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Theoretical analysis of ultrasonic relaxation in advanced tellurite glasses

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Abstract

This work deals with the theoretical analysis of ultrasonic relaxation in the glass system xNb_2O_5 - (100- x) TeO_2 glasses, and $4 \leq x \leq 22.5$ mole %. The parameters of the theoretical analysis deal with the transmission of ultrasonic waves through dual-well potential. The parameters are alternate potential energy, centers of energy loss, and elongation or contraction of the two-well potential. Moreover, the deformation potential is found to be sensitive to the variations of the Nb_2O_5 content. The number of centers of energy loss is related to the elastic moduli as a function of Nb_2O_5 content. The analysis revealed a sensitive effect of variation of Nb_2O_5 contents for elongation or contraction of the dual-well potential. Due to the increase of Nb_2O_5 from 4 to 22.5 mole %, the potential energy changed from 4-92 to 5.06 eV, longitudinal and transverse deformation potential changed from 0.18 to 0.346 and from 0.06 to 0.141 eV, respectively. The increase in the deformation potential of the presented dual-well state could be attributed to the increase of the experimental elastic parameters like Young's and shear moduli of the present glasses.

Keywords

Glasses, Tellurite, Ultrasonic relaxation.

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1. Introduction

Tellurite glasses are the topic of wide investigation due to their interesting physical properties which lead to sophisticated applications and are considered these glasses multifunctional glasses or smart glasses [1–17]. Also, transparent tellurite glass-ceramics for photonics applications and application to the case of thallium tellurite glasses [8, 9]. Moreover, novel 3.1μm and enhanced 2.7 μm emissions in Er^{3+} doped fluorotellurite glasses ceramic and copper nanoparticles–containing tellurite glasses: an efficient surface-enhanced Raman scattering substrate have been reported [10, 11]. Recently, visible to the infrared down conversion of Er^{3+} doped tellurite glass for luminescent solar converters, float, borosilicate, and tellurites as cover glasses in Si photovoltaics, performances under sunlight, and plasmonic effect of bimetallic TiO_2/Al_2O_3 nanoparticles in tellurite glass for surface-enhanced Raman scattering applications have been reported [12–14]. Besides that, a recent applications like white light source and optical thermometry based on zinc-tellurite glass tri-doped with Tm^{3+} / Er^{3+} /Sm^{3+}, functional thermal stable samples of MoO_3–V_2O_5-TeO_2 glasses, and radiation attenuation factors of borotellurite glasses have been achieved [15–17].

The transmission of the ultrasonic waves over a glass structure experiences attenuation. This attenuation is caused due to the energy dissipation of the ultrasonic wave into the glass structure. So, the determination of attenuation coefficient (α) over their transmission in a glass structure at lower temperatures (T) presented a pronounced peak. The ultrasonic attenuation coefficient ($\alpha - T$) relation requires extensive distributions of relaxation times and low energy excitations [18–22]. Such a relation can be related to the oscillations of oxygen atoms in unequal dual-well potentials [23–27]. The energy from the ultrasonic waves enforced oxygen atoms to exceed the obstacle height (activation energy (E_h)). This process can be achieved upon the raise of temperature (T). The needed time to overcome this height is named relaxation time (τ). The relaxation time can be related to both temperature and wall height E_h. The increase of the ultrasonic frequency (f) formed additional oscillating oxygen atoms in the dual-well at higher temperatures. Such a procedure is called thermal relaxation.

The purpose of the present study is to throw more light on the analysis of the ultrasonic relaxation at low temperatures of xNb_2O_5 - (100- x) TeO_2 glasses, $4 \leq x \leq 22.5$ mole % [18] through the usage of the central force model [24–27].

Table 1. Dependence of Young's modulus (Y) [18], shear modulus (S) [18], molar volume (V_m) [18], theoretically computed mutual potential energy (U_0), area under the curve $\int C(E)dE$ of $\alpha - T$ relationship, number of loss centers n, the longitudinal elongation (e$_L$%), the transverse elongation % (e$_S$%), theoretically determined normal cation–anion separation δx (longitudinal and transverse), deformation potential (D_{th}) (longitudinal and transverse) on NbO content of the glass system xNb$_2$O$_5$ - (100-x) TeO$_2$ glasses, 4 $\leq$x$\leq$ 22.5.

