PHYSICAL REVIEW B VOLUME 60, NUMBER 5 1 AUGUST 1999-I

Ionic conductivity and relaxation dynamics in lithium tellurite glasses

A. Pan and A. Ghosh*
Department of Solid State Physics, Indian Association for the Cultivation of Science, Jadavpur, Calcutta 700032, India

(Received 25 January 1999)

The ionic conductivity and relaxation in the lithium tellurite glasses with varying Li_2O content have been investigated in the frequency range 10 Hz to 2 MHz and in the temperature range from 373 K to just below the glass transition temperature. The composition dependences of the dc conductivity and the activation energy of these glasses have been compared with those of other glasses formed with traditional network formers such as B_2O_3, P_2O_5, and SiO_2 containing the same Li^+ ions as modifiers and explained in terms of composition dependent network structure of these glasses. The frequency-dependent electrical data have been analyzed in the framework of the conductivity and modulus formalisms. Both these formalisms have provided for qualitative similarities in the compositional variation of the relaxation times, interaction between the cations, and the dc conductivity. The finite frequency window has been suggested as the reason for the difference between the numerical values of the stretched exponent β obtained from the modulus formalism and the values of $(1-n)$ obtained from the conductivity formalism. [S0163-1829(99)07129-5]

I. INTRODUCTION

Oxide glasses based on heavy metal glass formers such as TeO_2 posses many interesting physical properties such as low melting point, high chemical durability, high refractive index, and good IR transmittivity, which make them suitable candidates for optical applications.[1] The role of modifier oxides in the vitreous transition of tellurite melts is extremely important unlike traditional glass formers such as SiO_2, B_2O_3, and P_2O_5.[2] When the alkali oxides are introduced in the tellurite network there exist different structural units at different alkali oxide contents. A study of Raman spectra[3] of the alkali tellurite glasses reveals that the structural unit changes from TeO_4 trigonal bipyramid to TeO_3 trigonal pyramid through TeO_{3+1} polyhedra with increasing content of alkali oxides. TeO_2 based glasses containing transition metal ions have been observed to have higher electronic conductivity than that of glasses based on traditional network formers.[4–6] A large number of studies on the ionic conductivity and relaxation in oxide glasses has been reported.[7–13] However, no clear consensus on these processes has emerged so far. To determine the mechanism for ionic conductivity in glasses, it is necessary to separate the contribution of the ionic concentration and mobility to the ionic conductivity.[14–16] Unfortunately, it has not been possible to determine ambiguously these two terms separately, although a few methods have been suggested.[17–21] The dependence of the relaxation mechanism on the concentration of the ionic charge carriers is another problem. The results reported in the literature are contradictory.[22–26] Generally, two formalisms, namely, the conductivity and modulus formalism have been adopted to study the problems.[27–29] But which of these two formalisms can describe the relaxation process much better has not been resolved so far. A correlation of the composition dependence of the ionic conduction and relaxation in glasses with their structure is another interesting problem.

In the present paper we have studied the conductivity and the relaxation mechanisms in lithium tellurite glasses. A comparison of the present results is also made with those of the glasses formed with different traditional network formers and a qualitative description of the variation of the conductivity and activation energy with alkali oxide content is given in terms of structure of the glasses. The ac conductivity and relaxation mechanisms have been analyzed in the framework of the conductivity and the modulus formalisms. Many present workers[30] have criticized the modulus formalism as one giving misleading information on the relaxation process. In the present work we have shown that the conductivity formalism provide for the same qualitative variation of relaxation parameters with composition as the modulus formalism.

II. EXPERIMENTAL

The starting compounds for the synthesis of the glasses of composition $x\mathrm{Li_2O}$-$(1-x)\mathrm{TeO_2}$, where $x=0.10$, 0.15, 0.20, 0.25, and 0.30, were TeO_2 (BDH, 98%) and Li_2CO_3 (Aldrich, 99+%). The appropriate mixtures of these chemicals were placed in a platinum crucible, heated at 450 °C for 2 h for decarbonization and then melted at a temperature in the range 680–760 °C depending on composition in air for about 15 min (chosen for minimum evaporation loss of the melts). The melts were then quenched on a preheated aluminum mould. The samples were immediately transferred to another furnace kept at 150 °C for annealing to remove residual stresses. It was not possible to get glasses with $x\mathrm{Li_2O} > 0.30$. X-ray diffraction patterns were recorded using Cu-K_α radiation in order to confirm the amorphous nature of the prepared samples. The glass transition temperatures were determined from the differential thermal analysis using a heating rate of 10 °C/min. The densities were measured using Archemedes' principle with acetone as an immersion fluid. The concentration N of Li ions were determined from the glass composition and density.

