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Physical and optical properties of zinc tellurite Glass embedded Silver Nanoparticles co-doped with Nd^{3+}/Er^{3+} ions

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Abstract

A series of Silver Nanoparticles embedded in the system of zinc-tellurite glass co-doped Neodymium/Erbium ions with the composition of $(69.5\text{-}2.5x)TeO_2–30ZnCl_2–xNd_2O_3–xEr_2O_3–(0.5+0.5x)AgCl$ concentration from 0.0 to 3.0 mol% (x=0, 1, 2 and 3) have successfully been synthesized by melt-quenching techniques. The amorphous nature of the glass was confirmed from x-ray diffraction technique. The decrease of refractive index from 2.446 to 2.429 is attributed to the generation of bridging oxygen atoms via the conversion of TeO_3 into TeO_4 units. Conversely the increase in refractive index to 2.436 is attributed to the generation of non-bridging oxygen atoms via the conversion of TeO_4 into TeO_3 units. From the absorption edge studies, the value of the optical band gap $E^I_{opt.}$ and Urbach energy (ΔE) have been evaluated. The value of $E^I_{opt.}$ lies between 2.34 and 2.83eV for the indirect transition while the Urbach energy values lies between 0.005 to 1.33eV. The experimental results indicate that Nd^{3+}/Er^{3+} rare earth ions co-doped in the system of zinc-tellurite glasses embaded silver nanoparticles are a good candidates for solid state laser as active medium.

Introduction

Tellurite glasses are very important material because of its unique properties that attract much attention in recent years. These glasses join good qualities such as low melting temperatures, good mechanical stability and excellent infra-red transmission [1-3] which lead to the eligible materials for photonic applications and optical devices [4-6]. The high non-linear refractive index of TeO_2 containing glasses is refers to the nonbonding lone electron pair $5s^2$ of tellurium. TeO_2-based glasses have large third-order nonlinear susceptibility and have been considered as favorable materials for use in optical amplifiers and nonlinear optical devices [7].

Dandan *et al.* [8] recently reported that one tellurium element could significantly enhanced the transmission capability, moisture resistance and transparency in the mid-infrared (2.7 μm) band lasers regions for their use as optical materials with suitable dopant rare earth ions. The glasses doped or co-doped with rare-earth ions have generated much interest due to the possibility of several promising applications such as optical data storage, visible laser, fiber amplifier, optical communication and sensor devices [9, 10]. The glass nanocomposites containing metallic NPs as silver nano-particles are among the motivating materials due to their high potency for the applications in various branches of science and technology, e.g.

photo-chemical materials, multi-dimensional and colorful industrial objects, non-linear nanophotonic fabrics, memory devices, and optical switches [11, 12].

However, the high concentration of dopant quenches the behaver of amplifiers and laser. Improving the up-conversion emission and the quantum efficiency is the key issue [13, 14]. Lack studies exist in the literatures on the characteristics of tellurite glass co-doped with Er_2O_3 and Nd_2O_3 which are rare earth elements. Trivalent rare earth (RE) ions doped tellurite glasses are widely investigated due to their unique electronic structure properties highly suitable for potential applications for optical sensing, lasing media, telecommunications and biochemical studies [15-17]. The mechanism behind the concentration quenching is still questionable. In light of the above, it appears obvious that it has to examine the role of co-doping on physical and optical properties of tellurite glasses. In this work, spectroscopic techniques employed to investigate the optical properties of Er^{3+}/Nd^{3+} co-doped tellurite glass system embedded with silver nanoparticles.

