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# Effect of Na<sub>2</sub>O/PbO substitution on physical and optical properties of Er<sup>3+</sup>-doped tellurite glasses

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Abstract. Er<sup>3+</sup>- doped tellurite glasses with chemical compositions (in mol%): 55TeO<sub>2</sub>-35ZnO-(5+x)PbO-2Bi<sub>2</sub>O<sub>3</sub>-(2-x)Na<sub>2</sub>O-Er<sub>2</sub>O<sub>3</sub> with x = 0; 0.5; 1.0; 1.5 and 2.0) were synthesized and explored their physical and optical properties. Density and refractive index were measured at room temperature and other physical properties such as ionic packing ratio, field strength, polaron radius, internuclear distance and molar refraction were derived. It was found that Na2O substitution by PbO causes both refractive index and glass density increase.

## 1. Introduction

TeO<sub>2</sub> based glass has been regarded as one of glass family characterized by its high stability against crystallization. The stabile glass belong to this glass family can be easily fabricated. Because of this reason, nowadays this glass family has become the subject of many studies, both mechanically, electricity, optically, magnetically, and physically. This glass is known to have a low corrosion resistance, high transparency, high refractive index value and high rare-earth ions solubility [1, 2].

Basically, tellurite cannot form glass by itself. This is because tellurite is a conditional glass former. Therefore, this material is only capable of forming glass when is combined with network modifying elements such as alkali metals, alkaline earth, heavy metals, transition metals, or oxides [3, 4]. Rodin and Sahar [5] added Na<sub>2</sub>O compound as a glass modifier to break the unit bond structure or increase the entropy value which can result in the molten tend to form a glass. Other compounds usually added to TeO<sub>2</sub> to form a glass are PbO, ZnO, and Bi<sub>2</sub>O<sub>3</sub>. PbO which is known for its highly ionic polarizability is added in order to increase the refractive index value, density, and bandgap energy. ZnO is added mainly because of its ability to enhance its ability to form glass optical properties of glass. Whereas, Bi<sub>2</sub>O<sub>3</sub> is usually added by considering its ability to create a glass having high refractive index and wide infra-red transmission [6-9].

Nowadays, research to seek a glass host for lasers, optical amplifiers and sensors has been one of the most attractive subjects. For this purpose, a certain amount of one or more rare earth ions is added into a stabile glass. Among them, erbium ion doped glass has been widely used in optical fibre amplifiers mainly due to its unique energy levels scheme resulting in emissions which are very useful in optoelectronics applications [10, 11, 12]. In order to fabricate  $Er^{3+}$ -doped glass to form an optical fibre or planar waveguide, glass composition needs to be slightly modified such that two nearly the same glass composition having a certain refractive index difference can be obtained. For this purpose, works devoted to study the effect of such compositional modification on the change in refractive index have been reported [13, 14, 15]. This work presents the effect of PbO/Na<sub>2</sub>O on physical and physical

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properties of  $Er^{3+}$ -doped glass. Discussion will be limited to its effect on change in density, oxygen packing density, polaron radius, inter-ionic distance, Field Strength and refractive index.

# 2. Experimental

Compositions of all tellurite glasses reported in this paper are tabulated in Table 1. Raw materials are prepared in glove box. Melting was carried out by placing raw materials in a platinum crucible and heated in an electric tube furnace at temperature 900°C for 30 minutes. In order to get homogeneous sample, molten was stirred twice. Casting was carried out by pouring the molten into a plate brass mould preheated at temperature of 245°C. Sample was subsequently annealed at temperature 260°C for 6 hours followed by cooling to room temperature at a cooling rate of 1°C/min. For optical characterization purpose, samples were cut to the size of 1.5 cm x 1.5 cm and polished down to optical quality.