The electrical measurements such as capacitance and conductance of the samples of thickness ~0.3–0.6 mm and diameter ~10 mm were carried out using a precision RLC meter (model 7600, QuadTech) in the temperature range

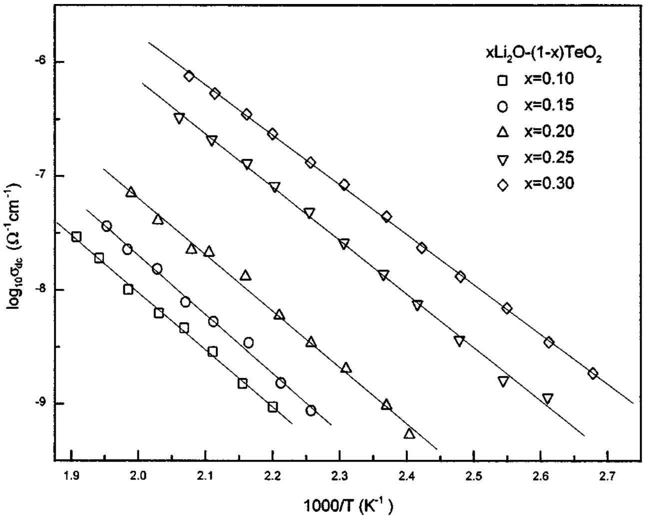

FIG. 1. Temperature dependence of the dc conductivity for different compositions of lithium tellurite glasses shown. The solid lines are the least squares straight line fits to the data.

373–543 K and in the frequency range 10 Hz–2 MHz. The parallel surfaces of the samples were coated with gold for electrodes.

III. RESULTS AND DISCUSSION

A. dc activation energy

The dc electrical conductivity was computed from the ac impedance plots. The variation of the dc conductivity with the reciprocal temperature for all compositions is shown in Fig. 1 which shows that the dc conductivity obeys the Arrhenius relation $\sigma_{\mathrm{dc}}=\sigma_0 \exp(-E_\sigma/kT)$. The values of the activation energy obtained from the least squares straight line fits for different compositions are given in Table I. The dependence of dc conductivity at 200 °C on the lithium ion content for the present tellurite glasses is compared in Fig. 2(a) with that of glasses formed with different traditional network formers such as SiO_2,[31] P_2O_5,[32] and B_2O_3 (Ref. 33) containing Li^+ ions. It is observed that the conductivity is strongly composition dependent for lower Li_2O content ($x Li_2O < 0.3$) for tellurite and borate glasses, while the conductivity shows a weak composition dependence for higher Li_2O content, i.e., $x Li_2O > 0.3$ for silicate, phosphate, and borate glasses. It is noteworthy that in the low Li_2O content region (i.e., below $x Li_2O = 0.30$), the data for phosphate and silicate glasses are not available. In the case of borate glasses as well as tellurite glasses, there exists a steep increase in the conductivity with the increase of Li_2O content in the region $x Li_2O < 0.3$. This may be due to the fact that, in both of these glasses different structural units exist for different Li_2O contents. According to Krogh-Moe,[34] the transformation of the structural units of alkali borate glasses from boroxol to diborates through tetraborate (triborate+pentaborate) occurs as $x Li_2O$ is increased from 0 to ~0.30. In the case of alkali tellurite glasses, the transformation occurs from TeO_4 trigonal bipyramids to TeO_3 trigonal pyramids in the region $0 < x Li_2O \leqslant 0.30$. However, for $x Li_2O > 0.30$, the basic structural units for phosphate glasses (PO_4) and silicate glasses (SiO_4) remain the same. It seems that the structural transformation as well as the increasing number of nonbridging oxygens are responsible for conductivity enhancement with the increase of Li_2O content in borate and tellurite glasses for $x Li_2O \leqslant 0.30$, while the conductivity for silicate, phosphate, and borate glasses becomes less composition dependent for $x Li_2O > 0.30$, as no major structural transformation occurs in this composition range. The conductivity enhancement in this region is solely due to the increasing number of nonbridging oxygens, which are the preferential sites for the alkali ions.

TABLE I. The dc activation energy, dc conductivity at 200 °C, high frequency dielectric constant, conductivity relaxation time at 200 °C, activation energy for the conductivity relaxation times, and the stretched exponential parameter for $x Li_2O$-$(1-x)TeO_2$ glasses.

mole fraction (x)	E_σ (eV) (±0.02)	$\log_{10}\sigma_{\mathrm{dc}}$ at 200 °C (Ω^{-1} cm^{-}) (±0.01)	ϵ_∞	$\log_{10}\tau_m$ at 200 °C (s) (±0.01)	E_τ (eV) (±0.03)	β (±0.01)
0.10	1.00	−8.54	34.40	−2.88	1.05	0.74
0.15	1.03	−8.28	35.88	−3.23	0.97	0.65
0.20	0.98	−7.67	29.73	−3.98	0.90	0.59
0.25	0.93	−6.68	26.74	−4.95	0.95	0.57
0.30	0.87	−6.27	22.22	−5.47	0.86	0.56

