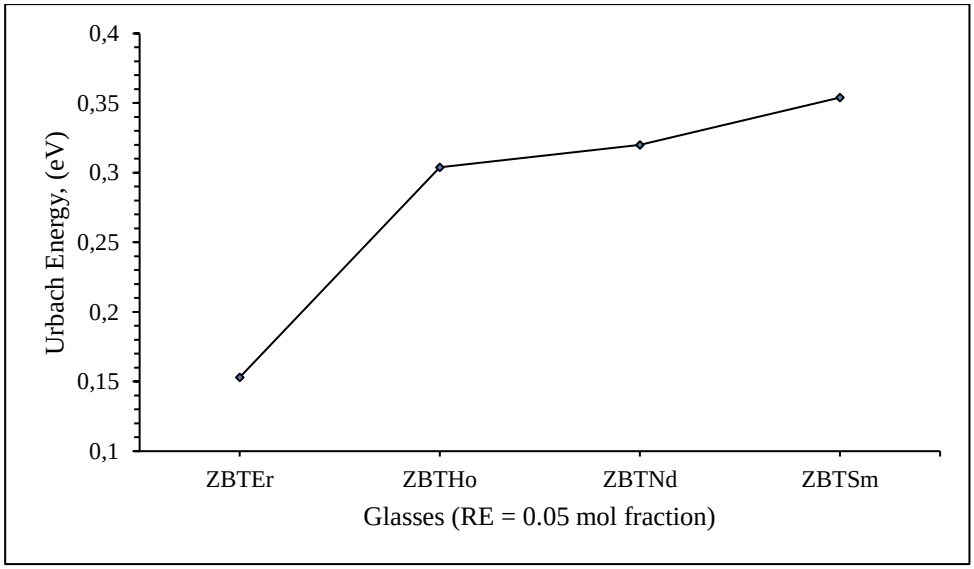

Figure 7 - Urbach energy of ZBTEr, ZBTHo, ZBTNd and ZBTSm glasses

Table 3 - Optical band gap and Urbach energy of ZBTEr, ZBTHo, ZBTNd and ZBTSm glasses

Sample (0.05)	Optical band gap	Urbach Energy
Samarium	2.980	0.660
Neodymium	3.151	0.320
Holmium	3.268	0.432
Erbium	3.380	0.153

Electronic polarizability and oxide ion polarizability

Electronic polarization was used to describe the deformation of electron clouds by applying an electromagnetic field. Electronic polarization affects inter-ionic interaction, refractive index, conductivity, optical basicity and non–linear optical properties. The early study of polarization in the field of glass materials was carried out by Fajans and Kriedl in 1948 [28]. Polarizability values have been

determined for three types of oxide glasses which are; 1. Acid oxides, 2. Ionic oxide, 3. High ion oxides.

The Lorentz-Lorenz equation explains the relationship between the refractive index and the molar volume as shown below:

$$R_m = \frac{(n_0^2 - 1)}{(n_0^2 + 2)} V_m \quad (8)$$

When Rm represents molar refraction, n0 signifies linear refractive index, and Vm implies molar volume. The Lorentz-Lorenz equation represents the average molar refraction for isotropic substances that are liquids, glasses and cubic crystals. The average electronic polarizability can be computed from the Lorentz-Lorenz equation by applying the number of the Avogadro to the given equation:

$$\alpha_m = \frac{3}{4\pi N_A} R_m \quad (9)$$

Where NA defines the Avogadro's number corresponding to the number of polarizable ions per mole. The value 4π/3 is known as a constant in the Lorentz function. Electronic polarization provides the magnitude of the electron response by incorporating the electromagnetic field into the electron clouds. With the α_m in (Å3), equation (3) can be altered into the given formulas:

$$\alpha_m = \frac{R_m}{2.52} \quad (10)$$

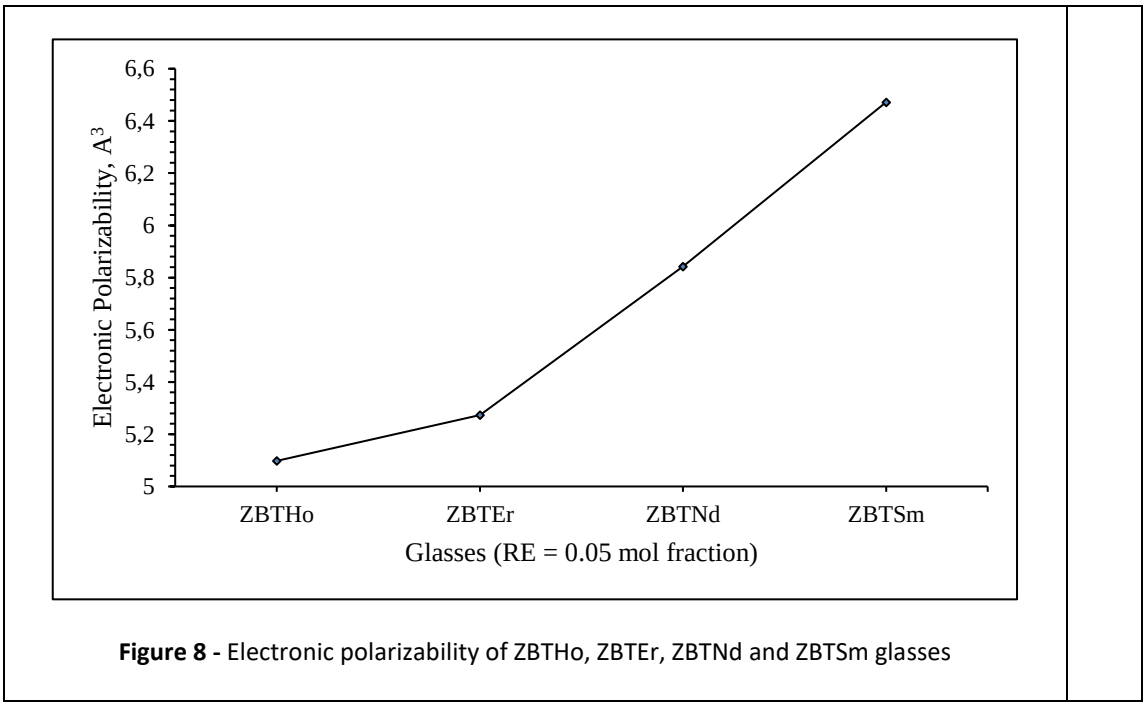

Figure 8 - Electronic polarizability of ZBTHo, ZBTEr, ZBTNd and ZBTSm glasses

Table 4 - Electronic polarizability, oxide ion polarizability, optical basicity and metallization criterion of ZBTHo, ZBTEr, ZBTNd and ZBTSm glasses

Glasses (0.05)	Electronic Polarizability			Metallization Criterion
		Oxide ion polarizability	Optical Basicity	
ZBTHo	5.098	2.361	1.151	0.404
ZBTEr	5.274	3.216	1.165	0.411
ZBTNd	5.843	3.306	1.173	0.483
ZBTSm	6.472	3.516	1.195	0.549

The obtained values of electronic polarizability for all glasses are depicted in Figure 8 and listed in Table 4. The pattern of electronic polarizability for all glasses is as follows ZBTSm>ZBTNd>ZBTEr>ZBTHo. This pattern shows that the ZBTSm glass has a higher tendency to be more polarized than the other glasses. The variations of electronic polarizability can be explained by the attraction between electrons and the nucleus and Hooke's law of potential energy. Furthermore, the values of electronic polarizability differ with various types of compounds. In other words, the values of polarizability depend on the density of charge distribution in both negative and positive ions. Hence, the ZBTSm glass has a higher density of charge distribution than the other glasses.

Moreover, the number of non-bridging oxygen has an important role in the tendency of glass to be polarized. Non-bridging oxygen has a lone pair which is independent of the chemical bonding and high mobility. ZBTSm glass may have a higher number of non-bridging oxygen than the other glass which contributes to high electronic polarizability. According to Fajan's rule, the polarizing power of the cation increases with decreasing its size and number of filled orbitals and with increasing its positive charge [28]. It is known that the samarium has a lower number of occupied 4f orbitals than the other glass system. Hence, this effect may increase the polarizing power of the cation according to Fajan's rule. Besides that, Sm^{3+} trivalent ions have a higher positive charge which leads to higher electronic polarizability.

