

# Quantifying Physical Parameters of Quaternary Glass with Composition Tellurium- Niobium - Zinc-Lithium Oxide

A.E.Al-Salami

**Abstract:** In the present work the tellurite glass a function of alkali metals as network intermediate with composition 85TeO<sub>2</sub>/5Nb<sub>2</sub>O<sub>5</sub>/5ZnO/5Li<sub>2</sub>O in mol% were been prepared. These glasses contain Li<sub>2</sub>O can be promising used in optical devices because it has advantage optical properties compared with other tellurite glasses. The physical parameters of these glass studied with respect to values of density,  $\rho$ , molar volume,  $V_m$ , linear refractive indices,  $n_0$ , nonlinear refractive index,  $n_2$ , electronic polarizability,  $\alpha_m$ , molar refraction,  $R_m$ , metallization criterion,  $M(n_0)$ , and third order non-linear optical susceptibility,  $\chi^{(3)}$ . The Sellmeier gap energy,  $E_s$ , and dispersion energy,  $E_d$  were calculated besides the theoretical third order of nonlinear optical susceptibility,  $\chi^{(3)}$ , calculated by used Lines model. Measure the thermal stability and the glass transition temperature by differential thermal analysis (Shimadzu DTA 50) which can be determine the glass transition temperature,  $T_g$ , onset crystallization temperature,  $T_c$ , and melting temperature,  $T_m$ . These glass have values as;  $\rho= 5.231 \text{ gm}\cdot\text{cm}^{-3}$ ,  $V_m= 28.75 \text{ cm}^{-3}$ ,  $n_0= 2.2228$  at 435.84 nm,  $\alpha_m= 6.478\text{\AA}^3$  and  $R_m=16.024 \text{ mol}^{-1}\cdot\text{cm}^3$  at 435.84nm. and  $E_s = 6.54 \text{ eV}$ ,  $E_d= 20.65 \text{ eV}$ .

**Keyword:** Oxide glass; linear refractive index; dispersion energy.

## I. INTRODUCTION

Tellurium dioxide based glasses are of scientific merit in view of their use in optical devices, which are currently being studied for use in telecommunications and computational system due to large optical nonlinearity [1]. In normal glass system, the modifier atoms cause the network to break. The structure of TeO<sub>2</sub> rich glasses consists of units such as TeO<sub>4</sub> having trigonal bipyramid (tbp), polyhedron (TeO<sub>3+1</sub>) and TeO<sub>3</sub> trigonal byramid (tp) structure with one of the equatorial position occupied by a lone pair of electrons. When a network modifier like metal oxide is added into the glass matrix, one of the Te- Oax bonds in TeO<sub>4</sub> polyhedra elongates, the bond length increases from 0.208 to 0.298 nm and TeO<sub>3+1</sub> structure are formed. The subscript 3+1 indicates that the fourth oxygen is nearby but is not within a true bonding range. When the Te-O bond length exceeds 0.298 nm, we get TeO<sub>3</sub> structural unit, i.e. TeO<sub>4</sub> converts to TeO<sub>3</sub> structural unit. TeO<sub>4</sub>, TeO<sub>3+1</sub> and TeO<sub>3</sub> structural units are usually labeled as  $Q_4^4$ ,  $Q_4^3$  and  $Q_3^3$ , respectively [2]. Here the subscript represents the coordination number of oxygen around Te atom and the superscript is the number of bridging oxygens linked to a Te atom. Many authors [3, 4] studied linear refractive index, linear refractive index, this order of

nonlinear susceptibility and optical energy gap of ternary glasses with composition TeO<sub>2</sub>/ Nb<sub>2</sub>O<sub>5</sub>/ZnO. They estimated that these glasses had high value of nonlinear optical response time and it can be used as suitable for all optic switching application. Yousef et al. [4] reported the value of,  $\chi(3)$ , for TeO<sub>2</sub>- Nb<sub>2</sub>O<sub>5</sub>- ZnO- CdS was equal  $7.8 \times 10^{-13}$  esu, hence these glass indicates that promising used for optoelectronic application. The linear refractive index,  $n_0$ , is related to the frequency,  $\nu$ , of light as follow [5];

$$n_0^2 - 1 = \frac{4\pi N e^2}{m} \sum_i \frac{f_i}{\nu_i^2 - \nu^2} \quad (1)$$