The dependence of the activation energy on the lithium ion content of lithium tellurite glasses is compared in Fig. 2(b) with that of silicate, borate, and phosphate glasses. A similar trend to the composition dependence of the conductivity is observed. The activation energy for borate glasses is much larger than that for tellurite glasses in the low alkali region $0 < x Li_2O < 0.30$ and there exists a steep fall in activation energy of borate glasses in comparison to tellurite glasses. The composition dependence of the activation energy in this composition range can be understood in the framework of the Anderson-Stuart model.[14] In this model, the total activation energy E_σ for ionic conduction is the sum of two parts:

$$E_\sigma = \Delta E_B + \Delta E_S, \tag{1}$$

where ΔE_B is the electrostatic binding energy and ΔE_S is the strain energy which are given, respectively, by the expressions

$$\Delta E_B = Z Z_0 e^2 \{1/(r+r_0) - 2/\lambda\}/\epsilon_\infty, \tag{2}$$

$$\Delta E_S = 4\pi G r_0 (r - r_D)^2, \tag{3}$$

where ϵ_∞ is the high frequency dielectric permittivity, λ is the jump distance between the cation sites, Z, r and Z_0, r_0 are the charges and radii of the cation and oxygen anion, respectively, G is the shear modulus, and r_D is the doorway radius which the mobile cation see while moving from one site to another. One of the outcome of the Anderson and Stuart model[14] is that the dielectric constant is a measure of looseness of the structure of the glass network. It also measures the ease with which a modifier cation can migrate through the glass network. In case of borate glasses, the dielectric constant arises from the nonbridging oxygen-modifier cation dipole moment, whereas in case of tellurite glasses, the di-

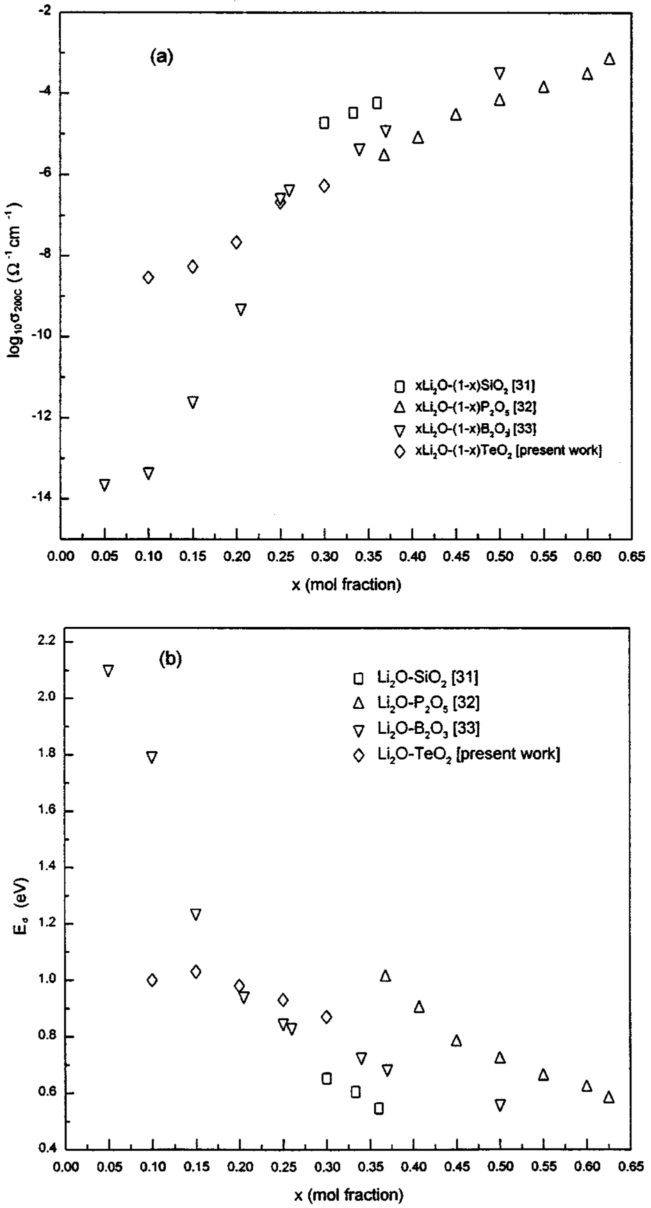

FIG. 2. (a) Composition dependence of the dc conductivity at 200 °C for tellurite, borate, phosphate, and silicate glasses. (b) Composition dependence of the dc activation energy for tellurite, borate, phosphate, and silicate glasses.