EXPERIMENTAL DETAILS

The glass samples were prepared by sing rapid melt quenching technique. The series of four binary glass system (coded as TZNEA0.5, TZNEA1.0, TZNEA1.5 and TZNEA2.0) consist of $(69.5\text{-}2.5x)TeO_2$–$30ZnCl_2$–xNd_2O_3–xEr_2O3–$(0.5+0.5x)AgCl_2$ with x = 0.0 to 3.0 mol %. The composition of the prepared samples and their codes are presented below:

TZNEA0.5	$69.5TeO_2$–$30ZnCl_2$–$0.5AgCl_2$
TZNEA1.0	$67.0TeO_2$–$30ZnCl_2$–$1Nd_2O_3$–$1Er_2O_3$–$1.0AgCl_2$
TZNEA1.5	$64.5TeO_2$–$30ZnCl_2$–$2Nd_2O_3$–$2Er_2O_3$–$1.5AgCl_2$
TZNEA2.0	$62.0TeO_2$–$30ZnCl_2$–$3Nd_2O_3$–$3Er_2O_3$–$2.0AgCl_2$

A homogenized materials batches of 10 g components were placed in a platinum crucible and being melted in a furnace (furnace from Nabertherm GmbH/1600^{o}C-8.0kW-400Volt) at 900^{o}C for 0.5hr. After the required viscosity was achieved the melt were then casted on a metal plate and annealed at 260 ^{o} C for 3 h. Finally, the temperature of the samples cooled down inside the furnace to room temperature for 12hr.

The glass has then been cut at the thickness of about 1.3-2.9 mm. The samples were polished with various types of sand papers, 1500 grid, 1200 grid and 1000 grid to obtain flat and smooth surface. The stoichiometric quantities of the chemicals were weighing out to get the required composition and are grounded in a mortar to obtain a homogeneous fine mixture. The glass structural characterized by using x-ray diffraction (Siemens D5000 X-Ray Diffractometer) with a copper target (Kα=1.54 Å) at a room temperature. The optical absorption of all samples is measured at room temperature by using a Perkin Elmer UV–VIS–NIR spectroscopy (UV-3101PC) in the range of 350–900 nm.

RESULTS AND DESCUSION

To check the amorphous state of the glass samples, X-ray measurements were performed. The short and medium-range orders in the binary system consist of $(69.5\text{-}2.5x)TeO_2$–$30ZnCl_2$–xNd_2O_3–xEr_2O_3–$(0.5+0.5x)AgCl$ glass system structures were tested by means of X-Ray diffraction. The *(Figure: 1)* represents the diffraction patterns of all the samples which shows no sharp Bragg peaks, indicating the amorphous nature of the prepared glasses. A broad humps over the region 26–36° for 2θ value was observed for the glass samples which represents the short range order of the high concentration of TeO_2 and $ZnCl_2$ composition corresponding to the diffraction effects due to the amorphous portion of the samples, which is the characteristic of the glass structure [7]. While, the low broad hump over the region 48°–54° for 2θ represents the low concentration of Er_2O_3 and Nd_2O_3 and Ag NPs in its amorphous nature in the samples.

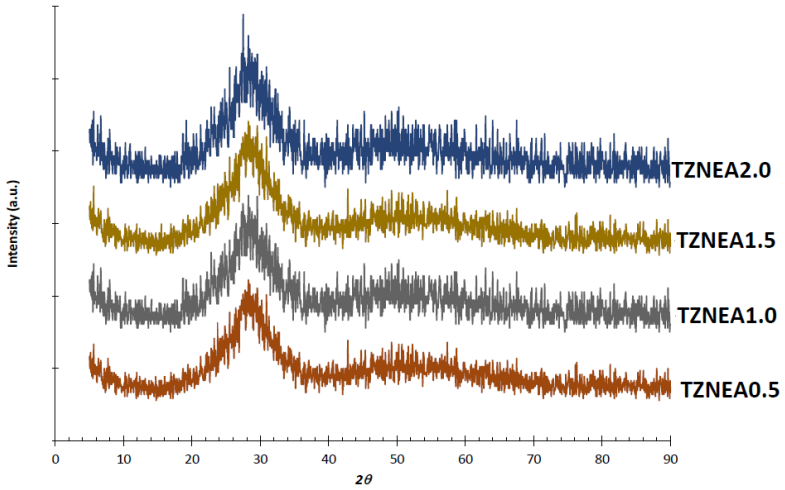

Figure-1: The diffractogram of XRD patterns of (69.5-2.5x)TeO_2–30$ZnCl_2$–xNd_2O_3–

The density (ρ) of each sample is measured by Archimedes method using toluene as the immersion liquid with an estimated error of ±0.003. The density of each sample is determined by the relation:

$$\rho = \rho_T \frac{W_{air}}{W_{air} - W_T} \ (gm/cm^3) \tag{1}$$

where ρ_T is the density of toluene (0.8669 gm/cm^3), W_{air} is the sample weights in air and W_T is the sample weights in toluene liquid [18].