Content mol% TeO$_2$	Content mol% Nb$_2$O$_5$	Y - GPa	S - GPa	V_m x10^{-6} m^3/mol	U_0 - eV	$\int C(E)dE$ - -	nx 10^{20} cm^{-3}	e$_L$ % -	e$_s$ % -	δ_{xth} long. nm	δ_{xth} trans. nm	D_{th} long. eV	D_{th} trans. eV
96	4	20.1	6.98	0.029	4.92	0.0411	6.31	40.03	-7.85	0.08	0.08	0.0181	0.063
92.5	7.5	22.27	7.76	0.030	4.95	0.0425	6.82	40.49	-7.81	0.08	0.08	0.201	0.070
90	10	24.53	8.58	0.031	4.96	0.0444	7.30	40.55	-7.77	0.08	0.08	0.225	0.079
87.5	12.5	26.98	9.47	0.314	4.98	0.0445	7.87	41.22	-7.70	0.08	0.08	0.243	0.085
85	15	29.77	10.48	0.324	5.00	0.0446	8.08	41.25	-7.62	0.08	0.07	0.267	0.108
82.5	17.5	33.71	11.95	0.332	5.02	0.0451	8.45	42.06	-7.57	0.08	0.07	0.302	0.122
80	20	38.04	13.59	0.341	5.04	0.0459	8.92	42.43	-7.51	0.08	0.07	0.340	0.139
77.5	22.5	39.18	14.00	0.347	5.06	0.0464	9.39	42.79	-7.47	0.08	0.07	0.346	0.141

2. Theoretical analysis and discussions

The applicability of the central force model [24–27] required adopting values of some physical parameters. The central force model [24–27] suggested oscillations of the light atoms in the glass structure in asymmetric two-well potential. These atoms are considered oxygen atoms. These atoms aim to surpass the wall elevation (activation energy). This aim is a result of the effect of ultrasonic energy on these atoms. The ultrasonic energy increases as the frequency (f) of these waves increase with the increment of the temperature. The increment of the ultrasonic energy formed extra oscillating oxygen atoms in a potential well. Therefore, the transmission of ultrasonic waves into the amorphous structure changes the equilibrium of the vibrating oxygen atom around the bottom of the well. This procedure created an energy change (ΔE) between the minima of the well. This energy shift can be presented by the deformation potential (D). The last parameter means the energy motion of the relaxing states in a strain field, so, it presents the interaction between ultrasonic waves within the asymmetric dual-well potential. In the central force model [24–27], it has been suggested an equation represents the internal friction ($Q-1$) of a set of oxygen atoms/volume (n) proceed in comparable dual-well potential with a wall

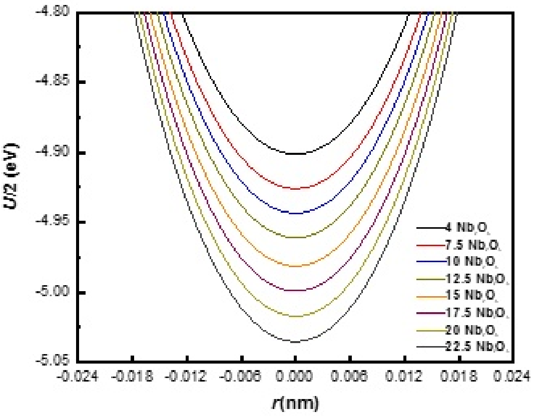

Figure 1. Variations of the configuration of the dual-wells for longitudinal vibrations of oxygen atoms at e= 0% for the glass system xNb$_2$O$_5$ – (100- x) TeO−2 glasses, 4 $\leq$x$\leq$ 22.5 .

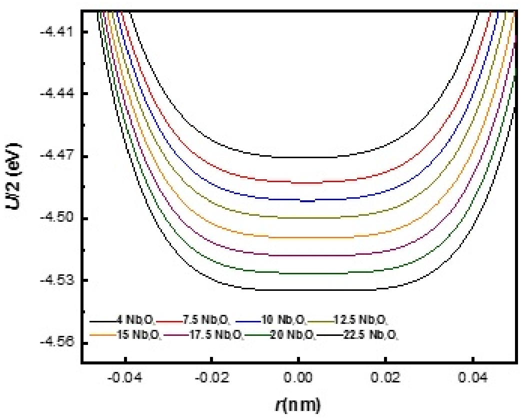

Figure 2. Variations of the configuration of the dual-wells for longitudinal vibrations of oxygen atoms at e=20% for the glass system xNb$_2$O$_5$ – (100- x) TeO−2 glasses, 4 $\leq$x$\leq$ 22.5 .