electric constant has the contribution of the lone pair of electrons on tellurium atoms as well. Thus the dielectric constants of tellurite glasses are much larger in magnitude than those for the borate glasses, scaling down the Coulomb part [cf., Eq. (2)] of the activation energy,[14] thereby decreasing the total activation energy for tellurite glasses. As the alkali ion content is increased, the dielectric constant increases in borate glasses due to increasing number of nonbridging oxygen-modifier bond and decreases in tellurite glasses due to the decreasing number of tellurium atoms. But the fraction of the increasing number of nonbridging oxygen-modifier bond is greater than that of decreasing tellurium atoms, thereby justifying the steep decrease of activation energy for borate glasses in comparison with tellurite glasses. From the structural point of view the coordination of boron in borate glasses changes from 3 to 4, increasing the network linkages with increasing alkali ion content, whereas in the case of tellurite glasses it changes from 4 to 3 by the cleavage of network linkages. Thus the strain energy part in tellurite glasses is expected to play a dominant role in the total activation energy, contrary to borate glasses. Unfortunately, we are unable to calculate the strain energy contribution quantitatively due to the lack of the values of the doorway radius for lithium tellurite glasses.

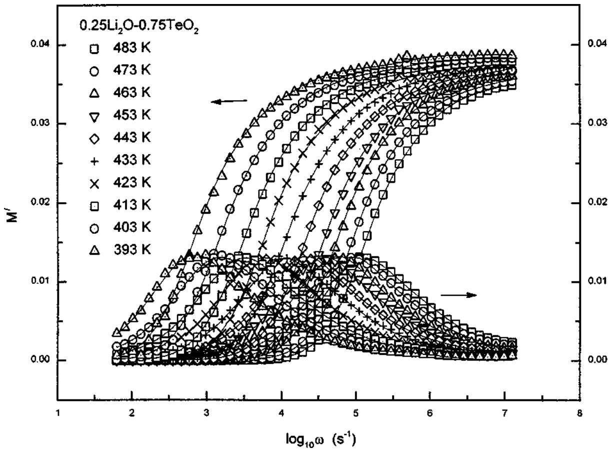

FIG. 3. Frequency spectra of the modulus of $0.25Li_2O$-$0.75TeO_2$ glass at several temperatures shown in the inset. The solid curves are the best fits to Eq. (4).

B. Frequency-dependent conductivity

As pointed out in Sec. I, we have adopted the conductivity as well as the modulus formalisms to study the frequency-dependent conductivity. The modulus formalism[18] is particularly suitable in the absence of well defined loss peaks. In the modulus formalism, an electric modulus $M^*(\omega)$ is defined in terms of reciprocal of the complex dielectric permittivity $\epsilon^*(\omega)$ by

$$M^*(\omega)=1/\epsilon^*(\omega)=M'(\omega)+iM''(\omega)$$

$$=M_\infty\left[1-\int_0^\infty \exp(-i\omega t)\{d\phi(t)/dt\}dt\right], \quad (4)$$

where M_∞ is the asymptotic value of $M'(\omega)$, the inverse of which gives the high frequency dielectric constant ϵ_∞ and $\phi(t)$ is the time evolution of the electric field within the material and is given by the KWW (Ref. 35) function

$$\phi(t)=\exp[-(t/\tau_m)^\beta], \quad 0<\beta\leq 1, \quad (5)$$

where β is a stretching exponent, tending to unity for Debye type of relaxation and τ_m is the conductivity relaxation time. The frequency dependence of $M'(\omega)$ and $M''(\omega)$ for a glass composition at different temperatures is shown in Fig. 3. $M'(\omega)$ shows a dispersion tending to M_∞ at higher frequencies, while $M''(\omega)$ exhibits an asymmetric maximum ($M''_{\max}$) centered at the dispersion region of $M'(\omega)$. It may be noted in Fig. 3 that the position of the peak $M''_{\max}$ shifts to higher frequencies as the temperature is increased. The frequency (ω_m) corresponding to $M''_{\max}$ gives the most probable conductivity relaxation time τ_m from the condition $\omega_m\tau_m=1$. The values of $\log_{10}\tau_m$ at 200 °C for different glass compo-

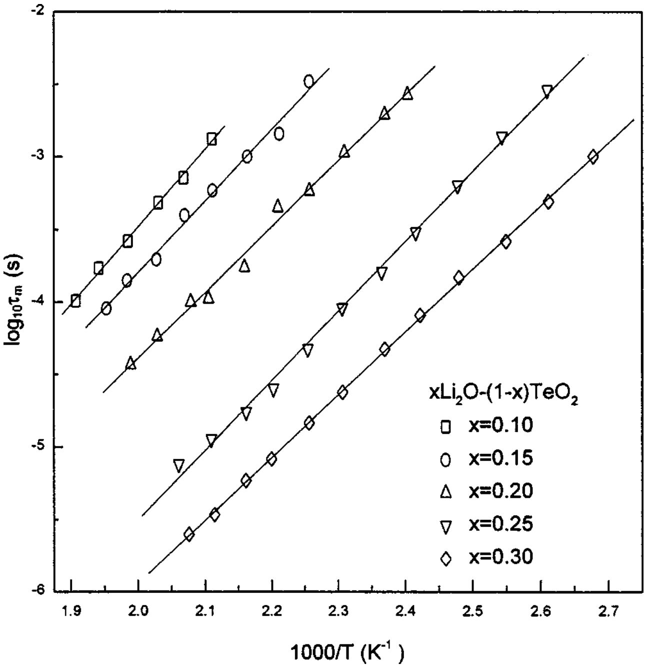

FIG. 4. Temperature dependence of the conductivity relaxation times obtained from the modulus formalism for different compositions of lithium tellurite glasses shown. The solid lines are the least squares straight line fits to the data.