The Molar volume (V_M) is given by:

$$V_M = \frac{M}{\rho} \ (cm^3.mol) \tag{2}$$

Where M is the average molecular weight of the glass and ρ is the density of glass samples. [19].

The nominal compositions of the prepared glasses and their corresponding codes together with some of their physical properties as density, refraction index and molar volume are listed in Table 1.

Table-1: Nominal composition of (69.5-2.5x)TeO_2–30$ZnCl_2$–xNd_2O_3–xEr_2O_3–(0.5+0.5x)AgCl glass system with codes, density (g/cm^3) and molar volume (V_m).

Glass	***mol fraction (mol%)***					***ρ***	***n***	***Vm***
Code	***TeO_2***	***$ZnCl_2$***	***Nd_2O_3***	***Er_2O_3***	***AgCl***	***(gm/cm^3)***		***(cm^3/mol)***
TZNEA0.5	*69.5*	*30*	*0*	*0*	*0.5*	*5.533*	*2.446*	*29.58*
TZNEA1.0	*67.0*	*30*	*1*	*1*	*1.0*	*5.586*	*2.432*	*31.09*
TZNEA1.5	*65.0*	*30*	*2*	*2*	*1.5*	*5.613*	*2.429*	*34.53*
TZNEA2.0	*62.0*	*30*	*3*	*3*	*2.0*	*5.716*	*2.436*	*40.61*

The *(Figure: 2)* represents the relationship between density, refraction index and molar volume as a function of Nd^{3+}/Er^{3+} contain.

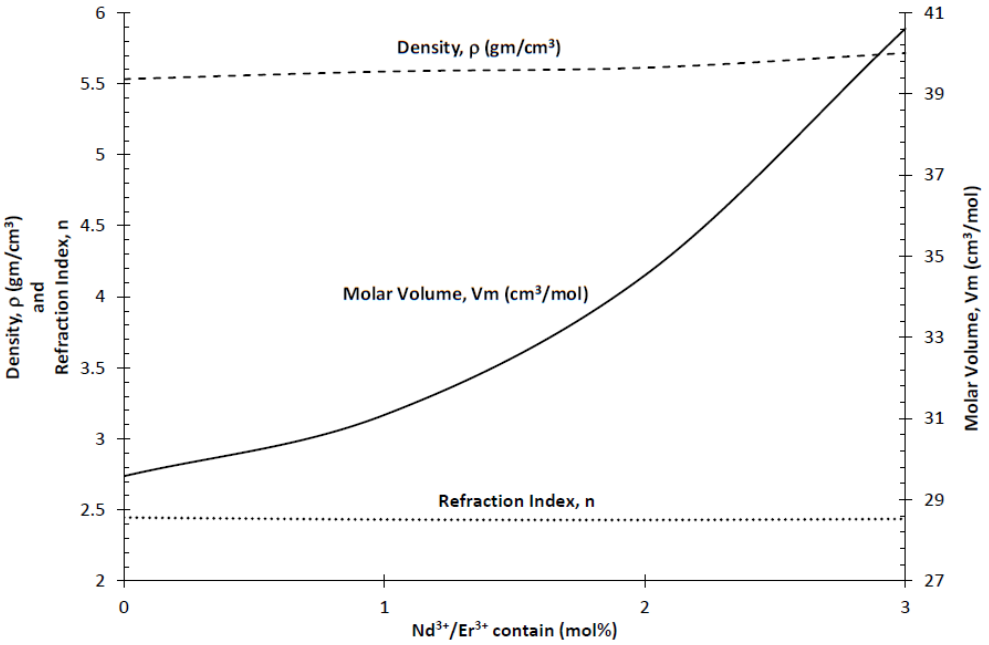

Figure-2: Density, molar volume and refraction index of Ag NPs embaded tellurite glass.