Where,  $e$ , is the electronic charge,  $m$ , is the mass charge and mass,  $N$ , is the number of molecules in a unit volume,  $\nu_i$ , is the resonance frequency, and,  $f_i$ , is the oscillator strength. When applied the Equ. (1) on the simple a single- oscillator model [6].

$$n_0^2 = \frac{4\pi N e^2}{m} \frac{f}{\nu_0^2 - \nu^2} + 1 \quad (2)$$

Moreover we can rewrite the Equ. (2) in formula as follow;

$$\frac{1}{n_0^2 - 1} = \frac{\pi m c^2}{e^2 N f} \left( \frac{1}{\lambda_0^2} - \frac{1}{\lambda^2} \right) \quad (3)$$

Where,  $c$ ,  $\lambda_0$ , and  $\lambda$  values are velocity of light, average resonance wavelength and wavelength of light, respectively. From the relation between,  $n_0$ , and  $\lambda$  in Equ. 3 obtained that the refractive index decreases with increasing wavelength. Although different oscillators like that bridging oxygens (BO), nonbridging oxygens (NBO), and cations exit practically in oxide glasses. Also it can be determine the parameter  $\lambda_0$  experimently from the graph of  $1/(n_2-1)$  versus  $1/\lambda^2$ , besides average oscillator strength,  $f$ , was computed from the values of the slope  $\left[ \left( \frac{\pi m c^2}{e^2} \right) \cdot (1/Nf) \right]$  and  $N$ .

The aim of the present paper computational physical prameters especially optical parameter provides the importance of the prepared glass can be used in optical application like that the nonlinear optical devices.

## II. PROCEDURE FOR PAPER SUBMISSION

Glasses with composition 85TeO<sub>2</sub>/ 5Nb<sub>2</sub>O<sub>5</sub>/ 5ZnO/ 5Li<sub>2</sub>O in mol% were melted with reagent grade ingredients of TeO<sub>2</sub> and pre-dried oxides; Batches (50g) were directly melted in an Au crucible.



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A.E.Al-salami, Department of Physics, King Khalid University, Faculty of Science/ P.O. Box 9004, Abha, Saudi Arabia.

**Table 1: Quantifying physical parameters of glass  
85TeO<sub>2</sub>- 5Nb<sub>2</sub>O<sub>5</sub>- 5ZnO- 5Li<sub>2</sub>O in mol% have been studied.**

Parameters	at different wavelength (in nm)				
	435.84	479.98	546.06	587	643.8
Density, $\rho$ , (in g·cm <sup>-3</sup> )	5.23				
Molar volume, $V_m$ , (in cm <sup>3</sup> )	28.75				
Molar volume of oxygen, $V_0$ , (in cm <sup>3</sup> )					
Refractive index, $n_0$ ,	2.2228	2.1857	2.1508	2.1358	2.1208
Abbe number, $\nu_d$ ,	17.7				
Molar refraction, $R_m$ , (in cm <sup>3</sup> · mol <sup>-1</sup> )	16.324	16.024	15.733	15.65	15.476
Polarizability, $\alpha_m$ , (in Å <sup>3</sup> )	6.478	6.359	6.243	6.193	6.141
Metallization criterion, $M(n_0)$	0.432	0.443	0.453	0.456	0.462
Sellmeier energy gap, $E_s$ , (in eV)	6.43				
Dispersion energy, $E_d$ , (in eV)	20.45				
Band gap, $E_g$ , (in eV)					
$n_2$ , in 10 <sup>-14</sup> cm <sup>2</sup> /W	8.4				
$\chi_{th. Lines}^{(3)} / d^2$ in 10 <sup>-13</sup> esu/ Å <sup>2</sup>	2.55				

The batches were given to a gold crucible and heated at a temperature of 850°C. The melt was allowed to cool to 750 °C and then cast in a graphite mould. Subsequently, the samples were transferred to an annealing furnace and kept at 300 °C for 2 h. Then the furnace was switched off and allowed to cool. From the glassy samples, prisms of the dimension 30x15x15 mm<sup>3</sup> were cut. The prisms were ground and polished using water as liquid component. The prisms were used to measure the linear refractive indices at wavelengths of 643.8, 589.3, 546.1, 479.98 and 435.8 nm.

The densities were measured by a helium pycnometer (AccuPyc 1330) with an accuracy of  $\pm 0.03$  %.

