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# **Nonlinear Optical Properties of Tellurite-based Glasses**

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**Abstract**: We have investigated the mechanisms which are contribute to the linear and nonlinear optical behaviors of the heavy metal containing tellurite glasses by using UV-Vis spectrophotometer and open aperture Z-scan experiments. The bandgap values of the studied tellurite glasses with WO<sub>3</sub>, Nb<sub>2</sub>O<sub>5</sub>, and ZnO modifiers were found among 2.58 eV and 3.01 eV. The results of the open aperture Z-scan experiments indicated that the studied tellurite glasses modified with different amounts of WO<sub>3</sub>, Nb<sub>2</sub>O<sub>5</sub>, and ZnO possessed larger nonlinear absorption coefficients (among 2.62x10<sup>-7</sup> cm/W and  $6.38x10^{-7}$  cm/W) than the nonlinear absorption coefficients of previously investigated tellurite glasses. When the linear and nonlinear absorption results of the studied modified tellurite glasses considered together, it can be reported that the mechanisms which are contributed to the nonlinear absorption coefficients caused mainly from two photon absorption and two photon assisted free carrier absorption.

Keywords: Tellurite glasses, Z-scan experiments, Nonlinear absorption

# Introduction

Because of their wide application area such as: amplifiers, modulators, lasers, sensors, optical limiters, the tellurite glasses still attracting interest (de Araujo et al., 2017). In addition, thermal, mechanical, chemical, and optical properties save several advantages to the tellurite glasses compared to the other glasses (Bilir et al., 2011, Bilir et al., 2011). The new trend to evolve suitable materials for desired application area is achieving by the combination and/or hosting of the some materials. The disadvantages to improve the optical properties of the some materials can be described as: (i) difficulties of the growth of the single crystals, (ii) prone to recrystallization of the inorganic materials and (iii) poor stability of the organic crystal phases at elevated temperatures (Yankov et al., 2012). It is possible to overcome these disadvantages by glassy materials. The tellurite glasses are very good candidates for hosting up to certain amounts of heavy metals and nanoparticles, due to their superior mechanical and chemical properties. On the other hand, the high polarizability is one of the unique property of the tellurite glasses (Tasheva et al., 2017). As well known that the polarization is the basic parameter of the nonlinear response of the materials, and under light of this truth, the tellurite based glasses should exhibit high third order nonlinear optical susceptibility (Yamane et al., 2000, Manning et al., 2012, Komatsu 2015).

In the literature, from the third order nonlinearity, generally the nonlinear refraction properties of the tellurite glasses had been studied (Tasheva et al., 2017, Souza et al., 2006, Kaur et al., 2017, Kim et al., 1993, Kim et al 1995, Castro-Beltran et al., 2011). In this study, we have investigated the linear and nonlinear optical characteristics of the WO<sub>3</sub>, Nb<sub>2</sub>O<sub>5</sub>, and ZnO doped tellurite glasses with UV-Vis spectrophotometer and open

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aperture Z-scan experiments. As the linear and nonlinear optical results of the studied glasses are considered together, it can be estimated that the nonlinear optical absorption causes from two photon absorption and two photon assisted free carrier absorption mechanisms. In the presence of TPA, the intensity dependent absorption is described by Eq. (1)

$$\alpha(I) = \alpha_0 + \beta I \tag{1}$$

where  $\alpha_0$  is the linear absorption coefficient and  $\beta$  is the nonlinear absorption coefficient. Experimental normalized transmittance data was fitted to Eq. (2) (Bahae et al., 1990)

$$T(z, S=1) = \frac{1}{\sqrt{\pi}q_0(z,0)} \int_{-\infty}^{\infty} \ln\left[1 + q_0(z,0)e^{-\tau^2}\right] d\tau$$
(2)

where  $q_0(z,0) = \beta I_0 L_{eff} / (1 + z^2 / z_0^2)$ , z is the position of the sample with respect to the focal position,  $z_0 = k\omega_0^2/2$  is the Rayleigh range,  $\omega_0$  is the beam radius at focus,  $I_0$  is the intensity of the incident light beam at the focus (z = 0),  $L_{eff} = [1 - \exp(-\alpha_0 L)]/\alpha_0$  is the effective thickness of the material and L is the sample length.

### Method

5 glass systems with the compositions of  $(1-x)TeO_2 + xZnO$  (x=0.15, 0.25, 0.35), 0.85TeO<sub>2</sub> + 0.15WO<sub>3</sub>, and 0.85TeO<sub>2</sub> + 0.15Nb<sub>2</sub>O<sub>5</sub> were synthesized using the conventional melt quenching technique. The oxide powders of each reagents with purities not less than 99.99% were used for the synthesis of glass samples. All the oxide powders with appropriate molar amounts were mixed thoroughly using an agate mortar. The batches of 7g of powders were prepared and were then melted in a platinum crucible in an electric muffle furnace under ambient conditions at temperatures ranges from 800 to 1100C according to the type and the amount of the modifier. The powders were kept at under these temperatures for an hour. Then, the glass melts were poured onto a stainless steel mold. The obtained glasses were thermally treated under their glass transition temperature (250<sup>o</sup>C) for 2 hours to remove thermal stresses due to the high synthesis temperatures.