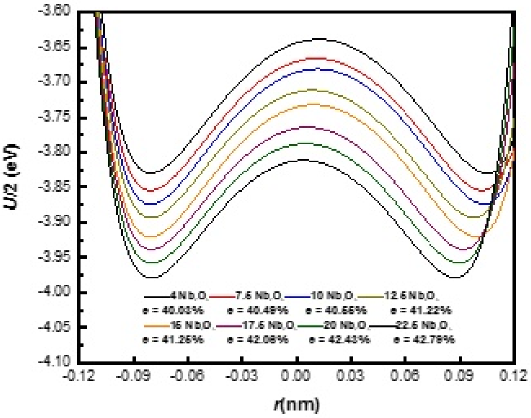

Figure 3. Variations of the configuration of the dual-wells for longitudinal vibrations of oxygen atoms at different equilibrium interatomic distances for the glass system xNb_2O_5 – (100- x) TeO−2 glasses, 4 ≤x≤ 22.5 .

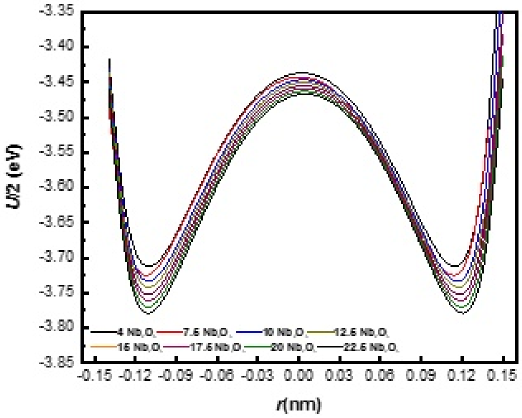

Figure 4. Variations of the configuration of the dual-wells for longitudinal vibrations of oxygen atoms at e= 60% for the glass system xNb_2O_5 – (100- x) TeO−2 glasses, 4 ≤x≤ 22.5 .

height E_h as;

$$Q^{-1} = \frac{nD^2\omega\tau}{4\rho v^2 kT(1+\omega^2\tau^2)} \tag{1}$$

when $\omega\tau(E) = 1$, the ultrasonic attenuation is maximum, and in this case, there is a distribution of both the activation energies and the relaxation times, so, Eq. 1 will be modified to

$$Q^{-1} = \frac{2nD^2}{\rho v^2}\int_0^\infty \frac{\omega\tau n(E)dE}{1+\omega^2\tau^2} \tag{2}$$

The oxygen atoms per unit volume (n) or the centers of energy loss can be taken as a summation of the probable activation energies as;

$$n = \int_0^\infty n(E)dE \tag{3}$$

The parameter $n(E)$ can be described in terms of the ultrasonic velocity (v) the angular frequency (ω) and the density (ρ) [20] as;

$$n(E) = E_h^{-1} e^{\frac{-E}{E_h}} = \frac{\rho v^2}{2zzD^2}\int_0^\infty C(E)dE \tag{4}$$

where zz is a constant and the $\int_0^\infty C(E)dE$ is the total number of centers of energy loss. The last parameter can be deduced from $\alpha - T$ relation.

The quantitative exploration of the central-force model [24–27] revealed that;

1. There is a predictable oscillation of oxygen atoms well with atomic limits,
2. Such a well had single minima corresponds to the equilibrium of oscillated oxygen atoms,
3. These minima agree with small O oscillations. The minima will flatted at higher anharmonic oscillations,
4. The energy of the transmitted ultrasonic wave (longitudinal or shear) disfigured the well,
5. The well had a potential energy U and an elongation e based on the longitudinal or shear oscillations of oxygen atoms as follows

$$U_L = -aa_1\left(\frac{1}{r}+\frac{1}{2el_0-r}\right)+aa_2\left(\frac{1}{r^m}+\frac{1}{(2el_0-r)^m}\right)$$

$$e = \frac{L}{2l_0}, b = \frac{al_0^{m-1}}{m}$$

$$U_T = \frac{-2a}{(e^2l_0^2+d^2)^{1/2}}+\frac{2b}{(e^2l_0^2+d^2)^{m/2}} \tag{5}$$

where aa_1 and aa_2 are constants, m takes values from 6-12, L is the cation-cation distance and l_0 expressed bond length in a definite glass structure. The term $U/2$ is a suitable expression for the potential energy U and in this case, characterized the alternate potential energy anion and cation (regarded as a more heavy atom).