Previously, Dimitrov and Sakka, 1996 illustrated the computation of the polarization of oxide ions based on optical band gap energy [29]. The deformity of the oxide ion's electron cloud is wider than that of the cation. This is due to the increased tendency of the cation electron to hold onto the cationic charge. This effect will contribute to the cation's electron cloud being not particularly polarised. The correlation between $\sqrt{E_g}$ and 1-R_m/V_m was presented by Duffy and Ingram in 1991 for a large number of simple oxides as shown in the given equation:

$$E_g = 20(1 - \frac{R_m}{V_m})^2 \qquad (11)$$

The oxide ion polarizability can be obtained by the substitution of Equation (6) into Equation (4) as shown by the following formula:

$$\alpha_{O_2^-}(E_g) = \left[\frac{V_m}{2.52}\left(1 - \sqrt{\frac{E_g}{20}}\right) - \sum \alpha_i\right](N_{O_2^-})^{-1} \qquad (12)$$

This equation has been accepted to be compliant with heavy metal oxide glasses.

The obtained values of oxide ion polarizability for all glass samples are illustrated in Figure 9 and listed in Table 4. The pattern of oxide ion polarizability for all glass samples is as follows; ZBTSm>ZBTEr>ZBTHo>ZBTNd. It is noted that ZBTSm has a higher number of oxide ion polarizability which reflects the high density of non-bridging oxygen in the glass system. Samarium oxide may enhance the formation of non-bridging oxygen in the glass system than erbium oxide, holmium oxide and neodymium oxide. The free electrons of the oxides in ZBTSm glass are independent of the nuclear charge and chemical bonding which improve the oxide ion polarizability. Moreover, the existence of localized states in the forbidden gap for different compounds leads to variations of oxide ion polarizability. The low number of polarizabilities in ZBTNd glass indicates that the neodymium oxide has low tendency to form the non-bridging oxygen in the tellurite glass as compared to other constituents.

Optical basicity and metallization criterion

The deep understanding of optical basicity was described by the work of Duffy and Ingram, 1971. Duffy and Ingram, 1971 proposed the theoretical calculation of the optical basicity for the multi-component oxide glasses as given below [30]:

$$\Lambda = X_1\Lambda_1 + X_2\Lambda_2 + \ldots\ldots + X_n\Lambda_n \qquad (13)$$

Where X_1, X_2,…., X_n correspond to the equivalent fractions of each oxide which contributes to the overall material stoichiometry and Λ_1, Λ_2,….,Λ_n correspond to the optical basicity of each individual oxide in the glass system.

The obtained values for the optical basicity of all glasses are shown in Figure 10 and Table 4. The pattern of optical basicity for the glasses is as follows; ZBTNd>ZBTHo>ZBTSm>ZBTEr. The low number of optical basicity for ZBTEr glass indicates higher in acidity. Meanwhile, the high number of optical basicity for ZBTNd glass corresponds to higher in basicity. It is known that a single neodymium oxide has higher number of optical basicity Λ (1.014) than erbium oxide Λ (0.929).

Hence, it is noted that ZBTNd glass has greater number of optical basicity than ZBTEr. Meanwhile, optical basicity values for holmium and samarium oxides are Λ (0.945) and Λ (0.984), respectively. The difference in optical basicity for single oxide contributes to the variations of overall optical basicity.