sitions are listed in Table I. It shows a decreasing trend with increasing Li_2O content, arising from the structural transformation in tellurite network. At very low composition, a few of the Te-O^- sites are available which makes the ion mobile over long distances at lower frequencies only, resulting in a large relaxation time. As the content of Li_2O is increased, more and more nonbridging oxygens are included in the network. Consequently, the ions remain mobile at higher frequencies making the relaxation time small. Figure 4 shows that the conductivity relaxation time also obeys the Arrhenius relation $\tau = \tau_0 \exp(E_\tau/kT)$ and the corresponding activation energy E_τ (Table I) for relaxation are found to be close to the activation energy for the dc conductivity (E_σ), which indicates that the charge carrier has to overcome the same energy barrier while conducting as well as while relaxing in consistence with fluctuation-dissipation theorem.[36] A masterplot for the electric modulus $M^*(\omega)$ is shown in Fig. 5 in which each frequency is scaled by peak frequency (ω_m) and $M'(M'')$ is scaled by $M_\infty(M''_{\max})$. The perfect overlap of the curves for all the temperatures into a single master curve indicates that the dynamical processes are temperature independent. The data for $M'(\omega)$ and $M''(\omega)$ in Fig. 3 were fitted simultaneously to Eq. (4). The values of the stretched exponential parameter β obtained from the fits are listed in Table I. They are found to decrease with increasing Li_2O content. As the alkali content is increased, the cation-cation distance (Table II) decreases and thus the interaction between the cations increases resulting in a decrease of β.

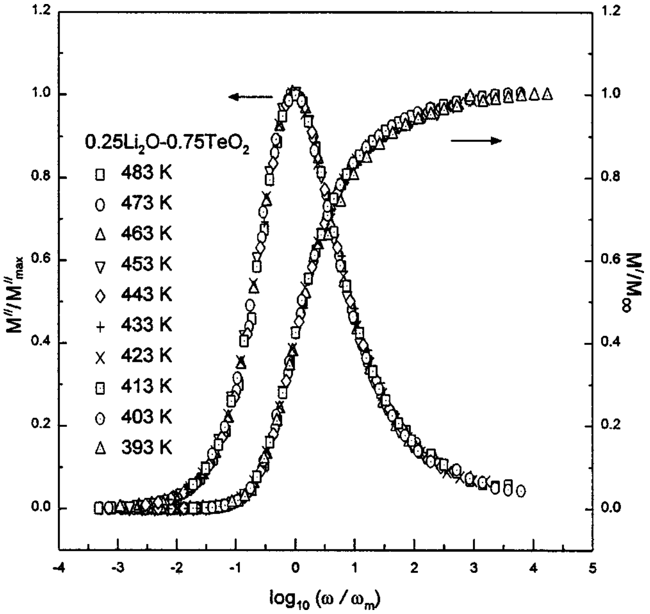

FIG. 5. Plot of M'/M_∞ and $M''/M''_{\max}$ versus $\log_{10}(\omega/\omega_m)$ for 0.25Li_2O-0.75TeO_2 glass for the same temperatures as shown in Fig. 3.

As already mentioned in the Introduction, we have also tried to analyze our data for tellurite glasses in the framework of the Almond-West formalism.[20] In this formalism the bulk frequency-dependent conductivity $\sigma'(\omega)$ (real part) in ionic crystal and glasses is described by

$$\sigma'(\omega) = \sigma_{dc}[1 + (\omega/\omega_H)^n], \tag{6}$$

where ω_H is the hopping frequency of the charge carriers and n is a frequency exponent parameter in the range $0 \leqslant n \leqslant 1$

TABLE II. The concentration of Li^+ ions and the average cation-cation separation calculated from composition and density; the mobile ion concentration, the hopping frequency at 200 °C, and the frequency exponent n obtained from the fits of Eq. (8) to the data and activation energy for the hopping frequency and dc conductivity calculated from Eq. (7) for xLi_2O-$(1-x)TeO_2$ glasses.