The densities of the glass system were found to increase from 5.533 to 5.716 gm/cm^3 and the molar volume V_m is increases as Nd^{3+}/Er^{3+} ions increases from 0 to 3 mol% as shown in Figure. 2. The increase in 'ρ' could increase the compactness of the glass network and as a result the overall bond lengths decrease. As rare earth is added to the Tellurite networks, converting bridging oxygens to non-bridging oxygens. This results in breaking of continuous networks of tellurium and the non-directed bonding of cations collapses the network into closely packed structure, changing the density of the prepared glasses. The increase in 'ρ' for the samples with 2% mol of rare earth ions may also be due to the higher density of the rare earths (Nd_2O_3=7.24 gm/cm^3, Er_2O_3=8.64 gm/cm^3) than that of TeO_2 (2.46 g/ gm/cm^3) but the density of AgCl (5.56 gm/cm^3) is almost similar to the density of TeO_2 (5.67 g/ gm/cm^3) this is why the varies of 'ρ' with adding more rare earth ions is more affective ($\Delta\rho\approx$0.061gm/cm^3 as average) than adding the NPs.

According to the literature [20], The absorption coefficient was calculated by:

$$\alpha(\omega) = 2.303\frac{A}{d}(cm^{-1}) \tag{3}$$

where A is a constant and d is the thickness of the glass.

The optical absorption at the fundamental edge in terms of the theory given by [21] and it can be written as [22, 23]:

$$\alpha(\omega) = A.\frac{(\hbar\omega - E_{opt.})^r}{\hbar\omega}(cm^{-1}) \tag{4}$$

Where $\hbar\omega$ is the incident photon energy, $E_{opt.}$ is the optical band gap and r is an index which can take values of 1, 2, 3, 1/2, and 3/2 depending on the nature of the interband electronic transition. The values of r is 1/2 (for allowed direct transitions), 3/2 (for direct forbidden transitions), 2 (for allowed indirect transitions) and 3 (for forbidden indirect transitions). From (4). The optical gap estimates by extrapolating the linear region of the curves to $(\alpha\hbar\omega)^{1/2}$=0 for all four samples as shown in (Figure: 3) [24]. These band gaps obtained from the above relations are interband transition, but later encompass the photon interaction [18].

The refractive index n of each sample (Table 1) is calculated according to the following equation [25]:

$$\frac{n^2-1}{n^2+1} = 1 + \sqrt{\frac{E^D_{opt.}}{20}} \quad (5)$$

where $E^D_{opt.}$ is the direct optical band gap energy of the glass obtained from absorption data.

The degree of disorder in amorphous and crystalline material is described by Urbach energy (ΔE) and is calculated using the relation,

$$\alpha(\omega) = B\frac{\hbar\omega}{\Delta E} \quad (6)$$

where B is a constant.

The measured and calculated values of the indirect optical band gap energy (E^I_{opt}) and the Urbach energy (ΔE) for all samples of tellurite glasses are summarized and listed in Table 2.

Table-2: The optical parameters o (69.5-2.5x)TeO_2–30$ZnCl_2$–xNd_2O_3–xEr_2O_3–(0.5+0.5x)AgCl glass system.

Glass code	*Optical band gap, $E^I_{opt.}$ (eV)*	*Urbach energy, ΔE (eV)*
TZNEA0.5	2.340	0.005
TZNEA1.0	2.810	1.330
TZNEA1.5	2.830	1.330
TZNEA2.0	2.600	0.970

Optical band gap studies in the UV region have proven to be an effective tool for investigating the electronic band structure of amorphous materials [26]. The principle based on the absorption of a photon with energy greater than the band gap energy. Direct and indirect optical transition can occur at the fundamental absorption edge. Both of them involve the interaction of an electromagnetic wave with an electron in the valence band. The (Figure: 3) shows the typical absorption spectrum of all tellurite glass samples.