The prepared glass was examined by X-ray diffraction, (Siemens D 6000) using CuK $\alpha$  radiation at 40 kV in the 2 $\theta$  range from 5 to 90<sup>0</sup>.

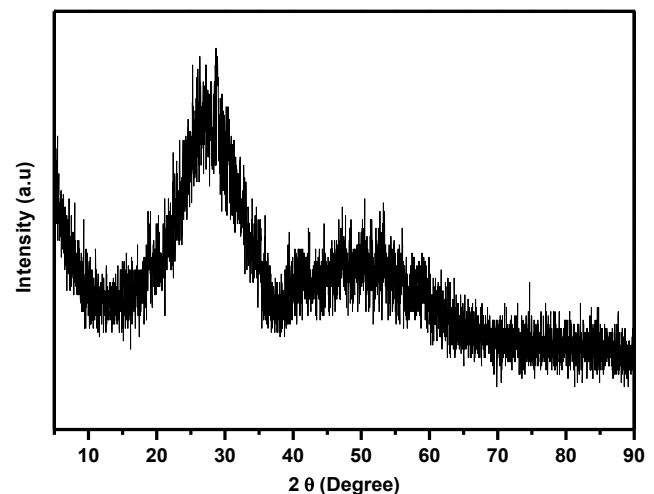
The glass samples were powdered in order to perform differential thermal (DTA) analysis using a Shimadzu DTA 50 instrument with a heating rate of 15 °C/min under air-atmosphere with an accuracy of  $\pm 3$  °C.

### III. RESULT AND DISCUSSION

Figure 1 show the X- ray diffraction patterns (XRD) of present glass with composition 85TeO<sub>2</sub>/ 5Nb<sub>2</sub>O<sub>5</sub>/ 5ZnO/ 5Li<sub>2</sub>O in mole% and it is not show any sharp peaks caused by crystalline phases. This indicate that The disappearance of sharp diffraction peaks indicates that the nonexistence of crystalline phase and the broad diffraction that's confirmed that amorphous nature of the prepared glasses.

#### A. Density and molar volume

The densities and molar volume of the prepared glasses are summarized in Table (1). The sample with composition 85TeO<sub>2</sub>/ 5Nb<sub>2</sub>O<sub>5</sub>/ 5ZnO/ 5Li<sub>2</sub>O in mol % has value of density ( $\rho = 5.231$  g.cm-3). The change in the density attributed to addition of atomic mass and atomic volume of



**Fig 1: X- ray diffraction patterns of prepared glass.**

modifier in the glass matrix. Also when oxide has low molecular weight substituted by a higher molecular weight leads to increase of density. So the density value of present glass lower than the density of other tellurite glasses systems [7] within composition such as 80TeO<sub>2</sub>- 5Nb<sub>2</sub>O<sub>5</sub>- 15ZnO- 5AgO ( $\rho = 5.3774$  g.cm-3), 75TeO<sub>2</sub>- 5Nb<sub>2</sub>O<sub>5</sub>- 15ZnO- 5TiO<sub>2</sub> ( $\rho = 5.3647$  g.cm-3) and 68TeO<sub>2</sub>- 5Nb<sub>2</sub>O<sub>5</sub>- 20ZnO- 7PbO ( $\rho = 5.6637$  g.cm-3).

#### B. Thermal parameters

The thermal profile in Fig. 2 of present glass clearly to determine the glass transition temperature,  $T_g$ ; softening temperature,  $T_s$ ; temperature for fiber fabrication,  $\Delta F = (T_s - T_g)$ ; onset crystallization temperature,  $T_c$ ; thermal glass stability  $\Delta T = (T_c - T_g)$ ; the peak of crystallization temperature,  $T_p$ ,

melting temperature,  $T_m$ , and finally thermal stability of anticrystallization,  $S$ , can be calculated as follow;  $S = \frac{\Delta T \cdot (T_p - T_c)}{T_g}$ , The results are obtained in Table (1), we know that the increase in  $T_g$  is an indication of strong

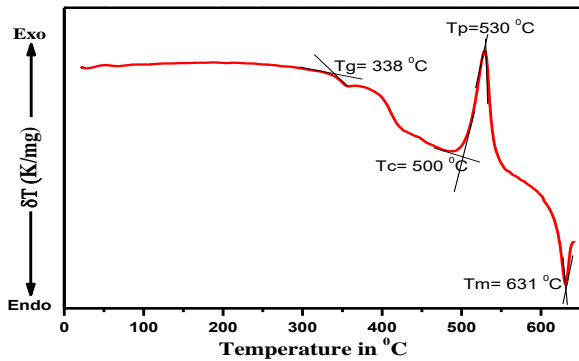


Fig 2: DTA curve of prepared glass.