mole fraction (x)	$\log_{10} N$ (cm^{-3}) (± 0.2)	R ($\times 10^{-8}$ cm) (± 0.01)	$\log_{10} N_0$ at 200 °C (cm^{-3}) (± 0.02)	$\log_{10}\omega_H$ at 200 °C (rad s^{-1}) (± 0.05)	n (± 0.01)	E_c (eV) (± 0.04)	$\log_{10}\sigma_{dc}$ at 200 °C (Ω^{-1} cm^{-1}) (± 0.02)
0.10	21.64	6.11	20.4	4.38	0.60	0.96	−8.64
0.15	21.83	5.31	21.02	4.18	0.61	0.94	−8.33
0.20	21.96	4.79	21.17	4.76	0.61	0.88	−7.69
0.25	22.06	4.44	21.18	5.82	0.65	0.90	−6.69
0.30	22.14	4.18	21.16	6.32	0.66	0.88	−6.27

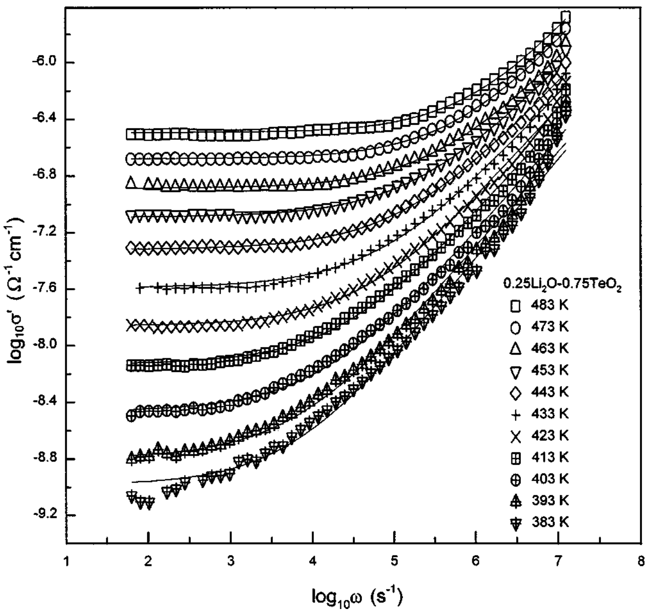

FIG. 6. Frequency spectra of the conductivity for $0.25Li_2O$-$0.75TeO_2$ glass at several temperatures shown in the inset. The solid curves are the best fits to Eq. (8).

measuring the interaction between the mobile ions. The value of σ_{dc} using Nerst-Einstein relation and assuming thermally activated hopping of charge carriers is given by

$$\sigma_{dc}=[N_0(Ze)^2\gamma\lambda^2\omega_H]/2\pi kT, \tag{7}$$

where N_0 is the mobile ion concentration, e is the electronic charge, γ is the geometrical factor for ion hopping, and λ is the average jump distance between the mobile ion sites. Thus the bulk ac conductivity can be written from Eqs. (6) and (7) as

$$\sigma'(\omega)=\{N_0(Ze)^2\gamma\lambda^2\omega_H/2\pi kT\}[1+(\omega/\omega_H)^n]. \tag{8}$$

In the present calculation, we have assumed the ion jump distance λ to be equal to the average cation-cation separation distance (R) and γ was taken to be $\frac{1}{6}$ assuming the present glass to be isotropic. The frequency spectra of the real conductivity $\sigma'(\omega)$ for a glass composition is shown in Fig. 6 at different measuring temperatures. It is observed that at lower frequencies, the conductivity is almost independent of frequency, approaching the dc conductivity. As the frequency is increased, the conductivity shows a dispersion which shifts to higher frequencies with the increase in temperature. The experimental conductivity data have been fitted to Eq. (8) with N_0, ω_H, and n as variables. The best fit of the conductivity spectra is exhibited as solid lines in Fig. 6 for a glass composition and for different temperatures. The results of the analysis are listed in Table II. An Arrhenius plot of the mobile charge carrier concentration N_0 is shown in Fig. 7(a) which reveals that they are not thermally activated, indicating that lithium tellurite glasses are strong electrolytes.[17] A comparison of the numerical values of N_0 with those of the concentration N of the lithium ions (Table II) obtained from the glass composition and density indicates that only 5–15% of the total lithium ions contribute to the electrical conduction. The reciprocal temperature dependence of hopping frequency (ω_H) is shown in Fig. 7(b) which indicates that ω_H obeys the Arrhenius relation. The values of the activation energy E_c (Table II) of the hopping frequency obtained from the slopes [Fig. 7(b)] are close to the dc activation energy (E_σ). The values of parameter n are weakly temperature dependent, but shows an increasing trend with increasing lithium content. This may be due to the fact that as the concentration of the lithium ions is increased, the interaction between them increases resulting in a higher value of n (n being a measure of interaction). The reciprocal of the hopping frequency (ω_H) shows a decreasing trend with increasing Li_2O content supported by the structural transformation in tellurite networks as explained earlier for the conductivity