Table. T Chemical composition and name of the samples							
Sample	mol% Composition						
	$TeO_2$	ZnO	PbO	Bi <sub>2</sub> O3	Na <sub>2</sub> O	Er <sub>2</sub> O <sub>3</sub>	
TZPBN 0	55	35	5.0	2	2.0	1	
TZPBNE 0.5	55	35	5.5	2	1.5	1	
TZPBNE 1.0	55	35	6.0	2	1.0	1	
TZPBNE 1.5	55	35	6.5	2	0.5	1	
TZPBNE 2.0	55	35	7.0	2	0.0	1	

Table. 1 Chemical composition and name of the samples

Glass characterization has been carried out to determine the physical and optical properties of glass. Density was measured using pycnometer and refractive index was measured by applying Brewster's angle method at room temperature using a spectrometer and a red laser pointer with a wavelength 642.5  $\pm$  0.3 nm. All measurements were carried out at room temperature.

# 3. Results and Discussion

### 3.1. Density

Glass density is a fundamental and significant part of the physical properties of glass. The change in density can be used to confirmed the compactness and structural change in the glass sample [13]. Density measurement was carried out using distilled water as an immersion liquid using equation:

$$\rho_{glass} = \frac{W_1}{W_1 - W_2} \rho_2 \tag{1}$$

where  $W_1$  is the weight of sample in air,  $W_2$  is the weight of sample in distilled water, and  $\rho_2$  is the density of distilled water [14]. From glass density, the molar volume of glass can then be calculated using equation:

$$V_m = \frac{M_{glass}}{\rho_{glass}} \tag{2}$$

where  $M_{glass}$  is the molecular wight of glass and  $\rho_{glass}$  is the density of the sample [15].

Knowing the  $V_m$ , oxygen packing density (OPD) which determines the arrangement of oxygen atoms in the oxide glass and can be estimated using equation:

$$OPD (g.atom/l) = \frac{1000 \times O}{V_m}$$
(3)

where *O* is the total number of oxygen atom in oxide glass. Internuclear properties can be calculated to determine the glass-forming structural system, including ion concentration  $\text{Er}^{3+}(N_{Er})$ , polaron radius  $r_p$ , inter-ionic distance  $r_i$ , and Field Strength *F* using the following equations:

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$$N_{\rm Er} (\text{ions or atom/cm}^3) = A_p \times N_A \times \rho_a / M_w$$
(4)

$$r_{p}(\text{\AA}) = \frac{1}{2} \left(\frac{\pi}{6N_{Er}}\right)^{1/3}$$
(5)

$$r_{i}(\text{\AA}) = \left(\frac{1}{N_{Er}}\right)^{1/3}$$
(6)  
$$F(\text{cm}^{-2}) = \frac{Z}{r_{p}^{2}}$$
(7)

where  $A_p$  is the mol% of  $Er^{3+}$  ion,  $N_A$  is the Avogadro number,  $\rho_g$  is the density of glass,  $M_w$  is the average molecular weight of the glass sample, and Z is the mass of dopant molecule [16]. The changes in density and molar volume of the glass samples are depicted in Figure 1 and the calculated values of  $N_{Er}$ ,  $r_p$ ,  $r_i$ , and F are tabulated in Table 2.



Figure 1. Density and molar volume of the glass sample at mol% PbO

It is shown that density increases and the molar volume decreases with the increase of PbO concentration. This is because the molar mass of PbO is greater than Na<sub>2</sub>O (PbO = 223.2 g/mol and Na<sub>2</sub>O = 105.9888 g/mol) [17]. Following equation (3), OPD is inversely proportional to  $V_m$  and molar volume is related to interatomic distance, the decrease of molar volume as the PbO concentration increase will result in increasing  $r_p$  and  $r_i$ . With this reason, it can be expected that the field strength will also increase with increasing PbO concentration.