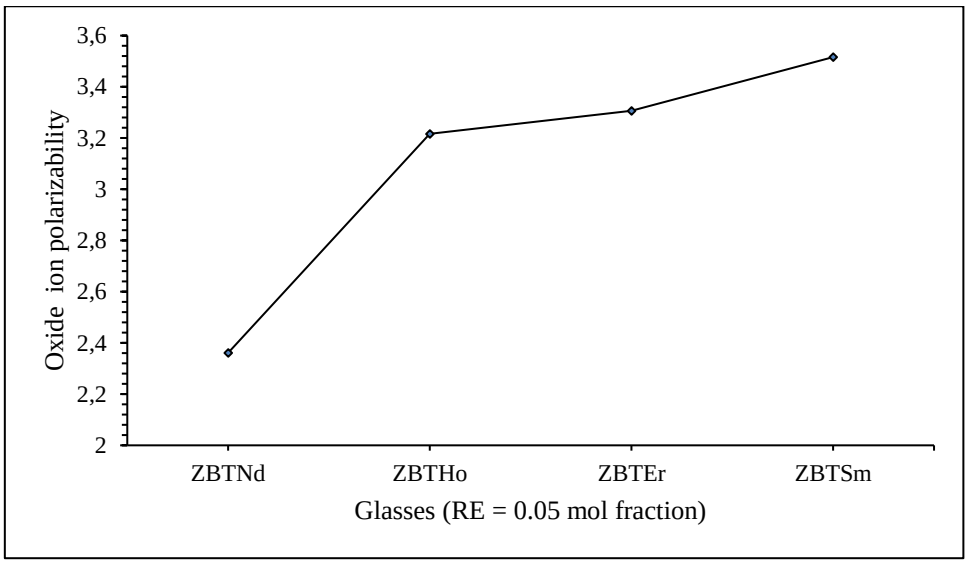

Figure 9 - Oxide ion polarizability of ZBTNd, ZBTHo, ZBTEr and ZBTSm glasses

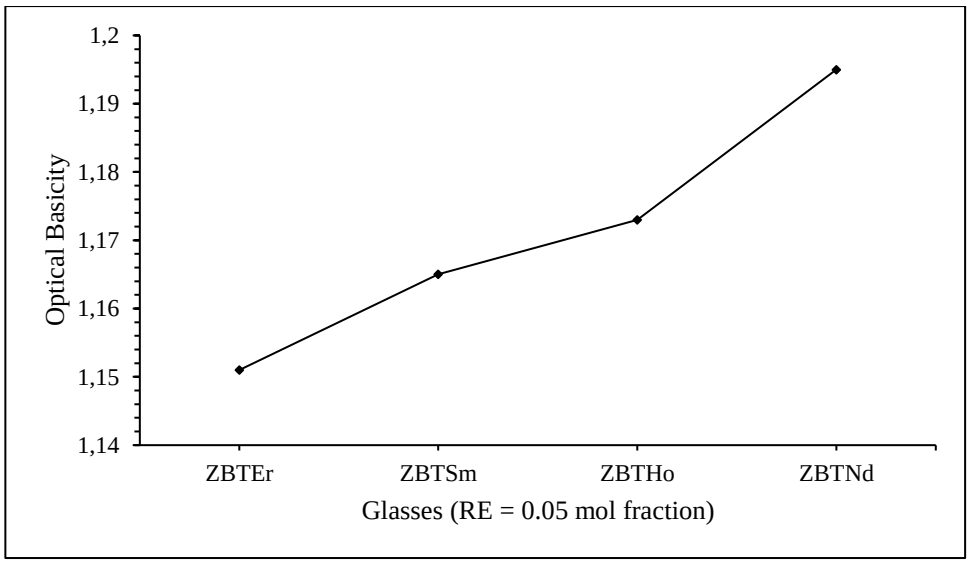

Figure 10 - Optical basicity of ZBTEr, ZBTSm, ZBTHo and ZBTNd glasses

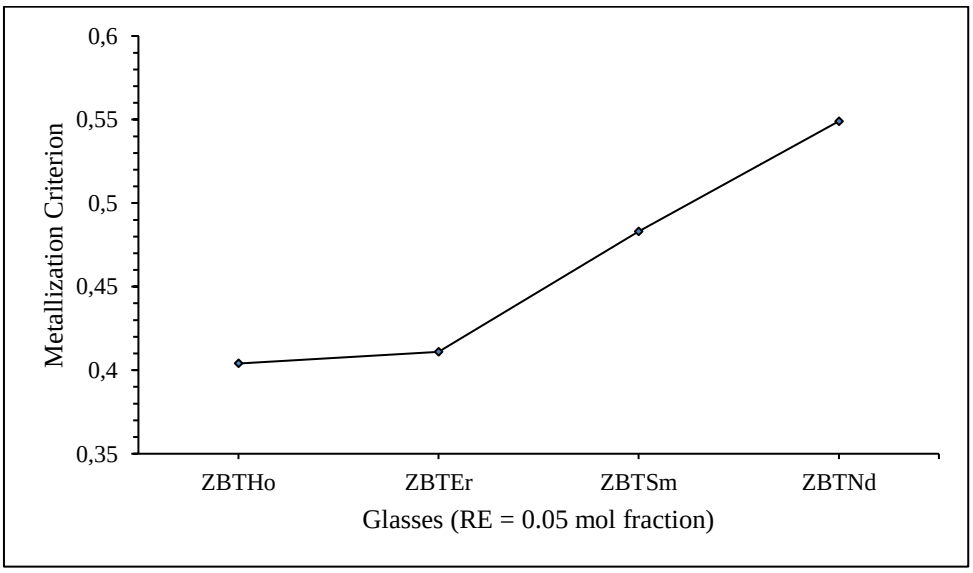

Figure 11 - Metallization criterion of ZBTHo, ZBTEr, ZBTSm and ZBTNd glasses

The concept of metallization of the condensed matter can be explained by the theory proposed by Herzfeld [31]. The condition of R_m/V_m= 1 in the Lorentz-Lorenz equation describes that the refractive index becomes infinite. This is in accordance with the metallization of covalent solid materials. In other words, the electrons become itinerant and acquires metallic status. The nature of metallic and non-metallic of oxide glasses can be predicted by the following conditions: $R_m/V_m<1$ (non-metal) and $R_m/V_m>1$ (metal). Subtracting by 1 gives the equation of metallization criterion as shown in the following expression:

$$M = 1 - \frac{R_m}{V_m} \quad (14)$$

This equation indicates that when the metallization criterion becomes zero, the transition to the metal states will occur. The metallization criterion based on refractive index and optical band gap can be calculated by transforming Equation (10) to the following expression:

$$M = 1 - \frac{(n_0^2 - 1)}{(n_0^2 + 2)} = \left(\frac{E_g}{20}\right)^{\frac{1}{2}} \quad (15)$$

The obtained values of the metallization criterion for all glasses are depicted in Figure 11 and tabulated in Table 4. The pattern of metallization criterion for the glasses is as follows; ZBTNd>ZBTSm>ZBTEr>ZBTHo. It is noted that all glasses are non-metallic and have a relatively large refractive index. The lower number of metallization criteria for ZBTHo