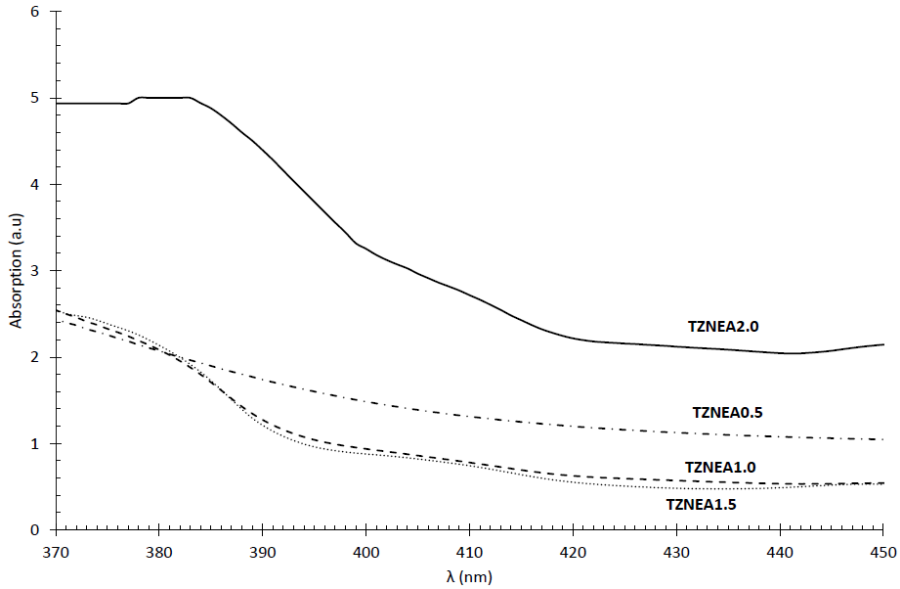

Figure-3: Variation of absorption coefficient with wavelength of (69.5-2.5x)TeO_2–30$ZnCl_2$–xNd_2O_3–xEr_2O_3–(0.5+0.5x)AgCl glasses.

The variation of the indirect optical energy band gap ($E^I_{opt.}$) versus photon energy (E) is shown in (Figure: 4), and the values of ($E^I_{opt.}$) is recorded in Table 2.

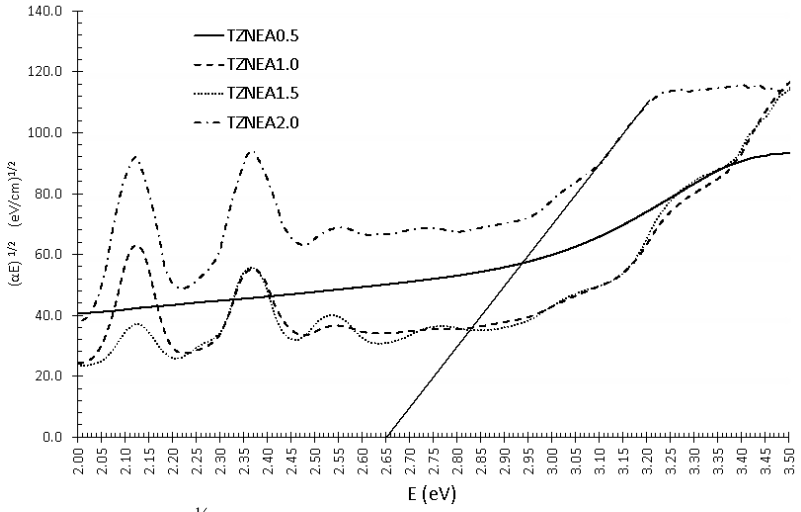

Figure-4: A plot of $(\alpha\hbar\omega)^{½}$ versus photon energy (E) for of $(69.5\text{-}2.5x)TeO_2–30ZnCl_2–xNd_2O_3–xEr_2O_3–(0.5+0.5x)AgCl$ glasses.

The values of Urbach energy (ΔE) are calculated from slopes of the linear regions of the curves at lower photon energies and taking their reciprocals corresponding to the expression given by [27, 28]:

$$\ln \alpha(\omega) = \frac{\hbar\omega}{\Delta E} - C \tag{7}$$

where C is a constant.