bonding between atoms in the glass matrix and increase in the rigidity of the network modifier  $\text{Li}_2\text{O}$ . The present glass has values of  $T_g = 338^\circ\text{C}$ ,  $\Delta T = 162^\circ$ ,  $\Delta F = 62^\circ\text{C}$ , and  $S = 14.38$ . These parameters are very important for fiber drawing of performs and hence to avoid the crystallization, it is advantage that  $S$ , and  $\Delta T$  have value as large as possible. The present glass has a high  $S$  and a  $\Delta T$ , in comparison with other tellurite glasses. Moreover, the factor  $T_g/T_m$  is a good measure of the glass stability it is equal 0.54 this leads to say the prepared glasses is in an excellent stability.

C. Optical properties

The refractive index is important parameter used in the fabrication of optical fiber amplifiers. The Lorentz- Lorenz equation related to molar refraction with refractive indices and molar volume as estimated by Fajans and Kredidl [8- 10] as follow:

$$n_0 = \left( \frac{\left[ 1 + \frac{8\pi N_A \cdot \alpha}{3V_m} \right]}{1 - \frac{4\pi N_A \cdot \alpha}{3V_m}} \right)^{1/2} \quad (4)$$

Where,  $R_m$ , is the molar refraction,  $V_m$ , the molar volume and,  $n_0$ , the linear refractive index.

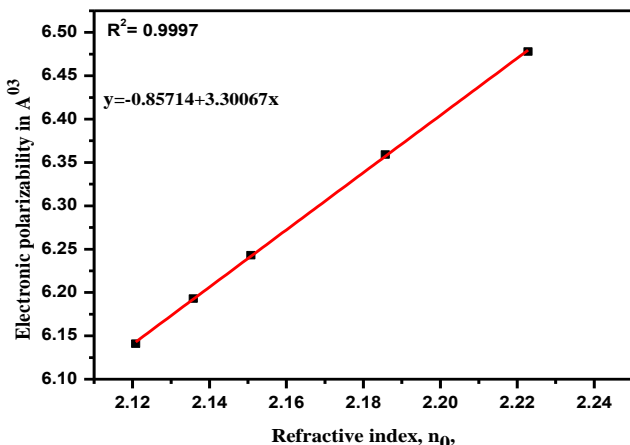


Fig 3: Relation between refractive index,  $n_0$ , and

electronic polarizability in  $\text{Å}^3$  at different wavelength of prepared glass.

Linear refractive index measured at the wavelengths 435.8, 479.98, 546.1 and 643.8 nm as well as the Abbe numbers calculated, polarizability,  $\alpha_m$  (in  $\text{Å}^3$ ) and molar refraction,  $R_m$  (in  $\text{cm}^3 \text{mol}^{-1}$ ) are summarized in Table 1. The value of linear refractive index,  $n_0$ , of glass  $85\text{TeO}_2/5\text{Nb}_2\text{O}_5/5\text{ZnO}/5\text{Li}_2\text{O}$  decreases from 2.2228 to 2.1208 when the wave length,  $\lambda$ , increase from 435.84 to 643.8nm and it has Abbe number ( $\nu = 17.7$ ). The results show that highest values of refractive index were obtained when lithium was incorporated into the tellurite in the present glasses by compare with other glasses system [7] like that  $80\text{TeO}_2-5\text{Nb}_2\text{O}_5-15\text{ZnO}-5\text{Ag}_2\text{O}$  ( $n_0 = 2.1684$  at 435.84nm),  $68\text{TeO}_2-5\text{Nb}_2\text{O}_5-20\text{ZnO}-7\text{PbO}$  ( $n_0 = 2.2123$  at 435.84nm) and  $57\text{TeO}_2-5\text{Nb}_2\text{O}_5-20\text{ZnO}-18\text{PbO}$  ( $n_0 = 2.2118$  at 435.84nm). We note that the change in the value of linear refractive indices related with the number of atoms/unit volume is not enough parameter to understand the increasing or decreasing in the value of refractive index,  $n_0$ , which can be attributed to other factor like that field strength. [11]. Here the prepared glass  $85\text{TeO}_2/5\text{Nb}_2\text{O}_5/5\text{ZnO}/5\text{Li}_2\text{O}$  has low high density does otherwise it has high value of refractive index. The refractive index is proportional to the inverted mole volume unit and the polarizability of ions in the glasses [12]. In the present paper the molar refraction,  $R_m$  and polarizabilities,  $\alpha_m$ , (in  $\text{Å}^3$ ), have been calculated using the following well-known Clasius- Mosotti relation [13- 14]:

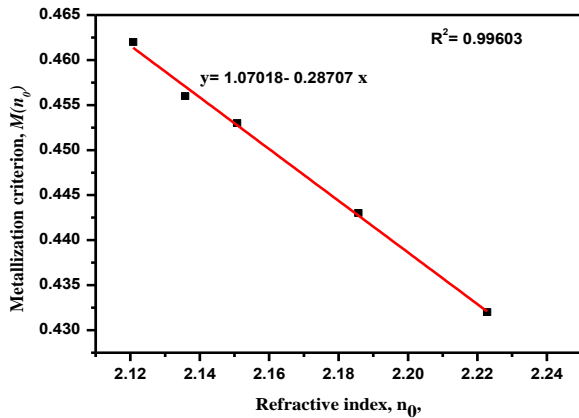
$$R_m = \left( \frac{n_0^2 - 1}{n_0^2 + 2} \right) V_m \quad (5)$$

$$\alpha_m = \frac{3}{4\pi N} \left( \frac{n_0^2 - 1}{n_0^2 + 2} \right) V_m \quad (6)$$

Where  $N$  is the Avagadro's number,  $V_m$  is the molecular weight and,  $\rho$ , is the density of the sample. The results of  $\alpha_m$  and  $R_m$  of glass  $85\text{TeO}_2/5\text{Nb}_2\text{O}_5/5\text{ZnO}/5\text{Li}_2\text{O}$  at different wavelength were obtained in table (1). Both the electronic polarizability and molar refraction are related to the structure of glass materials. Moreover the electronic polarizability is depend on the numerical value of the electrons responds to an applied electrical field. The relation between of electronic polarizability versus linear refractive index is shown in Fig. 3. It is found that from these figure the relation as; ( $\alpha_m = -0.85714 + 3.30067 \cdot n_0$ ). The prepared glass has highest value of electronic polarizability ( $\alpha_m = 6.478 \text{Å}^3$  at 435.84nm) and highest value of  $R_m = 16.024 \text{mol}^{-1} \cdot \text{cm}^3$  at 435.84nm by comparing with other tellurite glass systems with composition  $68\text{TeO}_2/5\text{Nb}_2\text{O}_5/20\text{ZnO}/7\text{Na}_2\text{O}$  ( $\alpha_m = 5.36 \text{Å}^3$ ,  $R_m = 13.5 \text{mol}^{-1} \cdot \text{cm}^3$  at 435.84nm),  $85\text{TeO}_2/5\text{Nb}_2\text{O}_5/5\text{ZnO}/5\text{Ag}_2\text{O}$  ( $\alpha_m = 6.38 \text{Å}^3$ ,  $R_m = 16.01 \text{mol}^{-1} \cdot \text{cm}^3$  at 435.84nm) and  $68\text{TeO}_2/5\text{Nb}_2\text{O}_5/20\text{ZnO}/7\text{PbO}$  ( $\alpha_m = 5.77 \text{Å}^3$ ,  $R_m = 14.55 \text{mol}^{-1} \cdot \text{cm}^3$  at 435.84nm). Hence it is concluding that the value of refractive index is strongly depends on the electronic poarizablty of the glass material, i.e. it is not only depend on the value of density of glass. Wang[13]



estimated the theory on metallization of condensed matter for different condition like that when,  $R_m/V_m=1$  , these means that the refractive index becomes infinite. It is in correspond to the metallization of covalent solid materials.