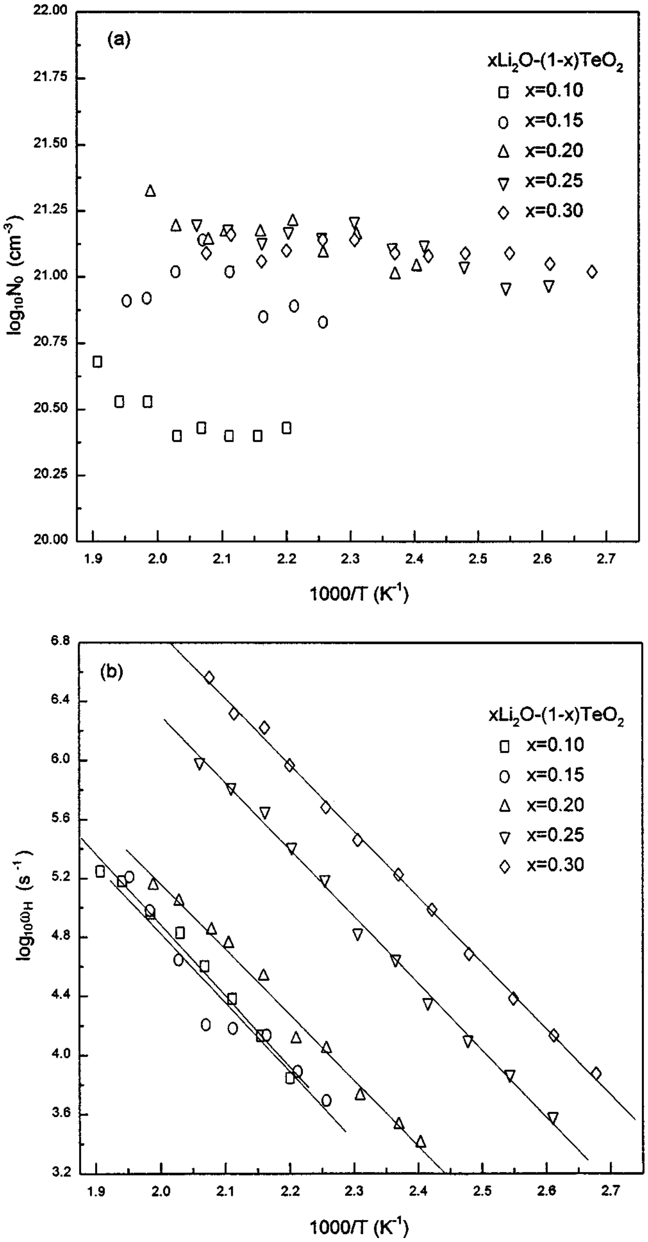

FIG. 7. (a) Temperature dependence of the ion concentration N_0 for all compositions of lithium tellurite glasses (shown) obtained from the fits of the conductivity isotherms to Eq. (8). (b) Temperature dependence of hopping frequency obtained from the conductivity formalism for all compositions of lithium tellurite glass (shown). The solid lines are the least squares straight line fits to the data.

relaxation time in the text. The values of the dc conductivity (Table II) were calculated from Eq. (7) using the values of the fitting parameters N_0, ω_H, and λ and were also found to agree well within 1% with the values obtained from the impedance plots. A comparison of the numerical values of n with those of the stretched exponential parameter β of the conductivity relaxation model shows that although the qualitative changes in n and β are in conformity with the fact that both represent the interaction between the modifiers, they do not, however, exactly obey the theoretical relationship between β and n, namely, $\beta = 1 - n$. The reason for numerically different values of β from those of $(1-n)$ may be the limited frequency window (10 Hz–2 MHz) of our measurements.

IV. CONCLUSIONS

The dc conductivity and conductivity relaxation of $x\mathrm{Li_2O}$-$(1-x)\mathrm{TeO_2}$ glasses with varying Li_2O content have been studied in wide frequency and temperature ranges. The dc conductivity and activation energy of these glasses were compared with those of other glasses formed with traditional glass formers, in particular, borate glasses and containing the same modifier Li^+ ions. In the lower Li_2O content region, the steeper increase of conductivity for tellurite and borate glasses was suggested to arise from the structural transformation and increasing number of nonbridging oxygen, while in the higher Li_2O content region for silicate, phosphate, and borate glasses, the weak increase in conductivity was due to the increasing number of nonbridging oxygens only. The ionic relaxation was analyzed in the framework of conductivity as well as modulus formalisms. The decreasing trend of conductivity relaxation time (τ_m) and stretched exponential parameter (β) with increasing Li_2O content, was explained in terms of structural transformation of tellurite network and cation-cation distance correlation, respectively. The composition dependence of the frequency exponent parameter (n) and the hopping rate (ω_H^{-1}) obtained from the conductivity formalism provided for the correlation with that of β and τ_m, respectively. The deviation of the theoretical relationship between β and $(1-n)$ was ascribed to the limited frequency window.