The *(Figure: 5)* shows the variation of $ln(\alpha)$ with the photon energy (E) and the values of its Urbach energy (ΔE) have been recorded in Table 2

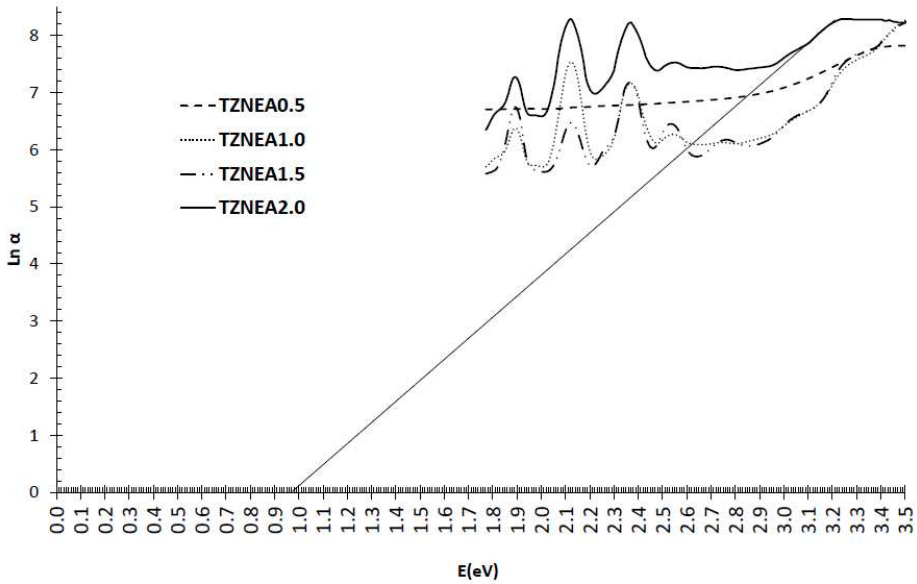

Figure-5: A plot of $ln(\alpha)$ versus photon energy ($\hbar\omega$) of $(69.5\text{-}2.5x)TeO_2–30ZnCl_2–xNd_2O_3–xEr_2O_3–(0.5+0.5x)AgCl$ glasses.

The (Figure: 6) introducing the variation of the optical bang gap energy ($E^I_{opt.}$) and Urbach energy (ΔE) with the concentration of RE contains.

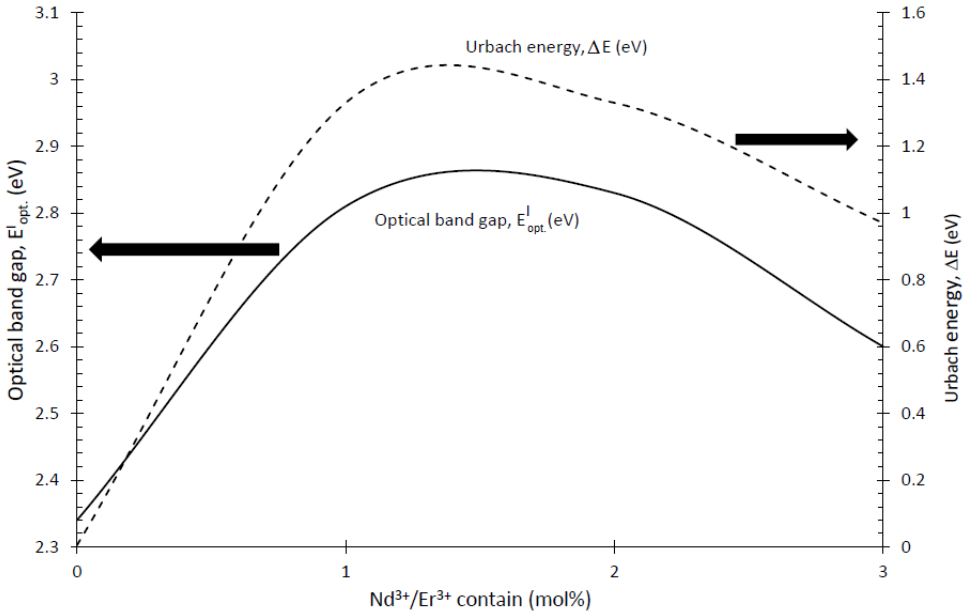

Figure-6: A plot of optical band gap energy ($E^{I}_{opt.}$) and Urbach Energy (ΔE) against Nd^{3+}/Er^{3+} mol% content for of $(69.5\text{-}2.5x)TeO_2–30ZnCl_2–xNd_2O_3–xEr_2O_3–(0.5+0.5x)AgCl$ glasses.