Tabel 2. Internuclear properties

Sampel	OPD (g.atom/l)	N <sub>Pb</sub> (ions/cm <sup>3</sup> )	$r_p(\text{\AA})$	$r_i$ (Å)	$F(x10^{-15} \text{ cm}^{-2})$
TZPBN 0.0	64.2460	1.2017	0.3790	0.9406	1.3924
TZPBNE 0.5	64.4925	1.3270	0.3667	0.9100	1.4875
TZPBNE 1.0	64.3174	1.4437	0.3565	0.8848	1.5735
TZPBNE 1.5	64.7729	1.5750	0.3463	0.8595	1.6676
TZPBNE 2.0	64.8103	1.6971	0.3378	0.8383	1.7528

#### 3.2. Refractive index

The Refractive index was measured using the Brewster angle using equation (18):

$$n = \tan \theta_p \tag{8}$$

where  $\theta_p$  and n are Brewster's angle and refractive index, respectively. The molar refraction  $R_m$  of amorphous materials can be determined by the Lorentz-Lorenz equation:

$$R_m = \frac{(n^2 - 1)}{(n^2 + 2)} V_m \tag{9}$$

where  $V_m$  is molar volume and n is refractive index. The molar refraction is proportional to the molecular polarizabilities  $\alpha_m$  as expressed in equation:

$$R_m = \frac{4\pi N_A}{3} \alpha_m = 2.52 \ \alpha_m \tag{10}$$

where  $\alpha_m$  used in (Å<sup>3</sup>) units [19]. The energy gap  $E_g$  can be determined equation:

$$E_g = 20 \left( 1 - \frac{Rm}{Vm} \right)^2 \tag{11}$$

where  $R_m$  is molar refraction and  $V_m$  is molar volume. The electric constant is the square of refractive index

$$\varepsilon = n^2 \tag{12}$$

where n is the refractive index of glass sample [20].



Figure 2. Measured refractive index and predicted refractive index

Figure 2 shows how the change in PbO concentration in the glass affect the change in refrative index. As shown the measured and the calculated values of refractive index tend to have the same trend that refractive index increase with the increase of PbO content. This results is linked to the higher value of ionic polarizability of  $Pb^{2+}$  as compared to that of  $Na^+$ .

Table 3 shows how the change in PbO content effect the value of  $V_m$ ,  $R_m$ ,  $\alpha_m$ ,  $\varepsilon$  and  $E_g$ . As seen  $V_m$ ,  $R_m$ ,  $\alpha_m$  and  $\varepsilon$  increase with the increase of PbO content while  $E_g$  decreases with the increase of PbO content suggesting that glass become less electrically conductive with the increase of PbO content.

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Sampel	$V_m$ (cm <sup>3</sup> )	$R_m$ (cm <sup>3</sup> )	$\alpha_m(\text{\AA}^3)$	Eg	3
TZPBN 0.0	25.0599	9.7816	3.8816	7,4340	2.9207
TZPBNE 0.5	24.9641	10.2277	4.0586	6,9692	3.0821
TZPBNE 1.0	25.0320	10.3764	4.1176	6,8556	3.1241
TZPBNE 1.5	24.8560	11.8720	4.7111	5,4574	3.7431
TZPBNE 2.0	24.8417	12.4679	4.9476	4,9622	4.0228

**Table 3.** Molar volume  $V_m$ , molar refraction  $R_m$ , electrical polarizability  $\alpha_m$ , energy gap  $E_g$ , and dielectric constant  $\varepsilon$ 

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### 4. Conclusions

 $Er^{3+}$  doped tellurite glass has been successfully prepared with a composition of 55TeO<sub>2</sub>-35ZnO-(5+x)PbO-2Bi<sub>2</sub>O<sub>3</sub>-(2-x)Na<sub>2</sub>O-xEr<sub>2</sub>O<sub>3</sub> where x = 0; 0.5; 1.0; 1.5; 2.0 mol%. From the results of the study, the density and refractive index increased along with the increase of PbO. Based on our calculation, it was found that the increase of PbO content increases the value of molar refractivity, electrical polarizability, dielectric constant and field strength but decreases the value of polaron radius and interatomic distance.

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