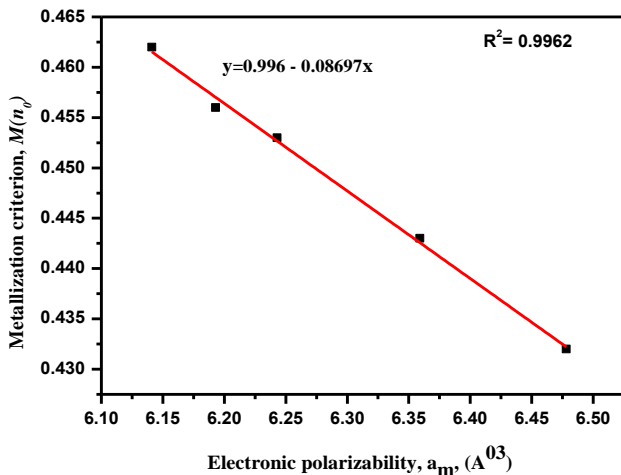


**Fig 4: Relation between refractive index,  $n_0$ , and metallization criterion,  $M(n_0)$  at different wavelength of prepared glass.**

Dimitrov and Komatsu [15] explained the condition for predicting the nature of material which the non-metallic ( $R_m/V_m < 1$ .) and the metal ( $R_m/V_m > 1$ ). The metallization criterion  $M(n_0)$  is subtracting from 1.0 as expression [14];

$$M(n_0) = 1 - \frac{(n_0^2 - 1)}{(n_0^2 + 2)} \quad (7)$$

The present glass  $85\text{TeO}_2/5\text{Nb}_2\text{O}_5/5\text{ZnO}/5\text{Li}_2\text{O}$  has  $M(n_0)=0.432$  at  $435.84\text{nm}$  lowest value comparing with  $\text{TeO}_2$  based glass within composition  $68\text{TeO}_2/5\text{Nb}_2\text{O}_5/20\text{ZnO}/7\text{Na}_2\text{O}$  ( $M(n_0) = 0.487$  at  $435.84\text{nm}$ ),  $85\text{TeO}_2/5\text{Nb}_2\text{O}_5/5\text{ZnO}/5\text{Ag}_2\text{O}$  ( $M(n_0) = 0.475$  at  $435.84\text{nm}$ ) and  $68\text{TeO}_2/5\text{Nb}_2\text{O}_5/20\text{ZnO}/7\text{PbO}$  ( $M(n_0) = 0.464$  at  $435.84\text{nm}$ ). According to the theory of metallization criterion when the width of both valence and conduction bands becomes large leads to a narrow band gap between them and increase in tendency for metallization of the glasses. Moreover the glass are have metallization criterion in the range 0.35 to 0.45 posses high nonlinear refractive indices. Hence it is predicted the prepared glass  $85\text{TeO}_2/5\text{Nb}_2\text{O}_5/5\text{ZnO}/5\text{Li}_2\text{O}$  has high nonlinear refractive index  $n_2=8.2 \cdot 10^{-15} \text{ cm}^2 \cdot \text{W}^{-1}$ .



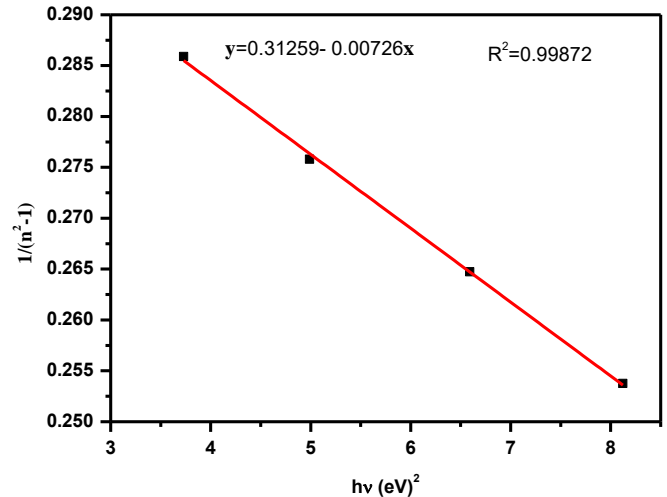
**Fig 5: Relation between electronic polarizability in  $A^03$  and metallization criterion at different wavelength of prepared glass.**

Figure 4 show the refractive index,  $n_0$ , depends on the metallization criterion  $M(n_0)$  by the relation as; [ $M(n_0)=1.07018 - 0.28707 \cdot n_0$ ] and Fig. 5 obtained the relation between the electronic polarizability and  $M(n_0)$  as; [ $M(n_0)=0.996 - 0.08697 \cdot \alpha_m$ ].