indicates that the glass containing holmium oxide tends to be more metallic than neodymium oxide. Meanwhile, the high number of metallization criteria for ZBTNd shows the tendency of glass to be more insulator. The high metallization criterion reflects the widen of the forbidden gap and reduction in overlapping electronic transition. However, all glasses are poor conductive materials and good for semiconductor devices.

Conclusions

The glass series identified as ZBTSm, ZBTHo, ZBTEr, and ZBTNd were fabricated using the conventional melt-quenching method, leading to distinct physical and optical properties among them. The density of these glasses showed a pattern of ZBTHo>ZBTSm>ZBTEr>ZBTNd, with ZBTHo glass being highlighted as an ideal material for radiation shielding. In terms of refractive index, the order was ZBTSm>ZBTHo>ZBTNd>ZBTEr, positioning ZBTEr glass as a promising candidate for optical communication applications due to its low dispersion. Optical absorption trends indicated a shift to longer wavelengths in the order of ZBTEr > ZBTHo > ZBTNd > ZBTSm. Additionally, the optical band gap followed the sequence ZBTEr>ZBTHo>ZBTNd>ZBTSm, influenced by Coulomb repulsion energy in rare-earth oxides. The electronic polarizability adhered to the pattern of ZBTSm>ZBTNd>ZBTEr>ZBTHo, with ZBTSm glass exhibiting a higher charge distribution density, enhancing its electronic polarizability. These findings underscore the exceptional optical characteristics of the selected rare-earth oxides, underscoring their utility in optical communications applications.

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Мырыш-боротеллурит шынысына неодим, гольмий, эрбий және самарий оксидтерін енгізу, физикалық және оптикалық салыстырмалы талдау