ACKNOWLEDGMENT

The financial support by the CSIR for the work is thankfully acknowledged.

---

*Author to whom correspondence should be addressed.

[1] B. Dubois, H. Aomi, J. J. Videau, J. Portier, and P. Hagenmuller, Mater. Res. Bull. **19**, 1317 (1984).

[2] S. Neov, V. Kozhukharov, I. Gerasimova, K. Krezhov, and B. Sidzhimov, J. Phys. C **12**, 2475 (1979).

[3] T. Sekiya, N. Mochida, A. Ohtsuka, and M. Tonokawa, J. Non-Cryst. Solids **144**, 128 (1992).

[4] A. Mansingh and V. K. Dhawan, Philos. Mag. B **47**, 121 (1983).

[5] B. W. Flynn, A. E. Owen, and J. M. Robertson, *Proceedings of the 7th International Conference on Amorphous Liquid Semiconductors*, edited by W. E. Spear (CICL, Edinberg, 1977), p. 678.

[6] K. Tanaka, T. Yoko, K. Kamiya, H. Yamada, and S. Sakka, J. Non-Cryst. Solids **135**, 211 (1991).

[7] C. A. Angell, Chem. Rev. **90**, 523 (1990).

[8] S. R. Elliott and A. P. Owen, Ber. Bunsenges. Phys. Chem. **95**, 987 (1991).

[9] A. Bunde, M. D. Ingram, and P. Maass, J. Non-Cryst. Solids **172-174**, 1222 (1994).

[10] D. L. Sidebottom, P. F. Green, and R. K. Brown, J. Chem. Phys. **108**, 5870 (1998).

[11] K. Funke, Prog. Solid State Chem. **22**, 111 (1993).

[12] P. Maass, M. Meyer, and A. Bunde, Phys. Rev. B **51**, 8164 (1995).

[13] K. L. Ngai, J. Non-Cryst. Solids **103**, 232 (1996).

[14] O. L. Anderson and D. A. Stuart, J. Am. Ceram. Soc. **37**, 573 (1954).

[15] S. W. Martin, J. Am. Ceram. Soc. **74**, 1767 (1991).

[16] J. L. Souquet and W. G. Perera, Solid State Ionics **40-41**, 595 (1990).

[17] M. D. Ingram, M. A. Mackenzee, W. Muller, and M. Torse, Solid State Ionics **28-30**, 677 (1988).

[18] Y. Moyamoto, M. Itoh, and K. Tanaka, Solid State Commun. **92**, 895 (1994).

[19] H. J. Scutt, Solid State Ionics **70-71**, 505 (1996).

[20] D. P. Almond, G. K. Duncan, and A. R. West, Solid State Ionics **8**, 159 (1983); **9&10**, 277 (1983).

[21] E. F. Hairetdinov, N. F. Uranov, H. K. Patel, and S. W. Martin, Phys. Rev. B **50**, 13 259 (1994).

[22] J. M. Seyer, J. M. Reau, H. Aomi, P. Hagenmuller, and M. Poulain, J. Non-Cryst. Solids **85**, 315 (1985).

[23] H. K. Patel and S. W. Martin, Phys. Rev. B **45**, 10 292 (1992).

[24] H. Kahnt, Ber. Bunsenges. Phys. Chem. **95**, 1021 (1991).

[25] K. L. Ngai and S. W. Martin, Phys. Rev. B **40**, 10 550 (1989).

[26] K. L. Ngai, J. N. Mundy, H. Jain, O. Kanart, and G. B. Jollenbeck, Phys. Rev. B **39**, 6169 (1989).

[27] P. B. Macedo, C. T. Moynihan, and R. Bose, Phys. Chem. Glasses **13**, 171 (1972).

[28] B. Rolling, A. Happe, K. Funke, and M. D. Ingram, Phys. Rev. Lett. **78**, 2160 (1997).

[29] B. Rolling, Solid State Ionics **105**, 185 (1998).

[30] S. R. Elliott, J. Non-Cryst. Solids **170**, 97 (1994); J. C. Dyre, *ibid.* **135**, 219 (1991).

[31] K. Otto, Phys. Chem. Glasses **7**, 29 (1966).

[32] S. W. Martin and C. A. Angell, J. Non-Cryst. Solids **83**, 185 (1986).

[33] S. A. Schtchukarev and W. R. Muller, Z. Phys. Chem. Abt. A **150**, 439 (1930).

[34] J. Krogh-Moe, Phys. Chem. Glasses **6**, 46 (1965).

[35] G. Williams and D. C. Watts, Trans. Faraday Soc. **66**, 80 (1970).

[36] K. L. Ngai, Comments Solid State Phys. **9**, 127 (1979); **9**, 141 (1980).