The value of,  $n_0$ , is function of different wave length,  $\lambda$  , as follow [16]:

$$\frac{1}{n^2(\lambda) - 1} = \frac{E_s^2 - E^2}{E_s E_d} \quad (8)$$

Where  $E_s$  is denote that Sellmeier gap energy, and  $E_d$  refer to is the dispersion energy. The graph between  $1/(n^2(\lambda)-1)$  and  $E^2 = (h\nu)^2$  for prepared glass  $85\text{TeO}_2/5\text{Nb}_2\text{O}_5/5\text{ZnO}/5\text{Li}_2\text{O}$  studied (see Fig. 4). From the linear relation can determine the value of  $E_s$  and  $E_d$  the present glass has  $E_s=6.43$  eV and  $E_d=20.45$  in eV these results seeing in Table 1. These value are lower than recorded in other tellurite glasses with composition  $85\text{TeO}_2/5\text{Nb}_2\text{O}_5/5\text{ZnO}/5\text{Ag}_2\text{O}$  ( $E_s = 6.76$  eV,  $E_d=20.6$  eV),  $68\text{TeO}_2/5\text{Nb}_2\text{O}_5/20\text{ZnO}/7\text{PbO}$  ( $E_s = 6.61$ eV,  $E_d=20.97$  eV) and  $57\text{TeO}_2/5\text{Nb}_2\text{O}_5/20\text{ZnO}/18\text{PbO}$  ( $E_s = 6.54$  eV,  $E_d=20.65$  eV). Decreasing in the  $E_d$  and  $E_s$  values attributed to the substitution of Li ions as ionic radii atoms in the glass network otherwise a decrease in dispersion energies infers to decrease in the covalent bond



**Fig 6: show the relation between  $1/(n^2 - 1)$  versus  $h\nu$  in  $(\text{eV})^2$  of prepared glass.**

The nonlinear refractive index,  $n_2$ , and the third order nonlinear optical susceptibility,  $\chi^{(3)}$ , can be calculated by Lines formula [16];

$$n_2 = \frac{(n_0^2 - 2)^2 (n_0^2 - 1)}{48\pi N n_0} \left( \frac{E_d}{E_0^2} \right) \quad (9)$$

$$(n_2 / d^2) = \frac{L(f \cdot f_L)^3 (n_0^2 - 1)^2 E_s^2}{n_0 [E_s^2 - E^2]^4} \quad (10)$$

Where  $E_0$  is the average oscillator energy and  $N$  is value of density of polarizable material and,  $\chi^{(3)}$ , in isotropic medium can be calculated from formula as;  $n_2 = \frac{12}{n_0} \text{Re} \chi^{(3)}$ .



Besides, we know that the  $\chi^{(3)}$  strongly depended on the  $n_2$  is defined by Miller's relation as follow  $\chi^{(3)}(n_0) = \chi^{(1)} x 10^{-10}$  esu, where

$$\chi^{(1)} = \left[ \frac{(n_0)^2 - 1}{4\pi} \right]^4. \text{ Here the present glass has}$$

$\chi_{th. Lines}^{(3)} / d^2 = 2.55 \cdot 10^{-13}$  esu/Å<sup>2</sup> and  $\chi_{th. Lines}^{(3)}$  [esu] = 2.04 ( $\chi_{th. Miller}^{(3)} / d^2$ ) [esu/Å<sup>2</sup>]. It can be suggest that these glasses contain Li<sub>2</sub>O can be promising used in optical devices because it has advantage optical properties compared with other tellurite glasses.

#### IV. CONCLUSION

Physical parameters of quaternary glass with composition 85TeO<sub>2</sub>/ 5Nb<sub>2</sub>O<sub>5</sub>/5ZnO/ 5Li<sub>2</sub>O in mol% have been calculated. It is found that the addition of Li<sub>2</sub>O to TeO<sub>2</sub>-based glasses due to increase refractive index and decrease in the density. These glass has advantage optical properties such as;  $\alpha_m = 6.478 \text{Å}^3$  at 435.84nm,  $E_g = 6.43$  eV  $E_d = 20.45$  in eV and  $\chi_{th. Lines}^{(3)} / d^2 = 2.55 \cdot 10^{-13}$  esu/Å<sup>2</sup>. A good correlation has been found between the measured linear refractive index and theoretical electronic polarizability.

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#### AUTHORS PROFILE

**A.E.Al-salami** Received Ph.D degree in experimental solid state from Physics Department, Faculty of Science, Cairo University in 2007 . Currently he is an Associate Prof. Physics Department, Faculty of Science , King Khalid University, Abha. current research includes Structure, Optical and Thermal properties of glasses based on tellurite oxide.