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ТҮЙІНДЕМЕ

Сирек жер элементтері оксидтерінің әртүрлі типтерінің мырыш боротеллурит шынысына әсерін зерттеу оптикалық құрылғыларда әлеуетті қолдану мүмкіндігін анықтау үшін маңызды. Мырыш боротеллуритті шыныға сирек жер элементтері оксидтерін қосу 4f-4f ауысуларының әсерінен оптикалық қасиеттерді жақсартатыны белгілі. Бұл жұмыста ZBTNd, ZBTHo, ZBTEr және ZBTSm деп белгіленген мырыштың боротеллурит шынысына әртүрлі сирек жер элементтері оксидтерінің әсерін салыстыру мақсат етілген. Шыны үлгілері балқыту әдісімен дайындалды. Шыныларға физикалық зерттеу тығыздық пен молярлық көлемді өлшеу арқылы жасалды. Неодим оксидіндегі атомдық радиус аз болғандықтан, ZBTNd шынысының басқа шыныларға қарағанда ең төмен тығыздығы бар екені анықталды. ZBTHo шынысының жоғары тығыздығы оның радиациядан қорғау ретінде пайдалану мүмкіндігін көрсетеді. ZBTNd шыны үшін молярлық көлемнің жоғары мәні тиімді талшықты оптика болып табылады, өйткені ZBTNd шыны серпімділік бойынша жақсы көрсеткішке ие. ZBTEr төмен дисперсиялық сипаттамаларға ие болғандықтан басқа әйнектерге қарағанда сыну көрсеткіші төмен екені анықталды. Дегенмен, ZBTEr әйнегі оптикалық байланыс қолданбалары ретінде пайдалану үшін жақсы көрсеткішке ие. Оптикалық сіңіру ZBTEr > ZBTHo > ZBTNd > ZBTSm бастап ұзағырақ толқын ұзындығына ауысатыны анықталды. ZBTEr шынысының оптикалық диапазондық энергиясы эрбийдің кулондық тебілу энергиясына байланысты басқа шыныларға қарағанда жоғары, ол неодим мен самарийден үлкен және гольмиден сәл жоғары. Барлық шынылардың электрондық поляризациялық үлгісі келесідей ZBTSm>ZBTNd>ZBTER>ZBTHo болады. ZBTEr үшін оптикалық негізділік ең жоғары болды, бұл қышқылдықтың жоғары екенін көрсетеді, ал ZBTNd шынысының қышқылдығы төмен, бұл негізділіктің жоғарырақ мәніне сәйкес келеді..

Түйін сөздер: теллуритті шыны, сирек жер элементтері оксидтері, оптикалық қасиеттер.

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Внедрение неодима, гольмия, эрбия и самария (оксидов) в цинкборотеллуритное стекло: физический и оптический сравнительный анализ

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АННОТАЦИЯ

Исследование влияния различных типов оксидов редкоземельных элементов на цинкборотеллуритное стекло важно для определения потенциального применения в оптических устройствах. Известно, что добавление оксидов редкоземельных элементов в цинкборотеллуритное стекло улучшает оптические свойства за счет эффектов 4f-4f-переходов. В данной работе мы стремимся сравнить влияние различных оксидов редкоземельных элементов на цинкборотеллуритное стекло, обозначенное как ZBTNd, ZBTHo, ZBTEr и ZBTSm. Образцы стекла были успешно изготовлены методом закалки в расплаве. Физическое исследование стекол было проведено путем измерения плотности и молярного объема. Установлено, что стекло ZBTNd имеет самую низкую плотность по сравнению с другими стеклами из-за малого атомного радиуса оксида неодима. Высокая плотность стекла ZBTHo указывает на его потенциал для использования в качестве защиты от радиации. Высокое значение молярного объема стекла ZBTNd является преимуществом оптоволокна, поскольку стекло ZBTNd имеет хорошие показатели эластичности. Установлено, что ZBTER имеет более низкий показатель преломления, чем другие стекла, из-за низких дисперсионных характеристик. Тем не менее, стекло ZBTER имеет хорошие характеристики для использования в приложениях оптической связи. Обнаружено, что оптическое поглощение смещается в длинноволновую область, начиная с ZBTEr > ZBTHo > ZBTNd > ZBTSm. Энергия оптической запрещенной зоны у стекла ZBTER выше, чем у других стекол, из-за энергии кулоновского отталкивания эрбия, которая больше, чем у неодима и самария, и немного выше, чем у гольмия. Характер электронной поляризуемости для всех стекол был найден следующим образом: ZBTSm>ZBTNd>ZBTER>ZBTHo. Оптическая основность ZBTEr оказалась самой высокой, что указывает на более высокую кислотность, в то время как стекло ZBTNd имеет самое низкое значение, соответствующее более высокой основности.

***Ключевые слова*:** теллуритное стекло, оксиды редкоземельных элементов, оптические свойства

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