The parameters are v, ρ, $\int C(E)dE$, $n(E)$, and T for each curve according to $\alpha - T$ relationship. The values of these parameters are based on that mentioned in Refs [17, 18]. The derived parameters from this model as a function of Nb_2O_5 are listed in Table 1. Based on the values of the mentioned parameters in Table 1, there are some remarkable features;

- The values of $\int C(E)dE$ are in inverse proportions with values of the activation energies [17],
- The increased content of Nb_2O_5 will increase the number of centers of energy loss. This increase can be related to the increment of $[NbO_6]$ at the expense of $[TeO_4]$ and $[TeO_3]$

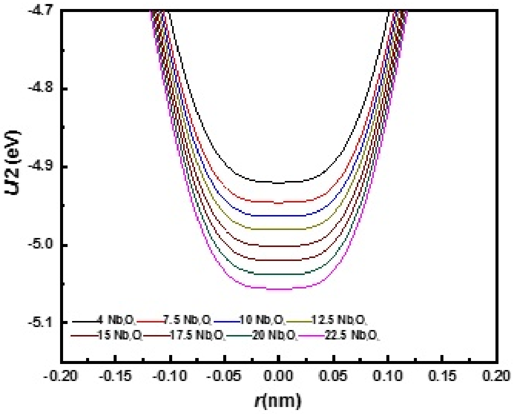

Figure 5. Variations of the configuration of the dual-wells for transverse vibrations of oxygen atoms at e= 0% for the glass system xNb_2O_5 – (100- x) TeO−2 glasses, 4 ≤x≤ 22.5 .

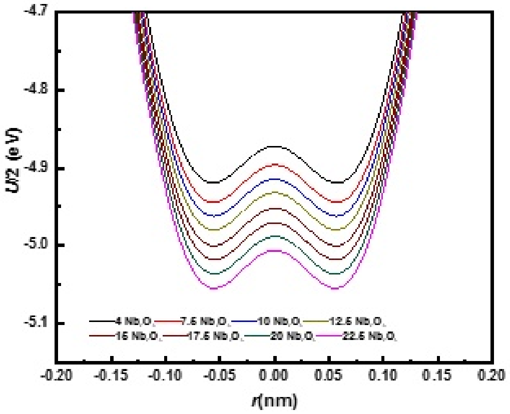

Figure 7. Variations of the configuration of the dual-wells for transverse vibrations of oxygen atoms at different equilibrium interatomic distances for the glass system xNb_2O_5 – (100- x) TeO−2 glasses, 4 ≤x≤ 22.5 .

structural units, in other words, the effect of the increment of the coordination number.

The deuced parameters from the applicability of the model are depicted in Figs (1-8) for longitudinal and shear oscillations. Based on these plots:

1. The oxygen atoms vibrate in a symmetric potential well at zero elongations and the well has one minimum as depicted in Figs. 1-2. Also, Fig. 1 presents the longitudinal oscillations while Fig. 2 represents the shear oscillations,

2. The decrement values of the potential energy as a function of Nb_2O_5 at zero elongation can be related to the relaxation of the tellurite network (effect of increasing molar volume as collected in Table 1. Such a relaxing network is created from the effect of increasing both the coordination number of Nb_2O_5 and the rigidity [17],

3. As the ultrasonic energy transfer to the oxygen atoms increases, the elongation will increase to a ratio of 20% for longitudinal oscillations and -2% for shear oscillations as shown in Figs. 3-4. This means enlargement of the anharmonic oscillations of oxygen atoms. Thus, two-well potential tends to be flat,

4. More ultrasonic energy transfers to the oxygen atoms enforced the elongation of the oscillations to increase. As the elongation exceeds 20% for longitudinal oscillations or -2% for shear oscillations, the flat potential well will have dual states, i.e., the dual well potential will be created. At some elongations ratios as listed in Table 1, the height of the dual

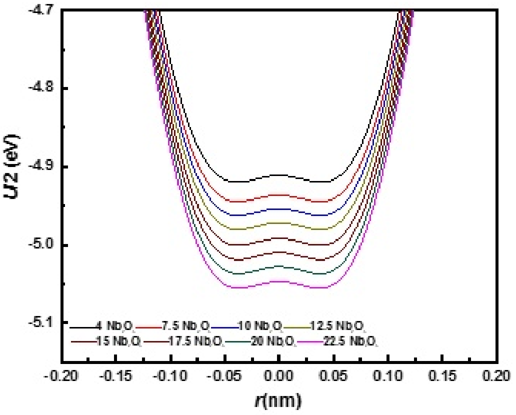

Figure 6. Variations of the configuration of the dual-wells for transverse vibrations of oxygen atoms at e=-2% for the glass system xNb_2O_5 – (100- x) TeO−2 glasses, 4 ≤x≤ 22.5 .