The linear absorption spectra of the glass samples were recorded using a Shimadzu-1800 model UV-Vis spectrophotometer under room temperature conditions. The nonlinear absorption characteristics were investigated by open aperture Z-scan experiments., A laser source with 532 nm wavelength, 10 Hz repetition rate and 65 ps pulse duration was used in the open aperture Z-scan experiments. In addition, a 30 cm focal length lens was used to enlarge the Rayleigh range.

#### **Results and Discussion**

The linear absorption spectra of the studied glasses which are recorded by UV-Vis spectrophotometer under room temperature conditions are given in Figure 1. As seen from the figure, the absorption edges are changing related to the mole percentage and/or type of network modifiers (WO<sub>3</sub>, Nb<sub>2</sub>O<sub>5</sub>, and ZnO).

The differences among the wavelengths at absorption edges can be attributed to the ratio of the non-binding oxygen atoms. The band gaps of the glasses were estimated by the linear extrapolation of the absorption edges to the wavelength axis. The calculated band gaps of the glasses are given in Table 1. It is clearly seen from the table that the increasing of the mole ratio of the ZnO in the tellurite glass causes to the narrowing at the band gap. This result can be attributed to the formation of the dopant states by the ZnO dopant at the bottom of the conduction band of the glass. On the other hand, the increasing of the ratio of the non-bridging oxygen atoms are causing to decrease at the band gap of the modified tellurite glasses.

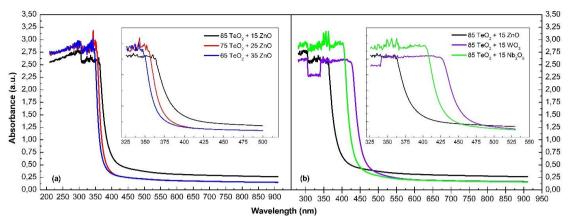


Figure 1. Linear absorption spectra of the studied glasses

The nonlinear absorption traces obtained from the open aperture Z-scan experiments of the studied glasses were shown in Figure 2. The nonlinear absorption coefficients of the modified tellurite glasses were obtained by fitting of the open aperture Z-scan results. The found nonlinear absorption coefficients ( $\beta_{eff}$ ) were given in Table 1. As we mentioned above the two photon absorption and two photon assisted free carrier absorption mechanisms contributed to the nonlinear absorption coefficients.

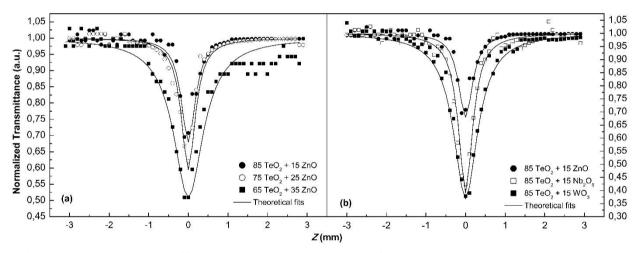


Figure 2. Comparison of the effective nonlinear absorption traces of samples according to (a) amount, and (b) type of the modifiers at 532 nm for 65 ps pulse duration

The narrowing of the band gap of the glasses lead to the increasing of the nonlinear absorption coefficient (Table 1).

Table 1. Comparison of the nonlinear absorption coefficients of the tellurite glasses related to the amount and/or type of the doped network modifiers

Material	Thickness (cm)	$\alpha_0(cm^{-1})$	$\omega_0(\mu m)$	$\beta_{eff}(cm/W)$	Band gap (eV)
85 mole% TeO <sub>2</sub> +15 mole% ZnO		0.688		$2.62 \times 10^{-7}$	3.07
75 mole% TeO <sub>2</sub> +25 mole% ZnO		0.402		$3.08 \times 10^{-7}$	3.24
65 mole% TeO <sub>2</sub> +35 mole% ZnO	0.5	0.338	54	$3.67 \times 10^{-7}$	3.32
85 mole% TeO <sub>2</sub> +15 mole% Nb <sub>2</sub> O <sub>5</sub>		0.352		$6.24 \times 10^{-7}$	2.85
85 mole% TeO <sub>2</sub> +15 mole% WO <sub>3</sub>		0.364		6.38x10 <sup>-7</sup>	2.73

#### Conclusion

The linear and nonlinear absorption properties of binary tellurite glasses have been investigated using absorption spectrophotometer and open aperture Z-Scan experiments, respectively. The band gap of the glass samples were determined using linear absorption spectra and found to be blue shifted at about 32 nm with the increasing amount of ZnO and red shifted at about 70 nm with the change of the type of the modifier. The linear optical absorption measurements showed the tunability of the band gaps of the glass samples, which has great significance for photonics applications, by changing the type and the amount of the modifiers. As for the nonlinear optical measurements, it was found that the two-photon absorption signals and the response times are relatively fast which is a desired feature for optical switching and photodetector applications.

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