The values of optical bang gap energy ($E^{I}_{opt.}$) are between 2.340eV and 2.830eV while the values of Urbach energy (ΔE) is between 0.005eV and 1.330eV (Table 2). The plot of $E^{I}_{opt.}$ and ΔE against Nd^{3+}/Er^{3+} mol% content presented to examine the clear nature of the optical gap energy and Urbach energy for or tellurite glass system (Figure: 6). It can be seen from the figure that the optical bang gap energy ($E^{I}_{opt.}$) and Urbach energy (ΔE) have a maximum values for the concentration of the sample TZNEA1.5 (2.830eV and 1.330eV) respectively. Meanwhile, the much lower value of the optical bang gap energy ($E^{I}_{opt.}$) and Urbach energy (ΔE) observed for the TZNEA0.5 and TZNEA2.0 presented glasses which suggests that the defects in these glasses are minimum [29].

The variation in band gap value is attributed to the structural changes that are taking place in the title glasses while co-doping RE content. From (Figure: 6), it is clearly shows the both of optical band gap energy ($E^{I}_{opt.}$) and Urbach energy (ΔE) are higher in the Nd^{3+}/Er^{3+} content of 1 and 2 mol% (where NPs of silver contain is 1 and 1.5 mol% respectively). The co-ordination number of Te atoms changes when the RE co-doping is introduced into the host matrix. Generally, more number of TeO_3 units has formed from TeO_4 to TeO_{3+1} and consequently band gap energy increases for concentration of 1 and 2 mol% [26].

The results are in good agreement with previous report in which the addition of rare-earth to the oxide glass shows a reduction in optical band gaps as well as Urbach energies with the densification of the glass network [29]. The estimated optical energy gap, $E^{I}_{opt.}$ at the fundamental absorption edge is higher in the co-doped glasses as compare to the host glass. This condition has also proposed related to the creation of higher number of non-bridging oxygen (NBO)s units after the incorporation of both Er^{3+}/Nd^{3+} ions into the host matrix. This attributed to the generation of large number of NBO which are more covalent in character than the bridging oxygen [18]. Consequently, transfer of electrons from the valence band to the conduction band becomes easier. The obtained optical energy gap values of the studied glasses are in a good agreement with the range of the values in various tellurite glasses reported by [20, 30, 31].

CONCLUSION AND RECOMMENDATIONS

The role of co-doping on optical of tellurite glasses with composition $(69.5\text{-}2.5x)TeO_2–30ZnCl_2–xNd_2O_3–xEr_2O_3–(0.5+0.5x)AgCl$ (x = 0.0 to 3.0 mol%) has successfully been synthesized by using conventional method. The amorphous nature of the prepared samples is confirmed by XRD. Density is an effective tool to explore the degree of structural compactness, modification of the geometrical configurations of the glass

network, change in coordination and the variation of dimensions of the interstitial holes. The Optical absorption behavior is measured using UV-VIS-NIR spectroscopy. It was observed that the maximum value of both of the optical band gap ($E^{I}_{opt.}$=2.830eV) and Urbach energy (ΔE=1.330eV) is at the concentration of Nd^{3+}/Er^{3+} at 1.5% contents for the third sample coded with TZNEA1.5. The results approved that the second glass sample has formed more number of TeO_3 units in the glass matrix. Both the optical band gap and the Urbach energy are found to be strong analytical functions of the co-dopant concentration. The results are in good agreement with other researchers work. The glass forming mechanism is understood. It is interesting to investigate the photoluminescence behavior and the structure properties by using Differential thermal analysis (DTA), Fourier transform infrared spectroscopy (FTIR) and Raman spectroscopy in these glass systems as a function of co-dopant concentration.

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