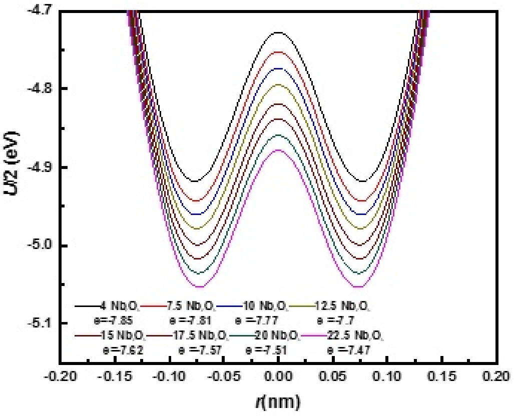

Figure 8. Variations of the configuration of the dual-wells for transverse vibrations of oxygen atoms at e=-10% for the glass system xNb_2O_5 – (100- x) TeO−2 glasses, 4 ≤x≤ 22.5 .

well will represent well the proper activation energies [18],
5. Ratios of elongations are in the range of 40.03%-42.79% as depicted in Fig. 5 for longitudinal oscillations. Also, the elongations for shear oscillation are in the range of -7.85%-7.47% as plotted in Fig. 6,
6. The Figs. (5-6) facilitate getting information about the separation between cation - cation (δxx) and separation between the minima of the states of the well (Dyy). The Dyy can be regarded as the ultrasonic activation energies,
7. More and more ultrasonic energy is transferred to the oxygen atoms, and the elongation of the oscillations will exceed the ratio of 60% for longitudinal oscillations and -10% for shear oscillations as depicted in Figs. (7-8), i.e., the elongations will not be in harmony with the proper ultrasonic activation energies [18],
8. At these ratios, the oscillations of the states of the well two-well exceed elasticity limits, i.e., the case of the applicability of the model will be down.
9. The values of the model's parameters correspond to the literature [13, 19, 28–33],
10. A pronounced parameter is the formed deformation potential $(D)th)$ from the disorder of the two-well potential. This parameter can be given as;

$$D_{th} = \frac{E\delta xx}{2n_b N_A V_m \delta yy} \tag{6}$$

where E is Young's modulus for longitudinal oscillations or shear modulus for shear oscillations, n_b represents a number of cation–anion–cation units/formula unit and N_A is Avogadro's number. The values of these parameters are taken from Ref. [17]. As listed in Table 1, the dependence deformation potential (D_{th}) (longitudinal and transverse) on Nb_2O_5 is correlated well to the values of the ultrasonic activation energies [18]. The values of Dth correspond well to the literature [19, 29]. The results of the obtained relaxation parameters are in good agreement with the extracted results from other experimental reports of tellurite based glasses [34–39].

3. Conclusion

The usage of the central force model to discuss the ultrasonic relaxation in xNb_2O_5 - (100- x) TeO_2 glasses, and $4 \leq x \leq 22.5$ mole %. The glasses show some amazing characteristics:
a) Firstly, as the Nb_2O_5 content increases in the tellurite network, the elastic moduli of such a network increase. This increment is associated with a decrement in both the alternate potential energy and the ultrasonic activation energy of the oscillated oxygen atoms,
b) The increment of [NbO_6] structural units in the tellurite network is congruent with the increment of the number of centers of energy loss increases,
c) A dual-well potential will be formed as a direct result of the oscillations of oxygen atoms. The oscillations are created from the effect of the ultrasonic energy,
d) The created potential well suffers from some changes as the supposed ultrasonic energy increases. The potential well will change from an equilibrium state to a flat-bottom state to a dual-well potential,
e) The last well corresponds well to the ultrasonic activation energy within the elastic limits,
f) More and more ultrasonic energy effects on the oxygen atoms will change the elasticity of the well and it will be no longer matching the proper activation energies. So, the well will be wracked,
g) The increment of the experimental elastic parameters of TeO_2-Nb_2O_5 glasses as a function of Nb_2O_5 mole % will increase the deformation potential of the presented dual-well state.

Conflict of interest statement:
The authors declare that they have no conflict of interest.

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